Above-threshold ionization in a bicircular field: quantum orbits unfolding in a plane

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Abstract. Above-threshold ionization (ATI) of atoms by a strong bicircular laser field is investigated using the strong-field approximation and the quantum-orbit theory. The bicircular field consists of two coplanar counterrotating circularly polarized fields with a frequency ratio of 2:1. The velocity map of the angle-resolved ATI spectra, both for direct and rescattered electrons, reflects the shape of a parametric plot of the bicircular field and its symmetries. It is shown that the main characteristics of the ATI spectra can be explained using only a few quantum orbits having short travel times. We also analyze a recently discovered [Phys. Rev. A 93, 052402(R) (2016)] bicircular-field-induced spin asymmetry of the ATI electrons and show that the momentum dependence of the spin-asymmetry parameter is stronger for longer wavelengths.

1. Introduction
The interaction of intense bicircular laser fields with atoms has received a great deal of recent attention [1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43]. A bicircular field consists of the superposition of two circularly polarized fields that rotate in the same plane, usually in opposite directions (counterrotating), with different frequencies. The case of a frequency ratio of 2:1 is the standard situation, but other frequency ratios and incommensurate frequencies are also considered. Counterrotating bicircular fields are known to generate very intense circularly polarized high-order harmonics that obey certain characteristic selection rules. The velocity map of angle-resolved above-threshold ionization (ATI) induced by such fields reflects the shape of the parametric plot of the bicircular field and its symmetries [33]. The strong-field approximation (with or without rescattering taken into account) well accounts for the experimental observations [32].

If the pertinent temporal integrations are carried out by the saddle-point method, quantum orbits result. They allow for a space-time visualization of the electronic trajectories subsequent to tunneling and before and after the rescattering event. Unlike the standard case of a linearly polarized driving field, the quantum orbits fill the entire polarization plane, with the electrons recolliding with their parent ions at arbitrary angles. It is important to discriminate between
backward and forward scattering orbits. The former shape the spectrum for comparably high energies, while the latter cause involved low-energy structures. Identifying the contributions of specific quantum orbits to the velocity map, it is possible to attribute certain features to certain quantum orbits. If the ionic dynamics are such that fine-structure splitting is important, then the rescattered electrons exhibit significant spin asymmetry. Since rescattered electrons are emitted on the attosecond time scale, the possibility opens up for the electron spin to enter attoscience as an observable [34].

Our theory, which is based on the S-matrix formalism introduced in [33] and [34], is briefly presented in Sec. 2. The numerical results and discussions are presented in Sec. 3 and, finally, the conclusions are summarized in Sec. 4. The atomic system of units (\( \hbar = e = m_e = 4\pi\varepsilon_0 = 1 \)) is used throughout the paper.

2. Theory

Using the S-matrix theory and the strong-field approximation (SFA) in [33] we derived the following expression for the averaged differential ionization rate,

\[
\frac{\mathcal{M}_p}(n) = \frac{2\pi p N_e}{2l + 1} \sum_{m=-l}^{l} |T_{plm}(n)|^2, \tag{1}
\]

where \( N_e \) is the number of equivalent electrons in the ionizing shell and \( T_{plm}(n) \) is the T-matrix element, which can be written as

\[
T_{plm}(n) = \int_{0}^{T} \frac{dt}{T} e^{i[p \cdot \alpha(t) + \mathcal{U}_1(t) + n \omega t]} \left[ (p + A(t) | r \cdot E(t) | \psi_{lm}) + T_{plm}^R(t) \right]. \tag{2}
\]

Here \( T = 2\pi/\omega \) is the optical period and \( n \) is the number of photons exchanged with the laser field, which is related to the energy-conserving condition \( n \omega = E_p + I_p + U_p \), where \( E_p = p^2/2 \) is the final electron kinetic energy in the continuum state \( |p\rangle \), \( \mathcal{U} \) is the electron’s drift momentum, which equals the momentum outside the field and \( \langle p | p \rangle = (2\pi)^{-3/2} \exp(ip \cdot r) \), \( I_p \) is the ionization potential of the atom in the bound state \( |\psi_{lm}\rangle \), \( \alpha(t) = \int t A(t') dt' \), \( A(t) = -\int t' E(t') dt' \) \( [E(t) \) is the electric field vector], and \( \mathcal{U}_1(t) = \int t A^2(t') dt'/2 - U_p t \) with \( U_p = \int_0^T A^2(t') dt/(2T) \) the ponderomotive energy. The first term of the integrand on the right-hand side of Eq. (2) corresponds to direct ionization, while the rescattering part of the transition amplitude is given by

\[
T_{plm}^R(t) = -i \int_{0}^{\tau_{max}} d\tau \left( \frac{2\pi}{1^{3/2}} \right)^{1/2} \langle p | V | k_s \rangle \langle k_s + A(t-\tau) | r \cdot E(t-\tau) | \psi_{lm} \rangle e^{i[(S_{k_s} - S_{k_s} - l^2)/\tau]}, \tag{3}
\]

where \( \tau_{max} \to \infty \), \( k_s = -\int_{-\tau}^{t} dt' A(t')/\tau \) is the intermediate electron stationary momentum and \( S_{k_s}(t) = \int t' dt' [k + A(t')]^2/2 \). In Sec. 3 we will analyze the influence of finite values of the parameter \( \tau_{max} \). The ground-state atomic wave function is modeled by a linear combination of Slater-type orbitals obtained by using the Hartree-Fock-Roothaan method [44]. For atoms having \( p \) ground state \( (l = 1) \) the magnetic quantum number is \( m = 0, \pm 1 \).

Within the quantum-orbit theory, the integrals in Eq. (3) are approximated by the sum over the saddle-point solutions of the equations

\[
\frac{1}{2} [k_s + A(t_0)]^2 = -I_p, \quad \frac{1}{2} [k_s + A(t)]^2 = \frac{1}{2} |p + A(t)|^2, \tag{4}
\]

which express energy conservation at the ionization time \( t_0 = t - \tau \) and the rescattering time \( t \). Not going into details, we only mention that in [33] these solutions were classified by
the multiindex \((\alpha, \beta, m)\) for backward-scattering solutions and \((\nu, \rho, \mu)\) for forward-scattering solutions. More details about quantum orbits can be found in references [45, 46, 47, 48, 49, 50, 51, 52].

We use a bichromatic circularly polarized field with the coplanar counterrotating components having the angular frequencies \(\omega\) and \(2\omega\). The corresponding electric-field vector in the \(xy\) plane, determined by the unit vectors \(\mathbf{e}_x\) and \(\mathbf{e}_y\), is defined by

\[
\mathbf{E}(t) = \frac{i}{2\sqrt{2}} \left[ E_1(\mathbf{e}_x + i\mathbf{e}_y)e^{-i\omega t} + E_2(\mathbf{e}_x - i\mathbf{e}_y)e^{-i2\omega t} \right] + \text{c.c.},
\]

where \(\text{c.c.}\) denotes the complex conjugate of the first term on the right-hand side and \(I_1 = E_1^2\) and \(I_1 = E_2^2\) are the component intensities. The bichromatic field \(\mathbf{E}(t)\) and its vector potential \(\mathbf{A}(t)\) obey the following dynamical symmetry: a rotation by the angle \(\alpha_j = -2\pi j/3\) about the \(z\) axis is equivalent to a translation in time by \(\tau_j = jT/3\), i.e. \(R_z(\alpha_j)\mathbf{E}(t) = \mathbf{E}(t + \tau_j)\) where \(j\) is an integer. The diagonal matrix elements of the rotation matrix \(R_z(\alpha_j)\) are \(\cos \alpha_j\), while the off diagonal elements are \(\pm \sin \alpha_j\).

In ATI of xenon atoms there are two continua corresponding to the two ground states of the \(\text{Xe}^+\) ion: \(2P_{3/2}\) \((I_{p3/2} = 12.14\) eV\) and \(2P_{1/2}\) \((I_{p1/2} = 13.44\) eV\). Because of this fine-structure splitting of 1.31 eV, it is possible to observe different momentum distributions of electrons having different spin in the case where the ATI rate shows a strong \(m = \pm 1\) asymmetry. This is the case for ATI by a circularly polarized field. This was predicted in [53] and recently observed [54]. For ATI by a bichromatic field this effect was considered in [34], where the normalized spin-asymmetry parameter was defined with respect to the maximal summed rate by the relation

\[
\tilde{\mathbf{A}}_p = (W_{p\uparrow} - W_{p\downarrow}) / \max_p(W_{p\uparrow} + W_{p\downarrow}).
\]

Here \(W_{p\uparrow}\) \((W_{p\downarrow})\) denotes the rate for electrons having spin up (down). The method of calculation of these rates is presented in [34].

3. Numerical results

In Figs. 1 and 2 we present numerical results for ATI of Ne atoms by a bichromatic \(\omega–2\omega\) laser field having the fundamental wavelength of 800 nm and equal intensity of both components, \(I_1 = I_2 = 2 \times 10^{14}\) W/cm\(^2\). The \(T\)-matrix element (2) is evaluated on a grid of points that satisfy the energy-conservation condition \(\hbar \omega = E_p + I_p + U_p\) and the resulting discrete distribution is smoothed. This is why no ATI rings are visible in the subsequent figures. In Fig. 1 the logarithm of the differential ionization rate of Ne atoms is presented as a function of the electron momentum. In the upper left panel we show the spectra of the direct electrons [electrons that reach the detector without interacting with the parent ion after ionization, as described by the first term in Eq. (2)]. Inspecting the lower left panel of Fig. 2 we conclude that the direct electrons are emitted predominantly in the direction opposite to the vector potential \(\mathbf{A}(t)\) at the time of ionization (i.e. in the directions determined by the emission angles \(\theta = 60^\circ, 180^\circ\) and \(300^\circ\)). The remaining panels of Fig. 1 present the differential ionization rate obtained taking into account only the rescattered electrons. The results shown in the left panels are obtained by two-dimensional numerical integration [scattering term in Eqs. (2) and (3)], using four \(2p\) Slater-type orbitals to represent the ground state of the Ne atom, for different values of the parameter \(\tau_{\text{max}}\): 0.66\(T\) (bottom panel), 1\(T\) (second panel from the bottom) and 5\(T\) (third panel from the bottom). For \(\tau_{\text{max}} > 5T\) the results do not change, which means that the contribution of electrons having travel times larger than five optical cycles can be neglected. It is interesting that the results obtained with \(\tau_{\text{max}} = 2T/3\) qualitatively reproduce the exact ATI spectra. This means that the short quantum orbits give the main contribution to the ATI rate.
Figure 1. (Color online) The logarithm of the differential ionization rate (in au) of Ne atoms presented in false colors in the electron-momentum plane for ionization by a bicircular $\omega-2\omega$ laser field with equal intensity of both components, $I_1 = I_2 = 2 \times 10^{14}$ W/cm$^2$ and the fundamental wavelength of 800 nm. The false-color scale covers four orders of magnitude. Left panels: results obtained by numerical integration. The subpanels from top to bottom correspond to the direct SFA (top) and rescattering improved SFA with the upper limit of integration over the travel time equal to 5, 1 and 0.66 optical cycles. Right panels: results obtained using the quantum-orbit formalism with the following quantum-orbit solutions (from top to bottom): $(\nu, \rho, \mu) = (1, 0, 0)$, $(\nu, \rho, \mu) = (\pm 1, 2, 0)$, $(\alpha, \beta, m) = (\pm 1, 1, 0)$ and the coherent sum of all these five contributions.
Figure 2. (Color online) Left: The $\omega$–$2\omega$ bicircular electric-field vector (upper left panel), vector potential (lower left), electron trajectory (upper right), and velocity (lower right) between the ionization (I) and rescattering (R) times that correspond to the orbit $(\alpha, \beta, m) = (1, 1, 0)$ and the energy $8U_p$. Right: Electron trajectories for three short forward-scattering-like quantum orbits and the energies as denoted in the legend. Laser and atomic parameters are as in the previous figure and the electron is emitted in the direction $\theta = 50^\circ$.

For a closer analysis, in the right-hand panels of Fig. 1 we present the results obtained using only some particular short quantum orbits. The top right panel displays the results obtained using only the shortest forward-scattering quantum orbit, which is denoted by the multiindex $(\nu, \rho, \mu) = (1, 0, 0)$ (notation of [33] is used). The second panel shows the contribution of the next forward-scattering quantum orbits $(\nu, \rho, \mu) = (\pm 1, 2, 0)$. These orbits come in pairs $(\nu = \pm 1)$, which leads to interference structures. The third panel corresponds to the pair of the shortest backward-scattering quantum orbits $(\alpha, \beta, m) = (\pm 1, 1, 0)$. These orbits dominate the high-energy part of the spectrum. Interference of the two solutions of the pair is again clearly visible. The coherent sum of the contributions of all five quantum orbits mentioned yields the spectrum shown in the bottom right panel. By comparison with the panels shown on the left-hand side we see that these five shortest quantum orbits qualitatively reproduce the exact spectrum.

In order to better understand the physics of the bicircular-field-induced ATI process, in Fig. 2 we analyze the relevant quantum orbits in more detail. In the left-hand panel we show for $0 \leq t \leq T$ the parametric plot of the electric field vector $(E_x(t), E_y(t))$ (upper left subpanel) and the corresponding vector potential $(A_x(t), A_y(t))$ (lower left subpanel). The field and the vector potential at the ionization time $t_0$ and the rescattering time $t$, which correspond to the shortest backward-scattering quantum orbit $(\alpha, \beta, m) = (1, 1, 0)$ are denoted by the filled circles and the letters I and R, respectively. We see that the field between ionization and rescattering approximately follows a line oriented by the angle $120^\circ$ with respect to the positive $x$ axis. As a consequence, the corresponding electron trajectory between Re$t_0$ and Re$t$, which is obtained projecting the quantum orbit onto the real $xy$ plane for real times, is almost parallel to the $y$ axis (see the upper right subpanel of the left panel in Fig. 2). The electron is “born at the tunnel exit”, a few atomic units away from the origin, moves in the direction of the positive $y$ axis and, when the $x$ component of the field changes its sign, turns around and moves back to the parent ion, approximately along the negative $y$-axis direction. Thereafter, the electron rescatters at
the origin and moves towards the detector, which is situated at the angle $\theta = 50^\circ$ with respect to the $x$ axis. In the lower right subpanel of the left panel of Fig. 2 we display the electron velocity. This velocity at the ionization time is different from zero, which allows the electron to return to the ion at the rescattering time $t$. The force in the negative $x$ direction [$-E_x(t)$; the field is shown in the upper left subpanel] is largely compensated by the initial momentum in this direction. After ionization the electron is first accelerated to positive momenta, then decelerated and bent around such that it recollides with substantial momentum. The vector potential is maximum at this return time $t$.

In the right-hand panel of Fig. 2 we show the electron trajectory for the three short forward-scattering-like quantum orbits $(\nu, \rho, \mu) = (1, 0, 0)$ (maroon line) and $(\nu, \rho, \mu) = (\pm 1, 2, 0)$ (red lines). For the maroon line the electron moves directly from the tunnel exit to the nucleus where it forward scatters and leaves in the direction determined by the angle $\theta = 50^\circ$ having the final energy $E_p = 1.6U_p$. Since the trajectory is short the probability of this process is higher. Between ionization and recombination the electron is close to the nucleus so that this probability is further enhanced by the influence of the Coulomb potential. The contribution of this solution is responsible for the experimentally observed blobs in the electron-momentum distribution [26]. The trajectories for the solutions $(\nu, \rho, \mu) = (\pm 1, 2, 0)$ (red solid line and red dot-dashed line) correspond to the energy $E_p = 0.8U_p$. The electron forward scatters under the angle $60^\circ$, then moves in the direction of the positive $y$ axis and then, finally, in the direction towards the detector.

Let us now present results for direct ATI of Xe atoms. We use the bicircular $\omega^{-2}\omega$ laser field with $I_1 = I_2 = 1 \times 10^{14}$ W/cm$^2$ and the fundamental wavelength of 1300 nm. Similarly as in [34], we exhibit in Fig. 3 the partial differential-ionization rate for initial states having the magnetic quantum number $m = -1$ (upper left panel), $m = +1$ (upper right panel) and the rate summed over $m = \pm 1$ (lower right panel; the contribution of the $m = 0$ term is zero [34]). It is obvious that we have a large asymmetry between the $m = -1$ and $m = +1$ case. As a consequence, the normalized spin-asymmetry parameter $A_p$, shown in the lower right panel, takes on large values. The results presented exhibit symmetry with respect to rotation by the angle $120^\circ$ about the $z$ axis and reflection symmetry with respect to axes at the angles $60^\circ$, $180^\circ$, and $300^\circ$ with respect to the $x$ axis. These symmetries are explained in [33]. Near the reflection axes the rate exhibits maxima, which are different for positions of $m = \pm 1$. The spin-asymmetry parameter and, therefore, the spin of the emitted electron, considerably changes with the electron energy for a fixed emission angle (look, for example, at $\theta = 60^\circ$). This effect was explained in detail in [34]. In the present paper we repeated the calculations of [34] for the fundamental wavelength of 1300 nm (800 nm was used in [34]). As a consequence of the longer wavelength, the oscillations of the rate are much larger and the observed structures change more rapidly than in the 800 nm case.

4. Conclusions
The theory of ATI by a bicircular field was presented in [33], while the corresponding spin-asymmetry effect was reported in [34]. In the present paper we confirmed the results of these references and analyzed some aspects of the bicircular-field-induced ATI in more detail. In particular, we investigated numerically the influence of the electron travel time on the rescattering process and found that the main contributions to the electron spectra come from short travel times (shorter than $2/3$ of the optical cycle). These numerical results agree with the results obtained using the quantum-orbit theory. We have analyzed how the relevant quantum orbits unfold in the bicircular-laser-field polarization plane.

Another important result of the present paper concerns the spin asymmetry of ATI by a bicircular field with a longer fundamental wavelength than considered in [34]. We found that the oscillations of the rate with the electron energy are much stronger and more rapid for the
Figure 3. (Color online) The differential ionization rate (in au) of Xe atoms, presented in false colors in the electron-momentum plane for ionization by a bicircular $\omega-2\omega$ laser field with equal intensity of both counterrotating components, $I_1 = I_2 = 1 \times 10^{14}$ W/cm$^2$, and the fundamental wavelength of 1300 nm. Upper panels: the rates for the initial states having the magnetic quantum number $m = -1$ (left) and $m = +1$ (right). Lower left panel: the rate summed over $m$. Lower right panel: normalized asymmetry parameter $A_p$.

longer wavelength of 1300 nm than for 800 nm used in [34]. This is an important result since spin-polarized electrons have many practical applications (for example, to study magnetic materials [55] and chiral molecules [56, 57]).

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