Strong interlayer coupling in CoO/NiO antiferromagnetic superlattices

M. J. Carey and A. E. Berkowitz
Department of Physics and Center for Magnetic Recording Research, University of California at San Diego, La Jolla, California 92093-0401

J. A. Borchers and R. W. Erwin
National Institute of Standards and Technology, Gaithersburg, Maryland 20899
(Received 29 September 1992)

A strong interlayer-exchange interaction is observed in short-repeat-distance CoO/NiO superlattices. This was investigated by measuring the exchange-anisotropy properties of superlattice CoO/NiO/$Ni_{81}Fe_{19}$ exchange couples and by examining NiO/CoO superlattices with neutron diffraction. Short-repeat-distance superlattices show a single ordering temperature which depends only on the [CoO]:[NiO] ratio. The magnetocrystalline anisotropy of the monoxide layers is strongly influenced by the interlayer coupling.

Epitaxial superlattices of ionic antiferromagnets (e.g., CoO/NiO and FeF_2/CoF_2) with short superlattice repeat distances exhibit single Néel temperatures (T_N) between the T_N's of the constituent layers.[1,2] Although any interlayer magnetic coupling is limited to the layer interfaces by the superexchange interaction, these results suggest that this interaction can have a strong effect far from these interfaces. The ordering behavior of magnetically coupled superlattices has recently been the subject of much interest both experimentally[3,4] and theoretically.[5,6] In the present study, the magnetic behavior of CoO/NiO superlattices and bilayers was examined by measuring the exchange-ansiotropy properties of CoO/NiO/$Ni_{81}Fe_{19}$ exchange couples, and CoO/NiO superlattices were investigated by neutron diffraction. These data determine the range of the interlayer interaction and the influence of this interaction on the magnetocrystalline anisotropies of the constituent layers.

Exchange anisotropy refers to the interfacial magnetic interaction between, for example, a ferromagnet (F) and an antiferromagnet (AF).[7,8] This often results in a shift of the hysteresis loop of the ferromagnetic film away from the zero-field axis. The magnitude of this loop shift is called the exchange field H_e, and is related to the strength of the magnetic interaction across the F-AF interface. Malozemoff[9] and Mauri *et al.*[10] have predicted that $H_e \propto \sqrt{AK}$, where A and K are the exchange constant and magnetocrystalline anisotropy of the antiferromagnet, respectively. H_e typically follows a linear $(1-T/T_b)$ temperature dependence,[11,12] where the blocking temperature (T_b) is the temperature above which H_e is zero. Typically, $T_b \leq T_N$. With the assumption that $H_e \propto \sqrt{AK}$ and $K(T)=K(0)(1-T/T_b)^2$ for cubic-anisotropy antiferromagnets, Malozemoff[13] has shown that $H_e \propto \sqrt{AK(0)}(1-T/T_b)$. Thus the linear temperature dependence of H_e follows from the cubic anisotropy of the antiferromagnet. Therefore, information about the ordering temperature and anisotropy of the antiferromagnet, which may be difficult to measure directly, can be inferred from the temperature dependence of the exchange anisotropy induced in the ferromagnet layer.

Of bulk CoO and NiO, CoO has the higher crystalline anisotropy in the (111) parallel-spin plane ($K_2 \lesssim 2\times10^5$ erg/cm^3 at 4.2 K) (Ref. 14) and lower Néel temperature ($T_N=293$ K),[14] while NiO has the lower crystalline anisotropy ($K_2=3.3\times10^2$ erg/cm^3 at 300 K) (Ref. 16) but a high T_N (523 K).[15] In this study, two oxide structures were produced by reactive sputtering from separate Co and Ni sources: CoO/NiO superlattices and CoO/NiO bilayers. The argon pressure was 2.00 m torr. During the oxide superlattice deposition, the Co and Ni sources were operated simultaneously and alternately shuttered. The oxygen partial pressure was 0.38 m torr before and typically 0.20 m torr during sputtering. For the oxide bilayers, the sources were operated sequentially with an oxygen partial pressure of 0.20 m torr before and 0.09 m torr during sputtering. The substrates were heated to 373 K before deposition. A rotating substrate table moved the substrates over each source each revolution, resulting in about 0.5 Å deposited per revolution. Both polycrystalline and epitaxial (111) films were produced under identical conditions. Films produced on Si(100) substrates were polycrystalline due to the amorphous native oxide layer on the Si. Transmission electron microscopy showed these films had an average grain size of about 200 Å. Epitaxial (111) monoxide superlattices were grown on (0001) α-Al_2O_3 substrates.[17] In both the epitaxial and polycrystalline films, only monoxide phases were detected by x-ray and neutron diffraction. For the exchange-anisotropy studies, a 300-Å layer of ferromagnetic $Ni_{81}Fe_{19}$ was deposited immediately after the oxide deposition, with the substrates backed by alnico magnets to induce an easy axis in the $Ni_{81}Fe_{19}$. While the temperature dependence of H_e was the same for polycrystalline and (111) couples, H_e was greater and the coercive force smaller in the polycrystalline couples. Thus polycrystalline couples were chosen for the exchange-anisotropy measurements. The neutron-diffraction studies were performed on epitaxial (111) monoxide superlattices due to the significantly higher diffraction peak intensities from

oriented films.

Magnetic measurements of the exchange couples were taken in flowing argon with a vibrating sample magnetometer. The easy axis of the $Ni_{81}Fe_{19}$ was parallel to the measuring-field direction. Each sample was cycled through enough hysteresis loops to minimize the effects of magnetic training.[18,19] H_e of the exchange couples was measured as a function of temperature in samples after cooling the samples from above the oxide T_N (T_b+50 °C) in a 3-kOe applied field. H_e typically increased by 3–5 Oe after the first field cooling, but no other significant changes were noticed.

A series of exchange couples was prepared using CoO/NiO superlattices with an overall CoO concentration of 55 vol%. Three superlattice periods were prepared: $[CoO(40\ Å)NiO(33\ Å)]_7$ ($\Lambda=73$ Å), $[CoO(20\ Å)NiO(16\ Å)]_{14}$ ($\Lambda=36$ Å), and $[CoO(10\ Å)NiO(8\ Å)]_{28}$ ($\Lambda=18$ Å) (where the number in the parentheses is the constituent layer thickness and the subscript is the number of bilayers). The number of superlattice periods was adjusted to keep the total film thickness constant at 510 Å. Two samples of each Λ were produced: one with CoO interfacing the $Ni_{81}Fe_{19}$ (CoO on top) and one with NiO interfacing the $Ni_{81}Fe_{19}$ (NiO on top). A $Co_{0.55}Ni_{0.45}O/Ni_{81}Fe_{19}$ exchange couple was also produced to compare alloy and multilayer properties. H_e vs T for this series is shown in Fig. 1. With the exception of the $\Lambda=73$ Å, CoO-on-top sample (which will be discussed below), all of the superlattice samples have the same blocking temperature as the $Co_{0.55}Ni_{0.45}O/Ni_{81}Fe_{19}$ sample, 378 K, *independent of whether low T_N CoO or high T_N NiO interfaces the $Ni_{81}Fe_{19}$*. This suggests that the CoO and NiO interfacial coupling has a strong, long-ranged effect which results in a single, intermediate T_N for the entire superlattice. The $\Lambda=73$ Å superlattices behaved differently. The $\Lambda=73$ Å CoO-on-top sample had $T_b=308$ K—near that of $Ni_{81}Fe_{19}$ on pure CoO.[12] The $\Lambda=73$ Å NiO-on-top sample showed considerable

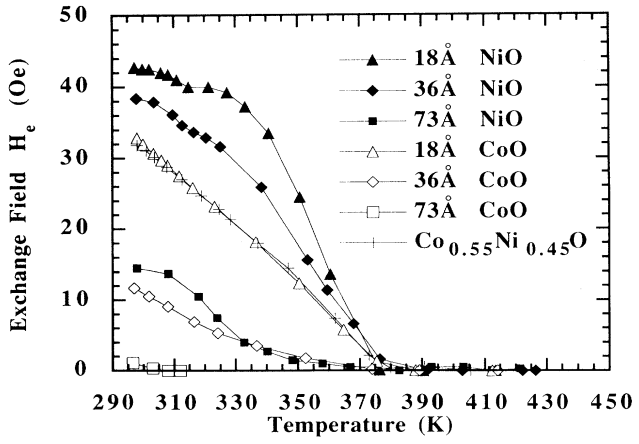

FIG. 1. The exchange field H_e as a function of temperature for the 510-Å superlattice CoO/NiO/300-Å $Ni_{81}Fe_{19}$ couples with 55-vol% CoO. The legend indicates the superlattice period and whether a CoO or NiO layer interfaces the $Ni_{81}Fe_{19}$.

magnetic training,[18,19] with H_e dropping 11 Oe (from 25 to 14 Oe) after repeated hysteresis loops, compared to a 1–2-Oe drop for the shorter-wavelength samples. Also, the easy direction of this sample could be rotated with the application of a field of only 100 Oe for 1 min at room temperature. These data suggest that the effect of the oxide-oxide interfacial coupling is much weaker in the long-wavelength samples than the short-wavelength samples.

The complex nature of the effects of the CoO/NiO interfacial coupling is made apparent by other trends in the H_e vs T data in Fig. 1. First, the temperature dependence of H_e is quite different for the CoO-on-top samples than the NiO-on-top samples. The CoO-on-top samples show the linear temperature dependence of H_e that is characteristic of cubic anisotropy antiferromagnets.[13] This would suggest that the coupling with the NiO raises the T_N of the CoO layers without affecting the temperature dependence of the cubic anisotropy. The NiO-on-top samples, however, show a more complicated H_e vs T behavior. For these samples, the slope of H_e vs T is small near room temperature. A kink in H_e vs T is present at 313 K, after which the slope decreases sharply. This temperature dependence is a distinct departure from the linear behavior expected for cubic-anisotropy antiferromagnets, indicating a more complex ordering. Since the 313-K kink is near the T_b of the $CoO/Ni_{81}Fe_{19}$ exchange couples,[12] it seems likely that the kink and slope change are due to a weakened magnetocrystalline anisotropy of the superlattice above the ordering temperature of the CoO. Second, the NiO-on-top samples have higher H_e's than the corresponding CoO-on-top samples. $NiO/Ni_{81}Fe_{19}$ couples have a significantly higher room-temperature H_e than $CoO/Ni_{81}Fe_{19}$, due to the higher T_b of the former.[12] In the superlattice case, however, both the NiO-on-top and the CoO-on-top couples have the same T_b. Since $H_e \propto \sqrt{AK(0)}(1-T/T_b)$, the higher H_e for the NiO-on-top samples is probably a result of a higher $K(0)$ induced in the NiO layer through the coupling with the adjacent high-anisotropy CoO. In fact, the room-temperature H_e of the $\Lambda=18$ Å, NiO-on-top sample is twice that found for field-cooled $NiO/Ni_{81}Fe_{19}$.[12] This means that the room-temperature K is four times larger in the superlattice, *even though T_b is significantly lower*. Assuming cubic anisotropy and using the 373- and 525-K respective T_b's of the superlattice and NiO couples, the resulting $K(0)$ of the superlattice is *16 times* that of NiO. Obviously, some care must be taken in using this interpretation since the NiO-on-top samples do not follow the linear $(1-T/T_b)$ temperature dependence. However, it is obvious that the CoO-NiO interfacial coupling not only affects the order behavior of the constituent layers, but significantly alters the anisotropy of these layers. Another remarkable feature is that H_e is much larger for the short-Λ NiO-on-top superlattice couple than for the $Co_{0.55}Ni_{0.45}O/Ni_{81}Fe_{19}$ couple. Thus the enhanced anisotropy in the NiO top layer is a direct result of coupling chemically distinct layers of high-anisotropy CoO with NiO rather than a direct alloying of the NiO and CoO properties. The third trend is that H_e

increases with decreasing Λ in both the CoO-on-top and NiO-on-top groups. The increase in the room-temperature exchange field from $H_e = 14$ Oe ($\Lambda = 73$ Å, NiO on top) to 43 Oe ($\Lambda = 18$ Å, NiO on top) represents a ninefold increase in the magnetocrystalline anisotropy of the antiferromagnet. Thus the influence of the interlayer coupling on the magnetocrystalline anisotropy depends strongly on the layer thicknesses.

Two superlattice CoO-NiO/$Ni_{81}Fe_{19}$ samples with an overall CoO concentration of 45 vol % were produced to investigate how T_b varies with the [CoO]:[NiO] ratio. These superlattices, [CoO(16 Å)NiO(20 Å)]$_{14}$ and [CoO(8 Å)NiO(10 Å)]$_{28}$, were similar to the $\Lambda = 36$ and 18 Å and NiO-on-top samples discussed above, with the exception that the [CoO]:[NiO] ratio was reversed. These two samples have the same T_b as that expected for $Co_{0.45}Ni_{0.55}O/Ni_{81}Fe_{19}$ exchange couples: $T_b = 420$ K.[12]

Neutron-diffraction studies were performed at the National Institute of Standards and Technology (NIST) research reactor on two triple-axis spectrometers: BT-2 and BT-9. The spins of the monoxides order in ferromagnetically aligned (111) planes which are antiferromagnetically aligned with adjacent (111) planes.[20] Thus the magnetic repeat distance is double the chemical repeat distance. The integrated intensity of the resulting half-order magnetic diffraction peak is related to the square of the average of the Ni^{2+} and Co^{2+} moments weighted by their concentration within each bilayer. The integrated intensity of the magnetic peak was measured as a function of temperature from 10 to 450 K. A CoO(1000 Å) film and two superlattices, [CoO(20 Å)NiO(15 Å)]$_{145}$ and [CoO(15 Å)NiO(20 Å)]$_{150}$, were epitaxially grown on α-Al_2O_3 substrates. The data from the [CoO(20 Å)NiO(15 Å)]$_{145}$, [CoO(15 Å)NiO(20 Å)]$_{150}$, and CoO(1000 Å) samples were fit to Brillouin functions with T_N's of 383, 410, and 305 K, respectively. An example of the data and fit is shown in the inset of Fig. 2 for the [CoO(20 Å)NiO(15 Å)]$_{145}$ sample. The data diverge from the Brillouin function above 380 K. The 305-K T_N of the CoO(1000 Å) sample is significantly greater than the 293-K T_N of bulk CoO,[15] and is $\cong T_b$ of CoO/$Ni_{81}Fe_{19}$ exchange couples.[12] The temperature dependence of the peak intensities are consistent with earlier neutron-diffraction studies on CoO/NiO superlattices.[2] *It should be noted that there is no anomaly in these data near 313 K.* Thus the sublattice magnetization is smoothly varying through the region where H_e vs T shows the slope change in the NiO-on-top samples. The structure in the H_e data must therefore result from the temperature dependence of the magnetocrystalline anisotropy of the superlattices. A more detailed discussion of the neutron-diffraction experiments will be presented elsewhere.[4]

Figure 2 shows a summary of the ordering temperatures of the superlattices and pure CoO and NiO films as measured by both exchange anisotropy (T_b) and neutron diffraction (T_N). Note that the polycrystalline T_b values are similar to the single-crystal T_N values. The ordering temperatures vary essentially linearly with the vol % NiO as observed by Takano, Terashima, and Bando.[2] The exception is the $\Lambda = 73$ Å CoO-on-top exchange couple,

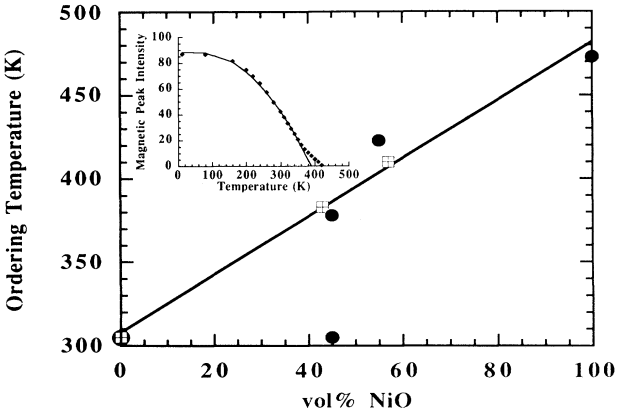

FIG. 2. The ordering temperature of CoO/NiO superlattices as a function of overall NiO concentration as measured by coupling with $Ni_{81}Fe_{19}$ (filled circles) and neutron diffraction (crossed squares). The anomalously low ordering temperature at 45-vol % NiO is the $\Lambda = 73$ Å, CoO-on-top sample. Magnetic peak intensity in arbitrary units vs temperature is plotted in the inset.

where the CoO top layer orders at the CoO value.

In the superlattices, the ordering behavior of both the CoO and NiO layers is strongly influenced, making a simple determination of the range of the interlayer-coupling effect in one constituent layer difficult. In order to define the range of the effect more precisely, exchange couples were produced with a trilayer structure of thick-NiO/thin-CoO/$Ni_{81}Fe_{19}$. The NiO was thick enough to retain "bulk" properties while the CoO layer thickness was varied in the range where strong coupling was observed in the multilayers. Samples with CoO thicknesses

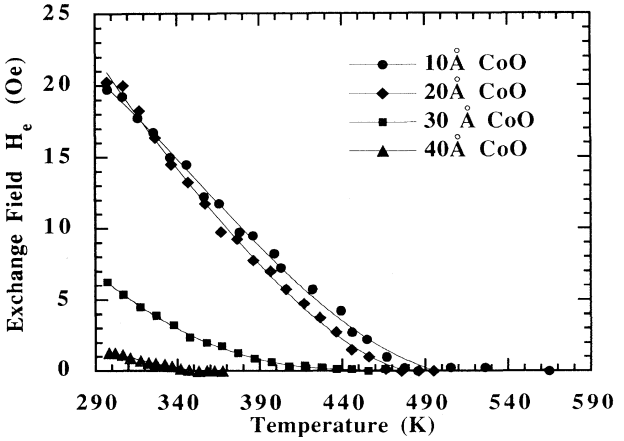

FIG. 3. The exchange field as a function of temperature for thick-NiO/thin-CoO/$Ni_{81}Fe_{19}$ couples. The CoO layer thickness is indicated in the legend. The total oxide thickness is 510 Å. The decrease in blocking temperature T_b with an increase in CoO thickness indicates the range of the magnetic coupling between the NiO and CoO. The 55-Å CoO data are not shown for clarity.

of $t_{CoO}=10$, 20, 30, 40, and 55 Å were produced. The NiO layer thickness was adjusted to keep the overall oxide film thickness 510 Å. H_e vs T for these samples is shown in Fig. 3. T_b is 480 and 470 K for trilayers with CoO layers, t_{CoO}, of 10 and 20 Å, respectively. These values are similar to those exhibited by $NiO/Ni_{81}Fe_{19}$ couples,[12] indicating that the effect of the interfacial coupling is strong enough to insure that the entire layer of low-T_N CoO behaves like the high-T_N NiO to which it is coupled. This T_b is significantly higher than that found for the superlattice $CoO/NiO/Ni_{81}Fe_{19}$ exchange couples discussed above. The reason is that the NiO layer in the trilayer is thick enough to retain "bulk" properties regardless of the interaction with the CoO layer. The CoO and NiO layers in the superlattices were in the same thickness range as the strongly coupled CoO layers in the trilayers. In the superlattices, the CoO and NiO strongly influence each other, with the NiO raising the CoO T_N, and vice versa. Lower T_b's are observed for $t_{CoO}=30$- and 40-Å trilayers (Fig. 3), indicating that the CoO near the interface with the $Ni_{81}Fe_{19}$ is less strongly influenced by the coupling with the NiO. The 308-K T_b of the trilayer with $t_{CoO}=55$ Å indicates that the CoO at the interface with the $Ni_{81}Fe_{19}$ is unaffected by the NiO, since this is the T_b obtained with $CoO/Ni_{81}Fe_{19}$ couples.[12] Thus the interfacial CoO/NiO coupling is effective over a range of <40 Å, and strongly effective over a range of <20 Å.

In conclusion, a strong effect is observed due to the interfacial magnetic coupling between CoO and NiO thin layers. This is exhibited by blocking temperatures in superlattice $CoO/NiO/Ni_{81}Fe_{19}$ exchange couples and Néel temperatures of short-repeat-distance ($\Lambda \leq 36$ Å) CoO/NiO superlattices which depend only on the overall [CoO]:[NiO] ratio. The T_b values of the polycrystalline exchange couples are in good agreement with the T_N values of single-crystal superlattices as determined by neutron diffraction. The magnitude and temperature dependence of the exchange fields for the superlattice $CoO/NiO/Ni_{81}Fe_{19}$ exchange couples depend strongly on the repeat distance Λ and whether CoO or NiO interfaces the $Ni_{81}Fe_{19}$. The data suggest that the oxide-oxide interlayer coupling strongly influences the magnetocrystalline anisotropy of the NiO and CoO layers within the superlattice, significantly increasing the anisotropy of the NiO layers. Exchange-anisotropy measurements yield a direct measure of the range of the interlayer-coupling effect. Data from $NiO/CoO/Ni_{81}Fe_{19}$ trilayers show the effect of the coupling is quite strong for thin CoO layers ($t_{CoO} \leq 20$ Å) but quickly falls to zero with increasing t_{CoO}.

The work at UCSD was supported by the NSF through Grant No. DMR-90-10908 and grants from the IBM Corporation and the Storage Technology Corporation.

[1]C. A. Ramos, D. Lederman, A. R. King, and V. Jaccarino, Phys. Rev. Lett. **65**, 2913 (1990).
[2]M. Takano, T. Terashima, and Y. Bando, Appl. Phys. Lett. **51**, 205 (1987).
[3]D. M. Lind (unpublished).
[4]J. A. Borchers, M. J. Carey, R. W. Erwin, A. E. Berkowitz, and C. F. Majkrzak (unpublished).
[5]A. S. Carriço and R. E. Camley, Phys. Rev. B **45**, 13 117 (1992).
[6]R. W. Wang and D. L. Mills, Phys. Rev. B **46**, 11 681 (1992).
[7]W. H. Meiklejohn and C. P. Bean, Phys. Rev. **102**, 1413 (1956).
[8]W. H. Meiklejohn, J. Appl. Phys. Suppl. **33**, 1328s (1962).
[9]A. P. Malozemoff, Phys. Rev. B **35**, 3679 (1987).
[10]D. Mauri, H. C. Siegmann, P. S. Bagus, and E. Kay, J. Appl. Phys. **62**, 3047 (1987).
[11]C. Tsang and K. Lee, J. Appl. Phys. **53**, 2605 (1982).
[12]M. J. Carey and A. E. Berkowitz, Appl. Phys. Lett. **60**, 3060 (1992).
[13]A. P. Malozemoff, J. Appl. Phys. **63**, 3874 (1988).
[14]A. J. Sievers III and M. Tinkham, Phys. Rev. **129**, 1566 (1967).
[15]T. Nagamiya, K. Yosida, and R. Kubo, Adv. Phys. **4**, 2 (1955).
[16]K. Kurosawa, M. Miura, and S. Saito, J. Phys. C **13**, 1521 (1980).
[17]M. J. Carey, F. E. Spada, A. E. Berkowitz, W. Cao, and G. Thomas, J. Mater. Res. **6**, 2680 (1991).
[18]D. Paccard, C. Schlenker, O. Massenet, R. Montmory, and A. Yelon, Phys. Status Solidi **16**, 301 (1966).
[19]C. Schlenker, S. S. P. Parkin, J. C. Scott, and K. Howard, J. Magn. Magn. Mater. **54-57**, 801 (1986).
[20]G. E. Bacon, *Neutron Diffraction* (Oxford University Press, London, 1975), pp. 444–453, and references therein.