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Effect of Ir substitution in the ferromagnetic superconductor $RuSr_2GdCu_2O_8$

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Abstract

A detailed study of the effect caused by the partial substitution of Ru by Ir on the magnetic and superconducting properties of the ruthenocuprate $Ru_{1-x}Ir_xSr_2GdCu_2O_8$; $0 \leqslant x \leqslant 0.10$; is presented. The combined experimental results of structural, electrical, and magnetic measurements indicate that Ir substitutes Ru for $x \leqslant 0.10$ with no significant structural distortions. Ir-doping gradually suppresses both the magnetic and the superconducting states. However, all samples were observed to attain the zero-resistance state at temperatures $\geqslant 2$ K up to the highest applied magnetic field of 18 T. The resistive upper-critical field H_{c2} as a function of temperature has been determined for these polycrystalline samples. Values of $H_{c2}(0)$ were found to be ~ 52 T, and weakly dependent on the Ir concentration. We have also observed that the superconducting transition width decreases and the slope of the resistive transition increases with increasing Ir doping, a feature which is much more pronounced at high applied magnetic fields. The double-peak structure observed in the derivative of the resistive curves has been related to an inhomogeneous nature of the physical grains which is enhanced due to the Ru substitution by Ir. This indicates that the Josephson-junction-array (JJA) model seems to be appropriated to describe the superconducting state in these ruthenocuprates. The low temperature $\rho(T)$ data along with the determined vortex thermal activation energy are consistent with a 2D vortex dynamics in these materials.

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1. Introduction

The study of the coexistence of superconductivity and magnetic ordering in the ruthenocuprate $RuSr_2GdCu_2O_8$ (Ru-1212) has attracted great interest since the original study of Bauernfeind et al. [1]. The Ru-1212 is a 1212-type layered cuprate structurally similar to the $YBa_2Cu_3O_{7-\delta}$-(YBCO), where Y and Ba are replaced by Gd (or Eu) and Sr, respectively, and the Cu–O chains replaced by RuO_2 layers [1,2]. In these materials the magnetic long-range ordering of the Ru sub-lattice occurs below a transition temperature $T_M \sim 130$ K while the superconductivity arising from the CuO_2 layers occurs below a critical temperature $T_c \sim 40$ K. Recent studies have indicated that the magnetic ordering and the superconducting state are essentially decoupled, being related only by the charge transfer between Ru and CuO planes [3]. In addition to this, there

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is no clear evidence about the exact nature of the magnetic structure of these materials up to now but it is accepted that, for low magnetic fields, there is an antiferromagnetic (AFM) order, whereas for high magnetic fields ($H \sim 2$ T) a spin-flop transition is observed [4], with Ru magnetic order essentially ferromagnetic (FM). Also, from experimental data and theoretical analysis, it was proposed that for temperatures lower than T_c, the magnetic-flux lines are present even without an external magnetic field, suggesting the creation of a spontaneous vortex phase (SVP) [5,6].

In fact, the genuine coexistence of superconductivity and magnetism at microscopic level is still controversial. Some experimental studies [7] have shown that the ruthenocuprates are microscopically uniform. On the other hand, several experimental results have indicated a possible phase-separation of superconducting (SC) and magnetic regions [8–10]. For instance, high-resolution transmission electron microscopy (HRTEM) and synchrotron X-ray diffraction analysis have suggested a phase separation in Ru-1212 compounds [8]. It was argued that such a phase separation arises from the rotation of the RuO_6 octahedra around the *c*-axis, resulting in the formation of small domains with characteristic lengths $\leqslant 200$ Å separated by sharp antiphase boundaries of reversed rotations [8]. Also, a phase separation between FM and AFM nanodomains inside physical grains of Ru-1212 has been proposed from a detailed analysis of magnetization data [9,10]. The authors have concluded that intragrain properties of the ruthenocuprates exhibit features of granular superconductors and a Josephson-junction-array (JJA) model was invoked to account for the intrinsic inhomogeneities of intragrain superconductivity [9,10]. Therefore, a discussion of whether both superconducting and magnetic phases originate from the same crystallographic structure, and features of this intimate coexistence on a microscopic scale are relevant questions for the understanding of these materials.

Considering that the coupling allowing for the coexistence of superconductivity and ferromagnetism in Ru-1212 compounds is very weak and strongly affected by chemical substitutions, the dilution of the magnetic Ru sublattice by different ions is an interesting approach to probe the coexistent phenomena. The partial substitution of Ru by Sn^{4+} was found to suppress the FM moment of the sublattice and to increase the onset of the SC transition. These features would reflect an increase in the transfer rate of holes to the CuO_2 planes [11]. Studies regarding substitution of Ru by both Ti and Rh revealed that both FM and SC transition temperatures are reduced upon increasing dopant concentration [12]. The substitution of Ru by Nb^{5+} results in a decrease of the magnetic ordering temperature and an increase in the Ru valence [13], whereas for Ta-substituted specimens an apparent suppression of the superconductivity of Ru-1212 has been observed [14]. In general, both magnetic and superconducting properties of the ruthenocuprates are affected by the ionic radius, valence, and magnetic character of the substituting ion. However, changes observed in alloying Ru-1212 compounds are usually accompanied by significant structural distortions due to differences in ionic radii. Within this scenario it is a difficult task to distinguish between changes arising from properties of the substituted ion and those from crystallographic distortions.

In the present work we have investigated the crystallographic, transport, and magneto-transport properties of $Ru_{1-x}Ir_xSr_2Gd_1Cu_2O_8$ compound in order to study the relationship between superconductivity and magnetism. We have found that Ir substitutes Ru up to 10% in Ru-1212 without appreciable structural changes. In addition to this, the combined data indicate a possible phase-separation in the Ru-1212 compound.

2. Experimental

Polycrystalline samples of $Ru_{1-x}Ir_xSr_2GdCu_2O_8$ (Ru(Ir)-1212); $0 \leqslant x \leqslant 0.10$; were prepared following a two-step procedure [15]. The two-step synthesis minimizes the formation of the $SrRuO_3$ phase, yielding samples with better quality [16]. Initially, the $Sr_2GdRu_{1-x}Ir_xO_6$ (Sr-2116) precursor was prepared by mixing stoichiometric quantities of high purity Ru, Ir, $SrCO_3$, and Gd_2O_3, grinding together and heating in air at 1250 °C for 12 h. Then, CuO was mixed to the Sr-2116 powders, ground together, pressed into pellets, and sintered at 1060 °C for 72 h in flowing O_2. The crystal structure of the samples was analyzed by X-ray powder diffraction (XRD) measurements using CuK_α radiation on a Bruker D8 Advance diffractometer. The diffraction patterns were collected in the 2Θ range 20–80° with a step of 0.01 and 8 s counting time. Rietveld refinements of crystal structures were performed using the GSAS software. The temperature dependence of the magnetoresistance $\rho(H,T)$ was measured by the standard four-probe method using a Linear Research Model LR-700 bridge operating at 16 Hz. In all transport measurements, copper electrical leads were attached to Ag film contact pads (made with Ag epoxy) on parallelepiped-shaped samples with typical dimensions of $5 \times 2 \times 1.5\ mm^3$. The magnetoresistance experiments were performed at the National High Magnetic Field Laboratory, Los Alamos, in the temperature range from 2 to 300 K and under magnetic fields H up to 18 T. Measurements at low applied magnetic fields H up to 0.5 T were performed in a home-made apparatus using a superconducting coil with very low remnant field. The samples were characterized by both magnetization $M(T)$ and ac magnetic susceptibility $\chi_{ac}(T)$ using a SQUID magnetometer from Quantum Design. Magnetization measurements in the remnant field ($\sim$1 Oe) of the superconducting magnet were performed in the temperature range from 5 to 300 K in both zero-field-cooled (ZFC) and field-cooled (FC) modes. The T-dependence of the ac magnetic susceptibility ($f = 155$ Hz) was measured with an excitation field of 2 Oe.

3. Results and discussion

The X-ray diffraction patterns of $Ru_{1-x}Ir_xSr_2GdCu_2O_8$-(Ru(Ir)-1212) for $x = 0.00$ and $x = 0.10$ are displayed in

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