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## Magnetic field dependence of the intragrain transition in $RuSr_2GdCu_2O_8$

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## Abstract

A characterization of the magnetic superconductor  $RuSr_2GdCu_2O_8$  through resistance measurements as a function of temperature and magnetic field up to 9 T is presented. The intragrain transition shows a two-step behavior with the increase in field, which is explained as a consequence of the effects of a spin-flop transition of the Ru-sub-lattice on the boundaries connecting intragranular structural domains.

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The onset of bulk superconductivity in the presence of a ferromagnetic (FM) component [1,2] in RuSr<sub>2</sub>-GdCu<sub>2</sub>O<sub>8</sub> [Ru-(1212)] makes this compound particularly suitable to study the interplay between these usually exclusive phenomena. In this work we show that the results obtained from a careful study of the derivative of the resistive superconducting (SC) transition curves in the presence of a DC magnetic field bias (Fig. 1) can be explained in terms of the effect of the magnetization of the Ru-sub-lattice on the antiphase boundaries connecting structural domains of coherently rotated RuO<sub>6</sub> octahedra [3]. Also, ac magnetic susceptibility  $\chi(T,H)$ measurements were performed with the same superimposed DC fields, looking for a correlation between transport and magnetic properties. Sharp SC transitions were observed for all the applied fields, as shown in Fig. 2(a). An expanded section of the SC transition region is displayed in Fig. 2(b).

Polycrystalline samples were prepared by conventional solid-state reaction. The room temperature X-ray diffraction pattern corresponds to Ru-(1212), with no

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spurious lines being observed. Two broad overlapped peaks in the derivative curves were observed at temperatures  $T_1$  and  $T_2$ . These peaks and the thermodynamic transition temperature  $T_{\text{th}}$  are identified by arrows for the H = 0 curve in Fig. 1.

The interval  $\Delta T_{\text{th},1} = T_{\text{th}} - T_1(0)$  is ~12 K, which is about one order of magnitude higher than for conventional cuprates. Traditionally, the existence of two peaks has been interpreted as intra- ( $T_1$ ) and intergrain ( $T_2$ ) SC transitions [4]. The increase in H up to 0.1 T leaves peak 1 unchanged in position, width and amplitude. For H > 0.3 T,  $T_1$  smoothly diminishes with an initial slope of ~7 K/T, while the peak strongly broadens. The behavior of  $T_2$  is analyzed elsewhere [5].

We explain the large  $\Delta T_{\text{th},1}$  interval for H = 0 in terms of spontaneous vortex phase (SVP) formation followed by flux expulsion in the structural domains. For temperatures below and near  $T_{\text{SC}}$  the internal field of the Ru sub-lattice is higher than the first critical field, and vortex lines are created. On cooling, the first critical field of the domains increases. When it becomes higher than the internal magnetization, Meissner effect occurs in the domains, with a partial expulsion of the vortex lines, leading to flux compression at the anti-phase boundaries. The result is a complex thread of magnetic



Fig. 1. The temperature dependence of the derivative of the resistive superconducting transition curves for RuSr<sub>2</sub>GdCu<sub>2</sub>O<sub>8</sub>. The DC magnetic fields *H* are indicated. The intragrain ( $T_1$ ) and intergrain ( $T_2$ ) peaks, and the thermodynamic transition temperature  $T_{\text{th}}$  are identified by arrows for H = 0 T. The lines are guides to the eyes.



Fig. 2. a) The temperature dependence of the AC magnetic susceptibility for  $RuSr_2GdCu_2O_8$ ; (b) an expanded section of the region of the superconducting transition. The DC magnetic fields are the same used in the resistance measurements. The lines are guides to the eyes.

field lines across the intragrain network, generating a variety of local effective fields. If the boundaries act as Josephson junctions, the structural domains will gradually become phase-locked as the temperature is decreased, until a maximum rate in this percolation process is reached at  $T_1$ .

The fact that peak 1 remains unchanged up to H = 0.1 T suggests that an external field of this strength has a little effect on the magnetization of the RuO<sub>2</sub> layers, which determines the vortex phase structure. The effect of the external field could be important if it promotes a re-arrangement in the magnetic order of the Ru moments. We recall here that when a magnetic field is applied to a Ru-(1212) sample, a change is observed in the neutron diffraction pattern at H = 0.4 T, a result interpreted as due to a spin-flop transition [6]. Magnetic measurements indicate that such transition should occur at a critical field of ~0.14 T [7]. These values are near to the fields at which we observed the onset of the decrease in  $T_1$  in Ru-(1212).

The  $\chi(T, H)$  curves also show a change in behavior for  $H \simeq 0.1-0.3$  T, evolving from an upward deviation to a drop [see Fig. 2(b)]. This implies that the contributions to the positive background (FM component and the paramagnetic signal of the Gd moments) diminish. A lower net magnetization at the neighboring CuO<sub>2</sub> planes leads either to a mixed state with a lower density of vortex lines, i.e., with an increased fraction of the superconducting volume, or, eventually, to prevent SVP formation. Then, the boundaries will be under the action of higher flux compression, as it is expulsed in a larger extent. Lower temperatures will be required to achieve intragrain percolation through a network of boundaries with an increased average local field. Also, the number of screened Gd paramagnetic ions in the domains will be increased. Thus, this picture of a magnetic transition leading to a state with a reduced Ru magnetization diminishes the positive components to the net magnetization, increases the superconducting fraction in the domains, and depletes the intragrain transition temperature.

In summary, we have provided evidence indicating that Ru-(1212) exhibits strong intragrain granularity effects. We propose that a phase-lock process occurs between structural domains of coherently rotated RuO<sub>6</sub> octahedra connected by antiphase boundaries, which act as Josephson-like junctions. SVP formation, flux expulsion from the domains and spin-flop transition of the Rusub-lattice are relevant points for this interpretation.

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