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Gradient corrections to the time-dependent Ginzburg-Landau equation for anisotropic perturbations of quasiparticles

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Following Gor'kov and Éliashberg [Zh. Eksp. Teor. Fiz. **54**, 612 (1968)], we derive the time-dependent Ginzburg-Landau equation from the nonequilibrium Green functions. Space-gradient terms appearing due to anisotropic perturbations of quasiparticles are evaluated. It is shown that the dominant contribution due to the normal current can be included with no increase in the complexity of the set of equations.

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I. INTRODUCTION

The Gor'kov[1] recovery of the Ginzburg-Landau (GL) theory[2] from Green functions made this very intuitive phenomenological theory acceptable for a wider community of physicists interested in superconductivity. At the same time, it has marked a systematic way to its various modifications. In particular, there was a clear promise that a nonequilibrium version of the Gor'kov approach can yield a desirable time-dependent extension of the GL theory.

The time-dependent equations for the gap Δ turned out to be nontrivial, however. In 1966, Abrahams and Tsuneto[3] and Kemoklidze and Pitaevskiĭ[4,5] obtained such equations for pure superconductors, but their validity is guaranteed only under very restrictive conditions. The problem follows from Rabi-like oscillations of a quasiparticle population with the frequency Δ (we use units with $\hbar=1$). No perturbation is sufficiently slow on the scale of this frequency because Δ vanishes, e.g., in cores of vortices.

To avoid the Rabi-like oscillations, Gor'kov and Éliashberg[6] derived the time-dependent Ginzburg-Landau (TDGL) equations for the case of alloys with paramagnetic impurities. In the dirty limit, when the mean scattering time τ is so short that $\tau\Delta \ll 1$, these oscillations are overdamped. In this case, the time dependence of the gap can be expressed by the microscopically justified TDGL equation.

The dirty limit $\tau \to 0$ implies the very short mean free path $l=v_F\tau$, where v_F is the Fermi velocity. The distribution of quasiparticles then achieves a form of local equilibrium; i.e., it becomes insensitive to various gradients in the system. As a result, nonlocal contributions expressed in the TDGL theory by space-gradient terms are exclusively due to the nonlocality of Cooper pairs. In various derivations[7–18] and implementations[19–46] of the TDGL theory, one thus invariably meets a single gradient term having the familiar form of the quantum kinetic energy with the covariant space gradient.

In this paper, we deal with theory for pure superconductors in which the distribution of quasiparticles can be perturbed from the local equilibrium. We adopt the time derivative as a term established by experience and focus on space gradients, which are not explored in the literature. One can expect that anisotropic perturbations lead to anisotropic gradient terms of the TDGL equation.

We will show that the anisotropic correction enters the kinetic energy via the drift velocity $\mathbf{v}_n$ as

$$\frac{1}{2m^*}(-i\nabla + m^*\mathbf{v}_n - e^*\mathbf{A})^2\psi + \alpha_{\mathrm{GL}}\psi + \beta_{\mathrm{GL}}|\psi|^2\psi = \Gamma_{\mathrm{GL}}\left(i\frac{\partial}{\partial t} - \mu^*\right)\psi. \quad (1)$$

Since the drift velocity is given by the normal current, $\mathbf{v}_n=\mathbf{j}_n/en_n$, this correction does not require any extension of the set of TDGL equations.

Plan of the paper

This paper is organized as follows. Section II provides a gradient expansion of the gap equation. In Sec. III, we employ the quasiparticle approximation to express selected terms of the gradient expansion as functions of the electron distribution. We also evaluate relations needed to transform the gap equation into the TDGL equation in an inhomogeneous system.

In Sec. IV, we solve the Boltzmann equation within the relaxation time approximation. This solution furnishes us with the electron distribution perturbed by an electric field and a temperature gradient. The magnetic field is included nonperturbatively.

Section V includes the main result of this paper. We first derive the anisotropic gradient correction for the gap and then transfer it into the anisotropic gradient term of the TDGL equation. We show that in a very good approximation, this gradient term can be expressed in terms of the drift velocity, as announced in Eq. (1). Section VI is devoted to the interpretation of the anisotropic gradient term.

Section VII provides the gradient correction of the second order. It is shown that this term does not contribute to the TDGL equation because it cancels with gradient terms resulting from the transformation of the gap into the GL function in a system with inhomogeneous temperature. In Sec. VIII, we present our conclusions.

Appendix A provides well known integrals over the BCS kernel. It is included for the convenience of the reader. Appendix B provides a gradient of the chemical potential induced by the gradient of the temperature. Evaluations of two integrals needed in Sec. V is moved to Appendix C.

II. GRADIENT EXPANSION

The time-dependent equation of the Ginzburg-Landau type for the gap is derived from the integral BCS-type equation

$$\bar{\Delta}(t,\mathbf{r}) + V\int dt' d\mathbf{r}' K(t,\mathbf{r},t',\mathbf{r}')\bar{\Delta}(t',\mathbf{r}') = 0. \tag{2}$$

Following Refs. 3, 6, 7, 9, 13, 15, and 19, we start from the equation for the complex conjugate gap $\bar{\Delta}$. The kernel K covers the propagators and statistics; the interaction strength is introduced by the BCS interaction V.

Our aim is to approximate this integral form by its gradient expansion to the first order in time and to the second order in space. In the first step, we derive the gradient expansion with an unspecified kernel K.

A. Gauge

The gap Δ and kernel K depend on the gauge of the electromagnetic field. Gradients of both functions can thus be large even if physical quantities smoothly change in time and space. To avoid large gradients, we shift the vector potential by a constant,

$$\tilde{\mathbf{A}} = \mathbf{A} - \mathbf{A}_0, \tag{3}$$

where the constant shift is adjusted so that this potential reaches zero at the reference point t, $\mathbf{r}$, i.e.,

$$\mathbf{A}_0 = \mathbf{A}(t,\mathbf{r}). \tag{4}$$

The transformation of the gap to the tilde gauge brings a phase factor,

$$\tilde{\Delta}(t',\mathbf{r}') = e^{ie^*(\mathbf{r}'-\mathbf{r})\mathbf{A}_0}\bar{\Delta}(t',\mathbf{r}'), \tag{5}$$

which compensates eventual fast oscillations of the gap. Here, $e^*=2e$ is the charge of the Cooper pair.

By substituting Eq. (5) into Eq. (2), we obtain the equation for the slowly varying gap,

$$\tilde{\Delta}(t,\mathbf{r}) + V\int dt' d\mathbf{r}' \tilde{K}(t,\mathbf{r},t',\mathbf{r}')\tilde{\Delta}(t',\mathbf{r}') = 0, \tag{6}$$

where

$$\tilde{K}(t,\mathbf{r},t',\mathbf{r}') = e^{ie^*(\mathbf{r}-\mathbf{r}')\mathbf{A}_0}K(t,\mathbf{r},t',\mathbf{r}') \tag{7}$$

is the gauge independent kernel. We expand these slowly varying functions in gradients.

We note that a similar transformation should also be done for the scalar potential and the time variable. We do not shift the scalar potential since we do not want to discuss time dependence in this paper.

B. Wigner mixed representation

The gradient expansion is conveniently performed in the Wigner mixed representation,

$$\tilde{K}(t,\mathbf{r},t',\mathbf{r}') = \int \frac{d\omega}{(2\pi)}\frac{d\mathbf{k}}{(2\pi)^3} e^{-i\omega(t-t')+i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \times \tilde{K}(\omega,\mathbf{k},\bar{t},\bar{\mathbf{r}})\big|_{\bar{t}=(1/2)(t+t'),\bar{\mathbf{r}}=(1/2)(\mathbf{r}+\mathbf{r}')}, \tag{8}$$

which represents the double-time function by the Fourier expansion in the difference time $t-t'$, keeping the center-of-mass time $\bar{t}=\frac{1}{2}(t+t')$ as the complementary variable. Similarly, the difference coordinate $\mathbf{r}-\mathbf{r}'$ enters via Fourier components while the center-of-mass coordinate $\bar{\mathbf{r}}=\frac{1}{2}(\mathbf{r}+\mathbf{r}')$ is kept.

In a homogeneous equilibrium system, the choice of the initial time and coordinate is arbitrary; therefore, the function $\tilde{K}$ does not depend on the center-of-mass variables. Out of equilibrium, we can assume that changes in the center-of-mass variables are slow in time and space. We can thus approximate all functions by their values at the reference point t, $\mathbf{r}$ and gradient corrections

$$\begin{aligned}\tilde{K}(\omega,\mathbf{k},\bar{t},\bar{\mathbf{r}}) &= \tilde{K}(\omega,\mathbf{k},t,\mathbf{r}) + \frac{1}{2}(t'-t)\partial_t\tilde{K}(\omega,\mathbf{k},t,\mathbf{r}) \\ &\quad + \frac{1}{2}[(\mathbf{r}'-\mathbf{r})\cdot\nabla]\tilde{K}(\omega,\mathbf{k},t,\mathbf{r}) \\ &\quad + \frac{1}{8}\sum_{\mu,\nu}(r'_\nu-r_\nu)(r'_\mu-r_\mu)\nabla_\nu\nabla_\mu\tilde{K}(\omega,\mathbf{k},t,\mathbf{r}).\end{aligned} \tag{9}$$

We use $\partial_t=\frac{\partial}{\partial t}$, $\nabla=\frac{\partial}{\partial \mathbf{r}}$, and $\nabla_\nu=\frac{\partial}{\partial r_\nu}$. Assuming that super currents are small, we can similarly expand the gap

$$\begin{aligned}\tilde{\Delta}(t',\mathbf{r}') &= \tilde{\Delta}(t,\mathbf{r}) + (t'-t)\partial_t\tilde{\Delta}(t,\mathbf{r}) + [(\mathbf{r}'-\mathbf{r})\cdot\nabla]\tilde{\Delta}(t,\mathbf{r}) \\ &\quad + \frac{1}{2}\sum_{\mu,\nu}(r'_\nu-r_\nu)(r'_\mu-r_\mu)\nabla_\nu\nabla_\mu\tilde{\Delta}(t,\mathbf{r}).\end{aligned} \tag{10}$$

In the expanded form, all functions have the same argument t, $\mathbf{r}$. In many formulas below, we keep this argument implicit.

C. Second-order expansion

Within expansions (9) and (10), the product $\tilde{K}\tilde{\Delta}$ in the integrand of the gap equation [Eq. (2)] has 16 terms. Neglecting cross terms proportional to the time derivative times the space gradient and all space gradients of higher than second order, one is left with eight terms. In this section, we discuss them one by one.

The zero order term reads

$$\tilde{L}\tilde{\Delta} = \int dt'\, d\mathbf{r}' \frac{d\omega}{(2\pi)} \frac{d\mathbf{k}}{(2\pi)^3} e^{-i\omega(t-t')+i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \tilde{K}(\omega,\mathbf{k},t,\mathbf{r})\tilde{\Delta}(t,\mathbf{r})$$
$$= \tilde{K}_{0,\mathbf{0}}\tilde{\Delta}, \qquad (11)$$

where $\tilde{K}_{0,\mathbf{0}} \equiv \tilde{K}\ (\omega=0, \mathbf{k}=\mathbf{0}, t, \mathbf{r})$. This term covers all nongradient terms of the GL theory.

The time derivative in expansion (10) results in the term

$$\tilde{L}_{\omega}\partial_t\tilde{\Delta} = \int dt'\, d\mathbf{r}' \frac{d\omega}{(2\pi)} \frac{d\mathbf{k}}{(2\pi)^3} e^{-i\omega(t-t')+i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}$$
$$\times \tilde{K}(\omega,\mathbf{k},t,\mathbf{r})(t'-t)\partial_t\tilde{\Delta}(t,\mathbf{r})$$
$$= i\frac{\partial \tilde{K}_{0,\mathbf{0}}}{\partial\omega}\partial_t\tilde{\Delta}. \qquad (12)$$

This term covers a part of the time derivative in the TDGL theory. It is supplemented by a cont0ribution from the time derivative in expansion (9),

$$\tilde{L}_{\omega t}\tilde{\Delta} = \int dt'\, d\mathbf{r}' \frac{d\omega}{(2\pi)} \frac{d\mathbf{k}}{(2\pi)^3} e^{-i\omega(t-t')+i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')} \tilde{\Delta}(t,\mathbf{r})$$
$$\times \frac{1}{2}(t'-t)\partial_t\tilde{K}(\omega,\mathbf{k},t,\mathbf{r})$$
$$= i\frac{1}{2}\frac{\partial^2 \tilde{K}_{0,\mathbf{0}}}{\partial\omega\partial t}\tilde{\Delta}. \qquad (13)$$

This term is the main cause of problems with the TDGL theory.

We should mention that the time derivative of the gap or the GL function is not yet fully clarified, and one finds many different forms of this term in the literature. The potential μ^* is either given by the scalar potential,[13,14,19,23–34] $\mu^* = e^*\varphi$, or by the electrochemical potential,[3,9,15,35–40] $\mu^* = 2\mu$, or it includes various nonequilibrium corrections.[16–18,41–45] The factor Γ_{GL} depends on the purity of the system and also on adopted approximations.

Complications hidden in Eq. (13) follow from mechanisms well described within studies of nonequilibrium superconductivity.[47] The time dependence is not our main aim; we thus merely sketch one of these mechanisms. The kernel depends on the distribution of quasiparticles. Since the energy of quasiparticles depends on the gap Δ, any time change of the gap drives quasiparticles out of equilibrium. The resulting nonequilibrium distribution supports a deviation of the gap from its local equilibrium value. One can naively expect that such mechanism yields $\tilde{L}_{\omega t} \propto \partial_t\tilde{\Delta}$ so that the whole term is as small as $\tilde{L}_{\omega t}\tilde{\Delta} \propto \tilde{\Delta}\partial_t\tilde{\Delta}$. This is not true because some relaxation processes have the effective mean scattering time of the form $\tau_{\mathrm{ef}} \propto 1/|\Delta|$. Equation (13) is thus linear in Δ and appreciably complicates the time derivative. This term has been discussed many times; therefore, we refer the reader to Refs. 9 and 47–49 and focus on the space gradients.

The space gradients are represented by five terms. The quadratic term of the expansion of $\tilde{\Delta}$ leads to

$$\tilde{L}_{\mathbf{kk}}\nabla\nabla\tilde{\Delta} \equiv \sum_{\mu\nu}\tilde{L}_{\mu\nu}\nabla_{\mu}\nabla_{\nu}\tilde{\Delta}$$
$$= \sum_{\mu\nu}\int dt'\, d\mathbf{r}' \frac{d\omega}{(2\pi)} \frac{d\mathbf{k}}{(2\pi)^3} e^{-i\omega(t-t')+i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}$$
$$\times \frac{1}{2}(r'_{\mu}-r_{\mu})(r'_{\nu}-r_{\nu})\tilde{K}(\omega,\mathbf{k},t,\mathbf{r})\nabla_{\mu}\nabla_{\nu}\tilde{\Delta}(t,\mathbf{r})$$
$$= -\frac{1}{2}\frac{\partial^2\tilde{K}_{0,\mathbf{0}}}{\partial k_{\mu}\partial k_{\nu}}\nabla_{\mu}\nabla_{\nu}\tilde{\Delta}. \qquad (14)$$

This is proportional to the second-order gradient of the gap; the kernel can thus be treated in the approximation of the local equilibrium. This contribution results in the nonlocal gradient term of the GL theory.

The linear term of the expansion of $\tilde{\Delta}$ multiplied with the zeroth order of $\tilde{K}$ yields

$$\tilde{L}_{\mathbf{k}}\nabla\tilde{\Delta} = \int dt'\, d\mathbf{r}' \frac{d\omega}{(2\pi)} \frac{d\mathbf{k}}{(2\pi)^3} e^{-i\omega(t-t')+i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}(\mathbf{r}'-\mathbf{r})$$
$$\times \tilde{K}(\omega,\mathbf{k},t,\mathbf{r})\nabla\tilde{\Delta}(t,\mathbf{r})$$
$$= -i\frac{\partial\tilde{K}_{0,\mathbf{0}}}{\partial\mathbf{k}}\nabla\tilde{\Delta}. \qquad (15)$$

This term provides a coupling between an anisotropic distribution of quasiparticles and gradients of the gap. Indeed, for an isotropic quasiparticle distribution, this is zero because the momentum derivative vanishes from symmetry reasons. If the distribution is anisotropic, e.g., due to a quasiparticle current, the momentum derivative is nontrivial. This term will be the major focus of our interest.

The linear term in gradients of $\tilde{\Delta}$ multiplied with the linear term of $\tilde{K}$ gives

$$\tilde{L}_{\mathbf{kkr}}\nabla\tilde{\Delta} = \sum_{\mu\nu}\int dt'\, d\mathbf{r}' \frac{d\omega}{(2\pi)} \frac{d\mathbf{k}}{(2\pi)^3} e^{-i\omega(t-t')+i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}$$
$$\times \frac{1}{2}(r'_{\mu}-r_{\mu})(r'_{\nu}-r_{\nu})(\nabla_{\mu}\tilde{K}(\omega,\mathbf{k},t,\mathbf{r}))\nabla_{\nu}\tilde{\Delta}(t,\mathbf{r})$$
$$= -\frac{1}{2}\sum_{\mu\nu}\frac{\partial^3\tilde{K}_{0,\mathbf{0}}}{\partial r_{\mu}\partial k_{\mu}\partial k_{\nu}}\nabla_{\nu}\tilde{\Delta}. \qquad (16)$$

Like contribution (15), this term is a scalar product of a vector material coefficient with the gradient of the gap. As we will see below, it has a very different physical content, however.

The last two terms are products of gradients of $\tilde{K}$ with the nongradient part of $\tilde{\Delta}$,

$$\tilde{L}_{\mathbf{kr}}\tilde{\Delta} = \int dt' d\mathbf{r}' \frac{d\omega}{(2\pi)} \frac{d\mathbf{k}}{(2\pi)^3} e^{-i\omega(t-t')+i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\tilde{\Delta}(t,\mathbf{r}) \times \frac{1}{2}(\mathbf{r}'-\mathbf{r})\,\nabla\,\tilde{K}(\omega,\mathbf{k},t,\mathbf{r}) = -i\frac{1}{2}\frac{\partial^2\tilde{K}_{0,\mathbf{0}}}{\partial\mathbf{r}\partial\mathbf{k}}\tilde{\Delta}, \tag{17}$$

and

$$\tilde{L}_{\mathbf{kkrr}}\tilde{\Delta} = \sum_{\mu\nu}\int dt' d\mathbf{r}' \frac{d\omega}{(2\pi)} \frac{d\mathbf{k}}{(2\pi)^3} e^{-i\omega(t-t')+i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')}\tilde{\Delta}(t,\mathbf{r}) \times \frac{1}{8}(r'_\mu - r_\mu)(r'_\nu - r_\nu)\nabla_\mu\nabla_\nu\tilde{K}(\omega,\mathbf{k},t,\mathbf{r}) = -\frac{1}{8}\sum_{\mu\nu}\frac{\partial^4\tilde{K}_{0,\mathbf{0}}}{\partial r_\mu\partial r_\nu\partial k_\mu\partial k_\nu}\tilde{\Delta}. \tag{18}$$

These two terms are gradient corrections to the coefficient $\tilde{L}$. We will not discuss their contributions in detail.

III. KERNEL

Further progress requires an explicit kernel K. In the limit of zero gap, it is the noninteracting retarded two-particle propagator

$$\tilde{K}(t,\mathbf{r},t',\mathbf{r}') = -i\theta(t-t')(G_\uparrow^>(t,\mathbf{r},t',\mathbf{r}')G_\downarrow^>(t,\mathbf{r},t',\mathbf{r}') - \lessdot), \tag{19}$$

where $G_\uparrow^>$ is the correlation function for spin-up holes, $G_\downarrow^<$ is the correlation function for spin-down electrons, and so on. We use the abbreviation $G_\uparrow^> G_\downarrow^> - G_\uparrow^< G_\downarrow^< = G_\uparrow^> G_\downarrow^> - \lessdot$ for the electron-hole conjugated parts.

Since the kernel [Eq. (19)] does not depend on the gap Δ, it cannot be used to derive the cubic term $\propto|\Delta|^2\Delta$ of the GL theory. This cubic term is not expected to have important nonlocal corrections (see the discussion in de Gennes.[50] We simply add the cubic term to the resulting equation.

A. Quasiparticle approximation

For the correlation functions, we will use the quasiparticle approximation. To this end, we have to express the correlation functions in the Wigner mixed representation.

The quasiparticle approximation of the electron correlation function is

$$G_\uparrow^<(\varpi,\mathbf{q};t,\mathbf{r}) = 2\pi\delta(\varpi - \xi_{\mathbf{q},t,\mathbf{r}}^\uparrow)f_{\mathbf{q},t,\mathbf{r}}^\uparrow, \tag{20}$$

where $f_{\mathbf{q},t,\mathbf{r}}^\uparrow$ is the quasiparticle distribution and $\xi_{\mathbf{q},t,\mathbf{r}}^\uparrow$ is the quasiparticle energy. The hole correlation is proportional to the hole distribution,

$$G_\uparrow^>(\varpi,\mathbf{q},t,\mathbf{r}) = 2\pi\delta(\varpi - \xi_{\mathbf{q},t,\mathbf{r}}^\uparrow)(1 - f_{\mathbf{q},t,\mathbf{r}}^\uparrow). \tag{21}$$

The kernel in the Wigner mixed representation reads

$$\tilde{K}(\omega,\mathbf{k},t,\mathbf{r}) = -i\int_0^\infty dt''\int d\mathbf{r}''\frac{d\nu}{2\pi}\frac{d\mathbf{p}}{(2\pi)^3}\frac{d\varpi}{2\pi}\frac{d\mathbf{q}}{(2\pi)^3} \times[G_\uparrow^>(\varpi,-\mathbf{q},t,\mathbf{r})G_\downarrow^>(\nu,\mathbf{p},t,\mathbf{r}) - \lessdot] \times e^{i(\omega-\nu-\varpi)t''-i(\mathbf{k}-\mathbf{p}+\mathbf{q})\cdot\mathbf{r}''}. \tag{22}$$

All functions have the same space and time coordinates t, $\mathbf{r}$. Therefore, we will keep these variables implicit.

The space integration in Eq. (22) yields a delta function in wave vectors, so that one can readily integrate over $\mathbf{p}$,

$$\tilde{K}(\omega,\mathbf{k}) = -i\int\frac{d\nu}{2\pi}\frac{d\varpi}{2\pi}\frac{d\mathbf{q}}{(2\pi)^3}\int_0^\infty dt'' e^{i(\omega-\nu-\varpi)t''} \times[G_\uparrow^>(\varpi,-\mathbf{q})G_\downarrow^>(\nu,\mathbf{k}+\mathbf{q}) - \lessdot]. \tag{23}$$

The time integration is straightforward

$$\tilde{K}(\omega,\mathbf{k}) = \int\frac{d\nu}{2\pi}\frac{d\varpi}{2\pi}\frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\omega-\nu-\varpi+i0} \times[G_\uparrow^>(\varpi,-\mathbf{q})G_\downarrow^>(\nu,\mathbf{k}+\mathbf{q}) - \lessdot]. \tag{24}$$

The infinitesimal complex shift $i0$ denotes that the pole is below the real frequency axis.

Now, we substitute the quasiparticle approximations [Eqs. (20) and (21)] into the kernel [Eq. (24)]

$$\tilde{K}(\omega,\mathbf{k}) = \int\frac{d\nu}{2\pi}\frac{d\varpi}{2\pi}\frac{d\mathbf{q}}{(2\pi)^3}\frac{1 - f_{-\mathbf{q}}^\uparrow - f_{\mathbf{k}+\mathbf{q}}^\downarrow}{\omega-\nu-\varpi+i0} \times 2\pi\delta(\varpi - \xi_{-\mathbf{q}}^\uparrow)2\pi\delta(\nu - \xi_{\mathbf{k}+\mathbf{q}}^\downarrow) \tag{25}$$

and integrate over frequencies

$$\tilde{K}(\omega,\mathbf{k}) = \int\frac{d\mathbf{q}}{(2\pi)^3}\frac{1 - f_{-\mathbf{q}}^\uparrow - f_{\mathbf{k}+\mathbf{q}}^\downarrow}{\omega - \xi_{-\mathbf{q}}^\uparrow - \xi_{\mathbf{k}+\mathbf{q}}^\downarrow + i0}. \tag{26}$$

For a finite gap, the denominator in Eq. (27) depends on quasiparticle energies that are always different from zero. The pole can appear only in the case where the frequency ω exceeds $2|\Delta|$. We restrict our attention to slow frequencies, $\omega \ll |\Delta|$, for which the ratio yields only the principal value

$$\tilde{K}(\omega,\mathbf{k}) = \int\frac{d\mathbf{q}}{(2\pi)^3}\frac{1 - f_{-\mathbf{q}}^\uparrow - f_{\mathbf{k}+\mathbf{q}}^\downarrow}{\omega - \xi_{-\mathbf{q}}^\uparrow - \xi_{\mathbf{k}+\mathbf{q}}^\downarrow}. \tag{27}$$

In equilibrium, the kernel [Eq. (27)] is identical to the $\Delta\to 0$ limit of the kernel familiar from the Bogoliubov–de Gennes theory.

B. Relation of the gap to the Ginzburg-Landau function

The GL function differs from the gap by a multiplicative factor. The GL function $\tilde{\psi}$ is a subsidiary function with a norm adjusted to the supercurrent,

$$\mathbf{j}_s = -\frac{e^*}{m^*}\,\mathrm{Im}\,\bar{\tilde{\psi}}\nabla\tilde{\psi}, \tag{28}$$

where $m^* = 2m$ is a mass of the Cooper pair. As shown by Gor'kov,[1] the gap and the GL function have the same complex phase, and their amplitudes are related as

$$\tilde{\psi} = c\tilde{\Delta}. \tag{29}$$

In an equilibrium homogeneous system, c is a constant, but it can depend on time and place out of equilibrium.

The current is given by the gradient of the complex phase [see Eq. (28)]. This gradient is captured by the condensate velocity $\mathbf{v}_s$ defined via the supercurrent as

$$\mathbf{j}_s = e^* \mathbf{v}_s |\tilde{\psi}|^2. \tag{30}$$

The velocity enters quasiparticle energies and results in the supercurrent. In equilibrium,

$$\mathbf{j}_s = e\mathbf{v}_s n \frac{|\tilde{\Delta}|^2}{k_B^2 T^2} I^2, \tag{31}$$

where T is the actual temperature, n is the electron density, and

$$I^2 = -\int_0^\infty dx \frac{1}{x}\frac{d^2}{dx^2}\frac{1}{e^x+1}. \tag{32}$$

Comparing Eqs. (30) and (31), one finds the constant

$$c = \sqrt{\frac{n}{2}}\frac{I}{k_B T}. \tag{33}$$

Below, we assume an inhomogeneous temperature. In this case, the constant c becomes a function of the position.

C. Relation of the gap equation to the time-dependent Ginzburg-Landau equation

The gradient approximation of the gap equation differs from the TDGL equation by a multiplicative factor cC, which can be established from the equilibrium homogeneous system. In this case, all gradients vanish, and the zeroth order term is proportional to the term $\alpha\psi$ of the GL equation,

$$\frac{1}{cC}\alpha\tilde{\psi} = \left(\frac{1}{V} + \tilde{L}\right)\tilde{\Delta}. \tag{34}$$

Using Eqs. (11) and (29) one finds

$$\frac{1}{C}\alpha = \frac{1}{V} + \tilde{K}_{0,\mathbf{0}}. \tag{35}$$

Since α is known,[1,50]

$$\alpha = \frac{6\pi^2 k_B^2 T_c}{7\zeta[3] E_F}(T - T_c), \tag{36}$$

we use relation (35) to establish factor C. Here, $\zeta[3] = 1.202\,057$ is Riemann's zeta function and T_c is the critical temperature.

The customary algebra of the BCS type is enclosed in Appendix A. The resulting relation [Eq. (A11)] yields the constant C as

$$C = \frac{8\pi^2}{7\zeta[3]}\frac{k_B^2 T_c^2}{n}. \tag{37}$$

With this constant, we have to multiply all $\tilde{L}$ coefficients to obtain terms compatible with the customary GL equation.

D. Anisotropic coefficient

To complete the general step, we express the anisotropic coefficient $\tilde{L}_\mathbf{k}$ in terms of the quasiparticle distributions $f^\uparrow$ and $f^\downarrow$. By using kernel (27) in coefficient (15), we find

$$\begin{aligned}\tilde{L}_\mathbf{k} &= -i\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{\partial}{\partial \mathbf{k}}\frac{1 - f^\uparrow_{-\mathbf{q}} - f^\downarrow_{\mathbf{k}+\mathbf{q}}}{\xi^\uparrow_{-\mathbf{q}} + \xi^\downarrow_{\mathbf{k}+\mathbf{q}}}\Bigg|_{\mathbf{k}=0} \\ &= i\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi^\uparrow_{-\mathbf{q}} + \xi^\downarrow_\mathbf{q}}\frac{\partial f^\downarrow_\mathbf{q}}{\partial \mathbf{q}} + i\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1 - f^\uparrow_{-\mathbf{q}} - f^\downarrow_\mathbf{q}}{(\xi^\uparrow_{-\mathbf{q}} + \xi^\downarrow_\mathbf{q})^2}\frac{\partial \xi^\downarrow_\mathbf{q}}{\partial \mathbf{q}}.\end{aligned} \tag{38}$$

According to formula (38), the anisotropic coefficient $\tilde{L}_\mathbf{k}$ preferably depends on the spin-down electrons. There is no reason for such nonsymmetry in spins. By substituting $\mathbf{q}-\mathbf{k}$ for $\mathbf{q}$, one obtains a formula for $\tilde{L}_\mathbf{k}$, which is identical to Eq. (38), except for interchanged spin-up and spin-down functions. In this way, one can derive an explicitly symmetric formula,

$$\begin{aligned}\tilde{L}_\mathbf{k} &= \frac{i}{2}\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi^\uparrow_{-\mathbf{q}} + \xi^\downarrow_\mathbf{q}}\frac{\partial f^\downarrow_\mathbf{q}}{\partial \mathbf{q}} + \frac{i}{2}\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1 - f^\uparrow_{-\mathbf{q}} - f^\downarrow_\mathbf{q}}{(\xi^\uparrow_{-\mathbf{q}} + \xi^\downarrow_\mathbf{q})^2}\frac{\partial \xi^\downarrow_\mathbf{q}}{\partial \mathbf{q}} \\ &\quad + (\uparrow \leftrightarrow \downarrow).\end{aligned} \tag{39}$$

Below, we discuss perturbations symmetric in spin-up and spin-down components. For such cases, formulas (38) and (39) give formally identical expressions. The second-order coefficient $\tilde{L}_{\mathbf{kkr}}$ is discussed in Sec. VII.

IV. PERTURBED DISTRIBUTION

To demonstrate a possible physical mechanism covered by the gradient term $\tilde{L}_\mathbf{k}\nabla\tilde{\Delta}$, we evaluate the anisotropic coefficient $\tilde{L}_\mathbf{k}$ from an electron distribution disturbed from equilibrium by an applied force and a temperature gradient. We assume that this perturbation is very small and keep only linear terms in the force and the temperature gradient.

We adopt the following simplifying assumptions. First, the quasiparticle energy is independent of spin and parabolic

$$\xi^\uparrow_{\mathbf{q},t,\mathbf{r}} = \xi_q = \frac{q^2}{2m} - E_F, \tag{40}$$

where E_F is the Fermi energy. Second, there is no external spin-dependent mechanism so that the distribution is also independent of spin

$$f^\uparrow_\mathbf{q} = f^\downarrow_\mathbf{q} = f_\mathbf{q}. \tag{41}$$

These assumptions allow us to simplify the integrand of Eq. (39) as

$$\tilde{L}_\mathbf{k} = i\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi_q + \xi_q}\frac{\partial f_\mathbf{q}}{\partial \mathbf{q}} + i\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1 - f_{-\mathbf{q}} - f_\mathbf{q}}{(\xi_q + \xi_q)^2}\frac{\partial \xi_q}{\partial \mathbf{q}}. \tag{42}$$

By integrating by parts in the second integral, one rearranges Eq. (42) as

$$\tilde{L}_{\mathbf{k}} = i\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{2\xi_q}\frac{\partial f_{\mathbf{q}}}{\partial \mathbf{q}} + \frac{i}{2}\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{2\xi_q}\frac{\partial}{\partial \mathbf{q}}(1 - f_{-\mathbf{q}} - f_{\mathbf{q}})$$

$$= \frac{i}{2}\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{2\xi_q}\frac{\partial}{\partial \mathbf{q}}(f_{\mathbf{q}} - f_{-\mathbf{q}}). \qquad (43)$$

As one can see, the anisotropic coefficient is given by the odd part of the quasiparticle distribution, $f_{\mathbf{q}} - f_{-\mathbf{q}}$. Accordingly, it vanishes in equilibrium even if there is a gradient of material properties, e.g., a gradient in the alloy composition.

A. Relaxation time approximation

For a simple estimate of the quasiparticle distribution, we use the Boltzmann equation

$$\frac{\partial f}{\partial t} + \frac{\mathbf{q}}{m}\nabla f + e(\mathbf{E} + \mathbf{v}\times\mathbf{B})\frac{\partial f}{\partial \mathbf{q}} = -\frac{1}{\tau}(f - f^{\mathrm{le}}). \qquad (44)$$

Relaxation processes are characterized by a single material parameter, the relaxation time τ. The velocity $\mathbf{q}/m$ corresponds to the parabolic energy band we assume here.

The major part of the force is the Lorentz force $e\mathbf{v}\times\mathbf{B}$, which is generally largely and has to be included nonperturbatively. In contrast, the electric force $e\mathbf{E}$ due to the time dependence of the vector potential will be treated as a small perturbation.

We split the distribution into a local equilibrium part and a perturbation $f = f^{\mathrm{le}} + f'$. The local equilibrium part $f^{\mathrm{le}}_{\mathbf{q}} = f_{\mathrm{FD}}[\beta(\xi_q - \mu)]$, where μ is a small departure of the chemical potential from zero due to the final temperature.

We assume that the flow of particles due to the electric force $e\mathbf{E}$ and an eventual temperature gradient is steady; i.e., the distribution does not depend on time. To the linear order in electric force or in the temperature gradient, the distribution satisfies the Boltzmann equation

$$\frac{\mathbf{q}}{m}\nabla f^{\mathrm{le}} + e\mathbf{E}\frac{\partial f^{\mathrm{le}}}{\partial \mathbf{q}} + e\mathbf{v}\times\mathbf{B}\frac{\partial f'}{\partial \mathbf{q}} = -\frac{1}{\tau}f'. \qquad (45)$$

The Lorentz force multiplies the perturbation. The scalar product of the Lorentz force with the momentum derivative of the local equilibrium vanished since the derivative $\frac{\partial}{\partial \mathbf{q}}f^{\mathrm{le}} = \mathbf{q}\frac{1}{m}\frac{\partial}{\partial \xi}f^{\mathrm{le}}$ is parallel to $\mathbf{q}$, while the Lorentz force $\frac{e}{m}\mathbf{q}\times\mathbf{B}$ is perpendicular to $\mathbf{q}$.

The space gradient of the local equilibrium part is proportional to the gradient of the temperature and the chemical potential. Therefore,

$$f' + \frac{\tau e}{m}\mathbf{q}\times\mathbf{B}\frac{\partial f'}{\partial \mathbf{q}} = -\frac{\tau}{m}\left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta\xi}[\xi\mathbf{q}\nabla\beta + \beta\mathbf{q}(e\mathbf{E} - \nabla\mu)]. \qquad (46)$$

The gradient of the chemical potential is linked to the temperature gradient,

$$\nabla\mu = \frac{\pi^2}{6E_F\beta^3}\nabla\beta. \qquad (47)$$

For details, see Appendix B. Since μ is proportional to the gradient of temperature multiplied by distance from some reference point, we take it as a small quantity. In this spirit, we have already used $\varepsilon = \beta(\xi - \mu) \approx \beta\xi$ in the right hand side of Eq. (46).

B. Zero magnetic field

In the zero magnetic field, the Boltzmann equation [Eq. (46)] directly provides the perturbation of the distribution

$$f^{0\prime} = -\frac{\tau}{m}\left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta\xi}[\xi\mathbf{q}\nabla\beta + \beta\mathbf{q}(e\mathbf{E} - \nabla\mu)]. \qquad (48)$$

The local equilibrium distribution is even in $\mathbf{q}$; therefore, it does not contribute to the coefficient [Eq. (43)]. The coefficient is fully given by the perturbation $f^{0\prime}$, which is odd in wave vector $\mathbf{q}$,

$$\tilde{L}^0_{\mathbf{q}} = -\frac{i}{2}\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi}\frac{\partial}{\partial \mathbf{q}}f^{0\prime}_{\mathbf{q}}. \qquad (49)$$

We have used $f^{0\prime}_{-\mathbf{q}} = -f^{0\prime}_{\mathbf{q}}$ to cancel the factor of 2 in the ratio. From expression (46), one can see that the anisotropic coefficient $\tilde{L}^0_{\mathbf{k}}$ is proportional to the electric force and space gradients, as indicated above.

C. Finite magnetic field

The perturbation of the distribution for the finite magnetic field can be readily evaluated from the zero-field value. To show this link we first express the zero-field perturbation [Eq. (48)] in the form

$$f^{0\prime} = \mathbf{F}^0\mathbf{q}, \qquad (50)$$

where

$$\mathbf{F}^0 = -\frac{\tau}{m}\left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta\xi}[\xi\nabla\beta + \beta(e\mathbf{E} - \nabla\mu)]. \qquad (51)$$

Note that the vector function $\mathbf{F}^0$ does not depend on the direction of the momentum $\mathbf{q}$.

We expect the perturbation in the finite magnetic field to be of similar form,

$$f' = \mathbf{F}\mathbf{q}, \qquad (52)$$

where $\mathbf{F}$ is also independent of the direction of the momentum; i.e., it is a function of the energy ξ.

The momentum derivative multiplied by the Lorentz force does not affect functions of the energy. Indeed, for component F_α, the following holds:

$$\frac{e}{m}\mathbf{q}\times\mathbf{B}\frac{\partial F_\alpha}{\partial \mathbf{q}} = \frac{e}{m}\mathbf{q}\times\mathbf{B}\frac{\partial F_\alpha}{\partial \xi}\frac{\mathbf{q}}{m} = 0 \qquad (53)$$

because the Lorentz force $(e/m)\mathbf{q}\times\mathbf{B}$ is perpendicular to the velocity $\mathbf{q}/m$. By substituting Eq. (52) into the Boltzmann equation [Eq. (46)], we find

$$\mathbf{F}\mathbf{q} + \frac{\tau e}{m}[\mathbf{q}\times\mathbf{B}]\mathbf{F} = \mathbf{F}^0\mathbf{q}. \qquad (54)$$

Equation (54) holds for any direction of $\mathbf{q}$, while $\mathbf{F}$ and $\mathbf{F}^0$ are independent of this direction. One can thus eliminate $\mathbf{q}$.

Using the vector identity $[\mathbf{q}\times\mathbf{B}]\mathbf{F}=[\mathbf{B}\times\mathbf{F}]\mathbf{q}$, one arrives at a simple equation for $\mathbf{F}$,

$$\mathbf{F}+\frac{\tau e}{m}[\mathbf{B}\times\mathbf{F}]=\mathbf{F}^0. \tag{55}$$

From this relation, we can evaluate $\mathbf{F}$ in terms of $\mathbf{F}^0$. To this end, we first project Eq. (55) on the magnetic field,

$$\mathbf{B}\mathbf{F}=\mathbf{B}\mathbf{F}^0. \tag{56}$$

The vector product of Eq. (55) with the magnetic field yields

$$\mathbf{B}\times\mathbf{F}+\frac{\tau e}{m}(\mathbf{B}\mathbf{F}^0)\mathbf{B}-\frac{\tau e}{m}B^2\mathbf{F}=\mathbf{B}\times\mathbf{F}^0. \tag{57}$$

We have used identity $\mathbf{B}\times[\mathbf{B}\times\mathbf{F}]=(\mathbf{B}\mathbf{F})\mathbf{B}-B^2\mathbf{F}$ and Eq. (56) to simplify the dual vector product. By substituting $\mathbf{B}\times\mathbf{F}$ from Eq. (57) into Eq. (55), one arrives at

$$\mathbf{F}=\frac{1}{1+\dfrac{\tau^2e^2}{m^2}B^2}\left(\mathbf{F}^0-\frac{\tau e}{m}\mathbf{B}\times\mathbf{F}^0+\frac{\tau^2e^2}{m^2}(\mathbf{B}\mathbf{F}^0)\mathbf{B}\right). \tag{58}$$

The relation between $\mathbf{F}$ and $\mathbf{F}^0$ preserves the vector along the magnetic field, while in the perpendicular plane it represents a rotation in the space on the Hall angle and a reduction in length. Since many physical quantities undergo this transformation, it is advantageous to express it in the matrix form,

$$\mathbf{F}=\mathsf{R}\mathbf{F}^0, \tag{59}$$

where R is a 3×3 matrix with elements

$$\mathsf{R}_{ij}=\frac{1}{1+\dfrac{\tau^2e^2}{m^2}B^2}\left(\delta_{ij}+\frac{\tau e}{m}\sum_k\epsilon_{ijk}B_k+\frac{\tau^2e^2}{m^2}B_iB_j\right), \tag{60}$$

where δ_{ij} is the Kronecker delta and ϵ_{ijk} is the Levi-Civita symbol.

Function (58) can be used to evaluate the anisotropic correction in the presence of the magnetic field,

$$\widetilde{L}_{\mathbf{q}}=-\frac{i}{2}\int\frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi}\frac{\partial}{\partial\mathbf{q}}f'_{\mathbf{q}}=-\frac{i}{2}\int\frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi}\frac{\partial}{\partial\mathbf{q}}\mathbf{F}\mathbf{q}. \tag{61}$$

In the right hand side of formula (58), the only function of the momentum is $\mathbf{F}^0$. The anisotropic correction for the finite magnetic field is a linear combination of the zero-field coefficient,

$$\widetilde{L}_{\mathbf{q}}=\mathsf{R}\widetilde{L}^0_{\mathbf{q}}. \tag{62}$$

Clearly, it is sufficient to evaluate the zero-field anisotropic correction. The finite field value is obtained from Eq. (62).

V. ANISOTROPIC CORRECTION

Now, we are ready to evaluate the anisotropic correction to the TDGL equation. In the first step, we evaluate the coefficient $\widetilde{L}_{\mathbf{k}}$ for the perturbed electron distribution. In the second step, we transform this gradient of the gap into the gradient of the GL function.

A. Anisotropic term in the gap equation

According to relation (62), it is sufficient to evaluate the coefficient $\widetilde{L}^0_{\mathbf{k}}$. After substitution of the zero-field perturbation $f^{0\prime}$ from Eq. (48) in the anisotropic coefficient [Eq. (49)], we have

$$\begin{aligned}\widetilde{L}^0_\alpha&=-\frac{i}{2}\frac{\tau}{m}\int\frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi}\frac{\partial}{\partial q_\alpha}\left.\frac{\partial f_{\mathrm{FD}}}{\partial\varepsilon}\right|_{\varepsilon=\beta\xi}\\&\quad\times[\xi\mathbf{q}\,\nabla\,\beta+\beta\mathbf{q}(e\mathbf{E}-\nabla\mu)]\\&=-\frac{i}{2}\frac{\tau}{m}\int\frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi}\Bigg[\left.\frac{\partial^2 f_{\mathrm{FD}}}{\partial\varepsilon^2}\right|_{\varepsilon=\beta\xi}\beta\frac{q_\alpha}{m}\\&\quad\times[\xi\mathbf{q}\,\nabla\,\beta+\beta\mathbf{q}(e\mathbf{E}-\nabla\mu)]\\&\quad+\left.\frac{\partial f_{\mathrm{FD}}}{\partial\varepsilon}\right|_{\varepsilon=\beta\xi}\left(\frac{q_\alpha}{m}\mathbf{q}\,\nabla\,\beta+\xi\nabla_\alpha\beta+\beta(eE_\alpha-\nabla_\alpha\mu)\right)\Bigg].\end{aligned} \tag{63}$$

Here, we denote a single component of the vector $\widetilde{L}^0_{\mathbf{k}}\equiv(\widetilde{L}^0_x,\widetilde{L}^0_y,\widetilde{L}^0_z)$ by $\widetilde{L}^0_\alpha$. The vectors in the vector notation form scalar products, e.g., $\mathbf{q}\nabla\beta\equiv\Sigma_\kappa q_\kappa\partial$.

The only nonzero contributions to the integral are due to even powers of q_α in the integrand,

$$\begin{aligned}\widetilde{L}^0_\alpha&=-\frac{i}{2}\frac{\tau}{m}\int\frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi}\\&\quad\times\Bigg[\left.\frac{\partial^2 f_{\mathrm{FD}}}{\partial\varepsilon^2}\right|_{\varepsilon=\beta\xi}\beta\frac{q_\alpha^2}{m}[\xi\nabla_\alpha\beta+\beta(eE_\alpha-\nabla_\alpha\mu)]\\&\quad+\left.\frac{\partial f_{\mathrm{FD}}}{\partial\varepsilon}\right|_{\varepsilon=\beta\xi}\left(\frac{q_\alpha^2}{m}\nabla_\alpha\beta+\xi\nabla_\alpha\beta+\beta(eE_\alpha-\nabla_\alpha\mu)\right)\Bigg].\end{aligned} \tag{64}$$

This expression reflects that in the isotropic system the perturbing gradient and a corresponding response have the same direction.

The integral over directions is conveniently achieved by the interchange $q_\alpha^2=\frac{1}{3}q^2=\frac{2}{3}(E_F+\xi)m$,

$$\begin{aligned}\widetilde{L}^0_\alpha&=-\frac{i}{2}\frac{\tau}{m}\int\frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi}\Bigg[\left.\frac{\partial^2 f_{\mathrm{FD}}}{\partial\varepsilon^2}\right|_{\beta\xi}\beta\frac{2}{3}(E_F+\xi)\\&\quad\times[\xi\nabla_\alpha\beta+\beta(eE_\alpha-\nabla_\alpha\mu)]\\&\quad+\left.\frac{\partial f_{\mathrm{FD}}}{\partial\varepsilon}\right|_{\beta\xi}\left(\frac{2}{3}(E_F+\xi)\nabla_\alpha\beta+\xi\nabla_\alpha\beta+\beta(eE_\alpha-\nabla_\alpha\mu)\right)\Bigg].\end{aligned} \tag{65}$$

The integrand depends on the momentum only via the energy. We can thus write it with the help of the single-spin density of states [defined in Appendix A, Eq. (A3)]

$$\begin{aligned}\tilde{L}^0_{\mathbf{k}} = -\frac{i}{2}\frac{\tau}{m}\int d\xi N_\xi \frac{1}{\xi}\Bigg[&\frac{\partial^2 f_{\rm FD}}{\partial\varepsilon^2}\bigg|_{\beta\xi} \beta\frac{2}{3}(E_F+\xi) \\ &\times[\xi\nabla\beta+\beta(e\mathbf{E}-\nabla\mu)] \\ &+ \frac{\partial f_{\rm FD}}{\partial\varepsilon}\bigg|_{\beta\xi}\left(\frac{2}{3}(E_F+\xi)\nabla\beta+\xi\nabla\beta+\beta(e\mathbf{E}-\nabla\mu)\right)\Bigg].\end{aligned} \tag{66}$$

We have restored the vector notation. Since the gradients and the electric field are the only vectors on the right hand side, there is no danger of confusion.

We split the anisotropic coefficient [Eq. (66)] into two terms,

$$\tilde{L}^0_{\mathbf{k}} = -\frac{i}{2}\frac{\tau}{m}(I_1+I_2), \tag{67}$$

with

$$\begin{aligned}I_1 &= \int d\xi N_\xi \frac{1}{\xi}\frac{\partial f_{\rm FD}}{\partial\varepsilon}\bigg|_{\beta\xi} \\ &\quad\times\left(\frac{2}{3}(E_F+\xi)\nabla\beta+\xi\nabla\beta+\beta(e\mathbf{E}-\nabla\mu)\right) \\ &= -\frac{1}{3\beta}(5N_0+2N_0'E_F)\nabla\beta\end{aligned} \tag{68}$$

and

$$\begin{aligned}I_2 &= \int d\xi N_\xi \frac{1}{\xi}\frac{\partial^2 f_{\rm FD}}{\partial\varepsilon^2}\bigg|_{\beta\xi}\beta\frac{2}{3}(E_F+\xi)[\xi\nabla\beta+\beta(e\mathbf{E}-\nabla\mu)] \\ &= \frac{7\zeta[3]}{3\pi^2}N_0\beta^2E_Fe\mathbf{E}+\frac{2}{3}\frac{1}{\beta}\left[\left(1-\frac{7\zeta[3]}{12}\right)N_0+N_0'E_F\right]\nabla\beta.\end{aligned} \tag{69}$$

Integrals I_1 and I_2 are evaluated in Appendix C. In formulas (68) and (69), we neglect terms as small as $1/(E_F\beta)^2$. Here, N_0 is the single-spin density of states at the Fermi level, and N_0' is the energy derivative of the single-spin density of states at the Fermi level. The anisotropic coefficient [Eq. (67)] with integrals (68) and (69) reads

$$\tilde{L}^0_{\mathbf{k}} = -\frac{i}{2}\frac{7\zeta[3]}{3\pi^2}\frac{\tau}{m}N_0E_F\beta^2e\mathbf{E}+\frac{i}{2}\left(1-\frac{7\zeta[3]}{18}\right)\frac{\tau N_0}{m\beta}\nabla\beta. \tag{70}$$

Within the parabolic band approximation, the density of states satisfies $N_0E_F=\frac{3}{4}n$, so that we get

$$\tilde{L}^0_{\mathbf{k}} = -i\frac{7\zeta[3]}{8\pi^2}\frac{\tau n\beta^2}{m}e\mathbf{E}+i\left(\frac{2}{3}-\frac{7\zeta[3]}{27}\right)\frac{\tau n}{mE_F\beta}\nabla\beta. \tag{71}$$

This is our final result for the anisotropic correction in the gap equation.

B. Anisotropic term of the Ginzburg-Landau equation

To transform the gradient correction $\tilde{L}^0_{\mathbf{k}}$ into the anisotropic term of the TDGL equation, we multiply it by c from Eq. (33) to transfer the gap into the GL function. Then, we multiply by C given by Eq. (37), so that it achieves a norm suitable for the TDGL equation,

$$cC\tilde{L}^0_{\mathbf{k}}\nabla\tilde{\Delta} = -i\frac{\tau}{m}e\mathbf{E}\nabla\tilde{\psi}-i\frac{8\pi^2}{3}\left(\frac{2}{7\zeta[3]}-\frac{1}{9}\right)\frac{\tau k_B^2T}{mE_F}(\nabla T)\nabla\tilde{\psi}. \tag{72}$$

We have neglected terms proportional to $e\mathbf{E}\nabla\ln c$ and $(\nabla T)\tilde{\psi}\nabla\ln c$ because c is proportional to the temperature gradient [see Eq. (33)], so that these terms are quadratic in the perturbing force.

We introduce the drift velocity of normal electrons

$$\mathbf{v}^0_n = \frac{2}{nm}\int\frac{d\mathbf{q}}{(2\pi)^3}\mathbf{q}f^{0\prime} = \frac{\tau e}{m}\mathbf{E}-\frac{\pi^2}{3}\frac{\tau k_B^2T}{mE_F}\nabla T \tag{73}$$

and a zero-drift anisotropy vector

$$\mathbf{s}^0_T = \frac{8\pi^2}{3}\left(\frac{2}{7\zeta[3]}+\frac{1}{72}\right)\frac{\tau k_B^2T}{mE_F}\nabla T, \tag{74}$$

where the numerical factor is $\frac{8\pi^2}{3}(\frac{2}{7\zeta[3]}+\frac{1}{72})=6.621\,23$. In terms of the drift velocity and the zero-drift vector, the anisotropic correction [Eq. (72)] reads

$$cC\tilde{L}^0_{\mathbf{k}}\nabla\tilde{\Delta} = -i\mathbf{v}^0_n\nabla\tilde{\psi}-i\mathbf{s}^0_T\nabla\tilde{\psi}. \tag{75}$$

Note that a nonzero gradient correction appears even if the so called normal current $\mathbf{j}_n=en_n\mathbf{v}_n$ is zero. Below, we show that the zero-drift anisotropy vector $\mathbf{s}_T$ is rather small. We expect that it can be neglected under usual conditions.

Finally, we express the anisotropic correction at the finite magnetic field. The drift velocity and the zero-drift vector at the finite magnetic field follow relation (59) as

$$\mathbf{v}_n = \mathsf{R}\mathbf{v}^0_n, \tag{76}$$

$$\mathbf{s}_T = \mathsf{R}\mathbf{s}^0_T. \tag{77}$$

Comparing Eqs. (76) and (77) with Eq. (62), one can see that the anisotropic correction to the TDGL equation is formally identical to the zero-field case,

$$cC\tilde{L}_{\mathbf{k}}\nabla\tilde{\Delta} = -i\mathbf{v}_n\nabla\tilde{\psi}-i\mathbf{s}_T\nabla\tilde{\psi}. \tag{78}$$

The TDGL equation modified by the $\tilde{L}_{\mathbf{k}}$ term reads

$$\begin{aligned}&-\frac{1}{2m^*}\nabla^2\tilde{\psi}-i(\mathbf{v}_n+\mathbf{s}_T)\nabla\tilde{\psi}+\alpha_{\rm GL}\tilde{\psi}+\beta_{\rm GL}|\tilde{\psi}|^2\tilde{\psi} \\ &\quad=\Gamma_{\rm GL}\left(-i\frac{\partial}{\partial t}-\mu^*\right)\tilde{\psi},\end{aligned} \tag{79}$$

Let us write the gradient correction in a more convenient form. Neglecting nonlinear terms, the GL equation [Eq. (79)] can be expressed as

$$\frac{1}{2m^*}[i\nabla - m^*(\mathbf{v}_n + \mathbf{s}_T)]^2\tilde{\psi} + \alpha_{\mathrm{GL}}\tilde{\psi} + \beta_{\mathrm{GL}}|\tilde{\psi}|^2\tilde{\psi}$$
$$= \Gamma_{\mathrm{GL}}\left(-i\frac{\partial}{\partial t} - \mu^*\right)\tilde{\psi}. \tag{80}$$

Going back to the general gauge, one has

$$\frac{1}{2m^*}[i\nabla - m^*(\mathbf{v}_n + \mathbf{s}_T) - e^*\mathbf{A}]^2\bar{\psi} + \alpha_{\mathrm{GL}}\bar{\psi} + \beta_{\mathrm{GL}}|\psi|^2\bar{\psi}$$
$$= \Gamma_{\mathrm{GL}}\left(-i\frac{\partial}{\partial t} - \mu^*\right)\bar{\psi}. \tag{81}$$

The anisotropic correction enters the TDGL equation as a vector field, which adds to the vector potential. As soon as this vector field is known, one can use the standard solvers for the TDGL theory and directly modify to cover the anisotropic correction.

In many cases, the temperature is not treated and the TDGL theory handles only the normal current and the supercurrent. Such approach applies only to cases in which temperature gradients are small because the GL parameter α_{GL} is a function of the temperature. We show below that in these cases, the zero-drift term $\mathbf{s}_T$ can be neglected.

C. Neglect of $\mathbf{s}_T$

Let us first estimate the relative amplitude of the correction term due to the drift. Such estimates are convenient in the London gauge $\mathbf{A}_{\mathrm{Lon}} = \mathbf{A} - (1/e^*)\mathrm{Im}\nabla\ln\psi$, for which one has

$$\mathbf{j}_s = -\frac{e^{*2}}{m^*}\mathbf{A}_{\mathrm{Lon}}|\psi|^2. \tag{82}$$

This relation defines $\mathbf{A}_{\mathrm{Lon}}$ as a function of the supercurrent.

The normal current $\mathbf{j}_n = en_n\mathbf{v}_n$ determines the drift velocity $\mathbf{v}_n$. The relative contribution of the anisotropic correction to the kinetic energy can be thus estimated as

$$\frac{m^*v_n}{e^*A_{\mathrm{Lon}}} = \frac{2|\psi|^2}{n_n}\frac{j_n}{j_s}. \tag{83}$$

In a conversion region or in the core of a moving vortex, the normal and supercurrents can be of comparable amplitudes. The ratio of superconducting to normal electrons $2|\psi|^2/n_n$ is small in the region of validity of the GL theory since it is restricted to the vicinity of the critical temperature T_c. Although not justified, the GL theory is often used for temperatures quite far from T_c. In such implementations, the gradient correction might be appreciable.

One can expect that in the majority of practical situations, the zero-drift anisotropic correction is negligible. Indeed, the upper estimate of the gradient of the GL function is the BCS coherence length $|\nabla\tilde{\psi}| < k_BT_c/v_F|\psi|$, where v_F is the Fermi velocity. From this, we find

$$|\mathbf{s}_T\nabla\tilde{\psi}| < \frac{k_B^3T_c^3}{E_F^2}l|\nabla\ln T||\psi|, \tag{84}$$

where $l = \tau v_F$ is a mean free path of electrons. Coefficients of the GL equation are of the order of the condensation energy per electron, which is $\sim k_B^2T_c^2/E_F$. The gradient correction is thus smaller than other terms by a factor $(k_BT_c/E_F)l|\nabla\ln T|$.

If the temperature gradient is imposed by external conditions on a macroscopic sample, the characteristic scale of the temperature gradient $1/|\nabla\ln T|$ appreciably exceeds the mean free path, making this term very small.

A temperature gradient in hot spots of microbridges is $1/|\nabla\ln T| \sim \sqrt{Kd/s}$, where K is the thermal conductivity, d is the bridge width, and s is the thermal coupling per area of the bridge to the heat bath.[51] With typical values for tin bridges,[51] $K \sim 0.025$ W/(cm K), $d \sim 10^{-7}$ m, and $s \sim 10$ W/(cm^2 K), one finds $1/|\nabla\ln T| \sim 10^{-6}$ m. The mean free path observed in the same samples[52] is also of the order of micrometers; i.e., the factor $l|\nabla\ln T|$ is of the order of unity. The temperature gradient contribution thus remains small as (k_BT_c/E_F), which we expect to be negligible in conventional superconductors. A more detailed discussion of temperature profiles in various narrow bridges can be found in Ref. 53.

The gradient in the core of a moving vortex has an even much shorter characteristic scale. One can expect the temperature changes on the Ginzburg-Landau coherence length, $1/|\nabla\ln T| \sim \xi_{\mathrm{GL}}$, so that the factor $l|\nabla\ln T|$ in clean materials might be larger than unity. Moreover, in the high-T_c materials, the ratio (k_BT_c/E_F) is not so small. Accordingly, one can be optimistic that the temperature gradient correction might become observable.

We note that the Boltzmann equation [Eq. (44)] does not include the electron-electron collisions. Since these binary collisions conserve the charge and the mean momentum, they tend to establish drifting Fermi-Dirac distribution $f_{\mathbf{q}} \to f_{\mathrm{FD}}(\xi_{\mathbf{q}-m\mathbf{v}_d})$. For this distribution, the zero-drift anisotropy vector vanishes, while the drift contribution remains unaffected. Briefly, strong electron-electron collisions further reduce the zero-drift anisotropic correction.

At least in conventional materials, the temperature gradient term is negligible; therefore, the conjugated TDGL equation [Eq. (81)] is simplified as

$$\frac{1}{2m^*}(i\nabla - m^*\mathbf{v}_n - e^*\mathbf{A})^2\bar{\psi} + \alpha_{\mathrm{GL}}\bar{\psi} + \beta_{\mathrm{GL}}|\psi|^2\bar{\psi}$$
$$= \Gamma_{\mathrm{GL}}\left(-i\frac{\partial}{\partial t} - \mu^*\right)\bar{\psi}, \tag{85}$$

which is a complex conjugated equation to the TDGL equation [Eq. (1)].

The TDGL equation [Eq. (1)] can be handled by standard tools developed for the TDGL theory without the anisotropic correction. Indeed, the new term merely adds to the vector potential.

VI. INTERPRETATION

The GL function ψ describes a gap Δ in the spectrum of single-electron excitation. If all electrons move with the mean velocity $\mathbf{v}_n$, the gap caused by their mutual interactions ought to be evaluated in the coordinate system moving with electrons. Briefly, the term usually interpreted as the kinetic energy of Cooper pairs has to be formulated as the kinetic energy of the relative motion of Cooper pairs with respect to the normal electrons.

The traditional view of the GL gradient term as the kinetic energy in the sample coordinate system has an advantage that the interaction of the Cooper pairs with the vector potential has a form that parallels the interaction of the quantum wave function with the vector potential. In particular, the supercurrent reminds the quantum-mechanical current. Modifying the gradient term, which parallels the kinetic energy, we have to clarify the supercurrent and the normal current.

A. Currents

The TDGL equation is often presented in the form

$$-\nabla\frac{\partial\mathcal{F}}{\partial\nabla\psi}+\frac{\partial\mathcal{F}}{\partial\psi}=\Gamma_{\mathrm{GL}}\left(-i\frac{\partial}{\partial t}-\mu^*\right)\bar{\psi}, \tag{86}$$

which combines the Lagrange variation of the density of free energy $\mathcal{F}$ with a phenomenological time derivative. A nonequilibrium free energy, which corresponds to the TDGL equations [Eqs. (1) and (85)], reads

$$\mathcal{F}=\frac{1}{2m^*}|(-i\nabla-m^*\mathbf{v}_n-e^*\mathbf{A})\psi|^2+\alpha_{\mathrm{GL}}|\psi|^2+\frac{1}{2}\beta_{\mathrm{GL}}|\psi|^4. \tag{87}$$

One can view this free energy as the usual GL free energy in a moving framework of coordinates.

The variational concept suggests the electric supercurrent as

$$\mathbf{j}_s^{\mathrm{an}}=\frac{\partial\mathcal{F}}{\partial\mathbf{A}}=\frac{e^*}{m^*}\,\mathrm{Re}\,\bar{\psi}(-i\nabla-m^*\mathbf{v}_n-e^*\mathbf{A})\psi. \tag{88}$$

This supercurrent with anisotropic correction covers all contributions of the GL function.

The background current is obtained sending the GL function to zero. It is given by the drift velocity $\mathbf{v}_n$ as

$$\mathbf{j}_b=en\mathbf{v}_n. \tag{89}$$

The total current is a sum of the supercurrent and the background current,

$$\mathbf{j}=\mathbf{j}_s^{\mathrm{an}}+\mathbf{j}_b=\frac{e^*}{m^*}\,\mathrm{Re}\,\bar{\psi}(-i\nabla-m^*\mathbf{v}_n-e^*\mathbf{A})\psi+en\mathbf{v}_n. \tag{90}$$

Introducing the normal density,

$$n_n=n-2|\psi|^2, \tag{91}$$

one readily rearranges the total current as

$$\mathbf{j}=\frac{e^*}{m^*}\,\mathrm{Re}\,\bar{\psi}(-i\nabla-e^*\mathbf{A})\psi+e\mathbf{v}_n n_n. \tag{92}$$

The first term

$$\mathbf{j}_s=\frac{e^*}{m^*}\,\mathrm{Re}\,\bar{\psi}(-i\nabla-e^*\mathbf{A})\psi \tag{93}$$

is the supercurrent of the isotropic GL theory. The second term

$$\mathbf{j}_n=e\mathbf{v}_n n_n \tag{94}$$

is the normal current given by the product of the drift velocity with the normal density.

The total current we have obtained from the theory with anisotropic corrections is identical to the customary GL theory. The only difference is in the fact that the contribution $e^*\mathbf{v}_n|\psi|^2$ is here attributed to the supercurrent while in the customary approach it is subtracted from the background current.

In the customary TDGL theory, one evaluates the normal current from the Ohm law,

$$\mathbf{j}_n=\frac{n_n}{n}\sigma_H\mathbf{E}, \tag{95}$$

where the Hall conductivity tensor $\sigma_H=\sigma\mathsf{R}$ is a product of the conductivity σ with the Hall rotation tensor R. Compared to the normal state, this current is reduced by the fraction of normal electrons. From this current, one can conveniently evaluate the drift velocity $\mathbf{v}_n$ and the anisotropic correction.

B. Relative motion of normal electrons with respect to Cooper pairs

Within the TDGL equation, the anisotropic gradient correction is naturally interpreted as the shift of the kinetic energy into the framework moving with normal electrons. One can alternatively formulate the request to work with the relative velocity in the framework moving with the Cooper pairs.

In the tilde representation, the supercurrent is covered by the gradient of the GL function [see Eq. (28)]. Writing the GL function via its modulus and phase, $\tilde{\psi}=|\tilde{\psi}|e^{i\theta}$, we get the supercurrent [Eq. (28)] as

$$\mathbf{j}_s=-\frac{e^*}{m^*}|\tilde{\psi}|^2\nabla\theta. \tag{96}$$

The momentum per Cooper pair evaluated from the supervelocity given by Eq. (30) is thus the gradient of phase

$$\mathbf{k}_s=m^*\mathbf{v}_s=\nabla\theta. \tag{97}$$

Neglecting the gradient of the modulus, the gradient of the GL function reads

$$\nabla\tilde{\psi}\approx|\tilde{\psi}|\nabla e^{i\theta}=i\mathbf{k}_s\tilde{\psi}. \tag{98}$$

In a system with a homogeneous supercurrent, this relation is exact. Since phases of the gap and the GL function are identical, we also have

$$\nabla\tilde{\Delta}\approx i\mathbf{k}_s\tilde{\Delta}. \tag{99}$$

Within this approximation, we can conveniently rearrange a sum of the nongradient term, the anisotropic correction, and the second-order gradient,

$$
\begin{aligned}
&\tilde{L}\tilde{\Delta} + \tilde{L}_{\mathbf{k}} \nabla \Delta + \tilde{L}_{\mathbf{kk}} \nabla \nabla \Delta \\
&\quad = \tilde{K}_{0,\mathbf{0}}\tilde{\Delta} - i\frac{\partial \tilde{K}_{0,\mathbf{0}}}{\partial \mathbf{k}} \nabla \tilde{\Delta} - \frac{1}{2}\frac{\partial^2 \tilde{K}_{0,\mathbf{0}}}{\partial \mathbf{k}\partial \mathbf{k}} \nabla \nabla \tilde{\Delta} \\
&\quad \approx \tilde{K}_{0,\mathbf{0}}\tilde{\Delta} + \frac{\partial \tilde{K}_{0,\mathbf{0}}}{\partial \mathbf{k}}\mathbf{k}_s\tilde{\Delta} + \frac{1}{2}\frac{\partial^2 \tilde{K}_{0,\mathbf{0}}}{\partial \mathbf{k}\partial \mathbf{k}}\mathbf{k}_s\mathbf{k}_s\tilde{\Delta} = \tilde{K}_{0,\mathbf{k}_s}\tilde{\Delta}.
\end{aligned}
\tag{100}
$$

We have used Eqs. (11), (14), and (15) to express $\tilde{L}$, $\tilde{L}_{\mathbf{k}}$, and $\tilde{L}_{\mathbf{kk}}$, respectively. As one can see, these three terms yield the kernel [Eq. (27)] evaluated at the momentum of a Cooper pair.

One can interpret the kernel as a balance between electrons entering and leaving the condensate of Cooper pairs. This process is open only for electron pairs of antiparallel spin and the sum momentum equal to the momentum per Cooper pair. When the argument of the integral kernel [Eq. (27)] is not isotropic, the pairing rate depends on a direction of the sum momentum $\mathbf{k}_s$. The difference is described by the anisotropic correction $\tilde{L}_{\mathbf{k}}\nabla\tilde{\Delta}$.

Briefly, to obtain the anisotropic gradient correction, one can either treat the motion of Cooper pairs in the framework moving with normal electrons or include the center-of-mass motion of normal electrons entering the moving Cooper pairs. These pictures are complementary and describe the same physical contribution to the TDGL theory.

VII. SECOND-ORDER GRADIENT

The anisotropic gradient correction $\tilde{L}_{\mathbf{k}}$ is zero in the absence of gradients in the system. Accordingly, $\tilde{L}_{\mathbf{k}}\nabla\tilde{\Delta}$ is, in fact, proportional to the second space derivative. To make the theory consistent, we have to also evaluate the second-order gradient term $\tilde{L}_{\mathbf{kkr}}$. This is done in this section.

Below, it is shown that $\tilde{L}_{\mathbf{kkr}}$ merely corrects a neglect commonly made while deriving the GL kinetic energy, but it does not bring any new physical contribution. The result oriented reader is encouraged to skip the algebra and continue from Eq. (112).

Many steps are parallel to the derivation of the first-order gradient $\tilde{L}_{\mathbf{k}}$ discussed above in detail. Here, we proceed faster.

From relation (16) and the kernel with the quasiparticle approximation [Eq. (27)], we arrive at

$$
\tilde{L}^{\alpha}_{\mathbf{kkr}} = \int \frac{d\mathbf{q}}{(2\pi)^3}\sum_{\nu} \nabla_{\nu}\frac{\partial}{\partial k_{\nu}}\frac{\partial}{\partial k_{\alpha}} \left.\frac{1 - f^{\uparrow}_{-\mathbf{q}} - f^{\downarrow}_{\mathbf{k+q}}}{\xi^{\uparrow}_{-\mathbf{q}} + \xi^{\downarrow}_{\mathbf{k+q}}}\right|_{\mathbf{k}=0}.
\tag{101}
$$

For the spin symmetric distribution, the second-order coefficient has an explicit form

$$
\begin{aligned}
\tilde{L}^{\alpha}_{\mathbf{kkr}} = \sum_{\nu} \nabla_{\nu}\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{4\xi^2_{\mathbf{q}}}\Bigg[&\frac{\partial \xi_{\mathbf{q}}}{\partial q_{\nu}}\frac{\partial f_{\mathbf{q}}}{\partial q_{\alpha}} + \frac{\partial f_{\mathbf{q}}}{\partial q_{\nu}}\frac{\partial \xi_{\mathbf{q}}}{\partial q_{\alpha}} - 2\xi_{\mathbf{q}}\frac{\partial^2 f_{\mathbf{q}}}{\partial q_{\nu}\partial q_{\alpha}} \\
&+ (1 - f_{-\mathbf{q}} - f_{\mathbf{q}})\left(-\frac{\partial^2 \xi_{\mathbf{q}}}{\partial q_{\nu}\partial q_{\alpha}} + \frac{1}{\xi_{\mathbf{q}}}\frac{\partial \xi_{\mathbf{q}}}{\partial q_{\alpha}}\frac{\partial \xi_{\mathbf{q}}}{\partial q_{\nu}}\right)\Bigg].
\end{aligned}
\tag{102}
$$

With the help of integration by parts, we rearrange it as

$$
\begin{aligned}
\tilde{L}^{\alpha}_{\mathbf{kkr}} = -\frac{1}{8}\sum_{\nu}\nabla_{\nu}\int \frac{d\mathbf{q}}{(2\pi)^3}\Bigg(&\frac{\partial^2(f_{-\mathbf{q}} + f_{\mathbf{q}})}{\partial q_{\alpha}\partial q_{\nu}}\frac{1}{\xi_{\mathbf{q}}} \\
&+ (1 - f_{-\mathbf{q}} - f_{\mathbf{q}})\frac{\partial^2 \xi_{\mathbf{q}}}{\partial q_{\nu}\partial q_{\alpha}}\frac{1}{\xi^2_{\mathbf{q}}}\Bigg).
\end{aligned}
\tag{103}
$$

Further progress requires an explicit quasiparticle distribution.

A. Local equilibrium approximation

Coefficient (103) depends on the even part of the quasiparticle distribution,

$$
f_{-\mathbf{q}} + f_{\mathbf{q}} = 2f^0_{\mathbf{q}}.
\tag{104}
$$

Within the relaxation time approximation, it implies that coefficient (103) depends on the local equilibrium part of the distribution. The distribution factor entering Eq. (103) then reads

$$
1 - 2f^0_{\mathbf{q}} = \tanh\left(\frac{\beta\xi_{\mathbf{q}}}{2}\right).
\tag{105}
$$

The second derivative of the parabolic energy band is a diagonal matrix

$$
\frac{\partial^2 \xi_{\mathbf{q}}}{\partial q_{\nu}\partial q_{\alpha}} = \delta_{\alpha\nu}\frac{1}{m}.
\tag{106}
$$

The second derivative of the distribution

$$
\frac{\partial^2 f^0_{\mathbf{q}}}{\partial q_{\alpha}\partial q_{\nu}} = \beta^2\frac{q_{\alpha}q_{\nu}}{m^2}\left.\frac{\partial^2 f^0}{\partial \varepsilon^2}\right|_{\varepsilon=\beta\xi} + \delta_{\alpha\nu}\beta\frac{1}{m}\left.\frac{\partial f^0}{\partial \varepsilon}\right|_{\varepsilon=\beta\xi}
\tag{107}
$$

contributes to integral (103) only by its diagonal terms. The integral thus reduces to two terms,

$$
\begin{aligned}
\tilde{L}^{\alpha}_{\mathbf{kkr}} = &-\frac{1}{8m}\nabla_{\alpha}\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi^2}\tanh\left(\frac{\beta\xi}{2}\right) \\
&-\frac{1}{4m}\nabla_{\alpha}\int \frac{d\mathbf{q}}{(2\pi)^3}\frac{1}{\xi}\left(\beta^2\frac{q^2_{\alpha}}{m}\frac{\partial^2 f^0}{\partial \varepsilon^2} + \beta\frac{\partial f^0}{\partial \varepsilon}\right)_{\varepsilon=\beta\xi}.
\end{aligned}
\tag{108}
$$

The integration over the direction of the momentum yields $\int q^2_{\alpha} = \frac{1}{3}\int q^2 = \frac{2}{3}m\int(E_F + \xi)$. We can thus express all integrations in terms of the energy integral,

$$
\begin{aligned}
\tilde{L}_{\mathbf{kkr}} = &-\frac{1}{8m}\nabla\int dEN_E\frac{1}{E^2}\tanh\left(\frac{\beta E}{2}\right) \\
&-\frac{1}{6m}\nabla\int dEN_E\beta^2\left.\frac{\partial^2 f^0}{\partial \varepsilon^2}\right|_{\varepsilon=\beta E}\frac{E_F + E}{E} \\
&-\frac{1}{4m}\nabla\int dEN_E\beta\left.\frac{\partial f^0}{\partial \varepsilon}\right|_{\varepsilon=\beta E}\frac{1}{E}.
\end{aligned}
\tag{109}
$$

We have introduced the density of states [Eq. (A3)]. From now on, we do not specify the vector component of the coefficient since the gradient and the coefficient are aligned as one can see from (108).

The linear approximation of the density of state near the Fermi level, $N_E \approx N_0 + N_0'E$, is sufficient because under space gradient, the relevant integration region is confined to the thermally activated energy region.

$\tanh(\varepsilon)$ and $\frac{\partial^2 f^0}{\partial\varepsilon^2}\big|_{\varepsilon=\beta E}$ are odd functions and $\frac{\partial f^0}{\partial\varepsilon}\big|_{\varepsilon=\beta E}$ is an even function of the energy E. Keeping even terms only, Eq. (109) reads

$$\begin{aligned}\widetilde{L}_{\mathbf{kkr}} = &-\frac{N_0'}{8m}\nabla\int dE\frac{1}{E}\tanh\left(\frac{\beta E}{2}\right)\\ &-\frac{N_0E_F}{6m}\nabla\int dE\frac{1}{E}\beta^2\left.\frac{\partial^2 f^0}{\partial\varepsilon^2}\right|_{\varepsilon=\beta E}\\ &-\frac{N_0'}{6m}\nabla\int dEE\beta^2\left.\frac{\partial^2 f^0}{\partial\varepsilon^2}\right|_{\varepsilon=\beta E}\\ &-\frac{N_0'}{4m}\nabla\int dE\beta\left.\frac{\partial f^0}{\partial\varepsilon}\right|_{\varepsilon=\beta E}.\end{aligned}\tag{110}$$

The third and the last integrals are independent of the temperature thus having zero gradient. We evaluate the second term with the help of the integral [Eq. (C7)]. In the first term we interchange the gradient with the integration, which leads to the integral over $1/\cosh^2\varepsilon$. In result

$$\widetilde{L}_{\mathbf{kkr}} = -\frac{N_0'}{4m\beta}\nabla\beta - \frac{7\zeta[3]}{6\pi^2}\frac{N_0E_F\beta}{m}\nabla\beta.\tag{111}$$

The first term of Eq. (111) is smaller than the second one by a factor $k_B^2T^2/E_F^2$. Within the accuracy we keep within this paper, we can ignore the first term and write the second-order anisotropic gradient correction as

$$\widetilde{L}_{\mathbf{kkr}}\nabla\widetilde{\Delta} = \frac{7\zeta[3]}{6\pi^2}\frac{N_0E_F}{mk_B^2T^3}(\nabla T)\nabla\widetilde{\Delta}.\tag{112}$$

This is the anisotropic gradient correction of the second order to the gap equation. Note that it does not depend on the normal current but exclusively on the gradient of the temperature.

B. Role of the second-order anisotropic coefficient in the GL equation

To convert the gap into the GL wave function we have to multiply it by c from Eq. (33). To convert the coefficient of the gap equation into the coefficient of the GL equation, we have to multiply it by C given by Eq. (37). By doing so, we arrive at the very simple expression

$$cC\widetilde{L}_{\mathbf{kkr}}\nabla\widetilde{\Delta} = \frac{1}{mT}(\nabla T)\nabla\widetilde{\psi}.\tag{113}$$

We have used that T is close to T_c to cancel them as $T_c/T=1$ after the gradient was taken. We have neglected a higher-order term proportional to $(\nabla T)^2$.

If we restrict our attention to perturbations caused by the temperature gradient, the amplitude of the second-order term [Eq. (113)] is much larger than the amplitude of the first-order term given by Eq. (74). Moreover, unlike $L_{\mathbf{k}}$, the term $L_{\mathbf{kkr}}$ is not imaginary but real; i.e., it has to affect the current conservation.

To clarify these points, we show that the second-order anisotropic correction [Eq. (113)] does not represent additional processes in the quasiparticle subsystem but merely complements the second-order gradient correction usually called the kinetic energy,

$$cC\widetilde{L}_{\mathbf{kk}}\nabla^2\widetilde{\Delta} = -\frac{1}{2m}c\nabla^2\widetilde{\Delta}.\tag{114}$$

Since $\psi=c\Delta$ and the factor c depends on temperature [Eq. (33)], to the first order in the temperature gradient, we have

$$cC\widetilde{L}_{\mathbf{kk}}\nabla^2\widetilde{\Delta} = -\frac{1}{2m}\nabla^2\widetilde{\psi} - \frac{1}{m}c\left(\nabla\frac{1}{c}\right)\nabla\widetilde{\psi}.\tag{115}$$

We have neglected terms proportional to $\nabla^2(1/c)$.

The logarithmic gradient of the factor c follows from Eq. (33), giving

$$cC\widetilde{L}_{\mathbf{kk}}\nabla^2\widetilde{\Delta} = -\frac{1}{2m}\nabla^2\widetilde{\psi} - \frac{1}{mT}(\nabla T)\nabla\widetilde{\psi}.\tag{116}$$

One can see that Eqs. (113) and (116) add by forming together the kinetic energy of the GL theory

$$cC\widetilde{L}_{\mathbf{kk}}\nabla^2\widetilde{\Delta} + cC\widetilde{L}_{\mathbf{kkr}}\nabla\widetilde{\Delta} = -\frac{1}{2m}\nabla^2\widetilde{\psi}.\tag{117}$$

Briefly, the second-order anisotropic correction merely covers a difference in the norm of the gap equation and the GL wave function. The current conservation has a simple expression within the GL theory. It shows that the correction $L_{\mathbf{kkr}}$ is vital to guarantee the current conservation.

VIII. CONCLUSIONS

We have shown that beyond the local equilibrium of quasiparticles, a new gradient term appears in the TDGL equation for the GL wave function. Within the relaxation time approximation, this term has two contributions in a form of the velocity vector field $\mathbf{v}_n+\mathbf{s}_T$ entering the kinetic energy. We expect that in the majority of cases, the dominant contribution results from the drift velocity $\mathbf{v}_n$ of normal electrons, while the zero-drift contribution $\mathbf{s}_T$ can be neglected. The drift velocity is directly related to the normal current, $\mathbf{v}_n=\sigma_H\mathbf{E}/(en_n)$, so that this correction does not require us to extend the set of the TDGL equations.

Perhaps, it is noteworthy that the inclusion of the anisotropic correction due to the normal currents is a step toward the consistency of the TDGL theory. Indeed, the normal current is a natural part of the TDGL theory as it is indispensable, e.g., in hot spots, in conversion layers, or in dissipative regions of moving vortices. The TDGL equation derived under the assumption of the local equilibrium of normal electrons is not consistent with the presence of the normal current. We should note, however, that for normal currents of amplitudes comparable to supercurrents, the anisotropic correction vanishes as $v_n\sim(|\psi|^2/n)j_s/(en)$, i.e., it effectively behaves as a nonlinear nonlocal correction. Such terms are usually neglected within the GL theory.

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APPENDIX A: EQUILIBRIUM KERNEL

To evaluate the equilibrium $\tilde{K}_{0,\mathbf{0}}$, we employ the Fermi-Dirac distribution,

$$f_{\mathbf{q}} = f_{\text{FD}}(\beta\xi_{\mathbf{q}}) = \frac{1}{e^{\beta\xi_{\mathbf{q}}}+1}, \tag{A1}$$

where $\beta = 1/k_BT$ is the inverse temperature in energy units. The distribution is independent of spin; therefore, relation (27) is simplified as

$$\tilde{K}_{0,\mathbf{0}} = -\frac{1}{2}\int \frac{d\mathbf{q}}{(2\pi)^3}(1-2f_{\mathbf{q}})\frac{1}{\xi_{\mathbf{q}}}. \tag{A2}$$

Since $k_BT_c \ll E_F$, it is possible to approximatively evaluate the integral in relation (A2). As usual, we introduce a single-spin density of states,

$$N_E = \int \frac{d\mathbf{q}}{(2\pi)^3}\delta(E-\xi_{\mathbf{q}}), \tag{A3}$$

in terms of which

$$\tilde{K}_{0,\mathbf{0}} = -\frac{1}{2}\int_{-E_F}^{\infty} dE N_E[1-2f_{\text{FD}}(\beta E)]\frac{1}{E}. \tag{A4}$$

In the spirit of the BCS model, the upper and lower limits has to be replaced by cutoffs $\pm\omega_{\text{cut}}$, i.e.,

$$\begin{aligned}\frac{1}{N_0}\tilde{K}_{0,\mathbf{0}} &= -\frac{1}{2}\int_{-\omega_{\text{cut}}}^{\omega_{\text{cut}}} dE[1-2f_{\text{FD}}(\beta E)]\frac{1}{E}\\ &= -\int_0^{(1/2)\beta\omega_{\text{cut}}}\frac{dy}{y}\frac{\sinh y}{\cosh y}\\ &\approx -0.818\,78 - \ln\left(\frac{1}{2}\beta\omega_{\text{cut}}\right)\\ &= -0.125\,63 - \ln(\beta\omega_{\text{cut}}).\end{aligned} \tag{A5}$$

The coefficient α/C from relation (34) and approximation (A5) results in

$$\frac{1}{C}\alpha = \frac{1}{V} - N_0[0.125\,63 + \ln(\beta\omega_{\text{cut}})]. \tag{A6}$$

At the critical temperature T_c, the GL parameter α equals zero. For $\beta = 1/k_BT_c$ and $C\alpha = 0$, we recover the BCS relation

$$k_BT_c = 1.1338\,\omega_{\text{cut}}e^{-1/VN_0}. \tag{A7}$$

For T close to T_c, we can expand in $T_c - T$,

$$\begin{aligned}\frac{1}{C}\alpha &= \frac{1}{V} - N_0\left(0.125\,63 + \ln\frac{\omega_{\text{cut}}}{k_BT}\right)\\ &= \frac{1}{V} - N_0\left(0.125\,63 + \ln\frac{\omega_{\text{cut}}}{k_BT_c} - \ln\frac{T}{T_c}\right)\\ &= N_0\ln\frac{T}{T_c} \approx \frac{N_0}{T_c}(T-T_c).\end{aligned} \tag{A8}$$

From Eqs. (A8) and (36) follows

$$\frac{1}{C} = \frac{7\zeta[3]E_FN_0}{6\pi^2k_B^2T_c^2}. \tag{A9}$$

Here, $\zeta[3] = 1.202\,057$ is Riemann's zeta function.

For the parabolic band, the density of states times the Fermi energy gives the electron density

$$\frac{4}{3}E_FN_0 = n. \tag{A10}$$

By using Eq. (A10) in Eq. (A9), one finds

$$C = \frac{8\pi^2}{7\zeta[3]}\frac{1}{n\beta^2} = 9.383\,538\frac{k_B^2T_c^2}{n}. \tag{A11}$$

APPENDIX B: GRADIENT OF THE CHEMICAL POTENTIAL

For a fixed density of electrons, the chemical potential depends on the temperature. The gradient of the chemical potential induced by the gradient of the temperature is evaluated in this Appendix.

The Coulomb forces are very strong; therefore, the system stays very close to the local neutrality. If we neglect the thermal expansion of the lattice, this implies that the density of electrons is constant $\nabla n = 0$.

The electron density

$$n = 2\int\frac{d\mathbf{q}}{(2\pi)^3}f = 2\int\frac{d\mathbf{q}}{(2\pi)^3}f^0 \tag{B1}$$

does not depend on the perturbation, which is odd in momentum. Accordingly, n depends on the local equilibrium part given by the Fermi-Dirac distribution

$$f_{\mathbf{q}}^0 = f_{\text{FD}}[\beta(\xi_{\mathbf{q}}-\mu)] = \frac{1}{e^{\beta(\xi_{\mathbf{q}}-\mu)}+1}, \tag{B2}$$

which includes a small deviation μ of the chemical potential from its zero-temperature value E_F. From Eqs. (B1) and (B2) follows

$$\nabla n = 2\int\frac{d\mathbf{q}}{(2\pi)^3}\left.\frac{\partial f_{\text{FD}}}{\partial\varepsilon}\right|_{\varepsilon=\beta\xi}(\xi\nabla\beta - \beta\nabla\mu). \tag{B3}$$

The neutrality condition $\nabla n = 0$ links the gradient of the chemical potential to the gradient of temperature

$$\beta \nabla \mu \int \frac{d\mathbf{q}}{(2\pi)^3} \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta\xi} = \nabla \beta \int \frac{d\mathbf{q}}{(2\pi)^3} \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta\xi} \xi. \tag{B4}$$

In terms of the density of states [Eq. (A3)], relation (B4) reads

$$\beta \nabla \mu \int dEN_E \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta E} = \nabla \beta \int dEN_E \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta E} E. \tag{B5}$$

For thermal energies $E \sim k_B T \ll E_F$, the density of states can be approximated by the linear expansion near the Fermi energy,

$$N_E \approx N_0 + \left.\frac{\partial N}{\partial E}\right|_{E=0} E = N_0 + N_0' E. \tag{B6}$$

The derivative of the Fermi-Dirac distribution is an even function of the energy; therefore, relation (B5) is reduced to

$$\beta \nabla \mu N_0 \int dE \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta E} = \nabla \beta N_0' \int dE \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta E} E^2. \tag{B7}$$

Using

$$\int dE \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta E} = -\frac{1}{\beta} \tag{B8}$$

and

$$\int dE \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\varepsilon=\beta E} E^2 = -\frac{1}{\beta^3}\frac{\pi^2}{3} \tag{B9}$$

one obtains

$$\nabla \mu = \frac{\pi^2}{3}\frac{1}{\beta^3}\frac{N_0'}{N_0} \nabla \beta. \tag{B10}$$

For the parabolic electron band structure, the density of states is of form $N_E \propto \sqrt{E+E_F}$. In this case,

$$\frac{N_0'}{N_0} = \frac{1}{2E_F}, \tag{B11}$$

and the gradient of the chemical potential achieves a simple form,

$$\nabla \mu = \frac{\pi^2}{6E_F \beta^3} \nabla \beta. \tag{B12}$$

From $\beta = 1/k_B T$ follows

$$\nabla \mu = -\frac{\pi^2}{6}\frac{k_B T}{E_F} k_B \nabla T. \tag{B13}$$

The gradient of μ is thus of the order of $k_B T/E_F$; i.e., it gives only a small contribution.

APPENDIX C: INTEGRALS I_1 AND I_2

In the integrand of Eq. (68), the product of functions $\frac{1}{\xi}\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}$ is odd in ξ. It is thus sufficient to keep the odd part of the remaining terms in the integrand. In the linear approximation of the density of states, $N_\xi = N_0 + N_0' \xi$, the first integral is simplified as

$$I_1 = \left[\left(\frac{5}{3}N_0 + \frac{2}{3}N_0' E_F\right) \nabla \beta + N_0' \beta (e\mathbf{E} - \nabla \mu)\right] \int d\xi \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\beta\xi}. \tag{C1}$$

The energy integral over distribution equals to the temperature

$$\int d\xi \left.\frac{\partial f_{\mathrm{FD}}}{\partial \varepsilon}\right|_{\beta\xi} = -\frac{1}{\beta}. \tag{C2}$$

Therefore,

$$I_1 = -\frac{1}{\beta}\left[\left(\frac{5}{3}N_0 + \frac{2}{3}N_0' E_F\right) \nabla \beta + N_0' \beta (e\mathbf{E} - \nabla \mu)\right]. \tag{C3}$$

The gradient of the chemical potential can be neglected in I_1 as it yields a contribution smaller by $(k_B T/E_F)^2$ than the gradient of temperature

$$I_1 = -N_0' e\mathbf{E} - \frac{1}{3\beta}(5N_0 + 2N_0' E_F) \nabla \beta. \tag{C4}$$

In the integrand of Eq. (69), the product of functions $\frac{1}{\xi}\frac{\partial^2 f_{\mathrm{FD}}}{\partial \varepsilon^2}$ is even in ξ. It is thus sufficient to keep the even part of remaining terms in the integrand. In the linear approximation of the density of states, $N_\xi = N_0 + N_0' \xi$, this integral is simplified as

$$\begin{aligned} I_2 = {} & \frac{2}{3}N_0 \beta^2 E_F (e\mathbf{E} - \nabla \mu) \int d\xi \frac{1}{\xi} \left.\frac{\partial^2 f_{\mathrm{FD}}}{\partial \varepsilon^2}\right|_{\beta\xi} \\ & + \frac{2}{3}\beta[(N_0 + N_0' E_F) \nabla \beta \\ & + N_0' \beta (e\mathbf{E} - \nabla \mu)] \int d\xi \xi \left.\frac{\partial^2 f_{\mathrm{FD}}}{\partial \varepsilon^2}\right|_{\beta\xi}. \end{aligned} \tag{C5}$$

By using

$$\int d\xi\,\xi \left.\frac{\partial^2 f_{\mathrm{FD}}}{\partial \varepsilon^2}\right|_{\beta\xi} = \frac{1}{\beta^2} \tag{C6}$$

and

$$\int d\xi \frac{1}{\xi} \left.\frac{\partial^2 f_{\mathrm{FD}}}{\partial \varepsilon^2}\right|_{\beta\xi} = \int dx \frac{1}{x}\frac{\partial^2 f_{\mathrm{FD}}}{\partial x^2} = \frac{1}{4}\int dx \frac{1}{x}\frac{\sinh\frac{x}{2}}{\cosh^3\frac{x}{2}}$$

$$= \frac{1}{2}\int_0^\infty \frac{dy}{y}\frac{\sinh y}{\cosh^3 x} = \frac{7\zeta[3]}{2\pi^2} = 0.426\,278, \tag{C7}$$

we can write down the second integral

$$I_2 = \frac{7\zeta[3]}{3\pi^2} N_0 \beta^2 E_F (e\mathbf{E} - \nabla\mu) + \frac{2}{3}\frac{1}{\beta}[(N_0 + N_0' E_F)\nabla\beta + N_0'\beta(e\mathbf{E} - \nabla\mu)]. \tag{C8}$$

The second term proportional to $e\mathbf{E} - \nabla\mu$ is small as $(k_B T/E_F)^2$ compared to the first term. Therefore, we neglect it,

$$I_2 = \frac{7\zeta[3]}{3\pi^2} N_0 \beta^2 E_F (e\mathbf{E} - \nabla\mu) + \frac{2}{3}\frac{1}{\beta}(N_0 + N_0' E_F)\nabla\beta. \tag{C9}$$

The first term of Eq. (C9) yields the only non-negligible contribution of the gradient of the chemical potential. We substitute its value driven by the temperature gradient from Eq. (47)

$$I_2 = \frac{7\zeta[3]}{3\pi^2} N_0 \beta^2 E_F e\mathbf{E} + \frac{2}{3}\frac{1}{\beta}\left[\left(1 - \frac{7\zeta[3]}{12}\right)N_0 + N_0' E_F\right]\nabla\beta. \tag{C10}$$

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