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Coexistence of ferromagnetism and cluster glass state in superconducting ferromagnet RuSr2Eu1.5Ce0.5Cu2O10

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Coexistence of ferromagnetism and cluster glass state in superconducting ferromagnet RuSr$_2$Eu$_{1.5}$Ce$_{0.5}$Cu$_2$O$_{10-\delta}$

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Systematic dc and ac magnetic susceptibility measurements performed on RuSr$_2$Eu$_{1.5}$Ce$_{0.5}$Cu$_2$O$_{10-\delta}$ (Ru1222) demonstrate a paramagnetic to ferromagnetic transition around 95 K. The third harmonic of the ac susceptibility reveals that the system undergoes a spin glass transition below 75 K. The features of the zero-field cooled and field cooled dc magnetization curves of Ru1222 material resemble those of a cluster glass state, rather than those of a canonical spin glass state. The magnetization versus applied field loops do not saturate, even at very high applied fields, resulting in the short range magnetic order in the system, which facilitates the formation of clusters that freeze at low temperature. The temperature dependence of the second and third harmonic ac susceptibilities further confirms the coexistence of a cluster glass state and ferromagnetic order in the Ru1222 system. © 2009 American Institute of Physics. [DOI: 10.1063/1.3063069]
the intercept of the straight line shown in the Fig. 1 the curves. ZFC curve has a peak at branch out, and the system enters into a glassy state. The TC transition around the peak temperature of RuSr$_2$Eu$_{1.5}$Ce$_{0.5}$Cu$_2$O$_{10-\delta}$. The inset shows the magnetization ($M$) vs $H$ curve at different temperatures in the range $-50,000 \text{ Oe} \leq H \leq +50,000 \text{ Oe}$.

FIG. 1. (Color online) ZFC and FC dc susceptibility curves of RuSr$_2$Eu$_{1.5}$Ce$_{0.5}$Cu$_2$O$_{10-\delta}$ measured at the applied magnetic field, $H =10 \text{ Oe}$. The inset shows the magnetization ($M$) vs $H$ curve at different temperatures in the range $-50,000 \text{ Oe} \leq H \leq +50,000 \text{ Oe}$.

Temperature range of 1.9–300 K. The linear and nonlinear ac susceptibilities were measured using a Physical Properties Measurement System (Quantum Design).

Figure 1 shows ZFC and FC dc susceptibility curves of the RuSr$_2$Eu$_{1.5}$Ce$_{0.5}$Cu$_2$O$_{10-\delta}$ sample measured at 10 Oe. The sharp rise of both the ZFC and the FC curves at $T_C=95 \text{ K}$ shows a PM to FM transition. Here, we have denoted $T_C$ as the intercept of the straight line shown in the Fig. 1 the $x$-axis. At temperatures close to $T_C$, the ZFC and FC curves branch out, and the system enters into a glassy state. The ZFC curve has a peak at $T_{\text{cusp}}=75 \text{ K}$ and the FC curve increases with decreasing temperature. The system undergoes a superconducting transition at $T_C=27 \text{ K}$ (the kinks in both the curves). In our papers, we show that Ru1222 undergoes the SG transition around the peak temperature of $\chi''(T)$, (see Fig. 2) resulting in the freezing spins. Therefore, the peak temperature is called the freezing temperature (denoted as $T_f$). This peak temperature corresponds to the peak in the ZFC curve with a few degrees of variation in the peak temperature because of the difference in the response of the system to dc and ac fields.

The temperature range of $70 \leq T \leq 100 \text{ K}$ is the critical region since the PM→FM→SG transitions take place in this region. SG behavior was confirmed by the typical features observed in the $\chi''(T)$ response of Ru1222: (i) the divergence of real part of $\chi''$ denoted as $\chi''|_{T_f}$ as $T → T_f$ according to the critical scaling law $\chi''|_{T_f} \propto e^{-\gamma}$, where $\gamma=(T-T_f)/T_f$ is the reduced temperature and $\gamma$ is the susceptibility exponent, which comes out to be 1.94 in our case, (see Fig. 3); (ii) the divergence of the magnitude of the peak value of $\chi''$ denoted as $|\chi''_{\text{max}}|$ in the limit of $H_{\text{ac}} \rightarrow 0$ (see inset of Fig. 3); and (iii) the temperature dependence of $\chi''$, which exhibits a sharp negative peak at $T_f$.

However, certain features are observed in the ZFC and FC curves which indicate that the Ru1222 system does not possess canonical SG behavior. A canonical SG system develops irreversibility between the ZFC and the FC curves below $T_f$ with the FC curve almost constant with decreasing temperature. A true SG state is realized by the freezing of individual spins at low temperature in the direction of the applied field. With decreasing temperature the magnetic moments reduce their thermodynamic energy and the moments will not prefer to deviate from the direction of the applied field, hence resulting in a constant FC magnetization. In Fig. 1, we observe that the irreversibility between the ZFC and the FC curves starts at a temperature just above $T_f$ with FC magnetization increasing with decreasing temperature. Such a behavior is observed in CG materials where FM interactions within the cluster tend to align the moments in the direction of the applied field, resulting in increased magnetization and a large difference between the ZFC and FC curves. Another possibility is that, since Ru1222 undergoes a PM to FM transition at $T_C$ followed by a SG transition, this system may also be classified as RSG, however, this possibility is ruled out since RSG materials show irreversibility far below $T_C$ and the ZFC curve develops a plateau rather than the sharp peak.

The inset of Fig. 1 shows magnetization ($M$) versus applied field ($H$) curves of Ru1222 measured at different temperatures. At low temperatures $M$−$H$ loops show a pronounced S-shaped curve that is characteristic of SG, which
dissolves into a linear curve with increasing temperature. The S-shaped curve also exhibits hysteresis and coercivity at low fields, indicating coexisting SG and FM states. Also, it can be observed that even under the highest magnetic field of 50 000 Oe the magnetization remains unsaturated, indicating that no canonical long range order exists in the Ru1222 system.13 This suggests that the formation of clusters is due to short range order and resulting in the CG state in this system.

To investigate this further, we have performed nonlinear ac susceptibility studies on our Ru-based system. Figure 2 shows the real and imaginary parts of the first harmonic of ac susceptibility, denoted as \( \chi' \) and \( \chi'' \), respectively, as functions of temperature, which were measured at different ac fields with zero dc bias. Figure 2(a) shows that the peak temperature of \( \chi'_1 \) is field independent whereas the peak height increases with increasing ac field. This is an unusual feature since it has been observed that \( \chi'_1 \) decreases with increasing ac field.4 This is because with an increase in the applied magnetic field, the magnetic energy associated with the external field becomes large enough compared to the thermodynamic energy of the magnetic moments to make it difficult for the magnetic moments to freeze in the direction of the applied field, and as a result \( \chi'_1 \) is reduced with increasing applied field. The unusual field dependence of \( \chi'_1 \) shows discrepancies from the canonical SG state in this system.

As discussed above, SG and FM coexist in Ru1222. This is confirmed by the appearance of an even harmonic (\( \chi'' \)) signal around the SG transition temperature. Figure 4 shows that \( \chi'' \) develops in the form of a sharp negative peak close to the peak temperature of \( \chi'_1 \), where the SG transition is supposed to occur, demonstrating the coexistence of SG and FM ordering. The magnetization is represented by Eq. (1). On the other hand, if there were a direct PM to SG transition, \( M \) would be expressed as an odd power series.11

\[
M = M_0 + \chi_1 H + \chi_2 H^2 + \ldots
\]  

Moreover, as can be observed in Fig. 4, \( \chi'' \) shows a nondiverging broad positive peak close to the temperature where \( \chi'_2 \) shows a peak. A similar feature is observed in other CG systems.9,13 In Fig. 2(b), \( \chi'' \) shows a kink (change in slope) around 64 K. With increasing applied ac field the kink shifts slightly toward the lower temperature, which suggests that the freezing of clusters is taking place at a temperature just below the CG transition (\( T_H = 75 \) K). A similar change in slope around 60 K is also visible in \( \chi'' \) (see Fig. 4). This can explain the sudden rise in the temperature dependence of the coercivity, \( H_C(T) \) around 60 K in Ref. 2. The small peak adjacent to the main peak in \( \chi'_1 \) and the change in the sign of \( \chi'_2 \) from negative to positive at the main peak temperature of \( \chi'_1 \) are unusual features observed for the first time. They may arise due to the field dependent dynamic response of intercluster or intracluster FM interactions. However, a further discussion on this issue is beyond the scope of this paper.

The dc and ac susceptibility studies presented in this paper suggest that a CG state exists in the Ru1222 system. Along with the short range FM order resulting in the formation of clusters, there are various other factors that support the presence of cluster formation in Ru1222. First, the Ru1222 system exhibits oxygen nonstoichiometry, which leads to the reduction of Ru5+ to Ru4+, resulting in cluster formation.13 Hence the random distribution of clusters with Ru5+−Ru5+, Ru5+−Ru4+, and/or Ru4+−Ru4+ exchange interactions may lead to CG state. Second, the synthesis of Ru1222 is always accompanied by the presence of clusters of minority RuSr2EuCu2O8 and SrRuO3 phases, which affect the magnetic and transport properties of the Ru1222 system.2,14

In conclusion, dc magnetization and nonlinear ac susceptibility measurements have revealed that the Ru1222 system undergoes PM to FM ordering, followed by a SG transition. However, the system does not possess a canonical SG state, rather a CG state is observed, which bears similar features to those of SG and RSG. At low temperature a clusterlike glassy state coexists with FM ordering. Superconductivity in Ru1222 is not affected by the cluster SG state or the FM ordering, so as a result, these three phenomena coexist. This is because superconductivity resides in CuO2 plane, whereas magnetism originates from RuO2 plane, and these are partially decoupled.1

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