ERRATA

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Erratum: Coherent scattering of electromagnetic radiation by a system of polarized particles [JETP 82 (4), 647–655 (1996)]

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A theory of coherent scattering of electromagnetic waves by a system of polarized atoms or molecules is developed. General expressions for the differential and total cross sections of the process are obtained. It is shown that only the orientation and alignment of the polarized ensemble can be revealed in coherent scattering. Properties of the angular distribution and the polarization of the scattered radiation, as well as circular dichroism, are discussed. Effects induced by dissipative processes are analyzed. © *1996 American Institute of Physics.* [S1063-7761(96)00904-3]

1. INTRODUCTION

Unshifted (Rayleigh) scattering of electromagnetic radiation, in which the quantum state of the scattering particle does not change, is coherent. This means that in the scattering of light[1)] by a system of N identical particles (atoms, molecules, etc.) occupying a region of space whose linear dimensions are small in comparison with the wavelength of the scattered radiation, the scattering amplitudes due to all the particles add together, and as a result the intensity of scattering of the entire system is proportional to N^2 (see Ref. 1, Sec. 59).

Recall that for coherence effects to appear the indicated spatial localization of the ensemble of scattering particles is of fundamental importance. Thus, if the distance between the particles (in what follows, for definiteness we will speak of atoms) significantly exceeds the wavelength of the scattered radiation, then interference of the scattering amplitudes of individual atoms can be neglected, and coherence effects are destroyed. In this case the scattering cross section of the entire system is obtained by multiplying the Rayleigh scattering cross section of a single atom by N (incoherent scattering). On the contrary, in a dense, unbounded medium separate small regions scatter light coherently; however, the total intensity of coherent scattering at all angles except zero vanishes (see, for example, Ref. 2, p. 19). The remaining coherent forward scattering is none other than the propagation of light in the medium, beautifully described by the equations of macroscopic electrodynamics. Weak Rayleigh scattering in the medium occurs due to various fluctuations (density, anisotropy, etc.).[2,3]

In the present paper we develop the theory of coherent scattering of electromagnetic radiation of a system of polarized atomic particles, i.e., we assume that an atom (molecule) of the medium has nonzero angular momentum j and the ensemble is nonequilibrium, so that states with different values of the projections m of the angular momentum on some direction are nonuniformly populated. General results for Raman and incoherent Rayleigh scattering by polarized atoms were obtained in our previous paper (Ref. 4). Recently, a study of Rayleigh scattering in a dense polarized gas[5] was carried out (scattering from fluctuations), generalizing the results of the theory of Rayleigh scattering in equilibrium media[3] to the case of gaseous media with polarized particles.

Note that coherent scattering by freely oriented (unpolarized) atoms does not require a special treatment. The cross section of incoherent scattering by freely oriented systems, as was shown by Placzek, divides into three independent parts: scalar, antisymmetric, and symmetric (see, for example, Ref. 1, Sec. 60). The cross section of coherent scattering contains only a scalar part.[2)] However, the cross section of coherent light scattering by polarized particles differs substantially from the cross section of incoherent scattering given in Ref. 4. In this paper, we succeeded in obtaining a compact expression for the cross section which makes it easy to analyze various effects associated with coherent scattering of electromagnetic radiation by a system of polarized particles. This latter circumstance is of no small importance for the solution of various spectroscopic problems.

The paper is organized as follows. A basic expression for the cross section of coherent light scattering by a polarized ensemble is obtained in Sec. 2. In Sec. 3, using the algebra of angular momentum, the cross section is separated into its angular parts and the result is presented in a compact form suitable for analysis. Section 4 discusses different new effects that can be observed in association with coherent scattering. Section 5, the Conclusion, throws out some ideas about possible experimental studies of the coherent scattering process.

2. SPECIFICATION OF THE INITIAL STATE OF THE POLARIZED ATOM. CROSS SECTION OF COHERENT SCATTERING

We assume that the state of a polarized atom having total angular momentum $j \neq 0$ is an incoherent mixture of states with different values of the projection m of the angular momentum onto some direction $\mathbf{n}$. In this case the density matrix of the atom is diagonal in m in the atomic coordinate system with axis z_a aligned with $\mathbf{n}$. As is well known, such polarization states arise if the external polarizing action is axially symmetric relative to $\mathbf{n}$.[6]

We will specify the state of the polarized atom not by $2j$ independent populations of the magnetic sublevels—the diagonal elements of the density matrix in the atomic coordinate system ρ^n_{mm}, but by the irreducible components of the density matrix, called the state multipoles:[6]

$$\rho^n_K=\sum_m (-1)^{j-m}(2K+1)^{1/2}\begin{pmatrix} j & j & K \\ m & -m & 0 \end{pmatrix}\rho^n_{mm}.$$

Equal populations of the magnetic sublevels are associated with an unpolarized atom, and in this case

$$\rho^n_K=(2j+1)^{-1/2}\delta_{K,0}. \tag{1}$$

In general, we have $2j+1$ various multipoles of state:

$$\rho^n_0=(2j+1)^{-1/2}, \quad \rho^n_1=3^{1/2}[j(j+1)(2j+1)]^{-1/2}\overline{m},$$

$$\rho^n_2=5^{1/2}[j(j+1)(2j-1)(2j+1)(2j+3)]^{-1/2}$$

$$\times[3\overline{m^2}-j(j+1)],\ldots,$$

$$\rho^n_{K>2j}=0,$$

where the bar denotes the statistical average, thus:

$$\overline{f}=\sum_m f_m\rho^n_{mm}.$$

As will be shown below, under conditions in which the dipole approximation applies, only the orientation ρ^n_1 and the alignment ρ^n_2 can be revealed in coherent scattering. Recall that in mirror-symmetric polarization states, when the magnetic sublevels with opposite values of the projection of the angular momentum are equally populated ($\rho^n_{mm}=\rho^n_{-m,-m}$), the orientation and all remaining multipoles of odd rank vanish.

Let us obtain the basic expression for the cross section of coherent light scattering by an ensemble of polarized particles. To start with, we will use the atomic coordinate system and denote by $d\sigma_{m'm}/d\Omega'$ the Rayleigh scattering cross section of an atom accompanied by the change in the projection of the angular momentum $m\rightarrow m'$. The cross section is given in the dipole approximation by the well-known Kramers–Heisenberg formula (Ref. 1, Sec. 59) (the atomic system of units is used)

$$\frac{d\sigma_{m'm}}{d\Omega'}=(\alpha\omega)^4|\langle\nu jm'|c_{ik}|\nu jm\rangle e'^*_i e_k|^2. \tag{2}$$

Here ω is the frequency of the electromagnetic radiation, α is the fine structure constant, $\mathbf{e}$ and $\mathbf{e}'$ are the unit polarization vectors of the incident and the scattered photon, and ν denotes the remaining set of atomic quantum numbers besides the angular momentum j and its projection m. Summation is assumed over the repeated Cartesian indices i and k. The operator c_{ik}, which we referred to in Ref. 4 as the scattering tensor, has the form

$$c_{ik}=d_i\hat{G}_{E_1+\omega}d_k+d_k\hat{G}_{E_1-\omega}d_i, \tag{3}$$

where d_i is the Cartesian component of the dipole moment operator of the atom, E_1 is the energy of its initial state, which does not change as a result of Rayleigh scattering,

$$\hat{G}_E=\sum_n \frac{|n\rangle\langle n|}{E_n-E-i0}$$

is the atomic Green's function (the summation is carried out over all possible states $|n\rangle$, including the continuum).

Let N_m particles, in an atomic ensemble consisting of N particles, have the projection of their angular momentum on the z_a axis of the atomic coordinate system equal to m. Obviously

$$N_m=\rho^n_{mm}N.$$

Here for simplicity we assume that all the particles of the ensemble have the same angular momentum j; however, this restriction is easily removed (see the clarifications of formula (12) in Sec. 3). If the ensemble scatters light incoherently (the distance between particles is significantly greater than a wavelength), then the scattering cross section of the entire ensemble is given by

$$\frac{d\Sigma^{(ic)}}{d\Omega'}=\sum_{m,m'} N_m\frac{d\sigma_{m'm}}{d\Omega'}=N\frac{d\sigma^{(ic)}}{d\Omega'},$$

where

$$\frac{d\sigma^{(ic)}}{d\Omega'}=\sum_{m,m'}\rho^n_{mm}\frac{d\sigma_{m'm}}{d\Omega'}$$

is the cross section of incoherent Rayleigh scattering by a polarized atom, whose structure was investigated in Ref. 4. Noting that the state of the atom does not change during the scattering process and the scattering amplitudes of each particle of the ensemble add if the scattering is coherent, we have [see formula (2)][3)]

$$\frac{d\Sigma^{(c)}}{d\Omega'}=\left|\sum_m N_m(\alpha\omega)^2\langle\nu jm|c_{ik}|\nu jm\rangle e'^*_i e_k\right|^2=N^2\frac{d\sigma^{(c)}}{d\Omega'} \tag{4}$$

where it is natural to call

$$\frac{d\sigma^{(c)}}{d\Omega'}=(\alpha\omega)^4\left|\sum_m \rho^n_{mm}\langle\nu jm|c_{ik}|\nu jm\rangle e'^*_i e_k\right|^2 \tag{5}$$

the cross section of coherent scattering. We have intentionally carried out the derivation of expression (5) in such elementary detail because it was specifically at this point in the treatment of coherent light scattering by an unpolarized ensemble that an error was made in Sec. 60 of Ref. 1, which led to the conclusion that the cross section of coherent scattering by a freely oriented system contains an antisymmetric and a symmetric part as well as a scalar part.

To wrap up this section, we write out the expression for the cross section of coherent light scattering (5) in an arbitrary coordinate system (in invariant form). We denote the projection of the angular momentum of the atom onto the z axis of this coordinate system by M, and the elements of the density matrix by $\langle\nu jM_1|\hat{\rho}|\nu jM_2\rangle$. Expanding the states

$|\nu jm\rangle$ in Eq. (5) over the states $|\nu jM\rangle$ and noting that the density operator of the polarized atom has the form

$$\hat{\rho}=\sum_m \rho^n_{mm}|\nu jm\rangle\langle \nu jm|$$

$$=\sum_{M_1,M_2}\langle \nu jM_1|\hat{\rho}|\nu jM_2\rangle|\nu jM_1\rangle\langle \nu jM_2|,$$

we easily find the invariant expression for the cross section:

$$\frac{d\sigma^{(c)}}{d\Omega'}=(\alpha\omega)^4\left|\sum_{M_1,M_2}\langle \nu jM_1|\hat{\rho}|\nu jM_2\rangle \times\langle \nu jM_2|c_{ik}|\nu jM_1\rangle e_i'^{*}e_k\right|^2. \quad (6)$$

3. SEPARATION OF THE GEOMETRIC AND DYNAMIC PARTS. DIFFERENTIAL AND TOTAL SCATTERING CROSS SECTIONS

To carry out the sum over the projections of the angular momenta in formula (6) and delineate its dependence on the various angular coordinates of the problem, we use the standard techniques of the angular momentum algebra and the method developed in Ref. 4. We define the irreducible components of the scattering tensor c_{ik} (3) by the relation

$$t_{kq}=\sum_{q_1,q_2}C^{kq}_{1q_1 1q_2}c_{q_1q_2}=\sum_{q_1,q_2}C^{kq}_{1q_1 1q_2}d_{q_1}[\hat{G}_{E_1+\omega}+(-1)^k\hat{G}_{E_1-\omega}]d_{q_2}, \quad (7)$$

where $C^{kq}_{1q_1 1q_2}$ is the Clebsch–Gordan coefficient, and the irreducible tensors composed from the polarization vectors are equal to

$$\{\mathbf{e}'^{*}\otimes\mathbf{e}\}_{kq}=\sum_{q_1,q_2}C^{kq}_{1q_1 1q_2}e'^{*}_{q_1}e_{q_2}. \quad (8)$$

By the vector components in formulas (7) and (8) we mean the corresponding spherical components:

$$a_0=a_z,\quad a_{\pm1}=\mp 2^{-1/2}(a_x\pm ia_y).$$

Correspondingly, $c_{q_1q_2}$ is the spherical component of the scattering tensor, defined by Eq. (3), where the Cartesian components of the dipole moment have been replaced by the corresponding spherical components. The scalar product of the scattering tensor c_{ik} and the tensor $e_i'^{*}e_k$ is expressed in terms of the corresponding irreducible tensors (see Ref. 4) as

$$c_{ik}e_i'^{*}e_k=\sum_{k,q}(-1)^{k-q}t_{kq}\{\mathbf{e}'^{*}\otimes\mathbf{e}\}_{k,-q}. \quad (9)$$

In the matrix elements of the irreducible parts of the scattering tensor (7) the dependence on the magnetic quantum numbers is extracted with the help of the Wigner–Eckart theorem. Using the abbreviated notation

$$T_k\equiv\langle \nu j\|t_k\|\nu j\rangle,\quad k=0,1,2,$$

for the irreducible matrix elements of the scattering tensor arising in the problem of coherent scattering, we have

$$\langle \nu jM_2|t_{kq}|\nu jM_1\rangle=(-1)^{j-M_2}\begin{pmatrix} j & k & j\\ -M_2 & q & M_1\end{pmatrix}T_k. \quad (10)$$

In the elements of the density matrix of the polarized atom it is also easy to extract the dependence on the magnetic quantum numbers and on the orientation of the polarization axis $\mathbf{n}$. Toward this end, invoking the law of transformation of the state multipoles under rotations, as in Ref. 4 we represent the density matrix in the form

$$\langle \nu jM_1|\hat{\rho}|\nu jM_2\rangle=\sqrt{4\pi}\sum_{K,Q}(-1)^{j-M_1}\times\begin{pmatrix} j & j & K\\ M_1 & -M_2 & -Q\end{pmatrix}Y^{*}_{KQ}(\mathbf{n})\rho^n_K, \quad (11)$$

where $Y_{KQ}(\mathbf{n})$ is the spherical harmonic of order K and index Q. Note that expression (11) is valid only for the axially symmetric polarization states considered here. For nonaxially symmetric polarization the state of the atom is a coherent superposition of states with different m, and its density matrix turns out to be nondiagonal in m even in the initial (atomic) coordinate system imposed by the external polarizing action. In this case, the spherical harmonic of $\mathbf{n}$ in formula (11) and below in formula (12) for the cross section is replaced by a linear combination of Wigner functions which will depend on the three Euler angles defining the orientation of the atomic coordinate system.

Substituting expressions (9)–(11) in expression (6) and, with the help of the orthogonality condition for the 3j-symbols, getting rid of the sum over magnetic quantum numbers, we obtain the following expression for the differential cross section of coherent scattering:

$$\frac{d\sigma^{(c)}}{d\Omega'}=4\pi(\alpha\omega)^4\left|\sum_{K,Q}(-1)^K(2K+1)^{-1}\rho^n_K T_K Y^{*}_{KQ}(\mathbf{n})\times\{\mathbf{e}'^{*}\otimes\mathbf{e}\}_{KQ}\right|^2. \quad (12)$$

Obviously, the index K in the sum in Eq. (12) can take only three values: $K=0,1,2$. This is a consequence of the fact that the expansion of a Cartesian tensor of second rank into irreducible parts includes only irreducible tensors of zeroth, first, and second rank [see Eqs. (7) and (8)]. Consequently, only the orientation and the alignment of the polarized system can be revealed in coherent light scattering,[4)] this result being in no way connected with the axial symmetry of the polarization state.

Above for simplicity we assumed that all the particles of a polarized ensemble have the same total angular momentum. However, all of our results here easily generalize to the case where the ensemble is an incoherent mixture of states with different j. Indeed, let W_j be the population of a level with some fixed j so that

$$\sum_m \rho^n_{mm}(j)=W_j,\quad \sum_j W_j=1.$$

Then in the basic expression for the coherent scattering cross section (5) we must replace ρ^n_{mm} by $\rho^n_{mm}(j)$ and sum over j. Notice that the original matrix ρ^n_{mm} satisfies the simpler normalization condition

$$\sum_m \rho^n_{mm}=1.$$

As a result, in formula (12) and the other formulas which we derive below, we must make the following substitution:

$$\rho^n_K T_K \rightarrow \sum_j W_j \rho^n_K T_K .$$

The coherent scattering cross section (12) has a significantly simpler structure than the incoherent scattering cross section of a polarized particle.[4] In the case under consideration of axially symmetric systems the coherent scattering cross section can be written in a still simpler form containing ordinary scalar and vector products of vectors. Toward this end, we express the spherical harmonic functions $Y_{1Q}(\mathbf{n})$ and $Y_{2Q}(\mathbf{n})$ in terms of irreducible tensors made up from the vector **n**:

$$Y_{1Q}(\mathbf{n})=\sqrt{\frac{3}{4\pi}}n_Q, \quad Y_{2Q}(\mathbf{n})=\sqrt{\frac{15}{8\pi}}\{\mathbf{n}\otimes\mathbf{n}\}_{2Q}.$$

We write the scalar constructions entering into expression (12) in the following form (the corresponding formulas can be found, for example, in Ref. 7):

$$\{\mathbf{e}'^*\otimes\mathbf{e}\}_{00}=\sqrt{-\frac{1}{3}}\,\mathbf{e}'^*\mathbf{e}, \quad \sum_Q Y^*_{1Q}(\mathbf{n})\{\mathbf{e}'^*\otimes\mathbf{e}\}_{1Q}$$
$$=i\sqrt{\frac{3}{8\pi}}[\mathbf{e}'^*\mathbf{e}]\mathbf{n}, \tag{13}$$

$$\sum_Q Y^*_{2Q}(\mathbf{n})\{\mathbf{e}'^*\otimes\mathbf{e}\}_{2Q}=\sqrt{\frac{15}{8\pi}}\left[(\mathbf{e}'^*\mathbf{n})(\mathbf{e}\mathbf{n})-\frac{1}{3}\mathbf{e}'^*\mathbf{e}\right].$$

After substituting relations (13) into formula (12), expanding the square of the absolute value and grouping all vector combinations, we can represent the differential cross section of coherent scattering by a system of polarized particles in the following final form:

$$\frac{d\sigma^{(c)}}{d\Omega'}=\frac{1}{3}(\alpha\omega)^4\Big\{a|\mathbf{e}'^*\mathbf{e}|^2+b_1|\mathbf{e}'\mathbf{n}|^2|\mathbf{e}\mathbf{n}|^2+b_2\,\mathrm{Re}[(\mathbf{e}'^*\mathbf{e})$$
$$\times(\mathbf{e}'\mathbf{n})(\mathbf{e}^*\mathbf{n})]+b_3[\xi_2\mathbf{k}\,\mathrm{Re}([\mathbf{n}\mathbf{e}'](\mathbf{n}\mathbf{e}'^*))$$
$$-\xi_2'\mathbf{k}'\,\mathrm{Re}([\mathbf{n}\mathbf{e}](\mathbf{n}\mathbf{e}^*))]+c_1|\mathbf{n}[\mathbf{e}'^*\mathbf{e}]|^2+c_{2+}$$
$$\times[\xi_2\,\mathrm{Re}((\mathbf{k}\mathbf{e}'^*)(\mathbf{n}\mathbf{e}'))+\xi_2'$$
$$\times\mathrm{Re}((\mathbf{k}'\mathbf{e}^*)(\mathbf{n}\mathbf{e}))]+\frac{1}{2}c_{2-}(\xi_2\mathbf{k}\mathbf{n}+\xi_2'\mathbf{k}'\mathbf{n})$$
$$+c_3\mathbf{n}\,\mathrm{Re}([\mathbf{e}'\mathbf{e}^*](\mathbf{e}'^*\mathbf{e})]+c_4[\xi_2(\mathbf{k}\mathbf{n})|\mathbf{e}'\mathbf{n}|^2$$
$$+\xi_2'(\mathbf{k}'\mathbf{n})|\mathbf{e}\mathbf{n}|^2]+c_5\mathbf{n}\,\mathrm{Re}[[\mathbf{e}'\mathbf{e}^*](\mathbf{e}'^*\mathbf{n})(\mathbf{e}\mathbf{n})]\Big\}. \tag{14}$$

Here **k** and **k**$'$ are unit vectors indicating the directions of propagation of the incident and scattered radiation, and

$$\xi_2=i\mathbf{k}[\mathbf{e}\mathbf{e}^*] \tag{15}$$

is the degree of circular polarization of the incident light, equal to ± 1 for right (left) circular polarization and zero for linear polarization, and

$$\xi_2'=i\mathbf{k}'[\mathbf{e}'\mathbf{e}'^*]. \tag{16}$$

The coefficients a, b_i, and c_i can be expressed in terms of the reduced matrix elements of the scattering tensor T_k [see Eq. (10)][5)] and state multipoles $\rho^n_0=(2j+1)^{-1/2}$, ρ^n_1, and ρ^n_2:

$$a=\frac{|T_0|^2}{2j+1}+\frac{1}{10}(\rho^n_2)^2|T_2|^2+\sqrt{\frac{2}{5(2j+1)}}\rho^n_2\mathrm{Re}(T_0T_2^*),$$

$$b_1=\frac{9}{10}(\rho^n_2)^2|T_2|^2, \quad b_2=-\frac{3}{5}\rho^n_2\left[\rho^n_2|T_2|^2+\sqrt{\frac{10}{2j+1}}\,\mathrm{Re}(T_0T_2^*)\right],$$

$$b_3=\frac{3}{\sqrt{10(2j+1)}}\rho^n_2\,\mathrm{Im}(T_0T_2^*), \quad c_1=\frac{1}{2}(\rho^n_1)^2|T_1|^2, \tag{17}$$

$$c_{2\pm}=\pm\rho^n_1\left[\frac{1}{\sqrt{2(2j+1)}}\,\mathrm{Re}(T_0T_1^*)\mp)\frac{1}{\sqrt{5}}\rho^n_2\,\mathrm{Re}(T_2T_1^*)\right],$$

$$c_3=\rho^n_1\left[\sqrt{\frac{2}{2j+1}}\,\mathrm{Im}(T_0T_1^*)+\frac{1}{\sqrt{5}}\rho^n_2\,\mathrm{Im}(T_2T_1^*)\right],$$

$$c_4=\frac{3}{2\sqrt{5}}\rho^n_1\rho^n_2\,\mathrm{Re}(T_2T_1^*), \quad c_5=\frac{3}{\sqrt{5}}\rho^n_1\rho^n_2\,\mathrm{Im}(T_1T_2^*).$$

For an unpolarized system ($\rho^n_1=\rho^n_2=0$) only the coefficient a is nonzero. If the system is aligned but not oriented ($\rho^n_2 \neq 0, \rho^n_1=0$), then the coefficients c_i vanish, and if the system is oriented but not aligned ($\rho^n_1 \neq 0$, $\rho^n_2=0$), then the coefficients b_i, c_4, and c_5 vanish. The expression for the differential cross section of coherent scattering (14) will be analyzed in more detail in the next section of this paper.

The total cross section is easily obtained from the differential cross section (14) by integrating over all scattering directions and taking into account the well-known relations

$$\int \mathbf{k}'d\Omega'=0, \quad \int e_i'e_k'^*d\Omega'=\frac{4\pi}{3}\delta_{ik}.$$

As a result, some of the vector combinations vanish and we obtain

$$\sigma^{(c)}=\frac{4\pi}{9}(\alpha\omega)^4(A+B|\mathbf{e}\mathbf{n}|^2+C\xi_2\mathbf{k}\mathbf{n}), \tag{18}$$

where the parameters A, B, and C can be expressed in terms of the coefficients (17):

$$A=a+c_1=\frac{|T_0|^2}{2j+1}+\frac{1}{10}(\rho_2^n)^2|T_2|^2$$

$$+\sqrt{\frac{2}{5(2j+1)}}\rho_2^n \operatorname{Re}(T_0T_2^*)+\frac{1}{2}(\rho_1^n)^2|T_1|^2,$$

$$B=b_1+b_2-c_1=\frac{3}{10}(\rho_2^n)^2|T_2|^2$$

$$-3\sqrt{\frac{2}{5(2j+1)}}\rho_2^n \operatorname{Re}(T_0T_2^*)-\frac{1}{2}(\rho_1^n)^2|T_1|^2,$$

$$C=c_{2+}+\frac{3}{2}c_{2-}+c_4=-\rho_1^n\left[\sqrt{\frac{2}{2j+1}}\operatorname{Re}(T_0T_1^*)\right.$$

$$\left.+\frac{1}{\sqrt{5}}\rho_2^n \operatorname{Re}(T_2T_1^*)\right].$$

Summation over the two independent polarizations of the scattered photon reduces to multiplying the total cross section (18) by two.

If the polarization of the scattered photon is not measured and we are interested only in the angular distribution of the scattered radiation, then we need to sum the differential cross section (14) over the two independent polarizations. This is easily done with the help of the well-known identity (Ref. 1, Sec. 45)

$$\sum_{\lambda}(\mathbf{a}\mathbf{e}_\lambda')(\mathbf{b}\mathbf{e}_\lambda'^*)=[\mathbf{k}'\mathbf{a}][\mathbf{k}'\mathbf{b}]. \tag{19}$$

Summing the parameter ξ_2' (16) over the polarizations obviously gives zero since it is equal to ± 1 for right (left) circular polarization.

With the help of an identity analogous to (19) it is also easy to average the differential cross section (14) and the total cross section (18) for coherent scattering over the polarizations of the incident photon, i.e., to go over to the case of scattering of unpolarized light.

Finally, we will show how to obtain an expression for the scattering cross section in the most general case of partial polarization of the incident radiation. The state of partial polarization is specified by the polarization density matrix, which in the basis of states with definite helicity (the basis vectors here are the unit vectors of right and left circular polarization of the incident radiation) is expressed in the standard way in terms of the Stokes parameters η_i:[6]

$$\begin{pmatrix}\rho^\gamma_{+1,+1} & \rho^\gamma_{+1,-1}\\ \rho^\gamma_{-1,+1} & \rho^\gamma_{-1,-1}\end{pmatrix}=\frac{1}{2}\begin{pmatrix}1+\eta_2 & -\eta_3+i\eta_1\\ -\eta_3-i\eta_1 & 1-\eta_2\end{pmatrix}. \tag{20}$$

Recall that the parameter η_2 specifies the degree of circular polarization, η_3 is the degree of linear polarization relative to the x and y axes of the coordinate system whose z axis points in the direction of propagation of the incident light (below we will call this the laboratory coordinate system), and η_1 is the degree of linear polarization relative to two mutually perpendicular axes rotated by an angle of 45° relative to the x and y axes of the laboratory coordinate system. The values of the Stokes parameters $\eta_1=\eta_2=\eta_3=0$ correspond to unpolarized light. In the general case $\Sigma_i\eta_i^2\leq 1$ holds, whereas in the state of pure polarization prescribed by the vector $\mathbf{e}$ (formulas (14) and (18) correspond to this case) we have $\Sigma_i\eta_i^2=1$ and $\eta_2=\xi_2$ (15).

Let $\mathbf{e}_\pm$ be the unit vectors of right (left) circular polarization of the incident photon which are expressed as follows in terms of the first two basis vectors of the laboratory coordinate system:

$$\mathbf{e}_{\pm 1}=\mp\frac{1}{\sqrt{2}}(\mathbf{e}_x\pm i\mathbf{e}_y).$$

To make the transition to the case of partially polarized incident radiation, it is necessary to average cross sections (14) and (18) over the two statistically-independent elliptical-polarization states (the eigenvectors of the polarization density matrix), whose incoherent superposition with statistical weights equal to the eigenvalues of the density matrix (20) constitutes partially polarized light. Such an operation is equivalent to the formal substitution

$$e_ie_k^*\rightarrow\sum_{\lambda,\lambda'}\rho^\gamma_{\lambda,\lambda'}e_{\lambda,i}e^*_{\lambda',k}$$

in expressions (14) and (18). Here ξ_2 given by (15) is replaced by η_2.

4. DISCUSSION OF RESULTS

Let us consider what effects polarization of the particles produces in coherent scattering. Obviously, the ensemble of polarized particles becomes asymmetric, which is expressed by the substantial change both in the angular distribution and in the polarization properties of the scattered radiation while the total cross section continues to depend on the direction of propagation and the polarization of the incident radiation. The new, experimentally observable effects arising here can be analyzed with the help of formulas (14) and (18). Before discussing some of them, note once more that due to the difference in the physics of the two scattering processes the structure of the cross section for coherent scattering differs substantially from the structure of the cross section for incoherent scattering by polarized particles.[4] As a consequence of this difference, only the orientation and alignment (ρ_1^n and ρ_2^n) of the polarized ensemble can be revealed in incoherent scattering. In addition, whereas the state multipoles enter linearly in the incoherent scattering cross section, the coherent scattering cross section also contains the product $\rho_K^n\rho_{K'}^n$. This latter circumstance leads to a number of qualitatively new effects. Thus, for example, the total cross section for coherent scattering of linearly polarized light ($\xi_2=0$) by an oriented but unaligned system ($\rho_1^n\neq 0$, $\rho_2^n=0$) continues to depend on the geometry as well as the orientation ρ_1^n of the system [see the second term in formula (18)]. Under these conditions the total cross section for incoherent scattering coincides with the cross section for scattering by an unpolarized particle[4] (the quantity ρ_1^n is proportional to the projection of a pseudovector, the average angular momentum, onto the vector $\mathbf{n}$ and cannot enter into the cross section linearly if the pseudoscalar ξ_2 is equal to zero).

4.1 Coherent scattering by a system of unpolarized particles

Substituting $\rho_1^n=\rho_2^n=0$ in formulas (17) and then substituting these results in the general expression (14), we obtain the differential cross section for coherent scattering by a system of unpolarized particles

$$\frac{d\sigma_{\mathrm{unp}}^{(c)}}{d\Omega'}=(\alpha\omega)^4\frac{|T_0|^2}{3(2j+1)}|\mathbf{e}'^*\mathbf{e}|^2, \tag{21}$$

which coincides with the scalar part of incoherent Rayleigh scattering by an unpolarized particle.[4] The total cross section in this case, naturally, does not depend on the parameters of the incident radiation:

$$\sigma_{\mathrm{unp}}^{(c)}=\frac{4\pi}{9}(\alpha\omega)^4\frac{|T_0|^2}{2j+1}.$$

Thus, the differential cross section for coherent scattering of light by a system of freely oriented (unpolarized) particles does not contain either an antisymmetric or a symmetric part, in contradiction to what is stated in Sec. 60 of Ref. 1 and, what is more, exactly coincides with the scalar cross section for incoherent scattering rather than differing from it by a factor of $2j+1$.

The absence of an antisymmetric and a symmetric part in the coherent scattering cross section of a system of unpolarized particles follows entirely from formula (5): in the average of the polarizability (the matrix element of the scattering tensor) over the orientations of a spherically symmetric system of particles, only the scalar part should remain. This result can be obtained from other, purely physical considerations. As was mentioned in the Introduction, the propagation of light in an unbounded medium from the microscopic point of view is simply its coherent forward scattering. But it is not hard to show that only for scalar forward scattering is the polarization of the light unchanged. Consequently, the presence of nonscalar parts in the cross section of coherent scattering by a system of unpolarized particles means that when light propagates in an isotropic medium its polarization changes.

Also note that the scattering of electromagnetic waves by macroscopic particles whose linear dimensions are small in comparison with the wavelength, first considered by Rayleigh (see Refs. 2 and 3), from a microscopic point of view is another example of coherent scattering. If with the help of identity (19) we sum cross section (21) over the polarizations of the scattered light and substitute it in Eq. (4), then we actually, by way of a microscopic derivation, reproduce the Rayleigh formula (in which we need to note that the particles are the particles of a gas, i.e., their dielectric constant is close to unity). In this regard we point out that our basic expression (4) could in general have been derived on the basis of considerations from classical electrodynamics similar to Rayleigh's arguments. In this case, however, we would have obtained a formula for the scattering intensity in a given direction, already summed over the polarizations of the scattered light and therefore less informative.

4.2 Structure of the differential cross section of coherent scattering. Effects induced by processes of dissipation of light energy

The expressions for the coherent scattering cross sections (14) and (18) contain the state multipoles ρ_1^n and ρ_2^n, defined in the atomic coordinate system with its quantization axis z_a aligned with the vector $\mathbf{n}$ (see Sec. 2). The orientation ρ_1^n is proportional to the average projection of the angular momentum (pseudoscalar) onto the vector $\mathbf{n}$ and, consequently, is a pseudoscalar. In addition, this pseudoscalar, as is obvious, is T-odd, i.e., it changes sign upon time reversal. At the same time, the alignment ρ_2^n is a T-even scalar. With this in mind, let us discuss the structure of the individual terms in the differential cross section of coherent scattering (14). Here for brevity we will refer to each term by its coefficient (e.g., the b_1 term, etc.).

The b_i coefficients do not depend on the orientation ρ_1^n [see formulas (17)]. The b_1-term, which is quadratic in the alignment ρ_2^n, cannot enter into the cross section of incoherent scattering by aligned systems, whereas the b_2- and b_3-term are linear in ρ_2^n so that the vector combinations entering into them should also show up in the incoherent scattering cross section.

Of especial interest is the b_3-term, containing T-odd vector combinations (these combinations are also pseudoscalars, but their pseudoscalarity is compensated by the pseudoscalars ξ_2 and ξ_2'). Since the scattering cross section is T-even, the coefficient b_3 should also be proportional to a T-odd parameter. The quantity $\mathrm{Im}(T_0T_2^*)$ entering into b_3 (17) is T-odd. This quantity is equal to zero if the scattering tensor [see Eqs. (3) and (7)] is Hermitian and, from a formal point of view, becomes nonzero only for a non-Hermitian scattering tensor. The anti-Hermitian part of the scattering tensor is T-odd and nonzero both for resonance scattering due to the width of the resonant level and for above-threshold scattering (the photon energy exceeds the ionization threshold, or dissociation threshold for molecules) due to the non-Hermiticity in this case of the Green's function (resolvent) $\hat{G}_{E_1+\omega}$. Non-Hermiticity, in principle arising as a result of taking radiative corrections into account, can be neglected. Thus, the b_3-term in (14) is nonzero only when one of the light-energy dissipation channels is open: the photo-ionization channel or the photodissociation channel in the case of above-threshold scattering; and the radiative, collisional, or some other channel for resonance scattering. The role of the T-odd parameter in this case falls to a physical parameter determining the light-energy dissipation rate (the width of the resonant level, the probability of photoionization or photodissociation) which is proportional to the matrix element of the anti-Hermitian part of the scattering tensor. Note that the b_3 term determines a number of qualitatively new effects in the process of coherent scattering by aligned systems: circular dichroism, some polarization properties (see below). Therefore we can say that these effects are dissipation-induced. As was recently shown by Manakov,[9] under certain conditions dissipative processes lead to polarization anomalies in the incoherent scattering of light by unpolarized atoms.[6)] The influence of dissipative processes on the incoherent scattering of light by oriented atoms is discussed in Ref. 10.

The c_1 term in expression (14), quadratic in the orientation, cannot enter into the cross section for incoherent scattering. All the remaining c terms are linear in ρ_1^n. The vector combinations entering into the $c_{2\pm}$ terms and c_3 term appear in the cross section for incoherent scattering by oriented systems (see Ref. 10). The last two terms in (14) contain products of the orientation and the alignment and show up only in the cross section for coherent scattering.

Note also that the vector combinations in the $c_{2\pm}$ terms and c_4 term are T-odd; however, the overall T-evenness of these terms is ensured by the T-oddness of the orientation ρ_1^n. At the same time, the c_3 term and c_5 term contain T-even vector combinations; therefore they differ from zero due to dissipation effects (the coefficients c_3 and c_5 (17) contain the T-odd quantities $\mathrm{Im}(T_0 T_1^*)$ and $\mathrm{Im}(T_1 T_2^*)$) like the b_3-term.

Finally, we note that the b_3-term, c_3-term, and c_5-term in (14), which are proportional to the T-odd dissipative parameter, vanish when integrated over all scattering directions and do not appear in the total cross section (18).

4.3 Circular dichroism

By circular dichroism we mean the difference in the result that light of left or right circular polarization has on a physical system.

Circular dichroism in the total cross section of coherent scattering (18) is observed only in the case of an oriented ensemble, i.e., for $\rho_1^n \neq 0$ ($\xi_2 = \pm 1$ in (18) for right or left circular polarization of the incident radiation). The absence of circular dichroism in the total cross section of any elementary process in unoriented systems is a simple consequence of parity conservation (see the analogous discussions for incoherent scattering of light in Ref. 4 and for multiphoton ionization of polarized atoms in Refs. 11 and 12). This result is obvious: the pseudoscalar ξ_2 cannot appear in the total cross section if the orientation ρ_1^n (a pseudoscalar) is equal to zero.

At the same time, circular dichroism in the differential cross section can be observed for zero orientation. This statement is quite obvious since it is possible to construct a number of pseudoscalar combinations from the vectors entering into the expression for the differential cross section. Recall that if a photon with nonzero degree of circular polarization ξ_2' (16), e.g., a circularly polarized photon, is recorded in the scattering, i.e., the scattered light is passed through a polarization filter, then the effect of circular dichroism is observed already in the scattering of light by unpolarized systems. The term responsible for this effect, $\xi_2 \xi_2' (\mathbf{k}\mathbf{k}')$, can be extracted from $|\mathbf{e}'^* \mathbf{e}|^2$ in the differential cross section (21). If a linearly polarized photon is recorded, or polarization of the scattered light is generally not recorded, then circular dichroism is absent in the scattering of light by unpolarized systems.

Speaking of circular dichroism in the differential cross section of coherent scattering by polarized systems, we have in mind specifically these cases: scattering without measurement of polarization of light or scattering with measurement of linear polarization ($\mathbf{e}'$ is the real vector, $\xi_2' = 0$). Here the effect of circular dichroism is due to the b_3-term, the $c_{2\pm}$-terms, and the c_4-term in (14), which are proportional to the degree of circular polarization of the incident light.

The $c_{2\pm}$-terms and c_4-terms are proportional to the orientation ρ_1^n [see Eqs. (17)], and the effect of circular dichroism due to them is preserved even in the total cross section (18). As was discussed above, terms of $c_{2\pm}$ type are also present in the cross section for incoherent scattering by oriented systems,[10] and a c_4-term only appears in the coherent scattering cross section for $\rho_2^n \neq 0$.

If the ensemble of particles is unoriented, then the $c_{2\pm}$-terms and c_4-term in (14) will be absent, and the effect of circular dichroism in the differential cross section, determined by the b_3-term, will be due only to the alignment of the system. It was shown above that the coefficient b_3 in (14) is nonzero only if a light-energy dissipation channel is open. Consequently, circular dichroism in the differential cross section for light scattering by aligned systems is dissipation-induced.

4.4 Polarization properties of the scattered radiation

Expression (14) for the coherent scattering differential cross section determines the intensity of scattering in the direction $\mathbf{k}'$ with definite polarization $\mathbf{e}'$, so it contains complete information about the polarization state of the scattered light. The polarization state of the scattered light is determined by the polarization density matrix, which is written in terms of the Stokes parameters η_i' analogous to the polarization density matrix of the incident light (20). With the help of formula (14) we can determine the Stokes parameters of the scattered light for arbitrary polarization of the ensemble of particles and arbitrary polarization of the incident light.

By way of an example, let us restrict ourselves to the case of scattering of electromagnetic radiation by an aligned ensemble, for which ρ_1^n equals zero. As is well known, such a polarization state of the ensemble arises if the polarizing excitation is not only axially symmetric, but also mirror-symmetric (e.g., excitation by unpolarized or linearly polarized light). An interesting non-optical method of aligning diatomic molecules with supersonic expansion of a buffer gas was developed in Ref. 13.

Note also that in the coherent scattering of long-wavelength radiation, when the frequency ω is small in comparison with the characteristic atomic (molecular) frequencies, the orientation of the ensemble is not revealed, and for any type of polarization the ensemble behaves as if it were purely aligned. Indeed, the orientation ρ_1^n enters into the expression for the coherent scattering cross section in combination with the reduced matrix element T_1 of the scattering tensor [see Eqs. (12) and (17)]. However, T_1 is proportional to the vector polarizability and is negligibly small in the low-frequency limit. This result follows directly from formula (7), in which for $k=1$ the two Green's functions essentially cancel each other out.

Let us derive explicit expressions for the Stokes parameters of the scattered radiation η_i' when the incident radiation is linearly polarized ($\mathbf{e}$ is the real vector, $\xi_2=0$). From the general expression (14) with the help of the identity (19) we find the differential cross section for coherent scattering by an aligned ensemble summed over the polarizations of the scattered light:

$$\frac{d\sigma_{\Sigma}^{(c)}}{d\Omega'} \equiv \frac{1}{3}(\alpha\omega)^4 S = \frac{1}{3}(\alpha\omega)^4\{a[\mathbf{k}'\mathbf{e}]^2 + b_1(\mathbf{en})[\mathbf{k}'\mathbf{n}]^2 + b_2(\mathbf{en})([\mathbf{k}'\mathbf{e}][\mathbf{k}'\mathbf{n}])\}.$$

In our case the first three terms in expression (14) are identical for right and left circular polarizations of the scattered radiation, so only the fourth term contributes to the degree of circular polarization η_2', and

$$\eta_2' = 2b_3(\mathbf{k}'[\mathbf{en}])(\mathbf{en})S^{-1}. \tag{22}$$

To find the degrees of linear polarization η_1' and η_3' we must first obtain an expression for the cross section from (14) in which the linear polarization vector of the scattered photon $\mathbf{e}'$ makes some arbitrary angle β with the x' axis of the coordinate system of the scattered photon (the z' axis is aligned with the vector $\mathbf{k}'$). It is easy to obtain this expression in the laboratory coordinate system with z axis aligned with the vector $\mathbf{e}$ (see Ref. 4), and then rewrite it in invariant form. Then assigning β the values 0, $\pi/2$, $\pi/4$, and $3\pi/4$, we obtain expressions for η_1' and η_3' (the x' axis lies in the plane of the vectors $\mathbf{e}$ and $\mathbf{k}'$):

$$\eta_1' = (\mathbf{k}'[\mathbf{en}])(\mathbf{en})\left\{ b_2 + 2b_1 \frac{\mathbf{en}}{[\mathbf{k}'\mathbf{e}]^2}([\mathbf{k}'\mathbf{e}][\mathbf{k}'\mathbf{n}])\right\} S^{-1}, \tag{23}$$

$$\eta_3' = \left\{ a[\mathbf{k}'\mathbf{e}]^2 + b_1(\mathbf{en})^2\left[(\mathbf{k}'\mathbf{n}]^2 - 2\frac{(\mathbf{k}'[\mathbf{en}])^2}{[\mathbf{k}'\mathbf{n}]^2}\right] + b_2(\mathbf{en}) \times([\mathbf{k}'\mathbf{e}][\mathbf{k}'\mathbf{n}])\right\} S^{-1}. \tag{24}$$

It follows from simple physical arguments that if the incident light is completely polarized (i.e., it is determined by the polarization vector $\mathbf{e}$) then the coherently scattered light will also be completely polarized. Indeed, it is not a single atom that coherently scatters light, but the atomic ensemble as a whole. The initial state of the system "photon plus atomic ensemble" was pure. The final state of the atomic ensemble after coherent scattering coincides with the initial state, therefore the scattered photon should also be found in a pure quantum-mechanical state. In the case under consideration here this fact is verified directly: with the help of formulas (22), (23), and (24) it is easy to show that $\Sigma_i \eta_i'^2 = 1$. Thus, the scattered light, generally speaking, is elliptically polarized.

The degree of circular polarization η_2' (22) is determined by a T-odd vector combination, so for linear polarization of the incident light the degree of circular polarization of the light scattered by an aligned ensemble is dissipation-induced and like the coefficient b_3 is nonzero only for above-threshold or resonance scattering, as was discussed above. In all other frequency ranges the scattered light will be linearly polarized. In this case, for an unpolarized ensemble, when $\eta_1'=0$ and $\eta_3'=1$ [see Eqs. (23), (24) and (17)], the scattered light is polarized along the x' axis, and for $\rho_2^n \neq 0$ at an angle $\beta' = \arctan(\eta_1'/\eta_3')/2$ to the x' axis. Measuring the degree of circular polarization of the scattered radiation, it is possible according to Eq. (22) to determine the alignment ρ_2^n. If $\eta_2'=0$ holds, then the alignment can be determined from the degrees of linear polarization η_1' (23) and η_3' (24). In this case it is not even necessary to examine the properties of the angular distribution of the scattered radiation.

Note also that the degree of circular polarization of the light scattered by an aligned ensemble will be nonzero only if the dissipation channels are open in all cases when the incident light has zero circular polarization (e.g., for scattering of unpolarized light).

5. CONCLUSION

The results obtained in this work may be useful in the design of experiments on coherent scattering of electromagnetic waves, in their interpretation, and also in the development of a number of new spectroscopic methods.

It should be borne in mind that to observe coherent scattering of electromagnetic radiation in the optical range by polarized particles it is necessary to localize a large quantity of free atoms (molecules) in a volume whose linear dimensions are small in comparison with the wavelength. Recent theoretical papers[14,15] maintain that such localization can in principle be achieved by methods of optical control of the motion of the atoms. Speaking of the possibility of experimentally observing the effect of coherent light scattering, we note that when the ratio of the wavelength λ to the mean distance between the atoms is of the order of 10^3, for which it is possible to localize up to 10^9 atoms in a volume on the order of λ^3, the scattering intensity will be comparable with the intensity of incoherent scattering of light by a gas containing $\sim 10^{18}$ atoms and, for a density of $\sim 10^{18} \text{cm}^{-3}$, occupying a volume on the order of 1 cm^3.

If we consider coherent scattering of long-wavelength radiation, then problems with the localization of particles should not arise. It can be shown that in this case the scattering intensity is small since the cross section (5) is proportional to ω^4. However, it should not be forgotten that we are talking about coherent scattering, whose intensity (4) is proportional to N^2. For a fixed particle density, the total number of particles N grows like λ^3 as one increases the wavelength λ and, correspondingly, the localization region, so that the intensity of coherent scattering in this case grows like λ^2. It is precisely for this reason that one observes Rayleigh scattering of electromagnetic waves from small particles, at the basis of which lies coherent scattering from the atoms (molecules) that make them up. Only recall that, as was discussed in Sec. 4, when long-wavelength coherent scattering from atoms is observed, only their alignment can be revealed (if we are not talking about atoms polarized in Rydberg states), whereas in the case of polarized molecules their orientation can also be revealed if the radiation frequency ω is not small in comparison with the characteristic molecular vibrational–rotational frequencies.

Note, finally, that in Sec. 4 we discussed by no means all the effects that can be observed in the process of coherent scattering of electromagnetic radiation by an ensemble of polarized atoms. The general expressions for the cross sections (14) and (18) allow us in a comparatively simple way to analyze any experimental situation. We hope that this study of coherent scattering of electromagnetic radiation by polarized systems will attract the attention of experimentalists.

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1)The word "light" is used here and below simply for brevity. The discussion is not limited to scattering of electromagnetic radiation in the optical range, but also significantly greater wavelengths.

2)Section 60 of Ref. 1 makes an invalid assertion about the structure of the coherent scattering cross section of freely oriented systems (see below, Sec. 4).

3)Weak ($\propto N$) incoherent Rayleigh scattering, which is accompanied by a change in the projection of the angular momentum of the atom, can of course be neglected.

4)The differential cross section of incoherent scattering contains state multipoles up to fourth rank inclusively, but only the orientation and the alignment remain in the total cross section.[4]

5)The quantities T_0, T_1, and T_2 are proportional to the scalar, vector, and tensor parts of the dynamic polarizability, introduced in Ref. 8.

6)It can be shown that it should not be possible to observe these anomalies in coherent scattering.

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