QUANTUM PHYSICS

Attoclock and tunnelling time

The time it takes for a particle to tunnel through a potential barrier, and even the interpretation of this phenomenon, have long drawn debate. By performing an attosecond angular streaking experiment in connection with ab initio calculations, researchers have concluded that tunnelling is instantaneous for atomic hydrogen.

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Quantum-mechanical tunnelling of a particle through a potential barrier is one of those phenomena that has made quantum mechanics famous, partly because the process seems impossible by human 'classical' intuition. Despite being trapped by a potential barrier, impenetrable by means of classical trajectories, a particle 'tunnels' through this barrier and can be found beyond it, albeit with an exponentially small probability. The first (but easier to clarify) issue is the rate with which an initially confined particle tunnels through the barrier; this rate is a statistical measure referring to the wave function of the particle dominated by the Gamow factor, the tunnelling probability, named after the Russian physicist George Gamow. The rate does not represent the time it takes the particle to transit the classically forbidden region under the barrier. This tunnelling time can only be formulated theoretically with additional assumptions and, obviously, the result depends on those assumptions. This has sparked many different more or less meaningful formulations that have been well compiled in a recent review[1]. Confusion often arises because the conclusion is narrowed down to a quantitative statement about tunnelling time — a statement that may be correct under given assumptions and still in contradiction to results obtained under different assumptions.

Hence, experiments cannot measure just tunnelling times, simply because they do not exist as such. Nevertheless, postulating to do so in the pioneering experiment[2] and follow-up experiments[3–5], stretching the limits of measuring time spans to incredibly short intervals of the order of 10 attoseconds (as) has been very fruitful. It has made us think again about the principles of quantum mechanics and what it is we measure. This is, by far, not as straightforward as one might believe.

What is measured in attosecond angular streaking experiments is the direction of the electron that tunnelled out. More specifically, the angle between the maximum of the asymptotic momentum distribution and the 'initial' momentum direction, given by the vector potential of the elliptically polarized laser light at maximal field amplitude during the pulse, is determined. This angular difference $\Delta\alpha$ translates into an ultra-short time difference $\Delta\tau = \Delta\alpha/\omega$ via the laser frequency ω.

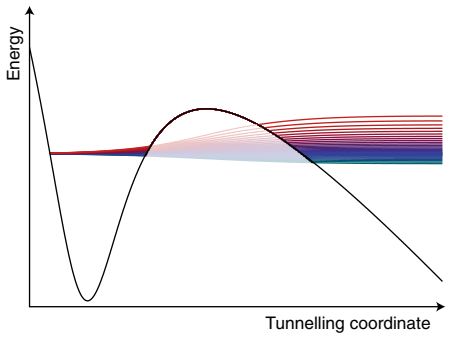

Fig. 1 | Non-adiabatic ionization. The sketch provides a snapshot in time and cut in space through the potential landscape for a bound electron in a strong laser field in a situation of non-adiabaticity. As a consequence, energy is not conserved and the tunnel exit changes depending on the energy of the electron.

Setting aside for the moment what the time difference derived in the way described above has to do with tunnelling, there was uncertainty regarding how to extract the initial electron momentum at maximal laser amplitude. While in the first publications this was done adiabatically (no additional initial momentum but the vector potential)[2], later a non-adiabatic variant was also offered in disagreement with simple theoretical models[5]. However, the explanation to the disagreement is that $\Delta\tau$ was determined in experiments performed on multi-electron systems (rare-gas atoms with helium having the fewest electrons), while theoretical work resorted to approximations and effective one-electron models.

What does one need in such a situation to get on firm ground? An accurate experiment on the simplest system, the hydrogen atom, such that results can be compared to ab initio calculations with no excuse for deviations. This is what U. Satya Sainadh and colleagues from Australia, China, South Korea and the United States have achieved in a joint experimental and theoretical effort with attosecond angular streaking[6]. It is often one of the toughest endeavours to perform accurate experiments on the simplest system. In particular, it is very difficult to produce atomic hydrogen as a target in the ground state and with sufficient density for high-precision experiments such as attosecond angular streaking. However, with the agreement of theory and experiment (within uncertainties), Sainadh and colleagues have provided evidence that measurements of the angular difference have been interpreted correctly, and that the non-relativistic Schrödinger equation can describe the process for the parameter regime of the experiments. Moreover, they draw the reasonable conclusion that tunnelling is instantaneous for hydrogen within the experimental accuracy of 1.8 as.

This leaves us with two questions: Where do the disagreements between theory and experiment in the earlier experiments come from? How should we think about tunnelling times given the outcome of angular streaking experiments?

The first question is retrospectively easier to answer. The electron dynamics with an optical period of 2.67 fs (at 800 nm wavelength) is clearly not fully adiabatic (Fig. 1). Hence, the non-adiabatic calibration of experimental angular streaking is called for. This was already pointed out in ref. [7], but did not receive too much attention in the heated discussion about tunnelling times, which brings us to the second question.

Keeping in mind that there is no unique definition, one needs a reasonable characterization of the tunnel exit that holds for non-adiabatic situations and reduces to the well-known (semi-classical) formulation in a conservative (energy preserving) system. We have proposed a 'minimal condition' that fulfils these criteria, namely that the momentum of the classical Hamiltonian

passes zero in one degree of freedom at the tunnel exit[8]. This acknowledges the notion that pure tunnelling dynamics in a semi-classical description is characterized by an imaginary momentum component in at least one degree of freedom and by the fact that all momenta are real in classically allowed regions.

Using classical backpropagation of an ionized wave packet (determined quantum mechanically by solving the Schrödinger equation), one arrives at the conclusion that the tunnelling time defined with this tunnel exit is zero for single-electron dynamics[9]. The same conclusion has been reached with different techniques[10] and certainly corroborates our 'quantum' intuition that for a single electron under a potential barrier, no (real) time passes.

At least two caveats remain for this intuition: it ignores interference and diffraction phenomena that may complicate determining tunnel exits in a meaningful way, and secondly, it is only valid for a single (or independent) electron ruled by a potential. Substantial interaction with other electrons will imprint an additional phase on the tunnelling electron that may provide a timescale for tunnelling referenced by the remaining electron(s). This renders our earlier statement that experiments cannot measure tunnelling times because they do not exist as premature. Nevertheless, thanks to the beautiful attoclock experiments in helium and now in hydrogen, we can be sure that a reasonably defined tunnelling time is zero in these cases. Yet, the legacy of tunnelling will continue. ❐

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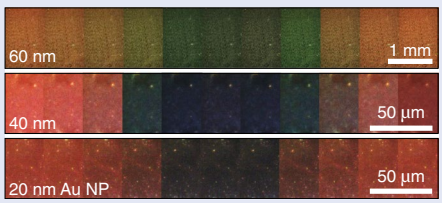
DISPLAYS

Tunable plasmonic pixels

Colour displays based on nanoplasmonics offer some advantages over conventional dye- or structural-based approaches. However, rapid and energy-efficient tuning of colours in plasmonic displays remains a challenge. Now, Jialong Peng, Hyeon-Ho Jeong and colleagues at Cambridge University, UK, have demonstrated a nanoplasmonic display with pixels that show tuning of resonance wavelength (colour), rapidly (>50 Hz) and over a huge 100 nm range (at visible wavelengths) (J. Peng et al., *Sci. Adv.* **5**, eaaw2205; 2019). Importantly, the tuning is energy efficient, with each pixel taking only 0.2 fJ per 1 nm wavelength shift.

Au nanoparticles (NPs) sit on an Au substrate (which is also the working electrode), but with carefully controlled particle–mirror spacing. The particles are electrochromic thanks to encapsulation in a conductive polymer shell (polyaniline); the shell also precisely dictates the particle–mirror gap and is tailored via bottom-up processing. The particles are randomly dispersed onto the planar Au substrate, but coverage can be engineered and the structures are incorporated into custom-built electromechanical cells, enabling tracking of both electrical dynamics and optical response (via dark-field microscopy; see image). The chemical state (and optical properties) of the shells is modified by applied voltage, swept from −0.2 to 0.6 V (at 50 mV s^{-1}). The resulting ~100 nm resonance shifts are not only reversible but the colour and



structures are shown to be stable for at least three months.

The corresponding author, Jeremy Baumberg, told *Nature Photonics* that they had previously been trying to make structural colour materials, using nanoassembly on a large scale. They had some success but realized that making electrically tunable wallpapers was going to require a lot of energy if using bulk structures, so they turned to surface photonics. Earlier work in collaboration with Nokia used holography to make deformable metallic structures like kirigami.

"We can make a scalable coating that allows switching of colour of a film with extremely low energy, which opens up the possibility of building-scale displays," Baumberg explained. "Scientists have been making plasmonic pixels made of noble metals for the last decade, but these all have fixed colours (the idea being to make permanent coloured images that don't fade). But switching in this way is new. Our display also can be viewed from any angle, and in any light conditions (except darkness) since it is a scattering-based colour which doesn't require a backlight."

The work was not without hurdles. Baumberg told *Nature Photonics* that it's a challenge to optimize all of the parameters at the same time. For example, it's not simple to make good blue colours, while keeping costs low, everything flexible, and with good electrical properties, and so on.

"It shows a nice route to making nanophotonics devices that can be scaled onto roll-to-roll processing, which is what I have been trying with a team for a few years," Baumberg stated. "I've been tired of seeing high-cost nanofabrication used for devices with the comment that it could be transferred as generally such high-accuracy nanostructuring is a big roadblock. Using solution growth is really important."

Key to the result, according to Baumberg, was the ability to trap light into tiny gaps, meaning that low energy is required for tuning due to the small volume of material where the optical properties need to be changed. The team is now looking to scale up to larger multipixel demonstrators, as well as to push the colour range. ❐

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