Ferromagnetism and superconductivity in pure and doped $RuSr_2GdCu_2O_8$

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$RuSr_2GdCu_2O_8$ (Ru-1212) is presently under particular interest, because of the coexistence of superconductivity (SC) ($T_c \approx 40$ K) and magnetism ($T_m \approx 140$ K) [1–4]. This system can be derived from the 123 high-T_c superconductors by replacing the CuO chains by RuO_2 layers. Ru-1212 is characterized by a sequence of CuO_2 double layers carrying the SC and RuO_2 layers responsible for the weak ferromagnetism (FM). The intrinsic nature of bulk SC and the uniform character of the magnetic interactions have been shown utilizing various techniques [2,5,6]. The magnetism in external zero field is dominated by the antiferromagnetic (AFM) order (G-type) of the Ru-sublattice in *c*-direction with a FM component ($< 0.1\mu_B$) lying in the *ab*-plane [8]. In fields below 10 kOe a purely FM state can be induced [9]. The Gd-moments order independently below 2.5 K, again revealing simple G-type AFM [8]. The electronic properties show widely the typical characteristics of strongly under-doped cuprates.

The coexistence of SC and weak FM motivated proposals of unconventional SC order parameters such as a Fulde–Ferrell–Larkin–Ovchinnikov phase [7]. In order to gain further insight into the interplay of magnetism and SC we synthesized a number of $Ru(Sr_{1-x}La_x)_2GdCu_2O_8$ samples. The single-phase polycrystalline samples were prepared using conventional ceramic techniques described elsewhere and were carefully characterized using X-ray diffraction [9]. The latter revealed the same primitive tetragonal structure (P4/mmm) of pure $RuSr_2GdCu_2O_8$ for all samples

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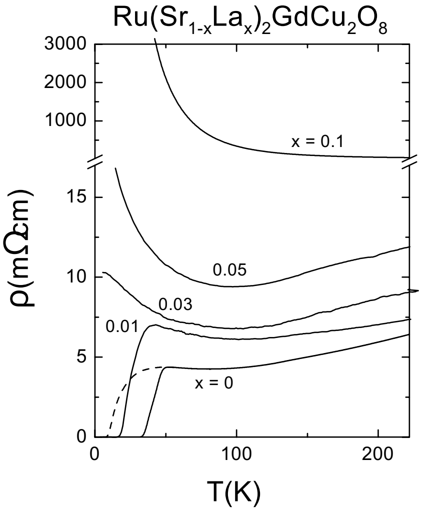

Fig. 1. Resistivity vs. temperature in zero external magnetic field. The dashed line represents the resistivity for $x = 0$ at $H = 100$ kOe.

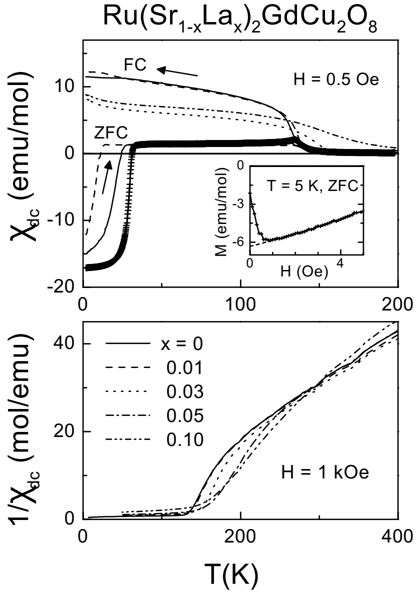

Fig. 2. Upper frame: DC magnetization vs. temperature for different La concentrations. For $x = 0$ and 0.01 the FC and ZFC branches are shown. The crossed line displays the real part of the AC-susceptibility χ'_{ac} measured with a stimulus of $H_{ac} = 0.1\ Oe_{rms}$ at a frequency of 1 kHz. The inset shows magnetization vs. field after zero-field-cooling for $x = 0$. Lower frame: Temperature dependence of the inverse susceptibility.

investigated ($x = 0, 0.01, 0.03, 0.05$ and 0.1). This crystallographic structure is closely related to that of other 1212-type cuprates. Planes of RuO_6 octahedra are connected via their apical oxygen ion with layers of CuO_5 square pyramids. With increasing La doping only weak shifts of the lattice parameters can be observed. In particular, the Cu–O–Ru bond angle ($\phi \approx 171^\circ$) remains unchanged up to La concentrations $x = 0.1$ [9]. Hence, one can expect that the magnetic exchange interaction and the charge transfer from Cu–O to Ru–O planes stays constant [10]. The substitution of La^{3+} on the Sr^{2+} site will be the main origin of the hole compensation in the Cu–O layers.

Fig. 1 displays the temperature dependence of the DC-resistivity at zero external field for the complete series of samples investigated. Focusing on the pure compound ($x = 0$), on decreasing temperatures the resistivity decreases, passes through a minimum close to 80 K and slightly increases just before the onset of SC at 50 K. The resistance drops from 90% to 10% of the initial onset value within 10 K and completely has vanished below 31 K. Probably due to the contribution of intrinsic magnetic interactions the transition appears to be quite broad even for ceramic samples. In an external field T_c is significantly shifted towards lower temperatures (dashed line, $H = 100$ kOe). On La doping the room temperature resistivity values are continuously increased. The sample with $x = 0.01$ behaves similar to the pure compound with the SC phase transition shifted to values which are approximately 10 K lower. For $x \geqslant 0.03$ the SC is fully suppressed. Again the resistivity passes through a minimum and reveals a semiconducting temperature characteristic below 100 K. Finally, for $x = 0.1$, $\rho(T)$ is strongly increased even at room temperature and increases for all temperatures below 300 K (see inset of Fig. 1). The complete suppression of SC by only 3% of La^{3+} substitution for Sr^{2+} and the onset of insulating behavior for $x \geqslant 0.1$ reveals that Ru-1212 is on the verge of a metal-to-insulator transition. The concentration of holes in the CuO_2 planes must be very low, because as mentioned above no change in the hole concentration due to structural reasons is to be expected.

The temperature dependence of the magnetic susceptibility $\chi_{DC} = M/H$ is displayed in Fig. 2. From the plot of the inverse susceptibility (lower frame) one can see, that the magnetic transition shifts slightly to higher temperatures with increasing La-concentration x while the effective magnetic moment remains nearly unchanged for $x < 0.1$. In this concentration range χ can be described assuming two independent Curie–Weiss contribution from the Gd ($J_{Gd} = \frac{7}{2}$) and the Ru ($S_{Ru} = \frac{1}{2}$, low spin) sublattices [9]. To emphasize the spontaneous FM contribution from Ru and to investigate the magnetic properties below the SC transition the DC magnetization was measured in a small external field of 0.5 Oe (upper frame of Fig. 2). Even though the onset of magnetic order is shifted to higher temperatures, the FC results reveal that the small ordered moment of the pure compound decreases on La doping. This points towards a more and more ideal AFM G-type structure. The Gd and Ru sublattices are still fully decoupled. From the ZFC measurements ($x = 0, 0.01$) we found evidence at least for a partial evolution of a Meissner state. The diamagnetic contribution of the FC magnetization curve can be estimated to amount 5% of the fully diamagnetic value displayed by the AC-susceptibility at low temperatures (crossed line in the upper frame of Fig. 2). The inset of Fig. 2 shows the field dependence of the ZFC magnetization at 5 K. Even for this low temperature smallest fields seem to enable the penetration of flux. Together with the incomplete Meissner-phase this points towards a vanishing lower critical field H_{c1} and matches previous results from the analysis of the frequency and field dependence of the AC-susceptibility [9]. The enhancement of the applied external fields by the intrinsic FM magnetization results in a spontaneous vortex phase. Nevertheless it should be emphasized, that the reduction of the FM component with increasing La doping coincides with the suppression of SC and the enhancement of resistivity in the normal conducting state.

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