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MULTIPHOTON IONIZATION PHOTOELECTRON SPECTROSCOPY OF XENON: EXPERIMENT AND THEORY*

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ABSTRACT

Photoelectron energy and angular distributions for resonantly enhanced multiphoton ionization (REMPI) of xenon via the three-photon-allowed $7s[\frac{3}{2}]_1^0$ and $5d[\frac{3}{2}]_1^0$ states have been studied both experimentally and theoretically. The electron kinetic energy spectra give the probability of leaving Xe^+ in either the $^2P_{1/2}$ or $^2P_{3/2}$ core. The measured branching ratio for leaving each ionic core is used to test the theoretical description of the REMPI process. Measurements of *both* the angular distributions and the [3+1] REMPI via the 5d state are adequately reproduced by multichannel quantum defect theory. However, measurements of angular distributions for the electrons resulting from [3+1] via the $7s[\frac{3}{2}]_1^0$ state into Xe^+ $^2P_{3/2}$ (core preserving) or Xe^+ $^2P_{1/2}$ (core changing) are in striking disagreement with theory.

INTRODUCTION

Coupling experimental and theoretical studies of REMPI and photoelectron spectroscopy (PES) can provide unique information about the photoionization dynamics of excited states. REMPI-PES studies of the rare gases have mostly been confined to xenon primarily because the lowest Rydberg states are easily accessible using available lasers. Recently, Blazewicz et al.[1] have shown that multichannel quantum defect theory (MQDT) can provide an excellent description of the [3+2] and [3+1] REMPI processes via the Xe $6s[\frac{3}{2}]_1^0$ and $6s'[\frac{1}{2}]_1^0$ states, respectively. We have extended this work to the $7s[\frac{3}{2}]_1^0$ and $5d[\frac{3}{2}]_1^0$ states of xenon. The apparatus for this experiment has been described elsewhere[1] and will not be discussed here.

RESULTS

Figure 1 shows the energy distribution of the photoejected electrons for the [3+1] REMPI process via the $5d[\frac{3}{2}]_1^0$ level of Xe at 357.6 nm. Electrons with an energy of 1.7 eV dominate the spectrum. This corresponds to a non-core-changing ionization process. In addition, a small signal at 0.4 eV is seen which corresponds to photoelectrons leaving the Xe^+ $^2P_{1/2}$ ionic core. The approximate ratio of the two observed signals is comparable with that predicted from MQDT calculations. Figure 1 also shows the polar plot of the measured angular distribution for the electrons resulting from REMPI via the $5d[\frac{3}{2}]_1^0$ level. The electron signal corresponding to leaving the $^2P_{1/2}$ core was too weak to obtain a reliable angular distribution measurement. Fitting the angular distribution data to a Legendre polynomial expansion required terms up to $P_8(\cos\theta)$:

$$Xe^+\ ^2P_{3/2} : I(\theta) = 1 + 1.23P_2 + 0.75P_4 + 0.20P_6 - 0.70P_8$$

The calculated angular distribution from MQDT for the $^2P_{3/2}$ electrons quantitatively reproduces the angular distribution observed in the experiment.

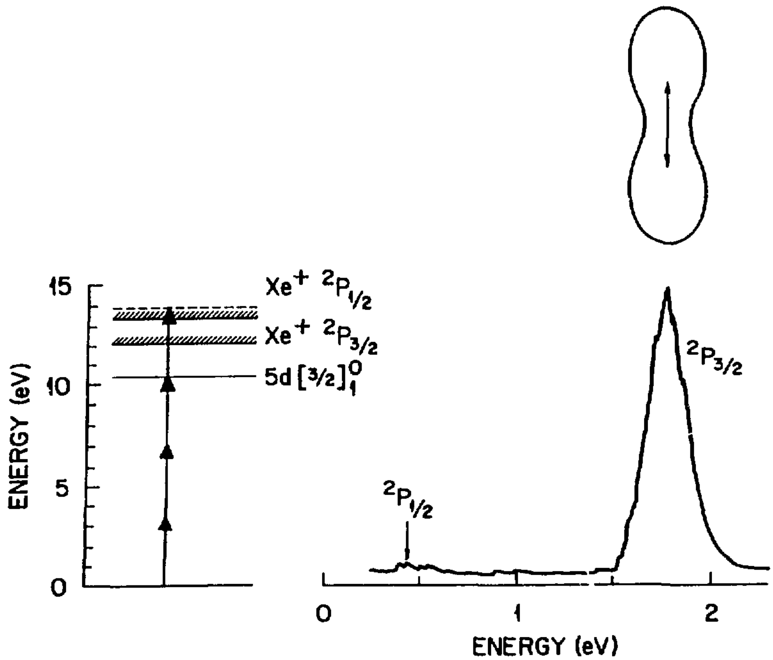

Figure 1. PES and angular distribution for REMPI via the 5d state.

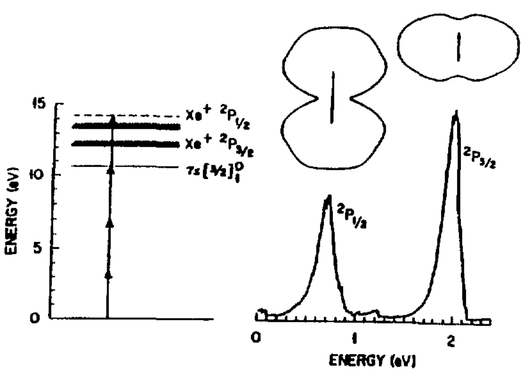

Figure 2. PES and angular distributions for REMPI via the 7s state.

Figure 2 shows the energy distribution of the photoelectrons for the [3+1] REMPI process through the $7s[\frac{3}{2}]_1^0$ state of Xe at 351.1 nm. Two groups of electrons are observed at 0.7 eV and 2.0 eV which correspond to leaving the $Xe^+\ ^2P_{1/2}$ and $^2P_{3/2}$ ionic cores respectively. The ratio of the two signals (approx. 2:1) compares favorably with that predicted from MQDT.

The polar plots of the experimental angular distributions for the two groups of electrons are shown above their respective energy distributions. As can be seen, the two groups yield quite different angular distributions. The experimental angular distributions were also fitted to a Legendre polynomial expansion. It was found that terms up to $P_8(\cos\theta)$ were needed to fit both the $^2P_{1/2}$ and $^2P_{3/2}$ data.

$$Xe^+\ ^2P_{3/2}: \qquad I(\theta) = 1 - 0.33P_2 + 0.02P_4 - 0.03P_6 - 0.04P_8$$

$$Xe^+\ ^2P_{1/2}: \qquad I(\theta) = 1 + 0.78P_2 - 0.42P_4 + 0.30P_6 - 0.16P_8$$

MQDT calculations semiqualitatively agree with the general shape of the experimental angular distribution for the $^2P_{1/2}$ electrons except for a predicted small maxima at $\pi/2$ where a sharp minima is observed in the experiment. MQDT calculations predict that the angular distribution for the $^2P_{3/2}$ core-preserving electrons should have a deep and somewhat broad minima at $\pi/2$ and a maxima at 0 and π. However the experiment shows that the angular distribution is somewhat isotropic with an unexpected maxima at $\pi/2$ and minima at 0 and π. This discrepancy is not understood at this time.

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REFERENCE

1. P. R. Blazewicz, X. Tang, R. N. Compton, and J.A.D. Stockdale, J. Opt. Soc. Am. B., 4(5), 770 (1987).