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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 LiYF₄ 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-earthdoped 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, upconversion 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 $Er: LiYF_4$

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(Er:YLF) [1]. There are two different ways to populate the upper laser level ${}^{4}S_{3/2}$ by a two-step pumping process (Fig. 1) [1]. First, dual wavelength pumping is possible, using $\lambda = 970$ nm to excite the ${}^{4}I_{11/2}$ by GSA and $\lambda = 810$ nm (ESA) to populate the ${}^{4}S_{3/2}$. 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 ${}^{4}I_{9/2}$. Every pump scheme yielded lasing of Er:YLF on the transition ${}^{4}S_{3/2} \rightarrow$ ${}^{4}I_{15/2}$ 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 ${}^{1}G_{4}$ level (Fig. 2). ${}^{1}G_{4}$ 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 ${}^{3}H_{4} \rightarrow {}^{3}F_{4}$ from being self-terminating as it transfers energy from the lower laser level to the ${}^{3}F_{2}$ 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 ${}^{1}G_{4}$. 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 Pr^{3+} and Yb^{3+} in Pr, Yb-codoped fluoride crystals was observed (Fig. 3) [4]:

1. A Pr^{3+} ion in its ${}^{1}G_{4}$ intermediate state is excited to ${}^{1}I_{6}$ by absorption of a pump photon.

2. This ESA is followed by a cross relaxation, transferring energy from the $Pr^{3+}({}^{1}I_{6} \rightarrow {}^{1}G_{4})$ to an Yb^{3+} ion $({}^{2}F_{7/2} \rightarrow {}^{2}F_{5/2})$.

Yb³⁺ ion (${}^{2}F_{7/2} \rightarrow {}^{2}F_{5/2}$). 3. The Yb³⁺ relaxes to its ${}^{2}F_{7/2}$ ground state, exciting a second Pr³⁺ from its ground state to the ${}^{1}G_{4}$ level.

This process yields a doubling of the population of the ${}^{1}G_{4}$ state of Pr^{3+} .

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 ${}^{3}P_{0}$ 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 ${}^{3}P_{0} \rightarrow {}^{3}F_{2}$ ($\lambda = 639.5$ nm, $P_{out} = 75$ mW) and ${}^{3}P_{0} \rightarrow {}^{3}F_{4}$ ($\lambda = 720$ nm, $P_{out} = 19$ mW), respectively.

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