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Dichroism Effects in the Ionization of Polarized Atoms by Short Circularly Polarized Laser Pulses

Alexei V. Meremianin * and Nikolai L. Manakov 

Physics Faculty, Voronezh State University, Voronezh 394018, Russia; manakov@phys.vsu.ru

* Correspondence: meremianin@phys.vsu.ru

Abstract: The dichroism effects, i.e., asymmetries of the ionization probability with respect to the inversion of either the atomic orientation (magnetic dichroism, MD) or the circular polarization degree of the photon beam (circular dichroism, CD), are investigated using the time-dependent perturbation theory (PT). It is shown that the magnitude of these effects depends not only on the polarization states of the atom and laser pulse but also on the intensity and duration of the latter. We find that the CD can also be observed in the ionization of oriented initial bound states, which is impossible in long-pulse ionization. Predictions of our PT analysis are supported by the results obtained by numerical solutions of the time-dependent Schrödinger equation (TDSE) describing the ionization of the excited $2P$ -states of the hydrogen atom.

Keywords: ionization; attosecond pulses; polarized atoms; circular dichroism; momentum distributions

1. Introduction

Ionization is one of the most important processes for determining the interaction of light with atoms and molecules. In the non-relativistic approximation, the cross-section of atomic photo-ionization is determined by the matrix element of the electric dipole transition between the initial bound state and the final continuum state [1]. In the single active electron approximation, the ionization of an initial atomic S -state ($l_i = 0$) via the single-photon absorption leads to only the P -state of the electron in the continuum. For initial states with $l_i > 0$, single-photon absorption leads to two possible states of the photoelectron: $l_i - 1$ and $l_i + 1$. The number of different ionization channels significantly increases when spin-orbital effects and electronic correlations are taken into account. This gives rise to the question of the possibility of the experimental determination of radial parameters, describing all ionization channels. It is well known that such a “complete experiment” requires the photon beam and initial atomic state to be polarized [2–6]. The process of the ionization of polarized atoms has been extensively studied, in both the single-photon [4,7–12] and many-photon regimes [13–18]. Recent experimental progress has made it possible to study the ionization of atoms and molecules through ultrashort (few-cycle) laser pulses [19–22]. Thus, it is of interest to analyze photon polarization effects arising in the ionization of polarized atoms by few-cycle electromagnetic pulses. First of all, we note that the dependence of the ionization probability on the polarizations of the ionizing pulse and atomic target, as well as on the direction of the photoelectron emission, is very complicated. Therefore, we limit our consideration only to the study of the asymmetries of the momentum distributions (MDs) of photoelectrons, which are referred to as the “dichroism effects”. In the case of circularly polarized (CP) laser pulses, two kinds of dichroism effects take place [23]:

- Circular dichroism in MD (CDMD), i.e., the difference in the differential ionization probability with respect to the rotation directions of the electric field of the ionizing pulse;
- Magnetic dichroism in MD (MDMD), i.e., the difference in the differential probability for two opposite directions of the atomic orientation.



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We emphasize that the above effects only take place in the ionization of *polarized* atoms, and they are absent in the ionization of unpolarized initial atomic states. For ultrashort (few-cycle) laser pulses, the ionization probability depends not only on the electron emission angles but also on its energy, which introduces an additional degree of freedom into the problem. The goal of the present work is to analyze the dependence of the ionization probability on both the electron emission angles and the electron momentum p in the case of the ionization of polarized atoms by few-cycle circularly polarized laser pulses.

In this article, we apply non-stationary perturbation theory (PT) to analyze the MDs of photoelectrons emitted by polarized atoms subjected to an ultrashort (i.e., few-cycle) CP electromagnetic pulse. First, in Section 2, we derive the parameterization of the ionization amplitude, A , in which its dependence on the photoelectron momentum p and the emission angle φ is separated. Next, in Section 3, we present expressions for the triply differential probability (TDP) of ionization. For the sake of simplicity, we limit our consideration to the case of purely polarized initial atomic states oriented or aligned along the pulse propagation direction. In Section 3.2, we discuss the novel CD effect arising in the ionization of aligned initial atomic states. Such an effect is absent in the case of ionization by monochromatic (i.e., long) pulses. This effect is of importance since aligned states naturally occur when atoms are excited by a linearly polarized light. In Section 4, we apply our PT analysis to interpret the numerical results in the ionization of the polarized 2P-states of the hydrogen atom. In Section 5, we discuss the derived results and the possibility of the experimental observation of the predicted dichroism effects. Section 6 contains some concluding remarks. Atomic units are used throughout the text unless otherwise specified.

2. Parameterization of the Ionization Amplitude

In the electric dipole approximation, the time-dependent Schrödinger equation describing the light–atom interaction has the form:

$$(H_0 - V_d)\Psi = i \frac{\partial \Psi}{\partial t}, \quad (1)$$

where H_0 is the unperturbed atomic Hamiltonian, Ψ is the atomic wave function, and the interaction operator, V_d , is defined by

$$V_d = (\mathbf{F} \cdot \mathbf{d}), \quad (2)$$

where $\mathbf{d}$ is the operator of the total electric dipole momentum of atomic electrons. For few-cycle pulses, the electric field-strength vector can be written as

$$\mathbf{F}(t) = \text{Re} \left(\mathbf{e} F_0(t) e^{-i(\omega t + \phi)} \right). \quad (3)$$

Here, ω is the pulse carrier frequency, $F_0(t)$ is the smooth pulse envelope function, ϕ is the carrier-envelope phase (CEP), and $\mathbf{e}$ is the complex vector of the pulse polarization. For CP pulses, the polarization vector can be written in the following form [24]

$$\mathbf{e} = \frac{1}{\sqrt{2}}(\mathbf{e}_x + i\zeta\mathbf{e}_y), \quad \zeta = \pm 1, \quad (4)$$

where $\mathbf{e}_x, \mathbf{e}_y$ are unit vectors of the Cartesian basis whose z-axis is directed along the pulse propagation. For right-hand CP (RCP) pulses, one has $\zeta = 1$, whereas for left-hand CP (LCP) pulses, one has $\zeta = -1$. The polarization vector is normalized by the condition $(\mathbf{e}^* \cdot \mathbf{e}) = 1$. Note also that for purely CP pulses, $(\mathbf{e} \cdot \mathbf{e}) = 0$.

In the first-order non-stationary PT, the ionization amplitude is defined by [25]:

$$A_{l_i m_i}^{(1)} = i \int_{-\infty}^{\infty} \langle \nu \mathbf{p} | \mathbf{d} \cdot \mathbf{F}(t) | l_i m_i \rangle e^{i(E - E_i)t} dt. \quad (5)$$

Here, $|l_i m_i\rangle$ is the initial state, with energy E_i , angular momentum l_i , and its projection m_i . $|\nu \mathbf{p}\rangle$ is the final state that satisfies the incoming wave boundary conditions, comprising the bound state $|\nu\rangle$ of the residual ion with energy E_ν and the continuum state $|\mathbf{p}\rangle$ of the electron, with momentum $\mathbf{p}$ and energy $E = p^2/2$. Although the integration limits in (5) should correspond to the pulse duration, it is convenient to extend those limits to infinity. The integral (5) can be calculated as follows

$$\int_{-\infty}^{\infty} \mathbf{F}(t) e^{i(E-E_i)t} dt = \frac{\mathbf{e}}{2} e^{-i\phi} \int_{-\infty}^{\infty} F_0(t) e^{i(E-E_i-\omega)t} dt + \frac{\mathbf{e}^*}{2} e^{i\phi} \int_{-\infty}^{\infty} F_0(t) e^{i(E-E_i+\omega)t} dt. \quad (6)$$

By introducing the Fourier transform $\hat{F}_0^\pm$ as

$$\hat{F}_0^\pm(\epsilon) = \int_{-\infty}^{\infty} F_0(t) e^{i(\epsilon \mp \omega)t} dt, \quad (7)$$

the above Equation (6) can be re-written as

$$\int_{-\infty}^{\infty} \mathbf{F}(t) e^{i(E-E_i)t} dt = \frac{e^{-i\phi}}{2} \hat{F}_0^+(E-E_i) \mathbf{e} + \frac{e^{i\phi}}{2} \hat{F}_0^-(E-E_i) \mathbf{e}^*. \quad (8)$$

In the case of the ionization process, the second term on the right-hand side of this identity can be neglected because $|E-E_i-\omega| \ll E-E_i+\omega$ and $|\hat{F}_0(\epsilon)|$ has a maximum at $\epsilon \approx \omega$. (Note that $E_i < 0$ for the initial bound state). Thus, the ionization amplitude (5) becomes

$$A_{l_i m_i}^{(1)} = \frac{e^{-i\phi}}{2} \hat{F}_0^+(E-E_i) \langle \nu \mathbf{p} | \mathbf{e} \cdot \mathbf{d} | l_i m_i \rangle. \quad (9)$$

Here, it can be seen that the dependence of the first-order PT amplitude on the pulse envelope is factorized. However, this is not so for higher PT orders (see Equation (12) below). By using the partial (i.e., multipole) expansion for the final state, the angular dependence of the dipole matrix element in (9) can be separated out using the standard methods of the quantum angular momentum technique [16,26,27]. This leads to the following expression for the ionization amplitude in the first PT order:

$$A_{l_i m_i}^{(1)} = e^{-i\phi} \sum_{l=l_i \pm 1} \mathcal{A}_{ll_i}^{(1)} \{ \mathbf{e} \otimes Y_l(\hat{\mathbf{p}}) \}_{l_i m_i}, \quad (10)$$

where $\mathcal{A}_{ll_i}^{(1)}$ represents the dynamic amplitude parameters, involving radial dipole matrix elements multiplied by the Fourier transform of the pulse envelope, $F_0^+(E-E_i) \langle \nu l | d | l_i \rangle$. The parameters $\mathcal{A}_{ll_i}^{(1)}$ are independent of the pulse polarization and the emission angle of the photoelectron, and the expression in curly brackets in (10) is the irreducible tensor product [26]:

$$\{ \mathbf{e} \otimes Y_l(\hat{\mathbf{p}}) \}_{l_i m_i} = \sum_{\mu=0, \pm 1} C_{1\mu l m_i}^{l_i m_i} e_\mu Y_{lm}(\hat{\mathbf{p}}), \quad m = m_i - \mu, \quad (11)$$

where e_μ is the μ -th spherical component of the photon polarization vector and $C_{1\mu l m_i}^{l_i m_i}$ is the Clebsch–Gordan coefficient. The parity conservation law in the dipole approximation demands the condition $(-1)^{l_i+l+1} = 1$ to be fulfilled. In the coordinate frame, whose z-axis is directed along the photon beam propagation, the non-zero spherical components of the polarization vector $\mathbf{e}$ are $e_{-1} = 1$ for an RCP pulse ($\xi = +1$) and $e_1 = -1$ for an LCP pulse ($\xi = -1$).

In the second-order PT, the ionization amplitude is defined by [25]:

$$A_{l_i m_i}^{(2)} = - \sum_{q \neq i} \int_{-\infty}^{\infty} \langle \nu \mathbf{p} | \mathbf{d} \cdot \mathbf{F}(t) | q \rangle e^{i\omega_{fq}t} \times \int_{-\infty}^t \langle q | \mathbf{d} \cdot \mathbf{F}(t') | l_i m_i \rangle e^{i\omega_{qi}t'} dt' dt. \quad (12)$$

Here, $|q\rangle$ is an intermediate state with energy E_q ; $\omega_{fq} \equiv E + E_\nu - E_q$; $\omega_{qi} \equiv E_q - E_i$; and $\mathbf{F}(t)$ is the electric field (3). Details of the calculations of the time integrals are provided in [28]. As a result, the ionization amplitude (12) is given by an integral over the energy E_q taken from the matrix element involving the product of two pulse envelope functions (7) with the function $(\mathbf{e} \cdot \mathbf{d})G_{E_q}(\mathbf{e} \cdot \mathbf{d}')$, where G_{E_q} is the stationary Green function of the atom (see [24]). Using the partial expansion of the final state, the angular dependence of the ionization amplitude (12) can be separated out, which leads to the following parameterization:

$$A_{l_i m_i}^{(2)} = e^{-2i\phi} \sum_{l=l_i, l_i \pm 2} \sum_{q=0, 2} \mathcal{A}_{q, l l_i}^{(2)} \{ \{ \mathbf{e} \otimes \mathbf{e} \}_q \otimes Y_l(\hat{\mathbf{p}}) \}_{l_i m_i}, \quad (13)$$

where the dynamic (or radial) amplitude parameters are independent of the electron emission angles and the pulse polarization. Note that the dependence of the dynamic parameters on the pulse envelope cannot be factored out as in the first-order PT case. Thus, the width of the functions $\mathcal{A}_{q, l l_i}^{(2)}(p)$ in momentum space is determined by both the bandwidth of the laser pulse and the dependence of the compound matrix elements on p . Only for $n = 1$ is the width of the functions $\mathcal{A}_{l l_i}^{(1)}(p)$ determined by the Fourier transform F_0^+ of the pulse envelope (see (9)).

At this stage, we should mention that due to the broadband nature of few-cycle pulses, the second-order PT amplitude should also contain the terms corresponding to absorption+emission and two-photon emission. However, these terms are small [24], and they are neglected in the treatment below. For the sake of simplicity, we limit our consideration only to the case of purely CP laser pulses. In this case, we have $\{ \mathbf{e} \otimes \mathbf{e} \}_{00} = -(\mathbf{e} \cdot \mathbf{e})/\sqrt{3} = 0$, and the term with $q = 0$ in (13) vanishes. Accordingly, below, we use the notation $\mathcal{A}_{l l_i}^{(2)} \equiv \mathcal{A}_{2, l l_i}^{(2)}$. Thus, it is easy to deduce that in the n -th PT order, the amplitude of the ionization by a CP pulse can be parameterized as

$$A_{l_i m_i}^{(n)} = e^{-in\phi} \sum_l \mathcal{A}_{l l_i}^{(n)} \{ \{ \mathbf{e} \}_n \otimes Y_l(\hat{\mathbf{p}}) \}_{l_i m_i}, \quad (14)$$

where $\{ \mathbf{e} \}_n$ denotes the tensor product of n vectors $\mathbf{e}$ [27,29], and the summation index l is limited by the triangle rule $|l_i - n| \leq l \leq l_i + n$. The parity conservation law leads to the condition $(-1)^{l+l_i+n} = 1$. This means that the parity of l is the same as that of l_i for even values of n and is the opposite to that of l_i for odd values of n . The total amplitude of the ionization by a few-cycle CP pulse is the sum of all PT orders:

$$A_{l_i m_i} = \sum_{n=1}^{\infty} e^{-in\phi} \sum_l \mathcal{A}_{l l_i}^{(n)} \{ \{ \mathbf{e} \}_n \otimes Y_l(\hat{\mathbf{p}}) \}_{l_i m_i}. \quad (15)$$

This equation can be simplified when an electron is detected in the photon polarization plane (the *in-plane* geometry) in the direction defined by the polar angle φ :

$$A_{l_i m_i}(p, \phi) = \sum_{n=1}^{\infty} \zeta^n e^{i[(m_i + \zeta n)\varphi - n\phi]} \mathcal{B}_{l_i m_i, \zeta}^{(n)}(p), \quad (16)$$

where the parameters $\mathcal{B}_{l_i m_i \zeta}^{(n)}$ are defined by

$$\mathcal{B}_{l_i m_i \zeta}^{(n)}(p) = \sum_l \mathcal{A}_{ll_i}^{(n)} C_{n(-\zeta n)l(m_i+\zeta n)}^{l_i m_i} Y_{l(m_i+\zeta n)}\left(\frac{\pi}{2}, 0\right). \quad (17)$$

Here, the summation runs over all values of $l > 0$, satisfying the conditions

$$(-1)^{l+n+l_i} = 1, \quad (-1)^{l+n+m_i} = 1. \quad (18)$$

Note that in (17), both the Clebsch–Gordan coefficients and the spherical harmonics can be evaluated in closed form [26]. However, we do not present the corresponding expressions here for the sake of brevity. Since the parameters $\mathcal{A}_{ll_i}^{(n)}(p)$ correspond to n -photon absorption, they have pronounced maxima at the values of p corresponding to the photoelectron energy close to $I_p + n\omega$, where I_p is the ionization potential.

Note that the relative magnitude of the parameters $\mathcal{A}_{ll_i}^{(n)}$ and $\mathcal{A}_{ll_i}^{(n\pm 1)}$ is not necessarily determined by the ratio F/F_a , where F is the peak pulse field strength and F_a is the atomic field strength. For example, for $F_a > F$, the second-order PT parameter $\mathcal{A}_{ll_i}^{(2)}$ can be larger than $\mathcal{A}_{ll_i}^{(1)}$ depending on p . Mathematically, this stems from the fact that compound matrix elements can be much larger than the dipole matrix element of the direct transition into the continuum.

From condition (18), it immediately follows that all parameters $\mathcal{B}_{l_i m_i \zeta}^{(n)}$ vanish when $(-1)^{l_i+m_i} = -1$. In other words, photoelectrons cannot be emitted in the photon polarization plane when the parity of m_i is opposite to that of l_i . For example, in the case of an initial P -state, we find that in-plane emission is forbidden for $m_i = 0$. Further, the symmetry properties of the Clebsch–Gordan coefficients and spherical harmonics entering Equation (17) lead to the conclusion that the parameters $\mathcal{B}_{l_i m_i \zeta}^{(n)}$ are invariant under the replacement $\zeta \rightarrow -\zeta$, $m_i \rightarrow -m_i$ up to a phase factor, i.e.,

$$\mathcal{B}_{l_i m_i \zeta}^{(n)} = (-1)^{l_i+n} \mathcal{B}_{l_i -m_i, -\zeta}^{(n)}. \quad (19)$$

3. Ionization Probability and Dichroism Parameters

Within the density matrix formalism, the TDP for the ionization of a polarized atom is defined by

$$\mathcal{W} \equiv \frac{d^3 W}{dp d\Omega_p} = \sum_{m_i, m_i' = -l_i}^{l_i} \mathcal{A}_{l_i m_i} \langle l_i m_i | \rho | l_i m_i' \rangle \mathcal{A}_{l_i m_i'}^*, \quad (20)$$

where ρ is the density operator of the polarized initial atom and Ω_p is the solid angle corresponding to the emission direction of the photoelectron. The angular dependence of the matrix elements of ρ can be separated out by introducing the “polarization momenta” P_{rm} [29,30], which are the irreducible tensor components of the polarization density matrix:

$$\langle l_i m_i | \rho | l_i m_i' \rangle = \sum_{r=0}^{2l_i} (-1)^{l_i-m_i} C_{l_i m_i' l_i -m_i}^{rm} P_{rm}, \quad m = m_i' - m_i. \quad (21)$$

(Often, the polarization is described by polarization state multipoles ρ_{rm} [30], which are connected with the polarization momenta by $\rho_{rm} = P_{rm}^*$).

Inserting (21) into (20) and noting Equation (16) for the ionization amplitude, one can perform summations over m_i, m_i' . However, this comes at the expense of introducing one summation over the momentum index coupling the orbital momenta of photoelectrons from both amplitudes in (20): l from $\mathcal{A}_{l_i m_i}$ and l' from $\mathcal{A}_{l_i m_i'}$. In atomic ionization, the momentum l_i is usually small ($l_i < 4$), so it is more convenient to analyze the expression (20) as it is.

For example, let us consider the ionization of an initial state, purely polarized along the photon beam propagation direction. In this case, the polarization density operator is

$$\rho = |l_i m_i\rangle \langle l_i m_i|, \quad (22)$$

and the non-zero polarization momenta in (21) are

$$P_{r0} = (-1)^{l_i - m_i} C_{l_i m_i l_i - m_i}^r, \quad r = 0, 1, \dots, 2l_i. \quad (23)$$

Accordingly, the TDP (20) becomes

$$\begin{aligned} \mathcal{W}_{l_i m_i}(\xi) &= |A_{l_i m_i}|^2 = \sum_{n, n'=1}^{\infty} \xi^{n+n'} e^{i(n-n')(-\phi+\xi\varphi)} \mathcal{B}_{l_i m_i, \xi}^{(n)} \mathcal{B}_{l_i m_i, \xi}^{(n')*} = \\ &= \sum_{n=1}^{\infty} \left(|\mathcal{B}_{l_i m_i, \xi}^{(n)}|^2 + 2 \operatorname{Re} \sum_{n'=n+1}^{\infty} \xi^{n+n'} e^{i(n-n')(-\phi+\xi\varphi)} \mathcal{B}_{l_i m_i, \xi}^{(n)} \mathcal{B}_{l_i m_i, \xi}^{(n')*} \right). \end{aligned} \quad (24)$$

Note that the major contribution to the magnitude of the expression in brackets comes from the first term. The point is that the overlap of the maxima of the $\mathcal{B}$ -parameters, corresponding to different PT orders (indicated by n), is weak. (Parameters $\mathcal{B}^{(n)}$ and $\mathcal{B}^{(n' \neq n)}$ do not overlap in the limit of monochromatic, i.e., infinitely long, pulses). Therefore, we conclude that the dependence of MD on φ should be weak, too. Consequently, MDs should be seen as a series of slightly distorted concentric circular patterns since the first term in (24) is independent of φ . We also note that the overlap of non-adjacent PT orders, like first and third, etc., is negligible, and only the term with $n' = n + 1$ contributes to the summation over n' in (24). Thus, Equation (24) simplifies to

$$\mathcal{W}_{l_i m_i}(\xi) = \sum_{n=1}^{\infty} |\mathcal{B}_{l_i m_i, \xi}^{(n)}|^2 + 2\xi \operatorname{Re} e^{i(\phi-\xi\varphi)} \sum_{n=1}^{\infty} \mathcal{B}_{l_i m_i, \xi}^{(n)} \mathcal{B}_{l_i m_i, \xi}^{(n+1)*}. \quad (25)$$

Similarly, one can obtain the TDP for the ionization of an initial state with $m_i < 0$, which is purely polarized oppositely to the laser pulse propagation:

$$\mathcal{W}_{l_i - m_i}(\xi) = \sum_{n=1}^{\infty} |\mathcal{B}_{l_i - m_i, \xi}^{(n)}|^2 + 2\xi \operatorname{Re} e^{-i(\phi+\xi\varphi)} \sum_{n=1}^{\infty} \mathcal{B}_{l_i - m_i, \xi}^{(n)} \mathcal{B}_{l_i - m_i, \xi}^{(n+1)*}. \quad (26)$$

Noting the symmetry property (19) of the $\mathcal{B}$ -parameters, we can re-write (26) as follows

$$\mathcal{W}_{l_i - m_i}(-\xi) = \sum_{n=1}^{\infty} |\mathcal{B}_{l_i m_i, \xi}^{(n)}|^2 + 2\xi \operatorname{Re} e^{-i(\phi-\xi\varphi)} \sum_{n=1}^{\infty} \mathcal{B}_{l_i m_i, \xi}^{(n)} \mathcal{B}_{l_i m_i, \xi}^{(n+1)*}. \quad (27)$$

When comparing (27) to (25), we conclude that the MD for the ionization of an atom, polarized along the pulse propagation, can be obtained from the MD for the ionization of an atom polarized oppositely to the pulse propagation by the replacement $\varphi \rightarrow -\varphi$.

3.1. Circular and Magnetic Dichroism Effects in MD

We define the magnitude of the absolute CDMD effect as

$$\Delta_{CD} = \mathcal{W}(\xi = +1) - \mathcal{W}(\xi = -1). \quad (28)$$

From (25), we obtain the absolute dichroism parameter for the ionization of a purely polarized state with the well-defined value of the magnetic quantum number $m = m_i$,

$$\Delta_{CD} = \sum_{n=1}^{\infty} \left(|\mathcal{B}_{l_i m_i, 1}^{(n)}|^2 - |\mathcal{B}_{l_i m_i, -1}^{(n)}|^2 \right) + 2 \operatorname{Re} e^{i\varphi} \sum_{n=1}^{\infty} \left(e^{-i\varphi} \mathcal{B}_{l_i m_i, 1}^{(n)} \mathcal{B}_{l_i m_i, 1}^{(n+1)*} + e^{i\varphi} \mathcal{B}_{l_i m_i, -1}^{(n)} \mathcal{B}_{l_i m_i, -1}^{(n+1)*} \right). \quad (29)$$

Note that $\Delta_{CD} \neq 0$, even for monochromatic pulses, when the second term describing the overlap of amplitudes corresponding to different PT orders vanishes. In this case, the MD will be seen as a series of concentric, perfectly circular patterns. For broadband laser pulses, the non-zero overlap of amplitudes will lead to distortions of the circular symmetry in one azimuthal direction since the second term in (29) is a combination of $\cos \varphi$ and $\sin \varphi$.

Now, let us consider the magnetic dichroism effect in MDs, which is defined as

$$\Delta_{MD} = \mathcal{W}(m_i, \xi) - \mathcal{W}(-m_i, \xi). \quad (30)$$

By considering (25) and (27), we obtain the absolute magnetic dichroism for the ionization by a CP pulse,

$$\Delta_{MD} = \sum_{n=1}^{\infty} \left(|\mathcal{B}_{l_i m_i, \xi}^{(n)}|^2 - |\mathcal{B}_{l_i m_i, -\xi}^{(n)}|^2 \right) + 2\xi \operatorname{Re} e^{i(\phi - \xi\varphi)} \sum_{n=1}^{\infty} \left(\mathcal{B}_{l_i m_i, \xi}^{(n)} \mathcal{B}_{l_i m_i, \xi}^{(n+1)*} + \mathcal{B}_{l_i m_i, -\xi}^{(n)} \mathcal{B}_{l_i m_i, -\xi}^{(n+1)*} \right). \quad (31)$$

As can be seen, the dependence of the MDs on p (i.e., the radial dependence of TDP in the emission plane) for both the CDMD and MDMD parameters, is the same, as it is mostly determined by the first term in (29) and (31). Thus, the corresponding MDs only differ in azimuthal asymmetry, which exhibits $\cos \varphi$ -like dependence in both cases.

3.2. CD Effect for Aligned States

So far, we have analyzed dichroism effects emerging in the in-plane electron emission for an oriented initial atomic state (see Equations (29) and (31)). Such states are characterized by polarization momenta P_{r0} with odd values of the momentum index $r = 1, 3, \dots$. For an aligned state, only polarization momenta P_{r0} with even values of r are non-zero, i.e., $r = 0, 2, \dots$. An example of such a state is given by the following expression for the polarization density operator [30]:

$$\rho = \frac{|l_i m_i\rangle \langle l_i m_i| + |l_i - m_i\rangle \langle l_i - m_i|}{2}. \quad (32)$$

This choice means that the initial atomic states with positive and negative magnetic projections m_i enter the statistical mixture with the same probability, which is equal to $1/2$. The corresponding polarization momenta are

$$P_{r0} = (-1)^{l_i - m_i} C_{l_i m_i, l_i - m_i}^{r0}, \quad r = 0, 2, 4, \dots, 2l_i. \quad (33)$$

Using Equations (25) and (27), we can write the TDP for the ionization of an aligned state defined by the density operator (32):

$$\mathcal{W}_{l_i m_i}^{(a)} = \frac{1}{2} \sum_{n=1}^{\infty} \left(|\mathcal{B}_{l_i m_i, \xi}^{(n)}|^2 + |\mathcal{B}_{l_i m_i, -\xi}^{(n)}|^2 \right) + \xi \operatorname{Re} e^{i(\phi - \xi\varphi)} \sum_{n=1}^{\infty} \left(\mathcal{B}_{l_i m_i, \xi}^{(n)} \mathcal{B}_{l_i m_i, \xi}^{(n+1)*} - \mathcal{B}_{l_i m_i, -\xi}^{(n)} \mathcal{B}_{l_i m_i, -\xi}^{(n+1)*} \right). \quad (34)$$

It can be seen that the first term in brackets is invariant under the exchange $\xi \rightarrow -\xi$, whereas the second term changes its sign. Thus, the first term in (34) does not contribute to the absolute CD (28) for the ionization of an aligned state, and the expression for Δ_{CD} becomes

$$\Delta_{CD}^{(a)} = \sin \varphi \operatorname{Re} e^{i(\phi-\pi/2)} \sum_{n=1}^{\infty} \left(\mathcal{B}_{l_i m_i, 1}^{(n)} \mathcal{B}_{l_i m_i, 1}^{(n+1)*} - \mathcal{B}_{l_i m_i, -1}^{(n)} \mathcal{B}_{l_i m_i, -1}^{(n+1)*} \right). \quad (35)$$

According to this equation, the CDMD effect emerges only due to the interference of the amplitude terms corresponding to different PT orders. Clearly, such an effect is only possible because of the broadband nature of few-cycle electromagnetic pulses. Note also that $\Delta_{CD}^{(a)}$ vanishes after the integration over the emission direction of photoelectrons defined by φ .

4. Numerical Results for the Ionization of the 2P-States of the Hydrogen Atom

To illustrate the theoretical analysis of Section 3, we present the MDs for the ionization of the 2P-states of the hydrogen atom, obtained by the direct numerical solution of the corresponding TDSE using the Qprop (ver. 3.2) software package [31].

We show the results for two values of the carrier frequency: $\omega = 0.08$ and 0.12 a.u. The electron-binding energy in the 2P-state is 0.125 a.u. Thus, for $\omega = 0.08$ a.u., the main ionization channel is two-photon absorption, whereas for $\omega = 0.12$ a.u., one-photon absorption is dominant. In Figure 1, we show the results for a laser pulse with $\omega = 0.08$ a.u. and a duration of $n_c = 2$ cycles of the frequency ω .

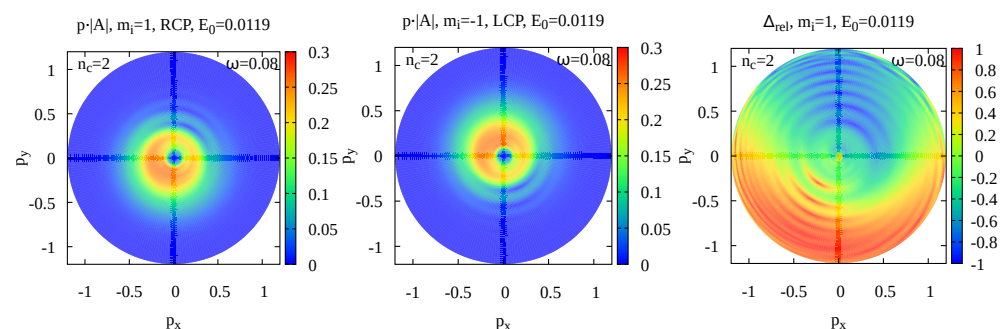


Figure 1. Absolute values of the ionization amplitude multiplied by p , $p|A|$, and the relative CDMD (36) for the ionization of the 2P-state of the hydrogen atom by a single CP pulse with a carrier frequency $\omega = 0.08$ a.u., peak electric field $E_0 = 0.0119$ a.u. (peak intensity $I_0 = 5 \times 10^{13}$ W/cm²), and duration of $n_c = 2$ cycles of the frequency ω .

Note that we show the color maps of the parameter $p|A|$ rather than the TDP, $\mathcal{W}$, for better visualization of the angular dependence details. The left panel corresponds to the ionization of an initial state with $m_i = 1$ by an RCP pulse. In the central panel, the pulse is an LCP, and the initial state has $m_i = -1$. In accordance with the above analysis (see Equations (25) and (27)), the two MDs are mirror images of each other with respect to the horizontal line. In other words, the image in the center is the same as the one on the left after the replacement $\varphi \rightarrow -\varphi$ and vice versa. The MDs in Figure 1 form a series of circular patterns distorted by cos-type angular factors, which enter Equation (25) via the complex exponent $\exp(-i\varphi)$. The right panel in Figure 1 shows the magnitude of the relative CD parameter, defined by

$$\Delta_{rel} = \frac{\mathcal{W}(\xi = +1) - \mathcal{W}(\xi = -1)}{\mathcal{W}(\xi = +1) + \mathcal{W}(\xi = -1)}, \quad -1 \leq \Delta_{rel} \leq 1. \quad (36)$$

It can be seen that the MD of Δ_{rel} exhibits circular patterns (corresponding to the first term in (29)), superimposed by azimuthal distortion with one maximum. Again, this is due

to the terms $\exp(\pm i\varphi)$ entering Equation (29). We note that the CDMD is large in regions where the TDP is small. In regions where the TDP is maximal, the magnitude of the relative CDMD is $\sim 20\%$, which is rather large compared to the CD in ionization by monochromatic (i.e., long) pulses [16].

Figure 2 shows the dependence of the parameter $p|A|$ and the relative CDMD on the pulse duration. Specifically, three pulse durations are considered: $n_c = 2$ (upper row), 3 (middle row), and 4 (lower row) cycles of the carrier frequency $\omega = 0.12$ a.u., with a peak pulse intensity of 10^{14} W/cm². One can observe that the azimuthal distortions of the MDs decrease as the pulse duration increases. This can be explained by Equation (25), where it can be seen that the dependence of the TDP on φ is determined by the overlap of the β -parameters corresponding to adjacent PT orders. This overlap vanishes for monochromatic pulses so that the color maps of the MDs in this case will comprise a series of infinitely thin circles with zero TDP in between. Interestingly, although the MDs for $n_c = 4$ appear as a series of regular circularly symmetric patterns, the CD effect is still substantial in this case. This is emphasized by the azimuthal color asymmetry seen in the panel for Δ_{rel} . Note also that the above results were given for a zero CEP, $\phi = 0$. The dependence of the TDP on ϕ mostly leads to an overall rotation of the azimuthal asymmetries in the color maps of the MDs.

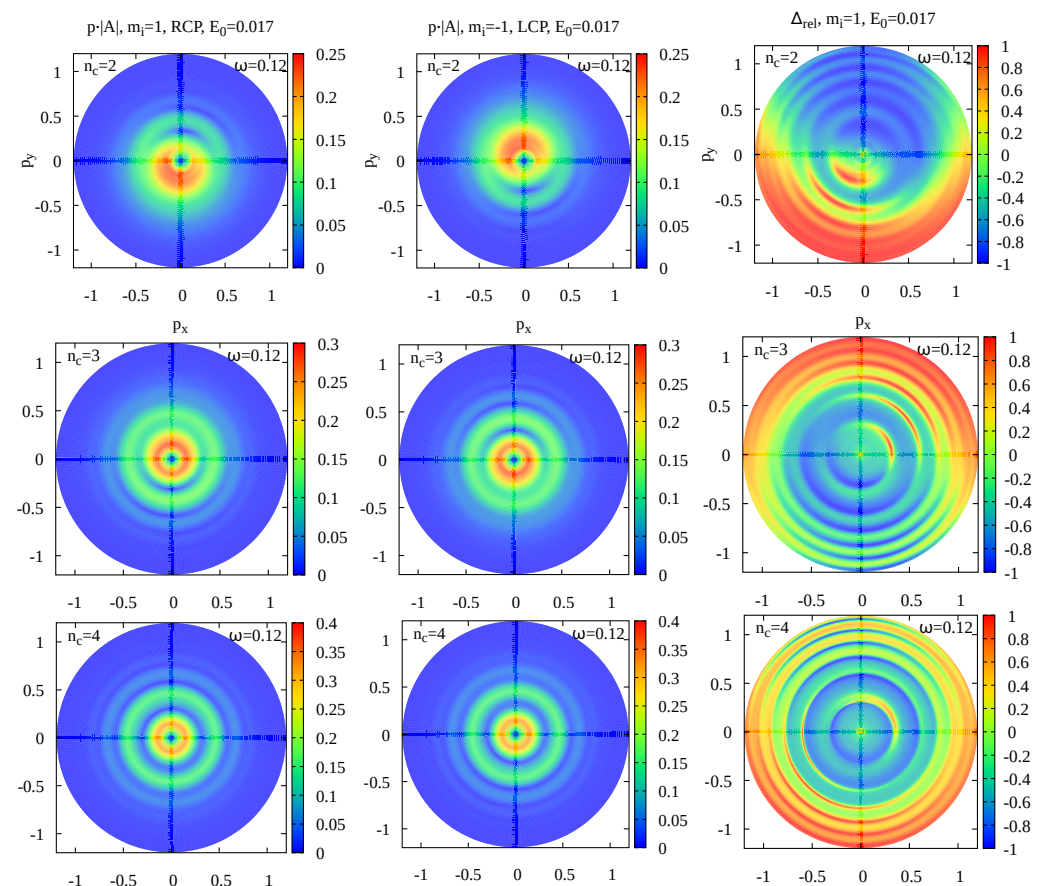


Figure 2. Absolute values of the ionization amplitude multiplied by p , $p|A|$, and the relative CDMD (36) for the carrier frequency $\omega = 0.12$ a.u., peak electric field $E_0 = 0.0169$ (peak intensity $I_0 = 10^{14}$ W/cm²), and three pulse durations: two, three, and four cycles of the carrier frequency, as indicated by n_c . Left column: $p|A|$ for an RCP pulse with $\phi = 0$; middle column: LCP pulse with $\phi = \pi$; right column: relative CDMD defined by (36).

Finally, in Figure 3, we present the results for the ionization of an aligned (see Equation (32)) initial 2P-state. As discussed in Section 3.2, we can observe that the azimuthal dependence of the TDP is much more pronounced than the radial one. Additionally, it can be seen that the relative CDMD vanishes along the line corresponding to

$\varphi = 0, \pi$, and changes sign under the replacement $\varphi \rightarrow -\varphi$, which is the signature of the factor $\sin \varphi$ entering Equation (35) for $\Delta_{rel}^{(a)}$.

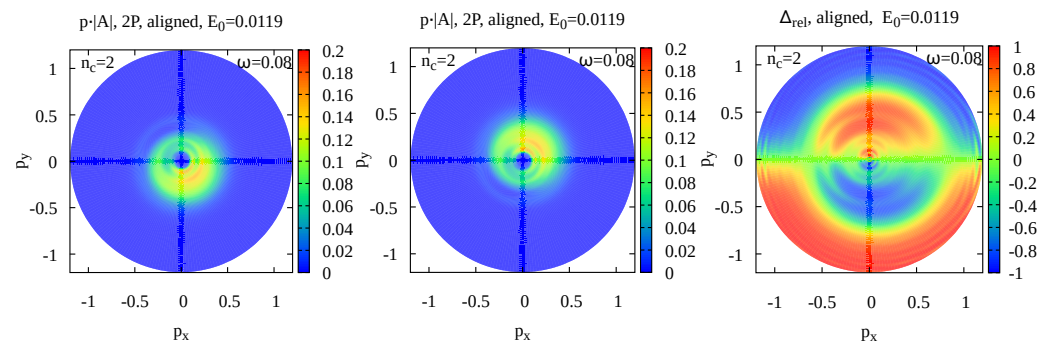


Figure 3. Absolute values of the ionization amplitude multiplied by p , $p|A|$, and the relative CDMD (36) for the ionization of the aligned 2P-state of the hydrogen atom (see (32)) by a single RCP pulse with a carrier frequency $\omega = 0.08$ a.u., peak electric field $E_0 = 0.0119$ a.u. (peak intensity $I_0 = 5 \times 10^{13}$ W/cm²), and duration of $n_c = 2$ cycles of the frequency ω .

5. Discussion

Above, we have presented the results of numerical TDSE calculations for the hydrogen atom only. However, our treatment was not specific to this atom. Rather, it is valid for an arbitrary many-electron atom beyond the single active electron approximation. Indeed, our consideration is based on the expansion (15) for the ionization amplitude. This expansion, however, is perfectly general, as the spherical harmonics $Y_{lm}(\hat{\mathbf{p}})$ form a complete basis set in the momentum space. The same is true for the set of tensor products $\{\mathbf{e}\}_{n\mu}$, which form a complete basis in the space of functions depending on the polarization vectors $\mathbf{e}$. The perturbation theory was only used to provide the physical interpretation for the terms of the expansion (15). In particular, within the PT, the summation in (15) should include only the terms satisfying the first condition from Equation (18). In addition, for the in-plane emission of electrons, the second condition in (18) should also be fulfilled. Next, PT was used to simplify the expression (24) for the ionization probability, where it was assumed that the non-adjacent terms of the ionization amplitude, for which the difference in the number of absorbed photons is larger than unity, do not overlap. The validity of all the above assumptions was confirmed by the TDSE results for the hydrogen atom. We expect this to be true for other atoms (or ions), provided the peak pulse electric field strength is smaller than the atomic field. Note that high-intensity few-cycle laser pulses are currently available in both the near IR (up to 10^{18} W/cm², $\omega = 1.5$ eV [32]) and XUV regions (up to 10^{14} W/cm² for $\omega = 36$ eV were reported [19,33]). Currently, experiments are being performed with alkali atoms like lithium [6] or potassium [34]. The electron-binding energy in these atoms is much smaller than that in the hydrogen atom. Therefore, an intensity range of $10^{12} \div 10^{13}$ W/cm² for few-cycle CP pulses in the optical region would suffice for the observation of dichroism effects.

Of course, PT formalism can also be used to obtain numerical values of the dynamic amplitude parameters $A_{li}^{(n)}$. The corresponding calculations, however, are rather complicated, even for the ionization of the hydrogen atom ground state when only first- and second-order PT terms are retained in the ionization amplitude [24]. The main complication is in the calculation of the integrals over the energy, taken from the compound matrix elements involving the Coulomb Green function. Numerical calculations of the parameters $A_{li}^{(n)}$ within the PT become practically inaccessible for higher PT orders ($n > 2$). Again, we emphasize that our conclusions are not related to the problem of the calculation of dynamic amplitude parameters since they are based on general assumptions about the properties of the coefficients in the tensor expansion (15).

Although we have analyzed the CDMD effects, assuming the initial bound state to be purely polarized, our results can easily be generalized to the case of partially polarized initial states based on Equation (20) and the parameterization (16) of the ionization amplitude. For partially polarized initial states, the elements of the density matrix (21) are determined by the set of polarization momenta P_{rm} , whose values should be provided either from the experiment or from calculations describing the atomic polarization process. Then, the probability of the corresponding ionization process will be determined by Equation (20) upon the substitution of the matrix elements (21).

6. Conclusions

We have analyzed the MDs of electrons emitted by polarized atoms subjected to a few-cycle CP electromagnetic pulse. Due to the large number of angular parameters inherent in the problem, we concentrated on the case of the emission of electrons in the pulse polarization plane (the “in-plane” emission geometry). We expect that other experimental geometries will not lead to the emergence of dichroism effects in the MDs other than those analyzed above. It is important to note that the results of our PT analysis are not specific to the hydrogen atom. Rather, they are applicable to the process of short CP pulse ionization of any atomic or molecular target. In Section 2, we established a selection rule, stating that the in-plane emission is forbidden in all PT orders when the initial orbital momentum quantum number and its projection have different parities. This selection rule is perfectly general and should hold for any kind of initial target.

Among the dichroism effects considered in this article, the most physically interesting is the CDMD effect in the ionization of aligned atomic states. Such an effect is absent in long pulse ionization since it is forbidden by symmetry arguments. Indeed, an aligned state has no right–left asymmetry, which is essential for the CD effect to occur. It is the short pulse itself that carries the asymmetry, and it is described by the parameter ϕ (i.e., the CEP). It is known that the CEP causes nonlinear CDMD effects in the short pulse ionization of spherically symmetric atomic S-states [24,35]. Here, we have demonstrated that such nonlinear CEP-caused CDMD effects take place in the general case of the ionization of initial bound states with non-zero orbital momentum. Such effects should be taken into account in investigations of the interaction of few-cycle XUV pulses with atomic inner shells, which have recently been reported [36,37].

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Abbreviations

The following abbreviations are used in this manuscript:

CEP	Carrier-envelope phase
CD	Circular dichroism
CP	Circular polarization
CDMD	Circular dichroism in momentum distribution
LCP	Left-hand circular polarization
MD	Momentum distribution

MDMD	Magnetic dichroism in momentum distribution
PT	Perturbation theory
RCP	Right-hand circular polarization
TDP	Triply differential probability
TDSE	Time-dependent Schrödinger equation

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