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Magnetic and structural properties of the new double perovskite family $Sr_2GdRu_{1-x}Re_xO_y$

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We study the synthesis and structural and magnetic characterization of the new compound family $Sr_2GdRu_{1-x}Re_xO_y$, with doping levels at 3%, 6%, 9%, and 12%. X-ray diffractograms revealed that the samples crystallize in a monoclinic structure, and exhibit a strong distortion of the octahedron $Ru(Re)O_6/GdO_6$ lattice. Magnetic characterization revealed non-ideal antiferromagnetic behavior, with Néel temperatures close to 25 K, and an interesting metamagnetic feature below applied magnetic fields between 1.0 and 1.4 T. The existence of a Re doping level limit that favors an ideal antiferromagnetic character of the perovskite is discussed. [http://dx.doi.org/10.1063/1.4796045]

I. INTRODUCTION

The discovery of superconductivity in the ruthenocuprates $RuSr_2GdCu_2O_8$ (Ru1212) and $RuSr_2(R_{1+x}Ce_{1-x})Cu_2O_{10}$ (R = Sm, Eu, and Gd) by Bauernfeind *et al.* in 1995,[1] and the report, two years later, by Felner *et al.*[2] of the coexistence of superconductivity and magnetism in these compounds renewed the interest of both theoreticians and experimentalists, in the study of the interplay between superconductivity and magnetism.

The distinguishing characteristic of these compounds in comparison with other magnetic superconductors such as Chevrel phases[3] or rare earth ternary borides[4] is the fact that the magnetic transition temperature is much higher than the critical superconducting temperature, making them unique materials. However, these compounds are extremely sensitive to the synthesis process, making their study difficult. With the aim of reducing the synthesis-dependent characteristics,[5–7] a method for ruthenocuprate production involving the synthesis of double perovskites Sr_2LnRuO_6 (Ln = lanthanide) as precursor oxides was developed.

In accordance with these ideas, we proposed to produce the new Re-doped ruthenocuprate $Ru_{1-x}Re_xSr_2GdCu_2O_y$ with its associated double perovskite family, as described elsewhere.[8] Although the first objective of their production was to provide the precursor for the novel so-called rhenium-cuprates, these oxides exhibit interesting characteristics, calling for a very detailed study. In this paper, we present the results of the synthesis and the structural and magnetic characterization of this new compound family, with x = 0.00–0.12.

II. EXPERIMENTAL

$Sr_2GdRu_{1-x}Re_xO_y$ with x = 0.00, 0.03, 0.06, 0.09, and 0.12 was prepared using the solid state reaction method. Stoichiometric quantities of high purity Aldrich powders $SrCO_3$ (99%), RuO_2 (99.9%), ReO_2 (99.7%), and Gd_2O_3 (99.9%) were dried at 200 °C for 24 h, mixed and ground in an agate mortar, and then pressed into pellets of approximately 5 mm diameter and 1 mm thickness. The samples were calcined at 850 °C for 24 h (with intermediate grindings), then heated at 1170 °C for 48 h, and finally, heat treated at 1250 °C for 24 h, always with intermediate grinding. The crystal structure was studied through X-ray powder diffraction, using a PanAlytical Pro diffractometer with Cu-kα radiation (1.5406 Å) with PiXcel detection. The diffraction patterns were analyzed through Rietveld refinement using GSAS software. Magnetic measurements were carried out on a Quantum Design MPMS SQUID magnetometer.

III. RESULTS AND DISCUSSION

Rietveld refinement of X-ray diffraction patterns revealed that the samples are single phase (see Fig. 1 for results for the x = 0.03 sample). The reliability factors for this refinement were $\chi^2 = 0.984(6)$, $R_F = 9.8(1)\%$, and $R_{wp} = 8.8(3)\%$. It was determined that the samples form a non-ordered perovskite and crystallize in a monoclinical structure with $P2_1n$ (#14) spatial group.

A remarkable Ru/ReO_6 octahedron distortion, with tilt angles up to 19°, was observed in all samples. For Re concentration higher than 6%, the presence of unreacted Gd_2O_3, limited to 6% of the phase, was observed, indicating that the solubility limit was reached for x = 0.09.

In Fig. 2, we show the temperature dependence of the dc susceptibility for the x = 0.03 sample. It can be seen that the system exhibits a magnetic transition at approximately 25 K. A Curie-Weiss fit revealed an antiferromagnetic (AFM) coupling, with Weiss constant $\Theta < 0$. The same behavior was observed for the samples studied.

A decrease in $|\Theta|$ with increasing Re content can be observed. However, this does not seem to affect the magnetic

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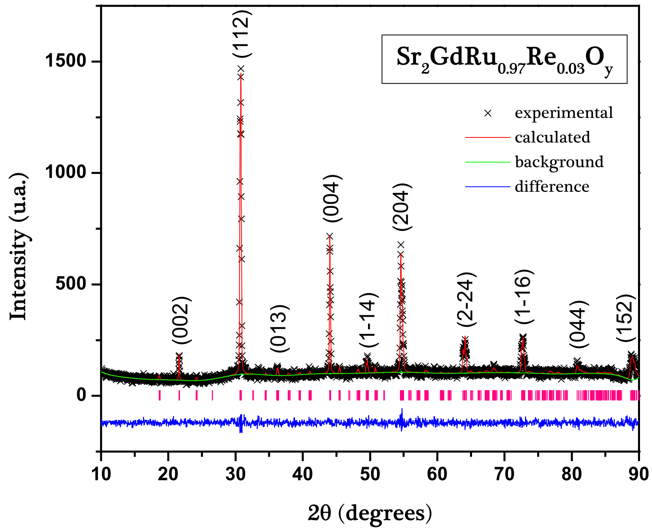

FIG. 1. Rietveld refinement of the X-ray diffraction pattern for the sample $Sr_2GdRu_{1-x}Re_xO_y$ with $x = 0.03$.

ordering temperature T_N (Fig. 3 and Table I), which has very similar values, except for the sample $x = 0.12$, which has a higher temperature, suggesting the existence of a doping level limit, after which the presence of Re starts to favor an ideal antiferromagnetic coupling.

The strong irreversibility in the ZFC (zero field cooled) and FC (field cooled) branches for the different Re contents (Fig. 4) suggests a non-ideal antiferromagnetic behavior, as confirmed by magnetization versus applied measurements (Fig. 5). The clear hysteretical cycle with no saturation reveals the presence of a weak ferromagnetic (FM) component.[9] The analysis of these measurements reinforces the doping limit hypothesis, since the ferromagnetic component diminishes as the Re level increases. In addition, it can be seen that the magnitude of the Weiss constant $|\Theta|$ decreases with the doping level in the sample, indicating a decrease in the exchange interaction.

The field dependence of the magnetization of all samples showed metamagnetic behavior, characterized by a quick superlinear increase in magnetization above a certain

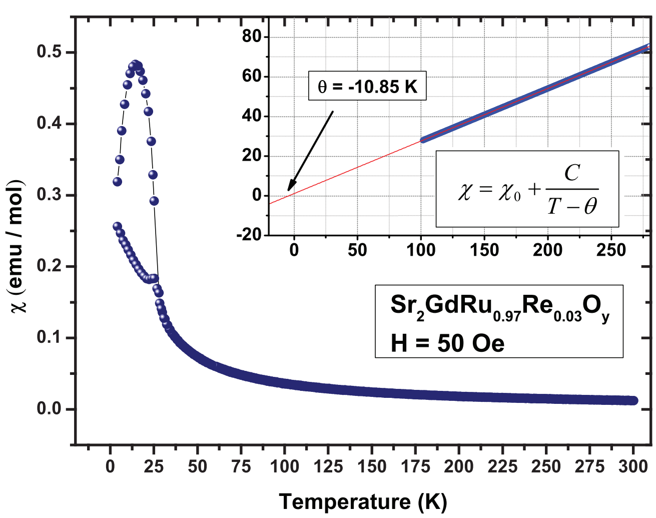

FIG. 2. Magnetic dc susceptibility for $x = 0.03$ sample, with applied field $H = 50\,Oe$. Inset: Curie-Weiss fit of paramagnetic region.

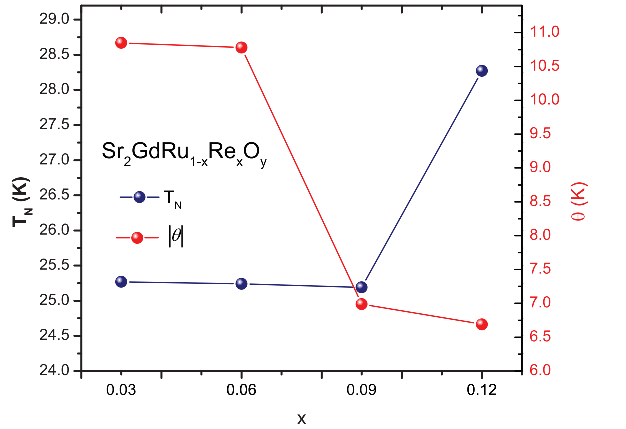

FIG. 3. Néel temperatures and Weiss constant magnitude as a function of Re content.

TABLE I. Néel temperatures and critical metamagnetic fields.

Sample	T_N (K)	H_{crit} (kOe)
$Sr_2GdRu_{0.97}Re_{0.03}O_y$	25.27	12.3
$Sr_2GdRu_{0.94}Re_{0.06}O_y$	25.24	10.2
$Sr_2GdRu_{0.91}Re_{0.09}O_y$	25.19	14.1
$Sr_2GdRu_{0.88}Re_{0.12}O_y$	28.27	9.9

critical field H_{crit}. This value decreases, increasing the Re doping level from 1.20 to 0.99 T. The metamagnetic behavior, evident from the sigmoidal shape in the hysteresis curve (Fig. 5), is defined as a transition from the AFM state, via

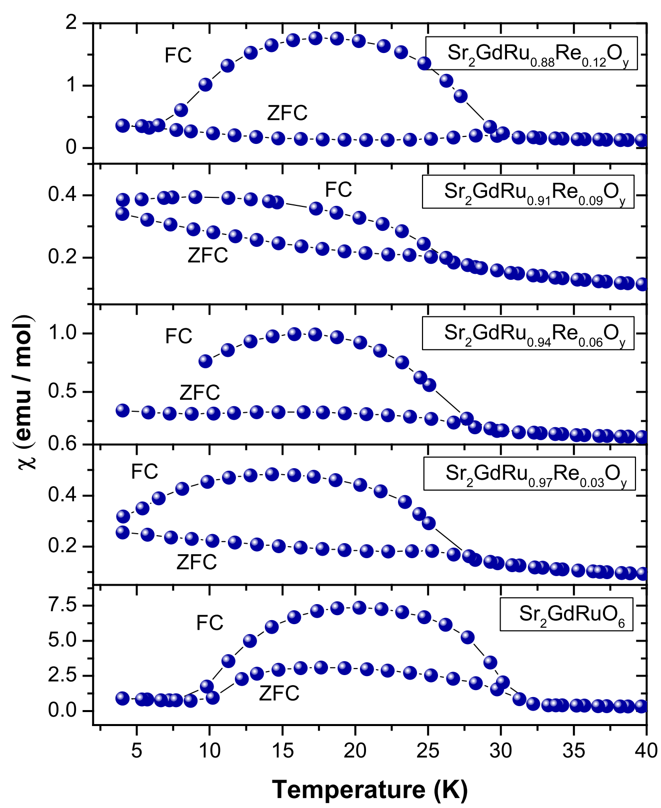

FIG. 4. Magnetic dc susceptibility as a function of temperature for $Sr_2GdRu_{1-x}Re_xO_y$.

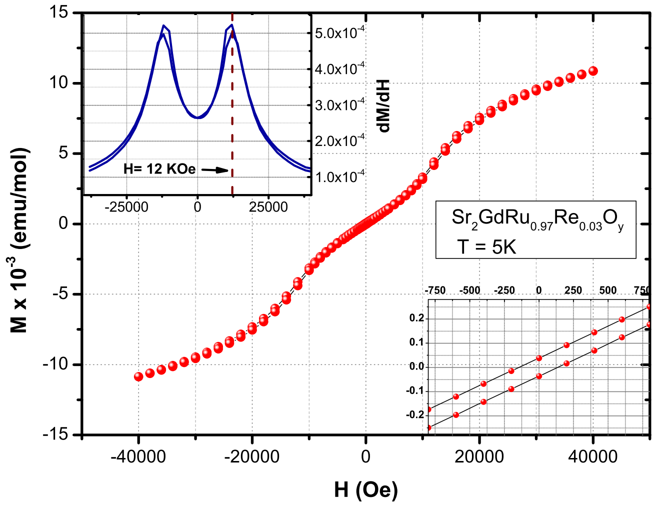

FIG. 5. Magnetization versus applied field for $Sr_2GdRu_{1-x}Re_xO_y$ with $x = 0.03$ at $T = 5$ K. Inset (left): First derivative, showing the metamagnetic critical field value. Inset (right): Magnification of low field region, showing the hysteretic characteristic.

phase transition or crossover, to a FM state, induced by the application of a magnetic field. As discussed above, these perovskites exhibit a tilt in the Ru-O-Ru angle, which favors the presence of a ferromagnetic component.[10] This, added to the fact that metamagnetism can occur in a near ferromagnetic metal, characterized by a maximum in magnetic susceptibility,[11] provides a scenario where the obtained results complement each other.

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