Nonlinear-optics phenomena due to dissipative or nonstationary processes

N. L. Manakov and A. G. Faĭnshteĭn

Voronezh State University
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It is shown that dissipative and nonstationary processes occurring during the propagation of light in a medium give rise to new nonlinear-optics effects, including optical rectification in a homogeneous isotropic medium and magnetization of anisotropic media by linearly polarized radiation. A classical and a quantum-mechanical analysis of these effects is given. The corresponding susceptibilities are calculated for an atomic gaseous medium. The density matrix is obtained for the steady state of the atom in a nonresonant light field.

§ 1. INTRODUCTION

It is well known that, in linear optics, absorption processes do not reduce only to the dissipation of light energy:

$$dW/dt = -\gamma W.$$

The presence of the T-odd (i.e., changing sign under time reversal) parameter γ may give rise to phenomena that are qualitatively different from those in nondissipative media [for example, elliptical polarization on reflection of linearly polarized light (Ref. 1, § 86), and the Maxwell effect (Ref. 1, § 102; see also the recent Ref. 2)]. In nonlinear optics, we again encounter a variety of phenomena due to dissipation (thermal self-focusing, photovoltaic effects, and so on) which, even though they were discovered relatively recently, have already found important practical applications. However, dissipation effects are commonly neglected in phenomenological studies of the properties of nonlinear susceptibilities, so that thermodynamic concepts can be used to establish relationships between phenomena of different physical nature.[1,3,4] When dissipation is taken into account, analysis based exclusively on consideration of spatial-symmetry and time-reversal properties does not yield such detailed information; nevertheless, it is often a very convenient means of elucidating the conditions under which a particular effect will arise,[9] and can often predict new phenomena.[6]

In this paper we consider the appearance of constant electric and magnetic moments **P** and **M** in a medium exposed to an electromagnetic wave. Inclusion of dissipation in the phenomenological description (§ 2) suggests that there are new nonlinear-optics phenomena, including optical rectification in a homogeneous isotropic medium and magnetization of anisotropic media by linearly polarized light, in which the induced **P** and **M** are proportional to γ. This T-odd parameter may also be due to processes of a different nature, including nonstationary processes, for example, those involving the ionization of atoms by the light field (it is precisely the ionization width that is responsible for the appearance of some unusual terms in the hyper-Raman cross section[7]). However, the most interesting practical effects are those produced by pulsed light fields for which the T-odd parameter is determined by the curvature of the envelope F of the light pulse:

$$\lambda = F^{-1}(dF/dt).$$

The induced moments **P**, **M** are then proportional to λ, and, together with $\lambda = \lambda(t)$, change sign during the propagation of the pulse.

The specific mechanisms responsible for these effects are elucidated in § 3 in terms of a simple classical model of the medium, namely, a set of noninteracting oscillators. Section 4 gives a quantum-mechanical calculation of the effects produced by pulsed fields, and gives numerical estimates for monatomic gases. Rigorous quantum-mechanical inclusion of dissipation involves the use of the density-matrix formalism and is employed in § 5 to calculate **P** and **M** for a gaseous medium when dissipation is due to nonresonant scattering of light.

§ 2. PHENOMENOLOGICAL ANALYSIS

Consider an infinite medium in which there are constant uniform electric and magnetic fields $\mathbf{F}_0$ and $\mathbf{B}_0$ and a propagating electromagnetic wave with electric vector

$$\mathbf{F}(\mathbf{r}, t) = \frac{1}{2}\{\mathbf{F}e^{i(\mathbf{kr}-\omega t)} + \text{c.c.}\}. \tag{1}$$

The T-even pseudoscalar (Stokes parameter)

$$\xi_2 = i\frac{\mathbf{k}}{k}\frac{[\mathbf{F}\times\mathbf{F}^*]}{\mathbf{F}\mathbf{F}^*}, \quad -1 \leqslant \xi_2 \leqslant 1$$

defines the degree of circular polarization of the wave.[8] In the absence of external fields, the medium will initially be considered to be homogeneous, isotropic, and centrally symmetric, so that its macroscopic properties can be specified by purely scalar parameters.

The polarization **P** and magnetization **M** induced in the medium must be expressible in terms of combinations of the vectors $\mathbf{F}_0$, $\mathbf{B}_0$, $\mathbf{F}$, $\mathbf{F}^*$, and $\mathbf{n} = \mathbf{k}/k$, and scalar parameters of the medium such as the T-even generalized susceptibilities $\chi, \eta, \ldots$ and the T-odd parameter γ defining, for example, the dissipation of light energy.

For a polar T-even electric polarization vector, the phenomenological expansion is

$$\mathbf{P} = \gamma\chi_1(\mathbf{F}\mathbf{F}^*)\mathbf{n} \tag{2a}$$

$$+\chi_2(\mathbf{F}\mathbf{F}^*)\mathbf{F}_0 + \chi_3\,\mathrm{Re}\,\{(\mathbf{F}_0\mathbf{F})\mathbf{F}^*\} + \gamma\chi_4\xi_2(\mathbf{F}\mathbf{F}^*)[\mathbf{F}_0\times\mathbf{n}] + \ldots, \tag{2b}$$

where we have retained only time-independent components of the vector **P** that are linear in the wave intensity, so that terms including $\mathbf{F}^2$ or $\mathbf{F}^{*2}$, which oscillate with frequency 2ω, are absent.

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Even the first term in (2) gives rise to a new phenomenon in isotropic media, namely, static polarization induced by an alternating field. This so-called optical rectification effect was previously considered to be possible only in piezoelectric or inhomogeneous media.[1]

The various terms in (2b) can be written in tensor form as $\alpha_{ij}(\mathbf{F}_0)_j$ where $\alpha_{ij} \propto |\mathbf{F}|^2$ is the correction to the static polarizability which takes into account the anisotropy induced by the alternating field in the medium. The dissipation term containing χ_4, which is nonzero only for $\xi_2 \neq 0$, gives rise to the antisymmetric part α_{ij}. The latter would seem to be inconsistent with symmetry conditions governing the static permittivity, which are established independently of the presence of T-odd parameters (for example, of the magnetic field $\mathbf{B}_0$) in the problem. However, these conditions follow from thermodynamic relationships for the free energy[1] that cannot be introduced when dissipative processes are taken into account.

The higher-order terms in (2), which contain γ, are also "unusual," but they are mainly responsible for corrections to the above effects, and are hardly of independent interest.

In the phenomenological expansion of the time-independent part of the magnetization $\mathbf{M}$ (a T-odd axial vector), the first term that is linear in intensity has the form $\eta\xi_2(\mathbf{FF}^*)\mathbf{n}$. It determines the inverse Faraday effect (magnetization of the medium by a circularly polarized field; Ref. 1, § 101) and is not connected with dissipation. The next terms are of the form given by (2b) with $\mathbf{F}_0$ replaced with $\mathbf{B}_0$, and can be interpreted as tensor corrections to the static magnetic susceptibility with antisymmetric part proportional to γ.

The most interesting result for $\mathbf{M}$ would seem to be connected with dissipation, and appears when we analyze the higher-order terms containing $\mathbf{F}_0^2$:

$$\mathbf{M}=\xi_2(\mathbf{FF}^*)\{\eta_1\mathbf{F}_0^2\mathbf{n}+\eta_2(\mathbf{F}_0\mathbf{n})\mathbf{F}_0\} +\gamma\eta_3\,\mathrm{Re}\,\{(\mathbf{F}_0\mathbf{F})[\mathbf{F}_0\times\mathbf{F}^*]\}. \quad (3)$$

The terms containing η_1, η_2 in this expression do not contain γ and produce the usual corrections $\sim \mathbf{F}_0^2$ to the inverse Faraday effect with allowance for anisotropy induced in the medium by the constant field $\mathbf{F}_0$. These corrections and the effect itself vanish for linearly polarized waves ($\xi_2 = 0$). However, the last term in (3), which contains γ, remains nonzero so that (for $\xi_2 = 0$, we put $\mathbf{F} = \mathbf{F}^*$)

$$\mathbf{M}=\eta_3\gamma(\mathbf{F}_0\mathbf{F})[\mathbf{F}_0\times\mathbf{F}]. \quad (4)$$

Thus, when dissipatation is present, the medium can become magnetized by the linearly polarized field. The magnetization (4) is perpendicular to the plane containing the vectors $\mathbf{F}_0$ and $\mathbf{F}$ and its magnitude is a maximum when $\mathbf{F}_0$ and $\mathbf{F}$ are at an angle $\pm \pi/4$ to one another (when $\mathbf{F}_0 \| \mathbf{F}$ and $\mathbf{F}_0 \perp \mathbf{F}$, we have $\mathbf{M} = 0$).

In spatially inhomogeneous and anisotropic media, when the expansion for $\mathbf{P}$ and $\mathbf{M}$ includes tensor characteristics of the medium, effects similar to those examined above may arise even in the absence of constant external fields. For example, the magnetization of a uniaxial crystal described by the tensor $\nu_i\nu_j$ (the vector $\boldsymbol{\nu}$ defines the direction of the anisotropy axis) in a linear field is given by an expression obtained from (4) by replacing $\mathbf{F}_0$ with $\boldsymbol{\nu}$ (this vector can also be the normal to the separation boundary between the two media, and so on).

It is also important to note that, in an anisotropic centrally symmetric medium, the optical rectification effect is possible even in the absence of dissipation in the case of nonzero circular polarization:

$$\mathbf{P}=\chi\xi_2(\mathbf{FF}^*)(\boldsymbol{\nu}\mathbf{n})[\boldsymbol{\nu}\times\mathbf{n}]. \quad (5)$$

As far as we know, the effect described by (5) was previously not discussed in the literature.

The expressions for the new nonlinear-optics effects given in this section were based exclusively on space-time parity considerations. To verify that such effects do actually occur (i.e., that the corresponding χ and η are nonzero), we must examine specific models of dissipative media. In the following sections, we shall confine our attention to the effects described by (2a) and (4).

§ 3. CLASSICAL MODEL OF THE MEDIUM

The mechanisms that lead to (2a) and (4) are essentially classical, and can be elucidated in terms of a simple oscillator model with friction:

$$m\ddot{\mathbf{r}}+\gamma\dot{\mathbf{r}}+\varkappa\mathbf{r}=\mathbf{f}. \quad (6)$$

The Lorentz force $\mathbf{f}$, acting in the wave field (1) on the oscillating charge e, has the form

$$\mathbf{f}(\mathbf{r},t)=\frac{e}{2}\left\{\mathbf{F}+\frac{1}{c}[\dot{\mathbf{r}}\times[\mathbf{n}\times\mathbf{F}]]\right\}e^{i(\mathbf{kr}-\omega t)}+\text{ c.c.},\quad \mathbf{n}=\frac{\mathbf{k}}{k}. \quad (7)$$

It depends on the position $\mathbf{r}$ and velocity $\dot{\mathbf{r}}$ of the charge, and this is responsible for the nonlinearity of the problem. The problem is linear in the dipole approximation

$$\mathbf{f}(\mathbf{r},t)=\mathbf{f}_d(t)=e\,\mathrm{Re}[\mathbf{F}\exp(-i\omega t)]$$

and the solution of (6) has the well-known form

$$\mathbf{r}_0(t)=\frac{1}{2e}\alpha(\omega)\mathbf{F}e^{-i\omega t}+\text{ c.c.},\quad \alpha(\omega)=e^2[\varkappa-m\omega^2-i\omega\gamma]^{-1}, \quad (8)$$

where α is the polarizability of the oscillator.

The anisotropy induced in the medium by the constant field $\mathbf{F}_0$ can be effectively taken into account by assuming that $\varkappa$ is a tensor, i.e.,

$$\varkappa_{ij}=\varkappa_\perp\delta_{ij}+(\varkappa_\|-\varkappa_\perp)\nu_i\nu_j,\quad \boldsymbol{\nu}=\mathbf{F}_0/F_0,\quad \varkappa_\|-\varkappa_\perp\propto F_0^2\,. \quad (9)$$

The same model describes a uniaxial crystal with axis along $\boldsymbol{\nu}$). The principal values $\varkappa_\|$ and $\varkappa_\perp$ of the tensor (9) determine the eigenfrequencies $\omega_\|$ and $\omega_\perp$ of the oscillator along the anisotropy axis ($\varkappa_\| = m\omega_\|^2$) and in the plane perpendicular to it ($\varkappa_\perp = m\omega_\perp^2$). The polarizability α is then also a tensor such as (9) with principal values

$$\alpha_{\|,\perp}=e^2[m(\omega_{\|,\perp}^2-\omega^2)-i\omega\gamma]^{-1}. \quad (10)$$

A. Magnetization of a medium by linearly polarized radiation

In the general case of an elliptically polarized wave, the end point of the vector $\mathbf{r}_0(t)$ given by (8) traces out an ellipse. In the linear field $\mathbf{F}(t) = \mathbf{F}\cos\omega t$, on the other hand, for which $\gamma = 0$, the oscillations of the charge occur along a straight line that does not lie along $\mathbf{F}$ because of anisotropy.

The oscillations of the charge lag in phase behind the oscillations of $\mathbf{F}(t)$ when friction is present. Since the phases of the complex principal values $\alpha_{\parallel}$ and $\alpha_{\perp}$ are different, the phases of the components of $\mathbf{r}_0(t)$ along and perpendicular to the anisotropy axis are also different. This means that, when friction is present, the end point of the vector $\mathbf{r}_0(t)$ traces out an ellipse even in the case of linearly polarized waves. It is clear that the oscillator has then a magnetic moment. To show this, consider a linear field and the average value of $\boldsymbol{\mu} = (e/2c)\mathbf{r}_0\times\dot{\mathbf{r}}_0$ over one period. Using (8) and (10), we obtain

$$\bar{\boldsymbol{\mu}}=\gamma\eta_\gamma(\mathbf{v}\mathbf{F})[\mathbf{v}\times\mathbf{F}],$$
$$\eta_\gamma=\frac{\omega\,\mathrm{Im}(\alpha_\perp\alpha_\parallel^*)}{2ec\gamma}=\frac{m\omega^2(\omega_\parallel^2-\omega_\perp^2)}{2e^5c}|\alpha_\parallel\alpha_\perp|^2 \qquad (11)$$

which agrees with (4).

The phase shift between the oscillator and field oscillations will also occur in the nonstationary problem without friction when one of the parameters of the medium or field, for example, the wave amplitude, is a function of time:

$$dF/dt=\lambda F,\quad |\lambda|\ll\omega,\ |\omega-\omega_{\parallel,\perp}|. \qquad (12)$$

The magnetic moment of the oscillator is then

$$\bar{\boldsymbol{\mu}}=\lambda\eta_\lambda(\mathbf{v}\mathbf{F})[\mathbf{v}\times\mathbf{F}],$$
$$\eta_\lambda=\frac{m\omega^2}{ce^3}\alpha_\parallel\alpha_\perp(\alpha_\perp-\alpha_\parallel)=\frac{m^2\omega^2(\omega_\parallel^2-\omega_\perp^2)}{ce^5}(\alpha_\parallel\alpha_\perp)^2. \qquad (13)$$

Expressions (11) and (13) can be used to estimate the order of magnitude of the magnetization $\mathbf{M} = n\bar{\boldsymbol{\mu}}$ per unit volume of the medium (n is the density of atoms or molecules of the material) if m and e are interpreted as the mass and charge of the electron and $\omega_{\parallel,\perp}$ as the atomic frequency or frequency of interband transitions in the solid. In crystals, the anisotropy $(\omega_\parallel - \omega_\perp)/\omega_{\parallel,\perp}$ is usually $\sim 10^{-2}$. The anisotropy induced by a constant field is of the order of $(F_0/F_{\mathrm{at}})^2$, where F_{at} is the characteristic intra-atomic field. In the neighborhood of a resonance in a monatomic gas

$$\omega-\omega_{\parallel,\perp}\approx\Delta,\quad \omega_{\parallel,\perp}\gg|\Delta|\gg\gamma/m,\lambda,|\omega_\parallel-\omega_\perp| \qquad (14)$$

the polarizability is $\alpha_{\parallel,\perp} \sim a_0^3\omega/\Delta$ (a_0 is the Bohr radius), so that η in (11) and (13) has the frequency dependence $\sim\Delta^{-4}$ which is more rapid than in the usual nonlinear-optics effects in media with cubic nonlinearity. When light scattering is the dissipation mechanism, the term $\gamma\dot{\mathbf{r}}$ in (6) must then be replaced with the radiation friction force $(2e^2/3c^3)\dddot{\mathbf{r}}$. The quantity γ in (11) is then replaced with

$$\gamma_{\mathrm{rad}}=2e^2\omega^2/3c^3 \qquad (15)$$

and, when $\omega\approx\omega_{\parallel,\perp}\gamma_{\mathrm{rad}}/m$, it is of the order of the natural width of the atomic levels that are being excited. The magnetization $\mathbf{M}$ vanishes in the limit as $\omega\to 0$, $\eta_\gamma,\eta_\lambda\propto\omega^2$ ($M\propto\omega^4$ when the radiation reaction is present). For optical frequencies generated by pulsed solid-state lasers, one would expect that $M\sim 10^{-6}$–10^{-4} erg·G^{-1}·cm^{-3} in resonant monatomic gases or crystals.

B. Optical rectification

In contrast to the magnetization $\bar{\boldsymbol{\mu}}$ that appears even in the dipole approximation (8), the optical rectification effect arises only when the forces $\mathbf{f}_\mathbf{k}$ and $\mathbf{f}_\mathbf{B}$, associated with spatial inhomogeneity and magnetic field of the wave are taken in the first order. According to (7),

$$\mathbf{f}_\mathbf{k}=\frac{ie\omega}{2c}(\mathbf{n}\,\mathbf{r})\mathbf{F}e^{-i\omega t}+\text{c.c.},$$
$$\mathbf{f}_\mathbf{B}=\frac{e}{2c}[\dot{\mathbf{r}}\times[\mathbf{n}\times\mathbf{F}]]e^{-i\omega t}+\text{c.c.},\quad \mathbf{n}=\frac{c}{\omega}\mathbf{k}, \qquad (16)$$

and the solution of (6) is now

$$\mathbf{r}(t)=\mathbf{r}_0(t)+\mathbf{r}_\mathbf{k}(t)+\mathbf{r}_\mathbf{B}(t)+\dots.$$

Assuming that the medium is isotropic ($\omega_\parallel = \omega_\perp\equiv\omega_0$), we find that

$$\bar{\mathbf{r}}_\mathbf{k}=0,\quad \bar{\mathbf{r}}_\mathbf{B}=\frac{\omega}{2ce^2}\alpha(0)\,\mathrm{Im}\,\alpha(\omega)\,(\mathbf{F}\mathbf{F}^*)\mathbf{n},$$

where $\alpha(0)$ is the static polarizability of the oscillator. Thus, the constant part of the dipole moment $\bar{\mathbf{d}} = e\bar{\mathbf{r}}$ induced by the alternating field,

$$\bar{\mathbf{d}}=\frac{1}{e}\alpha(0)\mathbf{f}_d=\gamma\chi_\gamma(\mathbf{F}\mathbf{F}^*)\mathbf{n},\quad \chi_\gamma=\frac{\omega^2}{2ce^3}\alpha(0)|\alpha(\omega)|^2, \qquad (17)$$

is due to the magnetic field of the wave and is produced by the radiation reaction $\mathbf{f}_d$. It is clear that $\mathbf{f}_d$ lies along the wave vector $\mathbf{k}$ and is nonzero only when friction is present ($\gamma\neq 0$), so that the oscillator absorbs light energy.

To avoid misunderstanding, we note that the electric quadrupole interaction $\mathbf{f}_Q$ of the oscillator with the field $\mathbf{F}(\mathbf{r},t)$ in (1) corresponds only to the potential part of $\mathbf{f}_\mathbf{k}$ in (16), whereas the nonconservative part of $\mathbf{f}_\mathbf{k}$ together with $\mathbf{f}_\mathbf{B}$ corresponds to the magnetic dipole interaction $\mathbf{f}_\mu$. Both $\mathbf{f}_Q$ and $\mathbf{f}_\mu$ then provide the same contribution to (17):

$$\chi_\gamma=\chi_\gamma^{(Q)}+\chi_\gamma^{(\mu)},\quad \chi_\gamma^{(Q)}=\chi_\gamma^{(\mu)}=\chi_\gamma/2.$$

In the case of a light pulse with the envelope given by (12), we must take into account the nonadiabatic corrections to $\mathbf{r}_0(t)$ and remember that, because of the spatial inhomogeneity of the light-pulse envelope, there is a change in the force $\mathbf{f}_\mathbf{k}$. Thus, for a traveling wave in a tenuous medium, the envelope of $\mathbf{F}$ is a function of the argument $\mathbf{n}\cdot\mathbf{r} - ct$, so that, in accordance with (12), we have $\partial F_i/\partial\mathbf{r} = -\mathbf{n}(\lambda/c)F_i$ and the expression for $\mathbf{f}_\mathbf{k}$ given by (16) acquires the factor $1 + i\lambda/\omega$. Effects due to nonstationarity (12) and spatial inhomogeneity of the envelope make comparable contributions to the constant dipole moment $\bar{\mathbf{d}}$ induced by the light pulse.

The resulting expression for $\bar{\mathbf{d}}$ can be written as the sum of terms respectively due to magnetic dipole and electric quadrupole interactions:

$$\bar{\mathbf{d}}=\lambda(\chi_\lambda^{(\mu)}+\chi_\lambda^{(Q)})(\mathbf{F}\mathbf{F}^*)\mathbf{n},$$
$$\chi_\lambda^{(\mu,Q)}=\frac{m}{2ce^3}\alpha(0)\alpha^2(\omega)\left[\frac{\omega_0^2+\omega^2}{2}\pm(\omega_0^2-\omega^2)\right], \qquad (18)$$

where, as in the case of (17), the entire effect in (18) is due to the light pressure alone. The absorption of light energy that is responsible for the light pressure is then due to an increase in the oscillation energy with increasing field amplitude F.

When F ($\lambda < 0$) decreases, the oscillator gives up energy to the wave, and $\bar{\mathbf{d}}$ changes sign. In contrast to (17), the moment $\bar{\mathbf{d}}$ in (18) does not vanish in the limit as $\omega \to 0$.

The absence of effects associated with the spatial inhomogeneity of the wave in (17) and (18) is due to the fact that we have used a highly simplified model of the medium. In real systems, the contribution of quadrupole terms is usually the dominant one (see below).

§ 4. QUANTUM-MECHANICAL CALCULATION OF EFFECTS IN THE FIELD OF A LIGHT PULSE

We shall confine our attention to a monatomic gaseous medium when we quantitatively examine the effects defined by (2a) and (4). The state ψ of an atom in the field (1) is a solution of the Schrödinger equation

$$i\hbar(\partial\psi/\partial t) = [H_0 + V(t)]\psi, \tag{19}$$

where the perturbation operator $V(t)$ has the form

$$\begin{aligned} &V(t) = V^{(+)}e^{-i\omega t} + V^{(-)}e^{i\omega t}, \\ &V^{(\pm)} = V_d^{(\pm)} + V_\mu^{(\pm)} + V_Q^{(\pm)} + \ldots, \quad V^{(-)} = V^{(+)+}, \\ &V_d^{(+)} = -\frac{1}{2} e\mathbf{r}\mathbf{F}, \quad V_\mu^{(+)} = -\frac{e}{4mc}[\mathbf{n}\times\mathbf{F}]\mathbf{L}, \\ &V_Q^{(+)} = -\frac{ie\omega}{4c}(\mathbf{F}\mathbf{r})(\mathbf{n}\mathbf{r})\left(1 + i\frac{\lambda}{\omega}\right) \end{aligned} \tag{20}$$

and $\mathbf{r}$ and $\mathbf{L}$ are operators for the position vector and angular momentum of the optical electron in the atom (the factor $1 + i\lambda/\omega$ in the quadrupole interaction operator V_Q takes into account the spatial inhomogeneity of the envelope of the light pulse; see § 3B).

When a nonresonant perturbation $V(t)$ is turned on adiabatically, it takes the atom from the nondegenerate state $|0\rangle$ with energy E_0 to the quasienergy state (QES) $|\psi_0\rangle$ (Ref. 9). The perturbation-theory expansion for $|\psi_0\rangle$ is[10]

$$\begin{aligned} |\psi_0(t)\rangle = \{&1 + G_{E_0+\hbar\omega}V^{(+)}e^{-i\omega t} + G_{E_0-\hbar\omega}V^{(-)}e^{i\omega t} \\ &+ G'_{E_0}[V^{(-)}G_{E_0+\hbar\omega}V^{(+)} + V^{(+)}G_{E_0-\hbar\omega}V^{(-)}] + \ldots\}|0\rangle, \end{aligned} \tag{21}$$

where G_E is the Green function of the optical electron of energy E, and contains summation (and integration) over the intermediate states $|n\rangle$ with energies E_n:

$$G_E = \sum_n \frac{|n\rangle\langle n|}{E - E_n}, \tag{22}$$

where G'_{E_k} denotes the sum (22) for $E = E_k$ with the term containing $|n\rangle = |k\rangle$ omitted. When nonadiabatic corrections are taken into account in the case of a smooth envelope (12), the wave function of the atom can be written in the form

$$\begin{aligned} &|\psi(t)\rangle = |\psi_0(t)\rangle - i\hbar\lambda|\psi^{(1)}(t)\rangle + \ldots, \quad |\lambda| \ll \omega_{n0}, \ |\omega_{n0} - \omega|, \\ &\hbar\omega_{n0} = E_n - E_0. \end{aligned} \tag{23}$$

Substituting this in (19) and recalling that $\partial V^{(\pm)}/\partial t = \lambda V^{(\pm)}$, we find that

$$\begin{aligned} |\psi^{(1)}(t)\rangle = \{&G^2_{E_0+\hbar\omega}V^{(+)}e^{-i\omega t} + G^2_{E_0-\hbar\omega}V^{(-)}e^{i\omega t} \\ &+ 2G'^2_{E_0}[V^{(-)}G_{E_0+\hbar\omega}V^{(+)} \\ &+ V^{(+)}G_{E_0-\hbar\omega}V^{(-)}] \\ &+ G'_{E_0}[V^{(-)}G^2_{E_0+\hbar\omega}V^{(+)} + V^{(+)}G^2_{E_0-\hbar\omega}V^{(-)}] + \ldots\}|0\rangle. \end{aligned} \tag{24}$$

Evaluating the average of the operator $\mathbf{d} = e\mathbf{r}$ in the first order in λ in the state defined by (23), and isolating the nonoscillating part of $\bar{\mathbf{d}}$, we obtain the following expression for the optical rectification effect:

$$\begin{aligned} &\bar{\mathbf{d}} = \lambda\chi_\lambda(\omega)(\mathbf{F}\ \mathbf{F}^*)\mathbf{n}, \\ &\chi_\lambda(\omega) = \chi_\lambda^{(\mu)}(\omega) + \chi_\lambda^{(\mu)}(-\omega) + \chi_\lambda^{(Q)}(\omega) + \chi_\lambda^{(Q)}(-\omega), \\ &\chi_\lambda^{(\mu)}(\omega) = -\frac{i\hbar e^3}{4mc}\langle 0|xG'_{E_0}L_zG^2_{E_0+\hbar\omega}y + 2xG'^2_{E_0}L_zG_{E_0+\hbar\omega}y|0\rangle, \\ &\chi_\lambda^{(Q)}(\omega) = -\frac{e^3}{4c}\langle 0|xG'_{E_0}yG_{E_0+\hbar\omega}xy + xG'_{E_0}xyG_{E_0+\hbar\omega}y \\ &\qquad + xG_{E_0+\hbar\omega}yG_{E_0+\hbar\omega}xy \\ &\qquad + \hbar\omega\{xG'_{E_0}y[xG^2_{E_0+\hbar\omega} - G^2_{E_0+\hbar\omega}x]y \\ &\qquad + 2xG'^2_{E_0}y[xG_{E_0+\hbar\omega} - G_{E_0+\hbar\omega}x]y \\ &\qquad + xyG_{E_0+\hbar\omega}[xG_{E_0+\hbar\omega} - G_{E_0+\hbar\omega}x]G_{E_0+\hbar\omega}y\}|0\rangle. \end{aligned} \tag{25}$$

Using the commutativity of L_z and G_E as well as simple transformations of the spectral sums, we can express the magnetic dipole part of (25) in terms of the polarizability $\alpha(\omega)$ of the ground state:

$$\chi_\lambda^{(\mu)}(\omega) + \chi_\lambda^{(\mu)}(-\omega) = \frac{e}{4mc\omega^2}\frac{d}{d\omega}\{\omega[\alpha(\omega) - \alpha(0)]\}.$$

This result is universal and is therefore valid for both quantum-mechanical and classical systems. In particular, the expression for $\chi_\lambda^{(\mu)}$ given by (18) follows from it.

As an example, Fig. 1 shows the dispersion relation $\chi_\lambda(\omega)$ for the ground state of hydrogen (the calculations make use of the Sturm expansion of the Coulomb Green function[11]). At frequencies $\hbar\omega = E_n - E_0$, the susceptibility χ_λ has second-order poles. As in § 3B, χ_λ remains finite as $\omega \to 0$. In atomic units,

$$\chi_\lambda(\omega \to 0) = \alpha[{}^{53}/_8 + 2.5\cdot 10^2\omega^2 + \ldots], \quad \alpha = {}^1/_{137}.$$

As ω increases, the susceptibility $\chi_\lambda(\omega)$ increases rapidly and the main contribution to χ_λ ($\sim 90\%$) is provided by the quadrupole part of (25). Calculations performed for inert gases in the model-potential approximation[11] yield values of χ_λ that are greater by two or three orders of magnitude as compared with hydrogen (4–6 orders of magnitude in the case of

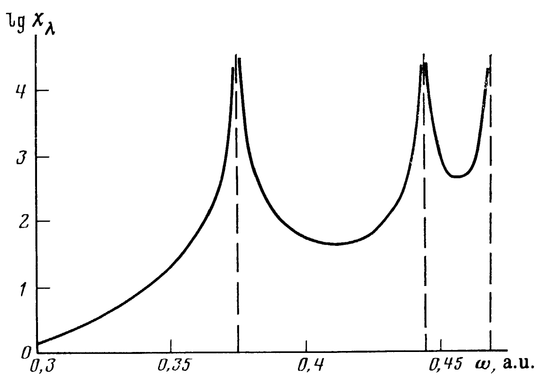

FIG. 1. Dispersion relation for $\chi_\lambda(\omega)$ (in atomic units; 1 a.u. $= 2.09\times 10^{-45}$ cgs). Dashed lines represent resonance frequencies.

the alkali metals). Thus, for frequencies in the optical range, the polarization per unit volume of the gas at atmospheric pressure, induced by a picosecond pulse ($\lambda \sim 10^{11} s^{-1}$) with amplitude $F \sim 5\cdot 10^5$ V/cm, turns out to be about 10^{-6} cgs. We note, for comparison, that a comparable polarization is induced in the gas by a constant field $F_0 \sim 10^2$ V/cm.

Direct calculation of $\chi_\lambda(\omega)$ is impossible for condensed media. Here, we can only relate χ_λ to the usual nonlinear susceptibility χ_{jklm} that determines the polarization of the isotropic medium with spatial dispersion in two fields $\mathbf{F}^{(1)}$ and $\mathbf{F}^{(2)}$ with frequencies ω and ω', propagating in the same direction $\mathbf{n}$. The component of the polarization vector at the difference frequency $\omega - \omega'$ is

$$P_j(t) = (i/2)\chi_{jklm}(\omega'-\omega;\ \omega, -\omega')F_k^{(1)}F_l^{(2)*}n_m e^{i(\omega'-\omega)t} + \text{c.c.} \tag{26}$$

For $\omega = \omega'$, the susceptibility χ_{jklm} of nondissipative media is equal to zero. For $\mathbf{F}^{(1)} = \mathbf{F}^{(2)}$ and comparable ω and ω', the resultant field $\mathbf{F}(t) = \mathbf{F}^{(1)}(t) + \mathbf{F}^{(2)}(t)$ can be written as a wave of frequency $(\omega + \omega')/2$ and envelope $\mathbf{F}$ varying with frequency $\delta = |\omega - \omega'|/2$. Expressing $\mathbf{P}$ in (26) in terms of $\mathbf{F}$ and $\lambda = F^{-1}dF/dt = -\delta \tan \delta t$ in this case, and passing to the limit as $\omega' \to \omega$, we obtain the following expressions for an isotropic medium if we take into account the symmetry properties of χ_{jklm}:

$$\mathbf{P} = \lambda\chi_\lambda(\omega)(\mathbf{F}\mathbf{F}^*)\mathbf{n},\quad \chi_\lambda(\omega) = \frac{\partial}{\partial\omega'}\chi_{xyyx}(\omega'-\omega;\ \omega, -\omega')\big|_{\omega'=\omega}. \tag{27}$$

It is interesting to note that, in this particular problem, the susceptibility χ_{jklm} ($\omega' = \omega$) is zero and the size of the effect is determined by its frequency dispersion in the neighborhood of $\omega = \omega'$. The result (27) was obtained for the special case of the envelope $\mathbf{F}$, but is valid even when $\mathbf{F}$ is an arbitrary function of time t. This is readily verified by expressing the response $\mathbf{P}(t)$, which is quadratic in $\mathbf{F}$, in the form of a double integral of $F_k(t-\tau)F_l(t-\tau')$ with respect to τ and τ', and then transforming it as in the derivation of the energy relations in dispersive anisotropic media.[1,12]

The determination of the magnetization of a gas in a constant electric field $\mathbf{F}_0$ by a pulse of linearly polarized radiation is based on (21), (23), and (24) and is analogous to the determination of $\bar{\mathbf{d}}$. The nonoscillating part of the magnetic moment is

$$\bar{\boldsymbol{\mu}} = \lambda\eta_\lambda(\omega)(\mathbf{F}_0\mathbf{F})[\mathbf{F}_0\times\mathbf{F}]. \tag{28}$$

The explicit expression for $\eta_\lambda(\omega)$ is very unwieldy (it contains several tens of composite matrix elements), and will not be reproduced here. The expression for $\eta_\lambda(\omega)$ becomes much simpler near resonances for which we can confine our attention to terms that are the most rapidly varying functions of frequency. Thus, near resonance with a p-level for $|\Delta| \ll \omega$, $\hbar\Delta = E_0 + \hbar\omega - E_p$ we have

$$\eta_\lambda(\omega) = \frac{e^3}{6\hbar m^2 c\omega\Delta^4} f_{p0}\alpha_p^{\mathrm{T}}, \tag{29}$$

where f_{p0} is the oscillator strength of the $|0\rangle \to |p\rangle$ transition and α_p^{T} is the tensor part of the static polarizability of the p-level that determines the splitting δ of the magnetic sublevels in a constant electric field $\mathbf{F}_0$: $\delta = E_{p,m=0} - E_{p,m=1} = \frac{3}{4}\alpha_p^{\mathrm{T}}F_0^2$. We note that, for the three-dimensional oscillator $f_{p0} = 1$ and $\delta/\hbar$ is the frequency difference $\omega_\parallel - \omega_\perp$ introduced in § 3, so that (29) becomes identical with the classical result (14) in the neighborhood of resonance.

In monatomic gases, the magnetization becomes appreciable only for frequencies ω approaching the resonance value. The characteristic values for atoms are $f_{p,0} \sim 1$, $\alpha_p^{\mathrm{T}} \sim 10^3$ a.u., so that the magnetization of the gas for $\Delta = 10$ cm^{-1}, $\lambda = 10^{11}$ s^{-1}, $F = 10^5$ V/cm, $F_0 = 10^4$ V/cm amounts to about 10^{-5} erg·G^{-1}·cm^{-3}, i.e., it is comparable with the magnetization of a diamagnetic medium in a constant magnetic field $B \sim 1000$ G.

By analogy with (26) and (27), the magnetization of a medium by a pulsed light field can also be looked upon as the limiting case of magnetization in the field of two frequencies ω and ω':

$$M_j(t) = (i/2)\eta_{jkl}(\omega'-\omega;\ \omega, -\omega')F_k^{(1)}F_l^{(2)*}e^{i(\omega'-\omega)t} + \text{c.c.} \tag{30}$$

When $\omega = \omega'$, the tensor η_{jkl} is antisymmetric in the indices k, l in the case of nondissipative medium and determines the inverse Faraday effect. $M_j(\omega = \omega')$ vanishes only when $\mathbf{F}^{(1)}$ and $\mathbf{F}^{(2)}$ are linearly polarized in the same direction. The transformation of (30) that is analogous to the transition from (26) to (27) then yields[1)]

$$M_j = \lambda F_k F_l \frac{\partial}{\partial\omega'}\eta_{jkl}(\omega'-\omega;\ \omega, -\omega')\big|_{\omega'=\omega}. \tag{31}$$

In particular, allowance for the spatial symmetry of η_{jkl} in the case of a uniaxial crystal enables us to rewrite (31) in the form

$$\mathbf{M} = \lambda\eta_\lambda(\omega)(\mathbf{v}\mathbf{F})[\mathbf{v}\times\mathbf{F}],$$

where

$$\eta_\lambda(\omega) = \frac{\partial}{\partial\omega'}\{\eta_{yzx}(\omega'-\omega;\ \omega, -\omega') + \eta_{yxz}(\omega'-\omega;\ \omega, -\omega')\}\big|_{\omega=\omega'}, \tag{32}$$

and we assume that the z axis is the crystal anisotropy axis.

§ 5. POLARIZATION AND MAGNETIZATION OF A GASEOUS MEDIUM BY A NONRESONANT MONOCHROMATIC LIGHT WAVE

In a nonresonant field in which the detuning from resonance $\hbar\Delta = |E_n - E_0 - \hbar\omega|$ exceeds the thermal energy kT of the atom, collisions between atoms do not affect the interaction between radiation and medium, and spontaneous scattering of light by atoms[13] is the principal mechanism for the dissipation of light energy. When spontaneous scattering is taken into account, the state of the atom in the field is no longer described by pure QES (21) because of the presence of effects due to radiation reaction. The simplest way of taking into account radiative damping is to introduce the width Γ_n of the excited levels, i.e., by substituting $E_n \to E_n - i\hbar\Gamma_n/2$, but this is valid only for resonance scattering of a low-intensity light wave. In the general case, on the other hand, the density matrix formalism has to be used to take into account the effect of the thermostat (in our problem, the electromagnetic vacuum) on the state of the system.

Under the influence of the light field, the system eventually reaches a steady state described by the density matrix $\rho(t) = \rho(t + 2\pi/\omega)$, which is periodic in time. It can be written as the sum of "even" and "odd" parts:

$$\rho(t) = \rho_{\rm even}(t) + \rho_{\rm odd}(t), \tag{33}$$

where $\rho_{\rm even}$ describes the "forced oscillations" of the system (with frequencies that are multiples of ω) and $\rho_{\rm odd}$ represents the effect of "friction" (dissipative processes) on these oscillations. It is precisely $\rho_{\rm odd}$ that determines the effects that we are considering.

The usual Bloch-type relaxation equations are unsuitable for the determination of $\rho(t)$ in a nonresonant field and we must resort to the more general integro-differential equations.[13] However, the determination of $\rho_{\rm odd}$ is then simpler: in a nonresonant field $\rho_{\rm odd}/\rho_{\rm even} \sim \Gamma/\Delta \ll 1$ (where Γ and Δ are the characteristic level width and detuning from resonance, respectively) and we can develop a perturbation theory in Γ. If, in addition, the interaction V with the light wave is also looked upon as a perturbation, simple closed expressions can be obtained for the first few terms of the expansion for $\rho_{\rm odd}$ in powers of V. In particular, when dissipation is due to the dipole scattering of light, we have in the first order in Γ

$$\rho_{\rm odd} = -i \sum_n \{\gamma_n \Omega_n [G_{E_0+\hbar\omega} \mathbf{r} \mathbf{R}_{n0} + \mathbf{R}_{n0} \mathbf{r} G_{E_n-\hbar\omega}] e^{-i\omega t}$$

$$+\gamma_n \Omega_n [G_{E_0}' \mathbf{r} G_{E_n-\hbar\omega} V^{(-)} \mathbf{R}_{n0} + G_{E_0}' V^{(-)} G_{E_0+\hbar\omega} \mathbf{r} \mathbf{R}_{n0}$$

$$+G_{E_0+\hbar\omega} \mathbf{r} \mathbf{R}_{n0} V^{(-)} G_{E_0+\hbar\omega} + G_{E_n-\hbar\omega} V^{(-)} \mathbf{R}_{n0} \mathbf{r} G_{E_n-\hbar\omega}$$
$$+\mathbf{R}_{n0} \mathbf{r} G_{E_n-\hbar\omega} V^{(-)} G_{E_n}' + \mathbf{R}_{n0} V^{(-)} G_{E_0+\hbar\omega} \mathbf{r} G_{E_n}'] + \ldots\} + \text{h.c.},$$

where $\gamma_n = 2e^2\Omega_n^2/3c^3$ and the vector operator is

$$\mathbf{R}_{n0} \equiv |n\rangle\langle n| \mathbf{r} G_{E_0+\hbar\omega} V^{(+)} + V^{(+)} G_{E_n-\hbar\omega} \mathbf{r} |0\rangle\langle 0|.$$

The various terms in the sum over n represent the effect of friction due to the transition from the ground state $|0\rangle$ to $|n\rangle$ with the absorption of a photon $\hbar\omega$ and the emission of a spontaneous Raman photon $\hbar\Omega_n = E_0 + \hbar\omega - E_n$. The sum over n is finite because it includes only components with $\Omega_n > 0$. In particular, when the external field frequency ω is less than the distance to the first resonance, the sum contains only the $n = 0$ term that corresponds to elastic scattering of light ($\Omega_0 = \omega$).

When the average electric or magnetic moments in the state (33) are evaluated, $\rho_{\rm even}$ does not contribute to the effects. The time-independent component of the moment is determined exclusively by the constant terms in the expression for $\rho_{\rm odd}$:

$$\bar{\mathbf{d}} = \mathrm{Sp}(e\mathbf{r}\rho_{\rm odd}^{(\rm const)}), \quad \boldsymbol{\mu} = \mathrm{Sp}\left(\frac{e\mathbf{L}}{2mc}\rho_{\rm odd}^{(\rm const)}\right). \tag{34}$$

The final expression can be written in the form

$$\bar{\mathbf{d}} = \frac{e^5}{6c^4} \chi_\gamma(\omega) (\mathbf{F}\mathbf{F}^*) \mathbf{n},$$

$$\chi_\gamma(\omega) = \sum_n \Omega_n^3 (\chi_{1,n}^{(Q)} + \chi_{2,n}^{(Q)} + \chi_{1,n}^{(\mu)} + \chi_{2,n}^{(\mu)}). \tag{35}$$

Each of the terms in χ_γ is the product of two matrix elements:

$$\chi_{1,n}^{(Q)} = -\omega \mathbf{M}_{1,n} \mathbf{M}_n^{(d)}, \quad \chi_{2,n}^{(Q)} = \omega \mathbf{M}_{2,n} \mathbf{M}_n^{(Q)},$$

$$\chi_{1,n}^{(\mu)} = -\frac{i}{m} \mathbf{M}_{3,n} \mathbf{M}_n^{(d)}, \quad \chi_{2,n}^{(\mu)} = -\frac{i}{m} \mathbf{M}_{2,n} \mathbf{M}_n^{(\mu)},$$

where $\mathbf{M}_n^{(d)}$, $\mathbf{M}_n^{(Q)}$ and $\mathbf{M}_n^{(\mu)}$ are the components of the dipole-dipole, dipole-quadrupole, and dipole-magnetic-dipole light-scattering tensors:

$$\mathbf{M}_n^{(d)} = \langle n | y G_{E_n-\hbar\omega} \mathbf{r} + \mathbf{r} G_{E_0+\hbar\omega} y | 0\rangle,$$

$$\mathbf{M}_n^{(Q)} = \langle n | xy G_{E_n-\hbar\omega} \mathbf{r} + \mathbf{r} G_{E_0+\hbar\omega} xy | 0\rangle,$$

$$\mathbf{M}_n^{(\mu)} = \langle n | L_z G_{E_n-\hbar\omega} \mathbf{r} + \mathbf{r} G_{E_0+\hbar\omega} L_z | 0\rangle,$$

and the matrix elements $\mathbf{M}_{i,n}$ ($i = 1,2,3$) are given by

$$\mathbf{M}_{i,n} = \langle 0 | y G_{E_0}' V_i G_{E_0+\hbar\omega} \mathbf{r} + y G_{E_0}' \mathbf{r} G_{E_n-\hbar\omega} V_i + \mathbf{r} G_{E_n-\hbar\omega} y G_{E_n-\hbar\omega} V_i$$
$$+ \mathbf{r} G_{E_n-\hbar\omega} V_i G_{E_n}' y + V_i G_{E_0+\hbar\omega} y G_{E_0+\hbar\omega} \mathbf{r} + V_i G_{E_0+\hbar\omega} \mathbf{r} G_{E_n}' y | n\rangle,$$

where $V_1 = xy$, $V_2 = y$, $V_3 = L_z$.

The magnetic-dipole part of (35) with $n = 0$ (elastic scattering) is expressed as in Section 4 in terms of the polarizability of the ground state $|0\rangle$:

$$\chi_{1,0}^{(\mu)} + \chi_{2,0}^{(\mu)} = \frac{\omega^2}{me^4} \alpha(\omega) [\alpha(\omega) - \alpha(0)],$$

which is identical in the case of the oscillator with the result of the classical analysis in § 3B.

Figure 2 shows the frequency dependence $\chi_\gamma(\omega)$ for the hydrogen atom. As $\omega \to 0$, the susceptibility becomes $\chi_\gamma \approx 360\omega^4$ a.u., i.e., it falls as in (17) with $\gamma = \gamma_{\rm rad}$ in (15). The contribution of magnetic dipole terms χ_γ at low frequencies is about 30%, and falls rapidly with increasing ω. Near resonance with the first excited level, $\chi \sim 1/\Delta^2$ as in (17). The appearance of third-order poles at resonances with higher-lying levels is a consequence of Coulomb degeneracy. The $n \neq 0$ terms in (35) provide no more than 10% of χ_γ, in practically the entire frequency range because $\Omega_n^3 \ll \omega^3$. Hence, χ_γ is determined largely by dissipation through elastic scattering. For atoms other than the hydrogen atom, the magnitude of χ_γ is greater than in Fig. 2 by two or three orders of magnitude. The polarization per unit volume of the

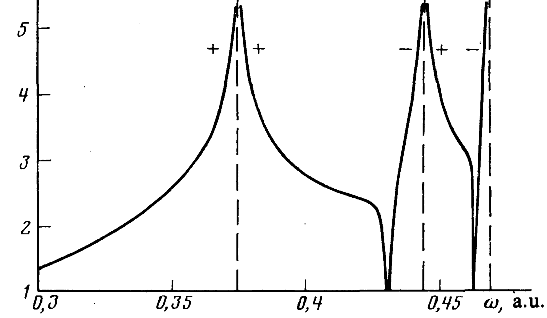

FIG. 2. Dispersion relation for $\chi_\gamma(\omega)$ (1 a.u. = 7.75×10^{47} cgs). The signs of $\chi_\gamma(\omega)$ at different frequencies are indicated against the curve.

gaseous medium in a field of about 10^5 V/cm is about 10^{-6} cgs.

According to (34), the magnetization of the atom in the linear field is

$$\bar{\mu}=\frac{e^5}{6mc^4}\eta_\gamma(\omega)(\mathbf{F}_0\mathbf{F})[\mathbf{F}_0\times\mathbf{F}].$$

The expression for η_γ will now be given but only for the case where ω is close to resonance with the p-level and provided we confine our attention to terms corresponding to elastic light scattering (it is precisely these terms that provide the dominant contribution to the effect):

$$\eta_\gamma(\omega)=\frac{3}{16}\frac{\omega}{\hbar m^2\Delta^4}f_{p0}{}^2\alpha_p{}^T,$$

where Δ, f_{p0} and α_p^T are the same as in (29). This result is in agreement with (11), so that estimates of **M** based on the classical model remain valid for resonant monatomic gases as well.

§ 6. CONCLUSION

In the microscopic determination of the effects defined by (2a) and (4) in Section 5, we confined our attention to the simplest (though universal) dissipation mechanism, i.e., spontaneous light scattering. The presence of a T-odd parameter in the problem may, of course, be due to other nonequilibrium processes as well. For example, in a resonant gaseous medium in which elastic collisions are important, the T-odd parameter will be determined by the transverse level widths, i.e., dissipation of the "coherence of atomic states." At the same time, inhomogeneous widths (Doppler, etc.) are not related to irreversibility and can themselves give rise to the above effects.

In precisely the same way, nonstationary processes are not exhausted by the case of the light pulse examined in § 4. In particular, it is very interesting to consider the effects defined by (2a) and (4) during the development of threshold processes, for example, when stimulated Raman scattering or the generation of harmonics (or simply the generation of laser radiation) occurs in the medium.

The manifestations of the new effects may be different in different cases. For example, when a particular amount of resonance radiation (analog of the π pulse) is transmitted by an anisotropic gaseous medium, the medium remains magnetized after the passage of the pulse, and the magnetization **M** subsequently oscillates at frequency $|\omega_\parallel-\omega_\perp|$ and relaxes during the lifetime of the excited resonance state.

All these problems are, however, connected with resonance processes. Their analysis in each individual case requires specialized techniques that lie outside the range of the present paper.

The above effects will provide us with new information on the nonlinear properties of different media and can be used to investigate different dissipative and transient processes. It may be more convenient to carry out experiments with intensity-modulated radiation and observe the electric or magnetic polarization oscillating at the modulation frequency.

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[1)] Similar corrections to the Pitaevskiĭ formula[3] for the magnetization of a transparent medium by an alternating field were obtained in Ref. 2 as a result of a macroscopic analysis of effects due to a nonmonochromatic wave in a medium with frequency dispersion. However, in contrast to (31), the final result [Eq. (2.22) in Ref. 2] does not agree with the microscopic calculation of $\eta_\gamma(\omega)$ for a monatomic gas.

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