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Development of high-polarization Fe/Ge neutron polarizing supermirror: Possibility of fine-tuning of scattering length density in ion beam sputtering

R. Maruyama [a,*], D. Yamazaki [a], K. Akutsu [b], T. Hanashima [b], N. Miyata [b], H. Aoki [a], M. Takeda [c], K. Soyama [a]

[a] J-PARC Center, Japan Atomic Energy Agency, 2-4 Shirakata, Tokai, Ibaraki 319-1195, Japan
[b] Neutron Science and Technology Center, CROSS-Tokai, 162-1 Shirakata, Tokai, Ibaraki 319-1106, Japan
[c] Materials Sciences Research Center, Japan Atomic Energy Agency, 2-4 Shirakata, Tokai, Ibaraki, 319-1195, Japan

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ABSTRACT

The multilayer structure of Fe/Si and Fe/Ge systems fabricated by ion beam sputtering (IBS) was investigated using X-ray and polarized neutron reflectivity measurements and scanning transmission electron microscopy with energy-dispersive X-ray analysis. The obtained result revealed that the incorporation of sputtering gas particles (Ar) in the Ge layer gives rise to a marked reduction in the neutron scattering length density (SLD) and contributes to the SLD contrast between the Fe and Ge layers almost vanishing for spin-down neutrons. Bundesmann et al. (2015) have shown that the implantation of primary Ar ions backscattered at the target is responsible for the incorporation of Ar particles and that the fraction increases with increasing ion incidence angle and increasing polar emission angle. This leads to a possibility of fine-tuning of the SLD for the IBS, which is required to realize a high polarization efficiency of a neutron polarizing supermirror. Fe/Ge polarizing supermirror with $m = 5$ fabricated under the same condition showed a spin-up reflectivity of 0.70 at the critical momentum transfer. The polarization was higher than 0.985 for the q_z range where the correction for the polarization inefficiencies of the beamline works properly. The result of the polarized neutron reflectivity measurement suggests that the "magnetically-dead" layers formed at both sides of the Fe layer, together with the SLD contrast, play a critical role in determining the polarization performance of a polarizing supermirror.

1. Introduction

A neutron polarizing supermirror is a stack of alternating layers of ferromagnetic and non-magnetic materials with a variation in bilayer thickness to extend the bandwidth of the neutron spin polarization [1,2]. High polarization and wide bandwidth play a critical role in determining the performance of polarizing supermirrors to meet a variety of research demands. To extend the bandwidth, it is important to increase the ratio *m* of the critical momentum transfer of the supermirror to that of natural nickel. Fe is frequently used for the magnetic layer of the neutron polarizing supermirror because it is a easily deposited ferromagnetic material with a large saturation magnetization and low coercivity. In addition, the radioactivation of Fe due to neutron irradiation is much less serious than the other ferromagnetic materials such as Co. Si or Ge is generally used for the non-magnetic layer because the neutron scattering length density (SLD) is close to that of Fe for spin-down neutrons. Deposition techniques involving evaporation and magnetron sputtering have been employed and shown to be effective in improving the performance of neutron polarizing multilayer mirrors and supermirrors [3–12].

We have developed neutron polarizing and non-polarizing supermirrors by using the ion beam sputtering (IBS) technique [13–17]. IBS has advantages over the other sputtering techniques that a separate ion source is used to produce an ion beam that is focused and incident on the target material for deposition. This allows the energy and current density of the bombarding ions to be controlled independently. Furthermore, the discharge is not formed over the entire chamber, but is confined to the ion source. This reduces the amount of sputtering gas that is required and results in better vacuum conditions. These advantages lead to the production of layers with high density and small grain size, making the IBS technique suitable for the fabrication of polarizing supermirrors with wide bandwidth and high reflectivity of spin-up neutrons [18,19]. High polarization efficiency, however, requires fine-tuning of the SLD. In the magnetron sputtering, the reactive sputtering in which a reactive gas (e.g. N_2 or O_2) is added to the noble gas (usually Ar) for discharge is a well-established technique and is

* Corresponding author.
E-mail address: ryuji.maruyama@j-parc.jp (R. Maruyama).

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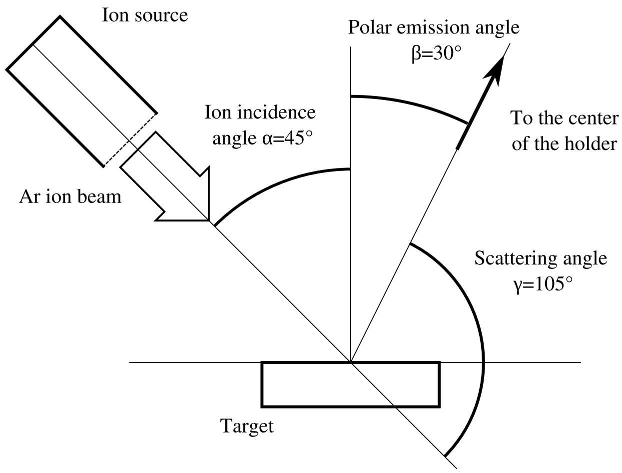

Fig. 1. Geometry of the IBS setup. These angles were fixed in this study.

widely used [9–12]. Until now, the possibility of using a reactive gas in IBS for neutron optical multilayers has not been explored.

In this study, we investigated the multilayer structure of the Fe/Si and Fe/Ge systems mainly using X-ray and polarized neutron reflectivity measurements and scanning transmission electron microscopy with energy-dispersive X-ray (STEM-EDX) analysis. Complementary use of these probes revealed that the incorporation of a considerable amount of Ar particles in the Ge layer, likely due to the implantation of primary Ar ions backscattered from the Ge target during the IBS process [20], results in a reduction in the SLD and contributes to the SLD contrast between the Fe and Ge layers almost vanishing for spin-down neutrons. The incorporation of primary ions offers a possibility of fine-tuning of the SLD for the fabrication of a polarizing supermirror using the IBS. This could be an alternative technique to the reactive sputtering in the magnetron sputtering. Fe/Ge polarizing supermirror with $m = 5$ fabricated under the same condition showed high performance not only in the reflectivity of spin-up neutrons but also in polarization arising from the small SLD contrast between the Fe and Ge layers. The simulation of the reflectivity profiles of the supermirror indicated that the "magnetically-dead" layers formed at top and bottom of the Fe layer, together with the SLD contrast, play a critical role in determining the polarization efficiency of a polarizing supermirror.

2. Sample preparation and experimental details

Fe/Si and Fe/Ge multilayers of 30 bilayers with a thickness of 10 nm (A and B) were fabricated by using an IBS system with dual bucket Ar^+ ion sources [13]. The thickness ratio was aimed at 1.0. The bilayer thickness of A and B was chosen because it is a representative of the sequence used for polarizing supermirrors.

The pressure before and during the sputtering process was approximately 1.0×10^{-5} and 3.0×10^{-2} Pa, respectively. One of the ion source is aimed at the target for sputtering. The other directly sees the substrate holder for the purposes such as polishing, which was not used in this study. The ion source for sputtering was operated under the condition of an ion energy of 600 V, current of 150 mA, and Ar mass flow of 20 sccm. The geometry of the IBS setting shown in Fig. 1, an ion incidence angle $\alpha = 45°$ and polar emission angle $\beta = 30°$ (i.e. scattering angle $\gamma = 180° - \alpha - \beta = 105°$), cannot be varied for this IBS system and hence it was fixed in this study. Fe, Si, and Ge targets with a purity higher than 99.99% were used. The sputtering rates were 4.21, 6.33 and 6.45 nm/min for Fe, Si, and Ge, respectively.

Si(100) substrates with an area and thickness of 73×25 mm^2 and 3 mm, 5×5 mm^2 and 0.2 mm, and 73×25 mm^2 and 0.6 mm were used for the X-ray and polarized neutron scattering, magnetic hysteresis, and TEM and STEM-EDX measurements, respectively. The Si substrates were polished so as to reduce the root-mean square (rms) surface roughness less than 0.3 nm. The deposition on these three kinds of substrates was performed in the same process.

The other sample was an Fe/Ge neutron polarizing supermirror with $m = 5$. The layer sequence was designed by using the algorithm proposed by Hayter and Mook, where the parameter ζ provides the layer sequence for a given minimum thickness corresponding to m-value and hence determines the reflectivity profile [21]. To obtain an appropriate design, polarized neutron reflectivity calculation was performed for obtained layer sequences with different ζ. The neutron reflectivity calculation was carried out by means of Parratt's dynamical approach [22]. The interface roughness was included in the Fresnel coefficients, as proposed by Névot and Croce [23]. An rms interface roughness of 0.75 nm was chosen in the calculation because it was a typical value in the simulation for supermirrors we had fabricated. The incident neutron beam was assumed to have a wavelength spread of 5.0% in FWHM and divergent angle of 1 mrad. The relation between the reflectivity for spin-up neutrons at the critical angle and number of layers is shown in Fig. 2. The value of $\zeta = 0.977$, corresponding to 4994 layers and a reflectivity of 0.70, was chosen as a compromise between the reflectivity and number of layers.

X-ray diffraction in a Bragg–Brentano geometry and reflectivity measurements were performed on a Rigaku Ultima III diffractometer. An incident X-ray beam with a wavelength of 0.154 nm was obtained by using Cu Kα radiation.

Magnetization measurements were performed by using a SQUID magnetometer (Quantum Design MPMS2) with a superconducting magnet capable of delivering magnetic fields up to 8.0×10^5 A/m. The magnetic field was applied parallel to the sample surface and hysteresis loops were measured at a temperature of 300 K. The error in the absolute value of the magnetization arises from the error in the sample area and the instrumental error. The former is estimated to be 1.4% whereas the latter is much smaller. By this estimation, an error of 1.4% should be taken into account when comparing measured values between different samples.

Polarized neutron reflectivity measurement was performed on the polarized neutron reflectometer SHARAKU with a horizontal scattering geometry installed at the Materials and Life Science Experimental Facility (MLF) of the Japan Proton Accelerator Research Complex (J-PARC) [24]. A pulsed neutron beam was obtained by the spallation reaction at the circulating mercury target where a high-energy proton beam with a repetition rate of 25 Hz was injected. The neutron flux is proportional to the proton beam power which was 150 kW at the present experiment. The MLF uses the event recording method as a standard data acquisition system [25]. Each signal that is a detection of neutrons, status of beamline optical components, conditions of the sample environment equipments, and so on, is recorded with the time stamp. The event data were converted to a histogram format as a function of the time-of-flight (TOF). The time bin lengths which define the TOF resolution can be arbitrarily chosen. The other factors to worsen the TOF resolution such as the neutron pulse width were enough smaller than the bin length corresponding to a TOF resolution of a few %. The incident beam was collimated by a pair of horizontal slits to an angular resolution of 3.4%. The polarizer and analyzer were Fe/Si polarizing supermirrors with $m = 4$, which provided a wavelength band from 0.2 to 0.84 nm. The polarization for the incident and reflected beams averaged over the wavelength bandwidth was 0.97 and 0.95, respectively, where the inefficiencies of the spin-flippers were included. The data were collected by a ^{3}He gas tube detector without spatial resolution.

TEM measurement was performed on a Hitachi H-9000NAR microscope. STEM-EDX measurements were performed on a Hitachi HD-2700 microscope equipped with an AMETEK EDAX Octane T Ultra W spectrometer. The acceleration voltage was 300 and 200 kV for the TEM and STEM-EDX, respectively. Samples used for the TEM and STEM-EDX measurements were prepared by a focusing ion beam system (Hitachi