Energy-Transfer Processes Among Non-Homogeneously Distributed Rare-Earth Ions and Impact on Amplification and Lasing

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ABSTRACT

Energy-transfer processes such as energy-transfer upconversion are often detrimental to the performance of rare-earth-doped amplifiers and lasers on the typical luminescence transitions in the near-infrared spectral region between 1-2 μm. In order to quantify the influence of these interionic processes on amplification and lasing, not only luminescence decay curves have to be measured, but also the population dynamics of the electronic level scheme need to be modeled. The usually encountered non-homogeneous ion distributions complicate the situation. Here we present a stochastic model of energy-transfer processes that takes a statistical ion distribution into account. The influence of energy-transfer upconversion and cross-relaxation on amplification and lasing on the 1.06 μm transition in Nd^{3+}, the 1.53 μm transition in Er^{3+}, or the 1.84 μm transition in Tm^{3+} under these conditions is investigated.

Keywords: resonator, laser, spectral coherence, stimulated emission, spontaneous emission.

1. INTRODUCTION

Energy-transfer processes, such as the ($^4F_{3/2}$, $^4F_{3/2}$) → ($^4I_{15/2}$, $^4G_{5/2}$), ($^4F_{3/2}$, $^4F_{3/2}$) → ($^4I_{13/2}$, $^4G_{7/2}$), and ($^4F_{3/2}$, $^4F_{3/2}$) → ($^4I_{11/2}$, $^2G_{9/2}$) upconversion processes in Nd^{3+} [1,2,3], the ($^4I_{13/2}$, $^4I_{13/2}$) → ($^4I_{15/2}$, $^4I_{9/2}$) upconversion process in Er^{3+} [4,5,6], and the ($^3H_4$, $^3H_6$) → ($^3F_4$, $^3F_4$) cross-relaxation process in Tm^{3+} [7,8], have a strong impact on the performance of amplifiers and lasers on the 1.06 μm transition in Nd^{3+}, the 1.53 μm transition in Er^{3+}, and the 1.84 μm transition in Tm^{3+}, respectively. Consequently, a correct quantitative understanding of these processes is required for the optimization of device performance. Also quenching processes may affect only a fraction of the active ions [6,9,10].

The usual rate-equation models [11,12,13,14,15] that are applied to model energy-transfer processes treat all ions spectroscopically equally, i.e., they assume a homogeneous ion distribution. This is typically not true, and in the best case the ion distribution is statistical. A non-homogeneous ion distribution leads to a significantly different spectroscopic behavior.

We show that distinguishing different classes of ions [6,8,9,10], in the simplest case single ions that can decay only intrinsically and ions with neighbors that can participate in energy-transfer or quenching processes, provides a more realistic picture of the ongoing spectroscopy. We have applied this approach to spectroscopic data in amorphous Al_2O_3 doped with Er^{3+}, Yb^{3+}, and Tm^{3+}. The evaluation of spectroscopic data in Nd^{3+}-doped crystalline materials is currently in progress.

2. QUENCHED IONS

Rare-earth-doped Al_2O_3 thin layers were deposited by RF reactive co-sputtering onto thermally oxidized silicon wafers [16] and subsequently micro-structured to obtain channel waveguides [17]. These waveguides can be integrated with silicon-on-insulator technology [18]. Various rare-earth-activated amplifiers [5,3,19] and lasers [20,21,22,23,24] on silicon have been demonstrated.

By measuring pump transmission, luminescence decay, and optical gain in Al_2O_3:Er^{3+} channel waveguides, we observed that a significant fraction of Er^{3+} ions, which increases with dopant concentration, amounting to 32% at an Er^{3+} concentration of 3.66×10^{20} cm^{-3}, is quenched by a fast relaxation process [6,14]. As confirmed in a rate-equation approach which treats active and quenched ions separately [6], it was shown that this fast quenching process limits the achievable gain in Al_2O_3:Er^{3+} to 2 dB/cm [5], see Fig. 1. Also in Al_2O_3:Yb^{3+}, quenching of a fraction of ions was detected [15], but the fraction was significantly smaller, amounting to only 11% at an Yb^{3+} concentration of 6.67×10^{20} cm^{-3}. In both cases, the reason may be either energy transfer to (rare-earth or transition-metal) impurity ions or ion pairs that exhibit fast energy-transfer upconversion in the case of Er^{3+} or cooperative upconversion in the case of Yb^{3+}. Further spectroscopic investigations to quantify are in progress.

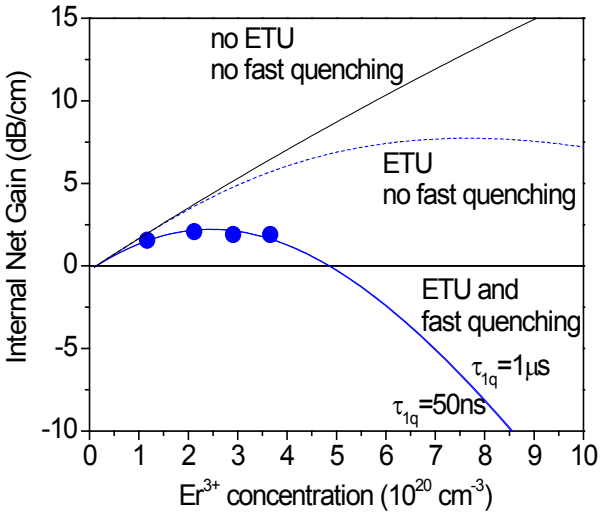

Figure 1. Internal net gain per unit length at 1533 nm versus Er^{3+} concentration, for a launched pump power of 100 mW at 976 nm and signal power of 1 μW at 1533 nm: measured data (dots) and calculations (lines) without quenching, only ETU quenching, as well as ETU and fast quenching. Two different values τ_{1q} = 50 ns and 1 μs of the fast quenching process were tested; the two resulting curves are almost identical. Figure taken from [6].

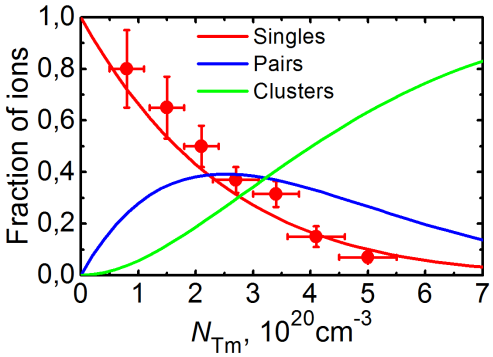

Figure 2. Fractions of spectroscopically distinct Tm^{3+} ions in amorphous Al_2O_3 versus Tm^{3+} concentration. Symbols*: experimental data for single ions determined from the luminescence decay.* Curves: *calculated data for single ions and ions with neighbors (pairs, clusters) using Eqs. (1) and (2). Figure taken from [8].*

3. STOCHASTIC MODEL FOR ENERGY-TRASFER PROCESSES

For the investigation of Al_2O_3:Tm^{3+}, we developed a stochastic model [8]. We consider two different ion classes, namely (i) single ions having a concentration N_{si} and (ii) ions with active neighbors, including ion pairs, multimers, and clusters, having a concentration N_{ni}. The sum of the two concentrations equals the dopant concentration N_d. All ions belonging to the same class of ions are considered spectroscopically identical. Single ions do not exhibit energy-transfer processes (CR and ETU), neither among their own class nor with ions from the other class, whereas ions with neighbors may participate in CR and ETU with other ions within their own class. Consequently, ions belonging to the different classes cannot communicate with each other. For each of these two ion classes a separate rate-equation system considering the spectroscopic processes relevant for this ion class is solved.

The probability for the occurrence of active neighbor ions in the first coordination sphere of an active ion is given by [8]

$$P_{m,n} = C_n^m p^m q^{n-m} = \frac{n!}{m!(n-m)!} p^m (1-p)^{n-m}. \quad (1)$$

Mathematically, n is the number of trials, m is the number of successful trials, and p is the probability of success. Physically, n corresponds to the coordination number of the first coordination sphere of nearest-neighbor rare-earth sites, m represents the tested number of active ions in the first coordination sphere, where m = 0 corresponds to a single ion, m = 1 to an ion pair, and m > 1 to a multimer or cluster, and $p = N_d/N_{max}$ represents the relative dopant concentration, where N_d is the absolute dopant concentration ($N_d = N_{Tm}$ in our case) and N_{max} is the maximum possible dopant concentration, i.e., for 100% replacement of the relevant host-matrix ions by rare-earth ions. Thus, statistically the fraction of single ions can be derived as [8]

$$x = \frac{P_{0,n}}{\sum_{m=0}^{n} P_{m,n}}. \quad (2)$$

This simple model is generally applicable to all rare-earth-doped materials and, although not taking all details into account, improves the accuracy of the spectroscopic description significantly. When applying it to the spectroscopic data of $Al_2O_3:Tm^{3+}$, we found good agreement for the fraction of single ions versus dopant concentration, see Fig. 2. The concentration-independent microscopic parameter of cross relaxation $(^3H_4, {}^3H_6) \rightarrow (^3F_4, {}^3F_4)$ was determined to be $C_{CR} = 5.83\pm0.26\times10^{-38}$ cm^6s^{-1}, such that the macroscopic parameter becomes $W_{CR} = C_{CR}N_{Tm}$.

Currently similar investigations on the energy-transfer-upconversion processes $(^4F_{3/2}, {}^4F_{3/2}) \rightarrow (^4I_{15/2}, {}^4G_{5/2})$, $(^4F_{3/2}, {}^4F_{3/2}) \rightarrow (^4I_{13/2}, {}^4G_{7/2})$, and $(^4F_{3/2}, {}^4F_{3/2}) \rightarrow (^4I_{11/2}, {}^2G_{9/2})$ in Nd^{3+} are in progress. Also in this case, the rate-equation analysis clearly indicates that the spectroscopic data cannot be described by a model that treats all ions equally.

4. CONCLUSIONS

Our investigation of energy-transfer processes among rare-earth ions indicates that models which treat all ions equally, hence assume a homogeneous ion distribution are unable to explain spectroscopic behaviour that is influenced by energy-transfer processes. A stochastic model has been proposed to take the influence of a statistical ion distribution into account.

ACKNOWLEDGEMENTS

I.C., P.L., and M.P. acknowledge financial support by the ERC Advanced Grant “Optical Ultra-Sensor” No. 341206 from the European Research Council.

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