Spin-dependent quantum theory of high-order above-threshold ionization

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The strong-field-approximation theory of high-order above-threshold ionization of atoms is generalized to include the electron spin. The obtained rescattering amplitude consists of a direct and exchange part. On the examples of excited He atoms as well as Li^+ and Be^{++} ions, it is shown that the interference of these two amplitudes leads to an observable difference between the photoelectron momentum distributions corresponding to different initial spin states: Pronounced minima appear for singlet states, which are absent for triplet states.

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I. INTRODUCTION

The ionization dynamics of atoms by a strong laser field is usually described by using the nonrelativistic Schrödinger equation and neglecting the influence of the electron spin. It has been assumed that this is justified for optical frequencies and laser intensities much lower than 3.5×10^{16} W/cm^2. For higher intensities, or longer wavelengths, the magnetic field component becomes important and one should use the Pauli or even the Dirac equation [1,2]. High-order atomic processes in strong fields are commonly described using the three-step model (see Refs. [3,4] and references therein): The electron, liberated in the first step, moves in the laser field and may be driven back to the atomic core in the second step. Finally, in the third step, the electron may elastically scatter off the core and leave towards the detector with a much higher energy than it could acquire in the first step alone. This is the so-called high-order above-threshold ionization (HATI) process [5,6]. The electron may also recombine to the ground state, emitting a high-energy photon in the high-order harmonic generation (HHG) process [7]. One may expect that, with increasing laser intensity, the energy of the emitted HATI electrons or HHG photons increases. However, due to the magnetic component of the Lorentz force, the returning electron has a drift momentum in the direction of propagation of the laser field, which drastically decreases the significance of the rescattering process [3,8]. Therefore, rescattering is only important for nonrelativistic electron energies. In this case, the probability of spin flips is small [9]. If the spin is not important, the rescattering effect can be treated using the simpler relativistic Klein-Gordon equation instead of the Dirac equation [10].

On the other hand, spin-polarized electrons are important in many areas of physics [11,12]. Spin dynamics in relativistic strong-field ionization has been analyzed in Ref. [13] for circularly and in Ref. [14] for linearly polarized laser fields. It has been proposed to use ionization with strong (but still nonrelativistic) circularly polarized laser fields for the creation of spin-polarized electrons [15]. This has been realized in a recent experiment [16]. However, since the probability of the electron returning to the core decreases with increasing ellipticity of the laser polarization, there is no rescattering for circularly polarized fields. This problem can be solved by using a so-called bicircular field (consisting of two coplanar counter-rotating circularly polarized fields with different frequencies), which enables rescattering [17]. In this case, the spin-dependent effects are due to the spin-orbit interaction.

Coulombic forces do not act directly on the spins of electrons. In addition to the mentioned spin-orbit coupling, spin-dependent effects in collisions may be facilitated via the “Pauli force,” i.e., the requirement that wave functions of identical fermions are antisymmetric. In this paper, we will show that spin effects in strong-field ionization can be important if the Pauli force is taken into account, even in the nonrelativistic regime. Using this, we will extend the semiclassical results from Ref. [18]. It is known that for the scattering of identical particles the scattering amplitude consists of the direct and exchange term [19]. The relative sign in the sum of these two terms depends on the spin state of the scattering particles (e.g., singlet or triplet). In calculations of the scattering cross sections for laser-assisted electron-atom scattering, the influence of the exchange effect was taken into account in Refs. [20–22]. Here, we consider spin-dependent rescattering in the HATI process. To this end, we reformulate and generalize the strong-field approximation (SFA) theory of HATI to include the spin-wave functions. In the obtained result, the direct and exchange rescattering amplitudes are explicitly separated. We illustrate our theory with examples of strong-field ionization of excited states of He, Li^+, as well as Be^{++} and find that considerable differences in the rescattering process are to be expected, depending on the spin state of the returning electron and residual ion. Specifically, ionization from singlet states leads to distinct minima in the photoelectron momentum distributions (PEMDs), which are absent for triplet states. The investigated effect could potentially be exploited

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TABLE I. Total spin S, charge of the nucleus Z, ionization potential I_p^S, and the effective charge α_S of the atomic core for He, Li^+, and Be^{++} for singlet and triplet $1s2s$ states.

Atom/ion	S	Z	I_p^S (eV)	α_S (a.u.)
He ($^1S_0$)	0	2	3.97	1.40
He ($^3S_1$)	1	2	4.77	1.552
Li^+ ($^1S_0$)	0	3	14.72	2.40
Li^+ ($^3S_1$)	1	3	16.62	2.571
Be^{++} ($^1S_0$)	0	4	32.25	3.40
Be^{++} ($^3S_1$)	1	4	35.31	3.578

in the future to measure spin dynamics during the strong-field ionization process.

II. THEORY

For our theoretical description, we assume initially helium, or a heliumlike ion, in one of the excited $1s2s\ ^{1,3}S$ states, that is in either the singlet ($^1S_0$, $S = M_S = 0$, ionization potential I_p^0) or triplet ($^3S_1$, $S = 1$, $M_S = -1,0,1$, I_p^1) state (see Table I). The corresponding wave functions are

$$|\Psi_{iSM_S}(\mathbf{r}_1,\mathbf{r}_2,t)\rangle = \psi_{iS}(\mathbf{r}_1,\mathbf{r}_2,t)|SM_S\rangle = \frac{e^{iI_p^S t}}{\sqrt{2}}[u_{1s}(\mathbf{r}_1)u_{2s}(\mathbf{r}_2) + (-1)^S u_{1s}(\mathbf{r}_2)u_{2s}(\mathbf{r}_1)]|SM_S\rangle, \quad (1)$$

where the spin-wave functions are $|11\rangle = |\uparrow\uparrow\rangle$, $|1,-1\rangle = |\downarrow\downarrow\rangle$, $|S0\rangle = [|\uparrow\downarrow\rangle - (-1)^S|\downarrow\uparrow\rangle]/\sqrt{2}$, $S = 0,1$. Since the difference in the ionization potential of the ground and excited state is very large, the ionization is possible only from the state u_{2s}. In the SFA, the final state after ionization by a strong laser field, $|\Psi_{\mathbf{p}S'M_S'}(\mathbf{r}_1,\mathbf{r}_2,t)\rangle$, has the same form as (1) but with $u_{2s}e^{iI_p^S t}$ replaced with the Volkov state $\chi_{\mathbf{p}}(t)$, which describes the electron in the laser field only. The variable $\mathbf{p}$ describes the asymptotic momentum of the emitted electron. Since the initial spin projection M_S is unknown, we should average over all values of M_S. Moreover, if the spin of the emitted electron is not measured, we have to sum over all $S'M_S'$. The differential ionization probability for emission of an electron with energy $E_{\mathbf{p}} = \mathbf{p}^2/2$ (atomic units are used throughout, unless otherwise stated) into the solid angle element $d\Omega_{\mathbf{p}}$ is then given by

$$w_S(\mathbf{p}) = \frac{|\mathbf{p}|}{2S+1}\sum_{M_S}\sum_{S'M_S'}|M_{\mathbf{p}S'M_S'iSM_S}|^2, \quad (2)$$

where $M_{\mathbf{p}S'M_S'iSM_S}(t,t')$ is the probability amplitude with initial time $t' \to -\infty$ and final time $t \to \infty$.

The total two-electron Hamiltonian of our system is

$$H(t) = \frac{\hat{\mathbf{p}}_1^2}{2} + \frac{\hat{\mathbf{p}}_2^2}{2} - \frac{Z}{r_1} - \frac{Z}{r_2} + \frac{1}{r_{12}} + (\mathbf{r}_1 + \mathbf{r}_2)\cdot\mathbf{E}(t), \quad (3)$$

where $\hat{\mathbf{p}}_j = -i\partial/\partial\mathbf{r}_j$, $r_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$, and $\mathbf{r}_j \cdot \mathbf{E}(t)$ is the interaction of the jth electron with the laser electric field $\mathbf{E}(t)$ in length gauge and dipole approximation. Since the Hamiltonian $H(t)$ does not contain a spin-dependent interaction, the spin state, e.g., singlet, remains fixed during all steps of the interaction. In Eq. (2) the averaging and summation over the spin indices gives $(2S+1)^{-1}\sum_{M_S}\sum_{S'}\sum_{M_S'}\delta_{S',S}\delta_{M_S',M_S} = 1$, so that the probability amplitude depends only on the total electron spin. We denote it by $M_{\mathbf{p}S}$, where the index i is also omitted since the initial state is fixed.

The ionization probability amplitude is given by [3,4]

$$M_{\mathbf{p}S}(t,t') = -i\int_{t'}^{t} dt_0 \int d\mathbf{r}_1 \int d\mathbf{r}_2 \phi_{\mathbf{p}S}^{(-)*}(\mathbf{r}_1,\mathbf{r}_2,t_0) \times(\mathbf{r}_1+\mathbf{r}_2)\cdot\mathbf{E}(t_0)\psi_{iS}(\mathbf{r}_1,\mathbf{r}_2,t_0), \quad (4)$$

where the two-electron antisymmetrized scattering state is

$$\phi_{\mathbf{p}S}^{(-)}(\mathbf{r}_1,\mathbf{r}_2,t_0) = \frac{1}{\sqrt{2}}[1+(-1)^S X_{12}]\Big[u_{1s}(\mathbf{r}_1)\chi_{\mathbf{p}}(\mathbf{r}_2,t_0) + i\int_{t_0}^{t} dt_1 \int d\mathbf{r}_1' \int d\mathbf{r}_2' \langle\mathbf{r}_1,\mathbf{r}_2|U(t_0,t_1)|\mathbf{r}_1',\mathbf{r}_2'\rangle \times\left(-\frac{Z}{r_2'}+\frac{1}{r_{12}'}\right)u_{1s}(\mathbf{r}_1')\chi_{\mathbf{p}}(\mathbf{r}_2',t_1)\Big], \quad (5)$$

with the Volkov state in the length gauge and dipole approximation given by $|\chi_{\mathbf{k}}(t)\rangle = |\mathbf{k}+\mathbf{A}(t)\rangle\exp[-iS_{\mathbf{k}}(t)]$, $\mathbf{E}(t) = -d\mathbf{A}(t)/dt$, $dS_{\mathbf{k}}(t)/dt = [\mathbf{k}+\mathbf{A}(t)]^2/2$, and $|\mathbf{q}\rangle$ is a plane-wave ket vector such that $\langle\mathbf{r}|\mathbf{q}\rangle = (2\pi)^{-3/2}\exp(i\mathbf{q}\cdot\mathbf{r})$. The operator X_{12} exchanges the electron coordinates $\mathbf{r}_1$ and $\mathbf{r}_2$ and $-Z/r_j + 1/r_{12}$ is the interaction of the jth (emitted) electron with the nucleus and the second (bound) electron. Here, $U(t,t_1)$ is the total time-evolution operator, which corresponds to the Hamiltonian $H(t)$.

Introducing only the first row term of Eq. (5) into (4), we obtain the SFA ionization probability amplitude

$$M_{\mathbf{p}S}^{\mathrm{SFA}}(t,t') = -i\int_{t'}^{t} dt_0 \langle\mathbf{p}+\mathbf{A}(t_0)|\mathbf{r}\cdot\mathbf{E}(t_0)|u_{2s}\rangle e^{i[I_p^S t_0 + S_{\mathbf{p}}(t_0)]}. \quad (6)$$

In deriving this result we used the orthogonality of the continuum and bound states and the relations $\langle u_{js}|u_{is}\rangle = \delta_{i,j}$ and $\langle u_{js}|\mathbf{r}|u_{js}\rangle = \mathbf{0}$.

In order to derive the first-order correction to the result (6), we introduce the remaining terms of Eq. (5) into (4). We use the single-active-electron approximation in the sense that only the emitted jth electron, liberated from the state $u_{2s}(\mathbf{r}_j)$ by the interaction $\mathbf{r}_j\cdot\mathbf{E}(t)$, is propagated in the laser field while the second electron remains bound. Furthermore, in the SFA [23] we approximate $\langle\mathbf{r}_1,\mathbf{r}_2|U(t_0,t_1)|\mathbf{r}_1',\mathbf{r}_2'\rangle$ with the product of one-electron Volkov propagators (for continuum states) and unit operators (for bound electron states), i.e., by

$$\sum_{i,j=1,2\ (i\neq j)} \delta(\mathbf{r}_i - \mathbf{r}_i')\int d\mathbf{k}\chi_{\mathbf{k}}(\mathbf{r}_j,t_0)\chi_{\mathbf{k}}^*(\mathbf{r}_j',t_1). \quad (7)$$

After a lengthy calculation, taking into account that only the combinations with $\chi_{\mathbf{k}}^*(\mathbf{r}_j,t_0)\mathbf{r}_j u_{2s}(\mathbf{r}_j)$ contribute, we obtain the rescattering or improved SFA (ISFA) ionization probability amplitude

$$M_{\mathbf{p}S}^{\mathrm{ISFA}}(t,t') = (-i)^2\int_{t'}^{t} dt_0 \int_{t_0}^{t} dt_1 e^{iS_{\mathbf{p}}(t_1)} \times\int d\mathbf{k}e^{-iS_{\mathbf{k}}(t_1)}[\mathcal{T}_{\mathrm{di}} + (-1)^S\mathcal{T}_{\mathrm{ex}}]e^{iS_{\mathbf{k}}(t_0)} \times\langle\mathbf{k}+\mathbf{A}(t_0)|\mathbf{r}\cdot\mathbf{E}(t_0)|u_{2s}\rangle e^{iI_p^S t_0}, \quad (8)$$

where the direct and exchange rescattering T-matrix elements are given by

$$\mathcal{T}_{\rm di} = \int \frac{d\mathbf{r}_2 e^{i(\mathbf{k}-\mathbf{p})\cdot\mathbf{r}_2}}{(2\pi)^3}\left[-\frac{Z}{r_2} + \int d\mathbf{r}_1 \frac{|u_{1s}(\mathbf{r}_1)|^2}{r_{12}}\right], \quad (9)$$

$$\mathcal{T}_{\rm ex} = \int \frac{d\mathbf{r}_1 d\mathbf{r}_2 e^{i(\mathbf{k}'\cdot\mathbf{r}_1 - \mathbf{p}'\cdot\mathbf{r}_2)}}{(2\pi)^3} u_{1s}^*(\mathbf{r}_1)\left(\frac{1}{r_{12}} - \frac{Z}{r_2}\right) u_{1s}(\mathbf{r}_2), \quad (10)$$

with $\mathbf{p}' = \mathbf{p} + \mathbf{A}(t_1)$ and $\mathbf{k}' = \mathbf{k} + \mathbf{A}(t_1)$ (t_1 is the rescattering time).

For He and He-like ions the state u_{1s} is given by $u_{1s}(\mathbf{r}) = \sqrt{Z^3/\pi}\, e^{-Zr}$. Using this, for the effective potential in the direct T-matrix element we obtain

$$-\frac{Z}{r} + \int d\mathbf{r}_1 \frac{|u_{1s}(\mathbf{r}_1)|^2}{|\mathbf{r} - \mathbf{r}_1|} = \frac{1-Z}{r} - \left(\frac{1}{r} + Z\right)e^{-2Zr}, \quad (11)$$

so that, with the notation $\mathbf{q} = \mathbf{p} - \mathbf{k}$, we get

$$\mathcal{T}_{\rm di} = -\frac{1}{2\pi^2}\left[\frac{Z-1}{\mathbf{q}^2} + \frac{\mathbf{q}^2 + 8Z^2}{(\mathbf{q}^2 + 4Z^2)^2}\right]. \quad (12)$$

In order to avoid the Coulomb divergences for small $\mathbf{q}^2$, we introduce the screening parameter μ such that in Eq. (11) $-1/r \to -e^{-\mu r}/r$ and in Eq. (12) $1/\mathbf{q}^2 \to 1/(\mathbf{q}^2 + \mu^2)$.

Taking into account that the effective shielding charges are different for singlet and triplet states, we obtain the wave function of the excited state [24],

$$u_{2s}^S(\mathbf{r}) = C_S\left(1 - \frac{\alpha_S r}{2}\right)e^{-\alpha_S r/2}, \quad C_S^2 = \frac{\alpha_S^3}{8\pi\left(1 - O_S^2\right)}, \quad (13)$$

where the overlapping integral is $O_S = \langle u_{1s}|u_{2s}\rangle = (2x)^{3/2}(x-1)/(x+1/2)^4$, $x = Z/\alpha_S$, and the charge Z and parameters α_S are given in Table I. For the dipole matrix element entering Eq. (6), we obtain

$$\langle \mathbf{q}|\mathbf{r}|u_{2s}^S\rangle = -i\frac{16\pi C_S\alpha_S}{(2\pi)^{3/2}} \frac{2\mathbf{q}^2 - \alpha_S^2}{\left(\mathbf{q}^2 + \alpha_S^2/4\right)^4}\mathbf{q}. \quad (14)$$

Expression (10) corresponds to the first Born approximation. A better approximation is to neglect the interaction with the core $(-Z/r_2)$ and to take into account only the electron-electron interaction $1/r_{12}$. In this case, an approximate result for the exchange channel is [19,25]

$$\mathcal{T}_{\rm ex} = \frac{8Z^4}{\pi^2}\frac{1}{\mathbf{k}'^2(\mathbf{q}^2 + 4Z^2)^2}. \quad (15)$$

Again, a screening parameter μ' can be introduced such that in Eq. (15) $\mathbf{k}'^2 \to \mathbf{k}'^2 + \mu'^2$.

III. NUMERICAL RESULTS

Based on these derivations, we will calculate PEMDs for singlet and triplet rescattering for three different target materials and suitable laser parameters. The five-dimensional integral in the ISFA matrix element (8) is approximately solved using the quantum-orbit theory [26]. We use both the forward- and backward-scattering quantum orbits. The integral over the intermediate electron momenta is solved using the saddle-point method, which gives $\mathbf{k} \to \mathbf{k}_s = -\int_{t_0}^{t_1}\mathbf{A}(t)dt/(t_1 - t_0)$, while the integrals over ionization time t_0 and rescattering time t_1 are calculated using the uniform approximation for the backward-scattering quantum orbits and the saddle-point method for the forward-scattering orbits. In this case, the rescattering T-matrix elements are on shell, i.e., $\mathbf{p}'^2/2 = \mathbf{k}'^2/2$.

To illustrate the influence of spin on the rescattering process, we present numerical results for HATI of excited states having $S = 0$ (left panels) and $S = 1$ (middle panels) in Fig. 1, using a linearly polarized laser field with amplitude $E(t) = E_0 \sin\omega t$ and angular frequency ω. We employ screening parameters $\mu = \mu' = 0.3$ a.u. (our results do not depend on this specific choice). Only the rescattering amplitude is taken into account. We estimated that the intensity used in our calculation is below saturation intensity. The presented PEMDs are typical for HATI by a linearly polarized laser field: They exhibit a cutoff at $10U_p + 0.538I_p$ along the $p_\parallel$ axis [6,27] and off-axis low-energy structures [28] [$U_p = E_0^2/(4\omega^2) \propto I\lambda^2$ is the ponderomotive potential].

For the laser parameters under investigation in this paper (see Fig. 1), effects of the laser magnetic field can still be safely neglected [29]. Specifically, we assumed the spin state to remain fixed during the electrons' motion in the continuum. The typical time scale from emission of the electron to the moment of rescattering is given by the laser frequency. We estimate the probability of a spin flip, due to the laser magnetic field, by using the Larmor frequency [18,30] $\omega_P = g_s\mu_B|\vec{B}|$ as $\omega_P/\omega \sim \sqrt{U_p}/c$. With the parameters employed for He, Li^+, and Be^{++} this yields $\omega_P/\omega \approx 4\times10^{-3}$, 5×10^{-3}, and 9×10^{-3}, respectively. We are therefore justified in neglecting spin-flip probabilities during the continuum motion. Spin flips during the rescattering process itself are also unlikely to occur. The maximum return energies of the electrons considered here are ≈ 120 eV. At such low impact energies, the main spin-dependent effect is the discussed exchange interaction [31]. The spin-orbit interaction, which could in theory facilitate spin flips, is only relevant in the relativistic regime, e.g., by scattering off targets with a high nuclear charge.

Analyzing the numerical results, clear differences between singlet and triplet rescattering can be identified. The main difference between the results shown in the left and middle panels of Fig. 1 is the appearance of minima in the off-axis direction, which are clearly visible for the $S = 0$ state, while they are absent for the $S = 1$ state. For a better visualization of the effect, we plot the normalized difference map (right column of Fig. 1), i.e.,

$$D(\mathbf{p}) = \frac{w_{S=0}(\mathbf{p}) - w_{S=1}(\mathbf{p})}{w_{S=0}(\mathbf{p}) + w_{S=1}(\mathbf{p})}. \quad (16)$$

These large differences are explained by the minima in the on-shell rescattering T-matrix element. Namely, from Eqs. (12) and (15), it follows that $\mathcal{T}_{\rm ex} > 0$ while $\mathcal{T}_{\rm di} < 0$. Since these amplitudes sum with the relative factor $(-1)^S$ in Eq. (8), the minima can appear only for $S = 0$. Thus, the effect is a direct consequence of different rescattering cross sections for singlet and triplet states. A more detailed discussion of the cross sections and their properties is given in Ref. [18].

The largest possible difference of -1 is located at the position of the minima in the right panels of Fig. 1. At the cutoff, the triplet yield always dominates the singlet yield: The differences are $D(\mathbf{p}) \approx -0.4, -0.65, -0.7$ for He, Li^+,

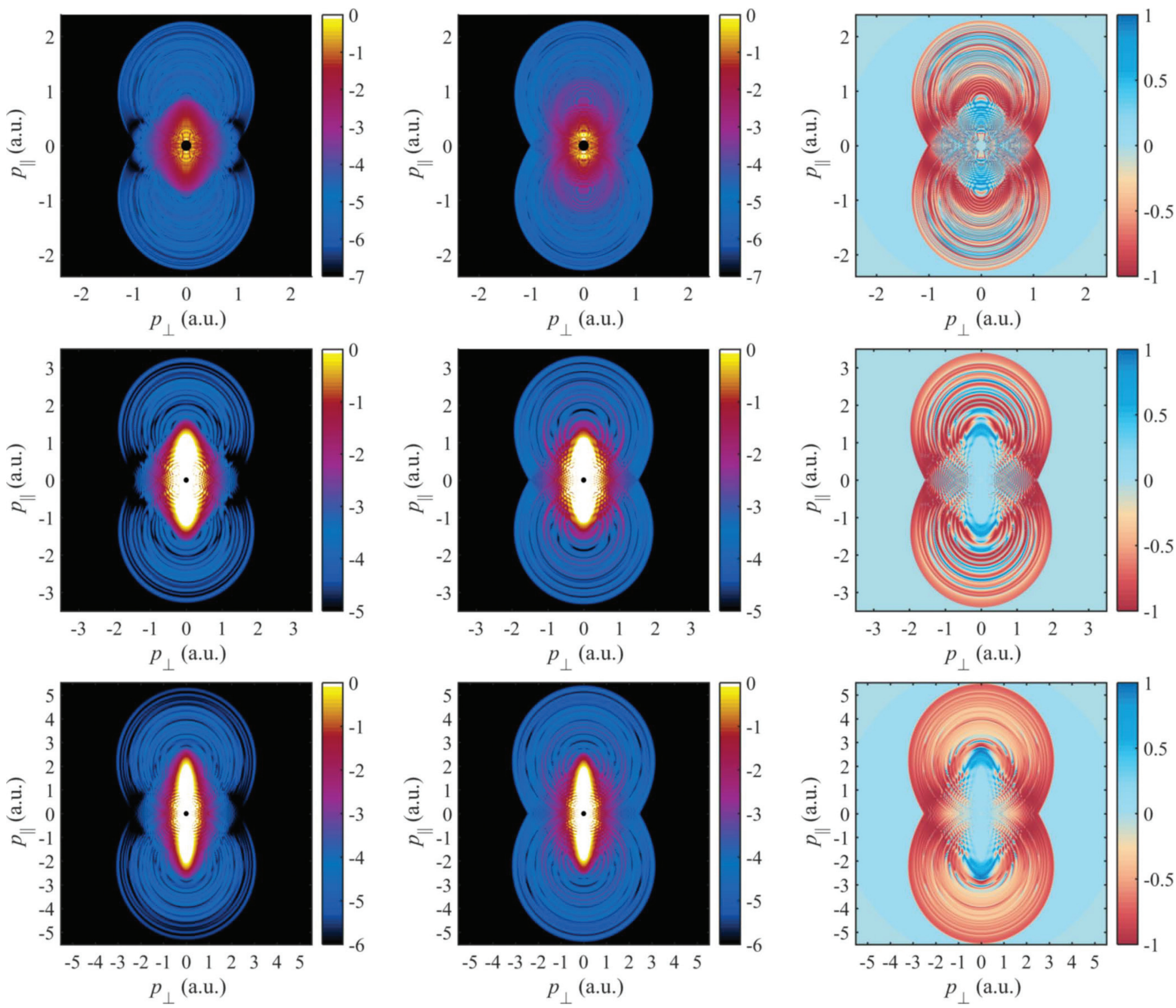

FIG. 1. The logarithm of the ionization rate, in arbitrary units, for singlet ($S = 0$, left) and triplet ($S = 1$, middle) for excited He (first row, $I = 7.65 \times 10^{11}$ W/cm^2, $\lambda = 9700$ nm), Li^+ (second row, $I = 4.55 \times 10^{13}$ W/cm^2, $\lambda = 1800$ nm), and Be^{++} (third row, $I = 6.2 \times 10^{14}$ W/cm^2, $\lambda = 800$ nm). In each singlet and triplet map the total yield was normalized to 1. The right column shows the normalized difference map $D(\mathbf{p})$ [see Eq. (16)].

and Be^{++}, respectively. For lower energies, the difference oscillates between positive and negative values.

Experimental verification of the proposed effect may prove to be challenging (for details, see Ref. [18]). However, if separate beams of excited singlet and triplet states could be obtained, we predict that one could observe minima in the PEMDs for the singlet states and for larger electron angles with respect to the laser polarization axis. Such minima do not appear for the triplet state. Thus, the fully quantum mechanical ISFA confirms the results from Ref. [18], which relied on classical trajectories. Remarkably, the minima appear at the same positions in the PEMD. Should a system, starting in the singlet state, undergo a spin flip during the laser interaction, e.g., by entering the relativistic regime, the strength of the minima would be changed, serving as a probe for spin dynamics.

IV. CONCLUSION

In summary, the influence of the electron spin on the rescattering step of the HATI process was ignored in previous investigations of strong-field ionization. We have *generalized* the SFA theory of HATI to include the initial spin states. In this case, the rescattering amplitude contains a direct and exchange part. Using examples of excited He, Li^+, and Be^{++}, we have shown that the interference of these two amplitudes may lead to observable differences between the photoelectron spectra corresponding to different initial spin states. This difference manifests most dominantly in the form of pronounced minima in the PEMDs which appear for singlet states, yet for triplet states such minima are absent. This effect is strong for a wide range of laser and excited atom and ion parameters. We have illustrated this using typical examples of wavelengths: from midinfrared (9700 nm [32]), via an optical parametric amplifier (1800 nm [28]) to the standard Ti:Sa laser (800 nm). Furthermore, the introduced spin-dependent quantum theory can be generalized to other high-order strong-field processes. For example, spin-dependent effects can be important in ionizing collisions during nonsequential double or multiple ionization [33–35].

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