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Up-conversion processes in laser crystals

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Abstract

Lasers which emit at higher frequencies than the pump light are usually called up-conversion lasers. The excitation of the active ion is due to multistep photon excitation, interionic energy transfer or photon avalanche. Er-, (Tm, Yb)-, and (Pr, Yb)-doped LiYF_4 laser crystals are successful examples where these processes are relevant.

Keywords: Up-conversion lasers; Energy transfer; Laser crystals; Rare-earth-doped materials

1. Introduction

Visible cw solid-state lasers are of great interest for applications like high-density data storage and laser displays. Their realization by using rare-earth-doped crystals is difficult due to the lack of suitable pump sources. A way to overcome this problem is the up-conversion of infrared pump photons by different energy transfer processes. The first up-conversion lasers required low temperatures, limiting their practical applicability. Today, up-conversion laser emission at room temperature and the availability of efficient infrared pump sources cause new interest in the development of such lasers.

2. Excited-state absorption pumping

The simplest mechanism is excited-state absorption (ESA) pumping as it occurs in Er:LiYF₄

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(Er:YLF) [1]. There are two different ways to populate the upper laser level $^4\text{S}_{3/2}$ by a two-step pumping process (Fig. 1) [1]. First, dual wavelength pumping is possible, using $\lambda = 970$ nm to excite the $^4\text{I}_{11/2}$ by GSA and $\lambda = 810$ nm (ESA) to populate the $^4\text{S}_{3/2}$. An easier way is the use of a single excitation wavelength. This excitation scheme requires an overlap between GSA and ESA, e.g. $\lambda = 970$ or 810 nm. At 810 nm the intermediate state is the $^4\text{I}_{9/2}$. Every pump scheme yielded lasing of Er:YLF on the transition $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$ at 551 nm. The maximum cw output power was 45 mW.

3. APTE and cooperative up-conversion

A second up-conversion mechanism is the APTE (addition de photons par transfer d'energie) as seen in Tm, Yb:YAG [2] and Tm, Yb:YLF [3]: Isolated Yb ions transfer successively their energy to various levels of a given Tm ion, yielding an excitation of the $^1\text{G}_4$ level (Fig. 2). $^1\text{G}_4$ is the upper laser level of three laser transitions in the spectral range

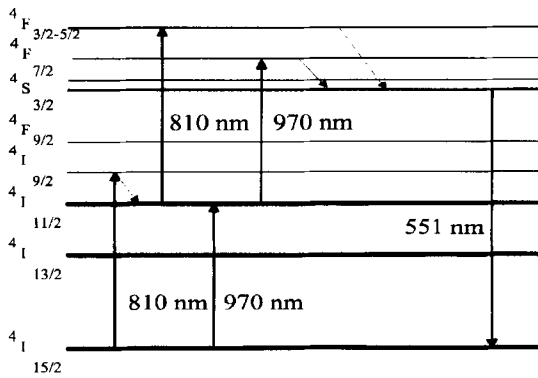


Fig. 1. ESA pump mechanism of Er:YLF [1].

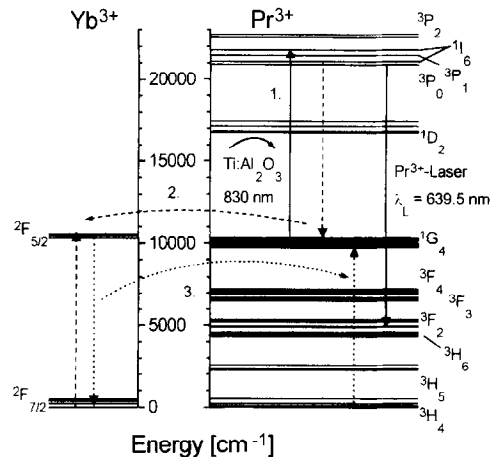


Fig. 3. Excitation scheme of Pr, Yb:YLF [4].

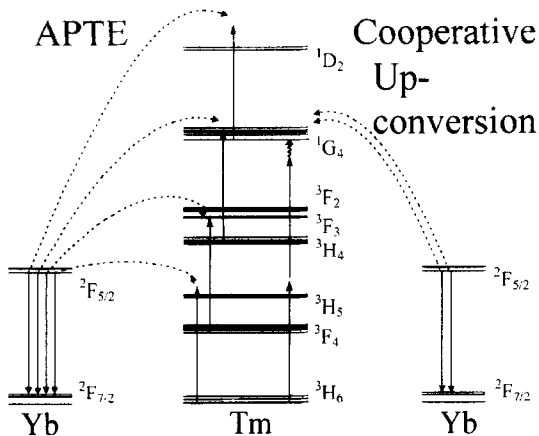


Fig. 2. Energy levels and transfer processes in Tm, Yb:YAG [2].

from 650 to 1568 nm. Furthermore, the APTE-process also prevents the laser transition $3H_4 \rightarrow 3F_4$ from being self-terminating as it transfers energy from the lower laser level to the $3F_2$ state. Besides the APTE, cooperative up-conversion is observed in Tm, Yb:YAG [2]. Energy is transferred simultaneously from two excited Yb ions to a single Tm ion (Fig. 2), yielding a population of the $1G_4$. Both processes can be observed at the same time in Tm, Yb:YAG. Their ratio depends on the concentration of dopants and on the excitation density.

4. Sensitized photon avalanche

Very recently, a sensitized photon avalanche mechanism between Pr³⁺ and Yb³⁺ in Pr, Yb-codoped fluoride crystals was observed (Fig. 3) [4]:

1. A Pr³⁺ ion in its $1G_4$ intermediate state is excited to $1I_6$ by absorption of a pump photon.
2. This ESA is followed by a cross relaxation, transferring energy from the Pr³⁺ ($1I_6 \rightarrow 1G_4$) to an Yb³⁺ ion ($2F_{7/2} \rightarrow 2F_{5/2}$).
3. The Yb³⁺ relaxes to its $2F_{7/2}$ ground state, exciting a second Pr³⁺ from its ground state to the $1G_4$ level.

This process yields a doubling of the population of the $1G_4$ state of Pr³⁺.

Three observations corroborate this hypothesis:

1. Neither a Pr:YLF nor an Yb:YLF exhibits any typical fluorescence under excitation at $\lambda \sim 830$ nm.
2. The population of $3P_0$ increases slowly, indicated by a rise time of the visible fluorescence of the order of some milliseconds.
3. A certain pump density is necessary to populate the upper laser level (threshold of fluorescence). Pumping with a Ti:sapphire laser at $\lambda \sim 830$ nm, laser emission was obtained on the transitions $3P_0 \rightarrow 3F_2$ ($\lambda = 639.5$ nm, $P_{\text{out}} = 75$ mW) and $3P_0 \rightarrow 3F_4$ ($\lambda = 720$ nm, $P_{\text{out}} = 19$ mW), respectively.

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