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Synchronous nonlinear wave interaction in Bragg diffraction in media with periodic structure

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A consistent theory is developed for nonlinear interaction of waves in a medium with an ideal periodic structure, for the case of Bragg diffraction of waves after Laue. Expressions are obtained for the second-harmonic amplitudes along the directions of the incident and diffracted waves. The regions of existence of "Bragg" synchronisms are identified, and the effectivenes of second-harmonic generation at various types of synchronism is analyzed. The theory is of interest for the description of nonlinear effects in optics of periodic structures and in x-ray optics.

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1. INTRODUCTION

The study of nonlinear wave processes in media with periodic structure, when its spatial period is of the order of the wavelength, is of considerable interest for a number of branches of physics. For example, if one speaks of electromagnetic waves, such processes can take place both in the optical band (media with artificial periodic structure,[1,5] integrated-optics elements,[6] liquid crystals,[7] etc.) and in the x-ray band (crystal gratings,[8-12] zeolite crystals).

It is known that Bragg diffraction of waves is possible in media with a periodic structure. Under Bragg-diffraction conditions the nonlinear interaction of the waves acquires entirely new features. In particular, it was shown recently[13-15] that in Bragg diffraction it is possible to realize new types of synchronism in isotropic media with spatially-periodic modulation of the linear and nonlinear susceptibilities. These synchronisms uncover additional possibilities of realizing synchronous interactions of optical waves in isotropic media, where the traditional synchronism method, based on birefringence, is not applicable[15] (this is particularly important for the feasibility of using the short-wave region of the spectrum), and also makes possible a new approach to the problem of frequency doubling in the x-ray band.[14] It should be noted that the synchronisms revealed in our studies include as particular cases two previously considered[1-3] types of synchronous interactions in a periodic structure.

In the present paper, using second harmonic generations (SHG) as an example, we describe a consistent theory of nonlinear interaction of waves that undergo Bragg diffraction in an isotropic medium. It is shown that, generally speaking, in Laue diffraction there exist six synchronism conditions that admit of simple interpretation in the language of the effective refractive indices (ERI) known in the dynamic theory of diffraction.[16-21] In turn, in each synchronism the harmonic generation proceeds simultaneously along six different channels. One of the channels, pertaining to the case of nonlinear diffraction, was discussed by Freund.[1] We have investigated the dependence of the effectiveness of synchronous SHG on the type of synchronism, the algebraic values of the deviation from the exact Bragg diffraction condition, and the parameters of the medium.

2. EQUATIONS FOR ELECTROMAGNETIC FIELDS IN A NONLINEAR MEDIUM WITH PERIODIC STRUCTURE

We consider the interaction of harmonic waves at frequencies ω and 2ω in a medium with quadratic nonlinearity. This process id described by two equations for the complex fields:

$$\operatorname{rot}\operatorname{rot}\mathbf{E}_1(\mathbf{r})-\frac{\omega^2}{c^2}(1+4\pi\chi_1)\mathbf{E}_1(\mathbf{r})=\frac{8\pi\omega^2}{c^2}\hat{\beta}\mathbf{E}_2\mathbf{E}_1^*, \qquad (1)$$

$$\operatorname{rot}\operatorname{rot}\mathbf{E}_2(\mathbf{r})-\frac{4\omega^2}{c^2}(1+4\pi\chi_2)\mathbf{E}_2(\mathbf{r})=\frac{16\pi\omega^2}{c^2}\hat{\beta}\mathbf{E}_1^2. \qquad (2)$$

Here $\mathbf{E}_j(t,\mathbf{r})=\mathbf{E}_j(\mathbf{r})\exp(i\omega_j t)+\text{c.c.}$, $\chi_j(\mathbf{r})$ is the linear

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susceptibility, $\hat{\beta}(\mathbf{r})$ is the quadratic-susceptibility tensor, $\omega_j = j\omega$, $j=1,2$.

In a medium with ideal spatially periodic structure, the susceptibilities can be expanded in a Fourier series in terms of the vectors of the reciprocal vector $\mathbf{h}$:

$$\chi_j(\mathbf{r}) = \frac{1}{4\pi}\sum_m \chi_{j,mh} e^{im\mathbf{hr}}, \quad \hat{\beta}(\mathbf{r}) = \frac{1}{4\pi}\sum_m \hat{\beta}_{mh} e^{im\mathbf{hr}}, \quad m=0, \pm1, \pm2\dots . \tag{3}$$

Let the waves propagate at an angle close to the Bragg-reflection angle $\theta \approx \theta_B = \arcsin(hc/2\omega)$. Then, besides the waves traveling in the principal direction, waves of different diffraction orders will appear in the medium. Confining ourselves to the two-wave approximation,[16-21] we represent the sought fields in the form of a sum of a transmitted (index 0) and reflected (indices $h, 2h$) waves

$$E_j(r) = \mathbf{e}_0 E_{j,0} \exp(ij\mathbf{k}_0\mathbf{r}) + \mathbf{e}_h E_{j,jh} \exp(ij\mathbf{k}_h\mathbf{r}), \tag{4}$$

where $E_{j,0}$ and $E_{j,jh}$ are the wave amplitudes, which undergo slow changes as a result of the Bragg diffraction and the nonlinear interaction,

$$\mathbf{k}_h = \mathbf{k}_0 + \mathbf{h}, \quad k_0 = n_0\omega/c, \quad n_0^2 = 1 + (\chi_{1,0} + \chi_{2,0})/2,$$

the vector $\mathbf{k}_0$ is directed along the "0" direction, $\mathbf{e}_0$ and $\mathbf{e}_h$ are the polarization unit vectors and are located either in the scattering plane $(\mathbf{k}_0, \mathbf{k}_h)$ (case of π polarization) or are perpendicular to this plane (σ polarization).

Substituting (3) and (4) in (1) and (2), and discarding the small second derivatives of the amplitudes, we obtain a system of abbreviated equations:

$$-i\frac{n_0 c}{\omega}\left(\cos\psi_0 \frac{\partial E_{2,0}}{\partial z} - \sin\psi_0 \frac{\partial E_{2,0}}{\partial x}\right) = \chi_{2,-2h} E_{2,2h} + \frac{\Delta}{2} E_{2,0} + P_{2,0}, \tag{5a}$$

$$-i\frac{n_0 c}{\omega}\left(\cos\psi_h \frac{\partial E_{2,2h}}{\partial z} + \sin\psi_h \frac{\partial E_{2,2h}}{\partial x}\right) = \left(\frac{\Delta}{2} - \alpha\right) E_{2,2h} + \chi_{2,2h} E_{2,0} + P_{2,2h}, \tag{5b}$$

$$-2i\frac{n_0 c}{\omega}\left(\cos\psi_0 \frac{\partial E_{1,0}}{\partial z} - \sin\psi_0 \frac{\partial E_{1,0}}{\partial x}\right) = \chi_{1,-h} E_{1,h} - \frac{\Delta}{2} E_{1,0} + P_{1,0}, \tag{5c}$$

$$-2i\frac{n_0 c}{\omega}\left(\cos\psi_h \frac{\partial E_{1,h}}{\partial z} + \sin\psi_h \frac{\partial E_{1,h}}{\partial x}\right) = \left(-\frac{\Delta}{2} - \alpha\right) E_{1,h} + \chi_{1,h} E_{1,0} + P_{1,h}, \tag{5d}$$

where the parameter

$$\alpha = (k_h^2 - k_0^2)c^2/\omega^2 \approx 2n_0^2(\theta - \theta_B)\sin 2\theta_B \tag{6}$$

characterizes the deviation from the Bragg condition

$$\theta = {}^1/_2 \sphericalangle(\mathbf{k}_0\mathbf{k}_h), \quad \psi_0 = \sphericalangle(\mathbf{k}_0, \mathbf{n}), \quad \psi_h = \sphericalangle(\mathbf{n}\mathbf{k}_h),$$

$\mathbf{n}$ is the normal to the surface of the sample, the parameter $\Delta = \chi_{2,0} - \chi_{1,0}$ characterizes the frequency dispersion of the medium, the quantities $|\alpha|, |\Delta|, |\chi_{j,jh}| \ll 1$ are much smaller than unity, and

$$P_{2,0} = \beta_0^{(0,0,0)} E_{1,0}^2 + 2\beta_{-h}^{(0,0,h)} E_{1,0}E_{1,h} + \beta_{-2h}^{(0,h,h)} E_{1,h}^2, \tag{7a}$$

$$P_{2,2h} = \beta_0^{(h,h,h)} E_{1,h}^2 + 2\beta_h^{(h,h,0)} E_{1,h}E_{1,0} + \beta_{2h}^{(h,0,0)} E_{1,0}^2, \tag{7b}$$

$$P_{1,0} = 2(\beta_0^{(0,0,0)} E_{2,0}E_{1,0}^* + \beta_{-h}^{(0,h,h)} E_{2,2h}E_{1,h}^* + \beta_h^{(0,0,h)} E_{2,0}E_{1,h}^* + \beta_{-2h}^{(0,h,0)} E_{2,2h}E_{1,0}^*), \tag{7c}$$

$$P_{1,h} = 2(\beta_0^{(h,h,h)} E_{2,2h}E_{1,h}^* + \beta_{-h}^{(h,h,0)} E_{2,2h}E_{1,0}^* + \beta_h^{(h,0,0)} E_{2,0}E_{1,0} + \beta_{2h}^{(h,0,h)} E_{2,0}E_{1,h}^*). \tag{7d}$$

Here $\beta_{mh}^{(a,b,c)} = \mathbf{e}_a\hat{\beta}_{mh}\mathbf{e}_b\mathbf{e}_c$ are the convolutions of the various Fourier components of the quadratic-polarizability tensor. The polarization factor, equal to $\cos 2\theta$ in the case of π polarization, is included in the quantities $\chi_{1,h}$ and $\chi_{2,2h}$ (in this case $\chi_{j,jh}^{\pi} = \chi_{j,jh}^{\sigma}\cos 2\theta$). In the case of a linear medium ($\beta \to 0$) the system (5) goes over into the Takagi equations.[16,17,20,21]

From the expressions for $P_{2,0}$, $P_{2,2h}$ we see that in the case of second-harmonic generation there take place both collinear interactions

$$k_{1,0} + k_{1,0} = k_{2,0}, \quad k_{1,h} + k_{1,h} = k_{2,2h}$$

(terms with β_0) and noncollinear interactions of two types

$$\mathbf{k}_{1,0} + \mathbf{k}_{1,h} - \mathbf{h} = \mathbf{k}_{2,0}, \quad \mathbf{k}_{1,0} + \mathbf{k}_{1,h} + \mathbf{h} = \mathbf{k}_{2,2h}$$

(the terms with β_{-h} and β_h, these interactions are shown dashed in Fig. 1) and

$$\mathbf{k}_{1,h} + \mathbf{k}_{1,h} - 2\mathbf{h} = \mathbf{k}_{2,0}, \quad \mathbf{k}_{1,0} + \mathbf{k}_{1,0} + 2\mathbf{h} = \mathbf{k}_{2,2h}$$

(terms with β_{-2h} and β_{2h}, see Fig. 1).

We consider next with the aid of Eqs. (5) the SHG by a plane wave, neglecting the reaction of the second-harmonic field on the fundamental-frequency wave $P_{1,0} = P_{1,h} = 0$. For convenience we omit the superior index of $\beta_{mh}^{(a,b,c)}$, with the exception of $\beta_0^{(0,0,0)} = \beta_0^{(0)}$, $\beta_0^{(h,h,h)} = \beta_0^{(h)}$. We put also $\chi_{1,h} = \chi_h$, $\chi_{2,2h} = \chi_{2h}$. From the energy conservation law it follows that in a medium without absorption we have $\chi_{jh}^* = \chi_{-jh}$, $\beta_{jh}^* = \beta_{-jh}$.

3. SECOND HARMONIC GENERATION IN LAUE DIFFRACTION

Let the normal $\mathbf{n}$ to the entry surface of the medium be perpendicular to the reciprocal-lattice vector (symmetrical case of Laue diffraction), in which case $\psi_0 = \psi_h = 0$ (Fig. 1). At the entry to the medium, at $z=0$, the wave amplitudes are equal to

$$E_{1,0}(0) = E, \quad E_{2,0}(0) = E_{1,h}(0) = E_{2,2h}(0) = 0. \tag{8}$$

Integrating Eqs. (5c) and (5d) at $P_{1,0} = P_{1,h} = 0$ we obtain the Pendellosung known from dynamic diffraction theory for the field of the fundamental radiation.[16-21] We represent the solution in the form of a sum of two elementary types of fields ($\pm$):

$$E_{1,0}(z) = \frac{E}{2}\exp\left\{-i(\Delta+\alpha)\frac{z_L}{4}\right\}\sum_{\pm}\left(1 \pm \frac{\alpha}{2\gamma_1}\right)\exp\left(\pm\frac{i\gamma_1 z_L}{2}\right),$$
$$E_{1,h}(z) = \frac{E}{2}\frac{\chi_h}{\gamma_1}\exp\left\{-i(\Delta+\alpha)\frac{z_L}{4}\right\}\sum_{\pm}\left\{\pm\exp\left(\pm\frac{i\gamma_1 z_L}{2}\right)\right\}, \tag{9}$$

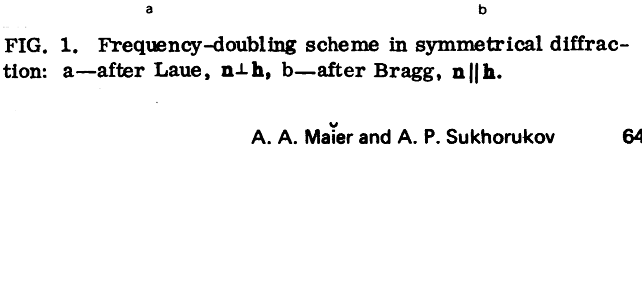

FIG. 1. Frequency-doubling scheme in symmetrical diffraction: a—after Laue, $\mathbf{n}\perp\mathbf{h}$, b—after Bragg, $\mathbf{n}\parallel\mathbf{h}$.

where

$$\gamma_1=(\alpha^2/4+\chi_1^2)^{1/2},\quad \chi_1^2=\chi_h\chi_{-h},\quad z_L=z\omega/cn_0\cos\theta.$$

Substituting (9) in (7a) and (7b) and integrating (5a) and (5b), we obtain expressions for the second-harmonic field amplitudes

$$E_{2,0}(z)=\frac{E^2}{8\gamma_2}\exp\left\{i(\Delta-\alpha)\frac{z_L}{2}\right\}\sum_{p,q}C_{p,q}\frac{\exp(i(p\gamma_1-\Delta)z_L)-\exp(iq\gamma_2z_L)}{p\gamma_1-q\gamma_2-\Delta},$$

$$E_{2,2h}(z)=-\frac{E^2}{8\gamma_2}\exp\left\{i(\Delta-\alpha)\frac{z_L}{2}\right\}\sum_{p,q}C_{p,q}\frac{(\alpha/2-q\gamma_2)}{\chi_{-2h}}\cdot \qquad (10a)$$

$$\cdot\frac{\exp(i(p\gamma_1-\Delta)z_L)-\exp(iq\gamma_2z_L)}{p\gamma_1-q\gamma_2-\Delta} \qquad (10b)$$

where

$$p=-1,\ 0,1,\quad q=-1,1,\quad \gamma_j=(\alpha^2/4+\chi_j^2)^{1/2},\quad \chi_j^2=\chi_{jh}\chi_{-jh},\quad j=1,\ 2,$$

$$C_{p,q}=(q\alpha/2+\gamma_2)F_0^{(p)}+q\chi_{-2h}F_{2h}^{(p)},$$

$$F_0^{(p)}=\beta_0^{(0)}\left(1+\frac{p\alpha}{2\gamma_1}\right)^2+2\beta_{-h}\frac{p\chi_h}{\gamma_1}\left(1+\frac{p\alpha}{2\gamma_1}\right)+\beta_{-2h}\frac{\chi_h^2}{\gamma_1^2},\quad p=\pm1,$$

$$F_0^{(0)}=\frac{2}{\gamma_1^2}(\beta_0^{(0)}\chi_1^2-\beta_{-h}\chi_h\alpha-\beta_{-2h}\chi_h^2),\quad p=0,$$

$F_{2h}^{(p)}$ is obtained from $F_0^{(p)}$ by the substitution

$$\beta_0^{(0)}\to\beta_{2h},\quad \beta_{-h}\to\beta_h,\quad \beta_{-2h}\to\beta_0^{(h)}.$$

We note that

$$\sum_p F_0^{(p)}=4\beta_0^{(0)}.$$

It is seen from (10) that when the condition

$$p\gamma_1-q\gamma_2=\Delta \qquad (11)$$

is satisfied the expressions for the fields acquire resonant terms that increase in proportion to the transversed distance z:

$$E_{2,0}=i\frac{C_{p,q}}{8\gamma_2}z_LE^2\exp\left\{iz_L\left(\frac{\Delta}{2}-\frac{\alpha}{2}+q\gamma_2\right)\right\},$$

$$E_{2,2h}=-i\frac{C_{p,q}}{8\gamma_2}\frac{(\alpha/2-q\gamma_2)}{\chi_{-2h}}z_LE^2\exp\left\{iz_L\left(\frac{\Delta}{2}-\frac{\alpha}{2}+q\gamma_2\right)\right\}. \qquad (12)$$

Relations (11) are the phase-synchronism conditions for frequency doubling in a medium with a periodic structure in Bragg diffraction after Laue. These conditions can be expressed in terms of the ERI (Ref. 21):

$$n_{j,0}^{(\pm)}=(1+\chi_{j,0}-\alpha/2\pm\gamma_j)^{1/2},\quad n_{j,h}^{(\pm)}=(1+\chi_{j,0}+\alpha/2\pm\gamma_j)^{1/2},\quad j=1,2,$$

the plus and minus signs correspond to the two elementary field types, see (9). Recognizing that $p=0$ and ±1 and that $q=\pm1$, we obtain from (11) the following six types of synchronism:

$$\begin{array}{lllll}
\text{I} & p=1, & q=-1: & \gamma_1+\gamma_2=\Delta, & n_1^{(+)}=n_2^{(-)},\\
\text{II} & p=0, & q=-1: & \gamma_2=\Delta, & n_1^{(+)}+n_1^{(-)}=2n_2^{(-)},\\
\text{III} & p=-1, & q=-1: & -\gamma_1+\gamma_2=\Delta, & n_1^{(-)}=n_2^{(-)},\\
\text{IV} & p=1, & q=1: & \gamma_1-\gamma_2=\Delta, & n_1^{(+)}=n_2^{(+)},\\
\text{V} & p=-1, & q=1: & \gamma_1+\gamma_2=-\Delta, & n_1^{(-)}=n_2^{(+)},\\
\text{VI} & p=0, & q=1: & \gamma_2=-\Delta, & n_1^{(-)}+n_1^{(+)}=2n_2^{(+)}.
\end{array} \qquad (13)$$

The synchronisms I and III–V are satisfied in the case of a deviation from the Bragg condition [see (6)]

$$\alpha_s^2=\left(\Delta+\frac{\chi_1^2-\chi_2^2}{\Delta}\right)^2-4\chi_1^2,$$

and synchronisms II and VI at a deviation

$$\alpha_s^2=4(\Delta^2-\chi_2^2).$$

It must be emphasized, however, that the regions of existence of the synchronisms do not coincide (see Fig. 2). On the regions of the boundaries, designated by dashed lines in Fig. 2, the conditions of the corresponding synchronisms are satisfied under the Bragg condition: $\alpha_s=0$. Near these boundaries, the case $|\alpha_s|\ll\chi_j$ is realized.

In synchronism I the fundamental-frequency field of the plus type interacts synchronously with the doubled frequency field of the minus type (Fig. 3). In synchronism II, a synchronous interaction of both types ($\pm$) of fields of fundamental frequency takes place with the field of minus type of the doubled frequency. The interpretation of synchronisms III–VI is analogous. In each synchronism (13), at the frequency 2ω only one type of field is synchronously excited in both directions "0" and "h": the minus type in synchronisms I–III ($q=-1$), and the plus type in synchronisms IV–VI ($q=1$) [see (12)].

It is known that the presence of only one type does not lead to extinction beats,[16-21] therefore the synchronously excited second harmonic has no extinction beats typical of the Pendellosung (9). The barely noticable extinction beats at the frequency 2ω (Fig. 4a) in synchronism I can be attributed to the weak nonsynchronous generation of a plus-type harmonic field.

In principle, however, two different types of synchronism can appear simultaneously: for example, II and IV at $\alpha_{sII}=\alpha_{sIV}$, $\Delta^2=(\chi_1^2-\chi_2^2)/3$, near the point D (Fig. 2). Then excitation of two elementary types of fields ($\pm$) simultaneously at the frequency 2ω leads to spatial extinction beats of the second-harmonic waves "0" and "h" (Fig. 4b) with a half-period (extinction length)

$$z_{2,e}=cn_0\pi\cos\theta/\omega\gamma_2.$$

The effectiveness of the synchronous SHG, as seen from (12), is determined by the coefficients

$$C_{p,q}^{(0)}=|C_{p,q}|/8\gamma_2,\qquad C_{p,q}^{(h)}=|C_{p,q}(^1/_2\alpha-q\gamma_2)/8\gamma_2\chi_{-2h}|. \qquad (14)$$

The effective nonlinear susceptibilities (ENS) $C_{pq}^{(0,h)}$ depend on the combination of several Fourier components of both the linear ($\chi_{\pm jh}$) and the nonlinear ($\beta_{\pm mh}$) susceptibilities and of the parameter α. This is a reflection of the fact that when SHG is produced in the periodic structure interference takes place between the contributions from the synchronous processes that occur si-

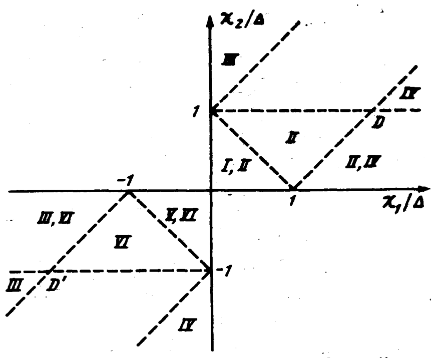

FIG. 2. Regions of existence of six "Bragg" synchronisms in Laue diffraction.

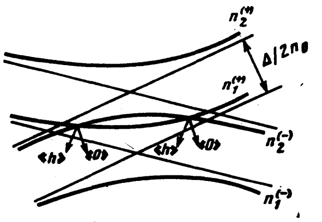

FIG. 3. Intersection of the dispersion hyperbolic surfaces for the effective refractive indices $n_j^{(\pm)}$ in reciprocal-lattice space (synchronism I), n_0 is the average refractive index.

multaneously in six different channels (different $\beta_{\pm mh}$, see above and Fig. 1). Under certain conditions, the contributions of the individual channels can mutually cancel each other, in which case the corresponding ENS vanishes and the effective SHG is impossible in this synchronism. The effect of "quenching" the synchronism in SHG in a periodic structure is the nonlinear analog of the effect of quenching or "forbiddenness" of certain reflections ($\chi_{jh}=0$) in linear Bragg diffraction.

Since usually the dispersion is large enough $\Delta \gtrsim \chi_1 + \chi_2$, greatest interest attaches to synchronisms I and II. We present therefore an analysis of the ENS for only these synchronisms. The treatment of the other ENS is similar.

Synchronisms far from the Bragg diffraction condition

Let the synchronisms be satisfied at large deviations from the Bragg diffraction conditions, $\alpha_s^2 \gg 4\chi_j^2$, in which case

$$\gamma_j \approx |\alpha_s|/2 + \chi_j^2/|\alpha_s|.$$

For the synchronism I ($\alpha_{sI}^2 \sim \Delta^2$) this case is realized at $(\chi_1^2+\chi_2^2)/\Delta^2 \ll 4$, and for II ($\alpha_{sII}^2 \sim 4\Delta^2$)—at $\chi_2^2/\Delta^2 \ll 4$.

In the region $\alpha_s > 0$ ($k_h > k_0$) we get from (10) and (14)

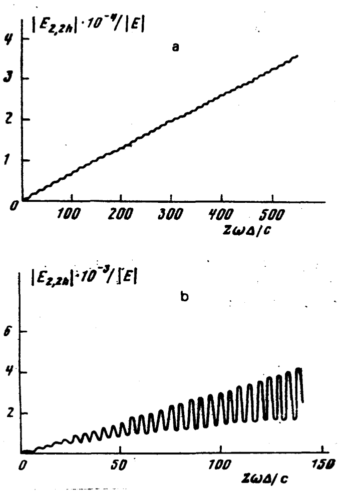

FIG. 4. Growth of the second-harmonic amplitude with increasing distance in Laue diffraction [results of numerical solution of the system (5)]: a—synchronism I is satisfied, $\alpha = \alpha_{sI}$, $\chi_{2h}/\chi_h = |\beta_{2h}|/|\beta_h|/=|\beta_h|/\beta_0 = 0.1$, $\beta_0 E/\Delta \approx 1.8\cdot 10^{-4}$, $\Delta \approx 1.5\chi_h$; b—two synchronism II and IV occur simultaneously, $\alpha = \alpha_{sII} = \alpha_{sIV}$; the parameters chosen correspond to the region near the point D (see Fig. 2), $\chi_{2h}/\chi_h = 0.48$, $|\beta_{2h}| = 0.1|\beta_h| = 0.01\beta_0$, $\beta_0 E/\Delta \approx 0.13\cdot 10^{-2}$, $z_{2,e}\omega\Delta/c \approx 20$.

$$C_{p,-1}^{(h)} \approx |F_{2h}^{(p)}|/4, \quad C_{p,-1}^{(0)} \approx |F_{2h}^{(p)}||\chi_{-2h}|/4\alpha_s, \tag{15}$$

$$\frac{|F_{2h}^{(p)}|}{4} \approx \begin{cases} |\beta_{2h} + 2\beta_h\chi_h/\alpha_s + \beta_0^{(h)}\chi_h^2/\alpha_s^2|, & p=1 \quad \text{(I)} \\ 2|\beta_{2h}\chi_1^2/\alpha_s^2 - \beta_h\chi_h/\alpha_s - \beta_0^{(h)}\chi_h^2/\alpha_s^2|, & p=0 \quad \text{(II)} \end{cases}. \tag{16}$$

Since $\alpha_s^2 \gg 4\chi_2^2$, it follows from (15) that the synchronisms will be much more intensive in the "h" direction than in the "0" direction: $C_{p,-1}^{(h)} \gg C_{p,-1}^{(0)}$. The relative effectiveness of synchronisms I and II along the channel β_{2h} is determined by the small quantity χ_1^2/α_s^2, while β_h and $\beta_0^{(h)}$ are practically the same in the other channels.

If the modulation of the linear susceptibility is vanishingly small, $\chi_j/\alpha_s \sim \chi_j/\Delta \to 0$, then only the ENS for synchronism I along the "h" direction remains different from zero: $C_{1,-1}^{(h)} = |\beta_{2h}|$, and is connected in this case with the interaction channel due to the modulation of the quadratic susceptibility. The condition for synchronism I assumes in this case the form $\alpha_{sI} = \Delta$; the nonlinear diffraction condition $\mathbf{k}_{2,2h} = 2\mathbf{k}_{1,0} + 2\mathbf{h}$ is then satisfied (this type of synchronism was first discussed by Freund[1]). The last case can be used to obtain effective SHG in an isotropic medium with large frequency dispersion.

In the region $\alpha_s < 0$ ($k_h < k_0$) we get from (10) and (14) the expressions

$$C_{p,-1}^{(0)} \approx |F_0^{(p)}|/4, \quad C_{p,-1}^{(h)} \approx |F_0^{(p)}\chi_{2h}/4\alpha_s|,$$

$$\frac{|F_0^{(p)}|}{4} \approx \begin{cases} |\beta_0^{(0)}(\chi_1/\alpha_s)^4 + \beta_{-h}\chi_h\chi_1^2/|\alpha_s|^3 + \beta_{-2h}\chi_h^2/\alpha_s^2|, & p=1 \quad \text{(I)} \quad (17) \\ 2|\beta_0^{(0)}\chi_1^2/\alpha_s^2 - \beta_{-h}\chi_h/\alpha_s - \beta_{-2h}\chi_h^2/\alpha_s^2|, & p=0 \quad \text{(II)} \end{cases}. \tag{18}$$

In this region the situation is reversed: the stronger synchronisms are those along the "0" direction:

$$C_{p,-1}^{(h)}/C_{p,-1}^{(0)} \sim |\chi_{2h}|/|\alpha_s| \ll 1,$$

and the ENS for synchronism II is $2|\alpha_s|/|\chi_h|$ times larger than for synchronism I in the case of a centrosymmetric medium ($\beta_0^{(0)}=0$) and $2\alpha_s^2/\chi_1^2$ times larger for a non-centrosymmetric medium ($\beta_0^{(0)} \neq 0$). At a vanishingly small modulation of the linear susceptibility $\chi_j/\alpha_s \to 0$ the effectivenesses of both synchronisms drop to zero.

Thus, in the region of large positive values of α, the synchronous SHG manifests itself primarily in the "h" direction, while in the region of negative values with large moduli—in the "0" direction. This result is understandable from physical considerations. As already mentioned, in the synchronism I–III ($q=-1$) a field of the minus type is excited synchronously at the frequency 2ω. As seen from Fig. 5c, if $\alpha \gg \chi_2$, then $|E_{2,2h}^{(-)}| \gg |E_{2,0}^{(-)}|$, therefore a second-harmonic wave will be effectively excited only in the "h" direction. At $\alpha \ll -\chi_2$, on the contrary, $|E_{2,0}^{(-)}| \gg |E_{2,2h}^{(-)}|$ and only a harmonic wave in the "0" direction will be effectively generated.

The opposite picture is observed for synchronisms IV–VI ($q=1$).

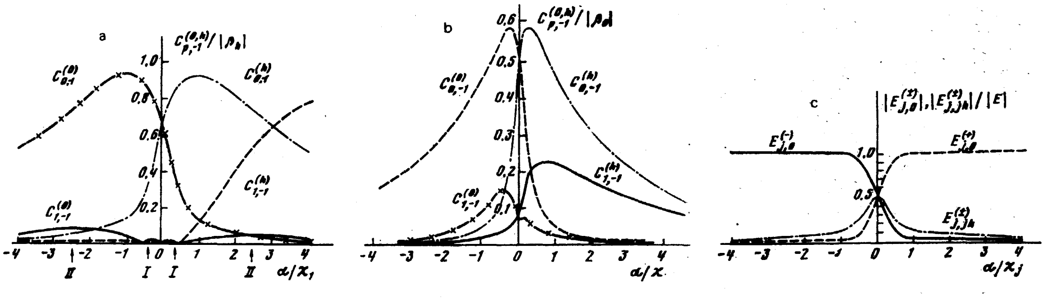

FIG. 5. Plots of effective nonlinear susceptibilities (ENS) in the "x-ray" (a, the arrows indicate the positions of the synchronisms) and "optical" (b) cases, c—dependence of the amplitudes of the two types of field on the parameter α.

Synchronisms near the Bragg diffraction condition

Let the synchronism be satisfied with small deviation from the Bragg condition: $|\alpha_s| \ll \chi_j$. For synchronism I this condition is satisfied at $|\chi_1+\chi_2-\Delta| \ll \chi_j$, and for II at $|\chi_2-\Delta| \ll \chi_j$. Then the ENS for synchronism I is equal to

$$C_{1,-1}^{(0)} \approx C_{1,-1}^{(h)} \approx {}^1/_8 |\beta_0^{(0)} + 2\beta_{-h}\chi_h/\chi_1 + \beta_{-2h}\chi_h^2/\chi_1^2 - (\beta_{2h} + 2\beta_h\chi_h/\chi_1 + \beta_0^{(h)}\chi_h^2/\chi_1^2)\chi_{-2h}/\chi_2|. \tag{19}$$

Similarly for synchronism II at $|\alpha_{sII}| \ll \chi_2$ we have

$$C_{0,-1}^{(0)} \approx C_{1,-1}^{(h)} \approx {}^1/_4 |\beta_0^{(0)} - \beta_{-2h}\chi_h^2/\chi_1^2 - (\beta_{2h} - \beta_0^{(h)}\chi_h^2/\chi_1^2)\chi_{-2h}/\chi_2|. \tag{20}$$

In the optical band, for a medium with induced or natural periodic structure, the typical situation is $\chi_{jh} = \chi_{-jh} = \chi_j$. In this case we get from (19) and (20)

$$C_{1,-1}^{(0)} \approx C_{1,-1}^{(h)} \approx {}^1/_8 |\beta_0^{(0)} - \beta_0^{(h)} + 2(\beta_{-h} - \beta_h) + \beta_{-2h} - \beta_{2h}|,$$

$$C_{0,-1}^{(0)} \approx C_{0,-1}^{(h)} \approx {}^1/_4 |\beta_0^{(0)} + \beta_0^{(h)} - \beta_{-2h} + \beta_{2h}|.$$

If the medium has no inversion center, then its quadratic susceptibility has a dc component β_0. An example of such a medium is lithium niobate[1)] with $\beta_0 \sim 10^{-7}$ cgs esu, in which it is possible to induce a periodic distribution of the refractive index with a modulation depth $\sim 10^{-4}$. Then usually $|\beta_0^{(0)}| \sim |\beta_0^{(h)}| \gg |\beta_h| \gg |\beta_{2h}|$ and the ENS of synchronism II is much larger than that of synchronism I:

$$C_{0,-1}^{(0)} \approx |\beta_0|/2 \gg C_{1,-1}^{(0)} \approx |\beta_h|.$$

If $\beta_0^{(0)} = \beta_0^{(h)} = 0$ and β_h is pure imaginary, then $C_{1,-1}^{(0)} \neq 0$, $C_{0,-1}^{(0)} \approx 0$ and synchronism I is more effective than synchronism II. In this case, however, the quantities $|\beta_h|$ and $|\beta_{2h}|$ are quite small and both synchronisms manifest themselves weakly.

In the x-ray band for a centrosymmetrical crystal

$$\chi_{jh} = \chi_{-jh} = -\chi_j < 0, \quad \beta_0^{(0)} = \beta_0^{(h)} = 0$$

and the ENS are determined by the pure imaginary quantities $\beta_h = -\beta_{-h}$, $\beta_{2h} = -\beta_{-2h}$ (see the Appendix). In this case

$$C_{1,-1}^{(0)} \approx C_{1,-1}^{(h)} \approx {}^1/_8 |\beta_{-2h} + \beta_{2h} - 2(\beta_{-h} + \beta_h)| = 0,$$

$$C_{0,-1}^{(0)} \approx C_{0,-1}^{(h)} \approx {}^1/_4 |\beta_{2h} - \beta_{-2h}| = |\beta_{2h}|/2 \neq 0.$$

Thus, in the x-ray band ($\chi < 0$) the synchronism I at $|\alpha_{sI}| \ll \chi_j$ cannot be observed in a centrosymmetrical crystal, while the synchronism II can appear in this case ($|\alpha_{sII}| \ll \chi_j$.

Absence of Bragg diffraction at one of the frequencies

If the Bragg diffraction of the fundamental wave is forbidden, $\chi_1 = 0$, and the diffraction of the excited second harmonic is possible, $\chi_2 \neq 0$, then

$$F_{2h}^{(0)} = F_0^{(0)} = 0, \quad F_0^{(1)} = \beta_0^{(0)}(1+\alpha/|\alpha|)^2, \quad F_{2h}^{(1)} = \beta_{2h}(1+\alpha/|\alpha|)^2.$$

Synchronism II loses its force, since $C_{0,-1}^{(0,h)} = 0$. Synchronism I appears only in the region $\alpha > 0$ at $\alpha_{sI} = (\Delta^2 - \chi_2^2)/\Delta$ and the corresponding ENS at synchronism are equal to

$$C_{1,-1}^{(0)} = |\chi_{-2h}(\beta_0^{(0)}\chi_{2h} - \Delta\beta_{2h})|/(\Delta^2+\chi_2^2), \tag{21a}$$

$$C_{1,-1}^{(h)} = C_{1,-1}^{(0)} |(\Delta^2 - \chi_2^2 + 2\gamma_2\Delta)/2\chi_{-2h}\Delta|. \tag{21b}$$

If the parameters satisfy the relation $\beta_0^{(0)}\chi_{2h} = \Delta\beta_{2h}$, then $C_{1,-1}^{(0)} = 0$, "quenching" of the synchronism takes place (above), and no synchronous generation of the harmonic occurs.

If there is no Bragg diffraction at either frequency, $\chi_1 = \chi_2 = 0$, then $C_{1,-1}^{(0)} = 0$, $C_{1,-1}^{(h)} = |\beta_{2h}|$ and we arrive at the earlier result [see (15) and (16)].

If the single-wave approximation $\chi_2 = 0$ is satisfied at the doubled frequency, and the two-wave approximation $\chi_1 \neq 0$ is satisfied at the fundamental frequency, then the synchronisms in the "0" direction appear only in the region of negative α_s: $C_{p,-1}^{(0)} = 0$ at $\alpha_s > 0$ and $C_{p,-1}^{(0)} = |F_0^{(p)}|/4$ at $\alpha_s < 0$, while the synchronisms in the "h" direction appear only for positive α_s: $C_{p,-1}^{(h)} = |F_{2h}^{(p)}|/4$ at $\alpha_s > 0$ and $C_{p,-1}^{(p)} = 0$ at $\alpha_s < 0$.

The considered general regularities of the behavior of the ENS can be easily seen on the plots of Fig. 5, which were constructed for the intermediate region $|\alpha| \leqslant 4\chi_1$. The positions of the synchronisms depend on the dispersion of the medium Δ and are marked for the concrete case on Fig. 5. The case a can be realized in a silicon crystal, at a fundamental-radiation wavelength $\lambda = 0.71$ Å (the MoK α line), $\mathbf{h} = [220]$, $2\mathbf{h} = [440]$, $\chi_{2h}/\chi_h = 0.17$ (Ref. 21). β_{jh} is pure imaginary, and the ratio $\beta_{2h}/\beta_h \approx {}^4/_3$ was calculated from formula (A.1) (Refs. 8 and 12) and $\beta_0^{(0)} = \beta_0^{(h)} = 0$. The case b can be realized in the optical band in a medium with artificially induced periodic structure. The ratio of the susceptibilities with spatial frequencies $2h$ and h was chosen to be of the order of the depth of modulation of the refractive index (see Fig. 4a). It was assumed here that $\chi_{jh} = \chi_{-jh} > 0$, $\beta_0^{(0)} = \beta_0^{(h)} > 0$ are real quantities, and β_{jh} has equal real and imaginary parts ($\beta_{jh}^* = \beta_{-jh}$).

Angular and frequency width of the "Bragg" synchronism

If the synchronism conditions (11) are not exactly satisfied

$$\Delta_{p,q}=|p\gamma_1-q\gamma_2-\Delta|\neq 0,$$

then beats of the second-harmonic amplitude appear

$$|E_2|\infty\sin(\Delta_{p,q}z_L/2)/(\Delta_{p,q}/2).$$

The first maximum is reached at a distance

$$z_{\rm coh}^{(p,q)}=n_0\cos\theta\lambda/2\Delta_{p,q}.$$

The width of the synchronism relative to α can be estimated from the formula

$$\frac{\partial\Delta}{\partial\alpha}z_L(\Delta\alpha)_{\rm coh}=\pi.$$

We have

$$(\Delta\alpha)_{\rm coh}\approx n_0\lambda\cos\theta/2z\left(\frac{\partial\Delta_{p,q}}{\partial\alpha}\right)_s, \tag{22a}$$

where $\Delta_{1,-1}=|\gamma_1+\gamma_2-\Delta|$ for synchronism I and $\Delta_{0,1}=|\gamma_2-\Delta|$ for synchronism II. If the deviation from the Bragg position in the synchronism is not small $|\alpha_s|\gtrsim\chi_j$, then $(\Delta\alpha)_{\rm coh}\sim\lambda/z$. In the optical band at $\lambda\sim10^{-5}$ cm and $z\sim0.1$ cm we have $(\Delta\alpha)_{\rm coh}\sim10^{-4}$, in the x-ray band at $\lambda\sim10^{-8}$ cm and $z\sim0.1$ cm we have $(\Delta\alpha)_{\rm coh}\sim10^{-7}$.

If the synchronism takes place at small deviation $|\alpha_s|\ll\chi_j$, then $\partial\Delta_{p,q}/\partial\alpha\to0$ and the width of the synchronism increases by $\sim(\chi_j z/\lambda)^{1/2}$ times and is estimated from the formula

$$(\Delta\alpha)_{\rm coh}\approx(n_0\lambda\cos\theta/2z(\partial^2\Delta_{p,q}/\partial\alpha^2)_s)^{1/2}. \tag{22b}$$

For the first types of synchronism we find

$$(\Delta\alpha)_{\rm coh\,I}\approx(\chi_1\chi_2\lambda/z(\chi_1+\chi_2))^{1/2},\quad(\Delta\alpha)_{\rm coh\,II}\approx(\chi_2\lambda/z)^{1/2}.$$

If the fundamental radiation is monochromatic $\Delta\omega/\omega_1\ll(\Delta\alpha)_{\rm coh}$, then the angle width of the synchronism coincides with the width of the synchronism in α, i.e., $(\Delta\theta)_{\rm coh}\sim(\Delta\alpha)_{\rm coh}$. Analogously, if the fundamental radiation has a narrow angular spectrum $\Delta\theta_1\ll(\Delta\alpha)_{\rm coh}$, then the relative frequency width of the synchronism coincides with the synchronism width in α: $(\Delta\omega)_{\rm coh}/\omega\sim(\Delta\alpha)_{\rm coh}$.

When the fundamental radiation has simultaneously a broad frequency spectrum and a broad angular spectrum $(\Delta\omega_1/\omega_1,\ \Delta\theta_1>(\Delta\alpha)_{\rm coh})$, then the effects of spatial and temporal non-coherence can cancel each other in part, i.e., in the fundamental-radiation band there will be, for example, two pairs of values θ', ω', and θ'', ω'' such that

$$\alpha(\theta',\omega')=\alpha(\theta'',\omega'')=\alpha_s(\theta_s,\omega_s).$$

Let $\Delta\omega_1/\omega_1>\Delta\theta_1>(\Delta\alpha)_{\rm coh}$, then the frequency interval in which the deviation of α from α_s does not exceed the value $(\Delta\alpha)_{\rm coh}$, and synchronism SHG is possible, is estimated from the formula $(\Delta\alpha)_{\rm coh}\approx\omega_1\cot\theta\Delta\theta_1$.

The role of absorption

In the presence of absorption, all the Fourier components of the linear susceptibility and the parameters expressed in their terms are complex quantities whose real and imaginary parts will be respectively designated r and i. The synchronism condition (11) takes now the form

$$p\gamma_{1,r}-q\gamma_{2,r}=\Delta_r,\quad p=0,\pm1,\quad q=\pm1. \tag{23}$$

In an absorbing medium, the second-harmonic field in the case of synchronism (23) reaches a maximum at a distance

$$z_{\rm abs}^{(p,q)}=\frac{cn_0\cos\theta|\ln[(q\gamma_{2,i}+\chi_{2,0,i})/(\chi_{1,0,i}+p\gamma_{1,i})]|}{\omega|-p\gamma_{1,i}+q\gamma_{2,i}+\chi_{2,0,i}-\chi_{1,0,i}|}. \tag{24}$$

For synchronism II ($p=0$, $q=-1$) in the x-ray band (Si crystal, $\mathbf{h}=[220]$, $\lambda=0.71$ Å, π polarization) we obtain according to the data of Ref. 21 the estimate $z_{\rm abs}^{(0,-1)}\sim0.15$ cm.

SHG in Bragg diffraction

The excitation of the harmonic can occur in a periodic structure in the case of the Bragg diffraction as in Fig. 1b.

It is then necessary to solve Eqs. (5) with boundary conditions specified on the two surfaces of the sample of the medium

$$E_{1,0}|_{z=0}=E,\quad E_{1,h}|_{z=0}=0,\quad E_{1,h}|_{z=t}=0,\quad E_{2,2h}|_{z=t}=0,$$

where t is the thickness of the sample.

The solution obtained for the second-harmonic fields contain resonant terms, which indicate the presence of synchronism. Deferring a detailed analysis of the effects that occur here to later publications, we present only the formulas for the two synchronism conditions

$$\alpha_{s\,\rm I}=(\chi_1^2-\chi_2^2)/\Delta\ ({\rm I}),\quad\alpha_{s\,\rm II}=\Delta\pm2\chi_2\ ({\rm II}).$$

Synchronism I is realized in the case of a detuning α lying between the regions of reflection of the fundamental wave and the second-harmonic wave. Synchronism II occurs at values of α corresponding to the boundaries of the harmonic-reflection band.

In contrast to Laue diffraction, we can go here to the limit of one-dimensional propagation of all the waves along the reciprocal-lattice vector: $\theta=\pi/2$. If furthermore the reflection of the fundamental wave is forbidden ($\chi_h=0$, $E_{1,h}=0$), then synchronism II becomes ineffective and synchronism I becomes one-dimensional.[2,3]

4. CONCLUSION

Experiments on the realization of the described synchronisms can be performed in the infrared, visible, and ultraviolet bands with laser sources in periodic structures with $d\sim\lambda$ (see Ref. 15) and with sufficiently large modulation of the linear susceptibility $\chi_j\sim|\Delta|$ by virtue of (15)–(18), or else by substantial modulation of the nonlinear susceptibility $\beta_{2h}\neq0$ [see (15) and (16)].

The value of α at the synchronism, α_s, is determined by the quantities χ_j and Δ, which are constant for a given sample. The deviation of $\alpha(\theta,\omega)$ from $\alpha_s(\theta_s,\omega_s)$ can be due either to a change of angle $\delta\theta=\theta-\theta_s$, or to a change of frequency $\delta\omega=\omega-\omega_s$. Therefore, by varying the angle of incidence of the radiation on the medium, we can change the frequency ω at which the synchronism occurs. The range of possible smooth tuning of the harmonic frequency is estimated from the formula $\delta\omega/\omega \sim \delta\theta \cot\theta$ and should correspond to the tuning range of the fundamental radiation.

The nonlinearity in the x-ray band, is usually due to the nonlinearity of the free electrons and is smaller than in optics by ten orders of magnitude (see the Appendix). Therefore the SHG harmonic even when synchrotron radiation for pumping is small.[14] Cases are possible, however, when at least one of the frequencies lies in the resonance (nuclear or electronic), and then the nonlinear susceptibility can increase by 3–4 orders,[22] while the SHG intensity can increase by a factor equal to the square of this number.

In the present article the nonlinear interaction of the waves in the periodic medium in the case of Bragg diffraction was considered with the second-harmonic generation as the example. Equations (5), which describe the degenerate case of three-frequency interaction ($\omega_3 = \omega_1 + \omega_2$, $\omega_1 = \omega_2 = \omega$) can be easily generalized to the case of the nondegenerate interaction ($\omega_1 \neq \omega_2$). It must be borne in mind here that satisfaction of the Bragg condition for one of the frequencies does not automatically mean satisfaction of this condition for the other frequencies. The developed approach can be used also to describe nonlinear effects cubic in the field (third-harmonic generation, Raman scattering).

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APPENDIX

Calculation of the Fourier components of the nonlinear susceptibility in periodic structures, with account taken of the specifics of these structures, was carried out in the papers known to us only for crystals in the x-ray band.[8,12] Freund and Levin[8] were the first to obtain an expression for the convolution of the Fourier components of the quadratic susceptibility tensor of a crystal in the x-ray band ($\omega_3 = \omega_1 + \omega_2$) (see also Ref. 12):

$$\beta_h^{(3,1,2)}(\omega_3, \omega_1, \omega_2) = \mathbf{e}_3 \hat{\beta}_h(\omega_3, \omega_1, \omega_2)\mathbf{e}_1\mathbf{e}_2$$
$$= i\tau F(\mathbf{h})\left(\frac{c}{\omega_3}(\mathbf{e}_1\mathbf{e}_2)(\mathbf{h}\mathbf{e}_3) - \frac{c}{\omega_1}(\mathbf{e}_2\mathbf{e}_3)(\mathbf{e}_1\mathbf{h}) - \frac{c}{\omega_2}(\mathbf{e}_1\mathbf{e}_3)(\mathbf{e}_2\mathbf{h})\right), \qquad \text{(A.1)}$$

where $\tau = e^3/2m^2c\omega_1\omega_2\omega_3$ is the quadratic increment to the polarizability of the free electron, the structure factor is

$$F(\mathbf{h}) = \sum_l f_l(\mathbf{h}) e^{i\mathbf{h}\mathbf{r}_l},$$

and $f_l(\mathbf{h})$ is the factor of atomic scattering of the l-th atom in the unit cell.

Equation (A.1) was obtained by transforming the general expression for nonlinear polarization.[23] In the derivation of (A.1) it was assumed that the wave functions are real, the charge distribution is centrosymmetric, and the fields are transverse. In the present paper we are interested in a frequency-degenerate case, when $\omega_1 = \omega_2 = \omega$, $\omega_3 = 2\omega$.

It must be emphasized that centrosymmetric crystals (including silicon) have a nonzero pure imaginary Fourier component of the quadratic susceptibility β_h (when the reciprocal-lattice vector $\mathbf{h} \neq 0$), even though the zeroth Fourier components $\beta_0^{(0)}$ and $\beta_0^{(h)}$ (for which $\mathbf{h} = 0$) vanish for such crystals. The reason is that the periodicity of the medium gives rise to a preferred direction, which is precisely characterized by the reciprocal-lattice vector.

We present an example of the calculation of the convolution $\beta_h = \mathbf{e}_h \hat{\beta}_h \mathbf{e}_0 \mathbf{e}_h$ in accord with formula (A.1) at $\omega_1 = \omega_2 = \omega$, $\omega_3 = 2\omega$ in the simple case of a σ-polarized fundamental wave and a π-polarized second-harmonic wave. For this case $\mathbf{e}_1\mathbf{h} = \mathbf{e}_2\mathbf{h} = 0$, $\mathbf{e}_3\mathbf{h} = \mathbf{e}_h\mathbf{h} = h\cos\theta$. Substituting the scalar productions in (A.1), we get $\beta_h = i\tau F(\mathbf{h})\cos\theta\, hc/2\omega$, $\beta_{-h} = \beta_h^* = -\beta_h$. Recognizing that $hc/2\omega \approx \sin\theta$, we can write the expression for β_h also in the form

$$\beta_h \approx i\tau F(\mathbf{h}) \sin 2\theta.$$

Thus, $\beta_h \propto \sin 2\theta$ and vanishes when $\theta = \pi/2$ or $\theta = 0$; furthermore, if $\theta \neq \pi/2$ and $\theta \neq 0$, then $\beta_h \neq 0$ even in centrosymmetric crystals.

For the Si crystals $\mathbf{h} = [220]$, $2\mathbf{h} = [440]$ and the ratio of the structure factors is equal to the ratio of the atomic scattering factor $F(2\mathbf{h})/F(\mathbf{h}) = f(2\mathbf{h})/f(\mathbf{h}) \approx 2/3$.[21] The quantity $\beta_h/F(\mathbf{h})$ is proportional to the reciprocal-lattice vector h (exactly for a σ-polarized fundamental wave, and approximately so for a π-polarized pump). Therefore $\beta_{2h}/\beta_h \approx 4/3$. If we assume the atoms to be pointlike and the crystal to be monotonic, then $F(\mathbf{h}) = F(2\mathbf{h})$. In this case we have for a σ-polarized fundamental wave $\beta_{2h} = 2\beta_h$.

Formula (A.1) was calculated for one unit cell of a crystal. In estimates of the nonlinearity we must multiply (A.1) by the cell density, which is $\sim 10^{23}$ for a monatomic crystal. If the wavelength of the fundamental radiation is $\lambda = 2$ Å, i. e., $\omega \approx 10^{19}$ sec^{-1}, then (A.1) yields $|\beta_h| \sim 10^{-18}$ cgs esu. The nonlinearity increases rapidly with increasing frequency in proportion to $1/\omega^4$.

1) In an anisotropic crystal $\hat{\chi}$ is a tensor and $\chi_{-jh} = \mathbf{e}_0 \hat{\chi}_{-jh} \mathbf{e}_h$, $\chi_{jh} = \mathbf{e}_h \hat{\chi}_{jh} \mathbf{e}_0$.

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Resonance processes in a two-level system in the presence of nonresonance fields

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A two-level system acted on by resonance and non-resonance fields is considered. It is shown that in a nonstationary regime the effect of nonresonance fields is proportional to the first power of the ratios of the amplitudes of the nonresonance fields to their detunings relative to resonance and depends on the initial phases of the fields. In a stationary regime in a system with damping the effect of nonresonance fields depends on the level of the resonance field. The analysis is based on the solution of the Bloch equations by the method of averaging, up to the third approximation of this method.

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There are well known resonance[1-6] and nonresonance[3-5,7] effects which appear when high-frequency fields interact with a quantum system. In the present paper we consider the behavior of a two-level system in fields which include simultaneously resonance and nonresonance frequencies. The main attention will be given to effects that arise when resonance and nonresonance fields act together. Effects of this kind occur when classical oscillators interact with high-frequency fields.[8,9]

The analysis of a two-level system in fields is based on the solution of the Bloch equations by the method of averaging, up to the third approximation of this method. It is well known[6] that the Bloch equations describe the behavior of a magnetized assembly of spins in the case of magnetic resonance. These equations are also used in optics[2] (the optical Bloch equations) in the study of the interaction of light with a two-level system. In optics one introduces an auxiliary vector of a fictitious electric spin $s=(s_1, s_2, s_3)$, or a speudospin vector whose components s_1 and s_2 are associated with the dipole moment of the system, while the third component s_3 is associated with inversion of the atom.[2]

1. Let us consider a two-level system described by the Bloch equations

$$\begin{aligned}\dot{s}_1&=-\omega_0 s_2-s_1/T_2,\\ \dot{s}_2&=\omega_0 s_1+\varkappa F(t)s_3-s_2/T_2,\\ \dot{s}_3&=-\varkappa F(t)s_2-(s_3-s_p)/T_1,\end{aligned} \tag{1}$$

where ω_0 is the frequency of the transition between the levels, $\varkappa$ is the gyromagnetic ratio in the case of magnetic resonance, and in optics $\varkappa=2d/\hbar$ (d is the magnitude of the dipole matrix element), $F(t)$ is the strength of the high-frequency magnetic (or in optics the electric) field acting on the system, T_1 is the longitudinal relaxation time (in optics the damping or inversion time), T_2 is the transverse relaxation time (in optics this is the damping time for the dipole moment), s_p is the equilibrium value to which the magnetization (the inversion) relaxes in the presence of noncoherent pumping in the case $F(t)\equiv 0$. A dot denotes differentiation with respect to the time. In the theory of magnetic resonance Eqs. (1) correspond to the case of orientation of the high-frequency magnetic field perpendicular to the static field. Here the components s_i are the projections of the magnetization vector.

Let the system be acted on by external linearly polarized fields

$$F(t)=F_1\cos(\omega_1 t+\zeta_1)+F_2\cos(\omega_2 t+\zeta_2), \tag{2}$$

where the field with frequency ω_1 is a resonance field, i.e., $\omega_1\approx\omega_0$, and that with frequency ω_2 is a nonresonance field, i.e., $\omega_2\neq\omega_0$. Substituting Eq. (2) in the equations (1) and using the substitution

