Multiphoton Ionization and Excitation of Atoms, Molecules, and Fullerenes

M. Braune, R. Hentges, A. Reinköster, D. Rolles[1], J. Viefhaus, and U. Becker

Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany
[1]Lawrence Berkeley National Laboratory, 1 Cyclotron Road, MS 4R0230, Berkeley, CA 94720, USA

The experiments were done at the new light-source FLASH (**F**ree-**E**lectron **LAS**er in **H**amburg) – formerly VUV-FEL – at the beamlines PG2 and BL2 with a two chamber setup. With this setup we performed angle resolved photoelectron spectroscopy measurements on various rare gases (He, Ne, Ar, Kr, Xe) as well as on the small molecules H_2, N_2, O_2, and CO, targets with well known cross sections [1]. Exemplarily we report here on results of Ne and Ar. Furthermore the ionization and fragmentation of C_{60} was studied.
Photoelectron spectra have been recorded at the beamline BL2 with a photon energy of about 38 eV. Results are shown for Ar in Fig. 1. The FEL delivered trains of 30 bunches with a repetition rate of 5 Hz. Bunches were separated by 1 μs and the single pulse length was 20–50 fs.

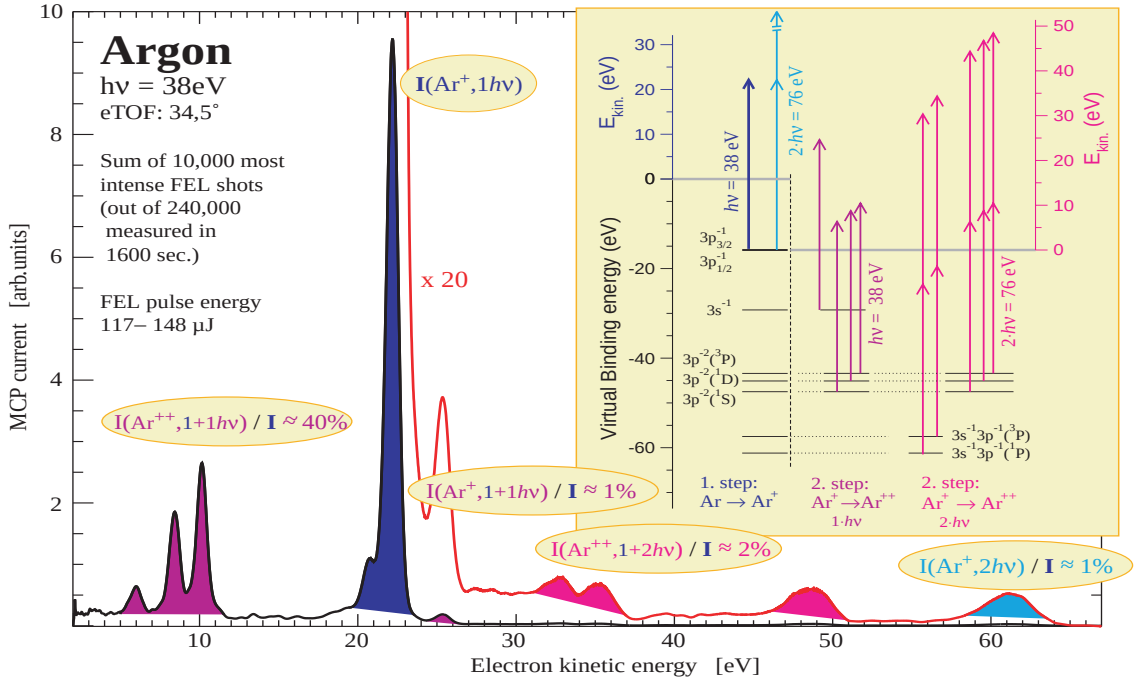

Figure 1: Photoelectron spectrum of argon recorded at the FEL. The inset indicates the observed transitions.

Besides the main single photoionization lines we recorded additional signals which we address to second step double ionization within the same FEL-pulse. This would mean that FEL pulses are able to create an ionic target of considerable density within the pulse duration. The kinetic energies of the electrons of most of these signals refer to second step ionization by a second fundamental wave photon. But especially for Ne and Ar there are as well some signals with higher kinetic energies which match double ionization energies by second harmonic radiation. The fact that these signals are rather strong (the 2nd harmonic radiation fraction should be less than 1%) and analog signals of third harmonic of similar strength were not observed may be a indication of two-photon absorption in the second ionization step.
A way to distinguish two-photon processes from processes due to 2nd harmonic radiation is to analyze the angular distribution of the outgoing electrons, since the angular patterns should differ from

a distribution of a dipole transition with momentum transfer of Δl=1 being governed by the well-known β-distribution (a 2nd order Legendre polynomial and the β-parameter) [1, 2]. Contributions from successive ionization with one- and two-photon processes are found:

$$\mathrm{Ne} \xrightarrow{1\cdot h\nu} \mathrm{Ne}^{+} \xrightarrow{2\cdot h\nu} \mathrm{Ne}^{2+} \quad ,$$

$$\mathrm{Ar} \xrightarrow{1\cdot h\nu} \mathrm{Ar}^{+} \xrightarrow{1\cdot h\nu} \mathrm{Ar}^{2+} \quad .$$

The corresponding angular distributions are shown in Fig. 2 for neon and argon.

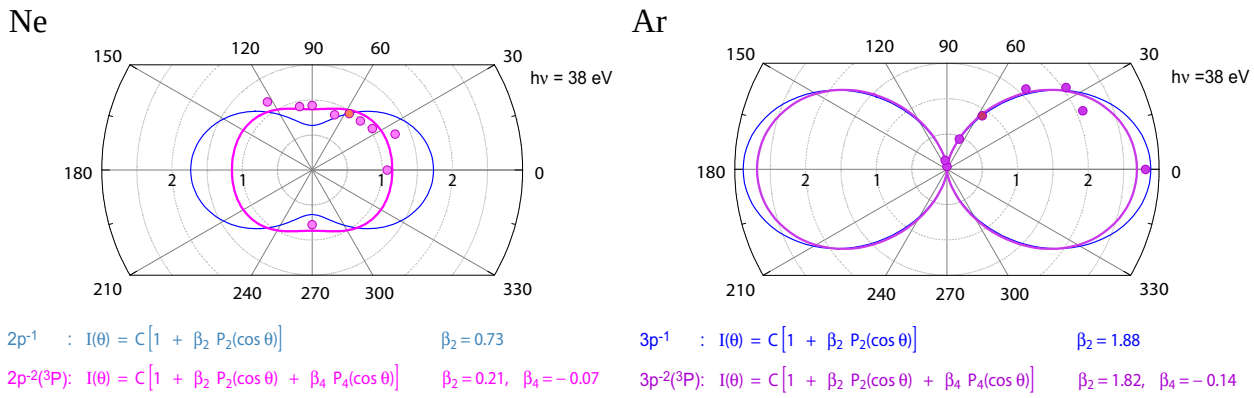

Figure 2: Photoelectron angular distribution for neon (left side) and argon (right side).

In June 2006 in experiments at the beamline BW3 additional measurements of N_2 core ionization were performed. These measurements concluded our studies of coherence of core electrons in homonuclear molecules [2]. As a final achievement, an improved electron–N^+-ion coincident angular distribution pattern could be determined containing experimental results of various beamtimes at BW3 over the last years, which is shown in Fig. 3. For the key instrument of these measurements – the position sensitive ion time-of-flight spectrometer – a new analysis tool for online visualization of angular distribution patterns was tested during the last beamtime.

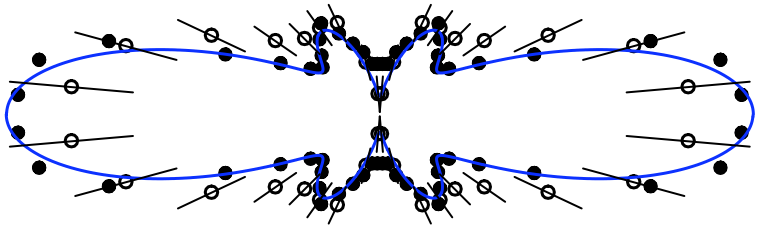

Figure 3: Coincident angular distribution pattern of N(1s) photoelectrons and N^+ ions at a photon energy of 419 eV. Open circles: our data, solid circles: results of the group of R. Dörner (*private communication*), blue line: calculations of B. Zimmermann (*private communication*).

References

[1] U. Becker and D.A. Shirley, *VUV and Soft X-Ray Photoionization*, Plenum Press, New York, 1996, Chapter V: Partial Cross Sections and Angular Distributions, p. 135–180.

[2] D. Rolles, M. Braune, S. Cvejanović, O. Geßner, R. Hentges, S. Korica, B. Langer, T. Lischke, G. Prümper, A. Reinköster, J. Viefhaus, B. Zimmermann, V. McKoy, and U. Becker, Nature 437, 711 (2005). —, Rad. Phys. Chem. 75, 1514 (2006).