Comparison of theory and experiment for nonadiabatic tunneling in circularly polarized fields

Ingo Barth and Olga Smirnova
Max Born Institute, Max-Born-Str. 2A, 12489 Berlin, Germany
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We compare results of the recent experiment by Herath *et al.* [T. Herath, L. Yan, S. K. Lee, and W. Li, Phys. Rev. Lett. **109**, 043004 (2012)] on strong-field nonadiabatic tunneling in circularly polarized laser fields with the original predictions of our theory [I. Barth and O. Smirnova, Phys. Rev. A **84**, 063415 (2011)] that stimulated these experiments. We show that the theory and experiment are in very good agreement. We also explain why the initial comparison performed by Herath *et al.* [T. Herath, L. Yan, S. K. Lee, and W. Li, Phys. Rev. Lett. **109**, 043004 (2012)] has suggested quantitative discrepancies with our theory. We confirm that these seeming discrepancies are removed with an accurate application of our theoretical model. We suggest an experiment for unique determination of the ionization preference of valence orbitals p_+ or p_-.

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Strong-field ionization can be understood as electron tunneling through the barrier created by the laser field and the core potential. Sensitivity of the tunneling rate in circularly polarized fields to the sense of electron rotation in the initial state [1,2] is a purely nonadiabatic effect, associated with the rotation of the barrier. The kinematics of the electron motion in a classically forbidden region is responsible for a higher ionization rate for an electron counter-rotating with respect to the laser field prior to ionization [1,2]. For example, a right circularly polarized laser field preferentially removes an electron from the valence p_- rather than p_+ orbital, creating a counter-rotating hole in the system. This asymmetry has recently been experimentally observed in an elegant experiment by Herath *et al.* [3]. Asymmetry in ionization from p_- and p_+ orbitals opens unique opportunities for production of intense ultrashort bursts of spin-polarized electrons (and ions) in strong-field ionization [4].

The experiment of Herath *et al.* [3] used two time-delayed (500-fs) circularly polarized laser pulses with same (RR) and opposite (LR) polarizations to induce double ionization of an Ar atom. By monitoring the yield of sequential double ionization (SDI) triggered by the RR or LR sequence of pulses, one can infer the sensitivity of ionization to the pulse helicity in each step. Indeed, as suggested in [3], the ratio of the SDI yields recorded in each experiment $\frac{I_{\text{SDI-LR}}}{I_{\text{SDI-RR}}}$ can be related to the ratios of the ionization rates $\frac{w_+^{p-}}{w_+^{p+}}$ from p_m orbitals of Ar (α) and Ar^+ (α') in right circularly polarized fields as follows:

$$\frac{I_{\text{SDI-LR}}}{I_{\text{SDI-RR}}} = \frac{\alpha\alpha' + 1}{\alpha + \alpha'}. \quad (1)$$

Performing a sequence of measurements in their ingenious setup, Herath *et al.* [3] managed to reconstruct α and α'. However, the comparison with the theory of [1] performed in [3] suggested a discrepancy, which we address below.

Time-delayed linearly polarized pump and probe laser pulses used in the experiment are reported [3] to have the mean intensities $\bar{I}_{\text{lin}} = 9 \times 10^{13}$ W/cm^2 and $\bar{I}'_{\text{lin}} = 1.4 \times 10^{14}$ W/cm^2. These values correspond to the amplitudes of the electric fields $\mathcal{E}_{\text{lin}} = 0.051$ a.u. and $\mathcal{E}'_{\text{lin}} = 0.063$ a.u. Herath *et al.* used a quarter wave plate to produce circularly polarized light, where the angle between the polarization of the incident wave and the optical axis of the crystal is adjusted to 45°. After the wave plate, the energy in the pulse stays the same but the amplitudes of the parallel and perpendicular components of the circularly polarized electric-field vector have reduced peak values compared to linear polarization, $\mathcal{E}_x = \mathcal{E}_y = \mathcal{E}_{\text{lin}} \cos(45°) = \mathcal{E}_{\text{lin}}/\sqrt{2}$. The same applies to the amplitude of the circularly polarized electric laser field, i.e., $\mathcal{E}_{\text{circ}} = \mathcal{E}_{\text{lin}}/\sqrt{2}$. The corresponding values for circularly polarized pump and probe laser pulses are $\mathcal{E}_{\text{circ}} = 0.036$ a.u. and $\mathcal{E}'_{\text{circ}} = 0.045$ a.u. The intensities of the circularly polarized pulses are $I_{\text{circ}} = \bar{I}_{\text{lin}}$ and $I'_{\text{circ}} = \bar{I}'_{\text{lin}}$.

Using these values for the amplitudes $\mathcal{E}_{\text{circ}}$ and $\mathcal{E}'_{\text{circ}}$ and the laser frequency $\omega = 0.057$ a.u. (800 nm), we get the Keldysh parameters $\gamma = 1.7$ for Ar and $\gamma' = 1.8$ for Ar^+. Applying analytical formulas Eqs. (6)–(9) from [1], we obtain the ratios of the ionization rates $\alpha = 5.8$ and $\alpha' = 6.2$ and the SDI ratio 3.1 in good agreement with the experimental result 3.6 before the focal averaging is taken into account. The ratios $\alpha = 5.8$ and $\alpha' = 6.2$ agree with the the experimental value $\alpha = \alpha' = 7.1$ within the error bars established in the experiment [3].

Interestingly, in contrast to common intuition, also shared by Herath *et al.* [3], that the intensity and the focal volume averaging generally tend to reduce observed strong-field effects, the asymmetry discussed here is an example of a strong-field effect that *benefits* from focal and intensity averaging. Indeed, we have shown in [1,2] that the ratio of the ionization rates depends on the laser intensity and increases as the intensity decreases. Increasing asymmetry of the ionization rates for larger values of the Keldysh parameter is a direct consequence of the nonadiabatic nature of the effect [1,2]. Thus, lower intensities at the wings of the pulse yield smaller ionization rates but larger ratios α and α'. Consequently, the laser intensity and the focal volume averaging can only enhance the ratio of the ionization rates. To illustrate this remark, we consider the spatiotemporal intensity profile of the Gaussian beam:

$$I(r,z,t) \sim \left[\frac{w_0}{w(z)}\right]^2 e^{-\frac{2r^2}{w(z)^2} - \frac{t^2}{2\sigma_t^2}}, \quad (2)$$

where w_0 is the waist size, $w(z) = w_0\sqrt{1 + z^2/z_R^2}$ is the z-dependent spot size, $z_R = \pi w_0^2/\lambda$ is the Rayleigh range, and σ_t is the temporal Gaussian width. Evaluating the corresponding averaged ratio

$$\bar{\alpha} = \frac{\int_0^\infty r\,dr \int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} w_+^{p_-}(I(r,z,t))\,dt}{\int_0^\infty r\,dr \int_{-\infty}^{\infty} dz \int_{-\infty}^{\infty} w_+^{p_+}(I(r,z,t))\,dt}, \quad (3)$$

that is independent of w_0, z_R, and σ_t, we obtain $\bar{\alpha} = 6.6$ and $\bar{\alpha}' = 6.7$ from Eq. (3). If only temporal averaging is performed, $\bar{\alpha} = 6.0$ and $\bar{\alpha}' = 6.3$. As we have anticipated, both averaged values are larger than the peak values obtained above, $\alpha = 5.8$ and $\alpha' = 6.2$. The averaged values are even closer to the experimentally estimated number $\alpha = \alpha' = 7.1$ and yield the theoretical result of the SDI ratio 3.4, in very good agreement with the experimental result 3.6. Thus, the experimental results [3] are in good agreement with the theoretical predictions [1,2].

Unfortunately, the theoretical predictions of [1,2] could not be tested in this experiment fully. Namely, the experiment has confirmed the existence of the asymmetry but the detection scheme could not establish the sign of the effect—the ionization preference of p_- vs p_+ or vice versa [3]. Herath *et al.* have suggested an interesting experiment utilizing an oriented atomic sample with known helicity. Such a sample can be produced by photodissociation on the excited state of a molecule such as ICl [5]. Here, we suggest another experiment for determining the value of α, using the spin-orbit splitting. Using the Stern-Gerlach-type experiment, four sublevels $M_{\frac{3}{2}} = \pm\frac{1}{2}, \pm\frac{3}{2}$ of a neutral iodine atom in the electronic ground state $^2P_{\frac{3}{2}}$ can be spatially separated and addressed separately [6]. Measuring only the ratio of the ionization rates α for each sublevel of the state $^2P_{\frac{3}{2}}$, one can extract the ionization rates for the orbitals p_m and determine the ionization preference of the valence orbitals p_+ or p_- in circularly polarized laser fields.

In summary, we have compared theoretical and experimental results for ratios of the ionization rates from valence orbitals $p_\pm$ of Ar and Ar^+ in circularly polarized laser fields and have found that they are in very good quantitative agreement, resolving the seeming discrepancy in the literature [3].

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