Circular dichroism of luminous energy, induced by the dissipation in light scattering by aligned atoms

M. Ya. Agre

Voronezh State University, 394693 Voronezh, Russia
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A compact expression for the cross section of light scattering by aligned atomic systems is derived. It is shown that in above-threshold or resonant scattering, when the channel of luminous energy dissipation is open, circular dichroism effects can be observed in the angular distribution and the degree of polarization of the scattered light. In such cases circular polarization of the scattered light is also induced when the incident light has no circular polarization.
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1. INTRODUCTION

When the magnetic sublevels of an atom are unevenly populated, the atom becomes polarized. The elementary photoprocesses involving polarized atoms (emission and absorption of light, the photoeffect, etc.) have been thoroughly studied both theoretically and experimentally (see, e.g., Ref. 1, the literature cited therein, and also Refs. 2 and 3).

The general theory of light scattering by polarized atoms developed in a recent paper[4] does not touch on a number of subtle effects important in various applications. Among such effects are, for instance, the so-called dissipation-induced effects, which can be observed only when the channels of luminous energy dissipation are open in the light scattering process. The atom in this case is characterized by a T-odd (i.e., changing sign under time inversion) dissipation parameter Γ, which determines the dissipation rate.

The manifestation of dissipation-induced effects in the scattering of light by oriented atoms was discussed in Ref. 5. For such a case the differential scattering cross section acquires a T-odd scalar combination of vectors of the form

$$\mathbf{j}\ \mathrm{Re}\{[\mathbf{e}\mathbf{e}'^*](\mathbf{e}^*\mathbf{e}')\},$$

where $\mathbf{e}$ ($\mathbf{e}'$) is the unit polarization vector of the incident (scattered) light, and $\mathbf{j}$ is the average angular momentum of the oriented atom. This leads to a dependence of the angular distribution and the degree of linear polarization of the scattered light on the orientation of the atom even when linearly polarized or unpolarized light is scattered.

This paper studies the effect of luminous-energy dissipation on the scattering of light by aligned atoms. The reader will recall that a type of polarization such as that due to alignment is possible if the atomic angular momentum $j_1 > 1/2$. Below we examine axisymmetric polarization states with a symmetry axis specified by a unit vector $\mathbf{n}$. The state of the polarized atom in this case is an incoherent mixture of states with different values m of the angular momentum in the direction of $\mathbf{n}$, and such polarization emerges when the external polarization is axisymmetric.[1] If the polarization is also mirror symmetric (say, unpolarized or linearly polarized electromagnetic radiation), the magnetic sublevels with opposite values of the projection of angular momentum on $\mathbf{n}$ have equal populations, so that orientation and other types of polarization determined by the multipoles of a state of odd rank are absent (Ref. 1)[1)]. Here alignment proves to be the simplest possible type of polarization, and for $j_1 = 1$ and 3/2 there can be no other type of polarization.

The important role of dissipation of luminous energy in the effects of circular dichroism discussed below can be perceived from simple symmetry considerations. Let $\mathbf{k}$ and $\mathbf{k}'$ be the unit vectors determining the direction of propagation of incident and scattered light. If the atom is unpolarized and we detect linear polarization of the scattered light by sending it through a polarization filter ($\mathbf{e}'$ is a real-valued vector), the scattering cross section, as is well known (see, e.g., Ref. 6, §60), cannot depend on the degree of circular polarization of the incident light, ξ_2: there is no way to construct a nonzero scalar from the pseudoscalar

$$\xi_2 = i\mathbf{k}[\mathbf{e}\mathbf{e}^*], \tag{1}$$

the vector $\mathbf{k}$, and two $\mathbf{e}'$ vectors.[2)] The situation is different for aligned atoms. In the event of axisymmetric polarization, alignment, being an irreducible tensor of rank 2, is proportional to an irreducible tensor of rank 2 built from the vector $\mathbf{n}$. Hence the terms proportional to aligning must contain two $\mathbf{n}$ vectors, and under the same conditions the cross section acquires a scalar combination of vectors of the form

$$\xi_2(\mathbf{n}\mathbf{e}')(\mathbf{k}[\mathbf{n}\mathbf{e}']), \tag{2}$$

which becomes

$$\xi_2(\mathbf{k}'\mathbf{n})([\mathbf{k}\mathbf{k}']\mathbf{n}) \tag{3}$$

after summation over the polarizations of the scattered light (i.e., in establishing the angular distribution of the scattered light). Combinations (2) and (3) are T-odd, with the result that they are present in the cross section only when a dissipation channel is open, when Γ is nonzero; hence such combinations determine dissipation-induced effects of circular dichroism. When dissipation channels are open, the cross section retains a dependence on ξ_2' even when the incident light is linearly polarized or unpolarized. The corresponding constructions have the form

$$\xi_2'(\mathbf{n}\mathbf{e})(\mathbf{k}'[\mathbf{n}\mathbf{e}]) \tag{4}$$

or

$\xi_2'(\mathbf{kn})([\mathbf{k}'\mathbf{k}]\mathbf{n})$

and determine the emergence of dissipation-induced circular polarization of the scattered light (circular dichroism in the final state).

Below we derive a compact expression for the cross section of light scattering by aligned atoms and discuss dissipation-induced effects in detail. Note that although we speak of atoms, the results can be applied to the scattering of light by aligned molecules, since all the information on the dynamical structure of the scattering target is contained in the reduced matrix elements of the scattering tensor.

2. THE CROSS SECTION OF LIGHT SCATTERING BY ALIGNED ATOMS

It is convenient to specify the state of a polarized atom with a total angular momentum $j_1 \neq 0$ by the irreducible components of its density matrix, which are known as the state multipoles (statistical tensors).[1] In the present case of axisymmetric polarization only the zeroth component (in a system of coordinates whose z axis is directed along $\mathbf{n}$) ρ_K^n of the state multipoles of rank $K=0, 1, \ldots, 2j_1$ is nonzero. Here we have $\rho_0^n=(2j_1+1)^{-1/2}$, ρ_1^n determines orientation, ρ_2^n determines alignment, etc. In the absence of polarization $\rho_{K\geq 1}^n=0$.

The general expression of the cross section of light scattering by a polarized atom, obtained in Ref. 4, has the form (we use the atomic system of units)

$$\frac{d\sigma}{d\Omega'}=(4\pi)^{1/2}\omega\omega'^3\alpha^4\sum_{K,k,k'}\rho_K^n(-1)^{j_1+j_2+k+K}\times\begin{Bmatrix} k & k' & K \\ j_1 & j_1 & j_2\end{Bmatrix}(2K+1)^{-1/2}\times T_kT_{k'}^*\sum_Q Y_{KQ}^*(\mathbf{n})\{\{\mathbf{e}'^*\otimes\mathbf{e}\}_k\otimes\{\mathbf{e}'\otimes\mathbf{e}^*\}_{k'}\}_{KQ}. \tag{5}$$

Here ω (ω') is the frequency of the incident (scattered) light, α is the fine-structure constant, j_2 is the angular momentum of the atom in the final state, and

$$T_k=\langle\nu_2 j_2\|t_k\|\nu_1 j_1\rangle,\quad k=0,\ 1,\ 2,$$

are the reduced matrix elements of the irreducible parts of the scattering tensor, which emerge when the dependence on the magnetic quantum numbers in the irreducible components of the scattering tensor, t_{kq}, is separated by the Wigner–Eckart theorem (the $\nu_{1,2}$ stand for the set of atomic quantum numbers in the initial and final states with the exception of the angular momentum and its projection). Equation (5) also incorporates the spherical function $Y_{kq}(\mathbf{n})$ and the irreducible tensor of rank K constructed from the polarization vectors. An irreducible tensor of rank K constructed from irreducible tensors $A_{k_1q_1}$ and $B_{k_2q_2}$ of rank k_1 and k_2, respectively, is defined in the following manner:

$$\{A_{k_1}\otimes B_{k_2}\}_{KQ}=\sum_{q_1,q_2}C^{KQ}_{k_1q_1k_2q_2}A_{k_1q_1}B_{k_2q_2}, \tag{6}$$

where the $C^{KQ}_{k_1q_1k_2q_2}$ are Clebsch–Gordan coefficients. In the case of a vector we have $a_{1q}=a_q$, where the a_q are its spherical components,

$$a_0=a_z,\quad a_\pm=\mp\frac{1}{\sqrt{2}}(a_x\pm ia_y). \tag{7}$$

The irreducible components of the scattering tensor have the form

$$t_{kq}=\sum_{q_1,q_2}C^{kq}_{1q_11q_2}(d_{q_1}\hat{G}_{E_1+\omega}d_{q_2}+d_{q_2}\hat{G}_{E_1-\omega'}d_{q_1}), \tag{8}$$

where d_q is the spherical component of the atomic dipole moment,

$$\hat{G}_E=\sum_n\frac{|n\rangle\langle n|}{E_n-E-i0} \tag{9}$$

is the resolvent of the atomic Hamiltonian, and E_1 is the initial-state energy of the atom.

Let us assume that the atom is polarized in such a way that only the alignment ρ_2^n is nonzero. We write the cross section (5) in a compact form convenient for analysis, namely, containing the ordinary scalar and vector products of the polarization vectors $\mathbf{e}$ and $\mathbf{e}'$, the vectors $\mathbf{k}$ and $\mathbf{k}'$ specifying the direction of propagation of the incident and scattered light, and the vector $\mathbf{n}$ defining the position of the alignment axis. We write the cross section of the light scattering by an aligned atom as a sum of two terms:

$$\frac{d\sigma}{d\Omega'}=\frac{d\sigma^{(\mathrm{unp})}}{d\Omega'}+\frac{d\sigma^{(\mathrm{al})}}{d\Omega'}. \tag{10}$$

Here

$$\frac{d\sigma^{(\mathrm{unp})}}{d\Omega'}=\frac{\omega\omega'^3\alpha^4}{2j_1+1}\left\{\frac{1}{3}|T_0|^2|\mathbf{e}'^*\mathbf{e}|^2+\frac{1}{6}|T_1|^2(1-|\mathbf{e}'\mathbf{e}|^2)+\frac{1}{10}|T_2|^2\left(1+|\mathbf{e}'\mathbf{e}|^2-\frac{2}{3}|\mathbf{e}'^*\mathbf{e}|^2\right)\right\} \tag{11}$$

is the cross section of light scattering by an unpolarized atom (see Refs. 4 and 6, §60) consisting of scalar, skew-symmetric and symmetric parts. The presence of each of these parts, determined by the irreducible components of the scattering tensor T_k, $k=0, 1, 2$, is possible only if the triangle condition $\Delta(j_1,j_2,k)$ is met.

The second terms on the right-hand side of Eq. (10), proportional to ρ_2^n, determines an addition to the cross section caused by the atom's alignment. The spherical function $Y_{2Q}(\mathbf{n})$ can be represented by an irreducible tensor of rank 2 (Eq. (6)) constructed from the vector $\mathbf{n}$ (Ref. 8):

$$Y_{2Q}(\mathbf{n})=\sqrt{\frac{15}{8\pi}}\{\mathbf{n}\otimes\mathbf{n}\}_{2Q}.$$

As a result, the sum over Q in (5) at $K=2$ can be expressed in terms of a scalar product (denoted by round brackets) of two irreducible tensors of rank 2 built from the corresponding vectors:

$$\sum_{Q}(-1)^{Q}Y_{2,-Q}(\mathbf{n})\{\{\mathbf{e}'^{*}\otimes\mathbf{e}\}_{k}\otimes\{\mathbf{e}'\otimes\mathbf{e}^{*}\}_{k'}\}_{2Q}$$

$$=\sqrt{\frac{15}{8\pi}}(\{\mathbf{n}\otimes\mathbf{n}\}_{2},\{\{\mathbf{e}'^{*}\otimes\mathbf{e}\}_{k}\otimes\{\mathbf{e}'\otimes\mathbf{e}^{*}\}_{k'}\}_{2}). \qquad (12)$$

At $k=k'=1$, and $k=0$ and $k'=2$ ($k=2$ and $k'=0$) the scalars (12) can be expressed in terms of ordinary products of the constituent vectors by employing the formulas from Ref. 8. The formulas for the more complicated cases with $k=k'=2$, and $k=1$ and $k'=2$ ($k=2$ and $k'=1$) are given in the Appendix. After separating all vector combinations, we arrive at the following expression for the second term on the right-hand side of Eq. (10):

$$\frac{d\sigma^{(\mathrm{al})}}{d\Omega'}=\frac{1}{\sqrt{2}}\omega\omega'^{3}\alpha^{4}\rho_{2}^{n}\Bigg\{a_{-}+a_{+}|\mathbf{e}'\mathbf{e}|^{2}+\frac{1}{3}b|\mathbf{e}'^{*}\mathbf{e}|^{2}$$
$$+c_{-}|\mathbf{n}\mathbf{e}|^{2}-c_{+}|\mathbf{n}\mathbf{e}'|^{2}+\frac{\sqrt{3}}{2}R_{11}|\mathbf{n}[\mathbf{e}'^{*}\mathbf{e}]|^{2}$$
$$-b\,\mathrm{Re}[(\mathbf{n}\mathbf{e}^{*})(\mathbf{n}\mathbf{e}')(\mathbf{e}'^{*}\mathbf{e})]-\frac{3}{\sqrt{7}}R_{22}$$
$$\times\mathrm{Re}[(\mathbf{n}\mathbf{e})(\mathbf{n}\mathbf{e}')(\mathbf{e}'^{*}\mathbf{e}^{*})]+d_{-}\xi_{2}'\mathbf{k}'\,\mathrm{Re}([\mathbf{e}^{*}\mathbf{n}](\mathbf{e}\mathbf{n}))$$
$$-d_{+}\xi_{2}\mathbf{k}\,\mathrm{Re}([\mathbf{e}'^{*}\mathbf{n}](\mathbf{e}'\mathbf{n}))\Bigg\}, \qquad (13)$$

where the degree of circular polarization of the incident light, ξ_2, equal to ± 1 for right-hand (left-hand) circular polarization and to zero for linear polarization, is defined in (1), and

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

The coefficients $a_{\pm}$, b, $c_{\pm}$, $d_{\pm}$, and $R_{kk'}$ are expressed in terms of $6j$-symbols and the reduced matrix elements of the scattering tensor, T_k:

$$a_{\pm}=\frac{1}{\sqrt{7}}R_{22}\pm\frac{1}{2\sqrt{3}}R_{11},\quad b=2\left(R_{02}-\frac{2}{\sqrt{7}}R_{22}\right),$$
$$c_{\pm}=R_{12}\pm\frac{3}{2\sqrt{7}}R_{22},\quad d_{\pm}=I_{02}\pm I_{12},$$
$$R_{kk'}=(-1)^{j_1+j_2}\begin{Bmatrix} j_2 & j_1 & k' \\ 2 & k & j_1\end{Bmatrix}\mathrm{Re}(T_kT_{k'}^{*}), \qquad (14)$$
$$I_{kk'}=(-1)^{j_1+j_2}\begin{Bmatrix} j_2 & j_1 & k' \\ 2 & k & j_1\end{Bmatrix}\mathrm{Im}(T_kT_{k'}^{*}).$$

Note that the last two terms on the right-hand side of Eq. (13) contain T-odd scalar combinations of vectors of the form (2) and (4). The scattering cross section is T-even, with the result that the coefficients $d_{\pm}$ must be T-odd. The $\mathrm{Im}(T_kT_{k'}^{*})$ in $d_{\pm}$ (see Eqs. (14)) is T-odd. This quantity is nonzero if the scattering tensor (8) is non-Hermitian. The skew-Hermitian part of the scattering tensor is T-odd and does not vanish in above-threshold scattering (the photon energy exceeds the ionization threshold) because of the non-hermiticity in this case of the resolvent $\hat{G}_{E_1+\omega}$ (Eq. (9)), while in resonant scattering the same is true due to the finite width Γ of the resonant level. In the latter case the resonant level is sure to have a multipole structure (for a resonance involving a singlet, $\mathrm{Im}(T_kT_{k'}^{*})=0$), with

$$\mathrm{Im}(T_kT_{k'}^{*})\sim\frac{\Gamma}{\Delta}\mathrm{Re}(T_kT_{k'}^{*}),$$

where Δ is of the order of the fine splitting of the resonant sublevels, so that the effects in question are determined in resonant scattering by the small parameter Γ/Δ.

Thus, the coefficients $d_{\pm}$ and the corresponding terms in (13) emerge only when a channel for dissipation of luminous energy is open in the process of light scattering: the photoionization channel in above-threshold scattering, and radiative, collisional, or other channels in resonant scattering. Here the physical parameter determining the rate of dissipation of luminous energy (the photoionization probability, and the width of the resonant level) acts as a T-odd parameter. The matrix element of the skew-Hermitian part of the scattering tensor is proportional to this parameter.

The effect of dissipation of luminous energy on the scattering of light by oriented atoms was studied in Ref. 5. Below we discuss a number of dissipation-induced effects emerging in the scattering of light by aligned atoms. Similar effects can be observed in the coherent scattering of electromagnetic radiation by aligned systems.[9]

Note that in the event of axisymmetric mirror polarization (with $j_1>3/2$) the state multipole ρ_4^n contributing to the scattering cross section (5) can also be nonzero. Here the cross section acquires a number of additional terms proportional to the T-even parameter $|T_2|^2$, so that the coefficients $d_{\pm}$ (see Eqs. (13) and (14)) of the T-odd combinations of vectors of types (2)–(4) do not change. Hence the discussed effects of circular dichroism are caused solely by atom alignment.

3. CIRCULAR DICHROISM IN THE ANGULAR DISTRIBUTION OF THE SCATTERED LIGHT

By circular dichroism we mean the change in the angular distribution of the scattered light that takes place when right-hand (left-hand) circular polarization of the incident light is replaced by left-hand (right-hand). Obviously, this effect is absent when the light is scattered by an unpolarized atomic target: there is no way in which a scalar combination can be built from the pseudoscalar ξ_2 (Eq. (1)) and the vectors $\mathbf{k}$ and $\mathbf{k}'$. The manifestation of circular dichroism is no less obvious when the light is scattered by an oriented atom: the orientation ρ_1^n is a pseudoscalar, and circular dichroism in the angular distribution[5] is retained in the total cross section as well.[4]

When the light is scattered by aligned atoms, the effect of circular dichroism is determined by the last term on the right-hand side of Eq. (13). To establish the angular distribution of the scattered light when the light polarization is not detected, we must sum the differential cross section (10) over two independent polarizations. This can easily be done in (11) and (13) by employing the well-known identity (see Ref. 6, §45)

$$\sum_{\lambda} (\mathbf{a}\mathbf{e}'_{\lambda})(\mathbf{b}\mathbf{e}'^{*}_{\lambda})=[\mathbf{k}'\mathbf{a}][\mathbf{k}'\mathbf{b}]. \tag{15}$$

We arrive at following expression for the angular distribution:

$$\frac{d\sigma^s}{d\Omega'}=\omega\omega'^3\alpha^4\Bigg\{\frac{1}{3(2j_1+1)}\Bigg[|T_0|^2+\frac{1}{2}|T_1|^2+\frac{7}{10}|T_2|^2-\left(|T_0|^2-\frac{1}{2}|T_1|^2+\frac{1}{10}|T_2|^2\right)|\mathbf{k}'\mathbf{e}|^2\Bigg]+\frac{1}{\sqrt{2}}\rho_2^n[A+B|\mathbf{k}'\mathbf{e}|^2+C(\mathbf{k}'\mathbf{n})^2+D|\mathbf{n}\mathbf{e}|^2+F\mathbf{k}'\mathbf{n}\,\mathrm{Re}\,[(\mathbf{n}\mathbf{e})(\mathbf{k}'\mathbf{e}^*)]+d_+\xi_2\mathbf{k}'\mathbf{n}([\mathbf{k}\mathbf{k}']\mathbf{n})]\Bigg\}, \tag{16}$$

where the coefficients A, B, C, D, and F can be expressed in terms of the parameters (14) introduced earlier as follows:

$$A=2a_-+a_++\frac{1}{3}b-c_+=\frac{1}{6\sqrt{7}}R_{22}-\frac{1}{2\sqrt{3}}R_{11}+\frac{2}{3}R_{02}-R_{12},$$

$$B=\frac{\sqrt{3}}{2}R_{11}-a_+-\frac{1}{3}b=\frac{1}{\sqrt{3}}R_{11}+\frac{1}{3\sqrt{7}}R_{22}-\frac{2}{3}R_{02},$$

$$C=c_++\frac{\sqrt{3}}{2}R_{11}=R_{12}+\frac{3}{2\sqrt{7}}R_{22}+\frac{\sqrt{3}}{2}R_{11},$$

$$D=2c_--b-\frac{3}{\sqrt{7}}R_{22}=2\left(R_{12}-R_{02}-\frac{1}{\sqrt{7}}R_{22}\right),$$

$$F=b+\frac{3}{\sqrt{7}}R_{22}-\sqrt{3}R_{11}=2R_{02}-\frac{1}{\sqrt{7}}R_{22}-\sqrt{3}R_{11}.$$

The last term on the right-hand side of Eq. (16), containing a T-odd combination of vectors of type (3), changes sign if right-hand circular polarization of the incident light is replaced by left-hand. Thus, the effect of circular dichroism in the angular distribution of the scattered light is dissipation-induced. Our analysis at the end of the previous section shows that this effect can be observed in both above-threshold and resonant scattering. In the latter case the effect is small (the coefficient d_+ contains the small parameter Γ/Δ).[3)]

The physical origin of circular dichroism emerging in the event of an open dissipation channel can be most readily understood on the basis of the following reasoning. If only a circularly polarized photon is detected in the scattering act, circular dichroism can be observed in the absence of dissipation (for instance, for unpolarized atoms this effect is caused by the term containing a combination of vectors of the type $\xi_2\xi'_2(\mathbf{k}\mathbf{k}')$). However, in this case circular dichroism vanishes from the angular distribution, which is determined by the sum of the scattering intensities over two oppositely directed circular polarizations of the scattered light. At the same time, when a luminous-energy dissipation channel is open and, because of this, processes in an atom become markedly irreversible, the intensity of successive emission of two photons by the atom changes somewhat. As a result the total scattering intensity (i.e., summed over two oppositely directed circular polarizations) proves to be different for right-hand and left-hand circular polarizations of the incident radiation, so that circular dichroism is retained in the angular distribution.

Note that circular dichroism is not observed in the total cross section of light scattering by aligned atoms, which is a simple consequence of parity conservation.[4] Integration of the cross section (16) over all scattering directions can easily be carried out by employing the following well-known identities:

$$\int k'_i d\Omega'=0, \quad \int k'_i k'_j d\Omega'=\frac{4\pi}{3}\delta_{ij},$$

and we arrive at the following expression for the total scattering cross section:

$$\sigma^s=\frac{4\pi}{3}\omega\omega'^3\alpha^4\Bigg\{\frac{1}{3(2j_1+1)}\sum_{k=0}^{2}|T_k|^2+\frac{1}{\sqrt{2}}\rho_2^n[3A+B+C+(3D+F)|\mathbf{n}\mathbf{e}|^2]\Bigg\}.$$

4. DISSIPATION-INDUCED POLARIZATION FEATURES OF THE SCATTERED RADIATION

Let us discuss some specific features of the polarization of light scattered by aligned atoms that emerge when luminous-energy dissipation channels are open.

If the scattered light is sent through a polarization filter that transmits only linearly polarized radiation, i.e., only the linear polarization of the scattered light is detected, circular dichroism is determined by the last term on the right-hand side of Eq. (13) ($\mathbf{e}'$ is a real-valued vector, and $\xi'_2=0$). Hence the circular dichroism effect, which can be observed in the powers of linear polarization (or the corresponding Stokes parameters[1]) of the light scattered by the aligned atoms, is also dissipation-induced.

Let us now assume that the incident light is linearly polarized. Then the difference between the scattering cross sections with detection of right- and left-hand circular polarizations is due solely to the penultimate term on the right-hand side of Eq. (13) (circular dichroism in the final photon state), which determines the degree of circular polarization of the scattered light:

$$\eta'_2=\frac{1}{\sqrt{2}}\omega\omega'^3\alpha^4\rho_2^n d_-(\mathbf{k}'[\mathbf{e}\mathbf{n}])(\mathbf{e}\mathbf{n})\left(\frac{d\sigma^s}{d\Omega'}\right)^{-1}. \tag{17}$$

Here $d\sigma^s/d\Omega'$ is given by the expression (16) with **e** the real-valued linear-polarization vector and $\xi_2=0$. Thus, when the polarization of the incident light is linear, the degree of circular polarization of the light scattered by the aligned atoms is dissipation-induced and hence is nonzero only in above-threshold or resonant scattering. It is also obvious that in all cases where the degree of circular polarization of the incident radiation is zero, the degree of circular polarization of the scattered light is nonzero only when a dissipation channel is open. The corresponding expression for η_2' can easily be obtained, say, for the case of scattering of unpolarized light, by averaging the numerator and denominator of (17) over two independent polarizations of the incident photon via an identity similar to (15). Here the T-odd combination of the vectors in the numerator of (17) is replaced by a T-odd combination of the form

$$\overline{(\mathbf{k}'[\mathbf{en}])(\mathbf{en})}=\frac{1}{2}(\mathbf{nk})([\mathbf{kk}']\mathbf{n}).$$

APPENDIX

Let us see how the scalar constructions

$$S_{kk'}=(\{\mathbf{a}\otimes\mathbf{b}\}_2,\{\{\mathbf{c}\otimes\mathbf{d}\}_k\otimes\{\mathbf{e}\otimes\mathbf{f}\}_{k'}\}_2) \tag{A1}$$

(see Eqs. (12) and (6)) can be expressed for $k=k'=2$, and $k=1$ and $k'=2$ in terms of scalar products of the constituent vectors.

The components of a Cartesian tensor A_{ak} can be used to construct three irreducible tensors

$$A_q^K=\sum_{q_1,q_2}C^{Kq}_{1q_11q_2}A_{q_1q_2},\quad K=0,\ 1,\ 2,$$

where $A_{q_1q_2}$ are the spherical components of the tensor constructed for each index in a way similar to the spherical components (7) of a vector. Using the well-known expression for the sum over the projections of the angular momenta of the product of three $3j$-symbols (see, e.g., Ref. 10), we can express a scalar built from three Cartesian tensors in terms of the corresponding irreducible tensors:

$$\begin{aligned}A_{ik}B_{ij}C_{jk}&=\sum_{q_1,q_2,q_3}(-1)^{q_1+q_2+q_3}A_{q_1q_2}B_{-q_1q_3}C_{-q_3-q_2}\\&=-\sum_{K,K',K''}[(2K'+1)(2K''+1)]^{1/2}\\&\quad\times\begin{Bmatrix}K & K' & K''\\ 1 & 1 & 1\end{Bmatrix}(A^K,\{B^{K'}\otimes C^{K''}\}_K).\end{aligned} \tag{A2}$$

Examining S_{22} of Eq. (A1), we use each pair of vectors to build a zero–trace symmetric tensor:

$$\begin{aligned}A_{ik}&=\frac{1}{2}(a_ib_k+a_kb_i)-\frac{1}{3}(\mathbf{ab})\delta_{ik},\\B_{ik}&=\frac{1}{2}(c_id_k+c_kd_i)-\frac{1}{3}(\mathbf{cd})\delta_{ik},\\C_{ik}&=\frac{1}{2}(e_if_k+e_kf_i)-\frac{1}{3}(\mathbf{ef})\delta_{ik}.\end{aligned} \tag{A3}$$

Here only the irreducible tensors of rank 2, $A_q^2=\{\mathbf{a}\otimes\mathbf{b}\}_{2q}$, and the like are nonzero. As a result, only one term containing the scalar S_{22} remains in the sum on the right-hand side of Eq. (A2). By employing Eqs. (A3), we can easily express this scalar in terms of scalar products of the vectors:

$$\begin{aligned}S_{22}=-\sqrt{\frac{3}{7}}\Bigg\{&\frac{1}{4}[(\mathbf{ac})(\mathbf{bf})(\mathbf{ed})+(\mathbf{ac})(\mathbf{be})(\mathbf{df})\\&+(\mathbf{ad})(\mathbf{bf})(\mathbf{ce})+(\mathbf{ad})(\mathbf{eb})(\mathbf{cf})+(\mathbf{ae})(\mathbf{bd})(\mathbf{cf})\\&+(\mathbf{ae})(\mathbf{bc})(\mathbf{df})+(\mathbf{af})(\mathbf{bd})(\mathbf{ce})+(\mathbf{af})(\mathbf{bc})(\mathbf{ed})]\\&-\frac{1}{3}[(\mathbf{ab})(\mathbf{ed})(\mathbf{cf})+(\mathbf{ab})(\mathbf{df})(\mathbf{ce})+(\mathbf{cd})(\mathbf{ae})(\mathbf{bf})\\&+(\mathbf{cd})(\mathbf{af})(\mathbf{be})+(\mathbf{ef})(\mathbf{ac})(\mathbf{bd})+(\mathbf{ef})(\mathbf{ad})(\mathbf{bc})]\\&+\frac{4}{9}(\mathbf{ab})(\mathbf{cd})(\mathbf{ef})\Bigg\}.\end{aligned}$$

When we examine S_{12} (Eq. (A2)), instead of constructing the symmetric tensor B_{ik} we construct a skew-symmetric tensor from the vectors **c** and **d**:

$$B_{ik}=\frac{1}{2}(c_id_k-c_kd_i).$$

Here only the irreducible tensor of rank 1, $B_q^1=\{\mathbf{c}\otimes\mathbf{d}\}_{1q}$, is nonzero, and only one term containing the scalar S_{12} remains in the sum on the right-hand side of Eq. (A3). The expression that we obtain for this scalar is

$$\begin{aligned}S_{12}=\frac{1}{4\sqrt{3}}\{&(\mathbf{ac})(\mathbf{bf})(\mathbf{de})+(\mathbf{ac})(\mathbf{be})(\mathbf{df})\\&-(\mathbf{ad})(\mathbf{bf})(\mathbf{ce})-(\mathbf{ad})(\mathbf{be})(\mathbf{cf})\\&+(\mathbf{af})(\mathbf{bc})(\mathbf{de})+(\mathbf{ae})(\mathbf{bc})(\mathbf{df})\\&-(\mathbf{af})(\mathbf{bd})(\mathbf{ce})-(\mathbf{ae})(\mathbf{bd})(\mathbf{cf})\}.\end{aligned}$$

1) State multipoles up to the fourth rank inclusive may manifest themselves in the differential cross section, and up to the second in the total cross section.[4]

2) If nondipole effects are taken into account, the cross section acquires constructions of the form $\xi_2(\mathbf{ke}')([\mathbf{kk}']\mathbf{e}')$, which results in dissipation-induced polarization anomalies in the scattering.[7]

3) The circular dichroism effect manifests itself much more vividly in a quadrupole resonance, where a T-odd parameter appears in the cross section as a result of interference of the resonant quadrupole and nonresonant dipole parts of the scattering tensor (similar to dissipation-induced effects in the scattering of light by oriented atoms[5]).

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