Evidence for Spin Singlet Pairing with Strong Uniaxial Anisotropy in URu_2Si_2 Using Nuclear Magnetic Resonance

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In order to identify the spin contribution to superconducting pairing compatible with the so-called "hidden order", ^{29}Si nuclear magnetic resonance measurements have been performed using a high-quality single crystal of URu_2Si_2. A clear reduction of the ^{29}Si Knight shift in the superconducting state has been observed under a magnetic field applied along the crystalline c axis, corresponding to the magnetic easy axis. These results provide direct evidence for the formation of spin-singlet Cooper pairs. Consequently, results indicating a very tiny change of the in-plane Knight shift reported previously demonstrate extreme uniaxial anisotropy for the spin susceptibility in the hidden order state.

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The heavy-fermion compound URu_2Si_2 exhibits unconventional superconductivity below $T_{SC} \sim 1.4$ K, coexisting with an unidentified long-range electronic order, the so-called "hidden order" (HO), which sets in below $T_{HO} \sim 17.5$ K [1]. This HO state is suppressed by hydrostatic pressure, whereupon antiferromagnetic (AF) order occurs [2,3]. Interestingly, superconductivity does not appear in the AF state, and thus a close interplay between the HO and the superconductivity is expected.

As concerns the gap symmetry of the superconducting (SC) state, the angular dependence of thermal conductivity [4,5] and specific heat measurements [6,7] reveal the existence of two point nodes and a horizontal line node. For the spin part of Cooper pairs, on the other hand, the suppression of the upper critical field H_{c2} at lower temperatures [4,8,9] is regarded as a result of the Pauli paramagnetic effect for spin-singlet Cooper pairing. Considering the suggested time-reversal symmetry breaking via the polar Kerr effect [10], magnetic torque [11], and muon spin rotation (μSR) measurements [12], spin-singlet chiral d-wave superconductivity with a symmetry of $k_z(k_x + ik_y)$ has been proposed.

As for the spin state, however, there is no definitive evidence. Thus, the spin-singlet pairing is only deduced from the suppression of H_{c2} [4,8,9]. Recent quantum oscillation experiments [13] proposed a large uniaxial anisotropy for the spin susceptibility, which might also explain the H_{c2} anisotropy as a consequence of Pauli paramagnetic effects, but is still an indirect measure. The NMR Knight shift (K) is a direct measure of the spin susceptibility, and thus has been employed to determine the spin state in many heavy fermion superconductors [14]. In a spin-singlet SC state, the suppression of the spin susceptibility causes a decrease of K values in any field direction, while in a spin-triplet state K should be invariant along some directions, depending on the anisotropy of the SC d-vector. In unconventional uranium superconductors, several examples such as UPd_2Al_3 for singlet [15–18] and UPt_3 for triplet [19,20] NMR measurements have established a solid basis of evidence for spin states.

For URu_2Si_2, the ^{29}Si Knight shift has been measured by Kohori *et al.* [21] using powder samples in the SC state. However, due to the breadth of powder spectra, the experimental resolution was not sufficient to determine whether K decreased or not. Then, more recently, K measurements have been performed using a high-quality single crystal in external fields along the a axis. Extremely narrow NMR spectra obtained with the single crystal led to the determination of K values with considerable accuracy (on the order of 0.001%). However, still *no* observable change in K below T_{SC} was detected in the basal plane (perpendicular to the c axis) in our study [22]. This somewhat surprising result suggested either the possibility of spin-triplet Cooper pairing or an extremely small quasiparticle spin susceptibility for $H||a$. This result has motivated us to extend the K measurements to the c axis, although working in the SC state becomes extremely difficult in this field direction due to the small H_{c2} and low T_{SC}. However, we present here the results of such experiments, which provide the first direct evidence for the formation of spin-singlet Cooper pairs in URu_2Si_2. The experiments also document an extreme directional anisotropy of the spin susceptibility along the crystallographic a and c axes in the HO state.

A single-crystal specimen of URu_2Si_2 grown by the Czochralski pulling method in a tetra-arc furnace under high-purity argon was utilized. To enhance the sample quality, subsequent annealing in an evacuated quartz ampoule was performed. In addition, this single-crystal sample was prepared with the ^{29}Si isotope enriched to 53% in order to enhance the ^{29}Si NMR signal intensity, since the 4.7% natural abundance of NMR-active nuclei of ^{29}Si is

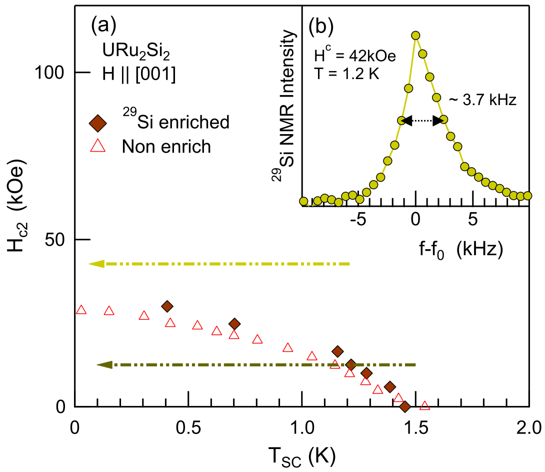

FIG. 1. (a) Temperature dependence of the upper critical field H_{c2} plotted for two single crystals URu_2Si_2: ^{29}Si-enriched (diamond) and nonenriched (triangle) [25], with external fields along the c axis. Temperature scans of ^{29}Si NMR spectra were performed under magnetic fields (42 and 12.5 kOe) as indicated by the horizontal two-dotted chain arrows. (b) ^{29}Si NMR spectrum under $H^c = 42$ kOe at $T = 1.2$ K. f_0 is the center frequency of the ^{29}Si spectrum. Full width at half maximum of the spectrum was about 3.7 kHz.

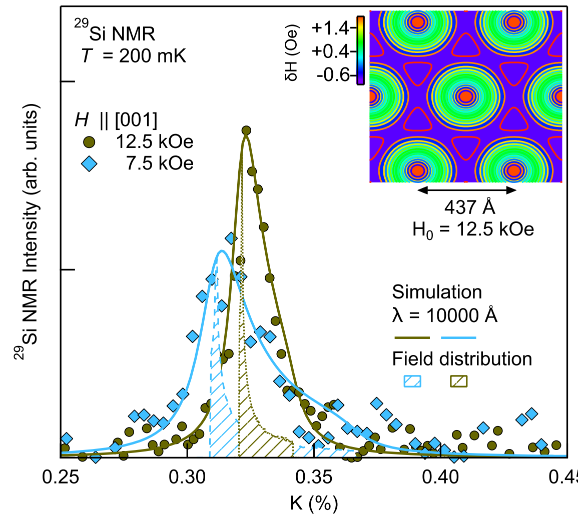

FIG. 2. ^{29}Si NMR spectra in the superconducting state with external fields $H^c = 7.5$ kOe and 12.5 kOe. The shaded area represents the field distribution (δH) due to the triangular vortex lattice, where the spectra are simulated with the field distribution convoluted with a Lorentzian broadening function. The inset shows the vortex lattice at applied field $H_0 = 12.5$ kOe.

inadequate. The excellent quality of crystals made by the same process has been confirmed in our ^{29}Si NMR studies already published [22–24].

NMR in the SC state was performed using a ^{3}He–^{4}He dilution refrigerator, where the single-crystal sample was immersed in the ^{3}He–^{4}He mixture to avoid rf heating. Since the crystal had a nearly perfect cylindrical shape (1.5 mm$\phi\perp$[100] $\times$ 2 mm $||$[100]), we aligned the crystal's [100] growth axis with the axis of the rf solenoid coil in order to obtain the highest NMR sensitivity and to minimize the distribution of rf fields. The direction of the external magnetic field was carefully adjusted using narrow [001] facets on the cylindrical sample, so that the [001] axis was exactly aligned with the magnetic field to within less than 1°. ^{29}Si NMR spectra were obtained by fast Fourier transform (FFT) of Hahn echo signals at a fixed frequency and magnetic field. The nuclear spin-lattice relaxation time T_1 was measured by the inversion-recovery method.

Figure 1(a) shows the upper critical field H_{c2} with a field along the c axis, which is determined via *in situ* ac susceptibility measurements carried out by tracking the resonant frequency of the NMR circuit. When the sample undergoes the SC phase transition, χ_{bulk} becomes negative owing to the Meissner shielding effect, leading to a change of inductance (L), and thus changing the resonant frequency of the LC series circuit with a constant capacitance (C). Nearly the same behavior of H_{c2} for the present sample as that for the nonenriched sample [25] confirms the sample quality [26] and the optimal magnetic field direction in this NMR setting. It should be noted that the superconductivity in URu_2Si_2 occurs only in an extremely clean limit [26,27].

A typical ^{29}Si NMR spectrum in the normal state with a field along the c axis is shown in Fig. 1(b). Full width at half maximum of the ^{29}Si NMR spectrum is about 3.7 kHz with a field of 42 kOe, which is nearly the same as that of previous NMR samples [22,24], and even that of a nonenriched sample [28], ensuring the high quality of the present sample. It also shows that the dipolar broadening of the NMR spectra due to ^{29}Si enrichment is negligible. ^{29}Si NMR measurements were performed at constant fields of $H^c = 12.5$ kOe and $H^c = 42$ kOe $> H_{c2}$. These temperature scans are indicated by the arrows in Fig. 1(a).

Figure 2 shows typical ^{29}Si NMR spectra of URu_2Si_2 in the SC state. The horizontal axis of Fig. 2 is plotted against a K scale converted from the relation $f = (\gamma_n/2\pi)[1 + K(T)]H^c$, where f is the frequency scale generated by the FFT of the echo signal, $\gamma_n/2\pi = 845.77$ Hz/Oe with external fields $H^c = 12.5$ and 7.5 kOe, respectively. In contrast to an almost symmetrical NMR spectrum at $H^c = 12.5$ kOe, the spectrum at lower field $H^c = 7.5$ kOe presents a slightly asymmetric shape. This is because the field distribution in the sample originates from Meissner shielding approximated by [29]

$$H(\boldsymbol{r}) = H\sum_{l,m}\frac{\exp[-\boldsymbol{G}_{l,m}^2\xi^2/2]}{1+\boldsymbol{G}_{l,m}^2\lambda^2}\exp[-i\boldsymbol{G}_{l,m}\cdot\boldsymbol{r}]. \quad (1)$$

Here, $\boldsymbol{G}_{l,m}$ is a reciprocal lattice vector of the vortex lattice, $\boldsymbol{r}$ is the position vector from the vortex core, λ is the penetration depth, ξ is the coherence length, and a triangular vortex lattice is assumed, as shown in the inset to Fig. 2. The experimental results are well reproduced by the field distribution (shaded area) convoluted with a

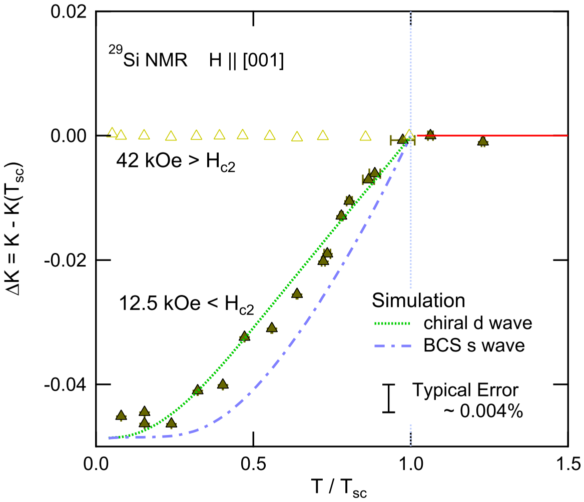

FIG. 3. Temperature dependence of differential ^{29}Si Knight shift: $\Delta K \equiv K - K(T_{SC})$ in both the normal and superconducting states. Temperatures on the horizontal scale are scaled to $T_{SC} = 1.2$ K at $H^c = 12.5$ kOe. The dotted (dot-dash) curve represents a simulation using the SC gap function of the chiral d-wave model (BCS s-wave model).

Lorentzian broadening function using the parameters $\lambda = 10000$ Å [30] and $\xi = 60$ Å. In general, higher fields can narrow such a field distribution by vortices, and in the case of $H^c = 12.5$ kOe, the field reduction is no more than $\delta H \sim 0.6$ Oe, as shown in the inset. This corresponds to about 0.005% on the K scale, which is rather smaller than the observed change of K discussed later. Therefore, the change of K below T_{SC} originates mainly from the change of electronic magnetic susceptibility.

Now we discuss the temperature dependence of the ^{29}Si Knight shift $K(T)$ as presented in Fig. 3. Values of $K(T)$ were determined by fitting ^{29}Si NMR spectra to a Lorentzian function, and the differential values $\Delta K(T) \equiv K(T) - K(T_{SC})$ under several fields are shown. Here $K(T_{SC})$ is the K value at $T_{SC} = 1.2$ K under $H^c = 12.5$ kOe. In the normal state above H_{c2}, no change in $K(T)$ is observed below $T = 1.2$ K. Thus, $K(T)$ is temperature independent for $H > H_{c2}$, indicating that a Fermi liquid state is realized at the lowest temperatures of the HO state, as discussed below. On the other hand, we observed a clear decrease of $\Delta K(T) \sim 0.05\%$ at $T = 100$ mK under low magnetic field well below H_{c2}. As already discussed, this decrease of K originates from the quenching of the spin susceptibility as Cooper pairs are being formed.

In order to quantify the decrease of $K(T)$, we derive the spin component of Knight shift $K_{\rm spin}$, which should decrease to zero when spin-singlet pairing occurs. Thus, $K_{\rm spin}$ can be estimated from the Sommerfeld coefficient $\gamma_{\rm el}$ obtained from specific heat measurements as

$$K_{\rm spin} = \frac{A_{\rm hf}}{N_A \mu_B} \chi^{\rm qp} = \frac{A_{\rm hf}}{N_A \mu_B} \frac{\gamma_{\rm el}(\mu_{\rm eff})^2}{\pi^2 k_B^2} R. \quad (2)$$

Here, N_A is Avogadro's number, μ_B is the Bohr magneton, $\chi^{\rm qp}$ is the quasiparticle susceptibility, $\mu_{\rm eff}$ is the effective moment, $k_{\rm B}$ is Boltzmann's constant, and R is the Wilson ratio. The details are given in Ref. [14] and in our previous report regarding the in-plane K for URu_2Si_2 [22]. In this way we deduce the estimate of $K^c_{\rm spin} = 0.057\%$. This estimate is nearly the same value as observed experimentally at the lowest temperature of 100 mK, suggesting that the experimental decrease of K corresponds to the quench of the quasiparticle spin susceptibility. Furthermore, temperature dependence of K in the SC state can be described using the Yosida function [31]. The dotted curve in Fig. 3 is simulated with the chiral d-wave model, whose gap symmetry of $k_z(k_x + ik_y)$ with a gap size of $2\Delta_0/k_B T_{\rm SC} = 3.52$ [14]. The residual density of states N_r relative to that for the normal state N_0 is set to $N_r/N_0 = 0.15$. It is noted that the fit to the experimental data is much better than that of the BCS full SC gap model shown as a dot-dash line in Fig. 3. However, this analysis does not indicate that the chiral d-wave model is a unique solution, since other SC gap functions with line or point nodes can also give a reasonable fit to the data.

In the case of spin-singlet pairing, spin susceptibility for any field direction should go to zero, so that $K_{\rm spin}$ for all directions should decrease. However, no change of in-plane K was detected in previous NMR measurements [22]. This indicates that the spin component of K along the magnetic hard axis $K^a_{\rm spin}$ is less than the experimental resolution of 0.002%. Considering that $K^c_{\rm spin}$ is about 0.057%, these results suggest a very strong Ising-type anisotropy for the spin component of susceptibility, thus $\chi^c_{\rm spin}/\chi^a_{\rm spin} > 25$. The hyperfine coupling constants are estimated to be fairly isotropic, i.e., $A^c_{\rm hf}/A^a_{\rm hf} \sim 1/2 \to 1$ [23,28,32–35], so that this cannot account for the ratio $K^c_{\rm spin}/K^a_{\rm spin} \gg 1$. This type of Ising anisotropy has been indicated by the results of several experiments. In order to explain the anisotropy of H_{c2}, taking the contributions from both the orbital and Pauli effects into account, $\chi^c_{\rm spin}/\chi^a_{\rm spin}$ was estimated to be at least ~60 [8]. On the other hand, the field-angle dependence of the effective g factor, which was estimated via quantum oscillation measurements, suggested much greater anisotropy $\chi^c_{\rm spin}/\chi^a_{\rm spin} > 1000$ [13]. Recent nonlinear magnetization measurements also show a clear Ising anisotropy of quasiparticles up to 60 K [36]. Therefore, our finding of an anisotropic K in the SC state can be naturally understood as a consequence of a very large Ising anisotropy in $\chi_{\rm spin}$. We should also note here that spin-triplet pairing with the $\boldsymbol{d}$ vector fixed along the c axis could also explain the behavior of $K(T)$ below $T_{\rm SC}$. Thus, K^c decreases while K^a is invariant. In this case, however, Cooper pairs would be formed with the spins aligned in the magnetic hard plane, which is extremely unlikely. Furthermore, recent nuclear relaxation rate $1/T_1$ measurements have revealed strong Ising-type magnetic fluctuations along the c axis [37],

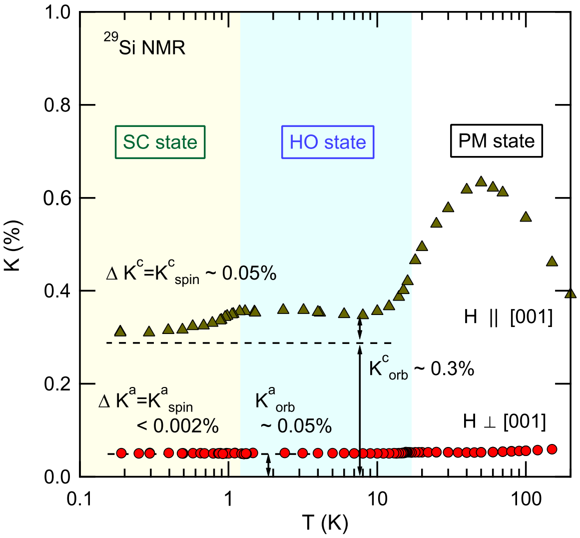

FIG. 4. Temperature dependence of [29]Si Knight shift with a field along each crystalline axis. The c-axis data below $T = 1.2$ K were taken with the present sample, while the others were taken with the sample used in Ref. [22]. The spin components of the Knight shift ($K^{a,c}_{\rm spin}$) estimated from ΔK, the reduction of K below $T_{\rm SC}$, are much smaller than the nearly temperature-independent orbital component ($K^{a,c}_{\rm orb}$).

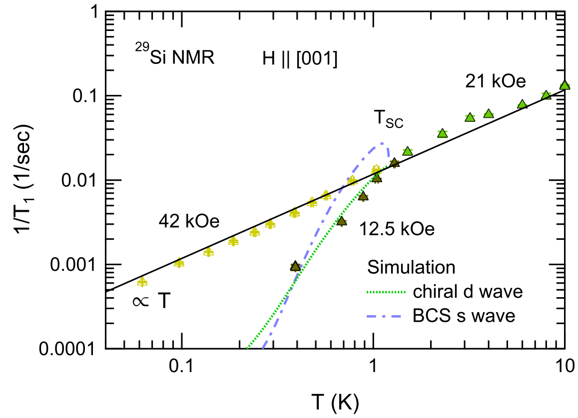

FIG. 5. Temperature dependence of nuclear spin-lattice relaxation rate $1/T_1$ with a field along the c axis. The data above $T = 1.5$ K were obtained with the previous sample [22]. $1/T_1$ was measured both in the normal state ($H^c = 42$ kOe) and in the superconducting state ($H^c = 12.5$ kOe). The solid line represents the Korringa law: $1/T_1 \propto T$. The dotted (dot-dash) line was calculated via a chiral d-wave (conventional s-wave) model with a SC gap of $2\Delta/k_B T_{\rm sc} = 3.52$.

consistent with the proposed anisotropic Knight shift ($K^c_{\rm spin} \gg K^a_{\rm spin}$). While Ising fluctuations such as these might stabilize spin-triplet pairing with spins aligned along the c axis [38,39], they would, however, be inconsistent with the present observation of a decrease in $K^c_{\rm spin}$.

To discuss such magnetic anisotropy further, $K(T)$ values in the whole temperature range are shown in Fig. 4. Above $T = 1.2$ K, K^c and in-plane K were taken with an earlier sample [22]. The temperature dependence of K in both the HO and paramagnetic (PM) states is in good accord with previous NMR data [21]. The decrease of $K^c_{\rm spin}$ in the SC state is only 1/7 of $K \sim 0.4\%$, suggesting that the orbital shift $K^c_{\rm orb}$ corresponding to, e.g., the Van Vleck susceptibility, is dominant in the HO state. Since $K^a_{\rm spin}$ at $T_{\rm SC}$ is estimated to be less than 0.002%, K in the basal plane originates mainly from the orbital component as well, i.e., $K^a_{\rm orb} \sim 0.05\%$. Here, the interesting problem is whether the strong reduction of $K^a_{\rm spin}$ relates to the HO or not, since high-resolution NMR detects a small decrease of $K^a \simeq 0.004\%$ below $T_{\rm HO}$ [23]. In addition, in-plane spin fluctuations from $5f$ electrons also mostly disappear below $T_{\rm HO}$, since fluctuations probed by $1/T_1$ are nearly the same as those from the reference compound $ThRu_2Si_2$ [37]. These results may suggest that the remaining spin fluctuations along the c axis in the HO state relate to the SC pairing mechanism. In order to examine this hypothesis, further experiments to investigate the spin anisotropy under hydrostatic pressure would be desirable. If antiferromagnetic order occurs instead of HO, a change of anisotropy might be expected.

Finally, we display the temperature dependence of $1/T_1$ in Fig. 5. Below $T = 1.2$ K, $1/T_1$ was measured both in the normal state ($H^c = 42$ kOe) and in the SC state ($H^c = 12.5$ kOe). $1/T_1$ results for the previous sample [22] above 1.5 K are also shown. In the normal state, $1/T_1 \propto T$ holds below $T = 10$ K to $T = 60$ mK, as indicated by the solid line; thus, the Korringa relation $1/T_1TK^2 = \text{const}$ is confirmed even at the lowest temperatures, which is characteristic of a Fermi liquid state. On the other hand, $1/T_1$ in the SC state shows a clear decrease below $T_{\rm SC}$, indicating the opening of the SC gap. No Hebel-Slichter peak as indicated by the dot-dash curve in Fig. 5 appears just below $T_{\rm SC}$, although it is known to be a characteristic of conventional s-wave superconductors [40]. Therefore, $1/T_1$ data below $T_{\rm SC}$ indicate anisotropic SC gap symmetry. In fact, the sharp drop of $1/T_1$ following $\sim T^3$ behavior can well be explained by the equivalent chiral d-wave model, as shown by the dotted curve in Fig 5 with the residual DOS $N_r/N_0 \sim 0.15$ used in the above Knight shift analysis.

In summary, we detect a clear reduction of the [29]Si Knight shift in the superconducting state of URu_2Si_2 along the magnetic easy (c) axis, while no change of in-plane Knight shift has been detected [22]. Our Knight shift measurements reveal that the superconducting state of URu_2Si_2 can be well understood as spin-singlet pairing with strong uniaxial spin anisotropy $\chi^c_{\rm spin}/\chi^a_{\rm spin} > 25$ in the HO state.

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