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NONLINEAR EFFECTS IN THE ELECTRODYNAMICS OF A TRANSPARENT MEDIUM

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Equations are deduced which describe the nonlinear interactions of electromagnetic waves in a transparent medium. The results are applied to the study of second harmonics arising in the propagation of the wave in the medium. The intensities and polarizations of the harmonics in quartz are determined.

1. Dynamic and kinetic equations describing nonlinear interactions between waves in a plasma, located in a strong magnetic field were obtained in [1]. In the present work, similar equations are obtained for an arbitrary medium.

Recently, the problems of the nonlinear electrodynamics of a medium have attracted interest in connection with the appearance of experimental possibilities for the study of nonlinear effects. For example, Franken et al [2] have reported that the appearance of a second harmonic was observed in the transmission of an intense monochromatic light beam in the optical frequency range through crystalline quartz. This phenomenon is very simply interpreted by means of the equations for wave interactions obtained in the present work.

2. For simplicity, we restrict ourselves to nonmagnetic media ($\mu \approx 1$) and write down Maxwell's equations in the form

$$\operatorname{rot}\mathbf{H} = \frac{1}{c}\frac{\partial \mathbf{D}}{\partial t}, \qquad \mathbf{D} = \mathbf{E} + 4\pi\mathbf{P}, \qquad \frac{\partial \mathbf{H}}{\partial t} = -c\operatorname{rot}\mathbf{E}. \quad (1)^*$$

The nonlinear effects are described by terms of second and higher orders in E, depending on the polarization vector **P** and **E**. By restricting ourselves to terms of second order, we can write (see [3])

$$\mathbf{P} = \mathbf{P}^{(1)} + \mathbf{P}^{(2)}; \quad (2)$$

$$P_\alpha^{(1)} = (4\pi)^{-1}\int_{-\infty}^{t} dt_1 \varphi_{\alpha\beta}^{(1)}(t - t_1) E_\beta(t_1), \quad (3)$$

$$P_\alpha^{(2)} = (4\pi)^{-1}\int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 \varphi_{\alpha\beta\gamma}^{(2)}(t - t_1, t_1 - t_2) E_\beta(t_1) E_\gamma(t_2), \quad (4)$$

$\alpha, \beta, \gamma = x, y, z$. The function $\varphi_{\alpha\beta}^{(1)}(\tau)$ in (3) determines the dielectric tensor

$$\varepsilon_{\alpha\beta} = 1 + \int_0^\infty \varphi_{\alpha\beta}^{(1)}(\tau) e^{i\omega\tau} d\tau, \quad (5)$$

$$\varepsilon_{\alpha\beta}(\omega) = \varepsilon_{\alpha\beta}^*(-\omega). \quad (6)$$

In what follows, only transparent media will be considered. We can then write [4]

$$\varepsilon_{\alpha\beta}(\omega) = \varepsilon_{\beta\alpha}^*(\omega). \quad (7)$$

For simplicity, we shall not consider spatial dispersion, i.e., we shall assume that $\varphi_{\alpha\beta}^{(1)}(t - t_1)$ does not depend on $\mathbf{r} - \mathbf{r}_1$, or $\epsilon_{\alpha\beta}(\omega)$ on **k**. Even in this case, $\varphi_{\alpha\beta\gamma}^{(2)}(t - t_1)$ does not in general depend on $\mathbf{r} - \mathbf{r}_1$, $\mathbf{r}_1 - \mathbf{r}_2$ (for example, for a cold plasma). In what follows, however, the dependence of $\varphi_{\alpha\beta\gamma}^{(2)}$ on the coordinates is not expressed explicitly, inasmuch as it does not play a decisive role and complicates the formulas. All the results obtained are easily generalized to the case of spatial dispersion.

It must be kept in mind that $\varphi_{\alpha\beta\gamma}^{(2)}$ vanishes in certain cases, for example, for bodies possessing a center of symmetry (in the absence of the dependence of $\varphi_{\alpha\beta\gamma}^{(2)}$ on $\mathbf{r}$, $\mathbf{r}_1$, or $\mathbf{r}_2$). In such a case, it is necessary to take the terms of third order in the field into account—$\varphi_{\alpha\beta\gamma\delta}^{(3)}$. We apply the method set forth below in this case; however, we shall not consider it specially since the effects determined below are chiefly connected with the term $\varphi_{\alpha\beta\gamma}^{(2)}$, which is quadratic in the field.

3. We first write down the fundamental relations of the linear approximation in a form convenient for subsequent expansion. The solution of the linearized equations of Maxwell has the form

$$\mathbf{E} = V^{-1/2}\sum_{\mathbf{p}=\mathbf{k},\,\mathbf{k}_-} c_p \mathbf{E}(\mathbf{p}) \exp\{i(\mathbf{pr} - \omega_p t)\},$$

$$\mathbf{H} = V^{-1/2}\sum_{\mathbf{p}=\mathbf{k},\,\mathbf{k}_-} c_p \mathbf{H}(\mathbf{p}) \exp\{i(\mathbf{pr} - \omega_p t)\}, \quad (8)$$

where the $c_\mathbf{p}$ are constants. Here and in what follows, we shall denote the wave vector of the

*rot = curl.

harmonic with positive frequency by **k**, and the wave vector of the harmonic with negative frequency by $\mathbf{k}_-$, so that

$$\mathbf{k}_- = -\mathbf{k}, \qquad \omega_{\mathbf{k}_-} = -\omega_{\mathbf{k}}, \qquad c_{\mathbf{k}_-} = c^*_{\mathbf{k}},$$

$$\mathbf{E}(\mathbf{k}_-) = \mathbf{E}^*(\mathbf{k}), \qquad \mathbf{H}(\mathbf{k}_-) = \mathbf{H}^*(\mathbf{k}). \tag{9}$$

$\mathbf{E}(\mathbf{p})$ and $\mathbf{H}(\mathbf{p})$ satisfy the equations ($\mathbf{p} = \mathbf{k}, \mathbf{k}_-$)

$$[\mathbf{p}\mathbf{H}(\mathbf{p})]_\alpha = -(1/c)\,\varepsilon_{\alpha\beta}E_\beta(\mathbf{p}), \quad \omega_{\mathbf{p}}H(\mathbf{p}) = c\,[\mathbf{p}\mathbf{E}(\mathbf{p})], \tag{10}$$

from which it follows that

$$[p_\alpha p_\beta - p^2\delta_{\alpha\beta} + (\omega_p^2/c^2)\,\varepsilon_{\alpha\beta}(\omega_p)]\,E_\beta(\mathbf{p}) = 0, \tag{11}$$

$$H_\alpha(\mathbf{k})\,H^*_\alpha(\mathbf{k}) = \varepsilon_{\alpha\beta}E^*_\alpha(\mathbf{k})\,E_\beta(\mathbf{k}). \tag{12}$$

Finally, to determine the meaning of $c_{\mathbf{p}}$ in (8), we must normalize the vectors $\mathbf{E}(\mathbf{k})$ and $\mathbf{H}(\mathbf{k})$. For this purpose, we write down the expression for the electromagnetic density of the medium W (which has meaning only for a transparent medium [4]). By using the expansion (8) and the relation (12), we have (see [4])

$$W = (16\pi)^{-1}\sum_{\mathbf{k}}\left[\frac{d(\omega\varepsilon_{\alpha\beta})}{d\omega}E^*_\alpha(\mathbf{k})\,E_\beta(\mathbf{k}) + H^*_\alpha(\mathbf{k})\,H_\alpha(\mathbf{k})\right]c^*_{\mathbf{k}}c_{\mathbf{k}}$$

$$= (16\pi)^{-1}\sum_{\mathbf{k}}\left[\frac{d(\omega\varepsilon_{\alpha\beta})}{d\omega} + \varepsilon_{\alpha\beta}\right]E^*_\alpha(\mathbf{k})\,E_\beta(\mathbf{k})\,c^*_{\mathbf{k}}c_{\mathbf{k}}, \tag{13}$$

where the summation is carried out over all harmonics with positive frequencies. As in [1], we normalize $\mathbf{E}(\mathbf{k})$ and $\mathbf{H}(\mathbf{k})$ so that W takes the form

$$W = \sum_{\mathbf{k}} c^*_{\mathbf{k}}c_{\mathbf{k}}\,\omega_{\mathbf{k}}, \tag{14}$$

i.e.,

$$(16\pi)^{-1}\left[\frac{d(\omega\varepsilon_{\alpha\beta})}{d\omega} + \varepsilon_{\alpha\beta}\right]E^*_\alpha(\mathbf{k})\,E_\beta(\mathbf{k}) = \omega(\mathbf{k}). \tag{15}$$

In such a normalization, $|c_{\mathbf{k}}|^2 = c^*_{\mathbf{k}}c_{\mathbf{k}}$ has the dimensions of action, and the quantity $|c_{\mathbf{k}}|^2/\hbar$ must be interpreted as the number of quasiparticles with energy $\hbar\omega_{\mathbf{k}}$.

4. Let us now consider the nonlinear term $\mathbf{P}^{(2)}$ in (2), which determines the interaction between the waves. Assuming this interaction to be sufficiently weak (amplitudes of $c_{\mathbf{k}}$ small), we shall seek the solution of the set (1)—(4) in the form

$$\mathbf{E} = V^{-1/2}\sum_{\mathbf{p}=\mathbf{k},\mathbf{k}_-} c_{\mathbf{p}}(t)\,[\mathbf{E}(\mathbf{p}) + \mathbf{E}'(\mathbf{p})]\,e^{i(\mathbf{p}\mathbf{r}-\omega_{\mathbf{p}}t)},$$

$$\mathbf{H} = V^{-1/2}\sum_{\mathbf{p}=\mathbf{k},\mathbf{k}_-} c_{\mathbf{p}}(t)\,[\mathbf{H}(\mathbf{p}) + \mathbf{H}'(\mathbf{p})]\,e^{i(\mathbf{p}\mathbf{r}-\omega_{\mathbf{p}}t)}, \tag{16}$$

where $\mathbf{E}(\mathbf{k})$ and $\mathbf{H}(\mathbf{k})$ are the polarization vectors of the normal waves of linear approximation, determined by the relations (10) and (15), $c_{\mathbf{p}}(t)$ are slowly varying amplitudes [in comparison with the exponents in (16)], $\mathbf{E}'(\mathbf{k})$ and $\mathbf{H}'(\mathbf{k})$ are small, slowly varying additions to the polarization vectors of the normal vibrations.

It will be clear from what follows that if $c_{\mathbf{p}}(t)$ is regarded as small of first order, then $\mathbf{E}'$ and $\mathbf{H}'$ will also be of first order smallness, and $dc_{\mathbf{p}}/dt$ will be of second order; the derivatives $d\mathbf{E}'/dt$, $d\mathbf{H}'/dt$ can also be regarded as small in comparison with $\mathbf{E}'$, $\mathbf{H}'$. Substituting (16) in (3), and limiting ourselves to terms up to second order smallness inclusively, we get

$$\frac{\partial P^{(1)}_\alpha}{\partial t} = \frac{V^{-1/2}}{4\pi}\sum_{\mathbf{p}=\mathbf{k},\mathbf{k}_-} e^{i(\mathbf{p}\mathbf{r}-\omega_{\mathbf{p}}t)}\Big\{i\omega_{\mathbf{p}}[\delta_{\alpha\beta} - \varepsilon_{\alpha\beta}(\omega_{\mathbf{p}})]\,c_{\mathbf{p}}E'_\beta(\mathbf{p})$$

$$+ E_\beta(\mathbf{p})\int_0^\infty \varphi^{(1)}_{\alpha\beta}(\tau)\,e^{i\omega_{\mathbf{p}}\tau}[\dot{c}_{\mathbf{p}}(t-\tau) - i\omega_{\mathbf{p}}c_{\mathbf{p}}(t-\tau)]\Big\}. \tag{17}$$

In the second term we expand $c_{\mathbf{p}}(t)$ in a Fourier integral

$$c_{\mathbf{p}}(t-\tau) = (2\pi)^{-1}\int_{-\infty}^{\infty}\tilde{c}_{\mathbf{p}}(\nu)\,e^{-i\nu(t-\tau)}\,d\nu. \tag{18}$$

Substituting this in (17) and carrying out the elementary transformations in the second term, we get

$$-iE_\beta(\mathbf{p})\int_{-\infty}^{\infty} d\nu\,e^{-i\nu t}(\nu + \omega_{\mathbf{p}})\,[\varepsilon_{\alpha\beta}(\nu + \omega_{\mathbf{p}}) - \delta_{\alpha\beta}]\,\tilde{c}_{\mathbf{p}}(\nu). \tag{19}$$

The smallness of the change in $c_{\mathbf{p}}(t)$ means that $\tilde{c}_{\mathbf{p}}(\nu)$ is substantially different from zero only for $\nu \ll \omega_{\mathbf{p}}$. We can therefore write in (19)

$$(\nu + \omega_{\mathbf{p}})\,[\varepsilon_{\alpha\beta}(\nu + \omega_{\mathbf{p}}) - 1]$$

$$\approx \omega_{\mathbf{p}}[\varepsilon_{\alpha\beta}(\omega_{\mathbf{p}}) - \delta_{\alpha\beta}] + \nu\frac{d}{d\omega}[\omega(\varepsilon_{\alpha\beta} - \delta_{\alpha\beta})]_{\omega=\omega_{\mathbf{p}}}. \tag{20}$$

Substituting (20) in (19), and carrying out the inverse summation of the Fourier components, we get

$$\frac{\partial P^{(1)}_\alpha}{\partial t} = \frac{V^{-1/2}}{4\pi}\sum_{\mathbf{p}=\mathbf{k},\mathbf{k}_-} e^{i(\mathbf{p}\mathbf{r}-\omega_{\mathbf{p}}t)}$$

$$\times\Big\{i\omega_{\mathbf{p}}[\delta_{\alpha\beta} - \varepsilon_{\alpha\beta}(\omega_{\mathbf{p}})]\,[E_\beta(\mathbf{p}) + E'_\beta(\mathbf{p})]\,c_{\mathbf{p}} + \frac{dc_{\mathbf{p}}}{dt}E_\beta(\mathbf{p})$$

$$\times\frac{d}{d\omega}[\omega(\varepsilon_{\alpha\beta} - \delta_{\alpha\beta})]_{\omega=\omega_{\mathbf{p}}}\Big\}. \tag{21}$$

Substituting (16) in (4), we compute $\partial P^{(2)}_\alpha/\partial t$ (with accuracy to terms of second order):

$$\frac{\partial P^{(2)}_\alpha}{\partial t} = -\frac{i}{4\pi V}\sum_{\mathbf{p}} e^{i\mathbf{p}\mathbf{r}}\sum_{\mathbf{p}'+\mathbf{p}''=\mathbf{p}} c_{\mathbf{p}'}c_{\mathbf{p}''}\,\sigma_{\alpha\beta\gamma}(\omega_{\mathbf{p}'}, \omega_{\mathbf{p}''})$$

$$\times E_\beta(\mathbf{p}')\,E_\gamma(\mathbf{p}'')\exp[i(\omega_{\mathbf{p}'} + \omega_{\mathbf{p}''})t], \tag{22}$$

where [1])

$$\sigma_{\alpha\beta\gamma}(\omega', \omega'') = {}^1/_2 (\omega' + \omega'') [\chi_{\alpha\beta\gamma}(\omega' + \omega'', \omega') + \chi_{\alpha\gamma\beta}(\omega' + \omega'', \omega'')], \tag{23}$$

$$\chi_{\alpha\beta\gamma}(\omega', \omega'') = \int_0^\infty d\tau' \int_0^\infty d\tau'' \varphi_{\alpha\beta\gamma}(\tau', \tau'') \exp[i(\omega'\tau' + \omega''\tau'')]. \tag{24}$$

Substituting (21) and (22) in (1) and (2), we equate the identical Fourier **p** components, use (10), and then eliminate $\mathbf{H}'(\mathbf{p})$ from the resulting equations. We then get a set of equations for $E'_\alpha(\mathbf{p})$:

$$\frac{c}{\omega_\mathbf{p}} c_\mathbf{p} \left(p_\alpha p_\beta - p^2\delta_{\alpha\beta} + \frac{\omega_\mathbf{p}^2}{c^2}\varepsilon_{\alpha\beta}\right) E'_\beta = -i\frac{dc_\mathbf{p}}{dt}\left[\varepsilon_{\alpha\beta} + \frac{d(\omega_\mathbf{p}\varepsilon_{\alpha\beta})}{d\omega_\mathbf{p}}\right] E_\beta - V^{-1/2} \sum_{\mathbf{p}'+\mathbf{p}''=\mathbf{p}} c_{\mathbf{p}'} c_{\mathbf{p}''} \sigma_{\alpha\beta\gamma}(\omega_{\mathbf{p}'}, \omega_{\mathbf{p}''}) E_\beta(\mathbf{p}') E_\gamma(\mathbf{p}'') \times \exp[i(\omega_\mathbf{p} - \omega_{\mathbf{p}'} - \omega_{\mathbf{p}''})t]. \tag{25}$$

This set differs from the set of equations (11) for $E'_\alpha(\mathbf{p})$ only in the presence of the right hand side. Consequently, its numerator is equal to zero. For the set of equations (25), it is necessary that the right side of this set be orthogonal to the solutions of the transposed set without the right-hand side, i.e., the vectors $\widetilde{E}'_\alpha(\mathbf{p})$ satisfying the equation

$$[p_\alpha p_\beta - p^2\delta_{\alpha\beta} + (\omega_\mathbf{p}^2/c^2)\varepsilon_{\beta\alpha}(\omega_\mathbf{p})]\widetilde{E}_\beta = 0. \tag{26}$$

On the basis of (7), one easily obtains

$$\widetilde{E}_\alpha(\mathbf{p}) = E^*_\alpha(\mathbf{p}). \tag{27}$$

Thus the consistency condition (25) takes the form

$$i\frac{dc_\mathbf{p}}{dt}\left\{\varepsilon_{\alpha\beta}(\omega_\mathbf{p}) + \frac{d}{d\omega_\mathbf{p}}[\omega_\mathbf{p}\varepsilon_{\alpha\beta}(\omega_\mathbf{p})]\right\} E^*_\alpha(\mathbf{p}) E_\beta(\mathbf{p}) + V^{-1/2} \sum_{\mathbf{p}'+\mathbf{p}''=\mathbf{p}} c_{\mathbf{p}'} c_{\mathbf{p}''} \sigma(\omega_{\mathbf{p}'}, \omega_{\mathbf{p}''}) E^*_\alpha(\mathbf{p}) E_\beta(\mathbf{p}') E_\gamma(\mathbf{p}'') \times \exp[i(\omega_\mathbf{p} - \omega_{\mathbf{p}'} - \omega_{\mathbf{p}''})t] = 0. \tag{28}$$

Using the normalization condition (15), we see that the expression in the curly brackets in (28) is equal to $16\pi\omega_\mathbf{p}$, and we get finally

$$i\frac{dc_\mathbf{p}}{dt} = \sum_{\mathbf{p}',\mathbf{p}''} V_{\mathbf{p}\mathbf{p}'\mathbf{p}''} c_{\mathbf{p}'} c_{\mathbf{p}''} \exp[i(\omega_\mathbf{p} - \omega_{\mathbf{p}'} - \omega_{\mathbf{p}''})t], \tag{29}$$

where

$$V_{\mathbf{p}\mathbf{p}'\mathbf{p}''} = -V^{-1/2}(16\pi\omega_\mathbf{p})^{-1}\sigma_{\alpha\beta\gamma}(\omega_{\mathbf{p}'}, \omega_{\mathbf{p}''}) E^*_\alpha(\mathbf{p}) E_\beta(\mathbf{p}') E_\gamma(\mathbf{p}'') \quad \text{for } \mathbf{p} = \mathbf{p}' + \mathbf{p}'', \tag{30}$$

$$V_{\mathbf{p}\mathbf{p}'\mathbf{p}''} = 0 \quad \text{for } \mathbf{p} \neq \mathbf{p}' + \mathbf{p}''. \tag{30a}$$

We have thus obtained an equation determining the change with time of the amplitudes of the interacting waves, which is identical in form with the dynamic equation for the waves obtained previously.[1] For calculation of $V_{\mathbf{pp}'\mathbf{p}''}$ in explicit form, it is necessary to know the function $\varphi^{(2)}_{\alpha\beta\gamma}(\tau_1, \tau_2)$, which determines the nonlinear response of the medium to the external electric field.

According to [3], the following expressions are valid for these functions:

$$\varphi^{(2)}_{\alpha\beta\gamma}(t_1, t_2) = (i/\hbar)^2 \langle [[Q_\alpha(t_1 + t_2), Q_\beta(t_2)]_-, Q_\gamma]_- \rangle_0. \tag{31}$$

However, for a number of concrete cases in which the quantum properties of the medium are inessential, in particular for a classical plasma, the quantity $\varphi^{(2)}_{\alpha\beta\gamma}(\tau_1, \tau_2)$ is easily computed directly by means of the kinetic or hydrodynamic equations, expressing the polarization vector of the medium in terms of the field intensity, with accuracy up to second order. Proceeding in this fashion in the case of a "cold" plasma in a strong magnetic field ($H^2/8\pi \gg nT$) we get the equations for $V_{\mathbf{pp}'\mathbf{p}''}$ found for this same case in [1] by a somewhat different method.

The "matrix elements" $V_{\mathbf{pp}'\mathbf{p}''}$ satisfy some general symmetry relations. Two of them are easily obtained immediately:

$$V_{\mathbf{pp}'\mathbf{p}''} = V_{\mathbf{pp}''\mathbf{p}'}, \tag{32}$$

$$V_{\mathbf{pp}'\mathbf{p}''} = -V^*_{\mathbf{p}_-\mathbf{p}'_-\mathbf{p}''_-}. \tag{33}$$

The first follows from (30) and (23), and the second is obtained if we take the complex conjugate of (30) and consider that $c^*_\mathbf{p} = c_\mathbf{p}$ [see (9)].

In contrast with (33) two other general relations are satisfied only for

$$\omega_\mathbf{p} = \omega_{\mathbf{p}'} + \omega_{\mathbf{p}''}, \quad \mathbf{p} = \mathbf{k}, \mathbf{k}_- \tag{34}$$

and have the form

$$V_{\mathbf{kk}'\mathbf{k}''} = V^*_{\mathbf{k}''\mathbf{k}_-\mathbf{k}'}, \tag{35a}$$

$$V_{\mathbf{kk}'\mathbf{k}''_-} = -V^*_{\mathbf{k}''\mathbf{k}'_-\mathbf{k}}. \tag{35b}$$

The relations (35) were obtained in [1] for the special case considered there on the basis of a concrete form of the matrix elements of $V_{\mathbf{kk}'\mathbf{k}''}$. We shall not give them here for the general case since this proof is rather cumbersome and the relation (35) is not necessary in this work. We only note that one can get (35a) by starting out from the correspondence between the quantum and classical theories of the interaction of electromagnetic waves in the medium.[2]) Then (35a) follows from

1)The analytic properties of the tensor $\sigma_{\alpha\beta\gamma}(\omega', \omega'')$ were studied by Kogan.[5]

2)We note that the effects of the interaction of the waves appear at such high intensities (the number of photons is $n_\mathbf{k} = c^*_\mathbf{k} c_\mathbf{k}/\hbar \gg 1$) that the quantum corrections are, as a rule, unimportant. Moreover, the quantum consideration, which is the skew symmetry of the matrix element of the S-matrix [see, for example, [6]) which describes the decay of a photon traveling in a transparent medium with energy $\hbar\omega_k$ and momentum $\hbar k$ into two waves: (ω_{k_1}, k_1); (ω_{k_2}, k_2), such that (34) is satisfied and $k = k_1 + k_2$. So far as (35b) is concerned, it is simply obtained from (35a) and (33).

By means of the dynamic equation (29) and the relation (35) one can study the decay instability of waves which was considered previously by Oraevskiĭ and Sagdeev, [7] and can also obtain the kinetic equation describing the interaction of a large number of waves with randomly distributed phases. All this was done in [1] for a plasma; however, inasmuch as a concrete form is not used here for the matrix elements, all these results are valid in an arbitrary transparent medium. [3)]

5. As an example of the application of the dynamic equation (29), let us consider the effect of frequency doubling in a transparent medium. A report on the observation of this effect in crystalline quartz is given in [2].

Let the wave $[\mathbf{k}, \omega(\mathbf{k})]$ be propagated in the medium. Then, on the basis of the dynamic equation (29), a wave appears with the double wave vector $\mathbf{k}_1 = 2\mathbf{k}$; its amplitude satisfies the equation

$$i\frac{dc_{\mathbf{k}_1}}{dt} = V_{\mathbf{k}_1\mathbf{k}\mathbf{k}}\, c_{\mathbf{k}}^2 e^{i(\omega_1-2\omega)t}, \qquad \omega_1 = \omega(\mathbf{k}_1) = \omega(2\mathbf{k}), \qquad \omega = \omega(\mathbf{k}). \tag{36}$$

We then get (neglecting the time dependence of the initial amplitude, which can be done when $c_{\mathbf{k}}$ is large in comparison with $c_{\mathbf{k}_1}$)

$$c_{\mathbf{k}_1}(t) = (2\omega - \omega_1)^{-1} V_{\mathbf{k}_1\mathbf{k}\mathbf{k}} c_{\mathbf{k}}^2 e^{i(\omega_1-2\omega)t}. \tag{37}$$

The equation of the wave with the wave vector $\mathbf{k}_1 = 2\mathbf{k}$ is the following on the basis of expansion of (8),

$$\mathscr{E}_1(\mathbf{r}, t) = V^{-1/2}\mathbf{E}(\mathbf{k}_1)\, c_{\mathbf{k}_1} e^{i(\mathbf{k}_1\mathbf{r}-\omega_1 t)} = V^{-1/2}\mathbf{E}(\mathbf{k}_1)(2\omega - \omega_1)^{-1} V_{\mathbf{k}_1\mathbf{k}\mathbf{k}} c_{\mathbf{k}}^2 e^{i(\mathbf{k}_1\mathbf{r}-2\omega t)}. \tag{38}$$

Thus it follows that the second harmonic is twice the frequency in comparison with the initial wave. [4)]

Equation (38) is valid for

$$|V_{\mathbf{k}_1\mathbf{k}\mathbf{k}} c_{\mathbf{k}} / (2\omega - \omega_1)| \ll 1, \quad |2\omega - \omega_1| \ll \omega_1. \tag{39}$$

The first of the inequalities (39) is the condition of the smallness of $c_{\mathbf{k}_1}$ in comparison with $c_{\mathbf{k}}$, while the second is the condition of the slowness of change of $c_{\mathbf{k}_1}(t)$. Both conditions can be satisfied simultaneously for

$$|V_{\mathbf{k}_1\mathbf{k}\mathbf{k}} c_{\mathbf{k}}/\omega_1| \ll |(2\omega - \omega_1)/\omega_1| \ll 1. \tag{40}$$

If some different waves with the same initial frequency ω are propagated in the medium, then they also can lead to the appearance of waves with double frequency (such a case is encountered below). The amplitudes of the latter will be determined by an equation similar to (37):

$$c_{\mathbf{k}_1} = (2\omega - \omega_1)^{-1} \sum_{\mathbf{k}_1=\mathbf{k}'+\mathbf{k}''} V_{\mathbf{k}_1\mathbf{k}'\mathbf{k}''} c_{\mathbf{k}'} c_{\mathbf{k}''} e^{i(\omega_1-2\omega)t}, \tag{41}$$

where $\mathbf{k}'$ and $\mathbf{k}''$ are the wave vectors of the initial wave.

We now consider in detail the form of the matrix element $V_{\mathbf{k}_1\mathbf{k}'\mathbf{k}''}$. It follows from (30) that one can describe it in the following fashion:

$$V_{\mathbf{k}_1\mathbf{k}'\mathbf{k}''} = -V^{-1/2}(\omega/8\pi\omega_1)\,\mathbf{E}^*(\mathbf{k}_1)\,\mathbf{u}(\mathbf{k}', \mathbf{k}''), \tag{42}$$

where the vector $\mathbf{u}(\mathbf{k}', \mathbf{k}'')$ is determined by the equation

$$u_\alpha(\mathbf{k}', \mathbf{k}'') = (2\omega)^{-1}\sigma_{\alpha\beta\gamma}(\omega, \omega)\, E_\beta(\mathbf{k}')\, E_\gamma(\mathbf{k}''). \tag{43}$$

We now apply these considerations to crystalline quartz which was investigated in [2]. The latter is a uniaxial crystal with symmetry class D_3, containing a three-fold symmetry axis (the z axis) and three two-fold symmetry axes in the xy plane; one of these axes is assumed directed along x. It then follows that the tensor $\sigma_{\alpha\beta\gamma}(\omega)$ will have the form (see [4], page 106)

$$\begin{gathered} \sigma_{x,xx} = -\sigma_{x,yy} = -\sigma_{y,xy} = 2\omega a, \\ \sigma_{x,yz} = -\sigma_{y,xz} = 2\omega b, \end{gathered} \tag{44}$$

where a and b are certain functions of the frequency ω.

Substituting (44) in (43), we get the vector $\mathbf{u}$ in the form

$$\begin{aligned} u_x &= a[E_x(\mathbf{k}')E_x(\mathbf{k}'') - E_y(\mathbf{k}')E_y(\mathbf{k}'')] \\ &\quad + b[E_y(\mathbf{k}')E_z(\mathbf{k}'') + E_y(\mathbf{k}'')E_z(\mathbf{k}')], \\ u_y &= -b[E_x(\mathbf{k}')E_z(\mathbf{k}'') + E_x(\mathbf{k}'')E_z(\mathbf{k}')] \\ &\quad - a[E_x(\mathbf{k}')E_y(\mathbf{k}'') + E_x(\mathbf{k}'')E_y(\mathbf{k}')], \\ u_z &= 0. \end{aligned} \tag{45}$$

The tensor $\epsilon_{\alpha\beta}$ is diagonal in the chosen set of coordinates, whence

$$\varepsilon_{xx} = \varepsilon_{yy} = \varepsilon, \qquad \varepsilon_{zz} = \varepsilon'. \tag{46}$$

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useful in obtaining general relations of the type (35) does not lead in sufficiently simple fashion to the explicit expressions (30) for the matrix elements. We shall therefore not consider it here.

[3)]A report was made in [8,9] on the experimental observation in crystals of a phenomenon that is the opposite of wave decay, namely, the harmonic $(\omega_1 + \omega_2, k_1 + k_2)$ appears in the superposition of two waves (ω_1, k_1), (ω_2, k_2). We note further that one of the decay cases was discussed previously by Akhmanov and Khokhlov[10] as a possible mechanism of wave amplification.

[4)]Inasmuch as $2\omega \neq \omega_1 = \omega(2k)$ as a consequence of the nonlinearity of the dispersion law, then such a wave must be considered as "forced."

It follows from the absence of absorption that ϵ, ϵ', and the polarization vectors $\mathbf{E}(\mathbf{k})$ are real.

We now consider three cases separately.

A. The incident ray is directed along the x axis (that is, along one of the two-fold axes). For arbitrary polarization, it yields ordinary and extraordinary waves having the same frequency and direction of propagation, but different values of the wave vectors and different polarizations; the ordinary wave is polarized along the y axis, and the extraordinary wave along the z axis. Consequently, $E_x(\mathbf{k}') = E_x(\mathbf{k}'')$, and the vector $\mathbf{u}$ has only an x component. Inasmuch as a secondary wave is also propagated along the x axis, $E_x(\mathbf{k}_1) = 0$ and consequently $\mathbf{E}(\mathbf{k}_1)\cdot\mathbf{u}(\mathbf{k}', \mathbf{k}'') = 0$. Therefore $V_{\mathbf{k}_1\mathbf{k}'\mathbf{k}''} = 0$. This means that the wave with double frequency cannot arise in this case.

B. The incident ray is directed along the y axis. For arbitrary polarization, it again yields an ordinary and an extraordinary wave. The first is polarized along the x axis, and the second along the z axis. For both waves, $E_y(\mathbf{k}) = 0$. For the secondary wave, $E_y(\mathbf{k}_1) = 0$ also. Consequently,

$$\mathbf{E}(\mathbf{k}_1)\,\mathbf{u}(\mathbf{k}',\mathbf{k}'') = E_x(\mathbf{k}_1)\,u_x(\mathbf{k}',\mathbf{k}''). \tag{46a}$$

It follows from (45) that $u_x(\mathbf{k}', \mathbf{k}'') \neq 0$ only in the ordinary wave. Consequently, in this case, only the ordinary wave produces a harmonic with double frequency, and the latter is also polarized along x.

The intensity of the wave with double frequency is determined by the energy flux

$$S = (c/4\pi)\,[\mathbf{EH}] = (c/4\pi)\,\mathbf{n}E^2 \tag{46b}$$

(the latter is written with account of the transverse character; the field intensity here is assumed to be real). The vector $\mathbf{n}$ is equal in magnitude to the index of refraction of the wave (here, the ordinary wave) and is directed along the wave vector.

The average value of the energy flux, expressed in terms of the complex intensity vector $\mathscr{E}$, has the form

$$S_1 = (cn_1/8\pi)\,\mathscr{E}_1^*\mathscr{E}_1, \tag{47}$$

where the index 1 denotes that the quantities refer to the secondary wave. We substitute $\mathscr{E}_1$ here from (38) and the expression for the matrix element from (42), (45):

$$|V_{\mathbf{k}_1\mathbf{k}\mathbf{k}}|^2 = V^{-1}\,(\omega^2/(8\pi)^2\omega_1^2)\,a^2E_x^2(\mathbf{k}_1)\,E_x^4(\mathbf{k}). \tag{48}$$

The values of the polarization vectors here are determined by the normalization condition (15), from which it follows that

$$E^2(\mathbf{k}) = 16\pi\omega\,(\omega d\varepsilon/d\omega + 2\varepsilon)^{-1} \approx 8\pi\omega/n^2,$$
$$E^2(\mathbf{k}_1) \approx 8\pi\omega_1/n_1^2, \tag{49}$$

where n and n_1 are the indices of refraction of the ordinary wave, corresponding to the first and second harmonics.

Substituting (49), (48), and (38) in (47), we finally obtain

$$S_1 = \frac{2\pi}{c}\frac{a^2}{n_1n^2(n_1-n)^2}S_0^2 = \frac{2\pi}{c}\frac{a^2}{n_1n^2(n_1-n)^2}S^2\cos^4\varphi, \tag{50}$$

where n and n_1 are the indices of refraction of the ordinary ray, corresponding to the wavelength λ (initial) and $\lambda/2$; S_0 is the energy flux of the initial ordinary wave, and S is the total energy flux or the initial ray in the crystal, φ is the angle between the direction of the polarization of the incident ray (at incidence on the crystal) and the x axis. Thus the intensity of the second harmonic here depends significantly on the polarization of the incident ray.

C. The incident wave is directed along the z axis, that is, the principal axis of the crystal; $E_z(\mathbf{k}) = 0$.

In this case the difference between the ordinary and extraordinary waves vanishes; the primary wave can have arbitrary polarization in the xy plane, i.e., degeneracy sets in. In this connection, it is necessary to make some observations relative to the initial equation (36). It was obtained from (25) by starting out from the fact that the right-hand side of (25) should be orthogonal to the solutions of the transposed system without the right-hand side. But, owing to the degeneracy, the homogeneous system now satisfies not a single vector as was assumed earlier, but all vectors lying in the xy plane. Therefore, the right-hand side of (25) must be orthogonal to any vector lying in the xy plane and, consequently, the x and y components for it must vanish.

Substituting $\mathbf{p} = \mathbf{k}_1$, $\mathbf{p}' = \mathbf{p}'' = \mathbf{k}$ and $\epsilon_{\alpha\beta}$ in (25) from (46), we get the right side of (25) in the form

$$\frac{dc_{\mathbf{k}_1}}{dt}\left(\varepsilon + \frac{d(\omega\varepsilon)}{d\omega}\right)\mathbf{E}(\mathbf{k}_1)$$
$$-2\omega V^{-1/2}c_{\mathbf{k}}^2\mathbf{u}(\mathbf{k},\ \mathbf{k})\exp[i(\omega_{\mathbf{k}_1} - 2\omega_{\mathbf{k}})t] = 0. \tag{51}$$

Here we have introduced the vector $\mathbf{u}$ in accord with (43), and taken into account the fact that $E_z(\mathbf{k}_1) = 0$ ($\mathbf{k}_1 \parallel Oz$) and $u_z(\mathbf{k}, \mathbf{k}) = 0$ [on the basis of (45)].

From Eq. (51) follows, in the first place, the previous Eq. (36), and in the second place the fact that $\mathbf{E}(\mathbf{k}_1)$ and $\mathbf{u}(\mathbf{k}, \mathbf{k})$ are parallel. Thus the polarization of the wave with double frequency is identical with the polarization vector $\mathbf{u}$ with components

$$u_x = a\,[E_x^2(\mathbf{k}) - E_y^2(\mathbf{k})],\ \ u_y = -2aE_x(\mathbf{k})\,E_y(\mathbf{k}),\ \ u_z = 0, \tag{52}$$

where $E_x(\mathbf{k})$, $E_y(\mathbf{k})$ are the components of the polarization vector of the initial wave. It then follows that if $E(\mathbf{k})$ makes the angle φ with the x axis, while the electric field of the secondary wave makes the angle φ_1, then

$$\varphi_1 = 2(\pi - \varphi). \tag{53}$$

Let us compute the intensity of the second harmonic in this case. By taking it into account that $E(\mathbf{k}_1)$ and $u(\mathbf{k}, \mathbf{k})$ in the matrix element (42) are parallel, we get

$$|V_{\mathbf{k},\mathbf{k}'\mathbf{k}''}|^2 = V^{-1}(\omega/8\pi\omega_1)^2 E^2(\mathbf{k}_1) u^2(\mathbf{k}, \mathbf{k})$$
$$= V^{-1}(a\omega/8\pi\omega_1)^2 E^2(\mathbf{k}_1) E^4(\mathbf{k}). \tag{54}$$

By substituting (54), (49), and (38) in (47), we get

$$S_1 = \frac{2\pi}{c}\frac{a^2}{n_1 n^2 (n_1 - n)^2} S^2, \tag{55}$$

where S_1 and S are the energy fluxes of the secondary and initial wave, and n_1 and n are the corresponding indices of refraction. In this case, the intensity of the wave of double frequency does not depend on the polarization of the incident beam.

We note that many qualitative features of the dependence of the intensity of the secondary wave on the polarization of the incident wave pointed out above are identical with the results of the analysis carried out in [2] by another method, and are confirmed by observation. The formula given in [2] for the intensity is in error.

From (55), one can make a numerical estimate of the effect under consideration. In fact, the quantity a^{-1} has the dimensions of the field intensity and must be equal in order of magnitude to the value of the atomic field. Substituting in (55) the expression $S \sim (c/4\pi)E^2$, where E is the field intensity in the medium and $n_1 - n \sim n^{-2}$, we get

$$S_1/S \sim 10^4 (aE)^2. \tag{55a}$$

For $a \sim 10^{-9}$ cm/V, $E \sim 10^5$ V/cm (a feasible value) we get $S_1/S \sim 0.01$ per cent.

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