Development of the Fast Camera System for High-Precision Photoelectron Imaging

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Recent development of photoelectron circular dichroism (PECD) studies significantly relies on photoelectron VMI (Velocity Map Imaging) measurements as well as circularly polarized radiation generated with undular light sources, which is especially advantageous for one-photon ionization in the vicinity of the molecular photoionization threshold region. On the other hand, multi-photon ionization based on laser-based photoionization has also been subject to the PECD research. Molecular electronic chirality can be further understood in comparison with the PECD data of one-photon and multi-photon ionization.[1, 2]

We have so far used the camera system of the VMI apparatus common for laser and synchrotron light sources, although the synchrotron measurements and laser-based measurements were executed in UVSOR and the Hiroshima University laboratory, respectively. Besides the wavelength regions, the critical difference of the UVSOR beam line and the pulsed laser system regarding our PECD study lies in the repetition rate and the photoelectron count rate. The pseudo-continuous radiation provided by the UVSOR beam line can generate photoelectrons with more than 100 kcps (counts per second) under our experimental conditions. The pulsed laser system runs in a 10 Hz repetition rate, where a single laser shot provides less than 100 photoelectrons corresponding to the 10 cps. The photoelectron count rate should be reduced to a rate so low that the light spots due to a single photoelectron arrival at the detector do not overlap each other, otherwise, the accurate photoelectron distributions are not obtained. The higher count rate, ensuring the better signal statistics, is especially critical in PECD measurements, where the difference of independently measured image data is analyzed. In place of the conventional frame-transfer type camera (1000 x 1000 pixels, 33 frames/s rate), which was matched with the low repetition pulsed laser, we introduced an event-driven type camera, whose nominal signal transfer rate is sufficiently fast to fully capture the signals provided by the UVSOR beam line.

The performance of the event-driven camera was examined with the BL5B beam line. The camera equipped with the VMI apparatus was linked with the local Ethernet LAN to store the event data. The sensitivity, temporal response, and transfer rate were methyl oxirane at 120 nm (Fig. 1). Although the photoelectron spot images were overlapped in a frame-type camera display (Fig. 1(a)-(c)), the stored event data were confirmed to be isolated from each other by the fast time resolution of 1 μs (Fig. 1(d)). We developed a program code to recover the image data from the event data. The results indicated the good linearity of the image intensity up to the photoelectron count rate of 500 kcps, which is nearly the upper limit of the detector damage.

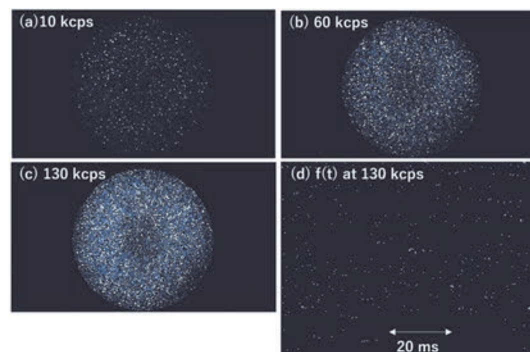

Fig. 1. Photoelectron image data measured with the event-driven camera equipped with the synchrotron radiation light source. The light spots due to photoelectron hitting on the imaging detector (1280 x 720 pixel size) accumulated in 33 camera frames are shown at a photoelectron count rate of (a) 10 kcps, (b) 60 kcps, and (c) 130 kcps. A time profile of the event-camera output in a particular pixel column of (c) is shown in (d).

[1] H. Kohguchi *et al,*, UVSOR Activity Report **50** (2023) 138.
[2] H. Kohguchi *et al*., UVSOR Activity Report **50** (2022) 136.

Transmission Spectra of Amorphous Chalcogenide Thin Films in the Vacuum Ultra-Violet Region

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Amorphous chalcogenide semiconductor materials, such as amorphous $GeSe_2$, amorphous As_2Se_3 and amorphous Se etc., show a variety of photoinduced phenomena. Therefore, these materials are expected as materials for optoelectronic devices. A lot of work has been done on the photoinduced phenomena of these amorphous semiconductor materials and various mechanisms have been proposed for these photoinduced phenomena [1]. Among those phenomena, the most prominent phenomena are the so-called photodarkening and photobleaching, which are parallel shifts of the optical absorption edge to lower and higher energy sides, respectively, after irradiation with light whose energy corresponds to the optical bandgap. These darkened and bleached states are removed by annealing near the glass-transition temperature. The X-ray diffraction and the volume change of the films before and after irradiation with bandgap light suggest that these phenomena result from the change in the local structure of the amorphous network. However, the details of the mechanisms are still unknown. We are interested in the changes in the optical properties at the higher energy regions. To obtain a wide knowledge of photoinduced phenomena, it is necessary to investigate the photoinduced effects over a wide energy region. In the previous reports, we reported the annealing effect on the VUV transmission spectrum of amorphous selenium [2]. In this report, we focus on the a-$GeSe_2$ thin film that exhibits photobleaching, and report on the investigation of its fundamental optical properties by measuring the VUV transmission spectrum.

Samples used for the measurement of the VUV transmission spectra were thin films of amorphous $GeSe_2$ deposited on a thin aluminum film by conventional evaporation technique. The sample thickness was about 180 nm. An aluminum film of the thickness of 200 nm was used to eliminate the higher order light from the monochromator in the VUV region. These measurements were carried out at room temperature on the BL5B beam line of UVSOR. The spectrum was measured with a silicon photodiode as a detector. Two 1.5mm diameter pinholes were inserted between the monochromator and sample to remove stray light. The intensity of the VUV light was monitored by measuring the TPEY of a gold mesh. The positions of the core levels for the samples were calibrated with reference to the 2p core level absorption of the aluminum film.

Figure 1 shows the VUV transmission spectrum of the amorphous $GeSe_2$ thin film in the wavelength range of 18 to 24 nm. For comparison, the VUV transmission spectra of amorphous As_2Se_3 and amorphous Se are also shown in the figure. As shown in the figure, each spectrum is very broad and multiple shoulders are observed. Main absorption peaks around 22nm correspond to the 3d core level of Se atoms. The absorption spectra observed in amorphous As_2Se_3 and amorphous Se are roughly consistent with the previous report [3]. Although the wavelength resolutions in the spectral measurements are all the same, the spin-orbit splitting of the $3d_{5/2}$ and $3d_{3/2}$ levels of Se atoms is clearly resolved in amorphous $GeSe_2$ and amorphous As_2Se_3, while it is not clearly resolved in as-deposited amorphous Se. The origin of broad spectra and several peaks is not clear. I think that these origins are related to the local structures of the amorphous network. The detailed experiments and analysis will be done in the next step. More detailed experiments are necessary to clarify the origin of the VUV transmission spectra.

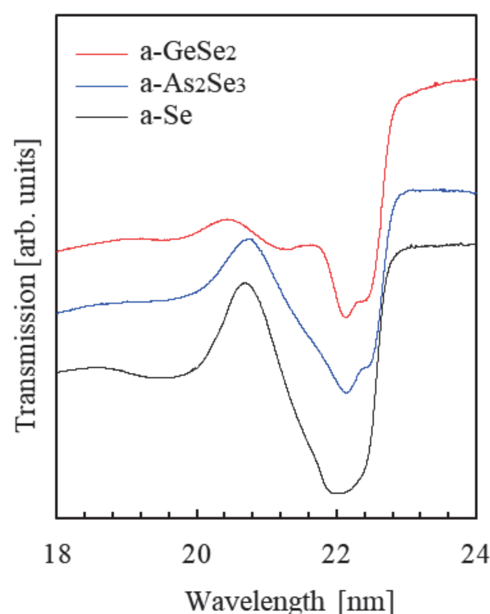

Fig. 1. VUV transmission spectra of amorphous $GeSe_2$, amorphous As_2Se_3, and amorphous Se thin films.

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[2] K. Hayashi, UVSOR Activity Report **50** (2022) 152.
[3] J. Bordas and J. B. West, Phil. Mag. **34** (1976) 501.

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Reflectance of La/B_4C Reflective Multilayers

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A wavelength in the soft X-ray (SX) region around 6.x nm has been proposed as a candidate for next-generation semiconductor lithography [1]. In this wavelength range, light penetration is significantly limited, necessitating the use of reflective optics based on multilayer structures. These multilayers typically consist of alternating layers of two materials that exhibit both low absorption and a large difference in refractive index. Ensuring a sharp interface between these materials is paramount for preserving the index contrast. Given that the periodic thickness of the film is approximately half the target wavelength at near normal incidence, the influence of the interface becomes increasingly critical as the wavelength decreases.

Currently, LaN/B-based multilayers are among the most reflective near 6.x nm [1]. These are fabricated via reactive magnetron sputtering using N_2 gas. However, the reactive gas can also interact with B, forming BNx compounds at the LaN/B interface and thereby degrading reflectance. To address this issue, we investigated La/B_4C multilayers as a promising alternative that eliminates the need for reactive sputtering (Fig. 1). B_4C has a negative enthalpy of formation and is thermodynamically stable in its standard state [2], while La is defined in its most stable phase [3]. This suggests that the La/B_4C pair can form stable, sharply defined interfaces in thin-film form.

The multilayers were deposited using ion beam sputtering at acceleration voltages of 1.0, 1.5, and 2.0 kV. X-ray reflectometry revealed the corresponding period lengths to be 3.63 nm, 3.52 nm, and 3.80 nm, respectively, indicating that ion energy primarily affects the interfacial structure.

Figure 2 presents preliminary reflectance spectra near 6.x nm for samples deposited at 1.0 kV and 2.0 kV. The observed variations in reflectance are believed to result from differences in interface quality induced by the ion energy.

In summary, the La/B_4C multilayer system demonstrates promising reflectance performance without the complications associated with reactive sputtering. The correlation between ion energy and interface sharpness highlights a critical parameter for further optimization. Ongoing and future work will aim to systematically improve reflectance by refining deposition conditions and enhancing interface control in La-based multilayers.

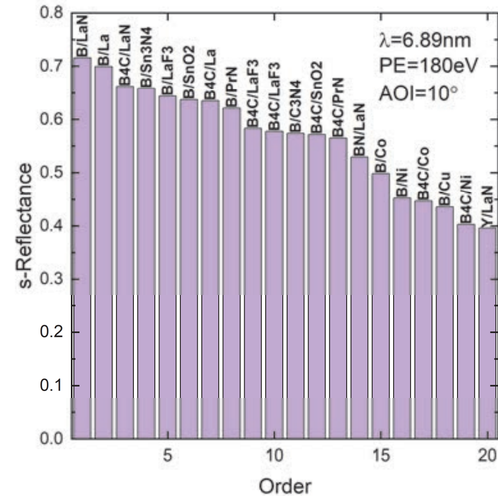

Fig. 1. A comparison of s-polarized reflectance for each material pair, with the horizontal axis indicating rank and the vertical axis displaying s-polarized reflectance. It is notable that the maximum value is B/LaN.

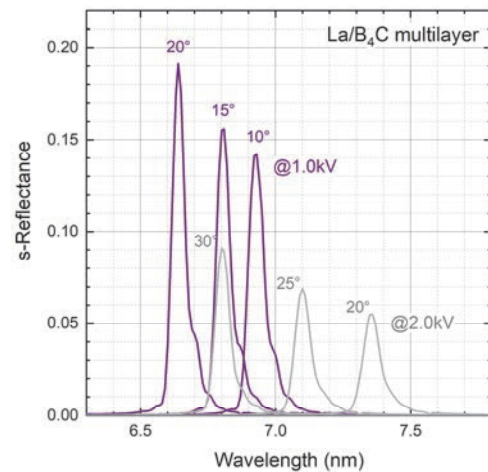

Fig. 2. A comparison of s-polarized reflectance for La/B_4C multilayers measured in the near normal incidence geometry (AOI: 10° – 30°).

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