ELSEVIER

Physica C 341–348 (2000) 481–482

PHYSICA C

www.elsevier.nl/locate/physc

Electrical and magnetic properties of $Ru_{1-x}M_xSr_2GdCu_2O_8$ (M = Ti, V and Nb)

J. T. Rijssenbeek[a], N. Mansourian-Hadavi[b], S. Malo[a], D. Ko[a], C. Washburn[b], A. Maignan[c], D. Pelloquin[c], T. O. Mason[b] and K. R. Poeppelmeier[a]

[a]Department of Chemistry and the Science and Technology Center for Superconductivity, Northwestern University, Evanston, IL 60208, U.S.A.

[b]Department of Material Science and Engineering, Northwestern University, Evanston, IL 60208, U.S.A.

[c]Laboratoire CRISMAT-ISMRA, UMR 6508, Université de Caen, France

Samples of $Ru_{1-x}M_xSr_2GdCu_2O_8$, with M = Ti, V and Nb; and $0 \leq x \leq 1$, were synthesized to shed light on the role of the RuO_2 planes in this 1212-type compound which is a ferromagnetic superconductor (T_c = 10-40 K) when x = 0. We show that the ferromagnetism and the superconductivity are weakened for M = Ti and Nb, and enhanced for M = V, although superconductivity is suppressed for all samples of higher doping levels (x > 0.2). Jonker analysis of the high-temperature electrical properties of $RuSr_2GdCu_2O_8$ shows that it falls within the characteristic range for other known superconductors.

1. INTRODUCTION

The remarkable coexistence of ferromagnetism and superconductivity in $RuSr_2GdCu_2O_8$ has made this material the focus of much recent research [1-4]. The RuO_2 layer in this material is at the origin of both the ferromagnetism as well as the carrier-creation mechanism in the CuO_2 planes, via some $Ru^{5+/4+}/Cu^{2+/3+}$ charge transfer. We have undertaken substitution studies in the RuO_2 sublattice to elucidate the nature of these phenomena. Niobium (V) and titanium (IV) were chosen because their d^0 configurations should weaken the magnetic interactions in the RuO_2 planes. Vanadium was chosen for its ability to mimic the mixed-valency of Ru^{4+}/Ru^{5+}, but with a different spin configuration. High-temperature thermopower and conductivity measurements were performed to compare these results with those of other known superconductors.

2. EXPERIMENTAL

Stoichiometric amounts of $SrCO_3$, Gd_2O_3, CuO, RuO_2 and $V_2O_5/Nb_2O_5/TiO_2$ powders were thoroughly ground, pelletized and then fired at 950 °C for 12 hours to decompose the carbonates. Subsequently, the temperature was increased to 1040 °C and maintained for 72 hours followed by cooling to room temperature over 12 hrs. Electron diffraction (ED) studies and EDS analyses were carried out using an 8100 Hitachi electron microscope equipped with a KEVEX analyzer. The superconductivity was characterized under true zero-field cooled conditions by AC susceptibility measurements performed using an AC-7000 Lake Shore Susceptometer with AC magnetic fields ranging from 10^{-2} to 3 Oe at a constant frequency of 133 Hz. The same instrument was used to measure the temperature dependent resistance via the classical four-probe method. Simultaneous four-point DC conductivity and Seebeck coefficient measurement were performed at various oxygen partial pressures (10^{-5}-1 atm) and different temperatures for Jonker analysis [5].

3. RESULTS AND DISCUSSION

X-ray diffraction and EDS analyses of $Ru_{1-x}M_xSr_2GdCu_2O_8$ show that the solubility behaviors differ markedly between M = Nb, and M = Ti and V. The former displays a complete solid solution with smoothly increasing lattice parameters as expected for substitution by the larger cation. The latter two cations have very limited solubilities (~5% and ~15%, for M = V and Ti respectively) as evidenced by the appearance of $SrMO_{3-\delta}$ at higher substitution levels.

PII S0921-4534(00)00553-0

3.1. Magnetism

The ferromagnetic transition (T_C = 136 K) and superconducting transition (T_c = 15 K) of the pure compound are in good agreement with previous reports [1-4]. These measurements confirm the coexistence of ferromagnetism and superconductivity in both the pure and nominally $Ru_{0.9}M_{0.1}$ samples. The highest T_c is exhibited for the V-substituted sample with an onset at 19 K, whereas those of Ti and Nb are 9 K and 13 K, respectively. A concurrent increase in the ferromagnetism is observed with vanadium substitution while a decrease is observed for Ti and Nb (Fig. 1). Temperature-dependent resistivity measurements confirm the improvement of the critical temperature from 20 K to 25 K.

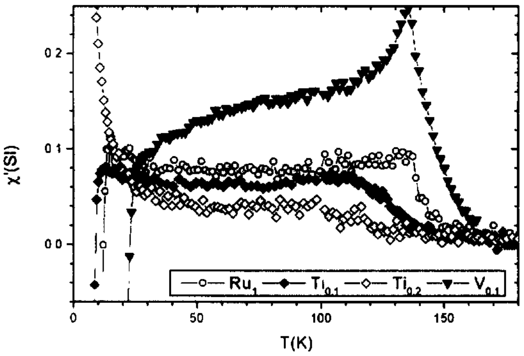

Figure 1. Real part (χ') of the AC susceptibility versus temperature registered with $H_{ac} = 10^{-2}$ Oe and f = 133 Hz. The $Ru_{1-x}Nb_x$ data are nearly identical to those of $Ru_{1-x}Ti_x$ and are omitted for clarity [6].

3.2. Thermopower

Jonker analysis, in which the thermopower (Q) is plotted versus the natural logarithm of conductivity, is used to examine the superconducting behavior of Ru-1212. The location and size of the "pear" shaped curve, the region of the curve where the data fall, the sign of the thermopower, and the spread of the data, all give a great deal of information with respect to the internal chemistry of a given material. The thermopower vs. ln conductivity data for Ru-1212 fall in the extrinsic region of the pear with very low thermopower values (~22 μV/K) typical of a superconducting material (Fig. 2). The narrow data distribution along the high conductivity leg of the curve, i.e. a smaller dependence of the electrical properties on the oxygen partial pressures, is further evidence for the self-doped and oxygen-stoichiometric character of the Ru-1212 material. Such behavior was also observed in $YBa_2Cu_4O_8$ (Y-124), another oxygen-stoichiometric superconductor [7].

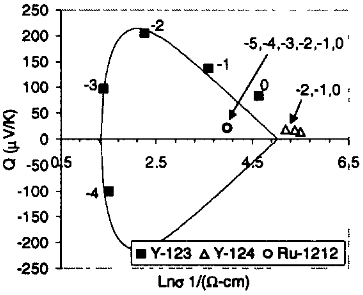

Figure 2. Comparative Jonker plot of the Ru-1212, Y-123 and Y-124 systems at 700 °C for different oxygen partial pressures (log values mentioned on the plot). Note the difference between the oxygen partial pressure dependence for the three compounds.

4. Conclusions

The results of these doping experiments indicate that there is no clear relationship between the ferromagnetism and the superconducting properties. Furthermore, the RuO_2 layer is believed to act as a hole reservoir for the CuO_2 planes and the fact that chemical substitutions in this layer rapidly destroy the superconductivity for $x > 0.1$, demonstrates that this charge creation mechanism is very fragile. The high-temperature thermopower data highlight the self-doped character of Ru-1212.

REFERENCES

1. L. Bauernfeind, W. Widder and H.F. Braun, J. Low Temp. Phys., 105 (1996) 1605.
2. C. Bernhard, J. L. Tallon, Ch. Niedermayer, Th. Blasius, A. Gonik, E. Brücher, R.K. Kremer, D.R. Noakes, C.E. Stronach and E.J. Ansaldo, Phys. Rev. B, 59 (1999) 14099.
3. D.J. Pringle, J.L. Tallon, B.G. Walker and H.J. Trodahl, Phys. Rev. B, 59 (1999) R11679.
4. A.C. McLaughlin, W. Zhou, J.P. Attfield, A.N. Fitch and J.L. Tallon, Phys. Rev. B, 60 (1999) 7512.
5. G.H. Jonker, Philips Res. Rep., 23 (1968) 131.
6. S. Malo, D. Ko, J.T. Rijssenbeek, A. Maignan, D. Pelloquin, V.K. Dravid and K.R. Poeppelmeier, Int. J. Inorg. Mater., submitted
7. B-S. Hong and T. O. Mason, J. Mater. Res., 6 (1991) 2054.