実験報告書様式（一般利用課題・成果公開利用）

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CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 承認者 Approver 提出日 Date of Report
課題番号 Project No. 2017A0226 実験課題名 Title of experiment Study on the reaction rate of silver photo-diffusion into amorphous Ge-chalcogenide by in situ neutron reflectivity measurement 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 Kazuhiko Soyama 装置名 Name of Instrument/(BL No.) SHARAKU (BL17) 実施日 Date of Experiment 10:00 April 13 – 10:00 April 16, 2017 10:00 June 19 – 9:00 June 21, 2017 9:00 June 22 – 10:00 June 23, 2017

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.

Introduction: Interestingly, silver dissolves and diffuses into amorphous chalcogenides by a light exposure and we are trying to clarify the *in situ* changes of the layer structure by time-resolved neutron reflectivity measurement. Recently, we measured the neutron reflectivity of $Ge_{33}S_{67}$/ Ag/ Si [1] and found that the reaction rate markedly depended on the thickness of the chalcogenide layer.

Experimental method: In this experiment, we intended to clarify the chalcogenide layer thickness dependence by measuring neutron reflectivity of several films with different chalcogenide layer thickness and different order of the film deposition as shown in Table 1. Static neutron reflectivity of the samples was measured before and after the light exposure for 60 min, fixing at three angles, to cover wider Q-range up to 0.08 $Å^{-1}$. Transient neutron reflectivity of the samples was measured fixing at one angle, in which Q-range is covered up to 0.04 $Å^{-1}$. Proton beam power in the present experiment was 150kW.

Ag 500 Å/ $Ge_{20}S_{80}$ 1250 Å / Si substrate	$Ge_{20}S_{80}$ 1250 Å / Ag 500 Å/ Si substrate
Ag 500 Å/ $Ge_{20}S_{80}$ 1500 Å / Si substrate	$Ge_{20}S_{80}$ 1500 Å / Ag 500 Å/ Si substrate
Ag 500 Å/ $Ge_{20}S_{80}$ 1750 Å / Si substrate	$Ge_{20}S_{80}$ 1750 Å / Ag 500 Å/ Si substrate
Ag 500 Å/ $Ge_{20}S_{80}$ 2000 Å / Si substrate	$Ge_{20}S_{80}$ 2000 Å / Ag 500 Å/ Si substrate

Table 1. List of the samples for which neutron reflectivity was measured in the present experiment. The indicated compound and thickness are the planned ones in the preparation.

2. 実験方法及び結果（つづき） Experimental method and results (continued)

Experimental results:

Fig.1(a) shows the neutron reflectivity profiles of Ag 500 Å/ $Ge_{20}S_{80}$ d Å (d = 1250, 1500, 1750, 2000 Å) before the light exposure and Fig.1(b) shows the SLD profiles, which are used as models for the calculated neutron reflectivity curves in Fig.1(a) (red curve). Actually, the neutron reflectivity profiles of the samples before the light exposure could not be fitted by the SLD profile models of the Ag/$Ge_{20}S_{80}$ structure. Therefore, we measured the X-ray diffraction of the samples and found that Ag changed to Ag_2S "before" the light exposure, and that the Ag_2S reacted by the light exposure. Fig.2(a) shows the neutron reflectivity profiles of the samples after the light exposure for 60 min, and Fig.2(b) shows the SLD profiles. The results indicate that (Ag/) Ag_2S/ $Ge_{20}S_{80}$ changes to one homogeneous Ag-diffused reaction layer by the light exposure. Fig.3 shows the time variation of the intensity of the integrated reflected neutron beam. This roughly indicates the change of the neutron reflectivity. As shown in the figure, the reaction is very fast and is completed within 1 (d = 1250, 1500, 1750 Å) or 2 (d = 2000 Å) min. In the previous neutron reflectivity measurement of Ag 500 Å/ $Ge_{20}S_{80}$ 1500 Å/ Si substrate, it was found that the film changed to one homogeneous reaction layer within 20 min [2]. Therefore, the reaction in the present experiment is considered to be different from the Ag photo-diffusion in Ag 500 Å/ $Ge_{20}S_{80}$ 1500 Å/ Si substrate.

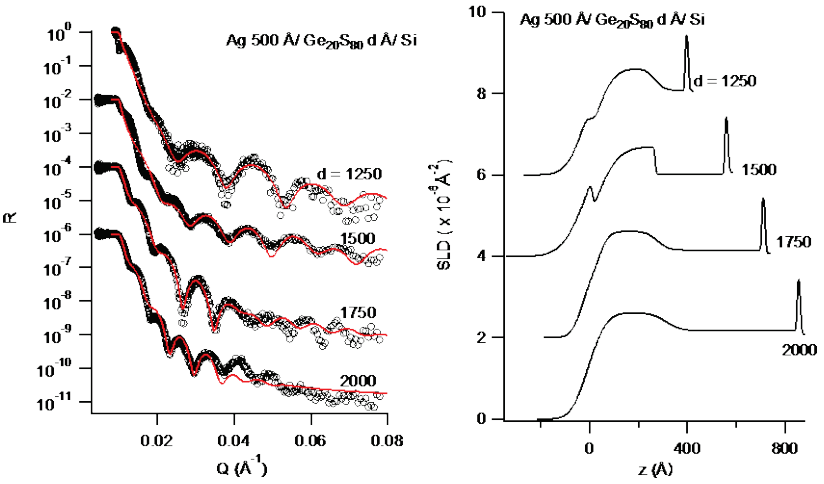

Fig.1 (a) (b)

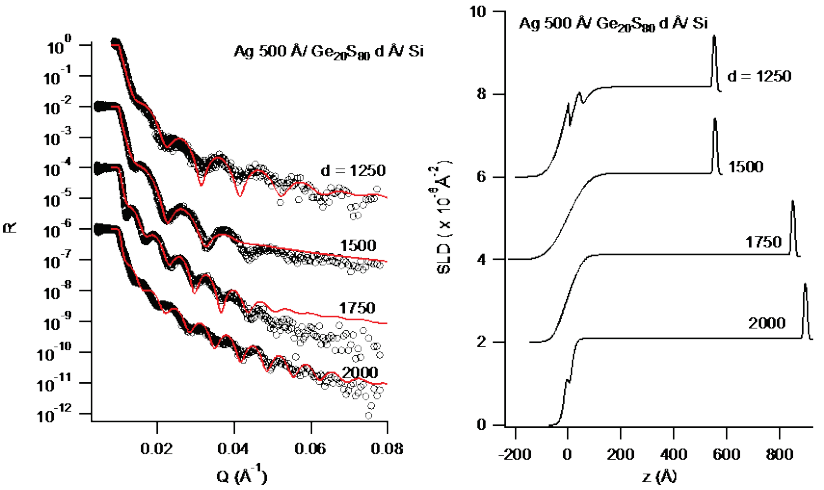

Fig.2 (a) (b)

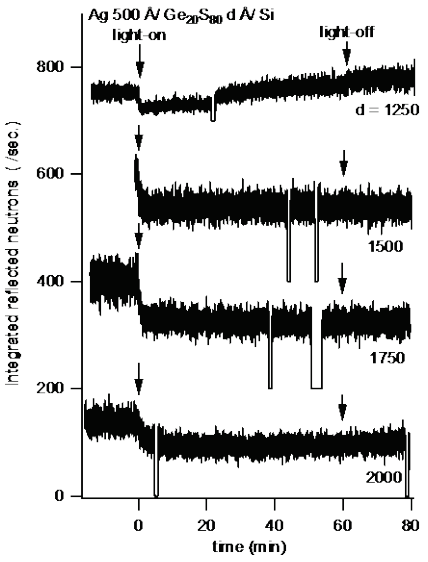

Fig. 3

[1] Y. Sakaguchi, *et al.*, J. Appl. Phys. 120 (2016) 055103.

[2] Y. Sakaguchi, *et al.*, Can. J. Phys. 92 (2014) 654.