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$EuSr_2RuCu_2O_8$ 的超导电性和磁性

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摘　要　研究了铁磁性超导体 $EuSr_2RuCu_2O_8$ 的结构、超导电性和磁性. 结果表明, 该化合物为欠掺杂超导体, 其超导转变起始温度为 T_c^{onset}=35K, 零电阻温度为 $T_c(\rho=0)$=10K, 铁磁转变居里温度为 T_c=130.2K.

关键词　超导电性　铁磁性　$EuSr_2RuCu_2O_8$ 化合物
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SUPERCONDUCTIVITY AND MAGNETIC PROPERTY IN $EuSr_2RuCu_2O_8$

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ABSTRACT　X-ray diffraction, resistivity, magnetization measurements were carried out for the ferromagnetic superconductor $EuSr_2RuCu_2O_8$. The temperature dependence of resistivity shows that this compound is an under-doped superconductor with T_c^{onset}=35K, $T_c(\rho=0)$=10K. The magnetization data shows that this compound exhibits the ferromagnetic transition at T_c=130.2K.

KEY WORDS　superconductivity(SC), ferromagnetism(FM), $EuSr_2RuCu_2O_8$ compound

1. Introduction

The problem of coexistence of superconductivity (SC) and ferromagnetism (FM) has attracted considerable attention since the original works of Ginzburg[1] and Matthias et al[2]. It was shown that singlet SC and FM are mutually exclusive and the SC can also be strongly suppressed by magnetic impurities. The competition between FM and SC were observed in the ternary compounds $HoMo_6S_8$, $HoMo_6Se_8$ and $ErRh_4B_4$.[3] But true microscopic coexistence was found only over a narrow temperature region when FM sets in and modifies itself to a spiral or domain-like structure.

The copper-ruthenium oxide $GdSr_2RuCu_2O_8$ has been reported to display uniformly coexisting ferromagnetism and superconductivity below 46K[4,5], however, some experimental studies

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have pointed towards antiferromagnetic order of Ru atoms below T_M ≈132K.Up to now, there is no clear understanding on the interaction of competing antiferromagnetic, ferromagnetic, and superconducting order parameters in $GdSr_2RuCu_2O_8$.

In this paper, we report the study on superconductivity and magnetic properties of $EuSr_2RuCu_2O_8$ where nonmagnetic Eu has replaced magnetic Gd. The merit of $EuSr_2RuCu_2O_8$ is that the magnetic behavior of the ruthenate lattice is not hidden by the magnetism from Gd [moment of 7.94μB and ordering temperature of 2.5K[4]].

2. Experimental Methods

$EuSr_2RuCu_2O_8$ sample was prepared by thoroughly reacting the mixture of Eu_2O_3, $SrCO_3$, RuO_2 and CuO, with the cation composition of Eu:Sr:Ru:Cu=1:2:1:2. These powders were high-grade purity (99.9%). The mixture was milled, decomposed at 960 ℃ in air for 24 hours. It was then ground milled, and die-pressed into pellets. The first sintering step took place in a flowing N_2 atmosphere at 1010 ℃. This step results in the forming of a mixture of the precursor material $Sr_2EuRuO6$ and Cu_2O and is directed towards minimizing the formation of $SrRuO_3$. The material was then reground and was sintered in a flowing O_2 for 4 days at 1060 ℃. Finally the sample was cooled slowly to room temperature in a flowing O_2 atmosphere.

The temperature dependence of resistivity was measured by the standard four-lead technique. The magnetization measurements on solid ceramic pieces in the range of 2~300K were performed in a commercial (Quantum Design) superconducting quantum interference device magnetometer (SQUID). The magnetization was measured by two different procedure: (1) the sample was zero-field-cooled(ZFC) to 2K, a field was applied and magnetization was measured as s function of temperature, (2) the sample was field-cooled (FC) from above 300K to 2K.

3. Results and Discussion

Fig.1 shows that $EuSr_2RuCu_2O_8$ sample is of a single phase and has a tetragonal structure with the space-group P4/mmm. The TEM and EDX experimental show that there is no impurity such as $SrRuO_3$ in the sample. The lattice parameters a (0.3841nm) and c (1.1566nm) were obtained from the X-ray diffraction pattern.

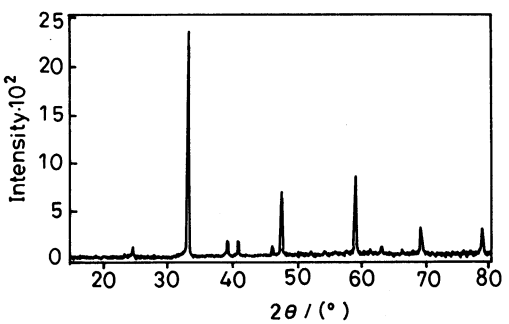

Fig.1 XRD pattern for $EuSr_2RuCu_2O_8$ sample

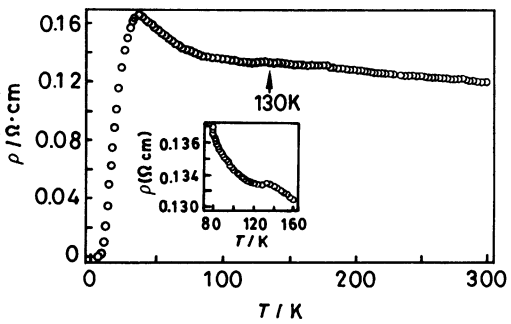

Fig.2 Temperature dependence of resistivity for $EuSr_2RuCu_2O_8$ sample

It can be seen from Fig.2 that the resistivity increases slowly with decreasing of temperature as semiconducting behavior above 130K. However, at near 130K, there is a small decrease of resistivity, which may be due to the ferromagnetic transition at this temperature as discussed as following. This behavior is similar to that in $SrRuO_3$, for which there is also a small decrease of resistivity at Curie temperature (T_c=165K)[6]. After this small decrease, resistivity increases again up to the maximum value at about 35K. And then an abrupt decrease of resitivity and zero resistivity was observed at 10K. This result showes that the sample is a superconductor with T^{onset}=35K, $T_c(\rho=0)$=10K. The shape of ρ(T) shown in Fig.2 is characteristic of an underdoped cuprate.

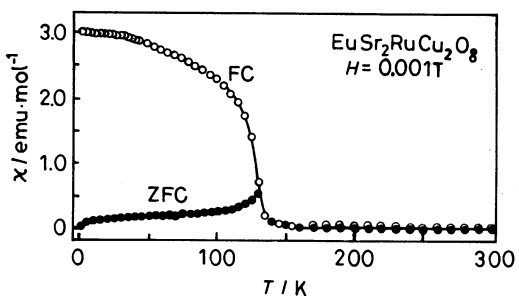

Fig.3 Temperature dependence of molar susceptibility χ(T), measured in 0.001T in ZFC and FC processes, respectively, for $EuSr_2RuCu_2O_8$ sample

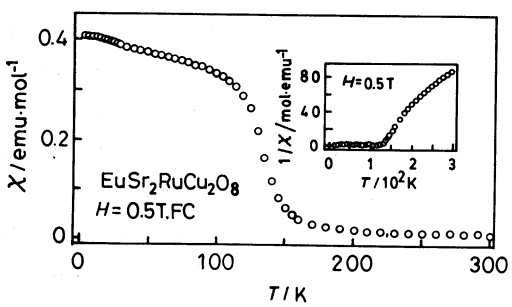

Fig.4 Temperature dependence of molar susceptibility χ(T), measured in 0.5T in FC process for the same sample. Inset : The temperature dependence of reciprocal susceptibility, the solid line shows the fit of Eq.(1) in the text for $EuSr_2RuCu_2O_8$ sample

The χ(T) curve (in Fig.3) measured with ZFC process exhibits a decrease of at about 35K, and a magnetic anomalous peak at about 130K. The former is due to the appearance of superconductivity below 35K, as shown in Fig.2. A similar decrease was observed at 32K in $Eu_{1.4}Ce_{0.6}RuSr_2Cu_2O_{10-\delta}$[7,8], at 42K in $Gd_{1.4}Ce_{0.6}RuSr_2Cu_2O_{10-\delta}$[8], and 45K in $GdSr_2RuCu_2O_8$[9,10]. It has been argued that this feature is due to the appearance of superconductivity, where the bulk Meissner phase is suppressed because the small negative magnetization resulting from intragranular supercurrents is swamped by the positive contribution from the ferromagnetic Ru. The latter anomalous peak is related to the ferromagnetic transition at near 130K. This ferromagnetic transition is also evident from the sudden onset of a magnetic susceptibility at about 130K as shown in the χ(T) curve measured at 10G with FC process. If the ferromagnetic transition T_c is defined as the temperature at which $d\chi/dT$, measured in a field of 10G with FC process, has the maximum value, T_c ($\approx$130.2K) was obtained from the χ(T) curve. The third feature in Fig.3 is that there is a significant difference between the susceptibilities measured with FC and ZFC processes. This is the characteristic of ferromagnet.

The magnetic susceptibility measured in a field of 0.5T (Fig.4) is similar to that measured in a field of 0.001T. Because there are Eu^{3+} ions in the compound, the susceptibility should contain the Van Vleck susceptibility χ_{vv}[11] due to the mixing of J=0, J=1 and J=2 level plus the thermal population of higher multiplets. The total susceptibility can be given by

$$\chi_{\text{total}} = \frac{C}{T-\theta} + \chi_0 + \chi_{vv} \tag{1}$$

where C is the Curie constant, θ is the paramagnetic Curie temperature, χ_0 is the temperature independent susceptibility. The inset of Fig.4 shows the temperature dependence of the reciprocal magnetic susceptibility measured at 0.5T with FC process for the same sample. The fit of Eq.(1) is represented by the solid line in the inset of Fig.4. It is clear that χ(T) in paramagnetic region can be described by the modified Curie–Weiss law and Van Vleck susceptibility term of Eu^{3+} ions. From this fitting, we can conclude the Curie temperature θ=137.5K, and temperature independence susceptibility χ_0=1.37×10^{-3}emu/mol.

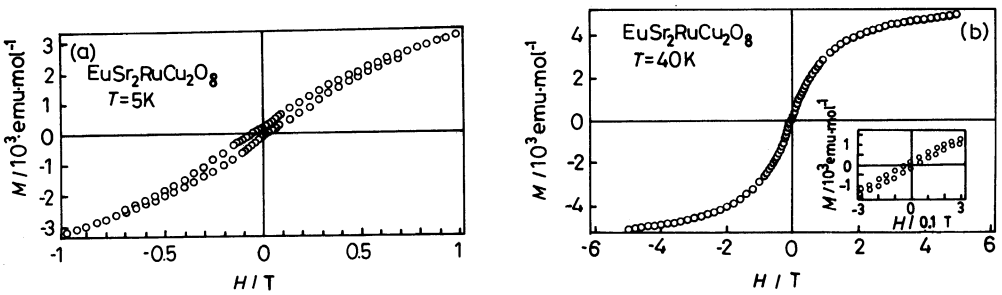

Fig.5 Isothermal magnetization loops at 5K (a) and 40K (b)

The M–H curves in Fig.5 show small hysteresis in the low–field region, and the hysteresis becomes larger with decreasing temperature. It is expected that this hysteresis results from the motion of the ferromagnetic domain walls, since the sample was measured in a ferromagnetic state below T_c. And the increase of magnetization by external magnetic field in higher field at low temperature indicates that this compound is an itinerant–electron ferromagnet, which is similar to $SrRuO_3$[12].

4. Conclusion

$EuSr_2RuCu_2O_8$ compound which has the similar structure to that of Y–123 exhibits ferromagnetism below 130.2K, and becomes superconducting below 10K.

References

1 V.L.Ginzburg, Zh.Eksp.Teor. **31**, 202(1956) [Sov.Phys.JETP **4**, 153(1957)]

2 B.T.Matthias, H.Suh, E.Corenzwit, Phys.Rev.Lett., **1**, 449(1958)

3 M.B.Maple, O.Fisher, Superconductivity in Ternary Compounds (Berlin, Springer–verlag Press, 1982) p.11

4 C.Bernhard, J.L.Tallon, Ch.Niedermayer, Th.Blasius, A.Golnik, E.Brücher, R.K.Kremer, D.R.Noakes, C.E.Stronach, E.J.Ansaldo, Phys.Rev.B **59**, 14099(1999)

5 J.W.Lynn, B.Keimer, C.Ulrich, C.Bernhard, J.L.Tallon, Phys.Rev.B **61**, 14964(2000)

6 J.J.Bouchard, J.L.Gillson, Mater.Res.Bull, **7**, 873(1972)

7 I.Felner, U.Asaf, Y.Levi, O.Millo, Phys.Rev. B **55**, 3374(1997)

8 E.B.Sonin, I.Felner, Phys.Rev. B **57**, 14000(1998)

9 J.L.Tallon, J.W.Loram, G.V.M.Williams, C.Bernhard, Phys.Rev. B **61**, 6471(2000)

10 C.Bernhard, J.L.Tallon, E.Brücher, R.K.Kremer, Phys.Rev.B **61**, 14960(2000)

11 J.H.van Vleck, The Theory of Electric and Magnetic Susceptibilities, (London, Oxford University Press, 1965) p.245

12 S.Uchida, Physica C 185∼189, 28(1991)