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Phase transition to the Fulde–Ferrell–Larkin–Ovchinnikov state in a quasi-one-dimensional organic superconductor with anion order

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Abstract. A theoretical study is presented on the effect of anion order on the phase transition from the normal state to a Fulde–Ferrell–Larkin–Ovchinnikov superconducting state in a quasi-one-dimensional organic superconductor $(TMTSF)_2ClO_4$. The temperature dependence of the upper critical field $H_{c2}(T)$ is examined. In the absence of anion order, the $H_{c2}(T)$ curve exhibits a one- to two-dimensional crossover with decreasing temperature T. For a sufficiently large anion order parameter, the dimensional crossover disappears and then a kink appears in the $H_{c2}(T)$ curve.

1. Introduction

The realization of the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) state has been suggested in low-dimensional organic superconductors [1, 2]. The low-dimensionality is advantageous to the FFLO state because the upper critical field H_{c2} is not so strongly reduced by the orbital pair-breaking effect when a magnetic field is applied parallel to the conductive layer. Moreover, anisotropy of the Fermi surface helps stabilize the FFLO state [3, 4].

Recently, Yonezawa *et al.* reported measurements of the superconducting onset temperature $T_c^{\rm onset}$ of a quasi-one-dimensional (Q1D) organic superconductor $(TMTSF)_2ClO_4$ in magnetic fields $\boldsymbol{H}$ aligned with the conductive plane. They found a characteristic field-direction dependence of $T_c^{\rm onset}$ suggestive of the transition to the FFLO superconducting phase [5]. Lebed showed that the observed H_{c2} in $\boldsymbol{H}$ parallel to the b' axis (parallel to the conducting ab plane and perpendicular to the most conductive a axis) agrees in magnitude with a theory in which the FFLO state with d-wave pairing symmetry is assumed [6], Croitoru *et al.* demonstrated how the orbital magnetism in the FFLO state causes the in-plane anisotropy of $T_c^{\rm onset}$ [7]. Miyawaki and Shimahara examined the temperature dependence of H_{c2} with a focus on the effect of the anisotropic Fermi surface and revealed that a novel dimensional crossover of the $H_{c2}(T)$ curve appears for $\boldsymbol{q} \parallel \boldsymbol{a}$ [8], where $\boldsymbol{q}$ is the center-of-mass momentum of Cooper pairs and $\boldsymbol{a}$ is the lattice vector of the most conducting chain of $(TMTSF)_2ClO_4$.

In this paper, the previous theory [8] is extended to investigate $H_{c2}(T)$ in $(TMTSF)_2ClO_4$ on the basis of a more realistic band structure. Specifically, the effect of anion (ClO_4^-) order [9], which is known to cause a splitting of the Q1D Fermi surface, is taken into account. It is shown that the finite anion gap Δ_{AO} gives rise to a qualitative difference in the temperature dependence of the FFLO upper critical field H_{c2}.

2. Model and method

We consider the Q1D tight-binding Hamiltonian [10]

$$H_{AO} = -\sum_{i,j,\sigma} t_{ij} a^\dagger_{i\sigma} a_{j\sigma} + \sum_{i,\sigma} \epsilon_i a^\dagger_{i\sigma} a_{i\sigma} - \sum_{i,\sigma} (\mu + \sigma\mu_e H) a^\dagger_{i\sigma} a_{i\sigma}, \tag{1}$$

where μ is the chemical potential and $\sigma\mu_e H$ ($\sigma = \pm$) is the Zeeman energy. In this model, two kinds of chains (A and B) are defined in the a direction. There is an energy difference between the A- and B-site due to the anion order. This energy difference is accounted for by the parameter

$$\epsilon_i = \begin{cases} \Delta_{AO} & \text{for } i \in A \\ -\Delta_{AO} & \text{for } i \in B \end{cases}. \tag{2}$$

For t_{ij}, only the nearest neighbor hopping is taken into account, and we define $t_{ij} = t_a$ (t_b) for the intra-chain (inter-chain) nearest neighbor hopping.

Diagonalizing this Hamiltonian, we obtain the dispersion [10, 11, 12]

$$\xi^s_\sigma(\boldsymbol{k}, H) = -2t_a \cos k_x - s\sqrt{(2t_b \cos k_y)^2 + {\Delta_{AO}}^2} - \mu - \sigma\mu_e H, \tag{3}$$

where the momentum k_x (k_y) is scaled by the inverse of the lattice constant a (b). There are two electron bands in the Brillouin zone $|k_y| < \pi/2$, corresponding to $s = \pm$. In the organic conductor $(TMTSF)_2ClO_4$, the hole number per site is 0.5 and thus the chemical potential μ is determined by the 1/4-filled condition. The anion gap Δ_{AO} is suggested to be 25 meV by magnetoresistance measurements [11]. Another estimation based on the Hückel method gives $\Delta_{AO} \sim 100$ meV [13]. From recent first-principles calculations, however, Δ_{AO} has been claimed to be nearly zero [14] or ∼14 meV [15]. We thus treat Δ_{AO} as a parameter that is less than $t_a \sim 300$ meV.

In the superconducting state, we take into account only intra-band Cooper pairing. We then obtain the following Bogoliubov-quasiparticle energy in an FFLO state with the superconducting gap function $\Delta(\boldsymbol{r}, \boldsymbol{k}) = \Delta(\boldsymbol{k}) e^{i\boldsymbol{q}\cdot\boldsymbol{r}}$:

$$E^s_{\boldsymbol{k}\sigma} = \sigma\zeta^s + E^s_{\boldsymbol{k}}, \tag{4}$$

where

$$E^s_{\boldsymbol{k}} = \sqrt{\xi^s_\sigma(\boldsymbol{k}, 0)^2 + \Delta(\boldsymbol{k})^2}, \tag{5}$$

$$\zeta^s = \frac{1}{2} \boldsymbol{v}^s_F(\boldsymbol{k}) \cdot \boldsymbol{q} - \mu_e H. \tag{6}$$

In Eq. (6), $\boldsymbol{v}^s_F(\boldsymbol{k})$ is the Fermi velocity for the s band.

The gap $\Delta(\boldsymbol{k})$ is determined self-consistently from

$$\Delta(\boldsymbol{k}') = \frac{1}{N} \sum_{s=\pm} \sum_{\boldsymbol{k}} V(\boldsymbol{k}', \boldsymbol{k}) \frac{1 - f(E^s_{\boldsymbol{k}\uparrow}) - f(E^s_{\boldsymbol{k}\downarrow})}{2E^s_{\boldsymbol{k}}} \Delta(\boldsymbol{k}), \tag{7}$$

where $V(\boldsymbol{k}', \boldsymbol{k}) = V\gamma(\boldsymbol{k}')\gamma(\boldsymbol{k})$ is the pairing interaction. According to Ref. [6], we assume a d-wave symmetry of the gap, i.e., $\Delta(\boldsymbol{k}) = \Delta_0\gamma(\boldsymbol{k})$ with $\gamma(\boldsymbol{k}) = \sqrt{2}\cos k_y$. The normal–superconducting phase boundary is determined by linearizing the above gap equation with respect to Δ_0. From the linearized gap equation, we can obtain

$$\ln\frac{T}{T_{\rm c}^{(0)}} = -\sum_{s=\pm}\int_{-\pi/2}^{\pi/2}\frac{dk_y}{2\pi}\frac{\rho^s(0,k_y)\gamma^2(k_y)}{N_d(0)}\sinh^2\frac{\beta\zeta^s}{2}\int_0^\infty dy\ln y \times\left[\frac{2\sinh^2 y}{\left[\cosh^2 y+\sinh^2(\beta\zeta^s/2)\right]^2}-\frac{1}{\cosh^2 y\left[\cosh^2 y+\sinh^2(\beta\zeta^s/2)\right]}\right] \tag{8}$$

with

$$N_d(0) = \sum_{s=\pm}\int_{-\pi/2}^{\pi/2}\frac{dk_y}{2\pi}\rho^s(0,k_y)\gamma^2(k_y). \tag{9}$$

Here, $\rho^s(0,k_y)$ is the Fermi surface value of the density of states defined through

$$\frac{1}{N}\sum_{\boldsymbol{k}}(\cdots) = \int d\xi\sum_{s=\pm}\int_{-\pi/2}^{\pi/2}\frac{dk_y}{2\pi}\rho^s(\xi,k_y)(\cdots). \tag{10}$$

Equation (8) determines $H_{\rm c2}$ as a function of $(T, \boldsymbol{q})$. An optimum value of $\boldsymbol{q}$ is fixed such that $H_{\rm c2}$ is maximized at a given temperature T, leading to an $H_{\rm c2}(T)$ phase boundary.

The optimum $\boldsymbol{q}$ occurs when it is directed along $\boldsymbol{a}$, as expected from the Fermi-surface nesting consideration [3, 4, 8, 16]. For the optimum value of $q = |\boldsymbol{q}|$, there are two possibilities associated with the nesting vectors $\boldsymbol{q}^+$ and $\boldsymbol{q}^-$ for the outer ($s = +$) and inner ($s = -$) Fermi surfaces (Fig. 1). In fact, when we plot $H_{\rm c2}$ as a function of q, we find that the $H_{\rm c2}(q)$ curve has two local maxima at low temperatures, though it has a single maximum at high temperatures. We shall denote the critical fields at the two local maxima by $H_{\rm c2}^{\pm}$.

3. Results

Figure 2 shows plots of the numerical results of $H_{\rm c2}^+(T)$. The FFLO state appears at low temperatures below $T^* \approx 0.56T_{\rm c}^{(0)}$, where $T_{\rm c}^{(0)}$ is the transition temperature at $H = 0$. When $\Delta_{\rm AO} = 0$ (black solid line), $H_{\rm c2}^+(T)$ curve exhibits a characteristic temperature dependence. Just below T^*, the $H_{\rm c2}^+(T)$ curve has a positive curvature as in the case of the 1D system (dashed line); however, as temperature decreases, the curvature becomes negative and a shoulder appears. As temperature decreases further, the curve exhibits an upturn similar to the 2D system (dotted line). This one- to two-dimensional crossover is typical for Q1D systems [8]. As $\Delta_{\rm AO}$ is increased, the magnitude of $H_{\rm c2}^+(T)$ decreases, and the crossover behavior becomes less pronounced for $\Delta_{\rm AO} \gtrsim 0.3t_a$. This is because the Fermi-surface mismatch between the up- and down-spin Fermi surfaces for $s = -$ at the nesting vector $\boldsymbol{q}^+$ is larger for larger $\Delta_{\rm AO}$.

When $\Delta_{\rm AO} = 0$, the critical field $H_{\rm c2}^+(T)$ is larger than $H_{\rm c2}^-(T)$ throughout the entire temperature region. However, numerical calculations show that the reduction of $H_{\rm c2}^-(T)$ by a finite $\Delta_{\rm AO}$ is less significant than that of $H_{\rm c2}^+(T)$. This is because the density of states $\rho^-(0,k_y)$ is larger than $\rho^+(0,k_y)$ on the nested Fermi surface. As a result, for $\Delta_{\rm AO} \gtrsim 0.3t_a$, the critical field $H_{\rm c2}^-(T)$ becomes larger than $H_{\rm c2}^+(T)$ at low temperatures. This means that the upper critical field $H_{\rm c2}(T)$ observed actually has a kink in its temperature dependence (Fig. 3).

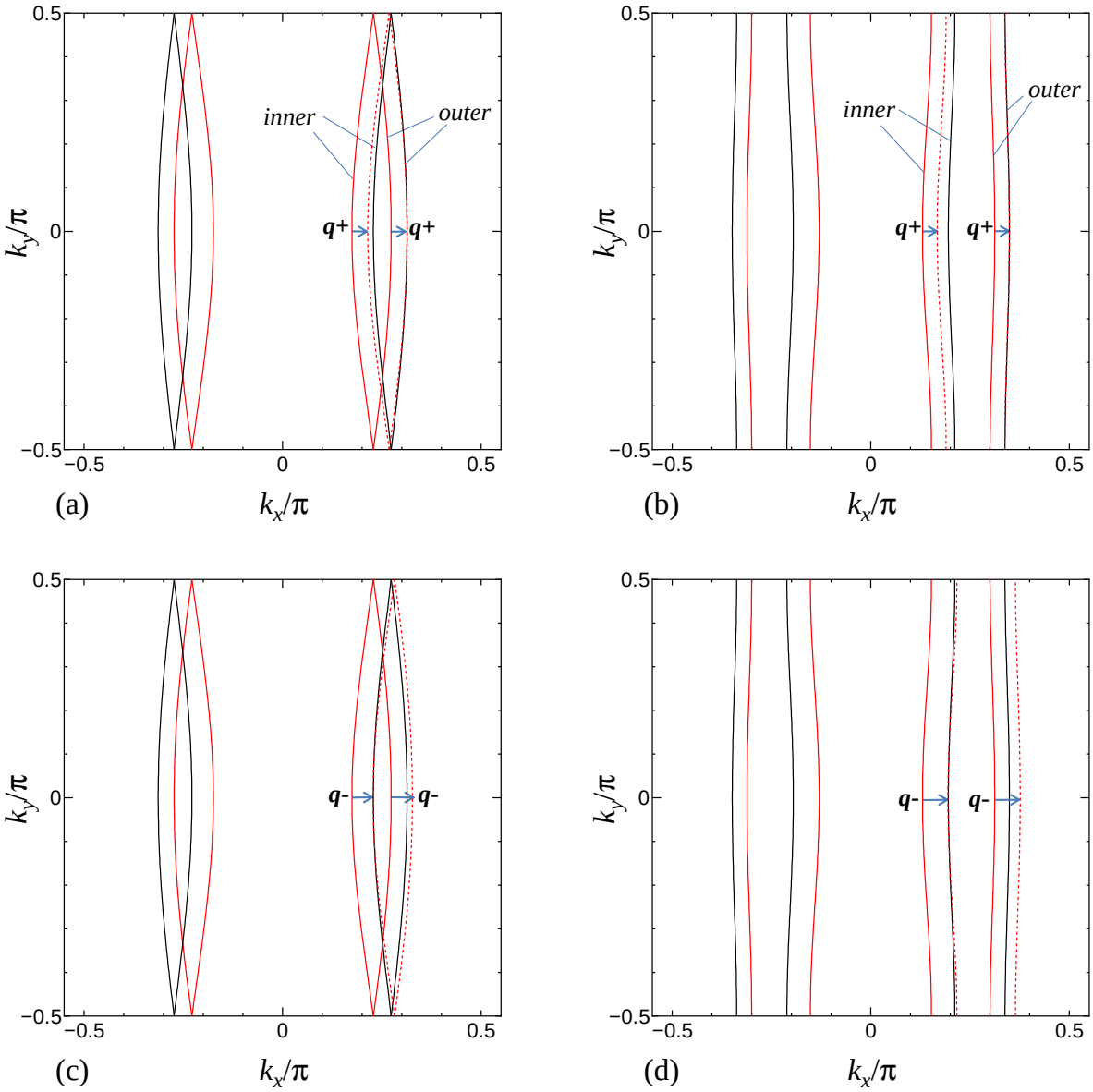

Figure 1. Fermi surfaces for (a),(c) $\Delta_{\rm AO} = 0$ and (b),(d) $\Delta_{\rm AO} = 0.3t_a$. Red and black solid curves are the Fermi surfaces of minority-spin and majority-spin electrons, respectively. The red dotted curves represent the minority-spin Fermi surface shifted by the nesting vector $\boldsymbol{q}^+$ ($\boldsymbol{q}^-$) for the outer (inner) Fermi surface (where the shifted Fermi surface is shown only for the $k_x > 0$ branch).

4. Conclusion

We have examined how the FFLO upper critical field $H_{\rm c2}(T)$ in a Q1D superconductor $(\mathrm{TMTSF})_2\mathrm{ClO}_4$ is modified depending on the anion order. In the absence of the anion gap $\Delta_{\rm AO}$, the $H_{\rm c2}(T)$ curve exhibits a dimensional crossover typical for Q1D systems. For $\Delta_{\rm AO} \gtrsim 0.3t_a$, the dimensional crossover disappears and a kink appears in the $H_{\rm c2}(T)$ curve at a low temperature. If the anion gap $\Delta_{\rm AO}$ is large enough, a kink of $H_{\rm c2}(T)$ should be observed in $(\mathrm{TMTSF})_2\mathrm{ClO}_4$.

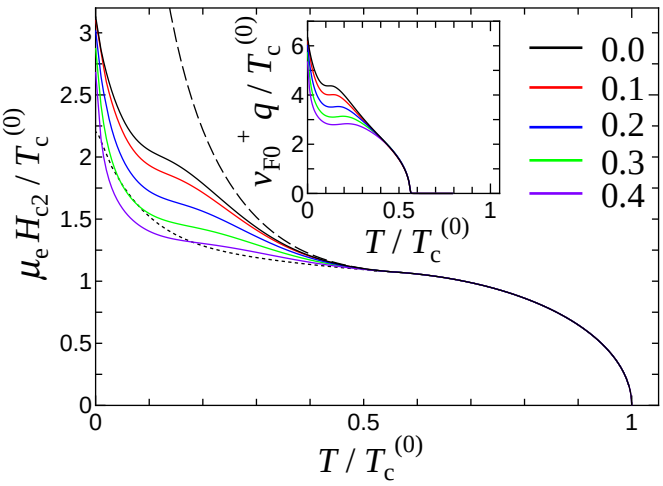

Figure 2. Temperature dependence of $\mu_e H_{c2}^+/T_c^{(0)}$ for $t_b = 0.1t_a$ and $\Delta_{AO} = 0$, $0.1t_a$, $0.2t_a$, $0.3t_a$, and $0.4t_a$. The temperature T is scaled by $T_c^{(0)} = T_c(H = 0)$. For reference, the results for a 1D superconductor ($t_b = 0$ and $\Delta_{AO} = 0$) and for a 2D $d_{x^2-y^2}$-wave superconductor (with a cylindrical Fermi surface) are plotted by dashed and dotted lines, respectively. The inset shows the T dependence of the optimum $v_{F0}^+ q/T_c^{(0)}$. Here, v_{F0}^+ denotes the Fermi velocity $|v_F^+(k_x, k_y = 0)|$.

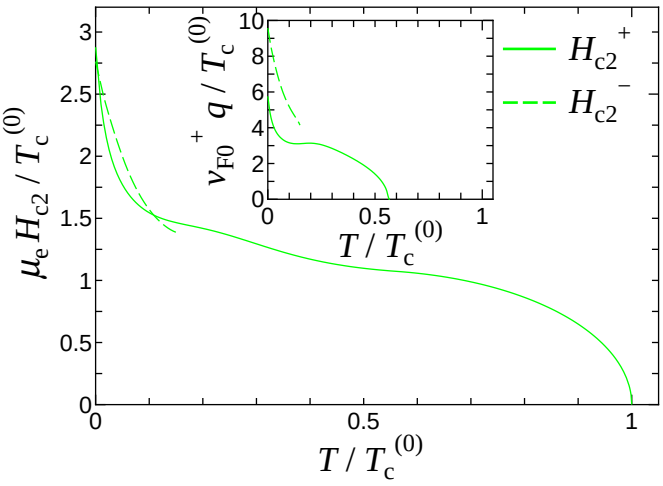

Figure 3. Phase diagram for $t_b = 0.1t_a$ and $\Delta_{AO} = 0.3t_a$. The solid line corresponds to H_{c2}^+ and the dashed line to H_{c2}^-. The inset shows the T dependence of the optimum $v_{F0}^+ q/T_c^{(0)}$. The phase boundary between the normal and superconducting states corresponds to the larger value of H_{c2}^+ and H_{c2}^-. The upper critical fields $H_{c2}^\pm$ cross each other at $T \approx 0.11T_c^{(0)}$, resulting in a kink in the phase boundary.

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