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OBSERVATION OF PARAMETRIC LUMINESCENCE IN A LITHIUM NIOBATE CRYSTAL EXCITED BY AN ARGON LASER

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The emission of visible light in the wavelength range $\lambda = 5200$ to 7000 Å by a lithium niobate crystal excited by an argon laser was observed, the value of λ being dependent on the angle between the direction of observation and the laser ray. The spectral power of the emission amounts to $\sim 4 \times 10^{-12}$ W/Å. The effect is attributed to the decay of each pump photon into two photons, the momenta of which satisfy the condition of spatial synchronism. Formulas for the main characteristics of the effect are obtained by using a simple model. Comparison of the experimental and theoretical results yields for the quadratic susceptibility of the crystal a value on the order of 4×10^{-8} abs. un. This value is in agreement with the value known from frequency-doubling experiments.

1. This paper is devoted to an investigation of a new type of scattering of light in a medium having nonlinear polarizability $P(t) = \hat{\chi}E^2(t)$. This effect, called "parametric luminescence"[1,2] (PL), is due to spontaneous decay of photons of the exciting light $\hbar\omega_3 \rightarrow \hbar\omega + \hbar\omega_2$, with $k_3 - k_1 - k_2 \equiv \Delta \approx 0$.

PL was first observed, in essence, by Akhmanov et al.[3], who used a pulsed laser with intensity ~ 10 MW cm^2, and regarded the PL as noise radiated by a parametric light amplifier. According to the estimate given in[2], the intensity of the PL should reach an appreciable value at much lower excitation levels, when the gain is certainly smaller than unity. A particular case of PL (at scattering angles $\vartheta \sim 0$) was observed also in[4]; the treatment of the PL in these papers is based on a premise which in our opinion is incorrect, namely on the notion of "one-dimensional" ($k_1 \parallel k_2 \parallel k_3$) interaction of the waves.

We present in this communication preliminary results of observation of "two-dimensional" PL in a lithium niobate crystal excited with an argon laser ($\lambda_3 = 4880$ Å) with a power P_3 up to 0.15 W, and also a simplified calculation of the main characteristics of the PL.

2. The experimental setup is shown in Fig. 1. The optical axis of the crystal (with length $l = 1.0$ cm) is parallel to the laser field e_3. The PL can be easily observed with the unaided eye in the form of colored rings (Fig. 2).

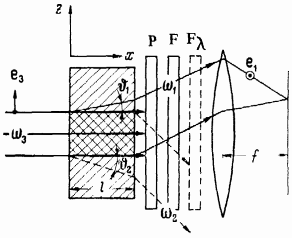

FIG. 1. Diagram of experiment. P – polaroid with axis perpendicular to the plane of the figure, F – filter transmitting the yellow-red part of the spectrum; F$_\lambda$ – interference filter.

Owing to the dispersion of the refractive index n, the condition $\Delta \approx 0$ leads to practically total polarization of the PL ($e_{1,2} \perp e_3$) and to a rigorous connection between the frequency $\nu = \omega/2\pi c$ and the angle ϑ (Fig. 3). The tuning curve $\vartheta(\nu)$ (solid line) is determined from the equation $\Delta = 0$. The experimental points were obtained with the aid of interchangeable interference filters. The measured angular width of the tuning curve at $\lambda \sim 6000$ Å is of the order of several minutes (inside the crystal). We note that PL with lower intensity and directivity was observed also at frequencies $\nu_1 \sim 1.85 - 1.92 \times 10^4$ cm^{-1} (5400—5200 Å); these waves correspond to the "additional" frequencies $\nu_2 \equiv \nu_3 - \nu_1 < 2 \times 10^3$ cm^{-1} ($\lambda_2 > 5\ \mu$), lying in the region of

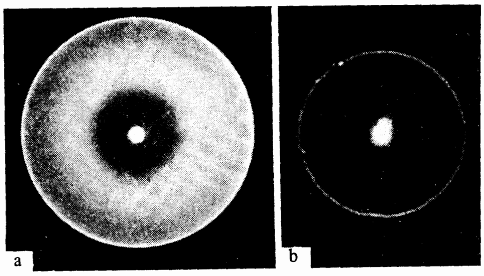

FIG. 2. a) Photograph of parametric luminescence, illustrating the distribution of radiation over the directions. The internal boundary of the ring is determined by the decrease of the sensitivity of the film when $\lambda < 7000$ Å (the central bright spot is connected with parasitic pump radiation); b) the same in the presence of a narrow-band filter ($\lambda = 6700$ Å, $\Delta\lambda \sim 50$ Å).

the infrared absorption of the crystal[1)].

The PL intensity was linearly dependent on P_3 (in the interval 0.03–0.13 W at a laser beam cross section 0.02 cm^2). An estimate of the absolute magnitude of the spectral power P_λ gave a value[2)] 4×10^{-12} W/Å (at $P_3 = 0.1$ W and $\lambda = 6328$ Å).

3. The PL intensity can be determined with the aid of the following illustrative considerations. Assume that a wave of frequency ω_2 propagates in a transparent crystal excited by a plane monochromatic wave ω_3. Owing to the effect of frequency subtraction, radiation of intensity S_1 is produced, the power conversion coefficient being equal to

$$K_{21} \equiv \frac{S_1 \cos\vartheta_1}{S_2 \cos\vartheta_2} = \frac{8\pi^3\omega_1{}^2\chi^2 S_3 l^2 F}{c^3 n_1 n_2 n_3 \cos\vartheta_1 \cos\vartheta_2}, \tag{1}$$

where $\chi \equiv |\mathbf{e}_1 \hat{\chi}\, \mathbf{e}_3 \mathbf{e}_1|$, n is the refractive index, and

$$F \equiv (\sin \Delta_x l / 2)^2 / (\Delta_x l / 2)^2.$$

In PL, the role of the radiation S_2 is played by quantum fluctuations having an intensity per mode and per unit of volume $\hbar\omega_2 v_2$, where v is the group velocity. For each mode of frequency ω_1 there are $v_1 \cos\vartheta_1 / v_2 \cos\vartheta_2$ modes of frequency $\omega_3 - \omega_1$, satisfying the condition $\Delta_y = \Delta_z = 0$; therefore

$$S_2 = \hbar\omega_2 v_1 \cos\vartheta_1 / \cos\vartheta_2.$$

Multiplying (1) by the density of the modes having the same polarization, we get the spectral brightness of the PL:

$$S_{\omega\Omega} = n^2 \hbar\omega^3 K / 8\pi^3 c^2, \tag{2}$$

where $K \equiv K_{21}\omega_2/\omega_1$. It follows from (2) that the effective temperature of the PL is $\hbar\omega/k \ln(1 + K^{-1})$.

The brightness S_Ω is equal to $\int S_{\omega\Omega} d\omega \equiv S^0_{\omega\Omega}\Delta\omega$, where $S^0_{\omega\Omega}$ is the maximum value of $S_{\omega\Omega}$ (at $F = 1$), and if $d\vartheta/d\omega \neq 0$, then

$$\Delta\omega_1 = 2\pi |\cos\vartheta_2| / |v_1^{-1} \cos(\vartheta_1 + \vartheta_2) - v_2^{-1}| l. \tag{3a}$$

Similarly, if we neglect the weak dependence of χ on

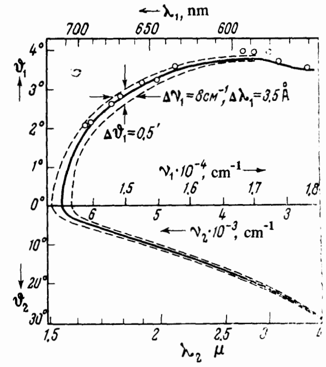

FIG. 3. Dependence of the scattering angles ϑ (inside the crystal) on the frequency ν for the visible (upper plot) and infrared (lower plot) regions of the spectrum. The dashed lines show the lines on which the spectral brightness of the luminescence decreases, in accordance with calculation, by a factor of about 2 at $l = 1$ cm (the width of the tuning curve has been magnified 40 times).

φ (at small values of ϑ), then $S_\omega = S^0_{\omega\Omega} 2\pi \sin\vartheta\Delta\vartheta$, where we have when $d\omega/d\vartheta \neq 0$

$$\Delta\vartheta_1 = 2\pi |\operatorname{ctg}\vartheta_2| / k_3 l = \Delta\omega_1 d\vartheta_1 / d\omega_1. \tag{3b}$$

Formulas (3) determine the width of the tuning curve (see Fig. 3) in the ideal case of a plane transparent scattering layer with small angular and spectral pump widths.

It is convenient to characterize the scattering efficiency by the following angular (at $d\vartheta/d\omega \neq 0$) and spectral ($d\omega/d\vartheta \neq 0$) scattering coefficients:

$$R_\Omega \equiv \frac{P_\Omega}{S_3 V} = \frac{2\pi\hbar\omega_1{}^4\omega_2\chi^2 n_1}{c^5 n_2 n_3 |v_1^{-1}\cos(\vartheta_1 + \vartheta_2) - v_2^{-1}|}, \tag{4a}$$

$$R_\omega \equiv \frac{P_\omega}{S_3 V} = 2\pi \sin\vartheta \cdot \frac{d\vartheta}{d\omega} R_\Omega = \frac{4\pi^2\hbar\omega_1{}^3\omega_2{}^2\chi^2}{c^4 n_3{}^2 \omega_3}. \tag{4b}$$

We note that formulas (4a) and (4b) are valid for an arbitrary form of the scattering volume v and width of the tuning curve, and also in the presence of absorption at the additional frequency[3)].

4. Subsituting in (4b) the experimental value of R_λ, which equals $P_\lambda/P_3 l \sim 4 \times 10^{-11} (\text{cm} \cdot \text{Å})^1$, we obtain $\chi_{yxy} \sim 2d_{15}^{2\omega} \sim 4 \times 10^{-8}$ absolute units. This coincides in order of magnitude with the result of measurement of χ by means of the frequency-doubling effect at $\lambda = 1.15\ \mu$[5].

According to (4), the experimental value of R_λ corresponds to a value of R_Ω equal to $4 \times 10^{-6} (\text{cm} \cdot \text{sr})^{-1}$. For comparison we indicate the following values of R_Ω for scattering by optical and acoustic lattice vibrations: 10^{-6}–10^{-7} (estimate by Loudon[6]) and 10^{-7} (measurements in quartz and in rock salt[7]).

5. The observed relatively large magnitude of the PL effect should make it a useful source of radiation with definite temporal and spatial coherence. It is obvious that by optimizing the conditions of the experiment it is possible to increase greatly the intensity of the PL. The observation of the PL should facilitate the problem of producing parametric light generators.

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[1)] As ν_2 approaches to the frequencies of the polar lattice vibrations, the PL obviously goes over into Raman scattering by polaritons [8,9]. We note that when using the concept of excitons in its broad meaning [10], the PL effects are generally indistinguishable from Raman-scattering effects.

[2)] Here and throughout the intensities and the scattering angles referred to the interior of the crystal.

[3)] In the case of a flat sample (when $R_\Omega = S_\Omega \cos\vartheta / S_3 l$), formula (4a) coincides with the result obtained in [2] by a probability method.

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