

electron-phonon coupling, electron-localized-mode interactions, or possible breakdown of the effective-mass approximation because of the large dc electric fields. However, the explanation for the frequency dependence of the mass proposed here is not considered conclusive until a more detailed theoretical treatment of the cyclotron resonance in this system is available.

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*National Research Council-Naval Research Laboratory Resident Research Associate.

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Time-Resolved Fluorescence Line-Narrowing Studies in $\text{LaF}_3:\text{Pr}^{3+}$ †

R. Flach, D. S. Hamilton, P. M. Selzer, and W. M. Yen

Department of Physics, University of Wisconsin, Madison, Wisconsin 53706

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We report the direct observation of energy migration and spectral diffusion in the 3P_0 state of $\text{LaF}_3:\text{Pr}^{3+}$ using the technique of time-resolved fluorescence line narrowing on the resonant $^3P_0 \rightarrow ^3H_4$ transition. The temperature dependence of the migration process has been measured and various possible transfer mechanisms are discussed. In addition a variation in the excited-state lifetime as a function of absorption frequency and a concentration broadening at zero delay in the nonresonant transition $^3P_0 \rightarrow (^3H_6)_1$ (the lowest Stark level) are reported.

We wish to report the direct observation of energy migration and spectral diffusion within the optically excited 3P_0 state of $\text{LaF}_3:\text{Pr}^{3+}$ using the technique of time-resolved fluorescence line narrowing on the resonant $^3P_0 \rightarrow (^3H_4)_1$ transition. ($(^3H_4)_1$ is the lowest Stark level.

Over the past few years the technique of fluorescence line narrowing (FLN)¹ has been repeatedly applied to overcome inhomogeneous broadening in solids. Using a pulsed, tunable, dye laser and time-resolved sampling of the fluorescence signal, this method can be expanded to in-

clude the study of dynamic effects within an inhomogeneously broadened fluorescence line. Applied to rare-earth ions in an amorphous host, it has recently made possible the direct observation of energy migration,² i.e., the nonradiative energy transfer between neighboring ions whereby the population of the excited-state level remains unchanged. By using a dye laser of considerably higher resolution and a Fabry-Perot interferometer as an analyzer, we believe we are the first to have directly observed an analogous effect for ions in a crystalline host. Obser-

variations of this type give insight into transfer mechanisms responsible for energy migration.

The excitation source used for our experiment is a pressure-scanned, pulsed, tunable, dye laser with a bandwidth of ≈ 1.5 GHz, and has been described elsewhere in detail.³ The analyzing system consists of a pressure-scanned Fabry-Perot interferometer followed by a narrow-band-pass interference filter. A Fabry-Perot spacer of 0.7 mm was used for most measurements to give a large free spectral range and an adequate signal-to-noise ratio. The signal from the photomultiplier was fed into a boxcar integrator where it was sampled at a preset time delay after the excitation pulse. By repeatedly scanning the Fabry-Perot interferometer at various time delays, the time evolution of the fluorescence profile was then recorded. A $\frac{1}{4}$ - μ sec gate aperture was used for all our measurements. With the laser pumping into the 3P_0 level all the studies pertaining to the migration effect were done on the resonant transition and at a dopant concentration of 5 mol%.

Figure 1 shows typical results of time-resolved sampling taken at two different temperatures. At lower temperatures we see a single, simple fluorescence line whose shape and width do not noticeably change with increasing delay. Also present is a small uniform background. When the temperature is further lowered this background

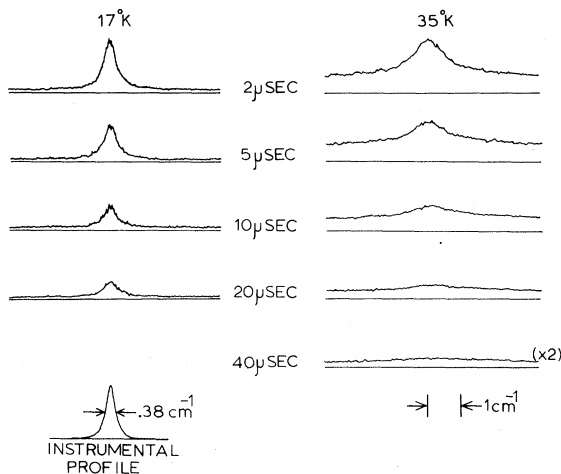


FIG. 1. Typical results of time-resolved studies taken at two different temperatures with a Fabry-Perot interferometer. [$^3P_0 \rightarrow (^3H_4)_1$ transition, 5-mol% Pr.] The virtually flat shape of the background is due to overlapping orders in the interferometer. The width of the narrowed fluorescence at a given temperature is equal to twice the homogeneous width plus the instrumental profile.

diminishes rapidly, and it appears to vanish at 5°K. At 35°K we see two obvious changes: First, the background becomes very significant, and persists to the very longest delays. Second, the narrowed fluorescence-line component now decays considerably faster. Both of these behaviors are characteristic of energy migration, provided the background originates from the inhomogeneously broadened, or "normal," fluorescence, i.e., the fluorescence which appears when the transition is pumped with a broad-band excitation source. To ascertain that this is the case, the time evolution of the signals was analyzed using a spectrometer (Spex Model No. 1401) instead of the Fabry-Perot interferometer (Fig. 2). At the cost of poorer resolution and signal-to-noise ratio, we eliminate the problem of overlapping Fabry-Perot orders which give rise to a seemingly flat background in Fig. 1. Figure 2 indicates that the background has the shape, width, and position of the full inhomogeneous fluorescence. Also, no significant changes are observed in its position, width, and time evolution when the laser is tuned from the high- to the low-energy side of the 3P_0 absorption curve. From these results it is clear that transfer of energy from the original excitation to ions that fluoresce at other positions in the inhomogeneously broadened fluorescence profile is occurring.

We proceed by addressing the nature of this transfer mechanism. We assume that strain broadening is microscopic, i.e., that it is random over a characteristic distance smaller than the average ion-ion separation. Assuming that

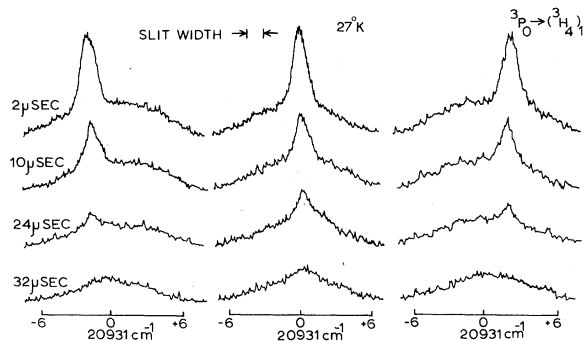


FIG. 2. Results of time-resolved studies taken with a spectrometer on the sample in Fig. 1. The laser is pumping at three different positions in the 3P_0 absorption band (low-energy side, center, and high-energy side). The narrowed fluorescence is in resonance with the laser frequency and shows the shape of the spectrometer profile.

the ion-ion interaction is electrostatic (exchange interaction is thought to be negligible for dilute rare-earth systems as in our case) the transfer rate $P_{1 \rightarrow 2}$ between two ions 1 and 2 can be written as^{4,5}

$$P_{1 \rightarrow 2} = K(\Delta E)R^{-s}. \quad (1)$$

$K(\Delta E)$ is a function of the energy mismatch $\Delta E = E_1 - E_2$, where E_1 and E_2 are the excitation energies of the two ions. Its qualitative behavior is determined by the particular transfer mechanism in question. R is the inter-ion spacing, and s is equal to 6, 8, or 10 depending on the multipolar nature of the interaction.

For resonant energy transfer between two similar ions $K(\Delta E)$ is proportional to a Lorentzian line-shape term, having a width twice the homogeneous width, $\Delta\bar{\nu}$, of the absorption line.^{2,4} It thus vanishes rapidly for energy differences ΔE much greater than a few homogeneous widths. This should result in a migration process starting from the initial excited-state population and gradually diffusing outwards towards the neighboring ions in the inhomogeneous profile. We would then expect to see a gradual increase in the actual width of the fluorescence line with increasing delay. Using Dexter's equation for resonance transfer⁴ we get an upper limit of $2 \times 10^{-2} \text{ cm}^{-1}/\mu\text{sec}$ for this increase in linewidth. From the appearance of the full inhomogeneous background at the earliest delays in all of our data, we conclude however that none of this type of spectral diffusion is taking place, and that instead $K(\Delta E)$ seems to be virtually independent of ΔE . We therefore proceed by considering other types of transfer mechanisms.

Another possibility is phonon-assisted energy transfer. Quite generally phonon-assisted energy transfer can be either direct or of higher order (Raman, etc.). The direct process, involving the emission or absorption of an acoustic phonon, has been treated by Orbach.⁵ For $kT \ll \Delta E$, this type of transfer should result in a strongly asymmetric background and a large difference between the transfer rates when the laser is pumping on the high- and low-energy side of the absorption profile. At higher temperatures, in the limit of $kT \gg \Delta E$, it can be shown^{2,5} that $K(\Delta E) \propto kT$. $K(\Delta E)$ is then virtually independent of ΔE . For temperatures above 30°K ($kT \approx 21 \text{ cm}^{-1}$, $\Delta\bar{E} \approx 4 \text{ cm}^{-1}$) this would then explain the simultaneous appearance of the entire inhomogeneous fluorescence at the earliest delays.

We have plotted the temperature dependence of

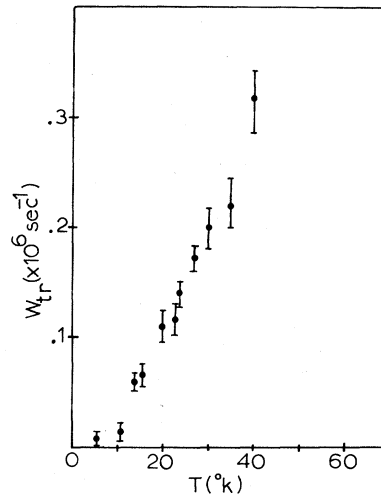


FIG. 3. Temperature dependence of the transfer rate W_{tr} . Above 10°K the dependence of the transfer rate is roughly linear (refer to text).

the transfer rate in Fig. 3, where W_{tr} is obtained by applying the formula of Inokuti and Hirayama⁶ to the height of the narrowed fluorescence in Fig. 1.⁷ Considering Eq. (1), it has been assumed that $K(\Delta E)$ is independent of ΔE as suggested by our data, and that $s = 6$ (dipole-dipole interaction).⁸ Although Fig. 3 qualitatively shows a linear dependence above 10°K, its slope does not intersect 0 as expected for the direct process.⁹ Higher-order phonon processes on the other hand would probably show a steeper temperature dependence (either exponential or approaching T^7 for a Raman process). Having ruled out resonant energy transfer as a viable mechanism, both the direct and higher-order phonon processes had to be invoked therefore to explain the observed temperature dependence. This conclusion seems to be indeed supported by recent measurements on higher-concentration samples¹⁰ which, while having a similar temperature dependence, also showed a pronounced asymmetry in the transfer rate at low temperatures.

Additional interesting effects that we have observed concern variations in linewidths and lifetimes. At 5°K and low concentrations (0.2 mol%) the nonresonant transition to the ($^3\text{H}_6$)₁ level shows an unexpectedly large residual width of 1 GHz. This width increases to $\approx 1 \text{ cm}^{-1}$ (= 30 GHz) at a concentration of 5 mol%, whereas we obtain an upper limit of only 200 MHz for the width of the resonant transition at this concentration. We attribute the large residual width of the nonreso-

nant transition to the fact that ions in qualitatively different crystal field environments can absorb at resonance but yet luminesce to slightly different nonresonant levels.¹¹ Such residual widths are not observed for Doppler-broadened lines in gases. This therefore constitutes a *major* difference in the behavior of inhomogeneously broadened lines between the two systems. Since we would expect that this residual width depends on the strength of the strain broadening, we attribute its increase with concentration to the corresponding increase in the inhomogeneous width (the latter increases from about 1 cm^{-1} at 0.2 mol% to over $\approx 7 \text{ cm}^{-1}$ at 5 mol%). Additional broadening could arise from resonance interactions in the ($^3\text{H}_6$)₁ level. At a concentration of 5 mol% we have also observed a variation in the excited-state lifetime as a function of absorption frequency. We attribute this to strongly concentration-dependent satellite lines that, at these higher concentrations, are now folded into the broadened main absorption line. Different crystalline field environments could also contribute to this effect.

These, and other related effects, will be discussed in a subsequent article.¹²

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⁹Also, as mentioned, for the direct process an asymmetric background should arise at lower temperatures. However below 7°K, where this asymmetry should appear, the background has already become vanishingly small. A search for variations in transfer rate with pump frequency is complicated by the fact that the excited-state lifetime varies with absorption frequency as discussed in the text.

¹⁰Selzer, Hamilton, Flach, and Yen, Ref. 8.

¹¹This interpretation is also in accordance with the fact that the corresponding line shapes are non-Lorentzian.

¹²R. Flach, D. S. Hamilton, P. M. Selzer, and W. M. Yen, to be published.

COMMENTS

Comments on $I = \frac{1}{2}$ Final States in Neutral-Current Reactions*

J. J. Sakurai

Department of Physics, University of California, Los Angeles, California 90024

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New tests of neutral-current models are proposed for $I = \frac{1}{2}$ final states produced in neutrino-induced reactions. The isoscalar analog of the conserved-vector-current hypothesis is also discussed.

One of the outstanding tasks in weak-interaction physics today is to determine the quantum numbers of the hadronic neutral current. Even

though neutral-current phenomena for semileptonic processes were first discovered in inclusive reactions,¹ it has been recognized for some