Diode-pumped midinfrared solid-state lasers

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he recent development of high power semiconductor lasers has led to a renewal of interest in resonantly-pumped solid-state lasers.¹ Solid-state lasers pumped by diode lasers are more efficient, compact, and rugged than those pumped by other sources such as lamps. The higher prices of diode lasers compared to other pump sources currently limits the use of diode-pumped solid state lasers to relatively low power applications. During the past several years, however, the available power of laser diode arrays has steadily increased, while the price per unit power has dropped considerably. If these trends continue as expected, diode-pumped solid-state lasers will find increasing applications.

The trivalent rare earth ions are natural candidates for resonant pumping due to their sharp absorption lines. Four of these ions, Nd^{3+} (neodymium), Dy^{3+} (dysprosium), Er^{3+} (erbium), and Tm^{3+} (thulium), have strong absorption lines² in the 800 nm region where high power AlGaAs diode laser arrays are commercially available. Of these ions, all but Dy^{3+} have laser transitions that have been pumped by diode lasers.

This article will review the trivalent rare earth laser transitions in the 2-3 μ m region that have been pumped by diode lasers. Several applications require wavelengths in this region. Lasers operating within the strong absorption bands of liquid water are useful for medical applications due to the high water content of most biological tissues. Two of these bands are centered at 1.94 μ m (with peak absorption coefficient α =1.1x10² cm⁻¹)³ and at 2.95 μ m (peak α =1.2×10⁴ cm⁻¹).⁴ These lasers are also useful for eye-safe lidar applications and as pump sources for par-

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ametric oscillators operating at mid-infrared wavelengths.

Diode pumped operation of the Nd^{3 +4}F_{3/2} \rightarrow ⁴I_{11/2} transition⁵ (at 1.06 µm in YAG) and ⁴F_{3/2} \rightarrow ⁴I_{13/2} transition (at 1.32 µm in YAG) has been investigated in various crystal hosts. Output energies as high as hundreds of millijoules,⁶ and powers of several watts,⁷ have been achieved for the 1.06 µm transition. Both of these Nd³⁺ transitions conform very closely to the ideal four-level laser model at room temperature. In both cases, the decay from the pump level to the upper laser level is essentially 100% efficient. The lower laser level decays rapidly to the ground state and has essentially no thermal population because its position is more than 2000 cm⁻¹ above the ground state.

In contrast to Nd³⁺, most of the trivalent rare earth transitions in the 2-3 μ m region do not follow the ideal four-level laser model. To date, four transitions in this spectral region, Tm³⁺ ³F₄ \rightarrow ³H₆ at 2.0 μ m, Ho³⁺ (holmium) ⁵I₇ \rightarrow ⁵I₈ at 2.1 μ m, Tm³⁺ ³H₄ \rightarrow ³H₅ at 2.3 μ m, and Er³⁺ ⁴I_{11/2} \rightarrow ⁴I_{13/2} at 2.8 μ m, have been diode-pumped. The wavelengths quoted are nominal values, since the precise wavelength depends on the particular crystal host material used. Only the 2.3 μ m Tm³⁺ transition is a nearly ideal four-level laser. Each of the others has unique operating characteristics that set it apart from the nearly ideal four-level laser model.

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Quasi-three-level lasers

The Tm³⁺ 2.0 μ m ³F₄ \rightarrow ³H⁶ and the Ho³⁺ 2.1 μ m ⁵I₇ \rightarrow ⁵I₈ laser transitions operate in what is referred to as the quasithree-level scheme. The Tm³⁺ transition is shown in Fig. 1. The lower laser level is one of the 13 crystal-field levels of the ³H₆ ground electronic state (hence the term quasithree-level), and the upper laser level is one of the 9 crystalfield levels of the ³F₄ state. For each electronic state, only the center of gravity of the crystal-field levels is shown.

The threshold and output characteristics of the quasithree-level laser system under longitudinal pumping have been modeled by Fan and Byer^{1,8} and independently by Risk.⁹ The threshold pump power is proportional to $[(L+T)+2f_1\sigma Nl]/f_2\sigma\eta_a$, where L is the round-trip cavity loss, T is the output transmission, σ is the gain crosssection, N is the density of dopant ions, *l* is the crystal length, η_a is the fraction of pump power absorbed, f_1 is the fraction of the ³H₆ population residing in the lower laser level crystal-field component, and f_2 is the fraction of the ³F₄ population residing in the upper laser level crystal-field component.

The first term, $(L+T)/f_2\sigma\eta_a$, is the usual four-level laser threshold ($f_2\sigma$ is the effective emission cross-section). In the second term, $f_1\sigma$ is the effective absorption crosssection, and therefore $f_1\sigma N$ is the absorption coefficient at the laser wavelength. The second term can therefore be thought of as a round-trip reabsorption loss, due to the population f_1 in the lower laser level.

For Tm³⁺ in YAG, the Boltzmann factors are $f_1=0.017$ and $f_2=0.46$ at room temperature for the 2.02 µm transition. Hence the term quasi-three-level laser is appropriate, since only 3.6% of the ground state population must be inverted to reach threshold, in contrast to the 50% inversion required in the three-level ruby laser. The population ratio f_1/f_2 decreases as the temperature is lowered. If the temperature is lowered sufficiently, the reabsorption loss term becomes negligible compared to the cavity loss term, and the quasi-three-level laser becomes a four-level laser.

The effect of the reabsorption loss can be reduced by using a shorter laser crystal. If the crystal is made too short, however, the threshold rises because insufficient pump power is absorbed. The absorbed pump power fraction is $\eta_a=1-\exp(-\alpha l)$, where α is the pump absorption coefficient. If the crystal is made shorter than $\approx 1/\alpha$, then $\eta_a \approx \alpha l$. Therefore, the reabsorption loss term (proportional to Nl/η_a) is not reduced appreciably as l is made substantially shorter than $1/\alpha$. The optimum crystal length that minimizes the threshold can be found by differentiating the threshold pump power expression with respect to $l.^9$ In any case, a large value of the pump absorption cross-section (α/N) is desirable. The resonant pumping scheme for the $Tm^{3+} 2.0 \mu m$ transition is shown in Fig. 1. The upper laser level is populated through the self-quenching process. This nearly resonant exchange of energy occurs through a dipoledipole interaction between nearby Tm^{3+} ions. The energy mismatch of the participating electronic levels is balanced by the emission of one or more phonons. The efficiency of the self-quenching process increases with increasing density of the Tm^{3+} dopant ion. For Tm^{3+} concentrations greater than approximately 5×10^{20} cm⁻³ (4% atomic substitution), the self-quenching dominates the decay of the ${}^{3}H_{4}$ state and the ${}^{3}F_{4}$ upper laser level is populated with a quantum efficiency of two.

Kintz et al.¹⁰ obtained cw laser emission at 2.02 μ m for this transition in YAG by pumping with a 200 mW, 785 nm laser diode array. The Tm³⁺ concentration was 12%, ensuring efficient population of the upper laser level. The slope efficiency (output power/absorbed pump power) was 56%, demonstrating a quantum efficiency of pumping greater than one. Tunable cw laser emission over the range 1.87-2.16 μ m in YAG (with 370 mW output power at 2.02



FIGURE 1. Resonant pumping scheme for Tm^{3+} laser transitions. The quasi-three-level 2.0 µm transition and the four-level 2.3 µm transition are shown. The 2.0 µm laser works best when the self-quenching is allowed, while the 2.3 µm laser prefers minimized selfquenching.



FIGURE 2. Resonant pumping scheme for the Ho³⁺ 2.1 μ m quasi-three-level laser transition.

 μ m), and over the range 1.85-2.14 μ m in YSGG (Y₃Sc₂Ga₃O₁₂), has been obtained for this transition with Ti:sapphire pumping,¹¹ and over the range 1.91-2.11 μ m in YSGG with diode laser pumping.¹² The short wavelength end of the tuning range is limited by reabsorption loss, due to the exponential increase in the Boltzmann ratio f_1/f_2 with decreasing wavelength. The long wavelength end of the tuning range is limited by the cavity losses.

The resonant pumping scheme for the 2.1 μ m Ho³⁺ transition is shown in Fig. 2. Both Tm³⁺ and Ho³⁺ ions are doped into the same crystal. The Ho³⁺ ⁵I₇ upper laser level is populated by the nearly resonant transfer of energy from the Tm³⁺ ³F₄ state. The Tm³⁺ ³F₄ state is populated by the self-quenching process as before. The threshold for the Ho³⁺ laser transition is typically lower than the Tm³⁺ 2.0 μ m threshold, since the Ho³⁺ concentration is kept low to minimize the reabsorption loss, while the Tm³⁺ concentration is held high enough to efficiently absorb the pump light and exploit the self-quenching.

Laser emission at 2.09 μ m was obtained by Allen et al.¹³ for this transition in YAG at 77 K using a 785 nm laser diode array pump. Room temperature operation was achieved by Fan et al.^{14,15} with a single-stripe laser diode pump, and by Kintz et al.¹⁶ with a laser diode array pump. Laser emission for this transition has also been obtained in the host crystal YLiF₄ (YLF) at 2.07 μ m at 77K,¹⁷ and at 2.08 μ m (26% slope efficiency)¹⁸ and 2.07 μ m (30% slope efficiency)¹⁹ at room temperature.

Bottlenecking in four-level lasers

If the Tm³⁺ concentration is held below approximately 2%, the ³H₄ lifetime is not shortened appreciably by the selfquenching process. In this case, the 2.3 μ m transition, shown in Fig. 1, can be operated with relatively low threshold. This is a four-level laser transition at room temperature since the ³H₅ state has essentially no thermal population due to its position more than 8,000 cm⁻¹ above the ground state. The rapid nonradiative (multi-phonon) decay from the ³H₅ lower laser level to the ³F₄ state permits cw operation.

Laser emission at 2.31 μ m was obtained by Thomas et al.²⁰ for this transition in YLF at room temperature with 15% slope efficiency using a pulsed alexandrite laser at 781 nm as the pump. Diode-pumped cw laser emission at 2.31 μ m was obtained by Kintz et al.¹⁸ at room temperature with 16% slope efficiency. Tunable laser emission in the range 2.29 to 2.42 μ m has been observed²¹ for this transition with a cw Ti:sapphire pump.





FIGURE 3. Resonant pumping scheme for the Er^{3+} 2.8 μ m four-level laser transition.

If the lifetime of the lower laser level is not negligible compared to the upper laser level lifetime, the population may build up, or bottleneck, in the lower laser level. This condition, referred to as self-termination, prevents cw operation. The short lifetime of the ${}^{3}H_{5}$ state relative to the ${}^{3}H_{4}$ state prevents self-termination of the Tm³⁺ 2.3 µm transition. The possibility of self-termination does, however, become an issue with the Er³⁺ 2.8 µm transition. The resonant pumping scheme for this four-level transition is shown in Fig. 3. For Er:YLF, the lifetime of the lower laser level is 13.2 ms, while the upper laser level lifetime is 4.2 ms.²²

The threshold pump power for a transition for which the lower laser level lifetime is not negligibly small is proportional to $(L+T)/f_2\sigma\eta_a\beta$, where $\beta=1-(f_1/f_2)(b_{21}\tau_1/\tau_2)$ is a factor that expresses the increase in cw threshold due to the non-negligible lower laser level lifetime τ_1 . The upper laser level lifetime is τ_2 , and b_{21} is the fraction of the decay (radiative plus non-radiative) from the upper laser level that reaches the lower laser level.

Continuous wave operation is allowed when $\beta > 0$. This

condition can be expressed as $f_1/f_2 < \gamma_{out}/\gamma_{in}$, where $\gamma_{in} = b_{21}/\tau_2$ is the rate of population entering the lower laser level, and $\gamma_{out} = 1/\tau_1$ is the rate of population leaving that level. That is, cw operation is possible even when the lower laser level lifetime is longer than that of the upper laser level lifetime, provided that the ratio of the Boltzmann factors is low enough. The partially bottlenecked four-level laser is therefore similar to the quasi-three-level laser in that it has a significantly lower threshold at reduced temperatures. At elevated temperatures, the partially bottlenecked laser eventually becomes self-terminating. In contrast to the quasi-three-level laser, however, the threshold of the partially bottlenecked laser does not increase with crystal length.

For the 2.8 μ m Er:YLF transition, $b_{21}=0.5$,²² and the Boltzmann ratio is $f_1/f_2=0.11/0.20$ at room temperature. The parameter β is therefore 0.14, implying that cw operation should be possible (although the threshold is raised by a factor of $1/\beta=7$ due to the non-negligible lifetime of the lower laser level). Room temperature cw laser emission was in fact observed²³ at 2.8 μ m in 8% Er:YLF pumped by a 797 nm laser diode array. The slope efficiency was 0.7%, with 147 mW threshold. The slope efficiency for cw diode-pumped laser emission from this transition was subsequently improved to 10% (with 23 mW threshold) by raising the Er³⁺ doping level from 8% to 30%.²⁴ The improved laser performance is attributed to the nonlinear upconversion process ${}^{4I}_{13/2}$ + ${}^{4I}_{13/2}$ - ${}^{4I}_{15/2}$ + ${}^{4I}_{19/2}$ that pumps population out of the lower laser level.²²⁻²⁴

Quasi-continuous-wave pumping

Pulsed laser diode arrays have been successfully employed in pumping the Nd³⁺ 1.06 μ m transition.⁶ These quasi-cw arrays typically have pulse durations of 200 μ s and

TABLE 1. Room temperature diodepumped mid-infrared solid state lasers, with highest reported efficiencies.

Laser transition	Wavelength	Slope efficiency
Tm ³⁺ ³ F ₄ Æ ³ H ₆	2.02 µm	56% ¹⁰
Ho ^{3+ 5} I7Æ ⁵ I8	2.09 µm	30%19
Tm ³⁺ ³ H ₄ Æ ³ H ₅	2.31 μm	16%18
Er ³⁺ ⁴ I ₁₁ ,Æ ⁴ I ₁₁	2.80 µm	10% ²

Diode-pumped lasers are already becoming competitive in applications where weight, size, and ruggedness are important issues.

have peak powers more than an order of magnitude higher than cw devices. The pulse duration is chosen to match the 230 µs lifetime of the Nd³⁺ ${}^{4}F_{3/2}$ upper laser level in YAG. The lifetimes of the upper levels of the Tm³⁺, Ho³⁺, and Er³⁺ lasers in the 2-3 µm region are typically in the range of 5-15 ms. Quasi-cw pulses with durations longer than those available commercially from laser diode arrays (≈200 µs) are therefore required to take advantage of the higher peak power.

With longer pulses the wavelength chirp (increase in diode laser wavelength with temperature) becomes a serious problem. Haden et al.²⁵ have demonstrated pulse durations as long as 1 ms with a chirp of 4 nm by using an improved thermal design. They estimate that longer pulses, possibly as long as 10 ms with a chirp as small as 5 nm, may be possible.

Future research directions

Diode pumping has proven to be an efficient means of producing laser emission in the 2 to 3 μ m region. As the available power from diode laser arrays increases and the price per unit power decreases, these lasers will become more important for practical applications. Diode pumped lasers are already becoming competitive in applications where weight, size, and ruggedness are important issues.

Most of the diode-pumped solid-state lasers developed to date have used AlGaAs diode arrays as the pump source. Significant progress in diode laser devices using other semiconductor materials is beginning to have an impact. For example, the recent development of higher power InGaAs strained-layer quantum well lasers in the range 0.9-1.1 μ m should allow higher efficiency diode-pumped operation of the Er³⁺ 2.8 μ m laser.²⁶ Extended pulse durations for quasi-cw diode laser devices will also have an impact on the development of solid state lasers in the 2-3 μ m region.

In addition to the practical applications, resonant pumping allows a more detailed study of processes such as cooperative upconversion and excited state absorption that are of scientific interest. The effects of physical processes such as these are more easily isolated when single electronic states can be excited. In turn, a more complete understanding of these effects leads to techniques for optimizing the performance of the solid state laser devices that are influenced by, and sometimes rely heavily upon, these effects.

REFERENCES

- 1. T. Y. Fan and R. L. Byer, IEEE J. Quantum Electron. 24, 895, 1988.
- 2. CRC Handbook of Laser Science and Technology, Vol. I, M.J. Weber, ed., CRC Press, Boca Raton, Fla., 1982.
- 3. J. A. Curcio and C. C. Petty, J. Opt. Soc. Am. 41, 302, 1951.
- 4. C. W. Robertson and D. Williams, J. Opt. Soc. Am. 61, 1316, 1971.
- 5. D. L. Sipes, Appl. Phys. Lett. 47, 74, 1985.
- 6. W. Hughes, A. Hays, D. DiBiase, J. Kasinski, and R. Burnham, in Technical Digest, Advanced Solid State Lasers, Optical Society of America, Washington, D.C., 1990, paper TuA1.
- T. M. Baer, D. F. Head, and P. Gooding, in Technical Digest, Advanced Solid State Lasers, Optical Society of America, Washington, D.C., 1990, paper TuB1.
- 8. T.Y. Fan and R. L. Byer, IEEE J. Quantum Electron. 23, 605, 1987.
- 9. W. P. Risk, J. Opt. Soc. Am. B 5, 1412, 1988.
- G. J. Kintz, R. Allen, and L. Esterowitz, in Technical Digest, Conference on Lasers and Electro-Optics, Optical Society of America, Washington, D.C., 1988, paper FB2.
- 11. R. C. Stoneman and L. Esterowitz, Opt. Lett. 15, 486, 1990.
- R. C. Stoneman and L. Esterowitz, proceedings of the Lasers and Electro-Optics Society Annual Meeting, IEEE, 1989, paper ELT2.3.
- 13. R. Allen, L. Esterowitz, L. Goldberg, J. F. Weller, and M. Storm, Electron. Lett. 22, 947, 1986.
- T. Y. Fan, G. Huber, R. L. Byer, in Technical Digest, Conference on Lasers and Electro-Optics, Optical Society of America, Washington, D.C., 1987, paper FL1.
- 15. T. Y. Fan, G. Huber, R. L. Byer, and P. Mitzscherlich, IEEE J. Quantum Electron. 24, 924, 1988.
- 16. G. J. Kintz, L. Esterowitz, R. Allen, Electron. Lett. 23, 616, 1987.
- 17. H. Hemmati, Appl. Phys. Lett. 51, 564, 1987.
- G. J. Kintz, L. Esterowitz, and R. Allen, in Technical Digest, Tunable Solid State Lasers, Optical Society of America, Washington, D.C., 1987, paper MC2.
- 19. H. Hemmati, Opt. Lett. 14, 435, 1989.
- M. D. Thomas, H. H. Zenzie, J. C. McCarthy, E. P. Chicklis, and H. P. Jenssen, in Technical Digest, Conference on Lasers and Electro-Optics, Optical Society of America, Washington, D.C., 1987, paper THJ5.
- R. C. Stoneman, G. H. Rosenblatt, and L. Esterowitz, in Tunable Solid State Lasers, Vol. 5 of OSA Proceedings Series, M. L. Shand and H. P. Jenssen, eds., Optical Society of America, Washington, D.C., 1989, p. 157.
- H. Chou and H. P. Jenssen, in Tunable Solid State Lasers, Vol. 5 of OSA Proceedings Series, M. L. Shand and H. P. Jenssen. eds., Optical Society of America, Washington, D.C., 1989, p. 167.
- G. J. Kintz, R. Allen, and L. Esterowitz, Appl. Phys. Lett. 50, 1553, 1987.
- 24. G. J. Kintz, L. Esterowitz, G. H. Rosenblatt, and R. C. Stoneman, proceedings of the Lasers and Electro-Optics Society Annual Meeting, IEEE, 1988, paper EL1.6.
- J. Haden, G. Harnagel, and J. Endriz, Postdeadline Papers, Advanced Solid State Lasers, Optical Society of America, Washington, D.C., 1990, paper TuPD8.
- R. C. Stoneman, J. G. Lynn, and L. Esterowitz, in Technical Digest, Conference on Lasers Electro-Optics, Optical Society of America, Washington, D.C., 1990, paper JWB1.