Visible cooperative upconversion laser in Er:$LiYF_4$

P. Xie and S. C. Rand

Division of Applied Physics, 1049 Randall Laboratory, University of Michigan, Ann Arbor, Michigan 48109-1120

Received March 30, 1992

We report trio upconversion laser action in Er:$LiYF_4$ with excitation at 1.55 μm resulting in cw emission at 0.55 μm to a temperature of 95 K and contrast its performance with earlier green erbium upconversion lasers.

Currently there is interest in developing compact short-wavelength laser sources for display and data-storage applications. Wide-gap semiconductor diode lasers,[1] harmonic generation by phase matching[2] or quasi-phase matching,[3] and upconversion lasers in bulk media[4] and fibers[5] are three promising approaches. However, wide-gap semiconductors present growth and doping problems that to date have prevented room-temperature operation by current injection. Harmonic generation requires critical alignment in bulk crystals and suffers reduced efficiencies in fibers and slab waveguides. Upconversion lasers depend on complex internal dynamics to upgrade photon energy by mechanisms that are themselves still the subject of intense inquiry. Hence, many questions remain to be answered before limitations of these various approaches can be assessed fully.

Initial upconversion laser research revealed low efficiencies and requirements for liquid-helium cryogenics. However, many different mechanisms of upconversion exist, and high-efficiency, high-temperature continuous operation is undoubtedly achievable in suitable media with appropriate techniques. To date, high efficiency and room-temperature operation have been demonstrated only under separate circumstances in selected laser crystals[6] and fibers,[5] respectively. In this Letter we explore capabilities of a cw cooperative upconversion laser that attains threefold upconversion of pump photon energy, 10% efficiency, and operation at temperatures as high as 95 K in an open-cavity configuration. Our device operates uniquely through cooperative upconversion, one of three known types of upconversion that may be categorized broadly by the preponderance of multiphoton,[6] avalanche,[7] or cooperative[8] processes. In the present case, inversion is shown to be due entirely to a cooperative energy-sharing process involving three atoms, similar in nature to the operating principle of a monolithic Er:CaF_2 trio laser that we reported previously.[9] By introducing a three-mirror astigmatically compensated cavity with Er:$LiYF_4$ as the gain medium, we have been able to study cavity losses in a fashion not possible in the original trio laser, to make precise assignments of excitation and emission dynamics, and to extend operation for what is to our knowledge the first time into the visible spectral region. We have found that the trio mechanism furnishes exceptional efficiency and stability even when the single-pump laser is tuned to the energy level in which the predominant trio interaction takes place.

Construction of a three-mirror cavity was motivated by unsuccessful attempts with 5% Er:CaF_2 to obtain green laser action in a monolithic design with expensive dielectric mirrors deposited directly on the ends of the gain medium. In that experiment, excited-state absorption was presumed to quench laser oscillation. The Brewster-oriented (θ_B = 55.6°) gain medium in the present experiment consisted of a 3-mm-thick crystal of 5% Er:$LiYF_4$ with its optic axis parallel to the crystal surface in the plane of incidence of the horizontally polarized pump field. This orientation permits gain extraction on both π- and σ-polarized transitions. The laser crystal was suspended on a cold finger in vacuum, together with all optics except for the external flat output mirror. A refractive index[10] of $n_e = 1.462$ mandated an interarm angle of $\theta = 26.2°$ for compensation of the astigmatism at 544 nm, introduced by the two 5-cm-radius spherical mirrors that were disposed symmetrically with respect to the gain medium. A tunable cw NaCl color-center laser provided resonant excitation of the $^4I_{13/2}$ level of Er^{3+} at 1.55 μm.

To study the inversion mechanism in this system, we measured the time and power dependencies of upconversion emission at two wavelengths, as shown in Fig. 1(a). The time-domain measurements reveal evolution of population in the $^4S_{3/2}$ and $^4I_{11/2}$ levels after impulse excitation of the $^4I_{13/2}$ level. They were made with an S-1 photomultiplier terminated in 50 Ω and amplified with a transimpedance amplifier with a bandwidth of dc to 13 MHz. Results in Fig. 1(a) show that population does not appear in either the $^4I_{11/2}$ or the $^4S_{3/2}$ state during the excitation pulse itself. Upconversion emission grows and peaks first in the $^4S_{3/2}$ state on a time scale much longer than the 100-μs pulse width. Its slow appearance cannot be accounted for by processes involving the absorption of more than one photon from the incident pulse by a single atom. Nevertheless, upconversion fluorescence from the $^4S_{3/2}$ level increases as the cube of incident intensity, when saturation of the pump transition is carefully avoided [Fig. 1(b)].

As in the Er:CaF_2 trio laser,[9] multiphoton excita-

0146-9592/92/171198-03$5.00/0

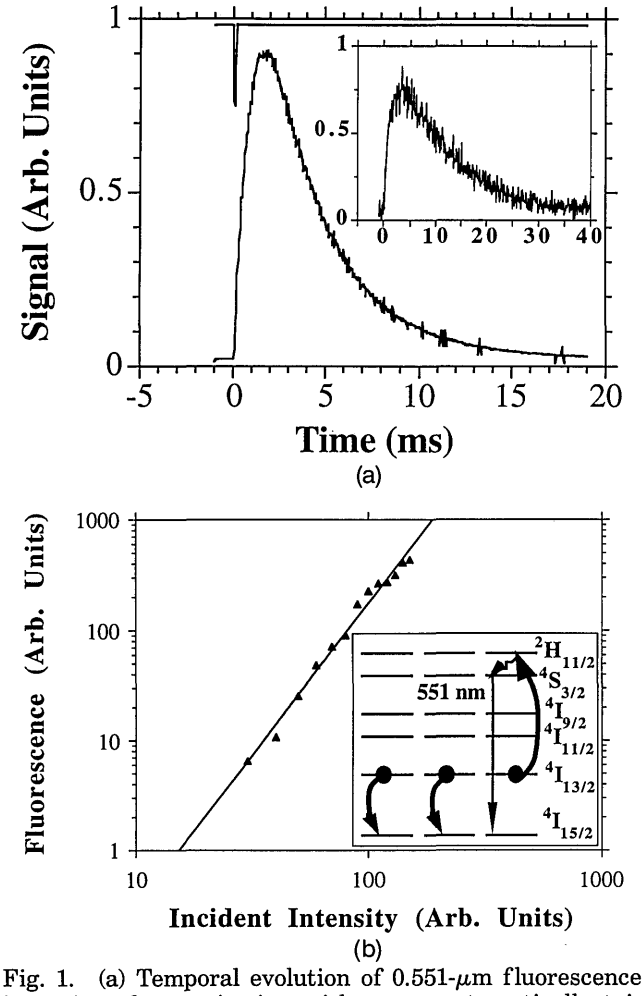

Fig. 1. (a) Temporal evolution of 0.551-μm fluorescence intensity after excitation with an acousto-optically tailored 100-μs infrared pulse (inverted upper trace) of peak intensity 2 kW/cm^2. The inset shows the evolution of pair-excited 0.98-μm fluorescence from the $^4I_{11/2}$ state under identical conditions. (b) Dependence of upconversion fluorescence intensity on incident intensity below 0.2 W/cm^2. The solid curve shows the power law with a best-fit slope of 2.8 $\pm$ 0.1. The inset shows the schematic of the $^4I_{13/2}$ trio process in erbium.

tion processes are ruled out in our Er:LiYF$_4$ laser by the time-dependence of fluorescence excitation shown in Fig. 1(a) at power levels of this experiment. The cubic intensity dependence and long fluorescence rise time, approaching the lifetime of the $^4I_{13/2}$ level, can be explained only by a trio process involving promotion of one Er^{3+} ion to the $^2H_{11/2}$ manifold at the expense of deexcitation of two $^4I_{13/2}$ neighbors. The inset of Fig. 1(b) illustrates the coopertive dynamics schematically. The predominance of this cooperative interaction is quite different from behavior observed when excitation wavelengths shorter than that of the first Er^{3+} resonance are used. It is important to recognize that this difference arises from the fact that upconversion dynamics depend strongly on the details of the initially prepared state, equivalent to the initial conditions of the process. Here we have deliberately prepared the ensemble of Er ions in the lowest excited state to isolate the multiatom or cooperative upconversion process and study its capabilities alone.

Upconversion laser experiments were performed with output couplers ranging between 2% and 16% transmission. Output versus input power at 9 K is illustrated in Fig. 2(a) for various output couplings. Thresholds in the range 20–50 mW were observed, and slope efficiency with 16% coupling was 11.6% up to a pump-limited maximum output power of 33 mW. At the intensities required to reach laser threshold, green fluorescence became strongly saturated. Hence, not surprisingly, laser output depended linearly rather than nonlinearly (as in Ref. 8) on pump intensity. Optimum output coupling, determined from a plot of output power versus coupler transmission [Fig. 2(b)] was ~9%. Internal gain and loss coefficients were calculated to be 22% and 2.0% per pass, respectively.

At 77 K, excitation of any Er absorption line in the range 1.45–1.55 μm resulted in laser output at $\lambda = 0.5516$ μm, corresponding to the $^4S_{3/2}(1)$–$^4I_{15/2}(4)$ transition with mixed σ and π polarization owing to Kramer's degeneracy. This laser transition terminates in the fourth $^4I_{15/2}$ Stark level from the bottom of the manifold, 200 cm^{-1} above the true ground state, and we found the maximum operating temperature to be 95 K on this line. For temperatures

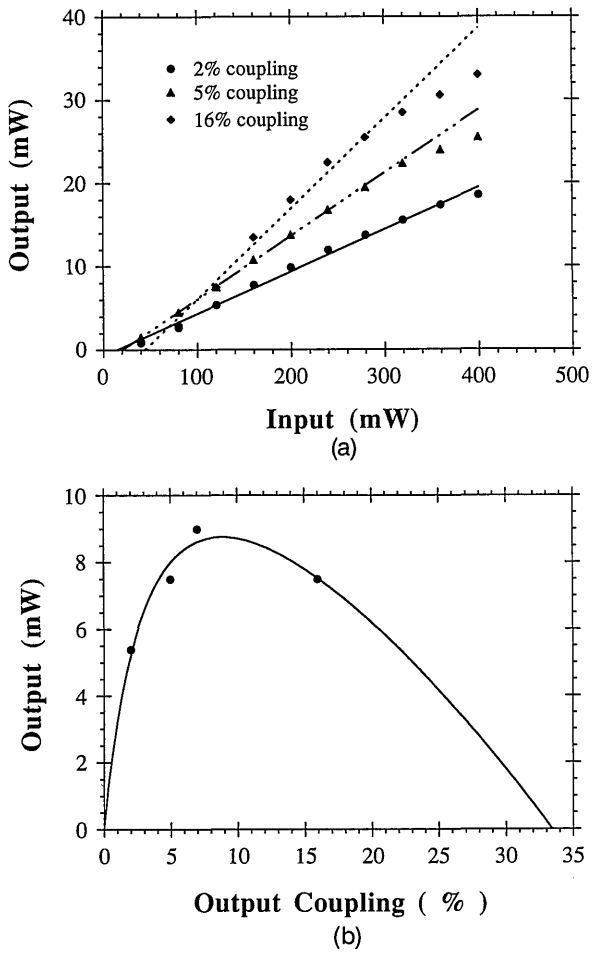

Fig. 2. (a) Laser output (0.5440 μm) versus input power at 9 K. (b) Output power versus output mirror reflectivity at a fixed pump power of 120 mW focused to a spot of radius 18 μm in the gain medium. The solid curve is a least-squares fit to the analytic output power formula.[11]

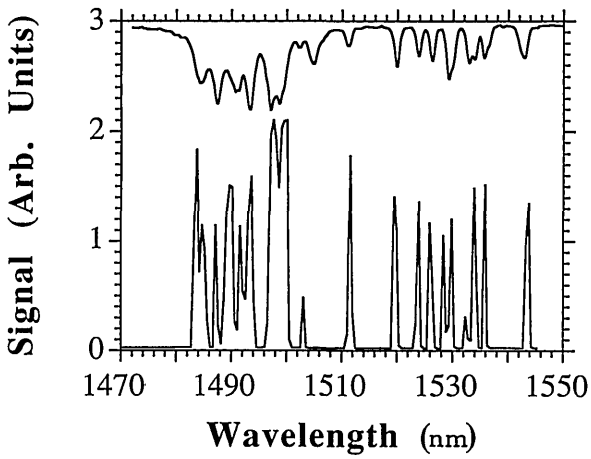

Fig. 3. Excitation spectrum of upconversion laser action in Er:$LiYF_4$ at 77 K and the erbium absorption spectrum (inverted curve). The absorptive transition from the lowest ground state to the lowest level of the first excited state corresponds to the line at $\lambda = 1.530$ μm.

below 15 K, the output wavelength shifted to $\lambda = 0.5440$ μm, corresponding to the $^4S_{3/2}(1)$–$^4I_{15/2}(6)$ transition. The laser-excitation spectrum is given in Fig. 3. All seven (Kramers degenerate) $^4I_{15/2}(1)$–$^4I_{13/2}(n)$ Stark components may be readily identified[12] in absorption and excitation in the figure, confirming that laser action results even from pumping into the $^4I_{13/2}(1)$ level ($\lambda_{ex} = 1.530$ μm) in which the trio interaction originates.

True cw operation was obtained under all pumping conditions of the green trio laser. This contrasts with recent observations of sustained oscillations in the pair-pumped Er:CaF_2 laser, which is also pumped cooperatively.[13] Superior stability of the trio laser may be related to the fact that the $^4S_{3/2}$ upper-state lifetime[14] is much shorter than that of the $^4I_{11/2}$ upper state of the pair laser. Our cw operation also contrasts sharply with self-pulsing observed on the 551-nm transition in Er:$LiYF_4$ when alternative excitation methods are used,[6] behavior that has been linked to excited-state absorption from the $^4I_{13/2}$ level.[4] Because the cooperative trio process relies on significant occupation of the $^4I_{13/2}$ level, this striking difference in behavior cannot result from the absence of self-absorption. Rather, the observed stability can only originate from different initial conditions and predominance of the cooperative upconversion process in our experiment. The steady output observed here must arise from the slow time constant and inherently sluggish response of the cooperative upconversion mechanism to changes in intracavity photon density compared with fast response times of multiphoton absorption mechanisms of upconversion.

In summary, we have demonstrated cooperative upconversion laser operation in the green spectral region with a cavity configuration amenable to the study of laser dynamics and the measurement of internal losses. We have confirmed spectroscopically that excitation mechanisms other than cooperative upconversion contribute negligibly to the inversion of this trio laser. True cw visible upconversion laser operation can be sustained by the spontaneous trio interaction alone, even when the pumped level coincides with the level in which the trio interaction originates. Cooperative pumping permits surprisingly efficient operation and apparently stabilizes the 551-nm inversion against self-Q-switching.

The authors gratefully acknowledge sponsorship by the U.S. Air Force Office of Scientific Research and equipment loans by B. Couillaud of Coherent, Inc., and J. Lekvitch of IntraAction.

References

1. M. Haase, J. Qiu, J. DePuydt, and H. Cheng, Appl. Phys. Lett. **59,** 1272 (1991); H. Jeon, J. Ding, W. Patterson, A. Nurmikko, W. Xie, D. Grillo, M. Kobayashi, and R. Gunshor, Appl. Phys. Lett. **59,** 3619 (1991).
2. L. Goldberg and M. K. Chun, Appl. Phys. Lett. **55,** 218 (1989); U. Österberg and W. Margulis, Opt. Lett. **12,** 57 (1987).
3. J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. **127,** 1918 (1962); N. Bloembergen and A. J. Sievers, Appl. Phys. Lett. **17,** 483 (1970); A. Feisst and P. Koidl, Appl. Phys. Lett. **47,** 1125 (1985).
4. L. F. Johnson and H. J. Guggenheim, Appl. Phys. Lett. **19,** 44 (1971).
5. J. Y. Allain, M. Monerie, and H. Poignant, Electron. Lett. **26,** 168, 262 (1989).
6. A. J. Silversmith, W. Lenth, and R. M. Macfarlane, Appl. Phys. Lett. **51,** 1977 (1987); R. M. Macfarlane, F. Tong, A. J. Silversmith, and W. Lenth, Appl. Phys. Lett. **52,** 1300 (1988); D. C. Nguyen, G. E. Faulkner, and M. Dulick, Appl. Opt. **28,** 3553 (1989); R. A. McFarlane, Appl. Phys. Lett. **54,** 2301 (1989).
7. M. E. Koch, A. W. Kueny, and W. E. Case, Appl. Phys. Lett. **56,** 1083 (1990).
8. P. Xie and S. C. Rand, Opt. Lett. **15,** 848 (1990).
9. P. Xie and S. C. Rand, Appl. Phys. Lett. **57,** 1182 (1990).
10. D. E. Castleberry and A. Linz, Appl. Opt. **14,** 2056 (1975).
11. A. E. Siegman, *Lasers* (University Science, Mill Valley, Calif., 1986), Chap. 12.
12. S. M. Kulpa, J. Phys. Chem. Solids **36,** 1317 (1975).
13. P. Xie and S. C. Rand, in *Conference on Lasers and Electro-Optics,* Vol. 12 of 1992 Technical Digest Series (Optical Society of America, Washington, D.C., 1992), paper CFE4.
14. S. A. Pollack, D. B. Chang, and M. Birnbaum, Appl. Phys. Lett. **54,** 869 (1989).