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Circular dichroism at equal energy sharing in photo-double-ionization of He

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Interference between dipole and quadrupole transition amplitudes in photo-double-ionization of He by an elliptically polarized vuv photon is shown to induce circular dichroism in the case of equal energy sharing. The magnitude of this retardation-induced dichroic effect is estimated and its impact on the nondipole asymmetries of the triply differential cross section is demonstrated.

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The process of photo-double-ionization (PDI) of He has attracted much interest during the last decade [1–3]. Experimental measurements of the PDI triply differential cross sections (TDCSs) have been interpreted using the electric-dipole approximation (EDA) for the electron-photon interaction. Within the EDA, the dependence of the TDCSs on the mutual angle, $\theta_{12}\equiv\theta$, between the photoelectron momenta, $\mathbf{p}_1$ and $\mathbf{p}_2$, and on the photon polarization is now well-studied for photon energies up to a few hundred eV. A most interesting feature of PDI is the circular dichroism (CD) effect, i.e., the dependence of the TDCS upon the handedness of an elliptically polarized photon (i.e., upon the sign of the degree of circular polarization, ξ, where $-1\leqslant\xi\leqslant+1$). The CD effect for PDI of He was first predicted theoretically [4] and then observed experimentally [5]. Up to now, all theoretical treatments of CD have employed the EDA (Refs. [1,3,4,6–13]) and their predictions are generally in agreement with experiments [12–16]. Measurement of CD provides a sensitive means for probing two-electron dynamics in PDI, since the photon-helicity-dependent CD term in the TDCS originates from an interference between real and imaginary parts of particular components of the (generally non-Hermitian) PDI amplitude [4,7]. Dichroic effects thus permit direct experimental measurements of this otherwise elusive "cross-interference" caused by the pseudoscalar origin of the parameter ξ [17].

A distinct feature of the CD effect within the EDA is that it vanishes at *equal* energy sharing ($p_1=p_2$), since in this case the PDI amplitude involves only a single θ-dependent scalar component (as explained below). One may expect this rule to fail if one treats the electron-photon interaction beyond the EDA, i.e., if one introduces the dependence of the TDCS upon the wave vector of the incident light wave (or upon its spatial inhomogeneity). As has been shown recently [18] (for the case of the quadrupole-induced asymmetry of the TDCS with respect to the direction of a linearly polarized photon beam), retardation effects can modify the TDCS considerably even at relatively small excess energies. Though existing measurements of the CD effect [12–16] do not show any signatures of nondipole effects, this fact is expected because all of them have been performed in the orthogonal geometry (i.e., when the photon wave vector, $\mathbf{k}$, is orthogonal to the detection plane spanned by the vectors $\mathbf{p}_1$ and $\mathbf{p}_2$), in which case the lowest-order retardation effects do not contribute to the TDCS [18].

In this Rapid Communication we analyze the impact of nondipole effects on the light polarization dependence of the TDCS in the vuv region of photon energies. We find that the most interesting polarization features, induced by the interference of dipole ($E1$) and quadrupole ($E2$) transition amplitudes, A_d and A_q, appear at equal excess energy sharing. For this case we predict a nonzero CD effect as well as CD-induced differences in the nondipole asymmetries of the TDCS with respect to (i) inversion of the photon beam direction and (ii) reflection of the electron pair with respect to the photon polarization plane.

The dipole-quadrupole TDCS for PDI of He by an elliptically polarized photon with complex polarization vector $\mathbf{e}$ $[(\mathbf{e}\cdot\mathbf{e}^*)=1]$ is

$$d^3\sigma/(dE_1 d\Omega_1 d\Omega_2)\equiv\sigma=\mathcal{A}|A_d+A_q|^2, \tag{1}$$

where $\mathcal{A}=4\pi^2\alpha p_1p_2/\omega$ is a normalization factor, $\alpha=1/137$, and atomic units are used. The quadrupole amplitude A_q may be parametrized as follows [18]:

$$A_q=g_1(\mathbf{e}\cdot\hat{\mathbf{p}}_1)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1)+g_2(\mathbf{e}\cdot\hat{\mathbf{p}}_2)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2)$$
$$+g_s[(\mathbf{e}\cdot\hat{\mathbf{p}}_1)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2)+(\mathbf{e}\cdot\hat{\mathbf{p}}_2)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1)], \tag{2}$$

where the dynamical factors are $g_1\equiv g(p_1,p_2,\cos\theta)$, g_2

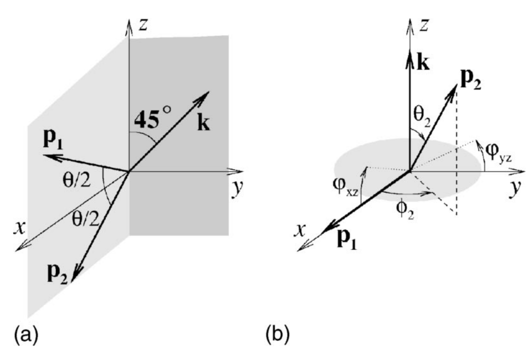

FIG. 1. Geometries suitable for observation of retardation-induced polarization effects. (a) Geometry at which the equal energy sharing CD effect is maximal. Electrons are detected in the xz plane, at angles of $\pm\theta/2$ with respect to the x axis. The photon wave vector $\mathbf{k}$ lies in the yz plane and makes an angle 45° with the z axis. (b) Geometry for observation of the retardation-induced asymmetries in the TDCS. The first electron is ejected along the x axis and the second one (whose angular distribution exhibits the asymmetry) along (θ_2,ϕ_2). (Concerning the angles φ_{xz} and φ_{yz}, see Fig. 3.)

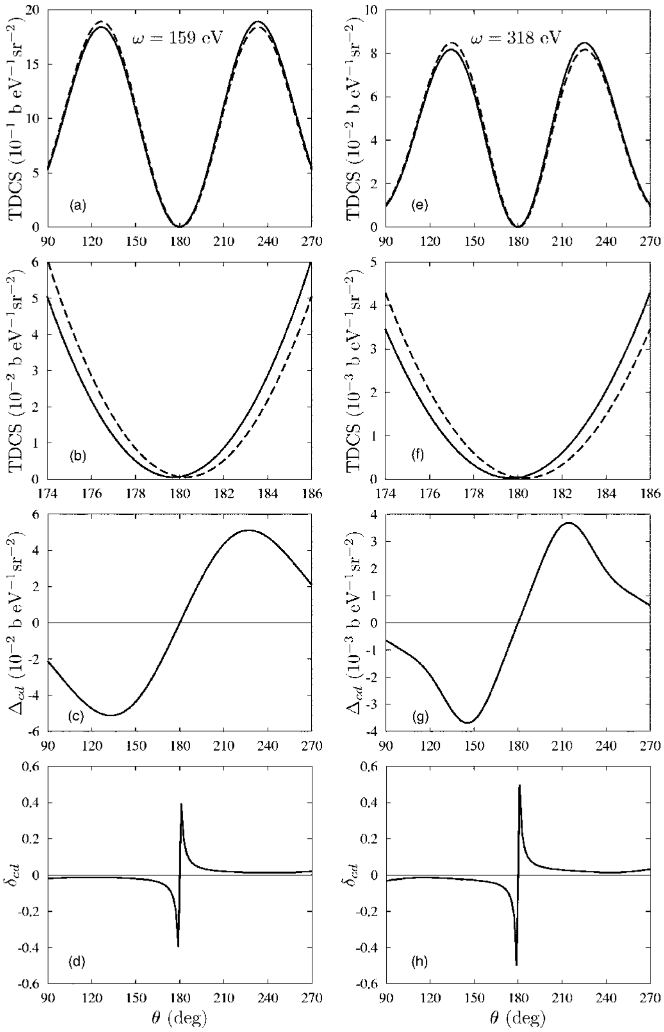

FIG. 2. The TDCS (9) and absolute (Δ_{CD}) and relative (δ_{CD}) CD parameters for PDI of He using circularly polarized light, plotted vs the mutual ejection angle θ. The geometry is as shown in Fig. 1(a). In (a),(b),(e),(f), full curve: $\sigma(\xi=+1)$; dashed curve: $\sigma(\xi=-1)$. The plots in (b),(f) show the TDCSs in the range of $174° < \theta < 186°$, in which the parameter δ_{CD} is large.

$\equiv g(p_2,p_1,\cos\theta)$, and $g_s \equiv g_s(p_1,p_2,\cos\theta) = g_s(p_2,p_1,\cos\theta)$. The dependence of the functions g and g_s on the mutual angle θ have been parametrized in Ref. [18] in terms of derivatives of the Legendre polynomials, $P_l(\cos\theta)$, and reduced matrix elements of the quadrupole momentum operator between the initial $^1S_0$-state, $|0\rangle$, and the D-wave component of the two-electron continuum state, $|\mathbf{p}_1\mathbf{p}_2\rangle$, with individual angular momenta l and $l'=l$, $l\pm 2$. The parametrization of the dipole amplitude, A_d, is well-known (see, e.g., Refs. [1,7]),

$$A_d = f_1(\mathbf{e}\cdot\hat{\mathbf{p}}_1) + f_2(\mathbf{e}\cdot\hat{\mathbf{p}}_2), \tag{3}$$

where $f_1 = f(p_1,p_2,\cos\theta)$, $f_2 = f(p_2,p_1,\cos\theta)$, and the function $f(p,p',\cos\theta)$ is given in Ref. [7] in terms of $P_l'(\cos\theta)$ and the reduced dipole matrix elements between $|0\rangle$ and the P-wave component of $|\mathbf{p}_1\mathbf{p}_2\rangle$ with individual angular momenta l and $l'=l\pm1$. Note that $f_1=f_2$ and $g_1=g_2$ for equal energy sharing.

For analysis of photon polarization effects, it is convenient to parametrize the TDCS (1) (neglecting the small terms $\sim|A_q|^2$) similarly to that for the dipole PDI [7]:

$$\begin{aligned}\sigma = \mathcal{A}\{&c_1|\mathbf{e}\cdot\hat{\mathbf{p}}_1|^2 + c_2|\mathbf{e}\cdot\hat{\mathbf{p}}_2|^2 + \mathrm{Re}\, c_3[(1-\ell)((\hat{\mathbf{p}}_1\cdot\hat{\mathbf{p}}_2)\\ &-(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2)) + 2\ell(\hat{\boldsymbol{\epsilon}}\cdot\hat{\mathbf{p}}_1)(\hat{\boldsymbol{\epsilon}}\cdot\hat{\mathbf{p}}_2)]\\ &+\xi\,\mathrm{Im}\, c_3(\hat{\mathbf{k}}\cdot[\hat{\mathbf{p}}_1\times\hat{\mathbf{p}}_2])\},\end{aligned} \tag{4}$$

where the coefficients c_i, however, are now $\mathbf{k}$-dependent:

$$c_1 = |f_1|^2 + 2\,\mathrm{Re}[f_1 g_1^*(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1) + f_1 g_s^*(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2)],$$

$$c_2 = |f_2|^2 + 2\,\mathrm{Re}[f_2 g_2^*(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2) + f_2 g_s^*(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1)],$$

$$c_3 = f_1 f_2^* + (f_1 g_s^* + f_2^* g_1)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_1) + (f_2^* g_s + f_1 g_2^*)(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}}_2). \tag{5}$$

The parameter ℓ in Eq. (4) is the degree of linear polarization of an elliptically polarized photon, $\ell = \mathbf{e}^2 = \sqrt{1-\xi^2}$, and the unit vector $\hat{\boldsymbol{\epsilon}}$ is directed along the major axis of the polarization ellipse. [Note that Eq. (4) may be easily rewritten also in terms of the Stokes parameters S_j [1]; in particular, $\xi \equiv i(\hat{\mathbf{k}}\cdot[\mathbf{e}\times\mathbf{e}^*]) = -S_3$.]

The *ab initio* parametrization of the TDCS in Eqs. (4) and (5) is independent of the dynamical model used for description of the correlated electrons and allows one to perform a complete analysis of nondipole effects for arbitrarily polarized photons. The CD effect is described by the term proportional to ξ in Eq. (4). Thus the absolute CD parameter, $\Delta_{\mathrm{CD}} \equiv \sigma(\xi=+1) - \sigma(\xi=-1)$, is

$$\Delta_{\mathrm{CD}} = 2\mathcal{A}\,\mathrm{Im}\, c_3(\hat{\mathbf{k}}\cdot[\hat{\mathbf{p}}_1\times\hat{\mathbf{p}}_2]). \tag{6}$$

If one keeps only the term $f_1 f_2^*$ in Eq. (5), the parameter Δ_{CD} in Eq. (6) (where $\mathrm{Im}\, c_3 = \mathrm{Im}\{f_1 f_2^*\}$) reduces to the known result for the dipole CD [4,7], whereas the nonzero imaginary parts of other terms in Eq. (5) determine the contribution of retardation effects to Δ_{CD}. At $p_1=p_2$, we have $f_1=f_2$, so that $\mathrm{Im}\{f_1 f_2^*\}=0$ and the dipole CD vanishes. However, the retardation-induced CD effect remains nonzero even at $p_1 = p_2$.

In order to analyze the dipole-quadrupole TDCS at equal energy sharing, it is convenient to use the parametrization of A_q in terms of the symmetrized combinations of $\hat{\mathbf{p}}_1$ and $\hat{\mathbf{p}}_2$: $\mathbf{p}_+ = (\hat{\mathbf{p}}_1+\hat{\mathbf{p}}_2)/2$ and $\mathbf{p}_- = (\hat{\mathbf{p}}_1-\hat{\mathbf{p}}_2)/2$. [Note that $(\mathbf{p}_+\cdot\mathbf{p}_-)=0$.] With these definitions, Eqs. (2) and (3) have the following forms:

$$A_d = f^{(g)}(\mathbf{e}\cdot\mathbf{p}_+) + f^{(u)}(\mathbf{e}\cdot\mathbf{p}_-), \tag{7}$$

$$\begin{aligned}A_q = {}&g_+^{(g)}(\mathbf{e}\cdot\mathbf{p}_+)(\hat{\mathbf{k}}\cdot\mathbf{p}_+) + g_-^{(g)}(\mathbf{e}\cdot\mathbf{p}_-)(\hat{\mathbf{k}}\cdot\mathbf{p}_-)\\ &+ g^{(u)}[(\mathbf{e}\cdot\mathbf{p}_+)(\hat{\mathbf{k}}\cdot\mathbf{p}_-) + (\mathbf{e}\cdot\mathbf{p}_-)(\hat{\mathbf{k}}\cdot\mathbf{p}_+)],\end{aligned} \tag{8}$$

where the symmetrized amplitudes are: $f^{(g)} = f_1 + f_2$, $f^{(u)} = f_1 - f_2$, $g_\pm^{(g)} = g_1 + g_2 \pm 2g_s$, and $g^{(u)} = g_1 - g_2$. For equal energy sharing, $f^{(u)} = g^{(u)} = 0$. [This description is very similar to the well-known parametrization of the dipole amplitude A_d in

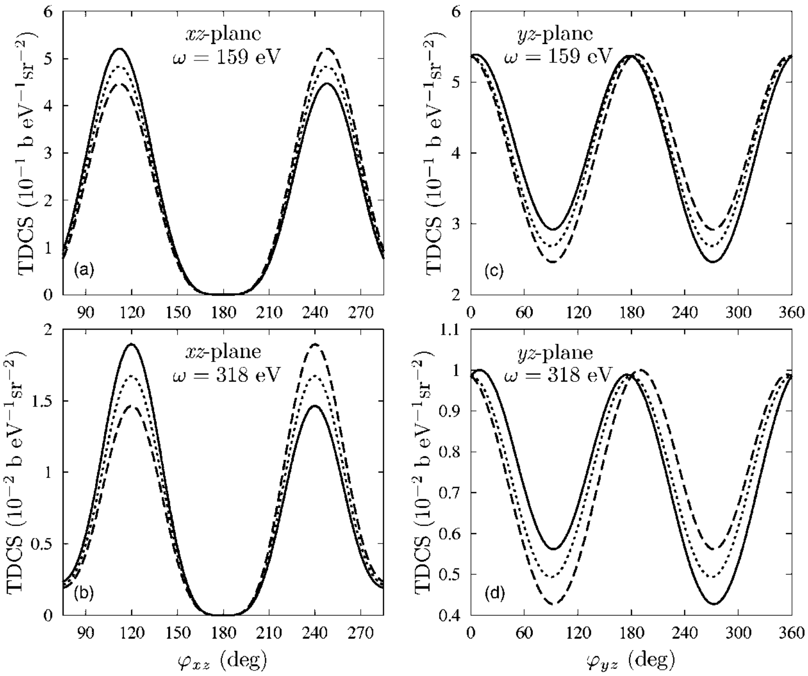

FIG. 3. The asymmetry of the TDCS (9) for PDI of He using circularly polarized light ($\xi = +1$). Full curves: the geometry is as in Fig. 1(b); dashed curves: direction of the photon beam is inverted (i.e., $\mathbf{k} \to -\mathbf{k}$); dotted curves: EDA results. φ_{xz} and φ_{yz} are the polar angles of the vector $\mathbf{p}_2$ in the planes xz (a), (b) and yz (c), (d), respectively [cf. Fig. 1(b)].

terms of the vectors $\hat{\mathbf{p}}_1 \pm \hat{\mathbf{p}}_2$ and the symmetrized (gerade and ungerade) amplitudes $a_{g,u}$ [1,8,19]; note that $f^{(g,u)} = 2a_{g,u}$.] The parametrization of our symmetrized amplitudes in terms of Legendre polynomials and reduced matrix elements follows immediately from that for the functions $f(p, p', \cos\theta)$ [7] and $g(p, p', \cos\theta)$, $g_s(p, p', \cos\theta)$ [18].

The TDCS in terms of the symmetrized amplitudes has a form identical to that in Eqs. (4) and (5) provided the substitutions, $\{\hat{\mathbf{p}}_1, \hat{\mathbf{p}}_2, f_1, f_2, g_1, g_2, g_s\} \to \{\mathbf{p}_+, \mathbf{p}_-, f^{(g)}, f^{(u)}, g_+^{(g)}, g_-^{(g)}, g^{(u)}\}$, are made. Although this parametrization does not simplify the general analysis, it leads to a simpler form of the TDCS for an elliptic polarization for the case of equal energy sharing:

$$\sigma^{(\mathrm{eq})} = \mathcal{A}[|f^{(g)}|^2 + 2\,\mathrm{Re}\{f^{(g)*}g_+^{(g)}\}(\hat{\mathbf{k}}\cdot\mathbf{p}_+)]|\mathbf{e}\cdot\mathbf{p}_+|^2 + \mathcal{A}\,\mathrm{Re}\{f^{(g)*}g_-^{(g)}\}(\hat{\mathbf{k}}\cdot\mathbf{p}_-)[2\ell(\hat{\boldsymbol{\epsilon}}\cdot\mathbf{p}_+)(\hat{\boldsymbol{\epsilon}}\cdot\mathbf{p}_-) + (\ell-1) \times(\hat{\mathbf{k}}\cdot\mathbf{p}_+)(\hat{\mathbf{k}}\cdot\mathbf{p}_-)] + (\xi/2)\Delta_{\mathrm{CD}}^{(\mathrm{eq})}, \tag{9}$$

where $2|\mathbf{e}\cdot\mathbf{p}_+|^2 = 2\ell(\hat{\boldsymbol{\epsilon}}\cdot\mathbf{p}_+)^2 + (1-\ell)[\hat{\mathbf{k}}\times\mathbf{p}_+]^2$ [20], and where the absolute CD parameter is [cf. Eq. (6)]:

$$\Delta_{\mathrm{CD}}^{(\mathrm{eq})} = 2\mathcal{A}\,\mathrm{Im}\{f^{(g)*}g_-^{(g)}\}(\hat{\mathbf{k}}\cdot[\mathbf{p}_-\times\mathbf{p}_+])(\hat{\mathbf{k}}\cdot\mathbf{p}_-). \tag{10}$$

The geometry for which $\Delta_{\mathrm{CD}}^{(\mathrm{eq})}$ has "kinematical" maxima may be deduced by supposing that the vectors $\mathbf{p}_-$ and $\mathbf{p}_+$ are directed along the z and x axes of a coordinate frame, so that the y-axis is directed along the vector product $[\mathbf{p}_-\times\mathbf{p}_+] = [\hat{\mathbf{p}}_1\times\hat{\mathbf{p}}_2]/2$ [see Fig. 1(a)]. In terms of $\theta_{\mathbf{k}}$ and $\phi_{\mathbf{k}}$, the spherical angles of the vector $\hat{\mathbf{k}}$, we obtain

$$2(\hat{\mathbf{k}}\cdot\mathbf{p}_-)(\hat{\mathbf{k}}\cdot[\mathbf{p}_-\times\mathbf{p}_+]) = (\hat{\mathbf{k}}\cdot\hat{\mathbf{z}})(\hat{\mathbf{k}}\cdot\hat{\mathbf{y}})\sin(\theta/2)\sin\theta = \sin(2\theta_{\mathbf{k}})\sin\phi_{\mathbf{k}}\sin^2(\theta/2)\cos(\theta/2). \tag{11}$$

The modulus of this expression is maximal for $\theta_{\mathbf{k}} = \pm\pi/4$ and $\phi_{\mathbf{k}} = \pm\pi/2$ [see Fig. 1(a)]. Thus, these configurations correspond to maxima of the CD effect.

In order to estimate the magnitude of retardation-induced polarization effects, we have used lowest-order perturbation theory in the interelectron interaction to calculate the functions $f(p, p', \cos\theta)$, $g(p, p', \cos\theta)$, and $g_s(p, p', \cos\theta)$. As in Refs. [11,21], for excess energies of the order of tens of eV, final state electron correlations are taken into account perturbatively to lowest order, and the He ground state is represented by the variational (uncorrelated) wave function having the effective nuclear charge $Z = 27/16$. This approach has been shown in Refs. [11,21] to provide reasonable predictions for the dipole TDCS (except for small mutual angles, $\theta \leqslant 60°$).

In Fig. 2 we present our results for the TDCS, Δ_{CD}, and the relative CD parameter, $\delta_{\mathrm{CD}} = \Delta_{\mathrm{CD}}/[\sigma(+1)+\sigma(-1)]$, for photon energies of 159 eV (2a–2d) and 318 eV (2e–2h), which correspond to excess energies of 80 eV and 239 eV [22]. The geometry is as in Fig. 1(a). Individual electron angular momenta up to $l = 5$ are accounted for in the amplitude A_d, and up to $l = 6$ in A_q. As Fig. 2 shows, there are two possibilities for observation of CD at equal energy sharing: (i) for $120° < \theta < 140°$, where the TDCS is maximum, the effect is 3–4%; (ii) for θ in the range 174°–178° or 182°–186°, where the TDCS is smaller, the effect is in the range of 20–60%.

As shown above, the CD effect is caused by the "cross-interference" term, $\mathrm{Im}\{f^{(g)*}g_-^{(g)}\}$, in the TDCS [cf. Eqs. (9) and (10)], whose measurement is possible, e.g., in two experiments having opposite signs of the parameter ξ. On the other hand, together with the "regular" interference parameters, $\mathrm{Re}\{f^{(g)*}g_{\pm}^{(g)}\}$, this term causes retardation-induced asymmetries in the TDCS (9). There are two kinds of these asymmetries for the case of an elliptic (or circular) polarization with respect to: (i) inversion of the photon beam, i.e., $\mathbf{k}\to-\mathbf{k}$, and (ii) reflection of the electron pair with respect to the photon polarization plane, i.e., $\theta_{1,2}\to(\pi-\theta_{1,2})$, where θ_1 and θ_2 are the polar angles of the photoelectrons in a reference frame with z-axis along $\mathbf{k}$ [see Fig. 1(b)]. To analyze these asymmetries, it is useful to note that

$$\sigma(\mathbf{k},\theta_1,\theta_2,\xi)=\sigma(-\mathbf{k},\pi-\theta_1,\pi-\theta_2,-\xi), \qquad (12)$$

which is obvious from Eq. (9). Therefore, for the case of linear polarization ($\xi=0$) one obtains: $\sigma(-\mathbf{k},\theta_1,\theta_2,0)=\sigma(\mathbf{k},\pi-\theta_1,\pi-\theta_2,0)$, i.e., the transformations (i) and (ii) are equivalent: in both cases the $E1$-$E2$ terms in Eq. (9) change their sign. (This "degenerate" asymmetry for the case of linear polarization has been discussed in Ref. [18].) The difference in (i) and (ii) for $\xi\neq 0$,

$$\sigma(-\mathbf{k},\theta_1,\theta_2,\xi)-\sigma(\mathbf{k},\pi-\theta_1,\pi-\theta_2,\xi)=\xi\Delta_{\mathrm{CD}}^{(\mathrm{eq})}, \qquad (13)$$

results from the invariance of the CD term in Eq. (9) with respect to the substitution $\mathbf{k}\to-\mathbf{k}$. Thus, for an elliptic polarization, the asymmetry (i) becomes equivalent to (ii) only for geometries in which CD vanishes.

To observe the asymmetries of the TDCS (9), a geometry in which one electron is detected in the polarization plane is convenient, as in Fig. 1(b). In Fig. 3 we present the TDCS (9) for the case of right circular polarization ($\xi=+1,\ell=0$), two excess energies (80 and 239 eV), and two geometrical configurations, defined as follows. In Figs. 3(a) and 3(b) the second electron is detected in the xz plane, so that the CD term in Eqs. (9) and (10) disappears; in Figs. 3(c) and 3(d), the second electron is detected in the yz plane, so that the CD term contributes to the TDCS (except for $\varphi_{yz}=n\pi/2$ with $n=0,1,2,3,4$). In Fig. 3 the full curves correspond to the photon beam directed along $\mathbf{k}$, as in Fig. 1(b); the dashed curves correspond to the photon beam directed along $\mathbf{k}'=-\mathbf{k}$. In accord with the general analysis above, one sees that in panels (a),(b) (when the CD term does not contribute to the TDCS), the TDCSs represented by the full curves and dashed curves coincide after reflection with respect to the axis $\varphi_{xz}=180°$ [i.e., $\varphi_{xz}\to(2\pi-\varphi_{xz})$ or, equivalently, $\theta_2\to(\pi-\theta_2)$], while in panels (c),(d) (when the CD term contributes to the TDCS), no such symmetry in φ_{yz} exists (except for $\varphi_{yz}=n\pi/2$). Note finally that the angular distribution of the second electron is always shifted in the "forward" direction (i.e., more electrons are ejected along the vector $\mathbf{k}$ than in the opposite direction), in agreement with intuitive assumptions on the role of retardation effects.

In summary, we have performed an accurate theoretical analysis of photon polarization effects in PDI taking into account lowest-order retardation corrections and have outlined two experimental arrangements for measuring CD effects in PDI of He for equal energy sharing. These effects do not exist in the EDA; they thus provide a polarization sensitive measure of nondipole corrections to the transition amplitudes. Our numerical estimates indicate that CD effects are significant even for electron energies as low as 40 eV and may be observable with current state-of-the-art capabilities. Owing to the significance of retardation effects for such a compact system as He, they may be expected to be even more important in PDI of more extended objects, e.g., molecules or clusters.

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[22] The EDA amplitude A_d for equal energy sharing is zero at $\theta=180°$, so that if one neglects the E2-E2 terms in Eq. (1), the parameter δ_{CD} has a singularity at $\theta=180°$. To avoid this singularity, we include the E2-E2 terms in Eq. (1) in our numerical results for δ_{CD} in Fig. 2.