In the conditions of the Earth's magnetosphere $d \sim (2$ to $3) \times 10^7$ cm, which agrees with the observational results.

The authors are grateful to R. Z. Sagdeev, A. A. Galeev, F. Koroniti, and A. B. Mikhaĭlovskiĭ for valuable advice and useful criticism. One of the authors (V. I. Shevchenko) thanks B. Buti and other participants of the Soviet-Indian Seminar on plasma astrophysics for fruitful discussions of the paper.

[1] B. Coppi, G. Laval, and R. Pellat, Phys. Rev. Lett. **16**, 1208 (1966).

[2] B. B. Kadomtsev, Fiz. Plazmy **1**, 710 (1975) [Sov. J. Plasma Phys. **1**, 389 (1975)].

[3] A. A. Galeev and L. M. Zeleny in Theoretical and Computational Plasma Physics, IAEA, Vienna, 1978, p. 93.

[4] A. B. Mikhaĭlovskiĭ and A. V. Timofeev, Zh. Eksp. Teor. Fiz. **44**, 919 (1963) [Sov. Phys. JETP **17**, 626 (1963)].

[5] I. P. Gudblud, A. I. Pyatak, and V. L. Sizonenko, Zh. Eksp. Teor. Fiz. **64**, 2084 (1973) [Sov. Phys. JETP **37**, 1051 (1973)].

[6] J. D. Huba, N. T. Gladd, and K. Papadopoulos, Geophys. Res. Lett. **4**, 125 (1977).

[7] D. A. Gurnett, R. R. Anderson, B. T. Truruntani, E. J. Smith, G. Paschman, G. Haerendel, S. J. Bame, and C. T. Russell, Preprint Univ. Iowa, 79-7, 1979.

[8] E. N. Parker, J. Geophys. Res. **62**, 509 (1957); P. A. Sweet, Proc. IAU Symp. No. 7, 1958, p. 123.

[9] C. T. Russell and R. C. Elphic, Space Sc. Rev. **22**, 681 (1978).

[10] A. B. Mikhaĭlovskiĭ, Teoriya plazmennykh neustoĭchivosteĭ (Theory of plasma instabilities) Vol. 2, Atomizdat, 1976 [English translation published by Plenum Press, New York].

[11] V. D. Shafranov, Voprosy Teor. Plazmy **3**, 3 (1963) [Rev. Plasma Phys. **3**, 1 (1967)].

[12] B. B. Kadomtsev, A. B. Mikhaĭlovskiĭ, and A. V. Timofeev, Zh. Eksp. Teor. Fiz. **47**, 2264 (1964) [Sov. Phys. JETP **20**, 1517 (1965)].

[13] A. B. Mikhaĭlovskiĭ, Nucl. Fusion **6**, 125 (1965).

[14] A. A. Galeev and R. Z. Sagdeev, Voprosy Teor. Plazmy **7**, 3 (1973) [Rev. Plasma Phys. **7**, 1 (1979)].

[15] V. E. Zakharov, Zh. Eksp. Teor. Fiz. **62**, 1745 (1972) [Sov. Phys. JETP **35**, 908 (1972)].

Translated by D. ter Haar

Photon coalescence in a dispersive medium when scattered by impurity centers without a change in its state

G. V. Vikhnina and S. I. Pekar

Semiconductor Institute, USSR Academy of Sciences

(Submitted 18 March 1979)
Zh. Eksp. Teor. Fiz. **78**, 600–608 (February 1980)

The probability of scattering multimode light by impurity centers, with absorption of an arbitrary number of photons and production of a single photon (photon-coalescence probability) is calculated for an isotropic medium with frequency dispersion but not with spatial dispersion of the dielectric constant in the transparency region. It is shown that the probability depends not only on the total number of centers but also on their distribution in space. The following cases are considered; 1) uniform concentration of the centers, 2) specified coordinate dependence of the concentration, 3) center concentration randomly fluctuating in space. The previously derived equations for the coalescence probability of two or three photons (uniform concentration of the scattering centers) differ from those obtained in the present paper, which makes use of consistent quantization of the field in a dispersive medium [S. I. Pekar, Sov. Phys. JETP **41**, 430 (1975)].

PACS numbers: 42.10.Ke, 78.50. – w

Multiphoton processes in a dispersive medium must be treated by quantizing the electromagnetic field in the medium. This quantization was considered in a number of papers on the basis of crystal microtheory.[1-3] In these papers they used not the complete system of the crystal basis functions, but only its excitonic excitations. The results of these papers are therefore valid only in a narrow spectral region near the exciton resonance. When photons coalesce, however, the frequency of the light wave changes severalfold, and to analyze the coalescence we must be able to quantize the field in a wide spectral region. This is why the results of Refs. 1–3 are not used in the theory of photon coalescence, and in particular in the present paper.

The coalescence of photons (the generation of multiple harmonics) on molecules of the host substance was previously considered a number of times (see, e.g., Refs. 4–8). In these studies the field was quantized in a wide spectral interval, the light waves were considered macroscopically, and the dielectric constant of the crystal $\varepsilon(\omega)$ was introduced phenomenologically. The field quantization, however, was not consistent: the electromagnetic-field energy operator was postulated in the form

$$\sum_k \hbar\omega_k a_k^+ a_k,$$

but the form of the operators a_k^+ and a_k was not derived and their commutation law was not proved, but was written in analogy with a nondispersive medium. Different connection of the operators a_k^+ and a_k with the vector potential of the field were assumed by different authors. The solution of the same problem led therefore to different results (cf., e.g., Refs. 5 and 7).

A consistent method of field quantization in a dispersive medium, applicable in a wide spectral interval and with $\varepsilon(\omega)$ introduced phenomenologically, was proposed in 1975.[9] In the absence of dissipation, i.e., when the electromagnetic field lies in the transparency region of the crystal, canonically conjugate quantities with a known commutation law were separated in the expression for the field energy, and the photons in a dispersive medium were obtained in consistent fashion. It is this quantization method which will be used here.

In a preceding paper[10] we calculated on the basis of this method[9] the probabilities of multiphoton processes occurring at local centers and accompanied by a change in the states of the centers. In a number of cases the expressions obtained for the probabilities turned out to be different from those used earlier. In the present paper we consider multiphoton processes in which the centers remain in the initial state, i.e., there is no energy exchange between the radiation and the center.

The unperturbed free-photon energy operator W is expressed with account taken of the dielectric constant $\varepsilon(\omega)$ [i.e., under the assumption that the phonon dispersion law is of the form $k=\omega\varepsilon^{1/2}(\omega)/c$, and not $k=\omega/c$], and contains consequently in part the interaction W' of the light with the molecules of the host substance. In the treatment of the scattering of light by the host molecules, the perturbation of the operator W is therefore not W' but some difficult-to-separate part of W'. This circumstance has escaped attention of most workers dealing with the interaction of light with the crystal host substance, who took the perturbation to be W'. Not seeing at the moment how to solve this problem correctly, we consider the interaction of the light with the impurity centers whose contribution to the polarization is not yet included in $\varepsilon(\omega)$. In that case we do not encounter the aforementioned difficulty.

We consider on the basis of Ref. 9 the processes of absorption of several photons by impurity centers, with generation of one photon with the summary frequency. The medium is assumed to be transparent, isotropic, having frequency but not spatial dispersion of $\varepsilon(\omega)$ and a magnetic permeability $\mu=1$, bounded by the parallel planes $z=0$ and $z=L$, with the dimensions of the cyclicity L_x and L_y in the x and y directions much larger than L. The operator of the photon energy in the medium is of the form[9]

$$W=\sum_{\mathbf{k}}\sum_{\nu=1}^{2}\sum_{l=0}^{s_k}\hbar\omega_{l\mathbf{k}}a_{l\mathbf{k}\nu}^{+}a_{l\mathbf{k}\nu}, \tag{1}$$

where $\mathbf{k}$ are wave vectors whose quasidiscrete spectrum is determined by the cyclicity conditions (the surface effects are not considered) and l numbers the roots of the dispersion equation

$$\varepsilon(\omega)\omega^2/c^2=k^2 \tag{2}$$

at fixed k. We denote these roots by $\omega_{l\mathbf{k}}^2$, where $l=0,1,\ldots,s_k$. The polarizations of the mode are numbered by the index $\nu=1,\ 2$, and $a_{l\mathbf{k}\nu}^+$ and $a_{l\mathbf{k}\nu}$ are the Bose creation and annihilation operators of a photon of mode $l,\mathbf{k},\nu$.

The operator of the vector potential of the field is

$$\mathbf{A}(\mathbf{r})=\sum_{l\mathbf{k}\nu}\mathbf{e}_{\mathbf{k}\nu}e^{i\mathbf{k}\mathbf{r}}(\hbar/Vm(\omega_{l\mathbf{k}})\omega_{l\mathbf{k}})^{1/2}(a_{l\mathbf{k}\nu}+a_{l,-\mathbf{k},\nu}^{+}), \tag{3}$$

where $\mathbf{e}_{\mathbf{k}\nu}$ is a unit vector of the polarization of the mode $l\mathbf{k}\nu$, and

$$m(\omega_{l\mathbf{k}})\equiv m_{l\mathbf{k}}=\frac{1}{2\pi c^2}\frac{\partial}{\partial\omega_{l\mathbf{k}}^2}[\omega_{l\mathbf{k}}^2\varepsilon(\omega_{l\mathbf{k}})]. \tag{4}$$

The energy operator of the interaction of one center with the electromagnetic field is of the form

$$U=-c^{-1}\mathbf{d}\mathbf{A}(\mathbf{r}); \tag{5}$$

here $\mathbf{d}$ is the operator of the dipole moment of the center, and $\mathbf{A}(\mathbf{r})$ is the operator of the vector potential of the field at the location $\mathbf{r}$ of the center. The number of values of the vector $\mathbf{k}$ in the frequency interval $d\omega$ and inside the solid angle $d\Omega$ is

$$\rho(\omega)\,d\Omega d\omega=\frac{V}{(2\pi)^3}dk_xdk_ydk_z=\frac{V\varepsilon^{1/2}(\omega)}{4\pi^2c}m(\omega)\omega^2d\omega d\Omega. \tag{6}$$

We present below expressions for the probabilities, per unit time, of the processes of interest to us. It is assumed that at the initial instant of time there are specified the numbers of the photons whose phases are perfectly random, and the usual perturbation theory[11] is used, in analogy with the procedure used by Hopfield.[1]

We use the following notation. The symbols ω_1^+, $l_2\omega_2^-\ldots$ denote that the process in question consists of production of one photon of frequency ω_1 and polarization e_1 and absorption of l_2 photons of frequency ω_2 and polarization e_2, etc. The presence of the factor $d\omega_1$, $d\Omega_1$, or d^3k_1 means that the photon ω_1 is produced in the frequency interval $d\omega_1$ and inside the solid angle $d\Omega_1$, or else in the element d^3k_1 of the wave-vector space. The radiation has the exact frequencies $\omega_2,\omega_3,\ldots,\omega_\lambda$. The state of the entire system is designated by the aggregate of indices $i,\ldots,n_{l\mathbf{k}\nu}\ldots$, where i is the set of quantum numbers characterizing the state of the mechanical subsystem, and $n_{l\mathbf{k}\nu}$ is the number of photons in the mode $l\mathbf{k}\nu$.

The probability of emission of a photon of frequency ω_1 upon coalescence of l_2 photons ω_2, l_3 photons $\omega_3,\ldots,l_\lambda$ photons ω_λ is equal to

$$\mathscr{P}(\omega_1^+,l_2\omega_2^-,\ldots,l_\lambda\omega_\lambda^-)d^3k_1=|\mathfrak{M}(\omega_1^+,l_2\omega_2^-,\ldots,l_\lambda\omega_\lambda^-)|^2\times\prod_{s=2}^{\lambda}\left(\frac{n_s}{m_s\omega_s}\right)^{l_s}\frac{\hbar^{F-2}J}{4\pi^2V^{F-1}c^{2F}m_1\omega_1}\delta(\omega_1-l_2\omega_2-\ldots-l_\lambda\omega_\lambda)d^3k_1, \tag{7}$$

where $F=1+l_2+\ldots+l_\lambda$ is the total number of photons participating in the process, the expression $\mathfrak{M}(\ldots)$ is given in the Appendix, and

$$J=\Big|\sum_j\exp(i\Delta\mathbf{k}\mathbf{r}_j)\Big|^2=\sum_{jj'}\exp(i\Delta\mathbf{k}(\mathbf{r}_j-\mathbf{r}_{j'})); \tag{8}$$

here j is the number of the impurity center, $\mathbf{r}_j$ is its coordinate, and $\Delta\mathbf{k}$ is the vector sum of the wave vectors of the absorbed photons minus the wave vector of the produced one. For example, in the case of generation of a photon ω_1 at the doubled frequency $\omega_1 = 2\omega_2$ we have $\Delta\mathbf{k} = 2\mathbf{k}_2 - \mathbf{k}_1$. The summation in (8) is over all the impurity centers of the crystal.

At $\lambda = 2$ and $l_2 = 2$ Eq. (7) describes the generation of photon ω_1 at double the frequency of the incident monochromatic light beam:

$$\mathscr{P}(\omega_1^+, 2\omega_2^-)d^3k_1 = |\mathfrak{M}(\omega_1^+, 2\omega_2^-)|^2\left(\frac{n_2}{m_2\omega_2}\right)^2\frac{\hbar}{4\pi^2V^2c^6m_1\omega_1}J\delta(\omega_1 - 2\omega_2)d^3k_1. \tag{9}$$

At $\lambda = 2$ and $l_3 = 3$ we have generation at triple the frequency of the incident light

$$\mathscr{P}(\omega_1^+, 3\omega_2^-)d^3k_1 = |\mathfrak{M}(\omega_1^+, 3\omega_2^-)|^2 \times\left(\frac{n_2}{m_2\omega_2}\right)^3\frac{\hbar^2}{4\pi^2V^3c^8m_1\omega_1}J\delta(\omega_1 - 3\omega_2)d^3k_1. \tag{10}$$

At $\lambda = 3$ and $l_2 = l_3 = 1$ we have emission of the photon ω_1 upon coalescence of the photons ω_2 and ω_3:

$$\mathscr{P}(\omega_1^+, \omega_2^-, \omega_3^-)d^3k_1 = |\mathfrak{M}(\omega_1^+, \omega_2^-, \omega_3^-)|^2 \times\frac{n_2n_3}{m_2\omega_2m_3\omega_3}\frac{\hbar}{4\pi^2V^2c^6m_1\omega_1}J\delta(\omega_1 - \omega_2 - \omega_3)d^3k_1 \text{ etc.} \tag{11}$$

We calculate on the value of J for various possible distributions of the centers in space.

a. *Equiprobable distribution of the centers in space.* Inasmuch as arbitrary mutual dispositions of the centers can be encountered in a large volume, the quantity (8) must be averaged over all the possible equiprobable dispositions of the centers over the cells. We carry out the averaging, as before,[12] under the assumption that: 1) not more than one center can be localized in a unit cell of the crystal; 2) the center can occupy in the cell only one position; 3) in all other respects all the distribution of the centers over the cells are equally probable. Expression (8) must be summed over all the integer lattice vectors $\mathbf{r}_j$ and $\mathbf{r}_{j'}$, with account taken of the identity of the centers, and must be divided by the number of terms, which is equal to $(V/\Delta)!N!(V/\Delta - N)!$; here Δ is the volume of the unit cell and N is the number of centers in the crystal. Recognizing that the number of terms with $\mathbf{r}_j \neq \mathbf{r}_{j'}$ in (8) is $N(N-1)$, and that the number of the distributions of all the centers over the cells, wherein the cells occupied are those having coordinates $\mathbf{r}_j$ and $\mathbf{r}_{j'}$ is equal to

$$\frac{2(V/\Delta - 2)!}{N!(V/\Delta - N)!},$$

and the summation over all the possible locations of the two particles reduces to

$$\frac{1}{2}\sum_{\mathbf{r}_j \neq \mathbf{r}_{j'}},$$

we obtain

$$J = J_1 + J_2, \tag{12}$$

$$J_1 = \frac{N^2L}{V}4\pi^2\delta(\Delta k_x)\delta(\Delta k_y)g(\Delta k_z), \tag{13a}$$

$$J_2 = N[1 - N\Delta/V], \tag{13b}$$

where

$$g(\Delta k_z) = \left(\sin^2\frac{\Delta k_zL}{2}\right)\Big/\left(\frac{\Delta k_zL}{2}\right)^2$$

and the fact is used that the dimensions of the medium along the axes x and y greatly exceed L.

The probability (7) is now represented by a sum of two terms:

$$\mathscr{P}d^3k_1 = \mathscr{P}_1d^3k_1 + \mathscr{P}_2d^3k_1$$

corresponding to the terms in (12). For simplicity we assume that the incident primary radiation propagates along the z axis, i.e., $\mathbf{k}_2 \| \mathbf{k}_3 \ldots \| \mathbf{k}_\lambda \| z$. It follows then from (13a) that $\mathscr{P}_1$ determines the probability of the production of a photon with $\mathbf{k}_1$ strictly parallel to z. The integral probability of this process is equal to

$$\begin{aligned} Q_1(l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-) &= \int\mathscr{P}_1(\omega_1^+, l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)d^3k_1 \\ &= |\mathfrak{M}(\omega_1^+, l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)|^2\prod_{s=2}^{\lambda}\left(\frac{n_s}{m_s\omega_s}\right)^{l_s} \\ &\quad\times\frac{2\pi LN^2\hbar^{F-2}}{\varepsilon_1^{1/2}\omega_1V^Fc^{2F-1}}g(\Delta k_z), \\ \omega_1 = l_2\omega_2 + \ldots + l_\lambda\omega_\lambda,& \quad \mathbf{k}_1 \| \mathbf{k}_2 \| \ldots \| \mathbf{k}_\lambda \| z, \end{aligned} \tag{14}$$

Q_1 as a sharp peak subject to the synchronism condition $\Delta k_z = 0$ or

$$\sum_{s=2}^{\lambda} l_s\omega_s\varepsilon_s^{1/2} = \omega_1\varepsilon_1^{1/2}$$

and tends rapidly to zero with increasing Δk_z. The term $\mathscr{P}_2$ determines the probability of production of the photon ω_1 with arbitrary direction $\mathbf{k}_1$. The probability of production of a photon with a wave vector in the solid-angle element $d\Omega_1$ is equal to

$$\begin{aligned} Q_2(l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)d\Omega_1 &= \int\mathscr{P}_2(\omega_1^+, l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)\frac{2\pi\varepsilon_1^{1/2}}{c}m_1\omega_1^2d\omega_1d\omega_1 \\ &= |\mathfrak{M}(\omega_1^+, l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)|^2\frac{\hbar^{F-2}\varepsilon_1^{1/2}\omega_1}{2\pi V^{F-1}c^{2F+1}}N\left(1 - \frac{N\Delta}{V}\right)d\Omega_1, \\ \omega_1 &= l_2\omega_2 + \ldots + l_\lambda\omega_\lambda. \end{aligned} \tag{15}$$

Unfortunately, the literature has dealt with coalescence of photons not with impurities, but only with molecules of the host material, and this makes it difficult to compare our results with the published ones. A comparison is nevertheless possible if: we ignore the remark made at the start of the article, i.e., there are no grounds for taking the perturbation to be U also when processes in the host substance are considered; b) the connection (3) between the creation and annihilation operators is replaced by a connection used by others, i.e., the coefficient (4) is replaced by the quantity $\varepsilon(\omega)/2\pi c^2$; c) the number N of the impurities is replaced by the number V/Δ of the molecules in the host substance. As a result, our formulas yield $J_2 = 0, Q_2 = 0$, and J_1 and Q_1 are given by expressions cited by others[4-8] for processes on the host molecules.[1)]

In the case of scattering by impurities, in contrast to the host substance, the term Q_2 (15) makes possible the coalescence of photons in those media in which the synchronism condition $\Delta\mathbf{k} = 0$ is satisfied. The ratio of the probability of production of a photon with $\mathbf{k}_1$ strictly parallel to z to the probability of production of a photon with arbitrary direction of $\mathbf{k}_1$ is of the form

$$\eta = Q_1(l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-) \Big/ \int Q_2(l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)\, d\Omega_1$$
$$= \frac{4\pi^2c^2L}{\varepsilon_1\omega_1^2}\frac{N}{V} g(\Delta k_z) |\mathfrak{M}(\omega_1^+, l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)|^2_{\mathbf{k}_1\|z}$$
$$\times\left(\int |\mathfrak{M}(\omega_1^+, l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)|^2\, d\Omega_1\right)^{-1}. \tag{16}$$

Let us estimate this ratio, assuming approximately that the last factor in the right-hand side of (16) equals 1/4. For

$$\omega_1 = 2\cdot 10^{15}\ \text{sec}^{-1},\quad N/V \sim 10^{16}\,\text{cm}^{-3},\quad \varepsilon_1 \sim 10,$$

we obtain

$$\eta \sim 10^6 Lg(\Delta k_z). \tag{17}$$

if L is given in centimeters.

According to Ref. 6, in the case of generation at the tripled frequency g assumes for various crystals values from 1 to 10^{-7}. It is seen from this that η can be considerably larger as well as considerably smaller than unity.

b. *Nonequilibrium distribution of the impurities*. We consider now the case of a nonuniform distribution of the impurities in the crystal. For example, according to a number of studies[13] the distribution of the impurities along the growing crystal can be given in real conditions not by a monotonic curve but by an oscillating one. The calculation of J in the case of a spatial dependence of the impurity concentration $\nu(\mathbf{r})$ and subject to the inequality $r_0 \ll \Delta k^{-1}, r_0 \ll L$ (r_0 is the average distance between the impurities) leads to the expression

$$J = V|\nu_{\Delta\mathbf{k}}|^2, \tag{18}$$

where $\Delta\mathbf{k}$ has already been defined, and

$$\nu_{\Delta\mathbf{k}} = \frac{1}{V}\int_V d\mathbf{r}\,\nu(\mathbf{r})\,e^{-i\Delta\mathbf{k}\mathbf{r}}. \tag{19}$$

Formulas (18) and (19) should be used if the function $\nu(\mathbf{r})$ is specified.

We consider now the case when the concentration fluctuates randomly in space. We put

$$\nu(\mathbf{r}) = \nu_0 + \Delta\nu(\mathbf{r}),\quad \nu_0 = \text{const} \tag{20}$$

and, regarding $\Delta\nu(\mathbf{r})$ as a random quantity, we introduce the correlation function

$$F(\mathbf{R}) = \frac{1}{V}\int d\mathbf{r}\,\Delta\nu(\mathbf{r})\,\Delta\nu(\mathbf{r}+\mathbf{R}). \tag{21}$$

Specifying $F(\mathbf{R})$ in Gaussian form

$$F(\mathbf{R}) = F_0 e^{-\beta R^2} \tag{22}$$

and using (18)–(21), we calculate J:

$$J = J_1 + J_3, \tag{23}$$

where J_1 is given by (13a), Q by (14), and

$$J_3 = |\Delta\nu_{\Delta\mathbf{k}}|^2V^2 = VF_0 \exp\left\{-\frac{(\Delta k_x)^2 + (\Delta k_y)^2}{4\beta}\right\}\frac{\pi}{\beta}\int_{-L}^{L} dz \exp\{-\beta z^2 + i\Delta k_z z\}, \tag{24a}$$

or, if the inequalities $L \gg \beta^{-1/2}, L \gg \Delta k^{-1}$, hold,

$$J_3 = VF_0(\pi/\beta)^{3/2}\exp\{-(\Delta k)^2/4\beta\}. \tag{24b}$$

Substituting J_3 in (7) we obtain, after integrating with respect to the frequency, the probability of production of a photon ω_1 in a solid-angle element $d\Omega_1$:

$$Q_3(l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)\,d\Omega_1 = |\mathfrak{M}(\omega_1^+, l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)|^2$$
$$\times\prod_{s=2}\left(\frac{n_s}{m_s\omega_s}\right)^{l_s}\frac{\hbar^{F-2}\omega_1\varepsilon_1^{1/2}}{2\pi V^{F-2}c^{2F+1}}F_0\left(\frac{\pi}{\beta}\right)^{3/2}\exp\left\{-\frac{(\Delta k)^2}{4\beta}\right\}d\Omega_1, \tag{25}$$
$$\omega_1 = l_2\omega_2 + \ldots + l_\lambda\omega_\lambda.$$

If the number of impurities N is replaced by the number of molecules of the host material V/Δ, then $F_0 = Q_3 = 0$.

If the concentration is homogeneous only along z, e.g., in the crystal growth direction, then

$$F = F_0\exp\{-\beta z^2\},$$
$$J_3' = (2\pi)^2V\delta(\Delta k_x)\delta(\Delta k_y)F_0(\pi/\beta)^{1/2}\exp\{-(\Delta k_z)^2/4\beta\}, \tag{26}$$

i.e., only a photon with $\mathbf{k}_1 \| z$ is produced. The probability of it production per unit time is

$$Q_3'(l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-) = |\mathfrak{M}(\omega_1^+. l_2\omega_2^-, \ldots, l_\lambda\omega_\lambda^-)|^2$$
$$\times\prod_{s=2}^{\lambda}\left(\frac{n_s}{m_s\omega_s}\right)^{l_s}\frac{2\pi h^{F-2}}{V^{F-2}c^{2F-1}\varepsilon_1^{1/2}\omega_1}F_0\left(\frac{\pi}{\beta}\right)^{1/2}\exp\left\{-\frac{(\Delta k)^2}{4\beta}\right\} \tag{27}$$
$$\Delta\mathbf{k}\|z,\quad \omega_1 = l_2\omega_2 + \ldots l_\lambda\omega_\lambda,$$

$$\eta' \equiv \frac{Q_1}{Q_3'} = \frac{\nu_0^2}{F_0}\left(\frac{\beta}{\pi}\right)^{1/2}Lg(\Delta k)\exp\left\{\frac{(\Delta k)^2}{4\beta}\right\}. \tag{28}$$

At $\Delta k^2 \approx 4\beta$ we have

$$\eta' \leqslant 3\frac{\nu_0^2}{F_0}\frac{1}{\Delta kL}. \tag{29}$$

According to Ref. 6, in this case of frequency tripling Δk assumes for various real crystals values from 1 to 10^4 cm^{-1}. Therefore, depending on the value of F_0L, the ratio η' can be both much larger and much smaller than unity.

We note that the perturbing macroscopic field that does not vary smoothly in space, which enters in the Schrödinger equation and in (5) should be chosen to be not $\mathbf{A}(\mathbf{r})$, but the local microscopic field $\tilde{\mathbf{A}}(\mathbf{r}) = \varkappa(\mathbf{r})\mathbf{A}(\mathbf{r})$, where the tensor $\varkappa(\mathbf{r})$ has the same periods of the lattice if the impurity does not force out a molecule of the host substance from its site. The macroscopic field is obtained from the microscopic one by averaging the latter over the volume of the unit cell of the crystal, i.e., the mean value of $\varkappa$ over the cell volume is $\bar{\varkappa} = 1$. It is shown in Ref. 14 that the mean value of $\varkappa$ along any straight line passing through the cell parallel to the edge of the cube is also equal to unity. It can be shown that the mean value of $\varkappa$ along any curved line that joins equivalent points of adjacent cells is also equal to unity. Therefore if the radius of the center is much larger than the lattice constant, then $\varkappa$ can be replaced by unity, i.e., ignored. If the radius of the cell is small, then $\varkappa$ must be introduced, and this leads to a some change in the numerical values of the matrix elements D_{ij} in Eq. (31).

APPENDIX

We use the following notation: λ is the number of modes (phonon species) participating in the process; s is the number of the mode ($1 \leqslant s \leqslant \lambda$); l_s is the number of vanishing or produced photons of mode s;

$$F=\sum_{s=1}^{\lambda} l_s$$

is the total number of photons of all modes participating in the process; q is the number of photon and numbers in succession first the photons of mode $s=1$, next of mode $s=2$, and so on $(1\leq q\leq F)$. For example, in $\mathfrak{M}(\omega_1^+, 2\omega_2^-, 5\omega_3^-)$ we have $\lambda=3$, $l_1=1$, $l_2=2$, $l_3=5$, and $F=8$; the first photon of the third mode has $q=4$. We note that the q that satisfy the inequality

$$\sum_{s'=1}^{s-1} l_{s'} < q \leq \sum_{s'=1}^{s} l_{s'},$$

number the photons of one mode s.

We put next

$$\omega^{(q)}=\begin{cases}-\omega_1 & q=1\\ \omega_s & q\neq 1\end{cases}, \quad \sum_{s'=1}^{s-1} l_{s'} < q \leq \sum_{s'=1}^{s} l_{s'}. \tag{30}$$

We designate the multidimensional quantum numbers of the mechanical subsystem by $i_0, i_1, \ldots,$ where i_0 are the initial and $i_1, \ldots, i_{F-1}$ are the intermediate subsystems. Then the quantity $\mathfrak{M}(\ldots)$ in (7) and in other equations takes the form

$$\mathfrak{M}(\omega_1^+, l_2\omega_2^-, \ldots, l_s\omega_s^-, \ldots, l_\lambda\omega_\lambda^-)=\hbar^{1-F} \sum_{i_1,\ldots,i_{F-1}} \sum_P P\Big\{ (\dot{\mathbf{D}}_{i_0 i_{F-1}}\mathbf{e}_1) \times\prod_{q=2}^{F} (\dot{\mathbf{D}}_{i_{F-q+1} i_{F-q}}\mathbf{e}_q) \Big/ \Big[\omega_{i_0 i_{F-q+1}} + \sum_{q'=q}^{F} \omega^{(q')}\Big]\Big\}. \tag{31}$$

Here P denotes the permutation of the numbers of two photons from different modes, say q'' and q'''. This interchanges the indices of the polarization vectors, $\mathbf{e}_{q''}\rightleftharpoons\mathbf{e}_{q'''}$, and of the frequencies, $\omega^{(q'')}\rightleftharpoons\omega^{(q''')}$. The sum $\sum_P P$ is taken over all the permutations of the numbers of photons from different modes, but the numbers of the photons of the same mode are not permuted.

For example, for generation of a photon ω_1 at double the frequency of the incident radiation ω_2: $\lambda=2$, $l_2=2$, $F=3$, $q=1, 2, 3$ ($q=2, 3$ number the photons of one mode $s=2$); $\omega^{(1)}=-\omega_1$, $\omega^{(2)}=\omega^{(3)}=\omega_2$, $\mathbf{e}_2=\mathbf{e}_3$. In this case Eq. (31) takes the form

$$\mathfrak{M}(\omega_1^+, 2\omega_2^-)=\hbar^{-2}\sum_{i_1 i_2}\sum_P P\left\{\frac{(\dot{\mathbf{D}}_{i_0 i_2}\mathbf{e}_1)(\dot{\mathbf{D}}_{i_2 i_1}\mathbf{e}_2)(\dot{\mathbf{D}}_{i_1 i_0}\mathbf{e}_3)}{(\omega_{i_0 i_2}+\omega^{(2)}+\omega^{(3)})(\omega_{i_0 i_1}+\omega^{(3)})}\right\}$$

$$=\hbar^{-2}\sum_{i_1 i_2}\left\{\frac{(\dot{\mathbf{D}}_{i_0 i_2}\mathbf{e}_1)(\dot{\mathbf{D}}_{i_2 i_1}\mathbf{e}_2)(\dot{\mathbf{D}}_{i_1 i_0}\mathbf{e}_3)}{(\omega_{i_0 i_2}+\omega^{(2)}+\omega^{(3)})(\omega_{i_0 i_1}+\omega^{(3)})}\right.$$

$$\left.+\frac{(\dot{\mathbf{D}}_{i_0 i_2}\mathbf{e}_2)(\dot{\mathbf{D}}_{i_2 i_1}\mathbf{e}_1)(\dot{\mathbf{D}}_{i_1 i_0}\mathbf{e}_3)}{(\omega_{i_0 i_2}+\omega^{(1)}+\omega^{(3)})(\omega_{i_0 i_1}+\omega^{(3)})}+\frac{(\dot{\mathbf{D}}_{i_0 i_2}\mathbf{e}_3)(\dot{\mathbf{D}}_{i_2 i_1}\mathbf{e}_2)(\dot{\mathbf{D}}_{i_1 i_0}\mathbf{e}_1)}{(\omega_{i_0 i_2}+\omega^{(2)}+\omega^{(1)})(\omega_{i_0 i_1}+\omega^{(1)})}\right\}$$

$$=\hbar^{-2}\sum_{i_1 i_2}\left\{\frac{(\dot{\mathbf{D}}_{i_0 i_2}\mathbf{e}_1)(\dot{\mathbf{D}}_{i_2 i_1}\mathbf{e}_2)(\dot{\mathbf{D}}_{i_1 i_0}\mathbf{e}_2)}{(\omega_{i_0 i_2}+2\omega_2)(\omega_{i_0 i_1}+\omega_2)}+\frac{(\dot{\mathbf{D}}_{i_0 i_2}\mathbf{e}_2)(\dot{\mathbf{D}}_{i_2 i_1}\mathbf{e}_1)(\dot{\mathbf{D}}_{i_1 i_0}\mathbf{e}_2)}{(\omega_{i_0 i_2}-\omega_1+\omega_2)(\omega_{i_0 i_1}+\omega_2)}\right.$$

$$\left.+\frac{(\dot{\mathbf{D}}_{i_0 i_2}\mathbf{e}_2)(\dot{\mathbf{D}}_{i_2 i_1}\mathbf{e}_2)(\dot{\mathbf{D}}_{i_1 i_0}\mathbf{e}_1)}{(\omega_{i_0 i_2}+\omega_2-\omega_1)(\omega_{i_0 i_1}-\omega_1)}\right\}$$

[1] J. J. Hopfield, Phys. Rev. **112**, 1555 (1958).
[2] V. M. Agranovich, Zh. Eksp. Teor. Fiz. **37**, 430 (1959) [Sov. Phys. JETP **10**, 307 (1959)].
[3] W. C. Tait and R. L. Weiher, Phys. Rev. **178**, 1404 (1969).
[4] N. Blombergen, Am. J. Phys. Soc. **35**, 989 (1967).
[5] R. London, Proc. Phys. Soc. London **80**, 952 (1968).
[6] P. D. Maker, Third-Harmonic Generation at Optical Frequencies, Russ. transl. in: Opticheskie kvantovye generatory (Lasers), transl. ed. Bunkin, Mir, 1966, p. 307.
[7] V. M. Faĭn, Fotony i nelineĭnye sredy (Photons and Nonlinear Media), Vol. 1, M., 1972.
[8] R. Loudon, The Quantum Theory of Light, Oxford, 1973.
[9] S. I. Pekar, Zh. Eksp. Teor. Fiz. **68**, 866 (1975) [Sov. Phys. JETP **41**, 430 (1975)].
[10] G. V. Vikhnina and S. I. Pekar, Zh. Eksp. Teor. Fiz. **74**, 2259 (1978) [Sov. Phys. JETP **47**, 1175 (1978)].
[11] W. Heitler, The Quantum Theory of Radiation, Oxford, 1974 (Russ. Transl., IIL, 1956).
[12] S. I. Pekar, Fiz. Tverd. Tela (Leningrad) **8**, 1115 (1966) [Sov. Phys. Solid State **8**, 890 (1966)].
[13] B. M. Turovskiĭ and M. G. Mil'vidskiĭ, Fiz. Tverd. Tela (Leningrad) **3**, 2519 (1961) [Sov. Phys. Solid State **3**, 1834 (1961)]. J. A. M. Dikhoff, Philips Tech. Rev. **25**, 195 (1963/64). V. I. Fistul', Sil'no legirovannye poluprovodniki (Strongly Doped Simiconductors), M., 1967.
[14] V. G. Grachev, M. F. Deĭgen, and S. I. Pekar, Fiz. Tverd. Tela (Leningrad) **15**, 2155 (1973) [Sov. Phys. Solid State **15**, 1431 (1973)].

Translated by J. G. Adashko