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THE ANISOTROPIC LOW-TEMPERATURE SPECIFIC HEAT OF ORGANIC SUPERCONDUCTOR κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ IN THE MAGNETIC FIELD

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We measured the low-temperature specific heat of the layered organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ for the magnetic field directed along and perpendicular to the conducting plane and found the difference between the two measurements. Our data indicate the existence of a nodeless superconducting state at zero field and low temperature. The field dependence of the specific heat anisotropy consists of two linear branches with the crossover field equal to the upper critical field perpendicular to the conducting plane.

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1. INTRODUCTION

The problem of superconductivity in the low-dimension organic metals continues to attract much attention. A wide discussion on the possibility of an unconventional superconducting state in the κ-(BEDT-TTF)$_2$X compound involves arguments both in agreement and in disagreement with the hypothesis. For κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, there is an extensive evidence in favor of the unconventional character of superconductivity: NMR [1], high-frequency conductivity [2], thermal conductivity [3] and specific heat data [4]. As to the penetration depth data, both the unconventional [5] and the conventional [6] behavior have been reported. Recent data of Carrington et al. [7] support the presence of low-lying excitations but do not give a definite answer as to their origin.

The previously reported data [8] of the specific heat of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ under the magnetic field up to 6 T perpendicular to the conducting plane and the temperature region 1.65–4.4 K demonstrated an almost linear field dependence of the specific heat for the field considerably below the perpendicular upper critical field $H_{c2\perp}$. Above $H_{c2\perp}$, the specific heat is field-independent within the experimental error. To obtain more information about the character of the low-temperature superconducting state, we performed specific heat measurements under the different orientations of the magnetic field.

2. EXPERIMENTAL

For the measurements, we used a modification of the standard *ac*-modulation technique; the experimental details are described in Ref. [9] and Ref. [10]. One single crystal with the total mass 0.45 mg was used. In addition to the specific heat, the magnetoresistance of the sample was measured. The Dingle temperature extracted from the Shubnikov–de Haas oscillation was about 0.5 K. The specific heat was measured mainly for

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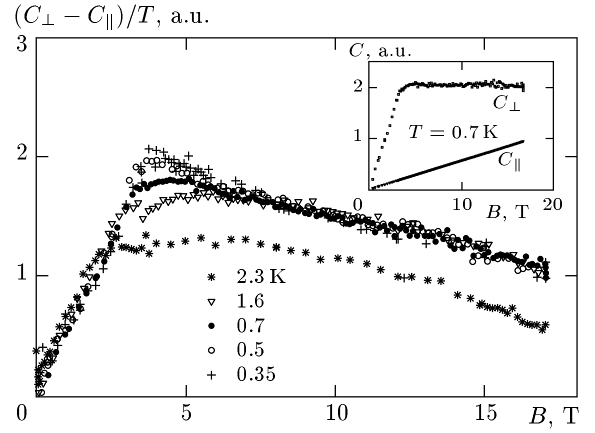

Fig. 1. The anisotropy of the field dependence of the specific heat at low temperatures. Inset: the extracted specific heat perpendicular ($C_\perp$) and parallel ($C_\parallel$) to the conducting layers

the magnetic field orientation along and perpendicular to the conducting plane. It was rather difficult to estimate the absolute value of the sample specific heat because of the small sample mass and an involved and significant field dependence of the specific heat of the thermometer. It was previously found that the specific heat of the thermometer is isotropic in the magnetic field. Our experimental setup makes it possible to rotate the sample *in situ*. Calculating the difference between the specific heat measured in the magnetic field parallel and perpendicular to the conducting plane, $C_\perp - C_\parallel = \Delta C$, we obtained the reliable value of the specific heat anisotropy ΔC.

3. RESULTS AND DISCUSSION

In Fig. 1, we plot the low-temperature specific heat anisotropy ΔC of the layered organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$, divided by the temperature. We can see that for each curve in Fig. 1, there are two regions: one is below $H_{c2\perp}$ (the upper critical field perpendicular to the conducting layers) and the other is above this field.

We first note that except in the region near $H_{c2\perp}$, all the curves coincide with each other. Using the data in Ref. [8], we can conclude that much below $H_{c2\perp}$, we have

$$C_\perp = A_1 T H, \qquad (1)$$

and at all fields,

$$C_\parallel = A_2 T H, \qquad (2)$$

where A_1 and A_2 are some constants. The field dependence of $C_\perp$ coincides with the one reported in the Ref. [8], although it is different from the one reported in Ref. [4], where a more abrupt increase of the electronic density of states in the magnetic fields below 0.03 T was observed.

The linear dependence of the specific heat on the magnetic field follows from the London model. It gives the electronic specific heat expressed as [11]

$$C(H,T) \approx -\frac{T\Phi_0 H}{32\pi^2}\frac{\partial^2}{\partial T^2}\left[\frac{1}{\lambda^2(T)}\frac{\alpha H_{c2}(T)}{H}\right], \qquad (3)$$

where Φ_0 is the flux quantum, $H_{c2}(T)$ is the upper critical field, and $\lambda(T)$ is the effective penetration depth in the plane perpendicular to the magnetic field. In the BCS approximation, Eq. (3) leads to the following field dependence of $C_\perp$ (see Ref. [12]) at the temperatures $T/T_c \ll 1$:

$$C_\perp \approx C_n \frac{H}{H_{c2}(0)}, \qquad (4)$$

with C_n being the electronic specific heat in the normal state. We recall that the London model is valid in the region $H_{c1} \ll H \ll H_{c2}$ and the Pauli breaking effect is neglected in that model; therefore, H_{c2} in Eq. (4) is actually the orbital upper critical field.

We note that the applicability of the London model does not depend on the superconducting pairing mechanism. The only important condition is a slow spatial variation of the order parameter over the length scale of the penetration depth. This condition is easily satisfied if the penetration depth of the magnetic field is much larger than the coherence length and the magnetic field is much lower than the upper critical field. In our case, the field dependence of the specific heat at low temperatures is almost linear up to the upper critical field.

Considerably above $H_{c2\perp}$, the data for the temperatures below 1.6 K almost coincide with each other. This indicates a BCS-like nodeless low-temperature superconducting state with the final gap in the excitation spectrum of the quasiparticle.

Using Eq. (4), we determined the upper critical field at the zero temperature as $H_{c2}(0.5\,\mathrm{K}) = 4$ T. The same value is obtained if we find the crossing point of the two linear branches. This is less than the value about 6 T reported by Sasaki et al. [13] and that about 5 T reported by Belin et al. [14], which were determined from

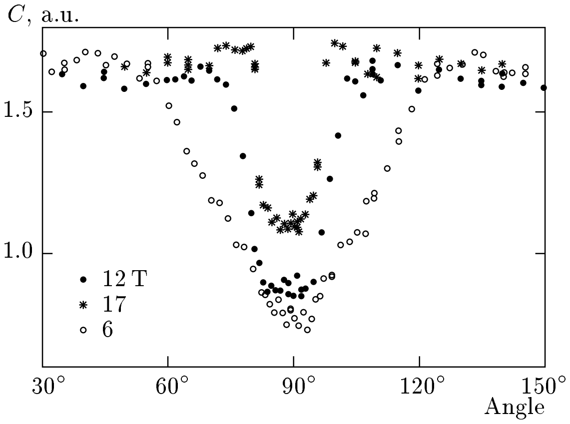

Fig. 2. The angular dependence of the specific heat at 0.5 K and different fields

the magnetic torque and the heat conduction measurements respectively. On the other hand, this is very close to the field of irreversibility at low temperatures reported in Ref. [13].

We note that the specific heat dependences on the magnetic field parallel to the layers is too strong for the expected Josephson coupling between the layers (see Ref. [15]). This problem requires a more detailed investigation of the low-temperature state for this orientation of the magnetic field. At present, we do not have any reasonable explanation of this fact.

The angular dependence of the specific heat is demonstrated in Fig. 2. It is noteworthy that the specific heat is almost independent of the angle in the region $\pm 3^\circ$ near the direction parallel to the conducting plane. We do not know the reason for this behavior but it can hardly be explained by the misalignment domen structure within the sample (with the planes inclined with respect to each other): such a misalignment has not been observed up to now [16].

4. CONCLUSIONS

In summary, we have demonstrated the nodeless superconductivity in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. The field dependence of the specific heat at the field direction parallel to the plane demonstrates the behavior that seems to be incompatible with the Josephson coupling between the layers. Further investigations are necessary in order to solve this problem. In addition, we estimated the upper critical field perpendicular to the layers to be about 4 T.

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