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 4p 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 \(^2P_{3/2}\) and \(^2P_{1/2}\) 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 (\(I = 0\)). 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 \(1.2 \times 10^{14} \text{ W/cm}^2\).

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 \(n \to 1\) (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: