Al Knight-shift measurement in the superconducting state of UNi_2Al_3

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Abstract

We report ^{27}Al Knight-shift (^{27}K) measurement on a single-crystal UNi_2Al_3 that reveals a coexistence of superconductivity and a spin-density-wave (SDW) type of magnetic ordering (T_{SDW} = 4.5 K). The spin part of ^{27}K, $^{27}K_s$ does not change down to 50 mK across the superconducting (SC) transition temperature $T_c \sim 0.9$ K. In contrast with the isostructural compound UPd_2Al_3 ($T_c \sim 2$ K), which was identified to be a spin-singlet *d*-wave superconductor, the behavior of ^{27}K strongly supports that UNi_2Al_3 belongs to a class of spin-triplet SC pairing state superconductor.

Key words: heavy-fermion compound; UNi_2Al_3; NMR; Knight shift; spin-triplet superconductivity

In heavy-fermion superconductors, respective isostructural hexagonal compounds UPd_2Al_3 and UNi_2Al_3 are of special interest, since antiferromagnetic (AF) long-range order with T_N =14.3 and 4.5 K is found to coexist with the superconductivity with T_c ∼2 and 1 K, respectively[1,2]. In UPd_2Al_3, neutron-diffraction (ND) experiments[3] revealed a commensurate magnetic structure with a large ordered magnetic moment μ=0.85μ_B on the uranium atoms. On the other hand, ND[4] and muon spin rotation (μSR)[5] experiments revealed that UNi_2Al_3 shows a spin-density wave (SDW) type of ordering and has a tiny magnetic moment of $\sim 0.2\mu_B$ modulated in amplitude within the basal plane.

As for the superconducting (SC) properties in UPd_2Al_3, it was reported that the NMR Knight shift decreases below T_c irrespective of the direction of the magnetic field, and that the nuclear spin-lattice relaxation rate, $1/T_1$ obeys a T^3 law in the SC state. This behavior is consistent with a spin-singlet *d*-wave superconductivity with a line-node gap[6,7]. In contrast, the SC nature in UNi_2Al_3 has been studied only by a few experiments owing to the difficulty of preparing a SC single crystal. Concerning the SC gap structure in UNi_2Al_3, line-node gap was reported from T_1 measurements as in UPd_2Al_3[8]. However, Knight-shift measurements, which give crucial information about the symmetry of the pairs, have not yet been reported due to the lack of the high-quality single crystal.

In this paper, we report measurement of the ^{27}Al Knight shift (^{27}K) in UNi_2Al_3 that allows us to determine the spin state of the Cooper pairs. The NMR measurement was carried out using the high-quality single crystal with the residual resistivity ratio ∼ 40 and residual resistivity $\rho_0 \sim 3.6\mu\Omega$cm. A precise report

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of the measurement will be published elsewhere[9].

As for ^{27}Al($I = 5/2$)-NMR spectra in UNi_2Al_3, when a static field $\boldsymbol{H_0}$ is applied to the c axis ($\boldsymbol{c}$) ($\theta = 0°$), five sharp peaks are well split by the nuclear electric quadrupole interaction. Here θ is defined as the angle between $\boldsymbol{H_0}$ and $\boldsymbol{c}$, which is the principal axis of the electric field gradient.The point symmetry of Al sites in the hexagonal crystal structure of UNi_2Al_3 is not axially symmetric, so that the quadrupole splitting ν_a along $\boldsymbol{a}$ is no longer equal to the quadrupole splitting ν_b along $\boldsymbol{b}$. For $\boldsymbol{H_0} \parallel \boldsymbol{a}$ ($\boldsymbol{H_a}$) ($\theta = 90°$), there exist two inequivalent Al sites. For one third of the Al sites $\boldsymbol{H_0}$ is parallel to $\boldsymbol{a}$ (denoted as the $\phi = 0°$ site) and for two thirds of the Al sites, the angle between $\boldsymbol{H_0}$ and $\boldsymbol{a}$ is $\pm 60°$ (denoted as the $\phi = 60°$ site).

From the $K(T)$ vs $\chi(T)$ plots in the normal state, the hyperfine coupling constants $A_{\rm hf}$ at $\phi = 0°$ and $\phi = 60°$ sites are estimated to be 3.5 and 4.2 kOe/$\mu_{\rm B}$ respectively, and $K_{\rm orb}$ at both sites is negligibly small. $K_{\rm s}$ at $\phi = 0°$ and $\phi = 60°$ sites is derived as 0.32 % and 0.39 % at 10K, respectively.

As temperature is cooling down across $T_c(H) \sim$ 0.75K that was determined by the ac-susceptibility and T_1 measurements as indicated in Figs. 1(a) and 1(b), the $K_{\rm s}^{\phi=60°}(T)$ does not change as seen in Fig. 1(c). The same result was also obtained for $\boldsymbol{H_a} = 4887$ Oe. This invariance of the shift for $\boldsymbol{H_a}$ =2438 and 4887 Oe reveals that the Knight shift, and therefore the spin susceptibility, does not change at all on passing through $T_c(H)$.

Now, we compare the ^{27}Al shift results in UNi_2Al_3 with those in the isostructural compound UPd_2Al_3. The shift associated with itinerant heavy electrons in UPd_2Al_3 exhibits a distinct decrease $\Delta K_{ab} \sim -0.15\%$, and $\Delta K_c \sim -0.10\%$ in the SC state[6,10,11]. The large residual Knight shift that remains even at the lowest T was ascribed to the localized part of $5f$ electrons that order antiferromagnetically with an atomic-like value of the saturation moment, $\mu \sim 0.85\mu_B$. The ΔK in UPd_2Al_3 reveals that the spin susceptibility of the heavy itinerant electrons diminishes in the SC state due to the formation of spin-singlet Cooper pairs. The value of the Knight shift for these electrons was found to be $K_{\rm s} \sim 0.14\%$ using γ=150 mJ/mol K^2 just above T_c and $A_{\rm hf}$ ~3.5 kOe/μ_B [12]. Here the Fermi liquid relation $\chi = \gamma$ in units of $\hbar = \mu_B = k_B$=1, and the relation of $K = A_{\rm hf}\chi/N\mu_B$ between the shift and the susceptibility are used. If the same calculation is applied to UNi_2Al_3, $K_{\rm s} \sim 0.16\%$ is calculated, using γ=140 mJ/mol K^2 just above T_c[13] and $A_{\rm hf}^{\phi=60} = 4.2$ kOe/μ_B. The value of 0.15% that is the Knight-shift decrease in UPd_2Al_3 is apparantly greater than the experimental error ($\leq$ 1 kHz) and is larger enough to be resolved from the present measurements. The invariance of ^{27}K against the onset of superconductivity

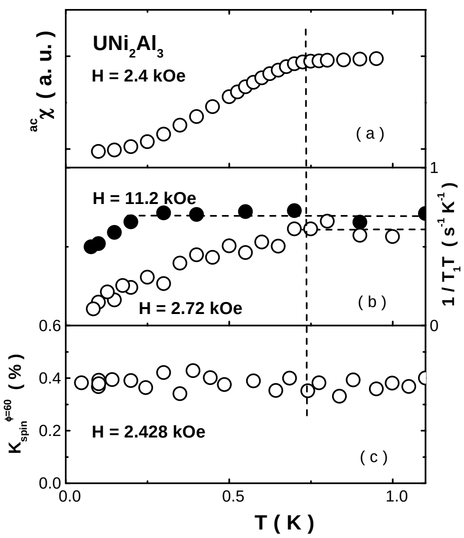

Fig. 1. T dependence of (a) ac susceptibility, (b) $1/T_1T$ for $\boldsymbol{a} \parallel \boldsymbol{H_0}$ =2.72 kOe (open circles), and 11.2 kOe (closed circles)[14], and (c) Knight shift at the ϕ=60° peak. The vertical dotted line indicates T_c(2.4 kOe).

suggests strongly that a spin-triplet pairing state is realized in UNi_2Al_3, with parallel spin pairing in the ab plane.

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References

[1] C. Geibel *et al.*, Z. Phys. B **83**, 305 (1991).

[2] C. Geibel *et al.*, Z. Phys. B **84**, 1 (1991).

[3] A. Krimmel *et al.*, Z. Phys. B **86**, 161 (1992).

[4] A. Schröder *et al.*, Phys. Rev. Lett. **72**, 136 (1994).

[5] Y. J. Uemura *et al.* Physica B **186-188**, 223 (1993).

[6] M. Kyogaku *et al.*, J. Phys. Soc. Jpn. **62**, 4016 (1993).

[7] R. Feyerherm *et al.*, Phys. Rev. Lett. **73** 1849 (1994).

[8] H. Tou *et al.*, Physica B **230-232**, 361 (1997).

[9] K. Ishida *et al.*, to be published in Phys. Rev. Lett.

[10] M. Kyogaku *et al.*, Physica B **186-188**, 285 (1993).

[11] Y.Kohori *et al.*, Solid State Comm. **95**, 121 (1995).

[12] H. Tou *et al.*, J. Phy. Soc. Jpn. **64**, 725 (1995).

[13] N. Aso *et al.*, Phys. Rev. B **61**, R11867 (2000).

[14] An unexpected decrease in $1/T_1T$ was observed below $\sim$ 0.2 K at H = 11.2 kOe, which is greater than $H_{c2} \sim 8$ kOe. The precise field dependence of $1/T_1T$ is now under investigation.