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PHOTOEMISSION DELAY

White Rabbit's clock

Without a very precise timer one can never catch up with the electron released in photoemission. Attosecond streaking spectroscopy allows to set such a chronometer clock to zero and reveals the role of electron correlations.

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Photoemission is not an instantaneous process. Following the absorption of a sufficiently high-energy photon, the electron can leave the atom only after a few tens of attoseconds[1]. Why does the electron hesitate before leaving the atom? The first reason is the interaction of the ionized electronic wave packet with the ionic potential. This can be seen as a half-scattering mechanism leading to a phase shift of the wave packet, which ultimately translates to a time delay[2]. The second reason is the influence of the remaining electrons—the so-called electronic correlation. Except for the hydrogen atom— the only one-electron system — for all other quantum mechanical systems photoemission must therefore be modelled as a many-body process. In a way we could think that the action of the fellow electrons on the ionized one is not negligible. And since quantum effects are never obvious, this action can result in either the retardation or advance of the electronic wave packet.

Recall the White Rabbit in *Alice in Wonderland*: constantly checking his clock in a race against time. Similarly, in photoemission delay experiments, without a sufficiently precise clock we can never be in time to catch the released electron. Now writing in *Nature Physics*, Marcus Ossiander and co-workers[3] show how to set the absolute zero time in photoemission with sub-attosecond accuracy, and reveal the presence of a correlation delay in helium, from which crucial information about the ionized system can be obtained.

The presence of only two electrons makes the helium atom the ideal test system for studying electron correlations. When the helium atom absorbs photons with the energy above 24.6 eV, one electron is released with kinetic energy corresponding to the difference between the photon energy and the ionization potential of the atom (direct photoemission). However, if the photon energy exceeds 65.5 eV, the electron emission can be accompanied by the excitation of the remaining electron into ionic states with higher energy (shake-up) or even a second ionization event (shake-off). Shake-up (or shake-off) lends itself well to investigate the correlation delay thanks to the presence of a strong electron-electron interaction.

The best technique for measuring the relative timing of electron photoemission is the attosecond streak camera[4]. An infrared laser pulse, or streaking pulse, with a well-

controlled electric-field waveform is used to impart substantial momentum to the electrons released by an XUV attosecond pulse. Thanks to the correspondence between the liberated electrons and a specific phase of the streaking field, it is possible to map the instant of release onto a change of the electron kinetic energy. This approach does not provide information on the absolute time of photoemission, but it can be used to clock the relative release time between electrons originating from different atomic shells[1] and bands in solids[5], or the time delay in photoemission compared to a well-known reference[6]. The presence of an external laser field leads to a substantial apparent time delay[7], which must also be taken into account.

The accuracy of the streak camera technique relies on the retrieval approach used to reconstruct the emitted electronic wave packet. *Ossiander et al* make a significant improvement in this direction: their data analysis routine, based on analytic fitting of the strong-field solution of the time-dependent Schrödinger equation, enables an unprecedented time resolution. This makes it possible to retrieve time delays with sub-attosecond accuracy, which in turn enables the precise determination of photoemission time. But the measurement itself does not provide the absolute zero time to be set on the clock. To this end, the authors need a quantum mechanical *ab-initio* calculation. It is the sub-attosecond agreement between the calculated and experimentally retrieved values that finally allows the absolute zero time to be set.

This achievement is accompanied by another important result: among the different time delay contributions, with the help of theory one can identify a correlation delay in the shake-up channel. This results from the back-action of the excited bound state onto the released electron wave packet in the presence of the streaking field[8]. So it is possible to extract information about the ionized system by interrogating the entangled outgoing electron. This incredibly short correlation time delay—just 6 attoseconds—is indeed governed by the charge asymmetry (effective dipole) induced by the shake-up ionization process. Thus, a precise retrieval of this correlation delay allows for the reconstruction of the effective dipole moment for the correlated shake-up wave packet.

Looking ahead, one could imagine that the precise determination of correlation delays will allow measuring the dynamics of the ionized system in a non-destructive way. But it remains to be seen if theory will be able to address with the same accuracy more complex atoms than helium, or even molecules.

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