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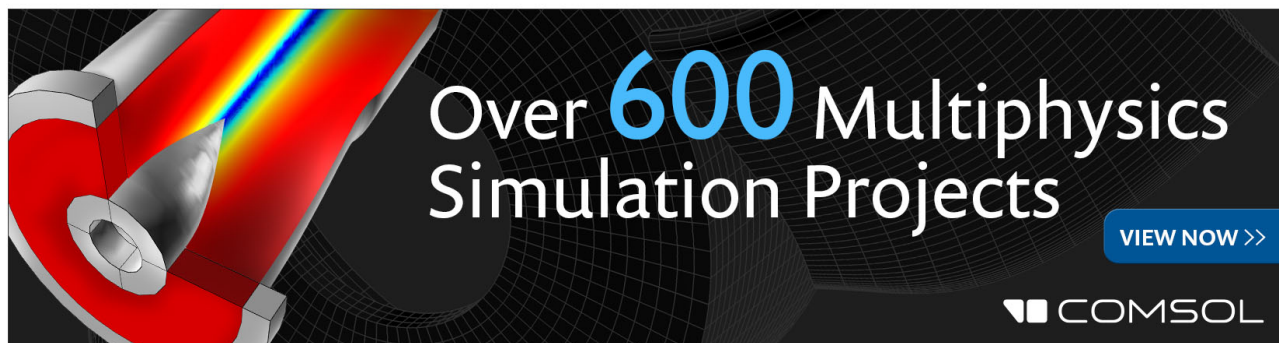
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Temperature- and magnetic-field-induced magnetization reversal in perovskite $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$

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Perovskite $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ exhibits magnetization reversal at low applied fields due to the competition between the single ion magnetic anisotropy and the antisymmetric Dzyaloshinsky–Moriya interaction. Below a compensation temperature (T_{comp}), a tunable bipolar switching of magnetization is demonstrated by changing the magnitude of the field while keeping it in the same direction. The present compound also displays both normal and inverse magnetocaloric effects above and below 260 K, respectively. These phenomena coexisting in a single magnetic system can be tuned in a predictable manner and have potential applications in electromagnetic devices. © 2011 American Institute of Physics. [doi:10.1063/1.3590714]

Magnetization reversal has attracted much attention in recent years, motivated both by interest in the fundamental physics as well as by the potential applications.^{1–4} As early as in 1948, Néel predicted that this phenomenon can occur in some ferrimagnetic materials such as spinel oxides,⁵ when the two antiferromagnetically coupled magnetic substructures exhibit a different temperature dependence of the magnetization. Recent studies have focused on some transition metal oxides with the perovskite structure, e.g., $\text{La}_{0.2}\text{Ce}_{0.8}\text{CrO}_3$, $\text{Sr}_2\text{YbRuO}_6$, and YbCrO_3 ,^{6–8} while the mechanisms of the magnetization reversal are different from that expected for ferrimagnetic systems. More interestingly, the coexistence of sign reversal of both magnetization and exchange bias field has been achieved in a single compound.^{6,7} Combining magnetization reversal effect with magnetoelectronics can exploit tremendous technological potential for device applications, for example, thermally assisted magnetic random access memories, thermomagnetic switches and other multifunctional devices, in a preselected and convenient manner. Unfortunately, such compounds are rare, and the low temperature operation is the main hindrance for their potential applications. Therefore, it is necessary to explore materials exhibiting magnetization reversal at higher temperature.

Very recently, the presence of magnetization reversal in $\text{BiFe}_{0.5}\text{Mn}_{0.5}\text{O}_3$ with high compensation temperature T_{comp} (the characteristic temperature where the material exhibits a net zero magnetization) of 208 K arouses much interest in this area.⁹ It highlights the possibility of designing materials with higher T_{comp} in perovskites with two transition metal magnetic ions at the *B* site. Actually, Azad *et al.*¹⁰ have reported the negative field-cooled (FC) magnetization below 210 K in $\text{LaFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ with the Fe and Cr cations randomly positioned at the *B* sublattice, whereas the origin of the magnetization reversal remains an open question. In order to explore materials exhibiting magnetization reversal at higher

temperature and understand the origin of negative magnetization in perovskites with two magnetic ions at the *B* site, the temperature- and magnetic-field-induced magnetization reversal in $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ was investigated in this letter. Perovskite $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ has the orthorhombic structure but does not display the 1:1 ordering of the Fe^{3+} and Cr^{3+} ions characteristics of the double perovskite. The observation of magnetization reversal in this system can be ascribed to the competition between the single ion magnetic anisotropy and the antisymmetric Dzyaloshinsky–Moriya (DM) interaction. Moreover, a bipolar switching of magnetization and both normal and inverse magnetocaloric effects (MCEs) were observed under low magnetic fields, indicating the potential applications of this material in spintronics.

$\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ ceramics were synthesized by the standard solid-state reaction. X-ray diffraction (XRD) powder patterns were collected using a Bede D¹ XRD unit with Ni-filtered $\text{Cu } K\alpha$ radiation at room temperature. Magnetic measurements were carried out using a vibrating sample magnetometer in a physical properties measurement system (PPMS-9T) of Quantum Design.

Figure 1 shows the experimental and calculated XRD patterns of $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$. No trace of any impurity phases is discernible for the studied sample. The XRD data were refined by the General Structure Analysis System Rietveld refinement program. The best fit of the observed diffraction peaks is obtained with the orthorhombic structure with space group *Pnma*, which is the same as that of the end members YFeO_3 and YCrO_3 .^{11,12} The calculated lattice parameters are $a=5.4987(1)$ Å, $b=7.9786(5)$ Å, and $c=5.5142(3)$ Å, respectively. The crystallographic data are similar to the neutron diffraction data as Azad *et al.*¹⁰ reported in $\text{LaFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$, which also did not show any signal of ordering between Fe^{3+} and Cr^{3+} cations in these perovskite oxides.

Figure 2 illustrates the temperature dependence of dc magnetization under FC mode at applied fields of ± 100 Oe. Intriguing magnetic behaviors were observed as the temperature decreases. Both these two curves exhibit the onset of

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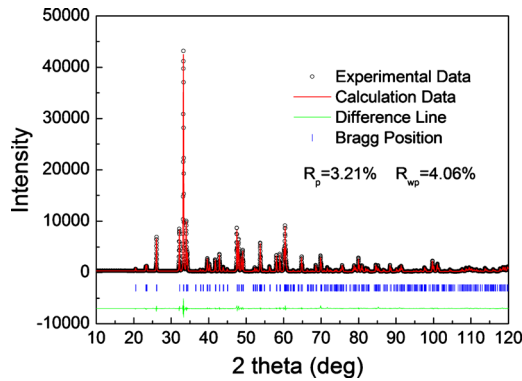


FIG. 1. (Color online) Results of Rietveld analysis of the XRD pattern for $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$. The observed and calculated patterns are denoted by open circles and solid lines, respectively. The solid lines at the bottom of the panel show the difference in the patterns. The Bragg peak positions are marked by vertical lines.

magnetic ordering at 274 K (see top inset of Fig. 2). On further cooling the compound under the external field +100 Oe, the magnetization increases, reaches a maximum at 260 K, then drops down and crosses the zero value of magnetization at $T_{\text{comp}}=248$ K. Afterward, the magnetization shows negative values, indicating that the direction of magnetization is against that of the applied field. On the other hand, a mirror image is seen if a field of -100 Oe is applied, namely, the magnetization at high temperature is negative and reverts to positive below T_{comp} . Compared to other compounds,^{1-10,13} $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ exhibits higher T_{comp} . T_{comp} decreases with increasing external field, and the negative magnetization is suppressed and vanishes at 3 kOe, as shown in Fig. 4(a). It suggests that the net magnetic moments initially in the opposite direction of the field are realigned in the same direction as the specimen in a higher magnetic field.

To understand the origin of magnetization reversal in $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$, it is essential to ascertain the magnetic ground state. The isothermal magnetization $M(H)$ at 5 K is depicted in the bottom inset of Fig. 2. There exhibits an obvious hysteresis loop between ± 2 kOe. No saturation is found even up to 50 kOe, instead the magnetization increases linearly

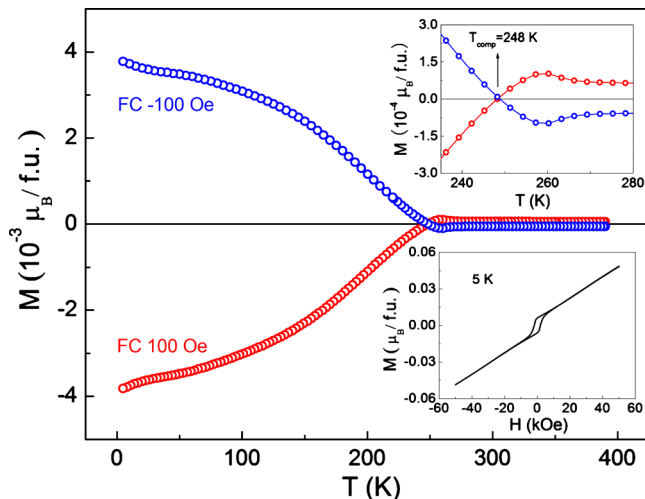


FIG. 2. (Color online) Magnetization as a function of temperature measured at $H = \pm 100$ Oe under FC condition for $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$. Insets show the compensation temperature (T_{comp}) and the isothermal magnetization at 5 K for top and bottom panels, respectively.

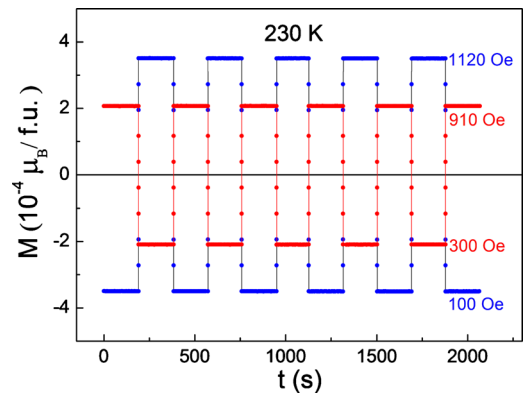


FIG. 3. (Color online) Switching of magnetization between negative and positive values at 230 K under various magnetic fields.

in the region of higher magnetic field. This kind of magnetization loop and the linear dependence at high field are usually attributed to the weak ferromagnetism caused by a departure from the collinearity of the moments in an antiferromagnet.^{8,12,13} The high field part of $M(H)$ curves can be represented as $M = \chi_{\text{AFM}}H + \sigma_S$, where $\chi_{\text{AFM}}H$ is the antiferromagnetic (AFM) contribution and σ_S is the saturation magnetization of the weak ferromagnetism. Thus, the ferromagnetic component in $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ can be obtained by subtracting the AFM contribution from the total magnetization. The remanent magnetization M_r is $0.0055 \mu_B/\text{f.u.}$ and the coercive field H_c is 2.2 kOe.

As discussed above, $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ shows AFM ordering with weak ferromagnetism and the Fe^{3+} and Cr^{3+} ions are distributed randomly at the B site. Evidently, the model proposed by Néel for ferrimagnetic systems is not applicable to the present compound. Normally, there are two mechanisms giving rise to a canting of the AFM alignment of moments: the single ion magnetic anisotropy and the antisymmetric DM interaction.^{14,15} YFeO_3 is a canted antiferromagnet with weak ferromagnetism due to DM interaction,¹¹ while in orthochromites the Cr^{3+} ions exhibit the single ion magnetic anisotropy arising from the effect of $t-e$ hybridization owing to the local site distortion and cooperative octahedral-site rotation.¹⁶ Hence, the substitution of Cr at the Fe site brings out the competition between the single ion magnetic anisotropy and the antisymmetric DM interaction. Noting the equal proportion of Fe and Cr in $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$, such competition is more prominent. As reported by Kadomtseva et al.,¹⁷ the net moments produced by these two factors are in opposite direction and they have different temperature dependence of magnetization. Consequently, as temperature decreases negative magnetization takes place at low fields. If the applied field is large enough, the average magnetic anisotropy is overcome by the energy induced by the field, the net magnetic moment is orientated parallel to the field direction, and the negative magnetization disappears. This can be demonstrated clearly by a tunable bipolar switching of magnetization at a given temperature below T_{comp} .

Figure 3 shows the switching of FC magnetization between negative and positive values in various magnetic fields at 230 K. The sample was cooled to 230 K under 100 Oe and then the external field was increased to 1120 Oe in order to switch the magnetization to a positive value of equal magnitude. The magnetization polarity switching can be cycled several times and shows good reproducibility of this field-

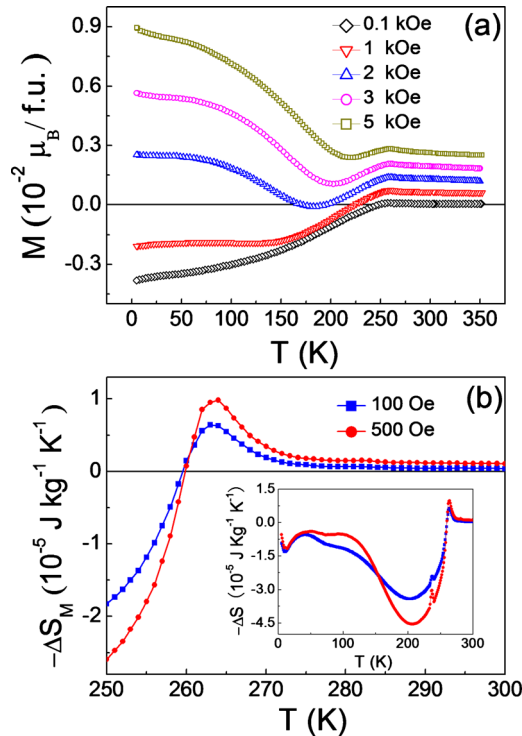


FIG. 4. (Color online) (a) FC magnetization versus temperature at different applied fields for $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$. Above 20 kOe, no magnetization reversal exists any more. (b) Magnetic entropy change (ΔS_M) as a function of temperature ($250 \leq T \leq 300$ K) in 100 and 500 Oe fields. Insert shows the ΔS_M - T curves in a reduced scale.

induced magnetization reversal. This phenomenon is also observed by varying the field to obtain other magnetization values, for example, as per sequence of 300 Oe—910 Oe—300 Oe—... Once the external field is switched off, the corresponding magnetization state ceases to exist rapidly without any noticeable decay in their values. As shown in Fig. 3, the alternation of the positive and negative magnetization states can be actuated by only changing the magnitude of the field while keeping it in the same direction. It can be tuned in a predictable way. This behavior can find application in magnetic data storage and switches devices such as nonvolatile magnetic memory, which facilitates two distinct states of magnetization. Compared with similar demonstration in a Prussian blue type molecular magnet and oxide $\text{BiFe}_{0.5}\text{Mn}_{0.5}\text{O}_3$,^{4,9} $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ displays a bipolar switching of magnetization at higher temperature.

The temperature-induced normal and inverse MCEs were also observed at low magnetic fields. According to the magnetic entropy change ΔS_M of the material with the variation in temperature and an external magnetic field, we can obtain ΔS_M from the temperature dependence of magnetization (M - T) at different fields, $\Delta S_M = \sum [(\partial M / \partial T)_{H_i} + (\partial M / \partial T)_{H_{i+1}}](1/2)\Delta H_i$, where $(\partial M / \partial T)_{H_i}$ and $(\partial M / \partial T)_{H_{i+1}}$ are the experimental values obtained from the M - T curves in magnetic fields H_i and H_{i+1} , respectively.¹⁸ As a direct consequence of the change in the sign of $\Delta M / \Delta T$,⁴ the polarity reversal of $-\Delta S_M$ can be observed in Fig. 4(b). The increase in $-\Delta S_M$ emerges at the onset of magnetic ordering temperature 274 K, and then it reaches a maximum at 262 K, where the $-\Delta S_M$ values start to decrease. On cooling below 262 K, the decrease in $-\Delta S_M$ continues and it crosses the $-\Delta S_M = 0$ axis at 260 K. Then, $-\Delta S_M$ moves to negative values (i.e.,

the inverse MCE) with further lowering the temperature. Obviously, an extraordinary coexistence of normal and inverse MCEs has been realized in $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$. It is well known that $-\Delta S_M > 0$ results in cooling in the conventional magnetic refrigeration cycle while $-\Delta S_M < 0$ induces a heating under the same refrigeration cycle. This phenomenon can be applied to a constant temperature bath proposed by Yusuf *et al.*⁴ If $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ is used as the working medium, at $T > 260$ K, the normal MCE will cause the temperature of the system to decrease, while at $T < 260$ K, the temperature of the system will increase due to the inverse MCE. Accordingly, in the vicinity of 260 K, any temperature fluctuation will be balanced by these two opposite MCEs. Therefore, a magnetic cooling/heating based constant temperature bath of 260 K can be realized in $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$.

In conclusion, we have demonstrated that perovskite $\text{YFe}_{0.5}\text{Cr}_{0.5}\text{O}_3$ exhibits magnetization reversal arising from the competition between the single ion magnetic anisotropy and the antisymmetric DM interaction. A tunable bipolar switching of magnetization and both normal and inverse MCEs at low applied fields were observed in this system above 230 K whereas the room temperature operation has not been realized yet. However, the present work shows a promising way of designing materials exhibiting magnetization reversal at higher temperature and proposes the potential applications of this effect in electromagnetic and magnetocaloric devices, such as nonvolatile memories and magnetic cooling/heating based constant temperature bath. To explore materials exhibiting magnetization reversal at room temperature, further theoretical and experimental work is needed.

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