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Experimental evidence for ferromagnetic spin-pairing superconductivity emerging in UGe_2: A ^{73}Ge-nuclear-quadrupole-resonance study under pressure

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We report that a different type of superconducting order parameter has been realized in the ferromagnetic states in UGe_2 via ^{73}Ge-nuclear-quadrupole-resonance experiments performed under pressure (P). Measurements of the nuclear spin-lattice relaxation rate $(1/T_1)$ have revealed an unconventional nature of superconductivity such that the up-spin band is gapped with line nodes, but the down-spin band remains gapless at the Fermi level. This result is consistent with that of a ferromagnetic spin-pairing model in which Cooper pairs are formed among ferromagnetically polarized electrons. The present experiment has shed light on the possible origin of ferromagnetic superconductivity, which is mediated by ferromagnetic spin-density fluctuations relevant to the first-order transition inside the ferromagnetic states.

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The coexistence of magnetism and superconductivity (SC) has recently become an important topic in condensed-matter physics. The recent discovery of SC in the ferromagnets UGe_2 (Refs. 1 and 2) and URhGe (Ref. 3) has been a great surprise because the Cooper pairs are influenced by a nonvanishing internal field due to the onset of ferromagnetism (FM), which is believed to prevent the onset of SC. In the ferromagnet UGe_2 with a Curie temperature $T_{\mathrm{Curie}}=52$ K at ambient pressure $(P=0)$, the emergence of P-induced SC has been observed in the P range of 1.0–1.6 GPa.[1,2] It is noteworthy that the SC in UGe_2 disappears above $P_c \sim 1.6$ GPa, beyond which FM is suppressed. The SC and FM in this compound have been shown to be cooperative phenomena.[4] The superconducting transition temperature (T_{sc}) is the highest at $P_x \sim 1.2$ GPa, where a first-order transition occurs from FM2 to FM1 as P increases. Here, it should be noted that the ferromagnetic moments are increased in the first-order transition from FM1 to FM2 as functions of temperature and pressure, as shown in Fig. 1(a).[5–7] The P-induced SC in UGe_2 coexists with FM1 and FM2 phases, exhibiting large magnetization of order $1\mu_B$ per U even for the case of $T_{\mathrm{Curie}} \sim 30$ K.[6] Therefore, it is proposed that the onset of SC is suitable for the formation of a spin-triplet pairing state rather than a spin-singlet pairing state.[2] However, there are few reports that address the type of order parameter realized in FM1 and FM2.

In a previous study, an unconventional nature of the SC was suggested from the measurement of the ^{73}Ge-nuclear-quadrupole-resonance (NQR) nuclear spin-lattice relaxation rate $1/T_1$.[6] However, it is not well understood whether or not the presence of a residual density of states (RDOS) at the Fermi level in the SC state is intrinsic, suggesting the occurrence of a possible extrinsic effect due to the presence of any impurity and/or imperfection in the sample.[6] In particular, it is unclear why SC emerges with the highest T_{sc} when the first-order transition occurs from FM1 to FM2 at $P_x \sim 1.2$ GPa. In order to gain insight into this issue, further experiments are required for understanding the P-induced evolution in the FM states and the order-parameter symmetry emerging in the FM states in UGe_2.

In this Rapid Communication, by performing ^{73}Ge-NQR measurements under pressure at zero field $(H=0)$ on a newly prepared sample, we report that the SC in this compound is caused by the formation of up-spin Cooper pairs, where the gap opens only at the up-spin band in FM1 and FM2 but not at the down-spin band.[8–10] The ferromagnetic spin-pairing SC is considered to be mediated by ferromagnetic spin-density fluctuations relevant to the first-order transition inside the ferromagnetic states.

A polycrystalline sample enriched by ^{73}Ge was crushed into coarse powder for the NQR measurement and annealed

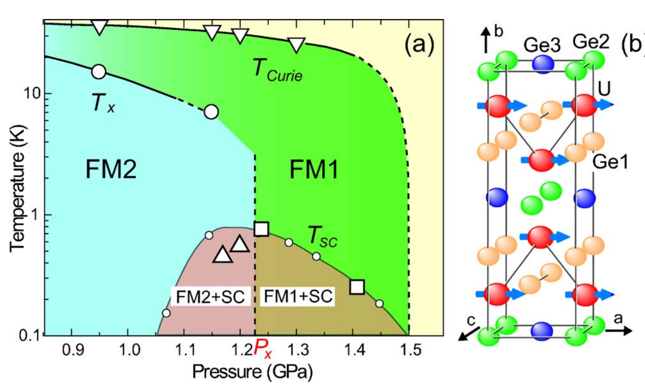

FIG. 1. (Color online) (a) Pressure versus temperature phase diagram of UGe_2 near the superconducting phase (Ref. 6). The T_{sc} values of FM1 (open squares) and FM2 (open triangles) and the P_x value determined in this study are plotted. (b) Crystal structure of UGe_2 with the ferromagnetic moment at the U site below T_{Curie}.

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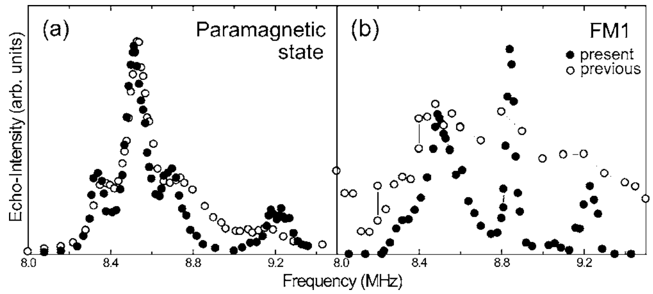

FIG. 2. Comparison of the ^{73}Ge-NQR spectra of the present and previous samples of UGe_2 in (a) the paramagnetic state and (b) FM1. The spectra at $P=1.9$ (a) and (b) 1.41 and 1.3 GPa are shown by solid and open circles, respectively, demonstrating that the present sample has better quality than the previous samples (Ref. 6).

to maintain its quality. The NQR experiments were performed by the conventional spin-echo method at $H=0$ in the frequency (f) range of 5–11 MHz at $P=1.17$, 1.2, 1.24, and 1.41 GPa. Hydrostatic pressure was applied by utilizing a NiCrAl-BeCu piston-cylinder cell filled with Daphne oil (7373) as a pressure-transmitting medium. The value of P at low temperatures was determined from the T_{sc} of Sn measured by a resistivity measurement. The possible distribution of the pressure inside the sample was less than 3% in the present experimental setup. A ^{3}He-^{4}He dilution refrigerator was used to obtain the lowest temperature of 50 mK. Figures 2(a) and 2(b) show the NQR spectra in the paramagnetic and FM1 phases, respectively. The linewidths in these NQR spectra are narrower for the present sample than for the previous sample, demonstrating that the quality of the present sample is significantly higher than that of the previous sample. Moreover, the NQR T_1 measurements reveal that the present sample exhibits the highest value of $T_{sc}=0.75$ K obtained thus far at $P=1.24$ GPa, to the best of our knowledge, ensuring higher quality than before.

Figure 3(a) shows the NQR spectra for the PM phase at 4.2 K and $P=1.9$ GPa where FM1 is completely suppressed. They reveal a structure consisting of separated peaks associated with three inequivalent Ge sites in one unit cell in the crystal structure illustrated in Fig. 1(b).[4,6] The number of Ge1 sites is twice that of the Ge2 and Ge3 sites in one unit cell. The Ge1 site is closely located along the uranium (U) zigzag chain, while the other two sites Ge2 and Ge3 are located outside this zigzag chain. From the analysis of the NQR spectra for the FM1 phase in previous experiment,[6] it was demonstrated that the onset of FM1 induces an internal field $H_{int}=0.9$ T at the Ge sites that additionally causes the Zeeman splitting in each Ge-NQR spectrum. Furthermore, the angle between the principal axis for the nuclear quadrupole Hamiltonian and the direction of H_{int} was determined as $\theta \sim \pi/3$. When the first-order transition occurs from FM1 to FM2, the spectral shape changes significantly from the spectra at $P=1.24$ and 1.41 GPa to those at $P=1.17$ and 1.2 GPa, as shown in Figs. 3(b)–3(e). From the analysis of the spectra, it is estimated that $H_{int}=0.9$ T for FM1 increases to $H_{int}=1.8$ T for FM2 [see Fig. 5(b)]. The sudden increase in H_{int} in the narrow P range should be relevant to the first-order transition at P_x. In such a case, the spectra near P_x are expected to reveal a mixture of both domains FM1 and FM2 in the narrow range of P close to P_x due to an inevitable distribution of P. In fact, the spectra at $P=1.2$ and 1.24 GPa are composed of spectra arising from FM1 ($P=1.41$ GPa) and FM2 ($P=1.17$ GPa) with the ratios of 1:9 and 7:3, respectively, as shown by the dashed lines in Figs. 3(b) and 3(c). By considering the inevitable P distribution ($\Delta P=0.04$ GPa) as a Gaussian function given by $\exp(-\{[P-P_0]/[\Delta P/(2\sqrt{\ln 2})]\}^2)$, we obtain $P_x=1.23$ GPa. The present experimental results reveal that the first-order transition occurs at $P_x=1.23$ GPa.

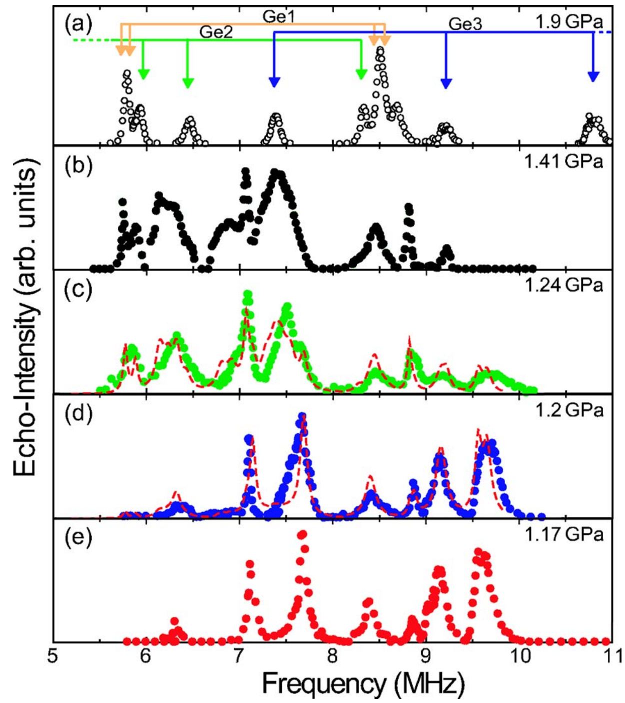

FIG. 3. (Color online) (a) ^{73}Ge-NQR spectra at 4.2 K in the P-induced paramagnetic phase. The NQR spectra in (b), (c), (d), and (e) represent the ferromagnetic phases at 1.4 K and $P=1.41$, 1.24, 1.2, and 1.17 GPa, respectively. The dashed lines in the figures indicate the simulated results (see text).

Figure 4(a) shows the T dependences of $1/T_1$ for FM1 and FM2 at pressures that are slightly lower and higher than $P_x=1.23$ GPa, respectively. Clearly, $1/T_1$ for FM1 and FM2 decreases without any indication of a coherence peak just below T_{sc}, which provides evidence for the uniform coexistence of the unconventional SC and ferromagnetism. In a previous study,[6] the line-node gap model with RDOS N_{res} at the Fermi level was applied to interpret the systematic evolution in the superconducting energy gap Δ and the fraction of the RDOS N_{res}/N_0. Here N_0 is the density of state (DOS) at the Fermi level in the FM phases. It should be noted that all the data of $1/T_1$ are uniquely determined in the present sample, but not in the previous sample.[6] Therefore, we could not exclude the fact that the RDOS is present in the previous sample due to some impurity effect. Similarly, for the present sample with higher quality than that of the previous sample, the application of the line-node gap model with the RDOS allows us to estimate $N_{res}/N_0=0.50$, 0.48, 0.29, and 0.30 and $T_{sc}=0.45$, 0.55, 0.75, and 0.25 K (±0.05 K) at $P=1.17$, 1.2,

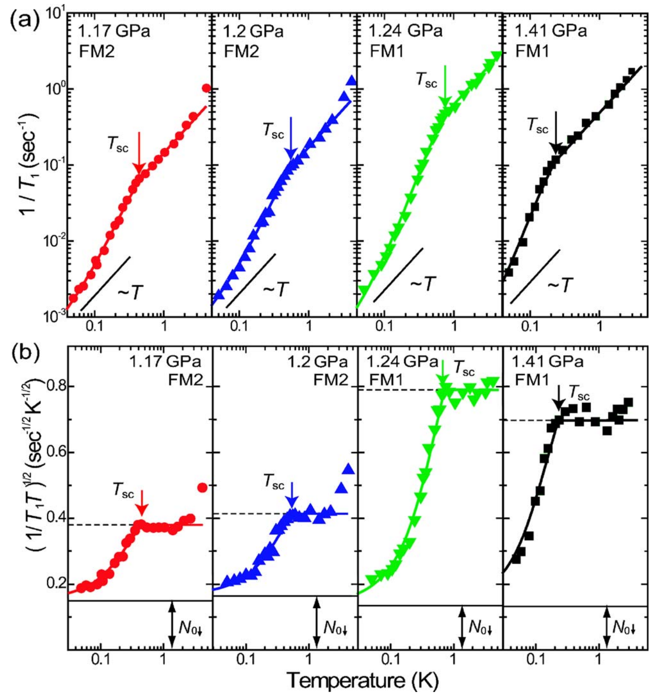

FIG. 4. (Color online) (a) Temperature dependences of $1/T_1$ for FM2 at P=1.17 and 1.2 GPa measured at f=7.68 MHz and for FM1 at P=1.24 and 1.41 GPa measured at 7.09 and 7.07 MHz, respectively. (b) Temperature dependences of $(1/T_1T)^{1/2}$ related to the DOS in either the SC state or the normal state at each P. The solid curves represent the results calculated based on the ferromagnetic spin-pairing model (see the text).

1.24, and 1.41 GPa, respectively. It should be noted that T_{sc} decreases from 0.45 to 0.25 K although N_{res}/N_0 does decrease from 0.50 to 0.30 at P=1.2 GPa in FM2 and at P=1.41 GPa in FM1. These results demonstrate that the presence of a RDOS in the superconducting state is not due to the impurity effect but intrinsic in origin. Although some impurity- and/or imperfection-based effects, if any, are not completely ruled out, we state that the observation of the highest T_{sc}=0.75 K, to our knowledge, ensures that the present sample is one of the best quality samples reported thus far.

First, we address whether or not the RDOS is associated with a self-induced vortex state in the SC+FM uniformly coexisting state. By assuming the Abrikosov triangular vortex lattice, a coherence length $\xi \sim 130$ Å,[7] and an internal magnetic field H=0.125 T,[11] we consider that only 3% of N_{res}/N_0 arises from the normal state inside the self-induced vortex core in the SC+FM state, which does not agree with the experimental result. Alternatively, in another promising scenario that explains the RDOS, we consider a nonunitary spin-triplet pairing model.[8] In this model, the superconducting energy gap opens only in the up-spin band parallel to the magnetization of FM phases, but not in the down-spin band which remains gapless. We begin by assigning possible nuclear relaxation processes of Ge-NQR T_1 in the ferromagnetic states. One of them is caused by the transversal component of fluctuations of internal magnetic fields at the Ge sites originating from intraband and interband transitions across the Fermi level at each up-spin and down-spin band. Another one is caused by only the interband spin-flip transition across the Fermi level between the up-spin and down-spin bands. By considering these relaxation processes, $1/T_1T$ in the FM state is expressed as

$$\frac{1}{T_1T} \propto 2t_2(T)\cos^2\theta + [t_1(T) + 2t_2(T) + t_3(T)]\sin^2\theta,$$

$$t_1(T) = \frac{1}{k_BT}\int_0^\infty dE\, N_\downarrow^2(E)f(E)[1-f(E)],$$

$$t_2(T) = \frac{1}{k_BT}\int_0^\infty dE\, N_\uparrow(E)N_\downarrow(E)f(E)[1-f(E)],$$

$$t_3(T) = \frac{1}{k_BT}\int_0^\infty dE\, N_\uparrow^2(E)f(E)[1-f(E)],$$

where $t_1(T)$, $t_2(T)$, and $t_3(T)$ indicate the former contributions and $t_2(T)$ represents the latter contribution which is possible only for θ=0. When the energy dependence of the DOS is neglected near the Fermi level, all contributions of $t_1=N_{0\downarrow}^2$, $t_2=N_{0\uparrow}N_{0\downarrow}$, and $t_3=N_{0\uparrow}^2$ are independent of temperature. Here, $N_{0\uparrow}$ and $N_{0\downarrow}$ are the DOSs at the up-spin and the down-spin bands at the Fermi level in the normal FM state, respectively. θ is the angle between the quantization axis of the ^{73}Ge-nuclear-quadrupole Hamiltonian and that of H_{int} at the Ge site in the FM state, which is estimated as $\theta \sim \pi/3$ from analysis of the NQR spectra in the FM state, and $f(E)$ is the Fermi distribution function. In the ferromagnetic spin-pairing model, a line-node gap of $\Delta(\phi)=\Delta_0\cos\phi$ is assumed only for the density of states $N_{s\uparrow}(E)$ at the up-spin band, but not for $N_{s\downarrow}(E)$ at the down-spin band. It should be noted that, if θ=0, $t_2(T)$ should behave as $1/T_1 \propto T^2$ well below T_{sc}. In the present case, because $\theta \sim \pi/3$, the gapless term t_1 gives rise to the RDOS at the Fermi level in the superconducting state, as shown in Fig. 4(b). In fact the experimental results are actually in good agreement with this theoretical model, as indicated by the solid lines in Figs. 4(a) and 4(b). Therefore, the SC energy gap Δ and $N_{0\uparrow}/N_0$ are estimated as $2\Delta/k_BT_{sc} \sim 3.7$, 3.8, 4.0, and 3.7 with $N_{0\uparrow}/N_0$=0.57, 0.57, 0.82, and 0.80 at P=1.17, 1.2, 1.24, and 1.41 GPa, respectively. Here, $N_0(P)=N_{0\uparrow}(P)+N_{0\downarrow}(P)$.

In order to gain further insight into this SC state, Fig. 4(b) shows the T dependence of $(1/T_1T)^{1/2}$ related to the DOS at the Fermi level in either the SC or the normal FM state. As shown in Fig. 5(c), the most interesting finding is that $N_{0\uparrow}(P)$ dramatically increases as P increases slightly from P=1.2 to 1.24 GPa across P_x=1.23 GPa, accompanying the sudden reduction of H_{int} shown in Fig. 5(b). By contrast, $N_{0\downarrow}(P)$ remains almost constant and the T-linear coefficient of the specific heat γ gradually increases with P across P_x.[12] These results reveal that the Fermi level in FM2 is located just above a sharp peak in the majority up-spin band, and as P increases across P_x, it shifts down toward the peak when it enters the FM1 phase. In the ferromagnetic spin-pairing SC state, the large DOS in the up-spin band in FM1 enhances

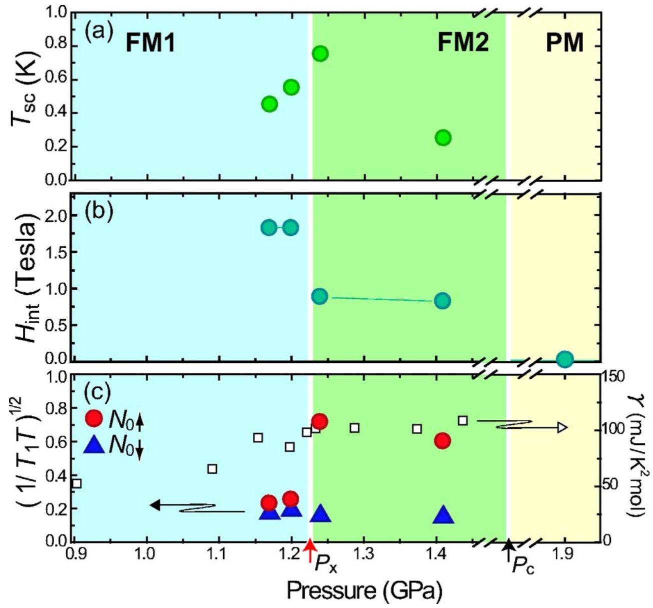

FIG. 5. (Color online) Pressure dependence of (a) T_{sc}; (b) internal magnetic field H_{int} at the Ge1 site; and (c) relative P dependence of $N_{0\uparrow}$ (solid circles) and $N_{0\downarrow}$ (solid squares) estimated from the ferromagnetic spin-pairing model on a scale of $(1/T_1T)^{1/2}$ and the T-linear coefficient of the specific heat γ (Ref. 13) (open squares) across P_x (see text). It should be noted that $N_{0\uparrow}$ dramatically increases when the first-order transition from FM2 at $P=1.2$ GPa to FM1 at 1.24 GPa occurs across $P_x=1.23$ GPa.

T_{sc}, leading to the highest value of $T_{sc}=0.75$ K, whereas its reduction in FM2 decreases T_{sc}, as shown in Figs. 5(a) and 5(c). However, it should be noted that, as P increases further up to $P=1.41$ GPa, even though $N_{0\uparrow}(P)$ for FM1 remains rather larger than that for FM2, $T_{sc}=0.25\pm0.05$ K for FM1 at $P=1.41$ GPa becomes lower than $T_{sc}=0.45$ K for FM2 at $P=1.17$ GPa. In this context, the large increase in $N_{0\uparrow}(P)$ is not always the main factor that increases T_{sc}. Rather, the first-order transition from FM2 to FM1 at P_x is responsible for the mediation of the up-spin Cooper pairing. The longitudinal FM spin fluctuations along the a axis[13] soften in energy at the critical end point for the first-order transition at P_x. Therefore, we suggest that these longitudinal FM fluctuations along the a axis would be a mediator of the ferromagnetic spin-pairing SC where the majority up-spin band in the FM phases is gapped, while the minority down-spin band is not.

In another context, it is predicted that T_x could be identified with the formation of a simultaneous charge- and spin-density wave (CSDW) induced by the imperfect nesting of the Fermi surface for the up-spin band and hence the superconducting pairing is mediated by CSDW fluctuations around P_x.[14] Although the NQR spectrum does not directly evidence an onset of static CSDW states, the remarkable increase in $N_{0\uparrow}(P)$ across P_x is relevant to the nesting at the Fermi surface for the up-spin band below T_x.

In conclusion, ^{73}Ge-NQR measurements under pressure on well-characterized UGe_2 have revealed that the superconducting energy gap opens only with the line node at the Fermi level in the majority up-spin band, but the down-band remains gapless. It is therefore concluded that ferromagnetic spin-pairing SC occurs in UGe_2. We have also shown that the first-order transition from FM1 to FM2 at $P_x=1.23$ GPa occurs because the Fermi level is located just on the peak in the DOS of the up-spin band. The ferromagnetic spin-density fluctuations emerging in the vicinity of the critical end point for this first-order transition are considered to be the mediator of the onset of SC realized in ferromagnetic UGe_2.

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