Construction of VUV photoelectron imaging apparatus with He(I) light source

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Introduction

Polyatomic molecules possess multiple electron orbitals that are nearly degenerate in energy, which leads to overlap of bands in photoelectron spectra. Spectral assignments of such overlapping band systems have been rather difficult. There have been two reasons for this difficulty. One is that most of the ultraviolet photoelectron spectra of polyatomic molecules so far have been measured at room temperature, and hot bands of low-frequency vibrations made the spectra congested: Oku et al. [1] have recently demonstrated that jet-cooled pyrazine exhibits much finer structure in the photoelectron spectrum than its vapor at room temperature, demonstrating the benefit of jet cooling in observation of polyatomic spectra. The other reason is that photoelectron angular anisotropy has not often been measured, although it is of great assistance for assigning those bands. Oku et al. have employed a hemispherical energy analyzer and integrated the signal for hours to observe jet-cooled spectra, while we expect photoelectron imaging will allow much more efficient data acquisition and direct visualization of photoelectron angular anisotropy as a function of photoelectron kinetic energy.

In the present study, we constructed a photoelectron imaging spectrometer with a He(I) light source to observe speed and angular distributions of photoelectrons and analyze the complex spectra of polyatomic molecules. This is also a part of our efforts for future photoelectron imaging experiments using a vacuum ultraviolet free electron laser (VUV-FEL). The FEL wavelength is 50-60 nm that almost coincide with the He(I) line, and VUV

experiments always encounter the influence of secondary–electron background due to its high photon energy. Thus, the present work also serves as a test for photoelectron imaging using VUV-FEL.

Experiment

The cross section of the photoionization chamber is shown in Fig. 1. The He discharge lamp, a continuum VUV light source, has a polarization degree of 88% and a photon flux of 1.0 \times 10¹³ photons/sec at 21.22 eV (58.4 nm). The He(I) radiation generated by a



FIG. 1. The cross section of the photoionization chamber; the elements indicated are: (1) a polarizer of He discharge lamp, (2) mu-metal shielding, (3) a stacked electrode, (4) a MCP with a phosphor screen, (5) a CCD camera.

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discharge lamp is originally unpolarized, with which photoelectron angular anisotropy can be determined by taking the propagation direction of light as the reference axis. However, it makes difficult to determine small anisotropy parameters that are typical of photoionization from valence orbitals. Therefore, we have polarized the radiation by using a set of mirrors, as reported by Heinzmann and coworkers [2]. Photon flux was reduced to 4.0×10^{11} photons/sec and the VUV light is gently focused onto the atomic beam. The apparatus consists of three vacuum chambers, a molecular beam source, a buffer and a photoionization chamber. Argon gas was continuously expanded into vacuum through a 25 µm nozzle at a stagnation pressure of 20 bar, and the typical operation pressure in the photoionization chamber is 1.3×10^{-7} Torr. At 380 mm downstream from the nozzle, Ar atomic beam with 6 mm diameter were photoionized by He lamp. The photoelectrons are accelerated in an electric field perpendicular to both the propagation directions of an atomic beam and the VUV light, and detected by an imaging detector (768×572 pixels). The photoelectron spectrometer is covered with a mu-metal shields placed inside of the photoionization chamber to avoid terrestrial magnetic field.

Results

We have integrated two images with and without the Ar atomic beam for 4 min and obtained the difference image as shown in Fig. 2. The background subtraction was necessary, since large background photoemission from the chamber was observed. We have traced back the cause of such photoemission and concluded that it is due to photoemission from the electrodes illuminated by scattered light. Currently, we are planning modification of the apparatus by FIG. 2. Photoelectron images of Ar at 21.22 eV



minimizing the light spot diameter in the interaction photon energy with background subtraction.

region and/or enlarging the electrode gap in which light passes through. With the current signal count rate, 100 min integration time would be necessary for measuring 5% sample diluted in a carrier gas, which is not satisfactory as performance of the imaging apparatus. We expected $\sim 2.8 \times 10^6$ electron counts per second (cps) in our experiment, while it seems this count rate is not accomplished at this point. A possible cause of this problem is that the photon flux is smaller than prediction for poor transmission through the polarizer. In order to figure out this problem and enhance the efficiency of data acquisition, we are considering to remove the polarizer.

Our current energy resolution of $\triangle E/E = 3.5$ % is also insufficient to resolve a spin-orbit splitting ~178 meV between ${}^{2}P_{3/2}$ and ${}^{2}P_{1/2}$ states of Ar⁺. This however is easily improved to \triangle E/E = 0.16 % by using our new detection system using 2048×2048 pixel CCD camera and subpixel center-of-gravity calculations.

Reference

[1] Oku M. et al., J. Phys. Chem. A 112, 2293 (2008).

[2] Schonhense G. et al., J. Phys. E.: Sci. Instr. 16, 74 (1983).