Circular Dichroism in Double Photoionization

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One-photon two-electron ionization of an atom by circularly polarized light yields triply differential cross sections different for left and right circular polarization. Necessary kinematical conditions for a finite dichroism are worked out. A numerical calculation for helium which employs correlated wave functions shows the angular correlations of the escaping electron pair to be very different for left and right circular polarization such that the effect should be observable in a coincidence experiment.

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It is well known that for single photoionization of an atom the polarization of the incident light enters only kinematically into the differential cross section; see, e.g., [1]. For circular polarization the angular distribution of photoelectrons is given by

$$\frac{d\sigma}{d\Omega_p}=\frac{\sigma_0}{4\pi}[1-\tfrac{1}{2}\beta P_2(\hat{\mathbf{k}}\cdot\hat{\mathbf{p}})]\,, \tag{1}$$

where σ_0 is the total cross section, β is the asymmetry parameter, $\hat{\mathbf{k}}$ is the direction of incident light, $\mathbf{p}$ is the momentum of the photoelectron, and P_2 is the second Legendre polynomial. Equation (1) holds for right and left circularly polarized light, and also for unpolarized light. For linearly polarized light one has simply to replace β by -2β and $\hat{\mathbf{k}}$ by the direction of the electric field strength in Eq. (1). In other words, switching from one polarization to another yields no new physical information. This, however, is not true for double photoionization. It is the purpose of this Letter to show that the light polarization, in particular the helicity of circular polarization, enters dynamically in a nontrivial way into the angular correlation pattern of the photoelectron pair.

Our following analysis for double photoionization is based on first-order perturbation theory and on the dipole approximation for the radiation field. As usual we assume randomly oriented initial target atoms, no observation of the photoions, and no spin analysis of the photoelectrons. Only under these circumstances does Eq. (1) hold for single photoionization. For double photoionization the triply differential cross section (TDCS) for the pair of photoelectrons detected in coincidence including energy analysis is then given by [2–4]

$$\frac{d^3\sigma}{d\Omega_a\,d\Omega_b\,dE_a}=C\sum_{M_f}\frac{1}{2J_i+1}\sum_{M_i}|\langle\Psi^-_{\mathbf{p}_a\mathbf{p}_b}|\hat{\epsilon}\cdot\mathbf{D}|\Phi_i\rangle|^2 \tag{2}$$

with the constant $C=4\pi^2 a a_0^2 E_\gamma p_a p_b$. Here, energies E and momenta $\mathbf{p}$ of the photoelectrons are labeled by indices a and b, α is the fine-structure constant, a_0 is the Bohr radius, E_γ is the incident radiation energy, $\mathbf{D}=\sum_n \mathbf{r}_n$ is the dipole operator in length form, and $\hat{\epsilon}$ is the polarization vector. Equation (2) averages over the initial magnetic sublevels M_i, and sums over the magnetic sublevels M_f of the photoion.

We focus now on circular polarization where $\hat{\epsilon}$ is a complex unit vector and employ a tensorial calculus which formally resembles that used by Fano and Macek [5] in their polarization analysis of dipole light emitted from oriented and aligned targets. To this end we write in Eq. (2) for convenience

$$\frac{1}{2J_i+1}\sum_{M_i}|\langle\Psi^-_{\mathbf{p}_a\mathbf{p}_b}|\hat{\epsilon}\cdot\mathbf{D}|\Phi_i\rangle|^2 =\langle\Psi_{\mathbf{p}_a\mathbf{p}_b}|(\hat{\epsilon}\cdot\mathbf{D})[\hat{\epsilon}^*\cdot(S\mathbf{D})]|\Psi^-_{\mathbf{p}_a\mathbf{p}_b}\rangle\,, \tag{3}$$

where the scalar operator $S=[1/(2J_i+1)]\sum_{M_i}|\Phi_i\rangle\langle\Phi_i|$ has been introduced. Since $S\mathbf{D}$ is a vector, we recouple in the right-hand side (rhs) of Eq. (3) according to [6]

$$(\hat{\epsilon}\cdot\mathbf{D})[\hat{\epsilon}^*\cdot(S\mathbf{D})]=\tfrac{1}{3}[\mathbf{D}\cdot(S\mathbf{D})]+\tfrac{1}{2}(\hat{\epsilon}\times\hat{\epsilon}^*)\cdot[\mathbf{D}\times(S\mathbf{D})] +T_2(\hat{\epsilon},\hat{\epsilon}^*)T_2(\mathbf{D},S\mathbf{D})\,, \tag{4}$$

where T_2 stands for a tensor of rank 2. Switching from left to right circularly polarized light is described by replacing $\hat{\epsilon}$ by its complex conjugate $\hat{\epsilon}^*$. The vector product term on the rhs of Eq. (4), the only term being odd with respect to this replacement, constitutes therefore the quantity sensitive to the helicity of circular polarization. Let us make the convention $\hat{\epsilon}$ ($\hat{\epsilon}^*$) to describe left (right) circularly polarized light, and let us denote the difference of TDCSs for left and right circularly polarized light, respectively, by $\Delta=\text{TDCS(left)}-\text{TDCS(right)}$. Further simplification is achieved by choosing the z axis as the direction $\mathbf{k}$ of the incident light. With $\hat{\epsilon}=(1/\sqrt{2})(1,i,0)$ and $\hat{\epsilon}\times\hat{\epsilon}^*=-i\hat{\mathbf{k}}$ we conclude

$$\Delta=-iC\sum_{M_f}\langle\Psi^-_{\mathbf{p}_a\mathbf{p}_b}|[\mathbf{D}\times(S\mathbf{D})]_0|\Psi^-_{\mathbf{p}_a\mathbf{p}_b}\rangle\,. \tag{5}$$

Basically, this identifies Δ as the expectation value of the pseudovector $\mathbf{D}\times(S\mathbf{D})$ [7].

We must now show that the dichroism Δ given by Eq. (5) is finite. Necessary kinematical conditions for this may be derived from the tensorial structure of the matrix element in Eq. (5) which ensures that Δ has the harmonic expansion [8]

$$\Delta=-i\sum_{L_aL_b}\gamma_{L_aL_b}\mathcal{Y}_{10}^{L_aL_b}(\hat{\mathbf{p}}_a,\hat{\mathbf{p}}_b) \tag{6}$$

with real coefficients $\gamma_{L_aL_b}$ and bipolar harmonics defined

by

$$\mathcal{Y}_{lm}^{l_a l_b}(\hat{\mathbf{a}},\hat{\mathbf{b}}) = \sum_{m_a m_b} \langle l_a m_a l_b m_b | lm \rangle Y_{l_a m_a}(\hat{\mathbf{a}}) Y_{l_b m_b}(\hat{\mathbf{b}}) \quad (7)$$

which are imaginary for $(l,m)=(1,0)$ such that Δ is real. The parametrization Eq. (6) may be proven by expanding the final state into partial waves,

$$|\Psi^-_{\mathbf{p}_a\mathbf{p}_b}\rangle = \sum_{l_a l_b lm} \mathcal{Y}_{lm}^{l_a l_b}(\hat{\mathbf{p}}_a,\hat{\mathbf{p}}_b)|\Psi^-_{p_a l_a p_b l_b lm}\rangle , \quad (8)$$

and by employing a Clebsch-Gordan series for two bipolar harmonics. This series (see [8] for details) shows then that the asymmetry parameters $\gamma_{L_a L_b}$ contain Clebsch-Gordan coefficient as factors,

$$\gamma_{L_a L_b} = \sum_{l_a l_b l'_a l'_b} \langle l_a 0 l'_a 0 | L_a 0\rangle \langle l_b 0 l'_b 0 | L_b 0\rangle \Gamma^{l_a l_b l'_a l'_b}_{L_a L_b} , \quad (9)$$

where l_a, l_b as well as l'_a, l'_b are the orbital angular momenta of the electrons labeled by indices a,b, respectively. Usually, the initial state and the final ion state are eigenstates of parity. Therefore, the ejected electron pair also has well-defined parity, i.e., for all contributing angular momenta l_a, l_b the sum $l_a + l_b$ is either even or odd. The Clebsch-Gordan coefficients in Eq. (9), as well as the anisotropy coefficients $\gamma_{L_a L_b}$, are therefore finite if and only if $L_a + L_b$ is even.

The above analysis is sufficient to prove easily that there is *no* dichroism in the following cases: (1) In noncoincident experiments. (2) If the momentum $\mathbf{p}_a$ is parallel or antiparallel to $\mathbf{p}_b$. (3) If the three vectors $\mathbf{p}_a, \mathbf{p}_b$, and the direction of incident light $\mathbf{k}$ are linearly dependent. (4) If the ejected electrons have equal energy. (5) If one of the escaping photoelectrons is an s electron.

Proof.—(1) Integration of $\mathcal{Y}_{10}^{L_a L_b}(\hat{\mathbf{p}}_a,\hat{\mathbf{p}}_b)$ over one of the unit vectors yields zero because $L_a + L_b$ is even. (2) For $\hat{\mathbf{p}}_a = \hat{\mathbf{p}}_b$ we employ in the definition of the bipolar harmonic a Clebsch-Gordan series. This shows the bipolar harmonic to be proportional to the Clebsch-Gordan coefficient $\langle L_a 0 L_b 0 | 10\rangle$ which is equal to zero for even values of $L_a + L_b$. For $\hat{\mathbf{p}}_b = -\hat{\mathbf{p}}_a$ we use $Y_{lm}(-\hat{\mathbf{p}}_a) = (-1)^l Y_{lm}(\hat{\mathbf{p}}_a)$ and argue as above. (3) With the z axis in the direction $\mathbf{k}$ we parametrize $\hat{\mathbf{p}}_a$ and $\hat{\mathbf{p}}_b$ by ordinary spherical angles. All terms in the bipolar harmonics in Eq. (5) contain factors $\sin[m(\varphi_a - \varphi_b)] = 0$ because $\varphi_a - \varphi_b = \pi$ and m is an integer. (4) For equal energies the TDCS is variant with respect to the exchange $\hat{\mathbf{p}}_a \leftrightarrow \hat{\mathbf{p}}_b$. The dichroism is then equal to zero because the bipolar harmonics in Eq. (6) are odd with respect to this exchange. (5) Consider Eq. (9) and put, e.g., $l_a = l'_a = 0$. A finite value for $\gamma_{L_a L_b}$ is obtained only for $L_a = 0$. There is no even value for L_b such that the bipolar harmonic in Eq. (6) is finite.

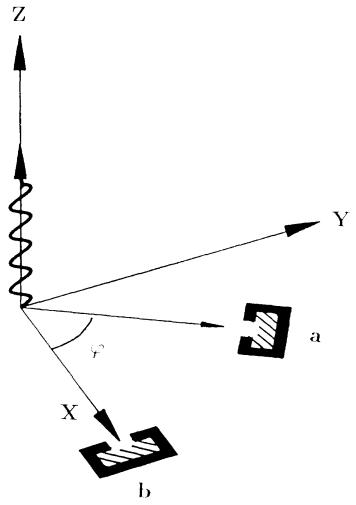

FIG. 1. Coordinate frame and experimental setup. Circularly polarized light is incident along the z direction. One electron counter (b) is in the x direction whereas the other one (a) can be rotated in the x-y plane.

In order to estimate the amount of the dichroism we have performed a numerical calculation for helium using correlated wave functions for both the initial and the final state. The ground state has been described by a Hylleraas-type function [9] given by

$$\Phi_i = N \exp[-\alpha(r_a + r_b) + \beta r_{ab}] \quad (10)$$

with $N = 1.474$, $\alpha = 1.858$, $\beta = 0.255$, and for the final state we start from a wave function which describes two outgoing electrons and satisfies exact Coulomb boundary conditions (see [10] for details),

$$\begin{aligned}\psi^-_{\mathbf{p}_a\mathbf{p}_b}(\mathbf{r}_a,\mathbf{r}_b) = {} & (2\pi)^{-3/2} e^{i\mathbf{p}_a\cdot\mathbf{r}_a} e^{\pi\alpha_a/2}\Gamma(1+i\alpha_a)\,{}_1F_1(-i\alpha_a;1;-i(p_a r_a + \mathbf{p}_a\cdot\mathbf{r}_a)) \\ & \times (2\pi)^{-3/2} e^{i\mathbf{p}_b\cdot\mathbf{r}_b} e^{\pi\alpha_b/2}\Gamma(1+i\alpha_b)\,{}_1F_1(-i\alpha_b;1;-i(p_b r_b + \mathbf{p}_b\cdot\mathbf{r}_b)) \\ & \times e^{-\pi\alpha_{ab}/2}\Gamma(1-i\alpha_{ab})\,{}_1F_1(i\alpha_{ab};1;-i(p_{ab} r_{ab} + \mathbf{p}_{ab}\cdot\mathbf{r}_{ab})) ,\end{aligned}$$

with

$$\alpha_a = -\frac{2}{p_a}, \quad \alpha_b = -\frac{2}{p_b}, \quad \alpha_{ab} = \frac{1}{2p_{ab}}, \quad \mathbf{p}_{ab} = \tfrac{1}{2}(\mathbf{p}_a - \mathbf{p}_b), \quad \mathbf{r}_{ab} = \mathbf{r}_a - \mathbf{r}_b ,$$

and employ for our calculation its odd-parity singlet part.

The experimental setup we investigate below is shown in Fig. 1. We chose a right-handed Cartesian coordinate frame with the z axis along the direction of incident light. We expect an observable dichroism, e.g., if the three vectors $\mathbf{p}_a, \mathbf{p}_b, \mathbf{k}$ are perpendicular to each other because the lowest-rank tensor in Eq. (6) shows $\Delta \propto (\mathbf{p}_a \times \mathbf{p}_b)\cdot\hat{\mathbf{k}}$; also see our statement

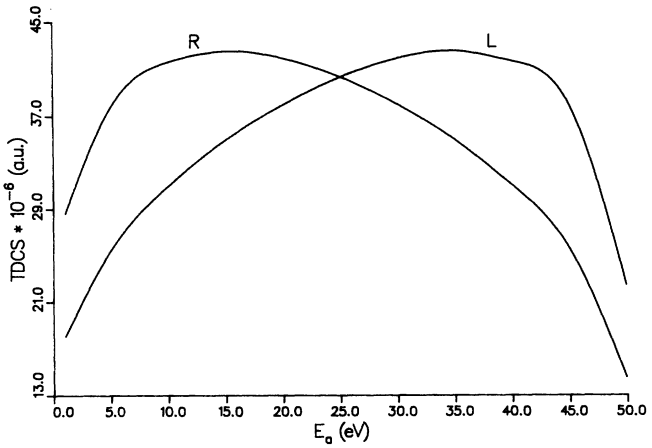

FIG. 2. The TDCS for He as a function of the energy E_a of the electron escaping in the y direction. The other electron escapes in the x direction; see Fig. 1. The total excess energy is 50 eV. L and R stand for left and right circular polarization.

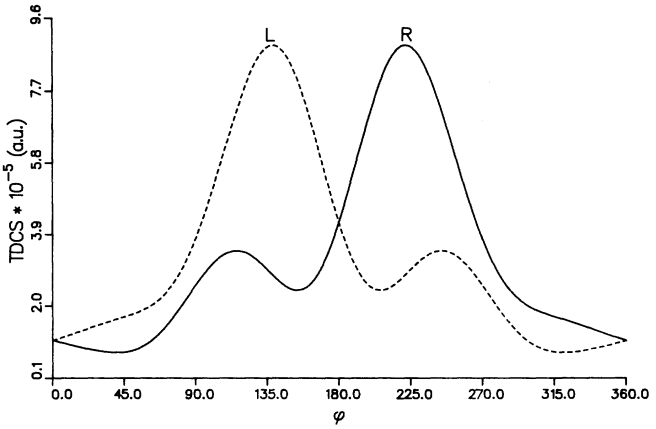

FIG. 3. Angular dependence of the TDCS for He. One electron with energy $E_b = 45$ eV is detected in the x direction, the other one with energy $E_a = 5$ eV is detected at the angle φ in the x-y plane; see Fig. 1. L and R stand for left and right circular polarization.

(3). We fix, therefore, one electron detector in the x direction perpendicular to $\hat{\mathbf{k}}$, and label this electron with the index b. In Fig. 2, the other electron (a) is detected in the y direction corresponding to $\varphi = 90°$ in Fig. 1. The total excess energy is $E_a + E_b = 50$ eV. The two curves represent the TDCS in atomic units for left (L) and right (R) circular polarization versus the energy E_b of the electron escaping in the x direction. It is clearly seen that the two TDCS are equal at 25 eV when both electrons have the same energy [statement (4)]. In this example the dichroism is largest for $E_>/E_< \approx 9$. Although the finite dichroism constitutes a broken chiral symmetry of the electron pair as a whole, Fig. 2 shows nevertheless a reduced symmetry: Except for an exchange of left and right circular polarization the two curves are invariant against reflection at the energy $E_a = E_b = 25$ eV.

Figure 3 elucidates the angular dependence of the dichroism at fixed energies. The total excess energy is again $E_a + E_b = 50$ eV with the asymmetric sharing $E_a = 5$ eV, $E_b = 45$ eV. The faster electron (b) escapes along the x direction whereas the slower one (a) escapes in the x-y plane at an angle φ relative to electron (b), i.e., $\hat{\mathbf{p}}_a \cdot \hat{\mathbf{p}}_b = \cos\varphi$; see also Fig. 1. The two TDCS curves obtained for left and right circular polarization cross each other for kinematical reasons at $\varphi = 0°$, corresponding to $\mathbf{p}_a \| \mathbf{p}_b$, and at $\varphi = 180°$, corresponding to $\mathbf{p}_a \| -\mathbf{p}_b$. Similarly as in Fig. 2 the electron pair shows a symmetry. The two TDCSs interchange under the transformation $\varphi \rightarrow 360° - \varphi$. From this it also follows that the TDCS for unpolarized light (being equal to the average of the two curves shown in Fig. 3) is invariant under $\varphi \rightarrow 360° - \varphi$. It is clearly seen that the dichroism reported in Figs. 2 and 3 is large and should be observable experimentally.

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