Transport, Thermal and Magnetic Properties of Pyrochlore Oxides $Y_{2-x}Bi_xIr_2O_7$

M. Soda [a], N. Aito [a], Y. Kurahashi [a], Y. Kobayashi [a] and M. Sato [a,1]

[a] *Department of Physics, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464-8602 Japan*

Abstract

Various macroscopic physical properties have been studied for the pyrochlore oxides $Y_{2-x}Bi_xIr_2O_7$ which exhibits the Mott insulator to metal transition with increasing x. The phase boundary has been found at $x = x_c \sim 0.5$, where the electronic specific heat coefficient γ has a maximum value. Hysteretic behavior of the magnetization-temperature (T) curve, which indicates the possible existence of the transition to the glassy phase, vanishes at around x_c. Measurements of the specific heat of $Y_2Ir_2O_7$ down to 0.95 K have revealed the peculiarity of its T-dependence.

Key words: pyrochlore oxides; $Y_{2-x}Bi_xIr_2O_7$; Mott transition

Pyrochlore oxides $A_2B_2O_7$ have the structure consisting of individual networks of the corner-linked A_4 and B_4 tetrahedra [1]. The strong magnetic frustration often arises from this structural origin. Another characteristic is that each magnetic moment at a corner of a tetrahedron has the anisotropy axis, which is parallel to the line connecting the corner with the center of gravity of the tetrahedron. Because there are four different directions of the axes, quite unusual magnetic behaviors are often observed: The so-called spin-ice behavior [2,3] and the unusual Hall resistivity [4].

$Y_2Ir_2O_7$ is a Mott insulator and becomes metallic with the Bi substitution for Y, and here, $Y_{2-x}Bi_xIr_2O_7$ has been adopted to see how the frustrated spin system in the Mott insulating phase behaves near the metal-insulator (M-I) phase boundary. Such works were carried out by the authors' group on pyrochlore ruthenates [5,6] and it was revealed that the Hall coefficient R_H of the system exhibits anomalous temperature(T)-dependence near the M-I boundary as in high-T_c Cu oxides. In the Mott insulating phase of the ruthenates, the valence of Ru is +4 and the spin $S = 1$, while the present system with $x = 0$ (in the Mott insulating phase) has $S = 1/2$. Then, the magnetic anisotropy does not exist in the present case and effects of the spin frustration are expected to be more significant.

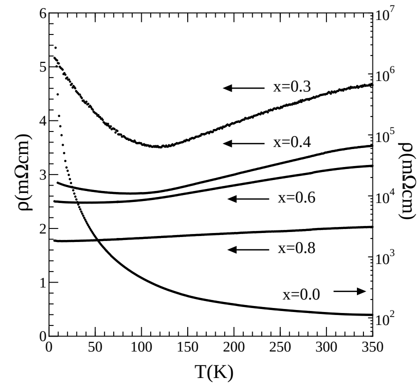

Fig. 1. T-dependences of the electrical resistivities ρ of $Y_{2-x}Bi_xIr_2O_7$ are shown for various x values.

Mixtures of Y_2O_3, Bi_2O_3 and Ir with proper molar ratios were ground, pressed into pellets and heated at 1000 °C for 24 h. Then, they were cooled in the furnace. The processes of the grinding and the heat treatments

1 Corresponding author. E-mail: e43247a@nucc.cc.nagoya-u.ac.jp

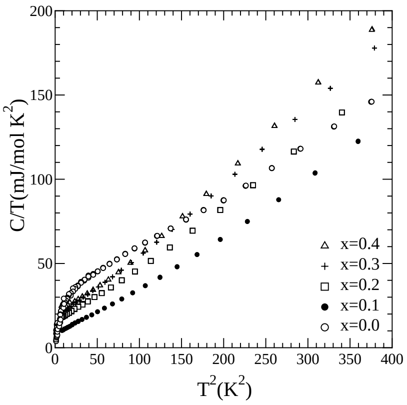

Fig. 2. Specific heat data divided by T, C/T are plotted against T^2, for $Y_{2-x}Bi_xIr_2O_7$ samples with various x values.

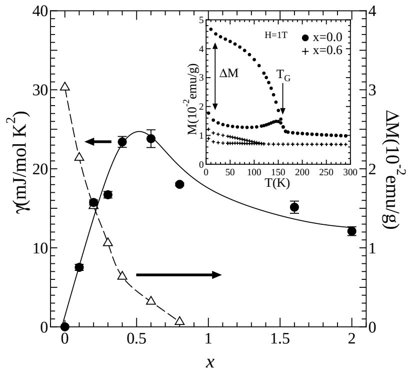

Fig. 3. Electronic specific heat coefficient γ (solid circles) of $Y_{2-x}Bi_xIr_2O_7$ is plotted against x. The difference ΔM (open triangles), estimated at low temperatures, between the magnetizations M measured with the magnetic field of 1 T with increasing T after zero field cooling and then, with decreasing T is also plotted. Inset shows the examples of the hysteretic behavior of M.

were repeated once more for the samples with nonzero x. For the sample with $x = 0$, the processes were repeated many times. The X-ray diffraction studies have revealed that all the samples are the single phase.

The electrical resistivities ρ were measured by the standard four probe method. The magnetic susceptibilities χ were measured by a SQUID magnetometer with the applied magnetic field(H) of 1 T. The specific heats were taken by the thermal relaxation method in most cases and by the adiabatic heat pulse method in the T region below 2 K for $x = 0$.

Figure 1 shows the $\rho - T$ curves of the samples of $Y_{2-x}Bi_xIr_2O_7$ with various x values. For $x = 0$, ρ exhibits the insulating behavior and at low temperatures, $d\rho/dT$ changes, with varying x, its sign at $x = x_c \sim 0.5$. A slight anomaly is observed in the T-dependence of ρ and the thermoelectric powers S in the region of small x at T_G, where the hysteretic behavior appears in the χ-T curve with decreasing T(See the inset of Fig. 3.). The anomalous behavior is quite similar to that observed in the pyrochlore ruthenates [5,6], where a kind of magnetic transition(possibly to spin glass phase) accompanied by very tiny spontaneous magnetic moment takes place. In the present case, however, the specific heat anomaly observed at T_G is much weaker than that of ruthenates and it is visible only for $x = 0$. The hysteretic behavior has been reported for $Y_2Ir_2O_7$ and other Ir pyrochlore systems [7,8].

Examples of the specific heat data of $Y_{2-x}Bi_xIr_2O_7$ are shown in Fig. 2 in the form of C/T-T^2 for various x values. With increasing x, the T region where the data fall on the straight line becomes very narrow, because the weight of the low frequency modes increases. One of the interesting characteristics of the data is that C/T for $x = 0$ exhibits peculiar behavior: The value at $T \to 0$ was found to be zero by the measurements down to ~ 0.95 K. It increases rapidly when T is increased from the lowest temperature studied here. It may indicate the remaining effects of the fluctuating quantum spins, though the magnetic transition exists at T_G, and therefore the entropy of the low T anomaly is very small as compared with the spin entropy $k_B\log2$. Similar peculiar behavior of $Y_2Ir_2O_7$ has also been reported in ref. [8], though their measurements are just down to 1.8 K.

Figure 3 shows the electronic specific heat coefficient γ of $Y_{2-x}Bi_xIr_2O_7$ as a function of x. It has a peak at a value which roughly corresponds to x_c, where the low T value of $d\rho/dT$ changes its sign. At around this x_c, ΔM defined in the inset of Fig. 3 as the measure of the strength of the hysteretic nature almost vanishes. Then, it can be said that along with the disappearance of the magnetic transition at T_G, the M-I transition takes place at low T with varying x, and γ becomes maximum at this phase boundary.

References

[1] M.A. Subramanian, G. Aravamudan and G.V. Subba Rao, Prog. Solid State Chem.**15** (1983) 55.

[2] A.P. Ramirez, A. Hayashi, R.J. Cava, R. Siddharthan and B.S. Shastry, Nature **399** (1999) 333.

[3] M. Kanada, Y. Yasui, Y. Kondo, S. Iikubo, M. Ito, H. Harashina, M. Sato, H. Okumura, K. Kakurai and H. Kadowaki, J. Phys. Soc. Jpn. **71** (2002) 313.

[4] S. Yoshii, S. Iikubo, T. Kageyama, K. oda, Y. Kondo, K. Murata and M. Sato, J. Phys. Soc. Jpn. **69** (2000) 3777.

[5] S. Yoshii and M. Sato, J. Phys. Soc. Jpn. **68** (1999) 3034.

[6] S. Yoshii, K. Murata and M. Sato, J. Phys. Soc. Jpn. **69** (2000) 17.

[7] N. Taira, M. Wakeshima and Y. Hinatsu, J. Phys. Condens. Matter **13** (2001) 5527.

[8] H. Fukazawa and Y. Maeno, cond-mat/0202169.