Continuous-wave, fourfold upconversion laser

P. Xie and S. C. Rand
Division of Applied Physics, 1049 Randall Laboratory, University of Michigan, Ann Arbor, Michigan 48109-1120

We report a new Er^{3+}:$LiYF_4$ cryogenic upconversion laser pumped by a fourfold upconversion process. Excitation at 1.5 μm results in laser emission at 701.5 nm on a transition with an upper state at nearly four times the pump photon energy. Mechanisms and the concept of cooperative upconversion enhancement are examined.

Recent demonstrations[1-5] of room-temperature upconversion lasers have stirred interest in their potential as practical sources of short wavelength radiation for display and data storage applications, as well as for communications and ultrashort pulse generation at visible and ultraviolet wavelengths.[6] To date however, little consideration has been given to the relative merits of various fundamental mechanisms for achieving upconversion, basic limits to achievable degrees of upconversion (ratio of upper laser level energy to incident photon energy), or to questions of stability of the highly nonlinear pumping processes involved in these devices. In this article we discuss two of these topics in the context of new results demonstrating a continuous-wave, fourfold upconversion laser. The important third issue of stability of solid state lasers in which nonlinear cooperative dynamics play an important role has been considered by Xie[7] and will be published separately.[8]

Upconversion fluorescence observed in Er:$LiYF_4$ due to irradiation with a continuous-wave (cw) NaCl laser at 1.5 μm at liquid-helium temperature is shown in Fig. 1(a). The upconversion mechanisms responsible for fluorescent emissions near 850 and 550 nm have been studied in past work and arise from cooperative (multi-atom) energy transfer processes[9] when cw excitation is restricted to 1.5 μm.

Emissions near 410, 650, 702 nm from higher lying states have not yet been studied as thoroughly, but are sufficiently intense to draw attention as potential laser candidates. The two near-ultraviolet lines at 407 and 413 nm arise from transitions with upper levels at roughly four times the incident photon energy, as indicated in Fig. 1(b). The fluorescent emission at 702 nm arises from Stark components of the $^2H_{9/2} \rightarrow {^4I}_{11/2}$ transition.

Laser experiments were performed in a three-mirror, astigmatically compensated[9] cavity consisting of two 5 cm radius total reflectors and an output mirror with 97% reflectively at 702 nm. A 3-mm-thick crystal of 5% Er:$LiYF_4$ inserted at Brewster's angle within the focusing arm of the laser served as the gain medium. Its optic axis was oriented parallel to the crystal surface in the plane of incidence of horizontally polarized pump radiation and the crystal was suspended on a cold finger in vacuum. The laser emission spectrum at 701.5 nm, assigned to the $^2H_{9/2}$ (1) $\rightarrow {^4I}_{11/2}$(3) transition,[10] and the variation of output power with input are shown in Fig. 2. The overall efficiency was 0.06% and observed slope efficiency was 0.09%.

The excitation spectrum of laser emission revealed a one-to-one correspondence with erbium absorption wavelengths in the 1.5 μm region. This result is shown in Fig. 3 and is similar to earlier findings for pair[9] and trio[11] lasers, but contrasts the restrictive wavelength dependence expected for multiphoton upconversion processes. It therefore has important implications for the inversion mechanism of the fourfold laser.[12]

For multiphoton absorption to be effective, several ground and excited state absorption (ESA) frequencies must overlap sharply. Hence, excited state resonances should appear in the excitation spectrum when this mechanism is operative.[13] For cooperative upconversion, only a single pump transition is relevant, terminating anywhere within the metastable manifold in which cooperative inter-
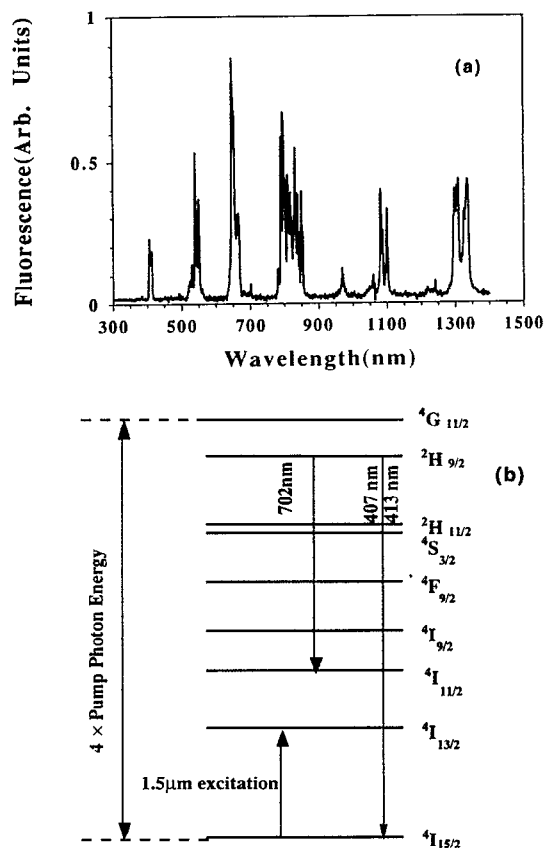

FIG. 1. (a) Upconversion fluorescence spectrum observed in Er^{3+}:$LiYF_4$ with excitation at $\lambda_{ex} = 1.5$ μm ($T = 10$ K), uncorrected for instrumental response. (b) Schematic diagram of Er^{3+} energy levels, showing emission at 410 and 701.5 nm from $^2H_{9/2}$ at nearly four times the incident photon energy.
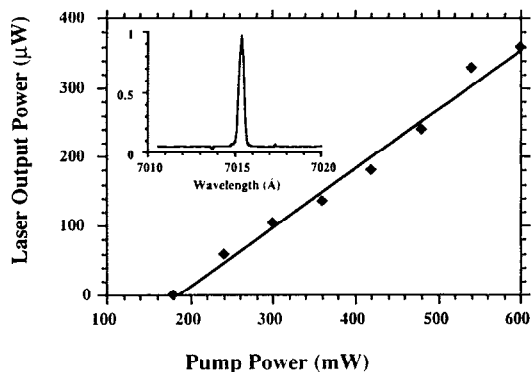
FIG. 2. Output vs input power of the cw fourfold upconversion laser operating at 701.5 nm ($T = 10$ K). Inset: Laser emission spectrum.

FIG. 4. Mechanism for enhanced quantum efficiency in cooperative upconversion systems. Following an initial excitation step (by direct pumping or upconversion), a cascade from higher to lower excited states occurs with branching ratios of η_i at each step. Wiggly arrows indicate nonradiative decay. A given atom can emit more than one photon per η_0 excitation if the upconversion rate exceeds the spontaneous decay rate of the lowest excited state (and detailed balance is satisfied).

actions occur. In this case only ground state absorption resonances should appear in the excitation spectrum. The results in Fig. 3 therefore provide ample evidence that absorption of light by excited ions does not occur and that the present fourfold laser operates by cooperative means.

This argument also rules out mechanisms combining cooperative and ESA processes. For example, ESA of 1.5 μm light from $^4S_{3/2}$ or $^2H_{11/2}$ states populated by trio upconversion[11] could in principle reach the high lying $^2H_{9/2}$ state responsible for laser action in this work, but such transitions are off resonant by over 218 cm^{-1} in LiYF$_4$ for all Stark levels[10] and are not present in the excitation spectrum. ESA transitions from $^4S_{3/2}$ to $^4G_{11/2}$ are still farther off resonance. On the other hand, quartet upconversion can provide very nearly *resonant* excitation of the $^4G_{11/2}$ state lying immediately above the upper laser level [generating the fluorescence at 380 nm in Fig. 1(a)]. The calculated energy defects on the quartet transitions $^4I_{13/2}$ $(3,3,3,4) \rightarrow {}^4G_{11/2}(1)$ and $^4I_{13/2}(4,4,4,3) \rightarrow {}^4G_{11/2}(4)$, for example, are only 4 and 7 cm^{-1} in LiYF$_4$, respectively. The bracketed arguments of the initial state indicate Stark levels of the four interacting atoms, and the single argument of the final state gives the Stark level of the acceptor. Taken together with the close match between the excitation spectrum and ground state absorption (Fig. 3), this argues strongly against any contributions by ESA pro-
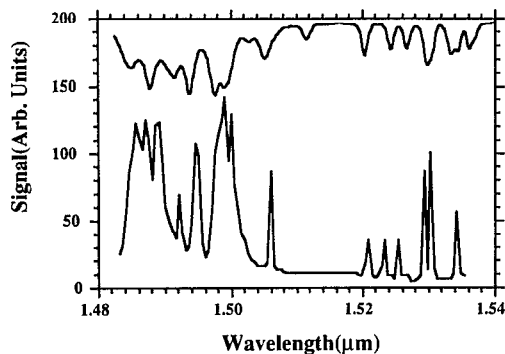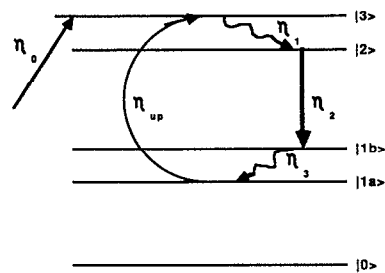

FIG. 3. Infrared excitation spectrum of fourfold upconversion laser emission at 701.5 nm ($T = 10$ K, lower trace) and, for comparison, the Er^{3+} absorption spectrum ($T = 9$ K, inverted).

cesses to $^2H_{9/2}$ population when 1.5 μm pumping is used.

The implication that laser operation may be sustained by a cooperative quartet process raises fundamental questions as to just how effective higher order *cooperative* upconversion processes can be in general. In this regard, we wish to point out that an enhancement mechanism exists for cooperative upconversion processes which can significantly extend their usefulness compared to multiphoton absorption. For emission terminating on or near levels in which strong cooperative interactions occur, no efficiency "penalty" is incurred for m-fold upconversion, where the integer m may be arbitrarily large. When upconversion emission terminates on an energy level which can renew cooperative upconversion, excited atoms are *recycled without external pumping* to enhance quantum efficiency by a factor of $(1 - \eta_1\eta_2\eta_3\eta_{up})^{-1}$, where η_{up} is the m-fold cooperative upconversion efficiency. η_1, η_2, and η_3 are branching ratios for the decay processes indicated in Fig. 4. Energy conversion between input wavelength λ_{in} and output at λ_{out} can be much higher than the maximum value $\eta_{max} = \lambda_{in}/\lambda_{out}(100/m)\%$ based on a picture in which only one of m atoms is upconverted following the absorption of m pump photons. Ignoring decay to the ground state, the energy efficiency takes on the enhanced value

$$\eta_E^* = (\lambda_{in}/\lambda_{out})\eta_0\eta_1(1 - \eta_1\eta_2\eta_3\eta_{up})^{-1} \qquad (1)$$

and $\eta_E^* > \eta_{max}$ when upconverted atoms reside long enough in their lowest excited state to participate repeatedly in upconversion. With recycling, energy efficiency can approach 100% [for $\eta_i = 1$ ($i = 0, 1...3$) with a negligible energy defect such that $\lambda_{out} = \lambda_{in}/(1 - \eta_{up})$].

In practice, theoretical enhancement of quantum efficiency by cooperative dynamics must be mitigated by losses due to an increasing number of intervening (radiative) levels on real atoms with increasing m. Branching ratios are typically less than unity. Also a statistical decrease is to be expected in the number of m-atom "clusters" with appropriate interatomic spacings, less than a nominal critical radius. However, the main conclusion is that a recycling mechanism exists whereby cooperative dynamics can mediate unexpectedly efficient upconversion emission for arbitrarily high degrees (m) of upconversion.

This has important implications not only for upconversion lasers, but also for other lasers in which cooperative nonlinear dynamics occur due to heavy rare-earth doping. One example is the 2.8 μm Er laser reported by Stoneman and Esterowitz.[14] These authors obtained an efficiency in practice which was marginally greater than the maximum theoretical quantum efficiency (ignoring cooperative dynamics). A calculation of enhanced quantum efficiency using the formula above, ignoring spontaneous radiative losses for simplicity, shows that for 980 nm pumping, complete recycling of terminal level population through pair upconversion results in a "renormalized" quantum efficiency of 2 (instead of 1) and a maximum energy efficiency of 70% (instead of 35%). Similar but less dramatic improvements due to cooperative enhancement of quantum efficiency are expected for 2.8 μm lasers pumped by other wavelengths.[8] Cooperative enhancement probably also accounts for the remarkable efficiency of the "self-terminating" Er trio laser at 855 nm.[11]

In the present experiment, we note that the 701.5 nm upconversion laser line terminates on the $^4I_{11/2}$ level of trivalent erbium. This level is known[15] to be effective in pair upconversion to the $^4F_{7/2}$ level, but is not the cooperative pumping level responsible for our inversion. The $^4I_{13/2}$ level, not the $^4I_{11/2}$ level, is responsible for cooperative pumping when excitation is tuned to 1.5 μm.[9,11] This situation therefore resembles, though it is not identical to, the general enhancement scheme of Fig. 4, and we suggest that enhanced upconversion efficiency through cooperative recycling may help account for successful operation of this laser, with its unprecedented (fourfold) degree of upconversion.

In summary, we have demonstrated operation of an upconversion laser in which population inversion is sustained in an upper laser level at nearly four times the incident photon energy,[16] through a fourfold upconversion process. The upconversion mechanism is thought to be exclusively cooperative in nature, arising from an atomic "quartet" interaction quite distinct from sequential energy transfer[13] or multiphoton absorption. The general notion of recycling of excited state population has been introduced and discussed as a possible mechanism for enhancing mth

order upconversion processes and accounting for successful operation of the present fourfold upconversion laser. Extension of this result to fourfold laser operation at 410 nm, 555 nm, and other wavelengths appears feasible. In the case of the 555 nm transition, the terminal level would be in the $^4I_{13/2}$ manifold, and especially strong cooperative enhancement of the efficiency may be possible.

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