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# The Fulde–Ferrell–Larkin–Ovchinnikov state in layered d-wave superconductors: in-plane anisotropy and resonance effects in the angular dependence of the upper critical field

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## Abstract

We study the anisotropy of the in-plane upper critical magnetic field coupled to the orbital motion and the spins of electrons in a layered  $d_{x^2-y^2}$  organic superconductor in the spatially modulated Fulde–Ferrell–Larkin–Ovchinnikov phase. We show that the interplay between the nodal structure of the order parameter and its spatial modulation results in the very peculiar angular dependence of the onset of superconductivity in the high-field regime. The principal axis of the field-direction dependence of the onset of superconductivity is tilted by  $\pi/4$  in the temperature range  $0.056 \lesssim T < 0.56$ . In some cases the resonance between the modulation wavevector and the vector potential of a parallel magnetic field may lead to anomalous cusps in the temperature and in-plane angular dependences of the onset of superconductivity. The obtained results support the interpretation of the recent experiments as evidence of the FFLO state.

## 1. Introduction

Several recent experiments [1–6] on layered organic crystals [7, 8] such as  $\kappa$ -(BEDT-TTF)<sub>2</sub>X (BEDT-TTF stands for bis-(ethylenedithia-tetrathiafulvalene)) or (TMTSF)<sub>2</sub>X (TMTSF stands for tetramethyl-tetraselenafulvalene) salts have led to a renewed discussion of a possible realization of the Fulde–Ferrell–Larkin–Ovchinnikov (FFLO) [9, 10] phase in the superconducting state under high in-plane magnetic field [1–3]. The existence of such a superconducting phase was predicted back in the 1960s by Fulde and Ferrell [10] along with Larkin and Ovchinnikov [9]. They pointed out that in clean superconductors at  $T < T^* = 0.56T_{c0}$  a paired state described by the spatially modulated order parameter becomes more favorable when the spin effect of the magnetic

field dominates over the orbital effect of the magnetic field. In the normal state under a magnetic field the Fermi surfaces of spin-up and spin-down electrons are displaced due to the Zeeman energy splitting. When the magnetic field is above the Pauli limit,  $\mu_B H_P = \Delta_0/\sqrt{2}$ , where  $\Delta_0$  is the superconducting gap at  $T = 0$  and  $H = 0$ , the Fermi surface mismatch should lead to a breaking of pairs with zero total momentum. However, the pairing between exchange-split Fermi surfaces can be done with non-zero total momentum, which results in the spatial modulation of the order parameter. Therefore, the superconducting state can be stable beyond the field set by the Pauli paramagnetic limit.

The conditions of the FFLO state formation are rather stringent [11–13]. Theoretically, it is predicted that the modulated superconducting phase can only occur if the

following hold. (i) The orbital pair breaking effect is sufficiently weaker than the Pauli paramagnetic limit. In layered singlet-paired superconductors the Zeeman response strongly dominates the orbital response at low temperatures for an in-plane magnetic field. (ii) The system is highly clean, since the FFLO state is very sensitive to the presence of nonmagnetic impurities, in contrast to the BCS state of s-wave pairing [14–16]. The known layered superconductors are available in high-purity single crystals,  $\xi_0 = \hbar v_F / \pi \Delta_0 > l$ , where  $l$  is the mean free path [17]. Furthermore, the highly anisotropic Fermi surface [12, 18, 19] favors the FFLO phase formation. This is an inherent property of layered conductors that exhibit a highly anisotropic structure and hence have features of a system with reduced dimensionality.

Although the main feature (property) of the FFLO state, the spatial oscillations of the order parameter, has not been observed directly yet, indications for a possible experimental observation of the FFLO state have been reported for layered organic superconductors, when external magnetic field is aligned along the conducting planes [20–22, 4]. The anomaly in the thermal conductivity for the clean organic sample  $\lambda$ -(BETS)<sub>2</sub>GaCl<sub>4</sub> [22], the calorimetric and magnetic torque evidence for the appearance of an additional first-order phase transition line within the superconducting phase in the in-plane high-field regime for organic sample  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu(NCS)<sub>2</sub> [2, 3], a second phase line forming a high-field low-temperature region obtained from rf penetration measurements in  $\lambda$ -(BETS)<sub>2</sub>GaCl<sub>4</sub> and  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu(NCS)<sub>2</sub> [5, 6], a kink in the diamagnetic susceptibility for the magnetic-field-induced organic superconductor  $\lambda$ -(BETS)<sub>2</sub>FeCl<sub>4</sub> [23] and an anomalous in-plane anisotropy of the onset of superconductivity in (TMTSF)<sub>2</sub>CIO<sub>4</sub> conductor [1, 17] have been interpreted as related to a stabilization of the superconducting phase with the modulated order parameter in real space. The authors of [24] attribute the characteristic dip structure in the interlayer resistance observed in the superconducting state of a magnetic-field-induced organic superconductor  $\lambda$ -(BETS)<sub>2</sub>FeCl<sub>4</sub> to the stabilization of the FFLO state, and it is related to the spatial modulation of the order parameter [25].

It has been shown that in the parallel orientation of the magnetic field, when accounting for the orbital effect, the in-plane anisotropy of the onset of s-wave superconductivity should change dramatically in the FFLO state, thus providing an interesting possibility to sample the direction of the FFLO modulation [26]. In this paper, we revisit the problem of the anisotropy of the upper critical field in the FFLO phase by considering the effects related to the  $d_{x^2-y^2}$ -pairing symmetry in layered organic superconductors. As was demonstrated in [27], the FFLO state in d-wave superconductors (wider) extends to a higher fields than that in s-wave superconductors.

Whether the spin part of the superconducting order parameter is a singlet or triplet is a current topic of debate [28]. Indeed, the NMR experiment with (TMTSF)<sub>2</sub>PF<sub>6</sub> salt at  $T_c$  and under pressure showed the absence of the Knight shift, supporting the triplet scenario of pairing [29], while the <sup>77</sup>Se NMR Knight shift in a

recent experiment with (TMTSF)<sub>2</sub>CIO<sub>4</sub> at low fields reveals a decrease in spin susceptibility  $\chi_s$  consistent with singlet spin pairing [30]. <sup>13</sup>C NMR measurements with  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu(NCS)<sub>4</sub> evidenced a Zeeman-driven transition within the superconducting state and stabilization of the inhomogeneous phase [4]. The temperature dependence of the NMR Knight shift (which measures the electron spin susceptibility) in the superconducting state provides a means to distinguish between the triplet and singlet pairing. The nuclear magnetic resonance relaxation-rate measurements reported in [31] show that the superconductivity is gapless with lines of zeros of the gap. Thus the d-wave pairing is most favored. More recent penetration depth measurements in the superconducting state of the  $\kappa$ -(BEDT-TTF)<sub>2</sub>X support a d-wave picture [32, 33]. Moreover, the sensitivity of  $T_{c0}$  to disorder in the  $\kappa$ -(BEDT-TTF)<sub>2</sub>X salts has been remarked, supporting the non-BCS mechanism of superconductivity [34].

## 2. The general setting

We model an organic layered superconductor as a stack of conducting layers. The single-electron spectrum of the system is taken as follows:

$$\xi_{\mathbf{p}} = \frac{p_x^2}{2m_x} + \frac{p_y^2}{2m_y} + 2t \cos(p_z d) - \mu, \quad (1)$$

where the effective mass approximation is used for description of the in-plane charge carrier motion and the tight-binding approximation for the motion along the  $z$ -direction, perpendicular to the conducting layers. The Fermi surface acquires the shape of a corrugated cylinder with an elliptical cross section. We assume that the corrugation of the Fermi surface, because of the coupling between layers, is small, i.e.  $t \ll T_{c0}$ , but sufficiently large to make the mean field treatment justified; namely, the coupling between layers,  $t$ , satisfies the condition  $T_{c0}^2/E_F \ll t \ll T_{c0}$  [35]. Here  $T_{c0}$  is the critical temperature of the system at  $T = 0$ . In this work we concentrate on the effects related to the d-wave pairing symmetry, therefore in the following the electron spectrum is assumed to be isotropic. Note that an effective mass anisotropy can be taken into account by the scaling transformation of the coordinates and orbital magnetic field [36].

For the in-plane magnetic field, with amplitude  $H$ , applied at angle  $\alpha$  with the  $x$ -axis, we may choose the vector potential along the  $z$ -axis, with amplitude  $A = -xH \sin \alpha + yH \cos \alpha$ . Assuming that the vector potential varies slowly at the interlayer distances and taking into account that the system is near the second-order phase transition, we employ the linearized Eilenberger equation for a layered superconductor in the form [37]

$$\hat{L}_2 f_{\omega}(\mathbf{n}_{\mathbf{p}}, \mathbf{r}, p_z) = \Delta \hat{p}(\mathbf{r}), \quad (2)$$

where

$$\hat{L}_2 = \Omega_n + \frac{\hbar}{2} \mathbf{v}_F \nabla_{x,y} + t \sin(p_z d) \left[ e^{i\mathbf{Q}\mathbf{r}} - e^{-i\mathbf{Q}\mathbf{r}} \right], \quad (3)$$

$\mathbf{Q} = (\pi dH/\phi_0)[- \sin \alpha, \cos \alpha, 0]$  with  $\phi_0 = \pi \hbar c/e$ ,  $\mathbf{v}_F = v_F \hat{\mathbf{p}}$  is the in-plane Fermi velocity,  $f_\omega(\hat{\mathbf{p}}, \mathbf{r}, p_z)$  is the anomalous quasiclassical Green function integrated over the energy near the Fermi surface,  $\Omega_n \equiv \omega_n + i\hbar \text{sign}(\omega_n)$ ,  $\hbar \equiv \mu_B H$  is the Zeeman energy, and  $\Delta(\mathbf{r})$  is the order parameter. We assume the d-wave pairing interaction in the form

$$V_d(\mathbf{p}, \mathbf{p}') = -\lambda 2\gamma(\hat{\mathbf{p}})\gamma(\hat{\mathbf{p}}'), \quad (4)$$

where  $\gamma_{d_{x^2-y^2}}(\hat{\mathbf{p}}) = \hat{p}_x^2 - \hat{p}_y^2 = \cos(2\varphi)$  with  $\varphi$ , the azimuthal angle of  $\mathbf{p}$  (angle between the momentum in the crystalline  $xy$ -plane and  $x$ -axis, which is the anti-nodal direction ( $\phi = 0$ )). Here the factor of 2 is introduced for normalization. Then the order parameter may be written

$$\Delta_{\hat{\mathbf{p}}}(\mathbf{r}) = \Delta_d(\mathbf{r})\gamma(\hat{\mathbf{p}}), \quad (5)$$

where  $\Delta_d(\mathbf{r})$  is defined as the self-consistency relation,  $\Delta_d(\mathbf{r}) = \lambda\pi T \sum_n 2\langle \gamma(\hat{\mathbf{p}}') f_{\omega_n}(\hat{\mathbf{p}}, \mathbf{r}, p_z') \rangle$ , with the averaging procedure,  $\langle \dots \rangle \equiv \int_{-\pi/d}^{\pi/d} \frac{d p_z}{2\pi} \int_0^{2\pi} d\varphi (\dots)$ . In this work we consider a model for a pure layered superconductor, meaning that the mean free path within the layers is much larger than the corresponding coherence length,  $l \gg \xi_0$ .

The FFLO state appears as a modulated order parameter with a wavevector  $\mathbf{q}$  whose direction is determined by the crystal field effects [38, 39] and the pairing symmetry [27]. The orientation of the FFLO modulation vector is arbitrary in the pure Pauli limited s-wave superconductor in the case of a Fermi surface with an elliptical cross section, which can be mapped by a scaling transformation to the isotropic case [36]. For a d-wave superconductor the symmetry of superconducting pairing fixes the directions of the FFLO modulation vector even in the absence of the crystal field effects [27]. If the orbital effects are essential, the actual direction will be determined by the interplay between the anisotropy of pairing and the crystal field effects. The situation when the latter are dominant is discussed in [38]. For the triclinic symmetry of the organic superconductors the FFLO modulation is pinned in a certain direction. The magnitude and the direction of the FFLO modulation vector are determined by the condition of the maximum value of the critical field (d-wave)

$$H_{cP2} = \max H_{c2}(q, \phi). \quad (6)$$

To describe correctly the angular dependence of the upper critical field in the FFLO phase the simple exponential solution  $f_\omega(\mathbf{n}_p, \mathbf{r}, p_z) \sim \exp(i\mathbf{q}\mathbf{r})$  is no longer valid in quasi-2D superconductors and we have to account for the orbital effects, which add the higher harmonics in FFLO modulation,  $\mathbf{q} \pm m\mathbf{Q}$ . Therefore, the solution of equation (2) should be written as

$$f_\omega(\hat{\mathbf{p}}, \mathbf{r}, p_z) = e^{i\mathbf{q}\mathbf{r}} \sum_m e^{im\mathbf{Q}\mathbf{r}} f_m(\omega_n, \hat{\mathbf{p}}, p_z). \quad (7)$$

Because of the form for  $f_\omega(\hat{\mathbf{p}}, \mathbf{r}, p_z)$  in equation (7) one can write the order parameter  $\Delta(\mathbf{r})$  as

$$\Delta(\mathbf{r}) = e^{i\mathbf{q}\mathbf{r}} \sum_m e^{i2m\mathbf{Q}\mathbf{r}} \Delta_{2m}. \quad (8)$$

From symmetry considerations it follows that  $\Delta_{-2m} = \Delta_{2m}$ . Substituting equations (7) and (8) into (2) one gets the following system of coupled equations:

$$L_n(\mathbf{q})f_0 + \tilde{t}f_{-1} - \tilde{t}f_1 = \Delta_0, \quad (9)$$

$$L_n(\mathbf{q} \pm \mathbf{Q})f_{\pm 1} \pm \tilde{t}f_0 \mp \tilde{t}f_{\pm 2} = 0, \quad (10)$$

$$L_n(\mathbf{q} \pm 2\mathbf{Q})f_{\pm 2} \pm \tilde{t}f_{\pm 1} \mp \tilde{t}f_{\pm 3} = \Delta_{\pm 2}, \quad (11)$$

$$L_n(\mathbf{q} \pm 3\mathbf{Q})f_{\pm 3} \pm \tilde{t}f_{\pm 2} = 0, \quad (12)$$

where  $L_n(\mathbf{q}) = \Omega_n + i\mathbf{v}_F\mathbf{q}/2$  and  $\tilde{t} = t \sin(p_z d)$ . Here, we have taken into account that  $\Delta_{\pm(2m+1)} = 0$  and we have introduced the notation  $f_m \equiv f_m(\omega_n, \mathbf{n}_p, p_z)$ . This hierarchy of coupled equations is broken on the level of  $f_{\pm 3}$  in order to obtain symmetric equations for the first two harmonics of the order parameter up to the second order with respect to  $t/T_{c0}$ . From equation (9) we can obtain function  $H_{c2}(q, \phi)$  to be minimized in order to obtain the absolute value and the direction of the modulation vector. In the Pauli limit, when neglecting the orbital motion, we obtain

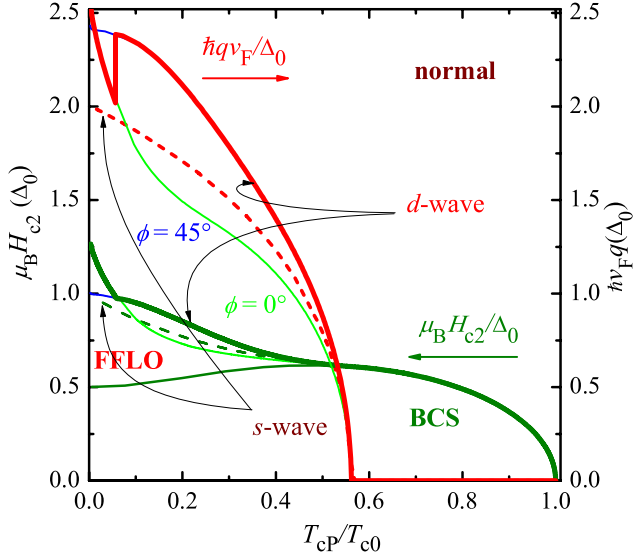
$$\ln\left(\frac{T_{c0}}{T_{cP}}\right) = \pi T_{cP} \sum_n \frac{1}{\omega_n} - \left\langle \frac{2\gamma^2(\hat{\mathbf{p}})}{L_n(\mathbf{q})} \right\rangle \equiv F(\tilde{h}_{cP}, \tilde{q}_{cP}), \quad (13)$$

where the reduced variables  $\tilde{h} = h/2\pi T$  and  $\tilde{q} = q/2\pi T$  are introduced,

$$F(\tilde{h}, \tilde{q}) \equiv \pi T \sum_n \frac{1}{\omega_n} - \frac{2}{g_1^v} - \frac{2(g_1^v - 2\Omega_n)^2}{g_1^v(g_1^v + 2\Omega_n)^2} \cos(4\phi), \quad (14)$$

$T_{cP}$  is the temperature of the onset of the superconductivity in the pure Pauli regime,  $g_1^v \equiv \sqrt{q^2 v_F^2 + 4\Omega_n^2}$ , and  $\phi$  is the angle the  $\mathbf{q}$  vector makes with the  $x$ -axis.  $H_{cP2}$  and the modulation vector maximizing  $H_{c2}$  are illustrated in figure 1. For a d-wave superconductor the paramagnetic upper critical field is never smaller than  $H_{cP2}$  for an s-wave superconductor [27], as shown in figure 1. The optimal direction of the modulation vector in a d-wave superconductor is  $\phi = \pm\pi/4, \pm 3\pi/4$  for  $T^{**} < T < T^*$  and  $\phi = 0, \pm\pi/2, \pi$  for  $0 < T < T^{**}$  with  $T^{**} \simeq 0.056T_{c0}$ . The magnitude of the  $\mathbf{q}$  vector monotonically increases from zero at the tricritical point to  $q \simeq 2.4\Delta_0$  at  $T < T^{**}$ , then drops discontinuously to  $q \simeq 2.4\Delta_0$  and finally approaches  $q \simeq 2.5\Delta_0$  for  $T \rightarrow 0$  [27].  $H_{cP2}$  and  $\mathbf{q}$  in s-wave pairing symmetry, shown as dashed lines in figure 1, are obtained from equation (13) by substitution  $\phi = \pi/8$ . If one accounts for the parity mixing between the d-wave and p-wave order parameter components, as has been done in [40–45] due to both time-reversal and spatial symmetry breaking in the FFLO state, a weak triplet pairing interaction between electrons with antiparallel spins remarkably enhances the upper critical field and increases the optimal value of the FFLO wavevector and temperature of the tricritical point.

From the system of equations (9)–(12) it is seen that, if  $L_n(\mathbf{q}) = L_n(\mathbf{q} \pm 2\mathbf{Q})$ , then the averaged equations (9) and (11) for  $\Delta_0$  and  $\Delta_{\pm 2}$  show that  $\Delta_{\pm 2}$  is of the same order as  $\Delta_0$ . To account for such degenerate or resonance situations



**Figure 1.** The upper critical field  $H_{c2}$  and the absolute value of the FFLO modulation vector  $q$  as a function of  $T_{c0}$  for s-wave and d-wave superconductors in the pure Pauli limit.

equations (11) and (12) are included in our consideration. From equations (11) and (12) one can obtain the equation for  $f_{\pm 2}$ ,

$$\left[ L_n(\mathbf{q} \pm 2\mathbf{Q}) + \frac{t^2}{L_n(\mathbf{q} \pm 3\mathbf{Q})} \right] f_{\pm 2} \pm \tilde{t} f_{\pm 1} = \Delta_{\pm 2}. \quad (15)$$

Substitution of the expression for the first harmonic of the pair amplitude,  $f_{\pm 1}$ , obtained from equation (10),

$$f_{\pm 1} = \mp \frac{\tilde{t} f_0}{L_n(\mathbf{q} \pm \mathbf{Q})} \pm \frac{\tilde{t} f_{\pm 2}}{L_n(\mathbf{q} \pm \mathbf{Q})}, \quad (16)$$

while accounting for the zero-order approximation with respect to  $(t/T_{c0})$  for  $f_0 \approx \Delta_0/L_n(\mathbf{q})$ , gives the equation for the second harmonic of the pair amplitude

$$\left[ L_n(\mathbf{q} \pm 2\mathbf{Q}) + \frac{t^2}{L_n(\mathbf{q} \pm 3\mathbf{Q})} + \frac{t^2}{L_n(\mathbf{q} \pm \mathbf{Q})} \right] f_{\pm 2} - \frac{\tilde{t}^2 \Delta_0}{L_n(\mathbf{q}) L_n(\mathbf{q} \pm \mathbf{Q})} = \Delta_{\pm 2}. \quad (17)$$

When substituting equation (16) for  $f_{\pm 1}$  and equation (17) for  $f_{\pm 2}$  into equation (9) one obtains the following equation for  $f_0$  with the precision up to the second order with respect to  $t/T_{c0}$ :

$$\left( L_n(\mathbf{q}) + \frac{\tilde{t}^2}{L_n(\mathbf{q} + \mathbf{Q})} + \frac{\tilde{t}^2}{L_n(\mathbf{q} - \mathbf{Q})} \right) f_0 - \sum_{\pm} \frac{\tilde{t}^2 \Delta_{\pm 2}}{L_n(\mathbf{q} \pm \mathbf{Q}) L_n(\mathbf{q} \pm 2\mathbf{Q})} = \Delta_0. \quad (18)$$

Making use of the self-consistency relation one obtains for  $T < T^*$  in a second-order approximation on the small parameter  $t/T_{c0}$

$$\Delta_0 \left( \frac{1}{\lambda} - \pi T_c \sum_n \left\langle \frac{2\gamma^2(\hat{\mathbf{p}})}{L_n(\mathbf{q})} \right\rangle_{\hat{\mathbf{p}}} + t^2 a \right) = \Delta_{\pm 2} t^2 c_{\pm}, \quad (19)$$

$$\Delta_2 \left( \frac{1}{\lambda} - \pi T_c \sum_n \left\langle \frac{2\gamma^2(\hat{\mathbf{p}})}{L_n(\mathbf{q} \pm 2\mathbf{Q})} \right\rangle_{\hat{\mathbf{p}}} + t^2 b_{\pm} \right) = \Delta_0 t^2 c_{\pm}, \quad (20)$$

where

$$a = \pi T \sum_{n, \xi = \pm} T_n(\mathbf{q}, \mathbf{q}, \xi \mathbf{Q})|_{T=T_{cP}}, \quad (21)$$

$$b_{\pm} = \pi T \sum_{n, \xi = \pm} T_n(\mathbf{q} \pm 2\mathbf{Q}, \mathbf{q} \pm 2\mathbf{Q}, \mathbf{q} \pm 2\mathbf{Q} + \xi \mathbf{Q})|_{T=T_{cP}}, \quad (22)$$

$$c_{\pm} = \pi T \sum_n T_n(\mathbf{q}, \mathbf{q} \pm \mathbf{Q}, \mathbf{q} \pm 2\mathbf{Q})|_{T=T_{cP}}, \quad (23)$$

with

$$T_n(\mathbf{g}_1, \mathbf{g}_2, \mathbf{g}_3) = \frac{1}{2} \int_0^{2\pi} d\varphi \frac{2 \cos^2(2\varphi)}{2\pi L_n(\mathbf{g}_1) L_n(\mathbf{g}_2) L_n(\mathbf{g}_3)}. \quad (24)$$

Taking into account the fact that the critical temperature when accounting for the orbital effects,  $T_c$ , is close to  $T_{cP}$ , equations (17) and (18) can be written as the following system of coupled equations:

$$\Delta_0 \left( -\frac{(T_{cP} - T_c)}{AT_c} + t^2 a \right) = \Delta_{\pm 2} t^2 c_{\pm}, \quad (25)$$

$$\Delta_{\pm 2} \left( -\frac{(T_{cP} - T_c)}{T_c B_{\pm}} + t^2 b_{\pm} + \delta_{\pm} \right) = \Delta_0 t^2 c_{\pm}, \quad (26)$$

where

$$A \equiv 1 - \frac{h}{T_{cP}} \frac{\partial T_{cP}}{\partial h} = \frac{1}{1 - \frac{\hbar \partial F(\hbar, \tilde{q})}{\partial \hbar} |_{T=T_{cP}}} \quad (27)$$

and

$$B_{\pm} \equiv \frac{A \frac{\hbar \partial \Lambda_{\pm}(\hbar)}{\partial \hbar} |_{T=T_{cP}}}{A - \frac{\hbar \partial \Lambda_{\pm}(\hbar)}{\partial \hbar} |_{T=T_{cP}}}, \quad (28)$$

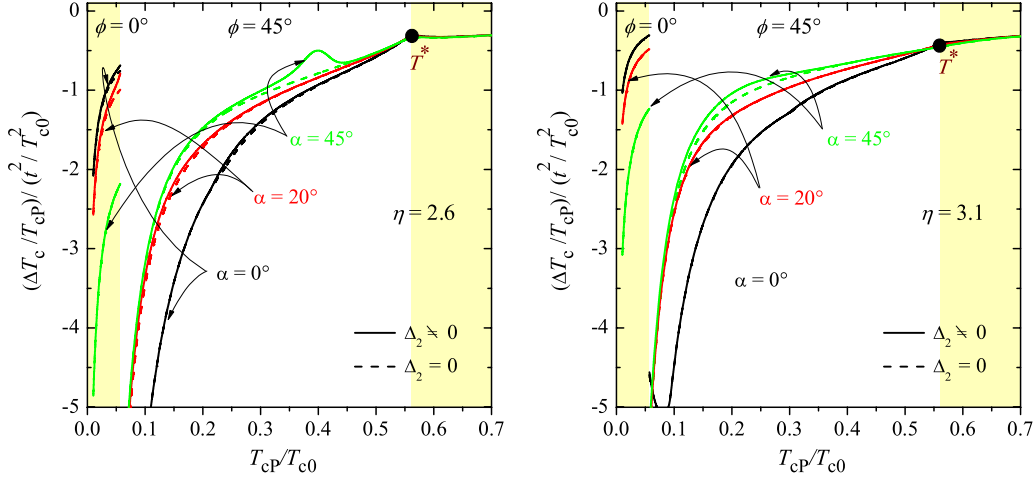
$$\Lambda_{\pm} = \pi T \sum_n \left\langle \frac{2\gamma^2(\hat{\mathbf{p}})}{L_n(\mathbf{q})} \right\rangle_{\hat{\mathbf{p}}} - \left\langle \frac{2\gamma^2(\hat{\mathbf{p}})}{L_n(\mathbf{q} \pm 2\mathbf{Q})} \right\rangle_{\hat{\mathbf{p}}}, \quad (29)$$

$$\delta_{\pm} = \Lambda_{\pm} |_{T=T_{cP}}. \quad (30)$$

It is possible to calculate  $T_n(\mathbf{g}_1, \mathbf{g}_2, \mathbf{g}_3)$  analytically, namely

$$\begin{aligned} T(\omega_n, \mathbf{g}_1, \mathbf{g}_2, \mathbf{g}_3) &= -8t^2 \sum_{k=1}^{N_k} \frac{4a}{p_k^2 p_{k+1} p_{k+2}} \\ &+ \frac{ig_k^2}{(g_k^- - g_k^+) (g_k^- - g_{k+1}^-) (g_k^- - g_{k+1}^+)} \\ &\times \frac{[1 + (g_k^-)^4]}{(g_k^- - g_{k+2}^-) (g_k^- - g_{k+2}^+) (2a - g_k^v)}, \end{aligned} \quad (31)$$

where we have introduced the following notations:  $g_k^v \equiv \sqrt{g_k^2 v_F^2 + 4\Omega_n^2}$ ,  $g_k = g_{k,x} - ig_{k,y}$ ,  $\mathbf{g}_1 \equiv \mathbf{q}$ ,  $\mathbf{g}_2 \equiv \mathbf{q} \pm \mathbf{Q}$ ,  $\mathbf{g}_3 \equiv$



**Figure 2.** Contribution of the orbital effect as a function of  $T_{cP}/T_{c0}$  for several field directions. The left panel is for  $\eta = 2.6$  and the right panel for  $\eta = 3.1$ . The regions showing the results obtained when  $\mathbf{q}$  is along the  $x$ -axis and for the conventional phase are highlighted in yellow, while the white region is for the case when the  $\mathbf{q}$  vector makes angle  $\phi = \pi/4$  with the  $x$ -axis.

$\mathbf{q} \pm 2\mathbf{Q}$  and

$$g_k^\pm \equiv i \frac{(2\Omega_n \pm g_k^v)}{g_k v_F}. \quad (32)$$

Here  $k$  is the cycling index with  $k = 1, 2, 3$ . The solution of the system of equations (25) and (26) is given as

$$\begin{aligned} \frac{T_{cP} - T_c}{T_c} &\equiv \frac{(aA + b_\pm B_\pm) t^2 + B_\pm \delta_\pm}{2} \\ &+ \frac{1}{2} [(aA + b_\pm B_\pm) t^2 + B_\pm \delta_\pm]^2 \\ &- 4AB_\pm [(ab_\pm - c_\pm^2) t^4 + a t^2 \delta_\pm]^{\frac{1}{2}}, \end{aligned} \quad (33)$$

where for  $\pm$  the values are chosen that maximize the critical temperature. Usually, the second harmonic of the order parameter,  $\Delta_{\pm 2}$ , can be neglected because the developed theory is valid up to the second order with respect to  $t/T_{c0}$ , and because  $t \ll T_{c0} \lesssim v_F Q$ . In this case the solution of the system is just  $(T_{cP} - T_c)/AT_c = -t^2 a$ . However, if  $\delta_\pm = 0$  (the resonance condition) then the term  $(- (T_{cP} - T_c)/T_c B_\pm + t^2 b_\pm + \delta_\pm)$  in the lhs of equation (26) is of the same order as the corresponding term in equation (25) (up to the second order with respect to  $t/T_{c0}$ ). Consequently,  $\Delta_{\pm 2}$  becomes of the order of  $\Delta_0$ , and one has to consider both equations in the system on equal footing [46–48].

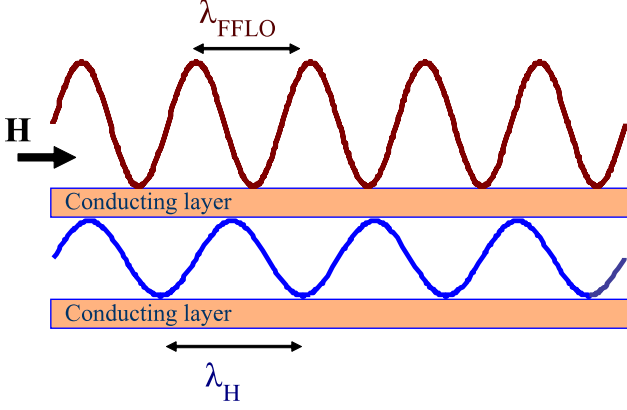
### 3. Results and discussion

To obtain the onset temperature of superconductivity, the summation over the Matsubara frequencies in equations (21)–(23), (30) was performed numerically. We used  $N = 10^3$  terms in the summation and this number suffices for the convergent result. The results presented here are in dimensionless units. In our numerical investigations we restrict ourselves to the following choice of parameters: the interlayer coupling is  $t = 1.8$  K [50],  $t/T_{c0} = 0.2$ , the Fermi velocity  $v_F = 7.5\text{--}10.0 \times 10^4$  m s $^{-1}$  [49], and the interlayer distance is  $d = 1.62$  nm. The Fermi velocity is characterized by the

dimensionless parameter  $\eta = \hbar v_F \pi d / \phi_0 \mu_B$ . In the following we consider two cases. In the first the symmetry of superconducting pairing fixes the direction of the FFLO modulation wavevector, characterized by  $\phi$ , the angle that  $\mathbf{q}$  makes with the  $x$ -axis: at high temperature of the FFLO phase  $\phi = \pm\pi/4, \pm 3\pi/4$ , while in the low-temperature phase  $\phi = 0, \pm\pi/2, \pi$ , thus making it fourfold degenerate. In the second case the crystal field effect leaves this degeneracy.

#### 3.1. FFLO wavevector is fixed by the pairing symmetry

Figure 2 presents the orbital-motion-induced normalized correction of  $\Delta T_c = T_c - T_{cP}$  as a function of  $T_{cP}/T_{c0}$  for different directions of the applied field. In the following, the direction of the external field is measured by  $\alpha$ , the angle the applied field  $\mathbf{H}$  makes with the  $x$ -axis. Due to the fourfold symmetry of the  $d_{x^2-y^2}$  pairing we plot  $\Delta T_c(T_{cP}/T_{c0})$  curves only for  $\alpha = 0, \pi/9, \pi/4$ . The left panel shows the results for  $\eta = 2.6$ , while the right panel is for  $\eta = 3.1$ . The dashed lines illustrate the solution  $P = -t^2 a$ , which is justified when the second harmonics of the order parameter are negligible,  $\Delta_{\pm 2} = 0$ . The solid lines exhibit the results obtained with equation (33), when accounting for the possibility of the resonance effect. The regions  $T_{cP} < T^{**}$  and  $T_{cP} > T^*$  are shadowed (online: highlighted in yellow). In the low-temperature region the modulation vector  $\mathbf{q}$  is along the  $x$ -axis, while at  $T_{cP} > T^*$   $|\mathbf{q}| = 0$  and it describes  $\Delta T_c$  in the conventional phase. The central white domain illustrates  $\Delta T_c$ , when the modulation wavevector  $\mathbf{q}$  makes the angle  $\phi = \pi/4$  with  $x$ -axis. The re-orientational transition at  $T_{cP} < T^{**}$  is accompanied by a strong change of the orbital correction, which should result in the sharp step on the  $T_c(H)$  curve. At some values of  $T_{cP}/T_{c0}$  for  $\eta = 2.6$  we see an essential discrepancy between the solid and dashed lines. This discrepancy is induced by the resonance effect discussed above. The physics of the resonance effect is as follows. As seen from equation (3), the vector potential of the



**Figure 3.** Schematic picture of the FFLO modulated order parameter and the oscillations of the interlayer coupling, explaining the physical origin of the resonances.

parallel magnetic field results in a modulation of the interlayer coupling,  $t \sin(\mathbf{Q} \cdot \mathbf{r})$ . The period of this modulation,  $\lambda_H = 2\phi_0/dH$ , may interfere with the period of the in-plane FFLO modulation vector  $\lambda_{\text{FFLO}}$  (see figure 3; for  $T = 0$   $\lambda_{\text{FFLO}}^0 = \pi \hbar v_F / \Delta_0 = \pi^2 \xi_0$ ), leading to the anomalies in  $\Delta T_c$ , when  $\lambda_{\text{FFLO}} = \lambda_H$ . Figure 2 (right panel) does not exhibit strong anomalies. We see only a small discrepancy between the solid and dashed lines at  $T_{\text{cP}}/T_{\text{c0}} \simeq 0.2$  for  $\mathbf{q} \perp \mathbf{H}$ , meaning that for the Fermi velocity parameter  $\eta = 3.1$  in the shown ranges of  $T_{\text{cP}}/T_{\text{c0}}$  and  $\phi = \pi/4$  resonance conditions are not precisely met.

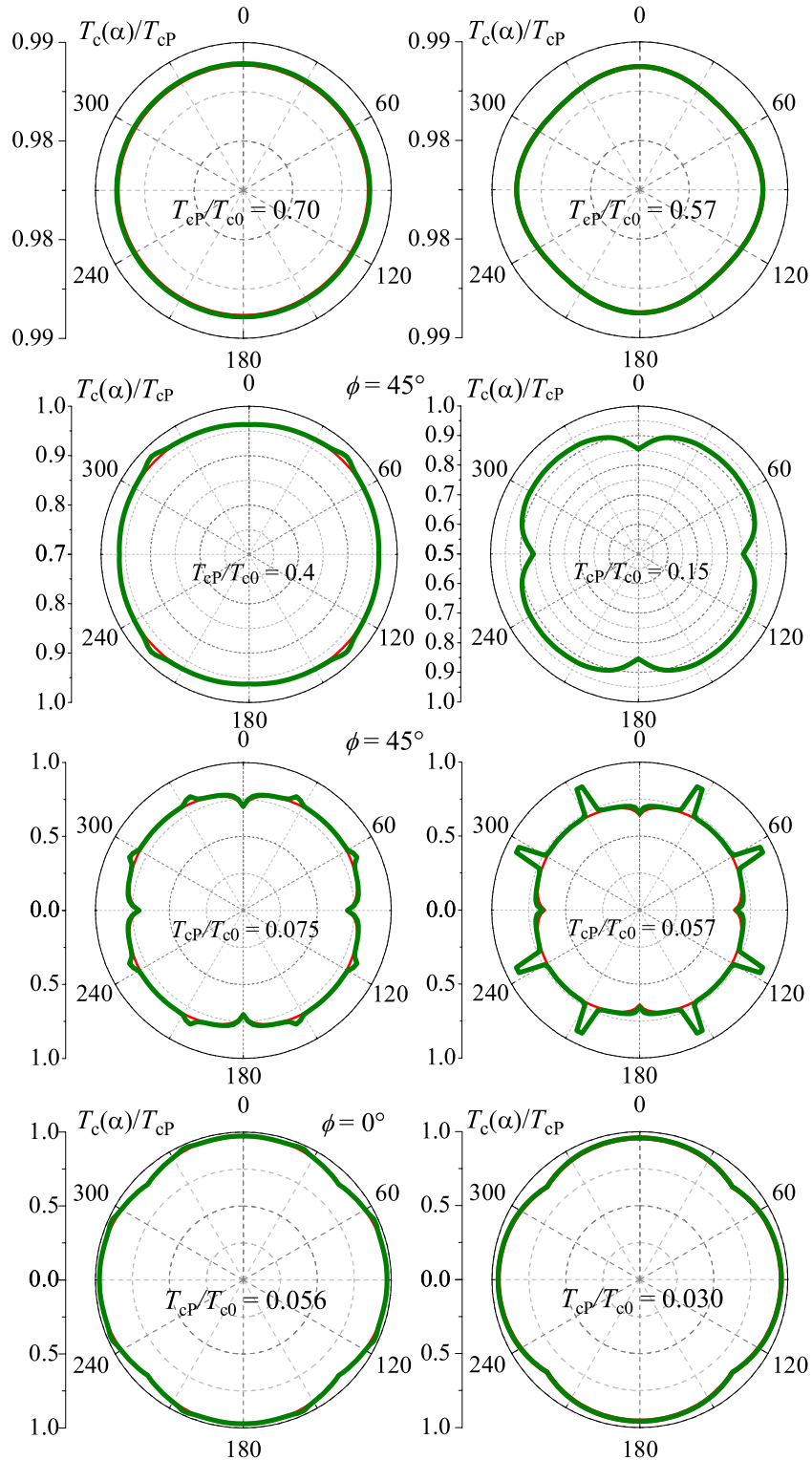
The existence of the nodes in the order parameter results in particular features of the anisotropy of the superconducting onset temperature induced by the spatially modulated FFLO phase. Figure 4 shows the magnetic field angular dependence of the normalized superconducting transition temperature,  $T_c(\alpha)/T_{\text{cP}}$ , calculated at  $T_{\text{cP}}/T_{\text{c0}} \simeq 0.03, 0.056$ , when the modulation vector is fixed by the symmetry of pairing to the direction of the maxima of the order parameter (fourth panel), at  $T_{\text{cP}}/T_{\text{c0}} \simeq 0.057, 0.075, 0.15$  and  $0.4$ , when it is fixed to the nodes of  $\Delta_{\mathbf{p}}(\mathbf{r})$  (second and third panels), and at  $T_{\text{cP}}/T_{\text{c0}} \simeq 0.57$  and  $0.7$ , corresponding to the conventional phase (first panel). The thick lines are the results accounting for the contribution of the second harmonics of the order parameter,  $\Delta_{\pm 2} \neq 0$ , while the thin ones are obtained when approximating it by  $\Delta_{\pm 2} = 0$ . In the polar plot the direction of each point seen from the origin corresponds to the magnetic field direction and the distance from the origin corresponds to the normalized critical temperature. While reducing  $T_{\text{cP}}/T_{\text{c0}}$  the figure gives evolution of the anisotropy of the upper critical field with the applied field strength. One can see that the field-angle dependence of the onset of superconductivity evidences two transitions. One is at  $T_{\text{cP}}/T_{\text{c0}} = 0.56$ , the transition from the conventional phase to the FFLO modulated phase. At this point the principal axes of the anisotropy are tilted by the angle  $\pi/4$ . The second transition occurs at  $T_{\text{cP}}/T_{\text{c0}} \simeq 0.056$ . At this temperature the direction of the FFLO modulation vector rotates by  $\phi = \pi/4$ , which is reflected in a change of the overall anisotropy of the onset of

superconductivity. The transition from the high-temperature FFLO phase to the low-temperature phase is accompanied by a strong decrease of the orbital effect, that makes  $T_c(\alpha)/T_{\text{cP}}$  dependence at  $T < T^{**}$  much closer to the pure paramagnetic limit. We see that, in addition to the overall anisotropy induced by the FFLO modulation (thin lines), cusps develop for certain directions of the applied field, when the resonance conditions are realized. These resonant cusps are obtained when the orbital effects of the field are taken in the second-order approximation. At the  $T_{\text{cP}} = T^{**}$  transition the resonant cusps almost vanish, making observation of such behavior in the experiment additional evidence for the FFLO phase formation.

### 3.2. Influence of the crystal field effects

So far we have neglected the influence of the crystal field on the pinning of the direction of the FFLO modulation wavevector. However, in real systems the crystal field effect is inevitably present and influences the direction of the FFLO modulation. In the case of weak crystal field effect it can lift the fourfold degeneracy of the direction of FFLO modulation, making it twofold degenerate. To illustrate this situation we suppose that the FFLO modulation vector is along the  $\phi = \pi/4, 5\pi/4$  directions and investigate the anisotropy of the superconducting onset in the temperature range  $T^{**} < T$ . Figure 5 displays the orbital-motion-induced normalized correction of the transition temperature,  $\Delta T_c$ , as a function of normalized temperature,  $T_{\text{cP}}/T_{\text{c0}}$ , for several orientations of the applied field, when the crystal field effect is sufficient to break the fourfold degeneracy to fix the direction of the FFLO modulation vector. The Fermi velocity parameter is  $\eta = 2.6$ . The dashed lines illustrate the result for  $\Delta_{\pm 2} = 0$ , while the solid lines are the solutions with  $\Delta_{\pm 2} \neq 0$ . Because the overall symmetry of the superconducting onset is twofold, we provide the results for large range of angles  $\alpha$  as compared to the case of section 3.1.

Figure 6 shows the magnetic field angular dependence of the normalized superconducting transition temperature,  $T_c(\alpha)/T_{\text{cP}}$ , calculated at  $T_{\text{cP}}/T_{\text{c0}} \simeq 0.057, 0.075, 0.15$  and  $0.4$ . We see that the shape of the field-angle dependence of the onset of superconductivity in the high-temperature FFLO phase is similar to that obtained for an s-wave superconductor [26]. However, the principal axis of the plot is not vertical but tilted by  $\pi/4$  and fixed along the direction of the modulation vector. For the considered parameters the direction of the maximum of the upper critical field is perpendicular to the FFLO modulation vector if we consider the non-resonant situation. Even in the presence of the ‘easy axis’ along the direction  $\pi/4$  induced by the crystal field, the fourfold degeneracy of the direction of the FFLO modulation vector is restored again for  $T < T^{**}$ . Therefore, the anisotropy in the low-temperature FFLO phase is presumably the same as that discussed in section 3.1. Hence the orbital induced reduction of the upper critical field is stronger for  $\phi = \pi/4, -3\pi/4$  than for  $\phi = 0, \pm\pi/2, \pi$ . Comparing the resonance induced changes in  $T_c(\alpha)/T_{\text{cP}}$  obtained for the d-wave superconductor with those induced in

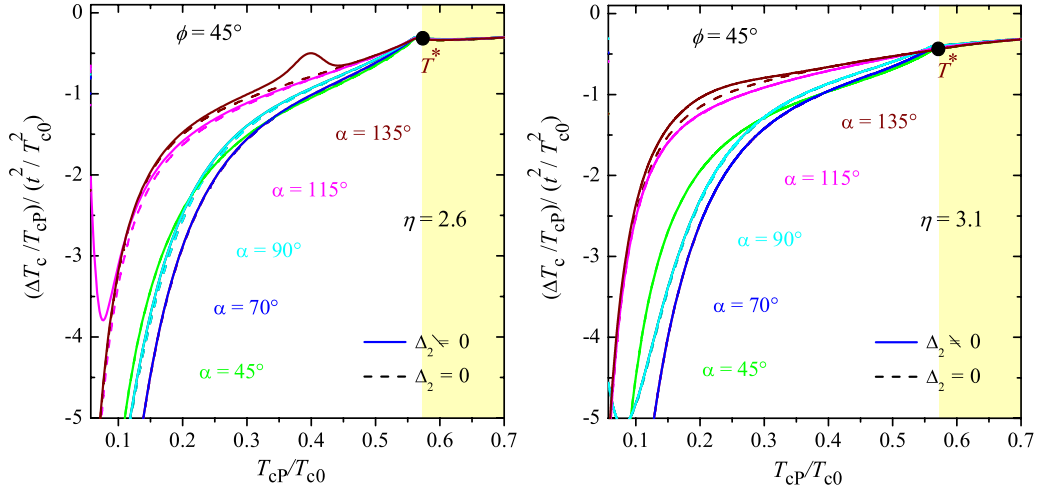


**Figure 4.** Normalized superconducting transition temperature,  $T_c(\alpha)/T_{cP}$  as a function of  $\alpha$  for several values of  $T_{cP}/T_{c0}$  and  $\eta = 2.6$ . The angle between the  $\mathbf{q}$ -vector and  $x$ -axis is  $\phi = \pi/4$  (second and third panels) and  $\phi = 0$  (last panel). The direction of the  $\mathbf{q}$ -vector is fixed by the symmetry of the order parameter.

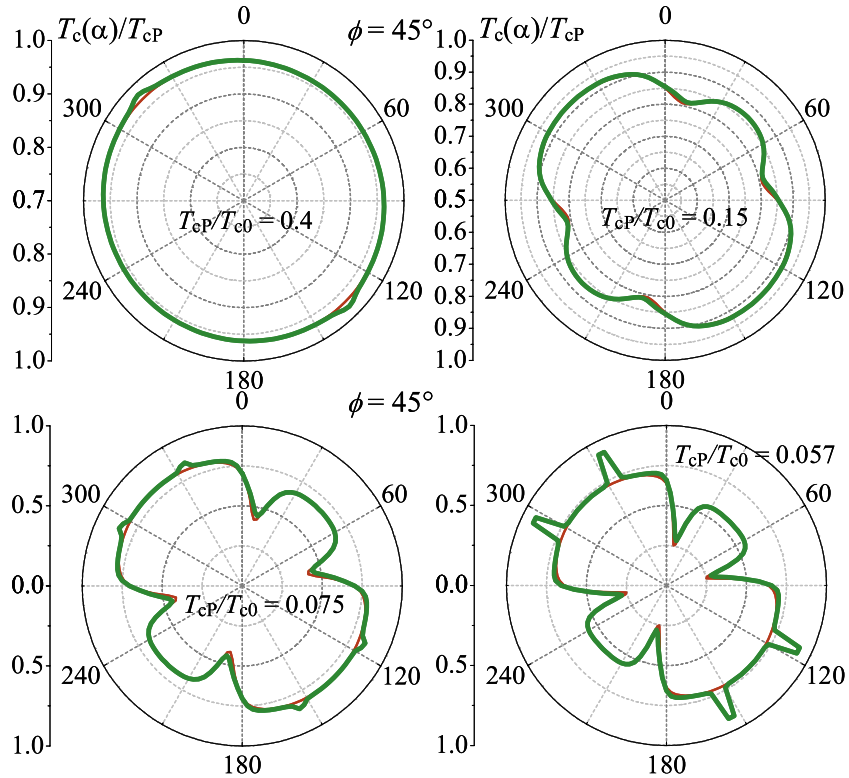
the s-wave superconductor [46], we see that the resonance is a more rare event in the former case than in the latter case. Incorporating into the model the terms beyond the second order produces additional, although much smaller, peaks in the angular dependence of the upper critical field.

The presence of additional p-wave pairing interactions can make the pinning of the direction of the FFLO modulation wavevector more complicated. This can occur only at high fields where the singlet pairing component of the gap function is strongly suppressed.





**Figure 5.** The same as in figure 2; however, the direction of the  $\mathbf{q}$ -vector is influenced by the crystal field.



**Figure 6.** The same as in figure 4; however, the direction of the  $\mathbf{q}$ -vector is influenced by the crystal field.

#### 4. Conclusion

In conclusion, we have described the anisotropy of the onset of superconductivity in layered d-wave superconductors under applied in-plane magnetic field in the Fulde–Ferrell–Larkin–Ovchinnikov phase. We demonstrated that the principal axis of the field-direction dependence of the onset of superconductivity is tilted by  $\pi/4$  and directed along the modulation vector in the range of temperatures  $T^{**} < T < T^*$ . The low-temperature FFLO phase is characterized by the principal axis pointing along the  $x$ -axis and by an essential reduction of the magnetic-field-induced orbital

effect as compared to the high-temperature FFLO phase. This makes the  $T_c(\alpha)/T_{cP}$  dependence much closer to the pure paramagnetic limit. A weak crystal field influences the direction of the FFLO wavevector modulation, making the direction of the maximum of the upper critical field perpendicular to the FFLO modulation vector. The resonance between the modulation vector of the FFLO phase and the vector potential of the magnetic field may lead to anomalous cusps in the field-direction dependence of the upper critical field analogous to those calculated previously in an s-wave conductor. We suggest that observation of characteristic cusps in the anisotropy of the onset of superconductivity as well as

the shift of the principal axis of the anisotropy of the upper critical field may serve as direct evidence for the appearance of the FFLO phase in layered superconductors.

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