Eur. Phys. J. D **23**, 333–336 (2003)
DOI: 10.1140/epjd/e2003-00097-5

THE EUROPEAN PHYSICAL JOURNAL D

Elliptic dichroism in hydrogen ionization by a coherent superposition of two harmonics

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Received 11 December 2002
Published online 29 April 2003 –

Abstract. The two-photon ionization of the hydrogen atom from its ground state by a two-colour electromagnetic field consisting of two odd harmonics of the same IR laser is analyzed. The influence of the state of polarization of the bichromatic field on the azimuthal angular distribution and the dependence of the elliptic dichroism on the photon frequencies are reported.

PACS. 32.80.Rm Multiphoton ionization and excitation to highly excited states (e.g., Rydberg states) – 32.80.Fb Photoionization of atoms and ions

1 Introduction

In the present work, we continue our recent investigations [1,2] regarding the circular dichroism in multiphoton two-colour ionization of hydrogen from its ground state. In [1] the bichromatic field consisted of an UV laser, circularly polarized, and its third harmonic, linearly polarized, while in [2] two odd harmonics were considered, that have different polarizations: one is linearly polarized and the other is circularly polarized. This constituted a certain generalization of earlier work by Taïeb *et al.* [3] and was stimulated by very recent experiments of Agostini and coworkers [4].

Here, we shall study the elliptic dichroism in the two-photon ionization of hydrogen by a coherent superposition of two odd harmonics of the same IR laser. In the dipole approximation the components of the bichromatic field are described by the vector potentials

$$\mathbf{A}_j(t) = \frac{1}{2}\left[A_{0j}\mathrm{e}^{-\mathrm{i}(\omega_j t+\gamma_j)}\mathbf{s}_j + \text{c.c.}\right], \quad j = 1, 2, \qquad (1)$$

where $\omega_1 = q_1\omega$, $\omega_2 = q_2\omega$ and ω is the fundamental laser frequency. A_{0j} $(j = 1, 2)$ are the amplitudes of the vector potentials of the two harmonics. The complex vectors $\mathbf{s}_j$, $(j = 1, 2)$ describe the polarizations of the components of the two-colour field and they are normalized by the condition $\mathbf{s}_j \cdot \mathbf{s}_j^* = 1$. The constant phases of the two components are denoted by γ_j, $j = 1, 2$. We shall investigate dichroic effects for identical elliptic polarizations. The two components of the bichromatic electromagnetic field propagate in the same direction, taken as the z-axis of our coordinate system.

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2 Basic equations

For ionizing the ground state of hydrogen by a bichromatic field of frequencies $q_1\omega$ and $q_2\omega$, with ω the frequency of an IR laser, we used q_1 and q_2 as two odd integers, that fulfill the condition $q_1 + q_2 > |E_1|/\hbar\omega$ (E_1 being the ground state energy). The final continuum state of energy $E = E_1 + (q_1 + q_2)\hbar\omega$ can be reached by the absorption of two different photons. Within second order time-dependent perturbation theory (TDPT) two channels of second order in the fine-structure constant α contribute to the above ionization process.

In the velocity gauge and in the dipole approximation, the amplitude of each two-photon transition is obtained from a second-order tensor with Cartesian components

$$\begin{aligned} \Pi_{ij}(\Omega) &\equiv \langle E- \mid P_i G_C(\Omega) P_j \mid E_1\rangle \\ &= \Upsilon\left[V_1(\Omega)\delta_{ij} + V_2(\Omega)n_i n_j\right], \end{aligned} \qquad (2)$$

where $\mathbf{P}$ is the momentum operator and G_C the Coulomb Green's operator. $|E-\rangle$ and $|E_1\rangle$ represent the final continuum and the initial ground state eigenvectors, respectively. The vector $|E-\rangle$ is normalized with respect to the scales of energy and solid angle. The parameter Ω has a specific value for each of the two transition channels, namely $\Omega_1 = E_1 + \hbar q_1\omega$ and $\Omega_2 = E_1 + \hbar q_2\omega$. The quantity $\Upsilon = \sqrt{m_e}/(\alpha c)$. Here m_e is the electron mass, c the speed of light, and $\mathbf{n} = \mathbf{p}/p$, with $\mathbf{p}$ being the photoelectron momentum. The dimensionless amplitudes V_1 and V_2 were derived by Gavrila [5] and by Klarsfeld [6]. They contain one or two hypergeometric Appell functions F_1 [7], respectively. The amplitudes $V_{1(2)}$ only depend on the parameter Ω of the Coulomb Green's function and on the energy of the photoelectron, E. The representation of V_1 and V_2 by these particular analytic expressions is only possible for hydrogen.

In this framework, each dimensionless amplitude of the two-photon ionization is then given by

$$\mathcal{T} = \alpha_1 (\mathbf{s}_1 \cdot \mathbf{s}_2) + \alpha_2 (\mathbf{n} \cdot \mathbf{s}_1)(\mathbf{n} \cdot \mathbf{s}_2), \tag{3}$$

where $\alpha_1 = V_1(\Omega_1) + V_1(\Omega_2)$ and $\alpha_2 = V_2(\Omega_1) + V_2(\Omega_2)$. The two amplitudes α_1 and α_2 ultimately depend on the frequencies of the two photons, hence on ω, q_1, and q_2. The expression for $\mathcal{T}$ in (3) is symmetric in $\mathbf{s}_1$ and $\mathbf{s}_2$ and hence an interchange of the states of polarization of the two fields does not change the amplitude of the process.

We analyze the configuration in which the two components of the bichromatic electromagnetic field are propagating in the same direction, taken as our z-axis. We shall discuss two distinct cases:

(i) identical elliptic polarization, described by the polarization vector $\mathbf{s} = \mathbf{e}_x \cos(\zeta/2) + \mathrm{i}\mathbf{e}_y \sin(\zeta/2)$, and
(ii) one harmonic is elliptically polarized and the other one is linearly polarized. The vectors $\mathbf{e}_x$ and $\mathbf{e}_y$ are unit vectors in the x- and y-directions, chosen along the semimajor and semiminor axes of the ellipses described by the radiation beam. The ellipticity ζ $(-\pi/2 \le \zeta \le \pi/2)$ determines the helicity $h = \sin\zeta$ of the field ($h > 0$ and $h < 0$ are for the left and the right elliptically polarized light, respectively). The values $\zeta = 0$ and $\zeta = \pm\pi/2$ correspond to linear and to circular polarizations, respectively.

In the case (i) of polarization of the photons of the two radiation beams, the differential ionization rate, evaluated in the (x, y)-plane has the form

$$\begin{aligned} G^{\pm}(\zeta, \phi) = K[&a_0(\zeta) + a_1(\zeta)\cos 2\phi \\ &+ a_2(\zeta)\cos 4\phi \pm a_3(\zeta)\sin 2\phi], \end{aligned} \tag{4}$$

where $K = \pi\alpha^2 m_e c^2 I_1 I_2/(\hbar q_1^2 q_2^2 k^4 I_0^2)$, with I_1 and I_2 being the intensities of the two components, respectively, and I_0 is the atomic unit for radiation intensity. $k \equiv \hbar\omega/|E_1|$ is a dimensionless measure for the energy of the laser photon. The signs $+$ and $-$ refer to the positive and negative helicity, respectively. ϕ describes the azimuthal direction of the ejected photoelectron momentum in the (x, y)-plane. The amplitudes a_j $(j = 0, 1, 2, 3)$ in (4) are given by

$$\begin{aligned} a_0 &= |\alpha_1|^2 \cos^2\zeta + \frac{1}{4}|\alpha_2|^2 \\ &\quad + \frac{1}{8}|\alpha_2|^2 \cos^2\zeta + \mathrm{Re}(\alpha_1\alpha_2^*)\cos^2\zeta \\ a_1 &= \left[\frac{1}{2}|\alpha_2|^2 + \mathrm{Re}(\alpha_1\alpha_2^*)\right]\cos\zeta \\ a_2 &= \frac{1}{8}|\alpha_2|^2\cos^2\zeta\ ,\ a_3 = \frac{1}{2}\mathrm{Im}(\alpha_1\alpha_2^*)\sin 2\zeta \end{aligned} \tag{5}$$

and depend on the photon frequencies through the two odd integers q_1 and q_2.

We remark that only the last term in (4) is not invariant under the transformations $\zeta \to -\zeta$ and $\phi \to -\phi$. It is responsible for the reduction of the rotational symmetry to a twofold one of the azimuthal photoelectron angular distribution (PEAD) (4), since $G^{\pm}$ is invariant *only* for $\phi \to \phi \pm \pi$ and determines the dichroic effects, characterized by

$$G^+ - G^- = \mathrm{Im}(\alpha_1\alpha_2^*)\sin 2\phi \sin 2\zeta. \tag{6}$$

Hence, the dichroic signal is modulated by the factor $\sin 2\phi$. Therefore dichroism vanishes for $\phi = 0, \pi/2$ or π, but we notice strong dichroic effects for $\zeta = \pi/4$.

In the case (ii) (one elliptically polarized and one linearly polarized harmonic) the differential ionization rate in the (x, y)-plane shows the same structure. Here, the amplitudes a_j $(j = 0, 1, 2, 3)$ are given by

$$\begin{aligned} a_0 &= |\alpha_1|^2\cos^2\frac{\zeta}{2} + \frac{1}{8}(2+\cos\zeta)\,|\alpha_2|^2 \\ &\quad + \mathrm{Re}(\alpha_1\alpha_2^*)\cos^2\frac{\zeta}{2} \\ a_1 &= \frac{1}{4}(1+\cos\zeta)|\alpha_2|^2 + \mathrm{Re}(\alpha_1\alpha_2^*)\cos^2\frac{\zeta}{2} \\ a_2 &= \frac{1}{8}|\alpha_2|^2\cos\zeta\ ,\ a_3 = \frac{1}{2}\mathrm{Im}(\alpha_1\alpha_2^*)\sin\zeta. \end{aligned} \tag{7}$$

As before, only the term $a_3 \sin 2\phi$ is responsible for the reduction of the rotational symmetry of the azimuthal angular distribution and for dichroic effects. The expression for a_3 indicates why in the present case the dichroic effects are largest for $\zeta = \pi/2$. This represents the case of circular dichroism (CD), that is observed if one of the odd harmonics is linearly polarized and the other one circularly polarized, as investigated in previous works [2,3].

The dichroic effects become transparent at the reversal of the helicity of the elliptically polarized beam and the elliptic dichroism (ED) in angular distribution can be represented by

$$ED = (G^+ - G^-)/(G^+ + G^-). \tag{8}$$

Since according to (4) $G^{\pm}$ is invariant under the operation $\phi \to \phi + \pi$, we can restrict our analysis of ED to the azimuthal range $[0, \pi]$.

3 Numerical results

We analyze elliptic dichroism (ED) in the (x, y)-plane perpendicular to the direction z of the propagation of the two harmonic radiation beams and we only consider the case of identical elliptic polarizations with $\zeta = \pi/4$, for which the dichroic effects are maximum. Moreover, we consider the situation in which the photoelectrons have the energy $E = E_1 + 12\hbar\omega$. We use the two well-known laser sources: a Ti:sapphire ($\omega = 1.55$ eV) and a Nd:YAG ($\omega = 1.17$ eV) laser.

First, we investigate the azimuthal angular distribution evaluated from equation (4). The quantity plotted

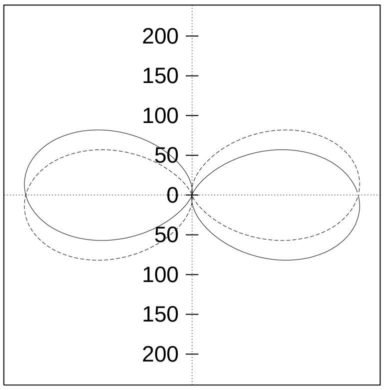

Fig. 1. The PEAD is shown in the (x, y) plane at $\theta = \pi/2$ as a polar diagram for the case (a) with $q_1 = 1$, $q_2 = 11$ for a Nd:YAG laser with $\omega = 1.17$ eV. A full line is used for positive helicity and dashed lines for negative helicity. The PEAD is considered for identical elliptic polarizations at $\zeta = \pi/4$.

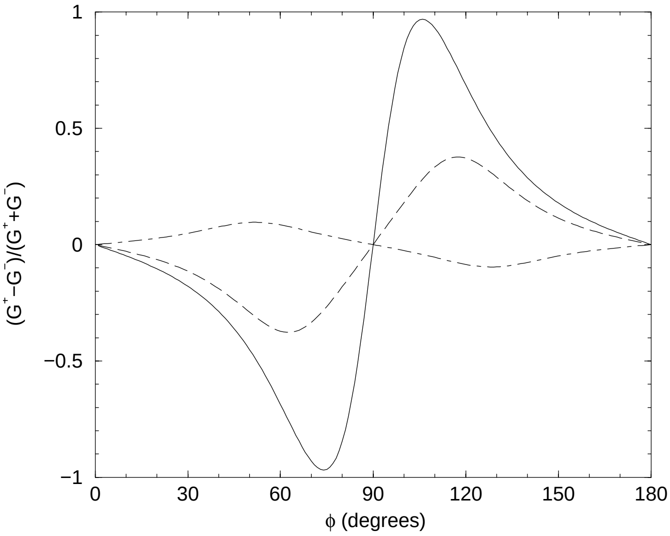

Fig. 2. The ϕ-dependence of ED for a Nd:YAG laser is presented as a Cartesian plot for $\zeta = \pi/4$ and for $q_1 = 1$, $q_2 = 11$ (full line), $q_1 = 3$, $q_2 = 9$ (dashed line) and $q_1 = 5$, $q_2 = 7$ (chain line).

is G/I_1I_2 ($\mathrm{W^{-2}\,cm^4\,s^{-1}}$) multiplied by the factor 10^{15}. Three possible combinations of odd integers q_1 and q_2 can be considered, that lead to different results, namely (a) $q_1 = 1$, $q_2 = 11$, (b) $q_1 = 3$, $q_2 = 9$, and (c) $q_1 = 5$, $q_2 = 7$. For a Nd:YAG laser, in all three cases both photons are required for ionization. Apparently, the asymmetric angular distribution reverses with the reversal of the sign of the light helicity ζ. The angular direction of the maximum of ionization changes very slowly for the different choice of the two integers. If the values of q_1 and q_2 are close to each other, the azimuthal angular distribution broadens and the asymmetry is less pronounced. The dependence of the amplitudes a_j in (5), mainly of a_0 and a_3, on the harmonic orders (given by q_1 and q_2) explains the influence of the photon frequencies on the shape of the azimuthal angular distribution. We find that the azimuthal angular distribution is aligned along a direction close to the the semimajor axis of the ellipses determined by the radiation field, that was chosen as the $x-$axis of the coordinate system. In Figure 1, we show the strong dichroic effect for the combination (a) $q_1 = 1$, and $q_2 = 11$, in which case the angle of the maximum of electron ejection for positive helicity (full line) is far away from the corresponding one for negative helicity (dashed line). Since the harmonic photon energies $9\hbar\omega$ and $11\hbar\omega$ are close to the atomic resonances located at 10.2 eV and 12.75 eV, respectively, the magnitude of the PEAD for the case (a) is appreciably larger than those for the cases (b) and (c).

In Figure 2 we illustrate as Cartesian plots for the Nd:YAG laser the dependence of ED on the azimuthal angle ϕ of the photoelectron momentum for the three combinations of integers q_1 and q_2 discussed before. As we expected, the strongest dichroic signal is achieved for the the combination $q_1 = 1$ and $q_2 = 11$ (full line). In this case, ED approaches 95 per cent for ϕ close to 75°, where it has its minimum and for ϕ close to 105°, where it has its maximum, since the frequency of the harmonic 11ω is close to the atomic resonance located at 12.75 eV. Evidently, the maximum and minimum values of ED are found symmetric with respect to $\phi = \pi/2$. The difference in the sign of ED between the data for (c) (chain line) and those for (a) (full line) and (b) (dashed line) is determined by the different sign of $\mathrm{Im}\,(\alpha_1\alpha_2^*)$ in (6). For given integers q_1 and q_2, the extrema of ED are located at the values of ϕ determined from the condition

$$a_2\cos 6\phi - (2a_0 + 3a_2)\cos 2\phi - 2a_1 = 0\ . \tag{9}$$

Since the three amplitudes a_0, a_1 and a_2 are strongly dependent on the value of the frequency, the positions of the extrema of ED are different for the three combinations (a), (b) and (c) analyzed here.

Next we consider for a Ti:sapphire laser with $\omega = 1.55$ eV the azimuthal angular distribution as Cartesian plots. Again, the plotted quantity is G/I_1I_2 ($\mathrm{W^{-2}\,cm^4\,s^{-1}}$) and is multiplied by the factor 10^{15}. We chose the same three combinations of integers q_1 and q_2, as before. Here too, the first two combinations (a) and (b) lead to an above-threshold ionization (ATI) process, since with $q_1 = 1$, $q_2 = 11$ or $q_1 = 3$, $q_2 = 9$ the hydrogen atom can be ionized directly by absorbing one photon from either the harmonic of order 11 or 9. This explains why the magnitude of PEAD's for the combinations (a) and (b) is larger than the one for (c). In Figure 3 we show the data for the most prominent case (a), again with full line for positive helicity and dashed line for negative helicity.

Correspondingly, the influence of the photon energies on the ϕ-dependence of ED is shown in Figure 4 for a Ti:sapphire laser. The three lines, full, dashed and chain, refer to the same cases (a), (b) and (c), respectively, as in Figure 2. As we expect the strongest dichroic signal is achieved for the the combination $q_1 = 5$, and $q_2 = 7$. Here ED approaches 95 per cent for ϕ close to 50°, where it has its minimum and for ϕ close to 130°, where it has its maximum. This is so, because the frequency of the harmonic of order 7 is close to the atomic resonance located at 10.2 eV.

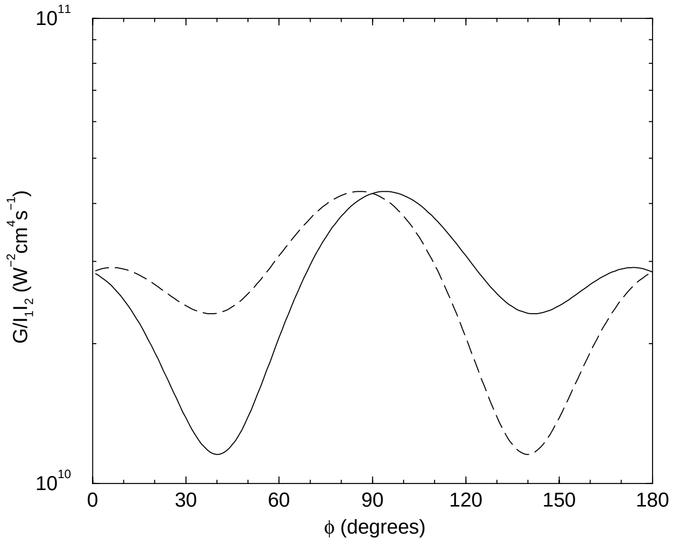

Fig. 3. Shows the PEAD as a Cartesian plot for a Ti:sapphire laser at $\omega = 1.55$ eV and for the combination (a) of harmonics with $q_1 = 1$ and $q_2 = 11$ with a full line for positive helicity and a dashed line for negative helicity at $\zeta = \pi/4$.

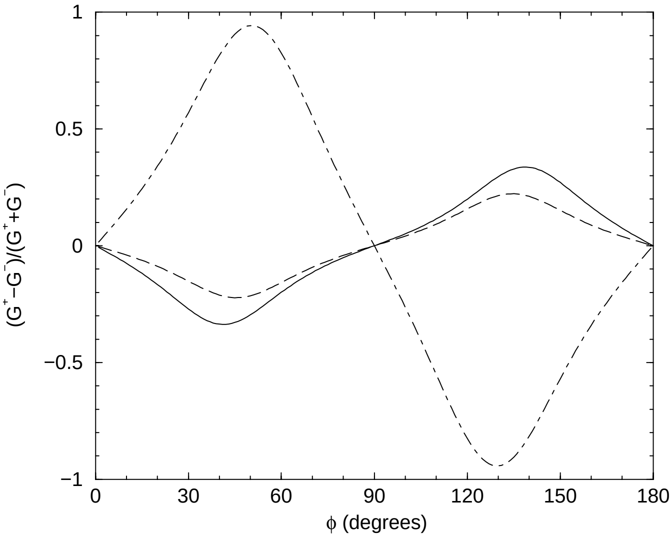

Fig. 4. Presents similar data as in Figure 2 for the ϕ-dependence of ED for a Ti:sapphire laser.

4 Conclusions

Summarizing, we have analyzed the elliptic dichroism in two-photon ionization of hydrogen from its ground state by the simultaneous action of two odd harmonics of a Nd:YAG or a Ti:sapphire laser field. The dichroic effects can be increased or decreased by the choice of the odd harmonic photon frequencies $q_1\omega$ and $q_2\omega$. Large ED effects are predicted for those combinations of odd harmonics which do not permit an ATI process by the absorption of a single harmonic photon, but where instead both harmonic photons are required for ionization and, moreover, where the frequency of one of the harmonics is close to one of the atomic resonances. This, in fact, should be true not only for the ionization of hydrogen, considered here, but also for other atoms which are more easily accessible to experimental work.

We acknowledge most valuable discussions with P. Agostini on the recent experiments on CD performed at his laboratory. This work was supported by a Special Research Project of The Austrian Academic Exchange Service for 2000/01 and partially supported by the Rumanian CNCSIS.

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