Double optical three-wave mixing

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This paper discusses the potential usefulness of double optical three-wave mixing as a mechanism for dynamic holography with a signal wave that is incoherent with the reference wave. It is shown that double optical three-wave mixing can lead to an effective third-order nonlinearity that not only exceeds the orientational Kerr nonlinearity of condensed matter, but also has a relaxation time of 10^{-15} s. Double optical three-wave mixing also has the unique capability of measuring dielectric permittivity in the microwave and far-IR regions by purely optical methods.

An important difficulty with using the method of dynamic holography[1] for phase-conjugation is the requirement that coherence be maintained between the conjugated wave and at least one of the high-power reference waves. As is well-known, the problem here is that the relaxation time τ associated with the basic mechanisms for the third-order nonlinearity $\chi^{(3)}$, including electronic processes, considerably exceeds[2] the period of the optical oscillations, so that when the relative phase between the conjugated and reference wave contains irregular jumps, the dynamic hologram written by these two waves cannot track the corresponding sudden changes in the interference pattern. Because of this, these phase changes are averaged away into a spatially uniform background.

On the other hand, phase conjugation via four-wave mixing allows the conjugation of weak signals from distant objects with a reflection coefficient that is practically independent of the energy of the signal pulse, an extremely attractive feature for various applications. Therefore, efforts to overcome these problems with coherence are currently of great interest.

The simplest and most natural escape from this predicament is to look for nonlinear mechanisms and specific materials for which $\chi^{(3)}$ has a suitably short relaxation time.

We have already noted that even the "fastest" mechanisms for $\chi^{(3)}$, i.e., nonlinearities due to anharmonicity of electronic oscillations in molecules and atoms, are as a rule very weak ($\chi^{(3)}\approx 10^{-15}$ cgs), and even these are quite slow ($\tau\approx 10^{-14}$ s).

On the other hand, the second-order nonlinearities $\chi^{(2)}$, which are responsible for processes like frequency (three-wave) mixing and second harmonic generation, possess *a priori* relaxation times that are smaller or on the order of the optical period.

Therefore, it is not surprising that attempts have been made[3–6] to use these nonlinearities for phase conjugation. However, the structure of the correction to the displacement $\mathbf{D}^{NL}$ due to $\chi^{(2)}$ is such that it is impossible to obtain a term with the "correct" space-time dependence from the point of view of phase conjugation, i.e., $\mathbf{E}_3^* \exp(-i\mathbf{k}_3\mathbf{r} - i\omega_3 t)$, for simple three-wave mixing with an arbitrary wave $\mathbf{E}_1$ (in this paper the amplitudes $\mathbf{E}$, frequencies ω, and wave vectors $\mathbf{k}$ are labeled in the way that is standard for four-wave mixing: 1, 2 are reference waves, 3 is the signal and 4 is the conjugated wave). In fact, the corresponding terms in the displacement have the form $\chi^{(2)}E_3^*E_1 \exp[i(\mathbf{k}_1 - \mathbf{k}_3)\mathbf{r} - i(\omega_1 - \omega_3)t]$, from which it is clear that phase conjugation requires that $\omega_1 = 2\omega$ along with $\mathbf{k}_1 = 0$, which is impossible. For this reason, the authors of Refs. 3–6 considered pseudo-phase conjugation, in which a wave with the phase-conjugated $\mathbf{E}_3$ wave front propagates in a direction which is not $-\mathbf{k}_3$, but which is subsequently corrected by a linear mirror. Because the quality of phase conjugation was correspondingly poor, further development of this method was not pursued.

However, there is one other possibility for using $\chi^{(2)}$ for phase conjugation, in an interaction geometry which is practically identical to four-wave mixing: double three-wave mixing with the participation of an intermediate wave $\mathbf{E}_s \sim \mathbf{E}_3^*\mathbf{E}_1$, which is generated by the original waves. The task of this paper will be to investigate the possibility of realizing such a process and to establish how competitive it is with traditional four-wave mixing caused by $\chi^{(3)}$.

POSTULATED INTERACTION PHENOMENOLOGY

Let us consider the wave interaction geometry shown in Fig. 1, bearing in mind that the interaction is via a quadratic nonlinearity $\chi^{(2)}$. Outwardly, the interaction geometry is practically identical to nondegenerate four-wave mixing: there are two reference waves $\mathbf{E}_{1,2}^* \exp(i\mathbf{k}_{1,2}\mathbf{r} - i\omega_{1,2}t)$, a signal wave $\mathbf{E}_3^* \exp(i\mathbf{k}_3\mathbf{r} - i\omega_3 t)$, and a conjugate wave $\mathbf{E}_4^* \exp(i\mathbf{k}_4\mathbf{r} - i\omega_4 t)$. It is assumed that the interaction actually takes place in two stages. First of all, the waves $\mathbf{E}_{1,3}$ give rise to a field $\mathbf{E}_s \sim \chi^{(2)}E_3^*E_1 \exp(i\mathbf{k}_s\mathbf{r} - i\omega_s t)$ via the process of difference frequency generation, where $\mathbf{k}_s = \mathbf{k}_1 - \mathbf{k}_3$, $\omega_s = \omega_1 - \omega_3$. Next, the field $\mathbf{E}_s$ participates in the process of sum frequency generation with the wave $\mathbf{E}_2$ to give a polarization $\mathbf{P}_s \sim \chi^{(2)}\mathbf{E}_2\mathbf{E}_s \sim \mathbf{E}_3^*\mathbf{E}_1\mathbf{E}_2 \exp[i\mathbf{q}_4\mathbf{r} - i\Omega_4 t]$. For this polarization to give rise to a wave $\mathbf{E}_4$ that is conjugate to $\mathbf{E}_3$, it is necessary that $\mathbf{q}_4 = \mathbf{k}_4 = -\mathbf{k}_3$, $\Omega_4 = \omega_3 = \omega$.

In order to fulfill these conditions it is necessary to impose the following restrictions on the frequency and wave vectors of the reference waves:

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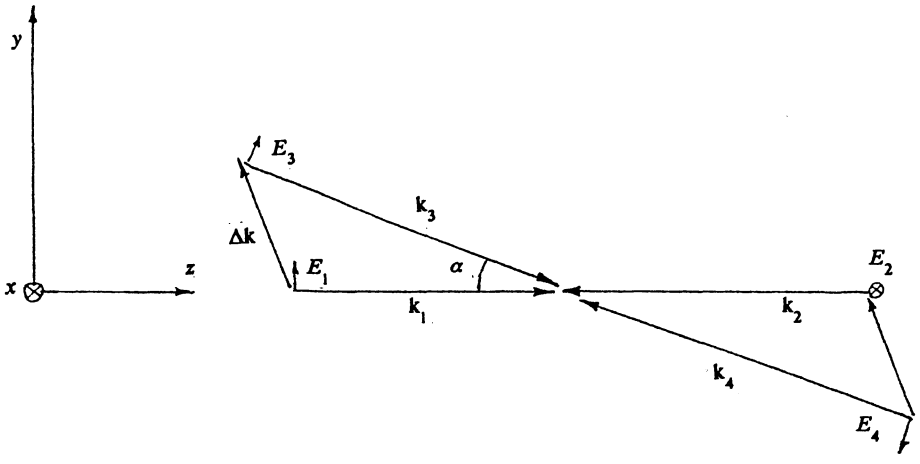

FIG. 1.

$$\omega_1=\omega+\Delta, \quad \omega_2=\omega-\Delta, \quad \mathbf{k}_1=-\mathbf{k}_2. \tag{1}$$

Here Δ is a free parameter equal to the frequency difference of waves $\mathbf{E}_1$ and $\mathbf{E}_3$. A reasonable choice of Δ can be made by starting from the following considerations. First of all, when $\mathbf{E}_1$ and $\mathbf{E}_3$ are incoherent, which is the case of primary interest, the quantity Δ has incoherence on the order of the Γ-linewidth of the signal $\mathbf{E}_3$. Therefore, it is necessary to choose $\Delta \geqslant (2\text{–}3)\Gamma$ in order to avoid the strong dispersion of $\chi^{(2)}$ near zero frequency Δ, which can lead to instability of the $\mathbf{E}_s$ oscillation. On the other hand, we need to fulfill the condition $k_1(\omega+\Delta)=k_2(\omega-\Delta)$. A natural possibility is to use waves $\mathbf{E}_{1,2}$ with different polarizations, in which case the maximum constant shift that is admissible for a given crystal is $\Delta=(\omega\Delta n)/(2n)$ (where $n_{\parallel,\perp}=(\varepsilon_{\parallel,\perp})^{1/2}$ are the principal values of the indices of refraction of the crystal, and Δn is its anisotropy). This implies $\Gamma \ll \Delta \ll \omega\Delta n/2n_\perp$. Among real crystals that possess rather large $\chi^{(2)} \approx 10^{-8}$–$10^{-7}$ cgs, the maximum $\Delta n/n_\perp \approx 0.05$ is observed in lithium niobate and perovskites similar to it.

When a Nd–YAG laser is used as the light signal to be conjugated ($\Gamma=6\ \text{cm}^{-1}$) these restrictions lead to the following condition: $1.1\cdot 10^{12}\ \text{s}^{-1} \ll \Delta \ll 4.7\cdot 10^{13}\ \text{s}^{-1}$, which corresponds to generation of an intermediate wave $\mathbf{E}_s$ in the far-IR and submillimeter region (with wavelength $\lambda_s \approx 50$–$100\ \mu\text{m}$).

Conditions (1) ensure phase-synchronous generation of the wave $\mathbf{E}_4$ conjugate to $\mathbf{E}_3$ in the interaction region. However, there is one other "internal" condition for synchronism for this two-stage process, whose fulfillment rigorously determines the efficiency of the process. Specifically, if the first stage of the process, i.e., generation of $\mathbf{E}_s$, is itself synchronous, an additional enhancement factor appears in the efficiency for double optical three-wave mixing, of order $(\alpha/\lambda_s)^2$ compared to the nonsynchronous generation of $\mathbf{E}_s$, where α is the size of the interaction region in the direction of propagation of the latter. Bearing in mind the need to take this synchronism into account, let us turn to quantitative consideration of the various types of double optical three-wave mixing.

DOUBLE OPTICAL THREE-WAVE MIXING WITH A NONSYNCHRONOUS FIRST STAGE

Let us therefore consider the interaction shown in Fig. 1, assuming that the difference frequency generation is nonsynchronous, i.e., $|\Delta\mathbf{k}|=|\mathbf{k}_1-\mathbf{k}_3| \neq |\mathbf{k}_s|=\varepsilon_s^{1/2}\Delta/c$. Here ε_s is the dielectric permittivity of the crystal at the frequency Δ. For the moment we will ignore the polarization structure of the wave $\mathbf{E}_s$, which is determined by the properties of the tensor $\hat{\chi}_-^{(2)}$, as it is unimportant for this case.

The Helmholtz equation for the wave $\mathbf{E}$ has the form

$$-\Delta E_s-\frac{\varepsilon_s\Delta^2}{c^2}E_s=2\pi\Delta^2c^{-2}\chi_-^{(2)}E_s^*E_1\exp(i\Delta\mathbf{kr}) \tag{2}$$

So as to not encumber the discussion, we limit ourselves to the Born approximation, for which $\mathbf{E}_3=\text{const}$. The problem becomes two-dimensional when this restriction is lifted, since $\Delta\mathbf{k}$ has y and z components in the geometry of Fig. 1. Furthermore, as we will see below, it becomes very onerous, without leading to any qualitatively new conclusions for the nonsynchronous case. If we consider only the particular solution to the inhomogeneous equation, which dominates within the interaction region, we have

$$E_s=\frac{2\pi\Delta^2}{c^2(\Delta k^2-k_s^2)}\chi_-^{(2)}E_s^*E_1\exp(i\Delta\mathbf{kr}). \tag{3}$$

Substituting this expression into the truncated Helmhotz equation for the wave $\mathbf{E}_4$ gives

$$\partial\frac{E_4}{\partial z}=-\frac{i\pi\omega^2}{2k_4c^2\cos\alpha}\chi_+^{(2)}E_sE_2=\frac{i\pi\omega^2}{2k_4c^2\cos\alpha}\kappa E_1E_2E_3^*, \tag{4}$$

$$E_4(L)=0, \quad \kappa=\frac{2\pi\Delta^2}{c^2(\Delta k^2-k_s^2)}\chi_+^{(2)}\chi_-^{(2)}.$$

The boundary condition corresponds to the usual statement that there is no conjugate wave at the input to the medium.

Equation (4) clearly implies that, formally, nonsynchronous double three-wave mixing in the Born approximation is completely equivalent to an effective four-wave mixing in a medium with a cubic nonlinearity $\chi^{(3)}=\kappa$. Let us estimate the achievable magnitude of this effective nonlinearity κ. For the maximum achievable Δ, angles of intersection of waves $\mathbf{E}_{1,3}$ in the crystal $\alpha\geqslant 10^{-2}$ rad, and $\chi^{(2)}_{+,-}\approx 5\cdot 10^{-8}$ cgs (the values are taken for lithium niobate from Ref. 7), it is easy to obtain $\kappa\approx 2\pi\chi_+\chi_-/[(2n_\perp^2\,\alpha/\Delta n)-\varepsilon_s]\approx 3\cdot 10^{-15}$ cgs. Thus, the effective cubic nonlinearity of nonsynchronous double optical three-wave mixing is quite comparable in size with electronic cubic nonlinearities of condensed matter[2] and has roughly the same relaxation time.

We have not established why double optical three-wave mixing with a nonsynchronous first stage is of any interest; we could easily interpret it as an additional mechanism for rapid electronic $\chi^{(3)}$ taking place in a noncentrosymmetric medium. Let us now discuss the possibility of implementing double optical three-wave mixing with a synchronous first stage.

DOUBLE OPTICAL THREE-WAVE MIXING WITH A SYNCHRONOUS FIRST STAGE

Let us first consider the criterion for synchronous generation of the intermediate wave $\mathbf{E}_s$, i.e., equality of $\Delta\mathbf{k}$ and $\mathbf{k}_s$, in more detail. If the medium were dispersionless $[\hat{\varepsilon}_s=\hat{\varepsilon}(\omega)=\hat{\varepsilon}(\omega-\Delta)]$, then because of the way the wave frequencies for $\mathbf{E}_{1,2,3}$ are related, synchronism would be possible only for strictly collinear propagation of all three waves. Accordingly, in a medium with normal dispersion $[\varepsilon_s<\varepsilon(\omega-\Delta)<\varepsilon(\omega)]$ synchronism is impossible in general for three waves of the same polarization type. Starting from the requirement $\mathbf{k}_1=-\mathbf{k}_2$, for a crystal with $\varepsilon_a<0$ (this includes practically all crystals with large $\chi^{(2)}$) the wave $\mathbf{E}_1$ should be **e**-type while wave $\mathbf{E}_2$ should be **o**-type. Accordingly, if wave $\mathbf{E}_3$ is **o**-type, then synchronism in a medium with normal dispersion is once more impossible ($|\mathbf{k}_s|+|\mathbf{k}_1|<|\mathbf{k}_3|$). The only remaining possibility is for waves $\mathbf{E}_{1,3}$ to be **e**-type and to generate a wave $\mathbf{E}_s$ of **o**-type. However, in order to implement double optical three-wave mixing in the geometry of Fig. 1 with this combination of polarizations for the waves, we require nonzero values of the components $\chi^{(2)}_{yxx}$ and $\chi^{(2)}_{xyy}$ simultaneously, which is possible only for crystals of triclinic syngony and monoclinic syngony of class m (see Ref. 7). Unfortunately, media with these crystal structures usually do not have refractive indices with sufficiently large anisotropy and quadratic nonlinearity; therefore, this type of synchronism is not very promising.

Nevertheless, there is one remaining possibility for achieving synchronous double optical three-wave mixing. The method, which is quite realistic, is in principle identical with synchronous third harmonic generation in gases. That is, suppose that all three waves $\mathbf{E}_{1,2,3}$ are **e**-polarized but that there are one or several absorption lines (bands) between the frequencies ω and Δ. In view of the large difference between ω and Δ ($\lambda_s\geqslant 50$ μm, while $\lambda\simeq 1.06$ μm), this situation is not at all unusual. When it does occur, the relation $\varepsilon_s>\varepsilon(\omega)\simeq\varepsilon(\omega-\Delta)$ often holds, and synchronism is entirely possible. On the other hand, in this geometry the requirement that certain components of $\chi^{(2)}$ be nonzero is considerably weakened [it is enough for any of the following components to be "nonzero": ($\chi^{(2)}_{yyy}$ or $\chi^{(2)}_{zyy}$) + ($\chi^{(2)}_{yyx}$ or $\chi^{(2)}_{zyx}$)], and the class of admissible symmetry types is considerably enlarged: in addition to the two classes mentioned above, the trigonal (class 3 or 3 m), tetragonal (4 or 4 mm), and hexagonal classes (6 or 6 mm) are also admissible, so that the class of crystals with suitable symmetry and satisfying the additional condition $\varepsilon_s>\varepsilon(\omega)$ now includes such well-known nonlinear crystals as proustite, KDP, and lithium niobate and iodate. Therefore, we will pause to discuss this situation in more detail.

Synchronism is observed for very specific angles α (and the angles β that corresponding to them; see Fig. 2) between the waves $\mathbf{E}_s$ and $\mathbf{E}_1$ ($\beta\approx\alpha\varepsilon_\parallel\,\omega/\varepsilon_{s\parallel}\,\Delta$). In explicit form, the conditions for synchronism have the following form:

$$\begin{aligned}&\cos\alpha-\xi^{1/2}\nu\cos\beta=1-\nu,\\&\sin\alpha=\xi^{1/2}\nu\sin\beta.\end{aligned}\tag{5}$$

Here $\nu=\Delta/\omega$, $\xi=\varepsilon_{s\parallel}/\varepsilon_\parallel$, and the small difference between $\varepsilon_\parallel(\omega)$ and $\varepsilon_\parallel(\omega-\Delta)$ has been neglected. Solving system (5) gives

$$\begin{aligned}&\sin(\alpha/2)=\nu(\xi-1)^{1/2}(1-\nu)^{-1/2}/2\quad\text{for }\nu\ll 1,\\&\alpha=\nu(\xi-1)^{1/2},\quad\beta=(\xi-1)^{1/2}.\end{aligned}\tag{6}$$

As we should expect, for $\xi=1$ (the dispersionless case) we have $\alpha\equiv\beta\equiv 0$. That is, the intermediate wave $\mathbf{E}_s$ can propagate both "along" the direction of the primary waves and "transverse" to them. The difference between these two cases is quite important: in view of the real geometry of the interaction region (i.e., the length L transverse to the dimension l) the problem is one-dimensional for $L\tan\beta\ll l$, and the boundary condition on $\mathbf{E}_s$ is imposed at the boundary of the nonlinear medium, while for $L\tan\beta\gg l$ the problem is two-dimensional and this boundary condition is imposed along one of the "lateral" boundaries of the interaction region. Therefore we will consider these two limiting cases separately.

SYNCHRONOUS DOUBLE OPTICAL THREE-WAVE MIXING WITH "LONGITUDINAL" INTERMEDIATE WAVES

The system of truncated Helmholtz equations for wave amplitudes $\mathbf{E}_4$, $\mathbf{E}_s$ in the Born approximation ($\mathbf{E}_3=$const) has the form

$$\begin{aligned}&\partial\frac{E_4}{\partial z}=-\frac{i\pi\omega}{c\varepsilon_\parallel^{1/2}\cos\alpha}\chi^{(2)}_+E_sE_2,\\&E_4(0)=E_3(0)=0,\\&\partial E_s\partial z=-\frac{i\pi\Delta}{c\varepsilon_{s\parallel}^{1/2}\cos\beta}\chi^{(2)}_-E_1E_3^*.\end{aligned}\tag{7}$$

The boundary conditions correspond to absence of the waves $\mathbf{E}_{4,s}$ at the input of the interaction region. From (7), by repeated differentiation of the first equation it is easy to obtain

$$\partial^2 \frac{E_4}{\partial z^2} = -\frac{\pi^2 \omega \Delta}{c^2 \varepsilon_{\parallel}^{1/2} \cos\alpha \varepsilon_{s\parallel}^{1/2} \cos\beta} \chi_+^{(2)} \chi_-^{(2)} E_1 E_2 E_3^*, \qquad (8)$$

$$E_4(0) = \partial E_4/\partial z|_{z=0} = 0,$$

so that for the amplitude of the wave $\mathbf{E}_4$ at the output of the medium we finally have

$$\mathbf{E}_4(L) = -\frac{\pi^2 \omega \Delta L^2}{2c^2 \varepsilon_{\parallel}^{1/2} \cos\alpha \varepsilon_{s\parallel}^{1/2} \cos\beta} \chi_+^{(2)} \chi_-^{(2)} E_1 E_2 E_3^*$$

$$= \frac{i\pi\omega^2 L}{2k_4 c^2 \cos\alpha} \kappa_1 E_1 E_2 E_3^*, \qquad (9)$$

$$\kappa_1 = \frac{i\pi\Delta^2 \chi_+^{(2)} \chi_-^{(2)}}{c^2 k_s^2 \cos\beta} (k_s L).$$

Here κ_1 has the sense of an equivalent effective cubic nonlinearity for the synchronous double optical three-wave mixing. A comparison of Eqs. (3) and (4) for nonsynchronous double optical three-wave mixing under the assumption $(\Delta k)^2 - k_s^2 \geqslant k_s^2$ reveals an enhancement factor for the synchronous case $\approx (k_s L) \approx 1200$ for $L = 1$ cm, so that the effective $\chi^{(3)}$ for synchronous double optical three-wave mixing $\chi_{\text{eff}}^{(3)} \approx 10^{-11}$ cgs, which is quite comparable to the strain-induced Kerr nonlinearity (stimulated Raman scattering) with three orders of magnitude better relaxation time.

These orders of magnitude indicate that rather large values of $R^{NL} = |E_4(L)/E_3(L)|^2$ are possible in principle for, say, picosecond pumping; therefore it is expedient to discuss this case as well.

The equations and boundary conditions for waves $\mathbf{E}_{4,3,s}$ have the form

$$\partial E_s/\partial z = -ia_s(E_1 E_3^* + E_4 E_2^*), \quad E_s(L) = 0,$$

$$\partial E_4/\partial z = iaE_s E_2, \quad E_4(0) = 0, \qquad (10)$$

$$\partial E_3^*/\partial z = iaE_s E_1^*, \quad E_3^*(L) = \text{const}.$$

Here the pump waves $\mathbf{E}_{1,2}$ are assumed to be traveling waves $a_s = \pi\Delta^2 \chi_-^{(2)}/c^2 k_s \cos\beta$; $a = \pi\omega^2 \chi_+^{(2)}/c^2 k_4 \cos\alpha$, and the boundary conditions correspond to absence of the waves $\mathbf{E}_{4,s}$ and a fixed amplitude of the waves $\mathbf{E}_3$ at the input to the medium.

The system (10) is easily reduced to the following equation for the wave $\mathbf{E}_4$ we are interested in:

$$\partial^3 E_4/\partial z^3 = b^2 \partial E_4/\partial z, \quad E_4(0) = 0; \quad \partial E_4/\partial z|_{z=L} = 0,$$

$$(\partial^2 E_4/\partial z^2 - aa_s|E_2|^2)_{z=L} = aa_s E_1 E_2 E_3^*(L), \qquad (11)$$

$$b^2 = aa_s(|E_1|^2 + |E_2|^2).$$

The solution to this system has the form:

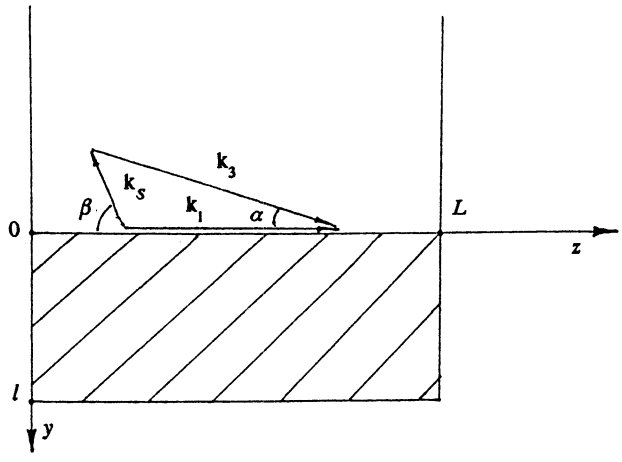

FIG. 2.

$$E_4(z) = \frac{E_1 E_2 E_3^*(L)}{|E_1|^2 + |E_2|^2 \text{ch}(bL)} [\text{ch}(b(L-z)) - \text{ch}(bL)] \qquad (12)$$

and for $L = z$ for the wave $\mathbf{E}_4(z = L)$:

$$E_4(L) = \frac{E_1 E_2 E_3^*(L)}{|E_1|^2 + |E_2|^2 \text{ch}(bL)} [1 - \text{ch}(bL)] \qquad (13)$$

For small bL, Eq. (13) corresponds exactly to the Born asymptotic form (9). For sufficiently large bL it is clear that (13) differs qualitatively from the corresponding expression for four-wave mixing (see, e.g., Ref. 8). For example, for real b (i.e., $\chi_{+,-}^{(2)}$ the same sign) there is no threshold for parametric oscillation; for large bL $R^{NL} \to R/(1+R)^2$, where $R = |\mathbf{E}_1|^2/|\mathbf{E}_2|^2$ is the ratio of intensities of the reference waves. Note especially the extremely steep dependence of the intensity of the wave $\mathbf{E}_4$ on the interaction length L (fourth power in the Born approximation).

SYNCHRONOUS DOUBLE OPTICAL THREE-WAVE MIXING WITH "TRANSVERSE" INTERMEDIATE WAVES

We now consider the case $L \tan\beta \gg l$, i.e., a "transversely" propagating intermediate wave $\mathbf{E}_s$. In this case, the geometry of the interaction region becomes extremely important, i.e., the boundary conditions on the wave $\mathbf{E}_s$ are imposed at the "lateral" boundaries of the region under discussion. For simplicity we consider the bounded two-dimensional interaction region (see Fig. 2) $0 \leqslant z \leqslant L$, $0 \leqslant y \leqslant l$. The system of equations that describes double optical three-wave mixing in this case is the following:

$$\partial E_s/\partial y = -ia_s(E_1 E_3^* + E_4 E_2^*), \quad E_s(0,z) = 0,$$

$$\partial E_4/\partial z = iaE_s E_2, \quad E_4(y,0) = 0, \qquad (14)$$

$$\partial E_3^*/\partial z = iaE_s E_1^*, \quad E_3^*(y,L) = \text{const}.$$

In contrast to (10), here it is not $\cos\beta$ but rather $\cos(\pi/2-\beta)$ that appears in the definition of α_s. In the Born approximation (i.e., neglecting the second term on the right hand side of the first equation of (14) and setting $E_3^*\equiv$const) the solution for $\mathbf{E}_4$ has the form

$$\mathbf{E}_4=-aa_sE_1E_2E_3^*zy. \tag{15}$$

Naturally, by virtue of the explicit dependence on y a field of this form is not strictly conjugate to $\mathbf{E}_3$. However, the corresponding change in the divergence is rather weak ($\sim\lambda/l$), and we will not discuss it here. In this case the nonlinear reflection coefficient for double optical three-wave mixing is not uniform in the transverse cross section of the primary beams ($\mathbf{E}_{1,2,3,4}$); its average value is given by

$$\bar{R}^{NL}=l^{-1}\left(\int_0^l|E_4(L)|^2dy\right)/|E_3|^2. \tag{16}$$

For comparison with the previous results it is convenient to use (16) to calculate the quantity $(R^{NL})^{1/2}\cdot|E_3|=\langle|E_4|\rangle$ (by this we understand the mean-square value of the absolute value):

$$\langle|E_4(L)|^2\rangle=\frac{\pi\omega^2L}{k_4c^2\cos\alpha}\kappa_2|E_1E_2E_3^*|,$$
$$\kappa_2=\frac{\pi\Delta^2\chi_+^{(2)}\chi_-^{(2)}}{3c^2k_s^2\cos\beta}(k_sl). \tag{17}$$

It is clear that this effective cubic susceptibility κ_2 for four-wave mixing, which is energetically equivalent to the double optical three-wave mixing under discussion, exceeds that for "nonsynchronous" double optical three-wave mixing by a factor of $\sim(k_sl)/3$. Although in a typical experimental situation ($l<L$) this enhancement factor is smaller than for the case $\tan\beta\ll l/L$, it nevertheless allows us to achieve an effective $\chi^{(3)}\approx3\cdot10^{-12}$ cgs, which also merits attention.

The dependence of $\langle|E_4|\rangle$ on L is the usual one for four-wave mixing in this case; however, the dependence on the width of the beam l should differ from the ordinary case, where it is due to the power density of the unchanging pulse energy.

As for the case $R^{NL}\sim1$ [Eqs. (14)], it can be reduced by a Laplace transform with respect to y and the substitutions $E_4=E_2S$, $E_3^*=E_1V$ to the following equation for S:

$$\partial^2s/\partial z^2+(b^2/p)\partial s/\partial z=0,\quad s(0)=0,$$
$$(\partial s/\partial z+aa_s|E_2|^2s)_{z=L}=-aa_s|E_1|^2E_3^*(L)/p. \tag{18}$$

Here $s=L(S)$, p is the argument of the Laplace transform, and $b^2=aa_s(|E_1|^2+|E_2|^2)$, as previously. The system (18) is solved by standard methods, and we have for the Laplace transform $e_4=L(\mathbf{E}_4)$ at the output of the interaction region the expression

$$e_4(L)=-\frac{E_3^*(L)}{p}\frac{\exp(-b^2L/p)-1}{\exp(-b^2L/p)+R}. \tag{19}$$

The inverse transform of this expression is extremely cumbersome and uninformative (it is the sum of a slowly converging series in $I_0(\{inbLy\}^{1/2})$, where I_0 is the modified Bessel function and n labels the term). Therefore, we will give no results for (numerical) calculations of $\bar{R}^{NL}$ here, because they add nothing qualitatively new to the results of the Born approximation.

DOUBLE OPTICAL THREE-WAVE MIXING AS A WAY TO MEASURE $\hat{\varepsilon}_s$

Up to now we have considered double optical three-wave mixing in terms of its possible usefulness for phase conjugation of the wave $\mathbf{E}_3$. However, there is still another rather attractive application for this phenomenon. Specifically, ε_s is unambiguously specified by the angle α at which synchronism is observed for the first stage of double optical three-wave mixing, and in the geometry discussed above (Fig. 1) $\varepsilon_\parallel(\omega)$ is likewise a function only of α and the known ratio Δ/ω from Eq. (6). Thus, double optical three-wave mixing provides a way to measure ε_s in the far-IR and submillimeter regions without the necessity of measuring radiation at the frequency Δ. The possibility of a purely optical measurement of ε_s is very attractive in view of the absence of reliable sources and detectors of radiation in these frequency regions; therefore, we will pause to discuss it in more detail.

First of all, let us specify the range of Δ within which it is possible to measure ε_s. In our earlier discussion, when the waves $\mathbf{E}_{1,3}$ were mutually incoherent and the condition $\mathbf{k}_1=-\mathbf{k}_2$ was required for synchronism of the second stage of double optical three-wave mixing, this region was $\Gamma\ll\Delta\ll(\omega\Delta n/2n_\perp)$.

However, there is no need to work with incoherent waves $\mathbf{E}_{1,3}$ in order to measure ε_s; therefore, the lower bound of the range of Δ that allows such measurements is determined by the requirement $k_sl\geqslant1$, which is necessary for the appearance of a synchronous peak in the angular dependence $|E_4|^2(\alpha)$. Thus, a sufficient requirement is $\lambda_s<l$ (for "transverse" synchronism of the first stage of double optical three-wave mixing, which is the more likely case).

Furthermore, if the goal is to measure ε_s, we can drop the requirement for synchronism of the second stage of double optical three-wave mixing; therefore, the limitation $\lambda_s>(2n_\perp/\Delta n)\lambda$ becomes superfluous.

Actually, if $|\mathbf{k}_1|\neq|\mathbf{k}_2|$ the signal amplitude will be weaker by a factor of $\sin(\Delta kL/2)/(\Delta kL/2)$, where $|\Delta k|=|\mathbf{k}_1|-|\mathbf{k}_2|$ in the geometry of Fig. 1. However, this weakening is independent of α; therefore, the peak corresponding to synchronism of the first stage of double optical three-wave mixing will be quite easy to observe when the attenuation of $\mathbf{E}_4$ is not too large, so that this signal is detectable.

In particular, lifting of the requirement $\mathbf{k}_2 = -\mathbf{k}_1$ implies that, in addition to the polarization combination of waves discussed above, interactions $E_1 E_3^* \rightarrow E_s$ of the type $oo \rightarrow o$, $oo \rightarrow e$, and $E_s E_2 \rightarrow E_4$ with arbitrary polarization combinations of the waves can be realized. This greatly enlarges the class of admissible crystal symmetries and allows us to separately measure $\varepsilon_{s\parallel}$ and $\varepsilon_{s\perp}$ in the region of λ_s from millimeter wavelengths out to ≈ 20 μm.

Thus, double optical three-wave mixing has real potential for realizing dynamic holography using signal waves that are incoherent with the reference wave. On the other hand, double optical three-wave mixing can be effectively used as a purely optical instrument for spectroscopy of the dielectric permittivity tensor of noncentrosymmetric crystals in the difficult-to-study submillimeter and far-IR regions.

Therefore, an experimental demonstration of these phenomena would undoubtedly be interesting.

*In this paper $\chi^{(n)}$ is everywhere defined as a quantity that connects the real polarization and the field corresponding to the complex amplitudes $P^{(n)} = 2^{(1-n)} \chi^{(n)} E^n$.

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