Enhanced Magnetism in Field-Cooled \([\text{Ni}_{80}\text{Fe}_{20}/\text{Mn}]_3\) Multilayers Studied Using Polarized Neutron Reflectometry

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Abstract. Here, the interfacial magnetic coupling in an exchange biased [Ni$_{80}$Fe$_{20}$/Mn]$_3$ multilayer system has been studied using polarized neutron reflectometry. Previous results on this system indicate the importance of the coupling between the Fe-Mn and Ni-Mn orbitals at the layer interfaces. Magnetic depth profiles of the multilayer were measured at low temperatures under field-cooled and zero-field-cooled conditions. While no definitive interfacial state was found, a magnetic moment enhancement of roughly 20-30% in the applied field direction was observed throughout the bulk of the NiFe layers in the field-cooled state as compared to the zero-field-cooled measurements. The origin of this enhancement also likely stems from Fe-Mn and Ni-Mn orbital coupling, but due to the interfacial roughnesses of the sample, the areas where this coupling plays an important role is no longer confined to the interface.

Layered magnetic structures consisting of neighboring ferromagnets (FM) and antiferromagnets (AFM) have played a key role in almost all facets of electronic technology since the discovery of giant magnetoresistance. A hallmark property of these systems is the exchange coupling between the two types of magnetic materials, which can lead to enhanced coercivity and exchange bias, namely as a shift in the hysteresis loop [1]. The use of this behavior to essentially “set” the magnetization direction of a FM layer is one of the main features of the read and write components of magnetic memory devices. The coupling between FM and AFM layers is especially dependent on the interfacial properties of the system, where roughness can be used to tailor exchange coupling [2, 3, 4]. Recent work has also found that spin-orbit coupling can play a role in augmenting exchange bias by enhancing orbital moments in the FM layers near the interface [5].

Here, we report on a Ni$_{80}$Fe$_{20}$ (NiFe)/Mn (FM/AFM) multilayer system studied using polarized neutron reflectometry (PNR). Previous results on similar samples have demonstrated the onset of exchange bias and enhanced coercivity at low temperatures and x-ray magnetic circular dichroism (XMCD) measurements showed evidence of an increased orbital moment under field-cooling conditions for both the Fe and Ni atoms as a result of overlapping d orbitals between Fe-Mn and Ni-Mn [6]. Such interactions could lead to an enhanced magnetic moment at the NiFe/Mn interfaces. The quality of the interfaces within the system also plays a role in determining its properties. Ion-beam bombardment was used to tailor the exchange bias effects in NiFe/Mn bilayers and was demonstrated to alter the Mn spin structure near the interface [7]. When extended to multiple repetitions and compared to composite [NiFe-Mn] layers, the presence of distinct interfaces also dictated the dominant magnetic coupling observed [8].

Due to the importance of the interface in the system presented here, it is worthwhile to correlate the structure of the multilayer and its magnetism. PNR offers a way to obtain a depth-resolved profile of both the nuclear and in-plane magnetic structure of thin films and layered systems and can thus be used to connect structural features such as disorder or interfaces with the magnetic moment at a given depth. Therefore PNR is an apt choice to study potential interfacial magnetic enhancement in NiFe/Mn multilayers.

1. Experiment
A [NiFe (~3.5nm) / Mn (~3.5nm)]$_3$ sample was deposited on an oxidized Si substrate using a dual ion-beam deposition technique, with conditions described in Ref. 6. A 6 nm Al capping layer was deposited on top of the multilayer stack. Due to the relatively low Neél temperature of the AFM Mn layers (~100 K) [6], no magnetic field was applied during deposition. Fig. 1 shows a schematic of the multilayer sample.

PNR was performed on the [NiFe/Mn]$_3$ multilayer in order to probe the magnetic moments in the FM NiFe layers and search for any interfacial effects. PNR uses small angle scattering in a reflection geometry to probe both the magnetic and nuclear depth profiles across a thin film or multilayer system. PNR provides information about the scattering length density (SLD)
in the direction of the scattering vector, $Q_z$, which is perpendicular to the sample surface, and is given as $Q_z = 4\pi \sin \theta \lambda$, where $\lambda$ is the neutron wavelength and $\theta$ the angle of the incident beam. The SLD contains both nuclear (e.g. atomic density and chemical makeup) and magnetic information with $SLD_{\text{total}} = SLD_{\text{nuclear}} \pm SLD_{\text{magnetic}}$ depending on the neutron polarization ($R^+$ or $R^-$). From measuring both $R^+$ and $R^-$ spin channels, where $+$ or $-$ gives the spin state of the incoming neutrons, the difference in SLD yields the magnetic SLD, which can be used to obtain a profile of the in-plane magnetic moment parallel to the neutron spin and applied field across the system. PNR measurements were made at the NREX reflectometer at the FRM-II research reactor in Germany. The program SIMULREFLEC was used to model the data and obtain SLD profiles as well as the magnetic moments of each layer [9].

2. Results

Magnetometry results on this system can be found in Ref. [6] and Ref. [8]. Both an increased coercivity and exchange bias behavior in Magnetization-Field hysteresis loops at low temperatures are clear evidence of exchange coupling [6]. Magnetization vs. Temperature studies show a large discrepancy between the field-cooled and zero-field-cooled magnetic states at low temperatures [6]. Below roughly 50 K, there was a significant increase in the overall magnetization of the multilayer as measured by magnetometry for the field-cooled state.

In order to determine the origins of this behavior, PNR was used to obtain magnetic depth profiles of the system under field-cooled and zero-field-cooled conditions. First, the sample was field-cooled in a 2.5 kOe field to the measurement temperature of 4 K, where a PNR scan was performed. The sample was then heated up to room temperature and re-cooled to 4 K under zero applied field. When this temperature was reached a field of 2.5 kOe was applied and a second PNR profile was obtained. PNR data were collected for the $R^+$ and $R^-$ channels, where the magnetization in the direction of the neutron polarization and applied magnetic field can be measured. The resulting reflectivity curves are shown in Fig. 2 for the zero-field-cooled case (a) and field-cooled case (b). The spin asymmetry between the two spin channels (defined as $\frac{R^+ - R^-}{R^+ + R^-}$), shown in (c), is proportional to the magnetization within a system.

Both the nuclear and magnetic depth SLD profiles were then modeled as to best fit the PNR data and the SLD profiles can be seen in Fig. 3. The magnetic SLD profiles are shown in Fig. 3 (a) for both the zero-field-cooled and field-cooled states. The magnetism within the sample is constrained to within the NiFe layers for both cases, although the interfaces between the NiFe and Mn layers are less sharp than previous measurements of similar samples, with roughnesses.
Figure 2. Reflectometry results for the zero-field-cooled (a) and field-cooled (b) states. The circles show the data and the solid lines are the best fit models. (c) Shows the experimental spin asymmetries \( \frac{R^+ - R^-}{R^+ + R^-} \), which is proportional to the magnetization of the film, for both conditions. The error bars like within the markers.

Table 1. The components of the magnetic moment parallel to the applied field direction for both cooling scenarios.

| Cooling Condition    | Bottom NiFe | Middle NiFe | Top NiFe |
|----------------------|-------------|-------------|----------|
| Zero-field-cooled    | 0.9 \( \mu_B \)/f.u. | 1 \( \mu_B \)/f.u. | 0.8 \( \mu_B \)/f.u. |
| Field-cooled         | 1.1 \( \mu_B \)/f.u. | 1.35 \( \mu_B \)/f.u. | 0.9 \( \mu_B \)/f.u. |

of approximately 10 to 20 Å, as opposed to 5 Å in Ref. 6. This may also result from the increased contrast between NiFe and Mn due to the negative SLD on Mn, as compared to the positive x-ray SLD’s of both materials, which was used to ascertain interfacial roughness values in [6]. A striking increase is noticeable between the magnetic SLD values for the field-cooled case as compared to the zero-field-cooled values. Table 1 shows the calculated moment for each NiFe layer, where the bulk moment is 0.9 - 1 \( \mu_B \)/f.u. [7].

3. Discussion
The best fit SLDs presented in Fig. 3 represent a model where it was not necessary to introduce an interfacial layer that was magnetically distinct from the bulk of the NiFe layers. The addition of an interfacial layer with either an enhanced or reduced magnetic moment did not significantly
Figure 3. SLD profiles for the fits that best fit the zero-field-cooled and field-cooled PNR measurements. The magnetic SLD profiles are shown for both states in (a). The SLD profiles Nuclear scattering and Nuclear ± Magnetic (+ and −) scattering are shown in (b) for the zero-field-cooled case and (c) for the field-cooled case.

improve or worsen the fit to the field-cooled data, so the presence of such a layer cannot be conclusively ruled out. The first scenario, of an enhanced interfacial moment, is what would likely be expected as a result of an orbital overlap across the interface observed in the field-cooled case, as was reported in Ref. 6. The second case, a reduced interfacial moment, may be indicative of a perpendicular anisotropy resulting from spin-orbit coupling at the interface [11], which has been observed in similar NiFe/Ru multilayers [12].

Although the fits are inconclusive with regards to an interfacial layer, it is striking that there is a relatively large enhancement of the NiFe moments throughout the bulk of the layers for the field-cooled measurements. When comparing the NiFe layers to previous data, it is also apparent that we are clearly looking at an enhancement for the field-cooled case rather than a reduction for the zero-field-cooled case. With the exception of the top NiFe layer, the bottom and middle NiFe layers have magnetic moments equal to that of the bulk value in the zero-field-cooled measurements. Additionally, magnetometry measurements on a similar sample shown in Ref. 6 show a coercive field of less than 500 Oe in the field-cooling direction, so it is evident that the PNR measurements are made in a saturated state.

The observation of the enhancement throughout the NiFe layers is likely a result of the increased interfacial roughness in this particular sample. Higher roughness would lead to a
widened interface region with areas of potentially intermixed NiFe and Mn, causing a larger amount of Ni and Fe atoms to neighbor Mn sites. As can be seen from the gradual change of SLD’s from Mn to NiFe in Fig. 3, the interfacial regions penetrate into a large fraction of the NiFe layers, which do not reach their maximum SLD values until almost the center of each layer, so an enhanced orbital moment may be expected throughout nearly the entirety of each NiFe layer rather than in only a layer near the interface. This would also explain why the degree of magnetization enhancement is much less in the top NiFe layer. This layer is only bordered by a single Mn layer and a larger fraction of this NiFe layer is near the bulk NiFe SLD value, indicating a smaller amount of NiFe and Mn mixing.

When comparing this conclusion to the results from Ref. 6, where the magnetization nearly doubled between the zero-field-cooled and field-cooled magnetometry measurements, here a magnetization enhancement of only roughly 20-30 % was seen in the bottom and middle NiFe layers. However, the magnetometry was performed under only a 100 Oe field, as compared to the 2.5 kOe measurement field here, so it is likely that previous results may not have been made in a completely saturated state. The increase in the NiFe moment observed by PNR does agree more with that observed in the XMCD measurements. Using XMCD an orbital enhancement of 15-20% was observed at 20 K. It is likely that here we are either seeing a further increase due to a lower measurement temperature (4 K) or that we are observing the same degree of orbital enhancement [6] with a slight additional increase of the spin moment.

4. Conclusions
Here we have presented a PNR study of a [NiFe/Mn]₃ multilayer system. PNR was used to obtain magnetic depth profiles of the system in both field-cooled and zero-field-cooled states. There was no evidence of an interfacial layer with either an increased or decreased magnetic moment in the field-cooled condition. However, an enhanced moment was observed in the bulk of the NiFe layers in the field-cooled state when compared to the field-cooled layer magnetizations and the bulk magnetic moment. Much as in previously reported results, the origins of this increased moment are likely to be from Fe-Mn and Ni-Mn orbital coupling. The larger interfacial roughness observed here result in more intermixing of the NiFe and Mn atoms, allowing the enhanced state to persist throughout the NiFe layer rather than to remain confined to the interface.

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