ATTOSECOND SCIENCE

Attoclocks play devil's advocate

An 'attoclock' that measures the relative release time of electrons during double ionization may force us to rethink our use of semi-classical models.

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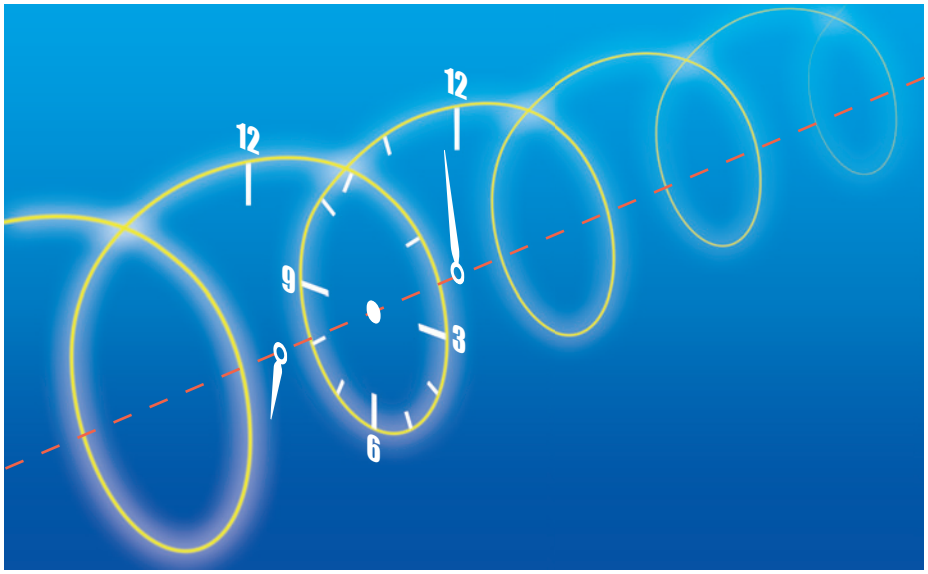

Electrons in an atom move on an attosecond timescale (1 as=10^{-18} s). The recent revolution in ultrafast-laser technology means that capturing this electron motion is no longer a dream, but it still remains a formidable challenge[1]. Under an intense laser field, atoms are ionized when electrons tunnel through the potential-energy barrier created by the nucleus. The released electron wave-packets are one of the most important aspects of attosecond science. But when, precisely, are they emitted? Adrian Pfeiffer and his co-workers now report in *Nature Physics*[2] their measurements on the ionization time of the first and second electrons during double ionization of argon atoms. On exciting the atoms with a strong elliptically polarized laser field, they achieve attosecond resolution using their fantastic hand-made 'attoclock'[3]. The researchers found that the time elapsed between the first and second ionization is significantly shorter than expected from a semi-classical model. It is easy, at first, to say that their model is just too simple. But the implications cannot be ignored: such models seem to be extremely useful, for example, in capturing electronic motion and for ultrafast molecular imaging[4]. So, is it time for a rethink? Or is the attoclock just playing devil's advocate?

To catch atomic motion in molecules as they undergo chemical reactions, we need a camera with femtosecond (10^{-15} s) shutter speed. This can be realized by pump–probe experiments using lasers with femtosecond pulse duration: a pump pulse triggers the reaction and a delayed probe pulse slices its evolution. This femtosecond pump–probe technique opened the door to a new field called femtochemistry[5] and has been widely used to investigate many ultrafast chemical reactions. Similarly, to catch atomic-scale electron motion, one may consider attosecond pump–probes. With current laser technology, it is possible to make sub-100-as pulses[6] in the extreme-ultraviolet (EUV) wavelength region. However, attosecond pump–probe experiments have not yet been achieved due to the low intensities of these pulses. Instead, infrared-femtosecond-laser-pump–attosecond-EUV-probe experiments[7], or attosecond metrology[8], have successfully probed ultrafast electron dynamics such as real-time electron motion[7] and time delay in photoionization[8].

The principle of the 'attoclock' now used by Pfeiffer *et al.*[2] is completely different from the above-described two-colour pump–probe schemes. They use circularly polarized light with a wavelength of about 740 nm. The electric-field vector of such light rotates one turn (360 degrees) in about 2.5 fs. This acts like the minute hand of the clock, providing sub-femtosecond temporal resolution. The amplitude of the electric-field vector varies within the pulse envelope and plays the role of the hour hand of the clock. To measure the direction and the amplitude of the electric-field vector, the team use strong-field ionization and detect photo-ionized electrons deflected by the electric-field vector. Strictly speaking, their ultrafast laser provides close-to-circularly polarized pulses due to technical reasons but compensation for the non-completeness of the circular polarization is trivial.

With this attoclock at hand, Pfeiffer *et al.* measured the ionization times of the first and second electrons ejected by strong-field double ionization of atomic argon[2]. We should note that it is their sophisticated detection scheme that made this ground-breaking experiment possible. They recorded the three-dimensional momenta of the two emitted electrons and the corresponding residual ion recoil by coincidence. Pfeiffer *et al.* used their experimental results to critically test the current understanding of strong-field double ionization. The results, it turns out, are likely to be controversial.

Usually, double ionization is classified into two categories, sequential and non-sequential. Sequential double ionization views the process as two separate independent steps: first, tunnel ionization creates a ground-state ion. Then tunnel ionization out of the ground-state ion takes place. This approximation is often called a single-active-electron approximation. However, double ionization by linearly polarized laser light is dominated by the recollision of the electron ionized in the first step as it is driven back to the ion by the laser field itself. This recollision is known to be the dominant non-sequential ionization mechanism. By using close-to-circularly polarized light, this recollision

mechanism is switched off, and we therefore usually consider that double ionization can be well described by a sequential model based on the single-active-electron approximation[9]. What Pfeiffer *et al.* found was that the second ionization step takes place much earlier than predicted by the sequential double-ionization model.

What does this really tell us? One might criticize the approximations used in the semi-classical model. First, how can we define the timing of the electron release? It is probably impossible to answer this philosophical question rigorously. The authors trace back the classical electron trajectory and determine the time when the trajectory had zero velocity. This fully classical definition is surely debatable but any alternative would not shift the timing by more than a half cycle. Second, is neglecting the Coulomb potential justified? It has recently been shown[10] that the angular distribution of the electron released by elliptically polarized laser fields is sensitive to the details of the electron-ion interaction, which would cast doubt on the validity of the minute hand of the attoclock. According to Pfeiffer *et al.*, the Coulomb correction is small in the intensity regime they studied and cannot explain the observed timing difference.

Third, is the theory used to calculate the tunnelling rate reliable? Fourth, is there any multi-electron effect during the ionization? Ideally, to answer these questions we would use time-dependent Schrödinger equation (TDSE) simulations. However, TDSE simulations for a multi-electron atom in an elliptically polarized strong field are currently not feasible due to exceedingly large computing time requirements. Very recently, Wang and Eberly[11] studied sequential double ionization with elliptical polarization using a classical-ensemble simulation that fully takes into account electron–electron interactions at all times. Such simulations might shed light on the multi-electron effects, though it is not clear how quantitatively the classical-ensemble model can describe strong-field ionization. Also, there remains uncertainty in the intensity and temporal profile of the pulses used in the experiments. The exploration of these issues will be an exciting prospect, and possibly at the same time sow the seeds of even more controversy. ❐

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THE PHYSICS OF BLOOMING

Watch it unfold

Their genesis is described with flowery eloquence in the *Geoponica*, a collection of twenty books of agricultural lore compiled during the tenth century: as the goddess Hera was suckling Heracles, drops of milk fell onto the ground and grew into lilies (the milk spilt into the sky became the Milky Way). Over the centuries, lilies — and in particular their blooms — have held mystical appeal, as a symbol of purity or rendered emblematically as the *fleur de lys* in coats of arms and flags. They've received less attention, however, from science, even if the mechanisms of how lilies and other flowers bloom, and thus how they reveal the bright colours of their petals and adopt their characteristic shape, are not fully understood.

Haiyi Liang and L. Mahadevan have taken a close look at the physical process of blooming and present a compact theory for the movements involved (*Proc. Natl Acad. Sci. USA* **108,** 5516–5521; 2011). The metamorphosis from bud to flower takes, depending on the flower, somewhere between a few hours and several days, suggesting that the process is driven by growth, rather than, for example, by flow of water.

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For the common lily *Lilium Casablanca* (pictured), which Liang and Mahadevan have studied, the process typically spans four and a half days. During that time, pressure builds up inside the bud, as the three inner petals grow inside the three outer sepals that embrace them. A locking mechanism between petals and sepals ensures that the bud remains intact during this period, but once a critical pressure is reached, the flower blooms relatively rapidly, as petals and sepals reverse their curvature and at the same time wrinkle around the edges.

These wrinkles hint at differential growth being part of the blooming process. It had been proposed that the relevant difference in growth rate is between the upper and lower sides of the petals and sepals, and that the midrib has an important role. But by shaving the midrib off petals, Liang and Mahadevan proved that it is not necessary for blooming. Furthermore, they find in their observations and through modelling that growth at the edges alone can induce the shape change.

So it's all about forces and stresses then, rather than mystique and elegance? Not quite, say Liang and Mahadevan. They see their study as "infusing a scientific aesthetic into a thing of beauty", but also expect, more pragmatically, that these findings could inspire new designs for artificial edge-activated bimorphs.

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