Observation of strong non-dipole effects in sequential multi-photon ionization using VUV FEL radiation

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One of the fundamental principles of light-matter interaction is the dipole approximation that is commonly assumed to be valid at low photon energies including the soft X-ray region. Although it has been shown at synchrotrons that this approximation has limitations, it has been unclear how they impact the ionization characteristics of ions. This is particularly interesting because of their abundance in outer space, however experimentally challenging to study.

Recently, ultrafast and ultra-intense Vacuum UltraViolet (VUV) pulses from free-electron lasers (FELs) have been used to efficiently study photoionization properties of noble gas ions by determining their photoelectron angular distributions [1]. In that scheme, singly charged ions that are created with an FEL pulse are further ionized within the very same light pulse that has a total duration of typically few to few hundreds of femtoseconds. We present evidence of a forward-backward symmetry breakdown in such a sequential ionization process in the vicinity of the Cooper Minimum of argon at 46 eV photon energy, measured at the VUV FEL FERMI in Italy. Our experimental setup consists of 16 independently working electron time-of flight detectors which are arranged in a plane with 38° rotation angle to the light propagation direction. While covering a photon energy range from 30.6 up to 66.6 eV, we present electron angular distributions of the first, second and even the third step of the sequential ionization process. In particular, we show that the electron angular distribution of ionic argon gains a pronounced forward-backward asymmetry with respect to the beam by interference between the electric dipole and the electric quadrupole amplitudes [2,3]. According to our preliminary experimental findings (see figure 1), the strength of this effect exceed the original theoretical predictions [2].

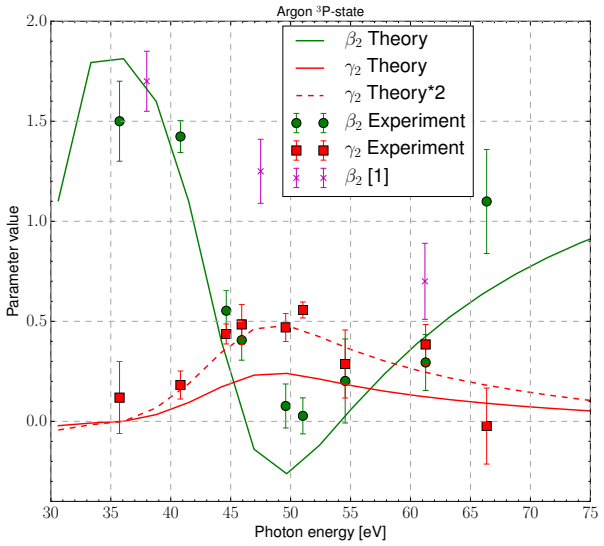

Figure 1. Preliminary results for the dipole asymmetry parameter β_2 (green) and the non-dipole asymmetry parameter γ_2 (red) as a function of photon energy for ionization of $Ar^+(3p^{-1})$ into $Ar^{2+}(3p^{-2})^3P$ compared with results of [1] and simulations [2]. While the experimental values of β_2 are in good agreement with theoretical predictions [2], γ_2 seems to be a factor of two larger than predicted (dashed red line).

References

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