SCATTERING OF LIGHT IN A MEDIUM WITH NONLINEAR POLARIZABILITY

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We consider the spontaneous scattering of light due to quadratic and cubic terms in the expansion of the polarization of the medium in powers of the electric field. The medium is assumed to be transparent at the frequency of the incident radiation and at the observed frequency. Formulas are obtained for the intensity of the scattered light and for its dependence on the frequency and on the observation direction.

Two new types of light scattering by a substance were observed recently[1–6] and called "parametric luminescence"[1,7] (or "parametric scattering"[3]) and "scattering of light by light."[8–10]

From the quantum point of view, parametric luminescence (PL) is the result of the spontaneous decay of the photons of the incident light ("pump") into pairs of photons ($\hbar\omega_3 \to \hbar\omega_1 + \hbar\omega_2$) as a result of interaction with matter, and is described by the third order of ordinary perturbation theory. When $\hbar\omega_2 \lesssim kT$ and in the presence of absorption at ω_2, the anti-Stokes process $\omega_3 + \omega_2 \to \omega_1$ is, of course, also possible. PL can be explained phenomenologically by assuming that the medium has a nonlinear polarizability of the type $\mathbf{P}_1 = \hat{\chi}\mathbf{E}_3\mathbf{E}_2$. Then, in the presence of a pump field E_3, the thermal and (if $\omega_1 < \omega_3$) quantum fluctuations of the field E_2 with frequency ω_2 lead to polarization of the medium, and consequently to emission of frequency ω_1. Thus, the quadratic polarizability of the medium produces, besides the so-called three-photon scattering with frequency $2\omega_3$ (see, for example, [11]), one more type of scattering—with frequencies ω and (if $\omega < \omega_3$) $\omega_3 - \omega$, where ω is in general arbitrary (in the interval from zero to $\omega_3 + kT/\hbar$).

The emission intensity at a given frequency has a sharp maximum in directions determined by the dispersion of the refractive index $n(\omega)$ and by the condition of the spatial synchronism $\mathbf{k}_1 + \mathbf{k}_2 = \mathbf{k}_3$, which, as is well known, can be satisfied in anisotropic crystals. These directions form a circular cone with an axis parallel (or almost parallel) to $\mathbf{k}_3$. The brightness of the PL at low pump intensities S_3 is proportional to S_3 and to the size of the scattering volume. However, at sufficiently large S_3, induced processes, which lead to a superlinear dependence, come into play. In this case the effect can be defined as noise radiation of a parametric light amplifier and called "parametric superluminescence"—in analogy with the term used to describe the noise of quantum amplifiers.

Analogously, scattering of light by light can be regarded as a four-photon process $\omega_3 + \omega_3' \to \omega_1 + \omega_2$, or else as radiation produced by cubic polarization $\mathbf{P}_1 \hat{\theta}\mathbf{E}_3\mathbf{E}_3'\mathbf{E}_2^*$ and fluctuations of the field E_2 (see [8] concerning the contribution of quadratic polarizability to this process).

In this paper we determine the intensity of the spontaneous scattering and its dependence on the frequency and the observation direction (emission line shape) with allowance for dispersion. Calculations of the PL and of the scattering of light by light are perfectly analogous, and therefore principal attention is paid to PL (Secs. 1 and 2). In Sec. 1 we consider PL in a medium that is transparent at all three frequencies. Since the procedure for solving the parameters χ and θ is well known and they have been measured with the aid of induced effects in a large number of substances, we start from a phenomenological description of the nonlinearity of the medium. The energy of the field contains in this case the terms χE^3 and θE^4, and therefore the scattering probability is determined by the first order of perturbation theory.[9] For simplicity we consider a uniaxial negative crystal, in which the synchronism condition is satisfied for the ordinary scattered waves and for the extraordinary pump wave. In Sec. 2 we calculate with the aid of the fluctuation-dissipation theorem the PL for the case when there is absorption at the "additional" frequency $\omega_2 = \omega_3 - \omega_1$. In Sec. 3 we consider the changes that must be introduced into the obtained expressions in the case of scattering by a cubic medium.

1. LIGHT SCATTERING IN A TRANSPARENT MEDIUM WITH QUADRATIC POLARIZABILITY

We consider an unbounded dispersive medium, in which we separate a scattering volume V with a polarizability that is quadratic in the field. The dielectric constant of the medium is modulated by a monochromatic plane pumping wave $E_3 \exp i(\mathbf{k}_3 \cdot \mathbf{r} - \omega_3 t) + \text{c.c.}$ The weak scattered field is expressed in terms of the creation and annihilation operators:

$$E = \int d\mathbf{k}\, i \frac{\sqrt{\hbar k v_k}}{2\pi n_k} a_\mathbf{k} e^{i\mathbf{k}\mathbf{r}} + \text{h.c.} \tag{1}$$

where v_k is the group velocity, and a δ-function normalization is used: $[a_{\mathbf{k}'}, a_\mathbf{k}^+] = \delta(\mathbf{k}' - \mathbf{k})$ (see [9]).

In the presence of pumping, the nonlinear polarizability χ leads to modulation of the field energy with frequency ω_3:

$$H(t) = -\int\limits_V d\mathbf{r} \int d\mathbf{k}\, d\mathbf{k}' \frac{\hbar\chi\sqrt{kk'v_k v_{k'}}}{8\pi^2 n_k n_{k'}} E_3 a_\mathbf{k}^+ a_{\mathbf{k}'}^+ e^{i(\mathbf{k}_3 - \mathbf{k} - \mathbf{k}')\mathbf{r} - i\omega_3 t} + \text{h.c.} \tag{2}$$

In (2) we have retained only the terms corresponding to the Stokes scattering (the case when $\omega > \omega_3$ is not considered, since the changes that should be introduced in this case are obvious). By χ is meant the sum $\chi_{ajk} \times (\omega, \omega', \omega_3)e_i e_j' e_{3k}$, where $\mathbf{e}$ are unit polarization vectors (we use Pershan's definition of χ [12]).

With the aid of (2) we obtain the rate of production of photon pairs in the $\mathbf{k}_1$ and $\mathbf{k}$ modes:

$$W_{k_1k} = 2\pi\hbar^{-2}|\langle 1,1|H|0,0\rangle|^2\delta(\Delta\omega),$$

where $\Delta\omega = \omega_3 - \omega_1 - \omega(\mathbf{k})$. The total rate of production of the $\mathbf{k}_1$ photons is

$$W = \int W_{k_1k}d\mathbf{k}.$$

Consequently the power dissipated at the frequency ω_1 in the direction of $\mathbf{k}_1$ in unit spectral and angular intervals $d\omega_1$ and $d\Omega_1$ is

$$P_{\omega\Omega} = \hbar\omega_1 Wd\mathbf{k}_1/d\omega_1 d\Omega_1 = CS_3Vv_2\int d\mathbf{k}f(\Delta\mathbf{k})\delta(\Delta\omega), \quad (3)$$

where

$$C = 2\pi\hbar\omega_1^4\omega_2 n_1\chi^2/c^5n_2n_3,$$

$$f(\Delta\mathbf{k}) = \left|\int_V d\mathbf{r}\exp i\Delta\mathbf{k}\mathbf{r}\right|^2 \Big/ 8\pi^3V, \quad \Delta\mathbf{k} = \mathbf{k}_3 - \mathbf{k}_1 - \mathbf{k}.$$

In (3), the smoothly varying quantities are taken outside the integral sign.

If we are not interested in the form of the emission line (i.e., in the dependence of $P_{\omega\Omega}$ on ω_1 and on the scattering direction), then we can replace $f(\Delta\mathbf{k})$ in (3) by $\delta(\Delta\mathbf{k})$. After integration, we obtain the following expression for the differential scattering coefficient (or the extinction coefficient[13]):

$$R_{\omega\Omega} \equiv P_{\omega\Omega}/S_3V = C\delta(q), \quad (4)$$

where

$$q \equiv k_{20} - k_2, \quad k_{20} \equiv k(\omega_3 - \omega_1), \quad \mathbf{k}_2 \equiv \mathbf{k}_3 - \mathbf{k}_1.$$

We introduce a spherical coordinate system ($\omega_1, \vartheta_1, \varphi_1$) with the axis parallel to $\mathbf{k}_3$. The synchronism condition $q = 0$ defines a surface $\omega_1(\vartheta_1, \varphi_1)$, in which $P_{\omega\Omega}$ is maximal. Since $k_{1,2}$ are ordinary waves, it follows that ω_1 depends only on ϑ_1. We note that at small values of ϑ the synchronism surface is a paraboloid of revolution: $\omega_1 - \omega_{10} = [k_1k_3/2k_2(v_1^{-1} - v_2^{-1})]\vartheta_1^2$.[1)] The function $\omega_1(\vartheta_1)$ can be called the tuning characteristic of the PL. The derivatives of $q(\omega_1, \vartheta_1)$ at $q = 0$ are equal to

$$\partial q/\partial\omega_1 = v_1^{-1}\cos(\vartheta_1 + \vartheta_2) - v_2^{-1} \equiv v^{-1},$$
$$\partial q/\partial\vartheta_1 = k_3\sin\vartheta_2, \quad (5)$$

where $\sin\vartheta_2 = k_1\sin\vartheta_1/k_2$. The ratio of these derivatives defines the slope of the tuning curve $d\omega_1/\vartheta_1$.

Integrating (4) with the aid of (5), we determine the power

$$P_\Omega = \int P_{\omega\Omega}d\omega_1,$$

dissipated in a unit solid angle, and the power

$$P_\omega = \int P_{\omega\Omega}d\Omega_1,$$

dissipated in a unit spectral interval. The results are conveniently expressed in terms of the angular and spectral scattering coefficients:[2)]

$$R_\Omega \equiv P_\Omega/S_3V = C|v|, \quad (6a)$$
$$R_\omega \equiv P_\omega/S_3V = 2\pi k_2C/k_1k_3. \quad (6b)$$

As will be shown in Sec. 2, these expressions are valid also in the case of absorption at the frequency ω_2.

Let us estimate from (6b) the total scattering coefficient, assuming that the condition $q = 0$ is satisfied in the entire frequency interval $0 - \omega_3$. If we neglect the $\chi(\omega_1)$ dependence and the contribution of the thermal fluctuations, then

$$R = \int_0^{\omega_3} R_\omega d\omega_1 = \pi^2\hbar\omega_3^5\chi^2/15c^4n_3^2. \quad (7)$$

In lithium niobate $\chi \sim 4\times10^{-8}$ absolute units,[5] so that at $\lambda_3 = 0.5\ \mu$ we get $R \sim 10^{-7}\ \text{cm}^{-1}$. Observation of the PL effect by means of such a small additional coefficient of pump absorption is hardly possible (the scattered radiation itself can be readily observed with the unaided eye using a pump of 0.1 W[1,5]).

Let us consider now the shape of the line $P_{\omega\Omega}(\omega_1 - \omega_1^*, \vartheta_1 - \vartheta_1^*)$, where ω_1^* and ϑ_1^* are connected by the condition $q = 0$. The line width in the case of a transparent sample is determined by the finite size of the scattering volume or the coherence volume of the pump $1/\Delta\mathbf{k}_3$. Let us consider first the former case ($\Delta\mathbf{k}_3 \sim 0$). Let the scattering volume have the form of a plane-parallel layer of thickness l and cross section A. We direct z along l, and let $\mathbf{k}_3 \| z$.[3)] Let also the scattering angles be small (or close to π): $|\tan\vartheta_{1,2}| \ll l/\sqrt{A}$, and then

$$f(\Delta\mathbf{k}) = \delta(\Delta k_x)\delta(\Delta k_y)g_0(\Delta k_z l/2)l,$$

where $g_0(\eta) = \sin^2\eta/2\pi\eta^2$. After integration, we get from (3)

$$R_{\omega\Omega} = C'g_0(x/2),$$
$$C' \equiv Cl/|\cos\vartheta_2|, \quad x(\omega_1, \vartheta_1) \equiv ql/|\cos\vartheta_2|. \quad (8)$$

Formula (8) with allowance for (5), determines the dependence of $P_{\omega\Omega}(\omega_1, \vartheta_1)$ in the case of a plane transparent sample.

Let us express the obtained results in terms of the spectral brightness of the PL, $S_{\omega\Omega} = P_{\omega\Omega}/A|\cos\vartheta_1|$ and in terms of the conversion coefficient K, which equals the ratio of the emission power at the frequency ω_1 to the incident power at the frequency ω_2, multiplied by ω_2/ω_1, in the experiment on the frequency subtraction:[4)]

$$K = \frac{(2\pi)^3\omega_1\omega_2\chi^2S_3l^2g_0}{c^3n_1n_2n_3|\cos\vartheta_1\cos\vartheta_2|}. \quad (9)$$

We denote by I_0 the spectral brightness of the equilibrium emission of one polarization at the temperature $\hbar\omega_1/k\ln 2$, and then (8) takes the form[5,7]

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1) If k_2 is the extraordinary wave, then the axis of the synchronism paraboloid for the frequency ω_1 is shifted through an angle $\alpha = (k_2/k_3)(\Delta n_2/n_2)\sin 2\theta_3$ towards the crystal axis ($\Delta n_2 = n_2^e - n_2^o$ is the birefringence at the frequency ω_2 and θ_3 is the angle between k_3 and the crystal axis); the shift of the paraboloid axis for ω_2 is $-\alpha(k_1/k_2)$.

2) χ^2 in (6b) should be taken to mean the quantity averaged over φ (the $\chi(\varphi)$ dependence can be neglected at small ϑ).

3) If $\mathbf{k}_3^3$ is not parallel to the z axis, then it is necessary to replace $\cos\vartheta_{1,2}$ by $\cos\tilde{\vartheta}_{1,2}$ in the formulas that follow, where $\tilde{\vartheta}_{1,2}$ are the angles between $\mathbf{k}_{1,2}$ and z.

4) We note that in a transparent medium K is smaller than the coefficient of parametric amplification by unity.

$$S_{\omega\Omega} = KI_0, \quad I_0 = \hbar\omega_1^3 n_1^2 / 8\pi^3 c^2. \tag{10}$$

If we equate (10) to the spectral brightness of the thermal radiation with temperature T_{eff}, then we obtain the following expression for the effective temperature of the PL:

$$T_{\text{эфф}}(\omega, \vartheta) = \hbar\omega / k \ln(1 + K^{-1}). \tag{11}$$

Let us return to the line shape. We define the effective[5)] spectral line width $\Delta\omega$ as the ratio

$$P_\Omega / P_{\omega\Omega}^0 = 2\pi \int g_0 \, d\omega_1$$

($P_{\omega\Omega}^0$ is the "resonant" power at q = 0). Taking (5) into account, we get

$$\Delta\omega_0 = 2\pi |v \cos\vartheta_2| l^{-1}. \tag{12}$$

Analogously, the angle width of the tuning curve at $\vartheta_{1,2} \neq 0, \pi$ (when $d\omega/d\vartheta \neq 0$) equals

$$\Delta\vartheta_0 = 2\pi |\cos\vartheta_2| / k_3 l \sin\vartheta_2. \tag{13}$$

We note that formulas (6a) and (12) become meaningless when $|v| = \infty$, particularly in the degenerate "one-dimensional" case, when $\omega_1 = \omega_2$ and $\vartheta_1 = \vartheta_2 = 0$. Then $\partial^2 q/\partial\omega_1^2 = -4c^{-1} dn_1/d\omega$, and we have in place of (12) (assuming in (4) that $\delta(q) = l\, g_0(x/2)$):

$$\Delta\omega_0^* = {}^4\!/_3 \sqrt{\pi c (l\, dn_1 / d\omega)^{-1}}. \tag{12a}$$

In the degenerate one-dimensional case it is necessary to add in (6a) the factor $2\Delta\omega_0/\Delta\omega_0$, so that R_Ω is proportional in this case to the square root of the layer thickness ($\sqrt{l}$).

In linear scattering, the non-monochromaticity and the divergence of the pump (as well as its coherence properties) do not affect the integral radiation power P_Ω (or P_ω), and only increase the line width. Let the pump field be δ-correlated in $\mathbf{k}_3$ with an intensity distribution function $g_3(\mathbf{k}_3 - \mathbf{k}_{30})$, and then the line shape will be given by formula (4) in which $\delta(q)$ is replaced $\int d\mathbf{k}_3 g_3 \delta(q)$. The broadening of the line due to the non-ideal pumping can be readily estimated by varying the equality q = 0. For example, the pump divergence $\Delta\theta_3$ in the principal section of the crystal leads at $\vartheta = 0$ to an additional spectral line broadening by an amount

$$\Delta\omega_\theta = \Delta\omega_0 l / l_{coh} \tag{12b}$$

where $l_{coh} = \lambda_3/\Delta n_3 \Delta\theta_3 \sin 2\theta_3$, and Δn_3 is the birefringence at the pump frequency. Here $P_{\omega\Omega}^0 \approx P_\Omega/\Delta\omega_\theta \sim l$, so that $P_{\omega\Omega}^0$ is proportional to l^2 only when $l \ll l_{coh}$.

2. SCATTERING IN ABSORPTION AT THE ADDITIONAL FREQUENCY

The probability calculation employed above is applicable when $\alpha_n l / |\cos\vartheta_n| \ll 1$, where α_n is the absorption coefficient at the frequency ω_n, n = 1, 2, 3. Interest attaches also to the case when the medium is transparent only at the frequencies ω_1 and ω_3[6)] (scattering at $\alpha_2 l > 1$ was observed in [3,5]). It is obvious that as ω_2 approaches the frequencies of the polar oscillations of the lattice, the PL goes over into scattering by polaritons. We consider here the intermediate case, when the absorption is still not too large ($\varepsilon'' \ll \varepsilon'$, where ε is the dielectric constant of the medium at the frequency ω_2), so that the longitudinal field component can be disregarded.

In the analysis of scattering in a non-transparent medium it is convenient to use the classical theory of scattering[13] and the fluctuation-dissipation theorem for the electromagnetic field.[13, 14] The field produced in the far zone by the nonlinear polarization is equal to

$$E_1 = \frac{\omega_1^2 e^{ik_1R}}{c^2 R} \int_V d\mathbf{r}\, \mathbf{e}_1 \mathbf{P}_1 e^{-i\mathbf{k}_1\mathbf{r}} = \frac{\omega_1^2 \chi E_3 e^{ik_1R}}{c^2 R} \int_V d\mathbf{r}\, E_2^* e^{i\mathbf{k}_2\mathbf{r}}. \tag{14}$$

We represent the fluctuation field in the form $E_2 = \int d\mathbf{p} E_\mathbf{p} \exp(i\mathbf{p}\cdot\mathbf{r})$. The direction of observation $\mathbf{k}_1$, and consequently also $\mathbf{e}_1$, is fixed; the synchronization condition $\mathbf{p} \approx \mathbf{k}_2 \equiv \mathbf{k}_3 - \mathbf{k}_1$ determines also $\mathbf{e}_2$ ($\mathbf{e}_{1,2}$ are perpendicular to $\mathbf{k}_{1,2}$ and to the crystal axis). From (14) we get the power dissipated in the interval $d\omega_1 d\Omega_1$:

$$P_{\Omega\omega} = \frac{cn_1}{2\pi} R^2 \langle E_1 E_1^* \rangle = \frac{n_1 \omega_1^4 |\chi|^2 S_3}{n_3 c^4} \left\langle \left| \int d\mathbf{r}\, d\mathbf{p}\, E_\mathbf{p} e^{i(\mathbf{p}-\mathbf{k}_2)\mathbf{r}} \right|^2 \right\rangle. \tag{15}$$

According to [13], the $\mathbf{p}$-correlator of the transverse field equals

$$\langle E_\mathbf{p} E_{\mathbf{p}'}^* \rangle = \frac{\hbar(N_2 + 1)\delta(\mathbf{p} - \mathbf{p}')}{2\pi^3 \varepsilon'' [1 + 4(p - k_{20})^2/\alpha_2^2]}, \tag{16}$$

where $N_2 = (\exp(\hbar\omega_2/kT) - 1)^{-1}$. In (16) we doubled the term describing the zero-point fluctuations of the field. We shall henceforth assume that $\hbar\omega_2 \gg kT$, so that $N_2 = 0$.

Substituting (16) in (15), we find that the line shape is now Lorentzian:

$$R_{\omega\Omega} = |C'| g_1(x, y), \quad g_1(x, y) = y\pi^{-1} / (x^2 + y^2), \tag{17}$$

where $y = \alpha_2 l / 2 |\cos\vartheta_2|$. The integration of (17) with respect to ω_1 or Ω_1, with allowance for (15), yields again Eq. (6)—as expected, absorption directly at the additional frequency does not change the integral scattering coefficients. In lieu of (12) and (13) we now have

$$\Delta\omega_1 = \pi\alpha_2 |v| / 2, \tag{18a}$$

$$\Delta\vartheta_1 = \pi\alpha_2 / 2k_3 \sin\vartheta_2. \tag{18b}$$

We note that if v_2 is taken to mean the speed of sound ($v_2 \ll v_1$), then (18a) coincides with the well known expression for the effective width of the Mandel'shtam–Brillouin components.

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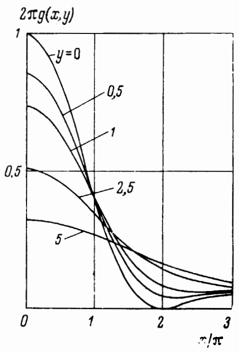
5)For the function g_0, the effective width is 13% larger than the width at the 0.5 level.

6)Absorption at the frequencies ω_1 and ω_3 causes, besides line broadening (in analogy with the broadening considered below), also a decrease of the effective scattering volume. The drop of the intensity can in this case, however, be compensated by the resonance in the $\chi(\omega_1)$ dependence.



Spectral brightness of scattered radiation vs. frequency (or scattering angle) at different absorption coefficients at the additional frequency.

In the intermediate case of a semitransparent layer (when $y \sim 1$), the line shape can be obtained in similar fashion. It is necessary here also to take into account the fluctuation field produced in the sample by the space outside the scattering layer (where $\varepsilon'' = 0$). When $y \lesssim 1$, the p-components of the field connected with the absorption inside the layer are no longer δ-correlated, leading to rather cumbersome calculations; we therefore present only the final results:

$$R_{\omega\Omega} = |C'|\, g(x, y),$$

$$g(x, y) = g_1 + \frac{(1 - e^{-y}\cos x)(x^2 - y^2) - 2xye^{-y}\sin x}{\pi(x^2 + y^2)^2}. \qquad (19)$$

This expression includes (8) and (17) as particular cases, namely, we have $g = g_0$ when $y \ll 1$ and $g = g_1$ when $y \gg 1$.[7)] A plot of $g(x, y)$ is shown in the figure. Since

$$\int_{-\infty}^{\infty} g\, dx = 1,$$

it follows that formulas (6) remain in force. The dependence of the line width on α_2 and l is determined by the resonant value of g:

$$\frac{\Delta\omega}{\Delta\omega_0} = \frac{1}{2\pi g(0, y)} = \frac{1}{2}\frac{y^2}{y + e^{-y} - 1}. \qquad (20)$$

3. SCATTERING OF LIGHT IN A MEDIUM WITH CUBIC POLARIZABILITY

In media with inversion centers, scattering is possible as a result of a real cubic polarizability θ.[8,9] Assume now that we have two incident light beams with frequencies ω_3 and ω_3'. The synchronism condition $\mathbf{k}_3 + \mathbf{k}_3' = \mathbf{k}_1 + \mathbf{k}_2$ can be satisfied also in isotropic media (at least when $\omega_1 \sim \omega_3$ and $\omega_2 \sim \omega_3'$). If we neglect dispersion, then the synchronism surface is an ellipsoid of revolution with interfocal distance $|\mathbf{k}_3 + \mathbf{k}_3'|$.[8,9] So far, spontaneous four-photon scattering was observed at $\omega_3 = \omega_3'$ and $\mathbf{k}_3 \perp \mathbf{k}_3'$.[6] Interest attaches also to the degenerate case $\mathbf{k}_3 = \mathbf{k}_3'$, when the tuning characteristic, with allowance for dispersion, has at $\omega_1 \sim \omega_3$ the form

$$\vartheta_1^2 = (\omega_1 - \omega_3)^2(\omega_1 / \omega_2 k_3)\, d^2k_3 / d\omega^2.$$

The effect should reach a considerable magnitude in the case of self-focusing of the incident radiation and when $\omega_1 \sim \omega_3$ or $\omega_1 \sim \omega_3 \pm \omega_0$ (ω_0—frequency of the molecular oscillations), when $\theta(\omega_1 = 2\omega_3 - \omega_2)$ has resonance maxima.

At real values of θ,[8)] the formulas obtained above remain in force, provided we make the substitutions $\omega_3 \to \omega_3' + \omega_3$, $\mathbf{k}_3 \to \mathbf{k}_3' + \mathbf{k}_3$, and $\chi \to \theta\mathbf{E}_3'$ (here $\theta \equiv \hat{\theta}_{ijkl}|e_{1i}e_{3j}e_{3k'}e_{2l}$, and account must be taken of the contribution of the different polarizations e_m). Thus, we get in place of (6)

$$P_\Omega = \frac{4\pi^2\hbar\omega_1^4\omega_2 n_1\theta^2|v|}{c^6 n_2 n_3 n_3'} VS_3S_3', \qquad (21a)$$

$$P_\omega = \frac{8\pi^3\hbar\omega_1^3\omega_2^2\theta^2}{c^6 n_3 n_3'|\mathbf{k}_3 + \mathbf{k}_3'|} VS_3S_3'. \qquad (21b)$$

If dispersion is neglected, (21a) coincides (apart from numerical coefficients) with the result of Robl.[8] Let us estimate (21b) at $\mathbf{k}_3 = \mathbf{k}_3'$ and $\omega_1 \sim \omega_3$. If the incident radiation has a diffraction divergence, then we get upon focusing of this radiation $VS_3^2 \sim 2P_3^2 n/\lambda$. Let $P_3 = 1$ MW, $\lambda_3 = 0.7\mu$, and $\theta = 10^{-13}$ cm^3/erg; then after a time $\tau = 10^{-8}$ sec; the number of photons scattered in a spectral interval of 1 cm^{-1} is $P_\omega\tau\lambda/\hbar = (2\pi)^7\theta^2P_3^2\tau / cn^2\lambda^4 \sim 10^3$.

4. CONCLUSION

The scattering model employed here agrees best with experiments in which a sample with "antireflection" surfaces is used. In this case, the conversion of the observed quantities to the parameters calculated above is trivial: it is necessary to take into account the refraction at the "output" surface, which leads, besides a change in the observed scattering angles, only to a decrease of the PL brightness by a factor n^2. We can expect that in the case of non-antireflection surfaces it is sufficient to take into account, in addition, only the losses for the single Fresnel reflection. The interference effects are significant in the case of specular boundaries and a sample of high optical grade, and even in this case the formulas presented above remain apparently in force, provided the bandwidth and the aperture of the receiver are sufficiently large: at small values of S_3 the interference leads to the appearance of a fine structure in the space-frequency distribution of the radiation, with the same average intensity.

Another limitation of the present paper is connected with the neglect of the induced effects, a procedure justified when $K \ll 1$ (see (9)). In practice, these effects become appreciable, even in a transparent crystal, only at pump intensities on the order of several megawatts per cm^2 (in the absence of mirrors) and at a sufficiently low pump divergence. It can be assumed that in this case formula (10) gives the correct order of magnitude of the spectral brightness of the superluminescence, provided K in this formula is taken to mean the conversion coefficient corresponding to these conditions.[7] At

[7)]A similar transition of the line shape from g_1 to g_0 takes place in the case of Mandel'shtam-Brillouin scattering when the attenuation of sound is decreased [15]. The line shape obtained in [15] is apparently close to the function g.

[8)]Resonant four-photon scattering is closely connected with Rayleigh scattering or Raman scattering and calls for a special analysis.

the present time, pulse lasers yielded a gain of several units.[4] The effective temperature of the PL should, according to (11), reach several tens of thousands of degrees.

The obtained expressions for the power of the scattered radiation (6) and (21) make it possible to measure the magnitude and dispersion of the polarizabilities χ[5] and θ. Such measurements are of particular interest in the case when ω_2 lies in the infrared or in the millimeter band, since they make it possible to determine the contribution of the vibrational levels. Observation of PL should also permit measurement of the dielectric constant in these ranges: n_2 can be determined from the synchronism condition if n_1 and n_3 are known,[3] and the line width, according to (20), determines ε''. We note finally, that PL is possible, in principle, also in optically active liquids in which $\chi \neq 0$ when $\omega_1 \neq \omega_2$.[16]

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[9)] Following the completion of this work, a paper dealing with PL in a transparent medium was published by Giallorenzi and Tang [[17]]. The expressions obtained in [[17]] for the radiation intensity coincide with formulas (4) and (6a) if $v = n/c$ (in the comparison it is necessary to take into account (12b) and the fact that the value of χ employed here is twice as large).

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