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Homogeneous broadening effect on temperature dependence of green upconversion luminescence in erbium doped fibers

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ABSTRACT

We study the green upconversion luminescence of Er^{3+} ions in an aluminosilicate optical fiber upon near infrared excitation at 787 nm. The dependence of the upconversion luminescence on temperature has been determined. As temperature drops from room to cryogenic temperatures, the upconversion green emission reaches a maximum around 40 K, and then decreases. A nearly quadratic dependence of the upconversion luminescence with excitation power is found, which is consistent with a sequential stepwise two-photon absorption process. These results have been explained with a semiclassical model that considers the inhomogeneous broadening of the optical transitions due to glass imperfections, and the dependence of the homogeneous linewidth broadening on temperature.

1. Introduction

Upconversion luminescence is a physical process where a high-energy photon is obtained from a material when two or more incident low-energy photons impinge on it. Rare earth (RE) elements are the most important materials which exhibit this behavior due to their unique atomic configuration that results in sharp luminescence emission lines [1]. RE ions have an optically active 4f electron layer shielded by the filled 5s and 5p outer shells. This shielding limits the interaction between the ions and the host material. Among the trivalent RE ions, the erbium ion (Er^{3+}) has been extensively studied because of its ability to emit light within the near-ultraviolet to the near-infrared range upon excitation with infrared light in a number of hosts [2,3]. The absorption and emission peaks can be engineered using different dopant–host combinations [4]. The host material has been shown to strongly modify the luminescence efficiency. Hosts with high phonon energies lead to a reduction of the luminescence efficiency due to non-radiative relaxation [5].

The upconversion process has found a wide range of applications like obtaining green laser emission upon excitation of Er^{3+} by infrared radiation either in crystals [6,7] and fibers [8,9]. Optical waveguides doped with proper ions have been proposed as temperature sensors based on measurements of the intensity ratio of the emission peaks around 530 and 550 nm [10]. More recently, biological applications for Er^{3+} doped nanomaterials such as in-vivo imaging [11–13] and intracellular temperature nanosensors [14] have been proposed. Other emerging applications include three-dimensional volumetric displays [15], and silicon solar cells [16].

The luminescence in materials with several metastable electronic states usually involves a variety of mechanisms. Most of the upconversion two-step processes rely on the ion excitation by ground-state absorption (GSA), subsequent upconversion by excited state absorption (ESA) and/or energy transfer upconversion (ETU), and depletion by luminescence and by multiphonon relaxation [17]. ESA and ETU mechanisms can coexist even though one of them usually dominates over the other. ESA involves only one optically active ion that is promoted to upper levels by sequential resonant absorption of two or more laser photons, and dominates in materials at low-dopant concentrations [18]. ETU is an interionic process that relies on the energy exchange between neighboring ions. It depends strongly on the distance between the ions and predominates at high dopant concentrations.

The dependence of the integrated intensity of the upconversion luminescence signal on the laser excitation intensity follows a power law. The slope of this power law in double-logarithmic representation is indicative of the number of photons involved to excite the emitting state. This slope decreases with increasing excitation power. The influence of the upconversion mechanism and the laser power on this slope have been studied in detail by Pollnau et al. [17].

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The upconversion luminescence of Er^{+3} in many different hosts has been extensively characterized mostly at room temperature due to the interest of erbium doped materials from both a scientific and application point of view [19–25]. Only few works have focused in the behavior of the upconversion signal obtained in these materials at cryogenic temperatures [7,18,26,27]. Typically, as temperature drops from room to cryogenic temperatures, the luminescence intensity increases, reaches a maximum, and then decreases when a further drop of the temperature is achieved. At high temperatures a similar behavior was found by dos Santos et al., where an Er^{3+}/Yb^{3+}-codoped chalcogenide glass was heated from 23 °C to 155 °C [28]. This general trend has been explained on the basis of a competition between nonradiative decays and off-resonance excitation processes. As temperature drops, radiative emission is enhanced due to the decrease of the nonradiative decay rates [29], while the upconversion efficiency diminishes due to a reduction of phonon-assisted absorption [26,28]. Vermelho et al. [26] characterized the upconversion luminescence intensity in erbium-doped tellurite glasses excited at 1540 nm. They observed a maximum green upconversion intensity at 120 K which was explained by the interplay between nonradiative multiphonon relaxations and phonon-assisted absorption at the second step of the stepwise three-photon upconversion process. Golab et al. [27] reported a maximum upconversion emission around 150 K in an Er:YVO_4 crystal pumped with a 808 nm diode laser. They ascribed the reduction in green emission at lower temperatures to the decrease of the $^4I_{9/2}$ level absorption coefficient and the reduction in green emission at higher temperatures to the decrease of the green emission quantum efficiency. Van der Ziel et al. [18] obtained a maximum of the green emission around 90 K in an erbium-doped YF_3 crystal under excitation to level $^4I_{11/2}$. In this case, the reduction in the green emission at lower temperatures resulted from the depopulation of the higher energy stark levels of the $^4I_{11/2}$ manifold, which allows a resonant match for energy transfer upconversion. The effect of temperature on greenupconversion lasing in Er:YALO with excitation around 807 nm ($^4I_{9/2}$) was reported by Scheps [7]. The maximum laser output was achieved at 34 K. Line narrowing was mentioned as a possible mechanism for the laser output quenching below this temperature.

In RE doped glasses the broadening of the absorption and emission line profiles arises from both the thermally excited low-frequency vibrational modes (homogeneous broadening) and the structural disorder of the host material (inhomogeneous broadening) (see the review work of Macfarlane and Shelby [30]). At room temperature the inhomogeneous and homogeneous linewidths are in the same order of magnitude [31,32]. As temperature decreases, the homogeneous broadening decreases highlighting the inhomogeneous character of the material. As a general trend the dependence of the homogeneous linewidth with temperature exhibits a power-law behavior with an exponent ranging from 1 to 2 [33]. At low temperatures tunneling modes contribute to the homogeneous linewidth resulting in a quasilinear dependence with temperature [34,35]. At high temperatures two-phonon Raman processes dominate resulting in a quadratic dependence with temperature [36]. In the intermediate range of temperatures a cross-over is expected [37] and other processes may contribute to the homogeneous width, as for example phonon assisted direct transitions (which also lead to a roughly linear dependence with temperature) [33,38]. The temperature at which this cross-over occurs varies depending on the ion and host considered [37].

Our aim is to experimentally and theoretically analyze the green upconversion luminescence of Er^{3+} ions in an aluminosilicate optical fiber excited at 787 nm. We measure the spectral changes of the upconversion luminescence signal from room temperature to 11 K. To characterize the upconversion mechanism involved we measure the luminescence dependence on the laser excitation power. We develop a semiclassical model that considers the inhomogeneous broadening of the optical transitions due to glass imperfections and we study the influence of different thermal effects on upconversion luminescence.

2. Experimental setup

We experimentally analyze the upconversion green luminescence corresponding to transitions $^4S_{3/2} \rightarrow {}^4I_{15/2}$ and $^2H_{11/2} \rightarrow {}^4I_{15/2}$ of the erbium ions in a single mode Al_2SiO_5-glass-based erbium doped fiber (Liekii, Ltd.) at temperatures from 11 to 300 K (see Er^{3+} ions energy level diagram in Fig. 1). We use the experimental setup depicted in Fig. 2. The fiber has a nominal ion concentration of 6.3×10^{25} ions/m^3 (0.5 mol.%), a peak absorption of 80 dB/m at 1530 nm, a length of 12.7 cm, a nominal mode field diameter (MFD) at 1550 nm of 6.5 μm, a fiber cladding of 245 μm, and a numerical aperture of 0.2. This fiber was rolled around a home-made, 35 mm diameter copper cylinder. The cylinder was affixed to the cold finger of an Edwards cryostat (Coldhead CS2/9) provided with a compressor (Cryodrive 1.5). High vacuum grease was applied to the fiber to ensure a good thermal contact with the cold finger (see photo in Fig. 2). The cold finger temperature was kept constant using a controller from Oxford Instruments (ITC 502) with an accuracy of 0.1 K. The EDF was fusion spliced to standard single mode fibers: at the input of the EDF, a S630-HP (Thorlabs) fiber with operating wavelength 630–860 nm and MFD 4.2 μm at 630 nm, which transmits the excitation power to the EDF; and at the output of the EDF, a 460-HP (Thorlabs) fiber with operating wavelength 450–600 nm and MFD 3.5 μm at 515 nm, which collects the green luminescence. These standard single mode fibers exited the cryostat through a home-made feedthrough, provided with two holes of 125 μm diameter, and were spliced again to undoped connectorized pigtails outside the cryostat with the same characteristics (S630-HP and 460-HP).

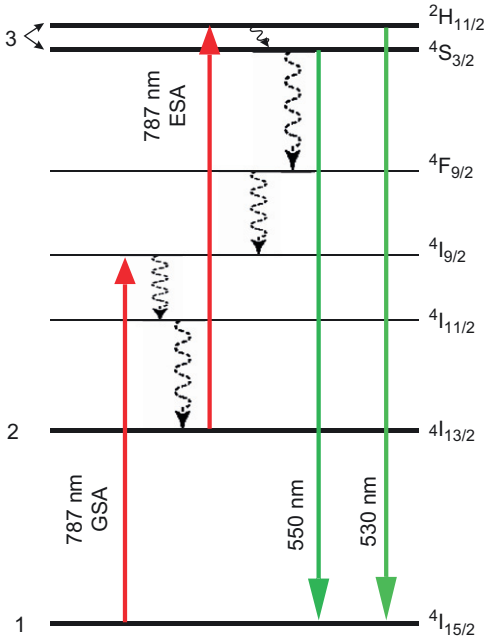

Fig. 1. Partial energy level diagram of Er^{3+} ions. The upward (red) arrows represent laser excitation at 787 nm. The downward solid (green) arrows represent green luminescence emission. Wavy lines represent fast nonradiative decays. GSA: Ground State Absorption; ESA: Excited State Absorption from $^4I_{13/2}$ level. (For interpretation of the references to color in this figure caption, the reader is referred to the web version of this paper.)