Spin-triplet superconductivity in the paramagnetic UCoGe under pressure studied by ^{59}Co NMR

Masahiro Manago,* Shunsaku Kitagawa, and Kenji Ishida
Department of Physics, Graduate School of Science, Kyoto University, Kyoto 606-8502, Japan

Kazuhiko Deguchi and Noriaki K. Sato
Department of Physics, Graduate School of Science, Nagoya University, Nagoya 464-8602, Japan

Tomoo Yamamura†
Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

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A ^{59}Co nuclear magnetic resonance (NMR) measurement was performed on the single-crystalline ferromagnetic (FM) superconductor UCoGe under a pressure of 1.09 GPa, where the FM state is suppressed and superconductivity occurs in the paramagnetic (PM) state, to study the superconducting (SC) state in the PM state. ^{59}Co-NMR spectra became broader but hardly shifted across the SC transition temperature. The Knight-shift change determined from fitting the spectral peak with a Gaussian was much smaller than the spin part of the Knight shift; this is in good agreement with the spin-triplet pairing suggested from the large upper critical field. The spectrum broadening in the SC state cannot be attributed to the SC diamagnetic effect but is related to the properties of spin-triplet superconductivity. The origins of the broadening are discussed herein.

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I. INTRODUCTION

Unconventional superconductivity mediated by a mechanism other than ordinary electron-phonon coupling is one of the most interesting phenomena in condensed-matter physics, and the symmetry of the pairing often differs from the *s* wave. Among the various unconventional superconductors reported so far, spin-triplet (odd-parity) superconductors are quite rare systems. Cooper pairs have spin degrees of freedom in the superconducting (SC) state in this case, leading to various exotic phenomena. The spin-triplet pairing was first identified in superfluid ^{3}He [1], and odd-parity pairing is most likely realized in a heavy-fermion superconductor UPt_3 [2]. The possibility of spin-triplet pairing was also noted in Sr_2RuO_4 [3–6], and this system has been intensively studied thus far to identify its pairing symmetry.

The family of ferromagnetic (FM) superconductors is another class of unconventional superconductors [7–11]. The odd-parity pairing is expected in this family because of the coexistence of ferromagnetism and superconductivity. The pairing mechanism of these systems is closely related to the FM instability, and this pairing glue can be tuned by the external magnetic field, resulting in the unusual field dependence of the SC transition temperature T_{SC}. For UCoGe, it was reported from a field-angle-controlled nuclear magnetic resonance (NMR) experiment that the Ising-type FM fluctuations along the *c* axis are essential for the pairing [12]. The changes in pairing strength with the fields were quantitatively analyzed by measuring the upper critical field H_{c2}, and this result supports the scenario of FM fluctuation-mediated superconductivity [13]. FM fluctuations also play an important role in superconductivity in URhGe; however, transverse fluctuations as well as those parallel to the *c* axis were also found to contribute to the pairing [14]. This is confirmed by the enhancement of the superconductivity by a uniaxial stress [15]. Therefore, these systems are suitable for studying the SC mechanism in detail.

UCoGe is a unique system because its SC phase remains beyond the suppression of the FM phase by pressure [16–18], and its superconductivity is expected to possess a spin-triplet symmetry on both the FM and paramagnetic (PM) sides. Actually, H_{c2} is far above the Pauli-limiting field estimated from T_{SC} at ambient pressure [19] and under pressure above P_c [17,18] when the field is perpendicular to the *c* axis. These results are consistent with the spin-triplet pairing. The absence of the Knight-shift decrease in the FM SC state also supports the spin-triplet scenario [10]. However, it is not trivial to achieve such a large H_{c2} in the *a* and *b* axes because the magnetic easy axis is the *c* axis and the spins of the Cooper pair cannot rotate freely in the presence of strong spin-orbit coupling. The absence of the Pauli-PM effect in the FM SC state is explained by the presence of the large exchange field [20]; however, this mechanism does not work on the PM side. The rotation of the $\boldsymbol{d}$ vector perpendicular to the field is therefore predicted under pressure with fields [21], where the $\boldsymbol{d}$ vector has been conventionally used to represent the spin state in the spin-triplet pairing and is perpendicular to the spins of the Cooper pairs. Such a rotation of the $\boldsymbol{d}$ vector leads

*Present address: Department of Physics, Kobe University, Kobe 657-8501, Japan; manago@crystal.kobe-u.ac.jp
†Present address: Institute for Integrated Radiation and Nuclear Science, Kyoto University, Kumatori 590-0494, Japan.

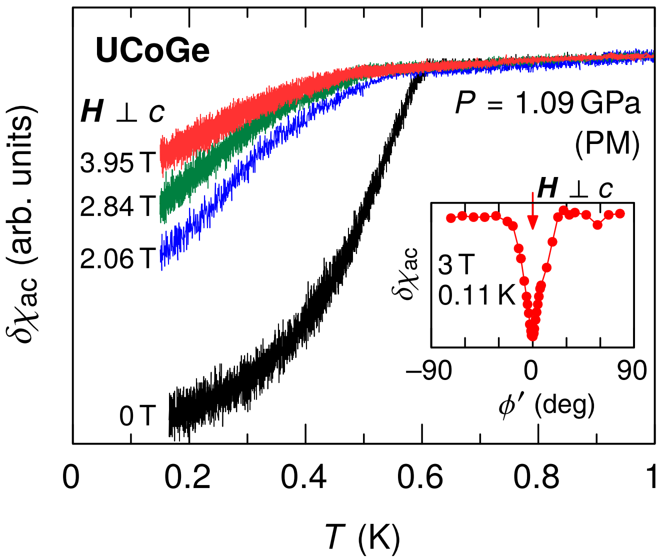

FIG. 1. Temperature dependence of ac susceptibilities $\delta\chi_{\mathrm{ac}}$ of UCoGe at some magnetic fields perpendicular to the c axis as measured using an NMR coil on cooling. Superconducting transition is detected in all fields. The frequencies are 8–9 MHz. Inset: field-angle dependence of $\delta\chi_{\mathrm{ac}}$ at 3 T and 0.11 K.

to an unchanged spin susceptibility in the SC state, resulting in the absence of the Pauli-PM effect. Because only a few experimental results on SC symmetry have been reported so far in the PM SC state under pressure, the properties of this state remain unclear.

To obtain microscopic information about the superconductivity of UCoGe on the PM side, we performed ^{59}Co NMR Knight-shift measurements under pressure. The result is in good agreement with the spin-triplet pairing suggested from the large H_{c2} when the field is applied to the ab plane. In this case, the NMR spectrum anomalously broadened, which cannot be understood by the SC diamagnetic effect but is related to the properties of spin-triplet superconductivity.

II. EXPERIMENT

A single-crystalline sample was used for ^{59}Co NMR measurements, and its FM and SC transition temperatures at ambient pressure are $T_{\mathrm{Curie}} = 2.5$ K and $T_{\mathrm{SC}} = 0.46$ K, respectively. This sample is the same one used in a recent nuclear quadrupole resonance (NQR) measurement under pressure [22]. Hydrostatic pressure was applied using a piston-cylinder-type cell with Daphne oil 7373 as a pressure medium, and the applied pressure was calibrated using an SC transition temperature of a Pb sample inside the cell. The thermometer was attached outside the pressure cell. Because the pressure cell and the thermometer are immersed in a ^{3}He-^{4}He mixture of a dilution refrigerator, thermal contact between the sample and thermometer is sufficient for the present measurement. The difference in T_{SC} of UCoGe determined by the cooling and warming processes is 0.01 K. The magnetic field was applied using a transverse SC magnet, enabling us to control the field angle precisely.

Figure 1 shows the temperature dependence of the ac susceptibility $\delta\chi_{\mathrm{ac}}$ of UCoGe at some magnetic fields with the field perpendicular to the c axis determined by the change of the tuning frequency of the LC circuit. The SC transition temperature of UCoGe increases to $T_{\mathrm{SC}} = 0.60$ K at 1.09 GPa. No FM transition was detected at this pressure [22]. The field was aligned from the local minimum of the field-angle dependence of $\delta\chi_{\mathrm{ac}}$ at $T = 0.11$ K with 3-T field, as shown in the inset of Fig. 1. A clear diamagnetic signal was observed in the narrow-angle region with $H \perp c$ because of the large H_{c2} anisotropy. The field direction in the ab plane could not be determined only from $\delta\chi_{\mathrm{ac}}$, and this was identified from the NMR spectra, as shown below.

The crystal structure of UCoGe possesses a $Pnma$ space group (No. 62, D_{2h}^{16}), and the Co site only has mirror symmetry with respect to the b plane [23]. The Co sites will split into two sites if the external magnetic field is not parallel to the ab or bc plane for the NMR measurements. The single site is observed with fields parallel to the ab plane, but the spectral positions differ in different field directions owing to the low symmetry of the local site. This feature enables us to deduce the full information about the applied field from the NMR spectrum.

The nuclear Hamiltonian of ^{59}Co ($I = 7/2$) in UCoGe consists of two parts. One is a Zeeman Hamiltonian of the nuclear magnetic moment, and the other is the electric quadrupole Hamiltonian arising from the coupling between the nuclear quadrupole moment and the electric field gradient (EFG). Then, the total Hamiltonian is represented as

$$\begin{aligned}\mathcal{H} &= \mathcal{H}_Z + \mathcal{H}_Q \\ &= -\gamma_N\hbar(1+\boldsymbol{K})\boldsymbol{I}\cdot\boldsymbol{H} \\ &\quad + \frac{\hbar\omega_Q}{6}\left\{\left[3I_z^2 - I(I+1)\right] + \frac{1}{2}\eta(I_+^2 + I_-^2)\right\},\end{aligned} \tag{1}$$

where γ_N is a gyromagnetic ratio of the nucleus, $\boldsymbol{H}$ is an external magnetic field, $\boldsymbol{K}$ is a Knight-shift tensor, ω_Q is a quadrupole frequency, and η is an asymmetric parameter of the EFG. This Hamiltonian is an expression in a particular coordinate, namely, the principal axis of the EFG. The z axis is the direction where the EFG is maximum, and the y is the second maximum direction. In the case of the ^{59}Co site in UCoGe, the z direction is $\sim 10^\circ$ tilted to the crystallographic c axis from the a axis, and the y axis is parallel to the b axis [24]. The quadrupole parameters are $\nu_Q \equiv \omega_Q/(2\pi) = 2.85$ MHz and $\eta = 0.52$ at ambient pressure [25], and these values slightly change as pressure increases. We used the values $\nu_Q = 2.795$ MHz and $\eta = 0.535$ determined at 1.09 GPa [22]. If the Zeeman Hamiltonian is a dominant term in the above Hamiltonian, seven lines will be observed, and they arise from $m \leftrightarrow m-1$ transitions ($m = 7/2, 5/2, \ldots, -5/2$). These line positions strongly depend on the field angle with respect to the EFG coordinate as well as the anisotropy of the Knight shift, and thus, the analysis of the spectral positions enables us to deduce the field direction. The Knight shift was measured at the central line arising from the $1/2 \leftrightarrow -1/2$ transition, and $^{59}\gamma_N/2\pi = 10.03$ MHz/T was used as a nuclear gyromagnetic ratio. The effect of the quadrupole shift was subtracted by using the numerical diagonalization of the nuclear Hamiltonian.

An attempt was made to attach the sample inside the pressure cell such that the crystallographic c axis is along the vertical direction; however, it was found that the c axis

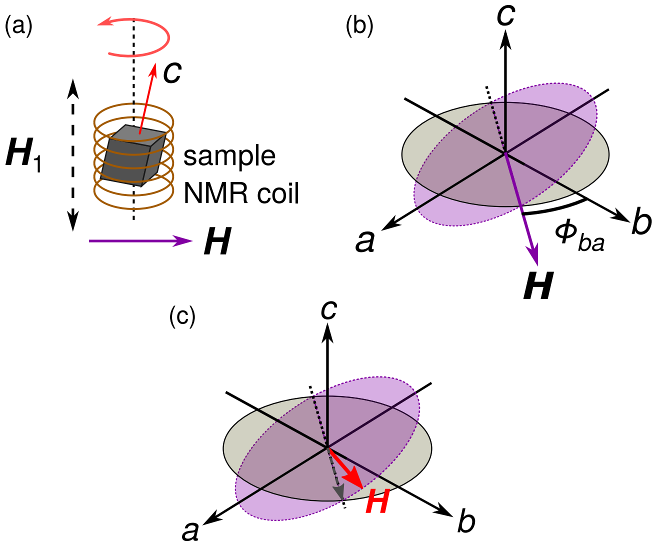

FIG. 2. (a) The schematic image of the static field H, NMR rf field H_1, and the sample directions. (b) The field-angle direction. The magnetic field is not perpendicular to the c axis in most directions and is perpendicular to the c axis only at a point. (c) The field slightly tilted toward the c axis.

was tilted. Because we could rotate the field angle only horizontally, the field direction was not aligned parallel to the a or b axis. The schematic image of the field alignment for the NMR measurements is shown in Fig. 2. This field direction perpendicular to the c axis was identified from the analyses of the field-swept NMR spectra at 0.8 K in the normal state, as shown in Fig. 3. Seven peaks were observed, which are due to the nuclear quadrupole splitting of ^{59}Co and consistent with $H \perp c$, although non-negligible broadening was visible

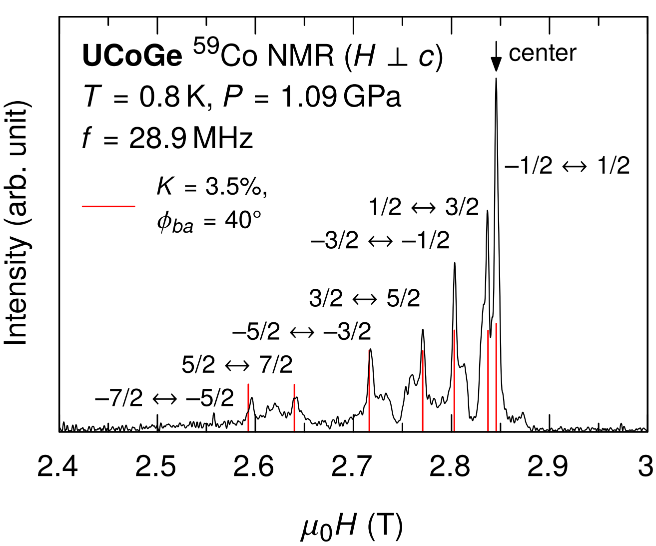

FIG. 3. Field-swept ^{59}Co NMR spectra with the field perpendicular to the c axis at 0.8 K (normal state). The vertical red line is the best fit of the spectra with $\phi_{ba} = 40^\circ$ by numerical diagonalization of the nuclear Hamiltonian. The vertical black arrow indicates the "central" ($1/2 \leftrightarrow -1/2$) peak, at which the Knight shift was measured. This peak is not located at the center of the seven splitting lines owing to the low-symmetric field direction.

at the satellite peaks. The vertical red line is the best fit of the calculated NMR peak positions by numerical diagonalization. The field angle tilts by $\phi_{ba} = 40^\circ$ from the b axis to the a axis. Further measurement of the NMR spectrum with a different field direction revealed that the c axis of the sample is tilted by 17° from the vertical direction. See Appendix A for the details on determining this angle.

III. RESULTS AND DISCUSSION

Figure 4 shows the ^{59}Co NMR spectra with a field of 2–4 T parallel to the ab plane, sufficiently smaller than $\mu_0 H_{c2} \sim 15$ T [17,18]. The Knight shift was analyzed by fitting the spectral peak with a Gaussian function, and the result is shown in Fig. 5. The Knight shift is expressed as $K(T) = K_{\rm spin}(T) + K_{\rm orb}$, where $K_{\rm spin}(T)$ is proportional to temperature-dependent spin susceptibility and the orbital shift $K_{\rm orb}$ is usually temperature independent. The spin and orbital components of the ^{59}Co site in UCoGe are evaluated from the ^{59}Co and ^{73}Ge NMR at ambient pressure [26]. The spin component $K_{\rm spin}$ along the b axis is not far from the estimated value from the experimental specific-heat coefficient in the normal state and hyperfine coupling constant [27]. The spin component is $K_{\rm spin} \simeq 1.1\%$ with this field direction at low temperatures below 1 K, as shown by the vertical arrow in Fig. 5, and it mainly originates from the spin susceptibility along the b axis because of the in-plane magnetic anisotropy [26]. The observed change across $T_{\rm SC}$ is an order of 0.1% and is much smaller than the spin part of the Knight shift. This result indicates that the spin susceptibility $\chi_{\rm spin}$ does not decrease in the SC state drastically, and the possibility of spin-singlet pairing is excluded, and spin-triplet superconductivity is suggested in this phase. We note that the NMR spectra became appreciably broader across $T_{\rm SC}$, as shown in the inset of Fig. 5 and in Fig. 6; this anomaly indicates that the Knight shift is actually measured in the SC state. In ordinary superconductors, this broadening is attributed to the effect of the vortex; however, the diamagnetic shift in the vortex state of UCoGe is estimated to be $\sim 10^{-4}\%$ at 2 T if we use parameters at ambient pressure [28], and therefore, the broadening originates from the change in the spin susceptibility.

To reveal the origin of the anomalous broadening of the NMR spectra, the spectra were measured with a tilted field. Figure 7 shows the NMR spectra with the field tilted by $\delta\theta_c \sim 5^\circ$ from the ab plane to the c axis. The projected field to the ab plane is $\phi_{ba} \simeq 24^\circ$ away from the b axis to the a axis. The Knight shift showed a small increase in the SC state without any significant broadening, as shown in Fig. 7. This indicates that the broadening of the spectrum in the $H \parallel ab$ plane arises from the following two effects. One is the Knight-shift decrease from the sample region where the field is exactly aligned in plane, and the other is the Knight-shift increase arising from the tiny misalignment of the field from the c axis. This misalignment is smaller than $\pm 5^\circ$.

It is surprising that the NMR spectra in the SC state exhibit a drastic change with tilting the field by only $\sim 5^\circ$ to the c axis. This result originates from the high sensitivity of UCoGe to the c component of the field. Such an anomalous field-angle dependence is also seen at the ambient pressure in H_{c2} [29] and nuclear spin-lattice relaxation rate $1/T_1$ [12]. This feature

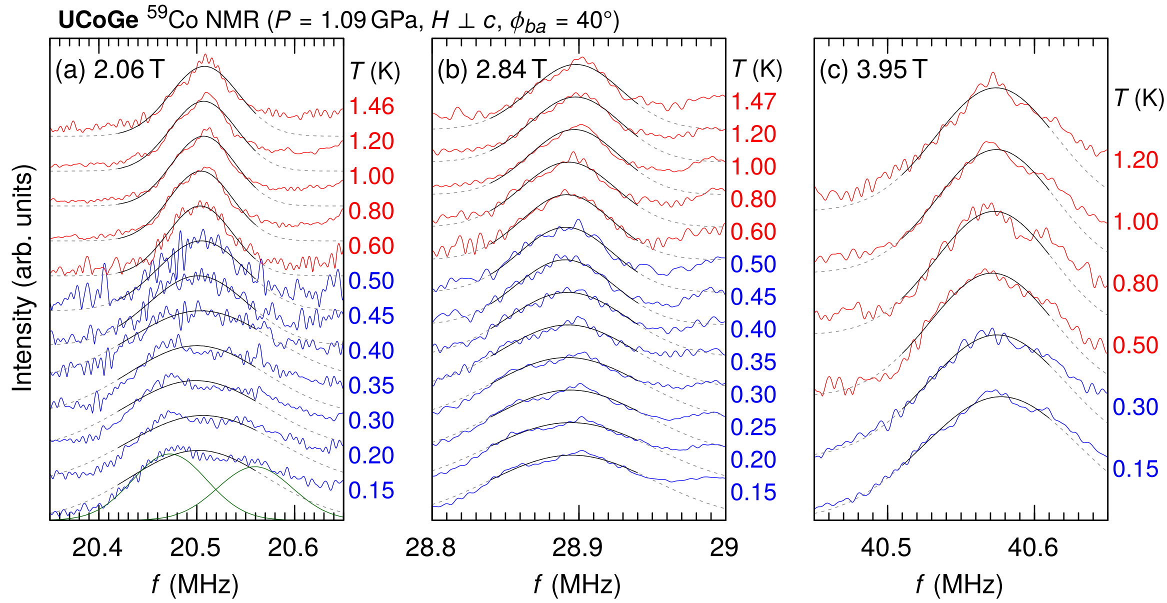

FIG. 4. ^{59}Co NMR spectra with field perpendicular to the c axis. The solid black lines indicate the fitted result obtained using a Gaussian and represent the fitting range. The range was shifted lower to avoid the effect of the first satellite peak at higher frequency near the central peak. The red (blue) lines represent the result of the normal (SC) state. The spectra were also fitted with two-component Gaussians at 2.06 T in the SC state, and each part is shown for 0.15 K with green lines (see the Appendix for details).

is considered to originate from the strong Ising anisotropy along the c axis of UCoGe, and the application of the field along this direction changes the electronic state.

The origin of this field-angle distribution is most likely due to the sample distortion by the pressure. The details of this discussion and the analysis based on the two-component Gaussian with $H \parallel ab$ are shown in Appendix B. The result of the two-component fitting at 2.06 T is shown in Fig. 5, and the increasing and decreasing parts are reasonably extracted in the SC state. Although clear two-component spectra are not seen at higher fields, the broadening of the spectra in the SC state suggests that two-component spectra persist even at higher fields. The Knight-shift decreases in the SC state for a field

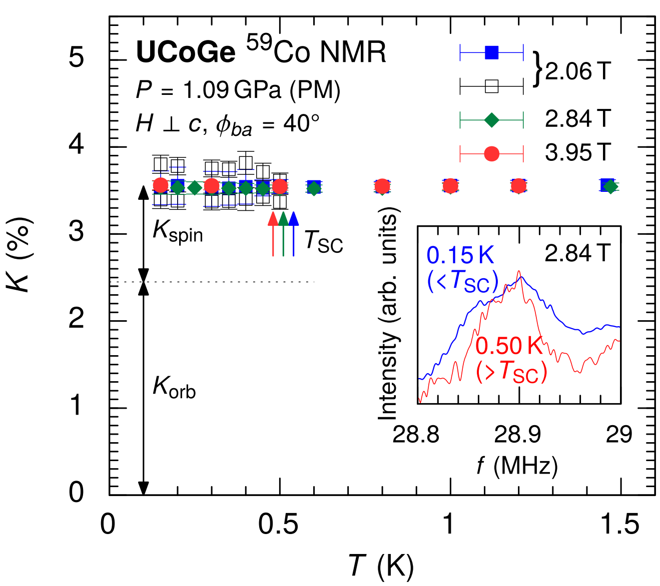

FIG. 5. Temperature dependence of the ^{59}Co NMR Knight shift of UCoGe with the field 40° away from the b axis to the a axis at the central ($1/2 \leftrightarrow -1/2$) line. The errors are estimated from the 90% (80% for the SC state at 2.06 T) width of the spectra. The open black squares indicate the result of the two-component fitting in the SC state (see text for details). Inset: ^{59}Co-NMR spectra at the central line at 2.84 T. Another peak at around 29 MHz originates from the first satellite owing to quadrupole splitting.

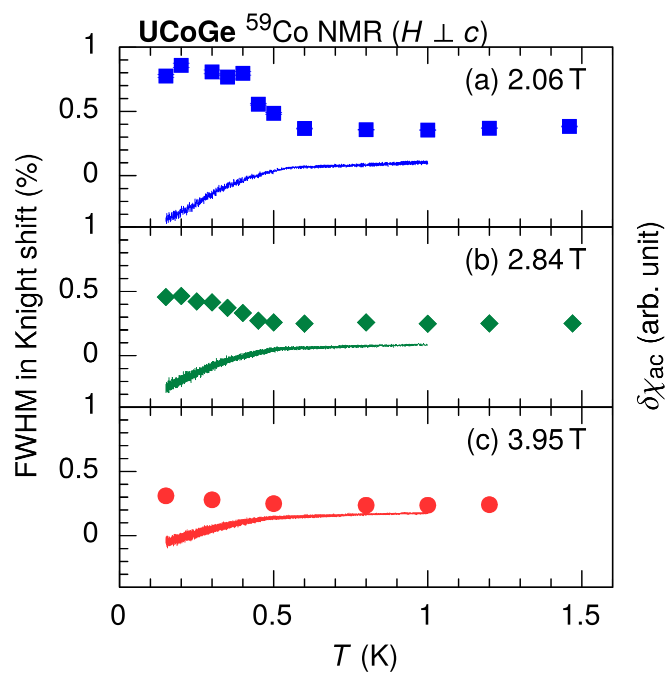

FIG. 6. Temperature dependence of the full width at half maximum (FWHM) of ^{59}Co NMR spectra at the central line represented in terms of the Knight-shift distribution. The ac susceptibilities are shown for comparison.

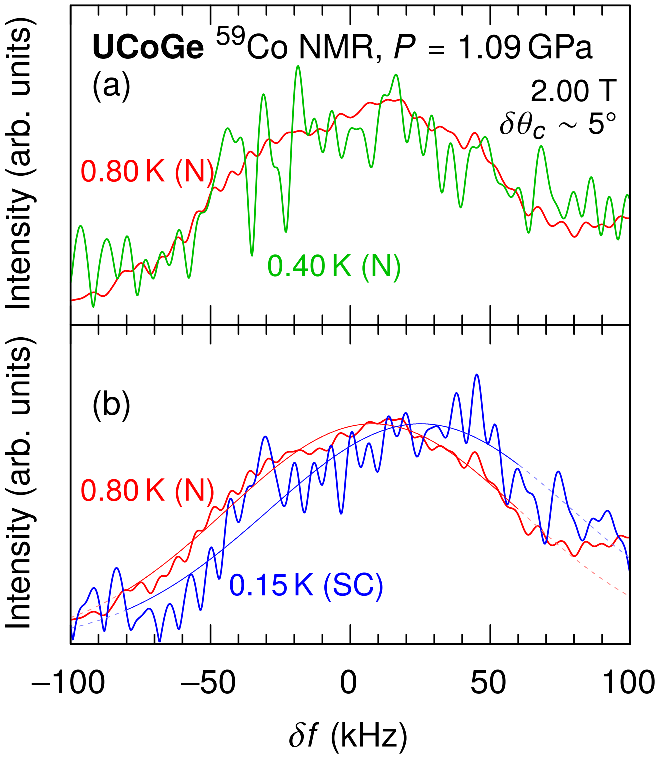

FIG. 7. ^{59}Co NMR spectra with 2.00-T field tilted to the c axis by $\delta\theta_c \sim 5°$ at 20.5 MHz. (a) The spectra in the normal state (0.80 and 0.40 K). No large shifts are observed in the normal state at these temperatures. (b) The spectra in the normal and SC states (0.80 and 0.15 K). The NMR spectrum slightly shifts to higher frequency in the SC state. The solid curves indicate results fitted with a Gaussian.

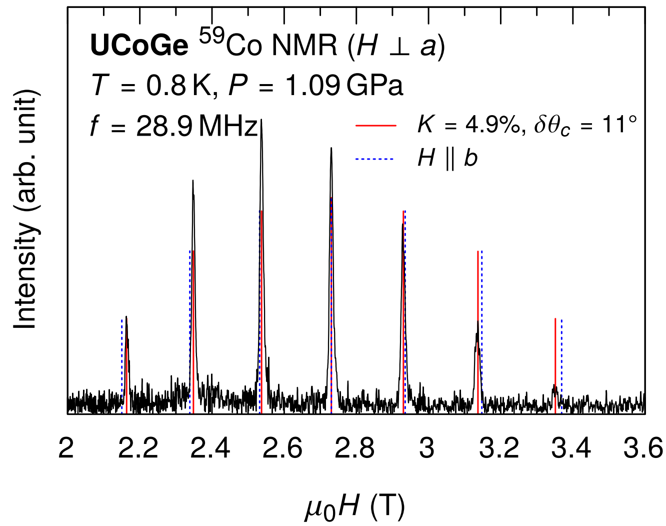

FIG. 8. Field-swept ^{59}Co NMR spectra with the field perpendicular to the a axis at 0.8 K (normal state). The vertical solid red lines indicate the best fit of the spectra with $\delta\theta_c = 11°$. The dashed blue lines indicate the calculated result with $H \parallel b$.

of 2–4 T are of the order of 10^{-1}%; this change corresponds to $\sim 10^{-1}\chi_{\rm spin}$. The increasing Knight shift is also of the order of 10^{-1}%.

It is not trivial that the Knight shift increases in the SC state in the spin-triplet pairing. A possible origin of the increase in spin susceptibility is the sharp density of states (DOS) around the Fermi energy [30]. If the DOS has a large slope at $E_{\rm F}$, a larger energy gain is obtained in the SC state, and the spin-triplet Cooper pairs have additional spin polarization. Although the details of the DOS in the normal state are not fully understood in UCoGe, a peak structure around $E_{\rm F}$ was revealed by a photoemission spectrum [31], and this could lead to redistribution of the Cooper pair spins. This effect would be most prominent when the field is along the easy axis (c axis), consistent with the experimental results. Another possibility is that the metamagnetism occurs coincident with the SC transition when the field has the c-axis component. A metamagnetic behavior at low temperatures is predicted using the theory of the quantum itinerant ferromagnetism [32]. This effect would also be anisotropic and sensitive to the c-axis field. We note that an increase of the Knight shift below 1 K was also observed in the FM SC state of UCoGe in the present sample at ambient pressure [10]; however, this anomaly started to occur at a temperature far above $T_{\rm SC}$. Such an increase in the Knight shift in the normal state was not observed in the PM state under pressure. Thus, we attribute this anomaly to the existence of the FM phase.

The decreasing Knight shift in the SC state is interpreted as the (partial) pinning of the $\boldsymbol{d}$ vector in the case of spin-triplet pairing. This result is in contrast to the case of the FM SC state, in which no decrease of the Knight shift was

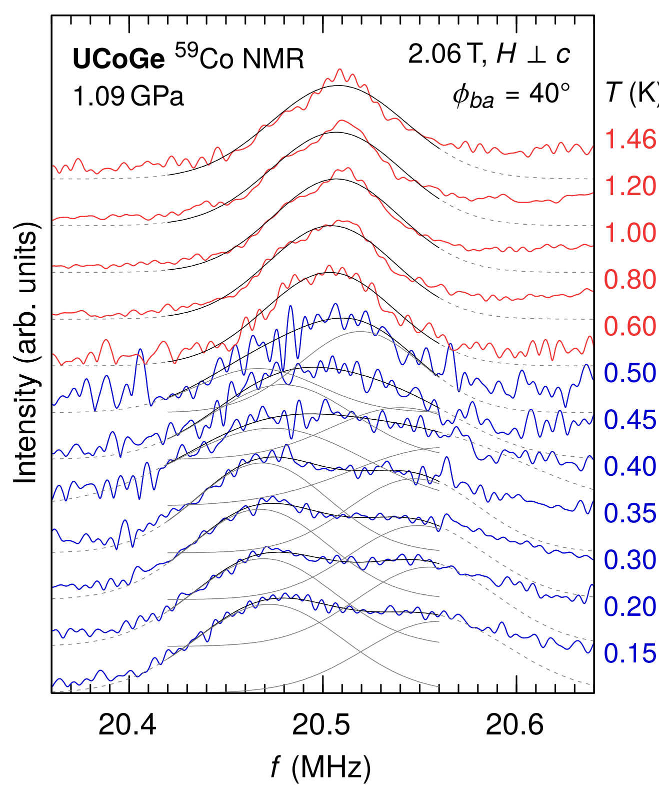

FIG. 9. ^{59}Co NMR spectra with the field perpendicular to the c axis, analyzed by two Gaussians in the SC state. The experimental data are the same as the ones shown in Fig. 4(a). The solid black lines indicate the best fit with the Gaussian(s). The solid gray lines indicate the partial components of two-component fitting.

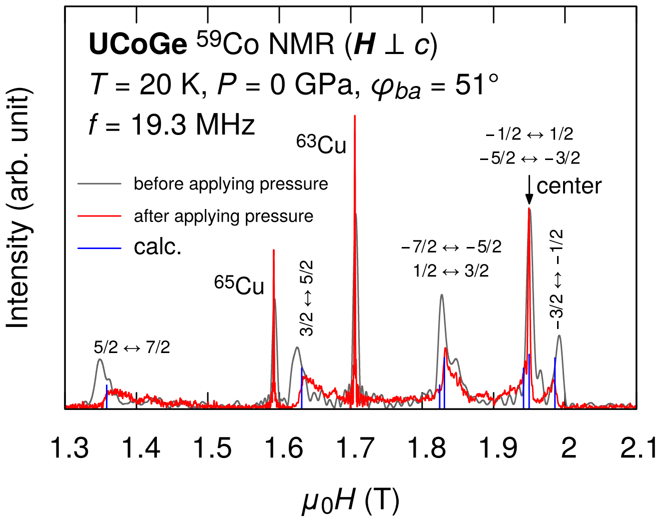

FIG. 10. Field-swept ^{59}Co NMR spectra at 0 GPa at 20 K at the PM state before (gray) and after (red) applying pressure. The vertical blue line is the best fit of the spectra with $\phi_{ba} = 51^\circ$.

detected within the experimental resolution when $H \parallel a$ and b [10]. The almost constant spin susceptibility in the FM SC state in these directions is attributed to the presence of the exchange field, which weakens the Pauli-PM effect [20]. It should be noted that exchange field is absent in the PM SC state under pressure. This suggests that the $\boldsymbol{d}$ vector would be perpendicular to the b axis in the ab plane and presumably parallel to the a axis, at least in the high-field region because the small spin susceptibility along the a axis [26] is in the favor of $\boldsymbol{d} \parallel a$. Thus, it is crucial to reveal whether the $\boldsymbol{d}$ vector rotates with the field parallel to the ab plane in the PM SC state. The field dependence of the broadening of the NMR spectra in the SC state implies that the decreasing component becomes weaker at higher field. This result may originate from the rotation of the $\boldsymbol{d}$ vector by the field. However, it is difficult to distinguish whether this field dependence is mainly ascribed to the gradual rotation of the $\boldsymbol{d}$ vector or the quasiparticle excitation from the present result. Because the possibility of multigap superconductivity is noted from thermal transport measurements in the FM SC state [33], it is inappropriate to speculate on the quasiparticle excitation based on a simple SC gap model. To evaluate the quasiparticle contribution accurately, it is important to reveal the SC gap structure under the magnetic field through, for instance, specific heat measurements, and future experiments may reveal the details of the $\boldsymbol{d}$-vector rotation.

Another interesting problem concerning the Knight-shift decrease is the decreasing amount of the Knight shift in the SC state, which is somewhat smaller than the simple estimate in the case of perfect pinning. If the $\boldsymbol{d}$-vector rotation occurs for a field much smaller than 2 T, the Knight shift becomes (almost) constant below T_{SC}. The existence of a residual DOS in the present sample may cause the reduction of the decreasing χ_{spin} as observed in $1/T_1$ without a field [22]. We also note that the value of K_{orb} is assumed to be unchanged by pressure when the spin part of the Knight shift is estimated. If K_{orb} changes under pressure, this could cause overestimation of the spin part. A more intrinsic reason for the small decrease of the Knight shift is also anticipated in heavy-fermion spin-triplet superconductors: it was reported that the Knight-shift decrease is much smaller than the expected χ_{spin} in UPt_3 even if the $\boldsymbol{d}$ vector is believed to be pinned along the field direction [34]. At present, this phenomenon remains unexplained.

IV. CONCLUSION

We demonstrated that the ^{59}Co NMR spectra are hardly shifted but become broader in the SC state of the single-crystalline UCoGe under a pressure of 1.09 GPa in the PM state when the field is perpendicular to the c axis. This result indicates that the spin-triplet pairing is realized in the PM state. The small decreasing component suggests the pinning of the $\boldsymbol{d}$ vector in the ab plane. Furthermore, the magnetic field dependence implies that the pinning is not so strong that the $\boldsymbol{d}$ vector can rotate to avoid the Pauli-PM effect; this is consistent with the large H_{c2} perpendicular to the c axis. An increasing Knight shift was also detected, arising from the sample region with a tilted field to the c axis. These anomalies cannot be interpreted by a spin-singlet pairing at all but are indicative of spin-triplet pairing.

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APPENDIX A: DETAILS OF THE SAMPLE TILTING

We estimate how much the sample is tilted inside the cell. Figure 8 shows the NMR spectrum with $H \perp a$. The field was aligned by monitoring the splitting of the central line: if the field is not parallel to the bc plane, the central peak splits into two lines owing to the low-symmetry nature of the Co site. This field direction is close to the b axis but is tilted toward the c axis due to the sample misalignment. The dashed blue lines indicate the calculated result with $H \parallel b$ and do not fit with the experimental result. The best fit was obtained when the field was tilted by $\delta\theta_c \sim 11^\circ$ from the b axis to the c axis, as shown by the solid red vertical lines. Then, it was revealed that the c axis of the sample is tilted by 17° from the vertical direction from this result combined with the field angle with $H \perp c$. This information was used to rotate the field by $\delta\theta_c \sim 5^\circ$ toward the c axis. There are two possible directions with $\delta\theta_c \sim 5^\circ$, and the tilted field is closer to the b axis than the original direction with $H \perp c$, as shown in Fig. 2(c).

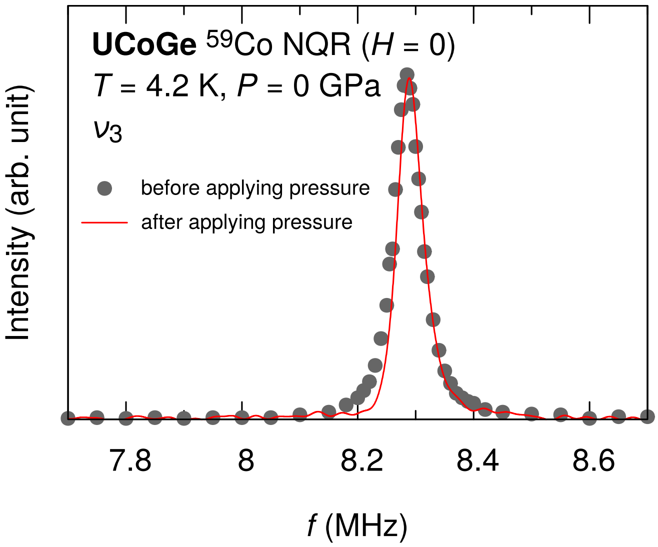

FIG. 11. ^{59}Co NQR spectra without field at 0 GPa at 4.2 K (the PM state) before (gray circles) and after (red line) applying pressure.

APPENDIX B: TWO-COMPONENT KNIGHT SHIFTS AND THEIR ORIGIN

The NMR spectra at 2.06 T were also analyzed based on the two-component Gaussian. The spectra and the fitting results are shown in Fig. 9. This analysis indicates that both the increasing and decreasing parts exist in the SC state at 2.06 T. They are on the order of $\Delta K \sim 0.1\%$, which is clear but much smaller than the spin part of the Knight shift $K_{\rm spin} \simeq 1.1\%$. We could not extract two components separately at higher fields owing to the smaller splitting.

It is not trivial that the ^{59}Co NMR Knight shift has two components in the SC state in the single-crystalline sample. We consider that this anomaly is due to the sample distortion by the pressure based on the following experimental results.

Field-swept NMR spectra were measured at ambient pressure in the PM state at 20 K before and after applying the pressure, as shown in Fig. 10. The field was applied perpendicular to the c axis and was tilted 51° away from the b axis to the a axis. This field direction differs from the one under the pressure measurement due to inevitable tilting of the sample inside the pressure cell (see Fig. 2). We found that the satellite peaks were broader after applying the pressure than before applying the pressure, while the central peaks did not broaden. Because the satellite peaks are more sensitive to the change in the EFG tensor, this broadening implies inhomogeneous EFG with respect to the field direction; more concretely, the quadrupole frequency ν_Q and/or the field angle with respect to the crystal distributes even after removing the pressure. However, the NQR spectrum at zero field at 4.2 K (PM state) did not broaden within the experimental error, as shown in Fig. 11. This result means that ν_Q and its distribution did not change after applying the pressure. Therefore, it is likely that the sample is distorted and the field angle distributes with respect to the crystal. We consider that this distortion was induced by applying pressure and existed at 1.09 GPa.

[1] A. J. Leggett, Rev. Mod. Phys. **47**, 331 (1975).
[2] H. Tou, Y. Kitaoka, K. Asayama, N. Kimura, Y. Ōnuki, E. Yamamoto, and K. Maezawa, Phys. Rev. Lett. **77**, 1374 (1996).
[3] K. Ishida, H. Mukuda, Y. Kitaoka, K. Asayama, Z. Q. Mao, Y. Mori, and Y. Maeno, Nature (London) **396**, 658 (1998).
[4] A. P. Mackenzie and Y. Maeno, Rev. Mod. Phys. **75**, 657 (2003).
[5] Y. Maeno, S. Kittaka, T. Nomura, S. Yonezawa, and K. Ishida, J. Phys. Soc. Jpn. **81**, 011009 (2012).
[6] A. P. Mackenzie, T. Scaffidi, C. W. Hicks, and Y. Maeno, npj Quantum Mater. **2**, 40 (2017).
[7] D. Aoki and J. Flouquet, J. Phys. Soc. Jpn. **81**, 011003 (2012).
[8] D. Aoki and J. Flouquet, J. Phys. Soc. Jpn. **83**, 061011 (2014).
[9] A. D. Huxley, S. J. C. Yates, F. Lévy, and I. Sheikin, J. Phys. Soc. Jpn. **76**, 051011 (2007).
[10] T. Hattori, Y. Ihara, K. Karube, D. Sugimoto, K. Ishida, K. Deguchi, N. K. Sato, and T. Yamamura, J. Phys. Soc. Jpn. **83**, 061012 (2014).
[11] A. D. Huxley, Phys. C (Amsterdam, Neth.) **514**, 368 (2015).
[12] T. Hattori, Y. Ihara, Y. Nakai, K. Ishida, Y. Tada, S. Fujimoto, N. Kawakami, E. Osaki, K. Deguchi, N. K. Sato, and I. Satoh, Phys. Rev. Lett. **108**, 066403 (2012).
[13] B. Wu, G. Bastien, M. Taupin, C. Paulsen, L. Howald, D. Aoki, and J.-P. Brison, Nat. Commun. **8**, 14480 (2017).
[14] Y. Tokunaga, D. Aoki, H. Mayaffre, S. Krämer, M.-H. Julien, C. Berthier, M. Horvatić, H. Sakai, S. Kambe, and S. Araki, Phys. Rev. Lett. **114**, 216401 (2015).
[15] D. Braithwaite, D. Aoki, J.-P. Brison, J. Flouquet, G. Knebel, A. Nakamura, and A. Pourret, Phys. Rev. Lett. **120**, 037001 (2018).
[16] E. Hassinger, D. Aoki, G. Knebel, and J. Flouquet, J. Phys. Soc. Jpn. **77**, 073703 (2008).
[17] E. Slooten, T. Naka, A. Gasparini, Y. K. Huang, and A. de Visser, Phys. Rev. Lett. **103**, 097003 (2009).
[18] G. Bastien, D. Braithwaite, D. Aoki, G. Knebel, and J. Flouquet, Phys. Rev. B **94**, 125110 (2016).
[19] N. T. Huy, D. E. de Nijs, Y. K. Huang, and A. de Visser, Phys. Rev. Lett. **100**, 077002 (2008).
[20] V. P. Mineev, Phys. Rev. B **81**, 180504(R) (2010).
[21] Y. Tada, S. Takayoshi, and S. Fujimoto, Phys. Rev. B **93**, 174512 (2016).
[22] M. Manago, S. Kitagawa, K. Ishida, K. Deguchi, N. K. Sato, and T. Yamamura, Phys. Rev. B **99**, 020506(R) (2019).
[23] F. Canepa, P. Manfrinetti, M. Pani, and A. Palenzona, J. Alloys Compd. **234**, 225 (1996).
[24] Y. Ihara, T. Hattori, K. Ishida, Y. Nakai, E. Osaki, K. Deguchi, N. K. Sato, and I. Satoh, Phys. Rev. Lett. **105**, 206403 (2010).
[25] T. Ohta, T. Hattori, K. Ishida, Y. Nakai, E. Osaki, K. Deguchi, N. K. Sato, and I. Satoh, J. Phys. Soc. Jpn. **79**, 023707 (2010).
[26] M. Manago, K. Ishida, and D. Aoki, Phys. Rev. B **97**, 075130 (2018).

[27] H. Tou, K. Ishida, and Y. Kitaoka, J. Phys. Soc. Jpn. **74**, 1245 (2005).
[28] K. Deguchi, E. Osaki, S. Ban, N. Tamura, Y. Simura, T. Sakakibara, I. Satoh, and N. K. Sato, J. Phys. Soc. Jpn. **79**, 083708 (2010).
[29] D. Aoki, T. D. Matsuda, V. Taufour, E. Hassinger, G. Knebel, and J. Flouquet, J. Phys. Soc. Jpn. **78**, 113709 (2009).
[30] K. Miyake, J. Phys. Soc. Jpn. **83**, 053701 (2014).
[31] S.-i. Fujimori, T. Ohkochi, I. Kawasaki, A. Yasui, Y. Takeda, T. Okane, Y. Saitoh, A. Fujimori, H. Yamagami, Y. Haga, E. Yamamoto, and Y. Ōnuki, Phys. Rev. B **91**, 174503 (2015).
[32] M. Brando, D. Belitz, F. M. Grosche, and T. R. Kirkpatrick, Rev. Mod. Phys. **88**, 025006 (2016).
[33] M. Taupin, L. Howald, D. Aoki, and J.-P. Brison, Phys. Rev. B **90**, 180501(R) (2014).
[34] H. Tou, Y. Kitaoka, K. Ishida, K. Asayama, N. Kimura, Y. Ōnuki, E. Yamamoto, Y. Haga, and K. Maezawa, Phys. Rev. Lett. **80**, 3129 (1998).