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Photoion-yield spectra of Xe^{2+} in the 4d-threshold energy region

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Abstract. Photoion yields from doubly charged Xe ions were measured in the 4d-threshold energy region. The experiment was performed by a photon–ion merged-beam technique with synchrotron radiation as a light source. The measured spectrum mainly consists of strong 4d $\rightarrow$ 5p transition lines at around 57 eV, a very broad peak at around 100 eV, and the preceding strong discrete peaks lying between 79 and 90 eV. The peak position of the broad peak changes very little from that in Xe^{+} photoion-yield spectra of Sano *et al*. The enhancement of discrete peaks in the Xe^{2+} spectrum implies a further collapse of nf-wavefunctions than that of Xe^{+}.

1. Introduction

The photoabsorption spectra of heavy atoms, such as Xe, Cs, Ba and the following lanthanides, show striking broad peaks called giant resonances in the 4d-threshold energy region. Many theoretical and experimental studies have been attracted to the giant resonances because these phenomena are good examples of the appearance of many-electron effect in atoms (for a review, see Connerade 1987). There are two plausible descriptions for the giant resonances. That is, the concept of a collective motion of several electrons, and of a single-electron motion in an effective potential, i.e. one-electron model (for a review, see Bréchignac and Connerade 1994). Calculations based on the former model have been performed mainly for closed- or half-closed-shell systems (for example, see Kutzner 1996, Amusia 1996); the results are in quantitatively good agreement with some experimental works, although a lot of systems remain unsolved. Despite its quantitative discrepancy between calculations and experiments, the latter model has often been adopted and is fairly successful in explaining the qualitative variation of spectral profiles in a series of experimental results for neutral atoms (Nagata *et al* 1990), where the profile of giant resonances systematically changes with increasing atomic number, in terms of the orbital collapse of excited nf waves. In the one-electron model, for lighter species such as Xe

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the giant resonance is attributed to the transition of the 4d electron to a virtual state in the inner well of the effective double-well potential for f electrons. For heavier atoms (larger than about $Z = 57$), nf-wavefunctions start to collapse into the inner well with increasing nuclear charge, which greatly affects the effective potential, and subsequently, 4d $\rightarrow$ nf discrete transitions appear in the spectra.

We are interested in how the giant resonances behave in the case of photoionization of atomic ions. The effective potential can be varied by means of removing electrons from atoms or ions as well as by changing the atomic number Z. One would expect the validity of the single-electron concept even for the atomic ion cases and/or to gain better understanding of the giant resonance (in other words, electron correlation) mechanism. In contrast to the neutral atom cases, there have been only a few experiments for the 4d photoionization of atomic ions so far. For Ba^+, Ba^{2+} (Lucatorto *et al* 1981) and I^+, I^{2+} (O'Sullivan *et al* 1996), the evolutions of photoabsorption spectra as a function of charge state are similar to each other. The discrete transitions begin to appear on the lower-energy side of the giant resonance from a singly charged target for both the Ba and I ion cases. This appearance of discrete transitions could be explained by the concept of orbital collapse. For Xe ions, the photoion-yield spectroscopy of Xe^+ has been reported by our group (Sano *et al* 1996). Some discrete peaks just below the giant resonance were assigned to the 4d $\rightarrow$ np, nf transitions due to the partial collapse of nf-wavefunctions.

2. Experimental

The experiment was performed on the beam line BL-3B at the Photon Factory using synchrotron radiation monochromatized with a 24 m spherical grating monochromator (Yagishita *et al* 1991). The experimental set-up was almost the same as described previously (Sano *et al* 1996) but with a few modifications: the ion–photon interaction cell was positioned closer to the parallel-plate charge analyser in order to reduce background noise arising between them, and the ion detectors in the charge analyser were relocated so that Xe^{3+} and Xe^{4+} photoionized from Xe^{2+} ions were detectable. Xe^{2+} ions were created with an electron-impact-type ion source and mass analysed by the Wien filter. Although we did not analyse the internal states of the ions, created Xe^{2+} ions are considered to include $^3P_{0,1,2}$, $^1D_2$ and $^1S_0$ terms. Since the state populations in Ar^+ and Kr^+ ions produced by the same type of ion source as ours were found to conform to the statistical weight (Kobayashi *et al* 1981, Itoh *et al* 1981); similarly, in our Xe^{2+} case the population of the $^3P_{0,1,2}$, $^1D_2$ and $^1S_0$ terms would be 9:5:1. The ion beam was deflected by 90° and merged with the photon beam in a 150 mm interaction region. The primary ions and photoionized product ions were separated by the electrostatic parallel-plate analyser. In the present experiment, two products, Xe^{3+} and Xe^{4+}, were detected simultaneously by channel electron multipliers combined with ion–electron converter plates. The primary current of the Xe^{2+} ion, typically 6 nA, was measured by the Faraday cup at the end of the parallel plate.

Photon energy was varied in the region of 4d photoionization from 52 to 150 eV with energy resolution $E/\Delta E \sim 160$ and $E/\Delta E \sim 250$ for the low- and high-resolution case, respectively. The intensity of the photon beam was monitored with a gold-coated photodiode. The photon flux is estimated to be of the order of 10^{11} s^{-1}. The energy scale was calibrated in the same manner as described in Sano *et al* (1996). The accuracy of the energy scale is estimated to be less than 0.2 eV.

In spite of modification on the interaction-region position, the signal-to-noise ratio was about 0.25 at best. The signal count rate was typically 150 s^{-1} at a maximum.

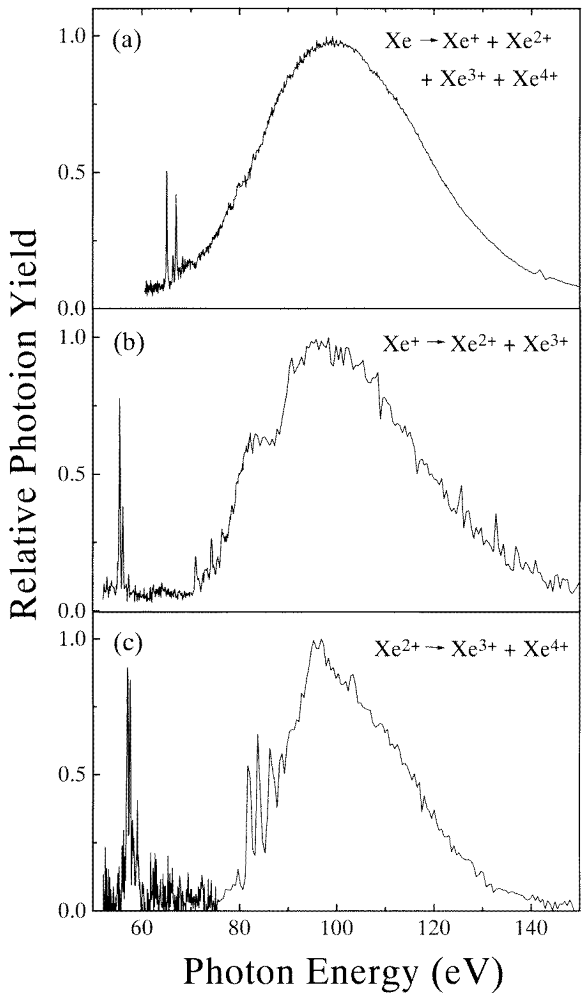

Figure 1. Photoion-yield spectra of Xe, Xe^+ and Xe^{2+}. Intensities are normalized at the maximum data points. (*a*) Total photoion yield from neutral Xe (Nagata *et al* 1990). (*b*) The sum of Xe^{2+} and Xe^{3+} yields from Xe^+ ions. The spectrum above 82 eV was measured by Sano *et al* (1996). The measurement below 82 eV was carried out with an energy resolution of $E/\Delta E \sim 240$ (to be published). (*c*) The sum of measured Xe^{3+} and Xe^{4+} yields from Xe^{2+} targets with the low-energy resolution of $E/\Delta E \sim 160$. Data points were acquired in 0.1 eV steps between 52 and 75 eV, and in 0.5 eV steps above 75 eV.

3. Results and discussion

For a comparison, the previous results for the photoionization of Xe and Xe^+ are shown together in figures 1(*a*) and (*b*), respectively. Figure 1(*c*) shows the sum of relative photoion yields of Xe^{3+} and Xe^{4+} from Xe^{2+} ions obtained in the measurements of low-energy resolution. Figure 2(*a*) shows the detailed structure around 90 eV obtained with the higher energy resolution. The structure observed below 60 eV was measured in detail with the low-energy resolution, and the result is shown in figure 2(*b*). Higher charged products than Xe^{4+} would be negligible in the energy region of present measurement because the triple ionization of Xe^{2+} is estimated to be about 140 eV from an electron-impact experiment by Müller *et al* (1984). Therefore, the sum of the Xe^{3+} and Xe^{4+} yield is equivalent to the total photoion yield from Xe^{2+} below 140 eV. Moreover, since radiative relaxations of 4d-hole states are very minor processes, the obtained spectra can be regarded as the photoabsorption spectra. The high-resolution spectrum was acquired in the energy region between 75 and 100 eV. It can be found in the high-resolution spectrum that each prominent peak preceding the broad peak consists of several smaller discrete peaks. Energies of some prominent peak

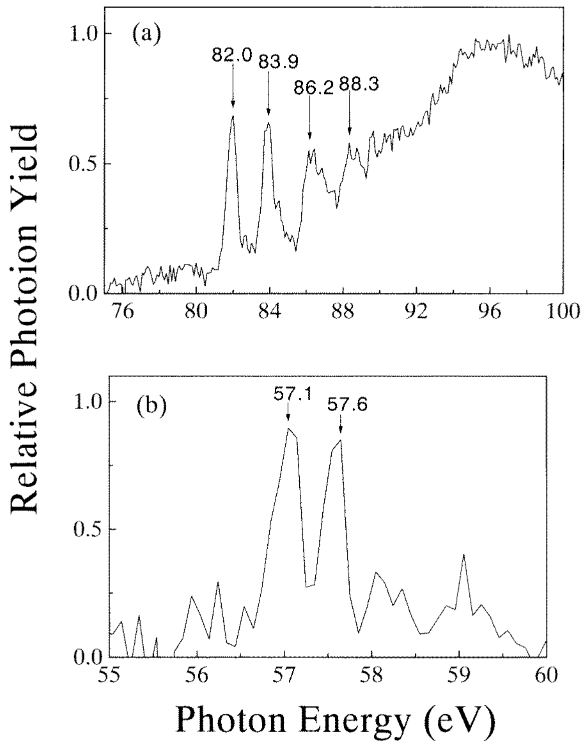

Figure 2. The detailed structures in the photoion-yield spectrum of Xe^{2+} (see figure 1(*c*)). Data points were acquired in 0.1 eV steps. The energies of several prominent peaks are determined in eV as shown above the arrows. (*a*) The spectrum around 90 eV, obtained with high-energy resolution. (*b*) The spectrum below 60 eV with low-energy resolution.

positions are determined and shown with the spectrum in figure 2(*a*). Statistical errors of yield intensities were evaluated to less than 5% for both resolutions in the energy region from 90 to 100 eV.

In figure 1(*c*) one can easily find three striking features: very strong sharp peaks just below 60 eV; a series of discrete peaks between 79 and 90 eV; and the strong broad peak at around 100 eV. The sharp peaks at around 55–60 eV have been assigned elsewhere (Koizumi *et al* 1997) to 4d $\rightarrow$ 5p transition lines by the multiconfiguration Dirac–Fock (MCDF) calculation. This transition never occurs in the 4d photoabsorption of neutral Xe (Haensel *et al* 1969) because the 5p state is fully occupied for neutral Xe. It is worth comparing the photoion-yield spectrum of Xe^{2+} with that of Xe^{+} from the viewpoint of orbital collapse and effective potential. In the Xe^{+} spectrum (see figure 1(*b*)) there exist several discrete peaks between 70 and 80 eV and a big shoulder above the 4d thresholds between 80 and 90 eV, which are not observed in the photoabsorption spectra of neutral Xe. Sano *et al* (1996) assigned discrete peaks to 4d $\rightarrow$ np and 4d $\rightarrow$ nf transitions due to the partial collapse of the f orbitals and suggested the shoulder as the 4d $\rightarrow$ εf giant resonance. On the other hand, in the Xe^{2+} spectrum, the strong discrete peaks take the place of the shoulder at the corresponding position just below the broad peak. This spectrum evolution implies the further collapse of nf waves with increasing effective charge, like Ba and I ion cases. The discrete peaks would be attributed to the 4d $\rightarrow$ np, nf transitions although we finally failed in our attempt to assign the discrete peaks and the broad peak of the Xe^{2+} spectrum by the MCDF calculation. For the broad peak, the width in the Xe^{2+} spectrum looks slightly narrower than that in Xe^{+} if the contribution of the discrete peaks is subtracted. This could be explained by the one-electron model because the 4d $\rightarrow$ εf transitions become discrete-like due to the collapse with the increase of effective charge. One can realize that the positions of the broad peaks change little between the Xe^{+} and Xe^{2+} spectra. This tendency is also observed in the spectra of Ba and I ion targets. In terms of effective potential, the position of the broad peak is expected to shift towards the higher energy side because the threshold energy of photoionization becomes higher with increasing ionicity. In fact, this consideration is consistent with the results of neutral atoms where peaks of the 4d-giant resonance move toward the higher energy side as Z increases (Nagata *et al* 1990, Richter *et al* 1989 and references quoted therein). The unchanging broad peak positions in the ion target cases are hardly explained by the simple one-electron model.

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