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LETTER TO THE EDITOR

Field-angle dependence of the zero-energy density of states in the unconventional heavy-fermion superconductor $CeCoIn_5$

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Abstract
Field-angle-dependent specific heat measurements have been made on the heavy-fermion superconductor $CeCoIn_5$ down to ∼0.29 K, in a magnetic field rotating in the tetragonal c-plane. A clear fourfold angular oscillation is observed in the specific heat with the minima (maxima) occurring along the [100] ([110]) directions. Oscillation persists down to low fields $H \ll H_{c2}$, thus directly proving the existence of gap nodes. The results indicate that the superconducting gap symmetry is most probably of d_{xy} type.

Recently a new class of heavy-fermion superconductors $CeTIn_5$ (T = Rh, Ir and Co) has been discovered. While $CeRhIn_5$ [1] shows pressure-induced superconductivity, $CeIrIn_5$ [2] and $CeCoIn_5$ [3] show superconductivity at ambient pressure at 0.4 and 2.3 K, respectively. The unique properties of $CeCoIn_5$ have attracted much attention in recent years. Various experiments such as specific heat [4, 5], thermal conductivity [4] and NMR T_1 measurements [6] have revealed that $CeCoIn_5$ is an unconventional superconductor with line nodes in the gap. From this, together with the suppression of the spin susceptibility below T_c [6, 7], this compound is identified as a d-wave superconductor. Recent rf penetration depth measurements [8] and the flux line lattice imaging study by means of small-angle neutron scattering [9] also seem to be consistent with the existence of line nodes running along the c-axis. In unconventional superconductors, identification of the gap node structure is of fundamental importance in understanding the pairing mechanism. Regarding this issue, it has recently been pointed out that the zero-energy density of states (ZEDOS) in the superconducting mixed state exhibits a characteristic oscillation with respect to the angle between $\boldsymbol{H}$ and the nodal direction [10–12]. An intuitive explanation of this effect employs a Doppler shift of the quasiparticle (QP) energy spectrum due to the local supercurrent flow [10–13]. More quantitative analysis of the effect has been carried out recently by Miranović *et al* [14] within the quasi-classical formalism, which also incorporates the contribution of QPs in the core states of vortices. Experimentally, the angle-dependent ZEDOS can be probed by means of thermal conductivity [15–17] or specific heat [18] measurements in rotating magnetic fields at low T. As for $CeCoIn_5$, angle-resolved thermal conductivity ($\kappa(\theta)$) measurements [17] have revealed a clear fourfold oscillation in a magnetic field rotating within the c-plane. It was argued that the line nodes exist along the [110] directions ($d_{x^2-y^2}$-type gap symmetry).

The interpretation of the thermal conductivity data is, however, necessarily involved because $\kappa(\theta)$ is proportional to the specific heat $C(\theta)$ as well as to the QP scattering time $\tau(\theta)$. The latter also depends on the field orientation but with opposite angular oscillation amplitude to the former. Which of these two contributes predominantly to $\kappa(\theta)$ is a subtle question, making it difficult to identify the gap node direction in some cases. It is therefore of vital importance to directly observe the ZEDOS by means of angle-resolved specific heat measurements. In this letter, we have examined the $C(H, \theta)$ of $CeCoIn_5$ in a magnetic field rotating within the c-plane. We observed a clear fourfold oscillation in $C(H, \theta)$ and argue that the oscillation originates from the nodal gap, most probably of d_{xy} symmetry.

A single crystal of $CeCoIn_5$ was grown by the so-called self-flux method [19]. The sample was cut into a thin plate ($\sim 2.3 \times 2.0 \times 0.7$ mm^3) with the largest plane oriented perpendicular to the c-axis. Field-angle-dependent specific heat measurements were made by a standard adiabatic heat-pulse technique using a ^{3}He refrigerator. A transverse magnetic field was generated by a split-pair superconducting magnet and applied parallel to the c-plane of the sample mounted on a quartz platform. The ^{3}He refrigerator was mounted on a mechanical rotating stage driven by a computer controlled stepping motor at the top of the Dewar, by means of which a quasi-continuous change of the field direction with the minimum step of 0.04° could be made. The angle-dependent specific heat data were collected at intervals of 2° in the range ±105° with respect to the a-axis. At the temperature of 380 mK, a 2° rotation of the refrigerator caused heating of the sample by ∼100 mK in a field of 5 T. In order to accurately evaluate $C(H, \theta)$ at constant temperature, each data point was taken after the sample temperature relaxed to within 0.1% of the initial temperature. The details of the experimental set-up will be published elsewhere [20]. The field dependence of the specific heat up to 12.5 T was also measured by the relaxation method using a commercial calorimeter (PPMS, Quantum Design Co.).

Figure 1 shows the field dependence of the specific heat $C(H)$ of $CeCoIn_5$ obtained at $T = 0.38$ K for $\boldsymbol{H} \parallel [100]$ and [110]. Throughout this letter, the nuclear spin contribution C_{nuc} has been subtracted from the data, assuming the form $C_{nuc} = (A_0 + A_1 H^2)/T^2$ with $A_0 = 7.58 \times 10^{-2}$ mJ K mol^{-1} and $A_1 = 6.90 \times 10^{-2}$ mJ K mol^{-1} T^{-2} [5]. $C(H)$ of $CeCoIn_5$ in the superconducting mixed state is quite unusual. This behaviour is very different from the one expected for ordinary s-wave superconductors in which $C(H)$ linearly increases with H due to the contribution of QPs trapped in the vortex cores. In many anisotropic superconductors, a power law dependence $C(H) \propto H^\beta$ with $\beta \sim 0.5$ has been observed and is attributed to the nodal QP excitations. The negative curvature of $C(H)$ in low fields seen in figure 1 is thus consistent with the existence of gap nodes in $CeCoIn_5$. However, the curvature of $C(H)$ changes sign above 2 T and becomes strongly positive at higher fields, as shown in the inset. Similar behaviour was previously reported for $\boldsymbol{H} \parallel [001]$ [5], as well as for Sr_2RuO_4 for $\boldsymbol{H} \parallel [110]$ [21]. Although the reason for this field dependence of the specific heat in $CeCoIn_5$ is unclear at the moment, it might be related to non-Fermi liquid behaviour near the quantum critical point. With increasing H in the c-plane, we observed a small but discernible anisotropy in $C(H)$ as shown in figure 1; $C(\boldsymbol{H} \parallel [100]) < C(\boldsymbol{H} \parallel [110])$.

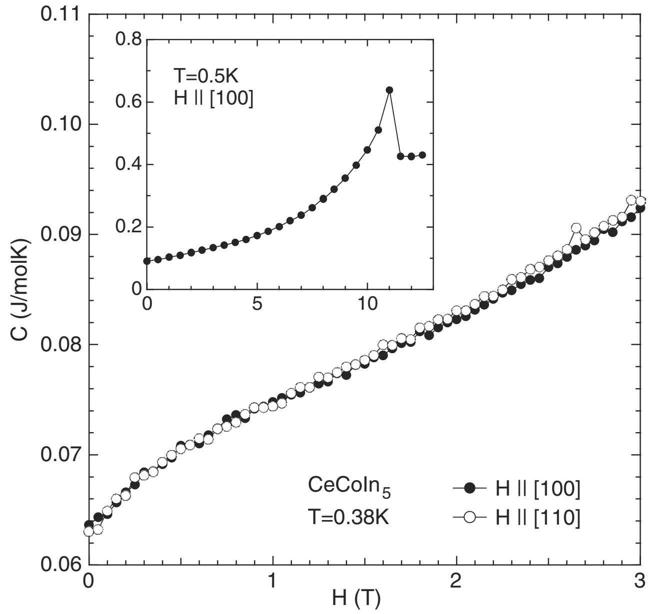

Figure 1. The field dependence of the specific heat $C(H)$ of $CeCoIn_5$ for $\boldsymbol{H} \parallel [100]$ (solid circles) and [110] (open circles), measured at $T = 0.38$ K. Inset: the overall field variation of $C(H)$ up to 12.5 T for $\boldsymbol{H} \parallel [100]$, measured at 0.5 K.

The in-plane anisotropy of $C(H)$ can be demonstrated more clearly by measuring its field-angle dependence, and some of the results are shown in figure 2. First of all, we examined the contribution of the addenda (the lower trace of figure 2(a)), which comprise the quartz platform, a thermometer (RuO_2 chip resistor) and a heater. As can be seen in the figure, there is virtually no angular dependence in the addenda specific heat, implying that the field-orientational dependence of the thermometer is negligible. By contrast, there is a small but distinct fourfold oscillation in the raw specific heat data with the sample on the platform (the upper trace of figure 2(a)). Comparing these two data sets, it is obvious that the observed angular oscillation is intrinsic to the sample. No appreciable twofold component is observed in the oscillation, indicating that the magnetic field is well oriented along the c-plane.

The specific heat $C(H, \theta)/T$ of $CeCoIn_5$ is shown in figure 2(b) for $H = 2$ T ($T = 0.29$ and 1 K) and 5 T ($T = 0.38$ K) as a function of the field angle θ measured with respect to the a-axis. The fourfold oscillation is clearly seen at the base temperature of 0.29 K but rapidly fades away at higher temperatures. $C(H, \theta)$ can be decomposed into constant and field-angle-dependent terms: $C(H, \theta) = C_0 + C_H(1 + A(\theta))$. C_0 is the zero-field term mainly due to thermally excited quasiparticles and phonons, whereas C_H and $A(\theta)$ are field dependent. The solid curves in figure 2(b) are the fitting results assuming a simple form $A(\theta) = A_4 \cos 4\theta$, with which we evaluated the amplitude of the fourfold angular oscillation. The sign of A_4 is negative, with the minima of $C(H, \theta)$ occurring along the a-directions. In figure 3, we plotted the field dependence of the relative amplitude $|A_4|$ as a function of H/H_{c2}. Most remarkably, $|A_4|$ decreases monotonically with H within the range of fields examined. The inset of figure 3 shows the temperature variation of the absolute amplitude $|A_4|C_H/T$ measured at $H = 2$ T ($H/H_{c2} = 0.175$). The fourfold angular oscillation in $C(H, \theta)/T$ rapidly diminishes with increasing T and vanishes above T_c, implying that superconductivity is responsible for its occurrence.

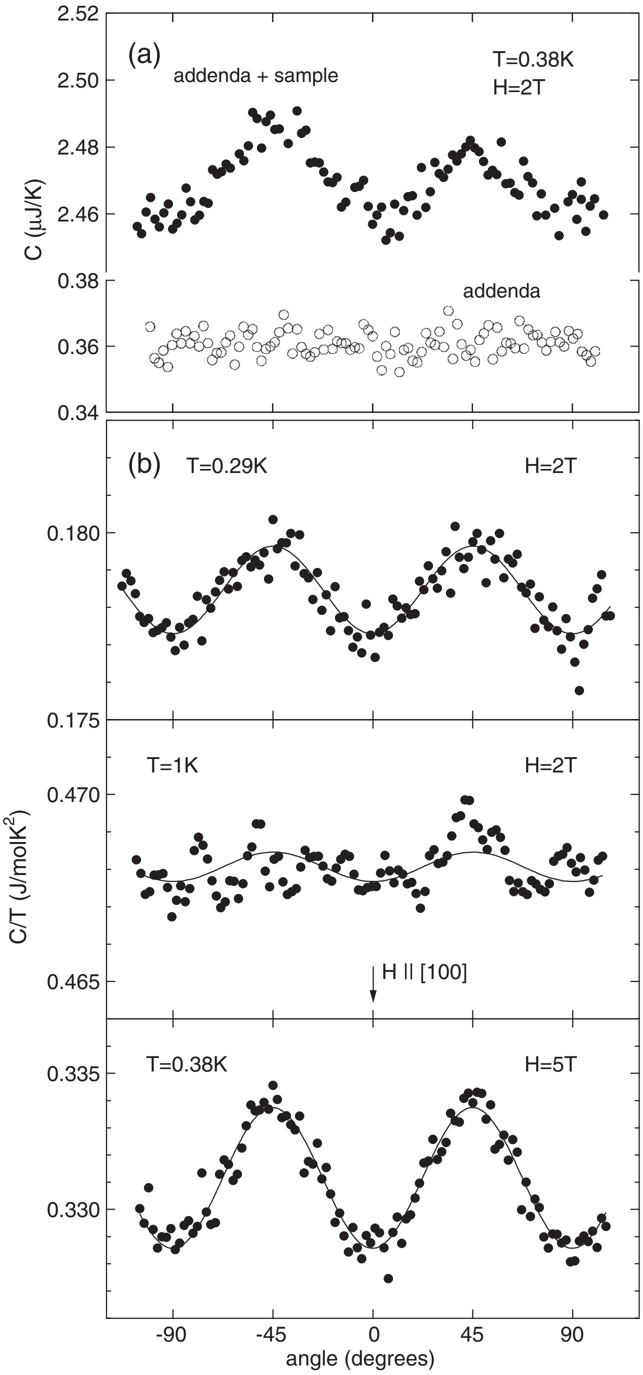

Figure 2. (a) The angular dependence of the total specific heat of $CeCoIn_5$ (solid circles) mounted on the platform, in a field of 2 T rotated in the c-plane at $T = 0.38$ K. $\theta = 0$ is the [100] direction. Open circles show the results without the sample. (b) The field-orientational dependence of the specific heat of $CeCoIn_5$ for $H = 2$ T measured at $T = 0.29$ and 1 K, and for $H = 5$ T measured at $T = 0.38$ K. The solid curves are the fits to $C(H,\theta)/T = (C_0 + C_H(1 + A_4 \cos 4\theta))/T$, where $C_0 = 0.0361$, 0.0631 and 0.4042 J mol^{-1} K^{-1} for $T = 0.29$, 0.38 and 1 K, and $A_4 = -0.0217$ (2 T, 0.29 K), -0.0157 (5 T, 0.38 K) and -0.0061 (2 T, 1 K).

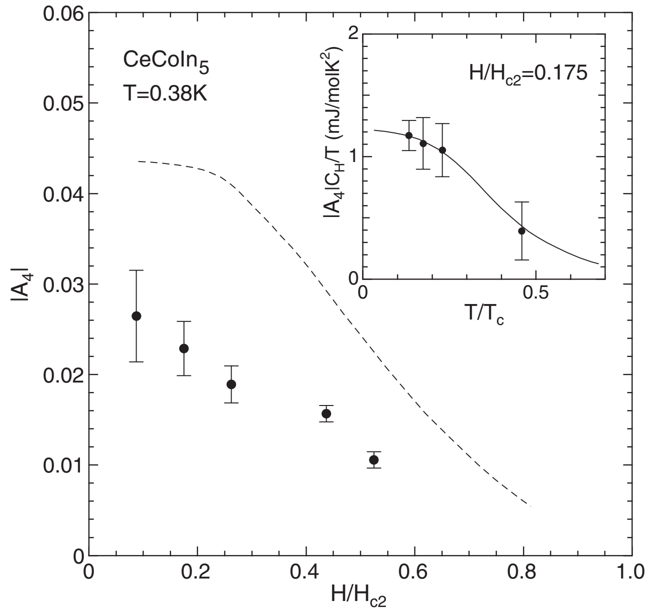

Figure 3. The field dependence of the amplitude $|A_4|$ of the fourfold oscillation at $T = 0.38$ K, plotted as a function of the reduced field H/H_{c2}. The broken curve shows the calculated field variation for $|A_4|$ at $T = 0$ assuming a d_{xy}-type gap node. Inset: the temperature dependence of the absolute amplitude $|A_4|C_H/T$ measured at $H/H_{c2} = 0.175$. The solid curve is a guide for the eyes.

We now discuss the origin of the fourfold oscillation in $C(H,\theta)$. First of all, we should pay attention to the in-plane anisotropy of H_{c2}, which is determined both by the gap topology and the Fermi surface anisotropy; $H_{c2} \parallel [100]$ is about 2.7% larger than $H_{c2} \parallel [110]$ [19]. The in-plane anisotropy of H_{c2} alone would also give rise to an angular dependence of the ZEDOS. This is because the gap amplitude, which is a decreasing function of the reduced field H/H_{c2}, becomes field-angle dependent under fixed H. This effect certainly becomes important when H is near H_{c2}, and enhances $C(H,\theta)$ for $\boldsymbol{H} \parallel [110]$ along which the system is closest to the normal state; the sign of the fourfold amplitude is the same as we observed. However, in the limit of low fields $H \ll H_{c2}$ the effect of the upper critical field can be disregarded. A crude estimate, assuming that $C(H)$ in figure 1 can be scaled linearly with $H_{c2}(\theta)$, shows that the $\sim$3% anisotropy in H_{c2} would give a fourfold term A_4 of the order of 1% at 2 T. This definitely overestimates the effect of H_{c2} anisotropy, but is still smaller than the observed one. Moreover, one expects the effect of the upper critical field to increase with increasing field. Instead, the oscillation amplitude is a decreasing function of H up to at least $H \sim 0.5H_{c2}$. The strong temperature variation of the fourfold amplitude of $C(H)/T$ shown in the inset of figure 3 is also hardly explained by the H_{c2} in-plane anisotropy effect, because the upturn in $C(H)/T$ does not change much in these temperature regions [5]. Since the effect of the H_{c2} anisotropy is incompatible with the data, we are led to conclude that the observed angular oscillation in $C(H,\theta)$ originates from the nodal structure, which has fourfold symmetry in the *ab*-plane.

In the mixed state of anisotropic superconductors the enhanced contribution to the ZEDOS is coming from the QPs with momentum along the gap node direction. The enhancement depends on the relative position of the gap and the magnetic field direction. Accordingly, the ZEDOS becomes field-angle dependent, and exhibits a characteristic oscillation with respect to the angle between $\boldsymbol{H}$ and the gap node directions. This ZEDOS oscillation is directly reflected in the field-orientational dependence of the specific heat at low T, which takes minima (maxima) for $\boldsymbol{H}$ parallel to the nodal (antinodal) direction [10, 12, 14]. The clear minima of $C(H, \theta)$ along [100], which persist down to low fields, indicate that there are gap nodes along these directions. Here we performed a microscopic calculation of the field-angle-dependent ZEDOS of a clean d-wave superconductor with a spherical Fermi surface. We solved numerically and self-consistently the quasi-classical Eilenberger equations. This formalism is a good approximation as long as $k_F\xi \gg 1$ (k_F being the Fermi wavenumber and ξ the coherence length), a condition which is met in $CeCoIn_5$. It takes into account both the Doppler shift effect and the vortex core contributions, on an equal footing and without any further approximation. The result is presented as the broken curve in figure 3. Although the predicted curve gives slightly a larger amplitude than the experimental one, it explains the overall field dependence of our experimental result reasonably well. The discrepancy between the predicted and observed amplitudes may partly arise from the fact that the measurements have been made at finite T ($T/T_c \sim 0.17$), as inferred from the strong temperature variation of the amplitude predicted in [12]. The actual amplitude however gradually levels off below 0.5 K. Impurities, which are always present, may reduce the oscillation amplitude at lower T. The present data thus strongly indicate that the gap symmetry of $CeCoIn_5$ is most probably of d_{xy} type.

The present results agree with the $\kappa(\theta)$ measurement on $CeCoIn_5$ [17] on the point that the nodal structure has fourfold symmetry in the c-plane, but disagree on the location of the nodes. $\kappa(\theta)$ shows a fourfold oscillation with the minima along the [010] and [100] directions, which is the same oscillation behaviour as for our $C(\theta)$ data. The authors of [17] assumed that this angular dependence of $\kappa(\theta)$ is dominated by the QP scattering time, which becomes largest (smallest) when the magnetic field is along the nodal (antinodal) directions, just the opposite to the ZEDOS contribution. It is considered that the QP scattering effect is important at high temperatures, but the ZEDOS contribution becomes predominant with decreasing T. If the angular oscillation of $\kappa(\theta)$ had come from the QP scattering term, then its amplitude would change sign on cooling. The observed amplitude of the fourfold term in $\kappa(\theta)$ however continues to increase down to the lowest T of 0.35 K [17]. Whether the amplitude changes sign at still lower temperatures or not would be an interesting issue, but our $C(\theta)$ data imply that the ZEDOS effect is already predominant in $\kappa(\theta)$ at temperatures above 0.35 K.

The present data also do not contradict the neutron scattering experiment of Eskildsen *et al* [9], who find a square vortex lattice oriented along the [110] direction. The result of a neutron scattering experiment was argued to support the picture of $CeCoIn_5$ being a $d_{x^2-y^2}$ superconductor. However, the structure and the orientation of the vortex lattice is determined by the combined effect of the Fermi surface anisotropy and the gap structure. Which effect will prevail is a subtle question and the answer depends on many factors: field value, temperature, degree of Fermi surface anisotropy etc. Thus, the orientation of the square vortex lattice in tetragonal crystals may not serve as a conclusive test for the positions of gap nodes. A model calculation by Nakai *et al* [22] shows a variety of stable vortex lattice structures in tetragonal d_{xy}-wave superconductors. It is also shown that the low-field square vortex lattice is oriented along the [110] direction which is consistent with the neutron scattering experiment. It is interesting to see that the square vortex lattice orientation should rotate by $\pi/4$ in high fields if d_{xy} is indeed the correct identification of the gap function.

In summary, we have performed angle-resolved specific heat measurements on the heavy-fermion superconductor $CeCoIn_5$ in a magnetic field rotating in the basal ab-plane. We observed a clear fourfold symmetry in $C(H, \theta)$ with the minima oriented along [100] directions, which comes from the field-angle oscillation of the zero-energy density of nodal quasiparticles.

The results imply that the superconducting gap node of $CeCoIn_5$ is most probably located along the [100] and [010] directions, suggesting the symmetry to be of d_{xy} type.

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