Attosecond Larmor Clock

Olga Smirnova¹, Jivesh Kaushal¹

¹Max Born Institute, Max Born Strasse 2a, 12489 Berlin, Germany jivesh.kaushal@mbi-berlin.de

We present a new clocking mechanism to time strong field ionisation processes. Using our recently developed analytical R-matrix (ARM) for circularly polarised fields [1], which is adopted from the R-matrix approach used in studying collision processes and nuclear resonance reactions [2], we can consistently include effects of arbitrary, long range potentials during ionisation and thus consider non-adiabatic dynamics. One application within this new scheme is the ability to time multiphoton ionisation process on attosecond scale using the spin-orbit interaction of the ionising electron with the core as the clock.

We consider multiphoton ionisation from 4*p* level in Kr atom. Experimentally, first a few femtosecond, infrared (right) circularly polarised field ionises the electron in the spin-orbital split levels, leaving behind a hole in a superposition of ${}^{2}P_{3/2}$ and ${}^{2}P_{1/2}$ state. Second, an attosecond, (left) circularly polarised XUV pulse, delayed w.r.t. the fs-pump pulse, excites the ion into an *s*-state, at which point the spin-orbital interaction is switched off (l = 0). This way we also impart a "start" and "stop" mechanism to our attosecond Larmor attoclock. Finally, the resulting ion signal can be measured by transient absorption spectroscopy [3].



Fig. 1: Ionisation time calculated from spin-orbit interaction for intensity 1.2×10^{14} W/cm².

Fig. 1 shows the resulting Wigner-Smith (WS) ionisation times vs. the number of absorbed photons at optimal momentum. We find that this WS time is related to the Larmor time. The WS time for multi-photon ionisation (blue) can be divided into two parts: 1) A WS time approaching the one-photon ionisation case in the limit $n \rightarrow 1$ (red) and 2) A clock-induced delay (green), due to the entanglement of the electron and hole wavepacket, leading to an additional phase dependence of $1/r^3$ -type which can either compress or stretch the hole wavepacket in the ion. This additional phase delay due to electron-hole interaction is the main difference from one-photon ionisation.

References:

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[2] P. G. Burke, *R-Matrix Theory of Atomic Collisions*, Springer Series on Atomic, Optical and Plasma Physics Vol. **61**, 2011.

[3] E. Goulielmakis et. al., "Real time observation of valence electron motion", Nature 466, 739 (2010).