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Structure of the non-uniform Fulde–Ferrell–Larkin–Ovchinnikov state in 3D superconductors

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Abstract

We demonstrate that in 3D superconductors the transition between a normal phase and a non-uniform superconducting phase is always of the first order in the pure paramagnetic limit. We also determine the transition temperature and the structure of the modulated 'lattice' by means of the generalized Ginzburg–Landau functional near the tricritical temperature, and the exact Gorkov equations in the whole temperature interval. © 1999 Elsevier Science B.V. All rights reserved.

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1. Introduction

As it has been demonstrated a long time ago by Fulde and Ferrell [1] and Larkin and Ovchinnikov [2] (FFLO) at low temperatures, when the magnetic field is acting on the electron spins only, the transition from normal (N) to modulated superconducting state (FFLO) must occur. Due to this nonuniform superconducting state formation, the paramagnetic limit at $T=0$ becomes larger than the usual Chandrasekhar–Clogston value $H_p(0)=\Delta_o/\sqrt{2}\,\mu_B \simeq 0.707\Delta_o/\mu_B$ [3], where $\Delta_o = 1.76T_c$ is the superconducting gap at $T=0$. The phase (H,T)-diagram for 3D superconductors was obtained by Saint-James et al. [3], assuming that the transition N → FFLO is of the second order. It occurs that the FFLO state appears only at $T<T^* \approx 0.56T_c$ [3], and the critical field temperature dependence is strongly influenced by the dimensionality of the system: for 3D superconductors: $H_{3D}^{FFLO}(0) = 0.755\Delta_o/\mu_B$ [1,2], for 2D superconductors it is larger: $H_{2D}^{FFLO}(0) = \Delta_o/\mu_B$ [4], and it diverges for 1D superconductors [5].

Up to now, there are no conclusive experimental evidences of the FFLO state formation (maybe except U Be_{13} [6], but for this heavy-fermion superconductor, the application of the standard theory of superconductivity is questionable). The main reason of the difficulties of experimental observation of such state is the orbital effect which is usually more important than the paramagnetic one. And actual critical field is determined mainly by the orbital effect. However, for heavy-fermion and low-dimensional superconductors (when the field is applied parallel to planes or chains) the orbital effect can be

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very weak, and we deal with paramagnetically limited critical field. The magnetic superconductors where the paramagnetic effect is strongly amplified by the internal exchange field are also good candidates for the FFLO state observations.

The problem of the exact structure of the FFLO state is not solved yet, even in the framework of the model of pure paramagnetic limit (except the 1D case where the superconducting order parameter in the FFLO state is described by the Jacobi elliptic function [5,7]). In this paper, we concentrate on the description of the 3D case, and we compare the four possible structures for the FFLO state: (i) the state with simple exponential modulation of the order parameter $\psi=|\psi_o|e^{iqx}$, (ii) the state with a simple cosine structure $\psi=|\psi_o|\cos qx$, (iii) the 2D 'lattice' state with $\psi\sim(\cos qx+\cos qy)$, and similarly (iv) the 3D 'lattice' state.

In Section 2, we study the region near the tricritical point $(T^*,H(T^*))$, where a generalized Ginzburg–Landau (GL) theory has been formulated [8]. We demonstrate that the first order transition temperature into 1D 'lattice' structure is slightly higher than transition temperatures into 2D and 3D 'lattice' states (the difference is of the order of 0.1% only). Near the tricritical point we are able to find the exact structure of the FFLO state, and we see that at the first order transition line N–FFLO state, it is very close to simple cosine structure, while as the temperature lowers this structure gradually transforms into the soliton-lattice structure.

However, as $T\to 0$, the transition into the cosine state becomes of the second order [9] while the results of Ref. [2] demonstrate that at $T=0$, the transition into 3D 'lattice' state is of the first order. This proves that such a state has a higher critical field and lower energy compared to the cosine one. In Section 3, we apply the exact Gorkov equations [10] for intermediate temperatures between 0 and T^*, and we show the possibility of a change in the modulation structure along the first order N/FFLO transition line.

2. Structure of the FFLO state near the tricritical point

First, we concentrate on the description of the FFLO phase near the tricritical point, where the characteristic wave vectors of the FFLO state are small compared with the inverse superconducting coherence length ξ_o^{-1}. For this region, the generalized GL theory may be formulated [8]. In contrast to the standard case, we need to take into account terms with a second order derivative of the order parameter $\sim(\psi'')^2$ as well as terms $\sim\psi^2(\psi')^2$ and ψ^6. The non-uniform state appearance is related with the change of a sign of the coefficient β at the gradient term $\beta|\nabla\psi|^2$. In the standard GL functional, β is positive, but it occurs to be a function of the field acting on the electron spins (paramagnetic effect), and goes to zero at $(T^*,H(T^*))$, being negative at $T<T^*$. Negative coefficient β means that the modulated state has lower energy compared with the non-uniform one. To find the modulation vector, it is needed to incorporate into the GL functional the term with a second order derivative. In addition, in the BCS theory, simultaneously with the vanishing of a gradient term, the coefficient γ at the fourth order $\gamma\psi^4$ vanishes too [3]. Due to this particular property, it is needed to add the higher order terms $\sim\psi^2(\psi')^2$ and ψ^6.

For a 3D superconductor in the paramagnetic limit, the generalized GL free energy density reads

$$F=\alpha|\psi|^2+\beta|\partial\psi|^2+\gamma|\psi|^4+\delta|\partial^2\psi|^2+\mu|\psi|^2|\partial\psi|^2+\eta\left[(\psi^+)^2(\partial\psi)^2+\psi^2(\partial\psi^+)^2\right]+\nu|\psi|^6, \tag{2.1}$$

where the coefficients are:

$$\alpha=-\pi N(0)(K_1-K_1^o),\quad \beta=\frac{\pi N(0)v_F^2K_3}{12},$$

$$\gamma=\frac{\pi N(0)K_3}{4},\quad \delta=-\frac{\pi N(0)v_F^4K_5}{80},$$

$$\nu=-\frac{\pi N(0)K_5}{8},\quad \mu=8\eta=-\frac{\pi N(0)v_F^2K_5}{6}.$$

v_F is the Fermi velocity, $N(0)$ the electron density of state, and

$$K_n(T)=2T\,\mathrm{Re}\left(\sum_{\nu=0}^{\infty}\frac{1}{(\omega_\nu-i\mathscr{H})^n}\right),\quad n\geqslant 1.$$

$\mathscr{H} = \mu_B H$, $\omega_\nu = (2\nu + 1)\pi T$ are Matsubara's frequencies at temperature T. Let $\mathscr{H}_o$ be the field corresponding to the second-order transition into the uniform superconducting state (U) at T, then we may set $\mathscr{H} = \mathscr{H}_o$ for all the coefficients except

$$\alpha \approx N(0)\frac{(\mathscr{H} - \mathscr{H}_o)}{2\pi T}\left|\mathrm{Im}\Psi'\left(\frac{1}{2} - i\frac{\mathscr{H}_o}{2\pi T}\right)\right|. \quad (2.2)$$

At the N/U first order transition, a constant order parameter ψ_o appears. The free energy (Eq. (2.1)) is $F = \alpha|\psi_o|^2 + \gamma|\psi_o|^4 + \nu|\psi_o|^6$. It is minimum in the U state, and vanishes at the transition, i.e., when $|\psi_o|^2 = -\gamma/2\nu$ and $\alpha_o^I = \gamma^2/4\nu$. If we choose the exponential order parameter $\psi(\boldsymbol{x}) = \psi_o e^{iqx}$, then the free energy reads $F = (\alpha + \beta q^2 + \delta q^4)|\psi_o|^2 + [\gamma + (\mu - 2\eta)q^2]|\psi_o|^4$. Analyzing the coefficient of $|\psi_o|^2$, we see that the field corresponding to the second order N/FFLO transition depends on the wave vector amplitude q, and that the actual field is the maximum one. Accordingly, we get $q_{max}^2 = -\beta/2\delta$ and $\alpha_o = \beta^2/4\delta = (10/9)\alpha_o^I$. Then we see that the coefficient on the quartic term is positive, thus indicating that the transition is of the second order. As $\alpha_o > \alpha_o^I$, this transition is more favorable than the N/U one. However, for the one, two, and three component cosine structures considered in Section 1, the same coefficient is negative, and consequently the actual N/FFLO transition is of the first order. In order to find the optimum state we need to add the sixth order term in the free energy, and study it after introducing following normalizations: $\psi = f\psi_o$, $x = \tilde{x}/q_{max}$, $C = -\gamma^3/8\nu^2$, and $\tilde{\alpha} = \alpha/\alpha_o^I$ is the only free parameter, we call it the 'external field'. The free energy per unit volume is

$$\tilde{F} = \frac{F}{C} = \frac{1}{\tilde{L}^3}\int\int\int_{[0,\tilde{L}]^3} \mathrm{d}\tilde{x}\,\mathrm{d}\tilde{y}\,\mathrm{d}\tilde{z}\left\{\tilde{\alpha}|f|^2 - \frac{20}{9}\left|\tilde{\nabla}f\right|^2 - 2|f|^4 + \frac{10}{9}\left|\tilde{\Delta}f\right|^2 + \frac{50}{9}|f|^2\left|\tilde{\nabla}f\right|^2 + |f|^6\right\}. \quad (2.3)$$

The dimensionless order parameter can be developed into a Fourier series $f = \sum_{\tilde{q}} a_{\tilde{q}} e^{i\tilde{q}\tilde{x}}$. We generate three- (respectively two-, one-) dimensional f functions with wave vectors $\tilde{\boldsymbol{q}} = 2\pi(n\hat{x} + m\hat{y} + p\hat{z})/\tilde{L}$ (respectively $\tilde{\boldsymbol{q}} = 2\pi(n\hat{x} + m\hat{y})/\tilde{L}$, $\tilde{\boldsymbol{q}} = 2\pi n\hat{x}/\tilde{L}$) where n, m, p are integers. Then we put this form for f with a finite number of harmonics ($|n|,|m|,|p| < 20$) in Eq. (2.3) and minimize with

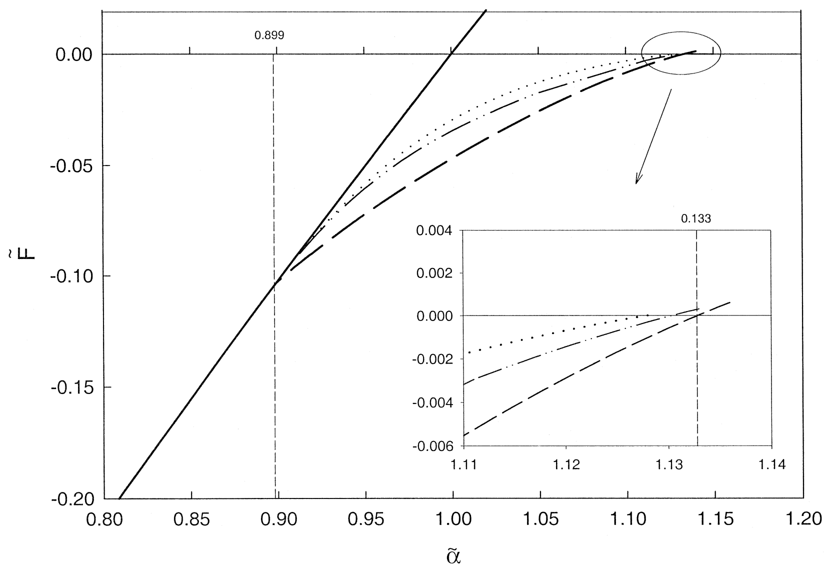

Fig. 1. Free energy as a function of the external field in the U phase (solid line), and in the FFLO phase for one-dimensional (dashed line), two-dimensional (dash–dotted line) and three-dimensional (dotted) structure of the order parameter.

respect to the amplitude of the harmonics and $\tilde{L}$. The free energy is finally drawn as a function of the external field. We compare it with the equilibrium free energy for the U state

$$\tilde{F}=\frac{2}{27}\left[9\tilde{\alpha}-8-(4-3\tilde{\alpha})\sqrt{4-3\tilde{\alpha}}\right].$$

In Fig. 1, we represent the $\tilde{F}$ free energy for each considered form of the order parameter. Among the modulated structures, the one-dimensional structure is always more favorable. The N/FFLO transition takes place at $\tilde{\alpha}=1.133$ and appears to be the slightly first order transition, whereas all the free energies converge at $\tilde{\alpha}=0.899$. The latter marks the FFLO/U transition which is of the second order. In Fig. 2, we show the period of the order parameter as a function of the external field in the FFLO phase. It diverges at FFLO/U transition. The order parameter can hardly be represented by a sinusoidal function unless near N/FFLO transition, and when the field decreases, the FFLO phase transforms into soliton-lattice phase. In Fig. 3, the form of the order parameter is represented for various external fields. When the modulation of the structure was one-dimensional, we could confirm all our results by using of a Newton–Raphson method in order to calculate directly the function which minimizes free energy (Eq. (2.3)).

3. Structure of the FFLO state at intermediate temperatures

In this section, we propose a scheme to calculate the first order transition line at every temperature when the transition is the slightly first order transition. It seems to be a reasonable assumption as we remind that the relative difference between the first order N/U and the second order N/FFLO transition lines on (H,T)-diagram was already only 5% [3]. It is also the situation near the tricritical point, as shown in Section 2. So, we suppose that the transition can be treated perturbatively.

We start with Gorkov equations [10], neglecting the orbital effect but taking into account the paramagnetic one where the electron spins interact with the magnetic field $\boldsymbol{H}$. The electron g-factor is assumed to be equal to 2. The axis of quantification is chosen along the magnetic field. Then, the order parameter $\Delta(\boldsymbol{r})$ is defined by a self-consistency equation, and it may vary spatially. When $\Delta(\boldsymbol{r})$ is small enough, the

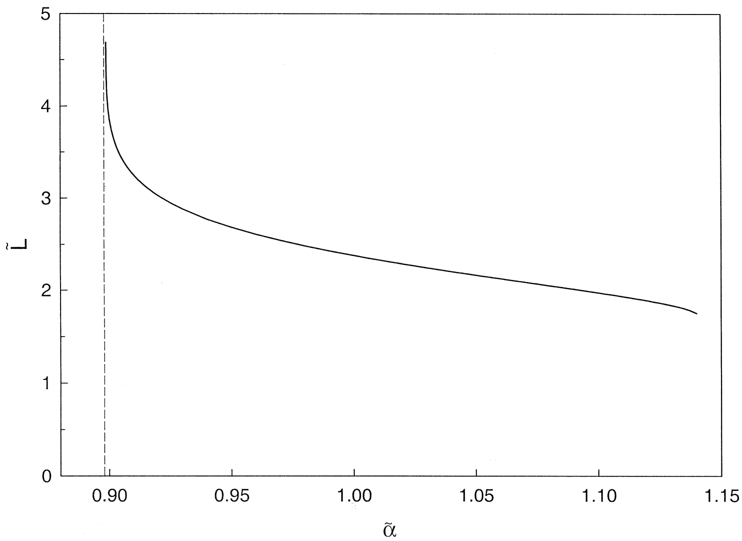

Fig. 2. Variation of the dimensionless period $\tilde{L}=2\pi/\tilde{q}=2\pi/q\sqrt{-\beta/2\delta}$ as a function of the external field. Note that the period L is also field dependent.

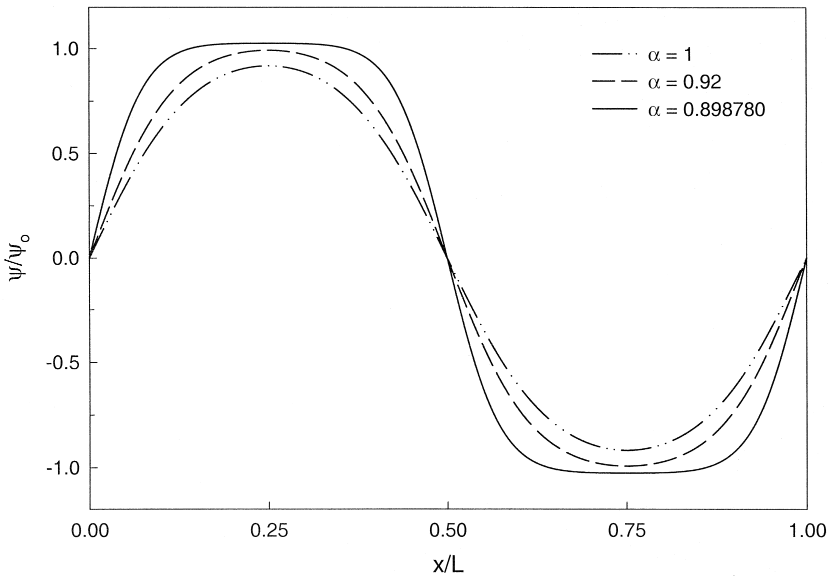

Fig. 3. Spatial dependance of the superconducting order parameter in the FFLO phase in one-dimensional structure for different fields—parameters α.

equation can be developed into a series on the Fourier components of $\Delta(\boldsymbol{r})$, which may be non-zero only for wave vectors $\boldsymbol{q}_i$ of the same amplitude q. Up to the fifth order, we obtain

$$\Delta_{\boldsymbol{q}}^* = K(q,H,T)\,\Delta_{\boldsymbol{q}}^* - \sum_{\boldsymbol{q_1}-\boldsymbol{q_2}+\boldsymbol{q_3}=\boldsymbol{q}} J_4\left(q,H,T;\hat{q}_1,\hat{q}_2,\hat{q}_3,\hat{q}\right) \Delta_{\boldsymbol{q1}}^*\Delta_{\boldsymbol{q2}}\Delta_{\boldsymbol{q3}}^* - \sum_{\boldsymbol{q_1}-\boldsymbol{q_2}+\boldsymbol{q_3}-\boldsymbol{q_4}+\boldsymbol{q_5}=\boldsymbol{q}} J_6\left(q,H,T;\hat{q}_1,\hat{q}_2,\hat{q}_3,\hat{q}_4,\hat{q}_5,\hat{q}\right) \times\Delta_{\boldsymbol{q1}}^*\Delta_{\boldsymbol{q2}}\Delta_{\boldsymbol{q3}}^*\Delta_{\boldsymbol{q4}}\Delta_{\boldsymbol{q5}}^*, \quad (3.1)$$

where the linear kernel is

$$K(q,H,T) = T|\lambda|\sum_{\omega_\nu}\int\frac{\mathrm{d}^3p}{(2\pi)^3} \times G_-^{(\mathrm{o})}\left(-\omega_\nu,\boldsymbol{p}+\frac{\boldsymbol{q}}{2}\right)G_+^{(\mathrm{o})}\left(\omega_\nu,-\boldsymbol{p}\frac{\boldsymbol{q}}{2}\right),$$

and

$$J_4 = T|\lambda|\sum_{\omega_\nu}\int\frac{\mathrm{d}^3p}{(2\pi)^3}G_-^{(\mathrm{o})}(-\omega_\nu,\boldsymbol{p}) \times G_+^{(\mathrm{o})}(\omega_\nu,-\boldsymbol{p}+\boldsymbol{q}_1) \times G_-^{(\mathrm{o})}(-\omega_\nu,\boldsymbol{p}-\boldsymbol{q}_1+\boldsymbol{q}_2) \times G_+^{(\mathrm{o})}(\omega_\nu,-\boldsymbol{p}+\boldsymbol{q}),$$

$$J_6 = -T|\lambda|\sum_{\omega_\nu}\int\frac{\mathrm{d}^3p}{(2\pi)^3}G_-^{(\mathrm{o})}(-\omega_\nu,\boldsymbol{p}) \times G_+^{(\mathrm{o})}(\omega_\nu,-\boldsymbol{p}+\boldsymbol{q}_1) \times G_-^{(\mathrm{o})}(-\omega_\nu,\boldsymbol{p}-\boldsymbol{q}_1+\boldsymbol{q}_2) \times G_+^{(\mathrm{o})}(\omega_\nu,-\boldsymbol{p}+\boldsymbol{q}_1-\boldsymbol{q}_2+\boldsymbol{q}_3) \times G_-^{(\mathrm{o})}(-\omega_\nu,\boldsymbol{p}-\boldsymbol{q}_1+\boldsymbol{q}_2-\boldsymbol{q}_3+\boldsymbol{q}_4) \times G_+^{(\mathrm{o})}(\omega_\nu,-\boldsymbol{p}+\boldsymbol{q}),$$

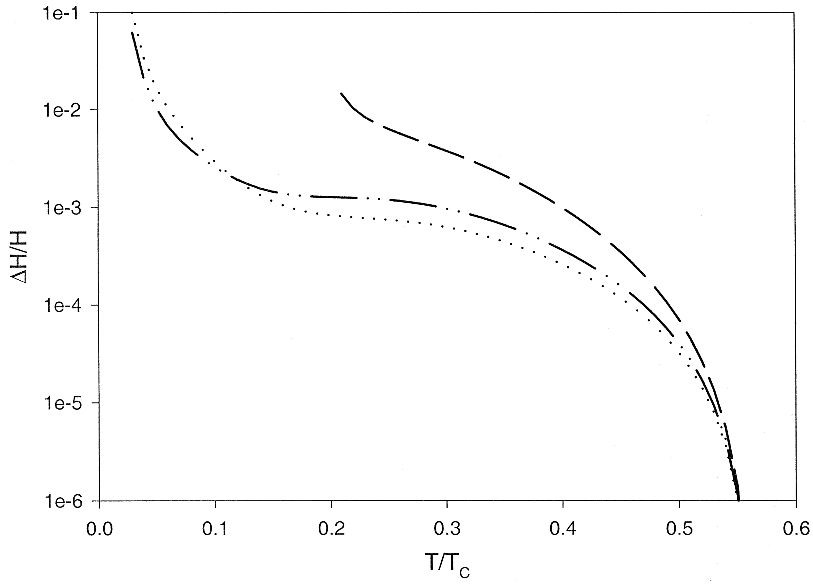

Fig. 4. Relative rise (in logarithmic scale) of the critical field of the first order N/FFLO transition vs. temperature, for different structures of modulation of the order parameter (dashed line is for 1D 'lattice' structure, dash–dotted line for 2D structure and dotted line for 3D structure), according to formula (3.3).

λ is the BCS coupling constant, and $G_{\pm}^{(o)}$ unperturbed Green's functions for spin up $(+)$ or down $(-)$ in the momentum representation

$$G_{\pm}^{(o)}(\omega_\nu,\boldsymbol{p}) = \frac{1}{i\omega_\nu - \xi_{\boldsymbol{p}} \pm \mu_B H}, \quad \xi_{\boldsymbol{p}} = \frac{p^2}{2m} - \varepsilon_F.$$

The free energy difference between N and FFLO phases is straightforwardly derived from Eq. (3.1)

$$\begin{aligned} F \propto & \sum_{\boldsymbol{q}} \tfrac{1}{2}(1-K(q,H,T))\,\Delta_{\boldsymbol{q}}^*\Delta_{\boldsymbol{q}} \\ & + \sum_{\boldsymbol{q}_1-\boldsymbol{q}_2+\boldsymbol{q}_3=\boldsymbol{q}} \tfrac{1}{4}J_4\,\Delta_{\boldsymbol{q}_1}^*\Delta_{\boldsymbol{q}_2}\Delta_{\boldsymbol{q}_3}^*\Delta_{\boldsymbol{q}} \\ & + \sum_{\boldsymbol{q}_1-\boldsymbol{q}_2+\boldsymbol{q}_3-\boldsymbol{q}_4+\boldsymbol{q}_5=\boldsymbol{q}} \tfrac{1}{6}J_6\,\Delta_{\boldsymbol{q}_1}^*\Delta_{\boldsymbol{q}_2}\Delta_{\boldsymbol{q}_3}^*\Delta_{\boldsymbol{q}_4}\Delta_{\boldsymbol{q}_5}^*\Delta_{\boldsymbol{q}}. \end{aligned} \tag{3.2}$$

We introduce dimensionless variables $\delta = \Delta/(\mu_B H)$, $j_4 = J_4(\mu_B H)^2$, $j_6 = J_6(\mu_B H)^4$. If we only consider the spatial modulations (i)–(iv) introduced in Section 1, then we can reduce Eqs. (3.1) and (3.2) to

$$\delta = K(q,H,T)\,\delta - j_4\delta^3 - j_6\delta^5,$$

$$F \propto (1-K(q,H,T))\frac{\delta^2}{2} + j_4\frac{\delta^4}{4} + j_6\frac{\delta^6}{6}.$$

where j_4 and j_6 now depend on q, H, T, and certainly on the geometry of the modulation.

The second order N/FFLO transition line is found by setting $K(q, H, T) = 1$, and maximizing H with respect to q. Solutions with $q \neq 0$, and forthcoming existence of the FFLO phase, are only found for $0 < T < T^* = 0.561T_c$, and we obtain the critical magnetic field $H(T)$ and wave vector $q(T)$. We then insert corresponding $q(T)$, $H(T)$ at temperature T inside j_4 and j_6, which are expected to vary smoothly. On the contrary, a significative variation is given by the coefficient of the quadratic term in an order parameter amplitude, in the free energy, near the second order transition line, and is approximated by

$$1 - K(q,H,T) \simeq \left(H_{\text{2nd order}}^{\text{FFLO}} - H\right)\left(\frac{\partial K}{\partial H}\right)_{q,T,\,\text{2nd order}},$$

as the q-variation is quadratic. For a given geometry, we can then predict the second order transition if $j_4 > 0$ and $j_6 > 0$, and the first order transition if $j_4 < 0$. In that case, the transition occurs when the energy difference vanishes. It is the weakly first order if we do not need the higher order coefficients, that is $j_6 > 0$, and $j_4/j_6 \ll 1$. Then, δ jumps from zero in the normal phase into $\sqrt{\frac{3}{4}\frac{j_4}{j_6}}$ in the FFLO phase. In such a case, the relative change of critical magnetic field at temperature T between the weak first order and the second order N/FFLO transition is

$$\frac{H^{\mathrm{FFLO}}_{\mathrm{1st\ order}} - H^{\mathrm{FFLO}}_{\mathrm{2nd\ order}}}{H^{\mathrm{FFLO}}_{\mathrm{2nd\ order}}} = \frac{\dfrac{3}{16}\dfrac{j_4^2}{j_6}}{H^{\mathrm{FFLO}}_{\mathrm{2nd\ order}}\left|\dfrac{\partial K}{\partial H}\right|_{q,T,\,\mathrm{2nd\ order}}} > 0. \quad (3.3)$$

For each geometry, j_4 and j_6 have been computed by numerical integration for temperatures neither too low nor too close to T^* (see Fig. 4). In cases (iii) and (iv), $j_4 < 0$ and $j_6 > 0$ at every temperature, and the condition $j_4/j_6 \ll 1$ is fulfilled. We can then apply the formula (3.3). We find that first order transition lines are higher than the second order one and join this last one when the temperature reaches T^*. The relative jump of critical magnetic field is never higher than 3%, and the lines cross at a temperature $T'' = 0.12T_c$. This means that modulation (iv) is more favorable than (iii) at $T < T''$, whereas it is the contrary at $T > T''$. In case (ii), we find again the sign change of j_4 at $T' = 0.132T_c$, in accordance with Ref. [9]. However, for this solution, the coefficient j_6 is positive only near T^* and then changes its sign. We cannot consider anymore that the first order transition is weak at all temperatures. We can just find that near T^*, when the first order transition into the structure (ii) is weak enough, it is more favorable than structures (iii) or (iv), the difference between the second order transition line and first order transition lines is becoming still smaller and smaller when the temperature approaches T^*. Let us note that it is coherent with the results of Section 2 in the vicinity of the tricritical point.

4. Conclusion

We then propose the following picture for the N/FFLO transition in 3D pure paramagnetic superconductors, (see Fig. 5). It is a first order one. Near

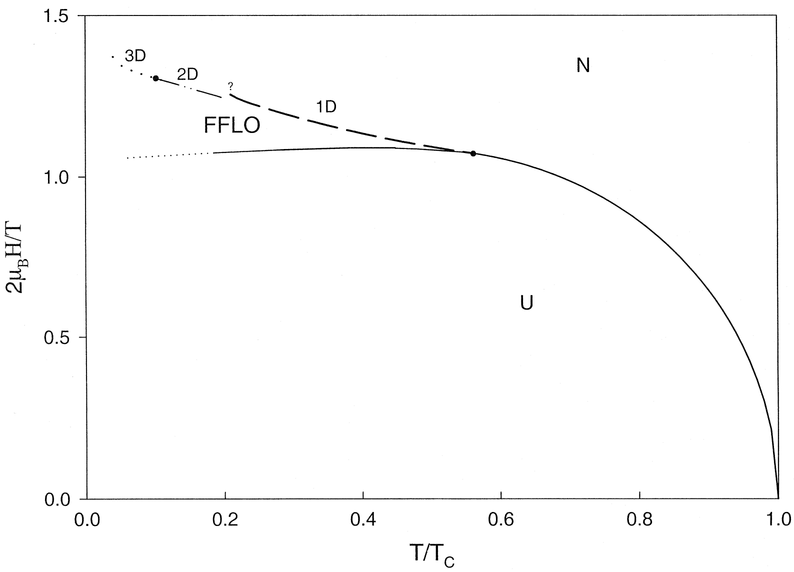

Fig. 5. Phase diagram of a 3D superconductor, in the paramagnetic limit. Solid line corresponds to the line transition to the uniform phase, dashed line is for a transition to a 1D 'lattice' structure, dash–dotted line to a 2D structure and dotted line to a 3D structure.

T^*, it provokes a slight enhancement of the transition line compared to the second order transition line. It is likely to lead to various spatial structures of the modulation: near T^*, the structure of the modulation is one-dimensional, when the temperature is lowered the modulation at the appearing of the FFLO phase becomes two-dimensional, then three-dimensional. However, the calculations we have done are not complete. Firstly, because we have not considered other spatial modulations which may have proved more stable. Secondly, because we have restricted ourselves to a weak first order transition scheme out of which the difficulty of the problem would be much more increased. Thus, the possibility of a strongly first order N → FFLO transition seems quite probable, for instance in the one-dimensional cosine structure. Whereas, we have demonstrated that N-FFLO transition is always a first order one and provided analysis of stability of different structures near T_c, more complete numerical calculations based on Eilenberger equations [11] are needed to find exact FFLO state structures at low temperatures.

Acknowledgements

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