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Study of Cooper Minimum in Argon by High order harmonic spectroscopy

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Over the past few years, it was recognized that high order harmonic generation (HHG) process in aligned molecules can encode molecular structure. In particular, many works have focused on investigating structural minima appearing in harmonic spectra. The studies have raised important question on the modelling of HHG such as the influence of the ionic potential on the recolliding electron, the contribution of multiple orbitals, the influence of the strong laser field and the role of propagation effects. Since HHG from molecule is quite complex to model, we use a more convenient test bench of HHG models by investigating the simpler case of high order harmonic emission from argon atoms which exhibits a local minimum associated to the Cooper minimum [1] observed in XUV photoionization of argon.

We have then studied experimentally the Cooper minimum in high order harmonic emission from argon using tunable infrared (1800-2000 nm) femtosecond laser pulses. We have performed a systematic experimental study of the position of the minimum as a function of the laser field (intensity and wavelength) and macroscopic parameters (gas pressure, beam focusing conditions). We find a systematic shift of more that 5 eV in the position of the minimum with respect to total photoionization cross section measurements [2, 3].

In order to extract physical insight of the process we use a semi-classical simulation based on a combination of Classical Trajectory Monte-Carlo (CTMC) [4] and Quantum Electron Scattering techniques (QUEST). As compared to TDSE calculations, and similarly to Strong Field Approximation (SFA) models, it allows to factorize the process in a product of the flux of returning electrons and the photorecombination cross section [5,6] and to easily separate the contribution from short and long trajectories. But in contrast to most SFA approaches, this new theoretical description (CTMC-QUEST) properly accounts for the influence of the ionic potential on the recolliding electron wavepacket and is thus able to reproduce accurately the experimental high harmonic spectra [7].

We therefore identify two contributions to the shift A first contribution arises from the difference between angle-integrated photoionization measurements with unpolarized light and the HHG recombination process which is highly differential with respect to both electron and polarization directions. More importantly, we show that the additional contribution to the shift is due to the shape of the recolliding electron wavepacket. Taking advantage of the versatility and ease of implementation of the CTMC-QUEST approach, we are now working to its extension to elliptically polarized laser field or complex molecules, in the perspective of achieving a complete 3D description of the process.

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