Nanoscale spin-dependent transport in a weak itinerant ferromagnet BaIrO$_3$

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Abstract. We have performed magnetoresistance (MR) measurements for a BaIrO$_3$ single crystal. As the magnetic field changes from 0 T to 14 T and back to 0 T, the longitudinal MR at 1.5 K traces out a butterfly-shaped hysteresis curve and the sign of the MR ratio for the field-ascending sweep changes from negative to positive near 6 T. The MR ratios for field-ascending and descending sweeps overlap each other around 13 T. A large anisotropic magnetoresistance (AMR) ratio per $\mu_B$, $(\rho_\parallel - \rho_\perp)/\rho_0 \sim 400\%$/$\mu_B$ at 1.5 K, is obtained, where $\rho_\parallel$, $\rho_\perp$, and $\rho_0$ are the longitudinal resistivity, the transverse resistivity, and the zero-field resistivity. The MR effects are interpreted based on a spin valve model considering a novel crystal structure and a local magnetic-anisotropy related to the spin-orbit coupling of this compound.

1. Introduction

Strongly correlated electronic systems are presently most extensively studied subjects in condensed matter physics. Especially, 3$d$ transition-metal oxides, such as high-$T_c$ superconductive cuprates, colossal magnetoresistive (CMR) manganites, and multiferroic materials, have attracted much attention because of their unique physical properties which come from competing and/or cooperating of multi-degrees of freedom (spin, orbital, charge, and lattice) [1]. The 3$d$ states in these oxides are well-localized, yielding strongly correlated narrow bands with a large on-site Coulomb repulsion $U$ and a small band width $W$. While 4$d$ and 5$d$ states in their transition-metal oxides are delocalized due to largely reduced $U$ and widened $W$ [2]. Therefore, in 4$d$ and 5$d$ transition-metal oxides, different physical phenomena from those in the 3$d$ systems are expected.

The 5$d$ transition-metal oxide BaIrO$_3$ has a monoclinic crystal-structure (space group C2/m) and features three face-sharing IrO$_6$ octahedra forming Ir$_3$O$_{12}$ trimers that are vertex linked to construct a zigzag chain along the $c$-axis [3]. The top and bottom triangle faces of each trimer are parallel to the $ab$-plane, forming the two-dimensional layers of corner-shared IrO$_6$ octahedra. Due to the peculiar crystal-structure, anisotropic properties are observed in electrical resistivity, magnetization, and thermoelectric power [4, 5]. BaIrO$_3$ exhibits a ferromagnetic transition with a small Ir$^{4+}$ moment of 0.04 $\mu_B$ at $T_c \sim 180$ K accompanying with a gap opening at the Fermi level. Below $T_c$, a hump-type anomaly corresponding to the gap opening in the density of state is observed in the electrical resistivity [4, 5]. In spite of the ferromagnetic ground state, the ordered
state below \( T_c \) was believed to be a CDW state. Thus, we expect that the coexistence of the spontaneous magnetization and the energy gap will lead to unprecedented physical properties in \( \text{BaIrO}_3 \). In the present study, we have performed magnetoresistance (MR) measurements in longitudinal and transverse geometries for the single crystal of \( \text{BaIrO}_3 \) to investigate the relationship between magnetic and transport properties of it.

2. Experimental

For this study, \( \text{BaIrO}_3 \) single crystals with a typical dimension of \( 1 \times 0.5 \times 0.2 \text{ mm}^3 \) were synthesized by the molten flux method using \( \text{BaCl}_2 \) as a solvent [5]. The MR measurements were performed by using a conventional dc four-probe technique in a 16 T superconducting magnet at 1.5 K. The electrical current was applied along the direction parallel to the \( c \)-axis of the sample. The applied magnetic field directions were parallel (\( B \parallel I \)) and perpendicular (\( B \perp I \)) to the current direction, corresponding to the configurations of longitudinal (\( \rho_\parallel \)) and transverse (\( \rho_\perp \)) MRs, respectively. The contribution from antisymmetrical part (Hall resistance) was carefully removed by subtracting \( \rho(-B) \) from \( \rho(B) \). In this paper, we define the MR ratio as a change in resistivity (\( \Delta \rho = \rho(B) - \rho_0 \)) normalized by the zero-field resistivity \( \rho_0 \). The magnetization for a polycrystalline sintered \( \text{BaIrO}_3 \) was measured with a commercial SQUID magnetometer (MPMS-XL7, Quantum Design, Inc.) in magnetic fields up to 7 T.

3. Results and Discussion

Figure 1(a) shows the MR ratio curves of the longitudinal configurations at 1.5 K. As the magnetic field changes from 0 T to 14 T and then to 0 T, the \( \Delta \rho_\parallel / \rho_0 \) signal at 1.5 K traces out a butterfly-shaped hysteresis curve and the sign of the MR ratio for the field-ascending sweep changes from negative to positive near 6 T. For the field-ascending sweep, the \( \Delta \rho_\parallel / \rho_0 \) has a minimum at 2 T and a typical inflection-point at around 10 T. On the other hand, a monotonic decrease of \( \Delta \rho_\parallel / \rho_0 \) with decreasing \( B \) is observed for the field-descending sweep. The MR ratios for these sweeps overlap each other around 13 T. For the transverse configuration (not shown here), the curve of \( \Delta \rho_\perp / \rho_0 \) shows a different behavior from that of \( \Delta \rho_\parallel / \rho_0 \). This discrepancy between \( \Delta \rho_\parallel / \rho_0 \) and \( \Delta \rho_\perp / \rho_0 \), so called anisotropic magnetoresistance effect (AMR) is observed in a number of itinerant ferromagnets [6]. For the \( \text{BaIrO}_3 \) single crystal, we obtain the AMR ratio per \( \mu_B \), \( (\rho_\parallel - \rho_\perp) / \rho_0 \sim 400 \% / \mu_B \) at 1.5 K. This value is considerably larger than those of

![Figure 1](image_url)

**Figure 1.** (a) Magnetoresistance as a function of magnetic field for a \( \text{BaIrO}_3 \) single crystal at 1.5 K. The magnetic field is applied parallel to the electrical current direction that is parallel to the \( c \)-axis of the sample. (b) Magnetization curve at 2 K for a polycrystalline \( \text{BaIrO}_3 \).
any previously reported itinerant ferromagnets, e.g., for Ni-based alloys, the AMR ratio per $\mu_B$ is about 20% at the most [6].

Figure 1(b) shows the magnetization curve of a polycrystalline sintered BaIrO$_3$ at 2 K in magnetic fields up to 7 T. A large hysteresis loop remains open and is not saturated up to 7 T. In our preliminary magnetization measurement for the polycrystalline sample at 4.2 K in pulsed magnetic fields up to 52 T, we observed that the hysteresis loop closes at around 14 T, corresponding to the overlap field between the field-ascending and the field-descending sweeps in the MR ratio curve. It is noted that the coercivity field in the magnetization curve is nearly equal to the field at the minimum in the MR ratio curve.

For ferromagnetic metals, the relation between the MR and the magnetization curves is described as $\Delta\rho/\rho_0 \propto [M/M_s]^2$ [7], where $M_s$ is the saturation magnetization. We therefore draw a expected magnetization-curve from the following relation, $(\Delta\rho/\Delta\rho_{\text{max}})^{1/2} = ([\rho(B) - \rho(B_1)]/\rho(B_2) - \rho(B_1))]^{1/2} \propto \pm M(B)/M(B_2)$, where $B_1 (= 2 T)$ and $B_2 (=14 T)$ are the field of the minimum in the MR ratio curve and the maximum field of the MR measurement, respectively. The analytic result for the longitudinal configuration at 1.5 K is shown in Fig. 2(a).

By an analogy with the relation between the MR and the magnetization curves of the AMR ferromagnetic metals, we assume that the field at the minimum of the MR ratio of BaIrO$_3$ corresponds to the coercive field in the expected magnetization-curve of it. Thus, the sign of $(\Delta\rho/\Delta\rho_{\text{max}})^{1/2}$ for the field-ascending sweep may become negative below $B_1$. The expected magnetization-curve shows a large hysteresis and a plateau $(M(B) \sim 2/3 M(B_2))$ at around 10 T for the field-ascending sweep. The butterfly-shaped MR curve (see Fig. 1(a)) and the appearance of the $\sim 2/3$ plateau in the expected magnetization-curve cannot be accounted for by a magnetic domain-wall motion in conventional itinerant ferromagnets. We discuss these results through a microscopic approach based on a novel crystal structure and a local magnetic-anisotropy related to the spin-orbit coupling of this compound.

The monoclinic BaIrO$_3$ has a complex structure, in which the Ir$_3$O$_{12}$ trimers are vertex linked to construct a zigzag chain along the c-axis resulting in multiplicity of Ir-O bond distances, thus creating four types of Ir$^{4+}$ sites [3]. Different nonequivalent Ir atoms are represented by Ir1,

Figure 2. (a) $[\{\rho(B) - \rho(2T)]/\{\rho(14T) - \rho(2T)])^{1/2}$ vs $B$ at 1.5 K. The inset shows the schematic diagram of Ir$_3$O$_{12}$ trimers in the monoclinic structure of BaIrO$_3$. The expected spin configurations on Ir$^{4+}$ sites of BaIrO$_3$ at (b) $B = 0$ T, (c) $B = 2$ T, (d) $B \sim 10$ T, and (e) $B = 14$ T for the field-ascending sweep.
Ir2, Ir3, and Ir4 in the inset of Fig. 2(a). Each trimer consists of two nonequivalent Ir atoms; Ir2 and Ir4 are located in the central octahedra of the Ir3O12 trimers, Ir1 and Ir3 are located in the other two octahedra. Accordingly there are two types of exchange-interactions between Ir4+ ions. One is an exchange-interaction mediated by apex oxygen atoms of corner-shared IrO6 octahedra between Ir1 and Ir3. The other is an exchange-interaction mediated by a triangle face of IrO6 octahedra between Ir1 and Ir2 atoms or Ir3 and Ir4 atoms. It is expected that a local magnetic-anisotropy related to the spin-orbit coupling on each Ir4+ site is classified into two groups, {Ir1} (Ir1 and Ir3) and {Ir2} (Ir2 and Ir4), according to the Ir-O-Ir bonds. As a result of the electrical resistivity [4, 5] and the band calculations [8] on BaIrO3, the dominant contribution to the electrical conductivity is the d-orbitals of the {Ir1} atoms. It might well be that the Ir4+ spins of {Ir1} site are more sensitive to a change of magnetic field than those of {Ir2} site. The compounds of Ir4+ (5d5, low spin) have a large spin-orbit coupling constant [9]. Thus, despite the small Ir4+ moment, the large spin-orbit coupling in BaIrO3 may provide the possibility of yielding a large magnetic-anisotropy through a large crystal field. We assume that the strength of the local magnetic-anisotropy of {Ir2} site is larger than that of {Ir1}. The magnetic easy axis of BaIrO3 is parallel to the c-axis [4, 5].

The (∆ρ/∆ρmax)1/2 behavior in Fig. 2(a) can be explained as follows based on the above assumptions; (i) At 14 T, the spin directions of all Ir4+ sites are parallel to the c-axis, and the moment of each Ir4+ site is equal in magnitude (Fig. 2(e)). Then, for the field-descending sweep (14 T to 0 T), the moments of {Ir1} site are gradually compressed. The same situation occurs for the field-ascending sweep (-14 T to 0 T), that is, the moments of {Ir1} site are compressed (Fig. 2(b)). (ii) At 2 T for the field-ascending sweep, the spins of {Ir1} site remain unchanged (Fig. 2(c)). The total moment becomes zero. (iii) Furthermore, when the field is raised to around 10 T, the spins of {Ir2} site begin to flip towards the field direction. It is possible that the effective moment of {Ir2} site becomes zero as shown in Fig. 2(d) because of the appearance of the ~2/3 plateau. We propose that the unique MR effects of BaIrO3 are caused by a nanoscale spin valve mechanism mentioned above.

4. Conclusions
In conclusion, we have investigated the MR effect of a BaIrO3 single crystal. We have observed the large AMR ratio per µB, (ρ∥ - ρ⊥)/ρ0 ∼ 400 %/µB at 1.5 K. As the magnetic field changes from 0 T to 14 T and back to 0 T, the ∆ρ∥/ρ0 signal at 1.5 K traces out a butterfly-shaped hysteresis curve and the sign of the MR ratio for the field-ascending sweep changes from negative to positive near 6 T. The MR results are interpreted based on a spin valve model considering a novel crystal structure and a local magnetic-anisotropy related to the spin-orbit coupling of this compound.

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