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Oscillatory biquadratic antiferromagnet/ferromagnet interface exchange coupling

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Abstract

A study of the influence of roughness at an antiferromagnet (AFM)/ferromagnet (FM) interface on the magnetic exchange coupling in epitaxial Co(0 0 1)/FCT-Mn(0 0 1) bilayers is presented. Co atomic monolayer thickness oscillations of the coercivity (H_C) and the exchange bias (H_E) are observed, which are clearly related to the modulated interface step density caused by the layer-by-layer growth mode of the Co(0 0 1) template films. Simulations based on a Stoner–Wohlfarth model suggest the existence of a strong intrinsic orthogonal coupling of the AFM/FM interface spins. This interaction is distinctly weakened by an increased interface roughness.

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Keywords: Exchange bias; Epitaxial growth; Co; Mn; Biquadratic coupling

Predominantly triggered by its distinct application potential in miniaturized magneto-electronic devices, the exchange anisotropy (EA) phenomenon stands since many years in the focus of magnetic research activities. Such an unidirectional EA is induced in a ferromagnet (FM) if it is brought in atomic contact with an antiferromagnet (AFM) [1]. Especially the most striking characteristic feature associated with EA, a shift of the hysteresis loop along the field axis (H_E), which is very often accompanied by an enhanced coercivity H_C, has attracted a lot of attention. For all efforts, a sufficient theoretical understanding of the EA is only slowly emerging. Most important, the microscopic origin of EA is still under debate, while it is evident that the atomic and magnetic structure at the AFM/FM interface plays a decisive role in the detailed interaction mechanisms.

In recent years, several theoretical models were developed to explain EA and the largely enhanced coercivities in coupled systems with *compensated* AFM interfaces. A particularly directive work was

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published by Koon [2]. He considered an orthogonal coupling between an FM and a compensated AFM accompanied by a slightly canted AFM spin structure at the interface responsible for the EA. It was Schulthess and Butler, who subsequently recognized that for perfectly flat interfaces this mechanism alone cannot account for H_E, but only for an enhanced H_C as the result of an additional uniaxial anisotropy induced by the interface spin–flop coupling [3]. Following the spirit of Malozemoff's early random interface field model [4], Schulthess and Butler concluded in the framework of a micromagnetic model that a H_E can only be achieved by randomly introducing (most likely roughness related) uncompensated AFM spins at the interface.

In order to systematically explore the role of roughness at a *compensated AFM/FM interface* in a well controlled way, we chose epitaxial Co/Mn bilayers, which were coherently grown on atomically clean and flat Cu(0 0 1) single crystals. By deposition under appropriate conditions (pressure below 5×10^{-11} mbar, $T \approx 330$ K, growth rate 1–2 monolayers (ML)/min) we have recently shown that Mn can be stabilized by molecular beam epitaxy (MBE) methods on high-quality Co(0 0 1) template layers in a metastable face centered tetragonal (FCT) structure up to 50–60 ML [5].

The FCT structure with a c/a of roughly 1.06 was established by a combination of several techniques, i.e. low-energy electron diffraction (LEED), X-ray photo/Auger electron diffraction (XPD/AED), and X-ray diffraction (XRD). Through extrapolating the structural and magnetic properties of high-temperature quenched manganese-rich M_xMn_{1-x} ($M = \mathrm{Cu, Fe, Ni, Pd}, \ldots$) bulk alloy crystals to a pure Mn content ($x = 0$), it is inferred that our FCT-Mn(0 0 1) films should be antiferromagnetic even at room temperature (RT), at least for large enough thicknesses where finite size effects are negligible [6,7].

For $c/a > 1$, as found for our layers, the bulk spin structure as measured with neutron diffraction is depicted in Fig. 1(a) [8]. If this spin arrangement is preserved for thin films and particularly also at the interface, a magnetically *compensated* FCT-Mn is expected with a c(2 × 2) antiferromagnetic in-plane *collinear spin structure*, which is actually an ideal magnetic structure to test the Koon/Schulthess/Butler (KSB) model [9].

The magnetooptical Kerr effect (MOKE) loops for two slightly different Co(0 0 1) template layer thicknesses, both covered with 25 ML FCT-Mn(0 0 1), are shown in Figs. 2(a) and (c). Prior to this magnetic measurement the samples were annealed at 440 K for 30 min in a magnetic field of 240 kA/m applied along the Co[1 1 0] easy-axis direction, in order to establish a well-defined exchange anisotropy direction in the Co-films. Both samples exhibit a moderate EA and a largely enhanced H_C (for uncovered or Cu-covered Co(0 0 1) layers in this thickness regime H_C lays

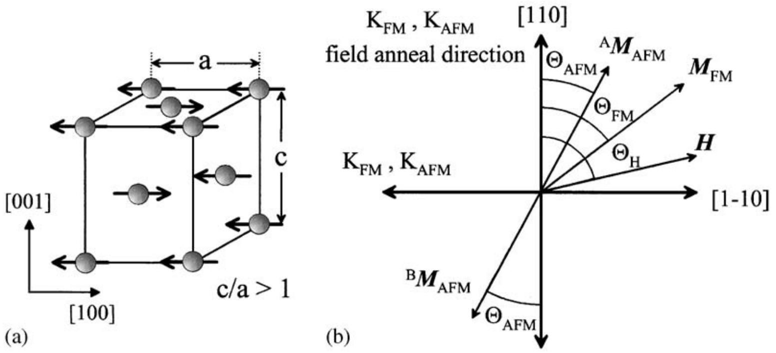

Fig. 1. (a) Schematic model of the $c(2 \times 2)$ antiferromagnetic collinear spin structure of tetragonally distorted γ-Mn(0 0 1) with $c/a > 1$. (b) Vector diagram involving the angles Θ_{AFM}, Θ_{FM}, and Θ_H related to the orientation of the AFM sublattice magnetization $^{A}\mathbf{M}_{AFM}$, the magnetization of the FM layer $\mathbf{M}_{FM}$ and the applied magnetic field $\boldsymbol{H}$ with respect to the easy axis of the AFM and the FM films designated by the corresponding anisotropy constants K_{AFM} and K_{FM}, respectively.

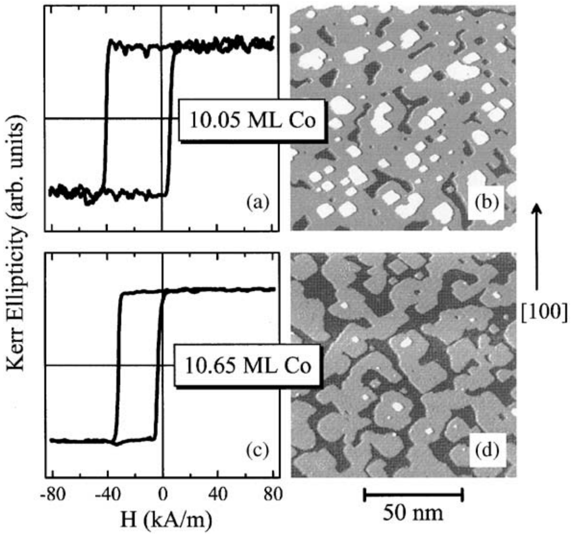

Fig. 2. Room temperature MOKE loops of (a) 10.05 and (c) 10.65 ML Co(0 0 1)/25 ML Mn bilayers, respectively, measured with H parallel [1 1 0] ($\Theta_H = 0°$, anneal field direction). (b) and (d) show the STM topographs for the uncovered 10.05 and 10.65 ML Co(0 0 1) layers before the Mn deposition.

well below 1 kA/m) even at RT, which is a clear evidence for an RT antiferromagnetic order in the FCT-Mn(0 0 1) layers.

While a high H_C value is expected for a compensated AFM in the framework of the KSB model, the sizeable EA (0.05 mJ/m^2) suggests that a certain amount of uncompensated Mn spins must be located at the Co/Mn interface. These uncompensated spins are most likely the result of a distinct roughness of the Co(0 0 1) base layers forming the template for the Mn films. Indeed, in the coherent growth regime (up to about 15 ML) the Co grows in an almost layer-by-layer growth mode on Cu(0 0 1), but nevertheless the Co surface displays for each coverage a certain amount of either channels and/or islands creating a stepped Co/Mn interface topology. This can be directly seen in the scanning tunneling microscopy (STM) pictures shown in Figs. 2(b) and (d). At the position of the interface step edges the Mn atoms experience a lowered symmetry and Mn coordination, most probably altering the local spin structure and creating a net uncompensated Mn moment.

Closer inspection of the magnetization loops measured at nominally filled [Fig. 2(a)] and half-filled [Fig. 2(c)] Co layers indicate that the exact interface topography has a measurable influence on the AFM/FM exchange coupling, especially H_C is distinctly larger for a nominally filled layer than for a half-filled layer. In Fig. 3 the results of a detailed high precision experiment, employing a shallow Co wedge and using MOKE with a spatial resolution <0.05 ML, are plotted. Both H_C and H_E oscillate with a Co ML period superimposed on a thickness dependent monotonic contribution (scaling roughly with $1/t_{Co}$).

The experimental results are characterized by several important observations: (1) The extrema of the oscillatory parts are located close to a nominally filled or half-filled Co template, respectively, as determined by an accurate STM study. (2) The oscillations in H_C and H_E are in anti-phase, e.g. at a filled Co layer H_C displays a maximum and H_E a minimum, respectively, in contrast to an earlier study were, for a thinner Mn overlayer (16 ML), no phase shift was found [10]. This apparent discrepancy is most likely explained by the changed magnetic properties of thin Mn layers in connection with the lowered magnetic anisotropy. An AFM thickness dependent study focusing on these aspects will be published

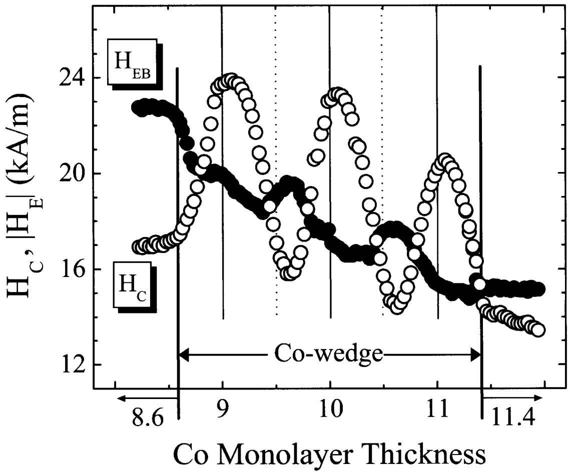

Fig. 3. High precision measurements of the Co thickness dependence of H_C and H_E of [1 1 0]-MOKE loops [see Figs. 2(a) and (c)] for a Co-wedge/25 ML Mn sample, measured at room temperature.

elsewhere [5]. (3) The oscillations are only seen in the coherent growth regime, where also a quasi layer-by-layer growth mode is observed. For thicker Co films, where strain relaxation and significant surface roughening sets in, the oscillations are effectively suppressed (not shown here).

All these facts clearly demonstrate that the H_C and H_E oscillations are induced by an altered interface step density. In the framework of the KSB model the lowered H_C and the enhanced H_E are ascribed to a decreased spin–flop coupling strength and an increased number of uncompensated interface spins, respectively. From the moderate oscillation amplitude of H_E we conclude that the number of uncompensated spins is only slightly influenced by the step density. On the other hand, based on the observation of a relative strong oscillation amplitude of H_C, is the orthogonal coupling significantly influenced by the step density.

Additional support for the spin–flop coupling model is given by magnetization reversal experiments with the field aligned at different angles Θ_H to the field anneal direction [see Fig. 1(b)]. For a nominally half-filled and a completely filled Co template layer [see Figs. 4(a) and (c)], a complete different behavior is noticed. Besides the larger coercivity for $\Theta_H = 0°$, also a much higher saturation field is observed in the hard-axis direction ($\Theta_H = 90°$) for a filled Co layer, both evidencing that the uniaxial anisotropy induced by the orthogonal interface coupling in the Co films is largest for an ideally flat interface.

Simulations based on a modified Stoner–Wohlfarth model, assuming to the film plane restricted homogenous rotations of the magnetizations, were performed. All relevant energy terms, like e.g. the FM Zeeman energy, the fourfold cubic FM in-plane anisotropy, the cubic AFM anisotropy, and the interface exchange interaction energy were taken into account. The interface exchange is phenomenologically described by two independent interaction energy contributions [for the definition of the angles see Fig. 1(b)]:

$$E_E = -J_{E1}\cos(\Theta_{FM} - \Theta_{AFM}) - J_{E2}\cos^2(\Theta_{FM} - \Theta_{AFM}), \quad (1)$$

with J_{E1} and J_{E2} the effective, over the interface area averaged, bilinear and biquadratic coupling constants, respectively, the former giving rise to the loop shift and the latter favoring an orthogonal alignment of the interface spins.

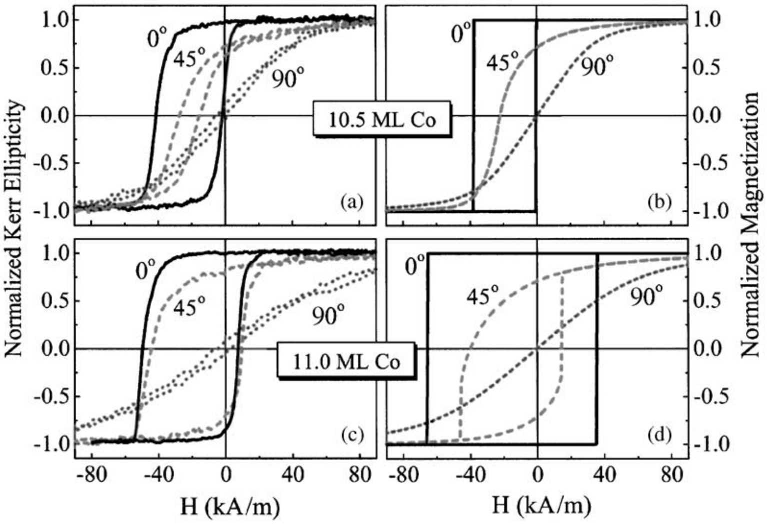

Fig. 4. Longitudinal component of the magnetization during a magnetization reversal obtained experimentally by MOKE [(a) and (c)] and calculated using a modified Stoner–Wohlfarth model [(b) and (d)] for different angles Θ_H of the applied field with respect to the anneal field direction [1 1 0].

Using reasonable values for the anisotropies ($K_4^{\mathrm{Co}} = 5\,\mathrm{kJ/m^3}$, $K_4^{\mathrm{Mn}} = 50\,\mathrm{kJ/m^3}$) and the coupling constants ($J_{\mathrm{E1}} = 0.07\,\mathrm{mJ/m^2}$, $J_{\mathrm{E2}} = 0.025\,\mathrm{mJ/m^2}$), a good agreement between the measured and the simulated magnetization reversal loops can actually be achieved for the 10.5 ML Co layer [see Figs. 4(a) and (b)]. All essential features are satisfactorily reproduced for all field directions, like for example the hard-axis saturation behavior and the asymmetrical reversal for the intermediate axis ($\Theta_H = 45^\circ$).

By enhancing the biquadratic coupling *almost four-fold* to $J_{E2} = 0.09\,\mathrm{mJ/m^2}$ and keeping all other parameters constant, also the 11.0 ML loops are basically reproduced [see Figs. 4(c) and (d)], except the H_{C} values which are always overestimated in a Stoner–Wohlfarth approach, simply because it neglects possible domain nucleations and thermal activations. Nevertheless, the simulations seem to support the KSB suggestion that for a compensated AFM/FM interface intrinsically a relatively strong orthogonal coupling exists and that this interaction is significantly lowered by topological interface roughness.

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