

Quantum tricritical fluctuations driving mass enhancement and reentrant superconductivity in URhGe

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Abstract.

The field-induced reentrant superconductivity (RSC) discovered near a quantum critical point (QCP) in a ferromagnetic superconductor URhGe highlights the close interplay between superconductivity and magnetism. While the origin of the RSC is broadly thought to be associated with quantum critical fluctuations, their exact nature had not been well identified. Here we review our recent ⁵⁹Co NMR study in a single crystal of URh_{0.9}Co_{0.1}Ge. Our measurements of the NMR spin-spin relaxation reveal a divergence of electronic spin fluctuations in the vicinity of the field-induced QCP at $H_R \approx 13$ T. The fluctuations observed are characteristic of a tricritical point, followed by a phase bifurcation toward quantum wing-critical points. We show that these tricritical fluctuations enhance the effective mass of the conduction electrons and, further, drive the RSC near the H_R .

1. Introduction

A quantum phase transition emerges between two competing electronic ground states driven by fluctuations of quantum mechanical rather than thermal. A textbook example of magnetic-field-induced quantum phase transitions occurs in the one-dimensional Ising chain [1, 2]. At zero temperature (T), the ferromagnetic (FM) order switches to a paramagnetic state at a critical value H_{cr} of a transverse field applied perpendicular to the Ising axis. The value of H_{cr} defines the quantum critical point (QCP).

An analogous quantum phase transition induced by a transverse field occurs in the heavy-fermion compound URhGe [3]. Although the uranium (U) $5f$ electrons exhibit itinerant character, their spin moments still possess a strong Ising anisotropy along the c axis, with zigzag chains of nearest-neighbour U ions along the a axis. At zero magnetic field, U moments of $0.4 \mu_B$ are aligned ferromagnetically along the c axis below the Curie temperature $T_{Curie} = 9.5$ K [4]. A transverse field H_b applied along the b axis gradually decreases the temperature of the FM transition, and eventually aligns the U moments along b at a critical field $H_R \sim 12$ T, resulting in



a sudden jump of magnetization in a narrow field range (i.e., metamagnetism)[3, 5, 6]. Reentrant superconductivity (RSC) emerges in this critical region [3, 7]. The RSC transition temperature $T_{sc}=0.42$ K is higher than that observed at zero field (which disappears for $H > 2$ T, Fig. 1), and is broadly thought to be associated with quantum critical fluctuations possibly emerging near H_R [3, 5, 7, 8, 9, 10, 11].

In this paper, we review our recent ^{59}Co NMR study in a single crystal of $\text{URh}_{0.9}\text{Co}_{0.1}\text{Ge}$ [12] and discuss the interplay between the RSC, electron-mass enhancement and quantum critical fluctuations, all induced near H_R by applied magnetic fields along the b -axis. Since both Rh and Ge nuclei have poor NMR sensitivity, we conducted ^{59}Co NMR in URhGe doped with 10% cobalt ($\text{URh}_{0.9}\text{Co}_{0.1}\text{Ge}$). The substitution of Co for Rh is isostructural and isoelectronic, although it precludes superconductivity, it only minimally affects the magnetic properties [13, 14]: H_R (13.4 T) and T_{Curie} (11.8 K) are only slightly enhanced by the Co substitution (Fig. 1), suggesting a gradual increase of magnetic correlation [13, 14], and the spin system retains its strong Ising character along the c axis. The resistivity data are also very similar to those previously reported for $x = 0$ (see for example the inset of Fig. 1) [10, 15]. Therefore, $\text{URh}_{0.9}\text{Co}_{0.1}\text{Ge}$ appears to be a suitable proxy, allowing us indirectly to probe by NMR the microscopic nature of magnetic fluctuations in URhGe .

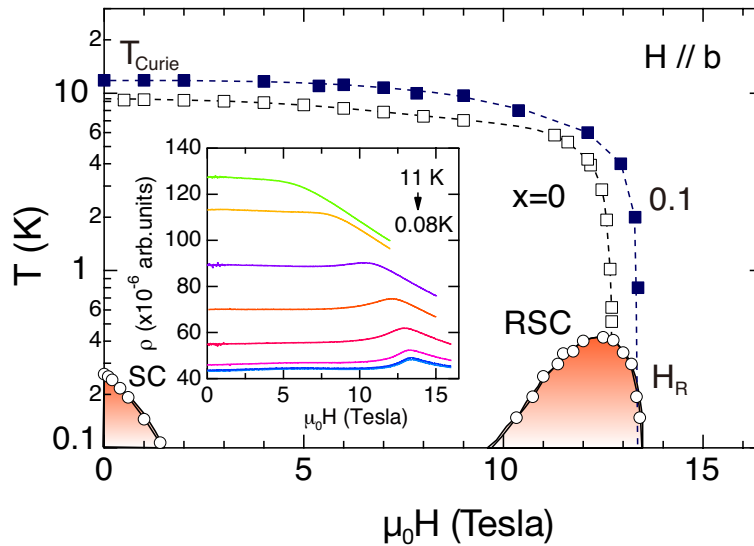


Figure 1. The temperature-field phase diagrams of $\text{URh}_{1-x}\text{Co}_x\text{Ge}$ constructed from resistivity measurement in single crystals of $x = 0$ and 0.1 [12]. The inset shows the field ($\parallel b$) dependence of resistivity $\rho(T, H)$ at different temperatures for $x = 0.1$.

2. Experimental Methods

We prepared a high quality single crystal of $\text{URh}_{0.9}\text{Co}_{0.1}\text{Ge}$ by the Czochralski pulling method [4, 10] and measured the field strength (H) and angle (θ) dependence of the spin-spin relaxation time (T_2) by using a sample rotator inside a superconducting magnet, where the θ is the angle from the b axis in the (bc) crystal plane, and the magnetic field components are thus $(H_b, H_c) = (H \cos \theta, H \sin \theta)$. ^{59}Co nuclei possess a high nuclear spin value, $I = 7/2$, and a relatively large nuclear gyromagnetic ratio, $^{59}\gamma_N = 10.03$ MHz/T, providing high efficiency of the NMR excitation pulses as well as high sensitivity of NMR detection. These advantages enabled the use of very short NMR pulses, typically around $1 \mu\text{sec}$, which was necessary to trace

the spin-echo decay with extremely short T_2 . All the NMR measurements reported here have been performed at $T = 1.6$ K, well below the T_{Curie} of 11.8 K of our crystal. The T_2 values were determined by fitting the τ dependence of the spin-echo intensity, measured on a center peak of NMR spectrum, to an exponential function, $M(2\tau) \propto \exp(-2\tau/T_2)$.

3. Results and discussion

Figure 1 show the T - H_b phase diagrams obtained for single crystals of $x = 0$ and 0.1. The transition temperature to the FM ordered state, T_{Curie} , is gradually suppressed by the magnetic field applied along the b -axis, H_b , and reaches zero at the critical field, H_R , where the U moments are aligned along the field direction. The inset of the figure shows the field dependence of the resistivity $\rho(T, H)$ for $x = 0.1$ at different temperatures. In general, magnetic transitions at high magnetic fields are not well defined in ferromagnets, because the application of the field transforms the phase transition into a broad crossover between the paramagnetic state and the FM state. However, when the field is perfectly aligned with the hard-magnetization axis in an Ising system, the magnetic transition remains clearly defined even at high fields [3, 10]. Indeed, as seen in the inset of Fig.1, the field dependence of the resistivity at low temperatures exhibits a distinct peak, defining H_R of 13.4 T in our crystal.

In URhGe ($x = 0$), the SC phase appears first below ~ 2 T, and then reappears at higher fields between ~ 8 and ~ 13.5 T with a higher $T_{\text{sc}} = 0.42$ K. On the other hand, neither the low field SC nor the RSC have been observed in substituted samples, including our crystal of URh_{0.9}Co_{0.1}Ge. This, however, does not necessarily mean a lack of electronic pairing interactions in these systems. In URhGe, in contrast to the robust FM order, SC is extremely sensitive to sample quality; it has been observed only in high quality sample with a very small ρ_0 (typically less than $30 \mu\Omega \text{ cm}$ [10]). This implies that the SC is unconventional, with the phase of the SC order parameter changing as a function of orientation; thus, it is easily destroyed when the electronic mean free path is reduced to a value comparable to the superconductor's coherence length [4]. We can thus naturally expect that a strong reduction of electronic coherence brings a strong suppression of T_{sc} in the substituted systems. The SC transition would thus likely appear even in doped crystals if the electronic mean free path could be made long enough.

Figure 2 (a) shows the field dependence of $1/T_2$ for $\theta = 0^\circ$ and 5° . Here the $1/T_2$ reflects the magnitude of longitudinal component (parallel to the applied field) of slow spin fluctuations near zero frequency ($\omega \sim 0$) [12]. As seen in the figure, $1/T_2$ for $\theta = 0^\circ$ ($H \parallel b$) shows a strong field dependence associated with the divergence of magnetic fluctuations near the H_R . The magnetic fluctuations become so strong near H_R that we lose the NMR spin-echo signal between 11.4 and 14 T, since for T_2 values smaller than $3 \mu\text{s}$ the NMR signal falls within the dead time of the spectrometer. On the other hand, this divergence of $1/T_2$ near H_R is rapidly suppressed by rotating the field (θ) from the b -axis. The field dependence of $1/T_2$ then changes to a crossover-like behavior for $\theta > 5^\circ$, where a broad maximum appears at 15.5 T, higher than that for $H_R(\theta = 0) = 13.4$ T. The behavior is connected with a phase bifurcation above the H_R , as see later.

In URhGe, the effective mass of the conduction electrons m^* has been reported to be enhanced in a relatively wide field range near H_R , which was naively thought to be induced by critical fluctuations [10, 15]. This is now confirmed directly from our NMR. In Fig.2(b), we plot the field strength and angle dependences of the A coefficient of the resistivity, which were extracted by fitting low temperature resistivity data to $\rho(H, T) = A(H)T^2 + \rho_0(H)$ in URhGe [15]. According to the Kadowaki-Woods ratio, the A coefficient has a relation with the effective mass m^* , namely $A \propto \gamma^2 \propto m^{*2}$. At $\theta = 0^\circ$ ($H \parallel b$), the A coefficient starts to increase above ~ 8 T and shows a clear maximum just before H_R , and then suppressed with further increasing field. By tilting the field away from the b -axis, the peak structure is rapidly suppressed and broaden, and shifts to higher fields above H_R . These characteristic behaviors of the A coefficient, and thus of the m^* ,

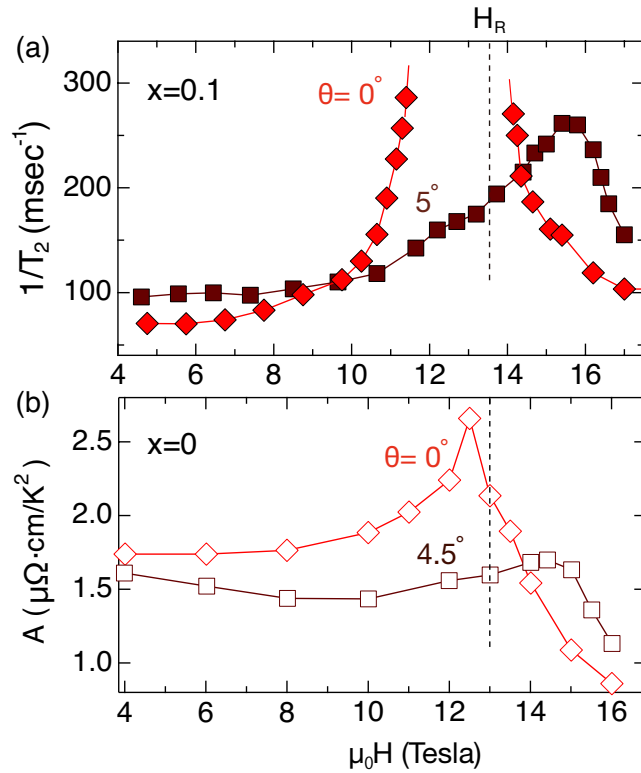


Figure 2. The field strength and angle dependences of (a) $1/T_2$ and (b) the A coefficient in $\text{URh}_{1-x}\text{Co}_x\text{Ge}$. The A coefficient was extracted by fitting low temperature resistivity data to $\rho(H, T) = A(H)T^2 + \rho_0(H)$ [15]. The field axis of the figures is scaled at the H_R of each sample.

are well understood in terms of the field dependent FM fluctuations, for both the strength and angle dependences, as seen in Fig. 2(a) and (b). This definitely supports the mass enhancement mechanism mediated by the critical fluctuations.

Next we see how the critical fluctuations are developed in the proximity of H_R . The contour plot of the $1/T_2$ in Fig. 3(a) represents the map of strength distribution of longitudinal FM fluctuations in the H_b - H_c fields plane. The fluctuations are enhanced most strongly around H_R , where the NMR signal becomes undetectable due to the extremely short T_2 (hatched grey area). We can also see the development of another weak singularity around $(H_b, H_c) \simeq (15.4, \pm 1.3)$ [Tesla], providing a clear signature of the phase bifurcation above H_R . These characteristics of quantum fluctuations are all connected with the tricritical nature of the phase transition, as discussed below.

In the transverse Ising system considered here, the order parameter is the magnitude of FM moment along the Ising axis, $\langle M_c \rangle$. The H_b is a tuning parameter of the quantum fluctuations driving the phase transition itself, while the H_c adds a perturbation conjugate to the order parameter. Then two types of magnetic phase diagram can be expected in the H_b - H_c plane, depending on the criticality. If the long-range FM order is continuously connected to the field-induced paramagnetic state, the H_b - H_c phase diagram simply involves an ordinary QCP at $T = 0$. This ordinary QCP induces solely the diverging susceptibility of the order parameter, i.e. the divergence of the transverse fluctuations along the c axis. On the other hand, if the system possesses a tricritical point (TCP), which prevents the continuous connection of the long-range order to the disordered state, one can generally expect the emergence of the phase bifurcation above the TCP [16], where the first order planar surface at $H_c = 0$ into two wings that continue away from the H_b -axis and end up at quantum wing-critical points (QWCPs) at zero temperature for a finite H_c . The $1/T_2$ behavior clearly supports this latter case. Another characteristic features of a TCP, with regard to magnetic excitations, is that it induces a diverging susceptibility not only for the order parameter but also for the physical

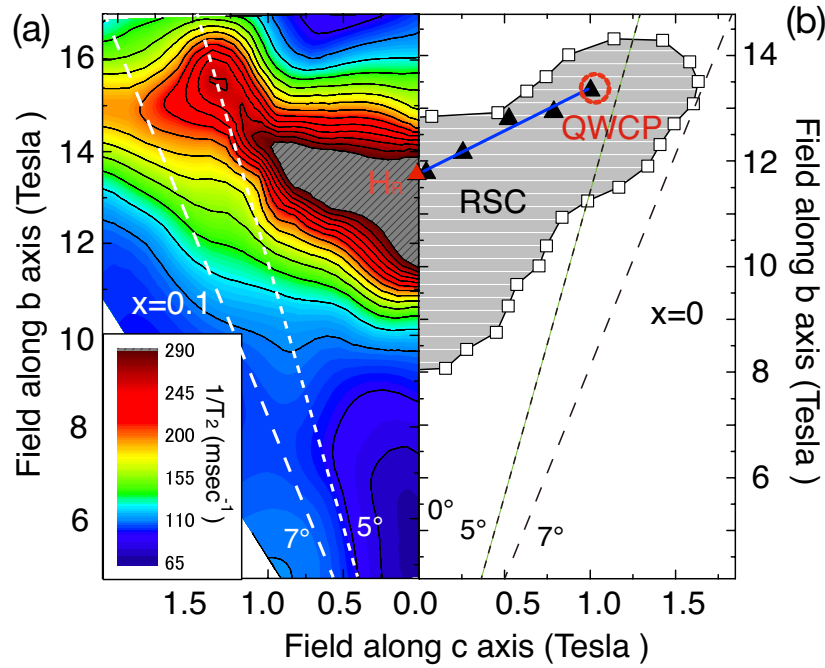


Figure 3. (a) The map of the strength distribution of magnetic fluctuations in the H_b - H_c fields plane around H_R . This contour plot of the $1/T_2$ at 1.6 K is constructed from the 104 data points in total measured with different combinations of H and θ , between $H=4.7$ and 17 T and between $\theta = 0^\circ$ and 11° , respectively [12]. (b) The phase diagram constructed from resistivity measurements by Lévy *et al.* in a single crystal of URhGe[9]. The open squares indicate the region where the RSC appears at $T \sim 40$ mK and the filled triangles indicates positions where first-order-like metamagnetic transitions occurs at $T \sim 500$ mK). Since their crystal had a different H_R value (12 T) from that of our crystal (13.4 T), the field axes are scaled to the H_R .

quantity conjugate to the tuning parameter driving the phase transition [17, 18]. Here the tuning parameter is H_b , and thus we can expect the divergence of the longitudinal ($\parallel b$) component of magnetic fluctuations, as observed experimentally.

Figure 3 further reveals the close interplay between the FM fluctuations and the RSC. In Fig. 3(b), we plot the H_b - H_c phase diagram constructed from resistivity measurements by Lévy *et al.* in a single crystal of URhGe[9]. The open squares indicate the region where the RSC has been observed at $T \sim 40$ mK and the filled triangles indicate the positions where first-order-like magnetic transitions have been observed at $T \sim 500$ mK. We can see that the RSC appears almost in the same limited region as that where we have observed the critical fluctuations on the H_b - H_c plane. Only about $\theta=7^\circ$ misorientation from the b -axis suppresses the RSC[9, 19], and as seen in Fig. 3 (a), such suppression is now definitely connected to the rapid decline of the critical fluctuations for $\theta > 7^\circ$. The RSC also exhibits the highest T_{sc} and the largest diamagnetic signal around H_R (not around the QWCPs), where we have detected the diverging fluctuations [3, 6]. These close relationships between the FM fluctuations and the RSC strongly suggests these quantum fluctuations as the pairing glue responsible for the RSC.

Finally, we note that, although we revealed a divergence of the longitudinal component ($H \parallel b$) of slow fluctuations ($\omega \sim 0$) near the TCP, this does not necessarily mean the absence of a similar divergence for the transverse component ($H \parallel c$) at $\omega \sim 0$. The NMR relaxation measurements, both $1/T_1$ and $1/T_2$, can not probe such fluctuations (which can probe the transverse fluctuations

at the NMR frequency of ~ 0.1 GHz), and we should recall that FM critical fluctuations may have strong enhance of fluctuations near zero frequency [12]. A divergence of the fluctuations is expected, in principle, for both the longitudinal and transverse components near the TCP, and we suggest that such tricritical nature of the fluctuations should be fully involved in understanding the mechanism of the RSC [20].

4. Summary

The experiments reported here indicates that URhGe possesses quantum tricriticality, associated with its low temperature TCP [7, 8], and hence develops strong longitudinal fluctuations perpendicular to the Ising axis (parallel to the applied field) at low temperatures. Such tricritical nature of the phase transition has already been proposed for URhGe; however, it was mostly relying on the observation of first-order-like behaviors near H_R [7, 8, 21, 22]. The present NMR go further, since they provide evidence by probing directly the nature of the associated fluctuations. We show that these tricritical fluctuations enhance the effective mass of the conduction electrons and, further, drive the RSC, most probably by reinforcing the SC pairing [11, 20, 23, 24], and partly through the mass enhancement [10, 15].

Acknowledgments

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References

- [1] see, for example, Sachdev S. Quantum Phase Transitions 1999 Cambridge Univ. Press
- [2] Kinross A W *et al.* 2014 *Physical Review X* **4**, 031008
- [3] Lévy F, Sheikin I, Grenier B, and Huxley A 2005 *Science* **309** 1343-1346
- [4] Aoki D, *et al.* 2001 *Nature* **413** 613-616
- [5] Hardy F, *et al.*, 2011 *Phys. Rev. B* **83** 195107
- [6] Aoki D and Flouquet J 2014 *J. Phys. Soc. Jpn.* **83** 061011
- [7] Lévy F, Sheikin I, and Huxley A, *Nature Phys.* **3** 460-463
- [8] Huxley A *et al.* 2007 *J. Phys. Soc. Jpn.* **76** 051011
- [9] Lévy F *et al.*, 2009 *J. Phys. Condens. Matter* **21** 164211
- [10] Miyake A, Aoki D, and Flouquet J 2008 *J. Phys. Soc. Jpn.* **77** 094709
- [11] Hattori K and Tsunetsugu H, 2013 *Phys. Rev. B* **87** 064501
- [12] Tokunaga Y *et al.* 2015 *Phys. Rev. Lett.* **114** 216401
- [13] Sakarya S *et al.* 2008 *J. Alloys and Comp.* **457** 51-56
- [14] Huy N T, and de Visser A 2009 *Solid State Comm.* **149** 703-706
- [15] Aoki D *et al.* 2011 *J. Phys. Soc. Jpn.* **80** SA008
- [16] Kirkpartrick T R and Belitz D 2015 *Phys. Rev. Lett.* **115** 020402
- [17] Misawa T, Yamaji Y and Imada M 2006 *J. Phys. Soc. Jpn.* **75** 064705
- [18] Misawa T, Yamaji Y and Imada M 2008 *J. Phys. Soc. Jpn.* **77** 093712
- [19] Yelland E A, *et al.* 2011 *Nat. Phys.* **7** 890-894
- [20] Mineev V P 2015 *Phys. Rev. B* **91** 014506
- [21] Aoki D, Knebel G, and Flouquet J, 2014 *J. Phys. Soc. Jpn.* **83** 094719
- [22] Kotegawa H *et al.* 2015 *J. Phys. Soc. Jpn.* **84** 054710
- [23] Hattori T *et al.*, 2012 *Phys. Rev. Lett.* **108** 066403
- [24] Tada Y *et al.* 2013 *J. Phys.: Conf. Ser.* **449** 012029