

(BL-3B)

## Laser photoionization electron spectroscopy of polarized rare gas atoms excited with synchrotron radiation

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We have performed laser photoionization spectroscopy of polarized rare gas atoms in order to study spin-orbit interactions of many electron systems and exploit the possibility of the complete photoionization experiment. Rare gas atoms in a gas cell are excited by linearly polarized synchrotron radiation (85-100nm) supplied from the beamline BL3B and aligned toward the electric vector. The light beam of a Nd:YAG laser (532nm) intersects with the synchrotron radiation at a right angle and ionize the aligned targets. The electric vector of the laser can be rotated by using a half wave plate. Photoelectrons, emitted in the direction perpendicular to both the laser and synchrotron radiation, are detected with a 160° hemispherical electrostatic analyzer.

A two-dimensional spectrum of Kr is shown in Fig. 1. Major spots due to high photoelectron yield are aligned on two straight lines with a slope of unity; these spots are assigned to two final ionic states of Kr<sup>+</sup> [<sup>2</sup>P<sub>1/2, 3/2</sub>]. Since electronic states of excited Kr atoms are designated by a J<sub>c</sub>-ℓ coupling scheme, a total angular momentum of the product ion J<sub>f</sub> can be expected to be equal to that of a core ion of an excited state J<sub>c</sub>. However, this picture seems to disagree with the present results in some cases. There are two possible explanations for this disagreement: the excited states designated by the J<sub>c</sub>-ℓ coupling are influenced by mixing of J<sub>c</sub> of 1/2 and 3/2, or an ejected electron exchanges angular momenta with the ion core during the photoionization process.

In Fig 2(a), the photoelectron yield for Ar(5s'[1/2]<sub>1</sub>)→Ar<sup>+</sup>(<sup>2</sup>P<sub>1/2</sub>) + e<sup>-</sup> (ℓ=1; j=1/2, 3/2) is plotted as a function of the angle ψ<sub>e</sub> between the electric vector of the laser and the direction of the linear momentum of photoelectrons. The angular distribution should be interpreted in terms of a p wave from excited Ar aligned toward the electric vector of synchrotron radiation. We have obtained an asymmetry parameter β of 1.96 from this result. The angular distributions have also been measured for photoelectrons produced by the process Ar(3d[1/2]<sub>1</sub>)→Ar<sup>+</sup>(<sup>2</sup>P<sub>f</sub>) + e<sup>-</sup> (ℓ=1 or 3; j=1/2, 3/2 or 5/2, 7/2) with J<sub>f</sub>=1/2 and 3/2. As shown in Fig. 2(b), the β values for these distribution curves are much smaller than that for photoelectrons from the 5s'[1/2]<sub>1</sub> state. We are planning the complete photoionization experiment to determine dipole matrix elements and a phase shift difference of each partial wave.

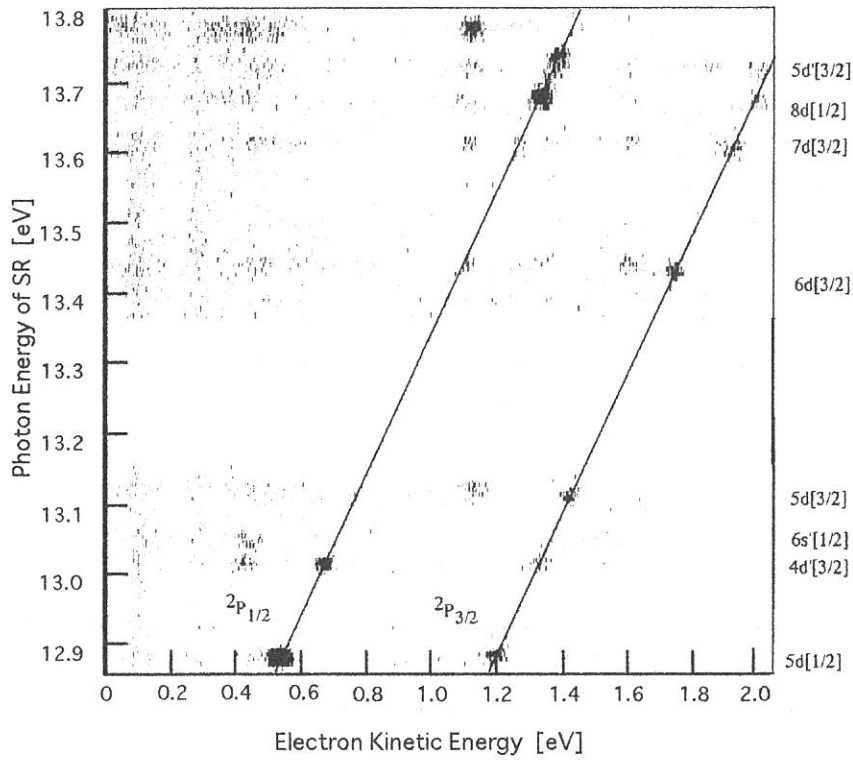


Fig 1. Two-dimensional spectrum of Kr. The electric vector of the laser is parallel to the direction of photoelectrons

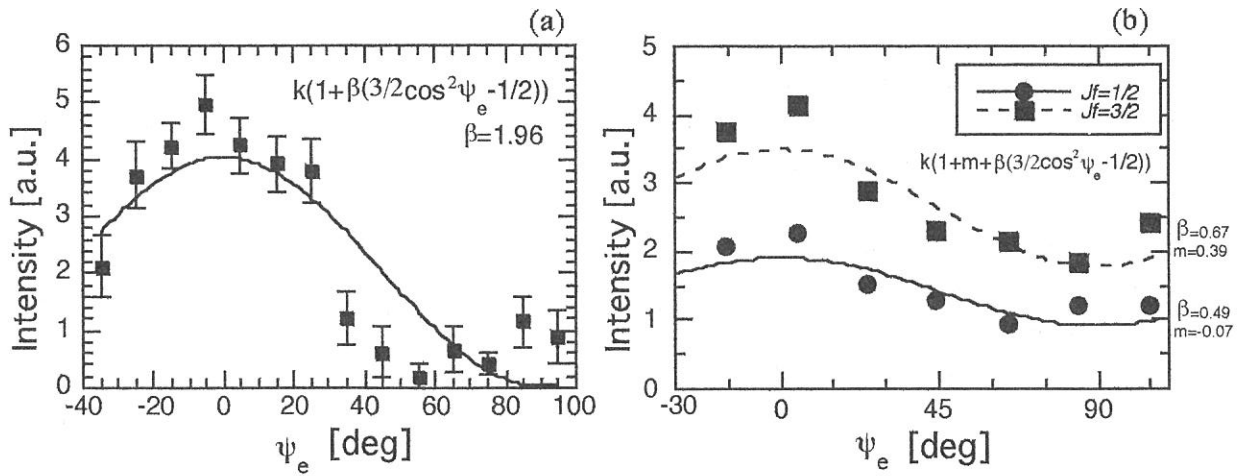


Fig. 2. Angular distributions of photoelectrons: (a)  $\text{Ar}(5s'[1/2]_1) \rightarrow \text{Ar}^+(^2P_{J_f})$  and (b)  $\text{Ar}(3d[1/2]_1) \rightarrow \text{Ar}^+(^2P_{J_f})$ . Photoelectron yields were measured as a function of  $\psi_e$ .

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## Spectator- and Participant-Like Behavior of a Rydberg Electron on Predissociation of Superexcited States of OCS

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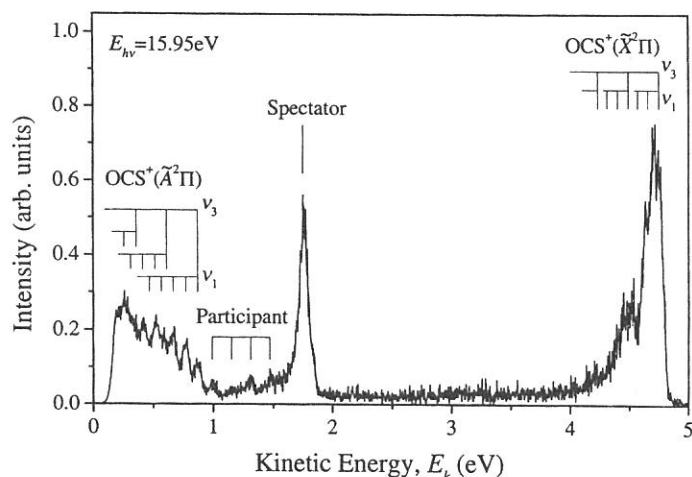
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Predissociation of superexcited states of OCS is studied by two-dimensional photoelectron spectroscopy using synchrotron radiation in the photon energy range of 15 - 16.5 eV.<sup>1)</sup> A two-dimensional photoelectron spectrum exhibits two kinds of characteristic patterns both of which are ascribed to autoionization of sulfur atoms. To clarify the point, we show a photoelectron spectrum of Figure 1 which is obtained as a cut through the two-dimensional spectrum along the electron kinetic energy axis at the photon energy of 15.95 eV.<sup>2)</sup> The superexcited atom S\* is produced by predissociation of a Rydberg state  $\text{OCS}^*(R_B)$  converging to  $\text{OCS}^+(\tilde{B}^2\Sigma^+)$ . The pattern of the first kind results from predissociation processes in which the effective principal quantum number  $n$  of the Rydberg electron is almost conserved. This suggests that the Rydberg electron behaves as a spectator because of its negligibly weak interaction with the ion core (spectator predissociation). On the contrary,  $n$  of S\* does not accord with that of  $\text{OCS}^*(R_B)$  in the pattern of the second kind, indicating that the Rydberg electron participates directly into the electron exchange mechanism controlling conversion from  $\text{OCS}^*(R_B)$  to a predissociating state (participant predissociation). With increasing  $n$ ,  $\text{OCS}^*(R_B)$  decays more preferentially by the spectator than by the participant predissociation. The spectator predissociation of  $\text{OCS}^*(R_B)$  proceeds through a two-step conversion which involves Rydberg states converging to  $\text{OCS}^+(\tilde{A}^2\Pi)$  and  $\tilde{X}^2\Pi$  and a dissociative multiple-electron-excited state  $\text{OCS}^*(SAT)$  asymptotically correlating with  $\text{S}^* + \text{CO}(\tilde{X}^1\Sigma^+)$ . In contrast, the participant predissociation may be accounted for by a direct conversion from  $\text{OCS}^*(R_B)$  to  $\text{OCS}^*(SAT)$ . The quantum yields are estimated from Figure 1 to be 0.07 and 0.02 for the participant and spectator predissociation, respectively, at the incident photon energy of 15.95 eV where  $\text{OCS}^*(R_B)$  states with  $n \sim 12$  lie. A simulation is performed to reproduce the partial cross section curve for the spectator predissociation by using a model in which the decay rates for the participant and spectator predissociation are assumed to be proportional to  $n^{-3}$  and  $n^0$ , respectively. The simulated and experimental cross section curves are in good agreement with each other at the photon energy higher than 15.8 eV.

### References

- 1) Y. Hikosaka, H. Hattori, T. Hikida, and K. Mitsuke, *J. Chem. Phys.* **107**, 2950 - 2961 (1997).
- 2) Y. Hikosaka, H. Hattori, and K. Mitsuke, *ibid.* **110**, 335-344 (1999).



**Figure 1.** Photoelectron spectrum of OCS at the photon energy of 15.95 eV where  $\text{OCS}^*(R_B)$  states with  $n \sim 12$  are located.