CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2014/7/19 承認者 Approver Masayasu Takeda 提出日 Date of Report 2014/6/18
課題番号 Project No. 2013B0159 実験課題名 Title of experiment In-situ neutron reflectivity measurements for photodoping of silver in Ge-chalcogenide films III 実験責任者名 Name of principal investigator Yoshifumi Sakaguchi 所属 Affiliation CROSS	装置責任者 Name of Instrument scientist Masayasu Takeda 装置名 Name of Instrument/(BL No.) *SHARAKU* (BL17) 実施日 Date of Experiment Feb 26 21:00 – Feb 28 10:00, 2014

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）
Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

Ag/$Ge_{33}S_{67}$ films

2. 実験方法及び結果 （実験がうまくいかなかった場合、その理由を記述してください。）
Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

Experimental method:

In order to clarify the change on the depth profile in Ag/amorphous (a-) Ge chalcogenide films by silver photo-doping, (silver diffusion) we investigate on the neutron reflectivity of Ag/a-Ge-S films under a light illumination. In the previous experiments, we measured neutron reflectivity profiles of Ag/a-$Ge_{40}S_{60}$ films (2012A) and Ag/a-$Ge_{20}S_{80}$ films (2013A). In the present experiment, we measure neutron reflectivity profiles of Ag/a-$Ge_{33}S_{67}$ films and try to find a compositional dependence on silver photo-diffusion.

In the experiment, we used a xenon lamp unit (MAX-303, ASAHI SPECTRA) as an excitation light source. The illumination area on the film was 25mm x 25mm. The measurements were performed on BL17 (*SHARAKU*) with unpolarized neutron beam mode. Full sets of neutron reflectivity profiles up to Q= 0.08 $Å^{-1}$ before and after an exposure to light from the xenon lamp have been obtained from reflected time-of-flight data at two angles. Time-resolved reflectivity profiles (Q < 0.04 $Å^{-1}$) under the exposure were obtained at one fixed angle of a detector.

Experimental results:

Fig.1 shows the neutron reflectivity of a-$Ge_{33}S_{67}$ 2000Å /Ag 500 Å / Si substrate (the thicknesses were given by a quartz crystal unit in a thermal evaporator) before a light exposure. The inset table summarizes the parameters used in the fitting. As shown in the table, we confirmed that there are only $Ge_{20}S_{80}$ and Ag layers,

without a reaction layer. After starting a light exposure, the total reflected beam intensity changed with time as shown in the inset in Fig.2, and the reflectivity profile changed with time as shown in Fig.2. By 1600s, there are changes in the amplitude and the width of the fringes, suggesting a development of silver diffusion to be one homogeneous layer. However, at 1600s, the reflected beam intensity suddenly drops. In addition, total reflection region disappears in the reflectivity profile measured after the 70min light exposure, as shown in Fig.3. In fact, we observed bubbles-like inhomogeneous surface with an optical microscope (the inset in Fig.3). The visible roughness on the surface coincides with the disappearance of total reflection on the neutron reflectivity profile. This type of change was also observed for $Ge_{33}S_{67}$ 1500Å /Ag 500 Å / Si substrate film. However, it was not observed for Ag 500 Å /$Ge_{33}S_{67}$ 1500 (2000) Å / Si films, and films with other Ge compositions (20 and 40%). There seems to be a specific condition to induce the change. Since this big change at 1600s occurs after further silver diffusion by the light exposure, this is attributed to a photo-structural change on homogeneous Ag-$Ge_{33}S_{67}$ compound. Considering the macroscopic inhomogeneity, we infer that phase separation occurred on the illuminated surface.

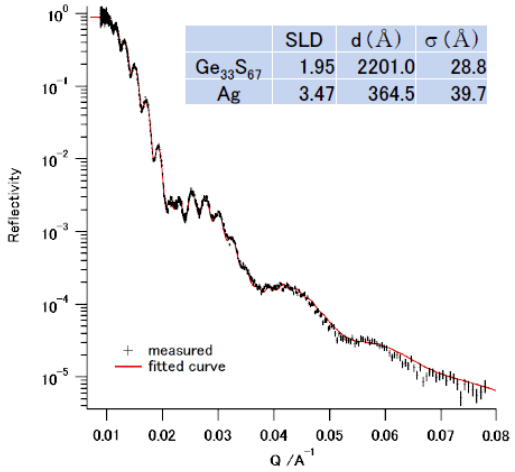

Fig.1 Neutron reflectivity profile before a 70 min exposure to the xenon lamp. SLD: scattering length density (x 10^{-6} $Å^{-2}$)

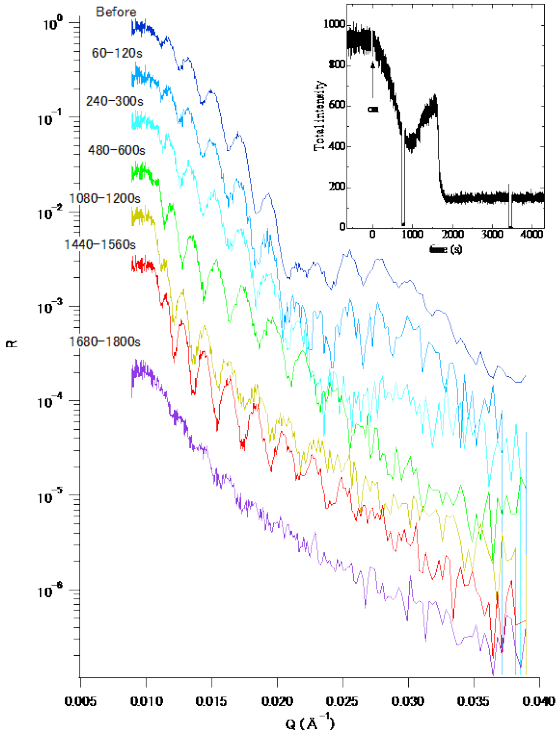

Fig.2 Time evolution of neutron reflectivity of $Ge_{33}S_{67}$ 2000Å/ Ag 500 Å / Si substrate film under light illumination. Inset: time variation of the total reflected intensity. Exposure time: 0-4200s. Neutron beam supply is off at the time when the total intensity is about zero

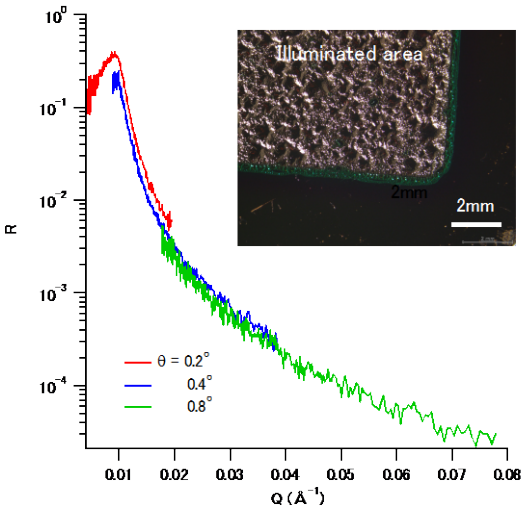

Fig.3 Neutron reflectivity profile after the 70 min exposure. Inset: Photograph of the film after the exposure.