Attosecond time delays in the photoionization of noble gas atoms studied in TDLDA

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Synopsis We perform time-dependent local density functional calculations of the quantum phase and time delays of valence photoionization of noble gas atoms. Results may be accessed by XUV-IR interferometric metrology.

In the standard RABITT (reconstruction of attosecond beating by interference of two-photon transitions) metrology, photoelectrons emitted by odd harmonics of an XUV pulse comb subsequently absorb or emit an IR photon, producing even-harmonic sidebands in the spectrogram. The ionization time delay is then determined by the ratio of the phase difference to the energy difference between these sidebands, directly connecting to the Wigner-Smith approach of the delay extraction from the emission quantum phase.

We compute these quantum phases and resulting Wigner-Smith time delays in the photoionization of valence electrons of various noble gas atoms, using a scheme of time-dependent local density approximation (TDLDA) [1] with Leeuwen and Baerends exchange-correlation functional. Analysis of the results with the goal to temporally access details of electron correlations has been carried out [2].

Numerical results for the 3*p* time delay of Ar (green curve) and the relative delay between 4*s* and 4*p* for Kr (red curve) are shown in Figure 1. For Ar, in addition, the calculations are compared with recent measurements [3] showing very good agreements. The minimum seen immediately above 45 eV is due to an accelerated ejection of 3*p* photoelectrons near the 3*p* Cooper minimum, which only exists in 3*p*→k*d* ionization channel. However, the energy dependence of this delay becomes softer due to the participation of the 3*p*→k*s* channel.

TDLDA 4*s*–4*p* relative delay for Kr exhibits a strong shape at lower energies centering 40 eV. This structure is due to the 4*s* Cooper minimum of Kr. In contrast, its 4*p* Cooper minimum at 67 eV generates a far weaker structure. This suggests the ability of the 3*s* minimum to affect the time behavior in a much stronger way than that of 3*p*.

These results suggest significant differences in the details of correlations in the valence photoionization among various noble gas systems.

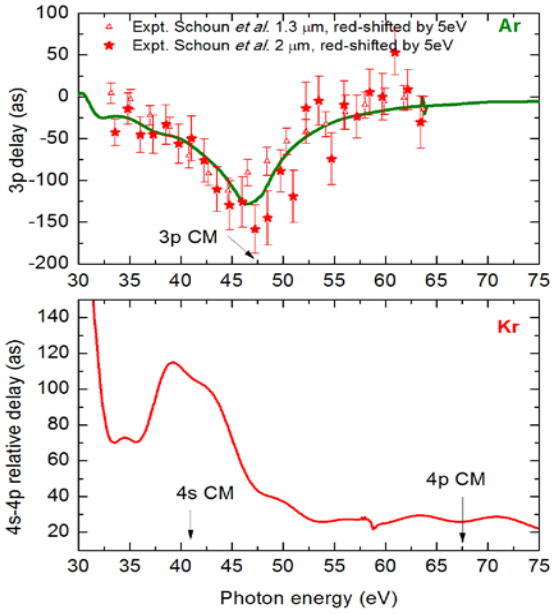

Figure 1. TDLDA Wigner-Smith time delay for Ar 3*p* and relative delay (4*s*–4*p*) for Kr valence electrons are shown. For Ar, the result is compared with the measured data [3].

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References

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