

Enhancement of superconductivity by pressure-induced critical ferromagnetic fluctuations in UCoGe

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(Received 3 August 2018; published 24 January 2019)

A ^{59}Co nuclear quadrupole resonance (NQR) was performed on a single-crystalline ferromagnetic (FM) superconductor UCoGe under pressure. The FM phase vanished at a critical pressure P_c , and the NQR spectrum just below P_c showed phase separation of the FM and paramagnetic (PM) phases below Curie temperature T_{Curie} , suggesting first-order FM quantum phase transition (QPT). We found that the internal field was absent above P_c , but the superconductivity is almost unchanged. This result suggests the existence of the nonunitary to unitary transition of the superconductivity around P_c . Nuclear spin-lattice relaxation rate $1/T_1$ showed the FM critical fluctuations around P_c , which persist above P_c and are clearly related to superconductivity in the PM phase. This FM QPT is understood to be a weak first order with critical fluctuations. $1/T_1$ sharply decreased in the superconducting (SC) state above P_c with a single component, in contrast to the two-component $1/T_1$ in the FM SC state, indicating that the inhomogeneous SC state is a characteristic feature of the FM SC state in UCoGe.

DOI: [10.1103/PhysRevB.99.020506](https://doi.org/10.1103/PhysRevB.99.020506)

Ferromagnetism and superconductivity have been considered to compete and mutually suppress one another [1], and the coexistence has been only reported in some compounds so far, where two phenomena arise from different atoms or sites [2–4]. However, such a generally accepted notion was forced to change after the discovery of superconductivity in a series of uranium (U)-based ferromagnets, namely, UGe_2 [5], URhGe [6], and UCoGe [7]. The superconducting (SC) phase in these compounds is embedded inside the ferromagnetic (FM) phase, and spin-triplet pairing is highly anticipated. Our ^{59}Co nuclear-quadrupole-resonance (NQR) measurements showed that ferromagnetism and superconductivity in UCoGe coexist microscopically [8], and that the U site is an origin of two phenomena [9]. One of the attractive features of these systems is that the FM quantum phase transition can be achieved experimentally with pressure or magnetic field, and thus, they are excellent systems for studying the relationship between the FM and SC phases. It was reported that the reentrant superconductivity in URhGe [10] and the robustness of superconductivity in UCoGe are related with the field-induced FM criticality [11]. The nuclear-magnetic-resonance (NMR) measurements revealed that the reentrant superconductivity in URhGe is associated with the tricritical fluctuations induced by the field [12,13]. We have shown from direction-dependent NMR measurements in UCoGe that the longitudinal critical FM fluctuations, which are regarded

as amplitude modes of magnons, play an essential role for superconductivity, and suggested that the FM fluctuations induce spin-triplet superconductivity with the theoretical model calculations [14–16]. This scenario, which differs from the ordinary electron-phonon coupling in the BCS model, is consistent with the theoretical work by Mineev [17], and is supported by recent thermodynamical measurements and analyses [18].

One of the remaining issues in UCoGe is an understanding of the FM criticality and its relationship with the superconductivity under pressure. UCoGe possesses a unique pressure-temperature phase diagram, in which superconductivity persists in the paramagnetic (PM) region beyond the FM criticality [19–21]. This phase diagram implies that the superconductivity is induced by the fluctuation related to a quantum critical point (QCP). In the case of antiferromagnetic (AFM) instability, SC phases are widely observed around the AFM QCPs in Ce-based heavy-fermion superconductors and iron-based superconductors. By contrast, the relationship between the FM QCP and the superconductivity is not straightforward because the first-order quantum phase transition (QPT) has been anticipated from the theoretical study of the itinerant ferromagnetism [22]. Actually, UGe_2 and URhGe exhibit first-order FM transitions by pressure or magnetic field [23,24], and the FM QCP does not exist at zero field. The first-order FM transition was reported at $P = 0$ in UCoGe [8,25], and it is necessary to examine how the FM criticality relates to the superconductivity in UCoGe.

Another important issue in UCoGe is the identification of a self-induced vortex (SIV) state in the coexisting phase. Careful magnetization and superconducting quantum

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interference device (SQUID) measurements showed that the Meissner state is absent although the Meissner-Ochsenfeld effect was observed [26,27]. We reported from the ^{59}Co NQR that the FM SC state is inhomogeneous because two nuclear relaxation components showing SC and non-SC behaviors were observed below the SC transition temperature T_{SC} , and suggested the realization of the SIV state [8]. However, we could not rule out the possibility that this inhomogeneity arises from the disorder- or impurity-induced non-SC part. It is, therefore, crucial to know whether the non-SC component disappears when the FM state is suppressed by pressure.

In this Rapid Communication, we report that the FM QPT of UCoGe is weakly first-order, and the details of the phase diagram are different from the case of a second-order QCP. We also found that the FM fluctuations are enhanced and T_{SC} increases around P_c , indicative of the positive relationship between the two phenomena. The strong FM fluctuations persist above P_c , and are likely to be responsible for the SC state in the PM side. UCoGe is also a member of the FM superconductors showing a first-order FM transition; however, the discontinuity of the magnetization at the transition is so weak that the development of the FM fluctuations is observed, which is a characteristic feature of the second-order transition. The NQR measurements above P_c suggest that the PM SC state is homogeneous, indicating that the whole part of the sample becomes superconducting. This result leads to a conclusion that the two-component nuclear relaxation in the FM SC state is not due to the disorder or impurity but is a characteristic feature in the FM superconductors.

We used a single-crystalline sample with the FM and SC transition temperatures $T_{\text{Curie}} = 2.5$ K and $T_{\text{SC}} = 0.46$ K, respectively. Details of sample preparation are described in a previous paper [14]. This sample has a residual resistivity $\rho_0 = 13 \mu\Omega\text{cm}$ along the b axis at ambient pressure [14], and the mean free path is calculated as $l \simeq 700 \text{ \AA}$ if we adopt a rough estimation used in the previous study [28]. This value is larger than the SC coherence length $\xi \simeq 120 \text{ \AA}$ [28]. Hydrostatic pressure was applied using a piston cylinder-type cell with Daphne oil 7373 as a pressure medium. Low-temperature measurements were carried out using a ^3He - ^4He dilution refrigerator down to 0.15 K. The ^{59}Co NQR was performed without applying static field. $1/T_1$ detects the FM fluctuations along the easy (c) axis, since the nuclear quantization axis at the Co site in NQR is almost parallel to the crystallographic a axis [29], and $1/T_1$ is determined with the magnetic fluctuations perpendicular to the quantization axis. The rf magnetic field H_1 was applied along the c axis, where a large signal intensity was obtained.

Figure 1 shows the temperature dependence of the ac susceptibility $\delta\chi_{\text{ac}}$ of UCoGe at several pressures measured with an NQR coil. The $\delta\chi_{\text{ac}}$ was determined by the change of the tuning of the LC circuit. T_{SC} slightly increases with increasing pressure and gets maximum at around 0.67 GPa. The SC transition width becomes sharper as pressure increases. These results are qualitatively consistent with the previous studies with bulk measurements [19–21].

Figure 2(a) shows the pressure dependence of the ^{59}Co NQR spectra at 4.2 K in the PM state. Three peaks arise from $\pm 1/2 \leftrightarrow \pm 3/2$ (ν_1), $\pm 3/2 \leftrightarrow \pm 5/2$ (ν_2), and $\pm 5/2 \leftrightarrow \pm 7/2$ (ν_3) transitions. The large asymmetric parameter $\eta \simeq 0.52$

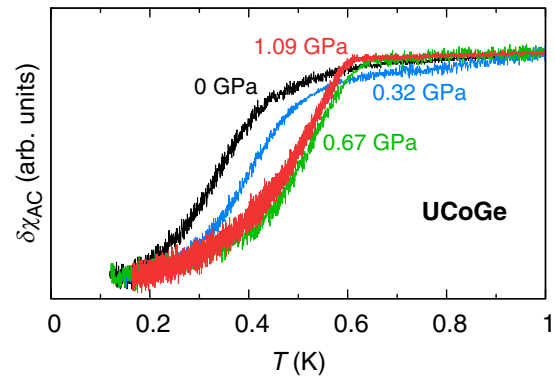


FIG. 1. ac susceptibilities of UCoGe at several pressures measured with an NMR coil at zero magnetic field. Frequencies are between 5 and 9 MHz.

makes ratios of these frequencies far from simple integers. The spectra slightly shift as pressure increases because the electric field gradient at the nuclear site changes by the lattice shrinkage. The unchanged linewidth indicates a small inhomogeneity of the applied pressure.

The temperature variation of the spectra at the ν_3 line at different pressures is shown in Figs. 2(b)–2(d). At ambient pressure, a FM signal appears below T_{Curie} because of an internal field at the nuclear site [8]. The PM signal disappears and only the FM signal was detected at sufficiently low temperature, suggestive of a homogeneous FM state. On the other hand, coexistence of the PM and FM signals persists

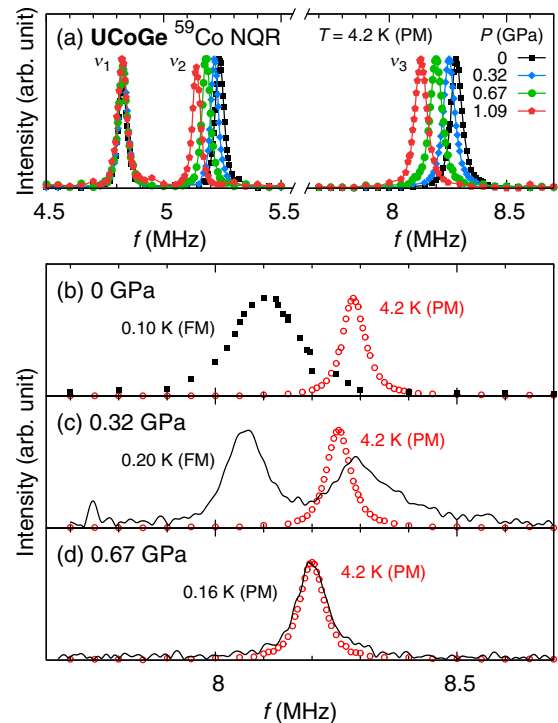


FIG. 2. ^{59}Co NQR spectra of UCoGe under zero field (a) at 4.2 K (PM state) arising from ν_1 , ν_2 , and ν_3 transitions, and (b)–(d) at various temperatures at ν_3 . The spectrum at $P = 0$ GPa and $T = 0.10$ K shown in (b) was from Ref. [8].

down to the lowest measurement temperature at 0.32 GPa. The PM signal becomes broader below T_{Curie} , which could be ascribed to the magnetostriction effect [30] because the existence of the partial FM regions with slightly different lattice constants leads to the local stress in the sample. The two peaks indicate the phase separation of the PM and FM phases and the first-order FM transition occurs at 0.32 GPa. Finally, no FM signal was detected at 0.67 GPa. These results indicate that the FM phase transition of UCoGe is already a first order at ambient pressure, and is completely suppressed at 0.67 GPa in our sample, indicating that the FM criticality of UCoGe is classified as the first-order QPT. The phase diagram of this sample is qualitatively in good agreement with the previous studies [19–21], although P_c is somewhat lower than the values in literature. This difference may reflect the remarkable sample dependence of the ferromagnetism of UCoGe [31]. The unchanged spectrum shown in Fig. 2(d) also indicates that an internal magnetic field was absent in the SC state at 0.16 K within the experimental resolution. Thus, a unitary state is realized in the PM SC state. The time-reversal symmetry has been anticipated in this phase because the FM transition is absent above T_{SC} [20], and the present result verified the absence of the internal field in the SC state from the microscopic point of view. We suggest that the nonunitary-unitary transition occurs around ~ 0.5 GPa in the present sample.

Figure 3 shows the results of nuclear spin-lattice relaxation rate $1/T_1$ divided by T at several pressures measured at the ν_3 line. The phase diagram of the present sample determined by the ac susceptibility and NQR is shown in the inset of Fig. 3. At ambient pressure, $1/T_1T$ exhibits strong enhancement around T_{Curie} due to strong FM fluctuations [8]. When the pressure increases, the peak temperature shifts lower owing to the suppression of the FM phase, and the FM fluctuations at T_{SC} are strongest at 0.67 GPa. A clear SC transition was observed even with strong FM fluctuations, and this is consistent with the scenario that the superconductivity is mediated by the Ising-type FM fluctuations [14]. At 1.09 GPa, the enhancement of the FM fluctuations becomes weaker and T_{SC} start to decrease, but the enhanced behavior still remains. The enhancement of $1/T_1$ usually implies a second-order FM QCP, but the clear first-order transition was observed in the NQR spectrum (Fig. 2). Thus, the FM transition of UCoGe is likely to be weakly first order near the tricritical point. We note the possibility that the hyperfine coupling constant might be changed by applying the pressure. In such a case, however, $1/T_1T$ and the Knight shift significantly change far above the magnetic ordering temperature, which was actually observed in CeRhIn₅ [32,33]. Because $1/T_1T$ is almost invariant at 60 K in UCoGe, the change of $1/T_1T$ is ascribed to the change of the spin fluctuations by pressure.

The SC state exists in the FM and PM sides in spite of the first-order FM transition by pressure. This is owing to the presence of strong FM fluctuations in both sides, and the discontinuity of the magnetism does not seriously affect the formation of the Cooper pairs. This is different from the case of UGe₂, where the FM phase vanishes with a first-order transition and neither critical fluctuations nor superconductivity was observed above P_c [34]. In addition, it is shown that T_{SC} of UCoGe is the highest at around the FM criticality. Thus,

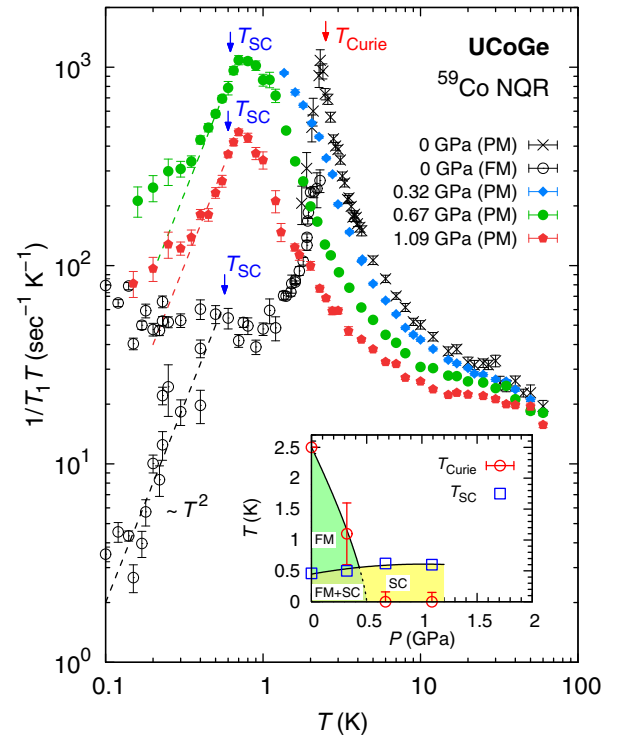


FIG. 3. ^{59}Co nuclear spin-lattice relaxation rate $1/T_1$ divided by temperature T under zero field in UCoGe. The result at 0 GPa was from Ref. [8]. The data below 1.4 K lacks for 0.32 GPa owing to the poor NQR intensity. $1/T_1$ was measured at the PM site except for 0 GPa below T_{Curie} , and was obtained at the ν_3 line (~ 8 MHz). Inset: pressure-temperature phase diagram of UCoGe determined by the ac susceptibility and NQR for the present sample.

the picture that the SC emerges around the FM criticality seems valid in UCoGe, and this system is similar to the case of the superconductors observed near the AFM phase. The remaining issue is whether UCoGe has a wing structure on pressure-field-temperature phase diagram characteristic to the quantum itinerant ferromagnet [22].

$1/T_1$ gives information about the SC gap structure as well as the magnetic fluctuations. Below T_{SC} , $1/T_1T$ rapidly decreases under all the pressures because of the opening of the SC gap. Line-nodal gap behavior was observed in the FM SC state at ambient pressure in the previous NQR [8], namely, $1/T_1T \sim T^2$. Similar line-node behavior is also confirmed with a thermal conductivity measurement [35]. Recently, it has been proposed that the line node of the SC gap is protected by the nonsymmorphic space-group symmetry, and it is expected that this gap structure persists in the PM SC state under pressure [36]. However, a deviation from this behavior was observed at 0.67 GPa and 1.09 GPa in the PM side at low temperatures. The deviation could be explained by a large residual density of states (DOS) in the SC state and also supports the nodal SC gap. The details of the gap structure are masked by this additional relaxation in the PM SC state. Since the multigap behavior was reported in the FM SC state at $P = 0$ of the recent high-quality samples by the thermal conductivity measurement [37], further measurements are necessary to reveal the SC gap structure of UCoGe.

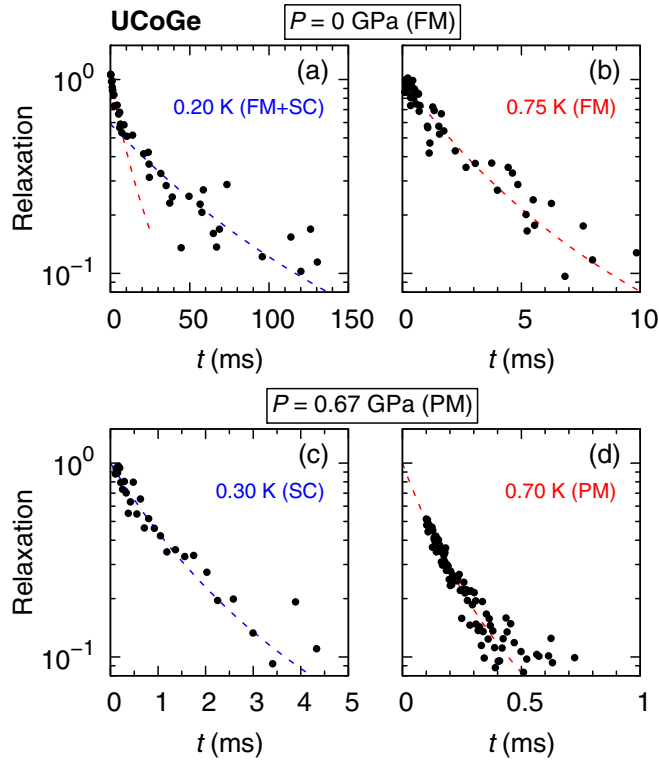


FIG. 4. ^{59}Co NQR relaxation curves of UCoGe at 0 (FM) and 0.67 GPa (PM) above and below T_{SC} . These data were measured at the ν_3 line (~ 8 MHz). The dashed lines are best fit of the results. The curves at ambient pressure denote the previous results [8]. The single-component relaxation persists in the SC state.

Figure 4 shows the nuclear relaxation curves at ambient pressure and 0.67 GPa, measured by ^{59}Co NQR. At the FM SC state, the relaxation curve shows two components below T_{SC} as shown in Fig. 4(a). The slower component shows the SC behavior, but the faster component shows the non-SC behavior (Fig. 3) [8]. The non-SC part was roughly 50% in the intensity and did not show large temperature dependence below T_{SC} . On the other hand, a single-component relaxation persists even below T_{SC} in the PM SC state [Fig. 4(c)]. These results indicate that the faster component in the FM SC state is an inevitable feature of UCoGe and originates from the FM phase, and the whole part of the sample exhibits superconducting in the PM SC state under pressure. In addition, the temperature-independent fraction of the faster component would be inconsistent with the nuclear spin diffusion by the presence of the diffusion center. Alternatively, we suggest that the faster relaxation could be explained by SIV owing to the coexistence of the FM and SC phases, as discussed in the previous paper [8]. In this scenario, the superconductivity is destroyed at the vortex core, which results in a normal-metal-like $1/T_1$. It is theoretically expected that the larger magnetization leads to a larger residual DOS owing to the partial pair breaking at the vortex core [38], and this tendency is consistent with experiments on the three FM superconductors [39]. Disorder-induced residual DOS behavior was seen in the present $1/T_1$ of UCoGe in the PM SC state under pressure as mentioned above; however, such a deviation was absent in the longer component of $1/T_1$ in the FM SC state at ambient

pressure. This difference is also explained by the formation of the vortex state, because the disorder region in the sample works as a pinning center of the vortex in the FM SC state and mainly contributes to the short- $1/T_1$ part. It should be noted that the large fraction of the non-SC part in the FM SC state is not evident because the internal field by the spontaneous magnetization at the FM state is two orders of magnitude smaller than H_{c2} along the c axis [26,27], and the estimation of the SIV region is of the order of the magnetization divided by H_{c2} [38]. The SIV region would be increased by the presence of a pinning center related to the disorder.

In addition to the SIV scenario, an interesting scenario to explain the non-SC fraction is the presence of unpaired electrons related to the spontaneous charge current resulting from the FM chiral SC state [40]. The scanning SQUID measurement revealed that the FM domain wall width is ~ 0.1 – 1 nm, and the size of the FM domains, an order of 10 μm , shows no large change across the SC transition [41]. If the FM chiral SC state is realized in UCoGe, it is expected that chiral SC domains are created and they coincide with the FM domains. This is the case for the so-called A symmetry expected from the theoretical works for the interpretation of the NMR results [14–16,42]. The spontaneous charge current would flow at the surface and the domain walls due to the opposite directions of the multidomain. With the multidomain structure, a state with finite sum of these currents can be more stable than a state with cancellation of these currents, and the Fulde-Ferrell-like supercurrent flows in the opposite direction inside the domain to cancel the net current in the domain. This current leads to spatially dependent pair breaking, and thus, this is also related to the two-component NQR relaxation in our sample [8] and a large residual specific-heat coefficient even in a high-quality sample [39]. Although the magnitude of this contribution to these experimental quantities and the relationship with the SIV state are still unclear, this scenario may explain the observed non-SC fraction. Further studies are needed to conclude the origin of this anomalous behavior in the FM SC state.

In conclusion, ^{59}Co NQR was performed on the FM superconductor UCoGe under pressure, and it was revealed that the FM fluctuations are enhanced around the critical pressure. This enhancement persists above P_c , and is closely related to the emergence of the SC phase in the PM side. The phase separation of the FM and PM phases indicates weakly first-order FM QPT. The nuclear relaxation curve has a single component in the PM SC state, which suggests that the fast relaxation in the FM SC state is a characteristic feature of UCoGe and is closely related to the interplay between the FM and SC states.

The authors would like to thank Y. Tokunaga, T. Hattori, D. Aoki, Y. Maeno, S. Yonezawa, A. Daido, Y. Yanase, J.-P. Brison, D. Braithwaite, A. Pourret, C. Berthier, A. de Visser, J. Flouquet, and V. P. Mineev for valuable discussions. This work was supported by Kyoto University LTM Center, and by Grant-in-Aid for Scientific Research (Grant No. JP15H05745), Grant-in-Aids for Scientific Research on Innovative Areas “J-Physics” (Grants No. JP15H05882, No. JP15H05884, and No. JP15K21732), and Grant-in-Aid for JSPS Research Fellow (Grant No. JP17J05509) from JSPS.

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