

# Quantifying the Importance of Relativistic Interactions in Atoms Using Photoionization by Circularly Polarized VUV Radiation

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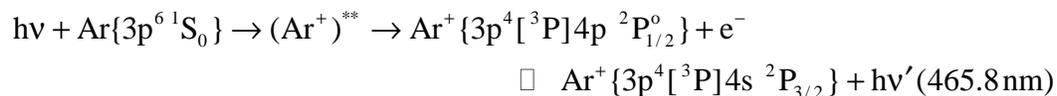
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One of the major goals of current atomic and molecular physics research is the quantification of the degree and nature of the correlated or organized motion of the electrons during dynamic processes. The measurable consequences of the correlated motions of the atomic electrons during photoionization provide us with an effective method to study and characterize the many-electron processes. One class of correlated motion results from relativistic interactions involving atomic electrons. Any photoionization process that changes the initial total spin of the residual ion-electron system is an unambiguous signature of the presence of relativistic interactions. This is due to the fact that photon fields couple only to the spatial parts of the wave functions and cannot change the total spin of the residual ion-electron system. Experiments probing the change in the total spin of the residual ion-electron system thus quantify certain relativistic aspects of the photoionization dynamics.

The organizing principle of our analysis is the determination of the partitioning of the single unit of angular momentum of the incident circularly polarized photon, 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. We extract from our measurements the expectation value of the z-component of the total spin of the residual ion-electron system using angular momentum coupling rules.

Ionizing photons of well-defined angular momentum are produced by a four-reflection quarter-wave retarder we have installed on ALS beam line 10.0.1. This retarder produces better than 99.7% circularly polarized radiation from the linearly polarized VUV radiation in the 35.5 to 37 eV photon energy range, typically with 1 % efficiency. It is based on earlier designs and prototypes.<sup>1,2</sup> In the reaction:



we measure the degree of circular polarization of the visible fluorescent photons from the excited residual ion in a direction that makes an angle of 30° with respect to the direction of the incident ionizing radiation. Note that for this residual ion state, the linear polarization of the fluorescence vanishes everywhere.

The top graph of Fig. 1 shows the total intensity of the fluorescence radiation at 465.8 nm as a function of the ionizing photon energy. The vertical lines denote the position of the doubly excited resonances in Ar. The bottom graph of Fig. 1 shows the orientation parameter  $O_0(J)$  of the excited residual ion and is obtained from the measured degree of circular polarization. By

definition, the orientation parameter  $O_0(J)$  is proportional to the expectation value of the z-component of the total angular momentum, i.e.,  $O_0(J) = J(J+1)\langle M_J \rangle$ . From conservation laws and angular momentum coupling rules one can derive<sup>3</sup> that for this excited ionic state, the orientation parameter  $O_0(S)$  resulting from the total spin  $S$  of the residual ion is proportional to  $O_0(J)$ . Similarly one can also show that<sup>3</sup> the orientation parameter resulting from the spin  $s_e$  of the electrons integrated over  $4\pi$  solid angle satisfies the relation  $O_0(s_e) = O_0(J)$ . Thus, the expectation value of the z-component of the total spin of the excited ion-photoelectron system is proportional to the measured  $O_0(J)$ .

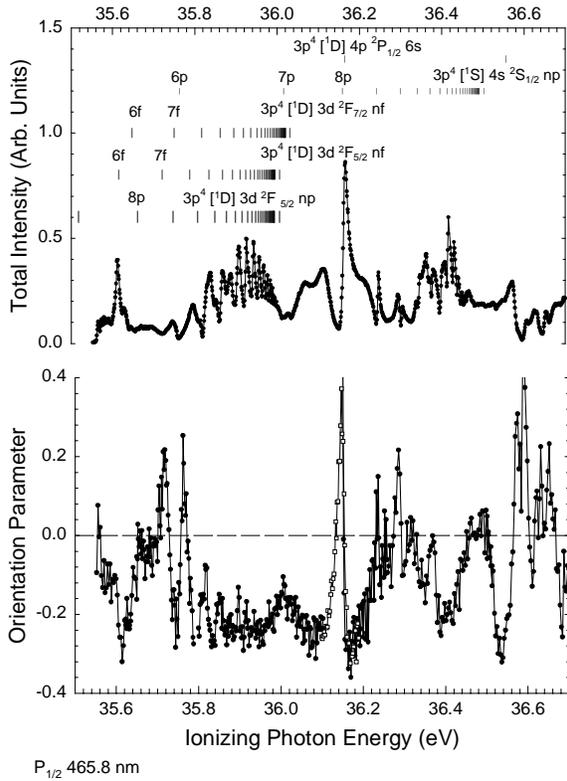


Figure 1. Top graph: Total Intensity of the fluorescence radiation at 465.8 nm at an incident photon resolution of 3 meV. The vertical lines show the position of doubly excited states of Ar. Bottom graph: Orientation parameter for the total angular momentum of the residual excited ion. A non-zero value for the orientation parameter is an unambiguous signature of relativistic interactions during the photoionization.

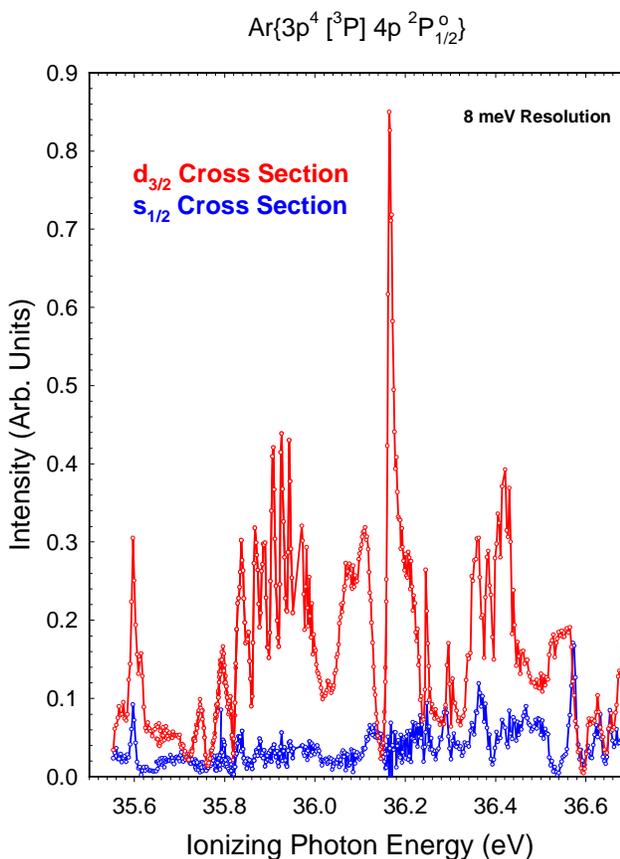
Since the absorbed photon cannot change the total spin of the system which is initially zero, any non-zero measurement of the final spin of the “residual ion + photoelectron” system is a measure of relativistic interactions during the photoionization. The bottom graph of Fig. 1 shows dramatically that over the entire energy range studied the relativistic interactions are significant and strongly energy-dependent.

In addition, the angular momentum coupling rules allows us to obtain the amplitudes of the allowed partial waves of the emitted photoelectron. Parity and angular momentum conservation restrict the emitted photoelectron to s and d partial waves with total angular momentum  $j_e=1/2$  and  $3/2$  respectively. We obtain the partial wave probabilities of the electrons integrated over  $4\pi$  solid angle in terms of the measured orientation coefficient  $O_0(J)$  as

$$|a_{1/2}|^2 = O_0(J) + \frac{1}{3} \quad \text{and} \quad |a_{3/2}|^2 = \frac{2}{3} - O_0(J) = 1 - |a_{1/2}|^2.$$

Multiplying each of these probabilities with the total intensity of the fluorescence provides us with the relative cross sections of  $s_{1/2}$  and  $d_{3/2}$  partial waves as shown in Fig. 2. As can be seen from Fig. 2, the d-wave cross section dominates the spectrum.

Figure 2: Relative cross sections for  $s_{1/2}$  and  $d_{3/2}$  partial waves of the photoelectron. The cross section for  $d_{3/2}$  partial-waves is larger almost everywhere.



Our method of assessing the importance of the relativistic interactions or determining the partial-wave probabilities is independent of any theoretical model and is equally applicable to resonant or non-resonant photoionization.

## ACKNOWLEDGMENTS

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