An analytic R-matrix approach to strong field ionization: Coulomb and correlation effects

Lisa Torlina[1], Misha Ivanov[1,2,3], Jivesh Kaushal[1], and Olga Smirnova[1]

[1]*Max Born Institute, Max Born Strasse 2a, 12489 Berlin, Germany*
[2]*Department of Physics, Humboldt University, Newtonstrasse 15, 12489 Berlin, Germany*
[1]*Department of Physics, Imperial College London, SW7 2AZ London, United Kingdom*
torlina@mbi-berlin.de

One of the most powerful ideas for describing scattering and ionization in multielectron systems is embedded in the R-matrix method [1], where space is divided into inner and outer regions, each associated with different dynamics. Its power comes from our ability to treat the two regions separately and make appropriate simplifications in each. While already well-established in the numerical community, we show that this approach can also offer distinct advantages when it comes to analytical or semianalytical methods. We develop a flexible analytical theory of strong-field dynamics in atoms and molecules: the analytical R-matrix method (ARM). Using this theory, we investigate 1. effects on ionization rates and electron spectra due to the potential of the core, and 2. excitations of the core during ionization due to electron-electron correlation.

The effects of electron-core interaction during strong field ionization are typically treated in the adiabatic approximation or using mixed quantum-classical models. Here, we develop a fully self consistent quantum approach, valid well into the non-adiabatic regime, which naturally includes the concept of trajectories. However, the trajectories are quantum: they include an imaginary component in general. Our theory provides insight into the dynamics of electron-core interaction in the laser field on a subcycle timescale. Focussing on circular fields, we study how ionization rate and photoelectron spectra build up over time, and find indications of electron trapping. In electron spectra, we also observe an overall shift towards low momenta, which we associate with a deceleration of the electron by the core. These non-adiabatic effects come about as a direct consequence of the imaginary part of the electron's trajectory.

Recent years have seen compelling experimental evidence that strong field ionization can generate ions in excited states, for example see [2-4]. In the standard adiabatic picture, it is typically assumed that only one electron is active while the others remain frozen in the ion. In this case, the only way to produce an excited ion is to remove an electron from a lower lying molecular orbital. The higher ionization potential implies that such direct channels are exponentially suppressed in general. However, we know that the ionization of multielectron atoms and molecules inherently involves interaction between all electrons. These interactions, in turn, can also give rise to excitations of the ion. We extend our theory beyond the single active electron approximation to take such correlation-driven channels into account. Applying our analysis to N_2 and CO_2 in linear fields, we show that coupling between the departing and core electrons can help remove the exponential penalty associated with the direct channel in certain cases. We expect these channels to play an important role in high harmonic experiments at 800nm.

References:

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