# Symmetry Breaking in the Sequential Photoionization of Argon

**M. Ilchen**[1,*1], **G. Hartmann**[2,3], **A. N. Grum-Grzhimailo**[1,4*2], **E. V. Gryzlova**[4], **A. Achner**[1], **A. Beckmann**[1], **J. Buck**[1,3], **C. Callegari**[5], **R. Cucini**[5], **A. De Fanis**[1], **E. Ferrari**[5], **P. Finetti**[5], **L. Glaser**[3], **A. Knie**[2], **A. O. Lindahl**[6], **O. Plekan**[3], **T. Mazza**[1], **E. Roussel**[5], **F. Scholz**[3], **I. Shevchuk**[3], **J. Seltmann**[3], **P. Walter**[3,6], **J. Viefhaus**[3], **and M. Meyer**[1]

[1] European XFEL GmbH, Holzkoppel 4, 22869 Schenefeld Hamburg, Germany

[2] University of Kassel, Institut für Physik, Heinrich-Plett-Straße 40, 34132 Kassel, Germany

[3]Deutsches Elektronen-Synchrotron (DESY), Notkestrasse 85, 22603 Hamburg, Germany

[4]Skobeltsyn Institute of Nuclear Physics, Lomonosov Moscow State University, Moscow 119991, Russia

[5]Elettra-Sincrotrone Trieste, I-34149 Basovizza, Italy

[6]SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, California 94025, USA

**Synopsis** A strong non-dipole effect in the vicinity of the Cooper minimum of argon ions has been investigated via sequential ionization by VUV FEL pulses of FERMI, Italy.

The basic principles of light matter interaction are continuously challenged by novel insights into ultrafast and non-linear processes. One of these fundamental principles 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 synchrotron radiation facilities that this approximation has limitations [1], it has been unclear how these non-dipole effects impact the characteristics of non-linear processes. Recently, ultrafast and ultraintense Vacuum UltraViolet (VUV) pulses from free-electron lasers (FELs) have been used to efficiently study the photoelectron emission characteristics of noble gas ions by determining their electron angular distributions [2]. 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. Besides the detailed study of non-linear processes in light-matter interaction, the scheme of sequential photoionization enables the efficient production and spectroscopic exploration of ionic species. Here, we present experimental evidence of a symmetry breakdown in such a sequential ionization process in the vicinity of the Cooper minimum of singly charged argon around 50 eV photon energy, measured at the VUV FEL FERMI in Italy (see Fig. 1). The angular distribution parameters $\beta_2$, $\beta_4$, and $\gamma_2$ have been determined for all final states of the second ionization step, i.e. $Ar^{2+}(3p^4)$ - $^1S$, $^1D$, $^3P$. The results are in agreement with and extend previous experimental studies [2] and reveal a pronounced non-dipole effect with very good statistical accuracy ($\gamma_2$-values). These are in good qualitative agreement with theoretical predictions [3, 4], but also indicate an underestimation of the effect in these calculations. Our new calculations, shown in Fig. 1, with optimized electron orbitals, are in better agreement with the experiment.

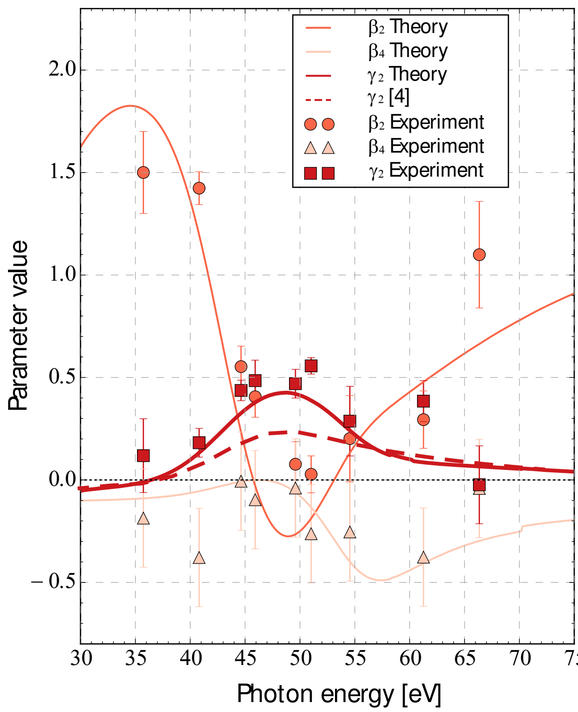


**Figure 1**. Preliminary results of the angular distribution parameters of electrons emitted from $Ar^+(3p^5)$ to $Ar^{2+}(3p^4)^3P$.

## References

[1] D. Lindle, *et al.*, *J. Electr. Spectr. Relat. Phenom.* **100**, 297 (1999).

[2] M. Braune, *et al.*, *J. Mod. Opt.* **63:4**, 324 (2016).

[3] E. V. Gryzlova *et al.*, *J. Phys. B* **46**, 164014 (2013).

[4] E. V. Gryzlova *et al.*, *J. Phys. B* **47**, 195601 (2014).

[1]E-mail: markus.ilchen@xfel.eu

[2]E-mail: grum@sinp.msu.ru