Tunneling in attosecond optical ionization and a dynamical time operator

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(Received 8 May 2017; published 29 August 2017)

The conundrum parameter-operator of time in quantum mechanics, as well as the time-energy uncertainty relation and the tunneling delay time, have recently been addressed in attosecond optical ionization experiments. Dirac's formulation of the electron's relativistic quantum mechanics (RQM) allows the introduction of a dynamical self-adjoint time operator, representing an internal time of the system. Its relation to the parametric (laboratory) time is derived. Its relevance to the tunneling measurements in these experiments is exhibited, namely, as the time it takes the wave packet to cross the exit point of the barrier, thus justifying considering this time operator an additional observable in RQM.

DOI: 10.1103/PhysRevA.96.022139

I. INTRODUCTION

The conundrum *parameter-operator* of time in quantum mechanics (QM), as well as the *time-energy uncertainty relation* and the *tunneling delay time,* have recently been addressed again in the development of attosecond optical ionization experiments [1–3]. The tunneling phenomenon, one of the earliest theoretical successes of QM, has been extensively debated in relation to the question of the time the particle spends in the barrier region. This has given rise to alternative definitions of tunneling times but has not been definitively resolved [3–5]. On the other hand, the technical development of attosecond pulses of extreme ultraviolet radiation has allowed photoionization processes where a tunneling delay time can be measured and compared to theoretical predictions, although using a time-energy uncertainty relation associated with the commutation relation rightfully objected to by Pauli [4,6,7].

Indeed the existence of a time-energy uncertainty relation analog to the position-momentum one, conjectured by Heisenberg early on, faced from the start Pauli's objection to the existence of a time operator, to quote [8, p. 63]: "... from the C.R. written above (cf. $[t,H] = i\hbar$) it follows that H possesses continuously all eigenvalues from $-\infty$ to $+\infty$, whereas on the other hand, discrete eigenvalues of H can be present. We, therefore, conclude that the introduction of an operator t is basically forbidden and the time t must necessarily be considered as on ordinary number ('c' number) in Quantum Mechanics." In the time dependent Schrödinger equation (TDSE), time appears as a parameter, not an operator [8,9]. This led to a variety of alternative proposals for a time-energy uncertainty relation and an extensive discussion of time in quantum mechanics throughout several decades [10–14]. Pauli's argument, sustained also by the fact that the system's stability requires the energy to have a finite minimum, is still the subject of current research, as well as the existence and meaning of a time-energy uncertainty relation [15,16]. The undisputed experimental corroboration of Schrödinger's equation supports the interpretation of the parameter t as the laboratory time. Its presence in the dynamical evolution of microscopical systems (TDSE) has been attributed to the entanglement of these systems with a macroscopic classical environment [17].

Now, it has been shown that Dirac's formulation of the electron's relativistic quantum mechanics (RQM) does allow the introduction of a dynamical time operator that is self-adjoint [18]. Consequently, it can be considered an additional system observable representing an internal time, that in the Heisenberg picture depends on the time parameter of the Schrödinger equation. This time operator is the generator of continuous momentum displacements, and consequently continuous energy displacements within both the positive and the negative energy branches, but not across the energy gap. In this way Pauli's objection is circumvented [19]. In the present paper it is shown that the dynamical time operator provides an equal footing of time and space in the analysis of the attosecond optical ionization processes, as suggested in Ref. [6].[1] These aspects are examined within the standard framework of RQM. The definition and main properties of the proposed time operator are recalled in Sec. II. In particular the ensuing time-energy uncertainty relation is shown to agree with Bohr's interpretation of the time uncertainty as the uncertainty in the time of passage at a point of the trajectory. On the assumption that this time of passage is a tunneling internal time, Sec. III develops its application to the attosecond optical ionization processes, based on its derived relation to the electron external (laboratory) tunneling time. Section IV advances conclusions and possible developments.

II. THE DYNAMICAL TIME OPERATOR IN RQM

A dynamical self-adjoint "time operator"

$$\hat{T} = \alpha \cdot \hat{\mathbf{r}}/c + \beta\tau_0 \tag{1}$$

has been introduced [18] in analogy to the Dirac free particle Hamiltonian $\hat{H}_D = c\alpha \cdot \hat{\mathbf{p}} + \beta m_0 c^2$, where $\alpha_i (i = 1,2,3)$ and

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[1] In this respect, Dodonov's quoted paper in Ref. [6], that claims that no unambiguous and generally accepted results have been obtained so far, refers only to the present author's paper of 1983 [20], where the necessary conditions to define a time operator are discussed, but not to the 2014 paper [18] that introduces a specific dynamical time operator in RQM.

β are the 4×4 Dirac matrices, satisfying the anticommutation relations [9,21,22]

$$\alpha_i\alpha_j + \alpha_j\alpha_i = 2\delta_{ij} \quad \alpha_i\beta + \beta\alpha_i = 0 \quad \beta^2 = 1. \tag{2}$$

τ_0 represents in principle an internal property of the system, determined to be the de Broglie period h/m_0c^2 [19,23,24].

In the Heisenberg picture the time evolution of the time operator is given by

$$\begin{aligned}\hat{T}(t) &= \alpha(t)\cdot\hat{\mathbf{r}}(t)/c+\beta(t)\tau_0\\ &= \alpha(0)\cdot\hat{\mathbf{r}}(0)/c+\beta(0)\tau_0+[\alpha(0)/c]\cdot(c^2\hat{\mathbf{p}}/\hat{H}_D)t\\ &\quad+\text{oscillating terms}\\ &= \hat{T}(0)+\{1-\beta(0)/\gamma\}t+\text{ oscillating terms},\end{aligned} \tag{3}$$

where use has been made of

$$\begin{aligned}&\alpha(0)\cdot(c\hat{\mathbf{p}}/\hat{H}_D)\\ &\quad=\{\hat{H}_D-\beta(0)m_0c^2\}/\hat{H}_D=\{1-\beta(0)m_0c^2/\hat{H}_D\}\\ &\quad=\{1-\beta(0)m_0c^2/m_0c^2\gamma\}=\{1-\beta(0)/\gamma\}\end{aligned} \tag{4}$$

and the fact that $\langle\hat{H}_D\rangle = m_0c^2\gamma$ is a constant of motion. $\gamma = \{1-(v_{gp}/c)^2\}^{-1/2}$ is the Lorentz factor with v_{gp} the group velocity. Thus $\hat{T}(t)$ exhibits a linear dependence on t together with a superimposed oscillation (Zitterbewegung), as occurs with the time development of the position operator $\hat{\mathbf{r}}(t)$.

Leaving aside the oscillating terms, its expectation value for a free wave packet is given as

$$\begin{aligned}\langle\hat{T}(t)\rangle &= \langle\hat{T}(0)\rangle+\{1-\langle\beta(0)/\gamma\rangle\}t\\ &= \langle\hat{T}(0)\rangle+\{1-\langle\beta(0)\rangle\{1-(v_{gp}/c)^2\}^{1/2}t.\end{aligned} \tag{5}$$

For low ("nonrelativistic") positive energies $[\langle\beta(0)\rangle \simeq 1, \langle\hat{H}_D\rangle \simeq m_0c^2, 1/\gamma \simeq 1-\frac{1}{2}(v_{gp}/c)^2+\cdots]$ a time lapse is given as

$$\begin{aligned}\langle\hat{T}(t_2)\rangle-\langle\hat{T}(t_1)\rangle &\simeq \{1-\langle\beta(0)\rangle+\tfrac{1}{2}\langle\beta(0)\rangle(v_{gp}/c)^2\}(t_2-t_1)\\ &\simeq \tfrac{1}{2}(v_{gp}/c)^2(t_2-t_1) \ll (t_2-t_1).\end{aligned} \tag{6}$$

Thus, in this case, electron parametric (external) intervals are enhanced with respect to dynamical (internal) intervals, that by definition [Eq. (1)] are related to the time it would take light to travel the distance.

On the other hand, for high ("relativistic") energies $[\langle\hat{H}_D\rangle \simeq cp, v_{gp} \simeq c, (1/\gamma) \simeq 0]$, one obtains

$$\langle\hat{T}(t_2)\rangle-\langle\hat{T}(t_1)\rangle \simeq (t_2-t_1), \tag{7}$$

and electron parametric (external) intervals coincide with dynamical (internal) intervals.

It also follows [18,19] that the time operator and the Dirac Hamiltonian satisfy the commutation relation

$$[\hat{T},\hat{H}_D] = i\hbar\{1+2<\beta K>+2\beta(\tau_0\hat{H}_D-m_0c^2\hat{T})\}, \tag{8}$$

where $K = \beta(2\mathbf{s}\cdot\mathbf{l}/\hbar^2+1)$, a function of the spin orbit coupling, is a constant of motion [21]. The last term vanishes at the origin of the proper system where $T = \beta\tau_0$ and $\hat{H}_D = m_0c^2$.

From this follows an uncertainty relation

$$\begin{aligned}(\Delta\hat{T})(\Delta\hat{H}_D) &\geqslant (\hbar/2)|1+2<\beta K>|\\ &= (3\hbar/2)\big|1+\tfrac{4}{3}\langle\mathbf{s}\cdot\mathbf{l}/\hbar^2\rangle\big|.\end{aligned} \tag{9}$$

For central potentials where the eigenstates are such that $\langle\hat{\mathbf{r}}\rangle = 0$ and $\langle\hat{\mathbf{p}}\rangle = 0$, the uncertainty of this time operator is related to the uncertainty in position $\Delta\mathbf{r}$, namely:

$$\Delta T^2 = \langle T^2\rangle-\langle T\rangle^2 \approx \langle r^2\rangle/c^2+\tau_0^2\{1-\langle\beta\rangle^2\} \approx \langle r^2\rangle/c^2, \tag{10}$$

whereas in the same way the energy uncertainty is related to the momentum uncertainty $\Delta\mathbf{p}$. Thus,

$$\Delta T \approx \Delta\mathbf{r}/c \qquad \Delta\hat{H}_D \approx c\Delta\hat{\mathbf{p}} \tag{11}$$

and

$$(\Delta\hat{T})(\Delta\hat{H}_D) \approx (\Delta\hat{\mathbf{r}})(\Delta\mathbf{p}) \geqslant (3\hbar/2). \tag{12}$$

The association of $\Delta\hat{T}$ with $\Delta\hat{\mathbf{r}}$, and of $\Delta\hat{H}_D$ with $\Delta\hat{\mathbf{p}}$, corresponds to Bohr's interpretation [25], namely, the width of a wave packet in space, complementary to its momentum dispersion (and thus to its energy dispersion), measures the uncertainty in the time of passage at a point of the trajectory.

III. TUNNELING TIME IN ATTOSECOND OPTICAL IONIZATION

The laser pulse opens the electron bound state at energy $E_0 = -I_p$ to tunneling through a practically static barrier created by an effective potential in the direction of the pulse polarization. This is justified by the fact that the laser center wavelength in the infrared range is much slower (0.0456–0.065 a.u.) than the oscillating frequency of the electron in the atom [$O(1)$ a.u.]. This means that for a bound electron, the time-dependent field varies adiabatically [3]. The barrier is thus modeled as [3,4,7]

$$V_{\text{eff}} = -\frac{Z_{\text{eff}}e}{|\mathbf{r}|}-\mathbf{F}\cdot\mathbf{r}. \tag{13}$$

The first term is the binding Coulomb potential and the second is the dipole interaction with a pulse of maximum intensity F. The barrier width d_B in any radial direction, say (θ_p,φ_p), of the electric field polarization is given by the difference between the entrance $r_{e,-}$ and exit $r_{e,+}$ points of the barrier (Fig. 1 of Ref. [7]), i.e., the solutions to the equation

$$-\frac{Z_{\text{eff}}e}{r}-Fr = -I_p$$

yielding

$$d_B(F) \doteq \{r_{e,+}-r_{e,-}\} = (I_p/F)\sqrt{1-4Z_{\text{eff}}eF/I_p^2} \tag{14}$$

as given in Eq. (13) of Ref. [7].

At this point it is important to note that in the presence of potentials dependent only on position, e.g., Coulomb type potentials, one has

$$[\hat{T},\hat{H}_D+V(\hat{\mathbf{r}})] = [\hat{T},\hat{H}_D] \tag{15}$$

and the same internal time uncertainty relation, Eq. (9), follows.

Considering that the sudden onset of the pulse spreads the stationary ground eigenstate ($\langle\hat{\mathbf{r}}\rangle = 0$, $\langle\hat{\mathbf{p}}\rangle = 0$) into a continuum, it sets the particle wave packet in motion by modifying the momentum distribution component in the direction of polarization. The time of passage at a point

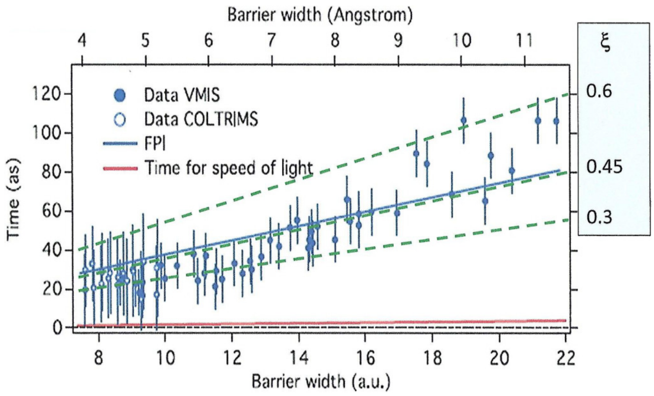

FIG. 1. Dependence on ξ of tunneling time vs experimental data (Fig. 3(d) of Ref. [2]).

of the barrier (say the exit point) acquires a finite internal time uncertainty related to the increasing position uncertainty, initially confined to the atomic size, but now occupying part of the much larger barrier width $d_B(F)$. As shown in Sec. II above, the minimum internal time uncertainty for spherical symmetry is

$$\Delta\hat{T} = \Delta\hat{\mathbf{r}}/c = \langle r^2\rangle^{1/2}/c, \tag{16}$$

where integration is carried over all directions. For a spherically symmetric state one can write $\Delta\hat{\mathbf{r}} = 4\pi\Delta r$ with the contribution in one radial direction being $\Delta r = \xi\{d_B(F)\}$, where the phenomenological parameter $\xi \leqslant 1$ allows one to consider that the wave packet, although spreading, occupies only a fraction of the barrier width. An estimation of ξ is given in the Appendix below.

The main assumption now is that this time of passage uncertainty at the exit point represents the time needed to cross this point and represents a tunneling internal time $\bar{\tau}_T$, namely:

$$\bar{\tau}_T = (1/4\pi)\Delta r/c \approx (1/4\pi)\xi\{d_B(F)\}/c. \tag{17}$$

However, from Eq. (6), the corresponding electron laboratory tunneling time Υ_T in the nonrelativistic regime is obtained through the enhancement from this internal time lapse as follows:

$$\begin{aligned}\Upsilon_T &\approx \bar{\tau}_T/\tfrac{1}{2}(cp/m_0c^2)^2\\ &\approx \{(1/4\pi)\xi d_B(F)/c\}/\tfrac{1}{2}(v_{gp}/c)^2 \gg \bar{\tau}_T.\end{aligned} \tag{18}$$

This approach is seen to yield a linear relation between laboratory electron tunneling time and barrier width, as has been experimentally obtained (Fig. 3(d) of Ref. [2]). Given a group velocity v_{gp}, the parameter ξ determines the slope of this linear dependence as well as the magnitude of the laboratory tunneling time. Reference [2] reports an electron tunneling time of 40 as for a barrier width of 13 a.u. = 6.88 Å. This gives a laboratory group velocity $v_{gp} = \frac{6.88}{40}$ Å/as, and a ratio $1/\frac{1}{2}(v_{gp}/c)^2 = 608.44$. It then follows

$$\Upsilon_T \approx (1/4\pi)608.44[\xi d_B(F)/c] \text{ as} = (16.14\xi)d_B \text{ (as)}, \tag{19}$$

where the barrier width is given in angstroms.

Figure 1 shows the dependence on ξ of the tunneling time vs barrier width for different values of ξ, together with the background of experimental data of Fig. 3(d) of Ref. [2], where the tunneling time of light—that by definition corresponds to the internal tunneling time—is also exhibited. Note that the value $\xi = 0.45$ practically reproduces the FPI (Feynman path integral) quantum mechanical result that is considered to realize a good fit to the experimental data [2].

The dependence on the field intensity is obtained using Eq. (14), namely:

$$\Upsilon_T \approx (1/4\pi)\xi\big[(I_p/F)\sqrt{1-4Z_{\text{eff}}eF/I_p^2}/c\big]\big/\big[\tfrac{1}{2}(v_{gp}/c)^2\big], \tag{20}$$

which gives the observed shape of the dependence of the barrier width on the field intensity (Fig. 3(b) of Ref. [2]).

IV. CONCLUSION

The dynamical time operator provides a plausible explanation within standard RQM of the tunneling times measured in the photoionization experiments. As an observable, it introduces an internal time, in addition to the parameter (laboratory) time in the TDSE, that has been shown to be an emergent property arising from the entanglement of a microscopic system with a classical environment in an overall closed time independent system, a property being apparent only to an internal observer [17,26]. There is no conundrum *parameter-operator* of time in quantum mechanics, as both times are seen to play a role in RQM and Pauli's objection is circumvented [19]. The tunneling internal time is assumed to be given by the dynamical time operator uncertainty considered, following Bohr, as a time of passage at, say, the exit point of the potential barrier. The derived enhancement at low energies of the tunneling internal time to exhibit the corresponding electron laboratory tunneling time yields the agreement with the measurements in attosecond optical ionization experiments. The point of view adopted certainly differs from other conceptions and definitions of tunneling time, some of which fail to agree with experimental data, as exhibited in Ref. [2].

Some aspects to be explored beyond this paper are the following. Based on the position observable, the time operator is seen to exhibit a Zitterbewegung behavior about its linear dependence on t. As occurs with the position one, its observation is beyond current technical possibilities. However it may be observable in systems that simulate Dirac's Hamiltonian, where a position Zitterbewegung has already been exhibited experimentally [27–29]. A corresponding time operator can be constructed in each case and perhaps its properties may be exhibited in similar experiments.

Finally, general relativity accords a dynamical behavior to space-time, firmly confirmed recently by the detection of gravitational waves. As a dynamical time is definitively incompatible with a time parameter, this becomes from the start a fundamental "problem of time" in quantum gravity [30,31]. Whether the time operator here introduced has a relevance in this problem is a venue to be considered [32].

ACKNOWLEDGMENTS

The author wishes to recognize an unknown reviewer for constructive comments and criticisms and the journal Optica for the permission to use Fig. 3(d) in Ref. [2].

APPENDIX: AN ESTIMATION OF ξ

The spreading of a free wave packet from an initial value Δq_0 is given as [33]

$$\Delta q(t) = \Delta q_0\left[1+\left(\frac{\Delta p_0}{m\Delta q_0}t\right)^2\right]^{1/2} = \Delta q_0\left[1+\left(\frac{\Delta H_0}{mc^2}\frac{ct}{\Delta q_0}\right)^2\right]^{1/2}, \tag{A1}$$

where, from Eq. (11), $\Delta H_0 \approx c\Delta p_0$ is considered as the energy spread produced by the sudden onset of the laser. For a spherically symmetric potential, this spread is $\Delta H_0 = F/\sqrt{2|E_0|}$, where E_0 is the ground state energy of the bound electron and F is the electric field strength [4]. Now Ref. [2] quotes in Fig. 3(d) a tunneling time of 40 as for a barrier width of 13 a.u. (6.88 Å) together with $F = (0.08/2\pi)$ a.u. $= 4.47\ \mathrm{W/cm^2}$ (see Fig. 2(b) of Ref. [2]). As the target is helium ($Z_{\rm eff} \approx 2$), one has for the ground state $\Delta q_0 \approx (\sqrt{3}/2)$ a.u., and $|E_0| = 24.59$ eV $= 0.904$ a.u. [1], to yield

$$\xi(t = 40\ \text{as}) \equiv \frac{\Delta q(t = 40\ \text{as})}{d_B} \approx 1.23\,\frac{\Delta q_0}{d_B} \approx 0.18. \tag{A2}$$

This value is of the order of the phenomenological adjustment in Fig. 1. It agrees with the assertion that ΔH_0 is small and spreads slowly, as quoted, and furthermore exhibited in Fig. 4(b) of Ref. [4]. The value will be increased if the effect of traveling through the barrier corresponds on average to free motion with an effective mass $m_{\rm eff} < m$.

Finally, from the Dirac equation [9,21,33], the laboratory time taken by a free wave packet to cover a distance $d_B = \langle r(t)\rangle - \langle r(0)\rangle$ is, neglecting the oscillating terms, given as $d_B/\langle c^2 p/H\rangle = d_B/v_{gp}$. Taking $v_{gp} = \sqrt{2|E_0|/m}$, one obtains

$$T = d_B/v_{gp} = (d_B/c)/(v_{gp}/c) = (d_B/c)/\sqrt{2|E_0|/mc^2} \approx 226\ \text{as} \tag{A3}$$

to compare with the 230 as quoted in Ref. [2].

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