Circular Dichroism in Laser-Assisted Short-Pulse Photoionization

A. K. Kazansky,[1,2,3] A. V. Grigorieva,[4] and N. M. Kabachnik[2,5,6]
[1]*Departamento de Fisica de Materiales, UPV/EHU, E-20018 San Sebastian/Donostia, Spain*
[2]*Donostia International Physics Center (DIPC), E-20018 San Sebastian/Donostia, Spain*
[3]*IKERBASQUE, Basque Foundation for Science, E-48011 Bilbao, Spain*
[4]*Fock Institute of Physics, State University of Saint Petersburg, Saint Petersburg 198504, Russia*
[5]*Institute of Nuclear Physics, Moscow State University, Moscow 119991, Russia*
[6]*European XFEL GmbH, Albert-Einstein-Ring 19, D-22761 Hamburg, Germany*
(Received 4 July 2011; published 14 December 2011)

A remarkable effect of circular dichroism, i.e., a difference in photoelectron spectra produced by right and left circularly polarized light in two-color multiphoton ionization of atoms, is predicted for the case when the atom is ionized by an extreme ultraviolet or x-ray femtosecond pulse in the field of a strong infrared laser pulse, both pulses being circularly polarized. We show that the sidebands formed in the spectra exhibit different circular dichroism often of different signs both in angle-resolved and angle-integrated experimental conditions. The effect can be used for detecting and measuring circular polarization of x rays in a spectral range where other methods are not effective.

DOI: 10.1103/PhysRevLett.107.253002 PACS numbers: 32.80.Fb, 32.80.Rm

Two-color multiphoton ionization experiments, in which an atom is ionized by an extreme ultraviolet (XUV) or x-ray photon in the presence of a synchronized strong field of a femtosecond optical laser, have been successfully realized first with the XUV sources based on high harmonic generation [1,2]. The laser-assisted XUV photoionization of atomic systems has been successfully used in the time-resolved investigation of core-level relaxation dynamics [3]. Further application of this method to study photoemission from solid surfaces [4,5] and adsorbates [6] has demonstrated its potential to investigate ultrafast electron dynamics in solids and surface-adsorbate systems where complex, correlated electron relaxation processes are expected. As an extension of these studies, two-color experiments at free electron lasers (FELs) have proved to be an effective method of studying photoionization dynamics as well as a useful instrument for characterizing the parameters of the FEL beams [7]. In particular, the strong optical laser field modifies the photoionization continuum leading to formation of a sequence of sidebands in the photoelectron spectra which exhibit a strong dependence on the temporal separation, intensity and duration of the XUV and visible or infrared pulses [8]. In addition, advantage has been taken from the polarization properties of the XUV and infrared (IR) photons in the two-color ionization experiments [9,10] providing direct insight into symmetry and phases of the outgoing electrons.

One of the most effective methods of studying the polarization effects in photoionization is the investigation of various types of dichroism (circular or linear) in photoelectron spectra, which is defined as the intensity difference in the electron emission for two different directions of either the polarization of the ionizing photons or of the target atom polarization. Studying dichroism in the photoionization of polarized atoms with synchrotron radiation sources allowed one to obtain complete information on photoionization amplitudes including their phases ([11,12] and references therein). Circular dichroism in photoemission from solids has been widely used for the investigation of local magnetization and other properties of solids and surfaces (see, e.g., [13] and references therein).

Some years ago, it was suggested to study elliptical and circular dichroism in two-color two- and three-photon ionization of initially unpolarized atoms [14,15]. This new type of dichroism measurement in the angular distribution of photoelectrons has the potential to yield the relative magnitudes and phases of the various interfering transition amplitudes. General analysis of the elliptical and circular dichroism in two-photon ionization of atoms, with no restriction either on the detection geometry or on the propagation direction of the fields, has been given in Ref. [14]. The theory was extended to the three-photon ionization in Ref. [15], where circular dichroism in angular distribution of photoelectrons produced by linearly polarized XUV photons in the circularly polarized optical laser field was investigated. In Refs. [14,15] analytical expressions for the photoelectron angular distributions in terms of photoionization amplitudes have been given for two- and three-photon two-color ionization. However, it was noticed [15] that an extension of this method to the multiphoton case entails prohibitively cumbersome computations.

With the advent of FELs, which greatly widened the area of application of two-color experiments for investigations of electron dynamics on a femtosecond time scale, analysis of polarization effects and their potential applications becomes quite actual. Up to the present time all FELs produce linearly polarized light. However, it is planned to obtain circularly polarized beams of XUV and x-ray radiation in the near future. Thus for planning future experiments it is timely to investigate theoretically the effects of circular polarization in two-color multiphoton ionization in conditions which are characteristic for modern FELs.

In this Letter we predict a notable effect of circular dichroism (CD) in short-pulse photoionization by circularly polarized XUV or x-ray photons in the strong field of a circularly polarized optical laser. The CD can be observed in angle-resolved as well as angle-integrated photoelectron spectra. The origin of the dichroism is found in the unequal population of the magnetic substates of the continuum atomic states. Previously, we developed a simple theoretical model [16] based on the strong-field approximation (SFA) [17] which reliably describes the sideband formation in photoelectron spectra; here we extend it for the description of CD in two-color multiphoton ionization.

CD in photoelectron emission is defined as the difference in electron emission probability for right and left circularly polarized light. In one-photon absorption, the CD can appear if either the target or the photoemission process itself has certain chiral properties. Thus angular distribution of photoelectrons from unpolarized (randomly oriented) atoms or nonchiral molecules is independent of the left or right direction of photon field rotation, CD is zero. Also for multiphoton ionization by several identical photons, CD is zero [18]. However, in two-color (XUV + IR) multiphoton ionization it can be nonzero. Roughly speaking, the ionization by the circularly polarized XUV photon orients the system along the beam direction. Because of this orientation, the following absorption (or emission) of the IR photons leading to the sideband formation is different for right and left circularly polarized optical photons [11]. Thus the CD should show up in the sideband structure. For simplicity, here we consider the XUV and IR beams copropagating along the z axis. In this case, the angular distribution of photoelectrons has to be axially symmetrical with respect to this axis, depending only on polar angle θ at which the electron is detected. Moreover, it should be symmetrical with respect to the plane perpendicular to the z axis ($\theta \leftrightarrow \pi - \theta$).

To describe XUV photoionization in a strong laser field, we use the time-dependent perturbation theory and the rotating wave approximation for interaction of an atom with the XUV field, considering the influence of the laser field within the SFA [16]. This approximation is adequate for comparatively fast photoelectrons (several tens of eV). Besides, we ignore the influence of the laser field on the bound ionic and atomic states which is a sufficiently good approximation for the moderate laser fields (10^{12}–10^{13} W/cm^2) considered here. Then the photoionization amplitude can be presented as (atomic units are used throughout unless otherwise indicated)

$$\mathcal{A}_{\vec{k}} \sim -i\int_{-\infty}^{\infty} dt\tilde{\mathcal{E}}_X(t)\langle\Psi_f\psi_{\vec{k}}(t)|\hat{D}|\Psi_0\rangle e^{i(E_b-\omega_X)t}, \quad (1)$$

where $\tilde{\mathcal{E}}_X(t)$ is the envelope of the XUV pulse, ω_X is its mean frequency, $E_b = E_f - E_0$ is the binding energy (positive) of the electron, Ψ_0 and Ψ_f are the initial atomic and final ionic wave functions, respectively, and $\hat{D}$ is the dipole operator, which for the circularly polarized XUV beam with the polarization vector $\vec{\epsilon}_X^{\,\pm}$ is given by

$$\hat{D}^{\pm} = (\vec{\epsilon}_X^{\,\pm}\vec{r}) = -\sqrt{4\pi/3}\,rY_{1,\pm1}(\hat{r}), \quad (2)$$

where plus and minus signs correspond to right and left circularly polarized XUV photon, respectively, and Y_{lm} is a spherical harmonic. The wave function $\psi_{\vec{k}}(t)$ in Eq. (1) describes the "dressed" photoelectron in the laser field, which is characterized by the final (asymptotic) momentum $\vec{k}$. Within the SFA, the wave function of the photoelectron is represented by the nonrelativistic Volkov wave function [19]:

$$\psi_{\vec{k}} = \exp\{i[\vec{k} - \vec{A}_L(t)]\vec{r} - i\Phi(\vec{k}, t)\}. \quad (3)$$

Here

$$\Phi(\vec{k}, t) = \frac{1}{2}\int_t^{\infty} dt'[\vec{k} - \vec{A}_L(t')]^2 \quad (4)$$

with $\vec{A}_L(t)$ being the vector potential of the laser field, which we define as $\vec{A}_L(t) = \int_t^{\infty} dt'\vec{\mathcal{E}}_L(t')$ ($\vec{\mathcal{E}}_L(t)$ is the IR laser electric field vector). For circularly polarized laser light, $\vec{\mathcal{E}}_L(t)$ is

$$\vec{\mathcal{E}}_L(t) = \frac{1}{\sqrt{2}}\tilde{\mathcal{E}}_L(t)[\hat{x}\cos\omega_L t \pm \hat{y}\sin\omega_L t], \quad (5)$$

where $\tilde{\mathcal{E}}_L(t)$ is the envelope of the laser pulse, ω_L is its mean frequency, $\hat{x}(\hat{y})$ is a unit vector along x (y) axis, and the plus (minus) sign corresponds to the right (left) circularly polarized IR light. To simplify the expressions, hereafter we consider ionization from an s subshell. Then separating the radial integral and the phase of the p wave for vanishing IR laser field, $d_{\rm sp} \equiv \exp(-i\delta_p)\mathcal{R}_{\rm sp}$, in the matrix element in Eq. (1) and collecting Eqs. (1)–(4), one can obtain for the right circularly polarized XUV pulse the following expression

$$\mathcal{A}^R_{\vec{k}} = -i\int_{-\infty}^{\infty} dt\tilde{\mathcal{E}}_X(t)d_{\rm sp}Y_{1,+1}(\theta_0(t),\phi_0(t))e^{i\Phi(\vec{k},t)}e^{i(E_b-\omega_X)t}. \quad (6)$$

The angles (θ_0, ϕ_0) give the direction of electron emission from the atom before propagation in the optical laser field. These angles are connected with the detection angles (θ, ϕ) after propagation in the IR field by the relations:

$$\theta_0(t) = \arccos(k_z/k_0(t)), \quad (7)$$

$$\exp(i\phi_0(t)) = \frac{[k_x - A_{Lx}(t)] + i[k_y - A_{Ly}(t)]}{[k_0^2(t) - k_z^2]^{1/2}}, \qquad (8)$$

where $k_0^2(t) = [\vec{k} - \vec{A}_L(t)]^2$. Similarly one can write the amplitude for the left circularly polarized XUV pulse, $\mathcal{A}_{\vec{k}}^L$, by changing the sign of the projection of the spherical harmonic. The square of the amplitude (6) determines the differential cross section. The CD in the angular distribution (CDAD) is usually characterized by the relative value:

$$\mathrm{CDAD} = \frac{|\mathcal{A}_{\vec{k}}^R|^2 - |\mathcal{A}_{\vec{k}}^L|^2}{|\mathcal{A}_{\vec{k}}^R|^2 + |\mathcal{A}_{\vec{k}}^L|^2}. \qquad (9)$$

Naturally, in this expression the helicity of the IR pulse is fixed (left or right). Alternatively, one can obtain the same result by reversing the polarization state of the IR field while the helicity of the XUV field is kept fixed.

Here we note, that all the above formulas are presented for the case of 100% circularly polarized light. If the XUV beam is partly circularly polarized and partly unpolarized, then the CDAD is proportional to the degree of circular polarization of the beam since the numerator in Eq. (9) contains only terms proportional to the degree of circular polarization, while the denominator contains the total cross section. Thus measurements of the CD can be useful for detecting and measuring the degree of circular polarization of XUV or x-ray beams as well as for monitoring the degree of circular polarization in other experiments with circularly polarized light.

Using the above described approach, we have calculated as an example the CD for ionization of He atom by the XUV pulse with energy of 92 eV and duration 2.4 fs in the field of the IR laser (800 nm) with the intensity of 3.5×10^{12} W/cm^2 and pulse duration of 30 fs. At the chosen XUV photon energy the photoelectron energy (disregarding the effect of IR field) is 67.4 eV. Since the matrix element of the dipole operator is a slowly varying function of energy (in the considered region out of resonances), we set it equal to a constant in our calculations. In Fig. 1 (left panel) the calculated double differential cross section for right circularly polarized XUV and IR pulses is shown. One can clearly see the sidebands separated by the energy interval ~1.6 eV which corresponds to the mean IR photon energy. The number of sidebands obviously depends on the emission angle as it was shown in Ref. [16]. The maximal number of sidebands is at 90°, minimal (zero) at 0° and 180°, where the electric field of IR pulse is perpendicular to the electron momentum.

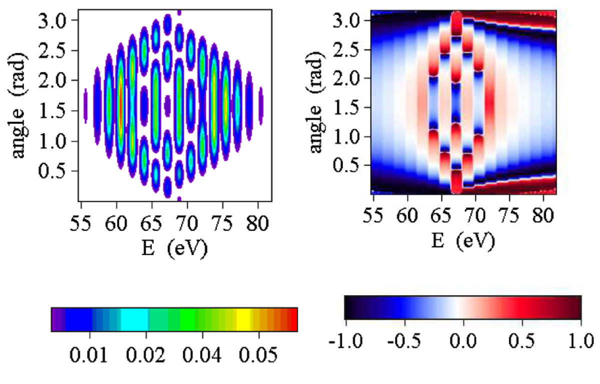

FIG. 1 (color online). Left panel: Double differential cross section (DDCS) calculated for photoionization of He by right-hand circularly polarized XUV photons at the energy 92 eV in the right-hand circularly polarized IR laser field (800 nm, 10^{12} W/cm^2). See other parameters in text. Right panel: Dichroism [Eq. (9)] calculated for the same parameters.

In Fig. 1 (right panel) the corresponding two-dimensional color-scale plot of the CDAD is shown as a function of electron energy and emission angle. Red (medium gray) color shows positive CD (cross section for right circularly polarized XUV photons is larger than for left ones). Blue (dark gray) color shows negative CD. One can see that CD strongly varies from one sideband to another. The sidebands of the lowest order (close to the central line at 67.4 eV) exhibit large CD of alternating sign at any angle. In turn, each of the lower-order sidebands shows CD quickly varying with emission angle (see discussion below). Higher-order sidebands exhibit small CD of negative sign, slowly varying with angle. We note that stripes of large CD at the angles close to zero and 180° have little meaning since the cross sections in these area are vanishingly small [see Fig. 1 (left panel)].

For a more detailed discussion of the results, in Fig. 2 we present the spectrum for right circularly polarized XUV and IR pulses and CD, calculated for a particular emission angle $\theta = 90°$, i.e., in the direction where a maximal number of sidebands can be observed. One can see that CD is practically constant within the spectral width of a sideband. It changes abruptly in between sidebands and becomes constant again. Similar spectral behavior of

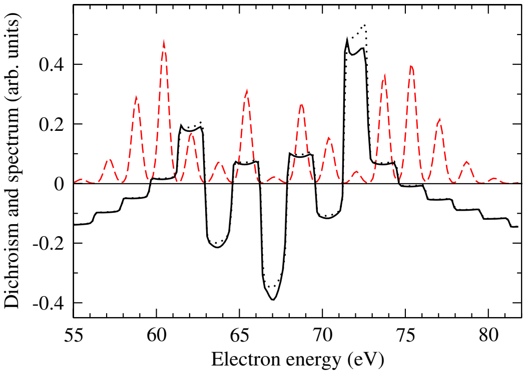

FIG. 2 (color online). Theoretical spectrum (in arbitrary units) for laser-assisted photoionization by right circularly polarized XUV light (red dashed line) and CD in absolute units (black solid line) at emission angle 90°. The dotted line shows the CD averaged over the time delay between IR and XUV pulses within one optical period. The IR laser pulse is right circularly polarized. The parameters are the same as in Fig. 1.

dichroism between two sidebands has been considered in Ref. [14]. The optimal choice for measuring the CDAD is the region of the maximum of the sideband, where the cross section is large. As we have already mentioned the CD alternates sign for lower-order sidebands, but becomes negative for higher-order sidebands. Particular behavior of the CDAD for low-order sidebands depends on the optical laser intensity and the kinetic energy of the emitted electron.

In a real two-color experiment, the phase of the IR laser is usually not stabilized. Therefore, it is important to check if the predicted effect still exists after integration over all possible phases. In calculations we simulated it by averaging over the time delay between IR and XUV pulses within one optical cycle. The result of the simulation is shown as a dotted line in Fig. 2 (similarly in Figs. 3 and 4). It is clear that the CD is practically not sensitive to the optical laser phase. This result is obvious for sufficiently long XUV pulse, covering many oscillations of the IR field. Indeed, a small shift of the XUV pulse within one optical period cannot significantly change the sideband structure formed by interference of electrons emitted in several periods. The CD which is determined by the difference of sideband intensities produced by right and left circularly polarized light remains almost unchanged when the phase is varied.

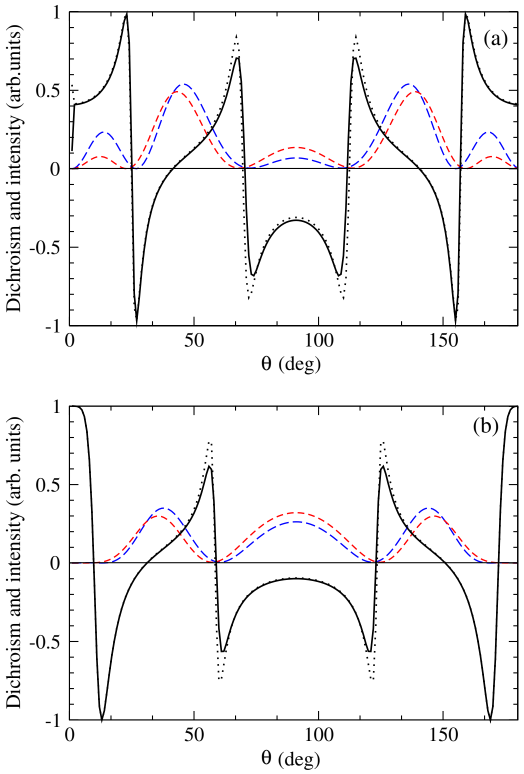

FIG. 3 (color online). Calculated angular distributions of photoelectrons (in arbitrary units) generated by right (blue long-dashed line) and left (red short-dashed line) circularly polarized light and angular distribution of CD in absolute units (black solid line): (a) for the central line at the energy of 67.35 eV and (b) for the second sideband at the energy of 70.8 eV. The dotted line shows the CD averaged over the time delay between IR and XUV pulses within one optical period. All parameters are the same as in Fig. 1.

In Fig. 3 the angular distribution of photoelectrons and angular dependence of CD is shown for the central line at 67.4 eV (a) and at the second sideband at 70.8 eV (b). As follows from symmetry considerations, the angular distributions and CD are symmetrical with respect to 90°. Angular distribution of the central line has several nodes; it is far from the $\sin^2\theta$ distribution, characteristic for the emission of p partial wave in single-photon ionization. The reason for such a complicated angular distribution is the fact that in a strong IR field the emitted electron can exchange several photons with the field. Less complicated angular distributions are expected for higher-order sidebands [compare Figs. 3(a) and 3(b)], since the probability of absorption or emission of IR photons decreases with the number of photons. Angular distributions for right and left circularly polarized photons are shifted with respect to each other. As a consequence, the CD shows a complicated behavior with zeros at the angles where the cross sections for right and left circularly polarized light are equal. A dispersive type of behavior connected with the change of sign is typical for the CDAD [15].

As we have already mentioned, the case when both XUV and IR beams are circularly polarized is equivalent to the CD in photoionization of the oriented target. Then, in contrast to the case when one of the beams is linearly polarized [15], not only the angle differential cross section, but also the angle-integrated one exhibits CD [11]. In Fig. 4 we show the spectrum integrated over all angles and corresponding dichroism. One sees, that CD in the integral cross section is much smaller (several per cent) than in the differential cross section (compare with Fig. 2). Nevertheless, it is probably easier to measure since the number of counts (statistics) should be larger in the integral case.

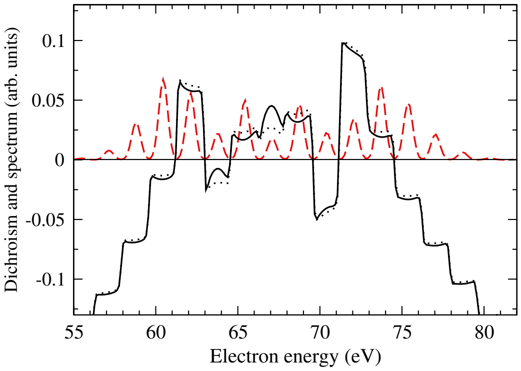

FIG. 4 (color online). Angle-integrated cross section (dashed curve) in arbitrary units and CD in absolute units (solid curve), calculated for the same parameters as in Fig. 1. The dotted line shows the CD averaged over the time delay between IR and XUV pulses within one optical period.

In conclusion, we have predicted a remarkable CD effect in two-color XUV + IR multiphoton ionization when both beams are circularly polarized. The effect can be observed both in angle-resolved and angle-integrated experiments. Particular calculations show that CD exhibits alternating sign for the neighbor sidebands. The CD effect can be used to measure circular polarization of the XUV or x-ray light in the energy region where other methods are not effective.

We are grateful to M. Meyer for many fruitful discussions. N. M. K. acknowledges the hospitality of Donostia International Physics Center (DIPC).

[1] T. E. Glover, R. W. Schoenlein, A. H. Chin, and C. V. Shank, Phys. Rev. Lett. **76**, 2468 (1996).
[2] E. S. Toma, H. G. Muller, P. M. Paul, P. Breger, M. Cheret, P. Agostini, C. Le Blanc, G. Mullot, and G. Cheriaux, Phys. Rev. A **62**, 061801(R) (2000).
[3] M. Drescher *et al.*, Nature (London) **419**, 803 (2002).
[4] L. Miaja-Avila, C. Lei, M. Aeschlimann, J. L. Gland, M. M. Murnane, H. C. Kapteyn, and G. Saathoff, Phys. Rev. Lett. **97**, 113604 (2006).
[5] G. Saathoff, L. Miaja-Avila, M. Aeschlimann, M. M. Murnane, and H. C. Kapteyn, Phys. Rev. A **77**, 022903 (2008).
[6] L. Miaja-Avila *et al.*, Phys. Rev. Lett. **101**, 046101 (2008).
[7] M. Meyer, J. T. Costello, S. Düsterer, W. B. Li, and P. Radcliffe, J. Phys. B **43**, 194006 (2010).
[8] M. Meyer*et al.*, Phys. Rev. A **74**, 011401(R) (2006).
[9] P. O'Keeffe, R. Lopez-Martens, J. Mauritsson, A. Johansson, A. L'Huillier, V. Véniard, R. Taïeb, A. Maquet, and M. Meyer, Phys. Rev. A **69**, 051401(R) (2004).
[10] M. Meyer *et al.*, Phys. Rev. Lett. **101**, 193002 (2008).
[11] N. M. Kabachnik, J. Electron Spectrosc. Relat. Phenom. **79**, 269 (1996).
[12] S. Baier, A. N. Grum-Grzhimailo, and N. M. Kabachnik, J. Phys. B **27**, 3363 (1994); A. Verweyen, A. N. Grum-Grzhimailo, and N. M. Kabachnik, Phys. Rev. A **60**, 2076 (1999).
[13] E. Arenholz, E. Navas, K. Starke, L. Baumgarten, and G. Kaindl, Phys. Rev. B **51**, 8211 (1995).
[14] N. L. Manakov, A. Maquet, S. I. Marmo, V. Veniard, and G. Ferrante, J. Phys. B **32**, 3747 (1999).
[15] R. Taïeb, V. Véniard, A. Maquet, N. L. Manakov, and S. I. Marmo, Phys. Rev. A **62**, 013402 (2000)
[16] A. K. Kazansky, I. P. Sazhina, and N. M. Kabachnik, Phys. Rev. A **82**, 033420 (2010).
[17] L. V. Keldysh, Sov. Phys. JETP **20**, 1307 (1965).
[18] P. Lambropoulos, Phys. Rev. Lett. **29**, 453 (1972).
[19] D. M. Wolkow, Z. Phys. **94**, 250 (1935).