

# Spontaneous vortex state and ferromagnetic behavior of type-II $p$ -wave superconductors

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The mixed phase in type-II superconductors with equal spin  $p$ -wave pairing is considered using the Ginzburg-Landau approach. Due to direct spin coupling of the condensate to magnetic field the mixed state acquires ferromagnetic properties. For sufficiently large Zeeman coupling a spontaneous vortex phase appears at  $H=0$  and exists for an arbitrarily large magnetic field. The Meissner phase therefore completely disappears. Vortices become thinner when  $H$  grows. There exists a value of Zeeman coupling above which, in the presence of external magnetic field, a mixed phase might occur even for temperatures above  $T_c$ . The structure of the vortex core is markedly different from the usual one. [S0163-1829(98)06137-2]

## I. INTRODUCTION

In the majority of conventional low- $T_c$  superconductors pairing occurs in the  $s$  channel. In this case the Cooper pair does not have total spin and an external magnetic field influences superconductors via coupling to orbital motion of the pairs only. Although high- $T_c$  cuprites are most probably  $d$ -wave superconductors, direct magnetic coupling to the spin of the Cooper pair should be still insignificant.<sup>1</sup> The situation might be different in certain cases of  $p$ -wave pairing. The magnetic field violates time-reversal invariance and is an extremely effective pair breaker for  $s$ -,  $d$ -, and certain  $p$ -wave states,<sup>2</sup> but it does not break pairs with parallel spins of constituent fermions.

The  $p$ -wave pairing is suspected to occur in a recently discovered new class of Ru-based superconductors  $\text{Sr}_2\text{YRu}_{1-x}\text{Cu}_x\text{O}_6$ .<sup>3</sup> At the same temperature of about 60 K, at which superconductivity sets in, these materials begin to exhibit basic ferromagnetic properties like hysteresis loop. Experimental observation of a positive remanence suggests the existence of spontaneous magnetization in the absence of an external magnetic field. Various conventional sources of ferromagnetism, independent from but coexisting with superconductivity,<sup>4,5</sup> cannot be ruled out. However, exact overlap of superconductivity and ferromagnetism naturally suggests that in these particular materials Cooper pairs might in fact be magnetic moments and that they themselves are responsible, at least partially, for overcoming the usual diamagnetic response of the superconductor. Of course, in principle, the critical temperatures of transition to ferromagnetic and superconducting states can simply accidentally coincide, but the "same-mechanism" scenario is nevertheless worth taking a look at.

In this paper we explore in some detail this possibility in the case of type-II superconductors using the phenomenological Ginzburg-Landau (GL) approach. Superconductors obtained by Wu and collaborators<sup>3</sup> are believed to be of type II, similar to high- $T_c$  copper oxides, although at present their unusual magnetic properties introduce ambiguities in stan-

dard direct methods of measurement of coherence length and penetration depth. Our study turned out to be interesting in its own right (even with no direct relation to the above experiment), since the vortex-matter physics happens to be quite nonstandard. The same GL equations might describe other physical systems. Microscopic derivation of GL equations for  $p$ -wave pairing has been recently performed by Xu *et al.*<sup>6</sup> in connection to  $\text{Sr}_2\text{RuO}_4$ .

Within the framework of the phenomenological GL theory nonzero spin of the Cooper pair is taken into account by introducing an order parameter of the vector type. It is directly coupled to an internal magnetic field through a Zeeman-like term in the free energy. An external magnetic field penetrates a type-II superconductor via creation of vortices in the bulk of the sample. Formation of each vortex is accompanied by both the energy loss due to vortex line formation, and the energy gain due to the energy of the penetrating field itself. Now, however, it is also accompanied by an additional energy gain due to direct interaction of the penetrating field with the spin of the condensate. If this interaction is sufficiently strong, the total line energy of a vortex can become negative and consequently instability develops. The usual hexagonal lattice vortex structure will form even for the system at zero external magnetic field. This state can be characterized as a spontaneous vortex state.<sup>7</sup> At the same time there appears a ferromagnetic moment of the superconductor associated with the bulk of the condensate that prevails over the conventional diamagnetic moment due to screening by supercurrents.

In Sec. II we formulate the model and investigate its main features. As in heavy-fermion systems,<sup>8</sup> there are two quartic terms in the GL free energy and there exist two qualitatively different superconducting states. One has rather conventional magnetic behavior and we concentrate on the more interesting state allowing ferromagnetism. Different types of  $H$ - $T$  phase diagrams are possible for different strengths of Zeeman coupling. Anticipating the derivation, we present the diagrams in Fig. 1. When the direct spin coupling increases, the magnetic behavior of the superconductor changes dra-

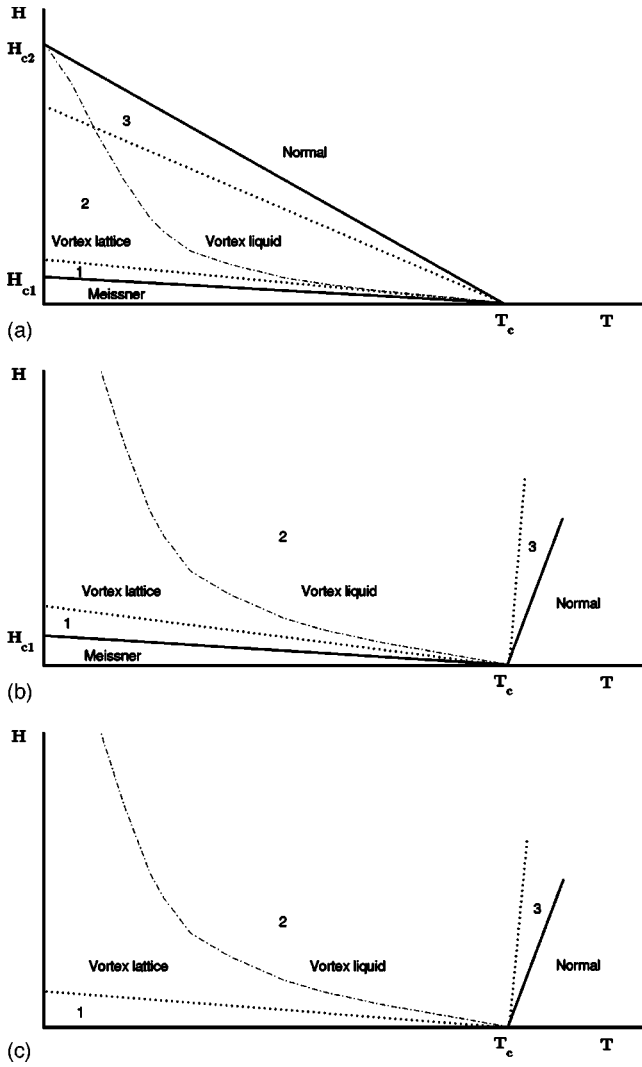


FIG. 1. Phase diagrams of equal spin  $p$ -wave superconductor with different strength of Zeeman coupling  $g$ : (a)  $g < g_{c2}$ , (b)  $g_{c1} < g < g_{c2}$ , (c)  $g > g_{c1}$ . The solid lines present lines of transition to the Meissner state and the normal state. The dashed line is vortex lattice melting line (not calculated in the present paper). Dotted lines are boundaries of validity of different approximations used.

matically: from essentially diamagnetic, even perfectly diamagnetic in the Meissner phase [Fig. 1(a)], to essentially ferromagnetic [Fig. 1(c)]. At nonzero magnetic field a mixed state can exist even beyond  $T_c$  because Zeeman coupling facilitates creation of the Cooper pairs along with the usual destruction of the condensate [Figs. 1(b) and 1(c)]. For large, but still of a quite realistic order of magnitude of the coupling, the Meissner phase disappears completely [Fig. 1(c)]. Below  $T_c$ , a spontaneous vortex phase appears at  $H=0$  and exists for an arbitrarily large magnetic field.

In Sec. III the model is studied using a London approximation. Transition to the normal state is studied in Sec. IV. In Sec. V we find the single-vortex solution numerically. The vortex core structure is quite different from that of an usual Abrikosov vortex in the  $s$ -wave superconductors. The phase diagram and the magnetization curve are also qualitatively calculated and discussed beyond London approximation. Vortices become thinner when  $H$  grows making room for

more vortices to squeeze in. Conclusions and some generalizations are discussed in Sec. VI.

## II. GENERAL PHENOMENOLOGICAL FORMULATION

Within the general framework of the Ginzburg-Landau approach in unconventional superconductors,<sup>9</sup> the  $p$ -wave pairing in an isotropic material is described by the order parameter  $\psi_i$  that has three complex components:  $i=1,2,3$ . In the absence of magnetic field the free-energy density is

$$F = \frac{\hbar^2}{2m^*} (\partial_j \psi_i) (\partial_j \psi_i)^* + F_{pot}, \quad (1)$$

$$F_{pot} = -\alpha \psi_i \psi_i^* + \frac{\beta_1}{2} (\psi_i \psi_i^*)^2 + \frac{\beta_2}{2} |\psi_i \psi_i|^2.$$

It has the following independent symmetries. The spin rotations, forming group  $SO_{spin}(3)$  act on the index of the order-parameter field, so that it transforms as a vector. Three-dimensional (orbital) space rotations forming different  $SO_{orbit}(3)$  group, act on spatial coordinates and the electric-charge transformations, forming the  $U(1)$  group, rotate the complex phase of the order parameter. Note two independent quartic terms. This is similar to that of heavy fermions (where order parameter usually has two components) or liquid  $^3\text{He}$  for which the order parameter is more complicated. The vacuum structure can be studied using methods developed in these fields.<sup>8,10</sup>

This free energy has been already considered by Burlachkov and Kopnin in connection with  $p$ -wave organic superconductors.<sup>11</sup> It is not, strictly speaking, the most general possible free energy for the  $p$  wave. Recently, an attempt has been made to derive the GL energy from reasonable microscopical models describing  $\text{Sr}_2\text{RuO}_4$ .<sup>6</sup> In principle, for strong spin-orbit coupling there is no separate spin rotation symmetry, just overall rotations. In this case one can add an additional two terms even for an isotropic superconductor.<sup>13</sup> It is sufficient, however, for the purposes of the present paper to consider free-energy (1) even not assuming weak spin-orbit coupling. Our results essentially depend only on the existence of Zeeman coupling in GL-free energy expansion. The Zeeman-like term that will be introduced below [see Eq. (6)] is also allowed by the symmetry considerations in the case of strong spin-orbit coupling as well as for weak breaking of full rotational symmetry by the crystal field. It is only necessary to have a nonunitary (broken time-reversal symmetry) superconducting phase with an order parameter that has at least two components. This generalization can be easily made.

We use the following convenient parametrization of the order parameter:

$$\psi = f(\mathbf{n} \cos \phi + i \mathbf{m} \sin \phi), \quad (2)$$

where  $f > 0$ ,  $\mathbf{n}$  and  $\mathbf{m}$  are arbitrary unit vectors and  $0 \leq \phi \leq \pi/2$ . Using this parametrization the homogeneous part of the free-energy density takes the form

$$F_{pot} = -\alpha f^2 + \frac{\beta_1}{2} f^4 + \frac{\beta_2}{2} f^4 (\cos^2 2\phi + (\mathbf{m}\mathbf{n})^2 \sin^2 2\phi), \quad (3)$$

which can be easily minimized to give the two phases. In phase I,  $\beta_2 > 0$ ,

$$\boldsymbol{\psi} = f \frac{\mathbf{n} + i\mathbf{m}}{\sqrt{2}}, \quad \mathbf{n} \perp \mathbf{m}, \quad \phi = \pi/4, \quad f^2 = \frac{\alpha}{\beta_1}, \quad (4)$$

while in phase II,  $\beta_2 < 0$ , various vacua are given by

$$\boldsymbol{\psi} = f e^{i\phi} \mathbf{n}, \quad \mathbf{n} = \pm \mathbf{m}; \quad f^2 = \frac{\alpha}{\beta_1 + \beta_2}. \quad (5)$$

In phase I both the spin-rotation  $\text{SO}_{spin}(3)$  symmetry and the superconducting phase  $\text{U}(1)$  symmetry are broken, but a diagonal subgroup  $\text{U}(1)$  survives. It consists of rotations by angle  $\theta$  around the axis  $\mathbf{n} \times \mathbf{m} \equiv \mathbf{l}$  that are accompanied by gauge transformations  $e^{i\theta}$ . The vacuum manifold in this phase is isomorphic to  $\text{SO}(3)$ . In phase II the superconducting  $\text{U}(1)$  symmetry is also broken, however the spin rotation  $\text{SO}_{spin}(3)$  is only partially broken down to its  $\text{SO}(2)$  subgroup. There is an additional unbroken discrete symmetry that consists of a simultaneous change of the gauge phase by  $\pi$  and change of the sign for  $\mathbf{n}$ . The vacuum manifold in phase II is therefore isomorphic to  $S_2 \otimes \text{U}(1)/Z_2$ . From Eqs. (4) and (5) it is seen that stability of phases is achieved for  $\beta_1 > 0$  in phase I and for  $\beta_1 + \beta_2 > 0$  in phase II. Energy densities of the condensate are  $-\alpha^2/2\beta_1$  and  $-\alpha^2/2(\beta_1 + \beta_2)$  correspondingly.

Gibbs free-energy density in the presence of electromagnetic interactions is

$$F = F_{pot} + F_{grad} + F_{magn} - \frac{B_i H_i}{4\pi} + \frac{H_i^2}{8\pi}, \quad (6)$$

$$F_{grad} = \frac{\hbar^2}{2m^*} (\mathcal{D}_j \psi_i) (\mathcal{D}_j \psi_i)^*,$$

$$F_{magn} = \frac{B_i^2}{8\pi} - \mu S_i B_i,$$

where  $\mathcal{D}_i \equiv \partial_i - i(e^*/c\hbar)A_i$ ,  $\text{rot } \mathbf{A} = \mathbf{B}$ . The second term in  $F_{magn}$  is the direct magnetic coupling to spin  $S_i \equiv -i\epsilon_{ijk}\psi_j^* \psi_k$  carried by the order-parameter field. Of course, due to this Zeeman-like term (even in the absence of external magnetic field), separate spin- and orbital-rotation symmetry groups are broken down to an overall rotation group:  $\text{SO}_{spin} \otimes \text{SO}_{orbit} \rightarrow \text{SO}_{rot}$ . In principle, one more independent gradient invariant is possible:<sup>8</sup>

$$(\mathcal{D}_j \psi_i) (\mathcal{D}_i \psi_j)^* + (\mathcal{D}_i \psi_i) (\mathcal{D}_j \psi_j)^*.$$

We will not introduce it and instead assume a larger symmetry even in the presence of external magnetic field. It is assumed that an external magnetic field is always oriented along the third ( $z$ ) direction:  $H_1 = H_2 = 0$ . In this case the free energy, Eq. (6) is still invariant under two-dimensional spin rotations  $\text{SO}_{spin}(2)$  in the  $xy$  plane, in addition to the orbital  $\text{SO}_{orbit}(2)$ . The additional derivative term does not respect this symmetry. We assume its coefficient to be small and thus avoid severe complications of considering noncylindrically symmetric vortices created by such a term. Its influence has been thoroughly studied in the context of heavy-fermion superconductors<sup>8</sup> and in the present context can be considered later perturbatively, similar to the recent treatment of the anisotropic  $d$ -wave situation.<sup>14</sup>

Using the parametrization of Eq. (2) allows us to make several interesting observations. We see that  $\mathbf{S} = f^2 \sin 2\phi \mathbf{n} \times \mathbf{m} \equiv f^2 \sin 2\phi \mathbf{l}$ . In phase I the projection of the spin of a Cooper pair  $\mathbf{S}$  on the quantization axis determined by the vector  $\mathbf{l}$  is equal to either  $+1$  or  $-1$ , reflecting spontaneous time-reversal symmetry breaking. In phase II this projection is equal to 0.

The gradient and magnetic parts of the free-energy density take form

$$F_{grad} = \frac{\hbar^2}{2m^*} (\partial_i f)^2 + \frac{\hbar^2}{2m^*} f^2 \left[ (\partial_i \phi)^2 + \cos^2 \phi (\partial_i \mathbf{n})^2 + \sin^2 \phi (\partial_i \mathbf{m})^2 - \frac{2e^*}{\hbar c} A_i \left( (\mathbf{m}\mathbf{n}) \partial_i \phi + \sin 2\phi \frac{\mathbf{n} \partial_i \mathbf{m} - \mathbf{m} \partial_i \mathbf{n}}{2} \right) + \left( \frac{e^*}{\hbar c} \right)^2 A_i^2 \right], \quad (7)$$

$$F_{magn} = \frac{1}{8\pi} \mathbf{B}^2 - \mu f^2 \sin 2\phi \mathbf{l} \mathbf{B}, \quad (8)$$

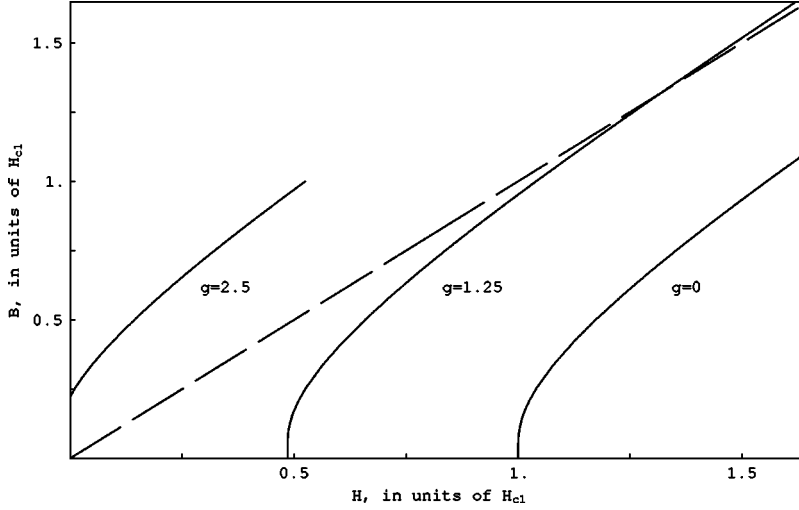


FIG. 2. Magnetic field  $B$  as a function of external magnetic field  $H$  for  $\kappa=10$  and different values of Zeeman coupling  $g$ .

from which the following equation for the field  $\mathbf{A}$  can be obtained:

$$\frac{e^*\hbar}{m^*c} f^2 \left( (\mathbf{n}\mathbf{m}) \partial_i \phi + \sin 2\phi \frac{\mathbf{n}\partial_i \mathbf{m} - \mathbf{m}\partial_i \mathbf{n}}{2} - \frac{e^*}{\hbar c} A_i \right) = \left( \frac{1}{4\pi} \nabla \times (\nabla \times \mathbf{A}) - \mu \nabla \times (f^2 \sin 2\phi \mathbf{l}) \right)_i. \quad (9)$$

The ‘‘supercurrent’’ Eq. (9) shows that superconducting velocity is given (in units of  $\hbar/m^*$ ) by  $\mathbf{n}\partial_i \mathbf{m}$  in phase I, while it is  $\partial_i \phi$  in phase II. Next we deduce, after integration along a closed contour, that the flux is quantized in units of  $\Phi_0 \equiv hc/e^*$  (for definiteness we assume that  $e^* > 0$ ). At this point it is convenient to introduce magnetic penetration depth  $\lambda \equiv (c/e^* f_0) \sqrt{m^*/4\pi}$ , where  $f_0$  should be taken from Eq. (4) for phase I or from Eq. (5) for phase II, and also coherence length  $\xi \equiv \hbar/\sqrt{2\alpha m^*}$  and dimensionless Ginsburg-Landau (GL) parameter  $\kappa \equiv \lambda/\xi$ .

The equation for the order-parameter field  $\psi$  reads

$$-\alpha \psi_i + \beta_1 (\psi_j \psi_j^*) \psi_i + \beta_2 (\psi_j \psi_j) \psi_i^* - \frac{\hbar^2}{2m^*} \mathcal{D}_j^2 \psi_i + i\mu \varepsilon_{ijk} \psi_j B_k = 0. \quad (10)$$

It is very bulky to be presented here in the parametrization of Eq. (2).

Being interested in the magnetic properties of a  $p$ -wave superconductor that arise because of direct Cooper-pair spin coupling to magnetic field, in the following sections we will concentrate on phase I only. Phase II is quite similar to usual  $s$ -wave superconductors in a sense that the condensate does not carry spin.

Equations (9) and (10) are difficult to solve analytically in the general case and we therefore have to resort to approximations. We will consider situations that possess translational symmetry along  $z$  axis, the direction of the magnetic field, and from now on spatial index  $i$  will take only two

values:  $i=1,2$ . This, in general, by no means precludes  $\mathbf{n}$  or  $\mathbf{m}$  to be oriented out of the 1-2 plane.

Two standard approaches are used in the following sections. The first (Sec. III) is for almost isolated vortices when distance between them  $a = \sqrt{\Phi_0/B}$  is larger than the magnetic penetration length  $\lambda$  (region 1 of  $H$ - $T$  diagram in Fig. 2). Here an analytic solution for an isolated vortex in the London approximation is used to show that a spontaneous vortex state can appear and to construct the magnetization curve. Then (Sec. IV) the opposite limit in which the superconductivity is very weak, i.e.,  $f \ll 1$ , allows linearization of the equations and can also be solved (region 3 in Fig. 1). Finally in Sec. V, a more complicated, but more important, intermediate regime is considered (region 2 in Fig. 1). Here different approximations and numerical methods are necessary.

### III. LONDON APPROXIMATION: DISAPPEARANCE OF MEISSNER PHASE

The London approximation assumes that the vacuum has the form that is determined by the homogeneous part of the free energy (3) almost everywhere except in some singular points. The approximation is applicable mainly in the case of the large GL parameter  $\kappa$ . In the presence of singularities, a vacuum state can gradually vary into another such state (still belonging the vacuum manifold defined earlier) and in this way a vortex is formed. The structure of the vortex core is outside of range of validity of this approach and will be investigated in Sec. V.

To derive the London equations we take the general vector equation for the superconducting current, Eq. (9), and substitute  $\psi$  in the form given by Eq. (4). This leads to

$$\frac{e^*\hbar}{m^*c} f_0^2 \left( \mathbf{n}\partial_i \mathbf{m} - \frac{e^*}{\hbar c} A_i \right) = \left( \frac{1}{4\pi} \nabla \times (\nabla \times \mathbf{A}) - \mu f_0^2 \nabla \times \mathbf{l} \right)_i, \quad (11)$$

where  $f_0^2 = \alpha/\beta_1$ .

To find the single-vortex solution, we impose, as usual, cylindrical symmetry and require appropriate behavior as  $r \rightarrow \infty$ , namely,  $\psi = (\mathbf{e}_x + i\mathbf{e}_y)\exp(iv\varphi)$ , or

$$\mathbf{n} = \mathbf{e}_x \cos v\varphi + \mathbf{e}_y \sin v\varphi,$$

$$\mathbf{m} = -\mathbf{e}_x \sin v\varphi + \mathbf{e}_y \cos v\varphi,$$

where  $\varphi$  is azimuthal angle and integer  $v$  stands for vorticity.  $\mathbf{l} \equiv \mathbf{n} \times \mathbf{m}$  always points upwards in the direction of external magnetic field so as to minimize the Zeeman term in the energy (8). This yields the equation for azimuthal component of the vector potential  $A(r)$ :

$$\frac{\hbar e^*}{m^* c} f_0^2 \left( \frac{v}{r} - \frac{e^*}{\hbar c} A \right) + \frac{1}{4\pi} \left( \ddot{A} + \frac{\dot{A}}{r} - \frac{A}{r^2} \right) = 0. \quad (12)$$

We observe that this equation that governs the behavior of a vortex in the  $p$ -wave superconductor is essentially the same as that for an  $s$ -wave superconductor. The solution is well known to be

$$A(r) = \frac{v\Phi_0}{2\pi\lambda} \left[ \frac{\lambda}{r} - K_1 \left( \frac{r}{\lambda} \right) \right].$$

Multiple vortices are unstable and we therefore set  $v = 1$ .

Let us now estimate the line tension of the vortex  $\varepsilon_V$  in the present case. Although Eq. (12) does not reflect the presence of Zeeman coupling  $-\mu S_i B_i$  of the spin to the magnetic field at all, the free energy does depend, of course, on  $\mu$ . Thus, the energetics changes considerably as compared to the  $s$ -wave vortex. Provided  $\mu > 0$ , the Zeeman term makes an additional negative ‘‘bulk’’ contribution  $\varepsilon_Z$  to the usual value of vortex line tension  $\varepsilon_L$ .<sup>15</sup> Summing up the two, one obtains

$$\varepsilon_V \equiv \varepsilon_L + \varepsilon_Z \approx \varepsilon_0 \ln \kappa - \mu \Phi_0 f_0^2 = \varepsilon_0 (\ln \kappa - g), \quad (13)$$

where the notation  $\varepsilon_0 = (\Phi_0/4\pi\lambda)^2$  was used and a convenient dimensionless parameter  $g$  was introduced by last equality. Since the relation

$$\mu = \frac{e^* \hbar}{2m^* c} g \quad (14)$$

holds,  $g$  can also be viewed as an effective Lande factor. However, in the present context, it is just a phenomenological parameter, in the spirit of the Ginzburg-Landau approach.

From Eq. (13) we see that lower critical field becomes renormalized and diminishes:

$$H_{c1}(g) = \frac{\Phi_0}{4\pi\lambda^2} (\ln \kappa - g). \quad (15)$$

Linear dependence of the lower critical field on  $g$  in Eq. (15) coincides with conclusions of earlier work by Tokuyasu *et al.*<sup>12</sup> Ultimately, in the case of

$$g \geq g_{c1} = \ln \kappa, \quad (16)$$

the line tension  $\varepsilon_V$  becomes negative and vortices will be created copiously until repulsive interaction between them overpowers the energy gain due to Zeeman coupling. This is

the most unusual situation in which the ground state of a superconductor at zero magnetic field spontaneously becomes inhomogeneous. Another situation of inhomogeneous ground state in the superconducting phase with spontaneously broken time-reversal symmetry was considered in connection to heavy-fermion systems by Palumbo *et al.*,<sup>16</sup> while inhomogeneous ground states of a superconductor described by a one-component order parameter in the presence of an external magnetic field were extensively studied by Vos *et al.*<sup>17</sup>

Peculiarity of the London approximation in our case results in the interesting fact that vortex interaction does not change at all as compared to the usual  $s$ -wave case<sup>15</sup> up to the values of external field where this approximation ceases to be valid. To show this we consider the free energy of collection of vortices separated by distances much larger than vortex core size. Additional contribution comes from the term  $-\mu L \int \mathbf{S} \mathbf{B} d^2x = -\mu L \int f^2 \mathbf{I} \mathbf{B} d^2x$ . It is equal to the total flux through the sample since, within the London approximation, the vortex core’s contribution to the integral is ignored. Flux is an additive function of the number of vortices and thus the above term does not influence the interaction between vortices.

As a straightforward consequence of such a lack of the interaction renormalization, we can get the  $B$ - $H$  curve of the vortex structure considered in weak fields  $B \approx \Phi_0/4\pi\lambda^2$  from that of the usual vortex structure simply by shifting the ordinate axis on the  $B$ - $H$  plot to the right by some amount proportional to  $\mu$  (see Fig. 2). A standard calculation<sup>18</sup> summing up  $z$  nearest neighbors ( $z = 6$  for triangular lattice) interactions and neglecting contributions of the cores gives

$$B = \frac{2\Phi_0}{\sqrt{3}\lambda^2} \left[ \ln \frac{4\pi\lambda^2}{3\Phi_0} [H - H_{c1}(g)] \right]^{-2}.$$

In the case  $g > g_{c1}$  the line, as expected, crosses the  $B = 0$  axis. This means that the Meissner phase completely disappears and the spontaneous vortex state is formed instead [see Figs. 1(b) and 1(c)]. The spontaneously created field, the remanence, is

$$B_r = \frac{2\Phi_0}{\sqrt{3}\lambda^2} \left[ \ln \frac{g - \ln \kappa}{3} \right]^{-2}. \quad (17)$$

This field rather than  $H_{c1}$  has a physical meaning under present circumstances. Now we turn to the opposite limit of situations in which the order parameter is small compared to its vacuum value.

#### IV. TRANSITION TO NORMAL STATE: ABSENCE OF $H_{c2}$ FOR $T < T_c$

The presence of strong Zeeman coupling should significantly modify also the transition to the normal state. When the order parameter becomes small, the quartic terms in the free-energy equations (1) and (6) are negligible compared to the quadratic ones. Simultaneously the magnetization becomes small and therefore one can replace  $B$  by  $H$  in the linearized equations. If the direction of the magnetic field is  $\hat{x}_3$ , the dependence of the order-parameter field on  $x_3$  has the form of a plane wave. It is also clear that the equation for  $\psi_3$  decouples and is the same as the usual  $s$ -wave equation. Its

eigenvalues are  $\alpha_{n3} = H(\hbar e^*/m^*c)(1/2+n)$ , where  $n$  is a natural number. Dependence of  $\psi_1, \psi_2$  on  $x_1, x_2$  is to be found from the following system of equations:

$$\frac{\hbar^2}{2m^*}(\mathcal{D}_1^2 + \mathcal{D}_2^2)\psi_i + \alpha\psi_i - i\mu H \varepsilon_{ij}\psi_j = 0, \quad (18)$$

where  $\mathcal{D}_j = \partial_j - i(e^*H/2\hbar c)\varepsilon_{ji}x_j$ ,  $\varepsilon_{ij}$  being the antisymmetric tensor and  $j, i = 1, 2$ . This coincides with the nonrelativistic Schrödinger equation for a spin 1/2 particle in magnetic field. It can be disentangled by changing variables to  $\psi_{\pm} = \psi_1 \pm i\psi_2$  and writing covariant derivative operators via  $\mathcal{D}_{\pm} = \mathcal{D}_1 \pm i\mathcal{D}_2$ . The eigenvalues are

$$\alpha_{n\pm} = H \frac{\hbar e^*}{2m^*c} (1 \pm g + 2n), \quad (19)$$

where definition (14) of  $g$  was used. If there are only positive eigenvalues for all components of the order parameter, the phase transition from a superconducting state to a normal one occurs at the value of an external magnetic field that corresponds to the lowest eigenvalue among  $\alpha_{n\pm}$ ,  $\alpha_{n3}$ . This argument gives us  $\psi_3 = \psi_+ = 0$ , while the  $\psi_-$  component determines the upper critical field

$$H_{c2}(g) = \frac{2m^*c}{\hbar e^*} \frac{\alpha}{1-g}. \quad (20)$$

If, however, the spectrum started from a negative eigenvalue, then linearization procedure that led us to Eq. (18) would have been inconsistent and the quartic terms in the free-energy equation (6) would have to be retained. Physically such a situation means that the phase transition to the normal state ceases to be of the second order (even neglecting fluctuations, as we do throughout this work). Either the transition becomes of the first order or, more probably, the vacuum rearranges and the transition disappears altogether for any value of magnetic field, no matter how large. The second possibility takes place in our idealized model.

To find the transition line, we therefore look for regions on the  $H$ - $T$  plane where the linearization of GL equation is still consistent and the second-order phase transition to the normal state takes place. Coefficients of the GL equations depend on temperature. Let us assume, for simplicity, that the dependence of  $\alpha$  is linear:  $\alpha = \alpha'(T_c - T)$ ,  $\alpha' > 0$  [recall that in our definitions  $\alpha$  is positive in the superconducting state; see Eqs. (1) and (4)], while the other coefficients are temperature independent. In reality this is true only near  $T_c$ , but necessary modifications for nonlinear behavior can be easily accommodated within the same framework of Ginzburg-Landau approach. Usually the transition line starts at  $(T_c, 0)$  and ends at  $(0, H_{c2})$ ; see solid line in Fig. 1(a). We find that the phase transition line starts naturally at  $(T_c, 0)$  and continues to higher fields, but in the case of

$$g > g_{c2} = 1 \quad (21)$$

turns to higher temperatures instead of lower ones [solid line in Figs. 1(b) and 1(c)].

Thus superconductivity at nonzero magnetic field can take place at  $T > T_c$ . This conclusion, although strange from the

traditional  $s$ -wave point of view, is rather natural under present circumstances. Indeed, from the beginning we assumed the ferromagnetic coupling of the magnetic field to the condensate that means that the field stimulates formation of Cooper pairs. For the particular pairing considered, pairing of  $p$ -wave type, the magnetic field is not acting as a pair-breaking agent. This by no means indicates that the resistive transition in highly fluctuating materials (Ginzburg number not very small) occurs at  $T > T_c$ . As is well known,<sup>20</sup> the resistive transition is associated with the vortex melting line [dashed line in Figs. 1(a), 1(b), and 1(c)], which is much lower than the classical line, especially for temperatures close to  $T_c$ .

At temperatures higher than  $T_c$  [see Figs. 1(b) and 1(c)] superconductivity appears at some finite field, which can be viewed as an analog of conventional  $H_{c2}$  and is given by the same formula (20) with negative  $\alpha$ . At fields slightly higher than this threshold value, magnetization of the sample due to developing superconducting state can be calculated by standard methods<sup>15</sup> taking into account nonlinear terms in the GL equations perturbatively. The result reads

$$M = \frac{(1-g)^2}{4\pi\beta_A[2\kappa^2 - (1-g)^2]} \left( H + \frac{2m^*c\alpha'}{\hbar e^*} \frac{T-T_c}{g-1} \right), \quad (22)$$

where  $\kappa = (m^*c/\hbar e^*)\sqrt{\beta_1/2\pi}$  and  $\beta_A = \langle (\psi_i\psi_i^*)^2 \rangle / \langle \psi_i\psi_i^* \rangle^2$ . Note that the magnetization is positive at  $T > T_c$ . The result (22) is valid in the region 3 on the  $H$ - $T$  diagram (see Fig. 1) and, of course, not in the vicinity of the point  $g = g_{c2} = 1$  itself. Similarly, one can analytically obtain magnetization in the case of  $g < 1$  and  $H \leq H_{c2}(g)$ ,  $T < T_c$ . It is again given by the same formula (22) and it is negative.

The unusual shape of the normal to mixed-state transition line in particular means that for  $T < T_c$  there is no  $H_{c2}$  at all [Fig. 1(b)]. The vortex density therefore is rising indefinitely with  $H$ . How could this happen? It turns out that vortices become thinner. If one defines (square of) coherence length as a ratio of the coefficient of the derivative term in free energy to that of the quadratic in the order-parameter term, it would be given by

$$\xi^2(T, H) \equiv \frac{\hbar^2}{2m^*[\alpha'(T-T_c) + \mu H]}. \quad (23)$$

It follows that for fixed  $T < T_c$ , the correlation length decreases when  $H$  increases. Vortex core size is reduced to allow more vortices to pass through. We will see in the next section that even far from the region considered here, vortices shrink to minimize the energy loss due to Zeeman coupling and will find the dependence almost identical to Eq. (23). One of the consequences of such behavior is that magnetization ceases to follow the linear law of Eq. (22) and becomes saturated.

It is worth noting that the condition for the existence of superconductivity beyond  $T_c$ , Eq. (21), is weaker than the condition of the existence of the spontaneous vortex state at  $H = 0$ , Eq. (16):  $g_{c1} \approx \ln \kappa > g_{c2} = 1$ . Therefore there exists a possibility that superconductivity in a magnetic field exists beyond  $T_c$  although there is no spontaneous vortex state at  $H = 0$ . The phase transition lines in this case are sketched in Fig. 1(b).

Having studied two regions of the fields in which analytic expressions can be obtained (regions 1 and 3 in Fig. 1), we now turn to the intermediate region (2 in Fig. 1) in which different approximations should be made and numerical methods are required. We also will determine where the borderline between the regions lies.

## V. BEYOND THE LONDON APPROXIMATION

In this section, we first investigate the structure of the vortex core of an isolated vortex in the presence of Zeeman coupling. Numerical results confirm the more qualitative conclusions of Sec. III. Then we present an approximation to the magnetization curve for intermediate fields  $B$  (region 2 in Fig. 1) for which the distance between the vortices is larger than the vortex size (although smaller than penetration depth). Since vortices are shrinking when magnetic field grows, this region covers the entire magnetic field range for  $T < T_c$ , if  $g > 1$  [see Figs. 1(b) and 1(c)].

### A. Isolated vortex

In order to study the vortex core structure we have to abandon the London approximation within which the field  $f$  is constant away from singular points. Due to cylindrical symmetry of the vortex, the coordinate dependence of variables is restricted to dependence on the distance from the center of the vortex:  $f(r)$  and  $A(r)$ . The vector potential is oriented azimuthally.

As in liquid  $^3\text{He}$  or heavy-fermion superconductors, there might be various kinds of topologically distinct vortices. The topological analysis we performed shows that there are solitons different from the usual ones considered below. The most interesting one is a Skyrmion of the field  $\mathbf{I}(x)$  (definition of vector  $\mathbf{I}$  is given in Sec. II). We will describe them elsewhere. We have good reasons to believe that the rather conventional Abrikosov vortex has lower energy and, since we are interested in energetics and neglect fluctuations, we concentrate on this type of vortex only. Now we consider the structure of the Abrikosov vortex.

With direction of  $\mathbf{l}|_z$  fixed throughout the volume of the superconductor, twofold discrete symmetry in the order-parameter field of phase I is generated. Correspondingly, the vortex can include two components,  $\sim(\mathbf{n} + i\mathbf{m})e^{ip\varphi}$  and  $\sim(\mathbf{n} - i\mathbf{m})e^{iq\varphi}$ , in the core region. Topological numbers  $p$  and  $q$  are not independent however. They satisfy the relation  $q = p + 2$ , due to definite transformation properties of the system as a whole under global rotations. As a result, in the case of  $g > 0$ , the presence of both components in the vortex core is obviously energetically unfavorable with respect to the Zeeman term in Eq. (6). For a detailed analysis, see Ref. 8. In addition, there is a possibility of still more complicated core structure in which both phases, I and II [see Eqs. (4) and (5)], are present inside the core. It was shown in slightly different context<sup>21</sup> that, at least for large  $\beta_2$ , this situation is not realized.

To consider the single-vortex problem, we introduce dimensionless variables. Energy density is measured in units of  $\alpha^2/\beta_1\kappa^2 = \varepsilon_0/2\pi\lambda^2$ . The absolute value of the order parameter  $\psi$  is given in units of its saturation value  $\sqrt{\alpha/\beta_1}$ , mag-

netic flux in the units of the elementary fluxon  $\Phi_0/2\pi$ , and length in the units of magnetic length  $\lambda$ :

$$F \equiv \frac{\alpha^2}{\beta_1\kappa^2}\tilde{F}, \quad f^2 \equiv \frac{\alpha}{\beta_1}\tilde{f}^2, \quad A \equiv \frac{\Phi_0}{2\pi\lambda}a, \quad (24)$$

$$B \equiv \frac{\Phi_0}{2\pi\lambda^2}b, \quad r \equiv \lambda\rho.$$

To simplify the notations, tilde marks will be dropped hereafter.

For a cylindrically symmetric situation the vortex line energy, which is an integral of Eq. (6) over the whole  $x-y$  plane, takes the form

$$\varepsilon_V = \varepsilon_0 \int_0^\infty \rho d\rho \left[ \frac{\kappa^2}{2}(1-f^2)^2 + \dot{f}^2 + f^2 \left( \frac{1}{\rho} - a \right)^2 - g f^2 \left( \dot{a} + \frac{a}{\rho} \right) + \left( \dot{a} + \frac{a}{\rho} \right)^2 \right], \quad (25)$$

where the condensation energy was subtracted.

Ginzburg-Landau equations take the form

$$\left( \ddot{a} + \frac{\dot{a}}{\rho} - \frac{a}{\rho^2} \right) + f^2 \left( \frac{1}{\rho} - a \right) - g f \dot{f} = 0; \quad (26)$$

$$\kappa^2(f - f^3) + g f \left( \dot{a} + \frac{a}{\rho} \right) - f \left( \frac{1}{\rho} - a \right)^2 + \left( \frac{\dot{f}}{\rho} + \dot{f} \right) = 0,$$

while boundary conditions read  $f(0) = a(0) = 0$ ,  $f(\infty) = 1$ ,  $\dot{a} + a/\rho|_{\rho=\infty} = 0$ .

Small  $\rho$  asymptotic behavior is similar to that of usual Abrikosov vortex, but modified by the presence of  $g$ :

$$f = c\rho \left[ 1 - [\kappa^2 + (g+1)b(0)] \frac{\rho^2}{8} \right], \quad (27)$$

$$b = b(0) + \frac{c^2}{2}(g-1)\rho^2.$$

Results of numerical integration for the GL parameter  $\kappa = 10$  are presented in Fig. 3. The profiles of reduced magnetic field and order parameter for several values of  $g$  are shown in Fig. 3(a). Our main observation is that the magnetic field inside the vortex core is affected drastically, but left almost unchanged outside the core. Changes in the behavior of  $f$  for different  $g$  are less apparent on this scale: the curves are almost indistinguishable. The order parameter rises linearly and reaches its asymptotic value on the scale of coherence length.

Detailed profiles of the magnetic field inside the core are given in Fig. 3(b). As  $g$  grows, the behavior of magnetic field qualitatively changes. For  $g$  exceeding the critical value of 1, see Eq. (27),  $b$  first rises and only then exponentially falls off instead of the usual monotonic decrease from the beginning. This is the response to the Zeeman interaction, which induces supercurrents proportional to the gradient of the square of the order parameter modulus; see Eq. (9).

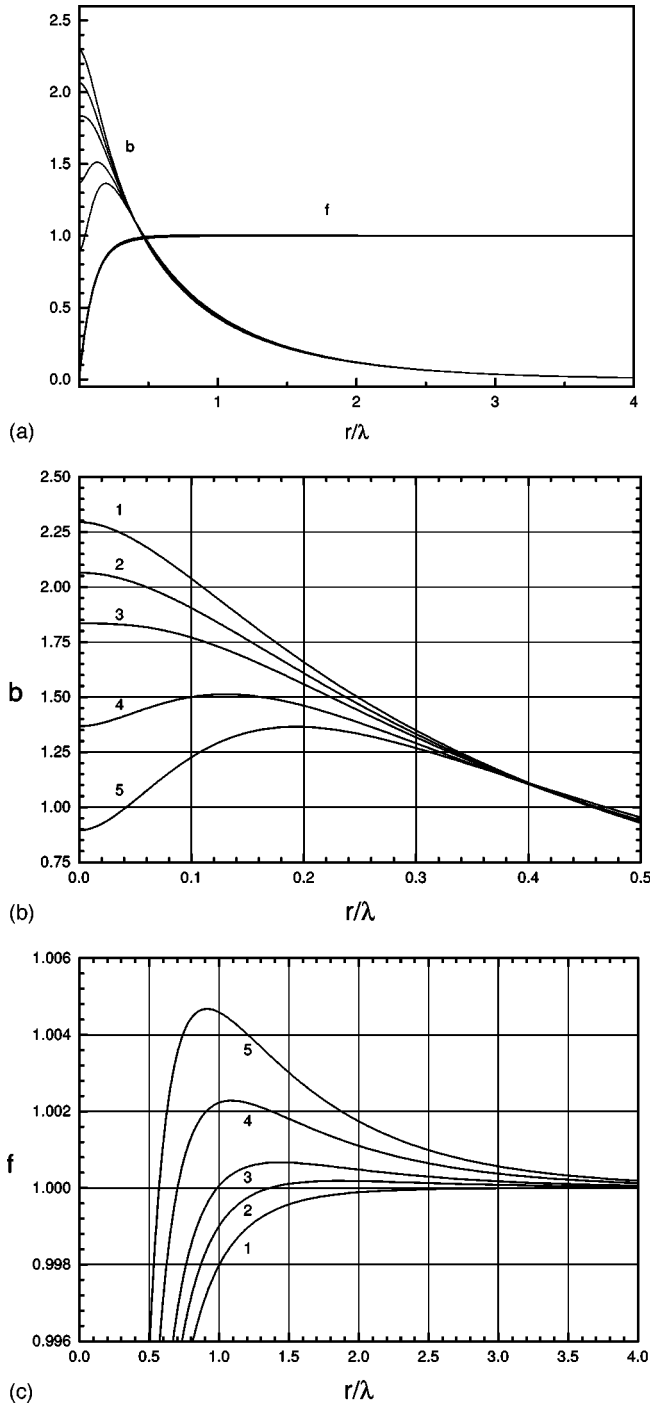


FIG. 3. Vortex structure for  $\kappa=10$  and different values of Zeeman coupling  $g=0(1), 0.5(2), 1(3), 1.25(4), 1.5(5)$ : (a) Large-scale magnetic field and order-parameter variations; (b) Magnetic field in the core; (c) Order-parameter profile close to its saturation value.

The way the order parameter approaches its asymptotic value differs markedly from the conventional monotonic increase [see Fig. 3(c)]. It rises linearly, but then surpasses the far asymptotic value of 1 and finally approaches it from above. It is worth noting that  $f$  becomes greater than 1 outside the core for any nonzero value of  $g$ , no matter how small. The maximum value of the order parameter increases with  $\kappa$  and may become experimentally detectable at  $\kappa=100$ .

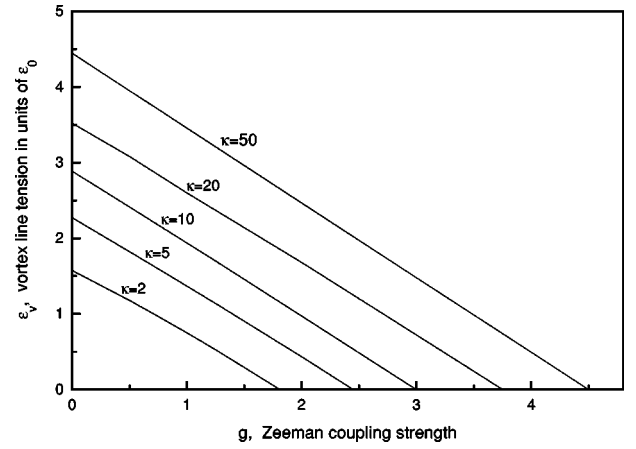


FIG. 4. Vortex line tension  $\epsilon_V$  as a function of Zeeman coupling strength  $g$  for different values of GL parameter  $\kappa$ .

Energy of the vortex is almost a linear function of  $g$ . It is given for several values of  $\kappa$  in Fig. 4. It becomes negative at some  $g_{c1}$ . This critical value of  $g$  as a function of  $\kappa$  is shown in Fig. 5. The high  $\kappa$  estimation equation (16),  $g_{c1}=\ln \kappa$ , is also shown for comparison.

### B. Vortex lattice

Having studied the two extreme cases of isolated vortices (region 1 in Fig. 1) and that of overlapping vortices, where intervortex distance  $a$  is about  $\xi$  (region 3 in Fig. 1), we now turn to the interesting intermediate region in between,  $\xi \ll a \ll \lambda$  (region 2 in Fig. 1). Beyond the London approximation of Sec. III, interaction between vortices is no longer independent of the Zeeman coupling. One expects the core size to be affected by magnetic field and the coupling parameter  $g$ , see Eq. (23), which in turn modifies the interaction energy. Let us introduce core size  $r_c \equiv \rho_c \lambda$  and assume, for simplicity, the steplike behavior of  $f$ . Then the Gibbs free-energy density, using dimensionless variables (24), reads

$$G = b^2 - b \ln \sqrt{\eta \rho_c^2 b} + \frac{1}{4} \kappa^2 \rho_c^2 b - 2bh - gb + \frac{1}{2} g \rho_c^2 b^2. \quad (28)$$

The first two terms represent the usual interaction energy and the vortex energy excluding the Zeeman coupling part. It is obtained by the standard method of summing up all the interactions using transition to the reciprocal lattice space; see Ref. 18. Then the summation in the reciprocal lattice space is replaced by the largest term at the origin plus the integral over a disk starting at  $\pi/a$  and terminating at  $\pi/r_c$ . The third term is the energy lost in the core due to melting of the condensate (which turns out to be rather insignificant). The fourth term is due to external magnetic field. The last two terms represent the Zeeman-coupling contribution and are important. They summarize the gain due to the ferromagnetism of Cooper pairs. It can be thought of as the homogeneous effect (the fifth term) minus the ferromagnetic energy loss in cores due to vanishing of magnetic moment there (the sixth term). Coefficient  $\eta$  is an unknown quantity of order 1



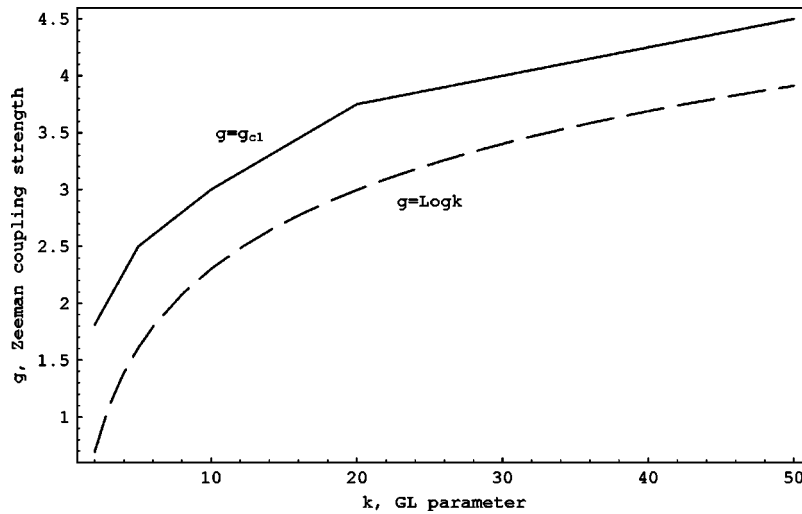


FIG. 5. Critical value of Zeeman coupling strength  $g_{c1}$  at which spontaneous vortex state develops as a function of GL parameter  $\kappa$ .

that reflects the estimated character of our calculation in this subsection. It is used later for joining together the two physically distinct regions 2 and 3.

The value of  $\rho_c$  is found from the condition  $dG/d\rho_c=0$  that gives

$$\rho_c = \sqrt{\frac{2}{\kappa^2 + 2gb}}. \tag{29}$$

Equation (29) shows that as magnetic field increases the core shrinks and is similar to Eq. (23) that we derived in region 3 where there is suppressed superconductivity. The shrinking of cores makes room for more vortices to squeeze in and allows internal magnetic field  $b$  to increase when  $h$  increases (of course, when the core becomes of microscopic size the whole approach ceases to be applicable). At the same time Eq. (29) makes it possible to obtain the boundary value of internal magnetic field  $b_{23}$ , below which the approximation used in the present subsection is valid. The vortices should be well separated in region 2 and we therefore require  $\rho_c b_{23} = 1$  and find  $b_{23} = \kappa^2 / 2(1 - g)$ , or  $B_{23} = H_{c2}(g) / 2$  in

physical units. This, in particular, means that for  $g > 1$ , region 3, where  $a \approx \xi$ , never extends to temperatures lower than  $T_c$ .

The magnetization curve is found from minimization of  $G$  with respect to  $b$ . It leads to

$$h = b - \frac{1}{4} \ln \frac{2b}{\kappa^2 + 2gb} - \frac{g}{2} \left( 1 - \frac{b}{\kappa^2 + 2gb} \right) - \frac{1}{4} \ln \eta. \tag{30}$$

Parameter  $\eta$  can now be determined from the physically transparent requirement that the expressions for magnetization (22) and (30) obtained in the regions 3 and 2 correspondingly have to coincide at  $b = b_{23}$ . In case of  $g < g_{c2} = 1$  these regions correspond to regions of an external field with high and intermediate flux density<sup>15</sup> inside the superconducting sample.<sup>19</sup> We plot the magnetization  $m$  in region 2 as a function of  $h$  for  $\kappa = 20$  and several values of  $g$  in Fig. 6. As  $g$  increases the value of  $H_{c2}(g)$  also increases and at  $g = g_{c2} = 1$  it should become infinite (see Sec. IV). At larger

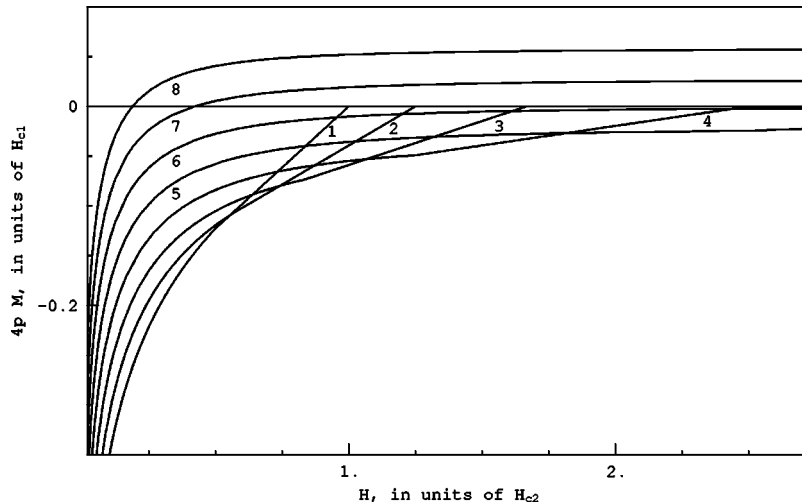


FIG. 6. Magnetization  $M$  of a vortex lattice as a function of external magnetic field  $H$  in the region 2.  $\kappa = 30$  and  $g = 0(1), 0.2(2), 0.4(3), 0.6(4), 0.8(5), 1(6), 1.2(7), 1.4(8)$ .

$g$ , as  $H$  grows, magnetization approaches a positive saturation value, which at high  $\kappa$  can be estimated as

$$m_s = \frac{1}{16\pi} \left[ (g-1) \left( 1 + \frac{1}{\beta_A} \right) - \ln g \right].$$

For  $g$  larger than critical value  $g_{c1}$  the remanence  $b_r \equiv b(h=0)$  exists and it is determined implicitly by Eq. (30).

## VI. SUMMARY AND DISCUSSION

In this paper the formation of spontaneous vortex phase in type-II superconductors with  $p$ -wave pairing was studied using the Ginzburg-Landau approach. Due to direct coupling of the Cooper pair's spin to magnetic field, in certain cases condensate acquires ferromagnetic properties. There are three cases depending on the strength of the effective Zeeman coupling, measured by dimensionless parameter  $g$ . First, if  $g < g_{c2} = 1$ , the phase diagram is similar to the usual one [Fig. 1(a)], although  $H_{c2}$  grows unlimitedly as  $g$  approaches 1. Second, when  $g_{c2} < g < g_{c1} \approx \ln \kappa$ , at magnetic field a mixed state can exist even beyond  $T_c$ : the direct coupling to magnetic field facilitates creation of the Cooper pairs along with the usual destruction of the condensate [Fig. 1(b)], while at  $T < T_c$ ,  $H_{c2}$  disappears. Vortices become thinner when  $H$  grows. The vortex core structure is as follows: the order parameter first increases almost linearly to a value slightly above its asymptotic value and then gradually decreases to it. Internal magnetic field first increases within the core before eventual exponential decrease at penetration depth distance. Third, when  $g > g_{c1}$ , the Meissner phase completely disappears [Fig. 1(c)]. Below  $T_c$  the vortex phase appears spontaneously at  $H=0$ , and exists for arbitrarily large external magnetic field for which the GL macroscopic approach is applicable.

Below we make several comments on the results presented in previous sections. First, there is the strange direction of the superconductor-normal phase transition line. Our mean field treatment completely neglects fluctuations. In strongly fluctuating superconductors (Ginzburg number not very small) to which high- $T_c$  cuprates, especially Ba-Ca-Sr-Cu-O compounds and similarly layered compounds with  $G_i \sim 0.1$ , and presumably the Ru-based compounds studied in Ref. 3, belong, the line where the order parameter vanishes ceases to describe actual phase transition. Instead, a much lower vortex lattice melting line appears. Vortex liquid is not a superconductor, as far as conductivity properties are concerned. Broad resistive transition in  $\text{Sr}_2\text{YRu}_{1-x}\text{Cu}_x\text{O}_6$  is presumably associated with melting<sup>22</sup> (see dashed line in Fig. 1).

Second, disorder can further restrict the vortex lattice region, and third, the form  $\alpha(T) = \alpha' (T - T_c)$  should be re-

placed by a more realistic one away from  $T_c$ . Similarly, the region of very large fields in which the vortex core becomes of microscopic size is beyond the reach of the macroscopic Ginzburg-Landau approach.

An additional issue is the value of the coefficient  $\beta_2$  of the additional term in the free energy (1). It might have a weak temperature dependence and can even change sign at certain temperature. Then the Zeeman-coupling effects described here will disappear and usual diamagnetic superconducting behavior sets in. Whether the resistivity results of Ref. 3 can be understood that way is not clear to us presently.

Finally, we would like to emphasize that although the novel superconducting system  $\text{Sr}_2\text{YRu}_{1-x}\text{Cu}_x\text{O}_6$ ,<sup>3</sup> that motivated the present research, has low orthorhombic symmetry  $Pbnm$ , a realization of the physical situation proposed by us is in principle possible, that is, nonunitary superconducting phases exist for this crystal symmetry. A weak spin-orbit coupling scheme obviously permits nonunitary phases. So actually does a strong spin-orbit coupling scheme, though this is less apparent due to the fact that irreducible representations of the point group  $D_{2h}$  of the crystal in question are all one dimensional. Nonunitary phases would correspond in this case to irreducible representations of the whole space group  $Pbnm$  with  $k \neq 0$  that are two dimensional.<sup>23</sup> One of our main conclusions about the existence of a spontaneous vortex state for sufficiently strong Zeeman coupling remains valid, at least as far as symmetrical crystalline directions for magnetic field are concerned. On the other hand, an estimate for the value of Zeeman coupling would vary considerably for different choices of a microscopic model. Such estimations are beyond the scope of our phenomenological approach. It is worth noting that Zeeman coupling is believed to be very small in  $\text{UPt}_3$ , in connection with which it was first considered by Tokuyasu *et al.*<sup>12</sup>

Experimentally, the distinct vortex core structure could be seen using scanning tunneling microscopy technique for high value of  $\kappa$ . Every spot should be surrounded by a ring. It is perhaps more difficult to see the decrease of magnetic field at the vortex core using electron tomography or Bitter technique.

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