Abstract

(001) oriented NiO/NiFe bilayers were grown on single crystal MgO (001) substrates by ion beam sputtering in order to determine the effect that the crystalline orientation of the NiO antiferromagnetic layer has on the magnetization curve of the NiFe ferromagnetic layer. Simple models predict no exchange anisotropy for the (001)-oriented surface, which in its bulk termination is magnetically compensated. Nonetheless exchange anisotropy is present in the epitaxial films, although it is approximately half as large as in polycrystalline films that were grown simultaneously. Experiments show that
differences in exchange field and coercivity between polycrystalline and epitaxial NiFe/NiO bilayers couples arise due to variations in induced surface anisotropy and not from differences in the degree of compensation of the terminating NiO plane. Implications of these observations for models of induced exchange anisotropy in NiO/NiFe bilayer couples will be discussed.

75.50.Ee,61.50.Cj,81.15.Cd,75.30.Et,85.70.Kh
I. INTRODUCTION

Exchange anisotropy refers to the effect that an antiferromagnetic (AF) layer grown in contact with a ferromagnetic (FM) layer has on the magnetic response of the FM layer. Exchange anisotropy is one of several magnetic interfacial interactions, which include interlayer coupling in multilayers, that have been intensively studied in recent years. The most notable changes in the FM hysteresis loop due to the surface exchange coupling are a coercivity enhanced over the value typically observed in films grown on a nonmagnetic substrate, and a shift in the hysteresis loop of the ferromagnet away from the zero field axis. The characteristics of the AF layer and the interface between the two layers that produce the strongest exchange bias are not well understood. Experimental studies and theoretical models indicate that intrinsic magnetic properties of the AF such as the magnetocrystalline anisotropy, exchange stiffness and crystalline texture as well as extrinsic properties such as grain size, domain size and interface roughness may influence the resulting response of the FM. Unfortunately, it is difficult to manipulate these properties independently, or to probe the magnetic structure of the bilayer interface directly.

Hysteresis loops of a NiO(500 Å)/NiFe(100 Å) bilayer couple measured below and above the NiO blocking temperature, T_b, are shown in Figure 1 and illustrate the effects of the interface exchange interaction. Above the blocking temperature (T_b=200°C < the Néel temperature, T_N = 240°C, the NiO spins are thermally fluctuating and the NiFe film shows evidence of an induced uniaxial anisotropy. The NiFe film has an easy axis coercivity of about H_cₑ=2 Oe, and a hard axis saturation field (not shown) of about H_s = 5 Oe. After cooling to room temperature in an external magnetic field, the NiO spins are frozen and the interfacial magnetic interaction induces a unidirectional anisotropy on the NiFe film which shifts the NiFe hysteresis loops away from the zero field axis by an amount H_E. The direction of the shift depends on the orientation of the NiFe layer magnetization during field cooling. In addition to the loop shift, the interfacial interaction increases the coercivity dramatically. Perpendicular to the loop shift direction, the hard axis loop (not shown) passes
nearly linearly through zero with almost no coercivity. The $1/t_{\text{NiFe}}$ thickness dependence of $H_E$ and $H_{ce}$ expected from the interfacial origin of these effects, is well established.\textsuperscript{10}

The dependence of $H_E$ and $H_{ce}$ on the NiO layer thickness, on temperature, and on cooling field have been documented\textsuperscript{10,11} but are not well understood at a microscopic level.

In polycrystalline NiO films at room temperature and with constant NiFe overlayer thickness, $H_E$ and $H_{ce}$ are constant for NiO layer thicknesses above about 500Å. As the NiO layer thickness drops below about 350Å, $H_E$ begins to drop, reaching zero for thicknesses below about 175Å. $H_{ce}$ increases slightly for NiO layer thicknesses below 300Å, peaks near 175Å, and decreases to near zero below 100Å. The decrease in $H_E$ in bilayers with thin NiO thickness is not accounted for by a reduction in the Néel temperature due to finite size effects\textsuperscript{12} and instead indicates a length scale associated with the frozen AF spin configuration.

For NiO layers thicker than 400Å, $H_E$ drops with increasing temperature, reaching zero at about 200°C, which is called the blocking temperature $T_b$. When the NiO layers are thinner than 400Å, the blocking temperature is reduced.\textsuperscript{11} $T_b$ is thought to be associated with a thermal activation energy for the domain configuration in the NiO. Experiments have shown that the decrease in $H_E$ with temperature can be accounted for by assuming a distribution of blocking temperatures\textsuperscript{5} possibly indicating a distribution of domain sizes or anisotropy energies.

Typically NiFe is deposited on top of NiO to form a NiFe/NiO exchange couple. A bias field (20-200 Oe) is applied during deposition to induce a uniaxial anisotropy in the NiFe layer. It is thought that the interaction of the aligned NiFe spins at the interface with the NiO during deposition influences the AF spins in the NiO since the applied bias field is too weak to induce ordering in the NiO spins directly. In turn the NiO spin arrangement at the interface induces a unidirectional surface anisotropy in the NiFe. Heating bilayers above $T_b$ and cooling in a field has been reported to increase\textsuperscript{11} and to decrease\textsuperscript{10,13} $H_E$ relative to the as-deposited values. Whether the magnitude of the bias field or the cooling field strongly influences $H_E$ has not been well established.\textsuperscript{14}

Exchange couples which incorporate FeMn, NiMn, PdMn, IrMn, Pd-Pt-Mn, NiO, and
NiCoO antiferromagnetic layers are currently under study for use in magnetoresistive sensors and magnetoresistive and spin-valve based hard disk readback heads. The exchange anisotropy is employed to achieve the optimum sensitivity bias configuration in the sensor and to reduce noise by stabilizing domains. In this paper we focus on the oxide AF materials which share the same rocksalt crystal structure. The AF spin configurations and exchange coupling properties of the Mn-based materials are significantly different from the oxide materials and thus must be considered separately. The oxide films proposed for applications are polycrystalline with relatively small grain sizes. Achieving a clearer understanding of how magnetocrystalline anisotropy and texture influence the exchange anisotropy, however, requires that films with a high degree of crystalline perfection be examined as well.

The NiO spin structure is relatively simple, however the large number of domain configurations and domain walls in a multidomain sample make theoretical models of exchange anisotropy in NiO/NiFe bilayers considerably more challenging. NiO has a cubic FCC NaCl crystal structure above its Néel temperature. Below the Néel temperature there is a slight distortion of the NiO lattice in a ⟨111⟩ direction ($\Delta \ell/\ell \approx 4.5 \times 10^{-3}$). A strong negative uniaxial anisotropy accompanies the contraction, resulting in an easy plane defined by $K_1 \approx 1 \times 10^6$ erg/cm$^3$. Sheets of ferromagnetically aligned spins form in the (111) planes defined by the contraction axis with the Ni spins in neighboring sheets oppositely aligned. Within a (111) plane the direction of the spin axis is determined by a second 3-fold anisotropy ($K_3$) that is roughly three orders of magnitude weaker than $K_1$.

The AF domain configurations in NiO have been studied both experimentally and theoretically. There are four possible (111) directions in a NiO crystal from which the contraction axis may choose, and 3 spin directions once the contraction axis is defined. Thus there are 4x3=12 distinct possible AF domain configurations in NiO below $T_N$. Since the four (111) directions in the cubic NiO are nominally equivalent, local inhomogeneities determine which (111) axis becomes the contraction axis in different regions of the crystal. Applied magnetic fields and strain can make one (111) direction more kinetically favorable, and thereby influence the distribution of AF domains. The magnetic susceptibility of the NiO is largest.
parallel to the contraction axis, and so this axis tends to align parallel to strong applied 
fields. It has proven to be experimentally nearly impossible, however, to create a macro-
scopic NiO specimen with a single contraction axis by cooling in a magnetic field. Once the 
sample temperature has been lowered below the Néel temperature, domain walls become 
strongly pinned and extremely large fields are required to change the AF domain 
configuration.

In this study, we compare the magnetic properties of polycrystalline and epitaxial (001) 
NiO/NiFe bilayers deposited simultaneously. We also compare epitaxial (001) NiO/NiFe 
bilayers with the deposition bias field, $H_b$, aligned along different in-plane NiO crystalline 
axes. The results of these studies are interpreted in terms of induced anisotropies at the 
NiO/NiFe interface.

II. EXPERIMENTAL METHODS

The single films and bilayers were grown in a multilayer deposition system using ion 
beam sputtering (IBS). The system has been described in detail elsewhere. Single crystal 
polished (001) oriented MgO substrates and Si substrates with native oxide layers were 
placed side by side in substrate holders with bias magnets. The bias magnets produce 
a uniform magnetic field, $H_b$, of about 300 Oe at the substrate surface. The substrate 
temperature was monitored but not controlled and reached about 80°C during deposition. 
The NiO layers were grown using a new IBS process. Briefly, this new process is simpler 
than the more widely used reactive sputtering technique in that an oxygen partial pressure 
is not required during NiO deposition. The NiO is deposited by directly sputtering a NiO 
target with a neutralized 750V, 30 mA Ar-ion beam which produces a deposition rate of 
about 0.2 Å/sec. The metal NiFe layers were then grown immediately on top of the NiO 
using a 500V, 20 mA ion beam without neutralization. The Ar gas pressure during deposition 
was about 0.25 mTorr. The Ni:O ratio of films produced using this process was measured 
using Rutherford back-scattering and was determined to be 1:1 to within 1%. NiFe films
were deposited from a Fe$_{19}$Ni$_{81}$ sputter target.

The morphology of the NiO/NiFe bilayers was probed using x-ray diffraction (XRD). Symmetric x-ray scans were performed on a 18 kW Rigaku rotating anode diffractometer with a scattered beam monochromator using Cu K$_\alpha$ radiation. Phi scans were performed on a four-circle goniometer using Cu radiation at Stanford University.

The magnetic properties of the bilayer films were measured with a vibrating sample magnetometer (VSM) equipped with two sets of orthogonal pick-up coils. The use of two sets of pick-up coils allows the simultaneous measurement of both the longitudinal and transverse magnetization curves, as is often done with Kerr magnetometry. The magnetization curve of a 500 Å thick NiFe film grown on MgO (001) and Si (not shown) shows that H$_b$ applied during deposition induces a uniaxial anisotropy in the NiFe. The value of the uniaxial anisotropy is determined from the hard axis saturation field, H$_s$, to be K = 2000 erg/cm$^3$ given that H$_s$ = 2K/M$_s$ in the polycrystalline NiFe film. In addition to K we also observe a four-fold magnetocrystalline anisotropy in the epitaxial NiFe film of about $K_1 = -500$ erg/cm$^3$ ($H_s = 2(K+K_1)/M_s$ for a ⟨110⟩ hard axis in the (001) plane). These values are consistent with those expected for NiFe films.

III. EXPERIMENTAL RESULTS

A comparison of the XRD spectra for NiO/NiFe bilayers grown simultaneously on MgO (001) and oxidized silicon is shown on Figure 2. The films deposited on oxidized Si wafers are polycrystalline as shown by the presence of (111), (002) and (022) NiO Bragg peaks. The average grain size calculated using the Scherrer formula from the full width at half maximum (FWHM) of the peaks is 100Å to 200Å. Because the NiO and MgO crystal structures are nearly identical with only slightly different lattice parameters, (MgO: a=4.213Å, NiO: a=4.177Å or 0.9% difference) the NiO (002) Bragg peak of the film on MgO is obscured under the strong substrate peak. No NiO (111) or (022) Bragg peak intensity was observed in the XRD spectra of the bilayer grown on MgO, however. Instead a strong (002) peak
from the NiFe deposited on the NiO was present with a correlation length limited by the
thickness of the film (100Å) and a rocking curve width of 1°-2° FWHM. Phi scans at the NiFe
(011) peak position show that the NiFe layer grown on top of the NiO layer is epitaxially
oriented relative to the MgO substrate (Figure 2b). The epitaxy of the NiFe shows that
the intermediate NiO layer is also oriented in-plane with respect to the MgO substrate. As
discussed in the previous section, NiFe films grown directly on MgO (001) were also found
to be epitaxial. Comparison of Kiessig fringes in the low-angle symmetric XRD spectra (not
shown) indicate that the interfaces of the epitaxial bilayer are rougher (8-12 Å) than the
polycrystalline bilayer (2-3 Å).

The hysteresis loops of polycrystalline and epitaxial NiO (500Å)/NiFe (100Å) bilayers
grown simultaneously are shown in Figure 3. In the epitaxial film the bias field during depo-
sition \( H_b \) was applied along a MgO [100] axis. Hysteresis loops parallel and perpendicular to
\( H_b \) are shown. The polycrystalline bilayer (Fig. 3a) illustrates the usual exchange anisotropy
behavior: there is a shift in the easy axis hysteresis loop of \( H_E = 52 \) Oe and an increase in
the NiFe coercivity from its free value of about \( H_{ce} = 2 \) Oe to \( H_{ce} = 30 \) Oe. The hard axis
loop shows almost zero coercivity and saturates at about \( 2H_E \). The loop parallel to \( H_b \) for
the epitaxial bilayer (Fig. 3b) shows a shift of \( H_E = 20 \) Oe and a coercivity of \( H_{ce} = 26 \)
Oe. The shape of the hard axis magnetization (perpendicular to \( H_b \) ) in the epitaxial film is
qualitatively different from the nearly linear hard axis loop observed in the polycrystalline
films. Figure 3c shows transverse magnetization data for the polycrystalline and epitaxial
films with \( H \perp H_b \), and indicates the nonlinearity in the epitaxial bilayer’s hard axis loop is
due to a nearly 90° reorientation of the magnetization vector.

In figure 4 another set of magnetization data for polycrystalline and epitaxial NiO/NiFe
bilayers grown simultaneously is shown but now with \( H_b \), the bias field applied during
deposition, aligned with an in-plane MgO [110] axis. The polycrystalline bilayer (Fig. 4a)
has an exchange field of \( H_E = 66 \) Oe and an easy-axis coercivity of \( H_{ce} = 34 \) Oe. The hard
axis loop once again saturates at about \( H_s = 2H_E \) and has coercivity less than 1 Oe. The
epitaxial bilayer (Fig. 4b) has \( H_E = 36 \) Oe, and \( H_{ce} = 42 \) Oe. The hard axis magnetization
data shown in figure 4b and 4c shows similar behavior to that seen in figure 3b and 3c. The similarity between figure 3b and 4b reveals that the nonlinearity observed in the hard axis magnetization curves of the epitaxial films is induced by \( H_b \) and is not referenced to the underlying crystal structure of the NiO. The variation in \( H_E \) for the polycrystalline samples shown in figures 3a and 4a results from uncontrolled variations in the deposition conditions and serves as a measure of the run-to-run reproducibility of the growth.

We have grown epitaxial bilayers in reverse order to better understand why this configuration typically shows lower exchange anisotropy than do bilayers with NiO on the bottom. Figure 5 shows easy axis magnetization loops for NiFe 100Å/NiO 500Å bilayers grown simultaneously on oxidized silicon and MgO (001) substrates. In this configuration the choice of substrate has much less effect on the hysteresis loop. The exchange shift is \( H_E = 13 \) Oe and \( H_{ce} = 5 \) Oe in both films. XRD shows that both NiO films are polycrystalline. These data demonstrate that the growth mode of NiO on NiFe is significantly different than that of NiFe on NiO, which explain the difference in the exchange anisotropy observed in the two configurations. Recently, however, large bias fields (0.04 erg/cm\(^2\)) were reported in “top” spin valves grown with the NiO layer on top of a NiFe layer using reactive RF sputtering.

IV. ANALYSIS

In order to model the field dependence of the NiFe magnetization, we start with the simplest energy equation that contains only a unidirectional anisotropy term and a Zeeman term describing interaction with the external field. The energy equation takes the form:

\[
E/M = -H \cos(\theta) - H_E \cos(\theta - \phi)
\]

where \( H_E \) is the effective unidirectional anisotropy field, \( \phi \) is the angle between the bias field \( H_b \) and the applied field \( H \), and \( \theta \) is the angle between \( H \) and the magnetization. (This form ignores the induced uniaxial anisotropy in the NiFe layer, which is small compared to \( H_E \).) Assuming the magnetization reorients by rotation following the minimum energy solution, the hard axis magnetization is:
\[
\frac{M(H)}{M_s} = \frac{H}{\sqrt{H^2 + H_E^2}}
\]  

(2)

Under these assumptions, the easy axis magnetization should have zero coercivity and change sign at \( H = H_E \), and the hard axis magnetization should approach saturation asymptotically. The best fit of equation 2 to the measured hard axis magnetization for the polycrystalline film in figure 3a predicts \( H_E = 83 \) Oe which is inconsistent with the measured easy axis value of \( H_E = 52 \) Oe. By increasing the size of uniaxial anisotropy above the usual value for soft NiFe alloys, we can consistently fit the easy and hard axis behavior observed in figure 3a, and 4a, and qualitatively account for the easy-axis coercivity.

\[
E/M = -H \cos(\theta) - H_E \cos(\theta - \phi) + H_K \cos^2(\theta - \phi)
\]  

(3)

The predicted analytical form of \( M(H) \) is complicated. However, we can consistently fit the easy and hard axis behavior with \( H_E = 53 \) Oe and \( H_K = 30 \) Oe, given that equation 3 predicts the magnetization approaches saturation with \( H_s \approx 2(H_E + H_K) \). The uniaxial term is significantly larger than that observed in single films of NiFe (\( H_K \) of NiFe = 5 Oe).\(^{31}\) The increase of \( H_K \) comes from the interfacial interaction with the NiO. Strain induced at the NiO/NiFe interface may be a source of uniaxial anisotropy, however, the small magnetostriction of the permalloy, combined with the small tetragonal distortion of the NiO below its Néel temperature make this an unlikely explanation for the large uniaxial anisotropy observed here.

The large uniaxial term in the energy equation needed to consistently model the easy and hard axis data helps to account for the coercivity in the easy axis loop. It is well known that, in the presence of a uniaxial term, the energy equation contains local energy minima in addition to global minima for a range of applied fields.\(^{34, 35}\) Local energy minima can pin the magnetization and temporarily delay the obtainment of the absolute energy minimum configuration. Since the magnetization loops show that NiFe moment reverses by rotation and follows the local energy minimum, the easy axis coercivity predicted by equation 3 is \( H_{ce} = H_K = 30 \) Oe. In single NiFe films, however, \( H_{ce} < H_K \) indicating the reversal occurs
through domain wall motion rather than rotation. The easy axis energy surfaces predicted by equation 1 do not contain local minima, however, those by equation 3 do. Thus the static interfacial interaction at the NiO/NiFe surface anisotropy can account for much of the coercivity observed in the easy axis loop.

It is interesting to note that the ratio of the $H_E$ to $H_{ce}$ for a wide range of IBS polycrystalline NiO/NiFe bilayers appears to have a characteristic maximum value. Figure 6 shows $H_E$ plotted vs. $H_{ce}$. The dotted line is a guide to the eye showing $H_E = 1.8 H_{ce} + 14$ Oe. Fig. 6 implies that increases in $H_E$ and $H_{ce}$ come together in the best polycrystalline NiO/NiFe bilayers. The data in figure 6 imply that $H_E$ and $H_K$ in equation 3 do not vary independently in NiO/NiFe bilayers. On the other hand, films with small $H_E$ and large $H_{ce}$ occur since there are many sources of coercivity in thin NiFe films, many not directly related to the surface exchange interaction. The highest $H_E$ to $H_{ce}$ ratio in NiO/NiFe bilayers published in the literature,\textsuperscript{10,11} deposited using reactive sputtering is approximately 2.2, which is similar to the value observed in the IBS films.

The slope of the line in figure 6 depends on the intrinsic anisotropies present in NiO. The $H_E/H_{ce}$ ratios we observe in coupled IBS NiFe/NiCoO bilayers typically lie above this line. Further, a typical $H_E/H_{ce}$ ratio for NiFe/FeMn bilayers is 25.\textsuperscript{36} The higher ratio observed in general in Mn-based AF exchange couples may be due to the higher magnetocrystalline anisotropy or the reduced symmetry of the Mn-based antiferromagnets.\textsuperscript{11} These differences produce a interface anisotropy that is closer to pure unidirectional in FeMn/NiFe bilayers compared to the unidirectional plus uniaxial anisotropy found in NiO/NiFe bilayers.

Turning now to the magnetization observed in the epitaxial bilayers in figures 3b, and 4b, the shape of the hard-axis magnetization curves can be predicted by adding a cubic anisotropy to the energy equation (1):

$$H_{k1} \sin^2(\theta - \phi) \cos^2(\theta - \phi)$$

(4)

The data in figure 3b are reasonably well reproduced with $H_E = 20$ Oe, $H_K = 30$ Oe. The cubic anisotropy produces an energy minimum perpendicular to the unidirectional anisotropy
(and to $H_b$) which qualitatively changes the hard axis loop shape.

In addition, the presence of a cubic anisotropy produces local energy minima in the energy surface describing the bilayer magnetization reversal. As discussed previously, these minima can be used to qualitatively account for the coercivity observed in the hard axis magnetization loop. Qualitatively, as $H$ decreases from a large positive value, the NiFe layer moment at first remains in a local energy minimum parallel to $H$, and then shifts suddenly from that minimum to the energy minimum derived from the unidirectional anisotropy term, perpendicular to $H$ and parallel to $H_b$. Transverse magnetization data (Fig. 3c, 4c) for $H \perp H_b$ reinforce this description. As the longitudinal magnetization ($M_x$) decreases, the transverse magnetization ($M_y$) increases abruptly and reaches a plateau as the NiFe layer moment settles into the global energy minimum perpendicular to $H$. This is in contrast to the transverse hard-axis behavior of the polycrystalline bilayer couple which shows a smooth rotation of the NiFe layer moment and no plateau.

Calculated magnetization curves that qualitatively reproduce the experimental data for the epitaxial bilayers are shown in figure 7a,b. The curves were calculated using an energy equation with a unidirectional and a cubic anisotropy:

$$E/(M \ast H_E) = -H/H_E \cos(\theta) - \cos(\theta - \phi) + H_{K1}/H_E \cos^2(\theta - \phi) \sin^2(\theta - \phi)$$

where $H_{K1}/H_E = 1.5$. The magnetization in Figure 7a is assumed to reverse by rotation and to find the absolute minimum energy configuration. The calculation reproduces the steps observed in the epitaxial hard axis loops. As in the case of the polycrystalline bilayers, if we assume the vector magnetization sticks in local energy minima and only achieves the absolute minimum when its path is unobstructed by an energy barrier, we can qualitatively account for the coercivity observed in the easy and hard axis loops as shown in figure 7b.

From the similarity of the hard axis loops in figure 3b,c and 4b,c, where the bias field is applied along a different NiO crystal axis, it is clear that the cubic term is induced by the bias field applied during deposition, and is not referenced to the NiO or the NiFe crystal axes. Thus the data are not consistent with a magneto-crystalline anisotropy in the NiFe
or the NiO. In contrast, epitaxial NiFe films deposited directly on MgO (001) show induced bulk uniaxial and cubic magnetocrystalline anisotropy terms which are nearly an order of magnitude smaller than those needed to describe the NiO/NiFe loops. The cubic anisotropy must arise from the same interfacial interaction with the NiO that produces the exchange anisotropy. Ferromagnetic resonance or Brillouin light-scattering measurements on these bilayer films may give a more quantitative determination of the anisotropy values.

The above analysis assumes the NiFe/NiO interfacial anisotropy is unmodified by rotation of the NiFe moment during magnetization reversal. This assumption is strictly false. The observation of a training effect and the presence of rotational hysteresis in NiFe/NiO bilayers even at very high applied fields\(^3\) clearly show that NiO spin dynamics are present during the NiFe reversal. However, a static induced surface anisotropy does describe many of the main features of the NiFe/NiO magnetization curves. The contributions of irreversible NiO spin dynamics on the NiFe loop are second-order effects. More experimental work is needed to measure and understand the NiO spin dynamics during the NiFe magnetization reversal process.

Once again it is interesting to compare the behavior of NiFe/NiO and NiFe/FeMn exchange couples. As with NiO/NiFe exchange couples, there is a strong deposition order dependence in NiFe/FeMn exchange couples. However, in this case it is the NiFe that should be deposited first in order to achieve a large exchange bias\(^{38}\). The order dependence of NiFe/FeMn exchange bias has been found to arise from changes in growth mode when the order of deposition is reversed. The (111) textured NiFe surface serves as a template for the antiferromagnetic \(\gamma\) phase of FeMn. In the absence of the NiFe template the FeMn does not achieve the \(\gamma\) phase and instead forms in the nonmagnetic \(\alpha\) phase, and no exchange bias is observed. In experiments where the \(\gamma\) FeMn was stabilized through epitaxy with a single crystal substrate, exchange bias is observed in NiFe deposited on top.\(^{6}\) Further, when the \(\gamma\) FeMn was grown in different crystalline orientations, exchange bias in the NiFe grown on top was observed in every case.\(^{6}\) The ratio of \(H_E\) to \(H_{ce}\) for the FeMn/NiFe bilayers was different for the different crystalline orientations, however.\(^{6}\) Changing the order of NiO/NiFe...
bilayer deposition does not change the crystalline phases of the individual layers, but instead produces more subtle changes at the interface that lead to differences in the exchange anisotropy.

V. DISCUSSION

There are two requirements for achieving shifts in the hysteresis loop of a ferromagnetic (FM) film deposited on an antiferromagnetic (AF) film and these requirements are sometimes at odds. First, there must be an uncompensated interaction at the interface. Second, spins in the AF layer must remain pinned as the FM film undergoes a reversal. The most obvious way to produce a magnetically uncompensated surface in the NiO is to terminate the NiO layer on the (111) face normal to the contraction axis. These (111) planes contain sheets of aligned spins. The anisotropy within this (111) plane is weak, however, and so the NiO spins may not be strongly pinned and may rotate within this plane during a NiFe reversal. In addition, the presence of domain walls in the NiO layer perpendicular to the interface will increases the degree of compensation due to averaging of the spin orientations over the surface. The presence of interface roughness makes it still more difficult to predict if the surface interaction will be uncompensated. The in-plane anisotropy is strongest when the contraction axis is forced into the plane of the film, but in this orientation the bulk NiO spin configuration predicts the surface will be compensated even at very short length scales. In polycrystalline NiO films, NiO spin rotations and domain wall dynamics are strongly influenced by grain boundaries and crystalline defects, in addition to the intrinsic magnetocrystalline anisotropy.

Our results clearly show that the crystalline structure of the NiO in NiO/NiFe thin film exchange couples does not strongly influence the spin structures that are responsible for the uncompensated interaction. Instead, it is the influence of the applied field indirectly through alignment of the NiFe layer and the subsequent NiFe/NiO interfacial interaction which determine the spin distribution at the NiO/NiFe interface. Coupling at the interface
between the NiO and the aligned NiFe layer must force the Ni spins in the NiO on average to be collinear, parallel to \( H_b \). These interactions force the spins at the surface of the NiO layer to distort producing an uncompensated interface nearly independent of the crystalline orientation or morphology at the interface. In other words, the interfacial NiO spin configuration is not the same as the bulk NiO spin configuration. The distorted spin configuration at the NiO surface must also be strongly coupled to the bulk of the NiO layer, anchoring the surface spins, so they do not significantly reorient during subsequent ferromagnetic reversals once the configuration is frozen in. This picture is consistent with the one presented by Schlenker\textsuperscript{40} and Stoecklein\textsuperscript{38} who suggest the frustrated interactions at the AF/F interface may be similar to those in a spin glass.

It is not consistent to assume that the formation of an aligned NiFe layer at the NiO surface during deposition can distort the NiO interfacial spins to create an uncompensated interaction, but yet the distorted configuration remains frozen during subsequent post deposition NiFe reversals. The time scale for the NiO spins dynamics must therefore be shorter during deposition than during routine room temperature magnetic measurements. It follows that the temperature during deposition is elevated close enough to \( T_N \) to allow the distortion of the surface NiO spin configuration during the short time scale of a deposition. Thus one could predict that NiFe deposition onto a cooled NiO thin film would produce no loop shift. In order to address this hypothesis, we have deposited IBS NiO/NiFe bilayers on substrates clamped to a thick Cu plate that is cooled to -120\(^\circ\)C. Large loop shifts were still observed in these films. The distorted spin region described in the previous paragraph that creates the uncompensated NiO surface may be relatively thin, and thus only the temperature at the surface of the growing film need approach \( T_N \) in order for a loop shift to be observed in the as deposited films. It is likely that even when the substrates are cooled from the back side, energy transported to the film surface by deposited material raises the surface temperature above \( T_N \). A more effective test of the above hypothesis would employ a low energy NiFe deposition technique such as evaporation in conjunction with substrate cooling. Stoecklein suggests that simply the presence of the AF/F interface produces the spin frustration needed
to produce a loop shift, however this model cannot explain the persistence of the loop shift during post deposition NiFe reversals.

It is easy to see that in as-deposited bilayers in which effectively only a thin surface layer has been field-cooled through $T_N$, we produce a different distorted NiO spin configuration than in field-annealed bilayers. In this way we can qualitatively account for changes in as-deposited and field-annealed $H_E$. During reverse-order deposition (NiFe first), the thermal development of the NiO surface layer will be different than during forward-order (NiO first). One can speculate that the presence of the thick metal NiFe layer will more efficiently cool the interface during reverse-order deposition, resulting in a thinner distorted layer and a smaller $H_E$. In analogy with spin glass models, the NiO/NiFe interface may possess a large number of nearly equal energy configurations, each producing a unique value of $H_E$. In spin glasses, configurations are separated by large energy barriers confining the system to a small region in phase space for short time scales. Changing the thermal and magnetic history of the interface, allows the spin configuration to quickly explore different local minima.

Clearly exchange couples based on polycrystalline rather than epitaxial NiO layers show the most promise for applications. In polycrystalline exchange couples the increased disorder of the NiO spins leads to larger exchange bias and smaller easy axis coercivity than in epitaxial films. The larger interface roughness in the epitaxial bilayers relative to the polycrystalline ones may account for the observed differences in the magnitude of the exchange field. The higher degree of crystalline perfection in the epitaxial bilayers leads to more complicated induced surface anisotropy. Stronger pinning in polycrystalline films may reduce the dynamics in the NiO spins and thus reduce the observed coercivity, however, our data indicate the easy axis coercivity observed in NiO/NiFe bilayers is primarily due to the static induced surface anisotropy. The ideal exchange anisotropy material for applications would have a non-hysteretic hard axis loop and an easy-axis loop with a large exchange bias and $H_E > H_{ce}$.

Our observations on epitaxial oxide-AF/NiFe exchange-coupled bilayers are consistent with those of Carey et al. who have done extensive characterization of exchange couples.
using epitaxial NiO, NiCoO films and NiO/CoO multilayers grown by reactive magnetron sputtering. They report consistently smaller $H_E$ in epitaxial relative to polycrystalline bilayers deposited under similar conditions. They also observe exchange anisotropy in both (111) and (001) oriented epitaxial bilayers, with consistently larger coercivity in the (111) relative to the (001) oriented films. Lai et al. report loop shifts in bilayers with epitaxial (001) and (111) NiO films grown by metal-organic chemical vapor deposition (MOCVD). They observe unusually large and nearly isotropic coercivity in both (001) and (111) oriented MOCVD based bilayer couples, however.

**VI. CONCLUSIONS**

We have shown that polycrystalline NiO/NiFe bilayers produce larger loop shifts than epitaxial bilayers deposited simultaneously. It appears that a larger surface anisotropy can be induced at a polycrystalline relative to an epitaxial interface. The presence of exchange anisotropy in (001)-oriented epitaxial NiO/NiFe layers shows that an uncompensated interface is produced independent of the NiO crystalline orientation. Our data show that in addition to the induced surface unidirectional anisotropy, an induced cubic surface anisotropy is needed to consistently model the hysteresis loops measured in epitaxial NiO/NiFe bilayers. Hysteresis loops of polycrystalline bilayers are most accurately modeled by an induced surface unidirectional plus uniaxial anisotropy. The induced surface anisotropies we observe are referenced only to the bias field applied during deposition, and are independent of the NiO crystal structure.

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FIGURES

FIG. 1. Hysteresis loops of the same polycrystalline NiO500Å/NiFe100Å bilayer film are shown, one at a temperature below the blocking temperature, $T_b$, of NiO and one above $T_b$. Above $T_b$, the NiFe behaves as a free layer, magnetically the same as a NiFe layer deposited on a non-magnetic substrate. Below $T_b$ the interfacial exchange interaction induces a surface anisotropy which shifts the NiFe loop away from the zero field axis and raises its coercivity.

FIG. 2. Comparison of x-ray spectra from NiO 500Å/NiFe 100Å bilayer films deposited simultaneously on polished single crystal MgO (001) and amorphous substrates. The bottom scan in a) shows that the NiO on the oxidized silicon substrate is polycrystalline with a grain size of approximately 150Å. The top scan shows the NiO (111) and (022) Bragg peaks are absent in the bilayer grown on MgO. The MgO (002) substrate peak obscures the presence of the NiO (002) peak. A strong reflection is present from the NiFe (001) planes. In b), phi scans at the NiFe (022) and the MgO (022) Bragg angles are shown. The NiFe layer is epitaxially oriented relative to the MgO substrate, confirming that the NiO layer is also epitaxial.

FIG. 3. Magnetization data for two NiO 500Å/NiFe 100Å bilayer films deposited simultaneously. In a) the easy-axis ($H$ parallel to $H_b$, the bias field during growth) and hard-axis ($H$ perpendicular to $H_b$) magnetization curves of the polycrystalline bilayer couple is shown. The easy-axis loop is shifted by $H_E = 52$ Oe due to interfacial exchange anisotropy with the NiO. b) shows the same measurement as in a) for an epitaxial (001) bilayer deposited on MgO. The bias field, $H_b$, applied during deposition was aligned parallel to an in-plane MgO (100) axis. The easy-axis loop is shifted by $H_E = 20$ Oe. Discontinuities in the hard-axis loop reveal the present of a cubic induced anisotropy term that produces a local energy minimum parallel to the applied field and perpendicular to $H_b$. In c) the transverse hard axis magnetization, $M_y$, for the polycrystalline (open circles) and epitaxial (filled circles) are compared. The smooth curve of the polycrystalline $M_y$ loop shows the magnetization vector rotates continuously as the applied field varies. The plateau in the epitaxial $M_y$ loop confirms that the NiFe moment turns discontinuously from a local energy well parallel to the applied field to the deep unidirectional well perpendicular to it.
FIG. 4. The same measurements as shown in figure 3 for a second set of simultaneously de-
posited polycrystalline and epitaxial bilayers except that here $H_b$, the bias field during deposition,
was applied parallel to an in-plane (110) axis. For the polycrystalline films in a) the easy-axis loop
is shifted by $H_E = 66$ Oe. In b) the epitaxial film has $H_E = 36$ Oe. c) shows the transverse hard
axis magnetization loops. The data are qualitatively similar to those in figure 3, particularly the
observation of discontinuities indicating a cubic anisotropy with minima referenced to the bias field
axis. Thus the cubic anisotropy is induced by the bias field and is not influenced by the orientation
of $H_b$ relative to the NiO crystal axis.

FIG. 5. Easy axis magnetization for two bilayer films deposited in reverse order simultaneously
on MgO and an oxidized silicon substrate. Both NiO layers grown on NiFe were polycrystalline.
The exchange bias fields and coercivities of the two films are the same. Differences in the growth
of NiO on NiFe compared to NiFe on NiO lead to the reduced exchange anisotropy observed in
these films.

FIG. 6. The exchange anisotropy field, $H_E$ for a wide variety of NiO-500Å/NiFe-$t_{NiFe}$ films
plotted vs. the easy-axis coercivity, $H_{ce}$. The dotted line is a guide to the eye indicating the
relationship $H_E = 1.8 H_{ce} + 14$ Oe, so the $H_E$ to $H_{ce}$ ratio has a limiting value of about 1.8. This
value appears to be a characteristic of the AF material since the ratio observed for NiCoO/NiFe
bilayers typically exceeds this value. AF/NiFe bilayers using Mn-based antiferromagnetic layers
greatly exceed this value, giving much less coercivity per unit exchange anisotropy shift.

FIG. 7. Calculated easy axis (dashed) and hard axis (solid) magnetization loops using an energy
equation with a unidirectional and cubic anisotropy (equation 5). In a) we assume the magnet-
ization achieves the absolute minimum energy configuration. In b) we assume the magnetization
remains in local minima until the path to the absolute minimum is unobstructed by an energy
barrier. The calculations qualitatively reproduce the features observed in the epitaxial NiO/NiFe
bilayers.