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 2016/6/28 承認者 Approver Dai Yamazaki 提出日 Date of Report 2016/6/27
課題番号 Project No. 2014B0178 実験課題名 Title of experiment In-situ neutron reflectivity measurements for photodoping of silver in Ge-chalcogenide films V 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 March 26 10:00 –March 28 10:00, 2015 February 29 10:00 – March 2 9:00, 2016 March 31 10:00 – April 2 10:00, 2016

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_xS_{100-x} films (x=20, 33, 40)

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_xS_{100-x} films under a light illumination. In the previous experiments (2012A, 2013A, 2013B, 2014A), we measured time-resolved neutron reflectivity for the films with different Ge compositions (x=20, 33, 40), different germanium sulfide thicknesses (1500 Å and 2000 Å) and different stacking orders (Ag/Ge-S/Si substrate or Ge-S/Ag/Si substrate). From the measurements, the changes in the depth profiles by silver photo-diffusion were clarified and it was found that the kinetics markedly depends on Ge composition, germanium sulfide thickness, and the stacking order. However, we could not follow so much fast changes within 2 minutes. One problem to investigate the fast changes is the limitation of Q-range in the transient measurement. The limited Q-range leads to less accuracy in the Fourier transform at thin thickness region such as 150 Å and it makes difficult to find a proper layer structure model. In the transient measurement, we fix the incident/scattering angle. Therefore, the covered neutron wavelength range determines the Q-range. On BL17 (SHARAKU), they conventionally use neutrons from 2.2 to 9.8 Å. When the incident/scattering angle, θ, is 0.4°, this time-of-flight beam provides the highest Q of 0.040 $Å^{-1}$. However, the neutron beam spectrum can be changed by adjusting the phases of three disk choppers on the beam line and the shortest wavelength can reach at least 1.0 Å. In this case, the highest Q for θ=0.4° is 0.088 $Å^{-1}$. This can improve the accuracy in the Fourier transform. In the present experiment, we performed time-resolved neutron reflectivity measurement for Ag/ a-Ge_xS_{100-x} under light illumination using

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

neutrons from 1.0 to 8.65 Å and tried to obtain transient neutron reflectivities with wider Q-range.

Experimental results:

Fig.1 shows the time-evolution of neutron reflectivity profiles of Ag 500 Å / $Ge_{20}S_{80}$ 1500 Å / Si substrate, in which spectrum is sliced with every 10 minutes during light exposure. The proton beam power in the measurements was 408kW (460kW@25Hz). Here, the Ge-composition is that of the bulky source sample at thermal evaporation and the Ge-composition of the film was finally evaluated as $Ge_{25}S_{75}$ by neutron reflectivity measurement. As shown in the figure, the Q-range was extended to 0.05 $Å^{-1}$ or more, but the data are scattered in high Q greater than 0.035 $Å^{-1}$ and the available Q-range is limited. In Fig.2, the neutron reflectivity profiles with every 1 or 2 minutes are displayed. The highest available Q is lower than that in the spectra in Fig.1. The reason why the highest Q did not reach our expectation (theoretically, it should be 0.088 $Å^{-1}$) is the weak signals at high-Q region. Of course, neutron reflectivity decreases exponentially with increasing Q, and the observable Q-range is limited by the dynamic intensity range of the instrument. However, in the present case, the roughness of the surface layer was large (47 Å) and it accelerates the decay. As a result, Q-range was not extended to our expectation, such as 0.05 $Å^{-1}$, which could improve the accuracy in Fourier transformation. Overall, it was found from the measurements that the sample preparation is very important factor to get data with wide Q-range.

Although Q-range was not extended to our expectation, the obtained neutron reflectivity profiles had enough quality to analyze. Using the experimental and analyzed data, we could present the results as shown in the following publication lists.

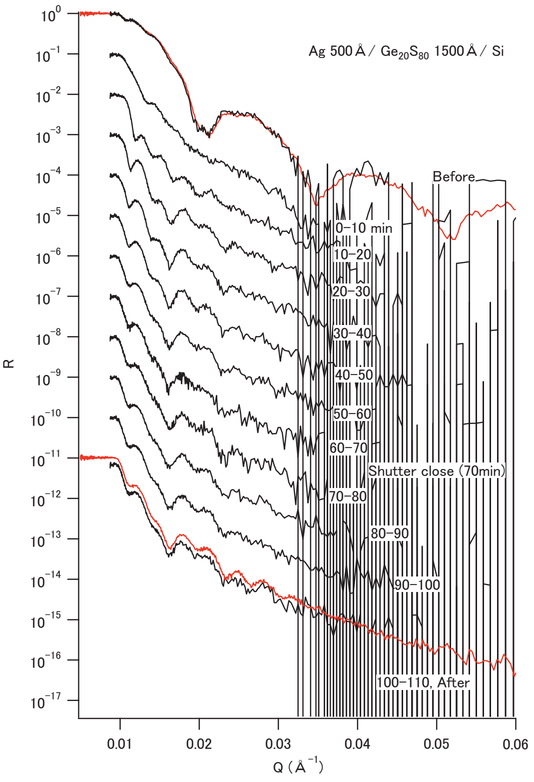

Fig.1

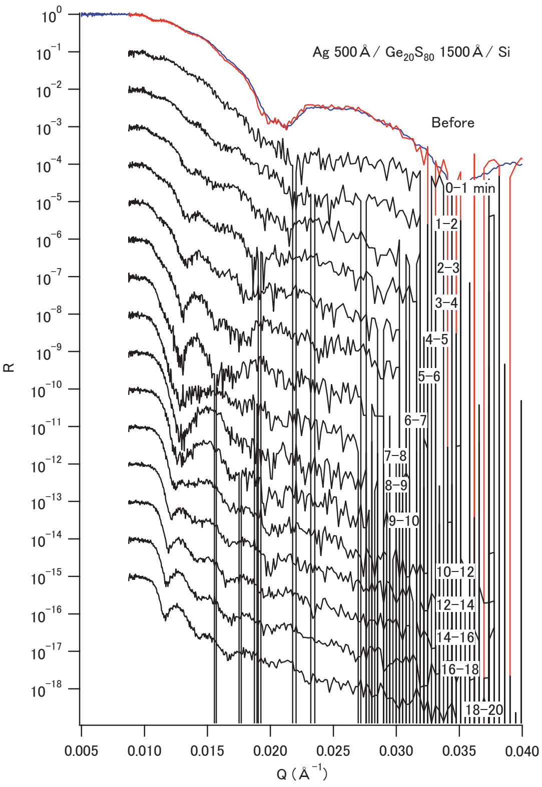

Fig.2