Interplay of Fulde-Ferrell-Larkin-Ovchinnikov and Vortex states in two-dimensional Superconductors

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Abstract

Clean superconductors with weakly coupled conducting planes have been suggested as promising candidates for observing the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state. We consider here a layered superconductor in a magnetic field of arbitrary orientation with respect to the conducting plane. In this case there is competition of Pauli spin-pair-breaking effects, favoring the FFLO state, and orbital-pair-breaking effects, favouring the Abrikosov vortex state. In previous work, phase transitions to phases with pairing in Landau levels with quantum numbers \(n > 0\) have been predicted. Here, we calculate the actual structure of the stable states below \(H_{c2}\) by minimizing the free energy. We find new order parameter structures differing from both the traditional Abrikosov and FFLO solutions. These include two-dimensional periodic structures with several zeros of the order parameter, as well as quasi-one-dimensional structures consisting of vortex chains separated by FFLO domains. We discuss the limit of high \(n\), where some interesting but yet unsolved questions appear.

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1 Introduction

Suppression of superconductivity by a magnetic field is a consequence of its interaction with either the magnetic moment due to the orbital angular momentum [1] or due to the spin [4] of the electrons. The recent discovery of several classes of layered compounds with nearly decoupled superconducting planes has renewed the interest in the interplay of spin and orbital pair-breaking mechanisms. These include high-$T_c$ cuprate superconductors, organic superconductors with very large upper critical fields, such as $(\text{TMTSF})_2\text{PF}_6$ [3], and hybrid ruthenate-cuprate compounds like RuSr$_2$GdCu$_2$O$_8$ [4, 5]. The orbital pair breaking effect, described by a vector potential $\vec{A}$, is usually much larger than the spin effect and dominates most of the phenomena observed in magnetic fields, e.g., the structure of vortices in type II superconductors. Nevertheless, the spin effect has attracted considerable interest [2, 6]; it has soon been realized that its relative importance can be greatly enhanced by reducing the spatial extension of samples in a direction perpendicular to the external field. For the case of an almost two-dimensional superconductor with applied field parallel to the conducting planes, the orbital upper critical field is extremely high and the spin pair-breaking mechanism becomes the dominant one.

Then, if the orbital pair breaking mechanism can be completely neglected, the homogeneous superconducting state at $T = 0$ becomes energetically favourable [2, 4] at an upper critical field given by $\mu H_c \approx \Delta_0/\sqrt{2}$, where $\Delta_0$ is the BCS gap at $T = 0$ (Chandrasekhar-Clogston limit). Later, a spatially inhomogeneous state (FFLO state) with higher critical field was theoretically predicted by Fulde and Ferrell [7], and Larkin and Ovchinnikov [8]. In the FFLO state the order parameter performs one-dimensional spatial oscillations, which reduce the pair-breaking effect of the external field. Other theoretical studies on the FFLO state include the relation to the mixed state [9, 10], the relevance of the shape of the Fermi surface [11, 12], heavy-fermion superconductivity [13], one-dimensional systems [14], the vicinity of the tricritical point [15], and a calculation of the lower critical field [16]; to mention only a few. Recently, it has been shown by one of us [17], that for superconductors with cylindrical (circular) Fermi surface one finds, at low temperature $T$, several different types of two-dimensional periodic structures with lower free energy than the traditional FFLO-state. Experimentally, while indications of spin paramagnetic effects have been observed, no clear evidence for an FFLO-like phase has been obtained so far. The recent discovery of several classes of compounds with nearly isolated conducting layers has, however, opened new possibilities to detect the FFLO state.

In most treatments, either the “pure” orbital effect or the “pure” spin
effect has been investigated. Here, we study, following Bulaevskii [18], a situation where both effects must be considered. The relative importance of the spin effect is enhanced by means of the following geometrical arrangement: Consider a quasi-two-dimensional thin film with a large component $\mathbf{H}_\parallel$ of the applied magnetic field parallel to the film, and a small perpendicular component $\mathbf{H}_\perp$. The smallness of the perpendicular component makes the spin effect comparable with the (much stronger) orbital effect. The upper critical field for such a situation has first been calculated by Bulaevskii [18] and later studied in the vicinity of the tricritical point [15]. Still later, Bulaevskii’s results were rediscovered in a different theoretical framework, and generalized to arbitrary temperatures and d-wave superconductivity by [19]. The essential result is a non-monotonic critical field curve $B_{c2}(T)$ which consists of several pieces belonging to different values of the Landau quantum number $n$. Such a behavior is in contrast to standard Ginzburg-Landau (GL) as well as microscopic theory, where the pair-wave function is always found to be in the lowest Landau level $n = 0$ at $H_{c2}$. Obviously, the gap at the Fermi surface opened by $\mathbf{H}_\parallel$ forces the single electrons in pairing states with higher Landau levels $n > 0$. We note that formally similar upper critical field curves may also occur in a different physical context, at extremely high fields when Landau quantization of the single electron levels must be taken into account [20, 21, 22, 23]. For the present situation, Landau quantization of the single electron levels can be neglected and the quasiclassical approximation holds.

The present communication addresses the question of the actual structure of the spatially inhomogeneous state below some part of the upper critical field curve characterized by a Landau quantum number $n$. This state is of interest because it is a consequence of a competition between two different pair breaking mechanisms. It is also of practical interest because the portion of the $H, T$ plane where it occurs may be easier accessible experimentally than the one corresponding to the “pure” FFLO state. Note also, that a perfect orientation of the external field is difficult to achieve; as a consequence, a small perpendicular component is always present even if one tries to avoid it.

2 Calculation and theoretical background

We use as a framework the quasiclassical Eilenberger equations [24], properly generalized to include the coupling between electron spins and magnetic field [25]. The notation used in the present work is the same as in Ref. [26]. We restrict ourselves to pure superconductors with an isotropic gap and a
circular Fermi-surface. The quasiclassical equations consist of the transport
equations for the Greens functions $f, f^+, g$ and the self-consistency equations
for the order parameter $\psi$ and the vector potential $\vec{A}$. The transport equa-
tions are given by:

\begin{align}
2\omega_s + \hbar \vec{v}_F(\hat{k}) \left( \vec{\nabla}_R - \imath (2e/\hbar c) \vec{A} \right) f(\vec{R}, \hat{k}, \omega_s) &= 2\psi(\vec{R}) g(\vec{R}, \hat{k}, \omega_s), \\
2\omega_s - \hbar \vec{v}_F(\hat{k}) \left( \vec{\nabla}_R + \imath (2e/\hbar c) \vec{A} \right) f^+(\vec{R}, \hat{k}, \omega_s) &= 2\psi^+(\vec{R}) g(\vec{R}, \hat{k}, \omega_s), \\
g(\vec{R}, \hat{k}, \omega_s) &= \left( 1 - f(\vec{R}, \hat{k}, \omega_s) f^+(\vec{R}, \hat{k}, \omega_s) \right)^{1/2}.
\end{align}

Here, the Fermi velocity is denoted by $\vec{v}_F(\hat{k})$, the Zeeman term is contained
in the quantity $\omega_s = \omega_l + \imath \mu B$, where $\omega_l$ are Matsubara frequencies, $\mu$ is the
magnetic moment of the electron and $B$ is the magnitude of the induction.

The self-consistency equations are given by

\begin{align}
\left( 2\pi k_B T \sum_{l=0}^{N_D} \frac{1}{\omega_l} + \log (T/T_c) \right) \psi(\vec{R}) &= \pi k_B T \sum_{l=0}^{N_D} \int d^2\hat{k}' \left[ f(\vec{R}, \hat{k}', \omega_s) + f(\vec{R}, \hat{k}', \omega_s^*) \right], \\
\vec{\nabla}_R \times \left( \vec{B}(\vec{R}) + 4\pi \vec{M}(\vec{R}) \right) &= \frac{16\pi^2 e k_B T N(E_F)}{c} \sum_{l=0}^{N_D} \int \frac{d^2\hat{k}'}{4\pi} \vec{v}_F(\hat{k}') \Im g(\vec{R}, \hat{k}', \omega_s), \\
\vec{M}(\vec{R}) &= 2\mu^2 N(E_F) \vec{B}(\vec{R}) - 4\pi k_B T N(E_F) \mu \sum_{l=0}^{N_D} \int \frac{d^2\hat{k}'}{4\pi} \Im g(\vec{R}, \hat{k}', \omega_s). \tag{5}
\end{align}

The magnetization $\vec{M}$ comes from the spins; the second term in Eq. (5) is due
to the interaction between spins and supercurrent and may be neglected in the
high $\kappa$-limit. These equations have to be supplemented by an expression
for the Gibbs free energy $G$. A functional $G$ leading to the above Euler-
Lagrange equations may be constructed as a straightforward generalization
of Eilenbergers original expression [24] and will not be written down here.

As a first step, the differential operator in the transport equations has
to be inverted using standard methods [27]. This calculation reproduces the
equation for $B_{c2}$ obtained previously [18, 19], as well as the eigenfunctions of
the linearized gap equation, which are given by

\begin{equation}
\phi_n(k, \vec{r}) = \frac{A}{\sqrt{n!}} \left( \frac{-1}{\sqrt{2}} \right)^n \exp \left( \frac{i}{\hbar} k x \right) \exp \left( -\frac{\kappa \perp}{2\hbar} (y - y_k)^2 \right) H_n \left( \sqrt{\frac{\kappa \perp}{\hbar}} (y - y_k) \right). \tag{7}
\end{equation}
Here, $x, y$ are the coordinates in the superconducting layer, $n = 0, 1, 2, \ldots$ is a discrete (Landau level) quantum number, $k$ is a continuous quantum number, and $H_n$ is the Hermite polynomial of order $n$. The remaining quantities in (7) are given by $\kappa_\perp = \frac{2|e|}{c}B \sin \Theta$ and $y_k = k/\kappa_\perp$, where $\Theta$ is the (small) angle between the external magnetic field and the superconducting plane. The value of $A$ may be chosen to normalize the complete, orthogonal set of eigenfunctions (6). The same form of the normal state vector potential as in previous work [19, 28] has been chosen.

We want to study the structure of the (stable) inhomogeneous state below $B_{c2}$ for arbitrary $n$. For given temperature $T$, angle $\Theta$, and ratio $T_c/E_F$, the correct value of $n$ is obtained by solving numerically the equation for $B_{c2}$ [19]. Given this $n$, the order parameter below the corresponding branch of the $B_{c2}$ curve can be constructed as a linear combination of eigenfunctions belonging to the infinitely degenerate set (6), which are labeled by the continuous quantum number $k$. We construct a general state with an order parameter which is quasi-periodic (periodic up to a phase factor) in two dimensions. Abrikosov’s method [29], used originally for $n = 0$ and $T \sim T_c$, may be applied in a straightforward manner to the present case of arbitrary $n$. It leads to an order parameter $\psi_n$ which is given by

$$\psi_n(X,Y) = AC_n \sum_{m=-\infty}^{m=+\infty} \exp \left(-\frac{2\pi i b}{a} \cos \alpha \frac{m(m+1)}{2}\right) \times$$

$$\cdot \exp \left(\frac{i}{\hbar} k_m (X + Y \cos \alpha) h (\sin \alpha (Y - mb))\right), \quad (8)$$

where

$$h(x) = \frac{A}{\sqrt{n!}} \left(-\frac{1}{\sqrt{2}}\right)^n \exp \left(-\frac{\kappa_\perp}{2\hbar x^2}\right) H_n \left(\sqrt{\frac{\kappa_\perp}{\hbar}} x\right). \quad (9)$$

Here, $\alpha$ is the angle between the two primitive lattice vectors $\vec{a}, \vec{b}$ (of length $a, b$) spanning the unit cell of the periodic structure we are interested in. An oblique system of coordinates $X, Y$ has been used in (8) whose relation to Cartesian coordinates $x, y$ is given by $x = X + Y \cos \alpha, y = Y \sin \alpha$. In the course of the calculation leading to (8), the continuous numbers $k$ have been restricted to a discrete set $k_m = (2\pi \hbar/a)m$ with integer $m$. The flux due to the perpendicular component $B_\perp = B \sin \Theta$ must be quantized; the corresponding fluxoid quantization condition has been used in the derivation of (8). It takes the form $\kappa_\perp ab \sin \alpha = 2\pi \hbar$ if we assume that each unit cell carries one flux quantum. Periodic order parameters for arbitrary $n$ have been reported previously [30, 31] in the literature.

Among all possible structures (8) the stable one, i.e. the one with the lowest free energy, must be found. To perform this task the quasiclassical
equations have to be solved near $B_{c2}$ and the free energy has to be calculated up to terms of fourth order in the order parameter magnitude. In contrast to the GL region, the coherence length is finite at low $T$; therefore all powers of the order parameter gradient must be taken into account. This large calculation, which generalizes Abrikosov’s solution to arbitrary $T$, has first been performed by Eilenberger \cite{32} for the ordinary vortex lattice, i.e. for $n = 0$. Later, Eilenberger’s mixed state calculations have been generalized by Rammer and Pesch \cite{33} to include strong coupling effects. The present calculation generalizes Eilenberger’s work to arbitrary Landau quantum numbers $n$. It makes extensive use of calculational techniques \cite{34, 27, 32, 35} developed by previous workers in this field and comprises the following main steps: (1) Solution of the linearized transport equations, (2) expansion of transport equations and free energy for small order parameter, (3) solving Maxwell’s equation, and (4) performing a large number of spatial and momentum integrations. While all second order free energy contributions can be calculated exactly, one has to perform an asymptotic approximation in integrals appearing in fourth order terms in order to obtain a simple final result. This approximation is similar to the one suggested by Delrieu \cite{35} and holds for not too low $T$ (see the discussion following Eq.(6) of Delrieu’s paper \cite{35}).

After a long calculation, which will be described in more detail elsewhere, one obtains the following expansion (note that dimensionless units \cite{28} will be used in what follows) for the free energy $G$:

$$G = \bar{G} + \tilde{\alpha}^2 \bar{G}^{(2)} + \tilde{\alpha}^4 \bar{G}^{(4)}$$ \hspace{1cm} (10)

Here, $\tilde{\alpha}^2$ denotes the spatial average of $\psi^2$, our small expansion parameter. The term $\bar{G}$ stems from the spatially constant part of the magnetic induction \cite{32}; it is unimportant in the present context since it does not depend on the structure variables $\alpha, a, b$. The second order contribution is given by

$$\bar{G}^{(2)} = \ln t + t \int_0^\infty ds \frac{1}{\sinh(st)} \left[ 1 - \cos(\mu \bar{B}s) f_1(s^2 \frac{\bar{B}}{2}^\perp) \right],$$ \hspace{1cm} (11)

where $t = T/T_c$, and $\bar{B}, \bar{B}^\perp$ are spatially averaged values of $B(\vec{R})$ and $B^\perp(\vec{R})$ respectively. The function $f_1$ depends on the Landau quantum number $n$ and is given by

$$f_1(x) = \exp(-\frac{x}{2}) L_n(x),$$ \hspace{1cm} (12)

where $L_n$ is the Laguerre polynomial of order $n$. The second order term (11) is also independent of $\alpha, a, b$, but the equation $\bar{G}^{(2)} = 0$ has to be solved in
order to find the upper critical field and the corresponding value of \( n \) (this equation for \( B_{c2} \) agrees with the one given in [19]).

The new information is contained in the fourth order term \( \bar{G}^{(4)} \). The magnetic part, say \( \bar{G}^{(4)}_M \), of \( \bar{G}^{(4)} \) is of order \( 1/\kappa^2 \). This magnetic contribution collects all the terms related to Maxwells equation. If Eilenbergers, or Abrikosovs results are to be rededuced, as a special case of the present theory for \( \vec{H}_\parallel = 0 \) \((n = 0)\), all contributions to the free energy, including \( G^{(4)}_M \), have, of course, to be taken into account (one can successfully check Eq.(10) against these classical results; the same is true for the FFLO upper critical field which may be derived by solving \( \bar{G}^{(2)} = 0 \) in the limit \( \vec{B}_\perp = 0 \)). In the present communication we consider the high-\( \kappa \) limit and neglect the contribution \( \bar{G}^{(4)}_M \). This approximation is used by most workers on the FFLO state, in particular in Larkin and Ovchinnikov’s original paper [8]. A complete treatment including this magnetic contribution as well as magnetization curves for the present geometry, will be reported in a separate publication [36]. Then, the final result for the fourth order term is given by

\[
\bar{G}^{(4)} = \frac{t}{4} \sum_{l=-\infty}^{+\infty} \sum_{j=-\infty}^{+\infty} f_1^2(x_{l,j}) S_{l,j},
\]

where \( f_1 \) has been defined in Eq.(12). The Matsubara sum \( S_{l,j} \) is defined by

\[
S_{l,j} = \sum_{i=0}^{Nd} \frac{2\omega_i^2 - \vec{B}_\perp x_{l,j}/2}{\omega_i^2 + \vec{B}_\perp x_{l,j}/2}^{3/2},
\]

and \( x_{l,j} \) is given by

\[
x_{l,j} = \frac{\pi^2}{\sin^2 \alpha} \left( \frac{L}{a} \right)^2 \dot{l}^2 + \left( \frac{a}{L} \right)^2 j^2 - 2\pi lj \frac{\cos \alpha}{\sin \alpha}.
\]

The minimization with respect to the flux-line structure is done at fixed \( \vec{B}_\perp, \vec{B}_\parallel \) (the minimization with respect to \( \vec{B}_\perp, \vec{B}_\parallel, \vec{\alpha} \) has to be done in separate steps). Therefore, stability conditions should not be applied with respect to the variables \( a/b, \alpha \) but with respect to \( a/L, \alpha \), where \( L \) is a magnetic length defined by \( L^2 = 2/\vec{B}_\perp \). Then, using the results for \( a/L, \alpha \), the ratio \( a/b \) is finally determined with the help of the fluxoid quantization condition.

The fourth order term (13) has a simple interpretation. As is well known, near \( T_c \) the conventional flux-line structure is completely determined by a single quantity, Abrikosov’s geometrical factor \( \beta_A = \bar{\psi}^4/\bar{\psi}^{22} \) (the bar denotes a spatial average). If this quantity is computed using a general order
parameter $\psi_n$, as given by Eq. (8), one obtains, after a lengthy calculation

$$\beta_{A,n} = \sum_{l=-\infty}^{+\infty} \sum_{j=-\infty}^{+\infty} f_l^2(x_{l,j}).$$

(16)

One notices that the free energy (13) differs from $\beta_{A,n}$ only by the temperature-dependent factor $S_{l,j}$ modifying each term in the double sum (for $n = 0$, $\beta_{A,n}$ takes its lowest value 1.1596 for a triangular lattice). The factor $B_{l,j}/2$ in $S_{l,j}$ (see Eq. (14), vanishes in the GL-limit; it reflects nonlocal correlations in Eilenbergers transport equations and implies an additional “microscopic” temperature dependence of the stable magnetic flux structure. One expects, that for higher $T$, the GL approximation $\beta_A$ may be used as a first approximation for the complete free energy (13). It should also be mentioned that the electrons magnetic moment $\mu$, which is the typical spin pair breaking parameter, does not explicitly occur in Eq. (13) (but only in $\bar{G}$, $G^{(2)}$, and $\bar{G}^{(4)}_M$). The spin effect influences, however, the free energy - and the resulting magnetic flux structure - in a very decisive way since it determines the value of the Landau quantum number $n$ in Eq. (13).

3 Results and discussion

To calculate numerically the stable flux structure the following two steps have to be performed: (1) The equation $G^{(2)} = 0$ has to be solved for a given temperature $T$ and angle $\Theta$ to obtain the upper critical field and the corresponding value of $n$. (2) Using these values the stable flux structure has to be determined by calculating the absolute minimum of (13) with respect to $a/L$, $\alpha$. Throughout the numerical calculations a value of $\mu = 0.1$ ($\mu = (\pi/2)k_BT_c/E_f$ in the present system of units) has been used. The calculations have been performed (for various $n$ in the range between 0 and 30) at two different temperatures $t = T/T_c = 0.5$ and $t = 0.2$, in order to investigate the influence of the above discussed low-temperature correlations.

The minimization procedure turned out to be more involved than one would expect from the simple form of Eq. (13). One finds a large number of minima belonging either to different lattice structures or to different basis vectors of the same lattice. The number of minima increases rapidly with increasing Landau quantum number $n$. The quantity shown in all plots is actually $\bar{G}_N^{(4)}$, which is $G^{(4)}$ multiplied by a factor of $16t^3/7\chi(3) \approx 2t^3$ (in order to normalize it for $t \to 1$ to the GL result). Fig. 1 shows a contour plot for $n = 10$, $t = 0.5$ of $\bar{G}_N^{(4)}$ in the most important part of the $\alpha,a/L$-plane; higher values of $a/L$ need not be considered, since they correspond
to lattices with exchanged axes $a, b$. The minima of the free energy are visible as dark regions. Two different types of minima may be distinguished in Fig.1 (and similar plots). A minimum of the first type is represented by an isolated dark region and corresponds to an “ordinary” two-dimensional lattice (several equivalent points in the $\alpha, a/L$-plane, which have the same free energy and belong to the same lattice, but to different basis vectors, may be found in Fig.1). The second type of minimum can be found in the straight ($\alpha$-independent) dark regions in the lower part of Fig 1. Near these “flat valleys” the free energy depends mainly on $a/L$, a small dependence on $\alpha$ along some lines of fixed $a/L$ exists, but is invisible in Fig 1. The minima in these flat valleys will be referred to as “quasi-one-dimensional” lattices, because of the small energy barrier along the lines of constant $a/L$; a more detailed discussion will be given below.

The phases with $n = 0, 1, 2$ occupy a relatively large portion of the phase diagram [18, 19] and are therefore of particular interest in view of a possible experimental detection of these new states. For the lowest Landau index $n = 0$, we recovered at both temperatures, as expected, the conventional triangular vortex lattice as stable state (global minimum of the free energy).

To create a state with $n = 1$, a possible choice for the tilting angle of the external magnetic field is $\Theta = 0.3$ (degree) at $t = 0.5$; another possible choice is $\Theta = 1.2$ at $t = 0.2$. The stable structure found for $n = 1$ is, at both temperatures, of the quasi-one-dimensional type. A contour plot of the magnitude of the order parameter is shown in Fig. 2; the structure consists of chains of vortices separated by a single line of vanishing order parameter. The latter may be considered as a fragment of the one-dimensional FFLO-state; for higher $n$ local minima appear whose order parameters look similar to Fig.2 except that $n$ lines of order parameter zeros appear between the vortex chains.

Let us discuss these quasi-one-dimensional structures - which have been verified as local, but not necessarily global, minima for many higher Landau indices in the range $n < 30$ - in more detail. They consist of two subsystems, vortices and one-dimensional FFLO-type oscillations. Both of these are of the “ordinary” type, i.e. they are basically unaffected by their mutual interaction; the phase change when encircling a vortex is $2\pi$ and the wave length as well as the behavior of the phase of the one-dimensional periodic structure agree with that of the one-dimensional FFLO state. For all of these states, the coupling energy between neighboring vortex chains is small. As a consequence, the energy barrier preventing motion of the vortex chains in the direction parallel to the chains is very small (for $n = 1$ the $\alpha$-dependence shows up in the sixth digit of the free energy). Thus, even small fluctuations will destroy the periodicity in the direction parallel to the chains, making the
material effectively one-dimensional. These decoupling of the vortex chains leads to a quasi-continuum of unit-cells (similar to the one shown in the lower part of Fig. 1 for $n = 10$) which is located near $a/L = 1.03$ for $n = 1$. Experimentally, one expects a pronounced change in transport properties if, by decreasing the angle $\theta$, the system makes a transition from $n = 0$ to $n = 1$.

As regards the next higher Landau level index, $n = 2$, stability calculations have been performed for the two sets of parameters $\Theta = 0.17$, $t = 0.5$ and $\Theta = 0.7$, $t = 0.2$. At $t = 0.5$ a triangular lattice is realized as the state of lowest free energy. At $t = 0.2$ a slightly distorted triangular lattice ($\alpha = 60.05$) is realized. The distortions are due to the microscopic factor $S_{ij}$ in Eq. (13). Thus, the influence of the microscopic correlations on these low-$n$ states is visible but very small. The order parameter for $n = 2$ is shown in Fig. 3. The triangular unit cell contains three order parameter zeros. These zeros have different vorticity; the phase change when encircling a zero is $+2\pi$ for two of the vortices and $-2\pi$ for the third. Recall that, for arbitrary $n$, the unit cell as a whole carries exactly one flux quantum.

For $n = 3$ at $t = 0.5$ we found again a quasi-one-dimensional stable state. The order parameter for $n = 3$ looks similar to Fig. 2, except for three dark lines appearing between the vortex chains. No stable quasi-one-dimensional states have been found for higher $n$ ($n > 3$). In the interval $3 < n < 10$ the global free energy minima are given by various types of two-dimensional lattices, including triangular, quadratic and oblique structures. With increasing $n$ the influence of the microscopic correlations on the lattice structure increases; the stable state does not only depend on $n$ but may change between $t = 0.5$ and $t = 0.2$ (keeping $n$ constant).

The phases with higher $n$ correspond to very small values of $\Theta$ (e.g. for $n = 8$ and $t = 0.5$ one has $\Theta = 0.05$). One expects the resulting stable states to be already very similar to the FFLO state, which corresponds to the limit $n \to \infty$, $\Theta = 0$ ($B_\perp = 0$). This FFLO-limit $n \to \infty$ of the present theory is a subtle step. An analytical limit of the calculation cannot be performed since in the present formulation extensive use has been made of the fluxoid quantization condition, which becomes meaningless if no perpendicular component exists. Nevertheless, one expects for general reasons of continuity, that the present theory comprises the FFLO theory in the sense, that, with increasing $n$, the numerical results become more and more similar to the FFLO state. Thus, for $t = 0.5$ we expected, as a natural candidate for a smooth transition to the FFLO state, the quasi-one-dimensional state to be stable for large $n$. At $t = 0.2$ we expected, in view of [17], a two-dimensional structure to be stable for large $n$.

However, contrary to our expectation, no single stable lattice could be found in our calculations which have been performed up to $n = 30$ for $t = 0.5$. 
and $n = 20$ for $t = 0.2$. Contour plots of the free energy look similar to Fig. 1 but with an increased number of local minima at higher $n$. At both temperatures the transition from $n$ to $n+1$ (keeping $t$ fixed and varying $\Theta$) leads generally not to the same but to a different lattice; among these stable states we found all kinds of oblique, triangular and rectangular two-dimensional structures. Quasi-one dimensional states, which are stable at $n=1,3$, have also been found as local minima in these high-$n$ calculations, but had always higher energy than the minima corresponding to two-dimensional states. The difference in free energy of the different local minima decreases with increasing $n$.

At present, we are unable to decide whether this strange vortex-liquid like limiting behavior, which would imply the instability of the FFLO state, is an artifact of our calculations or corresponds to reality. There are some arguments in favor of the latter possibility: Adding a perpendicular component of the magnetic field, no matter how small, implies flux quantization; this changes the conditions which determine the stable state in a discontinuous way. Further, the size of the unit cell and the number of vortices (with different vorticity) per unit cell increases with $n$. It seems reasonable to assume that the energy differences between different vortex arrangements (lattice structures) become small for large $n$. More work is required to settle this question.

4 Conclusion

We considered a geometrical arrangement with competition of orbital and spin pair breaking effects in a two-dimensional superconducting state. This arrangement comprises the traditional vortex state (for $B_{\parallel} = 0$) as well as the FFLO state (for $B_{\perp} = 0$). Minimizing the free energy we found several new structures below $B_{c2}$ with pairing states in Landau levels $n > 0$. These include two-dimensional structures with vortices of different vorticity as well as mixtures of one-dimensional periodic order parameter oscillations and vortex chains. The latter show a fluid-like behavior, as regards vortex motion in the direction parallel to the chains. This feature could be used, besides more local spectral techniques, to identify this new state experimentally. Topics to be treated in future work include the high-$n$ limit, contributions from finite values of $\kappa$, magnetization curves, a more systematic account of unit cell structures, and the transitions between pairing states belonging to different $n$. 
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FIGURE CAPTIONS

1. Contour plot of the fourth order free energy contribution $\bar{G}^{(4)}_N$ for $n = 10, \ t = 0.5$ as a function of $\alpha, a/L$. Minima of free energy must be sought in dark regions.

2. Contour plot of $|\psi|^2$ as a function of $x, y$. Stable structure for Landau level $n = 1$ at $t = 0.5$ with lattice parameters $a/b = 2.75, \ \alpha = 69.83$. A small energy barrier exists against translation of the vortex chains relative to each other.

3. Contour plot of $|\psi|^2$ as a function of $x, y$. Stable structure for Landau level $n = 2$ at $t = 0.5$ with lattice parameters $a/b = 1.00, \ \alpha = 60.00$ ($a/L = 1.90$). The unit cell carries one flux quantum and contains three order parameter zeros.
Fig. 1. Klein, Rainer, Shimahara
Order parameter for $n=1$
Order parameter for n=2

Fig. 3 Klein, Rainer, Shimahara