LUMINESCENCE AND GENERATION IN $CaF_2:Dy^{2+}$ *CRYSTALS EXCITED WITH A RUBY LASER*

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The results are presented of an investigation of the luminescence and generation in $CaF_2:Dy^{2+}$ crystals excited by a ruby laser. Giant generation pulses of ~ 30 nsec duration were obtained at the wavelength of $\lambda = 2.36\ \mu$.

FLUORITE crystals, activated with divalent dysprosium, have a strong absorption band in the 0.72 μ region and a ruby laser can therefore be used to excite luminescence and stimulated emission in these crystals. The use of a powerful monochromatic source opens up new avenues of investigation of the mechanism of light generation in crystals.

The purpose of the present work was: to determine the temperature dependence of the lifetime of Dy^{2+} ions at the luminescent level; to investigate the possibility of high-frequency modulation of the stimulated emission from $CaF_2:Dy^{2+}$ by the exciting energy; to obtain giant nanosecond generation pulses from $CaF_2:Dy^{2+}$; and to investigate the temperature dependence of the generation threshold of $CaF_2:Dy^{2+}$.

1. INVESTIGATION OF LUMINESCENCE

To determine the temperature dependence of the lifetime of Dy^{2+} ions at the luminescent $^5I_7$ level, we used an evacuated glass tube with double walls, into which the $CaF_2:Dy^{2+}$ sample was placed. Liquid nitrogen vapor was passed along the tube. The crystal temperature was measured with a thermocouple. The luminescence was excited using a Q-switched ruby laser (RL). Figure 1 shows the temperature dependence of the lifetime τ of Dy^{2+} ions at the luminescent level $^5I_7$ (in the range 77—260°K). The curve in Fig. 1 represents samples with an initial Dy^{3+} concentration of about 0.03%, which was reduced to Dy^{2+} by additive coloration or by γ-ray irradiation (10^7 r).

Kiss[1] estimated the radiative lifetime at the $^5I_7$ level as 0.4 sec. According to our estimates, the experimentally determined lifetime is shorter than the radiative lifetime and, therefore, the former is mainly due to nonradiative transitions (NT). The theory of these processes in ionic crystals is given in Malkin's papers.[2] The mechanism of energy transfer from an excited ion to the lattice involves the modulation of the Stark splitting of the ion level in the lattice field. Since the Debye frequency of fluorite ($\Theta = 470$°K) is much less than the optical quantum frequency ($^5I_7 \rightarrow {}^5I_8$), the NT's are accompanied by the simultaneous emission of a number of phonons. The temperature dependence of the NT's appears at temperatures T comparable with the Debye temperature Θ but it is absent at $T \ll \Theta$. This is confirmed by the experimental data.

We investigated crystals with a total concentration of Dy^{2+} and Dy^{3+} ions amounting to 0.03%. When the ratio of the concentrations of the two components was altered, the lifetime at the luminescent level changed within the limits 14—19 msec. Consequently, the NT's of Dy^{2+} could have occurred without the participation of Dy^{3+}. Therefore, it was interesting to calculate the probability of the NT $^5I_7 \rightarrow {}^5I_8$ for Dy^{2+}.

2. GENERATION IN $CaF_2:Dy^{2+}$

$CaF_2:Dy^{2+}$ crystals ($l = 20$ mm, D = 5 mm), with silver or dielectric coatings, were used to obtain generation (at $\lambda = 2.36\ \mu$) by excitation with a ruby laser. The latter was used either under the usual pulse conditions or under the giant pulse conditions. A $CaF_2:Dy^{2+}$ crystal was kept at T = 77°K and was excited through its lateral surface ("transmission"). When the laser was used under the usual pulse conditions, the threshold excitation energy of $CaF_2:Dy^{2+}$ was 0.1 J (the exit mirror had a reflection coefficient of 95%) and there was a delay before generation. The delay time was due to the need to accumulate energy before generation. The

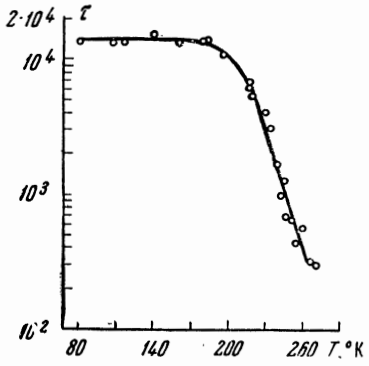

FIG. 1. Temperature dependence of the lifetime (in μsec) of Dy^{2+} ions at the $^5I_7$ level.

generation in $CaF_2:Dy^{2+}$ repeated the spikes of the ruby laser generation, and the spikes appeared simultaneously within 10^{-6} sec. This indicated the possibility of modulation, at a frequency of not less than 1 Mc, by means of optical pumping (for example, using semiconductor light sources). It was interesting to note that if the ruby laser excitation had no spikes,[3] the $CaF_2:Dy^{2+}$ generation had likewise no spikes (at a resolution of $\sim 10^{-7}$ sec).

3. DEPENDENCE OF THE GENERATION THRESHOLD ON TEMPERATURE

The temperature dependence of the generation threshold of $CaF_2:Dy^{2+}$ crystals is shown in Fig. 2. It is evident from this figure that when the temperature is increased (from nitrogen temperature) by 40 deg, the generation threshold rises by one order of magnitude and continues to rise, although the lifetime of Dy^{2+} ions at the $^5I_7$ level is constant in this range of temperatures (cf. Fig. 1). The increase in the threshold may be explained by a strong broadening of the luminescence line due to a shortening of the lifetime at the lower laser level as a result of an increase in the probability of the

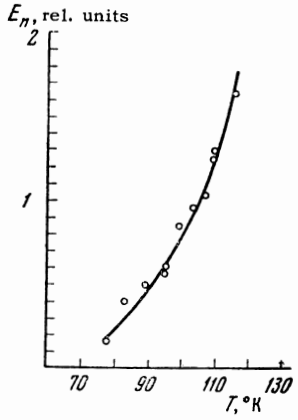

FIG. 2. Temperature dependence of the generation threshold of $CaF_2:Dy^{2+}$.

NT's when the temperature rises. In fact, the curve in Fig. 2 can be regarded as the temperature dependence of the width of the lower laser level. For this reason, the generation in $CaF_2:Dy^{2+}$ at room temperature meets with considerable difficulties, even if a ruby light source is used.

4. GENERATION OF GIANT PULSES AT THE WAVELENGTH OF 2.36 μ

A. Experimental part. When $CaF_2:Dy^{2+}$ was excited with a ruby laser working in the giant pulse mode (0.5 J energy, 30 nsec duration) no generation was obtained, although the crystal had a threshold generation energy of 0.1 J when excited with a ruby laser operating in the normal mode. However, if a ruby laser produced several giant pulses of ~ 30 nsec duration, which followed one another at 100—200 μsec intervals, generation was observed on the application of the second and later pulses. The general nature of the pulse sequence of the two lasers is shown in Fig. 3,[1)] which indicates that the second and third pulses of ruby excited giant pulses in $CaF_2:Dy^{2+}$. A giant pulse from $CaF_2:Dy^{2+}$ was followed by several small spikes, whose intensity was two orders of magnitude smaller. (In the case represented by Fig. 3, these small spikes were absent after the second giant pulse from $CaF_2:Dy^{2+}$.) As a rule, the number of these spikes lay within the limits 2—7 (sometimes they were absent) and the last spike followed the giant pulse after an interval of 100—200 μsec.

FIG. 3. Giant pulses of a $CaF_2:Dy^{2+}$ laser excited by a Q-switched ruby laser. The scanning rate is 100 μsec/division (the ruby signal is the upper one, the $CaF_2:Dy^{2+}$ signal the lower; the polarities are different).

[1)] For the best observation of the generation with an oscilloscope, the outputs of the infrared and red receivers were shunted with capacitors in order to increase the time constant.

The duration of the giant generation pulses of $CaF_2:Dy^{2+}$ was 30—40 nsec, the first pulse followed 100—200 nsec after the application of the exciting RL pulse, and the second fluorite pulse was delayed by 30—40 nsec.

B. Discussion of results. 1. The absence of generation in $CaF_2:Dy^{2+}$ when excited with a single RL pulse may be explained as follows. It is probable that the lifetime of Dy^{2+} ions at the levels in the absorption band (λ = 6943 Å) is longer than 30 nsec and these levels become saturated in a time interval less than 30 nsec,[2)] the maximum number of ions reaching the absorption band levels being insufficient to establish a negative temperature state and to compensate for the resonator losses. In order to obtain generation in $CaF_2:Dy^{2+}$ when excited with a single pulse of several tens of nanoseconds duration, it is necessary to establish first a near-threshold population of the lower component of the term $^5I_7$. This situation is obtained when $CaF_2:Dy^{2+}$ is excited with several giant pulses, of which the first establishes a near-threshold population in the lower component of the $^5I_7$ term and the remaining give rise to generation.

2. The appearance of small spikes can be explained by the existence of a metastable level $^5I_6$,[1] for which the lifetime is ~100—150 μsec. However, if all the excited Dy^{2+} ions do not bypass the $^5I_6$ level on relaxing, the following effect should be observed. When $CaF_2:Dy^{2+}$ is excited, with a 30—40 nsec pulse, the luminescence should rise exponentially with a time constant 100—150 μsec. We carried out this experiment, using an infrared radiation receiver with a time constant of ~15 μsec. The rise time of the luminescence front was ~15 μsec. Hence, we could conclude that not all the excited Dy^{2+} ions relax through the $^5I_6$ level.

3. The experimentally determined delay times of the giant pulses of $CaF_2:Dy^{2+}$, relative to the exciting pulses of the ruby laser, were in order-of-magnitude agreement with the calculations. The expression obtained by one of the present authors[4] for the generation delay time resulting from the instantaneous Q switching has the form

$$t_d \approx (1-2)\ln(\rho_{max}/2\rho_0)/v(\alpha_b-\alpha_0),$$

where ρ_0 and ρ_{max} are the initial and maximum energy densities for this type of oscillation, α_b is the negative absorption coefficient at t = 0, α_0 is the loss coefficient, and v is the velocity of light in the crystal. In our case, the beginning of the second and subsequent excitation pulses corresponds to the moment of Q switching, and their duration represents the Q-factor rise-time. If the concentration of Dy^{2+} ions is $N_0 \approx 6\times10^{17}$ cm^{-3}, $\alpha_0 = 0.05$ and the maximum value of α_b is 0.5 cm^{-1}, then the minimum delay time is

$$t_d \approx 5 \text{ nsec}.$$

This value is less than the delay time of the second giant pulse of Dy^{2+}. The discrepancy can be explained by the finite Q-factor rise-time (excitation pulse duration 30 nsec). The reason for the delay of the first Dy^{2+} pulse by 100—200 nsec is the small value of the difference $\alpha_b - \alpha_0$.

The duration of the Dy^{2+} generation pulse after the instantaneous Q switching, calculated using formulas given in[4], is ~2—5 nsec.

In conclusion, the authors express their gratitude to V. V. Osiko for the supply of $CaF_2:Dy^{2+}$ crystals.

[1] Z. J. Kiss, Phys. Rev. **137**, A1749 (1965).

[2] B. Z. Malkin, FTT **4**, 2214 (1962), Soviet Phys. Solid State **4**, 1620 (1963); FTT **5**, 1062 (1963), Soviet Phys. Solid State **5**, 773 (1963).

[3] A. K. Sokolov and T. N. Zubarev, FTT **6**, 2590 (1964), Soviet Phys. Solid State **6**, 2065 (1965).

[4] A. M. Prokhorov, Radiotekhn. i élektron. **8**, 1073 (1963).

[2)] Ejection of ions from this band to higher levels is also possible.

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