

Larkin-Ovchinnikov-Fulde-Ferrell state in quasi-one-dimensional superconductors

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The properties of a quasi-one-dimensional (quasi-1D) superconductor with an *open Fermi surface* are expected to be unusual in a magnetic field. On the one hand, the quasi-1D structure of the Fermi surface strongly favors the formation of a nonuniform state [Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) state] in the presence of a magnetic field acting on the electron spins. On the other hand, a magnetic field acting on an open Fermi surface induces a dimensional crossover by confining the electronic wave functions along the chains of highest conductivity, which results in a divergence of the orbital critical field and in a stabilization at low temperature of a cascade of superconducting phases separated by first-order transitions. In this paper, we study the phase diagram as a function of the anisotropy by taking into account on the same footing the paramagnetic and the orbital effects of the field. We discuss in detail the experimental situation in the quasi-1D organic conductors of the Bechgaard salts family and argue that they appear as good candidates for the observation of the LOFF state, provided that their anisotropy is large enough. Recent experiments on the organic quasi-1D superconductor $(\text{TMTSF})_2\text{ClO}_4$ are in agreement with the results obtained in this paper and could be interpreted as a signature of a high-field superconducting phase. We also point out the possibility to observe a LOFF state in some of the recently discovered quasi-2D organic superconductors due to the particular topology of their Fermi surface.

I. INTRODUCTION

In 1963, Larkin and Ovchinnikov, and independently Fulde and Ferrell, predicted the existence of a nonuniform superconducting state [hereafter referred to as the Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) state] in the presence of a magnetic field acting on the electron spins.¹ These authors noted that the destructive influence of Pauli paramagnetism on superconductivity can be mitigated by pairing spin-up and spin-down electrons with a nonzero total momentum whose value depends on the magnetic field. In this way, the pairing condition, which requires that opposite spin electrons with equal energy and a given total momentum should be paired, can be fulfilled with improved accuracy over some parts of the Fermi surface. On other parts of the Fermi surface, it may then not be possible to pair electrons at all, but the LOFF state can nonetheless be more stable than the uniform solution. This superconducting state occurs only at temperatures smaller than $T_0 \simeq 0.56 T_{c0}$ where T_{c0} is the zero-field superconducting transition temperature. The phase transition is of first order from the LOFF state to the ordinary uniform superconducting state and of second order to the normal metallic phase.

Although this nonuniform state was predicted many years ago, there has been up to now no unequivocal experimental evidence of its existence.² This can be explained by several reasons. For an isotropic dispersion law, the LOFF state leads only to a small increase of the zero-temperature critical field as given by the Shandrasekhar-Clogston (or Pauli) limit,³ and its region of existence in the H - T plane, although not known exactly, is very narrow.¹ Moreover, when orbital effects of the field are considered in a type-II superconductor, the

LOFF state can only exist if the diamagnetic effect is weak enough compared to the paramagnetic effect. The precise criterion obtained by Gruenberg and Gunther⁴ for clean superconductors with an isotropic dispersion law is $H_{c2}^{\text{orb}}(0)/H_P > 1.28$, where $H_{c2}^{\text{orb}}(0)$ and H_P are the zero-temperature orbital critical field and the Pauli limited field, respectively. Finally, the LOFF state is very sensitive to impurities and is destroyed when the elastic mean free path becomes smaller than the coherence length.⁵

Quasi-one-dimensional (quasi-1D) superconductors (weakly coupled chains systems with an *open Fermi surface* consisting in two quasi-1D sheets) appear very particular with respect to the existence of a LOFF state. The fundamental reason is that, because of the quasi-1D structure of the Fermi surface, the partial compensation of the Pauli pair-breaking (PPB) effect by a spatial modulation of the order parameter is much more efficient than in a system with an isotropic dispersion law.^{6,8,9} Moreover, it has been shown recently that the magnetic field induces a dimensional crossover which makes the orbital critical field H_{c2}^{orb} diverge,⁷⁻⁹ thus increasing the relative strength of the PPB effect compared to the orbital effect. Noting also that quasi-1D systems such as can be found experimentally in the organic conductors of the Bechgaard salts family can be made very clean, strongly anisotropic superconductors should be very good candidates for the observation of a LOFF state.

The effect of a magnetic field on the phase diagram of a quasi-1D superconductor has recently received a lot of attention.⁷⁻¹⁰ It was shown that for $T_{c0} \ll t_z$ a high magnetic field stabilizes at low temperature a cascade of superconducting phases separated by first-order transitions, which ends in a strong reentrance of the superconducting phase (the magnetic field is parallel to the y axis;

t_z is the coupling in the z direction between the chains parallel to the x axis). The existence of this cascade of superconducting phases in high magnetic field is a consequence of the two properties of a quasi-1D superconductor noted above: the magnetic-field-induced dimensional crossover which freezes the orbital mechanism of destruction of the superconductivity and the efficiency of the LOFF state in compensating the PPB effect.⁷⁻⁹ In the reentrant phase, the dimensional crossover is almost complete: The system exhibits a quasi-2D behavior and the critical temperature is mainly determined by the PPB effect. For small enough t_z ($t_z \sim T_{c0}$), the cascade of phase transition disappears and the reentrant phase appears more as a slow decrease of the critical temperature than as a real reentrance of the superconducting phase. The critical temperature T_c then decreases as $\sim 1/H$, a consequence of the existence of a LOFF state, leading to an upward curvature of the critical field $H_{c2}(T)$.⁹

In this paper, we determine the transition line $T_c(H)$ [or $H_{c2}(T)$] as a function of the parameter t_z/T_{c0} in the presence of both orbital and PPB effects. The present work is an extension of the work of Dupuis and Montambaux⁹ (DM) with special attention devoted to the appearance of a LOFF state and to the crossover between the low-field regime and the high-field regime (or quantum regime in the terms used in Ref. 9).

In the next section, we determine the transition line in the absence of orbital effects of the field. At low field, $T_c - T_{c0} \sim (\mu_B H)^2/T_{c0}$ which leads to a downward curvature of the upper critical field. We show that below $T_0 \simeq 0.56 T_{c0}$, the LOFF state is more stable than the uniform superconducting state. For $\mu_B H \gg T$, the critical temperature between the LOFF state and the normal state varies as $1/H$, leading to an upward curvature of the critical field $H_{c2}(T)$. The effect of disorder is discussed. In Sec. III, we study the effect of a small coupling $t_z \sim T_{c0}$ between chains on the phase diagram obtained in Sec. II. For a large anisotropy (i.e., $t_z/T_{c0} \sim 1$), the phase diagram obtained in Sec. II is only slightly modified by the orbital effects. At low field, the critical temperature T_c is now dominated by the orbital effects of the field and decreases linearly with H . However, the upward curvature of $H_{c2}(T)$ at $\mu_B H \gg T$, which results from the existence of a LOFF state, subsists when the quantum effects of the field are fully taken into account in the calculation of T_c . For a smaller anisotropy (i.e., $t_z/T_{c0} > 2$), the phase diagram becomes more complicated due to the stabilization at low temperature of the cascade of superconducting phases studied by DM. The interplay between this cascade, which is induced by the orbital effects of the field, and the appearance of the LOFF state is studied in detail as a function of the anisotropy t_z/T_{c0} . In Sec. IV, we discuss the experimental situation in the quasi-1D superconductors of the Bechgaard salts family and argue that they appear as good candidates for the observation of a LOFF state, provided that their anisotropy is large enough. Recent experimental results obtained by Lee *et al.*¹¹ with the organic quasi-1D superconductor (TMTSF)₂ClO₄ (TMTSF=tetramethyltetraselenafulvalene) are discussed. In the Conclusion, we point out the possibility to

observe a LOFF state in some of the recently discovered quasi-2D organic superconductors due to the particular topology of their Fermi surface.

II. PAULI PARAMAGNETISM

In this section, we consider a strongly anisotropic superconductor subject to a magnetic field acting on the electron spins. The open Fermi surface is described by the dispersion law ($\hbar = k_B = 1$ in the following and the Fermi energy is chosen as the origin of the energies)

$$\epsilon_{\mathbf{k}}^{\alpha} = v(\alpha k_x - k_F) + t_z \cos(k_z c), \quad (1)$$

where v is the Fermi velocity for the motion along the chains (x axis) and c the distance between chains. $\alpha = \text{sgn}(k_x) = +/-$ labels the right/left sheet of the Fermi surface. We do not consider explicitly the y direction parallel to the magnetic field which does not play any role for a linearized dispersion law.

At high temperature (or low magnetic field), the order parameter Δ is uniform. Its value is obtained from the self-consistency equation

$$\frac{1}{\lambda} = \frac{T}{S} \sum_{\mathbf{k}, \omega_n} \frac{1}{\epsilon_{\mathbf{k}}^{\alpha 2} - (i\omega_n + h)^2 + \Delta^2}, \quad (2)$$

where $\lambda > 0$ denotes the BCS attractive interaction and $h = \mu_B H$ is the Zeeman energy (the g factor is assumed to be equal to 2). $\omega_n = \pi T(2n + 1)$ is a Matsubara frequency and S is the area of the system. The difference $F(T, H)$ between the free energies of the superconducting state and of the normal state can be obtained from¹²

$$F(T, H) = \int_0^{\Delta} \frac{dg}{d\Delta'} \Delta'^2 d\Delta', \quad (3)$$

where the function $g(\Delta) = 1/\lambda$ is defined by (2). Expanding the self-consistency equation in powers of Δ , we obtain the Ginzburg-Landau (GL) expansion of the free energy,

$$F(T, H) = A\Delta^2 + \frac{B}{2}\Delta^4 + \frac{C}{3}\Delta^6, \quad (4)$$

where

$$\begin{aligned} A &= \lambda^{-1} - \chi(0), \\ B &= \frac{N(0)}{2} \frac{1}{(2\pi T)^2} \text{Re} \zeta \left(3, \frac{1}{2} + \frac{h}{2i\pi T} \right), \\ C &= -\frac{3N(0)}{8} \frac{1}{(2\pi T)^4} \text{Re} \zeta \left(5, \frac{1}{2} + \frac{h}{2i\pi T} \right), \end{aligned} \quad (5)$$

where $\zeta(a, z) = \sum_{n=0}^{\infty} (n+z)^{-a}$ is the generalized zeta function. $N(0)$ is the density of states per spin at the Fermi level. $\chi(0)$ is the Cooper pair susceptibility,

$$\begin{aligned} \chi(q) &= \frac{T}{S} \sum_{\mathbf{k}, \omega_n} \frac{1}{(i\omega_n - \epsilon_{\mathbf{k}}^{\alpha} - h)(-i\omega_n - \epsilon_{q-\mathbf{k}}^{-\alpha} + h)} \\ &= N(0) \left[\ln \left(\frac{2\gamma\Omega}{\pi T} \right) + \Psi \left(\frac{1}{2} \right) \right. \\ &\quad \left. - \frac{1}{2} \sum_{\alpha} \text{Re} \Psi \left(\frac{1}{2} + \frac{\alpha v q + 2h}{4i\pi T} \right) \right], \end{aligned} \quad (6)$$

evaluated at zero total momentum, and Ψ is the digamma function. Here q is the momentum along the chains. γ is the exponential of the Euler constant and Ω is the cutoff energy for the attractive interaction. The GL expansion (5) agrees with the one obtained by Maki and Tsuneto in the case of an isotropic system with the dispersion law $\epsilon_{\mathbf{k}} = k^2/2m$.¹³ As long as the Cooper pairs are formed with states of opposite momenta, the shape of the Fermi surface does not play any role. This appears clearly when we make the usual replacement $S^{-1} \sum_{\mathbf{k}} \rightarrow N(0) \int d\epsilon$ in the self-consistency equation (2). The critical temperature is determined by $A = \lambda^{-1} - \chi(0) = 0$. For low field $h \ll T$, using $\lambda^{-1} = N(0) \ln(2\gamma\Omega/\pi T_{c0})$, we obtain a downward curvature of the transition line [or equivalently of the critical field $H_{c2}(T)$]:

$$T_{c0} - T_c \simeq \frac{7\zeta(3)}{4\pi^2} \frac{h^2}{T_{c0}}, \quad (7)$$

where $\zeta(3) \simeq 1.20$. As pointed out by Maki and Tsuneto,^{13,14} the transition between the normal state and the uniform superconducting state becomes of first order when the coefficient of the quartic term in the GL expansion changes sign. This corresponds to the point (h_0, T_0) of the transition line determined by

$$\text{Re} \zeta \left(3, \frac{1}{2} + \frac{h_0}{2i\pi T_0} \right) = 0 \text{ and } A = 0. \quad (8)$$

The first equality in (8) leads to $h_0/2\pi T_0 \simeq 0.304\,092$. From $A = 0$ and $\lambda^{-1} = N(0) \ln(2\gamma\Omega/\pi T_{c0})$, we then deduce $T_0 \simeq 0.56 T_{c0}$.

Up to now, we have ignored the possibility to observe a nonuniform superconducting state. It is therefore necessary to consider the more general equation

$$\frac{1}{\lambda} = \chi(q), \quad (9)$$

where $\chi(q)$ is the Cooper pair susceptibility defined in (6). In principle, one should also consider the possibility of a nonuniform superconducting state with a finite momentum of the Cooper pair along the z direction. We have verified that in our model such a state is never stable. The wave vector of the modulation of the order parameter at the transition is determined by the maximum of the susceptibility, which can be obtained from the derivatives

$$\chi'(q) = -N(0) \frac{v}{8\pi T} \sum_{\alpha} \text{Re} \frac{\alpha}{i} \Psi' \left(\frac{1}{2} + \frac{\alpha v q + 2h}{4i\pi T} \right), \quad (10)$$

$$\chi''(q) = -N(0) \left(\frac{v}{4\pi T} \right)^2 \sum_{\alpha} \text{Re} \zeta \left(3, \frac{1}{2} + \frac{\alpha v q + 2h}{4i\pi T} \right), \quad (11)$$

where Ψ' and Ψ'' are the first and second derivatives of the digamma function. The last equation was obtained using $\Psi''(z) = -2\zeta(3, z)$. It can be seen from Eq. (10) that $\chi'(0) = 0$ independently of the value of the field. For $h/T < h_0/T_0$, $\chi''(0) < 0$, so that $q = 0$ corresponds

to a maximum of the susceptibility. For $h/T > h_0/T_0$, $\chi''(0) > 0$, showing that $q = 0$ corresponds to a local minimum of the susceptibility. The maximum of $\chi(q)$ is reached for a finite value of the total momentum. Thus, the temperature T_0 below which the LOFF state is more stable than the uniform superconducting state corresponds exactly to the temperature below which we showed that the transition between the normal state and the uniform superconducting state would have become of first order in the absence of the LOFF state. An analogous result has been obtained by Dieterich and Fulde in their study of the magnetic field dependence of the Peierls instability in one-dimensional conductors, a problem which bears some similarities with the one considered in this section.¹⁵ For h/T slightly above h_0/T_0 , we find using Eq. (10) that the maximum of the susceptibility is obtained for

$$q^2 = 6 \left(\frac{4\pi T}{v} \right)^2 \frac{\text{Re} \Psi'' \left(\frac{1}{2} + \frac{h}{2i\pi T} \right)}{\text{Re} \Psi^{(4)} \left(\frac{1}{2} + \frac{h}{2i\pi T} \right)}, \quad (12)$$

where $\Psi^{(4)}$ is the fourth derivative of the digamma function. For large field $h \gg T$, Eq. (6) shows that the maximum of the susceptibility should be reached for $q \simeq \pm 2h/v$. This leads to the critical temperature

$$T_c \simeq \frac{\pi T_{c0}^2}{4\gamma h}, \quad (13)$$

a result which was previously obtained by DM. Thus, at low temperature, the variation of T_c as $1/H$, which is a consequence of the existence of the LOFF state, leads to a divergence and an upward curvature of the critical field $H_{c2}(T)$. The susceptibility $\chi(q)$ as a function of q is shown in Fig. 1 for different values of the magnetic field. The transition line T_c and the wave vector q of the order parameter are shown in Fig. 2.

The divergence of the critical field is of course not physical. At low temperature, the effect of disorder will become more and more important and will lead to a finite critical field. Following the standard treatment,¹⁶

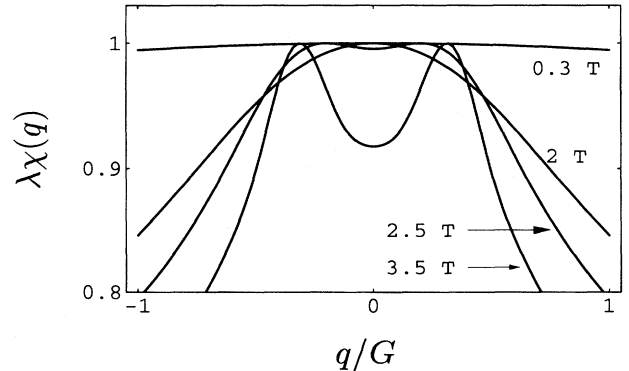


FIG. 1. Cooper pair susceptibility $\chi(q)$ on the transition line (H, T_c) as a function of q/G for different values of the magnetic field. $G = -eHc$ is the magnetic wave vector introduced in Sec. III.

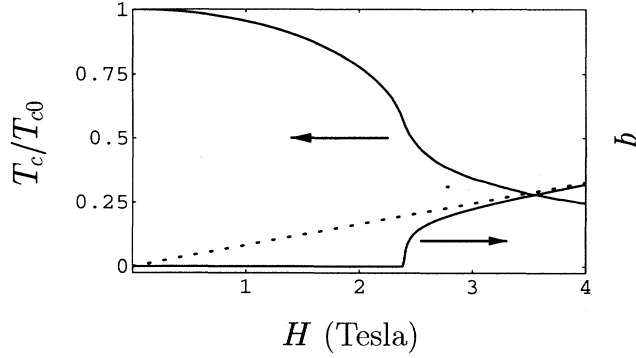


FIG. 2. Transition line T_c and wave vector q of the modulation of the order parameter vs magnetic field without the orbital effect. The dotted line corresponds to $q = 2h/v$.

impurity scattering is taken into account by including self-energy and vertex corrections in the Cooper pair susceptibility. Using the results of Ref. 9, we find that the critical temperature in presence of disorder T_c^{dis} is given by

$$\frac{T_c^{\text{dis}} - T_c}{T_c} \simeq -\frac{\pi}{32T_c\tau} + \frac{3\pi}{32T_c\tau} \frac{T_c^2}{h^2} \quad (14)$$

for $|T_c^{\text{dis}} - T_c| \ll T_c \ll h$. Thus, the disorder becomes important at low temperature when $T_c \sim \pi/32\tau$. In Bechgaard salts where $1/\tau$ can be of the order of 100 mK, the disorder will be inefficient down to very low temperature so that the upward curvature of the upper $H_{c2}(T)$ will persist in a very broad range of temperature.

In the present model, the stability of the LOFF state strongly relies on the use of a linearized dispersion law. It is therefore necessary to verify that a finite curvature of the dispersion law at the Fermi level does not modify significantly the preceding results. Instead of the linearized dispersion law given by Eq. (1), we consider the following tight-binding model:

$$\epsilon_{\mathbf{k}} = t_x \cos(k_x a) + t_y \cos(k_y b) + t_z \cos(k_z c) - \mu, \quad (15)$$

where μ is the Fermi energy and a , b , and c the lattice parameters. The transfer integrals t_x , t_y , and t_z verify the condition $t_y, t_z \ll t_x$ which ensures that the Fermi surface is open for a sufficient filling ($\mu \sim t_x$). If we expand $\epsilon_{\mathbf{k}}$ around $\pm k_F$ defined by $\mu = t_x \cos(k_F a)$, we obtain

$$\begin{aligned} \epsilon_{\mathbf{k}} \simeq v(|k_x| - k_F) + \frac{1}{2}(|k_x| - k_F)^2 a^2 t_x \cos(k_F a) \\ + t_y \cos(k_y b) + t_z \cos(k_z c), \end{aligned} \quad (16)$$

where $v = at_x \sin(k_F a)$ is the Fermi velocity along the chain direction. Because of the curvature of the dispersion law around the Fermi level, it is not possible to find a particular value of q , allowing us to fulfill the pairing condition $\epsilon_{\mathbf{k}} + h = \epsilon_{\mathbf{q}-\mathbf{k}} - h$ for one-half of the phase space. However, it will be possible to neglect the curvature of the dispersion law if

$$\frac{1}{2}q^2 a^2 t_x \cos(k_F a) \ll T, \quad (17)$$

where $q \sim \pm 2h/v$ is the total momentum of the Cooper pair in the LOFF state. Using $k_F a \sim 1$ and $t_x \sim vk_F$, this inequality can be rewritten as

$$h^2 \ll T t_x. \quad (18)$$

For $t_x/2 \sim 3000$ K and using $h = \mu_B H = 0.67H$ K, we obtain the condition $H \ll 30$ T at $T = 100$ mK. Thus, in the magnetic field and temperature ranges which can be experimentally reached, the use of a linearized dispersion law is justified.

III. ORBITAL AND PAULI EFFECTS

In this section, we study how the orbital effects of the magnetic field modify the phase diagram obtained in the preceding section. In the gauge $\mathbf{A}(0, 0, -Hx)$, the order parameter is determined by the integral equation⁷⁻⁹

$$\lambda^{-1} \Delta(x, q_z) = \int_{|x-x'|>d} dx' K(x, x', q_z) \Delta(x', q_z), \quad (19)$$

$$\begin{aligned} K(x, x', q_z) = \frac{N(0)\pi T \cos[2\mu_B H(x-x')/v]}{v \sinh[|x-x'|2\pi T/v]} \\ \times J_0 \left(\frac{4t_x}{\omega_c} \sin \left[\frac{G}{2}(x-x') \right] \right) \\ \times \sin \left[q_z \frac{c}{2} - \frac{G}{2}(x+x') \right], \end{aligned} \quad (20)$$

where K takes into account both the PPB and orbital effects. J_0 is the zeroth-order Bessel function, $G = -eHc$, and $\omega_c = Gv$. The cutoff d is related to the energy Ω . Taking advantage of the conservation of the transverse momenta in the chosen gauge, we have introduced the Fourier transform $\Delta(x, q_z)$ of the order parameter with respect to z . q_z only shifts the origin of the x axis and can therefore be set equal to zero when determining the critical temperature. Without any loss of generality, the solution of the integral equation (19) can be written as a Bloch function⁹

$$\Delta_Q(x) = e^{iQx} \tilde{\Delta}_Q(x), \quad (21)$$

where $\tilde{\Delta}_Q(x)$ has the periodicity π/G and the magnetic Bloch wave vector Q is restricted to $] -G, G[$. Each phase is characterized by this vector Q which plays the role of a pseudomomentum for the Cooper pairs in the magnetic field. The kernel $K(x, x')$ takes into account all the quantum effects of the field. In Sec. III A we will compare the exact mean-field results obtained with $K(x, x')$ with those obtained in the eikonal (or semiclassical phase integral) approximation where the quantum effects of the field are completely neglected. In this approximation, the kernel becomes⁹

$$K^{(\text{eik})}(x, x', q_z) = \frac{N(0)\pi T \cos[2\mu_B H(x - x')/v]}{v \sinh[|x - x'|2\pi T/v]} \times J_0 \left(\frac{2t_z}{v}(x - x') \sin \left[q_z \frac{c}{2} - \frac{G}{2}(x + x') \right] \right). \quad (22)$$

If $t_z = 0$, the orbital effects vanish for a field parallel to the y direction and the phase diagram is then as shown in Fig. 2. In the following, we study the orbital effects of the field for increasing coupling between chains.

A. Large anisotropy

We first consider the case of a small coupling $t_z/T_{c0} = 1.33$. For each value of the field, we determine numerically from Eq. (19) the vector Q which maximizes the critical temperature and the corresponding T_c .

In a first step, we neglect the PPB effect. The results are shown in Fig. 3 for the eikonal approximation, where quantum effects of the field are not taken into account. We recover the standard results for a system of weakly coupled superconducting chains (or planes).¹⁷ Close to T_{c0} , the critical temperature decreases linearly with H . In this regime, the coherence length $\xi_z(T)$ is much larger than the spacing c between chains and the superconducting state is a triangular Abrikosov vortex lattice. At lower temperature, the coherence length becomes of the order of the spacing between chains so that vortices can fit between chains, thus quenching the orbital mechanism of destruction of the superconductivity and leading to a divergence of the critical field. The superconducting state is then a triangular Josephson vortex lattice with a periodicity in the transverse direction equal to $2c$. The crossover between these two regimes is sometimes referred to as a (temperature-induced) dimensional crossover.¹⁸ It should be noted here that this dimensional crossover is different from the magnetic-field-induced dimensional crossover which results from the magnetic-field-induced localization of the wave functions and which

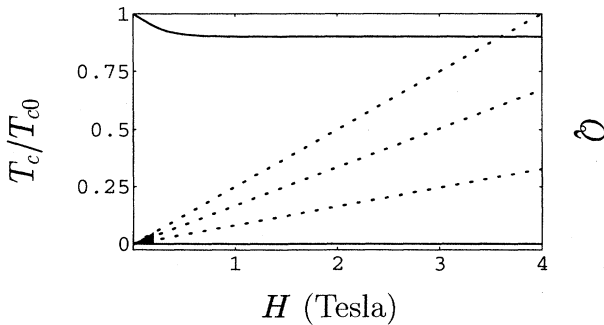


FIG. 3. Transition line T_c and magnetic Bloch wave vector Q for $t_z/T_{c0} = 1.33$ without the PPB effect in the eikonal approximation [Eq. (22)]. The degeneracy of T_c with respect to Q at low field is shown symbolically by a shaded triangle. The three dotted lines correspond to $Q = 2h/v, G - 2h/v, G$.

is not taken into account in the eikonal approximation.⁹ The values of Q corresponding to the highest T_c are shown in Fig. 3. At low field, all the values of Q lead to the same critical temperature. As pointed out by DM, this degeneracy allows one to construct the Abrikosov vortex lattice by taking a linear combination of the function $\Delta_Q(x, q_z)$. In Fig. 3, the degeneracy of T_c with respect to Q is shown symbolically by a shaded triangle. At higher field, when the superconducting state becomes a Josephson vortex lattice, the degeneracy is lifted in favor of $Q = 0$. It is worth pointing out that these results obtained in the eikonal approximation can also be obtained in the Lawrence-Doniach model¹⁹ where the critical temperature is obtained from⁹ (restoring the q_z dependence)

$$-v^2 \frac{\partial^2 \Delta}{\partial x^2} + t_z^2 [1 - \cos(q_z c - 2Gx)] \Delta = \frac{16\pi^2}{7\zeta(3)} T_{c0}^2 \left(1 - \frac{T_c}{T_{c0}}\right). \quad (23)$$

The results obtained in the exact calculation are similar to those obtained in the eikonal approximation, except for the reentrance which occurs at high field ($\omega_c \gg t_z$, which corresponds to $H > 1$ T for the parameters used in Fig. 3) as a consequence of the magnetic-field-induced dimensional crossover.⁹

We now consider both the PPB and orbital effects (Fig. 4). In both descriptions (exact and eikonal), we

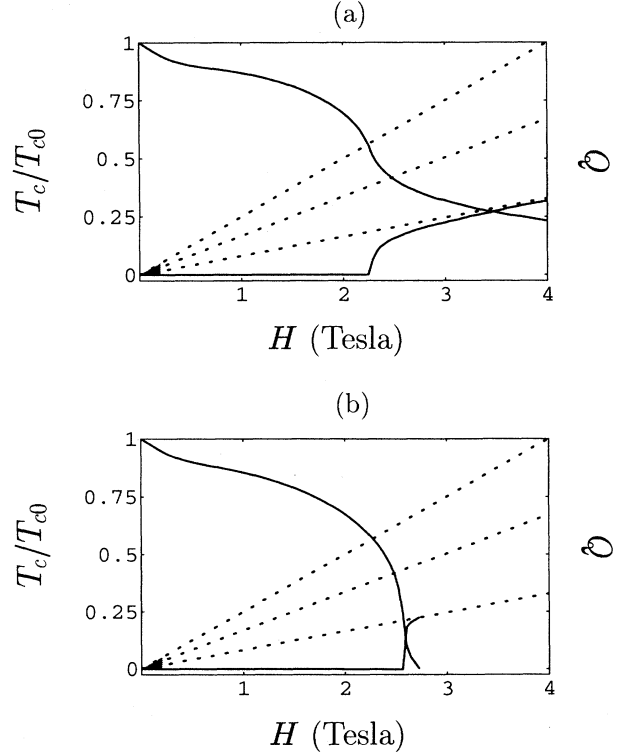


FIG. 4. Transition line T_c and magnetic Bloch wave vector Q for $t_z/T_{c0} = 1.33$ with both the PPB and orbital effects. (a) Exact calculation [Eq. (20)]. (b) Eikonal approximation [Eq. (22)].

obtain a linear behavior at low field showing that the critical temperature is limited by the orbital effect. The value of Q is degenerate in this field range. At higher field, the degeneracy is lifted in favor of $Q = 0$ when the periodicity of the vortex lattice becomes of the order of $2c$. This temperature-induced dimensional crossover from the Abrikosov vortex lattice towards the Josephson vortex lattice is accompanied by a weak upward curvature of $H_{c2}(T)$. At lower temperature, the two descriptions differ considerably. In the exact description [Fig. 4(a)], the orbital effect appears very weak and the phase diagram in this field range is similar to the one obtained by considering only the PPB effect (Fig. 2). We observe a transition to a LOFF state characterized by a finite value of Q which means an additional spatial modulation for the Josephson vortex lattice. For very high field, we find $Q \simeq 2h/v$. The transition line shows a pronounced upward curvature which is a consequence of the existence of the LOFF state. In the eikonal description [Fig. 4(b)], the orbital effect modifies in an important way the phase diagram shown in Fig. 2. The divergence of the critical field H_{c2} is suppressed²⁰ and the region of stability of the LOFF state is very narrow. The upward curvature of the transition line is now restricted to very low temperatures.

Thus, the pronounced upward curvature of $H_{c2}(T)$ found in the preceding section, which was a consequence of the existence of the LOFF state, persists only if the quantum effects of the field are fully taken into account. In the following, we shall not consider the eikonal approximation any more.

B. Smaller anisotropy

For larger values of the coupling between chains $t_z/T_{c0} = 2.67$ and 2.93 , the phase diagrams are shown in Figs. 5 and 6. The low-field regime, where the value of Q is degenerate, is now followed by a phase $Q = G$, which is itself followed by a phase $Q = 0$. This is the cascade of superconducting phases which has been studied by DM. This cascade appears between the low-field regime where the superconducting state is an Abrikosov vortex lattice and the very-high-field regime where the superconducting state is a Josephson vortex lattice. The transition to the LOFF state appears in the last phase $Q = 0$: The GL regime and the cascade of phases are dominated by the orbital effects of the field. Figures 5 and 6 show that the shape of the transition line is very sensitive to the value of t_z/T_{c0} .

If we further increase the coupling between chains (Figs. 7 and 8), the number of phases in the cascade increases. The transition to the LOFF state appears before the reentrant phase. In Fig. 7(b), the transition corresponds to a shift of Q within a phase $Q = G$.

For $t_z/T_{c0} = 6.67$ (Fig. 9), the cascade of phase transitions appears at lower temperature. The transition to the LOFF state occurs in the beginning of the cascade. At low temperature, we thus observe an alternance of phases $Q = 2h/v$ and $Q = G - 2h/v$. In Fig. 10, we have shown the eigenvalue λ_Q of the kernel $K(x, x')$ associated with the eigenfunction $\Delta_Q(x)$ for different values of the

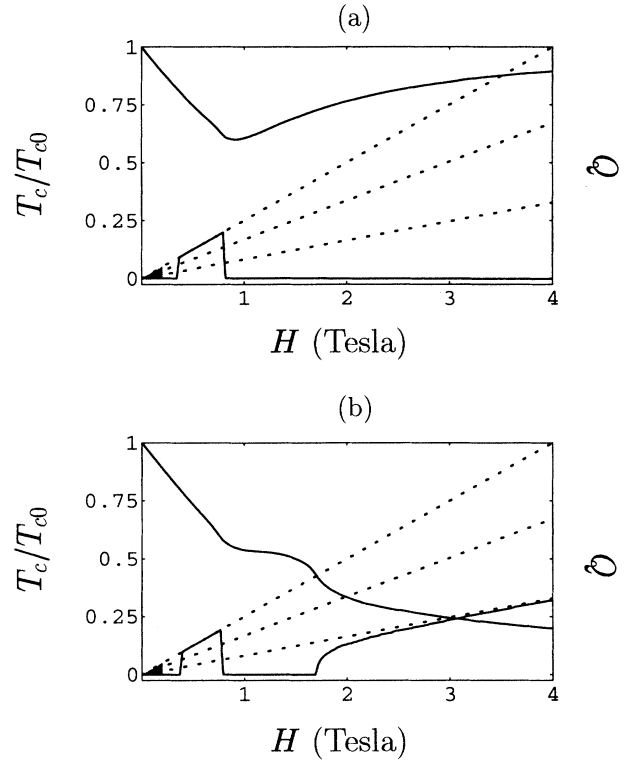


FIG. 5. Transition line T_c and magnetic Bloch wave vector Q for $t_z/T_{c0} = 2.67$ in the exact calculation [Eq. (20)]. (a) Without the PPB effect. (b) With both the PPB and orbital effects.

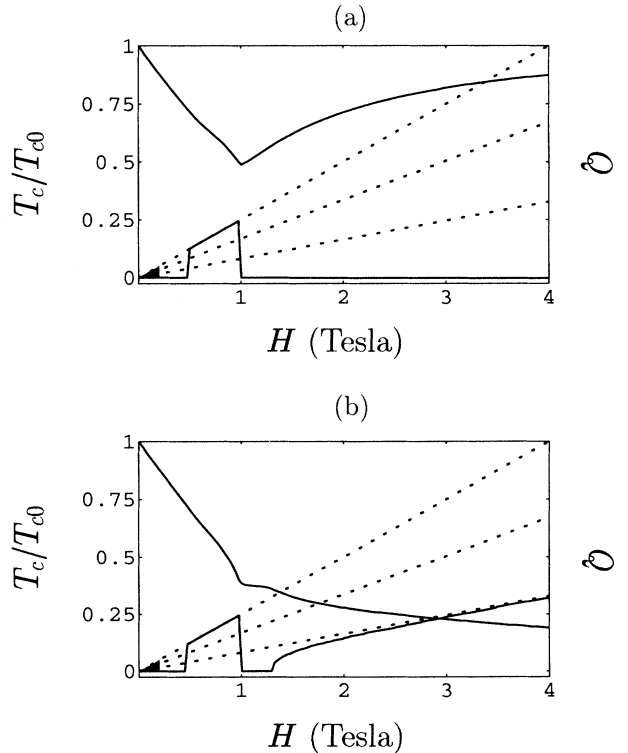
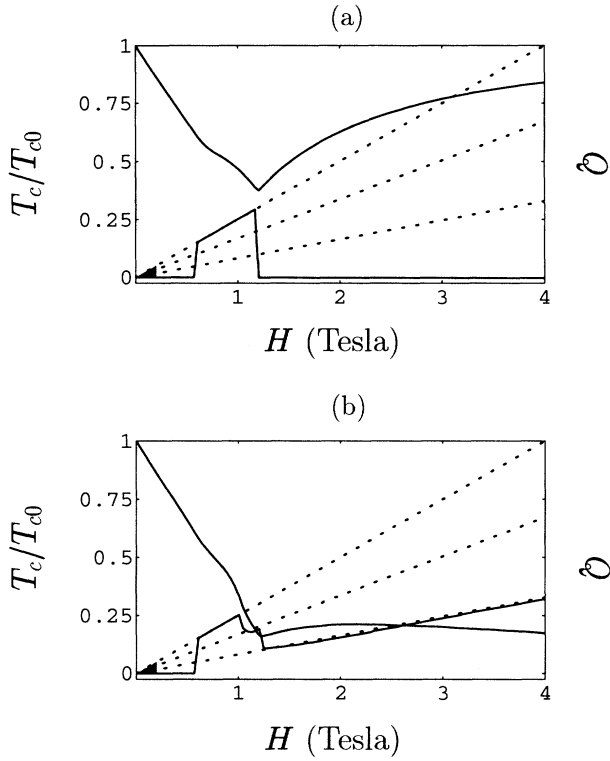
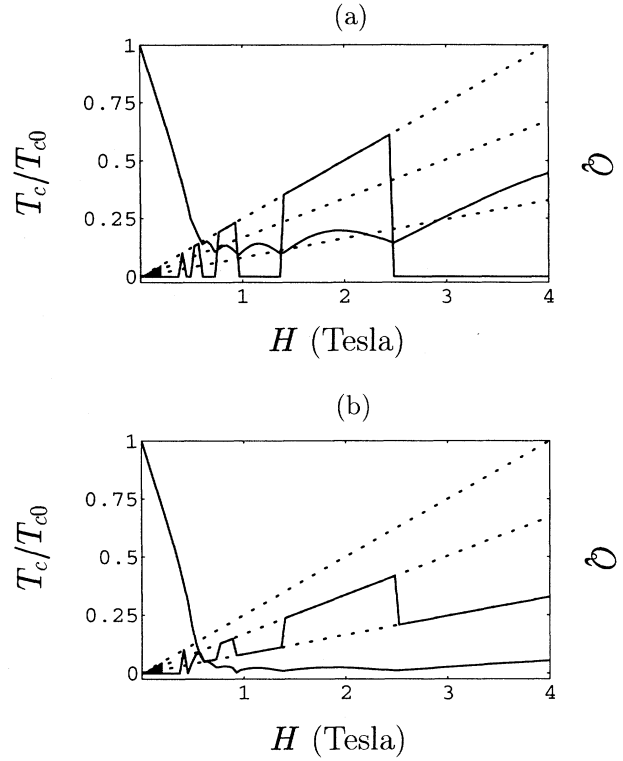
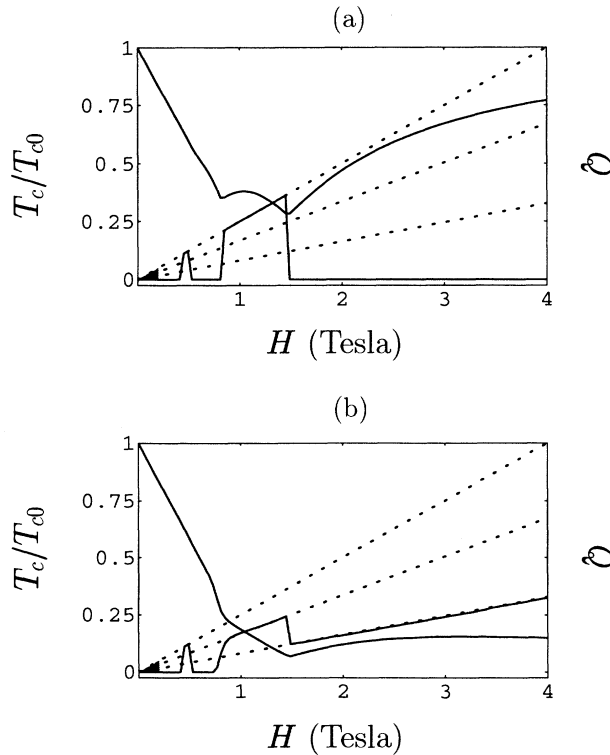


FIG. 6. As in Fig. 5, but for $t_z/T_{c0} = 2.93$.

FIG. 7. As in Fig. 5, but for $t_z/T_{c0} = 3.33$.FIG. 9. As in Fig. 5, but for $t_z/T_{c0} = 6.67$.FIG. 8. As in Fig. 5, but for $t_z/T_{c0} = 4$.

magnetic field. The parameters used in this figure are the same as the ones of Fig. 9.

IV. BECHGAARD SALTS

In this section, we discuss the experimental situation with the Bechgaard salts. We concentrate on $(\text{TMTSF})_2\text{ClO}_4$ which appears as the most promising

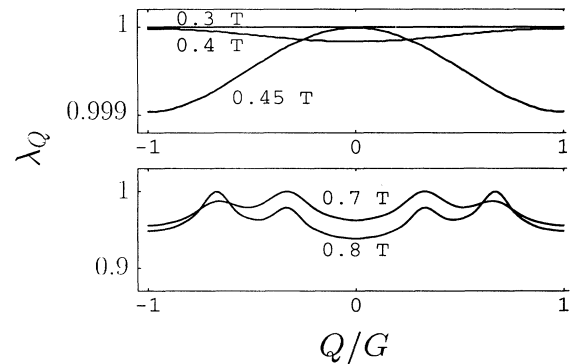


FIG. 10. Eigenvalue λ_Q of the kernel $K(x, x')$ vs Q for different values of the magnetic field. The parameters are the same as in Fig. 9. At low field ($H = 0.3$ T), λ_Q is independent of Q . At higher field ($H = 0.4$ and 0.45 T), this degeneracy is lifted in favor of $Q = 0$ or $Q = G$. At high enough field ($H = 0.7$ and 0.8 T), λ_Q is maximum for either $Q = 2h/v$ or $Q = G - 2h/v$.

material with respect to the effects discussed in this paper. Several comments are in order here. First we should wonder whether the model we have used is adequate to describe the superconductivity in the Bechgaard salts. Many experimental results seem to show that the intrachain interactions are repulsive and indicate that the superconductivity is not of conventional type.^{21,22} For example, NMR measurements obtained by Takigawa *et al.*²³ are clearly not compatible with isotropic *s*-wave pairing in zero field. However, DM argued that a model based on local attractive interactions (which would lead to isotropic *s*-wave pairing in zero field) should remain valid from a qualitative point of view. The reason is that the unusual behavior of a quasi-1D superconductor in a high magnetic field is due only to the magnetic-field-induced dimensional crossover (i.e., the localization of the one-particle wave functions along the chains of highest conductivity) and does not rely on a particular model of superconductivity.²⁴ Second, one should wonder whether a BCS mean-field analysis can be justified in a system of weakly coupled chains. Such an analysis requires well-defined quasiparticles in the normal state and in particular a coherent transverse (in the *y* and *z* directions) electronic motion. From a theoretical point of view, this problem has recently attracted a lot of attention. Many authors²⁵ have given some arguments in favor of a Fermi liquid behavior at low enough temperature in a system of weakly coupled chains²⁶ while the opposite point of view has been adopted by Anderson and collaborators.²⁷ In (TMTSF)₂ClO₄, the NMR relaxation rate²⁸ shows that the behavior of the system changes drastically when the temperature decreases below about 10–30 K. This result, together with the absence of a correlation gap as can be seen from resistivity measurements, strongly suggests that this compound undergoes a single-particle dimensionality crossover at a temperature $T_{x1} \sim 10$ K, below which the transverse electronic motion becomes coherent. Other convincing experiments are those concerned with the angular Lebed' resonances.²⁹ The origin of these resonances, which occur in various physical quantities (thermodynamics or transport) when the field is tilted in the (*y*, *z*) plane, can be simply understood from a semiclassical argument. The semiclassical electronic trajectory is of the form $y = b(t_y/\omega_{cy}) \cos(\omega_{cy}y/v)$ and $z = c(t_z/\omega_{cz}) \cos(\omega_{cz}z/v)$, where $\omega_{cy} = -eHc \cos(\theta)$ and $\omega_{cz} = -eHb \sin(\theta)$. Here *b* is the spacing between chains in the *y* direction and θ is the angle between **H** and the *z* axis. For certain orientations θ of the field ("magic" angles), the two magnetic frequencies ω_{cy} and ω_{cz} are commensurate, $\omega_{cz}/\omega_{cy} = p/q$ (*p*, *q* integer), leading to a periodic electronic motion which results in the Lebed' resonances. Clearly, this analysis based on the consideration of the semiclassical orbits is meaningful only if the electronic motion is coherent in both the *y* and *z* directions. From our point of view, the absence of coherent transverse electronic motion in the Bechgaard salts would therefore be very difficult to reconcile with the existence of these angular oscillations.³⁰

Since the shape of the transition line very strongly depends on the value of the ratio t_z/T_{c0} , one should wonder what the value of this parameter is in the Bechgaard

salts. There are two opinions in the literature concerning the values of the transfer integrals $t_b = t_y/2$ and $t_c = t_z/2$.³¹ According to many authors, $t_b = 200$ – 300 K and $t_c = 5$ – 10 K, as obtained from band calculation.³² These values seem to be supported by recent measurements of a new type of angular oscillations of the conductivity.³³ The second opinion is that the values of t_b and t_c are strongly reduced with respect to their bare values, due to 1D fluctuations.^{22,25} From a renormalization group calculation and using experimental results of the NMR relaxation rate, Bourbonnais *et al.* estimated $t_b \simeq 30$ K,²⁸ which leads to $t_c \simeq 0.5$ – 1.5 K. While the first point of view yields values of the transfer integrals which would make the observation of the LOFF state difficult or even impossible (see the numerical calculations in Sec. III and Ref. 9), the second point of view makes the compound (TMTSF)₂ClO₄ a very good candidate for the effects discussed in this paper.

Lee *et al.* have recently investigated the superconducting transition in (TMTSF)₂ClO₄ from resistive measurements performed between 1.2 K and 60 mK and up to 7 T.¹¹ The magnetic field was oriented along the *b'* axis, since this corresponds to the orientation for which quantum effects of the field are expected.⁹ Although they do not give a definite answer for the existence of high-field superconductivity in (TMTSF)₂ClO₄, these results might be interpreted as the signature of a high-field superconducting phase (see Ref. 11 for a detailed analysis of the experimental results). Such an interpretation would imply an anisotropy $t_z/T_{c0} \sim 3.5$ – 4 which leads to $t_c \sim 2$ K. For this value of the anisotropy, the GL regime and the reentrant phase are separated by a few superconducting phases separated by first-order transitions [see Figs. 7(b) and 8(b)].

V. CONCLUSION

We have presented a detailed analysis of the interplay of the Pauli paramagnetism and the orbital effects of the field in a quasi-1D superconductor with an open Fermi surface. We have calculated the transition line (*H*, *T_c*) as a function of the anisotropy t_z/T_{c0} . As a result of their quasi-1D Fermi surface, quasi-1D superconductors appear as very good candidates for the observation of a LOFF state, provided that their anisotropy is large enough. We have shown that when the interchain coupling is sufficiently weak, the transition between the LOFF state and the normal state is characterized by an upward curvature of the critical field $H_{c2}(T)$. We have also argued that the organic superconductor (TMTSF)₂ClO₄ is a good candidate for the observation of a LOFF state. The experimental results of Lee *et al.*¹¹ on this compound are in agreement with the results obtained in this paper and could be interpreted as a signature of high-field superconductivity. Since the zero-field critical temperature T_{c0} decreases with pressure,³⁴ it should be possible to study the evolution of the resistivity curves $R(H, T)$ measured by Lee *et al.* as a function of the anisotropy t_z/T_{c0} . A disappearance of the high-field "anomalies" of the resistivity when pressure

is increased would support the existence of a high-field superconducting phase at ambient pressure. Although we only considered in this paper the case of quasi-1D systems, the results can easily be extended to the case of quasi-2D conductors. DM argued that similar quantum effects of the field should be present in a weakly coupled-planes system in a magnetic field perpendicular to the low-conductivity axis, leading to a cascade of superconducting phases similar to the one predicted for quasi-1D systems. They also pointed out that for a dispersion law which is isotropic in the highest-conducting plane (for example, $\epsilon_{\mathbf{k}_{\parallel}} = k_{\parallel}^2/2m_{\parallel}$ where \mathbf{k}_{\parallel} and m_{\parallel} are the wave vectors and the effective mass in the highest-conducting plane), the LOFF state will not compensate significantly the PPB effect so that its region of stability in the H - T plane will be very narrow or even nonexistent. This means that both the LOFF state and the cascade will be very difficult to observe in this case. However, some of the recently discovered quasi-2D organic superconductors, like, for example, the salts of the BEDT-TTF

family, present some flat parts on their Fermi surface.³⁵ While such a topology of the Fermi surface is usually expected to favor the formation of a spin-density-wave state, it could also increase the efficiency of the LOFF state in compensating the PPB effect. Experimental results indicate that the critical field parallel to the highest-conducting planes is closed to the Pauli limit.³⁵ Thus, the existence of a LOFF state in quasi-2D materials cannot be *a priori* excluded.

Note added. After completion of this work, the author became aware of a related work on the LOFF state in quasi-2D superconductors.³⁶

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