Cryogenic Tm:YAG Laser in the Near Infrared*

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Abstract— **Thulium laser operation on the $^3H_4 - {}^3H_6$ transition at 823 nm has been demonstrated in bulk Tm:YAG by quasi-cw pumping with a fiber-coupled diode array at 782 nm. Prior demonstrations of this laser transition have been limited primarily to fluorozirconate glass host materials in waveguide configurations. Population trapping has been identified as an important consideration in the design and operation of lasers on this transition. The use of a crystalline host should enable both high peak and average power operation, and the basic principles discussed here should be extensible to other crystalline hosts and to room temperature operation.**

Index Terms—**Near-infrared lasers, Solid-state lasers, Tm-doped lasers**

I. Introduction

THE wavelength diversity of efficient laser systems capable of both high peak and average power is very limited because of the large number of requirements that must be satisfied for such systems. Here, we report on initial results for Tm:YAG operating on the $^3H_4 - {}^3H_6$ transition in the 0.82 μm wavelength range. It can be pumped with efficient diode lasers in the 0.78 – 0.8 μm wavelength range. The transition has a small quantum defect for low thermal dissipation. The upper state lifetimes can be long, on the order of a millisecond for good energy storage. It also has sufficient gain bandwidth to support sub-ps-long pulses depending on the host material and temperature of operation. In short, this transition has many of the same properties as the 1-μm laser transition in Yb^{3+} for operation at high power. However, operation on this transition has been demonstrated mostly in Tm-doped glass waveguides [1]-[18], primarily fluorozirconate (ZBLAN) fibers, which are limited by the host material and waveguide form to low average power and energy storage. It has been suggested [19] that Tm:YAG could be used to build a multi-kW laser using this transition, but to our knowledge, no such laser operation has been demonstrated. There is also a report [20] with little information on an upconversion-pumped Tm, Yb:$LiYF_4$ laser on this transition by using 960 nm light to excite the Yb^{3+}. However, upconversion pumping requires two pump photons to generate one output photon, thereby limiting the efficiency. Extending operation on this transition to crystalline host materials and single-photon pumping opens up this wavelength range to efficient sources with the potential for high peak and average powers.

One difficulty with this transition is that the lower laser level is significantly populated at room temperature, which raises the laser threshold, and therefore limits operation. However, operation at cryogenic temperature depopulates the lower laser level, reduces laser threshold, increases efficiency, and greatly mitigates thermo-optic effects in crystalline host materials [21]-[23]. Here, we have used cryogenic cooling to enable laser operation on this transition in a crystalline material, although extension to room temperature operation should be possible similar to the 1-μm transition in Yb^{3+}. More importantly, we have identified population trapping to be a key consideration for this transition.

II. Spectroscopy of $^3H_4 - {}^3H_6$ transition in Tm^{3+}

In this laser transition as shown in Fig. 1, pump photons excite Tm ions from the ground-state $^3H_6$ manifold to the $^3H_4$ upper laser level manifold and then stimulated emission occurs from the $^3H_4$ manifold back to the ground-state manifold. Also shown in Fig. 1 is the most commonly used laser transition in Tm^{3+}, the $^3F_4 - {}^3H_6$ around 2 μm, the $^3H_4 - {}^3H_5$ around 2.3 μm, and $^3H_4 - {}^3F_4$ transition around 1.5 μm. These latter two transitions use the same $^3H_4$ upper-state manifold as the transition of interest here. The $^3H_4 - {}^3H_6$ transition is similar to other familiar transitions in rare earth ions such as the 1 μm laser transition in Yb^{3+} and the 0.94 μm transition in Nd^{3+}, in that both the pump and laser levels are within the same manifold leading to a small quantum defect. Alternatively, the $^3H_4$ manifold can be excited by upconversion using multiple pump photons to make one excited state [12], [20], but here the focus is on using a single pump photon per excited state.

The Tm:YAG absorption spectrum at 80 K and 300 K are shown in Fig. 2. The pump wavelength band can be easily accessed by efficient diode lasers. The fluorescence spectra at 80 and 300 K in a 1% doped Tm:YAG is shown in Fig. 3.

One of the issues with this transition is concentration quenching of the $^3H_4$ lifetime by energy transfer. For efficient 2 μm lasers, the Tm concentration is deliberately made sufficiently high that the $^3H_4$ lifetime is quenched by cross relaxation to obtain two $^3F_4$ excited states per pump photon (the 2-for-1 effect). For operation on the $^3H_4 - {}^3H_6$ transition, the doping concentration should be kept around 1% or less to

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mitigate this quenching [24]; at 1% doping in YAG the $^3H_4$ lifetime is approximately 400 μs at 80 K [25]. The non-radiative multi-phonon relaxation rate from $^3H_4$ in Tm:YAG is a on the same order as the radiative relaxation rate, so the radiative quantum efficiency even at low doping is on the order of 50% [24].

III. Key Physics and Rate Equation Analysis

A rate equation analysis can elucidate some of the physics that has held back the development of lasers on this transition, particularly in crystalline hosts. In particular, the presence of the intermediate-state $^3H_5$ and $^3F_4$ manifolds complicates the physics. Their presence causes two key effects that must be considered. The first is concentration quenching of the $^3H_4$ lifetime, analogous to concentration quenching in Nd laser materials, which was discussed above, and the second is trapping of excited-state population in the long-lived $^3F_4$ manifold. This analysis shows that population trapping in $^3F_4$ can have a detrimental effect on laser performance.

A. Rate Equations

In this first-order rate equation analysis, the populations of only $^3H_6$ (level 1), $^3F_4$ (level 2), and $^3H_4$ (level 3) are considered. The relaxation of the $^3H_5$ population to $^3F_4$ is assumed to be instantaneous with unity efficiency. Terms are included for multi-phonon relaxation from level 3 and for concentration quenching. Concentration quenching is treated as a process that can be characterized by a single exponential. Higher-lying manifolds are ignored; processes that excite ions to these higher lying manifolds can become important at high excited-state population densities through upconversion and excited-state absorption. Within these assumptions, the rate equations including pumping and stimulated emission are

$$\frac{dN_3}{dt} = \frac{I_p}{h\nu_p}\left(N_1\sigma_a(\lambda_p) - N_3\sigma_e(\lambda_p)\right) - \frac{I_s}{h\nu_s}\left(N_3\sigma_e(\lambda_s) - N_1\sigma_a(\lambda_s)\right) - \frac{N_3}{\tau_{r3}} - \frac{N_3}{\tau_{nr3}} - \frac{N_3}{\tau_{cr}} \quad (1a)$$

$$\frac{dN_2}{dt} = \frac{\beta_{32}N_3}{\tau_{r3}} + \frac{N_3}{\tau_{nr3}} + \frac{2N_3}{\tau_{cr}} - \frac{N_2}{\tau_2} \quad (1b)$$

$$\frac{dN_1}{dt} = \frac{-I_p}{h\nu_p}\left(N_1\sigma_a(\lambda_p) - N_3\sigma_e(\lambda_p)\right) + \frac{I_s}{h\nu_s}\left(N_3\sigma_e(\lambda_s) - N_1\sigma_a(\lambda_s)\right) - \frac{N_3}{\tau_{cr}} + \frac{\beta_{31}N_3}{\tau_{r3}} + \frac{N_2}{\tau_2} \quad (1c)$$

where N_x is the population of level x and $N = N_1 + N_2 + N_3$ and N is the dopant concentration. I_p is the pump intensity, and $h\nu_p$ is the pump photon energy. $\sigma_a(\lambda)$ and $\sigma_e(\lambda)$ are the effective absorption and emission cross sections as a function of wavelength, and λ_p and λ_s are the pump and signal wavelengths. τ_{r3} is the radiative lifetime of level 3 and τ_2 is the lifetime of level 2. β_{32} and β_{31} are the branching ratios of the radiative relaxations from level 3, and $\beta_{32} + \beta_{31} = 1$. τ_{nr3} is the inverse of the multi-phonon relaxation rate from level 3 and τ_{cr} is the inverse of the concentration-quenching rate under the simplification that this process can be considered to be exponential.

In (1a) the first term on the right-hand side describes pumping, the second term describes stimulated emission, the third term is spontaneous emission, the fourth term is multi-phonon relaxation, and the last term is cross relaxation with a Tm ion in the ground state manifold.

To elucidate the effect of trapping, the population of level 2 can be solved under steady-state pumping giving

$$N_2 = N_3\tau_2\left(\frac{\beta_{32}}{\tau_{r3}} + \frac{1}{\tau_{nr3}} + \frac{2}{\tau_{cr}}\right). \quad (2)$$

A limiting case is for only radiative relaxation from level 3 in which case (2) simplifies to

$$N_2 = N_3\beta_{32}(\tau_2/\tau_{r3}). \quad (3)$$

β_{32} is typically on the order of 0.1 – 0.2 for many Tm-doped materials [24, 26], and since the fluorescence lifetime of level 2 is typically 5–10 times longer than radiative lifetime of level 3, in the absence of trapping mitigation approaches, the population of level 2 is expected to be around that of level 3 in steady state. Since the laser transition has a lower level in the ground-state manifold, the populations of both levels 2 and 3 can be significant just to reach transparency at the laser wavelength, and bleaching at the pump wavelength can start to be significant.

If multiphonon relaxation and concentration quenching are present, then N_2 can be substantially larger than N_3. Concentration quenching is particularly detrimental because one cross relaxation event, which reduces the population of level 3 by one, increases the level 2 population by 2. This property is the well-known 2-for-1 process in Tm^{3+}, which helps the 2 μm transition be efficient, but clearly is not desired for the transition of interest here. This relation in (2) applies both above and below laser threshold.

B. Example: Tm:YAG

For example in Tm:YAG from Caird [24], $\beta_{32} = 0.16$, $\tau_{r3} = 1.45$ ms, $\tau_{nr3} = 1.7$ ms, and $\tau_2 = 9.9$ ms. If non-radiative relaxation were to be negligible, then $N_2 = 1.1N_3$ in steady state. However, since multi-phonon relaxation is significant, at best $N_2 = 6.9N_3$ at low Tm^{3+} concentration. Even a modest multiphonon relaxation rate causes significant population trapping because this form of relaxation is efficient in feeding level 2. This effect is particularly large for YAG because the multiphonon relaxation rate is significantly larger than for other candidate hosts such as $LiYF_4$ or $YAlO_3$.

Concentration quenching makes the situation much worse. For 1% doping in YAG, the lifetime of level 2 drops to around 0.45 ms [25], which means $\tau_{cr}(1\%) = 1.06$ ms and that $N_2 = 26N_3$. Consequently, even for only modest Tm^{3+} doping, population trapping is a large effect in steady state. For a room temperature laser, the required population in $^3H_4$ to reach transparency will be a few percent. Achieving transparency will not be physically possible if the Tm^{3+} doping is too high,

unless an approach is implemented to mitigate population trapping. Even at low doping, bleaching of the pump absorption will be much more significant than an equivalent Yb^{3+} laser.

A previous rate equation analysis for Tm:YLF operating as an upconversion laser at 1.5 μm on the $^3H_4 - {}^3F_4$ transition by Diening et al. [27] also shows this population trapping effect. In their particular situation the material was co-doped with Yb and pump photons excite the Yb, which transfers its energy to Tm. At room temperature in steady state, this analysis showed the $^3H_6$ population dropping to less than 25% of the population while the $^3F_4$ population increasing to 40% of the population and the $^3H_4$ population being <20% of the population.

IV. Laser Experiments

A proof-of-principle laser was demonstrated using 1%-doped Tm:YAG cryogenically cooled to liquid nitrogen temperature. Cryogenic cooling was used to reduce the amount of $^3H_4$ population needed to reach inversion, thereby mitigating the trapping effect. Additionally, quasi-cw pumping was used since the $^3F_4$ is unoccupied at the beginning of the pump pulse (provided the pulse repetition rate is low enough) and approaches its steady state value only for pumping times on the order of the $^3F_4$ lifetime or longer; for pumping times much shorter than the $^3F_4$ lifetime, trapping is mitigated.

The laser was configured as an end-pumped rod with the length of the antireflection coated gain element being 1 cm. A fiber-coupled diode array at 782 nm with a core diameter of 400 μm and 0.22 NA was used to pump the laser, and its output was focused through a flat dichroic mirror with high reflectivity at 823 nm and high transmission at 780 nm to a spot of 825 μm in diameter in the gain element. The output coupler was also flat, and the resonator was about 20 cm long with the Tm:YAG located near the center.

Figure 4 shows the results for pumping with 5 ms long pulses at 50 Hz pulse repetition frequency. The output power is defined as the output energy in the pulse divided by the pump pulse length, and similarly for the absorbed pump power. The fraction of the incident pump power that was absorbed was 42%. Up to 21 W peak power was obtained with a slope efficiency of 36%.

Figure 5 shows the output power as a function of time for a single 20-ms-long pump pulse. The output power drops over the length of the pump pulse, and in fact, if the pump pulse is left on, the laser ultimately goes below laser threshold. The nominal time constant of the decay of the output power is similar to the $^3F_4$ lifetime, which is expected based on the rate equations. We have ruled out a thermal effect as the cause of this decay, given our previous experience with cryogenic lasers.

In summary, we have extended laser operation of the $^3H_4 - {}^3H_6$ transition at 823 nm using single-photon pumping in Tm^{3+} to a crystalline host material. This demonstration used YAG as the host material, but we expect this to be readily extensible to other crystalline host materials. More importantly, we have identified population trapping in the $^3F_4$ manifold as a key consideration in the design and operation of this laser transition. For this initial proof-of-principle laser demonstration, a combination of quasi-cw pumping and cryogenic laser operation mitigated trapping. Other approaches to reduce the $^3F_4$ lifetime, such as co-doping to quench the lifetime [28, 29], stimulated emission on the $^3F_4 - {}^3H_6$ transition, or upconversion out of the $^3F_4$ transition can mitigate this population trapping. This opens up the potential for relatively simple and efficient high peak and average power lasers operating in this wavelength range.

V. Acknowledgements

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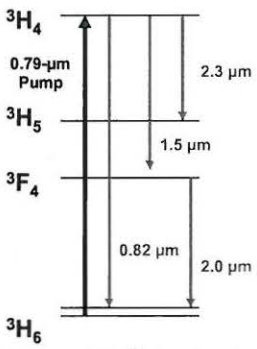

Fig. 1. Energy level diagram of Tm^{3+} showing the pump transition in black and laser transitions from the $^3H_4$ manifold and below in gray.

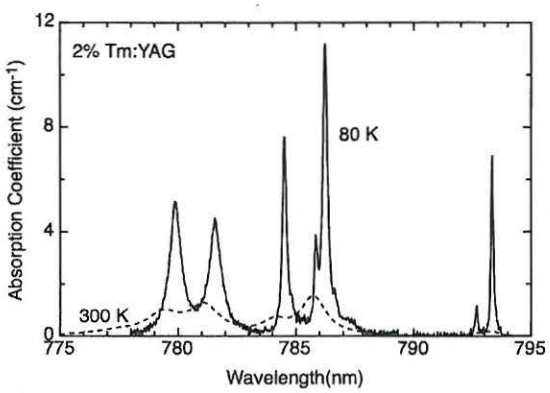

Fig. 2. Absorption spectra at 80 and 300 K for 2% Tm:YAG. The pump diode in the laser experiment was at the absorption feature at 782 nm.

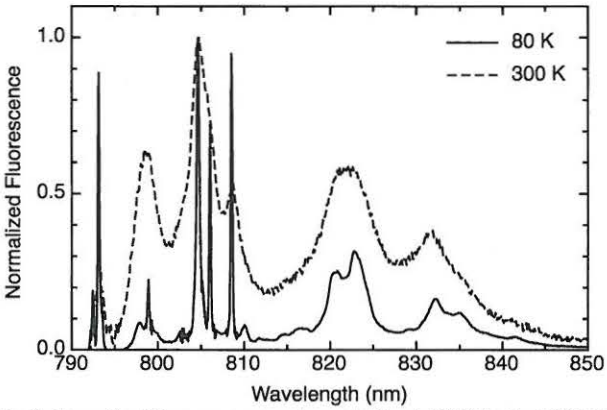

Fig. 3. Normalized fluorescence spectrum at 80 and 300 K in Tm:YAG. The laser operated on the line at 823 nm.

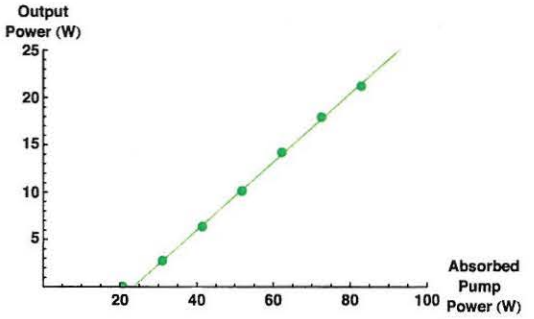

Fig. 4. Output power as a function of absorbed input power for 10% transmission output coupler and a 5 ms pump pulse at 500 Hz pulse repetition frequency. The powers are defined as the energy divided by 5 ms.

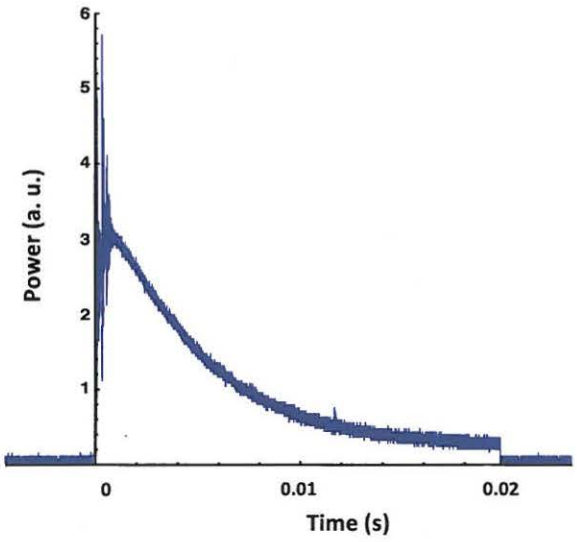

Fig. 5. Output power at 823 nm as a function of time for a 20-ms-long pump pulse. In this example, the output power averaged over the pump pulse is 11 W for an absorbed pump power averaged over the pump pulse of 75 W. The pump pulse starts about 250 μs before the Tm laser reaches threshold.

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