[1)] We note that in the presence of a mismatch ($\Delta \neq 0$) it is impossible to obtain equations in closed form for the intensities $I$ and $I_1$ (cf. [13]).

[2)] This phenomenon was apparently first pointed out by Ostrovskii, [18] who analyzed it qualitatively.


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# Resonance excitation of light and dynamic electro-optical effects


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An analysis is reported of the propagation of a weak plane light wave in a gas placed in strong constant and uniform alternating external electric fields. The frequency of the latter is in the radio band and the frequency of the incident light wave is close to the frequency of one of the allowed transitions in the gas molecules. The time modulation of the molecular transition frequency due to the Stark effect in the external electric field is taken into account. The degree of modulation and the mean intensity of the light wave transmitted through the medium under consideration are investigated as functions of the amplitude of the external alternating field, the constant external field, and the frequency of the incident light wave. A number of features of this functional dependence is noted. The possibility of observing these effects in a gas of molecules of the symmetric spinning-top type is discussed. Possible applications of these effects are examined.


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## 1. INTRODUCTION

Modulation of electromagnetic waves can be produced in media in which the refractive index is a function of the electric field. This type of modulation of light waves by an external electric radio-frequency field has been observed in dielectrics[1–4] and has subsequently found application in lasers where it is used for mode locking.[5–7] The modulation is also possible in the electron plasma of semiconductors and in gas plasma in an external magnetic field with an rf component modulating the cyclotron frequency.[8–13] The modulation of waves by an external electric low-frequency rf field in gas plasma, due to the modulation by this field of the electron mean free time, has been discussed by Kumar *et al.*,[14] and that due to the hydrodynamic modulation of the plasma density by this field has been discussed by Kumar *et al.*,[14] Aliev and Silin,[15] and Ostrovskiĭ and

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Stepanov.[16] Appreciable progress has also recently been achieved in the description of the modulation of light waves by acoustic waves in condensed dielectrics in the region in which they are transparent.[17–20]

In a previous paper,[21] we examined the modulation of light in dielectrics by an external uniform electric rf field, with the transition frequency in the medium modulated by the linear Stark effect. A matrix approach was adopted in the theory in which an initial determination was made of the light eigenwaves in the medium, and the solution of the subsequent boundary-value problem was then reduced to the expansion of the light field given on the boundary in terms of the eigenwaves. (This approach was proposed by the present author[9] and has been used by him[10,12,13,21] in the solution of a number of problems. An approach to the theory of modulation of light by the periodically inhomogeneous field of an acoustic wave based on a similar procedure was proposed and used by Singer and Tamir-Berman[18] and by Chu and Tamir.[19,20]) It was shown in a previous paper[21] that the nature of the modulation of the light wave in the medium was qualitatively different, depending on the density of the particles participating in the particular transition or, more specifically, on the ratio of the wavelength of the modulating field to the characteristic resonant absorption length for the light wave in the medium in the absence of modulation. In particular, it was predicted that, when this parameter was small, there would be an oscillation in the intensity of light transmitted by a layer of the medium under consideration, depending on the amplitude of the radio-field oscillations (the wave modulation being weak). When this parameter was large, i.e., for a relatively high density of particles, the analysis given previously[12,21] was restricted to the case of nondegenerate states for a homogeneously broadened transition line. Moreover, the medium was assumed to be spatially homogeneous. In this paper, we generalize the theory to the case of degenerate states, inhomogeneously broadened transition lines, two external electric fields (one constant and one alternating), and a medium that is spatially inhomogeneous in the longitudinal direction. Attention is drawn to a number of new effects that may be of practical interest. Results obtained by this general analysis are then applied to a gas of molecules of the symmetric spinning-top type.

## 2. BASIC RELATIONSHIPS

Consider the propagation of a weak light wave in a medium exposed to a strong homogeneous rf field of frequency $p$. If this wave is excited by a monochromatic source of frequency $\omega$, then, in general, its electric field can be written in the form

$$\mathbf{e}_2(\mathbf{r},t)=\frac{1}{2}\sum_n \mathbf{E}_n(\mathbf{r})e^{-i(\omega+np)t}+\text{c.c.} \tag{1}$$

The linear polarization response in the medium, induced by the field (1), has the same form, namely,

$$\mathbf{p}_2(\mathbf{r},t)=\frac{1}{2}\sum_m \mathbf{P}_m(\mathbf{r})e^{-i(\omega+mp)t}+\text{c.c.}, \tag{2}$$

where

$$\mathbf{P}_m=\sum_n \hat{\varkappa}^{(mn)}(\mathbf{r},\omega)\mathbf{E}_n. \tag{3}$$

The infinite dimensional matrix $\hat{\varkappa}^{(mn)}$ can be regarded as the generalized susceptibility of the medium. We shall be concerned below with a plane linearly polarized (say, along the $x$ axis) wave propagating in the direction of the $z$ axis, assuming that each of the matrix elements $\varkappa^{(mn)}$ is a scalar, which is valid provided the medium is isotropic and the rf field polarized along the same $x$ axis. We shall also suppose, at the beginning, that the medium is homogeneous $[\varkappa^{(mn)}=\varkappa^{(mn)}(\omega)]$. When this is so, the Maxwell equations and (1)–(3) can be written in the form

$$d^2E/dz^2=QE,\quad Q=-\beta^2(1+4\pi T). \tag{4}$$

where $E$ is the column matrix with elements $E_n$, the components of the vectors $\mathbf{E}_n$ along the $x$ axis ($-\infty<n<\infty$), and

$$\langle\beta\rangle_{mn}=k_n\delta_{mn},\ k_n=(\omega+np)/c,\ \langle T\rangle_{mn}=\varkappa^{(mn)}(\omega).$$

Let $S$ be a transformation that diagonalizes the matrix $Q$, i.e., the matrix $\Lambda=S^{-1}QS$ is diagonal ($\langle\Lambda\rangle_n=\Lambda_l\delta_{nl}$). The general solution of (4) can then be written in the form

$$E=Se^{z\sqrt{\Lambda}}C_1+Se^{-z\sqrt{\Lambda}}C_2, \tag{5}$$

where $C_1$, $C_2$ are arbitrary constants. In the usual representation, Eq. (5) has the form

$$E=\sum_{\alpha,l}C_{\alpha l}q_{\alpha l}(z)\varphi_l(t)e^{-i\omega t}, \tag{6}$$

where

$$\varphi_l(t)=\sum_n\langle S\rangle_{nl}e^{-inpt},\quad q_{\alpha l}(z)=\exp[-(-1)^\alpha z\sqrt{\Lambda_l}], \tag{7}$$

and $C_{\alpha l}$ are arbitrary constants ($\alpha=1,2$) and $e_2=\text{Re}E$ is the component of the electric field vector of the light wave along the $x$ axis. According to (6), a plane wave in the above medium will, in general, be a superposition of an infinite number of eigenwaves determined by functions of $t$ and $z$ with constants $C_{\alpha l}$. In contrast to the case of a stationary medium, it is clear that these waves will contain factors $\varphi_l(t)$ that are periodic functions of time. Their period is determined by the period of the variation in the external modulating field. The sum in (6) describes a plane wave propagating in the $z$ direction for $\alpha=1$.

Suppose now that the medium under consideration occupies the half-space $z\geqslant 0$ and a plane monochromatic wave is incident normally on the boundary $z=0$. In general, the wave may be reflected by the boundary and this may be accompanied by appreciable transformation into components with new frequencies $\omega+np$ ($n\neq 0$). The general approach to the solution of this problem and the conditions for the effective transformation of energy into these components have been formulated[13] for the case of an electron plasma in an external modulating magnetic field. However, in the situations that we shall discuss below, the transformation accompanying reflection will be relatively weak. The field $e_2$ on the $z=0$ boundary can therefore be specified to be monochromatic. Using (5) and (6), we can then write the wave field for $z \geqslant 0$ in the form

$$E=E^{(0)}\sum_{l}\langle S^{-1}\rangle_{l0}q_{1l}(z)\varphi_l(t)e^{-i\omega t}, \tag{8a}$$

or, using (7),

$$E=E^{(0)}\sum_{n,l}q_{1l}(z)\langle S\rangle_{nl}\langle S^{-1}\rangle_{l0}e^{-i(\omega+np)t}. \tag{8b}$$

Equation (8a) is an expansion of the field in the eigenwaves in the medium, and the expression given by (8b) is an expansion over the monochromatic components. It is clear from (8b) that the propagation of the wave in the above medium is accompanied by the appearance of components with the new frequencies $\omega+np$, i.e., the original wave becomes modulated. This modulation becomes appreciable for values of $z$ for which there is an appreciable dependence of the $q_{1l}$ on $l$. Expressions of the form given by (8) are valid for any dielectric in a uniform field modulated periodically in time.

Let us begin by considering the case where the susceptibility $\varkappa^{(mn)}(\omega)$ is due to the influence of two nondegenerate energy states of the medium, i.e., we shall consider the susceptibility of a set of two-level particles subjected to a strong modulating electric field $\mathbf{e}_1 = \mathbf{e}_{10}\cos pt$ (parallel to the $x$ axis) and a weak light field $\mathbf{e}_2$. We shall also suppose that the diagonal matrix elements $d_{aa}$, $d_{bb}$ of the projection of the dipole-moment operator along the $x$ axis are not zero and not equal to each other. If the transition line between the state under consideration is homogeneously broadened, then, provided

$$|d_{ab}e_{10}|,\ \hbar p \ll \hbar\omega_{ba}$$

($d_{ab}$ is a nondiagonal matrix element of the operator representing the projection of the dipole moment along the $x$ axis, $\omega_{ba}=(E_b-E_a)/\hbar$ is the frequency of the transition under consideration), the main influence of the field $\mathbf{e}_1$ on the susceptibility of the medium to the field $\mathbf{e}_2$ is due to the modulation of the transition frequency by the linear Stark effect[22] so that the expression for $\varkappa^{(mn)}(\omega)$ can be written in the form[21,23,24]

$$\varkappa^{(mn)}(\omega)=\varkappa_0\delta_{mn}+[N\Delta\rho|d_{ab}|^2(\omega+np)/\hbar\omega_{ba}]$$
$$\times\sum_{s}\left(\frac{J_{s-m}(\Delta)J_{s-n}(\Delta)}{\omega-\omega_{ba}+sp+i/\tau_\perp}+\frac{J_{m-s}(\Delta)J_{n-s}(\Delta)}{\omega+\omega_{ba}+sp+i/\tau_\perp}\right), \tag{9}$$

where $\varkappa_0$ is the susceptibility of the material but is unrelated to the particular transition, $J_\nu(\Delta)$ is the Bessel function, $N$ is the density of particles executing the given transition, $\Delta\rho=\rho_{bb}-\rho_{aa}$ is the population difference between the corresponding energy levels, $\Delta=(d_{bb}-d_{aa})e_{10}/\hbar p$, $e_{10}$ is the component of the vector $\mathbf{e}_{10}$ along the $x$ axis, and $\tau_\perp$ is the characteristic relaxation time of the nondiagonal elements of the density matrix.

If the transition line is inhomogeneously broadened and the energy levels are degenerate, then (9) must include additional summation over $\omega_{ba}$ and the complete or partial removal of degeneracy by the Stark effect must be taken into account. Confining our attention to the resonance case $\omega\to\omega_{ba}$, and omitting the nonresonance term in (9), we obtain

$$\varkappa^{(mn)}(\omega)=\varkappa_0\delta_{mn}+N\sum_{s,\mu}\frac{|d_{ab}|^2}{\hbar}J_{s-m}(\Delta_\mu)J_{s-n}(\Delta_\mu)U_\mu(\omega+sp), \tag{10}$$

where

$$U_\mu(\Omega)=\int\frac{\Delta\rho_\mu(\omega_{ba})}{\Omega-\omega_{ba}+i/\tau_\perp}d\omega_{ba}; \tag{11}$$

in which the sum over $\mu$ is connected with the degeneracy $[\Delta_\mu=(d_{bb}^{(\mu)}-d_{aa}^{(\mu)})/\hbar p]$. This analysis includes the case where, in addition to the alternating external field $\mathbf{e}_1$, a constant external electric field $\mathbf{e}_0$, parallel to the $x$ axis, is also applied to the medium. In the latter case, the functions $U_\mu(\Omega)$ describe the profiles of the inhomogeneously broadened components of the Stark-split (in $\mathbf{e}_0$) transition line: $U_\mu(\Omega)=U(\Omega-\delta\omega_\mu)$. For the linear Stark effect, $\delta\omega_\mu\propto e_0$. The total width of the corresponding profile $U_\mu(\Omega)$ will, in general, be denoted by $\Delta\omega_t$.

The problem is thus to diagonalize the matrix $Q$ with allowance for (10). It has been shown[21] that the character of the solutions corresponding to Eq. (4) is different, depending on the parameter

$$\gamma=2\pi\omega|\varkappa^{(n0)}(\omega)|/p\varepsilon_0 \tag{12}$$

(we have in mind here the maximum value in $n$ for $n\neq0$, and the quantity $\varepsilon_0=1+4\pi\varkappa_0$ is assumed to be independent of $\omega$). For small values of this parameter, the corresponding solution was given in our previous paper[21] (the solution of a similar problem for electron plasma in a uniform external magnetic field $\mathbf{H}=\mathbf{H}_0-\mathbf{H}_1\times\cos pt$, the alternating component of which produces a time modulation of the frequency of cyclotron resonance, was obtained in another previous paper[13]). For such values of $\gamma$, the above solution remains valid even in the case of the degenerate states that we are considering and for an inhomogeneously broadened transition line:

$$E=E^{(0)}\left[e^{\bar\lambda_0 z}+\frac{4\pi}{\varepsilon_0}\sum_{n}^{(n\neq0)}\frac{k_n^2\varkappa^{(n0)}(\omega)}{k^2-k_n^2}(e^{\bar\lambda_0 z}-e^{\bar\lambda_n z})e^{-inpt}\right], \tag{13}$$

where

$$\bar\lambda_n=i(k_n-k)\varepsilon_0^{1/2}+i\frac{2\pi k_n\varkappa^{(nn)}(\omega)}{\varepsilon_0^{1/2}},\quad k=\frac{\omega}{c}. \tag{14}$$

We note that, according to (13), the modulation of the light wave transmitted by the layer of the medium under consideration is, in general, directly determined by the matrix $\varkappa^{(mn)}(\omega)$, so that measurements of this modulation can be used as a means of determining the susceptibility matrix. Measurements of the modulation for small values of $\gamma$ can therefore be used to carry out a more complete investigation of transitions in the medium as compared with currently used methods. We also note that measurements of this modulation provide us with a spectroscopy of transition lines at very low concentration of the particles in which the transitions take place.

## 3. SOLUTION OF THE PROBLEM FOR LARGE VALUES OF $\gamma$

We must now consider the case of large $\gamma$. This case can easily be reached in practice because the frequency $p$ of the rf field is smaller by several orders of magnitude than the frequency of the light wave. We shall consider $z \ll c/p$ and, correspondingly, we shall set $\beta = k$ in (4). The matrix $Q$ then takes the form

$$Q = -k^2(1+4\pi T), \tag{15}$$

i.e., the problem reduces to the diagonalization of the matrix $T$. Next, we suppose that the distance up to the frequencies of the Stark components neighboring a particular component $\mu = \tilde{\mu}$, i.e., the interval $|\delta\omega_{\tilde{\mu}} - \delta\omega_\mu|$, exceeds the corresponding interval $|p\Delta_{\tilde{\mu}}|$ of the modulation of the component $\delta\omega_{\tilde{\mu}}$ by the field $\mathbf{e}_1$:

$$|\delta\omega_{\tilde{\mu}} - \delta\omega_\mu| > |p\Delta_{\tilde{\mu}}|. \tag{16}$$

Under these conditions, when the frequency $\omega$ of the incident light wave satisfies the condition

$$|\omega - \omega_{ba}^{(0)} - \delta\omega_{\tilde{\mu}}| \leqslant p\Delta_{\tilde{\mu}} \tag{17}$$

where $\omega_{ba}^{(0)}$ is the center of the transition line under consideration in the absence of external fields $\mathbf{e}_0$ and $\mathbf{e}_1$, the expression given by (10) must contain only the resonance term corresponding to $\mu = \tilde{\mu}$:

$$\varkappa^{(mn)}(\omega) \approx \varkappa_0\delta_{mn} + N\sum_s \frac{|d_{nb}|^2}{\hbar} J_{s-m}(\Delta) J_{s-n}(\Delta) U(\omega + sp - \delta\omega_{\tilde{\mu}}). \tag{18}$$

For simplicity, we have omitted the subscript $\tilde{\mu}$ on $d_{ab}$ and $\Delta$.

It may easily be verified that the transformation $S$, that diagonalizes the matrix (18), and the inverse transformation $S^{-1}$, are such that

$$\langle S\rangle_{nl} = \langle S^{-1}\rangle_{ln} = J_{l-n}(\Delta). \tag{19}$$

Under these conditions, the matrix $\Lambda = S^{-1}QS$ is diagonal ($\Lambda_n = \Lambda_l\delta_{nl}$) and the elements $\Lambda_l$ have the form

$$\Lambda_l = -k^2\varepsilon_0 - 4\pi k^2 N |d_{ab}|^2 \hbar^{-1} U(\omega + lp - \delta\omega_{\tilde{\mu}}). \tag{20}$$

Equations (7), (8), (19), and (20) form the solution of the above problem for $\gamma \gg 1$ provided (16) and (17) are satisfied. Using (7) and (19), we can write the expressions for the time-dependent components of the characteristic waves in the medium in the form

$$\varphi_l(t) = \exp(-ilpt + i\Delta\sin pt). \tag{21}$$

Moreover, for weak resonant absorption,

$$2\pi N |d_{ab}|^2 \hbar^{-1} \varepsilon_0^{-1/2} U(\omega_{ba}^{(0)}) \ll 1, \tag{22}$$

which is of the greatest interest in practice, the expression for the coordinate components of the characteristic waves is

$$q_{1l}(z) = \exp(ik\varepsilon_0^{1/2} z + \lambda_l z), \tag{23}$$

where

$$\lambda_l = i\frac{2\pi k |d_{ab}|^2}{\hbar\varepsilon_0^{1/2}} NU(\omega + lp - \delta\omega_{\tilde{\mu}}). \tag{24}$$

It is clear from (23) and (24) that, for example, when

$$p \gtrsim \Delta\omega_t, \tag{25}$$

$$\Delta \gtrsim 1, \tag{26}$$

an appreciable dependence of $q_{1l}$ on $l$ can be seen over a characteristic length

$$z \sim z_x = [2\pi k |d_{ab}|^2 \hbar^{-1}\varepsilon_0^{-1/2} N \operatorname{Im} U(\omega_{ba}^{(0)} + \delta\omega_{\tilde{\mu}})]^{-1}, \tag{27}$$

which is also the characteristic length for resonant absorption in the absence of the alternating external field $\mathbf{e}_1$. According to (8b), this may be accompanied by appreciable modulation of the initially monochromatic light wave.

## 4. DYNAMIC ELECTROOPTICAL EFFECTS

Let us now consider in greater detail the solutions obtained for $\gamma \gg 1$. We suppose, to begin with, that the frequency $\omega$ of the incident light wave is a resonance frequency with respect to the frequency of the corresponding Stark component of the transition line in the constant field $\mathbf{e}_0$($\omega = \omega_{ba}^{(0)} + \delta\omega_{\tilde{\mu}}$). We shall consider the modulation of the wave intensity under conditions (25)–(27). This modulation can be easily examined by starting with (8a), (19), (21), and (23) where, in view of (25) and (27), we may substitute $q_{10} = \exp(ik\varepsilon_0^{1/2}z + \lambda_0 z)$ and $q_{1l} = \exp(ik\varepsilon_0^{1/2}z)$ for $l \neq 0$. For a Doppler-broadened transition line at $p \geqslant 1.25\Delta\omega_t$, $0.5 \leqslant \lambda_0 z \leqslant 1.5$, the expression obtained in this way is accurate to within 0.3%. The corresponding dependence of $|E|^2$ on $t$ for $z = 0.94 z_x$ is shown by the solid curve in Fig. 1. It is clear that several intensity pulsations are present within each period $2\pi/p$.

It is interesting to compare this modulation picture

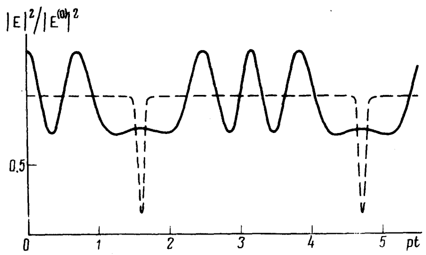


FIG. 1. Relative intensity of transmitted light wave as a function of time (solid curve). Broken curve shows the corresponding dependence predicted by the quasistatic expression.

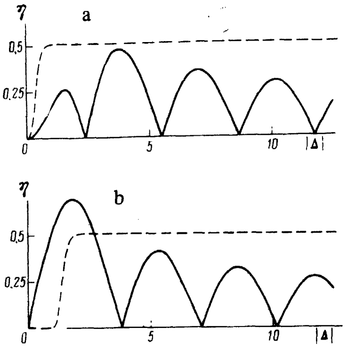


FIG. 2. Relative modulation of the transmitted light wave as a function of the amplitude of the external alternating electric field: a—incident wave frequency $\omega = \omega_{ba}^{(0)} + \delta\omega_{\bar{\mu}}$; b—incident light wave frequency $\omega = \omega_{ba}^{(0)} + \delta\omega_{\bar{\mu}} \pm p$ ($p$ is the frequency of the external electric field). Broken curves show the corresponding results predicted by the quasistatic expression.

with the corresponding picture predicted by the well-known quasistatic expression

$$E = E^{(0)} \exp[ik\varepsilon_0^{1/2} z + i\Delta n(t) kz], \tag{28}$$

where

$$\Delta n(t) = \frac{2\pi N |d_{ab}|^2}{\hbar \varepsilon_0^{1/2}} U(\omega - \delta\omega_{\bar{\mu}} + p\Delta \cos pt)$$

is the instantaneous value of the complex refractive index in the modulating field. The corresponding dependence of $|E|^2$ on $t$ is shown in Fig. 1 by the broken curve. As can be seen, the dependence represented by the solid curve is very different from that shown by the broken curve. This difference is due to the dynamic effects which occur in the propagating wave under condition (25).

Let us now consider the relative intensity modulation

$$\eta = (|E|^2_{max} - |E|^2_{min}) / |E^{(0)}|^2$$

as a function of $\Delta$. This modulation is practically zero ($\eta \approx 0$) when $\Delta$ is any of the roots of the Bessel function $J_0(\Delta)$, and is appreciable between the roots. The dependence of $\eta$ on $|\Delta|$ for $z = 0.36 z_x$, $p \geqslant 1.25\Delta\omega_t$ is shown in Fig. 2a by the solid curve. For comparison, the broken curve in this figure shows the dependence predicted by the corresponding quasistatic approximation. As can be seen, the "dynamic" curve oscillates along the $\Delta$ axis, whereas the quasistatic curve gives a monotonic dependence. It is also clear that the minima on the solid curve are very sharp, so that the positions of these minima can be determined with high precision. In the above example, the values of $\eta$ at these minima do not exceed 0.003.

We note that similar dynamic effects can also be seen whenever the frequency $\omega$ of the incident light wave differs from the resonance value by an integral number of frequencies of the modulating field:

$$\omega \approx \omega_{ba}^{(0)} + \delta\omega_{\bar{\mu}} + \nu p \tag{29}$$

($\nu = 0, \pm 1, \pm 2, \ldots, |\nu| \lesssim |\Delta|$). In this more general case, we have from (8a), (19), (21), and (23), using (25) and (27),

$$|E|^2 \approx |E^{(0)}|^2 |1 - (1 - e^{\lambda_\nu z}) J_\nu(\Delta) e^{-i\nu pt + i\Delta \sin pt}|^2. \tag{30}$$

In particular, it follows from (30) that, when $\nu \neq 0$, the dependence of the modulation parameter $\eta$ on $\Delta$ is also an oscillating function, and the minima of $\eta$ are determined from the condition $J_\nu(\Delta) = 0$ and are again sharp. As an example, the solid curve in Fig. 2b shows $\eta$ as a function of $|\Delta|$ under the exact relationship $\omega = \omega_{ba}^{(0)} + \delta\omega_{\bar{\mu}} \pm p$ ($|\nu| = 1$) and $z = 0.36 z_x$ (the broken curve shows the quasistatic approximation).

We now consider the average (over the period of the modulating field) relative intensity of the field oscillations in the wave

$$W(z) = \frac{p}{2\pi |E^{(0)}|^2} \int_0^{2\pi/p} |E|^2 \, dt.$$

In other words, the quantity $W$ is the transmission coefficient (in fact, the intensity transmission coefficient) of the medium of thickness $z$ for these particular waves. In general, we have from (8a), (19), (21), and (23)

$$W = \sum_l J_l^2(\Delta) \exp(2 \operatorname{Re} \lambda_l z). \tag{31}$$

In particular, under the conditions given by (25), (26), (27), and (29), we have approximately

$$W \approx 1 - [1 - \exp(2 \operatorname{Re} \lambda_\nu z)] J_\nu^2(\Delta). \tag{32}$$

It is clear from this expression that $W$ is also an oscillating function of $\Delta$, the maxima of which are determined by the roots of the equation $J_\nu(\Delta) = 0$. Thus, the maxima of the mean intensity of the transmitted wave correspond to minima on its modulation depth.

Figure 3a shows $W$ as a function of $|\Delta|$ for $z = z_x$ and

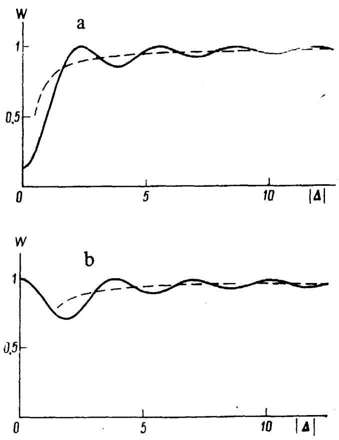


FIG. 3. Intensity absorption coefficient for the light wave as a function of the amplitude of the external alternating electric field: a—incident wave frequency $\omega = \omega_{ba}^{(0)} + \delta\omega_{\bar{\mu}}$; b—incident light wave frequency $\omega = \omega_{ba}^{(0)} + \delta\omega_{\bar{\mu}} \pm p$. Broken curves are predicted by the quasistatic expression.

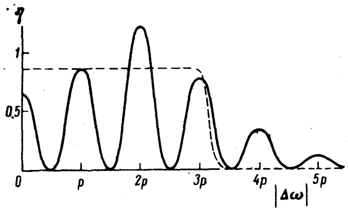


FIG. 4. Relative intensity modulation in the transmitted light wave as a function of the frequency difference ($\Delta\omega = \omega - \omega_{ba}^{(0)} - \delta\omega_{\bar{\mu}}$) between the incident light wave and the corresponding Stark component of the transition line in an external constant electric field. Broken curve corresponds to the quasistatic expression.

exactly satisfies the relation $\varphi = \omega_{ba}^{(0)} + \delta\omega$. The broken line in this figure shows, for comparison, the corresponding dependence obtained in the quasistatic approximation. Similar results for $\omega = \omega_{ba}^{0} + \delta\omega_{\bar{\mu}} \pm p$, $z = z_x$ are shown in Fig. 3b. As can be seen, the broken curves are not oscillating functions. Thus, the oscillations in $W$ as a function of $\Delta$ (solid curves) are also a dynamic effect.

Finally, consider $\eta$ and $W$ as functions of the frequency $\omega$ of the incident wave (or the frequency of the Stark component $\delta\omega_{\bar{\mu}}$) for fixed $\Delta$, i.e., for a fixed amplitude and frequency of the modulating field. It is readily seen from (30) and (32) that, when (25)–(27) are satisfied, this dependence is again an oscillating function and the maxima of $\eta$ and the minima of $W$ correspond to (29). The corresponding dependence of $\eta$ and $W$ on $\Delta\omega = \omega - \omega_{ba}^{(0)} - \delta\omega_{\bar{\mu}}$ for $|\Delta| = 3$, $p = 2.5\Delta\omega_t$, and $z = z_x$ is shown in Figs. 4 and 5, respectively, by the solid curves. The dashed curves show the corresponding results based on the quasistatic expression. Since the dashed curves are not oscillatory, it is clear that the oscillations in $\eta$, $W$ as functions of $\omega$ (solid curves) constitute another dynamic electrooptical effect. We also note that the minima in $\eta$ and the maxima in $W$ correspond to

$$\omega \approx \omega_{ba}^{(0)} + \delta\omega_{\bar{\mu}} + (l + 1/2)\,p \tag{33}$$

($l = 0, \pm 1, \pm 2, \ldots$), and, moreover, the magnitude of $\eta$ at the minima does not exceed 0.01 in the above example, whereas the magnitude of $W$ at the maxima is close to unity and does not differ from it by more than 0.001. It is also interesting to note that appreciable wave intensity modulations are observed within the detuning range $|\Delta\omega|$ exceeding the transition frequency modulation interval by roughly a factor of two (see Fig. 4). On the other hand, in the quasistatic description, these intervals are practically equal.

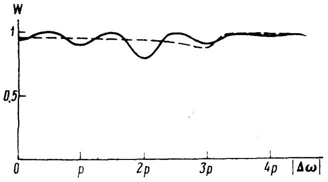


FIG. 5. Light wave intensity transmission coefficient as a function of the difference ($\Delta\omega = \omega - \omega_{ba}^{(0)} - \delta\omega_{\bar{\mu}}$) between the frequencies of the incident light wave and the corresponding Stark component of the transition line in a constant external electric field. The broken curve corresponds to the quasistatic expression.

The above effects have been examined for the case of a homogeneous medium and, in particular, it was assumed that the constant external field $\mathbf{e}_0$ was uniform. The influence of any nonuniformity of this field along the $z$ axis can also be taken into account. Consider the case where the inhomogeneity of the medium may be due to the dependence of $\varkappa_0$, $N$, $\delta\omega_\mu$, and $\Delta\omega_t$ on $z$. We also suppose that the frequency interval between the chosen component $\mu = \bar{\mu}$ and the neighboring Stark components (i.e., the quantity $|\delta\omega_{\bar{\mu}} - \delta\omega_\mu|$) exceeds both the interval $|p\Delta_{\bar{\mu}}|$ of the modulation of the component $\delta\omega_{\bar{\mu}}$ by the field $\mathbf{e}_1$ and the interval of the variations $\delta_z\omega_{\bar{\mu}}$ of this component in $z$ due to the nonuniformity of the constant field $\mathbf{e}_0$:

$$|\delta\omega_{\bar{\mu}} - \delta\omega_{\mu}| > |p\Delta_{\bar{\mu}}|, \quad |\delta_z\omega_{\bar{\mu}}|.$$

In this case, when the frequency $\omega$ of the incident light wave satisfies (17), and in (10) we can retain only the resonance term corresponding to $\mu = \bar{\mu}$, we arrive at the expression given by (18) in which $\varkappa_0$, $N$, $\delta\omega_{\bar{\mu}}$, and $\Delta\omega_t$ may depend on $z$. Next, using the $z$-independent transformation (19), we obtain (8) and (21), where the functions $q_{1l}(z)$ are determined by

$$d^2q_{1l}/dz^2 = \Lambda_l q_{1l} \tag{34}$$

subject to the boundary condition $q_{1l}(0) = 1$, $q_{1l}(+\infty) = 0$. The quantities $\Lambda_l$ in these equations are given by (20) and, in the present case of an inhomogeneous medium, are functions of $z$.

Thus, the original time-dependent problem has been reduced to a system of stationary equations (34). If we consider the case of a smooth inhomogeneity, as compared with the length of the light wave, due to the inhomogeneity of the field $\mathbf{e}_0$, and if (22) is satisfied, we can readily obtain the expression given by (23) for $q_{1l}(z)$ in the geometric-optics approximation in which we must now use a more general expression for $\lambda_l$ than is given by (24), namely:

$$\lambda_l = i\frac{2\pi k N |d_{ab}|^2}{z\hbar \varepsilon_0^{1/2}} \int_0^z U(\omega + lp - \delta\omega_{\bar{\mu}})\, dz. \tag{35}$$

It is clear from this expression that the nonuniformity of the external constant field $\mathbf{e}_0$ leads to a weakening of the above effects of electrooptical oscillations. For example, for a constant gradient $\partial e_0/\partial z$, these effects are practically absent if the interval of variation in the frequency of the Stark component ($|\delta_z\omega_{\bar{\mu}}|$) over the complete length of observation due to the nonuniformity exceeds the interval ($p\Delta_{\bar{\mu}}$) of modulation of this frequency by the field $\mathbf{e}_1$. The above effects are, therefore, most clearly defined in a homogeneous medium.

## 5. THE CASE OF A GAS OF MOLECULES OF THE SYMMETRIC SPINNING-TOP TYPE

As an example of a medium in which the above effects can be observed, consider a gas of molecules of the symmetric spinning-top type. We shall be interested in molecular transitions with a change in the vibrational and rotational quantum numbers, $v$ and $J$, respectively. To be specific, let us consider the transition $v, J \to v+1, J+1$ which is subject to the linear Stark effect (an analogous analysis can be given for the $v, J \to v+1, J-1$ transition). In this case, the subscript $a$ in the initial state is a set of two numbers, namely, $v, J$, whilst the subscript $b$ in the final state represents $v+1, J+1$, so that

$$\omega_{ba}^{(0)}=(E_{v+1,J+1}^{(0)}-E_{v,J}^{(0)})/\hbar.$$

In the constant electric field $\mathbf{e}_0$, the transition frequency splits into a series of Stark components $\omega_{ba}^{(\mu)}=\omega_{ba}^{(0)}+\delta\omega_\mu$ differing by the values of $\mu$ and, if the Stark splitting energy exceeds appreciably the hyperfine interaction energy and is much smaller than the separation between the rotational levels, the expression for $\delta\omega_\mu$ can be written in the form[22]

$$\delta\omega_{MK}=\frac{2MKde_0}{J(J+1)(J+2)\hbar}, \tag{36}$$

where $d$ is the absolute magnitude of the dipole moment of the molecule. As can be seen, the subscript $\mu$ corresponds to the set of two numbers in this case, namely, $M, K$ ($M\hbar$ is the component of the total angular momentum along the direction of the field $\mathbf{e}_0$ and $K\hbar$ is the component of this angular momentum along the molecular axis; $M, K=0, \pm1, \pm2, \ldots, \pm J$). The matrix elements $d_{ab}^{(MK)}$ of the dipole-moment operator for the corresponding transitions are given by[22]

$$|d_{ab}^{(MK)}|^2\sim\frac{[(J+1)^2-M^2][(J+1)^2-K^2]}{(J+1)^2(2J+1)(2J+3)}d^2. \tag{37}$$

Next, if we consider the field $\mathbf{e}_1$, we obtain the analog of (36) for $\Delta_{MK}$:

$$\Delta_{MK}=-\frac{2MKde_{10}}{J(J+1)(J+2)\hbar p}. \tag{38}$$

It is clear from this expression that the degeneracy in the subscripts $M, K$ is partly removed and partly remains. Assuming, for convenience, that different values of the subscript $\mu$ correspond to different values of the product $MK$ (for example, by assuming that $\mu=MK$), we shall take for $|d_{ab}^{(\mu)}|^2$ in (10) the sum of all values $|d_{ab}^{(MK)}|^2$ corresponding to the particular value of the product $MK$.

Let us consider in greater detail the given transition from the rotational level $J=1$. In this case, $\mu=0, \pm1$, i.e., the transition line splits into three components in the electric field. Suppose, for example, that $\tilde{\mu}=1$, i.e., consider the component with the maximum frequency. In (18), we then have

$$\Delta=-de_{10}/3\hbar p,\quad |d_{ab}|^2\sim0{,}3d^2,\quad \delta\omega_{\tilde{\mu}}=de_0/3\hbar.$$

Let us now take a numerical example for the $CH_3CN$ gas, the molecules of which have dipole moment $d=3.92\times10^{-18}$ e.s.u.[22] When the frequency of the incident wave is close to the frequency $\omega_4$ of the molecular vibrational mode (when this is so, $\omega_{ba}^{(0)}\approx1.73\times10^{14}$ rad/sec) and the gas temperature is $T=290\,^\circ$K, the Doppler absorption linewidth corresponding to pressures $P\lesssim1$ Torr in the absence of the external fields $\mathbf{e}_0$, $\mathbf{e}_1$ and the profile

$$\operatorname{Im} U\propto\exp[-(2\Delta\omega/\Delta\omega_t)^2]$$

($\Delta\omega=\omega-\omega_{ba}^{(0)}$) is $\Delta\omega_t\approx3.53\times10^8$ rad/sec. In accordance with the foregoing, we can then set $p=1.25\Delta\omega_t\approx4.3\times10^8$ rad/sec and, hence, we find that $|\Delta|\approx2.88e_{10}$ e.s.u. For example, when $e_{10}\sim0.3$–$3$ e.s.u., we have $|\Delta|\sim1$–$10$. Using (16), we find that the magnitude of the constant field $\mathbf{e}_0$ should exceed $|e_{10}|$. The resonance absorption length $z$ will, in general, depend on the gas pressure and the composition of the mixture, and may vary within broad limits. For example, for pure $CH_3CN$ gas at $P\sim0.1$–$0.01$ Torr, estimates of this length for the $v=0\to v=1$ transition yield $z_x\sim0.3$–$3$ cm. The wavelength of the modulating field ($\lambda\sim5\times10^2$ cm) is then found to exceed the above figure by two or three orders of magnitude (i.e., $\gamma\gg1$).

Let us now consider possible applications of the above effects. We note, above all, that the dependence of the light transmission coefficient of the medium on the frequency of the incident wave is a quasiperiodic function (Fig. 5) with a typical "period" of $10^9$ rad/sec when the layer width is $z\lesssim1$ cm. This property may be used in the selection of axial modes in laser cavities under high resolution. The above effect can also be used to produce various types of modulation in the light beam and to control the operation of lasers by varying the rf field parameters. Next, for an rf field of fixed frequency and amplitude, these effects can be used for the analysis of time deviations of the frequency of the incident light wave and, conversely, for fixed parameters of the incident wave they can be used to analyze the time variations of the rf field frequency. We also note that the effects can be used in high-resolution spectroscopy. For example, owing to the fact that the modulation parameter of the light wave plotted as a function of $\Delta$ (Figs. 2a and b) exhibits sharp minima, these minima can be used to determine accurate values for the Stark constants of vibrational-rotational transitions, to identify these transitions, and so on.


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# Role of collective and induced processes in the generation of Mössbauer $\gamma$ radiation

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It is shown that the principal role in the generation of coherent Mössbauer coherent radiation is played by processes of collective spontaneous radiation. Observation of induced Mössbauer $\gamma$ radiation in the decay of strongly excited polyatomic systems encounters unsurmountable difficulties.

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The question of the possibility of extending the principle of laser generation of the $\gamma$-ray band is constantly discussed in the literature of the last fifteen years (see the reviews[1–3]). It was assumed that the main idea of lasing in the optical band, i.e., amplification of the light with the aid of stimulated emission, could be directly realized also in the $\gamma$ band. No account was taken, however, of the peculiarities of the electromagnetic waves in the $\gamma$ band, or of the peculiarities of the Mössbauer $\gamma$ radiation. The need for using the latter has already been repeatedly emphasized. In the present article we consider in greater detail the kinetics of emission of extended resonatorless systems of two-level emitters that are strongly excited and are uncorrelated at the initial instant, and show that in the Mössbauer energy region the stimulated processes make a negligibly small contribution to the radiation intensity. The principal role is played here by processes of collective spontaneous emission, which replace stimulated emission when it comes to generation of coherent $\gamma$ photons.

The semiclassical approximation of the quantum equations of field dynamics is of the form (for a detailed derivation see[4]):

$$\frac{dn}{dt}+\frac{n}{\tau}=F,\quad \frac{dF}{dt}+\frac{1}{2}\left(\frac{1}{\tau}+\frac{1}{T_2}\right)F=\frac{1}{T_0^2}(nR+S),$$
$$\frac{dS}{dt}+\frac{S}{T_2}=FR-\varkappa F,\quad \frac{dR}{dt}=-2F, \tag{1}$$

where

$$n=\sum_{\mathbf{k}}\langle a_{\mathbf{k}}^{+}a_{\mathbf{k}}\rangle,\quad R=\sum_{i=1}^{N}\langle\sigma_z^{(i)}\rangle$$

are respectively the number of quanta in the sample volume and the population difference; $\tau=L/c$; $1/T_2$ is the width of the Mössbauer transition line;

$$\frac{1}{T_0^2}=\frac{2\overline{|g_{\mathbf{k}}|^2}}{\hbar^2}=\frac{4\pi f}{V\hbar\omega}\overline{|M|^2};$$

$V$ is the volume of the sample; $f$ is the probability of the Mössbauer radiation; $M$ is the matrix element of the nuclear-transition current density; the bar denotes averaging over the directions of $\mathbf{k}$: $\sigma_+$, $\sigma_-$, $\sigma_z$ are Pauli spin operators;

$$F=\frac{i}{\hbar}\sum_{\mathbf{k}}\sum_{i}\left(g_{\mathbf{k}}a_{\mathbf{k}}^{+}\sigma_-^{(i)}\exp(i\mathbf{k}\mathbf{r}_{0i})-\text{H.c.}\right), \tag{2}$$

$$S=\varkappa N_2+\sum_{\mathbf{k}}\sum_{i\neq j}\langle\sigma_+^{(i)}\sigma_-^{(j)}\rangle\exp[-i\mathbf{k}(\mathbf{r}_{0i}-\mathbf{r}_{0j})], \tag{3}$$

$$\varkappa=\sum_{\mathbf{k}}1=8\pi\frac{V\omega^2}{(2\pi c)^3}\Delta\omega=\frac{V\omega^2}{2\pi c^3\tau}. \tag{4}$$

Summation with respect to $\mathbf{k}$ is carried out over a spherical layer of thickness

$$\Delta k=\frac{\pi}{2c}\left(\frac{1}{\tau}+\frac{1}{T_2}\right),$$

and account is taken in (4) of the fact that $\tau\ll T_2$ for the

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