We report on the experimental and theoretical studies of cooling field ($H_{FC}$) and temperature ($T$) dependent exchange bias (EB) in Fe$_x$Au$_{1-x}$/Fe$_{19}$Ni$_{81}$ spin glass (SG)/ferromagnet (FM) bilayers. When $x$ varies from 8% to 14% in the Fe$_x$Au$_{1-x}$ SG alloys, with increasing $T$, a sign-changeable exchange bias field ($H_E$) together with a unimodal distribution of coercivity ($H_C$) are observed. Significantly, increasing in the magnitude of $H_{FC}$ reduces (increases) the value of $H_E$ in the negative (positive) region, resulting in the entire $H_E$~$T$ curve to move leftwards and upwards. In the meanwhile, $H_{FC}$ variation has weak effects on $H_C$. By Monte Carlo simulation using a SG/FM vector model, we are able to reproduce such $H_E$ dependences on $T$ and $H_{FC}$ for the SG/FM system. Thus this work reveals that the SG/FM bilayer system containing intimately coupled interface, instead of a single SG layer, is responsible for the novel EB properties.

Recently, spin glasses (SG) have attracted much attention and stimulated intensive studies because the combination of quenched spin spatial randomness and frustration create a complex free energy landscape with multiple local energy minima and finding the stable spin states formidable challenging 1–3. In spite of many theoretical attempts of developing renormalization group 1,4, mean-field approximation 5 and Monte Carlo simulation 3,6–8 based analyses, the low-temperature ($T$) spin phases as well as the out-of-equilibrium aging dynamics of such materials remain matters of strong debates. Not surprisingly, the complex nature of SG spin structures gives rise to the unique and diverse macroscopic properties in many fields of science. Exchange bias (EB) for ultrahigh-density magnetic recording is one of such areas 9. The EB effect in coupled ferromagnet (FM)/antiferromagnet (AFM) system has been studied extensively because of its critical role in spintronic devices and intriguing spin physics. The EB phenomenon manifests itself as a shifted and broadened magnetization-applied field ($M$-$H$) hysteresis loop along the $H$ axis when FM/AFM systems are cooled below an EB blocking temperature ($T_B$) under an external magnetic cooling field ($H_{FC}$) 10,11. Therefore, EB is both $H_{FC}$- and $T$-dependent normally. Most of the EB studies show that large enough $H_{FC}$ can either saturate a negative EB field ($H_E$) for FM typed interfacial couplings ($J_{IF}>0$) 12,13 or induce a positive $H_E$ for AFM typed $J_{IF}$ ($<0$) 14–16. On the other hand, the thermal energy at high $T$ will weaken $J_{IF}$ and thus $H_E$ often decreases monotonically with increasing $T$ and finally vanishes at $T_B$. Under certain circumstances, a sign inversion of $H_E$, i.e., a small positive EB, can be observed in a narrow $T$ region just below $T_B$ for some EB systems 17,18. These abnormal phenomena are argued to be a result of the unidirectional coercivity ($H_{C}$) enhancement along the $H_{FC}$ direction. Observation of this effect requires a modest $H_{FC}$ and it has been only reported in quite few AFM based materials since the origin of positive EB is very different from systems with negative $J_{IF}$ 14. Therefore, the study of $H_{FC}$ together with $T$ dependences of EB is of critical importance for the understanding of EB mechanism.

1National Laboratory of Solid State Microstructures and Department of Physics, Nanjing University, Nanjing 210093, P. R. China. 2College of Sciences, Northeastern University, Shenyang, 110819, P. R. China. 3Department of Physics, Tongji University, Shanghai, 200092, P. R. China. 4Collaborative Innovation Center of Advanced Microstructures, Nanjing, 210093, P. R. China. 5MOE Key Laboratory for Anisotropy and Texture of Materials, Northeastern University, Shenyang, 110819, P. R. China. Correspondence and requests for materials should be addressed to Y.H. (email: huyong@mail.neu.edu.cn) or J.D. (email: jdu@nju.edu.cn)
As mentioned previously, when SG is involved, the EB dependences on $H_{FC}$ and $T$ can be different from conventional FM/AFM systems. Nayak et al.\textsuperscript{25} found that zero-field-cooled (ZFC, $H_{FC}=0$) $H_E$ in Heusler compound Mn$_5$PtGa is comparable to the field-cooled (FC) value, due to the coexistence of field-induced irreversible magnetic behavior and a SG-like phase. Sabayasachi et al.\textsuperscript{26} found that even a strong $H_{FC}$ of 80 kOe cannot saturate the $H_C$ of nanocrystalline La$_{17}$Sr$_2$FeO$_{19}$ at 5 K and this behavior was attributed to the randomness of glassy magnetic phase. Especially, by using a canonical SG alloy (CuMn), Ali et al.\textsuperscript{21} first reported the sign-changeable behavior of $T$-dependent $H_E$ in SG/FM bilayer systems. Subsequently, Yuan et al.\textsuperscript{25} and Ali et al.\textsuperscript{21} observed the similar $H_E \sim T$ trend in other SG materials (FeAu and FeCr). Without performing any $H_{FC}$-dependent studies, they interpreted those abnormal $T$-dependent EB behaviors either in the framework of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction theory\textsuperscript{21,22} or by considering the existence of a $T$-driven SG-to-AFM phase transition\textsuperscript{23}. However, due to the diverse and entangled spin interactions existed in SG and the fact these interactions can be affected by the field cooling process, the $T$-dependent sign-changeable $H_E$ should be greatly influenced by $H_{FC}$ in the SG/FM bilayers. Theoretically, Usadel and Nowak\textsuperscript{24} have attempted to reproduce the EB phenomena in the SG/FM bilayer systems by applying an Ising model considering a short-range Gaussian distribution of interactions to simulate the SG material. While they were able to predict the decrease in the magnitude of $H_E$ at low $T$ when increasing $H_{FC}$, the model failed to obtain the $H_E \sim T$ behaviors as seen in Ref. 21–23. To the best of our knowledge, the experimental observations of the $H_{FC}$ dependence of EB in SG/FM bilayer systems has remained unreported yet, and there are no unified non-phenomenological models that can interpret both the $T$ and $H_{FC}$ dependences of EB in SG based systems.

In this paper, the effects of $H_{FC}$ and $T$ on EB in the FeAu/ NiFe SG/FM bilayers were first investigated experimentally. Phenomena of $H_E$ sign inversion at the temperatures just below $T_p$, the decrease (increase) in $H_E$ magnitude with increasing $H_{FC}$ at low $T$ (high $T$), the unimodal distribution of $H_C$ against $T$, and the $H_{FC}$-independent $H_E$ have all been observed. Then using Monte Carlo technique, we exploited a short-range SG vector model, with a combination of disorder and frustration as well as thermodynamic relaxation and successfully reproduced and interpreted the EB phenomena governed by $H_{FC}$ and $T$. Distinct from the AFM based systems, this study suggests that the EB effect and especially the sign-changeable (positive EB) behavior in the narrow $T$ region just below $T_p$ in the SG/FM bilayer systems may be inherent, and exists in all exchange biased SG/FM bilayers.

Results

The quantities of $H_E$ and $H_C$ are calculated based on $H_E = (H_{C1} + H_{C2})/2$ and $H_C = (-H_{C1} + H_{C2})/2$, where $H_{C1}$ and $H_{C2}$ denote the coercive fields at the descending and the ascending branches of the $M$-$H$ hysteresis loop, respectively. The temperature dependences of $H_E$ and $H_C$ in FeAu/ NiFe bilayers are shown in Fig. 1, where $x = 4\%$, 8\%, 11\% and 14\% and $H_{FC}$ is 5 kOe for these measurements. In order to confirm the SG nature of the FeAu layers, ZFC-FC curves were measured with an applied field of 50 Oe. The inset in Fig. 1 shows typical features of spin glass behaviors for a FeAu single layer with $x = 11\%$. The value of the freezing temperature ($T_f$) for this sample is about 30 K, below which the ZFC and FC curves become bifurcated. DC memory effect\textsuperscript{25} has also been observed in this sample (not shown), providing a further proof of the SG state. Similar SG behaviors can be found in all other FeAu single layer films with $x$ varying from 4\% to 14\%.

Figure 1(a) shows a clear sign-change in $H_E$ versus $T$ in the FeAu/ NiFe bilayers with $x = 8\%$, 11\% and 14\%. With increasing $T$ from the lowest value of 2 K, $H_E$ increases abruptly from a negative value and changes its sign at $T_0$, which is named the compensation temperature\textsuperscript{21–23}. When $T$ is increased further, $H_E$ increases to a positive maximum at another temperature defined as $T_p$. After that, $H_E$ decreases gradually and falls below to zero when $T$ approaches to $T_p$. As for the $H_C$, dependence on $T$, it increases initially with $T$ and also shows a concurrent peak at around $T_p$ as shown in Fig. 1(b). It is noteworthy that $H_C$ shows a maximum positive value of about 40 Oe when $x = 8\%$, much larger in magnitude than any other previous reported results (normally less than 10 Oe), even for a thin FM layer of 2.2 nm in ref. 21. However, the sign-changeable behavior of $H_E$ with $T$ is absent when $x = 4\%$, possibly because the value of $T_p$ is lower than 2 K and beyond the present temperature measuring range.

Figure 1 unambiguously demonstrates that for the SG/FM bilayers the $H_E \sim T$ curve depends strongly on the composition of the SG layer. Previously, such sign-changeable behavior of $H_E$ against $T$ was explained by the competition between long-range oscillatory RKKY couplings from the spins deep inside the SG layer and those close to the interface\textsuperscript{21–23}. In the present work, besides reconsidering the sign-changeable behavior, we focus on studying the $H_E$ and $H_C$ influenced by $H_{FC}$. First we discuss the evolution of $M$-$H$ hysteresis loops versus $T$. Figure 2 shows the $M$-$H$ hysteresis loops obtained between 2 K and 20 K for the Ta(4 nm)/Fe$_{11}$Au$_{89}$(50 nm)/FeNi(5 nm)/Ta(2 nm) sample, after cooled from 300 K to 2 K under $H_{FC} = 50$ kOe. The $M$-$H$ hysteresis loop measured at 2 K clearly shows that a negative EB was established after field cooling. As shown in Fig. 2(a), when $T$ increases from 2 K to 7 K, the ascending branch of the hysteresis loop shifts rightward while the descending branch keeps almost unchanged. With further increasing in $T$ from 8 K to 20 K, as displayed in Fig. 2(b), the hysteresis loop shrinks from both sides towards the center but the shift of ascending branch is more significantly than the descending branch. Therefore, the different $T$-dependent variation behaviors of the two loop branches lead to the concurrent peak on both the $H_E \sim T$ and $H_C \sim T$ curves, coinciding with the appearance of $T_p \sim 7$ K as
shown in Fig. 1. These results indicate that the $T$-dependent magnetization reversal mechanisms and/or the thermodynamic spin relaxation at the descending and ascending branches may be quite different.

Experimental study of the $H_{FC}$ dependent EB has been performed and representative results are presented in Fig. 3. As displayed in Fig. 3(a,c), when $H_{FC}$ increases from 0.2 kOe to 50 kOe, the $H_{E}$~$T$ curve shifts toward upper-left while the $H_{C}$~$T$ curves keep almost overlapped. Accordingly, $T_0$ decreases from 5.5 K to 4.2 K. The peak position of the $H_{E}$~$T$ curve moves towards lower $T$ slightly and the maximum positive value of $H_{E}$ increases significantly from 10 Oe to 18 Oe. Figure 3(b,d) show the corresponding
simulation results. The sign inversion in $H_E$ with $T$ can be reproduced and $H_E \sim T$ curve shift can also be repeated. It is noticed that simulated $T_0$, $T_p$, and $H_E(T_p)$ results have certain deviation from the experiments, but the theoretical $H_E \sim T$ curves agree well with experimental measurements. On the other hand, the calculated trend of $H_C$ versus $T$ is different from the experimental results obtained both by Ali et al.\(^21\) and us, but in agreement with those reported in ref. 22. Another disagreement with the present experimental results is that the theoretical $H_C \sim T$ curves are $H_FC$-dependent at the temperatures just below $T_B$, i.e., $H_C$ for larger $H_FC$ is slightly larger. These discrepancies will be addressed later.

At low $T$, the $H_EC$ dependence of $H_E$ is also interesting. The experimental and simulation results as shown in Fig. 4 indicate that the entire $M-H$ hysteresis loop moves rightwards with a strong enough $H_FC$. As a result, $H_E$ decreases with increasing $H_FC$ while $H_C$ is insensitive to $H_FC$. Furthermore, there is a slight vertical magnetization shift observed experimentally under strong $H_FC$, arising from a minor magnetization in the FeAu SG. Increase in $H_E$ at low $H_FC$ ($\leq 1$ kOe) is very small because saturation in the FeNi (FM) has not been achieved.

Based on the consistent $H_E$ results between experiment and simulation, we interpret the above experimental phenomena relying on our simulation method. At first, we have excluded that the EB phenomena in the SG/FM bilayers depend solely on interfaces [see Supplementary Material A]. In other words, the configurations/spins inside the SG influence the FM spins through the SG/FM interface and the SG/FM interface itself also plays a significant role in establishing EB. Thus in the low $T$ region, we calculate the interfacial exchange energy density ($\varepsilon_{IF}$) to interpret the relative EB behavior, which can be expressed as

$$\varepsilon_{IF} = - \frac{1}{A} \sum_{i<j}^{\text{IF \& FM}} \mathbf{\mu}_i \cdot \mathbf{\mu}_j,$$

(1)

where $J_{ij}^{IF}$ is the interfacial coupling strength between the FM and SG spins, $A$ is the interface area and $\mathbf{\mu}$ denotes the magnetic moment of the interfacial spin belonging to the FM or SG. Since this is the dominant energy term influencing the $H_E$ in SG/FM bilayers and meanwhile a low enough $T$ will suppress thermal fluctuation, the change of $\varepsilon_{IF}$ mainly determines the evolution of spin configuration at the interface during isothermally magnetizing at low $T$. Therefore, it provides us the opportunity to image the
M-H behaviors microscopically and helps us to elucidate how $H_E$ is influenced by $H_{FC}$. Figure 5(a) shows the results calculated for $T = 2.6$ K. In addition, other parameters including the spin energy barrier and the $x$ component of spin under specific fields are calculated simultaneously in order to provide a clear physical picture during isothermally magnetizing at low $T$, with the results shown in Fig. 5 (b,c), respectively. At low $T$ after field cooling, as shown in Fig. 5(a), $\varepsilon_{IF}$ for $H_{FC} = 0.2$ kOe is higher than that for $H_{FC} = 0.2$ kOe, and the value of $\varepsilon_{IF}$ keeps constant with decreasing $H$ in the positive direction. Then $H$ changes its sign and increases in magnitude, and the value of $\varepsilon_{IF}$ begins to decrease around $H = -100$ Oe. For $H_{FC} = 0.2$ kOe, $\varepsilon_{IF}$ can reduce to a lower value. With further increase in $H$ along the negative direction, $\varepsilon_{IF}$ begins to increase, corresponding to the magnetization reversal at the descending branch, and then reaches a comparable stable value both for $H_{FC} = 0.2$ kOe and 50 kOe. After the reversal, $\varepsilon_{IF}$ is unchanged approximately again so long as $H$ is applied in the negative direction. Once $H$ turns back to the positive direction and increases exceeding 100 Oe, $\varepsilon_{IF}$ decreases rapidly for both $H_{FC}$ accompanied by magnetization reversal at the ascending branch. Moreover, the rapid decrease of $\varepsilon_{IF}$ occurs at a larger $H$ for $H_{FC} = 50$ kOe.

The above hysteretic behavior of calculated $\varepsilon_{IF}$ and the $H_{EC}$ dependent EB at low $T$ ($T < T_c$) can be understood as follows. After field cooling, the bilayers undergone different $H_{FC}$ possess different $\varepsilon_{IF}$ as a result of the competition between $H_{FC}$ and $J_{IF}$. Meantime, the competition also establishes a $H_{EC}$ dependent energy barrier ($E_b$) configuration, as shown in Fig. 5(b). These energy barriers confine spin rotation, and $E_b$ for $H_{FC} = 50$ kOe is apparently higher than that for $H_{FC} = 0.2$ kOe. The reason is that $H_{FC}$ is applied...
along with the SG anisotropy and high $H_{FC}$ drags more spins to align with it, so the anisotropy energy of these spins is minimized simultaneously. This will result in a higher $E_b$ according to the Stoner-Wohlfarth theory. When $H$ decreases along the direction of $H_{FC}$, no extra energy is required to change the configurations of bilayers, and the systems are called in frozen states. When $H$ reverses its direction to become opposite to $H_{FC}$ (termed negative direction), there is a critical field ($H \approx -100$ Oe) beyond which the spins in the FM single layer can be reversed. However, in the SG/FM bilayers the SG spins, via $J_{IF}$, can impede this process, while the interfacial spins still tend to rearrange towards low exchange energy direction. The rearrangement of SG spin is also influenced by $E_b$ and thus $\varepsilon_{IF}$ for $H_{FC} = 0.2$ kOe decreases to a lower value. It is worth noting that the lower the $\varepsilon_{IF}$ is, the tighter is the bond between spins. To reverse
the FM spins to the negative direction, larger $H$ is needed for $H_{\text{FC}} = 0.2 \text{ kOe}$ to overcome the exchange energy. Hence the magnitude of $H_{\text{C1}}$ for $H_{\text{FC}} = 0.2 \text{ kOe}$ is larger than that for $H_{\text{FC}} = 50 \text{ kOe}$.

When the magnetization reversal at the descending branch is accomplished, $\varepsilon_{\text{IF}}$ is increased abruptly up to a nearly identical value for both $H_{\text{FC}}$ and the bilayer spin configuration become frozen again. We have to resort to other approaches to interpret the subsequent magnetizing process. Accordingly, the SG spin orientations at the interface (denoted by $\mu^x$) under large negative $H$ are calculated and shown in Fig. 5(c), and $\mu^x = 2.2 \mu_B$ indicates that the spins are aligning with $H_{\text{FC}}$ while $\mu^x = 0$ means that the spins are perpendicular to $H_{\text{FC}}$. Remarkably, the SG spins for $H_{\text{FC}} = 0.2 \text{ kOe}$ show random orientations in the film plane, whereas for $H_{\text{FC}} = 50 \text{ kOe}$ all spins keep aligning with $H_{\text{FC}}$ although in both cases $\varepsilon_{\text{IF}}$ has almost the same value. The spin configuration difference under different $H_{\text{FC}}$ will influence the magnetization reversal at the ascending branch around $H = 100 \text{ Oe}$. For $H_{\text{FC}} = 0.2 \text{ kOe}$, randomly oriented SG spins are more easily to drag the FM spins to deviate from the negative field direction via $J_{\text{IF}}$ under weak $H$, minimizing the interfacial exchange energy. It resembles that $J_{\text{IF}}$ favors the reduction in the reversal field of FM spin, as illustrated in the top panel of Fig. 5(d). In comparison, for $H_{\text{FC}} = 50 \text{ kOe}$, collinearly aligned SG spins couple to the FM spins via positive or negative $J_{\text{IF}}$. For the FM spins pointing to the negative field direction, the interfacial coupling with negative $J_{\text{IF}}$ restrains the FM reversal while positive $J_{\text{IF}}$ favors the FM reversal. However, the FM reversal for positive $J_{\text{IF}}$ must overcome the anisotropy energy. Therefore, in both cases $J_{\text{IF}}$ demotes FM reversal, and thus the FM reversal is driven only by $H$ [see the bottom panel of Fig. 5(d)]. As a result, $H_{\text{C2}}$ for $H_{\text{FC}} = 0.2 \text{ kOe}$ is smaller than that for $H_{\text{FC}} = 50 \text{ kOe}$ and $H_{\text{C2}}$ for $H_{\text{FC}} = 0.2 \text{ kOe}$ is more pronounced.

On the other hand, the influence of $H_{\text{IC}}$ on $H_b$ at low $T$ will be briefly discussed here. It is well-known that $H_{\text{C2}}$ is determined by the density of pinned sites, such as defects, which controls the nucleation and propagation of domain walls in FM$^{26,27}$. In the present SG/FM bilayers, $H_{\text{C2}}$ also depends on the SG spin behaviors at the interface$^{26}$. As mentioned above, $H_{\text{FC}}$ can reduce $|H_{\text{C1}}|$ while enhance $H_{\text{C2}}$. We speculate that the decrement of $|H_{\text{C1}}|$ and the increment of $H_{\text{C2}}$ are comparable due to invariable $E_b$ during isothermally magnetizing. Therefore, $H_b$ becomes $H_{\text{FC}}$-independent.

When the $M$-$H$ hysteresis loops are measured at the temperatures between $T_b$ and $T_a$ ($T_b < T < T_a$), $H_b$ may change its sign to become positive and its magnitude is also $H_{\text{FC}}$-dependent. However, in contrast to $T < T_{\text{sp}}$ now $H_b$ increases with increasing $H_{\text{FC}}$, as the results calculated at $T = 12.52 \text{ K}$ shown in Fig. 6(a). The $M$-$H$ hysteresis loop for $H_{\text{FC}} = 50 \text{ kOe}$ has a larger width and shifts more towards right than that for $H_{\text{FC}} = 0.2 \text{ kOe}$. For the phenomenon of positive $H_b$ in SG/FM bilayers is directly due to the RKKY type interaction influenced by $T$ to a different extent$^{21,22}$ or the $T$-driven SG-to-AFM phase transition$^{23}$. However, all these interpretations lack microscopic evidence and do not take into account the influence of $H_{\text{FC}}$ on positive $H_b$. 

**Figure 6.** (a) Calculated $M$-$H$ hysteresis loops at $T = 12.52 \text{ K}$ after cooling under $H_{\text{FC}} = 0.2 \text{ kOe}$ and $50 \text{ kOe}$, and (b) the corresponding calculated interfacial exchange fields ($H_{\text{IF}}$) as a function of magnetic field, where arrows indicate the magnetizing directions.
Different from the case of low $T$, thermal fluctuations are enhanced at these temperatures, although
the SG spins can be still frozen below $T_c$. Especially for the FM spins with weak anisotropy, their orien-
tations will be randomized by thermal energy, so $\xi_B$ cannot well describe the behaviors of SG spins and
is no longer suitable to interpret the phenomena at these temperatures. Therefore, we turn to calculate
the average interfacial exchange field ($H_I$), which arises from the SG and interacts with the FM spins
through the interface as

$$H_I = \frac{1}{A} \sum_{i=FM} f_{ij}^I \mu_j,$$

(2)

Here $f_{ij}^I$ is the interfacial coupling between the FM and SG spins, $A$ is the interface area and $\mu_i$ represents
the magnetic moment of the SG spins at the interface that are directly coupled to the FM spin. Apparently,
$H_I$ is influenced by the SG and thus is $T$- and $H$-dependent. As shown in Fig. 6(a, b), $H_I$ remains positive
until $H_{C1}$ and then changes to negative before $H_{C2}$. Similar to the external magnetic field effect, $H_I$ favors
the spins to align with it and thus impedes the $H$-driven magnetization reversals for both branches. For
$H_{FC} = 0.2$ kOe or 50 kOe, $|H_I|$ in the fourth quadrant is generally larger than that in the second quadrant
with same applied magnetic field, i.e. $|H_I| (H_{C1}) > |H_I| (H_{C2})$. Hence a larger $H$ is needed to realize
magnetization reversal at the ascending branch, i.e., $H_{C2}$ is larger than $|H_{C1}|$. It results in positive $H_I$. For
larger $H_{FC}$ of 50 kOe, $H_I$ before $H_{C1}$ is close to that for $H_{FC} = 0.2$ kOe, leading to similar $H_{C2}$ for both $H_{FC}$.
However, in the fourth quadrant, $|H_I|$ for $H_{FC} = 50$ kOe is much larger than that for $H_{FC} = 0.2$ kOe [see Fig. 6(b)],
leading to a larger $H_{C2}$ [see Fig. 6(a)]. Consequently, the positive $H_I$ is larger for higher $H_{FC}$.

Discussion

Different from conventional AFM/FM bilayers, the spin configuration can be rearranged in the present
FeAu/FeNi (SG/FM) bilayers even at very low $T$ (e.g. 2 K) due to intrinsic randomness and frustration
in the SG layer. Significantly, our work reveals that this reconfiguration can be controlled by $H_{FC}$ and
behaves quite differently in different $T$ regions. When $T < T_c$, the spin reconfiguration in the SG can
easily occur. For small $H_{FC}$, the polarization of SG is weak. In the SG/FM bilayers, a saturated FM layer
will couple to the SG spins unidirectionally and result in prominent asymmetry of FM reversal at both
branches of an $M$-$H$ loop and a negative $H_I$. However, for large $H_{FC}$, the polarization of SG is enhanced,
impeding the reconfiguration process. Thus the asymmetry of FM reversal is weakened and $H_I$ decreases.
The experimental and theoretical findings are both consistent with the explanations proposed by Usadel
and Nowak. On the other hand, when $T_c < T < T_{SP}$, the thermal fluctuation cannot be ignored and
even may smear out other energy landscapes. The $\xi_B$ adopted at the low $T$ below $T_c$ is not suitable for
the explanation of positive EB effect. Therefore, more direct effect from SG to FM, i.e. $H_I$ is considered.
Positive (negative) $H_I$ favors the FM spins to align with (against) $H_{FC}$ and thus forms FM (AFM) type
interfacial coupling to the FM spins along the $H_{FC}$ direction. As the calculated results of $H_I$ shown in
Fig. 6(b), an FM-to-AFM transition type of interfacial couplings does occur and this process is $H$-driven,
which is different from the $T$-driven phase transition model suggested by Ali et al. Hence a positive
$H_I$ appears when $T_c < T < T_{SP}$. Moreover, although the thermal fluctuation deteriorates thermal stability,
large $H_{FC}$ leads to strong polarization of SG and enhance the system stability, which is beneficial to
$H_I$ and favors enhancement in $H_E$ [see Fig. 6]. As a result, the positive EB for large $H_{FC}$ is more obvious
than that for small $H_{FC}$.

Now we will discuss some of discrepancies between experiment and simulation briefly. In the actual
FeAu/FeNi bilayers, when $T$ is decreased, the exchange and anisotropy energies are enhanced gradually
to induce $H_E$ below $T_S$ and this process also contributes to $H_E$ enhancement. With further decreasing $T$,
due to the polycrystalline nature of the FM layer, the easy axes of the FM grains cannot fully lie along with
the magnetizing direction, leading to reduction of $H_E$. Therefore, the variation of $H_E$ versus $T$ is
non-monotonic. In the present simulation, a single crystal model including a uniaxial anisotropy and a
saturated FM was adopted to study the EB properties and the magnetizing directions were set to be along
with the easy axis. Therefore, the calculated $M$-$H$ hysteresis loops are more rectangle-shaped and the
calculated values of $H_E$ are larger than those obtained from experiments. Significantly, the increase of anisotropy energy at low $T$ in the simulation further contributes to the increase of $H_E$ when
the $M$-$H$ hysteresis loops are measured along the easy-axis direction. As a result, $H_E$ has a monotonic
and sharp increase with decreasing $T$. Also, due to the saturated FM in the initial state, the experimental
result of increase in $H_E$ with initially increasing $H_{FC}$ is not obtained in simulation. Since the model only
considers the most dominant energy terms concerning EB, a perfect agreement between theory and
experiment cannot be achieved.

To summarize, the dependence of EB properties in FeAu/FeNi SG/FM bilayers on $H_{FC}$ and $T$ have
been studied experimentally and numerically. A sign-changeable behavior of $H_E$ versus $T$ is observed,
accompanied by a nonmonotonic behavior of $H_E$ versus $T$. More significantly, a phenomenon of decrease
(increase) in magnitude of $H_E$ at low (high) $T$ with increasing $H_{FC}$ is first observed in experiment while
$H_E$ is always insensitive to $H_{FC}$ below $T_p$. It is also found that the mechanisms of negative and positive
$H_E$ influenced by $H_{FC}$ are quite different. Therefore, the highlight of this paper includes exhibiting the $H_{FC}$
modulation of the $T$-dependent EB behaviors in SG/FM bilayers experimentally and interpreting these
behaviors simultaneously by using a unified vector model.
Methods
Sample fabrication and measurement. The samples were deposited on silicon wafer by DC magnetron sputtering at room temperature with a stacking sequence of Ta(4 nm)/Fe/FeAu/FeNi(5 nm)/Ta(2 nm). Here, x denotes the atomic fraction of iron in FeAu alloy. The FeAu layer was co-sputtered with tilted iron and gold guns and the sample composition was characterized by Energy Dispersive X-Ray Spectroscopy (EDX). By fixing the sputtering power of iron and varying that of Au, x was varied from 4% to 14%. A 4 nm Ta buffer layer was deposited to promote the SG/FM morphology and a 2 nm Ta capping layer was deposited to prevent sample from oxidation. FeNi represents Fe$_{19}$Ni$_{81}$, which is used as the magnetic pinned layer. The base pressure for sputtering was better than 7.0 × 10$^{-6}$ Pa and the working Ar pressure was kept at 0.3 Pa during film deposition. Another series of Fe$_{1-x}$Au$_x$ single layer films with the identical composition and thickness (i.e., 50 nm) were also deposited on Kapton substrates as reference samples for the low-T SG characterization by means of the ZFC and FC $M-T$ measurements.

Magnetic hysteresis loop measurements were performed in a SQUID-VSM (Quantum Design) with the applied magnetic field (in the range from −5 kOe to 5 kOe) parallel to the film plane. Before each $M$-$H$ hysteresis loop measurement at a specific $T$, a magnet reset procedure (oscillatory demagnetization between ±1 Tesla) was performed in order to train the sample to an approximately equilibrium state to eliminate the EB training effect and reducing the residual magnetic field to less than 3 Oe. This will ensure that the obtained $H_E$ and $H_C$ are accurate with an error less than 3 Oe.

Theoretical model and calculation details. On atomic scale, the real coarse-grained SG/FM bilayers are simulated by a finite number of spins placed on the nodes of a simple cubic lattice. By means of effective size scaling, a lateral dimension of 40 × 40 units in the film plane and a film thickness of 5 monolayers are used with periodic boundary conditions only applying in the plane. Simulations were performed with the Heisenberg model, with the Hamiltonian written as

\[ H = -J_{FM} \sum_{\langle \mathbf{i}, \mathbf{j} \rangle \in \text{FM}} \mathbf{\mu}_i \cdot \mathbf{\mu}_j - \sum_{\mathbf{i} \in \text{FM}} \left( K_{ij} \cos \alpha_i + \sum_{\mathbf{j} \in \text{FM}} K_{ij} \cos \beta_i \right) - H \sum_{\mathbf{i} \in \text{FM}} \mathbf{\mu}_i \]

where $\mathbf{\mu}$ denotes the magnetic moment of the spin at site $i$. The first term describes the direct exchange energy in the FM. The next two terms are the magnetocrystalline and shape anisotropy energies of the FM, where $\alpha_i$ ($\beta_i$) is the angle between $\mathbf{\mu}_i$ in the FM and the x (z) axis. The fourth term is the Zeeman energy of the FM, where H is applied along the x axis. From the fifth to seventh term represent the direct exchange, magnetocrystalline anisotropy, and Zeeman energies of the SG. Here $\gamma_i$ is the angle between $\mathbf{\mu}_i$ in the SG and the x axis. And the last term is the interfacial direct exchange energy between FM and SG. Detailed scaling and parameterizing processes have been described in the Supplementary Material B.

The simulation protocol mimics the experimental process. Initially, a cooling process under an $H_{FC}$ is performed on the SG/FM bilayers, where the FM spins are all pointing to the $H_{FC}$ direction and the SG spins are randomly oriented, from $T=300$ K to a target temperature. Then, an isothermal magnetization is recorded by cycling $H$ from 5 kOe to −5 kOe to extract $H_E$ and $H_C$. As for the update of spin state, a path-related Metropolis algorithm is adopted during the Monte Carlo simulation, which has been also introduced in the Supplementary Material B in details. This calculation is repeated for 10$^4$ times per spin to find the equilibrium state of the system, and then an additional 10$^5$ steps are taken to measure the thermodynamic average of the magnetization. Finally, 200 independent realizations of the disorder are averaged to reduce the statistical errors.

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Author Contributions
J.D. and Y.H. initiated the study. W.R. prepared the samples by magnetron sputtering. W.R., B.Y. and W.Z. performed the magnetic measurements by SQUID-VSM. Y.H., A.D., M.X. and J.D. analyzed the magnetic results. Y.H. and A.D. developed the theoretical model and did the simulation calculations. Y.H., S.Z. and J.D. prepared the manuscript. All the authors contributed to discussions of the project.

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