RESONANCE PHENOMENA IN NONLINEAR OPTICS

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Submitted to JETP editor September 21, 1964

J. Exptl. Theoret. Phys. (U.S.S.R.) 48, 931-938 (March, 1965)

A study is made of the interaction between light waves in a continuous medium when there is a resonance between multiples of the frequencies of the light waves and the frequencies of electromagnetic transitions of the medium. The nonlinear effect of generation of a third harmonic is treated for the case of passage of monochromatic radiation through a medium which has a resonance for the second harmonic. The conditions are stated under which a sharp increase of the intensity of the third harmonic is to be expected. It is shown that in a strong light field and in the case of spatial matching [$k(3\omega) = 3k(\omega)$, where k is the wave vector of the light wave] the nonlinear absorption is extraordinarily small.

1. INTRODUCTION

In linear electrodynamics there is the well known phenomenon of anomalous dispersion, which occurs in cases in which the frequency of the electromagnetic field is close to one of the characteristic frequencies of the medium. In this case both the real and the imaginary parts of the dielectric permeability tensor are much larger than their values far from the resonance. The elucidation of analogous properties in the nonlinear terms of the dielectric permittivity is of considerable interest, in particular in connection with the problem of the generation of multiple frequencies of light, for which the choice of material to a large extent determines the possibility of observing nonlinear optical effects. If the medium in question has a discrete energy spectrum, then the nonlinear terms in the permittivity increase sharply when the frequency ω of the beam of light, or a multiple $n\omega$ of this frequency, coincides with one of the characteristic frequencies of the medium. Not all of these resonances are equivalent, however, from the point of view of a maximum value of the intensity of the harmonic generated. In a previous paper by one of the writers[1] it was shown that a resonance with the fundamental or a generated harmonic is in no way distinguished as compared with a nonresonance situation.

In fact, as one comes closer to the resonance (which can be done either by changing the frequency of the incident light or by changing the energy levels with external static fields) there is on one hand a sharp increase of the corresponding nonlinear term in the permittivity, but on the other hand there is a sharp increase of the linear term in the permittivity which is responsible for absorption of the fundamental or the higher optical harmonic, which in itself leads to a decrease of the intensity of the harmonic generated. The analysis shows[1] that these two processes compensate each other, so that there is practically no appreciable change in the intensity. Precisely this situation has been observed experimentally.[2,3]

If, however, there is a resonance with an intermediate frequency (for example, in experiments on the production of the third harmonic, a resonance in the corresponding nonlinear term of the permittivity can occur with the doubled frequency[4]), so that it is not a resonance for the ordinary linear part of the permittivity in the vicinity of the frequency of the incident light or near that of the generated light, the situation is very different. As we come near a resonance of this type the intensity of the generated harmonic begins to increase sharply. In this sense substances with energy-level systems that provide resonances with an intermediate multiple frequency are markedly distinguished relative to other materials from the point of view of efficient transformation of one frequency of light into another.

The present paper is devoted to a study of nonlinear optical effects under conditions of resonance with an intermediate frequency. In particular, we consider the problem of the generation of light waves of frequency 3ω when light of frequency ω passes through a medium which has characteristic transition frequencies which are in resonance with the frequency 2ω. In Sec. 2 we examine the properties of the nonlinear terms in the dielectric process which are decisive for the generation of a third harmonic under resonance conditions. Section 3 contains the derivation of the nonlinear equations that describe the phenomena in question.

Section 4 is devoted to the solution of these equations for weak light waves. In Sec. 5 we study the question of the maximum possible transformation of the fundamental frequency into the third harmonic for strong light waves, and also analyze the mechanisms which limit this process, such as two-photon absorption, and so on.

2. THE NONLINEAR DIELECTRIC POLARIZATION

To terms of third order in the electric field the dielectric polarization vector is of the form

$$D_\alpha(t) = D_\alpha^{(1)} + D_\alpha^{(2)} + D_\alpha^{(3)}, \tag{1}$$

$$D_\alpha^{(1)} = \int_{-\infty}^{\infty} d\omega \varepsilon_{\alpha\beta}^{(1)}(\omega) E_\beta(\omega) \exp\{-i\omega t\}, \tag{2}$$

$$D_\alpha^{(2)} = \int_{-\infty}^{\infty} d\omega_1 \int_{-\infty}^{\infty} d\omega_2 \varepsilon_{\alpha\beta\gamma}^{(2)}(\omega_1, \omega_2) E_\beta(\omega_1) E_\gamma(\omega_2) \times \exp\{-i(\omega_1+\omega_2)t\}, \tag{3}$$

$$D_\alpha^{(3)} = \int_{-\infty}^{\infty} d\omega_1 \int_{-\infty}^{\infty} d\omega_2 \int_{-\infty}^{\infty} d\omega_3\, \varepsilon_{\alpha\beta\gamma\delta}^{(3)}(\omega_1, \omega_2, \omega_3) \times E_\beta(\omega_1) E_\gamma(\omega_2) E_\delta(\omega_3) \exp\{-i(\omega_1+\omega_2+\omega_3)t\}, \tag{4}$$

where $E_\alpha(\omega)$ is the Fourier component of the electric field of the light wave, $\epsilon_{\alpha\beta}^{(1)}$ is the linear dielectric polarizability, and $\epsilon_{\alpha\beta\gamma}^{(2)}$ and $\epsilon_{\alpha\beta\gamma\delta}^{(3)}$ are the second-order and third-order dielectric polarizabilities.

We shall be interested in the interaction of light with media which possess a center of inversion. In such media the second-order dielectric polarizability is determined by quadrupole and magnetic-dipole transitions which are small in comparison with the dipole terms, so that we neglect them. By using perturbation theory we can get the following expressions for $\epsilon^{(1)}$ and $\epsilon^{(2)}$:

$$\varepsilon_{\alpha\beta}(\omega) = \delta_{\alpha\beta} - \frac{4\pi}{\hbar} \times \sum_{m,n} \rho_n \left(\frac{d_{nm}^\alpha d_{mn}^\beta}{\omega - \omega_{mn} + i\Gamma_{mn}} - \frac{d_{nm}^\beta d_{mn}^\alpha}{\omega + \omega_{mn} + i\Gamma_{mn}} \right), \tag{5}$$

$$\begin{aligned}
\varepsilon_{\alpha\beta\gamma\delta}^{(3)}(\omega_1, \omega_2, \omega_3) = {} & -\frac{2\pi}{3\hbar^3} \sum_P P(\beta\omega_1, \gamma\omega_2, \delta\omega_3) \sum_{k,l,m,n} \rho_n \\
& \times \{[d_{nk}^\alpha d_{kl}^\beta d_{lm}^\gamma d_{mn}^\delta / (\omega_1+\omega_2+\omega_3-\omega_{kn}+i\Gamma_{kn}) \\
& \times (\omega_2+\omega_3-\omega_{ln}+i\Gamma_{ln})(\omega_3-\omega_{mn}+i\Gamma_{mn})] \\
& - [d_{nk}^\beta d_{kl}^\alpha d_{lm}^\gamma d_{mn}^\delta / (\omega_1+\omega_{kn}+i\Gamma_{kn}) \\
& \times (\omega_2+\omega_3-\omega_{ln}+i\Gamma_{ln})(\omega_3-\omega_{mn}+i\Gamma_{mn})] \\
& + [d_{nk}^\beta d_{kl}^\gamma d_{lm}^\alpha d_{mn}^\delta / (\omega_1+\omega_{kn}+i\Gamma_{kn}) \\
& \times (\omega_1+\omega_2+\omega_{ln}+i\Gamma_{ln})(\omega_3-\omega_{mn}+i\Gamma_{mn})] \\
& - [d_{nk}^\beta d_{kl}^\gamma d_{lm}^\delta d_{mn}^\alpha / (\omega_1+\omega_{kn}+i\Gamma_{kn}) \\
& + (\omega_1+\omega_2+\omega_{ln}+i\Gamma_{ln}) \\
& \times (\omega_1+\omega_2+\omega_3+\omega_{mn}+i\Gamma_{mn})]\}.
\end{aligned} \tag{6}$$

Here d_{mn}^α is the matrix element of the operator for the αth component of the dipole moment per unit volume of the medium; $\hbar\omega_{mn} = \mathscr{E}_m - \mathscr{E}_n$, where the $\mathscr{E}_n$ are the energy levels of the medium; ρ_n is a diagonal element of the equilibrium density matrix; P(a, b, c) is an operator of permutation of the quantities a, b, c; and $\Gamma_{mn} = (\Gamma_m + \Gamma_n)/2$, where $\hbar\Gamma_n$ is the energy width of the level $\mathscr{E}_n$.

The expression (6) is rather complicated. In the linear effect we are considering, however—the production of the third harmonic—the elements of the third-order tensor $\epsilon_{\alpha\beta\gamma\delta}^{(3)}(\omega_1, \omega_2, \omega_3)$ that are involved are those with the following arguments:

$$-\omega, \omega, \omega; \quad -3\omega, 3\omega, \omega; \quad -\omega, -\omega, 3\omega;$$

$$\omega, \omega, \omega; \quad -\omega, \omega, 3\omega.$$

Besides this, we shall be interested in the situation in which the energy-level system is such that there is resonance with the intermediate frequency 2ω; that is, there are some levels for which the difference $\omega_{mn} - 2\omega$ is small. Accordingly, in Eq. (6) we confine ourselves to the consideration of terms that have this small difference in the denominator. To simplify the expressions given below, we confine ourselves to the treatment of the case in which the harmonic generated is polarized in the same plane as the incident light, so that all of the formulas that follow involve elements of the tensor (6) which are diagonal in all of the indices. Then the five elements of the third-order nonlinear tensor that are indicated above take the forms

$$\begin{aligned}
\varepsilon^{(3)}(-\omega, \omega, \omega) &= -{}^8/_3\pi \sum_{m,n} \rho_n |\mu_{nm}|^2 / (2\omega - \omega_{mn} + i\Gamma_{mn}), \\
\varepsilon^{(3)}(-3\omega, 3\omega, \omega) &= -{}^2/_3\pi \sum_{m,n} \rho_n |\nu_{nm}|^2 / (2\omega - \omega_{mn} - i\Gamma_{mn}), \\
\varepsilon^{(3)}(-\omega, \omega, 3\omega) &= -{}^2/_3\pi \sum_{m,n} \rho_n |\nu_{nm}|^2 / (2\omega - \omega_{mn} + i\Gamma_{mn}), \\
\varepsilon^{(3)}(\omega, \omega, \omega) &= -4\pi \sum_{m,n} \rho_n \nu_{nm} \mu_{mn}^* / (2\omega - \omega_{mn} + i\Gamma_{mn}), \\
\varepsilon^{(3)}(-\omega, -\omega, 3\omega) &= -{}^2/_3\pi \sum_{m,n} \rho_n [4\mu_{nm}\nu_{mn}^* / (2\omega - \omega_{mn} + i\Gamma_{mn}) + 2\mu_{nm}\nu_{mn}^* / (2\omega - \omega_{mn} - i\Gamma_{mn})],
\end{aligned} \tag{7}$$

where

$$\nu_{nm} = 2\hbar^{-3/2}\sum d_{nk}d_{km}(\omega_{kn}-\omega)/(3\omega-\omega_{kn})(\omega+\omega_{kn}),$$

$$\mu_{nm} = \hbar^{-3/2}\sum_k d_{nk}d_{km}/(\omega-\omega_{kn}). \quad (8)$$

When there is no resonance, the ratio $D^{(3)}/D^{(1)}$ is a quantity of the order of $(E/E_a)^2$, where E_a is the intraatomic field. It follows from (7) and (8) that when there is a resonance at the frequency 2ω this ratio attains the order of magnitude of $(E/E_a)^2\omega/\Gamma$.

The intensity with which the third harmonic is produced can be increased in comparison with the nonresonance case by a factor $(\omega/\Gamma)^2$ (see below). We may state that the ratio ω/Γ can be rather large; in particular, for a number of rare earths dispersed in crystals it can reach values $\sim 10^5$. Still larger values can be achieved at low temperatures in gases, for resonances with forbidden transitions.[7]

3. THE SYSTEM OF NONLINEAR EQUATIONS WHICH DESCRIBE THE TRANSFORMATION OF OPTICAL FREQUENCIES

Let us consider a plane, linearly polarized, electromagnetic wave of frequency ω which is propagated in a continuous medium.

As the monochromatic light travels through the medium multiple harmonics will appear; in an isotropic medium or a medium possessing a center of inversion the third harmonic is most strongly produced. We confine ourselves to the case in which the intensity of the third harmonic is small enough so that it does not cause nonlinear processes such as the generation of a ninth harmonic. Starting from Maxwell's equations and the material equation (1), we find a system of equations connecting the amplitudes E_1 and E_3 of the first and third harmonics (see, for example, [8]):

$$d^2E_1/dz^2 + (\omega/c)^2[\varepsilon^{(1)}(\omega)E_1 + {}^3/_4\varepsilon^{(3)}(-\omega,\omega,\omega)|E_1|^2E_1$$
$$+ {}^3/_2\varepsilon^{(3)}(-3\omega,3\omega,\omega)|E_3|^2E_1$$
$$+ {}^3/_4\varepsilon^{(3)}(-\omega,-\omega,3\omega)E_3(E_1^*)^2] = 0, \quad (9)$$

$$d^2E_3/dz^2 + (3\omega/c)^2[\varepsilon^{(1)}(3\omega)E_3 + {}^1/_4\varepsilon^{(3)}(\omega,\omega,\omega)E_1^3$$
$$+ {}^3/_2\varepsilon^{(3)}(-\omega,\omega,3\omega)|E_1|^2E_3] = 0. \quad (10)$$

We look for the solution of these equations in the form

$$E_1(z) = A_1(z)\exp\{ik(\omega)z\}, \qquad E_3 = A_3(z)\exp\{ik(3\omega)z\},$$
$$k(\omega) = (\omega/c)[\varepsilon^{(1)}(\omega)]^{1/2}, \qquad k(3\omega) = (3\omega/c)[\varepsilon^{(1)}(3\omega)]^{1/2}, \quad (11)$$

in which we shall assume that $A_1(z)$ and $A_3(z)$ are slowly varying functions of the variable z, in comparison with the rapidly oscillating exponentials. This allows us to derive the following system of first-order nonlinear differential equations for $A_1(z)$ and $A_3(z)$:

$$dA_1/dz + \alpha|A_1|^2A_1 + \beta|A_3|^2A_1$$
$$+ \gamma A_3(A_1^*)^2e^{-i\Delta kz} = 0, \quad (12)$$

$$dA_3/dz + \tilde{\beta}|A_1|^2A_3 + \tilde{\gamma}A_1^3e^{i\Delta kz} = 0. \quad (13)$$

Here we have introduced the notations $\Delta k = 3k(\omega) - k(3\omega)$ and

$$\alpha = -3i\omega^2\varepsilon^{(3)}(-\omega,\omega,\omega)/8c^2k(\omega),$$
$$\beta = -3i\omega^2\varepsilon^{(3)}(-3\omega,3\omega,\omega)/4c^2k(\omega),$$
$$\gamma = -3i\omega^2\varepsilon^{(3)}(-\omega,-\omega,3\omega)/8c^2k(\omega),$$
$$\tilde{\beta} = -27i\omega^2\varepsilon^{(3)}(-\omega,\omega,3\omega)/4c^2k(3\omega),$$
$$\tilde{\gamma} = -9i\omega^2\varepsilon^{(3)}(\omega,\omega,\omega)/8c^2k(3\omega). \quad (14)$$

The second term in Eq. (12) describes two-photon absorption, and the third term in (12) and the second in (13) describe the process of absorption of a photon of frequency 3ω and simultaneous emission of a photon of frequency ω, so that the result of such an act of interaction is that the energy $2\hbar\omega$ is absorbed in the medium. The last terms in (12) and (13) do not admit of a simple quantum-mechanical description, since they describe processes that depend essentially on the relation between the phases of the interacting optical harmonics.

4. THE CASE OF A WEAK LIGHT-WAVE FIELD

If the electric field E_0 of the incident light wave of frequency ω is weak, namely if it satisfies the condition

$$|\tilde{\gamma}E_0^2/\Delta k| \ll 1, \quad (15)$$

then, as will be seen from what follows, the intensity with which the third harmonic is produced will be much smaller than the intensity of the fundamental frequency. In this case we can neglect the last two terms in (12) and the second term in (13); the result of this is that we get a rather simple system

$$dA_1/dz + \alpha|A_1|^2A_1 = 0,$$
$$dA_3/dz + \tilde{\gamma}A_1^3e^{i\Delta kz} = 0, \quad (16)$$

whose solution under the boundary conditions $A_1(0) = E_0$, $A_3(0) = 0$ is of the form

$$A_1(z) = E_0(1+\alpha' E_0^2 z)^{-1/2} \exp[i(\alpha'/2\alpha'') \times \ln(1+\alpha' E_0^2 z)], \quad (17)$$

$$A_3(z) = E_0 \frac{\tilde{\gamma}}{\alpha'} \int_0^{\alpha' E_0^2 z} d\zeta (1+\zeta)^{-3/2} \times \exp\left[i\frac{3\alpha'}{2\alpha''} \ln(1+\zeta) + i\frac{\Delta k \zeta}{\alpha' E_0^2} \right], \quad (18)$$

where $\alpha = \alpha' + i\alpha''$. When we make an expansion in powers of the small parameter (15) in the last expression, we get

$$A_3(z) = i\tilde{\gamma} E_0^3 \Delta k^{-1} \left\{ 1 - \exp\left[i\Delta kz - \frac{3}{2}\left(1 - i\frac{\alpha'}{\alpha''}\right) \times \ln(1+\alpha' E_0^2 z) \right]\right\}, \quad (19)$$

from which it follows that the maximum intensity of the third harmonic is given by the formula

$$|A_3|^2 = |\tilde{\gamma}|^2 E_0^6 / |\Delta k|^2. \quad (20)$$

Returning to the formulas (7) and (14), which give the coefficient $\tilde{\gamma}$ as a function of the frequency ω, we can see the resonance character of the effect of third-harmonic production: the expression (20) has a sharp maximum at the point $2\omega = \omega_{mn}$ (we note that for $\omega = \omega_{mn}$ or $3\omega = \omega_{mn}$ not only $\tilde{\gamma}$ but also Δk has a sharp maximum, and since it is the ratio of these quantities that occurs in (20), the intensity of the third harmonic is essentially unchanged as compared with the nonresonance case).

5. THE CASE OF A STRONG LIGHT-WAVE FIELD

Let us consider the case in which the electric field in the incident light wave is strong, so that we have the condition opposite to (15):

$$|\Delta k / \tilde{\gamma} E_0^2| \lesssim 1. \quad (21)$$

This inequality facilitates the resonance behavior of the coefficient $\tilde{\gamma}$. If the medium is transparent to the first and third harmonics and is strongly anisotropic (but, as before, possesses a center of inversion), then, as is well known,[8-10] directions can be chosen along which Δk is extraordinarily small. Let us set $\Delta k = 0$ in (12) and (13); the corrections to the solution so obtained are of the order of $\Delta k/\tilde{\gamma}E_0^2$. Even after these simplifications, however, the system of equations (12) and (13) is still complicated; it is a system of four nonlinear first-order differential equations, because the amplitudes A_1 and A_3 are complex. If, however, we consider the case of exact resonance $(2\omega - \omega_{mn} = 0)$, then the situation is much simpler, since the coefficients that appear in (12) and (13) satisfy the relations

$$\operatorname{Im}\alpha = \operatorname{Im}\beta = 0, \quad 3\beta = -\tilde{\beta}, \quad \gamma = |\gamma| e^{i\psi}, \\ \tilde{\gamma} = 3|\gamma| e^{-i\psi}, \quad |\gamma|^2 = \alpha\beta/2. \quad (22)$$

We can now look for the solution of Eqs. (12) and (13) in the form

$$A_1(z) = R_1(z)\exp\{i\varphi_1\}, \\ A_3(z) = R_3(z)\exp\{-i(3\varphi_1 - \psi - \pi)\}, \quad (23)$$

where R_1 and R_3 are real and satisfy the following system of equations:

$$dR_1/dz + \alpha R_1^3 - \beta R_3^2 R_1 - (\alpha\beta/2)^{1/2} R_3 R_1^2 = 0, \quad (24)$$

$$dR_3/dz + 3\beta R_1^2 R_3 - 3(\alpha\beta/2)^{1/2} R_1^3 = 0. \quad (25)$$

In the phase plane (R_1, R_3) these equations have a straight line of singular points (Fig. 1)

$$R_3 = \eta R_1, \quad \eta = (\alpha/2\beta)^{1/2}. \quad (26)$$

It is not hard to show that points on this line are points of stable equilibrium: small deviations from them have the result that the system of interacting light waves with the amplitudes R_1 and R_3 returns asymptotically to the original point along a line perpendicular to the line $R_3 = \eta R_1$. There is another line of singular points, $R_1 = 0$, but the states of the system of interacting waves R_1 and R_3 on this line are unstable.

Outside the lines $R_3 = \eta R_1$ and $R_1 = 0$ the equation for the determination of the phase trajectories is of the form

$$dR_3/dR_1 = -3[2\eta + (R_3/R_1)]^{-1}. \quad (27)$$

Equation (27) can be integrated without difficulty and leads to the following equations for the characteristics:

$$\ln R_1 + \Phi(\eta, R_3/R_1) = 0, \quad (28)$$

$$\Phi(\eta, t) = \begin{cases} \dfrac{1}{2}\ln\left|\dfrac{t^2+2\eta t+3}{3}\right| + \dfrac{\eta}{(3-\eta^2)^{1/2}}\tan^{-1}\dfrac{t(3-\eta^2)^{1/2}}{3+\eta t}, & \eta \leqslant 3 \\ \dfrac{\eta+(\eta^2-3)^{1/2}}{2(\eta^2-3)^{1/2}}\ln\left|\dfrac{t+\eta-(\eta^2-3)^{1/2}}{\eta-(\eta^2-3)^{1/2}}\right| - \dfrac{\eta-(\eta^2-3)^{1/2}}{2(\eta^2-3)^{1/2}}\ln\left|\dfrac{t+\eta+(\eta^2-3)^{1/2}}{\eta+(\eta^2-3)^{1/2}}\right|, & \eta > 3. \end{cases} \quad (29)$$

The phase trajectories for some values of the parameter η are shown in Fig. 1. A phase trajectory meets the line (26) at a right angle, and, as the figure shows, the point of intersection determines the maximum value of the amplitude of the third harmonic, $R_3(\eta)$ that can be attained for a

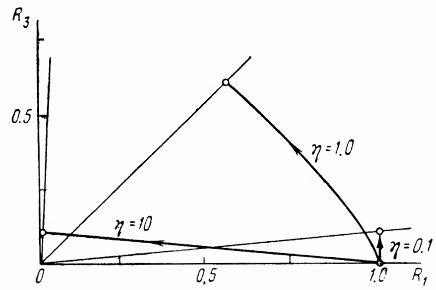

FIG. 1. Phase trajectories for three different values of the parameter η; R_1 and R_3 are the amplitudes of the first and third harmonics.

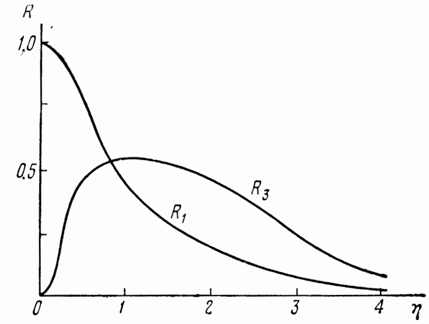

FIG. 2. Dependences on the parameter η of the amplitudes R_1 and R_3 of the first and third harmonics at the point of stable equilibrium.

given value of η, and the corresponding value of the other amplitude, $R_1(\eta)$. From (26) and (28) we easily find

$$R_3(\eta) = \eta \exp\{-\Phi(\eta, \eta)\},$$

$$R_1(\eta) = \exp\{-\Phi(\eta, \eta)\}. \tag{30}$$

Curves of these amplitudes are plotted in Fig. 2. It can be seen that $R_3(\eta)$ is small both for $\eta \ll 1$ and for $\eta \gg 1$, and reaches its maximum value $R_3(1) = 2^{-1/2}e^{-1/4}$ at $\eta = 1$. It follows that the maximum intensity of the third harmonic which can be reached by conversion from the first harmonic by means of a medium which has a resonance for the second harmonic amounts to about 30 percent of the incident first-harmonic intensity of the light.

The impossibility of reaching large values of R_3 for large η is explained by the fact that along with the process of interconversion of the harmonics there is a strong two-photon absorption of the first harmonic, which is represented by the term αR_1^3 in Eq. (24). In the opposite case of small η, when the two-photon absorption is weak, it is also impossible for R_3 to become large, since processes of reverse transition from the third harmonic to the first are strong here. It must be pointed out that as the amplitude of the third harmonic grows the nonlinear absorption (in particular, the two-photon absorption) of the first harmonic decreases, so as to become equal to zero when the condition (26) is satisfied.

In conclusion the authors thank N. G. Basov, O. N. Krokhin, and A. N. Oraevskiĭ for their interest in this work and for a discussion.

[1] A. M. Afanas'ev and É. A. Manykin, JETP **48**, 483 (1965), Soviet Phys. JETP **21**, 323 (1965).

[2] Miller, Kleinman, and Savage, Phys. Rev. Letters **11**, 146 (1963).

[3] R. C. Miller, Phys. Rev. **134**, A1313 (1964).

[4] Maker, Terhune, and Savage, Quantum Electronics, Proc. of the Third International Conference, Paris, 1964, **2**, page 1559.

[5] S. A. Pollack, Abstracts of Electronics Division **13**, 234 (1964).

[6] M. A. El'yashevich, Spektry redkikh zemel' (Spectra of the Rare Earths), Gostekhizdat, 1953.

[7] L. A. Borisoglebskiĭ, UFN **66**, 603 (1958), Soviet Phys. Uspekhi **1**, 211 (1958).

[8] Armstrong, Bloembergen, Ducuing, and Pershan, Phys. Rev. **127**, 1918 (1962).

[9] Maker, Terhune, and Savage, Phys. Rev. Letters **8**, 21 (1962).

[10] S. A. Akhmanov and R. V. Khokhlov, JETP **43**, 351 (1962), Soviet Phys. JETP **16**, 252 (1963).

Translated by W. H. Furry
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