Quasiclassical free energy of superconductors: Disorder-driven first-order phase transition in superconductor/ferromagnetic-insulator bilayers

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In the seminal work by G. Eilenberger, Z. Phys. **214**, 195 (1968), a closed-form expression for the free energy of inhomogeneous spin-singlet superconductor in terms of quasiclassical propagators has been suggested. However, deriving this expression and generalizing it for superconductors or superfluids with general matrix structure, e.g., spin-triplet correlations, has remained problematic. Starting from the Luttinger-Ward formulation, we discuss here the general solution. Besides ordinary superconductors with various scattering mechanisms, the obtained free-energy functional can be used for systems, such as superfluid ^{3}He and superconducting systems with spatially inhomogeneous exchange field or spin-orbit coupling. Using this result, we derive the simplified expression for the free energy in the diffusive and hydrodynamic limits. As an example of using this formalism, we show that impurity scattering restores the first-order phase transition in superconductor-ferromagnetic insulator bilayers making this system similar to the bulk superconductor with the homogeneous built-in exchange field.

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I. INTRODUCTION

Quasiclassical approximation [1,2] is one of the basic tools in the theory of Fermi systems. It is based on the separation of scales when the characteristic wave numbers and frequencies of interest are much smaller than the Fermi wave vector and energy. The technique has been widely successful in the description of many superconductor (S)/superfluid systems and effects, including dirty superconductors [3], superfluid ^{3}He [4–6], superconducting hybrid structures [7,8], transport properties of mesoscopic superconducting devices [9], and superconductors with spin-splitting fields [10].

The quasiclassical equilibrium theory is, however, partially incomplete with regard to expressing the free energy directly in terms of the quasiclassical propagators. Such an expression has been introduced by Eilenberger [1] for the particular case of spin-singlet superconductors where the correlation functions have trivial spin structures. Different forms of variational functionals yielding the quasiclassical equations as their saddle points have also been discussed in the framework of nonlinear σ models [11–14]. Although the expression by Eilenberger has been used in many subsequent works, there appears to be an open question in how it relates to the general Luttinger-Ward free-energy functional, [4,15] which, in its typical formulation, requires (usually numerical) coupling constant integration [5,16]. Furthermore, its extension to systems with general matrix structures, e.g., spin-triplet superconducting correlations, has not been discussed. As a consequence, analytical results are available in limited tractable special cases.

In the present paper, we resolve the above issues by evaluating a coupling-constant integral [5,16] analytically, and obtain a free-energy functional in terms of propagators of general matrix structure. We demonstrate that different versions of the free energy discussed in the previous works [1,11,12,17] are recovered by the general Eilenberger-type expression and, for the sake of example, show how it can be reduced to a simpler form in the limit of a short mean free path and in the hydrodynamic limit. Moreover, we apply the results to study the superconducting phase transition in superconductor-ferromagnetic insulator (S/FI) bilayers [10,18,19] and discuss how impurity scattering changes the order of the phase transition.

This paper is structured as follows. In Sec. II, we derive a quasiclassical free-energy functional applicable to generic situations. In Sec. III, we show how the functional is simplified in the diffusive limit. In Sec. IV, we derive the free energy in the hydrodynamic limit both for the spin-singlet and for the spin-triplet pairing. In Sec. V, we discuss the S/FI systems and how they are affected by impurity scattering. We summarize the results in Sec. VI. Longer mathematical derivations can be found in the Appendices.

II. GENERAL FORMULATION

A general expression for the free energy of a many-body fermionic system has been derived by Luttinger and Ward [15]. Later, this expression has been adopted by Serene and Rainer [5] to describe the superfluidity of a Fermi liquid using the expansion in small parameters determined by the ratio of pairing energy to the Fermi energy. The same approach works for the BCS model of superconductivity in metals. This expansion is formulated in terms of the quasiclassical propagator [1],

$$\hat{g} = \frac{i}{\pi} \fint d\xi_p \hat{\tau}_3 \hat{G}, \tag{1}$$

where $\hat{G}(\boldsymbol{r}, \boldsymbol{p}, \omega)$ is the exact Green's function (GF) and $\xi_p = p^2/2m - E_F$ is the kinetic energy of electrons relative to the Fermi level.

The quasiclassical GF $\hat{g}(\boldsymbol{n}_p, \boldsymbol{r}, \omega)$ is a 4×4 matrix in a combined spin and Gor'kov-Nambu space and depends on the direction of quasiparticle momentum $\boldsymbol{n}_p = \boldsymbol{p}/p$, the position in real-space $\boldsymbol{r}$, and the Matsubara frequency ω.

Integration in (1) is implemented in the vicinity of the Fermi sphere, and the off-shell contribution is neglected resulting in the following expression for the free energy [5]:

$$\Omega = \frac{1}{2}\mathrm{Tr}\bigg[\hat{\Sigma}\hat{g} - \frac{1}{\pi}\int d\xi_p \ln\big(-i\hat{\Sigma} - \hat{G}_0^{-1}\big)\bigg] + \Phi[\hat{g}], \tag{2}$$

where $\hat{\Sigma}$ is the self-energy and the last term is the functional generating the self-energy $\hat{\Sigma} = -2\delta\Phi/\delta\hat{g}^T$. The normal- (N-) state part has been subtracted from Ω, Φ, ln, and $\hat{\Sigma}$.

The generalized trace operator in Eq. (2) defined as $\mathrm{Tr} = \pi T N_0 \sum_{\omega_n} \int \frac{d\Omega_p}{4\pi}$ tr contains a Matsubara sum, Nambu and spin traces, integration over $\boldsymbol{n}_p$ directions, and the density of states at the Fermi-level N_0.

The superconducting pairing is determined by a contribution to the generating functional in (3), $\Phi_\Delta[\hat{g}] = -\mathrm{Tr}(\hat{\Delta}[\hat{g}]\hat{g})/4$, where $\hat{\Delta} = \hat{\Delta}[\hat{g}]$ is given by the self-consistency relation for the gap function, which is a linear functional that describes all possible types of pairings. In addition, there are other contributions to Φ, e.g., from various scattering mechanisms, including potential impurity scattering, spin-orbital, and spin-flip relaxations [20].

The operator $\hat{G}_0^{-1} = i(\omega\hat{\tau}_3 + \boldsymbol{v}_F \cdot \hat{\nabla}) - \hat{V}$ contains a spatial derivative in the direction determined by the Fermi velocity $\boldsymbol{v}_F = v_F \boldsymbol{n}_p$ and the spin-dependent potential-energy $\hat{V} = \hat{V}(\boldsymbol{r})$. Therefore, calculation of the logarithmic term in (2) is rather nontrivial. One way to do this is based on the observation (cf. Ref. [16]) $\int d\xi_p \partial_\lambda \,\mathrm{Tr}\, \ln(-i\lambda\hat{\Sigma} - \hat{G}_0^{-1}) = \pi\ \mathrm{Tr}\ \hat{\Sigma}\hat{g}_\lambda$ resulting in the general expression for the free-energy density of a nonuniform superconductor or Fermi superfluid [4–6,16],

$$\Omega[\hat{g}, \hat{\Sigma}] = \frac{1}{2}\int_0^1 d\lambda\ \mathrm{Tr}[\hat{\Sigma}(\hat{g} - \hat{g}_\lambda)] + \Phi[\hat{g}], \tag{3}$$

$$0 = \boldsymbol{v}_F \cdot \check{\nabla}\hat{g}_\lambda + [\hat{M}_\lambda, \hat{g}_\lambda], \quad \hat{g}_\lambda^2 = 1. \tag{4}$$

We denote $\hat{M}_\lambda = \hat{\Lambda} + \lambda\hat{\Sigma}$ and $\hat{\Lambda} = (\omega + i\hat{V})\hat{\tau}_3$. Here, $\hat{g}_\lambda = \hat{g}_\lambda[\hat{\Sigma}]$ is the quasiclassical GF, regarded as a functional of the variational self-energy. It satisfies the Eilenberger equation and the normalization condition (4). The potential energy can include a Zeeman term $\hat{V} = \boldsymbol{\sigma} \cdot \boldsymbol{h}$ with a general texture of exchange field $\boldsymbol{h} = \boldsymbol{h}(\boldsymbol{r})$ as well as spin-orbit coupling (SOC). The latter, however, is more conveniently included in the covariant differential operator defined as $\check{\nabla}_k = \nabla_k - ie[\cdot, \hat{\tau}_3 A_k] - i[\cdot, \mathcal{A}_k]$, where A_k are the components of the vector potential and $\mathcal{A}_k = \mathcal{A}_{kj}\sigma_j$ is the SU(2) gauge field for the SOC.

Expression (3) can be used for any weakly coupled superconducting or superfluid state with arbitrary pairing interactions and fields $\boldsymbol{A}(\boldsymbol{r})$, $\boldsymbol{h}(\boldsymbol{r})$, and $\hat{\boldsymbol{A}}(\boldsymbol{r})$. However, the remaining λ integration necessitates solving Eq. (4) for the auxiliary propagator $\hat{g}_\lambda$ for many λ's. This makes the functional (3) less convenient for numerical work and hinders analytical calculations except in certain limiting cases, such as, e.g., in the dirty limit with small impurity scattering time τ_{imp} or in the Ginzburg-Landau regime close to the critical temperature.

A simpler free-energy functional without λ integration has been suggested by Eilenberger [1] for the particular case of a spin-singlet superconductor and in the absence of spin-rotating fields (i.e., collinear $\boldsymbol{h}$ and $\boldsymbol{A} = 0$) but without a systematical procedure for extending the result beyond this case. Below, we discuss a way to extend it.

A. λ integration

The λ integral in (3) can be evaluated using an approach suggested in Ref. [20]. Let us note the general relation,

$$\mathrm{Tr}[\hat{\Sigma}(\hat{g} - \hat{g}_\lambda)] = \partial_\lambda\,\mathrm{Tr}[\hat{M}_\lambda(\hat{g} - \hat{g}_\lambda)] + \mathrm{Tr}[\hat{M}_\lambda \partial_\lambda \hat{g}_\lambda]. \tag{5}$$

Here, the first term on the right-hand side is a full λ derivative and easily integrated, but further treatment is needed for the second term. To calculate its contribution, we note that the variation of GF preserving the normalization condition $\hat{g}^2 = 1$ can, in general, be written as $\delta g = [\delta\hat{W}, \hat{g}]$ where $\delta\hat{W}$ is a matrix with infinitesimal coefficients. Hence, the derivative can be represented as

$$\partial_\lambda \hat{g}_\lambda = [\hat{W}_\lambda, \hat{g}_\lambda]. \tag{6}$$

Using Eq. (4), the last term in Eq. (5) can be written as

$$\mathrm{Tr}[\hat{M}_\lambda \partial_\lambda \hat{g}_\lambda] = \mathrm{Tr}[(\boldsymbol{v}_F \cdot \check{\nabla} g_\lambda)\hat{W}_\lambda]. \tag{7}$$

To proceed, let us now assume that there exists a functional density $E[\hat{g}]$ whose variation over the GF components yields the gradient term

$$\delta \int d^3r E[\hat{g}] = \int d^3r\ \mathrm{Tr}[(\boldsymbol{v}_F \cdot \check{\nabla}\hat{g})\delta\hat{W}]. \tag{8}$$

Then, from Eq. (7) we get

$$\int d^3r\ \mathrm{Tr}[\hat{M}_\lambda \partial_\lambda \hat{g}_\lambda] = \frac{d}{d\lambda}\int d^3r\, E[\hat{g}_\lambda]. \tag{9}$$

Finally, we can perform the λ integration to obtain the general expression for the free-energy functional,

$$\Omega[\hat{g}, \hat{\Sigma}] = \tfrac{1}{2}E[\hat{g}_1[\hat{\Sigma}]] + \Phi[\hat{g}] + \tfrac{1}{2}\,\mathrm{Tr}[\hat{\Lambda}(\hat{g}_n - \hat{g})] + \tfrac{1}{2}\,\mathrm{Tr}\,[(\hat{\Lambda} + \hat{\Sigma})(\hat{g} - \hat{g}_1[\hat{\Sigma}])]. \tag{10}$$

where $\hat{g}_n \equiv \hat{g}_{\lambda=0} = \mathrm{sgn}(\omega)\hat{\sigma}_0\hat{\tau}_3$ and we have chosen $E[\hat{g}_n] = 0$. Using Eqs. (4), (8), the saddle-point equations $(\delta/\delta\hat{g})\Omega = 0$, $(\delta/\delta\hat{\Sigma})\Omega = 0$ can be reduced to $\hat{\Sigma}_* = -2(\delta/\delta\hat{g}^T)\Phi$ and $\hat{g}_* = \hat{g}_1[\hat{\Sigma}]$, which, indeed, correspond to the quasiclassical equations.

The value of the functional at the saddle point gives the free energy,

$$\Omega = \tfrac{1}{2}E[\hat{g}_*] + \Phi[\hat{g}_*] + \tfrac{1}{2}\,\mathrm{Tr}[\hat{\Lambda}(\hat{g}_n - \hat{g}_*)]. \tag{11}$$

The gradient functional $E[\hat{g}]$ remains to be determined.

B. The functional E

In spin-diagonal systems, the gradient terms of the expression given by Eilenberger [1] constitute $E[\hat{g}]$. In the presence of general spin-triplet correlations, the situation is more complicated, and we need to find a functional satisfying Eq. (8).

Let us first state the result,

$$E[\hat{g}] = \tfrac{1}{2}\,\mathrm{Tr}(\hat{g}[\hat{\tau}_t, \hat{g}]\boldsymbol{v}_F \cdot \check{\nabla}[\hat{\tau}_t, \hat{g}]^{-1}), \tag{12}$$

where $\hat{\tau}_t$ is an arbitrary matrix field normalized to $\hat{\tau}_t^2 = 1$. In the singlet case, we can denote $\hat{\tau}_t = \hat{\boldsymbol{\tau}} \cdot \boldsymbol{t}$ where $\hat{\boldsymbol{\tau}} = (\hat{\tau}_1, \hat{\tau}_2, \hat{\tau}_3)$ and $\boldsymbol{t} = (t_x, t_y, t_z)$ are vectors normalized as $t^2 = 1$. The field can be inhomogeneous in space. Indeed, using the properties $\hat{g}^2 = 1$, $\hat{\tau}_t^2 = 1$, and $\delta\hat{g} = [\delta\hat{W}, \hat{g}]$, a straightforward calculation (see Appendix A) yields the variation (8) for any texture $\hat{\tau}_t(\boldsymbol{r})$. The gradient functional is not unique.

The above functional can be found as follows: We first express the Green's function in terms of Riccati parameters [21–23] a, b which are $n \times n$ matrices ($n = 2$ if the GF has only Nambu + spin structure), and

$$\hat{g} = \begin{pmatrix} (1-ab)^{-1} & 0 \\ 0 & (1-ba)^{-1} \end{pmatrix}\begin{pmatrix} 1+ab & 2a \\ -2b & -1-ba \end{pmatrix}. \tag{13}$$

This form automatically satisfies the normalization condition $\hat{g}^2 = 1$. Moreover, the Eilenberger equations (4) imply that $\hat{a}, \hat{b}$ obey Riccati equations [21,23],

$$\boldsymbol{v}_F \cdot \check{\nabla} a - (2\omega + a\bar{\Delta})a + \Delta = 0, \tag{14}$$

$$\boldsymbol{v}_F \cdot \check{\nabla} b + (2\omega - b\Delta)b + \bar{\Delta} = 0. \tag{15}$$

It is relatively straightforward to find an Ansatz functional that has Riccati equations as its saddle point. For example, one can use the functional (11) with (see Appendix B and Ref. [24])

$$E = \mathrm{Tr}[(a^{-1} - b)(\boldsymbol{v}_F \cdot \check{\nabla})(a^{-1} + b)^{-1}]. \tag{16}$$

Rewriting (16) in a parametrization-independent way yields Eq. (12) with $\hat{\tau}_t = \hat{\tau}_3$. To obtain the free energy in a form similar to that suggested by Eilenberger, we can consider Nambu components of the quasiclassical propagator $\hat{g} = (g, f; \bar{f}, \bar{g})$ where the normal $g, \bar{g}$ and anomalous parts $f, \bar{f}$ have similar matrix structure as a, b. Then, the general form (12) with $\hat{\tau}_t = \hat{\tau}_3$ yields

$$E = \tfrac{1}{2}\,\mathrm{Tr}[gf(\boldsymbol{v}_F \cdot \check{\nabla})f^{-1} + \bar{g}\bar{f}(\boldsymbol{v}_F \cdot \check{\nabla})\bar{f}^{-1}], \tag{17}$$

which clearly reduces to Eilenberger's result in the spin-diagonal case.

The expression (12) is not defined at points where $[\hat{\tau}_t, \hat{g}_\lambda]$ is not invertible. Such points, if they occur inside the region swept by the λ integration, produce imaginary winding number contributions (see Appendix A). For example, in the singlet case, $E[\hat{g}_n] \hat{=} i\boldsymbol{v}_F \cdot \nabla\psi$ (excluding the Matsubara sum and angle average), where ψ is the polar angle of rotation of the unit vector $\boldsymbol{t}$ around the z axis. Since the free energy is real valued, such contributions are removed by taking the real part. Moreover, in practice, one should choose $\hat{\tau}$ to avoid singularities in $E[\hat{g}_{\lambda=1}]$. Close to the normal state where $\hat{g} \approx \hat{\tau}_3$, $\hat{\tau}_1$ is a stable choice. Alternatively, given a decomposition $\hat{g}_0(x) = U_0(x)^{-1}\hat{\tau}_3 U_0(x)$ for some fixed $\hat{g}_0(x) \approx \hat{g}(x)$, one can choose $\hat{\tau}_t(x) = U_0(x)^{-1}\hat{\tau}_1 U_0(x)$. This is also applicable in the spin-diagonal problem.

Writing $\hat{g} = \hat{U}\hat{\tau}_3\hat{U}^{-1}$, we can also recognize $\hat{W}_\lambda = (\partial_\lambda \hat{U})\hat{U}^{-1}$ so that

$$\int_M d\lambda\, ds\, \mathrm{tr}[\partial\hat{g}_\lambda \hat{W}_\lambda] = \int_M d(E_s ds + E_\lambda d\lambda), \tag{18}$$

where $M = [0, 1] \times [-\infty, \infty]$, $E_s = -\,\mathrm{tr}[\hat{\tau}_3\hat{U}_\lambda^{-1}\partial\hat{U}_\lambda]$, $E_\lambda = -\,\mathrm{tr}[\hat{\tau}_3\hat{U}_\lambda^{-1}\partial_\lambda\hat{U}_\lambda]$, and $\partial = \boldsymbol{n}_p \cdot \check{\nabla}$ is the long derivative vs the coordinate s along the quasiclassical trajectory. Hence, the gradient term can also be expressed as a Berry/Wess-Zumino term [25] associated with the quasiclassical Green's function. A kinetic term of this type was obtained in Refs. [11,12] for the action of the ballistic σ model, which is closely related to the present problem.

Finally, to evaluate the term in Eq. (10), we can substitute $\check{\nabla}\hat{g}$ from Eq. (4) into Eq. (12). Direct calculation gives (for $\check{\nabla}\hat{\tau}_t = 0$),

$$\Omega = \tfrac{1}{2}\,\mathrm{Tr}(\hat{\Sigma}\hat{g} + \hat{\Lambda}\hat{g}_n - [\hat{\tau}_t, \hat{\Lambda} + \hat{\Sigma}][\hat{\tau}_t, \hat{g}_1[\hat{\Sigma}]]^{-1}) + \Phi[\hat{g}], \tag{19}$$

whose real part is equal to Eq. (3) if integrated over space. Comparing to Eq. (2), we, hence, found an expression for the (subtracted) ξ_p-integrated logarithm, in terms of the quasiclassical propagator $\hat{g}$.

III. APPLICATION: DIFFUSIVE LIMIT

The free energy can be further simplified in the dirty limit when the impurity scattering rate τ_{imp}^{-1} is the largest among energy scales, apart from the Fermi energy. In this limit, we can eliminate the momentum integration and express the energy in terms of the momentum-averaged GF, which we denote as $g_s = \langle g \rangle$.

The expression which has been used [26–29] for the dirty superconductors with spin-singlet s-wave pairing described by the pairing constant V reads

$$\frac{F_s}{N_0} = \frac{|\Delta|^2}{V} - \frac{\pi T}{2}\sum_\omega \mathrm{tr}\Big\{(\omega_n + i\boldsymbol{h}\cdot\boldsymbol{\sigma})\hat{\tau}_3\hat{g}_s + \hat{\Delta}\hat{g}_s - \frac{D}{4}(\check{\nabla}\hat{g}_s)^2\Big\}. \tag{20}$$

The saddle point of this expression yields the Usadel equation [3] for $\hat{g}_s$ and the self-consistency equation for $\hat{\Delta}$, and, therefore, (20) is naturally considered as the free-energy candidate. A similar expression can also be derived from diffusive nonlinear σ models [13,30]. In order to discuss this result in the Luttinger-Ward framework, where $\hat{\Delta}$ is handled in a slightly different way, we need to first substitute in the saddle-point value $|\Delta|^2/V = \frac{1}{4}\,\mathrm{Tr}\ \hat{\Delta}\hat{g}$,

$$\frac{F_s}{N_0} = -\frac{\pi T}{2}\sum_\omega \mathrm{tr}\Big\{(\omega_n + i\boldsymbol{h}\cdot\boldsymbol{\sigma})\hat{\tau}_3\hat{g}_s + \frac{1}{2}\hat{\Delta}\hat{g}_s - \frac{D}{4}(\check{\nabla}\hat{g}_s)^2\Big\}. \tag{21}$$

Here, we allow arbitrary coordinate dependence of exchange field $\boldsymbol{h}(\boldsymbol{r})$, the presence of SOC and the vector potential in the covariant gradient operator $\check{\nabla}$. This expression can be directly derived from Eq. (11) by including the impurity scattering: The terms without gradients in (21) are obtained immediately from the Λ and Φ terms in (11) by replacing the exact GF with $\hat{g}_s$. Below, we explain how to obtain the gradient terms as well.

Within the Born approximation, the impurity scattering can be described by the self-energy and the corresponding contribution to the generating functional given by

$$\hat{\Sigma}_{\rm imp} = \hat{g}_s/2\tau_{\rm imp}, \quad \Phi_{\rm imp} = {\rm Tr}\left(\hat{1} - \hat{g}_s^2\right)/8\tau_{\rm imp}. \tag{22}$$

To obtain the free-energy functional in the limit $\tau_{\rm imp} \to 0$, we expand the solution of Eq. (4) in spherical harmonics,

$$\hat{g} \approx \hat{g}_s + \boldsymbol{n}_p \cdot \hat{\boldsymbol{g}}_a, \quad \hat{\boldsymbol{g}}_a = -l\hat{g}_s\check{\nabla}\hat{g}_s, \tag{23}$$

where $l = v_F\tau_{\rm imp}$. The second equation above for the anisotropic contribution $\hat{\boldsymbol{g}}_a$ follows from the Eilenberger equation (4).

We first evaluate $\Phi_{\rm imp}$,

$$\Phi_{\rm imp} = \frac{{\rm Tr}\,\hat{\boldsymbol{g}}_a^2}{24\tau_{\rm imp}} = -\frac{D}{8}\,{\rm Tr}(\check{\nabla}\hat{g}_s)^2, \tag{24}$$

where $D = v_F l/3$ is the diffusion constant. Here, we noted the normalization condition $\hat{g}^2 = 1$ averaged over directions implies $\hat{g}_s^2 \simeq \hat{1} - \hat{\boldsymbol{g}}_a^2/3$ where due to the $1/\tau_{\rm imp}$ factor in (24) it is now important to retain the second-order term in l. The last equality follows from $l(\hat{g}_s\check{\nabla}\hat{g}_s)(\hat{g}_s\check{\nabla}\hat{g}_s) \simeq -l(\check{\nabla}\hat{g}_s)^2$, which holds in leading order due to the normalization condition.

A similar contribution appears from the gradient term functional E (12). We can first observe from Eq. (12) that, for matrices $\hat{g}_s$ without angular dependence, $E[\hat{g}_s] = 0$ because of the angular average in Tr.

In the leading order in l, the anisotropic correction (23) can be considered as a variation of the GF. Then, we can calculate the value of the functional $E[\hat{g}]$ by using its defining property (8),

$$\int d^3r\, E[\hat{g}_s + \boldsymbol{n}_p \cdot \hat{\boldsymbol{g}}_a] = \int d^3r\ {\rm Tr}\ \hat{W}(\boldsymbol{v}_F \cdot \check{\nabla}\hat{g}_s) + O(l^2), \tag{25}$$

where the matrix $\hat{W}$ is such that

$$[\hat{W}, \hat{g}_s] = -l\hat{g}_s(\boldsymbol{n}_p \cdot \check{\nabla})\hat{g}_s. \tag{26}$$

This implies

$$\hat{W}(\boldsymbol{v}_F \cdot \check{\nabla})\check{g}_s = v_F l(\boldsymbol{n}_p \cdot \check{\nabla}\check{g}_s)^2 + \check{g}_s\hat{W}\check{g}_s\check{\nabla}\check{g}_,s \tag{27}$$

so that, taking into account that $\check{g}_s\check{\nabla}\check{g}_s = -\check{\nabla}\check{g}_s\check{g}_s + O(l^2)$, we obtain

$${\rm Tr}\ \hat{W}(\boldsymbol{v}_F \cdot \check{\nabla})\check{g}_s \simeq \frac{v_F l}{2}\,{\rm Tr}(\boldsymbol{n}_p \cdot \check{\nabla}\check{g}_s)^2. \tag{28}$$

Then, Eq. (8) yields the gradient term $E \simeq \frac{D}{2}\,{\rm Tr}(\check{\nabla}\hat{g}_s)^2$ so that $\frac{1}{2}E + \Phi_{\rm imp} = \frac{D}{8}\,{\rm Tr}(\check{\nabla}\hat{g}_s)^2$. This leads to the free-energy functional in the diffusive limit (21).

IV. APPLICATION: GRADIENT ENERGY IN THE HYDRODYNAMIC LIMIT

As a further example, we derive the second-order gradient terms in the energy functional, valid both for the spin-singlet superconductor and for the spin-triplet superfluid. This regime is often referred to as the hydrodynamic or London approximation.

For this purpose, we can use a similar approach as in the dirty regime Sec. III. Gradient expansion of the Eilenberger equation gives

$$\hat{g} \approx \hat{g}_s + \boldsymbol{n}_p \cdot \hat{\boldsymbol{g}}_a + \hat{g}_{2s}, \quad \hat{\boldsymbol{g}}_a = -\frac{v_F}{2S}\hat{g}_s\check{\nabla}\hat{g}_s, \tag{29}$$

where $\hat{g}_s = (\omega\hat{\tau}_3 + \hat{\Delta})/S$ is the GF in the locally homogeneous approximation with $\hat{\Delta} = \hat{\Delta}(\boldsymbol{n}_p, \boldsymbol{r})$ and $\hat{g}_{2s}$ is of the second order in gradients. The free-energy Ω is given by Eq. (11) with $\Phi = -\frac{1}{4}\,{\rm Tr}\,[\hat{\Delta}[\hat{g}]\hat{g}]$.

We will consider two characteristic cases. First, the spin-singlet states described by the order parameter $\hat{\Delta}_s = \Delta\begin{pmatrix} 0 & e^{i\varphi} \\ e^{-i\varphi} & 0\end{pmatrix}$ and $S = \sqrt{\omega^2 + \Delta^2}$, where Δ is the real-valued amplitude which can be anisotropic in momentum space $\Delta = \Delta(\boldsymbol{n}_p, \boldsymbol{r})$. Second, the unitary spin-triplet states described by the spin-dependent order parameter $\hat{\Delta}_t = (\boldsymbol{\sigma} \cdot \boldsymbol{d})\begin{pmatrix} 0 & e^{i\varphi} \\ e^{-i\varphi} & 0\end{pmatrix}$ where $S = \sqrt{\omega^2 + \boldsymbol{d}^2}$ and vector $\boldsymbol{d}$ is real.

The general expression for the variation of the gradient term (8) together with (29) yields in leading order in gradients, $E \simeq \frac{v_F^2}{4}\,{\rm Tr}[(\partial\hat{g}_s)^2/S]$ where we denote $\partial = \boldsymbol{n}_p \cdot \check{\nabla}$. The calculation is the same as in the previous section, substituting $l \mapsto v_F/(2S)$. The $\hat{g}_{2s}$ part only contributes to the energy in the other terms in Eq. (11): $\delta\Omega \simeq -\frac{1}{2}\,{\rm Tr}\{(\omega\hat{\tau}_3 + \hat{\Delta}[\hat{g}_s])(\boldsymbol{n}_p \cdot \boldsymbol{g}_a + \hat{g}_{2s}) + \frac{1}{2}\hat{\Delta}[\boldsymbol{n}_p \cdot \boldsymbol{g}_a]\boldsymbol{n}_p \cdot \boldsymbol{g}_a\}$. We now assume the weak-coupling limit and evaluate this by noting [5] that ${\rm Tr}[(\omega\hat{\tau}_3 + \hat{\Delta})(\boldsymbol{n}_p \cdot \hat{\boldsymbol{g}}_a + \hat{g}_{2s})] = {\rm Tr}[S\boldsymbol{n}_p \cdot \hat{g}_s\hat{\boldsymbol{g}}_a + S\hat{g}_s\hat{g}_{2s}] = \frac{v_F^2}{8}\,{\rm Tr}[(\partial\hat{g}_s)^2/S]$ where we have used that $\hat{g}^2 = 1$, $\{\hat{g}_s, \hat{\boldsymbol{g}}_a\} = 0$, and $\{\hat{g}_s, \hat{g}_{2s}\} \simeq -(\boldsymbol{n}_p \cdot \hat{\boldsymbol{g}}_a)^2$.

Then, the free energy in the hydrodynamic regime is given by

$$\Omega = \frac{1}{2}\,{\rm Tr}\left[\frac{v_F^2}{8S}(\partial\hat{g}_s)^2 + |\omega| - \omega\tau_3\hat{g}_s - \frac{1}{2}\hat{\Delta}[\hat{g}_s]\hat{g}_s\right]. \tag{30}$$

Here, $\hat{g}_s = (\omega\hat{\tau}_3 + \hat{\Delta})/S$ is the 4×4 Nambu-spin matrix.

For the above two forms of the order parameter, the first term in Eq. (30) yields the gradient terms,

$$\Omega_s = \frac{N_0 v_F^2}{4}\int\frac{d\Omega_p}{4\pi}[y_{5/2}(\partial\Delta)^2 + y_{3/2}\Delta^2(\partial\varphi)^2], \tag{31}$$

$$\Omega_t = \frac{N_0 v_F^2}{4}\int\frac{d\Omega_p}{4\pi}\left[y_{5/2}(\partial|\boldsymbol{d}|)^2 + y_{3/2}\boldsymbol{d}^2(\partial\varphi)^2 + y_{3/2}\frac{(\boldsymbol{d}\times\partial\boldsymbol{d})^2}{\boldsymbol{d}^2}\right], \tag{32}$$

where we denote $y_{5/2} = \pi T\sum_\omega \omega^2/S^5$ and $y_{3/2} = \pi T\sum_\omega 1/S^3$. In the isotropic case, where $\int\frac{d\Omega_p}{4\pi}(\partial\varphi)^2 = \frac{1}{3}(\nabla\varphi)^2$ and similarly for Δ, Eq. (31) reduces to that in Ref. [31]; (32) agrees with Ref. [5] neglecting Fermi-liquid corrections and for weak coupling.

V. APPLICATION: S/FI BILAYER

The free energy can be used to locate first-order phase transitions. Below, we consider a prototypical example in the superconducting phase, the transition to the normal state

[32–37] induced by an interaction with magnets. To be specific, we consider the problem introduced in Ref. [19], a S/FI bilayer with specular scattering at the interface. In the clean limit, this system does not exhibit a first-order transition, but it is restored by processes that mix scattering trajectories [38]. In Ref. [19], the clean limit and Fermi-liquid interactions were considered; below, we, instead, study how the transition is restored by impurity scattering. To be more realistic, Fermi-liquid effects and interface roughness could also be included, but, for simplicity, we do not consider them below. Moreover, we will consider only uniform phases ignoring possibility of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) phase (which could be included, see Ref. [39]), which likely is reasonable in the presence of disorder [40].

We assume the FI lies in the half-space $z < 0$, superconductor in the layer $0 < z < d$, and vacuum in $z > d$. The quasiclassical description of the problem is as discussed in the previous sections and Ref. [19] with additional boundary conditions (b.c.s) at the FI and vacuum interfaces,

$$\hat{g}(\hat{p}, z = 0) = \hat{S}_{\hat{p}}\hat{g}(\underline{\hat{p}}, z = 0)\hat{S}_{\hat{p}}^{\dagger}, \quad n_z > 0\,, \tag{33}$$

$$\hat{S}_{\hat{p}} = e^{-i\Theta(n_z)\hat{\mu}\cdot\boldsymbol{\sigma}\hat{\tau}_3/2}, \quad \underline{\hat{p}} = (n_x, n_y, -n_z), \tag{34}$$

where $\Theta(\hat{p})$ is the spin-mixing angle and $\hat{\mu}$ is the unit vector in the direction of the magnetization of the FI layer. Here, we write $\hat{p} = (n_x, n_y, n_z)$. The vacuum interface boundary condition at $z = d$ is similar with $\Theta = 0$ and $n_z < 0$. We choose $\hat{\tau}_t = \hat{\tau}_3$ in which case the boundary conditions do not result to boundary contributions in the free energy (see Appendix A).

A. Thin-film solution

We first need the thin-film limit $d \to 0$ solution for $\hat{g}$, which can be found from Ref. [19], and is as follows. First, expand in polynomials in z:

$$\hat{g}(\hat{p}, z) = \hat{G}_0(\hat{p}) + \frac{d - z}{d}\hat{G}_1(\hat{p}) + \cdots. \tag{35}$$

Substituting this to the Eilenberger equation gives

$$\hat{G}_1(\hat{p}) = \frac{d}{v_F n_z}[M_{\hat{p}}, \hat{G}_0(\hat{p})]. \tag{36}$$

The b.c.s imply $\hat{G}_0(\underline{\hat{p}}) = \hat{G}_0(\hat{p})$, and ($n_z > 0$),

$$\hat{G}_0(\hat{p}) + \hat{G}_1(\hat{p}) = \hat{S}_{\hat{p}}[\hat{G}_0(\hat{p}) + \hat{G}_1(\underline{\hat{p}})]\hat{S}_{\hat{p}}^{\dagger}. \tag{37}$$

Suppose, now, $M_{\hat{p}} = M_{\underline{\hat{p}}}$. Then,

$$\frac{v_F n_z}{d}[\hat{S}_{\hat{p}} - \hat{S}_{\hat{p}}^{\dagger}, \hat{G}_0(\hat{p})] = \{\hat{S}_{\hat{p}} + \hat{S}_{\hat{p}}^{\dagger}, [M_{\hat{p}}, \hat{G}_0(\hat{p})]\}. \tag{38}$$

Because $\hat{S}_{\hat{p}} + \hat{S}_{\hat{p}}^{\dagger} = 2\cos(\Theta/2)$ [19],

$$[\tilde{M}_{\hat{p}}, \hat{G}_0(\hat{p})] = 0, \quad \hat{G}_0(\hat{p})^2 = 1. \tag{39}$$

$$\tilde{M}_{\hat{p}} = M_{\hat{p}} + i\boldsymbol{h}_{\hat{p}}\cdot\boldsymbol{\sigma}\hat{\tau}_3, \quad \boldsymbol{h}_{\hat{p}} = \hat{\mu}\frac{v_F|n_z|}{2d}\tan\frac{\Theta}{2}, \tag{40}$$

which is formally similar to the bulk Eilenberger equation with an effective exchange field. For simplicity, we below consider the case of $\boldsymbol{h}_{\hat{p}} = h_0|n_z|\hat{\mu}$ where the spin-mixing angle Θ is constant.

In the clean limit without molecular fields, we have $\hat{\Sigma} = \Delta\hat{\tau}_1$, and

$$\hat{G}_0(\hat{p}) = \frac{(\omega + i\boldsymbol{h}_{\hat{p}}\cdot\boldsymbol{\sigma})\hat{\tau}_3 + \Delta\hat{\tau}_1}{\sqrt{(\omega + i\boldsymbol{h}_{\hat{p}}\cdot\boldsymbol{\sigma})^2 + \Delta^2}}, \tag{41}$$

with an angle-dependent spin-splitting field.

For the self-consistency relation, we consider here the weak-coupling s-wave case of $\hat{\Delta}[\hat{g}] = B\hat{\tau}_1\Delta_1[\hat{g}]$, $\Delta_1[\hat{g}] = \frac{\lambda}{4N_0}\,\mathrm{Tr}[B\hat{\tau}_1\hat{g}]$ where $B(\omega)$ provides the BCS frequency cutoff at $\omega > \omega_c$ and λ is the coupling constant. Eliminating the cutoff as usual by adding and subtracting,

$$\Delta_1 = \Delta + \lambda\Delta\ln\frac{T_{c0}}{T} + \frac{\lambda}{4N_0}\mathrm{Tr}\left[\hat{\tau}_1\hat{g}_1[\Delta] - \frac{\Delta}{|\omega_n|}\right], \tag{42}$$

where the terms are cutoff independent.

In the clean limit at $T = 0$, the self-consistency equation becomes

$$\begin{aligned} 0 &= \frac{\Delta_1 - \Delta}{\lambda\Delta} \\ &= \ln\frac{\Delta_0}{\Delta} + \mathrm{Re}\,\mathrm{arsinh}\left(\frac{ih_0 - 0^+}{\Delta}\right) \\ &\quad + \mathrm{Re}\sqrt{1 - (\Delta/h_0)^2}, \end{aligned} \tag{43}$$

where Δ_0 is the order parameter at $h_0 = 0$, $T = 0$. For $h_0 < \Delta$, the solution is $\Delta = \Delta_0$; for $\Delta_0 < h_0 < h_{c,2} = \frac{e}{2}\Delta_0$ there is a solution $\Delta > 0$; and for $h_0 > \frac{e}{2}\Delta_0$ only the normal state exists [19].

B. Impurity effect

Including an impurity scattering self-energy with Ansatz $\hat{\Sigma}_{\mathrm{imp}} = \sum_{\pm}\frac{1}{2}(1 \pm \hat{\mu}\cdot\boldsymbol{\sigma})(\Sigma_{\pm,3}\hat{\tau}_3 + \Sigma_{\pm,1}\hat{\tau}_1)$ in the Eilenberger equation, the solution of Eq. (39) reads

$$\hat{G}_0(\hat{p}) = \sum_{\pm}\frac{1 \pm \hat{\mu}\cdot\boldsymbol{\sigma}}{2}\frac{(\omega_{\pm} \pm ih_0|n_z|)\hat{\tau}_3 + \Delta_{\pm}\hat{\tau}_1}{\sqrt{(\omega_{\pm} \pm ih_0|n_z|)^2 + \Delta_{\pm}^2}}, \tag{44}$$

where $\omega_{\pm} = \omega_n + \Sigma_{\pm,3}$, $\Delta_{\pm} = \Delta + \Sigma_{\pm,1}$. The self-consistency equations (22) for $\hat{\Sigma}_{\mathrm{imp}}$ or, equivalently, for $\omega_{\pm}$ and $\Delta_{\pm}$, read

$$\omega_{\pm} = \omega_n \pm \frac{\sqrt{(\omega_{\pm} \pm ih_0)^2 + \Delta_{\pm}^2} - \sqrt{\omega_{\pm}^2 + \Delta_{\pm}^2}}{2i\tau_{\mathrm{imp}}h_0}, \tag{45}$$

$$\begin{aligned} \Delta_{\pm} &= \Delta \pm \frac{\Delta_{\pm}}{2i\tau_{\mathrm{imp}}h_0} \\ &\quad \times \ln\frac{\omega_{\pm} \pm ih_0 + \sqrt{(\omega_{\pm} \pm ih_0)^2 + \Delta_{\pm}^2}}{\omega_{\pm} + \sqrt{\omega_{\pm}^2 + \Delta_{\pm}^2}}. \end{aligned} \tag{46}$$

The self-consistency equation for Δ can be written as

$$\begin{aligned} 0 = \frac{\Delta_1 - \Delta}{\lambda} &= \pi T\sum_{\omega_n,\pm}\left((\Delta_{\pm} - \Delta)\tau_{\mathrm{imp}} - \frac{\Delta}{2|\omega_n|}\right) \\ &\quad + \Delta\ln\frac{T_{c0}}{T}. \end{aligned} \tag{47}$$

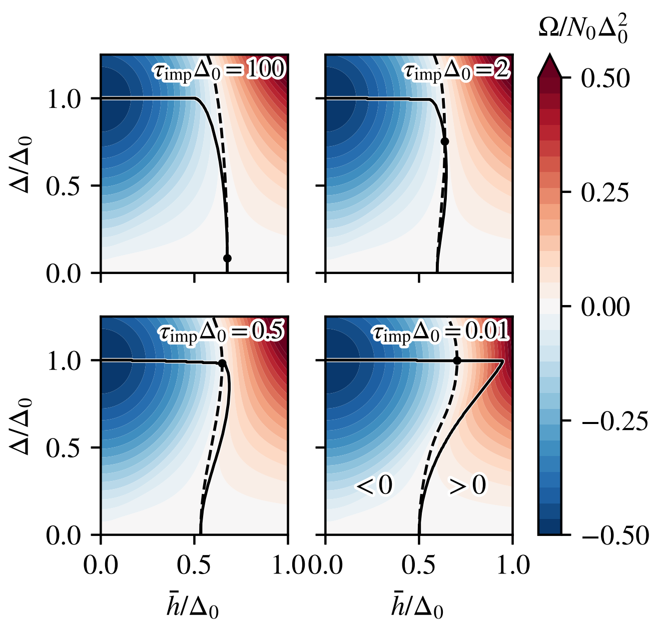

FIG. 1. Self-consistent $\Delta(\bar{h})$ relation at $T=0$ (solid lines), locus of $\Omega(\Delta,\bar{h})=0$ (dashed lines), and value of $\Omega(\Delta,\bar{h})$ (colors), from Eqs. (47), (51), and (60). The crossing points (dots) mark the location of the first-order transition.

For $\tau_{imp}h_0, \tau_{imp}\Delta \ll 1$, the solutions to Eqs. (45) and (46) are

$$\omega_\pm \simeq \frac{\Delta_\pm}{\Delta}\bar{\omega}_\pm, \qquad \Delta_\pm \simeq \Delta + \frac{1}{2\tau_{imp}}\frac{\Delta}{\sqrt{\bar{\omega}_\pm^2+\Delta^2}}, \tag{48}$$

where $\bar{\omega}_\pm \equiv \omega_n \pm i\bar{h}$ and $\bar{h}=\frac{1}{2}h_0$ is the angle-averaged exchange field. In this limit, the resulting $\hat{G}_0$ is the same as an isotropic bulk Green's function with uniform spin-splitting field $\bar{h}$, and at $T=0$, Eq. (47) reduces to Eq. (43) with the last term on the right-hand side omitted. Hence, for $\tau_{imp}\to 0$, $\Delta=\Delta_0$ for $0<\bar{h}<\Delta_0$. Solutions to Eq. (47) for $\tau_{imp}>0$ are shown in Fig. 1. They show the well-known supercooling/heating region where the superconducting and normal states are separated by a local free-energy maximum.

C. Thin-film condensation energy

Evaluating the condensation energy corresponding to the above solutions is now a mechanical exercise in substituting the above expressions into Eq. (19) and evaluating the Matsubara sum and the direction integral. We will below do this analytically to a degree—however, one could as well substitute self-consistent numerical solutions into Eq. (19).

The Φ_Δ functional can be rewritten using Eq. (42),

$$\Phi_\Delta[\hat{g}_1[\Delta]] = -\tfrac{1}{4}\,\mathrm{Tr}\{\hat{\Delta}[\hat{g}_1]\hat{g}_1\}, \tag{49}$$

$$\simeq -\frac{1}{4}\,\mathrm{Tr}\{B\Delta\tau_1\hat{g}_1\} - N_0\frac{\Delta_1-\Delta}{\lambda}\Delta, \tag{50}$$

where the second line is in the leading order in $\lambda\to 0$. Below, we compute the value Ω_* of the free energy at self-consistency ($\Delta_1=\Delta$); the full functional is recovered by adding the second term above,

$$\Omega = \Omega_* - N_0(\Delta_1-\Delta)\Delta/\lambda \tag{51}$$

see Eqs. (42) and (47). Ω_* is also cutoff independent.

Considering first the clean limit, for $d\to 0$, we can substitute Eq. (41) into Eq. (19),

$$\Omega_* = \frac{1}{2}\,\mathrm{Tr}\left\{\Delta\hat{\tau}_1\hat{G}_0 + \omega\hat{\tau}_3\hat{g}_n - \frac{[\hat{\tau}_3,\Delta\hat{\tau}_1]}{[\hat{\tau}_3,\hat{G}_0]} - \frac{1}{2}\Delta\hat{\tau}_1\hat{G}_0\right\} \tag{52}$$

$$= \pi N_0\int\frac{d\Omega_p}{4\pi}T\sum_{\omega_n,\pm}\left(|\omega_n| - \frac{2(\omega_n\pm ih_{\hat{p}})^2+\Delta^2}{2\sqrt{(\omega_n\pm ih_{\hat{p}})^2+\Delta^2}}\right) \tag{53}$$

$$= \pi N_0 T\sum_{\omega_n,\pm}\left(|\omega_n| - \frac{h_0\mp i\omega_n}{2h_0}\sqrt{(\omega_n\pm ih_0)^2+\Delta^2}\right). \tag{54}$$

The result is identical to the condensation energy of a superconductor in exchange field, [34–36] averaged over the direction dependence of the effective field. At $T=0$, this gives the condensation energy,

$$\Omega_* = -\tfrac{1}{2}N_0\Delta^2 + \tfrac{1}{3}N_0h_0^2\,\mathrm{Re}\left[1-\left(1-\Delta^2h_0^{-2}\right)^{3/2}\right]. \tag{55}$$

It is $\Omega_*<0$ for all h_0 if $\Delta>0$. Hence, the transition is always of the second order as found in Ref. [19] based on a Ginzburg-Landau expansion.

Let us then consider the effect of impurity scattering, with $\Phi=\Phi_\Delta+\Phi_{imp}$. From Eq. (22), we have $\Phi_{imp}=\frac{1}{8\tau_{imp}}\mathrm{Tr}[1-4\tau_{imp}^2\hat{\Sigma}_{imp}^2]$, $\mathrm{Tr}\,\hat{G}_0=\mathrm{Tr}[2\tau_{imp}\hat{\Sigma}_{imp}]$, and, hence,

$$\Omega_* = \frac{1}{2}\,\mathrm{Tr}\left\{\tau_{imp}\Delta\hat{\tau}_1\hat{\Sigma}_{imp} + \omega\hat{\tau}_3\hat{g}_n - \frac{[\hat{\tau}_3,\Delta\hat{\tau}_1+\hat{\Sigma}_{imp}]}{[\hat{\tau}_3,\hat{G}_0]} + \frac{1}{4\tau_{imp}} + \tau_{imp}\hat{\Sigma}_{imp}^2\right\}. \tag{56}$$

Direct calculation and using Eqs. (44)–(46) give

$$\int\frac{d\Omega_p}{4\pi}\frac{[\hat{\tau}_3,\Delta\hat{\tau}_1+\hat{\Sigma}_{imp}]}{[\hat{\tau}_3,\hat{G}_0]} = \sum_\pm\frac{1\pm\hat{\mu}\cdot\boldsymbol{\sigma}}{2}\hat{\tau}_0Q_\pm, \tag{57}$$

$$Q_\pm = \tau_{imp}[\Sigma_{\pm,3}^2+\Sigma_{\pm,1}^2] + \tau_{imp}[\Sigma_{\pm,1}\Delta+\Sigma_{\pm,3}\omega_n] + \frac{1}{2}\sqrt{(\omega_\pm+ih_0)^2+\Delta_\pm^2}, \tag{58}$$

and furthermore,

$$\Omega_* = \pi N_0 T\sum_{\omega_n,\pm}\left\{|\omega_n| - \frac{h_0\mp i\omega_n}{2h_0}\sqrt{(\omega_\pm\pm ih_0)^2+\Delta_\pm^2} + \frac{1}{4\tau_{imp}} + \frac{\mp i\omega_n}{2h_0}\sqrt{\omega_\pm^2+\Delta_\pm^2}\right\} \tag{59}$$

$$= \pi N_0 T\sum_{\omega_n,\pm}\left\{|\omega_n| - \frac{\omega_n\pm i\frac{1}{2}h_0}{2}(D_\pm+D_\pm^{-1})\right\}, \tag{60}$$

where $D_\pm = \frac{1}{\pm ih_0}[\sqrt{(\omega_\pm\pm ih_0)^2+\Delta_\pm^2}-\sqrt{\omega_\pm^2+\Delta_\pm^2}]$. For $\tau_{imp}\to 0$, $D_\pm\to(\omega_n\pm i\bar{h})/\sqrt{(\omega_n\pm i\bar{h})^2+\Delta^2}$, and the result becomes the spin-split BCS condensation energy [34–36], $\Omega_* = -\frac{\Delta^2}{2}+\bar{h}^2-\bar{h}\,\mathrm{Re}\sqrt{\bar{h}^2-\Delta^2}$ at $T=0$. For $\tau_{imp}\to\infty$, $\omega_\pm\to\omega_n$, $\Delta_\pm\to\Delta$ and one finds Eq. (54). Hence, as could be expected, strong impurity scattering restores the

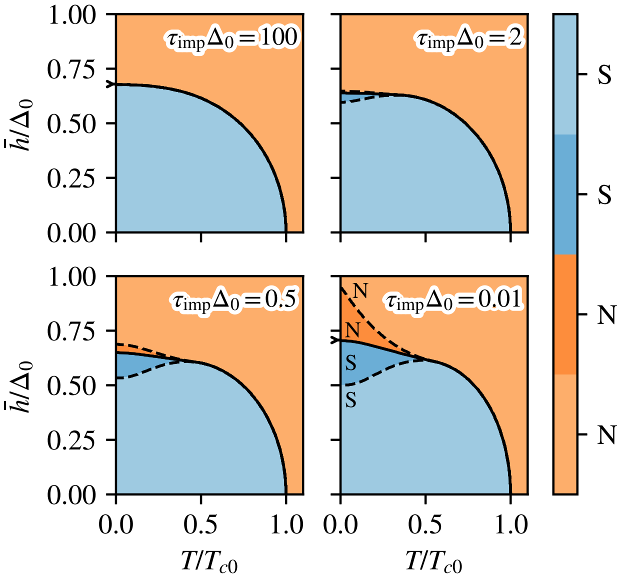

FIG. 2. Phase diagram for different $\tau_{\rm imp}$'s, indicating S and N states. The supercooling/heating region in which the N/S transition is first order is indicated by dashed lines. In the top-left panel, the arrowhead on the y axis indicates the critical field $\bar{h} = \frac{e}{4}\Delta_0$, and in the bottom-right panel, the Chandrasekhar-Clogston field is $\bar{h} = \Delta_0/\sqrt{2}$.

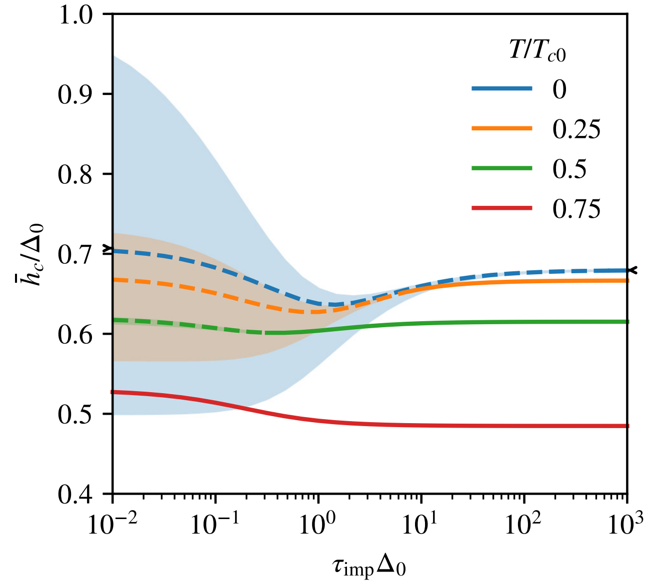

FIG. 3. Critical field $\bar{h}_c$ for $T/T_{c0} = 0, 0.25, 0.5, 0.75$ (top to bottom). The dashed line indicates a first-order transition and shading the supercooling region. The arrowheads on the y axis indicate $\bar{h}/\Delta_0 = e/4$ and $1/\sqrt{2}$.

Chandrasekhar-Clogston first-order transition to the normal state at $\bar{h} = \Delta_0/\sqrt{2}$.

The stability boundary $\Omega = 0$ from Eqs. (47), (51), and (60) is plotted in Fig. 1 for several $\tau_{\rm imp}$'s at $T = 0$ together with the self-consistent $\Delta(\bar{h})$ relations (47). The self-consistent Δ is a local extremum of the computed Ω. As $\tau_{\rm imp} \to 0$, the critical field $\Omega_*[\Delta(\bar{h}_p), \bar{h}_p] = 0$ approaches $\bar{h}_p \to \Delta_0/\sqrt{2}$.

The phase diagram is shown in Fig. 2, indicating how decreasing $\tau_{\rm imp}$ gives rise to a region with first-order transition to the normal state at low temperatures and high effective fields. The clean/dirty crossover in the limit $d \to 0$ shown occurs at $\tau_{\rm imp} \sim \hbar/\Delta$. The corresponding critical field $\bar{h}_c$ at which the N/S transition occurs is shown in Fig. 3 as a function of the impurity scattering time at a few different temperatures. It turns out to be nonmonotonic in $\tau_{\rm imp}$.

VI. SUMMARY AND DISCUSSION

We have rigorously derived the free-energy functional (10), (12), and (19) of a superconducting system in terms of the quasiclassical propagators. We obtained convenient expressions in terms of Riccati amplitudes (16) and in the diffusive limit (21). The functional generalizes the well-known Eilenberger free energy for the systems with arbitrary types of pairing and interacting with spin-dependent fields. The result fills an important gap in the theory of superconductivity between the Eilenberger free energy and the Luttinger-Ward functional. It can be used to analyze thermodynamic properties of many superconducting systems, some of which attract intense interest nowadays. Among them, there are exotic states in unconventional superconductors [41–44] and various hybrid systems [8] including those with spin-triplet superconducting correlations produced either by the exchange field and/or by the SOC [7,45]. Superconductor/ferromagnet systems are studied quite intensively in view of spintronic applications [46,47]. With the help of free-energy expressions found in this paper it is possible to analyze complicated behavior of competing superconducting phases, such as 0-π Josephson junctions [8], cryptoferromagnetism [48–52], FFLO states [53,54] modified by different geometrical factors [27,55], and configurations with different vorticities [56,57] in such systems using rigorous microscopic calculations.

As an application to the superconductor/ferromagnet systems, we studied the superconducting phase transition in a S/FI bilayer. In the clean noninteracting limit, this system has a second-order phase transition as a function of effective exchange field [19], differing from a bulk superconductor in the homogeneous spin-splitting field [32,33]. We show that even very small amount of impurity scattering in thin superconducting films restores the first-order phase transition.

The interplay of SOC and external magnetic field generates proximity-induced topological superconductivity in Majorana nanowires [58]. The ground states of such systems taking into account the important orbital effect and Abrikosov vortex formation [59,60] can be found by calculating the free energy, which can be performed using our expressions with arbitrary impurity scattering rates.

Our results can be directly applied to study the free energy of spin-triplet superconductors and superfluids [61], such as superfluid ^{3}He under various conditions [62]. Even though the spin-triplet superfluity in ^{3}He has been studied for many years, the Eilenberger-type free-energy expression is derived only in the present paper, advancing the theory of spin-triplet paired states. This tool should be particularly useful to study different competing and spatially inhomogeneous phases for the confined topological superfluids [6,44,63,64], exotic disordered phases [65,66], and vortex states, such as double-core vortices [67–74] and recently found half-quantum vortices [75,76].

ACKNOWLEDGMENTS

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APPENDIX A: DERIVATION OF EQ. (12)

We now derive Eq. (12). We assume $\hat{\tau}^2=1$, $\hat{g}^2=1$, $\partial_\lambda \hat{g}=[\hat{W},\hat{g}]$, and $\partial_\lambda\hat{\tau}=0$. Moreover, we denote $\partial\equiv\frac{\boldsymbol{v}_F}{v_F}\cdot\check{\boldsymbol{\nabla}}$ as the derivative operator in the Eilenberger equation.

From the above, it follows, with standard matrix calculus, $\partial(\hat{a}^{-1})=-\hat{a}^{-1}(\partial a)\hat{a}^{-1}$, $\delta(\hat{a}^{-1})=-\hat{a}^{-1}(\delta a)\hat{a}^{-1}$, and, moreover, $\partial\hat{g}\hat{g}=-\hat{g}\,\partial\hat{g}$, and $\delta\hat{g}\hat{g}=-\hat{g}\,\delta\hat{g}$. Denote $\hat{Z}\equiv[\hat{\tau},\hat{g}]^{-1}$. We can observe that $\hat{Z}\hat{g}=-\hat{g}\hat{Z}$ and $\hat{Z}\hat{\tau}=-\hat{\tau}\hat{Z}$.

Equipped with the above, consider then the variation vs $\hat{g}$ of $E_s=\frac{1}{2}\operatorname{tr}\hat{g}[\hat{\tau},\hat{g}]\partial[\hat{\tau},\hat{g}]^{-1}$,

$$\begin{aligned}2\delta E_s &= \delta\ \operatorname{tr}\hat{g}[\hat{\tau},\hat{g}]\partial Z\\ &= \operatorname{tr}\ \delta(\hat{g}[\hat{\tau},\hat{g}])\partial\hat{Z}-\operatorname{tr}\ \partial(\hat{g}[\hat{\tau},\hat{g}])\delta\hat{Z}+\operatorname{tr}\ \partial(\hat{g}[\hat{\tau},\hat{g}]\delta\hat{Z})\\ &= 2\,\delta E_1-2\,\delta E_2+2\,\delta E_3.\end{aligned}\tag{A1}$$

We write $\delta E=\delta E'+\delta E''$ where $\delta E'$ do not contain terms $\propto\partial\hat{\tau}$. We have

$$\begin{aligned}2\,\delta E_1' &= -\operatorname{tr}(\delta\hat{g}[\hat{\tau},\hat{g}]+\hat{g}[\hat{\tau},\delta\hat{g}])\hat{Z}[\hat{\tau},\partial\hat{g}]\hat{Z}\\ &= -\operatorname{tr}\{\delta\hat{g}[\hat{\tau},\partial\hat{g}]\hat{Z}+\partial\hat{g}\hat{Z}(\hat{\tau}\hat{g}[\hat{\tau},\delta\hat{g}]-\hat{g}[\hat{\tau},\delta\hat{g}]\hat{\tau})\hat{Z}\}\\ &= -\operatorname{tr}\ \delta\hat{g}[\hat{\tau},\partial\hat{g}]\hat{Z}-\operatorname{tr}\ \partial\hat{g}[\hat{\tau},\delta\hat{g}]\hat{Z}\\ &\quad -2\operatorname{tr}(\partial\hat{g})\hat{Z}\hat{g}(\delta\hat{g}-\hat{\tau}\,\delta\hat{g}\,\hat{\tau})\hat{Z}.\end{aligned}\tag{A2}$$

The term $\delta E_2'$ is obtained by exchanging ∂ and δ in the above expression. We then find

$$\begin{aligned}\delta E_1'-\delta E_2' &= \operatorname{tr}[\delta\hat{g}\hat{Z}\hat{g}(\partial\hat{g}-\hat{\tau}\,\partial\hat{g}\,\hat{\tau})-\partial\hat{g}\hat{Z}\hat{g}(\delta\hat{g}-\hat{\tau}\,\delta\hat{g}\,\hat{\tau})]\hat{Z}\\ &= \operatorname{tr}\ \delta\hat{g}\hat{Z}(\hat{\tau}\hat{g}\,\partial\hat{g}\,\hat{\tau}-\hat{g}\hat{\tau}\,\partial\hat{g}\,\hat{\tau})\hat{Z}\\ &= \operatorname{tr}\ \delta\hat{g}\,\partial\hat{g}\,\hat{\tau}\hat{Z}=\operatorname{tr}[\delta W,\hat{g}]\partial\hat{g}\,\hat{\tau}\hat{Z}\\ &= \operatorname{tr}(\partial\hat{g})\delta W.\end{aligned}\tag{A3}$$

Moreover,

$$\begin{aligned}&\delta E_1''-\delta E_2''\\ &= \tfrac{1}{2}\operatorname{tr}\ \hat{g}[\partial\hat{\tau},\hat{g}]\hat{Z}[\hat{\tau},\delta\hat{g}]\hat{Z}-\tfrac{1}{2}\operatorname{tr}\ \delta(\hat{g}[\hat{\tau},\hat{g}])\hat{Z}[\partial\hat{\tau},\hat{g}]\hat{Z}\\ &= \tfrac{1}{2}\operatorname{tr}\ \partial\hat{\tau}\,\hat{Z}\{[\hat{g},\delta(\hat{g}[\hat{\tau},\hat{g}])]+\hat{g}[\hat{\tau},\delta\hat{g}]\hat{g}-[\hat{\tau},\delta\hat{g}]\}\hat{Z}\\ &= 0.\end{aligned}\tag{A4}$$

We then find

$$\delta E_s=\operatorname{tr}(\partial\hat{g})\delta W+\tfrac{1}{2}\operatorname{tr}\ \partial(\hat{g}[\hat{\tau},\hat{g}]\delta\hat{Z}).\tag{A5}$$

The functional (12) then, indeed, has the claimed variation in the interior. Note that the above calculation did not assume a specific form for the matrix $\hat{\tau}$.

We can also evaluate the variation vs $\hat{\tau}$,

$$\begin{aligned}\delta_\tau E_s &= -\tfrac{1}{2}\operatorname{tr}\ \delta\hat{Z}\,\hat{g}\,\partial[\hat{\tau},\hat{g}]+\tfrac{1}{2}\operatorname{tr}[\delta\tau,\hat{g}]\partial(\hat{Z}\hat{g})\\ &\quad -\tfrac{1}{2}\operatorname{tr}\ \partial([\delta\tau,\hat{g}]\hat{Z}\hat{g})\\ &= \tfrac{1}{2}\operatorname{tr}[\delta\tau,\hat{g}]\hat{Z}(\hat{g}\,\partial[\tau,\hat{g}]+(\partial[\tau,\hat{g}])\hat{g}+(\partial g)[\tau,\hat{g}])\hat{Z}\\ &\quad +\operatorname{tr}\partial(\delta\tau\,\hat{Z})=\operatorname{tr}\ \partial(\delta\tau\,\hat{Z}),\end{aligned}\tag{A6}$$

which is a full derivative.

Integrating Eq. (8) now reduces to an application of the Stokes theorem. In particular, Eq. (A5) implies

$$\operatorname{tr}\ \partial\hat{g}_\lambda W_\lambda=\partial_\lambda E_s-\partial_s E_\lambda,\tag{A7}$$

where $E_\lambda=\frac{1}{2}\operatorname{tr}(\hat{g}_\lambda[\hat{\tau},\hat{g}_\lambda]\partial_\lambda[\tau,\hat{g}_\lambda]^{-1})$ and we write $\partial_s\operatorname{tr}\hat{X}\equiv\boldsymbol{n}\cdot\boldsymbol{\nabla}\ \operatorname{tr}\hat{X}=\boldsymbol{n}\cdot\operatorname{tr}\check{\boldsymbol{\nabla}}\hat{X}=\operatorname{tr}\ \partial\hat{X}$. Hence,

$$\begin{aligned}&\int_0^1 d\lambda\int d^3r\ \operatorname{Tr}[v_F\,\partial\hat{g}_\lambda W_\lambda]\\ &= \left\langle\int d^2\rho\, v_F\int_{\partial M}d\boldsymbol{l}\cdot\boldsymbol{E}\right\rangle_{\hat{p},\omega}\\ &= \int d^3r\,(E[\hat{g}_1]-E[\hat{g}_0])\\ &\quad+\left\langle\int d^2\rho\int_0^1 d\lambda\, v_F(E_\lambda|_{s=\infty}-E_\lambda|_{s=-\infty})\right\rangle_{\hat{p},\omega},\end{aligned}\tag{A8}$$

where $\langle X\rangle_{\hat{p},\omega}=\pi TN_0\sum_{\omega_n}\int\frac{d\Omega_p}{4\pi}X$ so that $\operatorname{Tr}X=\langle\operatorname{tr}X\rangle_{\hat{p},\omega}$. The line integral is over the boundary of $M=[0,1]\times[-\infty,\infty]$ with $d\boldsymbol{l}=(d\lambda,ds)$ and $\boldsymbol{E}=(E_\lambda,E_s)$. The spatial integral is decomposed to an integral over the coordinate s along $\boldsymbol{n}$ and the perpendicular coordinate $\boldsymbol{\rho}$.

The last boundary term vanishes under the average over momentum directions if $v_F(-\hat{p})=v_F(\hat{p})$. It also vanishes if the boundary conditions for $\hat{g}_\lambda$ are equal $\hat{g}_\lambda(s=\infty)=\hat{g}_\lambda(s=-\infty)$ or if they are independent of λ (e.g., the normal state at infinity). This also indicates the boundary term can be neglected when studying local effects in infinite systems.

We can also consider a finite-size system residing in $z>0$ with the scattering boundary condition of Eq. (33) at $z=0$,

$$\hat{g}(\hat{p},z=0)=\hat{S}_{\hat{p}}\hat{g}(\underline{\hat{p}},z=0)\hat{S}_{\hat{p}}^{-1},\quad n_z>0,\tag{A9}$$

where $\underline{\hat{p}}=(n_x,n_y,-n_z)$. Here, $\hat{S}$ is the boundary scattering matrix, independent of λ. If $[\hat{S},\hat{\tau}]=0$, then,

$$\int_{n_z>0}\frac{d\Omega_p}{4\pi}E_\lambda[\hat{g}_\lambda(z=0;\hat{p})]=\int_{n_z<0}\frac{d\Omega_p}{4\pi}E_\lambda[\hat{g}_\lambda(z=0;\hat{p})].\tag{A10}$$

From this, it follows that, in the directional average of Eq. (A8), the terms for $n_z<0$ (which are at the end of the trajectory) cancel those for $n_z>0$ (at the start of the trajectory). Hence, the hard-wall scattering boundary condition appears only via g_λ but does not generate additional terms in the free energy, provided $[\hat{\tau},\hat{S}]=0$ at the boundary.

We need to observe that the above results assume $[\hat{\tau},\hat{g}]$ is invertible everywhere in M since Eq. (A7) does not apply at the singularities where $\boldsymbol{E}$ is not defined. Such points give additional contributions that have to be subtracted, i.e., ∂M includes also clockwise contours C_* (with infinitesimal interior) circling each singularity lying inside $[0,1]\times[-\infty,\infty]$.

Each gives a contribution,

$$\oint_{C_*} \mathrm{tr}[gZ^{-1}dZ]. \tag{A11}$$

Note that because $\mathrm{tr}\,[\hat{g}\hat{Z}^{-1}[A,\hat{Z}]] = -2\,\mathrm{tr}[\hat{g}A]$, gauge fields do not contribute, and we replaced $\partial \mapsto \partial_s$ and $dZ = \partial_s Z\,ds + \partial_\lambda Z\,d\lambda$. Writing $g = U\tau_3 U^{-1}$, $Z = U(\begin{smallmatrix} 0 & w \\ \bar{w} & 0 \end{smallmatrix})U^{-1}$ (due to $gZ + Zg = 0$), we have

$$\oint_{C_*} \mathrm{tr}[gZ^{-1}dZ] = \oint_{C_*} \left[\frac{1}{2}d\ \mathrm{tr}(\ln\bar{w} - \ln w) - \mathrm{tr}\ \tau_3 U^{-1}dU\right] = i\pi m, \tag{A12}$$

where m is an integer. Namely, the last term is regular (we assume U is nonsingular) and gives no contribution for an infinitesimal loop, whereas the first terms yield a winding number. The number, and whether singularities are even present, depends on the choice of $\hat{\tau}$. As the free energy is real valued, these contributions then can be subtracted by taking the real part.

We find Eq. (12), indeed, gives the bulk contribution to the derivative term. It is also the only contribution relevant under quite general conditions.

APPENDIX B: RICCATI PARAMETRIZATION

In Ricatti parametrization, the gradient functional can be expressed as

$$E(\hat{g}) = \tfrac{1}{2}\mathrm{Tr}[\boldsymbol{v}_F\cdot(\hat{a}\nabla\hat{b} - \nabla\hat{a}\hat{b})(\hat{a}\hat{b})^{-1}(1+\hat{a}\hat{b})(1-\hat{a}\hat{b})^{-1}]. \tag{B1}$$

It is straightforward to check that the variation of this expression by $\hat{a}$ and $\hat{b}$ yields gradient terms in the Ricatti equations.

This expression can be written in the compact form

$$E(\hat{g}) = \mathrm{Tr}\ \boldsymbol{v}_F\cdot[(\nabla\hat{a}^{-1} + \nabla\hat{b})(\hat{a}^{-1} - \hat{b})^{-1} + \tfrac{1}{2}\nabla\ \ln(\hat{a}\hat{b})]. \tag{B2}$$

The last term is a full derivative and can be neglected.

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