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Spin-glass behavior, spin fluctuations, and superconductivity in $Sr_2Y(Ru_{1-u}Cu_u)O_6$

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Muon spin rotation measurements of $Sr_2Y(Ru_{1-u}Cu_u)O_6$ (for $u=0.1$) reveal two distinct muon sites: one located in a SrO layer (which is superconducting at low temperatures) and the other in a $Y(Ru_{1-u}Cu_u)O_4$ layer (which is magnetically ordered at low temperatures). A precursor spin-glass state due to the Ru moments is detected in high fields (≈3.3 kOe) in $Y(Ru_{1-u}Cu_u)O_4$ layers, with a spin-glass temperature of $T_G = 29.25$ K. The $Y(Ru_{1-u}Cu_u)O_4$ layers order ferromagnetically in the *a-b* planes at the Néel temperature, $T_N \approx 23$ K. This in-plane ferromagnetism alternates direction between adjacent $Y(Ru_{1-u}Cu_u)O_4$ planes, resulting in a net antiferromagnetic structure. Although the onset of superconductivity is observed both by electron spin resonance and by dc susceptibility to occur for temperatures up to about $T_{c,\mathrm{onset}} \approx 49$ K, this superconductivity is adversely affected by the Ru moments that fluctuate for $T > T_N$ producing magnetic fields that break pairs in the SrO layers. The muons, as well as other probes, sense the more-robust static superconductivity for $T < T_G$. In fact, resistance measurements only show zero resistance below T_N, at which temperatures the Ru moments that fluctuated for $T > T_N$ are frozen in-plane. Hence strictly speaking, the superconducting transition temperature is the same as T_N, which is far below $T_{c,\mathrm{onset}}$. Below T_N there are no pair breaking fluctuating magnetic fields in the SrO layers where the hole condensate resides.

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I. INTRODUCTION

$Sr_2Y(Ru_{1-u}Cu_u)O_6$, with $u \leqslant 0.15$, is an interesting compound [Fig. 1 (Ref. 1)] because it (i) has no cuprate planes,[2] (ii) superconducts at an onset temperature of $T_c \approx 45-49$ K,[3–9] (iii) has only two types of layers, $(SrO)_2$ and $Y(Ru_{1-u}Cu_u)O_4$, (iv) superconducts in its SrO layers,[5] (v) exhibits ferromagnetism in the *a-b* planes of its $Y(Ru_{1-u}Cu_u)O_4$ layers, whose ferromagnetic moments alternate direction from one adjacent magnetic $Y(Ru_{1-u}Cu_u)O_4$ layer to the next, forming a net antiferromagnetic structure,[10] (vi) has Cu ions that spin order at ≈86 K,[9] and (vii) exhibits spin-glass behavior of its $Y(Ru_{1-u}Cu_u)O_4$ layers in a narrow range around 29.25 K (as we shall show here).

In previous studies it was found that the muons stop at two types of sites, $\mu_{O(1,2)}$ and $\mu_{O(3)}$, the first of which is actually two nearly identical sites approximately at the center of four oxygen ions in a $Y(Ru_{1-u}Cu_u)O_4$ layer [we treat these two O(1,2) sites as equivalent]. The second muon stopping site is $\mu_{O(3)}$, and is on the edge of a SrO layer and between two oxygen ions in that layer, with two more oxygen ions above and below it in $Y(Ru_{1-u}Cu_u)O_4$ layers (see Fig. 1). At low temperatures the $Y(Ru_{1-u}Cu_u)O_4$ layers are

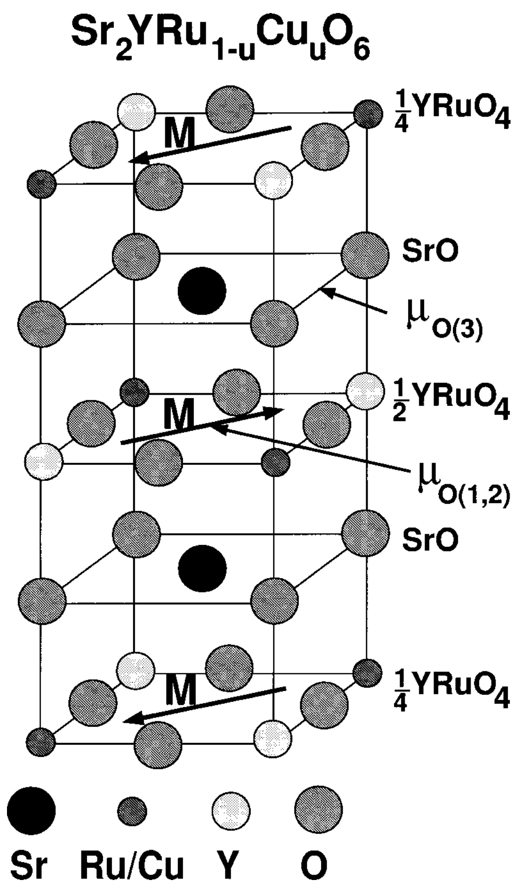

FIG. 1. The crystal structure of $Sr_2Y(Ru_{1-u}Cu_u)O_6$ is shown, along with the probable muon sites, $\mu_{O(1,2)}$ and $\mu_{O(3)}$, according to Ref. 1. The arrows **M** represent the average magnetic polarization of the Ru moments at temperatures below ≈23 K.

ferromagnetic sheets,[5] but are stacked in the c direction antiferromagnetically. Consequently for temperatures less than 23 K, the magnetic field (due to local moments) at a $\mu_{O(3)}$ site is zero, while the field at a $\mu_{O(1,2)}$ site is ≈3 kG.[8]

II. MEASUREMENTS

Our pressed-powder samples of $Sr_2Y(Ru_{1-u}Cu_u)O_6$ were polycrystalline and were prepared using a standard solid-state reaction.[4] They were characterized using an energy-dispersive x-ray analyzer, by high-resolution x-ray diffraction, and by neutron powder diffractometry.[9] These studies indicated that the sample material was phase pure to <1%. Measurements were made employing muon spin rotation (μSR), magnetic susceptibility, and resistance. All experiments were conducted using the same sample material and at an applied magnetic field of ~3.3 kOe. Both the dc susceptibility and the resistance were also measured at a smaller field of 10 Oe for comparison.

Since neutron powder diffractometry measurements[9] also indicate that the Ru spins order ferromagnetically in the planes at 23 K, the remainder of this paper will examine what is happening between 23 and 50 K. This is the most interesting region for us to study, because it contains interesting data: a spin-glass state.

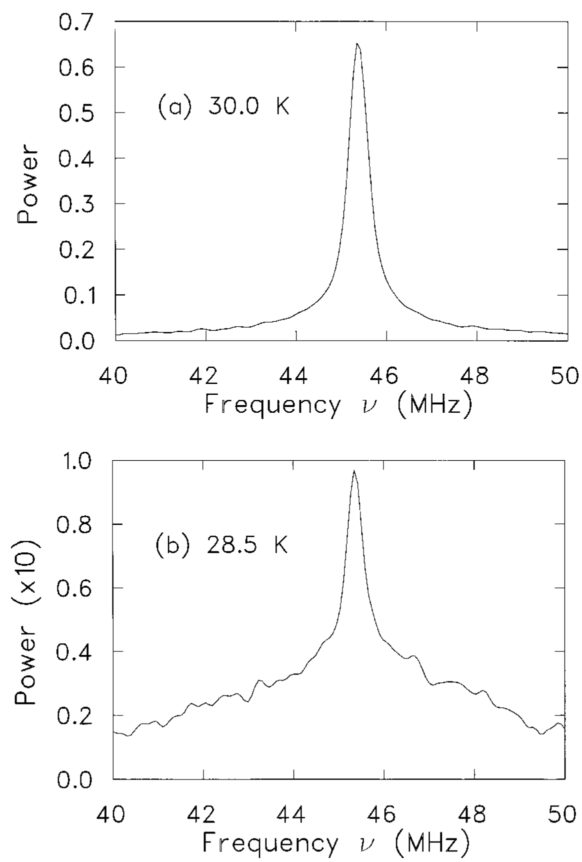

FIG. 2. Fourier power spectra [obtained by transforming the time spectra $H(t)$] versus frequency ν, are shown for $Sr_2Y(Ru_{0.9}Cu_{0.1})O_6$ at (a) 30.0 K and (b) 28.5 K. Above $T_G = 29.25$ K, as shown in frame (a), only a narrow peak is observed. However, below T_G [see frame (b)], the spin fluctuations begin to slow, introducing an additional (much broader) peak (corresponding to the fast relaxing signal which characterizes $\mu_{O(1,2)}$). The time-domain data $H(t)$ were smoothly truncated by multiplication prior to transformation; the multiplying function was $\exp(-\sigma^2 t^2)$, where $\sigma = 0.5\ \mu s^{-1}$.

A. Muon spin rotation

The μSR experiments were performed at the TRIUMF cyclotron facility using the standard time-differential techniques.[11,12] A low-background detection apparatus was employed, which vetoed events from muons that missed the sample, thereby making it possible to also accurately extract small minority components of the signal. For these experiments, the material was pressed into a pellet having a diameter of about 2 cm and a thickness of about 2 mm.

The time-domain muon data $H(t)$ were acquired in a 3.34 kOe transverse magnetic field as a function of temperature. Fourier power spectra of these data, shown in Fig. 2, feature a single narrow peak at 30.0 K [Fig. 2(a)] which splits below ~30 K [Fig. 2(b)] into a narrow peak (corresponding to muons stopped at the $\mu_{O(3)}$ sites) on top of a very broad peak (reflecting the fast relaxing signal associated with muons stopped at the $\mu_{O(1,2)}$ sites). The time spectra $H(t)$ were fit to a power-law relaxation function of the form

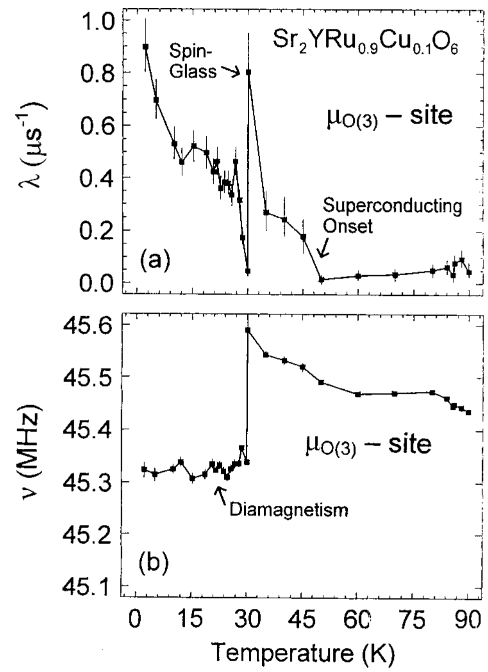

FIG. 3. The (a) muon relaxation rate $\lambda_{O(3)}$ and (b) precession frequency $\nu_{O(3)}$ in $Sr_2YRu_{0.9}Cu_{0.1}O_6$ vs temperature T taken in a transverse magnetic field of 3.34 kOe. The errors indicated are statistical, corresponding to one standard deviation.

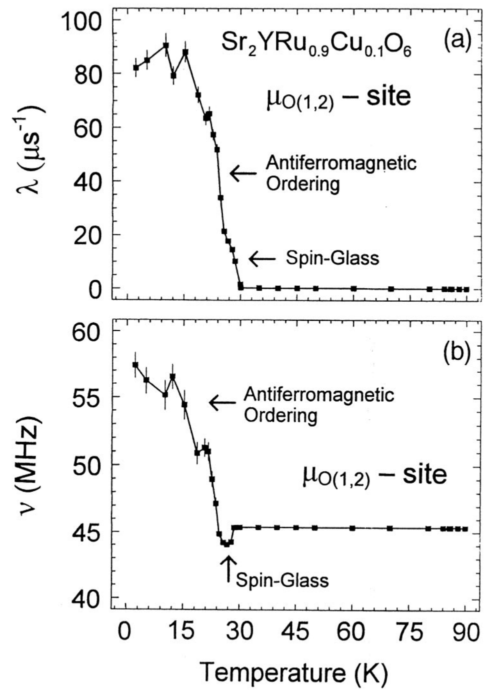

FIG. 4. The (a) muon relaxation rate $\lambda_{O(1,2)}$ and (b) precession frequency $\nu_{O(1,2)}$ in $Sr_2YRu_{0.9}Cu_{0.1}O_6$ vs temperature T, taken in a transverse magnetic field of 3.34 kOe. The errors indicated are statistical, corresponding to one standard deviation.

$$G_{xx}(t) = \sum_i A_i \exp[-(\lambda_i t)^{p_i}]\cos(2\pi\nu_i t + \phi),$$

where ϕ is the phase, and for each i, A is the amplitude, λ is the relaxation rate, ν is the frequency, p is the power exponent, and i refers to the site, either $\mu_{O(3)}$ or $\mu_{O(1,2)}$. For the signal associated with the $\mu_{O(3)}$ site in the SrO layer, p was approximately unity, and so was fixed to unity (corresponding to an exponential relaxation rate). But for the $\mu_{O(1,2)}$-site signal from the $Y(Ru_{1-u}Cu_u)O_4$ layer, the power p obviously varied, and was allowed to vary within the range from $p=0.5$ to $p=2.0$, being fixed at $p=2$ when the fitted power was indistinguishable from 2.

Figures 3(a) and (b) show the relaxation rate $\lambda(T)$ and the spin precession frequency $\nu(T)$ for muons stopped at the $\mu_{O(3)}$ sites. From the earlier muon and neutron results,[9] we know that the in-plane magnetic polarization [of adjacent $Y(Ru_{1-u}Cu_u)O_4$ layers] alternates direction, thereby resulting in a net magnetic field due to local moments; this field is zero in the SrO layers (at the $\mu_{O(3)}$ sites). The $\mu_{O(3)}$-site data for λ exhibit an initial rise (as temperature decreases) below 50 K, presumably associated with superconductivity, and show an increasing relaxation rate λ as a function of decreasing temperature below 29 K. Moreover, the muons stopped at $\mu_{O(1,2)}$ sites in the $Y(Ru_{1-u}Cu_u)O_4$ layers sense the Ru ordering transition at 23 K in both λ and ν, but the muons stopped at $\mu_{O(3)}$ sites in SrO layers do not. This, coupled with the fact that the muons at the $\mu_{O(3)}$ sites sense strong relaxation (presumably from superconductivity) below ~30 K (in λ), suggests that the Ru moments may already be confined to the *a-b* planes for temperatures below ~30 K.

The low-temperature diamagnetic shift observed in $\nu(T)$ for the $\mu_{O(3)}$ sites [Fig. 3(b)], is about 200 kHz, which corresponds to a field of about −15 G. This 0.5% shift in field is largely due to the magnetization of the sample induced by the 3.34 kOe applied field. Data taken earlier in 500 Oe (Ref. 8) showed a comparable 0.4% diamagnetic shift. These same data also exhibited a hysteresis in λ upon cooling in zero field, normally consistent with the presence of vortices. The local magnetic field shift expected, $-8\pi M/3$, for an antiferromagnet or spin glass having the geometry of our sample, with the applied field perpendicular to its flat side, can be estimated to be about −10 G if we assume the Ru moments are about 1 Bohr magneton and their fields inside a local-field sphere cancel at the site of the muon. This field could likely explain all of the −15 G shift observed for muons at this site. Thus the shift in $\Delta\nu(T)$ arising from the formation of vortices is very small in comparison.

Figures 4(a) and (b) present the relaxation rate $\lambda(T)$ and the precession frequency $\nu(T)$ for the $\mu_{O(1,2)}$ sites. Above 30 K, the $\mu_{O(1,2)}$ site data show no depolarization due to motional narrowing from the Ru moments (which are rapidly fluctuating). However, as the temperature is decreased, a slight rise in $\lambda(T)$ is observed from 30 K down to 23 K, followed by a much sharper rise below 23 K. Interestingly, as temperature decreases from ≈30 K, $\nu(T)$ remains relatively constant until 29.25 K, where it exhibits a large diamagnetic dip [labeled "Spin-Glass" in Fig. 4(b)], followed by a sharp rise at 23 K (coinciding with Ru ordering). We define $T_G = 29.25$ K to be the spin-glass temperature.

The detailed ordering of the $Y(Ru_{1-u}Cu_u)O_4$ layer is best illustrated by Fig. 5, which shows the power exponent p vs temperature T for the relaxation function employed to fit the $\mu_{O(1,2)}$-site signal produced by the muons near the face cen-

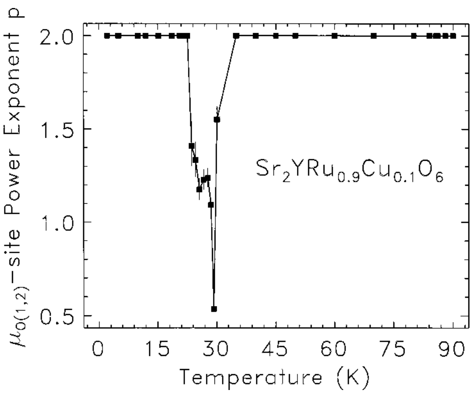

FIG. 5. The power exponent p at the $\mu_{O(1,2)}$ site as a function of temperature T in $Sr_2YRu_{0.9}Cu_{0.1}O_6$ taken in a transverse field of 3.34 kOe. The errors shown are statistical, corresponding to one standard deviation.

ter of the $Y(Ru_{1-u}Cu_u)O_4$ plane. For most of the temperature range, the power exponent for this plane is $p=2$, corresponding to a Gaussian relaxation envelope. However, near 30 K, p descends dramatically from $p=2$ to $p=0.5$, the exponent characteristic of a dilute spin-glass state in the fast-fluctuation limit.[13]

Upon closer examination, there appears to be a very narrow temperature range near T_G where the transverse field data show evidence for an $\exp[-(\lambda t)^{0.5}]$ decay (see Fig. 5). Such a time dependence has often been seen in spin-glass materials[13] and is associated with a range of magnetic environments, some with faster than average and others with slower than average depolarization rates. In transverse fields, the internal-field distribution of a dilute spin glass is expected to be reflected by an exponential decay (i.e., $p=1$) for static fields and by root-t exponential decay, $\exp[-(\lambda t)^{0.5}]$, if the spins fluctuate, assuming a simple time-correlation function with an Edwards-Anderson order parameter.[13,14]

Thus it may be that as the Ru spins of the $Y(Ru_{1-u}Cu_u)O_4$ layers slow down when temperature is reduced from $\approx$30 to $\approx$23 K, their effect on λ for the muons is similar to that of a dilute spin glass: Once the temperature is reduced several degrees below the narrow temperature region for which spin-glass-like behavior is evident, the relaxation is appropriate to a Gaussian decay of the $\mu_{O(1,2)}$ signal with a large depolarization rate. This is evidence for a single Gaussian distribution of fields and in fact is consistent with our zero-field measurements that show precession of the muon spin due to the local in-plane fields.

B. dc magnetization

The dc-magnetization data were acquired using a Quantum Design superconducting quantum interference device magnetometer on an elongated sample of 75.9 mg in a parallel field (i.e., the field parallel to the long dimension). Data taken upon zero-field cooling in 10 Oe are shown in Fig. 6(a). Upon close examination, these data reveal a slight diamagnetic response below $T_{c,\mathrm{onset}} \approx 49$ K, followed by a much sharper diamagnetic response below about 29.25 K. This confirms our earlier contention that the fluctuating Ru moments tend to interfere with superconductivity until they begin to freeze out at about 30 K (Ref. 8). The small bump observed at $T_N=23$ K corresponds to the spin ordering of the Ru moments.[9] For temperatures $T_N<T<T_G$, the Ru spins are most likely confined to the *a-b* plane, but fluctuate.

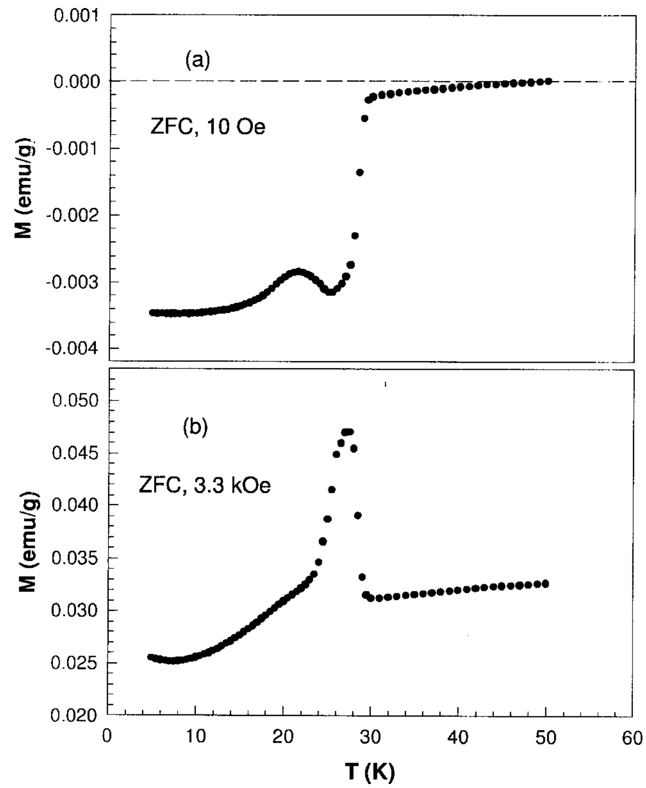

FIG. 6. The dc magnetization versus temperature for $Sr_2YRu_{0.9}Cu_{0.1}O_6$ taken upon zero-field cooling at (a) 10 Oe and (b) 3.3 kOe.

Figure 6(b) shows the dc magnetization versus temperature curve taken upon zero-field cooling at 3.3 kOe. As is clear from the data, a prominent peak in the paramagnetism is observed just below 30 K, thereby confirming the μSR spin-glass response. Interestingly, the bump at 23 K is reduced compared with the low-field data of Fig. 6(a) indicating that the applied field of 3.3 kOe affects the spin ordering somewhat. Moreover, the spin-glass effect observed just below 30 K is also field dependent since it is absent from the low-field data of Fig. 6(a), as well as from earlier low-field (500-Oe) μSR data.[8]

C. Resistance

The resistance is shown in Fig. 7 for the applied fields of 10 Oe and 3.3 kOe. Notice that zero resistance is only achieved at about $T_c \approx 23$ K, which is also the temperature T_N at which all of the Ru moments stop fluctuating and become ordered. This coincidence of T_N and T_c can be understood by realizing that as the material cools, the Ru moments fluctuate less and less, until all of the Ru moments become ordered for temperatures at and below $T_N=23$ K.

These data again support our contentions that fluctuating moments (i) act to destroy superconductivity and (ii) provide

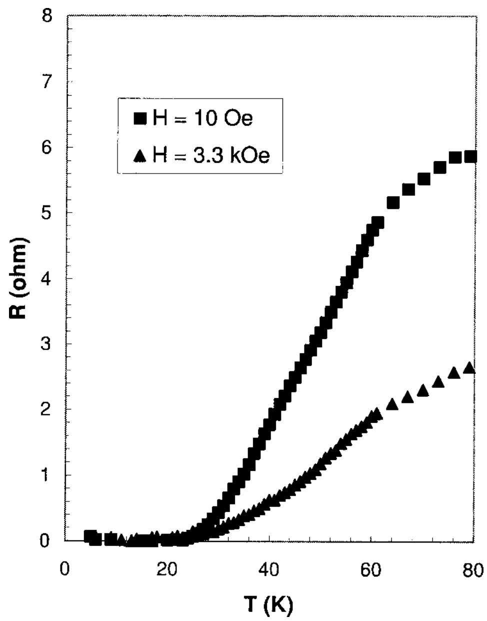

FIG. 7. The resistance of $Sr_2YRu_{0.9}Cu_{0.1}O_6$ taken after field cooling as a function of temperature T in an applied field of 10 Oe (squares) and 3.3 kOe (triangles).

a natural explanation of why $T_{c,\text{onset}}$ is $\approx$49 K, but T_c itself is only $\approx$23 K and coincides with T_N.

D. Fluxons

Since no hysteresis was observed in λ at the $\mu_{O(3)}$ site upon cooling in a field, after which the field was turned off and on, the data indicate weak pinning (because λ was unchanged). Therefore the data are consistent with a set of isolated sheets of pancake vortices, as in $Bi_2Sr_2CaCu_2O_8$,[15–20] which would be the case if the superconducting hole condensate resided in the SrO layers, with the vortex c-axis correlation length reduced by the intervening magnetic layers.

Although the electron spin resonance sees evidence for fluxons above 30 K, those fluxons may be short lived (GHz frequencies) compared with the time scales of μSR experiments (megahertz frequencies), which only see the longer-lived fluxons.

III. CONCLUSION

To summarize, muon spin rotation (μSR), dc magnetization, and resistance measurements of $Sr_2Y(Ru_{1-u}Cu_u)O_6$ (for $u=0.1$) have been performed. Analyses of the μSR spectra indicate the existence of two distinct muon sites: one signal corresponding to two magnetically similar sites near the center of the $Y(Ru_{1-u}Cu_u)O_4$ layers, and the other (weaker) feature due to muons located in the SrO layers. Just below 30 K, at a temperature of 29.25 K, a unique magnetic state associated with the Ru moments of $Y(Ru_{1-u}Cu_u)O_4$ is observed in high applied magnetic fields where the power p of the exponential tends toward 0.5, indicative of a varying distribution of exponential relaxation functions consistent with a dilute spin-glass state (in the fast-fluctuating limit). This state exists only over a very narrow temperature range (perhaps only over a few degrees Kelvin) and is corroborated by the high-field dc magnetization shown in Fig. 6(b). Below 29.25 K, the fluctuation rate of the Ru spins continues to slow, until at 23 K a first-order ferromagnetic transition is observed in these layers.[9] The polarization direction of the local ferromagnetic order alternates between magnetic layers, resulting in a net antiferromagnetic system. This unique stacked antiferromagnetic structure results in zero net magnetic fields in the SrO layers. At 10 Oe, the dc magnetization shows a slight diamagnetism starting at about 49 K before dropping dramatically below 30 K. From this we set the superconducting onset to be approximately 49 K. Zero resistance is not achieved until all of the Ru moments have stopped fluctuating and order at $T_N \approx 23$ K. Hence the true superconducting transition temperature is actually 23 K when the Ru moments are frozen, although the onset of superconductivity occurs at 49 K, which would be the critical temperature for superconductivity if the Ru moments were frozen instead of fluctuating. Above this temperature, fluctuating superconducting currents may be present in the sample, but it is clear that the fluctuating Ru moments act to suppress sustained superconductivity up to 49 K.

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