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Superconductivity Induced by Longitudinal Ferromagnetic Fluctuations in UCoGe

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From detailed angle-resolved NMR and Meissner measurements on a ferromagnetic (FM) superconductor UCoGe (TSC ~ 2.5 K and TC ~ 0.6 K), we show that superconductivity in UCoGe is tightly coupled with longitudinal FM spin fluctuations along the c axis. We found that magnetic fields along the c axis (H || c) strongly suppress the FM fluctuations and that the superconductivity is observed in the limited magnetic-field region where the longitudinal FM spin fluctuations are active. These results, combined with model calculations, strongly suggest that the longitudinal FM spin fluctuations tuned by H || c induce the unique spin-triplet superconductivity in UCoGe. This is the first clear example that FM fluctuations are intimately related with superconductivity.

The discovery of superconductivity in ferromagnetic (FM) UGe2 opened up a new paradigm of superconductivity [1,2], since most unconventional superconductivity has been discovered in the vicinity of an antiferromagnetic phase [3]. From the theoretical point of view, in an itinerant FM superconductor with the presence of a large energy splitting between the majority and minority spin Fermi surfaces, exotic spin-triplet superconductivity is anticipated, in which pairing is between parallel spins within each spin Fermi surface. In addition, it has been argued that critical FM fluctuations near a quantum phase transition could mediate spin-triplet superconductivity [4]. However, there have been no experimental results indicating a relationship between FM fluctuations and superconductivity.

Among the FM superconductors discovered so far, UCoGe is one of the most readily explored experimentally because of its high superconducting (SC) transition temperature (TSC) and low Curie temperature (TC) at ambient pressure [5]. Microscopic measurements have shown that superconductivity occurs within the FM region, resulting in the microscopic coexistence of ferromagnetism and superconductivity [6,7]. Studies of the SC upper critical field (Hc2) and its angle dependence along each crystalline axis have reported remarkable enigmatic behavior [8,9]: superconductivity survives far beyond the Pauli-limiting field along the a and b axes, whereas Hc2 for fields along the c direction (Hc2) is as small as 0.5 T. Colossal Hc2 for fields along the a and b axes seems to suggest spin-triplet pairing. In addition, a steep angle dependence of Hc2 was reported when the field was tilted slightly from the a axis toward the c axis [9]. The observed characteristic Hc2 behavior is one of the mysterious features of SC UCoGe, and its origin can be related to the mechanism of the superconductivity.

Unlike the three-dimensional crystal structure, magnetic properties are strongly anisotropic [8]. The magnetization has Ising-like anisotropy, with the c axis as a magnetic easy axis, and direction-dependent nuclear-spin lattice relaxation rate (1/T1) measurements on a single crystalline sample have revealed the magnetic fluctuations in UCoGe to be Ising-type FM ones along the c axis (longitudinal FM spin fluctuations) [10]. Here, we report from precise angle-resolved 1/T1 and Hc2 measurements how the longitudinal FM spin fluctuations are sensitively affected by the fields along the c axis (Hc) and are linked with the superconductivity. In addition, with the aid of model calculations, we unveil the role of the FM fluctuations as pairing glue in this compound, concomitantly resolving the abovementioned puzzle of Hc2.

A single crystal of UCoGe was grown by the Czochralski pulling method in a tetra-arc furnace under high-purity argon. A 1.65 × 1.65 × 1.89 mm3 sample with a mass of 55.8 mg was cut by spark erosion from the single crystalline ingot. The FM transition temperature TC was evaluated to be 2.55 ± 0.1 K from the Arrot plots, and onset and midpoint SC transition temperatures were determined from ac susceptibility as 0.70 and 0.57 K, respectively. Clear anomalies in the specific heat were observed at TSC and Tc, confirming that two anomalies are the bulk transitions. Resistivity along each direction was measured and is shown in Fig. 1. The sample showed a large residual resistivity ratio of approximately 30 along the b axis. The temperature dependence below 2 K is approximately expressed as ρ(T) = ρ0 + AT2, as shown in the inset. From the anisotropy of the A coefficient, the mass anisotropy is estimated as m*c/m*b ≈ 1.65 using a relation of m* = A. It is worth noting that a huge ratio of Hc2 to

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The angle dependence of $1/T_1$ is measured at the central peak in each spectrum. The recovery curves $R(t) = 1 - m(t)/m(\infty)$ of the nuclear magnetization $m(t)$, which is the nuclear magnetization at a time $t$ after a saturation pulse, can be fitted by the theoretical function for $I = 7/2$ with a single component throughout the measured field and temperature range. Thus, the electronic state is considered to be homogeneous in the whole region, and reliable $1/T_1$ values were obtained.

As shown in Fig. 2(a), $1/T_1$ along the $a$ and $b$ axes, which is much larger than $1/T_1$ along the $c$ axis due to the Ising-type anisotropy, shows a broad peak around 4 K due to the FM critical fluctuations. That the peak temperature is slightly higher than $T_{\text{Curie}}$ is conceivably due to a slight misalignment, since the peak is suppressed and shifts to higher temperatures when the external field is inclined by 3° away from the $b$ axis. The anisotropy of $1/T_1$ is largest around the peak temperature and extremely sensitive to the field angle. Figure 2(b) shows the angle dependence of $1/T_1(\theta)$ measured at 20, 4.2, and 1.7 K. For magnetic fields in the $bc$ plane, $1/T_1(\theta)$ is expressed as

$$\frac{1}{T_1(\theta)} = \frac{1}{T_1} \cos^2 \theta + \frac{1}{T_1} \sin^2 \theta.$$  \hspace{1cm} (2)

This equation can fully explain the smooth variation at 20 K but not the sharp angle dependence observed at 4.2 and 1.7 K, which shows a cusp centered at $\theta = 0^\circ$. The steep angle dependence of $1/T_1(\theta)$ is a characteristic feature of the FM fluctuations at low temperatures.

To further examine this behavior, the angle dependence of $1/T_1$ was measured at 1.7 K under three different applied fields ($\mu_0H \gg 0.28$ T). Although the cusp in $1/T_1$ versus angle becomes sharper with increasing $H$, as
A crucial question is whether such a characteristic feature of the FM fluctuations is related to superconductivity in UCoGe. To answer this question, we investigated the angle-dependent SC Meissner signal at 85 mK with high-frequency ac magnetic susceptibility measurements by observing the tuning frequency of the NMR circuit. When the single-crystal UCoGe undergoes a SC phase transition, $\chi_{\text{bulk}}$ becomes negative due to the Meissner effect and thus the frequency increases in the SC state. A shift of the tuning frequency was monitored as a function of $\theta$. The angular dependence of $H_{c2}$ in the $ac$ plane, which is determined from the onset of the Meissner signal, is shown in the inset of Fig. 4, since unusual enhancement of $H_{c2}$ along the $b$ axis was reported in a field larger than 4 T [9]. The $H_{c2}$ enhancement is another interesting phenomenon in UCoGe but beyond the scope of this Letter. The angular dependence of $H_{c2}$ is qualitatively the same as in a previous report [9], but the values are different by a factor of 3 from each other, as shown in the main panel of Fig. 4. As pointed out above, this steep angle dependence of $H_{c2}$ cannot be explained at all with the anisotropic Ginzburg-Landau formula, even if the field dependence of $m^*$ is taken into account. In contrast, if $H_{c2}$ is plotted against $H^c$, as shown in Fig. 3(c), we notice that superconductivity

\begin{align}
\langle \delta H^c \rangle^2 \propto \frac{1}{\cos^2 \theta} \left( \frac{1}{T_1} \frac{1 + \sin^2 \theta}{2} \right). \tag{3}
\end{align}

Figure 3(c) plots $\langle \delta H^c \rangle^2$ at 1.7 K against $H^c$, exhibiting a strong suppression approximately as $H^{-1/2}$, as shown by the broken line in the inset. It should be emphasized that $\langle \delta H^c \rangle^2$ at 20 K is independent of $H^c$, since the angle dependence of $1/T_1$ is well-fitted by Eq. (2); the strong suppression of $\langle \delta H^c \rangle^2$ by $H^c$ is observed only at low temperatures, where FM spin fluctuations are enhanced.
on the results in Fig. 3(c), we postulate that

\[ H_c \approx C_23 \]

where \( C_23 \) is approximately the Fermi velocity. Based on the results in Fig. 3(c), we postulate that \( \delta(H^F) \approx (1 + c\sqrt{H^F/H_c^0}) \), with a magnetic field \( H_c^0 \approx 1 \text{T} \) and a parameter \( c \approx O(1) \). It is worth noting that this form for \( \delta(H^F) \) is quite different from the ordinary mean-field-like dependence but that an anomalous \( \sqrt{H^F} \) dependence is a likely consequence of the semi-metallic-like 5f-band structure in UCoGe. Two spin-triplet SC states coexisting with ferromagnetism have been proposed by Mineev, of the form \( d \sim (a_1k_x + ia_2k_y, a_3k_y + ia_4k_x, 0) \) near the \( \Gamma \) point corresponding to point node symmetry (A state) and \( d \sim (b_1k_x + ib_2k_y, b_3k_y + ib_4k_x, 0) \) corresponding to horizontal line node symmetry (B state), where \( a_i \) and \( b_i \) are coefficients [12]. We calculated the angle dependence of \( H_c2 \) by solving the Eliashberg equation for each of the SC states, and the results agree well with the experiments when we use \( c = 5 \) for the A state, as shown with the red curve in the main panel of Fig. 4.

The temperature dependence of \( H_c2 \) along the \( a \) and \( c \) axes was calculated based on the A state with \( c = 1, 3 \), and 5, and the results are shown in Fig. 5. It is found that the qualitative behaviors of \( H_c2 \) are independent of the value of \( c \). Interestingly, the similar anisotropic \( H_c2 \) behavior with the enhancement of \( H_c2 \) along the \( a \) axis at low temperatures was actually observed [9]. These agreements strongly support the scenario that the longitudinal FM spin fluctuations in UCoGe mediate the spin-triplet superconductivity with the A-state symmetry, since the A state can explain the experimental observation consistently but the B state cannot reproduce the experimental results. We stress that the unusually strong angular dependence of \( H_c2 \) is extremely difficult to explain with a mean-field-like \( \delta(H^F) \) dependence but may be readily modeled with that deduced from the present experiment.

In conclusion, we demonstrate that a huge anisotropic ratio of \( H_c2 \) cannot be explained by the anisotropic Ginzberg-Landau model, since the conductivity of UCoGe is rather 3D. We found from the angle-resolved NMR measurements that the magnetic fields along the \( c \)-axis \( H_c \) strongly suppress the critical FM fluctuations with Ising anisotropy and that their suppression is intimately related to the unusual anisotropic properties of the superconductivity. The theoretical calculations based on the experimentally obtained \( \delta(H^F) \) dependence explain the strong suppression of \( H_c2 \) along the \( c \) axis. Our experimental results and theoretical analysis strongly suggest that the critical FM fluctuations with Ising anisotropy mediate the spin-triplet superconductivity in UCoGe.

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