Opt Quant Electron (2018) 50:214
https://doi.org/10.1007/s11082-018-1480-6

Molecules in a bicircular strong laser field

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Received: 14 October 2017 / Accepted: 18 April 2018 / Published online: 25 April 2018

Abstract Strong-field ionization of nonlinear planar triatomic molecules by a bicircular laser field is analyzed within the improved molecular strong-field approximation. Our calculations include additional interaction between the liberated electrons and atomic or ionic centers of the parent molecular ion. The used bicircular field consists of two counterrotating circularly polarized fields having angular frequencies $r\omega$ and $s\omega$, with integer r and s. In the case when the laser-field-polarization plane is parallel to the plane of the considered molecule (example of ozone molecule is analyzed), the corresponding photoelectron spectra are not rotationally symmetric. On the other hand, when these planes are mutually perpendicular, for the $(r\omega, s\omega) = (\omega, 3\omega)$ bicircular field, the electron spectra satisfy the corresponding rotational symmetries. Analyzing the obtained spectra and the corresponding symmetries, one can extract information about molecular orientation and structure. This technique may also be useful for more complex polyatomic molecules.

Keywords Improved molecular strong-field approximation · High-order above-threshold ionization · Reflection and rotational symmetry

This article is part of the Topical Collection on Focus on Optics and Bio-photonics, Photonica 2017.

Guest Edited by Jelena Radovanovic, Aleksandar Krmpot, Marina Lekic, Trevor Benson, Mauro Pereira, Marian Marciniak.

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1 Introduction

Above-threshold ionization (ATI) of atoms or molecules by a strong laser field is an interesting nonperturbative quantum-mechanical phenomenon discovered on atomic targets almost forty years ago (Agostini et al. 1979). In this process, an atomic or molecular electron becomes free after absorption of more photons than the minimum number necessary for ionization. The spectrum of the ionized electrons consists of peaks separated by the photon energy ω. This is the first step of more complex phenomena induced by a strong laser field. Since the middle of 90-ties, several novel phenomena whose energy spectra are characterized by a plateau in the electron energy spectra were discovered Paulus et al. (1994). The mentioned plateau region is due to the contribution of electrons that are driven back by the laser field to the parent ion and then elastically scattered off it. This process was named high-order above-threshold ionization or HATI [see, for example, review article (Becker et al. 2002) and references therein; HATI process includes both the (directly) ionized electrons in ATI process and rescattered electrons]. The HATI process is very similar to another laser induced process in which the returned electron recombines with the parent ion and a high-energy photon is emitted (Kulander et al. 1993). This process is called high-order harmonic generation (HHG). The energy spectra of HATI and HHG processes are characterized by a plateau which manifests itself 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. The key features of the HATI and HHG spectra were explained using the three-step model (Kulander et al. 1993; Corkum 1993). Both processes have been analyzed on molecular targets in the past decade. As molecules possess more complexity than atoms, various additional parameters must be taken into account when exploring molecular HATI and HHG. For example, double-slit or multiple-slit interference effects were observed in the HHG spectra (Lein et al. 2002; Kanai et al. 2005; Vozzi et al. 2005; Odžak and Milošević 2009) and in the HATI process (Busuladžić et al. 2008; Okunishi et al. 2009; Kang et al. 2010; Li et al. 2015). It was also shown that molecular symmetry and internuclear distance play an important role in (H)ATI of molecules (Busuladžić et al. 2008; Petersen et al. 2015).

In this paper we are interested in molecular high-order above-threshold ionization by a bicircular strong laser field. This and all other nonlinear processes governed by two counter-rotating circularly polarized fields having different angular frequencies have attracted much attention in the past few years. We will briefly survey recent references devoted to this topic. Atomic HHG by such a field was considered as early as 1995 (Eichmann et al. 1995; Long et al. 1995; Zuo and Bandrauk 1995) and was investigated theoretically in subsequent years using mostly *S*-matrix theory approach (Alon et al. 1998; Becker et al. 1999; Milošević et al. 2000; Milošević and Sandner 2000; Milošević et al. 2001; Milošević and Becker 2000). It was shown that the harmonic-generation efficiency is high and that only circularly polarized harmonics are generated. More recently, high-order harmonic generation by a bicircular field has again attracted attention as a source of strong circularly polarized harmonics which can be used for various applications (Fleischer et al. 2014; Pisanty and Ivanov 2014; Pisanty et al. 2014; Kfir et al. 2015; Milošević 2015b, c; Medišauskas et al. 2015; Milošević 2015a; Fan et al. 2015; Chen et al. 2016). Most of the above-cited papers are devoted to the atomic HHG generated by a bicircular laser field. Molecular HHG induced by a bicircular field was studied in Baykusheva et al. (2016), Mauger et al. (2016), Odžak et al. (2016), Reich and Madsen (2016). It was shown that there is a strong asymmetry in the emission of high harmonics of opposite helicities for the BF_3 molecule.

This asymmetry depends on the molecular orientation and may be used for its determination (Odžak et al. 2016). A general theory of HHG generated by a bicircular laser field on N-fold rotationally symmetric molecules was presented in Reich and Madsen (2016).

There are fewer papers that are devoted to atomic or molecular (H)ATI governed by a bicircular laser field. Above-threshold detachment electron spectra of atomic negative ions generated by a bicircular laser field were presented in Kramo et al. (2007), Hasović et al. (2008). This process is an 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 Odžak et al. (2017). Bicircular (H)ATI of atomic targets was recently analyzed in detail in Mancuso et al. (2015), Hasović et al. (2016), Milošević and Becker (2016), Mancuso et al. (2016). This includes a comparison between experimental data and numerical results based on the improved strong-field approximation (Mancuso et al. 2016). The agreement between theoretical calculations and experimental results is good. More recently, molecular HATI was investigated in Busuladžić et al. (2017). We were able to identify two rotational and two reflection symmetries which are satisfied in ATI of homonuclear diatomic molecules. The two rotational symmetries are valid both for HATI process, i.e., both for the direct and rescattered electrons.

2 Theory

We consider a polyatomic molecule as a system of N atomic (ionic) centers and an electron which can be freed under the influence of the laser field with the electric field vector $\mathbf{E}(t)$ (Hasović and Milošević 2012, 2014; Hasović et al. 2015). We apply the S-matrix theory to the molecular ATI and HATI processes. The atomic system of units ($\hbar = e = m = 4\pi\varepsilon_0 = 1$) is used throughout the paper. We choose the interaction with the laser pulse in dipole approximation and length gauge. The presented approach is an extension of our ATI theory of diatomic molecules (Milošević 2006).

As mentioned previously, a bicircular laser field is a superposition of two coplanar counterrotating fields having the angular frequencies $r\omega$ and $s\omega$, which are integer multiples of the same fundamental frequency $\omega = 2\pi/T$. It is defined by

$$\mathbf{E}(t) = \frac{i}{2}\left(E_1\hat{\mathbf{e}}_+ e^{-ir\omega t} + E_2\hat{\mathbf{e}}_- e^{-is\omega t}\right) + \text{c.c.}, \tag{1}$$

where $\hat{\mathbf{e}}_\pm = (\hat{\mathbf{e}}_x \pm i\hat{\mathbf{e}}_y)/\sqrt{2}$, with $\hat{\mathbf{e}}_x$ and $\hat{\mathbf{e}}_y$ the real unit polarization vectors along the x and y axes, respectively. In (1), E_j and $I_j = E_j^2$ are the electric-field vector amplitude and the intensity of the jth field component with the helicities h_j ($h_1 = 1,\ h_2 = -1$), respectively. Introducing arbitrary phases ϕ_1 and ϕ_2 in the definition of the bicircular field (1), the field components are (Milošević and Becker 2016)

$$\begin{aligned} E_x(t) &= \left[E_1 \sin(r\omega t + \phi_1) + E_2 \sin(s\omega t + \phi_2)\right]/\sqrt{2}, \\ E_y(t) &= \left[-E_1 \cos(r\omega t + \phi_1) + E_2 \cos(s\omega t + \phi_2)\right]/\sqrt{2}. \end{aligned} \tag{2}$$

A change of the phase ϕ_1, for a fixed value of ϕ_2, corresponds to a rotation of the field around the z axis by the angle $\alpha = s\phi_1/(r+s)$.

We now present corresponding matrix elements within the S-matrix theory of (H)ATI process on arbitrary polyatomic molecules. We first present the matrix element describing the

ATI process. As it was the case for diatomic molecules (Milošević 2006), the differential ionization rate for an infinitely long T-periodic laser field, is

$$w_{\mathrm{fi}}^{(0)}(n) = 2\pi p_{\mathrm{f}} \left| T_{\mathrm{fi}}^{(0)}(n) \right|^2, \tag{3}$$

where the $T^{(0)}$-matrix element is defined by

$$T_{\mathrm{fi}}^{(0)}(n) = \int_0^T \frac{dt}{T} \mathcal{T}_{\mathrm{fi}}^{(0)}(t) e^{in\omega t}, \tag{4}$$

with

$$\mathcal{T}_{\mathrm{fi}}^{(0)}(t) = \mathcal{F}_{\mathrm{fi}}^{(0)}(t) e^{i\mathcal{U}_{\mathrm{f}}(t)} = \sum_{n=-\infty}^{\infty} T_{\mathrm{fi}}^{(0)}(n) e^{-in\omega t}, \tag{5}$$

$\mathcal{U}_{\mathrm{f}}(t) = \mathbf{p}_{\mathrm{f}} \cdot \boldsymbol{\alpha}(t) + \mathcal{U}(t)$, $\mathcal{U}(t) = \int^t d\tau \mathbf{A}^2(\tau)/2 - U_{\mathrm{p}} t$, $U_{\mathrm{p}} = \int_0^T dt \mathbf{A}^2(t)/(2T)$ the ponderomotive energy of the electron in the laser field, and $\boldsymbol{\alpha}(t) = \int^t d\tau \mathbf{A}(\tau)$. The superscript (0) on the left-hand side of Eq. (4) denotes the matrix element accounting for contribution of direct electrons to the corresponding spectra. For the neutral polyatomic molecules, our final result within dressed length gauge is

$$\mathcal{F}_{\mathrm{fi}}^{(0)}(t) = \sum_{j=1}^{N} e^{-i\mathbf{p}_{\mathrm{f}} \cdot \boldsymbol{\rho}_j} \sum_a c_{ja} \langle \mathbf{p}_{\mathrm{f}} + \mathbf{A}(t) | \mathbf{E}(t) \cdot \mathbf{r} | \psi_a \rangle, \tag{6}$$

where $\boldsymbol{\rho}_j$ is defined by Eq. (24) in Hasović and Milošević (2012). More information about the presented theoretical approach can be found in Milošević (2006).

Using the S-matrix formalism, we were able to develop a theory which also takes into account the electron rescattering off the molecular centers, and which is often called Improved Strong-Field Approximation (ISFA). For the corresponding differential ionization rate with absorption of n photons from the laser field, we get

$$w_{\mathrm{fi}}^{(\mathrm{ISFA})}(n) = 2\pi p_{\mathrm{f}} |T_{\mathrm{fi}}^{(0)}(n) + T_{\mathrm{fi}}^{(1)}(n)|^2, \tag{7}$$

where

$$T_{\mathrm{fi}}^{(1)}(n) = \int_0^T \frac{dt}{T} \mathcal{F}_{\mathrm{fi}}^{(1)}(t) e^{i[U_{\mathrm{f}}(t) + n\omega t]}, \tag{8}$$

is the transition amplitude corresponding to the contributions of rescattered electrons. The subintegral function in Eq. (8) is given by

$$\begin{aligned} \mathcal{F}_{\mathrm{fi}}^{(1)}(t) = -ie^{-iS_{\mathbf{k}_{\mathrm{st}}}(t)} \int_0^{\infty} d\tau \left(\frac{2\pi}{i\tau} \right)^{3/2} e^{i\left[S_{\mathbf{k}_{\mathrm{st}}}(t') - \Delta E(\{\mathbf{R}_0\})\tau\right]} \\ \times \sum_{j=1}^{N} e^{i\mathbf{K}_{\mathrm{st}} \cdot \boldsymbol{\rho}_j(\{\mathbf{R}_0\})} V_{\mathrm{e}\mathbf{K}_{\mathrm{st}}}^{j} \sum_{l=1}^{N} e^{-i\mathbf{k}_{\mathrm{st}} \cdot \boldsymbol{\rho}_l} \sum_a c_{la} \langle \mathbf{k}_{\mathrm{st}} + \mathbf{A}(t') | \mathbf{E}(t') \cdot \mathbf{r} | \psi_a \rangle, \end{aligned} \tag{9}$$

with $t' = t - \tau$, $\mathbf{K}_{\mathrm{st}} = \mathbf{k}_{\mathrm{st}} - \mathbf{p}_f$, $\mathbf{k}_{\mathrm{st}} = -\int_{t'}^{t} dt'' \mathbf{A}(t'')/\tau$ the stationary electron momentum, and $S_{\mathbf{k}}(t) = \int^t dt' [\mathbf{k} + \mathbf{A}(t')]^2/2$. The energy-conservation condition for (H)ATI process is

given by the relation $\mathbf{p}_f^2/2 = n\omega - \Delta E - U_p$, where $\Delta E(\{\mathbf{R}_0\})$ is the ionization energy which includes vibrational degrees of freedom. $V^j_{e\mathbf{K}_{st}}$ is the Fourier transform of the rescattering potential on the j center. It is clear from Eq. (7) that the T-matrix element in the ISFA, $T_{fi}^{ISFA}(n) = T_{fi}^{(0)}(n) + T_{fi}^{(1)}(n)$, is a coherent sum of the direct ATI amplitude $T_{fi}^{(0)}(n)$ and its correction due to rescattering, the amplitude $T_{fi}^{(1)}(n)$. The relative factor between these two amplitudes is clearly defined by Eqs. (4)–(9). The differential ionization rate of the HATI process includes both the direct and rescattered electrons and is given by Eq. (7).

We assume that the ionization process occurs from the highest occupied molecular orbital (HOMO) of the molecule, which is written as a linear combination of atomic orbitals (Hasović and Milošević 2012). The calculation of the HOMO was done using the GAMESS quantum chemistry package within the cc-pVTZ basis set (Schmidt et al. 1993). Finally, in order to obtain complete spectra which include both contributions (i.e., the direct and rescattered electrons), a coherent sum of these contributions must be taken into account (Hasović et al. 2015).

3 Numerical results

We are going to demonstrate the application of the presented theory on a nonlinear triatomic planar molecule. More precisely, we consider the HATI process of the ozone molecule. Ozone is a triatomic molecule consisting of the three oxygen atoms with the internuclear distance of 1.278Å and the O–O–O angle of 116.8°. The structure of O_3 molecule belongs to the C_{2v} point group and its highest occupied molecular orbital (HOMO) has the $^1A_1$ symmetry. The vertical ionization energy of the ozone molecule is $I_p = 12.73$ eV. We suppose that the ionization process occurs from the HOMO of the molecule. As mentioned at the end of the previous section, the calculation of the HOMO was done using the GAMESS quantum chemistry package (Schmidt et al. 1993) within the cc-pVTZ basis set, which includes four s, three p, two d, and one f atomic orbital. The so obtained HOMO wave function of the O_3 molecule is presented in the xz plane in the upper left panel of Fig. 1. Geometry optimization was done within the RHF level of calculation (Hasović and Milošević 2012).

We consider the ionization process by the $\omega - 2\omega$ and $\omega - 3\omega$ bicircular laser fields. Normalized electric-field vector $\mathbf{E}(t)$ (black lines) and the corresponding normalized vector potential $\mathbf{A}(t)$ (red lines) for the $\omega - 2\omega$ ($\omega - 3\omega$) field, with equal intensities of the field components, are presented in the upper (lower) panels of Fig. 1. The bicircular field $\mathbf{E}(t)$ and the vector potential $\mathbf{A}(t)$ obey the following dynamical symmetry: The rotation by the angle $\alpha_j = -2\pi jr/(r+s)$ about the z axis is equivalent to the translation in time by $\tau_j = jT/(r+s)$, i.e.,

$$R_z(\alpha_j)\mathbf{E}(t) = \mathbf{E}(t + \tau_j), \tag{10}$$

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 $\pm\sin\alpha_j$. It was shown that the spectra of photons generated in HHG and laser-assisted recombination (LAR), as well as the spectra of (H)ATI electrons, obtained by placing atoms in a bicircular field, exhibit the same type of the symmetry (Kramo et al. 2007; Hasović et al. 2008, 2016; Milošević 2015a; Odžak and Milošević 2015). Electron-ion recombination is a process, which can occur in the absence of the laser field. In this case, the process is characterized by energy transfer from the free electron to a photon. If the process occurs in a laser field, the incident electron may

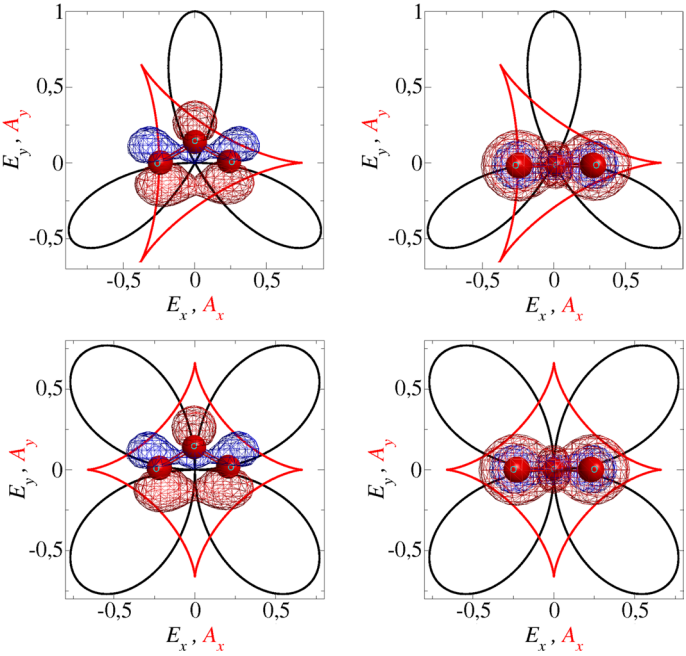

Fig. 1 (Color online) Polar diagram of the electric-field vector $\mathbf{E}(t)$, $0 \leq t \leq T$ (black lines) and the corresponding vector potential $\mathbf{A}(t)$ (red lines) of the $\omega - 2\omega$ (upper panels) and $\omega - 3\omega$ (lower panels) bicircular fields having equal intensities of the field components. The equilibrium geometry of the ozone molecule and the corresponding HOMO orbital is shown for the parallel (left panels) and perpendicular (right panels) orientations of the molecular plane with respect to the polarization plane of the laser field

exchange energy with the laser field before it recombines with the target ion. This may result in the emission of a high-energy photon. For more information about laser-assisted phenomena governed by a bicircular laser field one can see recently published papers (Odžak and Milošević 2015; Čerkić et al. 2017; Korajac et al. 2017).

We will now examine if there is any type of rotational symmetry in the spectra of HATI electrons when planar molecules are ionized by different types of bicircular field. We consider two orientations of the molecular plane with respect to the plane of the field-polarization vector, i.e., xy plane. We first calculate the HATI spectra (using ISFA) for the case of parallel orientation of the molecular plane with respect to the plane of the field-polarization vector (left panels of Fig. 1) and then we rotate the molecular plane by 90 degrees and calculate the HATI spectra for perpendicular orientation (right panels of Fig. 1). The calculated momentum distributions of the HATI electrons for these cases are presented in the (p_x, p_y) plane in Fig. 2. The corresponding bicircular field has equal component intensities $I_1 = I_2 = 10^{14}$ W cm^{-2} and the fundamental wavelength of 800 nm.

The HATI spectrum of the ozone molecule, oriented parallel to the plane of the $\omega - 2\omega$ bicircular field, is presented in the panel (a) of Fig. 2. The spectrum consists of three broad peaks at the emission angles $\theta \approx 60°, 180°$ and 300°. These directions are opposite to the direction of the vector potential at the ionization time (i.e., peaked values of the vector potential), as one can see from the left upper panel of Fig. 1. Although the field obeys the rotational symmetry by an angle of 120° about the z axis, the presented

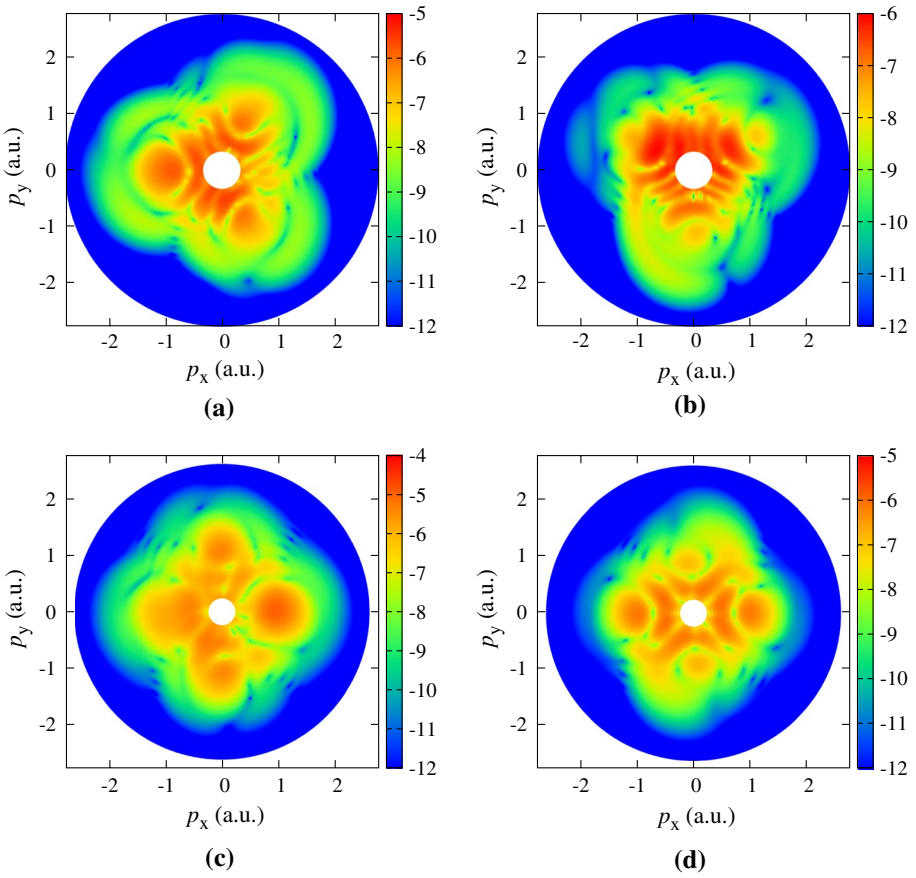

Fig. 2 (Color online) Logarithm of the differential ionization rate (HATI spectra) of the O_3 molecule presented in the momentum plane for ionization by the $\omega - 2\omega$ panels **a** and **b** and $\omega - 3\omega$ panels **c** and **d** laser fields having equal field component intensities $I_1 = I_2 = 10^{14}$ W cm^{-2} and the fundamental wavelength of 800 nm. The plane of the ozone molecule is parallel panels **a** and **c** or perpendicular panels **b** and **d** to the polarization plane of the laser field

spectrum is not rotational invariant because the ozone molecule, oriented parallel to the polarization plane of the laser field, does not posses any type of rotational symmetry.

A similar conclusion holds for the spectra presented in panels (b) and (c). The spectrum of the ozone molecule placed perpendicular to the $\omega - 2\omega$ field plane [panel (b)] looks similar to the spectrum of the parallel orientation [panel (a)]. In this case, the ozone molecule obeys a rotational symmetry about the z axis by an angle of 180°, but it is still different in comparison to the 120° rotational symmetry of the $\omega - 2\omega$ field and, therefore, the spectrum is not rotational symmetric. The HATI spectrum of the ozone molecule which is oriented parallel to the $\omega - 3\omega$ field is shown in the panel (c). The spectrum consists of four broad peaks at the emission angles $\theta \approx 0°, 90°, 180°$ and 270°, which are opposite to the direction of the vector potential at the ionization time (red line in the lower left panel of Fig. 1). The spectrum also does not posses any type of rotational symmetry because the molecule, oriented parallel to the field plane, is not rotational invariant about the z axis.

Finally, we present the HATI spectrum of the ozone molecule oriented perpendicular to the $\omega - 3\omega$ laser field plane [panel (d)]. It is obvious that this spectrum is rotational symmetric about the z axis by the angle of 180° because the laser field, as well as the ozone molecule, obeys the same type of rotational symmetry. The symmetry of the HATI spectra can be used as an indicator of the orientation of the molecule or molecular plane with respect to the laser field polarization plane. We expect that molecules like BH_3 or BF_3 will obey the 120° rotational symmetry if placed parallel to the $\omega - 2\omega$ bicircular field and the HATI spectra can be used as a measure of the molecular orientation.

4 Conclusions

We applied the the improved molecular strong-field approximation theory to investigate the HATI process of the ozone molecule in the presence of the $\omega - 2\omega$ and $\omega - 3\omega$ bicircular laser fields. The molecular plane was oriented parallel or perpendicular to the polarization plane of the laser field. It was shown that, in contrast to the atoms, the HATI spectra of polyatomic molecules do not obey rotational symmetry in general. For the $\omega - 2\omega$ case, the HATI spectrum shows three broad peaks, as expected, but rotational symmetry is absent. For the $\omega - 3\omega$ bicircular field, the HATI spectrum is rotational symmetric only if the plane of the molecule is perpendicular to the polarization plane of the laser field because, in this case, the molecule possesses the same type of rotational symmetry with respect to the z axis as the field. We propose that the symmetry of the HATI spectra of polyatomic molecules can be used as an indicator of the orientation of the molecule.

Acknowledgements Support by the Federal Ministry of Education and Science, Bosnia and Herzegovina, is gratefully acknowledged. This work was also supported in part by the Deutsche Forschungsgemeinschaft (DFG) within the Priority Programme Quantum Dynamics in Tailored Intense Fields (QUTIF).

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