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LETTER TO THE EDITOR

Thermal expansion and magnetostriction studies in a heavy-fermion superconductor, $CeCoIn_5$

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Abstract
We have measured the iso-field thermal expansion and magnetostriction in a heavy-fermion superconductor, $CeCoIn_5$, with tetragonal structure. The thermal expansion at zero field shrinks for the [100] direction in the superconducting state, while it expands for [001]. The magnetic field effect on the superconducting transition at the superconducting transition temperature T_c is remarkable for [001]. The longitudinal magnetostriction for $H \parallel [001]$ shows a continuous field dependence with a bend at the upper critical field H_{c2} below T_c, but this feature changes into a steplike transition below about 0.7 K, indicating that the transition changes from the second to the first order. We have also measured the thermal expansion at high temperatures, which is compared with those in $CeIrIn_5$ and $CeRhIn_5$.

There has been an increasing interest in heavy-fermion superconductors, since the interplay of magnetism and superconductivity gives rise to a variety of characteristic superconducting ground states such as a multiple superconducting phase and an unconventional form of superconductivity with a low-symmetry order parameter. In particular, some Ce-based compounds with the tetragonal $ThCr_2Si_2$-type structure were found to become superconductors at low temperatures in the vicinity of a quantum critical point where their antiferromagnetic ordering temperatures are tuned toward zero by applying pressure and/or chemical alloying [1–3]. The superconducting transition temperature T_c is, however, usually below 1 K, and the quantum critical point is sited in a rather high pressure range from 1 to 10 GPa. The proximity to the magnetic order suggests that the unconventional superconductivity in heavy-fermion compounds is most likely mediated by magnetic fluctuations, which is also

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responsible to the development of the heavy-fermion state with a large effective mass of quasi-particles. In spite of stimulating experimental and theoretical efforts, the detailed mechanism is still controversial.

Quite recently, a new series of the Ce-based heavy-fermion system $CeTIn_5$ (T = Co, Ir or Rh) was discovered, which has the tetragonal $HoCoGa_5$-type crystal structure [4–6]. The characteristic feature of this structure is that alternating layers of $CeIn_3$ and TIn_2 are stacked sequentially along the [001] direction (*c*-axis). $CeRhIn_5$ orders antiferromagnetically at $T_N = 3.8$ K at ambient pressure, but becomes a superconductor with $T_c = 2.1$ K at pressures higher than about 1.6 GPa [4]. On the other hand, $CeIrIn_5$ and $CeCoIn_5$ show superconductivity below $T_c = 0.4$ and 2.3 K, respectively, at ambient pressure [5,6]. Alloying studies on pseudo-binary compounds $CeIr_{1-x}Rh_xIn_5$ and $CeCo_{1-x}Rh_xIn_5$ revealed that superconductivity in these compounds is stable in a wide range of composition and even coexists with a magnetically ordered phase [7]. Therefore, $CeTIn_5$ is an ideal candidate to study the relation between magnetism and superconductivity in the heavy-fermion superconductors.

$CeCoIn_5$ has the highest transition temperature T_c among the known heavy-fermion superconductors. The electronic specific heat coefficient C/T increases approximately as a function of $-\sqrt{T}$ on cooling in a magnetic field sufficiently larger than the upper critical field H_{c2} in superconductivity. In addition, the resistivity exhibits linear temperature dependence below 20 K, and the magnetic susceptibility does not saturate at low temperatures but increases with decreasing temperature. These non-Fermi-liquid features indicate that $CeCoIn_5$ is situated close to the quantum critical point [5]. The specific heat and thermal conductivity show power-law temperature dependences in the superconducting state, which suggest an unconventional nature of the superconductivity with a line node [8]. NMR Knight shift measurements revealed a suppression of the spin susceptibility below T_c, implying a singlet pairing of Cooper pairs in the superconducting state [9]. Recently, fourfold symmetry was observed in the angular dependence of thermal conductivity in magnetic fields, and the symmetry of the superconducting order parameter was identified as the $d_{x^2-y^2}$ type, similar to high-T_c cuprates [10].

Another intriguing feature in the superconducting state is a steplike phase transition at H_{c2} observed in the thermal conductivity for $H \parallel [001]$ [10]. They claimed that this is the first material which shows a first-order phase transition at H_{c2} because the steplike change is most likely caused by an entropy jump. Such a first-order phase transition has already been predicted by theory when the Pauli paramagnetic effect is sufficiently strong compared with the orbital effect [11–13]. Although a strong paramagnetic effect was indeed observed in $CeCoIn_5$ [6,14], it is necessary to establish the first-order phase transition by another experimental method.

Thermal expansion and magnetostriction measurements by using the three-terminal capacitance method are powerful tools to detect phase transitions even in a very small sample because this method is basically highly sensitive in detection. To obtain more insight into the magnetic field effect on the superconducting transition, we performed thermal expansion and magnetostriction measurements for $CeCoIn_5$.

The single crystals were grown by the so-called self-flux method [6, 15]. The crystal structure was identified as the tetragonal $HoCoGa_5$-type with lattice constants $a = 4.612$ Å and $c = 7.549$ Å. The sample has a rectangular shape with dimensions of 1.72 mm × 3.45 mm × 1.04 mm. The parallel-plate capacitance cell was made of an oxygen-free high-conductivity copper and was placed in a superconducting magnet with a maximum field of 8 T. We checked the pressure effect from a movable electrode, which is pressurized to the sample by a spring in the capacitance cell, and confirmed that there is no such effect on the data.

Figure 1 shows the longitudinal thermal expansion under magnetic fields applied along (*a*) [100] and (*b*) [001] directions. In both panels, the data under magnetic fields

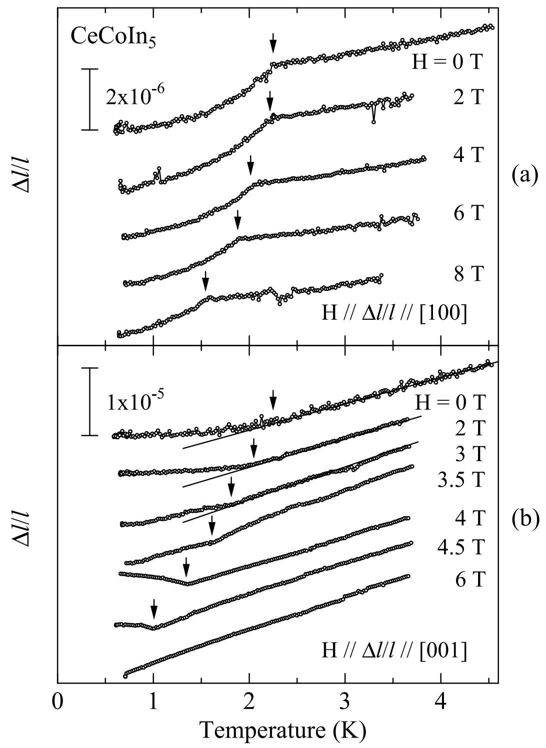

Figure 1. Iso-field thermal expansion $\Delta\ell/\ell$ as a function of temperature for (*a*) [100] and (*b*) [001] in $CeCoIn_5$.

are shifted downward for clarity. The thermal expansion for [100] is almost linear in the temperature dependence above $T_c = 2.25$ K and decreases in the superconducting state with decreasing temperature, showing that the lattice along [100] contracts in association with evolution of superconductivity. On the other hand, the temperature dependence of thermal expansion along [001] is also nearly linear above T_c. The superconducting transition at T_c for $H \leqslant 3$ T is less pronounced than that in [100], but the data start to deviate upward at T_c on cooling from a linear extrapolation from high temperatures which is shown by thin solid curves. The superconducting transition for $H > 3$ T becomes clear and shifts to a lower temperature with increasing magnetic field: see the data at 4 T. At $H = 6$ T, where the superconducting state is suppressed by the magnetic field, the linear decrease of the thermal expansion continues to the lowest temperature. It is noted that the linear decrease above T_c for [001] is significantly larger than that for [100], probably reflecting the quasi-two-dimensional crystal structure. The linear temperature dependence may be related to the non-Fermi-liquid nature observed in the specific heat, electrical resistivity and susceptibility measurements mentioned above [6].

Next we show in figure 2 the temperature dependence of volume thermal expansion, which was calculated from $\Delta V/V = 2(\Delta\ell/\ell)_{[100]} + (\Delta\ell/\ell)_{[001]}$, and its temperature derivative $\alpha_V = (1/V)(\Delta V/\Delta T)$ at $H = 0$.

The volume thermal expansion decreases linearly down to T_c with decreasing temperature. It decreases more rapidly in the temperature range from T_c down to 1.5 K, but decreases gradually with further decreasing temperature. Correspondingly, the coefficient of volume

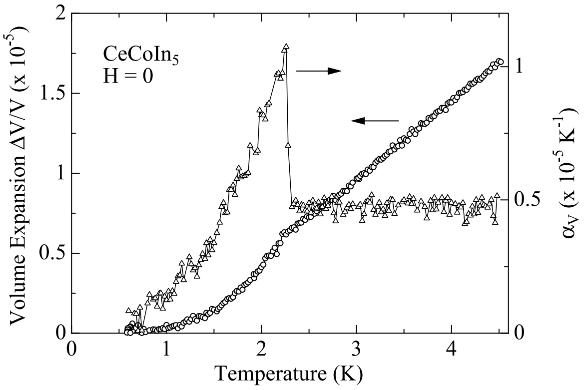

Figure 2. Temperature dependences of the volume thermal expansion and the volume thermal expansion coefficient in $CeCoIn_5$.

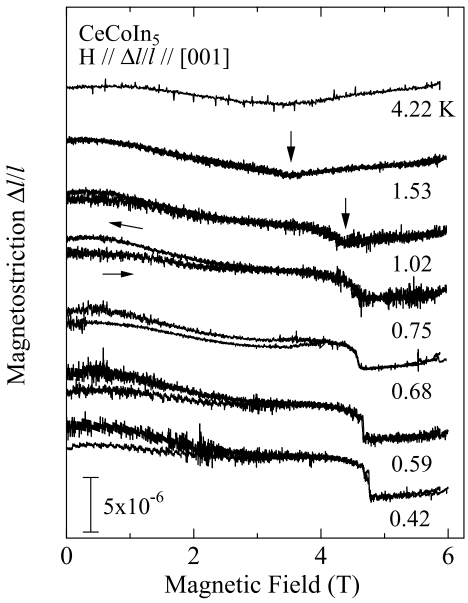

Figure 3. Longitudinal magnetostrictions along the [001] direction in $CeCoIn_5$.

thermal expansion shows a lambda-shape temperature variation below T_c, where it is nearly constant at high temperatures and exhibits a jump at T_c followed by a continuous decrease with decreasing temperature.

The initial slope of the pressure dependence of the superconducting transition temperature dT_c/dp can be estimated from the Ehrenfest relation defined by $dT_c/dp = V_m \Delta\alpha_V / \Delta(C/T)$, where V_m is the molar volume, and $\Delta\alpha_V$ and $\Delta(C/T)$ correspond to changes at T_c of the volume thermal expansion coefficient and the specific heat divided by temperature, respectively. By using experimental values of $V_m = 97.1$ cm^3 mol^{-1}, $\Delta\alpha_V = 6.0 \times 10^{-6}$ K^{-1} and $\Delta(C/T) = 1.7$ J K^{-2} mol^{-1} [14], we calculated dT_c/dp as 0.34 K GPa^{-1}. This is somewhat smaller than but agrees reasonably well with the recently reported value of $dT_c/dp = 0.5$ K GPa^{-1} which was determined from the resistivity and susceptibility measurements under pressure [16].

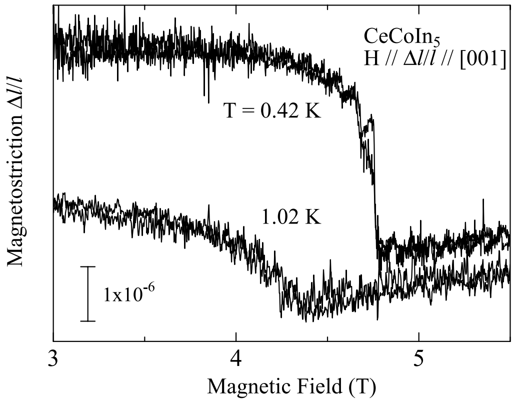

Figure 4. Magnetostriction for [001] around H_{c2} in $CeCoIn_5$.

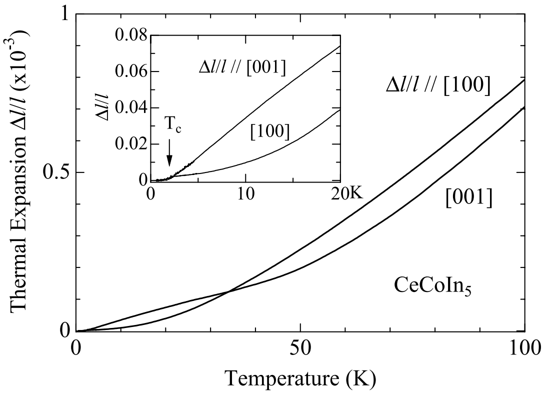

Figure 5. Temperature dependence of the thermal expansion for [100] and [001] in $CeCoIn_5$.

Figure 3 shows longitudinal magnetostriction curves for $H \parallel [001]$ at several temperatures below 4.22 K. The data were obtained by tentatively correcting background changes, though the signal from the sample is very small relative to the background change. The magnetostriction at 4.22 K, which is above T_c, changes smoothly in the field-up and down cycle. Below T_c, we observed a weak anomaly around 3.5 T, which shifts towards higher fields at lower temperatures. This anomaly corresponds to the transition from the superconducting to normal state at H_{c2} [6]. It becomes obvious with decreasing temperature and changes into a steplike transition below about 0.7 K. Another characteristic feature in the superconducting state is a hysteresis of the magnetostriction in a low field range, which becomes larger upon decreasing temperature. This behaviour is most likely due to the irreversibility of a magnetic flux motion based on the pinning effect.

In order to see the transition around H_{c2} more precisely, we show in figure 4 magnetostriction curves at selected temperatures in enlarged scales. At 1.02 K, the magnetostriction shows a continuous field dependence with a sharp bend at 4.4 T, indicating the transition is of second order. On the other hand, the magnetostriction at 0.42 K exhibits jump-down and up changes at H_{c2} with increasing and decreasing field, respectively, accompanying a very small hysteresis of $\Delta H < 200$ Oe. A change of magnetostriction at H_{c2} is about 2×10^{-6}, that is a change of about 20 Å length in our sample. This clear contrast indicates that the transition nature at H_{c2} changes from the second to the first order around 0.7 K.

Figure 5 shows the temperature dependence of thermal expansion $\Delta\ell/\ell$ along [100] and [001] directions between 0.5 and 100 K. The low-temperature region is enlarged as shown in the inset.

The thermal expansion for both directions decreases almost linearly down to 70 K, showing a small anisotropy. The thermal expansion for [100] decreases monotonically on further cooling down to T_c. On the other hand, the decrease of the thermal expansion for [001] becomes slow in the temperature range from 50 to 30 K and a rapid decrease appears again below about 20 K. This rapid decrease continues down to T_c. The qualitative features of the thermal expansion in $CeCoIn_5$ are very similar to those in another ambient-pressure superconductor, $CeIrIn_5$, in which the rapid decrease for [001] appears below about 10 K, reflecting the development of the heavy-fermion state via the Kondo effect [17]. The temperature dependence of thermal expansion in these two superconductors is in striking contrast with the data in the antiferromagnetic compound $CeRhIn_5$, which were well explained on the basis of the crystalline electric field model [17]. The quantitative analysis of the thermal expansion in $CeCoIn_5$ will be published elsewhere.

In summary, we have thus confirmed the first-order phase transition at H_{c2} in $CeCoIn_5$ via iso-field thermal expansion and magnetostriction measurements. The anomaly at T_c in the thermal expansion for [001] is weak in low fields but changes into a clear bend with increasing magnetic field. Furthermore, the transition at H_{c2} in the magnetostriction becomes a steplike change below about 0.7 K. The temperature variations of thermal expansion for [100] and [001] below 100 K are very similar to the data in another ambient-pressure superconductor, $CeIrIn_5$.

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