A New Paradigm to Quantify Relativistic Electron-Electron Interactions in Multielectron Atoms Using Photoionization by Circularly Polarized VUV Radiation

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We have developed a new paradigm to quantify relativistic electron-electron interactions in multielectron isolated atomic systems. This new paradigm utilizes the property that circularly polarized synchrotron radiation has a specific intrinsic angular momentum and complements the well established methods of using the wave nature of synchrotron radiation to study the details of nanoscale systems. Since the angular momentum is conserved in isolated atomic systems, the one unit of angular momentum of the incident circularly polarized photon is absorbed by the atomic system and transferred to atomic electrons as their orbital and spin angular momenta. After photoionization of the atomic system, we measure the degree of circular polarization of the visible fluorescent radiation from the residual excited ion. In the last step of our algorithm we extract a measure of the relativistic interactions between the electrons using the well-known angular momentum coupling rules.

To produce ionizing photons of well-defined angular momentum, we have installed, calibrated and characterized a four-reflection quarter-wave retarder based on earlier suggestions and prototypes.[1, 2] The measurements show the retarder produces better than 99.7% circularly polarized radiation from the linearly polarized VUV radiation from the 10.0.2 beam line in the 35.5 to 37 eV photon energy range typically with 1 % efficiency. Fig. 1 (a) and (b) show polar plots of the intensity of radiation without and with the retardation respectively, as measured by using a 45^{o} gold mirror linear polarization analyzer placed in front of a photodiode.

The degree of circular polarization of the visible fluorescent photons from the excited residual ion is measured in the forward direction with respect to the incident ionizing radiation. In addition, to completely determine the partial wave amplitudes of the emitted photoelectron, when the total angular momentum of the excited ion is larger than ½, the degree of linear polarization of the visible fluorescent radiation is measured in a direction perpendicular to the direction of the incident ionizing photon's helicity. Measurements are obtained for the visible fluorescent radiation from the decay of fine-structure resolved $3p^4\,[^3P]\,4p\;^2P_{1/2,\,3/2}$, $3p^4\,[^3P]\,4p\;^2D_{3/2,\,5/2}$, and $3p^4\,[^1D]\,4p\;^2F_{7/2}$ excited states of Ar^+.

The algorithms we have developed give the expectation value of the z-component of a variety of angular momenta associated with the "residual ion + photoelectron" system or its components. These are the total angular momentum, the total orbital and total spin angular momenta, the orbital and spin angular momenta of the $3p^4$ core-electrons, the orbital and spin angular momenta of the 4p valence electron, and the spin angular momentum of the photoelectron. In addition, we also obtain the amplitudes of the allowed partial waves of the emitted photoelectron. Although we do not measure the photoelectron, we can deduce from our analysis the total spin polarization of all of the electrons emitted to 4π solid angle.

Since the absorbed photon cannot change the total spin of the system which is initially zero, any non-zero measurement of the final spin of the “residual ion + photoelectron” system is a measure of the relativistic interactions during the photoionization. Our measurements show that over the entire energy range studied the relativistic interactions are significant.

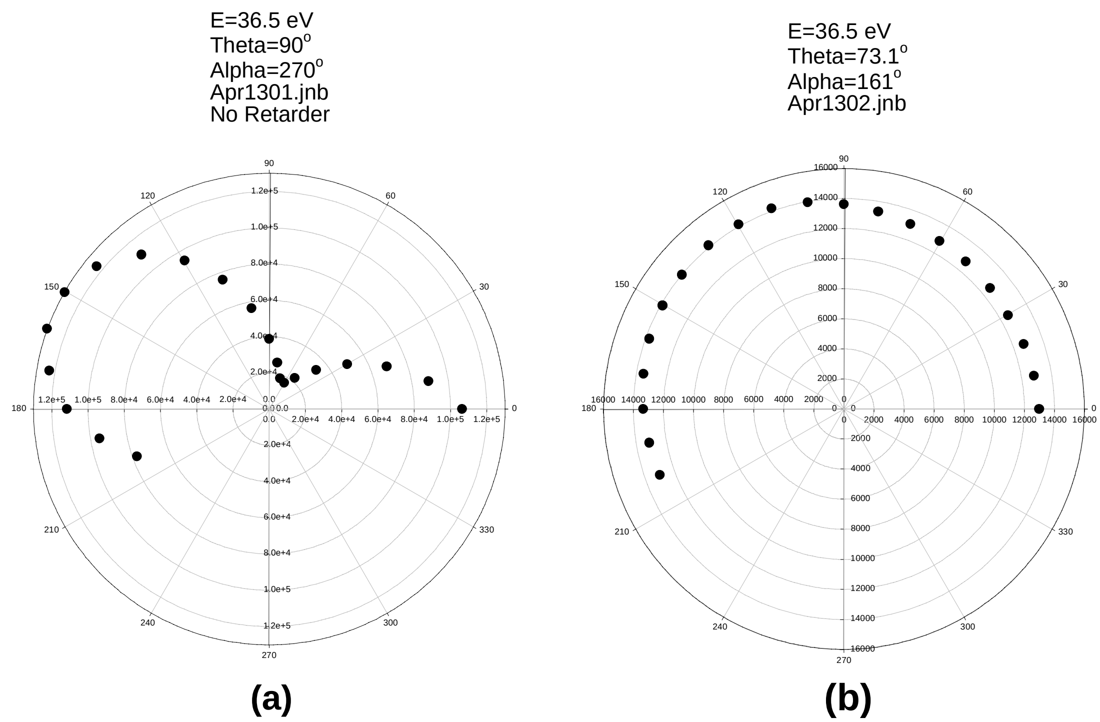

Figure 1. Polar Plots of the intensity of the radiation as a function of the angle of the 45° Gold mirror linear polarization analyzer.
(a): The retarder is out of the incident radiation’s path, the radiation is linearly polarized;
(b): The retarder is in the incident radiation’s path, the radiation is circularly polarized

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