ELLiptic Dichroism in Angular Distributions in Free-Free Transitions in Hydrogen

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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 light¹. This concept has been extended to the case of atomic or molecular interactions with a radiation field. In particular, the notion of elliptic dichroism in angular distribution (EDAD) refers to the difference between the differential cross sections (DCS) of laser assisted signals for left and right elliptically polarized (EP) light².

We discuss here the effect of the photon helicity in laser induced and inverse bremsstrahlung for high energy scattering of electrons by hydrogen atoms. We demonstrate that it is possible to find EDAD for high scattering energies of the electrons if the dressing of the atomic target by the laser field is taken into account. We consider higher optical frequencies of the laser field and we restrict ourselves to the use of moderate field intensities. In this case we can employ a hybrid treatment of the problem³: the interaction between the scattered electron and the laser field is described by Volkov solutions, while the laser-dressing of the atomic electron is evaluated within the framework of time-dependent perturbation theory (TDPT). Using this approximation, we can demonstrate that EDAD becomes a non-vanishing effect provided second order TDPT is used to describe the dressing of the atomic electron by the elliptical laser field. Moreover, we analyze the role of the virtual transitions between the bound and continuum states and we show that these transitions are essential in
order to be able to predict the existence of EDAD effects.

The basic equations will be presented in section II. In section III we shall consider in some
detail the case of two photon transitions in the weak field limit. The helicity dependence of
the DCS for two photon absorption/emission by the colliding system in interaction with the
EP laser field will be discussed in section IV.

BASIC EQUATIONS

We consider free-free transitions in electron-hydrogen scattering in the presence of an EP
laser field of polarization vector \( \vec{\varepsilon} \) given by

\[
\vec{\varepsilon} = \cos (\xi/2) \left[ \vec{e}_i + i \vec{e}_j \tan (\xi/2) \right],
\]

where \( \xi \) is the ellipticity, \(-\pi/2 \leq \xi \leq \pi/2\), and \( \vec{e}_{i,j} \) are orthogonal unit vectors in the
polarization plane. We are particularly interested to know whether the DCS are sensitive
to the helicity of the EP photons, defined by \( \eta = i \vec{n} \cdot (\vec{\varepsilon} \times \vec{\varepsilon}^*) \equiv \sin \xi \), with \( \vec{n} \) the direction
of propagation of the EP laser beam. Right hand EP has \( \eta < 0 \), it corresponds to \(-\pi/2 \leq \xi < 0\). Left hand EP that has opposite helicity, \( \eta > 0 \), corresponds to \( 0 < \xi \leq \pi/2 \). For
optical frequencies we adopt the dipole approximation, thus the resulting electric field can
be described by

\[
\vec{E}(t) = i \frac{\mathcal{E}_0}{2} \vec{\varepsilon} \exp (-i\omega t) + c.c.,
\]

where the intensity of the laser field is given by \( I = \mathcal{E}_0^2 \).

We assume that at moderate laser field intensities the interaction between the laser field
and the atomic electron can be described\(^3\) by TDPT. We find it necessary to use second
order perturbation theory and, following Florescu et al\(^4\), the approximate solution for the
ground state of an electron bound to a Coulomb potential in the presence of an EP laser
field can be written in the form

\[
|\Psi_1(t)\rangle = e^{-iE_1t} \left[ |\psi_{1s}\rangle + |\psi_{1s}^{(1)}\rangle + |\psi_{1s}^{(2)}\rangle \right].
\]

Here \( |\psi_{1s}\rangle \) is the unperturbed ground state of the hydrogen atom, of energy \( E_1 \) and
\( |\psi_{1s}^{(1)},(2)\rangle \) denote the first and second order laser field dependent corrections, respectively.
We made use of the published expressions\(^4,5\) of these corrections.
The scattering electron of kinetic energy $E_k$ and momentum $\vec{k}$ in interaction with the field (2) can be described by the well known Gordon-Volkov solution

$$\chi_{\vec{k}}(\vec{r},t) = \frac{1}{(2\pi)^{3/2}} \exp \left\{ -iE_k t + i\vec{k} \cdot \vec{r} - i\vec{k} \cdot \vec{a}(t) \right\},$$  \hspace{1cm} (4)

where $\vec{a}(t)$ describes the classical oscillation of the electron in the electric field $\vec{E}(t)$. The amplitude of this oscillation is given by $a_0 = \sqrt{I/\omega^2}$. Using Graf’s addition theorem of Bessel functions, the Fourier expansion of the Gordon-Volkov solution (4) leads to a series in terms of ordinary Bessel functions $J_N$ since one has

$$\exp \left[ -i\vec{k} \cdot \alpha(t) \right] = \exp \left\{ -iR_k \sin (\omega t - \phi_k) \right\} = \sum_{N=-\infty}^{N=\infty} J_N(R_k) \exp \left[ -iN(\omega t - \phi_k) \right].$$  \hspace{1cm} (5)

Following the definitions of the arguments and phases given in Watson’s book, we can write

$$\mathcal{R}_k = a_0 \cos(\xi/2) \sqrt{(\vec{k} \cdot \vec{e}_i)^2 + (\vec{k} \cdot \vec{e}_j)^2 \tan^2(\xi/2)} \equiv a_0 |\vec{k} \cdot \vec{e}|$$  \hspace{1cm} (6)

and

$$\exp(i\phi_k) = \frac{\vec{k} \cdot \vec{e}}{|\vec{k} \cdot \vec{e}|}.$$  \hspace{1cm} (7)

We recognize that a change of sign of the helicity of the EP photons, corresponding to the replacement $\vec{e} \rightarrow \vec{e}^*$, will lead to a change in sign of the dynamical phase $\phi_k$. Therefore, by searching for the signature of helicity in the angular distributions of the scattered electrons, it will be crucial to look for the presence of the dynamical phase in the expressions of the DCS.

For high scattering energies, the first order Born approximation in terms of the interaction potential is reliable. Neglecting exchange effects, this potential is given by $V(r, R) = -1/r + 1/|\vec{r} + \vec{R}|$, where $\vec{R}$ refers to the atomic coordinates. Then, the $S$–matrix element reads

$$S_{fi}^{B1} = -i \int_{-\infty}^{+\infty} dt <\chi_{\vec{k}_f}(t)\Psi_1(t)|V|\chi_{\vec{k}_i}(t)\Psi_1(t)>,$$  \hspace{1cm} (8)

where $\Psi_1(t)$ and $\chi_{\vec{k}_i,f}(t)$ are given by the dressed atomic state (3) and by the Gordon-Volkov states (4), respectively. $\vec{k}_i(f)$ represent the initial(final) momenta of the scattered electron. After Fourier decomposition of the $S$-matrix element (8), the DCS for a scattering process involving $N$ laser photons can be written in the standard form

$$\frac{d\sigma_N}{d\Omega} = (2\pi)^4 k_f(N)|T_N|^2.$$  \hspace{1cm} (9)
$N$ is the net number of photons exchanged between the colliding system and the laser field $\mathbf{E}$, thus the scattered electrons have the final energy $E_f = E_i + N\omega$. ($N \geq 1$ refers to the absorption and $N \leq -1$ to the emission of laser quanta, while $N = 0$ corresponds to the elastic scattering process.)

In the foregoing equation (9), the nonlinear transition matrix elements $T_N$, obtained from the $S$-matrix element (8), have the following general structure

$$T_N = \exp (iN\phi_q) \left[ T_N^{(0)} + T_N^{(1)} + T_N^{(2)} \right]. \quad (10)$$

$\phi_q$ is the dynamical phase defined in (7), referring here to the momentum transfer $\mathbf{q} = \mathbf{k}_i - \mathbf{k}_f$ of the scattered electron. The first term in equation (10),

$$T_N^{(0)} = -(2\pi)^{-2} f_{el} B_1 J_N(R_q), \quad (11)$$

would yield the well-known Bunkin-Fedorov formula. $f_{el} B_1$ is the amplitude of elastic electron scattering in the first-order Born approximation: $f_{el} B_1 = 2 \frac{(q^2 + 8)}{(q^2 + 4)^2}$. The other two terms in the transition matrix element (10) are related to the atomic dressing by the laser field. These terms were discussed in considerable detail in our preceding work. The second term, $T_N^{(1)}$, refers to first order dressing of the atom in which case one of the $N$ photons exchanged between the colliding system and the radiation field is interacting with the bound electron, while the third term $T_N^{(2)}$ refers to second order dressing and here two of the $N$ photons exchanged during the scattering interact with the atomic electron. For an $EP$ field, these dressing terms are given by

$$T_N^{(1)} = \frac{\alpha_0 \omega}{4\pi^2 q^2} \frac{q \cdot \varepsilon}{\mathbf{q}} \left[ J_{N-1}(R_q) - J_{N+1}(R_q) \right] J_{1,0,1}(q; \omega), \quad (12)$$

and

$$T_N^{(2)} = \frac{\alpha_0 \omega^2}{8\pi^2 q^2} \left\{ J_{N-2}(R_q) \left[ |\mathbf{q} \cdot \varepsilon|^2 q^{-2} \mathcal{T}_1(q; \omega) + \mathcal{T}_2(q; \omega) \cos \xi e^{-2i\phi_q} \right] + J_{N+2}(R_q) \left[ |\mathbf{q} \cdot \varepsilon|^2 q^{-2} \tilde{\mathcal{T}}_1(q; \omega) + \tilde{\mathcal{T}}_2(q; \omega) \cos \xi e^{2i\phi_q} \right] + J_{N}(R_q) \left[ |\mathbf{q} \cdot \varepsilon|^2 q^{-2} \mathcal{T}_1(q; \omega) + \tilde{\mathcal{T}}_2(q; \omega) \right] \right\}. \quad (13)$$

The five radial integrals, denoted by $J_{1,0,1}$, $\mathcal{T}_1$, $\mathcal{T}_2$, $\tilde{\mathcal{T}}_1$ and $\tilde{\mathcal{T}}_2$ in the foregoing equations (12) and (13), depend not only on the absolute value of the momentum transfer $q$ but also on the photon frequency. For the numerical evaluations performed in the present work, we used
the analytic expressions for the above five radial integrals which are presented explicitly elsewhere$^{8-10}$.

The transition matrix elements $T_{N}^{(1)}$ and $T_{N}^{(2)}$ in $^{12}$-13 are written in a form which evidently permits to analyze their dependence on the dynamical phase $\phi_q$. We recognize immediately that $T_{N}^{(0)}$ and $T_{N}^{(1)}$ do not depend on the helicity of the photon. On the contrary, $T_{N}^{(2)}$ exhibits such an explicit dependence. This dependence is determined by the phase factors $e^{\pm 2i\phi_q}$ by which $T_2$ is multiplied in $^{13}$. This demonstrates the necessity to describe target dressing in second order TDPT. In order to stress the important role of the virtual transitions to the continuum, we shall analyze small scattering angles. Here the dressing of the target is considerable and the EDAD effect can be large.

**WEAK FIELD LIMIT**

For small arguments of the Bessel functions, 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 $^{13}$ only. We discuss in some detail the case of two photon absorption, $N = 2$. The corresponding matrix element is

$$T_2 = \frac{\alpha_0^2}{8\pi^2 q^2} \left[ (\vec{q} \cdot \vec{\varepsilon})^2 \mathcal{A}(q; \omega) + \cos \xi \mathcal{B}(q; \omega) \right], \quad (14)$$

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

$$\mathcal{A}(q; \omega) = -\frac{q^2}{2^2} \left[ f_{el}^{B1} - \frac{4\omega}{q^3} j_{1,0,1} - \frac{4\omega^2}{q^4} T_1 \right],$$

$$\mathcal{B}(q; \omega) = \omega^2 T_2. \quad (15)$$

If we consider $N = -2$ (i.e. two photon emission), then the complex conjugate of the EP polarization vector $\vec{\varepsilon}$ in (14) has to be used and the invariant amplitudes for the appropriate value of $q$ have to be evaluated since $q = |\vec{k}_i - \vec{k}_f|$ is a function of $N$.

The DCS formula, derived from the transition matrix element (14), turns out to be

$$\frac{d\sigma_2}{d\Omega} = \alpha_0^4 \frac{k_f}{k_i} \frac{1}{2^2 q^2} \left\{ |(\vec{q} \cdot \vec{\varepsilon})^2| \mathcal{A} |^2 + \cos^2 \xi | \mathcal{B} |^2 + 2 \cos \xi \operatorname{Re} \left[ (\vec{q} \cdot \vec{\varepsilon})^2 \mathcal{A} \mathcal{B}^* \right] \right\}. \quad (16)$$

This formula is sensitive to a change of helicity only if $\operatorname{Im} \mathcal{A} \neq 0$ and $\operatorname{Im} \mathcal{B} \neq 0$. This happens if virtual transitions to continuum states are energetically allowed$^{8-9}$. 
EDAD, defined as the difference between the DCS for left hand (LH) and right hand (RH) elliptic polarizations, follows from (16) as
\[ \Delta E = -\alpha_0 \frac{k_f}{k_i} \frac{q_i q_j}{2q^4} \sin(2\xi) \text{Im}(A^*B) \quad \text{where} \quad q_{ij} = \vec{q} \cdot \vec{e}_{ij}. \tag{17} \]
\( \Delta E \) depends on the ellipticity \( \xi \), its maximum value corresponds to \( \xi = \pi/4 \). \( \Delta E \) is also symmetric with respect to the replacement \( \xi \to \pi/2 - \xi \).

**RESULTS AND DISCUSSION**

We discuss numerical results for EDAD in laser-assisted electron-hydrogen scattering at high energies of the ingoing particle. We concentrate our analysis on the cases in which the number of exchanged photons between the scattering system and the laser field is \( N = \pm 2 \) since here the effects turn out to be large enough to be accessible to observation. On the basis of the formalism developed above, we present the DCS evaluated from (9) for a fixed scattering angle \( \theta \) as a function of the azimuth \( \varphi \). We also present the dichroism \( \Delta E \) in the same azimuthal plane. As initial energy of the scattered electrons we have taken \( E_i = 100 \text{eV} \) and we have chosen a laser frequency that is close to an atomic resonance, namely \( \omega = 10 \text{eV} \). We show numerical results for the moderate field intensity \( I = 3.51 \times 10^{12} \text{Wcm}^{-2} \). The initial electron momentum \( \vec{k}_i \) is taken to point along the \( z \)-axes; the EP laser beam propagates along the same axes.

In figure 1 we plot the DCS at \( \theta = 20^\circ \) for two photon emission \( (N=-2) \) in panel (a) and two photon absorption \( (N=2) \) in panel(b). The signals obtained for LH polarization \( (\xi = \pi/4) \) are represented by full lines and those obtained for RH polarization \( (\xi = -\pi/4) \) by dotted lines. None of the two axes of the ellipse are symmetry axes for the DCS, but a change of helicity \( LH \leftrightarrow RH \) is equivalent to a reflection in each of the planes \( xOz \) and \( yOz \). The elliptic dichroism is shown in panel (c). In this geometry
\[ q_i q_j = k_f^2 \sin^2 \theta \sin \varphi \cos \varphi \tag{18} \]
and \( \Delta E \) has an overall \( \sin(2\varphi) \) dependence that determines its four-leaved clover pattern. The outer clover leaves correspond to absorption, while the inner ones are obtained for emission. The signs of the leaves are different for emission and absorption, respectively.
The nonlinear signals exhibit also a strong ellipticity dependence, shown in figure 2, where the DCS for two photon absorption are plotted for \( LH \) elliptic polarization using three values of the ellipticity, namely \( \xi = 10^\circ \), \( \xi = 45^\circ \), and \( \xi = 80^\circ \). It is interesting to note that although the DCS may be so different, in this geometry they will always lead to a four-leaved clover pattern. According to (17) only the magnitude of the clover leaves will be modulated by the ellipticity. Summarizing we find that EDAD in free-free transitions at high projectile energies in a EP laser field can only be predicted if laser dressing of the target atom is taken into account in second order TDPT including virtual bound-continuum transitions irrespective of the scattering configuration considered.

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FIG. 1: (a) DCS for two photon emission as a function of the azimuthal angle $\varphi$ at the scattering angle $\theta = 20^\circ$. Full line is used for $LH$ elliptic polarization and dotted line for $RH$ elliptic polarization. (b) Same as panel (a) but for two photon absorption. In panel (c) the dichroism $\Delta E$ is shown. The outer clover corresponds to $N=2$, while the inner one is obtained for $N=-2$. 
FIG. 2: (a) DCS for two photon absorption ($N=2$) as a function of the azimuthal angle $\varphi$ for LH helicity and ellipticity $\xi = 10^\circ$. The scattering angle is $\theta = 10^\circ$. The rest of the parameters are the same as in figure 1(b). (b) same as in panel 2(a) but for $\xi = 45^\circ$. (c) same as in panel 2(a) but for $\xi = 80^\circ$. 
