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Coupled perpendicular magnetization in Fe/Cu/Fe trilayers

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Abstract

Ultrathin epitaxial Fe films on Cu(100) with perpendicular magnetization have been used as templates for the preparation of FCC Fe/Cu/Fe trilayers. The magnetic anisotropy and the coupling of these films have been studied by in-situ magneto optical Kerr effect measurements and Kerr microscopy. The magnetic coupling of both Fe layers is found to be dominated by magnetostatic interaction. Adsorbate-induced spin reorientation in the top layer also causes spin reorientation in the bottom layer. The governing role of the Fe-vacuum interface for the magnetism of the whole trilayer is demonstrated.

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Epitaxial Fe layers on Cu(100) substrates are the subject of extensive research due to interesting magnetic and structural phases in the ultrathin film limit [1–3]. In particular, the appearance of perpendicular magnetization for films thinner than 11 monolayers (ML) [4] has stimulated extensive work and is commonly attributed to the contribution of the Fe–vacuum interface to the magnetic anisotropy energy [5]. In the present paper, FCC-Fe films with perpendicular magnetization are

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used as templates to form FCC Fe/Cu/Fe trilayers. It will be shown that the magnetization of both Fe layers is coupled with the easy axis along the surface normal. The perpendicular coupling as well as the observed formation of magnetic domains is found to be determined mainly by magnetostatic interaction between the Fe layers. Such systems of coupled perpendicular magnetization are exceptional and might be of particular relevance for giant magnetoresistive or magneto-optic applications [6].

The experiments were carried out in an ultrahigh vacuum (UHV) chamber system with a base pressure of 8×10^{-11} mbar [7]. The Cu substrates

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have been cleaned by cycles of Ar^+ ion sputtering, followed by annealing up to 750 K, until no contaminations of the Cu could be detected by Auger electron spectroscopy (AES). The films were grown by molecular beam epitaxy with evaporation rates of 0.3–0.5 ML/min, as calibrated with a quartz microbalance. During Fe deposition the substrate was kept at room temperature (RT = 295 K), while for Cu deposition the substrate was cooled to LT = 125 K. The magnetism of the samples was investigated in-situ by integral magneto optical Kerr effect (MOKE) measurements and in-situ Kerr microscopy [7].

Polar and longitudinal MOKE hysteresis loops, with the magnetic field applied perpendicular and parallel to the sample surface respectively, have been measured to determine the evolution of the magnetic anisotropy of the films after the preparation of each individual layer of the Fe/Cu/Fe trilayer. A 3 ML Fe film on Cu(100) shows the known result of perpendicular magnetization of this system. Deposition of just 1 ML of Cu on top of this film causes a spin reorientation into the plane. Intriguingly, the easy magnetization direction switches again out-of-plane after the deposition of the second Fe layer of 3 ML thickness. The magnitude of the observed polar Kerr signal for the trilayer is approximately twice the signal of the 3 ML Fe and thus corresponds to the sum of the signals of both individual Fe layers. Clearly, it is the presence of the top Fe layer that induces a perpendicular magnetization also in the bottom Fe layer. To probe the role of interfaces and magnetostatic interlayer interaction, the dependence of the trilayer magnetization on the thickness of both individual Fe layers as well as the sensitivity to electronic modifications at the Fe-vacuum interface is investigated and presented in the following.

The magnetization of the trilayer was investigated first as a function of the bottom Fe layer thickness, t_1 , with the thickness of the top Fe layer, t_2 , and of the Cu spacer, t_{Cu} , kept constant. A wedge of $t_1 = 0-15$ ML Fe was grown at RT on Cu(100) and investigated by MOKE at 200 K. The remanent (\blacktriangle) and saturation (\bigtriangleup) Kerr signals of polar MOKE loops as a function of Fe thickness are shown in Fig. 1. The well-known



Fig. 1. Polar remanent (\blacktriangle), polar saturation (\triangle) and longitudinal saturation (\diamondsuit) Kerr intensity of MOKE loops of a Fe wedge on Cu(100) as a function of Fe thickness, t_1 , taken at T = 200 K. Polar remanent (\bullet), polar saturation (\bigcirc) and longitudinal saturation (\square) Kerr intensity of MOKE loops of a trilayer, 3 ML Fe/2 ML Cu/ t_1 ML Fe, as a function of the bottom Fe layer thickness, t_1 , taken at 90 K.

magnetic phase transitions from ferromagnetic perpendicular to partly antiferromagnetic perpendicular ordering at ~ 4 ML and to in-plane (\diamond) at $t_1 \sim 12 \text{ ML}$ can be recognized [2]. For $4 \text{ ML} < t_1 < t_$ 12 ML, only the two top Fe layers are coupled with the magnetization pointing out-of-plane, whereas the other layers are antiferromagnetically coupled giving a zero net MOKE signal. Above 12 ML the magnetization reorients into the plane. The polar saturation signal drops to zero since the available field was not sufficient to saturate the film along the hard magnetization axis. By applying an inplane magnetic field, square hysteresis loops were obtained, thus indicating an easy magnetization axis parallel to the film plane. The longitudinal MOKE signal (\diamond) increases as a function of t_1 . Based on this sample, a trilayer of 3 ML Fe/2 ML Cu/t_1 ML Fe on Cu(100) was formed. The polar remanent (\bullet) and saturation (\bigcirc) Kerr signal, measured at 90 K, of the trilayer in Fig. 1 is clearly the sum of the signal of the Fe wedge plus a constant offset corresponding to the contribution of the top layer. Since the Cu-covered Fe wedge showed in-plane anisotropy, it is obvious that the top Fe layer controls the anisotropy in the bottom Fe layer. Both Fe layers are ferromagnetically coupled with perpendicular magnetization for all thickness $t_1 < 12 \text{ ML}$. Above $t_1 = 12 \text{ ML}$, both

layers show in-plane magnetization (\Box). It is now the bottom layer (\diamond) that pulls the top layer magnetization into the plane. The interaction is probably influenced by a structural transition from FCC to BCC in the bottom Fe layer [8], as well as the magnetostatic field of the bottom layer.

To further probe the role of the top layer on the total anisotropy of the trilayer, samples with varying top Fe layer thickness, t_2 , and constant t_1 and t_{Cu} were analyzed. For MOKE measurements at 135 K shown in Fig. 2, a Fe wedge of $t_2 = 0-4$ ML was grown on top of a 2 ML Cu/3 ML Fe bilayer. Again, the top Fe layer causes perpendicular magnetization in the trilayer, after in-plane magnetization was observed for the Cu/ Fe bilayer. The polar remanent (\bullet) and saturation (\bigcirc) Kerr signal of the trilayer is shown in Fig. 2(a). The data clearly show that: (i) For $t_2 > 1.3$ ML Fe, the saturation signal (\bigcirc) of the trilayer is larger than the polar MOKE signal of the Fe bottom layer (\triangle and dashed line). This means that also the magnetization in the bottom layer partially contributes to the polar MOKE signal of the



Fig. 2. (a) Magnetization of a trilayer, t_2 ML Fe/2 ML Cu/3 ML Fe, as a function of top Fe layer thickness, t_2 . Remanent (•) and saturation (\odot) Kerr intensity of polar MOKE loops before O₂ adsorption, taken at 135 K. Saturation (\triangle and dashed line) Kerr intensity of polar MOKE loop of 3 ML Fe/Cu(100) (bottom Fe layer). Saturation (\Box) Kerr intensity of longitudinal MOKE loops after O₂ adsorption. (b) Kerr images of the remanent state at $t_2 = 2.5$ ML (left) and $t_2 = 3.5$ ML (right) taken on a similar sample at 140 K.

trilayer. (ii) For $t_2 > 2 \text{ ML}$ Fe, the intensity of the Kerr signal of the trilayer corresponds to the sum of the signals of both individual Fe layers, assuming a linear proportionality between the thickness of the Fe layers and the MOKE signal. The magnetizations of the two lavers are out-ofplane aligned. (iii) Saturation and remanence have the same value only in the thickness range between $2.2 \text{ ML} < t_2 < 2.8 \text{ ML}$. Within this thickness range, square-shaped hysteresis loops are observed and clearly prove that the easy axis of magnetization for the whole trilayer is perpendicular. The magnetizations of the two Fe layers are ferromagnetically coupled. Outside this region, S-shaped magnetization loops were measured. For $t_2 >$ 3.5 ML and $t_2 < 1.5$ ML the remanence is zero. This thickness region of t_2 strongly depends on the relative thickness t_1 , t_2 , t_{Cu} , and on the sample temperature as well. By lowering the temperature, the interval of t_2 in which remanence and saturation have the same value becomes broader.

In-situ Kerr microscopy was used to analyze the magnetization of the trilayer in more detail. The Kerr images in Fig. 2(b) were taken at 140 K on a similar sample [9]. They show the magnetic domain configuration of the remanent state at positions corresponding to a top layer thickness of around $t_2 = 2.5 \text{ ML}$ (left) and $t_2 = 3.5 \text{ ML}$ (right) at the center of each image. Within these images the t_2 is increasing from left to right by ~0.2 ML. The black and white contrast in the image represents magnetic domains with opposite magnetization perpendicular to the plane. At $t_2 < 2.5 \text{ ML}$ and $t_2 > 3.5 \text{ ML}$ a transition from a single domain magnetization state to a multidomain state with domains of a few micrometers in size is observed. The appearance of the S-shaped magnetization loops can thus be identified as the result of the formation of domains with zero net magnetization when integrating over the area that is probed by the MOKE laser beam. As discussed in a previous work [9], the domain formation is driven by magnetostatics, and result of the competition between the gain of magnetostatic energy and the energy needed to create domain walls, which itself is related to the anisotropy.

The governing role of the Fe-vacuum interface for the trilayer magnetization is demonstrated in the following by gas adsorption experiments. The adsorption of small amounts of oxygen causes a modification of the electronic structure of the Fe-vacuum interface, and thus of the magnetic surface anisotropy. Changes in the magnetization are monitored by Kerr microscopy directly during the adsorption process. This technique has already proven to be very successful on LT (140 K) grown single Fe layers on Cu(100) [5]. Oxygen-induced changes in the magnetization of t_2 ML Fe/2 ML Cu/3 ML Fe with increasing O2 exposure are shown in the Kerr images in Fig. 3. Within these images, taken at zero field, the top layer thickness increases from $\sim 2.6 \text{ ML}$ to 3 ML. The transition from a single to a multi-domain state is visible at $t_2 = 2.9 \text{ ML}$. Exposure of the trilayer to oxygen



Fig. 3. Kerr images of the remanent state of t_2 ML Fe/2 ML Cu/3 ML Fe captured during oxygen adsorption at 125 K. Inset: longitudinal MOKE loop of the trilayer taken after the adsorption of 1.94 L O₂. Image size: $400 \,\mu\text{m} \times 110 \,\mu\text{m}$.

initially causes a transition of the whole sample into a multi-domain state after \sim 1–1.2 Langmuir (L) due to a gradual decrease of the surface anisotropy constant, K_S. Magnetic contrast disappeared at 1.8 L of O_2 . The magnetization loop depicted in the inset of Fig. 3 was taken after O₂ adsorption by longitudinal MOKE and shows that both layers exhibit uncoupled in-plane magnetization with slightly different coercivities. In Fig. 2, the saturation signal of longitudinal MOKE loops after O_2 adsorption (\Box) is shown as a function of t_2 . We find that, independent of t_2 , the magnetization of the whole trilayer has switched into the plane. The observed spin reorientation is attributed to an adsorbate-induced change of the magnetic interface anisotropy energy at the Fe-vacuum interface. Oxygen adsorption is reported to cause a charge transfer from iron to oxygen leaving behind Fe states with a majority character and causing a decrease of spin polarization and the density of states at the Fermi energy [10,11]. The strong response of the trilayer magnetism to modification of the electronic structure at the Fe-vacuum interface underlines the importance of this interface for the perpendicular magnetization. At the same time, the dependence of the bottom layer magnetization on the top layer magnetization is evident.

The experiments presented in this paper clearly prove the sensitivity of the magnetic anisotropy in epitaxial FCC-Fe layers to modification of the Fe-vacuum interface by Cu and O₂ adsorbates. The change of $K_{\rm S}$ upon Cu coverage is found to be much stronger than reported previously [12], probably due to slight differences in the interface morphology. An increase of the film roughness results in a smaller contribution of $K_{\rm S}$ [13], making less favorable out-of-plane easy axis of magnetization. In other words, it becomes easier to induce a spin reorientation transition into the plane by modifying the electronic structure of the Fe-vacuum interface. The perpendicular coupling is obviously dominated by magnetostatic interaction between the layers, as concluded from the domain formation in the film and the absence of oscillatory coupling with Cu thickness. Magnetic domains are formed within the film either if the domain wall energy decreases or if the magnetostatic energy

increases. This is the case when t_2 is small compared to t_1 due to the reduced total perpendicular anisotropy, which decreases the domain wall energy. On the other hand, domain formation for large thickness $t_1 + t_2$ decreases the magnetostatic energy.

In conclusion, perpendicular coupling was found for FCC Fe/Cu/Fe trilayers and ascribed to magnetostatic interaction between the Fe layers. The perpendicular magnetic anisotropy arises at the Fe–vacuum interface and governs the magnetization of the whole trilayer. The strongly reduced indirect exchange coupling in this system [14] is currently being investigated as the result of interface roughness, as predicted by theory [15].

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