EUROPHYSICS LETTERS 1 May 2000

Europhys. Lett., **50** (3), pp. 375–381 (2000)

Influence of the paramagnetic effect on the vortex lattice in 2D superconductors

M. Houzet(*) and A. Buzdin

Centre de Physique Théorique et de Modélisation de Bordeaux, ERS-CNRS 2120
Université Bordeaux I - 33405 Talence Cedex, France

(received 7 February 2000; accepted in final form 6 March 2000)

PACS. 74.60.Ec – Mixed state, critical fields, and surface sheath.

Abstract. – In 2D superconductors under a high magnetic field, when the upper critical field $H_{\mathrm{c}2}$ is determined by both the orbital and paramagnetic effects, new solutions for the superconducting order parameter, corresponding to higher Landau levels, must be realized. In order to determine the structure of the vortex lattice in the new phases, we derive a modified Ginzburg-Landau functional in the high-κ limit. Within this model, we show that the stability of the structures depends on a new criterion which replaces the Abrikosov parameter β_{A}. The triangular lattice is not always favored. We calculate the most favorable structures at different temperatures, and we find changes of the symmetry, as well as the transition order.

In 1964, Larkin and Ovchinnikov [1] and Fulde and Ferrel [2] predicted the appearance of a new superconducting state, when the magnetic field is acting on the electron spins only. This state is now called the "FFLO" state. It is characterized by a modulated superconducting order parameter. The (H,T)-phase diagram was calculated by Sarma and Saint-James [3], assuming that the transition from normal (N) to FFLO state is second order. The FFLO state may appear only at temperatures $T < T^* \approx 0.56T_{\mathrm{c}}$, the temperature of the tricritical point (TP) where the normal, uniform superconducting and FFLO states coexist.

Up to now, there are no unambiguous evidences of the FFLO state formation. The main reason for the difficulty of experimental observation of such state is the orbital effect which is usually more important than the paramagnetic one. However, it could be suppressed in a 2D superconducting film, when the field is applied parallel to the plane. Moreover, it is possible to control the relative strength of both the orbital and paramagnetic effects by tilting the magnetic field, as the paramagnetic effect does not depend on the inclination. In the pure paramagnetic limit, the N→FFLO transition in 2D superconductors is found to be second order with the order parameter depending on one coordinate only [4]. The situation is different in the presence of the orbital effect as it forces the solutions for the order parameter to be the Landau functions of the different Landau levels. The upper critical field $H_{\mathrm{c}2}$ was calculated first at $T = 0$ [5], and then at finite temperature [6]. Contrarily to the usual situation where the paramagnetic effect is neglected, the Landau functions of the lowest level

(*) E-mail: houzet@pth.u-bordeaux.fr

do not always give the highest critical field, and a cascade of transitions to higher Landau levels may take place as the temperature is decreased.

In the mixed state, the order parameter forms the vortex lattice. In this letter, we discuss its structure just below the upper critical field, when both paramagnetic and orbital effects are present. For this, we derive a Generalized Ginzburg-Landau (GGL) functional similar to that obtained in the pure paramagnetic limit [7]. It is valid when the characteristic modulation length is small compared to the coherence length. This is obtained when the paramagnetic effect is much more important than the orbital one, and for temperatures and magnetic fields near TP. We also consider the high-κ limit by neglecting the magnetic-field inhomogeneities. We find again the succession of increasing Landau-level states along the critical line as the temperature is decreased. Then, we give the approach to determine the structure of the vortex lattice that is formed by the order parameter. We show that the stability of the structures depends on a new criterion which replaces the Abrikosov parameter β_{A}. We calculate the most favorable structures among the lattices with one flux quantum per unit cell at different temperatures, and we find that changes of the symmetry, as well as the transition order, occur as the temperature is decreased. At some temperatures, transition happens to be first order, and finally we complete our description by analysis of this situation.

In the experiments, when the field direction is not very close to the parallel orientation, only the very first Landau level states would appear, and not in the immediate vicinity of TP. In this case, the GL-like theory presented in this paper strictly would not apply. However, we expect that it would be qualitatively correct in the whole temperature interval below TP (as the standard GL theory gives a qualitatively correct description of the properties of superconductors at temperatures well below T_{c}).

In the pure paramagnetic limit, the GGL functional is derived from Gorkov's equations [8], supposing both a small value and weak variation of the order parameter over the coherence length $\xi_0 = \hbar v_{\mathrm{F}}/2\pi T_{\mathrm{c}}$. Such a situation occurs near TP, where the characteristic wave vector of the FFLO state is small compared to ξ_0^{-1}. In the standard GL functional, the coefficient β at the gradient term $\beta\left|\partial\psi\right|^2$ is positive, but it occurs to be a function of the field I acting on the electron spins (paramagnetic effect), and vanishes at TP, being negative at $T < T^*$ [3]. Negative coefficient β means that the modulated state has lower energy comparing with the nonuniform one. To find the modulation vector it is necessary to incorporate into the GL functional the term with second-order derivative $\sim \left(\partial^2\psi\right)^2$. In addition, in the BCS theory, simultaneously with the vanishing of the gradient term, the coefficient γ at the quartic term $\gamma\left|\psi\right|^4$ vanishes too. Due to this particular property, it is also necessary to add the higher-order terms $\sim \psi^2\left(\partial\psi\right)^2$ and eventually ψ^6. The corresponding free-energy density functional near TP is [7]

$$\begin{aligned}\frac{\mathcal{F}}{N(0)T_{\mathrm{c}}^2} &= 0.86\frac{I-I_0(T)}{I^*}\left|\psi\right|^2 + 3.0\frac{T-T^*}{T^*}\xi_0^2\left|\boldsymbol{\nabla}\psi\right|^2 + 0.15\frac{T-T^*}{T^*}\left|\psi\right|^4 + 3.1\xi_0^4\left|\boldsymbol{\nabla}^2\psi\right|^2 + \\ &+0.85\xi_0^2\left\{\left|\psi\right|^2\left|\boldsymbol{\nabla}\psi\right|^2 + \frac{1}{8}\left[\left(\psi^*\right)^2\left(\boldsymbol{\nabla}\psi\right)^2 + \psi^2\left(\boldsymbol{\nabla}\psi^*\right)^2\right]\right\} + 0.011\left|\psi\right|^6 . \qquad (1)\end{aligned}$$

$N(0)$ is the density of states at the Fermi level, $I_0(T)$ is the effective field of the second-order transition from normal to uniform superconducting (S) state, and $I^* = I_0(T^*) \approx 1.07T_{\mathrm{c}}$.

The orbital effect in the presence of the magnetic field $\boldsymbol{H} = \mathrm{curl}\,\boldsymbol{A}$ is now described in the quasiclassical limit in Gorkov's equations, when $p_{\mathrm{F}} \gg \frac{e}{c}H\xi_0$, which means here a large effective mass for the orbital effect. (The Landau quantization effects we discuss in next section are those of *the Cooper pairs*, and not due to the occupation of the Landau levels by electrons in the normal metal at very high magnetic field.) As a result, it appears in (1) by the usual

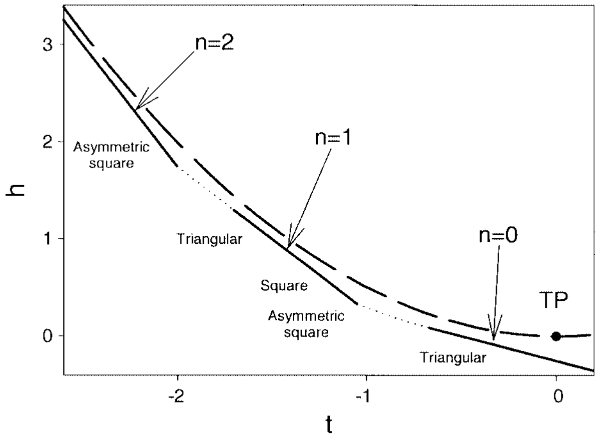

Fig. 1 – The enhancement of the upper critical field is represented against temperature, relative to the N/S critical field of the pure paramagnetic limit. The dashed line is the second-order N/FFLO transition line in the pure paramagnetic limit. The solid line represents the cascade of second-order transitions to the n-th Landau level states in the general case. Description of the superconducting vortex lattice is also given just below the upper critical line. The first-order critical lines are represented as dotted line. The tricritical point (TP) is also represented. The first-order transition lines between different superconducting states are not drawn.

substitution $\boldsymbol{\nabla} \to D = \left(\boldsymbol{\nabla} - \frac{2ie}{\hbar c}\boldsymbol{A}\right)$, and the addition of the new term $\left(3.1\,(2eH/\hbar c)^2\,|\psi|^2\right)$. In general, the free energy of a superconductor in magnetic field also contains the magnetic contribution. In the high-κ limit, the screening is unimportant, and inhomogeneities of the field may be neglected, so this contribution is inessential. Moreover, the effective field acting on the spins is $I = \mu_{\mathrm{B}} H$. We discuss later the criterion of slow variation of the order parameter, satisfied near TP. All the new physics arising in the following comes from the new terms in (1) when compared to the usual GL functional.

In order to calculate H_{c2}, we need to solve the linear eigenvalue problem. Near TP, it is given by

$$0.86\frac{H_{\mathrm{c2}} - H_0(T)}{H^*}\psi - 3.0\frac{T - T^*}{T^*}\xi_0^2\boldsymbol{D}^2\psi + 3.1\left(\xi_0^4\boldsymbol{D}^4\psi + (2eH^*/\hbar c)^2\,\psi\right) = 0\,, \tag{2}$$

where $\boldsymbol{H}_{\mathrm{c2}} = \mathrm{curl}\,\boldsymbol{A}$. The eigenvalues of the operator $\boldsymbol{D}^2$ are well known, *i.e.* the Landau levels $-\frac{4|e|H}{\hbar c}(n+1/2)$. Thus the critical field $H_{\mathrm{c2}}(T)$ is the maximum one solving (2). We introduce the Maki parameter $\alpha = \sqrt{2}H_{\mathrm{c2}}^{\mathrm{orb}}(0)/H_P(0)$ [9] which characterizes the relative strength of orbital and paramagnetic effects: $H_{\mathrm{c2}}^{\mathrm{orb}}(0) = 0.561\Phi_0/2\pi\xi_0^2$ is the pure orbital critical field for 2D superconductors at $T = 0$, $H_P(0) = \Delta_0/\mu_{\mathrm{B}}\sqrt{2}$ is the paramagnetic Chandrasekhar-Clogston limit [3], Δ_0 is the BCS gap parameter. Then we define the dimensionless temperature $t = (T - T^*)\,/\delta T$, and field $h = (H - H_0(T))\,/\delta H$, where $H_0(T) = I_0(T)/\mu_{\mathrm{B}}$, $\delta T/T^* \simeq 2.9/\alpha$, and $\delta H/H^* \simeq 14/\alpha^2$. Therefore, eq. (2) gives $h_{\mathrm{c2}} = -(n+1/2)t - \left[(n+1/2)^2 + 1\right]/2$. Maximization shows that there is a cascade of transitions (see fig. 1) between the successive Landau levels when the temperature is decreased. The 0-th level is favored when $t > -1$, and, for $n > 0$, the n-th level is favored when $-(n+1) < t < -n$. A similar result was already found at any temperature [6] and near TP in [10]. As our GGL functional is valid only near TP, the superconductor must be in the limit $\delta T \ll T^*$ and $\delta H \ll H^*$. It is realized when the paramagnetic effect is more important than the orbital one. Note that the Maki parameter is related to the orbitally limited slope of the upper critical field at T_c: $\alpha = 0.67\mu_B \left|\frac{\partial H_{c2}}{\partial T}\right|_{T_c}$.

It is interesting to compare h_{c2} with the critical field h_c^{para} of the pure paramagnetic limit. In the last case, the critical field is calculated with the order parameter of the form $\psi \propto e^{iqx}$, where q is the vector of modulation. Solutions with $q \neq 0$ exist at $t < 0$ and minimization gives $h_c^{para} = t^2/2$. It is the critical field of the transition to the nonuniform (or FFLO) state at $T < T^*$. We note that h_{c2} is everywhere lower than h_c^{para} as the coexistence of both orbital and paramagnetic effects must destroy superconductivity more easily than when only one effect is present.

In order to find the structure of the order parameter near the second-order critical line at some temperature T, we must calculate the free energy of the different vortex structures which may appear at magnetic field H, slightly lower than $H_{c2}(T)$, and look for the minimum one. However, the allowed structures must satisfy the nonlinear GGL equation (1). We will use Abrikosov's Ansatz [11] in looking for the order parameter among *a priori* structures. These ones will be some particular linear combinations of the Landau functions of the n-th level solving (2) at H_{c2}, and forming quasiperiodic structures. Thus, we introduce the spatial averages (denoted by upper bar) that are calculated on the unit cell of the corresponding vortex lattice. We subtract the linear equation obtained at H_{c2} to the nonlinear one at H. By conserving the terms up to the fourth order, the functional (1) looks like

$$\mathcal{F} \propto A\,(h - h_{c2})\,\overline{|\psi|^2} + \widetilde{\beta}_A(\overline{|\psi|^2})^2,$$

where

$$A = \frac{11.7}{\alpha^2}\left(1 + \frac{\delta H}{H^*}\left\{t\left(n+\frac{1}{2}\right) + \left[\left(n+\frac{1}{2}\right)^2 + 1\right]\right\}\right) \approx \frac{11.7}{\alpha^2},$$

and, after some simplifications, we introduced the new coefficient $\widetilde{\beta}_A$ replacing the Abrikosov one, such as

$$\widetilde{\beta}_A = \frac{0.434}{\alpha}\left\{\left[t + \frac{2}{3}\left(n+\frac{1}{2}\right)\right] I_4 + \frac{2}{3} I_{22}\right\},$$

where $I_4 = \overline{|\psi|^4}/(\overline{|\psi|^2})^2$, and $\frac{2|e|H^*}{\hbar c} I_{22} = \overline{|\psi|^2|\boldsymbol{D}\psi|^2}/(\overline{|\psi|^2})^2$. When $\widetilde{\beta}_A$ is positive, the transition is effectively second order. We calculate the free energy $\mathcal{F}(H) \propto -A^2(h-h_{c2})^2/4\widetilde{\beta}_A$. The most favorable structure should minimize the coefficient $\widetilde{\beta}_A$. Conversely, when $\widetilde{\beta}_A$ is negative, the transition becomes first order, and all calculations must be redone including the sixth-order term in the functional. Note that the usual Abrikosov parameter which only includes the $\overline{|\psi|^4}$-term could not be used; in fact, its contribution to the free energy is always negative below T^*.

As usual, the vortex lattice must have an integer number of flux quanta per unit cell. In the following, we only consider structures with only one flux quantum. In the usual GL theory, such a restriction was justified as it gives always lower β_A coefficient [12]. Here, such a justification does not hold, and we choose it for simplification. We compute them by following the presentation of Eilenberger [13]. The area of the unit cell is $\eta = \Phi_0/H$, and we define the magnetic length $L = \sqrt{\eta}$, which determines the length scale for the variations of the order parameter. Near TP, the slowness of these variations is thus ensured as $L \gg \xi_0$. At a translation, when using the Landau gauge $\boldsymbol{A} = (0, -Hy, 0)$, the functions of the n-th level are

$$\psi_n(x,y) = \frac{(2\sigma)^{\frac{1}{4}}}{(2^n n!)^{\frac{1}{2}}} e^{-\frac{\pi y^2}{\eta}} \sum_p \mathsf{H}_n\left(\sqrt{2\pi}\left(\frac{y}{L} + p\sqrt{\sigma}\right)\right) e^{2i\pi p\frac{x+iy}{L}\sqrt{\sigma} + i\pi p^2\zeta},$$

where H_n are the Hermite polynomials, $\zeta = \rho + i\sigma$ is a complex number which defines the basis vectors of the unit cell $\boldsymbol{r}_{\mathrm{I}} = L/\sqrt{\sigma}$ and $\boldsymbol{r}_{\mathrm{II}} = L\zeta/\sqrt{\sigma}$. Many choices of the basis vectors are equivalent. According to [3], [14], unique mapping of all the structures is made in the complex domain $\{|\zeta| \geqslant 1; 0 < \rho < \frac{1}{2}\}$. Note that $\zeta = e^{i\frac{\pi}{2}}$ corresponds to the square lattice, whereas $\zeta = e^{i\frac{\pi}{3}}$ corresponds to the triangular one. Now we introduce

$$\Lambda_n = \frac{\sqrt{\sigma}}{2^n n!}\left\{\left|\sum_p \mathsf{H}_n\left(2p\sqrt{\pi\sigma}\right)e^{2i\pi p^2\zeta}\right|^2 + \left|\sum_p \mathsf{H}_n\left((2p+1)\sqrt{\pi\sigma}\right)e^{2i\pi\left(p+\frac{1}{2}\right)^2\zeta}\right|^2\right\},$$

which vanishes at odd n. Then,

$$I_4 = \frac{1}{4^n}\binom{2n}{n}\sum_{q=0}^{n}\frac{\binom{n}{q}^2}{\binom{2n}{2q}}\Lambda_{2q},$$

$$I_{22} = \frac{1}{2.4^n}\binom{2n}{n}\sum_{q=0}^{n}\left[4(n-q)+1\right]\frac{\binom{n}{q}^2}{\binom{2n}{2q}}\Lambda_{2q}.$$

At temperature t and corresponding Landau level n, minimization of $\widetilde{\beta}_{\mathrm{A}}$ is done numerically on the vortex lattice structure, that is on the complex parameter ζ, in the domain defined above. We detail our results in the following.

In the temperature range where $n = 0$, $\widetilde{\beta}_{\mathrm{A}} \propto \left(t + \frac{2}{3}\right)\Lambda_0$. Except for the temperature-dependent factor, it is the same quantity that is computed in the GL theory, where the most favorable lattice is found to be the triangular one. We still obtain this result at temperatures higher than $-\frac{2}{3}$. However, at lower temperatures, $\widetilde{\beta}_{\mathrm{A}}$ is negative, and the transition cannot be of the second order. We calculate the first-order transition later.

In the temperature range where $n = 1$, several structures appear as the temperature is decreased. For $-1.16 < t < -1$, the structure is asymmetric. For instance, at $t = -1.08$, energy is minimized for $\zeta = -1.52i$. It corresponds to a quasi-unidimensional structure (fig. 2) where vortices form parallel chains, and they are separated by lines where the amplitude of the order parameter is limited. Such structures evolve continuously to the symmetric square lattice, which is favorable down to $t = -1.2$. For $-1.7 < t < -1.2$, the structure is triangular. At lower temperatures, the transition becomes first order as the sign of $\widetilde{\beta}_{\mathrm{A}}$ changes. Let us now describe the symmetric square (fig. 3) and triangular structures which are very similar apart from the change of symmetry. The order parameter vanishes in several points of the unit cell defined by $\boldsymbol{r}_{\mathrm{I}}$ and $\boldsymbol{r}_{\mathrm{II}}$: in the corners, where the phase decreases by 2π around each pole, and in the middle of the segments limiting the cell, where the phase increases by 2π. On the whole there is one positive "flux quantum" in the unit cell as specified in the definition of the Eilenberger functions. (Compare to the $n = 0$ case where the order parameter vanishes only once in the middle of the cell.)

In the temperature range where $n = 2$, the most favorable structure is square asymmetric when $-2.84 < t < -2$. For instance, at $t = -2.5$, $\zeta = -1.96i$. The structure is also quasi-unidimensional and there are on the whole two positive and one negative flux quanta in each cell. At $t < -2.84$, the transition becomes first order.

We are also interested in the pure paramagnetic limit. It does not appear explicitly in $\widetilde{\beta}_{\mathrm{A}}$. However, in this limit δT and δH go to zero, thus it corresponds to the high-n limit. In this

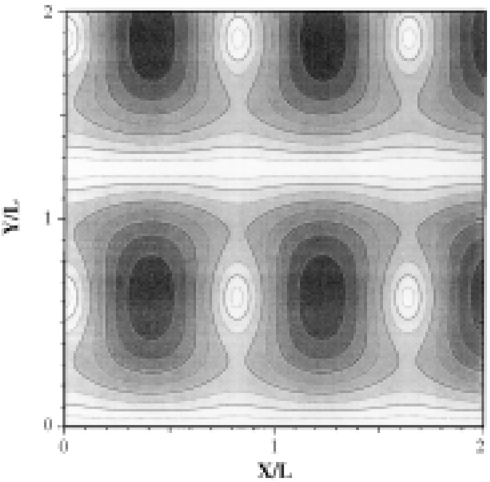

Fig. 2

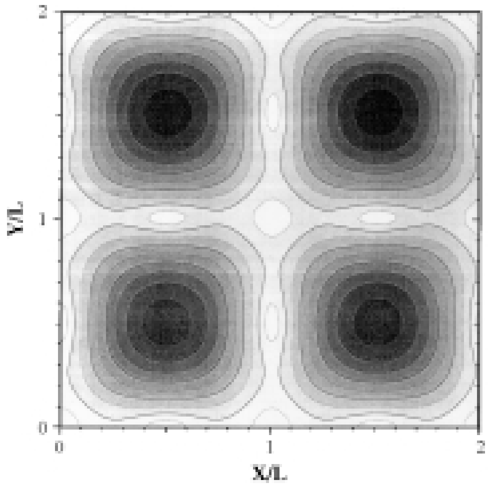

Fig. 3

Fig. 2 – The amplitude of the order parameter is represented for the asymmetric lattice at $n = 1$, $\zeta = 1.52i$. It is minimum in white zones and maximum in dark zones.

Fig. 3 – The square lattice at $n = 1$.

case, $\widetilde{\beta}_{\mathrm{A}}$ seems to be always positive and the transition is thus second order. However, the evolution from the two-dimensional functions used here to the one-dimensional sinusoidally modulated order parameter $\psi \propto \sin qx$ of this limit (see [4], [7]) remains obscure.

In order to have a complete picture of the transition near TP, we complete the description by studying the effect of the sixth-order term in the scheme of a weakly first-order transition. With the same arguments as previously, we determine the free-energy functional at given temperature T and magnetic field H

$$\mathcal{F} \propto A(h - h_{\mathrm{c2}})\overline{|\psi|^2} + \widetilde{\beta}_{\mathrm{A}}(\overline{|\psi|^2})^2 + \widetilde{\gamma}_{\mathrm{A}}(\overline{|\psi|^2})^3 \,,$$

where $I_6 = \overline{|\psi|^6}/(\overline{|\psi|^2})^3$, and $\widetilde{\gamma}_{\mathrm{A}} = 0.011 I_6$. Note that we retain only the leading sixth-order term. Now the first-order transition takes place when $\widetilde{\beta}_{\mathrm{A}}$ is negative. At the temperature where it vanishes, we calculate the free energy when the magnetic field H is just under the critical one which is still H_{c2}: $\mathcal{F} \propto -2\,[A\,|h - h_{\mathrm{c2}}|]^{\frac{3}{2}}\,/3\sqrt{3\widetilde{\gamma}_{\mathrm{A}}}$. Thus, the most favorable configuration is realized when I_6 is minimum. When the temperature is decreased and $\widetilde{\beta}_{\mathrm{A}} < 0$, the first-order critical field $H_{\mathrm{c2}}^{(1)}$ is the maximum one following $(h_{\mathrm{c2}}^{(1)} - h_{\mathrm{c2}}) = \widetilde{\beta}_{\mathrm{A}}^2/4A\widetilde{\gamma}_{\mathrm{A}}$.

Such a calculation may be done at each level where the weakly first-order transition takes place. We only report the results at the level $n = 0$. Here, the transition to the superconducting transition becomes first order when the temperature goes down $\frac{2}{3}$, and the lattice remains triangular. (As an example, for the square lattice $I_6 = 1.50$, whereas for the triangular one $I_6 = 1.42$). The first-order critical line for this lattice crosses the second-order critical line for the $n = 1$ level at $t = -1.05$. Below this temperature, previous results on the second-order transition apply.

In conclusion, we have described the influence of simultaneous strong paramagnetic and weak orbital effects on 2D superconductors, in the framework of a Ginzburg-Landau–like theory. We have demonstrated that the FFLO state manifests by inducing a cascade of transitions, of the first and second order, by investigating the vortex structures among single quantized flux-per-cell lattices. We have found that the vortex structure may be quite different from the standard triangular lattice with apex angle 60°. Further calculations should include the possibility of multiquantized structures. Numerical calculations on the basis of the Eilenberger equations [15] are necessary to extend the domain of validity of the present work.

The Ginzburg-Landau framework we have used has quite limited conditions of validity. However, we stress again that in real compounds, only the first Landau-level states should appear on the critical line, and in such case we expect that our results give at least a qualitative view of what would happen. Note also that the cascade of transitions into the higher Landau-level states is possible only below the tricritical point at $T^* \approx 0.56 T_c$. The systems where the predicted effects may be observed are superconducting films, which must be in the clean limit (as recently reported in Be films [16]), as the FFLO state is sensitive to impurities. Quasi–two-dimensional organic superconductors of the κ-(BEDT-TTF)$_2$X family [17] may be good candidates too, provided that the energy of the interlayer coupling is smaller than their critical temperature T_c.

REFERENCES

[1] LARKIN A. I. and OVCHINNIKOV YU. N., *Sov. Phys. JETP*, **20** (1965) 762.
[2] FULDE P. and FERRELL R. A., *Phys. Rev. A*, **135** (1964) 550.
[3] SAINT-JAMES D., SARMA G. and THOMAS E. J., *Type II Superconductivity* (Pergamon, New-York) 1969.
[4] BURKHARDT H. and RAINER D., *Ann. Phys. (Leipzig)*, **3** (1994) 181.
[5] BULAEVSKII L. N., *Sov. Phys. JETP*, **38** (1974) 634.
[6] BUZDIN A. I. and BRISON J. P., *Europhys. Lett.*, **35** (1996) 707.
[7] BUZDIN A. I. and KACHKACHI H., *Phys. Lett. A*, **225** (1997) 341.
[8] ABRIKOSOV A. A., GORKOV L. P. and DZYALOSHINSKII I. E., *Methods of Quantum Field Theory in Statistical Physics* (Dover Publications, Inc.) 1963.
[9] MAKI M., *Physics*, **1** (1964) 127.
[10] BUZDIN A. I. and KULIC M., *J. Low Temp. Phys.*, **54** (1984) 203.
[11] ABRIKOSOV A. A., *Sov. Phys. JETP*, **5** (1957) 1174.
[12] LASHER L., *Phys. Rev. A*, **140** (1965) 523.
[13] EILENBERGER G., *Phys. Rev.*, **153** (1967) 584; EILENBERGER G., *Phys. Rev.*, **164** (1967) 628.
[14] ZHITOMIRSKII M. E. and LUKYANCHUK I. A., *Sov. Phys. JETP*, **74** (1992) 1046.
[15] EILENBERGER G., *Z. Phys.*, **214** (1968) 195.
[16] ADAMS P. W., HERRON P. and MELETIS E. I., *Phys. Rev. B*, **58** (1998) R2952.
[17] ZUO F., BROOKS J. S., MCKENZIE R. H., SCHLUETER J. A. and WILLIAMS J. M., *Phys. Rev. B*, **61** (2000) 750.