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Upconversion Luminescence Transients

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Abstract

Inhomogeneous active-ion distributions in laser materials lead to strong deviations of upconversion versus direct luminescence transients from the quadratic law of energy-transfer upconversion. Measured luminescence decay curves in LaSc₃(BO₃)₄:Nd³⁺ and GdVO₄:Nd³⁺ confirm experimentally the predicted deviations. Differences in energy migration within the metastable level of Nd³⁺ are identified.

For several decades, energy-transfer upconversion (ETU) in rare-earth-ion doped systems has been studied intensively, partly because of the fundamental interest in the physical nature of this process, but also because of the availability of near-infrared pump sources for the ETU excitation of visible luminescence and laser emission and because ETU can introduce a loss channel for devices emitting in the infrared region.

We investigate fundamentally the behavior of infrared luminescence emitted directly from a metastable level after excitation by a short laser pulse and visible luminescence emitted after ETU from this metastable level to higher-lying levels [1]. Although these two luminescences are connected by the same metastable level and influenced by the same ETU process, they probe different classes of ions. Whereas the infrared luminescence probes all ions, the visible luminescence probes only the class of ions susceptible to ETU [2]. A simple analytical model predicted that such luminescence decay curves exhibit a super-quadratic dependence of upconversion on direct luminescence decay [3].

The Nd³⁺ ion can serve as a model system for such investigations. It exhibits strong ETU from the metastable ⁴F₃/₂ level (Fig. 1). When doped into oxide matrices, the ⁴F₃/₂ level is the only metastable level within the 4f subshell. The Nd³⁺ energy levels excited by ETU decay by fast multiphonon relaxation and, hence, the weak visible fluorescence emitted from these levels represents a quasi instantaneous reaction on the dynamics of the ⁴F₃/₂ metastable level.

LaSc₃(BO₃)₄ and GdVO₄ crystals with different Nd³⁺ concentrations were provided by the University of Hamburg, Germany, and the General Physics Institute, Moscow, Russia, respectively. Experimental results obtained after short-pulse laser excitation near 800 nm (for an example, see Fig. 2) show that the upconversion (VIS) decay is much faster than the square of the direct (IR) decay, indicating the superquadratic behavior of ETU that fundamentally derives from the inhomogeneous active-ion distribution in the host. The experimental decay curves are described in a new model taking into account isolated, paired, and clustered ions. Comparison between LaSc₃(BO₃)₄ and GdVO₄ shows that static ETU in LaSc₃(BO₃)₄ [4] leads to a more
superquadratic behavior, in agreement with model predictions. These findings question many interpretations of upconversion luminescence in solid-state laser materials.

Fig. 1. Partial energy-level scheme of Nd$^{3+}$ indicating the relevant processes: pump excitation at 800 nm, direct (LUM$_1$) and upconversion (LUM$_2$) luminescence, ETU processes, and fast multiphonon relaxation (dashed arrows).

Fig. 2. Measured direct (IR) and upconversion (VIS) luminescence intensities in LaSc$_3$(BO$_3$)$_4$:10\%Nd$^{3+}$ together with model results (solid lines) for direct (IR) and upconversion (VIS) decay as well as the exponential asymptote and the square of direct (IR) decay [1].

References


