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Exchange bias in the Fe/$KCoF_3$ system: A comprehensive magnetometry study[a)]

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Exchange bias was studied in the Fe/$KCoF_3$ ferromagnet/antiferromagnet system. $KCoF_3$ can be deposited onto single crystal of Fe, either in the polycrystalline or single crystal form, depending on growth conditions. The samples were grown by molecular beam epitaxy on Ga-terminated GaAs (100) wafers. We study effects of the crystal state of the fluoride, thickness of the Fe film, crystallographic orientation of the Fe, and temperature on exchange bias. The structures with single crystal $KCoF_3$ show that the exchange bias is well correlated with the coercivity at low temperatures and vanishes at a temperature close to the Néel temperature. Both the magnitude of the exchange bias and the blocking temperature of the samples with the polycrystalline fluoride were significantly reduced compared to the single crystal structures. As the Fe film thickness was increased, the exchange bias decreased for all samples. In contrast, the blocking temperature remained unchanged for the samples with the single-crystal fluoride. The exchange bias measured along the easy anisotropy axis of the Fe was slightly larger than that measured along the hard axis. In addition, all samples exhibited a weak training effect.
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The exchange bias originates from exchange interactions among magnetic atoms at the ferromagnet/antiferromagnet interface and it manifests itself as a shift of magnetization hysteresis loops. It also contributes to magnetic anisotropy as a unidirectional anisotropy component and can be measured using torque magnetometry or ferromagnetic resonance techniques. In spite of numerous works on exchange bias systems during the past decade the mechanism of exchange bias is still not fully understood.[1–4]

Exchange bias exists in a variety of nanostructured systems including granular and layered structures. The system we studied consists of a single crystal (100) Fe film and a $KCoF_3$ film, which is a model Heisenberg antiferromagnet. The key feature of this system is that the antiferromagnet can be grown on the Fe film in either single crystal or polycrystalline forms. It has been recently found that the crystalline form of the fluoride significantly modifies magnetocrystalline anisotropy of the adhering Fe film.[5,6] This article is devoted to a comprehensive study of exchange bias in this interesting system.

A molecular beam epitaxy system was used to deposit the exchange bias system. A few monolayers of Fe were deposited on a Ga terminated (100) GaAs substrate as a seed layer. Then, single crystal (100) Fe films with thicknesses from 1 to 3 nm were deposited on 80 nm Ag templates. Films of $KCoF_3$ with a thickness of 30 nm were deposited onto the Fe layer using an electron-gun evaporator. Single crystals of $KCoF_3$ grow at low deposition rates (below 0.1 nm/s) and at elevated temperatures ($\sim 180\,°C$). Room temperature deposition with the rates above 0.2 nm/s resulted in a columnar growth of polycrystalline film, with a typical diameter near 5 nm. Moreover the direction of growth for these columns varies from grain to grain. For the single crystal $KCoF_3$ a typical lateral grain size is significantly larger (above 50 nm) but dislocation defects at the Fe/fluoride interface are visible, separated by distances of 20–50 nm. The exchange bias was measured for $5\ K \leqslant T \leqslant 300\ K$, using a SQUID magnetometer. In addition some results were verified by ferromagnetic resonance measurements at 24 K.

Typical magnetic hysteresis curves measured along the easy and hard axis are presented in Fig. 1 for the sample with 1.3 nm thick Fe (001) and a single crystal $KCoF_3$. They are superpositions of hysteresis loops representing the two Fe layers. The central part of the loop refers to the exchange-biased Fe layer and it consists of two magnetization jumps, which are asymmetric with respect to the zero field. The approach to magnetic saturation at higher fields reflects magnetization behavior of the thinner seed layer, which is separated by a thick Ag buffer layer from the exchange biased

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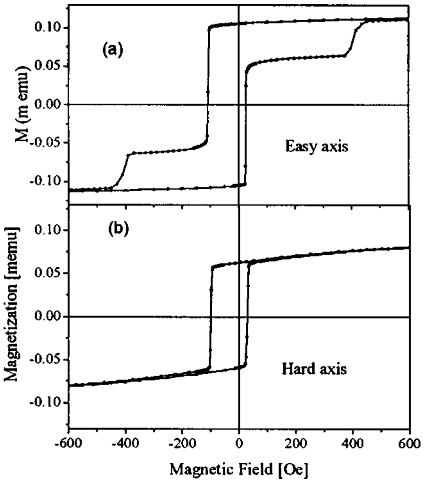

FIG. 1. Hysteresis loops for the single crystal Fe/KCoF$_3$ bilayer deposited on Ag template with 5 monolayers thick Fe seed layer. Magnetic field during cooling and measurement was applied along: (a) easy magnetization axis [001] and (b) hard magnetization direction [011] for Fe (001).

layer. Note that the high field jumps are symmetric about zero and not biased.

It is worth noting that the Fe seed layer shows abrupt magnetization reversal when magnetized along the easy magnetization direction and there is a gradual change and a substantially smaller hysteresis in the case of the magnetic field applied along the hard axis. This behavior of the seed layer is characteristic of single domain magnetization. In contrast the thicker exchange-biased layer does not exhibit such distinct differences for different crystallographic orientations with respect to applied field, which seems to indicate its multidomain structure. Our studies are focused on the layer showing exchange bias effects, therefore the magnetic behavior of the seed layer will be skipped in the further discussion.

We measured the temperature dependence of the exchange bias for several samples with Fe thicknesses varying from 1.05 to 2 nm. Typical results for the samples with single crystal and polycrystalline fluoride are presented in Fig. 2. In both cases the thickness of the Fe (001) film is 1.05 nm. These SQUID measurements were additionally verified by ferromagnetic resonance measurements for selected samples at 24 K. We found reasonably good agreement between the shift of the hysteresis loop and unidirectional anisotropy evaluated from the anisotropy curves measured by ferromagnetic resistance.

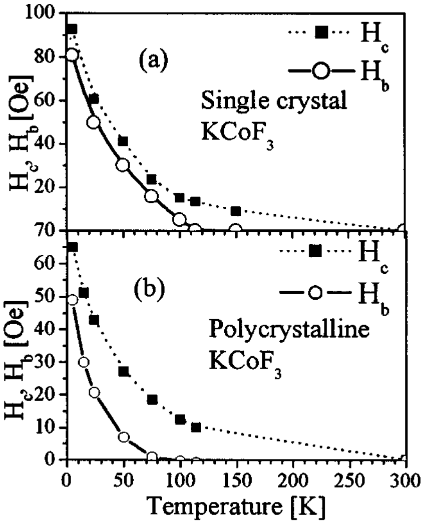

FIG. 2. Temperature dependences of coercive force (H_c) and exchange bias (H_b) for Fe/KCoF$_3$ structures with single crystal Fe and (a) single crystal fluoride and (b) polycrystalline fluoride.

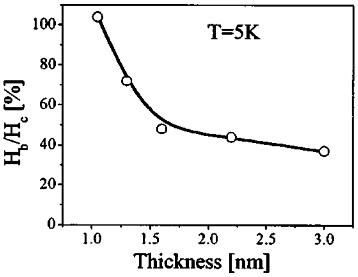

FIG. 3. The dependence of exchange bias to coercivity (H_b/H_c) ratio on the Fe film thickness in samples with single crystal fluoride. SQUID measurements were done at 5 K.

We found that for the samples with single crystal fluoride, there is a good correlation between temperature dependences of the coercivity and the exchange bias with a similar decrease with temperature as demonstrated in Fig. 2(a). Actually such correlation is expected for exchange bias systems in single domain models[7] because both quantities are functions of the magnetization, anisotropy, and thickness of the constituent magnetic layers. The exchange bias to coercivity ratio for the thinnest samples was in the range from 90% to 104% at 5 K and it decreased to zero at the temperature close to the Néel temperature (114 K). In contrast, samples with polycrystalline fluoride had a smaller initial exchange bias compared to the coercive field (65%–90%) and it decreased much faster with the increasing temperature as seen in Fig. 2(b). As a result the blocking temperature was substantially lower (from 50 to 75 K).

Samples deposited on GaAs wafers with different roughness had significantly different coercivities measured at 5 K—H_c varied from 65 Oe for epitaxy-ready GaAs substrates to over 300 Oe for the roughest substrate surfaces. The enhancement of the coercivity for rougher substrates can be explained by perturbed growth of the film on such surfaces and consequently increased number of defects, which obstruct domain wall movements. Because of the scatter in coercivity values on one hand and correlation between exchange bias and coercivity on the on the other, we indicated in our initial articles[5,6] that selected samples with polycrystalline fluoride (with high H_c) had larger exchange bias than those with single crystalline KCoF$_3$, however the ratio H_b/H_c for the large group of samples measured is larger for the single crystalline fluoride. This fact is demonstrated in Fig. 3, which shows the H_b/H_c ratio dependence on the thickness of the Fe film for the single crystal fluoride samples. Since we made a few samples with each thickness the points in the graph represent averaged results for samples with the same thickness. The ratio of exchange bias to coercivity decreases quickly from 104% to about half of this value as the Fe layer thickness is increased by only 4 atomic layers. Surprisingly, decay of exchange bias is much slower for larger thicknesses of Fe and it is still above 35% for

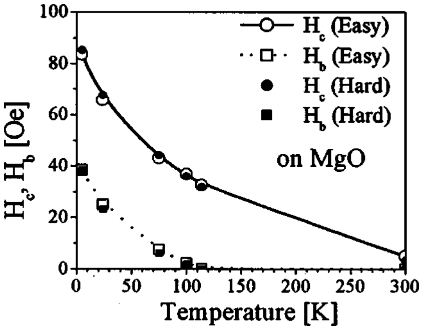

FIG. 4. The temperature dependence of coercivity and exchange bias for single crystal Fe/$KCoF_3$ bilayer deposited on MgO substrate.

samples as thick as 3 nm. This suggests that the dependences of the coercivity and exchange bias on thickness (d) does not both follow the $1/d$ law[7] as for the majority of exchange bias systems.[7,8] Our data indicate that the exchange bias follows the $1/d$ dependence reasonably well while the coercivity does not. One possibility to explain this is that the exchange bias is an interface phenomenon while the coercivity of the Fe film depends on both interface and bulk effects. This idea is supported by the fact that there is no dramatic change in the coercivity around the Néel temperature.

The samples with the polycrystalline fluoride showed similar decay of the H_b/H_c ratio with the Fe thickness for the thinnest Fe films, however their results were reduced compared to those for the samples with the single crystal $KCoF_3$. One possibility to explain this is that above 1.5 nm the interface topography between the Fe and the fluoride remains the same while below this thickness the interface roughness could change with Fe thickness. The exchange bias depends on the direction of the applied field with respect to the crystallographic axes. Both the coercive field and exchange bias were about 10% smaller when measured along a hard anisotropy axis (110) than those measured along an easy magnetization direction (100).

To understand the role of the substrate, we deposited a Fe/$KCoF_3$ structure with 10 monolayers of Fe and 30 nm of single crystal fluoride on an MgO substrate with a similar Fe seed layer and Ag template. In Fig. 4 we again plot the behavior of the bias and coercive fields as a function of the temperature. In this case the exchange bias curve was again parallel to the coercivity curve and it again approached zero close to 114 K, but the decay of the coercivity had a slightly different character than that for the films deposited on GaAs. For the sample on the MgO substrate, the coercivity and exchange bias do not depend on the crystallographic direction. Coercivity at the Néel temperature decreased to only about half of its low temperature value and it was distinctly larger at room temperature than that for the samples deposited on GaAs. This suggests that magnetoelastic effects may be responsible for the observed differences in the temperature dependence of coercivity and exchange bias for samples deposited on different substrates. This interpretation involves different mismatches between the substrate and template structures and different thermal expansion coefficients for GaAs and MgO substrates. Magnetoelastic effects may be quite an important factor for understanding this behavior because of the giant magnetostriction of $KCoF_3$.[8,9]

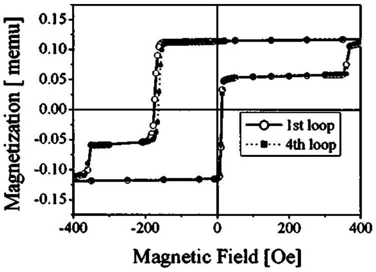

FIG. 5. Training effect in the exchange biased structure. Dashed line and filled squares represents the fourth cycle of the hysteresis loop. The sample with 1.05 nm of Fe and 30 nm single crystal fluoride was deposited on epitaxy-ready GaAs.

It is also important to mention that samples with the polycrystalline or the single crystal antiferromagnet followed the theoretical prediction of a model for the temperature dependence of exchange bias only if unstable grains play a substantial role.[10] However, in this case one expects the blocking temperature to be substantially below the Néel temperature, which is not observed for the single crystal sample. We note that the temperature dependence of the exchange bias is a very rich and complex phenomenon and can depend significantly on the grains and magnetic properties of the ferromagnet and antiferromagnet.[11] After a few hysteresis loops one of the hysteresis branches was shifted toward lower fields by a few Oersteds. The positive branch of the hysteresis loop remained unchanged as illustrated in Fig. 5. In the case of FMR measurements, a few rotations of the bias field were enough to significantly reduce the unidirectional anisotropy. This reduction in exchange bias was from 5% to 7%. This behavior is consistent with our interpretation[12] based on Kouvel's model.[13] As expected, there is no training effect for the seed layer—magnetization reversal at higher fields is unaffected by the number of cycles. Both poly- and single-crystal fluoride samples exhibited a similar training effect. This is curious in that one normally associates training with a distribution of pinning sites and local anisotropies. One might expect that the distribution should narrow for the single crystal film and widen for the polycrystalline film. One possibility is that the training may be most strongly affected by defects at the interface region, which exist in both the poly- and single-crystal fluoride structures.

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