PASSAGE OF ELECTROMAGNETIC RADIATION THROUGH A RESONANT MEDIUM IN THE PRESENCE OF AN INTENSE MONOCHROMATIC WAVE

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Submitted January 20, 1969

Zh. Eksp. Teor. Fiz. 59, 195–200 (July, 1970)

The passage of weak electromagnetic radiation through a resonant medium consisting of two-level atoms in the presence of an intense monochromatic field is considered in the case of antiparallel and parallel propagation of the weak and intense waves. In the first case expressions are obtained for the absorption coefficient and the refractive index of the medium for the weak wave. It is shown that the absorption coefficient is always negative at the frequency of the "three-photon transition," and the refractive index of the medium exhibits "anomalous" dispersion in the neighborhood of this frequency. In the second case it is shown that new regions of amplification appear in the spectrum of the weak field as a result of two-photon decay involving a change of the frequency.

1. INTRODUCTION

RECENTLY a considerable amount of attention has been given to nonlinear optical phenomena in metallic vapors. Under the influence of intense radiation, a pronounced change of the atomic states occurs, as a result of which a number of new effects arise which are not present in linear optics. Among such phenomena are the Stark effect in variable fields,[1,2] stimulated Raman scattering in electronic levels,[3] stimulated radiation of resonance lines of an atom,[2,3] etc.

Stimulated resonant three-photon (an atom absorbs two quanta of frequency ω' from an intense field, emits a quantum of frequency $2\omega' - \omega_0$, thus passing to the excited state) and four-photon (the decay of two quanta of the intense field into two other quanta with a change of the frequency) interactions were observed in[4,5]. Since these effects are purely resonant in character, one can limit their theoretical investigation to a two-level atom in an intense monochromatic pumping field and weak radiation. Such a problem has been considered in a number of articles. The polarizability of an atom in the field of two weak waves with frequencies ω' and ω was calculated in[6], and it was shown that in third-order perturbation theory (also see[7]) additional terms with frequencies $2\omega' - \omega$ and $2\omega - \omega'$ appear in the polarizability. In[8] the problem is solved exactly in a strong field, and the conditions under which the component of the polarization at the Raman frequency $2\omega' - \omega$ is comparable in order of magnitude with the component at the frequency ω are determined. In[9] the value of the atomic polarizability obtained in[8] is used in the field equations, and the problem of transmission is solved in the case of an exact resonance with a strong field ($\omega' = \omega$) (also see[10]). The appearance of the Raman frequency $2\omega' - \omega$ is associated with a resonant four-photon interaction. In particular, this leads to a broadening of the spectral line upon passage through a resonant medium (see[5]). However, it is impossible to use the results obtained in[9,10] in the real situations described in[4,5], where $\epsilon = \omega_0 - \omega' \neq 0$. In addition, in[8-10] it is actually assumed that the relaxation time $\tau_{rel} \ll \tau$ (where τ denotes the time of interaction of the radiation with the atoms). Since $\tau \sim \epsilon^{-1}(1+\xi)^{-1/2}$ where ξ is the Stark parameter introduced below, then in[4,5] $\tau \sim 10^{-12}$ sec, but $\tau_{rel} \sim 10^{-7}$ to 10^{-10} sec, and the assumption $\tau_{rel} \ll \tau$ is violated. A "three-photon" interaction in implicit form is also contained in[8,9] (the polarizability of an atom at the frequency ω of the weak field has a resonance at $\omega \approx 2\omega' - \omega_0$). However, the manifestation of this resonance in the real problem of transmission, and also the influence of the four-photon interaction on the "three-photon" process were not investigated in[8,9].

The passage of a weak wave through a resonant medium in the presence of an intense monochromatic field is considered in the present article. In this connection, fundamental attention is given to the case of propagation of the weak wave along the direction of propagation of the intense wave, since the effects of accumulation only occur in this case. In this connection a two-photon decay, leading to a coupling of the components of the weak field at the frequencies ω and $2\omega' - \omega$, occurs only in the case of parallel propagation of the waves (more precisely, in a narrow "forward" cone in virtue of the small difference of the medium's index of refraction from unity). Therefore, we shall consider the cases of parallel and counter waves separately. In addition, our investigation corresponds to the case $\tau \ll \tau_{rel}$. In this connection, in order to not obtain erroneous effects it is necessary to switch on the interaction correctly (see[11,12]).

2. THE CASE OF OPPOSING WAVES

The quasiclassical equations of a resonant medium containing two-level atoms and the radiation fields have the form[13]

$$\pm\frac{\partial A_{1,2}}{\partial x}+\frac{1}{c}\frac{\partial A_{1,2}}{\partial t}=-\overline{p\rho e^{\mp ikx}}, \tag{1}$$

$$\partial\rho/\partial t+i\varepsilon\rho=(A_1e^{ikx}+A_2e^{-ikx})\sqrt{1-q^2|\rho|^2}, \tag{2}$$

where

$$p = -\frac{2\pi|M|^2}{c\hbar\,\omega'}\Delta_0, \quad q = \frac{2|M|}{c\hbar}. \tag{3}$$

Here A_1 and A_2 denote the amplitudes of the weak and intense fields, ρ is the transition current, M is the matrix element, and Δ_0 is the initial value of the excess population. In order to simplify the calculations, irreversible relaxations are neglected in Eq. (2) (a consideration of the relaxations is given in Sec. 4), and the interaction is switched on at $t = -\infty$.[11,12] The bar in Eq. (1) denotes averaging over the spatial period of the wave.

Further, we shall assume that $|A_1(t, x)| \ll |A_2(x)|$ and we confine our attention to the linear approximation in A_1. In analogy to the field, we also represent the transition current in the form $\rho = \rho_1 + \rho_2$, where $|\rho_1| \ll |\rho_2|$. Separating the equations for $A_{1,2}$ and $\rho_{1,2}$ and assuming that $\partial\rho_2/\partial t = 0$ (a monochromatic wave), we find

$$\frac{\partial A_1}{\partial x} + \frac{1}{c}\frac{\partial A_1}{\partial t} = -p\overline{\rho_1 e^{-ikx}}, \tag{4}$$

$$\frac{\partial^2\rho_1}{\partial t^2} + \varepsilon^2(1+\xi)\rho_1 = \left[\frac{\partial A_1}{\partial t} - i\varepsilon\left(1+\frac{\xi}{2}\right)A_1\right]\frac{e^{ikx}}{\sqrt{1+\xi}} + i\varepsilon\frac{\xi}{2}A_1^*\frac{e^{-ikx-2ik(\xi)x}}{\sqrt{1+\xi}}. \tag{5}$$

Here the following dimensionless parameters have been introduced for the intensity and nonlinear wave vector of the intense wave:

$$\xi = \frac{q^2}{\varepsilon^2}|A_2(0)|^2, \quad k(\xi) = k + \frac{p}{\varepsilon\sqrt{1+\xi}}. \tag{6}$$

In Eq. (4) we do not consider inhomogeneous broadening of the atomic line, i.e., we assume $\epsilon \gg \Delta\omega_{\text{Doppler}}$ which is well satisfied in the experiments mentioned in the introduction.

From Eq. (5) one can express ρ_1 terms of A_1 and substitute into (4). Then for the spectral component $A_1(\omega, x)$ the equation

$$\frac{\partial A_1(\omega,x)}{\partial x} = \frac{i\omega}{c}A_1(\omega,x) - \frac{p}{2(1+\xi)}\left\{\left(\sqrt{1+\xi}-1-\frac{\xi}{2}\right)\left[\frac{-i}{\omega'-\omega-\varepsilon\sqrt{1+\xi}} + \pi\delta(\omega'-\omega-\varepsilon\sqrt{1+\xi})\right] + \left(\sqrt{1+\xi}+1+\frac{\xi}{2}\right)\left[\frac{-i}{\omega'-\omega+\varepsilon\sqrt{1+\xi}} + \pi\delta(\omega'-\omega+\varepsilon\sqrt{1+\xi})\right]\right\}A_1(\omega,x). \tag{7}$$

is obtained. From here one can obtain the following values for the absorption coefficient $\alpha(\omega)$ and the index of refraction $n(\omega)$ for the weak wave:

$$\alpha(\omega) = \frac{\pi p}{1+\xi}\left[\left(\sqrt{1+\xi}-1-\frac{\xi}{2}\right)\delta(\omega'-\omega-\varepsilon\sqrt{1+\xi}) + \left(\sqrt{1+\xi}+1+\frac{\xi}{2}\right)\delta(\omega'-\omega+\varepsilon\sqrt{1+\xi})\right], \tag{8}$$

$$n(\omega) = 1 + \frac{cp}{2\omega(1+\xi)}\left(\frac{\sqrt{1+\xi}-1-\xi/2}{\omega'-\omega-\varepsilon\sqrt{1+\xi}} + \frac{\sqrt{1+\xi}+1+\xi/2}{\omega'-\omega+\varepsilon\sqrt{1+\xi}}\right),$$
$$\omega \neq \omega' \pm \varepsilon\sqrt{1+\xi}. \tag{9}$$

The second term in (8) describes resonant absorption in the presence of a strong wave. Here the absorption is shifted by the Stark effect. The first term in (8) is not present in the linear approximation and leads for arbitrary ξ to negative absorption at the "three-photon" frequency[1] (which is also shifted due to the Stark effect). In connection with this an "anomalous" dispersion (see Eq. (9)) of the index of refraction appears in the neighborhood of the "three-photon" frequency. (In fact, the resonance in the first term of (8) is due to many-photon processes in general so that the "three-photon" term is somewhat conditional for arbitrary ξ, in connection with which we use it in quotation marks.)

It is of interest to note that $\alpha(\omega)$ and $n(\omega)$ are related by the usual dispersion relation.

3. THE CASE OF PARALLEL WAVES

In the case of parallel waves the equations of the medium take the form

$$\frac{\partial A}{\partial x} + \frac{1}{c}\frac{\partial A}{\partial t} = -p\rho, \tag{10}$$

$$\partial\rho/\partial t + i\varepsilon\rho = A\sqrt{1-q^2|\rho|^2}. \tag{11}$$

Just as in the previous case we shall assume that $A = A_1(t, x) + A_2(x)$ and $\rho = \rho_1 + \rho_2$, where $|A_1| \ll |A_2|$, $|\rho_1| \ll |\rho_2|$ and $\partial\rho_2/\partial t = 0$. Then in the linear approximation in A_1 we find

$$A_1(\omega,x) = C_1(\omega)\exp\left\{\frac{px}{\varepsilon\sqrt{1+\xi}}r_1\right\} + C_2(\omega)\exp\left\{\frac{px}{\varepsilon\sqrt{1+\xi}}r_2\right\}, \tag{12}$$

where

$$C_{1,2}(\omega) = \left(\frac{1}{2} \mp i\frac{z^2-\xi/2}{2z\sqrt{\xi-z^2}}\right)A_1(\omega,0) \pm i\frac{\xi/2}{2z\sqrt{\xi-z^2}}A_1^*(2\omega'-\omega,0), \tag{13}$$

$$r_{1,2} = i + \frac{i\pm\sqrt{\xi-z^2}}{2}\left[\frac{1}{z-\sqrt{1+\xi}} + \frac{1}{z+\sqrt{1+\xi}} + i\pi\delta(z-\sqrt{1+\xi}) + i\pi\delta(z+\sqrt{1+\xi})\right]. \tag{14}$$

Here $z = (\omega' - \omega)/\epsilon$.

As is evident from Eq. (12), $A_1(\omega, x)$ is determined by the spectral components of the incident radiation at the frequencies ω and $2\omega' - \omega$. An additional exponential amplification appears in the frequency range

$$(\omega'-\omega)^2 < \varepsilon^2\xi. \tag{15}$$

Here the maximum amplification occurs at the frequencies

$$\omega' \pm \varepsilon\sqrt{\xi(\xi+1)/(\xi+2)} \tag{16}$$

and the width of the maxima is proportional to $\sqrt{\xi}$. The formation of the maxima (16) associated with the presence of nonlinearity ($\xi \neq 0$) is the result of two-photon decay with a change of the frequency, which was mentioned in the Introduction. One can also verify this by starting from a qualitative analysis of the conservation laws $2\mathbf{k}' = \mathbf{k}_1 + \mathbf{k}_2$, $2\omega' = \omega_1 + \omega_2$, using expressions (6) and (9) as the indices of refraction for the intense and weak waves, respectively.

From the expansion of formula (12) in powers of x we have

$$A_1(\omega,x) = A_1(\omega,0)\left\{1 - \frac{\pi px}{2(1+\xi)}[(\sqrt{1+\xi}-1-\xi)\times\delta(\omega'-\omega-\varepsilon\sqrt{1+\xi}) + (\sqrt{1+\xi}+1+\xi)\delta(\omega'-\omega+\varepsilon\sqrt{1+\xi})]\right.$$

[1]The diameter of the "three-photon" scattering is calculated in [12].

$$+\frac{ipx}{2(1+\xi)}\left(\frac{\sqrt{1+\xi}-1-\xi}{\omega'-\omega-\varepsilon\sqrt{1+\xi}}+\frac{\sqrt{1+\xi}+1+\xi}{\omega'-\omega+\varepsilon\sqrt{1+\xi}}\right)\Big\} \quad (17)$$

(here the fact that $-A_1(\omega, 0) = A_1^*(2\omega' - \omega, 0)$ has been used); it is clear that at small distances the absorption and dispersion laws physically correspond to formulas (8) and (9), i.e., there is amplification at "three-photon" frequencies and absorption at atomic frequencies, just as in the case of waves moving in opposite directions. However, these processes rapidly come to a stop with increasing distance. This is associated with the exponential amplification in the region (15) (for $\xi \gg 1$ the "three-photon" and atomic frequencies coincide with the maxima given by expression (16)).

4. ALLOWANCE FOR IRREVERSIBLE RELAXATION

In order to not "smear" the line of the "three-photon" amplification, it is necessary to assume the atomic line sufficiently narrow, $\epsilon \gg \Gamma$. In this connection we still assume that the medium absorbs little:

$$\frac{\Gamma}{2}\int_{-\infty}^{+\infty}\frac{q^2|A|^2}{\varepsilon^2+q^2|A|^2}dt \ll 1.$$

Then from Eqs. (1) and (2) (with relaxation taken into account)

$$\frac{\partial\rho}{\partial t}+\left(i\varepsilon+\frac{\Gamma}{2}\right)\rho=(A_1e^{ikx}+A_2e^{-ikx})\sqrt{1-q^2|\rho|^2}$$

by a method similar to the one described in Sec. 2 one can obtain the equation

$$\frac{\partial A_1}{\partial x}=\frac{i\omega}{c}A_1+\frac{ip}{2(1+\xi)}\left[\frac{\sqrt{1+\xi}-1-\xi/2}{\omega'-\omega-\varepsilon\sqrt{1+\xi}-{}^1/_2 i\Gamma(1+\xi/2)}\right.$$
$$\left.+\frac{\sqrt{1+\xi}+1+\xi/2}{\omega'-\omega+\varepsilon\sqrt{1+\xi}-{}^1/_2 i\Gamma(1+\xi/2)}\right]A_1, \quad (18)$$

where the parameter ξ now depends on x:

$$\frac{\partial\xi}{\partial x}=-p\frac{\Gamma}{\varepsilon^2+\Gamma^2/4}\frac{\xi}{\sqrt{1+\xi}}$$
$$\xi(0)=\frac{q^2}{\varepsilon^2+\Gamma^2/4}|A_2(0)|^2. \quad (19)$$

As is clear from Eq. (18), the physical picture of the transmission has not changed, only the "three-photon" and atomic lines now have a width. In this connection the presence of the nonlinearity ξ leads to an increase of the width of these lines by a factor of $(1 + \xi/2)$ times.

In conclusion we note that in a real situation it is important to take the finite width of the intense field into consideration. However, in this case the problem becomes much more complicated and, as estimates show, the physical picture of transmission is not changed if the width of the incident intense field is small in comparison with the detuned resonance.

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Translated by H. H. Nickle
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