Dimensionality-driven changeover to first-order superconducting phase transitions in the Pauli paramagnetic limit

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Superconducting (SC) phase transitions in strongly type-II superconductors in the Pauli paramagnetic limit are considered within the framework of the Gorkov–Ginzburg–Landau approach in the lowest Landau level approximation for both *s*- and *d*-wave electron pairings. Simple analytical expressions for the quadratic and quartic coefficients in the order parameter expansion of the SC free energy are derived without relying on gradient or wave number expansions. The existence of a changeover from continuous to discontinuous SC phase transitions that are predicted to occur in the clean limit is shown to depend on the dimensionality of the underlying electronic band structure but to be independent of the type of the electron pairing. Such a changeover can take place in the quasi-two-dimensional (2D) regime below a critical value of a three-dimensional–two-dimensional (3D-2D) crossover parameter. In the 3D limit, wherein the normal to SC phase transitions are of second order and the SC phase is spatially modulated along the field direction, the transition line to a uniform SC phase is of first order and is usually very close to the normal to SC transition line.

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I. INTRODUCTION

In recent years, a number of unusual superconducting (SC) states have been discovered in different materials.[1] Most of these materials are strongly type-II superconductors, possessing highly anisotropic or even quasi-two-dimensional (2D) electronic structures. Of special interest in the present paper are SC materials that show peculiar clean-limit features at high magnetic fields and low temperatures, notably the recently discovered family of heavy-fermion compounds $CeRIn_5$ (R=Rh, Ir, and Co)[2] and some of the organic charge transfer salts of the type $(BEDT\text{-}TTF)_2X$.[3,4]

The heavy-fermion compound $CeCoIn_5$, for example, which is believed to be an unconventional (*d*-wave) superconductor[5] similar to the high-T_c cuprates, exhibits the highest T_c ($\sim$2.3 K) among Ce-based heavy-fermion compounds. This material is characterized by exceptionally strong Pauli paramagnetic pair breaking[6,7] due to its extremely large electron effective mass and small Fermi velocity, which could lead to discontinuous (first-order) SC phase transitions at sufficiently high magnetic fields.[8–10]

Recently, Bianchi *et al.*[11] observed a changeover of the second-order SC phase transition to a first-order transition in specific heat measurements performed on this material as the magnetic field is increased above some critical values for both parallel and perpendicular field orientations with respect to the easy conducting planes. A similar effect was very recently observed by Lortz *et al.*[12] in the nearly 2D organic superconductor κ-$(BEDT\text{-}TTF)_2Cu(NCS)_2$ but only for a magnetic field orientation parallel to the SC layers, where the orbital (diamagnetic) pair breaking is completely suppressed. Under these conditions, the usual (uniform) SC state is expected to be unstable with respect to the formation of a nonuniform SC state, as predicted more than 40 years ago by Fulde and Ferrel[13] and by Larkin and Ovchinnikov[14] (FFLO). The corresponding SC order parameter is spatially modulated along the field direction with a characteristic wave number q, whose kinetic energy cost is compensated by the Pauli pair-breaking energy. The critical temperature, T_{FFLO}, for the appearance of the FFLO phase is found to be equal to $0.56T_c$. At the corresponding tricritical point, the normal (N), the uniform, and nonuniform SC phases are all met.

The possibility of a changeover to first-order transitions can effectively be investigated within the Ginzburg–Landau (GL) theory of superconductivity since for the uniform SC phase (i.e., for $q=0$) the coefficient (usually denoted by β) of the quartic term in the GL expansion changes sign at a temperature T^*, which coincides with T_{FFLO}.[15] The identity of T^* with T_{FFLO} is peculiar to the clean limit of a superconductor with no orbital pair breaking. In conventional *s*-wave superconductors, electron scattering by nonmagnetic impurities shifts T_{FFLO} below the critical temperature T^*,[16] allowing discontinuous phase transitions at temperatures $T_{FFLO}<T\leq T^*$, since (following Anderson's theorem) β is not influenced by nonmagnetic impurities. In superconductors with an unconventional electron pairing, wherein β is strongly affected by nonmagnetic impurity scattering, the situation is reversed, i.e., $T^*<T_{FFLO}$.[16]

The interplay between orbital and spin depairing in a pure *s*-wave isotropic 3D superconductor was first discussed by Gruenberg and Gunther,[17] who conjectured (i.e., without presenting any result for the coefficient β) that for $T<0.56T_c$, the N-SC transition is of the second order, whereas at lower fields there should be first-order transitions to a uniform SC phase. Houzet and Buzdin[18] essentially confirmed this picture by exploiting order-parameter and gradient expansions in the GL theory to find that $T^*<T_{FFLO}$, so that at temperatures $T^*<T<T_{FFLO}$, there are second-order transitions to either the Larkin–Ovchinnikov (LO) or the Fulde–Ferrel (FF) phase. It should be noted, however, that the orbital effect was treated there by using gradient expansions, which is a strictly valid approximation only at very low magnetic fields.

In contrast to these papers, Adachi and Ikeda recently found[19] that, in a clean, *d*-wave, quasi-2D (layered) superconductor, the orbital effect always shifts T_{FFLO} below T^*. In

this work, the authors used an order-parameter expansion in the Gorkov Green's function approach to the GL theory up to the sixth order, avoiding the restrictions of gradient expansion by exploiting the lowest Landau level (LLL) approximation for the condensate of Cooper pairs. Accounting for impurity scattering destroys the FFLO phase and, in contrast to the pure paramagnetic situation, somewhat reduces T^*. The effect of SC thermal fluctuations was found in this work to broaden the discontinuous mean-field transition at T^* into a crossover. The reliance on (FFLO) wave number expansion and on extensive numerical computations in this work has saved formidable analytical efforts, leaving, however, interesting questions unanswered. In particular, the origin of the relative shift of T_{FFLO} below T^* by the orbital effect found in this paper, in contrast to all the earlier studies, remains unknown.

Houzet and Mineev[20] partly clarified the situation by considering two different models of paramagnetically limited superconductors: a 3D isotropic s-wave superconductor and a quasi-2D d-wave superconductor under a magnetic field perpendicular to the conducting layers. Within their microscopically derived GL functional approach, which exploited gradient expansions, it was found in the clean limit that for the former model, $T^* < T_{\mathrm{FFLO}}$, whereas for the latter model, the relation is reversed, that is, $T^* > T_{\mathrm{FFLO}}$. It was not clear, however, from these findings whether the difference in behaviors was due to the difference in types of pairing or to the difference in Fermi surface dimensionalities characterizing the two models.

In the present paper, we develop a formalism based on order-parameter expansion within the Gorkov theory for a strongly type-II superconductor, with both s- and $d_{x^2-y^2}$-wave electron pairings at high magnetic fields, which is sufficiently simple to yield useful analytical expressions for the SC free energy to any desired order in the expansion. By exploiting the LLL approximation, the fundamental interplay between spin induced paramagnetic and orbital diamagnetic effects at an arbitrary magnetic field is studied, within a model of anisotropic electron systems covering the entire 3D-2D crossover range, without relying on gradient or wave number expansions. These advantages enable us to shed a different light on the yet undecided debate concerning the order of the SC phase transitions in the presence of a strong Zeeman spin splitting and to push our investigation into the unexplored region of very low temperatures, where quantum magnetic oscillations have been shown to be observable in the heavy-fermion compounds.[21,22]

Specifically, it is found that the relevant parameter controlling the relative position of T_{FFLO} with respect to T^* is the dimensionality of the electronic orbital motion in the crystal lattice, through its influence on the orbital (diamagnetic) pair-breaking effect. For a 3D Fermi surface (isotropic or anisotropic), on which the electron motion along the magnetic field direction reduces the cyclotron kinetic energy, the shift of T^* to low temperatures is larger than that of T_{FFLO}. In this case, the kinetic energy of Cooper pairs associated with their motion along the field can compensate for the spin-splitting effect, thus leading to an increase in β and the disappearance of the first-order transition. The corresponding phase diagram is similar to that suggested in Ref. 17, in which the N-SC transition is of second order. However, due to the strong dependence of α and β on the FFLO wave number q, the SC free energy has a nonmonotonic dependence on q, so that the transition between the nonuniform and uniform (along the field) SC states is of first order. For spin-splitting energies that are not too large as compared to the critical paramagnetic value, the corresponding continuous and discontinuous transition lines are found very close to each other, so that in the presence of thermal fluctuations they might be indistinguishable.

In the quasi-2D limit (i.e., for quasicylindrical Fermi surfaces), the enhanced orbital pair-breaking shifts T_{FFLO} below T^*, which is in agreement with Adachi and Ikeda.[19]

II. ORDER PARAMETER EXPANSION IN THE PRESENCE OF SPIN-SPLIT LANDAU LEVELS

Our starting point is an effective BCS-type Hamiltonian with a $d_{x^2-y^2}$-wave pairing interaction similar to that exploited, e.g., by Agterberg and Yang.[16] The conventional s-wave situation can similarly be worked out and so will not be presented in detail here. The thermodynamical potential (per unit volume) for the corresponding d-wave superconductor, as expanded in the order parameter with nonlocal normal electron kernels, may be written as

$$\Omega = \frac{\Delta_0^2}{V} + \sum_{m=1} \frac{(-1)^m}{m} \tilde{\Omega}_{2m}\{\Delta(\mathbf{R},\mathbf{r})\}, \tag{1}$$

where $\tilde{\Omega}_{2m}\{\Delta(\mathbf{R},\mathbf{r})\}$ is a functional of the SC order parameter, $\Delta(\mathbf{R},\mathbf{r})$, that has a power-law dependence $\sim|\Delta_0|^{2m}$ on the spatial average amplitude, Δ_0, of the order parameter, and V is a BCS coupling constant (given in units of energy $\times$ volume). The corresponding d-wave order parameter depends on both the center of mass ($\mathbf{R}$) and the relative ($\mathbf{r}$) coordinates of a condensate of electron pairs: $\Delta(\mathbf{R},\mathbf{r}) = \Delta(\mathbf{R})\varphi(\mathbf{r})$. It should be self-consistently determined from the corresponding pair-correlation functions. Only stationary solutions are considered, neglecting quantum and thermal fluctuations. In addition, the order parameter in the mean-field approximation is selected as a hexagonal vortex lattice. Actually, this assumption is not very important since the second-order term in the order-parameter expansion does not depend on the vortex lattice structure, whereas the lattice structure dependence of the quartic term is very weak (see Refs. 23 and 24).

For the underlying system of normal electrons, we assume a simple model of quadratic energy dispersion $\varepsilon(k_x,k_y,k_z) = \hbar^2(k_x^2+k_y^2)/2m^* + \hbar^2 k_z^2/2m_z^*$ and an anisotropic effective mass tensor: $m^* \leq m_z^*$. A quasi-2D situation is characterized by a sufficiently large anisotropy parameter $\chi_a = \sqrt{m_z^*/m^*}$, corresponding to an elongated Fermi surface with a Fermi momentum k_F and a Fermi energy $\varepsilon_F \equiv \hbar^2 k_F^2/2m^*$, which is truncated by the Brillouin zone (BZ) face at $k_{z,\max} = \pi/d$, where d is the lattice constant perpendicular to the easy planes. A parameter determining the dimensionality of the Fermi surface may be defined by $v_0 = \sqrt{\frac{\varepsilon_{z,\max}}{\varepsilon_F}}$, where $\varepsilon_{z,\max} \equiv \hbar^2 k_{z,\max}^2/2m_z^*$ is the maximal value of the electron energy along the field. Thus, in the 2D limit, $k_{z,\max} \ll k_F$, we have

$v_0 \to 0$, while the system may be regarded 3D (isotropic or anisotropic) if $k_{z,\max} \simeq k_F$, for which the Fermi surface is contained entirely within the first BZ, namely, for $v_0=1$.

At any order of the expansion [Eq. (1)], the nonlocal electronic kernel of the corresponding functional [see, e.g., Eqs. (3) and (5) and Eqs. (25) and (26)] consists of a product of $m=1,2,\ldots$, pairs of normal electron Green's functions in a constant magnetic field, $\mathbf{H}=H\hat{z}$ (i.e., perpendicular to the easy conducting layers), which are written in the form $G_{\uparrow\downarrow}(\mathbf{R}_1,\mathbf{R}_2,\omega_\nu)=G_{0\uparrow\downarrow}(\mathbf{R}_2-\mathbf{R}_1,\omega_\nu)g(\mathbf{R}_1,\mathbf{R}_2)$, where the gauge factor is given by $g(\mathbf{R}_1,\mathbf{R}_2)=e^{-(i/2a_H^2)[\mathbf{R}_1\times\mathbf{R}_2]\cdot\hat{z}}$, $a_H=\sqrt{c\hbar/eH}$, and the gauge invariant part can be calculated by the well known expression[25]

$$G_{0\uparrow\downarrow}(\mathbf{R}_2-\mathbf{R}_1,\omega_\nu)=\frac{1}{2\pi a_H^2}\int\frac{dk_z}{2\pi}e^{ik_z(Z_2-Z_1)}\times\sum_n\frac{e^{-\rho^2/4}L_n(\rho^2/2)}{\mu-\varepsilon_{nk_z\uparrow\downarrow}+i\hbar\omega_\nu+i\,\mathrm{sign}(\omega_\nu)\hbar\Gamma}. \tag{2}$$

Here, $\omega_\nu=\pi k_BT(2\nu+1)/\hbar$, where $\nu=0,\pm1,\ldots$, is the Matsubara frequency, $\varepsilon_{nk_z\uparrow}=\hbar\omega_c(n+1/2+x^2-g/2)$ and $\varepsilon_{nk_z\downarrow}=\omega_c(n+1/2+x^2+g/2)$ are the spin-split normal electron energy levels, $\omega_c=eH/m^*$ is the in-plane electronic cyclotron frequency, $x^2=\xi^2k_z^2\equiv\frac{k_z^2}{2m_z^*\omega_c}$ is a dimensionless longitudinal (parallel to the magnetic field) kinetic energy, $\omega_c g\equiv eH/m_0$ is the Zeeman spin-splitting energy, Γ is the impurity scattering relaxation rate, and $\mu=\hbar\omega_c(n_F+1/2)\approx\varepsilon_F=\hbar^2k_F^2/2m^*$ is the chemical potential. The spatial variables are dimensionless in-plane (perpendicular to the magnetic field) coordinates, $\rho=\frac{\mathbf{R}_{2\perp}-\mathbf{R}_{1\perp}}{a_H}$, and longitudinal coordinates: $Z_1=\mathbf{R}_1\cdot\hat{z}$ and $Z_2=\mathbf{R}_2\cdot\hat{z}$.

A. Quadratic term

In the expansion [Eq. (1)], the second-order term, which describes the SC condensation energy of spin-singlet electron pairs, propagating from initial ($i=1$) to final ($i=2$) coordinates $\mathbf{R}_i\pm\frac{\mathbf{r}_i}{2}$, is given by

$$\Omega_2=\frac{\Delta_0^2}{V}-\frac{1}{\mathcal{V}_0}\int d^3\mathbf{R}_1d^3\mathbf{R}_2\tilde{\Gamma}_2(\mathbf{R}_1,\mathbf{R}_2)\tilde{K}_2(\mathbf{R}_1,\mathbf{R}_2)\equiv\frac{\Delta_0^2}{V}-A_0\Delta_0^2, \tag{3}$$

where $\mathcal{V}_0=SL_z$ is the volume of the system. The vertex part, $\tilde{\Gamma}_2$, is a product of two order parameters multiplied by the gauge factors, $g(\mathbf{R}_2,\mathbf{R}_1)$, which are functions of the center of mass coordinates only, due to the cancellation by the corresponding phase factors of the order parameters, namely,

$$\tilde{\Gamma}_2(\mathbf{R}_1,\mathbf{R}_2)=g^*(\mathbf{R}_1,\mathbf{R}_2)g(\mathbf{R}_2,\mathbf{R}_1)\Delta(\mathbf{R}_1)\Delta^\star(\mathbf{R}_2). \tag{4}$$

The kernel $\tilde{K}_2$ is a product of two translational invariant Green's functions, convoluted with the corresponding factors of the order parameters, which depend only on the relative pair coordinates, namely,

$$\tilde{K}_2(\mathbf{R}_1,\mathbf{R}_2)=k_BT\sum_\nu\int d^3\mathbf{r}_1d^3\mathbf{r}_2\varphi(\mathbf{r}_1)\varphi^*(\mathbf{r}_2)G_{0\uparrow}^*\left(\mathbf{R}_2-\mathbf{R}_1+\frac{\mathbf{r}_2-\mathbf{r}_1}{2},\omega_\nu\right)G_{0\downarrow}\left(\mathbf{R}_1-\mathbf{R}_2+\frac{\mathbf{r}_2-\mathbf{r}_1}{2},\omega_\nu\right). \tag{5}$$

The factor of the order parameter that depends on the pair center of mass coordinates is written, in the LLL approximation,[26] as[27]

$$\Delta(\mathbf{R})=c(Z)\sum_n e^{i\pi n^2/2}\phi_n(\mathbf{R}_\perp),$$

$$\phi_n(\mathbf{R}_\perp)=\exp\left[i\frac{2\pi n}{a_xa_H}X-\left(\frac{Y}{a_H}-\frac{\pi n}{a_x}\right)^2\right], \tag{6}$$

where $a_x=(\sqrt{2\pi}/3)^{1/4}$ is the hexagonal vortex lattice spacing in units of magnetic length and $c(Z)=c_0e^{iqZ}$ $[c_0=(\frac{2\pi}{a_x^2})^{1/4}\Delta_0]$ is the Fulde–Ferrell modulation factor. It should be noted that in the present calculations, the LO modulation has not been considered; all derivations were made for the FF state. This approach is justified since our main purpose here is in examining the existence of a changeover from continuous to discontinuous transitions and in studying how the appearance of a modulated SC phase affects this changeover. Obviously, this restriction does not allow us to discuss any structural transitions inside the modulated state. Exploiting the fact that the kernel $\tilde{K}_2(\mathbf{R}_1,\mathbf{R}_2)$ depends only on the difference $\mathbf{R}_1-\mathbf{R}_2$, one may first carry out the integration in Eq. (3) over the in-plane mean coordinates $\mathbf{R}_\perp=(\mathbf{R}_{\perp,1}+\mathbf{R}_{\perp,2})/2$ to get the following average vertex part:[28]

$$\langle\tilde{\Gamma}_2\rangle=\frac{1}{\mathcal{V}_0}\int\tilde{\Gamma}_2(\mathbf{R}_1,\mathbf{R}_2)d^2R_\perp=|c_0|^2\frac{a_x}{\sqrt{2\pi}}e^{(-\rho^2/2)-iq(Z_2-Z_1)}=\Delta_0^2e^{(-\rho^2/2)-iq(Z_2-Z_1)}, \tag{7}$$

where $\Delta_0^2=\frac{1}{\mathcal{V}_0}\int d^3R|\Delta(\mathbf{R})|^2$ and then integrate over the rest of the coordinates $\rho=(\mathbf{R}_{\perp,2}-\mathbf{R}_{\perp,1})/a_H$ and $\rho_z=(Z_2-Z_1)/a_H$.

Since, among other things, we are interested in the effect of quantum magnetic oscillations, we apply a technique of exact summation over Landau levels (LLs) that was suggested in Ref. 29. It is similar to the Poisson summation formula, which transforms the summation over LLs into a summation over the harmonics of the inverse magnetic field and allows us to separately deal with the uniform (quasiclassical) contribution and the various quantum corrections. This technique can be briefly described as follows: Let us consider the integral representation of the Green's functions, $[n_F-n-x^2\pm i\omega]^{-1}=\int_0^\infty d\tau e^{\pm i\tau[n_F-n-x^2\pm i\omega]}$, and perform the summation over LLs by using the well known identity, $\Sigma_{n=0}^\infty z^nL_n(t)=(1-z)^{-1}\exp(\frac{tz}{z-1})$, where $z=e^{\pm i\tau}$ and $t=\rho^2/2$. By taking advantage of these relations, the gauge invariant part of the Green's function for $\omega_\nu\geq0$ can be transformed to

$$G_{0\uparrow\downarrow}(\mathbf{R}_2-\mathbf{R}_1;\omega_\nu) = \frac{1}{2\pi a_H^2\hbar\omega_c}\int\frac{dk_z}{2\pi}e^{ik_z(Z_2-Z_1)} \times\int_0^\infty d\tau\frac{e^{i\tau[n_F-x^2+g+i\tilde{\omega}_\nu+i\tilde{\Gamma}]}}{(1-e^{-i\tau})}\exp\left(\frac{\rho^2}{4}\frac{1+e^{i\tau}}{1-e^{i\tau}}\right), \quad (8)$$

where $\tilde{\omega}_\nu\equiv\omega_\nu/\omega_c$ and $\tilde{\Gamma}\equiv\Gamma/\omega_c$. For $\omega_\nu<0$, one should replace τ with $-\tau$ (or ω_ν with $-|\omega_\nu|$).

The scattering of electrons by nonmagnetic impurities is taken into account here as a self-energy correction to the single electron Green's functions by using the standard relaxation time approximation. Vertex corrections to the quadratic kernel $\tilde{K}_2(\mathbf{R}_1,\mathbf{R}_2)$ (as well as to higher order ones), which are known to exactly cancel the self-energy insertions in the very weak magnetic field regime of conventional s-wave superconductors (see, e.g., Ref. 30), are not so crucial in the strong magnetic field regime of both the s- and d-wave situations that are investigated here and will therefore be neglected in our calculations, as done, e.g., in Refs. 19 and 31. In any event, for the high magnetic field and relatively clean superconductors that are considered here, the length scale, a_H, corresponding to the diamagnetic pair breaking is much smaller than the electron mean free path v_F/Γ, and the effect of impurity scattering is marginal.

By utilizing this approximation, we rewrite the kernel in the following form:

$$\tilde{K}_2(\rho,\rho_z) = \frac{k_BT}{(2\pi a_H^2\hbar\omega_c)^2}\sum_\nu\int dz_1dz_2\int\frac{dk_{z,1}}{2\pi}e^{ik_{z,1}(\rho_z+\sigma_z/2)} \times\int\frac{dk_{z,2}}{2\pi}e^{ik_{z,2}(\rho_z-\sigma_z/2)}\int_0^\infty d\tau_1e^{i\tau_1[n_F-\xi^2k_{z,1}^2+g+i\tilde{\omega}_\nu+i\tilde{\Gamma}]} \times\int_0^\infty d\tau_2e^{-i\tau_2[n_F-\xi^2k_{z,2}^2-g-i\tilde{\omega}_\nu-i\tilde{\Gamma}]}J(\tau_1,\tau_2,\rho), \quad (9)$$

where $\sigma=(\mathbf{r}_{\perp,2}-\mathbf{r}_{\perp,1})/a_H$, $\sigma_z=(z_2-z_1)/a_H$, and

$$J(\tau_1,\tau_2,\rho) = \int d^2\mathbf{r}_{\perp,1}d^2\mathbf{r}_{\perp,2}f^*(\mathbf{r}_2)(1-e^{-i\tau_1})^{-1} \times\exp\left(\frac{(\rho+\sigma/2)^2}{4}\frac{1+e^{i\tau_1}}{1-e^{i\tau_1}}\right)(1-e^{i\tau_2})^{-1} \times\exp\left(\frac{(\rho-\sigma/2)^2}{4}\frac{1+e^{-i\tau_2}}{1-e^{-i\tau_2}}\right). \quad (10)$$

In Eq. (9), we use the representation $\varphi(\mathbf{r})=\delta(z)f(\mathbf{r}_\perp)$, where $f(\mathbf{r}_\perp)=\int\frac{d^2k}{(2\pi)^2}f_\mathbf{k}e^{i(\mathbf{k}\cdot\mathbf{r}_\perp)}$ describes the two types of electron pairing, the symmetric s-wave pairing with

$$f_{sk} = \frac{1}{2}[\cos(k_xd)+\cos(k_yd)] \quad (11)$$

and $d_{x^2-y^2}$-wave pairing,

$$f_{dk} = \frac{1}{2}[\cos(k_xd)-\cos(k_yd)], \quad (12)$$

producing nodes in the order parameter along the $k_x=\pm k_y$ directions. The δ dependence on z enables us to readily perform the first two integrations in Eq. (9).

By using the resulting expression for $\tilde{K}_2(\rho,\rho_z)$ and the vertex function $\langle\tilde{\Gamma}_2\rangle$, one can calculate the nontrivial coefficient A_0 in Eq. (3) by performing the integrals over ρ and ρ_z,

$$A_0 = \int d^2\rho d\rho_z\tilde{K}_2(\rho,\rho_z)e^{-\rho^2/2-iq\rho_z}, \quad (13)$$

with the other integrals incorporated in the kernel as appearing in Eq. (9). It is convenient to perform the integration over ρ first since both the function $J(\tau_1,\tau_2,\rho)$ and the vertex part have a Gaussian dependence on ρ, which can be readily carried out with the result

$$J(\tau_1,\tau_2) = \int d^2\rho e^{-\rho^2/2}J(\tau_1,\tau_2,\rho) = \frac{2\pi J_p(\tau_1,\tau_2)}{2-e^{-i\tau_1}-e^{i\tau_2}}, \quad (14)$$

where

$$J_p(\tau_1,\tau_2) = \int d^2\mathbf{r}_{\perp,1}d^2\mathbf{r}_{\perp,2}f(\mathbf{r}_1)f^*(\mathbf{r}_2)e^{-(\gamma\tau/8)\sigma^2} \quad (15)$$

and $\gamma_\tau=\frac{2+e^{-i\tau_1}+e^{i\tau_2}}{2-e^{-i\tau_1}-e^{i\tau_2}}$. It should be noted here that the type of pairing influences the SC condensation energy through the functional dependence of J_p on the pairing function $f_\mathbf{k}$.

By performing the straightforward calculation of $J_p(\tau_1,\tau_2)$ for both functions, one obtains

$$J_{sp}(\tau_1,\tau_2) = \frac{1}{4}(1+e^{-(1/4)\gamma_\tau d^2})^2\simeq 1, \quad (16)$$

$$J_{dp}(\tau_1,\tau_2) = \frac{1}{4}(1-e^{-(1/4)\gamma_\tau d^2})^2\simeq\frac{1}{4}\left(\frac{\gamma_\tau d^2}{4}\right)^2, \quad (17)$$

where the last approximate step is obtained in the limit $\frac{\gamma_\tau}{4}d^2\ll 1$. This can be justified by noting that the scale of the function γ_τ is of the order of unity, whereas d (in units of the magnetic length) is much smaller than 1. In the opposite limit, $J_{sp}(\tau_1,\tau_2)=J_{dp}(\tau_1,\tau_2)=\frac{1}{4}$.

Thus, by noting that the integration of A_0 over the center of mass coordinates yields just the total volume of the system and performing the integration over the relative coordinate ρ_z,

$$\int d\rho_z\int\frac{dk_{z,1}}{2\pi}e^{ik_{1,z}\rho_z}\int\frac{dk_{z,2}}{2\pi}e^{ik_{z,2}\rho_z}e^{i\tau_1[n_F-\xi^2k_{z,1}^2+g+i\tilde{\omega}_v+i\tilde{\Gamma}]} \times e^{-i\tau_2[n_F-\xi^2k_{z,2}^2-g-i\tilde{\omega}_v-i\tilde{\Gamma}]}e^{-iq\rho_z} = \int\frac{dk_z}{2\pi}e^{i\tau_1[n_F-\xi^2(k_z+q/2)^2+g+i\tilde{\omega}_v+i\tilde{\Gamma}]} \times e^{-i\tau_2[n_F-\xi^2(k_z-q/2)^2-g-i\tilde{\omega}_v-i\tilde{\Gamma}]}, \quad (18)$$

one obtains for a d-wave superconductor the following:

$$A_0^{(d)} = \frac{6k_BT}{(\hbar^2 k_{z,\max}^2/2m^*)^2 a_H^2} \sum_\nu \int \frac{dk_z}{2\pi} \int_{-\infty}^{\infty} d\tau_1 d\tau_2 \times \frac{e^{i\tau_1[n_F - \xi^2(k_z+q/2)^2 + g + i\tilde{\omega}_\nu + i\tilde{\Gamma}]} e^{-i\tau_2[n_F - \xi^2(k_z-q/2)^2 - g - i\tilde{\omega}_\nu - i\tilde{\Gamma}]}}{(2 - e^{-i\tau_1} - e^{i\tau_2})^3}. \quad (19)$$

A similar expression can be derived for an *s*-wave superconductor. Below, we will present only the final result for this case [see Eqs. (22) and (24)].

Equation (19) is an *exact* representation for the coefficient of the quadratic term in the order-parameter expansion [Eq. (3)], which includes low temperature quantum corrections and quantum magnetic oscillations. It can be written as a sum of contributions from poles at the 2D lattice: $\tau_1 = 2\pi n_1$ and $\tau_2 = 2\pi n_2$, where $n_{1,2} = 0, 1, \ldots$. The dominant (zero harmonic) quasiclassical contribution arises from the pole at $n_1 = n_2 = 0$, whereas the quantum corrections are associated with the poles at $n_1 = n_2 \neq 0$. It is easy to see that the oscillating terms correspond to the off-diagonal poles, $n_1 \neq n_2$, in the (τ_1, τ_2) plane.

In the present paper, we are mainly interested in the quasiclassical contribution for which a further simplification can be achieved. By changing to new variables, $\tau_2 = \rho_0 + \frac{\tau}{2}$, $\tau_1 = \rho_0 - \frac{\tau}{2}$, and exploiting the expansion $2 - e^{-i\tau_1} - e^{i\tau_2} \simeq \rho_0^2 - i\tau$ near the "quasiclassical" pole $\tau_1 = \tau_2 = 0$, one carries out the integral over τ in Eq. (19) to have

$$A_0^{(d)} = \frac{6k_BT}{(\hbar^2 k_{z,\max}^2/2m^*)^2 a_H^2} \sum_\nu \int dk_z \int_0^{\infty} d\rho_0 e^{2i\rho_0[\xi^2 q k_z + g + i\tilde{\omega}_\nu + i\tilde{\Gamma}]} \times \{n_F - \xi^2[k_z^2 + (q/2)^2]\}^2 e^{-\rho_0^2\{n_F - \xi^2[k_z^2 + (q/2)^2]\}}. \quad (20)$$

Note that the lowest order expansion of the denominator in Eq. (19) about $\tau_1 = \tau_2 = 0$ is kept under the entire range of integration since the important integration interval is on the order $\tau \sim \rho_0^2 \ll \rho_0 \sim \frac{1}{\sqrt{n_F}} \ll 1$. Note also that throughout this paper, we assume that $n_F \gg 1$.

It is convenient to rescale variables as

$$u \equiv \sqrt{n_F}\rho_0, \quad x_0 \equiv \xi q, \quad v \equiv \frac{\xi k_z}{\sqrt{n_F}} \quad (21)$$

and neglect the energy of an electron pair along the z axis, $(\xi q)^2$, with respect to the Fermi energy, n_F. By performing the explicit summation over the Matsubara frequencies, one obtains, in terms of the new variables, the following result:

$$A_0^{(d)} = N(0)\lambda_d \frac{2\pi k_B T}{\sqrt{\mu\hbar\omega_c}} \int_0^{\infty} du \frac{1 - e^{-(2\omega_D/\sqrt{\mu\hbar\omega_c})u}}{\sinh\left(\frac{2\pi k_B T}{\sqrt{\mu\hbar\omega_c}} u\right)} e^{-(2\tilde{\Gamma}/\sqrt{n_F})u} \times \cos\left(\frac{2g}{\sqrt{n_F}} u\right) \Theta_2^{(d)}(u, x_0), \quad (22)$$

where $\lambda_d = 3(\frac{k_F d}{\pi})^4$, $N(0)$ is the electron density of states per spin at the Fermi energy $[N(0) = \frac{\sqrt{m^* m_z^*} k_F}{2\pi\hbar^3}]$, and

$$\Theta_2^{(d)}(u, x_0) = \int_0^1 d\nu (1 - v^2)^2 \cos(2x_0 uv) e^{-u^2(1-v^2)}. \quad (23)$$

A similar result is obtained for an *s*-wave superconductor. In this case, $\lambda_d \to \lambda_s = 1$ and

$$\Theta_2^{(s)}(u, x_0) = \int_0^1 d\nu \cos(2x_0 uv) e^{-u^2(1-v^2)}. \quad (24)$$

For $g = x_0 = \tilde{\Gamma} = 0$, Eqs. (22) and (24) reduce to the quadratic term derived by Helfand–Werthammer.[32]

B. Quartic term

The quartic term in the perturbation expansion [Eq. (1)], which corresponds to a closed loop diagram with four vertices, is given by

$$\Omega_4^{(s,d)} = \frac{1}{\mathcal{V}_0} \int d^3\mathbf{R}_1 d^3\mathbf{R}_2 d^3\mathbf{R}_3 d^3\mathbf{R}_4 \times \tilde{\Gamma}_4(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, \mathbf{R}_4) \tilde{K}_4(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, \mathbf{R}_4), \quad (25)$$

where the kernel, containing the gauge invariant factors of the four electron Green's functions, is

$$\begin{aligned} \tilde{K}_4(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, \mathbf{R}_4) &= k_B T \sum_\nu \int d^3\mathbf{r}_1 d^3\mathbf{r}_2 d^3\mathbf{r}_3 d^3\mathbf{r}_4 \varphi(\mathbf{r}_1)\varphi^*(\mathbf{r}_2)\varphi(\mathbf{r}_3)\varphi^*(\mathbf{r}_4) \\ &\times G_{0\uparrow}^*\left(\mathbf{R}_2 - \mathbf{R}_1 + \frac{\mathbf{r}_2 - \mathbf{r}_1}{2}, \omega_\nu\right) \\ &\times G_{0\downarrow}\left(\mathbf{R}_3 - \mathbf{R}_2 - \frac{\mathbf{r}_3 - \mathbf{r}_2}{2}, \omega_\nu\right) \\ &\times G_{0\uparrow}^*\left(\mathbf{R}_4 - \mathbf{R}_3 + \frac{\mathbf{r}_4 - \mathbf{r}_3}{2}, \omega_\nu\right) \\ &\times G_{0\downarrow}\left(\mathbf{R}_1 - \mathbf{R}_4 - \frac{\mathbf{r}_1 - \mathbf{r}_4}{2}, \omega_\nu\right) \end{aligned} \quad (26)$$

and the vertex part is

$$\tilde{\Gamma}_4(\mathbf{R}_1, \mathbf{R}_2, \mathbf{R}_3, \mathbf{R}_4) = g^*(\mathbf{R}_1, \mathbf{R}_2) g(\mathbf{R}_2, \mathbf{R}_3) g^*(\mathbf{R}_3, \mathbf{R}_4) g(\mathbf{R}_4, \mathbf{R}_1) \times \Delta(\mathbf{R}_1)\Delta^*(\mathbf{R}_2)\Delta(\mathbf{R}_3)\Delta^*(\mathbf{R}_4), \quad (27)$$

which consists of the gauge factors $g(\mathbf{R}_i, \mathbf{R}_j)$ and the order parameter values at the four center of mass positions for two electron pairs.

Since the dependence of the order parameter on the relative pair coordinates is separable from that of the center of mass coordinates, the latter dependence is selected to have the usual Abrikosov lattice structure,

$$\Delta(\mathbf{R}) = c_0 e^{iqZ} e^{-(1/2)(|u|^2 - u^2)} \sum_{n=0,\pm1,\pm2,\ldots} e^{iq_n u - q_n^2/4}, \quad (28)$$

where $q_n = 2\pi n/a_x$ and $u = X + iY$. To simplify the calculation of the vertex part, we exploit several assumptions. By substituting Eq. (28) for Eq. (27), one may keep only the diag-

onal terms with $q_{n1}=q_{n2}=q_{n3}=q_{n4}=p$, since all off-diagonal terms are small by the Gaussian factor $\sim\exp[-(q_{n4}-q_{n1})^2-(q_{n4}-q_{n1})^2]$. Furthermore, we may replace the summation over p with an appropriate integration. Both of these assumptions are equivalent to neglecting particular vortex lattice structures, corresponding to the replacement of the Abrikosov structure parameter, β_A, with $\frac{\sqrt{\pi}}{a_x}$,[27] which yields only a small error.

With the above assumptions, the vertex part reduces to

$$\tilde{\Gamma}_4(\mathbf{R}_1,\mathbf{R}_2,\mathbf{R}_3,\mathbf{R}_4)=\frac{a_x\sqrt{\pi}}{2\pi}|c_0|^4\exp[iq(Z_1-Z_2+Z_3-Z_4)]\times\exp\left(-\frac{1}{4}\sum|\rho_l|^2\right)\times\exp\{1/4[(u_1-u_3)^2+(u_2^*-u_4^*)^2]\},\tag{29}$$

where $\rho_l=u_{l+1}-u_l$. Since the dominant contribution to the quartic term arises from small propagation distances, $|u_l|\leq 1$,[24,33] one may expand the last exponential on the right hand side of Eq. (29), up to leading order, under the integrals over the angular variables in Eq. (25). An additional angular dependence is due to the kernel, K_4, through its dependence on the absolute values of linear combinations of "external," $\mathbf{R}_{l+1}-\mathbf{R}_l$, and "internal," $\mathbf{r}_{l+1}-\mathbf{r}_l$ $(l=1,\ldots,4)$, coordinates [see Eq. (26)]. Since the characteristic size of $|\mathbf{r}_{l+1}-\mathbf{r}_l|\sim d$ is much smaller than the scale of $|\mathbf{R}_{l+1}-\mathbf{R}_l|\sim a_H$, the dependence of the kernel on $\mathbf{r}_{l+1}-\mathbf{r}_l$ (and, consequently, its dependence on the angular variables) may be neglected at large $|\mathbf{R}_{l+1}-\mathbf{R}_l|$. Therefore, the integration over angular variables in this region involves only the last exponential in Eq. (29), resulting in

$$\langle e^{(1/4)[(u_1-u_3)^2+(u_2^*-u_4^*)^2]}\rangle\approx 1+\frac{1}{4}\langle(u_1-u_3)^2+(u_2^*-u_4^*)^2\rangle=1,$$

since $\langle u_l^2\rangle=\langle u_l u_k^*\rangle=0$ $(l\neq k)$, whereas for small values of $|\mathbf{R}_{l+1}-\mathbf{R}_l|$, this exponential is always close to 1 and the remaining integration over angular variables can be performed in closed form (see below). Thus, one can approximate the vertex part by the following simple expression:

$$\tilde{\Gamma}_4(\mathbf{R}_1,\mathbf{R}_2,\mathbf{R}_3,\mathbf{R}_4)=\frac{a_x\sqrt{\pi}}{2\pi}|c_0|^4\exp[iq(Z_1-Z_2+Z_3-Z_4)]\times\exp\left(-\frac{1}{4}\sum|\rho_l|^2\right),\tag{30}$$

which depends only on nearest neighboring coordinates.

By making use of Eq. (30), the remaining calculation of the quartic term is similar to that used for the quadratic term but is considerably massier. Below, we present only an outline of the derivation. Since integrations over z_i are trivial, from now on, we shall use only 2D vector notations with integrations over Z_i that are explicitly written.

Combining Eqs. (25), (26), and (30), our starting expression for the quartic term is given by

$$\Omega_4^{(s,d)}=\frac{k_BT}{V_0}\left(\frac{1}{2\pi\hbar\omega_c}\right)^4\frac{a_x\sqrt{\pi}}{2\pi L_z}|c_0|^4\sum_\nu\int dZ_1dZ_2dZ_3dZ_4\times\int\prod_{i=1}^4\frac{dk_{z,i}}{2\pi}e^{-ik_{z,1}(Z_2-Z_1)}e^{ik_{z,2}(Z_3-Z_2)}e^{-ik_{z,3}(Z_4-Z_3)}\times e^{ik_{z,4}(Z_1-Z_4)}e^{iq(Z_1-Z_2+Z_3-Z_4)}\int d^2\mathbf{r}_1d^2\mathbf{r}_2d^2\mathbf{r}_3d^2\mathbf{r}_4\times f(\mathbf{r}_1)f^*(\mathbf{r}_2)f(\mathbf{r}_3)f^*(\mathbf{r}_4)\Theta_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4;\{k_{z,i}\};\omega_\nu),\tag{31}$$

where the function $\Theta_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4;\{k_{z,i}\};\omega_\nu)$ includes integration over all electron pair coordinates:

$$\begin{aligned}\Theta_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4;\{k_{z,i}\};\omega_\nu)&=\frac{1}{L_xL_y}\int d^2\mathbf{R}_1d^2\mathbf{R}_2d^2\mathbf{R}_3d^2\mathbf{R}_4\exp\left(-\frac{1}{4}\sum|\rho_l|^2\right)\\&\times\int_0^\infty d\tau_1e^{-i\tau_1[n_F-x_1^2-g-i\tilde{\omega}_v-i\tilde{\Gamma}]}\frac{\exp\left(\frac{R_{12}^2}{4}\frac{1+e^{-i\tau_1}}{1-e^{-i\tau_1}}\right)}{1-e^{i\tau_1}}\\&\times\int_0^\infty d\tau_2e^{i\tau_2[n_F-x_2^2+g+i\tilde{\omega}_v+i\tilde{\Gamma}]}\frac{\exp\left(\frac{R_{23}^2}{4}\frac{1+e^{i\tau_2}}{1-e^{i\tau_2}}\right)}{1-e^{-i\tau_2}}\\&\times\int_0^\infty d\tau_3e^{-i\tau_3[n_F-x_3^2-g-i\tilde{\omega}_v-i\tilde{\Gamma}]}\frac{\exp\left(\frac{R_{34}^2}{4}\frac{1+e^{-i\tau_3}}{1-e^{i\tau_3}}\right)}{1-e^{i\tau_3}}\\&\times\int_0^\infty d\tau_4e^{i\tau_4[n_F-x_4^2+g+i\tilde{\omega}_v+i\tilde{\Gamma}]}\frac{\exp\left(\frac{R_{41}^2}{4}\frac{1+e^{i\tau_4}}{1-e^{-i\tau_4}}\right)}{1-e^{-i\tau_4}}.\end{aligned}\tag{32}$$

Here, the coordinates, $\mathbf{R}_{i,i+1}$, in Eq. (32) are the linear combinations of $\rho_l=\mathbf{R}_{l+1}-\mathbf{R}_l$ and $\eta_l=\mathbf{r}_{l+1}-\mathbf{r}_l$:

$$\mathbf{R}_{12}=\rho_1+\frac{1}{2}\eta_1,\quad \mathbf{R}_{23}=\rho_2-\frac{1}{2}\eta_2,$$

$$\mathbf{R}_{34}=\rho_3+\frac{1}{2}\eta_3,\quad \mathbf{R}_{41}=\rho_4-\frac{1}{2}\eta_4.\tag{33}$$

The Gaussian integration over ρ_l reduces Eq. (32) to

$$\begin{aligned}\Theta_4(\mathbf{r}_1,\mathbf{r}_2,\mathbf{r}_3,\mathbf{r}_4;\{k_{z,i}\};\omega_\nu)&=\int_0^\infty d\tau_1d\tau_2d\tau_3d\tau_4\frac{(2\pi)^3}{\varkappa}\\&\times\exp\left[-\frac{1}{8\varkappa}(\eta_1-\eta_2+\eta_3-\eta_4)^2\right]e^{-i\tau_1[n_F-x_1^2-g-i\tilde{\omega}_v-i\tilde{\Gamma}]}\\&\times e^{i\tau_2[n_F-x_2^2+g+i\tilde{\omega}_v+i\tilde{\Gamma}]}e^{-i\tau_3[n_F-x_3^2-g-i\tilde{\omega}_v-i\tilde{\Gamma}]}\\&\times e^{i\tau_4[n_F-x_4^2+g+i\tilde{\omega}_v+i\tilde{\Gamma}]},\end{aligned}\tag{34}$$

where $\varkappa=4-e^{i\tau_1}-e^{-i\tau_2}-e^{i\tau_3}-e^{-i\tau_4}$. It should be noted here that Eq. (34) has been obtained by exploiting the fact that the dominant contributions to the integrals originate in the regions where $\tau_i \ll 1$.

Furthermore, by noting that in the above equation the η_i and k_z dependences are factorized, one can separately perform the integrations over both sets of variables. For a d-wave superconductor, we obtain

$$\int d^2\mathbf{r}_1 d^2\mathbf{r}_2 d^2\mathbf{r}_3 d^2\mathbf{r}_4 f(\mathbf{r}_1) f^*(\mathbf{r}_2) f(\mathbf{r}_3) f^*(\mathbf{r}_4) \times \exp\left[-\frac{1}{8\varkappa}(\eta_1-\eta_2+\eta_3-\eta_4)^2\right] = \frac{1}{4^3}(1-e^{-d^2/\chi})^4(3+2e^{-d^2/\chi}+e^{-2(d^2/\chi)})^2 \simeq \frac{9}{16}\frac{d^8}{\varkappa^4}, \tag{35}$$

where the last approximation is valid under the same conditions discussed in the derivation of the quadratic term. Thus, the quartic term is transformed to

$$\Omega_4^{(d)} = \frac{k_B T}{(\hbar\omega_c)^4}\frac{a_x\sqrt{\pi}}{(2\pi)^3}|c_0|^4\frac{9d^8}{16a_H^8}\sum_\nu \int dk_z \times \int_0^\infty \frac{d\tau_1 d\tau_2 d\tau_3 d\tau_4}{(4-e^{i\tau_1}-e^{-i\tau_2}-e^{i\tau_3}-e^{-i\tau_4})^5} \times e^{-i(\tau_1-\tau_2+\tau_3-\tau_4)(n_F-\xi^2k_z^2-\xi^2(q/2)^2)} e^{-(\tau_1+\tau_2+\tau_3+\tau_4)(\tilde{\omega}_\nu+\tilde{\Gamma})} \times e^{i(\tau_1+\tau_2+\tau_3+\tau_4)(g+\xi^2k_z q)}, \tag{36}$$

where an additional integration over $\zeta=(\tau_1-\tau_2+\tau_3-\tau_4)/2$ for small τ_i can be performed by expanding $e^{i\tau_i}$ in the denominator of Eq. (36) up to the second order. By rescaling the remaining independent variables as

$$\varrho = \sqrt{n_F}\frac{\tau_1+\tau_2+\tau_3+\tau_4}{2}, \quad s = \frac{\sqrt{n_F}}{2}(\tau_3-\tau_1),$$

and summing up over ν, one obtains the final result for the quartic term as follows:

$$\Omega_4^{(d)} = B^{(d)}\Delta_0^4\int_0^\infty d\varrho \frac{1-e^{-(2\omega_D/\sqrt{\mu\hbar\omega_c})\varrho}}{\sinh\left(\frac{2\pi k_B T}{\sqrt{\mu\hbar\omega_c}}\varrho\right)} e^{-(2\tilde{\Gamma}/\sqrt{n_F})\varrho} \times \cos(2\varrho g_0)\Theta_4^{(d)}(\varrho,q), \tag{37}$$

where $B^{(d)}=c_4^{(d)}B_0$ with $c_4^{(d)}=\frac{3}{16}(\frac{k_F d}{\pi})^8$, $B_0=(\frac{\sqrt{\pi}}{a_x})\frac{\pi k_B T}{(\mu\hbar\omega_c)^{3/2}}N(0)$, and

$$\Theta_4^{(d)}(\varrho,q) = \int_0^1 dv(1-v^2)^4 e^{-(1/2)\varrho^2(1-v^2)}\cos(2vq\varrho) \times \left(\int_0^\varrho ds e^{-s^2(1-v^2)}\right)^2. \tag{38}$$

The result for an s-wave superconductor can be obtained from Eq. (37) by replacing the factor $(1-v^2)^4$ in the definition of $\Theta_4^{(d)}$ and the factor $c_4^{(d)}$ in the normalization coefficient $B^{(d)}$ with unity. The s-wave quartic term for zero spin splitting is equivalent to that obtained in Ref. 24.

III. RESULTS AND DISCUSSION

The analysis presented in the previous sections enables us to write a GL-type expansion of the SC contribution to the thermodynamic potential for an s- or $d_{x^2-y^2}$-wave pairing up to the second order in Δ_0^2 as follows:

$$\Omega^{(s,d)} = \alpha^{(s,d)}(t,b,q)\Delta_0^2 + \frac{1}{2}\beta^{(s,d)}(t,b,q)\Delta_0^4 + \frac{1}{3}\gamma^{(s,d)}(t,b,q)\Delta_0^6 + \cdots. \tag{39}$$

For the quadratic term, we have

$$\alpha^{(s,d)}(t,b,q) = N(0)\left[\frac{1}{\lambda} - \frac{c_2^{(s,d)}}{\varsigma(T)}\int_0^\infty d\rho \frac{(1-e^{-2\rho/\varsigma(T_D)})}{\sinh\left(\frac{2\rho}{\varsigma(T)}\right)} e^{-2\rho/l} \times \cos\left(\frac{2g}{r_F}\rho\right)\Theta_2^{(s,d)}(\rho,q)\right],$$

$$\Theta_2^{(s,d)}(\rho,q) = \int_0^{v_0} d\nu \vartheta_2^{(s,d)}(v)\cos(q\rho v/\chi_a) \times \exp[-(1-v^2)\rho^2/2a_H^2], \tag{40}$$

where $\lambda=N(0)V$; $\varsigma(T_D)\equiv\hbar v_F/\pi k_B T_D$, where T_D is the Debye temperature and $v_F=\sqrt{2\varepsilon_F/m^*}$ is the in-plane Fermi velocity; $\varsigma(T)\equiv\hbar v_F/\pi k_B T$ is the thermal mean free path; $r_F=\sqrt{2n_F}a_H$ is the electronic cyclotron radius at the Fermi energy; and l the mean free path due to impurity scattering.

The differences between s-wave and d-wave SCs are given by $c_2^{(s)}=1$, $c_2^{(d)}=3(\frac{k_F d}{\pi})^4$, and $\vartheta_2^{(s)}(v)=1$, $\vartheta_2^{(d)}(v)=(1-v^2)^2$.

The quartic term has a similar structure:

$$\beta^{(s,d)}(t,b,q) = B_0\frac{c_4^{(s,d)}}{a_H}\int_0^\infty d\rho \frac{(1-e^{-2\rho/\varsigma(T_D)})}{\sinh\left(\frac{2\rho}{\varsigma(T)}\right)} e^{-2\rho/l} \times \cos\left(\frac{2g}{r_F}\rho\right)\Theta_4^{(s,d)}(\rho,q),$$

$$\Theta_4^{(s,d)}(\rho,q) = \int_0^{v_0} d\nu \vartheta_2^{(s,d)}(v)\cos(q\rho v/\chi_a)\exp[-(1-v^2) \times \rho^2/4a_H^2]\left(\int_0^{\rho/\sqrt{2}a_H} ds e^{-s^2(1-v^2)}\right)^2, \tag{41}$$

where $B_0=(\frac{\sqrt{\pi}}{a_x})\frac{N(0)\pi k_B T}{(\varepsilon_F\hbar\omega_c)^{3/2}}$ and $c_4^{(s)}=1$, $c_4^{(d)}=\frac{3}{16}(\frac{k_F d}{\pi})^8$, $\vartheta_4^{(s)}(v)=1$, and $\vartheta_4^{(d)}(v)=(1-v^2)^4$.

On the basis of the above formulas, below, we discuss the H-T phase diagram for different values of the relevant pa-

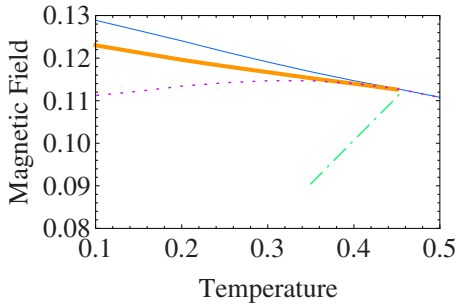

FIG. 1. (Color online) Phase transition lines for a 3D system with s-wave pairing: Shown are the second-order N-SC transitions for order parameters with FFLO modulation (thin solid line) and without FFLO modulation (dotted line). The FFLO-BCS transition obtained by minimizing the GL free energy (including quartic corrections) with respect to the FFLO wave number, which is of the first order, is represented by the thick solid line. The dashed-dotted line represents the location of vanishing $d\alpha/dq^2$ (see text for details). The value of the spin-splitting parameter is $\sigma=1.8$.

rameters. Three independent dimensionless parameters, $(2a_H/r_F)g$, $2a_H/\varsigma(T)$, and qa_H/χ_a, control the basic integrals in these equations. The first two parameters measure the strength of the spin and thermal pair-breaking mechanisms, respectively, relative to the orbital (diamagnetic) depairing. The third parameter determines the relative strength of the compensating FFLO mechanism. The value of the spin pair-breaking parameter, $\sigma \equiv g(2a_H/r_F)_{H=H_{c20}^{\rm orb}}$, where $H_{c20}^{\rm orb}$ is the upper critical field at $T=0$ in the absence of spin pair breaking, is related to the well known Maki parameter,[9] $\alpha_M = \frac{(\hbar e/m_0 c)H_{c20}^{\rm orb}}{1.76 k_B T_{c0}}$, by $\sigma=1.1\alpha_M$. Here, T_{c0} is the transition temperature at zero magnetic field.

As we shall show below, the situation $T_{\rm FFLO}>T^*$, where T^* is the temperature at which $\beta(t,b_{c2},q=0)=0$, is realized in 3D systems (corresponding to $v_0=1$), regardless of the spin-splitting strength and the type of electron pairing. A typical phase diagram is shown in Fig. 1 for s-wave pairing and spin pair-breaking parameter $\sigma=3$.

As long as $T>T^*$ [so that $\beta(t,b_{c2},q=0)>0$], the N-SC phase transition is of second order and the (reduced) critical field, $b_{c2}(t)$, can be determined as the maximal value of $b \equiv H/H_{c20}^{\rm orb}$ obtained from the equation $\alpha(t,b,q)=0$ for all values of q, at the (reduced) temperature $t \equiv T/T_{c0}$. The solution of this equation for $q=0$ yields a transition line, $b=b_{c2}^{(0)}(t)$, ignoring the possibility of a FFLO state. The tricritical point, $T_{\rm FFLO}$, is defined as the maximal temperature at which $b_{c2}(t)>b_{c2}^{(0)}(t)$. It can be alternatively determined from the equation $\frac{d\alpha(t,b_{c2},q=0)}{dq^2}=0$, which is equivalent to the condition for vanishing of the coefficient of $|\nabla\Delta|^2$ in a gradient expansion of the SC free energy.[18]

For $T<T^*$ and sufficiently strong spin pair breaking, there can be a changeover to first-order SC transitions, but since $\beta(t,b_{c2},q\neq 0)>\beta(t,b_{c2},q=0)$ (see Fig. 2), the segment of the $b_{c2}(t)$ line with first-order transitions arises only at very low temperatures. For moderate σ values, the coefficient $\beta(t,b_{c2},q)$ at optimal q is always positive and the N-SC transition is of the second order at an arbitrarily low temperature.

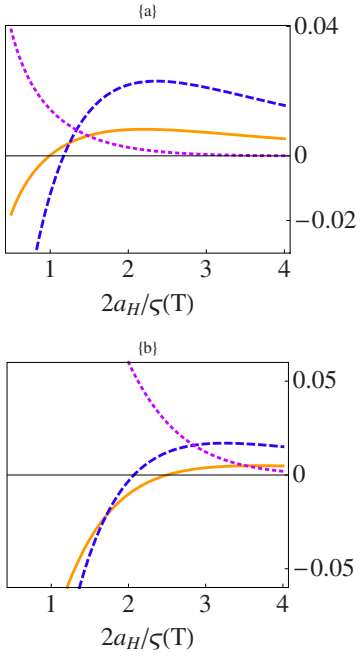

FIG. 2. (Color online) The GL coefficients $\beta(t,b,q=0)$ (dashed lines), $\frac{d\alpha(t,b,q=0)}{dq^2}$ (solid lines), and $\frac{d\beta(t,b,q=0)}{dq^2}$ (dotted lines) as functions of the parameter $\frac{2a_H}{\varsigma(T)}$ for (a) $v_0=0.4$ and (b) $v_0=1$. The curves are plotted with α measured in units of $N(0)$ and β in units of $N(0)\sqrt{\pi}/a_x\varepsilon_F\hbar\omega_c$.

The transition within the SC region from the nonuniform (FFLO) to uniform (BCS) phase at $T>T^*$ cannot be obtained just by analyzing the quadratic term $\alpha(t,b,q)$ since the SC order parameter is finite there. It can be obtained by minimizing the SC free energy (including both quadratic and quartic terms) with respect to the modulation wave number q. By neglecting the sixth and higher order terms in the expansion, the corresponding (standard) GL free energy, $\Omega(q) \simeq -\theta(\alpha)\frac{\alpha^2}{2\beta}$ [$\theta(\alpha)$ being the Heaviside step function], which has a single minimum at $q\neq 0$ for a field near b_{c2} (see Fig. 3, upper panel), develops a double-well structure (see Fig. 3, middle panel) as a function of q upon decreasing the field below b_{c2} at a given temperature T (due to the symmetry $q\leftrightarrow -q$, only positive values may be considered). One of these minima is always at $q=0$, and it becomes energetically favorable at a critical field for a first-order phase transition from the FFLO to the uniform BCS phase. The second (metastable) minimum at $q\neq 0$ completely disappears upon further field decrease (see Fig. 3, lower panel). The origin of the developing minimum at $q=0$ is the quickly decreasing values of $\beta(q)$ with the decreasing values of q toward $q=0$, where α is negative.

At temperatures T below T^*, the first two terms in the expansion of the thermodynamic potential are not sufficient to correctly describe the uniform SC state since for negative β values, the scale of the SC free energy is determined by the sixth-order term. In contrast, the free energy of the nonuniform state, where $\beta(t,b,q\neq 0)>0$, can be obtained from the standard GL functional (with the assumption that the contribution of the sixth-order term is small compared to that of the quartic term). The characteristic q dependences of the GL

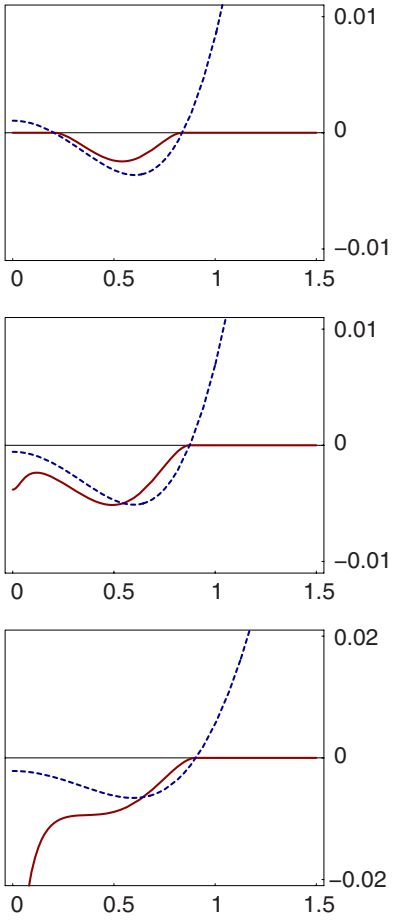

FIG. 3. (Color online) The dependence of the GL coefficient α (dashed lines) and the mean-field SC free energy, $-\theta(\alpha)\frac{\alpha^2}{2\beta_A\beta}$ (solid lines), on the modulation parameter, $\tilde{q}\equiv(\frac{2}{\sigma})\sqrt{\frac{m_x^*}{m_z^*}}(qa_{H_{c20}^{\rm orb}})$, in a 3D system ($v_0=1$) are shown at $t=0.4$ and a decreasing magnetic field. The value of the spin-splitting parameter is $\sigma=3$. Upper panel: $b=0.1142$, i.e., near the tricritical point, just below the normal-FFLO SC transition. It is seen that $\alpha<0$ in a small region around $\tilde{q}=0.6$, where the SC free energy has a minimum. Middle panel: $b=0.114$, where a uniform ($q=0$) metastable SC state is present. Lower panel: $b=0.1139$, where a uniform ($q=0$) equilibrium SC state is present, while a metastable SC state exists at $q\neq 0$.

coefficients, α and β, and the mean-field free energy, $-\theta(\alpha)\frac{\alpha^2}{2\beta_A\beta}$, for $T<T^*$ are illustrated in Fig. 4. Whereas at high fields (Fig. 4, upper panel) the minimum of the SC energy occurs in a region where $\beta>0$, at lower fields (see Fig. 4, lower panel) it approaches the expanding temperature domain of negative β. Thus, even for moderate spin splitting and low temperature, the transition line from a nonuniform to a uniform SC state cannot be determined without knowing the sixth-order term. It is clear, however, that this transition is of the first order.

It should be noted that if one attempts to determine the FFLO-BCS phase boundary from the equation $\frac{d\alpha(t,b,q=0)}{dq^2}=0$, it will greatly overestimate the size of the FFLO phase as compared to that obtained by minimizing $-\frac{\alpha^2}{2\beta}$ (see Fig. 1). This remarkable difference is due to the strong q^2 dependence of the quartic coefficient β (see Fig. 2).

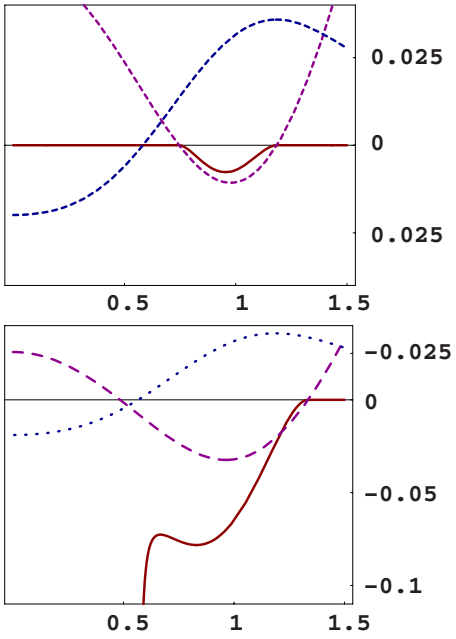

FIG. 4. (Color online) The GL coefficients, α (dashed lines) and β (dotted lines), and the mean-field SC free energy (solid lines) as functions of the modulation parameter, $\tilde{q}$, in a 3D system ($v_0=1$) at a relatively low temperature $t=0.25$ and decreasing field values $b=0.118$ (upper panel) and $b=0.117$ (lower panel). Note the vanishing of β inside the region where $\alpha<0$, around which the used approximation, $-\theta(\alpha)\frac{\alpha^2}{2\beta_A\beta}$, for $\Omega(q)$ breaks down.

The suppression of the orbital effect in the considered 3D systems, with ellipsoidal Fermi surfaces contained entirely within the BZ, is due to the factor $1-v^2$ appearing in the Gaussian exponents of Eqs. (40) and (41). The recovery of this effect in quasi-2D systems with truncated ellipsoidal Fermi surface, where $v_0<1$, can reverse the relation between $T_{\rm FFLO}$ and T^*. Figure 2(b), in which the GL coefficients are shown for $\sigma=1.8$, $v_0=0.4$ and s-wave pairing, illustrates the situation with $T_{\rm FFLO}<T^*$, which occurs for all values of v_0 below a critical dimensionality $v_{0,\rm cr}\approx 0.44$ (see Fig. 5), and only weakly depends on the spin-splitting parameter σ.

The corresponding phase diagram (see Fig. 6) for v_0 below this crossing point is quite different from that found for the 3D systems shown in Fig. 1. First of all, since $\beta<0$, one may use Eqs. (40) and (41) to determine the phase diagram only under the assumption that the sixth-order coefficient γ is positive (see Ref. 19). In this case, a discontinuous SC transition occurs at $\Omega(\Delta_0^2)=0$ with $\Delta_0^2=(\frac{3|\beta|}{4\gamma})$ and $\alpha=\frac{3\beta^2}{16\gamma}>0$, and the corresponding critical field, $b_{c2}(t)$, should be larger than $b_{c2}^0(t)$, which is obtained from the equation $\alpha(q=0)=0$. Thus, at a temperature below T^*, the N-SC phase boundary includes a segment of first-order transitions, which may end at zero temperature or at a finite temperature, depending on the spin-splitting strength. This dependence appears because of the competition between the decreasing explicit dependence of β on decreasing temperature and its increasing implicit dependence through $q(T)$ at the FFLO state. The boundary between the BCS and FFLO states should be determined by minimizing the free energy [Eq. (39)] with respect to q. This may be restricted to the explicit dependence

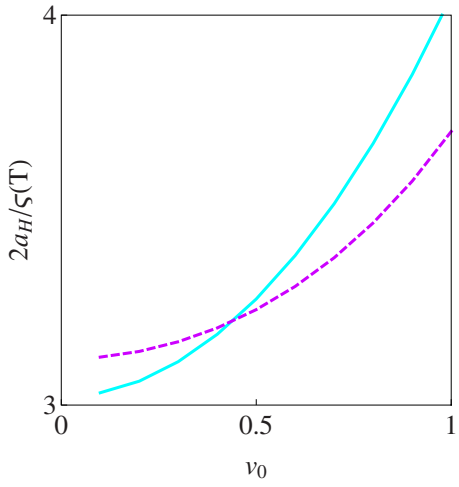

FIG. 5. (Color online) Solutions $\frac{2a_H}{\varsigma(T)} \propto T$ of the equation $\beta(q=0)=0$, which corresponds to T^* (dashed line), and the equation $\frac{d\alpha(q=0)}{dq^2}=0$, which corresponds to T_{FFLO} (solid line), vs the dimensionality crossover parameter v_0 for $\sigma=2.5$.

on q since the order parameter is determined by $\frac{\partial\Omega}{\partial\Delta^2}=0$. Consequently, the positive sign of $\frac{d\beta(t,b,q=0)}{dq^2}$ [see Fig. 2(b)] results in a partial cancellation of the leading contribution to $\frac{\partial\Omega}{\partial q^2}$, which is proportional to $\frac{d\alpha(t,b,q=0)}{dq^2}$ and negative in the FFLO part of the phase diagram. Moreover, since for the discontinuous transition the order parameter is finite just below the transition, the higher order terms in Δ_0^2 [Eq. (39)] should be taken into account. As a result, T_{FFLO} should be smaller than T^0_{FFLO}, which is the temperature obtained from the equation $\frac{d\alpha(t,b,q=0)}{dq^2}=0$, as schematically shown in Fig. 6.

IV. CONCLUSIONS

It is shown that the expected changeover to first-order SC transitions in clean, strongly type-II superconductors in the Pauli paramagnetic limit can take place in materials with quasicylindrical Fermi surfaces, regardless of the type of the electron (s- or d-wave) pairing interaction, which leads to superconductivity. This finding clarifies the debate on this topic in literature.[17–20]

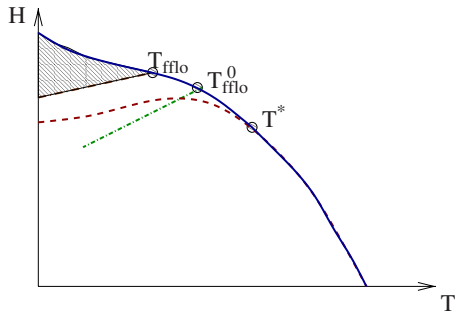

FIG. 6. (Color online) Schematic phase diagram for a quasi-2D system. The shaded area corresponds to a nonuniform SC (FFLO) phase, the dashed line corresponds to $\alpha(t,b,q=0)=0$, and the dashed-dotted line can be obtained from $\frac{d}{dq^2}\alpha(t,b,q=0)=0$.

The observation of such a changeover in the heavy-fermion compound $CeCoIn_5$ for a magnetic field orientation perpendicular to the easy conducting plane[11] is consistent with the quasi-2D character of its electronic band structure.[34] The interesting situation of a 2D superconductor under a magnetic field parallel to the conducting plane, for which a changeover to discontinuous SC transitions was very recently reported,[12] is more subtle since the vanishingly small cyclotron frequency characterizing this case does not allow the utilization of the LLL approximation employed here (for a recent review, see, e.g., Ref. 35). It should be noted that under certain circumstances, vortex states with higher Landau levels (HLLs) could be important even in the present situation of vortex lines parallel to a large magnetic field component. Adachi and Ikeda in Ref. 19, indeed, presented a phase diagram for a quasi-2D system with $H\|c$, in which $n=1$ LL was found to be dominant at sufficiently low temperatures.

In our opinion, this result should be considered with caution since it was obtained by analyzing only the quadratic term without accounting for q modulation and first-order phase transition. Both effects lead to an increase in the upper critical field for the LLL at low temperature, which can compete with the upper critical field for the $n=1$ LL. It should be stressed that the above discussion does not mean that the effect of HLL can be completely ignored. However, the problem is very subtle, and to correctly treat it, one should know the sixth-order term (at least for LLL), which is not yet available in our calculations.

Finally, in comparing our results to the available experimental data, one should note that for the typical values of the Maki parameter realized in the relevant materials, the first-order transition line, which separates the nonuniform SC phase from the uniform one, and the second-order N-SC transition line, which is predicted for superconductors with a 3D Fermi surface, are very close to each other. Thus, they might be indistinguishable in the presence of the strong thermal fluctuations expected for such a strongly type-II superconductor under high magnetic fields.[36]

Indeed, quite recently, a sharp rise of the thermal conductivity with the decreasing field just below H_{c2} at low temperatures was reported[37] for the heavy-fermion superconductor URu_2Si_2, whose Fermi surface may be characterized as 3D. This observation might indicate the existence of a latent heat associated with a first-order phase transition, which is not inconsistent with our theoretically predicted phase diagram for a 3D Fermi surface, since the width of the experimentally observed transition is $\sim 0.1H_{c2}$,[37] while the maximal field difference between the calculated transition lines shown in Fig. 1 is within $0.1H_{c2}$.

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