Nonlinear theory of radiation dragging

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The nonlinear problem of passage of electromagnetic radiation through a resonant gaseous medium is considered. A system of equations is obtained for the medium and for the field, with the dragging taken into account up to the terms of fourth order in the field. The Maxwell equation with cubic nonlinearity has an essentially nonlocal structure and depends on the type of the pumping wave (standing or traveling). In the order quadratic in intensity, the scattered field contains photons with frequencies equal to that of the pumping field as well as shifted in frequency by $\pm\Delta$. The appearance of photons with greatly differing mean free paths alters substantially the character of the diffusion of the excitations in the medium. The nonlinear effects lead to interaction of two light beams. Two traveling waves then attract and repel each other, depending on the sign of the detuning Δ. Interaction of the traveling wave with the scattered field of the standing wave leads to a phenomenon of the wave-front-reversal type.

1. INTRODUCTION

Effects connected with radiation dragging play an important role in the spectroscopy of gases. Even in a low-density gas ($n\lambda^3 \ll 1$, where λ is the resonant wavelength) it is easy to realize conditions such that the dimension of the medium exceeds the photon mean free path.

The existing theory of radiation dragging is based on the assumption that the external and scattered fields are weak, so that the propagation of the excitation in the resonant gas is linear in the field intensity and is described by an equation of Holstein-Biberman type.[1–5]

In the absence of collisions, the frequencies of the scattered photons are changed only by the Doppler effect. We can thus distinguish in the radiation-dragging problem two limiting cases. If the atom motion is negligible, the frequency of the diffusing radiation does not differ from that of the pump wave.[4,5] In the case of strongly inhomogeneous broadening the correlation between the frequencies of the incident and scattered fields becomes, on the contrary, very weak and is preserved only accurate to the Doppler width.[3] These limiting cases can be realized, for example, by varying the detuning $\Delta = \omega_0 - \omega_{at}$ of the external-field frequency ω_0 from zero, so that at $kv_0 \ll \Delta$ (v_0 is the characteristic thermal velocity) the atoms can be regarded as immobile, whereas at $kv_0 \gg \Delta$ we get strong Doppler broadening.

This paper is devoted to nonlinear theory of radiation dragging. When an electromagnetic wave propagates through a resonant medium under conditions of noticeable saturation and absorption, the scattered field acquires a high intensity and must be taken into account together with the mean field.

Nonlinear effects lead to a mutual influence of the mean and scattered fields and alter the scattered-radiation spectrum via the field-induced frequency splitting. Nonlinear equations for the medium and the field were obtained earlier[6] under conditions of strong inhomogeneous broadening, when the field splitting was less than kv_0 and the frequency correlation in the radiation diffusion could be neglected. In the present paper we consider the case of immobile atoms ($kv_0 \ll \Delta$) when the scattered-radiation spectrum is altered only by field effects and the frequency correlation in the diffusion is most strongly manifested.

The Maxwell equation for the mean field is derived (Sec. 3) in an approximation cubic in the field. Allowance for the radiation dragging in the pump-wave region makes this equation spatially nonlocal. The distribution function of the excited atoms is obtained accurate to terms of fourth order in the field (Sec. 4). In this approximation, the shifted components of the atom resonance-fluorescence spectrum alter the radiation-diffusion rate, since the mean free paths of photons with shifted and unshifted frequencies can differ greatly.

We show that the form of the nonlinear Maxwell equation depends strongly on the pump-wave spatial structure. In particular, if the medium is excited by a standing wave the resultant scattered field can lead to effects of the wave-front-reversal type. As an example of the manifestation of nonlinear nonlocal effects, we consider the interaction of two light beams, which can either attract or repel each other, depending on the sign of the resonance detuning.

2. INITIAL EQUATIONS

The Hamiltonian of a system of two-level atoms that interact with a radiation field can be expressed in the resonance approximation in the form ($\hbar = c = 1$)

$$\sum_{\mathbf{k}} (\omega_{\mathbf{k}}-\omega_0) c_{\mathbf{k}}^+ c_{\mathbf{k}} - \frac{\Delta}{2} \sum_j \hat{\sigma}_j^3 + \sum_j \left[\hat{\sigma}_j^+ (V_{0j}+\hat{V}_j) + \text{H.a.}\right],$$

$$V_{0j}=V_0(\mathbf{r}_j), \quad V_0(\mathbf{r}) = -\mathbf{d}\mathbf{E}_0(\mathbf{r}), \tag{1}$$

$$\hat{V}_j=\hat{V}(\mathbf{r}_j), \quad \hat{V}(\mathbf{r}) = g\sum_{\mathbf{k}} c_{\mathbf{k}} e^{i\mathbf{k}\mathbf{r}}, \quad g=d(2\pi\omega_0)^{1/2}.$$

Here d is the dipole matrix element, the frequency of the external classical field $E_0(\mathbf{r})e^{-i\omega_0 t}$ + c.c. differs little ($\Delta = \omega_0 - \omega_{at}$) from that of the atomic transition, the operators of the j-th atom are described by Pauli matrices $\hat{\sigma}$, $\hat{\sigma}^+$, and $\hat{\sigma}^3$, and $\hat{V}(\mathbf{r})$ is the operator of the quantized electromagnetic field. The atoms are immobile, are randomly dis-

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tributed in space, and have a density n satisfying the condition $n\lambda^3 \ll 1$. We consider for simplicity a scalar model of the interaction and disregard the polarization properties of the medium (level degeneracy) and of the radiation.

The Heisenberg equations of motion for the medium and field operators are

$$\left(i\frac{d}{dt}+\Delta\right)\hat{\sigma}_j(t)=-\hat{\sigma}_j^3(t)\,(V_{0j}+\hat{V}_j(t)), \tag{2}$$

$$i\frac{d}{dt}\hat{\sigma}_j^3(t)=2\hat{\sigma}_j(t)\,(V_{0j}+\hat{V}_j(t))-\text{H.a.}, \tag{3}$$

$$i\frac{d}{dt}c_{\mathbf{k}}(t)=(\omega_{\mathbf{k}}-\omega_0)\,c_{\mathbf{k}}(t)+g\sum_j\exp(-i\mathbf{k}\mathbf{r}_j)\,\hat{\sigma}_j(t). \tag{4}$$

Solving (4) and neglecting the relativistic retardation over the size of the atom system, we get

$$\hat{V}_j(t)=\hat{V}_{0j}(t)+\sum_{j'}\overset{0}{D}_{jj'}\hat{\sigma}_{j'}(t). \tag{5}$$

The first term of this equation

$$\hat{V}_{0j}(t)=g\sum_{\mathbf{k}}c_{\mathbf{k}}\exp[i\mathbf{k}\mathbf{r}_j-i(\omega_{\mathbf{k}}-\omega_0)t]$$

is the field zero-point oscillation operator, and the second is the scattered-field operator expressed in terms of the photon Green's function:

$$\overset{0}{D}_{jj'}\equiv D^0(\mathbf{r}_j-\mathbf{r}_{j'}),\quad D^0(\mathbf{r})=-\frac{\gamma}{2}\begin{cases}\dfrac{e^{ik_0r}}{k_0r}, & r\neq 0\\ i, & r=0\end{cases}. \tag{6}$$

$$\gamma=2k_0^3d^2,\quad k_0=\omega_0.$$

The value of this function at coinciding points ($r=0$) takes into account the effect of the radiation field on the emitting atom itself, an effect that leads to damping of the upper atomic state with a relaxation constant. The lower is taken to be the ground state.

We represent the operators $\hat{\sigma}_j(t)$ and $\hat{\sigma}_j^3(t)$ as sums of mean values and fluctuating parts:

$$\hat{\sigma}_j(t)=\sigma_j+\hat{f}_j(t),\quad \hat{\sigma}_j^3(t)=\sigma_j^3+\hat{f}_j^3(t),$$
$$\sigma_j=\langle\hat{\sigma}_j(t)\rangle_0,\ \sigma_j^3=\langle\hat{\sigma}_j^3(t)\rangle_0,\ \langle\hat{f}_j(t)\rangle_0=\langle\hat{f}_j^3(t)\rangle_0=0. \tag{7}$$

The averaging $\langle\ldots\rangle_0$ is over the initial state of the "atom + field" system, where there are no scattered photons and the atoms are in the ground state.

Substituting (5) in (2) and (3) and separating the relaxation terms, we obtain in the quasistationary approximation ($d/dt \ll \gamma$) the following equations for σ_j and σ_j^3:

$$\nu\sigma_j=-\sigma_j^3V_j-\sum_{j'\neq j}D_{jj'}\langle\hat{f}_j^3(t)\hat{f}_{j'}(t)\rangle_0,\quad \nu=\Delta+i\gamma/2,$$
$$i\gamma(1+\sigma_j^3)=2\sigma_j^*V_j+2\sum_{j'\neq j}\overset{0}{D}_{jj'}\langle\hat{f}_j^+(t)\hat{f}_{j'}(t)\rangle_0-\text{c.c.}; \tag{8}$$

$$V_j=V_{0j}+\sum_{j'\neq j}\overset{0}{D}_{jj'}\sigma_{j'}. \tag{9}$$

These equations describe the interaction between atoms located at points $\mathbf{r}_j$ and a monochormatic field V_j that is the sum of the incident and scattered fields, and with the nonmonochromatic field due to the fluctuations of the atomic operators.

The fluctuating parts of the atomic operators are determined by the zero-point oscillations of the magnetic field and give the shifted components of the resonance-fluorescence spectrum. These components are known[7] to appear in fourth order in the external field and are quadratic in the amplitude of the zero-point oscillations.

We confine ourselves hereafter to nonlinear effects of order V^3 and V^4. They can be adequately described by the equations for $\hat{f}_j(t)$ and $\hat{f}_j^3(t)$ in the linear approximation in $\hat{V}_0$. Taking Fourier transforms with respect to time, we obtain (the frequency ω is reckoned from the external-field frequency ω_0)

$$(\omega+\nu)\hat{f}_j(\omega)=-\hat{f}_j^3(\omega)V_j-\sigma_j^3\Big[\hat{V}_{0j}(\omega)+\sum_{j'\neq j}\overset{0}{D}_{jj'}\hat{f}_{j'}(\omega)\Big],$$

$$(\omega+i\gamma)\hat{f}_j^3(\omega)$$
$$=2V_j\hat{f}_j^+(-\omega)-2V_j^*\hat{f}_j(\omega)+2\sigma_j^*\Big[\hat{V}_{0j}(\omega)+\sum_{j'\neq j}\overset{0}{D}_{jj'}\hat{f}_{j'}(\omega)\Big]$$
$$-2\sigma_j\Big[\hat{V}_{0j}^+(-\omega)+\sum_{j'\neq j}\overset{*0}{D}_{jj'}\hat{f}_{j'}^+(-\omega)\Big]. \tag{10}$$

Equations (8), (9), and (10) are a closed system for the medium and the field, with the radiation reabsorption taken into account.

3. EQUATION FOR MEAN FIELD IN THE CUBIC APPROXIMATION

To obtain the macroscopic Maxwell equation we must average (9) over the locations of the atoms in space. This can be done by using the impurity diagram technique.[8]

In the approximation linear in the field (which we designate by $\mathscr{V}_j$) we have $\sigma_j^3=-1$ and $\sigma_j=\mathscr{V}_j/\nu$, and Eq. (9) takes the form

$$\mathscr{V}_j=V_{0j}+\frac{1}{\nu}\sum_{j'\neq j}\overset{0}{D}_{jj'}\mathscr{V}_{j'}. \tag{11}$$

Its solution

$$\mathscr{V}_j=V_{0j}+\frac{1}{\nu}\sum_{j'}D_{jj'}V_{0j'} \tag{12}$$

is expressed in terms of the photon Green's function in the medium at zero frequency, $D_{jj'}=D_{jj'}(\omega=0)$, where $D_{jj'}(\omega)$ satisfies the equation

$$D_{jj'}(\omega)=D_{jj'}^{(0)}+\frac{1}{\omega+\nu}\sum_{j_1\neq j}\overset{0}{D}_{jj_1}D_{j_1j'}(\omega). \tag{13}$$

Averaging (13) over the disposition of the atoms (we denote this operation by angle brackets without a subscript), we obtain the macroscopic Green's function of the photon

$$D(\mathbf{r}\omega)=\langle D_{jj'}(\omega)\rangle=-\frac{\gamma}{2}\frac{e^{ik(\omega)r}}{k(\omega)r},\quad \mathbf{r}=\mathbf{r}_j-\mathbf{r}_{j'},$$
$$k(\omega)=k_0[1+2\pi\chi(\omega)], \tag{14}$$

where $\chi(\omega)=-nd^2/(\omega+\nu)$ is the linear susceptibility of the gas at the frequency ω.

Let $u(\mathbf{r})$ be the average (macroscopic field: $u(\mathbf{r}) = \langle V_j \rangle \approx \langle \mathcal{V}_j \rangle$. Averaging of (12) leads, in the leading order in the parameter $n\lambda^3$, to the linear Maxwell equation

$$[\nabla^2+\omega_0^2(1+4\pi\chi_0)]\,u(\mathbf{r})=0, \tag{15}$$

where $\chi_0 = \chi(\omega = 0)$.

In the order quadratic in the field the atom-excitation probability is no longer zero

$$(1+\sigma_j^3)/2 \approx |\mathcal{V}_j|^2/|\nu|^2 \equiv w_j, \tag{16}$$

and yields the spatial distribution of the energy stored in the resonant medium.

When averaging of $|\mathcal{V}_j|^2$ over the disposition of the atoms

$$\langle |\mathcal{V}_j|^2 \rangle = \langle \mathcal{V}_j \mathcal{V}_j^* \rangle = |u|^2 + \langle\langle \mathcal{V}_j \mathcal{V}_j^* \rangle\rangle$$

it suffices to use for the irreducible part $\langle\langle \dots \rangle\rangle$ the "ladder" approximation in which only terms proportional to $|D_{jj'}|^2$ are retained, since the remaining terms oscillate and are small in terms of the parameter $n\lambda^3$. As a result we obtain for the macroscopic distribution function $w(\mathbf{r}) = \langle w_j \rangle$ of the excitations the integral equation

$$w(\mathbf{r}) = w_c(\mathbf{r}) + \int d^3\mathbf{r}'\, Q_0(\mathbf{r}-\mathbf{r}')\, w(\mathbf{r}'),$$
$$w_c(\mathbf{r}) = \frac{|u(\mathbf{r})|^2}{|\nu|^2}, \tag{17}$$
$$Q_0(\mathbf{r}) = \frac{n}{|\nu|^2}|D(\mathbf{r},\omega=0)|^2 = \frac{\varkappa_0}{2\pi r^2}e^{-2\varkappa_0 r},$$

where $\varkappa_0 = 2\pi k_0 \,\mathrm{Im}\,\chi_0$ is the linear absorption coefficient at the frequency $\omega = 0$. The points $\mathbf{r}$ and $\mathbf{r}'$ belong to the region occupied by the atom. The atom density is assumed constant, so that $nw(\mathbf{r})$ is the probability of exciting the atom by a coherent mean field. The integral term in (17), which we designate by $w_s(\mathbf{r})$ (so that $w = w_c + w_s$), is due to the action of the scattered incoherent field on the atom. We note that for immobile atoms the excitations are transported in the medium, in the approximation quadratic in the field, by photons whose frequency is equal to that of the external field. An equation of this type was obtained in Refs. 4 and 5.

The solution of (17) can be written in the symbolic form

$$w = w_c + w_s, \quad w_s = K_0 w_c, \tag{18}$$

where the integral operator K_0 with kernal $K_0(\mathbf{r}_1, \mathbf{r}_2)$ satisfies the equation

$$K_0 = Q_0 + Q_0 K_0. \tag{19}$$

The function $K_0(\mathbf{r}_1, \mathbf{r}_2)$ determines the excitation of an atom at a point $\mathbf{r}_1$ by a scattered field generated by coherent pumping at a point $\mathbf{r}_2$. For a system of finite size it does not reduce, generally speaking, to a function of the coordinate difference. We refer to K_0 hereafter as the excitation propagation function at the frequency $\omega = 0$.

We proceed to now derive the Maxwell equation in the approximation cubic in the field. We estimate the contribution of the fluctuation operators $\hat{f}_j$ and $\hat{f}_j^3$, which enter in Eq. (8) for the mean values. To this end we must calculate $\hat{f}_j^3$ and $\hat{f}_j$ in the approximations linear and quadratic in V_j, respectively. We obtain from (10)

$$\hat{f}_j^3(\omega) = \frac{2}{\nu^*} V_j^* \sum_{j_1} G_{jj_1}(\omega)\hat{\mathcal{V}}_{0j_1}(\omega) + \dots,$$
$$\hat{f}_j(\omega) = -\frac{2}{\nu}\sum_{j_1 j_2} G_{jj_1}(\omega)\, G^*_{j_1 j_2}(-\omega)\, V_{j_1}^2 \hat{\mathcal{V}}^*_{0j_2}(-\omega) + \dots, \tag{20}$$
$$G_{jj'}(\omega) = \delta_{jj'}/(\omega+\nu) + D_{jj'}(\omega)/(\omega+\nu)^2.$$

We have written out here only the terms that contribute to the mean values $\langle \dots \rangle_0$ contained in Eq. (8).

Accurate to the oscillating terms, the contribution of the fluctuations to σ_j is determined by the quantity

$$\sum_{j'\neq j} D_{jj'}{}^0 \langle \hat{f}_j^3(t) \hat{f}_{j'}(t) \rangle_0 = \frac{2i}{\pi|\nu|^2} V_j^* \sum_{j'} V_{j'}^2 \int d\omega \frac{D_{jj'}(\omega) D^*_{jj'}(-\omega)}{(\omega+\nu)(\omega-\nu^*)^2} = 0,$$

since all the poles of the integrand are in the same half-plane. In the approximation cubic in the field the fluctuations therefore do not contribute to (9), which takes then the form

$$V_j = V_{0j} + \frac{1}{\nu}\sum_{j'\neq j} D_{jj'}^0 V_{j'}\left(1 - \frac{2|V_{j'}|^2}{|\nu|^2}\right). \tag{21}$$

The solution of (21) can be written with the aid of the linear approximation (12) in the form

$$V_j = \mathcal{V}_j - \frac{2}{\nu|\nu|^2}\sum_{j'} D_{jj'}\mathcal{V}_{j'}|\mathcal{V}_{j'}|^2, \tag{22}$$

which facilitates the averaging. Averaging of the nonlinear term in (22) leads to terms of three types:

$$\langle D_{jj'}\mathcal{V}_{j'}|\mathcal{V}_{j'}|^2 \rangle = D(\mathbf{r}-\mathbf{r}')\,u(\mathbf{r}')\,\{|u(\mathbf{r}')|^2 + 2\langle\langle |\mathcal{V}_{j'}|^2 \rangle\rangle\} + u^2(\mathbf{r}')\langle\langle D_{jj'}\mathcal{V}_{j'}^* \rangle\rangle. \tag{23}$$

In the approximation assumed here we can put $\langle \mathcal{V}_j \rangle \approx u(\mathbf{r})$ and $\langle\langle |\mathcal{V}_j|^2 \rangle\rangle \sim w_s(\mathbf{r})$, with the latter quantity entering in (23) with double the weight. The cause of this statistical factor is that in averaging of a product of three fields $\mathcal{V}_j |\mathcal{V}_j|^2$ the quantity appears in twice as many pairings for the scattered field than in the calculation of $\langle |\mathcal{V}_j|^2 \rangle$. The last term of (23) has an anomalous form since it contains not the intensity of the mean field but the square of its amplitude.

The result is the following Maxwell equation with cubic nonlinearity:

$$\{\nabla^2 + \omega_0^2 + 4\pi\omega_0^2\chi_0[1 - 2(w_c(\mathbf{r}) + 2w_s(\mathbf{r}))]\}\,u(\mathbf{r}) - \frac{8\omega_0^2\chi_0^*}{|\nu|^2}u^*(\mathbf{r})\int d^3\mathbf{r}'\, K_0(\mathbf{r},\mathbf{r}')\, u^2(\mathbf{r}') = 0. \tag{24}$$

The last term of this equation makes a finite contribution only for a mean field of the standing-wave type. In the case of a traveling wave, $u^2(\mathbf{r})$ oscillates and this term can be omitted. The Maxwell equation takes then the simpler form

$$\{\nabla^2 + \omega_0^2 + 4\pi\omega_0^2\chi_0[1 - 2(w_c(\mathbf{r}) + 2w_s(\mathbf{r}))]\}\,u(\mathbf{r}) = 0. \tag{25}$$

In this equation, the part nonlinear in the field describes the saturation due to both the coherent pump field (w_c) and to the scattered field (w_s). These contributions do not add up to the total excitation probability w, which is determined by the Holstein-Biberman equation (17).

If the light beam has a large enough transverse dimension $\varkappa l \gtrsim 1$, the nonlinear susceptibility of the medium becomes a nonlocal quantity. The nonlocal interaction can therefore be significant in propagation of intense light waves.

4. NONLINEAR EFFECTS IN THE TRANSPORT OF EXCITATIONS IN A MEDIUM

We derive in this section an expression for the excitation distribution in a medium, in the form of an expansion in powers of the field up to terms of fourth order. We confine ourselves for simplicity to a traveling wave.

In the approximation considered, the atom-excitation probability takes the form

$$W_j=\frac{1+\sigma_j^3}{2}$$

$$=\frac{|V_j|^2}{|\nu|^2}-\frac{2|V_j|^4}{|\nu|^4}+\frac{2}{\gamma}\operatorname{Im}\sum_{j'\neq j}D_{jj'}^0\langle\hat f_j^+(t)\hat f_{j'}(t)\rangle_0. \quad (26)$$

The first two terms describe excitation of the atom by the mean and scattered fields at the pump-field frequency, while the last term contains the shifted resonance-fluorescence spectrum components.

For macroscopic averaging of $|V_j|^4$, the linear approximation for the field, $V_j\approx\mathscr{V}_j$ (12), is sufficient. $\langle|V_j|^2\rangle$ is calculated with the aid of (22) for a field with cubic nonlinearity. This leads to the appearance of terms of fourth order in the field, of the type $\langle\mathscr{V}^*D|\mathscr{V}|^2\mathscr{V}\rangle$.

The product of the fluctuations, averaged over the vacuum, is calculated with the aid of (20). In the "ladder" approximation it suffices to retain in the resultant sums over the atoms only the diagonal (non-oscillating) terms proportional to $|D_{jj'}|^2$, so that

$$\frac{2}{\gamma}\operatorname{Im}\sum_{j'\neq j}D_{jj'}^0\langle\hat f_j(t)\hat f_{j'}(t)\rangle_0$$

$$=\int\frac{d\omega}{2\pi}\frac{\rho(\omega)}{|\omega+\nu|^2}\sum_{j'}|D_{jj'}(\omega)|^2\frac{|V_{j'}|^4}{|\nu|^4}, \quad (27)$$

where $\rho(\omega)$ is the known spectrum of the resonant fluorescence for the shifted components[7]:

$$\rho(\omega)=\frac{4\gamma|\nu|^2}{[(\omega-\Delta)^2+\gamma^2/4][(\omega+\Delta)^2+\gamma^2/4]}. \quad (28)$$

In the macroscopic averaging of (27) we can put $\langle|D|^2|V|^4\rangle=\langle|D|^2\rangle\langle|V|^4\rangle$. We get then the propagation function for the excitations of frequency ω:

$$K(\mathbf{r}_1\mathbf{r}_2,\omega)=\frac{n}{|\omega+\nu|^2}\langle|D_{j_1j_2}(\omega)|^2\rangle, \quad (29)$$

which satisfies the integral equation

$$K(\mathbf{r}_1\mathbf{r}_2,\omega)=Q(\mathbf{r}_1-\mathbf{r}_2,\omega)+\int d^3\mathbf{r}Q(\mathbf{r}_1-\mathbf{r},\omega)K(\mathbf{r}\mathbf{r}_2,\omega),$$

$$Q(\mathbf{r},\omega)=\frac{n}{|\omega+\nu|^2}|D(\mathbf{r},\omega)|^2=\frac{\varkappa(\omega)}{2\pi r^2}e^{-2\varkappa(\omega)r}, \quad (30)$$
$$\varkappa(\omega)=2\pi k_0\operatorname{Im}\chi(\omega).$$

The functions K_0 and Q_0 considered above coincide with K and Q considered above coincide with K and Q at $\omega=0$.

The macroscopic excitation-distribution function $W(\mathbf{r})=\langle W_j\rangle$ takes ultimately the form

$$W(\mathbf{r})=w_c(\mathbf{r})-2\tilde w(\mathbf{r})+\int\frac{d\omega}{2\pi}\frac{I(\mathbf{r}\omega)}{|\omega+\nu|^2},$$

$$\frac{I(\mathbf{r}\omega)}{|\omega+\nu|^2}$$

$$=\int d^3\mathbf{r}'K(\mathbf{r}\mathbf{r}',\omega)\{2\pi\delta(\omega)[w_c(\mathbf{r}')-4\tilde w(\mathbf{r}')]+\rho(\omega)\tilde w(\mathbf{r}')\}, \quad (31)$$
$$\tilde w(\mathbf{r})=\langle|V_j|^4\rangle/|\nu|^4=w_c^2+4w_cw_s+2w_s^2.$$

The quantities w_c and w_s in these equations are expressed by Eqs. (17) and (18) in terms of the mean field $u(\mathbf{r})$ that satisfies the nonlinear Maxwell equation (25).

The problem of the passage of resonant radiation and the migration of excitations in a medium reduces thus to the solution of the nonlinear Maxwell equation (25) and of Eq. (30) for the excitation propagation function $K(\mathbf{r}_1,\mathbf{r}_2,\omega)$.

Those effects in the excitation distribution (31) which are nonlinear in the intensity are described by the terms that contain $\tilde w$. The terms with unshifted frequencies are in this case small corrections. Principal interest attaches to the last term of (31), which contains the shifted components of the scattered-field spectrum. The appearance of photons with greatly differing mean free paths alters substantially the character of the excitation diffusion in the medium. At larger Δ, for example, the medium may turn out to be transparent to the pump photons, and its excitation will be determined by the resonant photons of frequency $\omega=-\Delta$. This can be easily seen in the case when $\varkappa(\omega)l<1$ for all frequencies. The probability of exciting atoms outside the pump wave can then be written in the form

$$W(\mathbf{r})\approx\frac{1}{2\pi}\int\frac{d^3r'w_c(\mathbf{r}')}{(\mathbf{r}-\mathbf{r}')^2}\left\{\varkappa_0+\frac{1}{2}\varkappa(-\Delta)w_c(\mathbf{r}')\right\}. \quad (32)$$

The second term becomes dominant if $\varkappa(-\Delta)w_c/2\varkappa_0=2|u|^2/\gamma^2>1$. For pump fields that are not too weak ($\gamma<u\ll\Delta$) the excitation diffusion has thus a nonlinear dependence on the field intensity and is determined by the shifted components of the scattered radiation.

The function $I(\mathbf{r},\omega)$ in (31) has the meaning of the spectral density of the scattered radiation. It can be shown that

$$I(\mathbf{r},\omega)=\left\langle\int dt\langle\tilde U_j^+(t)\tilde U_j(0)\rangle_0e^{i\omega t}\right\rangle, \quad (33)$$
$$\tilde U_j(t)=\tilde V_j(t)-u(\mathbf{r}).$$

For a small scattering volume, $I(\mathbf{r}\omega)$ is proportional to the spectrum of the resonance fluorescence of a single atom.

5. DISCUSSION

Thus, when a strong electromagnetic field passes through a resonant medium of sufficiently large optical thickness the nonlinear susceptibility of the gas becomes nonlocal because of the dragging of the scattered radiation. This can manifest itself in various nonlinear optical effects.

By way of example we consider the interaction of two light beams propagating at a distance ρ between them. The scattered field generated by one of the beams (of thickness a) excites atoms in the region where the other beam passes and produces there an effective susceptibility [see Eq. (25)]:

$$\chi_{eff}(\rho) = -4\chi_0 w_s(\rho) \sim -\chi_0 \frac{\varkappa_0 a^2}{\rho} w_c.$$

The spatial inhomogeneity of this susceptibility leads to attraction (at $\Delta > 0$) or repulsion (at $\Delta < 0$) of the second (test) beam. We note that beam attraction and self-focusing occur at the same sign of the detuning. The deviation angle of the test beam can be estimated from the relation $\theta \sim (\pi l/\rho) \cdot \mathrm{Re}\chi_{eff}$ (l is the length of the light beams). Under conditions of noticeable saturation ($w_c \sim 1$) and absorption ($a \sim \rho \sim l \sim 1/\varkappa$) the value of θ can become of the order of $\mathrm{Re}\chi_0 \sim 10^{-3}$.

Another property of the nonlocal susceptibility is due to the fact that the scattered field depends strongly on the spatial structure of the pump wave. In particular, for a standing wave the Maxwell equation (24) contains an anomalous nonlinear term, which leads to an effect of the wave-front-reversal type. The reversal of a test signal from a region where the atoms are acted upon by a strong pump wave is well known.[9] It can be seen from (24) that the test signal can be reflected also from a region containing not a coherent standing wave but only scattered radiation produced by this wave. In other words, propagation of excitations in a medium cause transport of not only the intensity but also of the square of the amplitude, so that the scattered radiation retains some memory of the correlation properties of the pump wave.

The amplitude of the inverted signal builds up to a value of the order of the test-signal amplitude over a length $l \sim k_0 \,\mathrm{Re}\,\chi_0 w_{s\,st}$. For the wave-front inversion to be substantial, it is necessary that this length be shorter than the absorption length $1/\varkappa$. This can be achieved under conditions of noticeable absorption $\varkappa R \sim 1$ (R is the width of the standing wave) and at a sufficient intensity of the standing-wave amplitude: $\Delta\gamma < u_{st}^2 \ll \Delta^2$.

The nonlinear effects in radiation dragging can thus influence significantly the propagation of radiation through a resonant medium.

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