Direct observation of the magnetic proximity effect in amorphous exchange-spring magnets by neutron reflectometry

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In this letter we report a direct observation of a magnetic proximity effect in an amorphous thin film exchange-spring magnet by the use of neutron reflectometry. The exchange-spring magnet is a trilayer consisting of two ferromagnetic layers with high $T_c$'s separated by a ferromagnetic layer, which is engineered to have a significantly lower $T_c$ than the embedding layers. This enables us to measure magnetization depth profiles at which the low $T_c$ material is in a ferromagnetic or paramagnetic state, while the embedding layers are ferromagnetic. A clear proximity effect is observed 7 K above the $T_c$ of the embedded layer, with a range extending 50 Å.

The magnetic proximity effect refers to an induced magnetic ordering in a normally non-magnetic material. This can e.g. occur at an interface in direct contact with a ferromagnet or antiferromagnet.[1] Although often overlooked, magnetic proximity effects are present in a variety of magnetic thin film heterostructures and can have profound effects on their magnetic behaviour.[2, 3] As an example, proximity effects in non-magnetic spacer layers separating two ferromagnets can give rise to long-range interlayer exchange coupling,[4] changes in ordering temperature[5] and/or non-oscillatory alignment of magnetic layers.[6, 7] Since layered magnetic structures are ubiquitous in modern technology, the understanding of magnetic proximity effects is of fundamental importance.

In this work we investigate the proximity effect in a ferromagnet-paramagnet system, more specifically in a trilayer[8] of an amorphous exchange-spring magnet.[3] Amorphous heterostructures are favourable when studying magnetic interface effects as they are free of step edges and grain boundaries and consist of well defined and smooth layers.[9] Our exchange-spring magnet consists of three ferromagnetic layers, shown schematically in Fig. 1. The top Co$_{85}$(AlZr)$_{15}$ layer (A) has a high $T_c$ and a small field-imprinted uniaxial anisotropy. The middle layer (B), which is magnetically isotropic Co$_{60}$(AlZr)$_{40}$, is engineered to have a $T_c$ lower than layer A and C. The bottom high-$T_c$ Sm$_8$Co$_{92}$ layer (C) has a large imprinted anisotropy, which can increase the measured coercivity of the adjacent layers. This sample structure has previously been used to indirectly demonstrate that a proximity induced magnetization exists in layer B well above its intrinsic ordering temperature $T_c^B$. The proximity effect results in exchange-spring behaviour at temperatures 50% above $T_c^B$ and exchange bias at even higher temperatures.[3] Here we provide direct evidence of the induced magnetization in the low-$T_c$ middle layer as well as a depth profile of the magnetization throughout the trilayer, using polarized neutron reflectivity measurements.[10]

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The samples were grown by dc magnetron sputtering in a UHV sputtering chamber at an Ar (99.999% purity) sputtering gas pressure of 2.0 mmHg. First, a 20 Å thick buffer layer of Al$_{70}$Zr$_{30}$ was deposited on a Si(100) substrate (with the native oxide) from an Al$_{70}$Zr$_{30}$ alloy target of purity 99%. The buffer layer promotes flat amorphous growth of the following layers. Subsequently, a 200 Å thick Sm$_8$Co$_{92}$ alloy film was grown by co-sputtering from elemental targets of Co (99.9% purity) and Sm (99.9% purity), after which a Co$_{60}$(Al$_{70}$Zr$_{30}$)$_{40}$ of 100 Å and a Co$_{85}$(Al$_{70}$Zr$_{30}$)$_{15}$ layer of 150 Å were grown by co-sputtering from the Co and AlZr targets. Finally, a 30 Å thick capping layer of Al$_{70}$Zr$_{30}$ was grown to protect the magnetic trilayer from oxidation. All films were grown at room temperature. Two permanent magnets provided a magnetic field of approximately 0.1 T parallel to the plane of the films during growth. This induces a uniaxial in-plane anisotropy in the layers which are magnetic at room temperature. The atomic flux as a function of magnetron power was determined for each target material through X-ray reflectivity measurements of films grown from a single magnetron. The power on each magnetron was then set to achieve a given composition while co-sputtering. Rutherford backscattering measurements have previously confirmed that this is a robust method for the materials in question. MOKE measurements were carried out on the samples to confirm the $T_c$ of the middle layer and that the magnetization loops of the trilayers were consistent with previously studied samples. More details on the growth and structural characterization can be found in refs. 3, 11 and 12.

Polarized neutron reflectivity experiments were carried out at the PBR reflectometer at NIST at a wavelength of $\lambda = 4.75$ Å and with an instrument resolution of $\Delta \lambda / \lambda = 0.01$. Four reflectivities, corresponding to the two non-spin-flip channels ($R^{++}$ and $R^{--}$) as well as the two spin-flip channels ($R^{+-}$ and $R^{-+}$), were measured out to $q = 0.2$ Å$^{-1}$ during a 6 hour measurement at $T_1 = 300$ K, $T_2 = 110$ K and $T_3 = 10$ K. With $T_B^B = 103 \pm 1$ K, these temperatures correspond to $T_1 \gg T_B^B$, $T_2 > T_B^B$ and $T_3 < T_B^B$. The uncertainty of $T_B^B$ corresponds to the step size of the magnetization measurements used to determine where the phase transition takes place. Samples were measured with an external applied field of $\mu_0 H = 10$ mT along the easy axis and the scattering plane perpendicular to this axis. Measurements of the spin-flip reflectivities returned mainly noise, consistent with the presence of a collinear magnetization state, which is physically reasonable in the given measurement configuration. The spin-flip reflectivities were therefore subsequently disregarded in the fitting process.

Data were fitted using the GenX 2.4.7 reflectivity package with the new MagRefl module 13, using the logarithm of the reflectivity as the figure of merit. The sample model included an oxide consisting of a 70/30 mixture of Al$_2$O$_3$/ZrO$_2$, a capping layer of Al$_{70}$Zr$_{30}$, a magnetic Co$_{85}$(AlZr)$_{15}$ layer, a magnetic Co$_{60}$(AlZr)$_{40}$ interlayer, a magnetic Sm$_8$Co$_{92}$ layer and a buffer layer of Al$_{70}$Zr$_{30}$ on a thin SiO$_2$ layer on a Si substrate. Most structural parameters were determined by fitting the results obtained at $T = 300$ K and $\mu_0 H = 10$ mT. Reflectivity measurements cannot alone determine the chemical composition of layers consisting of more than two elements, but the careful validation of the sample preparation procedure stated above justifies fixing the stoichiometry of the layers to the intended values and only allowing their densities to vary during the fitting at $T = 300$ K and $\mu_0 H = 10$ mT.
mT. In the simulations, the low-$T_c$ layer was sliced into 10 slices and their magnetic moments were fitted to a sum of two power laws with the same exponent, one power law corresponding to a decaying magnetization from each interface, induced by the neighbouring ferromagnetic layers as illustrated in Fig. 1. A power law decay of the magnetization was used as it is the functional form of the long-range exchange interaction, as described in refs. [2] and [14]. The resulting step-wise magnetic profile was furthermore smoothed out by allowing a small, linked chemical roughness for each slice. As the chemical scattering length density (SLD) of each slice is also linked, the resulting chemical SLD profile of the low-$T_c$ layer acts like an uniform layer, as opposed to the non-trivial magnetic SLD profile. Only the magnetization of the the three ferromagnetic layers and their thicknesses were allowed to vary when fitting the data obtained at $T = 300$ K and $T = 10$ K, or stated differently, the chemical SLD and most structural parameters such as densities are fixed from the fit done at $T = 300$ K and $\mu_0 H = 10$ mT, while changes in the magnetic SLD is allowed. Additionally, changes in thickness were needed to account for the thermal expansion in the layers. The thermal expansion was determined to be 1.2 % when heating the sample from $T = 10$ K to $T = 300$ K which corresponds to $4.1 \times 10^{-5}$ K$^{-1}$. Notice that the determined thermal expansion only holds for the combined film and substrate, where the substrate provides elastic boundaries defining the changes in the lateral direction with temperature. The resulting polarized neutron reflectivity results (scaled in $q$) and fits are shown in Fig. 2(a), (b) and (c).

The determined chemical and magnetic SLD profiles at $\mu_0 H = 10$ mT and $T = 300$ K are shown in Fig. 3(a) as a function of the distance from the substrate. The chemical SLD profile yields interface roughnesses equal to or less than 8 Å. Magnetic SLD profiles of the middle layer region are shown in Fig. 3(b) at all investigated temperatures and applied fields. It is clearly seen that at the highest temperature $T = 300$ K and in the small field of $\mu_0 H = 10$ mT (green line) there is no significant magnetization in the middle of the layer. Furthermore, the magnetization at the interfaces decays sharply, consistent with weak magnetic proximity effects at this temperature. At the lowest temperature, $T = 10$ K (black line), the middle layer is magnetized, as expected since this is well below the ordering temperature of layer B. Interestingly, a significant magnetization is seen throughout the middle layer at $T = 110$ K (red line) which is 7 K above $T_c^{B}$. This is clear evidence for a long range magnetic proximity effect, where a magnetization is induced in layer B well above its intrinsic ordering temperature due to the proximity to the high-$T_c$ layers A and C. As there is a significant magnetization in layer B, which is 100 Å thick, it can be deduced that the range of the proximity effect is at least 50 Å at $T = 110$ K. This is consistent with previous experimental results and requires an explanation in terms of long-range exchange interactions. [3]. Monte Carlo simulations have shown that in this temperature range a large region of induced magnetization can be expected in layer B if beyond nearest neighbour magnetic interactions are accounted for. Atomic correlations in terms of regions of higher Co-density can further amplify the proximity effect, as discussed in ref. [2].

To investigate the temperature dependence of the magnetic susceptibility of the system, the absolute value of the derivative of the magnetic SLD profiles in Fig. 3(b) was calculated. Four half-widths at half maxima were extracted, describing the change in magnetization at the

![FIG. 3. (a) Chemical and magnetic SLD profiles at $\mu_0 H = 10$ mT and $T = 300$ K for the entire exchange-spring magnet heterostructure. (b) Magnetization profiles of the low-$T_c$ Co$_{60}$ (AlZr)$_{40}$ middle layer and its interfaces at $\mu_0 H = 10$ mT, $T = 10$, 110 and 300 K.](image)

### TABLE I. Half-width half maxima of the derivative of the magnetic SLD profiles.

| Temperature (K) | I (Å) | II (Å) | III (Å) | IV (Å) |
|-----------------|-------|--------|---------|--------|
| 10±1            | 7.8   | 7.7    | 8.6     | 8.3    |
| 110±1           | 8.0   | 11     | 11      | 8.7    |
| 300±1           | 7.5   | 7.2    | 8.3     | 7.9    |
two interfaces of layer B. The results are provided in Table I, the annotations of the interfaces I-IV are defined in Fig. 3 (b). The widths I and IV describe the change in the magnetization in layers A and C and these are found to be independent of temperature. This is expected as the changes in the temperature are small as compared to the intrinsic ordering temperature of the layers. The widths II and III describe the changes in the magnetization profile of layer B. At \( T = 10 \text{ K} \), the layer is fully magnetic and the deduced magnetization profile reflects the distribution of the elements in the sample. In this case, only minute proximity effects are expected, as the magnetic susceptibility of all the layers is small. Same is valid for the results obtained at \( T = 300 \text{ K} \), as this temperature is well above \( T_c \) of layer B, while far below \( T_c \) of layers A and C. Under these conditions, finite size effects can become prominent, giving rise to a decay of the magnetization in the interface region 16. These effects appear to be too small to be observed in the current experiment. We therefore conclude that the changes in the magnetization profile of layers A and C must be small. The changes in layer B are more pronounced, as seen in Fig. 3 (b) and Table I A clear maximum in width is seen at \( T = 110 \text{ K} \), which is close to the ordering temperature of layer B (103±1 K). This observation reflects the expected changes in the magnetic susceptibility, which is largest at \( T_c \). Monte Carlo simulations involving beyond nearest neighbour magnetic interactions show that not only is the peak in the magnetic susceptibility shifted to higher temperature, but it is also significantly broadened, due to proximity effects. 3 This is well reflected in the present results.

To explore the consistency of these results we compare the neutron reflectivity results with magnetization measurements of two separate films, one with the same composition as layer B and another one with similar composition to layer C. Two samples consisting of single layers of Sm\(_{10}\)Co\(_{90}\) and Co\(_{60}\)(AlZr)\(_{40}\) were measured in both a remanent state and in the presence of a field (\( \mu_0 H = 100 \text{ mT} \)) using the magneto-optical Kerr effect (MOKE). This field value is well above the saturation field of layer B and was chosen to ensure a collinear state in all layers. Both saturation and remanent magnetization values were extracted from full hysteresis loops at each temperature. The magnetic response normalized to their values at \( T = 10 \text{ K} \) are plotted for both the MOKE and neutron reflectometry measurements in Fig. 4. For the Sm\(_{10}\)Co\(_{90}\) sample, the moments measured in the remanent state and at \( \mu_0 H = 100 \text{ mT} \) by MOKE are essentially identical (only the latter is therefore shown in Fig. 4) and are consistent with the Sm\(_{8}\)Co\(_{92}\) layer magnetization measured by neutron reflectometry. MOKE measurements also clearly show the ferromagnetic-paramagnetic phase transition of Co\(_{60}\)(AlZr)\(_{40}\) and how the single-layer is magnetized far above its \( T_c \) in the presence of an external field of \( \mu_0 H = 100 \text{ mT} \). The Co\(_{60}\)(AlZr)\(_{40}\) layer is expected to be fully saturated by the field from the Sm\(_{8}\)Co\(_{92}\) layer in the exchange-spring magnet trilayer and the magnetization measurements of this layer by neutron reflectometry correspond very well to the MOKE values. The amplitude of the proximity effect is expected to scale with the magnetic susceptibility of the paramagnetic layer. We therefore conclude that the proximity effect at a ferromagnetic-paramagnetic interface is comparable with the presence of an external field, while the magnetic profile will be different in these two cases.

In conclusion, the results prove the existence of an induced magnetization throughout the middle layer well above its intrinsic ordering temperature caused by a magnetic proximity effect. Earlier measurements of the magnetic moments of single layer samples by MOKE magnetometry are consistent with the corresponding trilayer moments measured by neutron reflectometry, reaffirming the analysis. The measured effect has a range of at least 50 Å at \( T = T_c + 7 \text{ K} \), which can only be explained by long-range interactions.

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