

Superconductivity and magnetism in $\text{Ru}_{1-x}\text{Ir}_x\text{Sr}_2\text{GdCu}_2\text{O}_8$ compounds

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Abstract

We carried out an investigation of the magnetic and superconducting properties of $\text{Ru}_{1-x}\text{Ir}_x\text{Sr}_2\text{GdCu}_2\text{O}_8$ compounds for Ir concentrations up to $x = 0.25$, which is the limit of solubility. This study consisted in the X-ray diffraction analysis of these materials, and the measurements of the temperature dependence of the electrical resistivity, and of the ac and dc magnetic susceptibilities. The substitution of Ir for Ru moves the magnetic transition of the Ru-sublattice near 130 K to lower temperatures, and it suppresses superconductivity fairly rapidly. The zero-resistance state drops from ~ 32 K for the pure Ru-1212 compound to ~ 13 K for $x = 0.10$. Further addition of Ir causes a gradual evolution from metallic to non-metallic behavior, with progressively larger upturns at low temperatures.

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The synthesis of the two ruthenium-based layered cuprates with general composition $\text{RuSr}_2\text{LnCu}_2\text{O}_8$ and $\text{RuSr}_2(\text{Ln}_{1+x}\text{Ce}_{1-x})\text{Cu}_2\text{O}_{10}$, where Ln = Sm, Eu, and Gd [1,2], triggered a great deal of interest in these materials, since they both display coexisting superconductivity (SC) and long-range magnetic order, including weak ferromagnetism (wFM) at moderately high temperatures (T). The coexistence of SC and wFM occurs over a T -span of about 40 K [3–5]. The $\text{RuSr}_2\text{LnCu}_2\text{O}_8$ compound (Ru-1212) is closely related structurally to $\text{LnBa}_2\text{Cu}_3\text{O}_7$ (Ln-123), in which a layer of vertex sharing RuO_6 octahedra replaces the “charge reservoir” CuO_4 squares of the 123-structure. It has been suggested that the coexistence of SC and wFM in these rutheno-cup-

rates can be sustained possibly because of the weak coupling between the Ru–O magnetically ordered layers and the SC Cu–O planes, leading to a SC order parameter of the Fulde–Ferrel–Larkin–Ovchinnikov type [6]. The behavior of the electrical resistivity ρ vs T in the normal phase, the value of the SC transition temperature (T_c), and the width of the SC transition are strongly dependent of the synthesis process in general, and of the oxygenation process in particular [1,2]. The onset of SC for the Ru-1212 $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ compound is ~ 45 K, while the value of the Curie temperature (T_C) is ~ 132 K [3,4].

In order to probe the coexistence of wFM and SC in Ru-1212-Gd compounds, by means of diluting the Ru magnetic sublattice, we carried out an investigation of Ir substituted materials with composition $\text{Ru}_{1-x}\text{Ir}_x\text{Sr}_2\text{GdCu}_2\text{O}_8$. The specimens for this work were synthesized from high purity powders of Ru, Ir, SrCO_3 , Gd_2O_3 , and CuO, using two different procedures: (1) the stoichiometric amounts of the powders were

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ground together extensively, and reacted in alumina crucibles at 970 °C for several days with several intermediate grindings, followed by pelletizing and annealing for 7 days at 1060 °C in flowing O₂; and (2) by synthesizing the Sr₂GdRu_{1-x}Ir_xO₆ (Sr-2116) precursors [7], followed by adding CuO, pelletizing and annealing for several days at 1060 °C in flowing O₂. Although the phase purity, and magnetic ordering characteristics yielded in the two processes were similar, the SC properties of the specimens produced from the Sr-2116 precursors were much superior and considerably more reproducible.

X-ray diffraction analysis suggests that the limit of solubility when Ir substitutes Ru in the Ru-1212-Gd materials is about 25%, a composition in which minute amounts of Sr-2116 and SrRu(Ir)O₃ start to become more noticeable. The lattice parameters are not sensitive to the Ir substitutions. The effect of the partial substitution of Ir in the magnetic ordering can be seen in Fig. 1, which displays the temperature dependence of the ac susceptibility for samples with iridium content 0 ≤ x ≤ 0.2. Taking the ordering temperature from the peaks in the χ_{ac} vs T data, we see that the magnetic ordering temperature drops from ~131 K in the pure material to ~109 K for x = 0.2. Similarly, the χ_{dc} vs T data (not shown) reveal that the magnetic transition is shifted to lower temperatures, and that it smears out as a result of Ir substitutions. The strong effect of the substitutions on the magnetic ordering temperature suggests that Ir occupies the Ru site in the structure. The effect of the Ir substitutions on the temperature dependence of the electrical resistivity is displayed in Fig. 2. The onset of SC in the undoped Ru-1212-Gd

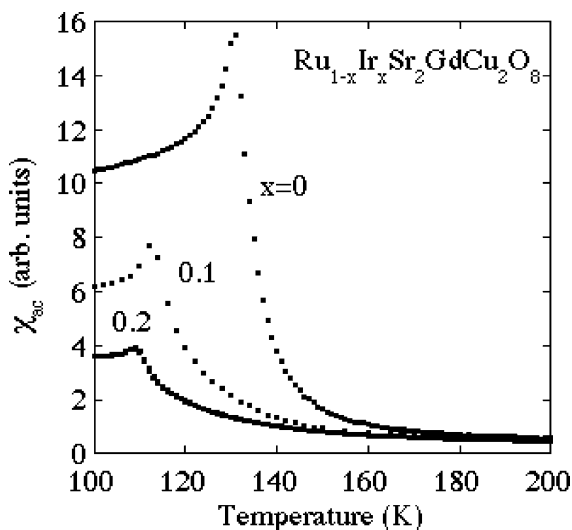


Fig. 1. ac magnetic susceptibility as a function of temperature in Ru_{1-x}Ir_xSr₂GdCu₂O₈ compounds, with x = 0, 0.1, and 0.2.

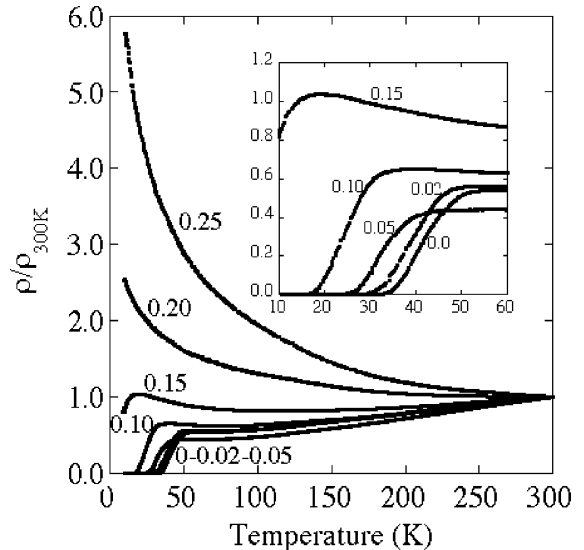


Fig. 2. Normalized electrical resistivity $\rho/\rho_{300\text{K}}$ vs temperature in Ru_{1-x}Ir_xSr₂GdCu₂O₈ compounds with 0 ≤ x ≤ 0.25. The inset shows the superconducting transition for the compounds with x < 0.15 in detail.

sample is $T_{c,\text{onset}} \sim 52$ K, while the zero-resistance state is reached at $T_{c,\text{zero}} \sim 32.5$ K. The values of $T_{c,\text{onset}}$ and $T_{c,\text{zero}}$ both drop precipitously with Ir substitutions, as indicated in Fig. 2, suggesting that Ir can change the optimal doping of charge carriers. For Ir contents $x \geq 0.2$, the ρ vs T data display semiconductor-like behavior.

In summary, we have shown that Ir can be partially substituted for Ru in the Ru-1212 materials up to 25% without affecting the formation of the crystallographic phase. The Ir substitution results in the depression of both the magnetic and superconducting transition temperatures, ultimately inducing non-metallic behavior.

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References

- [1] L. Bauernfeind et al., Physica C 254 (1995) 151.
- [2] L. Bauernfeind et al., J. Low Temp. Phys. 105 (1996) 1605.
- [3] J.L. Tallon et al., IEEE Trans. Appl. Supercond. 9 (1999) 1696.
- [4] C. Bernhard et al., Phys. Rev. B 59 (1999) 14099.
- [5] I. Felner et al., Phys. Rev. B 55 (1997) R3374.
- [6] W.E. Pickett et al., Phys. Rev. Lett. 83 (1999) 3713.
- [7] T.P. Papageorgiou et al., Physica C 377 (2002) 383.