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Journal
Journal of the Korean Physical Society, 63(3)

ISSN
0374-4884

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Publication Date
2013-08-01

DOI
10.3938/jkps.63.341

Journal of the Korean Physical Society, Vol. 63, No. 3, August 2013, pp. 341∼344

Pressure Evolution of the Metamagnetic Transition in UCoAl As Measured Using ^{59}Co NMR

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(Received 31 May 2012, in final form 29 July 2012)

We have performed NMR measurements under pressure in UCoAl with a quantum critical endpoint of the metamagnetic transition from the paramagnetic phase to the ferromagnetic (FM) phase. ^{59}Co-NMR sensitively detects the evolution of the internal field caused by applying the external field. The metamagnetic field H_m increases with increasing pressure consistently with other experimental methods, accompanied by the suppression of the magnetization in the field-induced FM phase and the magnetization jump at H_m. The loss of the NMR signal on approaching the QCEP indicates the development of the spin fluctuations.

PACS numbers: 68.37.Ef, 82.20.-w, 68.43.-h
Keywords: Quantum Critical Endpoint, UCoAl, NMR
DOI: 10.3938/jkps.63.341

I. INTRODUCTION

The terminal point of a first-order phase transition at 0 K, that is, a quantum critical endpoint (QCEP), is a fascinating target as a new type of quantum critical point. In itinerant ferromagnets with a tricritical point (TCP), the metamagnetic transition from the paramagnetic (PM) phase to the ferromagnetic (FM) phase possesses the wing structure of a first-order transition in the temperature(T)–pressure(P)–magnetic field(H) phase diagram [1–5]. The first-order metamagnetic (PM–FM transition) transition at low temperatures encounters a critical end point (CEP) above which the transition changes to a crossover. The temperature of CEP (T_{CEP}) reaches 0 K at the QCEP of P_{QCEP} and H_{QCEP}. Among several candidates, UCoAl is a good example with the moderately wide wing for which we can easily tune pressure from the vicinity of TCP, which presumably is present under a small negative pressure, to the PM region exceeding the QCEP [4].

UCoAl crystallizes in a ZrNiAl-type hexagonal structure stacked by U-Co(1) and Co(2)-Al layers [6]. It is a paramagnet, but a uniaxial pressure effect and chemical substitution have revealed that it is located in the vicinity of a FM critical point [7–9]. The FM state at zero field is thought to be already suppressed at a negative pressure of ~ -0.2 GPa [9]. A first-order metamagnetic

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transition to the FM state with $\sim 0.3\ \mu_B$/U occurs at $H_m \sim 0.7$ T for $H \parallel c$ axis at low temperatures [6]. The FM moment mainly originates from U-5f electrons [10]. The transition changes to a crossover at temperatures above $T_{\rm CEP} \sim 11 - 13$ K [4,9,11]. Strong longitudinal magnetic fluctuations are observed in the vicinity of the CEP [11]. The $T_{\rm CEP}$ is suggested to reach the QCEP at 0 K under $P_{\rm QCEP} \sim 1.5$ GPa and $H_{\rm QCEP} \sim 7$ T [4].

II. EXPERIMENTS AND DISCUSSION

The single crystal was grown using the Czochralski method in a tetra-arc furnace [12]. Powdered single crystals were used for NMR measurements at ambient pressure with an orientation $H \parallel c$. We used a single crystal for NMR measurements under pressure with the field applied along $H \parallel c$. Pressure was applied by using a piston-cylinder cell used Daphne7474 as a pressure-transmitting medium. [13] The pressure at low temperatures was estimated by using a Pb manometer. NMR was performed using a standard spin-echo method. Magnetization (M) measurements was performed using a Magnetic Property Measurement System (MPMS:Quantum Design). Several NMR data at ambient pressure have already been reported in another paper [11].

Figure 1 shows NMR spectra for (a) 60 K and 0 GPa, (b) 4.2 K and 0 GPa, and (c) 1.6 K and 0.45 GPa. As shown in Fig. 1(a), 19 lines are totally observed in the PM phase at high temperatures for two Co sites (the nuclear spin $I = 7/2$) and the Al site ($I = 5/2$). Seven lines originating in the Co(2) site appear with almost an equal interval between each line due to the nuclear quadrupole interaction. At low temperatures, such clear lines disappear, and the spectrum changes discretely at H_m due to the drastic change in the internal field H_{int} caused by the metamagnetic (PM–FM) transition because of the resonance condition $f_0 = \gamma(H + H_{int})$ for the central transition, where f_0 is the NMR frequency, γ is the gyromagnetic ratio, and H is the external magnetic field. H_m is ~ 0.7 T at ambient pressure while it increases to ~ 2.2 T at 0.45 GPa. As shown in the figure, the signals from the Co(1) site and the Al site are overlapped and cannot be separated at low temperatures, so we evaluated the internal field from the satellite lines ($\pm 5/2 \leftrightarrow \pm 3/2$ transitions) of the Co(2) site. The field dependence can be detected continuously by changing the NMR frequency f_0.

Figure 2(a) shows the field dependence of the internal field, H_{int} at the Co(2) site at different pressures. The H_{int} was estimated from the shift in the resonance line for the frequency-swept spectrum at a fixed field. The magnetization at ambient pressure is also plotted [12]. A distinct metamagnetic transition can be observed at the metamagnetic field H_m by using NMR measurements through the hyperfine field from the U site. Figure 2(b) shows M vs. H_{int} when treating the magnetic field as

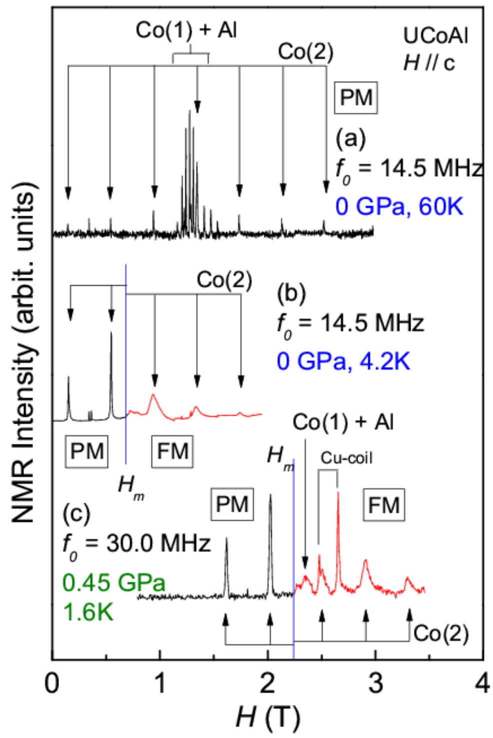

Fig. 1. (Color online) NMR spectra for UCoAl measured at (a) 60 K and 0 GPa, (b) 4.2 K and 0 GPa, and (c) 1.6 K and 0.45 GPa. At high temperatures, 19 NMR lines are observed for two Co sites and the Al site. The spectra at low temperatures are discontinuous on crossing H_m. H_m increases under pressure.

an implicit parameter. The slope corresponding to the hyperfine coupling constant has already been given by the temperature dependence of the NMR shift and the susceptibility [11]. The estimated value $A^c_{spin} = 2.58$ T/μ_B also reproduces the field dependence well, suggesting that the field dependence of A^c_{spin} is small. The H_{int} in the high-field FM phase is suppressed gradually under pressure while the slopes in the PM phase are almost overlapped for the different pressures. The jump at H_m (ΔH_{int}) also decreases with increasing pressure. This behavior is almost consistent with previous magnetization data [9]. At 1.14 GPa, we could not observe the NMR signal at fields around H_m due to the fast relaxation, suggesting that $T_{\rm CEP}$ at 1.14 GPa is close to 1.6 K because of a suppression of $T_{\rm CEP}$ from ~ 12 K at ambient pressure. This is almost consistent with the estimate of $T_{\rm CEP} \sim 4$ K based on resistivity measurements at the corresponding pressures [4].

Figure 3 shows the pressure evolutions of H_m and ΔH_{int}. The pressure evolution of H_m is displayed along with the estimates obtained using the resistivity and the magnetostriction [4]. In the resistivity measurements, another anomaly is detected after exceeding the QCEP as denoted by H^* [4]. The pressure dependences of H_m from all the measurements agree with one another. The

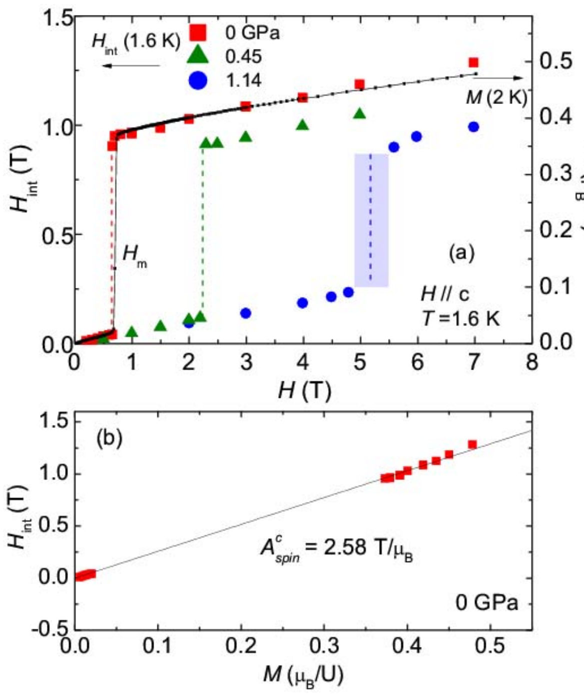

Fig. 2. (Color online) (a) Field dependence of the internal field at the Co(2) site measured at 1.6 K and different pressures, and the magnetization at ambient pressure. Each measurement is performed under $H \parallel c$. The internal field increases drastically at the metamagnetic transition at H_m. Sharp transitions are observed at 0 GPa and 0.45 GPa. The internal field at the Co(2) site scales well to the magnetization. At 1.14 GPa, the NMR signal could not be observed due to the fast relaxation time, implying that it is located in the vicinity of the QCEP. (b) Magnetization vs. the internal field. The slope corresponds to the hyperfine coupling constant, which is almost unchanged between the PM phase and the FM phase.

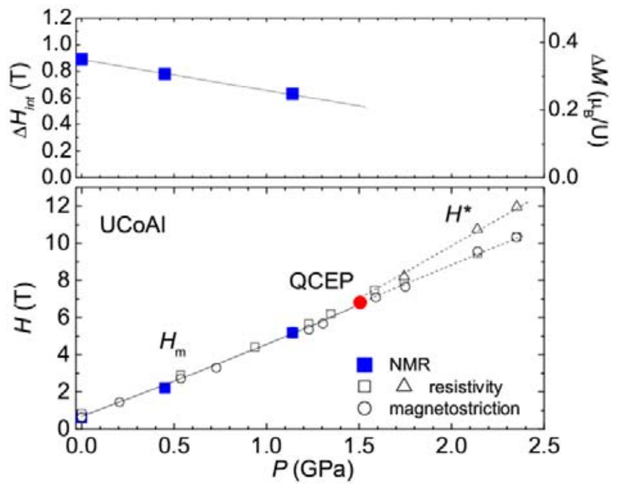

Fig. 3. (Color online) Pressure dependences of H_m (bottom panel) and ΔH_{int} (top panel). The H_m estimated from the resistivity and the magnetostriction is cited from Ref. 4. The H_m increases monotonously with increasing pressure up to the QCEP, which is estimated to be located at $P_{\rm QCEP} \sim 1.5$ GPa and $H_{\rm QCEP} \sim 7$ T. [4] ΔH_{int} also decreases monotonically at least up to 1.14 GPa. ΔM is estimated using the hyperfine coupling constant $A^c_{spin} = 2.58$ T/μ_B [11].

loss of the NMR signal at 1.14 GPa and ~ 1.6 K indicates that it is close to the QCEP, consistent with the estimate of $P_{\rm QCEP} \sim 1.5$ GPa [4]. The ΔH_{int} decreases monotonically under increasing pressure at least up to 1.14 GPa. If we use the hyperfine coupling constant $A^c_{spin} = 2.58$ T/μ_B with the assumption that it is independent of pressure, a jump in the magnetization ΔM under pressure can be obtained. It should be noted that A^c_{spin} is unchanged between the PM phase and the FM phase, as shown in Ref. 11 and Fig. 2(b). The simple extrapolation gives $\Delta M \sim 0.2\mu_B$/U at the QCEP, which lower than the $\Delta M \sim 0.34\mu_B$/U at ambient pressure, but is still robust. In the vicinity of the CEP at ambient pressure ($T_{\rm CEP} \sim 12$ K), the strong longitudinal magnetic fluctuations are observed [11]. The strong fluctuations are expected in the vicinity of the QCEP; however their detailed character is under investigation. In addition, the origin of another anomaly at H^* at high-pressure and high-field, which might be related to an instability in the Fermi surface, will be investigated by using NMR measurements.

III. CONCLUSIONS

We have observed the metamagnetic transition under pressure in UCoAl by using NMR measurements. NMR can sensitively detect the evolution of the hyperfine filed from the U site due to the metamagnetic transition. The metamagnetic field H_m increases with increasing pressure and is accompanied by the suppression of the magnetization in the FM phase and a jump in the magnetization at H_m. Measurements under higher pressures are desired to investigate the magnetic properties in the vicinity of the QCEP and the origin of the branch off of the metamagnetic line.

ACKNOWLEDGMENTS

This work has been partly supported by Grants-in-Aid for Scientific Research (Nos. 20102005, 22340102, and 22740231, and 24340085) from the Ministry of Education, Culture, Sports, Science and Technology (MEXT) of Japan.

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