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Influence of the FM/AFM interface morphology on the exchange coupling in epitaxial Co(001)/fct-Mn(001)

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The Co thickness dependence of the exchange interaction between ferromagnetic (FM) fct-Co(001) and antiferromagnetic (AFM) fct-Mn(001) thin layers, epitaxially grown on Cu(001) single crystals, is studied. In the pseudomorphic Co growth regime, oscillations of the coercivity (H_C) and the exchange bias (H_E) with a Co atomic monolayer period are observed. These oscillations are explained by a biquadratic exchange interaction component at the FM/AFM interface which, in this thickness range, is modulated by the layer-by-layer growth mode of Co on Cu(001). © 2004 American Institute of Physics. [DOI: 10.1063/1.1669114]

Since the first observation of exchange coupling between a ferromagnet (FM) in atomic contact with an antiferromagnet (AFM) almost half a century ago,¹ this issue has triggered an enormous amount of both theoretical and experimental activities.² The renewed, and in the last decade strongly increased, interest can be ascribed to the technological importance of exchange bias materials for the functionality of magnetoelectronic devices.³ Despite numerous efforts, a complete understanding of the FM/AFM exchange bias effect and its characteristic signatures [the shift of the magnetization loop along the field axis (H_E) and the largely enhanced coercivity (H_C)] has not yet emerged.

It is evident that the atomic and magnetic structures at the AFM/FM interface play a decisive role in the detailed interaction mechanism and thus for the magnetic properties of the coupled system. Even in the case of two single-crystalline films in atomic contact, which are both well ordered and from which their bulk spin structure are known, many different factors including lattice mismatch, strain relaxation, and changing step densities can lead to crystallographic and/or magnetic reconstructions and relaxations at the interface, which will certainly have an influence on the exchange coupling behavior.

In order to address the influence of the interface morphology, we performed an experimental study using single-crystalline Co/fct-Mn(001) bilayers with well defined interfaces grown on Cu(001) substrates. Recently, we have shown that epitaxially stabilized metastable fct-Mn(001) is antiferromagnetic and able to induce a unidirectional anisotropy in Co(001) layers, even at room temperature.⁴ The Co(001) template films grow on Cu(001) coherently and up to a certain thickness where the strain relaxation sets in a quasi

layer-by-layer mode. This allows us to study the FM thickness dependence and thus, as we will see, the influence of the step density on the exchange coupling in a well-controlled way.

The Co(001)/Mn(001) bilayers were epitaxially grown on atomically clean and flat Cu(001) single crystals (miscut $<0.1^\circ$) at 330 K in a multichamber molecular beam epitaxy (MBE) system (VG-Semicon V80M) with a base pressure better than 1×10^{-11} mbar. The Co layers were evaporated using an e-gun evaporator with feedback control of the flux, whereas the Mn films were prepared using a temperature stabilized and extensively degassed Knudsen cell. All nominal thicknesses were controlled by calibrated quartz-crystal monitors, with an accuracy of roughly $\pm 3\%$. During the growth, the pressure never rose above 5×10^{-11} mbar and the growth rates were 1–2 monolayers (ML)/min. Under these conditions, it was shown that the Mn adopts a slightly strained γ (fcc) structure with a c/a ratio of roughly 1.06, before it transforms at 50–60 ML to the thermodynamically stable complex α -Mn structure.⁴

The exchange coupling between the layers was studied in a number of different samples, employing both uniform Co films, as well as wedge-shaped Co layers using a moveable shutter positioned close to the substrate. The Co wedge slope for the sample used in this study was roughly 0.8 ML/mm. The uniform films were covered with 25 ML Mn, and the wedge with 16 ML Mn. To establish a well-defined exchange bias direction, all samples were annealed after the completed growth procedure for 30 min at 440 K in a magnetic field of 240 kA/m, which was aligned parallel to the [110]-Co easy axis direction. The magneto-optical longitudinal Kerr effect (MOKE) was used to characterize the magnetic behavior on a local scale with a focused HeNe-laser spot size of roughly 120 μm , resulting in a spatial resolution of 0.1 ML for the wedged samples. All measurements re-

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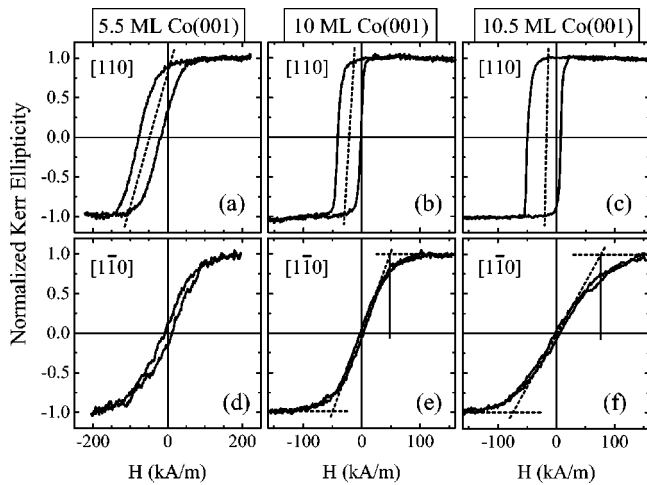


FIG. 1. Typical MOKE loops of Co/25 ML Mn bilayers for different nominal Co thicknesses, measured with H parallel [110] (anneal field direction) (a)–(c), or parallel [1 $\bar{1}$ 0] (perpendicular to anneal field) (d)–(f). Note the difference in field scales.

ported in this paper were performed at room temperature. Representative MOKE hysteresis loops of uniform Co/Mn bilayers with different nominal Co thickness (D_{Co}), and measured along the easy and hard axis of the unidirectional anisotropy, are collected in Fig. 1. Both H_E and H_C increase with decreasing D_{Co} , which is the consequence of the interface character of the exchange coupling, and has been observed several times before.² However, below 20 ML Co there is an indication that H_C , and somehow less pronounced but also H_E , seem to change in a *nonmonotonic* fashion [see Fig. 2(a)]. This becomes even more prominent by plotting the data versus the inverse Co thickness, resulting in a distinct scatter of the data points above $1/D_{Co} = 0.05 \text{ ML}^{-1}$ [see Fig. 2(b)]. This peculiar but completely reproducible behavior is demonstrated in more detail with the 10 and 10.5 ML Co samples.⁵ Despite showing almost the same H_E , H_C is sig-

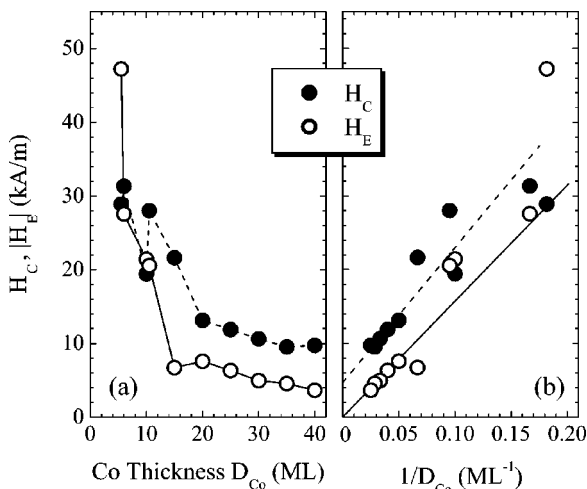


FIG. 2. (a) Dependence of the coercivity (H_C) and the exchange bias (H_E) on the Co thickness for the Co/25 ML Mn bilayers. The lines are guides to the eye. (b) The same data as in (a) now plotted vs the inverse Co thickness; here the lines are linear fits to the data points at low $1/D_{Co}$ values.

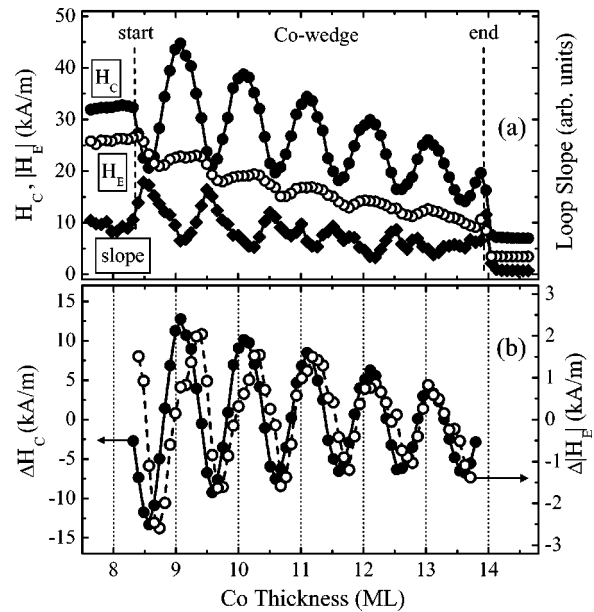


FIG. 3. (a) High precision measurements of the Co thickness dependence of H_C , H_E , and the loop slope of [110]-MOKE loops for a Co-wedge/16 ML Mn sample. (b) Effective oscillatory part ΔH_C and ΔH_E obtained by subtracting the thickness dependent nonoscillatory contribution from the data in (a). Note the different scales for ΔH_C and ΔH_E ; both data sets are scaled to each other to demonstrate the strong correlation between them.

nificantly smaller and the zero-field susceptibility of the hard-axis loops is larger for the thinner film, which is certainly counterintuitive [see Figs. 1(b), 1(c), 1(e), and 1(f)]. Actually, these observations point to an extra uniaxial anisotropy induced in the Co films which changes strongly and nonmonotonously with the thickness. Additional support for this interpretation, pointing to an interface-induced anisotropy, is given by the fact that the 10.5 ML easy-axis loop [Fig. 1(c)] shows slightly less slanting than the 10 ML loop [Fig. 1(b)] and distinctly less than the one at 5.5 ML [Fig. 1(a)].

The results of a detailed high precision experiment, employing a shallow wedge with D_{Co} in the coherent layer-by-layer growth regime,⁶ give a definite answer to the exact D_{Co} dependence of the exchange coupling [see Fig. 3(a)]. H_C and H_E actually oscillate with a ML period. The oscillatory components are achieved by subtracting a $1/D_{Co}^n$ power law from the data points in order to get rid of the intrinsic thickness dependence of the exchange coupling.

Several conclusions can be drawn from the plotted data in Fig. 3(b): (1) The oscillation amplitudes are also thickness dependent, e.g. decreasing with increasing D_{Co} , again indicating an *interface related* phenomenon. (2) Both ΔH_C and ΔH_E oscillate with roughly the same phase and with the same relative amplitude (at least in this Co and Mn-thickness range), indicating a *common origin*. (3) At a filled layer H_C has a maximum, and at a half-filled layer, a minimum,⁷ indicating that the *oscillating step density* of the Co template could be a determinant, e.g. effectively *lowering* H_C for a *rougher* interface.

A phenomenological understanding of the observed results is given in the framework of a modified Stoner–

Wohlfarth model, where a homogenous rotation of the FM magnetization in the presence of an applied field is assumed and restricted to the film plane.^{8,9} Based on these assumptions, the total areal energy density reads

$$E = E_{Zec} + E_A^{FM} + E_A^{AFM} + E_E. \quad (1)$$

The first term is the Zeeman energy of the FM layer in the applied field. The second and third terms are the magnetic anisotropy energies of the FM and the AFM, respectively, and the fourth term is the exchange interaction energy between the FM and the AFM layers. The interaction mechanism is postulated to be localized at the interface region and consists of two contributions:

$$E_E = -J_{E1} \cos(\theta_{FM} - \theta_{AFM}) + J_{E2} \cos^2(\theta_{FM} - \theta_{AFM}), \quad (2)$$

where θ_{FM} and θ_{AFM} are the angles between the FM magnetization and the FM anisotropy axis, and the AFM sublattice magnetization and the AFM anisotropy axis, respectively. The first term is the usual direct unidirectional exchange anisotropy with a bilinear coupling constant J_{E1} , giving rise to a shifted hysteresis loop. The second term is a so-called “spin-flop” term with a biquadratic coupling constant J_{E2} , which favors an orthogonal coupling of the AFM and FM spins at the interface.¹⁰

The biquadratic coupling constant J_{E2} could have different origins: (1) it was shown recently that a biquadratic term is intrinsically present as a higher order contribution both for uncompensated and compensated interfaces,¹¹ and (2) due to the frustrations by competing AFM exchange interactions between the AFM sublattices and the FM spins, a preferential 90° alignment between the FM and AFM spins is induced.

Based on these arguments and on earlier results reported for the similar epitaxial system NiMn/Co(001), where the observed hysteresis loops could only be satisfactorily understood by postulating a quite strong biquadratic interaction,¹² it is plausible that a sizable J_{E2} also exists in our system. The observed ML oscillations of H_C , as well as the zero-field susceptibilities, are then tentatively explained by a periodically changed frustration state correlated with the modulated step density at the interface.

The proposed simple model can at least qualitatively explain all the experimental observations described above. For large anisotropies of the AFM, θ_{AFM} is small and the biquadratic coupling term in Eq. (2) becomes mathematically equivalent to a *uniaxial interface anisotropy* in the FM film. This implies that all properties related to the biquadratic term, i.e., the amplitude of the coercivity oscillations and the zero-field susceptibility of the hard-axis loops, should scale with the inverse thickness. Furthermore, being an effective uniaxial anisotropy term, one expects from the biquadratic coupling a simultaneous modification of both, the coercivity and the zero-field susceptibility of the hard-axis loops. For example, an enhanced H_C should be accompanied by a decreased hard-axis susceptibility and vice versa, depending on the sign of the J_{E2} change. All these expectations are indeed

in full agreement with the experimental results described above.

Additional support for our model is given by the following consideration. If the easy axis of the uniaxial anisotropy lays orthogonal to the unidirectional easy direction, also in the easy-axis direction of the bilinear coupling, a hard-axis loop is expected under certain conditions, depending mainly on the value of J_{E1}/J_{E2} and the FM layer thickness. As can be seen in Fig. 1, this is actually the case. By plotting the loop shearing of the [110] MOKE loops versus the Co thickness of the wedge sample, as shown in Fig. 3(a), this effect can be observed in more detail. Indeed, for small D_{Co} a larger loop shearing is observed for half-filled FM layers, indicating a dominant orthogonal coupling for these conditions.

Up until now, we have gained only a qualitative phenomenological understanding of the step-induced biquadratic interactions. Further experiments at different temperatures, different angles of the applied external field, Mn thicknesses, and probably other well-defined AFM/FM combinations, are definitely needed to come to a more complete understanding of the exact role of the steps for the coupling mechanisms at the AFM/FM interface.

In summary, we have observed a pronounced influence of the Co thickness on H_C and H_E with a Co atomic monolayer period in high quality epitaxial Co/fct-Mn(001) bilayers. We presented experimental evidence that this phenomenon is caused by an additional biquadratic exchange interaction (favoring an orthogonal alignment of the spins at the AFM-Mn/FM-Co interface) which is modulated by the oscillating interface roughness induced by the layer-by-layer growth mode of Co on Cu(001). This observation demonstrates the prominent influence of the interface structure on the exchange coupling in epitaxial AFM/FM bilayer systems.

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