Room-temperature giant magnetotranstance effect in single-phase multiferroics

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Single-phase multiferroic materials are usually considered useless because of the weak magnetoelectric effects, low operating temperature, and small electric polarization induced by magnetic orders. As a result, current studies on applications of the magnetoelectric effects are mainly focusing on multiferroic heterostructures and composites. Here we report a room-temperature giant effect in response to external magnetic fields in single-phase multiferroics. A low magnetic field of 1000 Oe applied on the spin-driven multiferroic hexaferrites BaSrCo\textsubscript{2}Fe\textsubscript{11}AlO\textsubscript{22} and Ba\textsubscript{0.9}Sr\textsubscript{1.1}Co\textsubscript{2}Fe\textsubscript{11}AlO\textsubscript{22} is able to cause a huge change in the linear magnetoelectric coefficient ($\alpha_E = \frac{dE}{dH}$) by several orders, leading to a giant magnetotranstance (GMT) effect at room temperature. The GMT effect is comparable to the well-known giant magnetoresistance (GMR) effect in magnetic multilayers, and thus opens up a door toward practical applications for single-phase multiferroics.

multiferroic, magnetoelectric effect, transtor

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1 Introduction

In electric circuits, voltage ($v$), current ($i$), charge ($q$), and magnetic flux ($\phi$) are four basic circuit variables. The relationships between them have defined three fundamental circuit elements—the resistor with resistance $R=\frac{dv}{di}$, the capacitor with capacitance $C=\frac{dq}{dv}$, and the inductor with inductance $L=\frac{dq}{di}$, as shown in Figure 1(a). The fourth element defined from the $q-\phi$ relationship was named transtor with transtance $T=\frac{dq}{d\phi}$ [1], which can be implemented by employing the magnetoelectric (ME) effects, i.e., the mutual control of magnetization ($M$) by electric fields ($E$) and electric polarization ($P$) by magnetic fields ($H$) [2-5], as illustrated in Figure 1(b). Each circuit element has a corresponding nonlinear memelement, including the memristor, memcapacitor, meminductor, and memtranstor. Especially, the memristor and memtranstor have been under intensive studies due to their promise in developing next-generation low power electronic devices such as nonvolatile memory, logic gates, and artificial neurons [6-12].

It is well known that the resistance of a resistor may strongly depend on applied DC magnetic fields, yielding various magnetoresistance (MR) effects. For instance, the resistances of some magnetic multilayers and tunneling junctions are very sensitive to external magnetic fields, resulting in the GMR effect [13,14]; a moderate magnetic field...
of several Tesla can greatly suppress the resistance by several orders in some perovskite manganites, known as the colossal magnetoresistance (CMR) effect [15,16]; some topological orders in some perovskite manganites, known as the colossal...sis not only provide a playground for fundamental sciences but also serve as a basis for many technological applications.

Similarly, the physical quantities of a transistor may also exhibit a notable dependence on external magnetic fields, which would give rise to the magnetotransistance (MT) effects. However, this subject has not been explored yet. In this paper, we report the discovery of a GMT effect at room temperature in single-phase magnetoelectric multiferroics. Therefore, we used these samples to test the magnetic field dependence of $\alpha_E$ at room temperature.

Polycrystalline samples of BaSrCo$_{2}$Fe$_{11}$AlO$_{22}$ and Ba$_{0.9}$Sr$_{1.1}$Co$_{1.1}$Fe$_{11}$AlO$_{22}$ were synthesized by the conventional solid-state reaction method [24]. Stoichiometric amounts of SrCO$_3$, BaCO$_3$, Co$_3$O$_4$, Fe$_2$O$_3$, and Al$_2$O$_3$ powders were mixed and calcined at 1000°C for 12 h in air. Then, the calcinated powders were reground and pressed into pellets, and sintered at 1200°C for 12 h in oxygen atmosphere. A post-annealing at 900°C for 72 h in a flow of oxygen was performed in order to reduce the oxygen vacancies and make the samples highly insulating. The samples were checked by powder X-ray diffraction (XRD) at room temperature. As shown in Figure S1 in Supporting Information, all the main diffraction peaks can be indexed to the Y-type hexagonal phase. The XRD results confirm that all the prepared samples are clean single-phase Y-type hexaferrites without detectable impurities.

The magnetic properties were measured by using a Magnetic Properties Measurement System (PPMS, Quantum Design). The samples were cut into thin plates with the typical size of 3 mm×5 mm×0.2 mm for electrical property measurements. Silver paste was painted on two surfaces of the samples to make electrodes. The dielectric permittivity and ME current were measured in a Cryogen-free Superconducting Magnet System (Oxford Instruments, Teslatron PT) using an LCR meter (Aglient 4980A) and an electrometer (Keithley 6517B), respectively. A poling procedure is
necessary in order to measure the ME current. First, an electric field (1 MV/m) and a magnetic field (50 kOe) perpendicular to each other were applied on the samples. Then, the magnetic field was reduced to 5 kOe to enter into the ferroelectric phase. After electric field was removed and the samples were short-circuited for 30 min, the ME currents were measured by sweeping $H$ from 5 kOe down to $-50$ kOe. The value of electric polarization was obtained by integrating the ME current with time.

The linear ME coefficients $\alpha_E$ was measured by a dynamic method as described in ref. [11]. The sample is placed in the middle of the solenoid. An AC current source (Keithley 6221) is connected to the solenoid to generate a small AC magnetic field ($\sim 2$ Oe). The induced $V_{ME}$ on the sample is detected by a lock-in amplifier (Stanford Research SR830). The probe is loaded in an Oxford TeslatronPT superconducting magnet system which provides the DC bias magnetic field.

### 3 Results and discussion

Figure 2(a) and (c) show magnetic-field control of electric polarization at 300 K for two samples, respectively. One unique feature of these Y-type hexaferrites is that the spin-induced electric polarization can be rapidly reversed by a low magnetic field due to the rotation of conical spin structure with an applied magnetic field [19,20]. This leads to the maximum of the ME effect occurring around zero magnetic field. The amplitude of $P$ is about 3 $\mu$C/m$^2$ at room temperature for both samples, which is three to four orders less than that of traditional ferroelectrics. Therefore, these single-phase multiferroics with room-temperature ME effects are generally considered useless. Figure 2(b) and (d) show the $M$-$H$ loops at 300 K. These hexaferrites are soft magnets with a very low coercivity, which is pivotal to the low-field ME effect.

We then focused on the linear ME coefficient $\alpha_E$ of two samples. The measurement configuration is shown in the inset of Figure 3. Both the DC and AC magnetic fields are applied perpendicular to the electric field, i.e., in the transverse configuration, because the spin-induced electric polarization in these hexaferrites is vertical to the applied magnetic field. The amplitude of $\alpha_E$ depends on the frequency of applied AC magnetic field and a broad maximum around 130 Hz is observed, which could be related to the mechanical-resonance enhancement of the ME effect [28].

Figure 4(a) and (b) show $\alpha_E$ as a function of applied in-plane DC bias magnetic field for two hexaferrite samples, respectively. For the BaSrCo$_{2}$Fe$_{11}$AlO$_{22}$ sample, $\alpha_E$ decreases rapidly from 27 mV/(cm Oe) at zero field to 0.24 mV/(cm Oe) at 1 kOe and 0.04 mV/(cm Oe) at 2 kOe. For the Ba$_{0.9}$Sr$_{1.1}$Co$_{11}$Fe$_{11}$AlO$_{22}$ sample, $\alpha_E$ drops from 44 mV/(cm Oe) at zero field to 0.52 mV/(cm Oe) at 1 kOe.

**Figure 2** (Color online) Magnetic field reversal of electric polarization at 300 K in (a) BaSrCo$_{2}$Fe$_{11}$AlO$_{22}$ and (c) Ba$_{0.9}$Sr$_{1.1}$Co$_{11}$Fe$_{11}$AlO$_{22}$; (b) The $M$-$H$ loop at 300 K for two samples, respectively. The arrows indicate the direction of sweeping magnetic field.
and 0.09 mV/(cm Oe) at 2 kOe. In other words, a low magnetic field of 2 kOe is able to cause a huge change in $\alpha_E$ by three orders of magnitude.

In comparison, the magnetic field dependence of the dielectric permittivity ($\varepsilon_r$) at 300 K is presented in Figure 4(c) and (d). $\varepsilon_r$ decreases slightly with increasing magnetic field. For BaSrCo$_2$Fe$_{11}$AlO$_{22}$, a high magnetic field of 50 kOe causes a relative change $\sim$2.8% and a low magnetic field of 2 kOe only induces a change of 0.8% in $\varepsilon_r$. Similarly, for Ba$_{0.9}$Sr$_{1.1}$Co$_{2}$Fe$_{11}$AlO$_{22}$, the relative change in $\varepsilon_r$ is $\sim$ 2.2% at 50 kOe and 0.5% at 2 kOe. In fact, the quantity of capacitance (or dielectric permittivity) is generally insensitive to external magnetic fields so that a large magnetocapacitance (or magnetodielectric) effect can hardly be observed. In strong contrast, the ME coefficient is much more sensitive to a magnetic field than the capacitance does.

The MT ratio, defined as $MT = [\alpha_E(H) - \alpha_E(0)]/\alpha_E(0) \times 100\%$, is plotted in Figure 5(a) and (b) for two samples, respectively. For BaSrCo$_2$Fe$_{11}$AlO$_{22}$, the MT ratio is as high as 67% at 100 Oe, 96.5% at 500 Oe, and 99% at 1000 Oe. Similarly, for Ba$_{0.9}$Sr$_{1.1}$Co$_2$Fe$_{11}$AlO$_{22}$, the MT ratio reaches 62% at 100 Oe, 95.7% at 500 Oe, and 98.8% at 1000 Oe. We also measured the MT behavior with different frequencies (50 Hz and 1 kHz) of the AC magnetic field for BaSrCo$_2$Fe$_{11}$AlO$_{22}$. As seen in Figure S2 in Supporting Information, although the absolute value of $\alpha_E$ relies on the frequency of the AC magnetic field, the GMT effect at room temperature was always observed regardless of the frequency. This giant effect at room temperature is comparable to the well-known GMR effect in magnetic multilayers, and thus can be directly used for magnetic sensors to probe low magnetic fields in many circumstances.

The high sensitivity of the ME coefficient $\alpha_E$ in response to the DC bias magnetic field is related to the sharp reversal
4 Conclusions

In summary, the fourth circuit element “transistor” defined from $T=\frac{dq}{d\phi}$ is characterized by the linear ME coefficient $\alpha_E=\frac{dE}{dH}$ which reflects the ability of a material to convert an external magnetic field into an electric field. It is found that the $\alpha_E$ of some single-phase multiferroic materials is very sensitive to external DC magnetic fields, giving rise to a GMT effect at room temperature. This phenomenon not only holds a promise for technological applications such as magnetic sensors but also opens up a fresh field in condensed matter physics. The variation of $\alpha_E$ of a material or device in response to external stimuli such as temperature, magnetic and electric fields, pressure, and light radiation, may give rise to many interesting new phenomena, and would deserve more research efforts in the future.

process of the conical spin structure in multiferroic Y-type hexaferrites [19-24]. As seen in Figure 2, these hexaferrites are very soft magnets with a low magnetic coercivity. Upon the reversal of magnetization by a small field, the spin-induced electric polarization is also reversed. The ME coefficient is pronounced in the vicinity of zero field because the spin-induced electric polarization changes most fast around zero field (i.e., $dP/dH$ is maximum) and then changes slightly with increasing magnetic fields. It is worthy to note that in some hexaferrites the spin-induced electric polarization does not reverse with applied magnetic fields [24]. Then, the ME coefficient $\alpha_E$ would be minimum rather than maximum around zero field. Although the spin-induced electric polarization is tiny and the absolute value of $\alpha_E$ is not as high as that of magnetoelectric composites [28], the MT ratio is remarkable because $\alpha_E$ is very sensitive to external magnetic fields. This phenomenon will totally change the viewpoint on single-phase multiferroics. A large electric polarization and a high ME coefficient are not necessarily required for applications but the sensitivity of the ME coefficient to external fields is more important. In previous studies, a large change of the ME voltage coefficient $\alpha_E$ with applied DC magnetic fields was ever observed in some magnetoelectric composites [8,29]. Nevertheless, little attention has been paid to the magnetotransistance effect in single-phase magnetoelectric multiferroics. Our study represents the first example of GMT effect in the low field regime at room temperature in single-phase multiferroics. Moreover, diverse MT effects could exist in many magnetoelectric materials, especially in spin-driven multiferroics.

Figure 5 (Color online) The MT ratio defined as $MT=\frac{\alpha_E(H)-\alpha_E(0)}{\alpha_E(0)} \times 100\%$ at room temperature for (a) $\text{BaSrCo}_2\text{Fe}_5\text{AlO}_{12}$ and (b) $\text{Ba}_0.9\text{Sr}_1.1\text{Co}_2\text{Fe}_5\text{AlO}_{12}$. The insets show a zoom-in view in the low field range.

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Supporting Information

The supporting information is available online at phys.scichina.com and link.springer.com. The supporting materials are published as submitted, without typesetting or editing. The responsibility for scientific accuracy and content remains entirely with the authors.

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