Modification of interface magnetic anisotropy by ion irradiation on epitaxial Cu/Ni/Cu(002)/Si(100) films

J.-S. Lee,1 K.-B. Lee,1,2,* Y. J. Park,2 T. G. Kim,3 J. H. Song,3 K. H. Chae,3 J. Lee,4 C. N. Whang,4 K. Jeong,4 D.-H. Kim,3 and S.-C. Shin3

1eSSC and Department of Physics, Pohang University of Science and Technology, Pohang 790-784, Korea
2Pohang Accelerator Laboratory, Pohang University of Science and Technology, Pohang 790-784, Korea
3Advance Analysis Center, KIST 39-1, Hawolgok-dong, Seungbuk-gu, Seoul 136-791, Korea
4Department of Physics, Yonsei University 134, Shinchon-dong, Sudaemoon-gu, Seoul, Korea
5Department of Physics and CNSM, Korea Advanced Institute of Science and Technology, Taejon 305-701, Korea

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Various x-ray scattering and magnetic measurements were employed to reveal changes in intrinsic structural and magnetic properties on epitaxial Cu/Ni(1)/Cu(002)/Si(100) thin films (t = 20, 30, 60, and 90 Å) before and after 1 MeV C⁺ ion irradiation. Torque magnetometer and grazing incidence x-ray diffraction measurements were carried out to understand relation between magnetic and structural properties, respectively. X-ray reflectivity measurements were performed to characterize interface roughness and intermixing. It is observed that effective magnetic anisotropy values of ion-irradiated films are negative over the entire nickel thickness range and the dominant factor of the reorientation of magnetic easy axis from surface normal to surface parallel is reduction of the interface magnetic anisotropy coefficient in spite of decreased interface mixing after ion irradiation.

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In recent years, magnetic thin films possessing an easy axis of magnetization perpendicular to surface have attracted many interests due to their advantages in magnetic and magneto-optical recording media.1–11 One of the advantages of perpendicular over longitudinal magnetization is that it leads to higher recording densities with higher thermodynamic stability.

Perpendicular magnetic anisotropy (PMA) has been observed in a wide range of magnetic thin films including alloys and superlattices. Their magnetic easy axis are often observed to rotate from perpendicular to in-plane direction depending on film thickness. There have been controversies regarding the origin of the PMA transition: whether magnetoelastic (ME) effect or interface magnetic anisotropy effects are major factors.8–11 Moreover, it has been recently demonstrated that ion irradiation can be used to modify magnetic anisotropy characteristics of thin films from perpendicular to lateral magnetic easy axis.12–15 Ion irradiation has been used to modify magnetic properties such as magnetic anisotropy, coercivity, and magnetic exchange fields for well-known PMA systems such as Co/Pt superlattice, Co/Pd, Pt/Co/Pt, Fe/Ag, Fe/Cu, Ni/Pd, and Cu/Ni/Cu.12–20

Since ions can be focused to a spot of nanometer scale, ion-beam lithography is expected to allow us to fabricate nanopatterned magnetic media. Though it is expected to have huge impacts on future applications in magnetic data storage devices, modification of magnetic properties by ion irradiation is still at its early stage in understanding their origins. For Co/Pt-based thin films, the change of magnetic anisotropy is attributed to blurred interfaces between Co and Pt due to ion-beam mixing effect.12,16–18 Since Ni and Cu have an opposite sign of heat of mixing to that of Co and Pt,19 it is anticipated that Cu/Ni/Cu sandwiches have opposite effects of ion irradiation on magnetic anisotropy. However, Cu/Ni/Cu thin films were reported to show similar trends of modification, the rotation of magnetic easy axis from perpendicular to surface parallel direction, by ion irradiation,14 and the authors attributed it to strain relief by ion-induced dislocation defects and ME effects of epitaxial Cu/Ni/Cu(002)/Si(100) thin films.

In this work, systematic studies of torque magnetometer, vibrating sample magnetometer (VSM), grazing incidence x-ray diffraction (GID), and x-ray reflectivity (XRR) measurements were carried out on both as-grown and ion-irradiated epitaxial Cu/Ni/Cu(002)/Si(100) thin films of different Ni thickness, which are the same samples of Ref. 14, in order to confirm the relation between structural and magnetic properties. Torque magnetometer measurements data provide us with information related to magnetic anisotropy energy while GID and XRR data provide with structural information such as strain, intermixing, and roughness of the interface.

Analysis of the data clearly shows that the modification of the PMA after ion irradiation is dominated by change of the interface magnetic anisotropy coefficient rather than by ME effects. Additionally, ion-beam mixing effects at the Ni/Cu interfaces appear to be negligible in the context of PMA changes.

Growth of the epitaxial Cu/Ni/Cu(002)/Si(100) films has been described in detail by Kim et al.14 The samples show clearly the transition of magnetic easy axis from perpendicular to in-plane directions after ion irradiation.

Various x-ray scattering and magnetic measurements were carried out to understand structural and magnetic properties of the epitaxial Cu/Ni(i)/Cu(002)/Si(100) films before and after ion irradiation. X-ray measurements were carried out on a bending magnet beam line 3C2 at the Pohang Accelerator Laboratory (PAL). The wavelength of x-rays was 1.54 Å. Torque magnetometer data were measured as the samples were rotated about the (010) crystallographic direction under
an applied magnetic field of 0.98 T, which saturates the nickel magnetic moments even along the hard axis of magnetization. The effective magnetic anisotropy values $K_{\text{eff}}$ for the samples were estimated from analysis of the torque magnetometer data. Values of the saturated magnetization $M_s$ were also measured by VSM for all the samples.

The epitaxial Cu/Ni/Cu(002)/Si(100) films after ion irradiation showed changes in both magnetization directions and lattice strains, relative to the as-grown state. In order to understand ME effect of the films, we adopted the second-order spin-pair (SP) model of Ha et al. The model is employed since it describes ME coupling coefficients, to a good approximation, with a few parameters in terms of the dipole interactions. According to the SP model, $K_{\text{eff}}$ can be expressed as follows:

$$K_{\text{eff}} = 2(K_1 + B_1 \varepsilon_1 + D_1 \varepsilon_1^2) + (B_b \varepsilon_1 + D_b \varepsilon_1^2) \tau - 2 \pi M_s^2 t.$$  

(1)

Here, $B_1$ is the first-order interface ME coupling constant, $D_1$ is the second-order interface ME coupling constant, $B_b$ is the first-order bulk ME coupling constant, $D_b$ is the second-order bulk ME coupling constant, and $K_1$ is an interface magnetic anisotropy coefficient. $B_1$, $D_1$, and $B_b$ can be derived from reported parameters of nickel\textsuperscript{8,22} while $M_s$ were measured by VSM. Therefore, there are only two adjustable fitting parameters $K_1$ and $D_b$ in our analysis.

$K_{\text{eff}}$ includes several magnetic anisotropy constants, which can be distinguished by their respective dependence on the in-plane strain ($\varepsilon_1$). In order to analyze the dependence of $K_{\text{eff}}$ on in-plane strains, the GID measurements were carried out to investigate $\varepsilon_\parallel$ as a function of nickel film thickness and ion irradiation. Figure 1 shows results of the GID measurements on the epitaxial Cu/Ni/Cu(002)/Si(100) films before and after ion irradiation. In the case of as-grown films, the copper Bragg peaks are positioned at their bulk value but the nickel ones are shifted from its bulk value.\textsuperscript{23} In the case of ion-irradiated films, the copper Bragg peaks do not move while the nickel ones are shifted toward the bulk value much more than as-grown films. The $\varepsilon_\parallel$ values of the nickel layer were obtained by fitting data with the combined Gaussian and Lorentzian resolution function, as presented as solid lines in the Fig. 1. Extracted values of strain from the as-grown and ion-irradiated samples followed a phenomenological power-law dependence of $t^{-2/3}$ ($t$=nickel film thickness), as reported by others.\textsuperscript{8,9} As shown clearly in Fig. 1, strains for irradiated samples of Ni film thickness, 20 and 30 Å, are still larger than those for as-grown samples of the thickness, 60 and 90 Å. Despite of their larger strains, the irradiated samples of thinner Ni layers have their magnetic easy axis in lateral directions while the as-grown samples of thicker layers, with smaller strains, have the perpendicular magnetic easy axis. This result strongly implies that ME is not the dominant factor for modification of the PMA by ion irradiation.

In order to investigate in detail whether the PMA transition after ion irradiation can be attributed to the strain relief with ion-induced dislocation defects and ME effects of epitaxial Cu/Ni/Cu(002)/Si(100) thin films, the $K_{\text{eff}}$ and $M_s$ were plotted as a function of the $\varepsilon_1$ of nickel film, as shown in Fig. 2. The result shows that the $K_{\text{eff}}$ data before and after ion irradiation have an additional tendency besides strain relief. A remarkable behavior of the $K_{\text{eff}}$ after ion irradiation is its shifts to negative values over the entire thickness range, which cannot be explained by magnetoelastic effect alone. If magnetoelastic effects followed by strain relief is the dominant factor causing the transition, it is anticipated that $K_{\text{eff}}$ for both as-grown and ion-irradiated films can be expressed approximately by a single set of the effective magnetic anisotropy coefficients and can be plotted by a single curve, which is not the case as shown in Fig. 2. We also note that the magnetization before and after ion irradiation decrease rapidly above strain value $\sim 1.4\%$ (thickness: below $\sim 30$ Å). The dramatic reduction in $M_s$ is consistent with the results of others.\textsuperscript{10,24,25}

In order to investigate the effects of ion-irradiation quan-
In summary, we have investigated relation between magnetic and structural properties on epitaxial Cu/Ni(t = 20, 30, 60, and 90 Å)/Cu(002)/Si(100) films before and after ion irradiation. The filled and open circles are data of the as-grown and ion-irradiated samples, respectively. The solid and dashed lines are the best fit using the second-order SP model. Since $\varepsilon_1$ has dependence on $t^{-2/3}$, the curves are not simple parabola. The negative value of the $K_{eff}$ indicates in-plane magnetization. Inset: the saturated magnetization $M_s$ before and after ion irradiation as a function of nickel film strain, where the solid and dashed lines are guide to the eye.

titively, magnetic anisotropy coefficients were estimated using the SP model mentioned above. The derived terms have nearly the same values for both kinds of samples, while two fitting parameters have the following values: $D_b^{as} = -1.74 \times 10^{10}$ ergs/cm$^3$, $D_b^{irr} = -1.07 \times 10^{10}$ ergs/cm$^3$ and $K_{1}^{as} = 0.978$ ergs/cm$^2$, $K_{1}^{irr} = 0.045$ ergs/cm$^2$. Superscripts as. and irr. represent as-grown and ion-irradiated values, respectively. The solid and dashed lines of Fig. 2 represent values generated with estimated magnetic anisotropy coefficients for as-grown and ion-irradiated films, respectively, while filled and open circles represent experimental values for the films. From these results, it is clear that the shift of the $K_{eff}$ values by ion irradiation is mainly due to the change in $K_1$ while change in $D_b$ just results in a slightly different curvature of the relation. Due to the relatively small number of data points, 4, compared to that of the fitting parameters, 2, the estimated values of $K_1$ and $D_b$ might have some errors. However, the major effect of ion irradiation on the change in $K_1$ cannot be disputable. Since $K_1$ represents interface magnetic anisotropy, ion-irradiation results in modification of interface magnetic property and it is the main factor of the PMA transition on epitaxial Cu/Ni/Cu(002)/Si(100) film after ion irradiation, while ME effects play minor roles in the transition. This result is opposite to the previous of Ref. 14 argument.

For Co/Pt-based thin films, the variation of $K_1$ was attributed to the intermixing effect at the interface due to ion irradiation. In order to investigate structural changes at the interfaces of the samples, XRR measurements were carried out. In Fig. 3, the XRR data clearly show an enhancement of the Kiessig fringes after ion irradiation of the Cu/Ni(60 Å)/Cu sample. Oscillations of short wavelength represent the total thickness of the overlayers while oscillations having longer wavelengths represent either each layer thickness or their combinations. After ion irradiation, the long oscillations were enhanced better than the as-grown film.
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*Electronic address: kibong@postech.ac.kr

22From SP model of Eq. (1), $B_1$, $D_1$, and $B_b$ can be clearly derived as follow: $B_1=(a_0/16)[3B_2−4(2+(2c12/c11))B_1]$, $D_1=−(a_0/4)[1−1/(2c12/c11)1+(2c12/c11)2/3+2(2c12/c11)−(2c12/c11)^2]$, $B_b=B_l[1+(2c12/c11)]$, where $a_0$ is nickel lattice constant, $c_11(2.5×10^2\text{dynes/cm}^2)$ and $c_12(1.6\times10^3\text{dynes/cm}^2)$ are elastic constants of the nickel. $B_1(6.2\times10^7\text{ergs/cm}^3)$ and $B_2(8.6\times10^7\text{ergs/cm}^3)$ are the two well known bulk ME coefficients.