Non-saturating upper critical field of organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl

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Abstract

The layered superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl in the magnetic field applied strictly parallel to the superconducting plane shows the upper critical field exhibiting non-saturating behavior toward 0 K. The relation to the spatially modulated order-parameter state predicted by Fulde-Ferrell and Larkin-Ovchinnikov is argued.

Key words: Upper critical field; Fulde-Ferrell-Larkin-Ovchinnikov state; organic superconductors

In layered superconductors, when a magnetic field is applied parallel to the conducting plane, the orbital magnetic effect due to the electron motion is depressed and a high magnetic field is required to break the superconductivity (SC) pairing [1]. This enables the spin polarization effect (Pauli paramagnetic effect) to work in determining the upper dritical field H_{c2} [2,3]. When the SC is of singlet pairing, H_{c2} is bounded by the Pauli paramagnetic limit H_P, due to compensation of the condensation energy by the spin polarization energy. For the weak-coupling BCS superconductors, H_P is given by $H_P^{BCS} = 1.84T_c$ at 0 K, where H_P^{BCS} and T_c are given in units of T and K, respectively [1]. In not a few organic superconductors, the upper critical field in the parallel magnetic field ($H_{c2\parallel}$) exceeds H_P^{BCS} [4]. As the reasons, the effects of the strong coupling and the many body effects in the normal state are pointed out [5].

We report the temperature dependence of $H_{c2\parallel}$ of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl under the pressure of 1∼2 kbar, applied to induce SC and adjust T_c so as to cover the whole temperature-versus-magnetic-field phase diagram with a 17 T superconducting solenoid, simultaneously keeping the T_c as high as possible. The H_{c2} was determined from the transition mid-point of the inter-plane resistance temperature dependence [6].

The $H_{c2\parallel}(T)$'s, normalized by $T_c(=H_P^{BCS}/1.84)$, of κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl under 1.0, 1.3 and 1.9 kbar are shown in Fig. 1. The change in the slope of $H_{c2\parallel}(T)$ appearing near T_c is ascribed to the dimensional crossover associated with the temperature dependence of the coherence length [6].

It is noteworthy that the temperature dependence of $H_{c2\parallel}$ is not of saturation type: at low temperatures $H_{c2\parallel}$ continues to increase toward 0 K as seen for the 1.9 kbar data. The value of $H_{c2\parallel}(0)/H_P^{BCS}$ reaches 1.3. The linear temperature dependence in the high-temperature side, except for the proximity to T_c, can be explained in terms of the orbital magnetic field effect according to the Ginzburg-Landau (GL) theory [7,8]. The tendency of the slope decreases with pressure is also consistent with the GL theory since the pressure increases the interlayer coupling, resulting in the enhancement of the orbital effect even in parallel magnetic fields [5].

In the low temperature side, when the Pauli paramagnetic effect dominates, H_{c2} is expected to saturate towards H_P, even if the value becomes larger than H_P^{BCS} due to the strong coupling and the many-body

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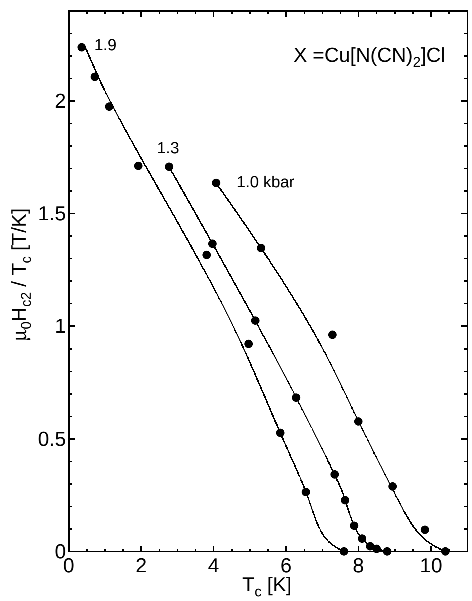

Fig. 1. Temperature dependence of H_{c2} defined by the midpoint of the resistive transition in parallel magnetic fields under pressures of 1.0, 1.3 and 1.9 kbar.

effect. For the orbital effect, H_{c2} is also expected to saturate [7]. It is also shown that H_{c2} in dirty superconductors saturates under the mixed contribution of the orbital and paramagnetic effects [9]. These all considerations contrasts with the almost linear temperature dependence as seen under pressure of 1.9 kbar in our experiment.

As a possible way to interpret the non-saturating behavior of $H_{c2\parallel}(T)$ at low temperatures, we point out the formation of the modulated order-parameter state predicted by Fulde-Ferrel and Larkin-Ovchinikov (FFLO) [10,11]. The FFLO state can be a solution of the gap equation in the Pauli paramagnetic limit region for clean superconductors. Theoretical studies have revealed that the two-dimensional clean superconductor in the parallel magnetic field is favorable to form FFLO state [12] and $H_{c2\parallel}$ can be larger than H_P [13]. In this case, H_{c2} exceeds H_P below $0.56T_c$ and increases all the way to T=0, resulting in non-saturating temperature dependence of $H_{c2\parallel}$ [14]. The clear observation of the quantum oscillations in the normal state of this material supports that the present salt is a clean superconductor. Indeed, the FFLO state is claimed in the organic superconductors like κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ [15] and λ-(BETS)$_2$GaCl$_4$ [16].

The angular dependence of the resistance in the proximity to the parallel field direction is extremely sharp: the control less than 0.1° is required. This is ascribable to the crossover with the vortex state caused by the magnetic field component perpendicular to the superconducting plane. The FFLO state can be influenced by the degree of excitation, for example by the current level, on the resistive transition. The current level dependence, showing the depression of the zero resistance at lower magnetic field, is in accordance with the expection, although the influence of the heating is not fully ruled out in the current dependence experiment [6].

In conclusion, H_{c2} for the pressurized κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl in the field direction exactly parallel to the superconducting plane shows non-saturating behavior at low temperature towards 0 K and exceeds the BCS Pauli paramagnetic limit. The behavior in $H_{c2\parallel}$ is consistent with the formation of the spatially modurated order parameter (FFLO) state.

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