Numerical verification of the theory of nonadiabatic tunnel ionization in strong circularly polarized laser fields

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Abstract. We verify the theory of nonadiabatic ionization of degenerate valence $p_{\pm}$ orbitals in strong circularly polarized laser fields by solving numerically the two-dimensional time-dependent Schrödinger equation for an effective one-electron potential of neon. The numerically calculated ionization ratios of the $p_-$ and $p_+$ orbitals agree well with the theoretical results, i.e. the counter-rotating electron tunnels preferably. However, for strong laser pulses and low laser frequencies, the adiabatic laser-dressed orbitals play an important role. In a Floquet treatment of a three-level model we find that in this regime the ionization ratio of initial $p_-$ and $p_+$ orbitals depends crucially on the orbital energy order of valence $s$ and $p_{\pm}$ orbitals. We also show that the emission angles of valence $p_-$ and $p_+$ electrons are different and should be observable in attoclock experiments.

Keywords: numerical solution of the time-dependent Schrödinger equation, nonadiabatic tunnel ionization, strong circularly polarized laser fields, laser-dressed orbitals


1. Introduction

Ionization in strong infrared (IR) laser fields is one of the fundamental non-linear processes in attosecond physics of atoms, molecules and nanostructures [1]. It can be controlled by varying the intensity, frequency, carrier-envelope phase and polarization of a few-cycle strong laser pulse [2]. In the case of linear polarization, the electron can travel back to the parent ion, leading to additional physical phenomena such as high harmonic generation [3, 4], non-sequential double ionization [5] and photoelectron scattering [6,7]. These recollision phenomena are absent in circular polarization, because the photoelectron cannot travel back to the parent ion. Assuming that there is no resonance-enhanced multiphoton ionization (REMPI) [8], the direct ionization is the only possible flavour of ionization in strong circularly polarized laser fields. The
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The experimental availability of femtosecond IR circularly polarized laser pulses and the existing theories of tunnel ionization in strong laser fields give us opportunities to compare experimental and theoretical results directly. Examples are the experimental and theoretical results of lateral widths of the electron momentum distribution [9–12] and the ratios of the ionization rates of valence $p_\pm$ orbitals of noble gas atoms [13–16]. Here the plus/minus sign denotes the magnetic (azimuthal) quantum number $m$. Attoclock experiments using near-circularly polarized pulses measure the offset angles of the emitted photoelectron at the detector [17–20]. It is one of the hottest topics in attosecond physics at the present day since it is connected to the determination of the time delay of direct ionization with attosecond resolution. For a very recent theoretical treatment including Coulomb effects see Refs. [21,22].

The theory of nonadiabatic tunnel ionization of atoms in strong laser fields, denoted as PPT theory, was established in the late 1960s [23, 24], see also Ref. [26]. This theory for short-range potentials, however, reported only the formulas for ionization of arbitrary atomic orbitals in linearly polarized laser fields as well as $s$ orbitals in circularly or elliptically polarized laser fields. Recently, this theory has been extended to nonadiabatic ionization of degenerate $p_0$ and $p_\pm$ orbitals in circularly polarized laser fields [13, 14], see also Refs. [27,28], with interesting features such as the wide-ranging controllability of spin polarization of the photoelectron [29] and of the rotating hole dynamics in the remaining ion [30]. Coulomb effects in nonadiabatic ionization by circularly polarized laser fields have been investigated theoretically in full detail, see Refs. [21,22]. It has been predicted that counter-rotating electrons that rotate against the sense of the circularly polarized field undergoes tunnel ionization easier than co-rotating electrons. In particular, for right circular polarization, the ionization rate of the $p_-$ orbital is always larger than the one of the $p_+$ orbital [13,14]. In other words, the ionization ratio between $p_-$ and $p_+$ orbitals is always above 1. In the low-frequency limit both ionization rates become equal. This prediction has recently been confirmed in an experiment on sequential double ionization of argon induced by two time-delayed strong circularly polarized laser pulses with either the same or opposite helicities [15,16].

In this work, we verify the prediction of PPT theory by numerical solution of the time-dependent Schrödinger equation. Since the circularly polarized laser field breaks the cylindrical symmetry and the numerical calculation in three dimensions is time-consuming, we perform two-dimensional numerical simulations for the ionization of the doubly-degenerate valence $p_\pm$ orbitals of a model neon atom, see section 2. We compare numerical results for ionization ratios with theoretical PPT results and we investigate the ionization dynamics in detail, such as population dynamics, snapshots of the electron probability densities and emission angles, see section 3. By further analysis, we find that for strong laser pulses and low laser frequencies, there are discrepancies between numerical and theoretical results, arising from adiabatic laser-dressed $p$ orbitals. In a 3-level model based on Floquet theory [31,32] we find in section 4 that the ionization ratios between initial $p_-$ and $p_+$ orbitals can be smaller than 1, in agreement with our numerical simulations. Section 5 concludes this work. Atomic units are used throughout
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2. Numerical solution of the 2D time-dependent Schrödinger equation

In this work, we perform simulations of the two-dimensional time-dependent Schrödinger equation (2D-TDSE) for the ionization dynamics of a valence \( p \) electron of a model neon atom in a strong circularly polarized laser pulse. Using atomic units, the 2D-TDSE for the one-electron wavefunction \( \psi(r, t) = \psi(x, y, t) \) in the semiclassical dipole approximation is given by

\[
\frac{i}{\hbar} \frac{\partial}{\partial t} \psi(r, t) = \hat{H}(t) \psi(r, t) = \left[ -\frac{\nabla^2}{2} + V_{\text{eff}}(r) + r \cdot \mathbf{E}_\pm(t) \right] \psi(r, t). \tag{1}
\]

Observables obtained from this theory are gauge-invariant. The effective atomic 2D potential \( V_{\text{eff}}(r) \) is chosen as

\[
V_{\text{eff}}(r) = -\frac{Z_{\text{eff}}(r)}{\sqrt{r^2 + \alpha}}, \tag{2}
\]

where we use a position-dependent core charge \( Z_{\text{eff}}(r) = 1 + 9 \exp(-r^2) \) to account for the screening of the nuclear charge by the inner electrons. It is similar to the empirical 3D potential in Ref. [33], but due to lower dimensionality the exponential function \( \exp(-r^2) \) and the soft-core parameter \( \alpha = 2.88172 \) a.u. are used to obtain the correct value for the energy of the valence \( 2p \) orbital which matches the negative first ionization potential of neon, \( E_{2p} = -I_p = -0.793 \) a.u. [34]. The numerically calculated energies of the \( 1s \) and \( 2s \) orbitals are \( E_{1s} = -2.952 \) a.u. and \( E_{2s} = -0.217 \) a.u., respectively. Our model does not reproduce the correct energy order of the \( s \) and \( p \) orbitals. In the real 3D neon atom, we have \( E_{2s} < E_{2p} \). We will discuss the influence of the orbital energy order on the laser-dressed orbitals in section 4.

The ionization dynamics is induced by a strong right (+) or left (−) circularly polarized laser pulse. The corresponding time-dependent electric field \( \mathbf{E}_\pm(t) \) is defined via vector potential \( \mathbf{A}_\pm(t) \) as

\[
\mathbf{E}_\pm(t) = -\frac{d}{dt} \mathbf{A}_\pm(t) \tag{3}
\]

with \( \lim_{t \to \pm \infty} \mathbf{A}_\pm(t) = \mathbf{0} \), in order to satisfy the condition of the far-field approximation of Maxwell’s equation [35,36]

\[
\int_{-\infty}^{\infty} \mathbf{E}_\pm(t) \, dt = \mathbf{0}. \tag{4}
\]

In this work, we define the circularly polarized vector potential \( \mathbf{A}_\pm(t) \) as

\[
\mathbf{A}_\pm(t) = -\frac{\mathcal{E}}{\omega} s(t) \left[ \sin(\omega t) \mathbf{e}_x \mp \cos(\omega t) \mathbf{e}_y \right] \tag{5}
\]

with the electric field amplitude \( \mathcal{E} \), laser frequency \( \omega \) and trigonometric pulse envelope [37]

\[
s(t) = \begin{cases} 
\sin^2 \left( \frac{\omega t}{2n} \right) & \text{for } t \in \left[ 0, \frac{2\pi n}{\omega} \right] \\
0 & \text{for } t \notin \left[ 0, \frac{2\pi n}{\omega} \right]
\end{cases}, \tag{6}
\]

unless otherwise stated.
where $n$ is the number of laser cycles. Our definition of left and right circular polarization assumes the point of view of the source for a laser field propagating along the positive $z$-direction [38, 39]. We note that the integral of $A_{\pm}(t)$ over the pulse duration is zero for integers $n \geq 2$. In our 2D-TDSE simulations, we use a 3-cycle ($n = 3$) right ($+$) circularly polarized laser pulse with four different electric field amplitudes $E = 0.06 \text{ a.u.}, 0.09 \text{ a.u.}, 0.12 \text{ a.u.}, 0.15 \text{ a.u.}$, corresponding to maximum laser intensities $I_{\text{max}} = 2.53 \times 10^{14} \text{ Wcm}^{-2}$, $5.69 \times 10^{14} \text{ Wcm}^{-2}$, $10.11 \times 10^{14} \text{ Wcm}^{-2}$, $15.79 \times 10^{14} \text{ Wcm}^{-2}$ and five different laser frequencies $\omega = 0.028477 \text{ a.u.}$, $0.037969 \text{ a.u.}$, $0.056954 \text{ a.u.}$, $0.075939 \text{ a.u.}$, $0.113908 \text{ a.u.}$, corresponding to laser wavelengths $\lambda = 2\pi c/\omega = 1600 \text{ nm}$, $1200 \text{ nm}$, $800 \text{ nm}$, $600 \text{ nm}$, $400 \text{ nm}$.

The time-dependent wavefunction $\psi(r, t)$ is propagated using the second-order split-operator method [40, 41] on a Cartesian grid. This spatial grid has $2048 \times 2048 = 4194304$ grid points with the same step sizes for $x$- and $y$-directions, $\Delta x = \Delta y = 0.1 \text{ a.u.}$ The grid ranges from $-102.35 \text{ a.u.}$ to $102.35 \text{ a.u.}$ in each direction. The real time step is chosen as $\Delta t = (\Delta x)^2/2 = 0.005 \text{ a.u.} \approx 0.121 \text{ as.}$ The initial and final times of the real-time propagation are $t_i = 0 \text{ a.u.}$ and $t_f = 1000 \text{ a.u.} \approx 24.2 \text{ fs}$, respectively. The final time $t_f$ is longer than the length of a 3-cycle pulse even for $\lambda = 1600 \text{ nm}$. The stationary normalized wavefunctions $\phi_{(n,m)}(r)$ with principal and azimuthal quantum numbers $(n, m)$ for 1s $(0, 0)$, 2s $(1, 0)$ and $2p_{\pm} (1, \pm 1)$ orbitals are obtained by imaginary-time propagation and orthogonalization under symmetry conditions corresponding to $m$. They are used as initial wavefunctions of the real-time propagation. In this work, we calculate the 2D ionization dynamics numerically for $4 \times 5 = 20$ different laser pulses starting either from the $2p_+ \text{ orbital } \phi_{2p_+}(r)$ or from the $2p_- \text{ orbital } \phi_{2p_-}(r)$.

To avoid unphysical reflections at the grid boundaries and to calculate ionization yields, we use an imaginary absorbing potential defined as

$$V_{\text{abs}}(r) = \begin{cases} -ia(r - r_{\text{abs}})^2 & \text{for } r > r_{\text{abs}} \\ 0 & \text{for } r \leq r_{\text{abs}} \end{cases}, \quad (7)$$

with $a = 0.1$ and $r_{\text{abs}} = 80 \text{ a.u.}$ Without absorbing potential, the norm of the time-dependent wavefunction is conserved, i.e. $\langle \psi(r, t)|\psi(r, t) \rangle = 1$. With absorbing potential, the norm of the wavefunction for $t > 0$ is always less than 1 and the time-dependent ionization yield can be calculated numerically as

$$Y_{(n,m)}(t) = 1 - \langle \psi_{(n,m)}(r, t)|\psi_{(n,m)}(r, t) \rangle, \quad (8)$$

where the index $(n, m)$ indicates the initialization of the wavefunction as $\psi_{(n,m)}(r, 0) = \phi_{(n,m)}(r)$. Furthermore, we also calculate the time-dependent depletion of the initial wavefunction as

$$D_{(n,m)}(t) = 1 - |\langle \psi_{(n,m)}(r, t)|\phi_{(n,m)}(r) \rangle|^2. \quad (9)$$

To compare the ionization yields and depletions at the final time $t_f$ for two initial wavefunctions $\phi_{2p_{\pm}}(r)$, we calculate the ratios

$$R_Y = \frac{Y_{2p_-}(t_f)}{Y_{2p_+}(t_f)}, \quad (10)$$
and

\[ R_D = \frac{D_{2p_-}(t_f)}{D_{2p_+}(t_f)}. \]  

(11)

Then, we compare these ratios with the ratios of the ionization rates \( w_{2p_\pm} \) using the simple formulas derived in Refs. [13, 14], that is the extension of the PPT theory of nonadiabatic ionization [23, 24], i.e.

\[ R_w = \frac{w_{2p_-}}{w_{2p_+}}. \]  

(12)

It can be shown, that the PPT ratios \( R_w \) using the simple formulas from Refs. [13, 14] are equal for 2D and 3D. We note that all ratios depend on the laser parameters \( \mathcal{E} \) and \( \omega \) of the right circularly polarized laser pulse. The PPT ratios for cw laser fields are gauge-invariant [14]. We evaluate these PPT ratios using the peak field strength of our laser pulses. The PPT ratio \( R_w \) depends only on the Keldysh parameter \( \gamma \) defined as

\[ \gamma = \frac{\omega}{\mathcal{E}} \sqrt{2I_p}, \]  

(13)

that discriminates between adiabatic tunneling (\( \gamma \ll 1 \)), nonadiabatic tunneling (\( \gamma \sim 1 \)) and multiphoton ionization (\( \gamma \gg 1 \)). The analytical expression of the PPT ratio \( R_w \) is [13, 14]

\[ R_w = \left( \frac{\sqrt{\zeta_0^2 + \gamma^2} + \zeta_0 \sqrt{1 + \gamma^2}}{\sqrt{\zeta_0^2 + \gamma^2} - \zeta_0 \sqrt{1 + \gamma^2}} \right)^2, \]  

(14)

where \( \zeta_0(\gamma) > 0 \) is the solution of the transcendental equation

\[ \text{artanh} \left( \frac{\zeta_0^2 + \gamma^2}{1 + \gamma^2} \right) = \frac{1}{1 - \zeta_0 \sqrt{\frac{\zeta_0^2 + \gamma^2}{1 + \gamma^2}}}. \]  

(15)

3. Results and discussion

The ratios \( R_Y \) (10) and \( R_D \) (11) obtained by 2D-TDSE numerical calculations for \( n = 3 \) and the PPT ratios \( R_w \) (12) are listed in table 1 and also shown in figure 1. This table and figure show that for not too strong fields the numerically calculated ratios \( R_Y \approx R_D \) agree well with the PPT ratios \( R_w \). The PPT ratios for short-range potentials are always larger than 1, i.e. in a strong right circularly polarized laser field the tunnel ionization of the valence 2\( p_- \) orbital of neon is preferred compared to the tunnel ionization of the valence 2\( p_+ \) orbital. However, there are some deviations between numerical and theoretical results. Below, we will discuss the origins of these deviations. We have also performed numerical calculations for a 6-cycle sin\(^2\) pulse with \( \mathcal{E} = 0.15 \) a.u. and \( \lambda = 1600 \) nm and for a Gaussian pulse with the same full width of half maximum of the intensity envelope. The results \( R_Y \approx R_D \approx 0.782 \) are almost the same for both pulse shapes. The results are similar to those for a 3-cycle laser pulse \( R_Y \approx R_D \approx 0.748 \), see table 1. The weak dependence on the pulse duration is also shown in the recent work [25].
Table 1. Numerically calculated ratios of the ionization yields $R_Y$, equation (10) and depletions $R_D$, equation (11), compared with PPT ratios of the ionization rates $R_w$, equation (12), for the $2p_{\pm}$ orbitals of the model neon atom. Results are shown for various laser parameters $\mathcal{E}$ and $\lambda = 2\pi c/\omega$ of the right circularly polarized laser pulse. The Keldysh parameter $\gamma$, equation (13), is also shown for each choice of parameters.

<table>
<thead>
<tr>
<th>$\mathcal{E}$ (a.u.)</th>
<th>$\lambda$ (nm)</th>
<th>$\gamma$</th>
<th>$R_Y$</th>
<th>$R_D$</th>
<th>$R_w$</th>
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<td>5.799</td>
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<td>1.383</td>
<td>1.682</td>
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<tr>
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For large Keldysh parameters $\gamma \gtrsim 1$, the Coulomb potential plays an important role. Due to the low mean electronic kinetic energy for $\gamma \gtrsim 1$ in a circularly polarized laser field, namely $\mathcal{E}^2/(2\omega^2) = I_p/\gamma^2$ [14], the slow photoelectron can interact with the attractive long-range potential for a longer time. Including Coulomb effects in the theoretical treatment [21] shows that for $\gamma \gtrsim 1$ the ratios of the ionization rates with Coulomb potential are smaller than the ones without long-range potential. For strong fields, e.g. $\mathcal{E} \geq 0.09$ a.u., they could be comparable with numerically calculated ratios, see figure 5 of Ref. [21]. However, for less strong fields, e.g. $\mathcal{E} = 0.06$ a.u., the numerically calculated ratios are significantly larger than the PPT ratios. This is astonishing, because it has not been predicted in theory yet.

For very small Keldysh parameters $\gamma \ll 1$, there are also strong deviations between the numerically calculated ratios $R_Y \approx R_D$ and the PPT ratios $R_w$. The numerically calculated ratios can be smaller than 1. That is in contrast to the prediction by the PPT theory [13, 14]. Numerically for $\gamma \ll 1$ the tunnel ionization of the valence $2p_{\pm}$ (rather than $2p_{-}$) orbital in a right circularly polarized laser field is preferred. This inversion is due to the laser-dressed $2p_{\pm}$ orbitals, which are not included in the PPT
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Figure 1. Numerically calculated ratios $R_Y$ of the ionization yields, equation (10), (colour solid) compared with PPT ratios $R_w$ of the ionization rates, equation (12), (black dashed without symbols) for the $2p_{\pm}$ orbitals of the model neon atom as a function of the Keldysh parameter $\gamma$, equation (13), for field amplitudes $E = 0.06 \text{a.u.}$ (violet triangles), $0.09 \text{a.u.}$ (blue squares), $0.12 \text{a.u.}$ (green diamonds) and $0.15 \text{a.u.}$ (red circles) of the right circularly polarized laser pulse. The symbols from left to right correspond to laser wavelengths $\lambda = 1600 \text{nm}, 1200 \text{nm}, 800 \text{nm}, 600 \text{nm}$ and $400 \text{nm}$. The ratios $R_Y$ and $R_D$, equation (11), are almost indistinguishable on the scale of the graph. Therefore, only $R_Y$ is sohwn.

theory and become important for high field amplitudes $E$ and low laser frequencies $\omega$. We will discuss this effect in detail in section 4.

To obtain more insight into the ionization dynamics of the degenerate $2p_{\pm}$ orbitals in a 3-cycle right circularly polarized laser pulse, we choose the example $E = 0.09 \text{a.u.}$ and $\lambda = 800 \text{nm}$. The corresponding numerical results for the time-dependent orbital populations, norm squares, depletions and ionization yields are shown in figure 2. The $2p_+$ or $2p_-$ orbital at the initial time $t = t_0 = 0 \text{a.u.}$ is depleted during the tunnel ionization process, while the laser-induced population of the initially empty $2p_-$ or $2p_+$ orbital remains negligible. The populations of $1s$ (not shown in figure 2) and $2s$ orbitals are also very small. The fact that the $2p_+/2p_-$ population and the norm square of the wavefunction at the final time are almost identical reveals that the populations of other excited orbitals at the final time are also negligible. It means that the depletion of the initial wavefunction and the ionization yield at the final time are similar, i.e. $D_{2p_+}(t_f) \approx Y_{2p_+}(t_f)$ and $D_{2p_-}(t_f) \approx Y_{2p_-}(t_f)$, leading to similar ratios $R_Y \approx R_D$, see also table 1 and figure 1. Importantly, figure 2 shows that the dynamics starting from the valence $2p_+$ and $2p_-$ orbitals are different. In particular, $D_{2p_-}(t_f) > D_{2p_+}(t_f)$ and $Y_{2p_-}(t_f) > Y_{2p_+}(t_f)$ mean that the electron from the $2p_-$ orbital is released easier than the electron from the $2p_+$ orbital. This clearly confirms the prediction of the PPT theory in Ref. [13,14] and shows that $R_Y \approx R_D \approx R_w > 1$, see table 1.

In the presence of the right circularly polarized laser pulse, electrons from the valence $2p_+$ or $2p_-$ orbital of neon can tunnel through the barrier, if the electric field is strong enough. Tunnel ionization is most probable at time $t_{\text{max}} = 3\pi/\omega$ where the
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Figure 2. Ionization dynamics of neon starting from the valence 2p$_+$ (a,b) and 2p$_-$ (c,d) orbital for the field amplitude $\mathcal{E} = 0.09$ a.u. and laser wavelength $\lambda = 800$ nm, corresponding to the Keldysh parameter $\gamma = 0.797$. Shown are the populations $\langle \psi(r,t) | \phi_{2p_{\pm}}(r) \rangle^2$ (a,d) (red solid), $\langle \psi(r,t) | \phi_{2s}(r) \rangle^2$ (b,c) (green solid) and $\langle \psi(r,t) | \phi_{2s}(r) \rangle^2$ (b,d) (blue dotted) of the 2p$_+$, 2p$_-$ and 2s orbitals, the norm square of the wavefunction $\langle \psi(r,t) | \psi(r,t) \rangle$ (a,c) (orange dash-dotted), the ionization yields $Y_{2p_{\pm}}(t)$ (b) and $Y_{2p_{-}}(t)$ (d) (black dash-dotted) and the depletions of the initial wavefunction $D_{2p_{\pm}}(t)$ (b) and $D_{2p_{-}}(t)$ (d) (black dashed).

The electric field has its maximum value and the corresponding field vector points along the negative $x$-direction. Snapshots of the numerically calculated electron probability density $|\psi(r,t_{max})|^2$ for $\mathcal{E} = 0.09$ a.u. are shown in figure 3. This figure clearly shows that the ionization probabilities for 2p$_\pm$ orbitals are different and the ionization from the 2p$_-$ orbital is preferred. The numerically calculated ratios $R_Y$ and $R_D$ agree well with the PPT ratios $R_w$ for different laser wavelengths, see table 1 for $\mathcal{E} = 0.09$ a.u. They are larger than 1, i.e. the bound state with the counter-rotating electron is ionized easier.

Figure 3 also shows the radius-dependent positions ($x_{max}(r)$, $y_{max}(r)$) corresponding to the angle where the electron probability density at $t = t_{max}$ is maximal. These different curves for initial 2p$_\pm$ orbitals mean that the emission angles $\varphi_{2p_{\pm}} = \arctan(y_{max}(r_{exit})/x_{max}(r_{exit}))$ at the minimum distance to the tunnel exit $r_{exit} = 7.38$ a.u. are different. They also depend on the field amplitude (cf. figure 5) and frequency. For fast electric field rotations (small wavelength), the emission angles deviate strongly from zero. This is in contrast to the semiclassical picture where
Figure 3. Snapshots of the electron probability density $|\psi(r,t_{\text{max}})|^2$ in a.u. at time $t = t_{\text{max}} = 3\pi/\omega$ where the right circularly polarized electric field has the maximum and the corresponding field vector points along the negative $x$-direction (orange arrows), for the field amplitude $E = 0.09$ a.u., different laser wavelengths $\lambda = 1600$ nm (a,d), 800 nm (b,e), 400 nm (c,f) and different initial orbitals $2p_+$ (a–c) and $2p_-$ (d–f). The density for radii larger than the minimum distance to the tunnel exit $r_{\text{exit}} = 7.38$ a.u. is scaled by the factor 3000 to better display both the bound-state inner region and the ionization part in the outer region. The red ($2p_+$) and green ($2p_-$) curves correspond to the radius-dependent angle where the electron probability density is maximal. The coordinates $x$ and $y$ are in atomic units.

Figure 4. (a) Electron probability density $|\psi(r,t_{\text{max}})|^2$ at the minimum distance to the tunnel exit, $r = r_{\text{exit}} = 7.38$ a.u., versus angle $\phi = \arctan(y/x)$ for right circular polarization, field amplitude $E = 0.09$ a.u., laser wavelength $\lambda = 800$ nm and different initial orbitals $2p_+$ (red) and $2p_-$ (green) with corresponding maxima at emission angles $\phi_{2p_-} < \phi_{2p_+} < 0$. (b) Emission angles $\phi_{2p_\pm}(r)$ versus radius $r$ for $2p_+$ (red) and $2p_-$ (green) orbitals and the difference $\Delta \phi(r) = \phi_{2p_+}(r) - \phi_{2p_-}(r)$ (black dashed). The radius $r = r_{\text{exit}}$ is shown as vertical line.
the electron tunnels exactly against the electric field vector, i.e. in the positive $x$-direction, corresponding to zero emission angle. Numerical calculations in figure 3 show that the emission angles are always smaller than 0 for right circular polarization ($\varphi_{\text{right}} < 0$). Although negative emission angles for right circular polarization due to the Coulomb potential ($\varphi_{\text{Coulomb}} < 0$) have recently been predicted theoretically in Ref. [21], this theory does not predict the different emission angles $\varphi_{2p_\pm}$ for $2p_\pm$ orbitals. Detailed analysis of the electron probability density $|\psi(r, t_{\text{max}})|^2$ at $r = r_{\text{exit}}$ versus angle $\varphi = \arctan(y/x)$ (see figure 4) shows that the emission angle for $2p_+$ orbital is closer to zero than for $2p_-$ orbital ($\varphi_{2p_-} < \varphi_{2p_+} < 0$). This is plausible because the azimuthal momentum component of the $2p_+/2p_-$ orbital is positive/negative (cf. Ref. [42]), adding to or subtracting from the mean emission angle $\bar{\varphi} = (\varphi_{2p_+} + \varphi_{2p_-})/2 < 0$, respectively. We expect that this non-zero mean emission angle $\bar{\varphi} < 0$ for right circular polarization is comparable with $\varphi_{\text{Coulomb}} < 0$ [21]. The non-zero emission angles for degenerate $E_\pm$ states in a linearly polarized laser field have already been observed in numerical calculations [42]. The $E_\pm$ states are eigenstates of the angular momentum component $\hat{L}_z$, similar to $P_\pm$ states. Due to the non-zero and opposite angular momenta of $E_\pm$ states, one finds $\varphi_{E_+} > 0$ and $\varphi_{E_-} = -\varphi_{E_+} < 0$ for linear polarization [42]. Figure 4 also shows the emission angles $\varphi_{2p_\pm}(r) = \arctan(y_{\text{max}}^{2p_\pm}(r)/x_{\text{max}}^{2p_\pm}(r))$ at $t = t_{\text{max}}$ as a function of $r$. It clearly shows that the difference of emission angles $\Delta \varphi(r) = \varphi_{2p_+}(r) - \varphi_{2p_-}(r)$ for $r \geq r_{\text{exit}}$ depends only weakly on $r$ and has a robust value of about $6^\circ - 10^\circ$. Furthermore, we expect that this difference does not change significantly with time, even after the end of the laser pulse. Therefore, we believe that different emission angles for degenerate valence $p_\pm$ orbitals of the atomic prepared state $P_\pm$ (e.g. ground state of a halogen atom) can be observed in attoclock experiments, see Refs. [17–20]. Recently, it has been theoretically predicted for one-photon ionization in a circularly polarized laser field that the time delay is sensitive to the emission angle of the photoelectron and that there is a pronounced difference between co- and counter-rotating electrons from $2p_\pm$ orbitals of the excited Li atom [43].

4. Laser-dressed orbitals

As already mentioned in the previous section, the numerically calculated ratios of the ionization yields $R_Y$ and depletions $R_D$ for high field amplitudes $E$ and low laser frequencies $\omega$, corresponding to low Keldysh parameters $\gamma = \sqrt{2T_p\omega/E}$, can be smaller than 1, see table 1 and figure 5. This cannot be reproduced by PPT theory for circular polarization [13, 14], because laser-dressed $2p_\pm$ orbitals are not considered in this theory. For the strong field $E = 0.15\,\text{a.u.}$, the strongly laser-dressed $2p_\pm$ orbitals are clearly visible in the inner region of the snapshots of the electron probability density $|\psi(r, t_{\text{max}})|^2$, see figure 5, particularly for long wavelength. For right circular polarization and orbital energy order $E_{2p} < E_{2s}$ as well as $\omega < E_{2s} - E_{2p}$ used in the 2D numerical calculations (see below), the initial $2p_\pm$ orbital is modified to the rotating $2p_z$-like orbital denoted as $2p\parallel$ orbital since this orbital is always aligned with the rotating electric
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Figure 5. Same as for figure 3, but for stronger field amplitude $E = 0.15$ a.u. to bring out the laser-dressed $2p_{\pm}$ orbitals. The density for radii larger than the minimum distance to the tunnel exit, $r_{exit} = 3.81$ a.u., is scaled by the factor 30.

field vector. In contrast, the initial $2p_\pm$ orbital is modified to the rotating $2p_y$-like orbital denoted as $2p_\perp$ orbital that is always aligned perpendicular to the electric field vector. The ionization of the $2p_\parallel$ orbital is preferred since in the semiclassical picture the electron tunnels preferably against the electric field vector and the ionization of the $2p_\perp$ orbital is suppressed. The ratios of the ionization yields $Y_{2p_\perp}(t_f)/Y_{2p_\parallel}(t_f)$ and depletions $D_{2p_\perp}(t_f)/D_{2p_\parallel}(t_f)$ are therefore smaller than 1. This implies that $R_Y = Y_{2p_\perp}(t_f)/Y_{2p_\parallel}(t_f) \approx R_D = D_{2p_\perp}(t_f)/D_{2p_\parallel}(t_f) < 1$. In the low-frequency limit $\omega \to 0$ (corresponding to $\gamma \to 0$), PPT theory for laser fields without envelope predicts that the ratio $R_w$ is exactly equal 1, see Refs. [13, 14]. In the simulation, in contrast, there is a slow adiabatic change of the initial $2p_\pm$ orbitals to laser-dressed $2p_\parallel$ and $2p_\perp$ orbitals during the laser pulse. If we wish to account for the laser-dressing in the PPT theory, we have to use non-degenerate $2p_\parallel$ and $2p_\perp$ orbitals for initialization instead of degenerate $2p_\pm$ orbitals.

We will investigate how the laser-dressed orbitals depend on the electric field amplitude, laser frequency, sense of circular polarization, orbital energies and orbital energy order. The shifts of the energy levels of atoms and atomic ions by dynamic Stark effects can be calculated theoretically based on the Floquet theory [31,32], see Ref. [44] for alternating laser fields and Refs. [45–47] for circularly polarized and arbitrarily strong laser fields using matrix diagonalization. However, these references do not discuss the shape of laser-dressed orbitals. Here, we apply the Floquet theory to one electron in a three-level model in a circularly polarized laser field without envelope to describe the laser-dressed orbitals $2p_\parallel$ and $2p_\perp$ qualitatively. We are aware that Floquet theories can be problematic for short pulses [48]. However, in this work, we would like to keep the theory as simple as possible. Since the TDSE results for a 3-cycle laser pulse and for a
6-cycle laser pulse are similar (see above), we expect that the adiabatic Floquet theory is applicable. We calculate not only energy shifts but also the Floquet states. The field-free energy eigenstates of our three-level model are the $2s$ and $2p_\pm$ orbitals. We assume that the choice of only these three orbitals is sufficient for qualitative description of orbital alignment and that the contributions of other field-free orbitals to the field-induced $2p_\|$ and $2p_\perp$ orbitals are not important. Multilevel Floquet theory with numerical matrix diagonalization could be used, but it is outside the scope of this work as we do not aim at quantitative agreement with the TDSE results.

According to the Floquet theory, we consider the periodic Hamiltonian

$$\hat{H}_F(t) = \hat{H}_F(t + nT) = \hat{H}_0 + \mathbf{r} \cdot \mathbf{E}_\pm(t) \quad (n \in \mathbb{Z})$$

(16)

where $\hat{H}_0 = -\nabla^2/2 + V_{\text{eff}}(r)$ is the time-independent Hamiltonian and $T = 2\pi/\omega$ is the laser period. To solve the corresponding TDSE

$$i \frac{\partial}{\partial t} |\psi_F(t)\rangle = \hat{H}_F(t)|\psi_F(t)\rangle$$

(17)

for the one-electron wavefunction $\psi_F(\mathbf{r}, t)$, we use the ansatz

$$|\psi_F(t)\rangle = |\phi_F(t)\rangle e^{-iE_F t}$$

(18)

where $E_F$ is the quasienergy and $\phi_F(\mathbf{r}, t) = \phi_F(\mathbf{r}, t + nT)$ is the periodic Floquet wavefunction. Inserting equation (18) into equation (17) yields

$$\left( \hat{H}_0 - E_F - i \frac{\partial}{\partial t} + \mathbf{r} \cdot \mathbf{E}_\pm(t) \right) |\phi_F(t)\rangle = 0.$$

(19)

As an ansatz for the Floquet wavefunction $\phi_F(\mathbf{r}, t)$ we use a superposition of the field-free $2s$ and $2p_\pm$ orbitals, i.e.

$$|\phi_F(t)\rangle = a|\phi_{2s}\rangle + b_+ e^{\mp i\omega t}|\phi_{2p_+}\rangle + b_- e^{\pm i\omega t}|\phi_{2p_-}\rangle.$$  

(20)

It satisfies the periodicity condition $\phi_F(\mathbf{r}, t) = \phi_F(\mathbf{r}, t + nT)$. The different signs $\mp$ and $\pm$ in equation (20) correspond to the sense of the circular polarization of the laser field $\mathbf{E}_\pm(t)$. The choice of the exponential factors in equation (20) is justified because for right circular polarization one photon is necessary for the transition $s \rightarrow p_+$. Thus there is an additional factor $e^{-i\omega t}$ in the $2p_+$ term compared to the $2s$ term. Likewise, one photon is necessary for the transition $p_- \rightarrow s$, thus there is an additional factor $e^{+i\omega t}$ in the $2p_-$ term. Applying $\langle \phi_{2s} |$, $\langle \phi_{2p_+} |$ and $\langle \phi_{2p_-} |$ to equation (19) and using $\hat{H}_0|2s\rangle = E_{2s}|2s\rangle$ and $\hat{H}_0|2p_\pm\rangle = E_{2p}|2p_\pm\rangle$ as well as the only one non-vanishing dipole matrix element $\mathbf{D} = \langle \phi_{2s} | \mathbf{r} | \phi_{2p_+} \rangle = \langle \phi_{2s} | \mathbf{r} | \phi_{2p_-} \rangle^*$, we find the three equations

$$
\begin{align*}
(E_{2s} - E_F) a + \mathbf{D} \cdot \mathbf{E}_\pm(t) b_+ e^{\mp i\omega t} + \mathbf{D}^* \cdot \mathbf{E}_\pm(t) b_- e^{\pm i\omega t} &= 0, \\
(E_{2p} - E_F + \omega) b_+ + \mathbf{D}^* \cdot \mathbf{E}_\pm(t) a e^{\pm i\omega t} &= 0, \\
(E_{2p} - E_F - \omega) b_- + \mathbf{D} \cdot \mathbf{E}_\pm(t) a e^{\mp i\omega t} &= 0.
\end{align*}

(21-23)

In our definition of the dipole matrix element, we have omitted the negative charge of the electron. Using the right $(+)$ or left $(-)$ circularly polarized electric field

$$\mathbf{E}_\pm(t) = E [\cos(\omega t) \mathbf{e}_x \pm \sin(\omega t) \mathbf{e}_y]$$

(24)
and the dipole matrix element

$$D = \frac{1}{\sqrt{2}} \left( \langle \phi_{2s} | r | \phi_{2p_x} \rangle + i \langle \phi_{2s} | r | \phi_{2p_y} \rangle \right) = D (e_x + i e_y), \quad (25)$$

where \( D = \langle \phi_{2s} | x | \phi_{2p_x} \rangle / \sqrt{2} = \langle \phi_{2s} | y | \phi_{2p_y} \rangle / \sqrt{2} \), we obtain

$$D \cdot E(t) = D \mathcal{E} [\cos(\omega t) \pm i \sin(\omega t)] = D \mathcal{E} e^{\pm i \omega t}. \quad (26)$$

Inserting this equation into equations (21)–(23) yields the system of time-independent equations in matrix notation

$$\begin{pmatrix} E_{2s} - E_F \\ D \mathcal{E} \\ D \mathcal{E} \\ D \mathcal{E} \end{pmatrix} = \begin{pmatrix} a \\ b_+ \\ 0 \\ b_- \end{pmatrix} = 0. \quad (27)$$

To obtain non-trivial solutions for the three quasienergies \( E_F \) and amplitudes \( a, b_\pm \), the determinant of the matrix in equation (27) is set to zero, leading to a cubic equation in \( E'_F \), i.e.

$$(E'_F - \Delta E)(E'_F - \omega^2) - 2(D \mathcal{E})^2 E'_F = 0, \quad (28)$$

where \( E'_F = E_F - E_{2p} \) is the relative quasienergy to the energy of the field-free \( 2p \) orbital and \( \Delta E = E_{2s} - E_{2p} \) is the energy difference between field-free \( 2s \) and \( 2p \) orbitals. We note that in the zero-field case (\( \mathcal{E} = 0 \)), the relative quasienergies are \( E'_F = \Delta E \) and \( E'_F = \pm \omega \), corresponding to \( E_F = E_{2s} \) and \( E_F = E_{2p} \). In our case, the non-zero laser frequency \( \omega \) is not equal to the energy difference \( \Delta E \), i.e. \( 0 \neq \omega \neq |\Delta E| \). Then, the cubic equation (28) has three real distinct solutions for the relative quasienergy \( E'_F \). We use Cardano’s formula [49] to obtain three \( (m = 0, \pm 1) \) relative quasienergies \( E'_{F,m} \) as

$$E'_{F,m} = (-1)^m 2 \sqrt{Q} \cos \left( \frac{1}{3} (\theta - m \pi) \right) + \frac{\Delta E}{3}, \quad (29)$$

where

$$\theta = \arccos \left( \frac{R}{\sqrt{Q^3}} \right), \quad (30)$$

$$R = \frac{\Delta E}{3} \left( (D \mathcal{E})^2 - \omega^2 + \frac{(\Delta E)^2}{9} \right), \quad (31)$$

$$Q = \frac{1}{3} \left( 2(D \mathcal{E})^2 + \omega^2 + \frac{(\Delta E)^2}{3} \right). \quad (32)$$

The corresponding Taylor series up to second order in \( D \mathcal{E} \) for \( \Delta E > \omega > 0 \) are

$$E'_{F,0} \approx \Delta E + \frac{2\Delta E (D \mathcal{E})^2}{(\Delta E)^2 - \omega^2}, \quad (33)$$

$$E'_{F,-1} \approx \omega - \frac{(D \mathcal{E})^2}{\Delta E - \omega}, \quad (34)$$

$$E'_{F,1} \approx -\omega - \frac{(D \mathcal{E})^2}{\Delta E + \omega}. \quad (35)$$
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Figure 6. Relative quasienergies $E'_{F,m} = E_{F,m} - E_{2p}$, equation (29), of the three laser-dressed orbitals $2\tilde{s}$ ($m = 0$, blue dotted), $2p_\perp$ ($m = -1$, green solid) and $2p_{\parallel}$ ($m = 1$, red solid), connecting to the field-free orbitals $2s$ ($m = 0$), $2p_-$ ($m = -1$) and $2p_+$ ($m = 1$), for the laser wavelength $\lambda = 1600$ nm, corresponding to the laser frequency $\omega = 0.0285$ a.u.. This graph assumes the case $\Delta E > \omega > 0$ with orbital energy difference $\Delta E = E_{2s} - E_{2p} = 0.576$ a.u. and dipole strength $D = 0.463$ a.u. The energies are plotted versus electric field amplitude $\mathcal{E}$ of the right circularly polarized periodic laser field. The field amplitude $\mathcal{E} = 0.15$ a.u. is shown as vertical black line.

In the zero-field limit ($\mathcal{E} \to 0$) and for $\Delta E > \omega > 0$, these relative quasienergies approach $E'_{F,0} = \Delta E$ and $E'_{F,\pm 1} = \pm \omega$. Therefore, by comparison for right circular polarization (see equation (27)) $E'_{F,0}$ corresponds to the field-free $2s$ orbital whereas $E'_{F,\pm 1}$ correspond to the field-free $2p_\pm$ orbitals. Figure 6 shows the relative quasienergies $E'_{F,m}$ ($m = 0, \pm 1$), equation (29), for $\Delta E > \omega > 0$ versus electric field amplitude $\mathcal{E}$, where the numerically calculated dipole strength $D = 0.463$ a.u. in our 2D model of neon atom is used. For other cases $\omega > \Delta E > 0$, $0 > \Delta E > -\omega$ and $0 > -\omega > \Delta E$, the meaning of the relative quasienergies $E'_{F,m}$ at the limit $\mathcal{E} \to 0$ are changed, because $m = 0$ always corresponds to the state with the highest energy, $m = -1$ to the state with the middle energy and $m = 1$ to the state with the lowest energy, see table 2 for the results. For right circular polarization and $\Delta E > \omega > 0$, these field-free $2s$, $2p_-$ and $2p_+$ orbitals modify adiabatically to field-dressed $2\tilde{s}$ ($m = 0$), $2p_\perp$ ($m = -1$) and $2p_{\parallel}$ ($m = 1$) orbitals (see below) with increasing field amplitude $\mathcal{E}$. There are no energy crossings between Floquet states. For the cases $\omega > \Delta E > 0$, $0 > \Delta E > -\omega$ and $0 > -\omega > \Delta E$, the adiabatic modification of the orbitals is different, see below and table 2.

The real amplitudes $a_m$ and $b_{\pm,m}$ of the field-free $2s$ and $2p_{\pm}$ orbitals contributing to the laser-dressed orbitals for $m = 0, \pm 1$ and right circular polarization are easily calculated using equation (27), resulting in

\[ b_{+,m} = \frac{D\mathcal{E}}{E'_{F,m} + \omega} a_m, \]

\[ b_{-,m} = \frac{D\mathcal{E}}{E'_{F,m} - \omega} a_m \]
At time $t_i.e.\text{ with normalization condition}$

$$a_m^2 + b_{+,m}^2 + b_{-,m}^2 = 1,$$  

i.e.

$$a_m = \left[1 + \frac{(D\mathcal{E})^2}{(E'_{F,m} + \omega)^2} + \frac{(D\mathcal{E})^2}{(E'_{F,m} - \omega)^2}\right]^{-1/2}.$$  

At time $t = t_{\text{max}} = 3\pi/\omega$, where the electric field vector points along the negative $x$-direction, see figure 5, the amplitudes $b_{x,m}$ and $b_{y,m}$ of the field-free $2p_x$ and $2p_y$ orbitals are calculated as

$$b_{x,m} = \frac{b_{+,m} + b_{-,m}}{\sqrt{2}},$$  

$$b_{y,m} = \frac{b_{+,m} - b_{-,m}}{\sqrt{2}}.$$  

Using equations (20), (40), (41), $\phi_{2s}(r) = \phi_{2s}(r)$ and $\phi_{2p}(r) = \phi_{2p}(r)e^{\pm i\varphi}$, the time-dependent electron probability density of the field-dressed orbitals for right circular polarization is

$$|\phi_{F,m}(r, t)|^2 = a_m^2|\phi_{2s}(r)|^2 + (b_{+,m}^2 + b_{-,m}^2)|\phi_{2p}(r)|^2$$  

$$+ 2a_m(b_{+,m} + b_{-,m})|\phi_{2p}(r)|^2 \cos(\omega t - \varphi)$$  

$$+ 2b_{+,m}b_{-,m}|\phi_{2p}(r)|^2 \cos(2(\omega t - \varphi)),$$

and for $t = t_{\text{max}} = 3\pi/\omega$

$$|\phi_{F,m}(r, t_{\text{max}})|^2 = a_m^2|\phi_{2s}(r)|^2 + (b_{+,m}^2 + b_{-,m}^2)|\phi_{2p}(r)|^2$$  

$$- 2\sqrt{2}a_m b_{x,m} \phi_{2s}(r)\phi_{2p}(r) \cos \varphi$$  

$$+ (b_{+,m}^2 - b_{-,m}^2)|\phi_{2p}(r)|^2 \cos(2\varphi).$$

The first two terms of equation (42) are time-independent and do not contribute to the electron dynamics of the dressed orbital. For $a_m \neq 0$, i.e. including the $2s$ orbital,
the term proportional to \( \cos(\omega t - \varphi) \) distorts the orbital and this distortion rotates with the electric field vector. If the relative quasienergy \( E'_{F,m} \) decreases with increasing field amplitude \( \mathcal{E} \), it can be shown that the prefactor \( a_m b_{x,m} \) is negative, leading to the distortion of the orbital to the positive \( x \)-axis at the time \( t = t_{\text{max}} \) when the electric field points along to the negative \( x \)-axis. If both \( b_{m,+} \neq 0 \) and \( b_{m,-} \neq 0 \), the term proportional to \( \cos(2(\omega t - \varphi)) \) contributes to the laser-induced modification of the field-free \( 2p_{\pm} \) orbitals and rotates with the electric field vector. In particular at time \( t = t_{\text{max}} \), this term is non-zero for \( b_{x,m} \neq b_{y,m} \), i.e. for the different populations of the \( 2p_x \) and \( 2p_y \) orbitals. This modified orbital is aligned along the rotating electric field vector if \( |b_{x,m}| > |b_{y,m}| \) and perpendicular to the rotating electric field vector if \( |b_{x,m}| < |b_{y,m}| \). These two possibilities correspond to the field-dressed rotating \( 2p_{||} \) and \( 2p_{\perp} \) orbitals, respectively. It can be shown analytically, that for the case \( \Delta E > \omega > 0 \) the conditions \( |b_{x,-1}| < |b_{y,-1}| \) and \( |b_{x,1}| > |b_{y,1}| \) are satisfied, therefore the field-free \( 2p_{-} \) and \( 2p_{+} \) orbitals are modified adiabatically to the \( 2p_{\perp} \) and \( 2p_{||} \) orbitals, respectively. Figure 7 confirms this conclusion for the case \( \Delta E > \omega > 0 \) by showing the weights \( |a_m|^2 \) and \( |b_{\pm,m}|^2 \) as well as \( |b_{x,m}|^2 \) and \( |b_{y,m}|^2 \) for a right circularly polarized laser pulse. The results for the cases \( \omega > \Delta E > 0, 0 > \Delta E > -\omega \) and \( 0 > -\omega > \Delta E \) are summarized in table 2.

Therefore, by deep analysis of the three-level Floquet theory, we confirm that the initial \( 2p_{\pm} \) orbitals for high field amplitudes \( \mathcal{E} \) and low laser frequencies \( \omega \) are strongly modified, see figure 5. In particular, for our 2D model of neon atom, where \( \Delta E = E_{2s} - E_{2p} > \omega \), the initial \( 2p_{-} \) and \( 2p_{+} \) orbitals are modified to laser-dressed
2p\(_{\perp}\) and 2p\(_{\parallel}\) orbitals that are perpendicular and parallel to the rotating electric field vector, respectively. Therefore, the ratios of the ionization yields \(R_Y\) and depletions \(R_D\) will be smaller than 1, because the tunnel ionization of the 2p\(_{\parallel}\) orbital is preferred over 2p\(_{\perp}\). Figure 1 indicates that in linear extrapolation to the low-frequency limit \(\omega \to 0\), smaller electric field amplitudes imply smaller ratios. This is qualitatively consistent with the low-frequency limit for the ratio of two orthogonally oriented orbitals ionized by a linearly polarized field [23]. Very recently, these discoveries with respect to orbital modifications and ratios of ionization rates are also confirmed by numerical TDSE calculations for the 2D model argon atom [25]. However, in a real 3D noble gas atom with \(\Delta E = E_{2s} - E_{2p} < -\omega\), we conjecture that the ionization behaviour is completely reversed, i.e. the initial 2p\(_{-}\) and 2p\(_{+}\) orbitals are modified to laser-dressed 2p\(_{\parallel}\) and 2p\(_{\perp}\) orbitals, respectively, see table 2. It leads to ratios \(R_Y\) and \(R_D\) larger than 1, which is in principle in accord with the PPT theory. However, in the limit \(\omega \to 0\), these ratios are expected to be much larger than 1, in contrast to the PPT theory for circularly polarized laser fields but in accord with the PPT theory for linearly polarized fields using p orbitals aligned parallel and perpendicular to the polarization axis.

5. Conclusion

The numerical results for ionization of the valence orbitals 2p\(_{\pm}\) of a model neon atom in a circularly polarized laser pulse obtained are generally in agreement with the theoretical results for short-range potentials [13, 14] as well as for the Coulomb potential [21]. In particular, the counter-rotating electron tunnels preferably. However for strong laser pulses and low laser frequencies, the discrepancies between numerical and theoretical results show that the laser-dressed p orbitals play an important role. In a 3-level model based on Floquet theory we find that, depending on the orbital energy order of the valence s and p\(_{\pm}\) orbitals, the p\(_{-}\) and p\(_{+}\) orbitals are adiabatically modified to laser-dressed p\(_{\perp}\) and p\(_{\parallel}\) orbitals or vice versa, where p\(_{\perp}\) and p\(_{\parallel}\) are aligned perpendicular and parallel to the instantaneous field. Since the laser-dressed p\(_{\perp}\) and p\(_{\parallel}\) have different ionization behaviours, we suggest that for strong laser fields and low laser frequencies, the laser-dressed p\(_{\perp}\) and p\(_{\parallel}\) should be used in the derivation of the PPT formulas instead of p\(_{\pm}\) orbitals, in order to obtain improved formulas for the ionization rates of the initial p\(_{\pm}\) orbitals.

Furthermore, we discover for the first time that the emission angles for valence p\(_{-}\) and p\(_{+}\) orbitals in circularly polarized laser fields are different. A related phenomenon for linear polarization has already been found in Ref. [42]. We believe that the emission angles at the tunnel exit correlate with the offset angles at the detector in the attoclock experiments [17–20]. Therefore, we expect that the different offset angles for atomic prepared states \(P_{\pm}\) (e.g. an m-prepared ground state of a halogen atom) can be observed in attoclock experiments.
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