## Separation between antiferromagnetic and ferromagnetic transitions in $Ru_{1-x}Cu_xSr_2EuCu_2O_{8+\delta}$

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The macroscopic magnetizations of  $\operatorname{Ru}_{1-x}\operatorname{Cu}_x\operatorname{Sr}_2\operatorname{EuCu}_2\operatorname{O}_{8+\delta}$  with *x* between 0 and 0.15 were investigated. A ferromagnetlike transition as well as an antiferromagnetlike transition appear around  $T_M$  in the low-field magnetization and around  $T_{AM}$  in the high-field differential susceptibility, respectively. The separation between them, which is accompanied by a flat plateau in the magnetic  $C_p$ , increases with *x*. Superparamagnetic M(H) and slow spin dynamics, i.e., characteristics of nanomagnetic clusters, were observed far above  $T_M$ . A comparison with  $\operatorname{Ru}\operatorname{Sr}_2(\operatorname{Eu}_{1-y}\operatorname{Ce}_y)\operatorname{Cu}_2\operatorname{O}_{10+\delta}$  and some manganites further suggests that a phase separation occurs, which can describe well the conflicting magnetic-superconductivity data previously reported.

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The puzzling bulk, yet granular, superconductivity (SC) in  $RuSr_2RCu_2O_{8+\delta}$  (Ru1212R) ruthenocuprates and  $\operatorname{RuSr}_2(R,\operatorname{Ce})_2\operatorname{Cu}_2\operatorname{O}_{10+\delta}(\operatorname{Ru}1222R)$  with  $R = \operatorname{Gd}$ , Eu, or  $Y^{1-4}$ , which coexists with weak ferromagnetism, is closely related to their magnetic structure. While a homogeneous canted antiferromagnetic (CAFM) spin order may coexist with more or less ordinary superconductivity, such as the proposed Meissner state or the  $\pi$ -phase SC,<sup>2,5</sup> magnetic inhomogeneity at length scales  $\geq \xi$  will unavoidably lead to a Josephson-junction-array-like superconductivity,<sup>4</sup> where  $\xi$  is the coherence length. In the case of Ru1222R, the reported data seem to indicate a rather complicated magnetic structure. Both the antiferromagnetic (AFM) like differentialsusceptibility maximum of the Ru ( $\chi_{Ru only}$ ) and the hyperfine splitting of the Mössbauer spectra, for example, occur at temperatures almost two times higher than  $T_M$ , where a ferromagnetic (FM) like transition occurs in the low-field fieldcooled magnetization  $(M_{FC})$ .<sup>1,6</sup> Either a phase separation or a multistage transition, therefore, should occur.<sup>1,6</sup> On the other hand, the situation of Ru1212R has been suggested to be different. The inflection point  $T_{AM}$  at  $\partial^2 (T \chi_{Ru only}) / \partial T^2$ =0, which should be the Néel temperature in simple antiferromagnets,<sup>7</sup> and  $T_M$  are in rough agreement for a Ru1212Eu sample.<sup>8</sup> Mean-field-like scaling has also been observed below  $T_M$  by both neutron powder diffraction (NPD) and zero-field nuclear magnetic resonance (ZFNMR).<sup>9,10</sup> It is therefore natural that simple canted ferromagnetism was assumed in many previous investigations. This model, however, faces a dilemma in accommodating the magnetizations and the ZFNMR and NPD data. NPD, for example, indicated that the Ru spins are antiferromagnetically aligned (G type) along the c axis with a very tight upper limit of the FM components, i.e., <0.1 and  $\approx 0.2 \mu_B/\text{Ru}$  at H=0 and  $0.4 \text{ T} \leq H \leq 7 \text{ T}$ , respectively.<sup>9</sup> The spontaneous magnetization of the sample, however, reaches  $M_r \approx 800$  emu/mole, i.e., an FM component of  $0.28\mu_B/\text{Ru}$  at H=0. The extrapolated zero-field magnetization of  $0.6\mu_B/\text{Ru}$  at 50 K,<sup>8</sup> which may serve as a lower limit for the FM component at 5 T, is again three times larger. The

ZFNMR data, in addition, demonstrate that the Ru spins should be aligned perpendicular to the c axis with a major (or dominant) FM component.<sup>10</sup> This unusual magnetic structure, which appears as G-type antiferromagnetism along the c axis in NPD but ordered along a, b with a large FM component in both magnetization and NMR, suggested that the magnetic structure of Ru1212R warranted a reexamination. It should be pointed out that both the extremely broad  $C_p$ peak and the super-paramagnet-like M(H) up to  $2T_M$  in Ru1212Gd already suggest that its magnetic transition is far from simple:<sup>3,8</sup> the spin correlations may exist up to  $2 T_M$ with a significant entropy and a correlation size as large as  $10^2 \mu_B - 10^3 \mu_B$ ,<sup>6</sup> both characteristic of phase separation. It is interesting to note that both  $T_M$  and  $T_{AM}$  of Ru1212R can be tuned by Cu doping.<sup>11</sup> The evolutions of M,  $\chi_{Ru only}$ , and  $C_n$  of  $\operatorname{Ru}_{1-x}\operatorname{Cu}_x\operatorname{Sr}_2\operatorname{EuCu}_2\operatorname{O}_{8+\delta}$  with  $0 \le x \le 0.15$ , therefore, were measured. The  $T_M$  drops more than 25 K with x while the variation in  $T_{AM}$  is negligibly small. A separation between  $T_M$  and  $T_{AM}$  is developed with x. This separation is further accompanied by a magnetic  $C_p/T$  with a flat plateau between  $T_M$  and  $T_{AM}$ . Hence, a mesoscopic phase separation is suggested.

Ceramic  $Ru_{1-x}Cu_xSr_2EuCu_2O_{8+\delta}$  samples with x between 0 and 0.15 were synthesized following a standard solid-state-reaction procedure. Precursors were first prepared by calcinating commercial oxides at 600-900 °C under flowing  $O_2$  at 1 atm. Mixed powder with a proper cation ratio was then pressed into pellets and sintered at 960 °C. The final heat treatment was done at 1065-1070 °C for 7 d in oxygen after repeatedly sintering and regrinding.<sup>4</sup> The structure of the samples was determined by powder x-ray diffraction using a Rigaku DMAX-IIIB diffractometer. The x dependence of the lattice parameters, i.e., the  $c \approx 11.553(2)$  to 11.550(2) Å for x=0 and 0.15, respectively, is slightly weaker than that reported for  $Ru_{1-r}Cu_rSr_2GdCu_2O_{8+\delta}$ . Minor impurity phases, likely SrRuO3 or oxides of (Sr,Cu), are below 5% at  $x \le 0.15$  (Fig. 1). The composition was measured by a JEOL JXA 8600 electron microprobe with attached wavelength-dispersive spectrometers. The local inho-



FIG. 1. The XRD of a  $(Ru_{0.9}Cu_{0.1})Sr_2EuCu_2O_{8+\delta}$  sample. +: data; solid line: the Rietveld fit; \*: the impurity line.

mogeneity of 1-x is within experimental resolution of  $\pm 0.05$ .<sup>12</sup> The magnetizations were measured using a Quantum Design superconducting quantum interference device (SQUID) magnetometer with an ac attachment and the specific heat was measured in a Quantum Design physical property measurement system with a specific-heat attachment.

Superconductivity appears in all the samples below a critical temperature  $T_c \approx 20-30$  K. A single-step jump of  $M_{FC}$  also appears with cooling at a higher temperature [Fig. 2(a)]. According to the scaling correlation  $(HM_0/MH_0)^{1/\gamma}$  $= t + (M/M_0)^{1/\beta}$ , the  $\partial M/\partial T$  of an ideal ferromagnet should decrease with  $t = (T - T_M)/T_M$  as  $1/t^{\gamma+1}$  above  $T_M$ , but increase as  $(-t)^{1-\beta}$  below, where  $0 < \beta < 1$ ,  $\gamma > 0$ ,  $H_0$ , and  $M_0$  are two critical exponents and two critical amplitudes, respectively. The situation for a CAFM magnet should be similar. Therefore, the inflection point of  $M_{FC}(T)$  at 5 Oe, i.e., the temperature at which  $\partial M_{FC}/\partial T$  peaks, is used as  $T_M$ [Fig. 2(a)]. The well defined  $T_M$  and the large FM component below  $T_M$  are in rough agreement with those reported for Ru1212Eu,<sup>8</sup> but rather different from those of  $Ru_{1-x}Cu_xSr_2GdCu_2O_{8+\delta}$ ,<sup>11</sup> where no clear FM transition can be identified with  $x \ge 0.1$ . Differences in both the rareearth elements and the synthesis procedures may contribute to the variation. It should be pointed out that the well defined  $T_M$  and the large  $M_{FC}$  of our samples make the analysis of  $M_{FC}$  and  $C_p$  easier and without significant interference from the minor impurities. A systematic decrease of  $T_M$  with x is observed, e.g.,  $T_M \approx 134$  K and 117 K at x=0 and 0.1, respectively [Fig. 2(a)]. It is also interesting to note that the reported bifurcation point between  $M_{ZFC}$  and  $M_{FC}$ , which should be very close to  $T_M$  if the domain pinning is strong, in  $Ru_{1-x}Cu_xSr_2GdCu_2O_{8+\delta}$  shows almost the same x dependence, i.e., down to  $\approx 115$  K and 100 K with x = 0.1 and 0.2, respectively.11

The  $\chi = M/H$  of a simple AFM magnet, which will be H independent far above its AFM transition, should have a maximum slightly above the Néel temperature,  $T_{AM}$ . It has been suggested, in fact, that the magnetic energy,  $E_m$ , and  $\chi$  should both depend on the pair-correlation functions  $\Gamma(r) = 3[S^z(0)S^z(r)]/S(S+1)$  as  $E_m \propto \Gamma_1$  and  $\chi \propto [1 + \Sigma_r \Gamma(r)]/T \approx [1 + f(T)\Gamma_1]/T$ , where  $\Gamma(r)$ ,  $\Gamma_1$ , and f(T) are the pair correlation with the pair distance = r, the corre-



FIG. 2. (a) Magnetizations. For the x=0 sample,  $\bigcirc: M_{FC}$  (5 Oe);  $+: H/M_{FC}$  (1 T);  $\textcircled{Oe}: 1/\chi_{Ru \ only}$ ; solid line: C-W fit. For the x=0.1 sample,  $\nabla: M_{FC}$  (5 Oe);  $\textcircled{V}: 1/\chi_{Ru \ only}$ . (b) Spin entropy.  $\bigcirc:$  the magnetic  $C_p/T$  of the x=0 sample;  $\bigtriangledown:$  that of the x=0.1 sample; solid line:  $\partial(T\chi)/\partial T$  of the x=0 sample; dashed line: that of the x=0.1 sample. (c) The evolution of  $T_{AM}-T_M$  with  $T_M$ . Oe: Cu-doped Ru1212Eu;  $\blacksquare:$  Ru1212Eu of Butera *et al.* (Ref. 8);  $\triangle:$  annealed Ru1222Gd;  $\Box:$  as-synthesized Ru1222Eu.

lation with the nearest neighbor, and a slowly varying function of *T*, respectively.<sup>7</sup> This leads to an approximation of  $C_p \propto \partial(T\chi)/\partial T$  if the short-range correlation  $\Gamma_1$  is dominant.  $T_{AM}$ , therefore, can be defined as the temperature of the  $\partial(T\chi)/\partial T$  peak,<sup>7</sup> which is observed close to the  $\chi$ -maximum temperature in three dimensions (3D) but much lower in 2D.<sup>13</sup> For CAFM magnets, an FM-like  $M_{FC}$  step may coexist with a  $\partial(T\chi)/\partial T$  peak. However,  $T_{AM} \approx T_M$  is expected, except for the possible *H*-induced transition shifts.<sup>14</sup>

To analyze the magnetization of  $Ru_{1-x}Cu_xSr_2EuCu_2O_{8+\delta}$ , the Eu/CuO<sub>2</sub> contributions were first eliminated using the procedure previously proposed,<sup>8</sup> i.e., with a Van Vleck susceptibility of free  $Eu^{3+}$  and a T independent  $\chi_0$  of  $8.7 \times 10^{-4}$  emu/mole for CuO<sub>2</sub>. For the undoped sample with x=0, the Ru contribution is H independent and follows a Curie-Weiss (C-W) fit only above 250 K with a C-W constant  $\approx 2.6 \mu_B/\text{Ru}$  and a Curie temperature of 127 K. Deviation from the C-W fit and large superparamagnetic M(H), however, develop at lower temperatures [Fig. 2(a)]. The Ru contribution to  $\chi$  at 1 T, for example, is more than 10% higher than that expected between 180 K and  $T_{AM}$  [Fig. 2(a)], indicating a dominant FM interaction. The 5-T differential Ru susceptibility after subtracting the Eu/CuO<sub>2</sub> contributions ( $\chi_{Ru \ only}$ ), however, shows an opposite downturn, suggesting significant AFM interactions [Fig. 2(a)]. In particular, a minimum of  $1/\chi_{Ru \ only}$  and a  $\partial(T\chi)/\partial T$  peak appear around 157 K [Fig. 2(a)] and  $T_{AM} \approx 138$  K [Fig. 2(b)], respectively, for the x=0 sample. Undoped Ru1212Eu, therefore, might be interpreted as a simple canted antiferromagnetic by either ignoring the 4-K difference between  $T_M$  and  $T_{AM}$ ,<sup>8</sup> or by regarding it as a small *H*-induced transition shift.

To further confirm the presumed  $T_{AM}$ , the magnetic specific heat was measured at zero field using a nonsuperconducting YBa<sub>2</sub>(Cu<sub>2.73</sub>Zn<sub>0.27</sub>)O<sub>7</sub> (YBCO) ceramic as the reference [Fig. 2(b)]. The raw specific heat of Ru1212 is well above that of YBCO between 80 and 180 K, but the two merge outside this region, a situation similar to the data of Ru1212Gd.<sup>3</sup> The magnetic  $C_p/T$ , i.e., the difference between Ru1212R and YBCO, shows a well-defined peak at 133 K, which is only slightly lower than the 138-K  $\partial (T\chi)/\partial T$  peak observed. This agreement between the  $C_p/T$ peak at zero field and the  $\partial (T\chi)/\partial T$  peak at 5 T again demonstrates that the procedure of Fisher<sup>7</sup> works reasonably well and that the *H*-induced transition shift is small in our case. It is also interesting to note the high-T tail of  $C_p/T$  and the non-C-W magnetization up to 180 K or higher [Figs. 2(a) and 2(b)]. Significant short-range spin orders, therefore,

should occur far above  $T_M$  and  $T_{AM}$ . With the Cu doping, however, the  $T_M$  and the  $T_{AM}$  evolve in different ways and the separation between them broadens. At x = 0.1, for example, the  $T_M$  is quickly suppressed to 117 K but the  $\partial(T\chi)/\partial T$  peak remains at 138 K [Figs. 2(a) and 2(b)]. The accompanying  $C_p/T$  appears to broaden with x as well [Fig. 2(b)]. In particular, the well-defined peak evolves into a flat plateau between  $T_M$  and  $T_{AM}$  [Fig. 2(b)]. It should also be pointed out that the separation at x = 0.1 is larger than the transition width in  $M_{FC}$ . Neither the sample inhomogeneity nor the experimental resolution, therefore, can account for the separation [Figs. 2(a) and 2(b)]. The AFM-like  $\partial(T\chi)/\partial T$  peak and the FM-like  $M_{FC}$  jump seem to carry distinct spin entropies of comparable strength.

It is therefore interesting to compare the data with that of Ru1222R, where two separate transitions have been observed in both magnetizations and Mössbauer spectra.<sup>1,6</sup> The  $T_M$  and  $T_{AM}$  of  $Ru_{1-x}Cu_xSr_2EuCu_2O_{8+\delta}$  $0 \le x \le 0.15$ , samples with the  $O_2/Ar$ -annealed  $RuSr_2(Gd_{1,4}Ce_{0,6})Cu_2O_{10+\delta}$ , and two as-synthesized Ru1222Eu samples are shown in Fig. 2(c).<sup>6</sup> The separation  $T_{AM} - T_M$  increases systematically with decreasing  $T_M$  in the Cu-doped Ru1212Eu: from an extrapolated zero separation at  $T_M \approx 140$  K to 25 K at  $T_M \approx 110$  K, where the data smoothly evolve into that of Ru1222R [Fig. 2(c)]. The observation of  $T_{AM} = T_M$  in the Ru1212Eu sample,<sup>8</sup> therefore, may be only a coincidence. Distinct AFM and FM transitions may coexist in both Ru1212R and Ru1222R.

These two transitions, as has been argued in the case of Ru1222R,<sup>1,6</sup> may be due to either a mesoscopic phase separation or a multistage transition. The magnetic properties between  $T_M$  and  $T_{AM}$ , however, will be different in these two



FIG. 3. (a) Cluster sizes for samples with igodots: x=0;  $\bigtriangledown$ : x=0.05;  $\blacksquare$ : x=0.10; and  $\blacklozenge$ : x=0.15 and for  $\bigcirc$ : as-synthesized Ru1222Eu (×4). Inset: The isothermal M(H) of the x=0 sample. (b) Relaxation of the remnant magnetizations at 160, 150, 140, and 130 K (from top to bottom) after field cooling at 50 Oe.

scenarios: some parts of Ru1212*R* should be in superparamagnetic states during phase separation, but should stay in a long-range spin-order state during a multistage transition. Evidence for the possible phase separation in Ru1222*R*, for example, is found in both the superparamagnetic M(H) with a magnetic cluster size of  $10^3 \mu_B$  and the slow spin dynamics far above  $T_{FM}$ .<sup>6</sup> Similar properties were therefore tested in Cu-doped Ru1212Eu.

The Langevin function with an additional linear term,  $a \cdot H + m \cdot [c \tanh(\mu H/k_BT) - k_BT/\mu H]$ , was used to fit the average magnetization in a M-H loop [inset, Fig. 3(a)].<sup>6</sup> The fit is reasonably good with the deduced  $\mu$  between  $100\mu_B$  and  $700\mu_B/c$ luster [closed symbols in Fig. 3(a)], which is four to five times smaller than those deduced in Ru1222R, but still far larger than that expected based on the spin fluctuations. A cluster of  $400\mu_B \approx 200$  Ru ions, for example, would be 4-5 nm or larger in an RuO layer. It should be further noted that  $\mu$  so deduced may be only a lower limit of the actual cluster/spin-correlation length.<sup>15</sup> The existence of such large clusters at  $T/T_M > 1.1$  will be difficult to be interpreted as a simple fluctuation. This deduced size, on the other hand, appears to be too small for a crystalline magnet, as is suggested in the multistage transition model.

The dynamic spin response was also studied. The logarithmic increase of  $M_{ZFC}$  at 5 Oe with time is almost unobservable, with the deduced rate of  $d \ln M/d \ln t < 10^{-3}$  well

within our experimental resolution, where 60 s < t < 3600 s is the time after the field switch. This is rather different from that of Ru1222Eu,<sup>6</sup> but in agreement with the unobservable relaxation of Ru1212R ac susceptibility reported between 1 s and 100 s.<sup>16</sup> The lack of relaxations under the above conditions is apparently related to the cluster size in Ru1212R [Fig. 3(a)], which is four to five times smaller and leads to quicker equilibrium. The slow spin dynamics, therefore, should either be explored in a shorter time window or after an enhancement of the energy barriers. Several different experimental conditions were then tested, and significant nonlogarithmic relaxations were observed in the remnant magnetization after a 50-Oe field cooling [Fig. 3(b)]. It is interesting to note that the energy barriers are  $\approx KV_c - \mu H$ and  $KV_c$ , respectively, for the  $M_{ZFC}$  and the remnant magnetization, where K and  $V_c$  are the magnetic anisotropy and the coherent volume, respectively. This may make the remnant magnetization a more favorable candidate for investigating the slow dynamics. The strong T dependence of the relaxation observed [Fig. 3(b)] suggests, in our opinion, that the relaxation observed is unlikely an artifact of the SQUID magnetometer, but supports the existence of superparamagnetic clusters.

As pointed out earlier, the phase-separation model may also offer a consistent interpretation for the conflicting NPD/ NMR and superconductivity data reported previously.<sup>6</sup> The conflict between the NPD and NMR data for the magnetic structure, for example, may be attributed to the fact that the two probes have different sensitivities to various magnetic species, such as those well documented in manganites.<sup>17</sup>

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Similarly, the spatial separation between AFM and FM species offers a natural mechanism for the unusual superconductivity observed.<sup>4</sup> Superconductivity can coexist with the AFM matrix. The finely dispersed FM clusters, on the other hand, depress the local SC order parameter and serve as tunnel barriers for the Cooper pairs. The superconductivity, therefore, may retain a significant part of the condensation energy, but appears only as a Josephson-junction array. Similarly, the critical temperature observed in the transport will naturally be much lower than that associated with the corresponding  $C_p$  anomaly,<sup>2,3</sup> and can be easily suppressed by external fields.<sup>12</sup> The intragrain penetration depth will also be much larger than those expected based on the proposed universal  $1/\lambda_2(T_c)$ .<sup>4</sup>

In summary, a systematic separation between  $T_M$  and  $T_{AM}$  is observed in Ru1212Eu with Cu doping, suggesting the coexistence of FM and AFM orders and the occurrence of a mesoscopic phase separation in the compound. The superparamagnetic M(H) as well as the slow spin dynamics further support the interpretation.

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