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## In-Plane Magnetic Field Anisotropy of the Fulde-Ferrell-Larkin-Ovchinnikov State in Layered Superconductors

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There is strong experimental evidence of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state formation in layered organic superconductors in a parallel magnetic field. We study theoretically the interplay between the orbital effect and the FFLO modulation in this case and demonstrate that the in-plane critical field anisotropy drastically changes at the transition to the FFLO state. The very peculiar angular dependence of the superconducting onset temperature which is predicted may serve for unambiguous identification of the FFLO modulation. The obtained results permit us to suggest the modulated phase stabilization as the origin of the magnetic-field angle dependence of the onset of superconductivity experimentally observed in  $(\text{TMTSF})_2\text{ClO}_4$  organic conductors.

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Layered superconductors exposed to an external magnetic field aligned parallel to their conducting layers have been the focus of theoretical and experimental investigations due to their remarkable anisotropic properties [1,2] favorable to the formation of the spatially modulated phase, known as the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [3,4]. In particular, in the family of organic layered superconductors  $(\text{TMTSF})_2X$ , where anion  $X$  is  $\text{PF}_6$ ,  $\text{ClO}_4$ , etc., very large upper critical fields, which exceed the Pauli paramagnetic limit, were reported [5–10]. In layered conductors the orbital motion of electrons is mostly restricted to the conducting crystal planes when hopping between adjacent layers is small. Thus the magnetic field applied parallel to the conducting planes causes only small diamagnetic currents and the orbital depairing is strongly weakened. Therefore, spin-singlet superconductivity is mainly limited by the Zeeman energy (Pauli spin polarization) of the quasiparticles. In contrast, the Pauli effect is negligible for a spin-triplet pairing because in this case Cooper pairs gain Zeeman energy without losing the condensation energy. The question concerning the singlet or triplet symmetry of the superconducting order parameter in layered organic conductors is a current topic of debate. Indeed, the nuclear magnetic relaxation (NMR) experiments with  $(\text{TMTSF})_2\text{PF}_6$  salts below  $T_c$  and under pressure showed the absence of the Knight shift, thus supporting the triplet scenario of pairing [11], while the  $^{77}\text{Se}$  NMR Knight shift in a recent experiment with  $(\text{TMTSF})_2\text{ClO}_4$  revealed a decrease in spin susceptibility consistent with singlet pairing [12].  $^{13}\text{C}$  NMR measurements with  $\kappa$ -(BEDT-TTF) $_2\text{Cu}(\text{NCS})_4$  also evidenced for a Zeeman-driven transition within the superconducting state and stabilization of FFLO phase [13]. For the singlet superconductivity, the FFLO phase can be a candidate for the enhancement of the upper critical

field [3,4]. Note that in the compound  $(\text{TMTSF})_2\text{ClO}_4$  the substantial anisotropy within the conducting  $\mathbf{a}$ - $\mathbf{b}^*$  plane is present. When a magnetic field is aligned along the high conductivity  $\mathbf{a}$  axis, the orbital currents are strongly quenched, which favors the FFLO phase appearance [14,15]. Interestingly, for a magnetic field applied along the  $\mathbf{b}^*$  axis, the 3D  $\rightarrow$  2D crossover occurs in the high-field regime and the coexistence of the hidden reentrant and FFLO phases can emerge [16].

Recently the in-plane angular dependence of the upper critical field,  $H_{c2}$ , of the organic superconductor  $(\text{TMTSF})_2\text{ClO}_4$  has been measured for wide temperature intervals [9]. The observed upturn of the  $H_{c2}$  curve at low temperatures has often been discussed in connection with the possibility of the FFLO state formation [17,18]. In addition, as shown in Ref. [10], the superconducting phase in a high magnetic field is more strongly suppressed by impurities than that in a low field, as expected in the FFLO scenario [19]. Furthermore, an unusual in-plane anisotropy of  $H_{c2}$  in the high-field regime was observed, which was again interpreted as evidence of FFLO state stabilization. This argument is based on the prediction of a very peculiar in-plane angular dependence of the FFLO critical field due to the orbital effects in thin superconducting films [20]. Motivated by these experimental findings we investigate in this work the influence of the spatially modulated superconducting phase on the in-plane anisotropy of the upper critical field in layered superconductors with  $s$ -wave pairing.

To describe the layered superconductors we consider a system of layers in the  $xy$  plane, stacked along the  $z$  axis. The single-electron spectrum is approximated by

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

where  $\mathbf{p} = (p_x, p_y, p_z)$  is the electron momentum. The in-plane motion is described within the effective mass approximation while the tight-binding approximation is used to describe the motion along the  $z$  direction. The corrugation of the Fermi surface due to the coupling between adjacent layers (interlayer distance  $d$ ) is assumed to be small, i.e.,  $t \ll T_{c0}$ , but sufficiently large to make the mean-field treatment valid,  $|\ln(T_{c0}/t)|T_{c0}/E_F \ll 1$  [21]. Here  $T_{c0}$  is the critical temperature of the system at zero magnetic field and  $E_F$  is the Fermi energy. We choose a gauge for which the vector potential  $\mathbf{A} = \mathbf{H} \times \mathbf{r}$  [ $\mathbf{r} = (x, y, 0)$  is a coordinate in the  $xy$  plane], i.e.,  $A_z = -xH \sin\alpha + yH \cos\alpha$ , where  $\alpha$  is the angle between the applied field, with amplitude  $H$ , and the  $x$  axis. As was demonstrated in Ref. [22], the anisotropic model with effective masses can be reduced to the isotropic one by a scaling transformation and corresponding renormalization of the magnetic field. Therefore, in the pure Pauli regime, the orientation of the FFLO modulation vector,  $\mathbf{q}$ , is arbitrary in the case of an elliptical Fermi surface. However, any deviation of the Fermi surface from the ellipticity fixes the direction of the modulation [20,23,24]. Hereinafter we assume for the sake of simplicity that these deviations from ellipticity are small and their role is just to pin the direction of the vector  $\mathbf{q}$ , which is supposed to be along the  $x$  axis. Performing this scaling transformation, we will thus consider from now on an isotropic in-plane spectrum, with mass  $m = m_x$ , and a magnetic field  $\mathbf{H} = H[(m_x/m_y)^{1/2} \times \cos\alpha, \sin\alpha, 0]$ . Taking into account that the system is near the second-order phase transition, the linearized Eilenberger equation on the anomalous Green function  $f_\omega(\mathbf{n}, \mathbf{r}, p_z)$  describing layered superconducting systems acquires the form (for positive Matsubara frequency  $\omega$  at temperature  $T$ ) [25]

$$\left[ \omega + ih + \frac{1}{2} \mathbf{v}_F \cdot \nabla + 2it \sin(p_z d) \sin(\mathbf{Q} \cdot \mathbf{r}) \right] f_\omega(\mathbf{n}, \mathbf{r}, p_z) = \Delta(\mathbf{r}). \quad (2)$$

Here  $h = \mu_B H$  is the Zeeman energy,  $\mathbf{v}_F = v_F \mathbf{n}$  is the in-plane Fermi velocity, and  $\mathbf{Q} = (\pi d H / \phi_0) \times [-\sin\alpha, (m_x/m_y)^{1/2} \cos\alpha, 0]$  with  $\phi_0 = \pi c/e$ . The order parameter is defined self-consistently as

$$\frac{1}{\lambda} \Delta(\mathbf{r}) = 2\pi T \operatorname{Re} \sum_{\omega>0} \langle f_\omega(\mathbf{n}, \mathbf{r}, p_z) \rangle, \quad (3)$$

where  $\lambda$  is the BCS pairing constant and the brackets denote averaging over  $p_z$  and  $\mathbf{n}$ . Here we considered a layered superconductor in the clean limit, meaning that the in-plane mean free path is much larger than the corresponding coherence length,  $\xi_0 = v_F / (2\pi T_{c0})$ . The upper critical field corresponds to the values of  $H$  for which the system of Eqs. (2) and (3) can be solved.

The solution of the Eilenberger equation (2) can be chosen without loss of generality as a Bloch function

$$f_\omega(\mathbf{n}, \mathbf{r}, p_z) = e^{i\mathbf{q}\mathbf{r}} \sum_m e^{im\mathbf{Q}\cdot\mathbf{r}} f_m(\omega, \mathbf{n}, p_z). \quad (4)$$

Equation (4) takes into account the possibility for the formation of the pairing state  $(\mathbf{k} + \frac{\mathbf{q}}{2}, \uparrow; -\mathbf{k} + \frac{\mathbf{q}}{2}, \downarrow)$  with finite center-of-mass momentum. At the same time, the order parameter can be expanded as

$$\Delta(\mathbf{r}) = e^{i\mathbf{q}\mathbf{r}} \sum_m e^{i2m\mathbf{Q}\cdot\mathbf{r}} \Delta_{2m}. \quad (5)$$

It is known [26] that in the absence of orbital effect, the FFLO state only appears at  $T < T^* \simeq 0.56T_{c0}$  and  $H > H^* \simeq 1.06T_{c0}/\mu_B$ , where  $(T^*, H^*)$  is the tricritical point. Therefore, the order of the magnitude of the magnetic field required to observe the FFLO state can be found from the relation  $\mu_B H \sim T_{c0}$ . Taking this into account one obtains  $v_F Q \sim v_F e d T_{c0} / \mu_B c \sim (d/a) T_{c0}$ , where  $a$  is the unit cell in the  $xy$  plane. Therefore  $v_F Q \gtrsim T_{c0}$ . Because of the assumption  $t \ll T_{c0} \lesssim v_F Q$  one has  $\sqrt{tT_{c0}} \ll v_F Q$ . This condition allows us to retain only the terms up to the first harmonics in Eqs. (4) and (5) [since we will retain only the terms up to  $(t/T_{c0})^2$  in the final expressions]. Substituting Eqs. (4) and (5) into Eq. (2) one gets

$$\begin{aligned} L(\mathbf{q})f_0 + \tilde{t}f_{-1} - \tilde{t}f_1 &= \Delta_0, \\ L(\mathbf{q} \pm \mathbf{Q})f_{\pm 1} \pm \tilde{t}f_0 &= 0, \end{aligned} \quad (6)$$

where  $L(\mathbf{q}) = \omega + ih + i\mathbf{v}_F \cdot \mathbf{q}/2$  and  $\tilde{t} = t \sin(p_z d)$ . If one neglects the Zeeman term these equations readily describe the reentrant phase predicted by Lebed [27], with critical temperature  $T_{c0}$  at fields  $H \gg \phi_0/(d\xi_0)$ . While keeping the terms up to the second harmonics within the same procedure would yield the Lawrence-Doniach equation [27]. Inserting the solution of Eqs. (6) into the self-consistency Eq. (3), keeping only the terms up to the second order in  $t/T_{c0}$ , and subtracting it with a similar equation relating  $\lambda$  with  $T_{c0}$ , we obtain

$$\begin{aligned} \ln \frac{T_{c0}}{T} &= 2\pi T \operatorname{Re} \sum_{\omega>0} \left\{ \frac{1}{\omega} - \left\langle \frac{1}{L(\mathbf{q})} \right\rangle \right. \\ &\quad \left. + \left\langle \frac{\tilde{t}^2}{L^2(\mathbf{q})} \left( \frac{1}{L(\mathbf{q} + \mathbf{Q})} + \frac{1}{L(\mathbf{q} - \mathbf{Q})} \right) \right\rangle \right\}. \end{aligned} \quad (7)$$

This equation defines the temperature dependence of the upper critical magnetic field  $H_{c2}$  in layered superconductors, when both the paramagnetic and orbital effects are accounted for.

In the limit  $t \ll T_c$  the magnitude of the FFLO modulation vector can be calculated by neglecting the orbital part in Eq. (7). When averaging over the Fermi surface, one gets the equation

$$\begin{aligned} \ln \frac{T_{c0}}{T} &= F(\tilde{h}, \tilde{q}) \equiv \sum_{n=0}^{\infty} ([n + 1/2]^{-1} - [n + 1/2 + i\tilde{h}]^2 \\ &\quad + v_F^2 \tilde{q}^2 / 4]^{-1/2}), \end{aligned} \quad (8)$$

with reduced variables  $\tilde{h} = h/2\pi T$  and  $\tilde{q} = q/2\pi T$ , which gives rise to a FFLO vector  $\mathbf{q}$  with a magnitude that maximizes the upper critical field, thus defining  $T_{cP}(H)$  and  $q_P(H)$ , in the pure Pauli limit [28]. Finally, averaging Eq. (7) over the Fermi surface one obtains the equation for the onset of superconductivity in layered superconductors,  $T_c(H)$ , in the presence of both Zeeman and orbital effects:

$$\frac{T_{cP} - T_c}{T_c} = \frac{1}{1 - \tilde{h} \partial F(\tilde{h}, \tilde{q}) / \partial \tilde{h}} \times 2\pi T \text{Re} \sum_{\omega > 0, \pm} \left\langle \frac{\tilde{r}^2}{L^2(\mathbf{q})L(\mathbf{q} \pm \mathbf{Q})} \right\rangle \Big|_{T=T_{cP}}. \quad (9)$$

The summation over the Matsubara frequencies is performed numerically. We used  $N = 10^4$  terms in the summation and this number suffices for convergence at  $T/T_{c0} > 10^{-2}$ . Figure 1 shows the variation of the normalized correction of the transition temperature,  $\Delta T_c = T_c - T_{cP}$ , as a function of reduced strength of the magnetic field,  $H/H_{P0}$  and angle  $\alpha$  (between  $\mathbf{H}$  and the  $x$  axis). Here  $H_{P0} = \Delta_0/\mu_B$  is the critical magnetic field at  $T = 0$  in Pauli limited two-dimensional superconductors [28]. [The  $(H, T)$ -phase diagram in this regime is given in the inset of the left panel.] The left panel describes the isotropic situation, typical for layered quasi-2D compounds [29], while the right panel exhibits results obtained for the highly anisotropic in-plane Fermi surface of layered conductors, exhibiting quasi-1D character [30]. We consider two opposite mass anisotropies. When  $m_x = 10m_y$   $\mathbf{q}$  is along the heavy mass direction, while in the case of  $m_x = 0.1m_y$  it is along the light mass direction. As it was intuitively expected, the orbital effects reduce the superconducting

onset temperature,  $\Delta T_c < 0$ . While increasing the applied magnetic field,  $\Delta T_c$  first decreases in most cases until the tricritical point,  $H^*$ , is reached and the curve of  $\Delta T_c$  exhibits a kink. At  $H > H^*$  the function  $\Delta T_c(H)$  strongly depends on the in-plane effective mass anisotropy and angle  $\alpha$ . For  $\alpha$  close to  $90^\circ$ ,  $\Delta T_c$  exhibits an upturn and  $T_c$  approaches the paramagnetic limit,  $T_{cP}$ , when  $H$  increases. In contrast, for small  $\alpha$  an increase of the magnetic field leads to a decrease of  $\Delta T_c$ . For intermediate angles,  $\Delta T_c$  can be a nonmonotonic function of the field strength. In the isotropic case and for  $H/H_{P0} \geq 0.75$  the largest correction to the onset temperature  $|\Delta T_c(\alpha)|$  occurs at  $\alpha \approx 20^\circ$ . For  $m_x/m_y = 10$  and  $H/H_{P0} \geq 0.8$ ,  $|\Delta T_c^{\max}|$  is at angles close to  $\alpha \approx 45^\circ$ , while for  $m_x/m_y = 0.1$  and  $H/H_{P0} \geq H^*$ ,  $|\Delta T_c^{\max}|$  is at angles close to  $\alpha \approx 0^\circ$ . One can infer that the strong field-direction dependence of the superconducting onset temperature  $T_c(\alpha)$ , appears at high magnetic fields when the FFLO state develops, while it is absent at low fields.

The change in the anisotropy of the superconducting onset temperature that is induced by the FFLO phase is particularly visible in Figs. 2 and 3, where the magnetic-field angular dependence of the normalized superconducting transition temperature,  $T_c(\alpha)/T_{cP}$ , at constant modulus of the in-plane magnetic field and  $t/T_{c0} = 0.2$ , is plotted. 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, when the orbital destructive effect is taken into account. We show here such dependence because this type of representation is essentially informative and was realized in the experiment [9]. For magnetic fields below  $H^*$  and  $m_x = m_y$ , one can see an expected isotropic

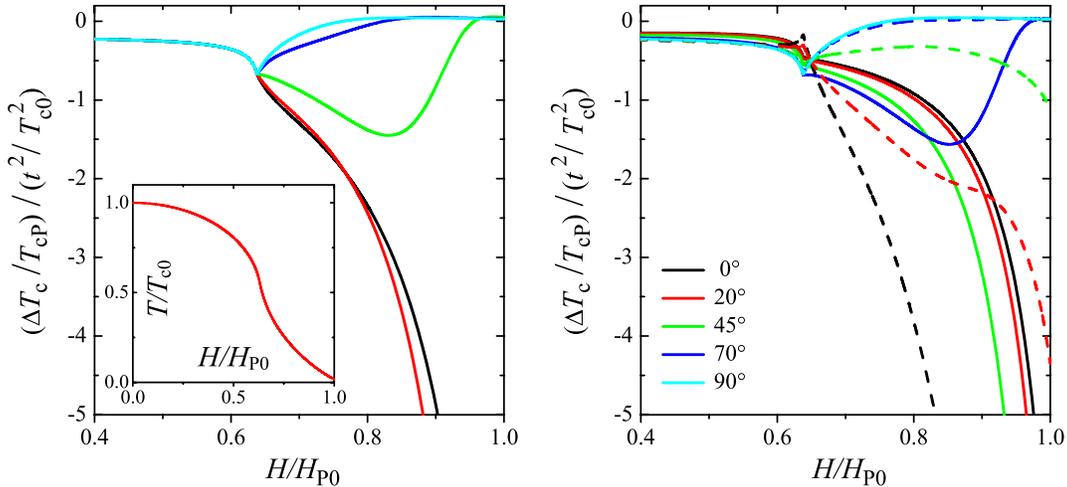


FIG. 1 (color online). Reduced critical temperature (with respect to the critical temperature in the pure paramagnetic limit) as a function of the in-plane magnetic field in a layered superconductor, for several angles  $\alpha$  between the field and  $x$  axis (equivalently FFLO modulation vector at  $H > H^*$ ). Left panel: isotropic regime with  $m_x = m_y$ . [Inset:  $(H, T)$ -phase diagram in the pure paramagnetic limit.] Right panel: anisotropic regime. Solid lines are for  $m_x = 10m_y$ ; dashed lines are for  $m_x = 0.1m_y$ . The calculations are performed for Fermi velocity  $v_F = 2 \times 10^7 \text{ cm} \cdot \text{s}^{-1}$  [15], and interlayer distance  $d = 1.3 \text{ nm}$ .

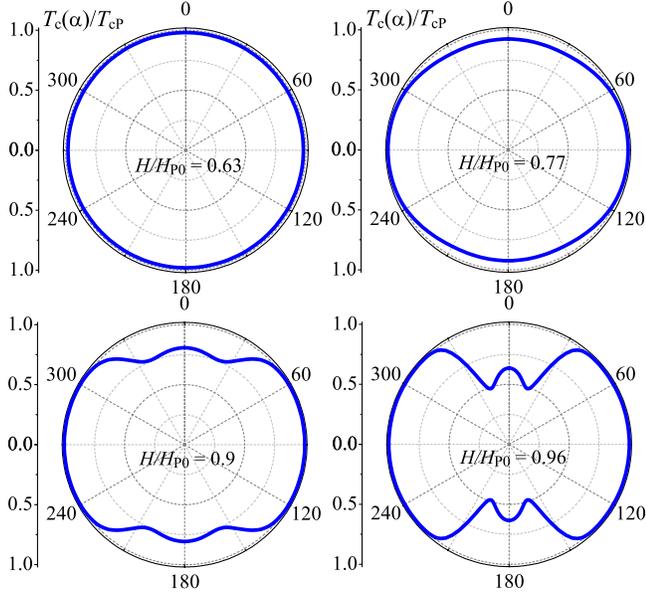


FIG. 2 (color online). Normalized superconducting transition temperature,  $T_c(\alpha)/T_{cp}$  as a function of the angle between the directions of the applied magnetic field and the vector  $\mathbf{q}$  for several values of  $H/H_{p0}$  and for  $m_x = m_y$ . Here  $t/T_{c0} = 0.2$ .

behavior of the upper critical field. When increasing  $H$  above  $H^*$ , a strong in-plane anisotropy of  $H_{c2}$  develops, which remains and becomes essentially pronounced at high fields. In particular, relatively strong dips at  $\alpha = \pm 18^\circ$  and  $\alpha = \pm 162^\circ$  with small peaks at  $0^\circ$  and  $180^\circ$  develop with the external magnetic field for the case of the isotropic in-plane Fermi surface. The maximum transition temperature is for the magnetic-field orientation perpendicular to the direction of the FFLO modulation vector along the  $x$  axis.

The peculiar features of the anisotropy of the superconducting transition onset with the orientation of the in-plane magnetic field strongly depend on the anisotropy of the Fermi surface. For example, comparing the curves in Fig. 3, one can notice the totally different in-plane field-direction dependence of  $H_{c2}$  above the tricritical point when the largest electron mass is along the  $x$  (solid line) or  $y$  direction (dashed line). For an in-plane mass anisotropy  $m_x = 10m_y$ , the positions of the dips are at angles  $\alpha = \pm 50^\circ$  and  $\pm 130^\circ$ , respectively. The positions of the small peaks are the same as in the isotropic case; however, they are strongly broadened. For opposite mass anisotropy  $m_x/m_y = 0.1$ , the dips are strengthened and are found now at  $\alpha = \pm 5^\circ$  and  $\pm 175^\circ$ . The fine structure at  $\alpha = 0^\circ, 180^\circ$  is relatively sharpened. Therefore the shape of the observed  $T_c(\alpha)$  dependence could permit us to determine the orientation of the FFLO modulation vector. However, the common feature of the field evolution of the in-plane upper critical field anisotropy observed in all considered cases is that the anisotropy becomes more pronounced with the field strength. Furthermore, the maximum critical temperature in the FFLO regime always corresponds to

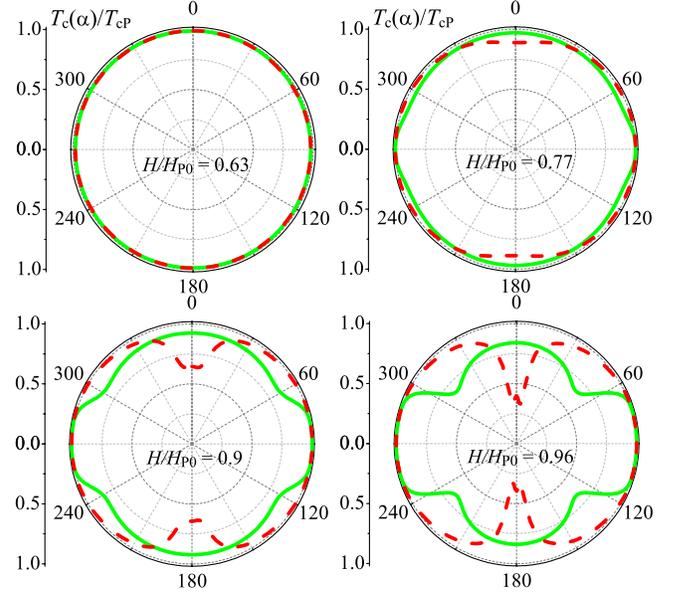


FIG. 3 (color online). The same as in Fig. 2 but for  $m_x = 10m_y$  (solid line)  $m_x = 0.1m_y$  (dashed line).

$\alpha = \pm 90^\circ$ , that is  $\mathbf{H}$  perpendicular to  $\mathbf{q}$ , irrespective of the effective mass ratio. This is in contrast with the usual behavior near  $T_{c0}$ , where the critical temperature is maximal for the magnetic field oriented along the lightest mass axis.

In conclusion, our results show that the FFLO modulation strongly interferes with the orbital effect and provides the main source of the in-plane critical field anisotropy. The superconducting onset temperature is maximal for the field oriented perpendicular to the FFLO modulation vector. The change of the anisotropy of the critical field as well as of its fine structure may give important information about the FFLO state and unambiguously prove its existence. Our calculations support the interpretation of the experimentally observed in-plane anisotropy of the onset of superconductivity in  $(\text{TMTSF})_2\text{ClO}_4$  samples as a realization of the FFLO state with the modulation vector close to the  $\mathbf{b}^*$  axis [9]. However, the compound  $(\text{TMTSF})_2\text{ClO}_4$  is in fact in the regime  $t \gtrsim T_{c0}$  ( $t \sim 2-7$  K and  $T_{c0} = 1.45$  K) [9,15]. In this case the FFLO vector can be changed by the orbital effect. Its orientation will result from the interplay of the Fermi surface nonellipticity, which favors pinning of  $\mathbf{q}$  in a certain direction, and the orbital effect, which prefers to orient  $\mathbf{q}$  perpendicular to  $\mathbf{H}$ . Nevertheless we expect that the obtained results will be qualitatively applicable in this case as well. We suggest that the predicted in-plane anisotropy of  $H_{c2}$  can be observed in experiments with  $\kappa$ -(BEDT-TTF) $_2\text{Cu}(\text{NCS})_2$  organic superconductors [31,32]. For this salt the angle-dependent magnetoresistance measurements [33] provide the estimate of the inter-layer transfer integral  $t \approx 1-2$  K, which is much smaller than  $T_{c0} = 9.1$  K, and the orbital effect should only

slightly change the modulation vector [34]. In this work we have assumed  $s$ -wave superconductivity; however, it is not an important ingredient in the present theory and we expect similar results in the case of  $d$ -wave pairing, which provides an additional source of pinning for the modulation vector [35].

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