

## CHCRUS

This is the accepted manuscript made available via CHORUS. The article has been published as:

## Nonlinear Dichroism in Back-to-Back Double Ionization of He by an Intense Elliptically Polarized Few-Cycle Extreme Ultraviolet Pulse

J. M. Ngoko Djiokap, N. L. Manakov, A. V. Meremianin, S. X. Hu, L. B. Madsen, and Anthony

F. Starace

Phys. Rev. Lett. **113**, 223002 — Published 26 November 2014 DOI: 10.1103/PhysRevLett.113.223002

## Nonlinear dichroism in back-to-back double ionization of He by an intense elliptically-polarized few-cycle XUV pulse

J.M. Ngoko Djiokap,<sup>1</sup> N.L. Manakov,<sup>2</sup> A.V. Meremianin,<sup>2</sup> S.X. Hu,<sup>3</sup> L.B. Madsen,<sup>4</sup> and Anthony F. Starace<sup>1</sup>

<sup>1</sup>Department of Physics and Astronomy, University of Nebraska, Lincoln, Nebraska 68588-0299, USA

<sup>2</sup>Department of Physics, Voronezh State University, Voronezh 394006, Russia

<sup>3</sup>Laboratory for Laser Energetics, University of Rochester, Rochester, New York 14623, USA

<sup>4</sup>Department of Physics and Astronomy, Aarhus University, DK-8000 Aarhus C, Denmark

(Dated: October 2, 2014)

Control of double ionization of He by means of the polarization and carrier-envelope-phase (CEP) of an intense, few-cycle XUV pulse is demonstrated numerically by solving the six-dimensional twoelectron, time-dependent Schrödinger equation for He interacting with an elliptically-polarized XUV pulse. Guided by perturbation theory (PT), we predict the existence of a nonlinear dichroic effect ( $\propto I^{3/2}$ ) that is sensitive to the CEP, ellipticity, peak intensity I, and temporal duration of the pulse. This dichroic effect (i.e., the difference of the two-electron angular distributions for opposite helicities of the ionizing XUV pulse) originates from interference of first- and second-order PT amplitudes, allowing one to probe and control S- and D-wave channels of the two-electron continuum. We show that the back-to-back in-plane geometry with unequal energy sharing is an ideal one for observing this dichroic effect that occurs only for an elliptically-polarized, few-cycle attosecond pulse.

PACS numbers: 32.80.Fb, 32.80.Rm, 02.70.Dh, 02.70.Hm

The quantum dynamics of two-electron atomic systems interacting with electromagnetic fields is a fundamental problem. It is well-known that electron correlation underlies the fundamental process of single-photon double ionization of He [1]. Owing to recent advances in producing XUV pulses by means of harmonic generation [2] or free-electron lasers [3–6], the nonlinear process of twophoton double ionization of He has been observed. The key role played by electron correlation in two-photon double ionization of He has subsequently been extensively investigated (see, e.g., Refs. [7–23]). All these investigations concern the case of linearly-polarized XUV pulses.

Use of elliptically-polarized light opens the possibility of investigating effects and target properties that are not accessible with linearly-polarized pulses. For example, investigations of atomic and molecular ionization by circularly- and/or elliptically-polarized ultrashort pulses have revealed "counter-intuitive angular shifts" in ionized electron angular distributions [24] (explained subsequently as due to a dynamical phase shift [25]; imprints of target orbital structures on photoelectron angular distributions [26, 27]; and the ability of a circularlypolarized pulse to serve as an attoclock for timing strong field and attosecond ionization processes [28]. In these works for atoms the three-dimensional time-dependent Schrödinger equation (TDSE) is solved using the singleactive-electron approximation. General formulations for single ionization of an atom [25] and double ionization of He [29] by an arbitrarily-polarized, few-cycle XUV pulse using perturbation theory (PT) have been validated numerically only for the case of a linearly-polarized pulse [29–32] owing to its axial symmetry, which reduces the numerical effort. None of these many prior numerical investigations has addressed the challenging

six-dimensional problem of a two-electron system interacting with an arbitrarily-polarized XUV pulse.

In this Letter we study double photoionization (DPI) of He by an intense, elliptically-polarized, few-cycle attosecond XUV pulse. Our focus is the dependence of the two-electron angular distributions on the carrierenvelope-phase (CEP) and the *helicity* of the pulse both by a PT analysis and by solving *ab initio* the sixdimensional TDSE for He. Owing to the large bandwidth of the few-cycle pulse, our numerical results reveal a new type of CEP-sensitive polarization asymmetry that is normally absent in single photon double ionization of He. The asymmetry is present in the two-electron angular distributions under a change of the rotation direction of the polarization of the attosecond pulse. The different angular distributions for opposite helicities of the pulse is our main finding, and we refer to this effect as "nonlinear dichroism" (ND). Its physical origin, within the framework of PT, is the interference of first-order  $(A_1)$  and second-order  $(A_2)$  transition amplitudes [cf. Fig. 1(a)]. In the absence of electron correlation,  $A_1 = 0$  and ND vanishes. Moreover, ND probes electron correlation on its natural timescale since ND vanishes also for long pulses.

For the pulse parameters employed here, PT in the pulse amplitude is valid and can be employed to both guide numerical calculations and interpret their results. A key fact is that experiments with elliptically-polarized pulses provide information that is in principle inaccessible to experiments with linearly-polarized pulses. We parameterize the pulse polarization vector,  $\mathbf{e}$ , as  $\mathbf{e} = (\hat{\boldsymbol{\epsilon}} + i\eta\hat{\boldsymbol{\zeta}})/\sqrt{1+\eta^2}$ , where  $\hat{\boldsymbol{\epsilon}}$  and  $\hat{\boldsymbol{\zeta}} = \hat{\mathbf{k}} \times \hat{\boldsymbol{\epsilon}}$  indicate respectively the major and minor axes of the polarization ellipse,  $\hat{\mathbf{k}}$  is the pulse propagation direction, and  $\eta$  is the ellipticity  $(-1 \leq \eta \leq +1)$ . [Note that the circular polarized polarized polarized polarized polarized pulses]



FIG. 1. (Color online) (a) Sketch of two-electron energy spectra produced when He absorbs one- or two-photons from a single 10- or 3-cycle pulse with  $\omega$ =65 eV. For the 3-cycle pulse, the one- and two-photon perturbation theory amplitudes ( $A_1$  and  $A_2$ ) overlap at  $\approx 4$  eV above the DPI threshold at 79 eV owing to the large pulse bandwidth. (b) The in-plane back-to-back (BTB) geometry for DPI of He with electron momenta  $\mathbf{p}_1$  and  $\mathbf{p}_2$  in the polarization plane orthogonal to the laser propagation direction  $\hat{\mathbf{k}}$ . The major and minor axes of the polarization ellipse are defined by the unit vectors  $\hat{\epsilon}$  and  $\hat{\zeta}$ . The BTB angle,  $\varphi$ , is the angle between  $\hat{\mathbf{p}}_1$  and  $\hat{\epsilon}$ .

ization degree,  $\xi$ , of the pulse is  $\xi = 2\eta/(1+\eta^2)$ .] Defining the triply differential probability (TDP) for DPI by  $d^3W/dEd\Omega_{\hat{\mathbf{p}}_1} d\Omega_{\hat{\mathbf{p}}_2} \equiv \mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e})$ , where  $\mathbf{p}_{1,2}$  are the electron momenta and  $E = (p_1^2 + p_2^2)/2$ , dynamical and phase information on the DPI process for a pulse with  $\xi \neq 0$  can be gained by measuring the difference of the TDPs for pulses with the electric field  $\mathbf{F}$  rotating in opposite directions, i.e., the dichroic effect [33]. We refer to this difference as the dichroism  $\Delta W_{\xi}$ ,

$$\Delta \mathcal{W}_{\xi} \equiv \mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e}) - \mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e}^*).$$
(1)

We describe the interaction of an atom in its  ${}^{1}S^{e}$ ground state with a short pulse electric field  $\mathbf{F}(t) = F_{0}(t)\operatorname{Re}[\mathbf{e} \ e^{-i(\omega t+\phi)}]$  having CEP  $\phi$ , carrier frequency  $\omega$ , and temporal envelope function  $F_{0}(t)$ . We neglect spinorbit interactions, so that both amplitudes  $A_{1}$  and  $A_{2}$  are scalars independent of the quantization axis. We adopt the same PT assumptions as in Refs. [25, 29]. Under these assumptions (see the Supplemental Material [34] for a discussion), the TDP equals

$$\mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e}) \approx \mathcal{C}[|A_1|^2 + 2\operatorname{Re}\left(A_1^*A_2\right)], \qquad (2)$$

where C is a normalization factor. The validity of Eq. (2) is determined by comparing with TDSE calculations.

Using Eq. (2) in (1), we see that  $\Delta W_{\xi}$  is comprised of two very different parts. One of them,  $\Delta W_{D1}$ , results from the interference of different terms in the first-order amplitude  $A_1$ ; it is the analog of conventional circular dichroism in single photon double ionization of He [1, 35– 41] and is linear in the pulse intensity *I*. The second part,  $\Delta W_{D12}$ , is due to interference of the first- and secondorder amplitudes, as occurs in single electron short-pulse ionization [25]. It is a nonlinear dichroic effect since  $\Delta W_{D12} \propto I^{3/2}$ . By choosing a geometry in which  $\Delta W_{D1}$ vanishes, one can thus measure the ND term  $\Delta W_{D12}$  directly. Such a geometry is back-to-back (BTB) emission of the two electrons [1]; cf. Fig. 1(b). For other geometries, PT indicates that the linear dichroism term  $\Delta W_{D1}$ is generally larger than  $\Delta W_{D12}$ . Note that  $\Delta W_{D12}$  vanishes upon averaging over the CEP  $\phi$ ; it also vanishes whenever the first-order amplitude vanishes due to selection rules. In the latter case ND originates from the interference between different terms in the second-order amplitude. This higher-order dichroism,  $\Delta W_{D2} \propto I^2$ , has the same general properties as  $\Delta W_{D12}$ , and its role is elucidated by our numerical calculations below.

The first-order PT amplitude,  $A_1$ , for single-photon DPI to the continuum  ${}^1P^o$ -state of the ionized electron pair with energy E can be parameterized as in Ref. [29]:

$$A_1 = e^{-i\phi} \left[ f_g(\rho)(\mathbf{p}_+ \cdot \mathbf{e}) + f_u(\rho)(\mathbf{p}_- \cdot \mathbf{e}) \right], \qquad (3)$$

where  $\rho \equiv (p_1, p_2, \theta)$ ,  $\theta$  is the mutual angle between the electron momenta, and  $\mathbf{p}_{\pm} \equiv (\hat{\mathbf{p}}_1 \pm \hat{\mathbf{p}}_2)/2$  (cf. [34] for discussion). The Pauli exclusion principle and parity conservation require the functions  $f_{g,u}$  to be symmetric and antisymmetric, i.e.,  $f_g(p_2, p_1, \theta) = f_g(p_1, p_2, \theta)$  and  $f_u(p_2, p_1, \theta) = -f_u(p_1, p_2, \theta)$ . Note that  $A_1$  vanishes for equal energy sharing  $(p_1=p_2)$  in the BTB geometry  $(\hat{\mathbf{p}}_1 = -\hat{\mathbf{p}}_2)$  since in that case both  $f_u(\rho)$  and  $\mathbf{p}_+$  vanish.

For an "in-plane geometry" (i.e.,  $\mathbf{p}_1$ ,  $\mathbf{p}_2$ , and  $\mathbf{e}$  all lie in the polarization plane), the first-order circular dichroism,  $\Delta W_{D1}$ , depends *only* on the degree of circular polarization  $\xi$  and is independent of both the CEP and the orientation (i.e.,  $\varphi$ ) of the polarization ellipse with respect to the electron momenta, as follows from the explicit expression for  $\Delta W_{D1}$  that one obtains using (3):

$$\frac{1}{\mathcal{C}}\Delta \mathcal{W}_{D1} = |A_1(\mathbf{e})|^2 - |A_1(\mathbf{e}^*)|^2 = \pm \xi \sin \theta \operatorname{Im}[f_g^* f_u].$$
(4)

Here  $\pm$  is the sign of the triple product  $(\hat{\mathbf{k}} \cdot [\hat{\mathbf{p}}_1 \times \hat{\mathbf{p}}_2])$ . Note that  $\Delta W_{D1}$  vanishes for the BTB geometry  $(\theta = \pi)$ .

For a sufficiently short pulse, one- and two-photon transitions (described by the first- and second-order amplitudes,  $A_1$  and  $A_2$ ) may each doubly-ionize an initial  ${}^{1}S^{e}$ -state leading to two-electron continuum states with the same energy E [cf. Fig. 1(a) for the 3-cycle case]. (Note that  $A_2$  includes both two-photon absorption and absorption/emission involving two photons.) By electric dipole selection rules,  $A_1$  leads to electron pairs in  ${}^1P^o$ states, while  $A_2$  leads to  ${}^1S^e$ - and  ${}^1D^e$ -states. In contrast to  $\Delta W_{D1}$ , the ND part of Eq. (1),  $\Delta W_{D12}$  [obtained within PT for unequal energy sharing and the BTB inplane geometry, cf. Fig. 1(b)], depends not only on  $\xi$  but also on the CEP, the orientation  $\varphi$  of the polarization ellipse, and on the product  $\xi \ell$ , where  $\ell = (1 - \eta^2)/(1 + \eta^2)$ is the degree of linear polarization. As explained in the Supplemental Material [34],  $\Delta W_{D12}$  equals

$$\Delta \mathcal{W}_{D12} = \mathcal{C}\xi \sqrt{2/(\ell+1)} \sin \varphi \operatorname{Im} \left\{ f_u^* \left[ e^{-i\phi} \left( 2h\ell - h_- \right) + (\ell \cos 2\varphi + 1) \right) + e^{i\phi} \left( 2h' + h'_- \left( \ell \cos 2\varphi + 1 \right) \right) \right\}, \quad (5)$$

where h,  $h_-$ , h', and  $h'_-$  are CEP- and  $\eta$ -independent dynamical parameters describing the amplitude  $A_2$  [34]. Equation (5) shows that the ND term  $\Delta W_{D12}$  involves the product of the dynamical parameter  $f_u^*$  of  $A_1$  [cf. Eq. (3)] and the dynamical h-parameters [34] characterizing  $A_2$ ; therefore  $\Delta W_{D12}$  vanishes unless the pulse bandwidth is sufficiently large that these parameters of  $A_1$ and  $A_2$  are nonzero at the same energy.

Unlike for linearly-polarized pulses, for ellipticallypolarized pulses the angular momentum projection Mis not conserved. This results in an "M-mixing problem" [42, 43] that we treat using ideas introduced in [43] and developed further in Refs. [24, 26–28, 44]. We solve the six-dimensional TDSE using a Finite-Element Discrete-Variable Representation and the Real-Space-Product algorithm [45] together with Wigner rotation transformations at each time step to the frame of the instantaneous electric field [24, 26–28, 42–44]. We calculate the TDP,  $\mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e})$ , for ionization of two electrons that share the energy  $E = E_1 + E_2$  above the DPI threshold, by projecting the continuum part  $\Phi_C(\mathbf{r}_1, \mathbf{r}_2, \phi, \mathbf{e})$  of the two-electron wave packet (at a time  $\approx 20$  a.u. after the pulse, ensuring convergence) onto field-free states,  $\Psi_{\mathbf{p}_1,\mathbf{p}_2}^{(-)}(\boldsymbol{r}_1,\boldsymbol{r}_2)$ , which are uncorre-lated symmetrized products of two Coulomb functions for Z = 2 [29, 46],

$$\mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e}) = |\langle \Psi_{\mathbf{p}_1, \mathbf{p}_2}^{(-)}(\mathbf{r}_1, \mathbf{r}_2) | \Phi_C(\mathbf{r}_1, \mathbf{r}_2, \phi, \mathbf{e}) \rangle|^2.$$
 (6)

Our calculations include 199 partial waves for four values of  $L: 0 \leq L \leq 3$ , so that effects of the small third-order PT amplitude are included. We assume a pulse envelope  $F_0(t) = F_0 \cos^2(\pi t/T)$  with  $-T/2 \leq t \leq T/2$ , where  $T \equiv n(2\pi/\omega)$  is the total pulse duration for n = 3 optical cycles. The temporal full-width at half-maximum of the pulse intensity profile is 0.364T = 1.1 cycles, which is comparable to those of the linearly-polarized, singlecycle pulses achieved experimentally [47, 48]. The spectral width  $\Delta \omega \simeq 1.44\omega/n$  [18] of the pulse intensity profile is 31.2 eV for  $\omega = 65$  eV (T = 190.9 as) and our peak pulse intensity is 2 PW/cm<sup>2</sup>. Significant interference, for  $\omega = 65$  eV, occurs at energies  $E \approx 4$  eV [29] above the DPI threshold energy ( $\approx 79$  eV) at which the PT amplitudes  $A_1$  and  $A_2$  are comparable [cf. Fig. 1(a)].

We present results of our numerical calculations for the BTB geometry [Fig. 1(b)] since the first-order circular dichroism  $\Delta W_{D1}$  vanishes [cf. Eq. (4)]. An additional virtue of the BTB scheme is that it guarantees a high accuracy of our numerical method in the XUV regime (with convergence of our results for a relatively low number of electron angular momenta) since the torque along the BTB axis is always zero [49]. All but one of the results in Figs. 2 – 4 are given for unequal energy sharing.

The strong CEP-dependence of the TDPs  $\mathcal{W}(\hat{\mathbf{p}}, \mathbf{e}) \equiv \mathcal{W}(\mathbf{p}_1, \mathbf{p}_2, \mathbf{e})|_{\hat{\mathbf{p}}_2 = -\hat{\mathbf{p}}_1}$  [cf. Eq. (6)] for  $\xi = \pm 0.8$  are shown in Figs. 2(a)-(d) for four CEPs. For each CEP, comparing the TDPs for  $\xi \to -\xi$  (or equivalently,  $\mathbf{e} \to \mathbf{e}^*$ ),



UES

BTB

φ=0

UES BTB

(a)

120

150

210

180

(c)

FIG. 2. (Color online) The TDP  $\mathcal{W}(\hat{\mathbf{p}}, \mathbf{e})$  (6) (in units of  $10^{-5}$  a.u.) vs.  $\varphi$  [cf. Fig. 1(b)] for DPI of He by a three-cycle XUV pulse (with  $\omega = 65$  eV, I = 2 PW/cm<sup>2</sup>, T = 190.9 as, a cos<sup>2</sup> envelope, and an ellipticity  $\eta = \pm 0.5$  or  $\xi = \pm 0.8$ ) for four CEPs: (a)  $\phi = 0$ , (b)  $\phi = \pi/3$ , (c)  $\phi = \pi/2$ , (d)  $\phi = 5\pi/6$ . All results are for the back-to-back geometry and unequal energy sharing (UES):  $E_1 = 0.7$  eV and  $E_2 = 3.3$  eV. In (c) we give for comparison  $\mathcal{W}^{(L=1)}(\mathbf{p}, \mathbf{e})$ ; see text for discussion.

150

210

one sees that the angular distributions,  $W(\hat{\mathbf{p}}, \mathbf{e})$  and  $W(\hat{\mathbf{p}}, \mathbf{e}^*)$  are mirror images of one another, which is the dichroic effect. For fixed CEP and  $\xi$ , the angular distributions are highly asymmetric under the transformation  $\varphi \to \varphi + \pi$  (or  $\hat{\mathbf{p}} \to -\hat{\mathbf{p}}$ ). In contrast, Fig. 2(c) shows the L = 1 part of the TDP,  $W^{(L=1)}(\hat{\mathbf{p}}, \mathbf{e})$ , which we find is CEP-independent and symmetric under the transformation  $\varphi \to \varphi + \pi$ . This is consistent with first-order PT, in which  $W^{(L=1)} \propto |A_1|^2$  [cf. Eq. (3)]. In the PT limit in which  $W(\hat{\mathbf{p}}, \mathbf{e}) \propto |A_1 + A_2|^2$ , the difference  $W(\hat{\mathbf{p}}, \mathbf{e}) - W(-\hat{\mathbf{p}}, \mathbf{e})$  thus measures directly the cross term  $2\text{Re}(A_1^*A_2)$ , as in the case of DPI of He by a linearly-polarized, few-cycle pulse [29].

The angular dependence of the dichroism  $\Delta W_{\xi}$  (1) is plotted in Figs. 3(a, b) for two CEPs; its dependence on ellipticity and intensity for  $\phi = \pi/2$  is shown in Figs. 3(c, d). One sees that  $\Delta W_{\xi}$ , which includes both the circular dichroism term  $\Delta W_{D1}$  (4) and higherorder dichroism terms [e.g.,  $\Delta W_{D12}$  (5) and  $\Delta W_{D2}$ ], is highly sensitive to the CEP, decreases with decreasing ellipticity, and scales approximately as  $I^{3/2}$  with intensity, with deviations originating from higher order terms. To estimate the contributions of each term, we plot in Figs. 3(a, b)  $\Delta W_{\xi}^{(L)}$  for the odd and even *L*components of the two-electron continuum wave packet, where  $\Delta W_{\xi}^{(L=1)} \approx \Delta W_{D1}$ ,  $\Delta W_{\xi}^{(L=0,2)} = \Delta W_{D2}$ , and thus  $\Delta W_{D12} \approx \Delta W_{\xi} - \Delta W_{\xi}^{(L=1)} - \Delta W_{\xi}^{(L=0,2)}$ . We see



FIG. 3. (Color online) Angular dependence of the dichroism,  $\Delta W_{\xi}$ , for two CEPs: (a)  $\phi = 0$ , and (b)  $\pi/2$ . The contributions of  $\Delta W_{\xi}^{(L=1)} \approx \Delta W_{D1}$ ,  $\Delta W_{\xi}^{(L=0,2)} = \Delta W_{D2}$ , and  $\Delta W_{D12}$  are also shown (see text for discussion). (c) Ellipticity-dependence of  $\Delta W_{\xi}$ . (d) Pulse intensitydependence of  $\Delta W_{\xi}$ ; results are scaled by  $(I/I_r)^{3/2}$ , where  $I_r = 2$  PW/cm<sup>2</sup>. Unless otherwise specified,  $\phi = \pi/2$ ,  $\xi = 0.8$ , I = 2 PW/cm<sup>2</sup>,  $E_1 = 0.7$  eV, and  $E_2 = 3.3$  eV.

that  $\Delta W_{\xi}^{(L=1)}$  is very small, consistent with 1st-order PT in which  $\Delta W_{D1}$  is zero in the BTB configuration. The non-zero  $\Delta W_{\xi}^{(L=1)}$  is CEP-independent, as expected for interference between 1st- and 3rd-order PT amplitudes.

The significance of the 2nd-order dichroism  $\Delta W_{D2}$ term depends on the CEP. For  $\phi = \pi/2$  [cf. Fig. 3(b)],  $\Delta W_{D2} \ll \Delta W_{D12}$  so that  $\Delta W_{\xi} \approx \Delta W_{D12}$  at all angles. However, for a CEP  $\phi = 0$  [cf. Fig. 3(a)] the magnitude of  $\Delta W_{D2}$  is comparable to that of  $\Delta W_{D12}$ , so that  $\Delta W_{\xi} \approx \Delta W_{D12} + \Delta W_{D2}$ . Thus for some values of  $\varphi$ , the 2nd-order part of the TDP ( $\propto |A_2|^2$ ) must be included in the PT analysis. Our results in Fig. 3 confirm the PT prediction that  $\Delta W_{D12} \sim \sin \varphi$  [cf. Eq. (5)], i.e.,  $\Delta W_{D12} = 0$  when electrons are emitted along the major axis of the pulse polarization ellipse. Figures 2 and 3 show also that  $\Delta W_{D2} = 0$  for  $\varphi = 0, \pi/2, \pi, 3\pi/2$ , indicating that  $\Delta W_{D2} \propto \sin \varphi \cos \varphi$ , as predicted by PT [34].

In Fig. 4(a) we show that the relative dichroism,  $\Delta W_{\xi}/[W(\hat{\mathbf{p}}, \mathbf{e}) + W(\hat{\mathbf{p}}, \mathbf{e}^*)]$ , is sensitive to the CEP and, for nearly all CEPs, is substantial. Its suppression for  $\phi = \pi/3$  is consistent with the similarity of the TDPs for  $\xi = \pm 0.8$  shown in Fig. 2(b); its large values near  $\varphi = \pi$  stem from the small values of the TDPs there. In Figs. 4(b, c, d) respectively we see that it decreases as one approaches equal energy sharing and as either the ellipticity or the intensity decrease.

In summary, by solving *ab initio* the six-dimensional two-electron TDSE for DPI of He by an ellipticallypolarized, intense few-cycle attosecond pulse, we have analyzed the dependence of the TDP on the pulse polar-



FIG. 4. (Color online) Angular dependence of the relative dichroism (RD),  $\Delta W_{\xi}/[W(\hat{\mathbf{p}}, \mathbf{e}) + W(\hat{\mathbf{p}}, \mathbf{e}^*)]$ , for the BTB geometry [cf. Fig. 1(b)]. Unless otherwise specified,  $\phi = \pi/2$ ,  $\xi = 0.8$ , I = 2 PW/cm<sup>2</sup>,  $E_1 = 0.7$  eV, and  $E_2 = 3.3$  eV. The panels show its sensitivity to (a) the CEP, (b) the energy sharing, (c) the ellipticity, and (d) the pulse intensity.

ization and CEP. For such few-cycle pulses, a new type of nonlinear (in the field intensity) dichroic effect in the twoelectron angular distributions [Eq. (1)] is predicted that can serve as a temporal measure of electron correlations (as it vanishes for long pulses). Our essentially exact numerical results show that, for pulse intensities that may be realized in the near future, PT can be successfully used to predict and explain characteristic features of this new polarization effect, which originates primarily from interference of the 1st- and 2nd-order PT amplitudes. Our results show that ND is highly sensitive to the pulse CEP. Accordingly, by tuning the CEP one can vary the relative contributions to the total ND of different PT amplitudes, thereby allowing one to determine their relative magnitudes. In the future, ND may be observed experimentally using reaction microscope techniques [50] with detection of electrons ionized in opposite directions in the pulse polarization plane for two helicities:  $\pm \xi$ . We note that linear dichroic effects in He have recently been employed to determine the polarization of an XUV freeelectron laser beam [51]. The ND predicted here, owing to its dependence on the large bandwidth of attosecond pulses and its sensitivity to the CEP, may be valuable for characterizing these much shorter pulses.

This work is supported in part by DOE Grant No. DE-FG03-96ER14646, by RFBR Grant No. 13-02-00420, by Russian Min. Educ. & Science Project No. 1019, by DOE NNSA Award No. DE-NA0001944, and by ERC-StG Project No. 277767 - TDMET. Our computations use Kraken (NICS) and Stampede (TACC), under Grant No. TG-PHY-120003; and Sandhills, Tusker, and Crane

at the Holland Computing Center, Univ. of Nebraska.

- [1] J.S. Briggs and V. Schmidt, J. Phys. B 33, R1 (2000).
- [2] H. Hasegawa, E.J. Takahashi, Y. Nabekawa, K.L. Ishikawa, and K. Midorikawa, Phys. Rev. A 71, 023407 (2005).
- [3] A.A. Sorokin, M. Wellhöfer, S.V. Bobashev, K. Tiedtke, and M. Richter, Phys. Rev. A 75, 051402(R) (2007).
- [4] A. Rudenko et al., Phys. Rev. Lett. 101, 073003 (2008).
- [5] M. Kurka *et al.*, New J. Phys. **12**, 073035 (2010).
- [6] A. Rudenko *et al.*, J. Phys. B **43**, 194004 (2010).
- [7] S. Laulan, H. Bachau, B. Piraux, J. Bauer, and G. Lagmago Kamta, J. Mod. Opt. 50, 353 (2003).
- [8] B. Piraux, J. Bauer, S. Laulan, and H. Bachau, Eur. Phys. J. D 26, 7 (2003).
- [9] S. Laulan and H. Bachau, Phys. Rev. A 68, 013409 (2003).
- [10] S.X. Hu, J. Colgan, and L.A. Collins, J. Phys. B 38, L35 (2005).
- [11] E. Foumouo, G. Lagmago Kamta, G. Edah, and B. Piraux, Phys. Rev. A 74, 063409 (2006).
- [12] E. Foumouo, Ph. Antoine, H. Bachau, and B. Piraux, New J. Phys. **10**, 025017 (2008).
- [13] J. Feist, S. Nagele, R. Pazourek, E. Persson, B.I. Schneider, L.A. Collins, and J. Burgdörfer, Phys. Rev. A 77, 043420 (2008).
- [14] X. Guan, K. Bartschat, and B.I. Schneider, Phys. Rev. A 77, 043421 (2008).
- [15] J. Feist, S. Nagele, R. Pazourek, E. Persson, B.I. Schneider, L.A. Collins, and J. Burgdörfer, Phys. Rev. Lett. 103, 063002 (2009).
- [16] A. Palacios, T.N. Rescigno, and C.W. McCurdy, Phys. Rev. Lett. 103, 253001 (2009).
- [17] Z. Zhang, L.-Y. Peng, Q. Gong, and T. Morishita, Opt. Express 18, 8976 (2010).
- [18] E. Foumouo, A. Hamido, Ph. Antoine, B. Piraux, H. Bachau, and R. Shakeshaft, J. Phys. B 43, 091001 (2010).
- [19] A. Palacios, D.A. Horner, T.N. Rescigno, and C.W. Mc-Curdy, J. Phys. B 43, 194003 (2010).
- [20] R. Pazourek, J. Feist, S. Nagele, E. Persson, B.I. Schneider, L.A. Collins, and J. Burgdörfer, Phys. Rev. A 83, 053418 (2011).
- [21] Z. Zhang, L.-Y. Peng, M.-H. Xu, A.F. Starace, T. Morishita, and Q. Gong, Phys. Rev. A 84, 043409 (2011).
- [22] L. Argenti, R. Pazourek, J. Feist, S. Nagele, M. Liertzer, E. Persson, J. Burgdörfer, and E. Lindroth, Phys. Rev. A 87, 053405 (2013).
- [23] W.-C. Jiang, L.-Y. Peng, W.-H. Xiong, and Q. Gong, Phys. Rev. A 88, 023410 (2013).
- [24] C.P.J. Martiny, M. Abu-samha, and L.B. Madsen, J. Phys. B 42, 161001 (2009).

- [25] E.A. Pronin, A.F. Starace, M.V. Frolov, and N.L. Manakov, Phys. Rev. A 80, 063403 (2009).
- [26] C.P.J. Martiny, M. Abu-samha, and L.B. Madsen, Phys. Rev. A 81, 063418 (2010).
- [27] M. Abu-samha and L.B. Madsen, Phys. Rev. A 84, 023411 (2011).
- [28] A.N. Pfeiffer, C. Cirelli, M. Smolarski, D. Dimitrovski, M. Abu-samha, L.B. Madsen, and U. Keller, Nature Phys. 8, 76 (2012).
- [29] J.M. Ngoko Djiokap, N.L. Manakov, A.V. Meremianin, and A.F. Starace, Phys. Rev. A 88, 053411 (2013).
- [30] L.-Y. Peng, E.A. Pronin, and A.F. Starace, New J. Phys. 10, 025030 (2008).
- [31] J.M. Ngoko Djiokap, S.X. Hu, W.-C. Jiang, L.-Y. Peng, and A.F. Starace, New J. Phys. 14, 095010 (2012).
- [32] J.M. Ngoko Djiokap, S.X. Hu, W.-C. Jiang, L.-Y. Peng, and A.F. Starace, Phys. Rev. A 88, 011401(R) (2013).
- [33] N.L. Manakov, M.V. Frolov, B. Borca, and A.F. Starace, J. Phys. B 36, R49 (2003), Sec. 1.1.
- [34] See Supplemental Material at [URL will be inserted by publisher] for a more detailed discussion of our PT analysis, including the assumptions leading to Eq. (2), the derivation of Eq. (5), and an explanation of the  $\varphi$ dependence of  $\Delta W_{D2}$  [cf. Fig. 1(b)].
- [35] J. Berakdar and H. Klar, Phys. Rev. Lett. 69, 1175 (1992).
- [36] J. Berakdar, H. Klar, A. Huetz, and P. Selles, J. Phys. B 26, 1463 (1993).
- [37] N.L. Manakov, S.I. Marmo, and A.V. Meremianin, J. Phys. B 29, 2711 (1996).
- [38] V. Mergel et al., Phys. Rev. Lett. 80, 5301 (1998).
- [39] A.S. Kheifets and I. Bray, Phys. Rev. Lett. 81, 4588 (1998).
- [40] A.Y. Istomin, N.L. Manakov, and A.F. Starace, Phys. Rev. A 69, 032713 (2004).
- [41] L. Avaldi and A. Huetz, J. Phys. B 38, S861 (2005).
- [42] H.G. Muller, Laser Physics 9, 138 (1999).
- [43] T.K. Kjeldsen, L.A.A. Nikolopoulos, and L.B. Madsen, Phys. Rev. A 75, 063427 (2007).
- [44] N.I. Shvetsov-Shilovski, D. Dimitrovski, and L.B. Madsen, Phys. Rev. A 87, 013427 (2013).
- [45] S.X. Hu, Phys. Rev. E 81, 056705 (2010).
- [46] L.B. Madsen, L.A.A. Nikolopoulos, T.K. Kjeldsen, and J. Fernández, Phys. Rev. A 76, 063407 (2007).
- [47] G. Sansone et al., Science **314**, 443 (2006).
- [48] E. Goulielmakis et al. Science **320**, 1614 (2008).
- [49] M. Gailitis, J. Phys. B 23, 85 (1990).
- [50] J. Ullrich, R. Moshammer, A. Dorn, R. Dörner, L.Ph.H. Schmidt, and H. Schmidt-Böcking, Rep. Prog. Phys. 66, 1463 (2003).
- [51] T. Mazza *et al.*, Nat. Commun. 5, 3648 (2014).