Molecular-Frame Photoelectron Angular Distributions of Halomethane Molecules at High Photoelectron Kinetic Energies.

C. Bomme[1], D. Anielski[1,2], R. Boll[1,2], S. Bari[3], E. Savelyev[1], B. Erk[1], J. Viefhaus[1], and D. Rolles[1]

[1]DESY, 22607 Hamburg, Germany
[2]Max-Planck-Institut für Kernphysik, 69120 Heidelberg, Germany
[3]European XFEL GmbH, 22761 Hamburg, Germany

Molecular-frame photoelectron angular distributions (MFPADs) provide deep insights into the molecular photoionization process that give access to an unprecedented level of detailed information such as phases of photoelectron waves [1, 2], localization of core holes [3], and double-slit interference [4, 5]. When interpreting MFPADs in terms of photoelectron diffraction [6, 7], direct information on the geometric and electronic structure of the molecule can be obtained, e.g., by comparing the measured diffraction patterns and MFPADs to single and multiple scattering calculations [8, 10, 11, 12].

Using circularly polarized light from the P04 beamline at PETRA III, we have determined the MFPADs of fluorine *1s* and iodine *3d* photoelectrons from CH_3F, CH_3I and CF_3I molecules by means of angle-resolved photoelectron-photoion coincidence spectroscopy.

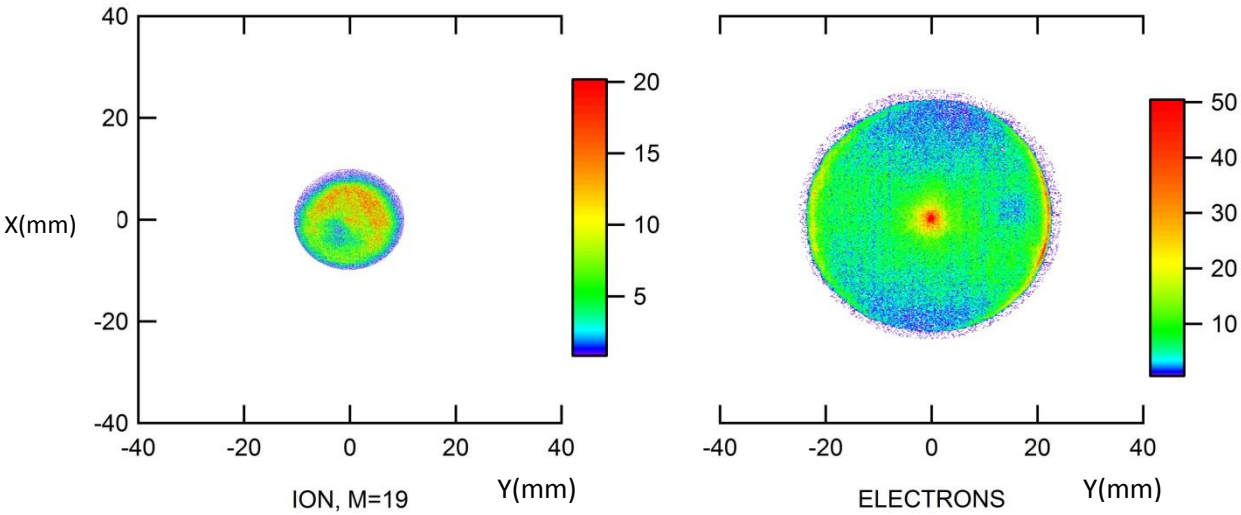

Figure 1: Two-dimensional momentum images of singly-charged fluorine ions (left) and 50 eV F(*1s*) photoelectron(right) measured in coincidence after ionization of CH_3F at 745eV photon energy.

With our double-sided velocity map imaging spectrometer, which is optimized for electron-ion coincidence measurements of high-kinetic energy electrons, we are able to determined MFPADs for photoelectrons up to 300 eV kinetic energy. Exemplary ion and photoelectron images are shown in Fig. 1.

Our measurements extend existing experimental data to higher photoelectron kinetic energies in order to allow a more systematic comparison to theory. Fig. 2 shows three-dimensional MFPADs obtained from the images shown in Fig.1 using the F function method[8,9,13] for parallel and perpendicular orientation between the light propagation direction and the main molecular axis. Here, the direction of the hydrogen atoms was not resolved, thus only the direction of the F-C axis is fixed in these MFPADs.

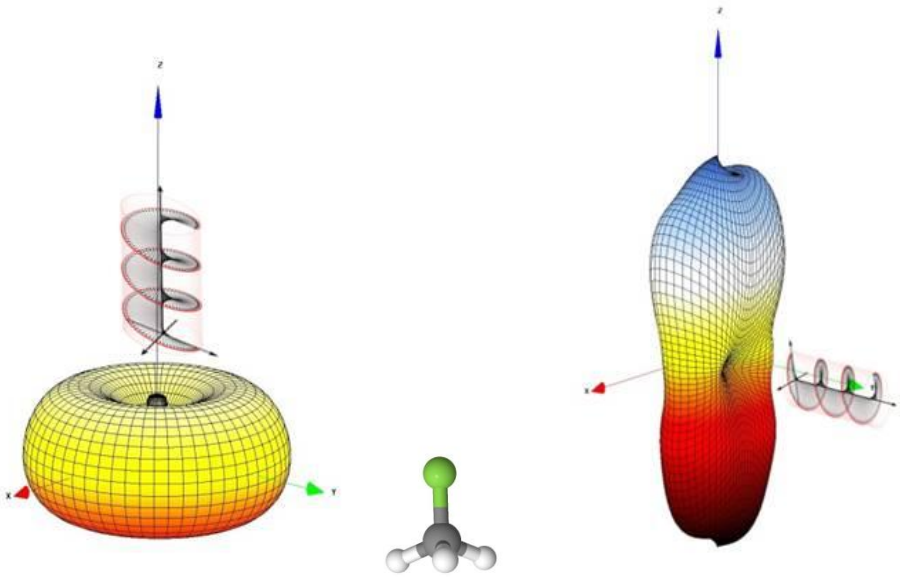

Figure 2: 3D MFPADs after F(*1s*) photoionization of CH_3F at 745eV photon energy. Light propagation axis and molecular F-C axis are parallel (left plot) and perpendicular (right).

For these high kinetic energies of 50eV and above, the MFPADs are dominated by diffraction effects that encode information on the molecular geometry in the MFPADs. In order to illustrate this in more detail, we are now in the process of comparing our experimental results to single and multiple scattering calculations similar to those used in earlier studies [11, 12] and to DFT calculations made by P. Decleva and M. Stener from Trieste University [13,14].

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