

## Strong-field ionization of homonuclear diatomic molecules by a bicircular laser field: Rotational and reflection symmetries

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We investigate above-threshold ionization (ATI) of homonuclear diatomic molecules by the so-called bicircular field using the improved molecular strong-field approximation. Bicircular field is a two-color laser field having coplanar circularly polarized counter-rotating components of frequencies  $r\omega$  and  $s\omega$ , with  $r$  and  $s$  integers. Our analysis includes the high-energy part of the corresponding spectra, i.e., high-order ATI (HATI). The obtained molecular (H)ATI spectra are more complicated than the corresponding atomic spectra. We have identified four symmetries which are satisfied in (H)ATI of homonuclear diatomic molecules. Two of these symmetries are general rotational symmetries valid both for direct and rescattered HATI electrons. The remaining two symmetries are reflection symmetries valid only for the direct ATI electrons. Analytical proof of these symmetries is also given. These symmetries are illustrated using numerical examples of HATI spectra of the  $N_2$  molecule for various molecular orientations.

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### I. INTRODUCTION

When atoms or molecules are subjected to an intense laser field various nonlinear phenomena can occur (see the review articles [1–7] and reference therein). Generally, these phenomena can be divided into two groups: processes that are possible only in the presence of a laser field and those which can take place without the laser field. In the present paper, we are interested in the first group. Examples of these nonlinear processes are high-order above-threshold ionization (HATI), high-order harmonic generation (HHG), and nonsequential double ionization (NSDI). During its interaction with a strong laser field, a quantum-mechanical system can absorb more photons from the field than is necessary for ionization of the considered target. After ionization, the freed electron may directly go to the detector without any interaction with its parent ion. This process is called above-threshold ionization (ATI) [8] and represents the first step of the previously mentioned high-order processes.

All these laser-induced processes can be described by the three-step model [9]. The first step of ionization was already explained. The second step is determined with the dynamics of the liberated electron in the strong laser field. Under the influence of the applied laser field, the electron can be driven back in the vicinity of its parent ion. Now, three different scenarios connected with the three mentioned phenomena can be seen. In the case of HATI, the returning electron elastically rescatters off the parent ion, moves away from it, and reaches the detector with a higher kinetic energy than in the ATI process [1]. For the HHG process, in the third step, the electron recombines with the parent ion and one high-energy photon is emitted [10]. The energy spectra of the HATI and HHG processes are characterized by a plateau which manifests as a broad energy interval of the spectrum in which the photoelectron (HATI) or photon (HHG) yield is practically constant. These intervals are followed by abrupt cutoffs. Finally, during the laser-driven recollision of the first

electron with the parent ion one more electron from the target may be ejected, which represents the third step for the NSDI process [6].

Nonlinear laser-induced molecular processes have attracted much attention in the last few years. Analyzing the corresponding spectra of the mentioned processes we are able to extract important information about molecular structure. This is possible for a linearly as well for an elliptically polarized driving field. In this paper we are interested in the molecular HATI process governed by a bicircular laser field.

More recently, the strong-laser-field scientific community has focused attention on the behavior of atoms and molecules in the so-called bicircular laser field [11–14]. A bicircular field consists of two coplanar corotating or counter-rotating circularly polarized fields of frequency  $r\omega$  and  $s\omega$  ( $r$  and  $s$  are integers). HHG by such a field has been considered as early as 1995 [15–17] and was investigated theoretically in subsequent years [18–25]. The approach used in Refs. [20–24] is based on the  $S$ -matrix theory. This theory of HHG has recently been generalized to multielectron atoms [26]. Selection rules for HHG by a bicircular field are also analyzed in detail in Refs. [27,28]. A distinction is made between the selection rules for atoms with closed and nonclosed shells. It is also shown that it is possible to introduce spin into attoscience with spin-polarized electrons produced by a bicircular laser field [29]. More recent papers [30–36] are devoted to the bicircular-field-induced HHG process on molecules. Symmetries of the HHG spectra were investigated for different molecular species and different forms of the applied bicircular laser field. HHG by a bicircular field is explored for a polyatomic molecule  $BF_3$  and it was shown that the observed asymmetry in the emission of high harmonics with opposite helicities strongly depends on the molecular orientation [34]. In Ref. [36] a general theory of HHG generated by a bicircular laser field on  $N$ -fold rotationally symmetric molecules was presented. NSDI by a bicircular field was investigated in Refs. [37–40].

Experimental results for the strong-field ionization by bicircular field were presented in Ref. [41]. Bicircular HATI from atomic targets was recently analyzed in detail in Refs. [42,43]. In Ref. [44] experimental data and numerical results based on the improved strong-field approximation (SFA) were compared and the agreement was good. It was found that the electron-ion rescattering is optimized (in the sense of the highest and longest plateau) when the ratio of the second harmonic field intensity and the fundamental driving field intensity is close to four. It was also shown that the electron-ion rescattering is absent in the case of the bicircular field with corotating components, but appears in the case of counter-rotating components of the driving field [44,45]. Also, in earlier papers [46] some features of the direct above-threshold detachment (ATD) spectra of atomic negative ions generated by a bicircular laser field were presented. The ATD process is analog of the ATI on atomic targets and considers detachment process from negative ions. High-order ATD, which takes into account the rescattering of the detached electron on the parent atom, was considered in Ref. [45]. All mentioned papers are devoted to laser-induced processes. In Ref. [47] the process of electron-ion radiative recombination assisted by bicircular field was considered. This process is called laser-assisted recombination (LAR). A similar but more complicated process which includes LAR with additional rescattering scenarios as well as the summation over all possible values of the magnetic quantum number  $m$ , which characterizes atoms with the  $p$  ground state, is considered in Ref. [48].

To our knowledge, there are no publications that extend atomic HATI by bicircular field to bicircular HATI from molecular targets. As molecules are multicenter systems and more parameters may influence electron-ion rescattering process, we should extend our improved molecular strong-field approximation (MSFA) theory in order to describe the molecular HATI process governed by a bicircular field. It is important to stress again that phenomena such as molecular HHG and molecular HATI can be a useful tool to gain insight into molecular structure and dynamics on the subfemtosecond time scale [49,50].

This paper is organized as follows. In Sec. II we define our counter-rotating bicircular field and the geometry of molecular HATI process and present a brief review of our extended MSFA theory. This is followed by consideration of general symmetries of molecules and applied bicircular laser field in Sec. III. Section IV summarize our results. We use atomic system of units.

## II. THEORY

Our MSFA for homonuclear diatomic molecules was presented in detail in Refs. [51–54]. It was used for simulation of experiments of different groups [55–61]. This theory was also generalized to heteronuclear diatomic molecules [62,63] and polyatomic molecules [64,65]. The theory was formulated for a general electric field vector  $\mathbf{E}(t)$  and applied to a linearly polarized field case and to a more general case of an elliptically polarized field in Ref. [54]. In the present paper we generalize it to the case of a bicircular laser field.

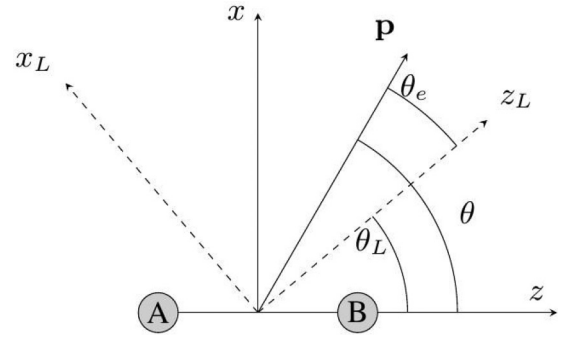


FIG. 1. Schematic presentation of the coordinate systems used in the paper. Molecule  $AB$  is along the  $z$  axis in the  $zx$  coordinate system, while the laser field is defined in the  $z_L x_L$  system. The ionized electron is emitted with the momentum  $\mathbf{p}$  whose direction is determined by the angle  $\theta$  ( $\theta_e$ ) with respect to the  $z$  ( $z_L$ ) axis.

### A. Notation

Let us first fix our notation. We suppose that the laser field, the molecule, and the emitted electron all lay in the same plane. The bicircular field is planar and diatomic molecules are linear so that we expect the main symmetry effects for planar geometry. HHG spectra of planar polyatomic molecules can be used to determine the orientation of the molecule in the plane [34] (for HHG by nonplanar molecules driven by a bicircular field, see Ref. [35]). The internuclear vector  $\mathbf{R}$  of our diatomic molecule  $AB$  is along the  $z$  axis, while the laser field is defined in the coordinate system  $z_L x_L$ , which is rotated with respect to the  $zx$  system by the angle  $\theta_L$  around the  $y = y_L$  axis, which is perpendicular to the polarization plane. The emitted electron momentum  $\mathbf{p}$  is in the direction determined by the angles  $\theta$  and  $\theta_e$  in the  $zx$  and  $z_L x_L$  system, respectively (see Fig. 1;  $\theta_e = \theta - \theta_L$ ). For the same bicircular field component strengths and relative phases fixed to zero ( $E_1 = E_2 = E_L$ ,  $\phi_1 = \phi_2 = 0$ , and  $x \rightarrow z$  and  $y \rightarrow x$  in the notation of Ref. [43]; we chose to follow the notation of Ref. [54]), the electric field vector has the form

$$\mathbf{E}(t) = \frac{E_L}{\sqrt{2}} \{ [\sin(r\omega t) + \sin(s\omega t)] \hat{\mathbf{e}}_{Lz} - [\cos(r\omega t) - \cos(s\omega t)] \hat{\mathbf{e}}_{Lx} \}. \quad (1)$$

Relation between the unit vectors of the corresponding coordinate systems is given by

$$\begin{aligned} \hat{\mathbf{e}}_{Lz} &= \hat{\mathbf{z}} \cos \theta_L + \hat{\mathbf{x}} \sin \theta_L, \\ \hat{\mathbf{e}}_{Lx} &= -\hat{\mathbf{z}} \sin \theta_L + \hat{\mathbf{x}} \cos \theta_L. \end{aligned} \quad (2)$$

### B. $T$ -matrix element

Generally, for a diatomic molecule with the internuclear vector  $\mathbf{R}$ , the  $T$ -matrix element of the (H)ATI process in which  $n$  photons of frequency  $\omega$  are absorbed from the laser field can be written as

$$T_{\mathbf{R}\mathbf{p}i}(n) = \int_0^T \frac{dt}{T} T_{\mathbf{R}\mathbf{p}i}(t) e^{in\omega t}, \quad (3)$$

with the time-periodic function, having the period  $T = 2\pi/\omega$ ,

$$\mathcal{T}_{\mathbf{R}\mathbf{p}i}(t) = \mathcal{F}_{\mathbf{R}\mathbf{p}i}(t)e^{i\mathcal{U}(t)} = \sum_{n=-\infty}^{\infty} T_{\mathbf{R}\mathbf{p}i}(n)e^{-in\omega t}, \quad (4)$$

with  $\mathcal{U}(t) = \mathbf{p} \cdot \boldsymbol{\alpha}(t) + \int^t d\tau \mathbf{A}^2(\tau)/2 - U_p t$ , where  $\boldsymbol{\alpha}(t) = \int^t d\tau \mathbf{A}(\tau)$ ,  $U_p$  is the ponderomotive energy, and  $\mathbf{A}(t) = -\int^t d\tau \mathbf{E}(\tau)$ , which is explicitly given by

$$\mathbf{A}(t) = \frac{E_L}{r\omega\sqrt{2}} \left\{ \left[ \cos(r\omega t) + \frac{r}{s} \cos(s\omega t) \right] \hat{\mathbf{e}}_{Lz} + \left[ \sin(r\omega t) - \frac{r}{s} \sin(s\omega t) \right] \hat{\mathbf{e}}_{Lx} \right\}. \quad (5)$$

The lowest-order term of the MSFA, which corresponds to the direct ATI electrons, within the dressed molecular length gauge [51], is described by the matrix element

$$\mathcal{F}_{\mathbf{R}\mathbf{p}i}^{(0)}(t) = \sum_{q=\pm 1} e^{iq\mathbf{p} \cdot \mathbf{R}/2} \langle \mathbf{p} + \mathbf{A}(t) | \mathbf{E}(t) \cdot \mathbf{r} | \Psi_q^{(0)} \rangle, \quad (6)$$

while the first-order term (improved MSFA), which corresponds to the rescattered electrons, is given by [53]

$$\begin{aligned} \mathcal{F}_{\mathbf{R}\mathbf{p}i}^{(1)}(t) &= -ie^{-iS_{\mathbf{k}_{st}}(t)} \int_0^\infty d\tau \left( \frac{2\pi}{i\tau} \right)^{3/2} \\ &\times e^{i[S_{\mathbf{k}_{st}}(\tau') - \Delta E(\mathbf{R})\tau]} \exp\left(-\frac{i}{2\tau} \frac{\partial^2}{\partial \mathbf{k}^2}\right) \\ &\times \sum_{q'=\pm 1} V_{\mathbf{e}\mathbf{K}}^{q'} \sum_{q=\pm 1} e^{i(q\mathbf{k} - q'\mathbf{K}) \cdot \mathbf{R}/2} \\ &\times \langle \mathbf{k} + \mathbf{A}(\tau') | \mathbf{r} \cdot \mathbf{E}(\tau') | \Psi_q^{(0)} \rangle_{\mathbf{k}=\mathbf{k}_{st}}, \end{aligned} \quad (7)$$

with  $\tau' = t - \tau$ ,  $\mathbf{K} = \mathbf{k} - \mathbf{p}$ ,  $\mathbf{k}_{st} = \int_t^{\tau'} dt' \mathbf{A}(t')/\tau$  the stationary electron momentum, and  $S_{\mathbf{k}}(t) = \int^t dt' [\mathbf{k} + \mathbf{A}(t')]^2/2$ . Here  $|\Psi_q^{(0)}\rangle = \sum_a c_{qa} |\psi_a^{(0)}\rangle$  represents the ground-state molecular wave function expressed as a linear combination of the atomic orbitals, which are presented by the linear combination of the Slater-type orbitals [51]. Finally,  $V_{\mathbf{e}\mathbf{K}}^{q'}$  is the Fourier transform of the rescattering potential on the  $q'$  center ( $q' = +1$  corresponds to the center A, while  $q' = -1$  corresponds to the center B) and  $\Delta E(\mathbf{R})$  is the ionization energy which includes vibrational degrees of freedom (for details, see Ref. [53]).

### III. SYMMETRY CONSIDERATIONS

#### A. Rotational symmetries

As homonuclear diatomic molecules are considered here (A and B represent the atoms of the same kind), both the ATI and HATI spectra should be invariant with respect to the rotation of the molecule about the  $y$  axes (that traverse molecular center; compare Fig. 8 in Appendix A) by  $180^\circ$ . This can be confirmed by considering the transition matrix element that corresponds to the molecule rotated about the  $y$  axes by  $180^\circ$ . It can be shown that the differential ionization rate, which is defined by ( $p = |\mathbf{p}|$ )

$$w_{\mathbf{R}\mathbf{p}i}(n) = 2\pi p |T_{\mathbf{R}\mathbf{p}i}(n)|^2, \quad (8)$$

is invariant with respect to the rotation of the coordinate system by the angle  $180^\circ$  (or  $-180^\circ$ ) about the  $y$  axis. According to the chosen notation (see Fig. 1), this rotation leads to the transformation

$$\theta \rightarrow \theta \pm 180^\circ, \quad \theta_L \rightarrow \theta_L \pm 180^\circ, \quad \theta_e \rightarrow \theta_e. \quad (9)$$

The proof is relegated to Appendix A. This rotational symmetry is generally valid, i.e., not only within the MSFA and improved MSFA.

Let us now consider the rotational symmetry of our bicircular field, having in mind that  $y = y_L$ . The bicircular field (1) obeys the following dynamical symmetry

$$\begin{aligned} \mathbf{E}'(t) &\equiv R_y(\alpha_j) \mathbf{E}(t) = \mathbf{E}(t + \tau_j), \\ \tau_j &= jT/(r+s), \quad \alpha_j = -r\omega\tau_j, \quad j - \text{integer}, \end{aligned} \quad (10)$$

where the prime denotes the vector rotated by the  $2 \times 2$  rotation matrix  $R_y(\alpha_j)$  by the angle  $\alpha_j$  about the  $y$  axis. Analogous relations are valid for the vectors  $\mathbf{A}(t)$  and  $\boldsymbol{\alpha}(t)$ . Therefore, a rotation by the angle  $\alpha_j$  about the  $y$  axis is equivalent to a translation in time by  $\tau_j$ . We will also use the fact that the Slater-type orbitals satisfy the relation  $\psi_a^{(0)}(\mathbf{r}) = \psi_a^{(0)}(\mathbf{r}')$  and that the scalar product is invariant with respect to rotation. Then, analogously to the atomic case considered in Ref. [43], and taking into account that the magnetic quantum number  $m_a$  is the same for all orbitals of the chosen molecule, we obtain  $\mathcal{F}_{\mathbf{R}\mathbf{p}i}^{(0)}(t) = \mathcal{F}_{\mathbf{R}'\mathbf{p}'i}(t)$  and analogous relation for the rescattering matrix element. The prime on  $i$  means that the molecular ground-state wave function is also rotated. This symmetry is generally valid, as shown in Appendix B, and we have

$$w_{\mathbf{R}'\mathbf{p}'i}(n) = w_{\mathbf{R}\mathbf{p}i}(n); \quad (11)$$

i.e., the differential ionization rate is invariant with respect to the simultaneous rotation of the vectors  $\mathbf{R}$  and  $\mathbf{p}$  around the  $y$  axis by the angle  $\alpha_j$ , with the fixed laser electric field vector. This is equivalent to the rotation of the vector  $\mathbf{E}(t)$  by the angle  $-\alpha_j$ , with fixed  $\mathbf{R}$  and  $\mathbf{p}$  [66]. Therefore, the differential ionization rate is invariant with respect to the transformation

$$\theta \rightarrow \theta, \quad \theta_L \rightarrow \theta_L - \alpha_j, \quad \theta_e \rightarrow \theta_e + \alpha_j. \quad (12)$$

For example, for  $r = 1$ ,  $s = 2$ , and  $\alpha_1 = -120^\circ$ , we obtain that the rate is invariant with respect to the transformation:  $\theta_L \rightarrow \theta_L + 120^\circ$ ,  $\theta_e \rightarrow \theta_e - 120^\circ$ . This is illustrated in Figs. 2 and 3. From Fig. 2 we see that the relative position of the molecule rotated by the angle  $-120^\circ$  and fixed field ( $\theta_L = 0^\circ$ ) in the upper panel is the same as the relative position of the unrotated molecule and the field rotated by the angle  $120^\circ$  in the bottom panel. Figure 3 illustrates the invariance with respect to the transformation (12) for  $\theta_L = 0^\circ$  and  $\alpha_1 = -120^\circ$ , i.e.,  $(\theta_L = 0^\circ, \theta_e)$  (upper panel)  $\rightarrow$   $(\theta'_L = 120^\circ, \theta'_e = \theta_e - 120^\circ)$  (lower panel), and  $w(\theta_L, \theta_e) = w(\theta'_L, \theta'_e)$ .

If one combines previously analyzed transformation (9), which leaves spectra of homonuclear diatomic molecule unchanged (i.e.,  $\theta_e \rightarrow \theta_e$ ), and rotation of the field (only) by, for example,  $-120^\circ$ , so that  $\theta \rightarrow \theta$ ,  $\theta_L \rightarrow \theta_L - 120^\circ$ , the resulting combination leads to the transformation  $\theta \rightarrow \theta + 180^\circ$ ,  $\theta_L \rightarrow \theta_L + 60^\circ$ ,  $\theta_e \rightarrow \theta_e + 120^\circ$ . Illustration of this result is given in Fig. 4. The spectrum shown in the upper panel ( $\theta_L = 30^\circ$ ), rotated by  $120^\circ$  ( $\theta_e \rightarrow \theta_e + 120^\circ$ ) is the same as

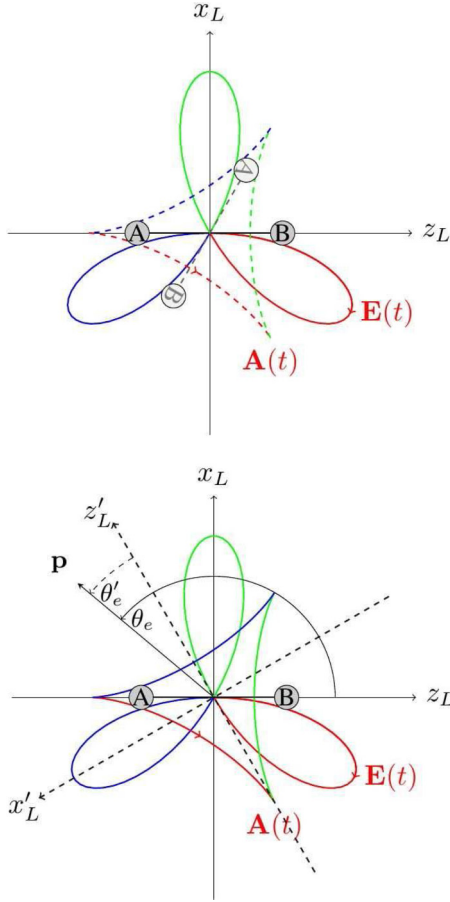


FIG. 2. Upper panel: Tips of the electric field  $\mathbf{E}(t)$  and the vector potential  $\mathbf{A}(t)$  are presented, with first third of a cycle in red, second third in green, and the last third in blue. Position of the molecule rotated by  $-120^\circ$  about the  $y$  axis is also shown. Lower panel: Position of the new laser field coordinate system (denoted with the primes) rotated by  $120^\circ$  about the  $y$  axis. The electron emission angles  $\theta_e$  and  $\theta'_e$  for two laser field coordinate systems are shown.

the spectrum shown in the lower panel ( $\theta_L = 30^\circ + 60^\circ$ ), i.e.,  $w(\theta_L = 30^\circ, \theta_e) = w(\theta'_L = 90^\circ, \theta_e + 120^\circ)$ .

### B. Reflection symmetries

For certain molecular orientation with respect to the laser field, ATI spectrum exhibits reflection symmetry, while HATI spectrum never does for the same reason as in atomic case. The vector potential  $\mathbf{A}(t)$  (see Fig. 2) obeys reflection symmetry about the axis at the angles  $\beta_j = -\alpha_j/2 = jr\pi/(r+s)$  with respect to the positive  $A_z$  axis. For atoms the differential ionization rate of the direct electron obeys the same reflection symmetry as well [43], but for molecules this is not the case. This is because the direct ATI  $T$ -matrix element  $T_{\mathbf{R}\mathbf{p}i}^{(0)}(n)$  has to be invariant (up to a phase factor) with respect to the reflection of the vector potential  $\mathbf{A}(t)$  and the molecule about the same axis.

However, for homonuclear diatomic molecules it is possible to show that for particular molecular orientations the direct differential ionization rate is invariant with respect to specific reflection transformations. Let us first fix the angle  $\theta_L = 0^\circ$

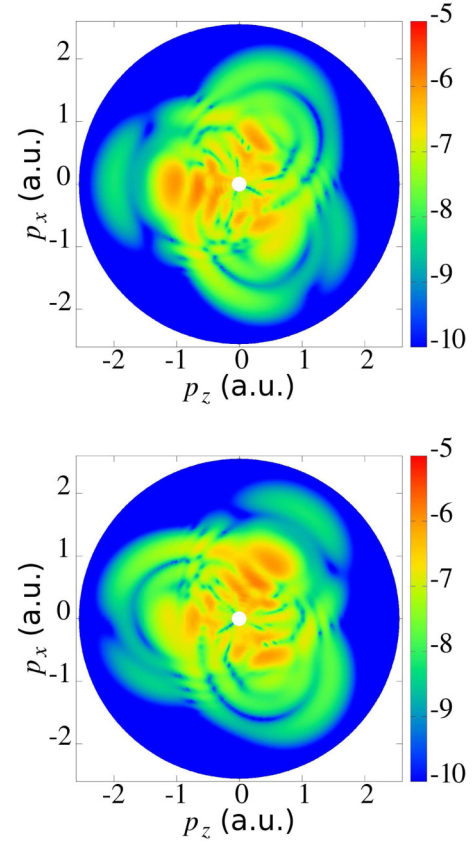


FIG. 3. Electron HATI spectra obtained using the improved MSFA (both the direct and the rescattered electrons are included) for the  $\text{N}_2$  molecule, laser intensity given by  $E_1^2 = E_2^2 = 1 \times 10^{14}$  W/cm $^2$ , and the fundamental wavelength of 800 nm. The electron momentum plane is defined with respect to the laser field coordinate system:  $p_z = p \cos \theta_e$ ,  $p_x = p \sin \theta_e$ . Upper panel:  $\theta_L = 0^\circ$ . Lower panel:  $\theta_L = 120^\circ$ . The false color scale covers five orders of magnitude.

so that  $\theta_e = \theta$ , and denote by double prime the vectors reflected with respect to the  $yz$  plane (see Fig. 5). Since this transformation corresponds to the inversion of the unit vector of the  $x$  axis,  $\hat{\mathbf{x}} \rightarrow -\hat{\mathbf{x}}$  we denote the corresponding operator by  $P_x(\theta_L = 0^\circ)$ . Using Eqs. (1), (2), and (5), we obtain

$$\begin{aligned} \mathbf{E}''(t) &\equiv P_x(\theta_L = 0^\circ)\mathbf{E}(t) = -\mathbf{E}(-t), & \mathbf{A}''(t) &= \mathbf{A}(-t), \\ \boldsymbol{\alpha}''(t) &= -\boldsymbol{\alpha}(-t), & \mathbf{R}'' &= \mathbf{R} = R\hat{\mathbf{z}}. \end{aligned} \quad (13)$$

In Appendix C it is shown that

$$w_{\mathbf{R}\mathbf{p}i}^{(0)}(n) = w_{\mathbf{R}\mathbf{p}'i}^{(0)}(n), \quad (14)$$

i.e., the direct differential ionization rate for the case  $z = z_L$  and  $x = x_L$  is invariant with respect to the transformation

$$\theta \rightarrow -\theta, \quad \theta_L = 0^\circ, \quad \theta_e \rightarrow -\theta_e. \quad (15)$$

Next, we fix the angle  $\theta_L = 90^\circ$  and denote by triple prime the vectors reflected with respect to the  $xy$  plane. In this case we have  $\hat{\mathbf{z}} \rightarrow -\hat{\mathbf{z}}$ , the corresponding operator is  $P_z(\theta_L = 90^\circ)$ , and

$$\begin{aligned} \mathbf{E}'''(t) &\equiv P_z(\theta_L = 90^\circ)\mathbf{E}(t) = -\mathbf{E}(-t), & \mathbf{A}'''(t) &= \mathbf{A}(-t), \\ \boldsymbol{\alpha}'''(t) &= -\boldsymbol{\alpha}(-t), & \mathbf{R}''' &= -\mathbf{R}. \end{aligned} \quad (16)$$



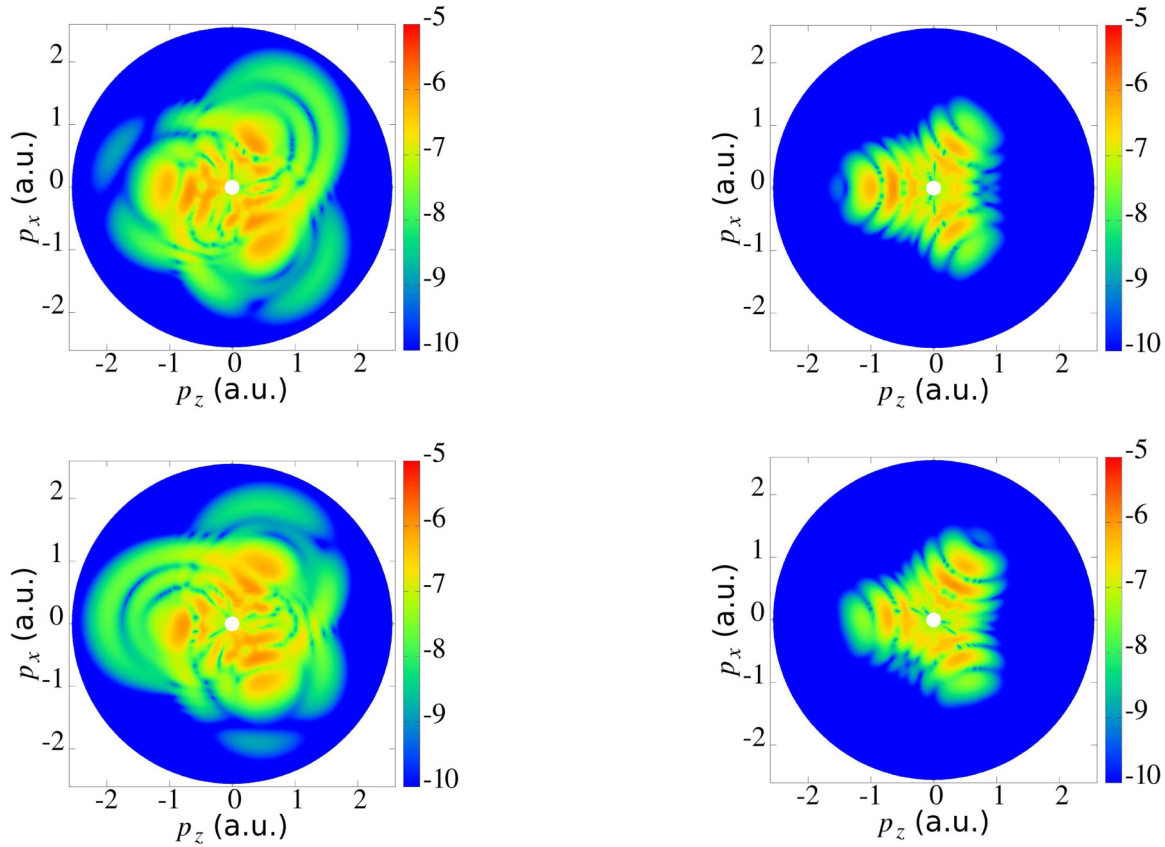


FIG. 4. Same as in Fig. 3, but for  $\theta_L = 30^\circ$  (upper panel) and  $\theta_L = 90^\circ$  (lower panel).

Again, the proof of the invariance of the direct differential ionization rate with respect to the transformation  $P_z(\theta_L = 90^\circ)$ , i.e., of the relation

$$w_{\mathbf{R}\mathbf{p}i}^{(0)}(n) = w_{\mathbf{R}\mathbf{p}'i}^{(0)}(n), \quad (17)$$

is relegated to Appendix C. This corresponds to the transformation

$$\theta \rightarrow 180^\circ - \theta, \quad \theta_L = 90^\circ, \quad \theta_e \rightarrow -\theta_e. \quad (18)$$

Therefore, we have found four different transformations, given by Eqs. (9), (12), (15), and (18), with respect to which the direct ATI electron spectra are invariant. By combining these transformations various new symmetry relations can be obtained. An example is shown in Fig. 6. The top panel shows the direct spectrum for  $\theta_L = 0^\circ$ . It is obvious that the transformation (15) is satisfied since the presented spectrum is

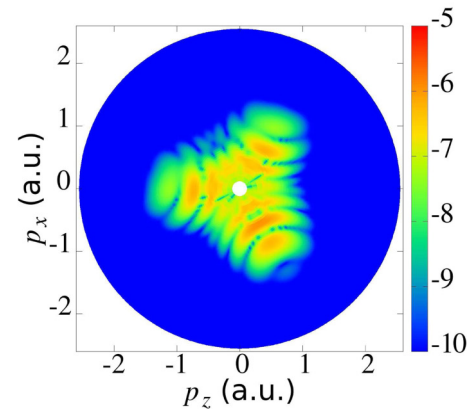


FIG. 6. Direct electron spectra for the same parameters as in Fig. 3, but for  $\theta_L = 0^\circ$  (top panel),  $\theta_L = 120^\circ$  (middle panel), and  $\theta_L = 60^\circ$  (bottom panel).

invariant with respect to the transformation  $\theta_e \rightarrow -\theta_e$  (this is the invariance with respect to the reflection about the  $p_z$  axis, i.e., the axis for which  $p_x = 0$ ). Combining the transformations (12) (for  $\alpha_1 = -120^\circ$ ) and (15) we obtain that for  $\theta_L = 120^\circ$  the direct spectrum should be invariant with respect to the transformation  $\theta_e \rightarrow 120^\circ - \theta_e$ . This situation is shown in the middle panel of Fig. 6. This spectrum is invariant with respect to the reflection about the axis obtained rotating the  $z$  axis (i.e., the  $p_z$  axis) for  $-120^\circ$  about the  $y$  axis. The bottom panel in Fig. 6 shows the situation where the direct electron spectrum for  $\theta_L = 60^\circ$  is invariant with respect to the reflection about the axis which is obtained by rotating the  $z$  axis for  $120^\circ$  about the  $y$  axis. This symmetry follows from Eq. (12) for  $\alpha_1 = 120^\circ$

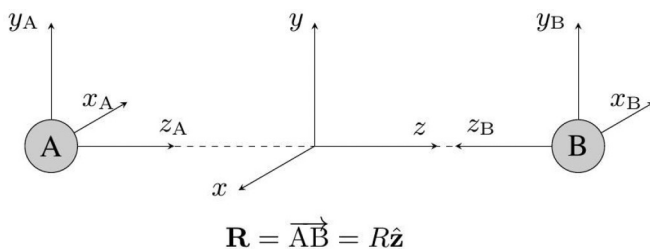


FIG. 5. Reflection of the molecule with respect to the  $yz$  plane.

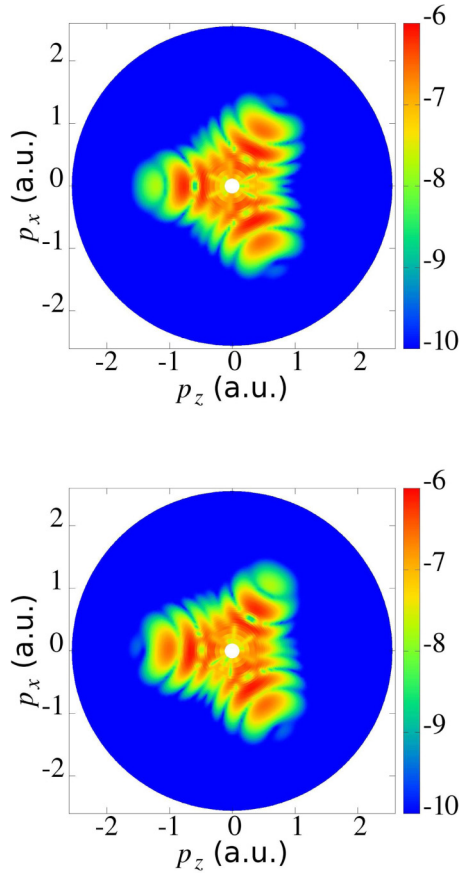


FIG. 7. Direct electron spectra for the same parameters as in Fig. 3, but for  $\theta_L = 90^\circ$  (upper panel) and  $\theta_L = 30^\circ$  (lower panel).

(which gives  $\theta_L \rightarrow \theta_L - 120^\circ$ ,  $\theta_e \rightarrow \theta_e + 120^\circ$ ), Eq. (9) (this further gives  $\theta_L \rightarrow \theta_L + 60^\circ$ ,  $\theta_e \rightarrow \theta_e + 120^\circ$ ), and Eq. (15), which leads to  $\theta_L = 60^\circ$ ,  $\theta_e \rightarrow -\theta_e - 120^\circ$ . It is obvious that the spectrum in the bottom panel of Fig. 6 is invariant with respect to this final transformation  $\theta_e \rightarrow -\theta_e - 120^\circ$ .

From Eqs. (17) and (18) it follows that the direct electron spectrum for  $\theta_L = 90^\circ$  exhibits reflection symmetry about the  $p_z$  ( $p_x = 0$ ) axis. This is clearly visible in the upper panel of Fig. 7.

Finally, let us consider combined transformations given by Eq. (12) for  $\alpha_1 = -120^\circ$  and Eq. (9) for  $-180^\circ$ . This gives that the differential ionization rate for  $(\theta_L, \theta_e)$  is equal to the rate for  $(\theta_L + 120^\circ - 180^\circ, \theta_e - 120^\circ)$ . On the other hand, from Eq. (18) it follows that the direct rate for  $\theta_L = 90^\circ$  is the same for the angles  $\theta_e$  and  $-\theta_e$ . From this we obtain that for  $\theta_L = 90^\circ - 60^\circ = 30^\circ$  the direct rate is invariant with respect to the transformation  $\theta_e \rightarrow -\theta_e + 120^\circ$ . This is presented in the lower panel of Fig. 7, where it is clearly visible that for  $\theta_L = 30^\circ$  the direct rate is invariant with respect to the transformation  $\theta_e \rightarrow 120^\circ - \theta_e$ . Notice that the same invariance with respect to  $\theta_e \rightarrow 120^\circ - \theta_e$  was found for  $\theta_L = 120^\circ$  (middle panel of Fig. 6), but that the corresponding spectra are different.

#### IV. CONCLUSIONS

The rotational and reflection symmetries, discovered in the ATI spectra of atoms ionized by a strong bicircular laser field,

cannot be directly generalized to the molecular systems. The reason is that the internuclear molecular axis can be oriented in an arbitrary direction with respect to the bicircular field. This is true even in the coplanar case considered in the present paper: The internuclear axis lays in the bicircular field plane, but still can be oriented by an arbitrary angle  $-\theta_L$  with respect to the axis of the first component of our bicircular field.

Nevertheless, in the present paper we were able to identify four symmetries (two rotational and two reflection symmetries) which are satisfied in ATI of homonuclear diatomic molecules. The first symmetry is with respect to the rotation of the molecule by  $\pm 180^\circ$  about the axis perpendicular to the internuclear axis and to the bicircular field polarization plane. The second rotational symmetry is related to the rotation of the  $r\omega$ - $s\omega$  bicircular electric field vector by the angle  $-\alpha_j = jr2\pi/(r+s)$  ( $j$  integer) around the axis perpendicular to the polarization plane. For the  $\omega$ - $2\omega$  bicircular field the corresponding rotation angle is  $-\alpha_1 = 120^\circ$ . Both mentioned rotational symmetries are valid both for the direct and for the rescattered HATI electrons. We have illustrated our results by few examples of the HATI spectra of the  $N_2$  molecule for different molecular orientation angles.

The direct ATI spectra of homonuclear diatomic molecules satisfy two additional reflection symmetries. The first one is with respect to the axis perpendicular to the internuclear  $z$  axis:  $\hat{x} \rightarrow -\hat{x}$  for  $\theta_L = 0^\circ$ , i.e., for  $z = z_L$ . The second one is the reflection symmetry:  $\hat{z} \rightarrow -\hat{z}$  for  $\theta_L = 90^\circ$ .

All four mentioned symmetries, given by Eqs. (9), (12), (15), and (18), can be combined in various ways, giving additional symmetry relations. This is illustrated by numerical examples of ATI spectra of the  $N_2$  molecule.

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#### APPENDIX A: INVARIANCE OF THE DIFFERENTIAL IONIZATION RATE WITH RESPECT TO THE ROTATION BY THE ANGLE $180^\circ$

Rotation of the molecule about the  $y$  axis (that traverse molecular center) by  $180^\circ$  leads to the transformations  $\mathbf{R} \rightarrow -\mathbf{R}$  (see Fig. 8) and also to the transformation of the atomic wave functions  $\psi_a^{(0)}$ , where  $a$  denotes a specific atomic Slater-type orbital. Rotation by  $180^\circ$  about the corresponding atomic  $y_J$  axis ( $J$  stands for the center A or B) can be considered as the transformation due to  $z_J \rightarrow -z_J$ ,  $x_J \rightarrow -x_J$ . First,  $z_J \rightarrow -z_J$  leads to  $\psi_a^{(0)} \rightarrow (-1)^{l_a+m_a} \psi_a^{(0)}$ , while  $x_J \rightarrow -x_J$  leads to  $\psi_a^{(0)} \rightarrow (-1)^{m_a} \psi_a^{(0)*}$ .

Altogether, for the direct electrons the rotation of the homonuclear diatomic molecule by  $180^\circ$  about the  $y$  axis leads to the transformation  $\sum_q c_{qa} e^{iq\mathbf{p}\cdot\mathbf{R}/2} \psi_a^{(0)} \rightarrow \sum_q c_{qa} e^{-iq\mathbf{p}\cdot\mathbf{R}/2} (-1)^{l_a} \psi_a^{(0)}$  [67]. Here,  $q$  is a counter that gets two values,  $+1$  and  $-1$ , that correspond to the atoms A and B, respectively, and  $c_{-1a} = (-1)^{p+m_a+l_a} c_{1a}$ . The factor  $(-1)^{m_a+l_a}$  comes from the fact that  $\hat{\mathbf{z}}_B = -\hat{\mathbf{z}}_A$ , while  $(-1)^p$  depends on

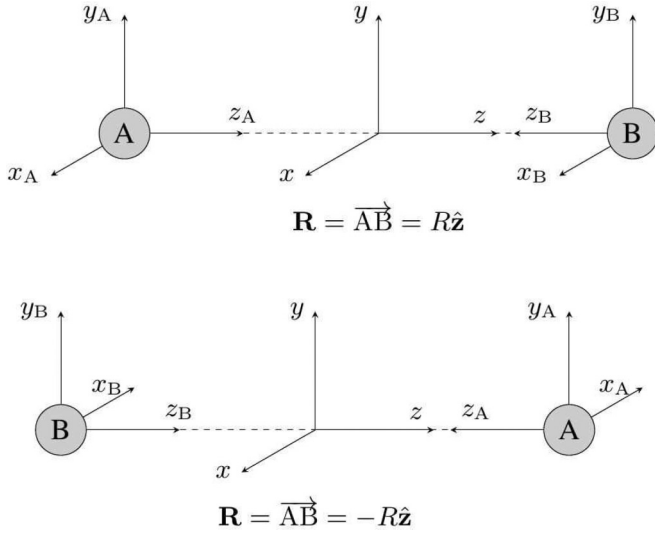


FIG. 8. Upper panel: Diatomic molecule AB with the denoted axes. Lower panel: The same molecule rotated by  $180^\circ$  about the  $x$  axis that passes through the center of the molecule.

the parity of the highest occupied molecular orbital (gerade or ungerade [51]). Taking all this into account, for the rotated molecule we get

$$\begin{aligned} & \sum_q c_{qa} e^{-iq\mathbf{p}\cdot\mathbf{R}/2} (-1)^{l_a} \psi_a^{(0)} \\ &= c_{1a} \psi_a^{(0)} [(-1)^{p+m_a+2l_a} e^{i\mathbf{p}\cdot\mathbf{R}/2} + (-1)^{l_a} e^{-i\mathbf{p}\cdot\mathbf{R}/2}]. \end{aligned}$$

This can be rearranged as  $(-1)^{p+m_a} \sum_q c_{qa} e^{iq\mathbf{p}\cdot\mathbf{R}/2} \psi_a^{(0)}$ , and, as  $p$  and  $m_a$  are the same for all atomic orbital  $a$ , the difference in the direct matrix element for homonuclear diatomic molecule rotated by  $-180^\circ$  is just in an overall sign (phase factor) which makes no difference in the differential ionization rate (8).

For the transformation of the rescattering matrix element we consider Eq. (7).  $\Delta E(\mathbf{R})$  is equal to the ionization potential and does not depend on the sign of  $\mathbf{R}$ . With an analysis analogous to the one for the direct matrix element, it can be shown that  $\mathcal{F}_{\mathbf{R}\mathbf{p}i}^{(1)}$  for rotated molecule (by  $-180^\circ$ ), after rearranging the order of summation, just differs in an overall phase factor. This rearranging of order of summation of contributions  $T^{++} + T^{--} + \tilde{T}^{+-} + \tilde{T}^{-+}$  corresponds to the transformation  $R_y(180^\circ)\tilde{T}^{+-} = \tilde{T}^{-+}$ , and analogous transformation for other contributions [ $R_y(180^\circ)T^{++} = T^{--}$  and vice versa]. For the definition of the partial rates see Ref. [53].

#### APPENDIX B: INVARIANCE WITH RESPECT TO THE ROTATION BY THE BICIRCULAR FIELD DYNAMICAL SYMMETRY ANGLE $\alpha_j$

The exact ionization probability amplitude for a neutral diatomic molecule with fixed internuclear distance  $\mathbf{R}$  can be written in the form [51,53]

$$\begin{aligned} M_{\mathbf{R}\mathbf{p}i}(t, t') &= -i \int_{t'}^t d\tau \langle \Phi_{\mathbf{R}\mathbf{p}}(t) | U_{\mathbf{R}}(t, \tau) \\ &\quad \times (\mathbf{r} - e_R \mathbf{R}) \cdot \mathbf{E}(\tau) | \phi_{\mathbf{R}i}(\tau) \rangle, \end{aligned} \quad (\text{B1})$$

where  $U_{\mathbf{R}}(t, \tau)$  is the total time-evolution operator which corresponds to the Hamiltonian

$$H(\mathbf{r}, \mathbf{R}, t) = K(\mathbf{r}, \mathbf{R}) + V(\mathbf{r}, \mathbf{R}) + (\mathbf{r} - e_R \mathbf{R}) \cdot \mathbf{E}(t), \quad (\text{B2})$$

with  $K$  being the kinetic energy operator and  $V$  being the potential energy operator which describes the interaction of the electron and the atomic (ionic) centers A and B in the absence of the laser field. The relative charge  $e_R$  (in atomic units) is equal to  $+1/2$  or  $-1/2$  depending from which center the electron is ionized. The wave vectors  $\Phi_{\mathbf{R}\mathbf{p}}(t)$  and  $\phi_{\mathbf{R}i}$  are solutions of the Schrödinger equation with the operators  $H$  and  $K + V$ , respectively.

We introduce the unitary rotation operator  $D(\alpha_j) = \exp(-iJ_y\alpha_j)$ , where  $J_y$  is the  $y$  component of the total angular momentum operator and the angle  $\alpha_j$  is given by Eq. (10). The electron coordinate operator transforms according to  $\mathbf{r}' = D(\alpha_j)\mathbf{r}D^\dagger(\alpha_j) = R_y(\alpha_j)\mathbf{r}$ , while the other vectors  $(\mathbf{R}, \mathbf{p}, \mathbf{E})$  are not affected by this operator, but are transformed to  $(\mathbf{R}', \mathbf{p}', \mathbf{E}')$  under rotation transformation. Acting from the left by  $D(\alpha_j)$  on the Schrödinger equation for the wave vector  $|\Phi_{\mathbf{R}'\mathbf{p}'}(t)\rangle$  we get

$$\left[ i \frac{\partial}{\partial t} - D(\alpha_j)H(\mathbf{r}, \mathbf{R}', t)D^\dagger(\alpha_j) \right] D(\alpha_j)|\Phi_{\mathbf{R}'\mathbf{p}'}(t)\rangle = 0.$$

Introducing the time  $t'' = t - \tau_j$  and using relations  $D\mathbf{r}D^\dagger = \mathbf{r}'$ ,  $K(\mathbf{r}', \mathbf{R}') = K(\mathbf{r}, \mathbf{R})$ ,  $V(\mathbf{r}', \mathbf{R}') = V(\mathbf{r}, \mathbf{R})$ , and  $(\mathbf{r}' - e_R \mathbf{R}') \cdot \mathbf{E}(t'' + \tau_j) = (\mathbf{r} - e_R \mathbf{R}) \cdot \mathbf{E}(t'')$  (scalar product is invariant with respect to the rotation), we obtain

$$\left[ i \frac{\partial}{\partial t''} - H(\mathbf{r}, \mathbf{R}, t'') \right] D(\alpha_j)|\Phi_{\mathbf{R}'\mathbf{p}'}(t'' + \tau_j)\rangle = 0,$$

so that

$$|\Phi_{\mathbf{R}\mathbf{p}}(t)\rangle = D(\alpha_j)|\Phi_{\mathbf{R}'\mathbf{p}'}(t + \tau_j)\rangle, \quad (\text{B3})$$

which is a molecular analog of the relation (A2) from Ref. [26] (see also first reference in Ref. [46] for above-threshold detachment process). Furthermore, for the eigenvectors of the time-independent Hamiltonian  $T + V$  we can write

$$|\phi_{\mathbf{R}i}(t)\rangle = D(\alpha_j)|\phi_{\mathbf{R}'i'}(t)\rangle, \quad (\text{B4})$$

where  $i'$  denotes the rotated ground-state vector. Introducing this into Eq. (B1) we obtain

$$\begin{aligned} M_{\mathbf{R}\mathbf{p}i}(t, t') &= -i \int_{t'}^t d\tau \langle \Phi_{\mathbf{R}\mathbf{p}}(t + \tau_j) | D^\dagger(\alpha_j) U_{\mathbf{R}}(t, \tau) \\ &\quad \times D(\alpha_j) D^\dagger(\alpha_j) (\mathbf{r} - e_R \mathbf{R}) \cdot \mathbf{E}(\tau) \\ &\quad \times D(\alpha_j) D^\dagger(\alpha_j) | \phi_{\mathbf{R}i}(\tau) \rangle \\ &= -i \int_{t'}^t d\tau \langle \Phi_{\mathbf{R}'\mathbf{p}'}(t + \tau_j) | U_{\mathbf{R}}(t + \tau_j, \tau + \tau_j) \\ &\quad \times (\mathbf{r} - e_R \mathbf{R}') \cdot \mathbf{E}(\tau + \tau_j) | \phi_{\mathbf{R}'i'}(\tau) \rangle \\ &= e^{-i\Delta E(\mathbf{R})\tau_j} M_{\mathbf{R}'\mathbf{p}'i'}(t + \tau_j, t' + \tau_j). \end{aligned} \quad (\text{B5})$$

Since the rate is defined in the limit  $t' \rightarrow -\infty$  and  $t \rightarrow \infty$ , we have proved that the relation (11) is generally valid.

We have confirmed that the differential ionization rate is invariant with respect to the rotation by the  $(r + s)$ -bicircular

field dynamical symmetry angle  $\alpha_j = -2\pi jr/(r+s)$ . This result is generally valid, i.e., not only in the MSFA and improved MSFA. Another way to confirm this invariance is to use a rotating frame of reference. In this case [36], by

the time-dependent unitary transformation  $\exp(-i\bar{\omega}J_y t)$ ,  $\bar{\omega} = (s-r)\omega/2$ , the bicircular field becomes a linearly polarized field having the frequency  $\bar{\omega} = (r+s)\omega/2$ . Neglecting the Coriolis term one can derive selection rules for HHG.

### APPENDIX C: INVARIANCE OF THE DIRECT DIFFERENTIAL IONIZATION RATE WITH RESPECT TO THE REFLECTIONS $P_x(\theta_L = 0^\circ)$ AND $P_z(\theta_L = 90^\circ)$

Let us first consider the reflection  $P_x(\theta_L = 0^\circ)$  (see Sec. III B). According to the result of Appendix A, the transformation  $x_j \rightarrow -x_j$  leads to  $\psi_a^{(0)}(\mathbf{r}) = \psi_a^{(0)}(\mathbf{r}') = (-1)^{m_a} [\psi_a^{(0)}(\mathbf{r}'')]^*$ . We also have  $d\mathbf{r} = d\mathbf{r}'$  and  $\mathbf{p} \cdot \mathbf{R} = \mathbf{p}' \cdot \mathbf{R}'$ . From Eq. (13) we obtain  $[\mathbf{p} + \mathbf{A}(t)] \cdot \mathbf{r} = [\mathbf{p}' + \mathbf{A}(-t)] \cdot \mathbf{r}'$ ,  $\mathbf{r} \cdot \mathbf{E}(t) = -\mathbf{r}' \cdot \mathbf{E}(-t)$ ,  $\mathbf{p} \cdot \boldsymbol{\alpha}(t) = -\mathbf{p}' \cdot \boldsymbol{\alpha}(-t)$ , and  $\int^t \mathbf{A}^2(\tau) d\tau = -\int^{-t} \mathbf{A}^2(\tau) d\tau$ . Using this and Eqs. (3), (4), and (6), we obtain

$$\begin{aligned} T_{\mathbf{R}\mathbf{p}i}^{(0)}(n) &= \int_0^T \frac{dt}{T} e^{i[\mathbf{p}\cdot\boldsymbol{\alpha}(t) + \int^t d\tau \mathbf{A}^2(\tau)/2 + (n\omega - U_p)t]} \sum_{q=\pm 1} e^{iq\mathbf{p}\cdot\mathbf{R}/2} \int \frac{d\mathbf{r}}{(2\pi)^{3/2}} e^{i[\mathbf{p}+\mathbf{A}(t)]\cdot\mathbf{r}} \cdot \mathbf{E}(t) \sum_a c_{qa} \psi_a^{(0)}(\mathbf{r}) \\ &= \int_0^T \frac{dt}{T} e^{-i[\mathbf{p}'\cdot\boldsymbol{\alpha}(-t) + \int^{-t} d\tau \mathbf{A}^2(\tau)/2 + (n\omega - U_p)(-t)]} \sum_{q=\pm 1} e^{iq\mathbf{p}'\cdot\mathbf{R}'/2} \\ &\quad \times \int \frac{d\mathbf{r}''}{(2\pi)^{3/2}} e^{i[\mathbf{p}'+\mathbf{A}(-t)]\cdot\mathbf{r}''} [-\mathbf{r}'' \cdot \mathbf{E}(-t)] \sum_a c_{qa} (-1)^{m_a} [\psi_a^{(0)}(\mathbf{r}'')]^*. \end{aligned} \quad (C1)$$

After the substitution  $q' = -q$ ,  $t' = -t$ ,  $\mathbf{r} = -\mathbf{r}''$ ,  $c_{qa} = c_{q'a}(-1)^{p+m_a+l_a}$ ,  $\psi_a^{(0)}(-\mathbf{r}) = (-1)^{l_a} \psi_a^{(0)}(\mathbf{r})$ , we get

$$\begin{aligned} T_{\mathbf{R}\mathbf{p}i}^{(0)}(n) &= \int_0^T \frac{dt'}{T} e^{-i[\mathbf{p}''\cdot\boldsymbol{\alpha}(t') + \int^{t'} d\tau \mathbf{A}^2(\tau)/2 + (n\omega - U_p)t']} \sum_{q'=\pm 1} e^{-iq'\mathbf{p}''\cdot\mathbf{R}''/2} \\ &\quad \times \int \frac{d\mathbf{r}}{(2\pi)^{3/2}} e^{-i[\mathbf{p}''+\mathbf{A}(t')]\cdot\mathbf{r}} \cdot \mathbf{E}(t') \sum_a c_{q'a} (-1)^{p+m_a} [\psi_a^{(0)}(\mathbf{r})]^* = (-1)^{p+m_a} [T_{\mathbf{R}\mathbf{p}''i}^{(0)}(n)]^*. \end{aligned} \quad (C2)$$

Taking into account the definition (8) and the fact that  $\mathbf{R}'' = \mathbf{R} = R\hat{\mathbf{z}}$ , we finally obtain the result (14).

Next, we consider the reflection  $P_z(\theta_L = 90^\circ)$ . Using  $\psi_a^{(0)}(\mathbf{r}) = \psi_a^{(0)}(\mathbf{r}''') = (-1)^{l_a+m_a} \psi_a^{(0)}(\mathbf{r}''')$  and Eq. (16), we obtain

$$\begin{aligned} T_{\mathbf{R}\mathbf{p}i}^{(0)}(n) &= \int_0^T \frac{dt}{T} e^{-i[\mathbf{p}'''\cdot\boldsymbol{\alpha}(-t) + \int^{-t} d\tau \mathbf{A}^2(\tau)/2 + (n\omega - U_p)(-t)]} \sum_{q=\pm 1} e^{iq\mathbf{p}'''\cdot\mathbf{R}'''/2} \\ &\quad \times \int \frac{d\mathbf{r}'''}{(2\pi)^{3/2}} e^{i[\mathbf{p}'''+\mathbf{A}(-t)]\cdot\mathbf{r}'''} [-\mathbf{r}''' \cdot \mathbf{E}(-t)] \sum_a c_{qa} (-1)^{l_a+m_a} \psi_a^{(0)}(\mathbf{r}'''). \end{aligned} \quad (C3)$$

With the substitution  $t' = -t$ ,  $\mathbf{r} = -\mathbf{r}'''$ ,  $\psi_a^{(0)}(-\mathbf{r}) = (-1)^{l_a} \psi_a^{(0)}(\mathbf{r})$ , with  $\psi_a^{(0)}$  real [67], and using  $\mathbf{R}''' = -\mathbf{R}$ , we get

$$\begin{aligned} T_{\mathbf{R}\mathbf{p}i}^{(0)}(n) &= \int_0^T \frac{dt'}{T} e^{-i[\mathbf{p}'''\cdot\boldsymbol{\alpha}(t') + \int^{t'} d\tau \mathbf{A}^2(\tau)/2 + (n\omega - U_p)t']} \sum_{q=\pm 1} e^{-iq\mathbf{p}'''\cdot\mathbf{R}'''/2} \\ &\quad \times \int \frac{d\mathbf{r}}{(2\pi)^{3/2}} e^{-i[\mathbf{p}'''+\mathbf{A}(t')]\cdot\mathbf{r}} \cdot \mathbf{E}(t') \sum_a c_{qa} (-1)^{m_a} [\psi_a^{(0)}(\mathbf{r})]^* = (-1)^{m_a} [T_{\mathbf{R}\mathbf{p}''i}^{(0)}(n)]^*. \end{aligned} \quad (C4)$$

From this we obtain the result (17).

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- [66] In fact, a simpler proof of the invariance of the differential ionization rate with respect to the rotation of the electric field vector by the angle  $-\alpha_j$  around the  $y$  axis can be obtained considering the transformed  $T$ -matrix element  $T'_{\mathbf{R}p_i}(n) = \int_0^T \frac{dt}{T} T'_{\mathbf{R}p_i}(t) e^{in\omega t} = e^{in\omega\tau_j} \int_{-\tau_j}^{T-\tau_j} \frac{dt}{T} T_{\mathbf{R}p_i}(t) e^{in\omega t}$ , where we used the relation (10). As

the field is periodic with the period  $T$ , it follows that  $T'_{\mathbf{R}p_i}(n)$  is equal to  $T_{\mathbf{R}p_i}(n)$  up to a phase factor.

- [67] In molecular calculations we are using real spherical harmonics [see Appendix A in S. Odžak and D. B. Milošević, *Phys. Rev. A* **79**, 023414 (2009)]. Therefore, we have  $\psi_a^{(0)*} = \psi_a^{(0)}$ . Information about rotation matrices for real spherical harmonics can be found in D. Pinchon and P. E. Hoggan, *J. Phys. A* **40**, 1597 (2007) and references therein.