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UPCONVERSION PROCESSES IN OPTICALLY PUMPED LASER CRYSTALS

H.P. JENSSEN

*Crystal Physics and Optical Electronics Laboratory, MIT,
Cambridge, MA 02139, U.S.A.*

INTRODUCTION.

Even though a not newly discovered phenomena, upconversion has gained renewed interest due to the 2.8 μm laser (1). This combined with the high inversion densities attainable with diode laser pumping, has led to the demonstration of cw, room temperature, operation of Er:LiYF₄ (2). It is expected that this will be only one of many similar lasers demonstrated in the near future. In this paper we will go back and examine earlier work on upconversion and relate that to the present Er laser. We will then examine other potential upconversion lasers.

EARLY WORK.

Upconversion is only a special case of the more general phenomena of energy transfer. Energy transfer was a process recognized early and the theory developed first by Foerster (3) and subsequently refined by Dexter (4). In short, energy transfer between two ions in a solid can take place due to the multipolar interaction between the ions. The transfer rate depends on the product of the oscillator strengths of the individual ions, their relative spatial separation and an overlap integral of the emission spectrum of the donor and the absorption spectrum of the acceptor. In mathematical form (for electric dipole-dipole interaction):

$$W_{tr} \propto \frac{f_d \cdot f_a}{R^6} \int I^d(em)(\lambda) \cdot I^a(abs)(\lambda) d\lambda$$

where:

W_{tr} is the transfer rate,

f_d, f_a are the donor and acceptor oscillator strengths

R is the distance between donor and acceptor

Several laser materials using energy transfer to aid pumping efficiency exist. Er, Tm, Ho in several hosts have been demonstrated, most notably LiYF₄ and YAG, the laser wavelength being in the 2 μm region. The 3 μm Dy laser in BaY₂F₈ and the 545nm LiTbF₄ laser are other examples for RE³⁺ lasers. More recently the Cr³⁺ sensitized Nd³⁺ laser has had a rapid development and is currently the most efficient lamp pumped solid state laser.

The total energy transfer rate in these materials is proportional to the product of the donor excited state population and the acceptor ground state population. At modest pumping rates the transfer rate is therefore proportional to the pump rate. In all the lasers mentioned however, the anticipated efficiency based on the pumping efficiency has not been realized. In most cases this can be related to either excited state absorption in the donor (Cr,Nd : GSGG) or upconversion as in the αβHo : YLF laser.

The discovery of upconversion can probably be credited the human eye being extremely sensitive to visible wavelengths. Thus the first reports on upconversion involved infrared to visible upconversion phosphors (5) such as (Yb,Er), green, (Yb,Ho), red, and (Yb,Tm), blue, even though these phosphors attained efficiencies only of the order of 10⁻⁴. Higher efficiency could of course be predicted if the pump intensity was greatly increased. Indeed the first reported upconversion laser involved flash lamp pumping (Yb,Er):BaY₂F₈ where green laser emission was obtained by infrared pumping (5).

UPCONVERSION.

As with energy transfer, upconversion takes place when an excited donor transfers its energy to an acceptor, Fig. 1. The only difference is that the acceptor ion is already excited to a metastable level. The total transfer rate now is therefore proportional to the product of the excited state population of both the donors and acceptors. Since both these populations in general are proportional to the pump rate, the upconversion is proportional to the square of the pump rate. In Fig. 1 let us assume that level 1 of the system is pumped at a rate W (s⁻¹cm⁻³). The steady state population is then, disregarding upconversion, given by :

$$\frac{dN_1}{dt} = 0 = W - \frac{N_1}{\tau_1}$$

$$\text{or } N_1 = W\tau_1$$

The upconversion rate can be expressed as:

$$W_{\text{up}} = \alpha N_1^2 = \alpha (W\tau_1)^2$$

We can immediately see that the upconversion rate in addition to be proportional to W^2 is also proportional to τ_1^2 . Thus a long fluorescent lifetime of level 1 is important. For a given oscillator strength the radiative lifetime has a λ^3 dependence. One therefore would expect the highest upconversion efficiencies where infrared levels are involved. Specifically the lowest energy manifolds of the trivalent Er, Ho, Tm and Yb are suitable candidates.

THE 2.8 μm ER : YLF LASER.

Fig.2 shows the energy levels of Er^{3+} in LiYF_4 . The 2.8 μm laser transition is between the $^4 I_{11/2}$ and the $^4 I_{13/2}$ manifolds. Conventional thinking led to the conclusion that "bottle necking" would prevent this laser from becoming practical. This is due to the very long fluorescent lifetime of the terminal laser level $\sim 15\text{ms}$. The population in this level would build up and terminate the laser action except at very low repetition rate pulsed pumping. When upconversion is included in the rate equations however, the effective fluorescent lifetime of the lower level shortens dramatically.

Let us assume that the lifetime of the $^4 I_{9/2}$ level is very short so that the reverse of the upconversion process does not take place. The decay rate of $^4 I_{13/2}$ is then, approximately given by:

$$\frac{dN_1}{dt} = \frac{N_1}{\tau_1} - \alpha N_1^2$$

For Er:YLF, $\tau_1 = 15 \cdot 10^{-3} \text{ s}$, $\tau_2 = 4 \cdot 10^{-3} \text{ s}$ and α is of the order of $10^{-15} \text{ cm}^{-3} \text{ s}^{-1}$. From this we find that the laser threshold is reached when $N_1 = 2 \cdot 10^{18} \text{ cm}^{-3}$. This corresponds to a stored energy of $\sim 0.2 \text{ J/cm}^3$. Pulsed threshold should therefore be relatively easy to achieve for this laser at room temperature.

Operation at reduced temperature improves the situation in two ways. First, since

the laser transition is from the bottom level of the $^4 I_{13/2}$ multiplet, the Boltzman distributions are more favorable at low temperature. Second, for the same reason, the reverse transfer process becomes even less likely and the effective upconversion parameter, α , will be larger.

A laser of the composition $\text{LiEr}_{0.3}\text{Y}_{0.7}\text{F}_4$ has been operated cw in this way. The 5mm by 50mm laser rod was cooled by flowing liquid N_2 and pumped by a 1000 W tungsten lamp. The threshold was at 250 W into the lamp and a few watts of output has so far been achieved.

FUTURE DEVELOPMENT.

It was mentioned earlier that upconversion was detrimental to the $\alpha\beta\text{Ho} : \text{YLF}$ laser. The upconversion process has been identified as the transfer from $\text{Er } ^4 I_{13/2}$ to Ho thereby raising the Ho from its $^5 I_7$ level to its $^5 I_5$ level, Fig.3 Since this is a near resonant process it can be very efficient. To make use of this process, the relative concentrations of Er and Ho have to be optimized for maximum upconversion. In the simplest picture, the $\text{Er } ^4 I_{13/2}$ will directly feed the $\text{Ho } ^5 I_7$ through energy transfer. This process should not be too efficient since the following upconversion rate is proportional to the product of the population of the $\text{Ho } ^5 I_7$ and the $\text{Er } ^4 I_{13/2}$. Possible laser transitions in this system are the $3.9 \mu\text{m } ^5 I_5$ to $^5 I_6$ transition and the $2.9 \mu\text{m } ^5 I_7$ transition.

Other possible upconversion lasers are systems with combinations of two or more of the RE ions Yb , Tm , Er and Ho . The parameters that will determine the practicality of these lasers are the temperature of operation, the availability of high intensity pump sources such as laser diodes and the availability of high quality crystals with optimum characteristics for the particular system.

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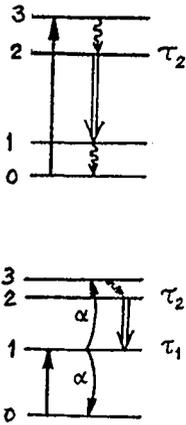


Fig. 1

Top: Normal relaxation

Bottom: Upconversion

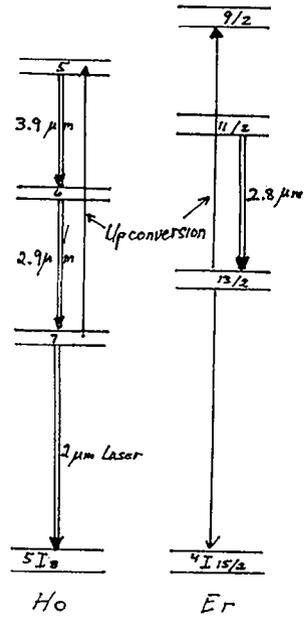


Fig. 3

Upconversion and possible laser transitions in the Ho, Er system.

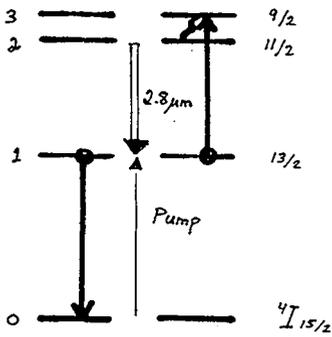


Fig. 2

The $2.8 \mu\text{m}$ Er upconversion laser.