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Molecular above-threshold ionization spectra as an evidence of the three-point interference of electron wave packets

Elvedin Hasović¹, Dejan B Milošević^{1,2,3}, Azra Gazibegović-Busuladžić¹, Aner Čerkić¹ and Mustafa Busuladžić⁴

1 Faculty of Science, University of Sarajevo, Zmaja od Bosne 35, 71000 Sarajevo, Bosnia and Herzegovina

2 Max-Born-Institut, Max-Born-Str. 2a, 12489 Berlin, Germany

3 Academy of Sciences and Arts of Bosnia and Herzegovina, Bistrik 7, 71000 Sarajevo, Bosnia and Herzegovina

4 Faculty of Medicine, University of Sarajevo, Čekaluša 90, 71000 Sarajevo, Bosnia and Herzegovina

E-mail: mustafabusuladzic@bih.net.ba

Abstract. We consider high-order above-threshold ionization (HATI) of polyatomic molecules ionized by a strong linearly polarized laser field. Improved molecular strong-field approximation by which the HATI process on polyatomic molecular species can be described is developed. Using this theory we calculate photoelectron angular-energy spectra for different triatomic molecules. Special attention is devoted to the minima that are observed in the calculated high-energy electron spectra of the ozone and carbon dioxide molecules. A key difference between these minima and minima that are observed in the corresponding spectra of diatomic molecules are presented.

1. Introduction

During laser-atom or laser-molecule interaction a few nonlinear phenomena of higher order can occur (see review articles [1, 2, 3, 4, 5] and references therein). The most important among them are the high-order above-threshold ionization (HATI) and high-order harmonic generation (HHG). These processes and key features of obtained photon and electron spectra can be explained using the so-called three-step model [6, 7]. In the first step of the HATI and HHG, quantum mechanical system (atom or molecule) absorbs more photons from the strong laser field than is necessary for ionization. Some of these electrons can go directly to detector. This process is called direct above-threshold ionization (ATI) and within it any additional interaction between electron and its parent ion is neglected. Due to the influence of the laser field, the ionized electron may also return to the parent molecular ion (second step) and elastically scatter off it (third step), before reaching the detector. In this process, the electron can absorb many more photons from the laser field than in the direct ATI process. This process was named high-order ATI (HATI). In the third step of the HHG process, the electron recombines with the parent ion and one high-energy photon is emitted. The energy spectra of atomic/molecular HATI and atomic/molecular 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.

Above-mentioned molecular laser-induced processes have received much attention in the last few years because of their ability to reveal some features of the internal structure of the molecule under investigation [8, 9]. More precisely, by analyzing their spectra one can extract information about molecular structure. In fact, even direct ionization (direct ATI) without rescattering reveals the initial symmetry of the molecular system. For example, the O_2 molecule shows a suppression in the low-energy electron spectra due to its π_g symmetry, while the N_2 molecule, having σ_g symmetry, does not show such a suppression [10, 11]. A more recent experimental and theoretical study of ATI on different polyatomic molecules was published in [12].

Here we are interested in the HATI process. If rescattering is included, more scenarios are possible than in the ATI process [13]. As molecules are multi-center systems, the ionization as well as rescattering can happen at different centers, causing interference structures in the electron spectrum. In the case of the HATI of arbitrary diatomic molecular species, there are four rescattering contributions to the ionization rate. In other words, our matrix element related to rescattering process in the case of diatomic molecules can be decomposed into four terms, each term describing one of the four possible scenarios [14]. All these contributions interfere in a complicated manner and can produce a complicated spectrum, which depends on the laser parameters, electron emission angle, and the molecular orientation. The most noticeable feature of the calculated HATI spectra is the existence of pronounced minima that are absent in atomic case. We have found an analytical formula for the minima position regardless of symmetry of considered molecules. It is given by the equation [14, 15]

$$R_0 |p_f \cos \theta - k_{st} \cos \theta_L| = (2m + 1)\pi, \quad (1)$$

where m is integer (i.e. $m = 0, 1, 2, \dots$), θ_L is the angle between the laser polarization vector and the internuclear axis R_0 , θ is the angle between the final momentum of the detected electron that is denoted by p_f and the internuclear axis R_0 , while k_{st} is the stationary intermediate electron momentum between the ionization and the rescattering (it is in the direction of the laser electric field vector). It is important to stress that the previously mentioned two-source two-rescattering-centers interference has been observed in experiments with unaligned molecules [16, 17].

Our next step is to calculate and analyze electronic spectra of polyatomic molecules. We will introduce a theory of strong-field ionization of arbitrary polyatomic molecules. In this sense, we follow the approach presented in [18]. The theory is so formulated that it takes into account a possible rescattering of the emitted electrons on the various atomic centers of polyatomic molecule. Then, we are going to present HATI spectra of some polyatomic molecules, such as O_3 and CO_2 , and to give a physical interpretation of the observed minima. Finally, the conditions for the destructive interference of all contributions are provided and the key differences between the HATI spectra of diatomic and polyatomic molecules are discussed. We use atomic system of units.

2. Improved molecular strong-field approximation theory for polyatomic molecules

We have developed a theory describing above-threshold ionization of arbitrary polyatomic molecules. A polyatomic molecule is modeled by an $(N + 1)$ -particle system, which consists of N heavy atomic (ionic) centers and an electron [19]. After the separation of the center-of-mass coordinate, the dynamics of this system is reduced to the relative electronic and nuclear coordinates. Two forms of the molecular strong-field approximation (MSFA), one with the field-free and the other with the field-dressed initial molecular bound state, are derived. The relevant time-dependent transition matrix element of the modified MSFA in the length gauge, for neutral

polyatomic molecules can be written as [19]

$$\mathcal{F}_{\text{fi,neut}}^{q\text{ML}}(t) = \sum_{j=1}^N g_q(\boldsymbol{\rho}_j, t) e^{-i[\mathbf{p}_f + \mathbf{A}(t)] \cdot \boldsymbol{\rho}_j} \sum_a c_{ja} \langle \mathbf{p}_f + \mathbf{A}(t) | \mathbf{E}(t) \cdot \mathbf{r} | \psi_a \rangle, \quad (2)$$

where \mathbf{p}_f is the final momentum of the detected electron and the factor $g_q(\boldsymbol{\rho}_k, t)$ enables us to treat both the undressed ($q = u$) and the dressed ($q = d$) initial state case. The transition matrix element (2) depends on the coordinates $\boldsymbol{\rho}_k(\{\mathbf{R}\})$ which are given as linear combinations of the relative coordinates $\mathbf{R}_1, \dots, \mathbf{R}_{N-1}$. The connection of the coordinates $\boldsymbol{\rho}_k(\{\mathbf{R}\})$ and the position vectors \mathbf{r}_j of atoms that are constituent of considered polyatomic molecules can be written as [19]

$$\boldsymbol{\rho}_j = \mathbf{r}_j - \mathbf{R}_{\text{c.m.}} + \frac{m_e}{M} \mathbf{R}_N, \quad (3)$$

where m_e is the electron mass and M is the mass of all atoms. It can be shown that $\boldsymbol{\rho}_j \approx \mathbf{r}_j$ [19]. The theory presented above was able to describe direct electrons only – electrons that leave the laser field and are collected on the detector right after the ionization.

In order to describe high-energy electrons contribution to the spectrum, we have generalized the above-mentioned theory so that it includes an additional interaction of the ionized electron with the molecular centers [20]. We obtained two terms – the first term describes the direct ATI (2), while the second one corresponds to the rescattered electrons, and, for the dressed case, has the form

$$\begin{aligned} \mathcal{F}_{\text{fi,d}}^{(1)}(t) &= i e^{-i S_{\mathbf{k}_{\text{st}}}(t)} \int_0^\infty d\tau \left(\frac{2\pi}{i\tau} \right)^{3/2} e^{i[S_{\mathbf{k}_{\text{st}}}(t') - \Delta E(\{\mathbf{R}_0\})\tau]} \\ &\times \sum_{j=1}^N e^{i\mathbf{K}_{\text{st}} \cdot \boldsymbol{\rho}_j(\{\mathbf{R}_0\})} V_{e\mathbf{K}_{\text{st}}}^j \sum_{l=1}^N e^{-i\mathbf{k}_{\text{st}} \cdot \boldsymbol{\rho}_l} \sum_a c_{la} \langle \mathbf{k}_{\text{st}} + \mathbf{A}(t') | \mathbf{E}(t') \cdot \mathbf{r} | \psi_a \rangle, \end{aligned} \quad (4)$$

where the stationary electron momentum between the ionization and the rescattering is

$$\mathbf{k}_{\text{st}} = -\frac{1}{\tau} \int_{t'}^t dt'' \mathbf{A}(t'') = \frac{1}{\tau} [\boldsymbol{\alpha}(t') - \boldsymbol{\alpha}(t)], \quad (5)$$

with $\boldsymbol{\alpha}(t) = \int^t dt'' \mathbf{A}(t'')$. The electron-molecular ion scattering potential, according to the independent atom model [21], is presented as the sum of the potentials for scattering off the individual atomic centers of the considered molecule. Also, we have $\mathbf{K}_{\text{st}} = \mathbf{k}_{\text{st}} - \mathbf{p}_f$, while $V_{e\mathbf{K}_{\text{st}}}^j$ is the Fourier transform of the rescattering potential at the atomic (ionic) center j . The semiclassical action is denoted by $S_{\mathbf{k}_{\text{st}}}(t) = \int^t dt'' [\mathbf{k}_{\text{st}} + \mathbf{A}(t'')]^2/2$. Finally, $\sum_a c_{la} |\psi_a^{(0)}\rangle$ is the ground-state electronic wave function, presented as a linear combination of atomic orbitals. The calculation of the highest occupied molecular orbital (HOMO) was done using the GAMESS quantum chemistry package within the cc-pVTZ basis set.

The differential ionization rate with absorption of n photons from the laser field having the angular frequency $\omega = 2\pi/T$ is given by

$$w_{\text{fi},q}^{(1)}(n) = 2\pi p_f |T_{\text{fi},q}^{(1)}(n)|^2, \quad (6)$$

where $T_{\text{fi},q}^{(1)}(n)$ is the Fourier transform of the matrix element (4) multiplied by the corresponding phase factor [20].

3. Numerical results

Now we are going to demonstrate an application of the presented theory on a simple example. All calculations are performed with the field-dressed initial molecular bound state, i.e. using (4). Let us consider the HATI process for both carbon dioxide and ozone molecule. Carbon dioxide is a linear triatomic molecule with carbon atom placed in the center, and oxygen atoms placed symmetrically on either side of the center. Due to the symmetry, the center of coordinate system is placed at the carbon atom. The distance between the carbon atom and either oxygen atom is 1.1621 Å. The HOMO of the CO₂ molecule is characterized by $1\pi_g$ symmetry, and its vertical ionization energy is $I_p = 13.777$ eV.

Ozone is a planar triatomic molecule consisting of three oxygen atoms with the internuclear distance of 1.278 Å. The O-O-O angle is 116.8°. Therefore, this molecule is more complex than the CO₂ molecule. The structure of the O₃ molecule belongs to the C_{2v} point group and its HOMO has 1A_1 symmetry. The vertical ionization energy of the ozone molecule is $I_p = 12.73$ eV. For both molecules we assume that the ionization process happens from the HOMO. According

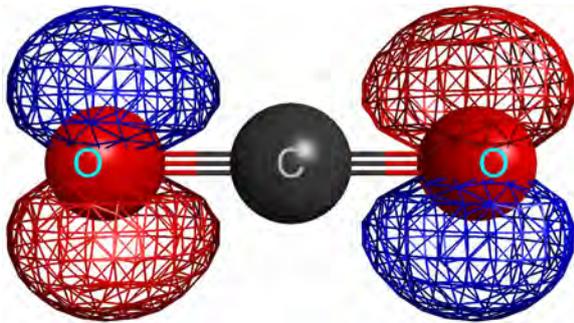


Figure 1. Coordinate-space wave function for the HOMO of the CO₂ molecule, presented in the xz plane.

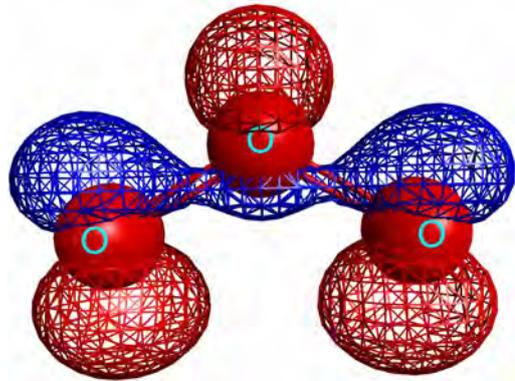


Figure 2. Coordinate-space wave function for the HOMO of the O₃ molecule, presented in the xz plane.

to this, the initial electronic state is the corresponding HOMO, written as a linear combination of atomic orbitals. As previously mentioned, the calculation of the HOMO was done using the GAMESS quantum chemistry package within the cc-pVTZ basis set. Such CO₂ and O₃ molecule HOMO wave functions are presented in figures 1 and 2 in the xz plane.

Geometry of the considered process on triatomic molecules is described in the next part of this section. The electron emission angle is defined as the angle between the laser polarization vector and the vector of the final momentum of the emitted electron. For diatomic molecules the internuclear axis is usually chosen as the reference axis (for example, the z axis) and the axial symmetry of the molecule is used to simplify the calculations. For nonlinear polyatomic molecules the situation is more complicated. It is the case for the planar O₃ molecule. Because of the absence of the axial or spherical symmetry we have to consider any possible orientation of the laser polarization vector with respect to the polyatomic molecule. Therefore, for a linearly polarized laser field we define the field vector as

$$\mathbf{E}(t) = E_0 \sin(\omega t) \hat{\mathbf{e}}_L, \quad (7)$$

where E_0 is the electric field vector amplitude and $\hat{\mathbf{e}}_L = e_x \hat{\mathbf{i}} + e_y \hat{\mathbf{j}} + e_z \hat{\mathbf{k}}$ is the unit laser-field vector. In practice, we determine the direction of this vector by the azimuthal angle θ_L and the polar angle φ_L , so that, in the Cartesian coordinates, it has the following form

$$\hat{\mathbf{e}}_L = \sin \theta_L \cos \varphi_L \hat{\mathbf{i}} + \sin \theta_L \sin \varphi_L \hat{\mathbf{j}} + \cos \theta_L \hat{\mathbf{k}}. \quad (8)$$

The emitted electrons can be detected in any direction too. The final momentum vector is defined by its intensity p_f , azimuthal angle θ_f and polar angle φ_f . We can write $\mathbf{p}_f = p_f \hat{\mathbf{e}}_f$, where the unit vector of the electron final momentum is defined as

$$\hat{\mathbf{e}}_f = \sin \theta_f \cos \varphi_f \hat{\mathbf{i}} + \sin \theta_f \sin \varphi_f \hat{\mathbf{j}} + \cos \theta_f \hat{\mathbf{k}} \quad (9)$$

For more details about the geometry of the process one can see [19, 20]. Now, we present high-

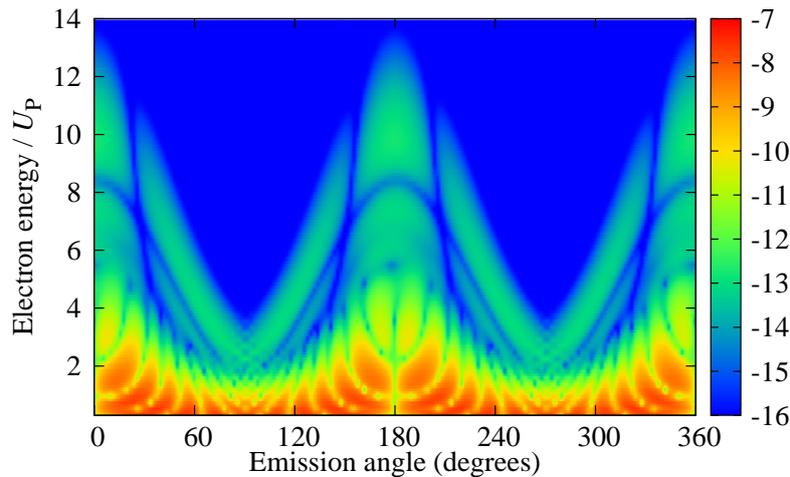


Figure 3. Logarithm of the differential ionization probability of the carbon dioxide molecule as a function of the electron kinetic energy and emission angle. The laser-field intensity is $1.2 \cdot 10^{14} \text{ W/cm}^2$, while the wavelength is 800 nm. The field polarization vector is placed in the xz plane with $\theta_L = 1^\circ$.

energy part of the electron spectra for the both considered molecules. In figure 3, the logarithm of the differential ionization probability of the carbon dioxide molecule as a function of the final electron kinetic energy $E_{\mathbf{p}_f}$ and azimuthal angle θ_f for linearly polarized laser field of the intensity $1.2 \times 10^{14} \text{ W/cm}^2$ and the wavelength 800 nm is presented. The laser-field polarization vector $\hat{\mathbf{e}}_L$ is placed in the xz plane with $\varphi_L = 0^\circ$ and $\theta_L = 1^\circ$. The most interesting feature of the ATI angular distribution of CO_2 molecule are distinct and sharp minima in the high-energy part of the spectrum. Similar minima are observed in the ATI angular distributions of diatomic molecules and are explained by the destructive interference of the T -matrix partial contributions [15]. The observed suppression in the high-energy part of the photoelectron spectrum of diatomic molecules can be fitted very well by the curve given by equation (1). It can be shown that the observed minima in the case of linear centrosymmetric triatomic molecule, like CO_2 molecule, can be fitted with the curve given by relation

$$x_1 |p_f \cos \theta_f - k_{\text{st}} \cos \theta_L| = 2 \left(\frac{m}{3} + 1 \right) \pi, \quad (10)$$

in analogy to the minima in diatomic molecule case [see equation (1)]. For the laser and molecular parameters we considered in the present paper, only the lowest value $m = 0$ contributes. If we increase the internuclear distance x_1 two times, the minima of the second order will appear

($m = 1$). So, in the case of linear triatomic molecules, minima have the same structure as in the diatomic molecule case. A very similar results were obtained for the HATI of triatomic molecules using the frequency-domain theory [22]. Finally, we conclude that the observed minima have their origin in the three center (three slit) interference, with position given by relation (10).

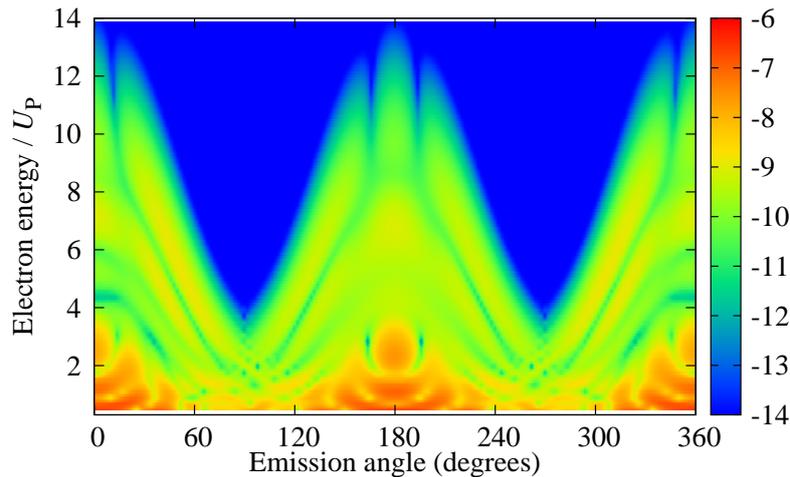


Figure 4. Logarithm of the differential ionization probability of the ozone molecule as a function of the electron kinetic energy and emission angle. The laser-field intensity is $1.2 \cdot 10^{14}$ W/cm², while the wavelength is 800 nm. The field polarization vector is directed along the z axis, i.e., $\theta_L = 0^\circ$.

The logarithm of the differential ionization probability of the ozone molecule as a function of the final electron kinetic energy E_{p_f} and azimuthal angle θ_f is presented in figure 4. Laser field intensity, wavelength and polarization are the same as in the previous case. The laser-field polarization vector \mathbf{e}_L is oriented along the z axis, i.e. $\varphi_L = 0^\circ$ and $\theta_L = 0^\circ$. The ozone molecule lies in the xz plane. The angular distribution, presented here, has similar features as angular distributions of diatomic molecules or atoms. The high-energy plateau is mostly extended in the laser-field direction, i.e., for angles $\theta_f = 0^\circ$ and $\theta_f = 180^\circ$, while the high-energy plateau disappears for electrons emitted in direction perpendicular to the laser-field polarization vector. The most interesting feature of the ATI angular distribution of ozone molecule are distinct minima in the high-energy part of the spectrum, detected for azimuthal angles $\theta \approx 10^\circ, 170^\circ, 190^\circ$, and 350° .

It is interesting to check what happen to the O_3 molecule spectrum and the corresponding minima if the internuclear angle is chosen to be $\alpha = 180^\circ$, i.e. if we assume that the O_3 molecule is linear. Sharp and clear minima appear in the spectrum, minima of the kind that can be fitted with curve given with relation (10), just as in diatomic case. This confirms our previous finding for the CO_2 molecule and the corresponding minima.

Let us go back to the proper ozone molecule, that is more complex, planar molecule. We will consider what mechanism causes the observed minima in the HATI spectrum of the O_3 molecule that are clearly visible on figure 4. Generally, for triatomic molecules there are twelve partial contributions: three partial contributions of the direct amplitude and nine partial contributions of the amplitude which describes rescattered electrons. It can be shown that for triatomic

molecules the partial contributions of the electrons rescattered on different atomic centers, which contribute to the high-energy part of the spectrum, interfere destructively. This is also the case for diatomic molecules. Then, the differential ionization probability is proportional to

$$w_{\text{fi}}(n) \propto \left| \sum_{j=1}^N e^{i\mathbf{K}_{\text{st}} \cdot \boldsymbol{\rho}_j(\{\mathbf{R}_0\})} V_{e\mathbf{K}_{\text{st}}}^j \right|^2. \quad (11)$$

For the ozone molecule the electron scattering amplitudes $V_{e\mathbf{K}_{\text{st}}}^j$ on the three atomic centers are equal, so that the differential ionization probability becomes

$$w_{\text{fi}}(n) \propto \left| e^{-iK_x x_1 + iK_z z_1} + e^{iK_z z_2} + e^{iK_x x_1 + iK_z z_1} \right|^2, \quad (12)$$

where $x_1 = 1.0304148692 \text{ \AA}$, $z_1 = -0.2012853882 \text{ \AA}$, and $z_2 = 0.4025707765 \text{ \AA}$ determine the equilibrium position of the oxygen atoms. If we put $z_2 = z_1 + \Delta z$, it can be shown that the differential ionization probability is proportional to

$$w_{\text{fi}}(n) \propto \left| e^{-iK_x x_1} + e^{iK_z \Delta z} + e^{iK_x x_1} \right|^2. \quad (13)$$

This proportion reduces to

$$w_{\text{fi}}(n) \propto (3 + 4 \cos \alpha \cos \beta + 2 \cos 2\alpha), \quad (14)$$

where $\alpha = K_x x_1$ and $\beta = K_z \Delta z$. If the function $f(\alpha, \beta) = 3 + 4 \cos \alpha \cos \beta + 2 \cos 2\alpha$ is equal to zero, the differential ionization probability will have the absolute minimum. The equation $3 + 4 \cos \alpha \cos \beta + 2 \cos 2\alpha = 0$ can be transformed to

$$\cos^2 \alpha + \cos \beta \cos \alpha + \frac{1}{4} = 0. \quad (15)$$

The solution of this equation can be written as

$$\cos \alpha_{1,2} = \frac{-\cos \beta \pm \sqrt{\cos^2 \beta - 1}}{2}. \quad (16)$$

The solutions of the previous quadratic equation will be real only if $\cos^2 \beta = 1$, i.e. $\cos \beta = \pm 1$, which leads to $\beta = k\pi$, $k = 0, 1, 2, \dots$. For even k the values of α become $\alpha = \frac{2m\pi}{3} + 2l\pi$, $m = 1, 2$ and $l = 0, 1, 2, \dots$, while if k is odd we have $\alpha = \pm \frac{\pi}{3} + 2l\pi$, $l = 0, 1, 2, \dots$. So, the differential ionization probability becomes zero if the next two conditions are fulfilled simultaneously

$$\begin{aligned} |k_{\text{st}} \cos \theta_{\text{L}} - p_{\text{f}} \cos \theta_{\text{f}}| \Delta z &= k\pi, \\ |k_{\text{st}} \sin \theta_{\text{L}} - p_{\text{f}} \sin \theta_{\text{f}}| x_1 &= \begin{cases} \frac{2m\pi}{3} + 2l\pi & \text{for even } k, \\ \pm \frac{\pi}{3} + 2l\pi & \text{for odd } k. \end{cases} \end{aligned} \quad (17)$$

From the previous two conditions we can find the values of electron kinetic energy and emission angles for which the ionization probability becomes zero. In the special case where the angle $\theta_{\text{L}} = 0^\circ$ the destructive interference conditions become

$$\begin{aligned} p_{\text{f}} \cos \theta_{\text{f}} = p_{\text{fz}} &= k_{\text{st}} - \frac{k\pi}{\Delta z}, \\ p_{\text{f}} \sin \theta_{\text{f}} = p_{\text{fx}} &= \frac{1}{x_1} \begin{cases} \frac{2m\pi}{3} + 2l\pi & \text{for even } k, \\ \pm \frac{\pi}{3} + 2l\pi & \text{for odd } k, \end{cases} \end{aligned} \quad (18)$$

with the electron kinetic energy and emission angle given by $E_{\mathbf{p}_f} = \frac{1}{2}(p_{fx}^2 + p_{fz}^2)$ and $\theta_f = \arctan(p_{fz}/p_{fx})$, respectively. As the previous two conditions have to be fulfilled at the same time, we cannot find an analytic function $E_{\mathbf{p}_f} = f(\theta_f)$, which determines the positions of the interference minima in the $(E_{\mathbf{p}_f}, \theta_f)$ plane, the way we could in the case of diatomic molecules and triatomic linear molecules. The absolute interference minima for the ozone molecule will appear only for certain values of the electron kinetic energy and certain values of the emission angles, and will be represented by a single point in the $(E_{\mathbf{p}_f}, \theta_f)$ plane. Therefore, the minima in the high-energy part of the angle-resolved electron emission spectrum of the ozone molecule, presented in the figure 4 cannot be explained as the absolute destructive interference minima. However, it does not mean that the interference has nothing to do with the appearance of the minima. If one plots the function $f(\alpha, \beta) = 3 + 4 \cos \alpha \cos \beta + 2 \cos 2\alpha$ in the (α, β) plane, as it was done in [20], one can conclude that the function $f(\alpha, \beta)$ has an absolute minimum, equal to zero, for $(\alpha, \beta) = (\pm\pi/3, \pm\pi), (\pm2\pi/3, 0), \dots$, which was confirmed analytically. However, there are relatively wide intervals of values of α and β , around the zero points, for which the function $f(\alpha, \beta)$ is close to zero. By closer investigation of angular distributions of emitted photoelectrons for fixed electron energies we have found out that there are clear minima for all photoelectron energies. One can extract values of the angles θ_f for which these minima occur. The angles $\alpha = K_x x_1$ and $\beta = K_z \Delta z$ corresponding to those values of θ_f are calculated. We concluded that these values are very close to the zero-point values $(\alpha, \beta) = (\pi/3, -\pi)$, and that the value of the function $f(\alpha, \beta)$ is close to zero, i.e. it is more than ten times smaller than its maximum value. Furthermore, by increasing the electron kinetic energy, the value of the function $f(\alpha, \beta)$ decreases, as well as the differential ionization probability. This is in agreement with the results presented in the energy-angular distribution in figure 4, where the minima in the high-energy part of the spectrum become more pronounced with the increase of the electron kinetic energy. Therefore, the minima in the high-energy part of the HATI spectra of the ozone molecule can be interpreted as the destructive interference of the three rescattered electron wave packets. It should be stressed that these minima do not correspond to the absolute interference minima where the coherent sum of the three wave packets completely vanishes.

4. Conclusions

We have developed an improved-molecular-strong-field-approximation-based theory of high-order above-threshold ionization of polyatomic molecules. The theory is formulated to include the rescattering of the emitted electron off the parent molecular ion. We applied this theory to calculate the energy- and angle-resolved HATI spectra of the ozone and carbon dioxide molecules. Obtained spectra exhibit clear minima for certain emission angles in the high-energy part of the spectrum. These minima originate from the three-center interference of the rescattered electron wave packets. We have concluded that the absolute destructive interference minima, that are characteristic of diatomic as well as triatomic linear molecules, do not cause the appearance of the minima in the HATI spectrum of planar polyatomic molecules.

Acknowledgments

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