Spin Glass Behavior in the Ru-1222 and Ru-1212 rutheno-cuprate families: a comparative study.

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Abstract

Strong evidences of a spin glass phase on polycrystalline $\text{RuSr}_2\text{Ln}_{1.5}\text{Ce}_{0.5}\text{Cu}_2\text{O}_{10-\delta}$ (LnRu-1222, Ln = Y, Ho and Dy) is provided by ac susceptibility ($\chi_{ac}$) and dc magnetization measurements. The fingerprint of a spin glass transition, the frequency-dependent cusp in $\chi_{ac}$ vs. $T$ measurements at low magnetic field, is observed for all LnRu-1222 samples. The change in the cusp position with frequency follows the Vogel-Fulcher law, which is commonly accepted to describe a spin glass with magnetically interacting clusters. The strong suppression of the cusp in the ac susceptibility measurement in the presence of magnetic fields as low as 500 Oe, and the results of thermoremanent magnetization (TRM) and isothermal remanent magnetization (IRM) measurements
are also evidences of a spin glass behavior. By comparing these results with measurements on a YRu-1212 sample we conclude that the glassy behavior is characteristic of the Ru-1222 phase, not being observed for the studied YRu-1212 sample. We suggest that the spin glass transition occur only in the Ru-1222 phase possibly due to the larger number of oxygen vacancies present in these compounds.

75.50.Lk, 75.40.Gb, 75.60.Ej,
I. INTRODUCTION

A large number of reports have recently appeared focusing on the magnetic and super-conducting properties of the rutheno-cuprates $RSr_2Ln_{2-x}Ce_xCu_2O_{10-\delta}$ (LnRu-1222)\textsuperscript{1-7} and $RuSr_2LnCu_2O_{8-\delta}$ (LnRu-1212)\textsuperscript{8-20}. However, despite the intensive research on these materials, some unanswered questions still remain. For instance, the oxygen non-stoichiometry, carrier concentration and valence state of Ru are not well understood yet\textsuperscript{21,22}. One of the most controversial questions is the exact type of magnetic ordering in the LnRu-1222 family. In contrast to the LnRu-1212 family, for which a consensus has been reached on the canted antiferromagnetic ordering for the Ru sublattice\textsuperscript{14,20}, the detailed magnetic ordering of the LnRu-1222 family is still lacking. Although the magnetic behavior of LnRu-1222 has been considered to be analogous to the magnetic response for LnRu-1212 samples, some recent results point towards various differences between them\textsuperscript{3,23,24}. In particular, strong evidences of spin glass behavior was found in GdRu-1222\textsuperscript{24}. In the present work we extend this previous study to other LnRu-1222 compositions (Ln = Y, Dy, Ho). The verification of the spin glass behavior for these new samples and the differences between these results with those obtained for an YRu-1212 sample lead us to conclude that the spin-glass phase is characteristic of the Ru-1222 phase. We propose that the presence of oxygen vacancies in the RuO\textsubscript{6} octahedra for Ru-1222 may cause the frustration of the magnetic ordering of the Ru ions, leading to a glassy behavior in this compounds, which does not usually occur for Ru-1212 samples. Studies comparing the oxygen non-stoichiometry for both families support such interpretation\textsuperscript{21,22}.

II. EXPERIMENTAL DETAILS

Samples of composition $RuSr_2Ln_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ (LnRu-1222) with Ln = Y, Ho, and Dy were synthesized through a high-pressure high-temperature (HPHT) solid-state reaction route\textsuperscript{25}. Briefly, a mixture of $RuO_2 + SrO_2 + SrCuO_2 + 3/4CuO + 1/4CuO_{0.011} + 3/4Ln_2O_3 + \ldots$
1/2CeO$_2$ was sealed in a gold capsule and submitted to a pressure of 6 GPa in a flat-belt-type-high-pressure apparatus, where it was allowed to react for 2 hours at 1200 °C. It is expected that this procedure provides samples with oxygen content close to nominal\textsuperscript{25}, i.e. 10. All LnRu-1222 samples present a tetragonal structure within the $I4/mmm$ space group as confirmed by X-ray powder diffraction (XRD) patterns obtained at room temperature (Philips-PW1800; CuK$_\alpha$ radiation). The lattice parameters are $a = b = 3.824(1)$, $3.819(1)$, and $3.813(1)$ Å and $c = 28.445(1)$, 28.439(1), and 28.419(1) Å for Ln = Dy, Y and Ho, respectively. The YRu-1212 was prepared using the same procedure, except that for this compound it was necessary to start from a slightly Ru poor composition in order to obtain a single phase sample of the desired stoichiometry\textsuperscript{26,27}. The lattice parameter obtained are $a = b = 3.818(1)$ Å and $c = 11.5222(3)$ Å. The details of the precise HPHT process are given in Ref. [27]. All samples are single phase within the XRD resolution, except for YRu-1222 which presents a small amount of $SrRuO_3$\textsuperscript{25}. All ac susceptibility measurements were performed in a commercial PPMS (Physical Properties Measurement System), while for the dc measurements a SQUID magnetometer MPMS-5 were employed, both equipments made by Quantum Design company.

**III. EXPERIMENTAL RESULTS**

The ac susceptibility ($\chi_{ac} = \chi'$ $+$ $i\chi''$) technique is a very powerful method to provide evidence of a spin-glass behavior. In this case, both components $\chi'$ and $\chi''$ of $\chi_{ac}$ present a sharp, frequency dependent cusp. The position of the cusp in $\chi'$ defines the freezing temperature $T_f$, which is coincident with the temperature of the inflection point in $\chi''$. It is also well known that dc magnetic fields as low as a few hundreds of Oersted may suppress this cusp. In Fig. 1 we present the ac susceptibility for all four studied samples measured at a frequency $\nu = 1000$ Hz for three different dc fields $H = 50$, 150 and 500 Oe. All samples present a well defined peak at temperatures $T_f = 87.42$, 103.3 and 101.5 $K$ for Dy-, Ho- and YRu-1222, and $T_N = 140.6$ $K$ for YRu-1212. All three LnRu-1222 samples present a strong
suppression of the peak observed in χ′, which almost disappeared for H = 500 Oe. Besides this suppression, the peak position is shifted to higher temperatures at higher fields. On the other hand, the peak for YRu-1212 present a much weaker dependence with the applied field and it is slightly shifted to lower temperatures with the increase of H. Another important difference between LnRu-1222 samples and the YRu-1212 one is the presence of an anomaly in the susceptibility at $T_M = 150 K$ for LnRu-1222. Sample YRu-1212 does not present any anomaly in χ′ above $T_p$. This anomaly is more prominent for Y- and HoRu-1222 than for DyRu-1222, and is smeared out when the applied field is increased. It is striking that the anomaly observed in LnRu-1222 samples occurs at the same temperature, independently of Ln, and also that this temperature is close to the position of the peak observed for YRu-1212. These results are the first evidence that the peaks observed in χ′ have different origins for Ru-1222 and Ru-1212 families. In Fig. 2 we present the dc susceptibility as a function of temperature for different magnetic fields. The curves for all LnRu-1222 show a similar behavior. The field cooled (FC) measurements present a ferromagnetic-like shape, while the zero-field cooled (ZFC) curve present a well defined peak at temperatures, for $H = 50 Oe$, $T_p = 83.7, 101.8$ and 140.8 K for Dy-, Ho- and YRu-1222 respectively. The two branch apart twice, one at $T_M \approx 150 K$ (for all LnRu-1222 samples), and again at $T_{irr} \approx T_f \sim 100 K$ (Y- and HoRu-1222) or 84 K (DyRu-1222). At $H = 50 Oe$ the ZFC and FC curves are not exactly reversible for any temperature below $T_M$, so $T_{irr}$ is not well determined. As the applied field is increased to 500 Oe, a significant reduction in the irreversibility is observed and both, ZFC and FC curves, tend to a ferromagnetic-like behavior. The irreversibility observed at temperatures above the peak in the ZFC curve is also greatly affected by the increase of the applied field, being hardly distinguishable at $H = 500 Oe$. Comparing the results for dc magnetization with the ac susceptibility, we observe: (1) the anomaly in χ′ occurs at the same temperature of the deviation from the paramagnetic behavior in the magnetization data (at $T_M$); (2) the peak position in χ′ roughly coincides with $T_{irr}$. The coincidence of the peak temperature in χ′ with $T_{irr}$, the drastic reduction of the irreversibility and the ferromagnetic-like behavior of both ZFC and FC curves with quite small applied
fields, are all consistent with the expected behavior of a spin-glass system. Now we turn our attention to the YRu-1212 sample again. The ZFC/FC irreversibility is also suppressed with the increase of $H$, but a significant irreversibility remains present even at $H = 500 \text{ Oe}$. Also, the peak in the ZFC curve is broadened at higher $H$, but it do not disappear (up to 500 Oe). Once more, the behavior of the YRu-1212 is quite different from the observed for LnRu-1222, which in turn is very similar with each other.

The most clear and conclusive way to experimentally separate an antiferromagnet from a spin-glass is to probe the frequency dependence of the peak in $\chi'$. As can be observed in Fig. 3, the peak shifts to lower temperatures and its intensity increases as the frequency of the excitation field is decreased, for all three LnRu-1222 samples. This frequency dependence of $\chi'$ is a typical feature of the dynamics of spin-glass systems\textsuperscript{28}. On the other hand, no significant changes in the peak are observed for the YRu-1212 sample, which is the expected behavior expected for an antiferromagnet. A quantitative measure of the frequency shift is obtained from $\Delta T_f/[T_f \log(\omega)]$. This quantity varies in the range of 0.004 - 0.018 for spin-glass systems\textsuperscript{28}, while for superparamagnets\textsuperscript{28} it is of the order of 0.3. From the FC susceptibility measurements at different frequencies, presented in Fig. 3, we could estimate $\Delta T_f/[T_f \log(\omega)] \approx 0.0031, 0.0021$ and 0.0018 respectively for Dy-, Ho- and YRu-1222, which is slightly low but yet consistent with expected behavior for spin-glasses.

There are basically two different possible interpretations of the spin-glass freezing: the first one assumes the existence of a true equilibrium phase transition at a finite temperature (canonical spin glasses\textsuperscript{29}). The second interpretation assumes the existence of magnetic clusters and, in this case, the freezing is a nonequilibrium phenomenon\textsuperscript{30}. For isolated clusters (superparamagnets), the frequency dependence of their freezing temperature (in this context more correctly referred as blocking temperature) has been predicted to follow an Arrhenius law

$$\omega = \omega_0 \exp[-E_a/k_B T_f],$$  \hspace{1cm} (1)

where $E_a$ is the potential barrier which separates two easy orientations of the cluster and
\( \omega \) is the driving frequency of the \( \chi_{ac} \) measurement. However, for magnetically interacting clusters, a Vogel-Fulcher law has been proposed, which has the form:

\[
\omega = \omega_0 \exp\left[-\frac{E_a}{k_B(T_f - T_0)}\right],
\]

where \( T_0 \) can be viewed as a phenomenological parameter which describes the intercluster interactions. Equation 2 implies a linear dependence of the freezing temperature with \( \frac{1}{\ln[(\omega_\tau0)^{-1}]} \), \( \tau_0 = 1/\nu_0 = 2\pi/\omega_0 \). In Fig. 4 we present Vogel-Fulcher plots for the three LnRu-1222 samples, which shows that our data follows the expected linear behavior. From the best linear fit we obtained \( \nu_0 \approx 1 \times 10^{12} \) Hz, \( T_0 = 83.14, 99.83, 98.80 \) K and \( E_a = 90.25, 74.35, 58.38 \) K for Dy−, Ho− and YRu−1222, respectively.

Also, the existence of the spin-glass behavior has been checked through the measurement of its remanent magnetization. The remanent state can be achieved by two different procedures. The first one consists on cooling the sample in a certain magnetic field \( H \) from \( T >> T_f \) down to the measurement temperature. After the temperature is stabilized the applied field is removed and the measurement is performed. This procedure gives us the thermoremanent magnetization (TRM). In the spin-glass case, the TRM quickly reaches its saturation value for low values of \( H \). The second way to measure the remanence is to cool the sample in zero field down to the measurement temperature, and only after that a magnetic field \( H \) is turned on. After a short time the field is removed (the temperature remains constant) and the measurement is performed. This provides the isothermal remanent magnetization (IRM), which saturates at a higher \( H \) than the observed for the TRM. Both TRM and IRM coincide in the saturated region\(^{28} \). Fig. 5 shows both TRM and IRM measurements for the three LnRu-1222 and the YRu-1212 samples. For the TRM experiments, all curves present an abrupt increase at lower fields, going through a small maximum just before reaching its saturation, while the IRM experiments, all curves present a much more gentle increase up to the saturation value. These results are in agreement with the expected behavior of a spin-glass, as exposed above. The TRM and IRM results for the YRu-1212 sample present a less pronounced dependence on the applied field, although it also increases
with $H$.

IV. DISCUSSION

The results presented in the previous section strongly support that all LnRu-1222 studied samples present a spin-glass transition at $T_f$, while the YRu-1212 sample is an antiferromagnet with spin canting. Once the magnetic behavior of the studied samples has been established, we have now to compare the different results obtained for each sample. This is done in two steps. First we compare the results for the LnRu-1222 to check for possible influences of the specific Ln ion on the spin-glass properties of the sample. A second and more important discussion is to find out why Ru-1222 and Ru-1212 present so distinct magnetic properties. In order to start these discussions, it is useful to review the crystallographic structure of both systems.

The structure of $\text{RuSr}_2\text{LnCu}_2\text{O}_{8-\delta}$ (LnRu-1212) is derived from that of $\text{REBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (usually known as RE-123; in a more general categorization scheme$^{31}$, it is written as $\text{CuBa}_2\text{RECu}_2\text{O}_{7-\delta}$, $\text{Cu} – 1212$). To go from the former to the last structure, it is necessary to replace the Cu in the charge reservoir by Ru, thus transforming the CuO chains in RuO$_2$ planes, and the Ba ions are substituted by Sr. The structure of $\text{RuSr}_2(\text{Ln,Ce})_2\text{Cu}_2\text{O}_{10-\delta}$ (LnRu-1222), on the other hand, results from that of LnRu-1212 when a three-layer fluorite-type block $(\text{Ln,Ce})_2 \text{O}_2$ substitutes the oxygen-free Ln layer, between the two adjacent CuO$_2$ planes. It is believed that in rutheno-cuprates the RuO$_6$ octahedra in the charge reservoir are mainly responsible for both the magnetic ordering and the hole doping of the superconductive CuO$_2$ planes, while the Ln ions do not order magnetically. The paramagnetic contribution of magnetic Ln ions is usually associated to the increase of the magnetization observed at low temperatures in these systems. Then it is clear that the Ln layer plays a less important role in determining the magnetic ordering in these ruthenates. In fact, it could be inferred from the data presented in Figs. 1 - 5 that the change of the Ln ion did not change qualitatively the magnetic behavior of the LnRu-1222 samples. However,
a few differences between them can be pointed, the more obvious of them being the variation of the freezing temperature as one goes from Ho to Dy or Gd\textsuperscript{24}. As shown in Fig. 6, this is related to the changes of the volume of the unit cell, due to the different ionic radii of the Ln ions. In the same figure is also plotted $T_M$ which is almost constant for the samples studied in this work but, for the GdRu-1222 sample previously studied, the value of $T_M$ is much higher\textsuperscript{24}. Remembering that the GdRu-1222 sample was prepared by a different technique\textsuperscript{32}, it is more likely that this difference is a consequence of the synthesis process than an intrinsic effect of the different Ln ions. Also, it is observed that the irreversibility observed in ZFC/FC magnetization curves at temperatures $T_f < T < T_M$ is quite small for GdRu-1222 and becomes more prominent as smaller Ln ions are considered. Both, different ionic radii and synthesis process, seem to be important in the determination of this irreversibility.

A more intriguing question to be answered is why all Ru-1222 samples we have studied present a clear spin-glass behavior while the YRu-1212 sample is an antiferromagnet. This is intriguing because the differences between both structures are restricted just to the Ln plane, which is isolated from the RuO\textsubscript{2} planes which order magnetically, as already discussed. Therefore, it seems probable that the Ln plane would affect the magnetic behavior of the compound by indirectly affecting the RuO\textsubscript{2} planes. In order to produce a spin-glass state it is necessary to have a frustration of the magnetic order, which is accomplished by disorder. One could first try to find this randomness in the mixing of Ce and Ln, which is only present in the Ru-1222 family. A random distribution of Ln could cause small local perturbations in the crystalline structure in such a way that the RuO\textsubscript{6} could be distorted, thus inducing the frustration of Ru magnetic moments. A more likely and simple explanation would come from oxygen non-stoichiometry, which could directly affect the RuO\textsubscript{2} planes. The different magnetic behavior of the two families (Ru-1212 and Ru-1222) could only be explained if the Ru-1222 system are more susceptible to present oxygen vacancies than the Ru-1212 family. In fact, a work recently reported\textsuperscript{22} concludes that GdRu-1212 can be obtained with a near stoichiometric oxygen content, while the GdRu-1222 phase is clearly oxygen-deficient.
even after 100-atm $O_2$ annealing. Changes observed in the valence of Ru in GdRu-1222 as the oxygen content varies suggest that the vacancies occur in the RuO$_2$ layers. The same work also shown that the oxygen content in GdRu-1212 is almost constant upon various annealings, while for GdRu-1222 a wider range in oxygen content is accessible. These results lead us to conclude that the most reasonable origin for the glassy behavior observed in our LnRu-1222 samples is the presence of oxygen vacancies. If this is true, our results show in a quite dramatic way the influence of the exact oxygen content of the sample. It is likely that some of the contradictory results reported on Ru-1222 in the literature are consequences of the difficulty to fully oxygenate samples of the Ru-1222 system. Although we do not observed any indication of a spin glass phase in the studied YRu-1212, we believe that degraded, oxygen-depleted Ru-1212 may also present a glassy behavior. It is important to notice that the samples we have studied in this work (as well as the GdRu-1222 sample, reported in Ref. [24]) were synthesized by routes which allow a higher oxygen content than more usual procedures.

V. CONCLUDING REMARKS

In summary, in this work we show that the frequency-dependent peak observed in the temperature dependence of the ac susceptibility $\chi_{ac}$, its suppression by a small magnetic field, combined with the remanent magnetization results, provide strong evidence of the existence of a spin glass phase over a significant temperature range in polycrystalline LnRu-1222. This is valid for samples with different $Ln$ and prepared by two different routes (considering also the GdRu-1222 sample, previously reported$^{24}$), indicating that the glassy behavior is a characteristic feature of the Ru-1222 family. This is to be contrasted with the existence of long-range antiferromagnetic order with spin canting for the YRu-1212 sample. We believe that oxygen vacancies could frustrate the long-range order of the Ru spins, leading to a spin glass phase. Recent results pointing that the Ru-1222 phase is usually oxygen deficient, while the Ru-1212 family can be synthesized with near stoichiometric oxygen content, corroborate
this idea and provide a possible explanation for the different behavior we found for these
two families.

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FIG. 1. Real part of the ac susceptibility as a function of temperature for different magnetic fields, for all four samples. Insets show the anomaly observed (in LnRu-1222 samples only) at $T_f < T < T_M$.

FIG. 2. dc magnetization as a function of temperature for different magnetic fields, for all four samples. Insets show the irreversibility observed (in LnRu-1222 samples only) at $T_f < T < T_M$.

FIG. 3. Real part of the ac susceptibility as a function of temperature for $H = 50$ Oe and four different frequencies, for all four samples. The peak position defines the freezing temperature $T_f$ (for the LnRu−1222 samples).

FIG. 4. Variation of the freezing temperature $T_f$ with the frequency of the ac field in a Vogel-Fulcher plot, for the three LnRu-1222 samples. The solid line is the best fit of Eq. 2.

FIG. 5. Thermoremanent magnetization (TRM, solid symbols) and isothermal remanent magnetization (IRM, open symbols) for all samples, measured as a function of the applied
field, at $T = 30$ K.

FIG. 6. Variation of the freezing temperature $T_f$ and of the magnetic transition $T_M$ as a function of the volume of the unit cells for LnRu-1222 compounds. The results for GdRu-1222 were extracted from Ref. [24].
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