

Nonlinear and hysteretic exchange bias in antiferromagnetically coupled ferromagnetic bilayers

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We report observations of nonlinear and hysteretic exchange bias in antiferromagnetically coupled hard/soft ferromagnetic bilayers. In such systems, the hard ferromagnetic layer acts as a tunable biasing layer replacing the antiferromagnet used in conventional exchange bias structures. It is shown that the reported behavior is due to the reciprocity of the exchange bias effect in conjunction with the presence of locally varying interfacial exchange which causes a preferential magnetization of the weakly exchange-coupled tuning regions during the exchange bias setting stage.

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The exchange bias effect refers to a hysteresis loop shift resulting from exchange coupling a ferromagnet (FM) to an antiferromagnet (AF).¹ More recently, it has been shown that related phenomena are also observed in coupling two ferromagnets with dissimilar magnetic properties.^{2–4} In the case of the FM/AF structures, the effect is typically controlled in an experiment by applying a magnetic field while cooling through the Néel temperature of the AF. In the case of FM/FM structures, this role of the AF is fulfilled by a hard FM layer and exchange bias setting is achieved by applying sufficiently large magnetic fields without the need for thermal preprocessing. The analogy between FM/AF and FM/FM systems is that in both cases the exchange bias is due to the exchange coupling between the layers and the net interfacial spin moments which are not reversed during the measurements of the hysteresis loop shift.^{5,6}

The continual interest in studying the exchange bias phenomenon is not only related to its practical importance in spintronics,⁷ but is also due to being one of the challenging problems in low dimensional magnetism.⁸ Recently, the research focus has been oriented toward the rich variety of related phenomena observed in exchange biased FM/AF systems, such as positive exchange bias,⁹ coercivity enhancement,¹⁰ asymmetrical hysteresis loop shapes,^{11,12} training effects,^{8,13} and thermally induced anomalous spontaneous magnetization reversal.¹⁴ However, a complete and quantitatively reliable understanding of exchange bias magnetic systems is an outstanding problem. This lack of understanding is primarily related to the fact that the defining physical phenomena are governed by the magnetic spin structures at the interface and within the AF layer, which are very difficult to access experimentally.¹⁵ Therefore, the use of FM/FM bilayers as model systems has been important in understanding core issues related to exchange bias such as the possible origins of the hysteresis loop asymmetry,¹² training effects and their correlation with the metastable nature of the tuning layer,^{16,17} and the effect of lateral magnetic domains on the exchange bias.⁶ The main advantage here is that the net interfacial moment can be quantified with very high reliability by a simple magnetization measurement of the ultrathin FM tuning layer.

In this paper, we demonstrate that the existence of a re-

ciprocal exchange bias effect can modify magnetization processes of the biasing layer during its setting stage (i.e., during the field cooling in the FM/AF case or the hard FM layer magnetization in the FM/FM case). This subtle effect can produce significant nonlinearities and even a hysteretic exchange bias behavior. We show that the essential ingredient is the presence of a laterally varying interfacial exchange coupling and that the effect disappears in the uniform case. Our experimental findings are confirmed by the complementary modeling of such FM/FM exchange bias systems.

The bilayer structures used in our experiment consist of a 15 nm thick CoPtCrB film, which is the adjustable hard-magnetic layer (HL), exchange coupled by means of a 6 Å Ru interlayer to a 1–2 nm thick CoCr film, which is the soft-magnetic layer (SL). All samples were prepared by magnetron sputter deposition onto ultrasmooth glass substrates. The magnetic multilayer structure was hereby grown on top of a suitable underlayer sequence to allow for a desirable grain-size distribution and orientation as well as for sufficient film adhesion. The structures were also covered with an overcoat layer for corrosion protection. In our samples, the thickness of the Ru spacer is adjusted to produce antiferromagnetic coupling.^{3,4,18} A schematic of this structure can be seen in Fig. 1(a), which also displays two examples of experimentally measured SL-hysteresis loops. All magnetization measurements were performed by means of an alternating-gradient magnetometer (AGM). The shift of the loop from the origin at zero field, denoted as h_{bias} and M_r , corresponds to the exchange bias on the SL and the remanent magnetization of the HL, respectively. As was previously demonstrated, the individual SL-hysteresis loops in Fig. 1(a) are closed,¹⁹ which verifies that no alteration of the HL magnetization is produced by the applied field due to the clearly separated switching field distributions of the two individual FM layers.^{3,4} In addition, training effects for our samples are small¹⁷ and their effect on the results of the present paper can be neglected. This allows independent control of both layers, where an arbitrary magnetization state M_r of the HL can be set at high magnetic fields in a first step, after which the bias field h_{bias} can be determined from the low-field SL-hysteresis loop without perturbing M_r . We will investigate the behavior of h_{bias} vs M_r obtained by using the following experimental

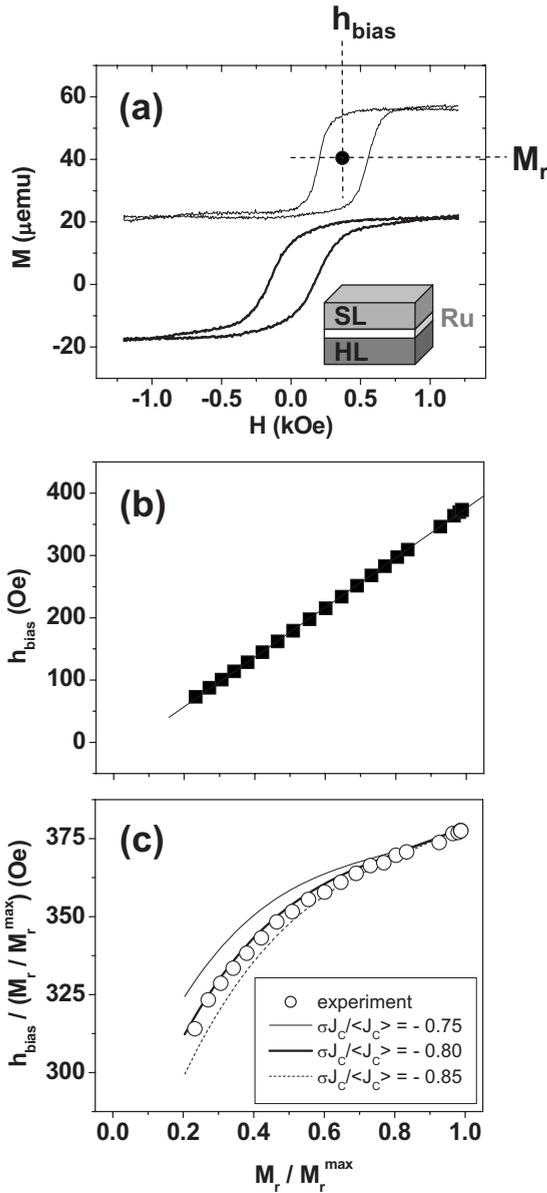


FIG. 1. (a) Low-field hysteresis loops of a 1.5 nm thick SL for two different magnetization states of the adjacent 15 nm HL film. The thick solid line shows the resulting SL-hysteresis loop for the ac-demagnetized state of the HL, while the thin solid line shows the case of positive saturation for the HL. The inset is a schematic of the sample structure. (b) Experimental data for the exchange bias field h_{bias} , as a function of M_r/M_r^{max} after applying different remagnetization steps to the HL. (c) Experimental data shown in (b), but with h_{bias} being normalized by the HL magnetization M_r/M_r^{max} . The lines correspond to model calculations for three different values of the interlayer coupling distribution width $\sigma J_c / \langle J_c \rangle$.

protocol:^{3,4} (i) The entire sample is at first demagnetized by applying an alternating field starting with a 10 kOe amplitude and decreasing it by 0.1% per cycle. This ac-demagnetization procedure was found to consistently produce final states with HL magnetization values of less than 5% of the remanent magnetization obtained after HL saturation. (ii) The sample is partially remagnetized by applying a set field, H_{set} , in the range from 0 to 10 kOe. (iii) After the

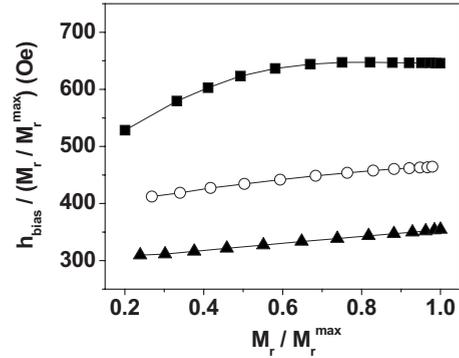


FIG. 2. Experimental data for the exchange bias field h_{bias} , normalized by the HL magnetization M_r/M_r^{max} , as a function of M_r/M_r^{max} for three different samples with varying SL-layer thickness: (▲) 1.5 nm, (○) 1.3 nm, (■) 1.1 nm CoCr film.

preconditioning steps (i) and (ii), a low-field hysteresis loop of the SL is measured using a maximum field of $H_m = \pm 1.2$ kOe, such that it is insufficient to produce switching of the HL. Steps (ii) and (iii) are then repeated for increasing values of H_{set} , producing different HL magnetization states M_r , and in turn altering the exchange bias h_{bias} of the adjacent SL.

A typical h_{bias} vs M_r curve is shown in Fig. 1(b). While the observed h_{bias} vs M_r relationship appears to be linear, a detailed data analysis reveals a more complex dependency. Indeed, as shown in Fig. 1(c), if the ratio h_{bias}/M_r is plotted, one observes a well-pronounced reduction for small M_r values and not a constant which would be expected for the truly linear case. This finding is a general observation as demonstrated in Fig. 2 for three other samples analyzed in the same manner. The large differences between the magnitudes of the exchange bias effect for the three curves are due to different thicknesses of the SLs and disappear (almost completely) after normalizing by the corresponding absolute magnetization of the SL.^{3,4} Thus, Fig. 2 shows that although the functional form does not appear to be unique but is rather sample dependent, the ratio h_{bias}/M_r is reduced for small M_r values in all cases.

Our experimental observation can be understood by assuming that the HL magnetization is not independent from the exchange interaction with the SL, but that there exists a “reciprocal” effect onto the HL, which affects the HL magnetization reversal during the setting stage, i.e., step (ii).²⁰ Therefore, the local regions of the HL, which produce a weaker exchange bias, would be preferentially magnetized during the initial stages in our experimental procedure and thus lead to the reduction of h_{bias}/M_r for smaller M_r values. On the other hand, the regions producing stronger exchange bias would be magnetized later at higher fields, increasing the h_{bias}/M_r ratio for high M_r values. A mechanism that would correlate preferential HL switching with the reduced SL exchange is easily identified because only the interlayer exchange coupling connects both aspects directly in our simple bilayer system.²¹ The exchange coupling is not only directly proportional to the local exchange bias of the SL, but during the setting stage also transfers a corresponding exchange field from the fully saturated SL onto the HL and

modifies its magnetization reversal. Correspondingly, a variation of local exchange coupling values is essential if such a correlation of the HL magnetization state with the SL bias is to be observable. The exchange coupling distribution can be attributed to the locally varying thickness of the interlayer spacer combined with the well-known thickness dependence of the interlayer exchange coupling. This explanation is supported by the fact that for our samples, the Ru layer has been grown on a granular template which had local height variations of the order of 1 nm. However, we could not unambiguously identify the exact physical origin of the exchange coupling distribution because our transmission electron microscopy (TEM) and x-ray diffraction (XRD) studies could not distinguish between the interface intermixing and film thickness variations with the necessary precision.

To test the validity of the above physical picture, we consider a previously devised microscopic model of the AF-coupled structure.¹² Main elements of the model are summarized in Fig. 3. The sample is described by means of two two-dimensional layers of Ising spins, each representing, respectively, a HL or SL grain. Adjacent HL and SL grains are coupled by a locally varying exchange coupling constant, J_c , which follows a Gaussian distribution with variance σJ_c and mean $\langle J_c \rangle$. The HL grains are assumed to be noninteracting and described by individual switching fields H_s , given by a Gaussian distribution with variance σH_s and average $\langle H_s \rangle$. The intrinsic coercivity of our experimental SL grains is small and therefore neglected in the model to reduce the number of model parameters. However, the SL grains are exchange coupled to their next neighbors by means of a fixed intergranular coupling constant J_{gg} , which is responsible for the overall hysteresis behavior of the SL. Mimicking the previously specified experimental procedure (i)–(iii) for setting the exchange bias, we can generate h_{bias} vs M_r curves which reproduce our experiments very well, as shown by the thick line in Fig. 1(c). This suggests that our simple model indeed captures the essential physics behind the nonlinear h_{bias} vs M_r dependence. We also verified that all our results depended on the ratio $\sigma J_c / \sigma H_s$ rather than on σJ_c and σH_s separately. Therefore, this ratio will be used to quantify the width of the exchange coupling distribution in the following discussion.

To explore the existence of a correlation between the HL magnetization state and the reduced SL bias, we computed the probability distribution $D(M_{\text{loc}}, J_{\text{cloc}})$ defined as the probability to find a local HL segment with an average local magnetic moment M_{loc} and average local exchange coupling J_{cloc} .²² Results of our model calculations are shown in Figs. 3(b)–3(g). The distribution patterns in the left column, Figs. 3(b), 3(d), and 3(f), are for $\sigma J_c / \sigma H_s = 1.6$ while those in the right column, Figs. 3(c), 3(e), and 3(g), represent data for $\sigma J_c / \sigma H_s = 0.16$. The top row [Figs. 3(b) and 3(c)], middle row [Figs. 3(d) and 3(e)], and the bottom row [Figs. 3(f) and 3(g)] correspond, respectively, to the demagnetized state of the HL with $M_r / M_r^{\text{max}} = 0$, HL magnetized to $M_r / M_r^{\text{max}} = 0.3$, and to $M_r / M_r^{\text{max}} = 0.5$ (i.e., 0%, 30%, and 50% of the saturation magnetization). The patterns in Figs. 3(b) and 3(c) have approximately a circular shape, demonstrating that after demagnetizing the model sample the local magnetization state

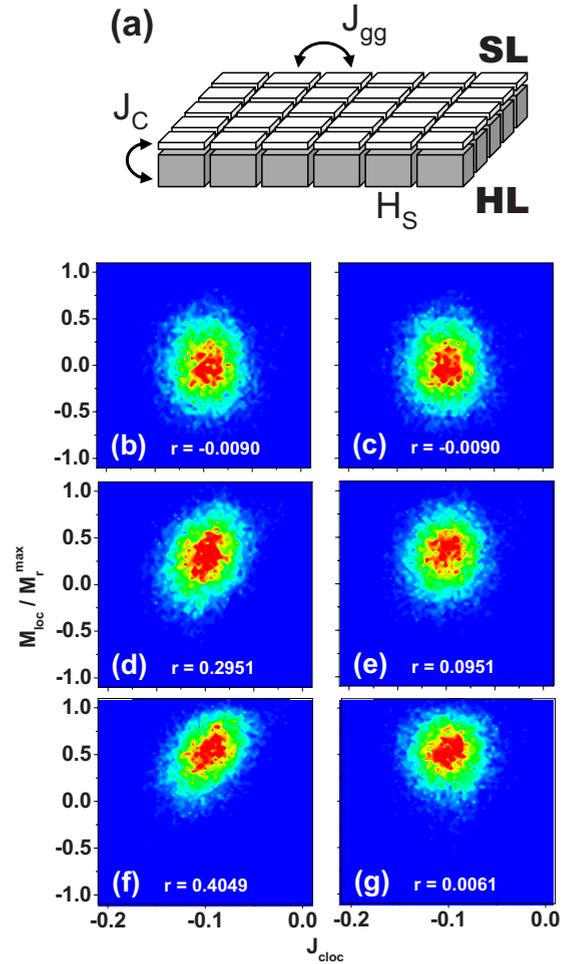


FIG. 3. (Color online) (a) Schematic of the bilayer model consisting of a SL and a HL. Both layers consist of vertically correlated grains, which are each represented by a single Ising spin. The layers are coupled with an antiferromagnetic coupling constant J_c , which varies from grain to grain following a Gaussian distribution of width σJ_c . The HL grains are independent from each other and characterized by a local switching field H_s , whose values follow a Gaussian distribution. The SL grains exhibit a next-neighbor exchange coupling of fixed strength J_{gg} . (b)–(g) Model calculation results. Characteristics of the HL-magnetic state, given by the probability distribution function $D(M_{\text{loc}}, J_{\text{cloc}})$, in which M_{loc} and J_{cloc} are averaged over a 5×5 grain area. (b), (d), and (f) are results for $\sigma J_c / \sigma H_0 = 1.6$, while (c), (e), and (g) are results for $\sigma J_c / \sigma H_0 = 0.16$. (b) and (c) correspond to random starting conditions, (d) and (e) to subsequent magnetization of the HL to $M_r / M_r^{\text{max}} = 0.3$, and (f) and (g) to subsequent magnetization of the HL to $M_r / M_r^{\text{max}} = 0.5$ in sufficiently high fields. The r values measure the correlation between M_{loc} and J_{cloc} in each HL-magnetic state.

M_{loc} and the coupling J_{cloc} remain uncorrelated. Moreover, these patterns have a similar shape, showing that the absence of correlations in the demagnetized state is independent of the exchange coupling distribution width. This is also verified by the small numerical values for the linear correlation coefficient r . Upon magnetizing the HL to 30% and 50% of the saturation magnetization, the $D(M_{\text{loc}}, J_{\text{cloc}})$ distribution patterns exhibit a tilt, which is a sign of developing correlations between M_{loc} and J_{cloc} . While being negligible for small

$\sigma J_c / \sigma H_s$, quite significant tilting and r values exist for a large $\sigma J_c / \sigma H_s$ ratio. Then, since for tilted distribution patterns shown in Fig. 3(d) or 3(f) smaller absolute J_{cloc} values coincide with the higher M_{loc} than they would if no tilting was present, the weakly coupled grains are likely to become preferentially magnetized earlier in the HL magnetization cycle. This is expected, of course, because the HL moments subject to a smaller exchange coupling J_{cloc} must overcome only weaker exchange bias fields to switch along the external field direction. Since the local exchange bias field onto the SL, h_{biasloc} , is proportional to J_{cloc} , these particular HL grains also produce only a diminished contribution to the overall h_{bias} on the SL. Therefore, due to the statistical dependence of J_{cloc} and M_{loc} variables, the simple linearity relationship between h_{bias} and M_r does not necessarily hold but needs to be replaced by

$$h_{\text{bias}} = \langle h_{\text{biasloc}} \rangle = \langle J_{\text{cloc}} M_{\text{loc}} \rangle \leq \langle J_{\text{cloc}} \rangle \langle M_{\text{loc}} \rangle = \langle J_c \rangle M_r. \quad (1)$$

Relation (1) verifies our assumption that for intermediate HL magnetization states, the presence of a nonvanishing distribution of interface exchange couplings will result in a reduction of the SL exchange bias below the value that would correspond to a volume-averaged coupling strength, a fact that will also be applicable to equally simple models of exchange bias in conventional FM/AM systems.⁸ Upon fully saturating, this reduction vanishes because all HL grains become magnetized independently from their J_{cloc} values and all correlations become effectively erased; hence the “ \leq ” sign in formula (1). This is further demonstrated by our model calculations in Fig. 1(c) for three different values of the exchange coupling distribution width σJ_c , showing that the bias field for the saturated HL is always the same. In addition, the reduction of h_{bias}/M_r is shown to be a very sensitive function of σJ_c , which might prove suitable as a measurement tool for determining the interlayer coupling distribution with a precision in the single digit percent level, at least in the case of FM/FM systems where the measurement of M_r values is straightforward, or for those FM/AM systems where the average uncompensated moment can easily be determined. Finally, using the here developed physical picture of the reduced exchange bias for low HL magnetizations, the different curve shapes shown in Fig. 2 can be explained in a straightforward fashion as corresponding to differing widths and shapes of the interlayer coupling distribution. Such sample-to-sample variations are very likely to occur if one considers the only 3 atom thick nature of the Ru coupling layers in our samples.

Given the fact that the HL in our bilayer system is a hysteretic magnetic entity, it is expected that the h_{bias} vs M_r relation is not only nonlinear, but also exhibits hysteresis. This assumption is experimentally verified in Fig. 4, showing data for a complete setting field H_{set} cycle. Figures 4(a) and 4(b) show, respectively, the H_{set} field dependence of M_r and h_{bias} , and Fig. 4(c) shows the corresponding direct comparison between M_r and h_{bias} . While the initial inspection of the experimental data plotted in Figs. 4(a) and 4(b) that was done in Ref. 4 suggested that both M_r and h_{bias} follow a very similar H_{set} dependency and have a linear relationship with each other, the more careful analysis of the data in the

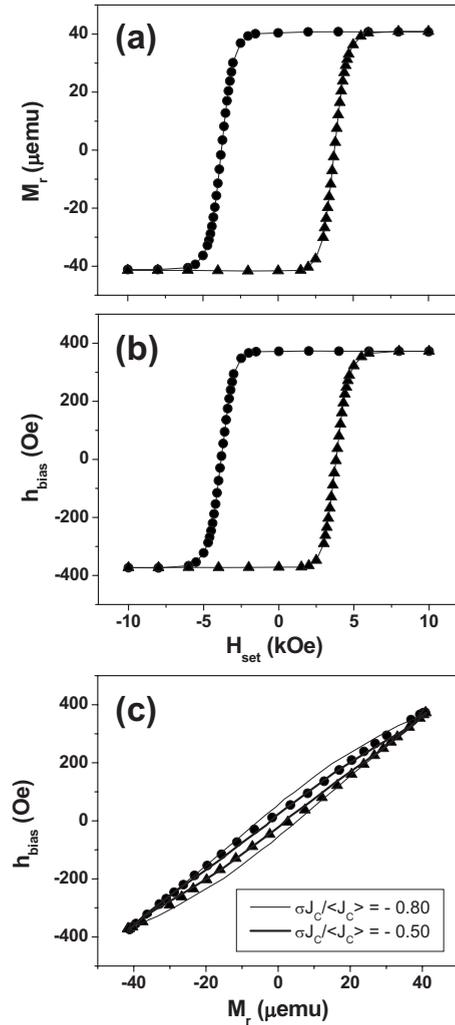


FIG. 4. Experimental data for an entire H_{set} cycle measured on an AF-coupled bilayer structure consisting of a 15 nm thick CoPtCrB-alloy HL and a 1.5 nm CoCr film as the SL: (a) remanent magnetization M_r of the HL, determined from the magnetization axis minor loop shift; (b) exchange bias field h_{bias} , determined from the field axis minor loop shift (a preliminary analysis of these data was shown in Ref. 4); (c) h_{bias} vs M_r . The different symbols correspond to the curve for decreasing H_{set} (●) and the curve for increasing H_{set} (▲). The lines shown in (a) and (b) are guides to the eyes, while the lines shown in (c) correspond to model calculations for two different values of the interlayer coupling distribution width $\sigma J_c / \langle J_c \rangle$.

present work reveals that M_r and h_{bias} are not fully synchronous and exhibit a phase shift which is responsible for the observed h_{bias} vs M_r hysteresis. These findings can also be understood within the previously developed physical picture. Upon applying a reversed H_{set} field, the most weakly coupled HL grains are preferentially switched at first and cause an inversion of M_r before the bias field h_{bias} can follow; hence the observed h_{bias} vs M_r hysteresis. Interestingly, this can result in HL magnetization states with the magnetic moment and the bias field pointing in the same direction despite the antiferromagnetic nature of the interlayer coupling, similarly to the situation responsible for positive exchange bias in conventional FM/AM structures.⁹ Our model calculations with

the parameter set yielding an excellent agreement with the H_{bias}/M_r vs M_r data in Fig. 1(c), i.e., $\sigma J_c/\langle J_c \rangle = -0.80$, are shown as solid lines in Fig. 4(c) and reproduce the experimentally observed behavior quite well, although the experimental hysteresis effect is slightly smaller. This, however, is not surprising because our model assumes completely independent HL grains—a condition which is experimentally not truly fulfilled simply due to the presence of dipolar interactions within the HL. Such interactions cause a commonly known phenomenon, namely, that dc-demagnetized states, such as the one populated in Fig. 4(c) at zero magnetization, show a higher magnetic correlation than ac-demagnetized states, which were the starting point for all plots in Fig. 1(c). Such a higher magnetic correlation will then reduce the effects of the interlayer coupling distribution, which is exactly what we observe given the good agreement between the experimental and modeled hysteresis loops obtained after reducing $\sigma J_c/\langle J_c \rangle = -0.50$. Thus, the various experimental results are consistent and are not only fully explained within the developed physical picture, but also described quantitatively by our simple bilayer model. It is worth mentioning that recent studies of the angular dependence of exchange bias in FM/AM systems also reported hysteretic behavior, which has been attributed to thermally driven relaxation

within the AF.²³ In view of our analysis, such hysteresis could also result from a fundamentally different mechanism, namely, from the nonthermal preferential reorganization of AF magnetic structure due to the reciprocal exchange bias effect produced by the FM.

In summary, we demonstrated that in exchange-biased FM/FM systems the exchange bias does not only act upon the SL, but also onto the HL and can be of crucial relevance in setting the HL magnetization state. In particular, variations of the interlayer coupling can result in correlating the magnetization state of the HL with those lateral coupling strength variations and hereby cause nontrivial changes in the observable exchange bias. We believe that these aspects are not limited to our FM/FM exchange bias systems, but also apply to conventional FM/AF materials. Here, interface topography, intermixing, as well as local composition gradients lead to lateral variations of the local coupling strength on the relevant length scale. These local coupling strength variations can then produce not only frustration effects near the interface, but can also influence the balance between the interface and the direct magnetic-field effects upon setting the magnetization state of the antiferromagnet, as in the case of the positive exchange bias.⁹

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- ¹⁹The hysteresis loops in Fig. 1(a) correspond to raw measurement data, which may show a slight opening that results from systematic shifting of the respective loop branches. This effect, which is due to the resonance frequency shift of a measurement baseline of the AGM instrument that was used in the experiment, has been removed from the data before performing any of the quantitative analysis reported in Figs. 1(b), 1(c), and 2.
- ²⁰Note that the observed behavior cannot be attributed to training effects. As shown in Ref. 17, the training effect does not exist in the limits of weakly magnetized and saturated HL and, therefore,

the curve shapes shown in Figs. 1(c) and 2 would have to be very different. In particular, a gradual monotonic reduction of H_{bias}/M_r for low HL magnetization could not be explained based on the training effect. We also note that the exchange bias in our FM/FM systems can produce asymmetric magnetization reversal. However, this is not the reason for the here reported exchange bias nonlinearity and hysteresis because a virtually identical behavior is observed even after the hysteresis loop asymmetry has been taken into account in the data analysis by means of more refined analysis schemes such as the one developed in Ref. 12.

²¹Contributions from dipolar interlayer coupling are negligible in comparison to interfacial exchange as discussed in Ref. 3.

²²Variables M_{loc} and J_{cloc} are averages calculated over local segments, which we chose here to be 5×5 spin blocks. This size is arbitrary and chosen only for the purpose of achieving a suitable display, but it does not change the physics. Smaller segment sizes, such as 1×1 spin blocks, for instance, have only two M_{loc} levels and do not allow a good graphic representation of the developing correlations.

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