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Dilution-induced enhancement of the blocking temperature in exchange-bias heterosystems

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The temperature dependence of the exchange bias field is investigated by superconducting quantum interference device magnetometry in $Fe_{1-x}Zn_xF_2$(110)/Fe14 nm/Ag35 nm, $x=0.4$. Its blocking temperature exhibits a significant enhancement with respect to the global ordering temperature $T_N=46.9$ K, of the bulk antiferromagnet $Fe_{0.6}Zn_{0.4}F_2$. The enhancement is attributed to fluctuations of the diamagnetic dilution which creates clusters on all length scales having a Zn dilution of $0\leqslant x\leqslant 1$. While the infinite clusters give rise to the well-known Griffiths phase, finite clusters also provoke a local enhancement of the exchange bias. The temperature dependence of the integral exchange bias effect is modeled by averaging all local contributions of the antiferromagnetic surface magnetization which exhibit a surface critical behavior.

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Recently, the exchange bias (EB) effect has attracted a great deal of interest in basic research and materials science,[1,2] owing to its relevance for technological applications. The EB effect describes a shift of the ferromagnetic hysteresis loop along the magnetic-field axis away from its origin at $H=0$, which is caused by the exchange coupling at the interface between ferromagnetic (FM) and antiferromagnetic (AF) materials after cooling or growth of the sample in a magnetic field to below the Néel temperature of the antiferromagnet.

Meiklejohn and Bean proposed a simple model of the EB effect which provides the basic, but powerful formula $H_{EB}=-J_{EB}S_{AF}S_{FM}/(M_{FM}t_{FM})$ for the exchange bias field.[3,4] Here S_{AF} and S_{FM} are the AF and FM spins at the interface, M_{FM} and t_{FM} are the magnetization and the thickness of the FM layer, while J_{EB} is the interface coupling constant. The above formula suggests that the exchange bias effect vanishes at the Néel temperature T_N of the bulk antiferromagnet. Note that, in general, the Curie temperature of the ferromagnet is much higher than T_N, and has no impact on the temperature dependence of H_{EB}. A rare exception was reported in Refs. 5 and 6, where FM/AF bilayers with comparable critical temperatures T_c and T_N exhibit an enhanced exchange bias field on approaching $T_c\gtrsim T_N$. Typically, the blocking temperature T_B, i.e., the temperature of the vanishing exchange bias effect, is smaller than T_N.[1] This behavior is usually attributed to finite-size effects which are known, e.g., from the dependence of the EB effect on the AF layer thickness.[4,7,8] Moreover Zaag *et al.*[9] pointed out that T_B does not track the ordering temperature in a simple manner. The authors of Ref. 10 showed that in FeF_2-Fe bilayers near T_N the temperature dependence of the exchange bias field follows the temperature dependence of the AF interface magnetic moment, which exhibits a power-law behavior owing to surface criticality. In the case of large grain sizes ξ_g, the corresponding critical exponent $\beta_s=0.8\pm0.04$ agrees with the surface critical exponent of the three-dimensional Ising system. With decreasing grain sizes ξ_g, however, the corresponding decrease of β_s was attributed[10] to either a finite-size effect, in accordance with an effective decrease in the lateral terrace size, or to an increase of the interface interaction. The latter gives rise to surface ordering which occurs independently of the bulk. Moreover, an increased interface interaction may explain a blocking temperature slightly above T_N.

In this paper, we investigate the temperature dependence of the exchange bias field H_{EB} in the $Fe_{0.6}Zn_{0.4}F_2$(110)/Fe(14 nm)/Ag(35 nm) heterostructure by superconducting quantum interference device (SQUID, Quantum Design MPMS-5S) magnetometry. Surprisingly, we find that the blocking temperature $T_B\approx 63$ K is much higher than the Néel temperature $T_N=46.9$ K of the diluted bulk antiferromagnet $Fe_{0.6}Zn_{0.4}F_2$. Such a huge enhancement of T_B with respect to T_N cannot be explained within the framework of a small proximity effect that originates from the mutual AF/FM interaction at the interface. Instead we propose that $Fe_{1-x}Zn_xF_2$ clusters, with an arbitrary concentration of ZnF_2, $0\leqslant x\leqslant 1$, originate from fluctuations of the diamagnetic dilution occurring during the natural growth of the AF bulk crystal. They appear on all length scales, where infinite clusters give rise to the well-known Griffiths phase where nonanalyticity at $T_N\leqslant T\leqslant T_N(x=0)=78.4$ K was predicted as a rigorous theoretical result.[11] However, finite clusters, which are already far more frequent than exponentially rare clusters of infinite size, are expected to provoke a local enhancement of the exchange bias. Here we present an analysis of the temperature dependence of the integral exchange-bias effect. To this end the local contributions to the AF surface magnetization, which exhibit critical behavior are averaged with respect to a phenomenological cluster distribution under the assumption of local quasicritical temperatures.

The $Fe_{0.6}Zn_{0.4}F_2$(110)/Fe(14 nm)/Ag(35 nm) sample is grown at $T=425$ K under ultra high-vacuum (UHV) conditions by depositing 14-nm Fe on top of the compensated (110) surface of the diamagnetically diluted AF substrate. The metallic thin film is deposited on an $Fe_{0.6}Zn_{0.4}F_2$ single crystal which has been polished to optical flatness with 0.3-μm diamond paste before transferring into the UHV chamber. Atomic force microscopy investigations of polished (001) surfaces of pure FeF_2 single crystals exhibit rough surfaces of such AF insulating substrates, with a corrugation of 0.8 nm on a local scale up to 0.3 μm while a full line scan of 10-μm length contains contributions of deep and rare scratches which give rise to a corrugation of 4.8 nm. After cooling in a field of $\mu_0H_{fc}=5$ mT applied parallel to the

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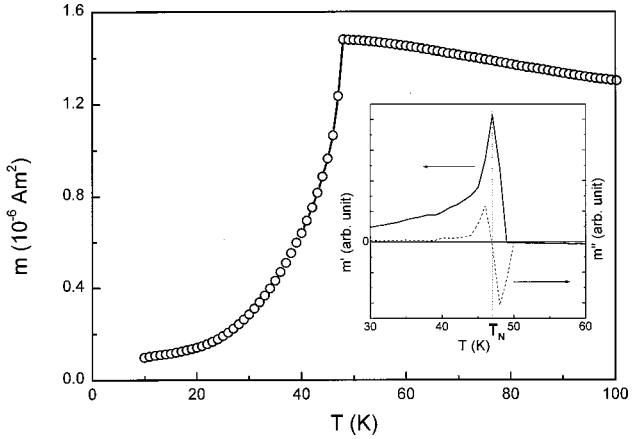

FIG. 1. Temperature dependence of the magnetic moment m of $Fe_{0.6}Zn_{0.4}F_2(110)$/Fe(14 nm)/Ag(35 nm). The inset shows the first (solid line) and second (broken line) derivatives with respect to temperature, thus defining the Néel temperature of the $Fe_{0.6}Zn_{0.4}F_2(110)$ substrate, $T_N = 46.9$ K.

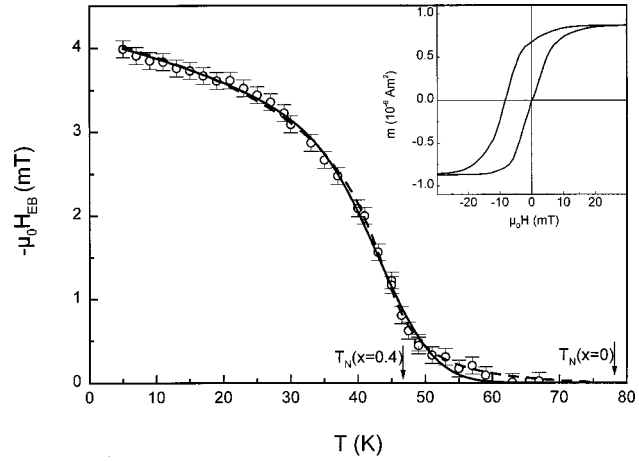

FIG. 2. Temperature dependence of the exchange bias field $\mu_0 H_{EB}$ (circles) obtained from magnetic hysteresis curves at the respective target temperatures $5\,K \leqslant T \leqslant 80\,K$ after cooling in a freezing field of 5 mT. The inset shows a typical hysteresis curve measured at $T = 5$ K. The solid and dashed lines show the results of the best fits of Eq. (1) involving Gaussian and Lorentzian distribution functions, respectively. Arrows at 46.9 and 78.4 K indicate the Néel temperatures of the diluted and the pure antiferromagnet, respectively.

planar [110] direction from $T = 100$ K to the respective target temperature, the exchange-bias field is determined from the shift of the hysteresis loop.

Figure 1 shows the temperature dependence of the magnetic moment m of the heterostructure. The Curie temperature of Fe is much higher than the global T_N of $Fe_{0.6}Zn_{0.4}F_2$. Hence the magnetic moment of Fe remains almost constant at all temperatures under investigation, and the m vs T dependence mainly reflects the temperature dependence of the parallel magnetic susceptibility of the antiferromagnet. However, the steep increase in the vicinity of T_N, on the one hand, and the delayed decay of the magnetic moment above T_N, on the other hand, very probably originate from the residual interface interaction. The inset of Fig. 1 exhibits the first and second derivatives of $m(T)$, which indicate the Néel temperature $T_N = 46.9$ K of the $Fe_{0.6}Zn_{0.4}F_2$ substrate. In contrast with this global AF critical temperature, however, the H_{EB} vs T dependence in Fig. 2 (open circles) shows a long tail above T_N with a blocking temperature $T_B \approx 63$ K.

The authors of Ref. 10 pointed out that in a FeF_2-Fe system the temperature dependence of H_{EB} should be directly proportional to the AF surface order parameter. They implicitly made the plausible assumption that the magnetic moment S_{AF} possesses the T dependence of the AF surface order parameter. In order to take into account the influence exerted by perturbations such as like strain and disorder on the EB effect, they introduced the "rounded" power law

$$H_{EB}(T) = H_{EB0} \int_0^\infty t^{\beta_s} P(T_C)\, dT_C, \tag{1}$$

where $t = 1 - T/T_C$ is the reduced temperature for $T < T_C$, and $t = 0$ for $T > T_C$, while $P(T_C)$ is the critical temperature distribution. A narrow Gaussian distribution function

$$P(T_C) = \frac{1}{\delta\sqrt{2\pi}} \exp[-(T_C - T_{C0})^2/2\delta^2] \tag{2}$$

has been used for a best fit of the H_{EB} vs T data, where H_{EB0}, T_{C0}, β_s, and δ are the fitting parameters.

In contrast with the assumption of weak perturbations in FeF_2, however, we stress the fact that in our case the diamagnetic dilution is a strong and intrinsic perturbation. It gives rise to a broad distribution function even in the case of an ideal antiferromagnet with random site dilution.

Previously, we showed that the dilution-induced Griffiths phase of the antiferromagnet $Fe_{1-x}Zn_xF_2$ gives rise to significant deviations from the Curie-Weiss behavior which is naively expected at $T > T_N$.[12] However, Griffiths-type clusters give rise to a continuous series of local phase transitions with quasicritical temperatures T_c within $T_N(x) \leqslant T_c \leqslant T_N(x=0)$.

In view of this experience, we fit Eq. (1) to the experimental data of Fig. 2 (circles) while alternatively using two phenomenological distribution functions, viz., the Gaussian distribution of Eq. (2) and the Lorentzian distribution

$$P(T_C) = \begin{cases} \dfrac{\varepsilon/2}{[\varepsilon^2 + (T_C - T_{C0})^2]\arctan\{[T_N(0) - T_{C0}]/\varepsilon\}} & \text{if } T_c \leqslant T_N(x=0) \\ 0 & \text{if } T_c > T_N(x=0) \end{cases}. \tag{3}$$

Here $T_N(0) = 78.4$ K is the Néel temperature of the pure FeF_2 while H_{EB0}, T_{C0}, β_s, δ, and ε are the fitting parameters. The Lorentzian distribution function was used previously in order to model the T_c distribution involved in the analogous phenomenon of a field-induced Griffiths phase.[13] It is normalized under the constraint $\int_{Tc0}^{T_N(0)} P(T_c)\, dT_c = \frac{1}{2}$, where a symmetric distribution with respect to T_{C0} is assumed. T_{C0} describes the centers of gravity of the distribu-

TABLE I. List of the parameters resulting from the best fits of Eq. (1) to the $\mu_0 H_{\mathrm{EB}}$ vs T data (Fig. 2, circles) involving Gaussian and Lorentzian distribution functions, respectively.

	$\mu_0 H_{EB0}$ (mT)	T_{C0} (K)	β_s	δ or ε (K)
Gaussian distribution	4.1 ± 0.1	45.0 ± 1.5	0.22 ± 0.04	6.1 ± 1.6
Lorentzian distribution	4.1 ± 0.1	44.4 ± 1.3	0.19 ± 0.03	4.0 ± 0.1

tion functions, respectively, and is expected to be in the vicinity of the global transition temperature $T_N(x)=46.9\,\mathrm{K}$.

Figure 2 shows the results of the best fits of Eq. (1) involving a Lorentzian (dashed line) and a Gaussian distribution (solid line), respectively. Inspection shows that, in particular, the tail of the H_{EB} vs T data at $T>50\,\mathrm{K}$ is modeled more accurately by the Lorentzian distribution. This is corroborated by the resulting fitting parameters which profit from smaller uncertainties in comparison with the parameters which result from the Gaussian distribution. The resulting fitting parameters are listed in Table I. Here we are going to discuss the parameters. Substitution of $t=1$ into Eq. (1) yields the physical meaning of $\mu_0 H_{\mathrm{EB0}}$, which is the zero-temperature limit of the exchange-bias field. Hence $\mu_0 H_{\mathrm{EB0}} \approx 4\,\mathrm{mT}$ is in accordance with the low-temperature value of the $\mu_0 H_{\mathrm{EB0}}$ vs T data shown in Fig. 2. As expected, $T_{C0}=45$ and 44.4 K, respectively, are close to the global transition temperature $T_N(x=0.4)=46.9\,\mathrm{K}$. The parameters $\delta=6.1$ and $\varepsilon=4.0$ characterize the widths of the Gaussian and Lorentzian distribution functions, respectively. Physically the most interesting parameters are the surface critical exponents $\beta_s=0.22$ and 0.19 for the Gaussian and Lorentzian distributions, respectively. These values are surprisingly low in comparison to $\beta_s=0.8$, the surface critical exponent of the three-dimensional Ising system found in the FeF_2-Fe bilayer system.[10] However, it is known that surface critical behavior depends on the ratio between the surface coupling J_s and the bulk coupling constant J_b, $r=J_s/J_b$.[10,14,15] The phase diagram of the semi-infinite Ising model exhibits a critical ratio at $r_c\approx1.50$.[14,16] In the case $r<r_c$ an "ordinary transition" occurs, which depends on the bulk critical behavior. At $r>r_c$ a surface transition occurs which is independent of the bulk transition, while a so-called "special transition" occurs at the multicritical point r_c.[15]

In our case, β_s is quite close to the value of the special transition where $\beta_s=0.237$ and 0.26 were reported in Refs. 16 and 17, respectively. As pointed out in the work of Ref. 10, β_s decreases with increasing interface exchange coupling because the surface spins tend to order independently of the bulk, i.e., with an exponent closer to the two-dimensional Ising value $\beta_s=\frac{1}{8}$.[18] In the case of the diluted antiferromagnet this tendency may be enhanced, because the diamagnetic dilution gives rise to broken magnetic "bonds." Hence the interaction of an AF surface moment with neighbors from the AF bulk is effectively reduced with respect to the exchange interaction between the AF and FM moments at the interface. Therefore the ratio of the effective surface and bulk interaction constants increases thus giving rise to a reduced β_s.

In conclusion, we have investigated the temperature dependence of exchange-bias field in $Fe_{1-x}Zn_xF_2(110)$/Fe14 nm/Ag35 nm, $x=0.4$. The blocking temperature is located more than 16 K above the Néel temperature of the diluted bulk antiferromagnet. The unusual enhancement of the blocking temperature is attributed to a wide distribution of quasicritical temperatures which correspond to clusters of various diamagnetic dilution $0\leqslant x\leqslant1$. The clusters originate from fluctuations of the dilution which are involved in the natural growth process. Clusters of infinite size (i.e., on the length scale of the sample) give rise to the well-known Griffiths phase. However, an enhancement of the blocking temperature requires only very frequently occurring clusters of finite size. Within the framework of this cluster model, the experimental data are fitted on averaging all local components which possess surface critical behavior. The resulting critical exponent $\beta_s\approx0.2$ is close to the value predicted for the so-called special transition.

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