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Rapid suppression of the energy gap and the possibility of a gapless hidden order state in $URu_{2-x}Re_xSi_2$

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ABSTRACT

We investigated the energy gap associated with the hidden order (HO) phase and the Grüneisen ratio in the $URu_{2-x}Re_xSi_2$ system using a combination of thermal expansion coefficient and specific heat measurements. As the HO phase transition is suppressed to lower temperature, the ratio between the energy gap and the HO transition temperature decreases three-fold. This rapid suppression of the energy gap potentially leads to a scenario of a 'gapless' HO state, in which the energy gap of the HO phase vanishes before the HO transition temperature is suppressed to 0 K. We also investigated the Grüneisen ratio in the vicinity of the Re substituent composition where the HO is suppressed. The Grüneisen ratio shows divergent behaviour at x=0.12 and 0.15, providing evidence for the existence of a quantum critical point, which is consistent with our hypothesis of a 'gapless' HO state.

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1. Introduction

The heavy fermion compound URu_2Si_2 exhibits unconventional superconductivity that coexists with a mysterious hidden order (HO) phase [1–5]. The central issue surrounding the HO phase is the identification of the order parameter underlying the entropy loss associated with the HO transition. Analysis of the specific heat indicates that a charge gap $\Delta = 11$ meV opens over about 40% of the Fermi surface [1]. Inelastic neutron scattering experiments further reveal that spin gaps open at both a commensurate wave vector $Q_0 = (1,0,0)$ and an incommensurate wave vector $Q_1 = (0.4,0,0)$, simultaneously with the opening of the charge gap [6, 7]. It has also been shown that gapping of the

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incommensurate spin fluctuations can account for the loss of entropy at the HO transition [7]. Therefore, the charge and spin gaps are clearly important features of the HO phase.

The substitution of Re for Ru in URu_2Si_2 rapidly suppresses superconductivity and the HO phase, and induces a ferromagnetic (FM) phase transition at higher Re substitution levels [8–10]. The systematic suppression of the HO with Re concentration x in $URu_{2-x}Re_xSi_2$ provides an opportunity to investigate the evolution of the energy gap, as well as the possible existence of a quantum critical point (QCP) associated with the suppression of the HO phase. Non-Fermi-liquid (NFL) behaviour occurs in the $URu_{2-x}Re_xSi_2$ system that persists deep within the FM phase and appears to vanish at a QCP in the vicinity of x=0.15 [8–10].

In this paper, we report the results of an investigation of the energy gap Δ associated with the HO phase and Grüneisen ratio in the $URu_{2-x}Re_xSi_2$ system by means of thermal expansion and specific heat measurements. Our results suggest a scenario of a 'gapless' HO state in which Δ vanishes before T_0 is suppressed to 0 K. This scenario is consistent with the existence of a QCP, indicated by the divergent Grüneisen ratio in the vicinity of the Re concentration where the HO is suppressed.

2. Experimental methods

Single crystals of $URu_{2-x}Re_xSi_2$ were grown in a tetra-arc furnace using the Czochralski method. Thermal expansion measurements were performed at temperatures down to 2 K in a Quantum Design DC-PPMS-9 with a Dilatometer Measurement Option (Model P680) and down to 0.4 K in a ^{3}He system using a capacitive dilatometer constructed of OFHC copper [11]. Specific heat measurements were performed at temperatures down to 1.8 K in a Quantum Design DC-PPMS-9 and down to 0.5 K with the ^{3}He option for the DC-PPMS-9, as well as in a home made ^{3}He calorimeter down to 0.8 K.

3. Results

The data collected for β/T and C_e/T as a function of T are shown in Figure 1. β is the volume thermal expansion coefficient, calculated from the linear thermal expansion coefficients in the ab-plane and along the c-axis. C_e is the electronic contribution to the specific heat, which was obtained by subtracting the phonon contribution to the specific heat from the total specific heat (the Debye temperature estimated from the phonon contribution is in the range 250 K to 280 K for all of the substitution levels investigated). β/T can be clearly divided into two groups with distinct features. For x<0.12, we observe an anomaly in β/T associated with the HO phase transition in the temperature range 10–20 K. The transition temperature and the size of the anomaly are

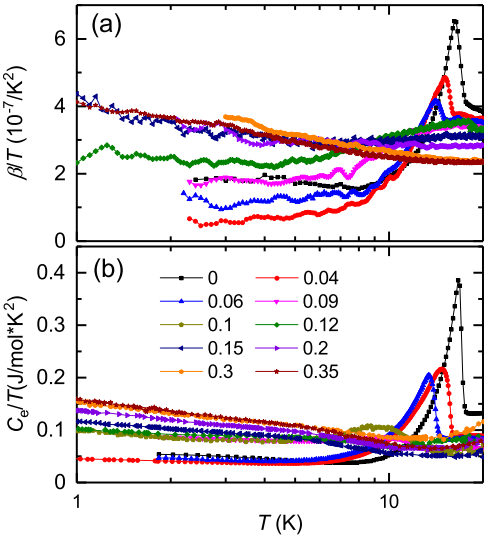

Figure 1. (Color online) (a) Electronic specific heat C_e divided by temperature T vs. T and (b) volume thermal expansion coefficient β divided by T vs. T for $URu_{2-x}Re_xSi_2$ single crystals with various values of x between 0 and 0.35.

suppressed systematically upon Re substitution. Similar behaviour can be seen in the specific heat data. Below the onset of the HO phase transition, β contains an exponential term arising from the gap in the HO phase, a linear term from the Fermi liquid background, as well as the phonon contribution term. Therefore, the $\beta(T)$ data can be fitted with a combination of these three terms, given by

$$\beta(T) = AT + B\exp\left(-\frac{\Delta}{T}\right) + CT^3, \tag{1}$$

as shown in Figure 2(a). Similarly, $C_e(T)$ contains an exponential term and a linear term from the Fermi liquid background, and can be fitted with the expression

$$C_e(T) = AT + B\exp\left(-\frac{\Delta}{T}\right), \tag{2}$$

as shown in Figure 2(b). The phonon contribution to the thermal expansion data is negligible compared with the two other terms, contributing less than 1% to the total thermal expansion. The coefficients A of the electronic term in the thermal expansion and specific heat data lie in the range 1×10^{-7} to 2×10^{-7} K^{-2} and 40 to 60 mJ/mol-K^2, respectively.

By fitting the above two expressions to the $\beta(T)$ and $C_e(T)$ data, one can extract the energy gap Δ of the HO phase. To account for the broadness of the phase transition, the error bars associated with T_0 are estimated using the entropy conserving construction described in Figure 3. The error bars for Δ are taken as the standard deviation of the fits. The energy gap extracted from $\beta(T)$ and $C_e(T)$ matches quite

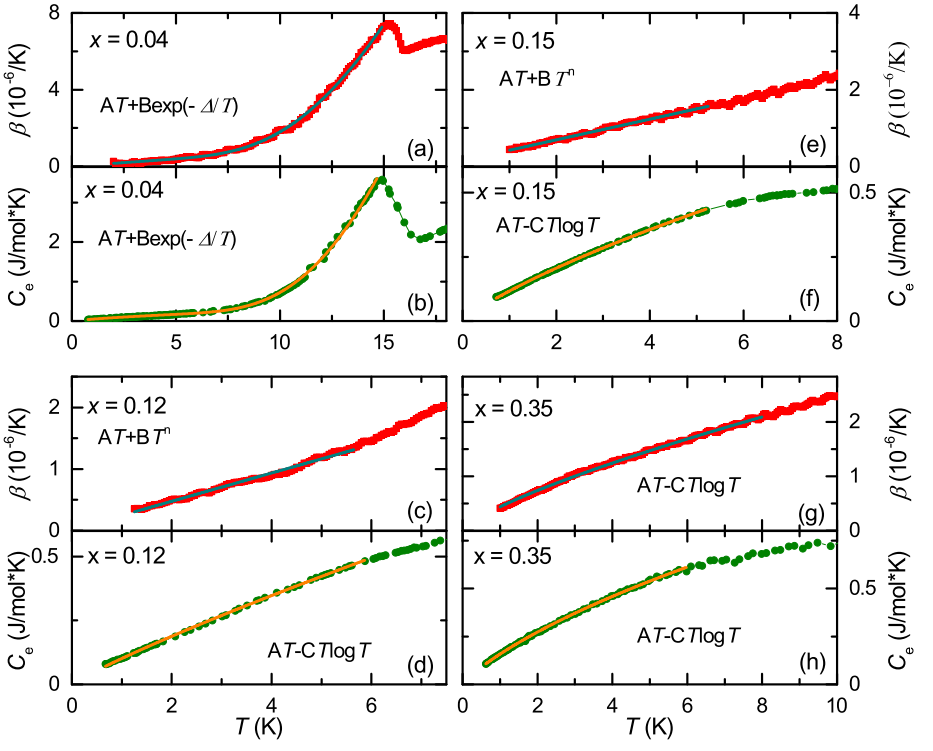

Figure 2. (Color online) The volume thermal expansion coefficient β (a, c, e, g) and electronic specific heat C_e (b, d, f, h) vs. temperature T for $URu_{2-x}Re_xSi_2$ single crystals with x=0.04, 0.12, 0.15 and 0.35, respectively. The lines represent the fits of the corresponding functions to the data.

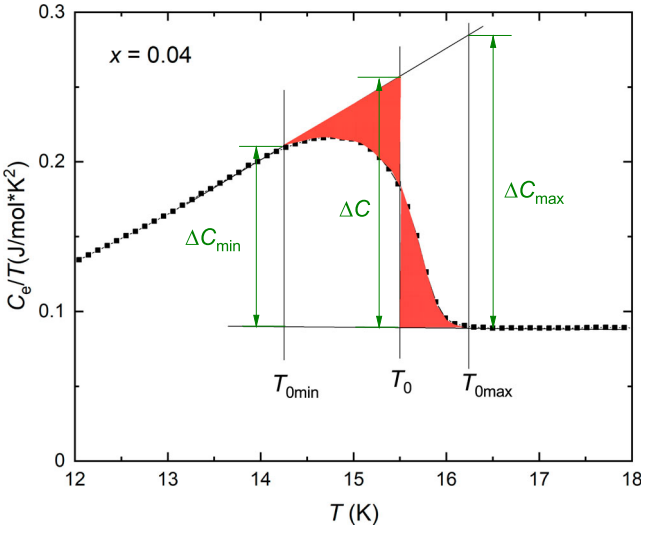

Figure 3. (Color online) Entropy conserving construction to evaluate the error bar in T_0 and ΔC. The two red regions have the same area.

well, as shown in Figure 4(a). The size of the gap decreases as T_0 is suppressed; however, it does not extrapolate linearly to zero at $T_0 = 0$ K. Instead, the energy gap is suppressed faster with x than T_0, leading to a reduction of the ratio between Δ and T_0, as shown in Figure 4(b). As x increases, the HO phase transition is broadened. Although the exact nature of the HO phase is still unknown, the temperature-dependence of β and C_e are reminiscent of a density wave [12, 13]. However, in mean field theory, both the transition temperature and energy gap of a density wave are exponential functions of $-1/\lambda$ (λ is the electron-electron coupling constant), leading to a constant Δ/T_0 ratio of 1.76, the weak-coupling BCS relation [12, 13]. In this context, the observed decrease of Δ/T_0 for the HO phase seems surprising.

It is noteworthy that the rapid suppression of the energy gap and the decrease of the Δ/T_0 ratio with T_0 appear to be universal for the HO phase, with respect to the tuning parameters employed in these experiments. Data for other URu_2Si_2-based substituted systems reported in the literature [14–17] are also included in Figure 4. These data represent substitutions on all three atomic sites for both electron and hole doping. All of the data seem to collapse onto

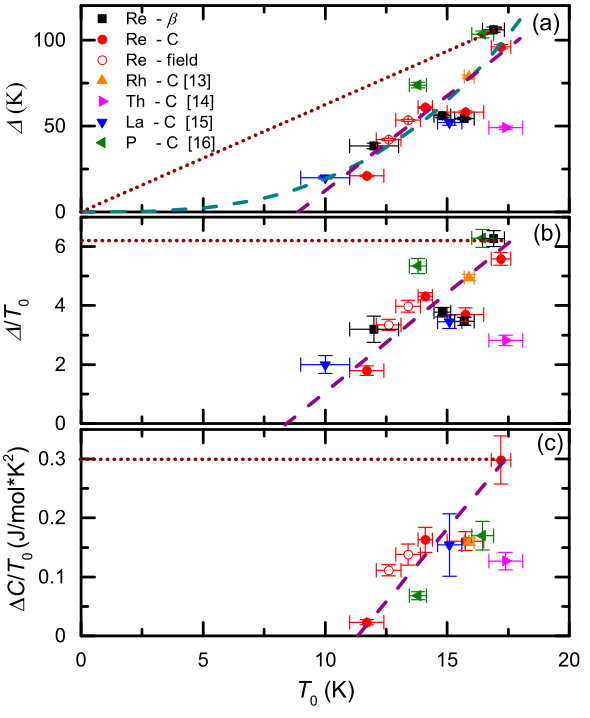

Figure 4. (Color online) (a) Energy gap Δ extracted from the volume thermal expansion coefficient and electronic specific heat, (b) energy gap divided by the HO transition temperature, Δ/T_0, and (c) jump in the electronic specific heat divided by the HO transition temperature, $\Delta C/T_0$, vs. T_0 for $URu_{2-x}Re_xSi_2$ single crystals. Data for other URu_2Si_2-based substituted systems taken from the literature are also presented for comparison [14–17]. The dotted line represents the behaviour expected from the 'BCS law of corresponding states' in which Δ scales with T_0, while the dashed lines are linear and power law extrapolations of the data points.

a single curve, except for the Th substitution into the U site, which leads to a faster suppression of Δ/T_0 and has a more drastic effect on the HO and superconducting transitions of URu_2Si_2 than other substitutions [17].

Another way to suppress the HO phase is to apply a magnetic field. In contrast to Re substitution, a magnetic field suppresses the HO phase to 0 K in a continuous manner [18–20]. We applied magnetic fields up to 14 T on a Re-substituted sample with x=0.06 as shown in Figure 5. The energy gap extracted from these data follows the same behaviour, which can be seen in Figure 4. This again illustrates the apparent universal manner in which the HO phase is suppressed, given that magnetic field and Re substitution suppress the HO via different mechanisms. It will be interesting to explore how the energy gap evolves as the HO is completely suppressed in a magnetic field. This requires a significantly higher magnetic field and will be investigated in a future experiment.

The rapid suppression of the energy gap potentially leads to a very interesting scenario: the energy gap vanishes before T_0 reaches zero. This can be seen from extrapolation of the Δ vs. T_0 data in Figure 4(a): Linear extrapolation of the energy gap terminates at 0 K for $T_0 = 8$ K; power-law extrapolation leads to a negligibly small energy gap below $T_0 = 4$ K. In both cases, the energy gap essentially vanishes at finite T_0. Unfortunately, the signatures of the HO phase transitions in transport and thermodynamic properties, which are themselves a reflection of the energy gap, can no longer be discerned below $T_0 = 10$ K, making it difficult to determine T_0 unambiguously. Extrapolation of T_0 from lower Re concentration leads to about 6 K at x=0.12, and 0 K at $x \approx 0.14$, as shown in Figure 6. Interestingly, this is close to the estimated location of the FM QCP. At x=0.12, fits of both $\beta(T)$ and $C_e(T)$ already yield a negligible value of the energy gap. This observation

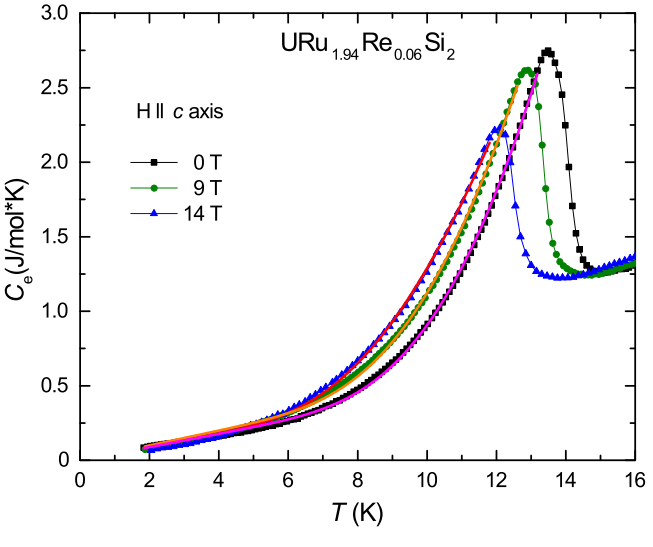

Figure 5. (Color online) Electronic specific heat C_e vs. T for $URu_{2-x}Re_xSi_2$ single crystal with x=0.06 in various magnetic fields applied along c-axis.

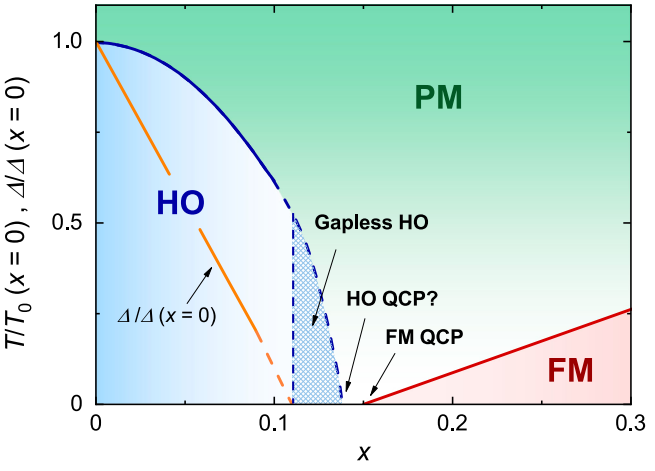

Figure 6. (Color online) Cartoon phase diagram of normalised temperature T/T_0 (x=0) and normalised energy gap Δ/Δ (x=0) vs. Re concentration x for $URu_{2-x}Re_xSi_2$ single crystals.

is reminiscent of a similar scenario in conventional superconductors containing paramagnetic impurities in which the intra-atomic exchange interaction between the impurity and conduction electron spins is ferromagnetic. In this case, the spin-dependent scattering of electrons by the paramagnetic impurities 'breaks' superconducting electron pairs and introduces states into the superconducting energy gap, reducing its magnitude. When the scattering becomes strong enough, the energy gap vanishes at all temperatures, a phenomenon referred to as 'gapless superconductivity' [21, 22]. In the present case, we seem to have observed a type of 'gapless hidden order' which may be consistent with aspects of HO that involve some type of density wave [1].

In the case of gapless superconductors, not only the energy gap Δ, but also the size of the jump in specific heat ΔC at the superconducting critical temperature T_c, is suppressed below the BCS law of corresponding states in which Δ and ΔC at T_c scale with T_c. Figure 4(c) shows $\Delta C/T_0$ at T_0 for $URu_{2-x}Re_xSi_2$ in zero and two magnetic fields, 9 T and 14 T, as well as other substitutions in URu_2Si_2 taken from the literature [14–17]. As mentioned above, the error bars have been estimated using the entropy conserving construction shown in Figure 3. It can be seen that $\Delta C/T_0$ decreases rapidly with decreasing T_0 and vanishes at around 10–15 K. This observation again underscores the possibility of a gapless HO phase. It is interesting that the suppressions of Δ and ΔC at T_0 are much faster than those observed in the simple case of superconductors containing paramagnetic impurities with a ferromagnetic intra-atomic exchange interaction. The much more rapid suppressions of Δ and ΔC at T_0 with T_0 are reminiscent of the behaviour of superconductors containing paramagnetic impurities with an antiferromagnetic intra-atomic exchange interaction which gives rise to the Kondo effect [22–25].

Our observations provide important information about the nature of the HO in URu_2Si_2 and may place constraints on potential models for the order parameter of the HO phase. It has been shown that the gapping of the incommensurate spin fluctuations at $Q_1 = (0.4,0,0)$ can account for the loss of entropy at the HO transition [7]. It has more recently been shown that the spin excitations are gapped over a wide range of incommensurate wave vectors that follow the Brillouin zone edges [26]. Given that the specific heat and the volume thermal expansion coefficient are both certain derivatives of the entropy, the gap inferred here is probably closely related to the gapping of the incommensurate spin excitations. Therefore, a detailed study of the spin excitation gap for this system using inelastic neutron scattering will potentially lead to similar results as ours and may yield insight into the nature of the HO phase.

As noted above, the transition temperature T_0 and even the existence of HO below T_0 = 10 K are uncertain. We could not rule out the possibility that the HO transition becomes first order and drops to $T_0 = 0$ K discontinuously at a certain concentration lower than x=0.12, which would be an interesting scenario as well. In the case of a continuous suppression of the HO phase to 0 K, a QCP is expected to exist, as shown in Figure 6. On the other hand, if the HO transition temperature T_0 drops to 0 K discontinuously at a first order transition, there would be no QCP. Instead, one would expect a tri-critical point, a wing-shaped structure, and quantum critical end points under a magnetic field, as has been observed for a first order ferromagnetic quantum phase transition [27].

In order to shed light on these two different scenarios, we searched for evidence of the existence of a QCP using volume thermal expansion and specific heat measurements. In previous studies, NFL behaviour has been observed in both specific heat and electrical resistivity measurements for a large range of Re concentrations, deep into the ferromagnetic region [8–10]. For example, the logarithmic term in specific heat persisting to x=0.35 has been related to NFL behaviour [8]. Strictly speaking, the observed NFL behaviour suggests the presence of strong quantum fluctuations, but do not directly imply that these fluctuations become critical. An interesting and useful approach for obtaining direct evidence for quantum critical fluctuations is to measure the Grüneisen ratio, which will diverge at the QCP [28]. [The Grüneisen ratio $\Gamma(T) = \beta(T)/C_e(T)$.] We performed measurements of the volume thermal expansion coefficient and specific heat down to 0.4 K and 0.5 K, respectively, on the x=0.12 sample to determine the Grüneisen ratio, the results of which are presented in Figure 7. We performed similar measurements on the x=0.15 sample as well. However, the low-temperature thermal expansion data have a significant amount of noise, making it difficult to extract useful information. Therefore, we have not presented the low temperature data on the x=0.15 sample.

For x=0.12, $C(T)/T$ diverges with decreasing T and can be described by a function with either a logarithmic or power law T-dependence. The power

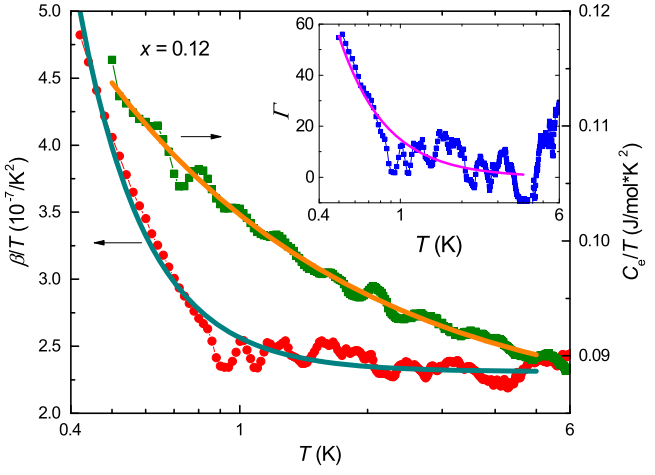

Figure 7. (Color online) Low temperature volume thermal expansion coefficient divided by temperature, β/T, and low temperature electronic specific heat divided by temperature, C_e/T, for x=0.12. The Grüneisen ratio Γ, estimated from the volume thermal expansion coefficient and electronic specific heat, is shown in the inset. The solid lines are power law fits.

law fit yields an exponent n=−0.7. Similarly, $\beta(T)/T$ also increases with decreasing T at low temperatures. However, $\beta(T)/T$ diverges faster than $C(T)/T$. A fit of the $\beta(T)/T$ data with a power law in T within the range 0.4–5 K yields an exponent n=−2.7. As a result, it is clear that the Grüneisen ratio diverges at low temperatures, providing strong evidence for the existence of a QCP in the vicinity of x=0.12. This is consistent with the scenario of a continuous suppression of the HO phase. It is worth noting that the quantum critical fluctuations are mainly along the c-axis with quasi-one-dimensional character, as can be seen in Figure 8. This one-dimensional character in the diverging Grüneisen ratio is consistent with the anisotropy of the uranium spins [6, 29].

4. Concluding remarks

To summarise, we investigated the energy gap associated with the HO phase in the $URu_{2-x}Re_xSi_2$ system by means of thermal expansion coefficient and specific heat measurements. As the HO transition is suppressed to lower temperature, the ratio between the HO energy gap and transition temperature decreases three-fold, indicating a change in the coupling strength. The fast suppression of the energy gap potentially leads to a scenario of a gapless HO state, in which the energy gap vanishes before the transition temperature is suppressed to zero. To test this hypothesis, we investigated the Grüneisen ratio in the vicinity of the concentration where the HO is suppressed. The divergence of the Grüneisen ratio at x=0.12 provides evidence for the existence of a QCP, consistent with our hypothesis.

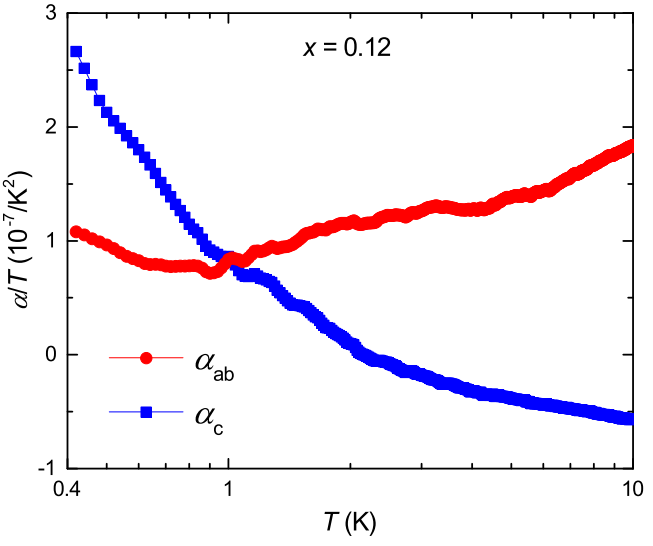

Figure 8. (Color online) Low temperature linear thermal expansion coefficient divided by temperature, α/T, within the *ab* plan and along the *c* axis, for x=0.12.

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Disclosure statement

No potential conflict of interest was reported by the authors.

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