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The effect of pressure on the superconductivity and magnetism of $RuSr_2GdCu_2O_8$

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Abstract
A new hybrid ruthenate–cuprate superconductor $RuSr_2GdCu_2O_8$ has been synthesized. This compound exhibits superconductivity and magnetic ordering around 40 K ($=T_c$) and 130 K ($=T_m$). The effect of pressure on the electrical resistance has been measured up to 2.1 GPa in order to make clear the interplay between the magnetism and the superconductivity. It is found that both T_c and T_m increase with pressure with rates of $\mathrm{d}T_c/\mathrm{d}P = 1.9$ K GPa^{-1} and $\mathrm{d}T_m/\mathrm{d}P = 5.7$ K GPa^{-1}, which indicates that the superconductivity does not compete with the magnetic ordering. An x-ray diffraction experiment under pressures up to 13 GPa has been carried out in order to clarify the relation between the electronic properties and the lattice compression.

1. Introduction

Recently the ruthenate–cuprate compounds have attracted much attention because of their interesting electronic properties. Among them, the hybrid ruthenate–cuprate compound $RuSr_2GdCu_2O_8$ (abbreviated as Ru⟨1212⟩) has been investigated extensively because it exhibits magnetic order around 130 K ($=T_m$) and superconductivity around 35 K ($=T_c$) [1–4]. For this compound, it is worthwhile to study the electronic state under external forces in order to make clear the interplay between the superconductivity and magnetic ordering.

In the present work we made an attempt to measure the electrical resistance and lattice parameters under high pressure to examine the stability of the electronic structures. The results are discussed briefly by using Grüneisen parameters of two characteristic temperatures, T_m and T_c.

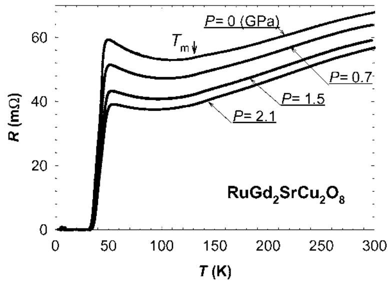

Figure 1. $R(T)$ for Ru⟨1212⟩ under high pressure.

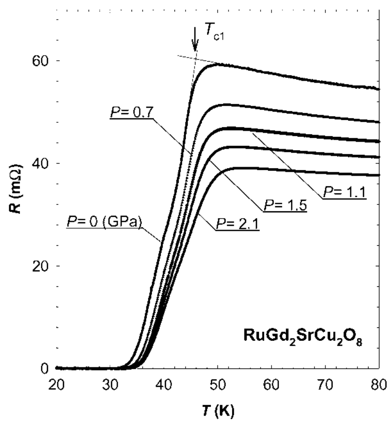

Figure 2. $R(T)$ for Ru⟨1212⟩ under high pressure at low temperature below 80 K.

2. Experimental details

The high-quality sample was synthesized by the solid-state reaction method. Electrical resistance was measured using the standard four-probe method, and hydrostatic pressure up to 2.1 GPa was generated by using a CuBe piston–cylinder device and a 1:1 mixture of Fluorinert FC70 and FC77 as a pressure medium. The details of the pressure apparatus have been reported elsewhere [5]. The pressure dependence of the lattice parameter was determined by x-ray (Mo Kα) powder diffraction using a Guinier-type focusing camera. Hydrostatic pressure was generated by tungsten carbide Bridgman-type anvils up to about 13 GPa.

3. Results and discussion

3.1. Electrical resistance at high pressure

Figure 1 shows the temperature dependence of the electrical resistance (R) of Ru⟨1212⟩ under high pressure. During cooling, $R(T)$ shows characteristic behaviour of an underdoped cuprate: $R(T)$ is semiconducting as seen from the upturn around 100 K and shows a sudden decrease to zero at 35 K. There is also small anomaly (inflection) at 130 K ($=T_m$) due to magnetic ordering. With increasing pressure, R decreases over the whole temperature range and the semiconducting upturn becomes small.

To show the $R(T)$ curve near T_c more clearly, $R(T)$ is shown on an expanded scale in figure 2. Here we defined two characteristic temperatures, T_{c1} and T_{c2}, for the temperatures at which superconductivity has its onset and disappears, respectively. T_{c1} is determined as the temperature where two straight lines intersect: one line is an extension of $R(T)$ in the range $T > T_c$ and the other is that of $R(T)$ in the range $T < T_c$. T_{c1} at ambient pressure is shown

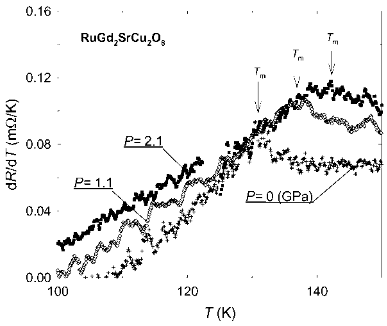

Figure 3. The temperature derivative $\mathrm{d}R/\mathrm{d}T$ for Ru⟨1212⟩ near the magnetic ordering temperature under high pressure.

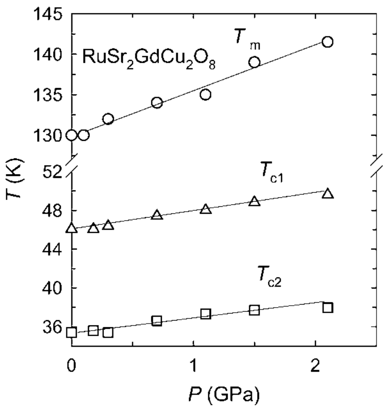

Figure 4. Pressure dependences of T_{c1}, T_{c2} and T_m for Ru⟨1212⟩. The lines are guides to the eye.

by an arrow in figure 2. T_{c2} is the temperature with $R(T) = 0.5$ mΩ. It is found that both T_{c1} and T_{c2} increase with increasing pressure, having the coefficients $\mathrm{d}T_{c1}/\mathrm{d}P = 1.9$ K GPa^{-1} and $\mathrm{d}T_{c2}/\mathrm{d}P = 1.6$ K GPa^{-1}.

Next we determine the pressure dependence of the magnetic ordering temperature T_m. In order to define T_m, the temperature derivative of the $R(T)$ curve was calculated and is shown in figure 3 as a function of T. A peak is clearly seen at each pressure. T_m is defined as the temperature showing the peak. T_m is found to increase with pressure with a coefficient of 5.7 K GPa^{-1}.

Pressure dependences of the critical temperatures T_{ci} $(i = 1, 2)$ and T_m are displayed in figure 4. Both T_{ci} and T_m increase with pressure. From these results, pressure is expected to increase both the number of hole carriers in the CuO_2 plane and the exchange interaction between the magnetic moments and carriers in the RuO_2 plane.

3.2. X-ray diffraction under high pressure

In order to investigate the crystal structure and lattice parameters under high pressure, x-ray diffraction measurement was carried out. At ambient pressure, the lattice parameters are $a = 3.836(2)$ Å and $c = 11.527(9)$ Å for the tetragonal cell, which are consistent with previous experiment [6]. Figures 5(a) and (b) show the pressure dependences of the relative lattice parameters a/a_0 and c/c_0 of the tetragonal cell and the relative volume V/V_0 respectively. Both a/a_0 and c/c_0 decrease with increasing pressure, at almost the same rate, below 6 GPa, but a small anisotropy is seen in their compression above 6 GPa: the c-axis is contracted more than the a-axis by pressure. This implies the possibility of phase transition. A more

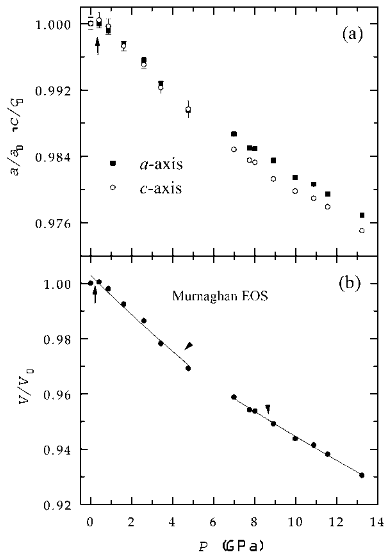

Figure 5. Pressure dependences of: (a) a/a_0 and c/c_0; (b) V/V_0. The Murnaghan EOS fitting curve for the experimental data is indicated by solid lines in (b) below and above 6 GPa.

detailed experiment is now planned to clarify this issue. It is also seen that all parameters do not decrease initially with increasing pressure but appear to remain constant or to increase slightly, and they begin to gradually decrease above 0.5 GPa as shown by an arrow in figure 5. A similar observation has been reported for the pressure dependences of the lattice parameters of TiO [7], which includes a large number of vacancies on both Ti and O lattice sites. The crystal lattice may expand provided that the ionic size of the vacancy is smaller than that of the overdoped atom (e.g. oxygen). The detailed explanation is given in [7]. The origin of the anomalous behaviour below 0.5 GPa in the present work may be the same as that for TiO.

To estimate the bulk modulus, we attempted a least-square fit of the data of $(V/V_0)(P)$ to the following first-order Murnaghan equation of state:

$$P = \left(\frac{B_0}{B_0'}\right)\left[\left(\frac{V_0}{V}\right)^{B_0'} - 1\right], \tag{1}$$

where B_0 denotes the bulk modulus at ambient pressure and B_0' its pressure derivative. Since there is an anomaly below 0.5 GPa and the lattice compressions of the *a*- and *c*-axes below 6 GPa are different from those above 6 GPa, we tried to fit the present data to equation (1) above and below 6 GPa. The solid lines for V/V_0 in figure 5(b) show the results of the fitting, assuming $B_0' = 4$. The values of B_0, determined in low-pressure ($P \leqslant 6$ GPa) and high-pressure ($P \geqslant 6$ GPa) regions, are 136(9) and 173(5) GPa respectively.

3.3. Calculation of Grüneisen parameters of T_m and T_c

In order to evaluate the stability of characteristic electronic states of Ru⟨1212⟩ compounds, we estimate the Grüneisen parameters Γ for T_{ci} and T_m. Γ is defined as

$$\Gamma(T_0) = -\frac{\partial \ln T_0}{\partial \ln V}, \tag{2}$$

where T_0 is a characteristic temperature. Using the isothermal bulk modulus B_T, equation (2) is rewritten as

$$\Gamma(T_0) = B_T \frac{\partial \ln T_0}{\partial P}. \tag{3}$$

Table 1. Pressure derivatives of T_c and T_m for Ru⟨1212⟩.

T_0	dT_0/dP (K GPa^{-1})	$d\ln T_0/dP$ (10^{-2} GPa^{-1})
T_{c1}	1.9	4.1
T_{c2}	1.6	4.5
T_m	5.7	4.4

The data obtained in sections 3.1 and 3.2 are summarized in table 1. By using these values, we got the following values for the Γs:

$$\Gamma(T_{c1}) = 5.6, \qquad \Gamma(T_{c2}) = 6.1, \qquad \Gamma(T_m) = 6.0. \tag{4}$$

It is very surprising that these values are almost the same. In other words, the superconductivity and magnetic ordering are enhanced by pressure in similar ways.

4. Conclusions

The electrical resistances of Ru⟨1212⟩ compounds have been measured at high pressure up to 2.1 GPa. It is found that both the superconducting transition temperatures and the magnetic ordering temperature increase with pressure. The lattice parameters and crystal structure are also examined at high pressure by using x-ray diffraction. Using these data, the Grüneisen parameters were determined. The results indicated that the Grüneisen parameters of T_c and T_m are almost the same.

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