Investigation of photodoping process in Ag/GeS_2 double layer films by using neutron beam

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1. Introduction

When metal layers like Ag and Cu deposited on the chalcogenide glass films such as As_2S_3, As_2Se_3, GeS_2, and $GeSe_2$, are illuminated by light, the metal diffuses abnormally into the chalcogenide glass layer. The photon energy of the light required for the doping should be larger than that of the optical gap of the chalcogenide glass. This abnormal diffusion of metal is called photodoping discovered by Kostyshin et al in 1965. The photodoping process using several methods, for example, optical transmittance, AFM, ellipsometry, RBS, and ATR have been investigated. The purpose of our research is to make clear the behavior of Ag in the photodoping phenomenon. Especially, information on the time dependence of the Ag doped layer is not fully understood through the various techniques. As an alternative method to know the detailed behavior of the doping process, we selected the neutron reflectometry in this time.

2. Experiment

<u>Sample preparation</u> : GeS_2 films were deposited on thin Si single crystals (30 x 30mm) with the thickness of 100-200nm by the vacuum evaporation at the vacuum less than 6 x 10^{-5} torr. The Ag layer of 10-70nm thick was deposited onto the GeS_2 film under shielding from the outside light to prevent the extra photodoping before experiments.

<u>Experimental procedure</u> : The experimental setup is shown in Figs. 1, 2 and 3. The sample film (30 x 30mm) is set on a sample holder, which is conventionally used on BL17. A laser diode (405nm) with a beam expander is prepared by applicants and the exposing light area is adjusted to the right position where neutron beam is irradiated. Since the original beam size of the laser diode is 1mm in diameter, the beam is expanded to 30mm in diameter at the sample position using a beam expander. The laser beam covers the squared area of 20mm x 20mm, where neutron beams are irradiated in the experiment.

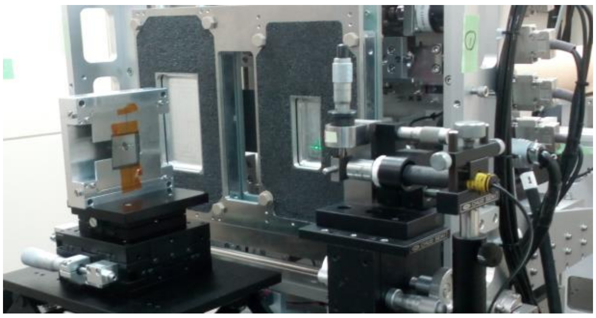

Fig. 1. Experimental setup in BL17

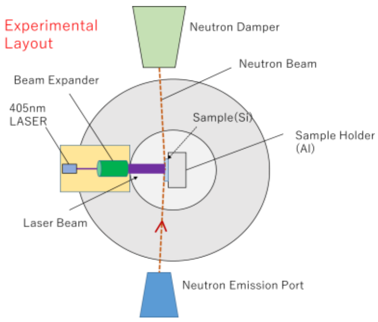

Fig. 2. Experimental setup

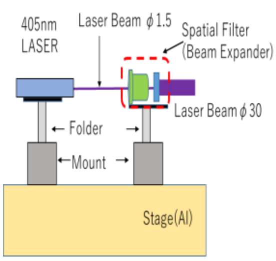

Fig. 3. Laser Setup

<u>Data analysis method</u> : From a full set of event data, a histogram of Time-of-Flight (TOF) spectrum is made considering a proper time resolution in the TOF (, which is related to Q-resolution), and a slicing time-region (, which depends on photoresponse in the reflectivity and is related to S/N ratio). The obtained reflectivity profile is compared to a simulated profile (model calculations), and are fitted, providing modified parameters such as scattering length density, layer thickness, and roughness. Time evolution of the reflectivity curve is also analyzed

by Fourier transformation, to evaluate changes in thickness. In addition, the experimental results obtained without neutron irradiation are analyzed and compared with the irradiated results to confirm the influence of neutron irradiation on photodoping.

3. Results

The following four samples such as a Si substrate, an a-GeS_2 film deposited on a Si substrate, an Ag film deposited on a Si substrate and an Ag/a-GeS_2 (30nm/200nm) film deposited on a Si substrate were measured. The time evolution of the neutron reflectivity curves for the two layered sample were obtained as shown in Fig.4 together with the fitting curves. The derived parameters corresponding to the thickness, densities and roughness of respective layers are shown in Table 1. The behavior of Ag photodoping was obtained through the Fourier analysis for the reflectivity curves and compared with the results of proceeding results using optical transmittance, AFM, ellipsometry, RBS, and ATR. As an example of the result, final ratio between Ag and doped layer was almost similar to the value estimated optical transmittance data measured with a spectrophotometer at Tokai University shown in Fig. 5.

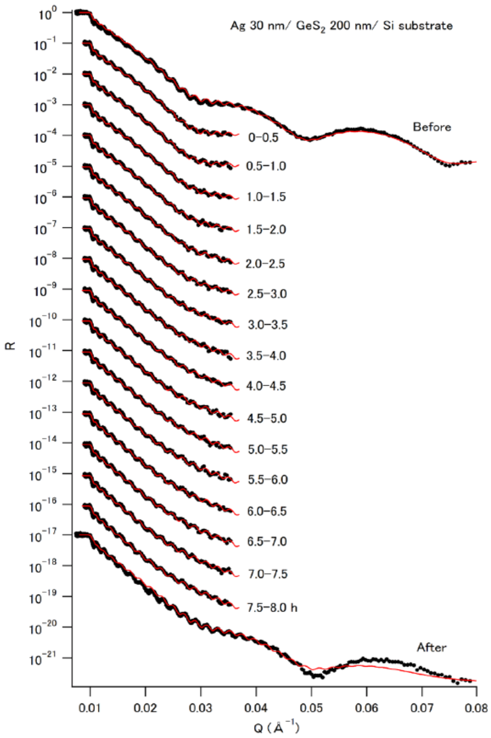

Fig. 4. Time dependence of neutron reflectance.
Black line: measured, red line: fitting.

Table 1

	Thickness(nm)	SLD	Roughness(nm)
Ag	13.2	3.47	3.44
Ag/GeS_2	11.7	2.81	0.388
a-GeS_2	258.5	2.06	0.376
SiO_2	1.5	3.47	0.3
Si		2.07	0.4

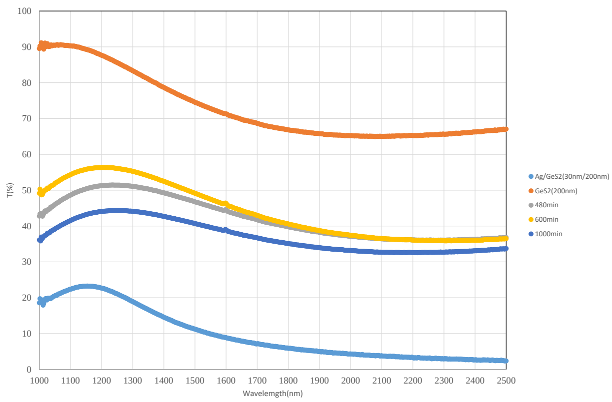

Fig.5. Time dependence of spectral transmittance.

4. Conclusion

The time evolution of the neutron reflectivity curves for the two layered sample were characterized and parameters corresponding to the thickness, densities and roughness of respective layers were derived. Through this experiment, it is suggested the neutron reflectivity method is effective tool to elucidate the mechanism of the photodoping phenomenon. We appreciate the cooperation of J-PARC and CROSS. As a further research, many samples with different compositions should be studied through enough machine time. In this time, net machine time was largely limited due to the system shut down of the beam line.