SCATTERING OF LIGHT BY LIGHT IN A NONCENTRALLY SYMMETRICAL MEDIUM

D. N. KLYSHKO and N. I. NAZAROVA

Moscow State University

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The main characteristics (intensity, dependence of frequency on scattering angle, line shape) of the radiation scattered by a transparent crystal not possessing a symmetry center are calculated. Scattering due to the quadratic and cubic terms in the expansion of the macroscopic polarizability in terms of the amplitude of the incident light (which is assumed to be monochromatic) is considered. It is shown that as a rule the efficiency of two consecutive three-photon processes is greater than the efficiency of a four-photon process. The effect of the finite cross section of the incident light beam is taken into account.

THE nonlinear dependence of the polarization of a transparent medium on the amplitude of the light field ($P^{(NL)} = \chi^{(2)}E^{(2)} + \chi^{(3)}E^{(3)}$) causes a unique light scattering, a distinguishing feature of which is an appreciable relative change of the frequency (on the order of unity) and large coherence scales (equal to the dimensions of the entire scattering region). The scattering due to the cubic polarizability $\chi^{(3)}$ was investigated in a number of works theoretically[1-3] and experimentally[4,5]. This phenomenon, called scattering of light by light (SLL) or four-photon scattering, is best treated as the result of elementary four-photon processes of the form $\omega_1 + \omega_1' \rightarrow \omega_s + \omega_a$, $\mathbf{k}_1 + \mathbf{k}_1' \rightarrow \mathbf{k}_s + \mathbf{k}_a$. Unlike scattering that is linear in the intensity S_1 of the incident light (see, for example,[3]) and is due to $\chi^{(2)}$, SLL causes field fluctuations also at the anti-Stokes frequencies.

As noted by Robl[1], in crystals without symmetry centers (in which $\chi^{(2)} \neq 0$) there is an additional SLL mechanism and, as will be shown below, $\chi^{(2)}$ makes in some cases (particularly at small angles between $\mathbf{k}_1$ and $\mathbf{k}_1'$) the principal contribution. This process is described by second-order perturbation theory (with a perturbation energy density $\mathbf{EP}^{NL}$ [1]), and can be represented as the result of two successive three-proton processes, the "virtual" wave being either the harmonic of the incident radiation ($\omega_2 \equiv 2\omega_1$) $\omega_1 + \omega_1 \rightarrow \omega_2 + \omega_s + \omega_a$ (we consider for brevity only the case $\omega_1' = \omega_1$, $\mathbf{k}_1' = \mathbf{k}_1$) or a difference wave ($\omega_0 \equiv \omega_1 - \omega_s = \omega_a - \omega_1$) $\omega_1 \rightarrow \omega_s + \omega_0$, $\omega_0 + \omega_1 \rightarrow \omega_a$.

The process in which ω_0 takes part was considered by Giallorenzi and Tang[6], who emphasized that this process limits the sensitivity of the infrared light receivers using parametric conversion of the frequency ($\omega_0 + \omega_1 \rightarrow \omega_a$) in the visible region (we note in this connection that the Bloembergen quantum counter, which can be regarded as a resonant frequency converter with increased output frequency, should also have a "noise" proportional to S_1^2).

Processes quadratic in the intensity of the incident pump light in crystals with large polarizability $\chi^{(2)}$ are of interest also in connection with the problem of producing sources of coherent light with a frequency exceeding the pump frequency. The threshold of excitation of the parametric generators, in which such crystals are used, can be lower than in the case of centrally-symmetrical media[7].

In this paper we consider SLL with account taken of all the three principal mechanisms: via virtual waves ω_0 and ω_2 and as a result of $\chi^{(3)}$. We use the following model: a flat nonlinear layer of thickness $2l$ is contained in an unbounded anisotropic dispersive transparent medium with homogeneous linear polarizability. A plane (Secs. 1 and 2) or almost-plane (Sec. 3) monochromatic pump wave with constant amplitude propagates in a direction perpendicular to this layer.

In Secs. 1 and 2 we use for the sake of clarity the classical calculation method, with the aid of a system of equations for the slowly-varying Fourier amplitudes of the field $E_i(z)$ ($i = 0, 2, a, s$), and the quantum uncertainty of the input amplitude $E_{a,s}(-l)$ is taken into account only in the next order. The starting point in Sec. 3, where the correctness of such a calculation is confirmed and the finite character of the cross section of the pump beam is also taken into account, are the Heisenberg equations for the operators $E_k(t)$. The employed approach is a generalization of the method of Louisell et al.[8], which makes it possible to treat quantum-mechanically the stationary problems of nonlinear optics, and particularly scattering processes.

1. CONVERSION COEFFICIENTS IN THE CASE OF A PLANE PUMP WAVE

Let $\mathbf{E}(\mathbf{r}, t) = \Sigma \mathbf{e}_i E_i(z) \exp(i\mathbf{k}_i\mathbf{r} - i\omega_i t) + \text{c.c.}$ and let the amplitudes of the scattered waves be much smaller than the pump amplitude; then the interaction of the waves in the nonlinear layer is described by the following system:

$$\begin{gathered} dE_2/dz = \beta_{21}e^{i\Delta_2 z}E_1, \\ dE_0/dz = \beta_{0s}e^{i\Delta_s z}E_s^* + \beta_{0a}^* e^{-i\Delta_a z}E_a, \\ dE_s/dz = \beta_{s0}e^{i\Delta_s z}E_0^* + (\beta_{as}'e^{i\Delta' z} + \beta_{as}''e^{i\Delta z})E_a^*, \\ dE_a/dz = \beta_{a0}e^{i\Delta_a z}E_0 + (\beta_{as}'e^{i\Delta' z} + \beta_{as}''e^{i\Delta z})E_s^*, \end{gathered} \tag{1}$$

where

$$\beta_{ij} = ib_i\chi_{ij}^{(2)}E_1, \quad \beta_{ij}' = ib_i\chi_{ij}^{(2)}E_2, \quad \beta_{ij}'' = ib_i\chi_{ij}^{(3)}E_1^2,$$

$$b_i = 2\pi\omega_i / cn_i', \quad n_i' = n_i\cos\alpha_i\cos\theta_i,$$

$$\Delta_s = k_1 - k_{sz} - k_{0z}, \quad \Delta_a = k_1 - k_{az} + k_{0z}, \quad \Delta_2 = 2k_1 - k_{2z},$$

$$\Delta' = k_{2z} - k_{sz} - k_{az}, \quad \Delta = \Delta_s + \Delta_a = \Delta_2 + \Delta';$$

χ_{ij} is the contraction of the nonlinear susceptibility tensor with the corresponding vectors e_i, n is the refractive index (with allowance for a correction proportional to $\chi^{(3)}|E_1|^2$), α is the angle between the ray and the wave vectors, and θ is the angle between the ray vector and the z axis (the scattering angle).

In an infinite layer, stationary interactions are produced only between waves satisfying the laws of conservation of the frequency and the transverse momentum, so that specification of the frequency, direction, and polarization of the observable wave (for example ω_a, k_a, and e_a) and of the pump wave at a known dispersion law $\omega_\lambda(k)$ leaves the polarizations and the signs of the longitudinal momenta of the remaining three waves undetermined. Usually, however, the wave detunings Δ are minimal for the forward waves ($\theta < \pi/2$) and for a definite combination of polarizations (for example $2k_1^o \to k_2^e \to k_a^e + k_s^o$ in a negative uniaxial crystal). We have therefore left out from (1) the polarization indices, and did not take into account the coupling with the backward waves.

According to (1), the harmonic is independent of the remaining waves:

$$E_2(z) = \beta_{21} E_1 \int_{-l}^{z} dz' e^{i\Delta_2 z'}. \qquad (2)$$

To find the amplitude of the spontaneously scattered field, for example at the frequency ω_s, it suffices to find the conversion coefficients K_{si} connecting the output amplitude $E_s(l)$ with the positive-frequency input amplitudes $E^*_{0,a}(-l)$. Putting therefore $E_i(-l) = 0$, we obtain the following amplitudes that differ from zero in first order in E_1:

$$E_s^{(1)}(z) = \beta_{s0} E_0^*(-l) \int_{-l}^{z} dz' e^{i\Delta_s z'}, \qquad (3a)$$

$$E_0^{(1)}(z) = \beta_{0s} E_s^*(-l) \int_{-l}^{z} dz' e^{i\Delta_s z'}, \qquad (3b)$$

$$E_0^{*(1)}(z) = \beta_{0a} E_a^*(-l) \int_{-l}^{z} dz' e^{i\Delta_a z'}. \qquad (3c)$$

Substituting (2) and (3) in the right sides of (1), we obtain, accurate to terms of order E_1^2,

$$E_s(l) = K_{s0} E_0^*(-l) + K_{sa} E_a^*(-l), \quad E_a(l) = K_{as} E_s^*(-l), \quad (4)$$

$$K_{s0} = \beta_{s0} 2l f(\Delta_s l), \quad f(x) = x^{-1} \sin x, \qquad (4a)$$

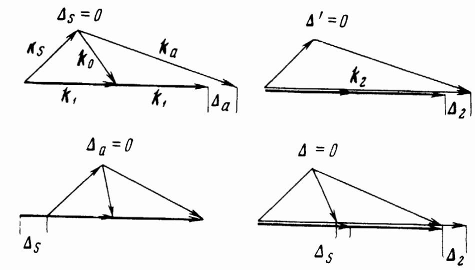
$$K_{sa} = \beta_{s0}\beta_{0a} 2l^2 f(\Delta_s l, \Delta_a l) + \beta_{sa}' \beta_{21} 2l^2 f(\Delta' l, \Delta_2 l) + \beta_{sa}'' 2l f(\Delta l), \quad (4b)$$

$$K_{as} = \beta_{a0}\beta_{0s} 2l^2 f(\Delta_a l, \Delta_s l) + \beta_{as}' \beta_{21} 2l^2 f(\Delta' l, \Delta_2 l) + \beta_{as}'' 2l f(\Delta l), \quad (4c)$$

$$f(x, y) = {}^1/_2 \int_{-1}^{1} dz e^{ixz} \int_{-1}^{z} dz' e^{iyz'} = i[f(x) e^{-iy} - f(y) e^{ix}]/(x + y).$$

Formula (4a) determines the amplitude of linear scattering (parametric luminescence—see, for example,[3,9] and the references given therein).

The function $|f(x, y)|$ has in the general case three resonant maxima, near which it is equal to $|f(x)/y|$ if $|y| \gg |x|$, to $|f(y)/x|$ if $|x| \gg |y|$, and to $|f(x+y)/x|$ if $x \approx -y$ and $|x| \gg |x+y|$. Thus, the connection between the frequency and the scattering direction in the case of coherent SLL is determined by the conditions $\Delta_s = 0$, $\Delta_a = 0$[1)], $\Delta' = 0$, $\Delta = 0$, and all three aforementioned mechanisms contribute to the last resonance (which can occur also in isotropic substances). These conditions specify four synchronism surfaces in k-space (see[3,9]). The arrangement of the wave vectors for different types of spatial synchronism is explained in the figure. The largest SLL effect as a result of $\chi^{(2)}$ is reached in the case of double resonance ($f(0, 0) = 1$), when $\Delta_2 = \Delta' = 0$ or $\Delta_s = \Delta_a = 0$.



Four arrangements of the wave vectors **k**, at which the scattering is maximal (the indices 1, 2, s, a, and 0 pertain respectively to the incident-radiation wave, its harmonic, and to the Stokes, anti-Stokes, and difference waves).

Let us compare with the aid of (4) the contributions of the different processes to the scattering amplitude at resonance, when $f(x) = 1$, $f(x, y) = \pm i/\Delta_i l$ (Δ_i—wave detuning of the corresponding non-synchronous interaction, see the figure). The ratio of the amplitude of quadratic scattering due to $\chi^{(2)}$ to the amplitude of linear scattering is of the order of β/Δ_i (or βl in the case of double resonance when $\Delta_i = 0$). In a lithium niobate crystal, the parametric gain factor β is equal to 1 cm^{-1} at S_1 10 MW/cm^2 (the last figure is decreased by four orders of magnitude if the polarization vectors of all three waves taking part in the non-synchronous process are parallel to the symmetry axis of the crystal). For waves of the same polarization, Δ_i reaches 10^3 cm^{-1}; when the polarizations are different, on the other hand, Δ_i can be much smaller (at a definite orientation of certain crystals, as is well known, $\Delta_2 = 0$; in addition, $\Delta_i = 0$ on the line of intersection of the synchronism surfaces Δ_a and Δ_s).

The ratio of the SLL amplitudes at the anti-Stokes frequency when $\Delta = 0$, due to $\chi^{(2)}$ and $\chi^{(3)}$, has according to (4c) the order of magnitude $(4\pi^2/n_0 \Delta_a \lambda_0)(\chi^{(2)2}/\chi^{(3)})$. Let $\chi^{(2)} = 10^{-8}$ $(cm^3/erg)^{1/2}$, $\chi^{(3)} = 10^{-15}$ cm^3/erg, $\Delta_a = 10^2$ cm^{-1}, $\lambda_0 = 2\mu$, and $n_0 = 2$; then this ratio is equal to 10^2.

2. INTENSITY OF SCATTERED LIGHT

To take into account the quantum fluctuations, we replace the slowly-varying amplitudes E_i and E_i^* by the operators E_k and E_k^+, which are proportional to the annihilation and creation operators and are determined by the relations

$$E(\mathbf{r}, t) = \int d\mathbf{k} E_{\mathbf{k}}(z) \exp i(\mathbf{kr} - \omega_{\mathbf{k}} t) + \text{h.c.}, \qquad (5)$$

$$\langle E_{\mathbf{k}}(-l) E_{\mathbf{k}'}^+(-l) \rangle = |c_{\mathbf{k}}|^2 \delta(\mathbf{k} - \mathbf{k}'), \qquad (6)$$

$$c_{\mathbf{k}} = i(\hbar\omega_{\mathbf{k}} u_{\mathbf{k}} / 4\pi^2 c n_{\mathbf{k}} \cos\alpha_{\mathbf{k}})^{1/2},$$

1) Scattering at $\Delta_a = 0$ determines the limiting sensitivity of a step-up frequency converter.

where the angle brackets denote averaging over the initial vacuum state and $u_k = \partial\omega_k/\partial k$ is the group velocity.

Let us fix $k = k_a$, and denote by $\tilde{k} = k_s$ the vector complementing k and defined by the equations

$$\omega_{\tilde{k}} = 2\omega_1 - \omega_k, \ \tilde{k}_\perp = k_\perp, \ \theta_{\tilde{k}} < \pi/2; \tag{7}$$

It then follows from a comparison of (5) with the definition of the amplitudes E_i that

$$E_k = E_a\delta(k - k_a), \quad E_k / E_{\tilde{k}} = (E_a / E_s) d\tilde{k}_z / dk_z = E_a u_{az} / E_s u_{sz},$$

so that we have in place of (4)

$$E_k(l) = (u_{az} / u_{sz}) K_{as} E_{\tilde{k}}^{\pm}(-l)$$

and when (6) is taken into account

$$\langle E_k^+(l) E_{k'}(l)\rangle = |K_{as}c_s|^2 (u_{az} / u_{sz}) \delta(k - k'). \tag{8}$$

The factor preceding the δ function determines the intensity of the fluctuations to the right of the nonlinear layer:

$$S = \int dk S_k, \quad S_k = (2\pi)^{-1} c n_a \cos\alpha_a \langle E_k^+(l) E_{k'}(l)\rangle / \delta(k - k').$$

In comparison with experiment, it is more convenient to use the concept of the spectral brightness of the radiation, i.e., the power per unit area, frequency, and solid angle (at a fixed direction k):

$$S_{\omega\Omega}(k) = S_k dk / d\omega d\Omega = S_k k^2 / u_k \cos\alpha_k,$$
$$S_{\omega\Omega}(k_a) = I_a |K_{as}|^2 \omega_s n_a' / \omega_a n_s', \tag{9}$$

where the quantity $I_a \equiv \hbar\omega_a k_a^2/8\pi^3 \cos\alpha_a$ can be defined as the spectral brightness of the vacuum fluctuations of the one polarization in the medium at the frequency ω_a.

We now assume that the pump beam has a Gaussian field distribution in the transverse cross section:

$$E_1(x, y) = E_1 \exp[-(x^2 + y^2) / w^2];$$

then

$$\int |E_1(x, y)|^4 dx\, dy = |E_1|^4 A/2,$$

where $A \equiv \pi w^2/2$ is the effective cross section of the beam, so that the power radiated in unit angle and spectral intervals (the spectral strength of the light) is equal to

$$P_{\omega\Omega}(k_a) = \int S_{\omega\Omega}(k_a) \cos\theta_a\, dx\, dy = {}^1/_2 A \cos\theta_a I_a |K_{as}|^2 \omega_s n_a'/\omega_a n_s'. \tag{10a}$$

Analogously, the power at the Stokes frequency is

$$P_{\omega\Omega}(k_s) = A \cos\theta_s I_s \frac{n_s'}{\omega_s}\left(\frac{\omega_0}{n_0'}|K_{s0}|^2 + \frac{\omega_a}{2n_a'}|K_{sa}|^2\right). \tag{10b}$$

The first term in (10b), which determines the power of parametric luminescence, coincides with the formula obtained by Kleinman[9] (and in the case of $\alpha_{0,s} = 0$ it coincides with the result of[3]), provided $|E_1|^2$ is expressed in terms of the pump power ($P_1 = e n_1 \cos^2\alpha_1 A |E_1|^2/2\pi$) and it is recognized that we are using double the defined value of $\chi^{(2)}$.

In experiment one usually measures integral power (integrated over the area of one of the resonance curves)[10], for example the light intensity

$$P_\Omega^{(i)} = \int P_{\omega\Omega}\, d\omega \equiv P_{\omega\Omega}(\Delta_i = 0)\, \delta\omega^{(i)}, \tag{11}$$

where $\delta\omega$ is the effective frequency band. For example, for anti-Stokes resonance at $\Delta = 0 (\Delta_a \neq 0)$ we have

$$\delta\omega = \pi l^{-1} |\partial\Delta / \partial\omega_a|^{-1} = \pi l^{-1} \cos\theta_s |u_s^{-1} - u_a^{-1} \cos\widehat{u_s k_a}/\cos\alpha_a|^{-1}. \tag{12}$$

As will be shown in the next section, formulas (10) and (12) are valid separately only if the cross section of the pump beam A is sufficiently large, and at the same time (11) remains in force in many cases of practical importance for arbitrary values of A.

3. INFLUENCE OF BOUNDEDNESS OF THE CROSS SECTION OF THE PUMP BEAM

According to (10)–(12), the nonlinear scattering power is $P_\Omega \sim P_1^2 l/A$, and to increase it it is necessary to focus the pump beam. This, however, decreases the length of the coherent interaction and simultaneously increases the width of the resonance (for waves with nonzero scattering angles θ_i). Let us examine the joint influence of these factors, using as an example a Gaussian pump beam with a plane phase front (i.e., under the condition $k_1 w^2 \gg l$):

$$E_1(r) = E_1 f(r), \quad f(r) = \exp\{ik_1 z - [(x - \alpha_1 z)^2 + y^2] / w^2\}. \tag{13}$$

A. We consider first scattering by $\chi^{(3)}$, described by a perturbation energy

$$H' = -{}^1/_2 \int dk\, dk' \chi^{(3)}_{kk'} E_1^2 E_k^+ E_{k'}^+ \exp\{i(\omega + \omega' - 2\omega_1)t\} f_2(k + k') + \text{h.c.},$$

$$f_m(k) \equiv \int_{-l}^{l} dz \int_{-\infty}^{\infty} dx\, dy \exp\{-ikr\} f^m(r) \tag{14}$$

$$= m^{-1}\pi w^2 \exp\{-(k_x^2 + k_y^2) w^2/4m\} \int_{-l}^{l} dz \exp\{iz(mk_1 - k_z - \alpha_1 k_x)\}.$$

The slowly varying operators E_k^+ are connected here with the Heisenberg-representation creation operators $a_k^+(t)$ by the relation $E_k^+ e^{i\omega t} = c_k^* a_k^+(t)$, so that $[E_k, E_{k'}^+] = |c_k|^2 \delta(k - k')$ and the Heisenberg equations take the form

$$dE_k/dt = i\hbar^{-1} |c_k|^2 E_1^2 \int dk' E_{k'}^+ \chi^{(3)}_{kk'} e^{it(\omega + \omega' - 2\omega_1)} f_2(k + k'). \tag{15}$$

We fix $k = k_a$; then, since f_2 has a sharp maximum at $k' = 2k_1 - k_a \approx k_s$, it follows that $\omega' \approx \omega_s + u_s(k' - k_s)$, so that in first approximation we have

$$E_a^{(1)} = E_a^{(0)} + 2\pi i\hbar^{-1} |c_a|^2 E_1^2 \chi^{(3)}_{as} \int dk' E_{k'}^+ f_2(k + k') \delta(u_s(k' - k_s)). \tag{16}$$

Let the initial state at t_0 be the vacuum state; then

$$\langle E_a^{(1)+} E_a^{(1)}\rangle = 2\pi (t - t_0) |c_a^2 c_s \chi^{(3)}_{as} E_1^2/\hbar|^2 \times \int dk' |f_2(k_a + k')|^2 \delta(u_s(k' - k_s)). \tag{17}$$

The energy in the "mode" k is equal to $\langle H_k\rangle = \hbar\omega\langle E_k^+ E_k\rangle/|c_k|^2$, and the radiated power is $P_k = d\langle H_k\rangle/dt$, so that

$$P_{\omega\Omega}(k_a) = {}^1/_2 A \cos\theta_a I_a b_a b_s |\chi^{(3)}_{as} E_1^2|^2 g_a(\Delta), \tag{18}$$

where the line shape is determined by the function

$$g_a(\Delta) = (u_{sz}/2\pi^2 A) \int dk' |f_2(k_a + k')|^2 \delta(u_s(k' - k_s))$$
$$= \int_{-l}^{l} dz\, dz' \exp\left[i\Delta(z - z') - \left(\frac{z - z'}{w_s}\right)^2\right], \quad w_s$$
$$\equiv \frac{w^2}{(u_{sx}/u_{sz} - \alpha_1)^2 + (u_{sy}/u_{sz})^2}. \tag{19}$$

We note that when we make the substitutions w^2

$\to 2w^2$ and $\Delta \to \Delta_s$, Eq. (19) describes the line shape of linear scattering at frequency ω_0, and therefore the subsequently discussed properties of the function $g(\Delta)$ can be readily transferred to this case (see[3,9]). At $\Delta = 0$, the maximum value of $g(\Delta)$ takes the form

$$g_a(0)=4l^2\left[\frac{\sqrt{\pi}\Phi(\xi)}{\xi}-\frac{1-e^{-\xi^2}}{\xi^2}\right]=4l^2\left(1-\frac{\xi^2}{6}+\ldots\right), \quad (20)$$

where $\Phi(\xi)$ is the probability integral and $\xi = 2l/w_s$. When $\xi \gg 1$ we have $g_a(0) = 2lw_s\sqrt{\pi}$, i.e., the at large scattering angles of the additional wave coherent interaction length is equal to $w_s\sqrt{\pi}$, which is the dimension of the scattering volume along the direction of this wave. From (20) follows the condition for applicability of the model of the plane pump wave to the calculation of $P_{\omega\Omega}$ in the case of scattering by $\chi^{(3)}$ namely $w_s \gg l$ (at $\alpha_1 = 0$ this condition takes the form $\tan\theta_s \ll w/l \sim \sqrt{A/l}$).²⁾ It is important, however, that the integral of $g(\Delta)$ does not depend on w:

$$\int g_a(\Delta)\,d\omega_a = 4\pi l\,|\partial\Delta/\partial\omega_a|^{-1} = 4l^2\delta\omega. \quad (21)$$

Thus, we actually have $P_\Omega \sim A^{-1}$ in this case. We note that the expressions that follow for P_Ω from (18) and (21) coincide with those following from (4c), (10a), and (11). The ratio of (21) to (20) determines the effective line width; in particular, at $\xi \gg 1$ it is equal to $\xi\delta\omega/\sqrt{\pi}$.

B. We now turn to scattering at the anti-Stokes frequency via the difference wave, which is described by the perturbation

$$H' = -\int dk\,dk' E_1 E_k^{+} e^{i(\omega-\omega_1)t}\left[{}^1/_2\chi^{(2)}_{kk'} E_{k'}^{+} e^{i\omega' t} f_1(k+k') + \chi^{(2)}_{kk'} E_{k'} e^{-i\omega' t} f_1(k-k')\right] + \text{h.c.}, \quad (22)$$

The first term yields we omit ($E_{k'}(0)$, since we are interested in the vacuum initial state)

$$E_{k'}^{(1)} = i\hbar^{-1}|c_0|^2\chi^{(2)}_{0s}E_1\int dk'' E_{k''}^{(0)+}\frac{f_1(k'+k'')\exp\{it(\omega'+\omega''-\omega_1)\}}{i(\omega'+\omega''-\omega_1)+\sigma}, \quad (23)$$

where $k' \approx k_0$, $k'' \approx k_s$, and we introduced a convergence parameter $\sigma = +0$. Substituting (23) in the second term of (22), we get

$$E_a^{(2)} = -2\pi|c_a c_0 E_1 \hbar^{-1}|^2\chi^{(2)}_{a0}\chi^{(2)}_{0s} \times \int dk'\,dk'' E_{k''}^{(0)+}\frac{f_1(k_a-k')f_1(k'+k'')}{i(\omega'+\omega''-\omega_1)+\sigma}\delta(\omega_a+\omega''-2\omega_1). \quad (24)$$

Just as in the derivation of (18), we obtain

$$P_{\omega\Omega}(k_a) = A\cos\theta_a I_a|\beta_{a0}\beta_{0s}|^2(\omega_s n_a'/\omega_a n_s')g_a(\Delta_a,\Delta_s),$$

$$g_a(\Delta_a,\Delta_s) = \frac{u_{az}u_{0z}}{(2\pi)^8 A}\int dk''\delta(u_s(k''-k_s))\left|\int dk'\frac{f_1(k_a-k')f_1(k'+k'')}{iu_0(k'-k_0)+\sigma}\right|^2 \quad (25)$$

$$= \frac{A}{4\pi^2}\int dq\exp\left\{-\frac{w^2q^2}{4}\right\}\left|\int_{-l}^{l}dz\int_{-l}^{z}dz'\exp\left\{iz\left(\Delta_a+\frac{p_0q}{2}\right) + iz'\left(\Delta_s-\frac{p_0q}{2}+p_sq\right)-\left(\frac{z-z'}{w_0}\right)^2\right\}\right|^2, \quad (26)$$

where q is a two-dimensional vector, p_i are two-dimensional vectors with components u_{ix}/u_{iz} and u_{iy}/u_{iz}, and $w_0 \equiv w\sqrt{2}/\tan\theta_0$ (in the derivation of the last expression for g_a we have assumed for simplicity that $\alpha_1 = 0$).

²⁾In [3] we wrote $l/\sqrt{A}$ in error. In addition, formula (2) of that reference contains an extra minus sign.

Let us examine the resonance g_a at $\Delta = \Delta_a + \Delta_s \approx 0$. Let $|\Delta_a w_0 + 1| \gg l/w_0$; then

$$\int g_a(\Delta_a,\Delta_s)\,d\omega_a = \frac{4\pi l}{\Delta_a^2}\left|\frac{\partial\Delta}{\partial\omega_a}\right|^{-1} \quad (27a)$$

and we get for P_Ω a value that coincides (apart from the factor 2 connected with the non-additivity of P_Ω in this case over the cross section of the pump beam) with the results of the preceding section (see formulas (10b), (11), and (12)). If the inverse inequality holds (small beam radius w or large angle θ_0), then

$$\int g_a(\Delta_a,\Delta_s)\,d\omega_a = \pi^2 w_0^2 l\,|\partial\Delta/\partial\omega_a|^{-1}, \quad (27b)$$

so that P_Ω does not depend on A. Results similar to (27) are obtained also for the areas of the other resonances ($\Delta_a \approx 0$ or $\Delta_s \approx 0$).

C. Finally, in the case of scattering via a harmonic (at $\alpha_1 = \alpha_2 = 0$), the line shape is described by the function (26) with the substitutions $\Delta_s \to \Delta_2$, $\Delta_a \to \Delta'$, and $\theta_0 \to 0$, so that we get for P_Ω expressions that are likewise independent of A at sufficiently small A.

The foregoing calculations can be extended without difficulty to the case of "two-beam" experiments[4], when $k_1 \neq k_1'$. At large angles φ between k_1 and k_1', second-order scattering by $\chi^{(2)}$ does not play any role (since the transverse momentum should be conserved with exponential accuracy in the production of the virtual waves k_0 and k_2), and the interpretation given in[4] for the experiment (in which $\varphi = 90°$) is correct. At the same time, the estimate given at the end of Sec. 1 shows that in many non-centrally-symmetrical crystals, at $\varphi \sim 0$, the intensity of the SLL due to $\chi^{(2)}$ should greatly exceed the scattering due to the cubic nonlinearity.

In conclusion we note that the foregoing calculation method, which uses Heisenberg's equations for the amplitude operators, has apparently certain advantages (e.g., compactness and parallelism with the classical methods of nonlinear optics) over the scattering-matrix method[6].

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