Continuous-wave trio upconversion laser

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We report operation of what we believe is the first continuous-wave laser which relies exclusively on cooperative upconversion by coupled ion trios to achieve population inversion.

The first laser to operate with an output wavelength shorter than that of the excitation was a pulsed erbium laser reported by Johnson and Guggenheim.[1] Since this early work, a variety of continuous-wave lasers have been reported in which upconversion occurs by different mechanisms, and inversions of upconverted energy levels have been sustained by avalanche processes,[2,3] direct pair-pumping,[4] and multiphoton absorption with variable contributions from energy transfer.[5,6] To date, cw upconversion lasers have been operated in Pr-, Er-, Tm-, and Nd-doped solids, exhibiting surprisingly high efficiencies (up to 30%) in simple and robust, monolithic designs. All can potentially be pumped by diode lasers, and furnish wavelengths typically twice as short as diode lasers. With the advent of room-temperature operation these devices should therefore offer attractive alternatives to harmonic generation schemes[7] for short-wavelength source applications.

In this letter we show that in addition to the known avalanche, two-atom and multiphoton upconversion mechanisms, steady-state inversions can also be maintained by cooperative transitions of atomic trios. Spontaneous cooperative fluorescence due to weakly coupled trios of rare-earth dopants in dielectric crystals has been studied previously.[8] However, to our knowledge this is the first report of cw stimulated emission sustained by trio-pumping alone. Furthermore the 0.855 μm upconversion laser reported here achieves remarkably efficient cw oscillation on a self-terminating transition.

A simple, but realistic picture of the basic dynamics in the current work is obtained with a six-level model of trivalent erbium, indicated schematically in Fig. 1. Here each manifold is represented by a single level and cooperative transitions are permitted only in level two, which is considered to be very long lived at low excitation densities. In reality level three of Er^{3+} is also long lived, but its population arises entirely from level two pair processes. Hence it contributes to effective losses in the erbium pair laser[4] and to quartic upconversion, but not to the cubic upconversion processes of interest here. In our model we draw on earlier results which justify omission of two-photon absorption source terms for level four population.[4] However we include the possibility of direct absorption of pump photons by ions maintained in level four by cooperative pair upconversion to reach the upper laser level. Diagonal density matrix elements for this nonlinear system are

$$\frac{d}{dt}\rho_{11} = \gamma_{51}\rho_{55} + \gamma_{41}\rho_{44} + \gamma_{31}\rho_{33} + \gamma_{21}\rho_{22} + \alpha\rho_{22}^2 + 2\beta\rho_{22}^3 - B_{12}I(\rho_{11} - \rho_{22}) \quad (1)$$

$$\frac{d}{dt}\rho_{22} = \gamma_{52}\rho_{55} + \gamma_{42}\rho_{44} + \gamma_{32}\rho_{33} - \gamma_{21}\rho_{22} + \kappa_{25}(\rho_{55} - \rho_{22}) - 2\alpha\rho_{22}^2 - 3\beta\rho_{22}^3 + B_{12}I(\rho_{11} - \rho_{22}) \quad (2)$$

$$\frac{d}{dt}\rho_{33} = \gamma_{53}\rho_{55} + \gamma_{43}\rho_{44} - \gamma_3\rho_{33} \quad (3)$$

$$\frac{d}{dt}\rho_{44} = \gamma_{54}\rho_{55} - \gamma_4\rho_{44} + \alpha\rho_{22}^2 - B_{46}I(\rho_{44} - \rho_{66}) \quad (4)$$

$$\frac{d}{dt}\rho_{55} = \gamma_{65}\rho_{66} - \gamma_5\rho_{55} - \kappa_{25}(\rho_{55} - \rho_{22}) \quad (5)$$

$$\frac{d}{dt}\rho_{66} = -\gamma_6\rho_{66} + \beta\rho_{22}^3 + B_{46}I(\rho_{44} - \rho_{66}), \quad (6)$$

where the spontaneous relaxation rate between level i and j is given by γ_{ij} and B_{ij} is the induced rate between i and j. The pair and trio upconversion coefficients are α and β,

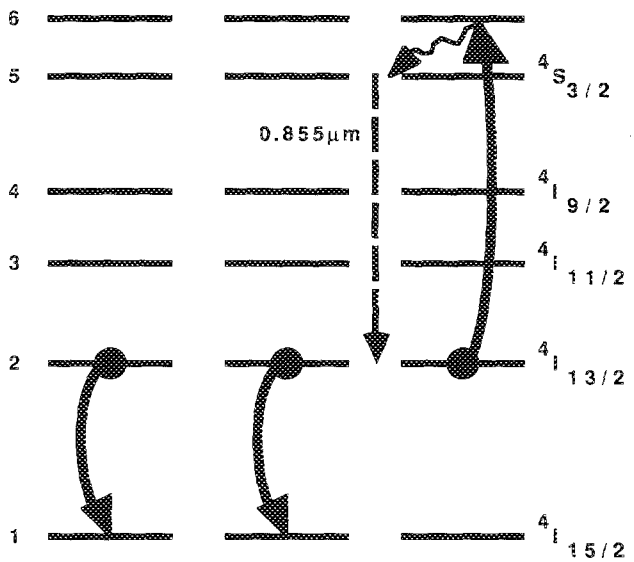

FIG. 1. Three-atom cooperative upconversion process responsible for steady-state inversion of level $^4S_{3/2}$ with respect to level $^4I_{13/2}$ in Er:CaF_2. The initial excited state, with three dopant ions in the $^4I_{13/2}$ state, is prepared by cw irradiation at 1.51 μm on the $^4I_{15/2} - {}^5I_{13/2}$ absorption resonance of trivalent erbium. The curved arrows indicate the dynamics schematically, with two atoms returning to the ground state while one is promoted to the upper laser level.

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respectively. κ_{ij} is the rate of stimulated emission from i to j and γ_i is the total decay rate of level i.

In Eq. (2), there is a quadratic loss term corresponding to a pair process and a cubic term for trio upconversion. In the former process two ions are lost for each transition, requiring the indicated factor of 2. One ion returns to ground while the other is upconverted to level four. In the trio process three atoms leave level two, requiring a factor of 3 in the cubic loss coefficient. Two of the excited ions return to ground and one is upconverted directly to level six with subsequent rapid decay to level five. Of central importance here is the result that in the absence of cavity losses or pair upconversion a steady-state inversion between levels two and five can be maintained by trio upconversion alone if

$$\beta \geqslant \frac{\gamma_5(\gamma_5+3\gamma_6)^2}{\gamma_6^2}\left(1+\frac{(\gamma_2+\gamma_{51}+2\gamma_5)\gamma_6}{(\gamma_5+3\gamma_6)B_{12}I}\right)^2. \qquad (7)$$

Conditions such as (7) for steady-state inversion in coupled-atom systems do not guarantee stable cw operation. The oscillator system is highly nonlinear and may exhibit unstable or even chaotic states. Hence a stability analysis is required to ensure theoretically stable operation of the laser. We have analyzed trio laser stability with a simplified four-level model (omitting levels three and four) by introducing small perturbations and linearizing system response near steady-state conditions. The Routh–Hurwitz criterion[9] then predicts stable oscillation whenever the highest state, the trio-pumped state, is short-lived compared to other levels in the system. This condition is well met in Er^{3+}.

To demonstrate an erbium trio laser experimentally, a 3-mm-thick crystal of 5% Er:CaF_2 was prepared with one flat surface and one convex surface of radius 2.5 cm. Both surfaces were antireflection coated in the range 1.4–1.6 μm. Additionally, in the range 0.8–0.9 μm, the curved surface was coated for total reflection ($R>99.9\%$) and the flat served as a 0.5% output coupler. The sample was pumped longitudinally at liquid-nitrogen temperature with a cw NaCl color center laser focused by a 5 cm lens, and it absorbed 74% of incident light at 1.51 μm. For incident intensities above a threshold of 10 mW, TEM_{00} laser emission was observed at 0.855 μm as shown in Fig. 2. A maximum of 64 mW cw output was obtained for 235 mW of absorbed pump power, for an overall efficiency of 26% (theoretical maximum is 60%) and a slope efficiency of 28%. No evidence of saturation spiking behavior was observed and amplitude fluctuations were 15 dB below the output level within the measurement bandwidth (dc 1 MHz).

The intensity dependence of upconversion fluorescence shown in Fig. 3 reveals that level five is populated by a process varying with the cube of the incident intensity. This is a key result of the present work, since with our method of excitation, there are only two possible channels for populating upper laser level five by a cubic process. One channel is a trio process and the other is absorption of a pump photon by pair-pumped ions in level four. Additional conceptual possibilities related to ground state

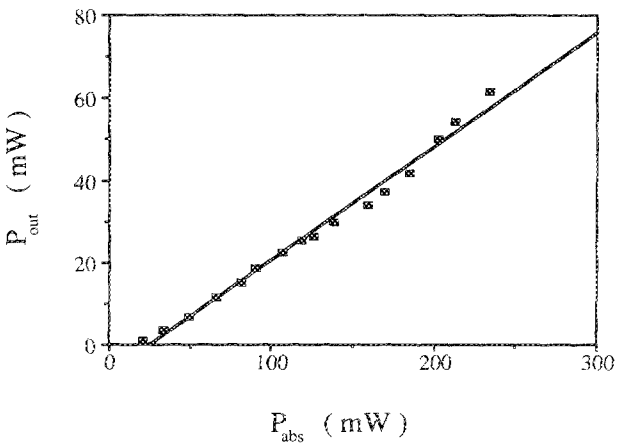

FIG. 2. Linear plot of cw trio-pumped laser output vs absorbed incident power. The solid curve is a linear guide to the eye.

two-or three-photon absorption are ruled out by the absence of prompt fluorescence from levels three and four, as discussed in our earlier paper.[4] We distinguished between the two remaining possibilities with time-resolved fluorescence measurements shown in the next figure.

Figure 4 shows the time dependence of fluorescence at 0.855 μm from the upper laser level, monitored with a fast photomultiplier following pulsed excitation below threshold. A rectangular pulse of 100 μs duration and 8 mW peak power was selected acousto-optically from the cw pump beam for this purpose. Signal averaging of 4000 scans was used to improve signal-to-noise ratio on a time scale of milliseconds with 1 MHz bandwidth.

Two components are evident in the experimental curve. The first is a prompt component with a rise time equal to the pulse duration, followed by a subsequent, rapid decay. This component can only be due to pump absorption by pair-upconverted ions in state $^4I_{9/2}$, since the signal decays when the pulse ends and yet two-photon absorption contributions to the $^4I_{9/2}$ population are insignificant under these conditions (see Fig. 4 inset and Ref. 4). This component therefore corresponds to the pair-mediated channel. The second component rises slowly,

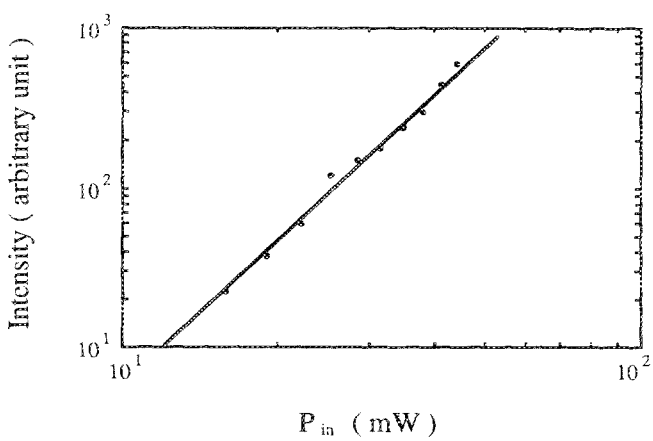

FIG. 3. Log-log plot of level five fluorescence vs excitation intensity. The solid curve illustrates a cubic dependence for 0.855 μm upconversion emission.

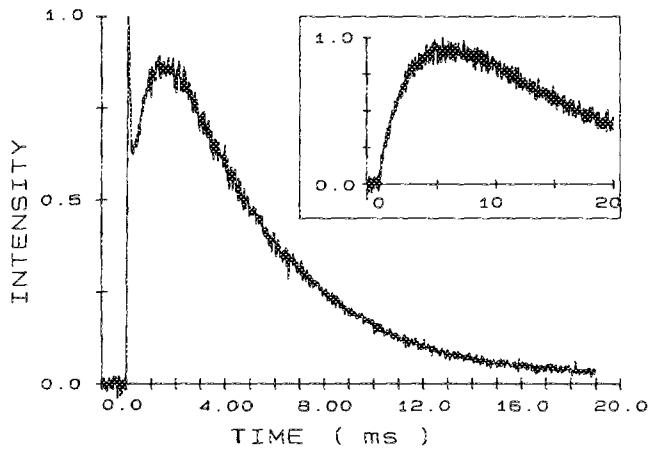

FIG. 4. Fluorescence intensity at 855 nm vs time, due to sample excitation by an acousto-optically tailored pulse of 100 μs duration and 8 mW peak intensity. Prompt emission is observed as a spike during the applied pulse. A slowly rising component appears long after the pulse terminates. The relative importance of these two excitation channels can be assessed by comparing areas under component curves. The inset shows the absence of prompt emission at 0.98 μm, ruling out level four two-photon and level six three-photon excitation.

reaching its maximum long after the excitation pulse is over. The only remaining process capable of furnishing cubic upconversion to $^4S_{3/2}$ is one involving three excited $^4I_{13/2}$ ions. Hence the second component corresponds to this trio contribution. Notice that long before the rate of trio upconversion reaches its peak in the steady state its contribution to upper laser level population exceeds the pair-mediated portion by a factor of 40, estimated from relative areas under the curve.

The overall efficiency of this trio-pumped laser is 26% at 235 mW pump power. Its slope efficiency is nearly 30%, using a linear approximation for the output curve versus input, and as mentioned earlier its threshold is only 10 mW. This trio laser consequently has much higher efficiency and lower threshold than the pair-pumped erbium laser,[4] a somewhat unexpected result due at least partly to the absence of water absorption at the emission wavelength as well as higher cavity Q. However the cubic depletion rate of the lower level may also play a significant role. In addition, amplitude fluctuations are much smaller in trio laser output, a result possibly due to the very short lifetime of level six which discourages back transfer.

In summary, we have demonstrated a continuous-wave laser pumped by cooperative upconversion from trios of excited, coupled erbium ions in CaF_2. Its unoptimized efficiency is high, indicating that trio mechanisms by themselves can be exploited for the development of novel, short-wavelength solid-state lasers. The erbium trio laser is remarkable in that cw operation is achieved on a self-terminating transition with lower laser level pumping. Hence this nonlinear pumping scheme not only offers the prospect of new upconversion lasers, but also of cw rare-earth lasers in other highly doped crystals on previously unusable, self-terminating transitions.

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