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Freezing field dependance of the exchange bias in uniaxial FeF2-CoPt heterosystems with perpendicular anisotropy

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Freezing field dependence of the exchange bias in uniaxial FeF$_2$–CoPt heterosystems with perpendicular anisotropy

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Abstract

The exchange bias effect is measured for the first time in FeF$_2$–CoPt heterosystems with perpendicular anisotropy. The exchange field exhibits a strong dependence on the axial freezing field. This behavior is explained in terms of the microscopic spin structure at the interface, which is established on cooling to below T_N. We calculate the dependence of the spin structure on the freezing field within the framework of an Ising model. It takes into account the Zeeman energy as well as an antiferromagnetic exchange coupling between the adjacent layers at the interface. © 2000 Elsevier Science B.V. All rights reserved.

1. Introduction

Since the pioneering observation in 1956 of the exchange bias effect on small ferromagnetic Co particles which are embedded in their antiferromagnetic oxide [1], there is a renewed interest in the investigation of the exchange bias effect in well-defined ferromagnetic (FM)/antiferromagnetic (AF) layered heterosystems. One of the best-understood model systems consists of single crystals or textured films of FeF$_2$ covered with various ferromagnets [2–6]. After field-cooling to below the Néel temperature of FeF$_2$, $T_N = 78$ K, the hyster-esis loop of the FM film is usually shifted towards the field direction opposite to the freezing field, H_F, by an amount $H_E < 0$, thus denoting a negative exchange bias field. The situation changes for high freezing fields, where positive exchange bias, $H_E > 0$, may be encountered [7]. This effect has been explained qualitatively by various authors, all of which assume AF coupling at the interface between FM and AF subsystems [7–9].

In order to facilitate a deeper understanding of the dependence of H_E on the strength of H_F and to develop a quantitative description, a novel experimental approach is presented in this paper. In contrast with the generally used in-plane anisotropy of both the AF crystal and the FM layer we investigate for the first time the exchange bias in an FeF$_2$/FM-system involving exclusively perpendicular uniaxial anisotropy.

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In accordance with the drastic reduction of the degrees of freedom of the spin variables in uniaxial systems, the complexity of possible spin structures [9–11] is largely reduced. Here we investigate the exchange bias arising at an $FeF_2(0\,0\,1)$ plane covered by a Co/Pt multilayer with uniaxial perpendicular magnetic anisotropy, and describe the experimental data within an Ising model approach. It takes into account the competition between the exchange and the Zeeman energies at the interface as well as its topographic roughness due to the substrate. It turns out that the theoretical description of the H_E vs. H_F data requires, again [7–9], AF coupling between the spins of the adjacent layers at the AF/FM interface.

2. Experimental procedures

We use the (0 0 1) plane of an FeF_2 single crystal with (1 0 0) (0 1 0) (0 0 1) orientation and volume $3 \times 3 \times 2.7\,mm^3$ as the AF partner of our uniaxial heterosystem. The (0 0 1) orientation is checked by conoscopy using a polarizing microscope. Before transferring the single crystal into the UHV chamber, the (0 0 1) plane is polished to optical flatness with 2.5 μm diamond paste. A multilayer $(Co3.5\,Å/Pt12\,Å)_3$ is deposited at 500 K under UHV conditions by thermal (Co) and electron beam evaporation (Pt) onto the (0 0 1) surface of the FeF_2 crystal. The deposition rate is controlled by piezoelectric quartz resonators during the growth process. In addition, the thickness is determined by ex situ X-ray small-angle scattering. The result agrees with the nominal thickness within an error of 10%. In order to prevent oxidation, the last Pt-layer is covered by an additional Pt-layer of 8 Å thickness. By this measure, ex situ investigation of the $FeF_2(0\,0\,1)/(Co\ 3.5\,Å/Pt\ 12\,Å)_3/Pt\ 8\,Å$ heterosystem becomes possible.

Perpendicular magnetic anisotropy has been ascertained by separate investigations of identical Co/Pt multilayers as prepared on glass substrates at 500 K in accordance with existing literature [12,13]. Despite their rather complex structure, the usefulness of Co/Pt multilayers becomes obvious when taking into account the advantages of combining perpendicular anisotropy on the one hand

with a high magnetic moment on the other hand. The latter one causes an adequate contrast between the magnetization of the FM layer and the large field-induced magnetization of the bulk antiferromagnet. For example, in Fe-layers perpendicular magnetic anisotropy is possible only in the ultrathin limit [14,15].

Magnetometric measurements were done with a commercial 5 T SQUID system (Quantum Design MPMS5S). Hysteresis loops are performed after heating the sample to 200 K and subsequent cooling down to 10 K in various applied axial magnetic freezing fields. While the exchange bias shifts the hysteresis loop along the field axis, a shift along the axis of the magnetic moment is occasionally encountered in FeF_2/FM systems [2]. It is due to piezomagnetism, which is allowed by symmetry in rutile-type AF compounds [16] and may be induced by residual shear stress [17]. Under these circumstances the exchange bias is safely determined by subtracting the ascending and descending branches of the hysteresis loop thus eliminating both the constant piezomagnetic and the large moment of the FeF_2 crystal being proportional to the field.

The resulting curve is best fitted by a Lorentz function. Its shift with respect to the line $H = 0$ determines the exchange bias field H_E. Note that the details of the fitting function do not change H_E within the errors of the fitting parameters. In contrast with the conventional determination of the exchange bias field by calculating $H_E = (H_1 + H_2)/2$ from the intercepts $H_{1,2}$ of $M(H)$ with the H axis, our novel method to extract H_E involves the data of the entire hysteresis loop and is, hence, assumed to be more accurate.

3. Experimental results

Figs. 1a, b and c show the hysteresis loops of the $FeF_2(0\,0\,1)/(Co\ 3.5\,Å/Pt\ 12\,Å)_3/Pt\ 8\,Å$ heterosystem for axial freezing fields $\mu_0 H = 0$, 0.2 and 5 T after subtraction of the quasi-linear magnetic contribution of the bulk antiferromagnet, respectively. Figs. 1a′–c′ (circles) display the results after subtracting the ascending and descending branches of the loops. The results of the best Lorentzian fits are
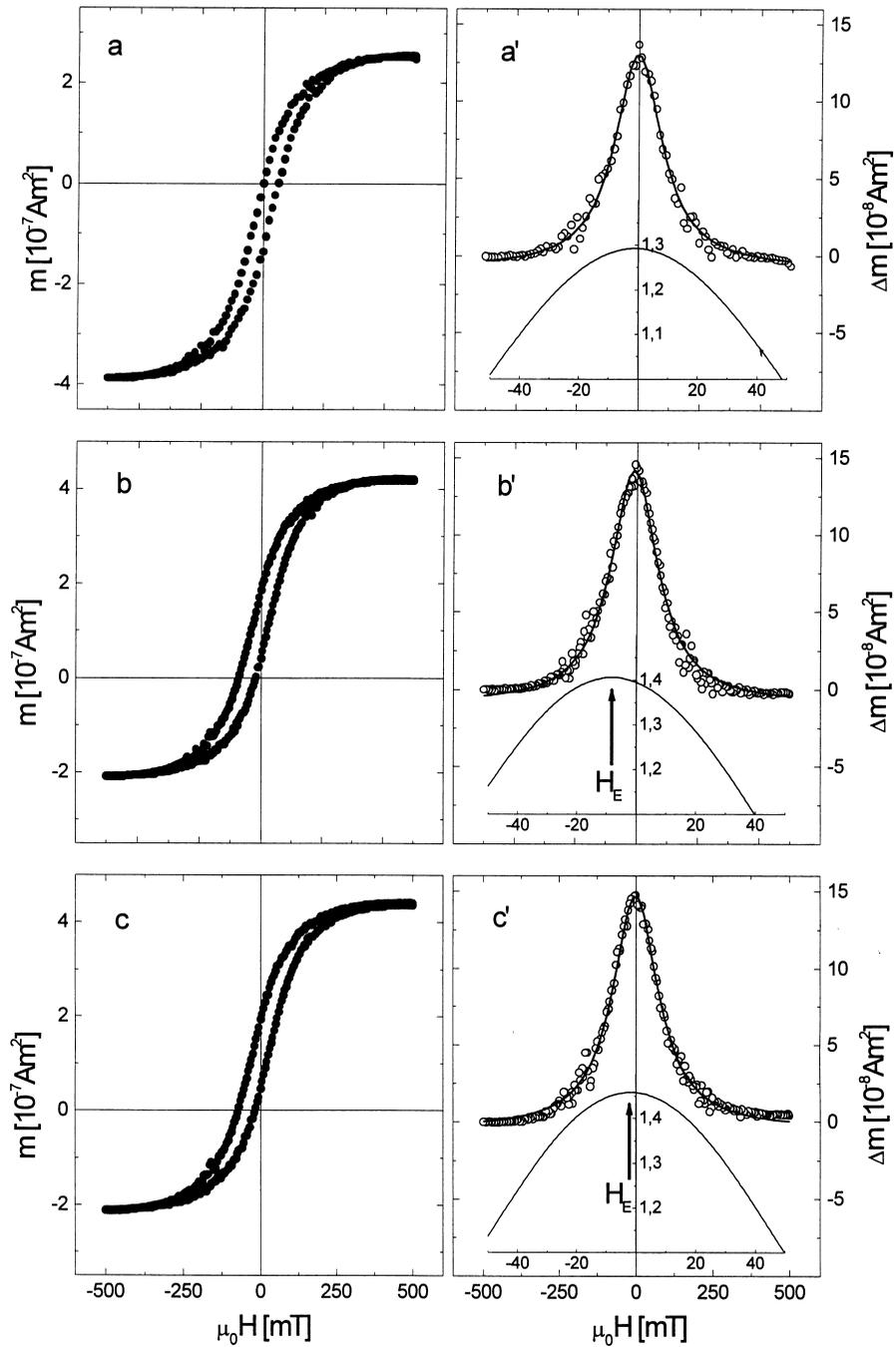
Fig. 1. Hysteresis loops (a–c) of the $FeF_2(0\,0\,1)/(Co\ 3.5\ \text{Å}/Pt\ 12\ \text{Å})_3/Pt\ 8\ \text{Å}$ system for freezing fields 0 (a), 0.2 (b) and 5 T (c) and difference curves (a′–c′) between descending and ascending branches (open circles, see text) with corresponding best fits to Lorentzian functions (solid lines).
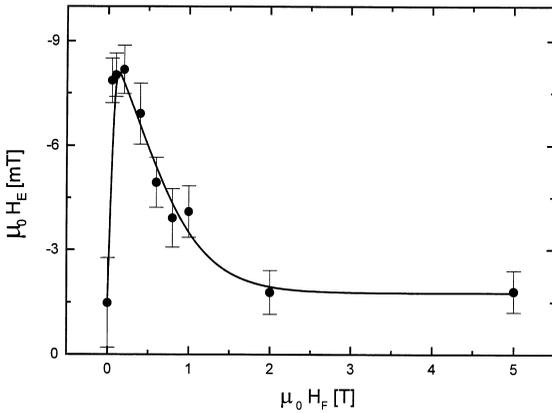
Fig. 2. Dependence of the exchange bias on the freezing field. Experimental data (circles with error bars) and their best fit to Eq. (10) (solid line) are shown.

shown by solid lines for the total field range and, in the insets, in the vicinity of the maximum. It is seen that the exchange bias field, H_E, as determined by the peak position of the fitting curve (arrows) is largest for a small field, $\mu_0 H_E(0.2\ \text{T}) = 8.5$ mT, whereas it decreases considerably in a high field, $\mu_0 H_E(5\ \text{T}) = 2.2$ mT.

This tendency is confirmed by Fig. 2, which shows the freezing field dependence of H_E for various values of $\mu_0 H_F$. In the weak H_F limit $|H_E|$ shows a steep increase with increasing freezing field up to a maximum at $H_F \approx 0.1$ T. Then $|H_E|$ decreases with increasing H_F and nearly vanishes for fields above 2 T. It is noticed that the value of H_E remains negative, in contrast with recent results on FeF_2-based systems with planar anisotropy [6,7]. It should be stressed that the measurements were done in arbitrary order at different values of H_F in order to avoid any systematic influence of a training effect on the data, although no effect of the number of measurements on the value of the exchange bias could be observed. This was proven by a series of measurements with constant freezing field. Within the errors of the exchange bias field no significant effect was observed.

4. Theory and discussion

Similar drastic changes of H_E vs. H_F as shown in Fig. 2 were reported previously for FeF_2Fe bi-

layers exhibiting in-plane anisotropy [7]. Qualitatively, the occurrence of positive exchange bias was explained by a competition between the AF–FM exchange interaction and the coupling energy between the AF surface layer and the magnetic field, H_F. In weak freezing fields the AF–FM exchange prevails over the Zeeman energy gained by the AF interface layer in the applied field. The dominant AF exchange coupling results in the usual negative exchange bias, $H_E < 0$. However, in high freezing fields and under the constraint of AF interface exchange interaction it may happen that the Zeeman energy overcomes the exchange energy. In that case, the FM and the AF topmost layers become aligned with H_F. When freezing in this unfavorable spin configuration, this will give rise to backswitching of the FM magnetization in zero external field, hence, producing a positive exchange bias, $H_E > 0$.

In order to describe quantitatively the strong dependence of the exchange bias on the freezing field H_F as shown in Fig. 2 or previously [7] we have to consider the actual spin structure at the AF/FM interface encountered at the Néel temperature, below which AF domain structure is established. From the above consideration we may anticipate that intermediate H_E values will occur in intermediate freezing fields H_F, which do not completely align either the FM or the AF spins at the interface. Since it is known that the exchange bias effect depends on the spin structure at the AF/FM interface, or more exactly, on the energy gained by exchange interaction, $\sum_{ij} J_{ij} S_i^{AF} S_j^{FM}$ [18–20], it will be useful to consider equilibrium thermodynamics of the interface at $T \approx T_N \ll T_C$ (= FM Curie temperature) under the constraint of a fixed external field, H.

The Curie temperature $T_C \approx 600$ K [12,13] of the FM Co/Pt-multilayer is much higher than $T_N = 78$ K of the antiferromagnet FeF_2. Hence, the Co/Pt-multilayer is magnetically ordered at all temperatures $T < T_C$. In the case of a uniaxial ferromagnet, at $T \ll T_C$, the net magnetic moment is determined by its domain structure and can be expressed by the number of up and down magnetic moments. Hence, the total magnetic moment of the FM interface layer reads

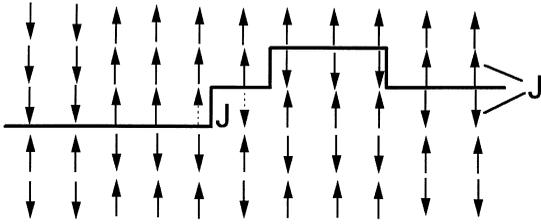$$M_F = (n_F^+ - n_F^-)m_F = (2n_F^+ - n_0)m_F, \qquad (1)$$

Fig. 3. Schematic spin structure at the FM/AF-interface (line) and its adjacent layers after cooling to below T_N in an axial freezing field $0 < H < H_S$. Arrows indicate the magnetic moments of the FM (above the interface) and AF system (below the interface), respectively. At the interface adjacent spins are coupled by the AF exchange constant J along the spin direction (solid arrows) and, in addition, by J' (dashed arrows) at the steps of the interface.

where m_F is the magnetic moment per atom and $n_0 = n_F^+ + n_F^-$ is the total number of FM interfacial up and down moments, $n_F^\pm$. In order to model the field dependence of the magnetization of the ferromagnet in the absence of exchange bias, i.e. at $T_N < T < T_C$, we make the simple linear ansatz

$$n_F^+(H) = \begin{cases} 0 & \text{for } H < -H_S, \\ (n_0/2)(1 + H/H_S) & \text{for } -H_S < H < H_S, \\ n_0 & \text{for } H > H_S, \end{cases}$$
(2)

where H_S is the saturation field value. Note, that the exchange bias of the magnetization reversal curve does not depend on the width of the hysteresis loop. Therefore, we completely neglect hysteresis for sake of simplicity and assume homogeneous domain nucleation and growth behavior. A typical situation encountered for $0 < H < H_S$ is depicted in Fig. 3 (above the interface). Since the magnetization reversal is strongly supported by the perpendicular anisotropy of the Co/Pt multilayer system [12,13] saturation is achieved at fairly low values of H_S. These are definitely lower than $H_S = M_S$ (in SI units), which would be expected for a FM thin film with mere shape anisotropy when exposed to a perpendicular magnetic field. Owing to the curvature of the experimental M vs. H curves (Figs. 1a–c) it seems reasonable to treat H_S as a fitting parameter.

Moreover, we assume that the spin structure of the ferromagnet which establishes on applying $H = H_F$ at $T > T_N$ is not affected by the ordering process of the exchange coupled antiferromagnet on cooling to below T_N, although the unidirectional anisotropy originates from this coupling. Hence, the number of up spins n_F^+ as given by Eq. (2) does not change on cooling towards T_N and below. Under this constraint it is now possible to calculate the spin structure and magnetization of the interface layer of the antiferromagnet within the framework of equilibrium thermodynamics. To this end we write down the partition function by taking into account the four possible configurations of the AF/FM spin pairs at the interface. They originate from the combinations of the spin values $S_F = \pm 1$ and $S_{AF} = \pm 1$, respectively. The energy function takes into account the exchange and the Zeeman energies. The latter one affects only S_{AF}, because the orientation of S_F is assumed to be independent of temperature at $T \ll T_C$ for a given freezing field. Hence, no thermodynamic consideration of S_F is necessary. Moreover, we assume that on cooling the ordering of the spins in the AF system starts at the AF/FM interface at $T = T_N + \delta T$, where $0 < \delta T \ll T_N$. This behavior is reasonable, because similar proximity effects have been observed on EuS precipitated in Co [21]. Therefore, exchange coupling between S_F and S_{AF} at the interface is taken into account, but neglected between S_{AF} and its AF bulk neighbors. Their ordering requires further cooling to $T \ll T_N$.

Once the spin structure at the interface is stabilized, the underlying antiferromagnet develops its domain structure from the interface into the bulk of the crystal. This final domain structure of the AF gives rise to the unidirectional anisotropy, which characterizes the exchange bias. Within these approximations the energy function, which controls the spin arrangement of the interface, reads

$$E = -JS_F S_{AF} - g\mu_B \mu_0 H S_{AF}.$$
(3)

For a given orientation of S_F there are two different states $S_{AF} = \pm 1$. This yields two partition functions $Z^\pm$ for $S_F = \pm 1$, respectively. They read

$$Z^\pm = 2\cosh((\pm J + g\mu_B \mu_0 H)/k_B T)$$
(4)

and allow to calculate the thermally averaged magnetizations

$$m^\pm = \frac{1}{\mu_0} \frac{\partial}{\partial H} k_B T \ln Z^\pm$$

$$= g\mu_B \tanh((\pm J + g\mu_B \mu_0 H)/k_B T). \quad (5)$$

Up to now no interface roughness has been taken into account. Let us now consider steps at the interface, which give rise to a new kind of interlayer exchange coupling J' between S_F and S_{AF} (Fig. 3, broken arrows). Since the steps may be regarded as (1 0 0) or (1 1 0) planes, which give rise to strongly enhanced exchange bias (e.g. $\mu_0 H_E \approx 50$ mT for $FeF_2(1\,0\,0)/Fe$ [2]) we anticipate $|J'| \gg |J|$. Hence, those spins S_{AF} which are coupled via J' to S_F do not participate in the above thermodynamic consideration, but are rigidly coupled to their neighboring FM spin.

Let $n_S^\pm$ be the number of spins S_F located at steps and $\tilde{n}_F^\pm$ the remaining spins on the terraces. The magnetic moment M_{AF} of the AF interface layer (Fig. 3, solid line) then reads

$$M_{AF} = \tilde{n}_F^+ m^+ + \frac{J'}{|J'|} n_S^+ m_0 + \tilde{n}_F^- m^- - \frac{J'}{|J'|} n_S^- m_0, \quad (6)$$

where m_0 is the absolute value of the magnetic moment of the spin in the AF system. With $n_F^\pm = \tilde{n}_F^\pm + n_S^\pm$, $n_0 = n_F^+ + n_F^-$ and the topographic roughness parameter $\alpha = n_S^\pm/n_F^\pm$, one obtains

$$M_{AF} = n_F^+(1-\alpha)[m^+ - m^-] + n_0(1-\alpha)m^-$$

$$+ \frac{J'}{|J'|} \alpha m_0(2n_F^+ - n_0) \quad (7)$$

with $0 \leqslant \alpha \leqslant 1$. This magnetization of the AF topmost layer is assumed to determine the domain structure of the bulk AF substrate (Fig. 3, below the interface) and to remain invariant both on cooling to $T \ll T_N$ and upon cycling between positive and negative saturation of the FM subsystem.

From Stoner–Wohlfarth model considerations [22] the exchange bias field H_E is known to be

$$H_E \propto [(1-\alpha)J + \alpha J'](\underline{M}_F \cdot \underline{M}_{AF}). \quad (8)$$

In the case of an Ising-system we replace $\underline{M}_{AF}$ and $\underline{M}_F$ by the scalar thermal equilibrium values. This yields

$$H_E \propto [(1-\alpha)J + \alpha J'](M_F M_{AF}). \quad (9)$$

With $\Delta m \equiv m^+ - m^-$ and $M_F = (2n_F^+ - n_0)m_F$ one finally obtains

$$H_E \propto J[(1-\alpha) + \alpha J'/J](2n_F^+ - n_0)m_F$$

$$\times (n_F^+(1-\alpha)\Delta m + n_0(1-\alpha)m^-$$

$$+ \frac{J'}{|J'|} \alpha m_0(2n_F^+ - n_0)). \quad (10)$$

Expression (10) is now best fitted to the H_E vs. H_F data inserting Eq. (2) with $H = H_F$ and Eq. (5) by letting $T = T_N = 78$ K. Here we assume that the spin structure at the interface, which establishes a $T = T_N$, will not change when cooling down to the measurement temperature, $T = 10$ K. We neglect, hence, all interactions both within the AF interface layer and between this layer and the ordered bulk. It will be left for the future to investigate the observed [23] subtle temperature dependence of the exchange bias field, H_E, within mean field approach. $J/k_B T_N$, H_S, α, $J'/|J'|$ and $g\mu_B/k_B T_N$ enter (10) as fitting parameters, while n_0, m_F and $J[(1-\alpha) + \alpha J'/J]$ are absorbed into the proportionality constant which transforms (10) into an equation. A constant offset $\mu_0 H_E = 1.4$ mT is added during the fitting procedure in order to describe the background of the H_E vs. H_F curve as defined by the data point at $H_F = 0$. It is considered as a small, hitherto unexplained systematic error in our procedure to determine H_E (Fig. 1). The result of the best fit is shown in Fig. 2 (solid line).

The data are well described within their error bars. As expected the microscopic exchange parameter $J/k_B T_N = -0.46$ is negative. Hence, AF coupling at the interface is favored. The maximum of the H_E vs. H_F curve originates from the competition between this AF exchange and the Zeeman energy. Moreover, $\mu_0 H_S = 0.12$ T is in acceptable agreement with the saturation fields revealed by the

hysteresis loops of Fig. 1, where about 80% of M_S is achieved at $\mu_0 H_S = 0.12$ T. The steep increase of H_E vs. H_F in the weak field limit, $H_F < H_S$, corroborates the rapid saturation of the FM overlayer, but unfortunately renders difficult, but not impossible [23] the measurement of additional data points in the interval $0 < H_F < H_S$. The subsequent decrease of H_E is understood by the weakening of the AF ordering owing to dominance of the Zeeman energy after reaching full FM saturation. The best-fitted value $\alpha = 0.2$ indicates a rough interface, where 20% of the FM spins are located at step positions. This roughness very probably originates from the mechanical polishing procedure of the (0 0 1)-surface of FeF_2. Note, that the proportionality (10) opens the possibility of positive exchange bias by reduction of α towards zero, which has the physical meaning of a perfect interface. Then expression (10) yields

$$H_E \propto J(2n_F^+ - n_0)m_F(n_F^+ \Delta m + n_0 m^-), \qquad (11)$$

which gives a continuous crossover from negative to positive exchange bias upon increasing the freezing field H_F. The proportionality constant which contains the product of $J[(1 - \alpha) + \alpha J'/J]$, n_0 and the magnetic moment of the Co atoms, m_F, reads $P = 0.11$ mT. Further, also the coupling constant J' turns out to be negative, since $J'/|J'| = -1$ is required to obtain the appropriate shape of the fitting function. The best-fitted value $g\mu_B/k_B T_N = 1.24$ T^{-1} is by a factor of 65 higher than expected from the single magnetic moment, $g\mu_B$, of Fe^{2+} in bulk FeF_2 letting $g = g_\parallel = 2.2$ [24]. This discrepancy is probably due to our neglect of any correlation between the AF interface and bulk spins. In a first approximation the above number corresponds to a cluster size of about four-nearest-neighbor distances, which appears reasonable for an antiferromagnet just above T_N. These clusters effectively enhance the field contribution which enters the weight factor $\tanh((\pm J + g\mu_B \mu_0 H)/k_B T_N)$ in Eq. (5). They thus determine the magnetic structure of the interface, which will not change upon cooling. Measurements at $T = 10$ K are, hence, able to provide information on the spin ordering taking place at T_N. Obviously, in future more exact treatments also the criticality of the AF system has to be accounted for.

5. Conclusion

We measured the freezing field dependence of the exchange bias of the $FeF_2(0\ 0\ 1)/(Co3.5$ Å/ Pt12 Å)$_3$/Pt8 Å heterosystem with uniaxial perpendicular anisotropy. The experimental H_E vs. H_F data are well described within the framework of an Ising-model. It takes into account the exchange and the Zeeman energy of the interface spins as well as the interface roughness. Our data are modeled by a fitting function which involves the roughness parameter $\alpha = 0.2$. It turns out that the shape of the fitting curve depends very sensitively on α. In the limit $\alpha \rightarrow 0$, even positive exchange bias fields H_E can be modeled, which were observed previously in planar FeF_2/Fe-systems [7]. The prominent maximum of the H_E vs. H_F curve originates from the competition between the AF interface coupling and the Zeeman energy, which favors the alignment of all moments parallel to the axial freezing field.

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