Tunneling phase time in photoionization: in search of a clock

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The electron phase time in laser-induced ionization is shown to permit an intuitive physical interpretation with a clear, transparent relation to the parameters of the potential barrier modulated by the laser field. We demonstrate that the phase time delay induced by the tunneling process can be isolated from the overall phase time of a tunneling electron by clocking the quantum electron wave-packet dynamics, as described by the time-dependent Schrödinger equation, against the classical Newtonian electron dynamics driven by the laser field. The electron phase delay time retrieved with the use of this clockwork is directly related to the width of the laser-field-controlled potential barrier. With this delay added, the quasiclassical model of laser-induced ionization and subsequent free-electron wave-packet dynamics is shown to accurately reproduce the results of the full quantum analysis. As physically discernable manifestations, the tunneling-induced phase delay time shows up in the energy distribution of photoelectrons and shifts the cutoff in the spectra of high-order harmonics emitted by recolliding electron wave packets. © 2016 Optical Society of America

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Since the birth of quantum mechanics, the problems and concepts related to time, its interpretation, and its measurement have been a focus of continuing debate [1–9]. Modern laser technologies enable time-resolved measurements on an unprecedented subfemtosecond time scale [10], offering access to the fastest processes of electron dynamics [11–14]. As laser-induced ionization plays a central role in a broad class of strong-field laser–matter interaction phenomena, electron tunneling once again becomes a focus of interest, as it serves a universal trigger, defining the time zero in many physical, chemical, and biological processes. In ultrafast optical physics, understanding the extremely fast dynamics of electron tunneling is critical to achieving the ultimate accuracy in attosecond metrology [10] and identifying the fundamental limitations of rapidly emerging petahertz optoelectronics [15–17].

Several experimental approaches have been developed in the past few years [18–23] to detect the dynamics of electron tunneling and to measure some of its key parameters. Physical interpretation of these parameters, however, encounters fundamental difficulties, as there is no consensus with regard to the possibility of direct tunneling time measurements and the uncertainty of such measurements in conventional quantum mechanics. The existing definitions of tunneling times, some of which are complex valued [5,6,24,25], are intended to quantify different aspects of tunneling, such as the phase delay or residence times. However, despite recent experimental breakthroughs toward a better understanding of electron arrival delays in laser-induced ionization [16,19,22,23], electron tunneling in the field of ultrashort laser pulses is especially difficult to interpret in terms of the available quantum approaches, since the complex-valued tunneling times originally defined for stationary potential barriers and plane-wave electron wave functions have to be applied to tunneling through potential barriers modulated by a rapidly oscillating laser field.

Here, we revisit the notion of the phase time for electron tunneling in laser-induced ionization. Our analysis suggests an intuitive physical interpretation for this time. We demonstrate that when clocked against the classical Newtonian electron dynamics driven by a laser field, the electron phase delay retrieved from the phase of the electron wave packet obeying the time-dependent Schrödinger equation is controlled by the width of the potential barrier. With this delay added, the semiclassical model of laser-induced photoionization and subsequent free-electron wave-packet dynamics accurately reproduces the results of the fully quantum analysis.

We start with a simple, yet instructive, insight into the phase time by representing the position of the maximum of a one-dimensional electron wave packet transmitted through a potential barrier [26] as \( z(t) = (p_0/m_e)t - \hbar(\partial \phi / \partial p)_{p_0} \), where \( t \) is the time, \( m_e \) is the electron mass, \( p_0 \) is the electron momentum right behind the barrier [26], and \( \phi \) is the phase shift of the wave packet induced by the transmission through the barrier. Since at the instant of time \( t \), a free-electron wave packet propagating without any potential barrier would have been centered at \( z_0(t) = (p_0/m_e)t \), the time delay acquired by the wave packet due to the potential barrier can be calculated as \( \Delta t = (m_e \hbar/p_0) \frac{\partial \phi}{\partial p} \) \( (\partial \phi / \partial p)_{p_0} \). With \( \phi \) understood as the phase of the wave function...
behind a potential barrier, the time Δt is known in the literature as the phase time after Bohm [26], Eisenbud [27], and Wigner [28].

This procedure, however, cannot be immediately extended to define the phase time in the case of tunneling ionization by a short-pulse laser driver. In that case, a tunneling electron is represented by a wave packet \( \psi_f(\vec{r}, t) = |\psi_f(\vec{r}, t)| \exp(i\eta(\vec{r}, t)) \) whose phase \( \eta(\vec{r}, t) \) acquires a complicated chirp in both space and time as an ultrashort laser pulse makes the electron tunnel through the field-modified potential barrier and then drives the subsequent electron wave-packet dynamics behind the barrier. In contrast to the regime where a transmitted electron propagates freely behind a stationary barrier, acquiring a simple phase shift \( \theta = (p_\theta^2/m_\theta) t - \phi \), in the case of laser-induced tunneling ionization, the phase shift component bearing the information on tunneling-induced delays needs to be isolated in a much more intricate phase \( \eta(\vec{r}, t) \), which includes the phase shift that an electron wave packet acquires as a part of its motion driven solely by an ultrabroadband laser driver field.

We will show now that this problem can be addressed by clocking the quantum electron wave-packet dynamics, as described by the time-dependent Schrödinger equation, against the classical Newtonian electron dynamics driven by the laser field. To this end, we first solve the time-dependent Schrödinger equation (TDSE) for the wave function \( \psi_f(\vec{r}, t) \) (where \( \vec{r} \) is the radial coordinate and \( t \) is the time) in the presence of the Coulomb potential of a hydrogen-like atom and a single-cycle laser field that is assumed to be linearly polarized along the \( z \) axis. The solution to the TDSE is represented as a sum, \( \psi_f(\vec{r}, t) = \psi_0(\vec{r}, t) + \psi_f(\vec{r}, t) = \sum_{n=0}^{\infty} \sum_{l=0}^{\infty} \alpha_n(l(t) u_{n,l}(\vec{r}) + \psi_f(\vec{r}, t) \) of negative- and positive-energy terms, \( \psi_0(\vec{r}, t) \) and \( \psi_f(\vec{r}, t) \), corresponding to the bound and free states of an electron [29–31], with \( u_{n,l}(\vec{r}) \) being the orthonormalized eigenfunctions of the stationary Schrödinger equation for a hydrogen-like atom. The analysis presented here is applicable to multielectron systems as long as the core electrons adiabatically follow field variations induced by the tunneling electron. In practice, this requirement is usually reduced to the inequality [32] \( \omega < \omega_j \), where \( \omega \) is the laser frequency and \( \omega_j \) is the frequency of transition between the ground and the first excited states of the ionized system that the electron tunneling leaves behind.

To facilitate comparison with the one-dimensional analysis of tunneling times, we reduce the treatment of the electron wave packet to one dimension by integrating the wave function \( \psi_f(z, \rho, t) \), found by numerically solving the TDSE, over the transverse coordinate \( \rho \). \( \psi_f(z, \rho, t) = \int_0^\infty \psi_f(z, \rho, t) d\rho \). We take \( t = T_0/2, T_0 \) being the field cycle, in most of our calculations, as the shape of tunneling electron wave packets can be clearly resolved after a field half-cycle. The wave function of the electron wave packet \( \psi_f(\vec{r}, t) \) calculated for different field intensities and carrier wavelengths at this moment of time is presented in Figs. 1(a)–1(c). Stronger driver fields and longer driver wavelengths are seen to increase the spatial extension of the electron wave packet. The momentum-representation transform of this wave packet, \( \tilde{\psi}_f(p_z, t) \), defines the distribution function of electrons transmitted through the potential barrier in the momentum \( p_z \) and, hence, energy \( E = (p_z^2/2m) \), \( T_{q0}(e) = |\tilde{\psi}_f(p_z, t)|^2 \), as shown in Figs. 1(a)–1(f).

With the phase \( \eta(p_z, t) \) of the electron wave packet \( \psi_f(p_z, t) \) taken in the momentum representation, the derivative, \( \hbar \partial \eta(p_z)/\partial p_z \), can be interpreted as the position \( z(p_z) \) of electrons with the momentum \( p_z \) and, hence, energy \( E = (p_z^2/2m) \) [blue lines in Figs. 1(d)–1(f)]. The inverse function \( e(z) \) defining the electron energy as a function of the electron position, calculated from the phase \( \eta(z, t) \), is shown as blue lines in Figs. 1(a)–1(c). The overall phase time delay of the electron wave packet can now be calculated as \( t_{ph}(e) = (m_\psi/\hbar^2) \partial \eta(p_z)/\partial p_z \) by applying the Bohm–Eisenbud–Wigner recipe to the phase \( \eta(p_z, t) \). The results of these calculations are plotted by blue lines in Figs. 2(a)–2(c). The spatial chirp of the electron wave packets in Figs. 1(a)–1(c), giving rise to a growing phase, is understood in terms of a simple picture where electrons with higher energies propagate further along \( z \) within a fixed time interval.

In the search for an appropriate clock that would help isolate the phase delay \( \Delta t_{ph}(e) \) solely due to tunneling in the overall phase \( \eta(p_z, t) \), we resort to a classical model, which treats an electron as a classical particle obeying the laws of Newtonian dynamics. In such a model, an electron that exits the barrier at the moment of time \( t_b \) acquires by the moment of time \( t \) a velocity \( v_{\psi-f}(t_b, t) = \int_{t_b}^{t} (E_0/\hbar^2) \sin[\eta(\vec{r}, \rho, t)/\hbar] \) d\tau. For \( t = T_0/2 \), this formula gives \( v_{\psi-f}(t_b) = (E_0/m_\psi)[1 + \cos(\eta(\vec{r}, \rho, t)/\hbar)] \).

The position of such an electron at \( t = T_0/2 \) is given by \( z_{\psi-f}(t_b) = (E_0/m_\psi)(T_0/2 - t_b) - \sin(\eta(\vec{r}, \rho, t)/\hbar) \). These classical solutions for \( v_{\psi-f}(t_b) \) and \( z_{\psi-f}(t_b) \) define the energy, \( E = m_\psi v_{\psi-f}^2/2 \), that an electron acquires at the moment of time \( t = T_0/2 \) as a function of its position [green dash-dotted line in Figs. 1(a)–1(c)]. On the other hand, solving the velocity equation for \( t_b \), we find the electron position at \( t = T_0/2 \) as a function of its energy \( E \).
of whether the tunneling phase time delay can have any discernible physical manifestations. In our search for the answer to this question, we consider a standard scenario of high-order harmonic generation [34,35], where electrons undergo tunneling photo-ionization and recollide with parent ions as part of their quasi-classical motion driven by a high-intensity laser field. The harmonic spectrum emitted by recolliding electrons features an extended plateau whose high-frequency cutoff is controlled by the ionization potential of the atom and the ponderomotive energy of recolliding electrons [34,35].

Two approaches have been used to analyze high-harmonic generation. In the first approach, the TDSE was used to calculate the dipole moment and its second-order time derivative responsible for harmonic generation [Figs. 3(a) and 3(b)]. Within each cycle of the driver field [the dashed line in Figs. 3(a) and 3(b)], electrons ionized within the first field half-cycle recollide with their parent ions within the second field half-cycle, emitting radiation with energy $E_{ph}(t)$. The ponderomotive energy of these electrons increases toward the end of the second field half-cycle, giving rise to chirped radiation [blue solid line in Figs. 3(a) and 3(b)]. The maximum photon energy, achieved toward the end of the driver field cycle [Figs. 3(b) and 3(c)], defines the cutoff $E_c$ in the harmonic spectra. For longer driver wavelengths, the cutoff is seen to be shifted toward higher frequencies, extending the plateau in the harmonic spectra, in agreement with earlier studies [36].

In the second, quasiclassical approach, the Newtonian dynamics of recolliding electrons driven by the laser field [34] is combined with an adiabatic model of ionization [31,33], yielding instantaneous ionization with no delay. For an electron ionized at the instant of time $t_b$, such a quasiclassical model dictates that $z(t) = (eE(t)/(m\omega))(\sin(\omega t) - \sin(\omega t_b))/\omega - (t - t_b)\cos(\omega t_b)$. Equating $z(t)$ to zero for a recollision event, the electron is ionized at the instant of time $t_b$, and the quasiclassical model predicts the time of recollision $t_r$. With this correction included, the position of the electron in the classical model [red dashed lines in Figs. 1(a)–1(f)] agrees very well with $z_{cl} = h\partial^2z_e/\partial p_e$, calculated in the full TDSE-based quantum model [blue lines in Figs. 1(a)–1(f)] within the entire spectrum of electron energies [Figs. 1(d)–1(f)].

![Image](https://via.placeholder.com/150)

**Fig. 2.** (a)–(c) Energy distribution of continuum-state electrons $T(e)$ at $t = T/2$ and the phase time (right axis) as a function of the electron energy calculated by using the TDSE-based quantum model (blue line) and the classical model with (red dashed line) and without (green dash-dotted line) correction for the nonzero exit point of electrons for $I_0 = 200$ TW/cm$^2$ [(a), c] and $400$ TW/cm$^2$ (b) and $\lambda_0 = 0.8$ μm [(a, b)] and 1.6 μm (c). (d, e) The difference $(z_{cl}(e)) - (z_{ad}(e))$ of the positions $z_{cl}$ and $z_{ad}$ as a function of the driver field intensity for (a) $\lambda_0 = 0.8$ μm, (b) 1.2 μm, and (c) 1.6 μm. The width of the potential barrier $D \approx I_p/(eeE_0)$ for a hydrogen atom ($I_p = 13.6$ eV) is shown by the dashed line.
solving this equation for \( t \), and plugging the result into the electron kinetic energy, \( E_k(t) = m_e \left[ e E_0 \cos(\omega t) - \cos(\omega t) \right] / m_e \omega^2 / 2 \), we find \( E_k(t) = I_p + E_k(t) \) [green dash-dotted lines in Figs. 3(b) and 3(c)]. The maximum value of \( E_k(t) \) calculated with this approach [shown with an arrow in Fig. 3(c)], is precisely \( E_k = I_p + 3.17 U_p \) being the average electron pondermotive energy, thus reproducing the cutoff of harmonic spectra [34].

Comparison of the fully quantum, TDSE-based analysis [blue lines in Figs. 3(b) and 3(c)] with quasiclassical calculations [green dash-dotted lines in Figs. 3(b) and 3(c)] reveals a clear discrepancy between the predictions of these two models. To identify the origin of this discrepancy, we add a correction to the classical equation of motion accounting for the nonzero electron exit point, \( z_{e\alpha}(t) = I_p/(eE(t)) \). This correction translates into a delay of the recollision time \( \Delta t_r \), as shown in Fig. 3(c).

With the \( z_{e\alpha}(t) \) correction included, predictions of the quasiclassical model agree very accurately with the fully quantum model, which indicates that the shift of propagating wave packets, which is infamously unreliable as a physical measurable [5, 6], allowing this time to be related to other, less fuzzily defined measurable parameters, such as a cutoff shift in high-harmonic spectra and the energy distribution of photoelectrons [see Supplement 1].

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See Supplement 1 for supporting content.

**REFERENCES**

1. E. U. Condon and P. M. Morse, Rev. Mod. Phys. 3, 43 (1931).

2. L. A. MacColl, Phys. Rev. 40, 621 (1932).

3. M. Böttiker and R. Landauer, Phys. Rev. Lett. 49, 1739 (1982).

4. A. M. Steinberg, Phys. Rev. Lett. 74, 2405 (1995).

5. R. Landauer and T. Martin, Rev. Mod. Phys. 66, 217 (1994).

6. E. H. Hauge and J. A. Stovneng, Rev. Mod. Phys. 61, 917 (1989).

7. A. S. Landsman and U. Keller, Phys. Rev. 547, 1 (2015).

8. L. DiMauro, M. Frolov, K. L. Ishikawa, and M. Ivanov, J. Phys. B 47, 030301 (2014).

9. L. Torlina, F. Morales, J. Kauhal, I. Ivanov, A. Khelifa, A. Zielinski, A. Scrinzi, H. G. Muller, S. Sukiasyan, M. Ivanov, and O. Smirnova, Nat. Phys. 11, 503 (2015).

10. P. B. Corkum and V. Krausz, Nat. Phys. 3, 381 (2007).

11. H. Akagi, T. Tobbe, A. Staudte, A. Shinier, F. Turner, R. Dörner, D. M. Villeneuve, and P. B. Corkum, Science 325, 1364 (2009).

12. M. Schultz, M. Fieß, N. Karpowicz, J. Gagnon, M. Korbman, M. Hofstetter, S. Neppel, A. L. Cavaliere, Y. Komminos, T. Mercouris, C. A. Nicolaides, R. Pazourek, S. Nagele, J. Feist, J. Burgdörfer, A. M. Azzeer, R. Ernstorfer, R. Kienberger, U. Kleinbegner, E. Goulielmakis, F. Krausz, and V. S. Yakovlev, Science 328, 1658 (2010).

13. J. Mauritsen, T. Remetter, M. Swoboda, K. Klünder, A. L’Huillier, K. J. Schafer, O. Ghafer, F. Kelkensberg, W. Siu, P. Johnsson, M. J. J. Vrakking, I. Znakovskaya, T. Uphues, S. Zherebtsov, M. F. Kling, L. Lépine, E. Benedetti, F. Ferrari, G. Sansone, and M. Nisoli, Phys. Rev. Lett. 105, 053001 (2010).

14. M. T. Hassan, T. T. Luu, A. Moulet, O. Raskazovskaya, P. Zhokhov, M. Garg, N. Karpowicz, A. M. Zelhtikov, V. Pervak, F. Krausz, and E. Goulielmakis, Nature 530, 66 (2016).

15. M. Schultz, E. M. Bothschafter, A. Sommer, S. Holzner, W. Schweinberger, M. Fieß, M. Hofstetter, R. Kienberger, V. Apalkov, W. S. Yakovlev, M. I. Stockman, and F. Krausz, Nature 493, 75 (2012).

16. F. Krausz and M. I. Stockman, Nat. Photonics 8, 205 (2014).

17. H. Mashiko, K. Oguri, T. Yamauchi, A. Suda, and H. Gotoh, Nat. Phys. 12, 741 (2016).

18. M. Ulbracker, T. Uphues, M. Schultz, A. Verhoeef, V. Yakovlev, M. Kling, J. Rauschenberger, N. Bakachnik, H. Schröder, M. Lezius, K. Kompa, H.-G. Muller, M. Vrakking, S. Hendel, U. Kleinbegner, U. Heinmznn, M. Dresher, and F. Krausz, Nature 446, 627 (2007).

19. P. Eckle, A. N. Pfeiffer, C. Cirelli, A. Staudte, R. Dörner, H. G. Muller, M. Böttiker, and U. Keller, Science 322, 1525 (2008).

20. A. J. Verhoeof, A. V. Mitrofanov, E. E. Serebryannikov, D. V. Kartashov, A. M. Zelhtikov, and A. Baltuška, Phys. Rev. Lett. 104, 163904 (2010).

21. A. V. Mitrofanov, A. J. Verhoeof, E. E. Serebryannikov, J. Lumeau, L. Glebov, A. M. Zelhtikov, and A. Baltuška, Phys. Rev. Lett. 106, 147401 (2011).

22. A. N. Pfeiffer, C. Cirelli, M. Smolarski, D. Dimitrovski, M. Abusamha, L. B. Madsen, and U. Keller, Nat. Phys. 8, 76 (2011).

23. D. Shafir, H. Soifer, B. D. Bruner, M. Dagan, Y. Mairesse, S. Patchkovskii, M. Y. Ivanov, O. Smirnova, and N. Dudovich, Nature 485, 343 (2012).

24. L. V. Keldysh, Zh. Eksp. Teor. Fiz. 47, 1945 (1964).

25. D. Sokolovski and J. N. L. Connor, Phys. Rev. A 42, 6512 (1990).

26. D. Bohm, Quantum Theory (Prentice-Hall, 1951).

27. L. Eisenbud, “Formal properties of nuclear collisions,” Ph.D. dissertation (Princeton University, 1948).

28. E. P. Wigner, Phys. Rev. 98, 145 (1955).

29. M. Nurhuda, A. Suda, and K. Midorikawa, New J. Phys. 10, 053006 (2008).

30. P. Béjot, E. Cormier, E. Hertz, B. Lavorel, J. Kasparian, J.-P. Wolf, and O. Faucher, Phys. Rev. Lett. 110, 043902 (2013).

31. E. E. Serebryannikov and A. M. Zheltikov, Phys. Rev. Lett. 113, 043901 (2014).

32. T. Brabec, M. Cote, P. Boulanger, and L. Ramunno, Phys. Rev. Lett. 98, 073001 (2005).

33. A. Karamatskou, S. Pabst, and R. Santra, Phys. Rev. A 87, 043422 (2013).

34. P. B. Corkum, Phys. Rev. Lett. 71, 1994 (1993).

35. M. Lewenstein, P. Balcou, M. Y. Ivanov, A. L’Huillier, and P. B. Corkum, Phys. Rev. A 49, 2117 (1994).

36. P. Colosimo, G. Doumy, C. I. Blaga, J. Wheeler, C. Hauri, F. Catoire, J. Tate, R. Chirila, A. M. March, G. G. Paulus, H. G. Muller, P. Agostini, and L. F. DiMauro, Nat. Phys. 4, 388 (2008).

37. A. S. Landsman, M. Weig, J. Maurer, R. Boge, A. Ludwig, S. Heuser, C. Cirelli, L. Gallmann, and U. Keller, Optica 1, 343 (2014).