Magnetic Proximity Effects in Antiferromagnet/Ferromagnet Bilayers: The Impact on the Néel Temperature

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We present a study of the ordering temperature of an ultrathin antiferromagnetic film in the proximity of a ferromagnetic layer. The Néel temperature of a single-crystalline antiferromagnetic Fe_xMn_{1-x} film on Cu(001) in contact with a ferromagnetic Ni layer was monitored by the discontinuity in the coercivity as a function of temperature by magneto-optical Kerr effect measurements. It decreases by up to 60 K if the magnetization axis of the ferromagnet is switched from out of plane to in plane by deposition of a Co overlayer. These results give clear evidence for a magnetic proximity effect in which the ferromagnetic layer substantially influences the ordering temperature of the antiferromagnetic layer.

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Antiferromagnetic layers are essential components of magneto-electronic devices [1] because of their insensitivity to external magnetic fields. The combination of ultrathin ferromagnetic (FM) and antiferromagnetic (AFM) layers opens fascinating possibilities for the engineering of magnetic metamaterials with desired magnetic properties [2]. In an antiferromagnet, the atomic spins order such that the overall magnetization cancels when averaged over a few lattice sites. Placed next to a ferromagnet, the spins at the interface couple to those of the ferromagnet by direct exchange interaction. A prominent consequence is a pronounced increase in the coercivity of the FM layer and a shift of the magnetization loop along the field axis, termed the exchange bias effect [3]. Because of its relevance for a number of applications, the fundamental investigation of the detailed physical mechanisms underlying the exchange bias effect as well as studies of fundamental properties of ultrathin antiferromagnetic films remain topics of very high current interest [4–6].

It is important for the understanding of the thermodynamic behavior of ultrathin AFM films as well as for device design to study the magnetic properties at *finite* temperatures. A particularly important parameter is the antiferromagnetic ordering temperature, the Néel temperature, above which the long-range antiferromagnetic spin order disappears. The ordering temperature, as in ferromagnets, depends on the strength of the exchange interaction between neighboring spins and the dimensionality of the system. In ultrathin FM films, it decreases with decreasing film thickness [7]. This behavior is well established and is called the finite-size effect. In contrast, the ordering temperature of ultrathin AFM films is extremely difficult to measure, since in such thin films susceptibility or neutron scattering data cannot be acquired with sufficient signal intensity due to the lack of material. The ordering temperature of these films is therefore inferred indirectly by observing the influence of the AFM layer on the magnetic properties of a neighboring FM layer. Similar finite-size effects have been observed in that way in ultrathin FeMn films [8], lowering the Néel temperature when the thickness of the AFM layer is decreased. Any influence that the FM layer may have on the ordering transition of the AFM layer, however, is always included in these indirect measurements. Such magnetic *proximity effects* can occur due to the interaction between two magnetic layers with different ordering temperatures. The layer with the higher ordering temperature induces magnetic order into the layer with the lower ordering temperature at temperatures at which the latter would be paramagnetic. Examples are indirectly exchange-coupled FM layers. There it was experimentally and theoretically observed that the coupling between two FM layers across a nonmagnetic spacer layer does indeed induce magnetic order in the layer with the lower ordering temperature at temperatures above its individual ordering temperature [9,10].

Proximity effects in AFM/FM bilayers are much less investigated. While it has been observed that the contact with an AFM layer can change the ordering temperature of an FM layer [11], the opposite, i.e., the influence of the FM layer on the Néel temperature of the AFM layer, has only been reported theoretically [12]. In this Letter, we address this matter from the experimental side.

We present magnetization measurements of AFM/FM bilayers in which the magnetization direction of the FM layer was spin engineered to exhibit either in-plane or out-of-plane anisotropy for otherwise identical interface conditions. By comparing the antiferromagnetic ordering temperature of single-crystalline AFM FeMn films in contact to an FM layer with in-plane or out-of-plane magnetization, we show unequivocally that the FM magnetization significantly influences the ordering transition of the AFM layer. We adjust the magnetic easy axis of the FM layer by depositing an overlayer, thus keeping the AFM-FM interface chemically and morphologically identical. Furthermore, variations in thickness and composition of the AFM FeMn film, as may occur in separate depositions,

are avoided. We observe an astonishing difference in the apparent Néel temperature of the AFM layer, which can thus unequivocally be assigned to a magnetic proximity effect. It can be explained by different AFM-FM coupling strengths for different FM magnetization directions, leading to a different proximity effect and thus to a different AFM ordering temperature. Additionally, we also find a dependence of the coupling on the interface topography, indicating a different sensitivity of the AFM-FM coupling on topographic details for in-plane and out-of-plane magnetization.

Single-crystalline epitaxial AFM FeMn films on Cu(001) are ideal systems to study the interaction between AFM and FM materials. They grow in a layer-by-layer mode [8], while the surface roughness is limited to islands and vacancy islands of monoatomic height [13]. Further deposition with a ferromagnetic material continues the layer filling, following the initial layer-by-layer growth [14]. This provides the unique opportunity to controllably vary the interface topography on the atomic scale by simply varying the atomic layer filling [5]. A three-dimensional noncollinear spin structure, similar to that of bulk FeMn, has also been found in ultrathin FeMn films [4]. Consequently, these films can couple to both in-plane as well as out-of-plane magnetized FM films [4,15].

FeMn, Co, and Ni were deposited at room temperature by thermal evaporation on a 5 mm diameter disk-shaped Cu(001) crystal. Film thicknesses and composition were determined by medium-energy electron diffraction during growth and by Auger electron spectroscopy, as outlined in Ref. [8]. The accuracy of the film thickness determination is about 5%, of the Fe_xMn_{1-x} composition about five atomic percentage points. The uniformity of these values across the sample was better than 2%. After growth of an Fe_xMn_{1-x} layer by coevaporation of Fe and Mn, 15 atomic monolayers (ML) of Ni were deposited on top. Five ML of Co were finally evaporated on one half of the sample with the other half covered by a mechanical shutter. It is known that the 5 ML Co overlayer leads to an in-plane magnetization of the Co/Ni bilayer [16], whereas 15 ML Ni/Cu(001) exhibit an out-of-plane easy axis of magnetization [17]. This has been verified using the magneto-optical Kerr effect (MOKE).

Figure 1 shows MOKE hysteresis loops taken at different temperatures in the range of $T = 226$ K to 336 K for a 15 ML Ni/7 ML $Fe_{47}Mn_{53}$/Cu(001) sample with and without a 5 ML Co overlayer. Polar MOKE geometry was used to record the hysteresis of the out-of-plane magnetized uncovered half of the sample [Fig. 1(a)], whereas the cobalt-covered other half was measured in longitudinal MOKE geometry [Fig. 1(b)] with the external magnetic field applied along the [110] axis. Square hysteresis loops are obtained along the easy axes of magnetization, namely, out of plane (in plane) for the uncovered (covered) half of the sample. The reason that the hysteresis loops of the cobalt-covered part are not entirely square is a slight misalignment of the sample azimuth and therefore some contribution from the [100] intermediate axis. Nevertheless, the key issue is the temperature dependent change of the coercivity H_c. In FM samples a monotonic decrease of H_c is expected as T is increased towards the magnetic ordering temperature. For the AFM/FM bilayers a clear discontinuity in the slope of H_c versus T indicates the ordering transition in the AFM layer.

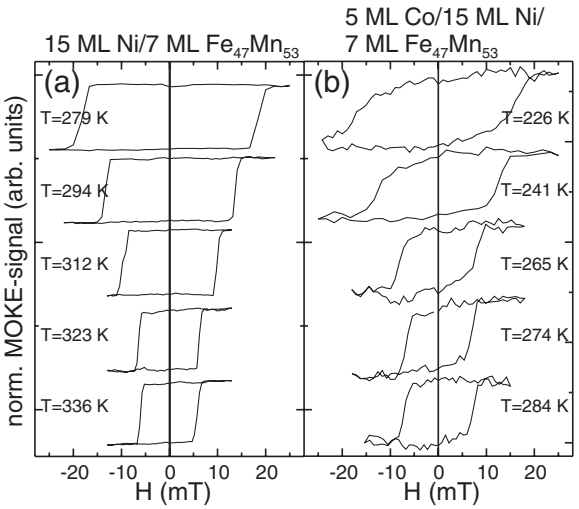

FIG. 1. Magnetic hysteresis loops of (a) 15 ML Ni/7 ML $Fe_{47}Mn_{53}$/Cu(001) and (b) 5 ML Co/15 ML Ni/7 ML $Fe_{47}Mn_{53}$/Cu(001) at different temperatures.

Figure 2 shows the coercivity of four different (Co)/Ni/Fe_xMn_{1-x} samples with $x = 39\%$, $x = 47\%$, $x = 50\%$, and $x = 64\%$ and thicknesses ranging from 6.9 to 8.1 ML as a function of temperature. Above a temperature T_{AFM}, H_c is similar to that of the FM layers, leading to a weak temperature dependence. Below this temperature, the AFM couples with the FM layers and therefore H_c is strongly increased. As described in Ref. [8], the intercept of tangents to the two slopes can thus be regarded as the ordering temperature T_{AFM}. This has been used previously to determine the ordering temperatures in FeMn/Co bilayers on Cu(001) [8]. The arrows in Fig. 2 mark the values of T_{AFM} that were obtained in this way. Above these temperatures, the coercive field for all samples, out of plane [Fig. 2(a)] as well as in plane [Fig. 2(b)], is similar and amounts to about 5 mT. This indicates that the coercivity is not so much determined by the magnetic anisotropy but by domain nucleation and pinning of domain wall motion, which depends on structural details and is similar for the in-plane and out-of-plane magnetized films.

In Fig. 2(a) and 2(b) one observes that for a constant thickness a decrease of the Mn content x of the AFM layer by about 15 percentage points is accompanied by a reduction of T_{AFM} by 20–30 K; e.g., compare the 7.0 ML data set (squares) with 6.9 ML data (solid circles). Keeping x fixed at about 50%, but decreasing the thickness by 1 ML (e.g., compare $Fe_{50}Mn_{50}$ with $Fe_{47}Mn_{53}$) leads to a similar

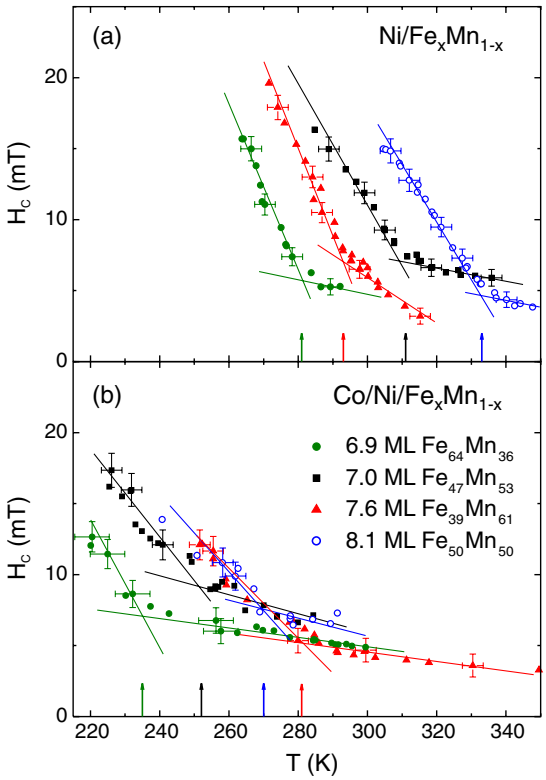

FIG. 2 (color online). Temperature dependent coercivity of the 15 ML Ni/Fe$_x$Mn$_{1-x}$ films (a) without and (b) with a 5 ML Co cap layer. Arrows denote T_{AFM} as evaluated from the discontinuity of the slopes of the $H_c(T)$ curves.

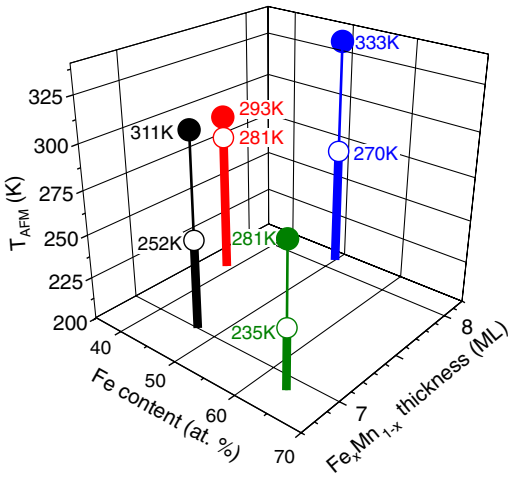

FIG. 3 (color online). Summary of Fig. 2 with the T_{AFM} values for the uncovered (●) and the cobalt-covered part (○) of the samples as a function of thickness and composition.

reduction of T_{AFM}. This applies to the Co-covered sample as well. The most interesting point is that when comparing Fig. 2(a) with Fig. 2(b), i.e., the out-of-plane versus the in-plane magnetized half of the sample, the temperatures at which the increase in the coercivity occurs are distinctly higher in the case of out-of-plane magnetization. This implies that a change in the direction of the magnetization axis of the FM layer obviously affects the Néel temperature of the AFM layer. Since the AFM-FM interface is *identical* in the Co-covered and the uncovered parts of the sample, the shift in T_{AFM} must be solely the result of magnetic proximity effects between FM and AFM.

Figure 3 summarizes the data of Fig. 2 in a three-dimensional plot. The open circles denote T_{AFM} for the Co-covered (in-plane magnetized) part of the sample and the solid circles T_{AFM} for the uncovered (out-of-plane) part, respectively. The values for in-plane magnetization compare qualitatively and quantitatively well with the behavior established by Offi *et al.* for in-plane magnetized FeMn/Co bilayers [8]: (i) increasing the Fe content x of the AFM layer at constant thickness reduces T_{AFM} and (ii) finite-size effects decrease T_{AFM} with decreasing thickness of the AFM layer at constant composition. The antiferromagnetic ordering temperatures of Fe$_x$Mn$_{1-x}$ layers as a function of composition x and Fe$_x$Mn$_{1-x}$ film thickness in Co/Ni/Fe$_x$Mn$_{1-x}$ (here) and Co/Fe$_x$Mn$_{1-x}$ (Ref. [8]) are thus very similar. This points towards a minor influence of the interface chemical composition as long as the topography and magnetization direction of the FM layer are the same. Since the surface of the Fe$_x$Mn$_{1-x}$ layer is mainly determining the topography of the interface to the FM overlayer, it is reasonable to assume a similar interface topography in Co/Ni/Fe$_x$Mn$_{1-x}$ and Co/Fe$_x$Mn$_{1-x}$.

In contrast, the Néel temperature of the AFM is increased by 46–62 K at close to integer atomic layer thicknesses of the AFM layer ($\approx$7.0 and $\approx$8.0 ML) if the magnetization direction is changed from in plane to out of plane, maintaining otherwise identical interface properties. We interpret the variation of the antiferromagnetic ordering temperature in AFM-FM systems as a proximity effect of the FM layer on the AFM layer. The magnetic coupling between FM and AFM acts like an effective field on the AFM sublattices, increasing the ordering temperature, in analogy to the case of two indirectly exchange-coupled FM layers [9,10]. The increase of the ordering temperature can thus be interpreted as a measure for the AFM-FM coupling strength.

Now we have to discuss why the Néel temperatures of the Fe$_x$Mn$_{1-x}$ films for in-plane magnetization of the adjacent FM layer are lower than for out-of-plane magnetization. In terms of AFM-FM coupling this would mean that the coupling is stronger for out-of-plane magnetization than for in-plane magnetization. Such differences in the AFM-FM interaction on the magnetization axis have indeed been reported. For CoO in contact to Co/Pt multilayers, Maat *et al.* found a higher exchange bias field for in-plane magnetization compared to out-of-plane magnetization [18]. The anisotropy in the CoO layer, induced by the textured growth of the sputter-deposited samples, had been evoked as explanation, and the different exchange bias

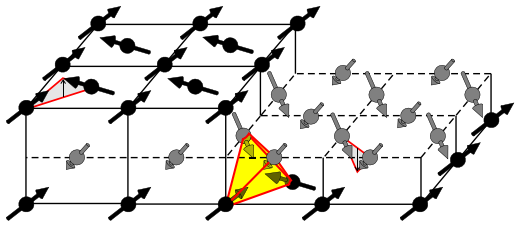

FIG. 4 (color online). Sketch of the $3Q$ noncollinear antiferromagnetic spin structure. Monoatomic steps at the (001) surface exhibit 90° different axes of the compensated spin component in the film plane and alternating sign of the uncompensated spin component perpendicular to the film plane.

field had been attributed to the projection of the AFM spins on the FM magnetization axis [18]. Marrows reported a similar behavior for a sputtered FeMn layer in contact to a Co/Pd multilayer [19], but pointed out that not only the AFM spin structure, but also details of the interfacial spin structure including defects and disorder have to be considered to discuss the smaller exchange bias field in the perpendicular direction.

Such details are more easily accessed if single-crystalline layers are involved, as in our study. Assuming a $3Q$-like spin structure [20] for the epitaxial Fe_xMn_{1-x} layers gives a consistent explanation of our results. Figure 4 shows a schematic illustration of this spin structure. The in-plane component of the surface atoms' spins in extended flat terraces is compensated. The situation is different for out-of-plane magnetization. As seen from Fig. 4, the out-of-plane spin component in flat terraces is not compensated. All the surface spins in the upper terrace of Fig. 4 are pointing up; in the lower terrace they are pointing down. This uncompensated spin structure suggests that for out-of-plane magnetization of the FM layer large islands and flat terraces would contribute significantly to the AFM-FM coupling by direct exchange interaction, leading to a larger coupling and thus a higher T_{AFM} compared to the in-plane case.

This model could be discussed further: if the coupling is mediated by the *out-of-plane* component from flat regions of the interface, at half-integer fillings of the bottom Fe_xMn_{1-x} layer the coupling of terraces of different height should be largely compensating each other. In addition, we have shown previously that the coupling between FeMn and *in-plane-magnetized* Co is taking place dominantly at monatomic steps of islands and vacancy islands at the interface, whereas atomically flat terraces do not significantly contribute [5]. A $3Q$-like spin structure would thus also explain the difference of ordering temperatures of the 7.6 ML $Fe_{39}Mn_{61}$ sample (Fig. 3). While for the other samples the difference between the ordering temperatures for in-plane and out-of-plane magnetization amounts to 46–63 K, it is only 12 K for this sample. Assuming similar finite-size effects and similar influence of the composition on the ordering temperature for both magnetization directions, this reduced difference would be in line with a significantly weaker AFM-FM coupling for out-of-plane magnetization at half-integer atomic layer filling of the interface compared to close to integer layer filling.

We have experimentally evidenced that an FM layer in direct contact with an AFM layer strongly influences the ordering temperature of the latter. This proximity effect depends on the strength of the coupling to the adjacent FM layer. It has important consequences for the interpretation of ordering temperatures of ultrathin AFM layers in general when they are in contact to FM layers, which is usually the case in all applications. The real Néel temperature of an isolated AFM layer, which for FeMn/Cu(001) is still unknown and may be even lower than what we found for the in-plane magnetized bilayers, may not be the relevant quantity for the design of functional structures containing AFM layers. In the case of (Co)/Ni/Fe_xMn_{1-x}/Cu(001) there exists a temperature interval in which the Fe_xMn_{1-x} layer is *ordered* if the FM layer is out-of-plane magnetized, but *disordered* if the magnetization is in the film plane. This suggests that the antiferromagnetic spin order of an AFM layer may be switched on and off simply be changing the magnetization axis of the adjacent FM layer at a temperature that can be tuned by thickness or composition.

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