TOPICAL REVIEW

Attoclock and the quest for tunnelling time in strong-field physics

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Abstract

The debate on tunnelling times have always been full of contradictions and the attoclock experiments that measure tunnelling delays in strong-field ionization are no exception. The current review presents the debate and discussions concerning the studies of tunnelling times based only on the attoclock technique. We review them with their implications and pitfalls identified due to lack of accurate strong field models that validate the observations in interpreting the measurements performed on noble gases. In order to provide a complete picture, the review begins with a background on some of the popular tunnelling time definitions, most of them conceived during the late 1980s debate, which are often cited in the attoclock literature. We then discuss various attoclock experiments on noble gas atoms and their interpretations in context of the tunneling time debate. The recently performed attoclock experiment and numerical modelling using atomic hydrogen are also presented as an attempt at resolving the controversy. We conclude with the current status of the debate.

1. Introduction

Quantum tunnelling is one of the key features of quantum mechanics. It is central to many chemical and biological processes [1], to electron transport in semiconductor diodes/ transistors [2, 3], molecular junctions [4], nano-structures [5, 6] etc Although quantum tunnelling is well understood and exploited in various applications, there is no consensus in the scientific community on ‘How long does it take for a particle to tunnel through the barrier?’ This question forms the very crux of the tunnelling time problem. Although the question is not a new one and it is very fundamental in its nature, it is still far from being settled, with the ongoing debate having generated a wide body of literature (for example please see [7–11] and references therein). The efforts to define, to understand, to measure and to interpret the value of tunnelling time continue.

Initially the discussion was limited to theory as the estimated tunnelling times were too short to experimentally measure. The emergence of semiconductor technology provided a practical platform to observe tunnelling times and revived the discussion in late 1980s leading to different perspectives and various definitions. As we will see in the next section, these debates of 1980-90s together with the experiments performed could not answer the question unambiguously keeping physicists in a limbo. It took another 20 years for the precision metrology using ultrafast laser technology to offer a new experimental approach in the form of attosecond angular streaking (AAS) [12]. The AAS, also known as the attoclock, is a precision measurement technique that enables one to access electron dynamics at few attoseconds time-scale using near-circular few-cycle laser pulses. The technique, conceived based on the Simpleman’s model [13–15], involves measuring the relative offset in the direction of photoelectrons1 most probable momentum in the polarization plane to the peak electric field direction of the ionizing pulse. The measured relative angular offset maps to time via the rotation period of electric field vector in an the ionizing pulse. As will be discussed later, the experimental offsets measured and interpreted in conjunction with theoretical models

1 Please note that, unless mentioned ‘photoelectrons’ in the subsequent discussion mean tunnelled electrons under strong-field ionization.
infer tunnelling delays. Therefore, it is of utmost importance to properly establish and verify validity of the models used for interpreting the results of the attoclock experiments.

The current review is focused on the discussion of tunnelling times (or tunnelling delays as they are called in the strong-field community) from the perspective of previous attoclock experiments [12, 16–21] and the intricacies of their interpretations [21–32]. It also presents the recent studies with atomic hydrogen ($H$) [33, 34] that were performed in an attempt to resolve the ongoing controversy on tunnelling delays. But before presenting and discussing those fairly recent results, we would like to give a brief overview of the tunnelling time definitions, which were often cited in the attoclock literature.

The article is divided into two parts. Part I reviews the tunnelling time debate and the various definitions that were introduced in order to assign a definite value to the ‘time spent by a particle under the barrier’. Of many definitions and ways that were used in understanding tunnelling dynamics, we are only presenting the most often cited throughout attoclock literature. Part II covers the revival of this debate after and in context of attoclock measurements. We argue that the crux of the debate lies in the failure of the theoretical models to accurately describe the ionization dynamics. As the only currently available theoretical model, for which validity, accuracy and precision are well established, is a numerical solution of the 3-dimensional Schrödinger equation (3D-TDSE) for one-electron systems, we also argue in favour of performing attoclock with $H$ as the only way to resolve the controversy. We then present the results of such a study. Finally, the current state of the debate is presented with concluding remarks.

Part I: Pre-attoclock debate on tunnelling times

2. Tunnelling time problem

Classically, a particle with total mechanical energy $E_n$ cannot overcome or cross a barrier $V$ if $V > E_n$. Nonetheless, in case of a quantum particle, there exists a finite probability for finding it on the other side of the barrier at finite barrier width. The difficulty in determining tunnelling time unambiguously is mostly due to two reasons: (i) Since the total mechanical energy (sum of kinetic and potential energy) of the particle is less than the potential barrier while travelling under it, the particle has a negative kinetic energy and an imaginary velocity under the barrier. Thus, this region is referred to as ‘classically forbidden’. (ii) The framework of quantum mechanics can calculate expectation values of a physical quantity represented by an operator. The expectation values are real for Hermitian operators, and they can be measured in the laboratory. However, time in quantum mechanics appears as a parameter rather than as an operator. Further, it has been argued that there exists no form of the time operator that can be both self-adjoint and also be conjugate with Hamiltonian $H$ [35–37]. Nevertheless, attempts have been made to assign a definite value to the ‘time spent by a particle under the barrier’.

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2.1. Phase times

The initial notable works on tunnelling times were done by Eisenbud [45], Wigner [46] and Bohm [47]. They considered the tunnelling models that were closer to the classical picture of waves getting scattered on a potential. A phase shift is acquired when the incident waves get scattered on a potential and the relative phase shifts $\partial \theta / \partial E_n$ were related to ‘time-delays’ by their energy derivative $\partial E_n / \partial \theta$. Hence these are known as ‘phase times’. Since the models considered only the stationary (time-independent) wavefunctions, the methods were called stationary-phase methods. This was further generalized and a comprehensive overview of these time-delays can be found in [48]. Later, Smith [49] introduced the collision lifetime that characterized the duration of a collision process in three dimensions. The collision lifetime is based on the idea that

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time as defined in (1), thereby shedding new light on the physical meaning of phase time. Hence the phase
time is often called 'Eisenbud-Wigner-Smith time' \( \tau_{\text{WES}} \) (Wigner-Smith time or Bohm-Wigner time),
especially in the recent literature concerning the attoclock. Incidentally, the above idea on which Smith’s
collision lifetime is based is essentially the same as the one on which a tunnelling time known as the dwell
time is based. Hence Smith’s collision lifetime is often regarded as the origin of the dwell time.

In all the above mentioned works the projectile is considered as a wave-packet, the peak of the
wave-packet is well-defined i.e. the wave-packet is localized sharply around a given wavenumber \( k \) such that
momentum \( p = \hbar k \) and the complex transmission amplitude is given by \( T(k) = |T(k)|e^{i\theta} \). If we consider a
rectangular opaque barrier of width \( a \), this would yield a transmission group delay as,

\[
\tau_{\text{WES}} \equiv \hbar \frac{d\theta}{dE_n} \approx \frac{2}{\nu \kappa}.
\]  

(1)

where \( \nu = \hbar k/m \) is the velocity and \( 1/\kappa = \hbar / \sqrt{2m(V-E_n)} \) is the penetration depth. There were strong
objections to this [50–52] since the wave-packet could get distorted considerably after its transmission
through the barrier, which was demonstrated with a simple example in [53]. Alternatively, methods
employing the centroid or centre of gravity of the wavepacket were formulated to calculate time delays
[54, 55]. In addition to the wavepacket distortion, the delay in (1) is independent of the barrier-width when
it becomes sufficiently large and therefore in such cases, it can lead to apparent superluminal speeds, namely
the Hartman effect [56]. This occurs due to the difference in the transmission coefficient of the faster
components of the wavepackets versus its slower counterparts. It can also at times lead to negative group
velocities, that seem to apparently violate causality [57, 58].

2.2. Tunnelling times using clocks

In one of his most influential papers on tunnelling time [50], Büttiker differentiates the subtlety in asking the
question ‘How long does a particle spend in the classically forbidden region?’ from that of ‘How long does it
take a particle to tunnel through the barrier?’ He states that:

‘... An approach given by Smith, and advanced by others yields a time \( \tau \), as the ratio of the number
of particles under the barrier to incident flux... This method does not distinguish between particles
which at the end of their stay in the forbidden region have been reflected, and those that were
transmitted. This time is the average dwell time of a particle in the barrier, and is not the traversal
time, if most particles are reflected...’

This formed the motivation for a second class of tunnelling times defined as traversal time. An extra
degree of freedom associated with the barrier is used as a clock to measure the amount of time the particle
interacted with the barrier. This variety of times has become popular after a series of publications by Büttiker
and later by Landauer as a part of the 1980’s tunnelling time debate [50–52, 59, 60].

2.2.1. Büttiker-Landauer time \( \tau_{BL} \)

Here the particles with energy \( E_n \) and mass \( m \) are incident upon a rectangular barrier of width \( d \) and height
\( V_0 \) with a small oscillation in its height, i.e. \( V(x, t) = V_0(x) + V_1(x) \cos(\omega t) \) (see figure 1). Considering the
frequency of the oscillation to be lower than the so called 'interaction time of the particle', it tunnels elastically
through the static barrier. At high frequencies the particle sees a time-varying potential and tunnels through it
inelastically either by absorbing or giving up a quanta of energy \( \hbar \omega \). The frequency of this oscillation \( \omega \) is
tuned over a wide range such that a crossover is achieved between the two behaviours. When the frequency
resonates with inverse of the interaction time we have \( \omega \tau_{BL} = 1 \). They have shown that the traversal time i.e.
the interaction time of the particle of mass \( m \) with the barrier (for \( E_n < V_0 \)) is given by \( \tau_{BL} = md/\hbar \kappa \)
for rectangular barriers, and by (2) for barriers of a more general shape that allows WKB approximation.

\[
\tau_{BL} = \int_{-d/2}^{d/2} \sqrt{\frac{m/2}{V_0(x) - E_n}} \, dx.
\]  

(2)

Experimentally, the traversal time can be measured by observing the crossover frequency in the energy
spectrum of the transmitted particles. At low frequencies the transmission energy side-bands (considering
only to the first order) of \( E_n \pm \hbar \omega \) appear equal in their magnitude. But at higher frequencies, they are
asymmetric due to the enhanced transmission probabilities for the higher energy side band (that gain a
quanta) than the lower ones (that lose a quanta). If \( T \) is the transmission probability of the static barrier, the
transmission probabilities of the side-bands \( T_{\pm} \) are,

\[
T_{\pm} = (V_1/2\hbar \omega)^2 (e^{\pm \omega \tau_{BL}} - 1)^2 \, T.
\]  

(3)
These are then used to yield a simple relation for the interaction time as,

\[
\left( \frac{T_+ - T_-}{T_+ + T_-} \right) = \tanh(\omega \tau_{BL}).
\]

(4)

Considering the special case of opaque barriers, the WKB approximation allows the formulation to extend beyond rectangular barriers and a general formula [60] was derived subsequently as

\[
\tau_{BL} = -\hbar \frac{\partial \ln |T|}{\partial V}.
\]

(5)

A good overview on the experimental attempts to determine the above discussed traversal times can be found in [62]. Several studies proposed and reported the observation of crossovers as suggested by [50] [63–67]. In [63, 64], macroscopic tunnelling rate was measured using Josephson junctions. Josephson junction is a quantum mechanical device where a resistor is sandwiched between two superconductors. The resistor acts like a barrier for the cooper pairs of electrons to tunnel through it (with propagation velocity v), creating a current that is carried along a closed path of length l, known as the transmission line. Considering the traversal time is longer than the delay through the transmission line 2l/v, the tunnelled waves can interfere among themselves back and forth, decreasing the overall tunnelling rate. On the other hand, if the traversal time is much smaller than the delay introduced due to increased length, they can no longer interfere. A crossover was indeed observed as predicted by the time modulated barrier model [50] while changing the length from which traversal times were calculated. An interesting set of experiments was reported in [67] where quantum heterostructures were used to fabricate rectangular barriers of various thickness and heights that changed dynamically as the electron traversed through them. They also revealed a crossover as predicted in the case of time-modulated barrier systems.

The tunnelling-current with applied voltage i.e. I-V characteristic features of a scanning tunnelling microscope (STM) were measured to calculate traversal times [65, 66] while a polarized beam is incident on its junction. When a linear polarized light is focused in a way that the electric field vector is aligned along the tip of the axis, a bias is formed inducing a current back and forth following the laser field. This occurs even with no external voltage being applied across it. Given the rectification property of the STM, it should generate a DC current if the laser frequency is small compared to the transit time of electrons and vice versa. They observed the electron transit time to be ~ 1.8 fsec that agrees qualitatively with the \( \tau_{BL} \). However, interpretation of such results using 1D models is not so straightforward, due to a complicated geometry involved in the STM set-up. This, in addition to other critical comments was later pointed out in [68] (see pp.146-147).

2.2.2. Larmor time

An alternative clock invoking an interaction between the spin of the particle and the barrier confined by a magnetic field was proposed by Baz’[69, 70] and Rybachenko [71, 72]. They considered spin-1/2 particles of energy \( E \) that are spin polarized in the Y direction (\( \langle S_y \rangle = \langle S_z \rangle = 0, \langle S_x \rangle = \hbar/2 \)) which were then influenced by the magnetic field \( \vec{B} = B\Theta(z)\Theta(d - z)\hat{x} \) confined within the barrier of width, \( d \) and height, \( V \). By comparing the spin orientation of transmitted particles due to Larmor precession relative to the incident particles, the tunnelling time could be computed. This is termed as Larmor time in the literature and is expressed as the (barrier) potential derivative of the phase acquired by the transmitted particles,

\[
\tau_{LM} = -\hbar \frac{\partial \theta}{\partial V}.
\]

(6)
Figure 2. An illustration of Larmor clock wherein a spin-1/2 particle (spin polarized in Y direction) of energy $E_n$ is incident on a rectangular barrier of width $d$ and height $V$. $\tau_{LM}$ is calculated by the Larmor precession angle $\theta$ undergone by the transmitted particle. Alternatively, the barrier height is modulated by Zeeman splitting $\pm \hbar \omega_L/2$ ($\omega_L$ is the Larmor frequency) under the influence of magnetic field $\hat{B}$ confined within the barrier. The side-bands formed in the energy spectrum enables one to calculate the traversal time $\tau_B = \sqrt{\tau_{BL}^2 + \tau_{LM}^2}$. The figure is adopted from [61].

Figure 3. (a) Experimental set-up of an optical Larmor clock is illustrated with light polarization is measured as it passes through two prisms (of refractive index $n_H$) arranged as shown with a birefringent liquid crystal (of refractive index $n_L$, with $n_H > n_L$) in between. (b) The set up from top view with a ray diagram is shown in comparison to the traditional tunnelling scenario. The figure is adopted from [75].

By considering only the $S_y$ components, Rybachenko found $\tau_{LM}$ to be independent of the barrier width. Later Büttiker [59] realized that the Zeeman splitting of the $S_x$ component could lead to $\langle S_x \rangle = \pm \hbar/2$, making the effective barrier height $\mp \hbar \omega_L/2$, where $\omega_L$ is the Larmor frequency. This could lead to a preferential tunnelling for particles with $\langle S_x \rangle = +\hbar/2$. This allowed him to reinterpret the problem using the approach of modulated barrier case and derived three characteristic times that explain the particle’s interaction with the barrier: dwell time, traversal time and reflection time. It must be noted that the dwell time here must not be confused with the average dwell time that Smith introduced. Usually, dwell time in the literature of tunnelling times refers to the definition given by Büttiker. Dwell time measures the average time that the particle interacts with the barrier irrespective of its final state and was defined as,

$$\tau_d = 1/v \int_0^d |\psi(x)|^2 dz,$$

(7)

with $v$ being the velocity of the incident particle and $\psi(x)$ being the stationary wavefunction inside the forbidden region $0 < x < d$. Traversal/reflection times ($\tau_T/\tau_R$) were defined as the transmitted/reflected particle’s interaction time with the barrier and were expressed using the transmittance/reflectance coefficients. Now, by considering the parallel component of spin ($S_x$), the traversal time was shown to vary linearly according to the barrier width, unlike what Ryabachenko calculated. This traversal time is sometimes also called Büttiker time [73] $\tau_B$ and should not be confused with the Büttiker-Landauer time $\tau_{BL}$. In fact for the rectangular barriers the traversal time was shown to be $\tau_B = \sqrt{\tau_{BL}^2 + \tau_{LM}^2}$ [74]. In the special case of opaque barriers the phase becomes insensitive to the barrier, therefore yielding $\tau_B = \tau_{BL}$ as in (5).

Although experimentally realizing Larmor clocks as suggested above is challenging, there were experimental studies done measuring the effect of weak transverse magnetic fields on the traversal times using heterostructures [76, 77]. Interestingly, an optical equivalent of electron is used to measure Larmor times [75]. The electron spin is replaced with the photon polarization and the uniform magnetic field by a birefringent liquid crystal. The Stokes parameter vector on the Poincaré sphere is measured to determine the precession caused by the anisotropy of the liquid crystal’s refractive index. The optical equivalent of tunnelling scenario is shown in figure 3. The light is incident on the first prism of refractive index $n_H$ with an
angle beyond the critical angle. The evanescent field is frustrated total internal reflected (FTIR) [78] through the liquid crystal of relatively lower refractive index \( n_0 \). The phenomenon of FTIR is optically equivalent to quantum tunnelling. By varying the wavelength of the light they could control the effective barrier thickness. However due to a strong anisotropy in the refractive index, the results could not determine traversal times unambiguously.

It is noteworthy to mention at this point, that the words ‘time-delays’, ‘dwell time’, ‘traversal time’, ‘Büttiker-Landauer time’ are not strictly adhered to their original definitions in all consequent literature. They were redefined/reused/generalized in many different ways in the course of history and hence should be carefully contextualized when read or used. For example Büttiker [50] called \( \tau_{BM} \) the traversal time but in [60] \( \tau_B = \sqrt{\tau_{BM}^2 + \tau_{LM}^2} \) was called the traversal time, which was later called Büttiker-Landauer time by others [9]. As stated before, for the special case of opaque rectangular barriers \( \tau_{LM} \approx 0 \) making \( \tau_B = \tau_{BL} \). Although in some recent literature, for some historical reasons, \( \tau_{BM} \) as in (5) was called the Büttiker-Landauer time, there exists an agreement among the tunnelling physics community that \( \tau_B \) be defined as the Büttiker-Landauer time in its broadest sense. Similarly the concept of time-delay was also understood and used in different contexts [45, 54, 79]. We have already seen two different ways of defining ‘dwell time’ and we are yet to introduce its usage from a classification point of view in the next section. Also ‘sojourn time’ is often used synonymously in the literature in place of dwell time.

2.3. Complex times

2.3.1. Pollak-Miller time

Extrapolating the classical problems to the realm of quantum mechanics often can lead to complex quantities. This is also true while considering classical collisional theories in quantum mechanical context. Pollak and Miller [80] took such an approach of considering the collisional time to be a time average of a flux-flux correlation function. The idea was partly motivated by the success of the kinetic definition by Smith [49] being equivalent to the phase time-delays. If tunnelling delays are regarded as the excess time taken by the wavepacket colliding with a non-zero potential, Smith showed that it is equivalent to an average time spent by the incoming flux of particles in the interaction region. Hence from a kinetic point of view, considering both the incoming flux of particles and particles in the interaction region, the delays would correspond to the ratio of these two numbers. This had led to the definition of Smith’s dwell time. Following this prescription, Pollak and Miller arrived at complex traversal times and they called it the quantal imaginary time [80] in the context of scattering theory. Later in [81] it was argued that the real part of their complex traversal time gives the Smith time, while the imaginary part of it gives the true interaction time in the tunnelling picture. This true interaction time is known as the Pollack-Miller time,

\[
\tau_{PM} = \hbar \frac{\partial \ln |T|}{\partial E_n}. \tag{8}
\]

2.4. Feynman path-integral technique and tunnelling times

In 1948 [82], Feynman gave a new and yet an equivalent description for the quantum mechanical formulation of calculating amplitudes and probabilities by generalizing the action principle. In his formulation, amplitudes are calculated by assigning a sum of infinite classical trajectories that a quantum particle can possibly take in space and time. This description was initially put to use in the tunnelling context by Sokolovski and Baskin [83]. They provided general expression for traversal times and constructed complex time parameters that can relate to the already existing traversal times mentioned above in the form of \( \tau_{LM} - i\tau_{BL} \). The complex time relating as \( \tau_{WES} - i\tau_{PM} \) was already shown by [74] in their attempt to extend Büttiker-Landauer times to arbitrary barriers. These two complex traversal times in the combinations as mentioned above were defined in a unified manner using Gell–Mann-Hartle decoherence functionals [84] with the path integral approach [85].

To briefly summarize the results of [85], the quantity of interest was the sum of all those times during which a Feynman path \( x(t) \) was inside the barrier as shown in the left of figure 4. Alternatively, one can also adhere to a definition of the time difference between the first instant of time a Feynman path entered the barrier to the last instant when it left the barrier (right side of figure 4). These different descriptions led to two different probability distributions \( f(\tau) \). Calculating the quasi average of these two different distributions led to two classifications of the traversal times namely dwell time/resident time, \( \tau_d \) and passage time, \( \tau_p \), respectively. Incidentally, these were related to the well known tunnelling times as:

\[
\tau_d = \tau_{LM} - i\tau_{BL}, \quad \tau_p = \tau_{WES} - i\tau_{PM} - \hbar/2E_n. \tag{9}
\]
Considering the transmission coefficient to be $T = |T|e^{i\theta}$, the tunnelling times $\tau_{BL}, \tau_{LM}$ derived using the potential derivatives of transmission coefficient ($\partial \ln T/\partial V$) were understood as the dwell time (see (5) and (6)) and the energy derivative of transmission coefficient ($\partial \ln T/\partial E$) were understood to be the passage time (see (1) and (8)). These classifications are entirely at the level of Feynman paths and can be physically interpreted as the time measured by barrier oscillations (for dwell times) and timing the traversing particle (for passage times)\[85\].

3. Summary of the pre-attoclock tunnelling time debate

A brief overview of tunnelling times and its history is presented, with some relevant experimental works related to them. It is clear from the above discussed literature, that the definition and interpretation of tunnelling time is not unique. The usage of various names itself poses an initial confusion for the reader to understand tunnelling times. The path integral approach had helped to some extent in generalizing tunnelling times by deriving the well known quantities not only in a unified manner but also devoid of any specific physical model.

In spite of various experiments performed in an attempt to validate different models of the tunnelling times, it is still a matter of debate among the physics community. For example, soon after the optical Larmor clock experiment [75] as mentioned above in section 2.2.2, Balcou and Deutriaux [86] successfully performed a similar experiment using FTIR. In their attempt to measure the traversal times, they ended up measuring two traversal times simultaneously—phase time-delays and the Larmor time. Interestingly, both of these observations agree with their respective theories, but which one to be called ‘the real tunnelling time’ is an open question. On the same note, though crossovers were observed in the experiments [63–67] discussed in section 2.2, no definitive conclusions were drawn in invalidating other available tunnelling theories over Buttiker-Landauer time [11] (see p. 223), [60] (see p. 311) and [73] (see p. 227). At this point, it is worthwhile to end the discussion on tunnelling time theories with what Landauer and Martin stated in [11] which would summarize the ongoing debate regarding tunnelling times:

‘... "Do we need to bother about traversal time; is it an indispensable concept"? Of course it is not. Physics is full of alternative viewpoints... Traversal time, like the uncertainty principle, is a useful conceptual tool. It does not permit us to find answers, unavailable in other ways. The concept can be avoided if it is not to your taste. There is no copyright on the expressions traversal time and tunnelling time; each author can choose an interpretation... We can only ask if this is a fruitful view, and we can ask if it is relatable to experiment... Attitudes toward traversal time are, as in any area of uncertainty, often tied to a particular background...’

Part II: Revival of the tunnelling time debate in context of attoclock

Technological and scientific advancements go hand-in-hand and latter are often triggered by technological breakthroughs. Ultrafast laser technology is one such great example that enabled us to probe light–matter interaction on time-scales and regimes that were not accessible before. Modern ultrafast lasers can achieve field-strengths that are close or even greater than the strength of a typical electric field experienced by electron in an atom. The new technology has led to discovery of new phenomena such as above-threshold ionization\[87\], HHG \[88, 89\], and also to development of attosecond metrology \[90\], generation of

\[\text{In } [85], \text{ the } \tau_{\text{BL}} = -\hbar \partial \ln |T|/\partial V \text{ was called a tunnelling time of resident (or dwell) time type, in the sense of being obtained from the segments of Feynman paths that lie only inside the barrier region. This does not mean that the Buttiker-Landauer time is the same as the dwell time } \tau_d \text{ which Buttiker introduced in [59] as seen in (7).}\]
attosecond pulses [91, 92], coherent control of molecular dynamics [93–96], diffraction imaging and time-resolved electron holography [97, 98], frustrated tunnel ionization [99], etc.

Quantum tunnelling through the suppressed Coulomb barrier is at the heart of all the above mentioned processes, thus providing an accessible arena for performing controlled experiments and studying tunnelling dynamics. With this motivation, the attoclock was designed and was demonstrated to be a promising tool for measuring tunnelling times. However, in addition to the difficulty in choosing the right definition of tunnelling time from the literature available, the inability to model the complex strong-field-atom interaction created the ambiguity in correlating the measured observable (angular offsets) to tunnelling time. In the subsequent sections, we present the semiclassical model of tunnel ionization and tunnelling times in the context of strong-field ionization. We then describe the attoclock technique based on the semi-classical model with a brief overview of the attoclock experiments and the ongoing debate regarding the interpretation of the attoclock observable. We finally present the recent experimental results with $H$ and the conclusions as they stand today.

4. Simpleman’s model and tunnelling times

It’s been more than half a century since the theory of tunnel ionization was proposed by Leonid Keldysh [100, 101]. He parametrized the characteristic momentum of electron in its bound state ($\sqrt{2p_f}$) to the maximum momentum transferred from the external field ($E/\omega_0$) by the Keldysh parameter

$$\gamma \equiv \sqrt{\frac{2p_f}{E/\omega_0}} \text{ which can be rewritten as } \frac{I_p}{2U_p}, \text{ where } U_p = \frac{E^2}{4\omega_0^2}. \tag{10}$$

Here $I_p$ is the ionization potential, $U_p$ is the ponderomotive energy, i.e. the cycle averaged kinetic energy of an electron gained in the laser field, $E$ is the peak field strength of the light pulse and $\omega_0$ is the angular frequency of the laser radiation. When the field is strong enough to deform the Coulomb potential and creates a barrier, the electron can penetrate through with a considerable probability. Typically this happens when the laser field strength is 10–30 times less than that of the Coulomb field at the respective electron orbit. At present, it is difficult to fully describe and comprehend this process of tunnel ionization due to the complex dynamics involved with its accurate modeling requiring one to solve the three dimensional time-dependent Schrödinger equation (3D-TDSE) with all the interactions included. Nevertheless, a semiclassical model that is also known as ‘Simpleman’s model’ [13–15] could provide us great insights towards understanding the strong-field phenomena. In this model, the tunnelling is considered as the first step, and the electron that appears at the tunnel exit is considered to be a free particle interacting with the field. In some instances, this step is followed by a third step where the electron is accelerated back towards the parent ion and can rescatter. During these last two steps, the external field strength is very strong and hence the residual ion-electron interaction is often neglected. This is known as strong-field approximation (SFA) [102, 103].

The first step of tunnel ionization would obviously make one ask the pertinent question about the tunnelling time. But the scenario here is different from all the tunnelling models mentioned in Part I, as the electron is not a free particle initially. Here, the quantum particle is tunnelling from a bound state into a continuum state. Nonetheless, such problems were also studied using the Wigner-Eisenbud-Smith delays in the context of photo-ionization by attosecond pulses [104–107]. A relative phase shift is acquired by the outgoing electron wavepacket, which interacts with the atomic potential for a very brief time (on attosecond timescales). These photoionization time delays are well modelled using phase times as discussed in section 2.1 and a very well discussed in [108] and references therein. In the case of single-photon ionization, the outgoing wavepacket has a well-defined peak, while the tunnelling wavepacket does not. That makes it problematic to use phase times for calculating tunnelling delays. Studies were reported in the context of tunnel ionization [109] deriving tunnelling times using Feynman approach (as was discussed in section 2.4 the different tunnelling times can be derived using Feynman path-integrals) and will be discussed later in section 6. Before that, we shall restrict ourselves to the discussion of a semi-classical approach that plays an important role in providing us with an intuitive description of tunnelling.

Semi-classically, the Coulomb potential is distorted by the external field $E$ creating an effective potential barrier $V_{eff} = -Z_{eff}/x - Ex$ as shown with a red thick line in figure 5. $Z_{eff}$ here is the effective nuclear charge that the active electron sees under the SAE approximation, considering the screening potential. In the current review where the discussion involves ionisation of neutral atoms, the asymptotic behaviour of it would go to $Z_{eff} = 1$ but otherwise it would vary as a function of distance [110–114] and we dropped $x$ in $Z_{eff}(x)$ for convenience. During tunnelling, the electron traverses through the barrier of length $d$ with an average velocity of $v_{avg}$. Hence the time a classical electron takes to tunnel through the barrier can be expressed as $\tau_{ cls} = d/v_{avg}$, known as the Keldysh time. $d$ is known as the classical tunnel exit point and can be derived by
using the strength of external field $E$ and the ionization potential $I_p$: $d = I_p/E$. Considering the initial energy of the electron to be $1/2\gamma^2 = -I_p$, the electron uses all its kinetic energy in overcoming the barrier leading to a final velocity $\gamma$ at the tunnel exit being zero, hence $v_{\text{avg}} \approx \sqrt{\frac{\hbar I_p}{E}}$. Consequently, the velocity of the electron traversing under the barrier is imaginary (and so is the time). By using the above expressions and (10) the Keldysh time is:

$$\tau_{\text{Keldysh}} = \frac{d}{v_{\text{avg}}} = \frac{i\sqrt{2I_p}}{E} = i\gamma/\omega_0.$$  

Therefore $\gamma$, which was originally known as the adiabaticity parameter, can now be thought as the ratio of tunnelling time to that of the laser period. In the case of $\gamma \ll 1$, the time period of each laser cycle is much larger than the tunnelling time. Therefore the atom follows the external field adiabatically with electron tunnelling through an oscillating barrier, as in the case of the Büttiker-Landauer tunnelling time, $\tau_{BL}$. Hence, the Keldysh time $\tau_{Keldysh}$ is a close counterpart of the $\tau_{BL}$ in the strong field regime. Interestingly, using the conventional definition of momentum ($\sqrt{\frac{2m(E_n - V)}{\hbar}}$) in (2), we see that $\tau_{BL}$ is also imaginary. However Büttiker-Landauer argued that the tunnelling time cannot be imaginary and hence suggested to consider the imaginary part of momentum or $\tau_{BL}$ for the real traversal time. It is worth to point out that the Keldysh time and Keldysh parameter prominently figure in a commonly used definition of 'tunnelling regime' of ionization - the famous $\gamma \ll 1$ criterion for adiabatic tunnelling. Since Keldysh parameter compares Keldysh time with field oscillation period, an implicit assumption that Keldysh time represents time taken by electron to tunnel out makes this criterion equivalent to a condition that tunnelling occurs through a static barrier. As a number of more recent studies argued for relaxing the tunneling regime criterion to $\gamma \lesssim 1$ or even further, it appears to indicate that Keldysh time may substantially exceed the actual tunnelling time. In section 7 we will discuss the recent attoclock experiments on atomic hydrogen which are well aligned with this conclusion.

Apart from the above said formulations, tunnelling times in strong field processes were also calculated [115] using the time-energy uncertainty relations (TEUR) as prescribed by Mandelstam-Tamm [116]. They were shown to be numerically equal to the value of Keldysh times [117, 118]. Adapting the ideas of TEUR and considering the process of tunnelling to be similar to a Bohr-Einstein weighing photon-box [119, 120], a symmetric tunnelling time $\tau_{\text{sym}}$ was derived [113]. Unlike the semi-classical case of considering the barrier length to be $d$, the author considers $d_{\pm} (d_{\text{eff}})$ as the barrier entrance (exit) point (see figure (5)). These can be calculated by simply solving for $x$ from $V_{\text{eff}}(x) = -I_p$ [113]:

$$d_{\pm} = \frac{I_p \pm \sqrt{I_p^2 - 4Z_{\text{eff}}E}}{2E}.$$  

This is a gedanken experiment where a photon placed in a box with a shutter, is allowed to escape as the shutter opens for a brief interval of time $\Delta t$. Bohr showed [119] that measuring the relative change in the mass and there by energy (since $E_{\text{c}} = mc^2$) of the box within an error of $\Delta E_{\text{c}}$ leads to the TEUR such that $\Delta t / \Delta E_{\text{c}} \geq 1/2$. $\Delta t$ can then be interpreted as the passage time for the photon to escape. An alternate explanation was given by Busch [120], where by measuring the relative energy of the photon (before and after the shutter is open) with in $\Delta E_{\text{c}}$ error could be used to infer $\Delta t$ using TEUR.
Here the $\tau_{\text{sym}}$ is defined as the sum of (i) the time it requires for the electron to interact with the external field to enter the barrier at the point $d$ and (ii) the time it takes to travel across the barrier and reach point $d$, from which it escapes to the continuum. This was shown to be inversely proportional to the field strength and is given by:

$$
\tau_{\text{sym}} = \frac{I_p}{4Z_{\text{eff}}E}.
$$

(13)

It is important to note that both $\tau_{\text{kin}}$ in (11) and $\tau_{\text{sym}}$ in (13) are inversely related to the field strength. This is due to the reduction in the area of the barrier as external field grows stronger. We have already calculated the barrier width to be $d = I_p/E$. The height of the barrier $h$ can be calculated by finding the maxima of the potential curve $\max(V_{\text{eff}}(x)) = V_{\text{eff}}(x_m)$ (see figure 5), where $x_m$ is the extremal point such that,

$$
\frac{\partial V_{\text{eff}}}{\partial x} \bigg|_{x=x_m} = 0 \quad \text{and} \quad \frac{\partial^2 V_{\text{eff}}}{\partial x^2} \bigg|_{x=x_m} < 0,
$$

(14)

$$
\Rightarrow x_m = \sqrt{\frac{Z_{\text{eff}}}{\tau}}.
$$

(15)

$$
h = I_p - V_{\text{eff}}(x_m) = I_p - 2\sqrt{Z_{\text{eff}}E}.
$$

(16)

It shall now be clear that the barrier height $h$ and the width $d$ both decrease together with the increasing field. Hence by considering an electron as a particle traversing a barrier, one shall expect to see a trend of decreasing tunnelling times as a function of increasing field strength.

All the above formulations of tunnelling times were made purely from a theoretical perspective and employed a semi-classical model for a static external strong field. But in reality, when strong ultrashort laser pulses are used, the external field is continuously changing. Hence it is important to devise an experiment, by which one can address these tunnelling times. One such experiment is the attosecond angular streaking technique, proposed by Keller and co-workers ten years ago [12]. We discuss this technique and how it revived the tunnelling time debate in the following sections.

5. Attosecond angular streaking

According to the simpleman’s model and considering the quasi-static ionization rates [110, 121–125], one can see that the ionization rate curve reaches its maximum with the peak electric field. Nonetheless, a finite tunnelling time would imply a delay in the release of a bound electron into the continuum, which is when the ionization event is said to be completed. Hence, a finite tunnelling time would imply a finite time delay between the maximum of ionization rate and the peak field of the ionizing pulse. Unfortunately, the expected tunnelling times are on the order of few tens to hundreds of attoseconds and also the time-dependent ionization rate is not directly accessible to experimentalists. Therefore, it is necessary to design a clock that can measure these tunnelling times precisely within tens of attoseconds timescales. The technique of AAS or ‘attoclock’, that was conceived based on the semi-classical model [22] offers precisely this. Tunnelling delays are defined and measured by using the time difference between the peak of the ionizing field and the instant when the electron appears in the continuum.

AAS uses few-cycle near-circularly polarized pulses as a clock to measure tunnelling delays. The field of such a pulse with a Gaussian envelope $G(t)$ is (see figure (6)):

$$
\vec{E}(t) = G(t) \left[ E_\ell \sin(\omega t)\hat{x} + E_p \cos(\omega t)\hat{y} \right].
$$

(17)

This has three advantages. Firstly, the circular polarized pulses prohibit recombination or re-scattering of electron into the ion core. Secondly, a few-cycle pulse is a short pulse that comprise of not more than 1–1.5 optical cycles around the peak of the pulse. This ensures that the field varies very quickly such that the maximum ionization probability occurs only at one point, i.e. at the peak field. Thirdly and most importantly, it enables electric field vector at an instant in time $\vec{E}(t)$ to be mapped to an angle in space. The tunnelling forms the first step of the AAS technique, with the moment when electric field reaches its maximum value serving as the time zero, $t_\text{peak}$. The tunnelled electron is then acted upon by the electric field of the pulse following its appearance at the tunnel exit at time $t_0$, reaching its final momentum of $\vec{p}_f$:

$$
\vec{p}_f = \int_{t_0}^{\infty} \vec{E}(t)dt.
$$

(18)
Hence the final momentum is equal to the field’s vector potential at the instant of ionization \( t_0 \). Since the vector potential changes its direction constantly in the polarization plane with a frequency \( \omega \), the angular frequency, it sweeps every degree for \( \frac{\pi}{\omega \times 180} \) seconds. For a typical 800 nm central wavelength pulses, the relation goes as \( 1^\circ \approx 7.4 \) a.s. If the electron appears in the continuum at \( t_0 = t_{\text{peak}} \), where \( t_{\text{peak}} \) corresponds to the time when electric field reaches its peak value, \( E_0 \), the corresponding ‘tunnelling delay’ is taken to be zero. When seen in the polarization plane (see figure 6), this would imply that the electron flies in a perpendicular direction to that of \( E_0 \). We denote this shift of \( 90^\circ \) relative to the direction of \( E_0 \) as \( \theta_{\text{streak}} \). Any finite tunnelling delay \( t_1 = t_0 - t_{\text{peak}} \) would change the initial condition for streaking thereby leading to an additional offset angle, \( \theta_{\text{delay}} \).

Experimentally, the angular offsets are measured by finding the peak of the streaked electrons’ momentum distribution in the polarization plane and the peak field direction, measured using basic polarimetry. Cold target recoil ion momentum spectrometer (COLTRIMS) [126] is used to measure the asymptotic momentum \( p_f \). COLTRIMS is essentially a mass spectrometer which in addition to the time-of-flight information, is also equipped with position sensitive detectors. This enables the reconstruction of the 3D momentum vectors of the ionized fragments. On further advancements [127], COLTRIMS with its capability of detecting both electrons and ions in coincidence helped in studying the molecular fragmentation process and reactions. Thus it is also known by as Reaction Microscope (REMI), and it is regarded as the ‘bubble chamber of the atomic physics’ [127]. A detailed chronological development of COLTRIMS/REMI can be found in [126–132]. The direction of the peak field in the polarization plane is determined using a basic polarimetry measurement with a polarizer and a power meter that measures relative intensity. The maxima of the relative intensity as a function of polarizer determines the peak field as shown in figure 6.

In addition to the pulse duration, another crucial parameter to characterize a few-cycle pulse is the carrier-envelope-phase (CEP) of the pulse. This is defined as the relative phase offset between the maxima of the carrier field to that of the pulse envelope (see figure 7(a)). It is important to note that for a circularly polarized few-cycle pulse, the CEP determines the direction of the maximal field. The figure 7(b)-(c) shows the direction of the maximal field for a CEP of 0 and \( \pi/2 \). Hence it is important to stabilize the CEP [133, 134] before performing the experiment. However, by introducing ellipticity and making the polarization near-circular i.e. for ellipticity \( \varepsilon \geq 0.8 \) where \( \varepsilon = \frac{\text{length of minor axis}}{\text{length of major axis}} \), the maximal field always lies very close to the major axis of the polarization ellipse. This is illustrated for a 6 fs pulse of ellipticity 0.84 in figure 7(d). The maximum angular deviation of the peak field from the major axis as a function of CEP is \( \approx 2.5^\circ \). Thus, using near-circularly polarized pulse relaxes the constraint of having a stable CEP. Unlike in the case of phase stabilized pulses, the random CEP fluctuations average out to an effective peak field along the major axis, giving rise to two points of maximal field (two ends of the major axis of the polarization ellipse). Therefore basic polarimetry is sufficient to determine the major axis of the polarization ellipse along which the effective peak field lies. This was experimentally illustrated in the attoclock setting in [16].

6. A brief review of attoclock debate

It has been a decade since the first attoclock experiment [12] was performed. Yet, the controversy and debate surrounding the tunnelling times in the context of strong field ionization has not subsided. Tunnelling delay times were first measured using helium [12, 16] as a target. The angular offsets were calculated by finding the difference between the momentum distributions using semi-classical simulations assuming adiabaticity and
Figure 7. (a) A 6 fsec linear polarized pulse having a Gaussian envelope (black line) with carrier-envelope offset phase (CEP) 0 (red) and π/2 (blue). Their corresponding circular polarized pulses in the polarization plane are shown in (b) and (c) using the same colour code, with the arrow pointing towards the $E_{\text{peak}}$. The black circle in (b)-(c) is the polarization ellipse and also happens to be the locus of the peak field, when CEP is not stabilized. (d) Parametric plots of a 0.84 elliptic polarized light shown as a function of CEP increasing from 0 to $7\pi/4$ in steps of $\pi/4$ (from left to right 0-3$\pi/4$ on top and $\pi-7\pi/4$ in the bottom). The black arrows point the direction of maximal field. The outer black dotted line correspond to the locus of maximal field for a circular polarized field with a random CEP. In case of 0.84 as ellipticity, a maximum angular deviation of peak field w.r.to CEP is $\sim 2.5^\circ$ occur away from the major axis. However as CEP gets randomized, the effective maxima is located along the major axis. The figure is adopted from [61].

...The experimentally measured momentum distributions. The experiment was performed in an intensity range of $2.3 \times 10^{14}$ W/cm$^2$ to $3.3 \times 10^{14}$ W/cm$^2$, corresponding to $\gamma = 1.45$ to 1.17. It was found that the tunnelling delays were zero within an uncertainty of 12 asec versus what $\tau_{\text{BL}}$ or $\tau_{\text{Keldysh}}$ would predict which is $\sim 500$ attoseconds. Thus, the authors concluded that there were no signs of real tunnelling delays. Further, the authors included the contributions of possible parent ion-electron interaction during the streaking process in their semi-classical simulations. Although the contribution from the Coulomb interaction towards the measured offset was non-zero, it remained constant across the intensity range. This was soon followed by a detailed description of a semi-classical model [22] that did not include any Coulomb contributions which also interestingly explained the experimental results successfully.

The semi-classical model in [22] uses the ADK formula in combination with the post-tunnelling classical trajectories of the unbound electron. But such a model had not gone uncontested. Although this theory would come handy for experimentalists to qualitatively (and quantitatively to some extent) gauge their results instead of relying on more computationally challenging TDSE simulations, it is prone to some limitations. To begin with, the authors themselves mentioned a limitation of such a theory that lies in a scenario where the ground state depletes at higher intensities. Later [23] pointed out that, in addition to the depletion effects, it is possible to ionize electrons before the field reaches $E_{\text{peak}}$ at higher intensities. This time was calculated to be more than the typical tunnelling delays. Using 3D-TDSE codes, [135] showed that counter-intuitive angular shifts can be observed in argon, due to the effect of the ever existing Coulomb field. They also showed that the theories like ADK or SFA can be misleading in predicting the most dominant direction of the ionized electrons. Hence, all these posed a challenge in interpreting the angular offsets in terms of tunnelling times. In fact, this was acknowledged in a successive experiment carried out on argon and helium again [17, 18], albeit in a broader intensity range ($\sim 3 - 15 \times 10^{14}$ W/cm$^2$) with 7 fs pulses:

‘...This Coulomb correction is especially sensitive to the ion-electron attraction at the beginning of the electron trajectory. In general however this angular offset $\theta$ is much more complicated and not fully explored and understood to date. ...’ [17]

It was noted that to provide an appropriate semi-classical theory it is important to know the initial conditions of the electron emerging into continuum. This depends strongly on the tunnelling geometry. To this end, a new semi-classical model was introduced, analyzing the system in parabolic coordinates, that includes the Stark effects, namely, tunnel ionization in parabolic coordinates with induced dipole and Stark shift (TIPIS) model. This model in conjunction with the AAS technique also concluded that there were no real

---

It was already well known that for ellipticity (around 0.1-0.5) the Coulomb field can alter the outgoing electron trajectory and affect the photoelectron momentum distribution in the polarization plane which was observed experimentally [136, 137] and then described within different approximations first qualitatively [138, 139] and on the quantitative level [140].
Table 1. Considering $T = |T_e|^n$ as the transmission coefficient, the four popular definitions of tunnelling time can be expressed as a derivative of transmission amplitude ($|T_e| \propto$ probability) or phase of transmitted wavepacket w.r.t to either energy of the initial wavepacket ($E_n$) or the barrier potential ($V$).

| Potential ($V$) | Amplitude ($|T|$) definition | Phase ($\theta$) definition |
|----------------|-------------------------------|-----------------------------|
| $\tau_{BL}$  | $-\hbar \frac{\partial \theta}{\partial (|T|)}$ | $-\hbar \frac{\partial \theta}{\partial (|T|)}$ |
| $\tau_{PM}$  | $\hbar \frac{\partial \ln |T|}{\partial n}$ | $\hbar \frac{\partial \ln |T|}{\partial n}$ |
| $\tau_{LM}$  | $\hbar \frac{\partial \ln |T|}{\partial V}$ | $\hbar \frac{\partial \ln |T|}{\partial V}$ |
| $\tau_{WES}$ | $\hbar \frac{\partial \ln |T|}{\partial E}$ | $\hbar \frac{\partial \ln |T|}{\partial E}$ |

tunnelling delays, but demonstrated how electron-electron correlations in the atom could possibly complicate the trajectories and thereby the interpretation of angular offsets.

The two sets of experiments with helium only went on to show zero tunnelling delays, partly because the latter one was done at the higher end of the intensity range that enters the over-the-barrier ionization regime. The claim for a finite tunnelling time in the context of strong-field ionization was only reported a year later [21, 109], when the experiment was transferred to a velocity-map imaging (VMI) apparatus [20], due to its better count rates even at lower intensities. This third set of experiments probed tunnelling delays in a wide range of intensities ($0.73-7.5 \times 10^{14}$ W/cm$^2$) varying the barrier size by almost 3 times. The measured angular offsets were non-zero after considering the Coulomb corrections obtained from the TIPIS model and were attributed to real finite tunnelling times.

The experimental observation was compared with the known popular tunnelling times $\tau_{BL}$, $\tau_{PM}$, $\tau_{LM}$ and $\tau_{WES}$ as given by their definitions mentioned in Part I. For a quick overview for the reader, the four definitions, although their origin is from different physical models, can be described as partial derivatives of transmission probability and phase of the tunnelling wavepacket as a function of barrier height or energy of the incoming wavepacket (see table 1). The tunnelling time definitions relying on the potential could be complex summed to be called as the resident time $\tau_D$ as both the times had their origin in measuring the interaction time with the barrier. In the formulation of Feynman path integral method, these correspond to summation of all times that the paths spend inside the barrier. On the contrary the complex summation of the energy dependent definitions are called as passage time $\tau_P$, since they relate to the phase of the outgoing wavepacket after it had interacted with the barrier. In terms of Feynman path integral formulation, these correspond to the total time that all the Feynman paths spend outside the barrier. Finally, from the probability distributions of $\tau_D$, $\tau_P$, individual tunnelling times could be identified as real and imaginary numerical values of their expectation values. The above method was employed for interpreting the attoclock data by modelling the tunnelling barrier as a triangle and the distribution function of tunnelling time $f(\tau)$ was calculated [109]. Different tunnelling times were numerically determined, but which one should be considered the correct one was an open question. The authors reported [109, 141] that the times calculated from the observed non-zero offsets were in good agreement with the Larmor time $\tau_{LM}$ (see figure 11(b)). In addition to this, they also showed that the peak value of $f(\tau)$ predicts the tunnelling time better.

The procedure of Feynman path integral is appealing, since, unlike the semi-classical models, it treats the whole problem quantum mechanically and is equivalent to considering the electron as a wavepacket. But it should be noted that the path integral approach goes to prove that the tunnelling time is not a deterministic quantity but a probability distribution, which further complicates the interpretation of the observed angular offsets as originating from tunnelling time. On the other hand, recent works [113] using the symmetric time $\tau_{sym}$ and entropic tunnelling time based on statistical approach [142] definitions, while treating the electron as a particle during tunnelling process together with the time-energy uncertainty relations, claimed to be in good agreement with the attoclock experimental data. A detailed review of this work and how its interpretation of tunnelling time is different from path integral approach can be found in [31].

As new results continued to be reported, the debate continued in the strong-field community as the angular offsets could not be reproduced by any of the known strong-field theoretical models. This is a serious concern as the calculations of angular offsets, from which time is deduced, strongly rely on the theoretical models used. This disagreement could be assigned to the non-adiabatic nature of interaction, which shall be taken care of in the process of calibrating intensities and ionization rates and/or the failure of modelling helium within the single-active electron approximation. The justification for using adiabatic models was argued in [24] by showing that non-adiabatic nature of interaction could not predict the qualitative trend as observed in the experiment. This argument was soon disputed by a paper that showed the opposite [25] and had put the assumptions of the semiclassical model TIPIS to question, challenging its validity. The argument for electron-electron correlations causing the offsets was apparently put to rest [30] by a 3D-TDSE code developed based on the hybrid anti-symmetrized coupled channels method [143], which can completely solve helium interacting with an elliptically polarized light. These results also could not reproduce the measured offsets in the attoclock setting for helium. Currently, the observations remain in disagreement with the state-of-the-art strong-field models, leaving the question of tunnelling time in strong-field physics open.
Recently, an attoclock experiment performed on a mixture of krypton and argon gases reported to have measured tunnelling times around ~ 100 as [19], by measuring relative offsets between the two species. The authors argue that by making use of relative offsets, they could cancel out the contributions of the Coulomb attraction by the parent ion. They used a higher wavelength laser pulses to satisfy the condition \( \gamma < 1 \), so as to overcome the complications due to non-adiabaticity. Wigner delays were used to find the most dominant quantum path that exit the tunnel at point \( x_{\text{exit}} \) at time \( t_{\text{exit}} \) with longitudinal momentum \( p_{\text{exit}} \). These are then used as the initial conditions for the classically propagating electron in the laser field. In spite of a compelling analysis, it still suffers from the disadvantage of inability to determine the initial wavepacket of electrons in its bound state, which is crucial in calculating Wigner delays. Argon and krypton are two complex systems that cannot be solved completely using 3D-TDSE. In experiments such as these, where precision is highly significant, it is necessary for one to closely examine the effects of approximations considered. Also, the effect of electron-electron correlations on the angular offsets were not discussed keeping the interpretation of the tunnelling times in doubt. In addition to that, Ar and Kr have very different ionization potentials, which could have resulted in different distributions of ionization times within the relatively long laser pulses used in those experiments.

6.1. Role of atomic \( H \) in resolving the debate

Previous experiments investigating tunnelling were performed on different multi-electron targets like Ar, Kr, and He. These target species are quite complex and interpretation of the results relies on the theoretical approximations that one chooses in their models. This makes it difficult to interpret the experimental results unambiguously. The difficulties are primarily due to (i) the inability to perform \textit{ab initio} simulations with exact potentials that can account for electron-electron correlations and (ii) relying on semi-classical models that fail to estimate the contributions from long-range parent ion-electron interactions. It is therefore believed that an experiment with \( H \) could possibly resolve the debate, as it requires no assumptions on tunnelling models to simulate the attoclock experiment using 3D-TDSE and those precise simulations can be compared with the experimentally measured offsets. Due to the simplicity of the \( H \), it serves as a physical two-body system that can be treated analytically and yields solutions in closed form. Hence \( H \) could be used as a point of reference for our understanding of the inherently complex dynamics of light-matter interactions. In fact, measurements on hydrogen atom of ever improving accuracy led to emergence and refinement of our most successful and quantitatively precise scientific theory - quantum mechanics - via development and validation of Bohr model in 1913, Schrödinger equation in 1925, Dirac equation in 1928 and Quantum Electrodynamics (QED) in 1948.

In context of strong-field physics \( H \) remains the only system for which accurate \textit{ab initio} calculations were performed and validated by comparison with experimental measurements. The importance of \( H \) in benchmarking strong-field physics was demonstrated experimentally [144] and with an excellent quantitative agreement between theory and experiment. Based on that quantitative agreement, by measuring the ionization cross-sections of \( H \) against various strong-field models and 3D-TDSE, few-cycle laser pulse intensities were calibrated within an accuracy of 1% [145]. Unfortunately, \( H \) is not naturally available in the earth's atmosphere and is a challenging task to produce in a laboratory. A secondary calibration standard was created using noble gases [146], that does not require a source of atomic \( H \). Later, the absolute CEP of few-cycle pulses was determined using \( H \) [147]. The same study also exposed the pitfalls of the commonly used strong-field models based on single-active electron approximation, that produced at least a 0.25 radian systematic offsets in tagging absolute CEP. Precise knowledge of experimental pulse parameters, such as peak intensity and absolute CEP, in the case of few-cycle laser pulses, in its turn allows further quantitative comparison between theory and experiments and validation of various approximate models based on that comparison. It is critical to properly establish validity of any model before using it to explain and interpret the experimental results unambiguously. It was shown that for \( H \) no approximate theory, short of high-quality 3D-TDSE solutions properly folded with well-characterized apparatus functions and averaged over focal and target geometry, could faithfully reproduce experimental electron momentum distributions.

Therefore, \( H \) offers unique opportunities for benchmarking and validating various experimental and theoretical techniques in strong-field physics. Careful experiments on \( H \) can yield data that quantitatively agrees with the theoretical predictions to within experimental uncertainty. Both the data and the predictions are validated by such agreement. Clearly, this advantage of \( H \) can be utilized also for measuring and understanding tunnelling delays using attoclock.

There were theoretical studies done to this end, but they too differed in their predictions [27–29]. While [27] and [29] argued in favour of instantaneous tunnelling, [28] calculated various well-known tunnelling times including the one formulated using Bohmian mechanics (see figure 10 (a)). All of these turned out to be non-zero according to their model. Zimmermann \textit{et al} [28] argued that Larmor time is the right choice of tunnelling time that one should use to describe a scenario like attosecond angular streaking and predicted
~ 100 attoseconds (at lowest intensity where they were calculated) as tunnelling time in strong-field ionization of atomic H. Later [29] used 3D-TDSE simulations to calculate the asymptotic photoelectron momentum distribution from H and subsequently used classical back-propagation technique to infer the initial conditions for the electron at the tunnel exit. By doing so, they could not only determine tunnelling to be instantaneous but also provided a relation of how the calculated angular offsets can be used to infer ‘proper’ tunnelling times. In addition to determining the fraction of electrons that ionize due to tunnelling process, they concluded that most of these electrons tunnel instantaneously. Therefore it is required of us to verify and validate these predictions experimentally.

Having first validated the theoretical model by its comparison with experimental measurements, we proceeded to replace the Coulomb potential with a short-range (Yukawa) potential, which was found to result in zero angular offsets corresponding to vanishing tunneling delays. This, we believe, definitively resolves the attoclock debate, proving conclusively that angular offsets are not caused by any finite time delays originating from one-electron tunnelling dynamics. Rather the measured (and calculated) angular offsets are caused entirely by electron scattering on the long-range Coulomb potential of the proton.

7. Attoclock with H

7.1. Central idea in determining tunnelling times

The issue of multi-electron interactions in modelling the potential could easily be overcome by using H. Though 3D-TDSE simulations can predict the exact values of angular offsets, they lack the access to internal dynamics that we are interested in, i.e. the long range interactions. Those simulations can not disentangle the contribution of the Coulomb force on the electron due to the ion from the real tunnelling time to the observed angular offsets. This was indeed the motivation for people to rely on semi-classical models wherein the whole process can be analyzed in two or three steps at a sub-system level, adding a more meaningful physical picture to the interpretation. But then the semi-classical models, in the present context, could not unambiguously evaluate the effects of long-range interactions as reviewed in section 6. Also, the Coulomb attraction experienced by the electron can not be eliminated in reality while performing experiments.

Performing experiments on negative ions, as some suggested, brings back the electron correlation issue, while not completely eliminating the relatively long-range induced dipole potential. It appears that the only way to surmount both multi-electron dynamics and Coulomb potential issues is to perform a numerical experiment on atomic hydrogen with a short-range (i.e. Yukawa) potential and to calculate the resulting angular offsets. That is exactly what Torlina et al did in [27]. They reported zero angular offsets for a Yukawa potential and concluded that tunnelling in H is instantaneous. However compelling, that purely theoretical argument failed to convince everyone. There always remains a possibility that a theory not validated by experiment could be simply wrong. Also, Torlina et al used unrealistic single-cycle circularly polarized CEP-locked pulse and determined angular offsets by finding the maximum of the calculated electron momentum distributions, which precluded experimental validation of their results even in principle. We adopted the same approach of replacing the Coulomb potential with a Yukawa potential in a numerical experiment. But first we validated our theoretical model (or rather two independently developed models) by performing attoclock experiment and corresponding calculations on H with well characterized realistic laser pulses. We confirm the conclusions of Torlina et al - the angular offsets are zero for the short-range Yukawa potential over the full range of intensities.

In order to determine the tunnelling delays unambiguously, we first compare our results with the full solutions of ab initio 3D-TDSE calculated accurately within the non-relativistic framework using the electric dipole approximation, that generate photoelectron momentum distribution projected on the polarization plane with the same pulse parameters as were used in the experiments. Once a good agreement is found, we then use the same numerical codes solving 3D-TDSE to artificially eliminate the parent ion-electron long-range attraction by choosing a short-range potential. [27]. The short range potential used is the Yukawa potential that binds the electron to proton, in such a way that the ground state energy of H atom is retained i.e. 13.6 eV as shown in figure 8(a). It is of the form,

\[ V(r) = -\frac{Z}{r} e^{-r/\alpha} \quad \text{where} \quad Z = 1.908 \ a = 1. \tag{19} \]

Therefore any offsets observed in the photoelectron momentum distribution generated by Yukawa potential would mean a real finite tunnelling delay. Therefore it is required of us to verify and validate these predictions experimentally.

6To this end numerical simulations were done with \( F^-/Cl^- \) ions using realistic parameters as used in [33] which showed no angular offsets [148]. However no comment was made by the authors to relate these results with tunnelling time.
Figure 8. (a) Potential curves showing both Coulomb potential (blue dot-dashed curve) given by \( V(r) = -1/r \) and a short range Yukawa potential (red thick curve) given by \( V(r) = Z/\beta r \exp(-\beta r) \) such that the electron is bound to a proton with the same binding energy as \( -13.6 \) eV or \(-0.5\) a.u. (b) Photoelectron momentum distribution projected into the polarization plane, using the full numerical solution of 3D-TDSE with Coulomb potential (shown with blue colour in (a)) at \( 1 \times 10^{14} \) W cm\(^{-2} \), with experimental parameters. (c) Photoelectron momentum distribution projected into the polarization plane, using the full numerical solution of 3D-TDSE with Yukawa (shown in red curve in (a)) potential at \( 1 \times 10^{14} \) W cm\(^{-2} \), with experimental parameters. The sub-figure (a) is adopted from [33].

Figure 9. The experimental observations are found to be in good agreement with the ab initio 3D-TDSE simulations with both Coulomb potentials provided by two independent groups marked as 1 and 2. The contribution of Coulomb field to the offsets is deducted by running the validated 3D-TDSE numerical codes with short range Yukawa potential shown in red and blue triangles. Same extraction procedure was used to determine the offset angles from experimental results and theoretical simulations for both Coulomb and Yukawa potentials. The numerical experiments confirm that the observed angular offsets are due to the electron scattering by the long-range interaction from the parent-ion. The figure is adopted from [33].

7.2. Experimental results
The experiment was performed using a few-cycle femtosecond laser (Femtopower Compact Pro) generating 6 fs pulses around 770 nm, which are converted to near-circular polarized pulses of ellipticity \( 0.84 \pm 0.01 \). The \( H \) gas beam-line is produced through an RF-discharge source [149, 150] that is integrated with the REMI chamber through a differential stage maintained under ultra-high vacuum [61]. As part of the gas beam line, a uniform field is applied through a pair parallel plates, to deflect out the ionized fragments (electrons or ions from the discharge source) either from \( H \) or \( H_2 \). In addition to this, the uniform DC-field of 26 V cm\(^{-1} \) produced by the plates and 23 V cm\(^{-1} \) produced within the spectrometer in the REMI reduces the lifetime of any metastable hydrogen atoms (\( H \) in 2s state) to few 100s of nanoseconds leaving only the ground state \( H \) in the interaction region. The electrons and ions from \( H \) are measured in coincidence in REMI and then the photoelectron momentum distribution projected onto the polarization plane is analyzed for determining the offset angles. The radially integrated momentum distribution is fitted with a double-Gaussian function to find the polar angle of the most probable asymptotic momentum of the tunnelled electrons. After a careful calibration of polarizer in the REMI’s frame of reference, polarimetry is performed in order to find the angular orientation of the peak field of the ionizing pulse and thereby the angular offsets.

The \( H \) experimental data are compared with the numerically modelled photoelectron momentum distributions using the same experimental parameters. The ab initio numerical simulations of 3D-TDSE are provided by two independent theoretical groups that are conceptually similar, and use Matrix Iteration Method to propagate the TDSE in time. Detailed descriptions of the numerical techniques used by the two groups can be found in [151, 152]. Specifically, the components of the vector potential \( \vec{A}(t) \) for the pulse with ellipticity \( \varepsilon \) were set as:
Figure 10. (a) Shows the tunnelling time estimates calculated for \( H \) using various tunnelling time definitions by Zimmerman et al [28]. The figure is adopted from [28]. (b) Shows the zoomed view of the Larmor time estimates (with same colour code) for \( H \) that was found to be most relevant (and also the least) for attoclock measurements in comparison to the possible tunnelling delays in experiment calculated by the validated 3D-TDSE numerical codes using Yukawa potential from [33]. The Larmor time curves in the graph are digitized from [28], with the permission of the author. Please note that (a) is shown in logarithmic scale while (b) is in linear scale in \( Y \)-axis.

\[
A_x(t) = -\frac{E_0}{\omega} G(t) \frac{\epsilon}{\sqrt{1 + \epsilon^2}} \cos(\omega t + \phi),
\]

\[
A_y(t) = -\frac{E_0}{\omega} G(t) \frac{1}{\sqrt{1 + \epsilon^2}} \sin(\omega t + \phi),
\]

\[
A_z(t) = 0.
\]

The envelope function \( G(t) \) was a Gaussian ramped on and off over three optical cycles each, respectively such that the FWHM was 6 fs in intensity. The electric field was obtained as \( E(t) = -\frac{\delta G(t)}{\delta t} \). Finally, the carrier-envelope phase \( \phi \) was varied in steps of \( \pi/4 \) from 0 to \( 2\pi \) and the results were averaged. A careful check was performed in order to see that the solutions reach converge to the required accuracy with respect to the parameters and a comparison has been made between the two independent calculations at an intensity of \( 1.65 \times 10^{14} \text{ W cm}^{-2} \), to ensure that the independent calculations mutually agree within the error caused by the fitting routine.

The experiment was performed in the intensity range of \( 1.65 \times 10^{14} \text{ W cm}^{-2} \) to \( 3.9 \times 10^{14} \text{ W cm}^{-2} \). The angular offsets from the experiment are directly compared to the \( \text{ab initio} \) simulations provided by two independent theoretical groups. An excellent agreement is found between the experimental data and the theoretical simulations (shown as circles in figure 9) from the two independent groups, which are also in mutual agreement. Thus, both the experimental data and the numerical simulations were mutually validated.

An expected trend of decreasing angular offsets (shown in figure 9 with black diamonds) is observed as was explained in the previous experiments. The explanation was that the increasing field-strength reduces the height and width of the barrier and thus decreases the time electrons spends under the barrier, leading to a faster tunnelling time and thereby lesser angular offsets. This explanation is however contested due to the lack of possibility to segregate the contributions of Coulomb attraction of the parent-ion to the angular offsets. As discussed earlier, we overcome this hurdle by performing computational experiments with the validated numerical code, albeit using a short range Yukawa potential. The photoelectron momentum distribution generated in such a way using both Coulomb and Yukawa potential at an intensity of \( 1 \times 10^{14} \text{ W cm}^{-2} \) is shown in figure 8(b) and (c) respectively.

The computations with Yukawa potential yield zero angular offsets (within the numerical and fitting uncertainty) due to tunnelling (triangles in figure 9). This holds true for the entire range of intensities where the experiment and calculations were performed. The computations were performed even below the lowest experimental intensity right from \( 1 \times 10^{14} \text{ W cm}^{-2} \) and the results are always the same. Thus our results confirm that the tunnelling delays in the strong-field ionization of \( H \) cannot exceed 1.8 attoseconds, which is the uncertainty in determining offsets. That upper limit applies to Yukawa potential, which presents a much larger barrier to tunnelling, and therefore it should also hold for the real Coulomb potential. That also leads to the definitive conclusion that the offset angles are entirely due to the Coulomb scattering of the unbound electron with no contribution from any tunnelling delays.

8. Comparison of attoclock experiments

We quickly compare and summarize the set-up and results of all the major attoclock experiments performed on \( \text{He} \) [109], \( \text{Ar-Kr} \) [19] and \( H \) [33]. The experimental method of all these experiments are same that is the
attoclock angular streaking technique with around similar ellipticity i.e. 0.84–0.87. While the experimental data of \(\text{He}\) was recorded using a VMI due to its higher count rate at lower intensities and higher repetition rate of 10 kHz, the latter two experimental data were recorded using REMI at 1 kHz repetition rate. Though the attoclock experiment with VMI has an advantage of recording data with higher count rate and with less acquisition time, it comes with the drawback of losing coincidence imaging\(^2\). REMI on the other hand allows one to record complete kinematic information of electron in coincidence with its parent ion, which is very crucial when there are background gases with ionisation potential either similar or lower than the target species as in \(\text{H}\) experiment or in cases where the target is a mixture as in the \(\text{Ar-Kr}\) experiment. Hence, in the \(\text{He}\) experimental set up it was important to ensure that there is negligible molecular contaminant while recording photoelectron momentum data. This was done by switching to ion measurements and checking the presence of any other molecular/atomic species. VMI primarily gives the projected 2D information of the 3D electron momentum distribution which had to be then reconstructed to calculate the angular offset in the plane of polarisation. The effort in switching from REMI to VMI does have its advantage in achieving higher resolution in momentum imaging of electrons, provided appropriate filters are used for reconstructing 3D momentum. This can be seen in the reduction of error bars for the measured angular offsets from 2–2.5\(^\circ\) to 1–1.5\(^\circ\) in figure 11(a). This is also true when we compare the error bars in \(\text{H}\) experiment that used REMI which had error bars ~ 2 – 2.5\(^\circ\) in figure 9. However it must be noted that the errors bars in the \(\text{Ar-Kr}\) experiment (see figure 11(c)) is smaller than that of the other two when done in REMI due to two possible reasons- higher count rates and fitting procedure. The total ionisation events of \(\text{He}\) are not as good as those of \(\text{Ar-Kr}\) due to high ionisation potential and low density in case of \(\text{H}\) since producing atomic \(\text{H}\) with larger dissociation ratio and therefore more count rates/ pulse is challenging. These could of course be surpassed by merely acquiring the data for longer time. But unfortunately, it was not possible in the case of \(\text{H}\), especially for lower intensities owing to the issues of laser stability. The second and most probable reason is due to the fact that the entire radially integrated electron momentum distribution is fitted with a double Gaussian function in both \(\text{H}\) and \(\text{He}\) experiments, while only the region of ± 50\(^\circ\) around the peak is fitted with Gaussian in case of \(\text{Ar-Kr}\) data. In addition to these, the experiments on lighter atoms had to consider propagation of errors arising from measuring the polarisation ellipse in order to determine the peak field, which was bypassed in the \(\text{Ar-Kr}\) experiment due to relative offset measurements, plotted in figure 11(c). In contrary to the experiments done on lighter atoms \(\text{H}\) and \(\text{He}\), the \(\text{Ar-Kr}\) employed longer wavelengths to bypass adiabaticity issues that were raised in [17]. The flip-side to this is that it would decrease the relative accuracy due to longer optical-cycle-to-angle conversion as compared to shorter wavelengths.

The obvious shall anyway be stated that though each set of experiments had their own set of advantages over the other, the results were quite contrasting when it comes to the conclusion they drew about the tunnelling time in strong-field ionisation. While the \(\text{He}\) set-up had the advantage of performing the experiment at higher momentum resolution with very high counts rate, it was handicapped by the lack of coincidence imaging. The \(\text{Ar-Kr}\) set-up employed clever technique of measuring relative angular offsets and longer wavelengths to ensure adiabatic tunnelling regime. In addition to the reduction of error bars as

\(^2\text{There are ways to do coincidence imaging with VMI on a single detector that are reported in [153, 154].}\)
explained before for the measured relative offsets, the configuration also claim to cancel offsets due to Coulomb drag. But this could only be true in a simple picture like SAE approximation and for distances far from the tunnel exit. They have also bypassed the issue of intensity calibration, which was an issue in interpreting He data, by directly plotting the data as the function of $\rho(\theta_{\text{max}})$ which is defined as the average electron momentum given by the radius of the maximal counts in the polarisation plane. Although it does not matter for the experimental data, intensity is an inevitable parameter in interpreting results through numerical simulations using TDSE. Effects like Stark shifts, ground state depletion and saturation, screening potential etc have been considered in interpreting data, which by themselves are a testimony to how complex the Ar-Kr system is to model. As mentioned earlier, determining the bound state wavefunction is key to estimate the Wigner phase times and it is not desired to have complex systems to be used as target species to determine ultra precision measurements like these. Conclusions from He data [28, 109] ruled out Wigner times as the possible explanation for tunnelling delays and proposed Larmor times as the real tunnelling time (see figure 11(b)). In contrast, [19] using Ar-Kr data concluded Wigner time as the time spent by an electron under the barrier during strong-field ionisation. They also argue that due to the variation in barrier width arising out of different binding energies, there would relatively be only a difference of 10 as or ~ 1° (see figures 11(c) and (d)) that the electron spends more time in case of Ar over Kr at the same intensity of external field. Such an interpretation is faulted in the recent reviews on attoclock [155, 156]. As there seems to be no consensus in the strong field community about the impact of electron-electron correlations on measured offsets pertaining to the He data [30, 155, 156], the Ar-Kr paper [19] do not address the issue of electron-electron correlations as they use the screening potential with SAE approximation [111, 114].

Though [19] could explain the experimental data of Ar in [17], it would be interesting to see if the same theory could also explain the He data in [17, 109] where the electron-electron correlations are predominant over other effects. Most of these disadvantages were circumvented by using the simplest atomic system H that could be benchmarked against 3D-TDSE which can be solved numerically to arbitrary precision in closed form under non-relativistic dipole approximation [33]. Subsequent analysis with Yukawa potential as described above confirms unambiguously that the entire angular offsets are due to Coulomb force exerted by the parent ion.

9. Current status of the discussion

The results from the first attoclock experiment with benchmarking atomic species, H are consistent with instantaneous tunnelling, i.e. there is zero time delay between the peak electric field and the appearance time of the continuum electron measured by the attoclock. Furthermore, it is also clear that the upper bound of 1.8 attoseconds is at least an order of magnitude smaller than the popular tunnelling time estimates as predicted in [28] for H, shown in figure 10(b). Thus in the attoclock setting, these definitions of tunnelling time as the time taken by an electron to traverse the barrier can be convincingly ruled out. The unbound electron’s scattering on the Coulomb potential of the proton results in the observed angular offsets. The intensity dependence of those offsets could be explained using a simple classical Rutherford-like scattering process, considering the tunnelling distance from Keldysh theory as the impact parameter and the vector potential of the driving pulse as the asymptotic velocity in the Rutherford formula [34]. It is interesting to see that this Keldysh-Rutherford model also qualitatively captures the frequency (wavelength) dependence of the estimated angular offsets, as predicted using Monte-Carlo simulations in [157].

Following the attoclock experiment with H, Kullie reinterpreted the experimentally measured offsets based on the definition of symmetric time [31, 113] and reported that the tunnelling time is finite [32]. The author completely neglected the effect of Coulomb potential on angular offsets and treated the measured angular offsets as a true measure of tunnelling time. Although replacing Coulomb potential with Yukawa potential [33] was questioned, no reason for the objection is found in the article. Kullie further argues that the idea of the Coulomb tail leading to angular offsets is a consequence of an imaginary time picture. He also claims that instantaneous tunnelling would mean a direct violation of special relativity as the speeds are super-luminal. In contrast to the above interpretation leading to finite tunnelling times, Pollak [158, 159] defines tunnelling flight time wherein the tunnelled wavepacket through a barrier is measured in a time-of-flight kind of measurement (that resembles closely to what is done in an attoclock measurement within the REMI). He went on to show that such tunnelling flight time vanishes when measured, yet does not violate special relativity, generalising Hartman effect [56]. The vanishing tunnelling times are also in agreement with the methodology of using a virtual detector at the end of the tunnel to understand strong field phenomena [29].

In addition to taking sides on whether tunnelling times are finite or vanishing, there is also an argument in favour of the very question about tunneling time being invalid and ambiguous [160]. In the context of attoclock, a recent article by Sokolovski et al indeed argues that the attoclock cannot reveal any information
regarding the time spent by the electron under the barrier [161]. However, in his article the author presumes that the $H$ attoclock experiment measures the Wigner-Smith delays, $\tau_{WES}$. But in reality the Wigner delays as calculated for $H$ [28] (blue colour curve in figure 10(a)) are two orders of magnitude higher than what attoclock measurement with $H$ [33] reports. As we already pointed out, attoclock does not measure any of the theoretically proposed delay times. It very specifically measures the time difference (delay) between the moment the electric field reaches its maximum value and the moment the peak of the electron wavepacket appears in the continuum. We put a strict upper limit on that delay for atomic hydrogen which is consistent with that delay being zero within our numerical and fitting errors. Whether that delay can be interpreted as ‘tunneling time’ or ‘time spent under the barrier’ is indeed an ill-posed question, as neither of those concepts is sufficiently well-defined and unambiguous to answer it.

Nevertheless, attoclock can still be a useful tool to explore the effects of electron-electron correlations on the measured angular offsets. Experimentally exploiting the advantages of all the three different experimental set-ups discussed in section 8 could definitely lead to a better design. For instance, measuring relative offsets of $H$–He at higher count rate is certainly something that the community can look forward as both the target species symbolise benchmarking species for understanding atomic system and electron-electron correlations. However the vast difference in the ionisation potential of these two species pose a challenge experimentally to exploit the right intensity regime to explore tunnelling delays.

10. Conclusion

It is important for the tunnelling time community to understand that the attoclock debate has its origin primarily in the inability of strong-field models to predict accurately the ionization dynamics of complex systems like noble gases. This, in turn, led to the claims of instantaneous [16–18] or finite [19, 21, 109] tunnelling times which, in the first place were not validated by accurate strong-field models. This has necessitated the experiment with $H$ and the conclusion that is drawn from the experiment (as analyzed the same way as other experiments, albeit accurately accounting for all the interactions) has effectively ruled out all common definitions of tunnelling times. In order to make this message clear, we presented a brief overview of the tunnelling time debate started in the late 1980’s, whose definitions had a major role to play in the current attoclock tunnelling delay experiments. We also described the technique of attoclock as it was conceived based on the semiclassical strong-field models. We then presented an overview of the attoclock measurements performed in the past decade and their contradicting results and claims. We argued that performing attoclock experiment on ($H$) was the only way to resolve this controversy. We then presented such a study of $H$ that puts an upper bound of 1.8 as on the possible tunnelling delays as defined in the attoclock setting. We do not claim that we have said the final word in this debate. Nonetheless, we hope that this review will clarify some misconceptions and benefit those who are interested in this question.

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References

[1] Devault D 1980 Quarterly Rev. Biophys. 13 387–564 (and references therein)
[2] Zener C 1934 Phil. Trans. R. Soc. A 145 523–9
[3] Esaki L 1958 Phys. Rev. 109 603–4
[4] Tao N 2010 Electron transport in molecular junctions Nanoscience And Technology: A Collection of Reviews From Nature Journals (Singapore: World Scientific) pp 185–93
[5] Bockrath M, Cobden D H, McEuen P L, Chopra N G, Zettl A, Thess A and Smalley R E 1997 Science 275 1922–5
[6] Grabert H and Devoret M H 2013 Single-Charge Tunneling: Coulomb Blockade Phenomena in Nanostructures vol 294 (New York: Springer)
[7] de Carvalho C A and Nussenzveig H M 2002 Phys. Rep. 364 83–174
[8] Winful H G 2006 Phys. Rep. 436 1–69
[142] Demir D and Güner T 2017 Ann. of Phys. 386 291–304
[143] Majety V P, Zielinski A and Scrinzi A 2015 New J. Phys. 17 063002
[144] Kielpinski D, Sang R and Litvinyuk I 2014 J. Phys. B: At. Mol. Opt. Phys. 47 204003
[145] Pullen M et al 2013 Phys. Rev.A 87 053411
[146] Wallace W et al 2016 Phys. Rev. Lett. 117 053001
[147] Khurmi C et al 2017 Phys. Rev. A 96 013404
[148] Douguet N and Bartschat K 2019 Phys. Rev. A 99 023417
[149] Pullen M G 2011 Above threshold ionisation of atomic hydrogen using few-cycle pulses PhD Thesis Griffith University
[150] Wallace W C 2017 Strong field ionisation of atomic Hydrogen PhD Thesis Griffith University
[151] Ivanov I 2014 Phys. Rev. A 90 013418
[152] Douguet N, Grum-Grzhimailo A N, Gryzlova E V, Staroselskaya E I, Venzke J and Bartschat K 2016 Phys. Rev. A 93 033402
[153] Lehmann C S, Ram N B and Janssen M H M 2012 Rev. Sci. Instrum. 83 093103
[154] Zhao A, Sándor P and Weinacht T 2017 J. Chem. Phys. 147 013922
[155] Hofmann C, Landsman A S and Keller U 2019 J. Mod. Opt. 66 1052–70
[156] Kheifets A S 2020 J. Phys. B: At. Mol. Opt. Phys. 53 072001
[157] Liu J, Fu Y, Chen W, Lü Z, Zhao J, Yuan J and Zhao Z 2017 J. Phys. B: At. Mol. Opt. Phys. 50 055602
[158] Petersen J and Pollak E 2017 The J. Phys. Chem. Lett. 8 4017–22
[159] Petersen J and Pollak E 2018 The J. Phys. Chem. A 122 3563–71
[160] Sokolovski D and Akhmatskaya E 2013 Ann. of Phys. 339 307–27
[161] Sokolovski D and Akhmatskaya E 2018 Commun. Phys. 1 47