Pressure effect on magnetic properties of a weak ferromagnet BaIrO$_3$

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Abstract. We have investigated the pressure effect on magnetic properties of a weak itinerant ferromagnet BaIrO$_3$, using a Cu-Be piston-cylinder type pressure cell. Under external pressure the ferromagnetic transition temperature $T_c$ decreases linearly with pressure at a large rate of $dT_c/dP \sim -17$ K/GPa. The critical exponent $\beta$ up to 0.86 GPa indicates the anomalous value ($\beta \sim 0.9$) and is nearly pressure-independent. Anomalous pressure effects on conductivity and magnetism suggest the existence of a nontrivial coupling among charge, spin and lattice in 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 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 $W$. 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 corner-shared to construct a zigzag chain along the $c$-axis [2]. The top and bottom triangle faces of each trimer are parallel to the $ab$-plane, forming the two-dimensional layers of the corner-shared IrO$_6$ octahedra. Owing to the peculiar crystal structure, anisotropic properties are observed in electrical resistivity, magnetization, and thermoelectric power [3, 4]. BaIrO$_3$ exhibits a ferromagnetic transition with a small Ir$^{4+}$ moment of 0.04 $\mu_B$ at $T_c \sim 180$ K accompanied by 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 [3, 4]. In spite of
the ferromagnetic ground state, the ordered state below $T_c$ was believed to be a CDW (charge density wave) state which was found by a optical conductivity measurement [3]. Thus, we expect that the coexistence of the spontaneous magnetization and the energy gap will lead to unprecedented physical properties in BaIrO$_3$.

Pressure is an important thermodynamical parameter to control the state of matter and can affect the magnetic properties of materials [5]. To our knowledge, there are little reports of high-pressure physical properties on oxide iridates so far. In the present study, we report the pressure effect of the magnetic properties on BaIrO$_3$, using high-pressure measurements of the magnetization in a Cu-Be piston-cylinder type pressure cell.

2. Experimental
For this study, polycrystalline sintered samples of BaIrO$_3$ were prepared by heating a mixture of stoichiometric amounts of BaCO$_3$ and Ir at 1050 °C for 12 h in air. The temperature dependence of magnetization $M$ at $H = 100$ Oe (field cooled condition; FC) and isothermal magnetization ($M$ vs $H$) at different temperatures were measured with a commercial SQUID magnetometer (Quantum Design MPMS-XL5S) in hydrostatic pressures up to 0.86 GPa with a piston-cylinder type pressure cell. The pressure cell is made of hardened Cu-Be alloy and has a cylindrical shape with an inner diameter of 2 mm and a length of 105 mm [6]. As a pressure transmitting medium a mixture of Fluorinert FC70:FC77 = 1:1 was used. The actual pressure was determined in each experiment by measuring the superconducting transition temperature of Sn which was put into the pressure medium together with a sample and placed about 20 mm away from the sample.

3. Results and Discussion
$M/H$ of BaIrO$_3$ as a function of temperature at ambient pressure and under different hydrostatic pressures are shown in Fig. 1(a). At ambient pressure, our sample shows a ferromagnetic transition temperature $T_c \approx 185$ K. Fitting to a modified Curie-Weiss law $M/H = \chi = C/(T - \theta_p) + \chi_0$ for $T > 190$ K yields the following parameters: the Curie-

![Figure 1](image-url)

**Figure 1.** (a) Temperature dependence of the susceptibilities ($M/H$) on polycrystalline BaIrO$_3$ at various pressures up to 0.86 GPa. The inset shows the pressure dependence of $T_c$. (b) Logarithmic plot of the magnetization as a function of the reduced temperature $1 - T/T_c$ at ambient pressure, 0.44, 0.54, and 0.86 GPa. The data are offset down at each pressure increment for clarity.
Weiss temperature $\theta_w = 183.4$ K (positive), the Curie constant $C = 2.11 \times 10^{-3}$ emu K/mol, the effective paramagnetic moment $\mu_{\text{eff}} = 0.13 \mu_B/Ir$ and the large temperature-independent term $\chi_0 = 2.6 \times 10^{-4}$ emu/mol, which is probably caused by the large spin-orbit coupling in this compound. $\theta_p$ is comparable to the magnetic ordering temperature and suggestive of a ferromagnetic spin coupling. The effective moment $\mu_{\text{eff}}$ is significantly smaller than the expected $S = 1/2$ moment ($\mu_{\text{eff}} = 1.73 \mu_B/Ir$) for the low-spin $\text{Ir}^{4+} (5d^5)$ configuration. These parameter values are in good agreement with the reported ones [3]. The inset of Fig. 1(a) shows the $S = 1$ effective paramagnetic moment $\mu$ in this compound.

The relative pressure dependence $\partial \ln T_c/\partial P$ in an homogeneous ferromagnet can be expressed as

$$\partial \ln T_c/\partial P = -\alpha/T_c^2 + (5/3)\kappa,$$  

where $\alpha$ is a slowly varying quantity and $\kappa$ is the compressibility. In general, the $\partial \ln T_c/\partial P$ for the weak itinerant ferromagnets is negative, suggesting that both $\alpha$ and $\kappa$ are positive, and that the first term on the right-hand side in Eq. (1) is dominant in comparison with the second term. For the Ba$_{1-x}$Sr$_x$IrO$_3$ series, $T_c$ decreases with decreasing lattice parameter [8], which is consistent with the present pressure effect in BaIrO$_3$. It suggests that the magnetic interaction of BaIrO$_3$ is tightly correlated with the lattice.

Recently, we reported that BaIrO$_3$ shows unconventional critical behavior [9]. The critical exponents have been determined to be $\beta = 0.82 \pm 0.03$, $\gamma = 1.03 \pm 0.03$, and $\delta = 2.20 \pm 0.01$, which roughly obeys the scaling relation $\delta = \gamma/\beta + 1$ of the second-order phase transition. We then roughly estimated the $\beta$ of BaIrO$_3$ under hydrostatic pressure by a power-law fitting in the logarithmic $M$ vs. $1-T/T_c$ plot near $T_c$. The $\beta$ is defined as $M \propto (1-T/T_c)^\beta$ for $T < T_c$, and is about 0.9 when hydrostatic pressure up to 0.86 GPa. The $\beta$ value is larger than the mean-field value ($\beta = 0.5$). It is noted that the $\beta$ is nearly pressure-independent, as indicated by the slope of solid lines in Fig. 1(b), suggesting that the ferromagnetic ordering and the gap opening of BaIrO$_3$ are not separated by external pressure. This result shows that unconventional critical properties of BaIrO$_3$ are preserved even under hydrostatic pressure.

![Figure 2](image-url.png)

**Figure 2.** Temperature dependence of magnetization curves on BaIrO$_3$ at (a) ambient pressure and (b) $P = 0.86$ GPa.
exponents of BaIrO$_3$ are not attributable to multicritical behavior. We believe that the anomaly of the critical exponents on BaIrO$_3$ is of intrinsic nature, which characterizes the unconventional phase transition of this compound.

Figures 2(a) and (b) show the temperature dependence of magnetization curves at ambient pressure and $P = 0.86$ GPa, respectively. Below 100 K, large hysteresis loops appear and the magnetizations are not saturated in magnetic fields up to 5 T at both ambient pressure and 0.86 GPa. At ambient pressure, the coercivity field increases with decreasing temperature and the value of the saturation (at 5 T) magnetization at 5 K is smaller than those of other temperatures. At 0.86 GPa, the temperature dependence of the saturation magnetization is weaker than that at ambient. The coercivity fields at 5 K and 100 K show no change with pressure, but those at 25 K and 50 K increase with pressure and are equal to that at 5 K. According to the pressure effect of transport properties on BaIrO$_3$ single crystal reported by Nakano [10], the electronic state of BaIrO$_3$ at ambient pressure varies with temperature; (i) the Mott insulator state ($T > 180$ K), (ii) a charge ordered state ($20$ K $< T < 180$ K), and then (iii) the Mott insulator state ($T < 20$ K) partially coexists with the charge ordered state again. We associate the drop of $M$ at 5 K with the nonlinear conduction. According to Ref. [10], the Mott insulating state is partially mixed at 5 K, which drops the saturation magnetization. Since this drop is suppressed at 0.86 GPa, the pressure seems to let the system homogeneous. This picture is consistent with the fact that the pressure suppresses the nonlinear conduction. In this respect, there exists a non-trivial coupling between magnetism, conduction, and lattice in this material.

4. Conclusions
In conclusion, we have investigated the pressure effect of magnetic properties on the weak itinerant ferromagnet BaIrO$_3$, using a Cu-Be piston-cylinder type pressure cell. Under external pressure the $T_c$ decreases linearly with pressure at $dT_c/dP \sim -17$ K/GPa. In addition, unconventional critical behavior was observed under pressure. The critical exponents $\beta$ under pressures up to 0.86 GPa are almost identical and show anomalous values ($\beta \sim 0.9$) in accordance with that at ambient pressure. These results suggest the existence of a nontrivial coupling among charge, spin and lattice in this compound.

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