INTERPLAY BETWEEN ORBITAL EFFECT AND THE NON-UNIFORM (FFLO) STATE IN QUASI-LOW-DIMENSIONAL SUPERCONDUCTORS

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in collaboration with

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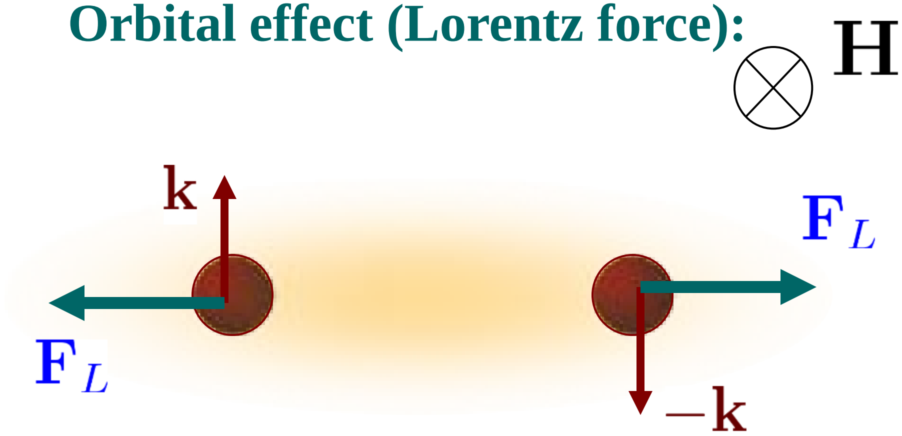
Outline

- Possible experimental evidence of the FFLO state in organic superconductors and other superconducting compounds;
- Motivation for the present research;
- Quasi-classical description of the FFLO state for layered superconductors;
- Strong interference of the FFLO modulation with the orbital motion as a main source of
    - in-plane upper critical field anisotropy,
    - resonance in-plane magnetic field effect,
    - FFLO modulation wave vector lock-in effect;
- Conclusions.

Breakdown of Cooper pairs by a magnetic field

Orbital effect (Lorentz force):



$$H_{c2}^{orb} = \frac{\Phi_0}{2\pi\xi_0^2}$$

Maki parameter

$$\alpha_M = \frac{\sqrt{2}H_{c2}^{orb}}{H_{c2}^{P}} \ll 1$$

Paramagnetic effect (singlet pair) :

$$s_z = \frac{1}{2} \qquad s_z = -\frac{1}{2}$$

$$H_{c2}^{P} = \frac{\sqrt{2}\Delta_0}{g\mu_B}$$

FFLO inventors

P. Fulde and R. A. Ferrell

A. I. Larkin and Yu. N. Ovchinnikov

P. Fulde, R. A. Ferrell, Phys.Rev. **135**, A550 (1964)
A. I. Larkin, Yu. N. Ovchinnikov, JETP **47**, 1138 (1964)

Superconducting order parameter behavior under paramagnetic effect

Standard Ginzburg-Landau functional:

$$F = a|\Psi|^2 + \frac{1}{4m}|\nabla\Psi|^2 + \frac{b}{2}|\Psi|^4$$

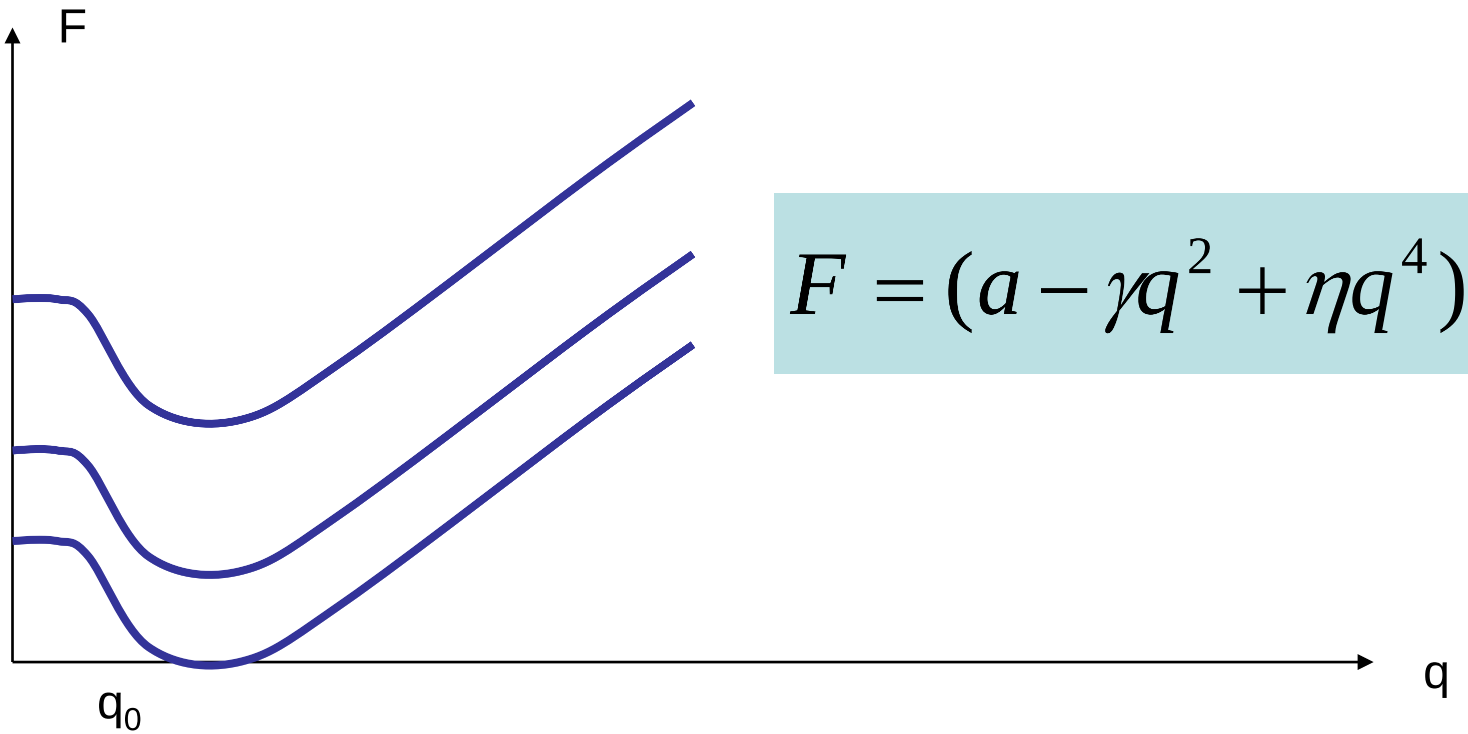
The minimum energy corresponds to Ψ=const

The coefficients of GL functional are functions of the Zeeman field h= $\mu_B H$ **!**

Modified Ginzburg-Landau functional ! :

$$F = a|\Psi|^2 - \gamma|\nabla\Psi|^2 + \eta|\nabla^2\Psi|^2 + \ldots$$

The **non-uniform** state $\Psi \sim \exp(iqr)$ will correspond to minimum energy and higher transition temperature



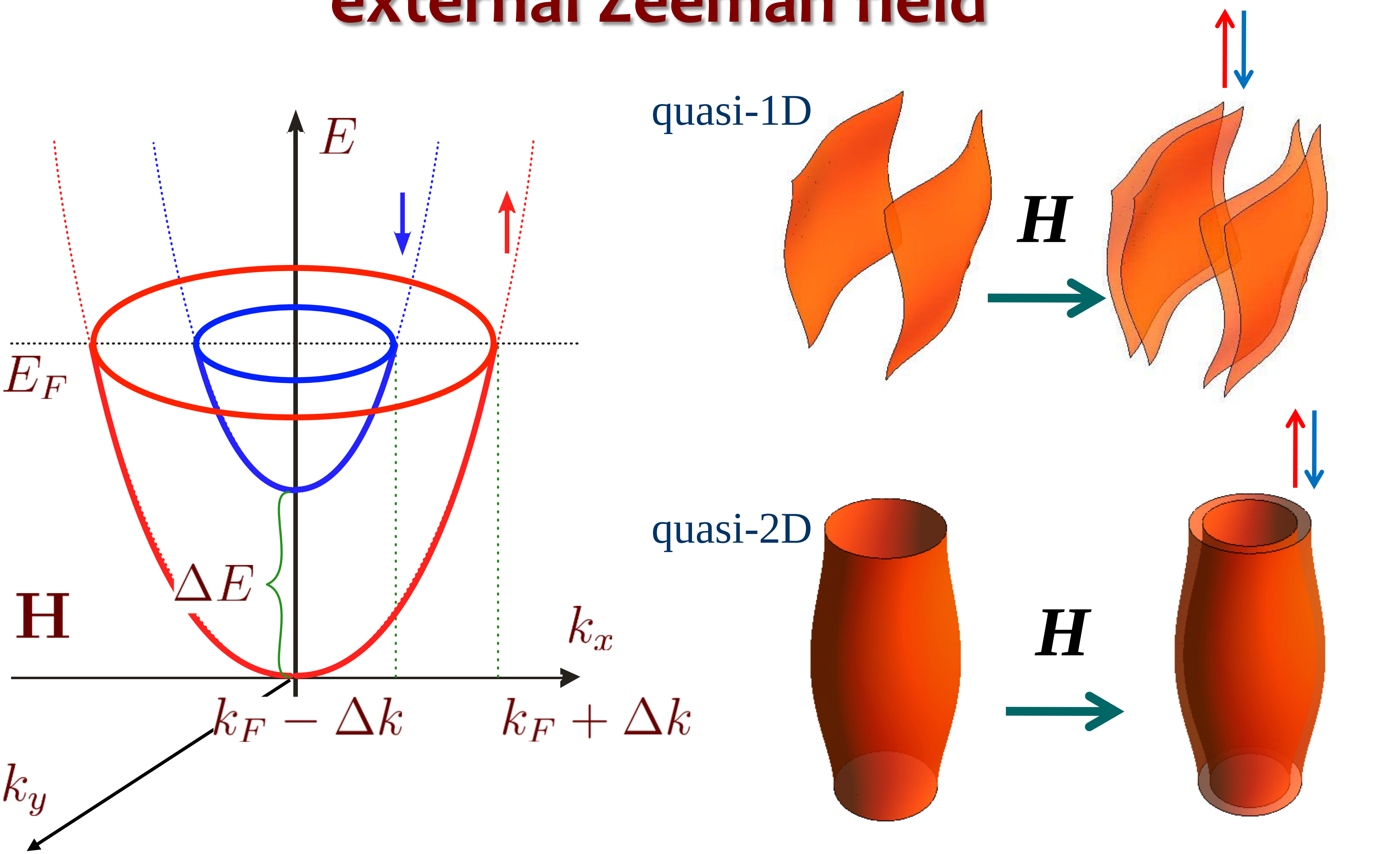
$$F = (a - \gamma q^2 + \eta q^4)\left|\Psi_q\right|^2$$

$\Psi \sim \exp(iqr)$ **- Fulde-Ferrell-Larkin-Ovchinnikov state (1964).**

Only in pure superconductors and in the rather narrow region.

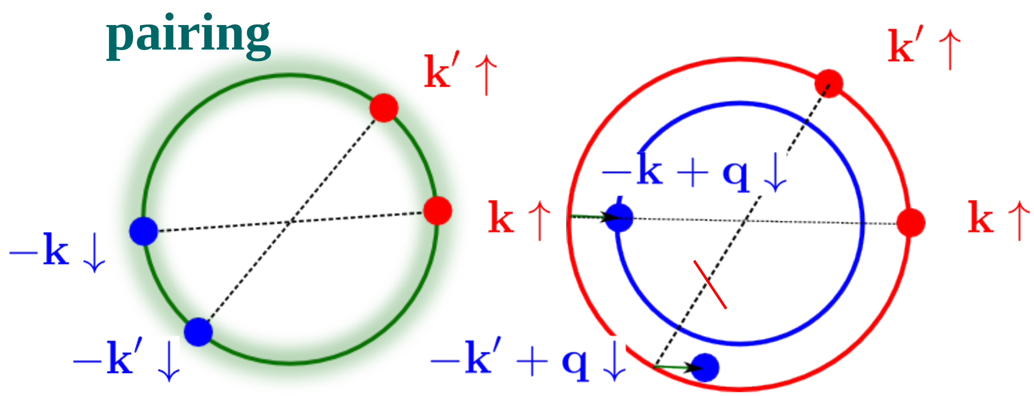
P. Fulde and R. A. Ferrell. Phys. Rev. **135**, A550 (1964).
A. Larkin and Y. Ovchinnikov. Sov. Phys. JETP **20**, 762 (1965).

Population polarization due to an external Zeeman field

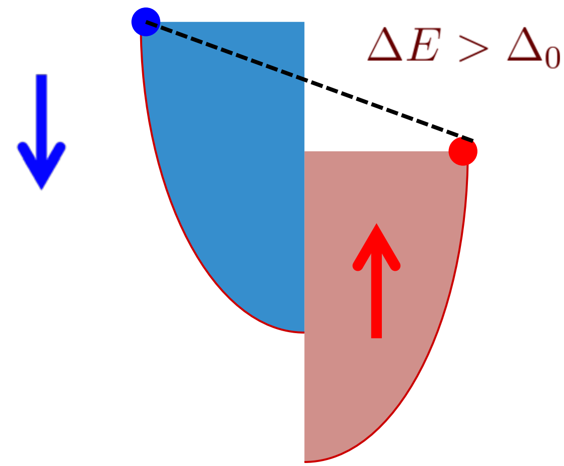


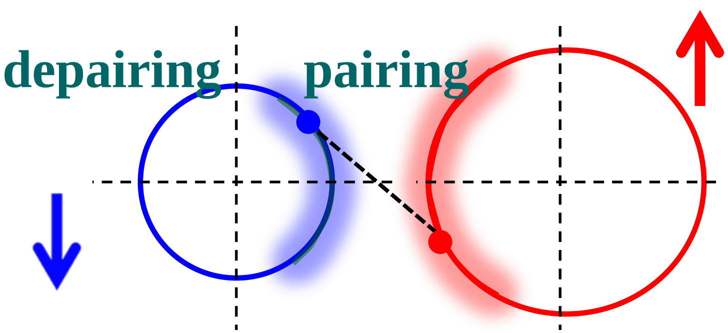
- Zeeman field splits conduction band;
- Fermi surfaces of the spin-up and -down bands are mismatched.

Fulde-Ferrell-Larkin-Ovchinnikov State

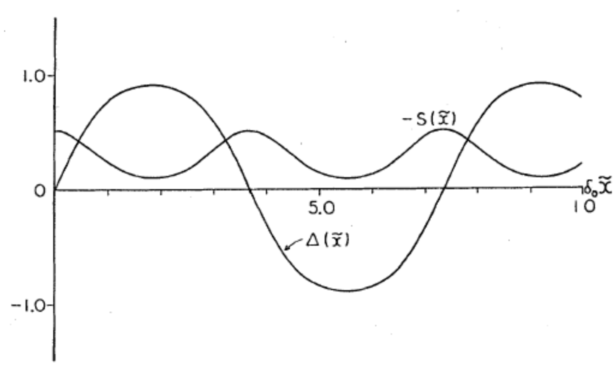


$\mathbf{k}+(-\mathbf{k})=0$ $\qquad$ $\mathbf{k}+(-\mathbf{k}+\mathbf{q})=\mathbf{q}$





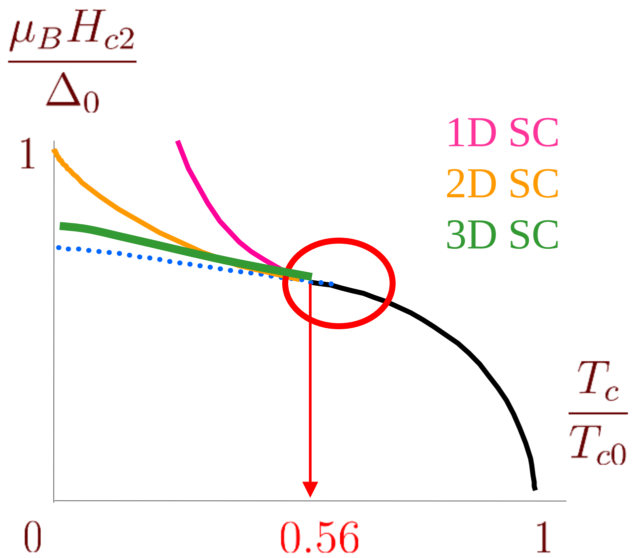
- Fermi surface mismatch $\longrightarrow$ pair breaking;
- It is difficult to form pairs with zero total momentum;
- Can lead to a pairing with non-zero total pair momentum;

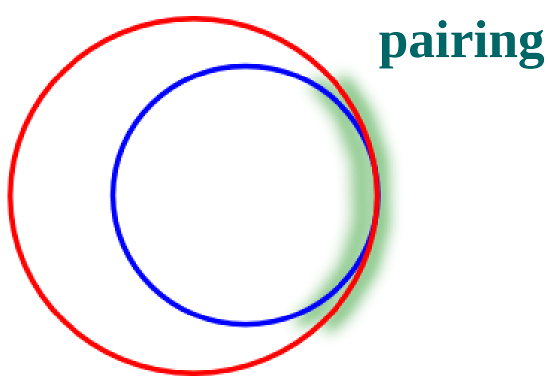


$$\varepsilon(\mathbf{k}+\mathbf{q}) - \mu_B H^{eff} \approx \varepsilon(-\mathbf{k}+\mathbf{q}) + \mu_B H^{eff}$$

if $$q \approx \frac{\mu_B H^{eff}}{v_F}$$

Requirements for the formation of the FFLO sate





- Strongly type-II superconductors with very large Ginzburg-Landau parameter

$$\kappa \equiv \frac{\lambda}{\xi} \gg 1$$

- Large Maki parameter, such that the upper critical field can easily approach the Pauli paramagnetic limit:

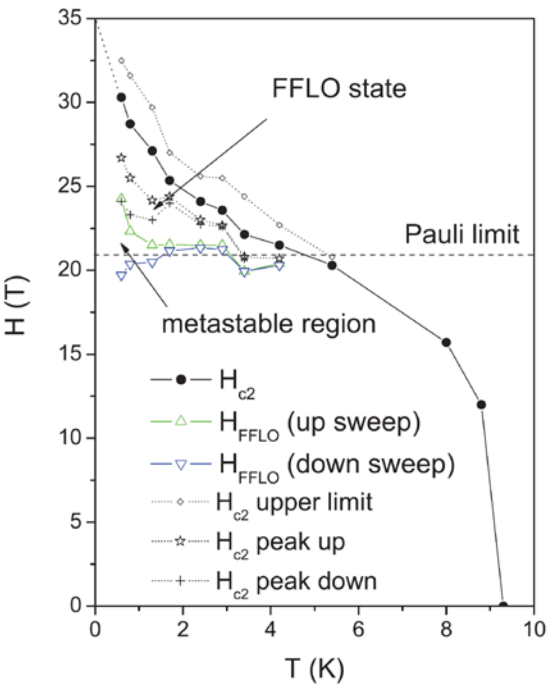
$$\alpha_M = \frac{\sqrt{2} H_{orb}}{H_P} > 1.8$$

- Very clean, $\xi \ll l$, since the FFLO state is very sensitive to the presence of impurities.

- Anisotropies of the Fermi surface and the gap function can stabilize the FFLO state. Stability of FFLO state crucially depends on the dimensionality of the system.

Thermodynamic evidence for FFLO phase in the layered organic superconductor

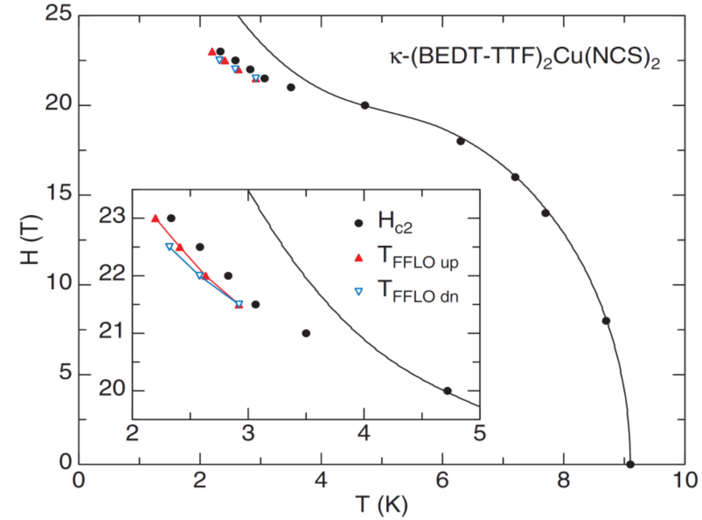
$$\kappa\text{-}\,(\mathrm{BEDT\text{-}TTF})_2\,\mathrm{Cu}\,(\mathrm{NCS})_2$$

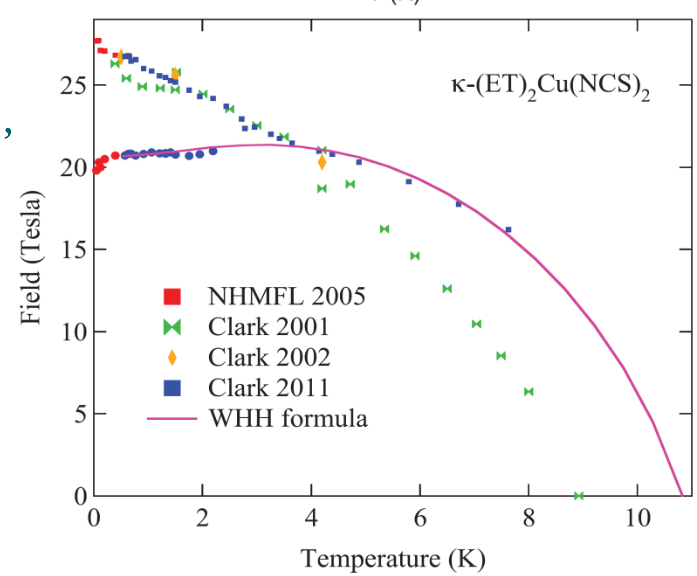


Magnetic torque measurements of magnetization: B. Bergk, et. al., Phys. Rev. B **83**, 064506 (2011)

High-resolution specific heat measurements: R. Lortz, et. al., Phys. Rev. Lett. **99**, 187002 (2007)

In-plane rf-penetration depth measurement: C. C. Agosta *et al.*, Phys. Rev. B **85**, 214514 (2012)





$$\xi = 9\ \mathrm{nm}$$

$$l = 100\ \mathrm{nm}$$

$$H_{orb} = 130\ \mathrm{T}$$

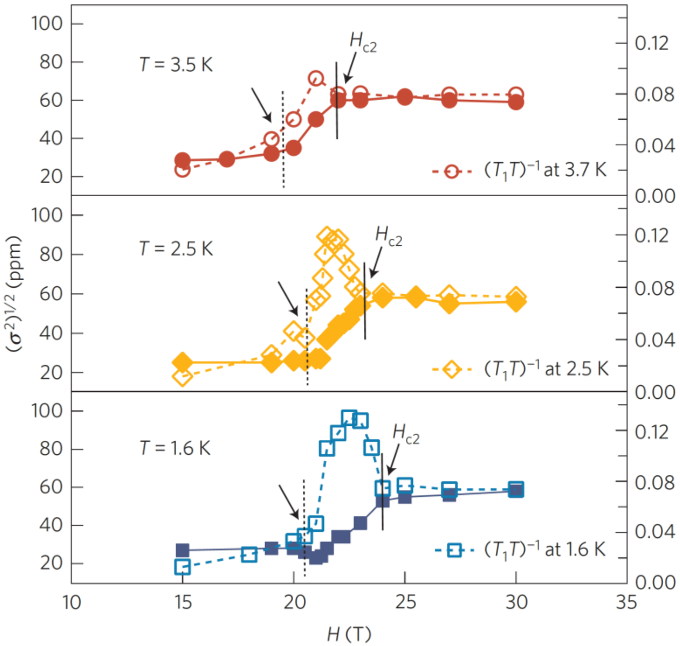
$$H_P = 23\ \mathrm{T}$$

$$\alpha_M = \frac{\sqrt{2}H_{orb}}{H_P} \approx 8 \gg 1.8$$

$$\xi \ll l \qquad \varepsilon_F / t_\perp = 3700$$

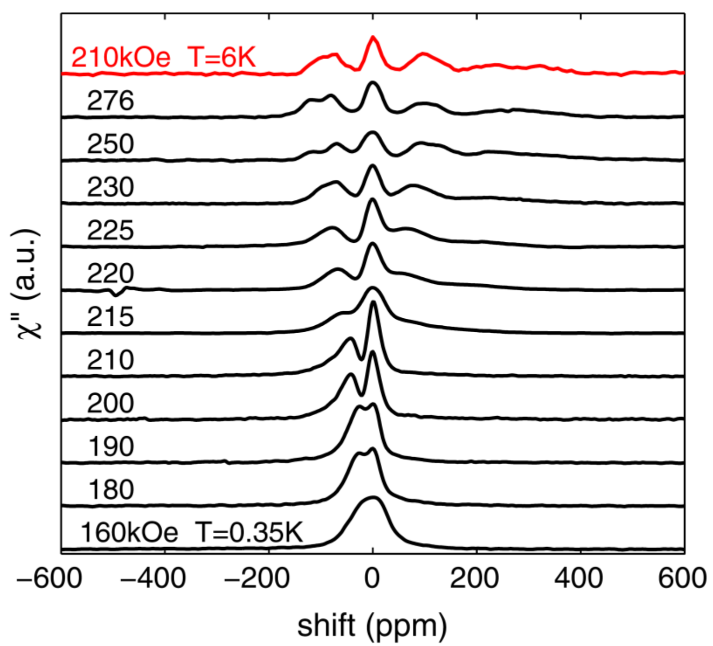
Microscopic evidence for FFLO phase in the layered organic superconductor

$\kappa\text{-}\left(\mathrm{BEDT\text{-}TTF}\right)_2\mathrm{Cu}\left(\mathrm{NCS}\right)_2$



Nuclear magnetic resonance (NMR) measurements: H. Mayaffre, et. al., Nat. Phys. 10, 928 (2014)

Appearance of spatially localized and spin-polarized quasiparticles forming the Andreev bound states as a hallmark for the FFLO state with nodes of Δ localising ABS.

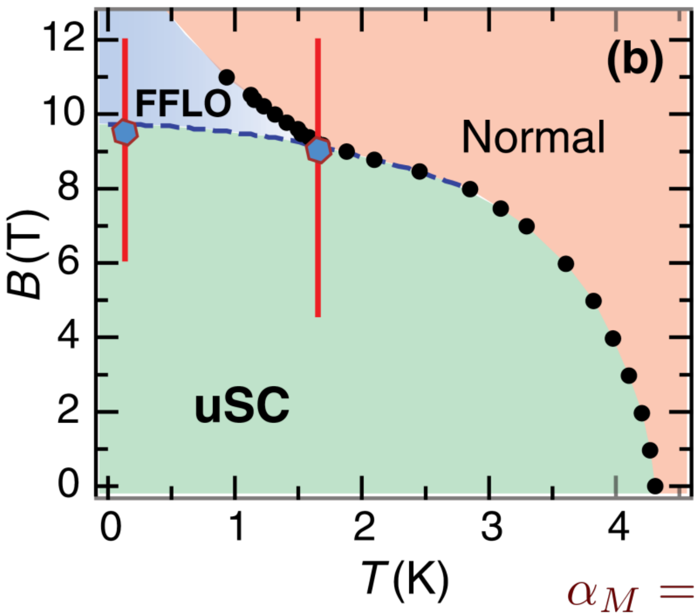


^{13}C NMR spectroscopy measurements: J. A. Wright, *et al.*, Phys. Rev. Lett. **107**, 087002 (2011).

• A change of behavior of electronic spin susceptibility at Hs is taken as evidence for a Zeeman driven phase transition within the superconducting state and stabilization of inhomogeneous superconductivity.

Evidence for FFLO phase in the layered organic superconductor

$$\beta''\text{-}\,(\mathrm{BEDT\text{-}TTF})_2\,\mathrm{SF_5CH_2CF_2SO_3}$$



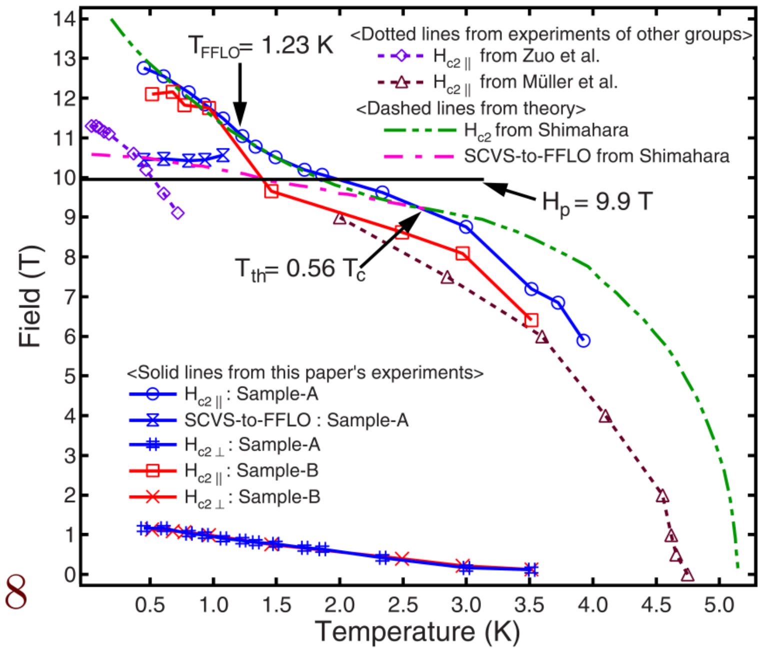
$$\xi = 15\ \mathrm{nm}$$

$$l = 100\ \mathrm{nm}$$

$$H_{orb} = 75\ \mathrm{T}$$

$$H_P = 9.73\ \mathrm{T}$$

$$\alpha_M = \frac{\sqrt{2}H_{orb}}{H_P} \approx 11 \gg 1.8$$



High-resolution specific heat measurements (thermodynamic technique): R. Beyer, et. al., Phys. Rev. Lett **109**, 027003 (2012)

[13]C NMR NMR studies: G. Koutroulakis, *et al.*, Phys. Rev. Lett. **116**, 067003 (2016).

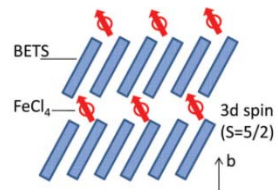
In-plane penetration depth measurements by use of TDO technique: K. Cho *et al.*, Phys. Rev. B **79**, 220507(R) (2009)

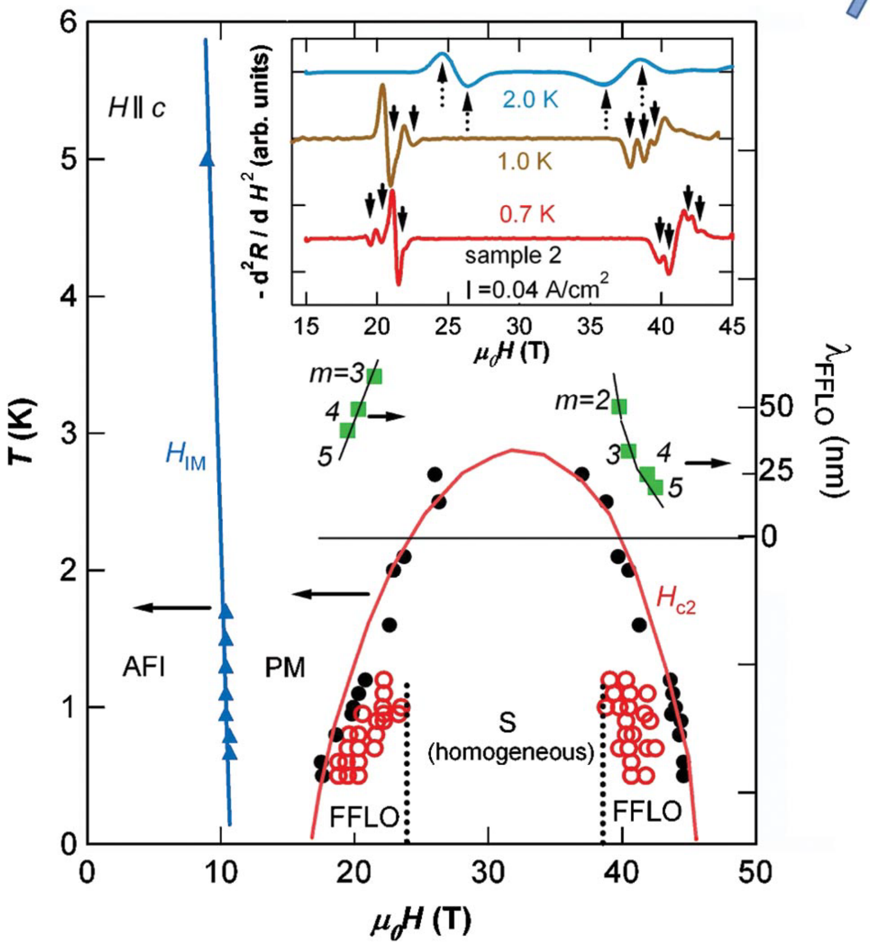
Direct evidence for FFLO phase. Intrinsic Josephson vortex pinning in the FFLO phase for parallel field orientation.

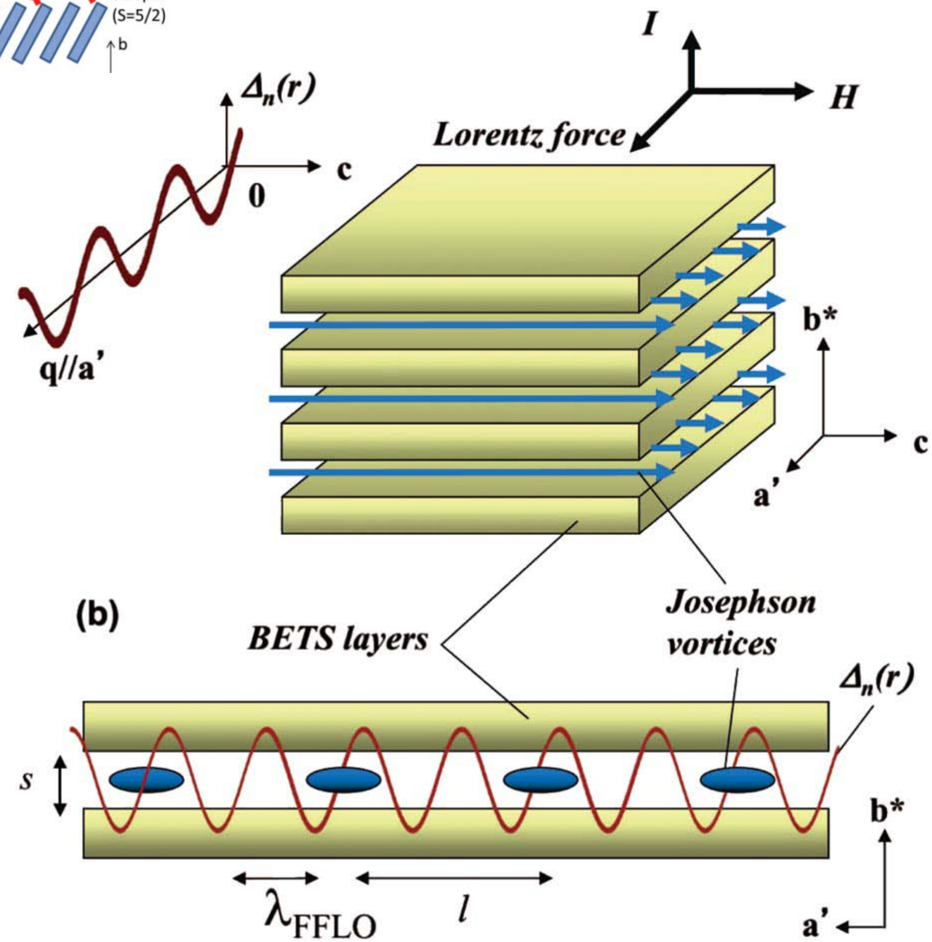
Jaccarino-Peter mechanism

$$\mu_B H^{eff} = \mu_B H - J \langle S \rangle$$

λ- $(BETS)_2$ $FeCl_4$



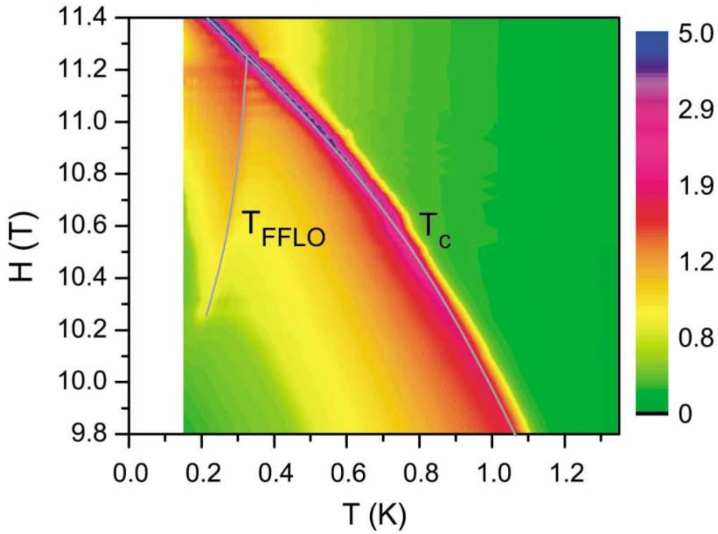




S. Uji *et al.*, Phys.Rev.Lett. **97**, 157001 (2006)

L. Bulaevskii *et al.*, Phys.Rev.Lett. **90**, 067003 (2006)

FFLO state in Heavy Fermion superconductors (thermodynamic and microscopic) $CeCoIn_5$



$$\alpha_M^{\parallel} \approx 4.5$$

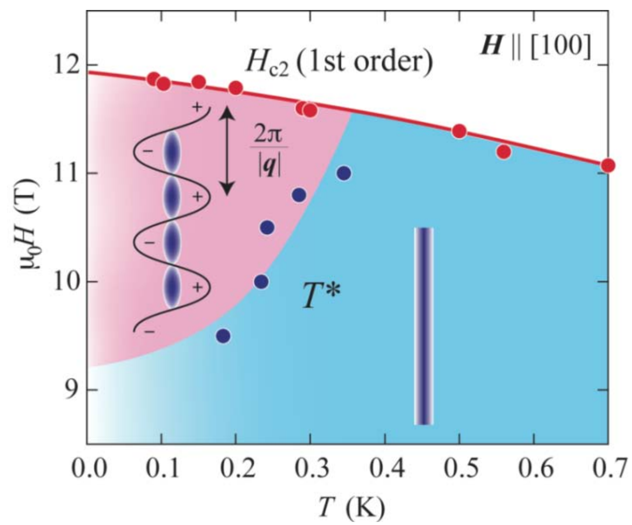
$$\alpha_M^{\perp} \approx 3.6$$

$$T_c = 2.3 \text{ K}$$

$$l = 81 \text{ nm}$$

$$\xi = 6 \text{ nm}$$

$$H_P = 7.3 \text{ T}$$

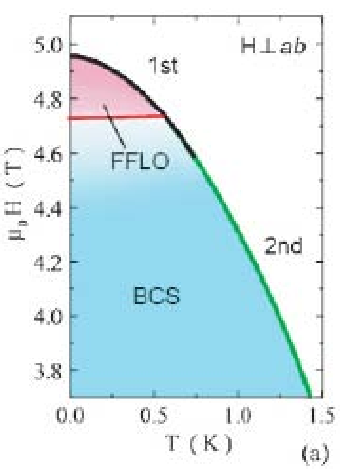


Specific heat measurements: (*i*) 2-order to 1-order phase transition; (*ii*) a second specific heat anomaly within SC state.

A. Bianchi, et. al., Phys. Rev. Lett. **91**, 187004 (2003)

Ultrasound velocity measurements: reveal an important decrease of the superconducting volume fraction at the FFLO transition.

T. Watanabe, et. al., Phys. Rev. B **70**, 020506 (R) (2004)



Penetration depth measurement (TDO): Two phase transitions were found.

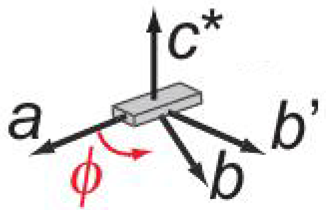
C. Martin *et al.*, Phys. Rev. B **71**, 020503(R) (2005)

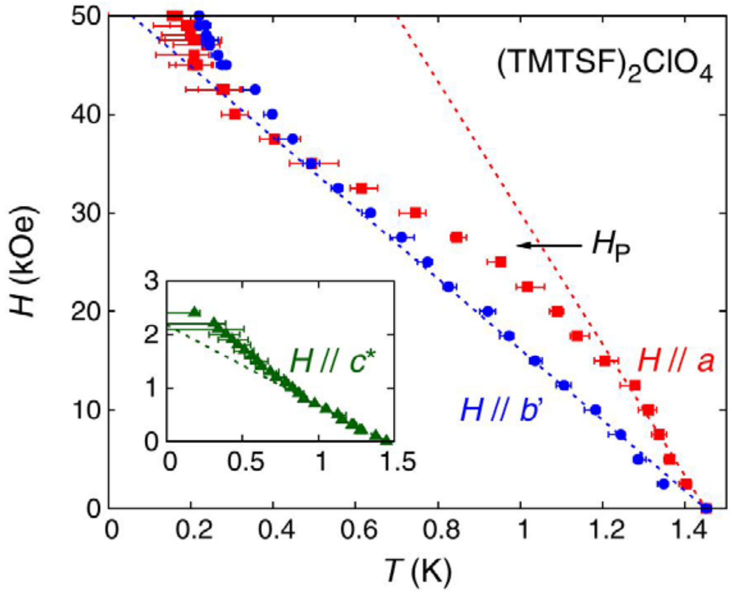
NMR measurement (TDO):

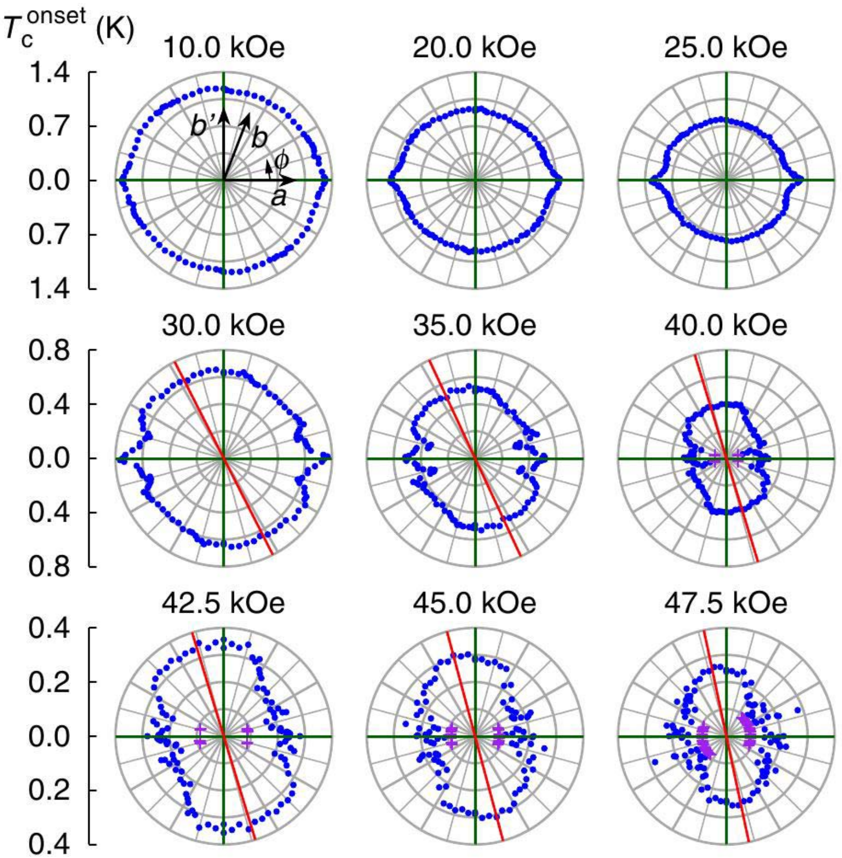
C. Martin *et al.*, Phys. Rev. B **71**, 020503(R) (2005)

Ultrasound and NMR results are consistent with the FFLO state which predicts a segmentation of the flux line lattice

Anomalous in-plane anisotropy of the onset of SC in $(TMTSF)_2ClO_4$

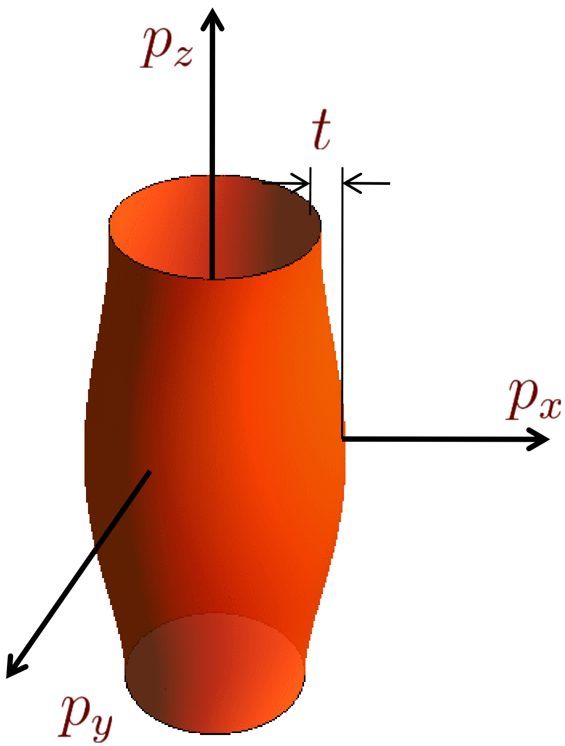


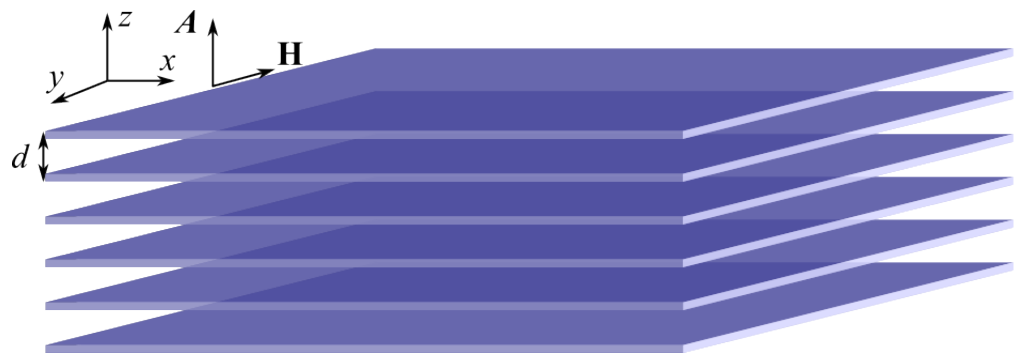




S. Yonezawa, S. Kusaba, Y. Maeno, P. Auban-Senzier, C. Pasquier, K.Bechgaard, and D. Jerome, Phys. Rev. Lett. **100**, 117002 (2008)

Model system





$$\mathbf{A} = \mathbf{H} \times \mathbf{r} \quad [\mathbf{r} = (x, y, 0)]$$

$$A_z = -xH \sin\alpha + yH \cos\alpha$$

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

We assume that the corrugation of the Fermi surface is small

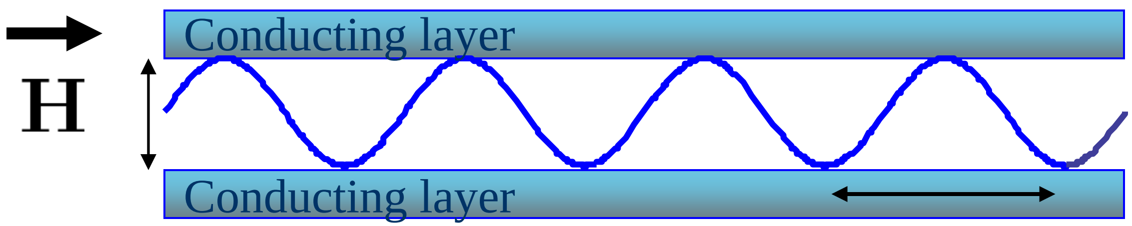
$$T_{c0}^2 / E_F \ll t \ll T_{c0}$$

Eilenberger equation for layered SC

Taking into account that the system is near the second-order phase transition, the linearized Eilenberger equation describing layered superconducting systems acquires the form

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

Here: $\mathbf{v}_F = v_F\mathbf{n}$ $\qquad$ $\mathbf{Q} = \frac{\pi d}{\phi_0} H \left(-\sin\alpha, \sqrt{\frac{m_x}{m_y}}\cos\alpha, 0\right)$



$$t\sin\left(\mathbf{Q}.\mathbf{r}\right)$$

$$\lambda = \frac{\phi_0}{\pi d H}$$

The order parameter is defined self-consistently as (*s*-wave)

$$\frac{1}{\lambda}\Delta\left(\mathbf{r}\right) = 2\pi T\,\mathrm{Re}\sum_{\omega>0}\left\langle f_\omega\left(\mathbf{n},\mathbf{r},p_z\right)\right\rangle \qquad \langle\ldots\rangle \equiv \int_{-\pi/d}^{\pi/d}\frac{d\ dp_z}{2\pi}\int_0^{2\pi} d\varphi\left(\ldots\right)$$

J. P. Brison et al., Physica C **250**, 128 (1995).

Choice of the solution

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

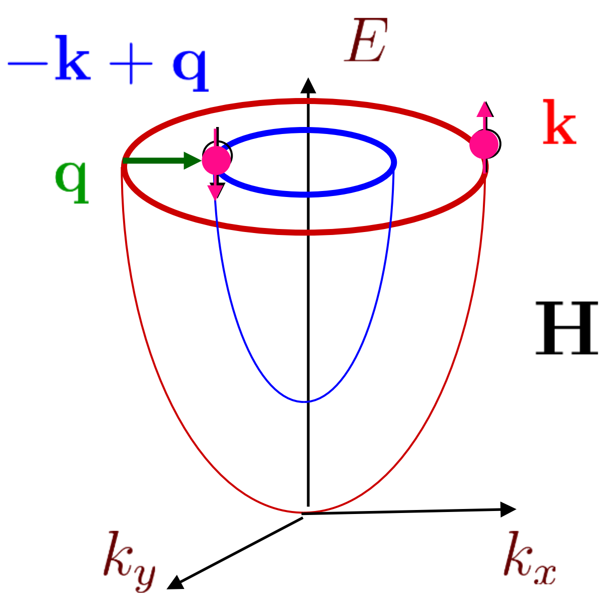
$$\left(\mathbf{k}+\frac{\mathbf{q}}{2}\uparrow;-\mathbf{k}+\frac{\mathbf{q}}{2}\downarrow\right)$$

$$f_\omega\left(\mathbf{n},\mathbf{r},p_z\right)=e^{i\mathbf{qr}}\sum_m e^{im\mathbf{Q}.\mathbf{r}}f_m\left(\omega,\mathbf{n},p_z\right)$$

$$t/T_{c0}\ll 1$$

At the same time, the order parameter can be expanded as

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



This expansion takes into account the possibility for the formation of the pairing state with finite center-of-mass momentum: $q\sim g\mu_B H/v_F$

- |**q**| is taken that maximizes H_{c2} calculated in the Pauli paramagnetic limit.
- **The direction of the FFLO modulation vector is fixed by the symmetry of the Fermi surface**.

M. D. C., M. Houzet, and A. I. Buzdin, Phys. Rev. Lett. **108**, 207005 (2012).

Solution

Making use of the self-consistency relation we finally obtain

$$\Delta_0\left(P+t^2a\right)=\Delta_{\pm2}t^2c_{\pm},$$

$$\Delta_{\pm2}\left(P+t^2b_{\pm}+\delta_{\pm}\right)=\Delta_0t^2c_{\pm},$$

Here: $P=-\left(T_{cP}-T_c\right)/AT_c$

$$A=1-\frac{\mu_BH}{T_{cP}}\frac{\partial T_{cP}}{\partial\mu_BH}$$

$$\delta_{\pm}=\pi T\sum_n\left\langle\frac{1}{L_n\left(\mathbf{q}\right)}\right\rangle-\left\langle\frac{1}{L_n\left(\mathbf{q}\pm2\mathbf{Q}\right)}\right\rangle\Bigg|_{T=T_{cP}}$$

$$\Rightarrow|\mathbf{q}\pm2\mathbf{Q}|=|\mathbf{q}|$$

$$T_c=T_{cP}/\left[1+A(S_{\mathrm{O}}+S_{\mathrm{R}})\right] \qquad S_{\mathrm{O}}\equiv t^2a$$

General case: $S_{\mathrm{R}}^{\pm}\equiv-\dfrac{\left(a-b_{\pm}\right)t^2-\delta_{\pm}}{2}-\dfrac{t^2}{2}\sqrt{\left(a-b_{\pm}-\delta_{\pm}/t^2\right)^2+4c_{\pm}^2}$

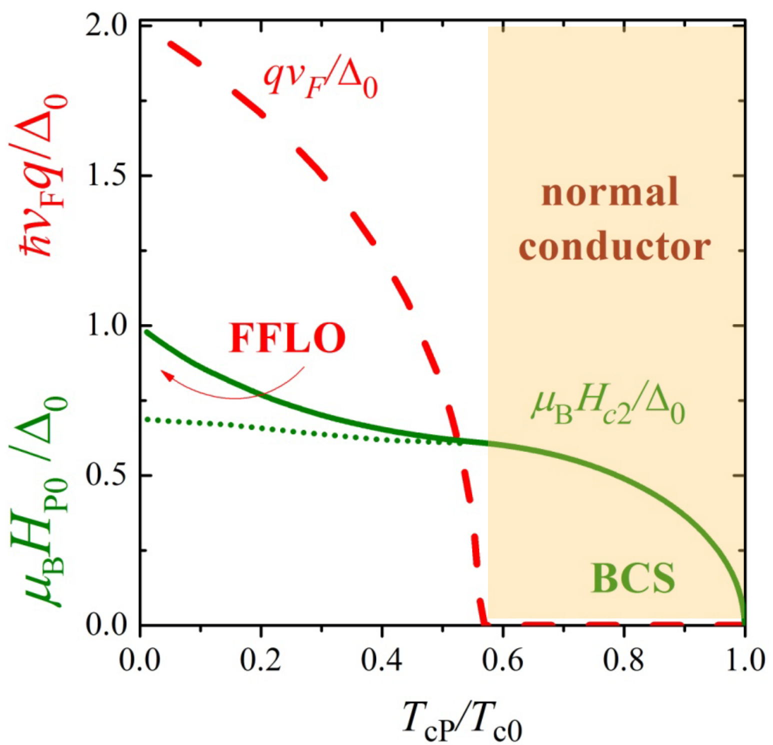
Resonance: $S_{\mathrm{R}}^{\pm}\equiv-t^2c_{\pm}$

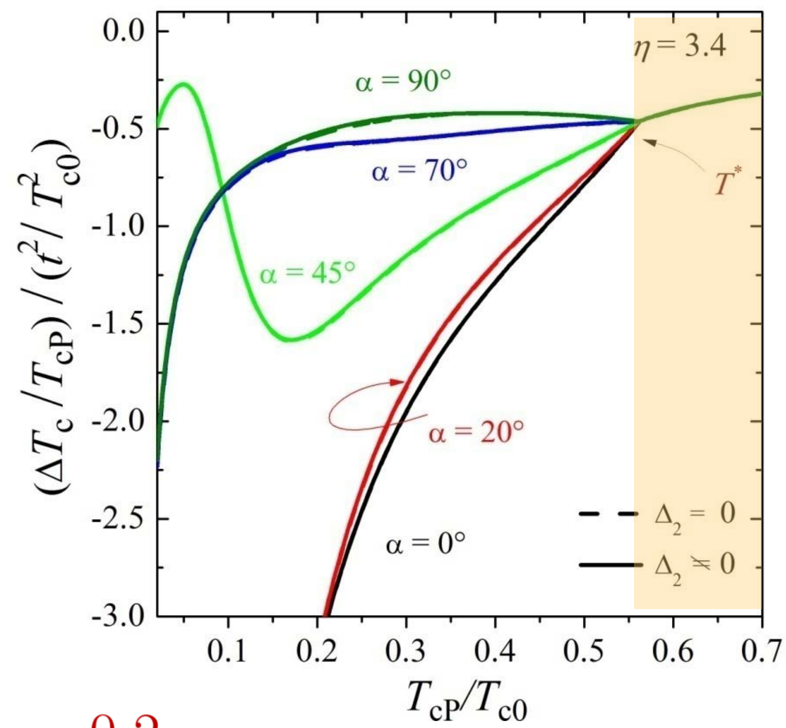
Out of resonance: $S_{\mathrm{R}}^{\pm}\ll1$ $\qquad t/T_{c0}\ll1$

Orbital correction (out of resonance)

Normalized orbital correction of the superconducting onset temperature as a function of in-plane magnetic field **H** for several angles between magnetic field and the FFLO modulation vector **q**.

$$v_F = 1.0 \times 10^5 m/s$$





$$t/T_{c0} = 0.2$$

$$d = 1.6\ nm$$

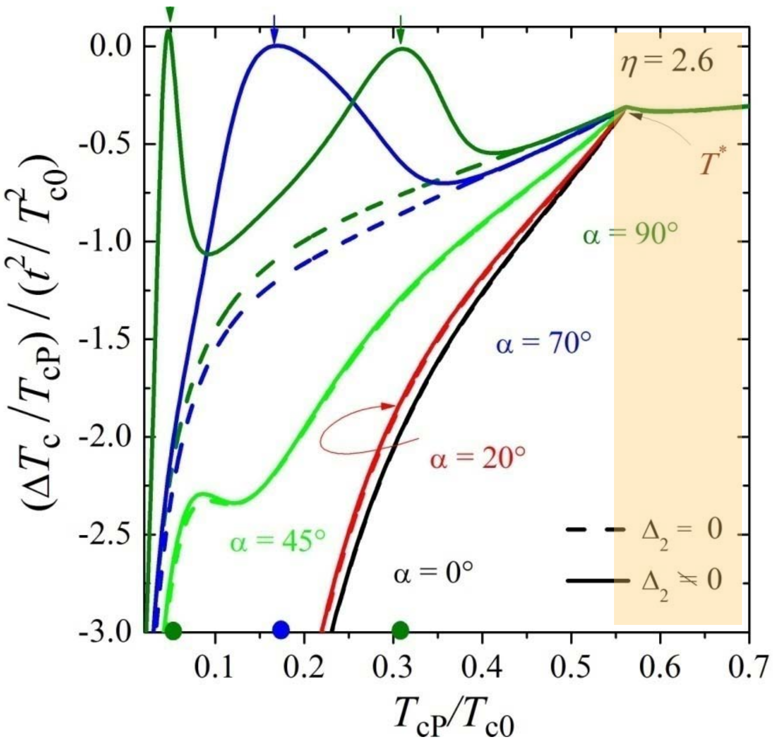
$$v_F = 1.0 \times 10^5 m/s \Rightarrow \eta = 3.4$$

$$\eta = \hbar v_F \pi d/\phi_0 \mu_B$$

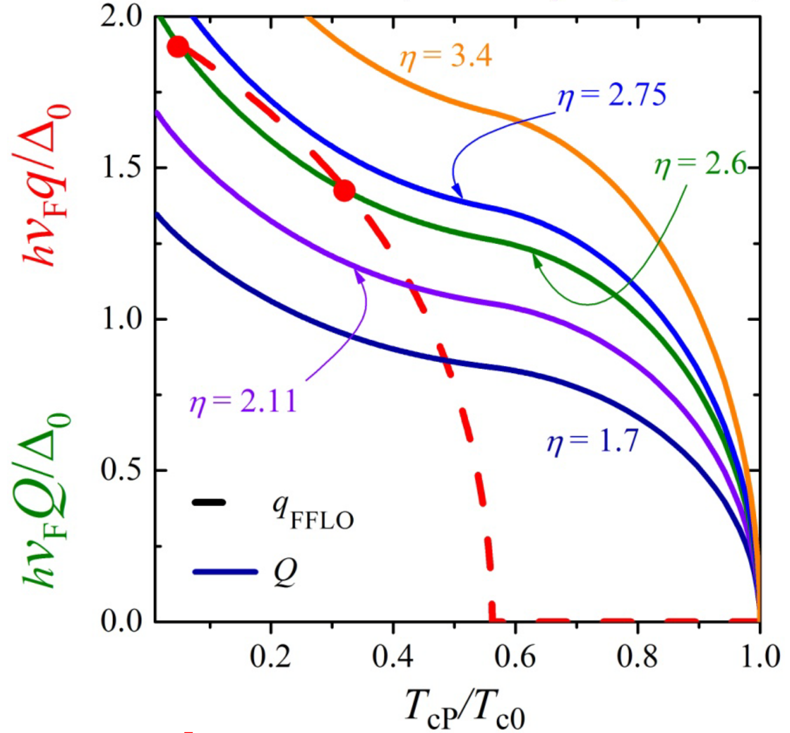
Orbital correction (resonance)

Absolute value of the wave vector **q** of the FFLO phase (dashed lines) and of the wave vectors **Q** (solid lines) versus the reduced temperature T_{cP}/T_{c0} calculated for several values of Fermi velocity $\eta = \hbar v_F \pi d/\phi_0 \mu_B$

$v_F = 0.75 \times 10^5 m/s$



$$\mathbf{Q} = \frac{\pi d}{\phi_0} H \left(-\sin\alpha, \sqrt{\frac{m_x}{m_y}}\cos\alpha, 0\right)$$



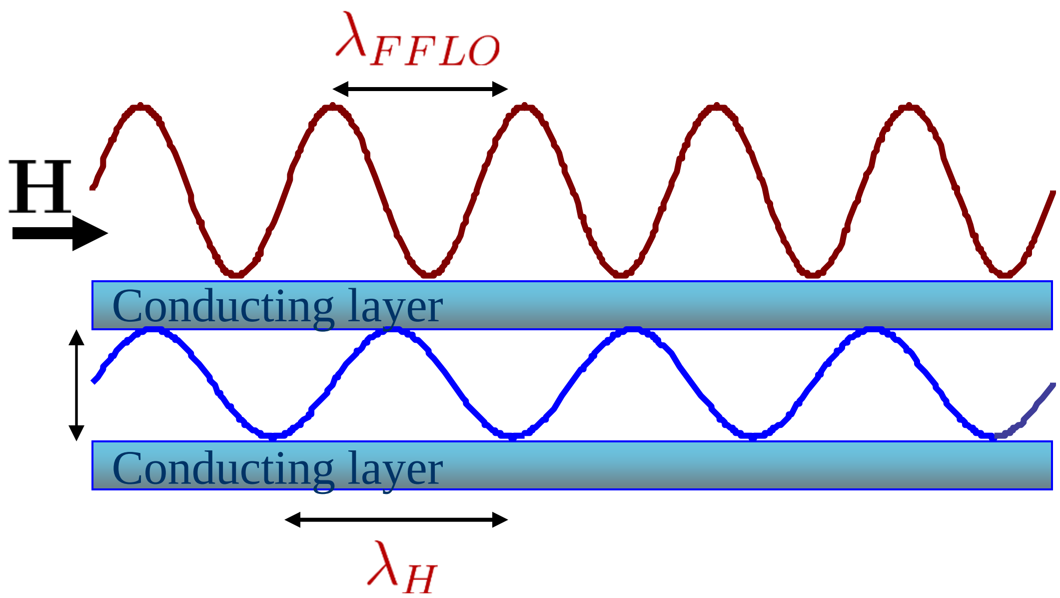
Resonance condition:

$$|\mathbf{q} \pm 2\mathbf{Q}| = |\mathbf{q}| \qquad \mathbf{q}.\mathbf{Q} = \mp Q^2 \;\Rightarrow\; q = \mp Q$$

Resonance condition

• Vector potential of the parallel magnetic field results in a modulation of the interlayer coupling;

$$\mathbf{Q} = \frac{\pi d}{\phi_0} H \left(-\sin\alpha, \cos\alpha, 0\right)$$



$$\Delta\left(\mathbf{r}\right) = \Delta_0 \cos\left(\mathbf{q}.\mathbf{r}\right) e^{i\phi_n(r)}$$

$$t\cos(\mathbf{Q}.\mathbf{r})$$

$$\Delta\left(\mathbf{r}\right) = \Delta_0 \cos\left(\mathbf{q}.\mathbf{r}\right) e^{i\phi_{n+1}(r)}$$

• The period of the magnetic wave vector induced modulation may interfere with the in-plane FFLO modulation leading to the anomalies in the critical field behavior;

Resonance condition: $|\mathbf{q} \pm 2\mathbf{Q}| = |\mathbf{q}|$ $\quad\quad$ $\mathbf{q}.\mathbf{Q} = \mp Q^2$

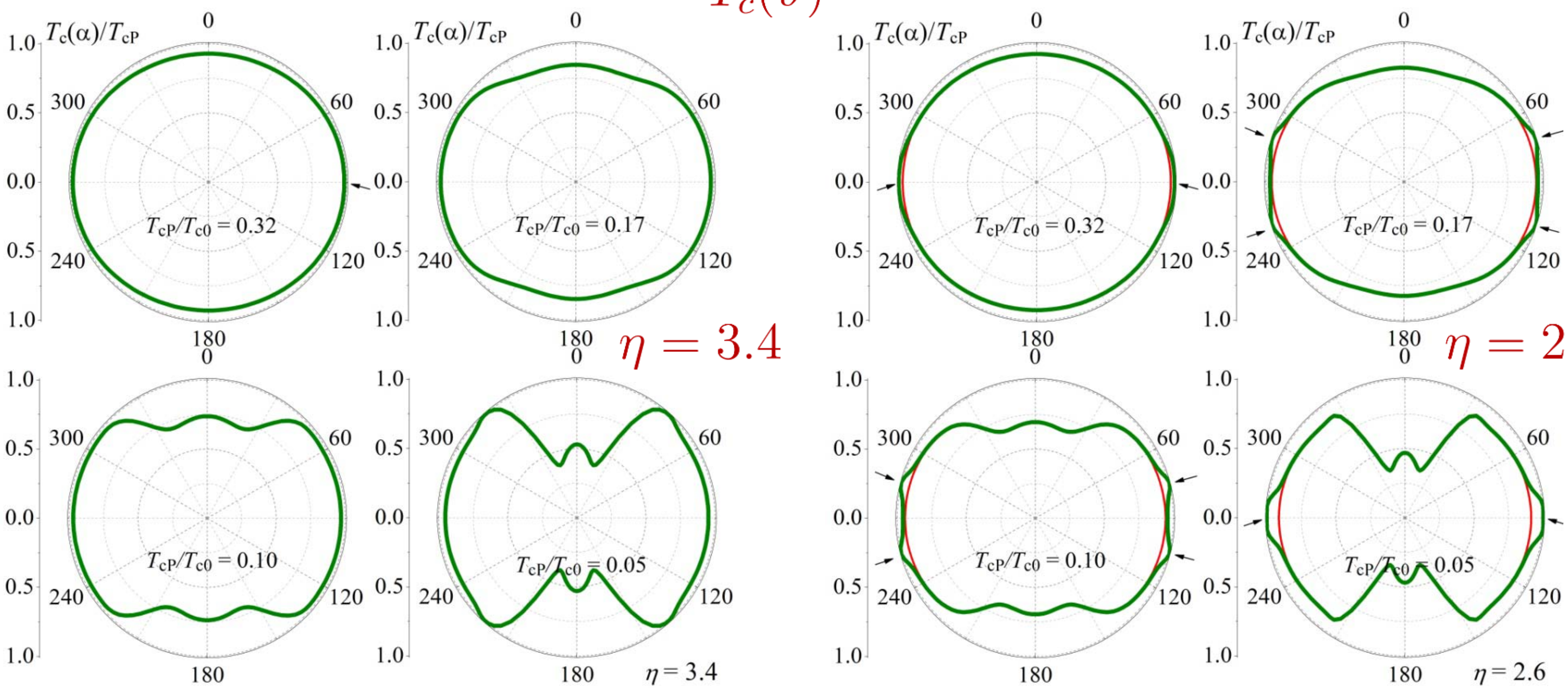
In-plane anisotropy of the onset of superconductivity

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 **q** for several values of T_{cP}/T_{c0}.

Non-resonance case

$T_c(\vartheta)$

Resonance case



$\eta = 3.4$

$\eta = 2.6$

$v_F = 1.0 \times 10^5 m/s$

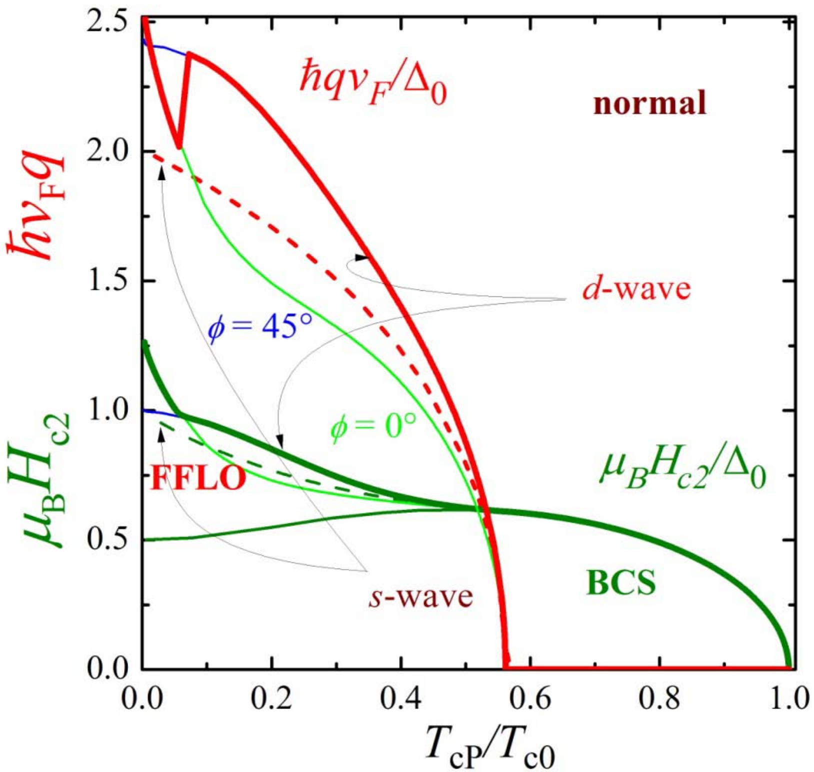
$v_F = 0.75 \times 10^5 m/s$

$t/T_{c0} = 0.2$

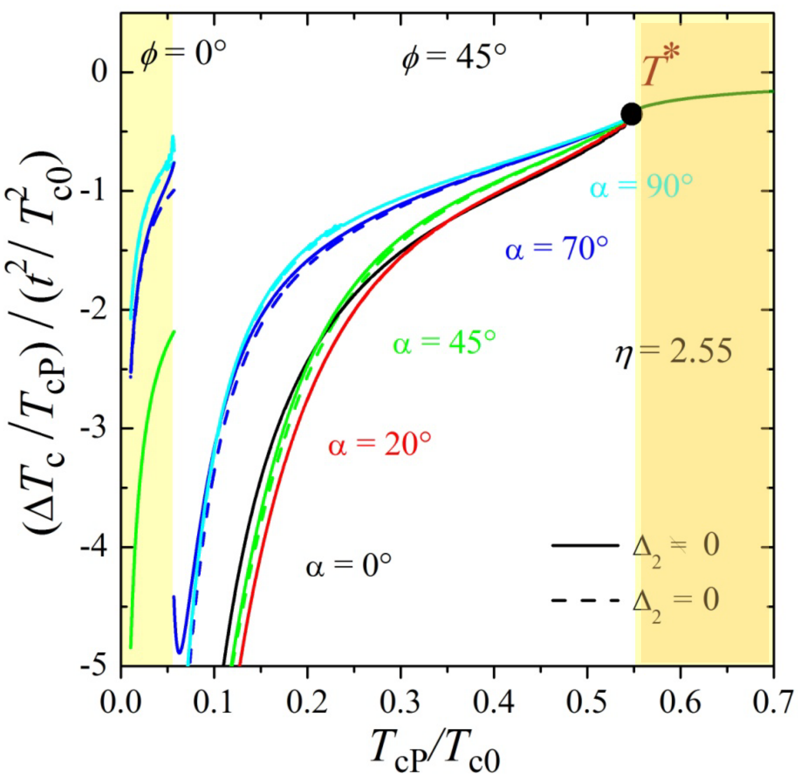
Orbital correction (*d*-wave)

Normalized correction of the superconducting onset temperature as a function of in-plane magnetic field **H** for several angles between magnetic field and the FFLO modulation vector **q**.

FFLO modulation vector



Orbital correction

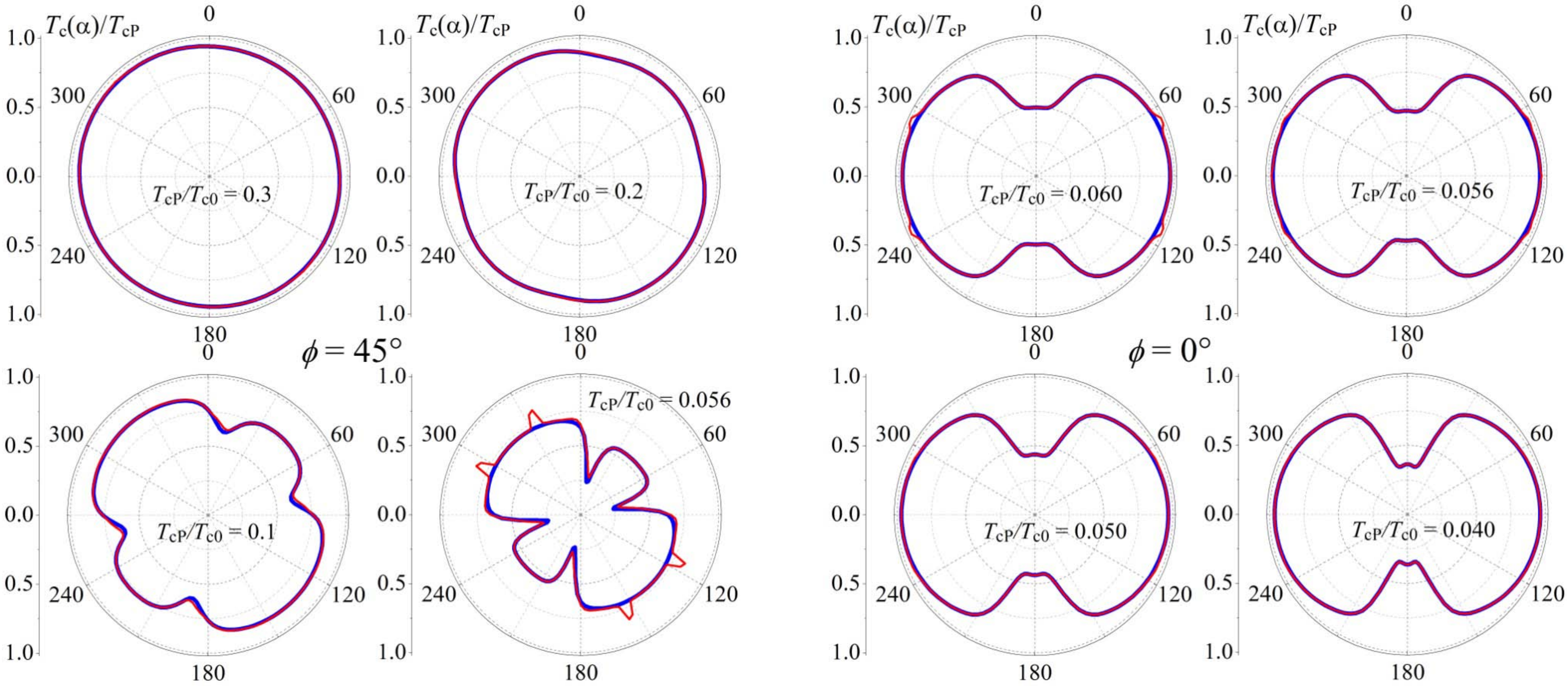


$$t/T_{c0} = 0.2$$

In-plane anisotropy of the onset of superconductivity (*d*-wave)

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 **q** for several values of T_{cP}/T_{c0}.

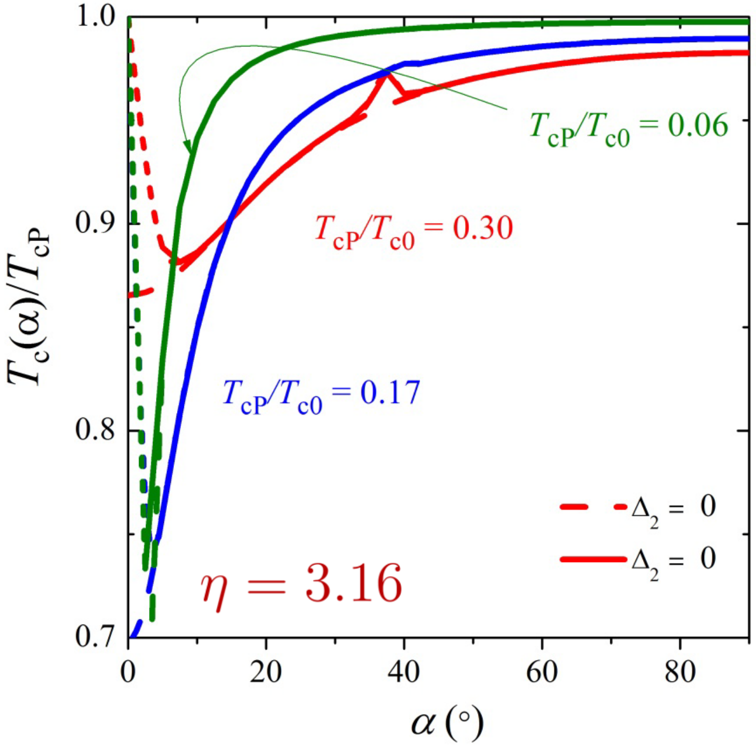
$\phi = 45°$ $T_c(\vartheta)$ $\phi = 0°$

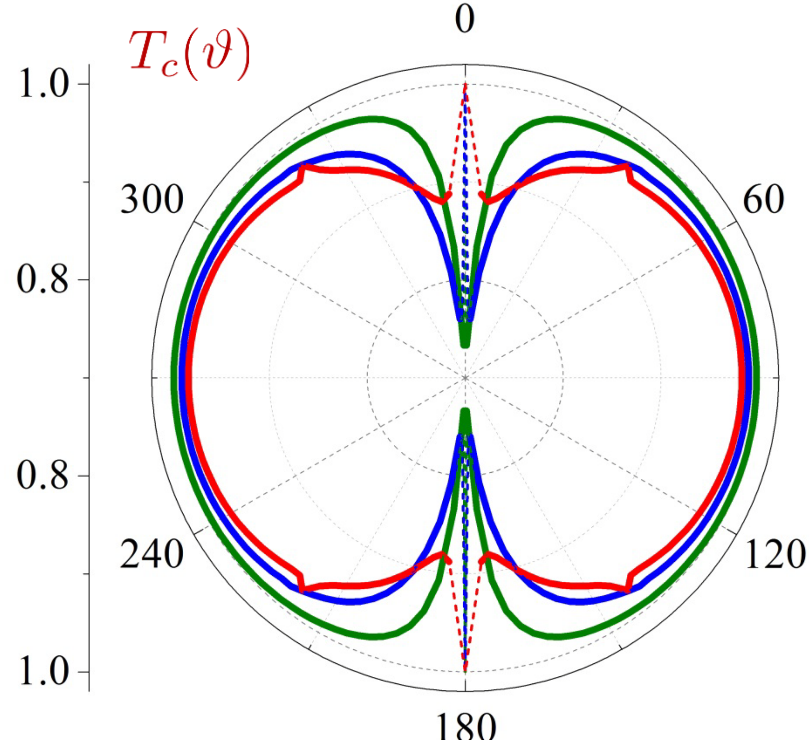


$t/T_{c0} = 0.2$

Anisotropy of the onset of superconductivity (quasi-1D)

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 **q** for several values of T_{cP}/T_{c0}.



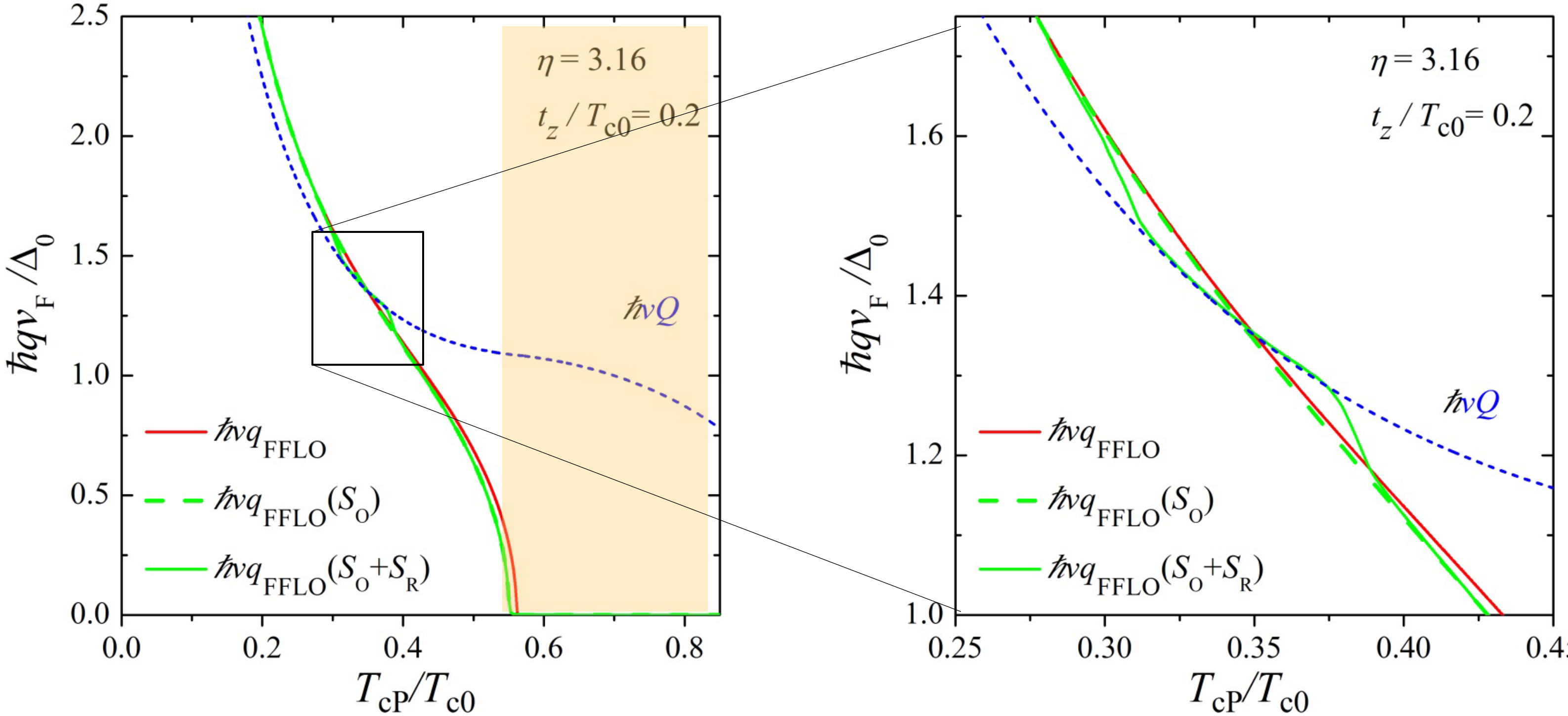


$t/T_{c0} = 0.25$

M. D. C. and A. I. Buzdin, Phys. Rev. B 89, 224506 (2014).

FFLO modulation vector *q* vs resonance

Influence of the orbital contribution on the absolute value of the FFLO modulation wave vector **q**:



$t/T_{c0} = 0.2$

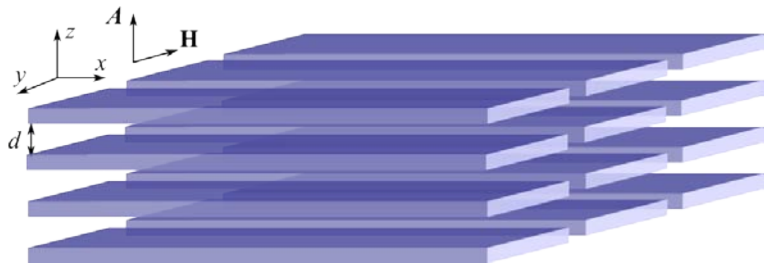
M. D. C. and A. I. Buzdin, Phys. Rev. B 89, 224506 (2014).

FFLO lock-in: a Frenkel-Kontorova study

Model:

$$\mathbf{A} = \mathbf{H} \times \mathbf{r} \quad [\mathbf{r} = (x, 0, 0)]$$

$$E_{\mathbf{p}} = \frac{p_x^2}{2m_x} + 2t_y \cos(p_y d_y) + 2t_z \cos(p_z d_z)$$

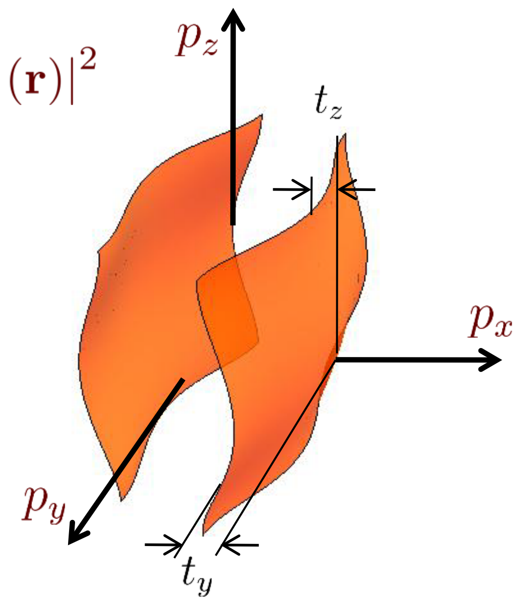


Expansion of the Eilenberger in small $\Delta(\mathbf{r})$

$$F_{sn} = \frac{1}{L_x} \int \mathrm{d}x \alpha \left|\Delta(\mathbf{r})\right|^2 + \beta \left|\partial_x \Delta(\mathbf{r})\right|^2$$

$$+\lambda \left[1 - \cos(2Q_x x)\right] \left|\Delta(\mathbf{r})\right|^2 + \mu Q^2 \left[1 - 7\cos(2Q_x x)\right] \left|\Delta(\mathbf{r})\right|^2$$

$$+\delta \left|\partial_x^2 \Delta(\mathbf{r})\right|^2 + \eta \sin^2(Q_x x) \left|\partial_x \Delta(\mathbf{r})\right|^2$$



At the transition line the solution is
$\Delta(\mathbf{r}) = \Delta_0 \cos[qx]$

We seek the solution in the form:

$$\Delta(\mathbf{r}) = \Delta_0 \cos[Qx + \varphi(x)]$$

with $\varphi'(x) \ll Q$

FFLO lock-in: a Frenkel-Kontorova study

In the vicinity of the resonance: $q \approx Q$

$$\langle F_{sn}^{\varphi} \rangle_Q = \frac{1}{L_x} \int \mathrm{d}x \frac{1}{2} \alpha \Delta_0^2 - \frac{1}{16} v_F^4 Q^2 \left(\varphi' - \delta q\right)^2 - \frac{5}{16} t^2 v_F^2 Q^2 + \frac{11}{32} t^2 v_F^2 Q^2 \cos\left[2\varphi\left(x\right)\right]$$

where: $\delta q \equiv Q - q$

A continuum limit approximation of the Frenkel-Kontorova Hamiltonian introduced by Frank and Van der Merwe leads to the sine-Gordon equation for $\varphi\left(x\right)$

$$\frac{\partial^2 \varphi}{\partial x^2} + v \sin\left[2\varphi\left(x\right)\right] = 0$$

$$v = 0 \longrightarrow \varphi\left(x\right) = Cx$$

$$v = \frac{t^2}{v_F^2} \longrightarrow \varphi\left(x\right) = \mathrm{am}\left[\frac{\sqrt{2v}}{\kappa} x, \kappa\right]$$

The normalized energy of the state with the spatial distribution of the order parameter:

$$\langle F_{sn}^{\varphi} \rangle = \frac{\delta q}{64} \frac{\pi \sqrt{2v}}{\kappa K\left(\kappa^2\right)} - \frac{v}{64}\left[1 - \frac{2}{\kappa^2} + \frac{4}{\kappa^2} \frac{E\left(\kappa^2\right)}{K\left(\kappa^2\right)}\right]$$

FFLO lock-in: a Frenkel-Kontorova study

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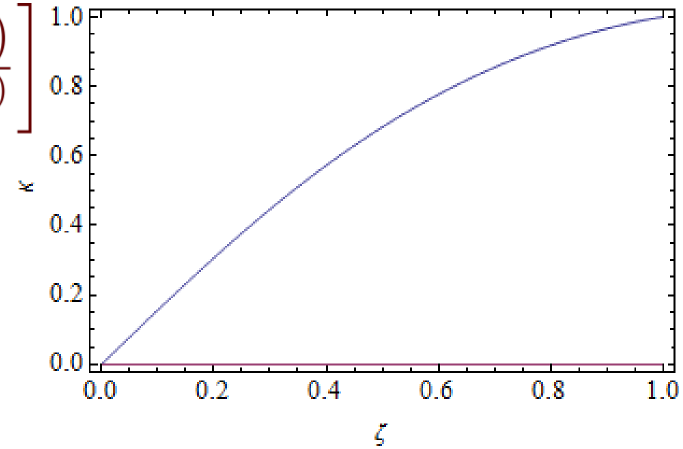
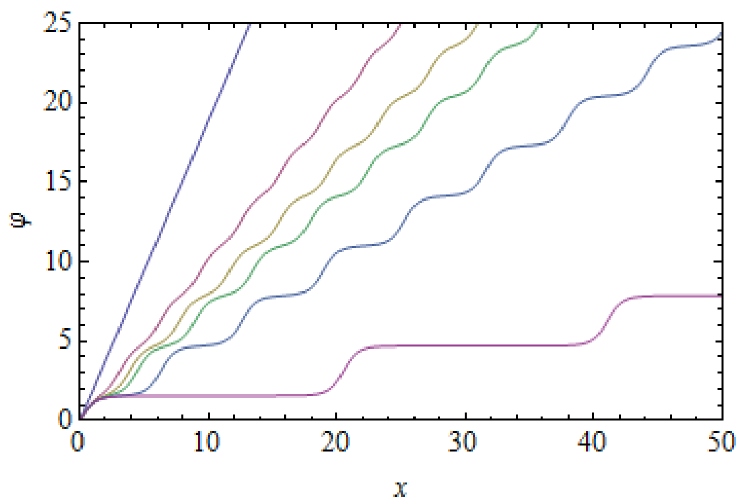
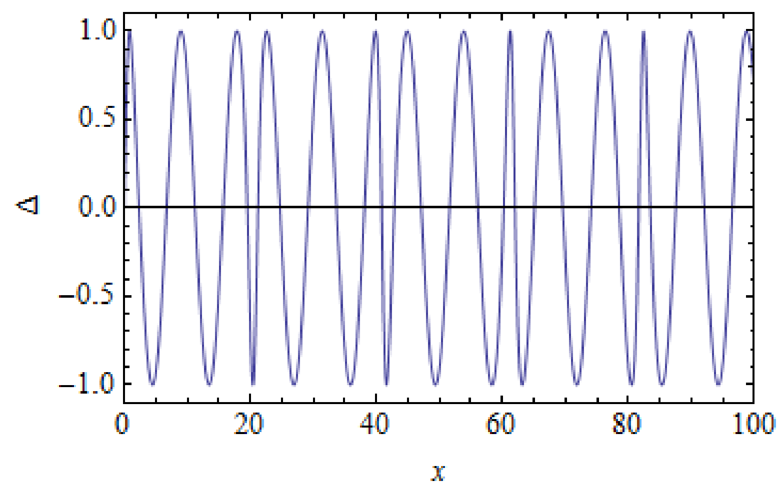
$$\langle F_{sn}^{\varphi} \rangle = \frac{\delta q}{64} \frac{\pi \sqrt{2v}}{\kappa K(\kappa^2)} - \frac{v}{64} \left[1 - \frac{2}{\kappa^2} + \frac{4}{\kappa^2} \frac{E(\kappa^2)}{K(\kappa^2)} \right]$$



Minimization of the energy:

$$\frac{\kappa}{E(\kappa^2)} = \sqrt{\frac{v}{v_c}} \qquad v_c = \pi^2 \delta q^2 / 8$$





M. Croitoru and A. Buzdin, PRB, Dec. 2016

Conclusions

- **FFLO modulation** strongly **interferes** with the **orbital effect** and provides the main source of the critical field anisotropy in quasi-LD superconductors.
- 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.
- As soon as the **vector potential** of the applied magnetic field is **commensurate** with the **wave vector of the FFLO phase** the **resonance** peaks appear in the field-direction dependence of the onset of superconductivity.
- At the r**esonance** the interplay between the orbital and paramagnetic effects may result in a **lock-in effect**.
- These effects can open up a new possibility to unambiguously evidence spatially modulated superconducting phase in quasi-LD conductors.

Thank you for attention!