EXCITATION OF $LaF_3(Nd^{3+})$ *CRYSTALS BY MONOCHROMATIC LIGHT*

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Relaxation rates from the absorption band to a metastable level and from the lowest laser level to the ground state are determined for a four-level system excited by a monochromatic light pulse. The derived formulas enable simple calculations of W_{43} and W_{12} from experiment.

PROKHOROV and co-workers[1-4] have shown that laser materials and the operation of lasers can be successfully investigated with the aid of monochromatic pumping. The diverse operating schemes of the lasers used as pump sources permit a broad range of laser property measurements.

It had previously been reported[5,6] that oscillation was produced in YAlG, $CaWO_4$, and LaF_3 crystals doped with Nd^{3+} and excited by light from a ruby laser. In the present work monochromatic pumping has been used to determine the particle relaxation time from the 0.53-μ absorption band of Nd^{3+} ions to the $^4F_{3/2}$ level and to measure the cross section for induced emission in $LaF_3(Nd^{3+})$ crystals.

1. EXCITATION OF FLUORESCENCE

Laser action exhibits an important dependence on the relaxation rate from the absorption band to a metastable level. Dianov et al.[7] have attempted to determine this rate experimentally from the populating rate of a metastable level in silicate glasses containing Nd^{3+}; the pump light came from a spark in air at the focus of a neodymium laser. The duration of the exciting light pulse did not exceed $\sim 0.5\ \mu$sec. The characteristic populating time of the $^4F_{3/2}$ level was taken to be shorter than 0.5 μsec.

The calculation is considerably improved by using single-pulse laser radiation to excite fluorescence in Nd^{3+}-activated materials. We begin by considering the excitation of fluorescence in a four-level system by a square pump pulse. With a low pump level and only a small change in the ground-state population, i.e., $N_1 \approx N_0$, an analytic solution is easily obtained; Fig. 1 shows the results. In the case of infinitely rapid relaxation from the absorption band to the metastable level the intensity of fluorescence during the pump pulse increases linearly with time and reaches its maximum at the end of this pulse. In the case of a finite relaxation rate the corresponding rate of fluorescence increase depends on the pump power and on the probability W_{43} of transitions from the absorption band to the metastable level. After the termination of the pump pulse the fluorescence intensity continues to increase according to the law $A(1 - e^{-W_{43}t})$. Thus the rate of relaxation from the absorption band to the metastable level can be measured after the pump pulse has terminated. The shape of the exciting pulse is not important if the pulse base duration obeys the relation $\tau_p < 1/W_{43}$.

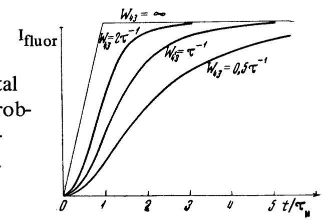

FIG. 1. Time dependence of crystal fluorescence intensity for different probabilities of the transition from the absorption band to the metastable level.

Crystals activated by Nd^{3+} ions have absorption bands in the regions of 0.53, 0.58, 0.74, 0.81, and 0.88 μ. We calculated the relaxation time $1/W_{43}$ in $LaF_3(Nd^{3+})$ crystals pumped by ruby laser light ($\lambda = 0.7\ \mu$) and by the second harmonic ($\lambda = 0.53\ \mu$) of a neodymium laser, corresponding, respectively, to transitions from the ground level to $^4F_{9/2}$ and $^4G_{7/2}$. In both cases the duration of the exciting pulse was 150 nsec. The 0.9-μ fluorescence was registered with an FÉU-28 photomultiplier having better than 5×10^{-8} sec time resolution. Figure 2 shows oscillograms of the leading edge of the fluorescence signal, on the basis of which we conclude that the relaxation time from the 0.53-μ absorption band to the metastable level is 1.2×10^{-7} sec and that the relaxation time from the 0.7-μ band is shorter than 10^{-7} sec. This difference is easily accounted for using the energy level scheme of Nd^{3+} ions in a $LaF_3(Nd^{3+})$ crystal.[8] It was shown in[7,9] that in Nd^{3+}-activated materials relaxation from an absorption band to a metastable

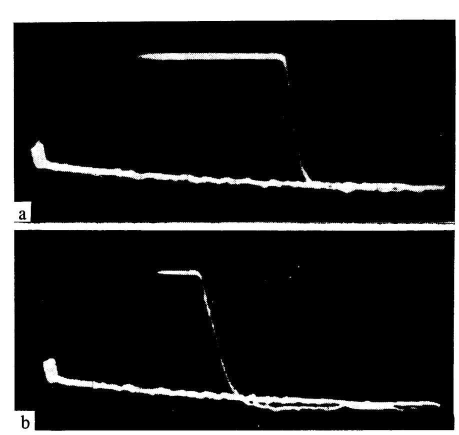

FIG. 2. Oscillograms of the leading edge of a fluorescence signal from a $LaF_3(Nd^{3+})$ crystal excited by: a—ruby laser radiation; b—the second harmonic of a neodymium laser (sweep scale 250 nsec/cm).

level proceeds in a cascade via intermediate levels. Therefore the difference between the relaxation times from the 0.53- and 0.7-μ bands is associated with a difference in the number of cascade steps and with a difference in the spacing of nearest levels. In the case of a cascade transition from $^4G_{7/2}$ to $^4F_{3/2}$ the maximum gap is ~1500 cm^{-1} for $^4G_{7/2} \rightarrow {}^2G_{7/2}$, thereby evidently determining the relaxation time from the 0.53-μ band. Similar relaxation time measurements from the same absorption bands to a metastable level were performed for KGSS-7 silicate glass and for an $YAlG(Nd^{3+})$ crystal; both results, 0.53 and 0.7 μ, were shorter than 10^{-7} sec.

2. EXCITATION OF LASER ACTION

It was noted in[10] that when $LaF_3(Nd^{3+})$ crystals are pumped by single-pulse lasers it is possible to determine the relaxation time $1/W_{21}$ of particles going from the lower laser level $^4I_{11/2}$ to the ground level $^4I_{9/2}$. In the present paper we present the results of a numerical calculation for the excitation of oscillation in a four-level system pumped by a 50-nsec square light pulse, allowing for the W_{21} transition probability. Figure 3a shows the time dependence, calculated with an electric computer, of laser emission for $W_{21} = 10^6$ sec^{-1}. The series of pulses following the first spike represents particle transitions from the lower laser level to the ground state. A computer calculation shows that after the first spike the populations of the upper and lower laser levels have been equalized. The next spike appears when the population difference $(N_3 - N_2)$ of these levels again reaches the threshold value as a result of transitions from the lower level to the ground level. The time interval between the first and second spikes is given by

$$\Delta t \approx \frac{1}{W_{21}} \ln \frac{K}{K-2}, \tag{1}$$

where $K = E_{pump}/E_{thresh}$. Equation (1) was obtained

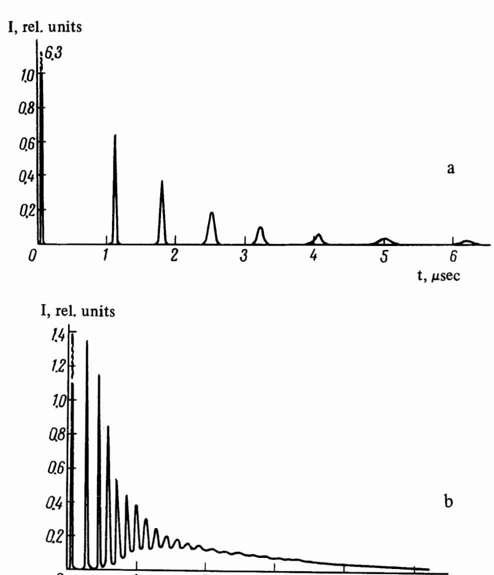

FIG. 3. Time dependence of laser radiation from a crystal excited by a square light pulse; $W_{21} = 10^{-6}$ sec. a—$K \approx 3$, b—$K \approx 12$.

using the approximation $\Delta t \ll 1/W_{32} = \tau$ (where τ is the particle lifetime on the upper laser level); this condition is fulfilled when $K > K_{min}$, where

$$K_{min} = 2\left[\left(\frac{W_{32}}{W_{21}}\right)^{W_{32}/W_{21}} - \frac{W_{32}}{W_{21}}\right]^{-1}. \tag{2}$$

K_{min} determines the minimum excess of pumping that is accompanied by the second radiation spike. The interval between the first and second spikes in this case, i.e., when $K = K_{min}$, is given by

$$\Delta t_{max} = \frac{1}{W_{21} - W_{32}} \ln \frac{W_{21}}{W_{32}}. \tag{3}$$

If the pumping considerably exceeds the threshold ($K > 10$) the spikes that follow the pump pulse are ultimately transformed into radiation that falls off exponentially (Fig. 3b). The characteristic time of the exponential decay is $\frac{1}{2}W_{21}$.

Equations (1) and (3) enable us to determine W_{21} from measurements of the time interval Δt between the first and second spikes.

As already mentioned, the foregoing calculation was performed for a four-level system. Considering the real level scheme of Nd^{3+} in LaF_3 crystals (with Stark splitting of the upper and lower laser levels), $E_{pump} \sim 7-8\ E_{thresh}$ is required to obtain the second spike. The threshold pump energy thus becomes very important. We therefore performed an experiment to determine the threshold population of the $^4F_{3/2}$ level in $LaF_3(Nd^{3+})$ crystals. The pump source was a freely oscillating ruby laser emitting a smooth pulse. The crystal was pumped from the end face as described in[6]. Since the pump pulse duration was only one-fifth of the particle lifetime on the $^4F_{3/2}$ level, the threshold population of this level was determined from the ruby laser energy absorbed in the $LaF_3(Nd^{3+})$ crystal. The $^4F_{3/2}$ threshold population in the experimental crystal was 6×10^{17} cm^{-3}. As we know, the population-difference threshold depends on losses in the crystal and on the induced-ratiation cross section σ. We determined σ by measuring the losses in the crystal using the method described in[11]. The cross section for induced emission in a $LaF_3(Nd^{3+})$ crystal ($\measuredangle$ CF 90°) was found to be 4×10^{-20} cm^2 ($\lambda_{laser} = 1.063\ \mu$).

The foregoing results indicate that measurements of the probability W_{21} of a transition from the lower laser level to the ground state, by means of the described procedure, require a radiation source in the 0.58-, 0.74-, or 0.81-μ region with a giant pulse yield of a few joules. Since high pump power usually damages a crystal it is evidently advisable to utilize a method of determining W_{21} that is similar to the aforedescribed one but avoids the production of high pump power. The essential modification in this technique, which is described in[10], consists in the fact that the resonator Q is switched on after the pump pulse has terminated. The trailing edge of the pump pulse should then not endure longer than the lifetime of the metastable level.

We must here mention work of Bondarenko et al.,[12] who reported an experiment with a ruby laser in which the Q was switched on at the end of a pump pulse, where the pump level was below the threshold, i.e., where free oscillation was interrupted. The emission output of this laser consisted of a series of spikes where the radiation frequency was downshifted continually in successive spikes. A similar pattern was observed when a ruby laser was pumped by a short pulse. As we have already shown, spiky oscillation can be associated with relatively slow relaxation of particles between ground-state doublet levels of Cr^{3+}, having the characteristic time 10^{-6} sec at 300°K,[13] while the frequency shifts result from the width of the upper laser level.

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