

Laser-assisted electron–molecular-ion radiative recombination

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We formulate a theory of laser-assisted radiative recombination of electron with diatomic homonuclear ion in a linearly polarized laser field using the S -matrix formalism. We investigate how the multicenter molecular structure modifies the differential power spectra and polarization properties of the emitted x-rays. The emitted spectra exhibit characteristic two-center interference minima whose positions depend on the molecular orientation, internuclear distance, symmetry of the molecular orbital, and the incident electron momentum. We show that the visibility of the interference minima is strongly influenced by the molecular orientation and by the component of the incident electron momentum perpendicular to the laser-field polarization vector. The minima remain comparatively robust against imperfect molecular alignment. In addition, the emitted x-rays remain linearly polarized, while their polarization direction depends on the emitted-photon energy and on the geometry of the recombination process. The obtained results demonstrate that laser-assisted radiative recombination provides direct access to structural and symmetry properties of molecular targets.

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I. INTRODUCTION

When atoms and molecules are exposed to a strong laser field many nonlinear phenomena may occur [1–12]. Particularly interesting are processes leading to the generation of high-frequency radiation, such as high-order-harmonic generation (HHG) [13–16], laser-induced bremsstrahlung [17–22], laser-assisted x-ray scattering [23–25], and laser-assisted radiative recombination [26–28]. These processes are important both for understanding strong-field dynamics and for developing compact coherent sources of ultraviolet and x-ray radiation.

Electron-ion radiative recombination is a process in which a free electron recombines with an ion, forming a bound atomic or molecular state and emitting a photon. When this process occurs in the presence of an external laser field, the electron can absorb or emit laser photons during its motion in the continuum. As a consequence, the emitted radiation spectrum is strongly modified and may extend to substantially higher photon energies than in the case of a field-free recombination. In particular, an electron with momentum $\mathbf{p}$ and kinetic energy $E_p = \mathbf{p}^2/2$ recombines with an ionic target into a bound state $|\psi_B\rangle$ with energy $E_B < 0$, while a photon with frequency $\omega_{\mathbf{k}}$ and polarization vector $\hat{\mathbf{e}}_{\mathbf{k}}$ is emitted. Simultaneously, the electron exchanges n photons with the laser field and the energy-conservation condition is

$n\omega = \omega_{\mathbf{k}} + E_B - E_p - U_p$, where ω denotes the laser frequency and U_p is the electron ponderomotive energy.

Laser-assisted electron recombination with atomic ions assisted by a monochromatic linearly polarized laser field has been studied theoretically in Refs. [26,27,29–35]. The same process assisted by the bichromatic fields was investigated in Refs. [36–40]. The influence of ultrashort laser pulses was analyzed in Refs. [41–45]. In addition, laser-assisted radiative recombination in a cold hydrogen plasma was analyzed in [46], while quantum-optical radiative recombination was investigated in [47]. Experimental investigations of laser-assisted recombination remain comparatively limited because the realization of the process requires synchronized electron, ion, and laser beams. Nevertheless, several important observations have been reported, including phase-dependent recombination [48] and polarization effects in electron- D^+ recombination [49]. Related studies have also considered laser-assisted antihydrogen formation [50–52], bound-state dressing effects [53], and recombination of twisted electrons with bare nuclei [54].

Compared with atomic targets, molecules exhibit a far richer response in strong laser fields due to their multicenter structure and anisotropic electronic orbitals [55–60]. Interference between contributions originating from different atomic centers can strongly affect both the spectral and polarization properties of the emitted radiation. Such effects are well known in molecular HHG, where destructive and constructive interference leads to characteristic minima and strong orientation dependence of the harmonic emission [57,58,61,62]. Similar interference phenomena are expected to play an important role in laser-assisted recombination with molecular ions, particularly for aligned or oriented targets. The differential power spectra and the polarization properties of the emitted x-rays are expected to be highly sensitive to the

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molecular orientation, internuclear distance, and symmetry of the molecular orbital involved in the recombination process. The analysis of these effects is therefore important for understanding how molecular structure is imprinted onto the strong-field-assisted recombination spectra.

In the present paper we formulate a theory of laser-assisted electron–molecular-ion radiative recombination with homonuclear diatomic molecular ions in a linearly polarized laser field. Our approach is based on the S -matrix formalism and is compatible with the molecular strong-field framework developed in Ref. [63]. Molecular orbitals are represented by the linear combination of the atomic orbitals, allowing us to investigate how the multicenter structure of the target modifies the emitted x-ray spectra and polarization properties.

Particular attention is devoted to the two-center interference minima and to their dependence on the molecular orientation, internuclear distance, degree of molecular alignment, and direction of the incident electron momentum. In addition, we study the polarization direction of the emitted x-rays using the Stokes parameters and discuss its connection with the geometry of the recombination process. The paper is organized as follows. In Sec. II we present the theoretical formulation of the process along with the description of the polarization properties of the emitted radiation, and the averaging over molecular alignment. Formal derivation of the S -matrix element and its explicit form for the laser-assisted electron-diatomic homonuclear molecular ion radiative recombination are presented in Appendixes A and B, respectively. Numerical results for the differential power spectra, interference effects, and polarization properties are presented and discussed in Sec. III. Finally, conclusions are given in Sec. IV. Atomic units are used unless otherwise stated.

II. THEORY

In this section we derive the differential power spectrum for laser-assisted radiative recombination with a molecular ion. Formal derivation of the transition amplitude of this process, based on the S -matrix formalism, is presented in Appendix A. The transition matrix element is then evaluated using Volkov continuum states for the incoming electron and Hartree-Fock-Roothaan molecular orbitals for the recombined electron. Finally, we derive analytical conditions for the two-center interference minima and discuss the polarization properties of the emitted x-rays.

A. Differential power spectrum

The S -matrix element for the transition from the initial state Ψ_a (state of the molecular ion and laser-dressed electron) to the final state Ψ_b (bound state of neutral molecule and an x-ray photon) due to interaction with the x-ray field, described by the Hamiltonian $H'_a(\tau)$, with τ the recombination time, is [see Eq. (A6)]

$$S_{fi} = -i \int_{-\infty}^{\infty} d\tau \langle \Psi_b(\tau) | H'_a(\tau) | \Psi_a(\tau) \rangle. \quad (1)$$

For diatomic molecules, the Hamiltonian $H'_a(t)$ is

$$H'_a(t) = -e\mathbf{E}_x(\mathbf{r}_e, t) \cdot \mathbf{r}_e - \sum_{j=A,B} e_j \mathbf{E}_x(\mathbf{r}_j, t) \cdot \mathbf{r}_j, \quad (2)$$

where $e = -1$ and e_j , $j = A, B$, are the electron charge and the charges of the atomic centers A and B , respectively, while $\mathbf{r}_e$ and $\mathbf{r}_j$, $j = A, B$, are the corresponding positions. We consider the x-ray radiation field as the quantized field $\mathbf{E}_x(\mathbf{r}, t) = \mathbf{E}_x^{(+)}(\mathbf{r}, t) + \mathbf{E}_x^{(-)}(\mathbf{r}, t)$ with

$$\begin{aligned} \mathbf{E}_x^{(+)}(\mathbf{r}, t) &= ic_{\mathbf{K}} \hat{\mathbf{e}}_{\mathbf{K}} a_{\mathbf{K}} e^{-i(\omega_{\mathbf{K}} t - \mathbf{K} \cdot \mathbf{r})}, \\ \mathbf{E}_x^{(-)}(\mathbf{r}, t) &= -ic_{\mathbf{K}} \hat{\mathbf{e}}_{\mathbf{K}}^* a_{\mathbf{K}}^\dagger e^{i(\omega_{\mathbf{K}} t - \mathbf{K} \cdot \mathbf{r})}, \end{aligned} \quad (3)$$

where $a_{\mathbf{K}}$ and $a_{\mathbf{K}}^\dagger$ are the photon annihilation and creation operators, respectively, $\hat{\mathbf{e}}_{\mathbf{K}}$ is the complex polarization vector, and $c_{\mathbf{K}}^2 = 2\pi\omega_{\mathbf{K}}/\mathcal{V}$, with $\mathcal{V}$ representing the quantization volume and $\omega_{\mathbf{K}}$ being the x-ray frequency. In the initial state there are no x-ray photons, $|\Psi_a(t)\rangle = |\psi_a(t)\rangle|0_{\mathbf{K}}\rangle$, while in the final state there is one x-ray photon with wave vector $\mathbf{K}$, $|\Psi_b(t)\rangle = |\psi_b(t)\rangle|1_{\mathbf{K}}\rangle$.

The coordinates $\mathbf{r}_e$ and $\mathbf{r}_j$, $j = A, B$, can be replaced by the Jacobi coordinates using the following relations [64]

$$\begin{aligned} \mathbf{r}_e &= \mathbf{R}_{\text{cm}} + \frac{M_{AB}}{M} \mathbf{r}, \\ \mathbf{r}_A &= \mathbf{R}_{\text{cm}} - \frac{m_B}{M_{AB}} \mathbf{R} - \frac{m_e}{M} \mathbf{r}, \\ \mathbf{r}_B &= \mathbf{R}_{\text{cm}} + \frac{m_A}{M_{AB}} \mathbf{R} - \frac{m_e}{M} \mathbf{r}, \end{aligned} \quad (4)$$

where m_e and m_j , $j = A, B$, are the masses of the electron and the atomic centers, respectively, $M_{AB} = m_A + m_B$, $M = M_{AB} + m_e$. Physically, $\mathbf{R}_{\text{cm}}$ is the position of the center-of-mass, $\mathbf{R}$ is the relative position of the two atomic centers, and $\mathbf{r}$ is the relative position of the electron with respect to the center-of-mass of the two atomic centers. Because $m_e \ll m_A, m_B$ we can assume that $m_e/M \approx 0$ and $M_{AB}/M \approx 1$. The states $|\psi_i(t)\rangle$ $i = a, b$, can be written as

$$|\psi_i(\mathbf{r}, \mathbf{R}, \mathbf{R}_{\text{cm}}; t)\rangle = |\Xi_i(\mathbf{R}_{\text{cm}}; t)\rangle |\Phi_i(\mathbf{r}, \mathbf{R}; t)\rangle, \quad i = a, b, \quad (5)$$

where we added the explicit dependence on the Jacobi coordinates. The state $\Xi_i(\mathbf{R}_{\text{cm}}; t)$ is the Volkov state in length gauge [63]. In particular, for a strong laser field, the continuum electron is assumed to interact only with the assisting laser field, while its interaction with the molecular ion prior to recombination is neglected. Consequently, the initial continuum state is described by a Volkov state. The masses of the nuclei A and B are much larger than the electron mass so that we can use the Born-Oppenheimer approximation in which the relative position of the nuclei is fixed to the equilibrium position coordinate, i.e., $\mathbf{R} = \mathbf{R}_0$ [65]. In this case, the S -matrix element becomes

$$S_{fi} = -i\delta(\mathbf{P}_{\text{cm},b} - \mathbf{P}_{\text{cm},a} + \mathbf{K}) \int_{-\infty}^{\infty} d\tau C(\tau) \mathcal{M}(\tau), \quad (6)$$

where $\mathbf{P}_{\text{cm},i}$, $i = a, b$, is the center-of-mass momentum in the initial ($i = a$) and final ($i = b$) state, $C(\tau) = iec_{\mathbf{K}} e^{i\omega_{\mathbf{K}}\tau}$, and the matrix element can be written as (see Appendix B for details)

$$\mathcal{M}(\tau) = \langle \Phi_b(\mathbf{r}, \mathbf{R}_0; \tau) | \hat{\mathbf{e}}_{\mathbf{K}}^* \cdot \mathbf{r} e^{-i\mathbf{K} \cdot \mathbf{r}} | \Phi_a(\mathbf{r}, \mathbf{R}_0; \tau) \rangle. \quad (7)$$

The δ -function which appears in Eq. (6) corresponds to the momentum conservation.

If we assume that the electron is directly recombined to the ground state emitting an x-ray photon, the initial state, which corresponds to the free electron in the laser field, can be modeled by the Volkov wave vectors

$$|\Phi_a(\tau)\rangle = |\mathbf{p}_i + \mathbf{A}(\tau)\rangle \exp\{-i[\mathbf{p}_i \cdot \boldsymbol{\alpha}(\tau) + \mathcal{U}(\tau) + E_{\mathbf{p}_i}\tau]\}, \quad (8)$$

where $\mathbf{p}_i$ is the initial electron momentum (the corresponding energy is $E_{\mathbf{p}_i} = \mathbf{p}_i^2/2$), $\mathbf{A}(\tau) = -\int^\tau dt \mathbf{E}(t)$ is the vector potential of the assisting field $\mathbf{E}(\tau)$, $\boldsymbol{\alpha}(\tau) = \int^\tau dt \mathbf{A}(t)$, and $\mathcal{U}(\tau) = \int^\tau dt \mathbf{A}^2(t)/2$. The final state is a bound state

$$|\Phi_b(\tau)\rangle = |\psi_B\rangle \exp(-iE_B\tau), \quad (9)$$

where $E_B < 0$ is the energy of the bound state. Dropping the δ -function corresponding to the momentum conservation and using the periodicity of the subintegral function in the S -matrix element we can write

$$\begin{aligned} S_{\text{fi}} &= -c_{\mathbf{K}} \int_{-\infty}^{\infty} d\tau e^{-i(E_{\mathbf{p}_i} - E_B + U_p - \omega_{\mathbf{K}})\tau} \mathcal{T}_{\text{fi}}(\omega\tau) \\ &= -2\pi c_{\mathbf{K}} \sum_n \delta(E_B + \omega_{\mathbf{K}} - E_{\mathbf{p}_i} - U_p - n\omega) T_{\text{fi}}(n), \end{aligned} \quad (10)$$

where

$$\begin{aligned} \mathcal{T}_{\text{fi}}(\omega t) &= e^{-i[\mathbf{p}_i \cdot \boldsymbol{\alpha}(t) + \mathcal{U}_i(t)]} \langle \psi_B | \hat{\mathbf{e}}_{\mathbf{K}}^* \cdot \mathbf{r} e^{-i\mathbf{K} \cdot \mathbf{r}} | \mathbf{p}_i + \mathbf{A}(t) \rangle \\ &= \sum_{n=-\infty}^{+\infty} T_{\text{fi}}(n) e^{-in\omega t}, \end{aligned} \quad (11)$$

$$T_{\text{fi}}(n) = \int_0^T \frac{dt}{T} \mathcal{T}_{\text{fi}}(\omega t) e^{in\omega t}. \quad (12)$$

Here the Fourier coefficient $T_{\text{fi}}(n)$ represents the probability amplitude for recombination accompanied by the net absorption ($n > 0$) or emission ($n < 0$) of n photons from the assisting laser field, $T = 2\pi/\omega$ is the assisting-field period, $\mathcal{U}_i(t) = \mathcal{U}(t) - U_p t$, with U_p the ponderomotive energy, and the δ -function $\delta(E_B + \omega_{\mathbf{K}} - E_{\mathbf{p}_i} - U_p - n\omega)$ corresponds to the energy-conservation condition. The differential power spectrum for the laser-assisted electron-ion recombination is [26,66,67]

$$S_n(\mathbf{K}, \mathbf{p}_i) = \frac{p_i \omega_{\mathbf{K}}^4}{2\pi c^3} |T_{\text{fi}}(n)|^2, \quad p_i = |\mathbf{p}_i|. \quad (13)$$

We represent our bound state by a linear combination of the atomic orbitals

$$\psi_B = \sum_{k=A,B} \sum_a c_{ka} \psi_a(\mathbf{r}_e - \mathbf{r}_k), \quad (14)$$

where the sum over a denotes the sum over atomic orbitals, while the sum over k denotes the sum over the atomic centers A and B . The bound-state wave functions (14) can be calculated using various quantum-chemistry softwares [65]. For our homonuclear diatomic molecules we use the self-consistent-field wave functions represented by the linear combination of the Slater-type orbitals, which are obtained using the Hartree-Fock-Roothaan method [68]. In this case we have

$$\begin{aligned} \mathcal{T}_{\text{fi}}(\omega t) &= e^{-i[\mathbf{p}_i \cdot \boldsymbol{\alpha}(t) + \mathcal{U}_i(t)]} \sum_{s=\pm 1} \sum_a e^{-is[\mathbf{p}_i - \mathbf{K} + \mathbf{A}(t)] \cdot \mathbf{R}_0/2} \\ &\quad \times c_{sa} \langle \psi_a | \hat{\mathbf{e}}_{\mathbf{K}}^* \cdot \mathbf{r} | \mathbf{p}_i - \mathbf{K} + \mathbf{A}(t) \rangle, \end{aligned} \quad (15)$$

where $s = +1$ ($s = -1$) corresponds to the contribution of the atomic center A (B). The coefficients c_{-1a} and c_{1a} are related by $c_{-1a} = s_{a\lambda} c_{1a}$ where $s_{a\lambda} = (-1)^{l_a - m_a + m_\lambda}$ for gerade symmetry and $s_{a\lambda} = (-1)^{l_a - m_a + m_\lambda + 1}$ for ungerade symmetry [69]. Here l_a and m_a are the orbital and magnetic quantum numbers of the atomic orbital a , respectively, and $m_\lambda = -1$ ($m_\lambda = +1$) for σ (π) molecular orbital. Finally, the matrix element $\langle \psi_a | \hat{\mathbf{e}}_{\mathbf{K}}^* \cdot \mathbf{r} | \mathbf{p}_i - \mathbf{K} + \mathbf{A}(t) \rangle$ can be calculated analytically for different atomic orbitals. The explicit analytical expressions for the required matrix elements are given in Appendix A of Ref. [57], in particular by Eqs. (A13) and (A14). The expression for the T -matrix element for the case of the atomic targets is given in [67]. Finally, we mention that, since the molecular electronic structure is described within the Hartree-Fock-Roothaan method, the recombination matrix elements already incorporate the average effect of all electrons through the molecular orbital. During the recombination process, however, only one electron is treated dynamically, corresponding to the single-active-electron approximation. Consequently, electron correlation beyond the Hartree-Fock mean field, dynamic polarization of the remaining electrons, and multielectron excitation processes are neglected.

B. Interference minima and the geometry of the process

The summation over s (i.e., over the atomic centers) in Eq. (15) leads to

$$\mathcal{T}_{\text{fi}}(\omega t) \propto \begin{cases} \cos\{\mathbf{R}_0 \cdot [\mathbf{p}_i - \mathbf{K} + \mathbf{A}(t)]/2\}, & s_{a\lambda} = +1 \\ \sin\{\mathbf{R}_0 \cdot [\mathbf{p}_i - \mathbf{K} + \mathbf{A}(t)]/2\}, & s_{a\lambda} = -1 \end{cases}, \quad (16)$$

and the interference minima appear for $\mathbf{R}_0 \cdot [\mathbf{p}_i - \mathbf{K} + \mathbf{A}(t)]/2 = (2m+1)\pi/2$, $m = 0, 1, \dots$, for $s_{a\lambda} = +1$ and for $\mathbf{R}_0 \cdot [\mathbf{p}_i - \mathbf{K} + \mathbf{A}(t)]/2 = m\pi$, $m = 1, 2, \dots$, for $s_{a\lambda} = -1$.

Our linearly polarized assisting field is

$$\mathbf{E}(t) = E_0 \sin(\omega t) \hat{\mathbf{e}}_z, \quad (17)$$

where E_0 is the electric-field amplitude and $\hat{\mathbf{e}}_z$ is the unit polarization vector. We assume that the initial electron momentum $\mathbf{p}_i$ is in the xz plane and that θ_i is the angle between the unit vector $\hat{\mathbf{e}}_z$ and the momentum $\mathbf{p}_i$. We investigate the case in which homonuclear diatomic molecule is placed in the xz plane and the x-ray is emitted orthogonally to this plane, i.e., $\mathbf{K} = K\hat{\mathbf{e}}_y$ (the emitted x-ray is polarized in the xz plane). In this case, the position of the molecule can be defined by the angle θ_L which is the angle between the unit vector $\hat{\mathbf{e}}_z$ and the internuclear axis ($\mathbf{R}_0$).

The argument of the exponential function of the T -matrix element [see Eqs. (12) and (15)] is proportional to the semiclassical action. This action is stationary if its first derivative over the recombination time is zero. This leads to the stationary-phase equation

$$E_B + \omega_{\mathbf{K}} = [\mathbf{p}_i + \mathbf{A}(t)]^2/2, \quad (18)$$

which represents the energy-conservation condition at the recombination time t (for a review of stationary-phase method see [70]). Here we have taken into account that $\mathbf{R}_0 \cdot \mathbf{K} = 0$ for our setup and we have neglected the $\mathbf{R}_0$ -dependent term in the action. For the initial electron momentum along the

assisting-field polarization direction ($\theta_i = 0^\circ$) the interference minima conditions are

$$\omega_{\mathbf{K}}^{(+1)} = -E_B + \frac{(2m+1)^2\pi^2}{2R_0^2 \cos^2 \theta_L}, \quad m = 0, 1, \dots, \quad (19)$$

$$\omega_{\mathbf{K}}^{(-1)} = -E_B + \frac{2m^2\pi^2}{R_0^2 \cos^2 \theta_L}, \quad m = 1, 2, \dots, \quad (20)$$

where the value of $s_{a\lambda}$ is indicated in the superscript. For $\theta_i \neq 0^\circ$, the analytical condition for the interference minima cannot be derived.

C. Polarization of the emitted x-rays

The polarization properties of the emitted x-rays can be analyzed using the Stokes parameters. Since in our geometry the emitted x-ray propagates along the y axis, its polarization vector lies in the xz plane. It is convenient to introduce the T -matrix vector $\mathbf{T}(\mathbf{K}, \mathbf{p}_i)$ as the product of the T -matrix element $T_{fi}(n)$ and the complex polarization vector $\hat{\mathbf{e}}_{\mathbf{K}}$ of the emitted x-ray and to decompose it along the real unit vectors $\hat{\mathbf{e}}_x$ and $\hat{\mathbf{e}}_z$:

$$\mathbf{T}(\mathbf{K}, \mathbf{p}_i) = T_{fi}(n)\hat{\mathbf{e}}_{\mathbf{K}} = T_x(n)\hat{\mathbf{e}}_x + T_z(n)\hat{\mathbf{e}}_z, \quad (21)$$

where $T_x(n)$ and $T_z(n)$ correspond to the x and z polarization directions, respectively. The polarization state of the emitted radiation is characterized by the Stokes parameters [71,72].

$$S_0 = |T_x|^2 + |T_z|^2, \quad S_1 = |T_x|^2 - |T_z|^2, \quad (22)$$

$$S_2 = 2\text{Re}(T_x T_z^*), \quad S_3 = 2\text{Im}(T_x T_z^*). \quad (23)$$

Here S_0 determines the total intensity of the emitted radiation (the differential power spectrum is proportional to S_0), while S_1 , S_2 , and S_3 describe its polarization properties.

For a linearly polarized assisting laser field, the emitted x-rays are also linearly polarized so that the circular-polarization Stokes parameter vanishes, $S_3 = 0$. In this case, the polarization state is solely determined by the parameters S_1 and S_2 . The polarization angle $\chi_{\mathbf{K}}$ between the electric-field vector of the emitted x-ray and the x axis can then be written as

$$\chi_{\mathbf{K}} = \frac{1}{2} \arctan\left(\frac{S_2}{S_1}\right). \quad (24)$$

As an example we mention a few limiting cases. If the emitted x-rays are polarized along the x direction, then $|T_x| > |T_z|$ and $S_1 > 0$, $S_2 = 0$, which leads to $\chi_{\mathbf{K}} = 0$. On the other hand, if the emitted x-rays are polarized along the z direction, then $|T_z| > |T_x|$ and $S_1 < 0$, $S_2 = 0$. In this case, the polarization angle is $\chi_{\mathbf{K}} = \pm\pi/2$, where the two values are physically equivalent since the polarization direction is unchanged under the transformation $\chi_{\mathbf{K}} \rightarrow \chi_{\mathbf{K}} + \pi$. Finally, for equal magnitudes of the two polarization components, $|T_x| = |T_z|$, and $S_1 = 0$ and $S_2 > 0$, the polarization vector is oriented along the diagonal direction in the xz plane and the corresponding polarization angle is $\chi_{\mathbf{K}} = \pi/4$. In general, the polarization angle $\chi_{\mathbf{K}}$ depends on the geometry of the process, parameters of the assisting field, and the energy of the emitted x-ray.

D. Averaging over molecular alignment

For realistic experimental conditions, the molecular axis is not perfectly aligned with respect to the assisting-field polarization direction. To investigate the influence of imperfect alignment on the recombination spectra, we average the differential power spectrum over the molecular orientation angle θ_L using an angular distribution function $W(\theta_L)$. The averaged differential power spectrum is defined as

$$\langle S_n(\mathbf{K}, \mathbf{p}_i) \rangle = \frac{\int_0^\pi d\theta_L \sin \theta_L W(\theta_L) S_n(\mathbf{K}, \mathbf{p}_i; \theta_L)}{\int_0^\pi d\theta_L \sin \theta_L W(\theta_L)}, \quad (25)$$

where on the right-hand side we have added an explicit dependence of the differential power spectrum on the molecular orientation angle θ_L . As a simple model of partial molecular alignment, we assume a Gaussian angular distribution around the assisting-field polarization direction

$$W(\theta_L) = N \exp\left(-\frac{\theta_L^2}{2\sigma^2}\right), \quad (26)$$

where σ characterizes the degree of molecular alignment and N is the normalization constant. In the limit $\sigma \rightarrow 0$, the molecular ensemble becomes perfectly aligned along the assisting-field polarization direction, while larger values of σ correspond to broader angular distributions. The averaging introduced here represents a simplified model of partial molecular alignment. In particular, we average only over the angle θ_L between the molecular axis and the assisting-field polarization direction, while the molecular axis is assumed to remain within the polarization plane. Therefore, the present treatment does not correspond to a full three-dimensional orientational averaging. Nevertheless, this simplified model is sufficient to investigate the robustness of the interference structures against imperfect molecular alignment and to estimate the extent to which the interference minima survive angular broadening. While the present calculations employ a simple averaging over the molecular orientation, the corresponding two- and three-dimensional averaging schemes have been described in [73].

III. NUMERICAL RESULTS

In the present paper we analyze laser-assisted electron-ion radiative recombination using the N_2^+ , O_2^+ , and He_2^+ molecular ions as examples. The final bound states are the states of the neutral N_2 , O_2 , and He_2 molecules. The bound state energy and the atomic orbitals used in a linear combination representing the corresponding highest-occupied molecular orbital (HOMO) are given in Table I. In particular, the HOMO of the N_2 molecule has five s (denoted by $1s, 1s', 2s, 2s', 3s$ in Table I), three p , three d , and one f atomic orbitals, the HOMO of the O_2 molecule has three p , one d , and one f atomic orbitals, while the HOMO of the He_2 molecule consists of two s , one p , and one d atomic orbitals. The equilibrium internuclear distances for the N_2 , O_2 , and He_2 molecules are 2.1 a.u., 2.3 a.u., and 5.6 a.u., respectively. For He_2 with the equilibrium distance between the atomic centers, only the contributions of the s atomic orbitals are relevant. We also analyze the

TABLE I. Final state HOMOs, bound state energies E_B , and atomic orbitals used to model the molecular orbital for the molecular species used in the paper.

Molecule	HOMO	$ E_B (\text{eV})$	Atomic orbitals
N_2	$3\sigma_g$	15.58	$1s, 1s', 2s, 2s', 3s, 2p, 2p', 2p'', 3d, 3d', 3d'', 4f$
O_2	$1\pi_g$	12.03	$2p, 2p', 2p'', 3d, 4f$
He_2	$1\sigma_u$	22.22	$1s, 1s', 2p, 3d$

case of the He_2 with shorter internuclear distance in which case $2p$ and $3d$ atomic orbitals should also be taken into account.

A. Differential power spectra and molecular interference

We start our analysis by investigating the differences between the differential power spectra calculated for the radiative recombination with atomic and molecular ions. Figure 1 presents a comparison between the differential power spectra obtained for selected diatomic molecular ions and those corresponding to atomic species with similar bound-state energies. The calculations are performed for an assisting laser field with intensity $2 \times 10^{15} \text{ W/cm}^2$ and wavelength of 800 nm (for these values of the parameters, the nondipole effects are still small [74,75]). The incident electron energy is 10 eV, with the momentum aligned along the laser polarization direction ($\theta_i = 0^\circ$), and the molecular axis is taken parallel to the assisting field ($\theta_L = 0^\circ$).

At lower emitted-photon energies (up to approximately 150 eV), the height of the plateaus for molecular ions and their corresponding atomic counterparts are very similar,

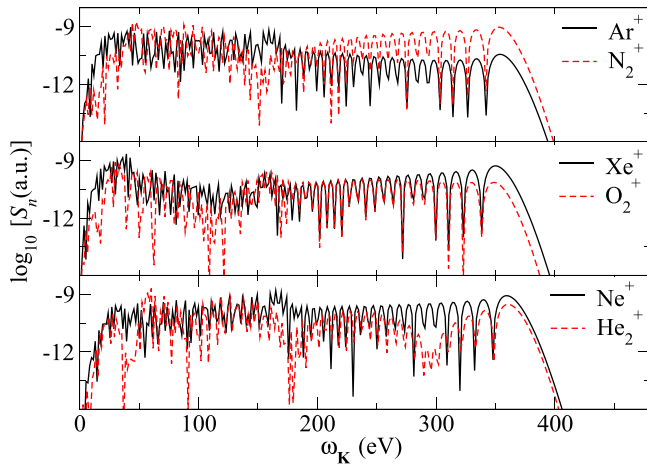


FIG. 1. Logarithm of the differential power spectrum as a function of the emitted photon energy ω_K for selected diatomic molecular ions (red dashed lines) and atomic species with similar bound-state energies (black solid lines). The upper, middle, and lower panels correspond to the pairs $\text{Ar}^+ - \text{N}_2^+$, $\text{Xe}^+ - \text{O}_2^+$, and $\text{Ne}^+ - \text{He}_2^+$, respectively. The assisting laser field has an intensity of $2 \times 10^{15} \text{ W/cm}^2$ and a wavelength of 800 nm. The initial electron energy is 10 eV, with the momentum aligned along the laser polarization direction ($\theta_i = 0^\circ$), and the molecular axis is oriented parallel to the laser field ($\theta_L = 0^\circ$).

indicating that, for the systems considered here, molecular effects are comparatively weak in this energy range. However, significant differences emerge in the high-energy region. In particular, for some molecular ions the differential power spectrum exceeds that of the corresponding atomic system, as observed for N_2^+ , where the x-ray yield becomes higher at large ω_K . In contrast, for other systems such as the O_2^+ and He_2^+ , the molecular spectrum is suppressed relative to the atomic case in the same energy range. The bound-state energies of the Ne, Ar, and Xe atoms are 21.56 eV, 15.76 eV, and 12.13 eV, respectively, i.e., they are almost the same as for the corresponding molecular species. Since the compared atomic and molecular species have very similar bound-state energies, the differences that appear in the spectra originate from the recombination matrix element. In particular, high-energy x-rays are emitted when the electron recombines with a large instantaneous momentum $\mathbf{p}_i = \mathbf{K} + \mathbf{A}(t)$. In this regime the recombination amplitude becomes increasingly sensitive to the momentum-space structure of the bound molecular orbital, i.e., on the relative contributions of the constituent atomic orbitals, their symmetry, and the interference between the recombination amplitudes associated with the two atomic centers. Since these properties differ substantially for the N_2 , O_2 , and He_2 molecules, qualitatively different behaviors are obtained in the high-energy part of the spectra. By contrast, in the low-energy region the recombination amplitudes of the corresponding atomic and molecular systems remain more similar, leading to comparable spectra. In addition, the close agreement between the spectra for Xe^+ and O_2^+ over most of the plateau indicates that the corresponding recombination matrix elements have a very similar momentum dependence. This is consistent with the fact that the recombination in both systems is dominated by p -orbital contributions. As a result, the molecular spectrum closely follows the atomic one and the noticeable differences appear only in the vicinity of the interference minimum and near the end of the plateau where the spectrum is most sensitive on the characteristics of the target. The most pronounced distinction between molecular and atomic targets is the presence of characteristic minima in the molecular spectra, which are absent for atomic systems. These minima may appear in different parts of the differential power spectrum and they arise from molecular interference effects, i.e., they are caused by the destructive interference of the contributions corresponding to the electrons recombined at one or the other atomic centers. For the N_2^+ ion, a clear minimum is observed around 150 eV, while for the O_2^+ ion it appears near 120 eV. In the case of He_2^+ , multiple minima are visible, located approximately at 40 eV, 90 eV, 180 eV, and 290 eV. These features highlight the role of two-center interference in shaping the recombination spectrum and underline the qualitative differences between molecular and atomic responses in strong-field-assisted recombination.

B. Dependence on the molecular orientation angle

The position of the interference minima changes as a function of the molecular orientation angle θ_L so that their dependence on the emitted-photon energy and the angle θ_L should be analyzed simultaneously. In Fig. 2 we present the

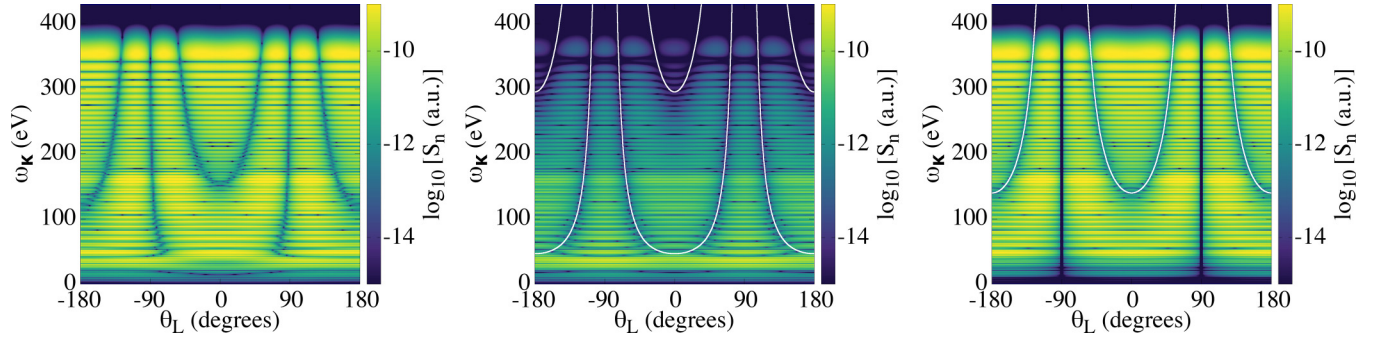


FIG. 2. Logarithm of the differential power spectrum as a function of the emitted photon energy $\omega_{\mathbf{K}}$ and the molecular orientation angle θ_L for electron recombination with the N_2^+ ion. The left panel shows the spectrum obtained by including all contributing atomic orbitals in the recombination amplitude. The middle panel presents the result obtained by retaining only the orbitals with $s_{a\lambda} = +1$ (corresponding to the s and d atomic orbitals), while the right panel shows the contribution from orbitals with $s_{a\lambda} = -1$ (corresponding to the p and f atomic orbitals). The white curves represent the corresponding interference-minimum conditions. The laser-field parameters are the same as in Fig. 1 and $\mathbf{p}_i$ is in the direction of the assisting field.

differential power spectrum as a function of the emitted photon energy $\omega_{\mathbf{K}}$ and the molecular orientation angle θ_L for the electron radiative recombination with the N_2^+ ion (the electron is recombined into the $3\sigma_g$ state). The left panel presents the full result obtained by including all contributing atomic orbitals in the recombination amplitude. The middle and right panels display the spectra calculated by retaining only the orbitals with $s_{a\lambda} = +1$ (corresponding to the s and d atomic orbitals) and only the orbitals with $s_{a\lambda} = -1$ (corresponding to the p and f atomic orbitals), respectively. When only orbitals with the same value of $s_{a\lambda}$ are included, pronounced interference minima become clearly visible. Moreover, the positions of these minima are reproduced accurately by the corresponding interference conditions [Eqs. (19) and (20)], shown in Fig. 2 by the white curves. The spectra obtained in this way possess the symmetries $\theta_L \rightarrow -\theta_L$ and $\theta_L \rightarrow \pi - \theta_L$, which is consistent with the symmetry properties of the underlying interference factors. In addition, the differential power spectrum calculated using only p and f atomic orbitals ($s_{a\lambda} = -1$) is zero for $\theta_L = \pm\pi/2$. This happens because, for these atomic orbitals, the matrix element $\langle \psi_a | \hat{\mathbf{e}}_{\mathbf{K}}^* \cdot \mathbf{r} | \mathbf{p}_i - \mathbf{K} + \mathbf{A}(t) \rangle$ contains $\cos \theta_L$ as a factor which vanishes for $\theta_L = \pm\pi/2$ (see Appendix A in [57]).

A comparison of the middle and right panels demonstrates that the contributions originating from the orbitals with $s_{a\lambda} = -1$ are considerably stronger than those associated with $s_{a\lambda} = +1$. Consequently, when all atomic orbitals are included simultaneously, the overall spectrum shown in the left panel is largely dominated by the features characteristic of the p and f orbitals. Nevertheless, the interference minima no longer coincide with the positions predicted separately for either $s_{a\lambda} = +1$ or $s_{a\lambda} = -1$. Instead, the minima are shifted due to the interplay and coherent superposition of contributions with different symmetry properties. An additional important feature of the full spectrum is the modification of its angular symmetry. While the symmetry under the transformation $\theta_L \rightarrow -\theta_L$ remains preserved, the symmetry with respect to $\theta_L \rightarrow \pi - \theta_L$ is broken if all orbital contributions are taken into account. This behavior originates from the interference between recombination channels associated with orbitals of opposite parity. In particular, the sum over the atomic orbitals

a which appears in the T -matrix element can be separated as

$$\begin{aligned} \mathcal{T}_{\text{fi}}(\omega t) \propto & \sum_{a_+} c_{1a_+} \cos\{\mathbf{R}_0 \cdot [\mathbf{p}_i + \mathbf{A}(t)]/2\} \\ & - i \sum_{a_-} c_{1a_-} \sin\{\mathbf{R}_0 \cdot [\mathbf{p}_i + \mathbf{A}(t)]/2\}, \quad (27) \end{aligned}$$

where the sum over a_+ (a_-) is the sum over the atomic orbitals with $s_{a\lambda} = +1$ ($s_{a\lambda} = -1$). For $\mathbf{p}_i$ in the direction of the assisting field, the scalar product $\mathbf{R}_0 \cdot [\mathbf{p}_i + \mathbf{A}(t)] \propto \cos \theta_L$. The transformation $\theta_L \rightarrow -\theta_L$ does not change the sign of $\cos \theta_L$, while the transformation $\theta_L \rightarrow \pi - \theta_L$ changes its sign. Consequently, when only the orbitals with $s_{a\lambda} = +1$ or $s_{a\lambda} = -1$ are taken into account, the change of the sign of the factor $\cos \theta_L$ does not affect the final result so that the power spectrum is invariant with respect to both transformations (see the middle and right panels of Fig. 2). On the contrary, when orbitals with different $s_{a\lambda}$ are taken into account the change of the sign of the factor $\cos \theta_L$ affects the final result and the power spectrum exhibits only the symmetry with respect to the transformation $\theta_L \rightarrow -\theta_L$ (see the left panel of Fig. 2).

Similar analysis can be performed for other molecular species. In Fig. 3 we present the differential power spectrum as a function of the emitted photon energy $\omega_{\mathbf{K}}$ and the molecular orientation angle θ_L for the electron radiative recombination with the O_2^+ (left panel) and He_2^+ (middle panel) molecular ions. As in the previous analysis, the spectra are used to investigate the role of the individual atomic-orbital contributions in determining the interference structure of the recombination signal. For the O_2^+ molecular ion, the contribution originating from the p orbitals ($s_{a\lambda} = -1$ because O_2 has $1\pi_g$ molecular orbital) is dominant. Consequently, the structure of the total spectrum is governed primarily by the interference pattern associated with the p -orbital component of the molecular wave function. In particular, the positions of the minima observed in the full spectrum coincide with those predicted for the p -orbital interference condition, demonstrating that the recombination dynamics is controlled mainly by the odd-parity orbital contribution (see the white lines in the left panel of Fig. 3). As a result, the overall angular and spectral dependence closely resembles the one for the isolated

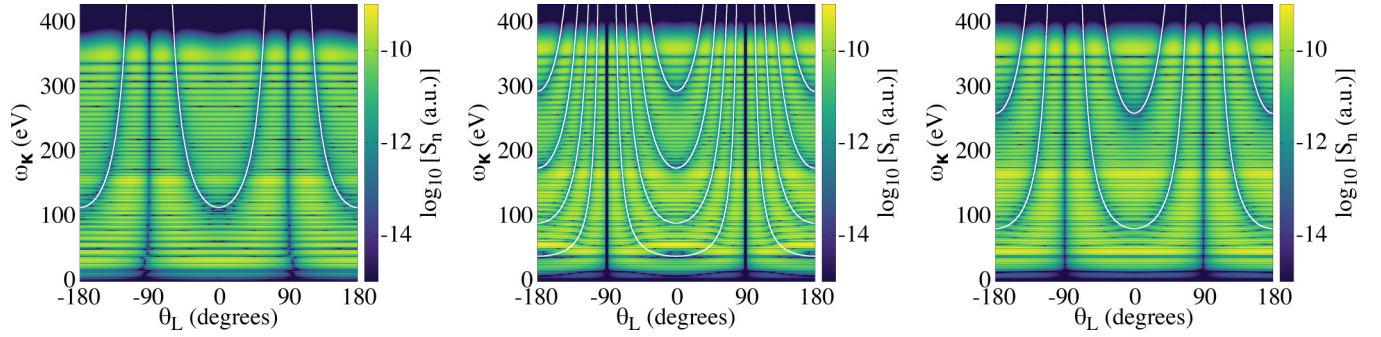


FIG. 3. Logarithm of the differential power spectrum as a function of the emitted photon energy ω_K and the molecular orientation angle θ_L . The left (middle) panel shows the spectrum corresponding to the O_2^+ (He_2^+) molecular ion with equilibrium internuclear distance. The right panel presents the results analogous to those presented in the middle panel but for the internuclear distance $R_0 = 3$ a.u. The white curves represent the corresponding interference-minimum conditions. The laser-field parameters are the same as in Fig. 1 and $\mathbf{p}_i$ is in the direction of the assisting field.

p -orbital contribution. In addition, the minima present for $\theta_L = \pm\pi/2$, which are caused by the presence of the factor $\cos\theta_L$ in the matrix element $\langle\psi_a|\hat{\mathbf{e}}_K^* \cdot \mathbf{r}|\mathbf{p}_i - \mathbf{K} + \mathbf{A}(t)\rangle$, are slightly blurred due to the small contributions of other atomic orbitals.

In contrast, the He_2^+ molecular ion represents a considerably simpler situation. For the equilibrium internuclear distance $R_0 = 5.6$ a.u. (the corresponding power spectrum is shown in the middle panel of Fig. 3), the molecular orbital is formed exclusively from s atomic orbitals. The molecular orbital is $1\sigma_u$ so that $s_{a\lambda} = -1$ for s atomic orbitals. Therefore, the interference structure of the spectrum is determined entirely by the s -orbital contribution. The minima appearing in the differential power spectrum are reproduced well by the analytical interference curves corresponding to the s orbitals, confirming the validity of the interference model in this case (see the white lines in the middle panel of Fig. 3). Owing to the absence of competing orbital contributions with different symmetry properties, the minima remain well defined and follow the predicted interference-minima conditions throughout the considered energy and angular ranges. Similarly as for the O_2 molecule, additional minima appear for $\theta_L = \pm\pi/2$. However, their origin is different. In particular, for the O_2 molecule they appear due to the fact that for p atomic orbitals (which are dominant in the molecular orbital of the O_2 molecule) the matrix element contains the term $\cos\theta_L$ which vanishes for $\theta_L = \pm\pi/2$. This is not the case for the He_2 molecule where, for the equilibrium internuclear distance, only s orbitals (with $s_{a\lambda} = -1$) are relevant. The matrix element does not vanish for $\theta_L = \pm\pi/2$. However, the sum over the atomic centers leads to the term $\sum_{a-} c_{1a-} \sin\{\mathbf{R}_0 \cdot [\mathbf{p}_i + \mathbf{A}(t)]/2\}$. In our case, the initial electron momentum is along the assisting-field polarization direction so that the argument of the sine function is proportional to $\cos\theta_L$ and thus the whole term vanishes for $\theta_L = \pm\pi/2$ even though the recombination matrix element is not zero.

Finally, we also present the differential power spectrum for the He_2^+ molecular ion calculated for a smaller internuclear distance (see the right panel of Fig. 3). In comparison with the spectrum obtained for the equilibrium separation $R_0 = 5.6$ a.u., the number of interference minima within the same photon-energy range is reduced. This behavior is

consistent with the dependence of the interference condition on the internuclear distance, since a smaller separation leads to a larger spacing between successive minima. Furthermore, the minima appearing for $\theta_L = \pm\pi/2$ become slightly blurred in comparison with the case of the equilibrium internuclear distance. This effect originates from the fact that, for the smaller separation, the molecular wave function is no longer determined exclusively by the s orbitals. Additional orbital contributions exist, and the coherent superposition of these channels partially washes out the sharp interference pattern characteristic of the pure s -orbital case. Nevertheless, the main interference structures remain visible, demonstrating that the recombination spectrum still retains clear signatures of two-center interference. In the end we stress that the richness of the interference structure is strongly affected by the internuclear distance, with longer separations producing more minima, while shorter distances lead to simpler interference features. This also becomes evident by comparing the left panel of Fig. 3 with the middle and right panels of the same figure even though the constituting orbitals and the bound-state energy are different for O_2^+ and He_2^+ molecular ions.

The presented results demonstrate that the molecular structure is directly imprinted onto the differential power spectrum of the emitted x-rays. In particular, for fixed assisting-field parameters, the positions of the interference minima are determined by the internuclear distance, molecular orientation, and symmetry properties of the molecular orbital. Consequently, the position of the minima in the (ω_K, θ_L) plane represents a characteristic fingerprint of the molecular target. Moreover, the interference structure provides direct information about the dominant atomic-orbital contributions forming the molecular state. In particular, the positions and symmetry properties of the minima allow one to determine whether the recombination process is governed predominantly by atomic orbitals with $s_{a\lambda} = +1$ or $s_{a\lambda} = -1$. The differential power spectrum therefore not only reflects the recombination dynamics, but also carries detailed information about the electronic and geometric structure of the molecule.

Finally, we investigate the influence of imperfect molecular alignment on the interference structure of the differential power spectrum. Figure 4 presents the alignment-averaged spectra for the He_2^+ molecular ion (with equilibrium

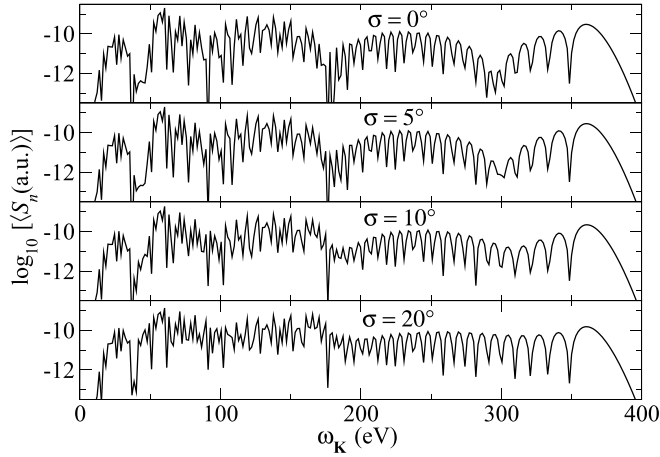


FIG. 4. Logarithm of the alignment-averaged differential power spectrum as a function of the emitted photon energy $\omega_{\mathbf{k}}$ for the He_2^+ molecular ion for different degrees of molecular alignment as indicated in the panels and for the same other parameters as in Fig. 1.

internuclear distance) calculated using the averaging procedure introduced in Sec. II D for different degrees of molecular alignment. The upper panel corresponds to the case of perfect alignment, while the remaining panels illustrate progressively broader angular distributions characterized by $\sigma = 5^\circ$, 10° , and 20° . For a strong molecular alignment ($\sigma = 5^\circ$), the interference minima remain clearly visible and their positions are only slightly modified in comparison with the perfectly aligned case. For a good alignment ($\sigma = 10^\circ$), the minima become smoother, particularly in the high-energy part of the spectrum, although the main interference pattern is still preserved. In the case of a moderate alignment ($\sigma = 20^\circ$), the higher-order minima become substantially blurred and several of them can no longer be clearly resolved, while the low-energy minima remain comparatively robust. This behavior can be understood from the interference conditions (19) and (20). The positions of the interference minima strongly depend on the molecular orientation angle through the factor $\cos \theta_L$. For higher-order minima, corresponding to larger emitted-photon energies, even relatively small variations of θ_L produce substantial shifts of the interference positions. Consequently, averaging over broader angular distributions washes out the high-energy interference structures more efficiently than the low-energy ones. The results presented in Fig. 4 therefore demonstrate that the low-order interference minima are comparatively robust against imperfect molecular alignment and should remain observable under experimentally realistic alignment conditions.

C. Influence of the incident-electron direction

We now discuss the influence of the angle θ_i between the incident electron momentum and the assisting-field polarization direction on the interference structure of the differential power spectrum. For $\theta_i = 0^\circ$, the incident electron momentum is parallel to the assisting field and the interference minima are well pronounced over a broad photon-energy range. However, as the angle θ_i increases, the minima gradually become blurred and eventually disappear, starting from the low-energy region

of the spectrum. For sufficiently large values of θ_i , the interference structure can become so weak that the minima are no longer clearly recognizable.

This behavior can be understood from the interference factor $\cos\{\mathbf{R}_0 \cdot [\mathbf{p}_i + \mathbf{A}(t)]/2\}$ or $\sin\{\mathbf{R}_0 \cdot [\mathbf{p}_i + \mathbf{A}(t)]/2\}$ appearing in the recombination amplitude. For $\theta_i = 0^\circ$, the scalar product $\mathbf{R}_0 \cdot [\mathbf{p}_i + \mathbf{A}(t)]$ depends only on the field-driven longitudinal motion of the electron, so that the interference conditions are satisfied in a well-defined and coherent manner. When $\theta_i \neq 0^\circ$, the incident electron momentum acquires a transverse component with respect to the assisting-field polarization direction. As a result, the phase entering the interference factor contains an additional contribution proportional to $\mathbf{p}_i \cdot \mathbf{R}_0$, which does not oscillate synchronously with the laser-driven motion. Consequently, electrons recombining at different times contribute with different interference phases, reducing the contrast of the destructive interference minima.

The effect is most pronounced in the low-energy region of the emitted spectrum. According to the stationary-phase equation (18), low-energy x-rays are emitted for recombination times for which $[\mathbf{p}_i + \mathbf{A}(t)]^2$ is relatively small. In this regime, the transverse component of the initial momentum associated with $\theta_i \neq 0^\circ$ becomes comparable to the laser-driven contribution $\mathbf{A}(t)$ and therefore strongly modifies the interference phase. The destructive interference condition is then fulfilled less efficiently over the contributing recombination times, which leads to the rapid blurring of the minima. In contrast, high-energy x-rays originate from recombination times for which the instantaneous momentum $\mathbf{p}_i + \mathbf{A}(t)$ is dominated by the laser-driven contribution $\mathbf{A}(t)$. In this case the relative influence of the transverse component of the incident momentum becomes weaker, so that the interference phase remains approximately synchronized with the laser-driven motion. Consequently, the interference minima in the high-energy part of the spectrum remain visible even for larger values of the angle θ_i .

Figure 5 illustrates the influence of the angle θ_i between the incident electron momentum and the assisting-field polarization direction on the interference structure of the differential power spectrum for electron recombination with the N_2^+ ion. The left and right panels correspond to $\theta_i = 45^\circ$ and $\theta_i = 75^\circ$, respectively. In comparison with the case $\theta_i = 0^\circ$, the interference minima become progressively blurred as the angle θ_i increases. This effect is particularly pronounced in the low-energy part of the spectrum, where the minima gradually disappear and the interference structure becomes difficult to identify. On the other hand, the minima in the high-energy region remain visible even for relatively large values of θ_i . This behavior reflects the increasing importance of the transverse component of the incident electron momentum for nonzero θ_i . In the low-energy region, where the instantaneous electron momentum is relatively small, the transverse momentum component strongly modifies the interference phase and weakens the destructive interference between contributions originating from the two atomic centers. In contrast, for high-energy x-rays the electron dynamics is dominated by the laser-driven contribution $\mathbf{A}(t)$, so that the influence of the transverse momentum component becomes comparatively weak and the interference minima remain well defined. A comparison

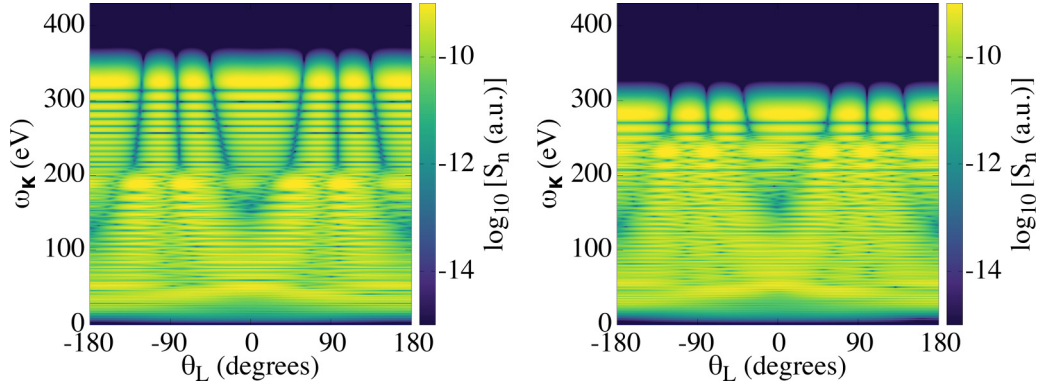


FIG. 5. Same as in the left panel of Fig. 2 but for the values of the incident-electron angle $\theta_i = 45^\circ$ (left panel) and $\theta_i = 75^\circ$ (right panel).

of the two panels clearly demonstrates that increasing θ_i reduces the visibility of the interference pattern primarily in the low-energy region, while the high-energy part of the spectrum preserves the characteristic signatures of two-center interference.

D. Polarization properties of the emitted x-rays

Apart from the differential power spectrum, it is also important to analyze the nature of the polarization state of the emitted x-rays. For a linearly polarized assisting laser field considered here, the emitted x-rays are linearly polarized. This is the case regardless of the value of the angle θ_i between the incident electron momentum and the assisting-field polarization direction. This behavior follows from the structure of the T -matrix components T_x and T_z . In particular, for a linearly polarized assisting field these components do not acquire a relative phase shift that would lead to a nonzero circular-polarization Stokes parameter S_3 , i.e., they oscillate synchronously. Consequently, the emitted radiation is linearly polarized and its polarization state is determined by the angle χ_K [Eq. (24)]. This behavior differs from the situation encountered in high-order harmonic generation with molecular targets. Although the recombination matrix element appearing in HHG is formally analogous to that entering the present laser-assisted recombination process, molecular HHG driven by a linearly polarized laser field generates elliptically polarized harmonics [76,77]. The origin of this effect lies in the fact that HHG is a multistep process involving ionization, continuum propagation, and recombination. During the propagation in the laser field, the electronic wave packet accumulates complex phases and multiple quantum trajectories contribute coherently to the emitted radiation. For molecular targets, the interference between different recombination channels and the introduced anisotropy produce a nontrivial relative phase between orthogonal components of the T -matrix element, resulting in nonzero ellipticity. In contrast, in the recombination process considered in this paper, the incident electron momentum is fixed externally and the emitted x-ray polarization is determined directly by the recombination event itself, without the additional phase accumulation associated with the continuum dynamics characteristic of HHG. As a result, the emitted x-rays remain linearly polarized, while their polarization direction depends on the emitted-photon energy and

on the geometry of the process. In Fig. 6 we present the polarization angle χ_K of the emitted x-rays as a function of the emitted-photon energy ω_K and the angle θ_i between the initial electron momentum and the assisting-field polarization direction. The calculations are performed for electron recombination with the N_2^+ ion with the molecular axis aligned parallel to the assisting laser field ($\theta_L = 0^\circ$). Two distinctive regions can be identified. For the low and the medium energy of the emitted x-rays, the polarization direction depends sensitively on both the emitted-photon energy and the direction of the incident electron momentum. On the other hand, for the high-energy x-rays, the polarization direction only slightly changes as a function of the angle θ_i . This can be explained using the stationary-phase equation (18). Since we neglected the $\mathbf{R}_0$ -dependent term, our classical analysis does not depend neither on the molecular orientation angle nor on the equilibrium internuclear distance. In Fig. 7 we present the solutions of the stationary-phase equation (18) for the parameters of Fig. 6 and for $\theta_i = 30^\circ$ (black solid line) and $\theta_i = 120^\circ$ (red dashed line). The photon energies 181.1 eV for $\theta_i = 30^\circ$ and

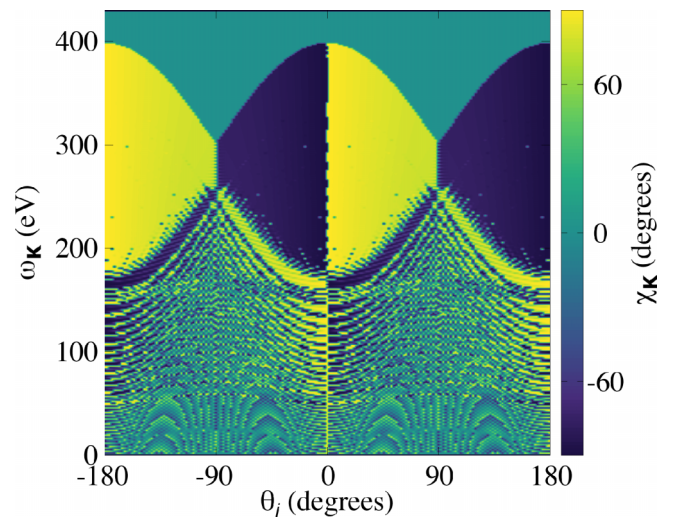


FIG. 6. The angle χ_K between the electric-field vector of the emitted x-ray and the x axis as a function of the emitted photon energy ω_K and the angle θ_i between the unit vector $\hat{\mathbf{e}}_z$ and the momentum $\mathbf{p}_i$ for radiative recombination with the N_2^+ ion. The molecular orientation angle is $\theta_L = 0^\circ$.

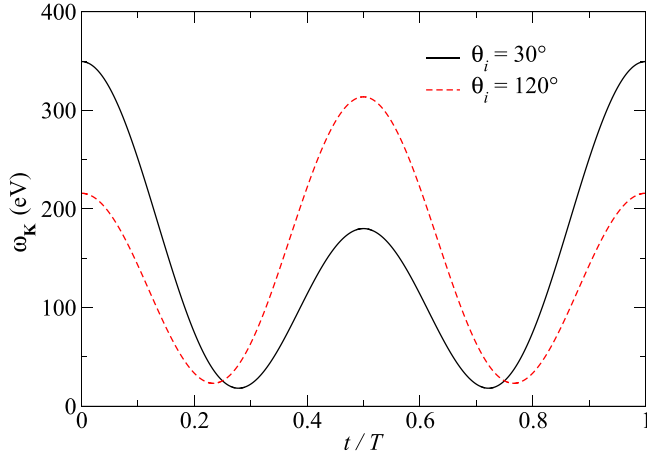


FIG. 7. Solutions of the stationary-phase equation (18) for the recombination time and for the parameters of Fig. 6 and for $\theta_i = 30^\circ$ (black solid line) and $\theta_i = 120^\circ$ (red dashed line).

215.9 eV for $\theta_i = 120^\circ$ correspond to the transition between the regimes with four and two stationary-phase solutions (more details regarding the change of the number of solutions of the stationary-phase equation can be found in [78]). These transition energies therefore separate the region of Fig. 6 where the polarization direction varies rapidly because of the interference of four trajectories, from the region where only two trajectories contribute and the polarization direction becomes almost constant. When only two solutions exist (they are always within the same half cycle of the assisting field), the emitted x-rays are polarized close to the polarization direction of the assisting field and their polarization only slightly changes as a function of the energy ω_K and the angle θ_i . On the other hand, when there are four stationary-phase solutions (they appear in pairs; each pair is within one half cycle of the assisting field), their partial contributions interfere which directly affects the polarization direction of the emitted x-rays which now changes rapidly as a function of ω_K and θ_i . The energy at which the transition happens depends on the angle θ_i . In addition, the recombination time corresponding to the high-energy x-rays depends on the angle θ_i . For example, for $\theta_i = 30^\circ$ the x-rays with maximum energy are recombined around $t = 0$ (or equivalently $t = T$), while for $\theta_i = 120^\circ$ the x-rays with maximum energy are recombined around $t = T/2$ (see Fig. 7). This affects the sign of the angle χ_K . Physically, both signs of the angle χ_K correspond to the x-rays linearly polarized along the same direction. These conclusions do not depend on the molecular orientation angle and they are valid for the recombination with the atomic ions as well. We have verified that the qualitative behavior discussed in Fig. 6 persists for other values of the molecular orientation angle θ_L , indicating that the energy dependence of the polarization direction is a general feature of the laser-assisted recombination process rather than a consequence of a particular molecular orientation.

However, the symmetry properties of the polarization angle χ_K as a function of the angle θ_i depend on the molecular orientation angle θ_L . In particular, for $\theta_L = 0^\circ$ the polarization angle χ_K is invariant under the transformation $\theta_i \rightarrow \theta_i + \pi$, while it changes the sign under the transformation $\theta_i \rightarrow -\theta_i$.

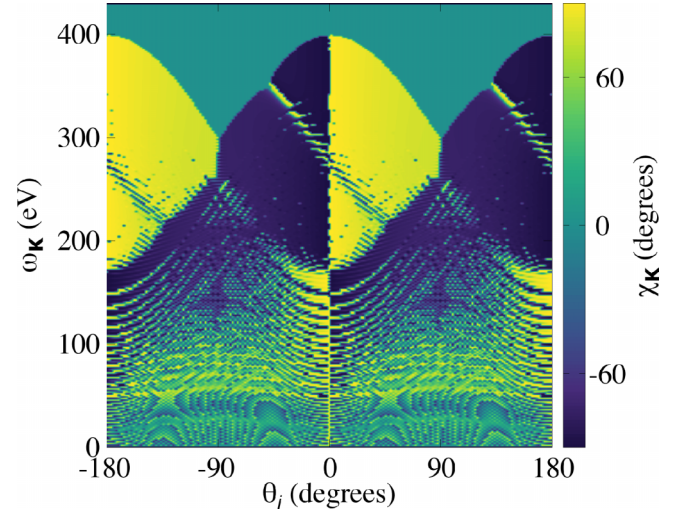


FIG. 8. Same as in Fig. 6 but for the molecular orientation angle $\theta_L = 45^\circ$.

Since a change of the sign does not affect the polarization direction of the emitted x-ray, the corresponding polarization pattern remains unchanged. For $\theta_L \neq 0^\circ$, however, only the invariance with respect to the transformation $\theta_i \rightarrow \theta_i + \pi$ is preserved. This can be understood by analyzing the behavior of the individual terms in Eq. (15) under these transformations. In particular, the transformation $\theta_i \rightarrow \theta_i + \pi$ corresponds to $\mathbf{p}_i \rightarrow -\mathbf{p}_i$. Furthermore, the molecule is invariant under a rotation by the angle π around the axis perpendicular to the polarization plane (the y axis), while the assisting field satisfies the half-cycle symmetry $\mathbf{E}(t + T/2) = -\mathbf{E}(t)$, along with $\mathbf{A}(t + T/2) = -\mathbf{A}(t)$ and $\boldsymbol{\alpha}(t + T/2) = -\boldsymbol{\alpha}(t)$. Consequently, all scalar products entering the transition amplitude remain invariant under the transformation $\theta_i \rightarrow \theta_i + \pi$. In addition, this transformation changes the recombination matrix element at most by an overall sign, which is identical for all contributing atomic orbitals (see Appendix A in [79]). As a result, the T -matrix element (12) is invariant under the transformation $\theta_i \rightarrow \theta_i + \pi$, up to an overall sign change.

On the other hand, the transformation $\theta_i \rightarrow -\theta_i$ preserves the polarization direction of the emitted x-ray only for $\theta_L = 0^\circ$ (or equivalently $\theta_L = \pi$; see Fig. 6). For other values of θ_L , the scalar product $\mathbf{p}_i \cdot \mathbf{R}_0$ is not invariant under the transformation $\theta_i \rightarrow -\theta_i$, which leads to the breaking of this symmetry. This behavior is illustrated in Fig. 8, where results analogous to those in Fig. 6 are shown for $\theta_L = 45^\circ$. A detailed inspection clearly reveals the absence of the symmetry $\theta_i \rightarrow -\theta_i$. The symmetry breaking is particularly pronounced in the vicinity of the interference minima, where the polarization angle of the emitted x-ray becomes highly sensitive to both the x-ray energy ω_K and the angle θ_i .

IV. CONCLUSIONS

In this paper we have formulated a theory of laser-assisted electron-ion radiative recombination with homonuclear diatomic molecular ions in the presence of a linearly polarized assisting laser field. Our approach is based on the S -matrix

formalism combined with molecular orbitals represented by the linear combination of atomic orbitals. This framework enabled us to investigate how the multicenter structure and symmetry properties of molecular targets modify both the differential power spectra and the polarization properties of the emitted x-rays.

The presented results demonstrate that the emitted spectra exhibit pronounced signatures of two-center interference. In contrast to the corresponding atomic systems, molecular targets display characteristic interference minima whose positions depend sensitively on the internuclear distance, molecular orientation, and symmetry of the molecular orbital involved in the recombination process. We have shown that the interference structures can be interpreted directly in terms of the contributions originating from atomic orbitals with different parity properties. In particular, the minima positions are accurately reproduced by the analytical interference conditions derived from the recombination amplitude.

Our analysis further revealed that the interference pattern strongly depends on the molecular orientation angle and on the direction of the incident electron momentum. For molecular orbitals dominated by contributions with the same value of the symmetry parameter $s_{a\lambda}$, the interference minima remain well pronounced and follow the predicted analytical curves. When contributions with different symmetry properties interfere coherently, the minima become shifted and the angular symmetry properties of the spectra are modified. In addition, increasing the angle between the incident electron momentum and the assisting-field polarization direction leads to a gradual suppression of the interference minima, particularly in the low-energy part of the spectrum, while the high-energy minima remain comparatively robust.

We have also investigated the influence of imperfect molecular alignment by averaging the spectra over a Gaussian angular distribution. The obtained results show that the low-order interference minima survive even for experimentally realistic alignment conditions, whereas the higher-order minima become progressively blurred as the angular distribution broadens. This demonstrates that the characteristic molecular interference structures should remain observable even in partially aligned molecular ensembles.

Apart from the spectral properties, we have analyzed the polarization state of the emitted x-rays using the Stokes parameters. We have shown that for a linearly polarized assisting laser field the emitted x-rays remain linearly polarized for all investigated geometries, in contrast to the situation encountered in molecular high-order harmonic generation. The polarization direction, however, depends strongly on the emitted-photon energy and on the direction of the incident electron momentum. In particular, rapid variations of the polarization angle occur in the spectral regions where multiple stationary-phase solutions contribute coherently to the recombination process. Furthermore, we have shown that the symmetry properties of the polarization angle depend on the molecular orientation.

The obtained results demonstrate that laser-assisted radiative recombination with molecular ions provides direct access to the structural and symmetry properties of molecular targets. In particular, the interference minima and polarization characteristics of the emitted x-rays carry detailed information

about the internuclear distance, molecular orientation, and dominant orbital contributions forming the molecular state. The presented theory therefore establishes a basis for further investigations of strong-field-assisted recombination process in molecular systems and may be useful for future applications in molecular imaging and ultrafast x-ray spectroscopy.

DATA AVAILABILITY

The data are available from the authors upon reasonable request.

APPENDIX A: FORMAL DERIVATION OF THE S-MATRIX ELEMENT FOR LASER-ASSISTED RADIATIVE RECOMBINATION

The laser-assisted radiative recombination can be considered as a reaction and treated using the corresponding S-matrix formalism (see, for example, Chapter 16 in [80]). In the initial state we have an electron e^- , laser photons γ , and a molecular ion M_2^+ , while in the final state we have a neutral molecule M_2 and an x-ray, so our reaction is

$$e^- + n\gamma + M_2^+ \rightarrow M_2 + \text{x-ray}. \quad (\text{A1})$$

In the infinite past ($t' \rightarrow -\infty$) the electron moves towards the molecular ion in the presence of the laser field. We denote the corresponding Hamiltonian by $H_a(t')$ and call this channel a (arrangement channel [80]). When the (laser-dressed) electron and the molecular ion are close enough to each other, the recombination may occur (for example, at the moment t'') and the corresponding (total) Hamiltonian can be written as $H(t'') = H_a(t'') + H'_a(t'')$, where $H'_a(t'')$ corresponds to the interaction with the x-ray [see Eq. (2) for our case of diatomic molecule]. The state $|\Psi^{(+)}(t'')\rangle$ which satisfies the Schrödinger equation with the total Hamiltonian $H(t'')$, i.e., $[i\partial/\partial t'' - H(t'')]\Psi^{(+)}(t'') = 0$, is related to the initial state $|\Psi_a(t'')\rangle$ [which corresponds to the Hamiltonian $H_a(t'')$] by the relation

$$|\Psi^{(+)}(t'')\rangle = |\Psi_a(t'')\rangle - i \int_{t'}^{t''} d\tau U_a(t'', \tau) H'_a(\tau) |\Psi^{(+)}(\tau)\rangle. \quad (\text{A2})$$

Here $U_a(t'', \tau)$ is the evolution operator corresponding to the Hamiltonian $H_a(t'')$. The relation between the state $|\Psi^{(+)}(t'')\rangle$ and the initial state $|\Psi_a(t'')\rangle$ can also be written as [80] $|\Psi^{(+)}(t'')\rangle = \Omega_+ |\Psi_a(t'')\rangle$, with the Møller operator $\Omega_+ = \lim_{t' \rightarrow -\infty} U^\dagger(t', t'') U_a(t', t'')$ where $U(t', t'')$ is the evolution operator corresponding to the total Hamiltonian.

At the final time $t \rightarrow \infty$ we have neutral molecule M_2 and an x-ray. Let us call this channel b and denote the corresponding Hamiltonian by $H = H_b + H'_b$. After the recombination the neutral molecule is formed and it does not interact with the emitted x-ray, so we have $H'_b = 0$ and $H_a = H_b$. Formally, one can write that the corresponding state $|\Psi^{(-)}(t'')\rangle$ is related to the final state $|\Psi_b(t'')\rangle$ by

$$|\Psi^{(-)}(t'')\rangle = |\Psi_b(t'')\rangle + i \int_{t''}^t d\tau U_b(t, \tau) H'_b(\tau) |\Psi^{(-)}(\tau)\rangle, \quad (\text{A3})$$

or using the Møller operator $|\Psi_b(t'')\rangle = \Omega_-^\dagger |\Psi^{(-)}(t'')\rangle$, $\Omega_- = \lim_{t \rightarrow \infty} U^\dagger(t, t'') U_b(t, t'')$.

The S -matrix element for the transition from the initial state to the final state is [80]

$$S_{fi} = \langle \Psi_b(t'') | \Omega_-^\dagger \Omega_+ | \Psi_a(t'') \rangle, \quad (\text{A4})$$

which can also be written as

$$S_{fi} = \lim_{t \rightarrow \infty} \langle \Psi_b(t) | \Psi^{(+)}(t) \rangle. \quad (\text{A5})$$

Using Eq. (A2), orthogonality of the states a and b , and the fact that $H'_b = 0$, $H_a = H_b$, the S -matrix element becomes

$$S_{fi} = -i \int_{-\infty}^{\infty} d\tau \langle \Psi_b(\tau) | H'_a(\tau) | \Psi_a(\tau) \rangle. \quad (\text{A6})$$

This result is our starting point in theoretical analysis of the laser-assisted radiative recombination. The presented formalism can be used for other laser-assisted or laser-induced reactions.

APPENDIX B: S -MATRIX ELEMENT FOR ELECTRON-ION RADIATIVE RECOMBINATION

For our Hamiltonian (2), the S -matrix element S_{fi} can be written as a sum of the contributions corresponding to the electron and two atomic centers, i.e., $S_{fi} = S_{fi,e} + S_{fi,e_A} + S_{fi,e_B}$. The contribution $S_{fi,e}$ can further be decomposed as $S_{fi,e} = S_{fi,e}^r + S_{fi,e}^{R_{cm}}$, where

$$S_{fi,e}^r = -i \int_{-\infty}^{\infty} d\tau C_e(\tau) \mathcal{M}_{R_{cm}}^r(\tau) \mathcal{M}_r^r(\tau), \quad (\text{B1})$$

with $C_e(\tau) = ie c_{\mathbf{K}} e^{i\omega_{\mathbf{K}}\tau}$ and

$$\begin{aligned} \mathcal{M}_{R_{cm}}^r(\tau) &= \langle \Xi_b(\mathbf{R}_{cm}; \tau) | e^{-i\mathbf{K} \cdot \mathbf{R}_{cm}} | \Xi_a(\mathbf{R}_{cm}; \tau) \rangle \\ &= \delta(\mathbf{P}_{cm,b} - \mathbf{P}_{cm,a} + \mathbf{K}), \end{aligned} \quad (\text{B2})$$

$$\mathcal{M}_r^r(\tau) = \langle \Phi_b(\mathbf{r}, \mathbf{R}; \tau) | \hat{\mathbf{e}}_{\mathbf{K}}^* \cdot \mathbf{r} e^{-i\mathbf{K} \cdot \mathbf{r}} | \Phi_a(\mathbf{r}, \mathbf{R}; \tau) \rangle. \quad (\text{B3})$$

Furthermore, the contribution $S_{fi,e}^{R_{cm}}$ is

$$S_{fi,e}^{R_{cm}} = -i \int_{-\infty}^{\infty} d\tau C_e(\tau) \mathcal{M}_{R_{cm}}^{R_{cm}}(\tau) \mathcal{M}_r^{R_{cm}}(\tau), \quad (\text{B4})$$

with

$$\begin{aligned} \mathcal{M}_{R_{cm}}^{R_{cm}}(\tau) &= \langle \Xi_b(\mathbf{R}_{cm}; \tau) | \hat{\mathbf{e}}_{\mathbf{K}}^* \cdot \mathbf{R}_{cm} e^{-i\mathbf{K} \cdot \mathbf{R}_{cm}} | \Xi_a(\mathbf{R}_{cm}; \tau) \rangle \\ &= i \hat{\mathbf{e}}_{\mathbf{K}} \cdot \nabla_{\mathbf{Q}} \delta(\mathbf{Q}), \end{aligned} \quad (\text{B5})$$

where $\mathbf{Q} = \mathbf{P}_{cm,b} - \mathbf{P}_{cm,a} + \mathbf{K}$, and

$$\mathcal{M}_r^{R_{cm}}(\tau) = \langle \Phi_b(\mathbf{r}, \mathbf{R}; \tau) | e^{-i\mathbf{K} \cdot \mathbf{r}} | \Phi_a(\mathbf{r}, \mathbf{R}; \tau) \rangle. \quad (\text{B6})$$

The term $\mathcal{M}_{R_{cm}}^{R_{cm}}(\tau)$ is proportional to the derivation of the δ -function and it describes sensitivity to spatial variation of the center-of-mass wave packet, i.e., how sharply localized the molecule is in the momentum space. For realistic systems, the center-of-mass motion is slow and the term $\mathcal{M}_{R_{cm}}^{R_{cm}}(\tau)$ can be neglected thus leading to $S_{fi,e} \approx S_{fi,e}^r$.

Analogous analysis can be performed for the contributions S_{fi,e_j} , $j = A, B$. These contributions can be written as $S_{fi,e_j} = S_{fi,e_j}^R + S_{fi,e_j}^{R_{cm}}$ where now $\mathbf{R}$ is the relevant relative coordinate. The contribution $S_{fi,e_j}^{R_{cm}}$ leads to

$$S_{fi,e_j}^{R_{cm}} = -i \int_{-\infty}^{\infty} d\tau C_j(\tau) \mathcal{M}_{R_{cm}}^{R_{cm}}(\tau) \mathcal{M}_{j,R}^{R_{cm}}(\tau), \quad j = A, B. \quad (\text{B7})$$

Here $C_j(\tau) = ie_j c_{\mathbf{K}} e^{i\omega_{\mathbf{K}}\tau}$ and

$$\mathcal{M}_{A/B,R}^{R_{cm}}(\tau) = \langle \Phi_b(\mathbf{r}, \mathbf{R}; \tau) | e^{\pm i\mathbf{K} \cdot \mathbf{R}_{B/A}/M_{AB}} | \Phi_a(\mathbf{r}, \mathbf{R}; \tau) \rangle, \quad (\text{B8})$$

with the upper (lower) sign corresponding to $\mathcal{M}_{A,R}^{R_{cm}}(\tau)$ [$\mathcal{M}_{B,R}^{R_{cm}}(\tau)$]. Moreover, $\mathcal{M}_{R_{cm}}^{R_{cm}}(\tau)$ is the same as for the contribution $S_{fi,e}$. Consequently, the term $S_{fi,e_j}^{R_{cm}}$, $j = A, B$, can be neglected because it is proportional to the derivation of the δ -function. On the other hand, the contribution S_{fi,e_j}^R is

$$S_{fi,e_j}^R = -i \int_{-\infty}^{\infty} d\tau C_j(\tau) \mathcal{M}_{R_{cm}}^R(\tau) \mathcal{M}_{j,R}^R(\tau), \quad j = A, B, \quad (\text{B9})$$

where $\mathcal{M}_{R_{cm}}^R(\tau)$ is the same as $\mathcal{M}_{R_{cm}}^r(\tau)$ which appears in $S_{fi,e}$ contribution [see Eq. (B2)] and it thus leads to the δ -function corresponding to the momentum conservation. On the other hand,

$$\begin{aligned} \mathcal{M}_{A/B,R}^R(\tau) &= \langle \Phi_b(\mathbf{r}, \mathbf{R}; \tau) | m_{B/A}/M_{AB} \hat{\mathbf{e}}_{\mathbf{K}} \cdot \mathbf{R} \\ &\times e^{\pm i\mathbf{K} \cdot \mathbf{R}_{B/A}/M_{AB}} | \Phi_a(\mathbf{r}, \mathbf{R}; \tau) \rangle. \end{aligned} \quad (\text{B10})$$

Within the Born-Oppenheimer approximation, $\mathbf{R} = \mathbf{R}_0$ and matrix elements $\mathcal{M}_{j,R}^R(\tau)$, $j = A, B$, are zero due to the orthogonality of the initial and final state. Consequently, $S_{fi,e_j}^R = 0$ and thus also $S_{fi,e_j} = 0$, $j = A, B$. As a result, within the Born-Oppenheimer approximation, only the term $S_{fi,e}^r$ is relevant and has to be taken into consideration. In the main part of the paper, instead of using the notation $\mathcal{M}_r^r(\tau)$ we have used $\mathcal{M}(\tau)$, while instead of $C_e(\tau)$ we used $C(\tau)$.

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