

THE EFFECT OF THE ANOMALOUS DISPERSION ON THE STIMULATED EMISSION SPECTRUM OF CRYSTALS

N. M. GALAKTIONOVA, V. F. EGOROVA, and A. A. MAK

State Optical Institute

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An effect of anomalous dispersion on the stimulated emission spectrum of $\text{CaF}_2:\text{Dy}^{2+}$ and $\text{CaF}_2:\text{U}^{3+}$ crystals is observed, leading to a decrease in the separation between laser modes and making the separation unequal. Under certain conditions the measured wavelength difference for two neighboring laser modes is $0.4 \lambda^2/2Ln_0$. It is found that the luminescence line in the $\text{CaF}_2:\text{Dy}^{2+}$ crystal is inhomogeneously broadened.

POPULATION inversion in an active medium gives rise to negative anomalous dispersion; recently this effect has been observed directly in ruby^[1]. The effect of negative dispersion on the laser spectrum was studied by Bennett^[2]. It was shown that negative dispersion leads to a drawing together of the frequencies of the laser modes (frequency pulling). The relative magnitude of this effect is of the order of 0.1%. Observation of this effect in crystals is made difficult by the relatively large fluorescence line widths which occur; an attempt to observe frequency pulling in a ruby crystal was made by McMurtry^[3].

It is well known that for a Lorentz line shape the magnitude of the mode pulling (which is determined by the ratio of the wavelength difference between axial modes $\Delta\lambda$ to the quantity $\Delta\lambda_d = \lambda^2/2Ln_0$) can be obtained from the expression^[2]

$$\Delta\lambda / \Delta\lambda_d = \gamma / (\gamma + \gamma_d),$$

where γ is the fluorescence line width, and $\gamma_d = \lambda^2\sigma/2\pi Ln_0$ is the width of the cavity resonance determined by the single-pass losses (for fluorite $n_0 = 1.42$).

It follows from this expression that the pulling effect becomes larger the narrower the fluorescence linewidth and the poorer the cavity Q. For a Gaussian line shape the dependence is qualitatively the same but the quantity $\Delta\lambda/\Delta\lambda_d$ depends also on the position of the mode with respect to the center of the fluorescence line; that is, the equal spacing of the modes may be perturbed.

We have investigated the stimulated emission spectra of crystals of $\text{CaF}_2:\text{Dy}^{2+}$ operating both continuously and pulsed at 2.36μ and also crystals of $\text{CaF}_2:\text{U}^{3+}$ operating pulsed as a three level laser

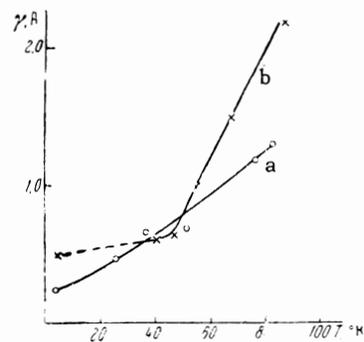


FIG. 1. Temperature dependence of the half-width of the fluorescence line in $\text{CaF}_2:\text{Dy}^{2+}$ (a) and in $\text{CaF}_2:\text{U}^{3+}$ (b, in this case the ordinate is $\gamma/10$).

($\lambda = 2.22 \mu$)^[4]. The temperature of the crystals was varied between 30 and 100°K. The temperature dependence of the fluorescence line width in $\text{CaF}_2:\text{Dy}^{2+}$ and $\text{CaF}_2:\text{U}^{3+}$ crystals is shown in Fig. 1. Spectroscopic studies established that in the temperature range used the line shape of the $\text{CaF}_2:\text{U}^{3+}$ line at 2.22μ was approximately Lorentzian; in the case of $\text{CaF}_2:\text{Dy}^{2+}$ the fluorescence line shape is Gaussian, which is evidence of inhomogeneous broadening.

A Fabry-Perot etalon and photoelectric detection were used to study the spectra. The length of the etalon was varied between 10 and 30 mm. The mirrors used had multilayer dielectric coatings with a reflectivity of 98% at 2.36μ . The spectrum was scanned by placing the etalon in a variable-pressure chamber. For continuous laser operation the pressure in the chamber was varied smoothly and the spectrum was obtained with a recording potentiometer. For pulsed laser operation the pressure was varied in steps and the spectrum was obtained point by point from oscilloscope pic-

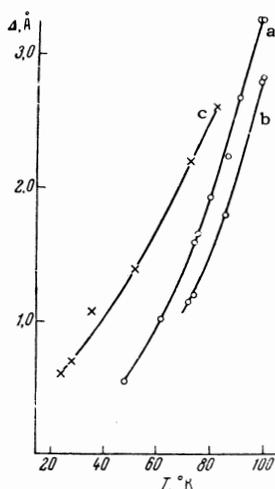


FIG. 2. Temperature dependence of the shift of the mode frequency (a and b) and of the fluorescence line (c) in $\text{CaF}_2:\text{Dy}^{2+}$ (the relative positions of the modes and the fluorescence line are arbitrary).

tures of the spikes for various pressures. The reproducibility of the results in this case was within 5%.

Special care was taken to ensure that the crystal temperature was constant during the experiment, since the temperature shift of the modes was very strong (cf. Fig. 2). A mode shift (of 0.1 Å) during pulsed operation was observed to occur during the emission pulse (due to crystal heating), and this was taken into account in reducing the data.

Examples of the stimulated emission spectrum are shown in Fig. 3. The width of all the modes observed is determined by the instrumental width of $(1/30)^{\text{th}}$ of the etalon interorder separation¹⁾

Figure 2 gives an example of the curve of the temperature shift of a mode in $\text{CaF}_2:\text{Dy}^{2+}$; also shown is a curve of the temperature shift of the fluorescence line.

The results obtained by averaging a series of measurements are shown in the table. The mode pulling is characterized in the table by the ratio $\Delta\lambda/\Delta\lambda_d$, where $\Delta\lambda$ is the measured difference in wavelength between neighboring axial modes. The variation of the quantity $\Delta\lambda$ from measurement to measurement did not exceed $\pm 4\%$.

The number of modes observed for $\text{CaF}_2:\text{Dy}^{2+}$

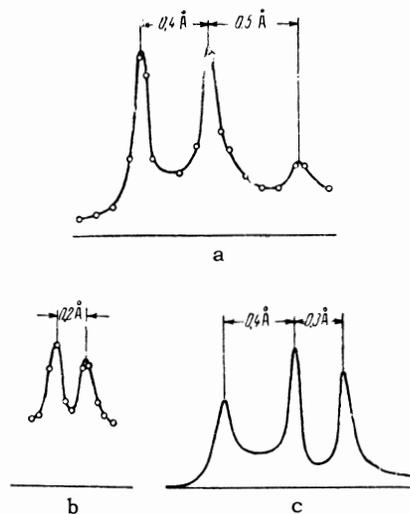


FIG. 3. The stimulated emission spectrum: a – for pulsed operation; $L = 40.5$ mm, $T = 97^\circ\text{K}$; b – pulsed operation with a confocal cavity; $L = 36.5$ mm, $T = 94^\circ\text{K}$; c – continuous operation; $L = 40.5$ mm, $T = 80^\circ\text{K}$

varied from 1 to 3, depending on the temperature of the crystal, the amount by which threshold was exceeded, and the transmission of the cavity mirrors. For sufficiently high temperatures the quantity $\Delta\lambda$ approaches the value $\Delta\lambda_d$. As the temperature is lowered, causing the fluorescence line to narrow, the quantity $\Delta\lambda$ decreases and in the case of $\text{CaF}_2:\text{Dy}^{2+}$ the equal spacing of the modes is not preserved. The relative magnitude of the pulling may be found from the above data and is very large; in a number of cases the quantity $\Delta\lambda/\Delta\lambda_d$ is between 0.6 and 0.7, and is 0.4 for $\text{CaF}_2:\text{U}^{3+}$. For c.w. operation the magnitude of $\Delta\lambda$ and the number of observed axial modes depended strongly on the degree to which threshold was exceeded; for pulsed operation this dependence was not observed. This may be explained by the fact that in continuous operation an increase in the pump power leads to a distortion of the cavity (cf. for example^[5]) and to a corresponding lowering of the Q. For pulsed operation the determination of the laser emission spectrum was carried out during the initial moments of laser action, at which time the crystal temperature and geometry had not changed.

It has been established that changing the diameter of the crystal from 3 to 5 mm does not effect the quantity $\Delta\lambda/\Delta\lambda_d$. The pulling effect was found to be quite significant for a confocal cavity. The quantity $\Delta\lambda/\Delta\lambda_d$ was found to be extremely small in the case of $\text{CaF}_2:\text{U}^{3+}$ at low temperatures, despite the large fluorescence linewidth observed for this crystal. The data given in the table for the intensities of the various modes make it possible to assert that the stimulated emission spectrum in

¹⁾Measurements of the mode width made by a photoelectric mixing technique for the case of cw stimulated emission in $\text{CaF}_2:\text{Dy}^{2+}$ gave a value of 1.8×10^5 cps (3.34×10^{-5} Å), which is in good agreement with the measured relaxation time (namely 5 μsec , $\Delta\nu/\Delta t \sim 1$). The total width of a group of modes having the same axial index (the width of an axial mode) was measured to be less than 2.7×10^8 cps (0.05 Å).

Crystal	L, mm	Mirror transmission	Operating regime	T, °K	Number of modes	$\Delta\lambda$, Å	Ratio of mode intensities	$\Delta\lambda/\Delta\lambda_d$
CaF ₂ :Dy ²⁺	29	20	c.w., threshold c.w., factor of 3 above threshold	~ 80	1	0.47	1:0.07	0.7
CaF ₂ :Dy ²⁺	40.5	20	c.w., threshold c.w., factor of 3 above threshold pulsed	~ 80	2	0.46	0.65:1	0.95
				~ 80	3	0.4; 0.3	0.5:0.5:1	0.83; 0.62
				98	3	0.43; 0.48	1:0.9:0.5	0.80; 1.0
CaF ₂ :Dy ²⁺	40.5	2	pulsed	~ 72	1			0.83
				74	2	0.4		0.93
				86	2	0.45		1.0
				100	2	0.48		
CaF ₂ :Dy ²⁺	36.5 confocal resonator	5	pulsed	94	2	0.2	0.7:1	0.74
CaF ₂ :U ³⁺	23	53	pulsed	28	1			
				46	2	0.3	1:0.7	0.30
				68	≥ 3	0.54		0.71
				86	> 4	0.76		

a number of cases is very asymmetrically located with respect to the fluorescence line.

The results we have obtained show that the effect of anomalous dispersion on the stimulated emission spectrum or crystals (leading to mode pulling) may be quite significant. Moreover, when the fluorescence line shape is Gaussian the equal spacing between the modes is perturbed. The magnitude of the mode pulling (if one excludes thermal effects) does not depend on the pump power within the experimental accuracy.

It should be pointed out that in a number of cases the total width of the laser emission spectrum in CaF₂:Dy²⁺ crystals is more than half the line width of the fluorescence, although for CaF₂:U³⁺ crystals the ratio is somewhat smaller. This fact and also the Gaussian line shape indicate that the broadening mechanism for the fluorescence line in the crystal of CaF₂:Dy²⁺ is inhomogeneous.

A rough estimate of the magnitude of the mode pulling using the expressions given in this paper shows that the magnitude of $\Delta\lambda/\Delta\lambda_d$ observed for CaF₂:Dy²⁺ should occur for a single pass loss which is 3–5 times greater than the observed losses as determined from the energy characteristics of the stimulated emission^[6]. An even larger discrepancy exists for CaF₂:U³⁺.

In order to give a complete quantitative interpretation of the results obtained here it would obviously be necessary to take account of effects related to the field distribution in the cavity and a series of other effects considered in^[7,9].

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