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# Two-dimensional superconductivity in the layered organic superconductor $\kappa_H$ -(DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](THF) with thick dielectric insulating layers

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We have systematically investigated the resistive superconducting transition in the layered organic superconductor  $\kappa_H$ -(DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](THF) [where DMEDO-TSeF is dimethyl(ethylenedioxy) tetraselenafulvalene and THF is tetrahydrofuran], which consists of two crystallographically independent conducting layers and one independent thick dielectric insulating layer. Applying a slight pressure of up to 0.35 GPa suppresses the superconducting phase. The angular dependence of the upper critical field and the short interlayer coherence length indicate that the present compound is a highly two-dimensional superconductor. The upper critical field parallel to the conducting layers exceeds the Pauli paramagnetic limit. These superconducting properties are consistent with the crystal structure in which the superconducting layers are well separated by a thick anion insulating layer.

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## I. INTRODUCTION

Most layered organic superconductors have one crystallographically independent conducting layer.<sup>1,2</sup> Two new organic superconductors with the same chemical composition,  $\kappa_L$ - and  $\kappa_H$ -(DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](THF) (see Fig. 1), have been developed.<sup>3,4</sup> The  $\kappa_L$  phase (onset  $T_c = 3.0$  K) has a disordered solvent molecule THF and exhibits an orthorhombic–monoclinic distortion phase transition at  $T_d = 209$  K.<sup>5–7</sup> The low-temperature structure consists of two monoclinic domains. On the other hand, the THF molecule of the  $\kappa_H$  phase (onset  $T_c = 4.8$  K) is ordered even at room temperature. Moreover, there are two crystallographically independent conducting layers (A and B) [see Fig. 1(b)] and both layers have a  $\kappa$ -type donor arrangement. We consider that both layers exhibit superconductivity because the volume fraction of superconductivity at 1.9 K is about 90% that of perfect diamagnetism.<sup>3</sup> This compound is a rare organic superconductor, having two independent conducting layers. Other organic superconductors with two independent conducting layers have one- and two-dimensional (2D) conducting layers,<sup>8,9</sup> whereas the present compound has two kinds of 2D  $\kappa$ -type conducting layers.

The dimensionality of the electronic states of the layered conductors is related to the magnitude of the interlayer interaction. The interlayer interaction depends on the distance between the conducting layers and the thick anion layers form a well-separated layered conducting system. Such a layered system has a highly 2D electronic state. Both  $\kappa_L$ - and  $\kappa_H$ -(DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](THF) superconductors have thicker anion layers than other organic superconductors.<sup>3</sup>

The present compounds have THF solvent molecules in the anion insulating layers. The THF molecule is a *polar* molecule and exhibits ferroelectric ordering in the insulating layer of the  $\kappa_H$  phase, even at room temperature [Fig. 1(c)]; the relationship between the insulating layers is antiferroelectric. The polarized THF polarizes the anion [Au(CN)<sub>4</sub>]<sup>−</sup>. This anion has four crystallographically nonequivalent CN units, in agreement with the polarization. The present organic superconductor,  $\kappa_H$ -(DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](THF), may exhibit Ginzburg's mechanism of superconductivity because its superconducting layers are sandwiched between dielectric insulating layers.<sup>10</sup>

The present paper reports the superconducting properties of the  $\kappa_H$  phase in terms of its transport properties. The results reveal that the present compound is a highly 2D superconductor with a short coherence length perpendicular to the conducting layer and that the superconducting phase is sensitive to the applied pressure.

## II. EXPERIMENT

Single crystals were prepared by electrocrystallization.<sup>3</sup> The crystal has the shape of hexagonal plate with the typical size of  $0.4 \times 0.3 \times 0.05$  mm<sup>3</sup>. High-pressure resistance measurements were performed using a clamped piston-cylinder cell consisting of a BeCu cylinder with Daphne 7373 oil as the pressure-transmitting medium; the sample was cooled to 1.6 K. The room-temperature pressure was determined by measuring the resistance of a Manganin wire with a pressure coefficient of 2.4%/GPa.<sup>11</sup> Because the pressure decreased by about 0.15 GPa between 300 and 50 K, this pressure was subtracted

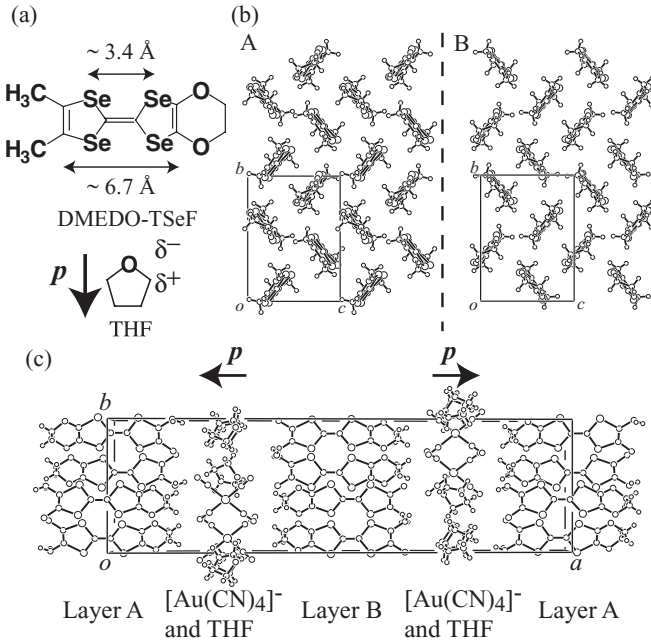


FIG. 1. (a) DMEDO-TSeF and THF molecules. THF is a polar molecule. (b) Crystal structure projected along the long molecular axis and (c) projection onto the  $ab$  plane of  $\kappa_H$ -(DMEDO-TSeF) $_2$ [Au(CN) $_4$ ](THF). A and B are two crystallographically independent conducting layers. The conducting layers of DMEDO-TSeF dimers are separated by insulating anion layers, where the insulating layer is composed of [Au(CN) $_4$ ] $^-$  and polar THF molecules. The arrows both in (a) and (c) denote the polarization vector  $\mathbf{p}$  of THF molecules.

from the pressures measured at room temperature.<sup>12</sup> The resistance measurements were performed by the four-probe method along the  $a^*$  axis (the interlayer resistance) with an ac current (0.01–0.3  $\mu$ A). For magnetoresistance measurements, the samples were mounted on a two-axis rotator in a cryostat in a 17 T superconducting magnet and were cooled to 1.6 K. For magnetoresistance measurements below 1.6 K, the samples were mounted in a dilution refrigerator in a 20 T superconducting magnet with one degree of rotational freedom with respect to the magnetic field and were cooled to 35 mK. All magnetoresistance measurements were performed at the National Institute for Materials Science.

### III. RESULTS

Figure 2(a) shows the temperature dependence of the electrical resistance for six different pressures. The resistance decreases with decreasing temperature, even at ambient pressure. In our measurements, the superconducting transition temperature at ambient pressure is  $T_c = 4.4$  K (midpoint  $T_c$ ). Applying a slight pressure drastically reduces the room-temperature resistance.  $T_c$  decreases gradually with increasing pressure and superconductivity disappears at around 0.35 GPa above 1.6 K. Figure 2(b) shows the temperature–pressure phase diagram. The superconducting phase exists in a narrow pressure range. The pressure dependence of  $T_c$  displays a power-law-like behavior.

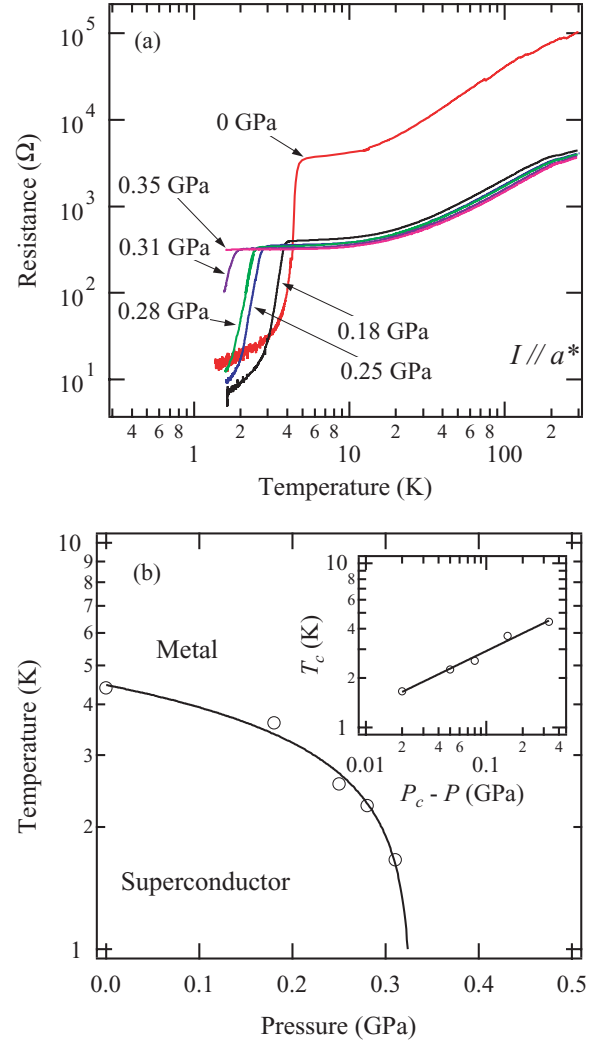


FIG. 2. (Color online) (a) Temperature dependence of the resistance for six pressures. (b) The phase diagram of  $\kappa_H$ -(DMEDO-TSeF) $_2$ [Au(CN) $_4$ ](THF). The inset in (b) shows the  $T_c \propto (P_c - P)^\alpha$  dependence, where  $P_c = 0.33(2)$  and  $\alpha = 0.35(8)$ . The solid line in (b) shows the same  $T_c \propto (P_c - P)^\alpha$  dependence.

Figure 3 shows the magnetic field dependence of the electrical resistance for various field directions and temperatures at ambient pressure. The upper critical fields,  $H_{c2}$ , are determined from 50% recovery of the extrapolated magnetoresistance  $R(H)$  in the higher field region in Fig. 3. In several highly 2D superconductors including high- $T_c$  cuprates, the curvature of the resistive transition field,  $H_{c2}(T)$ , is reported to vary depending on the definition of the recovery percentage and in the low resistance limit, the shape of  $H_{c2}(T)$  perpendicular to the conducting sheet approaches the irreversibility line.<sup>13,14</sup> Moreover, flux-flow resistance gives a finite resistance even in the superconducting state. In  $\kappa$ -(BEDT-TTF) $_2$ Cu(NCS) $_2$  [BEDT-TTF: bis(ethylenedithio)tetrathiafulvalene], the slope of  $H_{c2}(T)$  determined by the specific heat measurement is larger than that obtained from the magnetoresistance measurement.<sup>15</sup> However, in the present compound, even when  $H_{c2}$  is defined by 10 or 90% recovery, the shape of  $H_{c2}(T)$  does not qualitatively change. This indicates the validity of magnetoresistance as a probe of  $H_{c2}$ .

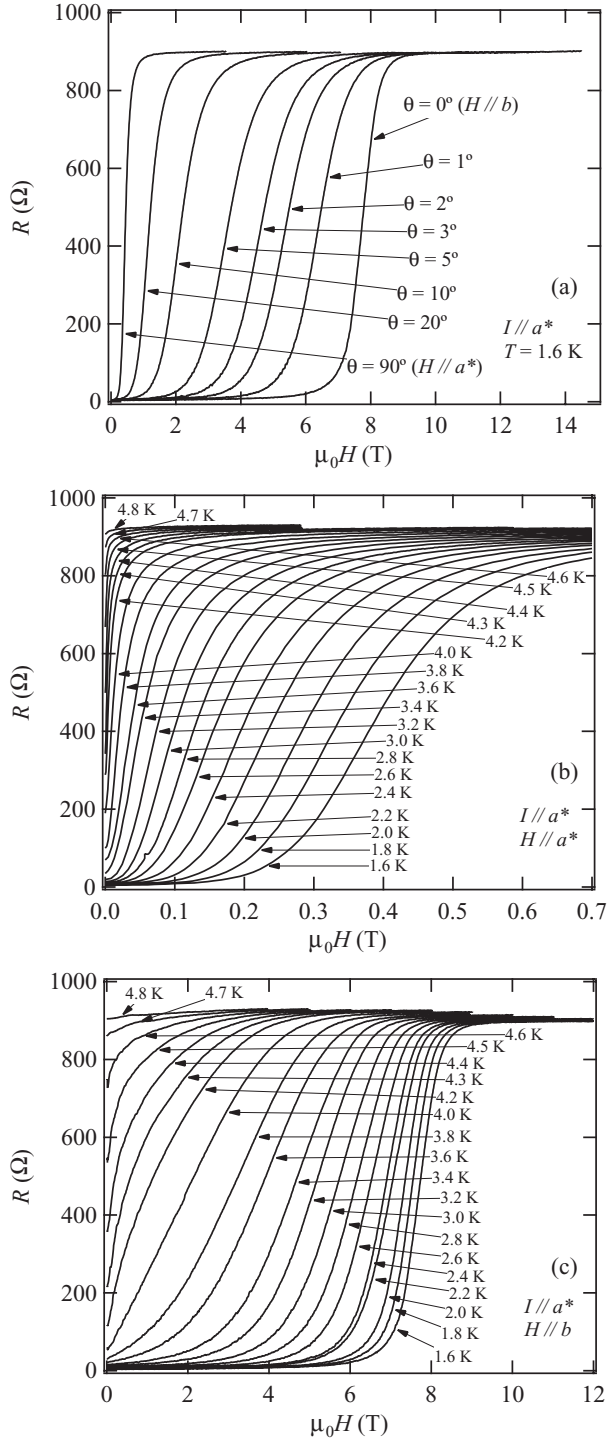


FIG. 3. Magnetic field dependence of resistance for various (a) field directions and [(b) and (c)] temperatures.

Figure 4 shows the angular dependence of the upper critical fields at 1.6 K. For anisotropic three-dimensional (3D) superconductors, the angular dependence of the upper critical field is given by

$$\left[ \frac{H_{c2}(\theta) \sin \theta}{H_{c2\perp}} \right]^2 + \left[ \frac{H_{c2}(\theta) \cos \theta}{H_{c2\parallel}} \right]^2 = 1, \quad (1)$$

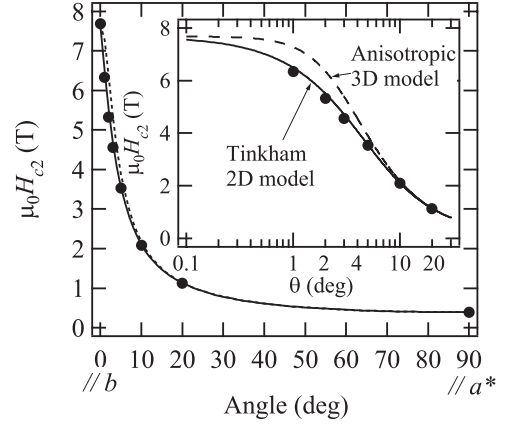


FIG. 4. Angular dependence of the upper critical field at 1.6 K. The solid and dotted lines indicate the results of calculations based on the Tinkham 2D model and the anisotropic 3D model, respectively. The inset shows the low-angle region.

where  $\theta$  is the angle of the field from the conducting plane ( $\parallel bc$  plane) and  $H_{c2\parallel}$  and  $H_{c2\perp}$  are, respectively, the upper critical fields parallel and perpendicular to the conducting plane.<sup>16</sup> For 2D superconductors, the angular dependence of the upper critical field varies according to:<sup>16</sup>

$$\left| \frac{H_{c2}(\theta) \sin \theta}{H_{c2\perp}} \right| + \left( \frac{H_{c2}(\theta) \cos \theta}{H_{c2\parallel}} \right)^2 = 1. \quad (2)$$

The difference between the 3D and 2D models is large near  $\theta = 0^\circ$ ; the 2D curve has a cusp for the magnetic field parallel to the conducting sheet. The observed upper critical fields ( $H_{c2\parallel}$  and  $H_{c2\perp}$ ) are well reproduced by the 2D model (Fig. 4). The present compound is recognized as being a 2D superconductor. A 2D angular dependence of the upper critical field has also been observed in some organic superconductors.<sup>7,17–21</sup>

Figure 5 shows the temperature dependence of the upper critical fields. We can estimate the Ginzburg–Landau (GL) coherence lengths at  $T = 0$  K using the following relation:<sup>22</sup>

$$-\frac{dH_{c2\perp}(T)}{dT} \Big|_{T=T_c} = \frac{\Phi_0}{2\pi\mu_0\xi_{\parallel}^2 T_c} \quad (3)$$

$$-\frac{dH_{c2\parallel}(T)}{dT} \Big|_{T=T_c} = \frac{\Phi_0}{2\pi\mu_0\xi_{\parallel}\xi_{\perp} T_c}, \quad (4)$$

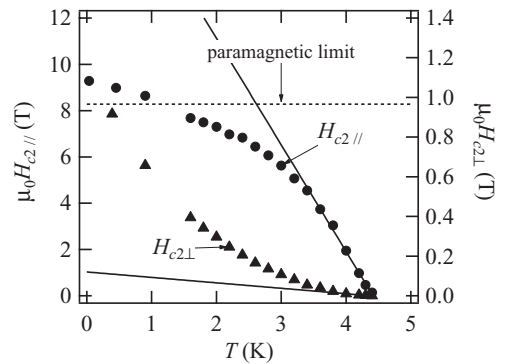


FIG. 5. Temperature dependence of the upper critical fields. The solid lines show the slope of  $H_{c2}(T)$  around  $T = T_c$ .



where  $\Phi_0$  is the flux quantum,  $\mu_0$  is the magnetic constant, and  $\xi_{\parallel}$  and  $\xi_{\perp}$  are, respectively, the GL coherence lengths parallel and perpendicular to the conducting layers at  $T = 0$  K. The coherence lengths estimated from the slope of  $H_{c2}(T)$  at  $T_c$  are  $\xi_{\parallel} = 530(20)$  Å and  $\xi_{\perp} = 3.1(3)$  Å. The transverse coherence length,  $\xi_{\perp}$ , is considerably shorter than the thickness of the effective conducting sheet,  $[a \sin(\beta)]/2 = 19.359(3)$  Å, indicating that the superconductivity is 2D. The superconducting anisotropy parameter,  $\gamma$ ,<sup>16</sup> is defined as

$$\gamma = \frac{\left. \frac{dH_{c2\parallel}(T)}{dT} \right|_{T=T_c}}{\left. \frac{dH_{c2\perp}(T)}{dT} \right|_{T=T_c}} = \frac{\xi_{\parallel}}{\xi_{\perp}}, \quad (5)$$

and it is 170(15).

#### IV. DISCUSSION

Although the present compound is a well-separated layered system at ambient pressure, the room temperature resistance rapidly decreases at a slight applied pressure  $\sim 0.18$  GPa. This indicates that the pressure drastically changes the value of the interlayer interaction. However, the reason of such a sensitive interlayer interaction is not clear. The temperature dependence of the resistance,  $R(T)$ , in  $\kappa$ -type BEDT-TTF conductors has a broad peak around 100 K.<sup>1,2</sup> Moreover, the  $\kappa_L$  phase of (DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](THF) and other  $\kappa$ -(DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](solvent) superconductors also have a peak structure around 100 K.<sup>3,4</sup> By contrast, the resistance of the present compound decreases monotonically with decreasing temperature despite its  $\kappa$ -type structure [Fig. 1(a)]. The origin of this difference between the  $\kappa_H$  phase and the other phases is not clear.

In the present compound, the conducting donor layers are separated by insulating layers composed of [Au(CN)<sub>4</sub>]<sup>−</sup> and THF molecules. Moreover, there are two crystallographically independent conducting layers, A and B. Therefore, this compound should be modeled by the  $S - I - S' - I \dots$  layer structure, where  $S(S')$  and  $I$  denote the superconductor and insulator, respectively. Although the present compound has two nonequivalent superconducting layers, it is not clear that the superconducting transition temperature of the layer A is the same as that of B. If  $T_c$  of the layer A differs from that of B, it is theoretically expected that the anisotropy of the London penetration depth rapidly decreases with decreasing temperature below the highest  $T_c$ .<sup>23</sup> However, our results cannot clarify this problem. The interlayer coherence length,  $\xi_{\perp}$ , is comparable to the distance between the Se atoms of the TSeF unit along the long molecular axis ( $\sim 3.4$  Å), as shown in Fig. 1(a). This indicates that Cooper pairs condense into the TSeF unit in the superconducting phase. The anisotropic coherence lengths are almost the same as those of the  $\kappa_L$  phase (see Table I) despite the difference in  $T_c$ .<sup>7</sup> The present compound has a much larger anisotropy parameter  $\gamma$  than other organic superconductors ( $\gamma \sim 10$ –50).<sup>2</sup> The values of  $\gamma$  for both the  $\kappa_L$  and  $\kappa_H$  phases are close to that of the high- $T_c$  cuprate, Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+x</sub>.<sup>2,16</sup>

There are two limits for the critical magnetic field at which superconductivity is destroyed. The first limit is the orbital effect. The expected zero temperature critical field is given by  $H_{\text{orb}} = 0.7 T_c |dH_{c2}/dT|_{T=T_c}$ .<sup>24</sup> For the present compound,

TABLE I. The midpoint superconducting transition temperature  $T_c^{\text{mid}}$ , the GL coherence lengths  $\xi_{\parallel}$  and  $\xi_{\perp}$ , the anisotropy parameter  $\gamma$ , the orbital limit  $\mu_0 H_{\text{orb}}$ , the Pauli limit  $\mu_0 H_P$ , and the Maki parameter  $\alpha$  for  $\kappa_L$ - and  $\kappa_H$ -(DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](THF). The results for the  $\kappa_L$  phase from a previous study.<sup>7</sup>

	$\kappa_L$ phase	$\kappa_H$ phase
$T_c^{\text{mid}}$ (K)	2.2	4.4
$\xi_{\parallel}$ (Å)	510(30)	530(20)
$\xi_{\perp}$ (Å)	3.5(5)	3.1(3)
$\gamma$	150(20)	170(15)
$\mu_0 H_{\text{orb}}$ (T)	$\sim 13$	$\sim 14$
$\mu_0 H_P$ (T)	$\sim 4.0$	$\sim 8.1$
$\alpha$	$\sim 4.6$	$\sim 2.5$

the orbital limit estimated from the slope of  $H_{c2\parallel}(T)$  at  $T_c$  in Fig. 4 is about 14 T. For a layered superconductor, the parallel critical field can be rewritten as

$$H_{c2\parallel} = \frac{\Phi_0}{2\pi\mu_0} \frac{\sqrt{12}}{\xi_{\parallel} t} \quad (6)$$

by considering the thickness of the superconducting layer,  $t$ .<sup>25</sup> The present compound satisfies the relationship between the insulating layer thickness and the coherence length ( $\xi_{\perp} < d_i/\sqrt{2}$ ), where we define the insulating layer thickness,  $d_i \approx 4.7$  Å, as the distance between the planes composed of the outside edge of the hydrogen atoms of DMEDO-TSeF molecules. Setting the corresponding values,  $\xi_{\parallel} = 530(20)$  Å and  $t = [a \sin(\beta)]/2 - d_i \approx 14.66$  Å, we find  $\mu_0 H_{c2\parallel} \sim 15$  T. This estimated value is comparable to experimentally obtained  $\mu_0 H_{\text{orb}}$ .

The second limit originates in the Pauli pair breaking mechanism; the external magnetic field destroys the spin-singlet state of the Cooper pair, imposing the Clogston–Chandrasekhar paramagnetic limit.<sup>26,27</sup> For a BCS superconductor, this is given by  $\mu_0 H_P = 1.84 T_c$  T/K. For the present compound, the paramagnetic limit is about 8.1 T when  $T_c = 4.4$  K. Figure 6 shows a plot of the critical fields of different materials,  $\kappa_L$  and  $\kappa_H$  phases, scaled by

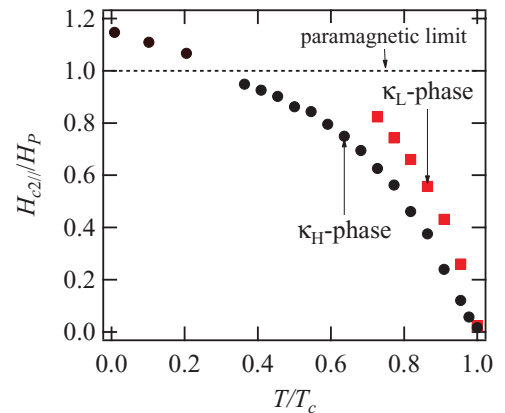


FIG. 6. (Color online) The critical fields with the magnetic field parallel to the conducting layers of  $\kappa_L$ - and  $\kappa_H$ -(DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](THF) as a function of temperature. The results for the  $\kappa_L$  phase are from a previous study.<sup>7</sup>

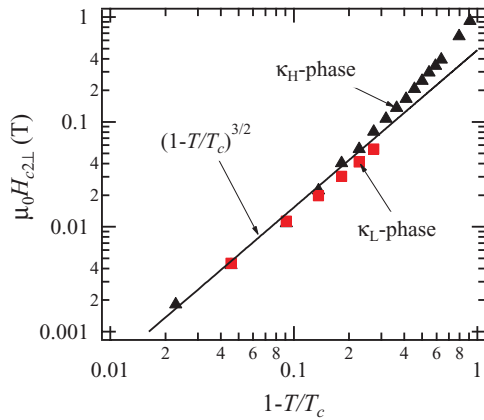


FIG. 7. (Color online) Upper critical fields perpendicular to the conducting layers. The solid line indicates the  $(1 - T/T_c)^{3/2}$  dependence. The results for the  $\kappa_L$  phase are from a previous study.<sup>7</sup>

their critical temperatures and Pauli limits. The upper critical field parallel to the conducting sheet of the  $\kappa_H$  phase exceeds the paramagnetic limit below  $T/T_c \sim 0.3$  and does not exhibit saturation behavior. This is similar to that of  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu(NCS)<sub>2</sub>.<sup>28</sup> However, the actual value of the Pauli limit is increased by the factor  $(1 + \lambda_{ep})^{1/2}$ , where  $\lambda_{ep}$  is the electron-phonon coupling constant.<sup>22,26</sup> The simple relationship,  $\mu_0 H_P = 1.84 T_c$ , is true in the weak-coupling limit. Therefore, the real value of  $H_P$  might be larger than our estimation.

These limits for the critical field give the Maki parameter,  $\alpha = \sqrt{2}H_{orb}/H_P$  (see Table I).<sup>29</sup> The Maki parameter of the  $\kappa_H$  phase is smaller than that of the  $\kappa_L$  phase. Although the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state may exist below  $T/T_c = 0.55$  for superconductors with  $\alpha > 1.8$ ,<sup>30</sup> the  $\kappa_H$  phase does not show a clear anomaly below  $T/T_c = 0.55$  in Fig. 6. Several research groups have recently found the phase transition from the conventional superconducting state to the FFLO state in several layered organic superconductors

by the penetration depth,<sup>31</sup> magnetic torque,<sup>32</sup> and specific heat measurements.<sup>33</sup> For the 2D organic superconductor  $\beta''$ -(BEDT-TTF)<sub>2</sub>SF<sub>5</sub>CH<sub>2</sub>CF<sub>2</sub>SO<sub>3</sub> with the anisotropy parameter  $\gamma = 18$ ,<sup>34</sup> the FFLO phase transition has been detected below  $T/T_c = 0.24$  using a tunnel diode oscillator.<sup>31</sup> However, the magnetoresistance does not show a clear anomaly in this temperature region.<sup>35</sup> This indicates the difficulty in detecting the FFLO phase transition by magnetoresistance measurements.<sup>36</sup>

Finally, we discuss the interlayer upper critical field. The interlayer upper critical field,  $H_{c2\perp}(T)$ , shows upward curvature and exhibits a  $(1 - T/T_c)^{3/2}$  dependence, which has been also observed in the  $\kappa_L$  phase, as shown in Fig. 7.<sup>7</sup> In the  $\kappa_H$  phase,  $H_{c2\perp}$  gradually deviates from this power-law dependence below  $T/T_c \sim 0.7$ . Many superconductors including high- $T_c$  cuprates exhibit this behavior but its origin is still controversial.<sup>37–40</sup>

## V. CONCLUSION

The room-temperature resistance decreases rapidly with increasing pressure. The superconducting phase is sensitive to the pressure and exists in a narrow pressure range. The angular dependence of the upper critical field and the short coherence length perpendicular to the conducting sheet demonstrate that  $\kappa_H$ -(DMEDO-TSeF)<sub>2</sub>[Au(CN)<sub>4</sub>](THF) is a highly 2D superconductor. The upper critical field parallel to the conducting layers exceeds the paramagnetic limit but is smaller than the orbital limit. Although the present compound may exhibit the FFLO state at low temperature, the magnetoresistance measurements have not detected the FFLO phase transition.

## ACKNOWLEDGMENTS

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