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Final Technical Report
June 1980

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RESEARCH STUDIES ON RADIATIVE COLLISION LASERS

Stanford University

S. E. Harris
J. F. Young

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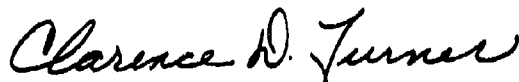
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19. KEY WORDS (Continue on reverse side if necessary and identify by block number) Lasers Radiative Collisions Laser Induced Collisions			
20. ABSTRACT (Continue on reverse side if necessary and identify by block number) This final report reviews the results of a program of theoretical and experimental studies of the physics and applications of radiative collisional processes involving two atoms and a photon simultaneously.			

1. INTRODUCTION

This contract has supported a program of theoretical and experimental studies of the physics and applications of radiative collisional processes. A radiative collisional process is one in which two atoms and a photon simultaneously participate. In one type of process, termed a laser induced or laser switched collision, energy is first stored in a state of some atom A. An incident laser is tuned so as to satisfy the energy defect between this storage state and a selected target state of a different atomic species B. In the presence of this incident laser beam, the collision cross section may be as large as 10^{-13} cm^2 , thereby allowing very rapid transfer of energy to the target species.

If the storage state of atom A is at a higher energy than that of the target state of atom B, then an incident laser photon will experience gain rather than loss, and stimulated emission may then occur at a characteristic frequency equal to the difference of the energy levels of the two species. In the visible, the gain linewidth for lasers of this type will generally be a few angstroms. The gain cross section will be quite low, but once stimulated emission is obtained, the energy storage will be very high.

If in a mixture of two species, more ions of higher ionization potential can be created (For example, by a selective photoionization process), then lasing may occur at a frequency characteristic of the difference of the ionization potentials of the two species. Such a radiative charge transfer laser would, again, have low gain and very high energy storage.

The collisional processes studied may also lead to a class of active upconverters, where upconversion gain is obtained from an electrically excited medium. For example, energy stored in an e-beam pumped inert gas might be used to efficiently upconvert CO_2 radiation into the visible.

Pair absorption processes are closely related to laser induced collisions and may provide a means for making new lasers.

Our research results on these topics are summarized in Section 2. Appendix A consists of the abstracts or title pages of the publications supported by this contract, and Appendix B lists our students who received a Ph.D. degree during the period of this work.

We note that much of this program is continuing under joint support of the Army Research Office and the Air Force Office of Scientific Research.

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2. SUMMARY OF RESEARCH RESULTS

A. Laser Induced Collisions

The laser induced collision process was predicted in 1972,^{1,2} and first demonstrated experimentally by Harris, et al. in 1976.³ Following that initial work, this program has supported efforts which have greatly expanded the state of knowledge of such effects, both experimentally and theoretically.

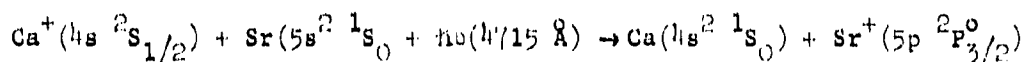
A theoretical analysis of laser induced collisions was developed⁴ to describe the linewidth, lineshape, and magnitude of laser induced collisions. The theory predicts that for laser induced collisional processes where the interaction Hamiltonian of the colliding species is of the dipole-dipole or dipole-quadrupole type, the cross section for laser induced collision should peak when the transfer laser is tuned to that wavelength (λ_{Reson}) which exactly satisfies energy conservation for the infinitely separated atoms. For quadrupole-quadrupole, charge exchange, and spin exchange processes, the peak should occur near to, but not at, the energy conserving wavelength, and could be much broader.

A number of these predictions were subsequently verified experimentally. Further details on the first dipole-dipole Sr-Ca system confirmed that very large (10^{-13} cm^2) collision cross sections were possible using practical sources.^{5,6} In addition, the first experimental observations of two new types of laser induced collisions were made: dipole-quadrupole⁷ and charge exchange.⁸ The dipole-quadrupole process is important because it expands

significantly the number of energy storage states which may be considered in designing a practical system. As expected, the process maximized at the $\lambda_{R=\infty}$ wavelength, and was nearly as strong as the dipole-dipole case.

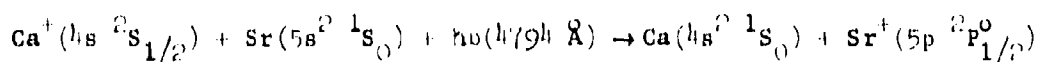
Charge exchange is of particular interest because the storage species can be the ground state of an ion. An intense laser field is then used to rapidly transfer this energy to an excited ionic state of a second species. Since ground state ions can be easily and efficiently created by a number of means, and represent relatively long-lived energy storage, laser induced charge transfer processes are particularly exciting for applications.

The first laser induced charge transfer collision which was observed⁸ is



An energy level diagram representing this reaction is shown in Fig. 1. In this system, the applied laser photon raises one of the two Sr valence electrons to a virtual state of $\text{Sr}(5\text{s}5\text{p } ^1\text{P}_1^o)$ character. As this Sr atom collides with a Ca^+ ion, the remaining unexcited Sr valence electron is transferred to the Ca^+ ion, leaving a Ca ground state neutral and an excited $\text{Sr}^+(5\text{p } ^2\text{P}_{3/2}^o)$ ion. Theory predicts that the transfer maximizes when the applied photon is detuned from $\lambda_{R=\infty}$. We estimate that in our experiments we induced a cross section of $5 \times 10^{-15} \text{ cm}^2$ at an applied laser power density at 4715 Å of $1.5 \times 10^9 \text{ W/cm}^2$.

Just recently, we believe that we have demonstrated a second laser induced charge transfer collision, represented by



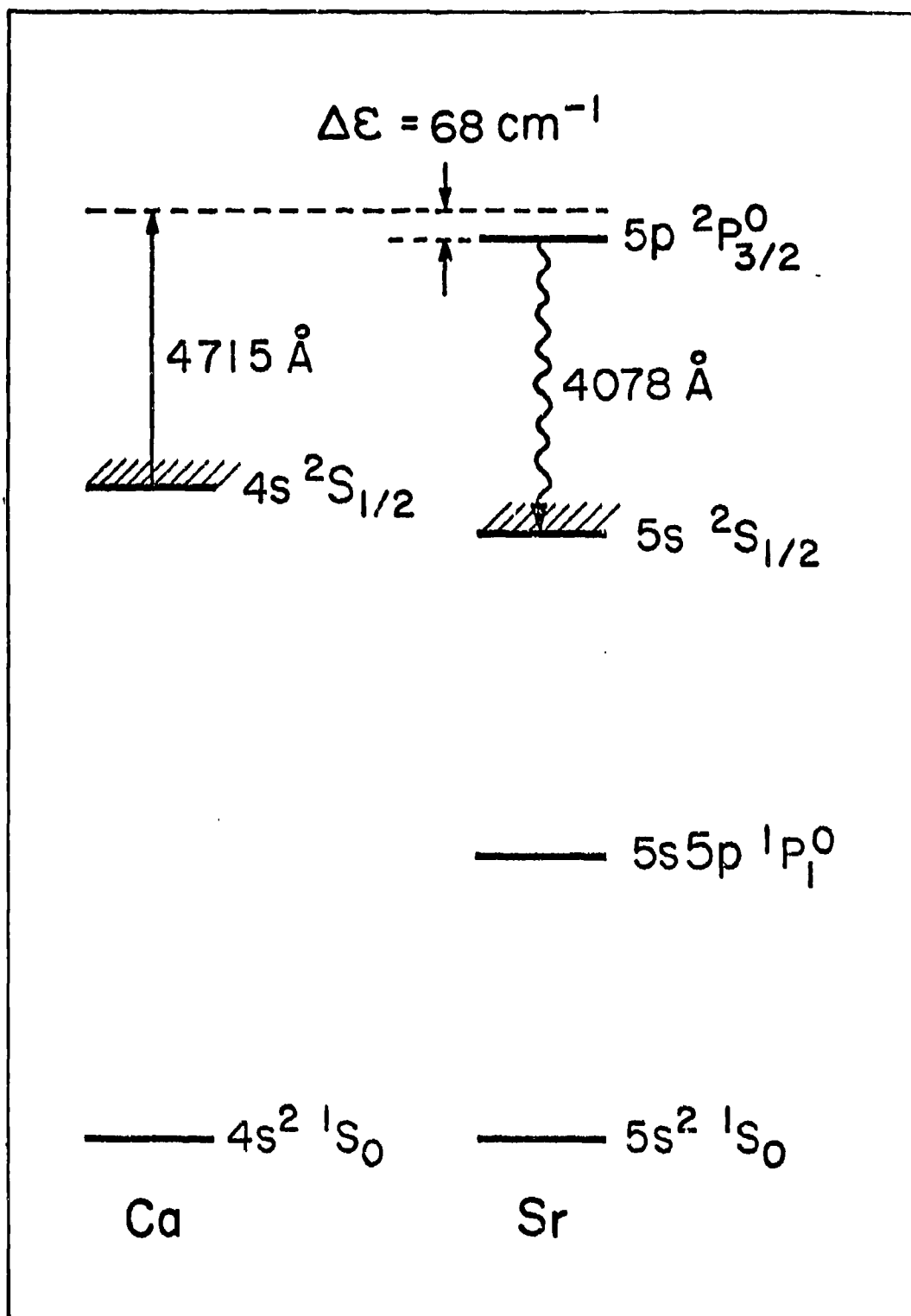


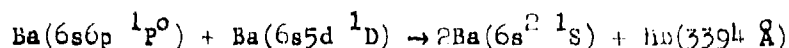
Fig. 1--Schematic of laser induced charge transfer collision.

This reaction differs from that above in that the target species is the $\text{Sr}^+(5p\ ^2P_{1/2}^o)$ ion rather than the $\text{Sr}^+(5p\ ^2P_{3/2}^o)$ ion. This second reaction is interesting in that its lineshape is broader and maximizes at a wavelength which is detuned a greater amount from $\lambda_{R=\infty}$. We are presently conducting further tests to verify the interpretation of our observations.

B. Atomic Pair Processes

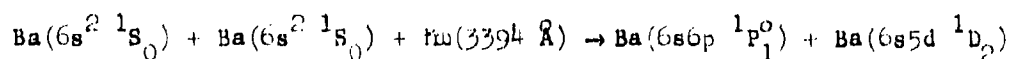
Pair absorption and pair emission refer to processes in which two atoms collide and absorb or emit a single photon at a wavelength corresponding to the sum of the energies of the excited states of the two collision partners. These processes are closely related to laser induced collisions and we have extensively studied them and their application to laser systems under this program.

The initial observation of the effect was radiative collisional fluorescence from the collision of two excited barium atoms,⁹ by the process

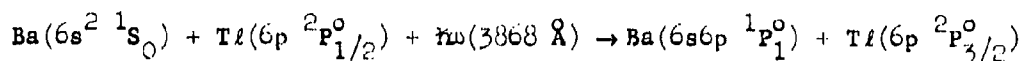


In this case, the emitted photon at 3394 Å has energy equal to the sum of the barium $6s6p\ ^1P^o$ and $6s^2\ ^1S$ states, and does not correspond to any of the energy level transitions of the individual barium atoms. The lineshape of the emission was similar to those observed for laser induced collisions.

Following the emission work, we observed the corresponding absorptive processes:^{9,10}

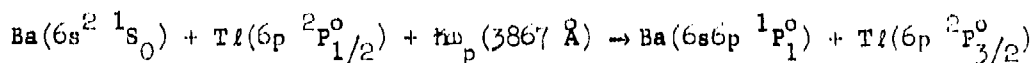


In addition, using a mixture of barium and thallium vapor, we observed the process



where the 3868 Å photon energy corresponds to the sum of the barium $6s6p \ ^1P_1^o$ and the thallium $6p \ ^2P_{3/2}^o$ levels. In both these experiments, the pair absorptions were found to be a few percent per centimeter for densities of $\sim 10^{17} \text{ cm}^{-3}$ and could be easily observed with simple white light absorption techniques.

The relatively large cross sections observed for pair absorption indicated that the process may be useful for pumping lasers. Such a technique was proposed by R. W. Falcone,¹¹ then one of the students working on this project. Just recently, we have successfully constructed a 1.5 μm atomic barium laser by optically pumping a pair absorption transition in barium-thallium vapor.¹² In particular, we have used the pair absorption process

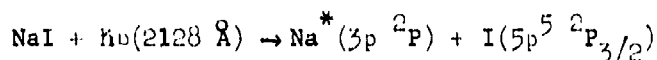


to pump the Ba $6s6p \ ^1P_1^o$ level. Subsequent lasing on the Ba $6s6p \ ^1P_1^o - 6s5d \ ^1D_2$ transition was observed. A preprint of a paper describing this work is included as Appendix C.

The pair absorption transitions described here are characterized by relatively large absorption widths ($\sim 1 \text{ \AA}$) and wavelengths in the near UV. Hence pair absorption is a method for channeling the energy of high powered, broadband, fixed wavelength lasers into specific target states of atoms which otherwise would not absorb the radiation. In particular, these techniques could be useful for the temporal and spectral compression of high powered excimer lasers.

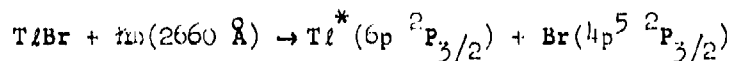
The application of pair processes to laser devices often require high densities of atomic excited states. During the contract period several techniques have been investigated for producing the required atomic densities.

We have demonstrated¹³ that photodissociation of NaI is a useful technique for producing large densities of Na $3p^2P$ state atoms, using the process



Subsequent lasing on the Na $3p^2P_{1/2} - 3s^2S_{1/2}$ transition was observed, with inversion densities of $\sim 10^{15}$ atoms/cm³.

We have also studied¹⁴ the photodissociation of TlBr



We found that, although the above process does occur, the $\text{Tl}^*(6p^2P_{3/2})$ atoms are quickly converted to the $\text{Tl}(6p^2P_{1/2})$ state via a "harpooning process". This work points out the importance of considering the potential curves for all possible products when considering photodissociation as a technique for the creation of excited states.

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2. J. C. White, "Inversion of the Na Resonance Line by Selective Photodissociation of NaI," Appl. Phys. Lett. 33, 325 (August 1978).
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2. J. F. Young: Conference on Multi-Photon Processes, Rochester, New York (June 1977).
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APPENDIX A

BIBLIOGRAPHY OF PUBLICATIONS SUPPORTED BY THE CONTRACT

- S. E. Harris and J. C. White, "Numerical Analysis of Laser Induced Inelastic Collisions."

Abstract: This paper studies the dynamics of (dipole-dipole) laser induced collision processes. Coupled equations are numerically integrated, first over time, and then over impact parameter. Transition probability and collision cross section are given as a function of the detuning from the $R = \infty$ frequency of the separated atoms, and of the incident laser power density. Numerical results are compared with approximate formulae for collision cross section at line center and in the wing.

- J. C. White, "Inversion of the Na Resonance Line by Selective Photodissociation of NaI."

Abstract: This letter reports the inversion and intense superfluorescent emission of the Na resonance line by selective photodissociation of NaI. The fifth harmonic of a Q-switched Nd:YAG laser at $2128 \text{ \AA}$ was used to photodissociate NaI to the unbound state $\text{Na}(3p^2P) + \text{I}(5p^5^2P_{3/2})$. Superfluorescence at the $5896\text{-}\text{\AA}$ resonance line was observed.

- J. C. White and G. A. Zdasink, "Branching Ratios for TlBr Photodissociation with $2660 \text{ \AA}$ Radiation."

Abstract: Selective molecular photodissociation as a pumping mechanism to invert atomic systems with respect to their ground levels has been used to construct a variety of laser sources.^{1,2} A more important application of these techniques, however, may be

the use of direct molecular photodissociation into atomic metastable states as a means of constructing stimulated Raman upconverters for infrared lasers. To date, only Carman and Lowdermilk³ have studied this possibility using metastable storage in atomic I following the flash photolysis of CF_3I . In the present work we examine the selective photodissociation of TlBr into the $\text{Tl}(6p\ ^2P_{3/2})$ metastable state as a potential candidate for improved Raman upconverters.

J. C. White, G. A. Zdasiuk, J. F. Young, and S. E. Harris, "Observation of Radiative Collisional Fluorescence."

Abstract: We report the observation of spontaneous radiative emission during the collision of two excited Ba atoms. The emission occurs at the sum energy of the excited atoms and is the emission analog of the laser-induced inelastic collision process. We measure a collision cross section for deexcitation by spontaneous radiative emission of $2.6 \times 10^{-20} \text{ cm}^2$.

R. W. Falcone, "Inversion of Atoms and Molecules to the Ground State by Optical Pumping."

Abstract: A technique is proposed for inverting atoms and molecules with respect to the ground state. The process is based on simultaneous excitation of colliding species by absorption of a photon. Visible wavelength lasers with millijoule energy outputs and several nanosecond pulse widths should be capable of demonstrating inversion of resonance lines in high-density metal-vapor mixtures.

S. E. Harris and J. F. Young, "Techniques for Rapid Laser Induced Energy Transfer Using Metastable States."

Abstract: We review recent experimental and theoretical results on two related methods of rapidly transferring stored population from

metastable states to selected target states of a different species. One method is kinetic-laser induced inelastic collisions, while the other is radiative-laser induced anti-Stokes scattering. Both methods utilize high peak power, short pulse, tunable lasers.

J. C. White, G. A. Zdasiuk, J. F. Young, and S. E. Harris, "Observation of Atomic-Pair Absorption with an Incoherent Source."

Abstract: We report the observation of atomic-pair absorption in barium and in barium-thallium vapors using a white-light source. At atomic densities on the order of 10^{17} atoms/cm³ we measure absorptions of 2.9% cm⁻¹ for Ba-Ba and 2.3% cm⁻¹ for Ba-Tl. In both cases, the absorption maxima occur at the predicted ($R = \infty$) wavelength and have a full width at half maximum of approximately 15 cm⁻¹.

W.R. Green, M.D. Wright, J.F. Young, and S.E. Harris, "Laser-Induced Charge Transfer to an Excited Ionic State."

Abstract: We report the observation of a laser-induced charge-transfer collision. In the presence of an intense laser beam, ground-state calcium ions collide with ground-state strontium atoms, selectively producing excited strontium ions and calcium neutrals. The laser-induced collision cross section has a line-width of about 50 cm⁻¹ and peaks ~ 70 cm⁻¹ from that wavelength which satisfies the energy defect of the separated atoms.

W.R. Green, M.D. Wright, J. Lukasik, J. F. Young, and S.E. Harris, "Observation of a Laser-Induced Dipole-Quadrupole Collision."

Abstract: Experimental observation of a laser-induced dipole-quadrupole collision between Sr and Ca is reported. The laser-induced collision cross section peaks at an applied photon energy

equal to the energy defect of the initial and final states of the infinitely separated atoms. A cross section of $3 \times 10^{-14} \text{ cm}^2$ was induced at a laser-power density of $7 \times 10^9 \text{ W/cm}^2$; the linewidth was 20 cm^{-1} .

S. E. Harris, J. F. Young, W. R. Green, R. W. Falcone, J. Lukasik,

J. C. White, J. R. Willison, M. D. Wright, and G. A. Zdasiuk,

"Laser Induced Collisional and Radiative Energy Transfer."

Abstract: This paper summarizes progress on two related methods of rapidly transferring energy which is stored in a metastable level of one species to a target level of a different species. The two methods are laser induced collision, and laser induced (two-photon) spontaneous emission and subsequent absorption. Both utilize high peak power, short pulse tunable lasers and both are capable of rapid and selective energy transfer. In particular the paper describes the recent experimental results of Green, et al., which demonstrate, we believe for the first time, laser induced charge transfer [1] and laser induced dipole-quadrupole collisions [2].

R. W. Falcone and G. A. Zdasiuk, "Pair Absorption Pumped Barium Laser."

Abstract: We report the first demonstration of the use of pair absorption transitions for optically pumping lasers. In a mixture of barium and thallium metal vapors, single-photon absorption of $3867 \text{ \AA}$ laser light caused simultaneous excitation of colliding ground state atoms to the Ba ($6s6p \text{ } ^1P_1^o$) and Tl ($6p \text{ } ^2P_{1/2}^o$) excited states. Excited state densities of about 10^{14} cm^{-3} were created, and subsequent laser emission on the Ba ($6s6p \text{ } ^1P_1^o$) $\rightarrow$ Ba ($6s5d \text{ } ^1D_2$) atomic transition at $1.5 \text{ }\mu\text{m}$ was observed.

APPENDIX B

STUDENTS RECEIVING Ph.D. DEGREES

K. S. Hsu (1977)

L. J. Zych (1978)

R. W. Falcone (1979)

W. R. Green (1979)

J. C. White (1979)

J. H. Newton (1979)

APPENDIX C

PAIR ABSORPTION PUMPED BARIUM LASER

by

R. W. Falcone and G. A. Zdziuk

Preprint

G.L. Report No. 3055

November 1979

supported by

NASA NGL-05-020-103

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Edward L. Ginzton Laboratory
W. W. Hansen Laboratories of Physics
Stanford University
Stanford, California

PAIR ABSORPTION PUMPED BARIUM LASER*

by

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ABSTRACT

We report the first demonstration of the use of pair absorption transitions for optically pumping lasers. In a mixture of barium and thallium metal vapors, single-photon absorption of 3867 Å laser light caused simultaneous excitation of colliding ground state atoms to the Ba ($6s6p\ ^1P_1^0$) and Tl ($6p\ ^2P_{3/2}^0$) excited states. Excited state densities of about 10^{14} cm^{-3} were created, and subsequent laser emission on the Ba ($6s6p\ ^1P_1^0 \rightarrow 6s5d\ ^1D_2$) atomic transition at 1.5 μm was observed.

*This work was jointly supported by the National Aeronautics and Space Administration under Contract NGL-05-020-103 and the U.S. Air Force under Contract F19628-77-C-0072.

PAIR ABSORPTION PUMPED BARIUM LASER

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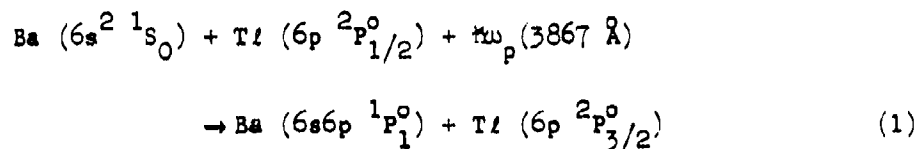
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We report the first demonstration of laser excitation of pair absorption transitions as a technique for optically pumping new types of lasers. In particular, we have simultaneously excited colliding ground state barium and thallium atoms to the Ba ($6s6p\ ^1P_1^o$) and Tl ($6p\ ^2P_{3/2}^o$) excited states by single-photon absorption of laser light at $3867\ \text{\AA}$. Subsequent laser emission on the inverted Ba ($6s6p\ ^1P_1^o \rightarrow 6s5d\ ^1D_2$) transition was observed at $1.5\ \mu\text{m}$.

Pair absorption, or simultaneous excitation, refers to a process in which two colliding atoms absorb a single photon at a wavelength corresponding to the sum energy of excited states of the individual species. After the collision the atoms separate, leaving both atoms in excited states. In the case of a dipole-dipole collisional interaction, one species makes a (dipole) allowed transition while the other species makes a (dipole) non-allowed transition. The use of laser excitation of pair absorption transitions for pumping new types of lasers was proposed in Ref. 1. The pair absorption process is a specific example of a general class of reactions now called laser induced collisions.^{2,3} Pair absorption has recently been observed in atomic metal vapors in the visible spectrum;⁴ previously it had been observed in molecular systems in the infrared.⁵

We have studied the pair absorption process



As shown in Fig. 1, the Ba atom is excited on an allowed transition and the Tl atom is excited on a non-allowed transition. The absorption process of Eq. (1) maximizes when the photon energy, $h\nu_p$, is equal to the sum of the energies of the atomic excited state products.

The absorption coefficient for pair absorption, $\alpha \text{ (cm}^{-1}\text{)}$, is given on line center by

$$\alpha = \frac{8\pi^6 \omega_p f_1 f_2 f_3 [\text{Tl}^o] [\text{Ba}^o]}{m^3 c \bar{v} \rho^2 (\Delta\omega)^2 \omega_1 \omega_2 \omega_3} \quad (2)$$

where e is the electronic charge; $\omega_p = 2\pi c/\lambda_p$ is the angular frequency of the absorbed light; f_1 , f_2 , f_3 and ω_1 , ω_2 , ω_3 are the oscillator strengths and frequencies of the atomic transitions $\text{Tl } (6p \ ^2P_{1/2}^o) \rightarrow \text{Tl } (7s \ ^2S_{1/2})$, $\text{Tl } (7s \ ^2S_{1/2}) \rightarrow \text{Tl } (6p \ ^2P_{3/2}^o)$, and $\text{Ba } (6s^2 \ ^1S_0) \rightarrow \text{Ba } (6s6p \ ^1P_1^o)$, respectively; $[\text{Tl}^o]$ and $[\text{Ba}^o]$ are the ground state number densities of the colliding atoms; m is the electron mass; c is the velocity of light; $\bar{v}$ is the relative velocity of the colliding atoms; ρ is the Weisskopf radius⁶ or dephasing radius of the collision (11 Å in this experiment); and $\Delta\omega$ is the frequency detuning of the pump laser relative to the atomic transition in Tl as indicated in Fig. 1. The excited state densities of both species, $\text{Ba}^* (6s6p \ ^1P_1^o)$ and $\text{Tl}^* (6p \ ^2P_{3/2}^o)$, are

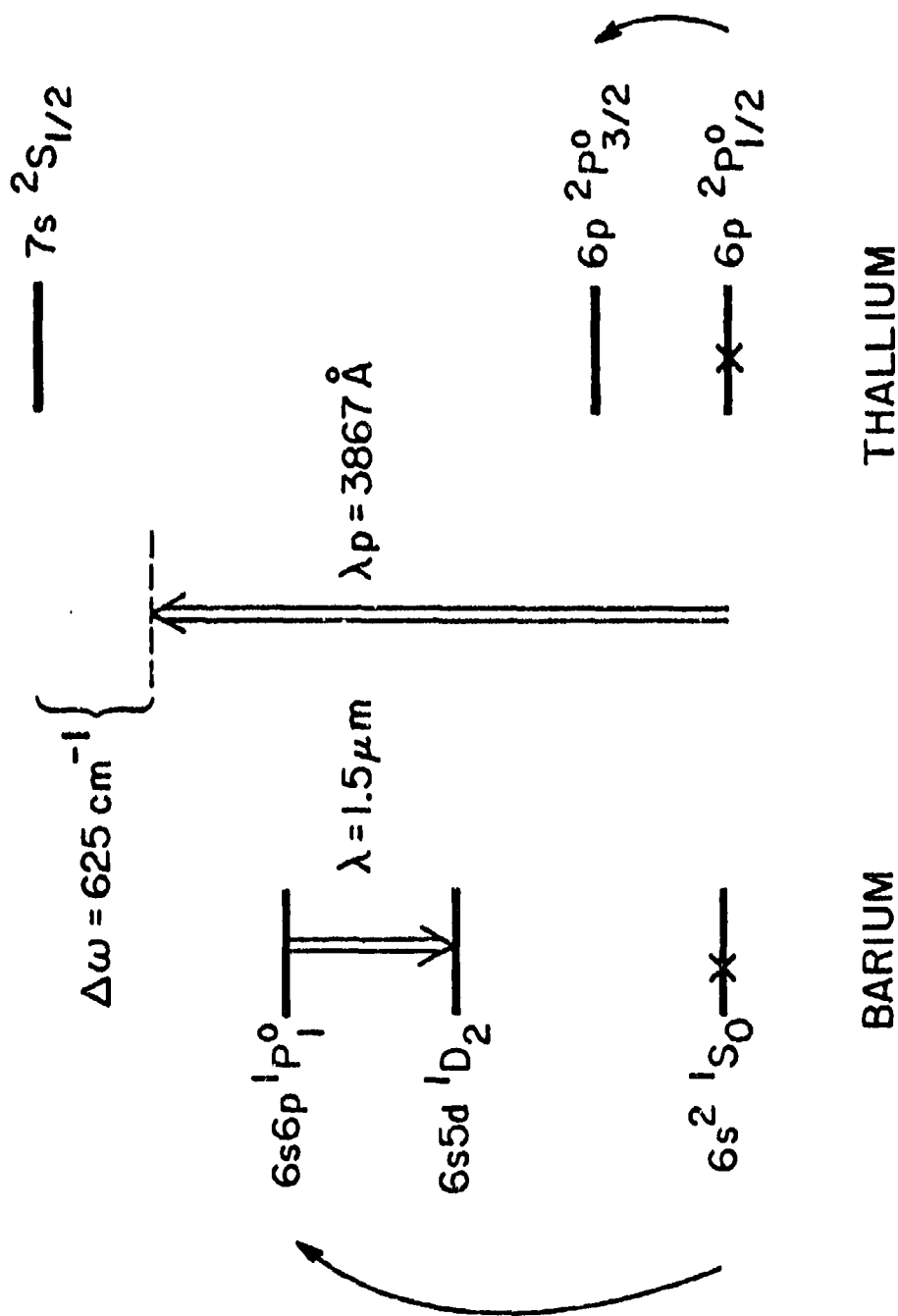


Fig. 1--Barium and thallium energy levels for the pair
absorption pumped atomic barium laser.

given by

$$[Ba^*] = [Tl^*] = \frac{P/A}{h\nu_p} \alpha \tau_p \quad (3)$$

where P/A is the power density and τ_p is the pulse length of the applied laser.

An experimental schematic is shown in Fig. 2. The 3867 Å radiation was produced by sum frequency generation in KD^*P of 6075 Å and 1.06 μm laser light using a Quanta Ray Nd:YAG dye laser system; pulse energies at 3867 Å of up to 5 mJ in a 5 ns pulse were available. This beam was collimated to a spot size of 0.15 cm² and directed into a metal vapor oven containing Ba and Tl heated to 1400°C over a vapor zone length of 10 cm. The cell also contained ~ 250 torr of argon gas to prevent metal vapor diffusion and condensation on the cold cell windows. The 1.5 μm output light from the cell was filtered with a spectrometer and detected with a room temperature InSb detector.

Figure 3 shows an absorption scan of the metal vapor cell in the region of pair absorption, made using a continuum discharge lamp. The shape of the curve agrees with the data of Ref. 4. A curve of growth analysis of the resonance line absorptions of Ba and Tl yielded ground state vapor densities of $[Ba^0] = 4 \times 10^{17} \text{ cm}^{-3}$ and $[Tl^0] = 2 \times 10^{17} \text{ cm}^{-3}$ in the cell. The measured absorption at 3867 Å was 35%, as compared with the predicted value of 80% at these densities using Eq. (2).

When the pump laser was tuned to 3867 Å, laser emission at $1.5000 \pm 0.0001 \text{ μm}$, corresponding to the Ba ($6s6p \ ^1P_1^o$) → Ba ($6s5d \ ^1D_2$) transition,⁷ was observed. The emission was narrow band (less than our 0.2 Å spectrometer

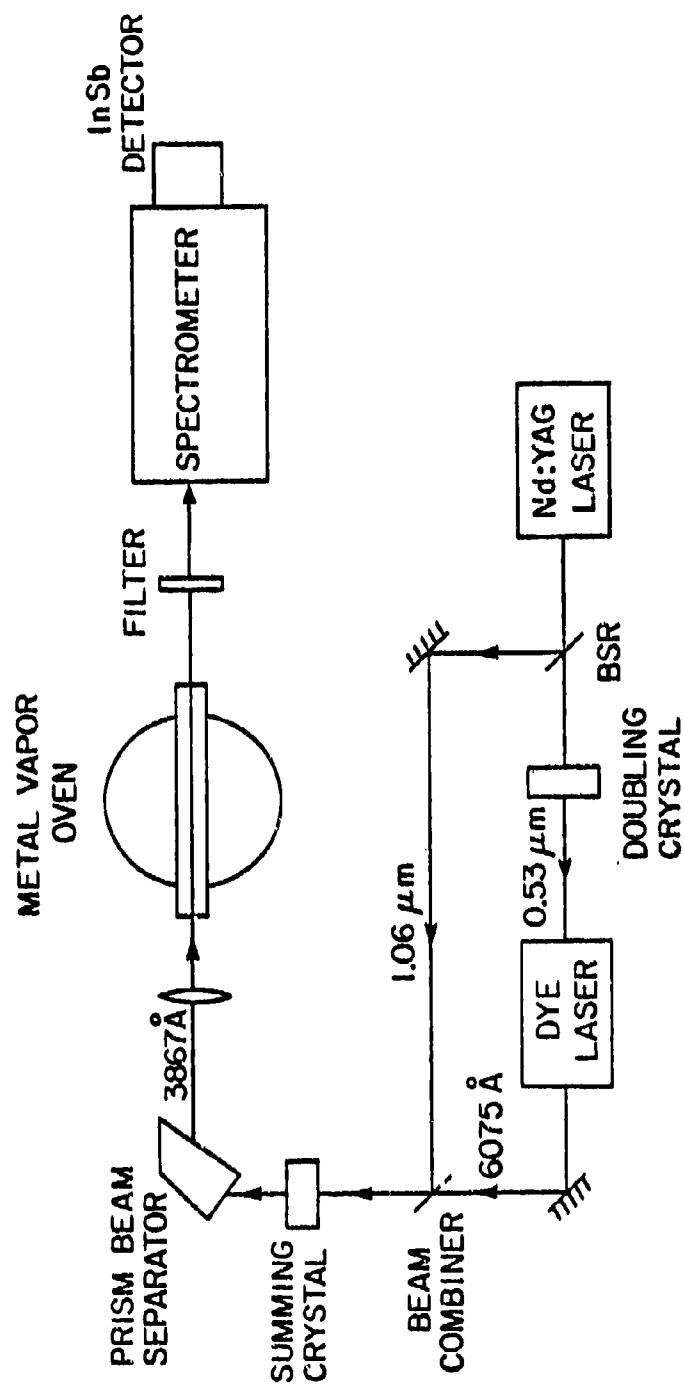


Fig. 2--Experimental schematic.

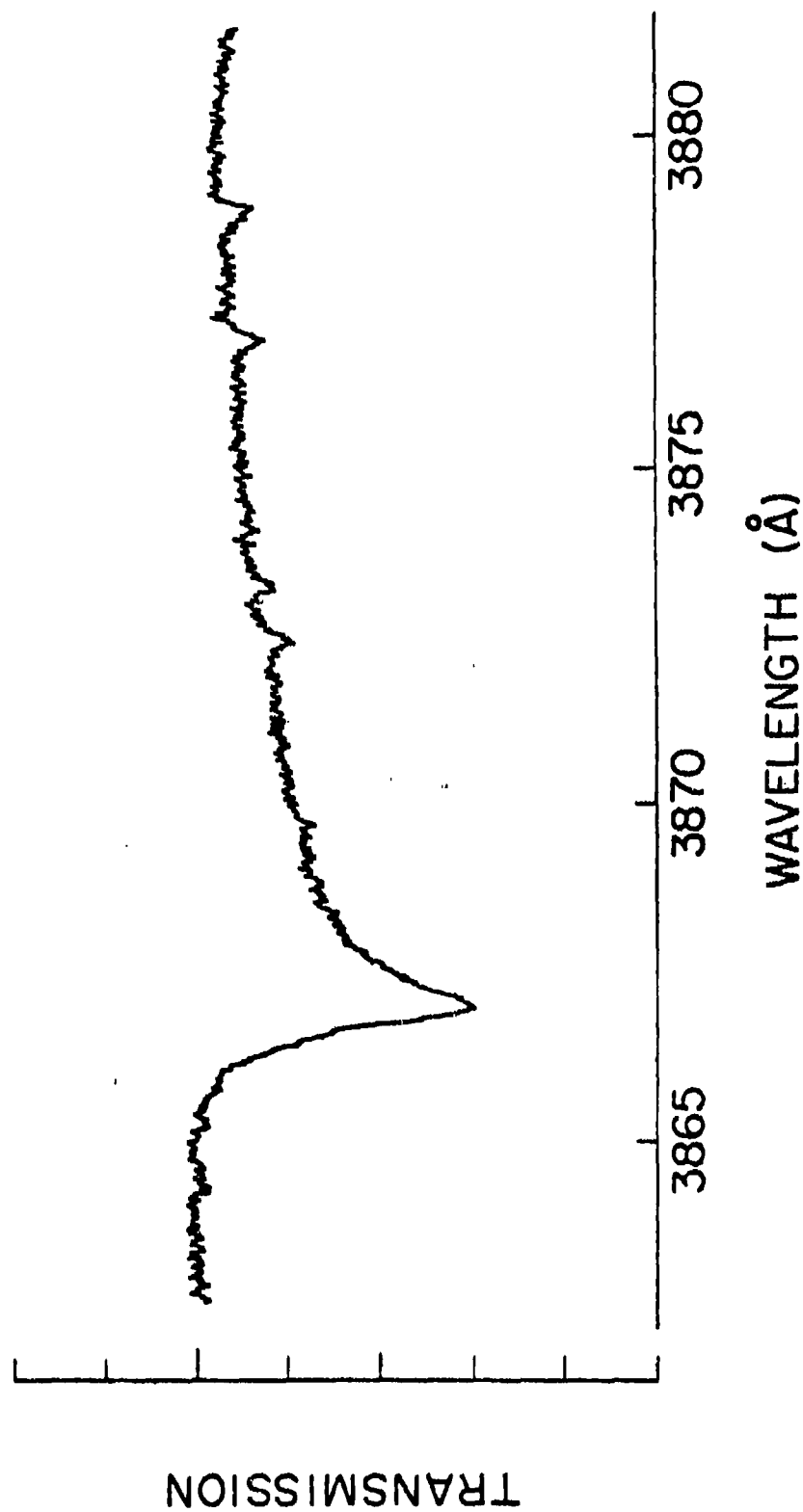


Fig. 3--Absorption scan showing barium-thallium pair absorption at 3867 Å.

resolution) and exhibited threshold behavior at a pump energy of 1 mJ. Spatial collimation was verified by placing a variable aperture between the detector and the cell. As the pump wavelength was tuned about 3867 Å, the observed emission wavelength at 1.5 μm remained constant. Our measurement of the time duration of the 1.5 μm emission was limited by the response time of the InSb detector to < 100 ns. [The spontaneous decay time of the Ba ($6s6p\ ^1P_1^o$) → Ba ($6s5d\ ^1D_2$) transition is ~ 200 ns.]⁷ The intensity of the emission as a function of pump laser wavelength is shown in Fig. 4. The intensity maximizes at a pump wavelength of 3867 Å and the profile has an asymmetry to longer wavelengths which is also seen on the absorption scan in Fig. 3.

The bandwidth of the pair absorption is about 1 Å (7 cm⁻¹). This absorption linewidth is characteristic of light induced collision processes² and is independent of both metal vapor density (in the linear absorption regime) and buffer gas density.

The measured cell absorption of 35% at 3867 Å implies an optically pumped excited state density of Ba ($6s6p\ ^1P_1^o$) of about 5×10^{14} cm⁻³ at the lasing threshold pump energy of 1 mJ per pulse. This estimated excited state density is consistent with calculations of the required threshold excited state density allowing for the 1×10^{14} cm⁻³ thermal population of the lower laser level.

This experiment demonstrates a new technique for optically pumping high densities of atomic (and molecular) species using the relatively large bandwidth cross section of pair absorption transitions. One of the most promising aspects of this technique is that new absorption wavelengths are created by mixing different species together; this provides a method for channeling the

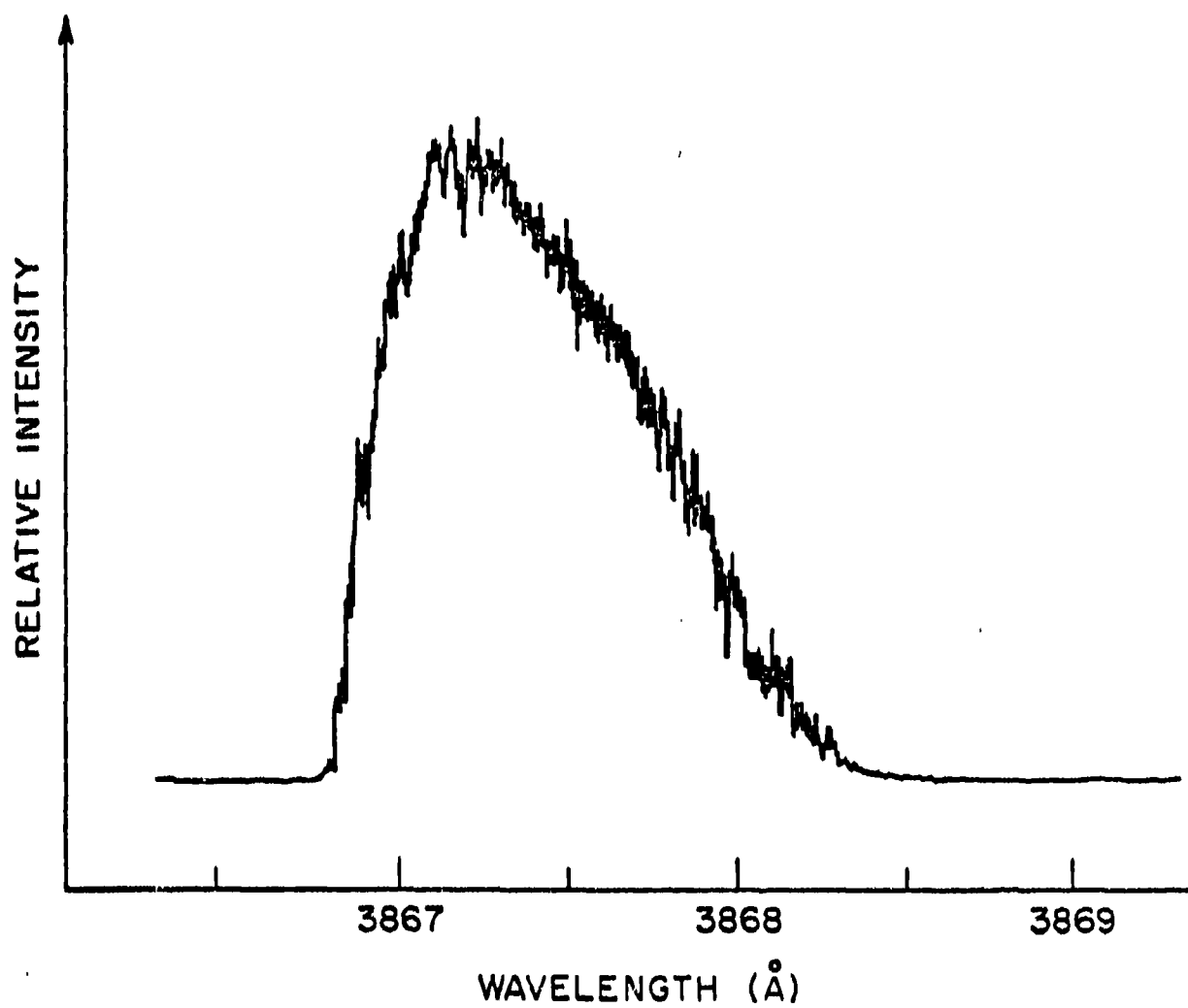


Fig. 4--Relative intensity of the $\lambda = 1.5 \mu\text{m}$ atomic barium laser as a function of pumping wavelength.

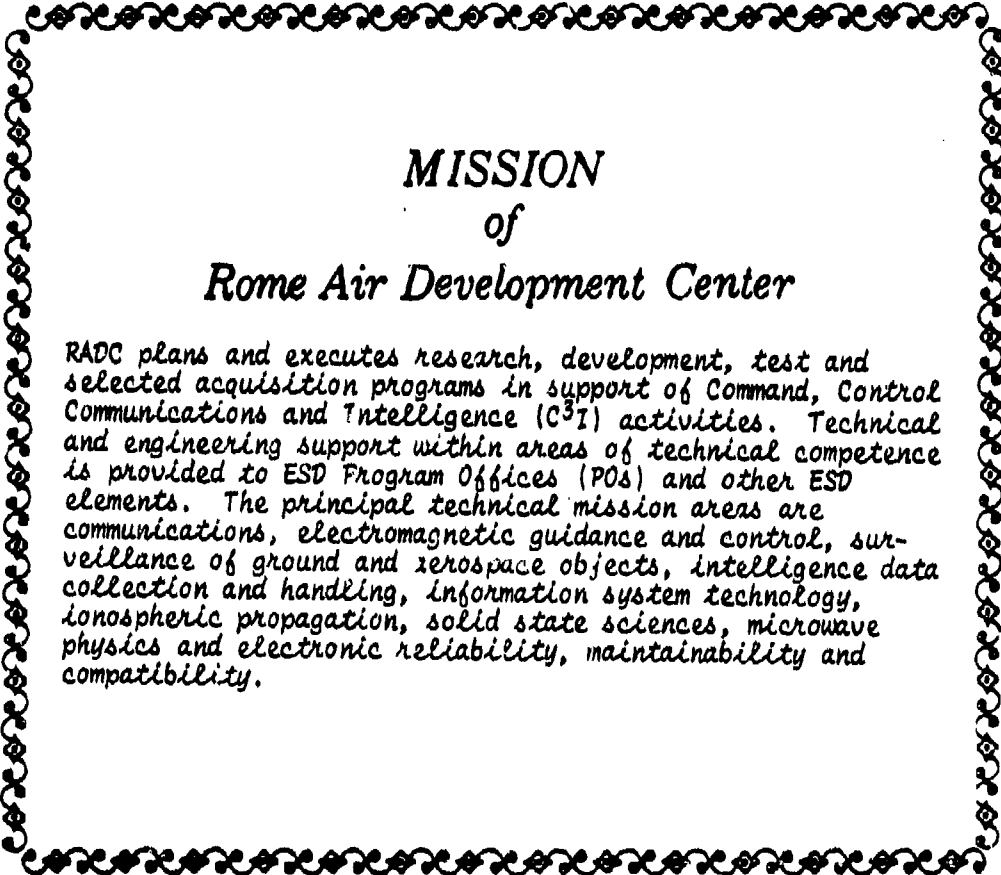
energy of high powered, fixed wavelength lasers into specific target states of atoms which otherwise would not absorb the radiation.

A further possibility is the use of pair absorption for the inversion of atoms and molecules to the ground state as pointed out in Ref. 1. A high density of one species will allow the inversion of the second species to the ground state when sufficient pump laser energy is applied at the pair absorption wavelength. One application of this technique would be the temporal and spectral compression of high powered excimer lasers. The energy stored in the inverted species by optical pumping with the excimer laser could be rapidly extracted in a narrow bandwidth pulse by Raman, two-photon, or other lasing process to the emptied ground state.

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