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Pressure effects on the superconductivity and magnetization in $\text{RuSr}_2\text{GdCu}_2\text{O}_8$

Yuh Yamada¹, Y Fujimoto¹, S Hatanaka¹, T Shibata¹, T Naka²,
J Tang², A Matsushita² and S Kubo¹

¹ Department of Material Science, Shimane University, Matsue, Shimane 690-8504, Japan

² National Institute for Materials Science, Tsukuba, Ibaraki 305-0047, Japan

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Abstract

A polycrystalline sample of $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ exhibits the superconducting transition at about 40 K and ferromagnetic order at about 130 K. We have investigated the crystal structure of this sample by means of a synchrotron radiation x-ray powder experiment and analysed the data by the Rietveld method. The results indicated the Cu sites to contain 5% Ru. The pressure dependences of the superconducting and ferromagnetic transition temperatures of this sample were measured. The superconducting transition temperature scarcely varies with pressure. The ferromagnetic transition temperature increases with increasing pressure at lower pressure. However, the increase of the ferromagnetic transition temperature became saturated above 1.6 GPa. The magnitude of the susceptibility decreases with increasing pressure below the ferromagnetic transition temperature.

1. Introduction

Coexisting ferromagnetism and superconductivity have recently been discovered in the 1212-type layered cuprate superconductor $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ (Ru1212) [1]. This material is isostructural with $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ with Y, Ba, and Cu1 (the chain copper atom) completely replaced by Gd, Sr, and Ru, respectively, and displays bulk superconductivity below $T_c = 0$ –46 K which is associated with the CuO_2 planes, and a Curie transition at $T_f = 132$ K. The ferromagnetism occurs in RuO_2 planes and is like that in SrRuO_3 which is an itinerant electron ferromagnet below $T_f = 165$ K. It is interesting to study the effects of pressure on this material to achieve an understanding of the coexisting ferromagnetism and superconductivity in these systems.

In this work, we will report on the pressure dependences of the superconducting and ferromagnetic transition temperatures of Ru1212.

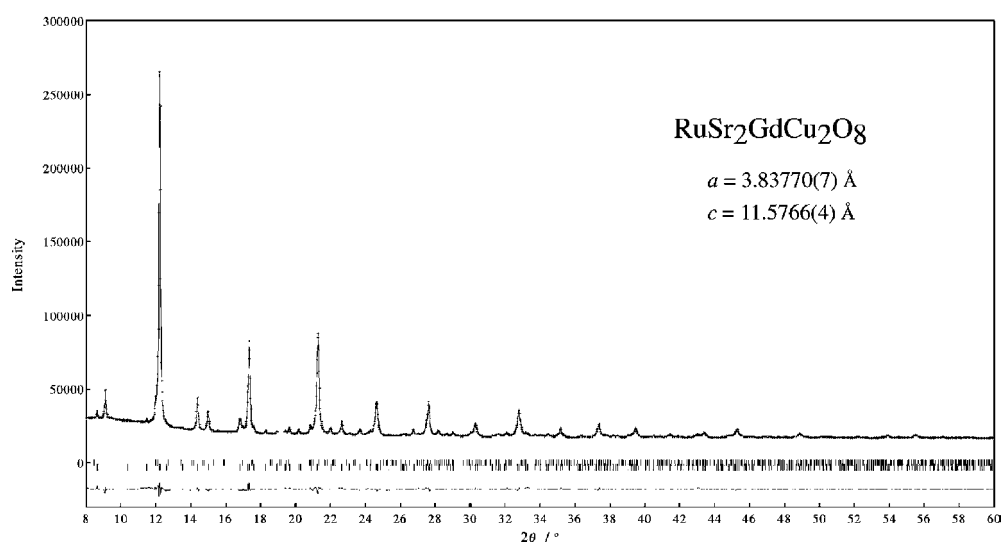


Figure 1. Observed, calculated, and difference synchrotron x-ray diffraction data for the $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ sample at RT.

2. Experimental procedure

A polycrystalline sample of Ru1212 was synthesized by solid-state reaction of a stoichiometric mixture of the oxides RuO_2 , Gd_2O_3 , CuO , and SrCO_3 [1]. These were decomposed at 900°C and die-pressed into pellets before being reacted in air at 1000 for six days. This was followed by reaction in flowing oxygen at 1050°C for six days and annealing at 530°C for 48 h under 5 atm oxygen. The final product was characterized by means of x-ray diffraction with $\text{Cu K}\alpha$ radiation. Powder synchrotron x-ray diffraction patterns for this sample were recorded on the SPring-8 beamline BL02B2 at room temperature. The wavelength of the incident x-rays is 0.57971 \AA . The diffraction patterns were fitted using the tetragonal $P4/mmm$ symmetry model. This sample was found to be a mixture of the RuSrO_3 phase and other impurity phases together with Ru-1212. We have analysed the data by means of two-phase profile fitting using the RIETAN2000 program [2].

The temperature dependence of the resistivity under pressure was measured with a DC four-probe method using a cubic-anvil apparatus up to $P = 9 \text{ GPa}$. The magnetization at high pressure was measured using the Faraday method up to $P = 2.3 \text{ GPa}$. The sample was filled into a Teflon cell with Fluorinert as the pressure-transmitting medium and pressurized in a piston-cylinder-type clamp made of NiCrAl alloy.

3. Results and discussion

The diffraction data analysed by Rietveld method for this sample are shown in figure 1. In this analysis, the structural parameters of Ru1212 reported by McLaughlin *et al* [3] were used as initial parameters. No refinement was attempted for the oxygen occupancy. The refined structural parameters at room temperature are listed in table 1. The mass fraction of the RuSrO_3 phase is 6.4% and the Cu sites contained 5% Ru. The length of the bond between Cu and apical oxygen O1 which was calculated from the structural parameters for this sample is 2.15 \AA . This bond length is unusually short as compared to those in the optimally doped oxide $\text{YBa}_2\text{Cu}_3\text{O}_7$

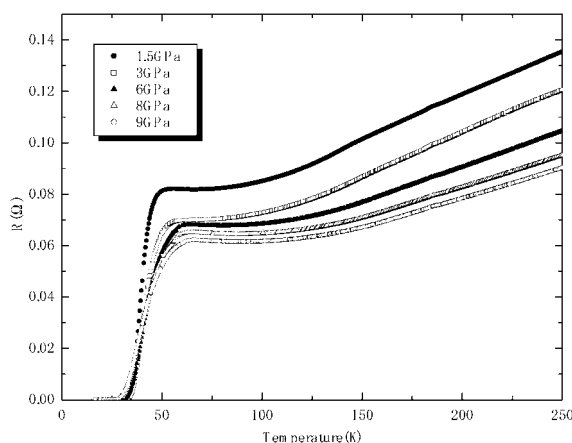


Figure 2. The temperature dependence of the electrical resistivity for RuSr₂GdCu₂O₈ measured at various pressures.

Table 1. The refined structural parameters for RuSr₂GdCu₂O₈ at RT. $R_{wp} = 1.44\%$, $R_p = 1.04\%$, $R_I = 2.11\%$ and $R_F = 1.32\%$.

Atom	Site	x	y	z	Occupancy	B
Ru	1(b)	0.0	0.0	0.5	1	0.04(5)
Sr	2(h)	0.5	0.5	0.3088(2)	1	0.43(5)
Gd	1(c)	0.5	0.5	0.0	1	0.48(4)
Cu/Cu	2(g)	0	0	0.1461(3)	0.95(1)	0.52(6)
Cu/Ru	2(g)	0	0	0.1461(3)	0.05	0.39(4)
O(1)	8(s)	0.03(2)	0.0	0.328(1)	0.25	1.25
O(2)	4(i)	0.0	0.5	0.136(1)	1	1.25(15)
O(3)	4(o)	0.088(5)	0.5	0.5	0.5	1.25

or the overdoped oxide TlSr₂CaCu₂O₇, as McLaughlin *et al* [3] indicated. This is suggested to lead to the existence of more holes in CuO₂ planes.

Figure 2 shows the temperature dependence of the electrical resistivity for Ru1212 measured at various pressures. At atmospheric pressure, we observed the superconducting transition at a temperature (T_c) of 38 K. The transition width ΔT_c , defined as the temperature interval over which the resistance dropped from 90 to 10% of the value of T_c , was about 6 K. The temperature dependence of the resistivity for this sample showed a slight enhancement near T_c indicating underdoped oxide superconductor. This is considered to be the effect of the slight replacement of Ru in CuO₂ planes.

The magnitude of the resistivity in the normal state decreases with increasing pressure. T_c scarcely varies and ΔT_c increases with increasing pressure.

The structural analysis suggested that this sample is an overdoped oxide superconductor. However, the behaviour of the pressure dependence at a glance appears similar to that of an optimally doped oxide superconductor, such as YBa₂Cu₃O₇.

The details of the pressure dependence of T_c show that the T_c for onset of superconductivity increases and the zero-resistivity temperature decreases with increasing pressure. In other words, T_c scarcely changes and ΔT_c increases with increasing pressure. We must consider the effect of the ferromagnetic moment in RuO₆ in order to understand the behaviour of the pressure dependence of T_c for Ru1212.

Figure 3 shows the temperature dependence of the magnetization of Ru1212 measured under a field of 20000 Oe at various pressures. An anomaly in the magnetic susceptibility

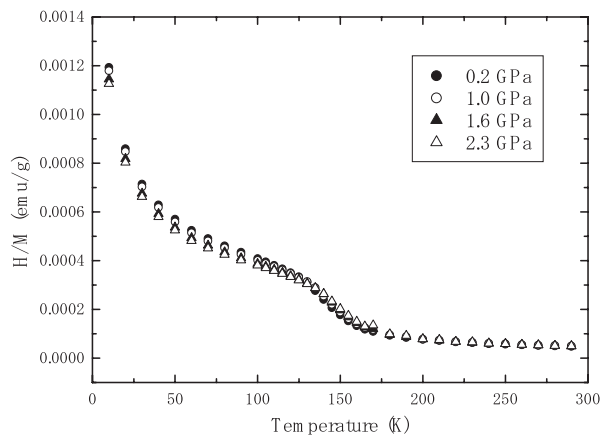


Figure 3. The temperature dependence of the magnetization for $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ measured at various pressures.

originating from the ferromagnetic ordering transition is observed at about 140 K. The ferromagnetic transition temperature (T_f) increases with increasing pressure at lower pressure. However, the increase of T_f became saturated above 1.6 GPa. Above the T_f -region, the magnitude of the susceptibility scarcely varies with pressure. On the other hand, the magnitude of the susceptibility decreases with increasing pressure below T_f . It is considered that the susceptibility below T_f consists almost entirely of the Curie term originating from the Gd moments and the Ru moments ordering ferromagnetically. It is of interest to consider which of the contributions, that from Ru or that from Gd, is dominant in the pressure effects. We also measured the pressure dependence of the magnetization for $\text{RuSr}_2\text{EuCu}_2\text{O}_8$. Eu^{3+} ions are paramagnetic, but do not follow a Curie law. Because of the proximity of excited J -multiplets to the $J = 0$ ground state, the susceptibility is usually described using the Van Vleck formula. In our results, decrease of the susceptibility was obtained with increasing pressure below T_f . We concluded that the decreasing of the susceptibility under pressure is due to the decreasing Ru ferromagnetic moment below T_f . Lynn *et al* [4] suggested, from a neutron diffraction study, that the magnetic order of the Ru is predominantly antiferromagnetic and the component of ferromagnetism for Ru1212 was produced by a canting originating from the slight rotation of the RuO_6 octahedra around an axis perpendicular to c . It is known that hydrostatic pressures improve the crystal symmetry—for example, the phase transition from orthorhombic to tetragonal structure for the La–Sr–Cu–O system. If the decrease in ferromagnetic moment with increasing pressure occurs due to the crystal symmetry, the ferromagnetism for Ru1212 may disappear in a higher-pressure region.

In summary, the results of a crystal structure analysis for Ru1212 indicated Cu sites containing 5% Ru and the Cu–O1 bond length which was calculated from the structural parameters for this sample is 2.15 Å. This bond length is unusually short compared to that in optimally doped oxide superconductor. T_c scarcely varies under pressure. T_f increases with increasing pressure at lower pressure. However, the increasing of the ferromagnetic transition temperature became saturated above 1.6 GPa. The magnitude of the susceptibility decreases with increasing pressure below T_f .

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