Journal of Physics Communications

OPEN ACCESS

RECEIVED
3 September 2024

REVISED
8 January 2025

ACCEPTED FOR PUBLICATION
30 January 2025

PUBLISHED
11 February 2025

PAPER

Universal behavior of tunneling time and barrier time-delay decoupling in attoclock measurements

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Keywords: ultrafast science, attosecond physics, tunneling and tunnel ionization time-delay, nonadiabatic effects, weak measurement, larmor clock and interaction time

Abstract

The measurement of the tunneling time-delay is hotly debated and remains controversial. In previous works, we showed that a model that accurately describes the time-delay measured by the attoclock experiment in adiabatic and nonadiabatic field calibrations. In the present work, we show that the tunneling time reveals a universal behavior with disentangled contributions. Even more remarkable is that the barrier tunneling time-delay can be convincingly defined and determined from the difference between the time-delay of adiabatic and nonadiabatic tunnel-ionization, which also show good agreement with experimental results. Furthermore, we illustrate that in the weak measurement limit, the barrier time-delay corresponds to the Larmor-clock time and the interaction time within the barrier.

1. Theoretical model

The interaction of an atom with a laser pulse in strong-field and attosecond science can be modeled in a simplified manner [1–4] as shown in figure 1. In [1] a simple tunneling model is developed with expressions involving basic laser and atomic parameters that describe the measurement result of the attoclock. This model provides insight into the temporal properties of tunneling ionization and sheds light on the role of time in quantum mechanics. We briefly present our model below, in which an electron can be tunnel-ionized by a laser pulse with an electric field strength (hereafter field strength) F. Throughout this work F stands for *the peak electric field strength at maximum* (quasistatic approximation [5]), and atomic units (*au*) are used ($\hbar = m = e = 1$). A direct ionization happens when the field strength reaches a threshold called atomic field strength $F_a = I_p^2/(4Z_{\text{eff}})$ [6, 7], where I_p is the ionization potential of the system (atom or molecule) and Z_{eff} is the effective nuclear charge in the single-active electron approximation (SAEA). However, for $F < F_a$ the ionization can happen by tunneling through an effective potential barrier including the Coulomb potential of the core and the electric field of the laser pulse. It can be expressed in a one-dimensional form in the length gauge due to the Göppert-Mayer gauge-transformation [8] by

$$V_{\text{eff}}(x) = V(x) - xF = -\frac{Z_{\text{eff}}}{x} - xF, \tag{1}$$

Compare figure 1. In the model the tunneling process can be described solely by the ionization potential I_p of the valence (interacting) electron and the peak field strength F, where the barrier height is given by $\delta_z = \sqrt{I_p^2 - 4Z_{\text{eff}}F}$. In figure 1 (for details see [1]), the inner $x_{e,-}$ and outer $x_{e,+}$ points are given by $x_{e,\pm} = (I_p \pm \delta_z)/2F$. The inner and outer points are called entrance and exit points, respectively. The barrier width is $d_B = x_{e,+} - x_{e,-} = \delta_z/F$, and its (maximum) height (at $x_m(F) = \sqrt{Z_{\text{eff}}/F}$) is δ_z. At $F = F_a$ we have $\delta_z = 0$ ($d_B = 0$), i.e., the barrier disappears and the direct (barrier-suppression) ionization begins, (green (dashed-doted) curve in figure 1).

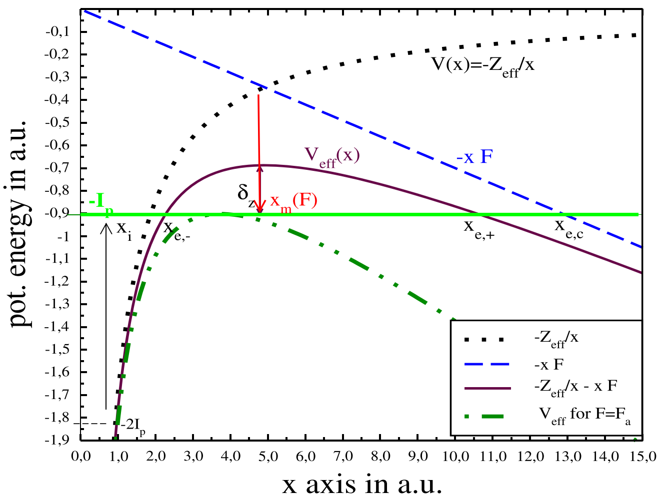

Figure 1. A sketch of the atomic and the effective potential curves, showing the inner $x_{e,-}$ and outer $x_{e,+}$ points, the width and height of the barrier formed by the laser field. $x_{e,\pm} = (I_p \pm \delta_z)/2F$, $d_B = x_{e,+} - x_{e,-} = \delta_z/F$, $\delta_z = \sqrt{I_p^2 - 4Z_{eff}F}$. The plot is for He-atom in the SAEA model with $Z_{eff} = 1.6875$ and $I_p \approx 0.9$ *au*. For different systems the overall picture remains the same. Reprinted (figure) with permission from [1], Copyright (2015) by the American Physical Society. Reproduced from [2]. © The Author(s). Published by IOP Publishing Ltd. CC BY 3.0.

1.1. Adiabatic tunneling

In the adiabatic field calibration by Landsman *et al* [9], we showed in [1] that the tunneling time-delay can be expressed by the following forms,

$$\tau_{T,d} = \frac{1}{2(I_p - \delta_z)}, \quad \tau_{T,i} = \frac{1}{2(I_p + \delta_z)} \tag{2}$$

In [1] was also shown that $\tau_{T,d}$ in equation (2) agrees well with the experimental result of Landsman *et al* [9]. Their physical reasoning is the following: $\tau_{T,d}$ is the time-delay of the adiabatic tunnel-ionization, with respect to the ionization at atomic field strength F_a, required to overcome the barrier and escape the atom into the continuum [1]. Furthermore, $\tau_{T,i}$ is the time needed to reach the entrance point $x_{e,-}$ from the initial point x_i, compare figure 1. At the limit of atomic field strength, we have $\lim_{F \to F_a} \delta_z = 0$, $\lim_{F \to F_a} \tau_{T,d} = \frac{1}{2I_p} = \tau_a$ (see below). For $F > F_a$, the barrier-suppression ionization sets on [10, 11]. On the opposite side, we have $\lim_{F \to 0} \delta_z = I_p$, $\lim_{F \to 0} \tau_{T,d} = \infty$, so nothing happens and the electron remains in its ground state undisturbed, which shows that our model is consistent. For details, see [1–3, 12, 13].

1.2. Nonadiabatic tunneling

In the nonadiabatic field calibration of Hofmann *et al* [14], we found in [4] that the time-delay of the nonadiabatic tunnel-ionization is descried by the relation

$$\tau_{dion}(F) = \frac{1}{2}\frac{I_p}{4Z_{eff}F} = \frac{1}{2I_p}\frac{I_p^2}{4Z_{eff}F} \tag{3}$$

$$= \frac{1}{2I_p}\frac{F_a}{F} = \tau_a\, \xi(F) \tag{4}$$

In [4] it was shown that equation (3) agrees well with the experimental result of Hofmann *et al* [14]. In addition, the result was confirmed by the numerical integration of the time-dependent Schrödinger equation (NITDSE) [4]. It is seen that in the limit $F \to F_a$, $\tau_{dion} = \tau_a$, whereas in the opposite case $F \to 0$, $\tau_{dion} = \infty$, similar to $\tau_{T,d}$. Hence, τ_a is always (adiabatic and nonadiabatic) a lower quantum limit of the tunnel-ionization time-delay and does not quantum mechanically vanish. $\xi(F)$ is an enhancement factor for field strength $F < F_a$.

2. The barrier time-delay

We have seen in the previous section that the experimental results of the tunneling time-delay, in the two field calibrations (adiabatic [9], nonadiabatic [14]), are in good agreement with our tunneling model, as given by the adiabatic and nonadiabatic tunneling time-delay $\tau_{T,d}$ and τ_{dion} of equations (2), (3) respectively.

In this section, we show that the time-delay due to the barrier itself, which can be considered as the actual tunneling time-delay, can be decoupled and determined by considering the adiabatic and nonadiabatic field calibrations together.

The time-delay due to the barrier itself is, in fact, the time-delay due to crossing the barrier region. However, the experiment does not verify whether the time spent in the barrier is affected by some reflections of the electrons inside the barrier before they escape the atom (see further below equation (8)). Apart from that, multiple reflections are related the regime of high harmonics generation, where electrons recombine by emitting high harmonics. In the attoclock experiment, the momentum distribution of photoelectrons (ions) during the tunnel-ionization process is considered to extract and determine the delay-time [15, 16].

Equation (2) can be decomposed into a twofold time-delay with respect to ionization at F_a, representing $\tau_{\mathrm{T,d}}$ in an unfolded form,

$$\begin{aligned}\tau_{\mathrm{T,d}} &= \frac{1}{2(I_{\mathrm{p}}-\delta_z)} = \frac{1}{2}\frac{I_{\mathrm{p}}}{4Z_{\mathrm{eff}}F}\left(1+\frac{\delta_z}{I_{\mathrm{p}}}\right)\\ &= \frac{1}{2I_{\mathrm{p}}}\frac{F_a}{F}\left(1+\frac{\delta_z}{I_{\mathrm{p}}}\right) = \tau_a(\xi(F)+\Lambda(F))\\ &= \tau_{\mathrm{dion}} + \tau_{\mathrm{dB}} \equiv \tau_{\mathrm{Ad}}\end{aligned} \tag{5}$$

Below we refer to $\tau_{\mathrm{Ad}} \equiv \tau_{\mathrm{T,d}}$ as adiabatic tunnel-ionization. From the second line of equation (5) follows that the adiabatic time-delay can be interpreted as a time-delay with respect to ionization at atomic field strength with $\tau_a = \tau_{\mathrm{Ad}}(F_a) = 1/(2I_{\mathrm{p}})$, where both terms ($\tau_{\mathrm{dion}}$, τ_{dB}) present real time-delay. Again $(\xi(F)+\Lambda(F))$ is an enhancement factor for field strength $F < F_a$.

The first term in the last line of equation (5), τ_{dion}, which is given in equation (3), is a time-delay solely because F is smaller than F_a and saturates at $F = F_a$, whereas the second term τ_{dB} is a time-delay due to the barrier itself and thus, can be considered as the actual tunneling time-delay [3]. We will show that it can be decoupled from the first term and is thus the tunneling time solely to the presence of the barrier itself. This can be seen from the factor δ_z/I_{p}, which relates the barrier height for $F < F_a$ to the ionization potential, which in turn represents the maximum barrier height ($\lim_{F\to 0}\delta_z = I_{\mathrm{p}}$). It vanishes when the barrier $\lim_{F\to F_a}\delta_z = 0$ vanishes, i.e. when saturation is reached at $F = F_a$.

Indeed, Winful [17] and Lunardi [18] proposed expressions that resemble the adiabatic tunnel-ionization time-delay given in equation (5).

$$\tau_g^{\mathrm{Winf}} = \tau_{\mathrm{si}} + \tau_{\mathrm{dwell}} \tag{6}$$

$$\tau_T^{\mathrm{Lun}} = \tau_{\mathrm{well}} + \tau_{\mathrm{barrier}} \tag{7}$$

In the quantum tunneling of a wave packet or a flux of particles scattering on a potential barrier, Winful showed that the group time-delay or the Wigner time-delay can be written in the form of equation (6), where τ_{dwell} is the dwell time which corresponds to our τ_{dB}, and τ_{si} is according to Winful a self-interference term which corresponds to our τ_{dion}. Importantly, Winful showed that the contribution of the first term is disentangled from the barrier time-delay [17]. In their work Lunardi *et al* used the so-called Salecker-Wigner-Peres quantum-clock (SWP-QC) with MC-simulation [18, 19]. They found that the tunneling time in the attoclock is given by the form of equation (7). The first term, which they called well-time is, according to the authors, the time (-delay) that elapses before reaching the barrier. This corresponds to our τ_{dion}. The second term, τ_{barrier}, corresponds to the barrier time (-delay) which corresponds to our τ_{dB}. The three results (ours confirmed with the NITDSE, Winful's and Lunardi's) clearly show that the separation of equation (5) presents a unified T-time picture (UTTP) [17], which allows us to conclude that the tunneling time-delay due the barrier itself is given by $\tau_{\mathrm{dB}} = \tau_{\mathrm{dwell}} = \tau_{\mathrm{barrier}}$ and can be determined from equations (3), (5) as follows:

$$\tau_{\mathrm{Ad}} - \tau_{\mathrm{dion}} = \tau_{\mathrm{dB}} = \frac{1}{2}\frac{1}{4Z_{\mathrm{eff}}}\frac{\delta_z}{F} = \frac{1}{2}\frac{d_{\mathrm{B}}}{4Z_{\mathrm{eff}}} \tag{8}$$

In equation (8), τ_{dB} is the barrier (tunneling) time-delay, usually referred to as the time spent within the barrier, τ_{barrier} by Lunardi or the dwell time τ_{dwell} by Winful. One also finds a linear dependence of τ_{dB} on the barrier width d_{B}. The barrier time-delay tends to infinity as $F \to 0$, leaving the stationary ground state of the atom undisturbed. The barrier time-delay tends to zero in the limit of $F \to F_a$ ($\delta_z = d_{\mathrm{B}} = 0$) because the barrier disappears, as is known from the seminal work of Hartman [20], where the linear dependence on the barrier width in equation (8) indicates that multiple reflections (if they exist) have a negligible influence on the tunnel-ionization. Obviously, τ_{dB} cannot be measured by the experiment directly since the first term is always present, τ_{dion}, τ_{si}, τ_{well}. However, as seen in equation (8), it can be determined taking both field calibrations into account, thus decoupling the barrier (tunneling) time-delay τ_{dB} in the tunnel-ionization of the attoclock experiment.

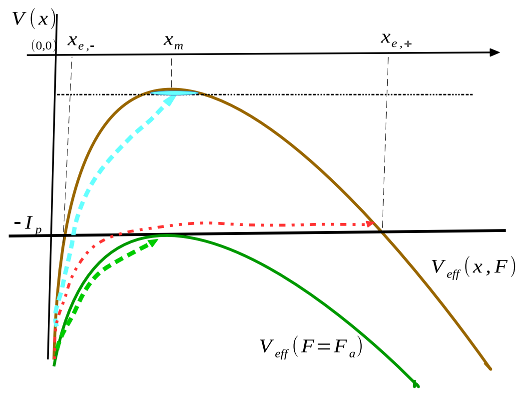

Figure 2. Illustration of tunnel-ionization of the horizontal (dashed-dotted, red) and vertical (dashed, blue) channels. The dotted line is a virtual state, as well as the BSI (green). Compare figure 1.

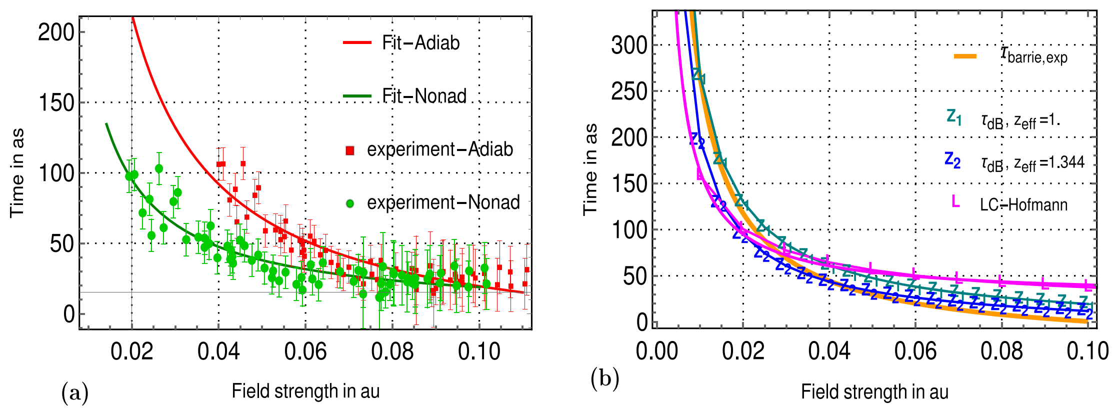

Figure 3. (a): Tunnel-ionization time-delay versus field strength. Experimental data of adiabatic [9] (red) and nonadiabatic [14] (green) field calibrations with their data-fit curves. (b): barrier time-delay $\tau_{\text{barrier,exp}}$ (orange) obtained by subtractions of the data-fit curves (shown left), and the barrier time-delay τ_{dB} of equation (8) for $Z_{\text{eff}} = 1, 1.344$ (green, blue). As well as the LC-curve (magenta) obtained by data-fit of LC-data given in [14].

It is worth noting that the decomposition given in equation (5) was derived in [3] concerning controversial issue of a quantum operator. However, its significance emerged when we found in [4] that the first term τ_{dion} agrees well with the experimental result of Hofmann [14] in nonadiabatic field calibration and the numerical integration of time-dependent Schrödinger equation. Furthermore, the relevance of numerical results of Winful and Lunardi in equations (6)-(7) was not clear for experimental finding of the attoclock. Our model and the present work are of crucial importance as we argue for a conceivable method to define and determine the barrier time delay, which is a hotly debated topic.

An illustration of the tunnel-ionization of adiabatic (horizontal) and nonadiabatic (vertical) channels is shown in figure 2. Fortunately, for the He atom we can consider the results of Landsman [9] (adiabatic calibration) and Hofmann [14] (nonadiabatic calibration) since they belong to the same experiment [15, 16]. In figure 3 (a), we show both experimental results with curves obtained by data fitting. The curves show a $1/F$-dependence as expected, subtracting them from each other gives the experimental barrier time, which is denoted by $\tau_{\text{barrier,exp}}$.

In figure 3 (b), we compare the barrier (tunneling) time-delay τ_{dB} of equation (8) with the experimental counterpart $\tau_{\text{barrier,exp}}$ (the orange curve). We plot τ_{dB} for two $Z_{\text{eff}} = 1.0,\ 1.344 = \sqrt{2I_{\text{p}}}$, expanding the range to a larger barrier width (smaller F) than specified in the experimental data. As can be seen in figure 3 (b), the agreement is very good. It undoubtedly shows that the time spent in the barrier (the time-delay caused by the barrier upon tunnel-ionization in attosecond experiment) is τ_{dB}, which can be determined (from the experimental data) by $\tau_{\text{barrier,exp}}$. The agreement (with $\tau_{\text{barrier,exp}}$ the orange curve) becomes better for $Z_{\text{eff}} = 1$ (Z_1 green curve) in the region of small field strength, since the barrier width $d_{\text{B}} = \delta_z/F$ is large and the tunnel-ionized electron escapes far from the atomic core. For larger field strength (near the atomic field strength) the barrier width is small and the agreement is better for $Z_{\text{eff}} = \sqrt{2I_{\text{p}}} = 1.344$ (Z_2 blue curve). Finally, all curves tend to zero for $F \to F_a$, since the barrier disappears as expected.

In figure 3 (b) we additionally present a curve (τ_{LC}) of Larmor clock (LC) time, which was also obtained by fitting of the LC data given by Hofmann [14]. The agreement with LC (magenta curve) is better for small F values, which is the limit of thick barrier (more on this in the next section). This is because Hofmann's LC time-delay is given for the nonadiabatic field calibration and our τ_{dB} becomes closer to nonadiabatic time-delay τ_{dion} for small F (thick barrier) (see equation 9 below). This again explains the disagreement for larger field strength $F = 0.06 - 0.1$, compare figure 3 (b). The notation τ_{dB} below is used to refer to the barrier (tunneling) time-delay τ_{dB} ($\equiv \tau_{barrier} \equiv \tau_{dwell}$) with the corresponding adiabatic and nonadiabatic tunnel-ionization time-delay τ_{Ad}, τ_{dion}, respectively.

3. Weak measurement limit

In the previous section we found that the LC time τ_{LC} tends closer to the barrier time-delay τ_{dB}, $\tau_{barrier,exp}$ for small field strength, compare figure 3 (b). The LC-time is usually considered in the context of the so-called weak measurement (WM) approach, which characterizes a system before and after it interacts with a measurement apparatus [21, 22]. Landsman [23] showed that the WM value of the time-delay corresponds to LC-time $\tau_{weak} = \tau_{LC}$ [23, 24]. Thus, for small field strengths ($F \ll F_a$) the barrier time-delay τ_{dB} in our model corresponds to the WM approach, which is clearly seen in figure 3 (b). Indeed, for $F \ll F_a$ the barrier width $d_B = \delta_z/F$ becomes large enough which makes the barrier thick . At the limit of thick or opaque barrier, d_B approaches the so-called classical barrier width d_C, $\lim_{F \ll F_a} d_B \approx I_p/F = d_C$ ($\equiv x_{e,c}$), compare figure 1. In this limit the barrier height $\lim_{F \ll F_a} \delta_z = I_p$, and the barrier time

$$\lim_{F \ll F_a} \tau_{dB} = \tau_a \frac{F_a}{F} \frac{(\delta_z \approx I_p)}{I_p} \approx \tau_a \frac{F_a}{F} = \tau_{dion} \tag{9}$$

This result is interesting, as it shows that the barrier time-delay τ_{dB} for a thick (or opaque) barrier is approximately equal to tunnel-ionization time-delay τ_{dion} (equation (3)) of the nonadiabatic field calibration [4, 14]. This explains the already mentioned agreement between the τ_{LC} given by Hofmann (data-fit curve) and τ_{dB} for small field strengths. The agreement is satisfactory considering that the fitted LC-curve is extended beyond the data range given in [14]. This leads us to the interaction time with the laser field in the barrier area, since the LC-time according to Steinberg [24, 25] is related to the interaction time within the barrier, which corresponds to the time spent in the barrier or the dwell time τ_{dwell} [26, 27], which in turn equals τ_{dB}, $\tau_{barrier}$ according to the UTTP (and τ_{dwell} of Winful in equation (6)).

The agreement shown in figure 3(b) (see also equation (9)) suggests that the WM value (LC-time) and the interaction time within the barrier region in the thick barrier limit, can be determined by τ_{dB} ($\approx \tau_{dion}$ of the nonadiabatic field calibration as given in equation (9)). We think this is similar to the measurement presented in [26] (measurement of the time spent in the barrier with LC). In addition, the back reaction of the measurement of the system can be found from $\tau_{T,i}$ as the following

$$\begin{aligned} \tau_{backr}^{WM} &= \lim_{F \ll F_a} \tau_{T,i} = \lim_{F \ll F_a} \frac{1}{2} \frac{(I_p - \delta_z)}{4 Z_{eff} F} \\ &= \lim_{F \ll F_a} \frac{1}{2} \frac{\varepsilon_F}{4 Z_{eff} F} \approx \frac{1}{4 I_p} = \lim_{F \to 0} \tau_{T,i} \end{aligned}$$

where $\varepsilon_F = (I_p - \delta_z) \sim 2 Z_{eff} F / I_p$ is small under the WM condition (linear dependence on F), and we used the expansion of $(I_p^2 - 4 Z_{eff} F)^{1/2}$ for small F.

However, the condition of WM is not necessary and we can assume that $\tau_{T,i}$ always represents the back reaction of the system, which is generally consistent with the interpretation as the time needed to reach the barrier entrance in the strong-field interaction, see discussion after equation (2). Finally, we note that $\tau_{T,d}$, $\tau_{T,i}$ can be interpreted respectively, as forward and backward tunneling [1] and we can decompose $\tau_{T,i}$ similarly as was done for equation (5), to

$$\begin{aligned} \tau_{T,d} &= \tau_{dion} + \tau_{dB} \, (\text{forward tunneling}) \\ \tau_{T,i} &= \tau_{dion} - \tau_{dB} \, (\text{backward tunneling}), \end{aligned} \tag{10}$$

showing again that

$$\tau_{dB} = \tau_{dwell},$$

since, according to Steinberg [24], we have $\tau_{dwell} = Re(\tau_T) = Re(\tau_R)$, where τ_T, τ_R are the transmission and reflection scattering channel times, respectively. They are the forward and backward (tunneling) scattering channels in our model. Of course, τ_{dB} is real, as we explained in our previous work [4]. In our forward and backward channels of adiabatic tunneling in equation 2, which is illustrated in figure 2 (dashed-dotted, red curve), the condition of a spatially symmetric barrier noted by Steinberg [24] is reflected by the fact that the horizontal channel happens along the (same) barrier width $d_B = x_{e,+} - x_{e,-}$ for forward and backward tunneling, compare figure 2. Finally we can write τ_T, (d, i) $= \tau_{dion} \pm \tau_{dB}$ and $\tau_{dion} = (1/2)\ (\tau_{T,d} + \tau_{T,i})$, $\tau_{dB} = (1/2)(\tau_{T,d} - \tau_{T,i})$, where the symmetrization results in τ_{dion} and the anti-symmetrization results in the barrier time-delay or the dwell time. For further details and discussion, we would like to refer readers to our previous work [3]. Forward and backward tunneling is equivalent to the transmission and reflection of the wave packet in the traditional quantum tunneling studies, which usually use numerical methods [17, 18, 28–31], whereas our model offers a simple tunneling approach with expressions that agrees well the measurement result of the attoclock experiment.

Conclusion and outlook

The tunneling time has a universal behavior referred to as UTTP, where the barrier (tunneling) time-delay can be defined by a simple subtraction of adiabatic and nonadiabatic tunnel-ionization time-delay, $(\tau_{Ad} - \tau_{dion}) = \tau_{dB} = \tau_{barrier} = \tau_{dwell}$, see equations (8), (and (6) and (7)). It is shown that τ_{dB} agrees well with time-delay $\tau_{barrier,exp}$, which is obtained by fitting the experimental data (see figure 3) as the difference between the time-delays resulting from the adiabatic and nonadiabatic field calibrations of the measurement of the attoclock experiment, i.e. those of Landsman [9] and Hofmann [14] respectively. More remarkably is that our result provides conceivable definitions of the tunnel-ionization time-delay (adiabatic and nonadiabatic), the barrier (tunneling) time-delay and the interpretation of the attoclock measurement. We also found that the barrier time-delay τ_{dB} corresponds to the LC-time and the interaction time [24–27]. This is particularly evident in the limit of thick barrier, where τ_{dB} approaches the time-delay in the nonadiabatic field calibration τ_{dion}, and where the back reaction is small and the weak measurement approach is justified. We assume that there is a similarity to the measurement presented in [26]. In the future, we will focus on faster-than-light tunneling or quantum superluminal tunneling (QST), e.g. [28], one of the most exciting phenomena in quantum physics. Our preliminary result shows that superluminality can be experimentally investigated using the attoclock scheme and furthermore by the numerical integration of the Schrödinger and Dirac equations, on which we are currently working. Furthermore, it also shows that the condition on QST for the barrier (tunneling) time-delay τ_{dB} is less severe than expected ($Z \sim 18$ for H-like atoms) and that the relativistic effects are not crucial. We think that the present work and future investigation on QST serve as inspiration for further experimental and theoretical studies.

Acknowledgments

I would like to thank C Hofmann for sending the experimental data and FPI data shown in the figures. I would like to thank Prof. Martin Garcia from the Theoretical Physics of the Institute of Physics at the University of Kassel for his kind support.

Competing interests statement

The author declares that there are no relevant financial or non-financial competing interests to report.

Conflict of interest

The author declares no conflict of interest.

Data availability statement

No new data were created or analysed in this study.

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