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Circular dichroism effects in atomic x-ray scattering

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Abstract

It is shown that a specific beyond-dipole-approximation polarization effect in elastic photon-atom scattering, resulting in circular dichroism (CD) when the scattered photon has a suitable observed *linear* polarization, has significance in the x-ray regime. Ground state atoms are considered, assumed to be randomly oriented. The cross section, given our assumptions, is written in terms of photon Stokes parameters and four real photon-polarization-independent atomic parameters (which depend on the vectors of the problem and the two invariant scattering amplitudes). Numerical results are given for the case of scattering from ground-state atoms with Z=29, Z=54 and Z=92. Attention is given to determining the regime where the dichroism effect may be experimentally observable. It is seen to be largest for intermediate angles, high Z and high (hundreds of keV) photon energies, increasing to an approximately 20% effect in cross sections. The parameter determining this effect is also responsible for the appearance of elliptically polarized scattered photons for

the case of a suitable linearly polarized incident beam.

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I. INTRODUCTION

Advances in the use of synchrotron x-ray and γ-ray sources allow the possibility of making detailed observations of photon polarization effects in the scattering of hard photons by atomic targets. A correct theoretical analysis of this problem requires a quantum electrodynamic description with an accurate account of relativistic and retardation effects. General discussions of both theoretical and experimental results concerning elastic x-ray and γ-ray scattering by atoms can be found in [1,2] and references therein. Polarization effects in elastic photon scattering have been investigated numerically, with particular attention being paid to linear polarization effects [3]. In addition there have been more detailed investigations into the contributions of relativistic and higher multipole effects to elastic scattering [4,5], though these works mainly studied the angular distribution of scattered photons rather than polarization effects. A recent high precision experiment involving the scattering of x-rays from neon attained cross sections accurate at the level of 1% [6], though polarization effects were not considered.

The problem of photon polarization effects has been completely analyzed only for the case of photon scattering by a relativistic free electron (e. g. [7,8]). Here we concentrate on a specific beyond-dipole-approximation polarization effect that appears in the elastic scattering of photons by ground-state atoms, giving rise to finite circular dichroism (CD) when the scattered photon has a fixed suitable linear polarization. (The existence of CD in the case that the scattered photon has a fixed circular polarization is well known - see discussion below.) We understand CD as the difference between cross sections (which may generally describe any process involving an incoming photon) for different (right-handed and left-handed) circular polarizations of the incident photon, with all other observables being held fixed (including the observed polarization of the final photon, in the case of scattering).

Apparently, the possibility of CD effects in light scattering (for the case that the scattered photon has a fixed linear polarization) was first pointed out for dipole-forbidden scattering in 1980 [9], and it was studied in detail for optical frequencies in light scattering by gases in [10]. In that work a nonrelativistic treatment was used, with account of retardation effects being taken only in the first nonvanishing order. The relativistic case of CD effects in scattering of hard photons by hydrogen-like ions was first investigated numerically in 1987 [11], and later analytical results were obtained for the simplest case of atoms with closed shells [12]. Terms responsible for these effects were identified in [3] and numerical results were obtained, but the emphasis there was on linear polarization effects.

The use of circular dichroism to investigate orientations in atomic and molecular targets is longstanding [13–15]. The approach has typically involved measuring the differences between the cross sections for the photoabsorption of right-handed and left-handed circularly polarized photons in the optical regime. Recently the possibility of also using circular dichroism effects in inelastic scattering as a tool for investigating target orientations has been proposed [16,17]. In these works the authors point out that, even for the case of randomly oriented targets, there is still the possibility of finite CD effects. This they refer to as *design-induced* CD (existing in the case of a randomly oriented target), which would tend to mask the CD effects arising from the target having some definite orientation in space. The aim is to identify the conditions under which design-induced CD vanishes for a randomly oriented target, so that that any observed CD effects would then be a clear indication of target orientation.

The conclusion [16,17] (for the case of inelastic photon scattering) is that design-induced CD can exist if the scattered photon has a fixed circular polarization, but it vanishes completely if the scattered photon has a fixed linear polarization. A similar assertion has been made for elastic (coherent) photon scattering, namely that CD is not present for the case of randomly oriented targets when the scattered photon has a fixed linear polarization [18]. We wish to point out that these conclusions are limited by the approximations made in the analyses: a more general analysis reveals that design-induced CD can occur when the scattered photon has a fixed linear polarization. We examine these CD effects and determine when they will vanish. For the purposes of our discussion we distinguish between TYPE-C CD (where the scattered photon has a fixed circular polarization) and beyond-dipole-approximation

TYPE-L CD (where the scattered photon has a fixed linear polarization). Of course in the general case of a fixed elliptical polarization of the scattered photon being observed, both types of CD may be present.

We point out that, given our assumptions, the parameter describing TYPE-C CD effects is also observable in measurements involving only linear polarizations, in contrast to the parameter describing TYPE-L CD effects [see Eq. (3) in next section]. Though both of these polarization effects fall under the general definition of circular dichroism (i.e. leading to differences in cross sections for left- and right-handed circularly-polarized incident radiation, with all other details of observation the same), they are nevertheless distinct effects, providing different information about the target wavefunction.

Here we specifically consider TYPE-L CD effects in the elastic scattering of x-rays by bound atomic electrons. We suppose the target atoms to be randomly oriented. We employ a fully relativistic approach for the description of atomic electrons, retaining all significant multipoles in the electron-photon interaction. We will present numerical estimates of the importance of TYPE-L CD effects for three ground-state atoms, with Z=29, Z=54, and Z=92. Attention is given to determining the regime where these effects are large and may be experimentally observable. The ideal experiment for observing such effects involves measuring the difference between the scattering cross sections for right-handed (RHC) and left-handed (LHC) circularly polarized incident photons, observing a fixed linear polarization (making an angle of 45^o with respect to the scattering plane) for the scattered photon.

The parameter determining this CD effect is also responsible for the appearance of elliptically polarized scattered photons from a linear polarized incident photon beam (even when the scattering is from an s-state target). One could therefore also measure this parameter by designing an experiment with a fixed linear polarization (making an angle of 45^o with respect to the scattering plane) for the incoming photon, measuring the difference between scattering cross sections for scattered RHC and LHC photons.

In Sec. II we discuss the general form of the scattering cross section for the case of elastic photon scattering from ground state-atoms under the assumption that polarization effects in the target are not being observed (scattering from s-state targets). A brief discussion is given regarding the consequences of the symmetry of the cross section under time reversal. Numerical results are given in Sec. III for TYPE-L CD effects in elastic photon scattering from ground-state atoms with Z=29, Z=54 and Z=92. These results suggest the situations in which the TYPE-L CD effect is most likely to be experimentally observable. General features are explained in terms of the basic scattering amplitudes. We summarize our conclusions in Sec. IV.

II. THEORY

We consider TYPE-L CD polarization effects in the elastic scattering of a photon by an atomic target. It is assumed that the polarization properties of the target are not observed. The target is effectively treated as if it had zero total angular momentum, corresponding to a summation, weighted according to the number of electrons present, over the magnetic substates of each subshell at the level of the scattering amplitude. This approach (exact for closed-subshell atoms) is justified since most of the atomic electrons are in closed subshells, with the scattering from inner shells being dominant (except for small angles where all subshells contribute to scattering, but where the effects under consideration here are not important, in fact vanishing in the forward direction). More details of this approach can be found in [1]. Results that go beyond this approximation, performing the more correct averaging over magnetic substates at the level of the cross section, indicate that the corrections for many-electron atoms tend to be small [19].

The initial (final) photon has momentum $\boldsymbol{k}_1$ ($\boldsymbol{k}_2$) and polarization $\boldsymbol{\epsilon}_1$ ($\boldsymbol{\epsilon}_2$). Given our assumption of scattering from s-state type targets these are the only vectors in the problem. We have $\hat{\boldsymbol{k}}_i = \boldsymbol{k}_i/|\boldsymbol{k}_i|$ and, since we are considering elastic scattering, $|\boldsymbol{k}_1| = |\boldsymbol{k}_2| = \omega/c$. The Stokes parameters of incident (scattered) photons are denoted by $\xi_1^{(1)}$, $\xi_2^{(1)}$ and $\xi_3^{(1)}$ ($\xi_1^{(2)}$, $\xi_2^{(2)}$ and $\xi_3^{(2)}$). Note that $\xi_1^{(i)}$ corresponds to linear polarization making an angle of 45^o with respect to the scattering plane, $\xi_2^{(i)}$ corresponds to circular polarization, and $\xi_3^{(i)}$

corresponds to linear polarization parallel or perpendicular to the scattering plane, for the incident ($i = 1$) and scattered ($i = 2$) photons. Here we follow the notation of [8] for the Stokes parameters - this notation differs from that used in [3], as is explained in the Appendix.

The elastic scattering amplitude A, with the assumptions above, can be written in terms of two (complex) invariant amplitudes M and N [1],

$$A = (\boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2^*)\ M + (\boldsymbol{\epsilon}_1 \cdot \hat{\boldsymbol{k}}_2)(\boldsymbol{\epsilon}_2^* \cdot \hat{\boldsymbol{k}}_1)\ N, \tag{1}$$

which depend only on the photon energy ω and θ, the scattering angle between $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$. (Note that in the electric dipole approximation the N amplitude vanishes, and the M amplitude is independent of θ, so that the only dependence on θ is through $\boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2^*$). This expression for the scattering amplitude leads to the scattering cross section

$$\begin{aligned} \frac{d\sigma}{d\Omega} &= |M|^2|\boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2^*|^2 + |N|^2|\boldsymbol{\epsilon}_1 \cdot \hat{\boldsymbol{k}}_2|^2|\boldsymbol{\epsilon}_2^* \cdot \hat{\boldsymbol{k}}_1|^2 + 2\ \mathrm{Re}\ (MN^*)\ \mathrm{Re}\left[(\boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2^*)(\boldsymbol{\epsilon}_1^* \cdot \hat{\boldsymbol{k}}_2)(\boldsymbol{\epsilon}_2 \cdot \hat{\boldsymbol{k}}_1)\right] \\ &\quad + \ 2\ \mathrm{Im}\ (MN^*)\ \mathrm{Im}\left[(\boldsymbol{\epsilon}_1 \cdot \boldsymbol{\epsilon}_2^*)(\boldsymbol{\epsilon}_1^* \cdot \hat{\boldsymbol{k}}_2)(\boldsymbol{\epsilon}_2 \cdot \hat{\boldsymbol{k}}_1)\right]. \end{aligned} \tag{2}$$

Alternatively the scattering cross section (again for the case of an s-state target) can be expressed in terms of the Stokes parameters and four real photon-polarization-independent parameters d_i

$$\frac{d\sigma}{d\Omega} = d_1 + d_1\xi_3^{(1)}\xi_3^{(2)} + d_2(\xi_3^{(1)} + \xi_3^{(2)}) + d_3(\xi_1^{(1)}\xi_1^{(2)} + \xi_2^{(1)}\xi_2^{(2)}) + d_4(\xi_1^{(1)}\xi_2^{(2)} - \xi_2^{(1)}\xi_1^{(2)}), \tag{3}$$

The four real parameters d_i are given in terms of the M and N amplitudes as

$$\begin{aligned} d_1 &= \frac{1}{4}\left[|N|^2 \sin^4\theta - 2\ \mathrm{Re}\ (MN^*)\cos\theta\sin^2\theta + |M|^2(1+\cos^2\theta)\right], \\ d_2 &= \frac{1}{4}\left[|N|^2 \sin^4\theta - 2\ \mathrm{Re}\ (MN^*)\cos\theta\sin^2\theta - |M|^2(1-\cos^2\theta)\right], \\ d_3 &= \frac{1}{2}\left[|M|^2\cos\theta - \mathrm{Re}\ (MN^*)\sin^2\theta\right], \\ d_4 &= \frac{1}{2}\left[\mathrm{Im}\ (MN^*)\sin^2\theta\right]. \end{aligned} \tag{4}$$

The unpolarized cross section (averaging over incident photon polarizations, summing over final photon polarizations), is given solely in terms of the parameter d_1 (all the Stokes parameters vanish in this case)

$$\frac{d\sigma}{d\Omega} = 2d_1. \tag{5}$$

Thus the parameter d_1 can be determined without any polarization measurements. The parameters d_2 and d_3 can in principle be determined from measurements involving only linear polarizations - a discussion of this as well as numerical results can be found in [3], though the notations and choices of invariant amplitudes differ, as described in the Appendix.

Our interests here are the polarization effects associated with the parameter d_4, appearing in the last term in Eq. (3). These effects are (1) TYPE-L CD (when $\xi_1^{(2)}$ is non-zero), and (2) the appearance of elliptically polarized scattered photons for the case of linearly polarized incident photons (when $\xi_1^{(1)}$ is non-zero). Note that TYPE-C CD effects, involving circular polarizations of both incoming and scattered photons (thereby involving $\xi_2^{(1)}\xi_2^{(2)}$) are determined by the parameter d_3. Looking at Eq. (4) we see that for the parameter d_4 to be finite requires the beyond-dipole-approximation N amplitude, which enters physical observables modulated by a $\sin^2\theta$ behavior. Thus TYPE-L CD is only important when retardation matters; it vanishes at forward and backward angle and has a geometric maximum at 90^o (the actual maximum shifts towards forward angle at high energy). By contrast d_3, characterizing TYPE-C CD, can still be finite even when the N amplitude vanishes or is neglected. A common approximation for Rayleigh scattering in the x-ray regime is to describe the M amplitude using form factors and (angle-independent) anomalous scattering factors, neglecting the N amplitude altogether [20]. Polarization effects involving d_4 are not present in this approximation.

We now return to the assertions in [16–18], mentioned earlier, that CD effects in scattering from randomly oriented systems vanish completely if one chooses to have a fixed linear polarization for the final photon. Though this is true within the approximations used in those analyses [nonrelativistic dipole approximation and A^2 approximation, neglecting $\boldsymbol{p}\cdot\boldsymbol{A}$ terms in the photon-electron interaction, $H_{\rm int} = (e^2/2mc^2)A^2 - (e/mc)\boldsymbol{p}\cdot\boldsymbol{A}$], in the more general case this condition of final linear polarization only suffices to exclude TYPE-C CD effects. TYPE-L CD effects can still be present if $\xi_1^{(2)} \neq 0$. In fact we see from Eq. (3) that

CD effects in the case of randomly oriented targets vanish completely only if one has a final photon linear polarization parallel or perpendicular to the scattering plane.

We conclude this section by making some observations on the implications of the fundamental symmetry of time reversal for these effects. Consider the action of time reversal on the invariant amplitudes and the photon variables (and the Stokes parameters):

$$M, N \leftrightarrow M^*, N^*; \quad \boldsymbol{k}_1 \leftrightarrow -\boldsymbol{k}_2; \quad \boldsymbol{\epsilon}_1 \leftrightarrow \boldsymbol{\epsilon}_2^*; \quad \xi_1^{(1)}, \xi_2^{(1)}, \xi_3^{(1)} \leftrightarrow \xi_1^{(2)}, \xi_2^{(2)}, \xi_3^{(2)}. \tag{6}$$

The combination of Stokes parameters that enters the d_4 term of Eq. (3) is odd under this symmetry (T-ODD), in contrast to the combinations associated with all the other terms. Therefore the parameter d_4 must also be T-ODD in order that the cross section be invariant under the time reversal operation. (The other three parameters must be T-EVEN.) Therefore the requirement that the parameter d_4 is T-ODD (and real) implies that it should be solely the result of interference between real (T-EVEN) and imaginary (T-ODD) parts of the invariant amplitudes.

The optical theorem relates the imaginary part of the elastic scattering amplitude at forward angle to the total cross section for absorption and scattering. If one writes the optical theorem in an expansion in the fine structure constant α, one relates the imaginary part of the 2nd-order amplitude for elastic scattering to the total cross section for absorptive scattering in lowest order (including photoeffect, bound-bound transitions and bound-electron pair production, depending on the energy involved). From this viewpoint polarization effects involving d_4 can be thought of (in leading order) as dissipation-induced effects related to the existence of dissipation channels (if they are present). A consequence of this observation is the lack of TYPE-L CD in scattering by a free electron in lowest order (as can be seen from the well-known Klein-Nishina formula; see e. g. [8]), where there is no dissipative channel - a free electron cannot absorb a photon.

III. NUMERICAL RESULTS

As discussed in the previous section, the polarization effects being considered here are determined by the parameter d_4, which specifies the magnitude of terms involving the Stokes parameters $\xi_2^{(i)}$, describing circular polarization, and $\xi_1^{(i)}$, describing the linear polarization making an angle of 45^o with respect to the scattering plane. Effects involving d_4 are maximized when one photon is circularly polarized and the other has a linear polarization making an angle of 45^o with respect to the scattering plane. In this case of maximum TYPE-L CD the cross section becomes

$$\frac{d\sigma}{d\Omega} = d_1 \pm d_4, \tag{7}$$

$\pm$ as $\xi_1^{(2)}$ and $\xi_2^{(1)}$ are of opposite (same) sign, where $\xi_1^{(2)}$ is fixed to be +1 or -1, and $\xi_2^{(1)}$ =+1 (RHC incident radiation) or -1 (LHC incident radiation); all the other Stokes parameters vanish.

Since d_4 is generally fairly small compared with d_1, the cross section to be measured is always of the order of d_1. Therefore in order to observe the effect one has to be able to measure a cross section of the order of d_1, and measure it sufficiently accurately to be able to detect the influence of d_4 in Eq. (7). For this reason we have chosen to present our results by giving the magnitude of d_1, and then expressing d_4 as a percentage of d_1.

It is not in fact the case that all four parameters d_i (i=1... 4) are truly independent [3]. There exists a relation between them,

$${d_1}^2 = {d_2}^2 + {d_3}^2 + {d_4}^2, \tag{8}$$

related to the fact that the overall phase of the scattering amplitude is not an observable. In principle, if all the other parameters are known, then d_4 can be determined from Eq. (8), apart from its overall sign. That only a sign is left undetermined is a consequence of the special case of treating the target as having no angular momentum; in the general case there will be undetermined amplitudes. However, since the parameter d_4 tends to be small compared to the others (which can all be of the same order of magnitude), it is preferable to have a direct calculation, and a direct observation, of this parameter.

We present numerical results for d_1 and d_4 for ground-state atoms with Z=29, Z=54 and Z=92. We consider all scattering angles and all energies for which the effects appear to be significant. Calculations were performed using the S-matrix approach in independent particle approximation (see [1] and references therein). Electron orbitals were obtained in a Dirac-Slater type central potential. This is a fully relativistic calculation, retaining all significant multipoles in the photon-atom interaction.

Figure 1 shows a contour plot of the magnitude of d_1 for Z=29, Z=54, and Z=92 as a function of angle and energy, given in units of r_0^2 ($r_0 = e^2/mc^2$ being the classical electron radius). Note this parameter changes by many orders of magnitude over the range of Z, energies and angles considered. In Figs. 2, 3 and 4 corresponding results are presented for d_4 (expressed as a percentage of d_1), for Z=29, Z=54, and Z=92 respectively. Results are given for all scattering angles and for photon energies ranging from 100 eV to 1 MeV. This includes the inner-shell threshold region for all but the lightest atoms (for which the effects are small).

As is seen in Fig. 1, as one goes to high energies, many times threshold, the elastic scattering cross section becomes very small except at forward angles (where d_4 vanishes). Thus, although d_4 can be largest relative to d_1 at high energy, the total elastic scattering cross section is becoming unimportant except at forward angles. Only for high Z is d_1 still sizeable away from forward angles at high energies. It is also the case that d_4 is largest relative to d_1 for high Z, rising to $\approx$ 20% of d_1 for Z=92. For very low Z the effect is of very little importance. From this we conclude that the TYPE-L CD effect is of experimental consequence in the regime where magnitude of the Rayleigh scattering cross section is still large (indicated by d_1), and where d_4 is significant in comparison to d_1. This is the case for high Z, intermediate scattering angles, and photon energies in the hundreds of keV range.

We now wish to discuss some of the common features of d_4 seen in Figs. 2, 3 and 4. At lower energies there is symmetry about 90^o, corresponding to the geometric maximum. At very high energies the situation has clearly changed - the maximum shifts towards forward angles and the parameter is now negative at large angles. This is due to competition between the two terms in the expression for d_4:

$$d_4 = \frac{1}{2} [\, \mathrm{Im}\ (M)\ \mathrm{Re}\ (N) - \mathrm{Re}\ (M)\ \mathrm{Im}\ (N)\,] \sin^2 \theta. \tag{9}$$

At low energies the second term tends to dominate, since this involves the real part of the M amplitude, which contains the large (at low energy) form factor contribution. At higher energies, however, the form factor is small at finite angle, so there is more interference between the two terms, which have opposite signs, causing the d_4 parameter to pass through zero and become negative.

Since both terms involve imaginary amplitudes which (for a given subshell) will vanish below the subshell threshold, we expect d_4 to abruptly increase as the photon energy exceeds atomic thresholds (and the corresponding dissipative channels are opened). There is clear evidence of such abrupt increases in our results. For Z=29 we see such an increase occurring at the K-shell threshold ($\approx$ 9 keV). For Z=54 we see this happening at the M-shell thresholds ($\approx$ 1 keV), at the L-shell thresholds ($\approx$ 5 keV), and at the K-shell threshold ($\approx$ 34 keV). These increases are not so distinct for the case of Z=92, as there are many outer shells with binding energies in the energy range up to 10 keV, whose individual contributions are not discernible on a large scale. The inner shells have large binding energies ($\approx$ 116 keV for the K-shell) and the situation is more complicated at these high energies due to interference between the two (comparable) terms in Eq. (9).

IV. CONCLUSIONS

A unique polarization effect of circular dichroism in elastic x-ray scattering, occurring when the scattered photon has a suitable fixed linear polarization, has been investigated for the case of scattering by randomly oriented ground-state atoms. Though it is a beyond-dipole-approximation effect, it has been shown to have significance, rising to $\approx$ 20% in cross sections for Z=92 at intermediate angles. The suitable polarization measurement involves measuring the difference in the scattering cross sections for right-handed and left-handed circularly polarized incident photons, for a final photon observed having a fixed linear polarization at 45^o with respect to the scattering plane. CD effects in scattering from randomly oriented targets vanish completely only if the final photon is chosen to have a linear polarization either parallel or perpendicular to the scattering plane.

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APPENDIX

Note that there is a sign error in Eq. (12) of reference [3] - it should read

$$\frac{d\sigma}{d\Omega} = \frac{1}{4}(|A_{||}|^2 + |A_\perp|^2) + \frac{1}{4}\xi_{1i}\xi_{1f}(|A_{||}|^2 + |A_\perp|^2) + \frac{1}{4}(|A_{||}|^2 - |A_\perp|^2)(\xi_{1i} + \xi_{1f}) + \frac{1}{4}(A_{||}A_\perp^* + A_{||}^*A_\perp)(\xi_{2i}\xi_{2f} + \xi_{3i}\xi_{3f}) + \frac{1}{4}i(A_{||}A_\perp^* - A_{||}^*A_\perp)(\xi_{3i}\xi_{2f} - \xi_{2i}\xi_{3f}). \quad \text{(A1)}$$

This equation describes the differential cross section for elastic photon scattering from an unpolarized target in terms of the Stokes parameters and combinations of two complex amplitudes. It corresponds to Eq. (3) of this paper. The amplitudes $A_{||}$ and $A_\perp$ are related to the M and N amplitudes through the relations

$$A_{||} = M\cos\theta - N\sin^2\theta, \quad A_\perp = M. \quad \text{(A2)}$$

A different notation for the Stokes parameters was used in [3]. In order to compare Eq. (A1) with Eq. (3) the following substitutions should be made, relating the two sets of Stokes parameters:

$$\xi_{1i} \to \xi_3^{(1)}, \quad \xi_{2i} \to \xi_1^{(1)}, \quad \xi_{3i} \to \xi_2^{(1)},$$
$$\xi_{1f} \to \xi_3^{(2)}, \quad \xi_{2f} \to \xi_1^{(2)}, \quad \xi_{3f} \to \xi_2^{(2)}. \tag{A3}$$

We can then rewrite Eq. (A1) as

$$\frac{d\sigma}{d\Omega} = d_1 + d_2(\xi_3^{(1)} + \xi_3^{(2)}) + d_3(\xi_1^{(1)}\xi_1^{(2)} + \xi_2^{(1)}\xi_2^{(2)}) + d_1\xi_3^{(1)}\xi_3^{(2)} + d_4(\xi_1^{(1)}\xi_2^{(2)} - \xi_2^{(1)}\xi_1^{(2)}), \tag{A4}$$

where

$$d_1 = \frac{1}{4}(|A_{||}|^2 + |A_\perp|^2), \quad d_2 = \frac{1}{4}(|A_{||}|^2 - |A_\perp|^2),$$
$$d_3 = \frac{1}{4}(A_{||}^* A_\perp + A_{||} A_\perp^*), \quad d_4 = \frac{i}{4}(A_{||}^* A_\perp - A_{||} A_\perp^*), \tag{A5}$$

and the identification with Eq. (3) of this paper is complete. A detailed discussion of other commonly used representations of the invariant amplitudes is given in [1].

REFERENCES

[1] P. P. Kane, L. Kissel, R. H. Pratt, and S. C. Roy, Phys. Rep. **140**, 75 (1986).

[2] R. H. Pratt, L. Kissel, and P. M. Bergstrom, Jr., in *X-ray anomalous (resonance) scattering: theory and experiment*, edited by K. Fischer, G. Materlik, and C. Sparks (Elsevier, North Holland, 1994), pp. 9–33.

[3] S. C. Roy, B. Sarkar, L. Kissel, and R. H. Pratt, Phys. Rev. A **34**, 1178 (1986).

[4] V. Florescu and M. Marinescu, Phys. Rev. A **42**, 3844 (1990).

[5] A. Costescu, P. M. Bergstrom, Jr., C. Dinu, and R. H. Pratt, Phys. Rev. A **50**, 1390 (1994).

[6] M. Jung, R. W. Dunford, D. S. Gemmell, E. P. Kanter, B. Krässig, T. W. LeBrun, S. H. Southworth, L. Young, J. P. J. Carney, L. LaJohn, R. H. Pratt, and P. M. Bergstrom, Jr., Phys. Rev. A **81**, 1596 (1998).

[7] W. H. McMaster, Rev. Mod. Phys. **33**, 8 (1961).

[8] V. B. Berestetskii, E. M. Lifshitz, and L. P. Pitaevskii, *Quantum Electrodynamics* (Pergamon Press, Oxford, 1982).

[9] N. L. Manakov, V. D. Ovsiannikov, and Z. Ozgo, Physica C **100**, 260 (1980).

[10] N. L. Manakov, Zh. Eksp. Teor. Fiz. **106**, 1286 (1994) [Sov. Phys. JETP **79**, 696 (1994)].

[11] N. L. Manakov, A. A. Nekipelov, and A. G. Fainshtein, Yad. Fiz. **45**, 1091 (1987) [Sov. J. Nucl. Phys. **45**, 677 (1987)].

[12] N. L. Manakov, S. I. Marmo, and A. V. Meremianin, J. Electron Spectrosc. Relat. Phenom. **79**, 327 (1996).

[13] L. Velluz, M. Le Grand, and M. Grosjean, *Optical circular dichroism: principles, measurements, and applications* (Academic Press, New York, 1965).

[14] *Circular dichroism: principles and applications*, edited by K. Nakanishi, N. Berova, and R. W. Woody (VCH Publishers, New York, 1994).

[15] *Applications of circularly polarized radiation using synchrotron and ordinary sources*, edited by F. Allen and C. Bustamante (Plenum Press, New York, 1985).

[16] T. Åberg and S. Heinäsmäki, Appl. Phys. B **65**, 131 (1997).

[17] T. Åberg and S. Heinäsmäki, J. Phys. B **30**, 4827 (1997).

[18] M. Y. Agre and L. P. Rapoport, Zh. Eksp. Teor. Fiz. **109**, 1203 (1996) [Sov. Phys. JETP **82**, 647 (1996)].

[19] J. P. J. Carney, L. Kissel, R. H. Pratt, S. C. Roy, and S. K. Sen Gupta, Radiat. Phys. Chem. **51**, 371 (1998).

[20] L. Kissel, B. Zhou, S. C. Roy, S. K. S. Gupta, and R. H. Pratt, Acta Cryst. A **51**, 271 (1995).

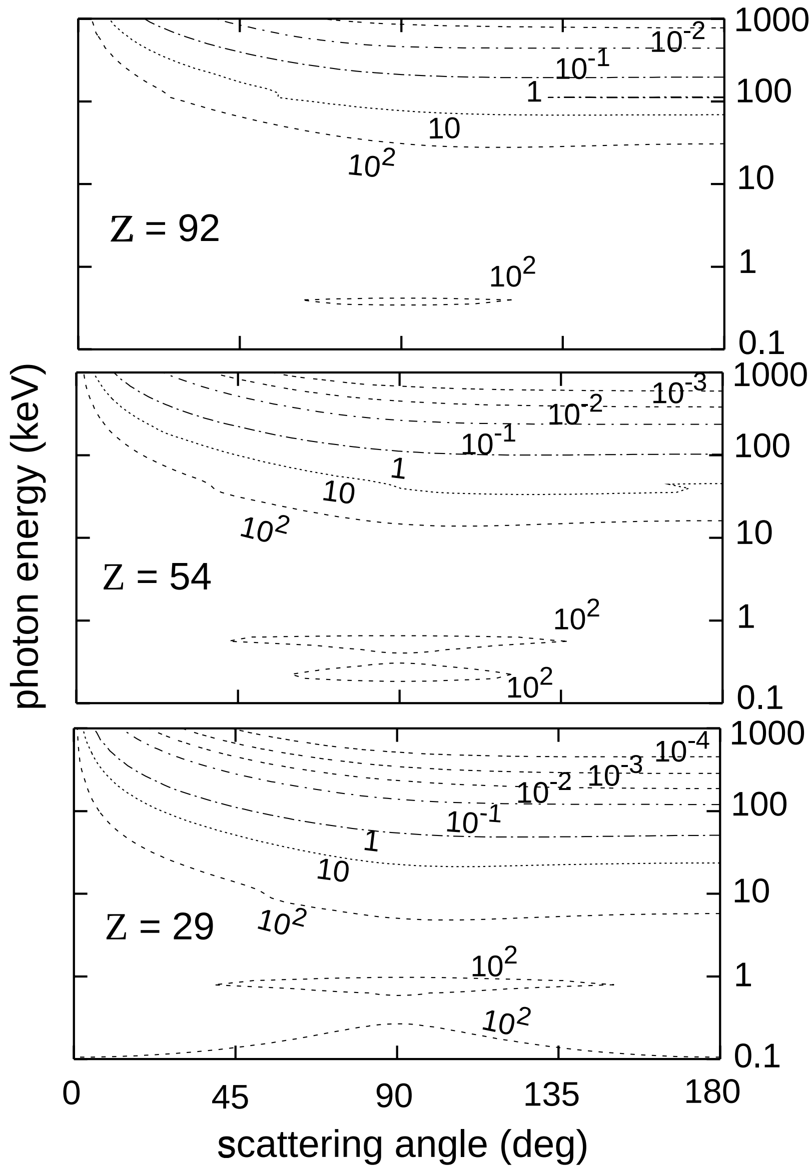

FIG. 1. Contour plot of the parameter d_1, related to the unpolarized cross section, in units of r_0^2, for Z=29, Z=54, and Z=92, as a function of scattering angle (abscissa) and energy (ordinate, on a log scale).

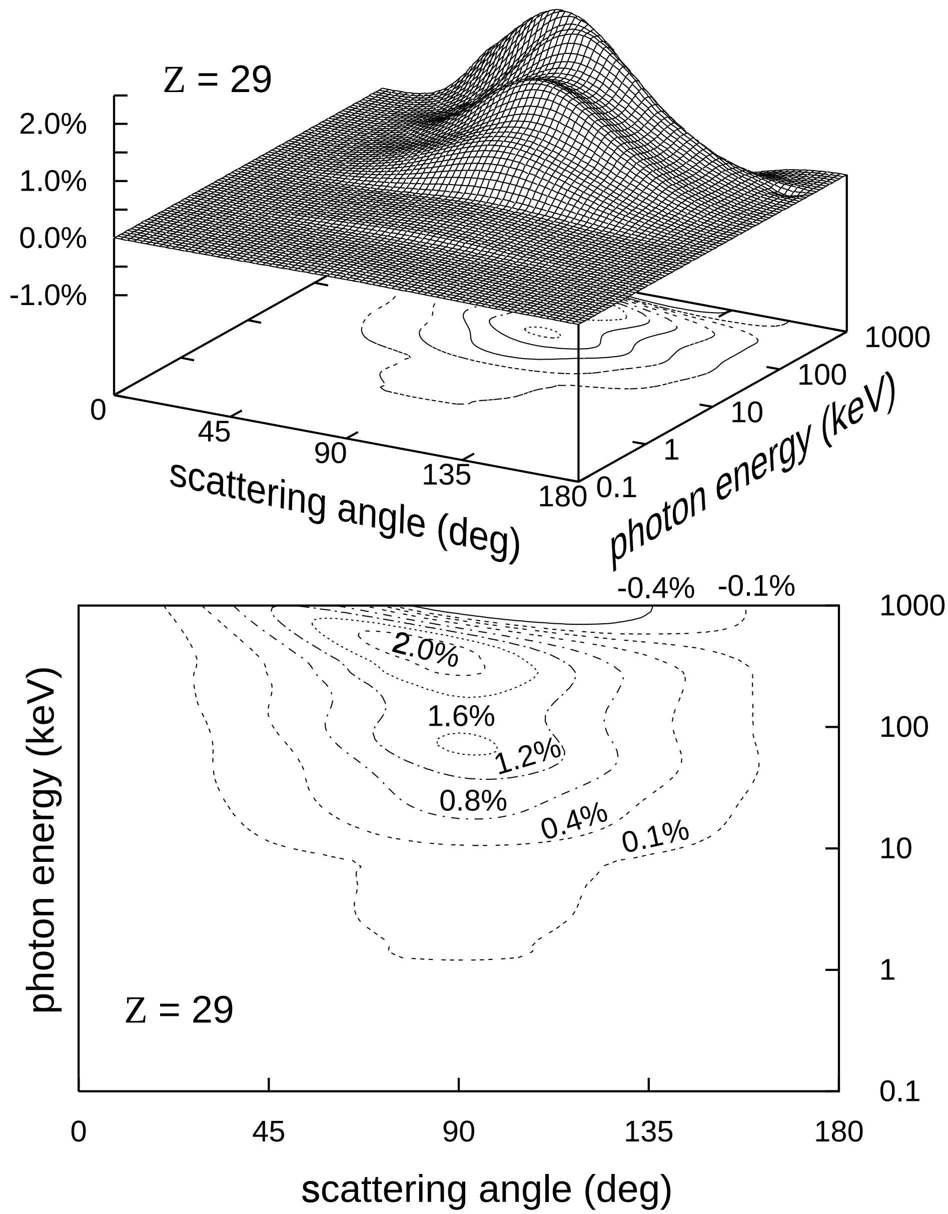

FIG. 2. Three dimensional and contour plot of the parameter d_4 expressed as a percentage of the parameter d_1 for Z=29, as a function of scattering angle (abscissa) and energy (ordinate, on a log scale).

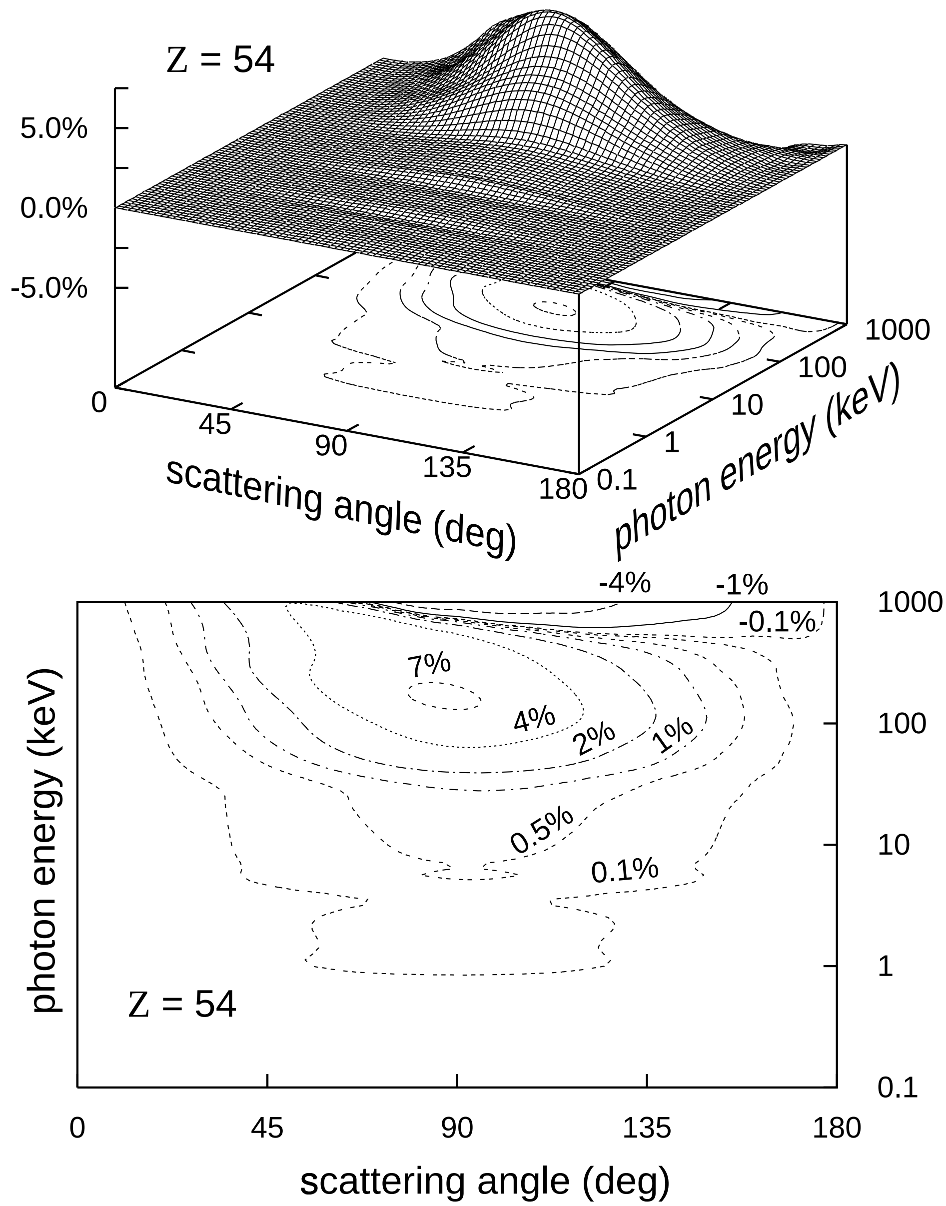

FIG. 3. Three dimensional and contour plot of the parameter d_4 expressed as a percentage of the parameter d_1 for Z=54, as a function of scattering angle (abscissa) and energy (ordinate, on a log scale).

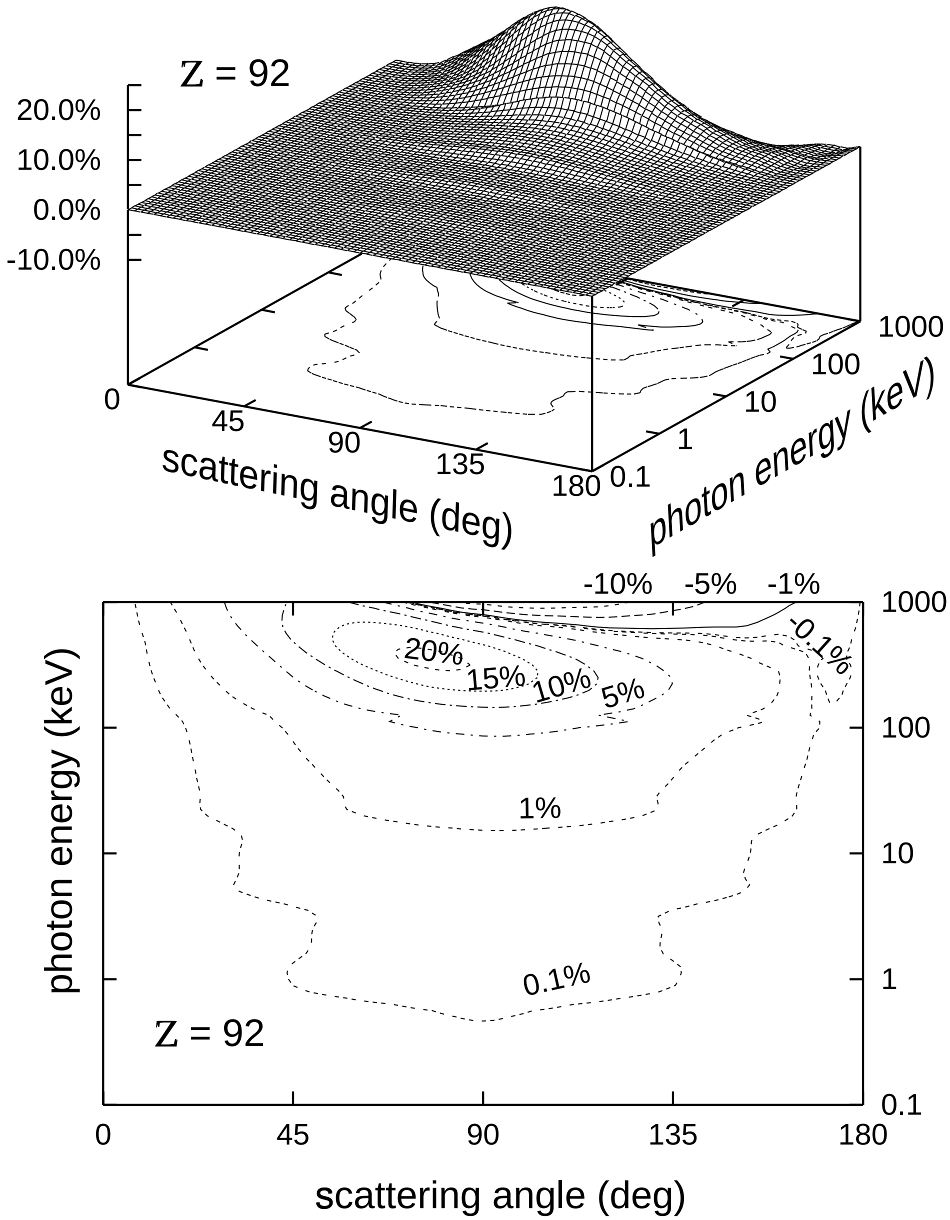

FIG. 4. Three dimensional and contour plot of the parameter d_4 expressed as a percentage of the parameter d_1 for Z=92, as a function of scattering angle (abscissa) and energy (ordinate, on a log scale).