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Out-of-plane exchange coupling between epitaxial Ni (50 Å) and NiO (600 Å) bilayers

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We have investigated the exchange coupling between an epitaxial Ni (50 Å) film with an out-of-plane magnetic easy axis and a NiO (600 Å) film by polar magneto-optic Kerr-effect measurements. The temperature dependences of exchange field H_E for both as-deposited and field-cooled states exhibit the same blocking temperature $T_B \sim 130\,°\mathrm{C}$. The exchange field H_E for the field-cooled state is lower than that for the as-deposited state. The hard-axis in-plane loop shows a much smaller value of H_E. No coercivity enhancement is observed. The data are suggestive of linear coupling across the ferromagnet/antiferromagnet interface. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1538315]

I. INTRODUCTION

In ferromagnet (FM)/antiferromagnet (AF) systems, exchange coupling between FM and AF layers causes the hysteresis loop of the FM layer to shift by the exchange field H_E. The exchange-biased FM layer often exhibits an enhanced coercivity H_C. Recent review papers[1,2] have summarized the experimental and theoretical studies performed to understand the mechanism of exchange coupling. Most investigations have been reported on exchange-biased FM/AF systems with a ferromagnetic in-plane easy axis. Recently, exchange coupling has been found in FM/AF systems with a ferromagnetic out-of-plane easy axis.[3–6] Pt/Co or Pt/CoFe multilayers are used as the FM layer with out-of-plane anisotropy. In Co/Pt multilayers exchange-biased with naturally oxidized thin CoO (10 Å), [3] a model has been proposed to show that out-of-plane AF spin components are necessary to obtain out-of-plane exchange coupling. In this article, we have investigated the out-of-plane exchange coupling between an epitaxial Ni film (50 Å) with out-of-plane easy axis and a textured NiO film (600 Å).

II. EXPERIMENTAL PROCEDURES

The details of epitaxial growth of Ni on Si(001) with a Cu seed layer have been reported elsewhere.[7] An out-of-plane magnetic easy axis is obtained if the Ni thickness is between 20- and 120 Å.[8–10] A Si(001) wafer was degreased, etched in dilute HF for 12 min, rinsed in de-ionized water, and dried with nitrogen gas. It was then loaded into an e-beam evaporation chamber with a base pressure of 4×10^{-7} Torr. The evaporation pressure was 6×10^{-7} Torr. A seed layer of 1000-Å Cu, followed by a layer of 50-Å Ni was evaporated at the rate of ~0.15 Å/s. A 600-Å NiO film was then deposited in 3 m Torr Ar atmosphere using rf magnetron sputtering from a NiO target at a deposition rate of 0.31 Å/s. The crystal structures were checked by x-ray diffraction with Cu K_α radiation (see Fig. 1). Both Cu and Ni layers grow epitaxially with (200) orientation. The NiO layer has a single NaCl-type fcc phase. For the as-deposited state, the NiO layer is polycrystalline [see Fig. 1(a)], but after annealing at 180 °C, it becomes highly (111) textured [see Fig. 1(b)]. This change in texture caused by the annealing has a strong effect on H_E, as we will discuss subsequently. During the preparation of samples, no external field was applied. The observation of exchange field in the as-deposited state implies that the Ni layer was in a single domain during NiO deposition which can be ascribed to the perpendicular component (~24 Oe) of the stray field emanating from the rf sputtering gun. A bare 50-Å-thick Ni film was also made for comparison.

Magnetic measurements were performed using a magneto-optic Kerr-effect (MOKE) magnetometer. A 660-nm laser with s-polarization was used. With a magnetic field perpendicular to the sample surface, measurements of polar MOKE loops were made on the as-deposited sample while heating up to 180 °C (above the blocking temperature of $T_B \sim 130\,°\mathrm{C}$). The sample was maintained at 180 °C for 10 min and then cooled to room temperature in a 3 kOe magnetic field applied perpendicular to the plane. The sample was then reheated and polar MOKE loops were measured at various temperatures. The field-cooling procedure was also performed with the magnetic field applied in-plane.

III. RESULTS AND DISCUSSION

Figure 2(a) shows polar MOKE loops at different temperatures for the as-deposited state. All loops exhibit a square shape, indicating an out-of-plane easy axis. The loop height decreases monotonically with temperature, while maintaining its square shape. Since only the temperature of the sample was varied, with no change in any other measurement parameters, the loop height is proportional to the magnetization of the sample. There is no reorientation of the easy axis, and the reversal mechanism along the easy axis does not vary with temperature. The same results have been observed for the field-cooled state and the uncoupled Ni film. Both the as-deposited and field-cooled states and the un-

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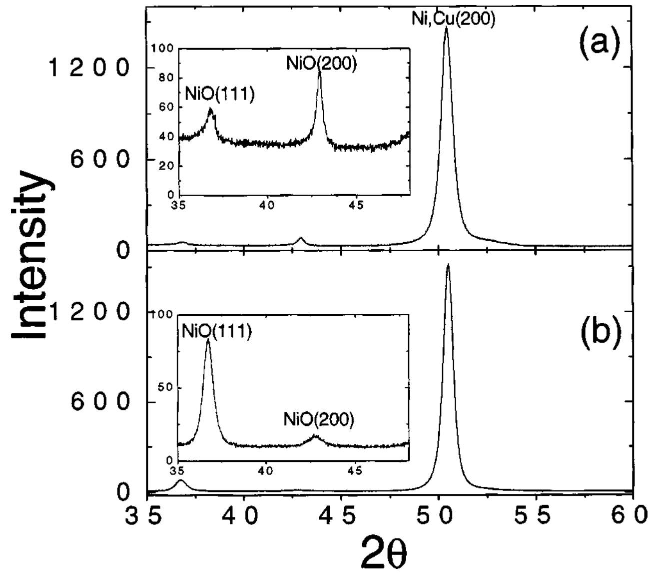

FIG. 1. X-ray diffraction spectra of a Si(100)/Cu(100 nm)/Ni(5 nm)/NiO(60 nm) sample. (a) As-deposited state. (b) Field-cooled state. The insets show the expanded peaks of NiO in the range of $35° < 2\theta < 45°$.

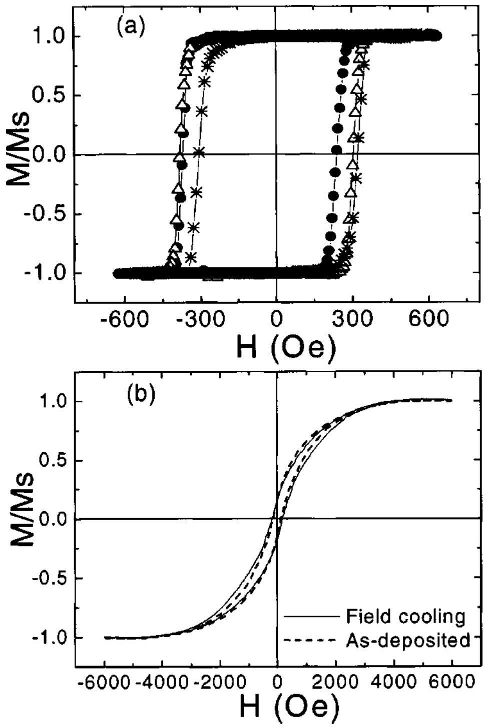

FIG. 3. (a) Polar MOKE loops at room temperature for as-deposited (●) and field-cooled (Δ) states and for the uncoupled Ni film (*). (b) Hysteresis loops for as-deposited and field-cooled states with magnetic field applied in-plane. These loops were measured using on alternate gradient force magnetometor.

coupled Ni film exhibit the same temperature dependence of saturation magnetization, as shown in Fig 2(b). We fit the data for the pure 50-Å Ni film to the phenomenological power-law $M_F(T) = (1 - T/T_C)^{\alpha}$, where M_F is the normalized saturation magnetization and α is a power-law exponent. Both the Curie temperature $T_C = 240\,°\mathrm{C}$ (below the bulk T_C for Ni) and the exponent of $\alpha = \sim 0.44$ are identical to the molecular-beam-epitaxy-grown films with comparable thickness,[11] an indicator of the quality of our epitaxial Ni film. This reduced Curie temperature is very close to the Néel temperature of bulk NiO (250 °C).

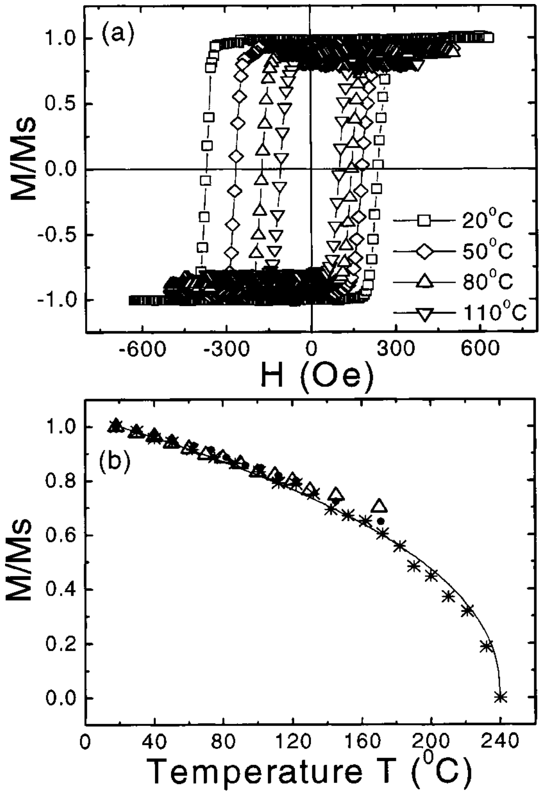

FIG. 2. (a) Polar MOKE loops at different temperatures for the as-deposited state. (b) Normalized saturation magnetization M_F for as-deposited (●) and field-cooled (Δ) states and for the uncoupled Ni film (*). The solid line is the power-law fit to $M_F = (1 - T/T_C)^{\alpha}$.

Figure 3(a) shows the polar MOKE loops at room temperature for as-deposited and field-cooled states and the uncoupled Ni film. The coercive fields for the bare Ni film, the as-deposited bilayer, and the field-cooled bilayer were $H_C = 323$, 303, and 333 Oe, respectively, demonstrating the absence of any enhancement. The uncoupled Ni film exhibits no loop shift, whereas the loop shifts for the as-deposited and field-cooled samples are $H_E = 65$ and 38 Oe, respectively. Figure 3(b) shows the in-plane hard-axis hysteresis loops at room temperature for as-deposited and field-cooled states. The in-plane loop shifts for the as-deposited and field-cooled states are much smaller ($H_E = 28$ and 10 Oe, respectively), with coercivities of 138 and 180 Oe. The saturation field (~ 4 kOe) of the hard-axis loops is much higher than the easy-axis switching fields, which suggests domain nucleation followed by domain wall propagation as the magnetization reversal mechanism along the easy axis. Neither the easy- nor hard-axis loops show any asymmetry for the increasing and decreasing fields, as is often observed for in-plane exchange biasing experiments.[12]

The temperature dependences of the exchange field H_E and coercivity H_C for the as-deposited and field-cooled states, and for the uncoupled Ni film, are shown in Figs. 4(a) and 4(b). In spite of the comparable values of T_C and T_N, no peak has been seen in either H_E or H_C, [13] for reasons related

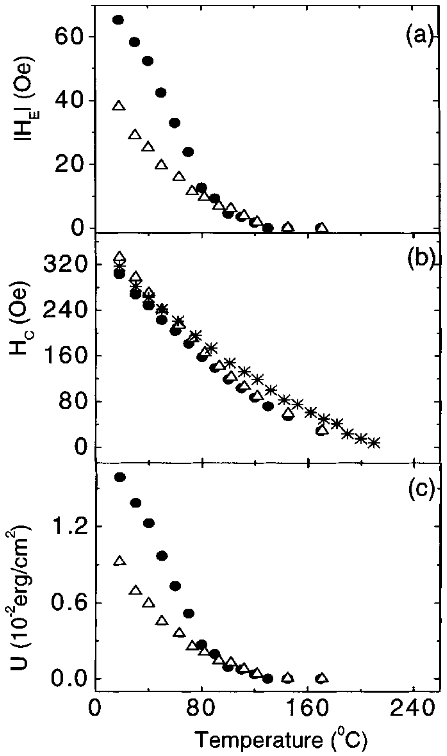

FIG. 4. Temperature dependences of (a) the exchange field H_E, (b) coercivity H_C, and (c) the exchange-coupling energy U. ●: as-deposited state; Δ: field-cooled state; *: pure Ni film.

to the anisotropy of the NiO that will be further discussed subsequently. The coercivity decreases almost linearly with increasing temperature for all three measurements. No coercivity enhancement has been observed. A similar lack of coercivity enhancement in exchange-coupled systems has been seen in $(\mathrm{Pt/Co_{0.9}Fe_{0.1}})n/\mathrm{FeMn}$ multilayers with perpendicular anisotropy[4] and in FeNi/FeMn bilayers[14] for FeMn thicknesses above 100 Å, and has been ascribed to the pinning of AF spins due to the AF anisotropy.

The AF NiO consists of an assembly of grains. According to the models proposed by Fulcomer and Charap[15] and its extension,[16] enhanced coercivity is due to the "dragging" of the AF moments in small AF grains at the interface which rotate when the magnetization of the FM layer is reversed, while the AF moments in large AF grains that do not rotate are responsible for the shift of the loop. In our sample, the square shape of the hysteresis loops implies that magnetization reversal for the out-of-plane direction in the FM is due to domain nucleation and domain wall propagation instead of rotation, as discussed previously. The strong out-of-plane anisotropy limits the modes of reversal and the exchange coupling between the interfacial FM and AF spins at the interface will not allow rotation of the interfacial spins of the AF grains during magnetization reversal, in which case no coercivity enhancement will occur.

The exchange field H_E for the as-deposited state is much larger than that for the field-cooled state at $T \lesssim 80\,°\mathrm{C}$, (as previously seen by Carey and Berkowitz[17] in NiFe/NiO with in-plane anisotropy), and becomes comparable at $T > 80\,°\mathrm{C}$. Both exhibit the same blocking temperature T_B $\sim 130\,°\mathrm{C}$, which is $\sim 120\,°\mathrm{C}$ below the bulk Néel temperature $T_N \sim 250°\mathrm{C}$ of NiO, in contrast to other FM/NiO systems with in-plane FM easy axis in which $T_B \sim 202\,°\mathrm{C}$.[17] The low blocking temperature for NiO has been ascribed to the rapid drop in the anisotropy constant K_{AF} with increasing temperature, dropping by a factor of 10 in going from room temperature to 130 °C.[18]

The exchange-coupling energy is determined by $U = t_F M_F(T) H_E(T)$, with M_F and t_F being the saturation magnetization and thickness of the FM layer, respectively. The temperature dependence of U is shown in Fig. 4(c), revealing similar behavior to the exchange field H_E. Even though M_F decreases monotonically with increasing temperature [see Fig. 2(b)], it still maintains a high value at the blocking temperature. The monotonic decrease of M_F can not be the reason for the disappearance of U and H_E at a temperature much below the Néel temperature.

As proposed by Malozemoff[19] and Mauri *et al.*,[20] domain wall formation in the AF suggests that the exchange-coupling energy and hence the exchange field are related to the domain wall energy $(2\sqrt{AK_{\mathrm{AF}}})$. The temperature dependence of H_E and U will be primarily controlled by the temperature dependence of the AF anisotropy constant K_{AF}. For NiO, the weak anisotropy constant $K_{\mathrm{AF}} (= 3.3 \times 10^2\ \mathrm{erg/cm^3})$ decreases rapidly with increasing temperature. As the AF anisotropy decreases, the net out-of-plane AF moment at the interface will relax quickly to zero with increasing temperature, causing the rapid disappearance of the exchange coupling (and the exchange field) at a blocking temperature that is much lower than the Néel temperature. Similarly, the peaks in the temperature dependence of H_C and H_E seen by Wu *et al.*[13] are ascribed to a competition between the sharp decrease of AF and FM magnetic order; in our case, the temperature dependence of the anisotropy of NiO causes a sharp decrease in the AF magnetic order at temperatures at which the FM magnetization still maintains a high value.

The magnetic order in NiO consists of sets of antiferromagnetically-coupled (111) planes with ferromagnetically ordered moments pointing in the $\langle 11\bar{2}\rangle$ direction.[21] There are four equivalent (111) planes and three equivalent $\langle 11\bar{2}\rangle$ directions for each plane, leading to 12 equivalent easy axes for the NiO spins.[21] Assuming the exchange-bias field arises from a linear spin–spin interaction term determined by $S_F S_{\mathrm{AF}} \cos\beta$, where S_F, S_{AF}, and β are the FM spin, AF spin, and the angle between them, respectively, the exchange coupling field is dependent on the availability of AF domains with easy axes parallel to the FM moment direction during the field cooling. A net AF moment at the interface is necessary for the appearance of both the in-plane and out-of-plane exchange-coupling field.[3,22,23] Field cooling in the presence of a FM layer has been shown to alter the populations of the equivalent AF domains in the NiO layer.[22] Fields parallel to the surface will increase the population of domains with the easy axis parallel to the interface, while an out-of-plane applied field will increase the population of domains with the easy axis perpendicular to the interface. In our as-deposited sample, the field is provided by the FM Ni layer and is directed out-of-plane, leading to a larger population of out-of-plane domains. Our observed in-plane exchange field is much smaller than the out-of-plane one, confirming that it is collinear exchange coupling that is responsible for exchange-bias coupling. Our interpretation is similar to the work on Co/Pt multilayers exchange biased with CoO,[3] in which the strength of the coupling was proportional to the population of AF domains parallel to the magnetization of the FM layer.

For the field-cooled sample, x-ray spectra in Fig. 1(b) shows that the NiO layer becomes highly (111) textured. The AF spins in NiO layer lie in the antiferromagnetically coupled (111) planes, making it harder to repopulate the AF domains with an easy axis parallel to the FM Ni moments. The population of AF domains with the easy axis parallel to the FM Ni moments will be lower for the field-cooled state with the dominant (111) texture than that for the as-deposited polycrystalline state, causing a lower exchange field for the field-cooled state.

IV. CONCLUSION

In summary, out-of-plane exchange coupling has been found in an epitaxial Ni/textured NiO bilayer that shows no coercivity enhancement. The lack of coercivity enhancement has been ascribed to the magnetization reversal mechanism in the FM Ni layer, in which the strong out-of-plane anisotropy limits the possible reversal modes. In this bilayer, for both as-deposited and field-cooled states, the temperature dependence of the exchange field H_E demonstrates the same blocking temperature of $T_B \sim 130\,^\circ\mathrm{C}$, much lower than the Néel temperature, which we attribute to the steep temperature dependence of the anisotropy constant of NiO. The lower H_E for the field-cooled state is attributed to the change in the texture of the AF NiO layer after annealing. The larger value for the out-of-plane exchange field as compared to the in-plane exchange field provides strong support for collinear exchange coupling.

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