Ultrafast resolution of tunneling delay time

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Received 8 August 2014; revised 1 October 2014; accepted 4 October 2014 (Doc. ID 220663); published 14 November 2014

The question of how long a tunneling particle spends inside the barrier region has remained unresolved since the early days of quantum mechanics. The main theoretical contenders, such as the Buttiker–Landauer, Eisenbud–Wigner, and Larmor time, give contradictory answers. On the other hand, recent attempts at reconstructing valence electron dynamics in atoms and molecules have entered a regime where the tunneling time genuinely matters. Here, we compare the main competing theories of tunneling time against experimental measurements using the attoclock in strong laser field ionization of helium atoms. The attoclock uses a close to circularly polarized femtosecond laser pulse, mapping the angle of rotation of the laser field vector to time similar to the hand of a watch. Refined attoclock measurements reveal a real (not instantaneous) tunneling delay time over a large intensity regime, using two independent experimental apparatus. Only two theoretical predictions are compatible within our experimental error: the Larmor time and the probability distribution of tunneling times constructed using a Feynman Path Integral formulation. The latter better matches the observed qualitative change in tunneling time over a wide intensity range, and predicts a broad tunneling time distribution with a long tail. The implication of such a probability distribution of tunneling times, as opposed to a distinct tunneling time, would imply that one must account for a significant, though bounded and measurable, uncertainty as to when the hole dynamics begin to evolve. We therefore expect our results to impact the reconstruction of attosecond electron dynamics following tunnel ionization.

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OCIS codes: (020.2649) Strong field laser physics; (020.4180) Multiphoton processes; (240.7040) Tunneling.

http://dx.doi.org/10.1364/OPTICA.1.000343

1. INTRODUCTION

The tunneling time problem is almost as old as quantum mechanics itself and is a highly debated subject [1]. Time is not a quantum observable, and therefore many conflicting theories have been developed over the decades [1–5], but definitive experiments were largely impossible and awareness of the controversy in the broader scientific world faded. More recently within the ultrafast science community, tunneling time is implicitly assumed to be deterministic and, in fact, very often posited to be instantaneous or imaginary [6,7]. Hence, even the best techniques for imaging valence electron dynamics in atoms and molecules are founded on a deterministic time at ionization [6,8–10]. On the other hand, it is widely accepted in the tunneling-time community that the time is not instantaneous or even deterministic [1–3,5,11,12].

Our refined attoclock measurements using two independent measurement apparatus provide substantial advances in these key issues for both communities. Over a range of intensities, we show that only two prominent tunneling time theories are compatible with our experiments—and that the compatible probability distribution of times is broad enough to have significant impact on current experimental interpretation and reconstruction of hole dynamics.

2. ATTOCLOCK MEASUREMENTS

The attoclock definition of tunneling delay time relies on two independent measurements based on two observables.
Observable 1 is the polarization axis of the elliptically polarized light and observable 2 is the electron momentum vector. The angular orientation of the most probable momentum vector with respect to the polarization ellipse yields time delays as the hands of a watch indicate time (see [13] for further detail). Prior attoclock measurements [13] found an upper limit on tunneling time of around 40 as, but within a narrow intensity range, and hence observed no significant trends. More recent attoclock measurements in helium and argon covered high intensities, corresponding to very fast tunneling delay times, and therefore could not resolve tunneling time within experimental error [14]. Another recent experiment measured the time an electron involved in high harmonic generation (HHG) exits the barrier [6]. However, the absolute timing of ionization, and therefore the tunneling time, was not accessible [7].

Tunnel ionization occurs when a strong laser field bends the binding potential of an atom, creating a barrier [Fig. 3(c)] whereby electrons can tunnel out, be accelerated by the field, and eventually register their momenta at the detector (Fig. 1). An important parameter in strong field ionization is \( \gamma = \omega \sqrt{2 I_p/F} = \sqrt{I_p/2 U_p} \) [15], where \( I_p \) is ionization potential, \( U_p \) is ponderomotive energy, \( \omega \) is the central frequency of the laser, and \( F \) is the peak electric field, which divides the “vertical channel” of multiphoton ionization (\( \gamma \gg 1 \)) from the “horizontal channel” of optical tunneling (\( \gamma \ll 1 \)). Our experimental regime is in the \( 0.8 < \gamma < 2.5 \) range, corresponding to “nonadiabatic tunnel ionization” [16]. In this regime, while the tunneling probability may be substantially modified from the quasi-static rates [15,17], phase-independent contributions due to multiphoton absorption are small (approximately 3.3% of the total rate for \( \gamma \sim 2 \) [16]), and tunneling remains the dominant ionization mechanism [18], widely used to investigate molecular orbitals [8,19,20] and electron rearrangement [8–10,18] after ionization.

The momentum distributions were measured by two different instruments: a cold target recoil ion momentum spectrometer (COLTRIMS) [21] and a velocity map imaging spectrometer (VMIS) [22] (see Fig. 1). Figure 2 shows good overlap between COLTRIMS and VMIS, with error bars higher on COLTRIMS. The use of both apparatus gives access to a wide intensity range of \( 0.73–7.5 \times 10^{14} \text{ W/cm}^2 \), corresponding to variation in the barrier width by about a factor of 3, from 7.5 to 24 au (0.4–1.27 nm), with width given approximately by \( I_p/F \). Such large variation in barrier width resulted in a comparable variation of measured tunneling delay time (Fig. 3), allowing for a detailed comparison with theory. Tunneling delay time was extracted from measurements using attosecond angular streaking [13,23].

The experiment uses elliptically polarized laser light, with parameters given in Fig. 2. At lower intensities, attoclock [13,23] measurements were performed with VMIS with the gas nozzle integrated into the repeller plate [see Fig. 1(c)]. The integration of the gas nozzle allows one to achieve target gas densities that are significantly higher as compared to setups employing cold atomic beams. Two-dimensional projections of the photoelectron momentum distribution were recorded in steps of 2° covering 180°. The three-dimensional momenta...
distribution, and thus the electron momenta distribution in the polarization plane, was retrieved by tomographic reconstruction \([24]\), shown in Fig. 1(d) (more detail in Supplement 1). The COLTRIMS measurements were performed in a similar way as described elsewhere \([14]\).

3. EXPERIMENTAL EXTRACTION OF TUNNELING DELAY

The extraction of tunneling delay time follows the method first presented in \([13]\). The electron located at the peak of the momenta distribution, given by \(|\langle \hat{\mathbf{k}} | \Psi \rangle|^2\), corresponds to the most probable electron trajectory. To locate this peak from measurements, radial integration is used, combined with a double Gaussian fit (corresponding to two trajectories \([25, 26]\)) to extract the angle at which the maxima in the distribution occurs [see Figs. 1(a)–1(b)], corresponding to ionization at the peak of the laser field. This angle \(\theta_m\) corresponds to the direction of the most probable electron momentum in the plane of polarization. The measured offset angle, \(\theta_m - \pi/2\), shown in Fig. 2, is used to extract tunneling time after subtracting the Coulomb correction, \(\theta_{\text{Coul}}\), and the streaking angle, \(\theta_{\text{str}}\), which includes rotation due to the drift created by the vector potential of the electric field, resulting in the experimentally measured tunneling time, \(\tau\), given by \(\omega \tau = \theta_{\text{str}} = \theta_m - \theta_{\text{Coul}} - \theta_{\text{str}}\). The semiclassical simulations that were compared with experiment

![Fig. 2](image-url)

Fig. 2. Measured offset angle \(\theta_m - \pi/2\), from which tunneling time is extracted. Black line and dots correspond to the Coulomb correction obtained using the TIPIS model \([14]\) with single trajectory and classical trajectory Monte Carlo (CTMC) simulations, respectively (more in Section 2 of Supplement 1). The error bars are the result of a geometrical sum of the errors from the fit of the angular distribution of the electrons and the error of the polarization characterization. For the measurements performed with the COLTRIMS apparatus, an additional source of uncertainty comes from the thermal spread of the gas jet (more in Section 1 of Supplement 1).

![Fig. 3](image-url)

Fig. 3. Comparison of theory to experiment. (a) Five theoretical predictions of tunneling time compared to experiment. (b) Same as (a), but zoomed into the lower time delay. (c) Potential resulting from the combined Coulomb–laser field. (d) Tunneling time as a function of barrier width. The measured tunneling time is significantly smaller than the free propagation time for electrons with kinetic energies characteristic of electron motion in Helium: for example, around 40 as and 230 as for tunneling time and free propagation time, respectively, for barrier width of 13 au.
were based on the TIPIS model [14] (more detail in Section 2 of Supplement 1).

It was found in [27] that, for our experimental parameter range and ionization of Helium, TIPIS agrees within one degree (much less at higher intensities) with the semiclassical simulations based on the well-known nonadiabatic Perelomov, Popov, and Terentev model [28]. In calculating $\tau$, nonadiabatic effects and an offset of the streaking angle from 90° were taken into account, resulting in a minor (less than 5 as and up to 15 as at the lowest intensity) correction to tunneling time (see Section 2 of Supplement 1).

For direct comparison with the experiment, analytic calculations are done for the Fourier component, $k$, that maximizes $|\langle \hat{k}|\Psi \rangle|^2$, corresponding to the peak of the electron momenta distribution, from which the tunneling time is experimentally extracted. The tunneling process acts as a momentum filter that maps different Fourier components of the bound-state wavefunction onto different momenta, $k$, at the tunnel exit with probability $\propto \exp(-k^2/2\sigma^2)$ [17,19], where $k$ is the momentum transverse to the direction of tunneling. The maximum of $|\langle \hat{k}|\Psi \rangle|^2$ is given by the transmission of a $\Phi(x, k_z = 0)$ component, in the partial Fourier transform [29] of the bound-state wavefunction: $\Psi_{\delta}(x, y, z) = \int_\mathbb{R} \Phi(x, k_z) e^{-ik_z y} dk_z dz$, where $x$ is the major axis of polarization. The tunneling times were calculated for this component $\Phi(x)$ within the short-range potential approximation [29], taking account of nonadiabatic effects. Nonadiabatic effects arise when electrons absorb energy from the laser field. These effects are therefore expected to become more significant at lower intensities (corresponding to higher $\gamma$), where multiphoton contributions play a bigger role. We found the analytic results to be robust to variation in barrier shape (resulting, for example, from a Coulomb correction or nonadiabatic effects), as long as the barrier width did not change significantly. In short, small variations in barrier shape result in correspondingly small variations in tunneling time. The contribution of nonadiabatic effects to tunneling time estimates is relatively small (particularly at lower $\gamma$), even when the ionization rates are substantially offset from the quasi-static. This is partly due to the exponential dependence of ionization probability on barrier width, versus the linear dependence of tunneling time [Fig. 3(d)].

4. COMPARISON OF EXPERIMENTAL RESULTS WITH THEORETICAL DEFINITIONS OF TUNNELING TIME

The four widely used interpretations of tunneling delay times were calculated by finding the transmission amplitude $\Phi(x)$ for the propagation through the barrier, given by: $T = |T|e^{i\theta}$, and using the definitions $\tau_{BL} = -\hbar \partial \ln |T| / \partial V$; $\tau_{LM} = -\hbar \partial \theta / \partial V$; $\tau_{PM} = \hbar \partial \ln |T| / \partial E$; and $\tau_{EW} = \hbar \partial \theta / \partial E + w / k$ for Buttiker–Landauer [3,30], Larmor [5], Pollack–Miller [30], and Eisenbud–Wigner times [4], respectively, where $V$ is barrier height and $E$ is electron energy.

These four alternative interpretational pictures have been derived using very different physical considerations. The Buttiker–Landauer time corresponds to the time it would take a classical particle to propagate through an inverted potential barrier. The physical argument as to why it should be considered tunneling time rests on the fact that the ratio of $\tau_{BL}$ to the period of barrier modulation serves as a nonadiabaticity parameter (much as the Keldysh $\gamma$), and therefore must correspond to whether the electron sees a static or an oscillating field while it is traversing the barrier. The Larmor time corresponds to the degree of rotation of a spin-polarized electron inside a magnetic potential barrier. The Pollack–Miller time was derived using flux–flux correlation functions and represents an average correlation time in a collision. The Eisenbud–Wigner time follows the peak of a near-chromatic wavepacket, calculating the delay of this peak due to the presence of a potential. Hence, an additional term, $w / k$, is present in $\tau_{EW}$, where $w$ and $k$ are the barrier width and electron velocity, respectively. This additional term corresponds to the propagation of the electron in the barrier region if that barrier were absent, and has to be added to get the total time [2], since the first term only gives a relative time shift [4].

The Eisenbud–Wigner time has been used extensively [31,32] to explain the relative single photon ionization delay between ionization of valence and core electrons in Neon and Argon, first observed experimentally [33,34]. Perhaps counterintuitively, it is not the actual time to absorb a photon, but rather an additional phase shift in the peak of the propagating wave packet induced by the presence of the ionic potential after that photon is absorbed. Calculating the shift in the peak is straightforward in single-photon ionization, where the total energy of the electron is above threshold, leading to a well-defined single peak and propagation of the entire wavepacket. However, the Eisenbud–Wigner time is much more disputed in tunneling [1], where the peak is absent inside the barrier [2], and moreover, a large part of the wavefunction remains confined. Further discussion of the four tunneling times in the context of strong field ionization can be found in Section 3.1 of [35].

The four tunneling times are shown in Fig. 3(a), where we only consider the experimental range corresponding to $\gamma \leq 2$, where multiphoton contributions are believed to be relatively small [16]. Although these times were derived using very different physical models, they can be expressed as expectation values using the tunneling time probability amplitude, $f(\tau)$, constructed with Feynman Path Integrals (FPIs) [11,30]. The FPI approach is particularly appealing, because the total transmitted wavefunction can be expressed as a sum of all possible paths, each path corresponding to a deterministic tunneling time and contributing $\exp(iS[x(\tau)]/\hbar)$. The quantity $f(\tau)$ represents the contribution to the total transmission amplitude of only those paths that spend an amount of time, $\tau$, inside the barrier, such that $T = \int_0^\infty f(\tau) d\tau$. The ambiguity in interpretation of tunneling time using this approach arises because the tunneling time probability amplitudes, $f(\tau)$, are complex and interfere. As pointed out by Landauer and Martin [1], while no one disputes the construction’s accuracy, it is not clear what procedure to use for calculating relevant physical quantities with FPIs.
Fig. 4. Normalized distribution where each probability value corresponds to an integer number of attoseconds of tunneling time. (a) Probability distribution at intensity = 1.625 × 10^{15} \text{ W/cm}^2, FWHM ≈ 80 as, and skewness = 0.9. (b) Probability distribution at intensity = 6.5 × 10^{14} \text{ W/cm}^2, FWHM ≈ 50 as, and skewness = 1.09.

The four tunneling delay time definitions shown in Fig. 3 can be viewed as expectation values, rather than deterministic quantities [1,2,11,30]. In particular, the Buttker–Landauer and the Larmor times correspond to the absolute and the real parts, respectively, of the following complex-valued average [1,11]: \( \langle \tau \rangle = \int_0^\infty \tau f(\tau) d\tau / T \). This definition has been widely used, in part because it can be expressed as the transition element of the evolution of a hole \( \langle \tau \rangle = \langle \Psi_i | \tau | \Psi_f \rangle \), where \( \Psi_i \) and \( \Psi_f \) are normalizing incident and transmitted parts of the wavefunction, respectively. Perhaps surprisingly, \( \langle \Psi_i | \tau | \Psi_f \rangle \) is far too large and only the real part of \( \langle \tau \rangle \), equivalent to \( \tau_{LM} \), is within our experimental uncertainty. The agreement of the Larmor time with our experimental data suggests that phase accumulated before the electron appears at the tunnel exit (which is typically lost when using saddle-point or WKB-type approximations) is significant in determination of tunneling time.

In addition to computing expectation values with \( \langle f(\tau) \rangle \), we use it directly to construct the probability distribution of tunneling times, adapting a known method [12], shown in Fig. 4. The peak of this probability distribution is shown along with other theoretical predictions in Fig. 3. This peak corresponds better to the experimental observable (which is the peak of the recorded electron momenta distribution) than the expectation value given by the other tunneling time definitions. This is because trajectories that begin to tunnel at the peak but have longer or shorter tunneling times than the most probable trajectory will not end up at the peak of the momenta distribution, but will nevertheless be included in any averaging procedure that extracts the expected value of tunneling time.

5. CONCLUSION

Historically, measured tunneling time varied with the nature of the experiment. Besides the attoclock measurements, only one other experiment was done at the single-particle level [36], thereby avoiding the possibility of a pulse-reshaping process [1]. Our findings are consistent with this single-photon experiment [36], in that a particle moving through a potential barrier takes significantly less time than free propagation over the same distance. Therein this yielded superluminal velocities [36]; we get tunneling times that are just a small fraction of the free propagation time [though not superluminal, as Fig. 3(d) shows] for electrons with kinetic energies characteristic of electron motion in Helium.

As Fig. 3 shows, of the five theoretical approaches, two cannot be excluded: the Larmor time and the probability distribution of tunneling times constructed using FPIs. These two are not mutually exclusive. In particular, the correctness of Larmor time (viewed as an average, rather than a deterministic, quantity) allows the existence of a probability distribution of tunneling times. This view is further supported by the correspondence between Larmor time and the weak measurement value of tunneling time, given by \( \tau_{LM} = \Re(\langle \Psi_i | \tau | \Psi_f \rangle) \) [37]. The probability distribution shown in Fig. 4 has a long asymmetric tail that lengthens, along with an increase in the position of the peak and the full width-half-maxima (FWHM), as intensity decreases. This suggests that both uncertainty and expected duration of tunneling time increase at lower intensities, corresponding to a larger barrier width.

Our results imply that the probability distribution of tunneling time, at all measured intensities, may add significant uncertainty to reconstruction of attosecond electron dynamics after strong field ionization. In particular, the FWHM is larger or comparable, depending on intensity, to the “universal attosecond response to removal of an electron” of about 50 as, found computationally [38]. A second important issue is the loss of coherence of the hole if tunneling time is probabilistic rather than deterministic. A hole is created due to a superposition of states formed during the tunneling process. This hole subsequently evolves on the timescale determined by the energy splitting of nearby valence orbitals from which tunneling takes place. The loss of coherence will depend on the time scale of the evolution of a hole [9] relative to the uncertainty in tunneling time. Hence, relatively slow hole dynamics (such as 6.2 fs in Krypton [9,10]) will result in little loss of coherence. On the other hand, in some cases, (such as CO₂ where the hole period is as short as around 1.2 fs [8]) the loss of coherence can be significant. As Fig. 4 shows, the long tail of the distribution can extend to a substantial fraction of a hole period, implying a substantial loss of coherence.

The timescale of tunneling, once an unresolvable question for theorists of the foundations of quantum mechanics, therefore meaningfully affects the reconstruction of electron dynamics using HHG [8] or pump–probe experiments [18], which is
the primary goal of ultrafast science. The implications for both ultrafast experiment and theory, especially for molecules and at time resolutions of current and future interest [39], are at once fundamental, practical, and approachable with existing technology.

**FUNDING INFORMATION**

European Research Council Advanced Grant (ERC-2012-ADG_338 20120216); Marie Curie IIF; Marie Curie COFUND; NCCR Molecular Ultrafast Science and Technology (NCCR MUST); Swiss National Science Foundation (SNSF).

**ACKNOWLEDGMENT**

A. S. L. thanks B. R. Doran and S. Mishra for assistance and comments.

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See Supplement 1 for supporting content.

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