TOKAI CROSS **Experimental Report** J-PARC	Date of Approval 2013/12/10 Approver Masayasu Takeda Date of Report 2013/09/06
課題番号 Project No. 2013A0203 実験課題名 Title of experiment In-situ neutron reflectivity measurements for photodoping of silver in Ge-chalcogenide films Ⅱ 実験責任者名 Name of principal investigator Yoshifumi Sakaguchi 所属 Affiliation CROSS	装置責任者 Name of responsible person Masayasu Takeda 装置名 Name of Instrument/（BL No.） SHARAKU（BL 17） 実施日時 Date and time of Experiment 2013/4/10-4/12 2013/5/20-5/22

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/$Ge_{20}S_{80}$ 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, we investigate on the neutron reflectivity of Ag/a-Ge-S films under a light illumination. In 2012A, we have measured for Ag/a-$Ge_{40}S_{60}$ films. In 2013A, we have measured for Ag/a-$Ge_{20}S_{80}$ films, which are supposed be more reactive and show faster change.

　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. Neutron reflectivity measurements have been made on BL17 (SHARAKU) with unpolarized neutron beam mode. We measured neutron reflectivity of the sample before and after making a light exposure in the Q-region up to 0.08 $Å^{-1}$. The light was exposed on the film for 60 minutes and neutron reflectivity measurements have also been made during the light exposure, fixing the angle of a detector.

Experimental results:

　Fig.1 shows the neutron reflectivity of Ag 500 Å / a-$Ge_{40}S_{60}$ 1500Å / Si substrate (the thicknesses were given by a quartz crystal unit in a thermal evaporator) before and after a light exposure. The black dots show the experimental data while the red solid curve shows the result of fitting. The inset table summarizes the

parameters used in the fitting. As shown in the table, before making a light exposure, there are Ag and $Ge_{20}S_{80}$ layers, and, in addition, there is silver-doped reacted layer between them. We infer that this 100 Å -depth-diffusion occurred thermally after thermal evaporation, due to its active reactivity. After 60-min light exposure, it is considered that the film gets to be one homogeneous Ag-doped layer and silver diffusion is completed.

Before light-on

	SDL	d (Å)	σ (Å)
Ag	3.47	376.6	20.8
Ag-$Ge_{20}S_{80}$	2.21	100.7	28.5
$Ge_{40}S_{60}$	1.7	1154.5	15.6

After light-off

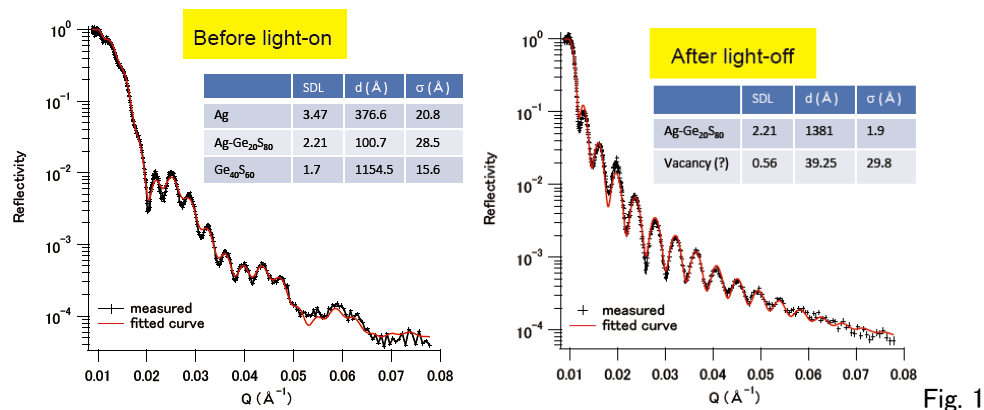|	SDL	d (Å)	σ (Å)
Ag-$Ge_{20}S_{80}$	2.21	1381	1.9
Vacancy (?)	0.56	39.25	29.8

Fig. 1

Fig.2(a) shows the time evolution of neutron reflectivity during and after stopping a light exposure. The reflectivity curve rapidly changes within 20 min. To see the details in the first 10min, we obtained time-slicing reflectivity up to 30 sec resolution from the data with event recording format and the result is shown in Fig.2(b). Although there are some noisy data points in the reflectivity curves with 30 sec resolution, we could get fundamental features in them.
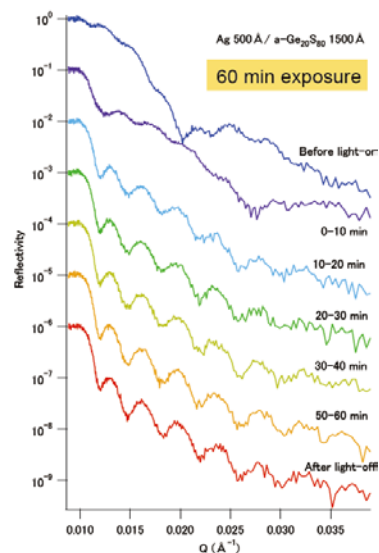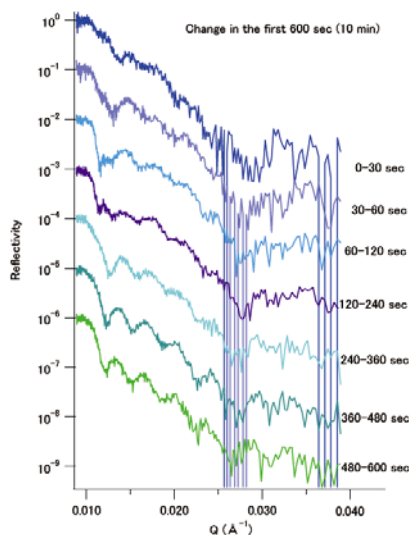

Fig.2(a)

Fig.2(b)

Fourier transformation has been applied for the above original reflectivity data. From the results of static measurements with wide Q-range up to 0.08 Å $^{-1}$, we could confirm the number of layers before and after making a light exposure, which is related to the number of peaks in the Fourier transformed curve. From the results of transient measurement with narrower Q-range up to 0.04 Å $^{-1}$, and the plots of the position and the height of the first peak, we observed (1) rapid change within 2 min (2) the following peak decay (3) a saturation after 20 min. Possible picture of the silver photo-diffusion based on this model-free analysis has been shown in the conferences listed in the following section.