Radiative interaction between atoms

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The effect of the scattered field on the passage of an intense light wave through a resonant medium is studied. Radiative interaction between the atoms, due to absorption and emission of scattered quanta, alters the spectral properties of the atoms and the constant of the interaction with the external field. These renormalizations are of nonlocal nature and become significant under conditions of appreciable absorption and saturation. To describe this group of phenomena in the low density approximation, a closed system of equations is obtained for the density matrix, for the correlation function, and for mean field strength. The renormalizations for weak saturation and absorption are found with the aid of the equations. The nonlinear susceptibility of the medium, which is nonlinear in the field intensity and density, depends on the size of the light beam; this results in a decrease of the absorption coefficient with increase of the beam diameter.

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§1. INTRODUCTION

We investigate here the influence of scattered light on the propagation of strong electromagnetic radiation through a resonant medium. The resonant field is the resonant-fluorescence field of individual atoms. In view of the large resonant scattering cross section, the mean free path of the quantum may become comparable with the dimensions of the system even at low atom densities. When the scattered-field intensity becomes large enough, account must be taken of its action on the atoms of the medium, in addition to the action of the mean field. In other words, the field acting on the atom can differ substantially from the mean field in the medium.

It is known that in an optically dense medium the difference between the effective field and the mean field is due to the singularity of the dipole-dipole interaction of the atoms at short distances. This effect is described by the Lorentz field,[1] which usually leads to local renormalization of the mean field. As applied to a resonant medium, the Lorentz correction becomes substantial if $n\lambda^3 \gtrsim 1$, where n is the density of the atoms and λ is the resonant wavelength.[2-4] We consider a low-density medium:

$$n\lambda^3 \ll 1. \tag{1}$$

In this case the interaction between the atoms is via exchange of real quanta, while the electrostatic interaction is negligible. However, even in this case, as will be shown below, the field acting on the atoms becomes renormalized. This takes place if the photon mean free path is comparable with the dimensions of the light beam and the saturation effect is of the order of unity.

The question considered is closely connected with the problem of radiation dragging.[5-8] In the radiation-dragging theory, the energy of the system is determined mainly by the number of excited atoms. The excitations are diffused via emission and absorption of quanta. This process, which is of noncoherent character, is described by an equation of the Holstein-Biberman type for the population of the upper levels.

In our case, the atoms are excited both coherently, on account of the mean electromagnetic field, and incoherently on account of absorption of the scattered quanta. To describe these processes we must use the equations for the atom density matrix and the equations for the correlation function, together with Maxwell's equations for the mean field. These equations are derived for a low-density gas of atoms in §§2, 3.

We study next these equations by perturbation theory in the form of an expansion in powers of the density (§§5, 6). The absorption of the scattered radiation leads to renormalization of the atomic characteristics, of the mean field, and of the dielectric constant. The effect of self-action of the field is nonlocal. Therefore the renormalized quantities depend on the shape and size of the system.

§2. BASIC EQUATIONS

The Hamiltonian of the system of two-level atoms interacting with a resonant electromagnetic field is of the form ($\hbar = c = 1$):

$$\sum_{\mathbf{k}} (\omega_{\mathbf{k}} - \omega_0) a_{\mathbf{k}}^+ a_{\mathbf{k}} + \sum_i [\sigma_+^{(i)} (\hat{V}(\mathbf{r}_i) + V_0(\mathbf{r}_i, t)) + \text{H. adj.}],$$
$$\hat{V}(\mathbf{r}) = g \sum_{\mathbf{k}} a_{\mathbf{k}} e^{i\mathbf{k}\mathbf{r}}, \quad g = -d(2\pi\omega_0/V)^{1/2}, \tag{2}$$
$$V_0(\mathbf{r}t) = V_0(\mathbf{r}) e^{-i\Delta t}, \quad V_0(\mathbf{r}) = -\mathbf{d}\mathbf{E}_0(\mathbf{r}).$$

Here ω_0 and d are the frequency and dipole moment of the transition, σ are Pauli matrices, $\hat{V}$ is the operator of the quantized scattered field, V_0 is an external class classical field with small detuning $\Delta = \omega - \omega_0$, and Ω is the volume of the system. The first term in (2) corresponds to the free field, the second describes the interaction of the atoms with the "bare" external field V_0 and with the scattered field $\hat{V}$.

We neglect recoil and assume that the particles move in linear trajectories $\mathbf{r}_i(t) = \mathbf{r}_{i0} + \mathbf{v}_i t$. To simplify the calculations we disregard likewise the polarization properties of the medium and of the radiation. The Heisenberg equations of motion are most compactly written in four-dimensional form. The reason is that

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the matrices $\sigma_\alpha=(1,\sigma_-,\sigma_+,\sigma_3)$ make up a complete set of matrices.

We introduce the four-component operator $\hat{R}_\alpha$ of the medium:

$$\hat{R}_\alpha(\mathbf{r}\mathbf{v}t)=\sum_i \sigma_\alpha^{(i)}\delta(\mathbf{r}-\mathbf{r}_i)\delta(\mathbf{v}-\mathbf{v}_i), \quad \alpha=0,1,2,3, \tag{3}$$

where $\hat{R}_0$ is the density operator in phase space, $\hat{R}_{1,2}$ are the positive- and negative-frequency parts of the dipole moment, and $\hat{R}_3$ is the population-difference operator. The equation for $\hat{R}$ is of the form

$$id\hat{R}/dt=(\mathcal{V}^+ +V_0^*)B\hat{R}+A\hat{R}(\mathcal{V}+V_0), \tag{4}$$
$$A_{00}=A_{0i}=A_{i0}=0, \quad A_{ij}=a_{ij},$$
$$B_{00}=B_{0i}=B_{i0}=0, \quad B_{ij}=b_{ij} \quad (i,j=1,2,3),$$
$$\hat{a}=\begin{pmatrix}0&0&-1\\0&0&0\\0&2&0\end{pmatrix}, \quad \hat{b}=\begin{pmatrix}0&0&0\\0&0&1\\-2&0&0\end{pmatrix}.$$

$d/dt=\partial/\partial t+\mathbf{v}\nabla$ denotes here the total derivative with respect to time. The Hermitian-adjoint vector $\hat{R}^*$ is obtained from $\hat{R}$ by permuting[1)] the components 1 and 2:

$$\hat{R}^+=\hat{T}\hat{R}, \quad T_{\alpha\beta}=\delta_{\alpha\beta}(\alpha,\beta=0,3)=(\sigma_1)_{\alpha\beta} \quad (\alpha,\beta=1,2).$$

Equation (4) is self-adjoint, since the matrices $\hat{A}$ and $\hat{B}$ satisfy the condition $\hat{T}\hat{A}\hat{T}=-\hat{B}$ and $\hat{T}\hat{B}\hat{T}=-\hat{A}$. The matrix adjoint to $\hat{S}$ will hereafter be taken to mean the matrix $-\hat{T}\hat{S}\hat{T}$

$$\hat{S}+\text{ adj }=\hat{S}-\hat{T}\hat{S}\hat{T}.$$

The quantized-field operator satisfies the abbreviated Maxwell's equation

$$(2i\omega_0\partial/\partial t+\omega_0^2+\nabla^2)\hat{V}=4\pi\omega_0^2 d^2\int d\mathbf{v}\hat{R}_1(\mathbf{r}\mathbf{v}t), \tag{5}$$

whose solution can be represented in the form

$$\hat{V}(\mathbf{r}t)=\hat{V}_0(\mathbf{r}t) -2\pi i\omega_0 d^2\sum_{\mathbf{k}}\int d\Gamma'\int_{-\infty}^{t}dt'\exp[i\mathbf{k}(\mathbf{r}-\mathbf{r}')-i(\omega_{\mathbf{k}}-\omega_0)(t-t')]\hat{R}_1(\mathbf{r}'\mathbf{v}'t'), \tag{5'}$$

where $\hat{V}_0(\mathbf{r}t)$ is the operator of the free field (of the zero-point oscillations), and $d\Gamma'=d\mathbf{r}'d\mathbf{v}'$ is the phase-space element. Using the resonance approximation, we can simplify the Green's function of the electromagnetic field. Indeed, the main contribution to the sum over $\mathbf{k}$ in (5′) is made by wave numbers those modulus is close to the resonant wave number $k_0=\omega_0$. Assuming wherever possible $k=k_0$, we integrate with respect to $k-k_0$ between infinite limits. The contribution made to the radiation field at a given point by atoms located at a finite distance from this point is determined by the retarded Green's function. On the other hand, the field produced by the atom at the location of the atom itself is determined, as is known, by the arithmetic mean of the retarded and advanced solutions of the wave equation. This is how the relaxation of an isolated atom in the course of radiation is described.

Under real conditions it is frequently possible to neglect the relativistic retardation and the slow dependence of the radiation period of the time: $l\ll c\tau$, where l is the linear dimension of the system and τ is a characteristic time of the order of the atom lifetime or of the order of the reciprocal Doppler width. As a result of these simplifications, Eq. (5′) takes the form

$$\hat{V}(\mathbf{r}t)=\hat{V}_0(\mathbf{r}t)+\int d\Gamma' D(\mathbf{r}-\mathbf{r}')\hat{R}_1(\mathbf{r}'\mathbf{v}'t),$$
$$D(\mathbf{r})=-\frac{\gamma}{2}\begin{cases}\exp(ik_0r)/k_0r, & r\neq 0,\\ i, & r=0,\end{cases} \quad \gamma=2k_0^3d^2, \tag{6}$$

where γ is the width of the upper working level of the isolated atom.[2)]

Thus, the slow dependence of the electromagnetic-field operator $\hat{V}(\mathbf{r}t)$ is determined by the state of the medium at the same instant of time. Equations (4) and (6) are the initial operator equations for the "atoms + radiation" system in the presence of an external field V_0.

§3. EQUATION FOR ATOMIC CORRELATORS

In the quantum and statistical averaging of (4) and (6) there appears a chain of equations that relate distribution functions of different order. We consider hereafter the case of strong inhomogeneous broadening (V_0 is the characteristic thermal velocity)

$$\Delta_0=kv_0\gg\gamma, \ |\Delta|. \tag{7}$$

We can then confine ourselves to calculation of only single-time correlators.[3)] The condition (1) allows us to close the resultant chain of equations in the second step, neglecting the irreducible part of the third-order correlator.

We introduce the atom-density matrix:

$$R_\alpha(\mathbf{r}\mathbf{v}t)=\langle\hat{R}_\alpha(\mathbf{r}\mathbf{v}t)\rangle,$$

where the angle brackets denote quantum-statistical averaging over the initial stage of the system with zero photon occupation numbers. With the aid of (6) we can exclude the electromagnetic field from Eq. (4). Although $\hat{V}$ and $\hat{R}$ cummute at the same instant of time, is useful to employ the sequence adopted in (4) for these operators, so that $\hat{V}$ is to the right of R and $\hat{V}^*$ to the left. This allows us to exclude the constribution from the zero-point oscillations of the field.[11,12]

The mean value of the product of two operators $\hat{R}$ can be represented in the form[4)]

$$\langle\hat{R}_{\alpha_1}(1)\hat{R}_{\alpha_2}(2)\rangle=C_{\alpha_1\alpha_2,\,\alpha}\delta(1-2)R_\alpha(1)+R_{\alpha_1}(1)R_{\alpha_2}(2)+R_{\alpha_1\alpha_2}(1,2). \tag{8}$$

Here $R_{\alpha_1\alpha_2}(1,2)$ is an irreducible two-particle correlation function that is symmetrical with respect to simultaneous permutation of the indices and the arguments: $R_{\alpha_1\alpha_2}(1,2)=R_{\alpha_2\alpha_1}(2,1)$. The irreducible part is small compared with the reducible $R_{\alpha_1}(1)R_{\alpha_2}(2)$ in terms of the parameter (1). The coefficients $C_{\alpha_1\alpha_2\alpha_3}$ are determined by the algebra of the Pauli matrices. We shall present below the explicit form of those coefficients which enter in the final equations. The equation for the Bloch vector $R(1)$ takes the form

$$i\,dR(1)/dt=\hat{\mathcal{H}}(1)R(1)+\int d\Gamma_2[D(1-2)\hat{A}(1)R_{\cdot 1}(1,2) +D^*(1-2)\hat{B}(1)R_{\cdot 2}(1,2)],$$
$$\mathcal{H}_{0\beta}=0, \quad \mathcal{H}_{\alpha 0}=-i\gamma\delta_{\alpha 3}, \quad \mathcal{H}_{ij}=H_{ij}, \tag{9}$$
$$\hat{H}=\begin{pmatrix}-i\gamma/2&0&-V\\0&-i\gamma/2&V^*\\-2V^*&2V&-i\gamma\end{pmatrix};$$

$$V(\mathbf{r}t)=V_0(\mathbf{r}t)+\int d\Gamma' D(\mathbf{r}-\mathbf{r}')R_1(\mathbf{r}'\mathbf{v}'t). \tag{10}$$

The Bloch operator $\hat{H}$ describes transitions in the atom on account of the coherent field V and the relaxation on account of the spontaneous emission. The mean field V is the sum of the bare (external incident field) and scattered fields. The integral term with the correlation function in (9) takes into account the interaction of the atoms via the scattered field and leads, as we shall show, to a renormalization of the quantities that enter in the Bloch operator.

The symbolic form of the tensor quantities $R\cdot_1(1,2)$ and $R\cdot_2(1,2)$ used in (9) as well as some equations that follow means that the first index is abritrary and the second is fixed. Thus, for example, $(\hat{A}(1)R\cdot_1(1,2)_\alpha = A_{\alpha\beta}R_{\beta 1}(1,2)$.

The equation for the correlation function $R(1,2)$ contains the mean value of three operators $\hat{R}$, which can be written in the form

$$\langle \hat{R}_{\alpha_1}(1)\hat{R}_{\alpha_2}(2)\hat{R}_{\alpha_3}(3)\rangle = \delta(1-2)\delta(1-3)C_{\alpha_1\alpha_2,\,\alpha}C_{\alpha'\alpha_3,\,\alpha}R_{\alpha}(1) + R_{\alpha_1}(1)R_{\alpha_2}(2)R_{\alpha_3}(3) + \{\delta(1-2)C_{\alpha_1\alpha_2,\,\alpha}[R_{\alpha}(1)R_{\alpha_3}(3) + R_{\alpha\alpha_3}(1,3)] + R_{\alpha_1\alpha_2}(1,2)R_{\alpha_3}(3) + (123)\to(231),\ (312)\} + R_{\alpha_1\alpha_2\alpha_3}(1,2,3).$$

The fully irreducible part $R(123)$ contains a higher power of the small parameter $n\lambda^3$ compared with the remaining functions, and will therefore be disregarded hereafter. As a result $R(1,2)$ satisfies the equation

$$i(\partial/\partial t + \mathbf{v}_1\nabla_1 + \mathbf{v}_2\nabla_2)R(1,2) = (\hat{\mathscr{H}}(1) + \hat{\mathscr{H}}(2))R(1,2) + \{[D(1-2)\hat{A}(1)\hat{C}_a(2) + D^*(1-2)B(1)\hat{C}_b(2)]R(1)R(2) + \int d\Gamma_3[D(1-3)\hat{A}(1)R(1)R_1(2,3) + D^*(1-3)B(1)R(1)R_2(2,3)] + 1\rightleftarrows 2\}, \tag{11}$$

$$\hat{C}_a \equiv C_{\alpha 1,\beta} = \begin{pmatrix} 0 & 0 & 1 & 0 \\ 1/2 & 0 & 0 & 1/2 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 \end{pmatrix}, \quad \hat{C}_b \equiv C_{2\alpha,\beta} = \begin{pmatrix} 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 1/2 & 0 & 0 & 1/2 \\ 0 & -1 & 0 & 0 \end{pmatrix}.$$

The matrices $\hat{C}_a$ and $\hat{C}_b$ are mutually adjoint: $\hat{T}\hat{C}_a\hat{T} = -\hat{C}_b$. The notation $1 \rightleftarrows 2$ corresponds to permutation of the arguments and indices of particles 1 and 2. The indices of the third particle, which are written out in explicit form, are not affected by this. Averaging (5), we obtain Maxwell's equation for $V(\mathbf{r}t)$:

$$(2i\omega_0\partial/\partial t + \omega_0^2 + \nabla^2)V = 4\pi d^2\omega_0^2 \int d\mathbf{v}R_1(\mathbf{r}\mathbf{v}t). \tag{12}$$

Equations (10) and (12) for the mean field are equivalent apart from small terms connected with the relativistic retardation. It is more convenient, however, to use Eq. (12), since it does not contain the bare field explicitly.

Thus, Eqs. (9), (11), and (12) constitute a closed system that describes the self-consistent interaction of the mean field with the resonant medium. The effect of the scattered field on the system is taken into account with the aid of the correlator $R(1,2)$. We emphasize that the closure of the system of equations is with respect to the parameter $n\lambda^3$. As for the amplitude of the external field and the dimensions of the system, they can be arbitrary.

§4. DRAGGING OF THE RADIATION

By way of illustration we consider the known problem of the diffusion of excitations in a medium of resonant atoms under radiation dragging conditions.[5-8]

Thus, assume that there is no coherent field, $V = 0$. The atom density matrix is diagonal ($R_1 = R_2 = 0$) and the density of the excited atoms $N = (R_0 + R_3)/2$ is assumed small: $N \ll f = (R_0 - R_3)/2$. The problem consists of obtaining for $N(\mathbf{R}\cdot\mathbf{v}t)$ an equation that describes the process of diffusion of excitations in the medium. We can assume an equilibrium distribution of the atoms in the ground state:

$$f(v) = \frac{n}{\pi^{3/2}v_0^3}\exp\left(\frac{-v^2}{v_0^2}\right).$$

Only two of all the components of the correlation function differ from zero, namely $R_{12}(1,2) = \varphi(1,2)$ and $R_{21}(1,2) = \varphi(2,1)$. We then obtain from (9) and (11)

$$i(d/dt+\gamma)N(1) = \int d\Gamma_2[-D^*(1-2)\varphi(1,2) + D(1-2)\varphi(2,1)],$$
$$i(\partial/\partial t + \mathbf{v}_1\nabla_1 + \mathbf{v}_2\nabla_2 + \gamma)\varphi(1,2) = N(1)D(1-2)f(2) - f(1)D^*(1-2)N(2) + \int d\Gamma_3[f(2)D(2-3)\varphi(1,3) - f(1)D^*(1-3)\varphi(3,2)]. \tag{13}$$

The kernel in the equation for φ is the function $D(\mathbf{r})$, which oscillates over distances on the order of the wavelength. Therefore in large volumes (much larger than λ^3) we can use a Fourier transformation. In the quasistationary approximation, the Fourier transform of the correlation function is

$$\varphi(\mathbf{p},\varkappa,\mathbf{v}_1,\mathbf{v}_2) = \frac{f(\mathbf{v}_1)F(\mathbf{p},\varkappa,\mathbf{v}_2) - f(\mathbf{v}_2)F^*(\mathbf{p},-\varkappa,\mathbf{v}_1)}{\mathbf{p}(\mathbf{v}_2-\mathbf{v}_1)+i\gamma},$$

where the function F satisfies an integral equation with a Cauchy kernel

$$F(\mathbf{p},\varkappa,\mathbf{v})\tilde{D}^{-1}\left(\mathbf{p}+\frac{\varkappa}{2},\mathbf{p}\mathbf{v}\right) = N(\varkappa,\mathbf{v}) + f(\mathbf{v})\int\frac{d\mathbf{v}_1F^*(\mathbf{p},-\varkappa,\mathbf{v}_1)}{\mathbf{p}(\mathbf{v}_1-\mathbf{v})-i\gamma},$$
$$\frac{1}{\tilde{D}(\mathbf{q},\omega)} = \frac{1}{D(\mathbf{q})} + \frac{\chi_0(\omega)}{d^2}, \quad \chi_0(\omega) = d^2\int\frac{d\mathbf{v}f(\mathbf{v})}{\mathbf{p}\mathbf{v}-\omega-i\gamma}. \tag{14}$$

Here $\tilde{D}(\mathbf{q},\omega)$ is the Green's function of the photon in the medium, and $\chi_0(\omega)$ is the linear susceptibility of the gas. An equation of this type is encountered, for example, when describing the correlation properties of the plasma[14,15] and is solved by methods known from the theory of singular integral equations.[16] As a result we obtain

$$F(\mathbf{p},\varkappa,\mathbf{v}) = \tilde{D}\left(\mathbf{p}+\frac{\varkappa}{2},\mathbf{p}\mathbf{v}\right)N(\varkappa,\mathbf{v}) + f(\mathbf{v})\int\frac{d\mathbf{v}_1N(\varkappa,\mathbf{v}_1)}{\mathbf{p}(\mathbf{v}_1-\mathbf{v})-i\gamma}\tilde{D}\left(\mathbf{p}+\frac{\varkappa}{2},\mathbf{p}\mathbf{v}_1\right)\tilde{D}^*\left(\mathbf{p}-\frac{\varkappa}{2},\mathbf{p}\mathbf{v}_1\right). \tag{15}$$

Substituting this solution in (14), we obtain an equation of the Holstein-Biberman type:

$$(d/dt+\gamma)N(\mathbf{r}\mathbf{v}t) = \gamma\int d\Gamma' K(\mathbf{r}-\mathbf{r}',\mathbf{v},\mathbf{v}')N(\mathbf{r}'\mathbf{v}'t),$$
$$K(\mathbf{r}\mathbf{v}\mathbf{v}') = \pi d^2 f(\mathbf{v})\int\frac{d\varkappa e^{i\varkappa\mathbf{r}}}{(2\pi)^3}\int\frac{d\mathbf{n}\delta(\mathbf{n}\mathbf{v}-\mathbf{n}\mathbf{v}')}{i\varkappa\mathbf{n}+4\pi k\,\mathrm{Im}\,\chi_0(k\mathbf{n}\mathbf{v})} \tag{16}$$

where $\mathbf{n}$ is a unit vector. Apart from numerical coefficients that take into account the polarization properties of the atoms and of the light, Eq. (16) coincides with the equation obtained by D'yakonov and Perel'.[7]

§5. CASE OF SMALL ABSORPTION

The solution of integral equations for correlation functions in the presence of an external field simplifies greatly in the case of small absorption of the radiation.

When the dimension of the medium l is less than the photon mean free path $\varkappa^{-1}$

$$\varkappa l<1, \quad \varkappa=2\pi k \operatorname{Im}\chi_0, \tag{17}$$

the scattered field is weak and its influence can be taken into account by perturbation theory. Leaving out of (11) the integral terms, we obtain for the correlation functions equations of the Bloch type (of double the dimensionality), with a right-hand side quadratic in $R_\alpha(\mathbf{r}\cdot\mathbf{v}t)$. In three-dimensional notation

$$R(1)=\{f(1),\ R_i(1)\},$$
$$R(1,2)=\left(\begin{array}{c|c} R_{00}(1,2) & g_j(1,2) \\ \hline g_i(2,1) & R_{ij}(1,2) \end{array}\right), \quad i,j=1,2,3,$$

Eqs. (11) decay into systems of equations for R_{00}, g_i, and R_{ij}.

For a scalar, the equation is trivial:

$$dR_{00}/dt_{12}\equiv(\partial/\partial t+\mathbf{v}_1\nabla_1+\mathbf{v}_2\nabla_2)R_{00}(1,2)=0.$$

The function R_{00} is determined by its initial value as $t\to-\infty$, when the external field was absent and there was no correlation between the atoms. We can therefore put $R_{00}=0$. The equations for the vector $g_i(1,2)$ and the tensor $R_{ij}(1,2)$ are

$$i\frac{d}{dt_{12}}g_i(1,2)=H_{ij}(2)g_j(1,2)+[D(2-1)R_i(1)a_{ij}(2)+\text{adj.}]R_j(2), \tag{18}$$

$$i\frac{d}{dt_{12}}R_{ij}(1,2)=H_{ii'}(1)R_{i'j}(1,2)+H_{jj'}(2)R_{ij'}(1,2)-i\gamma\delta_{is}g_j(1,2)$$
$$-i\gamma\delta_{js}g_i(2,1)+{}^1/_2\{D(1-2)a_{ii'}R_{i'}(1)[\delta_{j2}f(2)+b_{jj'}R_{j'}(2)]+\text{adj.}+1\rightleftarrows2\}. \tag{19}$$

The distribution function of the atoms remains unchanged in the assumed approximation:

$$(\partial/\partial t+\mathbf{v}_1\nabla_1)f(1)=0$$

and coincides with the equilibrium spatially homogeneous distribution $f(v)$. The Bloch equation in three-dimensional form is written in the following manner:

$$i\frac{dR_i(1)}{dt}=H_{ij}(1)R_j(1)-i\gamma\delta_{is}f(1)+\int d\Gamma_2[D(1-2)a_{ij}R_{js}(1,2)+\text{adj.}]. \tag{20}$$

We consider further the case of a traveling wave:

$$V(\mathbf{r}t)=V\exp[i(\mathbf{k}\mathbf{r}-\Delta t)], \quad |\mathbf{k}|=k_0.$$

After going over into a rotating coordinate frame, the operator $\hat{H}(\mathbf{r}t)$ ceases to depend explicitly on the coordinate and the time:

$$\hat{H}(\mathbf{r}t)\to\hat{H}(1)=\begin{pmatrix} -i\gamma/2-\Delta_1 & 0 & -V \\ 0 & -i\gamma/2+\Delta_1 & V^* \\ -2V^* & 2V & -i\gamma \end{pmatrix}, \tag{21}$$

$\Delta_1=\Delta-\mathbf{k}\cdot\mathbf{v}_1$, and the Green's function (7) of the photon acquires an additional phase factor

$$D(\mathbf{r})\to\bar{D}(\mathbf{r})=D(\mathbf{r})e^{-i\mathbf{k}\mathbf{r}}. \tag{22}$$

We obtain now the stationary solution for the correlation functions. The coordinate dependence is determined by the function $\bar{D}(\mathbf{r})$. In the action of the operator $\mathbf{v}\nabla$ on $\bar{D}$ it suffices to differentiate only the exponential $\exp[1(k_0r-\mathbf{k}\cdot\mathbf{r})]$. The remaining functions vary little over the wavelength, and they need not be differentiated. Then Eqs. (18) and (19) reduce to algebraic Bloch equations with a certain right-hand side that describes the radiative interaction of the atoms.

Substituting the solutions of these equations in the integral term of Eq. (20), we retain only the nonoscillating terms with $|\bar{D}(\mathbf{r}_1-\mathbf{r}_2)|^2$, which make a contribution proportional to the linear dimension of the system. The integral term in Eqs. (20) is now expressed in terms of the product of the vectors $R_i(1)R_j(2)$ and adds a correction to the operator $\hat{H}(1)$. If we use for $R_j(2)$ the lowest-order approximation in the density:

$$R_j(2)=i\gamma\hat{H}_{js}^{-1}(2)f(2),$$

Eq. (20) reduces to a Bloch equation with a renormalized interaction

$$i\frac{dR_i}{dt}=(H_{ij}+\delta H_{ij})R_j+i\gamma\delta_{is}f, \quad \delta\hat{H}=\hat{a}\hat{h}\hat{b}+\text{adj.}, \tag{23}$$

$$h_{ij}(1)=\langle{}^1/_2u_{ij,11}^{-1}(1,2)[1+i\gamma H_{ss}^{-1}(2)]$$
$$-i\gamma u_{ii',13}^{-1}(1,2)[\delta_{i'j}+i\gamma(\xi-\hat{H}(1))_{i'j}^{-1}]H_{2s}^{-1}(2)\rangle; \tag{24}$$

$$\langle\ldots\rangle=\int d\Gamma_2 f(2)|\bar{D}(1-2)|^2\ldots,$$
$$\hat{u}(1,2)=\xi-\hat{H}(1)-\hat{H}(2), \quad \xi=(\mathbf{v}_1-\mathbf{v}_2)(k_0\mathbf{n}_{12}-\mathbf{k}),$$
$$\mathbf{n}_{12}=(\mathbf{r}_1-\mathbf{r}_2)/|\mathbf{r}_1-\mathbf{r}_2|.$$

The correction $\delta\hat{H}$ to the Bloch operator is linear in the atom density.

The interaction of the atoms via the scattered field is incoherent and is determined by the sum of the intensities of the fields scattered by the different atoms. This manifest itself in the fact that the integration in $\hat{h}(1)$ is carried out with the kernel $|\bar{D}|^2$, which describes the law that governs the decrease of the scattered-field intensity. The structure of the operator $\delta\hat{H}$ coincides with the structure of $\hat{H}$. Therefore allowance for the interaction of the atoms via the scattered field reduces to a renormalization of the Bloch operator

$$\hat{H}+\delta\hat{H}=\begin{pmatrix} -i\gamma_\perp-(\tilde{\Delta}-\mathbf{k}\mathbf{v}) & 0 & -V_\perp \\ 0 & -i\gamma_\perp+(\tilde{\Delta}-\mathbf{k}\mathbf{v}) & V_\perp^* \\ -2V_\parallel^* & 2V_\parallel & -i\gamma_\parallel \end{pmatrix}. \tag{25}$$

The transition frequency and the longitudinal and transverse relaxation times are renormalized, and in addition two constants of the interaction with the average field appear, which can be naturally called longitudinal and transverse. The renormalized quantities depend on the size and shape of the volume occupied by the atom and by the field, and vary slowly with the coordinate.

We note that the operator (25) is T-self-adjoint.

§6. RENORMALIZATION IN A WEAK FIELD

In an arbitrary field V, to find the renormalizations one must find the inverse Bloch operators $\hat{H}^{-1}$ and $\hat{u}^{-1}$. To simplify (24), we consider a weak field, when the saturation parameter is small:

$$W(\Delta)=2|V|^2/(\Delta^2+\gamma^2/4)\ll1. \tag{26}$$

The expansion, in terms of the field, in the operators contained in (24) begins with V^2. Retaining terms up to V^3, we obtain

$$i\gamma_\perp+\tilde{\Delta}=i\gamma/2+\Delta-\langle W(\Delta_2)(\xi+i\gamma)^{-1}\rangle, \tag{27}$$
$$\gamma_\parallel=\gamma[1+\langle W(\Delta_2)((\xi-\Delta_1)^2+\gamma^2/4)^{-1}\rangle],$$
$$\left.\begin{matrix} V_\perp \\ V_\parallel^* \end{matrix}\right\}=\left.\begin{matrix} V \\ V^* \end{matrix}\right\}[1+\langle W(\Delta_2)(\xi+i\gamma)^{-1}(\xi-\Delta_1+i\gamma/2)^{-1}\rangle].$$

The renormalizations in (27) are proportional to the intensity of the average field. This is perfectly natural, since the intensity of the scattered field is determined by the density of the excited atoms, which increases with increasing pump field in the case of weak saturation.

We see that the longitudinal and transverse components of the Bloch operator are generally speaking differently renormalized. The renormalization depends on the correlation between the position of the given atom and the Doppler frequency shift of the radiation scattered by other atoms.

Expressions (27) take the simplest form for slow atoms ($v \ll v_0$):

$$\begin{aligned} \gamma_{\parallel}&=2\gamma_{\perp}=\gamma[1+W(0)\varkappa l_1\gamma/4\pi^{1/2}\Delta_0], \\ V_{\parallel}&=V_{\perp}=V[1-W(0)\varkappa l_2(\gamma/2\Delta_0)^2], \\ \bar{\Delta}&=\Delta-\pi^{-1}W(0)\,(\gamma/2\Delta_0)^2\varkappa l(k\mathbf{v}-\Delta\mathbf{k}/k), \\ \{l_1, l_2, l_3, \mathbf{l}\}&=\int d\mathbf{r}'\,(\mathbf{r}-\mathbf{r}')^{-2}\{1/n_{\perp}, 1/n_{\perp}^2, 1/(1+n_{\perp}^2)^{1/2}, (\mathbf{n}+\mathbf{k}/k)/n_{\perp}^2\}. \end{aligned} \tag{28}$$

Here $\mathbf{n}=(\mathbf{r}-\mathbf{r}')/|\mathbf{r}-\mathbf{r}'|$, $n_{\perp}=|\mathbf{n}\times\mathbf{k}/k|$. The parameters l_i are proportional to the linear dimension of the system and depend on the shape and size of the volume of the scattering medium, as well as on the position of the atom. In particular, for an atom located near the center of a spherical volume of diameter L, we have $l_1=\pi^2 L$. The parameter l_2 diverges logarithmically at small angles, owing to the low-velocity approximation. The cutoff at the angles $\sim|\Delta/\Delta_0|$, ($|\Delta|\gtrsim\gamma$), leads to the appearance of an additional logarithmic factor, so that $l_2=\pi L\ln|\Delta_0/\Delta|$.

In the low-velocity approximation, the renormalizations of the longitudinal and transverse quantities are equal and do not depend on the atom velocity (with the exception of $\bar{\Delta}$). As a result of the interaction with the scattered radiation, the atomic widths increase, and the constant of the interaction with the external field decreases.

We note that for atoms moving along a light beam and at resonance with the field ($\mathbf{v}=\Delta\mathbf{k}/k^2$), the effective detuning is not renormalized ($\bar{\Delta}=\Delta$). The renormalizations are proportional to the fraction of the absorbed energy of the scattered field $W\varkappa l$. The appearance of the parameter γ/Δ_0 is due to the fact that the only atoms that interact resonantly with one another are those having close velocities along the direction of the line joining them.

The interaction of the atoms leads also to a renormalization of the dielectric constant of the medium. Substituting in Maxwell's equation (12) the stationary solution of the equations (23), obtained accurate to V^3, we obtain the nonlinear (in the field and in the density) susceptibility of the medium

$$\begin{aligned} \mathrm{Re}\,\chi&=\mathrm{Re}\,\chi_0\Big[1-\frac{\pi^{1/2}W(0)\gamma}{4\Delta_0}\Big(1+\frac{\varkappa l_3}{\pi}\Big)\Big], \\ \mathrm{Im}\,\chi&=\mathrm{Im}\,\chi_0[1-{}^1/_2 W(0)\,(1+\varkappa l_1\gamma/2^{3/2}\pi^{1/2}\Delta_0)], \\ \chi_0&=-\frac{nd^2}{\Delta_0}\Big(\frac{2\Delta}{\Delta_0}-i\pi^{1/2}\Big). \end{aligned} \tag{29}$$

It follows from this expression that the contribution to the susceptibility from the reradiation processes leads to an effective increase of the saturation parameters, making the medium more transparent.

We note that if the field saturates the entire Doppler contour with $\varkappa l\sim 1$, the susceptibility renormalization becomes equal to unity.

§7. DISCUSSION. CONCLUSION

We see thus that when a strong light wave passes through a medium with noticeable absorption, the scattered field exerts a substantial influence both on the atomic characteristics of the medium and on the strong field itself. In other words, allowance for the "dragging" of the radiation in the region of the pump field leads to changes in the line width, in the detuning from resonance of the transition, and in the amplitude of the interaction with the field.

We shall name the emission and absorption of scattered-field quanta "radiative interaction of atoms." It becomes significant when the size of the system is of the order of the absorption length and the saturation effect is of the order of unity. In alkali-metal atom vapors, the Doppler-broadened contour saturates in fields 10–100 W/cm^2, and the absorption length is ~1 cm at densities $n\sim 10^{12}$–10^{13} cm^{-3}. Thus, the effects of radiative interaction can be observed with the aid of both cw lasers and pulsed lasers, if the pulse duration exceeds the spontaneous relaxation time.

The field acting on the atom is a sum of the coherent mean field and of the incoherrent scattered field, which is not monochromatic and contains random phases. For this reason, the change of the atomic characteristics is determined by the intensity of the scattered field, and the kernel of the integral correction terms behaves like r^{-2}.

Inasmuch as in a radiation field the momentum flux is equal to the energy flux (divided by the speed of light), the radiative correction of the atoms is also a source of their ponderomotive interaction (light pressure). Therefore the mechanical forces coincide with Coulomb's law for a certain effective charge that depends on the pump field.[17]

It is interesting to note that renormalization of atomic quantities is determined not by the interaction potential of the particles, but by the force. This situation differs from the pair-collision theory, in which the change of the spectral characteristics is expressed in terms of the interaction potential. It is known that the interaction of atoms in the case of short-range forces leads to a local change of the width and shift of the line; in this case, owing to the short particle interaction time in the collision, the coupling constant with the external field remains unchanged. In our case the interaction of the atoms is long-lasting and it must be taken into account in a self-consistent manner. As a result the time of correlation of the atoms with one another becomes long, and this manifests itself, in particular, in a change of the coupling constant with changing field. In addition, the renormalized quantities are nonlocal.

In connection with the change of the coupling constant, a comparison with the Lorentz correction to the mean

field is of interest. Owing to the singularity of the dipole interaction, the Lorentz correction to the mean field leads to a local change of the coupling constant. Accordingly, the dielectric constant becomes also a local quantity that is nonlinear in the density. It can be stated that (29) is the analog of the Lorenz-Lorentz formula, represented in the form of an expansion in the atom density. The main difference is that the increments nonlinear in the density are nonlocal and proportional to the mean-field intensity. The dependence of the optical properties of the medium on the size of the region of interaction with the field can lead to different physical consequences. The simplest example of this kind can be the dependence of the coefficient of absorption of a light beam on its diameter. According to (39), when the transverse dimension increases (i.e., when the diaphragm that limits the beam is increased), the transparency of the medium increases and the absorption coefficient decreases.

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[1)]It is understood here that a vector column does not go over into a vector row.

[2)]The reason why the numerical coefficient in the formula for γ differs from 4/3 is that no account is taken of the polarization and of the angular distribution of the spontaneous emission.

[3)]We note that for immobile atoms it is necessary to use two-time correlation functions.[9,10]

[4)]Small terms of the order of $1/N$ (N is the number of particles), which ensure conservation of the normalization,[13] have been left out of the reducible part.

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