EFFECT OF STIMULATION OF NONRADIATIVE TRANSITION BY INTENSE OPTICAL EXCITATION

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In the case of Stokes excitation of a crystal, the radiation is preceded as a rule by nonradiative relaxation from the excited to the radiative level. The nonradiative relaxation probability depends on the temperature and is usually considered to be a characteristic of the impurity center and independent of the intensity of the excited light.

Our investigation of the probability of nonradiative relaxation in CaF_2 (Sm^{2+}) crystals has revealed the effect of stimulation of nonradiative transitions in the intense field of the exciting-light wave.

We observed the luminescence buildup kinetics following excitation by subnanosecond pulses of the second harmonic of a mode-locked neodymium laser. The exciting pulses constituted a series of spikes of duration <1 - 1.5 nsec (the limit of the time resolution of the apparatus) and average energy 0.1 - 0.2 J each.

The level and transition scheme of $CaF_2(Sm^{2+})$ is shown in Fig. 1. The excitation and emission of the crystal are identified with allowed transitions (1 → 3 and 2 → 1 on Fig. 1) between the states of the 4f and 4f-5d configurations in the Sm^{2+} ion [1 - 6]. We denote the probability of the nonradiative transition 3 → 2 by W_{32}. There are no other nonradiative transitions in this system, since the luminescence quantum yield from level 2, when excited in the $(14 - 30) \times 10^3$ cm^{-1} region, is equal to unity and the lifetime τ_2 of the radiative level at $T \leq 77°K$ is constant at $\sim 2 \times 10^{-6}$ sec [1, 2]. When level 3 is excited by a pulse having the form F(t) and under the conditions $\tau_2 \gg t_p$ (pulse duration) and $\tau_2 \gg 1/W_{32}$, the growth kinetics of the luminescence from level 2 is given by

$$I_g(t) = \text{const} \int_0^t \exp(-W_{32}t)\left[\int_0^t F(t)\exp(W_{32}t)\,dt\right]dt. \quad (1)$$

We can thus estimate W_{32} from $I_g(t)$.

Fig. 2 shows oscillograms of the luminescence signal of a $CaF_2(Sm^{2+})$ crystal, obtained at 77 and 300°K and at different excitation densities P/S from ~400 to 360 MW/cm^2.[1)]

The lower limit was determined by the amplitude sensitivity of the receiving section of the apparatus, namely the 18ELU-F9 photomultiplier[2)] and the 6LOR-02M oscilloscope. As seen from Figs. 2b and 2s, at $P/S \lesssim 1$ kW/cm^2 and $T = 77°K$ the luminescence intensity increases over a time much

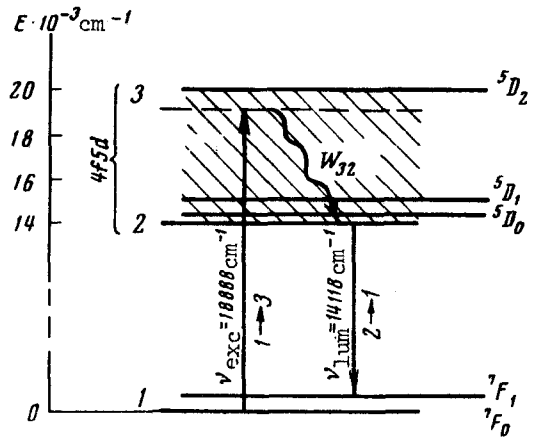

Fig. 1. Level scheme of CaF_2 (Sm^{2+}) crystal.

[1)] To vary the excitation density and maintain the total amplitude of the registered signal constant, we used different combinations of neutral filters to attenuate either the exciting light or the luminescence of the sample by an equal factor.

[2)] The authors take the opportunity to thank L.I. Andreeva, Z.M. Semichastonva, and B.M. Stepanov for supplying the photomultiplier.

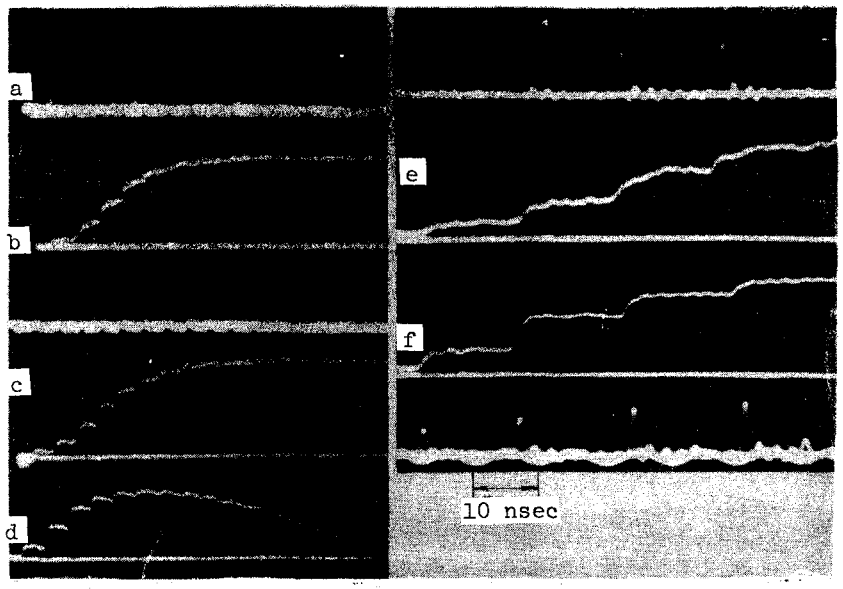

Fig. 2. Oscillograms of exciting pulses (a) and of the kinetics of luminescence growth at different excitation power densities: b) P/S ∿ 400 W/cm^2, S = 40 mm^2, T = 77°K; c) P/S ∿ 360 MW/cm^2, S = 1 mm^2, T = 77°K; d) P/S ∿ 4.5 kW/cm^2, S = 13 mm^2, T = 300°K; e) P/S ∿ 1 kW/cm^2, S = 1 mm^2, T = 77°K; f) P/S ∿ 2 kW/cm^2, S = 33 mm^2, T = 77°K. The distance between exciting spikes corresponds to 16.5 nsec.

longer than the duration of one spike, i.e., the case $t_p \ll 1/W_{32}$ is realized, and the time of nonradiative relaxation estimated from (1) is $1/W_{32} \gtrsim 10$ nsec. When the excitation power density is increased, this time becomes shorter, ∿3 nsec at P/S ∿ 2 kW/cm^2 (Fig. 2f), and at P/S > 100 kW/cm^2 it becomes comparable with the duration of the exciting spike, i.e., $1/W_{32} < 1.5$ nsec. Thus, when the exciting-light power density changes from 400 W/cm^2 to several dozen kW/cm^2 and more, the nonradiative transition probability W_{32} increases from ∿10^8 to ≳5 × 10^{10} sec^{-1}.

The observed effect can be connected with two factors: 1) the temperature dependence of W_{32}, if an increase of the excitation density raises the local temperature of the impurity center (the heating can usually be disregarded, for at P/S ∿ 300 kW/cm^2 and total light absorption the sample temperature rise would be only $\Delta t \sim 2 \times 10^{-3}$ deg); 2) interaction between the impurity center and the field of the exciting or radiated wave, leading to a change in the probability of the nonradiative transitions. The local temperature of the impurity center can be monitored by means of the change of τ_2. As follows from the form of $\tau_2(T)$ [2] and Fig. 2d, the value of τ_2 at 300°K decreases to a value $\lesssim 4 \times 10^{-8}$ sec. The absence of noticeable luminescence damping at 77°K within a time 10^{-7} sec (Figs. 2b and 2c) allows us to conclude that τ_2 should exceed 10^{-6} sec, and consequently the local temperature of the center cannot exceed 100°K [2]. The interaction of the impurity center with the field of the light wave can become manifest in an increase of W_{32} as a result of a shift of the excited states of the impurity center. This effect is connected with intra-center transitions and depends only on the intensity of the exciting light. In this case, as well as in the case of the temperature effects, the changes of $I_g(t)$ in formula (1) are connected only with changes of the parameter W_{32}.

Experiment has shown, however, that at excitation power densities $P/S \lesssim 10$ kW/cm^2 two processes are observed in the luminescence growth kinetics $I_g(t)$. One is fast and corresponds to the intensity jumps on the oscillograms, which occur after a time $\lesssim 1.5$ nsec. The other is slower, with duration $\gtrsim 3$ nsec. The ratio of the first process to the second decreases (Figs. 2b and 2f) with increasing number of the exciting spike (and with increase in its intensity).

The observed effects can be explained by assuming that intense excitation makes possible stimulated emission of non-equilibrium ("hot") phonons. As shown in [7], for stimulated nonradiative transitions to occur it is necessary to have an inverted population and a coupling between the particles via the photon or phonon field. For the $3 \to 2$ transition the level population inversion condition is satisfied only for the first spikes. It is for these that the maximum amplitudes of the fast jumps are observed on the oscillograms of Fig. 2. Estimates show that at densities $P/S \sim (1 - 10)$ kW/cm^2 $= (2.6 - 26) \times 10^{12}$ quanta/cm^2 (at $\lambda = 530$ nm), at which the effect of stimulation of nonradiative transitions is already observed, the distance between excited centers is $\sim(0.8 - 0.4) \times 10^{-4}$ cm and is comparable with the wavelength of the exciting light. To effect a coupling between centers via the phonon field at such distances between centers, it is necessary to have a phonon lifetime $\tau_{ph} > 10^{-10}$ sec, which seems little likely to us for CaF_2.

It follows from the aggregate of presented facts and estimates that stimulated nonradiative transitions are observed in $CaF_2(Sm^{2+})$ and the coupling between centers can be realized via the field of the exciting light wave.

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ENERGY SPECTRA OF NEUTRAL COMPONENTS IN SCATTERING BY A SOLID TARGET

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The radiation emitted by an ion-bombarded solid target includes a group of fast particles, ions as well as neutral atoms, the energies of which are comparable with the energy of the incident ions. These particles are partners of one or several successive pair collisions between the ion and the target atom [1]. The angular and energy distributions of these particles carry considerable information both on the microscopic properties of the bombarded sample (its crystal structure [2], the thermal lattice vibration [3], the composition of the surface [4]) and on the potentials of the interaction between the two colliding atomic systems [1].

In many cases, for example when metals are bombarded with inert-gas ions of energy $10^3 - 10^4$ eV, most fast scattered particles are neutral, and the ions constitute only several per cent [5]. In spite of this, almost all the published data pertain to the ionic component of the scattering. There have been