Circular dichroism in free-free transitions of high energy electron-atom scattering

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Abstract

We consider high energy electron scattering by hydrogen atoms in the presence of a laser field of moderate power and higher frequencies. If the field is a superposition of a linearly and a circularly polarized laser beam in a particular configuration, then we can show that circular dichroism in two photon transitions can be observed not only for the differential but also for the integrated cross sections, provided the laser-dressing of the atomic target is treated in second order perturbation theory and the coupling between hydrogenic bound and continuum states is involved.

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I. INTRODUCTION

Dichroism is a well known concept in classical optics where it denotes the property shown by certain materials of having absorption coefficients which depend on the state of polarization of the incident light \[1\]. This concept has been further extended to the case of atomic or molecular interactions with a radiation field. In particular, the notion of circular dichroism in angular distribution (CDAD) refers to the difference between the differential cross sections (DCS) of laser assisted signals for left \((L)\) and right \((R)\) circularly polarized \((CP)\) light \[2\].

Here we investigate the effect of the photon state of polarization, \(i.e.\) of its helicity, in laser-assisted high energy electron-hydrogen scattering. We show under what conditions CDAD is observable at high scattering energies as a result of \textit{target dressing} by the laser field. We consider optical frequencies and moderate field intensities and apply a hybrid calculational approach \[3\]. The interaction between the projectile and the field is treated exactly, while the interaction between the atom and the field is treated in perturbation theory. First order Born approximation is used to evaluate the scattering amplitude. We demonstrate that CDAD is encountered, provided i) the electromagnetic field is a superposition of two laser beams, one of which is linearly polarized \((LP)\) and the other is a \(CP\) field, ii) second order dressing of the target by the electromagnetic field is included. In addition, iii) the role of the virtual transitions to the continuum is shown to be essential for the observation of CDAD. Finally, we demonstrate that for a special configuration not only CDAD but also CD for the integrated cross sections can be observed. Atomic units are used.

II. THEORY

We consider electron-hydrogen scattering in the presence of an electromagnetic field that is a superposition of two laser beams. One beam is \(LP\), with polarization vector \(\vec{e}\), while the other is \(CP\) with polarization vector \(\vec{\varepsilon}\). The beams can have different directions of propagation. For simplicity, we discuss the case where the two beams have the same frequency
ω and intensity I. In dipole approximation the resulting field is

$$\vec{E}(t) = \frac{i\vec{E}_0}{2} (\vec{e} + \vec{e}^*) \exp(-i\omega t) + \text{c.c.}, \quad (1)$$

where the intensity \(I = E_0^2\). We want to know whether the DCS are sensitive to the helicity of the CP photons, defined by

$$\xi = i\vec{n} \cdot (\vec{e} \times \vec{e}^*), \quad (2)$$

which explicitly depends on the direction \(\vec{n}\) of propagation of the CP beam. As shown in [2], [4] and [5], for high energies of the projectiles CDAD does not occur for a CP laser field alone, since the first order Born approximation leads to real scattering amplitudes. We therefore present the theory for the above superposition of fields.

According to [3], at moderate laser field intensities the field-atom interaction can be described by time-dependent perturbation theory (TDPT). We consider second order dressing of the hydrogen ground state by the field (1). The approximate solution for an atomic electron in an electromagnetic field reads

$$|\Psi_1(t)\rangle = e^{-i\vec{E}_1 t} \left[ |\psi_{1s} > + |\psi_{1s}^{(1)} > + |\psi_{1s}^{(2)} > \right], \quad (3)$$

where \(|\psi_{1s} >\) is the unperturbed ground state of hydrogen, of energy \(E_{1s}\). \(|\psi_{1s}^{(1)},(2) >\) denote first and second order corrections, respectively. On account of [6] and [7] these corrections can be expressed in terms of

$$|\vec{w}_{1s}(\Omega) > = -G_C(\Omega) \vec{P} |\psi_{1s} >, \quad (4)$$

and

$$|w_{ij,1s}(\Omega',\Omega) > = G_C(\Omega') P_i G_C(\Omega) P_j |\psi_{1s} >, \quad (5)$$

where \(G_C(\Omega)\) is the Coulomb Green’s function and \(\vec{P}\) the momentum operator of the bound electron. For the field (1) there are five values of the argument of the Green’s functions necessary in order to write down the approximate solution (3), namely \(\Omega^\pm = E_1 \pm \omega, \Omega'^\pm = E_1 \pm 2\omega, \tilde{\Omega} = E_1\).

A projectile of kinetic energy \(E_k\) and momentum \(\vec{k}\), moving in the field (1), is described by the Volkov solution

$$\chi_\ell(\vec{r},t) = \frac{1}{(2\pi)^{3/2}} \exp \left\{ -iE_k t + i\vec{k} \cdot \vec{r} - i\vec{k} \cdot \vec{a}(t) \right\} \quad (6)$$
\( \vec{a}(t) \) represents the classical oscillation of the electron in the field \( \vec{E}(t) \), its amplitude is \( \alpha_0 = \sqrt{I/\omega^2} \). Using Graf’s addition theorem [8], the Fourier expansion of (6) yields a series in terms of ordinary Bessel functions \( J_N \)

\[
e^{-i\vec{k} \cdot \vec{a}(t)} = \exp\left\{-i\alpha_0 \vec{k} \cdot \vec{e} \sin \omega t - iR_k \sin (\omega t - \phi_k)\right\}
= \sum_N J_N(\mathcal{Z}_k) \exp(-iN\omega t) \exp(iN\psi_k).
\] (7)

According to the definitions of the arguments and phases given in Watson’s book [8], we have

\[
\mathcal{Z}_k = \alpha_0 |\vec{k} \cdot \vec{e} + \vec{k} \cdot \vec{\varepsilon}|, \quad R_k = \alpha_0 |\vec{k} \cdot \vec{\varepsilon}|,
\] (8)

and

\[
\exp(i\psi_k) = \frac{\vec{k} \cdot \vec{e} + \vec{k} \cdot \vec{\varepsilon}}{|\vec{k} \cdot \vec{e} + \vec{k} \cdot \vec{\varepsilon}|}, \quad \exp(i\phi_k) = \frac{\vec{k} \cdot \vec{\varepsilon}}{|\vec{k} \cdot \vec{\varepsilon}|}.
\] (9)

\( R_k \) and \( \phi_k \) refer to the \( CP \) field alone, while \( \mathcal{Z}_k \) and \( \psi_k \) are related to the superposition (11). Using (9), we recognize that a change of helicity of the \( CP \) photons, \( i.e. \ \vec{\varepsilon} \rightarrow \vec{\varepsilon}^* \), leads to a change in sign of the dynamical phases \( \phi_k \) and \( \psi_k \). Therefore, looking for the signature of helicity in the angular distributions of laser-assisted signals, we have to observe the presence of these dynamical phases in their DCS.

For high scattering energies, the first order Born approximation in terms of the interaction potential is reliable. Neglecting exchange effects, this potential is \( V(r, R) = -1/r + 1/|\vec{r} + \vec{R}| \), and the \( S \)-matrix element reads

\[
S_{B1}^{if} = -i \int_{-\infty}^{+\infty} dt <\chi_{\vec{k}_f}(t)\Psi_1(t)|V|\chi_{\vec{k}_i}(t)\Psi_1(t)>,
\] (10)

where \( \Psi_1 \) and \( \chi_{\vec{k}_{i,f}} \) are given by (3) and (6). \( \vec{k}_{i,f} \) are the initial(final) electron momenta.

The DCS for a process in which \( N \) photons are involved is

\[
\frac{d\sigma_N}{d\Omega} = (2\pi)^4 k_f(N) k_i |T_N|^2.
\] (11)

The scattered electrons have the final energy \( E_f = E_i + N\omega \) where \( N \) is the net number of photons exchanged between the colliding system and the field (11). \( N \geq 1 \) refers to absorption, \( N \leq -1 \) to emission and \( N = 0 \) describes the elastic process. The nonlinear transition matrix elements \( T_N \) in (11) have the general structure

\[
T_N = \exp(iN\psi_q) \left[ T_N^{(0)} + T_N^{(1)} + T_N^{(2)} \right].
\] (12)
ψ_q is the dynamical phase in (9) evaluated for the momentum transfer \( \vec{q} = \vec{k}_i - \vec{k}_f \). The first term in (12),

\[
T^{(0)}_N = -\frac{1}{4\pi^2} f_{el}^B J_N(\mathcal{Z}_q),
\]

yields the Bunkin-Fedorov formula [9] (target dressing is neglected). Here \( T_N = \exp(i N \phi_q) T^{(0)}_N \) and the Bessel function \( J_N(\mathcal{Z}_q) \) contains all the field intensity dependences of the transition matrix element. \( f_{el}^B \) is the amplitude of elastic scattering in the first order Born approximation, \( f_{el}^B = 2(q^2 + 8)(q^2 + 4)^{-2} \).

The remaining terms in (12) describe the dressing of the atom by the field (1), they were discussed in detail in [5]. In the case of \( T^{(1)}_N \) one of the \( N \) photons exchanged between the colliding system and the field is interacting with the bound electron, while in \( T^{(2)}_N \) two of the \( N \) photons interact with the atomic electron. These dressing terms in (12) are

\[
T^{(1)}_N = \frac{\alpha_0 \omega}{4\pi^2 q^2} \left( \frac{q}{|q|} \right) J_{1,0,1}(\tau^+, \tau^-, q)
\]

and

\[
T^{(2)}_N = \frac{\alpha_0^2 \omega^2}{8\pi^2 q^2} \left( \frac{q}{|q|} \right)
\]

\begin{align*}
&\times \{ J_{N-2}(\mathcal{Z}_q)[|\vec{q} \cdot \vec{e} + \vec{q} \cdot \vec{\varepsilon}|^2 q^{-2} \mathcal{T}_1 + (1 + 2 \vec{\varepsilon} \cdot \vec{e}) e^{-2i\psi_q} \mathcal{T}_2]

&\quad + J_{N+2}(\mathcal{Z}_q)[|\vec{q} \cdot \vec{e} + \vec{q} \cdot \vec{\varepsilon}|^2 q^{-2} \mathcal{T}_1 + (1 + 2 \vec{\varepsilon} \cdot \vec{e}^*) e^{2i\psi_q} \mathcal{T}_2]

&\quad + J_N(\mathcal{Z}_q)[|\vec{q} \cdot \vec{e} + \vec{q} \cdot \vec{\varepsilon}|^2 q^{-2} \mathcal{T}_1 + 2(1 + \Re \varepsilon^* \cdot \vec{e}) \mathcal{T}_2]\}
\end{align*}

The five radial integrals, \( J_{1,0,1}, \mathcal{T}_1, \mathcal{T}_2, \mathcal{T}_1, \mathcal{T}_2 \) in (14)-(15), depend on \( q = |\vec{q}| \) and on the parameters of the Coulomb Green’s functions. The integral \( J_{1,0,1} \) is a function of the two parameters \( \Omega^\pm \) through \( \tau^\pm = 1/\sqrt{-2\Omega^\pm} \) (see [10]), while \( \mathcal{T}_1, \mathcal{T}_2, \mathcal{T}_1, \mathcal{T}_2 \) depend on four parameters [5]. \( \mathcal{T}_1 \) and \( \mathcal{T}_2 \) are multiplied by \( J_{N\mp 2} \) if both photons are absorbed/emitted by the atomic electron. In the last line of (15), \( \mathcal{T}_1 \) and \( \mathcal{T}_2 \) are multiplied by \( J_N \). Here two photons interact with the atomic electron, but one is emitted and the other is absorbed. For our numerical calculations we used the analytic expressions for the above five radial integrals presented in [5] and [10]-[11]. Equivalent expressions were published for the case of single photon transitions in [12]-[14] and for two photon absorption/emission in [15].

The transition matrix elements for first and second order dressing in (14)-(15) are written in a form that permits to analyze their dependence on the dynamical phase \( \psi_q \). We see that \( T^{(0)}_N \) and \( T^{(1)}_N \) do not depend on the helicity [2]. On the other hand, \( T^{(2)}_N \) has an explicit dependence on \( \xi \), determined by the phase factors that multiply \( \mathcal{T}_2 \) in (15). Due to the
structure of $T_N^{(2)}$, it is evident that in the absence of the \textit{LP} component of the field \[1\] the dynamical phase $\psi_q$ is absent and hence there is no CDAD. Indeed, both polarization terms multiplying $T_2$ would be zero since $\vec{\varepsilon}^2 = \vec{\varepsilon}^* = 0$.

This demonstrates the essential role of the \textit{LP} beam and the necessity to include second order dressing of the target. In order to stress the important role of the virtual transitions to the continuum, we shall consider small scattering angles. Here the dressing of the target is considerable and the CDAD effect can be large.

**III. WEAK FIELD LIMIT**

For small arguments of the Bessel functions, \textit{i.e.} either for weak fields at any scattering angle or for moderate fields at small scattering angles, we can keep the leading terms in \[12\] only. We discuss in some detail the case $N = 2$. The corresponding matrix element is

$$T_2 = \frac{\alpha_0^2}{8 \pi^2 q^2} \left[ (\vec{q} \cdot \vec{e} + \vec{q} \cdot \vec{\varepsilon})^2 A + (1 + 2\vec{\varepsilon} \cdot \vec{\varepsilon}) B \right],$$

(16)

where the amplitudes $A$ and $B$ depend on $q$ and on $\omega$

$$A(q; \omega) = -\frac{q^2}{2^2} \left[ f_{e1}^R - \frac{4\omega}{q^2} J_{1,0,1} - \frac{4\omega^2}{q^4} T_1 \right],$$

(17)

$$B(q; \omega) = \omega^2 T_2.$$  

(18)

The DCS derived from \[16\] are

$$\frac{d\sigma_2}{d\Omega} = \frac{\alpha_0^4}{k_f k_i 2q^4} \frac{1}{|q|^4} \left| (\vec{q} \cdot \vec{e} + \vec{q} \cdot \vec{\varepsilon})^2 A + (1 + 2\vec{\varepsilon} \cdot \vec{\varepsilon}) B \right|^2$$

$$+ 2\text{Re}[(\vec{q} \cdot \vec{e} + \vec{q} \cdot \vec{\varepsilon})^2 (1 + 2\vec{\varepsilon} \cdot \vec{\varepsilon}^* A B^*)].$$

(19)

They depend on the change of helicity only if $\text{Im} A \neq 0$ and $\text{Im} B \neq 0$. As shown in \[5\] and \[10\], this is true if virtual transitions to continuum states are energetically allowed, \textit{i.e.} if $\omega > |E_1|$ or $2\omega > |E_1|$.

CDAD, defined as the difference between the DCS for \textit{LCP} and \textit{RCP}, follows from \[19\] as

$$\Delta_C = -\frac{k_f \alpha_0^4}{k_i q^4} \text{Im} Q \text{Im} (A^* B),$$

(20)

where $Q = (\vec{e} + \vec{\varepsilon})^2 (\vec{e} \cdot e + \vec{q} \cdot \vec{\varepsilon}^*)^2$ and thus $\text{Im} Q$ gives the angular dependence of $\Delta_C$. We easily verify that the three conditions i)-iii), quoted in the introduction, are all necessary to have
\[ \Delta_C \neq 0, \] namely (i) \( Q = 0 \) if the LP field is absent, (ii) \( B \) stems from second order target dressing, and (iii) \( A \) and \( B \) become real as soon as \( 2\omega < |E_1| \).

Next we study the angular dependence of \( \Delta_C \). Two cases are of major interest:

(I) If \( \vec{e} \parallel \vec{e}_i \), then the superposition of LP and CP is equivalent to elliptic polarization (EP) and we write \( \Delta_C \equiv \Delta_E \). Hence

\[ \Delta_E = \frac{k_f \alpha_0^4}{k_i q^4} q_i q_j \left( \sqrt{2} + 1 \right)^2 \text{Im} (A^*B), \tag{21} \]

where \( q_{ij} = \vec{e}_{ij} \cdot \vec{q} \) are the projections of \( \vec{q} \) on the axes \( \vec{e}_{ij} \) of the CP vector \( \vec{e} = (\vec{e}_i + i\vec{e}_j) / \sqrt{2} \). Here \( \Delta_E \) leads to CDAD but integrating it over \( \varphi \) in the azimuthal plane yields zero. Expression (21) is comparable to that of elliptic dichroism in photoionization [16].

(II) If \( \vec{e} \parallel \vec{e}_j \), then CDAD reads

\[ \Delta_C = -\frac{k_f \alpha_0^4}{k_i q^4} \left[ \frac{q_i^2}{\sqrt{2}} + q_i q_j - \frac{q_j^2}{\sqrt{2}} \right] \text{Im} (A^*B). \tag{22} \]

Taking in addition \( \vec{e} \parallel \vec{k}_i \), then the term proportional to \( q_j^2 \) gets \( \varphi \)-independent and its contribution survives in the azimuthally integrated cross sections. Thus, the choice of a privileged direction in the problem, namely that of \( \vec{k}_i \), introduces an additional asymmetry. Therefore, the signature of the photon helicity prevails not only in the angular distribution but also in the integrated cross sections. To the best of our knowledge, this is the first case of a laser-assisted process in which CD in the integrated cross sections is encountered.

Finally, we stress the importance of the form of our \( T \)-matrix element (16), since a more general structure

\[ T_2 = (\vec{e}_1 \cdot \vec{e}_2) M + (\vec{e}_1 \cdot \vec{v}_2)(\vec{e}_2 \cdot \vec{v}_1) N \] \tag{23} \]

was also found in discussions of dichroism in other processes, like two-photon ionization [17]–[19], two-photon detachment of \( \text{H}^- \) [20] or elastic X-ray scattering by ground state atoms [21]. Of course, the meaning of the vectors \( \vec{v}_{1,2} \) is specific to each process. In all these cases dichroism is caused by interferences between the real and imaginary parts of the amplitude and two terms with different angular behavior are needed to get such interferences. In the examples above, \( M \) and \( N \) were complex quantities. Our conditions, \( \omega > |E_1| \) or \( 2\omega > |E_1| \), serve the same purpose. Contrary to our problem, in photoionization or photodetachment of unpolarized systems there is no equivalent for our privileged direction \( \vec{k}_i \) and hence only CDAD is observable.
IV. RESULTS AND DISCUSSION

We present numerical results for CDAD and CD in laser-assisted electron-hydrogen scattering at high energies. We analyze the DCS for $N = \pm 2$ since here the CDAD effects are large enough. Using the above formalism, we evaluated the DCS (11) in the azimuthal plane as a function of $\varphi$ for a fixed scattering angle $\theta = 20^\circ$ and for the initial scattering energy $E_i = 100$ eV. Our laser frequency was $\omega = 10$ eV, taken close to an atomic resonance in order to enhance the CD effects, and we chose the moderate field intensity $I = 3.51 \times 10^{12}$ Wcm$^{-2}$. The initial electron momentum $\vec{k}_i$ was taken parallel to the LP vector $\vec{e}$, both pointing along the $z -$axis and the LP beam propagated in the $(x, y)$-plane.

In case (I) the $CP$ beam propagated in the $y -$direction and the corresponding $CP$ vector $\vec{\varepsilon} = (\vec{e}_y + i\vec{e}_x)/\sqrt{2}$ has helicity $\xi = 1$, known as $LCP$, while $\vec{\varepsilon}^*$ has opposite helicity $\xi = -1$, representing $RCP$. In this configuration we have $\vec{k}_i || \vec{\varepsilon} || \text{Re}(\vec{\varepsilon})$.

In Figure 1(a) we present for $N = 2$ with $E_f = E_i + 2\omega$ the $\varphi -$dependence of the DCS at $\theta = 20^\circ$. The data for $LCP$ are shown by a dotted line and for $RCP$ by a dashed line. Clearly, the laser-assisted signals depend on the helicity of the photon. In Figure 1(b) we present the results for CDAD (21). Since here $\Delta E \sim \cos \varphi$, the ”+” and ”−” in the two lobes indicate this dependence. If $\Delta E$ gets integrated over $\varphi$, the net CD effect is zero. Similar results and conclusions are obtained for $N = -2$.

For case (II) the $CP$ beam propagated in the $x -$direction and the $CP$ vector is $\vec{\varepsilon} = (\vec{e}_y + i\vec{e}_z)/\sqrt{2}$ so that now $\vec{k}_i || \vec{\varepsilon} || \text{Im}(\vec{\varepsilon})$. The LP beam propagated as before.

Figure 2 shows the DCS (11) and the CDAD (22) for $N = 2$ in panel (a) and for $N = -2$ in panel (b). Dotted lines are for $LCP$ and dashed ones for $RCP$. Full lines are used for $\Delta C$ (22) where explicitly

$$q_j^2 - \sqrt{2} q_i q_j - q_i^2 = (k_i - k_f \cos \theta)^2 - k_i^2 \sin^2 \theta \sin^2 \varphi$$

$$+ \sqrt{2} (k_i - k_f \cos \theta) k_f \sin \theta \sin \varphi. \quad (24)$$

Due to this angular dependence, the integration of $\Delta C$ over $\varphi$ does not vanish. Since the final momentum $k_f$ depends on $N$, the shape of the azimuthal dependence of $\Delta C$ is different for absorption and emission. Our data show that the maximum value of dichroism can amount up to 2/3 of the assisted signal. This is comparable to or even larger than the effect predicted for X-ray scattering [21] or for two-photon ionization [19]. Similar to the case of
X-ray scattering, the dichroism in our case is increasing with increasing laser frequency. In free-free transitions at high scattering energy the dichroic effects stem from target dressing, which is increasing with the photon frequency. Target dressing is significant for rather small scattering angles. We therefore expect that dichroism is large in this angular domain, and not near $\theta = \pi/2$ as for X-ray scattering and two-photon ionization.

V. SUMMARY AND CONCLUSIONS

Summarizing, we considered scattering of high energy electrons by hydrogen atoms in the presence of a laser field of moderate power but higher frequencies. The field had two components of equal frequency and intensity. One of the components was circularly, the other linearly polarized. The two laser beams were permitted to propagate in different directions. In the first order Born approximation we showed that in the above scattering configuration CDAD becomes observable in two-photon transitions if laser-dressing of the atomic target is carried out in second order TDPT and transitions between atomic bound and continuum states are energetically allowed, requiring higher laser frequencies. Since the scattering probabilities decrease with increasing $\omega$, we preferred to choose $\omega$ close to an atomic resonance to enhance the signals. Thanks to the above laser configuration and second order target dressing, the $T$-matrix elements become complex as a prerequisite for predicting CDAD in the Born approximation, since the elastic Born-amplitude is real.

We conclude that at high projectile energies CDAD in free-free transitions is a second order field-assisted effect occurring under special conditions only. In particular, the matrix elements of the process considered have to be complex. Similar effects might occur, if higher order terms of the Born series are taken into account, since we know that then the scattering matrix elements become complex.

Finally, we stress the role of the two asymmetries which were introduced into the scattering configuration in order to obtain helicity dependent nonlinear signals. One asymmetry came in by the $LP$ laser. This was sufficient to achieve CDAD. A second asymmetry was determined by the momentum $\vec{k}_i$ of the ingoing electrons. The two asymmetries together, more precisely $\vec{k}_i || \vec{e} || \text{Im}(\vec{\varepsilon})$, then led to CD that even persists if the DCS are integrated over $\varphi$. In photoionization and photodetachment there is no equivalent to $\vec{k}_i$. Hence, in those processes only CDAD is encountered. The analysis of the structure of the two photon
transition matrix element led us to a general formula for CDAD and to our understanding of
the physical reasons of the configuration necessary to obtain CDAD and CD, respectively.

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Figure Captions

Fig. 1: Refers to case (I), i.e. $\vec{k}_i || e || \text{Re}(\varepsilon)$. We present for $N = 2$ the DCS as function of the angle $\varphi$ at the scattering angle $\theta = 20^\circ$. The initial electron energy is $E_i = 100$ eV, the radiation frequency is $\omega = 10$ eV and its intensity $I = 3.51 \times 10^{12}$ Wcm$^{-2}$. The panel (a) shows the data for $LCP$ as dotted line and the data for $RCP$ as dashed line. Clearly, the laser-assisted signals depend on the helicity of the photons. In panel (b) the CDAD effect is visible but integration over $\varphi$ yields zero.

Fig. 2: Treats case (II), i.e. $\vec{k}_i || e || \text{Im}(\varepsilon)$. For the same parameter values as in Fig. 1, we show the $\varphi$-dependence of the DCS in panel (a) for $N = 2$ and (b) for $N = -2$. Signals for $LCP$ are dotted lines and signals for $RCP$ are dashed lines. The CDAD effects are represented by full lines. Integrating these data over $\varphi$, a non-vanishing CD effect remains. The dependence of the effects on photon emission/absorption is apparent.
