Isothermal low-field tuning of exchange bias in epitaxial Fe/Cr$_2$O$_3$/Fe

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Moderate dc magnetic fields of less than 1 T allow tuning the exchange bias in an epitaxially grown Fe 10 nm/Cr$_2$O$_3$ 2.7 nm/Fe 10 nm trilayer between negative and positive bias fields. Remarkably, this tunable exchange bias is observed at least up to 395 K which exceeds the Néel temperature of bulk Cr$_2$O$_3$ (307 K). The presence of spontaneous exchange bias and the absence of training effects at room temperature suggest the existence of stable interface moments independent of antiferromagnetic long range order in Cr$_2$O$_3$. Furthermore, the coercivity remains constant, independent of the exchange bias field. In contrast, large training associated with nonequilibrium spin configurations of antiferromagnetically ordered Cr$_2$O$_3$ appears below 50 K. © 2007 American Institute of Physics. [DOI: 10.1063/1.2801519]

The majority of today’s realized spintronic devices$^{1–3}$ utilize the exchange bias$^4$ (EB) effect.$^{5–8}$ The latter is a fundamental proximity effect taking place at the interfaces of magnetic heterostructures including ferromagnetic (F)/antiferromagnetic (AF), F/ferromagnetic, soft F/hard F systems, and others. Conventionally, EB is established by field cooling the heterostructure below the Néel temperature $T_N$, or by applying a magnetic field during the fabrication process. EB manifests itself in the shift of the hysteresis loop along the magnetic field axis and a concomitant increase in the coercivity $\mu_0 H_C$. The EB magnitude $\mu_0 H_C$ depends on intrinsic parameters such as the exchange coupling at the F/AF interface, interface roughness, individual F and AF microscopic structure, or their thicknesses. However, it has been reported that EB can be tuned by various extrinsic parameters. In this context, a broad spectrum of experimental protocols has been employed: cooling in different magnetic fields,$^9$ cooling in zero field from different magnetization protocols has been employed: cooling in different magnetic fields, cooling in zero field from different magnetization protocols,10 cooling in combinations of dc and ac fields,11 or saturating the F in large negative fields and then measuring the remaining loop with different waiting times.12,13 Other irreversible approaches such as high-temperature annealing14 or ion irradiation15 have also been used. Nogués et al.$^16$ reported that EB can be tuned at room temperature by subjecting the system to pulsed fields as high as 55.6 T. All these attempts to tune EB externally involve tedious experimental treatments or impractically large magnetic fields not easily available, which make them unsuitable for practical applications.

In this letter, we present an epitaxially grown Fe 10 nm/Cr$_2$O$_3$ 2.7 nm/Fe 10 nm trilayer structure and demonstrate that EB can be tuned at room temperature by subjecting the system to moderate dc magnetic fields, $-1 \leq \mu_0 H_{dc} \leq 1$ T. The motivation of using Cr$_2$O$_3$ as the AF layer is based on our recent observation of an appreciable moment in single Cr$_2$O$_3$ films.$^{17}$ There the direction of the induced moment could be controlled by an applied dc field. Coupling of a tunable pinning moment to an adjacent F film is anticipated to provide tunable EB.

Moreover, EB above $T_N$ of the AF has recently been observed in other F-AF systems such as Co/NiF$_2$ (Ref. 18), Fe$_2$O$_4$/CoO (Ref. 19), and Ni$_{80}$Fe$_{20}$/Co$_3$O$_4$ (Ref. 20). There it is primarily attributed to either strain or proximity effects at the F/AF interfaces. In our trilayer, the EB is stable up to the highest measured temperature $T \approx 395$ K, which is far above the bulk Néel temperature of Cr$_2$O$_3$, $T_N \approx 307$ K. Here, we explore the possibility of using dc fields to control the EB effect. Unlike previous studies, our system does not require any specific field-cooling procedure.

The trilayer system, Fe 10 nm/Cr$_2$O$_3$ 2.7 nm/Fe 10 nm, is prepared in a molecular beam epitaxy chamber at a base pressure of $1 \times 10^{-10}$ mbar. The (0001)-oriented c-Al$_2$O$_3$ substrate is heated to and maintained at 573 K during the deposition process. Note that no external magnetic field was present during the deposition. A stoichiometric Cr$_2$O$_3$ film was grown by thermal evaporation of Cr metal in an O$_2$ partial pressure of $2.2 \times 10^{-6}$ mbar.$^{16}$ Growth rates of Fe and Cr$_2$O$_3$ were 0.37 and 0.28 nm/min.

Analysis of the large-angle x-ray diffraction pattern (Max-B, Rigaku D) in Fig. 1 illustrates that single-crystalline Fe and Cr$_2$O$_3$ films are formed. The (110)-oriented bcc Fe peak shows pronounced Laue oscillations originating from coherently scattering Fe (110) lattice planes. The distribution of scattered intensity around the central maximum at $2\theta = 44.67^\circ$ is fitted by the one-dimensional Laue diffraction function.

![Figure 1](https://via.placeholder.com/150)

**FIG. 1.** (Color online) X-ray diffraction pattern of Fe 10 nm/Cr$_2$O$_3$ 2.7 nm/Fe 10 nm trilayer. Solid lines are the fit to the Laue function according to Eq. (1). The inset shows small angle x-ray reflection data (circles) and the best fit (line) by using LEPTOS-2 program.
The hysteresis loop shift (EB) always means that the magnetic structure of the system has a reversible component that lifts the degeneracy of the state magnetized in the opposite direction. In our case, EB free from any training effect persists well above bulk $T_N$ of Cr$_2$O$_3$, and therefore, barring large proximity effects, it appears unlikely that the EB originates from exchange coupling with the antiferromagnet. It may be explained by the presence of magnetically hard regions that are only reversed by strong dc fields. Such regions may appear, for example, due to the formation of chemically modified interface regions such as Fe$_3$O$_4$ with high Curie temperatures. The presence of magnetically hard regions reaching even into the Fe film is consistent with recent neutron reflectometry studies.

We find that the sign of $\mu_0H_{eb}$ can be controlled exclusively by $\mu_0H_{dc}$ in our trilayer. This is reminiscent of the scenario of changing the sign of $\mu_0H_{eb}$ depending on the strength of the cooling fields. Note, however, that our EB tuning takes place isothermally where the pinning magnetization is reversed by a magnetic field only without breaking and reestablishing AF long range order during a field cooling process. Competition between AF exchange coupling of the pinning and the Fe magnetizations with the Zeeman interaction energy of the pinning system determines the sign of $\mu_0H_{eb}$. For $\mu_0H_{dc}=0.2$ T, AF coupling remains dominant over the Zeeman energy, resulting in regular (negative) EB. For larger $\mu_0H_{dc}$, the Zeeman interaction overcomes the AF exchange coupling which aligns the pinning moments parallel to larger $\mu_0H_{dc}$, leading to positive EB. Note that recently Cheon et al. pointed out that EB fields can be reversed in all EB systems below the blocking temperature via reversal of the uncompensated pinning magnetization. However, the isothermal field tuning described here takes place at easily accessible magnetic fields of less than 1 T, and does not depend on long range AF order.

Figure 2(b) shows $\mu_0H_{eb}=\mu_0(H_z+H_\perp)/2$ and $\mu_0H_{H_\perp} = \mu_0(H_z-H_\perp)/2$ versus $\mu_0H_{dc}$, where $\mu_0H_{H_\perp}$ and $\mu_0H_{dc}$ are the field values at which the magnetization becomes zero on right and left branch of the hysteresis loop, respectively. While $\mu_0H_{H_\perp}$ remains unchanged to within 1% of its mean value of $\sim$2 mT, $\mu_0H_{dc}$ varies in the range $-0.9<\mu_0H_{dc}\approx 1.2$ mT depending on the dc field. Note that identical EB fields have been determined from alternating gradient force magnetometry using electromagnets to generate the dc fields. Hence, trapped flux in the superconducting coils of the SQUID magnetometer is unambiguously excluded. It is also important to mention that the results are independent of the duration the system is subjected to the dc field. Indeed, we measured the same hysteresis loops with exposure times of 10, 100, and 300 s. The results presented in this letter correspond to exposure duration of 100 s.

Training of the exchange bias effect offers a unique tool to test the deviation from the equilibrium of the AF layer. To this end, we measured the training effect after tuning the exchange bias by various dc fields. Shown in Fig. 3(a) are the results at 300 K with $\mu_0H_{dc}=1$ and $-1$ T, respectively. Note that these loop shifts resemble positive EB. Clearly, the hysteresis loops remain unchanged implying the absence of any aging phenomena and, hence, indicating that this unusual high temperature exchange bias phenomenon originates from a stable interface moment. The latter is independent of the AF long range order which establishes at lower temperatures.
sent the best fit of $\mu_0 H_{eb}$ vs $n$ data to a recently developed phenomenological theory,  

$$\mu_0 [H_{eb}(n + 1) - H_{eb}(n)] = - \gamma (\mu_0 H_{eb}(n) - H_{eb}^0)^3,$$  

(2)

where $\gamma = 2 \times 10^{-4}$ (mT)$^{-2}$ and $\mu_0 H_{eb}^0 = 0.4$ mT were obtained as the two fitting parameters. $\gamma$ contains the interface exchange coupling and the damping constant governing the relaxation dynamics of the AF spin configuration while $\mu_0 H_{eb}^0$ is the quasiequilibrium exchange bias field.  

In conclusion, we found that exchange bias resulting from pinned interface magnetization in an epitaxial Fe/Cr$_2$O$_3$/Fe trilayer is isothermally tunable by moderate dc magnetic fields at room temperature. This behavior is distinct from regular exchange bias in the sense that it does not rely on field-cooling treatments and shows no training effect. It indicates that the pinning magnetization is independent of the long range AF order in the Cr$_2$O$_3$ spacer. Regular exchange bias with pronounced training behavior is, however, recovered below 50 K. Isothermally tunable exchange bias with moderate fields at room temperature is expected to impact future spintronic devices.

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