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Exchange bias training relaxation in spin glass/ferromagnet bilayers

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A canonical spin glass (SG) FeAu layer is fabricated to couple to a soft ferromagnet (FM) FeNi layer. Below the SG freezing temperature, exchange bias (EB) and training are observed. Training in SG/FM bilayers is insensitive to cooling field and may suppress the EB or change the sign of the EB field from negative to positive at specific temperatures, violating from the simple power-law or the single exponential function derived from the antiferromagnet based systems. In view of the SG nature, we employ a double decay model to distinguish the contributions from the SG bulk and the SG/FM interface to training. Dynamical properties during training under different cooling fields and at different temperatures are discussed, and the nonzero shifting coefficient in the time index as a signature of slowing-down decay for SG based systems is interpreted by means of a modified Monte Carlo Metropolis algorithm. *Published by AIP Publishing.*
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Exchange bias (EB) refers to a coupling between an antiferromagnet (AFM) and a ferromagnet (FM) and gives rise to a shift and/or a broadening of the hysteresis loop characterized by EB field (H_E) and coercivity (H_C).[1–4] However, the cycled loop measurement quantified by the loop index n generally induces an EB degeneration, called the EB training.[5] As a dynamical aspect of EB, training helps us to unravel the mechanism of EB and eliminate the EB instability in spintronic devices. Thus, to elucidate training, diverse theoretical models have been proposed.[5–12] First, an empirical power-law function of $H_E \propto n^{-1/2}$ was used to fit experimental data.[5] However, a good fitting has to exclude the first data point of $n = 1$, and significantly, there has been a lack of theoretical understanding. Therefore, using micromagnetic calculations, Hoffmann[6] suggested that the symmetry of AFM magnetocrystalline anisotropy may play a crucial role in training. Moreover, Binek *et al.* gave a physical meaning to the empirical $n^{-1/2}$ dependence by discretizing the Landau-Khalatnikov model and proposed that the dynamical property of the pinning interface layer may determine training.[7–10] Similarly, Xi *et al.* derived a single exponential decay function to fit the experimental data based on the Kolmogorov-Avrami model.[11,12] Finally, the Monte Carlo simulation has also proven to be a powerful tool to study the mechanism of training.[13–15] Unfortunately, most of them focus on the EB training in magnetically ordered systems and the knowledge of training in complex disordered and frustrated systems is still lacking.

Spin glasses (SGs) are prominent disordered and frustrated spin systems.[16,17] Magnetic structures in the SGs are *metastable* because they are stuck in stable configurations other than the lowest energy configuration. The EB training in SG/FM bilayers has been reported previously.[18–22] Experimentally, Ali *et al.*[18] gave the SG thickness dependence of training in CuMn/Co bilayers and presented that H_E was unchanged after the second loop cycle when the SG thickness was large enough. Yuan *et al.*[19] studied the training in FeAu/FeNi bilayers and conjectured that the irreversible spin structure rearrangement was triggered by an FeNi magnetization reversal and magnetic field, not by irreversible movement of domain walls in the SG layer. In the same system, Rui *et al.*[20] and Zhang *et al.*[21] analyzed the training from the evolution of coercive fields at different loop branches and improved the empirical power-law to fit data well. Theoretically, Usadel and Nowak[22] predicted that the SG states in SG/FM bilayers were rather stable even for the training of $n = 10$. Although the training phenomena have been observed in varied SG/FM bilayers as mentioned above, a deep understanding of the magnetic relaxation mechanism of the training has remained absent yet. In this letter, we still choose the FeAu/FeNi SG/FM bilayers as a representative system and demonstrate the dynamical properties of EB during training linked to the nature of SG.

A multilayer sample with a stacking sequence of Ta (4 nm)/$Fe_{11}Au_{89}$ (50 nm)/$Fe_{19}Ni_{81}$ (5 nm)/Ta (2 nm) was deposited on the silicon wafer by DC magnetron sputtering at room temperature, and the detailed descriptions of sample fabrication and characterization on the SG nature can be referred to Refs. 20 and 23. Then, the samples were cooled from room temperature to 2 K under a specific H_{FC} ranging from 0.2 kOe to 5 kOe. Finally, the magnetization hysteresis loops between 5 kOe and −5 kOe were measured repeatedly till $n = 20$. H_E and H_C were obtained by $H_E = (H_{CR} + H_{CL})/2$ and $H_C = (H_{CR} - H_{CL})/2$, where H_{CR} and H_{CL} were the coercive fields at the ascending and descending branches, respectively.

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Magnetic operations and measurements above were performed in a SQUID-VSM (Quantum Design).

Theoretically, a paradigmatic model in the statistic physics that could exhibit the SG complexities is the Sherrington-Kirkpatrick model.[24] In a dilute alloy, the coupling distribution can approximate to the Edwards-Anderson form[25] or the simpler "$\pm J$" form.[26,27] Additionally, it is more appropriate to replace the ideal Ising model by the Heisenberg vector model to mimic the magnetic moments of SGs at finite temperatures.[22,23] Hence, the Hamiltonian of spin i in the presence of the magnetic field is written as

$$H_i = -K_i V_i (\mathbf{S}_i \cdot \boldsymbol{e}_i)^2 - \mathbf{S}_i \cdot \left(J_{ij} \sum_j \mathbf{S}_j + g\mu_B \mathbf{H} \right), \qquad (1)$$

where $\mathbf{S}_i$ denotes the vector of magnetic moment and $\boldsymbol{e}_i$ denotes the unit vector along the easy axis. For simplicity, only the dominant terms related to EB are considered. The simulation procedure mimics the experimental one. As for the SG spin update, we use a modified Monte Carlo method. The magnetic parameters and the simulation method used are described in Ref. 28 in detail.

At first, the H_{FC} dependence of 2-K training is studied and the results are shown in Fig. 1. For different H_{FC}s, both values of H_E and H_C drop abruptly between the first and the second loop cycles, and H_E and H_C do not cease to decrease even when n reaches 20. In some literatures,[20,21,29] it has been found that some existing training theories based on the ordered AFM/FM systems become incompetent to cope with the training phenomena observed in the SG/FM bilayers. However, at the temperatures below the SG freezing point, it

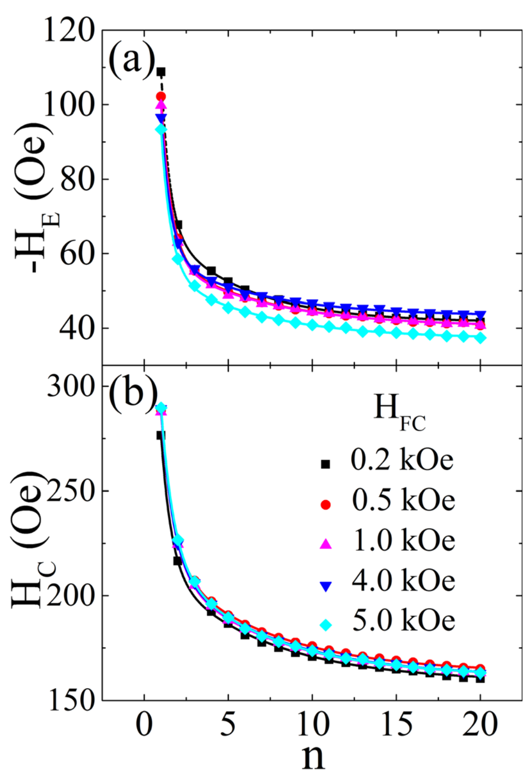

FIG. 1. Training in FeAu/FeNi spin glass/ferromagnet bilayers at 2 K after cooling under different fields. Symbols are the experimental data of (a) exchange bias field and (b) coercivity and solid curves are the fitting results obtained from Eq. (2).

is reasonable that the relaxation still obeys an exponential decay. Furthermore, we need to distinguish the contributions from the SG bulk and the SG/FM interface to training. Therefore, we suggest a tentative function for training in order to study the mechanism in SG/FM bilayers,

$$\begin{aligned} \mp H_{\mathrm{E/C}}(n) = \mp H_{\mathrm{E/C}}(\infty) + A_s^{EB/CO} \exp\left[-\frac{n - n_0^{EB/CO}}{\tau_s^{EB/CO}} \right] \\ + A_i^{EB/CO} \exp\left[-\frac{n - n_0^{EB/CO}}{\tau_i^{EB/CO}} \right], \end{aligned} \qquad (2)$$

where $-H_{\mathrm{E}}(\infty)$ and $H_{\mathrm{C}}(\infty)$ are the limiting values referring to an infinite number of cycles of H_{E} and H_{C}. Superscript EB/CO corresponds to $H_{\mathrm{E}}/H_{\mathrm{C}}$ and subscripts s and i correspond to the contributions of the SG bulk and the SG/FM interface. The parameter A has a dimension of field and its value can reflect the weight between the SG bulk and the SG/FM interface in training, analogous to a kind of amplitude of decay. Parameters τ, n_0, and discretized n have the identical dimension of time, where τ is a characteristic relaxation time length and its temperature dependence can be well described by the Arrhenius law.[11] In other words, τ determines a relaxation velocity, and the smaller is the τ, the faster is the relaxation. The parameter n_0 is defined as a shifting coefficient and its physical meaning will be discussed later. The functional curves are well fitted to the experimental data with the results also depicted in Fig. 1, and the values of various parameters for different H_{FC}s are summarized in Fig. 2.

From Eq. (2), we consider that the values of H_{E} and H_{C} derived from the nth loop cycle consist of three parts: the value in the time-independent equilibrium state plus the residual values after decays related to SG bulk and SG/FM interface. The trends of $-H_{\mathrm{E}}(\infty)$ and $H_{\mathrm{C}}(\infty)$ with increasing H_{FC} are consistent with the results obtained in Ref. 23 and unnecessary to be discussed further. For decay amplitudes, A_s is higher than A_i for both H_{E} and H_{C}. If an exponential term is equal to unity, A depends on the field cooling process. Therefore, H_{FC} is slightly destructive to A^{EB}, while it has no effect on A^{CO}.[23] On the other hand, τ is relatively fixed with H_{FC}, i.e., $\tau_s^{EB} = (0.57 \pm 0.04)$, $\tau_i^{EB} = (5.6 \pm 0.5)$, $\tau_s^{CO} = (0.72 \pm 0.05)$, and $\tau_i^{CO} = (6.1 \pm 0.1)$. According to the Arrhenius law, τ is a function of attempt frequency, energy barrier, and temperature. We have demonstrated that a large H_{FC} induces a high energy barrier in the SG layer,[23] combined with τ independent of H_{FC}. Therefore, at a finite temperature, the attempt frequency should depend on H_{FC} as well; in other words, a large H_{FC} reduces the magnetic viscosity which is controlled by interactions. Meantime, τ_s is nearly ten times smaller than τ_i, causing that the field decay related to SG bulk is faster than the SG/FM interface. Consequently, the SG bulk plays a crucial role in training with small n, and for large n, the SG/FM interface becomes dominant. Similar behaviors are also observed in some AFM/FM systems.[30–32] Interestingly, for SG/FM bilayers, there exists an additional shifting coefficient n_0, and its value is equal to 1.0 approximately. When $n = 1$, Eq. (2) can be written as $\mp H_{\mathrm{E/C}}(1) \approx \mp H_{\mathrm{E/C}}(\infty) + A_s^{EB/CO} + A_i^{EB/CO}$ and

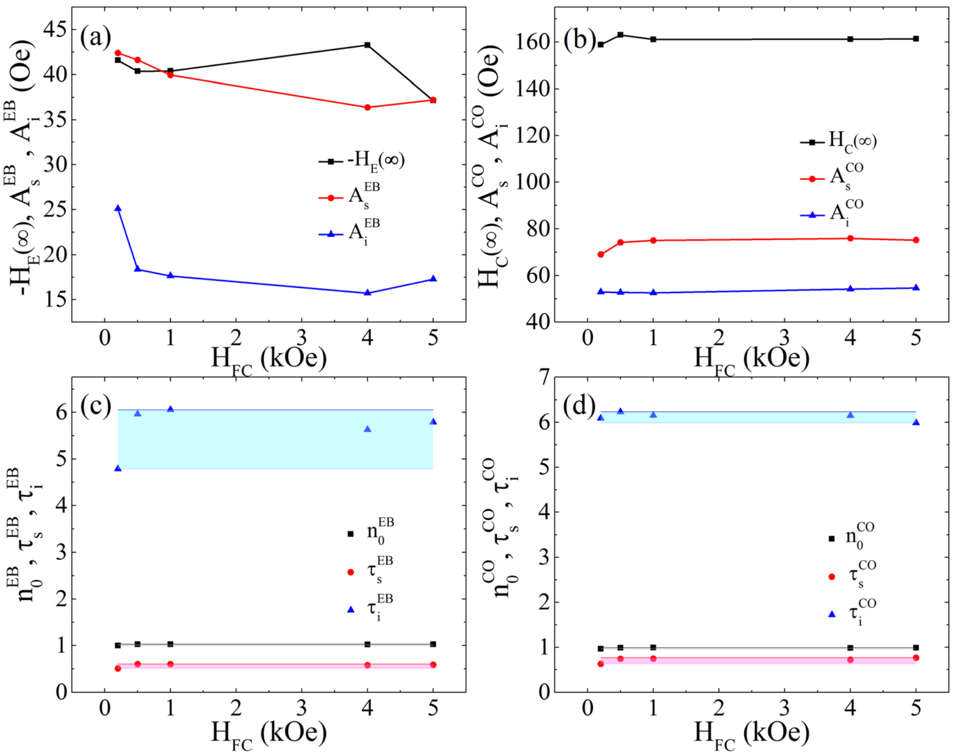

FIG. 2. Fitting parameters of Eq. (2) as functions of cooling field. Superscript is used to distinguish from exchange bias and coercivity and subscript is used to distinguish from spin glass bulk and spin glass/ferromagnet interface. Shadow regions in (c) and (d) show fluctuations.

thus describes the case without any decay. For larger n, the existence of n_0 is prone to induce the sustained reduction in H_E and H_C with increasing n.

For further verifying the training relaxation properties in SG/FM systems, the temperature dependent training is studied. We used our previous experimental data plotted in Figs. 5(a) and 5(b) in Ref. 21 to discuss this issue. The 5-K training changes the sign of H_E from negative to positive and the 7-K training increases H_E further towards the positive field direction. Both the 5-K and 7-K training behaviors also obey the double exponential decay presented in Eq. (2), with the fitting parameters listed in Table I. Remarkably, A is temperature dependent and decreases monotonically with elevated T, also confirming that A strongly depends on the frozen configurations in the SG bulk and the SG/FM interface. Also, τ is temperature dependent. With increasing temperature from 2 K to 5 K, positive H_E appears during training and τ increases. However, based on the Arrhenius law, a lower attempt frequency or a higher energy barrier is favored to enhance τ, in contradiction with the results obtained in the AFM/FM systems.[1] In Ref. 23, we have proposed that the positive H_E near the blocking temperature is due to the interfacial coupling that changes its type from SG to AFM. Hence, the training rearranges the configuration in the SG bulk and SG/FM interface, and H_E increases towards the positive field direction and meanwhile the thermal excitation is suppressed. However, n_0 is temperature independent. Therefore, n_0 should be material dependent and linked to some intrinsic properties in the SG.

To interpret n_0, the training with small n is simulated using the Monte Carlo simulation with the Metropolis algorithm modified in Ref. 28, and the phenomena in SG/FM and AFM/FM bilayers are shown for comparison. The results are presented in Fig. 3. Under a weak H_{FC}, a negligible H_E and no training are obtained for AFM/FM bilayers. On the contrary, training is observed in the SG/FM bilayers and the trend is also fitted by Eq. (2) using a set of the individual parameters with simulation unit, and thus the results in Fig. 3(a) well prove the validity of the present model and the method. Normally, the EB phenomena are linked to the change of magnetization at the interface. Therefore, Fig. 3(b) gives the simulation results of magnetization at the SG/FM and AFM/FM interfaces during loop cycles. During the first loop cycle, the irreversible magnetization reversal at the SG/FM interface is remarkable and it is attributed to the energy minimization of the interactions between the SG and FM interfacial spins and between the intralayer and interlayer SG spins. Hence, the resultant EB phenomenon is hardly to be interpreted by using a regular training theory. From $n \geq 2$, the magnetization variation at the SG/FM interface becomes periodic and multistep as compared to that at the AFM/FM interface. For example, when $n = 2$, under large H marked as Nos. 1, 3, and 5 in Fig. 3(b), H plays a major role in the magnetic relaxation in the SG layer, similar to the case

TABLE I. Fitting parameters of Eq. (2) for different temperatures.

T (K)	$-H_E(\infty)$ (Oe)	A_s^{EB} (Oe)	A_i^{EB} (Oe)	n_0^{EB}	τ_s^{EB}	τ_i^{EB}	$H_C(\infty)$ (Oe)	A_s^{CO} (Oe)	A_i^{CO} (Oe)	n_0^{CO}	τ_s^{CO}	τ_i^{CO}
2	24.38	34.34	11.76	0.98	0.51	3.98	157.03	73.98	52.99	0.99	0.73	5.68
5	−5.50	15.95	4.99	0.99	0.62	5.31	192.15	32.04	29.78	0.97	0.80	6.14
7	−9.07	6.44	2.38	1.00	0.64	4.78	199.14	13.86	12.54	0.97	0.72	6.15

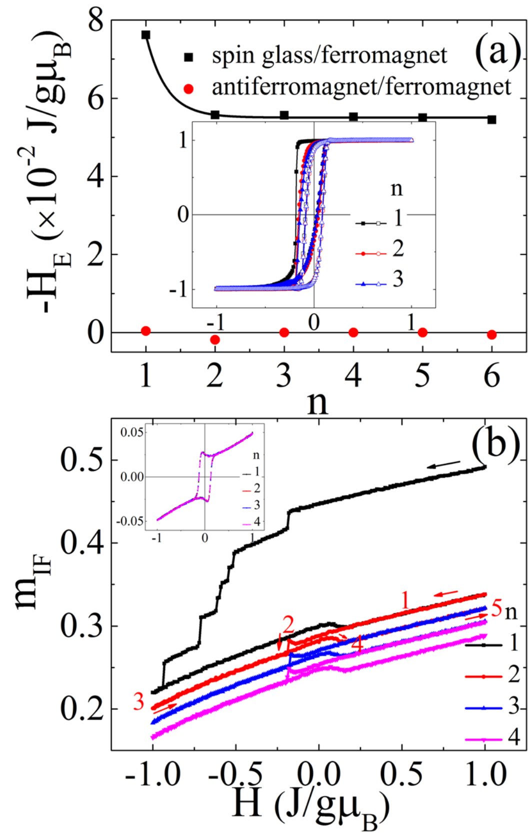

FIG. 3. Simulation results of training at $T = 0.001\,J/k_B$ after cooling under $H_{FC} = 0.2\,J/g\mu_B$. (a) Exchange bias training in spin glass/ferromagnet and antiferromagnet/ferromagnet bilayers, where the solid curve is the fitting result obtained from Eq. (2) and the inset shows the magnetization hysteresis loops of the ferromagnet component coupled to a spin glass (solid symbols) or an antiferromagnet (open symbols) during the first three loop cycles. (b) Evolution of the magnetization at the spin glass/ferromagnet interface during the first four loop cycles, where the arrows point out the magnetizing directions, the numbers are used to distinguish the multistep relaxation, and the inset gives the corresponding evolution of the magnetization at the antiferromagnet/ferromagnet interface.

in the AFM. However, under weak H, the magnetization reversal in the FM turns to become a dominant source to influence the relaxation behavior in the SG via interfacial couplings, as Nos. 2 and 4 shown in Fig. 3(b). During field cooling, the magnetization at the interface has a positive surplus value, since some SG spins are ferromagnetically coupled to a saturated FM. When H decreases in the positive direction and increases in the opposite direction, it can reduce the surplus magnetization and cause training after perpetual repetition of field cycles. However, during the magnetically hysteretic measurement, the FM magnetization reversal from positive to negative induces an increase in the surplus magnetization [seen at No. 2]. It is implied that the AFM coupling at the interface plays a crucial role and the peak value even approaches to the magnetization during the previous loop cycle, equivalent to a partial training recovery triggered by interactions. At No. 4, the increase of H in the positive direction enhances the surplus magnetization, while the second FM magnetization reversal hampers the increase. In other words, the SG typed couplings in the hysteresis loops are competing with H and can effectively slow down the training relaxation. Similarly, Zhou *et al.*[33] supposed that there existed a "trajectory" of the surplus magnetization during training in IrMn/YIG AFM/FM bilayers, and the surplus magnetization at the AFM/FM interface is oscillating and damping about the field axis due to FM magnetization reversal. On the contrary, the change of surplus magnetization at the SG/FM interface triggered by the FM magnetization reversal is smoother and unidirectional, also indicating that the training relaxation time is longer in the SG/FM bilayers. Consequently, on the one hand, n_0 is necessary to compensate the training between the first and the second loop cycles. On the other hand, for further training with $n \geq 2$, n_0 is used to increase the decay factor, i.e., the exponential term excluding A, and thus slows down the training relaxation for every n. It is attributed to the entangled exchange couplings in SG/FM bilayers and thus not observed in the ordered AFM/FM systems.

In summary, dynamical properties of the EB training in SG/FM bilayers are studied under different H_{FC} and at different temperatures. The results indicate that the training behaviors failed to be interpreted by the present theories derived from the AFM/FM systems. Both the SG bulk and the SG/FM interface contribute to the training based on the different magnetic relaxation properties. Significantly, a nonzero shifting coefficient needs to be considered for the EB training in the SG/FM bilayers to reflect an abrupt jump after $n = 1$ and slowing-down decay at every n as compared to the AFM/FM systems. Based on the modified Monte Carlo method, it is proved that a multistep and staggering evolution of the surplus magnetization at the interface emerges during every loop cycle due to the coexistence of FM and AFM couplings in the SG. The theory of double decay with an adjustable time index is also suitable for the training in other SG materials and SG-like magnets and phases.

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