

# Photon Factory Activity Report 2001 #19B

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## Doubly excited states of ethylene in the vacuum ultraviolet range as studied by dispersed uv-visible fluorescence spectroscopy

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### Introduction

Superexcited states play an important role in the photoabsorption of molecules in the vacuum ultraviolet range [1]. One of the hot topics concerning these states is the dynamics and spectroscopy of doubly excited states [2,3]. Less has been known about the doubly excited states of polyatomic molecules compared with those of diatomic molecules. We were successful in observing the doubly excited states of methane in the cross section curves of uv-visible fluorescences from dissociation fragments in the photoexcitation of methane [4]. In the present experiment, we have measured the cross sections for the emission of fluorescences emitted by dissociation fragments in the photoexcitation of ethylene ( $C_2H_4$ ) as a function of incident photon energy in the range of 15 – 40eV.

### Experimental

Synchrotron radiation dispersed by a 2400 lines/mm grating in the 3m-NIM at BL-20A was used as an incident light source. Fluorescences emitted by the neutral fragments produced in the photoexcitation of ethylene were dispersed by a 300mm uv-visible monochromator equipped with a 300lines/mm grating and a CCD detector [4]. This liquid  $N_2$ -cooled CCD detector covers the wavelength range of which width is 280nm in one frame. Several fluorescence lines and bands were observed in the fluorescence wavelength range of 350 – 680 nm. Among them, cross sections for the  $CH(A,B \rightarrow X)$ , Balmer- $\beta$ ,  $\gamma$ ,  $\delta$ , and  $\epsilon$ , and  $C_2(d \rightarrow a, \Delta v = -1, 0)$  fluorescences were put on an absolute scale using the  $N_2^+(B)$  partial ionization cross section as described in reference [4].

### Results and Discussions

In Fig. 1 the cross sections for the emission of (a) the  $CH(A \rightarrow X)$  and (b) Balmer- $\beta$  fluorescences in the photoexcitation of ethylene are plotted against the incident photon energy along with the lowest dissociation limits for the neutral dissociation and dissociative ionization processes involved.

Peaks indicated by arrows in Fig. 1 are attributed to the superexcited states of ethylene. The peaks around 27 eV in Fig. 1(b) seem to originate from doubly excited states of ethylene since they lie above the ionization threshold of the inner-most valence electron as shown in the upper part of Fig.1 (b).

### References

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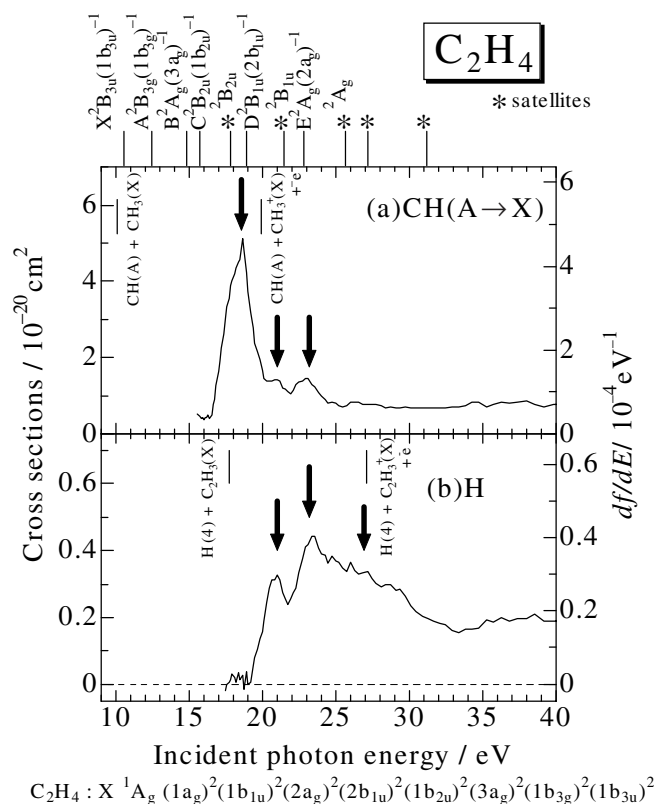


Fig.1 Cross sections for the emission of (a) the  $CH(A \rightarrow X)$  and (b) Balmer- $\beta$  fluorescences as a function of incident photon energy in the photoexcitation of ethylene. The oscillator strengths for the same processes are shown on the right axes. Vertical bars on the upper part of the figure indicate ionization potentials of ethylene. The lowest dissociation limits for the neutral dissociation and dissociative ionization processes involved are also indicated.

## Stark quantum beat of He

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Since the energy splitting between  $M=0$  and  $M=\pm 1$  is very small, it is not possible to measure it in the usual strength electric field and a conventional monochromator. However, if the interference between two levels occurs, the energy splitting between  $M=0$  and  $M=\pm 1$  can be measured. In this research, the Stark energy splitting between magnetic quantum number  $M=\pm 1$  and  $0$  of He( $1s^2S_0-1snp^1P_1$  Series) was measured through the quantum beat. The experimental values of the energy splitting were compared with calculated values.

The experiment was performed using synchrotron radiation as a light source on beam line 20A of KEK. The fluorescence excitation spectrum and the decay curve of He ( $1s^2S_0-1snp^1P_1$  Series) were measured. The synchrotron radiation was monochromitized by the 3m normal incidence monochromator, and focused in the interaction region using three mirrors. He gas supplied through the supersonic jet nozzle in the interaction region was excited by the monochromitized light, and fluorescence from the He Rydberg state was detected using MCP (Micro Channel Plate). A supplied the static electric field of 0-5kV/cm was to the interaction region. When a static electric field vertical or parallel to an electric vector of light is supplied, the atom was excited to  $M=\pm 1$  or  $M=0$ , respectively, because the ground state of He is  $M=0$ . To excite both levels simultaneously, the static electric field was supplied at  $45^\circ$  for an electric vector of light.

Fig.1 shows the quantum beat spectrum in He  $7^1P_1$  obtained by the experiment. The energy splitting in the electric field can be deduced from the quantum beat spectrum. Fig.2 show the experimental and calculated values of the beat frequencies (corresponding to energy splitting) versus the square of the electric field strength. The experimental values were indicated by the dots and the calculated values were indicated by the line. The calculated values were obtained by using the perturbation theory[1]. It can be confirmed at the experimental and calculated value almost agree. Experimental error was assumed to be  $\pm 4\%$ , mainly caused by the distortion of the electric field.

Now calculation is being performed to higher principal quantum numbers.

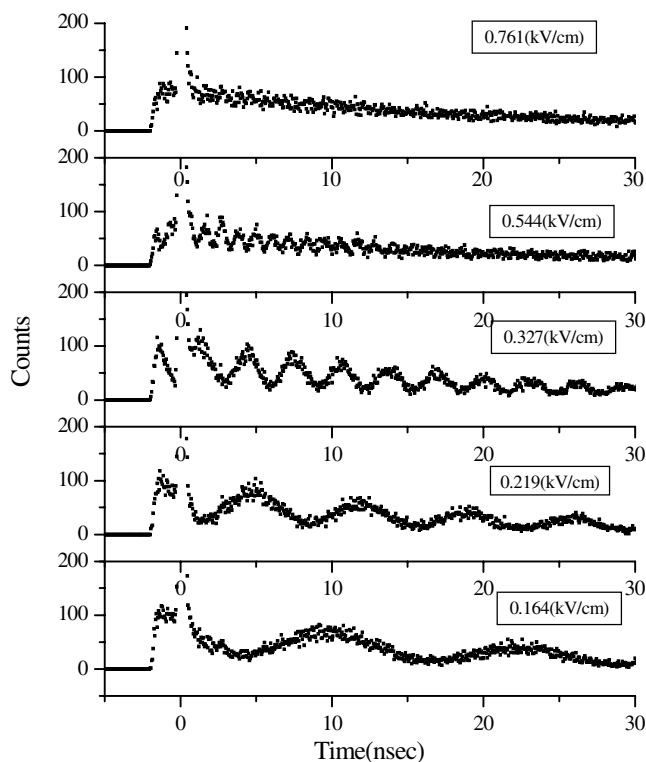


Fig.1. The quantum beat spectrum in He $7^1P_1$  state

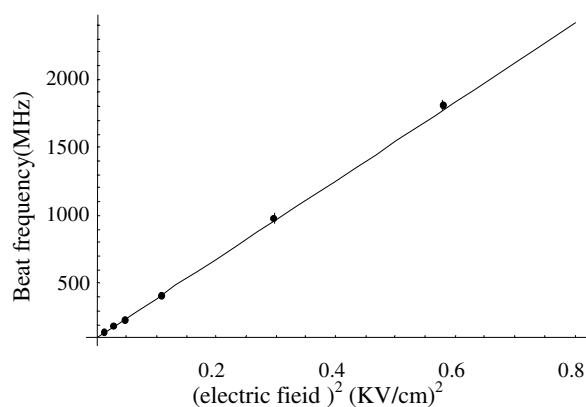


Fig.2. The beat frequency versus the square of the electric field strength ( $7^1P_1$ ). The experimental values were indicated by the dots and the calculated values were indicated by the line.

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## Stark quantum beat of Kr in V.U.V. region

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### Introduction

We have obtained Stark Quantum Beat (SQB) spectrum under a static electric field in vacuum ultraviolet fluorescence decay spectra of Kr atom resonance lines. Kr was excited by pulsed vacuum ultraviolet light in a synchrotron single bunch operation. SQB spectroscopy is based on the quantum interference effect between two levels which are in close proximity. When an atom under the static electric field was excited by polarized light to magnetic sublevels  $M=0$  and  $|M|=1$  simultaneously, SQB appears in the fluorescent decay spectra due to the quantum interference effect between the two levels. From the beat period we can determine the frequency and the energy between the levels.

### Experiment

The experiments were performed on beamline 20A. The synchrotron radiation was monochromatized by a 3-m normal incidence Eagle mounted scanning monochromator equipped with a 2400 lines/mm gold-coated grating. When the entrance and exit slit were set to be 10  $\mu\text{m}$ , the monochromator has a resolution ( $E/\Delta E$ ) of about 60,000. Synchrotron radiation in a single bunch operation had a pulse width of approximately 50 psec and a repetition period of 624 nsec. The target gas was supplied by the jet nozzle and crossed perpendicularly to the excited light. Fluorescence emitted from the excited Kr atom was detected by the MCP and set to detect a 45° gradient to the direction of the electric field.

### Result

Fig.1 shows the obtained fluorescence excitation spectrum of Kr covering the Rydberg series converging to  $^2P_{3/2}$  and  $^2P_{1/2}$  ionization thresholds by synchrotron multi bunch operation. We applied a DC electric field less than 1[kV/cm] to the interaction region and observed the Stark quantum beat spectra. Fig.2 shows time resolved fluorescence spectra of the Kr  $8d[1/2]_1$  state at field free and applied electric field by single bunch operation. The spectra were Fourier transformed and shown in the right side. The observed beat frequencies varied proportionally to the square of the external electric field. The theoretical energy gap between the  $M=0$  and  $|M|=1$  levels can be calculated using a  $jl$ -coupling scheme.

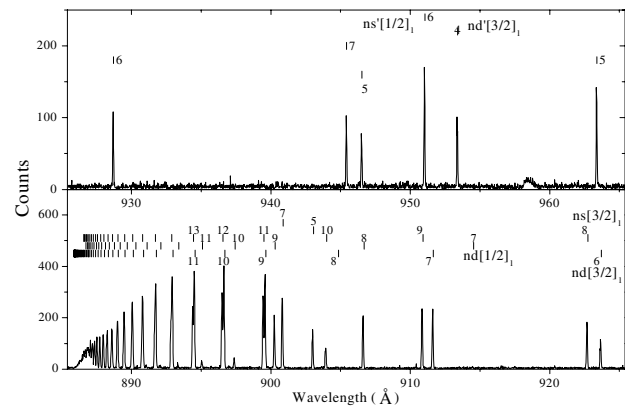


Fig.1 Fluorescence spectrum of Kr.

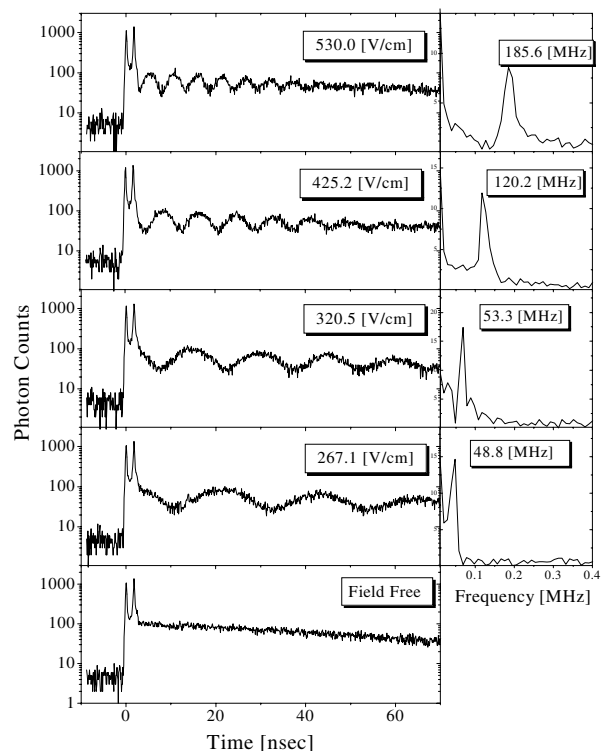


Fig.2 Left figures shows SQB spectra of the Kr  $8d[1/2]_1$  state. Figures on the right show Fourier spectra of the left ones.

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# SAXS measurement of supercritical xenon

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### Introduction

For disordered materials such as liquids, solutions, fluids and amorphous solids, concept of inhomogeneity is of peculiar importance. Small-angle scattering methods by use of x-ray and neutron give the most direct information on the inhomogeneity of molecular distribution. A supercritical fluid, which is defined as a fluid above its critical temperature and pressure, is essentially characterized by the large inhomogeneity. Because of much higher intensity of x-ray source than neutron one, a small-angle x-ray scattering (SAXS) method will be the most powerful and appreciable experiment for the study.

Recently we performed the SAXS study of supercritical fluids with hydrogen bonding such as water[1], methanol, ethanol, and alcohol aqueous solutions[2]. In this report, we describe the SAXS measurements for supercritical xenon, which contrastively is a simple fluid with only van der Waals interaction, to discuss the behaviours of the inhomogeneity.

Fig. 2 shows the Ornstein-Zernike plots for the same thermodynamic states as those shown in Fig. 1. The both plots show the satisfying linearity, though it is difficult to perform a SAXS measurement in supercritical xenon due to the large value of the absorption coefficient for xenon atom. The linearity indicates that little uncertainty was caused by the experimental and analytical procedure.

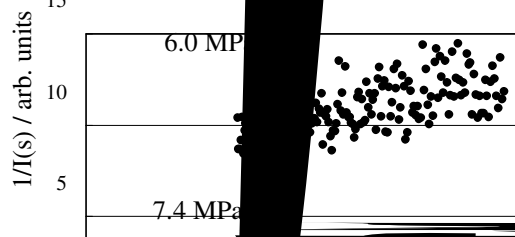


Fig. 1: SAXS intensities for supercritical xenon at 302.0 K.

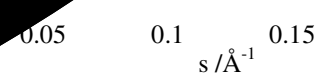


Fig. 2: Ornstein-Zernike plots for supercritical xenon at 302.0 K.

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## Dissociation process of the $\text{Kr}_2^+ \text{C}_2 \ ^2\Pi_{1/2u}$ state

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### Introduction

Studies on the photoionization of rare gas dimers and their ions are important to understand the nature of the van der Waals interaction. In the past two decades, many theoretical and experimental studies on the homonuclear rare gas dimers have been performed. We have already observed threshold photoelectron (TPE) spectra of Kr gas and measured vibrational structures of  $\text{Kr}_2^+$ [1]. After that, we also observed threshold photoelectron photoion coincidence (TPEPICO) spectra of  $\text{Kr}_2$ . Unexpectedly, we found that at the peak of the  $\text{Kr}_2^+ \text{C}_2 \ ^2\Pi_{1/2u}$  TPE spectrum, the threshold photoelectrons are detected in coincidence not with  $\text{Kr}_2^+$  ions, but with  $\text{Kr}^+$ [2]. This observation points to a rapid predissociation of the  $\text{Kr}_2^+ \text{C}_2 \ ^2\Pi_{1/2u}$  state to  $\text{Kr}^+(\ ^2P_{3/2}) + \text{Kr}(^1S_0)$ .

### Experiment

The measurements were carried out at the beamline 20A of the Photon Factory. The experimental setup and technique were same as our previous study[2]. In brief, photoelectrons extracted by a penetrating field were focused by a lens system and then led to a hemispherical electrostatic analyzer. The Time-of-Flight (TOF) ion mass analyzer enabled us to select mass-identified photoions.

When a dissociation product has a kinetic energy in the ionization region, it is expected that the peak of the TOF spectrum has wide width. Since no competition between dissociation processes is expected and the only final state is  $\text{Kr}^+(\ ^2P_{3/2}) + \text{Kr}(^1S_0)$ , the kinetic energy of the fragment  $\text{Kr}^+$  in the ionization region should be dominated by only one energy. Generally, if the kinetic energy of the ion is narrowed, if the ions are released symmetrically, and if all ions are detected, the TOF spectrum would have a rectangular shape, the center of which is the flight time of  $\text{Kr}^+$ , and its width  $W$  given by;

$$W = \frac{2\sqrt{2m}}{qE_s} \sqrt{U} \quad (1)$$

[3], where  $m$  is the mass of the ion,  $q$  is the ionic charge,  $E_s$  is the electric field in the collision region, and  $U$  is the kinetic energy of the ion.

We expected it could be observed the rectangular TOF peak, which has the width estimated as  $\sim 10$  sec from the energy difference between the vibrational level of the  $\text{Kr}_2^+ \text{C}_2 \ ^2\Pi_{1/2u}$  state and the first dissociation limit ( $\sim 550$  meV).

### Results

In Fig. 1 is shown the observed TOF spectrum at the photon energy of the  $\text{Kr}_2^+ \text{C}_2 \ ^2\Pi_{1/2u}$  state ( $v=2$ ). It seems that the spectrum shape is not rectangular and the width is narrower than the estimated width ( $\sim 10$   $\mu\text{sec}$ ). This indicated the  $\text{C}_2 \ ^2\Pi_{1/2u}$  state loses energy before dissociating to  $\text{Kr}^+ + \text{Kr}$ . We propose that this energy consumption can be ascribed to the radiative transition of the  $\text{C}_2 \ ^2\Pi_{1/2u}$  state to the intermediate repulsive  $\text{B}_2 \ ^2\Pi_{1/2g}$  state correlated to the first dissociation limit. We found the shape of the TOF peak could be made up by accumulation of the rectangular spectrum under concerning the effect of the life time. The fitting which designates the potential energy of the  $\text{B}_2 \ ^2\Pi_{1/2g}$  state and the life time as parameters was performed.

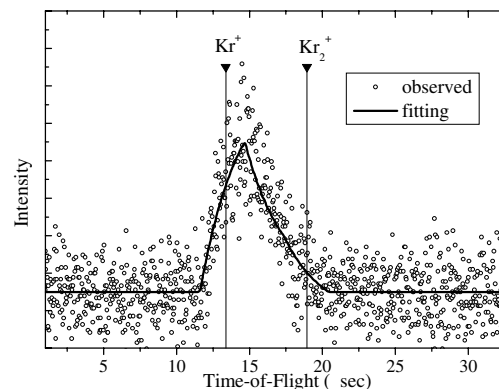


Fig. 1 The TOF spectrum at the photon energy of the  $\text{Kr}_2^+ \text{C}_2 \ ^2\Pi_{1/2u}$  state

The result of the fitting is also shown in Fig. 1. From the fitting, the potential energy of the  $\text{B}_2 \ ^2\Pi_{1/2g}$  state from the first dissociation limit and the life time of the radiative transition were obtained as  $\sim 50$  meV and  $\sim 2.7$  sec. These values were obtained experimentally for the first time and the obtained potential energy of the  $\text{B}_2 \ ^2\Pi_{1/2g}$  state is comparable with the theoretical prediction[4].

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## Dissociation of inner valence states of $O_2^+$ studied by threshold photoelectron-photoion coincidence spectroscopy

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### Introduction

Breakdown of the independent-particle orbital picture is characteristic of inner valence electrons in molecules. Therefore, for molecular photoionization of the inner valence shells, this leads to many final states of configuration interaction satellites. An attractive objective is to obtain detailed information on dissociation processes of the inner valence states, because these processes are closely related to the electronic character and potential energy surface of the final states of the molecular ions. Experimental investigation is still limited, even for diatomic molecules, owing to the small cross sections for photoionization into inner valence states. In contrast, modern configuration-interaction calculations have become capable of accurately describing these inner-valence states.

One of the effective experimental approaches is threshold photoelectron-photoion coincidence spectroscopy, where fragment ions are detected simultaneously with threshold photoelectrons so as to define the binding energy of the initial inner valence states. The experimental setup is that used for Auger electron-threshold electron coincidence measurements [1], except the hemispherical electrostatic analyzer equipped with a position sensitive detector is here used to detect energetic fragment ions.

### Experimental results

Measurements were carried out on beamline BL20A at the Photon Factory, equipped with a 3-m normal incidence monochromator. The top panel of Fig. 1 shows a threshold electron spectrum of  $O_2$  in the inner valence region (25 - 45 eV). The spectrum exhibits several broad peaks, which are not always given definite assignments, as well as discrete peaks probably assignable to Rydberg states of  $O_2^+$  converging to  $O_2^{2+}$ . We have measured kinetic energy spectra of fragment  $O^+$  in coincidence with threshold electrons at different photon energies, mainly on the broad peaks. The kinetic energy spectra of  $O^+$ , thus obtained, depend on the final molecular states, reflecting potential energy curves of related electronic states and their dissociation dynamics. Typical coincidence spectra are shown in the middle and bottom panels, which are measured for  $O_2^+(^2\Sigma_u^-)$  and  $O_2^+(^4^2\Pi_u)$ , respectively. These ion states dissociate mainly to the limits  $O^+(^2P) + O(^2P)$  and/or  $O^+(^2P) + O(^1D)$ , though dissociation to other limits are also recognizable. For the  $O_2^+$  states of higher binding energies, relatively slow  $O^+$  is found to be dominant. This shows that neutral O atoms, dissociation partners of  $O^+$ , are electronically excited.

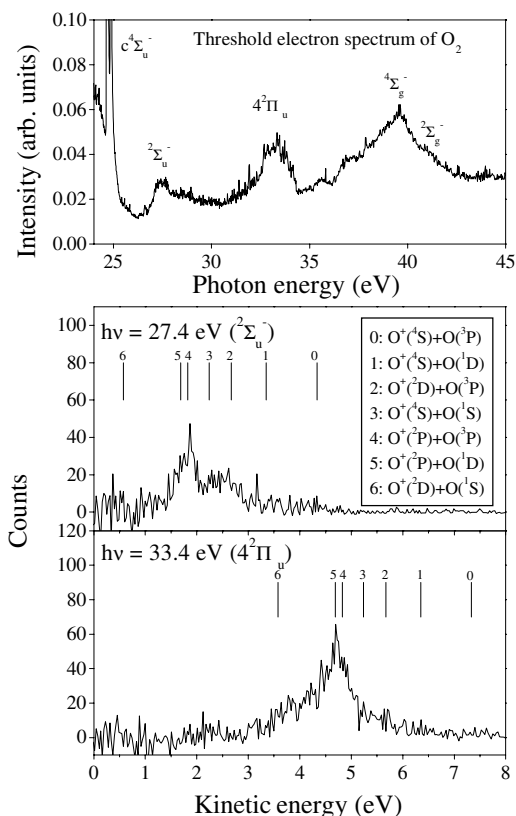


Figure 1. A threshold electron spectrum of  $O_2$  (top), and kinetic energy spectra of fragment  $O^+$  measured in coincidence with threshold electrons (middle and bottom).

### References

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## How are threshold electrons produced on the Xe 4dnp resonance?

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### Introduction

Our scientific interest, for several years, has been to elucidate the decay dynamics following photoionization and photoexcitation of innershell electrons using Auger and/or photoelectron spectroscopy in coincidence with threshold electrons.

It is well known that threshold electrons are produced following the  $4d \rightarrow np$  excitation in Xe. However, the question; which process is responsible for the formation of threshold electron on these resonances, is still unanswered. Heimann et al [1] suggested that the origin of the threshold electrons is shake-off, that is, direct emission of two electrons from decay of the np state. In this case, the two emitted electrons share the available energy and it is probable that one emerges as a threshold electron. On the other hand, Hayaishi et al [2] proposed, based on coincidence measurements between threshold electrons and ions, that a cascade or two-step model can also explain the resonant production of threshold electrons.

Here, we report coincidence measurements between threshold electrons and Auger electrons associated with the decay process  $Xe^* 4d^{-1}np \rightarrow Xe^{2+} 5p^{-2}$  and  $5s^{-1}5p^{-1}$ , which may give an answer to the above question.

### The present experiment

The experiment consists in detecting the threshold electrons and the associated energetic electrons in coincidence [3]. The measurements were made at BL16B of the Photon Factory.

We present, in Fig.1, the Auger electron spectra measured on the  $Xe^* 4d^{-1}np$  ( $n=6, 7$ , and  $8$ ) resonances in coincidence with threshold photoelectrons, together with the spectrum at the  $4d_{5/2}$  threshold showing the energy levels of the  $Xe^{2+} 5p^{-2}$  state. It can be seen that the Auger peak associated with the  $Xe^{2+} 1D$  state is strongly enhanced at the  $Xe^* 4d^{-1}7p$  resonance.

In Fig.2, the difference in arrival times between Auger and threshold electrons is shown for the different resonance peaks. It is apparent that, on the  $4d^{-1}7p$ -peak, the electrons, which we detect as threshold electrons, are slightly energetic. This indicates that a two-step process is present for the production of pseudo-threshold electrons on the  $4d^{-1}7p$ -peak. First, the  $4d^{-1}7p$  state decays into a  $Xe^+$  state, whose binding energy is very close to, but slightly above, the  $Xe^{2+}(1D)$  state and, second, this  $Xe^+$

state produces a low energy electron via autoionization into  $Xe^{2+}(1D)$  state. The most plausible candidate for the  $Xe^+$  state is the  $Xe^{+*}(5p^{-2}(1S)8p)$  state.

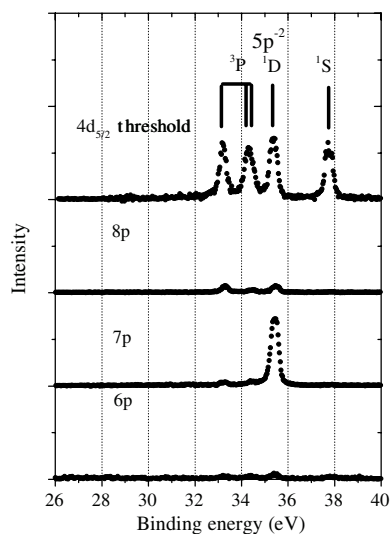


Figure 1 Auger spectra measured on the  $Xe 4d^{-1}np$  resonances in coincidence with threshold electrons.

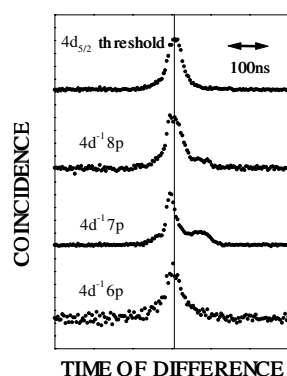


Figure 2. Coincidence spectra between Auger and threshold electrons at the  $Xe 4d^{-1}np$  resonances.

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## Ar 2s Auger spectrum observed in coincidence with the 2s photoelectron in the sub-natural linewidth regime

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### Introduction

The 2s hole in Argon is extremely short lived, due to its fast decay involving the 2p shell (Coster-Kronig decay). As a consequence, the resulting Auger spectrum is broadened by 2.25 eV. This prevents the observation of the detailed spectroscopy of the final Ar<sup>++</sup> states produced, as well as their relative intensities. Previous measurements of the Ar 2s Auger spectrum produced by electron impact [1] or more recently by photon impact [2] were faced with this 2.25 eV limit and hardly resolved the 2 groups of states corresponding to the Ar<sup>++</sup> 2p<sup>-1</sup>3p<sup>-1</sup> states and the Ar<sup>++</sup> 2p<sup>-1</sup>3s<sup>-1</sup> states, respectively.

Our approach to overcome this limit uses photoionization with synchrotron radiation and consists in measuring the Ar 2s Auger spectrum in coincidence with the associated 2s photoelectron. Energy conservation shows then that the final state is defined with a precision that depends only on the combined resolutions of the photons and of both electron detectors. The limitation associated with the hole lifetime is thus removed, as was shown by revealing, for the first time, the structure of the Ar<sup>++</sup> 2p<sup>-1</sup>3p<sup>-1</sup> states [3]. This situation is referred to as the "sub-natural linewidth regime" [4].

### The present experiment

Our experiment [5] uses here a high luminosity threshold electron spectrometer, dedicated to the detection of the photoelectron, and a hemispherical electrostatic analyzer, equipped with a two dimension position sensitive detector for the detection of the Auger electron. These measurements on Argon were made on the BL16B beam line at the Photon Factory.

The complete coincident Auger spectrum reveals the spectroscopy of the Ar<sup>++</sup> dication with a hole located in the 2p innershell. The Ar<sup>++</sup> 2p<sup>-1</sup>3p<sup>-1</sup> states were observed with better statistics and resolution than previously [3], as shown in the figure. Population of the Ar<sup>++</sup> 2p<sup>-1</sup>3p<sup>-1</sup> (<sup>1</sup>S) is now demonstrated by the peak at 40.5 eV that was obscured previously by the large error bars.

Furthermore, we located the Ar<sup>++</sup> 2p<sup>-1</sup>3s<sup>-1</sup> states and revealed satellite lines, predicted to be of Ar<sup>++</sup> 2p<sup>-1</sup>3p<sup>-2</sup>nl

configurations [6]. Special care was taken to obtain relative intensities of the different Auger transitions. Work is now in progress to accomplish an accurate theoretical description of the observed processes.

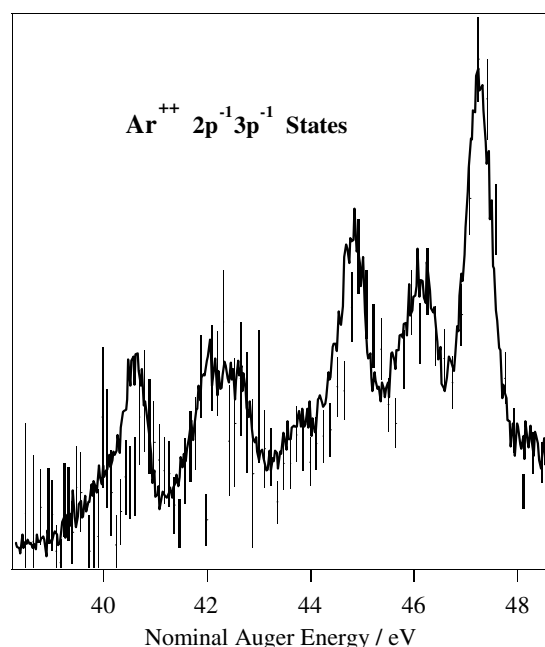


Figure: Ar L<sub>1</sub>L<sub>2,3</sub>M<sub>2,3</sub> Auger lines observed in coincidence with the 2s photoelectron, revealing the Ar<sup>++</sup> 2p<sup>-1</sup>3p<sup>-1</sup> final states. Solid line gives the present measurement, error bars are those from reference [3].

### References

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coincidence spectroscopy, threshold electron detection allows to choose singly-charged ion state, and the decay can be discussed from the kinetic energy of the coincident electron. On handling threshold electron-electron coincidence spectra, two-dimensional plot of the coincidence yields, as a function of both incident photon energy and electron kinetic energy, is the most powerful representation. The two-dimensional plot, however, has not come out yet, because of unfavorable coincidence rate had been achieved hitherto.

#### **Experimental results**

In order to get effective coincidence rate on threshold electron-electron coincidence measurement, we have developed a coincidence spectrometer which consists of a high-luminosity threshold electron analyzer and a hemispherical electrostatic electron analyzer equipped with a position sensitive detector [3]. We have applied the spectrometer to the study on double photoionization of molecular oxygen in the photon energy region of 46 - 50.5 eV. The achieved coincidence rate allows us to plot the coincidence yields in two-dimensional presentation (Fig. 1). Two electrons ejected on each double photoionization event share energy difference between incident photon energy and ionization energy for  $O_2^{2+}$ , and kinetic energy of one of electrons observed is restricted to essentially zero. The available energy increases linearly with the incident photon energy, and therefore formation of  $O_2^{2+}$  states results in diagonal lines on the two-dimensional plot. Besides the diagonal lines due to formation of  $O_2^{2+}$  states, we can see on the two-dimensional plot three or more vertical stripes running parallel to the photon energy axis. These are assignable

