Polarization anomalies caused by dissipation processes in the scattering of light by gases

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Allowing for dissipation in the scattering of light by a freely orienting system leads to distinctive polarization asymmetry, e.g., to the emergence of circular polarization of the scattered radiation in the scattering of photons with linear polarization. The phenomenon is similar to the emergence of ellipticity in the light reflected from an absorbing-medium boundary (a metal) and is caused by the skew-Hermitian part of the scattering amplitude. This paper studies the asymmetry in dipole-allowed scattering for various dissipation mechanisms: the photoelectric effect, radiative reaction, and resonant scattering. The polarization–angular structure of dipole-forbidden light scattering is analyzed, and we calculate numerically the cross section of such scattering and the degree of asymmetry for hydrogen and cesium atoms.

1. INTRODUCTION

Dissipation of light energy in the propagation of light through a medium leads to several effects whose specific features are determined by the irreversible nature of the dissipation process. For instance, Baranova, Bogdanov, and Zel'dovich[1] and Manakov and Faĭnshteĭn[2] examined new effects associated with the variation in the electric and magnetic properties of a medium in the presence of dissipation, such as gyrotropic properties induced by a constant electric field (the electrical analog of the Faraday effect) in a conducting liquid and optical rectification (induction of a static electric polarization by a monochromatic wave) in a centrally symmetric medium. Note also the classical emergence of elliptical polarization in the reflection of linearly polarized radiation from an absorbing-medium boundary (a metal).[3] Formally, the new effects emerge because the problem contains a T-odd parameter Γ characterizing the intensity of dissipation.

This paper examines similar effects in the scattering of light by a gaseous medium. Allowing for dissipation in this case leads to distinctive anomalies in the polarization–angular dependence of the cross section: for one thing, scattered photons are circularly polarized because of scattering of linearly polarized radiation by unpolarized particles. The role of the parameter Γ mentioned earlier is taken in scattering processes by the skew-Hermitian part of the scattering amplitude, this part being related to amplitudes of other physical processes than the present one, which constitute the sources of dissipation.

Although there may be various dissipation mechanisms, the "anomalous" terms in the scattering cross section usually arise because of interference of the Hermitian and skew-Hermitian parts of the partial amplitudes and have the same polarization–angular structure. Let $\mathbf{e}_1,\mathbf{k}_1,\omega_1$ and $\mathbf{e}_2,\mathbf{k}_2,\omega_2$ be the polarization vectors, the wave vectors, and the frequencies of the incident and scattered photons ($|\mathbf{e}_m|^2=1$, $\mathbf{k}_m=(\omega_m/c)\mathbf{n}_m$, $m=1,2$), and $A_m=i\mathbf{n}_m\cdot(\mathbf{e}_m\times\mathbf{e}_m^*)$ and $l_m=\mathbf{e}_m\cdot\mathbf{e}_m=\sqrt{1-A_m^2}$ the degree of circular and linear polarization of the mth photon ($-1\leqslant A_m\leqslant 1$). As will be shown below, in the presence of dissociation, the cross section $d\sigma/d\Omega$ is different for right-hand and left-hand circularly polarized photons, so that

$$\Delta\left(\frac{d\sigma}{d\Omega}\right)\equiv\frac{d\sigma(A_1,A_2)}{d\Omega}-\frac{d\sigma(-A_1,-A_2)}{d\Omega}$$
$$=f(\omega_1,-\omega_2,\theta)I_1-f(-\omega_2,\omega_1,\theta)I_2, \qquad (1)$$

where $\theta=\cos^{-1}(\mathbf{n}_1\cdot\mathbf{n}_2)$ is the scattering angle, and

$$I_1=A_1\ \mathrm{Re}\{[\mathbf{e}_2\cdot(\mathbf{n}_1\times\mathbf{n}_2)](\mathbf{e}_2^*\cdot\mathbf{n}_1)\},$$
$$I_2=A_2\ \mathrm{Re}\{[\mathbf{e}_1\cdot(\mathbf{n}_2\times\mathbf{n}_1)](\mathbf{e}_1^*\cdot\mathbf{n}_2)\}. \qquad (2)$$

Note that the right–left asymmetry in the scattering of light, determined by Δ, arises in the absence of dissipation processes as well, but in right–left asymmetric media, e.g., in a chiral molecular gas.[4] The vector structure of Δ differs from the right-hand side of Eq. (1).

The term with I_2 in Eq. (1) describes the onset of circular polarization in the scattering of a linearly polarized photon, and I_1 describes the reciprocal effect. Since I_1 is proportional to l_2 and I_2 to l_1, both I_1 and I_2 vanish if the two photons are 100% linearly or 100% circularly polarized. On the other hand, the effect peaks when one photon is circularly polarized and the other linearly polarized. Suppose, for instance, that the incident photon is linearly polarized ($\mathbf{e}_1=\mathbf{e}_1^*$) at an angle β to the scattering plane and that β is neither 0 nor 90°. Then the scattered radiation contains photons with circular polarization whose number is determined by the term with I_2 in Eq. (1), and the numbers of left-hand ($A_2=1$) and right-hand ($A_2=-1$) circularly polarized photons scattered in the same direction differ. The quantities $I_{1,2}$ are at their maximum in scattering through 90° and $\beta=45°$ and disappear in the total cross section integrated over angles.

Interestingly, when light is reflected by a metal surface, the difference ΔR in the reflection coefficients for photons of different circular polarizations is also described by Eqs. (1)

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and (2), with the subscripts 1 and 2 referring to the incident and reflected waves, $\omega_1=\omega_2\equiv\omega$, and the function $f(\omega,-\omega,\theta)$ proportional to the absorption coefficient (the imaginary part of the dielectric constant) of the reflecting medium.

The effects can also be described in terms of Stokes parameters, which prove convenient in the study of polarization phenomena and make it possible to allow for partial polarization of photons. In this connection some results that will prove useful in what follows can be recalled (see, e.g., Ref. 5). The Stokes parameters of the incident and scattered photons are denoted by $\xi_1^{(1)},\xi_2^{(1)},\xi_3^{(1)}$ and $\xi_1^{(2)},\xi_2^{(2)},\xi_3^{(2)}$, respectively. They are defined with respect to two coordinate systems, x_1,y_1,z_1 and x_2,y_2,z_2, whose z axes are directed along the wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, the x axes coincide and are perpendicular to the scattering plane $[x\|(\mathbf{k}_1\times\mathbf{k}_2)]$, and the y_1 and y_2 axes lie in the scattering plane. The parameters $\xi_2^{(i)}$ give the degrees of circular polarization ($\xi_2^i=A_i$), the $\xi_3^{(i)}$ characterize the linear polarization along the x_i or y_i axis ($\xi_3^{(i)}=+1$ or $\xi_3^{(i)}=-1$ corresponds to full polarization along the x_i or y_i axis), and the $\xi_1^{(i)}$ characterize linear polarization in a similar manner in directions that form an angle of $\frac{1}{4}\pi$ or $-\frac{1}{4}\pi$ with the x axis. For a fully polarized photon, $\xi_1=l\sin 2\varphi$ and $\xi_3=l\cos 2\varphi$, where φ is the angle between the x axis and the semimajor axis of the polarization ellipse, and $l=\sqrt{\xi_1^2+\xi_3^2}$ is the degree of maximum linear polarization introduced earlier.

Introducing, as usual, the photon density matrix $\rho_{\alpha\beta}$ by replacing $e_\alpha e_\beta^*$ with $\rho_{\alpha\beta}$ and expressing $\rho_{\alpha\beta}$ in terms of the Stokes parameters, we reduce Eqs. (2) to

$$I_1=\tfrac{1}{2}\xi_2^{(1)}\xi_1^{(2)}[1-(\mathbf{n}_1\cdot\mathbf{n}_2)^2],$$

$$I_2=\tfrac{1}{2}\xi_1^{(1)}\xi_2^{(2)}[1-(\mathbf{n}_1\cdot\mathbf{n}_2)^2].$$

This implies that the features just described are due to terms proportional to $\xi_1^{(i)}\xi_2^{(k)}$ in the scattering cross section. In this regard it must be noted that the polarization–angular dependence of the scattering cross section of a photon scattered by an unpolarized free electron in relativistic theory has the form[5]

$$\frac{d\sigma}{d\Omega}=\frac{r_0^2}{12}\{2f+g[(1+\xi_3^{(1)})(1+\xi_3^{(2)})+(1-\xi_3^{(1)})(1-\xi_3^{(2)})\cos^2\theta+2\xi_1^{(1)}\xi_1^{(2)}\cos\theta]+h\xi_2^{(1)}\xi_2^{(2)}\cos\theta\}, \tag{3}$$

where r_0 is the classical electron radius, and f, g, and h are simple functions of the frequencies of the incident and scattered photons. As shown in Ref. 5, the lack of terms proportional to $\xi_1^{(i)}\xi_2^{(k)}$ is due to the hermiticity of the scattering amplitude in second-order quantum electrodynamic perturbation theory used in deriving Eq. (3), and is not forbidden by general symmetry considerations as it is, for example, in relation to terms proportional to $\xi_1^{(i)}$ and $\xi_2^{(i)}$ or $\xi_1^{(i)}\xi_3^{(k)}$ and $\xi_2^{(i)}\xi_3^{(k)}$. The known nonrelativistic expression for the scattering cross section of light scattered by a freely orienting system accompanied by a transition between states of the same parity in the dipole approximation,[5]

$$\frac{s\sigma^{(0)}}{d\Omega}=r_0^2\left\{\frac{G^0}{3}|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2+\frac{G^1}{6}(1-|\mathbf{e}_1\cdot\mathbf{e}_2|^2)^2+\frac{G^2}{10}\left(1+|\mathbf{e}_1\cdot\mathbf{e}_2|^2-\frac{2}{3}|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2\right)\right\}, \tag{4}$$

when written in terms of the Stokes parameters, has the same polarization–angular structure as Eq. (3). The only thing that changes is the formulas for the invariant parameters f, g, and h, which in terms of the scalar (G^0), skew-symmetric (G^1), and symmetric (G^2) scattering cross sections have the form

$$f=G^1+\tfrac{1}{5}G^2,\quad g=G^0-\tfrac{1}{2}G^1+\tfrac{1}{10}G^2,\quad h=2G^0+G^1-G^2.$$

For a bound electron, the scattering amplitude may have a skew-Hermitian part by the first nonvanishing order, and the lack of terms of type (2) in Eq. (4) is a result of the dipole approximation [the independence of the amplitude from the photon wave vectors in the vector combinations in (2)]. The terms proportional to $\xi_1^{(i)}\xi_2^{(k)}$ in the radiative corrections to Eq. (3) were calculated in Ref. 6, and in the scattering cross section of hard photons scattered by a relativistic atom in Ref. 7.

Below, the functions $f(\omega_1,-\omega_2)$ in Eq. (1) are obtained for various dissipation mechanisms accompanying the scattering of light by a freely orienting system. Since the nondipole effects in the interaction of light and the scatterer must be taken into account, as mentioned earlier, Sec. 2 studies the general structure of the scattering cross section with allowance in the first nonvanishing order for the magnetic dipole and electric quadrupole terms in the radiative interaction operator and examines the case where $\hbar\omega_1$ exceeds ionization energy $|E_i|$ of the initial state $|i\rangle$ of the atom. Here the irreversible process that leads to terms of type (2) in $d\sigma/d\Omega$ is the photoelectric effect. Note that after absorbing a photon $\hbar\omega_1$ and emitting a photon $\hbar\omega_2$, the atom goes into a bound state instead of becoming ionized. The anomaly arises because scattering occurs against the background of an open photoionization channel (the atom can be said to be virtually ionized and then recombined in the scattering process).

One universal dissipation mechanism independent of the relation between $\hbar\omega_{1,2}$ and $|E_i|$ is radiative reaction caused by light scattering. Indeed, fixed- angle scattering of light always occurs against the background of an open channel of scattering into other angles and of transitions into other final states allowed by energy conservation and selection rules. Allowing for radiative reaction requires calculating the radiative corrections to the scattering amplitude. Section 3 examines such a situation for $\hbar\omega_1<|E_i|$. Of course, in the optical frequency range considered here, the functions f in Eq. (1) that result from radiative corrections are small but increase at resonance. Here, allowing for radiative corrections requires only introducing resonant level widths. Resonant scattering is examined in Sec. 4.

Nondipole effects were discussed above in connection with ordinary scattering accompanied by a transition between atomic levels $|i\rangle$ and $|f\rangle$ of the same parity. In dipole-forbidden scattering (between states of opposite parities), type (2) terms in the cross section are no longer small cor-

rections; their presence for $\hbar\omega_1 > |E_i|$ was noted in Ref. 8. Section 5 studies the polarization–angular dependence of the dipole-forbidden scattering cross section and gives the results of numerical calculations of $d\sigma/d\Omega$. In the process, a general expression is derived for the scattering cross section in the simplest possible form, which contains only scalar products of vectors present in the problem and trivial combinations of reduced matrix elements of a two-photon transition. Note that the general structure of the dipole-forbidden scattering cross section has been studied in several papers (see, e.g., Refs. 8 and 9), but the results proved to be extremely complicated since the angular part is expressed in terms of tensor products of six vectors, which are difficult to analyze, and the radial part in terms of cumbersome combinations of reduced matrix elements incorporating the Wigner $9j$ and $12j$ symbols.

2. LIGHT SCATTERING CROSS SECTION WITH NONDIPOLE INTERACTION EFFECTS

In the most general case, the scattering cross section for a centrally symmetric system contains only combinations of the vectors $\mathbf{e}_i$ and $\mathbf{n}_i$ $(i=1,2)$, with $d\sigma/d\Omega$ a linear function of each vector $\mathbf{e}_1$, $\mathbf{e}_1^*$, $\mathbf{e}_2$, and $\mathbf{e}_2^*$. If nondipole effects are considered only in the first nonvanishing order, the dependence of the cross section on $\mathbf{n}_1$ and $\mathbf{n}_2$ is quadratic, so that the general structure of the polarization–angular dependence can be established on the grounds of phenomenological considerations by computing the number of linearly independent combinations of the above vectors. Choosing the following 12 combinations has proved expedient (the orthogonality condition $(\mathbf{e}_k\cdot\mathbf{n}_k)=0$ and normalization have been used in building the combinations):

$$
\begin{aligned}
&\Phi_0=(\mathbf{e}_1\cdot\mathbf{e}_1^*)(\mathbf{e}_2\cdot\mathbf{e}_2^*)=0,\\
&\Phi_1=(\mathbf{e}_1\cdot\mathbf{e}_2^*)(\mathbf{e}_2^*\cdot\mathbf{e}_2)=|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2,\\
&\Phi_2=(\mathbf{e}_1\cdot\mathbf{e}_2)(\mathbf{e}_1^*\cdot\mathbf{e}_2^*)=|\mathbf{e}_1\cdot\mathbf{e}_2|^2,\\
&(\mathbf{n}_1\cdot\mathbf{n}_2)\Phi_i,\quad i=0,1,2,\\
&|\mathbf{e}_1\cdot\mathbf{n}_2|^2\quad |\mathbf{e}_2\cdot\mathbf{n}_1|^2,\\
&\mathrm{Re}\,E_1,\quad \mathrm{Im}\,E_1,\\
&\mathrm{Re}\,E_2,\quad \mathrm{Im}\,E_2,
\end{aligned}
\tag{5}
$$

where

$$E_1=(\mathbf{e}_1\cdot\mathbf{e}_2^*)(\mathbf{e}_1^*\cdot\mathbf{n}_1)(\mathbf{e}_2\cdot\mathbf{n}_1),\quad E_2=(\mathbf{e}_1\cdot\mathbf{e}_2)(\mathbf{e}_1^*\mathbf{n}_2)(\mathbf{e}_2^*\cdot\mathbf{n}_1),$$

In dipole scattering, the cross section is independent of $\mathbf{n}_i$, with only the coefficients of Φ_i being nonzero, in accordance with (4). In certain special cases, the number of invariant parameters determining $d\sigma/d\Omega$ also decreases. For instance, in elastic scattering ($\omega_1=\omega_2$), symmetry under time reversal (see Sec. 87 in Ref. 5), i.e., invariance of the cross section under the substitutions

$$\mathbf{n}_{1,2}\to-\mathbf{n}_{2,1},\quad \mathbf{e}_{1,2}\to\mathbf{e}_{2,1}^*, \tag{6}$$

requires that the coefficients of $|\mathbf{e}_1\cdot\mathbf{n}_2|^2$ and $|\mathbf{e}_2\cdot\mathbf{n}_1|^2$ be equal and that the coefficient of $\mathrm{Im}\,E_2$, which changes its sign under the transformations (5), vanish. Employing vector algebra, it is fairly easy to establish how the vector combinations in (2) are related to $\mathrm{Im}\,E_{1,2}$:

$$I_1=\mathrm{Im}(E_2+E_1),\quad I_2=\mathrm{Im}(E_2-E_1). \tag{7}$$

In terms of the Stokes parameters,

$$
\begin{aligned}
E_{1,2}=-\frac{\sin^2\theta}{4}\{&\xi_1^{(1)}\xi_1^{(2)}\pm\xi_2^{(1)}\xi_2^{(2)}+(1-\xi_3^{(1)})(1-\xi_3^{(2)})\\
&\times\cos\theta+i(\xi_1^{(1)}\xi_2^{(2)}\pm\xi_2^{(1)}\xi_1^{(2)})\}.
\end{aligned}
\tag{8}
$$

Note that the vector combination in (5) and the additional combination of four vectors,

$$|(\mathbf{e}_1\cdot\mathbf{n}_2)(\mathbf{e}_2\cdot\mathbf{n}_1)|^2,$$

determine the polarization structure of scattering in the general case, too (without employing the multipole expansion of the photon vector potential); the only difference is that the coefficients of the combinations become functions of the scattering angle θ. Here the Φ_i and $(\mathbf{n}_1\cdot\mathbf{n}_2)\Phi_i$ combine in such a way that the cross section contains ten terms with coefficients depending on $\omega_{1,2}$ and θ. The cross section can also be written in terms of the Stokes parameters if one employs Eq. (8) and the following relations:

$$
\begin{aligned}
|\mathbf{e}_k\cdot\mathbf{n}_i|^2=\tfrac{1}{2}(1-\xi_3^{(k)})\sin^2\theta,\quad 4|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2&=(1+\xi_3^{(1)})\\
\times(1+\xi_3^{(2)})+(1-\xi_3^{(1)})&(1-\xi_3^{(2)})\cos^2\theta\\
+2(\xi_1^{(1)}\xi_1^{(2)}+\xi_2^{(1)}\xi_2^{(2)})&\cos\theta.
\end{aligned}
$$

Here $\xi_2^{(2)}\to-\xi_2^{(2)}$ as $\mathbf{e}_2^*\to\mathbf{e}_2$.

We now give the results of quantum mechanical calculations of the scattering cross section for a freely orienting system accompanied by a transition between the states $|i\rangle\equiv|n_iJ_iM_i\rangle$ and $|j\rangle\equiv|n_fJ_fM_f\rangle$ of the same parity, with nondipole interaction effects in the first nonvanishing order taken into account. The interaction operator for incident and scattered photons of the $E1$-, $M1$-, or $E2$-type can be written as follows($\mathbf{e}=\mathbf{e}_1$ or $\mathbf{e}=\mathbf{e}_2$):[1)]

$$V=V_d+V_m+V_q, \tag{9}$$

where

$$
\begin{aligned}
&V_d=-\mathbf{d}\cdot\mathbf{e},\\
&V_m=-\boldsymbol{\mu}\cdot(\mathbf{n}\times\mathbf{e})=i\sqrt{2}\boldsymbol{\mu}\cdot\{\mathbf{n}\otimes\mathbf{e}\}_1,\\
&V_q=-\frac{i}{3}\sum_{ij}Q_{ij}e_ik_j=-\frac{i}{\sqrt{6}}Q_2\cdot\{\mathbf{n}\otimes\mathbf{e}\}_2,
\end{aligned}
$$

with $\mathbf{d}$, $\boldsymbol{\mu}$, and Q_{ij} the electric dipole, magnetic dipole and electric quadrupole operators, and $\{\mathbf{a}\otimes\mathbf{b}\}_p$ the irreducible tensor product defined in the standard way.[10] Due to the selection rules, the $E1$, $M1$, and $E2$ operators do not interfere with each other in the scattering amplitude A_{fi}, so that

$$A_{fi}=A_{fi}^{(0)}+A_{fi}^{(2)}, \tag{10}$$

where $A_{fi}^{(0)}=\langle f|\{V_d^{(2)\dagger}G_{E_i+\omega_1}V_d^{(1)}+V_d^{(1)}G_{E_i-\omega_2}V_d^{(2)\dagger}\}|i\rangle$ is the dipole scattering amplitude,[5] with the superscripts in V referring to the incident (1) and scattered (2) photons; G_E is the Green's function of the atom, and

$$A_{fi}^{(2)}=\langle f|\{(V_m^{(2)\dagger}+V_q^{(2)\dagger})G_{E_i+\omega_1}(V_m^{(1)}+V_q^{(1)})$$
$$+(V_m^{(1)}+V_q^{(1)})G_{E_i-\omega_2}(V_m^{(2)\dagger}+V_q^{(2)\dagger})\}|i\rangle \qquad (11)$$

is the nondipole part of the amplitude, which is of order α^2 compared to $A_{fi}^{(0)}$. Since we are interested only in the corrections to the cross section that contain terms of order $n_{1i}n_{2k}$, to simplify formulas we have not allowed in Eq. (9) for the interaction operators of the $M2$- or $E3$-type. Although these operators do interfere with V_d in the scattering amplitude and yield corrections of order α^2, they do not lead to terms like (1) in the cross section. The cross section with the first nondipole correction has the form

$$\frac{d\sigma}{d\Omega}=\frac{r_0^2\omega_1\omega_2^3}{2J_i+1}\sum_{M_iM_f}|A_{fi}|^2\approx\frac{d\sigma^{(0)}}{d\Omega}+\frac{d\sigma^{(2)}}{d\Omega}, \qquad (12)$$

where $d\sigma^{(0)}/d\Omega$ is specified in Eq. (4), and

$$\frac{d\sigma^{(2)}}{d\Omega}=\frac{2r_0^2\omega_1\omega_2^3}{2J_i+1}\sum_{M_iM_f}\mathrm{Re}(A_{fi}^{(0)*}A_{fi}^{(2)}). \qquad (13)$$

Since neither the $M2$ nor the $E3$ interaction, quadratic in $\mathbf{n}_i$, are taken into account, the cross section does not contain the Φ_i and $|\mathbf{e}_k\cdot\mathbf{n}_m|^2$ of (5), so that the general structure of $d\sigma^{(2)}/d\Omega$ is as follows:

$$\frac{d\sigma^{(2)}}{d\Omega}=r_0^2\omega_1\omega_2^3\left\{\sum_{i=0}^{2}a_i\Phi_i(\mathbf{n}_1\cdot\mathbf{n}_2)+b_1\mathrm{Re}\,E_1\right.$$
$$\left.+b_2\,\mathrm{Re}\,E_2+g_1\,\mathrm{Im}\,E_1+g_2\,\mathrm{Im}\,E_2\right\}. \qquad (14)$$

If the scattering amplitude is Hermitian, time-reversal symmetry leads to the invariance of the cross section under the substitutions (see Sec. 87 of Ref. 5)

$$\mathbf{n}_k\to-\mathbf{n}_k,\quad \mathbf{e}_k\to\mathbf{e}_k^*,\quad k=1,2,$$

also reversing the sign of $\mathrm{Im}\,E_{1,2}$, which means that the g parameters in Eq. (14) must vanish. Thus, the "anomalous" terms in $d\sigma/d\Omega$ are nonzero only if the amplitude A_{fi} in (10) is non-Hermitian, i.e., if among the virtual (intermediate) states through which scattering proceeds there are states in which real transitions are energy-allowed (such transitions are precisely the dissipation processes accompanying scattering, as discussed in Sec. 1).

In the simplest case the nonhermiticity of A_{fi} is related to the possibility of photoionizing an atom at $\omega_1>|E_i|$ and is due to the Green's function $G_{E_i+\omega_1+i0}$ acquiring a skew-Hermitian part at a positive energy in $A_{fi}^{(0)}$ and $A_{fi}^{(2)}$

$$G_{E_i+\omega_1+i0}\equiv\frac{1}{H_0-E_i-\omega_1-i0}$$
$$=\frac{\mathscr{P}}{H_0-E_i-\omega_1}+i\pi\delta(H_0-E_i-\omega_1), \qquad (15)$$

where H_0 is the atomic Hamiltonian and $\mathscr{P}$ denotes the Cauchy principal value. Now, if we separate both $A_{fi}^{(0)}$ and $A_{fi}^{(2)}$ in Eq. (13) into their Hermitian and skew-Hermitian parts via (15), we arrive at an expression for the "anomalous" part of the cross section, $d\sigma_{\mathrm{anom}}^{(2)}/d\Omega$, determined by the last two terms in Eq. (14):

$$\frac{d\sigma_{\mathrm{anom}}^{(2)}}{d\Omega}=\frac{2\pi r_0^2\omega_1\omega_2^3}{2J_i+1}\sum_{M_iM_f}\mathrm{Im}\sum_{JM}\{\langle i|V_d^{(1)\dagger}|EJM\rangle$$
$$\times\langle EJM|V_d^{(2)}|f\rangle A_{fi}^{(2)'}+\langle i|V_m^{(1)\dagger}+V_q^{(1)\dagger}|EJM\rangle$$
$$\times\langle EJM|V_m^{(2)}+V_q^{(2)}|f\rangle A_{fi}^{(0)'}\}. \qquad (16)$$

Here $|EJM\rangle$ is a state in the continuous energy spectrum of the atom with energy $E=E_i+\omega_1=E_f+\omega_2$, and $A_{fi}^{(k)'}$ is the Hermitian part of $A_{fi}^{(k)}$. Clearly, $d\sigma_{\mathrm{anom}}^{(2)}$ is determined by a distinctive interference of the amplitude of ionization from the states $|i\rangle$ and $|f\rangle$ to the state $|EJM\rangle$ accompanied by absorption of photons of frequencies ω_1 and ω_2, respectively, which suggests a relation between the polarization anomalies in the cross section and the presence of an open ionization channel in the atom.

The projections of angular momenta in Eq. (13) are summed by the standard methods of angular-momentum theory.[10] For the dipole amplitude, the expansion in irreducible parts has the well-known form

$$A_{fi}^{(0)}=\sum_{pm}C_{pmJ_fM_f}^{J_iM_i}\alpha_p(\omega_1,\omega_2)\{\mathbf{e}_1\otimes\mathbf{e}_2^*\}_{pm}, \qquad (17)$$

where

$$\alpha_p(\omega_1,\omega_2)$$
$$=(-1)^{J_i+J_f}\sqrt{\frac{2p+1}{2J_i+1}}\sum_J\begin{Bmatrix}1&1&p\\J_i&J_f&J\end{Bmatrix}\langle n_fJ_f\|\mathbf{d}\cdot\{G_{E_i+\omega_1}^J$$
$$+(-1)^pG_{E_i-\omega_2}^J\}\mathbf{d}\|n_iJ_i\rangle \qquad (18)$$

are the invariant amplitudes determining the cross sections of scalar, skew-symmetric, and symmetric scattering in Eq. (4), with

$$G^p=\omega_1\omega_2^3|\alpha_p(\omega_1,\omega_2)|^2, \qquad (19)$$

$C_{b\beta c\gamma}^{a\alpha}$ and $\{^{abc}_{def}\}$ are the Clebsch–Gordan coefficients and the Wigner $6j$ symbols, and G_E^J is the partial Green's function corresponding to the angular momentum J, whose value is determined by the "triangle rule" for the $6j$ symbol in α_p.

If we allow for (9) and write the operator $V_m^{(k)}+V_q^{(k)}$ in the form

$$V_m^{(k)}+V_q^{(k)}=i\sqrt{2}\sum_{\alpha=1,2}\sum_m(-1)^m\{\mathbf{n}_k\otimes\dot{\mathbf{e}}_k\}_{\alpha,-m}\hat{T}_{\alpha m}^{(k)},$$

where

$$\hat{T}_1^{(k)}=\boldsymbol{\mu},\quad \hat{T}_2^{(k)}=\frac{(-1)^k\alpha\omega_k}{\sqrt{12}}Q_2,$$

we arrive at the expansion of $A_{fi}^{(2)}$ in the irreducible parts similar to Eqs. (17) and (18):

$$A_{fi}^{(2)}=2\sum_{\alpha,\beta=1}^{2}\sum_{pm}(2p+1)C_{pmJ_fM_f}^{J_iM_i}B_p^{\alpha\beta}$$

$$\times\{\{\mathbf{n}_1\otimes\mathbf{e}_1\}_\alpha\otimes\{\mathbf{n}_2\otimes\mathbf{e}_2^*\}_\beta\}_{pm},$$

$$B_p^{\alpha\beta}(\omega_1,\omega_2)=\frac{(-1)^{J_i+J_f}}{\sqrt{(2p+1)(2J_i+1)}}\sum_J\left[\begin{Bmatrix}\alpha\beta p\\ J_fJ_iJ\end{Bmatrix}\right.$$

$$\times\langle fJ_f\|\hat{T}_\beta^{(2)}G^J_{E_i+\omega_1}\hat{T}_\alpha^{(1)}\|iJ_i\rangle$$

$$+(-1)^{\alpha+\beta+p}\begin{Bmatrix}\beta\alpha p\\ J_fJ_iJ\end{Bmatrix}$$

$$\left.\times\langle fJ_f\|\hat{T}_\alpha^{(1)}G^J_{E_i-\omega_2}\hat{T}_\beta^{(2)}\|iJ_i\rangle\right]. \qquad (20)$$

Clearly, the amplitude contains 14 independent invariant parameters $B_p^{\alpha\beta}$ three for $M1-M1$, five for $E2-E2$ scattering, and six "mixed" terms. Only ten of these with $p=0,1,2$ at $\alpha=\beta$ and $p=1,2$ at $\alpha\neq\beta$ contribute to the cross section (13):

$$\frac{d\sigma^{(2)}}{s\Omega}4r_0^2\omega_1\omega_2^3\sum_{\alpha,\beta}\sum_p\mathrm{Re}\{\alpha_p^*(\omega_1,\omega_2)B_p^{\alpha\beta}$$

$$\times(\omega_1,\omega_2)(\{\mathbf{e}_2\otimes\mathbf{e}_1^*\}_p\cdot\{\{\mathbf{n}_1\otimes\mathbf{e}_1\}_\alpha\otimes\{\mathbf{n}_2\otimes\mathbf{e}_2^*\}_\beta\}_p)\}. \qquad (21)$$

Having employed the rules of correspondence of spherical and Cartesian components of tensors and carried out some lengthy calculations, we can write the tensor products of the six vectors in (21) in terms of combinations of ordinary scalar products defined in (5). One such relation is

$$(\{\mathbf{e}_2\otimes\mathbf{e}_1^*\}_2\cdot\{\{\mathbf{n}_1\otimes\mathbf{e}_1\}_2\otimes\{\mathbf{n}_2\otimes\mathbf{e}_2^*\}_2\}_2)$$

$$=-\tfrac{1}{4}\sqrt{\tfrac{3}{7}}[(1+|\mathbf{e}_1\cdot\mathbf{e}_2|^2-\tfrac{4}{3}|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2)(\mathbf{n}_1\cdot\mathbf{n}_2)$$

$$-\tfrac{10}{9}\,\mathrm{Re}\,E_1+2\,\mathrm{Re}\,E_2+i\tfrac{14}{9}\,\mathrm{Im}\,E_1]. \qquad (22)$$

At this point it is convenient to introduce special notation for the products $\alpha_p^*B_p^{\alpha\beta}$ in (21) corresponding to the contribution of magnetic (M_p) and quadrupole (Q_p) effects and their interference ($C_p^\pm$):

$$M_p=\alpha_p^*B_p^{11},\quad Q_0=\sqrt{\tfrac{3}{5}}\,\alpha_0^*B_0^{22},$$

$$Q_1=\tfrac{1}{\sqrt{5}}\,\alpha_1^*B_1^{22},\quad Q_2=\sqrt{\tfrac{3}{7}}\,\alpha_2^*B_2^{22},$$

$$C_1^\pm=\sqrt{\tfrac{3}{5}}\,\alpha_1^*(B_1^{21}\pm B_1^{12}),\quad C_2^\pm=\tfrac{1}{\sqrt{3}}\,\alpha_2^*(B_2^{21}\pm B_2^{12}).$$

In terms of these quantities the coefficients in Eq. (14) acquire the form

$$a_0=-\mathrm{Re}(M_1-M_2-C_1^--C_2^-+Q_1+Q_2),$$

$$a_1=-\tfrac{2}{3}\,\mathrm{Re}(M_0+2M_2+Q_0-2Q_2),$$

$$a_2=\mathrm{Re}(M_1+M_2-C_1^-+C_2^-+Q_1-Q_2),$$

$$b_1=\mathrm{Re}\,z,\quad b_2=-2\,\mathrm{Re}(M_1+M_2-Q_1+Q_2), \qquad (23)$$

$$g_1=-\mathrm{Im}\,z,\quad g_2=2\,\mathrm{Im}(C_1^+-C_2^+),$$

$$z=M_1+M_2+C_1^--C_2^-+Q_1-\tfrac{2}{9}\,Q_2$$

$$+\tfrac{2}{3}(M_0^*+2M_2^*-Q_0^*+2Q_2^*).$$

The general formulas (14) and (23) simplify when the angular momenta J_i and J_f of the levels $|i\rangle$ and $|f\rangle$ are small, e.g., at $J_i=0$ and $J_i=\frac{1}{2}$. If fine structure is ignored, the $M1-M1$ terms (with $\alpha=\beta=1$) vanish for transitions between states with different angular momenta ($l_i\neq l_f$) and at $l_i=l_f=0$. The formulas for $d\sigma/d\Omega$, written in terms of the radial matrix elements with the optical–electron Green's function $g_l(E;r,r')$ for the two most interesting cases, are

(a) $s-s$ scattering ($l_i=l_f=0$):

$$\frac{d\sigma}{d\Omega}=r_0^2\{\sigma|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2+\sigma^s[\mathrm{Re}\,E_1+|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2(\mathbf{n}_1\cdot\mathbf{n}_2)]$$

$$+\sigma^a\,\mathrm{Im}\,E_1\}, \qquad (24)$$

where

$$\sigma^0=\tfrac{1}{9}\omega_1\omega_2^3|A_{00}^{(1)}|^2,$$

$$\sigma^s=-\frac{\alpha^2}{90}\,\omega_1^2\omega_2^4\,\mathrm{Re}(A_{00}^{(1)*}A_{00}^{(2)}),$$

$$\sigma^a=\frac{\alpha^2}{90}\,\omega_1^2\omega_2^4\,\mathrm{Im}(A_{00}^{(1)*}A_{00}^{(2)}),$$

with $A_{l_fl_i}^{(k)}$ the radial dipole and quadrupole scattering amplitudes,

$$A_{l_fl_i}^{(k)}=\langle n_fl_f|r^k\{g_k(E_i+\omega_1)+g_k(E_i-\omega_2)\}r'^k|n_il_i\rangle,$$

determining at $|i\rangle=|f\rangle$ the dipole and quadrupole polarizabilities of the $|i\rangle$ state.

(b) $s-d$ scattering ($l_i=0$ and $l_f=2$). In this case the following three atomic parameters are nonzero:

$$B_2^{12}=-\frac{\alpha^2\omega_2}{10\sqrt{2}\omega_2}\langle r^2\rangle,\quad B_2^{21}=\frac{\alpha^2\omega_1}{10\sqrt{2}\omega_2}\langle r^2\rangle,$$

$$B_2^{22}=\frac{\alpha^2\omega_1\omega_2}{30\sqrt{14}}A_{20}^{(2)},\quad \langle r^2\rangle=\langle n_f,l_f=2|r^2|n_i,l_i=0\rangle.$$

In $d\sigma^{(0)}/d\Omega$ only the symmetric-scattering amplitude, $\alpha_2=\sqrt{2/3}A_{20}^{(1)}$, is nonzero. In view of the unwieldiness of the general expression, only the expression for the cross section with the nondipole correction containing I_1 and I_2 is given below:

$$\frac{d\sigma}{d\Omega}=r_0^2\omega_1\omega_2^3\{\tfrac{1}{10}|\alpha_2|^2(1+|\mathbf{e}_1\cdot\mathbf{e}_2|^2-\tfrac{2}{3}|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2)$$

$$+\psi(\omega_1,-\omega_2)I_1-\psi(-\omega_2,\omega_1)I_2\}, \qquad (25)$$

where

TABLE I. Frequency dependence of the $1s-2s$ and $1s-4s$ cross sections for the hydrogen atom (numbers in parentheses denotepowersoften:$(-n)=10^{-n}$).

	$1s-2s$			$1s-4s$		
$\omega/\lvert E_{1s}\rvert$	σ^0	σ^s	σ^a	σ^0	σ^s	σ^a
1.01	2.17(−1)	−6.59(−7)	−3.90(−6)	3.57(−2)	1.97(−9)	−3.47(−7)
2	2.47(−2)	1.69(−7)	−3.96(−6)	3.27(−3)	−9.58(−8)	−2.87(−7)
6	6.71(−4)	7.74(−6)	−5.83(−6)	8.54(−5)	5.34(−7)	−4.12(−7)
10	1.15(−4)	1.09(−5)	−6.04(−6)	1.45(−5)	7.83(−7)	−4.37(−7)
20	9.71(−6)	1.44(−5)	−5.66(−6)	1.22(−6)	1.06(−6)	−4.16(−7)

$$\psi(\omega_1,-\omega_2)=\frac{\alpha^2}{60}\left[\frac{4}{21}\,\omega_1\omega_2\,\mathrm{Im}(A_{20}^{(1)*}A_{20}^{(2)})+\left(3\frac{\omega_1}{\omega_2}-\frac{\omega_2}{\omega_1}\right)\langle r^2\rangle\mathrm{Im}\,A_{20}^{(1)*}\right].$$

Because of the presence of $M1-E2$ terms $B_2^{\alpha\beta}$, the coefficients of I_1 and I_2 in Eq. (25) prove to be different, in contrast to Eq. (24).

To give an idea of the magnitude of the asymmetry effects, Table I lists the scattering cross sections for the $1s-2s$ and $1s-4s$ transitions in the hydrogen atom, calculated by using the Coulomb Green's function. As expected, for $\omega\gtrsim|E_i|$ both σ^a and σ^s are small compared to the dipole cross section σ^0, but at $\omega\sim 20|E_i|$ the magnitude of $\Delta(d\sigma/d\Omega)$ is comparable to $d\sigma/d\Omega$. For the case of the ground state of hydrogen considered here such frequencies already correspond to the x-ray region, but for the highly excited atomic levels the asymmetry can be significant in the optical range, too. Note also that when $\omega\gg|E_i|$, the numerical calculations must allow for higher multiplicity effects and for relativistic corrections, which by themselves do not lead to asymmetry but can change the numerical value of the cross sections.

3. SCATTERING ASYMMETRY DUE TO RADIATIVE CORRECTIONS

When $\hbar\omega_1<|E_i|$, the amplitude A_{fi} in (12) is Hermitian and anomalies can arise only if the radiative corrections to scattering are taken into account. To lowest order in α, these corrections are represented by the Feynman diagrams in Fig. 1 (Sec 40 in Ref. 11), to which diagrams with interchanged photon lines ω_1 and ω_2 must be added. The skew-Hermitian part of the radiative-correction amplitude A_{fi}^{rad} is determined by the poles of the diagrams, and corresponds to virtual transitions to the energy-allowed state $|n\rangle$. It can be expressed in terms of the product of two ordinary transition amplitudes from the $|i\rangle$ and $|f\rangle$ states to the intermediate bound state $|n\rangle$. For instance, for the diagram (d) in Fig. 1, the pole has the form depicted in Fig. 2 ($\hbar\omega_n=E_i+\hbar\omega_1-E_n$). The prime on the sum over n in Fig. 2 means that only the $|n\rangle$ states with $\omega_n>0$ are taken into account. The x's in Fig. 1 designate the places where the diagrams must be dissected to obtain the pole terms. Dissecting the horizontal electron lines yields the contribution of virtual Raman-scattering processes of the type depicted in Fig. 2. At $|i\rangle=|f\rangle$ the same pole parts also determine the skew-Hermitian part of the polarizability tensor for $\hbar\omega_1<|E_i|$ calculated in Ref. 12. For $|i\langle\neq|f\rangle$ the dissections of the lateral electron lines in the diagrams (a) and (b) in Fig. 1 and in the diagrams with interchanged photon lines must also be taken into account. These dissections yield the contribution of spontaneous hyper-Raman scattering processes ($E_i+\hbar(\omega_1-\omega_2-\omega_{n'})\to E_{n'}$), with emission of an additional photon $\hbar\omega_{n'}$ and transition to a virtual state $|n'\rangle$ with $E_{n'}<E_F$, followed by the transition $|n'\rangle\to|f\rangle$ with absorption of a photon of frequency $\omega_{n'}=\omega_1-\omega_2+(E_i-E_{n'})/\hbar$. The states $|n\rangle$ and $|n'\rangle$ belong to different sets and have opposite parities. The polarization and direction of propagation of the photons ω_n and $\omega_{n'}$ are not fixed and are summed over.

Our problem does not require a full QED calculation of A_{fi}^{rad} since the regular part of A_{fi}^{rad} does not lead to asymmetry and provides only small corrections to $d\sigma^{(0)}/d\Omega$. On the other hand, the pole parts of the Feynman diagrams, as is known,[5] require no renormalization and, according to what has been said earlier, can easily be written explicitly. The asymmetry in scattering is described by the second term in the cross section

$$\frac{d\sigma}{d\Omega}=\frac{\omega_1\omega_2^3}{c^4(2J_i+1)}\sum_{M_iM_j}\{|A_{fi}|^2+2\,\mathrm{Re}(A_{fi}^{(2)*}A_{fi}^{\mathrm{rad}})\},\quad(26)$$

which can be obtained from (12) by adding A_{fi}^{rad} to the am-

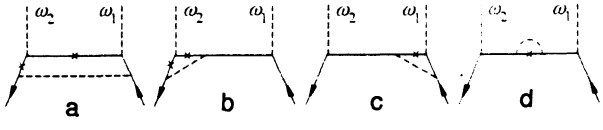

FIG. 1.

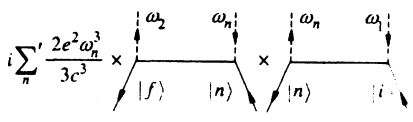

FIG. 2.

plitude $A_{fi}=A_{fi}^{(0)}+A_{fi}^{(2)}$. As the expression in Fig. 2 shows, asymmetry has the additional smallness of order α^3 compared to the case where $\hbar\omega_1>|E_i|$. This is due to the smallness of the radiation widths of the atomic levels.

Thus, the corrections are generally small and only point to the important fact that polarization anomalies in the scattering are caused by radiative reaction. An exception is the scattering at frequencies $\omega_1=\omega_t$ (the transparency frequencies), at which the dipole amplitude $A_{fi}^{(0)}$ vanishes. Such frequencies always exist, for example, in $s-s$ and $s-d$ scattering, for which $A_{fi}^{(0)}$ is the only atomic parameter α_p in Eq. (18) (α_0 or α_2) that changes sign in each interval between resonances $|E_i-E_{n,l=1}|<\hbar\omega_1<|E_i-E_{n+1,l=1}|$ (see Ref. 13). At $\omega_1\simeq\omega_t$ the main scattering is the quadrupole, determined by the amplitude $A_{fi}^{(2)}$ in Eq. (20) with $\alpha=\beta=2$ and $p=J_f$, and radiative correction effects could be observed.

For $s-s$ scattering at $\omega_1\approx\omega_t$, the scattering cross section (26) assumes the form

$$\frac{d\sigma}{d\Omega}=\frac{\omega_1\omega_2^3}{c^4}\left\{A_Q^2|(\mathbf{e}_1\cdot\mathbf{e}_2^*)(\mathbf{n}_1\cdot\mathbf{n}_2)+(\mathbf{e}_1\cdot\mathbf{n}_2)(\mathbf{e}_2^*\cdot\mathbf{n}_1)|^2+\frac{2e^4A_Q}{27c^3}(\Pi^{\text{Raman}}+\Pi^{\text{hyper-Raman}})\operatorname{Im}E_1\right\}. \tag{27}$$

Here

$$A_Q=\frac{e^2\omega_1\omega_2}{60c^2}\langle f|r^2\{g_2(E_i+\hbar\omega_1)+g_2(E_i-\hbar\omega_2)\}r'^2|i\rangle$$

is the radial part of the quadrupole scattering amplitude, Π^{Raman} and $\Pi^{\text{hyper-Raman}}$ are the pole parts of A_{fi}^{rad} corresponding to spontaneous Raman and hyper-Raman processes such as

$$\Pi^{\text{Raman}}=\sum_n{}^{\cdot}\sum_{L=0,2}(L+2)\left(\frac{E_i-E_{nL}}{\hbar}+\omega_1\right)^3\times\langle f|r[g_1(E_i+\hbar\omega_1)+g_1(E_{nL}-\hbar\omega_2)]r'|nL\rangle\times\langle nL|r[g_1(E_{nL}-\hbar\omega_1)+g_1(E_i+\hbar\omega_1)]r'|i\rangle,$$

$$\Pi^{\text{hyper-Raman}}=\sum_{n'}{}'\sum_{l=0,2}(l+2)\left(\frac{E_i-E_{n',L=1}}{\hbar}+\omega_1-\omega_2\right)^3\times\langle f|r|n',L=1\rangle\langle n',L=1|r\{g_l(E_{n'}+\hbar\omega_2)\times rg_1(E_i+\hbar\omega_1)+g_l(E_{n'}-\hbar\omega_1)rg_1\times(E_i-\hbar\omega_2)+[g_l(E_{n'}-\hbar\omega_1)+g_l\times(E_{n'}+\hbar\omega_2)]rg_1(E_{n'}+\hbar\omega_2-\hbar\omega_1)\}r|i\rangle.$$

As Eq. (27) clearly shows, the asymmetric part of $d\sigma$ has a smallness of only first order in α compared to the symmetric part (quadrupole in the case at hand) of the scattering. For the ground state of hydrogen the transparency frequencies lie in the UV range, and for alkali atoms in the optical range. For instance, for elastic 1s\–1s scattering in hydrogen we have (in atomic units)[13]

$$\omega_t^{(1)}=0.429538,\quad \omega_t^{(2)}=0.463438,\ \ldots.$$

The atomic parameters exhibit strong dispersion dependence. Allowing for the more complicated resonance structure of Π^{Raman} and $\Pi^{\text{hyper-Raman}}$ than that of A_Q and for the numerical factors in (27), one can expect the asymmetry effects at $\omega_1\approx\omega_t$ to amount to several percent.

4. ASYMMETRY IN RESONANT SCATTERING

As ω_1 approaches the resonant transition frequency E_r-E_i, it becomes impossible to use perturbation theory techniques to take radiative corrections into account, and radiative reaction is allowed for by introducing the radiative width Γ of the resonant level: $E_r\to E_r-i\Gamma/2$. In the resonant case it also proves possible to compensate for the smallness of order α^2 in $d\sigma$ caused by the fact that the interaction is nondipole. To this end one must use the resonance on the dipole-forbidden $|i\rangle$–$|r\rangle$ transition, in which $A_{fi}^{(2)}$ becomes comparable in magnitude to the nonresonant dipole amplitude $A_{fi}^{(0)}$.

If there is resonance, one can retain in the amplitude $A_{fi}^{(2)}$ specified by Eq. (12) only the resonant term in the Green's function $G_{E_i+\omega_1}$:

$$A_{fi}^{(2)}=\frac{A_{fi}^{\text{res}}}{\Delta+i\Gamma/2},\quad \Delta=E_i+\omega_1-E_r,$$
$$A_{fi}^{\text{res}}=\sum_{M_r}\langle f|V_m^{(2)\dagger}+V_q^{(2)\dagger}|n_rJ_rM_r\rangle\times\langle n_rJ_rM_r|V_m^{(1)}+V_q^{(1)}|i\rangle. \tag{28}$$

Here for the sake of simplicity we have assumed that the resonant level $|r\rangle\equiv|n_rJ_rM_r\rangle$ with $E_r>E_i$ has no multiplet structure (or that the detuning Δ is much smaller than the multiplet splitting). Combining (13) with (28), we arrive at the following expression for the "anomalous part" of the cross section [cf. Eq. (16)]:

$$\frac{d\sigma_{\text{anom}}}{d\Omega}=\frac{r_0^2\omega_1\omega_2^3}{2J_i+1}\sum_{M_iM_f}\operatorname{Im}(A_{fi}^{(0)}A_{fi}^{\text{res}*})\frac{\Gamma}{\Delta^2+\Gamma^2/4}. \tag{29}$$

The angular structure of $A_{fi}^{(0)}$ and A_{fi}^{res} has the same form (17) and (20) as in the nonresonant case, so that after separation of the angular parts, Eq. (29) is reduced to the last two terms on the right-hand side of Eq. (14). It is very clear that the change in sign of $\operatorname{Im}E_{1,2}$ under time reversal discussed in Sec. 2 is balanced by the reversal of sign in the width Γ, which by its very meaning is a T-odd parameter.

The coefficients in Eq. (14) are specified in (23) with obvious changes in the reduced matrix elements following from (28). For one thing, all the coefficients except g_1 and g_2 contain the factor $\Delta/(\Delta^2+\Gamma^2/4)$. In the resonant case one must also allow for purely multipole scattering,

$$\frac{d\sigma^{(4)}}{d\Omega}=\frac{r_0^2\omega_1\omega_2^3}{(2J_i+1)(\Delta^2+\Gamma^2/4)}\sum_{M_iM_j}|A_{fi}^{\text{res}}|^2, \tag{30}$$

which is no longer small. As a result, the angular structure of the resonant scattering cross section incorporates all ten vector combinations discussed in Sec. 2 in relation to the general case.

Because the general formulas for the coefficients in Eq. (30) are cumbersome, we write only the formula for the total resonant scattering cross section with $l_i=l_j=0$. Here only a quadrupole resonance with the d-level of the state $|r\rangle$ is possible, and the cross section has the form

$$\frac{d\sigma}{d\Omega}=\frac{r_0^2\omega_2\omega_2^3}{3}\left\{|\alpha_0|^2|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2+\frac{Q_{\rm res}^2}{\Delta^2+\Gamma^2/4}[|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2\cos^2\theta\right.$$
$$+2\ \mathrm{Re}\ E_1\cos\theta+|\mathbf{e}_1\cdot\mathbf{n}_2|^2|\mathbf{e}_2\cdot\mathbf{n}_1|^2]+\frac{2\alpha_0 Q_{\rm res}}{\Delta^2+\Gamma^2/4}$$
$$\left.\times[\Delta(\mathrm{Re}\ E_1+|\mathbf{e}_1\cdot\mathbf{e}_2|^2\cos\theta)-\Gamma\ \mathrm{Im}\ E_1]\right\},\qquad(31)$$

where $\alpha_0=\frac{1}{3}A_{00}^{(1)}$ [see Eq. (24)], and

$$Q_{\rm res}=\frac{\alpha^2\omega_1\omega_2}{20\sqrt{3}}\langle f|r^2|d\rangle\langle d|r^2|i\rangle.$$

The asymmetry effects become especially important when the dipole and quadrupole scattering amplitudes become equal and all the terms in (31) are of the same order of magnitude, i.e., at

$$\Gamma\sim\frac{Q_{\rm res}}{\alpha_0}\sim\alpha^2|E_i|.\qquad(32)$$

As is known, radiation widths are of order $\alpha^3|E_i|$, smaller than Γ in (32). However, the necessary value of Γ can easily be obtained if we allow for collisional broadening. Taking collision broadening into account consistently requires using the density matrix of the resonant levels. Without going into detail, we only note that here the scattering cross section will be a structure similar to (31), where Γ must be interpreted as the transverse width, which determines the relaxation rate of the off-diagonal elements of the density matrix. Next, the quadrupole scattering cross section $d\sigma^{(4)}/d\Omega$ [the second term on the right-hand side of Eq. (31)] also contains longitudinal widths, which determine the damping rate of the statistical tensors corresponding to the magnetic sublevels of the state $|r\rangle$. Finally, in addition to describing resonant scattering, $d\sigma^{(4)}/d\Omega$ describes collision-induced resonance fluorescence, which leads to a spread in the scattered photon frequency by a quantity of order Δ.

5. DIPOLE-FORBIDDEN SCATTERING

Now let us discuss the scattering accompanied by a transition between the levels $|i\rangle$ and $|f\rangle$ of opposite parities forbidden in the electric dipole approximation. For states with $|J_i-J_f|\equiv\Delta J\leqslant 3$, the amplitude of such scattering is determined by the interference of the $E1$- and $(M1+W2)$-interaction. Hence the cross section has a smallness of order α^2 compared to $d\sigma^{(0)}/d\Omega$, as the cross section defined in Eq. (13) has, but contrary to the latter the angular structure incorporates not only the products $n_{1i}n_{2k}$ but also the bilinear combinations $n_{1i}n_{1k}$ and $n_{2i}n_{2k}$. As a result the dipole-forbidden scattering cross section generally incorporates all the vector combinations specified in Eqs. (5) with coefficients depending only on frequencies:

$$\frac{d\sigma}{d\Omega}=r_0^2\omega_1\omega_2^3\left\{\sum_{i=0}^{2}[A_i+B_i(\mathbf{n}_1\cdot\mathbf{n}_2)]\Phi_i+C_1|\mathbf{e}_1\cdot\mathbf{n}_2|^2\right.$$
$$\left.+C_2|\mathbf{e}_2\cdot\mathbf{n}_1|^2+D_1\ \mathrm{Re}\ E_1+D_2\ \mathrm{Re}\ E_2+F_1I_1+F_1I_2\right\}.\qquad(33)$$

To achieve a higher symmetry in the coefficients we have used the combinations of the I_1 and I_2 of Eqs. (2) rather than the Im E_1 and Im E_2 of Eqs. (7). As before, when the amplitude has a skew-Hermitian part, $F_{1,2}\neq 0$. For one thing, when $\omega_1>|E_i|$, we can write an expression like Eq. (16) for the "anomalous part" of the cross section. But now all the parameters A_i-F_i in Eq. (33) are of the same order of magnitude and the asymmetry effects contain no smallness parameter for all $\omega_1>|E_i|$.

Expanding the scattering amplitude in irreducible parts as in Sec. 2, we can represent the cross section in an invariant form containing no projections of the angular momenta J_i and J_f:

$$\frac{d\sigma}{d\Omega}=2r_0^2\omega_1\omega_2^3\sum_p\sum_{\alpha_1\alpha_2}\{X_p^{(\alpha_1)}X_p^{(\alpha_2)*}$$
$$\times(\{\{\mathbf{n}_1\otimes\mathbf{e}_1\}_{\alpha_1}\otimes\mathbf{e}_2^*\}_p\cdot\{\mathbf{e}_2\otimes\{\mathbf{e}_1^*\otimes\mathbf{n}_1\}_{\alpha_2}\}_p)$$
$$+2\ \mathrm{Re}[X_p^{(\alpha_1)}Y_p^{(\alpha_2)*}(\{\{\mathbf{n}_1\otimes\mathbf{e}_1\}_{\alpha_1}\otimes\mathbf{e}_2^*\}_p$$
$$\times\{\mathbf{e}_1^*\otimes\{\mathbf{e}_2\otimes\mathbf{n}_2\}_{\alpha_2}\}_p)]+Y_p^{(\alpha_1)}Y_p^{(\alpha_2)*}$$
$$\times(\{\{\mathbf{n}_2\otimes\mathbf{e}_2^*\}_{\alpha_1}\otimes\mathbf{e}_1\}_p\cdot\{\mathbf{e}_1^*\otimes\{\mathbf{e}_2\otimes\mathbf{n}_2\}_{\alpha_2}\}_p)\},\qquad(34)$$

where

$$X_p^{(\alpha)}\equiv X_p^{(\alpha)}(\omega_1,-\omega_2)=\frac{1}{\sqrt{2J_i+1}}\sum_J\left[\begin{Bmatrix}\alpha\ 1\ p\\ J_fJ_iJ\end{Bmatrix}\right.$$
$$\times\langle fJ_f\|\mathbf{d}G^J_{E_i+\omega_1}\hat{T}_\alpha\|iJ_i\rangle+(-1)^{\alpha+1-p}\begin{Bmatrix}1\ \alpha p\\ J_fJ_iJ\end{Bmatrix}$$
$$\left.\times\langle fJ_f\|\hat{T}_\alpha G^J_{E_i-\omega_2}\mathbf{d}\|iJ_i\rangle\right],\qquad(35)$$

$$\hat{T}_1=\boldsymbol{\mu},\quad \hat{T}_2=-\frac{\alpha\omega_1}{\sqrt{12}}Q_2,\quad Y_p^{(\alpha)}\equiv X_p^{(\alpha)}(-\omega_2,\omega_1).$$

Calculating the tensor products in Eq. (34) by employing relations of type (22), we arrive at explicit expressions for the coefficients A_i-F_i in terms of bilinear combinations of X and Y. The corresponding formulas are given in the Appendix. Together with (33) they completely solve the problem of the polarization–angular dependence of the cross section of dipole-forbidden scattering by a freely orienting system.

These results serve as a direct generalization of Placzek's theory of $E1-E1$ scattering,[5] which was precisely based on the expansion (17) for $A_{fi}^{(0)}$. In the case at hand the initial amplitude A_{fi} contains four partial amplitudes corresponding to absorption of a dipole ($E1$) photon with emission of a photon of the $M1$ or $E2$ type $(Y^{(1)},Y^{(2)})$ and, vice

TABLE II.

$\omega/\lvert E_i\rvert$	Atom	a_1	a_2	a_3	a_4	b_1	b_2
1.02	H	−2.24(−9)	8.13(−10)	1.86(−9)	1.44(−10)	3.77(−6)	−3.06(−6)
	Cs	−3.40(−11)	7.70(−11)	4.08(−10)	3.69(−12)	7.70(−7)	−4.38(−7)
1.1	H	−1.17(−9)	4.30(−10)	8.76(−10)	1.01(−10)	4.25(−6)	−3.22(−6)
	Cs	1.99(−10)	−5.18(−11)	5.91(−10)	2.49(−12)	8.58(−7)	−5.32(−7)
1.6	H	−8.36(−11)	5.52(−11)	7.74(−11)	1.17(−11)	5.90(−6)	−3.30(−6)
	Cs	9.55(−10)	−2.19(−9)	2.43(−9)	5.04(−10)	1.45(−6)	−9.77(−6)
2.0	H	−2.29(−11)	1.70(−11)	2.59(−11)	3.20(−12)	6.45(−6)	−3.02(−6)
	Cs	1.03(−9)	−5.29(−9)	4.66(−9)	1.51(−9)	2.04(−6)	−1.18(−6)

versa, absorption of an $M1$ or $E2$ photon with emission of an $E1$ photon ($X^{(1)}$,$X^{(2)}$). As was the case with $A_{fi}^{(0)}$, each can be expanded in three irreducible parts with $p=\alpha,\alpha\pm1$, given by Eqs. (35) and similar to α_p in (18). As a result the cross section is determined by 12 invariant amplitudes $X_p^{(\alpha)}$ and $Y_p^{(\alpha)}$ this number is equal to the number of coefficients in (33). Owing to interference, the relation between X,Y and A_i-F_i is even more complicated than between α_p and G^p in (19).

As the formulas in the Appendix imply, the parameters $F_{1,2}\equiv F_{1,2}(\omega_1,-\omega_2)$ in (35) transform into each other under permutation of arguments $[F_2(\omega_1,-\omega_2)=-F_1(-\omega_2,\omega_1)]$ and determine the asymmetric part $\Delta\sigma$ of the cross section in Eq. (1) with $f(\omega_1,-\omega_2)=2F_1(\omega_1,-\omega_2)$. The coefficients $F_{1,2}$ are determined by the imaginary part of the product $X_p^{(\alpha)}Y_p^{(\beta)*}$ of amplitudes. In forward scattering, which may be of interest, for example, in connection with stimulated Raman scattering, the asymmetry effects disappear and the polarization dependence of the cross section is determined by the same three combinations Φ_i as for dipole scattering [Eq. (4)] with coefficients A_i+B_i. At $\theta=\frac{1}{2}\pi$ the asymmetry effects are at their maximum and, as Eq. (8) implies, the cross section contains terms proportional to $\xi_1^{(1)}\xi_1^{(2)}$ and $\xi_2^{(1)}\xi_2^{(2)}$, which vanish in the dipole-allowed scattering through an angle of 90°. Asymmetry disappears after integrating over θ, and the total cross section σ, when integrated with respect to θ, summed over the scattered-photon polarizations of the scattered photon, and averaged over the incident-photon polarizations, assume the form

$$\sigma=\frac{8\pi r_0^2\omega_1\omega_2^3}{3}\sum_{p=\alpha,\alpha\pm1}\sum_{\alpha=1,2}\frac{2p+1}{2\alpha+1}\sigma_p^{(\alpha)}. \tag{36}$$

Here the $\sigma_p^{(\alpha)}=|X_p^{(\alpha)}|^2+|Y_p^{(\alpha)}|^2$ are the partial cross sections of $E1-M1$ and $E1-E$ scattering (at $\alpha=1$ and 2, respectively) similar to the invariant cross sections G^p in (4), which determine the total cross section of $E1-E1$ scattering,

$$\sigma^{(0)}=\frac{8\pi}{9}r_0^2\omega_1\omega_2^3\sum_{p=0,1,2}G^p.$$

The selection rules for scattering are specified by the $6j$ symbols in the $X_p^{(\alpha)}$. For instance, at $J_i=J_f=\frac{1}{2}$ the parameters $X_1^{(2)}$, $X_0^{(1)}$, and $X_1^{(1)}$ are nonzero, and at $J_i=0$ only the $X_p^{(\alpha)}$ with $p=J_i$ are nonzero. Of special interest is the scattering cross section for a transition with $\Delta J=0$, since one-photon transitions between such states are highly forbidden and light scattering may prove effective in studying the properties of levels with high angular momenta J_f. The cross section in this case is expressed solely in terms of the parameters $X_3^{(2)}\equiv X$ and $Y_3^{(2)}\equiv Y$ although it does contain all the vector combinations of Eq. (33):

$$\begin{aligned}\frac{d\sigma}{d\Omega}=&\tfrac{1}{15}r_0^2\omega_1\omega_2^3\{[|X|^2+|Y|^2+2\ \mathrm{Re}(XY^*)\cos\theta]\\&\times(5|\mathbf{e}_1\cdot\mathbf{e}_2|^2-2|\mathbf{e}_1\cdot\mathbf{e}_2^*|^2+5)+3|X|^2|\mathbf{e}_2\cdot\mathbf{n}_1|^2\\&+3|Y|^2|\mathbf{e}_1\cdot\mathbf{n}_2|^2+2\ \mathrm{Re}(XY^*)(3\ \mathrm{Re}\,E_2-4\ \mathrm{Re}\,E_1)\\&+7\ \mathrm{Im}(XY^*)(I_1+I_2)\}.\end{aligned} \tag{37}$$

Note that in this example $F_2=F_1$. This follows from the relation

$$F_1(\omega_1,-\omega_2)=-F_1(-\omega_2,\omega_1),$$

which is valid in the lack of interference of $M1$ and $E2$ interactions but invalid in the general case.

Table II lists the results of calculations of the parameters [(see Eq. (37)]

$$a_1=\tfrac{7}{15}\omega_1\omega_2^3\ \mathrm{Im}(XY^*),\quad a_2=\tfrac{2}{15}\omega_1\omega_2^3\ \mathrm{Re}(XY^*),$$

$$a_3=\tfrac{1}{15}\omega_1\omega_2^3|X|^2,\quad a_4=\tfrac{1}{15}\omega_2\omega_2^3|Y|^2$$

for the transition from the ground state to the first excited f state in hydrogen atoms ($1s-4f$) and cesium atoms ($6s-6f$). The frequencies are given in units of the ionization potential of the initial state (for cesium $|E_{6s}|=31406\ \mathrm{cm}^{-1}$). The table shows that the "anomalous" terms are of the same order of magnitude as the other terms in the cross section. Note the different nature of the frequency dependence of the cross sections for hydrogen and cesium: in the chosen frequency range the cross section for hydrogen decreases while that for cesium increases with frequency.

For the $J_i=0\to J_f=1$ transition the coefficients A_0, B_0, A_2, and B_2 vanish, while the rest can be expressed simply in terms of the parameters introduced in the Appendix:

$$R_1^{\pm}(\omega_1,-\omega_2)=\sqrt{\tfrac{3}{5}}X_1^{(2)}\pm X_1^{(1)},\quad Q_1^{\pm}=[R_1^{\pm}(-\omega_2,\omega_1)]^*.$$

If the multiplet structure is ignored, the matrix elements of $X_1^{(1)}$ are real for all frequencies ω_1, and the skew-Hermitian part is present only in the radial matrix element

$$\rho(\omega_1,-\omega_2)=\langle n_f,l_f=1|rg_2(E_i+\omega_1)r'^2 + r^2 g_1(E_i-\omega_2)r'|n_i,l_i=0\rangle$$

in the expressions for $R_1^{\pm}$ and $Q_1^{\pm}$:

$$R_1^{\pm}=\frac{\alpha}{\sqrt{6}\omega_1}\left(\tfrac{1}{5}\omega_1^2\rho(\omega_1,-\omega_2)\mp\langle n_f,l_f=1|r|n_i,l_i=0\rangle\right).$$

Table II lists the coefficients [see Eq. (33)]

$$b_1\equiv\omega_1\omega_2^3A_1=\tfrac{1}{2}\omega_1\omega_2^3(|R_1^-|^2+|Q_1^-|^2),$$

$$b_2\equiv\omega_1\omega_2^3F_1=-\tfrac{1}{2}\omega_1\omega_2^3\mathrm{Im}(R_1^-Q_1^+-R_1^+Q_1^-+R_1^+Q_1^+)$$

for $1s-2p$ scattering in hydrogen and $6s-6p$ scattering in cesium for estimates. As in the previous example, the matrix elements have been calculated for Cs by employing Green's functions in the model potential approximation.[13] Here the absolute value of the cross section is greater by several orders of magnitude than that for $s-f$ and the frequency dependence is smoother.

The resonant case can be analyzed along the same lines as in Sec. 4. Here, too, the most interesting case is the resonance on the quadrupole $|i\rangle-|r\rangle$ transition with a width Γ, a resonance absent in the E\1–E1 scattering from the initial state $|i\rangle$ (the amplitude $X_{1,2,3}^{(2)}$ in (34) are resonant.) However, the coefficients $F_{1,2}$ are now determined by the interference of $X_p^{(2)}$ and the nonresonant amplitudes $Y_p^{(\alpha)}$, which are of the same order in α as the $X_p^{(2)}$. Hence the asymmetric part $\Delta\sigma$ of the cross section containing $\Gamma/(\Delta^2+\Gamma^2/4)$, which has a smallness of order $\Gamma/|E_i|\sim\Gamma/\omega$ compared to the resonant terms $|X_p^{(2)}|^2$. Nevertheless, the absolute value of $\Delta\sigma$ near resonance is related to the cross section $\sigma^{(0)}$ of E1–E1 scattering as follows:

$$|\Delta\sigma|\sim\frac{\alpha^2|E_i|\sigma^{(0)}}{\Gamma},$$

with the result, for instance, that the intensity of the circularly polarized component of the radiation emerging in dipole-forbidden resonant scattering of linearly polarized light is of the same order of magnitude as, or even greater than, the intensity of ordinary $E1-E1$ scattering in the nonresonant case.

6. CONCLUSION

The results of the investigation show that dissipation in light scattering may occasionally lead to distinctive effects whose analysis requires measuring additional characteristics besides the standard set of traditional scattering characteristics such as the extinction coefficient (the total cross section), the degree of depolarization, and the inversion coefficient. One such additional characteristic may be the degree of asymmetry, defined as the ratio of $\Delta\sigma$ to the total cross section. Since in resonant scattering

$$\Delta\sigma\sim\frac{\Gamma}{\Delta^2+\Gamma^2/4},$$

measuring the degree of asymmetry can be used to develop an extremely sensitive polarization method of measuring resonant level widths. For one thing, the results of Sec. 4 imply that the probability of resonant two-photon excitation by linearly and elliptically polarized photons depends on the sign of the degree of circular polarization.

The scattering system in this investigation is assumed to be freely orienting in space and its polarization states before and after scattering are not fixed. Dissipation also leads to anomalies when the scattering is produced by polarized particles: for instance, the scattering cross section of unpolarized light depends on the orientation of the vector J_i of the initial state, and the vector J_f "prefers" a certain orientation when unpolarized or linearly polarized light is scattered by an unpolarized target.

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APPENDIX: COEFFICIENTS IN EQ. (33) FOR THE DIPOLE-FORBIDDEN SCATTERING CROSS SECTION

It has proved convenient to use the following quantities when writing the coefficients in Eq. (33):

$\sigma_p^{(\alpha)}=|X_p^{(\alpha)}|^2+|Y_p^{(\alpha)}|^2$, the partial cross section in(36),

$\Pi_p^{(\alpha)}=X_p^{(\alpha)}Y_p^{(\alpha)*}$, the product of $E1\to M1,E2$ and M1,$E2\to E1$ scattering amplitudes in(34),

$$R_1^{\pm}=\sqrt{\tfrac{3}{5}}X_1^{(2)}\pm X_1^{(1)},\quad Q_1^{\pm}=\sqrt{\tfrac{3}{5}}Y_1^{(2)}\pm Y_1^{(1)},$$

$$R_2^{\pm}=\tfrac{1}{\sqrt{3}}X_2^{(2)}\pm X_2^{(1)},\quad Q_2^{\pm}=\tfrac{1}{\sqrt{3}}Y_2^{(2)}\pm Y_2^{(1)},$$

$$A_0=\tfrac{1}{3}(\sigma_0^(1)+2\sigma_2^{(1)}+2\sigma_2^{(2)}+\sigma_3^{(2)},$$

$$A_1=\tfrac{1}{2}(|R_1^-|^2+|Q_1^-|^2-|R_2^+|^2-|Q_2^+|^2-\tfrac{4}{15}\sigma_3^{(2)}),$$

$$A_2=-\tfrac{1}{3}(\sigma_0^{(1)}+2\sigma_2^{(1)}+2\sigma_2^{(2)}-3|R_2^+|^2-3|Q_2^+|^2+\sigma_3^{(2)}),$$

$$B_0=-\tfrac{3}{2}\,\mathrm{Re}(\Pi_0^{(1)}-4\Pi_2^{(1)}+3R_2^-Q_2^--\Pi_3^{(2)}),$$

$$B_1=\mathrm{Re}(R_1^-Q_1^--R_2^+Q_2^+-\tfrac{4}{15}\Pi_3^{(2)}),$$

$$B_2=\tfrac{2}{3}\,\mathrm{Re}(\Pi_0^{(1)}+2\Pi_2^{(1)}+2\Pi_2^{(2)}+\Pi_3^{(2)}),$$

$$C_1=-\tfrac{1}{3}|Y_0^{(1)}|^2+\tfrac{1}{2}|Q_1^+|^2-\tfrac{1}{2}|Y_2^{(2)}+\tfrac{1}{\sqrt{33}}Y_2^{(1)}|^2+\tfrac{1}{5}|Y_3^{(2)}|^2,$$

$$C_2=-\tfrac{1}{3}|X_0^{(1)}|^2+\tfrac{1}{2}|R_1^+|^2-\tfrac{1}{2}|X_2^{(2)}+\tfrac{1}{\sqrt{3}}X_2^{(1)}|^2+\tfrac{1}{5}|X_3^{(2)}|^2,$$

$$D_1=\mathrm{Re}(R_1^+Q_1^-+R_1^-Q_1^++R_2^+Q_2^-+R_2^-Q_2^+-\tfrac{8}{15}\Pi_3^{(2)}),$$

$$D_2=-2\,\mathrm{Re}[\tfrac{1}{3}\Pi_0^{(1)}-\tfrac{1}{2}R_1^+Q_1^++\tfrac{2}{3}(\Pi_2^{(1)}-\Pi_2^{(1)}) + \tfrac{1}{2}R_2^-Q_2^--\tfrac{1}{5}\Pi_3^{(2)}],$$

$$F_1 = -\tfrac{1}{2}\,\mathrm{Im}(\tfrac{2}{3}\Pi_0^{(1)} + R_1^- Q_1^+ - R_1^+ Q_1^- + R_1^+ Q_1^+ + R_2^+ Q_2^-$$
$$- R_2^- Q_2^+ - R_2^- Q_2^+ - R_2^- Q_2^- - \tfrac{14}{15}\Pi_3^{(2)}),$$
$$F_1 \equiv F_1(\omega_1, -\omega_2), \quad F_2 = -F_1(-\omega_2, \omega_1).$$

Notwithstanding their somewhat cumbersome nature, these formulas graphically demonstrate the partial structure of the scattering cross section. For one thing, the low index used in the expression for $A_i - F_i$ designates the rank of the tensor product in Eq. (34) and determines the selection rules for the matrix elements in (25), while the upper index, $\alpha = 1$ or 2, symbolizes the $E1-M1$ or $E1-E2$ transition type, respectively. [1)]In what follows, with the exception of Sec. 3, the atomic system of units is used, with $r_0^2 = \alpha^4$, where α is the fine-structure constant.

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