Effect of disorder on quantum phase transition in the double layered ruthenates (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$

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(R$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$ is characterized by complex magnetic states, spanning from a long-range antiferromagnetically ordered state over an unusual heavy-mass nearly ferromagnetic (NFM) state to an itinerant metamagnetic (IMM) state. The NFM state, which occurs in the $0.4 > x > 0.08$ composition range, freezes into a cluster spin glass (CSG) phase at low temperatures [Z. Qu et al., Phys. Rev. B 78, 180407(R) (2008)]. In this article, we present the scaling analyses of magnetization and the specific heat for (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$ in the $0.4 > x > 0.08$ composition range. We find that in a temperature region immediately above the spin freezing temperature $T_f$, the isothermal magnetization $M(H)$ and the temperature dependence of electronic specific heat $C_e(T)$ exhibit anomalous power-law singularities; both quantities are controlled by a single exponent. The temperature dependence of magnetization $M(T)$ also displays a power-law behavior, but its exponent differs remarkably from that derived from $M(H)$ and $C_e(T)$. Our analyses further reveal that the magnetization data $M(H,T)$ obey a phenomenological scaling law of $M(H,T) \propto H^{\lambda} f(H/T^\beta)$ in a temperature region between the spin freezing temperature $T_f$ and the scaling temperature $T_{\text{scaling}}$. $T_{\text{scaling}}$ Systematically decreases with the decease of Ca content. This scaling law breaks down near the critical concentration $x = 0.1$ where a CSG-to-IMM phase transition occurs. We discussed these behaviors in term of the effect of disorder on the quantum phase transition.

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I. INTRODUCTION

How a phase transition is affected by disorder is an important problem extensively studied in condensed matter physics. For a classical phase transition, the system behaves as a clean system extensively studied in condensed matter physics. This is a weak effect.

In this scenario, thermodynamics quantities are expected to display “anomalous” power-law singularities controlled by a single continuously varying exponent, e.g., the specific heat $C(T) \propto T^\gamma$, the magnetic susceptibility $\chi(T) \propto T^{\delta-1}$, and the zero-temperature isothermal magnetization $M(H) \propto H^\nu$, where $\nu$ is a nonuniversal Griffiths exponent and ranges from 0 to 1.18,19

In many itinerant systems close to QPTs, various behaviors reminiscent of the quantum Griffiths singularities have been observed, and their underlying physics is still under debate.15–17 Theoretically, it has been suggested that itinerant systems should show quantum Griffiths phenomena similar to those in insulating magnets.1,18 However, further theoretical studies suggest that perturbations in itinerant systems could significantly modify this scenario. For example, Landau damping was argued to prevent the tunneling of the rare regions and suppress the Griffiths singularities at low temperatures.20,21 Moreover, it has also been proposed that for the Ising case the QPT is fundamentally modified by the overdamping,22 while in the Heisenberg system the standard quantum critical point (QCP) scenario should hold and the power-law quantum Griffiths singularities could be retained.23 The recent work by Dobrosavljević and Miranda considered the correlation among individual rare regions in itinerant systems and pointed out that such correlations destabilize the quantum Griffiths phase, leading to a generic formation of a cluster spin glass (CSG) phase preceding uniform ordering.24 In the meantime, considerable effort has been devoted to experimental study of the effect of disorder on QPTs. Many experimental results associated with the quantum Griffiths phases have been reported in itinerant systems such as magnetic semiconductors Fe$_1-x$Co$_x$S$_2$,25 the heavy fermion system CePd$_{1-x}$Rh$_x$,26 and the metal alloy Ni$_{1-x}$V$_x$.27 However, the effect of disorder on...
QPTs was rarely investigated in the transition metal oxides (TMOs).

Here we focus on the double layered ruthenates (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$. The end member Ca$_3$Ru$_2$O$_7$ ($x = 1$) is an antiferromagnet with $T_f = 56 \text{ K}$, whose ground state consists of ferromagnetic (FM) bilayers stacked antiferromagnetically along the $c$ axis.\textsuperscript{28–31} Sr substitution for Ca continuously suppresses the $T_f$, from 56 K for $x = 1$ to 30 K for $x = 0.4$, and eventually results in the collapse of the AFM order for $x < 0.4$.\textsuperscript{32–35} For $0.4 > x > 0.08$, an unusual heavy-mass nearly ferromagnetic (NFM) state is observed, which is characterized by an extremely large Wilson ratio $R_w$ (e.g., $R_w \sim 700$ for $x = 0.2$).\textsuperscript{34} Despite considerably strong FM correlations, the system does not develop a long-range order at low temperatures but freezes into a CSG phase due to the close proximity to a 2D FM instability.\textsuperscript{34,35} When Ca content is decreased below 0.08, the system enters an itinerant metamagnetic (IMM) state.\textsuperscript{34} Angle-resolved photoemission spectra of (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$ measured at 20 K reveal a qualitative change of electronic properties near $x \sim 0.3$–0.4, which is suggested to be associated with a QPT.\textsuperscript{36} Specific heat and resistivity measurements down to 0.3 K have proved the existence of non-Fermi-liquid (NFL) behavior near $x = 0.1$.\textsuperscript{34,37} corroborating the existence of a QPT in this system. Since the system is tuned via chemical substitution, strong disorder is introduced into the system, making the double layered ruthenates (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$ a good candidate to explore the effect of disorder on QPT in TMOs.

In this article, we present the scaling analyses of magnetization and the specific heat for (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$ in the $0.4 > x > 0.08$ composition range. We find that in a temperature region immediately above the spin freezing temperature $T_f$, the isothermal magnetization $M(H)$ and the temperature dependence of electronic specific heat $C_e(T)$ exhibit anomalous power-law singularities; both quantities are controlled by a single exponent. The temperature dependence of magnetization $M(T)$ also displays a power-law behavior but its exponent differs remarkably from that derived from $M(H)$ and $C_e(T)$. Moreover, the magnetization data $M(H, T)$ are found to obey a phenomenological scaling law of $M(H, T) \propto H^\beta (H/T)\gamma$ in a temperature region immediately above $T_f$. The scaling temperature $T_{\text{scaling}}$ below which the scaling equation holds systematically decreases with the decease of Ca content. This scaling law breaks down near the critical concentration $x = 0.1$, where a CSG-to-IMM phase transition occurs. We discussed these behaviors in term of the effect of disorder on QPTs.

### II. EXPERIMENT

High-quality single crystalline samples of (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$ were grown using a floating-zone technique. Magnetization measurements were performed using a commercial SQUID magnetometer (Quantum Design) with the magnetic field applied in the basal plane. The specific heat was measured using a thermal relaxation method in a Quantum Design PPMS. The experiments were performed on single crystals that had been characterized in our earlier works.\textsuperscript{34,37–39}

![FIG. 1. (Color online) The temperature dependence of magnetization $M(T)$ under an applied field of 0.005 T with field-cooling (filled symbols) and zero-field-cooling (open symbols) histories for (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$ with $x = 0.3$ (a), 0.25 (b), 0.2 (c), 0.15 (d), and 0.1 (e). The temperature dependence of AC susceptibility measured at 10 Hz, 100 Hz, 1 kHz, and 10 kHz for $x = 0.03$ and 0.1 are also shown in (a) and (e). The magnetic field is applied perpendicular to the $c$ axis. $T_f$ (arrow) marks the freezing temperature of the CSG phase. The dash lines represent fittings to $T^{-\beta}$. The data for $x = 0.3$ and 0.1 are quoted from from our earlier works.\textsuperscript{34,39}](https://link.to/fig1)

### III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of magnetization $M(T)$ taken under an applied field of 0.005 T with field-cooling (FC) and zero-field-cooling (ZFC) histories for typical samples (the data for $x = 0.3$ and 0.1 are quoted from our earlier works\textsuperscript{34,39}). The samples freeze into a CSG phase below $T_f$. This is evidenced by (i) the irreversibility of $M(T)$ curves between ZFC and FC histories below $T_f$ [shown in Figs. 1(a)–1(d)] and (ii) a frequency dependence of the peak temperature in the real part of AC susceptibility [e.g., see the data for $x = 0.3$ in Fig. 1(a)]. For $x = 0.1$, $T_f$ decreases to $\sim 1 \text{ K}$, which could not be detected in present measurements. However, distinct irreversibility is observed in $M(T)$ between ZFC and FC histories below $\sim 10 \text{ K}$ [shown in Fig. 1(e)]. Given that ferromagnetic correlations are still considerably strong for $x = 0.1$, as manifested in the large Wilson ratio ($\sim 110$ at 2 K), and that quantum fluctuations are significantly enhanced,\textsuperscript{34} the irreversible characteristic of $M(T)$ below 10 K may imply formation of ferromagnetic clusters with dynamic nature. Further investigation is needed to demonstrate this point of view. Additionally, we note that all samples with $0.4 > x > 0.08$ show power-law singularities both in DC magnetization and AC susceptibility, i.e., $\propto T^{-\beta}$, in a temperature region immediately above $T_f$. Those dashed lines in Fig. 1 represent power-law fitting and the power-law exponents derived from fittings are shown in Fig. 4(a).
the temperature dependence of the crossover field. Upper insets show the temperature dependence of the exponent $\alpha$.

Figs. 2(a)–2(e) show the temperature dependence of the power-law field dependence in a high-field region, for a given temperature. We have conducted power-law fitting for the data in high-field region, but a power-law dependence in a low-field region, following a power-law manner, i.e., $H^* \propto T^\beta$ (see lower insets to Fig. 2). This means that $M(H)$ is approaching the power-law dependence on cooling, suggesting that the system develops toward the criticality. Power-law singularity is also found in the specific heat. The electronic specific heat data were obtained by subtracting the phonon contribution from the total specific heat. As shown in Fig. 3, $C_e/T$ shows a power-law temperature dependence for $x = 0.15$, $0.2$, $0.25$, and $0.3$, i.e., $C_e/T \propto T^{-\gamma}$, in a temperature region right above the freezing temperature of the CSG phase. In contrast, the $x = 0.1$ sample does not exhibit a similar power-law behavior in $C_e/T$ vs. $T$.

Figure 4(a) summarizes the exponents $\alpha$, $\beta$, and $\gamma$ extracted from $M(H)$, $M(T)$, and $C_e(T)$ as described above. It is interesting that the exponent $\gamma$ approximately equals the exponent $\alpha$ and falls into the range of 0 to 1 for $x = 0.15$–0.3. This power-law singularity appears to be consistent with the prediction of the quantum Griffiths model. However, several deviations from the prediction of the quantum Griffiths model are noticed. The exponent $\beta$ derived from $M(T)$ is apparently larger than the exponents $\alpha$ and $\gamma$ and does not fall into the expected range of 0 to 1 (see Fig. 4(a)]. Meanwhile, the anomalous power-law behaviors do not persist down to the...
The inconsistency between the exponents $\alpha$, $\beta$, and $\gamma$ can also be understood in light of the interaction between the droplets. In the present system, there is extremely strong FM fluctuations in the composition range of $0.4 > x > 0.08$ as manifested in the large Wilson ratios.

The interactions between magnetic droplets are then expected to be FM. Therefore, the magnetization of the system should grow faster than isolated droplets on cooling; this leads to a super-Curie temperature dependence, resulting in a power-law behavior with the exponent larger than 1. If this is the case, the evolution of the exponent $\beta$ determined from $M(T)$ should follow the same trend as that of the FM fluctuations, which are exactly observed here. For example, at a temperature immediately above $T_f$, the FM fluctuations revealed by the Wilson ratio for $x = 0.3$ are apparently much weaker than those for $x = 0.2$ and 0.25, and the $\beta$ for $x = 0.2$ and 0.25 are strikingly larger than that for $x = 0.3$ [see Fig. 4(b)]. In the high-field region, spin fluctuations are suppressed. Thus, the exponent $\alpha$ extracted from $M(H)$ does not equal the exponent $\beta$ derived from $M(T)$. The consistency of the exponent $\gamma$ with $\alpha$ indicates that the electronic specific heat is dominated by the slow dynamics of droplets, as expected in the quantum Griffiths model, but less sensitive to the interaction between droplets.

To further reveal the underlying physics of the anomalous power-law behaviors above $T_f$, we have performed the phenomenological scaling analyses of magnetization using the data discussed above. Given that $M(H)$ shows a linear-to-power-law crossover at a crossover field $H^*$, analytically we have $\chi(T)H^* = AH^\alpha F(x)$ at $H = H^*$, where $A$ is a constant. Based on this equation, we propose a phenomenological scaling equation $M(H,T) \propto H^\delta F(x)$, where $F(x = H/H^*(T))$ is the scaling function. As discussed above, $\chi = M/H$ is proportional to $T^{-\beta}$. Therefore, it can be shown that $H^* \propto T^\delta$ with $\delta = \beta/(1 - \alpha)$. As shown in Fig. 4(b), it is noticed that the values of $\delta$ experimentally determined from $H^*(T)$ (see the insets in Fig. 2) approximately equal to those calculated with the equation $\delta = \beta/(1 - \alpha)$, suggesting the self-consistency of the proposed phenomenological scaling law.

The validity of this phenomenological scaling expression is demonstrated in the scaling plots shown in Fig. 5, where $M/H^\alpha$ is plotted as a function of $H/T^\delta$. The magnetization data amazingly collapse into a single curve in a temperature range immediately above $T_f$ for the samples with $x = 0.3, 0.25, 0.2$, and 0.15. The characteristic temperature below which the scaling behavior occurs is defined as the scaling temperature $T_{\text{scaling}}$. At temperatures above $T_{\text{scaling}}$, the $M(H,T)$ data deviate from the universal curve on the $M/H^\alpha$ vs. $H/T^\delta$ scaling plot. We present an example for the $x = 0.3$ sample in the inset of Fig. 5(a), where the $M(H,T)$ data clearly do not collapse into a universal curve for $T > T_{\text{scaling}} = 13$ K. $T_{\text{scaling}}$ systematically decreases with the decrease of Ca content. When the Ca content $x$ decreases to 0.1 near which a CSG-to-IMM phase transition occurs, the scaling law breaks down and the magnetization data clearly do not collapse into a single curve [shown in Fig. 5(e)]. The breakdown of the phenomenological scaling behavior at $x = 0.1$, as well as the absence of the anomalous power-law behavior of electronic specific heat in this sample, suggests that the underlying physics near this critical composition might differ. We have added $T_{\text{scaling}}$ to the magnetic phase diagram of
FIG. 5. (Color online) Scaling plots of $M/H^\alpha$ vs. $H/T^\beta$ for the samples with $x = 0.3$ (a), 0.25 (b), 0.2 (c), 0.15 (d), and 0.1 (e). Inset to (a) shows the gradual deviation from the universal curve when the temperature exceeds the scaling temperature $T_{\text{scaling}}$.

(Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$, as shown in Fig. 6. Interestingly, $T_{\text{scaling}}(x)$ appears to follow the extrapolation of Néel temperature $T_N(x)$, implying that the phenomenological scaling behavior reported here might be a fingerprint of some energy scale in a disordered itinerant system close to a quantum phase transition.

FIG. 6. (Color online) The updated magnetic phase diagram of (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$.

IV. CONCLUSION

In summary, we have conducted the scaling analyses of magnetization and electronic specific heat for the NFM states in (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$. For $0.4 > x \geq 0.15$, the isothermal magnetization and the electronic specific heat exhibit anomalous power-law singularity that is controlled by a single exponent in a temperature region immediately above $T_f$. $M(T)$ also follows a power-law behavior. However, the extremely strong FM fluctuations lead its exponent to differ from those derived from the isothermal magnetization and the electronic specific heat. The magnetization data $M(H,T)$ are found to obey a phenomenological scaling law of $M(H,T) \propto H^\alpha f(H/T^\beta)$ also in a temperature region immediately above $T_f$. The scaling temperature $T_{\text{scaling}}$ systematically decreases with the decrease of Ca content. This scaling law, as well as the anomalous power-law behavior, break down near the critical concentration $x = 0.1$ where a CSG-to-IMM phase transition occurs. These results demonstrate the existence of the slow dynamics of rare regions arising from the effect of disorder on QPT in (Sr$_{1-x}$Ca$_x$)$_3$Ru$_2$O$_7$. The finding of the $M(H,T)$ scaling law implies novel physics associated with the slow dynamics of rare regions. Further theoretical work is needed to reveal its nature.

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