Experimental report

06/07/2016

Proposal: 6-05-962 **Council:** 10/2014

Title: Time-resolved neutron reflectivitymeasurements for silver photo-diffusion into Ge-chalcogenide films

Research area: Materials

This proposal is a new proposal

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Samples: Ag film on silicon wafer
Ge-S film on silicon wafer
Ag/Ge-S films on silicon wafers

Instrument	Requested days	Allocated days	From	To
D17	2	2	08/07/2015	10/07/2015

Abstract:

Silver photo-diffusion into amorphous chalcogenide films has attracted much interest because of the potential applications such as non-volatile memory devices. So far, the diffusion kinetics has been studied by mainly Rutherford Backscattering and the studies revealed their unusual diffusion profile, in which silver concentration abruptly drops off at the interface. However, strong helium ion beam also induces silver diffusion and therefore, it is desirable to use in-situ techniques to exclude any assumptions on the photo-induced changes. Time-resolved X-ray/neutron reflectivity is a suitable technique to clarify the time evolutionary changes. Strong X-rays of synchrotron radiation can also induce silver diffusion and use of neutrons is safer and suitable approach to exclude the possibility of the changes by the probe beam. So far, we performed time-resolved neutron reflectivity measurements on BL17 (SHARAKU) in J-PARC and 30 second time-resolution has been achieved. In this proposal, we perform time-resolved neutron reflectivity on D17 in ILL and try to find much faster dynamics up to 6 second resolution, which is the possible shortest time-resolution with the instrument.

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So far, we performed time-resolved neutron reflectivity measurements of Ag/Ge-S films under light illumination on BL17 (*SHARAKU*) in J-PARC. Using the intensive pulsed neutron source (the proton beam power was 300kW) and a delicate event-data recording system, we achieved 30 second time-resolution in the shortest case. Previously, it has been proposed that the silver photo-diffusion proceeds by a progression of the diffusion front and the diffusion is completed by the front reaching the end of the chalcogenide layer. However, the neutron reflectivity results showed that a metastable silver-rich reaction layer is formed first, and then silver diffusion is taken place by passing across the Ag-rich reaction layer/ Ag-poor reaction layer interface, almost fixing the position of the interface. In this way, we have demonstrated that time-resolved neutron reflectivity technique is powerful and useful technique to investigate on the silver photo-diffusion dynamics.

From our previous time-resolved neutron reflectivity measurements in J-PARC, it was found that there are two silver diffusion processes with different reaction time. The fast process corresponds to the silver diffusion from pure silver layer to metastable silver-doped reaction layer, and the slower one corresponds to the metastable Ag-rich reaction layer to the Ag-poor reaction layer. For $Ag/Ge_{20}S_{80}$ films, the fast one is about 2 min and the slower one is 20 min. Regarding the fast process, the data of 30 seconds period are not enough to analyze the process in detail. Also, there could be some other elemental processes in the *initial* diffusion stage, and it is worth going to a pioneering time region.

Therefore, we have performed time-resolved neutron reflectivity measurement on D17 at ILL. The experiment was very successful. Using the special technique to optimize the neutron intensity for fast measurements [1], we could obtain the transient data with 5 seconds time-resolution with good signal to noise ratio, which are comparable to the static data. From the time variation of the reflectivity at constant Q, it was found that there could be a new fast reaction (diffusion) process within 10 seconds. In addition, using 2-

dimensional detectors, the time variation of Yoneda peak was clarified. The result suggested that the interface roughness was also affected by the silver photo-diffusion. These results were presented at the annual meeting of the Japanese Society for Neutron Science held on December 11, 2015.

[1] R. Cubitt, T. Saerbeck, R. A. Campbell, R. Barker and P. Getfreund, J. Appl. Cryst. 48 (2015) 2006.