



**CASCADE AND TWO-PHOTON LASING
FROM TWO-PHOTON EXCITATION OF
CESIUM 6^2D**

DISSERTATION

Nathan D. Haluska, Captian, USAF
AFIT-ENP-DS-17-S-026

**DEPARTMENT OF THE AIR FORCE
AIR UNIVERSITY**

AIR FORCE INSTITUTE OF TECHNOLOGY

Wright-Patterson Air Force Base, Ohio

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DISSERTATION

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in Partial Fulfillment of the Requirements for the
Degree of Doctor of Philosophy

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Abstract

Cascade and two-photon lasing have been studied utilizing two-photon pumping of the cesium $6^2D_{3/2}$ and $6^2D_{5/2}$ states, with both pulsed and CW sources. Two-photon pumping may enable a new generation of wavelength agile diode pumped alkali lasers.

For the high intensity pulse regime, we report our on-resonance observations from two-photon pumping the Cs $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ and $6^2S_{1/2} \rightarrow 6^2D_{5/2}$ transitions. Cascade lasing via Amplified Spontaneous Emission (ASE) is observed on over 17 transitions. Many stimulated emission lines possess 100 to 229 μJ of energy, slope efficiencies of 4 to 7%, low thresholds favorable for diode pumping, and together have combined optical conversion efficiency as high as $10\% \pm 3\%$. This agile system could have applications in infrared counter measures, target illuminator lasers, beacon lasers, underwater communication, and other tactical laser applications. Also in the high energy pulse regime, we report the theory and observation of two-photon lines. The first arises from the $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ transition in which a 1381 nm line is observed. A second two-photon line is observed around 3120 nm, from the $5^2D_{1/2} \rightarrow 6^2S_{1/2}$ transition. The 1381 nm line is shown to have >9 nm of tuning, a slope efficiency of $\sim 1.2\%$, and lasing threshold of <140 kW/cm^2 . Four wave mixing, six wave mixing, and two-photon lasing are discussed as potential mechanisms; and future two-photon laser experiments are suggested.

A CW laser (via ASE) on the $3.096\mu\text{m}$ $7^2P_{1/2} \rightarrow 7^2S_{1/2}$ line is also generated by two-photon pumping the cesium $6^2D_{3/2}$ state with a focused, single pass, 1.6W Ti:sapphire laser. This provides a peak corrected power of 750 μW , absorption corrected efficiency of 0.7%, and lasing thresholds <10 kW/cm^2 . Our CW results demonstrate that a two-photon diode pumped alkali laser is feasible. We also develop a hyperfine

resolved two-photon absorption theory and validate it with two-photon absorption measurements, and 3 μm lasing spectra. The absorption and emission spectra enable the calculation of the $6^2D_{3/2}$ energy level value of $22588.8203 \pm .0003 \text{ cm}^{-1}$. Finally we observe CW nonlinear enhancement on the 1359 nm line and generation of the 1381 nm two-photon line.

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First and foremost I would like to thank God for making me OK enough at physics, and giving me a beautiful and supportive wife who lets me do physics. I want to thank my wife for her patience and strength while dealing with my long work hours. Without her, I would not be where I am today, and I'd also be pretty boring. She still points out that in my master thesis acknowledgments I promised to get her a pet cow. Yes, you can still get a pet miniature cow, no I don't know where we're gonna put it. I wish I could come up with something more romantic or clever to say, but I'm a physicist, not a bard.

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CASCADE AND TWO-PHOTON LASING FROM TWO-PHOTON EXCITATION OF CESIUM 6²D

I. Introduction

In this work we outline innovative techniques to build off of the recent success of Diode Pumped Alkali Lasers (DPALs) [75, 6]. High power diode arrays can produce 10 to 100 kW/cm² intensities with good electrical to optical efficiency. The challenge is to enable this efficient but poor beam quality diode energy to propagate over long distances.

In 2001 [54, 55] it was shown that diode light can be phased together in an alkali vapor gain medium by abortion on D_2 line, fine structure mixing, and lasing on the D_1 line back to the ground state. This mixing is necessary to invert the alkali vapor and enables dramatically improved coherence quality of the laser emission on the D_1 line. We seek to exploit similar schemes in alkali vapor involving higher lying excited states.

1.1 Dissertation Goals and Potential Applications

Our goal with this dissertation is to assess the feasibility of two-photon pumping alkali vapors to create near-term two-photon DPAL analogs and identify future fundamental research applications. We define two-photon pumping to mean the simultaneous absorption of two photons through a virtual state. For example, the $n^2S_{1/2} \rightarrow n^2D_{5/2}$ or $n^2S_{1/2} \rightarrow n + 1^2S_{1/2}$ are two-photon allowed transitions. This is covered in greater detail below and in Chapter IV. This is not to be confused with sequential pumping where two pump sources operate in serial—e.g.

$n^2S_{1/2} \rightarrow n^2P_{3/2} \rightarrow n^2D_{5/2}$ or $n^2S_{1/2} \rightarrow n^2P_{3/2} \rightarrow n+1^2S_{1/2}$. Our experimental work is done in Cs, but these processes have analogs in Na, K, and Rb.

Although the normal DPAL is ideal for high efficiency applications, the operational laser wavelengths are constrained by the near infrared $D_1 \ ^2P_{1/2} \rightarrow ^2S_{1/2}$ transitions at 770, 795, and 894 nm in K, Rb and Cs. Extended wavelength capabilities and ranges, or "wavelength agility," from a potential two-photon DPAL are desirable for many applications including beacons, illuminators, underwater communications, and infrared counter measures [14, 72, 4]. We provide a fuller description of these processes in the next section, as well as Chapters IV and VI.

We also assess the feasibility of two-photon lasing in cesium vapor. This process is introduced in the next section, and we provide additional details in Chapter II, V, and VII. Although we do not prove two-photon lasing in this work, we do demonstrate several useful properties of cesium vapor that prove a two-photon laser is theoretically possible, as well as many experimental conclusions that suggest it may have occurred in our results.

1.2 Wavelength Agility from Diode Pumping

There are several ways to use alkali vapors to generate wavelengths from the UV to the far infrared. In this work we focus on cascade lasing (or one-photon lasing), and two-photon lasing, with some discussion on Multi Wave Mixing (MWM), and Stimulated Raman Scattering (SRS).

All of these processes can lead to wavelength agility, however we also need to consider compatibility with modern high power laser diodes. In general, one and two-photon lasing require inversions but do not require highly coherent pump sources; conversely, MWM and SRS do not necessarily require an inversion but rely on the coherence properties of their pump sources.

Cascade Lasing

Cascade lasing accounts for much of AFITs recent work in alkali vapor laser technology [97, 13, 64]. However, it's also the oldest example of an alkali laser. It was first suggested in 1958 (potassium) [83], and realized in 1962 (cesium) [78]. With the recent success of DPALs, it has been tested as recently as 2016 [13, 64]. Many of these experiments do not involve cavities and instead rely on the high gain inherent to alkali vapors to generate cascade lasing from Amplified Spontaneous Emission (ASE). This document focuses on stimulated emission beams (ASE), or "lasing" without a cavity. We are interested in techniques that put significant populations in higher energy states than typically used in standard DPAL devices. These populations are easily inverted to non-ground state levels, which leads low lasing thresholds. In our case we utilize two-photon pumping, where prior to this work, no continuous wave cascade lasing has been observed. Even though the two-photon pump makes the overall process non-linear, we refer to this process as cascade lasing, one-photon lasing or linear lasing, to distinguish between other mechanisms. We further expand on the background in Chapter IV.

Two-Photon Lasing

We use the term two-photon laser to describe a laser whose stimulated emission yields four quanta from two incident quanta. Degenerate two-photon stimulated emission is illustrated in Figure 1; where some external process first puts the system in state $|4\rangle$, two photons are injected with the same (degenerate) angular frequency ω , and $2\hbar\omega$ is the energy of the $|4\rangle \rightarrow |1\rangle$ transition. This process is resonantly enhanced by the presence of real states $|3\rangle$ and $|2\rangle$ if they are close to virtual state $|v\rangle$. Therefore it is also possible to achieve a larger gain on the two-photon transition if we tune $|v\rangle$ closer to a real state. This process is referred to as non-degenerate

two-photon stimulated emission as we now require two different incident frequencies. As evident from Figure 1, we expect the properties of the two-photon stimulated emission process to possess potentially distinct coherence and statistical properties from the one-photon laser.

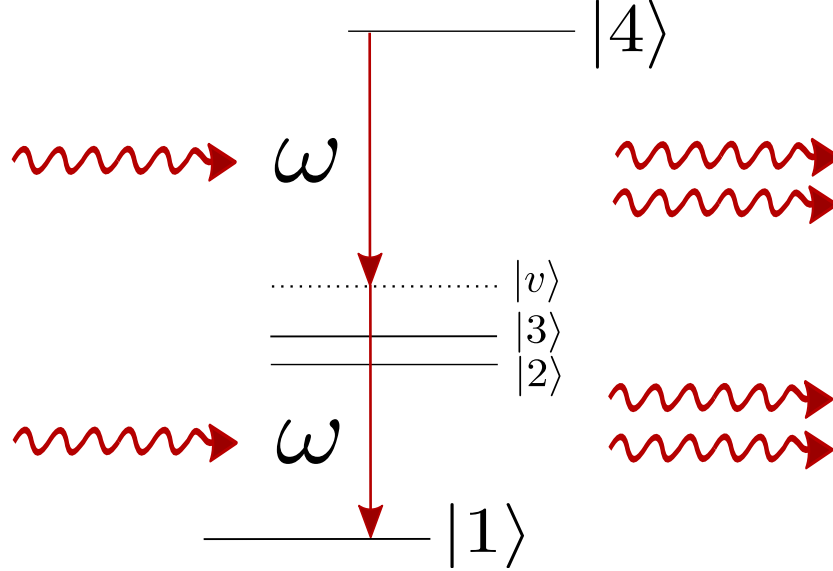


Figure 1. Degenerate two-photon stimulated emission from $|4\rangle \rightarrow |1\rangle$ via virtual level $|v\rangle$. Given a system in state $|4\rangle$, two incident quanta with frequency ω will yeild 4 total quanta.

Historical Synopsis

Two-photon processes were first considered by Maria Goppert-Mayer in 1929 and 1931 [42, 43]. These references are in German, but English translations can be found using the cited URL in reference [62]. For example, translated titles of Goppert’s works offer some insight into her thoughts: “On The Probability of Two Light Quantum Working Together in an Elementary Act.” and “Elementary Acts With Two Quantum Jumps.” Goppert-Mayer utilized time-dependent perturbation theory to describe transition probability (but not the transition rate). More modern versions of the quantum optics two-photon transition rate can be found in [26, 58]. Note that in

the full quantum mechanical treatment the Hamiltonian in the Schrodinger picture is not time dependent so time dependent perturbation theory is used by switching to the Heisenberg picture. However, we focus on intense light fields, thus a semi-classical treatment is preferable. Our treatment in Appendix A is semi-classical and follows the methods of [9, 57].

The two photon laser was first proposed in 1964—after the invention of the ruby laser in 1960. This “Double Quantum Stimulated Emission Device” was proposed in close succession by [93] and [77]. In 1967 when Lambropoulos analyzed the “Quantum Statistics of a Two-Photon Quantum Amplifier,” he suggested that the statistics will be distinct from thermal sources and one-photon lasers [56]. Lambropoulos also utilized a full quantum mechanical treatment of atoms, though the interaction was confined to a single mode. At this point in the history we can already see two groups interested in two-photon lasers: those interested in the properties as a laser device, and those interested in the implications for quantum optics experiments.

The first demonstration of two-photon gain was reported by Loy in 1978 [59]. In his letter, he reported a two-photon gain of only 0.2% over a 40 cm length in NH_3 . The fundamental limitation was the pressure scaling. Essentially the only way to raise gain was to add more NH_3 , i.e. raise the pressure, but doing so would just increase the already fast population relaxation. Thus, the author concluded that “the NH_3 system excited by two-photon adiabatic inversion does not appear to be promising for achieving two-photon laser oscillation” [59].

In 1981 a German team claimed to demonstrate a pulsed two-photon laser [67]. As we note below, it is unlikely that this was a true two-photon laser, it was more likely a 5 wave mixing process (the odd parity here is the result of a quadruple transition). This was the first group to realize the potential of the large two-photon gain in alkali vapors. The experimental setup utilized three focused pulsed dye lasers, two from

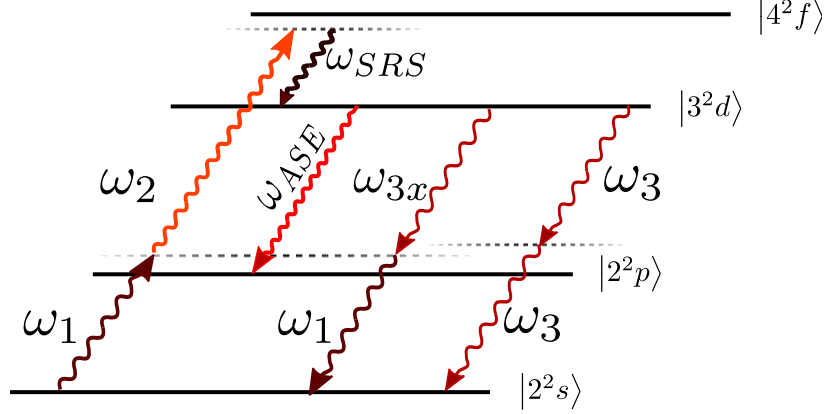


Figure 2. The first claimed two photon laser demonstration [67]. Li Energy level diagram. Shown (left to right) 1) The pump from $s \rightarrow d$ via $\omega_1 + \omega_2 - \omega_{SRS}$. Followed by the three potential excitation process: 2) Parasitic ASE via ω_{ASE} . 3) Non-degenerate two photon stimulated emission utilizing ω_1 as a seed (note ω_{3x} was NOT seeded): $d \rightarrow s$ via $\omega_1 + \omega_{3x}$. 4) Degenerate two photon stimulated emission tuning the third pulsed dye laser through resonance with itself: $d \rightarrow s$ via $\omega_3 + \omega_3$.

one direction and one from the other, and a Li heat pipe.

The energy levels are shown in Figure 2. The system is pumped via a resonant electric dipole, resonant electric quadrupole, stimulated Raman scattering process to move population from $2s \rightarrow 3d$. Parasitic amplified stimulated emission (ASE) from ω_{ASE} is inhibited¹ by the pumping process: ω_1 and ω_2 are tuned in wavelength to be very closely resonant with the $2P$ state $\Delta\omega \approx 80 \text{ cm}^{-1}$. The two fields are then tuned in power (ω_1 set to high power, ω_2 is tuned as low as possible). The net effect of this wavelength and power tuning is that population is also transferred to the $2P$ state by the pump process. This $2P$ population effectively quenches any one-photon $3d \rightarrow 2p$ ASE through inhibition of the population inversion.

Their paper detailed 2 two-photon processes as shown in Figure 2. The first involved non-degenerate two-photon stimulated emission where ω_{3x} is the unseeded observed wavelength, ω_1 acted as a seed (ω_3 is turned off). The important observa-

¹The authors call this super-fluorescence and note it ω_{SF} , but we believe this is ASE.

tions were that the slope of the non-degenerate emission was $\propto 1/(\Delta\lambda)^4$ and more importantly a resonant dip is observed when $\Delta\nu < 30 \text{ cm}^{-1}$ which is likely the only experimental evidence of a two-photon process collapsing to two single photon processes. The second two-photon process identified was amplification of degenerate two-photon stimulated emission on ω_3 . Their results showed 20% amplification of the ω_3 probe beam when the beam was at max power and the heat pipe was at 720 °C.

Although the paper was very short and offered little experimental insight, the authors claimed to demonstrate “non-degenerate two-photon light generation and degenerate two-photon amplification in Li vapor,” and a two-photon gain of around 20% on a dye laser probe beam [67]. Unfortunately this paper was hindered by the confusion of super-fluorescence and cascade lasing, lack of experimental details, insufficient evidence of two-photon lasing as opposed to MWM, and limited conclusions. This lead to criticism that we discuss in section 1.2.

For the remainder of the 1980s, only theory was published, see for example [111]. In 1992, Daniel Gauthier detailed a continuous wave two-photon laser in an atomic beam of barium atoms [38]. However, this technique involved the energy levels of dressed-atom doublets. Where energy splitting of dressed atom states is on the order of tens of MHz—which required strict pump/seed line widths. Gauthier’s research group spent the next few years looking for better options. The group published several theoretical papers [18], and the group also realized the advantage of alkali vapors. In 1997 Hope Concannon, a PhD student of Gauthier, showed a 30% CW amplification in a pumped/seeded scheme in potassium vapor utilizing two-photon stimulated Raman scattering [20]. Concannon estimated a 300x improvement in two photon gain compared to the earlier work [38, 111]. The advantages of alkali vapor as a gain medium are evident. Although the CW two-photon Raman laser is interesting for nonlinear and quantum optics work, the tunability is limited to

tens of MHz. Gauthier’s research group at Duke did some more experimental work with a second PhD student, Oliver Phister, in this area in the late 1990’s [74]; and some theoretical work in the early 2000’s [33], focused on their success in K vapor. A chapter in Progress in Optics was dedicated to this subject in 2003 [37] which provides an excellent overview of the subject matter.

This concludes most of the work to date in the gas laser realm. In this prospectus we do not include a thorough review of emerging work in solid state devices but we note that the challenges are more difficult: the cross-sections are several orders of magnitude lower, material survivability and temperature fluctuations/control are all much more important in solid state devices. For example, a team in 2011 saw two-photon spontaneous emission from a Type III-V semiconductor quantum dot [69]. Like the articles above, the key challenge was overcoming the one-photon gains. They achieved this by building a “double” hetero-structure. The first hetero-structure being the 72.5 nm radius InAs quantum dots embedded in GaAs. The second was a 130 nm 1D quantum well structure made by decreasing the quantum dot density in the middle of the slab. This density miss-match formed a cavity that favored the two-photon transition. The difficulty here is making the nano-scale structure, cooling it to 4.5K, and limiting the Ti:sapphire pump beam to 180 nW. This is clearly a very different device then the alkali vapor lasers outlined above.

The underlying theme from this historical perspective is that alkali vapors make an excellent test bed for two photon lasers, whether resonant dipole, dressed states, or stimulated Raman scattering. We build on this idea in Chapter II and V, and in Chapter VIII we suggest a new approach to a two-photon laser in our conclusions.

Multi Wave Mixing

Multi Wave Mixing (MWM) refers to multiple electromagnetic fields interacting within a dielectric medium to produce an additional field[10]. For our purposes, we note that Four Wave Mixing (FWM), and Six Wave Mixing (SWM) are possible effects observed in our experiments. As an example, with the correct resonance scheme, two-photon pumping can create cascade lasing on multiple fields from population inversions. These fields can lead to MWM. Most notably, MWM does not require a population inversion.

Although a population inversion is not required, there are frequency matching (energy conservation) and phase matching (momentum conservation) conditions [10]. These processes also occur on or near real transitions, and the wave vectors $\mathbf{k}_i$ are therefore sensitive to index of refraction variations. This leads to axial and non-axial emission. In gasses the off axis angle of the non-axial emission is generally small (we do not measure these in this work). MWM processes are also sensitive to coherence effects, collisions, and other broadening mechanisms; and it is difficult to see if high power laser diodes could be used to efficiently pump a FWM or SWM process with desirable output characteristics like beam quality. However, our surrogate pump sources do not have these limitations so MWM effects must be considered.

Cascade lasing can create additional fields until limited by a lack of inversion (e.g. to the ground state), and MWM can enable that population to return to the ground state more effectively than fluorescence. This coupling can therefore enhance both processes while obfuscating the linear physics from the non-linear physics. For instance, in recent AFIT work Sulham and Brown [14, 97] both report lasing behavior in environments where one type of lasing is difficult to distinguish from FWM. Sulham reports lasing on the Rb and Cs blue transitions from two-photon pumping. As we explore further in Chapter V, this blue emission is frequently attributed to a FWM

process. Brown also observed SRS at anti-Stokes and Stokes lines, this too is often attributed to a FWM process. For example, the pump on the $^2P_{1/2}$ state can lead to a SRS Stokes line from a virtual state to the $^2P_{3/2}$ state; if the Stokes line is strong enough the SRS process of the pump and the Stokes fields can couple with the pump field a second time to produce the anti-Stokes line, which describes a FWM process. Thus FWM can also obfuscate multiple nonlinear effects. We provide specific examples of MWM and SWM in Chapter V where we compare two-photon lasing with SWM.

Competition Between Cascade Lasing, Two-Photon Lasing and Multi Wave Mixing

In the previous few sections we note multiple linear (cascade lasing lines) and non-linear (two-photon pumping, two-photon lasing, FWM, SWM) processes. When many of these processes are present, it creates an environment where it is difficult to distinguish the process which require an inversion (cascade and two-photon lasing) from those that do not (FWM, SWM).

There are a few examples of this competition in literature: The original two-photon laser publication [67] that we cover in section 1.2, was quickly followed by criticism from Jackson [51] who presented several arguments that favored a MWM process over a two-photon laser processes. Sparbier [94] discusses some similar observations where the presence of two-photon emission could originate from both stimulated and MWM processes. In both [67] and [94], no observations made could decouple the impact of the ASE processes from the MWM processes.

In 1987, Boyd developed one technique to distinguish ASE from MWM [11]. In this article, Boyd two-photon pumped the 3D states in Na vapor with GW/cm^2 pump intensities from a highly focused pulsed dye laser, and monitored the forwards and

backwards ASE on the $3D \rightarrow 3P$ line and two FWM lines. Boyd demonstrated that pump intensities $< 0.3 \text{ GW/cm}^2$ favored ASE. This ASE exponentially decreased to zero as the pump intensity was increased from 0.3 to 10 GW/cm^2 . Pump intensities $> 10 \text{ GW/cm}^2$ show purely 4WM processes. The key conclusions are that the FWM can overtake ASE at large intensities; and the FWM intensity observed were $I_{FWM} \propto I_P^2$ (where I_P is the pump laser intensity).

The primary take away is that multiple linear and nonlinear process can coexist with MWM. However, past work has shown that the MWM processes favor large intensities (GW/cm^2). Our unfocused pulsed dye pump intensities are $< 5 \text{ MW/cm}^2$. Therefore intensity will be an important distinguishing characteristic. We expand on this discussion in Chapter V.

1.3 Dissertation Contributions

Prior two-photon excitation of alkali vapors have observed the production of blue wavelengths, with minimal IR observations, and much of this work was in Rb. The only work done in cesium was by Sulham [97] and Hamadani [45]. Sulham looked at the Cs 7D states with a much weaker two-photon cross section; and Hamadani focused on the production of blue and photo-ionization. However, it is important to consider both an optimal two-photon pump and every possible emission process simultaneously to understand which processes perform the best and the impact they could mutually have on a future system. We expand on this previous work by observing the efficiency and lasing threshold of every lasing line from 386 nm to 3614 nm with a wider range of densities of 10^{14} to $10^{17}/\text{cm}^3$, He buffer gas pressures of 0 to 10 Torr, and pump energies of 1 to 30 mJ/pulse. We therefore assess the feasibility of a wavelength agile DPAL on nearly every dipole allowed transition below the Cs 6D states. Our wide range of both operating conditions and data collection wavelengths capture additional

nonlinear processes and provides a detailed picture of how the energy flows through all (excluding the 6D to 7P transitions) possible transitions. Previous work using surrogate pulsed pump sources only speculated that a CW wavelength agile DPALs may be possible [64, 36, 97]. In this work we also assess the utility of CW pumping to further demonstrate the utility of two-photon DPAL.

We build off prior nonlinear optics work. Previous multi-photon nonlinear processes in alkali vapors required focused pump intensities $\sim \text{GW}/\text{cm}^2$, and were limited to MWM or SRS. Our pulsed work analyzes multi-photon processes from an unfocused pump over longer path lengths and we show innovative two-photon lasing process are theoretically possible. Similar observations occur at kW CW intensities.

1.4 Organization of Dissertation

Chapter II contains background theory on two-photon absorption and emission, and other important alkali vapor properties. In Chapter III we cover key experimental methods and techniques such as heat pipe design and operation.

Chapters IV and V cover the pulsed laser demonstrations. This chapter explores on-resonance one-photon cascade lasing processes from high intensity pulsed two-photon pumping.² In Chapter V we report off-resonance non-linear behavior from high intensity pulsed two-photon pumping.³ This chapter details two two-photon processes that may be produced by two-photon lasing or Six Wave Mixing (SWM).

Chapters VI and VII are from our low power, focused, CW two-photon excitation work. In Chapter VI we report our results on a 3 μm CW cascade lasing from a two-photon pump.⁴ In Chapter VII we develop a hyperfine theory for two-photon

²Chapter IV is based on a draft publication titled: “Efficient cascade lasing on over 17 wavelengths from two-photon excitation of cesium 6²D.”

³Chapter V is from a second draft publication titled “Two-photon and nonlinear stimulated emission from two-photon excitation of cesium 6²D.”

⁴Chapter VI is from a third draft publication titled “Continuous-wave 3 μm cesium laser on the 7² $P_{1/2} \rightarrow 7^2S_{1/2}$ transition from a two-photon pump.”

absorption and refine the NIST values for the cesium $6^2D_{3/2}$ state which are currently only known to ± 30 MHz. In this chapter we also cover the non-linear and off-resonance behavior from our CW two-photon pump. Finally we present our concluding remarks in Chapter VIII.

II. Background and Theory

In this chapter we cover some required background theory on two-photon processes and properties of the alkali vapors.

2.1 Rational for Alkali vapor

The alkali vapors have several unique properties that make two-photon pumping, both cascade and two-photon lasers, possible.

Large Two-photon Cross-Sections

Alkali vapors have very large two photon cross-sections, perhaps the strongest in nature. Thorough derivations for the two-photon absorption cross section can be found in Appendix A and B. For the degenerate case the Two-Photon Absorption (TPA) cross section is from equation 99, from Appendix A and additional details can be found in [57, 58, 26, 9]. We define the two-photon absorption cross section $\sigma_{if}^{(2)}$ from equation 100 as:

$$\sigma_{if}^{(2)}(\nu) = \frac{\pi^2 \nu}{5 h^3 c^2 \epsilon_0^2} \sum_n \frac{1}{g_i g_n} \frac{\mu_{fn}^2 \mu_{ni}^2}{(\nu_{ni} - \nu)^2} \rho_f(\nu_{fi} = 2\nu) \quad (1)$$

or

$$\sigma_{if}^{(2)}(\nu) = \frac{9}{5} \frac{\nu}{256 \pi^4 h c^2} \sum_n \frac{g_f}{g_i} \frac{\lambda_{fn}^3 A_{fn} \lambda_{ni}^3 A_{ni}}{(\nu_{ni} - \nu)^2} \rho_f(\nu_{fi} = 2\nu) \quad (2)$$

where i indicates the initial state, f the final state, A_{fn} is the A coefficient for the $f \rightarrow n$ transition, μ_{fn} is the dipole moment, λ_{fn} is the wavelength, g is the degeneracy, $\nu_{ni} - \nu$ is the frequency separation from the virtual state to the real state n , ρ_f is the density of final states or lineshape, and the sum over n represents a sum over

the resonant-real energy levels.¹ We have set the index of refraction to 1. The factor of 1/5 arises from the angular average of the two dipole moments and only applies to the case where the two exciting fields are in the same direction with the same polarization. This cross section is defined by the upward transition rate:

$$\left\langle R_{i \rightarrow f}^{(2)} \right\rangle = \sigma_{if}^{(2)} \frac{I^2}{h\nu} \quad (3)$$

Using equations 1, 3 and the notation of Figure 1 we can write the rate equation for the two-photon stimulated process:

$$\frac{dn_4}{dt} = -\sigma_{41}^{(2)} \frac{I^2}{h\nu} \left(n_4 - \frac{g_4}{g_1} n_1 \right) \quad (4)$$

where $\sigma_{41}^{(2)}$ is the stimulated emission cross section, and n_4 and n_1 are the population densities of levels $|4\rangle$ and $|1\rangle$. For both the degenerate and non-degenerate cases, the order of magnitude of $\sigma_{if}^{(2)}$ is governed by $\nu \lambda_{fn}^3 A_{fn} \lambda_{ni}^3 A_{ni} / (\nu_{ni} - \nu)^2$. That is, the magnitude of the cross-section is proportional to the product of the Einstein A coefficients, and the alkali vapors have some of the largest A coefficients. Next, the position of the P states are conveniently close to resonance and the $\nu_{ni} - \nu$ term(s) or $\Delta\nu$ are relatively small. All these factors give large two-photon cross sections on the order of $10^{-20} \text{ cm}^4/\text{W}$ to $10^{-24} \text{ cm}^4/\text{W}$ for various alkali transitions.

Although our experimental work is in cesium, there are no clear experimental values for cesium vapor's two-photon cross sections. As a quick comparison with experiment, we take the data for the rubidium $5^2S_{1/2} \rightarrow 5^2D_{5/2}$ recently studied at AFIT by Gallagher [36]. These calculations start with the Einstein A coefficients in Table 1. We use $\mu_{ni}^2 = g_n \frac{3\epsilon_0 h \lambda_{ni}^3}{16\pi^3} A_{ni}$ to be consistent with most sources.

¹This form also implies that we multiply by a sum over the upper degeneracies divided by the sum of the lower degeneracies. We define $\mu_{ni}^2 = g_n \frac{3\epsilon_0 h \lambda_{ni}^3}{16\pi^3} A_{ni}$ and recover the expected ratio $\frac{g_f}{g_i}$ in the final expression.

Table 1. A coefficients and dipole moments for ^{85}Rb

	$\lambda_{ij}(\text{Vac})$	$A_{ij}(10^7 \text{1/s})$	$\mu_{ij}(10^{-29} \text{C} \cdot \text{m})$	Ref
$5^2S_{1/2} \rightarrow 5^2P_{1/2}$	794.978nm	3.6129	5.0754	[96]
$5^2S_{1/2} \rightarrow 5^2P_{3/2}$	780.24nm	3.8117	7.1623	[96]
$5^2P_{3/2} \rightarrow 5^2D_{5/2}$	775.978nm	0.529	2.30	[71]
$5^2P_{3/2} \rightarrow 5^2D_{3/2}$	776.156nm	0.088	-	[71]
$5^2P_{1/2} \rightarrow 5^2D_{3/2}$	762.102nm	0.41	-	[71]

Using the data in Table 1 and equation 99 with a Doppler broadened line-shape at $T=400 \text{ K}$, $\Delta\bar{\nu} = 35 \text{ cm}^{-1}$, $\rho_3(2\nu_0) = .94/\delta\nu_D$, $\delta\nu_D = 7.16 \times 10^{-7} \nu_0 \sqrt{T/M}$, and $\nu_0 = 2\nu_{\text{pump}}$, the TPA cross-section is reported in the first row of Table 2. The remaining rows in Table 2 provide a comparison with experimental values, many were taken with sub-Doppler measurements and converted by Gallagher to Doppler broadened measurements. We can see that our theory accurately captures the value within the uncertainty of the upper A coefficient ($\sim 50\%$).

The values provided in Table 2 may seem small, but in the context of TPA cross sections, these are extremely large. For comparison, typical TPA cross sections are $\approx 3 \cdot 10^{-31} \text{ cm}^4/\text{W}$ [10, 108], where [108] gives data from many laser dyes. In 2001, Drobizhev [30] reported a “record high intrinsic” TPA cross-section of $\approx 2 \cdot 10^{-27} \text{ cm}^4/\text{W}$ in a dendrimer synthetic polymer.

The last notable detail is that in the non-degenerate cases, we can tune the two-photon cross-sections into resonance (decreased $\Delta\nu$) but at some point the perturbation theory will break down (e.g. in Section 1.2 we note that $\Delta\nu < 30 \text{ cm}^{-1}$ results in an on-resonance dip). We return to this non-degenerate theory in Chapter V. The two-photon gain can be further improved by increasing the number density.

Table 2. Degenerate two-photon absorption coefficient for the $5^2S_{1/2} \rightarrow 5^2D_{5/2}$ transition comparison with experimental data. Note the cross sections from [109, 17, 82] were corrected by Jeff Gallagher for Doppler free to Doppler Broadened

Doppler-Free $\sigma^{(2)}(cm^4/W)$	Doppler Broadened $\sigma^{(2)}(cm^4/W)$	Isotope	Theory/Exp.	Ref
5.02×10^{-18}	7.72×10^{-21}	85	Theory	This work
4×10^{-20}	1.30×10^{-21}	87	Exp.	[109]
1.2×10^{-18}	5.58×10^{-20}	85	Exp.	[17]
6.5×10^{-20}	3.51×10^{-21}	85	Theory	[82]
5×10^{-20}	2.7×10^{-21}	85	Exp.	[82]
N/A	6.78×10^{-21}	85	Exp.	[36]
N/A	2.5×10^{-21}	85	Theory	[36]

Large Number Densities

A summary of the alkali vapor pressure curves is given in Figure 3. The blue lines show the number densities from vapor pressures up to the melting point. In this work with Cs we use temperatures of 150 to 330 °C or densities of 2.0×10^{14} to $8.8 \times 10^{16}/cm^3$. We treat this as the ground state density, however, at high temperatures this population may have to compete with Cs_2 dimer population.

We can see that K, Rb and Cs can obtain practical number densities for laser devices at reasonable temperatures. The success of DPAL devices is directly related to this property. Na and Li have vapor pressure curves shifted to the right, and therefore require much higher temperature to obtain similar number densities.

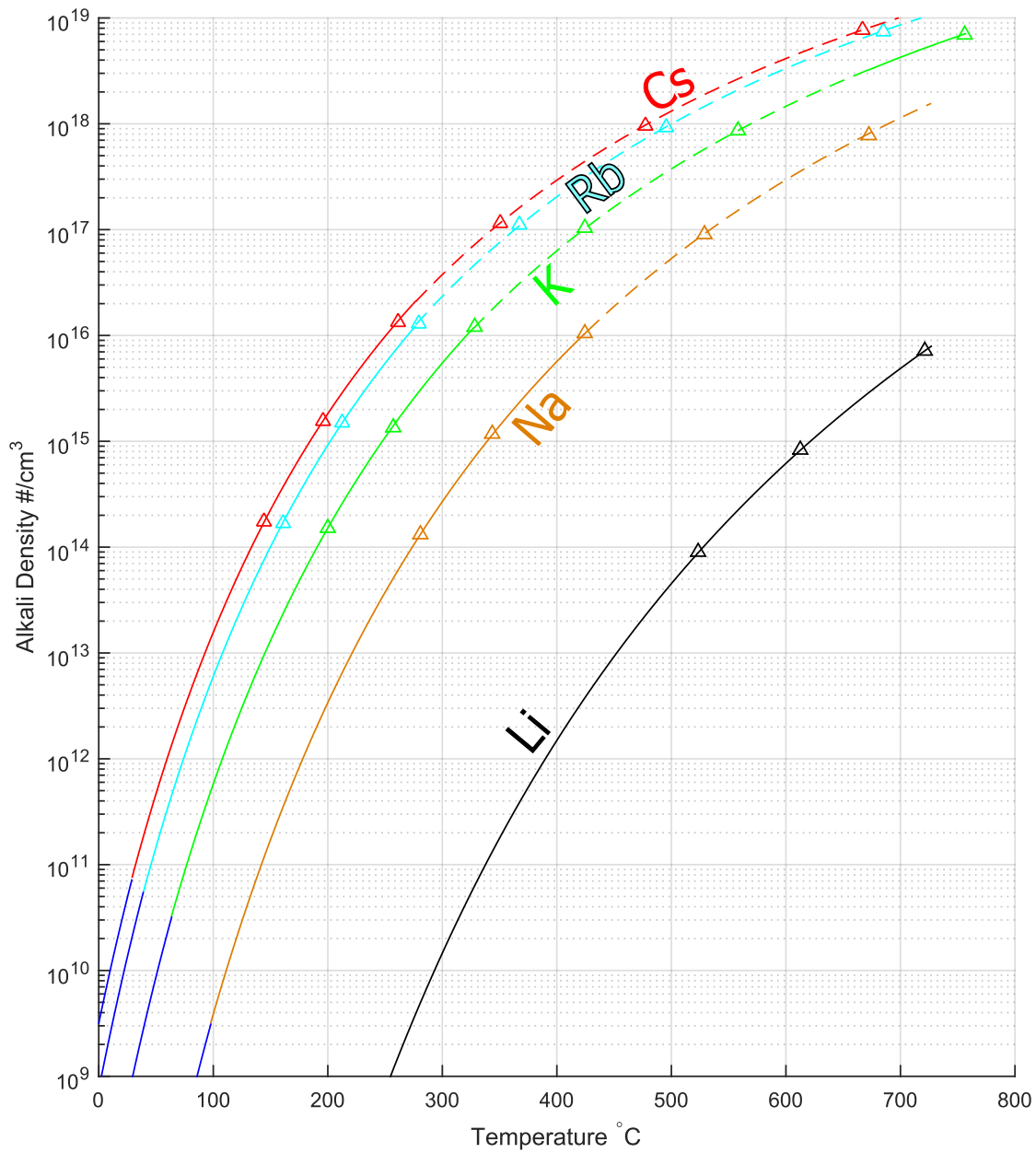


Figure 3. Alkali Vapor Pressures. Solid blue lines give solid phase data, dashed lines give extrapolated or interpolated data [2, 8]. Δ =CRC data [47].

Large Fine Structure Mixing Rates

Another useful property of the alkali vapors is their fast kinetics. Specifically, the spin-orbit mixing between the upper excited doublet states Cs 7^2P , 6^2D for Cs-He and Cs-Cs collisions. For Cs-He collisions this process is represented as



where M_J is any alkali metal in state J. We return to the discussion of rates in Chapter IV where we provide rates for the Cs-He and Cs-Cs collisions for the 7P and 6D states in Table 4. As an example the $6^2D_{5/2} \rightarrow 6^2D_{3/2}$ mixing rate is

$$k_{\frac{5}{2} \rightarrow \frac{3}{2}} = \sigma_{HeCs6D} \bar{g}_{HeCs} n_{He} + \sigma_{HeCs6D} \bar{g}_{CsCs} n_{CsT} \quad (6)$$

where σ_{HeCs6D} is the fine structure mixing cross section due to He-Cs collisions of $6^2D_{5/2} \rightarrow 6^2D_{3/2}$, the average collision speed is $\bar{g} = \sqrt{8kT/\pi\mu}$, n_{He} is the He number density, n_{CsT} is the Cs number density at temperature T, and the reduced mass $\mu = m_1 m_2 / m_1 + m_2$ (note that $\bar{g}_{HeCs} \neq \bar{g}_{CsCs}$). For 5 Torr He (at room temperature fill) and 237 °C heat pipe operating temperature, and cross-section values from Table 4 this rates is 6.2 ns which is comparable to our 10 ns pump pulse width. This fast mixing enables inversions on transitions that would not otherwise be possible. This includes cascade lasing inversions (Chapter IV) as well as two-photon lasing inversions (Chapter V).

III. Experimental Methods

In this chapter we discuss the evolution of our experimental work. This includes some experiments that were of limited interest/success, as well as the procedures for proper operation of our Cs heat pipe.

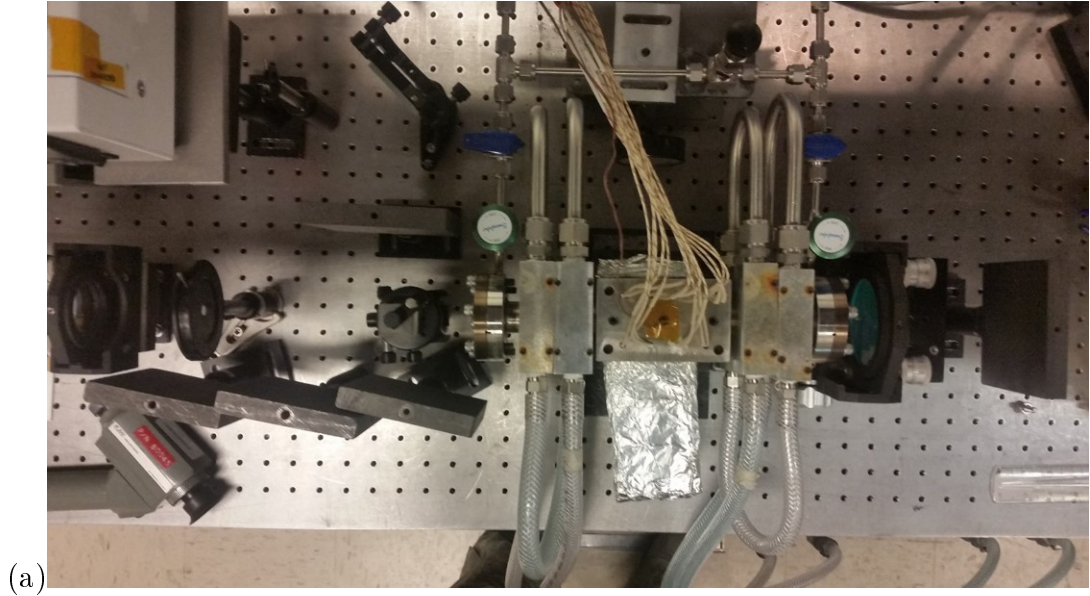
3.1 Initial Experimental Work

Our initial lab work focused on two experiments that were intended to serve as learning tools for the lab equipment as opposed to beneficial dissertation results. In this section we cover these inauspicious results and experiments in the hopes that future students can benefit from them. The two experiments in this section are an SRS experiment and a Cs DPAL.

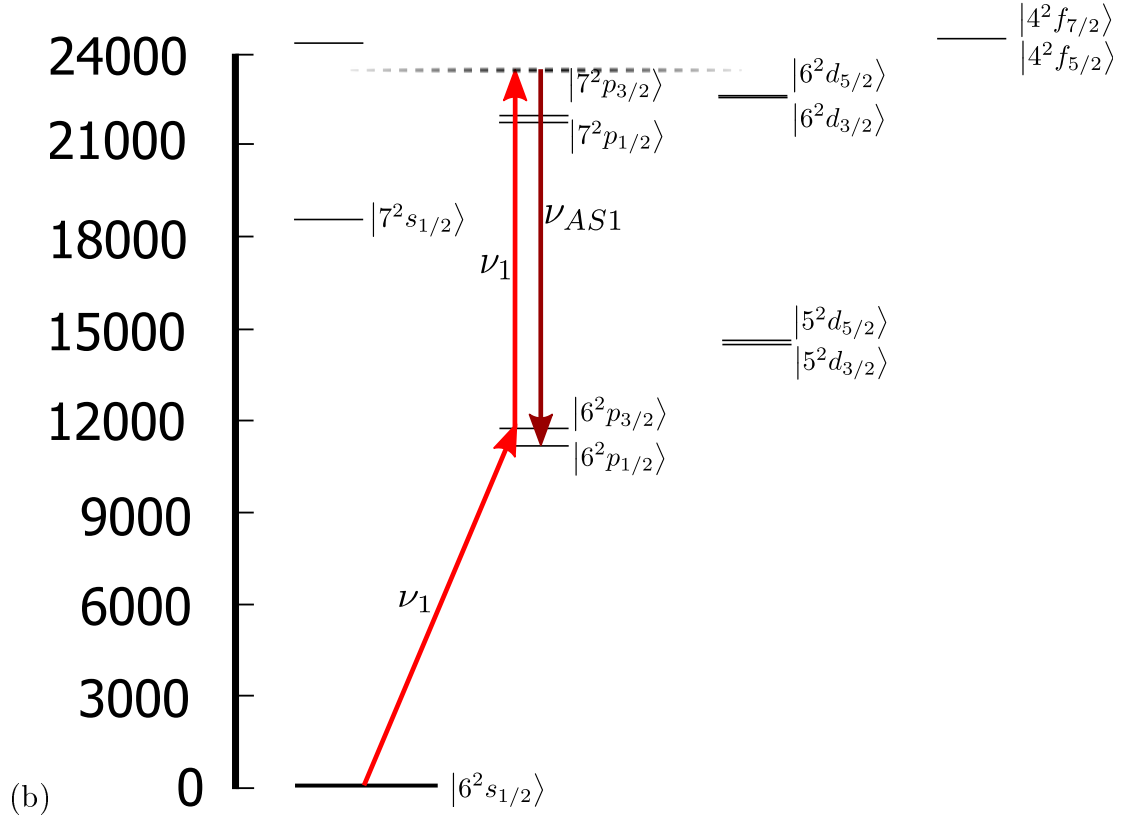
Our first experiment was an attempt to repeat Brown's potassium SRS experiment [14], in cesium, with some minor modifications. In this experiment a pulsed dye laser pumped the system to one of the 6P states, and we hypothesized SRS off one of the upper 7S or 6D states to the other 6P state. A cavity was used in an attempt to enhance the effect. Figure 4 shows the energy levels for generating the first anti-stokes line ν_{AS1} .

No SRS was observed. Our first speculation is that the A coefficients suggest that SRS to 7P states will have higher Raman gain. Another potential explanation is that the wings of the pulsed dye laser in potassium experiment played a role in seeding the processes (it is not uncommon in nonlinear optics for this to occur [76, 15]). By contrast the shift in cesium is from 852 nm to 813 nm well outside the pulsed dye tuning curve.

Next we made a basic Cs surrogate DPAL with ethane and the same cavity configuration to see if the heat pipe was working properly. We show these results in



(a)



(b)

Figure 4. Cs vapor SRS Experiment. a) Picture of our cavity around the heat pipe, before our temperature zone reconfiguration. b) The energy levels for the SRS experiment.

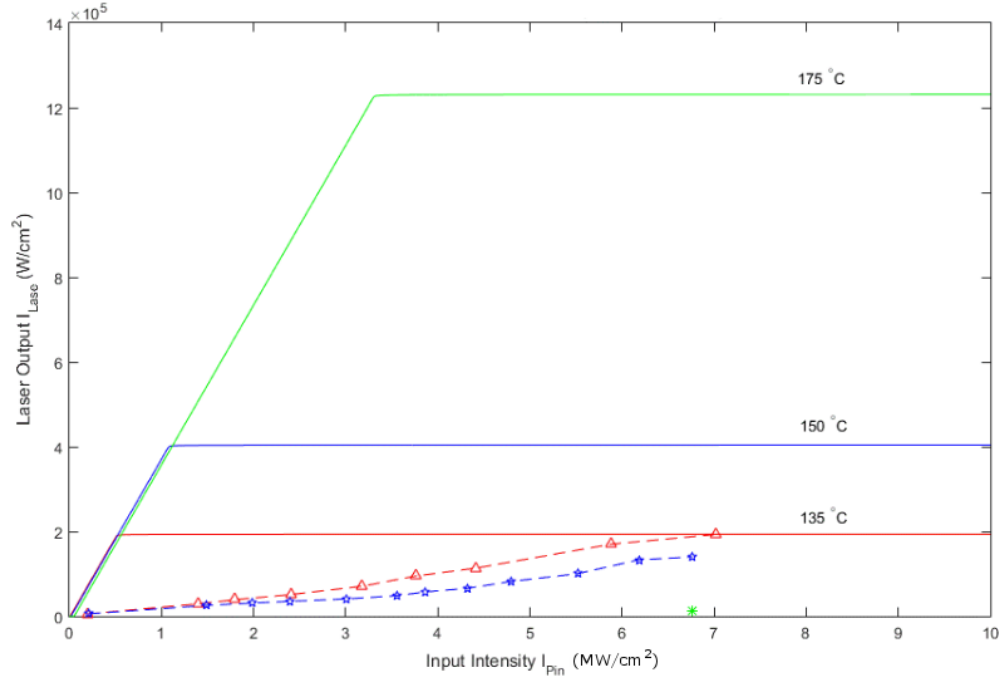


Figure 5. Performance of a Cs ethane optically pumped alkali laser to test heat pipe operation. Heat pipe was filled at room temperature with 400 Torr of ethane. The solid lines show the theoretical output from a line narrow 3 level DPAL model at **135 °C**, **150 °C**, and **170 °C**. The dashed lines and points show our data.

Figure 5 with a comparison to a 3 level DPAL model. This simple check showed that the heat pipe was working to some degree at low temperatures. There were also large window losses from contamination, reactions with the ethane, and poor resonator alignment, all of which could be a source of poor slope efficiency, but we digress. More importantly, performance degraded significantly at higher temperatures, although other problems were clearly present.

In summary, these initial experiments showed that high temperature heat pipe operation, heat pipe cleanness, and pulsed dye laser wavelength calibration would all be issues.

3.2 Cesium Heat Pipe Operation

Alkali heat pipes are generally made from a stainless steel tube lined with mesh to hold the molten alkali metal. Further details on heat pipe construction materials can be found in [28]. Assembly requires a well purged dry box, and a spring holds the mesh in place. This initial assembly is followed by bake out of key components under vacuum at higher temperatures than required for experiments to drive out water vapor and H_2 gas. The dry box must be used again to add 1 to 3 gm of alkali to the mesh near the center of the heat pipe. For low temperature cesium experiments, operation consists of a central oven region (up to 130 °C) and chilled outside (10 to 18 °C) the oven region to prevent contamination on the windows. However, this creates issues at higher oven temperatures as we detail below. In a successful heat pipe, this temperature gradient sets up a convective flow where the vapor generated at the central oven, flows out to and condenses on the cold mesh, and capillary forces would replenish the liquid alkali in the central region. However, we operate the system in an oven mode for these experiments. Control of thermal gradients and temperature zones of the heat pipe is critical to long term stable performance.

Some of the major issues associated with transitioning from low to high temperature operation include:

A true heat pipe is a thermal device used to cool a hot region with a cold region some distance away. To operate a true cesium heat pipe, temperatures exceeding 450 °C are required [28]. This is due to several viscous limitations and drag forces created by the liquid vapor interaction region.

Past heat pipe experiments were performed at low temperatures and did not move enough vapor to establish a significant convective flow. To minimize window contamination, temperatures outside of the oven region were controlled to 10 to 18 °C. For example, at 120 °C the mass flow rate is constrained by the vapor pressure

or number density $\sim 5 \times 10^{13}/\text{cm}^3$ and we estimate the flow to be laminar $v \approx \frac{r^2}{8\mu} \frac{\Delta P}{L}$ (see [28] Chapter 2). Note this is driven by a ΔP pressure drop so if the heat pipe is not sufficiently evacuated there will be diffusion through other gases). Under these conditions the mass flow rate is approximated by

$$\dot{m} = m_{Cs} n_{CsT} \frac{\pi r^4}{8\mu} \frac{\Delta P}{L} \quad (7)$$

where r is the pipe radius (1.25 cm) (L is the axial distance (~ 20 cm) that describes the pressure drop, μ is the cesium vapor viscosity (temperature dependent), and $m_{Cs} = 132$ amu. For our condition we have $v \approx 11$ m/s, $\dot{m} \approx 6.4 \times 10^{-8}$ kg/s, and our 1 gm supply will last several hours if it is not resupplied by the wick. Our experience suggests this is likely even slower. If we repeat this for 330 °C, the ratio of mass flow rates is $\dot{m}_{330}/\dot{m}_{120} \approx 4 \times 10^6$. Hotter temperatures move vapor much faster because the cesium density and pressure gradient both increase exponentially.

At higher temperatures (but still below the critical 450 °C), the wicking will begin to deplete but the heat pipe is still not hot enough for capillary forces to move liquid cesium back in place fast enough. This problem is further exacerbated when the chiller zones are set to temperatures below alkali freezing point. A second issue is that there is so much alkali partial pressure that vapor still gets to the windows. We show some examples of the escalation of window contamination in Figure 6. Also, the condensed alkali on the cold zones is trapped because its below the freezing point. We have observed cesium stalactites that eclipsed the pump laser beam.

To rectify these high temperature issues, we added two requirements. First, the entire mesh lattice must be saturated with liquid alkali. The second requirement is to insure that the cold regions are above the melting point so that no alkali is frozen or lost to the circulation process. These requirements are also given in reference [28]. However, we still need to protect the windows, and one way to achieve this at high

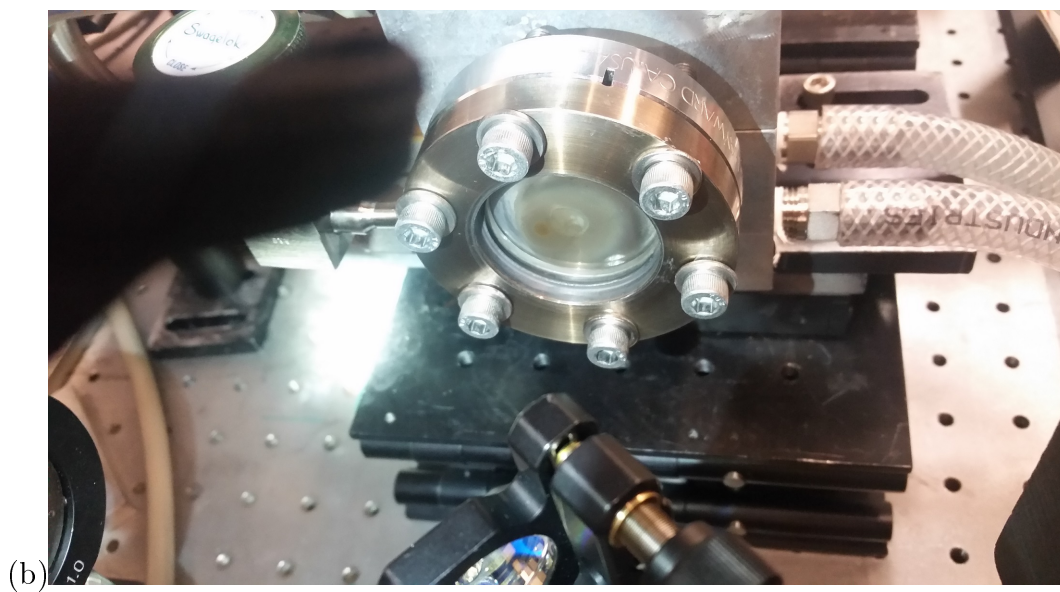
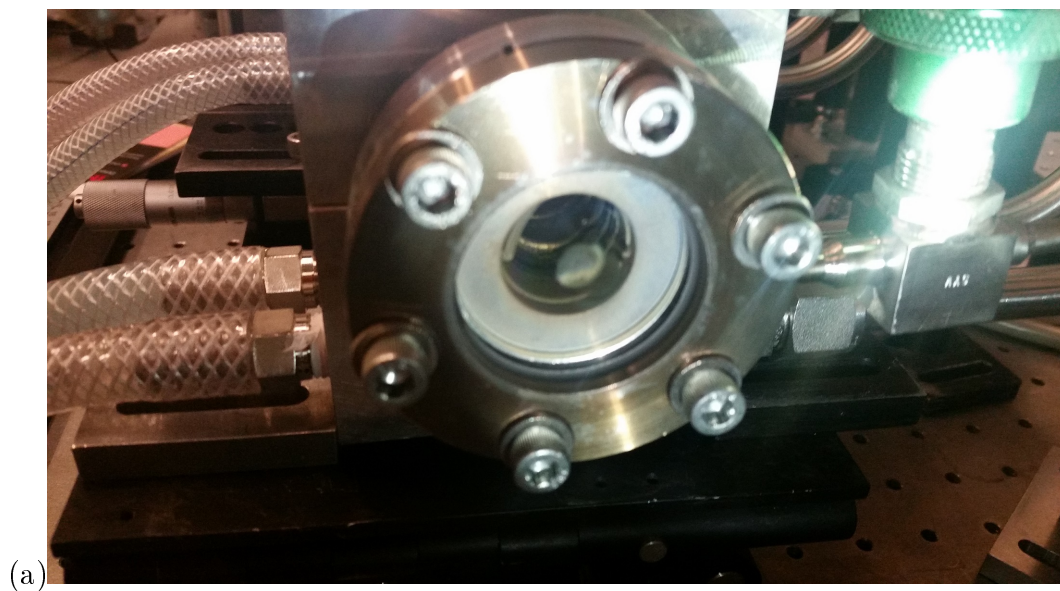


Figure 6. a) The start of semi-permanent window contamination. b) Fully developed window contamination.

temperatures is to insure the windows are not colder than the cold zones. Through trial and error we converged on 180 to 200 °C window temperatures, 38 °C cold zones (10 °C above the melting point), and 49 °C intermediary zones. This worked well if the oven zone was below 240 °C, maybe 270 °C.

The final heat pipe operating procedure, was how to set the temperature zones when in the off or standby state. Because we are still below the recommended cesium heat pipe temperature, the liquid alkali needs more time to flow back to the center. We accomplish this condition in a “turn off mode” by imposing an inverted gradient when off. The windows were kept above 160 °C, the cold zones were left at 38 °C, and the intermediary zone and central oven zone were allowed to reach near room temperature ~ 30 °C, still slightly above the melting point.

In addition to heat pipe operating procedures, significant work was required to minimize any contamination sources. To this end, we replaced much of the vacuum lines and installed a getter on the helium supply near the cell. The culmination of these operating procedures, new vacuum lines, and filtration, lead to an astonishing >18 months with no maintenance (e.g. no window replacements or addition of Cs, which was previously a weekly to bi-monthly process).

3.3 Pulsed Experiment Configuration

Our first good sets of experimental data came from two-photon pumping the cesium 6D states with our improved heat pipe. We detail exact procedures and equipment in Chapters IV and V of this document, and in this section we introduce the basic setup, appearance, and techniques. Our pulsed apparatus is shown in Figure 7 with pictures in Figure 8. In addition to the heat pipe, the other important piece of equipment is a pulsed dye pump source with LDS 867 dye that provided 1- 30 mJ or 150 kW to 5 MW/cm² of peak pump intensity, with in a 10 ns pulse at our (883 nm

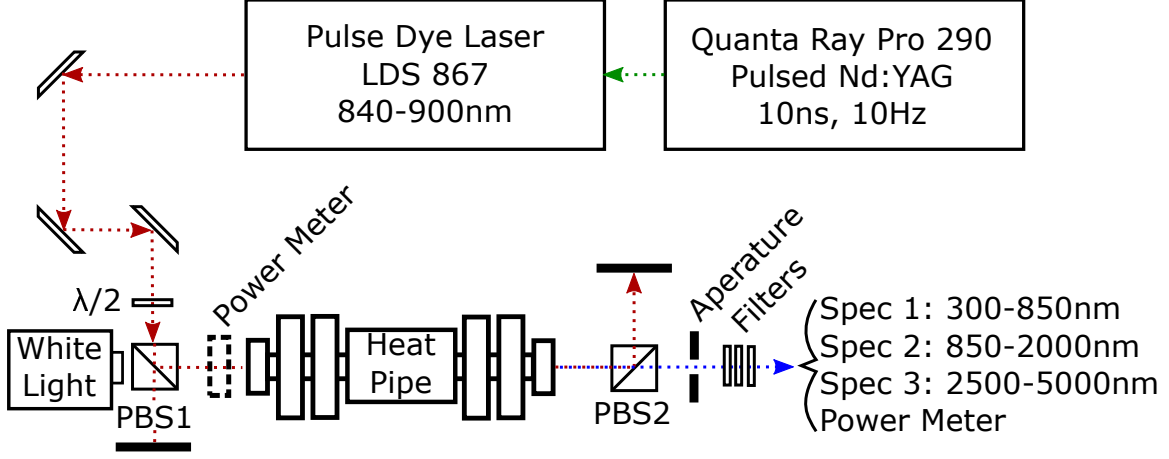


Figure 7. Experiment apparatus for pulsed work.

to 885 nm) two-photon wavelengths. A key advantage of this setup is that no pump focusing lens is required, and we therefore have longer path lengths.

The challenge in working with this source is that the pulse energy exhibits 20% pulse to pulse power or energy variation. This is worse if the pulsed dye laser energy output is tuned by varying the flashlamp energy of the Quanta Ray Nd:YAG laser. For example, alignment of optics requires low power, so we set the amplifier (flashlamp energy) on the Nd:YAG laser to near zero. However, this can change both the temporal and spatial intensity profiles. We minimize these effect by leaving the laser at full power, and tuning the power with a half wave plate (broadband over our wavelengths of interest) and Polarization Beam Splitting (PBS) cube. In other words, the Quanta Ray is always set to full so the spatial and temporal profile is consistent. Furthermore, we track the variation by using energy meters as opposed to just an average power measurement. The energy meter has the advantage of a smaller form factor to manipulate around optics, we can always average ~ 500 pulses and have both the average energy/pulse and the standard deviation (generally around 10%).

The next step was to collect data on lasing lines produced from this pump. The strong pump is convenient, but it makes putting sensitive instruments in and around

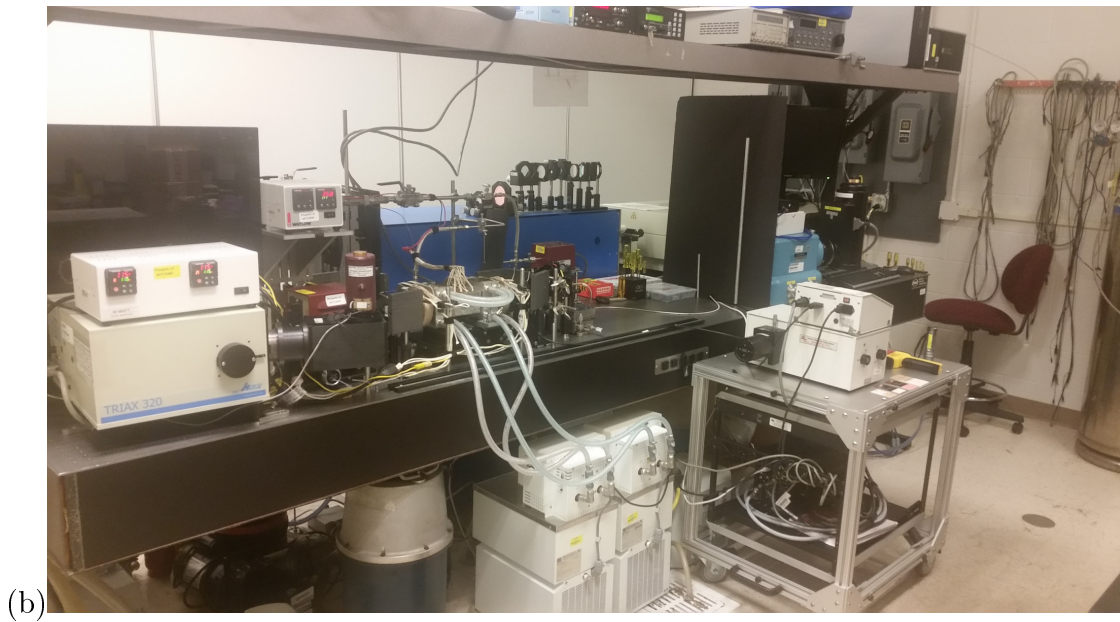
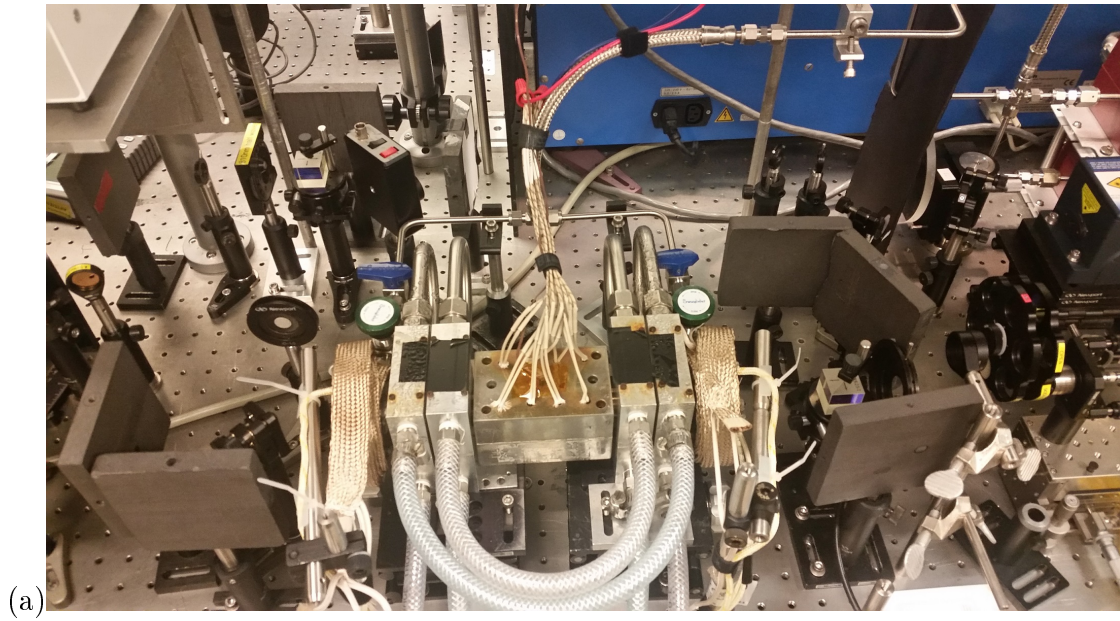


Figure 8. Pictures of the experiment apparatus for pulsed work: a) close up of the heat pipe with PBS cubes and fiber collimators to the spectrometers, and b) extended view with the three spectrometers.

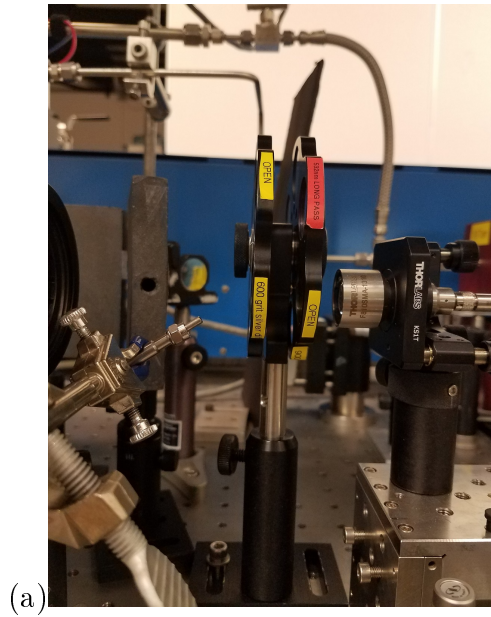


Figure 9. Close up of the collection optics. a) Heat pipe pumped from the right, left is a fiber looking at a 600 grit silver diffuser plate that goes to our spectrometer in the 300-850 nm range; with the filter wheel set to move the diffuser plate, the laser light hits the collimator on the right that goes to our 900-2000 nm spectrometer. b) Heat pipe is pumped from the left, output hits a fused silica substrate high power mirror to filter out the pump and let 3000 nm light through; this light hits two gold mirrors and a 35 mm CaF_2 coupling lens to our 3000-5000 nm spectrometer.

it difficult. Past AFIT work used prisms to split the beams. However, this made it difficult to estimate the true intensity of the output beams and each output wavelength could only be monitored one at a time. We instead favor the use of a second PBS cube to act as a filter for the pump. Our lasing lines are produced by ASE and we assume they are unpolarized; the PBS cube this will therefore hurt our transmission by $\sim 50\%$ and we can measure this transmission and correct for it later. The advantage of this approach is that we can now point sensitive spectrometers at the heat pipe and cover a large span of wavelengths much faster and get accurate energy output measurements for each of the beams.

We cover a range of 300 nm to 3700 nm with three spectrometers. The details for coupling the spectrometers are shown and explained in Figure 9. We were not able to take data with all three spectrometers simultaneously, so measurements were repeated under conditions of interest for each spectrometer. We also pumped the heat pipe from left to right for the visible and near IR spectrometer, and from right to left for the IR spectrometer. Each of the spectrometers are equipped with an appropriate detector or array for the wavelength range, and the integration times long enough to average 500 pulses or more. For the visible and near IR spectrometer (arrays) with was down through software, for the IR spectrometer (single element detector), measurements were taken with an o-scope and we averaged the area of the pulse trace for 500 pulses.

Many of the lasing lines could be measured directly with a assortment of filters and sensitive energy meter. For example, after the pump beam is filtered by the PBS, the D_2 line at 850 nm could be measured with at 850 nm band pass filter with 10 nm band pass, followed by a energy meter. We detail the 12 filters and measurements in further detail in Chapter IV.

In Figure 10 we show the apparatus with the pump set to the Cs $6^2D_{3/2}$ two-

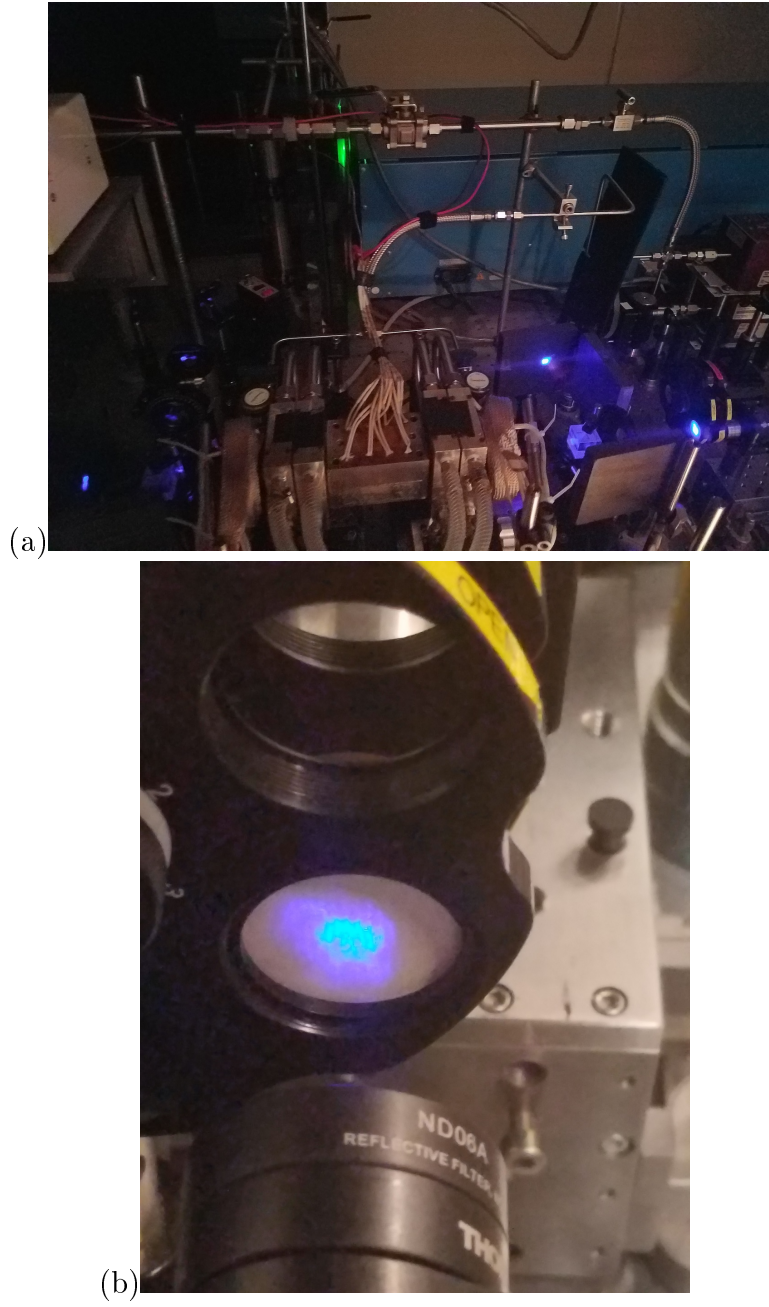


Figure 10. Pulsed excitation of the Cs $6^2D_{3/2}$ state at 200°C, 15 mJ/pulse pump. a) View of the heat pipe. b) zoomed in view of the silver diffuser where the blue beam profile mimics the spatial intensity pattern of the pulsed dye pump.

photon wavelength, the heat pipe oven set to 200 °C, and the pump energy set to 15 mJ/pulse. In Figure 10b we can also see that the blue beam spot is clearly lasing. Although an impressive amount of blue is visible, in Chapters IV and V we show even more power in the IR lines.

3.4 CW Experiment Configuration

The CW laser experiment apparatus is shown in Figures 11 and 12. The major changes are the pump source is now a 2 W (1.6 W into heat pipe at our two-photon wavelengths) Ti:sapphire laser with < 50 kHz bandwidth. We again tune power with the half wave plate and Polarization Beam Splitting (PBS) cube. The spectrometers are removed in-favor of a single InSb detector with several filters. However, we have to focus the power down which gives us a very small gain length. Our focus spot diameter (radius ω_0) is estimated by

$$2\omega_0 = M^2 \frac{4\lambda_{pump}}{\pi} \frac{F}{D} = 59 \mu\text{m} \quad (8)$$

where $M^2 \approx 1.05$ is the beam quality for the pump, F is the focal length of the lens 150 mm (200 mm also used) and D is the beam diameter at the lens ~ 3 mm. The gain length will be similar to the Rayleigh range

$$L_g \approx Z_R = \frac{\pi\omega_0^2}{\lambda_{pump}} = 3.1 \text{ mm} \quad (9)$$

The primary uncertainty in these quantities that comes from the beam size on the lens after it has traveled through a 5x beam expander and propagated ~ 4 m. We estimate unfocused spot size with burn paper.

The data acquisition is also different. We use lock-in detection and send the output to an analog to digital converter that interfaces with a Matlab script to record

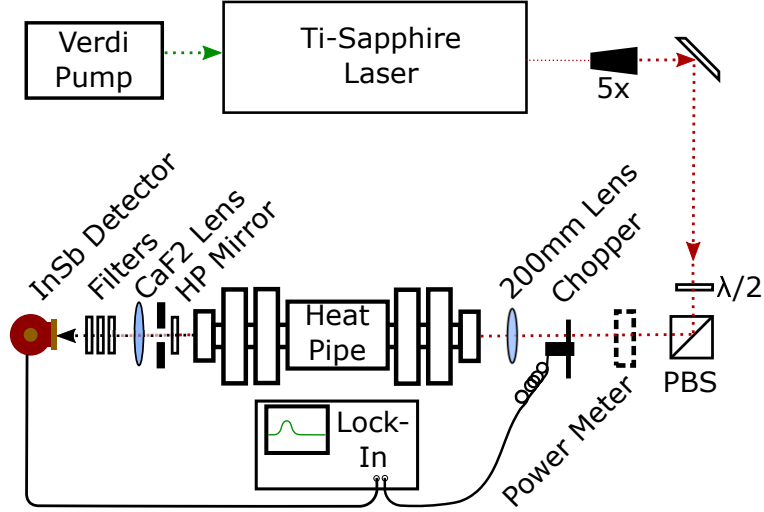


Figure 11. Experiment apparatus for CW work.

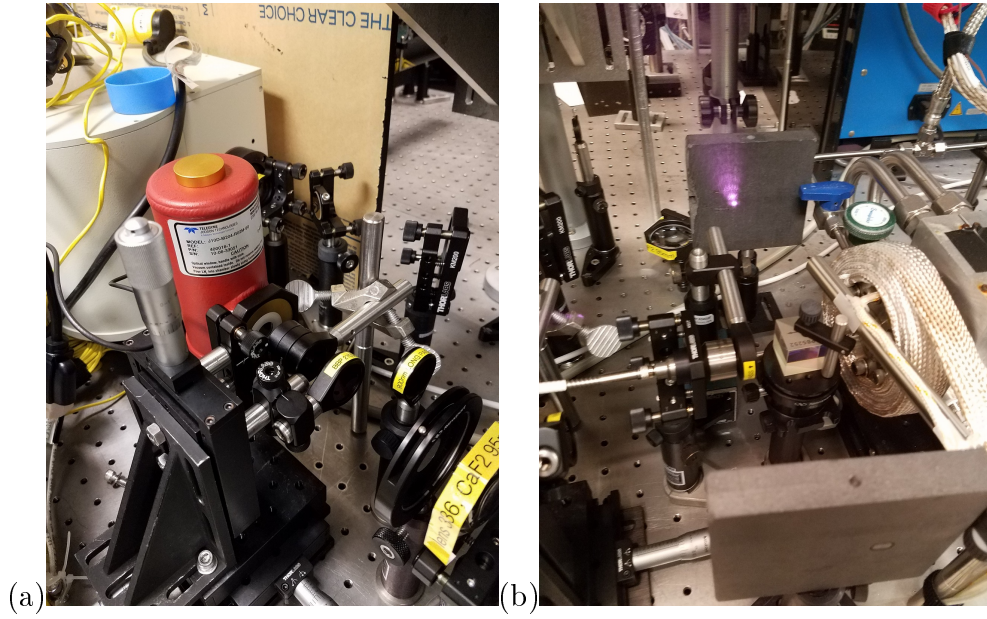


Figure 12. (a) Coupling of the heat pipe to the InSb cryo-cooled detector. (b) Fluorescence coupling to spectrometer, this was taken before we added a collimation lens between the heat pipe and the PBS cube.

pump laser wavelength (from a wave-meter) and data from the lock-in, this allows pump laser wavelength vs output laser spectra to be formed.

In addition to the apparatus shown in Figure 11, we also used two additional configurations. We use our near IR spectrometer to observe the fluorescence of populations involved in creation of the 3 μm laser. In Figure 12b, we show the modification to our experiment; the InSb detector is removed and replaced with a fiber collimator to our near IR spectrometer. We filter out the pump (to protect the spectrometer) in the same way as the pulse experiments: after the pump beam exits the heat pipe it is re-collimated with a lens hits a PBS cube and the power is dumped on a graphite block, the residual pump that passes through the PBS cube (about 1/500 to 1/1000) is blocked by an IBS coated high efficiency long pass filter with 5 orders of magnitude of blocking below 900 nm and $>99\%$ transmission from 900 nm to 2000 nm.

In the final configuration (not shown) we use the pump that comes off the second PBS cube and pick a small amount off and monitor the signal with a amplified large area Si-detector. We swap this out with the InSb detector on the lock-in and are now able to record absorbance spectra. In practice, this was only successful for our 5 Torr He cases, due to large sources of noise from the heat pipe such as etalon effects. We attempted to improve this by picking off some of the pump before the heat pipe, chopping it at a second frequency, directing it at the same Si-detector, and splitting the output to a second lock-in and the second chopper frequency. By monitoring the input of the pump power, we hoped to remove and pump wavelength variations which should have given us a signal to noise improvement of $\sim 10\times$. However, there was a lot of cross talk between the two legs, and wavelength transmission effects through the heat pipe were different enough that the second leg did not help. The pump also had to propagate ~ 10 m from an optics bench in an adjacent lab to reach the heat pipe so turbulence effects over the beam-path may also be significant. To bottom line

is that sensitive absorption effects take a lot of time and finesses to properly set up. Further details regarding the experimental equipment can be found in Chapter VI.

IV. Cascade Lasing from Two-Photon Excitation of Cesium 6^2D

In this chapter we report our on-resonance observations from two-photon pumping the cesium $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ and $6^2S_{1/2} \rightarrow 6^2D_{5/2}$ transitions. This sets up our understanding of the system as a whole, so that we can consider more complex nonlinear (two-photon) process in Chapter V. A broad survey is taken to assess the agility of the cesium 6^2D states in the generation of cascade lasing from 455 to 3650 nm. The experiment utilized a heat pipe with 0 to 10 Torr of He, cesium densities of 10^{14} to 10^{17} per cm^3 ; it is pumped, single-pass, by a pulsed dye laser with 1 to 30 mJ/pulse. Cascade lasing, generated by amplified spontaneous emission with no aid of a cavity, is observed on over 17 transitions, or nearly every dipole allowed transition below the $6D$ levels. Many lasing lines possess 100 to 229 μJ of energy, slope efficiencies of 4 to 7%, low thresholds favorable for diode pumping, and together have combined optical conversion efficiencies as high as $10\% \pm 3\%$. This agile system could have applications in infrared counter measures, target illuminator lasers, beacon lasers, underwater communication, and other tactical laser applications.

4.1 Background

Since proposed in 2001 [54, 55], the Diode Pumped alkali laser (DPAL) continues to be an efficient and viable option to phase together the energy from high power laser diodes. Recent demonstrations in DPALs have shown scaling to kW-class devices in both potassium [75] and cesium [6]; significant interest also exists in rubidium[75].

Although the DPAL is ideal for high efficiency applications, the operational laser wavelengths are constrained by the near infrared $D_1 \ ^2P_{1/2} \rightarrow \ ^2S_{1/2}$ transitions; i.e. 770, 795, and 894 nm in K, Rb and Cs. Extended wavelength ranges, or "wavelength agility," is desirable for many applications including beacons, illuminators, under-

water communications, and infrared counter measures [14, 72, 4]. To achieve these wavelengths of interest, many have looked into pumping to higher lying states of alkali vapors. There are three primary strategies found in literature that relate to cascade lasing:

1. Blue/UV pump $n^2S \rightarrow (n+1, 2)^2P$ [83, 78, 88, 13, 64]
2. Sequential pump $n^2S \rightarrow n^2P \rightarrow n^2D$ [114, 63, 85, 86, 101, 103, 1]
3. Two-photon pump $n^2S \rightarrow (n+0, 1)^2D$ [45, 97, 34]

The first method was suggested in 1958 (potassium) [83] and [78]. With the recent success of DPALs, it has been tested as recently as 2016 [13, 64]. It is widely agreed that lasing generated results from cascade lasing via Amplified Spontaneous Emission (ASE). In the context of DPALs, this scheme is limited due to the lower maturity of blue laser diodes.

Sequential and two-photon pumping of the alkali's have primarily focused on the production of blue lasing. The mechanism for blue generation (for methods 2 and 3) is generally thought to be some combination of ASE and four wave mixing [97, 34]. The sequential method is the most prevalent as it does not require an exotic (blue/UV) or intensity dependent (two-photon) source. Conversion efficiencies as high as 2.6% have been reported [101]. Potential advantages of two photon pumping include: simplified pump mechanism, larger population transfer to the upper level—population is not lost to an intermediate level—and larger range of potential wavelengths because of inversions to the intermediate level. In both pump cases, the question still remains as to whether the dominant mechanism is multi-wave mixing or cascade lasing. In this work we focus on two-photon pumping cesium and consider the entire wavelength space generated, we further limit the consideration to cesium because, large cascade lasing cross sections immediately follow the pump, and have the potential to achieve

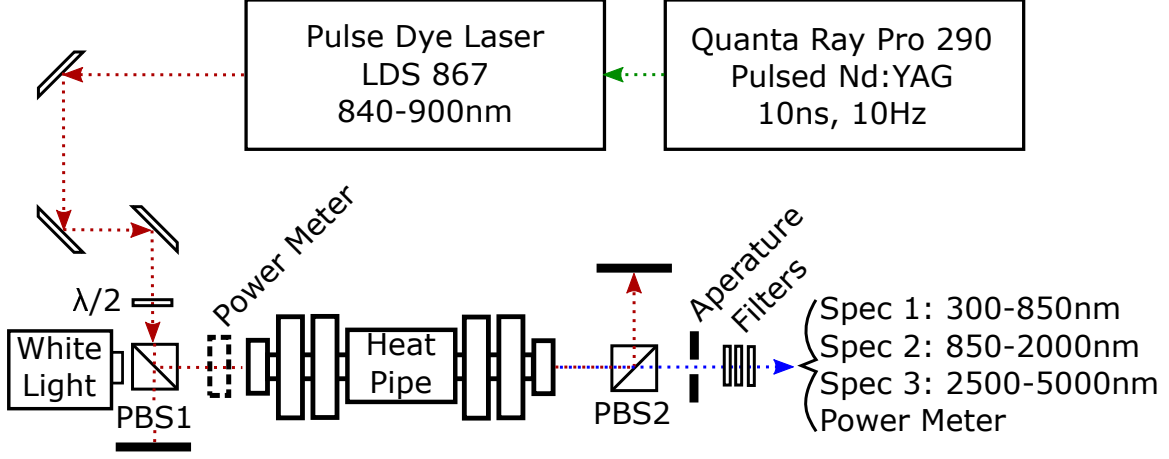


Figure 13. Experiment apparatus

a nearly depleted ground state. Other wavelength generation methods exist such as multi wave mixing [34], lasing without inversion [112], and Stimulated Raman/Hyper Raman Scattering [14, 105]. We see some of these effects, but we have isolated them for a later discussion to better focus on cascade lasing.

4.2 Experiment

The experiment utilizes a pulsed dye laser to single-pass-pump a mirror-less cesium heat pipe with normal incident uncoated sapphire windows. The dominant mechanism for lasing is Amplified Spontaneous Emission (ASE). Three spectrometers, and 13 filtered power measurements are used to characterize 17 cascade lasing transitions. These 17 transitions do not include the 12.1, 14.6 and 15.6 μm 6D to 7P transitions because they are blocked by the sapphire windows. A schematic is given in Figure 13 and the relevant energy levels with cascade lasing lines are illustrated in Figure 14.

The pump laser is a Sirah Precision Scan D pulsed dye laser, whose oscillator and two amplifiers are circulated with LDS 867 dye. It is pumped by a Quanta Ray Pro 290 Nd:YAG laser that is doubled to 532 nm, with around 930 mJ/pulse. This gives 10 Hz, 10 ns pulses tunable from 840 nm to 900 nm, with ~ 1.5 GHz spectral width.

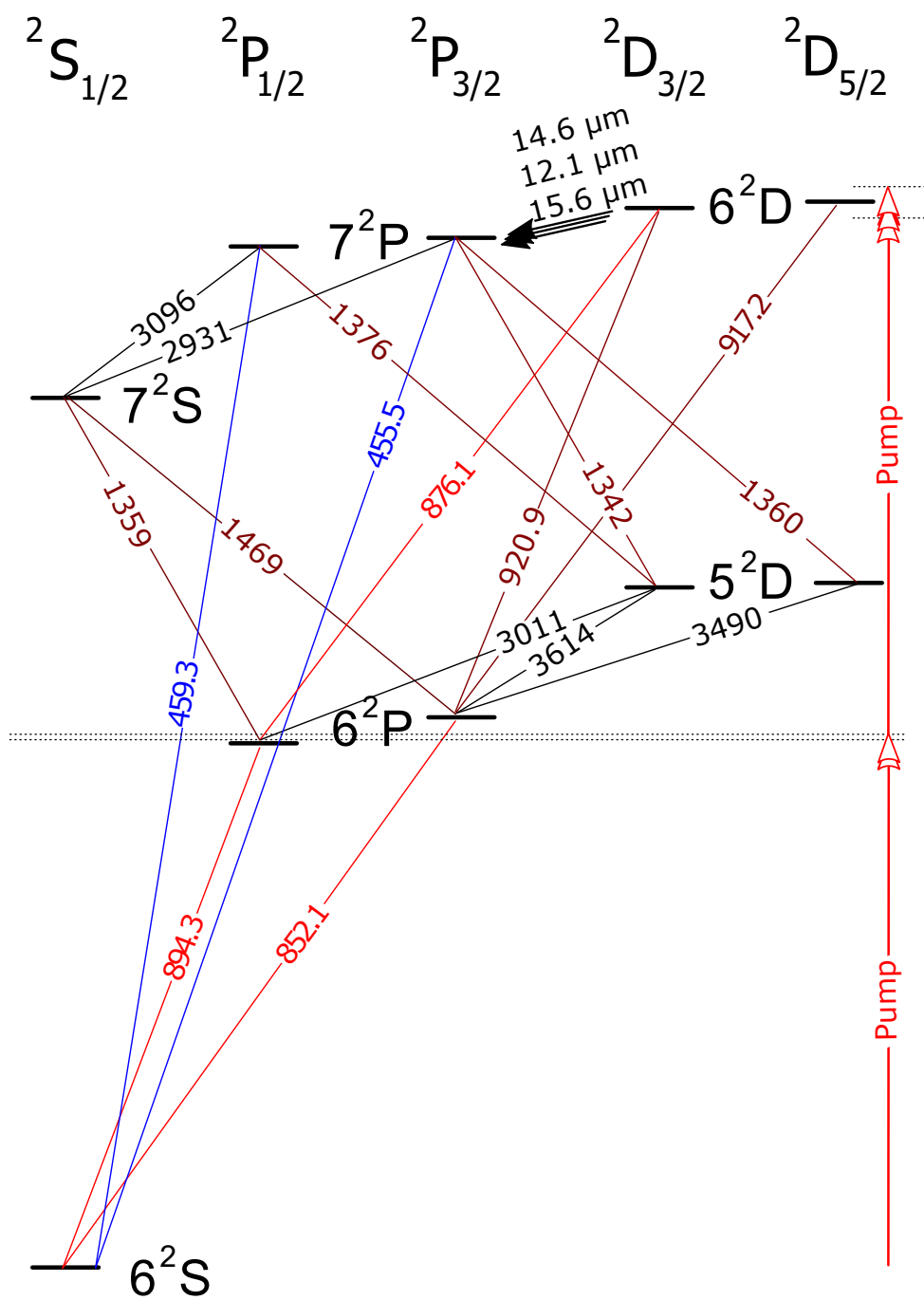


Figure 14. Energy diagram with all observed cascade lasing lines. Lines are in nm except for the 6D to 7P lines. The 6D to 7P transitions are not transmitted through sapphire windows.

To pump the $6^2D_{3/2}$ and $6^2D_{5/2}$ states, the laser is tuned to 885.39 nm and 883.72 nm, with ~ 28 mJ/pulse.

Pulse energy is tuned with a half wave plate and Polarizing Beam Splitter (PBS). This not only gives the capability to tune the pump energy from 1.5 mJ to 28 mJ, but also insures a more consistent spatial and temporal pump pulse.

Wavelength calibration of the pulsed dye laser was done with a High Finesse WS7; however, some drift from this is seen over time. This creates some additional error for our low pump intensity measurements.

The cesium heat pipe consists of a stainless steel tube, with brazed, normal-incident, uncoated sapphire windows. Inside the heat pipe is 3-5 layers of mesh that spans the heat pipe to within 1 cm of the sapphire windows. The heat pipe was filled with 0-10 Torr of He and pressure was monitored with a MKS 10 Torr Baratron attached to a MKS 670 controller. To function properly the mesh must be saturated with liquid cesium, i.e. it must be above the melting point everywhere in the cell. Two cold zones must also be supplied or the vapor will condense on the windows. Seven temperature zones were used: The windows on either end of the cell were kept to 200 °C with Watlow Ez-zone Heaters and heat wraps. Four intermediary zones were created with flowing water, Neslab RTE-111 chillers, and aluminum blocks; chillers kept the blocks at 38 °C (cold zone next to window) and 48 °C (cold zone next to oven). The central oven zone was made with a 15 cm aluminum block and heated with a Watlow Ez-zone heater from 150 °C to 330 °C. This configuration resulted in > 18 months of operation with no maintenance (i.e no window swaps or addition of cesium).

The heat pipe is 29 cm long, and 2.5 cm diameter. The oven length in the center of the heat pipe is 15 cm which roughly corresponds to the gain length. The pump beam travels 3.6 m from the dye laser to the end of the heat pipe. The fiber collimator

to the spectrometers is 40 cm away from the center of the heat pipe, which allows for ample spatial discrimination of the beams.

Three spectrometers were required to resolve the lines from 455 nm to 3600 nm given in Figure 14. The first was an Acton SpectraPro 275, with 5 μm slits, focal length of .275 m, and a Princeton Instruments PI-Max2 ICCD. The input beam was attenuated with two ND filters, OD 1.6 total. This spectrometer was used to analyze wavelengths from 455 nm to 850 nm.

Spectrometer 2 was a McPherson 218, with a focal length of .3 m, a grating of 600 grooves/mm and 1.25 μm blaze. It was attached to a Roper Scientific OMA V 1x1024 InGaAs strip detector with a peak sensitivity of 2.2 μm .

To cover the 2.5 μm to 4 μm window, Jobin Yvon Triax 320, a .3 m spectrometer with a 300 grooves/mm 4000 nm blaze grating. This was attached to an Electro-Optical-Systems IS-020-E-LN7 single element InSb detector. This detector was attached to a LeCroy Wavepro 7300 3 GHz oscilloscope. Measurements were performed with 250 μm slits. In addition the broad band polarizing beam splitting cube 2 depicted in Figure 13 was swapped with a 45 deg dielectric mirror on a fused silica substrate that gave <.05% transmission at pump wavelengths and > 50% transmission from 2.9 μm to 3.7 μm . The spectrometer was free-space coupled with two gold mirrors and matched with a 35 mm CaF_2 Lens.

White light measurements were performed with a 100 W Earling collimated lamp, and spectrometer intensity calibration was done with a calibrated Oriel 63355 lamp. Both were powered by an Oriel 68831 radiometric power supply.

Energy measurements of the pump were performed with an PE50BF-C head attached to a Nova II meter. Each reading was taken with 500 to 1000 pulses to track the average and standard deviation. For our pump this gives good energy measurements for energies from .3 mJ to > 100 mJ. On the low side a Laser Probe RjP-445

head was attached to a Rm-3700 meter. Again 500 to 1000 pulses were averaged. This worked well for pump energies from .6 μJ to 500 μJ . Energy measurements were taken with the following filters: 455 nm 10 nm bandpass, 532 nm long pass, 850 nm 10 nm band pass, 900 nm long pass, 1100 nm long pass, 1325 nm 50 nm band pass, 1500 nm 50 nm band pass, and 2700 nm to 3975 nm broadband pass filter. Transmission of the filters and other key optics in the apparatus were measured with a Cary 5000 Spectrophotometer (200 nm to 3000 nm), and a Bomem MB 155S FT-IR spectrometer (>1000 nm).

4.3 Results and Discussion

The Two-Photon Pump

The key enabler of our experiment is the degenerate two-photon pump $6^2S \rightarrow 6^2D$. Rubidium $5^2S_{1/2} \rightarrow 5^2D_{5/2}$ and cesium $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ have extraordinarily large two-photon cross sections, perhaps the largest in nature. The fundamental physics involved can be understood from [10, 36]. For our purposes—i.e. degenerate two-photon absorption—it is convenient to use the form:

$$\sigma^{(2)}(\nu) \propto \nu \frac{\lambda_1^3 A_1 \lambda_2^3 A_2}{(\Delta\nu)^2} \quad (10)$$

where λ_1 and A_1 refer to the wavelength and A coefficient of the first real transition involved in the two-photon transition (usually the D1 or D2 line), λ_2 and A_2 are the wavelength and A coefficient for the $P \rightarrow D$ transition, and $\Delta\nu$ is the frequency separation of the virtual state from the real state. This enables us to take an accurate experimentally verified value such as the Rb $5^2S_{1/2} \rightarrow 5^2D_{5/2}$ value of 6.78×10^{-21} cm^4/W [36] and use ratio of the Cs to Rb values to apply it to our case given in Table 3. This behavior is evident in our lasing spectra shown in Figure 15, which compares

Table 3. Two-photon pumping cross sections for the 6D states

Transition	$\lambda_1, \lambda_2(\text{nm})$	$A_1 (\cdot 10^7/s)$	$A_2 (\cdot 10^7/s)$	$\Delta\nu (cm^{-1})$	$\sigma^{(2)} \left(\frac{cm^4}{W} \right)$
$6^2S_{1/2} \rightarrow 6^2D_{5/2}$	852, 917	3.28	1.52	416	$0.39 \cdot 10^{-21}$
$6^2S_{1/2} \rightarrow 6^2D_{3/2}$	894, 876	2.86	1.8	116	$3.2 \cdot 10^{-21}$

the two different pump wavelengths at 237.5 °C, 28 mJ/pulse, and 5 Torr He buffer gas. We note that this is a lasing spectra (from ASE), because we are measuring large energies/pulse (too large to be fluorescence), and the emission is contained in a beam (position of fiber collimator is very sensitive to any off-axis deviations).

Impact of Pressure

With the pump cross-sections understood, the next step was to determine an optimal amount of buffer gas . Fill pressures of 0 Torr He, 1 Torr He, 5 Torr and 10 Torr He were used. In Figure 16 we show the 0 Torr vs 5 Torr He case, where we have used the sum of all the lasing energies to simplify the comparison. To estimate the total energy we used several filtered measurements, corrected for known transmission losses, and added the backwards lasing component discussed below. The 1 and 10 Torr cases are not shown, the 1 Torr He data is only slightly above the 0 Torr He data, and the 10 Torr He data is very similar to the 5 Torr He data.

We hypothesize that this small amount of helium creates beneficial mixing in the $6^2S_{1/2}$ hyperfine states, and the upper 6D and 7P fine structure states. White light measurements were performed on the D_1 and D_2 lines, but the high number densities, combined with the large instrument line-shapes, meant that the results were dominated by the absorption feature wings. Despite these complications, we did not identify any trend with pressure; and the data formed error bounds for the number densities as shown in Figure 16.

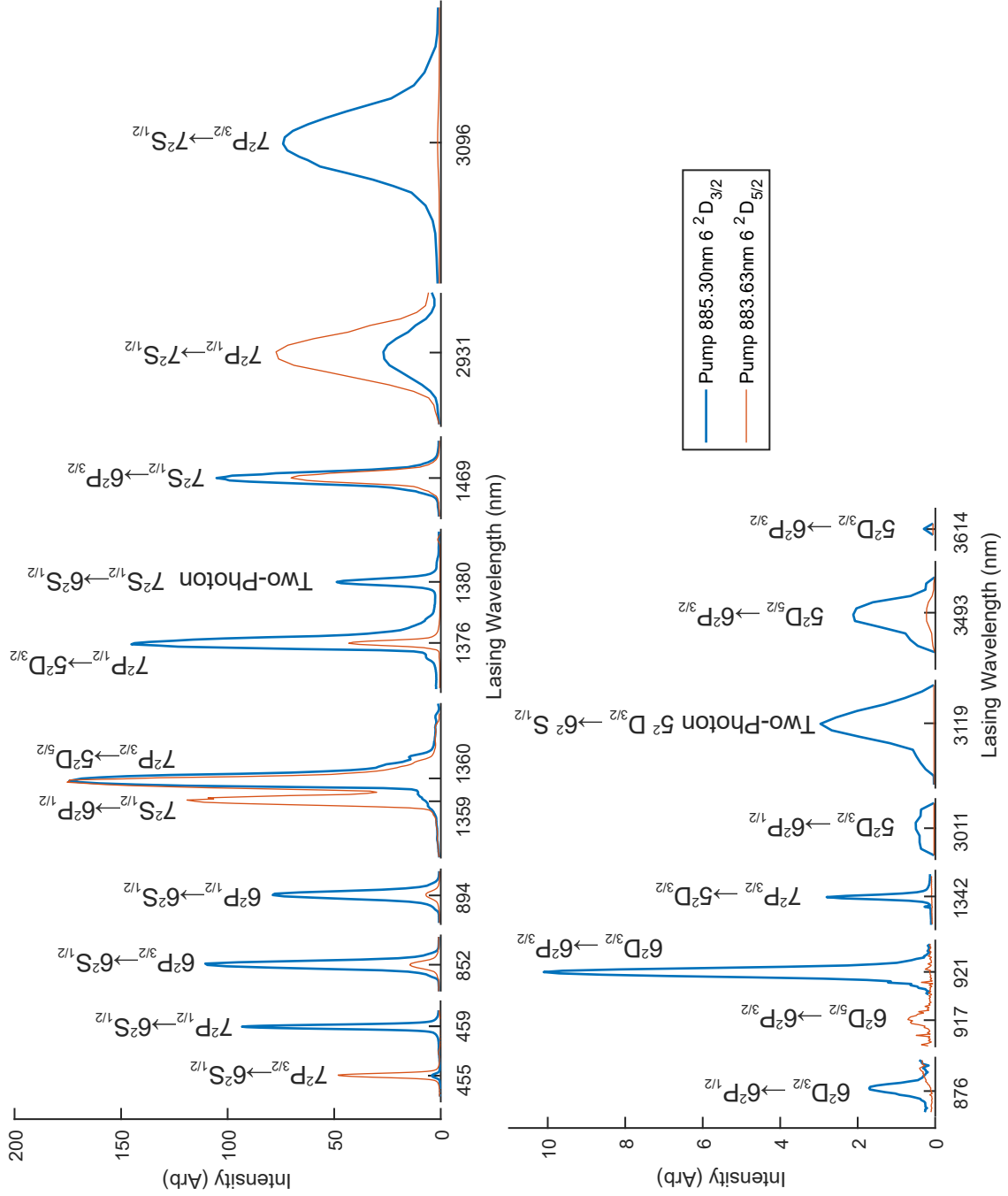


Figure 15. Calibrated lasing spectra from pumping the $6^2D_{3/2}$ (blue) and $6^2D_{5/2}$ (orange) states at a pump energy of 28 mJ, 237.5 °C, 5 Torr He. The wavelength axis is calibrated, but the intensity axis of the 3 μ m spectral window is slightly lower than the intensity of the 455 nm-1469 nm window. The two-photon lines share a virtual state with the pump and are the subject of Chapter V.

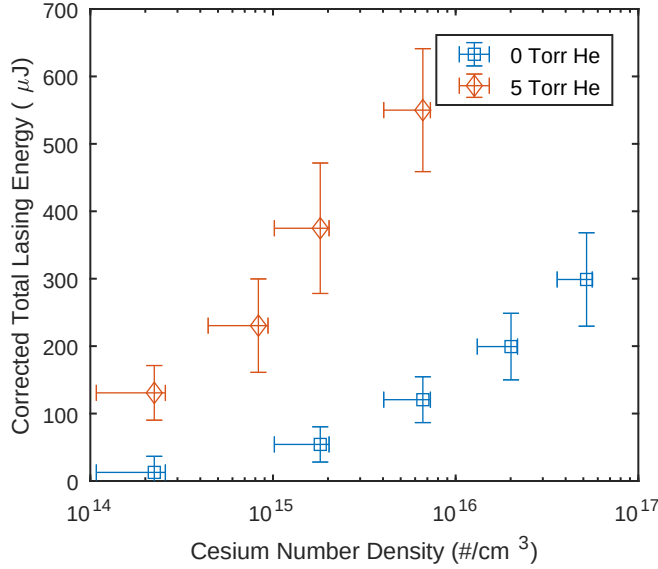


Figure 16. Impact of pressure. The orange $\diamond$ give total lasing energy (from all lines except the pump and $3\mu\text{m}$) at 5 Torr He, the blue $\square$ give total lasing energy at 0 Torr He. Pump: wavelength $6^2D_{3/2}$, energy 14.6 mJ. Error bounds based on the standard deviation of 500 measurements and white light data.

Historically, a small amounts of inert gas have been used in alkali heat pipes for various reasons, but we are unaware of any low pressure comparisons. Many of the above references focus on vacuum conditions such as [97, 34]. However, Hamadani [45] pumped the same transitions in cesium with a heat-pipe of similar dimensions and stated that much of their data was taken with 3 Torr of He or Ar. Clearly small amounts of He buffer gas yields beneficial enhancement to lasing and is consistent with earlier work, but the underlying mechanism is not well understood.

Intensity Scaling

With the motivation of Section 4.3, all further data focuses on 5 Torr of He. To observe how the laser behavior scales with pump intensity, data was taken by tuning a half-wave plate, measuring pump energy, recording spectra, and measuring lasing

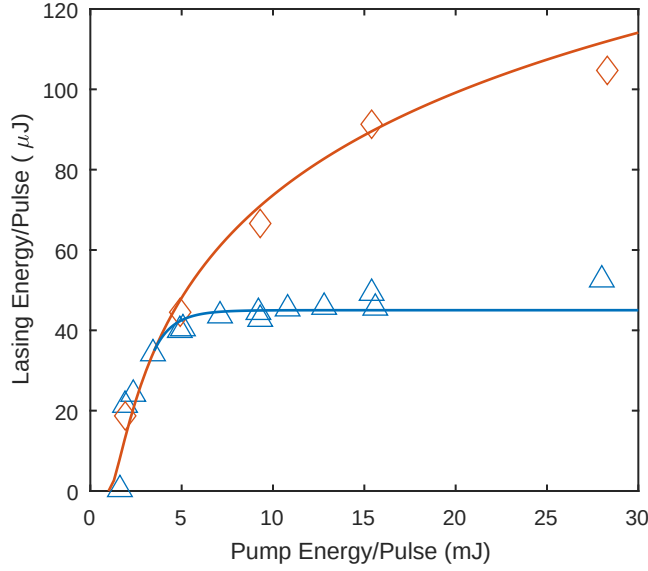


Figure 17. Intensity scaling of 1469 line from the $6^2 D_{3/2}$ state at 5 Torr He, and 237.5°C. Lasing energy from: $\diamond$ (orange) corrected direct measurement; $\triangle$ (blue) calibrated integrated spectra. Solid lines are fits to equation 12.

energy through the filters given above. This process was repeated for several pump intensities. Our initial goal was to fit the laser intensity E_L to

$$E_L = E_m \left(1 - \exp \left[\frac{\eta}{E_m} (E_P - E_t) \right] \right) \quad (11)$$

to obtain the slope efficiency η , lasing threshold E_t and bleached limit E_m as detailed in [64]. However our data is dominated by radial effects at high intensities. An example of this is given in Figure 17, where the filtered energy measurements diverge from the integrated spectral measurements. This is caused by two sources. The first is the fiber collimator (for the spectrometer) captures a 6.3 mm diameter portion of the >9 mm diameter laser beams, and the energy sensor (for the filtered measurements) is a 10 mm x 10 mm square which captures the whole blue beam (the larger wavelengths may diverge more). Second, our pump source effectively grows in size with intensity because more of the beam is above lasing threshold. Therefore we modify our fit to

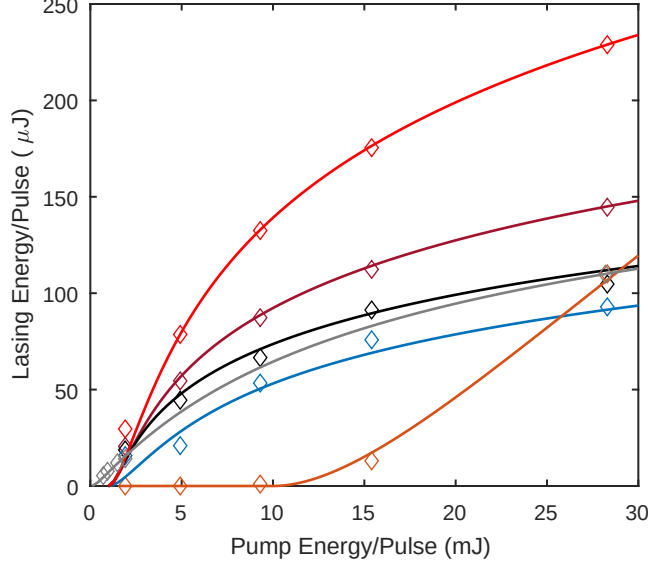


Figure 18. Filtered Energy measurements: Intensity scaling of the 459 nm (blue), 852 nm (orange), 1360 nm (red), 1376 nm (dark red), 1469 nm (black) lines and 3.1 μm (gray). Data from the $6^2D_{3/2}$ pump at 5 Torr He, and 237.5°C.

equation 12, and treat the pump $E_P(r)$ as a Gaussian, and integrate radially:

$$E_L(E_p) = 2\pi\xi \int_0^{E_p(r)=E_t} E_m \left(1 - \exp \left[-\frac{\eta}{E_m} \left(E_{p0} e^{-\frac{r^2}{2c^2}} - E_t \right) \right] \right) r dr \quad (12)$$

where c is a Gaussian standard deviation that is related to the fixed pump beam radius (we use a value of $c=2.175$ mm which corresponds a $1/e^2$ diameter of 8.7 mm), and ξ is an additional parameter that is set to 1.3 for all fits. This ξ factor is included because our pump is not Gaussian, and our initial fits overestimated E_m and η . This factor decreases the fit parameters E_m and η so that η matches with our low power observations where radial effects are less significant. We utilize two different limits of integration for each measurement type: the raw filtered measurements are integrated from 0 to lasing threshold; and the integrated spectral measurements are integrated from 0 to 3.15 mm (the collection radius of the fiber collimator). This

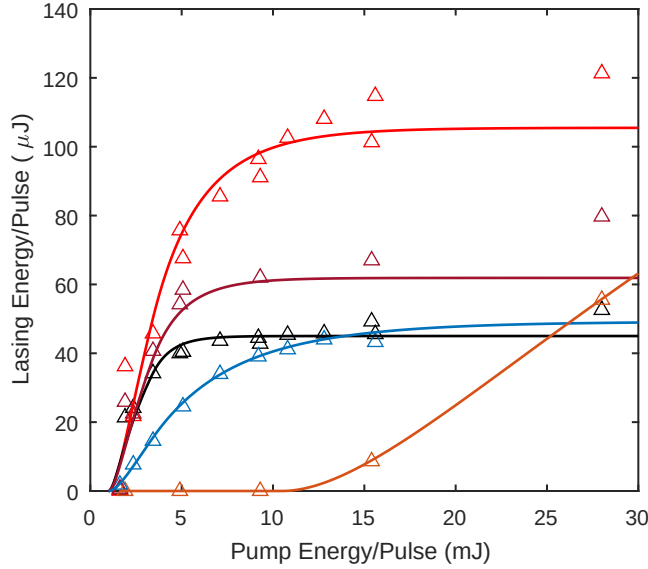


Figure 19. Calibrated integrated spectral measurements: Intensity scaling of the 459 nm (blue), 852 nm (orange), 1360 nm (red), 1376 nm (dark red), and 1469 nm (black) lines data from the $6^2D_{3/2}$ pump at 5 Torr He, and 237.5 °C.

allows us to simultaneously fit equation 12 to both our filtered energy measurements and integrated spectral measurements.

Figure 18 shows the lasing energy measurements fit, and Figure 19 shows the fits for lasing energies calculated from the integrated spectral intensities. The wavelengths shown are the strongest transitions. Overall, equation 12 provides a good improvement over equation 11, by allowing us to consider both measurement types when radial effects are present.

Pumping $6^2D_{3/2}$ vs $6^2D_{5/2}$

Equation 12 is also fit to $6^2D_{5/2}$ data, and plotted in Figure 20. The $6^2D_{5/2}$ results are weaker because of the two-photon cross sections given in Table 3, and shown in the spectra from Figure 15. Specifically, Figure 20 compares the three strongest infrared transitions and the blue transition. The strongest line is essentially the same for both

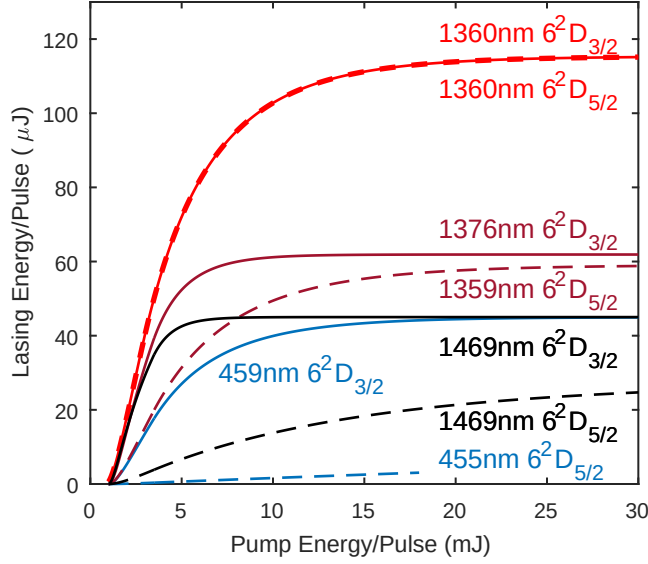


Figure 20. Impact of two-photon pump cross section on cascade lasing. Solid lines denote lasing from pumping to $6^2D_{3/2}$, and the dashed lines are for pumping to $6^2D_{5/2}$.

cases, though we note below that the 1360 nm line from the $6^2D_{5/2}$ pump case is slightly weaker. After this line the degradation becomes increasingly evident.

For the 3 μm window we note from Figure 15 that cascade lasing pumps the 3096 nm line from the $6^2D_{3/2}$ pump, and the $6^2D_{5/2}$ pump favors the 2932 nm line. The weaker lines also insinuate some of the competition that occurs from cascade lasing, for instance the 1342 nm line is only seen lasing from the $6^2D_{3/2}$ pump, even though the parent $7^2P_{3/2}$ state is heavily populated by both pumps. One possible explanation for this, and the existence of a strong 1376 nm line from the $6^2D_{5/2}$ pump case, is fine structure mixing.

Lasing from Spin Orbit Mixing

Fine structure mixing plays an important role in our cesium He mixture. When the single valence electron in cesium is excited to high levels the expectation value $\langle r \rangle$ increases, and the energy separation to near by levels becomes small, which implies

Table 4. Fine structure mixing cross sections for the $6^2D_{3/2} \rightleftharpoons 6^2D_{5/2}$ and $7^2P_{3/2} \rightleftharpoons 7^2P_{1/2}$ transitions

Transition	P	$Q^{(P)} (cm^2)$	T(C)	Ref.
$6^2D_{3/2} \rightarrow 6^2D_{5/2}$	Cs	$270 \cdot 10^{-16}$	210	[98]
$6^2D_{5/2} \rightarrow 6^2D_{3/2}$	Cs	$210 \cdot 10^{-16}$	210	[98]
$6^2D_{3/2} \rightarrow 6^2D_{5/2}$	He	$23 \cdot 10^{-16}$	77	[21]
$6^2D_{5/2} \rightarrow 6^2D_{3/2}$	He	$54 \cdot 10^{-16}$	77	[21]
$7^2P_{1/2} \rightarrow 7^2P_{3/2}$	Cs	$107 \cdot 10^{-16}$	170	[70]
$7^2P_{3/2} \rightarrow 7^2P_{1/2}$	Cs	$121 \cdot 10^{-16}$	170	[70]
$7^2P_{1/2} \rightarrow 7^2P_{3/2}$	He	$12 \cdot 10^{-16}$	177	[27]
$7^2P_{3/2} \rightarrow 7^2P_{1/2}$	He	$11 \cdot 10^{-16}$	177	[27]

large collisional cross sections and transfer rates. This has been well studied and the relevant cross sections are summarized in Table 4.

Spin orbit mixing is very prevalent in our data, the best example is the lasing from 1376 nm and 3096 nm lines from the $6^2D_{5/2}$ pump case as seen in Figure 15. If spin orbit mixing were absent we would not see this line lasing, and we would instead see a strong 1342 nm line. These results confirm that spin-orbit mixing can generate cascade lasing. To better connect this phenomena to the values presented in Table 4, we plot the lasing intensity of the 1376 nm line for 5 Torr He and 0 Torr He for various cesium densities in Figure 21. The vacuum data shows a very weak 1376 nm line until enough cesium density is available to fuel the Cs-Cs collisional mixing. The 5 Torr case shows a stronger more log-linear trend because of Cs-He mixing as well as improvements we give in section 4.3.

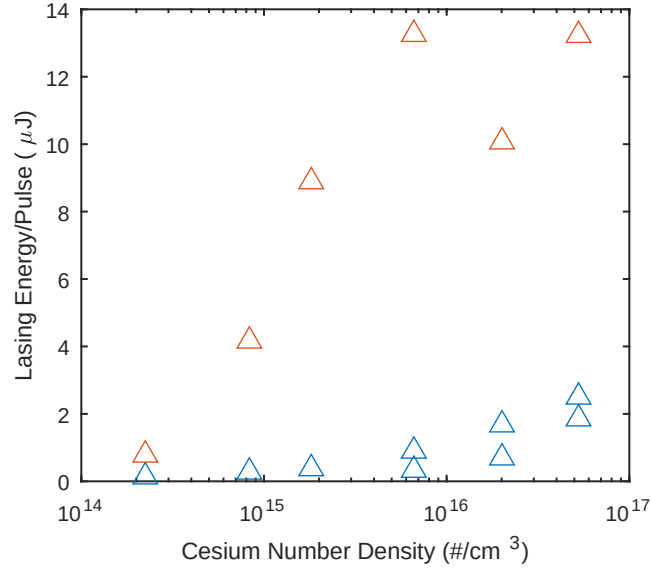


Figure 21. Cascade lasing from fine structure mixing of the $6^2D_{3/2} \rightleftharpoons 6^2D_{5/2}$ and $7^2P_{3/2} \rightleftharpoons 7^2P_{1/2}$ transitions. $\triangle$ (orange) 5 Torr He; $\triangle$ (blue) 0 Torr He. Data from $6^2D_{5/2}$ pump.

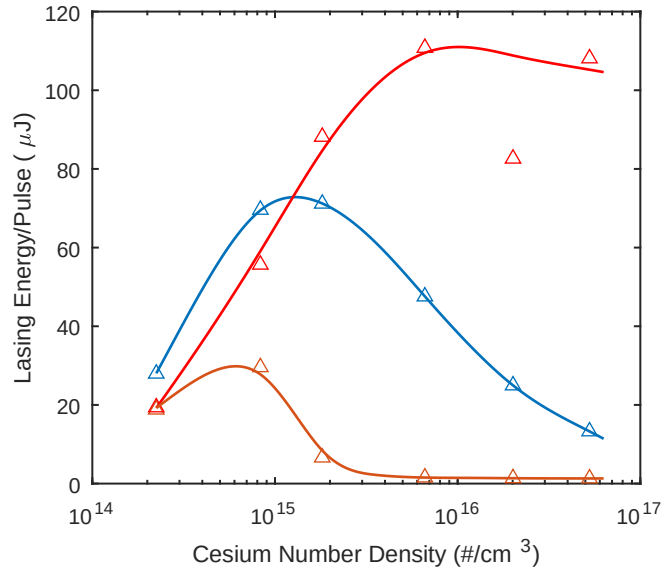


Figure 22. Impact of optical trapping on cascade lasing of the 459 nm (blue $\triangle$), 852 nm (orange $\triangle$), 1360 nm lines (red $\triangle$) when pumping to the $6^2D_{3/2}$ with 14.6 mJ/pulse.

Optical Trapping

Optical trapping plays a key role in cascade lasing transitions connected to the ground state. This is not immediately obvious from the maximum pump energy spectra presented in Figure 15, which shows a strong D_1 and D_2 line. Therefore we include the D_2 line Figures 18 and 19 to show that significant pump energies are required. Impacts of optical trapping are seen in other lines as well. In Figure 22 we plot the 459 nm, 852 nm, and 1360 nm lines at several cesium densities. We draw lines through the data to better illustrate the trends we observed. This figure shows how the D_2 line, with its larger A coefficient, is trapped first. At slightly higher temperatures the 459 nm line also starts to exhibit trapping.

The key result is that, even if an inversion exists, optical trapping can prevent lasing. Transitions further removed from the ground state will be more resilient to optical trapping effects. For instance, the 3096 nm line may peak at slightly higher number densities than the 1360 nm line. One interesting consequence is that optical trapping can serve as control mechanism for particular wavelengths. Therefore cesium density and pump intensity can be used to control the lasing threshold of trapped transitions. In Figures 18 and 19, we estimate lasing thresholds for the D_1 and D_2 line to be an order of magnitude larger than the others given below.

Fit Parameters at Various Cesium Densities

To obtain better trends in lasing threshold, slope efficiency, and bleached limit, we repeat the process of fitting equation 12 at two additional cesium densities for the 1360 nm line, shown in Figure 23, the 1376 nm line, and the 1469 nm line (not shown).

The fit parameters are summarized in Table 5. Values for Table 5 are taken from fitting equation 12 then correcting the slope efficiency and lasing threshold for the

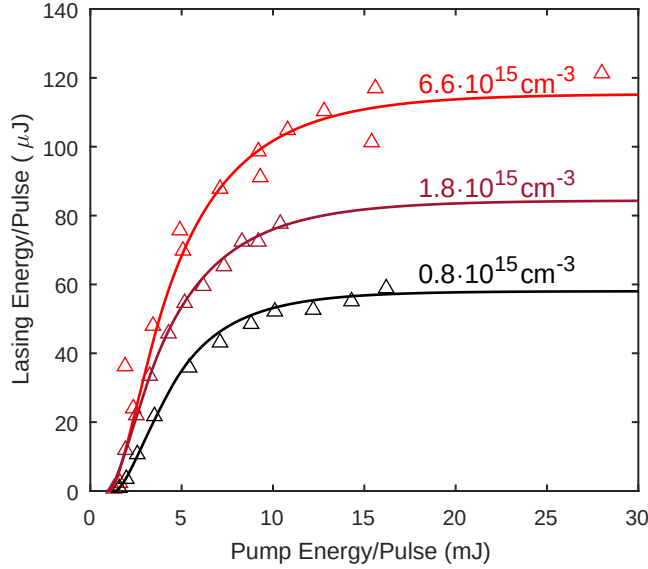


Figure 23. Intensity scaling of the 1360 nm line from the $6^2D_{3/2}$ pump at 5 Torr He, and 237.5 °C or $6.6 \cdot 10^{15} \text{ cm}^{-3}$ (red), 200 °C or $1.8 \cdot 10^{15} \text{ cm}^{-3}$ (dark red), 180 °C or $0.8 \cdot 10^{15} \text{ cm}^{-3}$ (black). Data shown from integrated spectral intensity.

sapphire window. These results show good slope efficiencies and low lasing thresholds. The slope efficiencies and bleaching limit both increase with cesium density, and threshold decreases. Higher efficiencies may be possible with appropriate cavity design.

Discussion and Sources of Error

The results shown in Table 5 would be incomplete without a discussion of the experimental errors. Most of the raw energy measurements are accurate to <10% and are not a considerable source of error; but pulsed dye pumps are known to have 20% pulse to pulse variation. All measurements used 500 averaged pulses, and report standard deviations as shown in Figure 16. We calculate the lasing cross-sections for each transition using A coefficients summarized in Table 6, and give the lasing energies with standard deviations for all transitions in Table 7 below, along

Table 5. Laser slope efficiency η , threshold E_t , and bleached limit E_M for various conditions from pumping the $6^2D_{3/2}$ state

$\lambda(\text{nm}):$	T $^{\circ}\text{C}$	η (%)	E_t (mJ)	E_m (μJ)
1360	237.5	5.3	.72	82.0
1360	200	4.1	.77	60.0
1360	180	3.5	1.11	41.3
1376	237.5	4.7	0.85	70.0
1376	200	4.5	0.85	53.0
1376	180	3.2	1.19	37.0
1469	237.5	7.1	0.85	33.0
1469	200	5.2	0.68	28.5
1469	180	4.8	1.11	24.5
459	237.5	2.1	0.85	32
3096	237.5	1.2	0.063	37

with the cross sections for each transition. A comparison is also made at maximum pump energy between the $6^2D_{3/2}$, $6^2D_{5/2}$, and the $6^2D_{3/2}$ at 4.9 mJ/pulse. The 4.9 mJ/pulse case is also interesting because it is close to maximum efficiency; when we take our raw total energy measurements and correct for transmission losses we see a 10% conversion efficiency, we can compare this to the addition of all the values in the 4.9 mJ/pulse case which comes out to $\sim 7\%$. Therefore we report the total peak conversion efficiency at $10 \pm 3\%$.

In our setup the PBS cubes reflected a moderate amount of blue and probably some infrared. Perhaps enough to create small cavity effects. With the setup briefly reconfigured with different optics, we saw, $\sim 75\%$ backwards lasing energy in the 1342 nm to 1469 nm spectral window, $\sim 21\%$ for the 2932 nm to 3600 nm window, and $\sim 10\%$ for the blue. These energies are incorporated into the total lasing energy for each line in Table 7. Large difference in forward and backwards lasing fields are expected when the source is a high intensity unidirectional pump generating ASE. We also expect larger divergence with longer wavelengths, therefore the decrease in the $3\mu\text{m}$ backwards fields is not unreasonable. However, the backwards blue field is

significantly smaller than the others, small enough that it is difficult to distinguish between a beam and a potential back reflection of the forward beam. We also note that a highly directional blue beam is seen by Hamadani [45], where four wave mixing was believed to be the source of the directionality.

Our 459 nm efficiencies (peak 0.5% off resonance at 200°C) are very similar to those reported by Hamadani (0.8%)[45]. As a side note there are non-linear lines and effects that occur off resonance that greatly enhance many features, which we cover in Chapter V. In general we expect similar fit parameters to Table 5 when pumping the rubidium $5^2S_{1/2} \rightarrow 5^2D_{5/2}$ transition. Sulham [97] reports slightly lower thresholds, consistent with the slightly higher two-photon cross section. However, lower lasing energy's and slope efficiencies are also reported. Potential reasons could be the use too low temperatures and, if not properly addressed, poor thermal zone control of the heat pipe can be a significant source of error. There are also more abstract errors created by a transitional period in our labs where the lab temperatures were widely fluctuating. This gives us two additional sources of error: First the fiber collimators that were precisely aligned to the lasing beams could shift off creating an additional $\pm 20\%$ difference. Second, this also caused slight shifts in the operation wavelength of our pulsed dye laser. This creates an interesting dilemma: at high pump intensities we see a large degree of saturation broadening which means tuning to the peak wavelength without the assistance of a wave meter is easy but central wavelength is not known accurately. Therefore, when analyzing the low intensity behavior, we see potentially higher thresholds, and lower slope efficiencies because we are not necessarily on resonance. We also have some error introduced by fitting equation 12. The pump radial intensity profile is not well known, and the spot is far from Gaussian. Also if we consider the limit where no radial effects are present—which implies that the spectral measurements in Figures 19 and 20 are accurate representations of slope

Table 6. Einstein A coefficients, and one-photon cross sections for the cascade lasing transitions at 237.5 °C

$\lambda(\text{nm}):$ Transition	A (s^{-1})	Ref.	$\sigma (cm^2)$
455.7: $7^2P_{3/2} \rightarrow 6^2S_{1/2}$	$1.84 \cdot 10^6$	[100]	$1.55 \cdot 10^{-13}$
459.4: $7^2P_{1/2} \rightarrow 6^2S_{1/2}$	$7.93 \cdot 10^5$	[100]	$6.84 \cdot 10^{-14}$
852.3: $6^2P_{3/2} \rightarrow 6^2S_{1/2}$	$3.28 \cdot 10^7$	[79]	$1.81 \cdot 10^{-11}$
876.4: $6^2D_{3/2} \rightarrow 6^2P_{1/2}$	$1.8 \cdot 10^7$	[71]	$1.08 \cdot 10^{-11}$
894.6: $6^2P_{1/2} \rightarrow 6^2S_{1/2}$	$2.86 \cdot 10^7$	[79]	$1.82 \cdot 10^{-11}$
917.5: $6^2D_{5/2} \rightarrow 6^2P_{3/2}$	$1.52 \cdot 10^7$	[48]	$1.04 \cdot 10^{-11}$
921.1: $6^2D_{3/2} \rightarrow 6^2P_{3/2}$	$2.66 \cdot 10^6$	[48]	$1.85 \cdot 10^{-12}$
1342.8: $7^2P_{3/2} \rightarrow 5^2D_{3/2}$	$1.3 \cdot 10^5$	[48]	$2.08 \cdot 10^{-13}$
1359.2: $7^2S_{1/2} \rightarrow 6^2P_{1/2}$	$6.23 \cdot 10^6$	[48]	$1.39 \cdot 10^{-11}$
1360.6: $7^2P_{3/2} \rightarrow 5^2D_{5/2}$	$1.10 \cdot 10^6$	[48]	$2.46 \cdot 10^{-12}$
1376.3: $7^2P_{1/2} \rightarrow 5^2D_{3/2}$	$1.59 \cdot 10^6$	[48]	$3.86 \cdot 10^{-12}$
1469.9: $7^2S_{1/2} \rightarrow 6^2P_{3/2}$	$1.14 \cdot 10^7$	[48]	$3.22 \cdot 10^{-11}$
2931.3: $7^2P_{3/2} \rightarrow 7^2S_{1/2}$	$4.05 \cdot 10^6$	[48]	$9.07 \cdot 10^{-11}$
3011.2: $5^2D_{3/2} \rightarrow 6^2P_{1/2}$	$9.13 \cdot 10^5$	[81]	$2.22 \cdot 10^{-11}$
3096.1: $7^2P_{1/2} \rightarrow 7^2S_{1/2}$	$3.52 \cdot 10^6$	[48]	$9.29 \cdot 10^{-11}$
3491.0: $5^2D_{5/2} \rightarrow 6^2P_{3/2}$	$7.81 \cdot 10^5$	[29]	$2.95 \cdot 10^{-11}$
3614.1: $5^2D_{3/2} \rightarrow 6^2P_{3/2}$	$1.07 \cdot 10^5$	[81]	$4.49 \cdot 10^{-12}$
12143: $6^2D_{3/2} \rightarrow 7^2P_{1/2}$	$9.0 \cdot 10^4$	[48]	$1.43 \cdot 10^{-10}$
14592: $6^2D_{5/2} \rightarrow 7^2P_{3/2}$	$6.3 \cdot 10^4$	[48]	$1.74 \cdot 10^{-10}$
15566: $6^2D_{3/2} \rightarrow 7^2P_{3/2}$	$8.6 \cdot 10^3$	[48]	$2.88 \cdot 10^{-11}$

efficiency and bleached limit—then we see a 30 to 50% reduction in slope efficiency, and a $\sim 35\%$ increase in bleached limit. Although this case is unlikely, we collect this 30% error with the above sources and estimate $\sim 50\%$ error to our fit parameters.

The pump beam size is estimated with burn paper and the blue beam can be seen visually, but IR beams are expected to have a larger divergence. No temporal measurements of the pump or lasing lines are presented. Our energy to intensity conversions assume 10 ns pulse widths but they could be as low as 6-8 ns, implying slightly higher thresholds; however, the half wave plate and PBS insured that the spatial and temporal properties of the beam were consistent while tuning intensity.

Table 7. Cross sections for the lasing transitions with measured energies (in μJ)

$\lambda(\text{nm}):$ Transition	$\sigma (cm^2)$	Pump 4.9 mJ	Pump 28 mJ	
		$6^2D_{3/2}$	$6^2D_{3/2}$	$6^2D_{5/2}$
455.7: $7^2P_{3/2} \rightarrow 6^2S_{1/2}$	$1.55 \cdot 10^{-13}$	0.9 ± 0.2	4.0 ± 0.8	47.8 ± 11
459.4: $7^2P_{1/2} \rightarrow 6^2S_{1/2}$	$6.84 \cdot 10^{-14}$	21 ± 4.0	93 ± 19	0.0
			$127 \pm 24^{*\dagger}$	
852.3: $6^2P_{3/2} \rightarrow 6^2S_{1/2}$	$1.81 \cdot 10^{-11}$	0.0	110 ± 50	14.0 ± 6.3
			$219 \pm 58^\dagger$	
876.4: $6^2D_{3/2} \rightarrow 6^2P_{1/2}$	$1.08 \cdot 10^{-11}$	6.3 ± 0.8	1.8 ± 0.8	0.0
894.6: $6^2P_{1/2} \rightarrow 6^2S_{1/2}$	$1.82 \cdot 10^{-11}$	0.0	79 ± 35	6.7 ± 3.0
917.5: $6^2D_{5/2} \rightarrow 6^2P_{3/2}$	$1.04 \cdot 10^{-11}$	0.0	0.0	0.9 ± 0.4
921.1: $6^2D_{3/2} \rightarrow 6^2P_{3/2}$	$1.85 \cdot 10^{-12}$	32 ± 15	10 ± 4.6	0.0
1342.8: $7^2P_{3/2} \rightarrow 5^2D_{3/2}$	$2.08 \cdot 10^{-13}$	0.6 ± 0.1	2.8 ± 0.3	0.0
1359.2: $7^2S_{1/2} \rightarrow 6^2P_{1/2}$	$1.39 \cdot 10^{-11}$	< 3	< 6	63 ± 8
1360.6: $7^2P_{3/2} \rightarrow 5^2D_{5/2}$	$2.46 \cdot 10^{-12}$	79 ± 11	229 ± 28	180 ± 23
1376.3: $7^2P_{1/2} \rightarrow 5^2D_{3/2}$	$3.86 \cdot 10^{-12}$	55 ± 7.0	145 ± 17	18 ± 2.2
1469.9: $7^2S_{1/2} \rightarrow 6^2P_{3/2}$	$3.22 \cdot 10^{-11}$	45 ± 6.0	105 ± 13	44 ± 5.5
2931.3: $7^2P_{3/2} \rightarrow 7^2S_{1/2}$	$9.07 \cdot 10^{-11}$	9 ± 5	33.1 ± 6.3	96.5 ± 19
3011.2: $5^2D_{3/2} \rightarrow 6^2P_{1/2}$	$2.22 \cdot 10^{-11}$	0.0	0.25 ± 0.05	0.0
3096.1: $7^2P_{1/2} \rightarrow 7^2S_{1/2}$	$9.29 \cdot 10^{-11}$	28 ± 10	82.4 ± 16	$0.9 \pm .17$
3491.0: $5^2D_{5/2} \rightarrow 6^2P_{3/2}$	$2.95 \cdot 10^{-11}$	0.0	$1.0 \pm .2$	$0.1 \pm .02$
3614.1: $5^2D_{3/2} \rightarrow 6^2P_{3/2}$	$4.49 \cdot 10^{-12}$	0.0	$0.03 \pm .005$	0.0
12143: $6^2D_{3/2} \rightarrow 7^2P_{1/2}$	$1.43 \cdot 10^{-10}$	Transmission blocked by Al_2O_3		
14592: $6^2D_{5/2} \rightarrow 7^2P_{3/2}$	$1.74 \cdot 10^{-10}$	Transmission blocked by Al_2O_3		
15566: $6^2D_{3/2} \rightarrow 7^2P_{3/2}$	$2.88 \cdot 10^{-11}$	Transmission blocked by Al_2O_3		

*Off Resonance, † Data taken at 200°C

Simplified Model

It is difficult to predict impact of an intense pump onto the 6D states and its 20 coupled transitions. However, we can gain much insight by looking at the two-photon saturation intensity and its relationship to particular thresholds. We first give the pump rate equation in the absence of all lasing:

$$\frac{dn_{6D_{3/2}}}{dt} = \frac{\sigma^{(2)} I^2}{h\nu} \left(\frac{g_{6D_{3/2}}}{g_{6S_{1/2}}} n_{6S_{1/2}} - n_{6D_{3/2}} \right) - \Gamma n_{6D_{3/2}} \quad (13)$$

where n_i is the population in level i , g_i is the degeneracy of level i , and $\Gamma = \sum_i A_{6D_{3/2} \rightarrow i}$. Next we define a pseudo saturation intensity $I_S^* = \sqrt{h\nu\Gamma/\sigma^{(2)}}$, the saturation parameter $S = I/I_S^*$, and the inversion $\Delta n = (g_{3/2}/g_{1/2}) n_{6S} - n_{6D_{3/2}}$. Because the A coefficients are large, we can assume steady state conditions are reached within the pump pulse and rewrite equation 13 as

$$\Delta n = \frac{\frac{g_{6D_{3/2}}}{g_{6S_{1/2}}} n_{6S_{1/2}}}{1 + S^2} \quad (14)$$

from which we define the saturation intensity as the intensity where $\Delta n = n_{6S}/2$ and obtain:

$$I_s = I_S^* \sqrt{2 \frac{g_{6D_{3/2}}}{g_{6S_{1/2}}} - 1} = 66 \frac{\text{kW}}{\text{cm}^2}. \quad (15)$$

To compare this with our 5 Torr case we consider the limit where rapid mixing 6D states and lasing to the $7^2P_{1/2}$ state effectively gives one upper level with a degeneracy equal to the sum of the three states (4+6+2):

$$I_s = I_S^* \sqrt{2 \frac{g_{6D_{3/2}} + g_{6D_{5/2}} + g_{7P_{1/2}}}{g_{6S_{1/2}}} - 1} = 126 \frac{\text{kW}}{\text{cm}^2} \quad (16)$$

and compares well with our .85 mJ/pulse or 110 to 143 kW/cm² threshold value. The range comes from the area uncertainty of the lasing spot size .6 cm² (the pump area) to .8 cm² (estimated from the size of the blue beam). This suggests a potential method to estimating thresholds of lines that are a few cascaded transitions away from the pump state. A possible interpretation is that this is the intensity at which a sufficient inversion will exist on the Cs 3096 nm $7^2P_{1/2} \rightarrow 7^2S_{1/2}$ transition causing it to be strongly lasing, and because cascade lasing is the dominant mechanism, once the 3096 nm line is lasing strongly the others will start lasing; i.e. a threshold. Using this method with the equivalent transition in Rubidium, $5^2S_{1/2} \rightarrow 5^2D_{5/2}$,

the two photon cross section from Gallagher [36], pulsed duration 5 ns, and the .3 mJ threshold from Sulham [97], with our spot diameter, we calculate $\sim 75 \text{ kW/cm}^2$ and using our saturation intensity method of equation 16, and obtain a value of 70 kW/cm^2 ; again giving a good estimate to an otherwise intricate problem. Its also important to note that Sulham estimates the threshold intensity to be $\sim 200 \text{ kW/cm}^2$, but this is because they used the spot size coming out of the pump laser, ignoring the divergence. Transitions connected directly to the 6D states are expected to have near zero thresholds. The next lines to lase, e.g. the 3096 nm line, are expected to have much lower thresholds then the saturation intensity, but higher than the 12 μm line. Indeed we see this in our data in Table 5, where the threshold more than an-order of magnitude less.

To gain further insight into the above trends and the 3096 nm threshold, we add to equation 14 the criteria that $6^2D_{3/2} \rightarrow 7^2P_{1/2}$ always reaches the bleached limit, and again consider the steady state behavior:

$$n_{7P_{1/2}} = \frac{1}{2}n_{6D_{3/2}} \quad (17)$$

$$\frac{dn_{7S_{1/2}}}{dt} = A_{7P_{1/2} \rightarrow 7S_{1/2}}n_{7P_{1/2}} - \Gamma_{7S_{1/2}}n_{7S_{1/2}} = 0 \quad (18)$$

$$n_T = n_{6S_{1/2}} + n_{6D_{3/2}} + n_{7P_{1/2}} + n_{7S_{1/2}} = f_B n_{CsT} \quad (19)$$

where n_{CsT} is the number density from vapor pressure of cesium at temperature T and f_B is the Boltzmann distribution of the ground hyperfine states, and $\Gamma_{7S_{1/2}} = \sum_i A_{7S_{1/2} \rightarrow i}$. Again, we define an inversion

$$\Delta n_L = n_{7P_{1/2}} - n_{7S_{1/2}} \quad (20)$$

and using equations 14, 17, 18, and 19, we obtain

$$\Delta n_L = f_B \cdot n_{CsT} \cdot \frac{S^2 \left(\Gamma_{7S_{1/2}} - A_{7P_{1/2} \rightarrow 7S_{1/2}} \right)}{\Gamma_{7S_{1/2}} + S^2 \left(4\Gamma_{7S_{1/2}} + A_{7P_{1/2} \rightarrow 7S_{1/2}} \right)}. \quad (21)$$

We define the threshold gain in the usual manner:

$$\gamma_{th} = \sigma_L \Delta n_L L_g \approx 20 \quad (22)$$

where L_g is the gain length ~ 10 cm.

$$I_t \approx I_S^* \sqrt{\frac{\gamma_{th} \Gamma_{7S_{1/2}}}{\Gamma_{7S_{1/2}} - A_{7P_{1/2} \rightarrow 7S_{1/2}}}} \frac{1}{\sqrt{f_b L_g \sigma_L n_{Cs}}} \propto \frac{1}{\sqrt{n_{Cs}}} \quad (23)$$

We find the threshold intensity to be ~ 100 W/cm² which is significantly less than our fitted measurement of ~ 11 kW/cm². If we were to include fine structure mixing within the 7P and 6D states, and a finite lasing intensity of the $6D_{3/2} \rightarrow 7P_{1/2}$ transition, the threshold would increase; but a more involved model is required. We can still draw two important conclusions: First, as shown in equation 23, $I_t \propto 1/\sqrt{n_{Cs}}$, this explains the decreasing thresholds with cesium density in Table 5, which also had remained a mystery in Sulham's work [97]. Second, the threshold is going to be very low and favorable for pumping with modern diodes.

4.4 Chapter Summary

Considerable evidence exists to support the feasibility of modern laser diodes to two-photon pump the Cesium 6^2D transitions and create a DPAL analog with high wavelength agility. We observe cascade lasing on 17 transitions, that have a broad range of supporting missions. Interesting non-linear effects exist, but the dominant mechanism is cascade lasing. Small mounts of inert buffer gas are believed to cause

beneficial mixing and enhancement to the overall system. Even without the aid of a cavity we observe low thresholds, high slope efficiencies, large bleaching limits, and scalability with cesium density. Many lines are intense enough to be viable pulsed dye sources in the difficult 1300 to 1500 nm range. It is also possible to manipulate the lasing with spin orbit mixing, and alter lasing thresholds of particular transitions by exploiting optical trapping. Simple methods have also been presented to understand lasing threshold behavior.

V. Two-Photon and Nonlinear Stimulated Emission from Two-Photon Excitation of Cesium 6^2D

In this chapter we report our non-linear on and off-resonance observations from two-photon pumping the entire cesium 6^2D states. We report the observation and theory of stimulated two-photon lines that arise from two-photon pumping the cesium $6D$ states. Large two-photon absorption and strong cascade lasing pump multi-photon processes. The first arises from the $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ transition in which a 1381 nm line is observed. A second two-photon line is observed around 3120 nm, from the $5^2D_{1/2} \rightarrow 6^2S_{1/2}$ transition. Theoretical predictions for degenerate and non-degenerate two-photon cross sections are given. Both lines show strong off-resonance enhancement and the 1381 nm line is shown to have >9 nm of tuning, with a slope efficiency of $\sim 1\%$, and lasing threshold of <140 kW/cm². The mechanisms of four wave mixing, six wave mixing, and two-photon lasing are discussed as potential mechanisms; and future two-photon laser experiments are suggested.

5.1 Background

Alkali vapors have a rich history in the field of lasers and nonlinear optics. Recent kW-class demonstrations in Diode pumped alkali lasers [75, 6] have reinvigorated interest in the utility of alkali lasers.

Potassium was first suggested for a laser gain medium in 1958 [83], and experimental observations in cesium were seen in 1962 [78]. The first nonlinear lasing behavior observed was Stimulated Electronic Raman Scattering (SERS) in potassium in 1967[92]. After this initial work, many examples were seen in potassium [107, 22, 5], rubidium [66], and cesium [23, 49, 106]. These methods produce tunable infrared emission in the 2-8 μm range from a one-photon pump.

Two-photon pumping has been used to demonstrate Stimulated Hyper-Raman

Scattering (SHRS) in sodium [24], potassium [14], and cesium [102]. These methods also tend to produce tunable infrared wavelengths. However, when the combination of pumping to higher levels produces a strong stokes (anti-stokes) field that in-turn produces an anti-stokes (stokes) [12, 14, 24] field, the processes become difficult to distinguish from four wave mixing (FWM).

FWM was reported in 1967 in potassium [80, 61] and in 1973 for cesium [90]. The primary area of interest for FWM has been the generation of blue and UV wavelengths. The use of two sequential (e.g. $S \rightarrow P \rightarrow D$) pumps has generated FWM at 455 nm in cesium [85] and 420 nm in rubidium [86, 101, 103, 1] with blue conversion efficiencies up to 2.6% [101]. Two-photon emission in sodium at 1.18 μm (tunable) was observed via FWM by two pumps: one in the UV $3S \rightarrow 3P$ and the second on a 600 nm virtual state for $4S \rightarrow 3S$ [46]. FWM from two-photon pumping has been reported in rubidium [86] and cesium [45, 35]. The two-photon pump also creates numerous cascade lasing wavelengths from Amplified Spontaneous Emission (ASE) [97]; the presence of these additional fields can also create Six Wave Mixing (SWM).

In many of the FWM cases, SWM also exists. This coexistence was noted first 1973 for cesium [90]. Two-photon pumped SWM was reported in rubidium [52], where two pulsed dye lasers were used to non-degenerately pump the 5D and 7S states. However, we note that their results were, in many cases, bi-directional and therefore largely due to cascade lasing as we discuss further below. A similar experiment [15] was performed in rubidium for the degenerate pump case in 1995, where 24 axial and non-axial emissions coupled with the pump were observed from 1.3 to 1.6 μm , providing clearer support of SWM processes. Other examples of two-photon pumped, ASE assisted, SWM include: 32 UV emission lines in potassium [110], tunable 722 nm [76], 830 nm and 1.16 μm lines in sodium [60], tunable 1.2 to 1.45 μm lines in

potassium [50], and 680 to 800 nm in mixed sodium-potassium vapors [16].

Strong two-photon pumps also have the potential to create other novel stimulated processes such as two-photon lasing. This was originally proposed by Sorokin in 1964 [91]. The first evidence of two-photon lasing was shown in 1992 through the dressed states of barium atoms [39]. Performance improved in 1997 with the two-photon Raman laser [19]. In this approach, three virtual levels are used to simultaneously absorb two degenerate pump photons and emit two degenerate lasing photons; the process moves population from the lower hyperfine ground state to the upper, and the population mismatch is maintained by an additional pump source. Despite being a high order optical process, 30% CW amplification of a probe was seen in potassium [19], and by monitoring other SRS/mixing process and minimizing the mixing process, they were able to confirm that the results were not multi wave mixing. A two photon laser could also be made by more traditional means. One such device could use a virtual level and a population inversion over a two-photon transition; but to the best of our knowledge this has not been achieved due to lack of proof of a population inversion. For example, in 1981, Nikolaus claimed to demonstrate a two-photon laser in lithium vapor that was pumped and seeded by three pulsed dye lasers [67]. Although interesting multi-photon behavior and gain was clearly present, Jackson [51] emphasized that several criteria were not explicitly proven. In 1995 another example [94] using two pulsed dye lasers to pump lithium vapor supported the conclusion that two-photon gain was present on several transitions, but—as we show below with results in cesium—the mechanism is not clear.

Complications arise in distinguishing ASE generated cascade lasing from multi wave mixing; this also applies to two-photon effects as well. In this work we cover multiple examples involving both one and two photon stimulated emission; and their behavior are in some cases consistent with non-axial FWM and SWM, and in some

cases inconsistent. In particular, we detail our observations of two-photon lines, and show that in the forward direction their behavior mimics the intensity scaling of the cascade lasing processes, while in the backward direction a 20% weaker field is still present.

5.2 Theory

We first cover relevant theory of two-photon absorption, two-photon stimulated emission, FWM, and SWM. For our particular case we are degenerately two-photon pumping cesium vapor. The two-photon pump is tuned between the $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ (where $\omega_P + \omega_P = \omega_{6D_{3/2}}$) and $6^2S_{1/2} \rightarrow 6^2D_{5/2}$ (where $\omega_P + \omega_P = \omega_{6D_{5/2}}$) transitions. These pump processes and key lasing emissions are shown in the energy level diagram in Figure 24. This figure is similar to Figure 14, but we drop the weaker cascade lasing processes that are not directly related to the nonlinear behavior of this chapter. The pump utilizes a virtual state, and is enhanced by the near resonance with the 6P states, which lead to exceptionally strong two-photon cross-sections, that can be bleached. The additional photon also alters our dipole selection rules from $\Delta L = 1 \rightarrow \Delta L = 0, 2$.

We also note two nonlinear two-photon emission lines described by $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ where $\omega_{1380}^{(2)} + \omega_P = \omega_{7S_{1/2}}$, and $5^2D_{3/2} \rightarrow 6^2S_{1/2}$ where $\omega_{3120}^{(2)} + \omega_P = \omega_{5D_{3/2}}$. Both cases utilize the virtual energy level of the degenerate pump. Throughout this work we refer to $\omega_{1380}^{(2)}$ and $\omega_{3120}^{(2)}$ as two-photon lines; however, the generation process could be two-photon lasing or SWM.

Degenerate Two-Photon Absorption

The most common type of two-photon absorption involves two pump photons of equal (degenerate) frequency. We derive the two-photon cross section using second

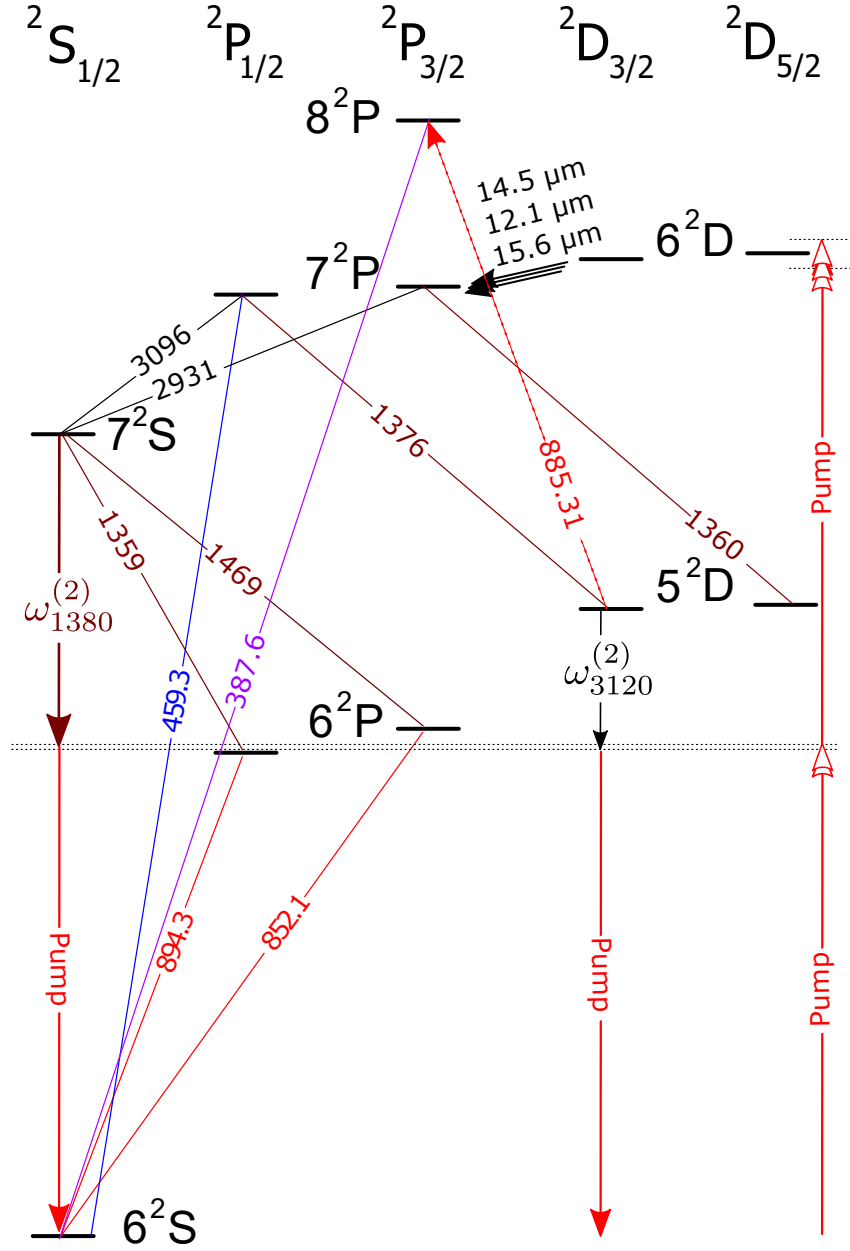


Figure 24. Energy diagram with key cascade lasing lines and nonlinear two-photon lines. The pump frequency ω_P is tuned between the degenerate two-photon pump processes described by $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ where $\omega_P + \omega_P = \omega_{6D_{3/2}}$ and $6^2S_{1/2} \rightarrow 6^2D_{5/2}$ where $\omega_P + \omega_P = \omega_{6D_{5/2}}$. We also note two nonlinear two-photon emission processes described by $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ where $\omega_{1380}^{(2)} + \omega_P = \omega_{7S_{1/2}}$, and $5^2D_{3/2} \rightarrow 6^2S_{1/2}$ where $\omega_{3120}^{(2)} + \omega_P = \omega_{5D_{3/2}}$.

order time-dependent perturbation theory. [10, 25] The electric field is defined as $\mathbf{E} = [Ee^{-i\omega t} + E^\dagger e^{i\omega t}] \hat{e}$, which has a time averaged intensity given by $I = 2nc\epsilon_0 E^2$. We define the upward absorption rate, R , from initial state i to final state f , as

$$\langle R_{i \rightarrow f}^{(2)} \rangle = \sigma_{if}^{(2)} \frac{I^2}{h\nu} \quad (24)$$

where $\sigma_{if}^{(2)}$ is the degenerate two-photon cross section. We assume a single pump polarization, and obtain

$$\begin{aligned} \sigma_{if}^{(2)}(\nu) &= \frac{9}{5} \frac{\nu}{256\pi^4 h n^2 c^2} \sum_n \frac{g_f}{g_i} \frac{\lambda_{fn}^3 A_{fn} \lambda_{ni}^3 A_{ni}}{(\nu_{ni} - \nu)^2} \\ &\quad \times \rho_f(\nu_{fi} = 2\nu), \\ &= \frac{\pi^2 \nu}{5h^3 n^2 c^2 \epsilon_0^2} \sum_n \frac{1}{g_i g_n} \frac{\mu_{fn}^2 \mu_{ni}^2}{(\nu_{ni} - \nu)^2} \\ &\quad \times \rho_f(\nu_{fi} = 2\nu). \end{aligned} \quad (25)$$

where ν is the two-photon frequency, g_i is the degeneracy of state i , λ_{ni} is the wavelength of the $n \rightarrow i$ transition, A_{ni} is the Einstein A coefficient for the same transition, ρ_f is the density of final states—for our case this is a Doppler broadened Gaussian for a frequency of ν —and μ_{ni} is the electric dipole matrix element given by

$$\mu_{ni}^2 = g_n \frac{3\epsilon_0 h \lambda_{ni}^3}{16\pi^3} A_{ni}. \quad (26)$$

It is also important to note the factor of 5 that appears in the denominator of equation 25. When computing this rate, we must average over the dipole moments. For the case of equal pump orientation and polarization: $\langle \mu_{in}^2(\hat{r} \cdot \hat{z}) \mu_{nf}^2(\hat{r} \cdot \hat{z}) \rangle = \mu_{in}^2 \mu_{nf}^2 \langle \cos^4(\theta) \rangle = \mu_{in}^2 \mu_{nf}^2 / 5$. This is analogous to the factor of 3 for one-photon transitions. By comparison, two-sequential pump transitions would have a factor of 1/9.

Although this could be used to distinguish between a two-photon process and two serial one-photon processes, the A coefficients of the upper transitions are not known well enough in cesium.

Theoretical predictions of the two-photon cross sections are given in Table 8 evaluated at line center. The first, and largest, connects the $6^2S_{1/2}$ to the $6^2D_{3/2}$ ($22588.8210 \text{ cm}^{-1}$) [32] via the 894 nm D_1 line ($11178.268 \text{ cm}^{-1}$) [41] intermediate level; the degenerate wavelength is 885.4 nm, and creates a virtual level at $(\nu_{ni} - \nu) = \Delta\nu$. The upper transition to $6^2D_{5/2}$ ($22631.6863 \text{ cm}^{-1}$) [32] utilizes the 852 nm D_2 line ($11732.3071 \text{ cm}^{-1}$) [40]. The degenerate wavelength is 883.7 nm. For comparison with experimental values, this method estimates the Rb $5^2S_{1/2} \rightarrow 5^2D_{5/2}$ as $7.7 \cdot 10^{-21} \text{ cm}^4/\text{W}$, in good agreement with the values reported in 2011 [36]. The degenerate cross section is a factor of two better in Rb partially because the $\Delta\nu = 35 \text{ cm}^{-1}$; however, cesium has longer wavelengths and a larger A coefficient for the upper $P \rightarrow D$ transitions, so using non-degenerate pumping maybe advantageous. We also note that our pump source bandwidth (1.5 GHz) is much less than the ground state hyperfine splitting. From the excited 6D state, direct 1 photon ionization will compete with lasing and relaxation. In the absence of these process the saturation intensity is low [44]:

$$I_s = I_S^* \sqrt{2 \frac{g_{6D_{3/2}}}{g_{6S_{1/2}}} - 1} = 66 \frac{\text{kw}}{\text{cm}^2} \quad (27)$$

where $I_S^* = \sqrt{h\nu\Gamma/\sigma^{(2)}}$ and $\Gamma = \sum_i A_{6D_{3/2} \rightarrow i}$. These I_s estimates are also applicable to the non-degenerate two-photon cross sections.

Non-Degenerate Two-Photon Absorption

To further enhance the cross-section, two non-degenerate pump fields can force $\Delta\nu$ to a semi-arbitrary position. For two fields, we define the electric field as $\mathbf{E} = \left(E_1 e^{-i\omega_1 t} + E_1^\dagger e^{i\omega_1 t}\right) \hat{e}_1 + \left(E_2 e^{-i\omega_2 t} + E_2^\dagger e^{i\omega_2 t}\right) \hat{e}_2$ and the time averaged intensity is

Table 8. Degenerate two-photon absorption cross-sections $\sigma_{if}^{(2)}(\nu_0)$ for the cesium 6S $\rightarrow$ 6D transitions; theoretical predictions of equation 25. The $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ transition has a degenerate wavelength of 885.4 nm, the virtual level separation given by $\Delta\nu$. The $6^2S_{1/2} \rightarrow 6^2D_{5/2}$ transition has a degenerate wavelength of 883.7nm.

Transition	$\Delta\nu(\text{cm}^{-1})$	$A_i, A_f(10^7/s)$	$\sigma^{(2)}(\text{cm}^4/\text{W})$
$6^2S_{1/2} \rightarrow 6^2D_{3/2}$	116	$2.86^a, 1.8^b$	$3.36 \cdot 10^{-21}$
$6^2S_{1/2} \rightarrow 6^2D_{5/2}$	416	$3.28^a, 1.52^c$	$0.372 \cdot 10^{-21}$

^a[79], ^b[71], ^c[48]

$I = 2nc\epsilon_0(E_1^2 + E_2^2) = I_1 + I_2$. We assume both pump fields have equal direction and polarization, and define the upward rate as

$$\langle R_{i \rightarrow f}^{(2)(2field)} \rangle = \sigma_{if}^{(2)(2field)} \frac{I_1 I_2}{h\nu} \quad (28)$$

where $\sigma_{if}^{(2)(2field)}$ now denotes the non-degenerate cross-section given by:

$$\begin{aligned} \sigma_{if}^{(2)(2field)}(\nu_1, \nu_2) &= \frac{9}{5} \frac{\nu}{256\pi^4 \hbar n^2 c^2} \sum_n \frac{g_f}{g_i} \lambda_{fn}^3 A_{fn} \lambda_{ni}^3 A_{ni} \\ &\times \frac{(\nu_1 + \nu_2 - 2\nu_{ni})^2}{(\nu_1 - \nu_{ni})^2 (\nu_2 - \nu_{ni})^2} \\ &\times \rho_f(\nu_{fi} = \nu_1 + \nu_2) . \end{aligned} \quad (29)$$

We choose the arbitrary convention of using the wavelength that connects the ground state to the virtual for ν , this does not effect the actual rate and allows for easier comparison across the degenerate and non-degenerate cases. The two fields now allow us to chose a $\Delta\nu$ as shown in Table 9. This amounts to ~ 20 x improvement in cross section, but at the requirement of two pump sources. If $I_{P1} = I_{P2}$ then we have a net gain of ~ 5 x which is still substantial and likely the best two-photon pump values possible in the alkali vapors. We limit $\Delta\nu$ to be greater than 30 cm^{-1} because

Table 9. Non-degenerate two-photon absorption cross-sections $\sigma_{if}^{(2)(2field)}(\nu_{p1}, \nu_{p2})$ for the cesium $6S \rightarrow 6D$ transitions; theoretical predictions of equation 29.

Transition	$\Delta\nu(\text{cm}^{-1})$	$\lambda_{p1}, \lambda_{p2}(\text{nm})$	$\sigma^{(2)}(\text{cm}^4/\text{W})$
$6^2S_{1/2} \rightarrow 6^2D_{3/2}$	30	892.20, 878.69	$6.56 \cdot 10^{-20}$
$6^2S_{1/2} \rightarrow 6^2D_{5/2}$	30	854.5, 918.6	$7.71 \cdot 10^{-20}$

perturbation theory will breakdown if $\Delta\nu$ becomes small. We also want to maintain the future possibility of diode pumping where the diode bandwidth may also be an issue. One example of this breakdown can be found in [67]—the first paper claiming two-photon lasing—and 30 cm^{-1} is consistent with peak behavior in their results.

Two-Photon Emission and Lasing

The above approach to non-degenerate absorption can be applied to emission as well. We define the two-photon emission cross section $\sigma_{fi}^{(2)(2field)}$ in the usual manner:

$$\sigma_{if}^{(2)(2field)} = \frac{g_f}{g_i} \sigma_{fi}^{(2)(2field)} \quad (30)$$

In pumping the cesium 6D states we see nonlinear behavior on the $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ and $5^2D_{3/2} \rightarrow 6^2S_{1/2}$ transitions given in Table 10. We use $18535.5286 \text{ cm}^{-1}$ for the $7^2S_{1/2}$ state [104], and A coefficients from Heavens [48]; for the $5^2D_{3/2}$ state we use $14499.2568 \text{ cm}^{-1}$ [31], and A coefficients from Safronova [81].

If a population inversion exists, then two-photon lasing is possible with the pump acting as one of the stimulating fields as shown in Figure 24. Such a population inversion could be temporal or steady state. The high gain in the alkali vapors cause the system to reach steady state in $< \text{ns}$, therefore even our 10 ns pulses achieve quasi CW behavior.

We therefore must prove that a steady state two-photon inversion is possible. We

Table 10. Non-degenerate two-photon emission cross-sections $\sigma_{fi}^{(2)(2field)}(\nu_{p1}, \nu_{p2})$ for the cesium $7S \rightarrow 6S$ and $5D \rightarrow 6S$ transitions; theoretical predictions of equation 30.

Transition	$\Delta\nu(\text{cm}^{-1})$	$\lambda_{p1}, \lambda_{p2}(\text{nm})$	$\sigma^{(2)}(\text{cm}^4/\text{W})$
$7^2S_{1/2} \rightarrow 6^2S_{1/2}$	115	892.20, 878.69	$4.56 \cdot 10^{-21}$
$7^2S_{1/2} \rightarrow 6^2S_{1/2}$	30	892.20, 1365	$4.93 \cdot 10^{-20}$
$5^2D_{3/2} \rightarrow 6^2S_{1/2}$	115	885.55, 3120	$1.23 \cdot 10^{-20}$
$5^2D_{3/2} \rightarrow 6^2S_{1/2}$	30	892.20, 3039	$1.73 \cdot 10^{-19}$

analyze the case of the $6^2S_{1/2} \rightarrow 6^2D_{5/2}$ pump with two-photon lasing on the $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ and assume the bleached limit for all fields. If we only consider optical transitions, there will be no inversion: the ground state population is n_0 , $6^2D_{5/2}$ is $3n_0$, $7^2P_{3/2}$ is $2n_0$, and $7^2S_{1/2}$ is n_0 . Therefore the inversion from $7^2S_{1/2}$ to $6^2S_{1/2}$ is zero. In this limit, it is unlikely that previous work such as [67] achieve steady state two-photon lasing in lithium. However, cesium (and Rb) has a comparatively larger fine structure splitting on the upper states, that have near gas kinetic mixing rates. For example, if we assume the fine structure mixing rate is very large, we now have n_0 in the ground state, $3n_0$ in $6^2D_{5/2}$, and $2\text{Exp}[\theta_{6D}]n_0$ in $6^2D_{3/2}$ where $\theta_{6D} = \Delta E_{6D}/kT$. If we assume we lase $6^2D_{5/2} \rightarrow 7^2P_{3/2}$, mix with in the 7P states and lase $7^2P_{1/2} \rightarrow 7^2S_{1/2}$, we now have a population in the $7^2S_{1/2}$ state of $\text{Exp}[\theta_{7P}]n_0$ and our two-photon inversion is $\Delta n = (\text{Exp}[\theta_{7P}] - 1)n_0$. If we can instead lase $6^2D_{3/2} \rightarrow 7^2P_{3/2}$, mix 7P and lase $7^2P_{1/2} \rightarrow 7^2S_{1/2}$, our two-photon inversion is $\Delta n = (\text{Exp}[\theta_{7P} + \theta_{6D}] - 1)n_0$.

In other words we use propose same inversion process as the conventional DPAL, where the mixing takes enough population out of the ground state to create an inversion. In our recent work [44] (Chapter IV) we showed the impact of these strong mixing rates where when pumping to the $6^2D_{5/2}$ state, strong cascade lasing is observed on the 3096 nm and 1376 nm lines that can only be produced from spin-orbit

mixing.

Again, in the lighter alkali vapors $\Delta n = (\text{Exp}[\theta] - 1)n_0 \approx 0$ and no definitive proof of a two-photon inversion has been published. Most literature points to multi wave mixing [51, 76, 15] as the dominant nonlinear mechanism.

Multi Wave Mixing

When two-photon pumping the 6D states the following four wave mixing mechanism is possible: $\omega_{BLUE} = 2\omega_P - \omega_{IR}^{12\mu m}$ with the non-axial phase matching condition $2\mathbf{k}_P = \mathbf{k}_{BLUE} + \mathbf{k}_{IR}^{12\mu m}$, where ω_{BLUE} refers to the $7P \rightarrow 6S$ transition, ω_P is the degenerate two-photon pump, and $\omega_{IR}^{12\mu m}$ is from the $6D \rightarrow 7P$ transition. Similar examples of the polarization and susceptibility can be found in [10, 110, 50] and for our case take the form

$$\begin{aligned} \mathbf{P}^{(3)}(\omega_{BLUE}) &= \epsilon_0 \chi^{(3)}(-\omega_{BLUE}, \omega_P, \omega_P, -\omega_{IR}) \\ &\quad \times E_P(\omega_P) E_P(\omega_P) E_{IR}(\omega_{IR}) , \end{aligned} \quad (31)$$

where $\chi^{(3)}$ is the third-order susceptibility:

$$\begin{aligned} \chi^{(3)}(-\omega_{BLUE}, \omega_P, \omega_P, -\omega_{IR}) &= \\ K \frac{N_i}{\epsilon_0 \hbar^3} \sum_{jkl} \frac{\mu_{ij} \mu_{jk} \mu_{kl} \mu_{li}}{(\omega_{ji} - \omega_P)(\omega_{ki} - 2\omega_P - i\gamma_{ki})} \\ &\quad \times \frac{1}{(\omega_{li} - \omega_{BLUE} - i\gamma_{li})} , \end{aligned} \quad (32)$$

and $\omega_{nm} = (E_n - E_m) / \hbar$, the damping rate $\gamma_{nm} = \frac{1}{2} (\Gamma_n + \Gamma_m) + \gamma_{nm}^{(c)}$, Γ_n is the total decay rate out of level n, and $\gamma_{nm}^{(c)}$ accounts for decoherence due to all other sources, (e.g. collisions), and K is a constant introduced by Orr [68]. The i subscript on the population N_i is included to point out that the population is distributed to the upper

levels via cascade lasing, individual populations are non trivial and dispersed among levels not involved in the mixing process.

For SWM with two-pump photons and a seed photon from the same pump we have the non-axial condition $\omega_{2photon} = 2\omega_P - \omega_{IR}^{12\mu m} - \omega_{IR}^{3\mu m} - \omega_P$ and $2\mathbf{k}_P = \mathbf{k}_{2photon} + \mathbf{k}_{IR}^{12\mu m} + \mathbf{k}_{IR}^{3\mu m} + \mathbf{k}_P$. Examples of axial phase matching in sodium and rubidium can be found in the references given above [60, 15, 76]. For this publication we limit our focus to the non-axial phase matching case as we shall explain in the results. The fifth-order polarization takes the form:

$$\begin{aligned} \mathbf{P}^{(5)}(\omega_{2photon}) = \\ \epsilon_0 \chi^{(5)}(-\omega_{2photon}, \omega_P, \omega_P, -\omega_{IR}^{12\mu m}, -\omega_{IR}^{3\mu m}, -\omega_P) \\ \times E_p^3(\omega_P) E_{IR}^{12\mu m}(\omega_{IR}^{12\mu m}) E_{IR}^{3\mu m}(\omega_{IR}^{3\mu m}) . \end{aligned} \quad (33)$$

where $\chi^{(5)}$ is the fifth-order susceptibility:

$$\begin{aligned} \chi^{(5)}(-\omega_{2photon}, \omega_P, \omega_P, -\omega_{IR}^{12\mu m}, -\omega_{IR}^{3\mu m}, -\omega_P) = \\ K' \frac{N_i}{\epsilon_0 \hbar^5} \sum_{jklmn} \frac{\mu_{ij} \mu_{jk} \mu_{kl} \mu_{lm} \mu_{mn} \mu_{ni}}{(\omega_{ji} - \omega_P)(\omega_{ki} - 2\omega_P - i\gamma_{ki})} \\ \times \frac{1}{(\omega_{li} - 2\omega_P + \omega_{IR}^{12\mu m} - i\gamma_{li})} \\ \times \frac{1}{(\omega_{mi} - 2\omega_P + \omega_{IR}^{12\mu m} + \omega_{IR}^{3\mu m} - i\gamma_{mi})} \\ \times \left[\frac{1}{(\omega_{ni} - \omega_P)} + \frac{1}{(\omega_{ni} - \omega_{2photon})} \right] . \end{aligned} \quad (34)$$

We have limited our consideration to the largest known terms. Given the complexity of $\chi^{(5)}$ its possible this form neglects some resonances. With the basic theory covered we now address the experiment.

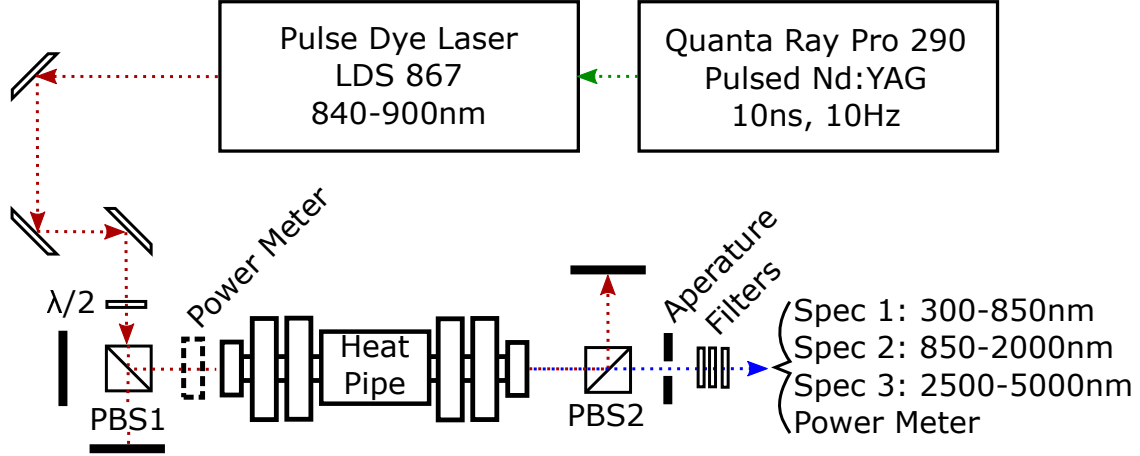


Figure 25. Experiment schematic. Backwards lasing data was also captured by directing the pump into PBS2.

5.3 Experiment

A pulsed dye laser is used to pump the cesium 6D states in the vicinity of the degenerate two-photon process of Table 8 and Figure 24. A schematic diagram of the experimental apparatus is provided in Figure 25. Three spectrometers are used to characterize over 20 lasing transitions at wavelengths ranging from 386 - 3641 nm. Only a single pulsed dye laser was available so the non-degenerate pump case was not studied.

The pump source is a Sirah Precision Scan D pulsed dye laser, with LDS 867 dye, and is pumped by a Quanta Ray Pro 290 Nd:YAG laser. This provides 10 Hz, 10 ns pulses, with ~ 1.5 GHz spectral width with a tunable range much greater than the 6D states. The pump has two amplification stages giving up to 30 mJ/pulse at 885.4 nm. No focusing of the pump was required and allowed for improved mode volume overlap for stimulated emission. The laser spot size was ~ 8.7 mm, yielding peak instantaneous intensities exceeding 4 MW/cm^2 . Pulse energy is tuned with a half wave plate and polarized beam splitter to maintain a constant pulse shape (tuning pump energy with the Nd:YAG pump can lead to shorter or longer pulse and add a

nonlinear influence the peak intensity).

The heat pipe consists of a stainless steel tube 29 cm long, 2.5 cm diameter. It is filled with 0-10 Torr of He. Seven temperature zones were used. We kept the cell windows at 200 °C with Watlow Ez-zone Heaters and heat wraps. Four intermediary zones were created by flowing heated water through aluminum blocks; these served as the cold zones at 38 °C (outer) and 48 °C (inner). The central oven zone was made with a 15 cm aluminum block and heated with a Watlow heater from 150 °C to 330 °C (Cs density of 2×10^{14} to $8 \times 10^{16}/\text{cm}^3$). This configuration resulted in > 18 months of operation with no maintenance.

Three spectrometers were required. The first (455 nm to 850 nm) was an Acton SpectraPro 275 (.275 m focal length with ± 0.1 nm resolution), with a Princeton Instruments PI-Max2 ICCD. The input beam was attenuated with PBS cube, and two ND filters (OD 1.6 total). Spectrometer 2 (850 nm to 2000 nm) was a McPherson 218 (.3 m focal length with ± 0.12 nm resolution) with a Roper Scientific OMA V 1x1024 InGaAs array. A fiber coupler was numerical aperture matched with the spectrometer to capture 6.3 mm of the 8.7 mm output beams. To cover the $2.5 \mu\text{m}$ to $4 \mu\text{m}$ window, we used a Jobin Yvon Triax 320 (.3 m focal length with a $\pm .3$ nm resolution) equipped with an Electro-Optical-Systems IS-020-E-LN7 single element InSb detector with oscilloscope detection. For the IR observations, the PBS2 depicted in Figure 25 was swapped with a 45 deg dielectric mirror on a fused silica substrate that gave <.05% transmission at pump wavelengths and >50% transmission from $2.9 \mu\text{m}$ to $3.7 \mu\text{m}$. The spectrometer was free-space coupled with two gold mirrors and matched with a 35 mm CaF_2 Lens. Data collection was performed with integration times of 100 ms with 500 accumulations to average 500 spectra. Spectrometer intensity calibration was done with a calibrated Oriel 63355 lamp, powered by an Oriel 68831 radiometric power supply.

Laser output energies were recorded with a Laser Probe RjP-445 sensor and Rm-3700 meter. Each reading was taken with 500 pulses to track the average and standard deviation. Measurements were taken with the following filters: 455 nm with 10 nm bandpass, 532 nm long pass, 850 nm with 10 nm band pass, 900 nm long pass, 1100 nm long pass, 1325 nm with 50 nm band pass, 1500 nm with 50 nm band pass, and 2700-3975 nm broadband pass filter.

Backwards measurements were taken with the McPherson 218 and Laser Probe RjP-445 sensor in the same positions by pumping the heat pipe in reverse through PBS2, from right to left in Figure 25. Further details can be found in [44] (Chapter IV).

5.4 Results

Off-Resonance Behavior of One-Photon Lines

In this section we cover some of the lasing spectra and energy measurements scanning over the 6D pump wavelengths at various intensities. We believe the lines are produced by cascade lasing with the exception of the transitions connected to the ground state where, in the absence of a population inversion, FWM, and SWM maybe the dominant mechanism [45, 35, 15].

In Figure 26 we plot the intensity of the strongest lines, from 455 nm to 1469 nm, as the pump is tuned across the 6D states; we compare this with the total measured lasing energy. We omit the D_1 and D_2 lines because their pump tuning is very narrow. The data features two sets of peaks centered on the $6^2D_{5/2}$ and $6^2D_{3/2}$ two-photon resonances. The overall behavior is consistent with the cross-sections given in Table 8 where the strong $6^2D_{3/2}$ two-photon cross section gives rise to much more interesting behavior. For instance, the $6^2D_{5/2}$ peaks exhibit, lower lasing energies, lower total lasing energy, and small, Gaussian-like wavelength tuning.

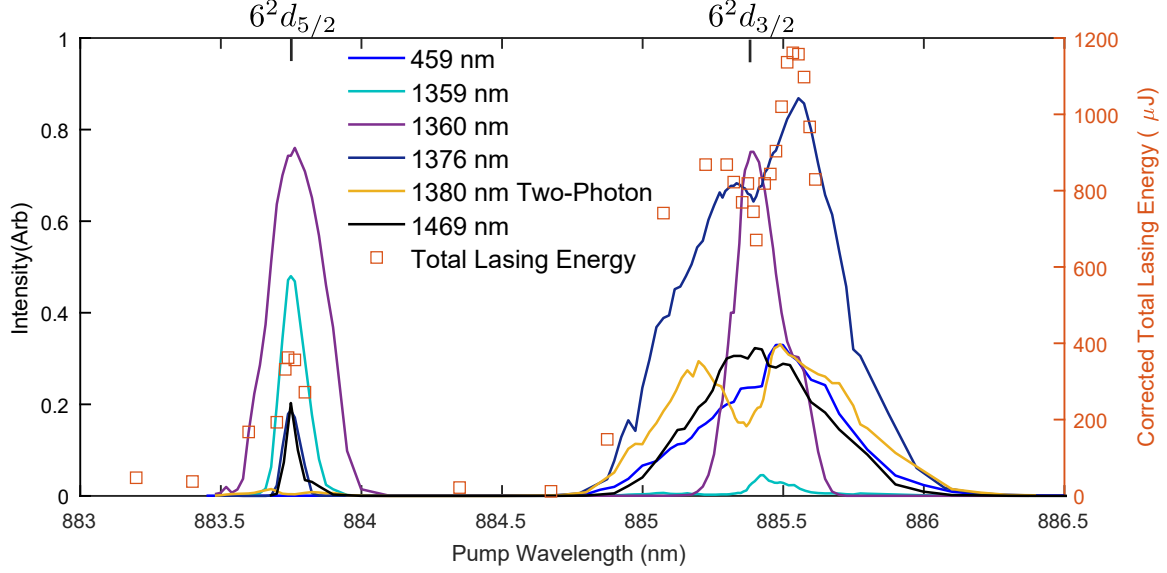


Figure 26. Pump wavelength tuning vs lasing spectra for the strong cascade lasing lines and two-photon line. Data taken at 237.5 °C, 5 Torr He, Pump 28 mJ/pulse. Right axis shows the total lasing energy (excluding 3 μm power) given by the orange squares.

In contrast, the $6^2D_{3/2}$ data shows that only some of the lasing wavelengths peak on-resonance. Off-resonance wavelengths follow the strongest one-photon cross sections path, and display asymmetrical off-resonance behavior. This behavior is highlighted in Table 11, where we present estimates for the cross-sections and compare the on and off-resonance lasing energies. These energies are estimated by a combination of calibrated spectra and energy measurements where we correct for known transmission losses from various filters and the cell windows, and backwards lasing energy when known. Tuning widths from the $6^2D_{3/2}$ state in Figure 26 are > 1 nm, and likely created by a combination of saturation broadening (saturation parameter $I/I_S \approx 80$ is very large), SHRS, and/or a broad range of phase matching conditions. However, we believe saturation broadening is the dominant mechanism because this leads to the creation of ASE fields required to pump some of the other processes; it is also a lower order optical process. For the one-photon lines we note that off-resonant enhancement is seen in the 459 nm line, and a strong enhancement is seen in the 1376

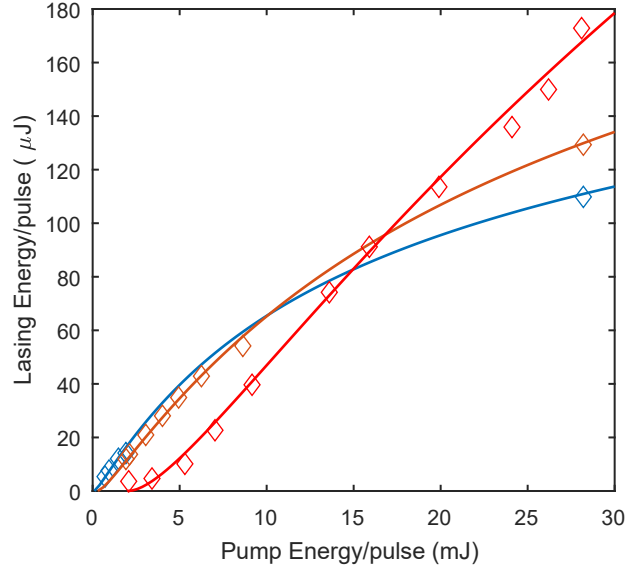


Figure 27. Off-resonance pump enhancement of the infrared cascade lasing 3096 nm line. $\diamond$ data points are from corrected, filtered, energy meter measurements and the solid lines are from fits. Plot shows the intensity (Energy) scaling of the 3096 nm (and 2932 nm) for on-resonant pump (885.399 nm in blue) and off-resonant pump (885.429 nm in orange and 885.499 nm in red) wavelengths. This plot is also representative of the enhancement seen on the 1376 line. Experimental conditions: 5 Torr He, and 237.5 °C.

nm line and 3096 nm line. All of these enhancements are strongly correlated to the presence of 2 two-photon lines on the $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ and $5^2D_{3/2} \rightarrow 6^2S_{1/2}$ that we discuss in the next section.

To gain further insight into the off-resonant enhancement of the 1376 nm and 3096 nm lines, we plot the lasing energy as a function of pump energy for the 3096 line in Figure 27. We include results for three pump wavelengths from 885.399 nm ($6^2D_{3/2}$ resonance) to 885.499 nm.

Our fitting method described in [44] (Chapter IV) accounts for a large part of the radial behavior differences between our filtered energy meter measurements which observe nearly all of the beam, and our integrated spectral measurements we see only

Table 11. Comparison of lasing energies at max pump energy for on-resonance pump (885.4 nm) and off-resonance pump (885.55 nm, 885.5 nm for 3 μ m) lasing energies. Experimental condiction: pump energy of 28.3 mJ/pulse (on-res.), 26.6 mJ/pulse (off-res.) 5 Torr He, at 237 °C.

$\lambda(\text{nm}): \text{Transition}$	Cross-Section $\sigma_{fi} \text{ cm}^2$	On-Res. (μJ)	Off-Res. (μJ)
459.45: $7^2P_{1/2} \rightarrow 6^2S_{1/2}$	$6.84 \cdot 10^{-14}$	74 ± 15	81.6 ± 16
852.35: $6^2P_{3/2} \rightarrow 6^2S_{1/2}$	$1.81 \cdot 10^{-11}$	110 ± 50	1.63 ± 1
894.59: $6^2P_{1/2} \rightarrow 6^2S_{1/2}$	$1.82 \cdot 10^{-11}$	79 ± 35	1.57 ± 1
1359.2: $7^2S_{1/2} \rightarrow 6^2P_{1/2}$	$1.39 \cdot 10^{-11}$	< 6	2.8 ± 0.4
1360.6: $7^2P_{3/2} \rightarrow 5^2D_{5/2}$	$2.46 \cdot 10^{-12}$	220 ± 28	70.8 ± 11
1376.3: $7^2P_{1/2} \rightarrow 5^2D_{3/2}$	$3.86 \cdot 10^{-12}$	145 ± 17	360 ± 58
1381.0: $7^2S_{1/2} \rightarrow 6^2S_{1/2}$	$\sigma^{(2)(2f)} I_P = 2.0 \cdot 10^{-14}$	25.6 ± 3.0	80 ± 13
1469.9: $7^2S_{1/2} \rightarrow 6^2P_{3/2}$	$3.22 \cdot 10^{-11}$	105 ± 13	84.1 ± 10
2931.3: $7^2P_{3/2} \rightarrow 7^2S_{1/2}$	$9.07 \cdot 10^{-11}$	33.1 ± 6.3	42.4 ± 8.1
3096.1: $7^2P_{1/2} \rightarrow 7^2S_{1/2}$	$9.29 \cdot 10^{-11}$	82.4 ± 16	120 ± 23
3120.0: $5^2D_{3/2} \rightarrow 6^2S_{1/2}$	$\sigma^{(2)(2f)} I_P = 5.5 \cdot 10^{-14}$	1.6 ± 0.3	21.0 ± 4.0

around half. This method fits is a modification of the form

$$E_L = E_m \left(1 - \exp \left[\frac{\eta}{E_m} (E_P - E_t) \right] \right) \quad (35)$$

where the slope efficiency is η , lasing threshold is E_t and bleached limit is E_M . The functional form for this model is based on a one photon pump mechanism, and does not capture nonlinear behaviors near threshold, but allows for estimation of the three key features. For our case model radial behavior with a Gaussian pump intensity

distribution and integrate:

$$E_L(E_p) = 2\pi\xi \int_0^{E_p(r)=E_t} E_m \left(1 - \exp \left[-\frac{\eta}{E_m} \left(E_{p0} e^{-\frac{r^2}{2c^2}} - E_t \right) \right] \right) r dr \quad (36)$$

where c is a Gaussian standard deviation that's related to the fixed pump beam radius (we use a value of $c=2.175$ mm which corresponds a $1/e^2$ diameter of 8.7 mm), and ξ is an additional parameter set to 1.3 for all fits. We use two different limits of integration for each measurement type: the raw filtered measurements are integrated from 0 to lasing threshold, the spectral measurements—which has an aperture—the limits of integration are 0 to 6.3 mm/2. This allows us to simultaneously fit equation 36 to both data sets. These fits shows that as the off-resonant wavelength increases from a deviation of 0.0 nm $\rightarrow$ 0.03 nm $\rightarrow$ 0.1 nm, the following effects are seen: the lasing threshold, E_t , increases from .064 mJ $\rightarrow$.21 mJ $\rightarrow$ 1.7 mJ; the bleached limit, E_m , increases from 39 μ J $\rightarrow$ 60 μ J $\rightarrow$ $> 200 \mu$ J; and the slope efficiency, η , remains fairly constant 1.2% $\rightarrow$ 0.9% $\rightarrow$ 1.0%. Errors in our fit parameters are around $\pm 40\%$ as detailed in previous work [44] (Chapter IV).

Off-Resonance Behavior of Two-Photon Lines

The two photon processes observed in Figure 26 and Table 11 also exhibit interesting behavior for off-resonance pump conditions. The wavelength tuning of these lines are determined by their creation mechanism. These two-photon lines can be created by axial SWM, but phase matching requires an additional on-axis field close to but not on the pump (some estimates are given in [15] typically >3 nm off the pump, but no estimates are given for our case. Creation of these lines are sometimes seeded by the wings of large pulsed dye pump widths [60, 15]. However, our pulsed dye source

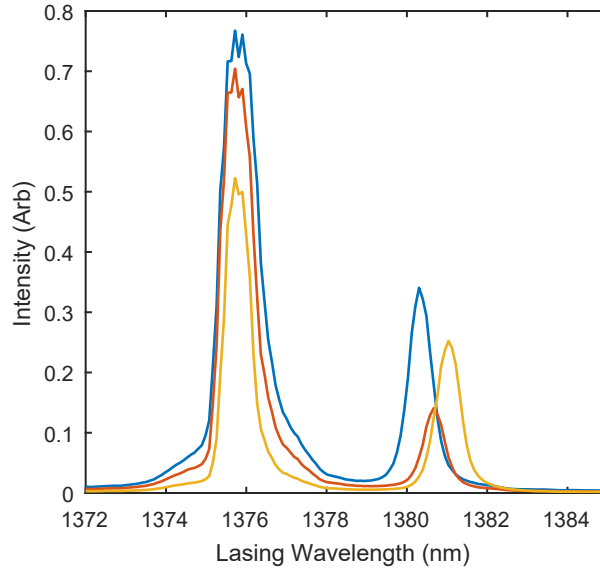


Figure 28. Example spectra to illustrate wavelength tuning for the 1380 nm two-photon line at on-resonance pump (orange 885.399 nm) minimum and off-resonance pump maximums (885.499 nm blue and 885.199 nm in yellow). The $7^2P_{1/2} \rightarrow 5^2D_{3/2}$ 1376 nm line is also shown. Experimental conditions: 237.5 °C, 5 Torr He, pump energy 15 mJ/pulse.

features a narrow bandwidth (~ 1.5 GHz). Therefore, the only options are non-axial SWM and two-photon lasing, which are indistinguishable from a wavelength tuning perspective because both use the same pump field.

We first look at the $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ or 1380 nm line. In Figure 28, we give three spectra of two-photon tuning through resonance. There are two important features in this figure. First, the two-photon line is very strong. We include the 1376 nm line, the strongest line in our system, with the spectra for comparison. This indicates potential future utility as a tunable source. Secondly, the line tunes with pump wavelength. This tuning is shown for a significant number of spectra in Figure 29, and we look at the error in Figure 30. Though a slight downward trend is seen, the effect is below our spectral resolution of ± 0.12 nm for our .3 m spectrometer. Therefore it is a two-photon process coupled to the pump, created by SWM or two-photon lasing.

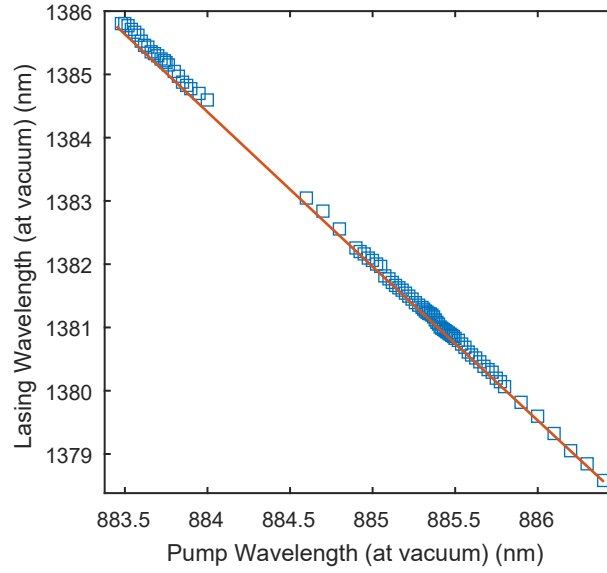


Figure 29. Wavelength tuning for the 1380 nm two-photon line vs theory. Data $\square$ (blue) and theory —(orange). Experimental conditions: 237.5 °C, 5 Torr He, pump 885.5 nm, 15 mJ/pulse.

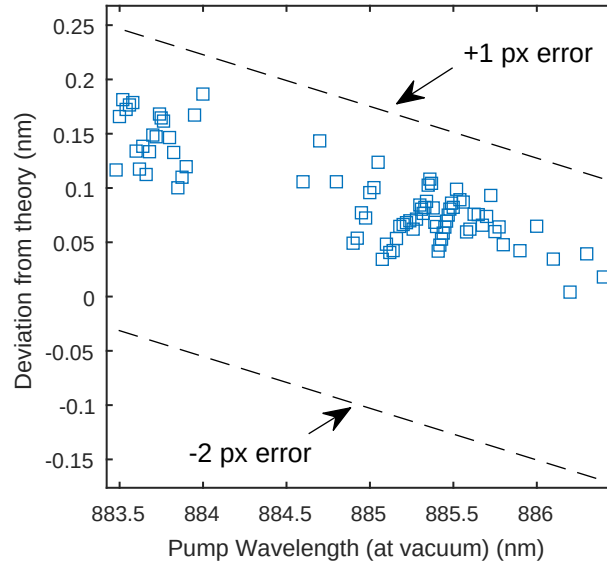


Figure 30. Deviation from theory. Instrument accuracy is ± 0.1 nm or ± 1.5 pixels for our 1x1024 InGaAs array.

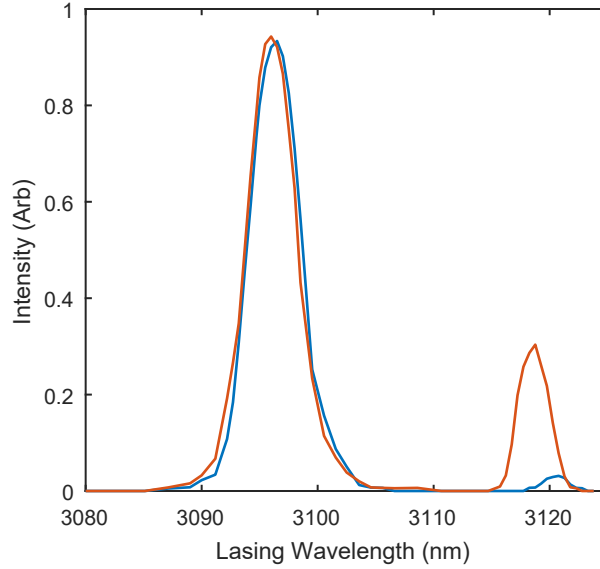


Figure 31. Wavelength tuning for the 3120 nm two-photon line at on-resonance pump minimum (885.399 nm blue) and off-resonance pump maximum (885.549 nm orange). The $7^2P_{1/2} \rightarrow 7^2S_{1/2}$ 3096 nm line is also shown. Experimental conditions: 237.5 °C, 5 Torr He, Pump 26 mJ/pulse.

The second two-photon line is seen around 3120 nm from the $5^2D_{3/2} \rightarrow 6^2S_{1/2}$ illustrated in Figure 31. Again we note the presence of a strong tunable line next to the 3069 nm line which is the strongest of the 3 μm cascade lasing lines. To be consistent with the values in Table 11, the 3096 nm one-photon line has a peak intensity when pumped at 885.499 nm. The wavelength tuning is plotted for the two-photon line in Figure 32. We report fewer points for the 3120 nm line due to faster data acquisition for the 1381 nm line.

Two-Photon Intensity Scaling

We performed intensity or energy scaling measurements of the 1381 nm two-photon line for on and off-resonance wavelengths. We use two separate sources to obtain intensity measurements. The first is data from integrated lasing energy from

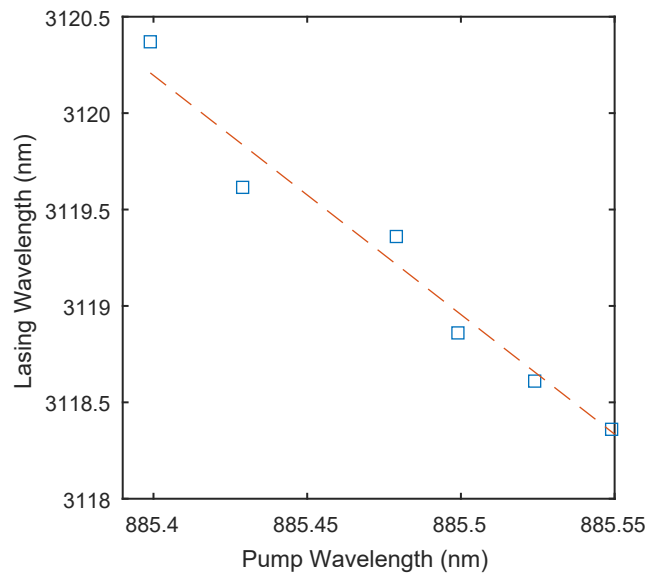


Figure 32. Wavelength tuning for 3120 nm two-photon line vs theory. Data $\square$ (blue) and theory --(orange). Experimental conditions: 237.5 °C, 5 Torr He, Pump 885.5 nm, 26 mJ/pulse.

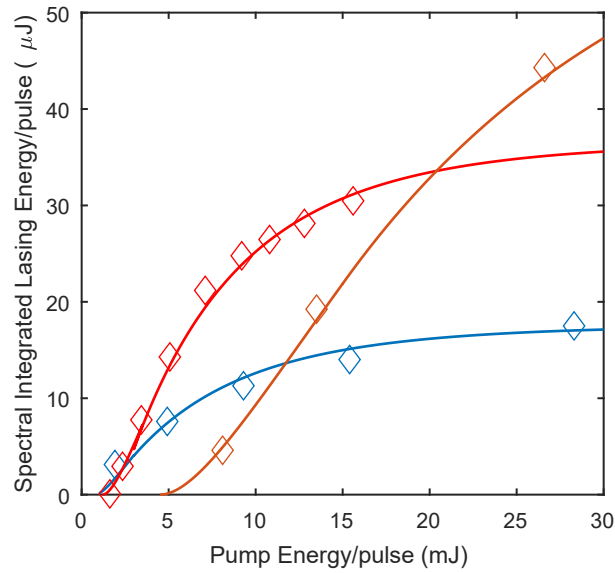


Figure 33. Energy (Intensity) scaling of the 1380 nm two-photon line for resonant (885.399 nm blue) and off-resonant (885.435 nm red and 885.549 nm orange) pump wavelengths data from integrated spectral energy. Experimental conditions: 237.5°C, 5 Torr He.

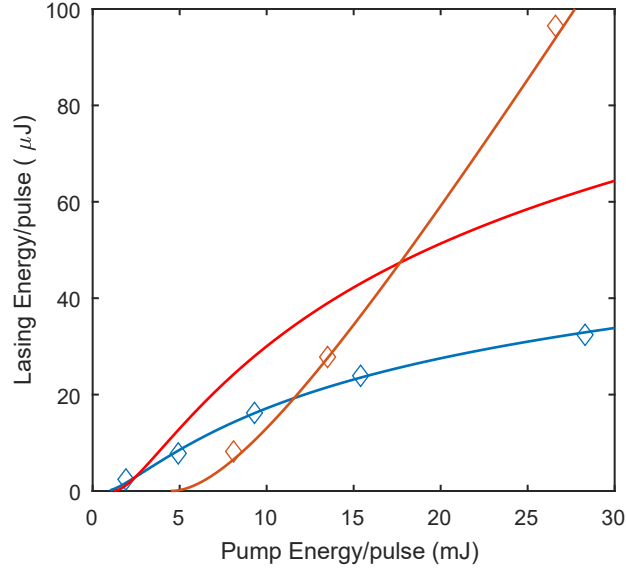


Figure 34. Energy (Intensity) scaling of the 1380 nm two-photon line for resonant (885.399 nm blue) and off-resonant (885.435 nm red and 885.549 nm orange) pump wavelengths. Data from corrected direct energy meter measurement. Experimental conditions: 237.5°C, 5 Torr He.

calibrated spectra that is corrected for various transmission losses in our apparatus. The second is from filtered energy meter measurements, and the measurements are again corrected for transmission losses. We fit the two data sets to equation 36. The data and fit to the integrated spectral energies are given in Figure 33. The data and fit to the direct energy measurements are given in Figure 34.

These fits provide estimates and trends for slope efficiency η , lasing threshold E_t , and bleached limit E_m . The trends in slope efficiency, in order of on to off-resonance pump wavelength 885.399 nm $\rightarrow$ 885.435 nm $\rightarrow$ 885.549 nm, is 0.4% $\rightarrow$ 0.8% $\rightarrow$ 0.7%. Lasing threshold trends as 0.80 mJ/pulse $\rightarrow$ 1.0 mJ/pulse $\rightarrow$ 3.8 mJ/pulse. And the bleached limits trend as 13.5 μ J $\rightarrow$ 28 μ J $\rightarrow$ >50 μ J. Results up to this point focus on 237.5 °C with 5 Torr He buffer gas. We elaborate on these choices in the next section.

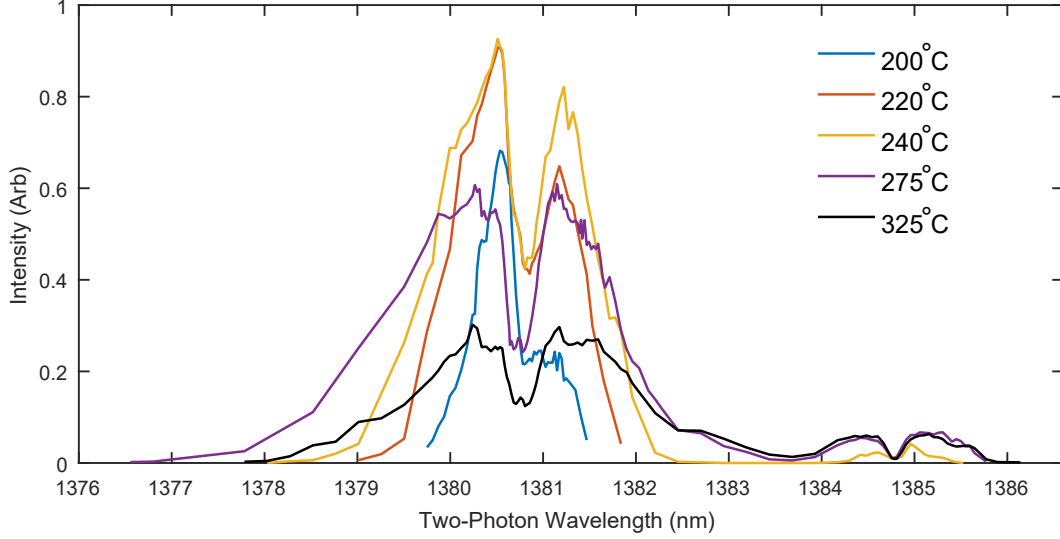


Figure 35. Two-photon wavelength tuning at 5 Torr He for various temperatures and pump energy of ~ 14.3 mJ/pulse.

Influence of Cesium Density and Buffer Gas Pressure

In Figure 35 we show the impact of cesium density scaling in the system. We plot the two-photon wavelength tuning for 5 cesium densities. At heat pipe temperatures of 200 °C we see a strong asymmetry that evolves into a resonance dips. The 1380.9 nm dip is from the $6^2D_{3/2}$ resonance and the 1384.8 nm dip is from the $6^2D_{5/2}$ resonance and occur at higher cesium densities. This dip is seen in multiple sources for similar two-photon processes [76, 60] [67]. Cited reasons for this dip include pump depletion and competition with other nonlinear optical processes. Above 275°C the the two-photon line can be completely resolved over 9 nm of pump wavelength tuning.

The impact of cesium density scaling on the energy scaling of the 1381 nm two-photon line for a fixed pump wavelength is shown in Figure 36. We see a decreasing lasing threshold behavior with increased cesium density: 1.4 mJ/pulse $\rightarrow$ 1.0 mJ/pulse $\rightarrow$ 1.0 mJ/pulse; increasing slope efficiency: 0.5% $\rightarrow$ 0.8% $\rightarrow$ 0.9%; and bleached limit 6.0 μ J $\rightarrow$ 15 μ J $\rightarrow$ 26 μ J.

The roll over point also trends with the number of atoms available. For the 150

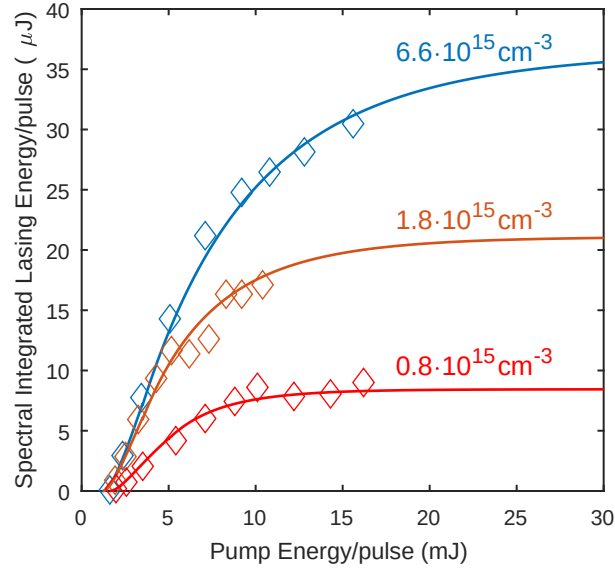


Figure 36. Cesium density and energy scaling of the 1381 nm two-photon line. Data from calibrated integrated spectral energy. Experimental conditions: 180 °C ($0.8 \cdot 10^{15}/\text{cm}^3$), 200 °C ($1.8 \cdot 10^{15}/\text{cm}^3$), and 237.5 °C ($6.6 \cdot 10^{15}/\text{cm}^3$), 5 Torr He, Pump wavelength of 885.435 nm. Include fit parameter discussion, include that at lower temperatures favor wavelengths closer to resonance.

°C case, the number density is $0.8 \times 10^{15}/\text{cm}^3$ the total number of atoms in our gain volume is 5×10^{15} atoms, and roll over occurs around 5 mJ/pulse or 1×10^{16} pairs of pump photons. For the 180°C case, the number density is 1.8×10^{15} the total number of atoms in our gain volume is 1×10^{16} atoms, and roll over occurs around 8 mJ/pulse or 2×10^{16} pairs of pump photons. For the 237.5°C case, the number density is 6.6×10^{15} the total number of atoms in our gain volume is 4×10^{16} , and roll over occurs around 15 mJ/pulse or 3×10^{16} pairs of pump photons. There appears to be a rough correlation between the number of photon pairs available at roll over location and the number of cesium atoms.

In our analysis of the on-resonance pumping behavior [44] (Chapter IV), we note a strong enhancement from the inclusion of 5 Torr of He buffer gas. We show this effect on the 1381 nm two-photon line in Figure 37. The vacuum case shows a asymmetric

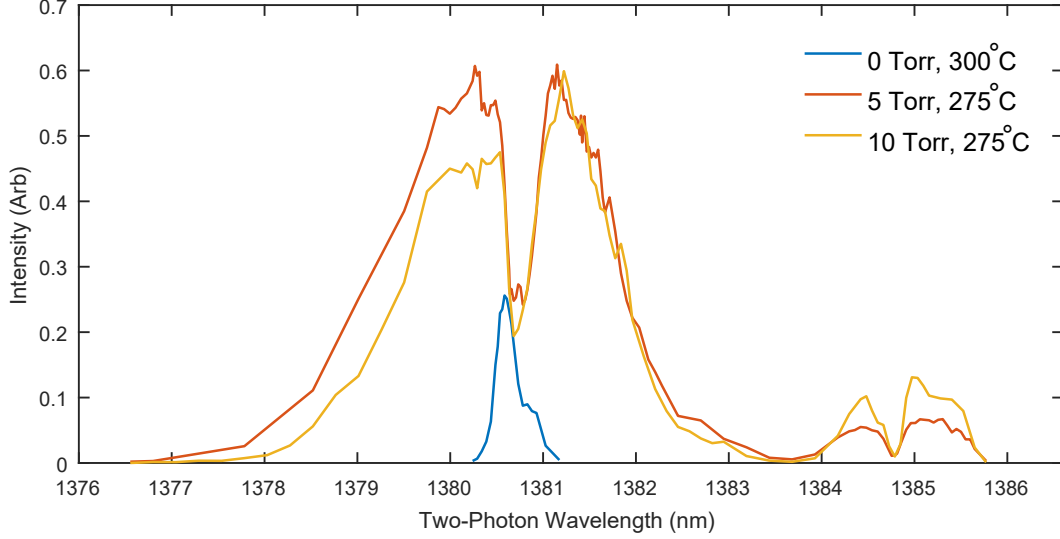


Figure 37. Two-photon wavelength tuning at high temperature for 0, 5 and 10 Torr of He buffer gas, and pump energy of ~ 14.3 mJ/pulse.

small peak, 5 Torr shows the same result from Figure 36, and 10 Torr shows an even more intriguing asymmetric decrease in the $6^2D_{3/2}$ structure and an enhancement to the $6^2D_{5/2}$ structure. This effect is not explored further in this work.

Additional Non-Linear Effects

Two additional lines observed are under unique circumstances. The first unexpected transition is a UV line associated with the $8^2P_{3/2} \rightarrow 6^2S_{1/2}$ shown in Figure 24. This process occurs in the blue wing of the $6^2D_{3/2}$ resonance at 885.399 nm where there is a fortuitous resonance at 885.31 nm associated with the $5^2D_{3/2} \rightarrow 8^2P_{3/2}$ transition. We estimate the energy to be <20 nJ of UV energy which may simply be fluorescence.

We also see a 887 nm line produced when the pump is slightly red shifted from the $6^2D_{3/2}$ state at cell temperature near 275 °C, with an energy <2 μ J. This only appears under a narrow range of pump wavelengths and cell temperatures. One possible explanation is an axial SWM process where the 887 nm line pairs with a

1378 nm emission analogous to the non-axial 1381 nm two-photon line pump pair. However, the associated 1378 nm line is not seen. It is also close to the strong 1376 nm ASE and the 1381 nm two-photon line, and may be too weak to see.

5.5 Discussion

Performance and Comparison with Prior Work

The two-photon processes are efficient, and perhaps some of the strongest two-photon process observed in the alkalis to date. For comparison, Jabbour [50] measured peak output energies ~ 10 nJ/pulse. Pong noted .05% efficiency [76] from which we estimate 5 μ J of two-photon energy; Nikolaus [67] achieved lasing energy of around 18 μ J from a 20% probe gain but this was from three pulsed dye sources of unknown energy so we cant estimate total efficiency. We present 80 μ J two-photon energy, conversion efficiencies $\sim 0.4\%$, slope efficiencies $\sim 0.9\%$, and threshold pump energies of .8 mJ/pulse or a peak intensity of ~ 130 kW/cm². We measure 9 nm tuning, which is significant given the single pump source and saturation broadening. Nikolaus [67] achieved around 40 nm of tuning but that required three focused pulsed dye sources. Pong estimated a narrow tuning range of 0.4 nm [76]. In most of the sources for two-photon emission effects in the alkalis output power, and efficiency were not available.

One of the reasons for the improved performance of this system is the large two-photon cross sections in cesium. In addition, we do not require any focusing of our pump beam; this provides improved mode volume overlap of the pump and lasing beams. By contrast, many of the above references [67, 60, 15] required focusing of the pump to GW/cm² intensities.

We do not observe any axial SWM effects. We attribute to them requiring higher pump energy (focus), and larger pump bandwidth, or the emission may be too weak

for our setup to detect in the present of large emission.

The on-resonance dips shown in Figures 26, 35, and 37 are also noted in other instances [67, 60, 76] of two-photon lines. Cited reasons include pump depletion due to stronger on-resonance absorption, and competition with other nonlinear optical processes. We believe potential causes include stronger on-resonance absorption where more of the energy goes into 1360 nm line which does not pump any two-photon lines. However, this effect is not well understood.

The addition of buffer gas (Figure 37) plays a significant role in our results. Many sources have reported similar enhancement to cascade lasing and multi-photon processes [45, 44, 76]. This effect is not well understood, and has been attributed to various situational sources such as a phase mismatch for MWM, or increasing hyper-fine and spin-orbit mixing for cascade lasing (Chapter IV).

We observe in Figure 36 two-photon scaling with cesium density up to 237.5 °C, and decreased performance at temperatures above 275 °C. Pong attributed a similar fall off with an unfavorable phase mismatch, and stated that the inclusion of Ar buffer gas provided a more favorable phase matching; but this was caused by a Ar resonance near their SWM wavelength. We note that no such resonance exists for our two-photon lines and He. Instead we observe temperature trends consistent cascade lasing lines, where the system as a whole is effected to increasingly higher levels by optical trapping as noted in our recent work [44] (Chapter IV). The high cesium number densities, and presence of buffer gas, may present an environments unfavorable to coherence effects.

Two-Photon Lasing vs Six Wave Mixing

In this section we present several arguments for and against two-photon lasing, followed by a similar discussion for SWM.

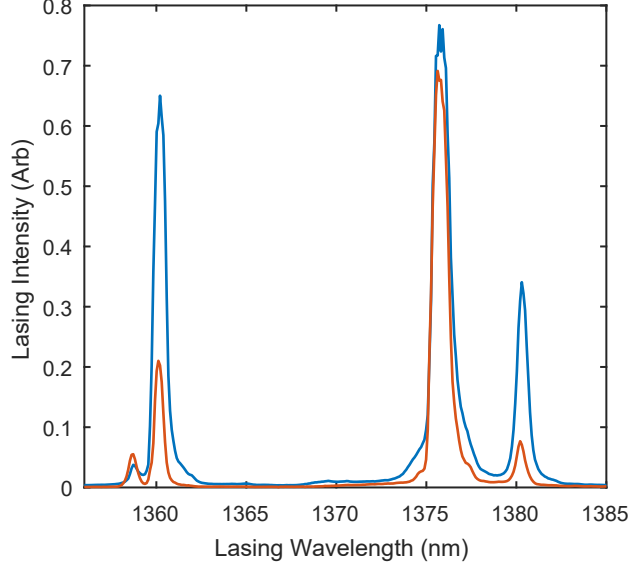


Figure 38. Forward (blue) vs backwards (orange) lasing at 237.5°C , 5 Torr He, pump wavelength 885.5 nm, and pump energy of 26 mJ/pulse.

Cesium vapor has several favorable traits for two-photon lasing. We have shown that the two-photon cross sections are large. There is also a good balance of large mixing rates, and optimal fine structure splitting in the 6D and 7P states to create an inversion. These arguments are supported by Figure 37 where we see strong enhancement with pressure, and by our previous work [44] (Chapter IV) where we showed that fine-structure mixing generated strong lasing on otherwise forbidden transitions.

The intensity scaling of these two-photon processes noted in Figures 33, 34, and 36 are similar to intensity scaling of cascade lasing lines. The rollover behavior in these figures, and correlates to the number of pump photons to number of atoms also favor a two-photon process.

In the criticism following the first claim of two-photon lasing, Jackson [51] noted a few requirements for a potential two photon laser. The first of these is the requirement of directionality. A SWM process will be unidirectional. However, we note a $\sim 20\%$ backwards two-photon field as shown in Figure 38. There is a large degree of

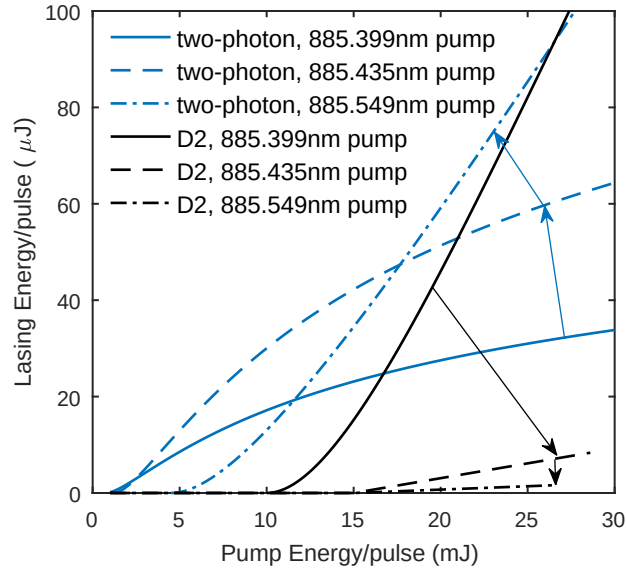


Figure 39. Two-photon line (1380 nm) vs D2 line. Arrows follow wavelength tuning from on-resonance to off-resonance. Experimental conditions: 237.5°C, 5 Torr He.

uncertainty in this measurement because we had to rearrange the optics to reverse the pump direction while leaving the spectrometer and heat pipe in the same place. The pump energy and wavelength were the same for both. This backward two-photon beam is unlikely a back reflection of the forward beam. In the worst case, a back reflected beam would be $\sim 12\%$ of the forward beam intensity for the case of no divergence and perfect cell alignment for back reflections. We do note however that the strong upper cascade lasing states will create backwards fields as well; and the second window back-reflected pump could couple with these fields. However, our intensity scaling data suggests that this is not the case as the backwards pump intensity (~ 3.2 mJ/Pulse) at 885.5 nm would be close to threshold (note the 885.435 and 885.549 nm curves in Figures 33 and 34). This data provides the strongest consideration for a non MWM process.

The second Jackson requirement was proof of overcoming the D_1 and D_2 lines, i.e. competition with one photon parasitic processes. In earlier work [44] (Chapter

IV), we show that optical trapping from the ground state can provide a possible mechanism to control lower lying one-photon processes by imposing a significantly higher lasing threshold. In Figure 39 we show on and off-resonance data of the D_2 line and compare it to the on and off-resonance case of the 1381 nm two-photon line. This figure illustrates the effect of wavelength tuning on both lines. There is a sizable region below 10 mJ/pulse where the D_1 and D_2 lines are not lasing. Furthermore, the effect of tuning on the two-photon cross sections and the fifth order susceptibility is minimal; yet, the two-photon line is significantly enhanced and the opposite happens to the D_2 line. In Figure 26, the cascade lasing lines which pump the D_1 and D_2 lines show a wide range of tuning with the pump. This also holds for the two-photon lines. Even the blue 459 nm line—often considered a FWM process in literature—shows similar tuning and off-resonant enhancement. The D_1 and D_2 lines on the other-hand show minimal tuning and no enhancement. Regardless of the mechanism for the D_1 and D_2 , this data shows that the two-photon line can compete with the one-photon processes.

However, there are several arguments against two-photon lasing as the mechanism. Although the two-photon line can compete with the one photon lines, the sheer number of one-photon processes would be harmful to a maintaining potential inversion. Furthermore, much of our data is from pumping to $6^2D_{3/2}$ state, where it is more difficult to generate an inversion because the 6D energy splitting can not directly contribute, though the 7P can still contribute with a limit of $\Delta n = (\text{Exp}[\theta_{7P}] - 1) n_0$.

Although we show a backward lasing field, this still does not constitute proof of a population inversion. The lack of proof of an inversion was the primary criticism given by Jackson [51] in his criticism of the first claimed two-photon laser by Nikolaus [67]. The primary argument for SWM is that an inversion is not required, and we note above that achieving an inversion is difficult. Another, advantage is the significant

history of similar FWM and SWM in the alkali vapors, given above. The strong cascade lasing we show in previous work [44] (Chapter IV) creates ideal conditions for generating FWM and SWM on transitions connected to the ground state. SWM could also explain the behavior seen in Figure 39. In general the SWM intensity is given by

$$I_{SWM} \propto \text{Exp}[-\alpha L] \times |\mathbf{P}^{(5)}|^2 \times G(\Delta k) \quad (37)$$

which contains an absorption piece $\text{Exp}[-\alpha L]$ and a phase matching piece $G(\Delta k)$. The absorption piece can explain the high threshold of the D_2 line. If the phase matching factor for the D_2 is narrow, it would explain the limited tuning behavior of the D_2 line. The absorption piece for the two-photon line would be comparatively smaller (see cross-sections in Table 11), and the on-resonant dip could explain the difference in behavior. However, no good explanation for this dip exists. It is not likely caused by the polarization. For the range of wavelength tuning in Figure 39, the susceptibility of the two-photon line in equation 34 and its analog for the D_2 line are nearly unchanged.

The tuning range on the D_2 line is significantly narrower than all the others which pump it. If we assume all the transitions to the ground state are FWM or SWM then Figure 26 suggests that these processes have similar widths. However, the data in Figure 39 suggests that the D_1 and D_2 line widths, or range of phase matching conditions, are substantially narrower than any other observed process.

We also note that the ratio of the D_2 to the two-photon polarizations from equation 34 and 33 and the analogs for the D_2 should favor D_2 over the two-photon by a factor of $\gg 10^3$ because the susceptibility of the D_2 process does not have a large $\Delta\omega$ term. Although, this does compete with the absorption which favors the two-photon line.

A major argument against SWM is the intensity scaling. From equation 37, we

expect the two-photon line SWM intensity

$$I_{SWM} \propto |\mathbf{P}^{(5)}|^2 \propto I_P^3 \times I_{3\mu m} \times I_{12\mu m} . \quad (38)$$

However, at high intensities we observe roll over , and at lower intensities we know the intensities of the 3 μm pump field in Figure 27 which implies $I_{SWM} \propto I_P^3$. But this behavior is not observed. The same is true of our 459 nm, D₁, and D₂ observations. For comparison, the FWM mixing observations in Boyd's sodium two-photon pump work [11] were proportional to I_P^2 but the pump intensities $> \text{GW}/\text{cm}^2$ (the pump source was focused).

Similar argument can be made for the cesium density scaling where $I_{SWM} \propto |\mathbf{P}^{(5)}|^2 \propto N^2$. Again this behavior is not observed in Figure 35 or 36. Finally, we note that pressure should add to the collisional decoherence terms in equation 34 but this is not observed in Figure 35.

Ultimately, both two-photon lasing and SWM require a more complete understanding of the ground and excited state populations. The multi wave mixing process has the easier criterion of no population inversion, but we do not yet understand how the predicted intensities match theoretical SWM process.

Future Work

The arguments from previous section all require populations to distinguish between SWM and two-photon lasing. One option is to look at the side fluorescence to see if any populations could be estimated. We also note that if an inversion can be proven on the blue, the D₁, or the D₂, then it is likely that an inversion will also exist on the two-photon process.

Another experiment of interest is to use two pulsed dye pumps to non-degenerately pump/seed the same nonlinear processes in cesium. Cesium's non-degenerate two-

photon cross sections appear to have an advantage over the other alkali vapors.

Finally we propose a novel two-photon laser that operates independently of any multi wave mixing mechanisms. Above we show that the strong mixing can create a two-photon inversion. We propose to use two pump sources to non-degenerately pump the cesium $6^2D_{5/2}$ state and then use 5 to 10 Torr of He buffer gas to spin orbit mix to the $6^2D_{3/2}$ state. Then, one or two other seed/probe pumps, possibly combined with one of the other pumps, will check for two-photon gain from the $6^2D_{3/2}$ state. We note that this gain must be extremely large to coexist with the $6D \rightarrow 7P$ lasing, but we have shown above that the D_1 and D_2 lines exhibit delayed turn on behavior and because these lines are trapped this should bottleneck many of the competing one-photon processes. Cavities may also be required to improve the gain on linear/nonlinear processes favorable to two-photon lasing.

5.6 Chapter Summary

The cesium 6D states provide an excellent platform to study MWM and multi photon effects in the alkali vapors. Observations suggest that two-photon pumping with diodes maybe possible, and theory suggests that out of Rb and Cs, Cs has the better non-degenerate two-photon cross sections due to its larger upper A coefficients.

Significant enhancement to some one-photon lasing lines, and 2 tunable two-photon lines are seen when pumping through the entire 6D states. The tuning behavior limits these two-photon lines to a two-photon lasing process, or a non-axial SWM process. We are not aware of another process capable of creating these two-photon lines with the the currently achieved powers and efficiencies. Much of the lasing from the two-photon pumped 6D states is cascade lasing. These fields fulfill the requirements for pumping non-axial FWM (e.g. 459 nm) and SWM processes (852 nm, 894 nm, 1381 nm two photon, and 3120 nm two-photon). However, the

intensity scaling of these states does not initially appear consistent with a multi wave mixing processes, especially the D_1 and D_2 lines.

The existence of a backward two-photon lasing field, and the ability to suppress lasing from the D_1 and D_2 lines suggest favorable conditions for two-photon lasing, but do not prove its existence. However, we note that Cs can theoretically enable a two-photon inversion, and our nonlinear lines are observed at significantly lower intensities than prior work. The large intensities and simple resolvability of the reported 2-photon lines, make this system an excellence candidate for further nonlinear/ quantum optic studies. We also present a two-photon laser concept that is distinguishable from wave mixing effects.

VI. Continuous-Wave 3 μm Cesium Laser on the $7^2P_{1/2} \rightarrow 7^2S_{1/2}$ Transition from a Two-Photon Pump

In this chapter we further justify our claim of low-threshold CW lasing from Chapters IV and V with a fully CW two-photon pumped demonstration. A CW laser on the 3.096 μm $7^2P_{1/2} \rightarrow 7^2S_{1/2}$ line is generated by two-photon pumping the cesium $6^2D_{3/2}$ state. The apparatus consists of a heat pipe at 150-270 $^{\circ}\text{C}$, pumped by a focused, single pass, 1.6 W Ti-sapphire laser. Lasing at 3 μm is developed from ASE with no cavity. This provides a peak corrected power of 750 μW , absorption corrected efficiency of 0.7%, and lasing thresholds $<10 \text{ kW}/\text{cm}^2$. We demonstrates scaling to $5\times$ lasing threshold, and efficiency can be improved with a resonator and non-degenerate pumping. Results suggest a two-photon diode pumped alkali laser is feasible.

6.1 Background

Recent kW-class demonstrations in Diode Pumped Alkali Lasers (DPALs) [75, 6], have motivated the search for additional or agile DPAL wavelengths [73]. In this work we limit our consideration to wavelengths near 3 μm , which have applications for infrared countermeasures [4]. Production of these IR wavelengths in the alkali vapors comes from three primary methods: single photon pump, two-sequential pumps, or a two-photon pump.

The single photon pump method was suggested in 1958 [83], realized experimentally in 1962 [78], and attempted as recently as 2015 [64]. Other notable examples include [88]. CW performance with the aid of resonators has been limited to 50 μW [78], and pulse demos have achieved slope efficiencies of 1.7×10^{-4} [64]. Another limitation of this approach is the low maturity and efficiency of blue/UV pump diodes. The primary focus of using sequential pumps has been production of the blue

wavelengths via four wave mixing (one potential infrared line connects the blue and two pump lines) [112, 113, 114, 63, 84, 86, 101, 103, 1]. Sequential pumping maybe capable of efficient generation near 3 μm , however we are currently unaware of any power and efficiencies estimates. This method also requires population to be left in an intermediary state, which is not required for two-photon pumping at high intensities.

The method we explore experimentally in this chapter utilizes two-photon pumping. Previous two-photon pumping demonstrations include [46, 45, 97, 34, 35, 44] and were all pulsed dye laser pumped. Many of these publication focus on production of blue wavelengths via four wave mixing. However, recent demonstrations [97, 44] have also focused on cascade lasing generated by Amplified Spontaneous Emission (ASE). In [44] (Chapters IV and V) we demonstrate quasi CW behavior of the 3 μm line from a pulsed dye laser source, with low thresholds $\sim 11 \text{ kW/cm}^2$ and modest slope efficiencies $\sim 1\%$. These results suggested favorable conditions for a true CW experiment.

6.2 Experiment

A schematic of our experimental apparatus is given in Figure 40, and the relevant energy levels are illustrated in Figure 41. The pump path shown in Figure 40 starts with an 18W Verdi laser that pumps a MBR 110E Ti-sapphire laser. This produces $\sim 2.5 \text{ W}$ near the 8 hyperfine two-photon pump transitions $6^2S_{1/2} \rightarrow 6^2D_{3/2}$. Ground state hyperfine splitting is resolved, yielding two sets of 4 pump transitions centered near 885.3990 nm and 885.3870 nm. The pump bandwidth is $< 50 \text{ kHz}$. A small part of the pump beam (not shown in Figure 40) is picked off to a High Finesse WSU wave meter to monitor the laser frequency. The beam is expanded 5x and then propagated a few meters to a separate optics bench. Laser power is controlled with a half wave plate and polarized beam splitter. Laser power is measured with a Coherent LM3

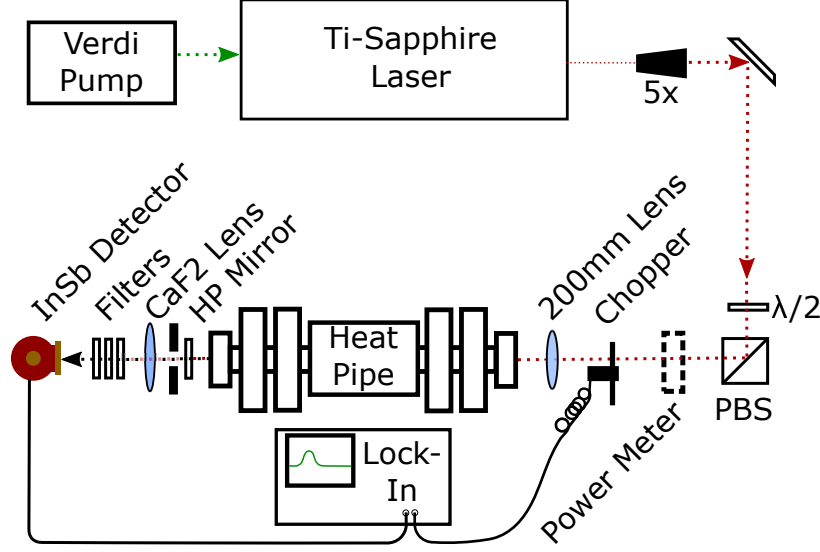


Figure 40. Experiment apparatus

sensor attached to a LabMax-TO meter. The beam is then amplitude modulated by a chopper wheel set near 500 Hz. To achieve intensities needed, the pump is focused by a 200 mm lens for some experiments and a 150 mm lens for others. We estimate diffraction limited focused spot diameters of $59 \mu\text{m}$ for the 150 mm lens ($\sim 66 \text{ kW}/\text{cm}^2$) and $79 \mu\text{m}$ for the 200 mm lens ($\sim 36 \text{ kW}/\text{cm}^2$). The lens is positioned to focus the pump near the center of the heat pipe.

The cesium heat pipe consists of a stainless steel tube, 29 cm long, and 2.5 cm in diameter. The heat pipe is vacuumed out at room temperature and 0 or 5 Torr of He is added before the cell is sealed off for the experiment. Pressure was monitored with a MKS 10 Torr Baratron attached to a MKS 670 controller. Seven temperature zones were used. Both windows were kept at 200°C with Watlow Ez-zone controllers and heat wraps. Four intermediary zones were created with flowing heated water from RTE-111 chillers through and aluminum blocks. The chillers kept the blocks at 38°C (outer zones) and 48°C (inner zones). The central oven zone was made with a 15 cm aluminum block and heated with a Watlow Ez-zone heater from 150°C to 270°C .

After the heat pipe, three configurations were used to acquire data. The first

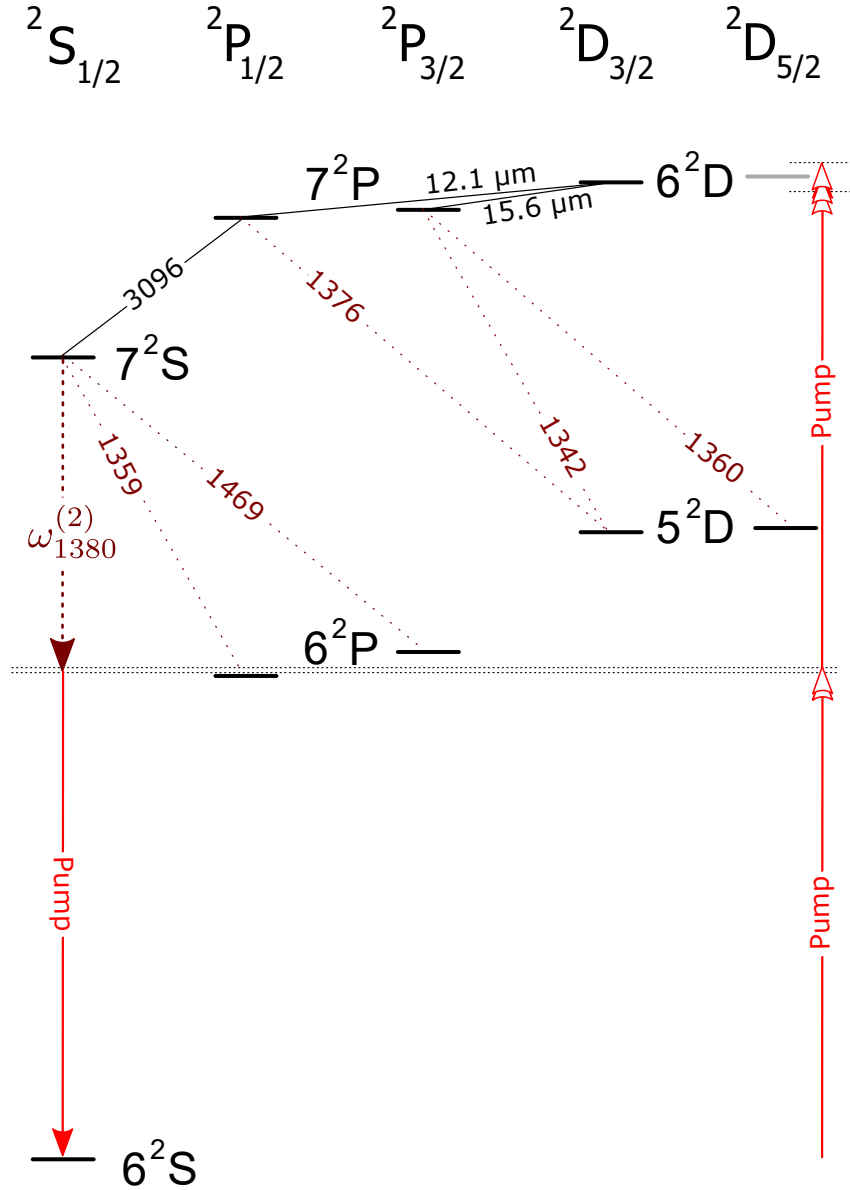


Figure 41. Energy diagram shows the primary lasing path (solid lines), and fluorescence (dotted lines). In this work we two-photon pump the $6^2D_{3/2}$ hyperfine states. The pump frequency ω_P is tuned between the degenerate two-photon pump processes described by $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ where $\omega_P + \omega_P = \omega_{6D_{3/2}}$. We also note a nonlinear two-photon emission processes described by $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ where $\omega_{1380}^{(2)} + \omega_P = \omega_{7S_{1/2}}$. Wavelengths are in nm except for the $6D \rightarrow 7P$ transitions which are in microns.

configuration, shown in Figure 40, was used to measure laser power. In this setup, the pump, the 3 μm beam and fluorescence beams exit the heat pipe. The expanding pump beam hits a high power mirror on a fused silica substrate. This mirror reflects $>99.5\%$ of the pump and transmits $>70\%$ of the 3 μm beam. The beam is then focused by a CaF_2 lens, the focused beam passes through a ND 1 filter, a ND 0.5 filter and a 3 to 4 μm broad band pass filter. This final filter removed any residual pump and the fluorescence from the other transitions. The focused beam was incident on a J10D-M204-R02M-60 cryocooled InSb detector, and processed by a Stanford Research System SR850 lock-in amplifier. The system was calibrated with an attenuated 10 mW 3.77 μm quantum cascade laser from Daylight Solutions and adjusted based on factory calibration from 3.77 to 3.096 μm .

The second configuration (not shown) measured the fluorescence of the transitions adjacent to the 3 μm line. In this setup, the output beams were re-collimated by a 150 mm lens followed by a second PBS cube to divert the remaining pump, a 1100 nm long pass filter, and a fiber coupler to a McPherson 218 spectrometer equipped with a cryocooled Roper Scientific 1x1024 InGaAs array. This system was calibrated with an attenuated 8 mW 1.33 μm diode laser. Further details can be found in [44] (Chapter IV).

The third configuration (not shown) measured absorption of the two-photon pump. In this setup, the residual pump beam was re-collimated by a 150 mm lens and a small portion of this beam was sent to a Thorlabs PDA100A amplified Si detector. The output of this detector was sent to the lock-in amplifier.

Transmission of the filters and other key optics in the apparatus were measured with a Cary 5000 Spectrophotometer (200 nm to 3000 nm), and a Bomem MB 155S FT-IR spectrometer (>1000 nm). Our recent pulsed experiments [44] (Chapters IV and V) took place in the same heat pipe. Optimal conditions for 1.3 μm lasing were

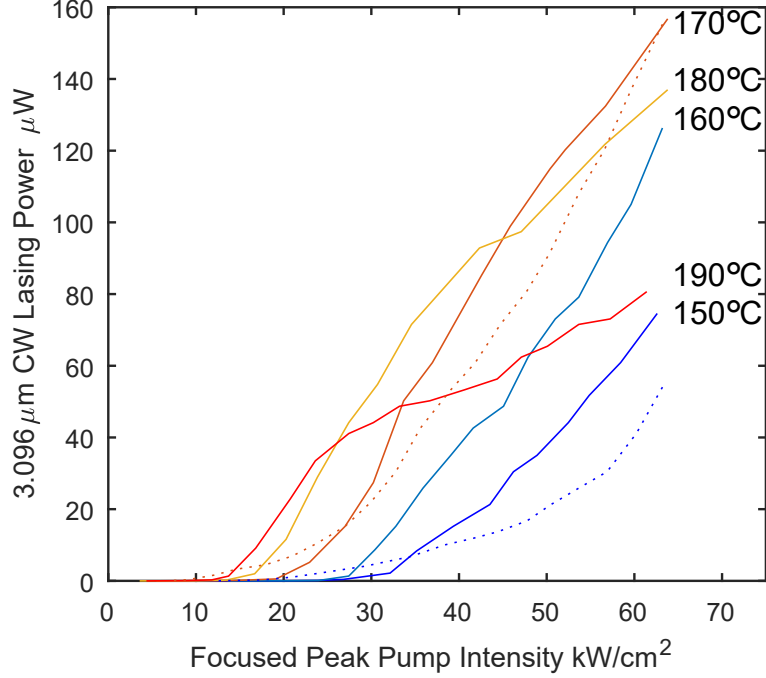


Figure 42. Laser intensity scaling of the $3.096 \mu\text{m } 7^2P_{1/2} \rightarrow 7^2S_{1/2}$ line with 5 Torr He buffer gas. Solid lines show data from 150 mm focusing lens. Dotted lines show a comparison with vacuum data (at 150 °C and 170 °C see Figure 43). Lines are formed by ~ 20 evenly spaced data points for a given temperature.

observed at 237.5 °C, and 5 Torr of He buffer gas.

6.3 Results and Discussion

We first present results for 5 Torr of He buffer gas with the 150 mm focusing lens in Figure 42. The raw measurements are corrected based on calibrations and transmission of known optics to estimate the forward and backward lasing power in the cell. We also assume the backward intensity is 20% of the forward intensity based on previous measurements from our pulsed work [44]. We were not able to repeat the backwards measurements for the present work so they should be at least 20% and are likely higher. Cesium densities based on the vapor pressure curves range from $2.2 \times 10^{14}/\text{cm}^3$ at 150 °C and $1.4 \times 10^{15}/\text{cm}^3$ at 190 °C.

Performance declines for Cs melt pool temperatures exceeding 170 °C. This is not consistent with our recent pulsed results [44], where the 3 μm lines which continue to improve beyond 240 °C. At 170 °C, the slope efficiency begins to decrease 33 kW/cm². For 180 °C this roll is difficult to discern, but occurs slightly earlier, ~ 25 kW/cm². For 190 °C we see the roll over knee is at 22 kW/cm².

There are two likely explanations for this roll over and both involve increase divergence of the 3 μm beam. The first is that as the gain/ unit length increases, there is a less preferential lasing direction. The second mechanism, proposed by Moran [64], is that at higher cesium densities, the length over-which bleaching occurs (an effective gain length) becomes smaller. As this length decreases, there is a less preferential lasing direction and the divergence increases. We can correlate this to the number of atoms becoming similar to the number of pump photon pairs that the system can cycle. The number of pump photon pairs that the system can cycle is the pump power at rollover times the rate limiting step. For our case this is the radiative lifetime of the 7S state (48 ns) [99] plus the 6P state lifetime (30 ns) [99]; from these lifetimes we estimate an 80 ns relaxation rate per cycle. As the cesium melt pool temperature increases from 170 °C $\rightarrow$ 180 °C $\rightarrow$ 190 °C, the number of atoms in the gain volume increase from $2.0 \times 10^{10} \rightarrow 2.6 \times 10^{10} \rightarrow 4.4 \times 10^{10}$. This occurs while the number of pump photon pairs that the system can cycle (from the above roll over estimates) decreases from $1.4 \times 10^{11} \rightarrow 1.1 \times 10^{11} \rightarrow 9.5 \times 10^{10}$. Overall these show good agreement with the explanations for increased divergence.

The data in Figure 42 clearly shows a decrease in threshold with an increase in cesium density or oven temperature. This trend is also observed in recent pulsed work [97, 44]. In [44] (Chapter IV) we present a simplified analytical model that shows the threshold intensity $I_t \propto 1/\sqrt{n_{Cs}}$ where n_{Cs} is the cesium density.

In Figure 42 we also include results without He. The comparison shows a decrease

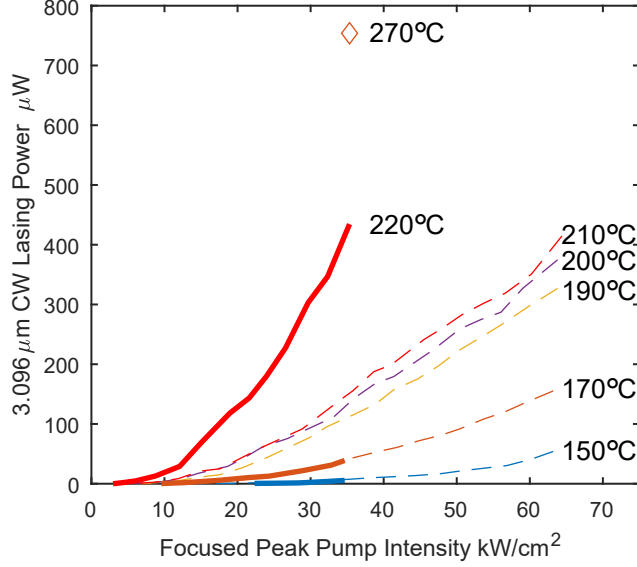


Figure 43. Laser intensity scaling of the $3.096 \mu\text{m } 7^2P_{1/2} \rightarrow 7^2S_{1/2}$ line without buffer gas. Plot shows lasing power vs the focused peak pump intensity for various heat pipe temperatures (Cs densities). The upper data point $\diamond$ shows the conditions of best performance (270°C with 200 mm lens). Thick lines show data from 200 mm focusing lens. Dashed lines show data from 150 mm lens.

in lasing threshold but also a decrease in performance for the 150 and 170 $^\circ\text{C}$ cases. The performance increase at 5 Torr is consistent with our pulsed work. We believe this mechanism involves the ground state hyperfine mixing [44]. However, it is unclear why the slopes are different, and the upper 6D fine structure may play also an important role. The 0 Torr thresholds are also less defined, therefore it is possible that when the system is lasing under more auspicious conditions the thresholds may appear to be larger. This trend of less defined and potentially decreased thresholds is shown in Figure 43, where we plot the same cases for 150 to 210 $^\circ\text{C}$ at 0 Torr He. For this case the lasing power continues to scale up to 210 $^\circ\text{C}$ (dashed lines). When we switch to a weaker focus lens of 200 mm (thick lines) we continue to see improvement in laser power up to 270 $^\circ\text{C}$. We attribute this improvement to a large gain volume and radius. This volume contains more atoms, but more importantly the larger dimensions and

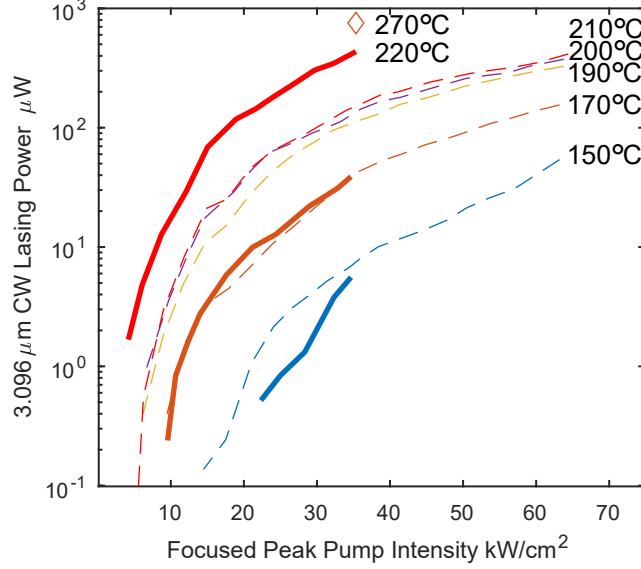


Figure 44. Log scale of output power of Figure 43.

may help to minimize divergence losses.

At lower temperatures the two different lenses show good agreement, and confirms that our focused spot size estimates are fairly accurate. We refine this with a log scale plot in Figure 44. These results also confirm a decrease in threshold with increased cesium density. Figures 42 and 43 both show thresholds in good agreement with our pulsed threshold of 11 kW/cm² (237.5 °C). Our best low threshold estimate is ~ 10 kW/cm² for the 220 °C vacuum case. Therefore our optimal power measurement at 270 °C will have a trend and threshold < 10 kW/cm². This makes diode pumping a strong possibility as modern DPAL pumps have > 30 kW/cm².

Our best performance of 750 μ W in vacuum, at 270 °C, and 200 mm lens, is also shown in Figure 43. For these conditions, the fraction of the pump power absorbed is modest, $< 7\%$, and limits laser efficiency. Therefore we estimate the absorption corrected optical to optical efficiency to be 0.7%. These results are consistent with our pulsed slope efficiency of $\sim 1\%$. We plan to examine the absorption data with a thorough discussion of the eight two-photon hyperfine absorption lines in a subsequent

study. We did not see further improvement beyond 270 °C which indicates a possible roll over. The roll over we see at 170 °C with 5 Torr He, was also reported by Sharma at 150 °C when pulse pumping with a blue source [88]. However, their work was at vacuum we do not see this roll over in vacuum until ~ 270 °C with the 200 mm focusing lens, or ~ 210 °C with the 150 mm lens. We repeat the calculation of number of atoms to pump photons. For the 150 mm lens and 210 °C case we estimate 8×10^{10} atoms and 3×10^{11} pump photon pairs that the system can cycle. For the 200 mm lens and 270 °C case, we estimate 9×10^{11} atoms and 3×10^{11} pump photon pairs that the system can cycle. These observations also support the roll over due to increased divergence hypothesis, as opposed to other explanations such as energy pooling suggested by Sharma [88]. We also note that a roll over near ~ 270 °C is consistent with the trends from our pulse work where we see the D_1 and D_2 lines roll over near 190 °C, the blue lines near 200 °C, and the $1.3 \mu\text{m}$ lines near 240 °C.

The output power is directional, and considerably larger than could be described by spontaneous emission. The 750 μW bi-directional lasing power, which corresponds to a 625 μW forward power, is constrained by a 1.7 cm diameter aperture located 25 cm from the center of the cell. If this power were purely fluorescence there would be >2 W of fluoresced power, which is greater than our pump input power. The fraction of the cesium density which can be transferred under fully bleached conditions is 1/4 the total density. We estimate this 1/4 from by assuming a best case bleached pump, and bleached 12 μm line. Under these conditions, we have n_0 in the $6^2S_{1/2}$ state, $2n_0$ in the $6^2D_{3/2}$ state, and n_0 in the $7^2P_{1/2}$ state. The maximum number of atoms in the $7^2P_{1/2}$ state is 1/4 of the 9.7×10^{11} atoms in the gain volume. With 2 W of fluoresced power we have 3×10^{19} 3 μm photons/s, The corresponding fluorescence rate for the $7^2P_{1/2} \rightarrow 7^2S_{1/2}$ transition would be $> 1 \times 10^8/\text{s}$, orders of magnitude larger than the $3.5 \times 10^6/\text{s}$ reported value [48]. When we repeat this process for our

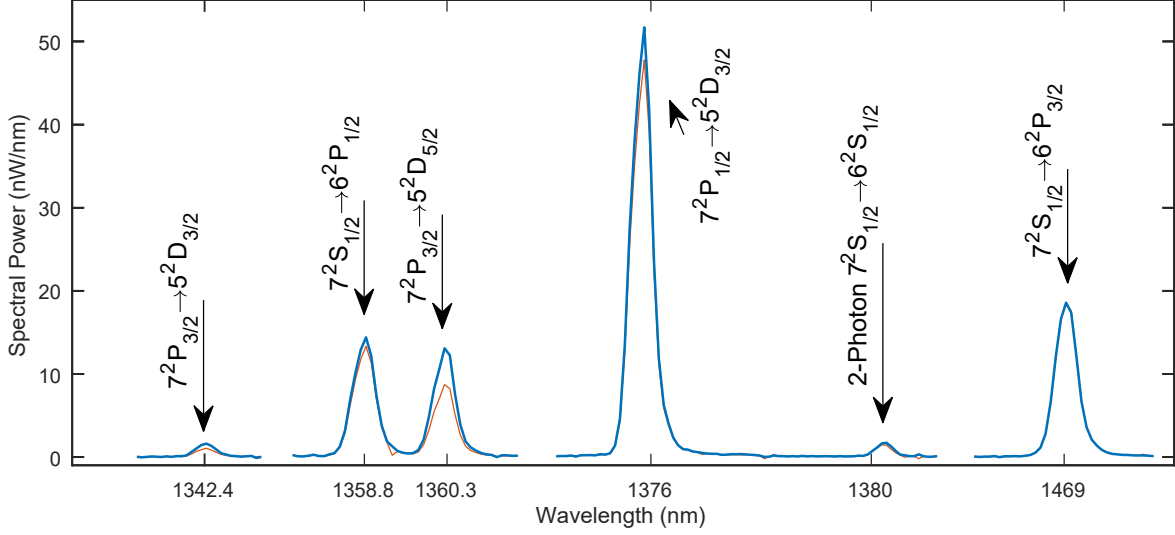


Figure 45. On-resonance fluorescence spectra of transitions circumscribing the $7^2P_{1/2} \rightarrow 7^2S_{1/2}$ lasing line. Data taken at 220 °C with no buffer gas. Blue line shows 885.3990 nm, orange line shows 885.3870 nm.

150 °C case, we still get an A coefficient requirement $> 4 \times 10^7$.

The fluoresce spectra during lasing on the 3096 nm $7^2P_{1/2} \rightarrow 7^2S_{1/2}$ transition are shown in Figure 45. These transitions are also shown in the energy diagram of Figure 41. From our experience with the pulsed version of this laser, we know that once the 12 μm $6^2D_{3/2} \rightarrow 7^2P_{1/2}$ lases strong enough, the $6^2D_{3/2} \rightarrow 7^2P_{3/2}$ transition will also lase, followed by lasing on the 2931 nm $7^2P_{3/2} \rightarrow 7^2S_{1/2}$. We use the fluorescence data from Figure 45 and A coefficients from [48] to compare the inversion on the 3096 nm line with the inversion on the 2931 nm line. These inversions suggest that at maximum pump power (65 kW/cm²) and 220 °C heat pipe temperature, <25% of the power is in the 2931 nm line and the remaining >75% is in the 3096 nm line. This is consistent with our pulsed data, where spectra show ~24% in the 2931 nm line at pump intensities near 500 kW/cm². Therefore, both of these lines could be useful as a 3 μm DPAL.

Other Considerations for a 3 μm DPAL

The two-photon pumped DPAL has some potential advantages and disadvantages relative to the high power, near infrared DPAL system. The obvious disadvantage is the total efficiency. Quantum efficiency limits performance 14% optical to optical efficiency (ratio of the 3 μm energy to the two pump photon energy). We have only shown $\sim 1\%$. However, there is significant opportunity for improvement with the addition of a cavity, non-degenerate pumping, and a more intense pump source (that does not require intense focusing) to achieve better mode volume overlap. High power laser diode bandwidths will also need to be minimized to operate this system near vacuum.

The advantages of the two-photon pump approach include wavelength agility (new DPAL missions), near diffraction limited beam quality (near vacuum pressures), and simplified circulator (heat is released optically, not into the gas). These all point to potential power scaling. Other mid IR laser technologies such as Quantum Cascade Lasers (QCL) maybe currently more efficient for mid IR sources. If we were to use 55% electrical-optical (E-O) efficient diode pumps and achieve the quantum efficiency limit, the best this system will do is 8% E-O efficiency. For comparison, QCL have demonstrated CW E-O efficiencies of 21%, with ~ 4 W of power, at 4.9 μm [3]. However, QCLs maybe limited in power scaling, and beam quality. The narrow A path to very high mid IR CW powers might be enabled by the two photon pumping scheme. Also, the threshold is low enough to utilize current diode technology, which provides a rapid path to realization.

The ideal choice of alkali is pump scheme dependent. If we are constrained to a degenerate two-photon pump, rubidium and cesium are the primary options. Rubidium has a clear advantage as the $5^2S_{1/2} \rightarrow 5^2D_{3/2}$ transition has a virtual state only 30 cm^{-1} off resonance. This gives a 2x better cross section than the cesium transi-

tion. However, non-degenerate pumping provides many additional options. Based on our knowledge of the A coefficients we speculate the following options to have merit. In sodium the best method is to two-photon pump $3^2S_{1/2} \rightarrow 5^2S_{1/2}$ and lase 3415 nm on $5^2S_{1/2} \rightarrow 4^2P_{3/2}$. In potassium a good scheme would be to pump $3^2S_{1/2} \rightarrow 6^2S_{1/2}$ and lase 3662 nm on $6^2S_{1/2} \rightarrow 5^2P_{3/2}$. In rubidium $nS \rightarrow (n+2)S$ and $S \rightarrow D$ pump options are available but cesium generally has larger A coefficients on the upper pump states. Finally, for cesium the best case is not actually the one present here but the $6^2S_{1/2} \rightarrow 6^2D_{5/2}$ and lase 2391 nm on $7^2P_{3/2} \rightarrow 7^2S_{1/2}$. Cesium also has a $nS \rightarrow (n+2)S$ path, but the two photon pump cross section is estimated to be 4x larger for the $S \rightarrow D$ pump since its also 20x larger than the degenerate cross-section employed in this work. However, the $nS \rightarrow (n+2)S$ scheme should possess lower pump thresholds because the lasing state is immediately inverted. Clearly there are many options that must be considered before committing to particular pump diodes. These of course must be weighted against the ability to create high-power narrow line-width pump diodes at the degenerate wavelengths, and the atmospheric transmissions of the various lines. Also the lighter alkali require shorter pump wavelengths that are more difficult to achieve and may lower the efficiency of both pump diodes and the theoretical limit of 3 μm laser photon to two pump photons. Our current assessment is that cesium has many advantages over the other alkali vapors but an improved knowledge of the upper Einstein A coefficients or atmospheric transmission could change this conclusion.

6.4 Chapter Summary

A CW 3 μm laser has been achieved by two-photon pumping with a 1 W pump source. Two-way laser power is estimated to be $>750 \mu\text{W}$, with a absorption corrected efficiency of 0.7%, and lasing threshold $<10 \text{ kW/cm}^2$. This can be signifi-

cantly improved with intense, non-degenerate, two-photon pumping and a resonator. Two-photon DPALs are an auspicious path to wavelength agile systems. The potential architecture does not require buffer gas and heat is released optically, which simplifies the requirements for a gas circulation system. Such a device maybe able to achieve 3 μm laser operation with greater output power and better beam quality compared to solid state alternatives.

VII. Continuous-Wave Nonlinear Effects Observed from a Two-Photon Pump

In this chapter we report our preliminary findings of the non-linear and off-resonant behavior from CW pumping the two-photon $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ transition. The first half of this chapter focuses on the hyperfine two-photon absorption theory, and measurements, as well as our $3\ \mu\text{m}$ emission spectra. We can also use these two pieces of information to gain a slight improvement of the NIST $6^2D_{3/2}$ energy level. The second half of the chapter is a first look at two non-linear features observed in the $1.3\ \mu\text{m}$ spectral window. One is the appearance of the 1381 nm two-photon line, and the second is a dramatic off-resonance enhancement to the 1359 nm line.

7.1 CW Hyperfine resolved absorption and emission spectra

Hyperfine Two-Photon Absorption Theory

Throughout this document, we reference the degenerate two-photon absorption, and in Appendix A we derive:

$$\begin{aligned} \sigma_{if}^{(2)}(\nu) = & \frac{9}{5} \frac{\nu}{256\pi^4 \hbar n^2 c^2} \sum_n \frac{g_f}{g_i} \frac{\lambda_{fn}^3 A_{fn} \lambda_{ni}^3 A_{ni}}{(\nu_{ni} - \nu)^2} \\ & \times \rho_f(\nu_{fi} = 2\nu), \end{aligned} \tag{39}$$

and for the non-degenerate case from Appendix B

$$\begin{aligned}
\sigma_{if}^{(2)(2field)}(\nu_1, \nu_2) &= \frac{9}{5} \frac{\nu}{256\pi^4 \hbar n^2 c^2} \sum_n \frac{g_f}{g_i} \lambda_{fn}^3 A_{fn} \lambda_{ni}^3 A_{ni} \\
&\times \frac{(\nu_1 + \nu_2 - 2\nu_{ni})^2}{(\nu_1 - \nu_{ni})^2 (\nu_2 - \nu_{ni})^2} \\
&\times \rho_f(\nu_{fi} = \nu_1 + \nu_2) .
\end{aligned} \tag{40}$$

We now seek to incorporate the hyperfine theory for the two-photon case. In Figure 46 we plot the energy levels of the hyperfine resolved $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ transition with data from [95, 32, 53]. We calculate the separation of these states based on the $F' = 4$ level to be 3.95 MHz from the NIST value of $22588.821 \text{ cm}^{-1}$ [32]. This offset calculation is made from the separations from [53] and balances the moments of the $2F+1$ degeneracies.

In the derivation of equations 39 and 40, we update the dipole selection rules to reflect the 2 quanta absorbed in the process. For the angular momentum we have $\Delta L = 0, 2$. For the hyperfine two-photon transitions [7] $\Delta F=0, 1, 2$ which makes all 8 transitions dipole allowed. For a further discussion on the relationships for the polarization of each individual photon and the magnetic quantum numbers see [7]. We now know we must sum over 8 transitions. We define the hyperfine resolved cross section as

$$\sigma_{if}^{(2HF)}(\nu) = \sum_{F \rightarrow F'} \sigma_{if}^{(2)}(\nu, 2\nu_{F \rightarrow F'}) \times f_F \times S_{F \rightarrow F'} \tag{41}$$

where we have used that convention that $\sigma_{if}^{(2)}(\nu, 2\nu_{F \rightarrow F'})$ contains a $\rho_f(\nu_{F \rightarrow F'} = 2\nu)$ (degenerate) or $\rho_f(\nu_{fi} = \nu_1 + \nu_2)$ (non-degenerate) line shapes. That is, each Gaussian, Lorentzian, or Voigt lineshape is centered at one of the 8 transitions. Next, f_F is the

statistical distribution of the ground state population F given by

$$f_F = \frac{(2F+1) e^{-E(F)/kT}}{\sum_F (2F+1) e^{-E(F)/kT}} \quad (42)$$

and $S_{F \rightarrow F'}$ is the two-photon hyperfine strength. Our discussion of the mirrors the one-photon analog found in Steck [95].

$$S_{F \rightarrow F'} = (2F' + 1) (2J + 1) \left\{ \begin{matrix} J & J' & 2 \\ F' & F & I \end{matrix} \right\}^2 \quad (43)$$

where the term in $\{ \}$ denotes the Wigner 6-j symbol. Note that $S_{F \rightarrow F'}$ has the requirement

$$\sum_{F'} S_{F \rightarrow F'} = 1. \quad (44)$$

In the Chapter 4 and Appendix A, we note a factor of $1/5$ that comes from the simultaneous average of two dipole moment operators. We can also use the theory from Steck[95] to verify this for the two-photon line strengths:

$$\sum_{F'} (2F' + 1) (2J + 1) \left\{ \begin{matrix} J & J' & 2 \\ F' & F & I \end{matrix} \right\}^2 |\langle F m_F | F' 2 m_F 0 \rangle|^2 = \frac{1}{5}, \quad (45)$$

where for the one-photon case

$$\sum_{F'} (2F' + 1) (2J + 1) \left\{ \begin{matrix} J & J' & 1 \\ F' & F & I \end{matrix} \right\}^2 |\langle F m_F | F' 1 m_F 0 \rangle|^2 = \frac{1}{3}. \quad (46)$$

We present the values for $S_{F \rightarrow F'}$ in Table 12, and verified that equation 44 and 45 hold.

As side note, we point out that this approach is different than Gallagher [36] and his source for $S_{F \rightarrow F'}$ [65] does not have the same meaning. They base the strength

Table 12. Cesium $S_{F \rightarrow F'}$ hyperfine line strengths (evaluation of equation 43). F denotes the ground state hyperfine level and F' denotes the $6^2D_{3/2}$ hyperfine level

	$F = 4$	$F = 3$
$F' = 2$	1/4	1/28
$F' = 3$	7/24	1/8
$F' = 4$	11/40	81/280
$F' = 5$	11/60	11/20

on an electric quadrupole transition stating “the two-photon transition operator is purely quadrupolar in nature.” In Appendix C we show these different definitions of $S_{F \rightarrow F'}$ are nearly equivalent.

We use the values from Table 12 and illustrate the impact of the line strengths in Figure 47 with Doppler broadening at 180°C. We can see that the lower frequency peak in Figure 47a (dark red two-photon line in Figure 46) is shifted $-30.6 \text{ MHz}/2$, where the factor of two is for the difference being shared by the two photons. The larger frequency peak in Figure 47b (blue two-photon line in Figure 46) is shifted $+39.17 \text{ MHz}/2$. We estimate the two sets of peaks to be separated by

$$\Delta\nu_{2p} = \frac{\Delta\nu_{GS}}{2} - \frac{\Delta\nu_{4 \rightarrow 2', 3', 4', 5'}}{2} + \frac{\Delta\nu_{3 \rightarrow 2', 3', 4', 5'}}{2}. \quad (47)$$

Two-Photon Absorption Measurements

The expected absorption behavior for the two-photon process in the low intensity limit is given by a modified bears law

$$-\alpha^{(2)} = \ln \frac{I}{I_0} = -\sigma_{if}^{(2HF)}(\nu) \times I_0 \times n_{Cs} \times L_g \quad (48)$$

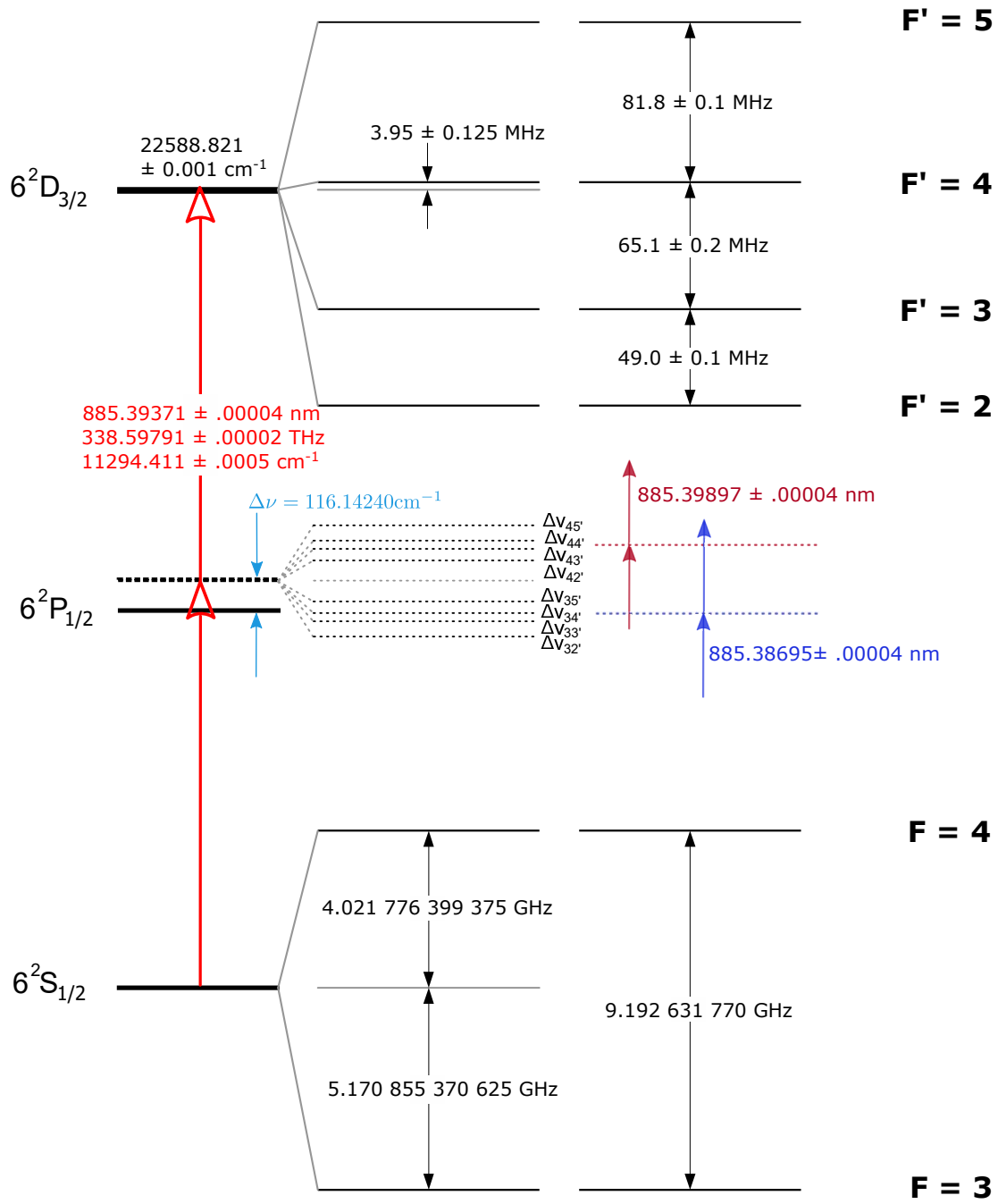


Figure 46. Energy diagram of the two-photon hyperfine energy levels on the eight $6^2S_{1/2} \rightarrow 6^2D_{3/2}$ transitions. Compiled with data from [95, 32, 53]

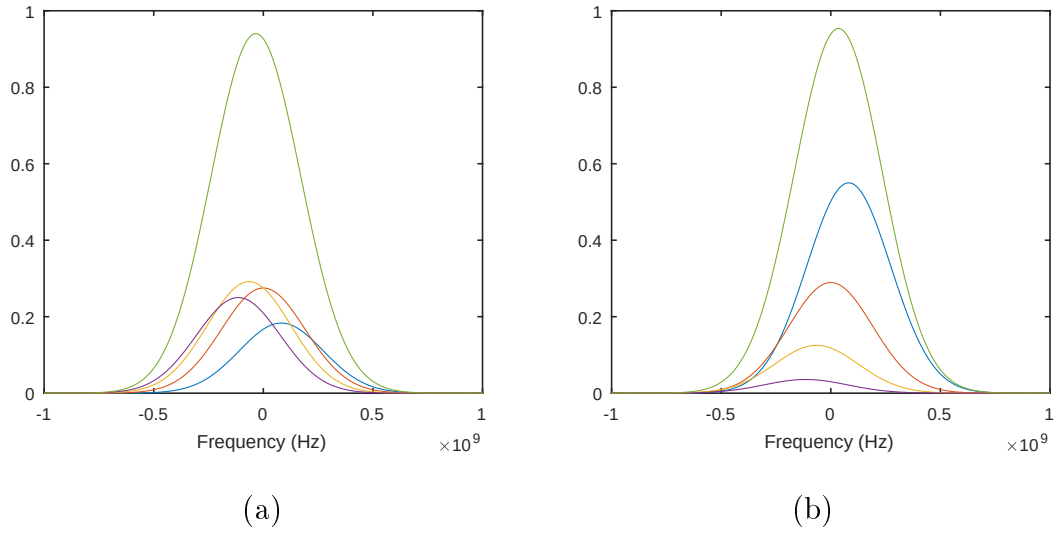


Figure 47. Simulations of of the eight Doppler broadened hyperfine line strengths. The ground state hyperfine splitting separates the eight transitions separate into two groups of 4 transitions as shown in Figure 46. The green line shows the sum of the four underlying hyperfine transitions and represents the expected Doppler broadened measurement. (a) The $\nu_4 \rightarrow \nu'_{2',3',4',5'}$ exhibits a 30.6 MHz red shift. (b) The $\nu_3 \rightarrow \nu'_{2',3',4',5'}$ exhibits a 39.17 MHz blue shift.

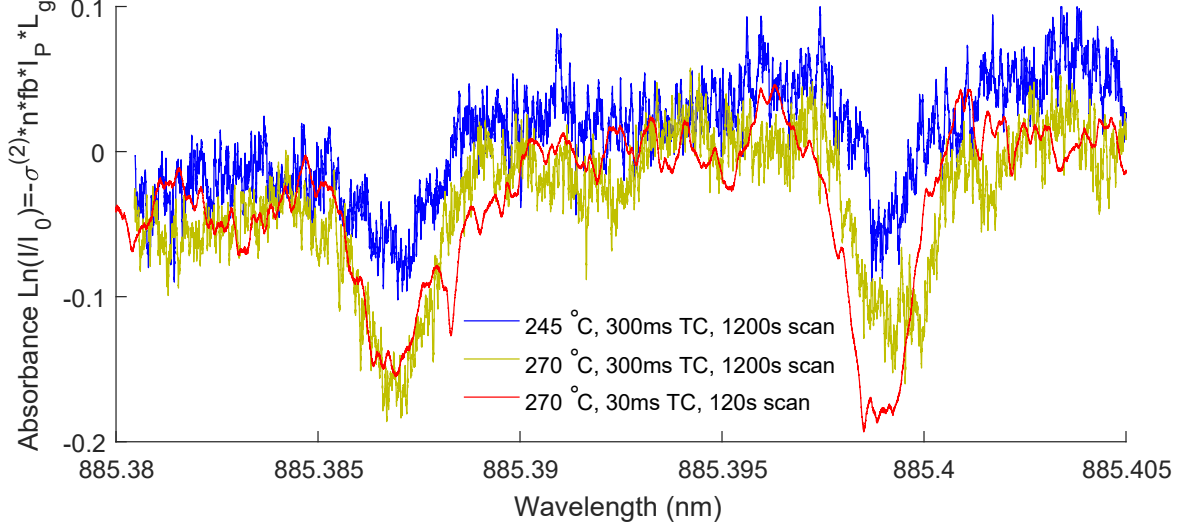


Figure 48. Two-photon absorption with resolved ground state hyperfine splitting. Experimental conditions: 245 °C ($8.4 \times 10^{15}/\text{cm}^3$) and 270 °C ($1.7 \times 10^{16}/\text{cm}^3$), 1.6 W, 200 mm lens, 5 Torr He. Note the absorbance doubles when we double the density.

where I is the transmitted pump intensity, I_0 is the initial pump intensity, n_{Cs} is the cesium density, and L_g is the gain length. This behavior is seen in Figure 48 where we note that the absorbance doubles when we double the density. We see good agreement with our two-photon cross section theory. From equation 41 and the values in Table 8 $\alpha^{(2)} \approx .2$ with $I_0 = 35 \text{ kW}/\text{cm}^2$, and $n_{Cs} \approx n_{Cs:T}/2 = 1.7 \times 10^{16}/\text{cm}^3/2$ from f_F (equation 42), we use the Rayleigh range for $L_g \approx 5 \text{ mm}$. This is a very good agreement considering the uncertainty in I_0 (uncertainty in focus spot size), $n_{Cs:T}$ (uncertainty in cell temperature), L_g (related to focus spot size), and $\sigma_{if}^{(2HF)}$ (Upper A coefficients not known accurately). These spectra were taken with 5 Torr He; but our attempts to measure the absorbance for 0 Torr He were unsuccessful due to strong noise sources that were interrelated to the heat-pipe. The level of this noise puts an upper bound on the absorbance of the 0 Torr He case of around $\sim 7\%$.

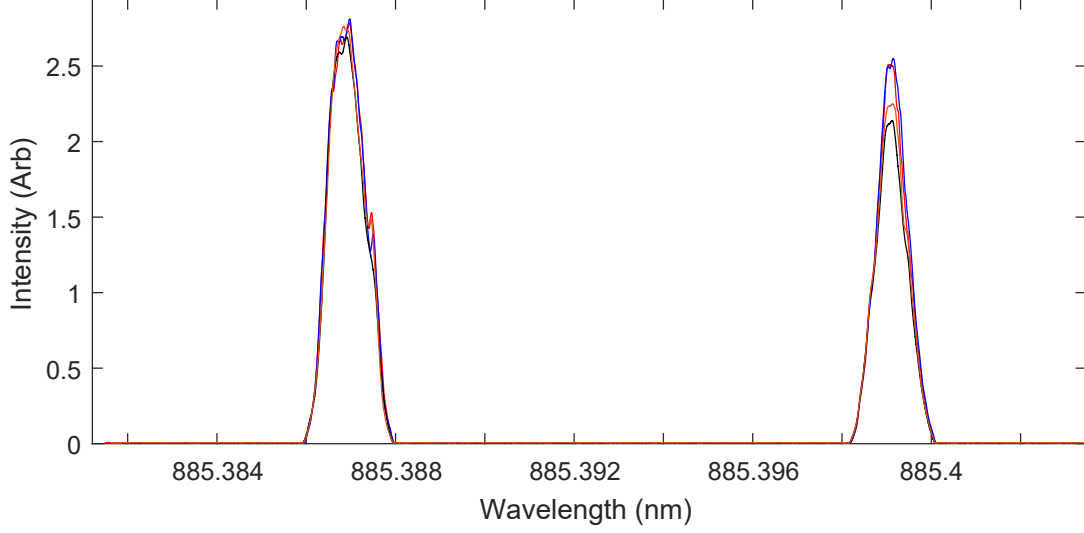


Figure 49. 3096 nm lasing spectra. Four spectra under identical conditions to illustrate the repeatability of the measurement. Experimental conditions: 180 °C, 1.5 W pump, 200 mm lens, vacuum.

Lasing Spectra

Next we cover our 3096 nm lasing spectra from two-photon pumping. In Figure 49, we show the data for 4 cases of different scan times and lock-in time constants. The shorter wavelength is expected to be less energetic from the grounds state hyperfine distribution f_F (equation 42). However, the lower wavelength has the largest peak for the vacuum case, but this difference is small. Conversely, the 5 Torr He absorption spectra in Figure 48 is consistent with equation 42.

Update to Cesium $6^2D_{3/2}$ Energy Level

The current NIST energy level for the cesium $6^2D_{3/2}$ energy level, $22588.821 \pm .001 \text{ cm}^{-1} (\pm 30 \text{ MHz})$ was taken in 1965 [32]. We realized that despite our noise sources and Doppler broadening, we could use information from equation 47 to attempt to estimate the hyperfine resolved two-photon wavelengths, and by extension the $6^2D_{3/2}$ energy level. Our primary tool for this is our High Finesse WSU wavemeter with ± 2

MHz of accuracy. However, this accuracy is based on its calibration source (ours is a 632 nm HeNe with ± 2 MHz of accuracy) but at 885.4 nm we are prone to some offset error. To improve the accuracy, we leverage the nearby D_1 line at 894 nm and its 4 hyperfine resolved transitions. The cesium ground state hyperfine splitting is of the best experimentally determined quantities in all of physics.[95] .

We present one of four D_1 absorption measurements in Figure 50. Due to the long path lengths and large-one-photon cross sections, these measurements were well resolved in comparison to the two-photon absorption measurements in Figure 48. We also have simple tools to accurately produce theoretical predictions of the hyperfine resolved one-photon cross sections which we also plot in Figure 50. We compare to theory only to get line centers, exact duplication of theory generally required some trial and error to account for various effects. The height of the lines is related to the number density should could be reduced by localized heating. The heat pipe was operated in a cold mode for this experiment with the oven off. But the windows were still $\sim 180^\circ\text{C}$ and had nearly 18 months of exposure to alkali. Therefore the absorption volume temperature, path length, and axial cesium density are difficult to determine. A static cell would be more ideal for this measurement. The large broadening is likely due to heating of the gas from the pump, window heaters, and trace alkali contaminates on the windows.

We estimates the centroids from the upper 30 to 40% of each peak to insure that the pedestal or adjacent lines are not influencing our measurement. To calculate the centroids we use

$$\bar{x} = \frac{\int_b^a x f(x) dx}{\int_b^a f(x) dx} . \quad (49)$$

Our four data sets gave us 16 error estimates the we average as: $\Delta_{WM} = 47 \pm 4$ MHz. This is the larges source of error. Other errors are $\sim .1$ MHz so we don't track them.

Next we use our lasing spectra in Figure 49 to estimate the two-photon transitions.

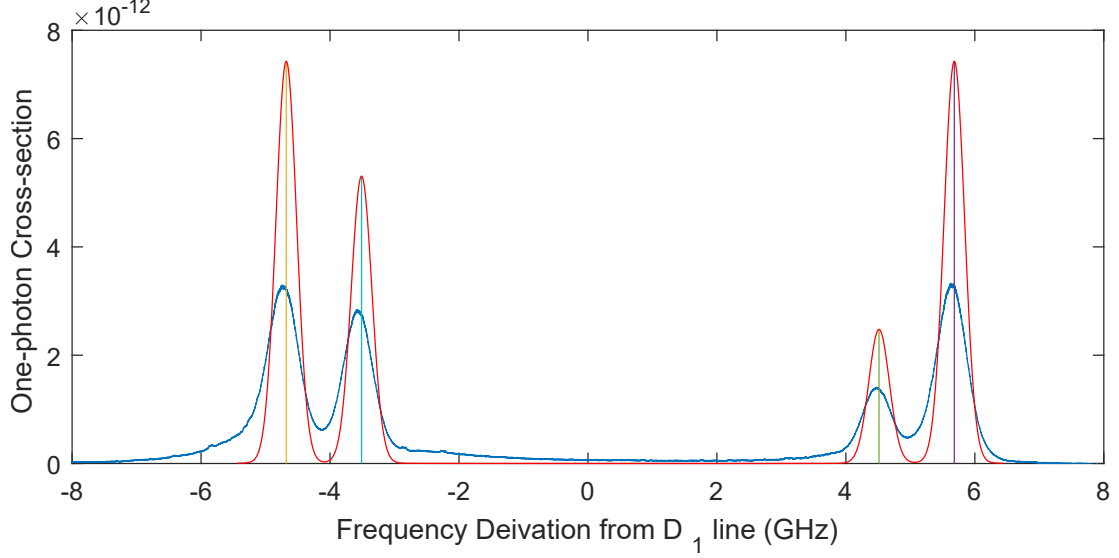


Figure 50. One-photon cross section based on absorption measurement of the D_1 line to estimate wave meter offset. Blue line shows measured data. Red line and vertical lines show theoretical cross-section and line centers. Experimental conditions: 25°C, 0 Torr He.

We obtain raw frequency measurements of

$$f_{R3} = f_{3 \rightarrow 2', 3', 4', 5'}^R + f_{3Error} = 338.600498 \text{ GHz} \quad (50)$$

$$f_{R4} = f_{4 \rightarrow 2', 3', 4', 5'}^R + f_{4Error} = 338.595840 \text{ GHz} \quad (51)$$

where f_{3Error} and f_{4Error} denote some experiment offset error constrained by the ground state hyperfine splitting. Recall from equation 47

$$\begin{aligned} \Delta\nu_{2p} &= \frac{\Delta\nu_{GS}}{2} - \frac{\Delta\nu_{4 \rightarrow 2', 3', 4', 5'}}{2} + \frac{\Delta\nu_{3 \rightarrow 2', 3', 4', 5'}}{2} \\ &= \frac{9.192631770 \text{ GHz}}{2} + \frac{30.6 \text{ MHz}}{2} + \frac{39.2 \text{ MHz}}{2} \end{aligned} \quad (52)$$

where the constraint on the data is $f_{R4} - f_{R3} = \Delta\nu_{2p}$ or

$$27.0 \text{ MHz} = f_{3Error} + f_{4Error}. \quad (53)$$

Next we correct our raw frequencies for the wave meter off set and hyperfine offset, and ground state splitting to estimate the $6^2D_{3/2}$ energy level. Our final system of three equations to solve is

$$f_{6D_{3/2}} = 2(f_{3 \rightarrow 2', 3', 4', 5'}^R + f_{3Error}) + 2\Delta_{WM} - \Delta\nu_{3 \rightarrow 2', 3', 4', 5'} - 5.170855370625 \text{GHz} \quad (54)$$

$$f_{6D_{3/2}} = 2(f_{4 \rightarrow 2', 3', 4', 5'}^R + f_{4Error}) + 2\Delta_{WM} + \Delta\nu_{4 \rightarrow 2', 3', 4', 5'} + 4.021776399375 \text{GHz} \quad (55)$$

$$27.0 \text{ MHz} = f_{3Error} + f_{4Error} . \quad (56)$$

We solve for $f_{6D_{3/2}}$ and convert to wave numbers and obtain

$$\text{Proposed: } f_{6D_{3/2}} = 22588.8203 \pm .0003 \text{ cm}^{-1} \quad (57)$$

$$\text{Current: } f_{6D_{3/2}} = 22588.821 \pm .001 \text{ cm}^{-1} \quad (58)$$

This approach validates our experimental capabilities, and could be used to update the current NIST value by nearly an order of magnitude better accuracy. The source of our final error $\pm .0003 \text{ cm}^{-1}$ (or $\pm 8 \text{ MHz}$) is that our wave meter error is doubled when converting from the two-photon wavelength to the $6^2D_{3/2}$ energy level.

7.2 Nonlinear Effects

In this section we cover two nonlinear effects. We first cover a dramatic enhancement seen on the $1359 \text{ nm } 7^2S_{1/2} \rightarrow 6^2P_{1/2}$ florescence data when tuning off-resonance. The second observation we report on is the CW appearance of the 1381 nm two-photon line.

Off-Resonant Enhancement to 1359 nm Line

Throughout this work we see a trend that the on-resonance behavior is expected by scanning through resonance can reveal nonlinear off-resonant behavior. In our pulsed work this is seen in the contrast of on-resonance behavior in Chapter 3 vs the off-resonance behavior of Chapter 4. The same trend holds for the CW case, but the effects are different. Specifically, the on resonance fluorescence data given in Figure 45 of Chapter 5, shows that both of the sets of hyperfine lines on from the two-photon pump of the $6^2D_{3/2}$ line look very similar and the magnitudes of each of the lines are consistent with reasonable populations and the estimated values of the A coefficients.

In Figure 51 we show two 3D plots of the fluorescence spectra as pump wavelength is scanned. The top plot in Figure 51 shows the impact of wavelength tuning near the 885.3990 nm two-photon hyperfine pump line. If we took a slice through the center we would replicate Figure 51 (we reverse the x-axis so that the 1376 nm peak does not eclipse the two-photon feature). On left we have the 1381 nm two-photon line, followed by the 1376, 1360, and the 1359 nm line. Note that as we scan the pump towards the red there is a notable resurgence on the 1359 nm. In the bottom 3D plot of Figure 51 we show the scan through the 885.387 nm line. For this case similar fluorescence levels are present, except for the 1359 nm line where we now note a 100x enhancement to the spectral power as we scan the pump towards the blue (within the solid angle seen by the detector).

An additional view of this data is shown as a log plot in Figure 52. This view highlights the separation of the expected symmetrical on-resonance pump behavior, from the nonlinear off-resonance peaks of the 1359 nm line. Again the two-photon pump resonances are largely separated by the ground state hyperfine splitting. We can resolve the two-photon 1381 nm line, as well as the 1376, 1360, 1359, 1342 nm lines. Figure 52 also shows that the fluorescence peaks at the same pump wavelength

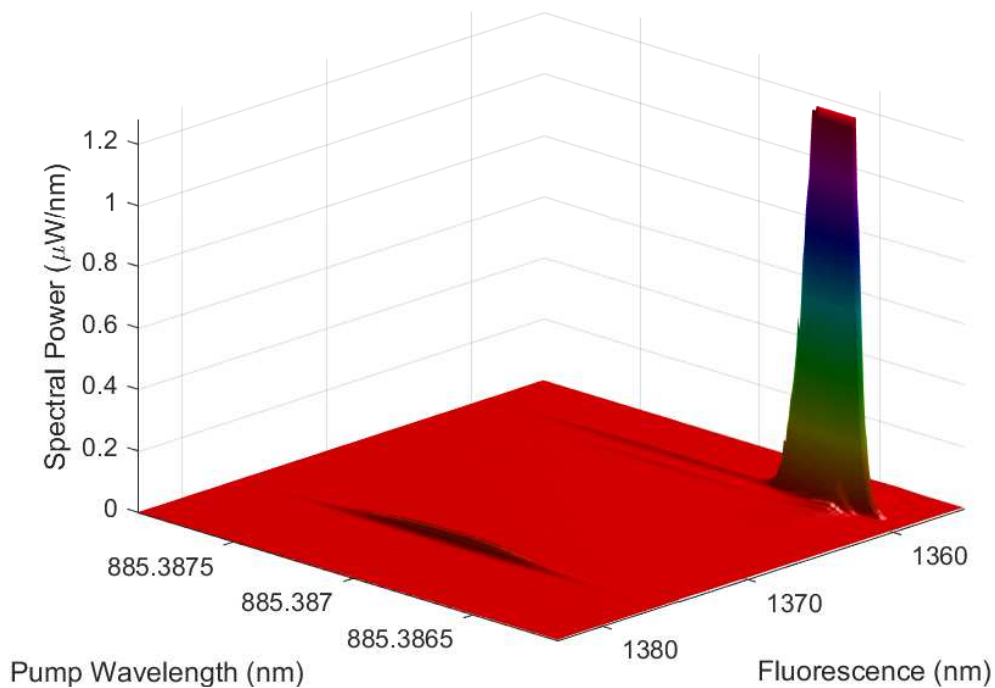
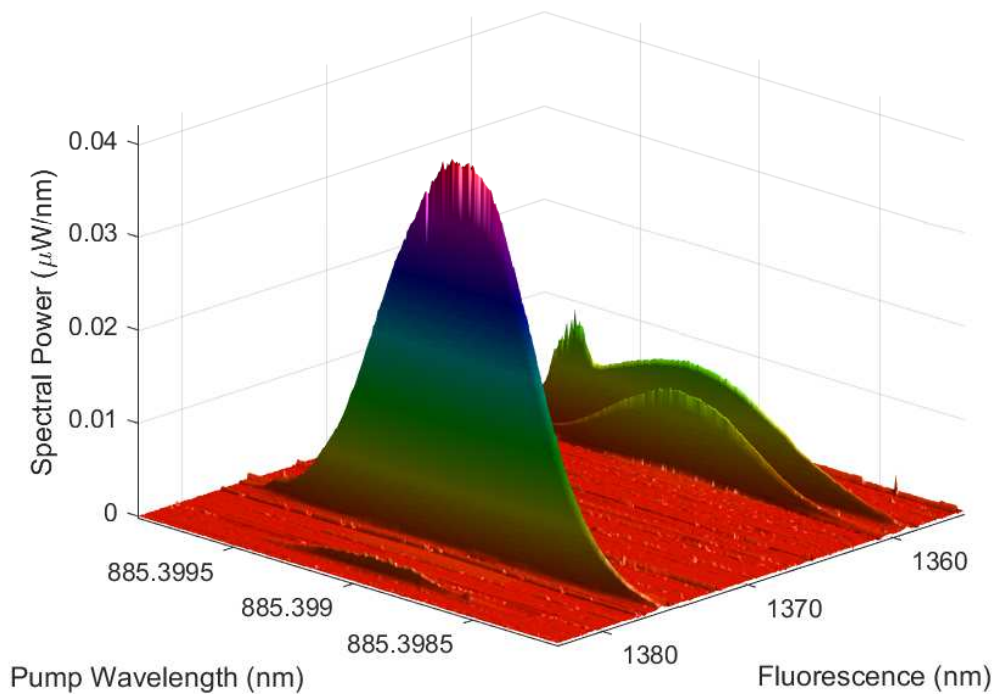


Figure 51. Top: fluorescence spectral tuning at $\nu_4 \rightarrow \nu_{2',3',4',5'}$, note expected behavior but small off-resonance resurgence on 1359 nm line. Bottom: fluorescence spectral tuning at $\nu_3 \rightarrow \nu_{2',3',4',5'}$, significant off-resonant behavior on 1359 nm line. Experimental conditions: pump 1.6W, 0 Torr He, 200 mm lens, 220 °C heat pipe.

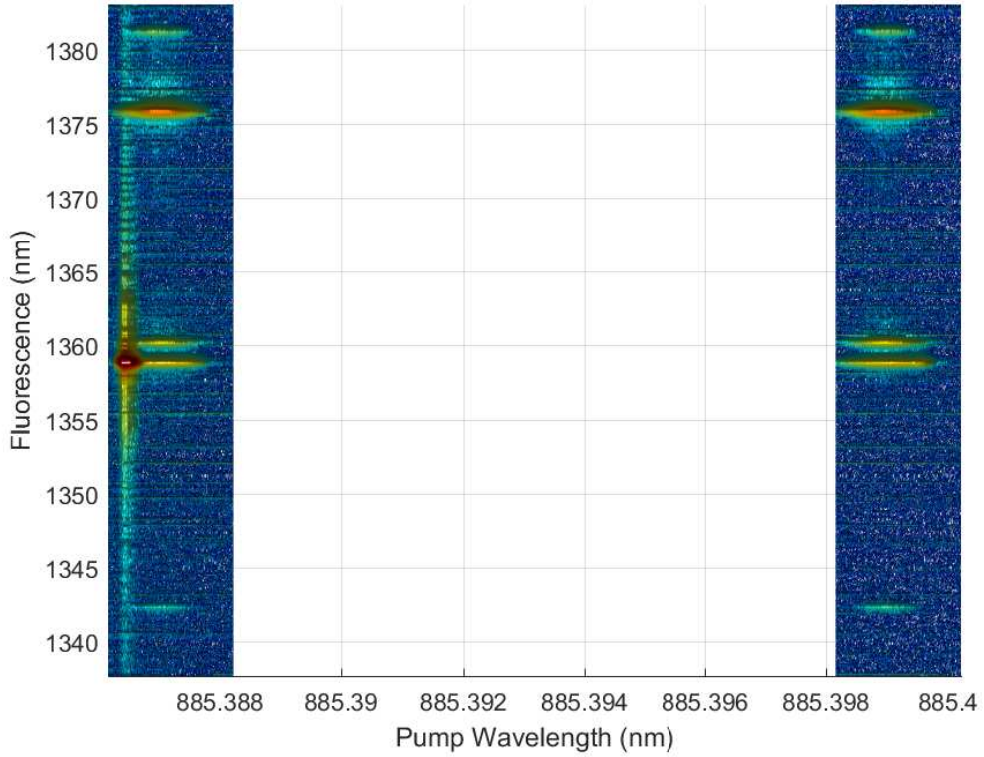


Figure 52. Log plot of fluorescence spectra while tuning pump wavelength. Note that the fluorescence peaks at the same position as the 3 μm laser emission in Figure 49, which is distinct from the 885.386 nm pump enhancement to the 1359 nm line.

as the 3 μm laser peaks from Figure 49. This indicates that the 3 μm laser data from Chapter VI is not influenced by the nonlinear behavior of the 1359 nm line.

Figures 51 and 52 contain data with a fixed pump intensity. We further investigate the enhancement to the 1359 nm line by tuning both intensity and wavelength. We limit our attention to the four two-photon pump transitions near 885.387 nm $\nu_3 \rightarrow \nu_{2',3',4',5'}$ transitions, because they have the strongest nonlinear behavior. The Cs density is kept at $3.7 \times 10^{15}/\text{cm}^3$ which corresponds to a heat pipe temperature of 220 $^\circ\text{C}$ (the same as Figures 51 and 52). We then integrate the spectral power to obtain the total power of the 1359 nm line for two-photon pump wavelengths from 885.387 nm to 885.386 nm.

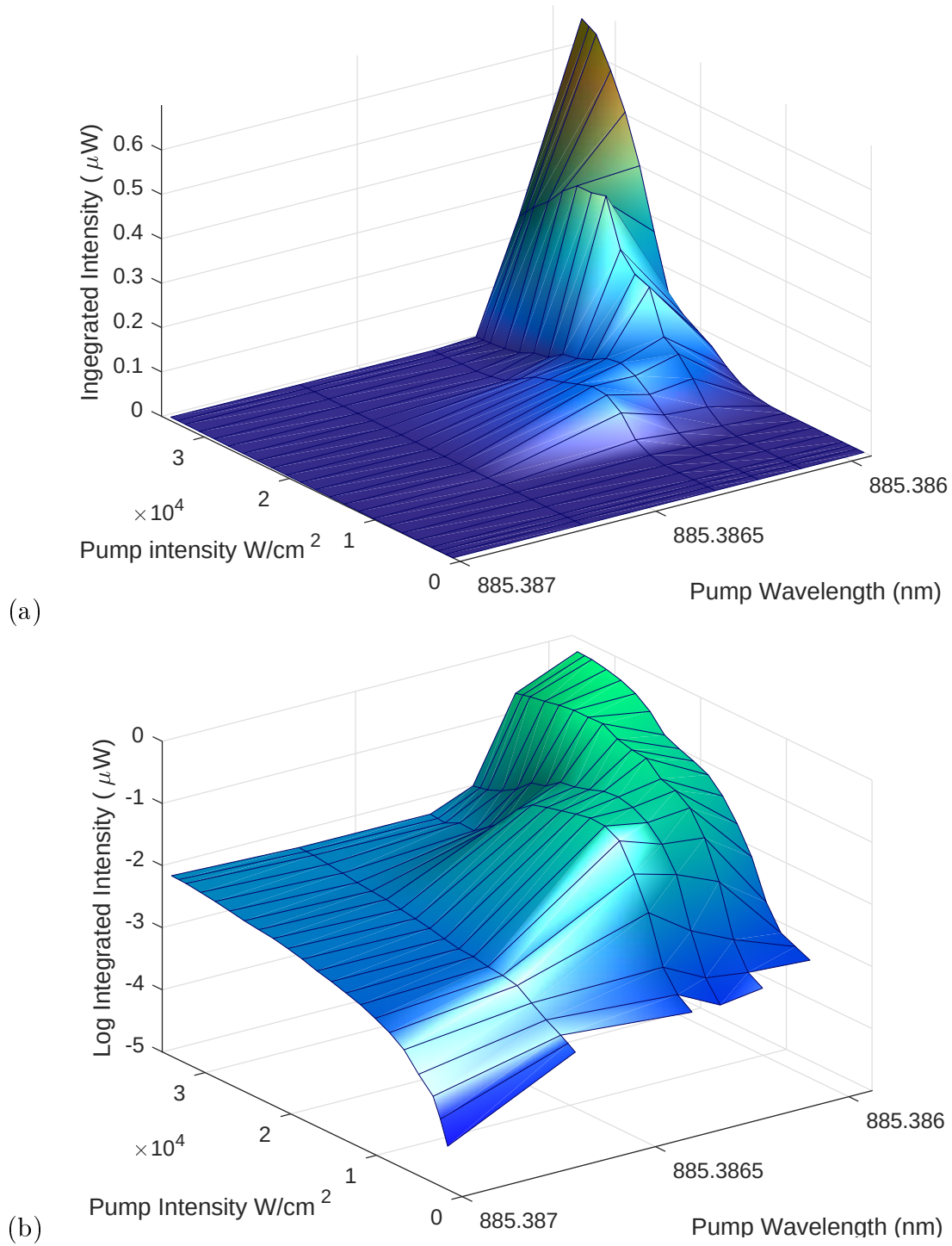


Figure 53. a) Pump wavelength and intensity tuning near $\nu_3 \rightarrow \nu_{2',3',4',5'}$ and integrated spectral intensity of the 1359 nm line. b) log plot. Experimental conditions: cell vacuum, 200 mm lens, 220 °C heat pipe.

These results are shown in Figure 53. The bottom plot in Figure 53, is a Log plot of the output power to highlight the tuning behavior. The left edge of the pump wavelength axis is at 885.387 nm line center (pure fluorescence), and as we can see that the 1359 nm power is both pump wavelength pump intensity dependent.

A likely explanation is a nonlinear process that involves the pump and the 1359 nm line, (e.g. a multi wave mixing process). This initially seems more likely than a shift in the energy levels (e.g. a dressed state) as this would be apparent in our 3 μm lasing spectra in Figure 49.

Shift is far enough off resonance that we can say it that the off-resonance behavior is not seen in the 3 μm data. Although its possible that this plays a minor role in the increased power seen in the 3096 nm line for the $\nu_3 \rightarrow \nu_{2',3',4',5'}$ over the $\nu_4 \rightarrow \nu_{2',3',4',5'}$ seen in Figure 49, we believe it is unlikely the 3 μm power (750 μW) is much greater than the 1359 nm power (0.7 μW). So even if the pump and the 3 μm play a role in the off-resonance enhancement of the 1359 nm line, we do not believe it will be a hindrance to the proposed 3 μm DPAL analog.

This intricate wavelength and pump tuning behavior is likely caused to multiple competing nonlinear processes such as FWM and SRS. We plot the tuning behavior from Figure 49 at a fixed pump wavelength of 885.3860 nm in Figure 54. In this figure we show that at low intensities, the 1359 nm output follows a near I_P^4 behavior, but around 24 kW/cm² a second resurgence is seen which likely indicates a second competing nonlinear processes. One possible explanation for this second enhancement is an SRS process from $6^2P_{1/2} \rightarrow 7^2P_{1/2}$, where $\omega_{6P_{1/2} \rightarrow 7P_{1/2}} = \omega_P + \omega_{SRS}$. At this time we do not understand the exact mechanism. However, it is significant to point out that this is a powerful nonlinear process that can be resolved at very low pump intensities.

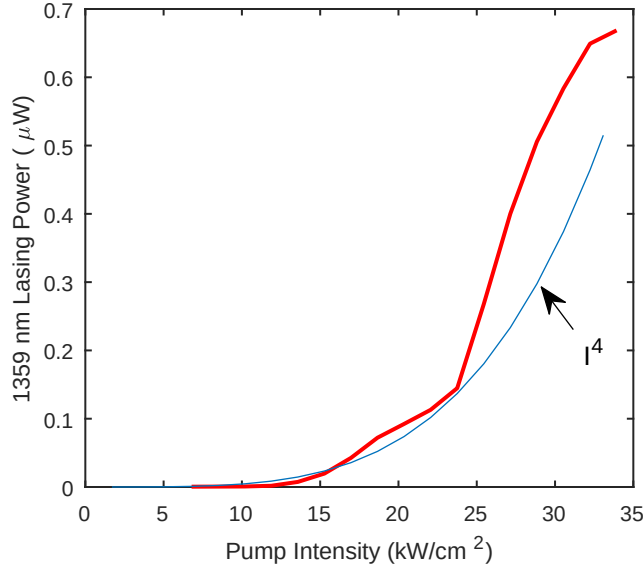


Figure 54. Peak intensity scaling of the off-resonant pumped 1359 nm line. Red line shows the 1359 nm power. Blue line shows an pseudo-fit I^4 line, roughly matched to the lower intensity data. Experimental conditions: pump wavelength 885.3860 nm, heat pipe temperature 220°C, vacuum, 200 mm lens.

CW Two-Photon Line

We originally did not expect to see the 1381 nm two-photon line because our upper limit of CW focused intensity is 66 kW/cm² and is well below our approximate 140 kW/cm² threshold estimate. Yet clearly the same line is present in Figures 45, 51, and 52. We can also see a subtle two-photon wavelength tuning slope from data similar to that in Figure 52 that is consistent with our two-photon wavelength tuning observed in Chapter V.

For this case, two-photon lasing is an unlikely explanation, because an inversion is not reasonable. We are therefore left with SWM and another option: two-photon stimulated emission [25]. In this process a stimulating field (our pump) provides a virtual state for the for an upper-state is emitted spontaneously to the virtual state. In this process one photon (the pump) is stimulated and a photon is added to the

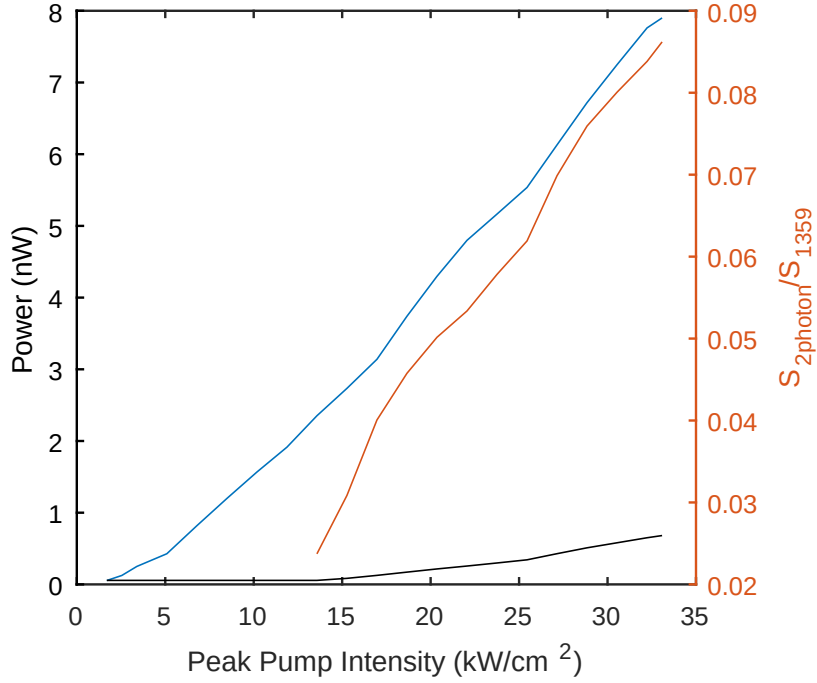


Figure 55. Comparison of 1359 nm intensity to the two-photon intensity. Blue line shows the 1359 nm Fluoresced power with in detector solid angle ($\propto I_P^2$), Black line shows the two-photon power seen by the detector ($\propto I_P^3$), Orange line (right y axis) is the ratio of the two-photon to the 1359 nm ($\propto I_P$). Experimental conditions: pump wavelength 885.3991 nm, heat pipe temperature 220°C, vacuum, 200 mm lens.

pump mode, with the same direction and polarization, while the second photon is emitted in an arbitrary direction. We can treat two-photon stimulated fluorescence as a nonlinear A coefficient where

$$A^{(2)} \propto \sigma^{(2)} I_P \quad (59)$$

The other option of SWM also has issues. As we discuss in Chapter 4, similar process have intensity requirements of GW/cm² (typical Intensity value from a focused pulsed dye laser). Also we did not observe the two-photon line at intensities below 140 kW/cm² in the pulsed case which raises more questions. Again the SWM

intensity is

$$I_{2p}^{(SWM)} \propto I_P^3 I_{3\mu m} I_{12\mu m} \quad (60)$$

where we also note that $I_{3\mu m} \propto I_P^2$ from the data in Figure 43. There for we expect a SWM intensity to be very high order. In Figure 55, we show plots dependent on pump intensity. The blue line is the fluorescence from the 1359 nm line (the on-resonance case not the nonlinear off-resonance case from the last section). This intensity is fluorescence based so $I_{1359} \propto n_{7S} A$. The second line in black is the two-photon intensity which is either $I_{2p}^{(SWM)} \propto I_P^3 I_{3\mu m} I_{12\mu m}$ or from the $A^{(2)}$ process $I_{2p}^{(A2)} \propto n_{7S} \sigma^{(2)} I_P$. The line in orange shows the ration of the two-photon line.

There are two important observations to be made. First, the two-photon power tuning curve is loosely approximated by $I_{2p} \propto I_P^3$. As was note from the Pulsed case in Chapter 4, the expected intensity dependence from a SWM process is not observed. Although Pong[76] reported a cubic dependence to a SWM scheme in sodium at 722 nm, this was from a high energy pulsed source. The second observation is that the ratio of the two-photon to the 1359 nm line is linear which is expected form the two-photon stimulated emission process:

$$\frac{I_{2p}^{(A2)} \propto n_{7S} \sigma^{(2)} I_P}{I_{1359} \propto n_{7S} A} \propto I_P \quad (61)$$

Although this appears consistent, there is minimal work on the subject [25]. Our attempts to estimate an $A^{(2)}$ for our conditions, with the methodology in [25] suggest values (10^2 to 10^3 /s) many orders of magnitude smaller than our observations ($\sim 10^5$ /s).

7.3 Chapter Summary

In this chapter we develop a hyperfine two-photon absorption theory and compare it with some absorption measurements. Our theory provides further evidence for the spatial average factor of $1/5$ for the two-photon case. We also compare this with the $3\text{ }\mu\text{m}$ lasing spectra. The data from these observations provides a revised estimate to the cesium $6^2D_{3/2}$ energy level. Much like our pulsed work covering on-resonance linear lasing processes in Chapter IV and off-resonance and nonlinear processes in Chapter V, our CW work also has nonlinear observations. We observe a strong off-resonant enhancement to the 1359 nm line, potentially caused by MWM within the upper states. Finally we also observe the 1381 nm two-photon line again and compare its behavior with SWM and two-photon stimulated emission.

VIII. Conclusions and Recommendations

8.1 Synopsis and Impact

The impact and feasibility of two-photon pumped Diode Pumped Alkali Lasers (DPAL) has been studied using pulsed and cw surrogate sources. Stimulated emission has been observed for a cesium cell without resonator for wavelengths from the ultraviolet, 387 nm, to the mid infrared, 3600 nm. Cascade lasing on 17 one-photon transitions can be as efficient as 7%, thresholds is modest, $< 120 \text{ kW/cm}^2$ and a CW system has also been demonstrated with a 1.6 W pump source. Several transitions have been observed to tune with pump wavelength, and have theoretical explanations consistent with two photon lasing.

Cascade Lasing

Our first experiment utilized a pulsed source to show potential scaling of over 17 wavelengths. In the near IR, wavelengths of intensest include the 1360 nm $7^2P_{3/2} \rightarrow 5^2D_{5/2}$ line with 5% slope efficiency, 100 kW/cm^2 threshold, and 220 μJ of energy (off-resonance cases show 71 μJ of energy); the 1376 nm $7^2P_{1/2} \rightarrow 5^2D_{3/2}$ line with 5% slope efficiency, 120 kW/cm^2 threshold, and 145 μJ of energy (off-resonance cases show 360 μJ of energy); and the 1469 nm $7^2S_{1/2} \rightarrow 6^2P_{3/2}$ line with 7% slope efficiency, 120 kW/cm^2 threshold, and 105 μJ of energy (off-resonance cases show 85 μJ of energy). In the mid IR transition of interest include the 3096 nm $7^2P_{1/2} \rightarrow 7^2S_{1/2}$ line with 1% slope efficiency, 11 kW/cm^2 threshold, and 82 μJ of energy (off res cases show 120 μJ of energy); and we expect the 2931 nm $7^2P_{3/2} \rightarrow 7^2S_{1/2}$ favorable properties. When alkali density is increased, slope efficiency showed improvement and threshold decreased. The blue 459 nm $7^2P_{1/2} \rightarrow 6^2S_{1/2}$ showed 2% slope efficiency, 120 kW/cm^2 threshold, and 75 μJ of energy (off-resonance cases show 82 μJ of energy); however,

this process has not been proved to be cascade lasing or MWM. Even though much of the past research in the area has focused on the blue generation, we show better efficiencies, and power performance in the near to mid IR where there are simple and obvious cascade lasing processes that are distinct from MWM. We observed the beneficial impact of small amounts of He buffer gas, and demonstrate lasing from fine structure mixing. At 0 Torr He, cesium densities of $6.6 \times 10^{15} \text{ /cm}^3$, and 15 mJ/pulse pump energy, the total lasing energy observed from 300 nm to 2000 nm was 100 μJ , and at 5 Torr He the total lasing energy observed was 550 μJ . When pumping the $6^2D_{5/2}$ state the 1376 nm transition is not expect to lase with out strong mixing because it is not connected on a dipole allowed transition to the pump state. At 0 Torr He, cesium densities of $6.6 \times 10^{15} \text{ /cm}^3$, and 15 mJ/pulse pump energy, the 1376 nm line contained $<.5 \mu\text{J}$ of lasing energy, and its existence was likely due to Cs-Cs collisions. However, when we added 5 Torr He, we see 13 μJ of lasing energy which clearly shows lasing from fine structure mixing. Optical trapping played a significant role for transitions connected to the ground state. We show that this can be used as a tool to control lasing thresholds on undesired one-photon processes. For example we observe (at max pump power) lasing on the D_1 and D_2 lines of around 200 μJ at 200 °C; we decreased this by increasing temperature, and observed 110 μJ at 237.5 °C. At 237.5 °C, the threshold of the D_1 and D_2 lines are also large $\sim 2 \text{ MW/cm}^2$. The line-width of the D_1 and D_2 lines is also very narrow, so we can detune our pump by $\sim .04 \text{ nm}$ and the lines will no longer lase. Other lines in the system show $>0.5 \text{ nm}$ of pump wavelength tuning. Our simplified model of these processes provides an adequate theory for estimating a variety of lasing thresholds and saturation intensities. The model addresses the source of the decreasing lasing threshold with laser intensity. This analysis also suggested 3 μm lasing thresholds $< 11 \text{ kW/cm}^2$.

Our CW pump case proves our assessment of low thresholds, and to the best of our knowledge is the first proof of CW lasing from two-photon pumping. In this work we demonstrate 750 μW of lasing power from a 1.6 W pump source with 0.7% efficiency, and threshold $<10 \text{ kW}/\text{cm}^2$. Our power measurements prove lasing from ASE because the solid angle captured by our detector from the imaging optics would correspond to 2 W of fluoresced power from 1.6 W of pump. The 3 μm field is therefore directional. There is significant opportunity to improve this performance up to a quantum limit of 14% by using a resonator and non-degenerate pumping. Our temperature performance limits are explained by divergence arguments. For instance, at 270 °C, or Cs density of $1.7 \times 10^{16}/\text{cm}^3$, the total number of atoms in our estimated gain volume is 9×10^{11} while the pump has 3×10^{11} photon pairs that the system can cycle. These similar values are indicative of high gain near bleached conditions where we expect path lengths to decrease and divergence to increase, therefore delivering less 3 μm power to our detector. A resonator will be able to address the divergence problem and increase system efficiency. The DPAL architecture and existing diode technology offers a rapid path to power scaling for a two-photon DPAL. Such a device could have significant application and impact for IR counter measures. The only real options available today near this wavelength are quantum cascade lasers. QCLs may be moderately efficient, but there is no clear path to power scaling with good beam quality. Advantages of the two-photon DPAL include rapid power scaling (diode technology exists), good beam quality (very low pressure), and a simplified circulator (heat is not transferred to the gas medium). Two-photon pumping and wavelength agile DPALs can also produce strong beams in the blue 455 and 459 nm, as well as near IR wavelengths at 1360, 1376 and 1469 nm. The near IR wavelengths may have applications in target illuminator lasers, and beacon lasers; and the blue could be used for underwater communication. Proof of CW lasing via ASE from two-photon

pumping is the most significant impact of this work.

A fully hyperfine resolved two-photon absorption theory is developed from first principals, including the degenerate pump case, the non-degenerate pump case, and hyperfine line strengths. Cross-sections are expressed in terms of both dipole moments and A coefficients. Two-photon theory also predicts a hitherto unmentioned factor $1/5$, this is verified with the line strength theory and the expectation value of two dipole operates. In the limit that the process becomes two sequential processes we get a factor of $1/9$ which indicated a potential advantage for two-photon processes. This makes it one of the most comprehensive adaptations of the theory. The accuracy of this theory is tested against known values for rubidium where $7.7 \times 10^{-21} \text{ cm}^4/\text{W}$ is predicted vs Gallagher's recent $6.8 \times 10^{-21} \text{ cm}^4/\text{W}$. We also test it with absorption spectra in cesium and predict and predict an absorbance of 0.20 and measure 0.18. Both of these tests are well with in the 30 to 50% uncertainty of the A coefficients.

This hyperfine theory is also consistent with our hyperfine resolved $3 \mu\text{m}$ lasing spectra. We even used this data to improve accuracy for the $6^2D_{3/2}$ Energy level to $22588.8203 \pm .0003 \text{ cm}^{-1}$.

Fundamental Physics Research: Two-Photon Lasing

In each of our pulsed and CW pump cases we also observe non-linear novel effects slightly detuned from resonance. Our pulsed off-resonance results show enhancement to cascade lasing because the two-photon lines allow more population to be cycled back to the ground state. This may have implications to future two-photon DPALs at very high pump intensities, as well as modern laboratory sources. We study multiple non-linear energetic process with low thresholds. In particular, we study two intense two-photon lines. The first arises from the $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ transition in which a 1381 nm line is observed with $81 \mu\text{J}$, $140 \text{ kW}/\text{cm}^2$ threshold, 1% slope efficiency, and 9 nm

of wavelength tuning. A second two-photon line is observed around 3120 nm, from the $5^2D_{1/2} \rightarrow 6^2S_{1/2}$ transition with 21 μJ and wavelength tuning $\gg 2$ nm (tuning was not measured to find lasing limits, efficiency and threshold were not measured). The strongest impact is the possibility to create a two-photon diode pumped alkali two-photon laser whose operational wavelength is tunable. Although we do not prove the existence of two photon lasing in this work, we proved that a two-photon inversion is theoretically possible in Cs and that key one-photon competing processes can be controlled. This method can also be used to create a tunable wavelength source in the lab that pulsed dye lasers can not normally achieve. Other wavelengths observed include a 387 nm $8^2P_{3/2} \rightarrow 6^2S_{1/2}$ line made possible by the favorable 885.31 nm $5^2D_{3/2} \rightarrow 8^2P_{3/2}$ resonance. This line is observed when our pump is shifted to the blue from the 885.4 nm on-resonance two-photon wavelength. At temperatures near 275 °C we also observe a weak 887 nm line that does not correspond to any known cesium transitions, and we speculate that this is formed by a nonlinear process.

We also compare the two-photon interpretation of our results with FWM and SWM as possible mechanisms. FWM and SWM are largely accepted as the dominant nonlinear mechanism in a wide variety of nonlinear alkali vapor research. Although no population inversion is required for SWM, we provide a simple mechanism for steady state inversion. This inversion only requires a small amount of buffer gas and a small energy separation on the upper states. These conditions exist in rubidium and cesium, but not in sodium and lithium where early two-photon alkali laser research was done. We observe backwards two-photon emission that is not consistent with SWM. Our intensity scaling, and cesium density scaling data are also inconsistent with SWM. Many similar processes in literature occur at GW/cm^2 intensities. Conversely, we see threshold near $\sim 140 \text{ kW}/\text{cm}^2$ and strong lasing by $500 \text{ kW}/\text{cm}^2$.

We also use our two-photon absorption theory to estimate both absorption and

emission cross-sections. This theory provides good agreement with the results we observed. The degenerate absorption cross-section for the $6^2D_{3/2}$ was estimated to be $3.4 \times 10^{-21} \text{ cm}^4/\text{W}$ which is nearly $10\times$ greater than the $6^2D_{5/2}$ estimated at $0.37 \times 10^{-21} \text{ cm}^4/\text{W}$. Our results show much stronger behavior from the $6^2D_{3/2}$ state. For the emission cross-sections we also show the potential for improvement from non-degenerate two-photon lasing. For the $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ transition, with the degenerate pump we predict $4.56 \cdot 10^{-21} \text{ cm}^4/\text{W}$, and for the non-degenerate pump we predict a $10\times$ improvement with $4.93 \cdot 10^{-20} \text{ cm}^4/\text{W}$. For the $5^2D_{3/2} \rightarrow 6^2S_{1/2}$ transition, with the degenerate pump we predict $1.23 \cdot 10^{-20} \text{ cm}^4/\text{W}$, and for the non-degenerate pump we predict an order of magnitude improvement with $1.73 \cdot 10^{-19} \text{ cm}^4/\text{W}$. These non-degenerate are likely the largest Doppler broadened two-photon cross sections ever predicted.

CW pump also has two notable non-linear effects. We note the appearance of the 1381 nm two-photon line, and a dramatic off resonance enhancement to the 1359 nm line. A mixing process may be responsible for the 1359 nm line, but the two-photon line again appears to be inconsistent with SWM. It may be consistent with two-photon stimulated emission where the 1381 nm photon is spontaneously emitted to the virtual state, the intensity trends support this hypothesis but the magnitude of the observations are stronger than any current theory suggests. Clearly these low threshold, CW, two-photon nonlinear observations were not expected and it is not clear if SWM is the dominant mechanism, which indicate opportunities for future work.

8.2 Recommendations

Near Term: Two-Photon DPAL

As we state in the last section, the most important impact of this work is proof of the CW lasing from two-photon pumping. The risk to future device development at the engineering physics level is largely constrained by the high power laser diodes. This is because these diodes are the most expensive part any initial system development. Therefore, we believe that further experimental and modeling work is needed to constrain the trade space of alkali vapor, degenerate/ non-degenerate pump wavelength(s), and atmospheric transmission. Indeed, a full model of the observed processes is essential and would guide both efficiency and scaling assessments.

Similar experimental work should investigate non-degenerate pump sources and the two-photon line shapes. Some may argue that the transitions we pump are too narrow for diode pumping. However, the two-photon process may be more robust as blue wing photons from one pump and red-wing photon from the other create a wider range of usable photons. Our two-photon absorption and emission theory is sound, but its biggest limitations are the A coefficient inputs for the upper states. Further experimental work on degenerate and non-degenerate two-photon absorption to Cs 6D and 7S states will help to improve our estimates of these A coefficients and cross-sections. Results will shed light on diode line width requirements and where the optimal virtual states are located. Also, because this work will scan through one-photon resonances, there will be a wealth of information on how two-photon process may be preferred to two serial one-photon processes. We predict that the simultaneous two-photon process to have the advantages of better population transfer and larger effective cross-section from the factor of $1/5$ vs two $1/3 \times 1/3 = 1/9$ from the serial process.

Sodium, potassium, rubidium, and cesium all have two-photon pump processes of

merit, and the ability to generate similar mid IR wavelengths. Another set of experiments and modeling is needed to explore generation of these mid IR wavelengths. Additional experiments may be needed to verify atmospheric transmission of these specific wavelengths. This information will provide optimal high power pump diode design wavelengths for a particular alkali vapor configuration.

At first glance, the engineering favors cesium as the longer wavelength diodes are generally more efficient; the 3 μm quantum efficiency limit is also more optimal with longer wavelength pump photons.

Sub-Doppler CW work could also assist in improving our understanding of the 3 μm lasing process. The only exotic optic needed is a curved high reflector on a fused silica substrate. This should let $>70\%$ of the 3 μm lasing through while focusing the pump beam back on itself. This should push the efficiency beyond our 1%. However, this is not a resonator for the 3 μm line, and the sub-Doppler two-photon line-shape (natural width) cannot be pumped by diodes. Therefore expanding this experiment to utilize a Doppler broadened pump with an appropriate 3 μm resonator could push the efficiency closer to the 14% quantum limit. Florescence could also be monitored with our techniques. This experiment would provide larger (natural width) more controlled two-photon cross sections so absorption measurements will be improved along with the ability to predict A coefficients and Energy levels.

Fundamental Physics Research: Two-Photon Lasing

Non-degenerate pumping with multiple sources combined with large alkali mixing rates provide a viable path to unambiguously demonstrating a true two-photon laser. The risks in two-photon lasing experiments has been greatly reduced in this work. We have shown two-photon inversion mechanisms exist, two-photon production paths, and cascade lasing from fine-structure mixing. We also show that some one-photon

parasitic processes can be controlled with alkali density and optical trapping. These characteristics all support a novel two-photon laser that operates independently of any multi wave mixing mechanisms. Above we show that the strong mixing can create a two-photon inversion. We propose to use two pump sources to non-degenerately pump the cesium $6^2D_{5/2}$ state and use 5 to 10 Torr of He buffer gas to spin orbit mix to the $6^2D_{3/2}$ state. Then, one or two other seed/probe pumps, possibly combined with one of the other pumps, will check for two-photon gain from the $6^2D_{3/2}$ state. If this gain is still not strong enough, cavities can also be used to enhance two-photon lasing. The use of cavities could also be applied under similar conditions to our $7^2S_{1/2} \rightarrow 6^2S_{1/2}$ two-photon mechanism. For example, a cavity on the 3096 nm pump line would increase the gain from a spin-orbit mixing process which, as we have previously shown, enables a two-photon inversion. Also, any experimental campaign to study cascade lasing from non-degenerate two-photon pumping (above) can also study nonlinear effects. Thus the duality of near-term device work and future fundamental physics research is preserved.

Repeating many of our measurements with additional off-axis fluorescence diagnostics may reveal critical information about the population levels that can further assist in separation of linear from nonlinear observations. This is because both two-photon lasing and multi wave mixing depend on populations levels.

Finally, the ease with which we can see non-linear effects in cesium is dramatic. We note that past work required focused pulsed dye lasers and GW/cm² of pump intensity. We use a CW 1 W Pump. These low thresholds make the system an excellent candidate for future study. Future observations of these nonlinear processes with additional diagnostics may be able to reveal the mechanisms for the nonlinear phenomena. Modeling the coherence effects from these conditions is vital to understanding possible mechanisms and fundamental nonlinear and quantum optics

research.

Appendix A. Resonant Degenerate Two-Photon Absorption

1.1 Setting Up The Problem

Our initial goal is to estimate applicability of two photon process to a creating a new type of Diode Pumped Alkali Laser (DPAL). The first step is to develop a method to compute a two photon absorption cross section. The general two photon absorption mechanism is shown in Figure 56. The two degenerate photons of angular frequency $\omega + \omega = \omega_{14}$, utilize virtual state $|n\rangle$. Although we can think of the virtual state to lie in the “wings” of states $|2\rangle$ and $|3\rangle$, this picture looks more like two serial one-photon processes. We also note that the two photons are not required to be equal in frequency, there is also a non-degenerate path $\omega_1 + \omega_2 = \omega_{14}$, that is not restricted to a particular virtual state. We derive a non-degenerate theory in the Appendix B. This process is most effective if $|1\rangle \rightarrow |4\rangle$ is not a dipole allowed transition, but the intermediary transistions (e.g. $|1\rangle \rightarrow |3\rangle$) are allowed. To analyze the physics of these processes we first consider the perturbation Hamiltonian.

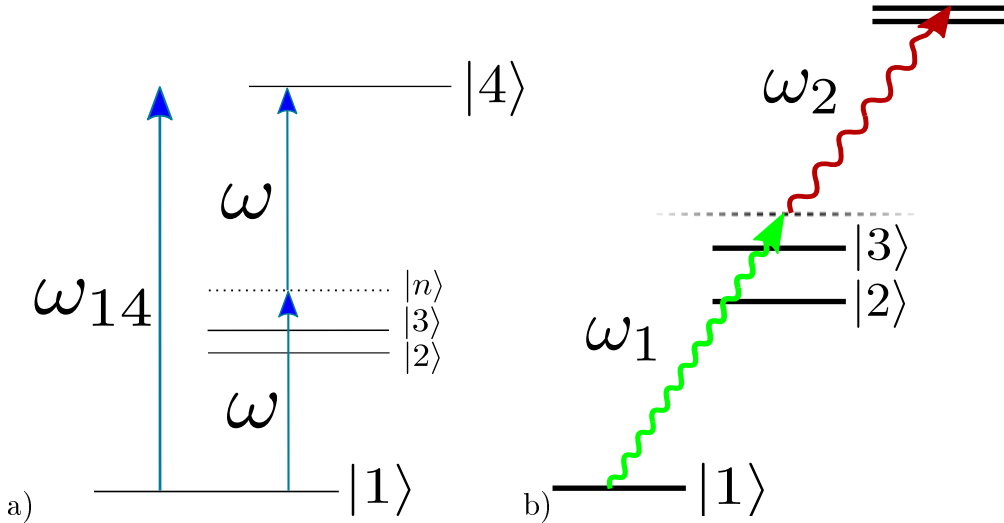


Figure 56. a) Degenerate two-photon absorption from $|1\rangle \rightarrow |4\rangle$ via virtual level $|n\rangle$. b) Non-degenerate two-photon absorption.

The Perturbation Hamiltonian

We first consider a perturbation to our system from an incident electric field. In the dipole approximation, the perturbation Hamiltonian takes the form

$$\hat{H}' = -\hat{\boldsymbol{\mu}} \cdot \mathbf{E} \quad (62)$$

where the dipole moment is $\hat{\boldsymbol{\mu}} = -e \sum_i \hat{\mathbf{r}}_i$, e is the charge on the electron, and $\hat{\mathbf{r}}_i$ is a vector from the nucleus to the electron. Note that this formulation pushes the limits of perturbation theory—and may break it—in that two photon processes are inherently non-linear and therefore relate to high intensities.

We define the incident electric field as:

$$\mathbf{E} = [Ee^{-i\omega t} + E^\dagger e^{i\omega t}] \hat{e} \quad (63)$$

where $\hat{e}$ is the polarization of the field. It is also useful to define the intensity I as

$$I = \frac{c}{n} u \quad (64)$$

where n is the index of refraction, and the energy density u ($\mathbf{E}$ is a real field) gives:

$$I = \frac{c}{n} \left(\frac{1}{2} \left(\epsilon \mathbf{E} \cdot \mathbf{E} + \frac{1}{\mu} \mathbf{B} \cdot \mathbf{B} \right) \right) \quad (65)$$

Using $B = \sqrt{\mu\epsilon}E$ we obtain:

$$I = \frac{c}{2n} \left(\epsilon |\mathbf{E}|^2 + \frac{\mu\epsilon}{\mu} |\mathbf{E}|^2 \right) = \frac{c\epsilon |\mathbf{E}|^2}{n} \quad (66)$$

For optical frequencies $\mu \simeq 1$ and $\epsilon = \epsilon_r \epsilon_0 = n^2 \epsilon_0$ where n is the index of refraction. Finally it is desirable to have I in its time averaged (or average over one period) form

because the frequency is large

$$\langle I \rangle = I = cn\epsilon_0 |\mathbf{E}|^2 = cn\epsilon_0 \lim_{t \rightarrow \infty} \frac{1}{t} \left| \int_0^t \mathbf{E} dt \right|^2 \quad (67)$$

$$I = 2nc\epsilon_0 E^2 \quad (68)$$

where we have relabeled the time averaged intensity as I for simplicity.¹ The next step is too use perturbation theory to obtain transition rates from $\hat{H}'$.

1.2 Second Order Time Dependent Perturbation Theory

Two photon process required second order time-dependent perturbation theory. Shnakar presents an elegant discussion of general higher order perturbation theory in Chapter 18 of reference [87]. We summarize this process in Fig 57. This approach seeks to find the transition probability through the propagator, i.e. $P_{i \rightarrow f} = |\langle f | U_s(t_0, t) | i \rangle|^2$. If we alter traditional perturbation theory by a constant phase, we can view the process as a combination of Zero-order propagators and the interaction Hamiltonian. This provides a good visual and analytic method that does not require the traditional iterative technique.

Mathematically we can express the propagator $U_s(t_0, t)$ from Figure 57, using the perturbation Hamiltonian from the Schrodinger picture H_s^1 , as:

¹Note: if instead $\mathbf{E}$ takes the form $\mathbf{E} = E \cos(\omega t - kz) \hat{i}$ then $I = nc\epsilon_0 E^2/2$

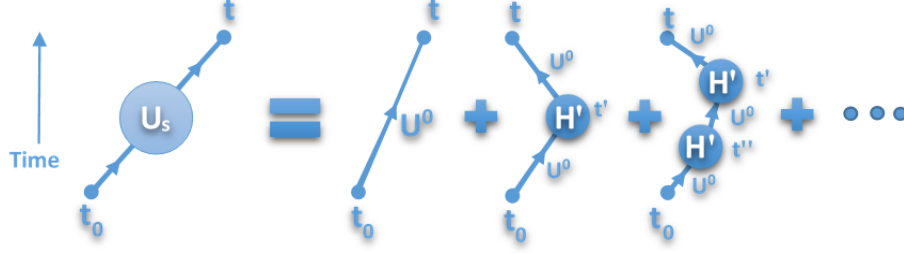


Figure 57. The propagator generated from a series of perturbations. Analogous to a Feynman diagram for perturbation theory. Figure is based on a similar one found in [87].

$$\begin{aligned}
 U_s(t_0, t) &= \underbrace{U_S^0(t_0, t)}_{0^{th}} - \frac{i}{\hbar} \int_{t_0}^t U_S^0(t_0, t) \underbrace{U_S^{0\dagger}(t', t_0) H_s^1 U_S^0(t', t_0)}_{1^{st}} dt' \\
 &+ \left(\frac{i}{\hbar}\right)^2 \int_{t_0}^t \int_{t_0}^{t'} U_S^0(t_0, t) \underbrace{U_S^{0\dagger}(t', t_0) H_s^1 U_S^0(t', t_0) U_S^{0\dagger}(t'', t_0) H_s^1 U_S^0(t'', t_0)}_{2^{nd}} dt'' dt' + \dots
 \end{aligned} \tag{69}$$

or

$$\begin{aligned}
 \langle f | U_s(t_0, t) | i \rangle &= \underbrace{\delta_{if}}_{0^{th}} e^{-iE_i(t-t_0)/\hbar} - \frac{i}{\hbar} \int_{t_0}^t \underbrace{e^{-iE_f(t-t')/\hbar} \langle f | H_s^1 | i \rangle e^{-iE_i(t'-t_0)/\hbar}}_{1^{st}} dt' \\
 &+ \left(\frac{i}{\hbar}\right)^2 \int_{t_0}^t \int_{t_0}^{t'} \underbrace{\sum_n e^{-iE_f(t-t')/\hbar} \langle f | H_s^1 | n \rangle e^{-iE_n(t'-t'')/\hbar} \langle n | H_s^1 | i \rangle e^{-iE_i(t''-t_0)/\hbar}}_{2^{nd}} dt'' dt' + \dots
 \end{aligned} \tag{70}$$

For a two-photon transition, the first two terms are zero: the first δ_{if} is 0 because the probability to spontaneously jump to final state is ~ 0 , and the second is 0 because one photon is not going to make a dipole allowed two photon transition (we are

ignoring the electric quadrupole which is at a much shorter wavelength). We now have

$$\begin{aligned} \langle f | U_s(t_0, t) | i \rangle = \\ \left(\frac{i}{\hbar} \right)^2 \int_{t_0}^t \int_{t_0}^{t'} \sum_n e^{-iE_f(t-t')/\hbar} \langle f | H_s^1(t') | n \rangle e^{-iE_n(t'-t'')/\hbar} \langle n | H_s^1(t'') | i \rangle e^{-iE_i(t''-t_0)/\hbar} dt'' dt' \end{aligned} \quad (71)$$

We can set $t_0 = 0$ and note that $P_{i \rightarrow f} = |\langle f | U_s(t_0, t) | i \rangle|^2$ is invariant under a constant phase² so we can pull out $e^{-iE_f t}$. Using equations 62 and 63 above the perturbation Hamiltonian takes the form:

$$\langle a | \hat{H}'(t) | b \rangle = \langle a | \hat{\mu} \cdot E | b \rangle e^{-i\omega t} + \langle a | \hat{\mu} \cdot E^\dagger | b \rangle e^{i\omega t} \simeq E \langle a | \hat{\mu} \cdot \hat{e} | b \rangle e^{-i\omega t} = H_{ab} e^{-i\omega t} \quad (72)$$

where we define $\omega = \omega_{fi} = (E_f - E_i)/\hbar$, we use the notation $E \langle a | \hat{\mu} \cdot \hat{e} | b \rangle = H_{ab}$ for the perturbation Hamiltonian, and we have made the rotating wave approximation and dropped the counter rotating component³ (i.e. the component relating to the $f \rightarrow i$

²This is the phase we alluded to above to make this propagator picture work for perturbation theory.

³At this point we ignore the orientation of the dipole operator to the field E. But we correct this in the integration later with “factor of 3 vs factor of 5” later.

emission)

$$\begin{aligned}
\langle f|U|i\rangle &= \\
&= \left(\frac{i}{\hbar}\right)^2 \int_0^t \int_0^{t'} \sum_n e^{-iE_f(-t')/\hbar} H_{fn} e^{-i\omega t'} e^{-iE_n(t'-t'')/\hbar} H_{ni} e^{-i\omega t''} e^{-iE_i(t'')/\hbar} dt'' dt' \\
&= \left(\frac{i}{\hbar}\right)^2 \sum_n H_{fn} H_{ni} \int_0^t \int_0^{t'} e^{-iE_f(-t')/\hbar} e^{-i\omega t'} e^{-iE_n(t'-t'')/\hbar} e^{-i\omega t''} e^{-iE_i(t'')/\hbar} dt'' dt' \\
&= \left(\frac{i}{\hbar}\right)^2 \sum_n H_{fn} H_{ni} \int_0^t \int_0^{t'} e^{i[(\omega_{fn}-\omega)t' + (\omega_{ni}-\omega)t'']} dt'' dt' \\
&= \left(\frac{i}{\hbar}\right)^2 \sum_n H_{fn} H_{ni} \frac{i}{\omega - \omega_{ni}} \int_0^t \left(e^{it'(\omega_{fn} + \omega_{ni} - 2\omega)} - e^{it'(\omega_{fn} - \omega)} \right) dt' \\
&= \left(\frac{i}{\hbar}\right)^2 \sum_n H_{fn} H_{ni} \frac{i}{\omega - \omega_{ni}} \int_0^t \left(e^{it'(\omega_{fi} - 2\omega)} - e^{it'(\omega_{fn} - \omega)} \right) dt' \tag{73}
\end{aligned}$$

The rotation wave approximation can be used again by noting that $e^{it'(\omega_{fn}-\omega)}$ is a counter rotating component (corresponding to absorption from the intermediary virtual state to the final state).

$$\begin{aligned}
\langle f|U_s(t_0=0,t)|i\rangle &= \left(\frac{i}{\hbar}\right)^2 \sum_n H_{fn} H_{ni} \frac{i}{\omega - \omega_{ni}} \int_0^t e^{it'(\omega_{fi} - 2\omega)} dt' \\
&= \left(\frac{i}{\hbar}\right)^2 \sum_n H_{fn} H_{ni} \frac{1 - e^{it(\omega_{fi} - 2\omega)}}{(\omega - \omega_{ni})(2\omega - \omega_{fi})} \\
&= \sum_n \frac{H_{fn} H_{ni}}{\hbar^2 (\omega_{ni} - \omega)} \frac{1 - e^{it(\omega_{fi} - 2\omega)}}{(2\omega - \omega_{fi})} \\
&= \sum_n \frac{H_{fn} H_{ni}}{\hbar^2 (\omega_{ni} - \omega)} \frac{e^{it(\omega_{fi} - 2\omega)} - 1}{(\omega_{fi} - 2\omega)}. \tag{74}
\end{aligned}$$

Now the transition probability can be written as

$$\begin{aligned}
P_{i \rightarrow f} = |\langle f | U_s(t_0, t) | i \rangle|^2 &= \sum_n \left| \frac{H_{fn} H_{ni}}{\hbar^2 (\omega_{ni} - \omega)} \right|^2 \left| \frac{e^{it(\omega_{fi} - 2\omega)} - 1}{(\omega_{fi} - 2\omega)} \right|^2 \\
&= \sum_n \left| \frac{H_{fn} H_{ni}}{\hbar^2 (\omega_{ni} - \omega)} \right|^2 \frac{4 \sin^2[(\omega_{fi} - 2\omega) \frac{t}{2}]}{(\omega_{fi} - 2\omega)^2}. \quad (75)
\end{aligned}$$

However we are really interested in the transition rate that can be related to experiment:

$$R_{i \rightarrow f} = \lim_{t \rightarrow \infty} \frac{d}{dt} P_{i \rightarrow f}. \quad (76)$$

We can obtain an expression for $R_{i \rightarrow f}$ by noting that the Sinc function is similar to the delta function in the distributional sense:

$$\delta(\omega_{fi} - 2\omega) = \frac{1}{2\pi} \int_{-\infty}^{\infty} e^{it'(\omega_{fi} - 2\omega)} dt' \quad (77)$$

From above we note

$$\begin{aligned}
\lim_{t \rightarrow \infty} \frac{d}{dt} \left(\frac{4 \sin^2[(\omega_{fi} - 2\omega) \frac{t}{2}]}{(\omega_{fi} - 2\omega)^2} \right) &= \lim_{t \rightarrow \infty} \frac{d}{dt} \left| \int_0^t e^{it'(\omega_{fi} - 2\omega)} dt' \right|^2 \\
&\rightarrow \lim_{t \rightarrow \infty} \frac{d}{dt} [4\pi^2 \delta(\omega_{fi} - 2\omega) \delta(\omega_{fi} - 2\omega)] \quad (78)
\end{aligned}$$

and we handle the product of delta functions as follows:

$$\begin{aligned}
\lim_{t \rightarrow \infty} \frac{d}{dt} [4\pi^2 \delta(\omega_{fi} - 2\omega) \delta(\omega_{fi} - 2\omega)] &= 2\pi \lim_{t \rightarrow \infty} \frac{d}{dt} \left(\delta(\omega_{fi} - 2\omega) \int_{-t/2}^{t/2} e^{it'(\omega_{fi} - 2\omega)} dt' \right) \\
&= 2\pi \delta(\omega_{fi} - 2\omega) \lim_{t \rightarrow \infty} \frac{d}{dt} \left(\int_{-t/2}^{t/2} e^{it'(\omega_{fi} - 2\omega)} dt' \right) \quad (79)
\end{aligned}$$

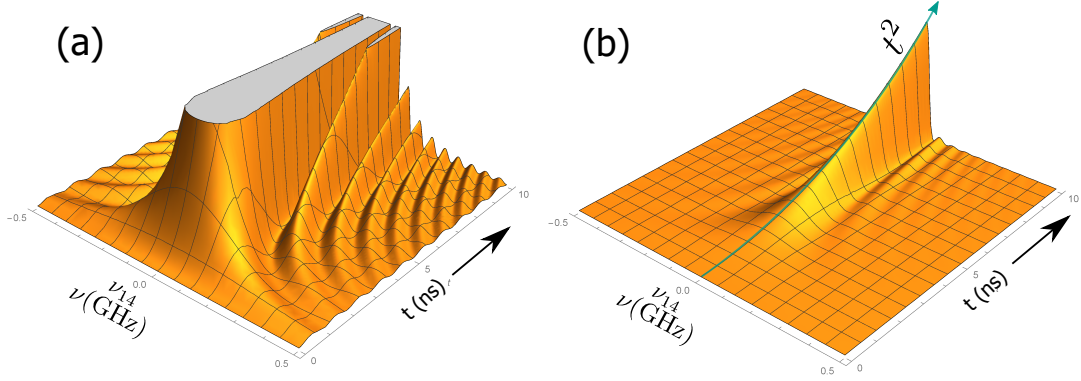


Figure 58. Illustration of the collapse of $\frac{4 \sin^2[(\omega_{fi}-2\omega)\frac{t}{2}]}{(\omega_{fi}-2\omega)^2} \rightarrow \delta$. (a) shows a 30x magnification of the vertical axis, and (b) shows the entire axis. Note that the quantum-like ripple behavior becomes negligible, the width of the central peak decreases, and the peak increases as t^2 . Thus as $t \rightarrow \infty$ we get a δ .

Because $\delta(\omega_{fi} - 2\omega) = 0$ unless $\omega_{fi} = 2\omega$:

$$\begin{aligned}
 2\pi\delta(\omega_{fi} - 2\omega) \lim_{t \rightarrow \infty} \frac{d}{dt} \left(\int_{-t/2}^{t/2} e^{it'(\omega_{fi}-2\omega)} dt' \right) &= 2\pi\delta(\omega_{fi} - 2\omega) \lim_{t \rightarrow \infty} \frac{d}{dt} \left(\int_{-t/2}^{t/2} e^{it'(0)} dt' \right) \\
 &= 2\pi\delta(\omega_{fi} - 2\omega) \lim_{t \rightarrow \infty} \frac{d}{dt} \left(\int_{-t/2}^{t/2} 1 dt' \right) \\
 &= 2\pi\delta(\omega_{fi} - 2\omega) \quad (80)
 \end{aligned}$$

Pictorially we can view this process in Figure 58 where we have plotted the surface of $\sin^2[2\pi\nu t]/(2\pi\nu)^2$ around the central frequency ν_{14} . Clearly the function behaves like a δ at large t .

Using this expression with 75 and 76 we obtain *Fermi's golden rule* for two photon absorption:

$$R_{i \rightarrow f} = \sum_n \left| \frac{H_{fn} H_{ni}}{\hbar^2 (\omega_{ni} - \omega)} \right|^2 2\pi\delta(\omega_{fi} - 2\omega) \quad (81)$$

This result is true for a single pure state, but it is unrepresentative of an actual physical result. We correct equation 81 by introducing a density of final states or line shape function $\rho_f(\omega)$.⁴ Examples include Lorentzian, Gaussian, and Voigt profiles. We can now rewrite 81 as:

$$R_{i \rightarrow f}^{(2)} = \int \sum_n \left| \frac{\langle f | \hat{\mu} \cdot \hat{e} | n \rangle \langle n | \hat{\mu} \cdot \hat{e} | i \rangle E^2}{\hbar^2 (\omega_{ni} - \omega)} \right|^2 2\pi \delta(\omega_{fi} - 2\omega) \rho_f(\omega_{fi}) d\omega_{fi}$$

$$R_{i \rightarrow f}^{(2)} = \sum_n \left| \frac{\langle f | \hat{\mu} \cdot \hat{e} | n \rangle \langle n | \hat{\mu} \cdot \hat{e} | i \rangle E^2}{\hbar^2 (\omega_{ni} - \omega)} \right|^2 2\pi \rho_f(\omega_{fi} = 2\omega) \quad (82)$$

Note the superscript (2) has been added to $R_{i \rightarrow f}^{(2)}$ to highlight a two-photon transition rate. In passing to the next section we note that this expression is only correct to a *factor of 3*, as will be explained below. We could also repeat this process for 1 photon (see section 12.5 from [10]) from i (initial/ground) to n (the final state) as:

$$R_{i \rightarrow n}^{(1)} = \left| \frac{\langle n | \hat{\mu} \cdot \hat{e} | i \rangle E}{\hbar} \right|^2 2\pi \rho_f(\omega_{ni} = \omega) \quad (83)$$

The Factor of 3 or 5

A general discussion of the following can be found in section 3.5 of [89]. The derivation of 82 in Boyd [10] implicitly assumes that the dipole operator is perfectly aligned with the incident field. However, it is unrepresentative of our case where we have an ensemble of atoms each with randomly orientated dipole operators (i.e. the dipoles is not aligned). Therefore, each atom effectively sees a different and randomly polarized incident field. We therefore take the expectation values of the dipole operator, i.e. an average over all angular orientations. Mathematically we need to find the average component of $\hat{\mu}^2$ in the $\mathbf{E}$ -direction ($\hat{e}$). From above we have

⁴Note: if ρ_f is a function of E rather than ω we lose a factor of $\hbar$ since $\delta(\omega_{fi} - 2\omega) = \delta(E_{fi} - 2E)/J = \hbar \delta(E_{fi} - 2E)$ where J is the Jacobian.

$$|\langle a | \hat{\mu} \cdot E | b \rangle|^2 = E^2 |\langle a | \hat{\mu} \cdot \hat{e} | b \rangle|^2 \quad (84)$$

Again, quantum mechanics tells us—in the absence of any alignment mechanism—we can't know the orientation of the dipole operator. We can arbitrarily chose $\mathbf{E}$ in the $\hat{z}$ direction:

$$|\langle a | \hat{\mu} \cdot \hat{z} | b \rangle|^2 = \mu_{ab}^2 (\hat{r} \cdot \hat{z}) \quad (85)$$

which is a function of $\hat{r} \cdot \hat{z}$. In general this value could range from zero to μ_{ab}^2 . In a given ensemble of atoms the angular expectation value is

$$\langle \mu_{ab}^2 (\hat{r} \cdot \hat{z}) \rangle = \mu_{ab}^2 \langle \cos^2(\theta) \rangle = \frac{\mu_{ab}^2}{4\pi} \int \int \cos^2(\theta) \sin(\theta) d\theta d\phi = \frac{\mu_{ab}^2}{3} \quad (86)$$

Hence we see a factor of $1/3$ for μ_{ab}^2 . So for the angular average two-photon case we have:

$$\langle R_{i \rightarrow f}^{(2)} \rangle = \langle \mu_{ab}^2 (\hat{r} \cdot \hat{z}) \mu_{bc}^2 (\hat{r} \cdot \hat{z}) \rangle = \mu_{ab}^2 \mu_{bc}^2 \langle \cos^4(\theta) \rangle = \frac{\mu_{ab}^2 \mu_{bc}^2}{5} \quad (87)$$

Therefore we assume the angular averaging for the two-photon process gets a factor of $1/5$.

Two Photon Absorption Cross Section

We now seek to link Fermi's golden rule to the absorption cross section. Our next objective is to obtain an expression for the absorption cross section in terms of Einstein A coefficient which can be found experimentally. The absorption cross section provides a key link between modeling and experimental laser results.

The One Photon Case

For single photon transition, we relate the time derivative for state N_n (the single photon transition rate) to the Einstein B coefficient or the single-photon absorption cross section by⁵:

$$\frac{dN_n}{dt} = R_{i \rightarrow n}^{(1)} N_i = B_{i \rightarrow n} u \rho_f (\nu_{ni} - \nu) N_i = \sigma_{in}^{(1)}(\nu) \frac{I}{h\nu} N_i \quad (88)$$

or

$$R_{i \rightarrow n}^{(1)} = B_{i \rightarrow n} \rho_f (\nu_{ni} - \nu) u = \sigma_{in}^{(1)}(\nu) \frac{I}{h\nu} \quad (89)$$

where we identify $R_{i \rightarrow n}^{(1)}$ as the angular average of the one-photon rate, the time average intensity I is given by equation 68, and $\sigma_{ij}^{(1)}(\nu)$ represents the single photon absorption cross section. Note some authors—e.g. Section 12.5 of [10]—define $R_{i \rightarrow j}^{(1)} = \sigma_{ij}^{(1)} I$ and absorb the other factors into the definition of $\sigma_{ij}^{(1)}$; we shall use the former representation. Before jumping to the two photon case note that equation 89 gives us a relationship between $B_{i \rightarrow n}$ and $\sigma_{i \rightarrow n}^{(1)}$ (again using i for initial/ground and n for final). The absorption $B_{i \rightarrow f}$ and emission $B_{f \rightarrow i}$ are related by:

$$B_{i \rightarrow n} = \frac{g_n}{g_i} B_{n \rightarrow i} \quad (90)$$

where g_i is the degeneracy of state i . B is related to the A coefficient by:

$$A_{n \rightarrow i} = \frac{\hbar \omega_{ni}^3 n^3}{\pi^2 c^3} B_{n \rightarrow i} \quad \text{or} \quad \frac{8\pi n^3 h \nu_{ni}^3}{c^3} B_{n \rightarrow i} \quad (91)$$

$$B_{i \rightarrow n} = \frac{g_n}{g_i} B_{n \rightarrow i} = \sigma_{in}^{(1)} \frac{I}{h\nu_{ni}} \frac{1}{u} = \sigma_{in}^{(1)} \frac{c}{h\nu n} \quad (92)$$

⁵Note this is in terms of ν not ω , and stimulated emission and spontaneous emission are not included

Putting the above pieces together we find the absorption cross section for a single photon:

$$\sigma_{in}^{(1)} = \frac{g_n}{g_i} \frac{\lambda_{ni}^2}{8\pi n^2} A_{n \rightarrow i} \rho_f(\nu_{ni} = \nu) \quad (93)$$

To relate this to equation 83 we note that the lineshape $2\pi\rho_f(\omega_{ni} - \omega) = \rho_f(\nu_{ni} - \nu)$.

We combine this with 83 and 86 and 68:

$$R_{i \rightarrow n}^{(1)} = \frac{\mu_{ni}^2 E^2}{3h^2} 4\pi^2 \rho_f(\nu_{ni} = \nu) = \sigma_{in}^{(1)}(\nu) \frac{2nc\epsilon_0 E^2}{h\nu_{ni}}$$

and we find:

$$A_{n \rightarrow i} = \frac{1}{g_n} \frac{16\pi^3}{3\epsilon_0 h \lambda_{ni}^3} \mu_{ni}^2 \quad (94)$$

or

$$\mu_{ni}^2 = g_n \frac{3\epsilon_0 h \lambda_{ni}^3}{16\pi^3} A_{n \rightarrow i} \quad (95)$$

Note that the degeneracy g_i seems to have vanished but we have implicitly assumed that the rate $R_{i \rightarrow n}^{(1)}$ will include a sum over final degenerate states to recover the degeneracy factor. With these expressions we can now address the two photon absorption cross section.

The Two-Photon Case

We define the two-photon cross section with (using the density of final states and summing over the upper degeneracies and dividing by the lower degeneracies to insure each transition is only counted once):

$$R_{i \rightarrow f}^{(2)} = \sum_n \frac{1}{5} \frac{\sum_{m_{jf}} \sum_{m_{jn}}}{\sum_{m_{ji}} \sum_{m_{jn}}} \left| \frac{\mu_{fnj} \mu_{ni} E^2}{\hbar^2 (\omega_{ni} - \omega)} \right|^2 2\pi \rho_f(\omega_{fi} = 2\omega) \quad (96)$$

$$= \sum_n \frac{1}{5} \frac{1}{g_i g_n} \left| \frac{\mu_{fn} \mu_{ni} E^2}{\hbar^2 (\omega_{ni} - \omega)} \right|^2 2\pi \rho_f(\omega_{fi} = 2\omega) = \sigma_{if}^{(2)} \frac{I^2}{h\nu} \quad (97)$$

where $R_{i \rightarrow f}^{(2)}$ is identified as the angular average of the two-photon rate. Using our definition from equation 95 the upper degeneracies are included in the definition of μ^2 . The $1/5$ comes from equation 87. Note that Boyd[10] defined this as: $R_{i \rightarrow f}^{(2)} = \sigma_{if}^{(2)} I^2$ as in the one photon case this is arbitrary but we put in a factor of $h\nu$ to be consistent with traditional one-photon processes.

$$\begin{aligned} \sigma_{if}^{(2)} &= \frac{h\nu}{I^2} \sum_n \frac{1}{g_i g_n} \frac{\mu_{fn}^2 \mu_{ni}^2 E^4}{5\hbar^4 (2\pi (\nu_{ni} - \nu))^2} \rho_f(\nu_{fi} = 2\nu) \\ &= \frac{h\nu}{(2nc\epsilon_0)^2} \sum_n \frac{1}{g_i g_n} \frac{\mu_{fn}^2 \mu_{ni}^2}{5\hbar^4 (2\pi (\nu_{ni} - \nu))^2} \rho_f(\nu_{fi} = 2\nu) \\ &= \frac{h\nu}{(2nc\epsilon_0)^2} \sum_n \frac{1}{g_i g_n} \frac{g_f \frac{3\epsilon_0 h \lambda_{fn}^3}{16\pi^3} A_{fn} \cdot g_n \frac{3\epsilon_0 h \lambda_{ni}^3}{16\pi^3} A_{ni}}{5\hbar^4 (2\pi (\nu_{ni} - \nu))^2} \rho_f(\nu_{fi} = 2\nu) \\ &= \frac{9}{5} \frac{h\nu}{(2nc)^2} \sum_n \frac{g_f}{g_i} \frac{\frac{\lambda_{fn}^3}{16\pi^3} A_{fn} \cdot \frac{\lambda_{ni}^3}{16\pi^3} A_{ni}}{h^2 (\nu_{ni} - \nu)^2} \frac{(2\pi)^4}{(2\pi)^2} \rho_f(\nu_{fi} = 2\nu) \end{aligned} \quad (98)$$

Simplifying we obtain:

$$\sigma_{if}^{(2)}(\nu) = \frac{9}{5} \frac{\nu}{256\pi^4 h n^2 c^2} \sum_n \frac{g_f}{g_i} \frac{\lambda_{fn}^3 A_{fn} \lambda_{ni}^3 A_{ni}}{(\nu_{ni} - \nu)^2} \rho_f(\nu_{fi} = 2\nu) \quad (99)$$

or

$$\sigma_{if}^{(2)}(\nu) = \frac{\pi^2 \nu}{5h^3 n^2 c^2 \epsilon_0^2} \sum_n \frac{1}{g_i g_n} \frac{\mu_{fn}^2 \mu_{ni}^2}{(\nu_{ni} - \nu)^2} \rho_f(\nu_{fi} = 2\nu) \quad (100)$$

Appendix B. Resonant Non-Degenerate Two-Photon Absorption

Much of this follows the derivation from Appendix A. We start with the Electric field:

$$\mathbf{E} = \left(E_1 e^{-i\omega_1 t} + E_1^\dagger e^{i\omega_1 t} \right) \hat{e}_1 + \left(E_2 e^{-i\omega_2 t} + E_2^\dagger e^{i\omega_2 t} \right) \hat{e}_2 \quad (101)$$

If the fields are propagating in the same direction, the corresponding time averaged intensity is

$$I = 2nc\epsilon_0 (E_1^2 + E_2^2) = I_1 + I_2 \quad (102)$$

Next we compute the matrix elements of the propagator,

$$\begin{aligned} \langle f | U_s | i \rangle &= \left(\frac{i}{\hbar} \right)^2 \int_{t_0}^t \int_{t_0}^{t'} \sum_n e^{-iE_f(t-t')/\hbar} \langle f | H_s^1(t') | n \rangle e^{-iE_n(t'-t'')/\hbar} \\ &\quad \times \langle n | H_s^1(t'') | i \rangle e^{-iE_i(t''-t_0)/\hbar} dt'' dt' \end{aligned} \quad (103)$$

The dipole matrix elements of the perturbation in the Schrodinger picture are:

$$\begin{aligned} \langle f | H_s^1(t') | n \rangle &= E_1 e^{-i\omega_1 t'} \langle f | \hat{r} \cdot \hat{e}_1 | n \rangle + E_1^\dagger e^{i\omega_1 t'} \langle f | \hat{r} \cdot \hat{e}_1 | n \rangle \\ &\quad + E_2 e^{-i\omega_2 t'} \langle f | \hat{r} \cdot \hat{e}_2 | n \rangle + E_2^\dagger e^{i\omega_2 t'} \langle f | \hat{r} \cdot \hat{e}_2 | n \rangle \end{aligned} \quad (104)$$

and using the RWA we obtain:

$$\begin{aligned} \langle f | H_s^1(t') | n \rangle &= E_1 e^{-i\omega_1 t'} \langle f | \hat{r} \cdot \hat{e}_1 | n \rangle + E_2 e^{-i\omega_2 t'} \langle f | \hat{r} \cdot \hat{e}_2 | n \rangle \\ &= H_{fn}^{(1)} e^{-i\omega_1 t'} + H_{fn}^{(2)} e^{-i\omega_2 t'} . \end{aligned} \quad (105)$$

Substitution into the matrix elements of the propagator yields

$$\begin{aligned}
\langle f|U_s|i\rangle &= \left(\frac{i}{\hbar}\right)^2 \sum_n H_{fn}^{(1)} H_{ni}^{(1)} \int_{t_0}^t \int_{t_0}^{t'} e^{-iE_f(-t')/\hbar} e^{-i\omega_1 t} e^{-iE_n(t'-t'')/\hbar} e^{-i\omega_1 t} e^{-iE_i(t'')/\hbar} dt'' dt' \\
&+ H_{fn}^{(1)} H_{ni}^{(2)} \int_{t_0}^t \int_{t_0}^{t'} e^{-iE_f(-t')/\hbar} e^{-i\omega_1 t} e^{-iE_n(t'-t'')/\hbar} e^{-i\omega_2 t} e^{-iE_i(t'')/\hbar} dt'' dt' \\
&+ H_{fn}^{(2)} H_{ni}^{(1)} \int_{t_0}^t \int_{t_0}^{t'} e^{-iE_f(-t')/\hbar} e^{-i\omega_2 t} e^{-iE_n(t'-t'')/\hbar} e^{-i\omega_1 t} e^{-iE_i(t'')/\hbar} dt'' dt' \\
&+ H_{fn}^{(2)} H_{ni}^{(2)} \int_{t_0}^t \int_{t_0}^{t'} e^{-iE_f(-t')/\hbar} e^{-i\omega_2 t} e^{-iE_n(t'-t'')/\hbar} e^{-i\omega_2 t} e^{-iE_i(t'')/\hbar} dt'' dt' \quad (106)
\end{aligned}$$

and a second application of the RWA provides

$$\begin{aligned}
\langle f|U_s|i\rangle &= \frac{1}{\hbar^2} \sum_n H_{fn}^{(1)} H_{ni}^{(1)} \frac{1}{\omega_{ni} - \omega_1} \frac{1}{\omega_{fi} - 2\omega_1} \left(e^{it(\omega_{fi} - 2\omega_1)} - 1 \right) \\
&+ H_{fn}^{(1)} H_{ni}^{(2)} \frac{1}{\omega_{ni} - \omega_2} \frac{1}{\omega_{fi} - (\omega_1 + \omega_2)} \left(e^{it(\omega_{fi} - (\omega_1 + \omega_2))} - 1 \right) \\
&+ H_{fn}^{(2)} H_{ni}^{(1)} \frac{1}{\omega_{ni} - \omega_1} \frac{1}{\omega_{fi} - (\omega_1 + \omega_2)} \left(e^{it(\omega_{fi} - (\omega_1 + \omega_2))} - 1 \right) \\
&+ H_{fn}^{(2)} H_{ni}^{(2)} \frac{1}{\omega_{ni} - \omega_2} \frac{1}{\omega_{fi} - 2\omega_2} \left(e^{it(\omega_{fi} - 2\omega_2)} - 1 \right) \quad (107)
\end{aligned}$$

Now we need to consider the probability $P = |\langle f|U_s|i\rangle|^2$. Clearly this will have a large number of cross terms so lets consider a limiting case (worst case): We know we will later include a density of final states $\implies$ the $H_{fn}^{(1)} H_{ni}^{(1)}$ and $H_{fn}^{(2)} H_{ni}^{(2)}$ go to zero. We can further assume that we are optimizing our two photon cross section by getting as close as we can to a particular virtual state so only state will dominate. In other words, we only care about one highly resonant state. We can now simplify the

matrix element $\langle f | U_s | i \rangle$ to:

$$\begin{aligned} \langle f | U_s | i \rangle \approx & \frac{1}{\hbar^2} H_{fn}^{(2)} H_{ni}^{(1)} \frac{1}{\omega_{ni} - \omega_1} \frac{1}{\omega_{fi} - (\omega_1 + \omega_2)} \left(e^{it(\omega_{fi} - (\omega_1 + \omega_2))} - 1 \right) + \\ & \frac{1}{\hbar^2} H_{fn}^{(1)} H_{ni}^{(2)} \frac{1}{\omega_{ni} - \omega_2} \frac{1}{\omega_{fi} - (\omega_1 + \omega_2)} \left(e^{it(\omega_{fi} - (\omega_1 + \omega_2))} - 1 \right). \end{aligned} \quad (108)$$

With this simplified form, and following the approach of Appendix A, the probability $P_{i \rightarrow f}$ can be expressed as

$$\begin{aligned} P_{i \rightarrow f} &= |\langle f | U_s(t_0, t) | i \rangle|^2 \\ &= \frac{1}{\hbar^4} \frac{\left(H_{fn}^{(1)} H_{ni}^{(2)} (\omega_1 - \omega_{ni}) + H_{fn}^{(2)} H_{ni}^{(1)} (\omega_2 - \omega_{ni}) \right)^2}{(\omega_1 - \omega_{ni})^2 (\omega_2 - \omega_{ni})^2} \\ &\quad \times \left(\frac{4 \sin^2[\frac{t}{2} (\omega_{fi} - (\omega_1 + \omega_2))]}{(\omega_{fi} - (\omega_1 + \omega_2))^2} \right). \end{aligned} \quad (109)$$

Again, following the discussion of Appendix A, the transition rate is given in terms of a delta function by:

$$\begin{aligned} R_{i \rightarrow f} &= \lim_{t \rightarrow \infty} \frac{d}{dt} P_{i \rightarrow f} \\ &= \frac{1}{\hbar^4} \frac{\left(H_{fn}^{(1)} H_{ni}^{(2)} (\omega_1 - \omega_{ni}) + H_{fn}^{(2)} H_{ni}^{(1)} (\omega_2 - \omega_{ni}) \right)^2}{(\omega_1 - \omega_{ni})^2 (\omega_2 - \omega_{ni})^2} 2\pi \delta(\omega_{fi} - (\omega_1 + \omega_2)) \\ &= \frac{1}{\hbar^4} \frac{|E_1|^2 |E_2|^2 \left(\mu_{fn}^{(1)} \mu_{ni}^{(2)} (\omega_1 - \omega_{ni}) + \mu_{fn}^{(2)} \mu_{ni}^{(1)} (\omega_2 - \omega_{ni}) \right)^2}{(\omega_1 - \omega_{ni})^2 (\omega_2 - \omega_{ni})^2} \\ &\quad \times 2\pi \delta(\omega_{fi} - (\omega_1 + \omega_2)) \end{aligned} \quad (110)$$

where we note that $|E_1|^2 |E_2|^2$ can be pulled out because they are propagating in the same direction. Next we assume the polarization of $\hat{e}_1 = \hat{e}_2$:

$$\begin{aligned}
R_{i \rightarrow f} &= \frac{1}{\hbar^4} \frac{|E_1|^2 |E_2|^2 |\mu_{fn}|^2 |\mu_{ni}|^2 ((\omega_1 - \omega_{ni}) + (\omega_2 - \omega_{ni}))^2}{(\omega_1 - \omega_{ni})^2 (\omega_2 - \omega_{ni})^2} 2\pi \delta(\omega_{fi} - (\omega_1 + \omega_2)) \\
&= \frac{1}{\hbar^4} \frac{|E_1|^2 |E_2|^2 |\mu_{fn}|^2 |\mu_{ni}|^2 (\omega_1 + \omega_2 - 2\omega_{ni})^2}{(\omega_1 - \omega_{ni})^2 (\omega_2 - \omega_{ni})^2} 2\pi \delta(\omega_{fi} - (\omega_1 + \omega_2)) . \quad (111)
\end{aligned}$$

Next we define the rate $\langle R_{i \rightarrow f}^{(2)(2field)} \rangle = \sigma_{if}^{(2)} \frac{I_1 I_2}{h\nu}$ and include the degeneracies, density of states, and obtain:

$$\sigma_{if}^{(2)}(\nu_1, \nu_2) = \frac{9}{5} \frac{\nu}{256\pi^4 \hbar c^2} \sum_n \frac{g_f}{g_i} \frac{\lambda_{fn}^3 A_{fn} \lambda_{ni}^3 A_{ni} (\nu_1 + \nu_2 - 2\nu_{ni})^2}{(\nu_1 - \nu_{ni})^2 (\nu_2 - \nu_{ni})^2} \rho_f(\nu_{fi} = \nu_1 + \nu_2) \quad (112)$$

Note that ν with no subscripts is now somewhat arbitrary. As a check with the one field result easier, we assume $\omega_1 \approx \omega_2$ (for many of our Cs cases there is only 1 or 2 nm of difference in wavelength) and include the density of final states:

$$\langle R_{i \rightarrow f}^{(2)(2field)} \rangle = \frac{1}{5\hbar^4} \frac{4 |E_1|^2 |E_2|^2 |\mu_{fn}|^2 |\mu_{ni}|^2}{(\omega_1 - \omega_{ni})^2} 2\pi \rho_f(\omega_{fi} = 2\omega_1) = \sigma_{if}^{(2)(2field)} \frac{I_1 I_2}{h\nu} \quad (113)$$

Note that a factor of 4 comes from the difference in the definition of E for the degenerate and non degenerate cases. Converting from angular frequency to frequency we obtain an identical result.

Appendix C. Different Approaches to $S_{FF'}$

In Chapter VII we derived a two-photon hyperfine absorption theory that requires a hyperfine line strength factor. In this work we consider two simultaneous dipole transitions and use

$$S_{F \rightarrow F'} = (2F' + 1) (2J + 1) \left\{ \begin{array}{ccc} J & J' & 2 \\ F' & F & I \end{array} \right\}^2 \quad (114)$$

However, Gallagher [36] and his source for $S_{F \rightarrow F'}$ [65] does not have the same meaning. Nez [65] bases the strength on an electric quadrupole transition stating “the two-photon transition operator is purely quadrupolar in nature.”

$$S_{F \rightarrow F'}^{(Quad)} = \frac{(2F' + 1) (2F + 1)}{(2I + 1)} \left\{ \begin{array}{ccc} J & 2 & J' \\ F' & I & F \end{array} \right\}^2 \quad (115)$$

We believe our equivalent factor is line strength times the Boltzmann factor:

$$S_{F \rightarrow F'} f_F = (2F' + 1) (2J + 1) \left\{ \begin{array}{ccc} J & J' & 2 \\ F' & F & I \end{array} \right\}^2 \frac{(2F + 1) e^{-E(F)/kT}}{\sum_F (2F + 1) e^{-E(F)/kT}} \quad (116)$$

But we note that Gallagher multiplies by f_F by $S_{F \rightarrow F'}^{(Quad)}$ factor in his approach. The 6J symbol is symmetric under permutation of the columns so those terms are the same. Ignoring the exponentials, we note that the two forms agree if

$$\frac{1}{(2I + 1)} = \frac{(2J + 1)}{\sum_F (2F + 1)} \quad (117)$$

Expanding the sum yields

$$\begin{aligned}
\sum_F (2F + 1) &= \sum_{n=0}^{2J} (2[I - J + n] + 1) = (2I + 1)(2J + 1) - 2J(2J + 1) + 2 \sum_{n=0}^{2J} n \\
&= (2I + 1)(2J + 1) - 2J(2J + 1) + 2J(2J + 1) \\
&= (2I + 1)(2J + 1)
\end{aligned} \tag{118}$$

Therefore these approaches are consistent. However, we note that Gallagher [36] and Nez [65] effectively ignore the exponentials within f_F which is not physical.

Bibliography

1. Alexander Akulshin, Dmitry Budker, and Russell McLean. Directional infrared emission resulting from cascade population inversion and four-wave mixing in rb vapor. *Opt. Lett.*, 39(4):845–848, Feb 2014. doi: 10.1364/OL.39.000845. URL <http://ol.osa.org/abstract.cfm?URI=ol-39-4-845>.
2. C. B. Alcock, V. P. Itkin, and M. K. Horrigan. Vapour pressure equations for the metallic elements: 298-2500k. *Canadian Metallurgical Quarterly*, 23(3):309–313, 1984. doi: 10.1179/cmq.1984.23.3.309. URL http://www.physics.nyu.edu/kentlab/How_to/ChemicalInfo/VaporPressure/Pressure.pdf.
3. Y. Bai, N. Bandyopadhyay, S. Tsao, S. Slivken, and M. Razeghi. Room temperature quantum cascade lasers with 27% wall plug efficiency. *Applied Physics Letters*, 98(18):181102, 2011. doi: 10.1063/1.3586773. URL <http://dx.doi.org/10.1063/1.3586773>.
4. H. H. P. T. Bekman, J. C. van den Heuvel, F. J. M. van Putten, and Ric Schlei-jpen. Development of a mid-infrared laser for study of infrared countermeasures techniques, 2004. URL <http://dx.doi.org/10.1117/12.578214>.
5. P. Bernage, P. Niay, and R. Houdart. Stimulated electronic raman scattering in potassium vapour. *Optics Communications*, 36(3):241 – 246, 1981. ISSN 0030-4018. doi: [http://dx.doi.org/10.1016/0030-4018\(81\)90367-9](http://dx.doi.org/10.1016/0030-4018(81)90367-9). URL <http://www.sciencedirect.com/science/article/pii/0030401881903679>.
6. A V Bogachev, Sergey G Garanin, A M Dudov, V A Eroshenko, S M Kulikov, G T Mikaelian, V A Panarin, V O Pautov, A V Rus, and Stanislav A Sukharev. Diode-pumped caesium vapour laser with closed-cycle laser-active medium cir-

- culation. *Quantum Electronics*, 42(2):95, 2012. URL <http://stacks.iop.org/1063-7818/42/i=2/a=A01>.
7. Keith D. Bonin and Thomas J. McIlrath. Two-photon electric-dipole selection rules. *J. Opt. Soc. Am. B*, 1(1):52–55, Mar 1984. doi: 10.1364/JOSAB.1.000052. URL <http://josab.osa.org/abstract.cfm?URI=josab-1-1-52>.
8. K.J. Bowles, United States. National Aeronautics, Space Administration, and Lewis Research Center. *Vapor Pressure of Potassium to 2170 Degrees K*. NASA technical note. National Aeronautics and Space Administration, 1968. URL <https://books.google.com/books?id=z64wuFJffeQC>.
9. Robert W. Boyd. Chapter 12 - optically induced damage and multiphoton absorption. In Robert W. Boyd, editor, *Nonlinear Optics (Third Edition)*, pages 543 – 560. Academic Press, Burlington, third edition edition, 2008. ISBN 978-0-12-369470-6. doi: <http://dx.doi.org/10.1016/B978-0-12-369470-6.00012-5>. URL <http://www.sciencedirect.com/science/article/pii/B9780123694706000125>.
10. Robert W. Boyd. *Non Linear Optics, Third Edition*. Academic Press, Burlington, MA, 2008.
11. Robert W. Boyd, Michelle S. Malcuit, Daniel J. Gauthier, and Kazimierz Rzaewski. Competition between amplified spontaneous emission and the four-wave-mixing process. *Phys. Rev. A*, 35:1648–1658, Feb 1987. doi: 10.1103/PhysRevA.35.1648. URL <https://link.aps.org/doi/10.1103/PhysRevA.35.1648>.
12. D J Bradley, G M Gale, and P D Smith. Stimulated stokes and anti-stokes electronic raman scattering by selectively excited potassium and rubidium atoms.

- Journal of Physics B: Atomic and Molecular Physics*, 4(10):1349, 1971. URL <http://stacks.iop.org/0022-3700/4/i=10/a=020>.
13. Kirk C. Brown and Glen P. Perram. Demonstration of a 459-nm pulsed, optically pumped cesium vapor laser. *Optics Communications*, 300:51 – 57, 2013. ISSN 0030-4018. doi: <https://doi.org/10.1016/j.optcom.2013.03.002>. URL <http://www.sciencedirect.com/science/article/pii/S0030401813002940>.
 14. Kirk C. Brown, Edward J. Hurd, Jeremy C. Holtgrave, and Glen P. Perram. Stimulated electronic raman and hyper-raman scattering in potassium vapor. *Optics Communications*, 309:21 – 25, 2013. ISSN 0030-4018. doi: <http://dx.doi.org/10.1016/j.optcom.2013.07.009>. URL <http://www.sciencedirect.com/science/article/pii/S0030401813006408>.
 15. Yao-Ju Chan and Wei-Tzou Luh. Six-wave mixing near-infrared emissions in rubidium vapor. *Chinese Journal of Physics*, 33(3):247–261, Jun 1995.
 16. B. K. Clark, M. Masters, and J. Huennekens. Wave mixing and amplified spontaneous emission in pure potassium and mixed sodium-potassium vapors. *Applied Physics B*, 47(2):159–167, 1988. ISSN 1432-0649. doi: [10.1007/BF00684083](https://doi.org/10.1007/BF00684083). URL <http://dx.doi.org/10.1007/BF00684083>.
 17. C. L. A. Collins, K. D. Bonin, and M. A. Kadar-Kallen. Absolute two-photon cross section of rb measured by differential absorption. *Opt. Lett.*, 18(20): 1754–1756, Oct 1993. doi: [10.1364/OL.18.001754](https://doi.org/10.1364/OL.18.001754). URL <http://ol.osa.org/abstract.cfm?URI=ol-18-20-1754>.
 18. Hope M. Concannon and Daniel J. Gauthier. Simple rate-equation model for two-photon lasers. *Opt. Lett.*, 19(7):472–474, Apr 1994. doi: [10.1364/OL.19.000472](https://doi.org/10.1364/OL.19.000472). URL <http://ol.osa.org/abstract.cfm?URI=ol-19-7-472>.

19. Hope M. Concannon, William J. Brown, Jeff R. Gardner, and Daniel J. Gauthier. Observation of large continuous-wave two-photon optical amplification. *Phys. Rev. A*, 56:1519–1523, Aug 1997. doi: 10.1103/PhysRevA.56.1519. URL <https://link.aps.org/doi/10.1103/PhysRevA.56.1519>.
20. Hope M. Concannon, William J. Brown, Jeff R. Gardner, and Daniel J. Gauthier. Observation of large continuous-wave two-photon optical amplification. *Phys. Rev. A*, 56:1519–1523, Aug 1997. doi: 10.1103/PhysRevA.56.1519. URL <http://link.aps.org/doi/10.1103/PhysRevA.56.1519>.
21. Tiffany L. Correll, Vlasta Horvatic, Nicolo Omenetto, James D. Winefordner, and Cedomil Vadla. Experimental evaluation of the cross-sections for the $\text{cs}(6d) - \text{cs}(7p_j)$ and $\text{cs}(6d5/2) - \text{cs}(6d3/2)$ collisional transfer processes induced by he and ar. *Spectrochimica Acta Part B: Atomic Spectroscopy*, 61(6):623 – 633, 2006. ISSN 0584-8547. doi: <https://doi.org/10.1016/j.sab.2005.08.002>. URL <http://www.sciencedirect.com/science/article/pii/S0584854705002156>. Special Honor Issue Dedicated to the memory of Prof. Howard V. Malmstadt.
22. D. Cotter, D.C. Hanna, P.A. Kärkkäinen, and R. Wyatt. Stimulated electronic raman scattering as a tunable infrared source. *Optics Communications*, 15(2):143 – 146, 1975. ISSN 0030-4018. doi: [http://dx.doi.org/10.1016/0030-4018\(75\)90270-9](http://dx.doi.org/10.1016/0030-4018(75)90270-9). URL <http://www.sciencedirect.com/science/article/pii/0030401875902709>.
23. D. Cotter, D.C. Hanna, and R. Wyatt. A high power, widely tunable infrared source based on stimulated electronic raman scattering in caesium vapour. *Optics Communications*, 16(2):256 – 258, 1976. ISSN 0030-4018. doi: [http://dx.doi.org/10.1016/0030-4018\(76\)90230-3](http://dx.doi.org/10.1016/0030-4018(76)90230-3). URL <http://www.sciencedirect.com/science/article/pii/0030401876902303>.

24. D. Cotter, D.C. Hanna, W.H.W. Tuttlebee, and M.A. Yuratich. Stimulated hyper-raman emission from sodium vapour. *Optics Communications*, 22(2):190 – 194, 1977. ISSN 0030-4018. doi: [http://dx.doi.org/10.1016/0030-4018\(77\)90017-7](http://dx.doi.org/10.1016/0030-4018(77)90017-7). URL <http://www.sciencedirect.com/science/article/pii/0030401877900177>.
25. D.P. Craig and T. Thirunamachandran. *Molecular Quantum Electrodynamics: An Introduction to Radiation-molecule Interactions*. Dover Books on Chemistry Series. Dover Publications, 1984. ISBN 9780486402147. URL <https://books.google.com/books?id=rpbdozIZt3sC>.
26. D.P. Craig and T. Thirunamachandran. *Molecular Quantum Electrodynamics: An Introduction to Radiation-molecule Interactions*. Dover Books on Chemistry Series. Dover Publications, 1984. ISBN 9780486402147. URL <https://books.google.com/books?id=rpbdozIZt3sC>.
27. J. Cuvellier, P. R. Fournier, F. Gounand, J. Pascale, and J. Berlande. Inelastic collisions involving excited cesium atoms at thermal energies. *Phys. Rev. A*, 11: 846–856, Mar 1975. doi: 10.1103/PhysRevA.11.846. URL <https://link.aps.org/doi/10.1103/PhysRevA.11.846>.
28. "Ryan McGlen David Reay", "Peter Kew". *Heat Pipes: Theory, Design and Applications sixth edition*. Butterworth-Heinemann, Oxford, UK, 2014.
29. D. DiBerardino, C. E. Tanner, and A. Sieradzan. Lifetime measurements of cesium $5d^2d_{5/2,3/2}$ and $11s^2s_{1/2}$ states using pulsed-laser excitation. *Phys. Rev. A*, 57:4204–4211, Jun 1998. doi: 10.1103/PhysRevA.57.4204. URL <https://link.aps.org/doi/10.1103/PhysRevA.57.4204>.
30. Mikhail Drobizhev, Aliaksandr Karotki, Aleksander Rebane, and Charles W.

- Spangler. Dendrimer molecules with record large two-photon absorption cross section. *Opt. Lett.*, 26(14):1081–1083, Jul 2001. doi: 10.1364/OL.26.001081. URL <http://ol.osa.org/abstract.cfm?URI=ol-26-14-1081>.
31. K B S Eriksson and I WenÅŸker. New wavelength measurements in cs i. *Physica Scripta*, 1(1):21, 1970. URL <http://stacks.iop.org/1402-4896/1/i=1/a=003>.
 32. Karl B rje Eriksson, JOHANSSO. I, and G Norlen. Precision wavelength measurements connecting cs i 6s 6p and 6d levels with a study of correction for phase change in infrared interferometry. *ARKIV FOR FYSIK*, 28(3):233–+, 1965.
 33. J. L. Font, J. J. Fern ndez-Soler, R. Vilaseca, and Daniel J. Gauthier. Multiphoton lasing in atomic potassium: Steady-state and dynamic behavior. *Phys. Rev. A*, 72:063810, Dec 2005. doi: 10.1103/PhysRevA.72.063810. URL <http://link.aps.org/doi/10.1103/PhysRevA.72.063810>.
 34. Baodong Gai, Rui Cao, Xusheng Xia, Shu Hu, Jinbo Liu, Jingwei Guo, Yannan Tan, Wanfa Liu, Yuqi Jin, and Fengting Sang. Modulation of a double-line frequency up-conversion process in cesium vapor. *Applied Physics B*, 122(6):165, 2016. ISSN 1432-0649. doi: 10.1007/s00340-016-6438-4. URL <http://dx.doi.org/10.1007/s00340-016-6438-4>.
 35. Baodong Gai, Rui Cao, Xusheng Xia, Shu Hu, Jinbo Liu, Jingwei Guo, Yuqi Jin, and Fengting Sang. Influence of pumping power density on output intensities and stabilities of competitive multi-line frequency up-conversions in cesium vapor. *Optics and Laser Technology*, 95:18–22, 2017. doi: 10.1016/j.optlastec.2017.03.044.
 36. Jeffrey Gallagher and Glen Perram. chapter Determining the Two-Photon Ab-

sorption Cross-Section for the $5^2S_{1/2} \rightarrow 5^2D_{5/2}$ Transition in Naturally Occurring Rubidium. Fluid Dynamics and Co-located Conferences. American Institute of Aeronautics and Astronautics, Jun 2011. doi: 10.2514/6.2011-4005. URL <http://dx.doi.org/10.2514/6.2011-4005>. 0.

37. Daniel J. Gauthier. Chapter 4 - two-photon lasers. volume 45 of *Progress in Optics*, pages 205 – 272. Elsevier, 2003. doi: [https://doi.org/10.1016/S0079-6638\(03\)80006-9](https://doi.org/10.1016/S0079-6638(03)80006-9). URL <http://www.sciencedirect.com/science/article/pii/S0079663803800069>.
38. Daniel J. Gauthier, Qilin Wu, S. E. Morin, and T. W. Mossberg. Realization of a continuous-wave, two-photon optical laser. *Phys. Rev. Lett.*, 68:464–467, Jan 1992. doi: 10.1103/PhysRevLett.68.464. URL <http://link.aps.org/doi/10.1103/PhysRevLett.68.464>.
39. Daniel J. Gauthier, Qilin Wu, S. E. Morin, and T. W. Mossberg. Realization of a continuous-wave, two-photon optical laser. *Phys. Rev. Lett.*, 68:464–467, Jan 1992. doi: 10.1103/PhysRevLett.68.464. URL <https://link.aps.org/doi/10.1103/PhysRevLett.68.464>.
40. V. Gerginov, C. E. Tanner, S. A. Diddams, A. Bartels, and L. Hollberg. High-resolution spectroscopy with a femtosecond laser frequency comb. *Opt. Lett.*, 30(13):1734–1736, Jul 2005. doi: 10.1364/OL.30.001734. URL <http://ol.osa.org/abstract.cfm?URI=ol-30-13-1734>.
41. V. Gerginov, K. Calkins, C. E. Tanner, J. J. McFerran, S. Diddams, A. Bartels, and L. Hollberg. Optical frequency measurements of $6s_{1/2} - 6p_{1/2}$ transitions in cs and their impact on the fine-structure constant. *Phys. Rev. A*, 73:032504, Mar 2006. doi: 10.1103/PhysRevA.73.032504. URL <https://link.aps.org/doi/10.1103/PhysRevA.73.032504>.

42. Maria Göppert-Mayer. über die wahrscheinlichkeit des zusammenwirkens zweier lichtquanten in einem elementarakt. *Naturwissenschaften*, 17(48):932–932, 1929. ISSN 0028-1042. doi: 10.1007/BF01506585. URL <http://dx.doi.org/10.1007/BF01506585>.
43. Maria Göppert-Mayer. über elementarakte mit zwei quantensprüngen. *Annalen der Physik*, 401(3):273–294, 1931. ISSN 1521-3889. doi: 10.1002/andp.19314010303. URL <http://dx.doi.org/10.1002/andp.19314010303>.
44. N. D. Haluska and G. P. Perram. Draft publication: Efficient cascade lasing on over 17 wavelengths from a two-photon pumped cesium 6d manifold. *Optics Communications*, 2017.
45. S. M. Hamadani, J. A. D. Stockdale, R. N. Compton, and M. S. Pindzola. Two-photon resonant four-wave mixing and multiphoton ionization of cesium in a heat-pipe oven. *Phys. Rev. A*, 34:1938–1943, Sep 1986. doi: 10.1103/PhysRevA.34.1938. URL <https://link.aps.org/doi/10.1103/PhysRevA.34.1938>.
46. W. Hartig. Two-photon resonant four-wave mixing in atomic sodium vapour. *Applied physics*, 15(4):427–432, 1978. ISSN 1432-0630. doi: 10.1007/BF00886163. URL <http://dx.doi.org/10.1007/BF00886163>.
47. W.M. Haynes. *CRC Handbook of Chemistry and Physics, 93rd Edition*. CRC Handbook of Chemistry and Physics. Taylor & Francis, 2012. ISBN 9781439880494. URL <https://books.google.com/books?id=-BzP7Rkl7WkC>.
48. O. S. Heavens. Radiative transition probabilities of the lower excited states of the alkali metals. *J. Opt. Soc. Am.*, 51(10):1058–1061, Oct 1961. doi: 10.1364/JOSA.51.001058. URL <http://www.osapublishing.org/abstract.cfm?URI=josa-51-10-1058>.

49. R. T. Hodgson. Stimulated electronic raman scattering from the $6s\ 2s_{1/2}$ to the $5d\ 2d_{5/2}$ state of cesium. *Applied Physics Letters*, 34(1):58–60, 1979. doi: 10.1063/1.90604. URL <http://dx.doi.org/10.1063/1.90604>.
50. Z. J. Jabbour, M. S. Malcuit, and J. Huennekens. Broadly tunable near-infrared six-wave mixing processes in potassium vapor. *Applied Physics B*, 52(4):281–289, 1991. ISSN 1432-0649. doi: 10.1007/BF00325406. URL <http://dx.doi.org/10.1007/BF00325406>.
51. DJ Jackson and JJ Wynne. What is the signature of a 2-photon laser. In *APPLIED PHYSICS B-PHOTOPHYSICS AND LASER CHEMISTRY*, volume 28, pages 238–238. SPRINGER VERLAG 175 FIFTH AVE, NEW YORK, NY 10010, 1982.
52. F. A. Korolev, V. V. Martynov, V. I. Odintsov, and A. O. Fakhmi. Stimulated and parametric emission in Rb vapor in two photon excitation of $5^2D_{3/2,5/2}$ and $7^2S_{1/2}$ levels. *Optics and Spectroscopy*, 40:600–603, June 1976.
53. A Kortyna, NA Masluk, and T Bragdon. Measurement of the $6\ d\ d\ j\ 2$ hyperfine structure of cesium using resonant two-photon sub-doppler spectroscopy. *Physical Review A*, 74(2):022503, 2006.
54. W.F. Krupke. Diode-pumped alkali laser, November 4 2003. US Patent 6,643,311.
55. William F. Krupke, Raymond J. Beach, V. Keith Kanz, and Stephen A. Payne. Resonance transition 795-nm rubidium laser. *Opt. Lett.*, 28(23):2336–2338, Dec 2003. doi: 10.1364/OL.28.002336.
56. P. Lambropoulos. Quantum statistics of a two-photon quantum amplifier. *Phys.*

- Rev.*, 156:286–297, Apr 1967. doi: 10.1103/PhysRev.156.286. URL <http://link.aps.org/doi/10.1103/PhysRev.156.286>.
57. V. Letokhov and V. Chebotayev. *Nonlinear Laser Spectroscopy*. Springer-Verlag Berlin Heidelberg, Germany, 1977.
 58. R. Loudon. *The Quantum Theory of Light*. OUP Oxford, 2000. ISBN 9780191589782. URL <https://books.google.com/books?id=AEkfajgqldoC>.
 59. M. M. T. Loy. Two-photon adiabatic inversion. *Phys. Rev. Lett.*, 41:473–476, Aug 1978. doi: 10.1103/PhysRevLett.41.473. URL <http://link.aps.org/doi/10.1103/PhysRevLett.41.473>.
 60. W. T. Luh, Yan Li, and J. Huennekens. 830 nm emission in sodium vapor. *Applied Physics B*, 49(4):349–359, 1989. ISSN 1432-0649. doi: 10.1007/BF00324185. URL <http://dx.doi.org/10.1007/BF00324185>.
 61. O. Lumpkin. Four-wave parametric interaction in potassium vapor. *IEEE Journal of Quantum Electronics*, 4(4):226–228, Apr 1968. ISSN 0018-9197. doi: 10.1109/JQE.1968.1075076.
 62. Berry R. Masters. Proximity of theory and experimental practice in göttingen’s physical institutes 1920-1933: Work of maria göppert as exemplar of an institutional culture, June 2010. URL http://quantum-history.mpiwg-berlin.mpg.de/news/workshops/hq3/hq3_talks/21_masters_appendix.pdf.
 63. T. Meijer, J. D. White, B. Smeets, M. Jeppesen, and R. E. Scholten. Blue five-level frequency-upconversion system in rubidium. *Opt. Lett.*, 31(7):1002–1004, Apr 2006. doi: 10.1364/OL.31.001002. URL <http://ol.osa.org/abstract.cfm?URI=ol-31-7-1002>.

64. P. J. Moran, R. M. Richards, C. A. Rice, and G. P. Perram. Near infrared rubidium $6^2P_{3/2,1/2} \rightarrow 6^2S_{1/2}$ laser. *Optics Communications*, 374:51–57, September 2016. doi: 10.1016/j.optcom.2016.03.090.
65. F. Nez, F. Biraben, R. Felder, and Y. Millerioux. Optical frequency determination of the hyperfine components of the $5s12-5d32$ two-photon transitions in rubidium. *Optics Communications*, 102(5):432 – 438, 1993. ISSN 0030-4018. doi: [http://dx.doi.org/10.1016/0030-4018\(93\)90417-4](http://dx.doi.org/10.1016/0030-4018(93)90417-4). URL <http://www.sciencedirect.com/science/article/pii/0030401893904174>.
66. P. Niay, P. Bernage, and H. Bocquet. Stimulated electronic raman scattering in rubidium vapour. *Optics Communications*, 29(3):369 – 374, 1979. ISSN 0030-4018. doi: [http://dx.doi.org/10.1016/0030-4018\(79\)90120-2](http://dx.doi.org/10.1016/0030-4018(79)90120-2). URL <http://www.sciencedirect.com/science/article/pii/0030401879901202>.
67. B. Nikolaus, D. Z. Zhang, and P. E. Toschek. Two-photon laser. *Phys. Rev. Lett.*, 47:171–173, Jul 1981. doi: 10.1103/PhysRevLett.47.171. URL <http://link.aps.org/doi/10.1103/PhysRevLett.47.171>.
68. B.J. Orr and J.F. Ward. Perturbation theory of the non-linear optical polarization of an isolated system. *Molecular Physics*, 20(3):513–526, 1971. doi: 10.1080/00268977100100481. URL <http://dx.doi.org/10.1080/00268977100100481>.
69. Yasutomo Ota, Satoshi Iwamoto, Naoto Kumagai, and Yasuhiko Arakawa. Spontaneous two-photon emission from a single quantum dot. *Phys. Rev. Lett.*, 107:233602, Nov 2011. doi: 10.1103/PhysRevLett.107.233602. URL <http://link.aps.org/doi/10.1103/PhysRevLett.107.233602>.
70. Paul W. Pace and J. B. Atkinson. Transfer of electronic excitation between the

- 72p $1/2$ and 72p $3/2$ states of cesium induced by collisions with cesium atoms. *Canadian Journal of Physics*, 52(17):1641–1647, 1974. doi: 10.1139/p74-214.
71. R. Payling, P. Larkins, and T. J. Vickers. Optical Emission Lines of the Elements. *Applied Spectroscopy*, 55:295, September 2001.
 72. W. Scott Pegau, Deric Gray, and J. Ronald V. Zaneveld. Absorption and attenuation of visible and near-infrared light in water: dependence on temperature and salinity. *Appl. Opt.*, 36(24):6035–6046, Aug 1997. doi: 10.1364/AO.36.006035. URL <http://ao.osa.org/abstract.cfm?URI=ao-36-24-6035>.
 73. Glen P. Perram. Wavelength diversity in optically pumped alkali vapor lasers, 2017. URL <http://dx.doi.org/10.1117/12.2256256>.
 74. Olivier Pfister, William J. Brown, Michael D. Stenner, and Daniel J. Gauthier. Two-photon stimulated emission in laser-driven alkali-metal atoms using an orthogonal pump-probe geometry. *Phys. Rev. A*, 60:R4249–R4252, Dec 1999. doi: 10.1103/PhysRevA.60.R4249. URL <http://link.aps.org/doi/10.1103/PhysRevA.60.R4249>.
 75. Greg A. Pitz, Donald M. Stalnaker, Eric M. Guild, Benjamin Q. Olier, Paul J. Moran, Steven W. Townsend, and David A. Hostutler. Advancements in flowing diode pumped alkali lasers, 2016. URL <http://dx.doi.org/10.1117/12.2217078>.
 76. B. J. Pong, J. E. Fan, C. C. Chu, W. T. Cheng, and T. S. Yih. Six wave mixing generated 722 nm emissions in sodium vapor. *Chinese Journal of Physics*, 34(2):122–129, Apr 1996.
 77. A. M. Prokhorov. Quantum Electronics. *Science*, 149:828–830, August 1965. doi: 10.1126/science.149.3686.828.

78. P. Rabinowitz, S. Jacobs, and G. Gould. Continuous optically pumped cs laser. *Appl. Opt.*, 1(4):513–516, Jul 1962. doi: 10.1364/AO.1.000513. URL <http://ao.osa.org/abstract.cfm?URI=ao-1-4-513>.
79. R. J. Rafac, C. E. Tanner, A. E. Livingston, K. W. Kukla, H. G. Berry, and C. A. Kurtz. Precision lifetime measurements of the $6p^2p_{1/2,3/2}$ states in atomic cesium. *Phys. Rev. A*, 50:R1976–R1979, Sep 1994. doi: 10.1103/PhysRevA.50.R1976. URL <https://link.aps.org/doi/10.1103/PhysRevA.50.R1976>.
80. M. Rokni and S. Yatsiv. Stimulated electronic raman effect and parametric anti-stokes radiation in potassium vapor. *IEEE Journal of Quantum Electronics*, 3(7):329–330, July 1967. ISSN 0018-9197. doi: 10.1109/JQE.1967.1074595.
81. M. S. Safronova and Charles W. Clark. Inconsistencies between lifetime and polarizability measurements in Cs. *Phys. Rev. A*, 69:040501, Apr 2004. doi: 10.1103/PhysRevA.69.040501. URL <https://link.aps.org/doi/10.1103/PhysRevA.69.040501>.
82. Kasturi Saha, Vivek Venkataraman, Pablo Londero, and Alexander L. Gaeta. Enhanced two-photon absorption in a hollow-core photonic-band-gap fiber. *Phys. Rev. A*, 83:033833, Mar 2011. doi: 10.1103/PhysRevA.83.033833. URL <http://link.aps.org/doi/10.1103/PhysRevA.83.033833>.
83. A. L. Schawlow and C. H. Townes. Infrared and optical masers. *Phys. Rev.*, 112:1940–1949, Dec 1958. doi: 10.1103/PhysRev.112.1940. URL <https://link.aps.org/doi/10.1103/PhysRev.112.1940>.
84. J. T. Schultz, S. Abend, D. Döring, J. E. Debs, P. A. Altin, J. D. White, N. P. Robins, and J. D. Close. Coherent 455 nm beam production in a cesium

- vapor. *Opt. Lett.*, 34(15):2321–2323, Aug 2009. doi: 10.1364/OL.34.002321. URL <http://ol.osa.org/abstract.cfm?URI=ol-34-15-2321>.
85. J. T. Schultz, S. Abend, D. Döring, J. E. Debs, P. A. Altin, J. D. White, N. P. Robins, and J. D. Close. Coherent 455 nm beam production in a cesium vapor. *Opt. Lett.*, 34(15):2321–2323, Aug 2009. doi: 10.1364/OL.34.002321. URL <http://ol.osa.org/abstract.cfm?URI=ol-34-15-2321>.
 86. J. F. Sell, M. A. Gearba, B. D. DePaola, and R. J. Knize. Collimated blue and infrared beams generated by two-photon excitation in rb vapor. *Opt. Lett.*, 39(3):528–531, Feb 2014. doi: 10.1364/OL.39.000528. URL <http://ol.osa.org/abstract.cfm?URI=ol-39-3-528>.
 87. R. Shankar. *Physics of Quantum Mechanics, Second Edition*. Springer, New York, NY, 2008.
 88. A. Sharma, N. D. Bhaskar, Y. Q. Lu, and W. Happer. Continuous wave mirrorless lasing in optically pumped atomic cs and rb vapors. *Applied Physics Letters*, 39(3):209–211, 1981. doi: 10.1063/1.92681.
 89. Anthony E. Siegman. *Lasers*. University Science Books, Palo Alto, CA, 1986.
 90. P. Sorokin and J. Lankard. Efficient parametric conversion in cesium vapor irradiated by 3470-a mode-locked pulses. *IEEE Journal of Quantum Electronics*, 9(2):227–230, February 1973. ISSN 0018-9197. doi: 10.1109/JQE.1973.1077452.
 91. P. P. Sorokin and N. Braslau. Some theoretical aspects of a proposed double quantum stimulated emission device. *IBM J. Res. Dev.*, 8(2):177–181, April 1964. ISSN 0018-8646. doi: 10.1147/rd.82.0177. URL <http://dx.doi.org/10.1147/rd.82.0177>.

92. P. P. Sorokin, N. S. Shiren, J. R. Lankard, E. C. Hammond, and T. G. Kazyaka. Stimulated electronic raman scattering. *Applied Physics Letters*, 10(2):44–46, 1967. doi: 10.1063/1.1754839. URL <http://dx.doi.org/10.1063/1.1754839>.
93. P.P. Sorokin and N. Braslau. Some theoretical aspects of a proposed double quantum stimulated emission device. *IBM Journal of Research and Development*, 8(2):177–181, April 1964. ISSN 0018-8646. doi: 10.1147/rd.82.0177.
94. J Sparbier, K-J Boller, and PE Toschek. Two-photon emission competing with cooperative and nonlinear processes. *Applied Physics B: Lasers and Optics*, 62(3):227–234, 1996.
95. Daniel A. Steck. Cesium d line data, December 2010. URL <http://steck.us/alkalidata/cesiumnumbers.pdf>.
96. Daniel A. Steck. Rubidium 85 d line data, 2013. URL <http://steck.us/alkalidata/rubidium85numbers.pdf>.
97. C. V. Sulham, G. A. Pitz, and G. P. Perram. Blue and infrared stimulated emission from alkali vapors pumped through two-photon absorption. *Applied Physics B*, 101(1):57–63, 2010. ISSN 1432-0649. doi: 10.1007/s00340-010-4015-9.
98. A. C. Tam, T. Yabuzaki, S. M. Curry, M. Hou, and W. Happer. Inelastic cross sections in $\text{Cs}(n^2d_J) + \text{Cs}(6^2s_{\frac{1}{2}})$ collisions. *Phys. Rev. A*, 17:1862–1868, Jun 1978. doi: 10.1103/PhysRevA.17.1862. URL <https://link.aps.org/doi/10.1103/PhysRevA.17.1862>.
99. Constantine E. Theodosiou. Lifetimes of alkali-metal—atom rydberg states. *Phys. Rev. A*, 30:2881–2909, Dec 1984. doi: 10.1103/PhysRevA.30.2881. URL <http://link.aps.org/doi/10.1103/PhysRevA.30.2881>.

100. A. A. Vasilyev, I. M. Savukov, M. S. Safronova, and H. G. Berry. Measurement of the $6s-7p$ transition probabilities in atomic cesium and a revised value for the weak charge Q_W . *Phys. Rev. A*, 66:020101, Aug 2002. doi: 10.1103/PhysRevA.66.020101. URL <https://link.aps.org/doi/10.1103/PhysRevA.66.020101>.
101. A. Vernier, S. Franke-Arnold, E. Riis, and A. S. Arnold. Enhanced frequency up-conversion in rb vapor. *Opt. Express*, 18(16):17020–17026, Aug 2010. doi: 10.1364/OE.18.017020.
102. Q.H.F. Vrehen and H.M.J. Hikspoors. Two-photon induced stimulated raman scattering in cesium vapor. *Optics Communications*, 21(1):127 – 131, 1977. ISSN 0030-4018. doi: [http://dx.doi.org/10.1016/0030-4018\(77\)90092-X](http://dx.doi.org/10.1016/0030-4018(77)90092-X). URL <http://www.sciencedirect.com/science/article/pii/003040187790092X>.
103. G. Walker, A. S. Arnold, and S. Franke-Arnold. Trans-spectral orbital angular momentum transfer via four-wave mixing in rb vapor. *Phys. Rev. Lett.*, 108: 243601, Jun 2012. doi: 10.1103/PhysRevLett.108.243601. URL <https://link.aps.org/doi/10.1103/PhysRevLett.108.243601>.
104. K.-H. Weber and Craig J. Sansonetti. Accurate energies of ns, np, nd, nf, and ng levels of neutral cesium. *Phys. Rev. A*, 35:4650–4660, Jun 1987. doi: 10.1103/PhysRevA.35.4650. URL <https://link.aps.org/doi/10.1103/PhysRevA.35.4650>.
105. R. Wyatt and D. Cotter. Tunable infrared generation using the $6s-5d$ raman transition in caesium vapour. *Applied physics*, 21(3):199–204, 1980. ISSN 1432-0630. doi: 10.1007/BF00886169. URL <http://dx.doi.org/10.1007/BF00886169>.
106. R. Wyatt and D. Cotter. A tunable picosecond IR laser generating multi-

- megawatt pulses in the range 3-8 microns. *Optics Communications*, 37:421–425, June 1981. doi: 10.1016/0030-4018(81)90134-6.
107. J J Wynne and P P Sorokin. Optically pumped stimulated emission and stimulated electronic raman scattering from k atoms. *Journal of Physics B: Atomic and Molecular Physics*, 8(4):L37, 1975. URL <http://stacks.iop.org/0022-3700/8/i=4/a=002>.
 108. Chris Xu and Watt W. Webb. Measurement of two-photon excitation cross sections of molecular fluorophores with data from 690 to 1050 nm. *J. Opt. Soc. Am. B*, 13(3):481–491, Mar 1996. doi: 10.1364/JOSAB.13.000481. URL <http://josab.osa.org/abstract.cfm?URI=josab-13-3-481>.
 109. W. Zapka, M. D. Levenson, F. M. Schellenberg, A. C. Tam, and G. C. Bjorklund. Continuous-wave doppler-free two-photon frequency-modulation spectroscopy in rb vapor. *Opt. Lett.*, 8(1):27–29, Jan 1983. doi: 10.1364/OL.8.000027. URL <http://ol.osa.org/abstract.cfm?URI=ol-8-1-27>.
 110. P.-L. Zhang, Y.-C. Wang, and A. L. Schawlow. Generation of coherent uv radiation by optical wave-mixing processes in atomic potassium. *J. Opt. Soc. Am. B*, 1(1):9–14, Mar 1984. doi: 10.1364/JOSAB.1.000009. URL <http://josab.osa.org/abstract.cfm?URI=josab-1-1-9>.
 111. Yifu Zhu, Qilin Wu, S. Morin, and T. W. Mossberg. Observation of a two-photon gain feature in the strong-probe absorption spectrum of driven two-level atoms. *Phys. Rev. Lett.*, 65:1200–1203, Sep 1990. doi: 10.1103/PhysRevLett.65.1200. URL <http://link.aps.org/doi/10.1103/PhysRevLett.65.1200>.
 112. A. S. Zibrov, M. D. Lukin, D. E. Nikonov, L. Hollberg, M. O. Scully, V. L. Velichansky, and H. G. Robinson. Experimental demonstration of laser os-

- cillation without population inversion via quantum interference in rb. *Phys. Rev. Lett.*, 75:1499–1502, Aug 1995. doi: 10.1103/PhysRevLett.75.1499. URL <https://link.aps.org/doi/10.1103/PhysRevLett.75.1499>.
113. A. S. Zibrov, L. Hollberg, V. L. Velichansky, M. O. Scully, M. D. Lukin, H. G. Robinson, A. B. Matsko, A. V. Taichenachev, and V. I. Yudin. Destruction of darkness: Optical coherence effects and multi-wave mixing in rubidium vapor. *AIP Conference Proceedings*, 551(1):204–217, 2001. doi: 10.1063/1.1354350. URL <http://aip.scitation.org/doi/abs/10.1063/1.1354350>.
114. A. S. Zibrov, M. D. Lukin, L. Hollberg, and M. O. Scully. Efficient frequency up-conversion in resonant coherent media. *Phys. Rev. A*, 65:051801, Apr 2002. doi: 10.1103/PhysRevA.65.051801. URL <https://link.aps.org/doi/10.1103/PhysRevA.65.051801>.

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