PHYSICAL REVIEW B **71**, 134509 (2005)

# Magnetic and superconducting properties of $RuSr_2Gd_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ samples: Dependence on the oxygen content and aging effects

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(Received 31 July 2004; revised manuscript received 24 November 2004; published 15 April 2005)

The magnetic and superconducting properties of $RuSr_2Gd_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ polycrystalline samples with different oxygen-doping levels are presented. A strong suppression of the superconducting temperature ($T_c$), as well as a reduction in the superconducting fraction, occurs as the oxygen content is reduced by annealing the samples in oxygen-deprived atmospheres. Drastic changes in the electrical resistivity are observed above $T_c$, possibly associated with oxygen removal, mainly from grain boundaries. However, the magnetic ordering is relatively less affected by the changes in oxygen content of the samples. The spin-glass transition is enhanced and shifted to higher temperatures with the reduction in oxygen content. This could be correlated with an increase in the spin disorder and frustration for the oxygen-depleted samples. Also, the same oxygen-vacancy-induced disorder could explain the reduction in the fraction of the sample showing antiferromagnetic order. We also report significant changes in the measured properties of the samples as a function of time.

DOI: 10.1103/PhysRevB.71.134509 PACS number(s): 74.25.Ha, 74.72.Jt, 75.50.Lk, 75.40.Gb

## I. INTRODUCTION

The rutheno-cuprate $RuSr_2Gd_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ (Ru-1222) and $RuSr_2GdCu_2O_{8-\delta}$ (Ru-1212) families have attracted considerable interest by the coexistence of magnetic order and superconductivity in these compounds.[1,2] The magnetic ordering of the Ru moments occurs at a temperature around 100 K, while the superconducting transition temperature ($T_c$) is usually no higher than 50 K. The fact that superconductivity occurs in the $CuO_2$ planes, while the magnetic order is related with the Ru ions, casts some doubt about the genuine coexistence of these two phenomena at a microscopic level. Also, the exact nature of the magnetic order is still in debate. In this context, the role played by the oxygen nonstoichiometry in the determination of the magnetic and superconducting properties of these compounds is still not completely clear.[3–5] In particular, the large number of oxygen vacancies observed in Ru-1222, bigger than in Ru-1212,[4] was associated with the presence of a spin-glass (SG) behavior observed in Ru-1222 samples.[5,6] Although reports on oxygen doping in Ru-1222 can be found in the literature,[7–10] the possible correlation of oxygen stoichiometry and the appearance of the SG phase has not been explored yet. In fact, this has been one of the motivations for the present work. We have studied polycrystalline samples of Ru-1222 with different oxygen-doping levels by performing magnetization, ac susceptibility, and resistivity measurements. For oxygen-depleted samples we observed a large variation in the resistivity and a suppression of the superconducting transition, while the magnetic order, above $T_c$, showed a subtle qualitative modification. A significant variation in the measured properties of the samples was also observed as a function of time, which may help to explain some contradictory results reported in the literature.

## II. EXPERIMENTAL DETAILS

Samples of composition $RuSr_2Gd_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$ were synthesized through a solid-state reaction route. High purity $RuO_2, SrCO_3, Gd_2O_3, CeO_2$, and CuO were mixed in stoichiometric proportions and calcinated at 1000, 1020, 1040, and 1050 °C each for 24 h with intermediate grindings. One pellet was annealed in a flow of high-pressure oxygen (100 atm) at 420 °C for 100 h and labeled as $O_2$ annealed. Two other pellets were annealed in air for 48 h at 1050 °C (labeled as air-annealed). One of them was further annealed in nitrogen at 420 °C for 24 h ($N_2$ annealed). In this way it was possible to vary the oxygen content in the studied samples. Though the actual oxygen content of the samples was not determined, qualitatively the same should be the maximum for the 100 atm $O_2$-annealed samples, moderate for the air-annealed, samples and minimum for the $N_2$-annealed samples.

X-ray diffraction (XRD) patterns were obtained at room temperature for all three samples (MAC Science: MXP18VAH; $F^{22}$Cu K$\alpha$ radiation). The ac susceptibility measurements were performed in a commercial PPMS (physical properties measurement system), while for the dc measurements a superconducting quantum interference device (SQUID) magnetometer MPMS-5 was employed, both pieces of equipment made by Quantum Design company. For the resistivity measurements the four-point technique was used.

 

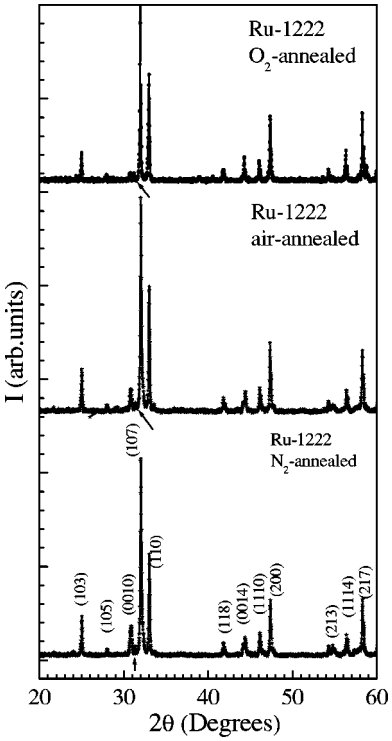


FIG. 1. X-ray diffraction patterns for all three samples studied.

### III. RESULTS AND DISCUSSION

Analysis of the XRD patterns (Fig. 1) revealed that all three samples crystallize in a tetragonal structure (space group $I4/mmm$) with the following lattice parameters for $O_2$-, air-, and $N_2$-annealed samples, respectively, $a=b=3.8327(7)$ Å and $c=28.3926(8)$ Å, $a=b=3.8427(7)$ Å and $c=28.4126(8)$ Å, and $a=b=3.8498(3)$ Å and $c=28.4926(9)$ Å. The increase in the lattice parameters from the $O_2$- to the $N_2$-annealed samples indicates that the different annealings are not only affecting the grain boundaries but leading to an overall reduction in the oxygen content in the bulk of the air- and $N_2$-annealed samples. Small impurity peaks were observed in the XRD patterns, which are the same for all samples. Despite the presence of these impurity peaks, our samples may be considered to be of very good quality if compared to those reported in the literature by several authors.[11]

The temperature dependence of the magnetization for all three samples is presented in Fig. 2. The samples were cooled in a zero magnetic field down to the lowest accessible temperature (2 K). After temperature stabilization, a magnetic field of 50 Oe was applied and the magnetization recorded as the temperature was raised [zero field-cooled curve (ZFC) ] up to 200 K. Then the temperature was decreased back to 2 K, keeping the same applied magnetic field, and the field-cooled (FC) curve was obtained. The ZFC curves show pronounced peaks at 68 K, 84 K, and 92 K for $O_2$-, Air-, and $N_2$-annealed samples, respectively, while the FC curves show monotonical increases with the reduction of the temperature. Besides the shift to higher temperatures of the

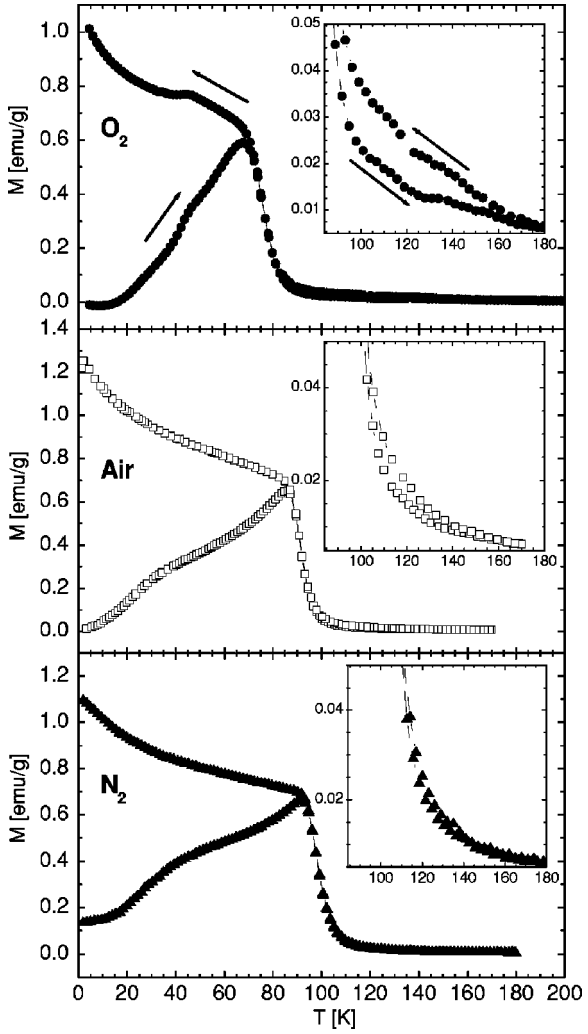


FIG. 2. Magnetization as a function of temperature and $H=50$ Oe, for all three samples. Insets show the suppression of the irreversibility observed near the onset of the magnetic transition, with the decrease in oxygen content.

peak in the ZFC curve, its height also increased with the reduction in the oxygen content of the samples, going from 0.58 emu/g for the $O_2$-annealed sample to 0.67 emu/g for $N_2$-annealed one. In the SG scenario, the frustration of the antiferromagnetic interaction is believed to be associated with the presence of disorder due to the large number of oxygen vacancies in this compound.[4,6] Thus, an increase in the number of oxygen vacancies would lead to a more disordered system, favoring the occurrence of the SG phase at higher temperatures. This interpretation is consistent with the results presented in Fig. 2. A small antiferromagnetic (AFM) transition can be detected at higher temperatures, around 174 K for all samples, although this transition becomes less pronounced with the reduction in oxygen content. At temperatures between the AFM transition (~174 K) and the SG freezing point (~80 K) a small irreversibility can be observed. As shown in the insets of Fig. 2, the irreversibility is greatly reduced for the air-annealed sample and it is almost completely absent in the $N_2$-annealed sample. These results point to the simultaneous occurrence of an ordered phase

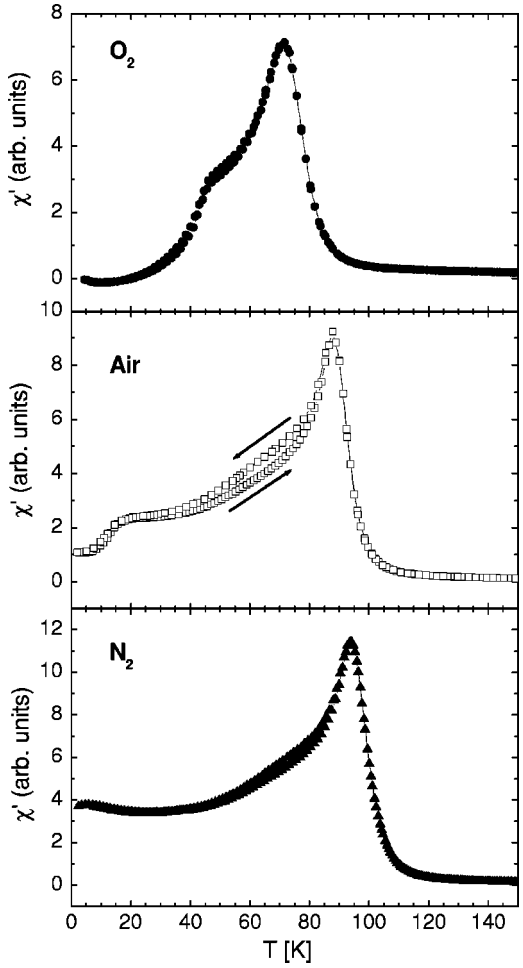


FIG. 3. Real part of the ac magnetic susceptibility as a function of temperature at an applied dc field of $H=50$ Oe, for all three samples. The amplitude and frequency of the excitation field were 1 Oe and 1000 Hz, respectively.

(AFM) and a disordered or frustrated phase,[12] whose configurations freeze into a SG at $T_f \sim 80$ K. It is likely that as oxygen vacancies are introduced in the sample, a larger fraction of the same becomes disordered, favoring the SG phase at the expense of the AFM phase. In fact, the almost reversible magnetization curve observed for the $N_2$-annealed sample, above the SG freezing temperature $T_f$, exhibits the expected behavior of a prevailing SG phase.[13]

Figure 3 presents the temperature dependence of the real part of the complex ac susceptibility $\chi_{ac}=\chi'+i\chi''$, measured with the same routines employed to obtain the magnetization curves. The measurements were performed at an applied field of $H=50$ Oe and revealed well defined peaks at temperatures $T_f=70.7$ K, 87.9 K, and 93.7 K for $O_2$-, air-, and $N_2$-annealed samples, respectively. The position of these peaks defines the freezing temperature of the spin system, where the ZFC and FC curves branch apart from each other in the magnetization measurements (see Fig. 2). For the ac susceptibility, the ZFC and FC curves are almost identical for all three samples, except for temperatures in the 40–90 K range. For these temperatures a small irreversibility is observed for all samples, being more pronounced for the air-annealed sample and less pronounced for the $O_2$-annealed sample. A clear feature present in these curves is a sudden decrease in $\chi'$, associated with the superconducting transition. We see that the superconducting phase is strongly suppressed by the reduction of the oxygen content in the samples. Both the transition temperature and the superconducting fraction are reduced until the complete suppression for the $N_2$-annealed samples, which does not present superconductivity down to 2 K.

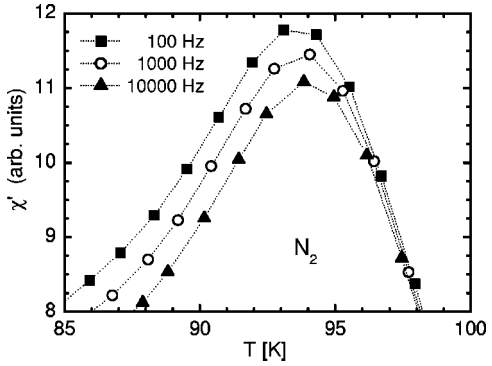


FIG. 4. Real part of the ac susceptibility as a function of temperature for $H=50$ Oe and frequencies of 100, 1000, and 10 000 Hz for the $N_2$-annealed sample.

The frequency dependence of the peak in $\chi'$ as a function of the temperature is a clear signature of a SG phase. To check this we repeated the $\chi' \times T$ measurements for three different frequencies and for all samples. For the sake of brevity, we show in Fig. 4 only the results for the $N_2$-annealed sample. Similar results were found also for the other samples. As observed in Fig. 4, the peak shifts to lower temperatures and its intensity increases as the frequency of the excitation field is decreased, which is the expected behavior for a SG system.[5,6,14] The results obtained through magnetization and ac susceptibility measurements show unequivocally the presence of a SG phase in all three samples studied, with the SG phase being more noticeable in the oxygen-depleted samples. On the other hand, a reduction in the oxygen content suppresses both the superconducting phase (below $T_f$) and the fraction of the sample that presents AFM order (above $T_f$).

The imaginary component of the complex susceptibility also presents a peak associated with the glassy transition, as can be observed in Fig. 5. This peak occurs at about 3 K below the corresponding peak in $\chi'$ and is weakly frequency dependent, as expected for a SG phase.[13] However, a second peak appears at 42.9 K, 57.7 K, and 65.7 K for the $O_2$-, air-, and $N_2$-annealed samples, respectively, as shown in Fig. 5 for a frequency of the driving field of 1000 Hz. This peak is strongly frequency dependent, being more intense and shifting to higher temperatures as the frequency is increased, as shown for the $N_2$-annealed sample in Fig. 6. It is important to notice that the strong frequency dependence of this second peak contrasts with the very weak frequency dependence observed for the SG peak, clearly indicating that it has a different origin. It shows a more intense signal in the FC branch, if compared with the ZFC branch, thus revealing a strong irreversibility. Also, the temperature range where this

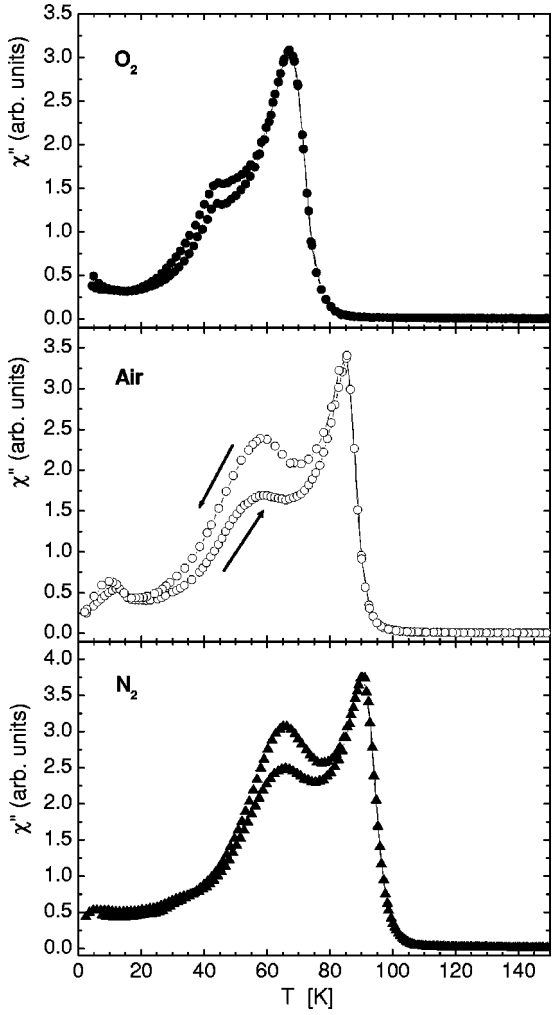


FIG. 5. Imaginary part of the ac magnetic susceptibility as a function of temperature at an applied dc field of $H=50$ Oe, for all three samples. The amplitude and frequency of the excitation field were 1 Oe and 1000 Hz, respectively.

peak and the irreversibility appear in the $\chi''$ measurements coincides with the irreversibility observed in $\chi'$ (see Fig. 3). For the $O_2$ sample this peak could tentatively be associated with the superconducting transition, which occurs at a close temperature. However, the same is not true for the other two samples. For the air-annealed sample, for instance, the peak related to the superconducting transition is located at about 47 K below the second peak position. A more likely explanation for the irreversibility and the second peak in $\chi''$ could be associated with the rotation of spins or spin-clusters, due to the excitation field applied during the ac susceptibility measurements. A recent report on samples of the Ru-1222 family shows[15] that for temperatures between 30 and 80 K, depending on the exact sample composition, the hysteresis loops are very narrow, with the coercivity field ($H_c$) being zero at a certain temperature within this interval. This coincides very well with the temperature range where we found an irreversible behavior in our $\chi''$ measurements. If $H_c$ is small enough, then the amplitude of our ac magnetic field (1 Oe) could rotate the existing magnetic domains. Since this spin rotation is a dissipative process, one should expect an

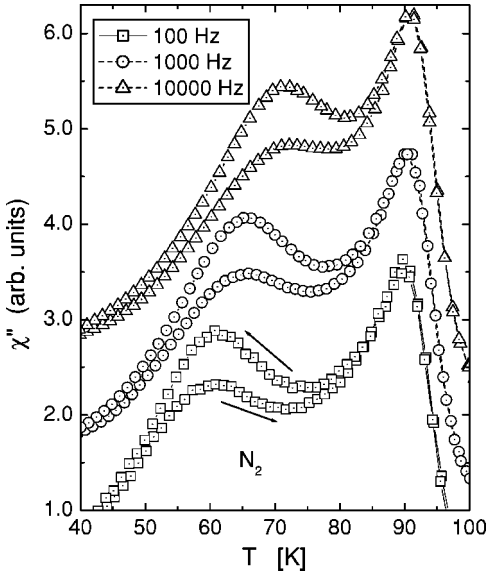


FIG. 6. Imaginary part of the ac susceptibility as a function of temperature for $H=50$ Oe and frequencies of 100, 1000, and 10 000 Hz, for the $N_2$-annealed sample. The data for 1000 Hz and 10 000 Hz were, respectively, shifted up by 1.0 and 2.0, in units of the arbitrary scale, for a better visualization.

increase in the dissipation (and then in $\chi''$) which would peak when $H_c=0$. Also, it was shown that the coercive field is larger for samples annealed in high pressure of oxygen, as compared with as-grown samples.[10] Therefore it can be inferred that this peak in $\chi''$ should be stronger for oxygen-depleted samples, in full agreement with our data (see Fig. 5). Although no clear change at this temperature can be detected in the magnetization curves (Fig. 1) for any of the three samples, it is possible to argue that this peak indicates a rearrangement of the spins in the sample, going from a more disordered state (SG phase) to a more ordered weak-ferromagnetic (W-FM) state. Our results alone are insufficient to clearly identify such reordering of the spins; however, recent results on zero-field muon spin rotation (ZF-μSR) have shown two different internal fields at low temperatures, one vanishing at temperatures above 90 K and the second one vanishing at 80 K.[16]

Another important property strongly affected by the change in the oxygen content of the samples is their resistivity, as shown in Fig. 7. By reducing the oxygen content of the sample both the resistivity and its variation with temperature increase [see Fig. 7(a)]. All samples present an exponential growth of resistivity with decreasing temperature. At low temperatures the behavior changes; for the $O_2$- and air-annealed samples, the superconducting transition can be undoubtedly identified, while for the $N_2$-annealed samples the resistivity continues to increase at an even higher rate. A different result was obtained when we repeated these measurements after a few months. A clear increase in the resistivity was observed, indicating a possible continuous loss of oxygen from the samples, which were kept in a dessecator all the time. After 5 months [see Fig. 7(b)], the resistivity measurements for the air-annealed sample (open squares) are similar to the result previously obtained for the $N_2$-annealed

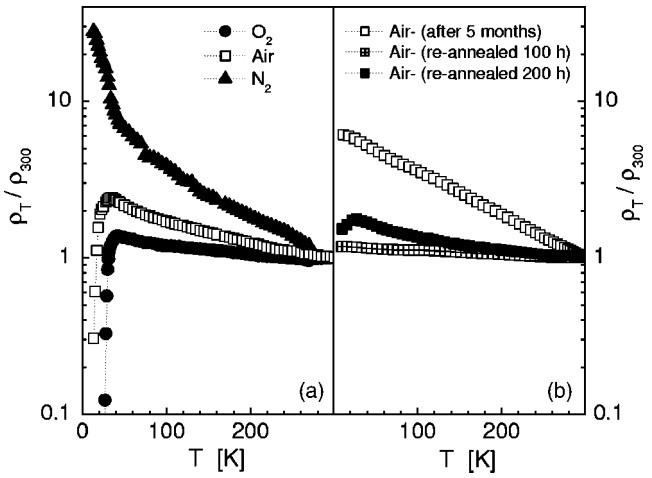


FIG. 7. (a) Normalized resistivity as a function of temperature, for all three samples. (b) Results for the air-annealed sample after degradation and reannealing in an $O_2$ atmosphere (see text).

sample, showing no sign of superconductivity. To evaluate if this sample degradation could be due to oxygen loss only, we tried to recover the original state of the air-annealed sample by annealing it in an oxygen atmosphere. Our attempts to reoxygenate the sample in a 1 atm oxygen atmosphere for 100 h at 450 °C and 100 h at 800 °C did not produce any meaningful change in its properties. Then it was annealed again for 100 h at 420 °C in 70 atm of $O_2$. The resistivity curve obtained after oxygenation, shown in Fig. 7(b) as crossed squares, presented a drastic reduction in its slope, becoming similar to the curve for the $O_2$-annealed sample. Nevertheless, the superconductivity could not be restored at all. After a second annealing, performed under the same conditions as the previous one, the superconducting transition could then be partially recovered in this sample [Fig 7(b), solid squares]. The obtained $T_c \approx 25$ K indicates that even after both annealings the sample still remains underdoped. Interestingly, the normal-state resistivity for the sample after both annealing processes shows a steeper change in resistivity with the decrease in temperature. An $M \times T$ curve for $H = 50$ Oe (ZFC followed by FC) was also measured for the reoxygenated sample as shown in Fig. 8. We can see that the basic features previously observed for the oxygen annealed sample are all present, including the irreversibility observed at temperatures immediately above the onset of the SG peak and the AFM transition at $T_c \approx 175$ K (see the inset of Fig. 8). Both resistivity and magnetic measurements indicate that the oxygen content in the sample increased with the annealings, but they were not enough to fully reoxygenate the sample. The fact that the resistivity is strongly affected by the degradation of the sample and by its reoxygenation, while the changes in magnetization and susceptibility are more subtle, seems to indicate that both processes affect primarily the grain boundaries. However, the complete suppression of the superconductivity, not detected even by inductive measurements, and the change in the AFM transition for the degraded samples point to the relevance of processes occurring in the bulk of the grains.

Our results indicate that although the oxygen can easily leave the sample, the reverse process of reoxygenation is

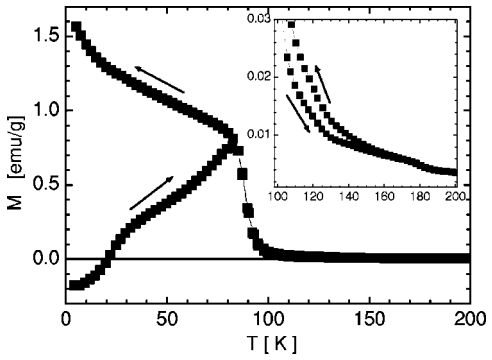


FIG. 8. Magnetization as a function of temperature and $H = 50$ Oe, for the air-annealed sample after degradation and reannealing. The inset shows the irreversibility near the onset of the magnetic transition.

quite tricky and difficult. Also, the oxygen content of Ru-1222 samples can change with time, and that aging process could be the origin for many discrepancies between results reported in the literature. For instance, the valence of the ruthenium ions is known to be affected by the change in oxygen content.[3] Our results indicate that not only the synthesis process may be responsible for the different values of Ru valence reported in the literature, due to the different initial oxygen content of the samples. The time delay between the samples' preparation and their measurements could also strongly affect the results.

## IV. DISCUSSION

The magnetic properties of Ru-1222 are very complex, with different magnetic transitions and possible phase separations. The significant role played by oxygen vacancies is still under study and would be benefit from a careful comparison of works from different groups. It is important to compare results obtained by different techniques in the attempt to reach a better understanding on the interesting properties of this rutheno-cuprate family. Recently, Felner *et al.*[15] presented a detailed Mössbauer study of Ru-1222 where they observed two magnetic transitions. The first was an AFM transition at 160 K, and the second was identified as the onset of a W-FM order at around 90 K. Interestingly, only 10–20 % of the sample volume ordered antiferromagnetically while its major fraction presents a magnetic order only below 90 K. These results gave strong support to the idea of magnetic phase separation in Ru-1222 polycrystalline samples.[17] Further, the Mössbauer spectra was rather broad and more than one subspectra, besides the one associated with the small AFM fraction, was necessary to fit the experimental results, indicating some degree of disorder or inhomogeneity in the sample. Both central conclusions presented in that work, the occurrence of phase separation, and the presence of disorder and inhomogeneity are consistent with our results. Similar conclusions were reached by Shengelaya *et al.*[16] based on muon spin rotation ($\mu$SR) experiments in $RuSr_2Eu_{1.4}Ce_{0.6}Cu_2O_{10}$ samples. They observed that a fraction (around 15%) of the sample orders magnetically at temperatures around 200 K, while the remaining fraction of the

sample orders only at temperatures below 90 K, in close connection with the strong increase in magnetization. Then, a well resolved internal field appears at a lower temperature close to 77 K. Another important result is that two different oscillating components were observed. This may indicate either two different muon stopping sites within the same magnetic phase or two different magnetic phases, with two different but close temperature transitions. Comparing these results with ours, we again find support for the idea of phase separation, with a possible occurrence of AFM clusters at temperatures between 90 and 200 K. The majority phase orders at a temperature of 77.6 K, which is close to the freezing temperature observed in our $O_2$-annealed sample. Our results have demonstrated a strong sensitivity of the magnetic properties of Ru-1222 to the sample synthesis process and oxygen content, which changes dramatically from sample to sample and, for the same sample, with time. However, some common ground has been achieved. It seems clear that two different magnetic phases are present in Ru-1222: one AFM phase, that shows a transition temperature around 160–200 K, and another phase that appears at temperatures around 70–90 K. The AFM phase presents spin canting that leads to a small ferromagnetic loop, and it is usually a minority phase corresponding to 10–20 % of the sample volume. Our results indicate that this fraction decreases with the reduction of the oxygen content. The second magnetic transition occurs at $\sim$90 K when the rest of the sample starts to show some magnetic order. The frequency dependence observed in our ac susceptibility data strongly suggests that a SG phase sets in at this temperature, leading to an additional ferromagnetic component. As the oxygen content is reduced and the system becomes more disordered (or more inhomogeneous) this SG phase is favored and both its volume fraction and freezing temperature increase. The dissipative peak observed at temperatures around 60 K (see Fig. 5), in the imaginary component of the ac susceptibility, seems to be related to a coercive field being close to zero in this temperature range.[10,15] This fact may indicate some reordering of the spins, from the SG phase into a more ordered WFM state. However, no significant change was observed in the magnetization curves, so if such reordering really occurs it would be quite subtle.

## V. FINAL REMARKS

In this work we have explored the influence of the oxygen content in the electrical and magnetic properties of polycrystalline samples of $RuSr_2Gd_{1.5}Ce_{0.5}Cu_2O_{10-\delta}$. It was observed that the reduction in the oxygen content favors the development of a spin-glass state at the expense of the antiferromagnetic phase. The presence of oxygen vacancies in the system may affect the magnetic ordering of the Ru sublattice in two ways: by changing the valence of some of the Ru ions and by distorting the crystalline lattice. Both mechanisms introduce disorder in the system and affect the antiferromagnetic interaction, favoring the appearance of a spin-glass phase. By reducing the charge density in the system, the oxygen depletion also strongly influences the resistivity and the occurrence of superconductivity in Ru-1222, which disappears completely for the $N_2$-annealed sample.

We also documented the deterioration of the samples in a time window of a few months and its recovery after annealing in a high-pressure atmosphere of oxygen. In a polycrystalline sample it is expected that oxygen first leaves the grain boundaries, thus strongly affecting the resistivity of the sample. Further reduction in the oxygen content occurs at the $RuO_2$ planes,[4] affecting strongly the magnetic order and leading to the spin-glass phase. In a later stage the oxygen from the $CuO_2$ planes possibly starts to leave the sample gradually suppressing the superconductivity. Similarly, in the reoxygenation process the grain boundaries are easily oxygenated, leading to a quick recovery in the resistivity of the sample. However, the oxygen diffusion inside the grains is difficult and slow, being effective only when the sample is annealed in high-oxygen pressure. Since this process was incomplete in our samples the spin-glass phase persisted even after a high oxygen filling. By further increasing the oxygen content it starts to get into the $CuO_2$ planes and then superconductivity is recovered. It seems that some rearrangement of the oxygen atoms inside the sample may take place at this point, which could eventually explain why the recovery of superconductivity occurs simultaneously with an increase in the normal-state resistivity as observed in this work. In conclusion, these results corroborate previous discussions on the importance of the oxygen-doping level in the electrical transport and magnetic properties of Ru-1222 samples.[5,6] Additionally, we have found that aging effects could be a possible explanation for some of the contradictory results found in the literature for this rutheno-cuprate family.

## ACKNOWLEDGMENTS

This work was partially supported by Brazilian agencies FAPESP and CNPq. Authors from NPL, India thank Professor Vikram Kumar Director for his keen interest in the present work.

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