

Effect of electric field on resistivity of a material has attracted considerable attention due to its rich physics as well as great potential in developing electronic devices. A large change in resistivity upon applied electric field, usually termed as the electroresistance effect, has been observed in a variety of transition metal oxides such as perovskite manganites, titanates, and magnetite. Although the underlying physical mechanism could be distinct from each other, these effects are believed to be intrinsic properties of the materials, in contrast to those extrinsic electroresistance observed in some metal-insulator-metal structures where the interfacial effects play a dominating role. In most cases, these intrinsic electroresistance effects happen at quite low temperatures and require high electric fields, which are apparent drawback for practical applications. Here we show that an intrinsic colossal electroresistance effect occurs around room temperature in LuFe$_2$O$_4$.

The mixed-valence material LuFe$_2$O$_4$ belongs to the family of the rare earth-iron oxide RFe$_2$O$_4$, where R stands for rare-earth elements from Dy to Lu and Y. It has a rhombohedral crystal structure consisting of alternate stacking of triangular lattices of rare-earth elements, iron and oxygen ions. As the Fe ions have an average valence of 2.5+, an equal amount of Fe$^{2+}$ and Fe$^{3+}$ ions coexist at the same site in the triangular lattice. The strong magnetic interactions between localized Fe moments develop as a two-dimensional (2D) ferromagnetic ordering below the Neel temperature $T_N \sim 250$ K. The system also involves a charge interaction between Fe$^{2+}$ and Fe$^{3+}$ ions. Compared with the average Fe valence of 2.5+, Fe$^{2+}$ and Fe$^{3+}$ ions are considered as having an excess and a deficiency of half an electron, respectively. Thus the coulombic interactions between Fe$^{2+}$ and Fe$^{3+}$ are accompanied by a charge frustration on the triangular lattice. The competing interactions between frustrated charges are settled by a $\sqrt{3} \times \sqrt{3}$ superstructure of Fe$^{2+}$ and Fe$^{3+}$ arrangements, similar to the stable configuration of Ising spins in a triangular lattice. As a consequence, LuFe$_2$O$_4$ exhibits a 2D charge ordered (CO) state below 330 K. Above 330 K, the 3D charge ordering transforms into a 2D ordering state. Spectacularly, the charge ordering in LuFe$_2$O$_4$ leads to a local electrical polarization because the centers of Fe$^{2+}$ and Fe$^{3+}$ do not coincide in the unit cell of the superstructure. Such kind of ferroelectricity associated with charge ordering is termed as "electronic ferroelectricity," which is in contrast to conventional ferroelectricity involving displacement of cation and anion pairs. The CO state in many systems could be melted by external forces such as magnetic field, electric field, and pressure, which generally leads to an insulating to metallic transition. For example, both the CO perovskite manganites and magnetite (Fe$_3$O$_4$) exhibit an electrically driven phase transition below the charge-ordering temperature $T_{CO}$. Considering the high $T_{CO}$ in LuFe$_2$O$_4$, one may expect a similar phase transition induced by electric field at relatively high temperatures.

The resistance and $I$-$V$ measurements were performed on polycrystalline LuFe$_2$O$_4$ samples using the circuit shown in the inset of Fig. 1. The details of sample preparation can be found in Ref. 17. The applied voltage pulse was supplied by a Keithley 2400 source meter up to 80 V. To protect the circuit against a burst of big current, the current limit is set to 100 mA. The typical sample has a size of $3.8 \times 1 \times 1$ (length $\times$ width $\times$ thickness) mm$^3$. An electrode was 

![FIG. 1. (Color online) Temperature dependence of resistance of LuFe$_2$O$_4$ measured in various applied voltages. The inset shows the measurement circuit. The applied voltage refers to the voltage supplied by the source meter.](image-url)
The resistance was thus measured using the conventional four-electrodes attached to the surface with a space of 1 mm. The voltage drop across the sample was monitored using two other electrodes made at each end of the sample using silver paint. The voltage drop across the sample was monitored using two other electrodes made at each end of the sample using silver paint. The voltage drop across the sample was monitored using two other electrodes made at each end of the sample using silver paint. The voltage drop across the sample was monitored using two other electrodes. The sudden jump of current at a threshold voltage decreases nearly exponentially with the increasing temperature so that a small voltage of a few volts is threshold voltage. We have confirmed that the temperature increase was less than 0.8 K. Moreover, we used voltage pulses and found that the temperature increase was less than 0.8 K. When the voltage is scanned back from the negative to the positive, a symmetric variation was observed. The relative

Figure 1 shows the temperature dependence of resistance of LuFe$_2$O$_4$ under various applied voltage pulses. The resistance was measured with a conventional four-probe configuration (see the inset of Fig. 1). In a low applied voltage (0.3 V), the resistance shows an insulating behavior with an apparent kink around 330 K where the 3D to 2D charge order transition occurs. In higher applied voltages, the resistance is nearly the same at low temperature. However, as temperature increases, an abrupt drop of resistance takes place, which results in a burst of current exceeding the current limit of 100 mA and makes the lower resistance undetectable. With increasing applied voltage, the abrupt change in resistance starts at lower temperature. These sharp resistance drops indicate that the applied voltages may induce an insulating-metallic transition at high temperatures.

Figure 2 shows the current-voltage ($I$-$V$) curves at various temperatures. These curves were measured by scanning pulsed voltage up to 80 V. The width of each pulse is 20 ms. To protect the circuit against a burst of big current, the current limit is set to 100 mA. At 240 K, the $I$-$V$ curve is close to linearity up to 80 V. Above 260 K, each $I$-$V$ curve deviates from the linearity and exhibits a big jump of current at a threshold voltage $V_{th}$. As shown in the inset of Fig. 2, the threshold voltage decreases nearly exponentially with the increasing temperature so that a small voltage of a few volts is able to cause the transition at 340 K. We have confirmed that the sharp transitions are not due to Joule heating. We monitored the temperature of the sample during the voltage scan and found that the temperature increase was less than 0.8 K. Moreover, we used voltage pulses ($\sim 0.02$ s) with 1 s pause between pulses in the measurements so that the Joule heating is limited to a tiny effect. These highly nonlinear $I$-$V$ curves further prove the point that an insulating-metallic transition can be induced by applied electric fields in LuFe$_2$O$_4$.

In order to better evaluate the electrically driven insulating-metallic transition, we measured the $I$-$V$ hysteresis loop at 300 K. The pulsed voltage is scanned with a step of 0.8 V and 1 s pause between pulses. As shown in Fig. 3(a), the $I$-$V$ loop looks quite symmetric with positive and negative voltages. The initial ascending branch, marked with 1, gives a threshold voltage \( \sim 22 \) V at which the current suddenly jumps up and exceeds the 100 mA limit. When the voltage is scanned back, the current remains undetectable till \( \sim 10 \) V and drops fast thereafter, giving a large hysteresis. When the voltage is scanned to negative, the switching phenomenon repeats at nearly the same threshold voltages. The final branch with increasing voltage, marked with 2, is slightly different from the initial branch, showing a smaller threshold voltage.

Figure 3(b) shows the resistance of LuFe$_2$O$_4$ as a function of the cycle of applied voltage at 300 K. We note that the resistance is obtained using the four-terminal configuration, i.e., calculated using the voltage drop between the voltage leads. The resistance decreases continuously with the increasing voltage, which suggests that the nonlinearity in the $I$-$V$ curves starts well before the switching. At the threshold voltage, the resistance becomes undetectable because the current exceeds the limit of 100 mA. When the voltage is scanned back, the resistance becomes detectable only below \( \sim 10 \) V and does not return to the initial value at zero voltage. The resistance increases further as the voltage is scanned to negative and reach a maximum at \( \sim -10 \) V. Then it decreases smoothly with the increasing negative voltage and becomes undetectable at the negative threshold voltage. When the voltage is scanned back from the negative to the positive, a symmetric variation was observed. The relative

FIG. 2. (Color online) Current-voltage ($I$-$V$) curves at various temperatures. The measurements were performed by scanning pulsed voltage up to 80 V using the circuit shown in Fig. 1. The sudden jump of current at a threshold voltage $V_{th}$ indicates an insulating-metallic transition. The inset shows the temperature dependence of $V_{th}$.
change in resistance \( (R/R_0) \) as a function of applied voltage at various temperatures is shown in Fig. 4. \( R_0 \) is the initial resistance at the lowest voltage. The relative ratio is nearly 100\% at the threshold voltages as the resistance changes by several orders, which is really a colossal electroresistance effect. We note that the measurements were performed on a bulk sample with a 3.8 mm length. Thus, the threshold voltage (~22 V) at room temperature corresponds to a quite small electric field of tens V/cm, which is beneficial for practical applications.

The electrically driven phase transition observed in LuFe\(_2\)O\(_4\) has some common features with that in the CO Pr\(_{1−x}\)Ca\(_x\)MnO\(_3\) manganites and magnetite (Fe\(_3\)O\(_4\)), but occurs at much higher temperatures and requires much smaller electric fields. It has been long believed that the resistive switch in manganites is due to the melting of the CO state triggered by applied electric field.\(^4\,16\,17\) Since LuFe\(_2\)O\(_4\) is also a CO system, we think that a similar mechanism could apply. In the CO state, the charge carriers are initially localized at each atomic site due to the repulsive electron-electron interaction. A high enough electric field may cause the dielectric breakdown of the CO state, which immediately leads to a large number of mobile charge carriers and, consequently, a metallic state appears. The fact that the threshold voltage decreases exponentially with the increasing temperature implies that thermally assisted fluctuations make the melting of the CO state much easier. Therefore, with the benefit of a high \( T_{CO} \) above room temperature in LuFe\(_2\)O\(_4\), a small electric field is able to break down the CO state in the vicinity of room temperature and induces an insulating-metallic transition.

The breakdown of the CO state by electric field can be evidenced by the dielectric response measurements. Since the electronic ferroelectricity in LuFe\(_2\)O\(_4\) is associated with charge ordering of Fe\(^{2+}\) and Fe\(^{3+}\) ions,\(^1\) the breakdown of the CO state would subsequently result in a suppression of the electrical polarization, which should be reflected in the dielectric response. In fact, this has been confirmed by our recent dielectric tunability experiments.\(^17\) Similarly, a strong suppression of the low-frequency dielectric constant by bias electric field was recently observed in the CO state of Pr\(_{1−x}\)Ca\(_x\)MnO\(_3\), which also exhibits a colossal electroresistance effect at low temperature.\(^18\) The authors considered it as an evidence for the melting of the polaron ordering.

We also examined the influence of magnetic field on the resistance and \( I-V \) characteristics. Unlike perovskite manganites where magnetic field has a strong action,\(^4\,5\) we found that the magnetoresistance effect in LuFe\(_2\)O\(_4\) is very small even in a 10 T magnetic field. It seems that the electronic properties of LuFe\(_2\)O\(_4\) are very sensitive to electric field but quite dull to magnetic field. The physics underlying this peculiar feature deserves further studies. Combining the multiple virtues of room temperature and colossal effects in low applied fields, LuFe\(_2\)O\(_4\) is not only a very promising material for many applications but also provides a playground for intriguing physics of strongly correlated electrons.

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