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Interplay between quantum fluctuations and reentrant superconductivity with a highly enhanced upper critical field in URhGe

Y. Tokunaga,[1,*] D. Aoki,[2,3] H. Mayaffre,[4] S. Krämer,[4] M.-H. Julien,[4] C. Berthier,[4] M. Horvatić,[4] H. Sakai,[1] T. Hattori,[1] S. Kambe,[1] and S. Araki[2,5]

[1]*ASRC, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan*
[2]*INAC/SPSMS, CEA-Grenoble/UJF, 38054 Grenoble, France*
[3]*IMR, Tohoku University, Ibaraki 311-1313, Japan*
[4]*LNCMI-CNRS (UPR 3228), EMFL, UGA, UPS, INSA, 38042 Grenoble, France*
[5]*Department of Physics, Okayama University, Okayama 700-8530, Japan*

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The ferromagnetic superconductor URhGe has been known to exhibit an extremely large enhancement of the upper critical field when the field is confined and rotated in the (*ab*) crystal plane. Our high-field ^{59}Co NMR measurements on 10% Co-doped URhGe up to 30 T prove that this unconventional behavior of the superconductivity (SC) is associated with a strong anisotropy of field-dependent quantum fluctuations near a tricritical point. These fluctuations are rapidly suppressed by the field component along the *c* axis, while being unaffected by any field component along the *a* axis. The observed close interplay between the SC and quantum fluctuations strongly supports a pairing mechanism mediated by these fluctuations in URhGe.

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Spin fluctuations near a ferromagnetic (FM) quantum critical point (QCP) have been supposed to create a binding force between quasiparticles with equal (triplet) spin pairs [1,2], analogous to the mechanism of a superfluid pairing in ^{3}He [3]. Such an unconventional spin-triplet superconductivity (SC), mediated by FM fluctuations, is now considered to be realized in a family of uranium (U) based compounds, UGe_2 [4], URhGe [5], and UCoGe [6]. Up to now, these are the only fully established examples of FM superconductors in which uniform SC exists deep inside the FM state. A common and characteristic feature of these FM superconductors is that they exhibit very large upper critical fields, by far exceeding the ordinary Pauli paramagnetic limit ($= 1.84 k_B T_{sc}/\mu_B$), as expected for triplet SC [7–11]. For URhGe, in particular, the upper critical field exceeds 28 T, although T_{sc} is less than 0.42 K. Such a large enhancement of the upper critical field, however, appears only when the field is rotated in the (*ab*) plane of hard magnetization; it characterizes the unconventional reentrant SC (RSC) phenomenon near a FM QCP [10], implying a strong interplay with magnetism.

At zero field, URhGe undergoes a ferromagnetic transition at $T_{Curie} = 9.5$ K. Below this temperature, U $5f$ moments ($\sim 0.4\mu_B$) are aligned ferromagnetically along the crystallographic *c* axis, and SC appears, coexisting with FM order, below $T_{sc} \sim 0.25$ K [5]. A magnetic field applied along the *b* axis (H_b) first suppresses the SC around 2 T, and then induces the RSC between 8 and 14 T with a maximum T_{sc} of 0.42 K, higher than at zero field [8]. Interestingly, at a similar field of $H_R \approx 12$ T, the FM moments are forced into alignment along the field direction ($\|b$); the transition is thus reminiscent of the textbook example of a quantum phase transition in a transverse Ising chain [12,13]. The RSC emerges in the vicinity of H_R.

Recently, ^{59}Co NMR performed on 10% Co-doped URhGe ($URh_{0.9}Co_{0.1}Ge$) has given evidence for diverging magnetic fluctuations for fields around H_R in the (*bc*) plane [14]. The observed fluctuations were characteristic of a tricritical point (TCP), showing a phase bifurcation towards two quantum wing-critical points (QWCPs) [15,16]. In addition, they have been found to be present in the same limited region around H_R in the (*bc*) plane as that where RSC has been observed [8,17]. These results thus suggest that these fluctuations are responsible for the RSC. Here, we performed the ^{59}Co NMR study at very high fields up to 30 T in the (*ab*) plane. The experiments reveal that the critical fluctuations are unaffected by any additional field component applied along the hard magnetic *a* axis (H_a), proving that the only tuning parameter of quantum fluctuations is H_b. This finding provides the key to understanding the peculiar phase diagram of the RSC, including the extremely large enhancement of the upper critical field in the (*ab*) plane, and gives further evidence that the RSC mechanism is mediated by the magnetic quantum fluctuations in this system.

Preparation of a high quality single crystal of $URh_{0.9}Co_{0.1}Ge$ was carried out by using the Czochralski pulling method [5,18]. The substitution of Co for Rh is isostructural and isoelectronic, and hence it only minimally affects the magnetic properties [19,20]. Although a strong reduction of electronic coherence precludes a SC transition, $URh_{0.9}Co_{0.1}Ge$ can be considered as a suitable proxy to investigate with ^{59}Co NMR the nature of magnetic fluctuations in URhGe [14]. The field dependence of the resistivity at low temperatures exhibits a distinct peak at $H_R(\|b) = 13.4$ T, which is slightly higher than that in URhGe. The T^2 coefficient A and the residual resistivity ρ_0, extracted by fitting the resistivity data to $\rho(H,T) = A(H)T^2 + \rho_0(H)$, also have a maximum at $H = H_R(\|b)$ [14]. ^{59}Co nuclei possess a high nuclear spin value ($I = 7/2$) with a relatively large nuclear gyromagnetic ratio ($\gamma_N = 10.054$ MHz/T [21]), providing high efficiency of the NMR excitation pulses as well as high sensitivity of NMR detection. These advantages enabled us to use very short NMR pulses (~ 1 μs), which was necessary in

*tokunaga.yo@jaea.go.jp

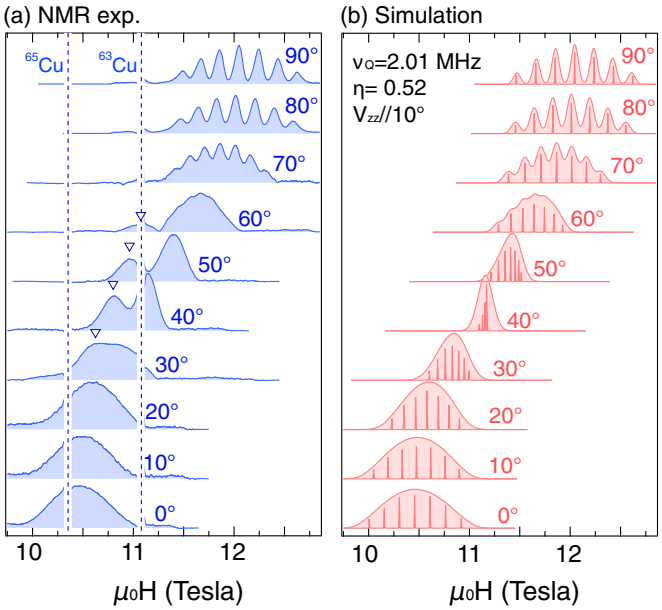

FIG. 1. (a) The θ_{ab} dependence of ^{59}Co NMR spectra recorded with a NMR frequency of 125.45 MHz at 1.6 K. (b) Numerical simulations, based on diagonalization of the total Hamiltonian consisting of the Zeeman term and a quadrupolar term, which well reproduce the overall θ_{ab} dependence of the spectra, except for a small satellite peak observed in the interval $\theta_{ab} = 30°$–$60°$ (inverted triangles). Here, the calculated bare NMR spectrum is convoluted with a Gaussian broadening function having a width of $\Delta \sim 0.05$ T for $H \| a$, and ~0.15 T for $H \| b$, respectively. The major source of Δ is magnetic, i.e., a distribution of the Knight shift values, since all seven NMR lines have a similar width.

the present experiments to measure extremely short transverse relaxation times T_2.

The field strength (H) and angle (θ_{ab}) dependences of the NMR spectrum and T_2 were measured at a temperature of $T = 1.6$ K, well below $T_{\rm Curie} = 11.8$ K for our crystal. Here, θ_{ab} is the angle in the (ab) plane relative to the b axis; thus, $(H_b, H_a) = (H\cos\theta_{ab}, H\sin\theta_{ab})$. The high-field data above 17 T were obtained using a 20 MW resistive magnet at the LNCMI-Grenoble. ^{59}Co NMR spectra were recorded at fixed frequency by sweeping the field in equally spaced steps and by summing the Fourier transforms of the spin-echo signals. T_2 values were determined by fitting the τ (pulse separation) dependence of the spin-echo intensity, measured at the center of the NMR spectrum, to an exponential function, $M(2\tau) \propto \exp(-2\tau/T_2)$.

Figure 1(a) shows the θ_{ab} dependence of the field swept Co NMR spectrum. For $H \| a$ ($\theta_{ab} = 90°$), the spectrum splits into seven sharp peaks, owing to the quadrupolar interaction between the nuclear quadruple moment (spin $\frac{7}{2}$) and the electric field gradient (EFG). For $H \| b$ ($\theta_{ab} = 0°$), on the other hand, the seven peaks merge into a single, broad peak, because of a significant broadening of each NMR line. The overall θ_{ab} dependence is also well reproduced by numerical simulations of the NMR spectra, except for a small satellite peak observed in the interval $\theta_{ab} = 30°$–$60°$ [Fig. 1(b)]. The numerical simulations are based on diagonalization of the total Hamiltonian consisting of the Zeeman term and the

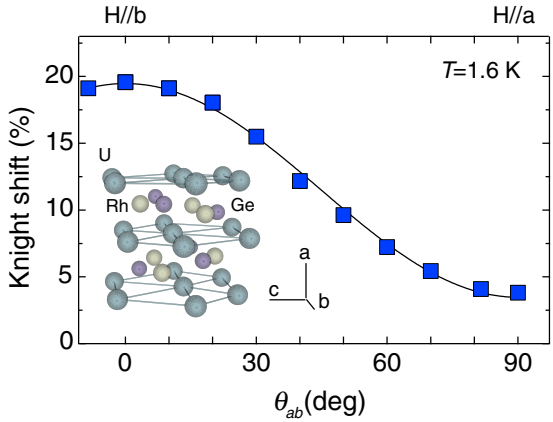

FIG. 2. Angular dependence of the Knight shift $K(\theta_{ab})$ in the (ab) plane, extracted from the numerical simulations shown in Fig. 1(b). The solid line is the fit to Eq. (2) (see the text). The inset shows the TiNiSi-type crystal structure of URhGe.

quadrupolar term,

$$\begin{aligned}\mathcal{H} &= \mathcal{H}_z + \mathcal{H}_q \\ &= -\gamma_N\hbar(1+\boldsymbol{K})\boldsymbol{I}\cdot\boldsymbol{H} \\ &\quad + \frac{\hbar\nu_Q}{6}\left\{(3I_Z^2 - \boldsymbol{I}^2) + \frac{1}{2}\eta(I_+^2 + I_-^2)\right\}. \end{aligned} \tag{1}$$

Here, we use the asymmetry parameter $\eta = 0.52$ with the principal Z axis of the EFG tensor being tilted by 10° from the a axis, as found in UCoGe [22–24]. The quadrupole frequency ν_Q is fixed at 2.01 MHz, while the Knight shift K is extracted as a fitting parameter at each angle.

In Fig. 2 we plot the θ_{ab} dependences of the Knight shift $K(\theta_{ab})$. $K(\theta_{ab})$ is associated with magnetizations in the (ab) plane as

$$K(\theta_{ab})H = A_a M_a \sin\theta_{ab} + A_b M_b \cos\theta_{ab}, \tag{2}$$

where $A_i = K_i/\chi_i$ ($i = a,b$) are the principal values of the hyperfine coupling tensor, connecting the spin polarization at U sites to the hyperfine field observed at Co nuclei, and M_i is the magnetization along the i direction. When both M_a and M_b are magnetic-field-induced magnetization ($M_a = M_b = 0$ at $H \to 0$, namely, the FM moments are aligned along the c axis at zero field [8]), they should be proportional to the applied field along each axis, $M_a = \chi_a H\sin\theta_{ab}$ and $M_b = \chi_b H\cos\theta_{ab}$. Then, Eq. (2) is reduced to

$$K(\theta_{ab}) = K_a \sin^2\theta_{ab} + K_b\cos^2\theta_{ab}. \tag{3}$$

As shown by a solid line in Fig. 2, this equation fully accounts for the data with $K_a = 2.9\%$ and $K_b = 19.6\%$. The large ratio K_b/K_a confirms that the magnetic susceptibility is very anisotropic even in the ab plane ($\chi_c > \chi_b > \chi_a$) [25].

Now we discuss the field-angle dependence of magnetic fluctuations measured by $1/T_2$. In Figs. 3(a) and 3(b), we compare $1/T_2$ data obtained in the (bc) and (ab) planes, where $1/T_2$ values reflect the magnitude of the longitudinal component (parallel to the applied field) of slow spin fluctuations near zero frequency ($\omega \sim 0$). That is, $1/T_2 \propto G_\parallel(0)$, where $G_\alpha(\omega) = \int_{-\infty}^{\infty} < h_\alpha(t)h_\alpha(0) > \exp(i\omega t)dt$ is the spectral density of the fluctuating hyperfine field [14]. For $\theta_{ab} = 0°(H \| b)$, $1/T_2$

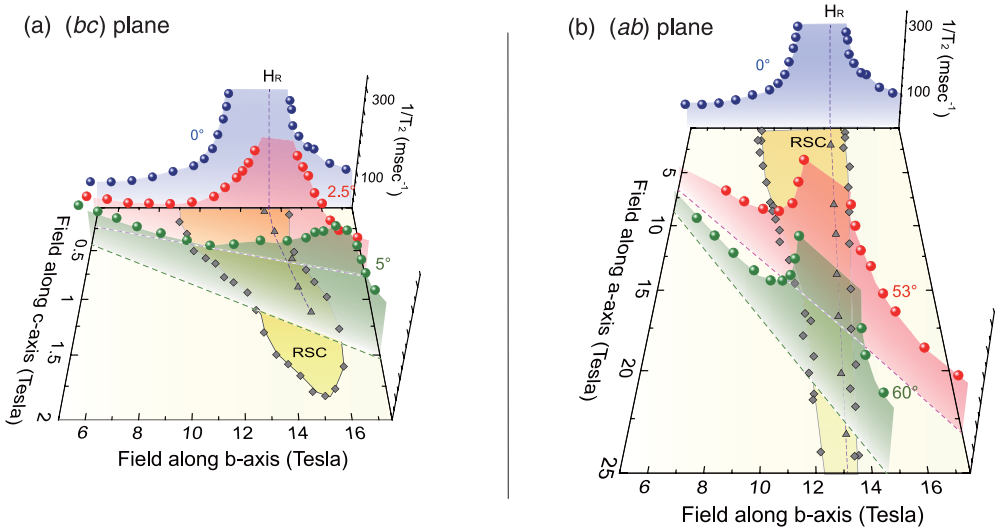

FIG. 3. The field and angle dependences of $1/T_2$ in the (a) (bc) [14] and (b) (ab) planes. The solid diamonds indicate the region where RSC has been observed from resistivity measurements at $T \sim 40$ mK in a single crystal of URhGe [17]. The solid triangles indicate the first-order-like metamagnetic transitions at $T \sim 500$ mK [17]. Since URhGe has a lower H_R (12 T) than $URh_{0.9}Co_{0.1}Ge$ (13.4 T), the data for URhGe are plotted here with field values scaled proportionally to the H_R values in $URh_{0.9}Co_{0.1}Ge$.

diverges strongly around $H_R(\|b) = 13.4$ T. In the (bc) plane, however, these critical fluctuations are suppressed rapidly by rotating the field (i.e., by adding H_c) [Fig. 3(a)]. A rotation by only 5° away from the b axis changes the divergence to a broad maximum, with the peak position shifted to $H_b \sim 15.4$ T.

In the (ab) plane, on the other hand, the critical fluctuations are never suppressed by the field rotation [Fig. 3(b)]. Regardless of the θ_{ab} angle of the applied field, the diverging fluctuations always appear near a fixed value of the field component along the b axis, that is, $H_b \sim H_R(0°) = 13.4$ T. This means that, as regards the total value of H, the critical field increases with increasing θ_{ab}, e.g., $H_R(\theta_{ab} = 53°) \sim 21$ T and $H_R(\theta_{ab} = 60°) \sim 25.5$ T. The critical fluctuations are thus suggested to be determined only by the magnitude of H_b. This can be seen more precisely by plotting $1/T_2$ vs $H_b = H \cos\theta_{ab}$ (Fig. 4), where all the data essentially collapse onto a single curve in the region where $H < H_R$. This result indicates that only the H_b field component is a tuning parameter for quantum fluctuations driving the system to a critical point, while H_a has practically no effect on the quantum fluctuations in the (ab) plane. The robustness of the quantum fluctuations against H_a suggests that they are developed in the (bc) plane, that is, perpendicular to the a axis. This is supported by the fact that the fluctuations are suppressed quickly by the application of H_c.

The transition at H_R thus approximates most simply to that of a transverse Ising chain [12,26], whose Hamiltonian is given by

$$\mathcal{H} = -J \sum_i \left(S_i^z S_{i+1}^z + h_x S_i^x \right), \tag{4}$$

where J (>0 for FM) is an exchange interaction between the nearest-neighbor spins and a field h_x is applied perpendicular to the Ising axis (z). The competition between J and h_x stimulates quantum fluctuations of S^z and leads the system to a QCP between FM ordering and a field-induced paramagnetic state along h_x. In an isolated one-dimensional (1D) Ising chain, the long-range FM order is expected only at $T = 0$, while additional 3D interchain couplings stabilize it even at $T > 0$. Similarly, in URhGe, the order parameter is the magnitude of FM moment, and H_b corresponds to h_x. H_b tunes the quantum fluctuations and finally, above H_R, aligns the FM moment along the b axis. On the other hand, H_c provides an additional perturbation conjugate to the order parameter, leading the system away from the critical point by stabilizing long-range FM order.

As regards magnetic excitations (magnons), if the transition were occurring at a QCP as in the case of the ordinary

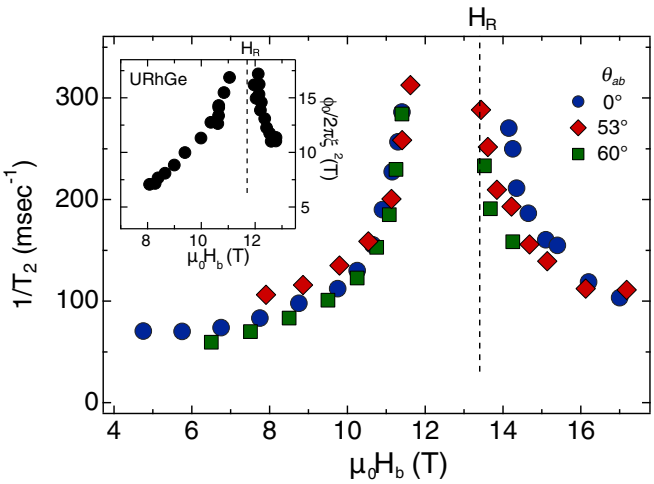

FIG. 4. The $1/T_2$ data in the (ab) plane are plotted against $H_b = H \cos\theta_{ab}$. The inset shows the H_b dependence of the geometric average of the coherence length, evaluated from the resistivity data in several different θ_{ab}, assuming constant anisotropy, in URhGe [10]. The quantity plotted is proportional to the inverse-square coherence length $\phi_0/2\pi\xi^2$, given in units of magnetic field.

transverse Ising chain, we would expect solely a diverging susceptibility of the order parameter, which in this instance would be a divergence of the fluctuations along the c axis [26]. However, URhGe involves a TCP [10,14,27–29], likely associated with the itinerant character of its ferromagnetism [30–32]. In that case, the susceptibility diverges not only for the order parameter but also for the physical quantity conjugate to the tuning parameter, H_b [33–35]. We can thus expect diverging fluctuations for both the b- and c-axis components, as was observed experimentally [14].

In the (ab) plane, on the other hand, one might expect quantum fluctuations driven by H_a [i.e., by adding a term $-h_y \sum_i S_i^y$ to Eq. (5)]. However, in our $1/T_2$ data we do not observe any additional enhancement of fluctuations by H_a up to 25 T, suggesting that the QCP or TCP (if any) along the a axis would only exist at very high fields. This is naturally expected in URhGe, since the a axis is magnetically much harder than the b axis; the slope of $M(H)$ for $H \| a$ is about five times smaller than that for $H \| b$ [36]. Such strong magnetic anisotropy (and magnetic excitations) appears to be inherent in the crystal structure. That is, the TiNiSi-type structure of this compound can be considered as an orthorhombically deformed hexagonal AlB_2-type structure, with the a axis of the TiNiSi lattice corresponding to the hexagonal c axis of the AlB_2 lattice (see the inset to Fig. 2). U atoms form zigzag chains extended along this a axis, thus breaking the local inversion symmetry at each U site [37,38]. All this makes the a axis inherently distinct from the other two crystal axes.

Next we focus on the interplay between the quantum fluctuations and RSC. In Fig. 3 we have plotted the region where RSC has been observed in resistivity measurements by Lévy *et al.* in a single crystal of URhGe [8,10,17]. We can see that while RSC is suppressed rapidly with a small field $H_c \sim 1.8$ T in the (bc) plane [Fig. 3(a)], in the (ab) plane it survives in a large region of fields with a large value of H_a in the presence of a practically constant value of $H_b \simeq H_R(\|b)$ [Fig. 3(b)]. The latter situation corresponds to an extremely large enhancement of the upper (total) critical field, to above 28 T when $\theta_{ab} \gtrsim 60^\circ$ [10]. In particular, our present NMR results reveal that only field rotation in the (ab) plane preserves the critical fluctuations around $H_b \simeq H_R(\|b)$ [Fig. 3(b)]. This provides a necessary condition for the RSC to survive up to high fields with a large H_a component in the (ab) plane, where $H = \sqrt{H_b^2 + H_a^2}$. On the other hand, in the (bc) plane the critical fluctuations are suppressed quickly by H_c, hence the RSC disappears in $H_c > 1.7$ T [Fig. 3(a)]. This close overlap between RSC and quantum fluctuations provide firm evidence that the pairing mechanism of SC is mediated by these fluctuations.

Finally, we consider how critical fluctuations induce the RSC. In the inset to Fig. 4 we show the field dependence of the geometric-average coherence length $1/\xi^2$ evaluated by Lévy *et al.* from resistivity data for the (ab) plane [10]. Remarkably, $1/\xi^2$ exhibits a very similar field dependence to that of $1/T_2$; it diverges around H_R and is only scaled by H_b [10]. This similarity suggests that the critical fluctuations drive the RSC by contributing to the enhancement of $1/\xi^2$. The critical fluctuations could enhance $1/\xi^2$ through the enhancement of the pairing mechanism [10,26,35] and/or of the effective mass of the conduction electrons [18,39]. We also remark that, when the fluctuations develop within the (bc) plane as observed here, the gap structure of the RSC is naturally expected to have p-wave symmetry with the antinode along the a axis, as discussed by Mineev [40] and Huxley *et al.* [28]. Indeed, this gap structure describes well the anisotropy of the temperature-dependent critical field for the superconductivity around $H = 0$ [9]; however, it has not yet been examined for the reentrant superconductivity. Further experiments will be required to confirm this point.

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