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Core-Shell and Valence-Shell Photoionization of Argon Clusters

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Synopsis Core- and valence-shell photoionization of variable size argon clusters were investigated using angle-resolved time-of-flight photoelectron spectroscopy. The Ar 3p outer-valence and Ar 3s inner-valence photoelectron spectra were measured as two formats: individual spectra and two-dimensional maps. The Ar 2p core-shell photoelectron spectra were measured only in individual spectra format. The photoelectron angular distribution parameters of Ar 3p, 3s, and 2p clusters as a function of photon energy and cluster size were determined. In the case of the 2p-shell, measurements were also spin-orbit resolved. Argon cluster with cluster sizes <N>=70 and 210-250 were studied.

Clusters have been studied for several decades in various areas of physics and chemistry, mainly because they are seen as a connection between single atoms and solids, or, in other words, the microscopic and the macroscopic world. Due to their special, closed-shell electronic structure, rare-gas clusters, in particular, have been intensively investigated.

We have performed systematic studies of the photoionization of rare gas clusters and especially their photoelectron angular distributions. We will report on the photoionzation of argon clusters with different average cluster sizes. The measurements were carried out at the Advanced Light Source (ALS) of the Lawrence Berkeley National Laboratory, using synchrotron radiation from beam line 10.0.1. Two-dimensional time-of-flight electron spectroscopy was used (more details are described in references [1-4]). A supersonic jet was used to produce the argon clusters. Argon gas was expanded through a 100µm diameter nozzle at different stagnation pressures and different nozzle temperatures. Based on empirical scaling laws, the average cluster sizes were <N>= 70 and 250 for Ar 2p, <N>= 210 for Ar 3p, and <N>=230 for Ar 3s.

Two time-of-flight photoelectron spectrometers were used to detect the photoelectrons. One TOF spectrometer was located at 0^0 and the other at 54.7^0, with respect to the photon electric field vector. Fig. 1 is an example of photoelectron spectrum. This simultaneous measurement of the photoelectrons intensity is crucial for the quantitative determination of the photoelectron angular distribution of rare gas clusters.

The Photoelectron angular distribution parameters of the Ar 2p, Ar 3p and Ar 3s shells in the cluster were determined. In particular, also the

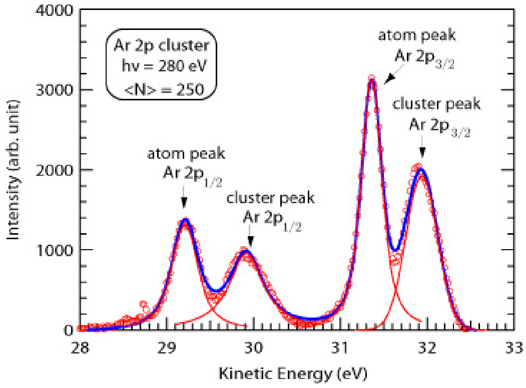

Fig. 1. Ar 2p photoelectron spectrum for Ar clusters with cluster size <N>=250 measured at a photon energy of 280eV. The spectra were recorded with one TOF at 54.7^0. The red and blue lines show the result of least-square fit lines.

angular distribution of the two-spin orbit components, Ar $2p_{1/2}$, and Ar $2p_{3/2}$, in the clusters were obtained. The results demonstrate that the photoelectron angular distributions of outer-valence photoelectrons in clusters are more isotropic than those of the corresponding free atoms. In contrast, the Ar 2p core photoelectron angular distribution of clusters is almost the same as for free atoms. For Ar 3s, the angular distribution parameters for the cluster surface and bulk components were obtained separately and are found to differ significantly from each other and from the atomic value.

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