90° coupling in (Fe/Cr/Fe)_{AFM}/Cr/Fe system epitaxially grown on GaAs(001)

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Abstract

Single-crystalline (Fe/Cr/Fe)_{AFM}/Cr/Fe structures were epitaxially grown on atomically flat GaAs(001). Choosing the same thickness of the antiferromagnetically (AFM) coupled Fe layers in the bottom (Fe/Cr/Fe)_{AFM} structure, their net magnetization is balanced to zero, in particular up to a spin–flop transition when the field is applied along the [1 1 0] direction. For the Cr thicknesses at which the top Fe layer is weakly magnetically coupled to the bottom (Fe/Cr/Fe)_{AFM} structure, at low fields, the magneto-optical Kerr effect and/or SQUID signal from the sample corresponds to the top Fe layer only. An influence of the Cr spin structure on the magnetization reversal in the Fe layer is reported. In particular, a strong increase of coercivity (by a factor of 12) is found at low temperatures. A 90° coupling is detected which affects the minor loops measured along the [−1 1 0] and [1 0 0] directions.

Keywords: Ferromagnetic layers; Epitaxial growth; GaAs(001); Magneto-optical Kerr effect; Interlayer coupling

1. Introduction

The coupling between ferromagnetic (FM) layers separated by a non-FM layer has become a subject of great interest during the past decades. This is due to fundamental interest in how such a coupling is mediated across a metallic \[1\] or non-metallic \[2\] spacer as well as due to technical applications of such coupled systems (e.g. \[3\]).

It is well known that the coupling can result in a non-collinear alignment of the magnetization directions of the two FM films that is phenomenologically explained with a biquadratic coupling term contributing to the total energy of the system. Perpendicular interlayer coupling is reported for a variety of different systems including the most explored Fe/Cr/Fe trilayers \[4\]. Very recent experiments have found a 90° interlayer coupling in FM/antiferromagnet (AFM)/FM systems with Mn \[5\] and NiO \[6,7\] as spacer layers. In particular, the first direct experimental observation of a room temperature (RT) in-plane 90° coupling in Co/NiO/FeNi trilayers was made by means of

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magneto-optical Kerr effect (MOKE) and X-ray magnetic circular dichroism [8]. A coexistence of exchange biasing and FM coupling in two FM layers separated by an NiO layer has also been reported [9].

To understand the coupling between two FM layers across the AFM spacer, the spin structure of the AFM must be considered. This is closely related to the exchange-bias phenomenon between the FM and AFM layers reported frequently [10,11] and analyzed in view of the spin structure in the AFM layer (e.g., [12]). Most of the exchange-biased systems incorporate AFM layers with localized moments (like Mn and NiO), whereas Cr is an itinerant AFM with an incommensurate spin density wave [13]. Nevertheless, in FeNi films deposited on the Cr(001) substrate exchange bias has also been observed and oscillatory behavior has been found [14].

In most studies of the interlayer magnetic coupling, identical FM layers were used which complicated the separation of their contributions to the total magnetization curve. The magnetic signal of two FM layers of different materials was clearly separated due to a difference in the intrinsic magnetic anisotropy [6] or using element-sensitive hysteresis loops taken by X-MCD [8].

It is important to understand details of the magnetization reversal in the FM layer grown on the AFM film of all magnetic moments uniaxially aligned. The most important question concerns the influence of the real surface (and real spin arrangement) of such an AFM film on the magnetization of the FM overlayer. The most standard (Fe/Cr/Fe)\textsubscript{AFM}/Cr/Fe system epitaxially grown on GaAs(001) is chosen for the experiments reported in this paper. With this system, the uniaxial spin alignment in the Cr film is achieved due to uniaxial anisotropy persisting in the underlying (Fe/Cr/Fe)\textsubscript{AFM} structure if grown on GaAs(001). In addition, the (Fe/Cr/Fe)\textsubscript{AFM}/Cr/Fe system allows to follow the magnetization reversal separately for one FM layer only in the case where both magnetically coupled FM layers are made of the same material. In particular, the analysis is applied for the Cr spacer thickness for which the top Fe layer is not, or only weakly, FM coupled to the bottom (Fe/Cr/Fe)\textsubscript{AFM} structure. This allows a detailed analysis of the coupling between the Fe layer and spin structure of the Cr film separating the top Fe layer from the (Fe/Cr/Fe)\textsubscript{AFM} structure.

2. Experimental

The sample preparation and characterization were carried out in a ultrahigh vacuum multichamber system equipped with molecular beam epitaxy, Auger electron spectroscopy (AES), low-energy electron diffraction (LEED), scanning tunneling microscopy and in situ MOKE. MOKE loops were collected in the longitudinal geometry by using an electromagnet (with a maximum field of 30 mT).

The GaAs substrates were cleaned by 500 eV Ar\textsuperscript{+} sputtering at 590°C. After the cleaning procedures were completed, no traces of contamination were detected in the AES spectra and sharp LEED patterns were observed. The cleaning procedure resulted in a Ga-terminated (4\times6)-like reconstruction, which was found to protect the Fe film against strong intermixing with As and Ga [15].

Fe and Cr were deposited at a rate of 1–1.5 ML/min by electron beam evaporation from thoroughly outgassed high-purity iron and chromium rods. The growth was carried out at varying temperatures at a pressure below 4 \times 10^{-10} mbar (which was the maximum pressure after a long deposition of Fe). Finally, the samples were coated with a 30 ML Au layer to prevent reactions with air.

Magnetization measurements were made ex situ using a Quantum Design SQUID magnetometer. Since the magnetic field was controlled by the current, all low-field loops for \(H<5\) mT were measured independent of high-field loops for \(H>250\) mT to avoid the creep of the remanent field in the superconducting magnet. The remanent field, which was of the order of 0.5 mT, was accounted for by the independent measurement of the magnetic field before and after each low-field loop.
3. Results and discussion

Progress in the preparation of the GaAs(001) substrate allows a Ga-terminated atomically flat surface of large terraces, separated by double-atomic steps [15]. An ultrathin Fe film can be grown directly on such a surface at RT, with no significant intermixing between GaAs and Fe. The plot of longitudinal Kerr signal in remanence vs. the Fe film thickness shows a linear dependence [15,16]. The line crosses the thickness axis very close to zero thickness confirming that magnetically dead layers are not present in the GaAs(001)/Fe interface [16].

We prepared the “artificial layered AFM” growing Fe (15ML)/Cr (9ML)/Fe (15ML) trilayer directly on the GaAs(001) substrate. The Cr thickness of 9ML corresponds to AFM coupling between the Fe layers (for the systems in which the short period oscillations cannot be obtained due to structural imperfections, a strong AFM coupling is typically observed for the Cr films thickness between 5 and 10–11 ML, e.g. [17]). Such a “layered AFM” shows an easy axis of magnetization oriented along the [110] direction. This is due to the uniaxial anisotropy of the easy axis along the [110] direction that dominates over the 4-fold anisotropy existing in the bcc Fe(001) films grown on GaAs(001) [18]. This uniaxial anisotropy allows omitting the usually applied external magnetic field during growth below the Neel temperature [10]. Then, a 5 ML thick Cr film was grown on top of this Fe/Cr/Fe “layered AFM”. Finally, the structure was completed with the top Fe layer 13 ML thick. The top Fe layer can be coupled to the bottom (Fe/Cr/Fe)\textsuperscript{AFM} structure in an oscillating manner (between AFM and FM) with varying thickness of the separating Cr layer. The strength of the interlayer coupling is weak at the Cr thicknesses at which the coupling changes sign. Also for very thin Cr layer (below 3–6 ML, depending on flatness of the Fe surface), the Fe layers are either FM coupled or simply uncoupled. Then, the hysteresis loop, which is measured for the complete sample, corresponds to the top Fe layer only, because the signals from the Fe layers of the bottom Fe/Cr/Fe “layered AFM” are balanced to zero. The magnetization reversal followed by SQUID measurements is shown in Fig. 1, when the external magnetic field was applied along the [110] direction. After the top Fe film is saturated (at a field of about 5mT), the signal remains constant until the field approaches the value at which the spin–flop transition occurs in the bottom (Fe/Cr/Fe)\textsuperscript{AFM} trilayer. This clearly confirms that the magnetization in both the Fe layers of the (Fe/Cr/Fe)\textsuperscript{AFM} structure is fully saturated, however in opposite directions. The details of the magnetization reversal, with increasing field only (after saturation at \(-H_{\text{max}}\), are shown in Fig. 2 (full dots). At about 60 mT, a very sharp transition is seen, followed by a continuous saturation at about 150 mT. Interpretation of this transition comes from a comparison of the absolute magnetization values achieved after each step of the hysteresis loop to the thickness of the Fe layers contributing to the total SQUID signal. The increase of the magnetization at 60 mT corresponds to the contribution of one of the AFM-coupled Fe layers and thus is interpreted as a spin–flop, i.e. a transition to the perpendicular orientation of magnetizations of the Fe layers [4]. Then, only this layer which is magnetized along the field contributes to the SQUID signal. From the magnetization value in saturation (i.e. for

![Fig. 1. SQUID magnetization curves for GaAs(001)/15Fe/9Cr/15Fe/5Cr/13Fe sample measured at RT along [110]. The top Fe layer saturates at a field of about 5 mT. Above this field, almost constant magnetization is measured showing that the Fe layers in the bottom 15 Fe/9 Cr/15 Fe trilayer are fully saturated in opposite directions. A sharp spin–flop transition is seen above 60 mT.](image-url)
15 + 15 + 13 ML of Fe all magnetized in parallel), one can conclude that the low-field loop corresponds actually to the top (13 ML thick) Fe layer, supporting the above interpretation. In this way, details of magnetization reversal can be followed in the top Fe layer only, with no contribution to the measured magnetization signal from the bottom (Fe/Cr/Fe)AFM structure (at the fields below the field at which the spin–flop transition occurs). After the magnetization reversal in the top Fe layer has occurred at low fields, the signal along the \([1\bar{1}0]\) direction increases linearly until all the spins in the Fe layers are oriented parallel (Fig. 2, inset, for positive fields only). Such a linear increase of the magnetization is typical for the AFM-coupled structure when it is measured along the hard axis of magnetization. Comparison of the magnetization curves measured along \([1\bar{1}0]\) and \([-1\bar{1}0]\) (Fig. 2) confirms that the uniaxial anisotropy of the easy axis oriented along the \([1\bar{1}0]\) direction exists in the Fe/Cr/Fe trilayer grown on GaAs(001).

Such a (Fe/Cr/Fe)AFM/Cr/Fe system of uniaxial anisotropy offers a unique possibility to follow magnetization reversal in the top Fe layer separately, with no contribution from the bottom (Fe/Cr/Fe)AFM structure. This is a simple way to separate the signal from one FM layer coupled to the second FM layer made of the same material. In order to test the method, we measured by SQUID the magnetization reversal along the \([1\bar{1}0]\) direction for the same sample (i.e. for the top Fe layer separated from the bottom (Fe/Cr/Fe)AFM structure with 5 ML of Cr), but at low temperatures (Fig. 2, open circles). At \(T=10\) K, a strong increase of the coercivity of the top Fe layer up to about 60 mT is observed in comparison to about 5 mT measured at RT. This effect has been observed previously for Fe films grown on Cr001) [19]. The origin of the coercivity increase at low temperatures is attributed to the spin–flop transition in Cr (coercivity starts to increase just below the spin–flop transition temperature), which influences the magnetization in the Fe layers. A similar observation was made by Hopster [20] who has shown how a single domain state of thin Fe films on Cr(001) transforms upon cooling into a state with locally varying in-plane magnetization directions. It has been suggested that the in-plane rotation is driven by frustration that favors 90° coupling [20]. This illustrates how the intrinsic structure in an AFM (Cr) can influence the properties of an interfacing FM layer (of Fe in this case) through the interface exchange coupling. In the bottom (Fe/Cr/Fe)AFM structure, the spin–flop transition occurs at low temperature (10 K) at a slightly higher field (75 mT) than that at RT (60 mT) (Fig. 2), again in agreement with other observations [21]. Phenomenologically, the higher field required to invoke the spin–flop transition at low temperature is easily understood due to the interlayer coupling that increases with the decreasing temperature [21].

When the magnetization reversal was examined along the \([1\bar{1}0]\) direction at low fields only, the magnetization in both Fe layers of the bottom (Fe/Cr/Fe)AFM structure was kept oriented antiparallel along the \([1\bar{1}0]\) direction due to the uniaxial anisotropy. After saturation (i.e. after the final step of the uniaxial-“layered-AFM” preparation), any magnetic coupling of the top Fe layer should be immediately seen by a shift of the minor loop to
positive or negative fields. This is due to the magnetization of the middle Fe layer (i.e. upper Fe layer of the \((\text{Fe/Cr/Fe})_{\text{AFM}}\) structure) which is pinned and cannot be reversed even at high fields. To which fields the loop would be shifted depends on the existing magnetic coupling (FM or AFM) and on magnetization orientation in the Fe layers of the \((\text{Fe/Cr/Fe})_{\text{AFM}}\) structure after the sample saturation. It is worth to note that in the case of any interlayer coupling existing between the top Fe layer and the \((\text{Fe/Cr/Fe})_{\text{AFM}}\) structure underneath, the magnetization of the \((\text{Fe/Cr/Fe})_{\text{AFM}}\) structure can be easily reversed adopting the magnetization orientation in the top Fe layer of the \((\text{Fe/Cr/Fe})_{\text{AFM}}\) structure. Then, the exchange field would be always negative regardless of whether the coupling is FM or AFM [22]. The minor loop corresponding to the top Fe layer, measured in this way, is shown in Fig. 3. The loop is of rectangular shape, symmetric and practically not shifted either to positive or to negative fields. In other words, the exchange-bias effect, expected for magnetization reversal of the Fe film being coupled to another Fe film of well-defined magnetization direction, does not exist. This lack of exchange biasing is an additional proof that actually the top Fe layer is only weakly magnetically coupled to the bottom \((\text{Fe/Cr/Fe})_{\text{AFM}}\) structure. It has been shown before for the Fe/Cr double superlattices that actually the exchange bias exists for the Cr spacer thicker than 13 Å [22] and oscillates with increasing thickness. Then, the magnetic properties of the top Fe layer are determined by the interface coupling to the spin lattice of the upper Cr layer, which is however dragged by the bottom \((\text{Fe/Cr/Fe})_{\text{AFM}}\) structure. The AFM order in the upper Cr film is stabilized by the presence of the Fe layer magnetized along the [1 1 0] direction and one could still expect exchange biasing caused by the interface coupling of the top Fe layer to the spin structure of the Cr spacer. The exchange-bias effect typical for AFM/FM interfaces has been reported mostly for the AFM layers with localized moments [10–12]. Only very recently, Yang et al. [14] have reported on the oscillatory exchange bias in epitaxial Ni$_8$Si$_{1-x}$Fe$_{19}$/Cr(001) system caused by the incommensurate spin-density waves in AFM Cr(001) layers.

![Fig. 3](image-url)

Fig. 3. In situ MOKE minor loops measured at RT for GaAs(001)/15 Fe/9 Cr/15 Fe sample along [1 1 0] (a), and for GaAs(001)/15 Fe/9 Cr/15 Fe/5 Cr/13 Fe sample along [1 1 0] (b), [−1 1 0] (c), and [1 0 0] (d). No loop is detected for GaAs(001)/15 Fe/9 Cr/15 Fe sample along [1 1 0] (a). This is due to AFM coupling between two Fe layers of the same thickness resulting in zero net ellipticity signals. Thus the loops measured for GaAs(001)/15 Fe/9 Cr/15 Fe/5 Cr/13 Fe sample actually correspond to the top Fe layer only. The loop which is not affected by 90° coupling is measured along the [1 1 0] direction, i.e. along the easy axis of magnetization (b). An opened loop of small coercivity is visible when the field is applied along [1 1 0] (c). This suggests that the [−1 1 0] direction is a preferable orientation for part of the spins in the top Fe layer. The observation is confirmed with the loop measured along [1 0 0] (d). At very low fields magnetization rotates to [−1 1 0] and only at higher fields (of about 5 mT) switches to [1 0 0] and saturates.
However, the exchange field they measured at RT is very small and vanishes for very thin Cr films due to reduced blocking temperature [14].

In order to analyze magnetic anisotropy in the top Fe layer, magnetization vs. the field was detected using in situ MOKE. We followed the magnetization reversal after each step of the structure deposition. No hysteresis loop was detected after deposition of the bottom (Fe/Cr/Fe)_{AFM} structure (Fig. 3a). This is due to the net magnetization of the AFM-coupled Fe layers of the same thickness balanced to zero at low fields. After the Fe/Cr/Fe)_{AFM}/Cr/Fe structure is completed, the measured MOKE loops correspond to the top Fe layer only (Fig. 3). The spins of the upper Cr spacer rotate minimizing their anisotropy energy, in consequence being aligned in the same direction as in the underlying (Fe/Cr/Fe)_{AFM} structure of the easy axis oriented along [110]. However, local distortions of the magnetization introduced by the interfacial steps make a frustration due to competing positive and negative exchange interactions (depending on the local thickness of the Cr layer). When the interfacial roughness correlation length is smaller than the domain wall width, the Cr layer cannot break into domains to locally satisfy the interfacial coupling [8]. This can result in average perpendicular coupling between the top Fe layer and the spins of the Cr spacer. In the case of a perfect 90° coupling, hysteresis loop of the top Fe layer measured along the [−110] direction should be of an easy-axis character whereas that measured along the [110] direction should be of a hard-axis behavior. Actually, the 90° coupling competes with the linear coupling which promotes the magnetization in the top Fe layer to be oriented along the [110] direction. Unless the top Fe layer is magnetically anisotropic with the easy magnetization axis oriented along [110], the loop which is not affected by the 90° coupling is measured along the [110] direction. The uniaxial anisotropy in the top Fe layer originates in the spin configuration which follows the anisotropy of the bottom (Fe/Cr/Fe)_{AFM} structure. This is the reason why the single-step rectangular loop is measured when the field is applied along the [110] direction (Fig. 3b).

Then, magnetization reversal in the same sample was examined at low fields along the [−110] direction. The minor loop corresponding to the top Fe layer is shown in Fig. 3c. It is clearly seen that at 10 mT magnetization is still unsaturated along [−110]. Nevertheless, an opened loop of small coercivity is visible suggesting that the [−110] direction is a preferable orientation for part of the spins in the top Fe layer. Note, that within this experiment, the magnetic field is applied along the hard-axis of the bottom (Fe/Cr/Fe)_{AFM} structure (i.e. along [−110]), whereas the magnetic response of the top Fe layer (which is actually measured) shows the easy-axis properties. This behavior suggests that perpendicular coupling between the top Fe layer and the bottom (Fe/Cr/Fe)_{AFM} structure exists. A kind of local energy minimum is reached for the magnetization of the top Fe layer oriented under 90° to the spin direction in the Cr spacer layer.

The top Fe layer can be easily magnetized, in particular along the high-symmetry [100] axis of the (001)-oriented bcc-Fe films. Thus, the loop measured along the [100] direction (as well as that measured along [010]) would be affected by any local energy minimum available for the rotating magnetization. In order to prove this idea, we examined the magnetization reversal in the top Fe layer along the [100] direction. The corresponding loop is shown in Fig. 3d. It is immediately seen that the magnetization switches in two steps. At low fields, exactly the same at which the magnetization switches when the field is applied along the [−110] direction, the magnetization drops down showing that in the top Fe layer the spins prefer to be oriented along [110]. The MOKE signal detected along [100] does not disappear completely, most likely due the part of the spins which are not frustrated and keep their orientation along the [110] easy axis. Only after a field of about 5 mT is applied, the magnetization switches to the field direction (i.e. to [100]) and saturates. The situation is fully symmetric, i.e. when the field is applied along the [010] direction, the magnetization reversal proceeds exactly in the same way with the preferred spin orientation along [−110] (not shown in the figure). This confirms that the magnetization alignment in the top Fe layer
originates in the spin coupling. Note, that an intermediate orientation along the [1 1 0] direction is not detected. This shows again that the minimum energy configuration is reached for the magnetization of the top Fe layer oriented perpendicular to the [1 1 0] direction, i.e. to the easy axis of magnetization in the bottom (Fe/Cr/Fe)_{AFM} structure and overlaying Cr film. The minimum exists due to the 90° coupling, not due to any other uniaxial anisotropy induced, e.g. by the growth conditions.

With the above analysis, we were able to follow the magnetization reversal separately in the top Fe layer only. We have shown that the anisotropy of the top Fe layer does not follow the anisotropy of the underlying Cr film. A local energy minimum exists for the magnetization of the top Fe layer oriented along the [\bar{1} 1 0] direction.

4. Conclusions

The results of our experiments show that the perpendicular alignment is a preferable orientation of magnetization in an Fe layer deposited on top of a Cr film. This means that 90° interlayer coupling exists between the Cr spin structure and the top Fe layer. Most likely such coupling is caused by the magnetic disorder in the Cr layer resulting from the interfacial roughness and competing positive and negative exchange interactions. This results in an average perpendicular coupling of the Fe layer to the spin lattice of the Cr spacer. Such surface spin frustration due to local variations of the Cr spacer thickness can be expected in the multilayer structures grown on GaAs(001) due to the substrate imperfections. The anisotropy of the top Fe layer, smaller than that of the bottom (Fe/Cr/Fe)_{AFM} structure, is crucial for the observation of the 90° interface coupling. Replacing Cr with a compensated AFM (like, e.g. NiO) allows a detailed analysis of the coupling between two Fe layers across the AFM spacer.

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