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Energy flow in manganese-neodymium-doped fluoride glasses

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Abstract Measurement of the time-response of the excited-state concentration of manganese and neodymium has confirmed our previous hypothesis of fast energy transfer between donors and acceptors in fluoride glasses.

I. INTRODUCTION

The absorption and fluorescence properties of solid-state materials can often be enhanced by codoping with two ions: one ion (referred to as the donor) which absorbs the energy from the pump and transfers the energy to a second ion (referred to as the acceptor) which subsequently lases or fluoresces. The basic models for such non-radiative energy transfer have been developed extensively by Forster¹, Dexter², and Inokuti et al.³ In a recent paper,⁴ we developed a multiple-mechanism model, in which we proposed that within a certain donor-acceptor separation distance, the energy transfer occurs in a qualitatively faster manner than when the donor-acceptor separation distance is greater than this distance. This effect appears to be particularly prominent in glasses.⁵⁻⁷

In this paper, we will present data from manganese (donor) and neodymium (acceptor) doped fluoride glasses. We find an anomalously fast manganese-neodymium energy transfer, achieved by pumping at 409 nm and observing neodymium emission at 870 nm. We demonstrate how the relevant parameters of the transfer can be systematically derived from the experimental data.

II. EXPERIMENTAL PROCEDURE

We measured the temporal response of two fluoride glasses to optical pumping, Crystal A (36 PbF₂, 24 MnF₂, 35 GaF₃, 2 AlF₃, Y F₃, 3.8 LaF₃, 0.2 Nd F₃) and Crystal B (same as Crystal A but with 2 LaF₃ and 2 NdF₃). Thus, Crystal B is considerably more heavily doped than Crystal A. The measurements were conducted with an experimental setup with a 100 ns impulse response (determined by measuring the detector response to a sub-nanosecond dye-laser pulse train). The 409 nm excitation pulses (corresponding to a sharp Mn⁺²

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absorption band) were obtained from a PRA-LN102 dye laser pumped at 337 nm by PRA-LN103 nitrogen laser. The fluorescent output was filtered through a band pass filter centered at 600 nm (passing Mn^{+2} emission) or alternately a high pass filter at 780 nm (passing only the Nd^{+3} 870 nm emission). The fluorescence was measured by a PM55 Hamamatsu photo-multiplier and recorded on a Tektronix 2430 digital scope.

III. RESULTS AND DISCUSSION

The theory and parameters in our model have been previously described in Ref. 7. Figs. 1 - 4 show the donor excited concentration 605 nm decay obtained from Crystals A and B (pumping at 409 nm). (Two time scales are shown.) The decay shows a definite initial fast decay followed by a slower decay. The analysis described in Ref. 7 was performed and the results obtained are shown in Table 1. The fit obtained to the donor data is shown in these figures.

The parameters of Table 1 were then used to calculate the predicted acceptor decay. An excellent fit is shown in Figs. 5 and 6.

The actual results obtained for r_1 and the fact that it changes with crystal composition are very interesting. Future work should be directed towards analyzing phenomena present in glasses on the order of tens of nanometers which could contribute to the energy transfer.

IV. SUMMARY

We have confirmed that our previously hypothesized fast transfer between Mn^{+2} and Nd^{+3} is indeed fact. The inadequacy of the standard model for the initial decay in these systems has been shown. The efficacy of studying the donor and acceptor excited-state concentration for determining the coefficients of the ionic interactions has been validated.

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	<u>Crystal A</u>	<u>Crystal B</u>
R_1	5.8 nm	3.6 nm
R_{01}	10.4 nm	8.4 nm
R_{02}	8.6 nm	5.4 nm
τ_{0D}	580 μsec	580 μsec
τ_{0A}	630 msec	380 μsec

Table 1: Results of the analysis of the lightly and heavily doped crystals (A and B respectively).

Fig. 1: Experimental donor decay data and fit obtained for Crystal A, long time scale.

Fig. 2: Experimental donor decay data and fit obtained for Crystal A, short time scale.

Fig. 3: Experimental donor decay data and fit obtained for Crystal B, long time scale.

Fig. 4: Experimental donor decay data and fit obtained for Crystal B, short time scale.

Fig. 5: Experimental acceptor excited state concentration and fit, obtained for Crystal A.

Fig. 6: Experimental acceptor excited state concentration and fit, obtained for Crystal B.