

Quasi-One-Dimensional Superconductors in Strong Magnetic Field

N. Dupuis,⁽¹⁾ G. Montambaux,⁽¹⁾ and C. A. R. Sá de Melo⁽²⁾

⁽¹⁾*Laboratoire de Physique des Solides, Université Paris-Sud, 91405 Orsay, France*

⁽²⁾*Materials Science Division, Science and Technology Center for Superconductivity, Argonne National Laboratory, Argonne, Illinois 60439*

(Received 22 October 1992)

We determine the phase diagram of a quasi-one-dimensional superconductor (weakly coupled chains system with an *open Fermi surface*) in a magnetic field. The usual Ginzburg-Landau regime is followed, when the field is increased, by a *cascade* of superconducting phases separated by first-order transitions, which ends in a strong reentrance of the superconducting phase. These new phases show a novel kind of symmetry of a laminar type. The Zeeman splitting does not completely suppress the reentrance in very strong field, the ground state being in this case a Larkin-Ovchinnikov-Fulde-Ferrell state.

PACS numbers: 74.20.-z, 74.70.Kn

According to the conventional view, superconductivity and magnetic field are incompatible. The fundamental reason is that in an external magnetic field the order parameter becomes frustrated. This orbital frustration raises the free energy of the superconducting state leading ultimately, as the field is increased, to a transition back to the normal state. The equilibrium state of type II superconductors was first described by Abrikosov using a phenomenological Ginzburg-Landau (GL) theory [1], which was later justified by Gor'kov in a microscopic model [2]. The Ginzburg-Landau-Abrikosov-Gor'kov (GLAG) theory treats the magnetic field in the semiclassical phase integral (also called eikonal) approximation. It neglects the quantum effects of the magnetic field and is valid only when $\omega_c \ll T, 1/\tau$ (ω_c is the characteristic magnetic energy and τ the elastic scattering time). In sufficiently clean materials, it is expected to break down at low temperature. The exact determination of $H_{c2}(T)$ then requires an exact treatment of the magnetic field.

The influence of Landau level quantization in isotropic superconductors, first investigated many years ago [3], has recently received a lot of attention. It has been proposed by Rasolt and Tešanović that superconductivity can exist in very strong magnetic field [4]. When only one Landau level is occupied, the supercurrents can be made to coincide with the orbital motion of the electrons in this Landau level if the periodicity of the vortex lattice is approximately equal to the orbit radius of the lowest Landau level. In this case, the orbital frustration vanishes and superconductivity is only limited by impurity scattering and the Pauli pair breaking effect.

It is well known that 3D strongly anisotropic conductors (i.e., weakly coupled chains systems) such as can be found experimentally in the organic conductors of the Bechgaard salt family show unusual properties in a magnetic field because of their quasi-1D *open Fermi surface* [5]. In these conductors, the semiclassical orbits in the presence of the field are open. Consequently, there is no Landau level quantization but the field induces a 3D/2D crossover [6,7]. Therefore, an interesting question is to

know if there is also in this case a mechanism which will naturally suppress the orbital frustration, leading to the possible existence of superconductivity in strong magnetic field. A first answer was given a few years ago by Lebed' [8], who has shown that the superconducting phase is always stable at low temperature and exhibits a strong reentrance in high magnetic field for equal spin triplet pairing. This reentrance can be simply understood with the following argument. Consider a strongly anisotropic superconductor described by the dispersion law ($\hbar = k_B = 1$ throughout the paper)

$$E_{\mathbf{k}} = v|k_x| + t_y \cos(k_y b) + t_z \cos(k_z c). \quad (1)$$

v is the Fermi velocity for the motion along the chain and t_y, t_z are the coupling between chains separated by the distances b, c . For a magnetic field along the y axis, the semiclassical electronic trajectories obtained from the equation of motion $d\mathbf{k}/dt = e\mathbf{v} \times \mathbf{H}$ are of the form $z = c(t_z/\omega_c) \cos(Gx)$, where $G = -eHc$ and $\omega_c = Gv$. The field localizes the electronic motion in the z direction. In very strong field ($\omega_c \gg t_z$), the amplitude of the trajectories becomes smaller than the distance between chains, showing that the electronic motion becomes localized in the (x, y) plane. The magnetic field being parallel to the plane of the electronic motion, the orbital frustration vanishes [there is no magnetic flux inside the 2D Cooper pairs located in the (x, y) plane]. In Bechgaard salts, the small value of $t_z \sim 20$ K allows one to reach the very strong field limit ($\omega_c \gg t_z$) for reasonable values of the field ($H \sim 20$ T).

In this Letter, we first determine the transition line $T_c(H)$ of a singlet or triplet quasi-1D superconductor. Our calculation goes beyond the semiclassical phase integral approximation and takes into account the quantum effects of the magnetic field. We show that the phase diagram originally proposed by Lebed' [8] is not complete: the GL regime is followed, when increasing the field, by a *cascade of superconducting phases* separated by first-order transitions, which ends with a strong reentrance of the superconducting phase. A description of

the ordered phase is given for temperatures slightly below $T_c(H)$: in strong field, the vortex lattice is replaced by a new superconducting state with a laminar structure. Moreover, it is shown that the reentrance at high field for a singlet superconductor is not completely destroyed by Zeeman splitting, the ground state being in this case a Larkin-Ovchinnikov-Fulde-Ferrell (LOFF) state. Finally, we discuss the application of our calculations to some organic conductors.

We consider a type II quasi-1D superconductor with the dispersion law $E_{\mathbf{k}}$ and the critical temperature $T_c \ll t_z \ll t_y$. This latter condition ensures that the smallest coherence length in the system is always much larger than the spacing between chains [$\xi_z(T) > \xi_z(0) \gg c$]. When quantum effects of the field are not taken into account, the discrete aspect of the system does not play an important role: the superconductivity is well described by the anisotropic GLAG theory. The superconducting state is an anisotropic vortex lattice, the normal state being restored at a field $H_{c2}(T) = \phi_0/2\pi\xi_x(T)\xi_z(T)$ (ϕ_0 is the flux quantum) [9].

In order to take into account the quantum effects of the magnetic field, we start from the Hamiltonian $\mathcal{H}_0 + \mathcal{H}_{\text{int}}$, where $\mathcal{H}_0 = E(\mathbf{k} \rightarrow -i\nabla - e\mathbf{A})$ is the standard Hamiltonian of a noninteracting quasi-1D system obtained by the

Peierls substitution [6]. \mathbf{A} is the magnetic vector potential. For the interacting part \mathcal{H}_{int} of the Hamiltonian, we use the (g_1, g_2) model where the constants g_1 and g_2 describe backward and forward scattering of two particles on opposite sides of the Fermi surface [10]. We look for a BCS mean field solution of the Hamiltonian $\mathcal{H}_0 + \mathcal{H}_{\text{int}}$ with an order parameter corresponding to singlet or equal spin triplet pairing state. Since we are interested in the strong field regime, we do not consider the possibility of triplet pairing with opposite spin which is strongly suppressed by Zeeman splitting contrary to equal spin pairing. Since the dispersion law $E_{\mathbf{k}}$ corresponds to a perfect electron-hole nesting with the vector $(2k_F, \pi/b, \pi/c)$, one should in principle consider on an equal footing the logarithmic divergences appearing in the electron-hole and electron-electron channels. We could consider a more realistic dispersion law with imperfect electron-hole nesting allowing the usual ladder approximation in the electron-electron channel [11]. However, these deviations from a perfect electron-hole nesting do not affect the superconducting instability and can therefore be discarded in the calculation. For a uniform magnetic field, \mathcal{H}_0 can be diagonalized [6] and the transition line $T_c(H)$ is given in the gauge $\mathbf{A}(0, 0, -Hx)$ by the linearized gap equation [8]:

$$\lambda^{-1}\Delta(x, q_z) = \int dx' K_{\sigma, \sigma'}(x, x', q_z)\Delta(x', q_z), \quad (2)$$

$$K_{\sigma, \sigma'}(x, x', q_z) = \frac{T}{bcv^2} \frac{\cos[(\sigma - \sigma')\mu_B H(x - x')/v]}{\sinh[|x - x'|2\pi T/v]} J_0 \left(\frac{4t_z}{\omega_c} \sin \left[\frac{G}{2}(x - x') \right] \sin \left[q_z \frac{c}{2} - \frac{G}{2}(x + x') \right] \right), \quad (3)$$

where J_0 is the zeroth-order Bessel function. $\Delta(x, q_z)$ is the Fourier transform of the order parameter with respect to z . We have also used the fact that the highest $T_c(H)$ is obtained for a uniform order parameter along the field. $\sigma = \sigma' = 1$ ($\sigma = -\sigma' = 1$) for triplet (singlet) superconductivity. $\lambda_{SS} = -(g_1 + g_2)/2$ and $\lambda_{TS} = (g_1 - g_2)/2$ are the coupling parameters for singlet and triplet superconductivity, respectively. $T_c(H)$ is clearly independent of q_z (which only shifts the origin of the x axis by $q_z c/2G$) allowing us to set $q_z = 0$. Using $K(x, x') = K(x + \pi/G, x' + \pi/G)$, the solution of Eq. (2) can be written without any loss of generality as $\Delta_Q(x) = e^{iQx}\tilde{\Delta}_Q(x)$ where $\tilde{\Delta}_Q(x + \pi/G) = \tilde{\Delta}_Q(x)$ and $-G < Q \leq G$. This form of $\Delta_Q(x)$ is more general than Lebed's choice [8] which corresponds to $Q = 0$. In order to determine $T_c(H)$ and $\tilde{\Delta}_Q(x)$, we have to solve Eq. (2) numerically. We first consider triplet pairing. In the low field regime ($\omega_c \ll T$), all the possible values of Q correspond to the same $T_c(H)$. In the quantum regime ($\omega_c \gg T$), this degeneracy is lifted. The highest $T_c(H)$ is always obtained for $Q = 0$ or $Q = G$. The phase diagram is shown in Fig. 1. The GL regime is followed by a regime where the solutions $Q = 0$ (Lebed's line) and $Q = G$ al-

ternate for increasing magnetic field, the last phase (very strong field) corresponding to $Q = 0$. Because these two solutions are characterized by a different structure of the order parameter, the field induces a *cascade of superconducting phases separated by first-order transitions*. The resulting phase diagram is somehow reminiscent of the field-induced spin-density-wave phases [5,12]. For singlet pairing, the best values of Q are $\pm 2\mu_B H/v$ and $\pm(G - 2\mu_B H/v)$. The shift $\pm 2\mu_B H/v$ of the value of Q displaces the Fermi surfaces of spin \uparrow and spin \downarrow relative to each other and compensates partially the effect of Zeeman splitting as is the case in a LOFF state (note that the order parameter remains uniform along the magnetic field direction) [13]. As a result, one half of the phase space is again available for pairing so that the reentrance in high field is not completely suppressed as shown in Fig. 1 [in the singlet case, the cascade also exists but at very low temperature: $T_c(H)/T_c \sim 10^{-3}$]. In the preceding discussion, we have assumed that the type of superconductivity remains the same as the field is increased. If $\lambda_{SS} > \lambda_{TS} > 0$, one would observe singlet superconductivity in weak magnetic field and triplet su-

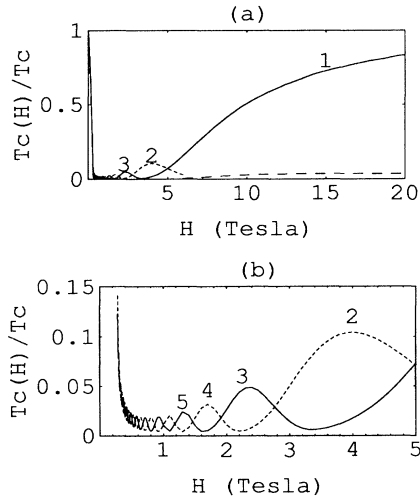


FIG. 1. Critical temperature $T_c(H)/T_c$ vs magnetic field ($t_z = 20$ K and $t_z/T_c \sim 13$). (a) The solutions $Q = 0$ (solid line) and $Q = G$ (dashed line) alternate for increasing magnetic field in the triplet case. Each phase is labeled by an integer N (see text). In the singlet case, $T_c(H)$ is strongly reduced but the reentrance in very strong field is not suppressed (long-dashed line). (b) The low temperature region for the triplet case.

perconductivity in strong magnetic field as a consequence of the Zeeman splitting effect.

The periodic part $\tilde{\Delta}_Q(x)$ of the order parameter is shown in Fig. 2. In the GL regime, $\tilde{\Delta}_Q(x)$ is localized around the points $x_n = n\pi/G$. Using the degeneracy of $T_c(H)$ with respect to Q , it is possible to recover the Abrikosov Gaussian solution $f(x - x_n) = \sum_Q e^{-iQx_n} \Delta_Q(x)$. In the quantum regime where the degeneracy with respect to Q is lifted, $\tilde{\Delta}_Q(x)$ becomes extended. This suggests that the usual vortex lattice structure is strongly modified or even suppressed when $\omega_c \gg T$. In very strong field ($\omega_c \gg t_z$), $\tilde{\Delta}_Q(x)$ is almost uniform.

In order to understand more precisely the evolution of the vortex lattice in the quantum regime, it is necessary to determine the order parameter for temperatures slightly below $T_c(H)$. To do this, we proceed as follows (we only consider the triplet case). We first consider the GL regime and construct the Abrikosov vortex lattice by taking a linear combination of the Gaussian functions $f(x - x_n - q_z c/2G)$ (we have restored the q_z dependence). Using the relation between f and Δ_Q , we can express the order parameter as a linear combination of the functions $\Delta_Q(x, q_z)$. We then take into account the discreteness along the z direction by requiring the vortex lattice to have the periodicity $a_z = Nc$ (N integer) so that the vortex cores can lie between two planes (which will minimize the free energy). In this case, only the $Q = 0$ or $Q = G$ solution is necessary to describe the Abrikosov vortex lattice:

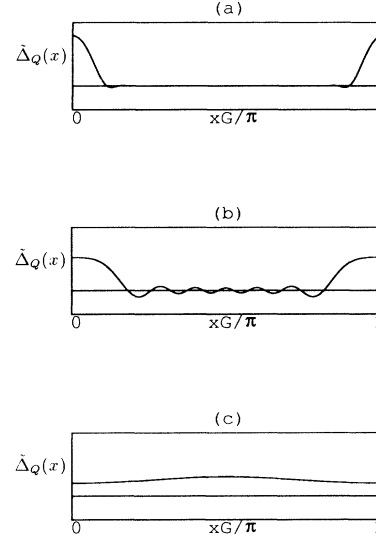


FIG. 2. Periodic part $\tilde{\Delta}_Q(x)$ of the order parameter. (a) $H = 0.13$ T, $Q = 0$. (b) $H = 0.48$ T, $Q = 0$. (c) $H = 5.8$ T, $Q = 0$. Note that $\tilde{\Delta}_{Q=0}(x)$ is real.

$$\Delta_{Q,N}(x, z = mc) = C \sum_{l=m \text{ mod } N} \Delta_{2l}^Q e^{i(Q+2lG)x}, \quad (4)$$

where $Q = 0$ (G) if N is odd (even). For simplicity, we consider a square lattice, but a similar analysis can be made for a triangular lattice. The solution (4) is naturally extended to the quantum regime where the best solution corresponds to $Q = 0$ or $Q = G$ depending on the value of the field. It can be seen that $|\Delta_{Q,N}(x, m)|$ has periodicity $a_x = \pi/NG$ and $a_z = Nc$ for every value of the field. We have the usual relation $Ha_x a_z = \phi_0$ showing that there is one flux quantum in the unit cell (a_x, a_z). In order to know completely the order parameter, we have to determine the value of N . In the GL regime, the periodicity is given by the coherence lengths. We have $a_x/a_z = \xi_x/\xi_z = v\sqrt{2}/t_z c$, which leads to $N_{GL} \sim (t_z/\omega_c)^{1/2}$. In the quantum regime, N labels the successive superconducting phases and decreases by one unit at each phase transition. In this regime, N is found to vary as $N_{QR} \sim t_z/\omega_c$: the periodicity a_z is not given by the coherence length ξ_z anymore, but by the amplitude of the semiclassical orbits (or equivalently the extension of the one-particle quantum states in the z direction). The cascade of phase transitions is then due to commensurability effects between the crystalline lattice spacing c and the periodicity a_z of the order parameter. In very strong field, the electrons become localized in the planes $z = mc$. Therefore, the last phase corresponds to $N_{QR} = 1$ (this choice $N_{QR} = 1$ is possible because of the square symmetry of the order parameter). The relation between the value of Q and the parity of N obtained in the GL regime remains true for every value of the field. Since $T_c \ll t_z$, N cannot be a monotonic de-

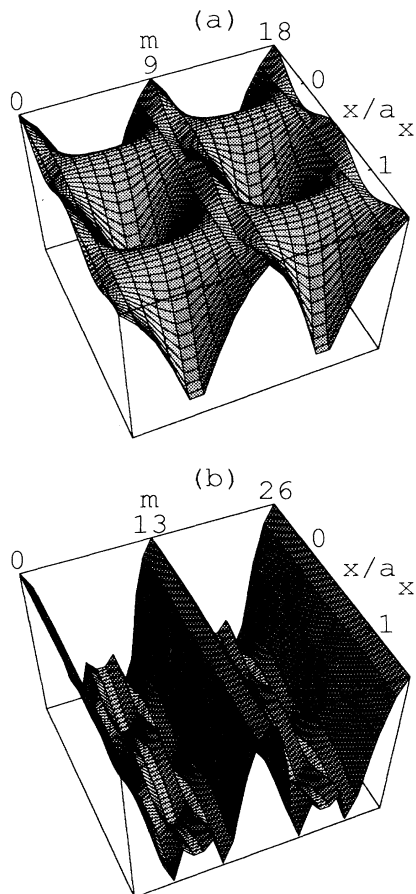


FIG. 3. Order parameter $|\Delta_{Q,N}(x,m)|$. (a) $H = 0.13$ T, $N = 9$, $Q = 0$. (b) $H = 0.48$ T, $N = 13$, $Q = 0$.

creasing function of the field, but has to increase strongly at the transition between the GL and quantum regimes. With the parameters used to obtain Fig. 1, $N_{GL} = 6$ at the end of the GL regime, while it is possible to distinguish 21 phases in the quantum regime. This increase of N corresponds to the disappearance of the vortex lattice as shown in Fig. 3 (the phase of the order parameter and the current distribution also support this analysis). In the quantum regime, the order parameter shows a symmetry of Lamellar type consistent with the one-particle quantum states which are localized in the z direction. In the last phase ($N = 1$), the electrons are mainly localized in the planes $z = mc$ which interact by Josephson coupling. This coupling arises when $t_z \ll \omega_c$ and follows from the magnetic field induced 2D localization. When $t_z \ll \omega_c$, the order parameter corresponds to a square lattice of Josephson vortices. For a superconducting state with a triangular symmetry, the system evolves from a triangular Abrikosov vortex lattice (for $\omega_c \ll T$) towards a triangular Josephson vortex lattice (for $\omega_c \gg t_z$). N is always even and the last phase corresponds to $N = 2$.

In Bechgaard salts like $(TMTSF)_2ClO_4$ and

$(TMTSF)_2PF_6$, the phase diagram proposed in this paper could be observed. The intermediate phases involve a low temperature range [$T_c(H) \sim 10$ mK] which can be reached experimentally only in the case of triplet superconductivity. The reentrance in high field should be visible even in the case of singlet superconductivity [$T_c(H) \sim 50$ mK]. As previously noticed [8], the precise alignment of the magnetic field along the y axis ($\Delta\theta < 1^\circ$) required in this field regime might be an experimental difficulty.

Finally, we would like to mention the case of quasi-2D superconductors described by the dispersion law $E_{\mathbf{k}} = k_{\parallel}^2/2m + t_z \cos(k_z c)$. Although the situation is different due to the presence of open and closed semiclassical orbits at the Fermi surface, we may wonder whether the phase diagram in the presence of a magnetic field parallel to the superconducting planes bears some similarities with the one of the quasi-1D superconductors which has been proposed in this Letter.

One of us (C.A.R. Sá de Melo) acknowledges support from NSF via STCS Grant No. STC 88-09854. Laboratoire de Physique Solides is associé au CNRS.

- [1] A.A. Abrikosov, Zh. Eksp. Teor. Fiz. **32**, 1442 (1957) [Sov. Phys. JETP **5**, 1174 (1957)].
- [2] L.P. Gor'kov, Zh. Eksp. Teor. Fiz. **36**, 1918 (1959) [Sov. Phys. JETP **9**, 1364 (1959)].
- [3] A.K. Rajagopal and R. Vasudevan, Phys. Lett. **23**, 539 (1966); L.W. Gruenberg and L. Gunther, Phys. Rev. **176**, 606 (1968).
- [4] For a review, see M. Rasolt and Z. Tešanović, Rev. Mod. Phys. **64**, 709 (1992).
- [5] See review articles and references therein in *The Physics and Chemistry of Organic Superconductors*, edited by G. Saito and S. Kagoshima (Springer-Verlag, Berlin, 1987).
- [6] L.P. Gor'kov and A.G. Lebed', J. Phys. (Paris), Lett. **43**, L433 (1984).
- [7] N. Dupuis and G. Montambaux, Phys. Rev. B **46**, 9603 (1992).
- [8] A.G. Lebed', Pis'ma Zh. Eksp. Teor. Fiz. **44**, 89 (1986) [JETP Lett. **44**, 114 (1986)]; L.I. Burlachkov *et al.*, Europhys. Lett. **4**, 941 (1987).
- [9] See, for instance, L.N. Bulaevskii, Int. J. Mod. Phys. B **4**, 1849 (1990).
- [10] See, for instance, J. Sólyom, Adv. Phys. **28**, 201 (1979).
- [11] In Bechgaard salts, a magnetic field along the z direction restores the logarithmic divergence in the electron-hole channel, a situation known as quantized nesting and responsible for the field-induced spin-density-wave phases [5,12]. For a field along the y axis, this situation does not occur because the deviation to the perfect nesting in the y direction, which is not affected by the field, is large enough to suppress the nesting.
- [12] For a review and references, see G. Montambaux, Phys. Scr. **T35**, 188 (1991).
- [13] P. Fulde and R.A. Ferrell, Phys. Rev. **135**, A550 (1964); A.I. Larkin and Yu.N. Ovchinnikov, Zh. Eksp. Teor. Fiz. **47**, 1136 (1965) [Sov. Phys. JETP **20**, 762 (1965)].