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Magnetoelectric Switching of Exchange Bias

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The perpendicular exchange bias field, \( H_{EB} \), of the magnetoelectric heterostructure \( \text{Cr}_2\text{O}_3(111)/(\text{Co}/\text{Pt})_3 \) changes sign after field cooling to below the Néel temperature of \( \text{Cr}_2\text{O}_3 \) in either parallel or antiparallel axial magnetic and electric freezing fields. The switching of \( H_{EB} \) is explained by magnetoelectrically induced antiferromagnetic single domains which extend to the interface, where the direction of their end spins controls the sign of \( H_{EB} \). Novel applications in magnetoelectronic devices seem possible.

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The linear magnetoelectric (ME) effect has been proposed decades ago to realize the control of electric and magnetic polarization properties by complementary fields in possible applications [1]. However, appropriate materials, which might fulfill the technologic demands, only recently became available. In particular, multiferroic manganites show convincing switching properties of their ferroelectric polarization by a magnetic field (e.g., \( \text{TaMnO}_3 \) [2] or \( \text{TbMn}_2\text{O}_5 \) [3]) or of their ferromagnetic magnetization by an electric field (e.g., \( \text{HoMnO}_3 \) [4]). Their very applicability, however, remains limited, since their ME properties are typical low temperature features.

A more favorable situation is met for the archetypical ME material chromium oxide, \( \text{Cr}_2\text{O}_3 \), which becomes magnetoelectric above room temperature, viz. below its antiferromagnetic (AFM) Néel temperature, \( T_N = 307 \) K [5]. This makes it interesting for devices involving the well-known exchange bias (EB) effect of exchange coupled ferromagnetic (FM) and AFM heterostructures [6]. They are widely used in magnetic random access coupled ferromagnetic (FM) and AFM heterostructures.

Our specimen was the layered heterostructure \( \text{Cr}_2\text{O}_3(111)/(\text{Co}/\text{Pt})_3 \) changes sign after field cooling to below the Néel temperature of \( \text{Cr}_2\text{O}_3 \) in either parallel or antiparallel axial magnetic and electric freezing fields. The switching of \( H_{EB} \) is explained by magnetoelectrically induced antiferromagnetic single domains which extend to the interface, where the direction of their end spins controls the sign of \( H_{EB} \). Novel applications in magnetoelectronic devices seem possible.

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In accordance with the phenomenological approach of Meiklejohn and Bean (MB) [6] there is now unanimity that a net interface magnetization of the antiferromagnet is necessary for generating EB. The simple MB expression

\[
\mu_0 H_{EB} = -J S_{AFM} S_{FM} / (M_{FM} t_{FM})
\]

(1)

describes the dependence of the bias field \( \mu_0 H_{EB} \) on a phenomenological coupling, \( J \), between the FM and AFM interface magnetizations \( S_{FM} \) and \( S_{AFM} \), respectively, where \( t_{FM} \) and \( M_{FM} \) are the thickness and the saturation magnetization of the FM layer. Equation (1) suggests that an extrinsic control of the EB field can be achieved by modifying the value of \( S_{AFM} \). Suitable control parameters are, e.g., temperature and magnetic freezing field [8]. Less well-known is the control of \( S_{AFM} \) using the piezomagnetic effect as observed, e.g., in \( \text{Fe}_2\text{O}_3\text{Zn}_0\text{F}_2/\text{Fe} \) [9]. Here we propose to achieve this goal by making use of a ME antiferromagnet, which allows to control interface magnetic moments by application of an external electric field [10].

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FIG. 1. Normalized hysteresis curves of Cr₂O₃(111)/Pt 0.5 nm/[Co 0.3 nm/Pt 1.5 nm] Pt 1.5 nm measured after magnetic field cooling in μ₀Hₑ₀ = 0.6 T and Eᵣ = 0 from T = 350 to 298 K (1) and after magnetoelectric field cooling to 250 K in μ₀Hₑ₀ = 0.6 T and Eᵣ = −500 kV/m (2) and Eᵣ = +500 kV/m (3), respectively. The lines are to guide the eyes. The exchange bias fields μ₀Hₑ₀ referring to the loops 2 and 3 are indicated by arrows.

rectangular with its center shifted to negative fields by an EB field, μ₀Hₑ₀ = −19.8 mT. A very similar hysteresis loop as shown by curve 2 with a negative EB shift, μ₀Hₑ₀ = −32.1 mT, emerging after negative MEFC, i.e., under the simultaneous application of an electric field antiparallel to the magnetic field. Here the sample was cooled from T = 350 to 250 K under the action of the freezing fields μ₀Hₑ₀ = 0.6 T and Eᵣ = −500 kV/m. Surprisingly, when inverting the sign of Eᵣ, i.e., when applying positive MEFC with μ₀Hₑ₀ = 0.6 T and Eᵣ = +500 kV/m, the EB also turns positive, μ₀Hₑ₀ = +30.3 mT (curve 3). The sign obviously follows the inversion of the electric field from Eᵣ < 0 to Eᵣ > 0, while the sign of μ₀Hₑ₀ has remained unchanged.

This amazing effect can be understood in terms of the so-called “magnetoelectric annealing” upon MEFC, which is known to create a single domain AFM state of the Cr₂O₃ crystal [13]. The two possible AFM domains A and B, say, differ by opposite orientations of the AFM vector, I = s₃ − s₂ + s₁ − s₄, where 1–4 denote adjacent spins within the magnetic unit cell as shown schematically in the lower panels of Fig. 2(a) and 2(b) [14]. In simultaneously applied fields Hₑ₀ and Eᵣ they have different ME energies, W_ME = −2αzz μ₀Hₑ₀Eᵣ, where αzz is the appropriate diagonal component of the magnetoelectric susceptibility tensor and αzz ≡ 0 refers to A and B, respectively [15]. AFM single domaining under MEFC is, hence, due to the energy difference between A and B domains. The formation of domain A upon cooling to below Tₘ is more probable than that of domain B if Hₑ₀Eᵣ < 0, and vice versa for Hₑ₀Eᵣ > 0.

FIG. 2. Schematic sketches of a FM/AFM bilayer with freezing fields (Hₑ₀ \pm Eᵣ), order parameters (M, I), and magnetic moments (S_FM, S_AFM), where the AFM layer is single domained (A and B, respectively) after MEFC (see text).

Bulk single domains are most convincingly demonstrated by their inherent ME susceptibility [5]. A net magnetic moment, Δm ∝ αzz E, is induced along the z axis by an external electric field, E, provided that the AFM system is single domained. This is shown in Fig. 3, where the magnetic moment of another heterostructure Cr₂O₃(111)/Co/Pt [16] has been measured by using superconducting quantum interference device (SQUID) magnetometry at T = 150 K in fields within the range |E| ≤ 400 kV/m. Where MFC of this sample in μ₀Hₑ₀ = 0.5 T and Eᵣ = 0 gives rise to a multidomain state with nearly vanishing slope (line 1, αzz = 0), MEFC with μ₀Hₑ₀ = 0.5 T and Eᵣ = −460 and +425 kV/m yields slopes of either sign, αzz < 0 (line 2) and αzz > 0 (line 3), due to the A and B domains, respectively. Note that the observed ME moments Δm add to the remanence of the sample, m(E = 0) = 1.8 × 10⁻⁸ A m².

Obviously AFM single domain formation under MEFC is at the heart of the observed switching of Hₑ₀. First of all we have to consider that merely 1%– 5% of the uncompensated AFM spins residing in the interface with the adjacent FM layer are responsible for the EB via S_AFM [17]. At T < Tₘ they occupy privileged positions, since they are robust against magnetic field cycles, which rotate the spins in both the FM and the topmost layers of the AFM component [18]. They are only loosely coupled to the FM, but strongly to the bulk of the AFM component. Hence, when inverting the orientation of I by MEFC, the orientation of the uncompensated spins, which constitute S_AFM, will also be inverted. This is schematically depicted in Fig. 2 for the cases μ₀Hₑ₀Eᵣ < 0 [2(a)] and μ₀Hₑ₀Eᵣ > 0 [2(b)], where the rhombohedral unit cells within the two domains of Cr₂O₃ have either outward (A) or inward directed (B) end spins.

It is now tempting to assume that these end spins are mainly responsible for the interface coupling. This immediately implies that S_AFM and S_FM are either parallel or antiparallel to each other, thus giving rise to (conventional) negative or (unconventional) positive EB as observed. Note
that this mechanism does not care about interface roughness. Its only prerequisite is the existence of complete AFM unit cells adjacent to the FM interface. Remarkably, the positive EB fields are systematically smaller by about 10% than the magnitudes of the negative ones. This appears reasonable when taking into account that the novel ME based coupling counteracts the conventional one for the case \( \mu_0 H_{it} > 0 \). In that case some uncompensated pinned spins, which are less tightly bound to the AFM bulk or even dangling, will rather follow the FM alignment and favor negative EB.

Let us first mention a fundamental test of our hypothesis, which states that merely the sign of the product \( H_{it}E_{it} \), and thus the kind of AFM domain, \( A \) or \( B \), decides the sign of \( H_{EB} \). To this end we have chosen all four sign combinations of the freezing fields with the magnitudes \( \left| \mu_0 H_{it} \right| = 0.6 \) T and \( \left| E_{it} \right| = 500 \) kV/m for MEFC from \( T = 350 \) to 298 K. Evaluation of the hysteresis loops measured at 298 K yields the following shifts: \( \mu_0 H_{EB}(+0.6 \) T, 500 kV/m) = + 17.3 mT, \( \mu_0 H_{EB}(-0.6 \) T, 500 kV/m) = + 17.5 mT, \( \mu_0 H_{EB}(+0.6 \) T, -500 kV/m) = - 19.8 mT, and \( \mu_0 H_{EB}(-0.6 \) T, -500 kV/m) = - 20.6 mT. Obviously, only the sign of \( H_{it}E_{it} \) counts for the sign of \( \mu_0 H_{EB} \).

Further we have tested our model by varying the strength of electric freezing field \( E_{it} \) as shown by a plot of \( \mu_0 H_{EB} \) as a function of \( E_{it} \) in Fig. 4. A virtually constant negative EB field, \( \mu_0 H_{EB} \approx - 20 \) mT, emerges after MEFC out of the positive remanent FM state from \( T = 350 \) to 298 K for any \( E_{it} \leq 0 \) in various magnetic freezing fields, \( \mu_0 H_{it} \approx 0.1, 0.3, \) and 0.6 T. Obviously, the formation of an A domain in \( E_{it} < 0 \) does not intensify the positive polarization of \( S_{AFM} \) compared to that obtained after MFC, i.e., in \( E_{it} = 0 \). This situation changes drastically when applying positive freezing fields, \( E_{it} > 0 \). At fairly low field values, \( E_{it} \leq 60 \) kV/m, \( \mu_0 H_{EB} \) changes from negative to positive values, which saturate at \( \mu_0 H_{EB} = 18 \) mT for \( E_{it} = 100 \) kV/m. The transition points, \( E_{fr} \), on the electric field axis shift to lower values as \( \mu_0 H_{it} \) is increased. This clearly hints at a competition between the two modes of field cooling: conventional MFC and MEFC as conjectured above.

In order to understand the inverse dependence of the threshold field \( E_{fr} \) on the magnetic freezing field, \( \mu_0 H_{it} \) (see enlarged plot of the transition curves in the upper inset in Fig. 4), we consider the energy of the AFM interfacial spins during the freezing process. First, we have to take into account the ME induced energy difference between domain types \( A \) and \( B \), \( W_{ME} \propto \alpha_{zz} \mu_0 H_{it} E_{it} \). Second, the magnetostatic Zeeman energy of the AFM interface spins, \( W_Z \approx - \mu_0 H_{it} S_{AFM} \), has to be considered. Third, the exchange interaction at the FM-AFM interface yields \( W_{EX} \propto - J S_{FM} S_{AFM} \), which is independent of \( H_{it} \) under the assumption of a single domain FM state. Hence, \( H_{EB} \) will vanish, if the ME energy \( W_{ME} \) is compensated by the MFC contributions \( W_Z \) and \( W_{EX} \),

\[
W_{ME} = W_Z + W_{EX}. \tag{2}
\]

Dividing both sides in Eq. (2) by \( \mu_0 H_{it} \) we readily obtain the electric threshold field,

\[
E_{fr} = c_1 + c_2/\mu_0 H_{it}, \tag{3}
\]

where \( c_1 \) and \( c_2 \) are fitting parameters. Indeed, as shown in the lower inset of Fig. 4 the experimental data \( E_{fr} \) vs
AFM single domain, we were able to switch the interface crystal. By inverting the electric freezing field and thus the consisting of a FM multilayer on top of a ME AFM single from high to low resistance values and vice versa. Procedures might be an interesting alternative to conventional power consumption, but simultaneously requires further tailoring magnetoresistive components with very low power consumption. For example, shifting the hysteresis loop from the electric field compensating the Zeeman contribution to EB field, \( H_{EB} \). This way remains invisible within the symbol widths of Fig. 1. Albeit being disappointingly small at the first glance, future use of all thin-film samples promises to improve the situation considerably. Similarly, it can be expected that the performance of the present switching mechanism might be further enhanced in proper thin-film samples, which then would promise applicability in future magnetoelectronic devices. For example, shifting the hysteresis loop from positive to negative magnetic fields by proper field-cooling procedures might be an interesting alternative to conventional current driven magnetic switching of spin valves from high to low resistance values and vice versa.

In summary, we investigated a new kind of EB system consisting of a FM multilayer on top of a ME AFM single crystal. By inverting the electric freezing field and thus the AFM single domain, we were able to switch the interface magnetization \( S_{AFM} \) and thus the EB field, \( H_{EB} \). This way of electrically controlling EB opens new possibilities for tailoring magnetoresistive components with very low power consumption, but simultaneously requires further research on ME EB systems. In the present case we propose single AFM unit cells to be related to the uncompensated pinned interface spins \( S_{AFM} \) discussed in the literature [17]. Their pinning mechanism remains unexplained. Further, it will be interesting if similar ME coupling can be realized with multiferroic materials.

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(\( \mu_0 H_\text{fr} \)) reveal linearity and are best fitted by \( c_1 = 8.63 \text{ kV/m} \) and \( c_2 = 5104 \text{ V}^2/\text{s/m}^3 \). The constant \( c_1 \), which formally equals \( E_0 \) in the limit \( H_\text{fr} \rightarrow \infty \), denotes the electric field compensating the Zeeman contribution to \( H_{EB} \) for large \( H_\text{fr} \).

Let us finally remark that the newly discovered switching effect due to magnetoelectric single domaining at the interface is giant compared to the ME shift of \( H_{EB} \) when applying an electric field to the AFM single domain after MEFC. Although a sizable magnetic moment arises in the bulk (Fig. 3, lines 2 and 3), only a minute change of \( S_{AFM} \) has been predicted [10] and experimentally confirmed [16]. For the present sample we have observed \( |\mu_0 \Delta H_{EB}| \approx 0.3 \text{ mT} \) for \( |E| = 500 \text{ kV/m} \) and \( T = 250 \text{ K} \), which remains invisible within the symbol widths of Fig. 1. Albeit being disappointingly small at the first glance, future use of all thin-film samples promises to improve the situation considerably. Similarly, it can be expected that the performance of the present switching mechanism might be further enhanced in proper thin-film samples, which then would promise applicability in future magnetoelectronic devices.

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