Gauge-invariant microscopic kinetic theory of superconductivity in response to electromagnetic fields

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Within a gauge-invariant microscopic kinetic theory, we study the electromagnetic response in the superconducting states. Both superfluid and normal-fluid dynamics are involved. We predict that the normal fluid is present only when the excited superconducting velocity v_s is larger than a threshold $v_L = |\Delta|/k_F$. Interestingly, with the normal fluid, we find that there exists friction between the normal-fluid and superfluid currents. Due to this friction, part of the superfluid becomes viscous. Therefore a three-fluid model, normal fluid and nonviscous and viscous superfluids, is proposed. For the stationary magnetic response, at $v_s < v_L$ with only the nonviscous superfluid, the Meissner supercurrent is excited and the gap equation can be reduced to the Ginzburg-Landau equation. At $v_s \geqslant v_L$, with the normal fluid and nonviscous and viscous superfluids, in addition to the directly excited Meissner supercurrent in the superfluid, a normal-fluid current is also induced through the friction drag with the viscous superfluid current. Due to the normal-fluid and viscous-superfluid currents, the penetration depth is influenced by the scattering effect. In addition, a modified Ginzburg-Landau equation is proposed. We predict an exotic phase in which both the resistivity and superconducting gap are *finite*. As for the optical response, the excited v_s oscillates with time. When $v_s < v_L$, only the nonviscous superfluid is present, whereas at $v_s \geqslant v_L$, normal fluid and nonviscous and viscous superfluids are present. We show that the excited normal-fluid current exhibits the Drude-model behavior, while the superfluid current consists of the Meissner supercurrent and Bogoliubov quasiparticle current. Due to the friction between the superfluid and normal-fluid currents, the optical conductivity is captured by the three-fluid model. Finally, we also study the optical excitation of the Higgs mode. By comparing the contributions from the drive and Anderson-pseudospin pump effects, we find that the drive effect is dominant at finite temperature whereas at zero temperature, both effects contribute.

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I. INTRODUCTION

In the field of superconductivity, electromagnetic responses have been attracting intensive attention in the past few decades for revealing the physics of superconductivity and exploring the novel properties [1–9]. For the stationary magnetic response, the induced diamagnetic supercurrent and the resulting magnetic-flux expulsion are known to be one of the fundamental phenomena in superconductors, referred to as the Meissner effect [10,11]. Analysis of the magnetic response in the early-stage works is based on the well-known Ginzburg-Landau phenomenological theory for pure superconductors [12]. As for the optical studies in superconductors, efforts are focused on the microwave and terahertz (THz) absorptions in both linear [13–21] and nonlinear [22–31] regimes. Particularly, a phenomenological picture based on the two-fluid model, which was first proposed by Tisza and London [32] and then developed by Landau [33] in bosonic liquid helium II, is widely used to capture the physics of the optical response in superconductors [1,2,5,18,19,21–23,34]. It is postulated that both the normal fluid and superfluid are present as separate fluids, each with its own density and velocity in the superconducting state. The normal fluid in the optical response exhibits the Drude-model behavior [1,2,5,18,19,21–23]. A superfluid on the other hand has no resistivity [1,2,5,18,19,21–23]. Recently, it was experimentally realized that through the intense terahertz (THz) field, one can excite the fluctuation of the superfluid density with the oscillation frequency at twice the optical frequency [26,28–31]. This oscillation so far is attributed to the excited Higgs mode, i.e., the fluctuations of the magnitude of the superconducting order parameter [35–47]. In most situations, a plateau of the superconducting order parameter is discovered after the THz pulse [28,29].

Within the framework of superconductivity theory established by Bardeen, Cooper, and Schrieffer (BCS) [48], microscopic theories of the above electromagnetic properties of superconductors have been developed for more than five decades [37,38,40–45,47,49–62]. In principle, a complete theory to calculate the electromagnetic properties must satisfy certain conditions. First, it should be capable of calculating both magnetic and optical responses in linear and nonlinear regimes. Second, it should include the scattering effect, which is inevitable in dirty superconducting metals [40,41]. Finally, it should satisfy the gauge invariance in superconductors [63–65], first revealed by Nambu [63,65] based on a gauge structure of vector potential **A**, scalar potential ϕ, and superconducting phase ψ. However, to the best of our knowledge,

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a microscopic theoretical description that satisfies all three conditions above is still absent in the literature.

Specifically, the electromagnetic properties of conventional superconductors were first discussed by Mattis and Bardeen (MB) within the BCS theory in the linear regime and dirty limit [49]. Based on the MB theory, Miller gave a dependence of the penetration depth δ on the mean free path l in the case of a stationary magnetic response [50]. This dependence was extended by Tinkham to the regime between clean and dirty limits later as $\delta = \delta_c\sqrt{1 + \xi_0/l}$ at low temperature [2] (ξ_0 and δ_c denote the coherence length and clean-limit penetration depth, respectively), in good agreement with the experiments [66–70]. This directly indicates that the Meissner supercurrent experiences a friction resistance from scattering. Nevertheless, a supercurrent should be nonviscous. The physical origin of the friction resistance on a supercurrent is still unclear in the literature, since the scattering effect in the early-stage works [2,49] is included through a hand-waving discussion and hence the microscopic scattering process is absent. As for the optical response, MB theory reveals that the optical absorption is realized by breaking the Cooper pairs into quasielectrons and quasiholes when the optical frequency is larger than twice the superconducting-gap magnitude [1,49,59]. In this regime, the MB theory successfully describes the experimentally observed complex conductivity [15–17,19,28]. However, at low frequency, it deviates from the experimental observation [15,17,28]. In addition to this deficiency, it is hard to extend the MB theory into the nonlinear regime, and hence, the excitation of Higgs mode is absent in this description. Most importantly, as an early-stage work, the MB theory [49], established in a specific gauge with finite vector potential alone, is not gauge invariant.

Theories for the excitation of the Higgs mode in superconductors are mostly based on the Liouville [37,38,41] or Bloch [40,42–45,47] equation derived in the Anderson pseudospin representation [71]. In these theories, the nonlinear term $\mathbf{A}^2$ is included, which leads to the pump of the quasiparticle correlation (pump effect) and then contributes to the excitation of the Higgs mode. However, no drive effect (linear term) is included in this description. Thus unphysical conclusions are immediately obtained. On one hand, no optical current is excited. On the other hand, the elastic scattering is ineffective since the pump effect alone is isotropic in the momentum space. Consequently, the Liouville [37,38,41] or Bloch [40,42–45,47] equation in the literature is insufficient to elucidate the complete physics. Moreover, with only a finite vector potential [37,38,40–45,47], the gauge invariance is also unsatisfied.

To date, the most effective method of calculating the electromagnetic properties in superconductors is provided by Gorkov's equation of Green function [51–54,56] and its derivatives. Specifically, in the Gorkov equation, the gauge invariance is satisfied. For the stationary magnetic response, it is demonstrated that the Gorkov equation can reduce to the Ginzburg-Landau theory [53,54]. Moreover, by calculating the scattering self-energy by assuming that the scattering in superconductors is the same as that in normal metals, the disorder effect on the penetration depth is discussed by Abrikosov and Gorkov [51], in consistency with the MB theory [50]. As for the optical case, it is reported that in appropriate limits, the obtained optical conductivity from the Gorkov equation can reduce to the MB theory in the dirty limit [55] and exhibits the two-fluid-model behavior in the weak scattering [52]. However, the Gorkov equation [54,56] actually is very hard to handle for a kinetic calculation of the temporal evolution or spatial diffusion in superconducting systems as too many variables are involved. The complex calculation also makes it difficult to explore the microscopic process and physical picture of both the electromagnetic properties and scattering effect.

To reduce the number of variables, two kinds of the transformations of Gorkov equation into the transportlike equation are developed in the literature. Specifically, based on Gorkov's equation, via τ_3-Green function $[G(x,x') = -i\tau_3\langle T\Psi(x)\Psi^\dagger(x')\rangle$ with $\Psi(x)$ being the Nambu-space field operator [54,56], $x = (t,\mathbf{r})$ denoting the time-space point, T being the chronological ordering [54], and $\langle\ldots\rangle$ representing the ensemble average, in the quasiclassical approximation [6,7,72] with an integration over the energy variable [72], Eilenberger derived a transport-like equation [57], which can reduce to Ginzburg-Landau equation near the critical temperature [73]. However, the Gauge invariance is lost during this derivation. It is fixed years later [74,75] by constructing the gauge-invariant τ_3-Green function by introducing the Wilson line [76]. The Eilenberger equation successfully describes topics like the Josephson effect in multilayer junctions [77–79], unconventional superconductivity [80–83], vortex behaviors [84–87], and disorder influence on superconductivity [88–91]. Particularly, for the stationary case in the dirty limit, the Eilenberger equation is further simplified into a diffusive Usadel equation [58], which is widely used to investigate the superconducting proximity effects in multilayered structures [8,9,92–97]. However, the specific scattering term in the Eilenberger equation is very hard to handle due to the relative-time (i.e., frequency) variable. Thus the relaxation-time approximation is usually taken. Therefore the microscopic process and physical picture of the scattering effect are lacking. Moreover, the relative-time variable also markedly enlarges the difficulty for the temporal evolution. Consequently, it is hard to apply the Eilenberger equation in the optical study.

Actually, in the optics [98] and spintronics [99] of semiconductors, to obtain the kinetic equation, a complete nonequilibrium approach with reduced relative-time variable by taking the equal-time approximation, has been well established. Similarly, considering the fact that the superconductivity in conventional superconductors is characterized by equal-time pairing [48], Yu and Wu proposed another transformation of the Gorkov equation into a transportlike equation in superconducting states through τ_0-Green function $[G(x,x') = -i\langle T\Psi(x)\Psi^\dagger(x')\rangle]$ [60]. Moreover, to retain the gauge invariance, a gauge-invariant τ_0-Green function [60] is constructed by introducing the Wilson line [76]. Then, a gauge-invariant kinetic equation is proposed. Thanks to the reduced relative-time variable, this equation is much easier to handle for the temporal evolution and hence the optical response in superconductors. Moreover, due to its gauge invariance, both the drive and pump effects mentioned above are kept. Particularly, it is revealed that the drive effect makes a dominant contribution in the Higgs-mode excitation [60], in sharp contrast to the conclusion by Liouville [37,38,41] or Bloch [40,42–45,47] equation in which only the pump effect is considered. Most importantly, the complete microscopic scattering process is constructed in this gauge-invariant theory, and the rich physics of the relaxation mechanism [60] and transport phenomena [62] is revealed. The experimentally observed plateau of the superconducting gap after the THz pulse [28,29] is also revealed as the consequence of the scattering effect [60]. However, in spite of the success in optical studies, as a gauge-invariant work for the electromagnetic response, this theory fails to apply to the magnetic case since it is incapable of giving the Meissner current and reducing to the Ginzburg-Landau theory. Therefore it is natural to conclude that this theory only describes the dynamics of quasiparticles [60,62]. The dynamics of superfluid is not directly involved in this description, but circumvented through the response of the gap in the Bogoliubov quasiparticle excitation.

In this work, we extend the kinetic theory by Yu and Wu [60] to include the superfluid, so that both normal-fluid and superfluid dynamics are involved in the theory. As a gauge-invariant theory for the electromagnetic response, our kinetic equation can be applied to study both the magnetic and optical cases. We first focus on the weak-scattering case in the present work. Rich physics is revealed. Specifically, in the electromagnetic response, we show that the superconducting velocity v_s is always excited. Particularly, a threshold $v_L = |\Delta|/k_F$ (Δ and k_F denote the superconducting order parameter and Fermi momentum, respectively) of superconducting velocity for the emergence of the normal fluid and hence the scattering is predicted from our theory, i.e., the normal fluid is excited only when $v_s > v_L$. Actually, similar threshold for the emergence of the normal fluid and scattering was first proposed by Landau to interpret the fluid viscosity in bosonic liquid helium II at large velocity [33]. Therefore we refer to this threshold as Landau threshold. Interestingly, we find that there also exists friction between the normal-fluid and superfluid currents. Due to this friction, part of the superfluid becomes viscous. Therefore the superfluid consists of the nonviscous superfluid and viscous one. Consequently, to capture the physics of the electromagnetic response in superconducting states, a three-fluid model at $v_s \geqslant v_L$ is proposed from our theory: normal fluid and nonviscous and viscous superfluids.

The physics behind these predictions can be understood as follows. It is established [100–105] that with a superconducting velocity, the quasiparticle energy spectrum is tilted as $E^{\pm}_{\mathbf{k}} = \mathbf{k}\cdot\mathbf{v}_s \pm E_k$ with $E^{+}_{\mathbf{k}}$ ($E^{-}_{\mathbf{k}}$) standing for the quasielectron (quasihole) energy and E_k being the BCS Bogoliubov quasiparticle energy. At a small superconducting velocity, the superconducting state behaves like the BCS state, in which all particles in the spherical shell by the BCS theory participate in the pairing. Thus there only exists superfluid. As for the case with a large superconducting velocity at $v_s \geqslant v_L$, in addition to the pairing (P) region with $|\mathbf{k}\cdot\mathbf{v}_s| < E_k$, there also exists the region with $|\mathbf{k}\cdot\mathbf{v}_s| > E_k$, in which the quasielectron energy $E^{+}_{\mathbf{k}}$ is smaller than zero or the quasihole energy $E^{-}_{\mathbf{k}}$ is larger than zero. As revealed in the previous works [103–106], the anomalous correlation in this region is

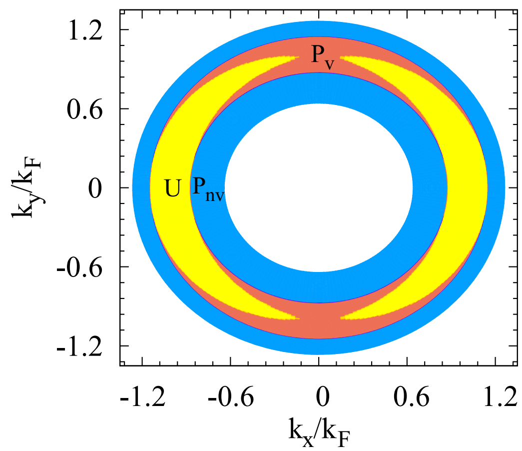

FIG. 1. Schematic showing the division in the momentum space when the superconducting velocity v_s is larger than the Landau threshold v_L. In the figure, the spherical shell by the BCS theory is divided into three parts: unpairing (U) region characterized by $|\mathbf{k}\cdot\mathbf{v}_s| > E_k$, denoted by yellow regions; nonviscous pairing (P_{nv}) region characterized by $kv_s < E_k$, denoted by orange regions; viscous pairing (P_v) region characterized by $kv_s > E_k$ and $|\mathbf{k}\cdot\mathbf{v}_s| < E_k$, denoted by blue regions.

destroyed. Thus the particles in this region no longer participate in the pairing and behave like normal particles. Following the terminology in the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state [106,107], this region is referred to as the unpairing (U) region. Then, both the normal fluid (from U region) and superfluid (from P region) are present. Particularly, as shown in Fig. 1, there exists a special region (P_v region characterized by $kv_s > E_k$ and $|\mathbf{k}\cdot\mathbf{v}_s| < E_k$) in the pairing region which shares the same momentum magnitude with U region. In conventional superconducting metals, due to the strong screening, the impurity scattering behaves as the short-range impurity scattering, which is isotropic in the momentum space. Therefore the particles in P_v region participate in the pairing but experience the scattering with those in U region, leading to the friction between the superfluid and normal-fluid currents. Consequently, the superfluid in P_v region becomes viscous. Whereas the superfluid in the remaining pairing region (P_{nv} region characterized by $kv_s < E_k$ shown in Fig. 1) is still nonviscous.

For the stationary magnetic response, when $v_s < v_L$, only superfluid is present. In this situation, we prove that the excited superfluid current is the Meissner supercurrent, and near the critical temperature, our gap equation reduces to the Ginzburg-Landau equation [12]. As for $v_s \geqslant v_L$, there exist normal fluid (from U region) and nonviscous (from P_{nv} region) and viscous (from P_v region) superfluids. The magnetic response is captured by the three-fluid model proposed above. Specifically, differing from the excited Meissner supercurrent in the superfluid, no current is directly excited from the magnetic flux in the normal fluid as it should be. Nevertheless, the normal-fluid current can be induced through the above mentioned friction drag with superfluid current. Moreover, due to this friction, the superfluid current is separated into the nonviscous and viscous ones. Consequently, thanks to the viscosity in superfluid current and presence of the normal fluid current, the penetration depth is influenced by the scattering. By only considering the viscous superfluid, the dependence of the penetration depth on the mean free path from our theory is exactly the same as that from Tinkham's discussion [2]. Nevertheless, since there also exist normal fluid and nonviscous superfluid, an extension of the penetration depth is revealed. In addition, at $v_s \geqslant v_L$, we also propose a modified Ginzburg-Landau equation, in which the calculation of the phenomenological parameters is restricted to the pairing region. Finally, at $v_s > \omega_D/k_F$ (ω_D denotes the Debye frequency), before the superconducting gap is destroyed, we predict an exotic phase in which the nonviscous superfluid vanishes, leaving only the viscous superfluid and normal fluid. Thus, interestingly, this phase shows finite resistivity but with a *finite* superconducting gap.

As for the optical response, the excited superconducting velocity v_s oscillates with time. When $v_s < v_L$, only the nonviscous superfluid is present, whereas at $v_s \geqslant v_L$, there exist normal fluid (from U region) and nonviscous (from P_{nv} region) and viscous (from P_v region) superfluids. We show that in the optical response, the normal-fluid current exhibits the Drude-model behavior as it should be. Whereas in the superfluid, we find that the superfluid current is excited and it consists of the Meissner supercurrent, which has the same form as that in the magnetic response, as well as the Bogoliubov quasiparticle current. At low temperature, few Bogoliubov quasiparticles are excited in the pairing region and hence the Bogoliubov quasiparticle current is marginal. In this case, the normal-fluid current and the superfluid current, which only consists of the Meissner supercurrent, are exactly the same as those in the original two-fluid model [1,2,5,18,19,21–23,34]. However, friction between the superfluid and normal-fluid currents is present. Due to this friction, the superfluid is separated into the nonviscous and viscous ones. This suggests that the optical response is also captured by the three-fluid model above. Then, based on this three-fluid model, an expression of the optical conductivity is revealed. Furthermore, we also give the expression of the optical excitation of the Higgs mode. A comparison between the contributions from the drive and Anderson pseudospin pump effects mentioned above is addressed. We point out that the previous conclusion by Yu and Wu [60] that the drive effect is dominant only holds at finite temperature, whereas at zero temperature, both effects contribute.

This paper is organized as follows. In Sec. II, we introduce our model and construct the gauge-invariant kinetic theory of the electromagnetic response in superconducting states. We derive the three-fluid model and perform the analytical analysis of the magnetic and optical responses in Sec. III. We summarize and discuss in Sec. IV.

II. MODEL

In this section, we first set up the Hamiltonian for the conventional superconducting states and present the gauge structure revealed by Nambu [63,65]. Then, we extend the previous theory by Yu and Wu [60] and present a gauge-invariant microscopic kinetic equation of the electromagnetic response in superconducting states.

A. Hamiltonian

The free Bogoliubov-de Gennes (BdG) Hamiltonian of the s-wave superconducting state reads

$$H = \int \frac{d\mathbf{r}}{2}\Psi^\dagger(x)\{[\xi_{\mathbf{p}-e\mathbf{A}(x)\tau_3} + e\phi(x)]\tau_3 + \hat{\Delta}(x)\}\Psi(x), \quad (1)$$

with

$$\hat{\Delta}(x) = |\Delta|[e^{i\psi(x)}\tau_+ + e^{-i\psi(x)}\tau_-]. \quad (2)$$

Here, the Nambu-space field operator reads $\Psi(x) = (\Psi_\uparrow(x), \Psi_\downarrow^\dagger(x))^T$; $\xi_\mathbf{p} = \varepsilon_\mathbf{p} - \mu$ and $\varepsilon_\mathbf{p} = \frac{\mathbf{p}^2}{2m}$ with m and μ being the effective mass and chemical potential; $\mathbf{p} = -i\hbar\nabla$; τ_i are the Pauli matrices in particle-hole spaces. In the present work, we consider a magnetic flux in the magnetic response of superconductors, and hence, the Zeeman effect of the magnetic field is neglected.

It is first revealed by Nambu that under a gauge transformation $\Psi(x)\to e^{i\tau_3\chi(x)}\Psi(x)$, to restore the gauge invariance of the BdG Hamiltonian [Eq. (1)], the vector potential $\mathbf{A}$, scalar potential ϕ, and superconducting phase ψ must transform as [63,65]

$$eA_\mu \to eA_\mu - \partial_\mu\chi(x), \quad (3)$$

$$\psi(x) \to \psi(x) + 2\chi(x), \quad (4)$$

where the four vectors are $A_\mu = (\phi, \mathbf{A})$ and $\partial_\mu = (\partial_t, -\nabla)$.

B. Kinetic equation

Following the previous work by Yu and Wu [60], we derive the gauge-invariant microscopic kinetic equation of the electromagnetic response in superconducting states in the presence of the electron-electron, electron-phonon, and electron-impurity scatterings.

1. Derivation of the free kinetic equation

We first present the derivation of the free kinetic equation in the absence of the electron-electron, electron-phonon, and electron-impurity interactions.

We begin with the lesser τ_0-Green function $G^<_{x_1x_2} = i\langle\Psi^\dagger(x_2)\Psi(x_1)\rangle$ [60]. The Gorkov equations of the lesser τ_0-Green function $G^<_{x_1x_2}$ read [54,98,103]

$$(i\overrightarrow{\partial}_{t_1} - \overrightarrow{H}_{\mathbf{p}_1,x_1})G^<_{x_1x_2} = 0, \quad (5)$$

$$G^<_{x_1x_2}(-i\overleftarrow{\partial}_{t_2} - \overleftarrow{H}_{\mathbf{p}_2,x_2}) = 0. \quad (6)$$

The gauge structure of the lesser τ_0-Green function is given by $G^<_{x_1x_2} \to e^{i\tau_3\chi(x_1)}G^<_{x_1x_2}e^{-i\tau_3\chi(x_2)}$ after a gauge transformation $\Psi(x)\to e^{i\tau_3\chi(x)}\Psi(x)$. As in the kinetic equation, only the center-of-mass coordinate $R = (T, \mathbf{R}) = (x_1 + x_2)/2$ is retained. It is hard to retain the gauge invariance in the kinetic equation derived from $G^<_{x_1x_2}$. To fix this, following the previous works [60,74,75], by introducing the Wilson line [76], the gauge-invariant Green function is constructed: $G^{g<}_{x_1x_2} = e^{-iW^R_{x_1}}G^<_{x_1x_2}e^{-iW^{x_2}_R}$. Here, $W^y_x = \mathrm{P}\int_x^y dx^\mu eA_\mu\tau_3$ with $dx^\mu = (dt, -d\mathbf{r})$. "P" indicates that the integral is path dependent. Then, after the gauge transformation $\Psi(x)\to e^{i\tau_3\chi(x)}\Psi(x)$,

$G^{g<}_{x_1x_2}$ transforms as $G^{g<}_{x_1x_2} \to e^{i\tau_3\chi(R)}G^{g<}_{x_1x_2}e^{-i\tau_3\chi(R)}$, in which only the center-of-mass coordinate is relevant.

By taking the difference of Eqs. (5) and (6) and replacing $G^{<}_{x_1x_2}$ with $G^{g<}_{x_1x_2}$, one has

$$i\widetilde{\partial_T}G^{g<}_{x_1x_2} - [e\phi(x_1)\tau_3 G^{g<}_{x_1x_2} - G^{g<}_{x_1x_2}e\phi(x_2)\tau_3] - [\tau_3\widetilde{\xi}_{\overrightarrow{\mathbf{p}}_1-e\mathbf{A}(x_1)\tau_3}G^{g<}_{x_1x_2} - G^{g<}_{x_1x_2}\widetilde{\xi}_{\overleftarrow{\mathbf{p}}_2-e\mathbf{A}(x_2)\tau_3}\tau_3] - [e^{-2iW^R_{x_1}}\hat{\Delta}(x_1)G^{g<}_{x_1x_2} - G^{g<}_{x_1x_2}\hat{\Delta}(x_2)e^{-2iW^{x_2}_R}] = 0, \quad (7)$$

in which $\widetilde{X}G^{g<}_{x_1x_2} = e^{-iW^R_{x_1}}[X(e^{iW^R_{x_1}}G^{g<}_{x_1x_2}e^{iW^{x_2}_R})]e^{-iW^{x_2}_R}$ and $G^{g<}_{x_1x_2}\widetilde{X} = e^{-iW^R_{x_1}}[(e^{iW^R_{x_1}}G^{g<}_{x_1x_2}e^{iW^{x_2}_R})X]e^{-iW^{x_2}_R}$. Then, via taking the path in the Wilson line to be the straight line [60,74,75] and defining the relative coordinate $r=(t,\mathbf{r})=x_1-x_2$, through the gradient expansion [98,99], by taking equal time, i.e., $t=0$ [60,98,99,103], the gauge-invariant kinetic equation of the density matrix $\rho_{\mathbf{k}}(\mathbf{R},T) = -iG^{g<}(\mathbf{R},T,\mathbf{k},t=0) = -i\int d\mathbf{r}e^{-i\mathbf{k}\cdot\mathbf{r}}G^{g<}(\mathbf{R},T,\mathbf{r},t=0)$ is obtained from Eq. (7).

It is pointed out that in the previous work by Yu and Wu [60], except the zeroth order, the higher-order gradient expansion on the last term on the left-hand side of Eq. (7), i.e., the superconducting order parameter $\hat{\Delta}$ accompanied by the Wilson line, is neglected by considering a fixed order parameter in semiconductor quantum wells from the proximity effect. This approximation is sublated in our work, considering the fluctuation of W and Δ in time and space in the electromagnetic response. To apply the higher-order gradient expansion on this term, we approximately take $e^{-2iW} \approx 1-2iW-2W^2$. This approximation is based on the fact that in conventional superconductors, the vector potential is much smaller than the Fermi momentum. Therefore, since one has $W \propto (\mathbf{A}\cdot\mathbf{r})$ after taking equal time, W can be treated as a small quantity.

Finally, the new gauge-invariant microscopic kinetic equation of the electromagnetic response in the superconducting states is written as

$$\partial_T\rho_{\mathbf{k}} + i[(\xi_k+e\phi)\tau_3 + \hat{\Delta}(\mathbf{R}),\rho_{\mathbf{k}}] + i\left[\frac{e^2A^2}{2m}\tau_3,\rho_{\mathbf{k}}\right] + \frac{1}{2}\{e\mathbf{E}\tau_3,\partial_{\mathbf{k}}\rho_{\mathbf{k}}\} + \left\{\frac{\mathbf{k}}{2m}\tau_3,\nabla_{\mathbf{R}}\rho_{\mathbf{k}}\right\} - \left[\frac{i}{8m}\tau_3,\nabla^2_{\mathbf{R}}\rho_{\mathbf{k}}\right] - \frac{1}{2}\{(\nabla-2ie\mathbf{A}\tau_3)\hat{\Delta}(\mathbf{R}),\partial_{\mathbf{k}}\rho_{\mathbf{k}}\} - \frac{i}{8}[(\nabla-2ie\mathbf{A}\tau_3)(\nabla-2ie\mathbf{A}\tau_3)\hat{\Delta}(\mathbf{R}),\partial_{\mathbf{k}}\partial_{\mathbf{k}}\rho_{\mathbf{k}}] - \left[\frac{e\mathbf{A}}{2m}\tau_3,\tau_3\nabla_{\mathbf{R}}\rho_{\mathbf{k}}\right] - \left[\frac{e\nabla_{\mathbf{R}}\cdot\mathbf{A}}{4m}\tau_3,\tau_3\rho_{\mathbf{k}}\right] = \partial_t\rho_{\mathbf{k}}|_{\mathrm{sc}}. \quad (8)$$

Here, $[A,B]=AB-BA$ and $\{A,B\}=AB+BA$ represent the commutator and anticommutator, respectively; $\mathbf{E} = -\nabla_{\mathbf{R}}\phi - \partial_T\mathbf{A}$ denotes the electric field. It is noted that on the right-hand side of Eq. (8), the scattering term $\partial_t\rho_{\mathbf{k}}|_{\mathrm{sc}}$ is added for completeness, whose explicit expression is given in the next section.

In Eq. (8), on the left-hand side, the second term represents the coherent term contributed by the BCS Hamiltonian. The third and fourth terms denote the pump and drive effect mentioned in the introduction, as addressed in the previous work by Yu and Wu [60]. The fifth and sixth terms stand for the diffusion terms. The seventh and eighth terms, which behave like the drive effect, are absent in Ref. [60]. They come from the higher-order gradient expansion of the superconducting order parameter accompanied with the Wilson line mentioned above. In the following section, it is shown that these two terms provide the kinetic-energy terms in the Ginzburg-Landau equation. Particularly, it is noted that with the gauge structure revealed by Nambu [Eqs. (3) and (4)] [63], Eq. (8) is gauge invariant after the gauge transformation $\rho_{\mathbf{k}}(R) \to e^{i\tau_3\chi(R)}\rho_{\mathbf{k}}(R)e^{-i\tau_3\chi(R)}$.

The order parameter is self-consistently determined by the gap equation:

$$\Delta(\mathbf{R}) = -V{\sum_{\mathbf{k}}}'\mathrm{Tr}[\rho_{\mathbf{k}}(\mathbf{R})\tau_-], \quad (9)$$

where V is the conventional s-wave attractive potential. $\sum_{\mathbf{k}}'$ here and in the following shows the summation is restricted in the spherical shell by the BCS theory [48].

The gauge invariant current is obtained by performing the Wilson line [76] technique on the current [54,56]

$$\mathbf{j} = -\frac{ie}{2m}\mathrm{Tr}[(i\nabla_{x'} - i\nabla_x)G^<_{x,x'} - 2e\mathbf{A}\tau_3G^<_{x,x'}]_{x'\to x+0^+} \quad (10)$$

and reads

$$\mathbf{j} = -\frac{ie}{2m}\mathrm{Tr}[-2i\partial_{\mathbf{r}}G^{g<}_{x,x'}]_{x'\to x+0^+} = \sum_{\mathbf{k}}\mathrm{Tr}\left[\frac{e\mathbf{k}}{m}\rho_{\mathbf{k}}\right]. \quad (11)$$

2. Derivation of scattering

We next present the scattering terms $\partial_t\rho_{\mathbf{k}}|_{\mathrm{sc}}$ in Eq. (8) due to the electron-electron Coulomb, electron-phonon, and electron-impurity scatterings. The scattering terms are derived based on the generalized Kadanoff-Baym (GKB) ansatz [98,99,103,108].

The specific scattering terms of the electron-electron Coulomb, electron-phonon, and electron-impurity interactions are written as (the detailed derivation of the scattering terms can be found in the previous works [60,99])

$$\partial_t\rho_{\mathbf{k}}|_{\mathrm{sc}} = -\pi\sum_{\mathbf{k}'}\sum_{\eta_1\eta_2}[S^{\eta_1\eta_2}_{\mathbf{k}\mathbf{k}'}(>,<) - S^{\eta_1\eta_2}_{\mathbf{k}\mathbf{k}'}(<,>) + \mathrm{H.c.}], \quad (12)$$

with

$$S^{\eta_1\eta_2}_{\mathbf{k}\mathbf{k}'}|_{\mathrm{ei}} = n_i|V_{\mathbf{k}\mathbf{k}'}|^2\delta(E^{\eta_1}_{\mathbf{k}'} - E^{\eta_2}_{\mathbf{k}})[\tau_3\rho^>_{\mathbf{k}'}\Gamma^{\eta_1}_{\mathbf{k}'}\tau_3\Gamma^{\eta_2}_{\mathbf{k}}\rho^<_{\mathbf{k}}], \quad (13)$$

$$S^{\eta_1\eta_2}_{\mathbf{k}\mathbf{k}'}|_{\mathrm{ep}} = |g^{\gamma_p}_{\mathbf{k}\mathbf{k}'}|^2[n^>_{\mathbf{k}-\mathbf{k}'}\delta(E^{\eta_1}_{\mathbf{k}'} - E^{\eta_2}_{\mathbf{k}} + \omega^{\gamma_p}_{\mathbf{k}-\mathbf{k}'}) + n^<_{\mathbf{k}-\mathbf{k}'} \times\delta(E^{\eta_1}_{\mathbf{k}'} - E^{\eta_2}_{\mathbf{k}} - \omega^{\gamma_p}_{\mathbf{k}-\mathbf{k}'})][\tau_3\rho^>_{\mathbf{k}'}\Gamma^{\eta_1}_{\mathbf{k}'}\tau_3\Gamma^{\eta_2}_{\mathbf{k}}\rho^<_{\mathbf{k}}], \quad (14)$$

$$S^{\eta_1\eta_2}_{\mathbf{k}\mathbf{k}'}|_{\mathrm{ee}} = \sum_{\mathbf{q}}\sum_{\eta_3\eta_4}|V_{\mathbf{q}}|^2\delta(E^{\eta_1}_{\mathbf{k}-\mathbf{q}} - E^{\eta_2}_{\mathbf{k}} + E^{\eta_3}_{\mathbf{k}'+\mathbf{q}} - E^{\eta_4}_{\mathbf{k}'}) \times[\tau_3\rho^>_{\mathbf{k}-\mathbf{q}}\Gamma^{\eta_1}_{\mathbf{k}-\mathbf{q}}\tau_3\Gamma^{\eta_2}_{\mathbf{k}}\rho^<_{\mathbf{k}}]\mathrm{Tr}[\rho^>_{\mathbf{k}'+\mathbf{q}}\Gamma^{\eta_3}_{\mathbf{k}'+\mathbf{q}}\Gamma^{\eta_4}_{\mathbf{k}'}\rho^<_{\mathbf{k}'}]. \quad (15)$$

Here, $\eta = \pm$; $\Gamma^{\pm}_{\mathbf{k}}$ represent the projection operators; n_i is the impurity density; $V_{\mathbf{q}}$ denotes the screened Coulomb potential; $g^{\gamma_p}_{\mathbf{kk'}}$ stands for the electron-phonon interaction, and $\omega^{\gamma_p}_{\mathbf{q}}$ represents the phonon energy with γ_p being the corresponding phonon branch; $\rho^{<}_{\mathbf{k}} = \rho_{\mathbf{k}}$ and $\rho^{>}_{\mathbf{k}} = 1 - \rho_{\mathbf{k}}$; $n^{>}_{\mathbf{k}} = 1 + n_{\mathbf{k}}$ and $n^{<}_{\mathbf{k}} = n_{\mathbf{k}}$ with $n_{\mathbf{k}}$ being the phonon distribution function.

As mentioned in the introduction, it is established [100–105] that with the superconducting velocity $\mathbf{v}_s$, the quasiparticle energy is tilted as $E^{\pm}_{\mathbf{k}} = \mathbf{k}\cdot\mathbf{v}_s \pm E_k$ with $E_k = \sqrt{\xi_k^2 + |\Delta|^2}$. In this situation, the projection operators are written as $\Gamma^{\pm}_{\mathbf{k}} = U_k^{\dagger} Q^{\pm} U_k$ with $Q^{\pm} = (1 \pm \tau_3)/2$. $U_k = u_k\tau_0 - v_k\tau_+ + v_k\tau_-$ represents the unitary transformation matrix from the particle space to the quasiparticle one with $u_k = \sqrt{1/2 + \xi_k/(2E_k)}$ and $v_k = \sqrt{1/2 - \xi_k/(2E_k)}$. It is noted that the effect of the superconducting velocity on the scattering process is neglected in Ref. [60] by taking the quasiparticle energies as the BCS ones (i.e., $E^{\pm}_{\mathbf{k}} = \pm E_k$).

III. ANALYTICAL ANALYSIS

In this part, with the new gauge-invariant microscopic kinetic equation [Eq. (8)] in Sec. II B, we analytically investigate the electromagnetic properties of superconductors including the magnetic and optical responses in the linear and nonlinear regimes in the weak-scattering limit.

A. Weak scattering

We first simplify the scattering terms by transforming the scattering terms into the quasiparticle space (i.e., $\partial_t \rho_{\mathbf{k}}|_{\rm sc} = U_k \partial_t \rho^q_{\mathbf{k}}|_{\rm sc} U_k^{\dagger}$). Considering the fact that the electron-phonon scattering is weak at low temperature, we mainly consider the electron-impurity scattering, which reads

$$\partial_t \rho^q_{\mathbf{k}}\big|_{\rm sc} = -n_i\pi \sum_{\mathbf{k}'} |V_{\mathbf{kk'}}|^2 U_k^{\dagger}\tau_3 U_{k'}\big\{Y_{kk'}\big(\rho^q_{\mathbf{k}} - \rho^q_{\mathbf{k}'}\big) - \big[\rho^q_{\mathbf{k}'}, Y_{kk'}\big]\big\} + \text{H.c.}. \quad (16)$$

Here, $Y_{kk'} = \sum_{\eta_1\eta_2} Q^{\eta_1} U^{\dagger}_{k'}\tau_3 U_k Q^{\eta_2}\delta(E^{\eta_1}_{\mathbf{k}'} - E^{\eta_2}_{\mathbf{k}})$.

In the present work, we consider a weak-scattering limit. In this situation, the scattering only causes the momentum (current) relaxation. Therefore one only needs to keep the leading contribution in the scattering terms, i.e., the diagonal terms in $\rho^q_{\mathbf{k}}$ (quasiparticle distribution) and $\partial_t \rho^q_{\mathbf{k}}|_{\rm sc}$ (scattering of the quasiparticle distribution), and Eq. (16) becomes

$$\partial_t \rho^q_{\mathbf{k}}\big|_{\rm sc} = -n_i\pi \sum_{\mathbf{k}'} |V_{\mathbf{kk'}}|^2 \Bigg\{(1-\eta_{kk'})\begin{pmatrix} \big(\rho^q_{\mathbf{k},11} - \rho^q_{\mathbf{k}',11}\big)\delta(E^+_{\mathbf{k}'} - E^+_{\mathbf{k}}) & 0 \\ 0 & \big(\rho^q_{\mathbf{k},22} - \rho^q_{\mathbf{k}',22}\big)\delta(E^-_{\mathbf{k}'} - E^-_{\mathbf{k}}) \end{pmatrix} + (1+\eta_{kk'})\begin{pmatrix} \big(\rho^q_{\mathbf{k},11} - \rho^q_{\mathbf{k}',22}\big)\delta(E^-_{\mathbf{k}'} - E^+_{\mathbf{k}}) & 0 \\ 0 & \big(\rho^q_{\mathbf{k},22} - \rho^q_{\mathbf{k}',11}\big)\delta(E^+_{\mathbf{k}'} - E^-_{\mathbf{k}}) \end{pmatrix}\Bigg\}, \quad (17)$$

where $\eta_{kk'} = (|\Delta|^2 - \xi_k\xi_{k'})/(E_k E_{k'})$.

On the right-hand side of Eq. (17), the first term denotes the intra-quasi-electron-band and intra-quasi-hole-band scatterings. The second term represents the interband scattering between the quasielectrons and quasiholes. Actually, as shown in Fig. 2(a), in the absence of the superconducting velocity, the interband scattering between the quasielectrons and quasiholes is forbidden by the energy conservation thanks to the BCS gap. Only the intraband scatterings exist. Nevertheless, as mentioned above, with a large excited superconducting velocity ($kv_s > E_k$) in the electromagnetic response [60,62], the quasiparticle energy spectrum is tilted [100–105]. Then, as shown in Fig. 2(b), the interband scattering between the quasielectrons and quasiholes is turned on. However, this unique scattering has long been overlooked in the literature.

In conventional superconducting metals, due to the strong screening, one can take the impurity scattering as the short-range one, i.e., $|V_{\mathbf{kk'}}|^2 \approx |V_0|^2$. Moreover, thanks to the large Fermi energy, we approximately take the emergence of the scattering around the Fermi surface by setting $|\xi_k|, |\xi_{k'}| < E_c$ in Eq. (17). E_c is the cutoff energy. Then, after the integration over the angle, Eq. (17) approximately becomes (refer to Appendix A)

$$\partial_t \rho^q_{\mathbf{k}}\big|_{\rm scat} = -\frac{1}{\tau_k}\Bigg[\frac{1+\tau_3}{2}\big(\rho^q_{\mathbf{k},11} - \rho^q_{\mathbf{k}',22}\big)\Big|^{|\xi_k|=|\xi_{k'}|}_{\delta\theta_{\mathbf{kk'}}=\frac{E_k}{kv_s}} + \frac{1-\tau_3}{2}\big(\rho^q_{\mathbf{k},22} - \rho^q_{\mathbf{k}',11}\big)\Big|^{|\xi_k|=|\xi_{k'}|}_{\delta\theta_{\mathbf{kk'}}=-\frac{E_k}{kv_s}}\Bigg]. \quad (18)$$

Here, $1/\tau_k = 2n_i\pi|V_0|^2 D_0\lambda_c(1 + 4u_k^2 v_k^2)$ with $D_0 = mk_F/(2\pi^2)$ denoting the density of states and λ_c being a dimensionless parameter; $\delta\theta_{\mathbf{kk'}} = (\cos\theta_{\mathbf{k}'} - \cos\theta_{\mathbf{k}})/2$. Consequently, the scattering term is simplified.

B. Three-fluid model

Based on Eq. (18), we next perform an analysis on the scattering and derive a three-fluid model in the electromagnetic response in the superconducting states. Specifically, it is noted that from Eq. (18), one always has $|\delta\theta_{\mathbf{kk'}}| = E_k/(kv_s)$. Therefore since $|\delta\theta_{\mathbf{kk'}}| = |(\cos\theta_{\mathbf{k}'} - \cos\theta_{\mathbf{k}})|/2 \leqslant 1$, the scattering term is nonzero only in the region $kv_s > E_k$. This is natural since when $kv_s > E_k$, as mentioned in the introduction, the unpairing (U) region with $|\mathbf{k}\cdot\mathbf{v}_s| > E_k$, in which the particles no longer participate in the pairing and behave like the normal particles, emerges [103–106]. Then the normal fluid is present. Hence the scattering in the unpairing (U) region is nonzero. Consequently, a threshold of superconducting velocity v_s for the emergence of normal fluid and hence scattering is predicted from our theory as

$$v_L = \frac{|\Delta|}{k_F}. \quad (19)$$

As mentioned in the introduction, we refer to this threshold in superconducting state as Landau threshold, following Landau in bosonic liquid helium II theory [33].

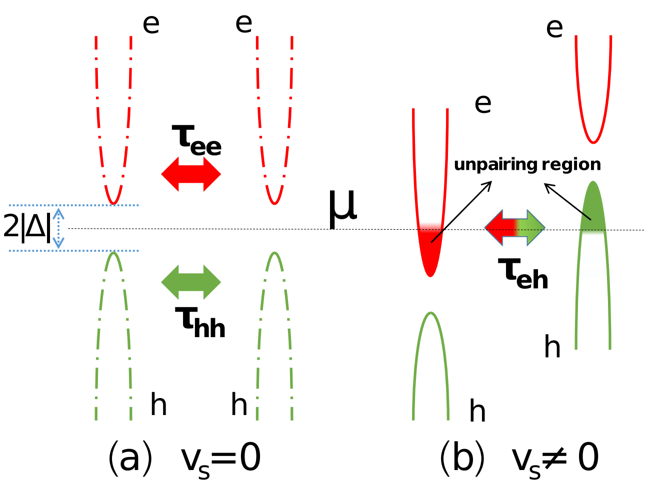

FIG. 2. Schematic showing the tilt of the quasiparticle energy spectrum and scattering processes. The chain (solid) curves represent the quasiparticle energy in the absence (presence) of a large superconducting velocity. The filled arrows represent the scattering process. In (a), the interband scattering between the quasielectrons and quasiholes is forbidden by the energy conservation. Only the intra-quasi-electron-band (denoted by τ_{ee}) and quasi-hole-band (denoted by τ_{hh}) scatterings exist. In (b), the presence of the large superconducting velocity ($kv_s > E_k$) tilts the quasiparticle energy spectrum and hence the unpairing regions (represented by red and green regions) emerge. In this case, the interband scattering between the quasielectrons and quasiholes (denoted by τ_{eh}) is turned on.

Besides U region, there also exists a special pairing region ($\mathrm{P_v}$ region) with $kv_s > E_k$ and $|\mathbf{k}\cdot\mathbf{v}_s| < E_k$, in which the scattering is also finite since $kv_s > E_k$. This is due to the fact that this region shares the same momentum magnitude with U region, as shown in Fig. 1. Since the short-range impurity scattering is isotropic in the momentum space, the particles in $\mathrm{P_v}$ region participate in the pairing but experience scattering with those in U region, and hence the superfluid from $\mathrm{P_v}$ region becomes viscous. This can also be understood as follows. In the first term on the right-hand side of Eq. (18), the particle with $\mathbf{k}$ is scattered by that with $\mathbf{k}'$. When the $\mathbf{k}$ particle is in $\mathrm{P_v}$ region ($|\mathbf{k}\cdot\mathbf{v}_s| < E_k$ but $kv_s > E_k$), one has $-3E_{k'} < \mathbf{k}'\cdot\mathbf{v}_s < E_{k'}$, and hence, the $\mathbf{k}'$ particle sits in U region. This indicates that the particles in $\mathrm{P_v}$ region experience the scattering from those in U region. By using similar analysis, one can find that the particles in U region experience the scattering from those in *both* U and $\mathrm{P_v}$ regions. The internal scattering in U region is natural since the particles in U region behave like the normal ones. Whereas the interscattering between $\mathrm{P_v}$ and U regions denotes the existence of the friction between the superfluid and normal fluid. Therefore the superfluid from $\mathrm{P_v}$ region becomes viscous. As for the remaining pairing region ($\mathrm{P_{nv}}$ region with $kv_s < E_k$), the superfluid in this region is still nonviscous.

Consequently, a three-fluid model for the electromagnetic response in the superconducting states at $v_s \geqslant v_L$ is predicted from our theory: normal fluid (from U region) and nonviscous (from $\mathrm{P_{nv}}$ region) and viscous (from $\mathrm{P_v}$ region) superfluids. Based on this three-fluid model, in the following sections, we show that the electromagnetic properties of the superconducting states including both the magnetic and optical responses can be well captured.

C. Magnetic response

In this part, by using the gauge-invariant kinetic equation, we investigate the stationary magnetic response in the superconducting states. The properties of the excited current and superconducting order parameter are addressed.

1. Solution of density matrix

In the stationary situation, one has $\partial_t\rho_{\mathbf{k}} = 0$, $\phi = 0$, and $e\mathbf{E} = 0$ in the kinetic equation. By expanding the density matrix as $\rho_{\mathbf{k}} = \rho_{\mathbf{k}0}\tau_0 + \rho_{\mathbf{k}-}\tau_- + \rho_{\mathbf{k}+}\tau_+ + \rho_{\mathbf{k}3}\tau_3$, Eq. (8) becomes

$$\Big(\varepsilon_k - \mu + \frac{\varepsilon_{\mathbf{p}-2e\mathbf{A}}}{4}\Big)\rho_{\mathbf{k}+} = \Big[\rho_{\mathbf{k}3} - \frac{i\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}0}\cdot(\nabla-2ie\mathbf{A})}{2} - \frac{\boldsymbol{\partial}_{\mathbf{k}}\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}3}:(\nabla-2ie\mathbf{A})(\nabla-2ie\mathbf{A})}{8}\Big]\Delta + \frac{\{\partial_t\rho_{\mathbf{k}}|_{\rm sc}\}_+}{2}, \tag{20}$$

$$\frac{\mathbf{k}}{m}\cdot\nabla\rho_{\mathbf{k}0} = i\Delta^*\rho_{\mathbf{k}+} - i\Delta\rho_{\mathbf{k}-} + \{\partial_t\rho_{\mathbf{k}}|_{\rm sc}\}_3 + \frac{i\boldsymbol{\partial}_{\mathbf{k}}\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}-}:(\nabla-2ie\mathbf{A})(\nabla-2ie\mathbf{A})\Delta}{8} - \frac{i\boldsymbol{\partial}_{\mathbf{k}}\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}+}:(\nabla+2ie\mathbf{A})(\nabla+2ie\mathbf{A})\Delta^*}{8}, \tag{21}$$

with $\rho_{\mathbf{k}-} = \rho^*_{\mathbf{k}+}$. Since $\mu \gg \varepsilon_{\mathbf{p}-2e\mathbf{A}}$ thanks to the large Fermi energy in the conventional superconductors, $\varepsilon_{\mathbf{p}-2e\mathbf{A}}$ on the left-hand side of Eq. (20) can be neglected.

Then, from Eqs. (20) and (21), by only keeping the diagonal terms in the density matrix in the quasiparticle space due to their leading contribution, the solution of the density matrix in the quasiparticle space is obtained as (refer to Appendix B)

$$\rho^q_{\mathbf{k}} = \begin{pmatrix} f(E^+_{\mathbf{k}}) & 0 \\ 0 & f(E^-_{\mathbf{k}}) \end{pmatrix} + (\mathbf{k}\cdot\mathbf{v}_s)\begin{pmatrix} a^+_{\mathbf{k}} & 0 \\ 0 & a^-_{\mathbf{k}} \end{pmatrix} + (\mathbf{k}\cdot\mathbf{v}_s)|\Delta|^2\begin{pmatrix} m^+_{\mathbf{k}} & 0 \\ 0 & m^-_{\mathbf{k}} \end{pmatrix} + \frac{(\mathbf{k}\cdot\mathbf{v}_s)^2}{2}\begin{pmatrix} b^+_{\mathbf{k}} & 0 \\ 0 & b^-_{\mathbf{k}} \end{pmatrix} + (\mathbf{k}\cdot\mathbf{v}_s)|\Delta|^2\begin{pmatrix} \delta m^+_{\mathbf{k}} & 0 \\ 0 & \delta m^-_{\mathbf{k}} \end{pmatrix}, \tag{22}$$

with

$$a^\pm_{\mathbf{k}} = \mp\partial_{E_k} f(E^\pm_{\mathbf{k}}), \tag{23}$$

$$b^\pm_{\mathbf{k}} = \partial^2_{E_k} f(E^\pm_{\mathbf{k}}) + \frac{\partial_{E_k} f(E^\pm_{\mathbf{k}})}{E_k}, \tag{24}$$

$$m^\pm_{\mathbf{k}} = \pm\Big[\frac{1}{E_k}\partial_{E_k} + \frac{1}{4\xi_k\varepsilon_k}\Big]\frac{f(E^\pm_{\mathbf{k}})}{E_k}, \tag{25}$$

$$\delta m^\pm_{\mathbf{k}} = \mp\frac{\xi}{\tau_k v_F}\theta\Big(\frac{kv_s}{E_k}\Big)\frac{\xi_k}{E_k}m^\pm_{\mathbf{k}}. \tag{26}$$

Here, $\mathbf{v}_s = \mathbf{p}_s/m$ (refer to Appendix B); the gauge invariant $\mathbf{p}_s = \nabla\psi/2 - e\mathbf{A}$ denotes the superconducting momentum

[60–63,65], $f(x)$ represents the Fermi distribution, and $\theta(x)$ is the step function.

As seen from Eq. (22), the first term in $\rho^q_{\mathbf{k}}$ represents the quasiparticle distribution of the FFLO-like state. The second term stands for the linear response of the quasiparticle state. The third term denotes the Meissner supercurrent response, which is proved in the following. The forth term represents the nonlinear response. The last term is the scattering contribution, which emerges at $kv_s > E_k$ as mentioned in Sec. III B.

2. *Excited current*

With Eqs. (22) and (11), by neglecting the nonlinear response, the excited current in the stationary magnetic response reads

$$\begin{aligned}\mathbf{j} &= \frac{2e}{m}\sum_{\mathbf{k}}\mathbf{k}\rho_{\mathbf{k}0} = \frac{2e}{m}\sum_{\mathbf{k}}\mathbf{k}\rho^q_{\mathbf{k}0}\\ &= \frac{2e}{m}\sum_{\mathbf{k}}\mathbf{k}\bigg[\frac{f(E^+_{\mathbf{k}})+f(E^-_{\mathbf{k}})+(\mathbf{k}\cdot\mathbf{v}_s)(a^+_{\mathbf{k}}+a^-_{\mathbf{k}})}{2}\\ &\quad+(\mathbf{k}\cdot\mathbf{v}_s)|\Delta|^2\frac{m^+_{\mathbf{k}}+m^-_{\mathbf{k}}+\delta m^+_{\mathbf{k}}+\delta m^-_{\mathbf{k}}}{2}\bigg].\end{aligned} \tag{27}$$

When $v_s < v_L$, no U region emerges and the momentum space belongs to the nonviscous pairing ($\mathrm{P_{nv}}$) region. Therefore, only the nonviscous superfluid is present. Then, one has $f(E^\pm_{\mathbf{k}}) \approx f(\pm E_k) + (\mathbf{k}\cdot\mathbf{v}_s)\partial_{E_k} f(E_k)$, and Eq. (27) becomes

$$\begin{aligned}\mathbf{j} &= e\mathbf{v}_s D_0\int\frac{d\Omega}{4\pi}\cos^2\theta_{\mathbf{k}}\int d\xi_k\left(4\varepsilon_{k_F}|\Delta|^2\frac{m^+_{\mathbf{k}}+m^-_{\mathbf{k}}}{2}\right)\\ &= e\mathbf{v}_s D_0\int\frac{d\Omega}{4\pi}\cos^2\theta_{\mathbf{k}}\int d\xi_k\rho_{m\mathbf{k}},\end{aligned} \tag{28}$$

with

$$\rho_{m\mathbf{k}} = \frac{4\varepsilon_{k_F}|\Delta|^2}{E_k}\partial_{E_k}\left[\frac{f(E^+_k)-f(E^-_k)}{2E_k}\right]. \tag{29}$$

In the pairing region, with $\rho_{m\mathbf{k}} \approx \frac{4\varepsilon_{k_F}|\Delta|^2}{E_k}\partial_{E_k}[\frac{2f(E_k)-1}{2E_k}]$, the current reads

$$\mathbf{j} = e\mathbf{v}_s N_0|\Delta|^2\frac{7R(3)}{4(\pi T)^2}, \tag{30}$$

which is exactly the same as the Meissner supercurrent in the literature [53,54]. Here, $R(x)$ is the Riemann zeta function and N_0 represents the electron density. Consequently, we refer to $\rho_{m\mathbf{k}}$ as the Meissner superfluid density. Particularly, it is noted that the excited Meissner supercurrent entirely comes from $m^\pm_{\mathbf{k}}$ terms, indicating that the third term in Eq. (22) gives rise to the Meissner supercurrent response.

For the case $v_s > v_L$, as mentioned in Sec. III B, the normal fluid (from U region) and nonviscous (from $\mathrm{P_{nv}}$ region) and viscous (from $\mathrm{P_v}$ region) superfluids are present. In this situation, considering the fact that $f(E^\pm_{\mathbf{k}}) \approx f(\pm E_k) + (\mathbf{k}\cdot\mathbf{v}_s)\partial_{E_k} f(E_k)$ in $\mathrm{P_v}$ and $\mathrm{P_{nv}}$ regions and $f(E^\pm_{\mathbf{k}}) \approx f(\mathbf{k}\cdot\mathbf{v}_s) \pm E_k\partial_{\mathbf{k}\cdot\mathbf{v}_s} f(\mathbf{k}\cdot\mathbf{v}_s)$ in U region, with $f(\mathbf{k}\cdot\mathbf{v}_s) \approx f(0) + (\mathbf{k}\cdot\mathbf{v}_s)\partial_0 f(0)$ near the Fermi surface, Eq. (27) becomes

$$\mathbf{j} = \mathbf{j}_{\mathrm{P_{nv}}} + \mathbf{j}_{\mathrm{P_v}} + \mathbf{j}_{\mathrm{U}}, \tag{31}$$

where

$$\mathbf{j}_{\mathrm{P_{nv}}} = e\mathbf{v}_s\sum_{\mathbf{k}\in\mathrm{P_{nv}}}\rho_{m\mathbf{k}}\cos^2\theta_{\mathbf{k}}, \tag{32}$$

$$\mathbf{j}_{\mathrm{P_v}} = e\mathbf{v}_s\sum_{\mathbf{k}\in\mathrm{P_v}}\left(1-\frac{\xi}{l}\right)\rho_{m\mathbf{k}}\cos^2\theta_{\mathbf{k}}, \tag{33}$$

$$\mathbf{j}_{\mathrm{U}} = -e\mathbf{v}_s\sum_{\mathbf{k}\in\mathrm{U}}\frac{\xi}{l}\rho_{m\mathbf{k}}\cos^2\theta_{\mathbf{k}}. \tag{34}$$

Here, $l = 3N_0\tau_k v_F/\pi^3[1/(2D_0E_k) + \partial_{(D_0E_k)}f(E_k)]$ denotes the mean-free path in the superconducting states.

The features of Eq. (31) can be well captured by the three-fluid model described in Sec. III B. Specifically, without the scattering ($1/l = 0$), the Meissner supercurrent ($\mathbf{j}_{\mathrm{P_{nv}}}$+$\mathbf{j}_{\mathrm{P_v}}$) is excited in the superfluid ($\mathrm{P_v}$ and $\mathrm{P_{nv}}$ regions) whereas no current ($\mathbf{j}_{\mathrm{U}} = 0$ when $1/l = 0$) is directly excited from the magnetic flux in the normal fluid (U region) as it should be. Nevertheless, in the presence of the scattering ($1/l \neq 0$), the normal-fluid current $\mathbf{j}_{\mathrm{U}}$ can be induced through the friction drag with the superfluid current mentioned in Sec. III B. Moreover, due to this friction, the superfluid current $\mathbf{j}_{\mathrm{P_v}}$ becomes viscous, while $\mathbf{j}_{\mathrm{P_{nv}}}$ is still nonviscous.

Thanks to the normal-fluid and viscous-superfluid currents, the penetration depth is influenced by the scattering. Particularly, by only considering the viscous superfluid current $\mathbf{j}_{\mathrm{P_v}}$, the penetration depth reads $\delta^2 = \delta_c^2/(1-\xi/l) \approx \delta_c^2(1+\xi/l)$ at the weak scattering, exactly the same as the one from Tinkham's discussion [2]. Nevertheless, due to the presences of both the normal-fluid current induced by friction drag and the nonviscous superfluid, the dependence of the penetration depth becomes

$$\delta^2 = \delta_c^2(1+\xi/l_{\mathrm{eff}}), \tag{35}$$

with the clean-limit penetration depth δ_c and effective mean-free path l_{eff} given by

$$\delta_c = \left(e^2\sum_{\mathbf{k}\in\mathrm{P}}\rho_{m\mathbf{k}}\cos^2\theta_{\mathbf{k}}\right)^{-\frac{1}{2}}, \tag{36}$$

$$\frac{1}{l_{\mathrm{eff}}} = \frac{\sum_{\mathbf{k}\in(\mathrm{P_v}+\mathrm{U})}\frac{\rho_{m\mathbf{k}}\cos^2\theta_{\mathbf{k}}}{l}}{\sum_{\mathbf{k}\in\mathrm{P}}\rho_{m\mathbf{k}}\cos^2\theta_{\mathbf{k}}}, \tag{37}$$

respectively.

3. *Modified Ginzburg-Landau equation*

In this part, we investigate the stationary magnetic response of the superconducting order parameter. We first focus on the case at $v_s < v_L$, in which only the nonviscous superfluid is present. In this situation, we prove that the gap equation in our theory [Eq. (9)] exactly reduces to the Ginzburg-Landau theory [12,53,54] (refer to Appendix C).

We next focus on the situation at $v_s > v_L$, in which both the normal fluid and superfluid are present. Specifically, with

Eq. (22), from the gap equation [Eq. (8)], one has

$$\Delta = V{\sum_{\mathbf{k}}}'\left[-\frac{\Delta}{E_k}\rho^q_{\mathbf{k}3}\right]$$
$$= -V{\sum_{\mathbf{k}}}'\frac{\Delta}{E_k}\bigg[\frac{f(E^+_{\mathbf{k}})-f(E^-_{\mathbf{k}})}{2}+(\mathbf{k}\cdot\mathbf{v}_s)\frac{a^+_{\mathbf{k}}-a^-_{\mathbf{k}}}{2}$$
$$+\frac{(\mathbf{k}\cdot\mathbf{v}_s)^2}{2}\frac{b^+_{\mathbf{k}}-b^-_{\mathbf{k}}}{2}+(\mathbf{k}\cdot\mathbf{v}_s)|\Delta|^2\frac{m^+_{\mathbf{k}}-m^-_{\mathbf{k}}}{2}$$
$$+(\mathbf{k}\cdot\mathbf{v}_s)|\Delta|^2\frac{\delta m^+_{\mathbf{k}}-\delta m^-_{\mathbf{k}}}{2}\bigg]. \tag{38}$$

By using the same expansion of $f(E^\pm_{\mathbf{k}})$ in each regions in Sec. III C 2, Eq. (38) becomes

$$\Delta\left[\sum_{\mathbf{k}\in\mathrm{P}}\frac{1-2f(E_k)}{2E_k}-\frac{1}{V}\right]-mv_s^2\Delta\lambda=0, \tag{39}$$

where

$$\lambda=\varepsilon_{k_F}\left[\sum_{\mathbf{k}\in\mathrm{P}}\frac{\cos^2\theta_{\mathbf{k}}\partial^2_{E_k}f(E_k)}{E_k}\right]. \tag{40}$$

Near the critical temperature, the superconducting order parameter can be treated as a small quantity. Then, with $\mathbf{v}_s\Delta = (\nabla\psi - 2e\mathbf{A})\Delta/(2m) \approx (-i\nabla - 2e\mathbf{A})/(2m)$, Eq. (39) can be transformed into

$$\left\{\frac{\lambda(\nabla-2ie\mathbf{A})^2}{4m}+[\alpha-\beta|\Delta|^2]\right\}\Delta=0, \tag{41}$$

with

$$\alpha=\sum_{\mathbf{k}\in\mathrm{P}}\left[\frac{1-2f(E_k)}{2E_k}\right]\bigg|_{|\Delta|=0}-\frac{1}{V}, \tag{42}$$

$$\beta=\sum_{\mathbf{k}\in\mathrm{P}}\left\{\frac{1}{2E_k}\partial_{E_k}\left[\frac{2f(E_k)-1}{2E_k}\right]\right\}\bigg|_{|\Delta|=0}. \tag{43}$$

Consequently, a modified Ginzburg-Landau theory is obtained. Particularly, it is noted that the calculation of the phenomenological parameters α and β is restricted to the pairing (P) region.

4. *Exotic phase with both finite resistivity and order parameter*

In this part, we show the volume proportion of the unpairing region ($V_{\mathrm{U}} = \sum_{\mathbf{k}\in\mathrm{U}}\Xi^{-1}$) and the viscous ($V_{\mathrm{P_v}} = \sum_{\mathbf{k}\in\mathrm{P_v}}\Xi^{-1}$) and nonviscous ($V_{\mathrm{P_{nv}}} = \sum_{\mathbf{k}\in\mathrm{P_{nv}}}\Xi^{-1}$) pairing regions during the magnetic response in Fig. 3 by performing a numerical calculation for a specific material, Pb, by self-consistently solving the gap equation [Eq. (39)]. Here, $\Xi = \sum'_{\mathbf{k}} 1$ is the volume of the spherical shell. As seen from the figure, when $v_s < v_L$, only the nonviscous superfluid ($V_{\mathrm{P_{nv}}} \neq 0$) is present. When $v_L < v_s < 9.5v_L \approx \omega_D/k_F$, the finite $V_{\mathrm{P_{nv}}}$, $V_{\mathrm{P_v}}$, and V_{U} indicate that the normal fluid (from U region) and nonviscous (from $\mathrm{P_{nv}}$ region) and viscous (from $\mathrm{P_v}$ region) superfluids are present. Actually, in most conventional superconducting materials, due to the large k_F, the value of the Landau threshold v_L is very small (for Pb, one has $v_L \approx 0.33$ nm/ps at $T = 0$ K and the corresponding vector potential is $eA \approx 2.9\times10^{-3}$ nm^{-1}) and hence hard to be detected.

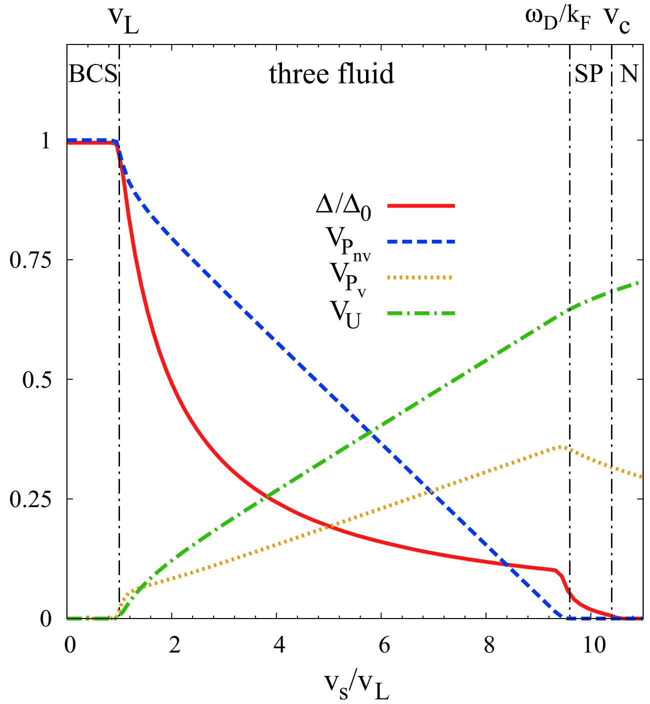

FIG. 3. Superconducting order parameter Δ and volume proportions of the unpairing region V_{U} and viscous $V_{\mathrm{P_v}}$ and nonviscous $V_{\mathrm{P_{nv}}}$ pairing regions versus superconducting velocities v_s. The order parameter is self-determined from the gap equation [Eq. (39)]. Δ_0 denotes the BCS superconducting order parameter at zero temperature. The used parameter in the calculation includes $E_F = 1.021$ eV [109], $\omega_D = 10.75$ meV [110], $m = m_e$, $\Delta_0 = 1.13$ meV [110], and $T = 0.02$ K. m_e represents the free electron mass. $v_L = |\Delta_0|/k_F$. The vertical chain line stands for the crossover. N denotes the normal state. SP denotes the special phase that has both finite resistivity and order parameter. v_c denotes the critical point into the normal state.

Interestingly, before the superconducting gap $|\Delta|$ becomes zero (i.e., at $v_s < v_c$, where v_c denotes the critical point into the normal state and $v_c \approx 10.4v_L$ here from the self-consistent calculation), with the increase of v_s after $\omega_D/k_F \approx 9.5v_L$, we find that the superconducting state falls into a special phase, in which the nonviscous superfluid vanishes ($V_{\mathrm{P_{nv}}} = 0$), leaving only the viscous superfluid ($V_{\mathrm{P_v}} \neq 0$) and normal fluid ($V_{\mathrm{U}} \neq 0$). This is because the increase of v_s at $v_s > v_L$ enlarges U and hence $\mathrm{P_v}$ regions. When $v_s > \omega_D/k_F$, as shown in Fig. 4, the spherical shell by the BCS theory is filled with U and $\mathrm{P_v}$ regions and $\mathrm{P_{nv}}$ region (nonviscous superfluid) vanishes. Particularly, due to the absence of the nonviscous superfluid, the resistivity in this phase is finite but the superconducting gap is *finite*.

In high-temperature superconductors[111–116] and strongly disordered superconductors [117–120], the phase with both finite resistivity and gap, known as pseudogap phase, has been widely studied. In the present work, we point out that in the conventional superconductors, the phase with both finite resistivity and gap can also be realized by tuning the magnetic flux. Nevertheless, to realize this special phase, the emergence point ω_D/k_F of this phase must be smaller than the critical point v_c at which the superconducting gap becomes

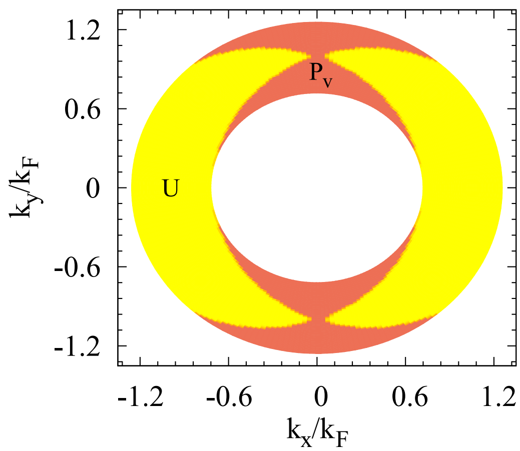

FIG. 4. Schematic showing the division in the momentum space (in the spherical shell by the BCS theory) when $v_s > \omega_D/k_F$. In this situation, the spherical shell by the BCS theory is divided into only two parts: U region, denoted by the yellow regions, P_{nv} region, denoted by the orange region.

zero. Thus small Debye frequency and low temperature are necessary. Consequently, materials Pb, Hg, and V, which possess small Debye frequency [110], are ideal candidates. For the experimental detection, the finite resistivity can be detected through the electrical methods [111–113,118–120], whereas the finite gap can be measured by using the scanning tunneling microscope [111,112,115,117,118,120,121] or angle-resolved photoemission spectroscopy [111,122].

D. Optical response

We next study the optical response in the superconducting states in both linear and nonlinear regimes. The properties of the optical current and excited Higgs mode are addressed.

1. Solution of density matrix

In the optical response, we first choose a specific gauge with zero superconducting phase for the convenience of the physical analysis, and considering the translational symmetry, the spatial gradient terms in Eq. (8) can be neglected. Then, the kinetic equation reads

$$\begin{aligned}\partial_T\rho_{\mathbf{k}} &+ i[(\xi_k+\mu_{\rm eff})\tau_3+|\Delta|\tau_1,\rho_{\mathbf{k}}]+i\left[\frac{p_s^2}{2m}\tau_3,\rho_{\mathbf{k}}\right]\\ &+\frac{1}{2}\{e\mathbf{E}\tau_3,\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}}\}+\{\mathbf{p}_s|\Delta|\tau_2,\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}}\}\\ &+\frac{i}{2}[\mathbf{p}_s\mathbf{p}_s|\Delta|\tau_1,\boldsymbol{\partial}_{\mathbf{k}}\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}}]=\partial_t\rho_{\mathbf{k}}|_{\rm sc}. \qquad (44)\end{aligned}$$

Here, the superconducting momentum $\mathbf{p}_s=-e\mathbf{A}+\frac{1}{2}\boldsymbol{\nabla}_{\mathbf{R}}\psi$ and the effective chemical potential $\mu_{\rm eff}=e\phi+\frac{1}{2}\partial_t\psi$, related by the acceleration relation $e\mathbf{E}=\partial_t\mathbf{p}_s-\boldsymbol{\nabla}\mu_{\rm eff}$, are gauge-invariant physical quantities [60,63]. Particularly, in the presence of the translational symmetry, the electric field reads $e\mathbf{E}=\partial_t\mathbf{p}_s-\boldsymbol{\nabla}\mu_{\rm eff}=i\omega\mathbf{p}_s$ in the optical response with ω being the optical frequency. On the left-hand side of Eq. (44), the third term represents the Anderson-pseudospin pump effect [37,38,40–45,47] and the forth one is the drive effect, exactly as those revealed in the previous work by Yu and Wu [60]. Whereas the last two terms on the left-hand side of Eq. (44), which stand for the Ginzburg-Landau kinetic effect, are absent in Ref. [60].

To obtain the solution, we transform Eq. (44) from the particle space into the quasiparticle one as

$$\begin{aligned}\partial_T\rho^q_{\mathbf{k}} &+ i[E_k\tau_3,\rho^q_{\mathbf{k}}]+i[\mu_{\rm eff}t_3,\rho^q_{\mathbf{k}}]+i\left[\frac{p_s^2}{2m}t_3,\rho^q_{\mathbf{k}}\right]\\ &+\frac{1}{2}\{e\mathbf{E}t_3+2\mathbf{p}_s|\Delta|t_2,\boldsymbol{\partial}_{\mathbf{k}}\rho^q_{\mathbf{k}}+[U_k^\dagger\boldsymbol{\partial}_{\mathbf{k}}U_k,\rho^q_{\mathbf{k}}]\}\\ &+\frac{i}{2}[\mathbf{p}_s\mathbf{p}_s|\Delta|\tau_1,\boldsymbol{\partial}_{\mathbf{k}}\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}}+2[U_k^\dagger\boldsymbol{\partial}_{\mathbf{k}}U_k,\boldsymbol{\partial}_{\mathbf{k}}\rho^q_{\mathbf{k}}]]\\ &+\frac{i}{2}[\mathbf{p}_s\mathbf{p}_s|\Delta|\tau_1,U_k^\dagger\boldsymbol{\partial}_{\mathbf{k}}\boldsymbol{\partial}_{\mathbf{k}}U_k\rho^q_{\mathbf{k}}+\rho^q_{\mathbf{k}}(\boldsymbol{\partial}_{\mathbf{k}}\boldsymbol{\partial}_{\mathbf{k}}U_k^\dagger)U_k]\\ &-\frac{i}{2}[\mathbf{p}_s\mathbf{p}_s|\Delta|\tau_1,2U_k^\dagger\boldsymbol{\partial}_{\mathbf{k}}U_k\rho^q_{\mathbf{k}}U_k^\dagger\boldsymbol{\partial}_{\mathbf{k}}U_k]=\partial_t\rho^q_{\mathbf{k}}|_{\rm sc}, \qquad (45)\end{aligned}$$

in which $t_i=U_k^\dagger\tau_iU_k$.

Then, from Eq. (45), the solution of the density matrix in the quasiparticle space is derived as (refer to Appendix D)

$$\rho^q_{\mathbf{k}}=\rho^{q0}_{\mathbf{k}}-(\mathbf{k}\cdot\mathbf{v}_s)\rho^{q1}_{\mathbf{k}}+\frac{(\mathbf{k}\cdot\mathbf{v}_s)^2}{2}\rho^{q2}_{\mathbf{k}}+mv_s^2\rho^{q3}_{\mathbf{k}}+\delta\rho^{qs}_{\mathbf{k}}, \qquad (46)$$

$$\rho^{q0}_{\mathbf{k}}=\begin{pmatrix}f(E^+_{\mathbf{k}}) & 0\\ 0 & f(E^-_{\mathbf{k}})\end{pmatrix}, \qquad (47)$$

$$\rho^{q1}_{\mathbf{k}}=\frac{\rho_{m\mathbf{k}}\tau_0}{4\varepsilon_{k_F}}, \qquad (48)$$

$$\delta\rho^{qs}_{\mathbf{k}}=-\frac{(\mathbf{k}\cdot\mathbf{v}_s)}{i\omega\tau_k}\theta\left(\frac{kv_s}{E_k}\right)(\partial_{E_k}\rho^{q0}_{\mathbf{k}3}+\hat{O}_kf_{\mathbf{k}})\tau_0, \qquad (49)$$

in which, $\mathbf{v}_s=-\frac{e\mathbf{E}}{i\omega m}$ (refer to Appendix); $\hat{O}_k=4u_k^2v_k^2(1/E_k-\partial_{E_k})$ and $f_{\mathbf{k}}=[3f(E^+_{\mathbf{k}})-3f(E^-_{\mathbf{k}})-f(E^+_{\mathbf{k}}+2E_k)+f(E^-_{\mathbf{k}}-2E_k)]/8$; the specific expressions of $\rho^{q2}_{\mathbf{k}}$ and $\rho^{q3}_{\mathbf{k}}$ are given by Eqs. (D6) and (D8) in Appendix, respectively.

As seen from Eq. (46), the first term in $\rho^q_{\mathbf{k}}$ represents the quasiparticle distribution. The second term, in which $\rho_{m\mathbf{k}}$ is exactly same as the Meissner-superfluid density [Eq. (29)] in the stationary magnetic response, stands for the Meissner response. The third and forth terms denote the nonlinear response. The last term is the scattering contribution, which emerges at $kv_s > E_k$ as mentioned in Sec. III B.

2. Optical current

We first investigate the properties of the optical current. In contrast to the two-fluid model in the literature [1,2,5,18,19,21–23,34], we show that the optical current is well captured by the three-fluid model described in Sec. III B. Specifically, with Eqs. (46) and (11), by neglecting the nonlinear response, the optical current reads

$$\begin{aligned}\mathbf{j}&=\frac{2e}{m}\sum_{\mathbf{k}}\mathbf{k}\rho_{\mathbf{k}0}=\frac{2e}{m}\sum_{\mathbf{k}}\mathbf{k}\rho^q_{\mathbf{k}0}\\ &=\frac{2e}{m}\sum_{\mathbf{k}}\mathbf{k}[\rho^{q0}_{\mathbf{k}0}-(\mathbf{k}\cdot\mathbf{v}_s)\rho^{q1}_{\mathbf{k}}+\delta\rho^{qs}_{\mathbf{k}}]. \qquad (50)\end{aligned}$$

At $v_s < v_L$ with only the nonviscous superfluid, the current is written as

$$\mathbf{j} = \frac{e^2\mathbf{E}}{im\omega}\sum_{\mathbf{k}}\cos^2\theta_{\mathbf{k}}\big[\rho_{m\mathbf{k}} - 4\varepsilon_{k_F}\partial_{E_k}f(E_k)\big]. \quad (51)$$

Besides the Meissner supercurrent ($\rho_{m\mathbf{k}}$), a Bogoliubov quasiparticle current $[4\varepsilon_{k_F}\partial_{E_k}f(E_k)]$ is also present in the superfluid during the optical response. The presence of the Bogoliubov quasiparticle current is natural, since the drive from the optical field causes the drift of the electron states, resulting in a center-of-mass momentum in superconducting states [60].

As for the case $v_s > v_L$ with the presences of the normal fluid and nonviscous and viscous superfluids, by using the same expansion of $f(E^{\pm}_{\mathbf{k}})$ in each region in Sec. III C 2, in the weak-scattering limit, the current becomes

$$\mathbf{j} = (\sigma_{\mathrm{P_{nv}}} + \sigma_{\mathrm{P_v}} + \sigma_{\mathrm{U}})\mathbf{E}, \quad (52)$$

with

$$\sigma_{\mathrm{P_{nv}}} = \frac{e^2}{im\omega}\sum_{\mathbf{k}\in\mathrm{P_{nv}}}\cos^2\theta_{\mathbf{k}}\big[\rho_{m\mathbf{k}} - 4\varepsilon_{k_F}\partial_{E_k}f(E_k)\big], \quad (53)$$

$$\sigma_{\mathrm{P_v}} = \frac{e^2}{m}\sum_{\mathbf{k}\in\mathrm{P_v}}\cos^2\theta_{\mathbf{k}}\left[\frac{\rho_{m\mathbf{k}}}{i\omega + (2\tau_k)^{-1}} - \frac{4\varepsilon_{k_F}\partial_{E_k}f(E_k)}{i\omega + \tau_k^{-1}}\right], \quad (54)$$

$$\sigma_{\mathrm{U}} = -\frac{e^2}{m}\sum_{\mathbf{k}\in\mathrm{U}}\cos^2\theta_{\mathbf{k}}\frac{4\varepsilon_{k_F}\partial_{\xi_k}f(\xi_k)}{i\omega + \tau_k^{-1}}. \quad (55)$$

Specifically, the excited superfluid current consists of the Meissner supercurrent ($\rho_{m\mathbf{k}}$) and Bogoliubov quasiparticle current $[4\varepsilon_{k_F}\partial_{E_k}f(E_k)]$, as mentioned above. Due to the presence of the friction between the superfluid and normal-fluid currents mentioned in Sec. III B, the superfluid current is separated into the nonviscous $\sigma_{\mathrm{P_{nv}}}\mathbf{E}$ and viscous $\sigma_{\mathrm{P_v}}\mathbf{E}$ ones, and the former (latter) exhibits zero (finite) resistance τ_k^{-1}, whereas the normal-fluid optical conductivity σ_{U} exhibits the well-known Drude-model behavior. Particularly, in the normal state with the normal fluid alone, it exactly reduces to $\sigma_{\mathrm{U}} = \frac{e^2N\tau}{m(1+i\omega\tau)}$ from the Drude model [123].

In the superconducting state, at low temperature, few Bogoliubov quasiparticles $[f(E_k)\approx 0]$ are excited in the superfluid. Thus the Bogoliubov quasiparticle current is marginal and the superfluid current only consists of the Meissner supercurrent. In this situation, if we neglect the friction between superfluid and normal-fluid currents, i.e., the viscous superfluid ($\sigma_{\mathrm{P_v}}$), the optical conductivity $\sigma = \sigma_{\mathrm{U}} + \sigma_{\mathrm{P_{nv}}}$ from our theory [Eqs. (53) and (55) with $f(E_k)\approx 0$] is exactly the same as the one from two-fluid model, $\sigma_{\mathrm{two}} = \frac{e^2\rho_m}{im\omega} + \frac{e^2\tau\rho_n}{m(1+i\omega\tau)}$ [1,2,5,18,19,21–23,34], in which ρ_m is the total Meissner-superfluid density and ρ_n denotes the total normal-fluid density. Nevertheless, the presence of viscous superfluid here suggests that the optical response is captured by the three-fluid model and the two-fluid model in the literature [1,2,5,18,19,21–23,34] is insufficient for a complete picture. Actually, although the viscous superfluid has been hinted in the stationary magnetic response in the literature [2,50], it has long been overlooked in the optical response.

3. Higgs mode

Finally, we discuss the optically excited Higgs mode. Comparison between the Anderson-pseudospin pump effect [37,38,40–45,47] [third term in Eq. (44)] and the drive effect [forth term in Eq. (44)] revealed in the previous theory [60] by Yu and Wu is addressed. Particularly, in Ref. [60], it is reported that in the excitation of the Higgs mode, the drive effect is the dominant effect and the pump effect is marginal. Nevertheless, as pointed out in Sec. II B 1, the Ginzburg-Landau kinetic-energy terms [seventh and eighth terms in Eq. (44)] are absent in Ref. [60]. With these two terms, we show that the previous conclusion in Ref. [60] only holds at finite temperature.

Specifically, with the solution of density matrix in the optical response [Eq. (46)], the gap equation [Eq. (9)] becomes

$$\Delta = V\sum_{\mathbf{k}\in\mathrm{P}}\frac{\Delta}{E_k}\bigg\{-a_k + \frac{(\mathbf{k}\cdot\mathbf{v}_s)^2\xi_k^2}{2(\omega^2 - E_k^2)}\bigg[\bigg(1 - \frac{\omega|\Delta|^2}{E_k^3}\bigg)\partial_{E_k} + \frac{1}{2\varepsilon_{k_F}}\bigg(1 + \frac{\omega|\Delta|^2}{\xi_k^2E_k\cos^2\theta_{\mathbf{k}}}\bigg)\bigg]\partial_{E_k}f(E_k) + \varepsilon_{p_s}a_k\frac{|\Delta|^2}{E_k^2}\times\frac{E_k\cos^2\theta_{\mathbf{k}} - \omega}{\omega^2 - E_k^2} + \frac{\varepsilon_{p_s}\omega a_k}{\omega^2 - E_k^2}\bigg\}, \quad (56)$$

in which $a_k = f(E_k) - 1/2$. The above gap equation is calculated in the pairing region alone. In principle, the superconducting gap is self-consistently determined by the above gap equation. Nevertheless, for a weak optical field at low temperature, one has $|\delta\Delta| = |\Delta_0 - \Delta| \ll |\Delta_0|$ with $\Delta_0 = V\sum_{\mathbf{k}\in\mathrm{P}}\frac{1}{2E_k}$ being the gap at zero temperature.

At low temperature, considering the large Fermi energy in conventional superconductors, from Eq. (56), $\delta\Delta$ reads

$$\delta\Delta = \Delta_0V\sum_{\mathbf{k}\in\mathrm{P}}\frac{f(E_k)}{E_k} - \delta\Delta^{\mathrm{pump}} - \delta\Delta^{\mathrm{drive}}, \quad (57)$$

with

$$\frac{\delta\Delta^{\mathrm{pump}}}{V} = \varepsilon_{p_s}\sum_{\mathbf{k}\in\mathrm{P}}\frac{\omega a_k\Delta_0}{E_k(\omega^2 - E_k^2)} \quad (58)$$

$$\frac{\delta\Delta^{\mathrm{drive}}}{V} = \varepsilon_{p_s}\Delta_0\sum_{\mathbf{k}\in\mathrm{P}}\bigg[2\varepsilon_{k_F}\frac{\xi_k^2\cos^2\theta_{\mathbf{k}}\partial^2_{E_k}f(E_k)}{E_k(\omega^2 - E_k^2)}\times\bigg(1 - \frac{\omega|\Delta_0|^2}{E_k^3}\bigg) + a_k\frac{|\Delta|^2}{E_k^3}\frac{E_k\cos^2\theta_{\mathbf{k}} - \omega}{\omega^2 - E_k^2}\bigg]. \quad (59)$$

On the right-hand side of Eq. (57), the first term directly leads to the decrease of the superconducting gap as a consequence of the thermal effect. Particularly, this term is finite after the THz pulse and hence causes a plateau of the superconducting gap, in consistency with the experimental findings [28,29]. We point out that the second term comes from the Anderson-pseudospin pump effect [37,38,40–45,47]. The third term arises from the drive effect [60]. Both effects in the excitation of the Higgs mode, proportional to $\varepsilon_{p_s} = p_s^2/(2m)$, oscillate at twice the optical frequency.

By comparing the relative contribution of these two effects, near the Fermi surface, at zero temperature in the absence of thermal effect, the ratio between the drive and pump effects is $r_{\mathrm{drive/pump}} \approx |\frac{\Delta-3\omega}{3\omega}|$, and in the THz regime, both effects contribute. Whereas at finite temperature, thanks to the large Fermi energy, the drive effect [Eq. (59)] becomes

$$\frac{\delta\Delta^{\mathrm{drive}}}{V} = \varepsilon_{p_s}\Delta_0 2\varepsilon_{k_F}\sum_{\mathbf{k}\in\mathrm{P}}\left[\frac{\xi_k^2\cos^2\theta_{\mathbf{k}}\partial_{E_k}^2 f(E_k)}{E_k(\omega^2-E_k^2)}\times\left(1-\frac{\omega|\Delta_0|^2}{E_k^3}\right)\right]. \quad (60)$$

Then, one finds $r_{\mathrm{drive/pump}} \approx |\frac{\varepsilon_{k_F}}{3\omega}\frac{(\omega-|\Delta|)|\Delta|}{T_{\mathrm{eff}}^2\cosh^3(\frac{|\Delta|}{2T_{\mathrm{eff}}})}|$, and hence, the drive effect plays a dominant role in the excitation of the Higgs mode.

Actually, the dominant role of the drive effect can also be understood as follows. It is noted that at low frequency and small order parameter, the drive effect [Eq. (60)] becomes

$$\delta\Delta^{\mathrm{drive}}/V = -2\varepsilon_{p_s}\Delta_0\lambda = -mv_s^2\Delta_0\lambda, \quad (61)$$

which is exactly the kinetic-energy term in the Ginzburg-Landau equation [first term in Eq. (41)]. Consequently, the drive effect in our microscopic theory is related to the kinetic energy in the Ginzburg-Landau theory, in which the vector potential is involved as $(\mathbf{k}_F\cdot\mathbf{A})^2/m^2 = 4\varepsilon_{k_F}\mathbf{A}^2/(2m)$ at finite temperature. Nevertheless, in the pump effect, the vector potential is involved as $\mathbf{A}^2/(2m)$. These two responses of the vector potentials are totally different, and thanks to the large Fermi energy, the drive effect makes the dominant contribution. Consequently, the Liouville [37,38,41] or Bloch [40,42–45,47] equation in the literature with the pump effect alone is insufficient to study the optical excitation of the Higgs mode. However, although the deficiency of the Liouville or Bloch equation has been hinted according to the Ginzburg-Landau theory, it has long been overlooked in the study of the Higgs mode in the literature.

Particularly, in the experiments for the detection of Higgs mode [26,28–31], the thermal effect is inevitable because of the intense THz field. This conclusion is supported by the experimentally discovered plateau of the superconducting gag after the THz pulse, which is attributed to the thermal effect as mentioned above. Therefore we believe that the experimentally observed excitation of the Higgs mode is dominated by the drive effect. This conclusion is also supported by our numerical calculation (refer to Appendix E).

IV. SUMMARY AND DISCUSSION

In summary, we extend the kinetic theory by Yu and Wu [60] to include the superfluid, so that both the normal-fluid and superfluid dynamics are involved. As a gauge-invariant theory for the electromagnetic response, our kinetic equation can be applied to both the magnetic and optical responses. We first focus on the weak-scattering case in the present work. Rich physics is revealed.

Specifically, in the electromagnetic response, we show that the superconducting velocity v_s is always excited by the electromagnetic field. Particularly, a threshold $v_L = |\Delta|/k_F$ of superconducting velocity v_s for the emergence of normal fluid and hence the scattering is predicted from our theory, i.e., the normal fluid and scattering appear only when $v_s > v_L$. We refer to this threshold as the Landau threshold, following Landau in bosonic liquid helium II theory [33]. Interestingly, we find that there also exists friction between the normal-fluid and superfluid currents. Due to this friction, part of the superfluid becomes viscous. Therefore the superfluid consists of nonviscous superfluid and viscous one. Consequently, we propose a three-fluid model at $v_s \geqslant v_L$: normal fluid and nonviscous and viscous superfluids. We show that from this three-fluid model, the physics of the electromagnetic response in the superconducting states can be well captured.

For the stationary magnetic response, in the case with $v_s < v_L$ in which only the nonviscous superfluid is present, we prove that the excited superfluid current is the Meissner supercurrent and near the critical temperature, the gap equation in our theory reduces to the Ginzburg-Landau equation [12]. As for the situation with $v_s \geqslant v_L$ where both the superfluid and normal fluid are present, differing from the excited Meissner supercurrent in the superfluid, no current is directly excited from the magnetic flux in the normal fluid. Nevertheless, through the friction drag with the superfluid current, the normal-fluid current is induced. Moreover, thanks to this friction, the superfluid is separated into the nonviscous and viscous ones. Thus the stationary magnetic response is captured by the three-fluid model. Moreover, because of the normal-fluid and viscous-superfluid currents, the penetration depth is influenced by the scattering. Particularly, by only considering the viscous superfluid current, the dependence of the penetration depth on the mean free path from our theory is exactly the same as the one from Tinkham's discussion [2]. Nevertheless, due to the presences of both the normal-fluid current induced by friction drag and the nonviscous superfluid current, an extension of the penetration depth is proposed.

In addition, when $v_s \geqslant v_L$, a modified Ginzburg-Landau equation is revealed, in which the calculation of the phenomenological parameters is restricted to the pairing region. Furthermore, at $v_s > \omega_D/k_F$, before the superconducting gap is destroyed, we predict an exotic phase, in which the nonviscous superfluid vanishes, leaving only the viscous superfluid and normal fluid. Thus, interestingly, this phase shows finite resistivity but with a finite superconducting gap. Actually, in high-temperature superconductors [111–116] and strongly disordered superconductors [117–120], the phase with both finite resistivity and gap, known as pseudogap phase, has been widely studied. We point out that in the conventional superconductors, the phase with both finite resistivity and gap can also be realized by tuning the magnetic flux.

As for the optical response, the excited superconducting v_s oscillates with time. When $v_s < v_L$, only the nonviscous superfluid is present whereas at $v_s \geqslant v_L$, normal fluid and nonviscous and viscous superfluids are present. We show that in the optical response, the excited normal-fluid current exhibits the Drude-model behavior as it should be, whereas in the superfluid, we find that the superfluid current is excited and it consists of the Meissner supercurrent, which has the same form as that in the magnetic response, as well as the Bogoliubov quasiparticle current. Particularly, at low temperature, few Bogoliubov quasiparticles are excited in the pairing region and hence the Bogoliubov quasiparticle current is marginal. Then the normal-fluid current and the superfluid current, which only consist of the Meissner supercurrent, are exactly the same as those in the original two-fluid model [1,2,5,18,19,21–23,34]. However, friction between the superfluid and normal-fluid currents is present, and due to this friction, the superfluid is separated into the nonviscous and viscous ones. The presence of viscous superfluid suggests that the optical response is also captured by the three-fluid model, whereas the two-fluid model [1,2,5,18,19,21–23,34] in the literature is insufficient for a complete picture. Actually, although the viscous superfluid has been hinted in the stationary magnetic response in the literature [2,50], it has long been overlooked in the optical response.

Based on the three-fluid model, the expression of the optical conductivity is revealed. We also give the expression of the optical excitation of the Higgs mode. By comparing the contributions from the drive and Anderson-pseudospin pump effects, we find that the drive effect is dominant at finite temperature whereas at zero temperature, both effects contribute. Actually, the drive effect in our microscopic theory is related to the kinetic energy in the Ginzburg-Landau theory, in which the vector potential is involved as $(\mathbf{k}_F\cdot\mathbf{A})^2/m^2 = 4\varepsilon_{k_F}\mathbf{A}^2/(2m)$ at finite temperature. Nevertheless, in the pump effect, the vector potential is involved as $\mathbf{A}^2/(2m)$. These two responses of the vector potentials are totally different, and thanks to the large Fermi energy, the drive effect makes the dominant contribution. Consequently, the Liouville [37,38,41] or Bloch [40,42–45,47] equation in the literature with the pump effect alone is insufficient to study the optical excitation of the Higgs mode. However, although the deficiency of the Liouville or Bloch equation has been hinted according to the Ginzburg-Landau theory, it has long been overlooked in the study of the Higgs mode in the literature. Particularly, in the experiments for the detection of Higgs mode [28–30], since the thermal effect is inevitable because of the intense THz field, we believe that the experimentally observed excitation of the Higgs mode is dominated by the drive effect.

Finally, we discuss the charge density in the superconducting state from the dynamic viewpoint. In the superfluid, from the BCS theory, the charge density with momentum $\mathbf{k}$ reads [60,124–127]

$$en_{\mathbf{k}} = e\sum_{\sigma}\langle c^{\dagger}_{\mathbf{k}\sigma}c_{\mathbf{k}\sigma}\rangle = 2ev_k^2 + 2e\frac{\xi_k}{E_k}f(E_k), \tag{62}$$

consisting of the charge densities of the condensate [124–127] $2ev_k^2$ and Bogoliubov quasiparticles [124–129] $2e\frac{\xi_k}{E_k}f(E_k)$, whereas in the normal state one has $en_{\mathbf{k}} = 2ef(\xi_k)$, hence, there exists the charge-density difference between the superconducting and normal states, which is related to the well-known particle-number nonconservation in the BCS theory. Interestingly, we find that this charge-density difference can be compensated by the Meissner-superfluid density $\rho_{m\mathbf{k}}$ [Eq. (29)] as

$$2ef(\xi_k) = 2ev_k^2 + 2e\frac{\xi_k}{E_k}f(E_k) - eC_k\rho_{m\mathbf{k}} + eO(|\Delta|^4), \tag{63}$$

with a prefactor $C_k = D_0\xi_k/(3N_0)$, guaranteeing the charge-density conservation in the superconducting states. As seen from the right-hand side of the above equation, in addition to the condensate and Bogoliubov quasiparticles, the charge density in the superconducting states also consists of the contribution from the Meissner density $\rho_{m\mathbf{k}}$. At zero temperature, as the Bogoliubov quasiparticles, i.e., thermal excitations, vanish, what remain are the condensate from the BCS ground state and the Meissner charge fluctuation on top of the condensate. By noticing that all the electromagnetic responses in superconductors at zero temperature come from the Meissner current, one can draw the conclusion that only the Meissner charge fluctuation contributes to the superconducting response and the condensate simply provides a rigid background. This is in contrast to the previous textbook understanding [4,93,102,130–133] that the supercurrent is a collective motion of the condensate [134].

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APPENDIX A: DERIVATION OF EQ. (18)

In this section, we derive Eq. (18). Specifically, by taking the impurity scattering as the short-range one, i.e., $|V_{\mathbf{k}\mathbf{k}'}|^2 \approx |V_0|^2$, after the integration over the angle in Eq. (17), one obtains

$$\begin{aligned}\partial_t\rho^q_{\mathbf{k}}\big|_{\rm sc} = &-n_i\pi D_0|V_0|^2\int\frac{d\xi_{k'}}{k_Fv_s}(1-\eta_{kk'})\begin{pmatrix}\big(\rho^q_{\mathbf{k},11}-\rho^q_{\mathbf{k}',11}\big)\big|_{\cos\theta_{\mathbf{k}'}=\frac{E^+_{\mathbf{k}}-E_{k'}}{k_Fv_s}} & 0\\ 0 & \big(\rho^q_{\mathbf{k},22}-\rho^q_{\mathbf{k}',22}\big)\big|_{\cos\theta_{\mathbf{k}'}=\frac{E^-_{\mathbf{k}}+E_{k'}}{k_Fv_s}}\end{pmatrix}\\ &-n_i\pi D_0|V_0|^2\int\frac{d\xi_{k'}}{k_Fv_s}(1+\eta_{kk'})\begin{pmatrix}\big(\rho^q_{\mathbf{k},11}-\rho^q_{\mathbf{k}',22}\big)\big|_{\cos\theta_{\mathbf{k}'}=\frac{E^+_{\mathbf{k}}+E_{k'}}{k_Fv_s}} & 0\\ 0 & \big(\rho^q_{\mathbf{k},22}-\rho^q_{\mathbf{k}',11}\big)\big|_{\cos\theta_{\mathbf{k}'}=\frac{E^-_{\mathbf{k}}-E_{k'}}{k_Fv_s}}\end{pmatrix}.\end{aligned} \tag{A1}$$

Thanks to the large Fermi energy, we approximately take the emergence of the scattering around the Fermi surface by setting $|\xi_k|, |\xi_{k'}| < E_c$ in Eq. (A1). Then, one has $|\xi_k| - E_c < \xi_{k'} < |\xi_k| + E_c$. By using the mean value theorem for integrals, Eq. (A1) becomes

$$\partial_t \rho^q_{\mathbf{k}}\big|_{\rm sc} = -2n_i\pi D_0|V_0|^2\lambda_c\frac{\xi_k(\xi_k+\xi_{k'})}{E_k^2}\begin{pmatrix}\big(\rho^q_{\mathbf{k},11}-\rho^q_{\mathbf{k}',11}\big)\big|^{|\xi_k|=|\xi_{k'}|}_{\cos\theta_{\mathbf{k}'}=\cos\theta_{\mathbf{k}}} & 0\\ 0 & \big(\rho^q_{\mathbf{k},22}-\rho^q_{\mathbf{k}',22}\big)\big|^{|\xi_k|=|\xi_{k'}|}_{\cos\theta_{\mathbf{k}'}=\cos\theta_{\mathbf{k}}}\end{pmatrix}$$
$$-2n_i\pi D_0|V_0|^2\lambda_c\left(1+\frac{|\Delta|^2}{E_k^2}\right)\begin{pmatrix}\big(\rho^q_{\mathbf{k},11}-\rho^q_{\mathbf{k}',22}\big)\big|^{|\xi_k|=|\xi_{k'}|}_{\cos\theta_{\mathbf{k}'}-\cos\theta_{\mathbf{k}}=\frac{2E_k}{k_Fv_s}} & 0\\ 0 & \big(\rho^q_{\mathbf{k},22}-\rho^q_{\mathbf{k}',11}\big)\big|^{|\xi_k|=|\xi_{k'}|}_{\cos\theta_{\mathbf{k}'}-\cos\theta_{\mathbf{k}}=-\frac{2E_k}{k_Fv_s}}\end{pmatrix}, \quad \text{(A2)}$$

with the dimensionless parameter $\lambda_c = 2E_c/(k_F v_s)$. It is noted that the first term on the right-hand side of Eq. (A2) is zero as a consequence of the particle-hole symmetry under the particle-hole transformation [135] $\xi_k \to -\xi_k$. Then, Eq. (18) is obtained.

APPENDIX B: DERIVATION OF EQ. (22)

We derive Eq. (22) in this part. Considering the large Fermi energy in conventional superconductors, one can neglect $\varepsilon_{\mathbf{p}-2e\mathbf{A}}$ on the left-hand side of Eq. (20). Then, by using Eq. (20) to substitute $\rho_{\mathbf{k}\pm}$ in Eq. (21), one has

$$\frac{\mathbf{k}}{m}\cdot\nabla\rho_{\mathbf{k}0} = \frac{\nabla|\Delta|^2\cdot\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}0}}{2\xi_k}+\frac{(\nabla|\Delta|^2\cdot\boldsymbol{\partial}_{\mathbf{k}})(\mathbf{p}_s\cdot\boldsymbol{\partial}_{\mathbf{k}})\rho_{\mathbf{k}3}}{2\xi_k}-\frac{(\nabla|\Delta|^2\cdot\boldsymbol{\partial}_{\mathbf{k}})(\mathbf{p}_s\cdot\boldsymbol{\partial}_{\mathbf{k}})(\rho_{\mathbf{k}3}/\xi_k)}{2}+\big\{\partial_t\rho_{\mathbf{k}}\big|_{\rm sc}\big\}_3. \quad \text{(B1)}$$

In the quasiparticle space, Eq. (B1) becomes

$$\frac{\mathbf{k}}{m}\cdot\nabla\rho^q_{\mathbf{k}0} = \nabla|\Delta|^2\cdot\left[\frac{\boldsymbol{\partial}_{\mathbf{k}}\rho^q_{\mathbf{k}0}}{2\xi_k}+\frac{\boldsymbol{\partial}_{\mathbf{k}}(\mathbf{p}_s\cdot\boldsymbol{\partial}_{\mathbf{k}})}{2\xi_k}\left(\frac{\xi_k\rho^q_{\mathbf{k}3}}{E_k}\right)-\frac{\boldsymbol{\partial}_{\mathbf{k}}(\mathbf{p}_s\cdot\boldsymbol{\partial}_{\mathbf{k}})(\rho^q_{\mathbf{k}3}/E_k)}{2}\right]+\frac{\xi_k}{E_k}\big\{\partial_t\rho^q_{\mathbf{k}}\big|_{\rm sc}\big\}_3. \quad \text{(B2)}$$

In the presence of a superconducting momentum $\mathbf{p}_s = -e\mathbf{A}+\frac{1}{2}\nabla_{\mathbf{R}}\psi$, i.e., the center-of-mass momentum, the superconducting state behaves like the FFLO-like state [100–106]. Consequently, at the weak-scattering limit, the solution of density matrix reads

$$\rho^q_{\mathbf{k}} = \rho^{q0}_{\mathbf{k}}+\delta\rho^q_{\mathbf{k}}+\delta\rho^{qs}_{\mathbf{k}}, \quad \text{(B3)}$$

with

$$\rho^{q0}_{\mathbf{k}} = \begin{pmatrix} f(E^+_{\mathbf{k}}) & 0\\ 0 & f(E^-_{\mathbf{k}})\end{pmatrix}. \quad \text{(B4)}$$

Here, $\rho^{q0}_{\mathbf{k}}$ is the quasiparticle distribution of the FFLO-like state with $E^\pm_{\mathbf{k}} = \mathbf{k}\cdot\mathbf{v}_s\pm E_k$ and $\mathbf{v}_s = \mathbf{p}_s/m$; $\delta\rho^q_{\mathbf{k}}$ denotes the disturbance from the FFLO-like state in the magnetic response in the absence of the scattering; $\delta\rho^{qs}_{\mathbf{k}}$ represents the scattering contribution. By substituting Eq. (B3) into Eq. (B2), one can construct $\delta\rho^q_{\mathbf{k}}$ as

$$\delta\rho^q_{\mathbf{k}} = (\mathbf{k}\cdot\mathbf{v}_s)\begin{pmatrix} a^+_{\mathbf{k}} & 0\\ 0 & a^-_{\mathbf{k}}\end{pmatrix}+(\mathbf{k}\cdot\mathbf{v}_s)|\Delta|^2\begin{pmatrix} m^+_{\mathbf{k}} & 0\\ 0 & m^-_{\mathbf{k}}\end{pmatrix}+\frac{(\mathbf{k}\cdot\mathbf{v}_s)^2}{2}\begin{pmatrix} b^+_{\mathbf{k}} & 0\\ 0 & b^-_{\mathbf{k}}\end{pmatrix}. \quad \text{(B5)}$$

Then, Eq. (B2) becomes

$$\frac{\mathbf{k}\cdot\nabla|\boldsymbol{\Delta}|^2}{m}\left[(\mathbf{k}\cdot\mathbf{v}_s)\frac{m^+_{\mathbf{k}}+m^-_{\mathbf{k}}}{2}+\frac{\partial_{E_k}\rho^q_{k0}}{2E_k}\right] = \frac{\mathbf{v}_s\cdot\nabla|\Delta|^2}{2\xi_k}\left[\partial_{E_k}\rho^{q0}_{k3}+\frac{a^+_{\mathbf{k}}+a^-_{\mathbf{k}}}{2}+(\mathbf{k}\cdot\mathbf{v}_s)\partial_{E_k}\left(\frac{a^+_{\mathbf{k}}-a^-_{\mathbf{k}}}{2}\right)+(\mathbf{k}\cdot\mathbf{v}_s)\frac{b^+_{\mathbf{k}}+b^-_{\mathbf{k}}}{2}\right]$$
$$+\frac{\mathbf{k}}{m}\cdot\nabla|\Delta|^2\left[\frac{\partial_{E_k}\rho^{q0}_{k0}}{2E_k}+\frac{(\mathbf{k}\cdot\mathbf{v}_s)}{E_k}\partial_{E_k}\left(\frac{\rho^{q0}_{k3}}{E_k}\right)+\frac{\varepsilon_{p_s}}{\xi_k}\frac{\partial_{E_k}\rho^{q0}_{k0}}{E_k}\right]+\frac{\mathbf{v}_s\cdot\nabla|\Delta|^2}{2\xi_k}\frac{(\mathbf{k}\cdot\mathbf{v}_s)}{E_k}\partial_{E_k}\rho^{q0}_{k0}+\frac{\rho^q_{k3}}{4\xi_kE_k\varepsilon_k}(\mathbf{k}\cdot\mathbf{v}_s)\frac{\mathbf{k}}{m}\cdot\nabla|\Delta|^2, \quad \text{(B6)}$$

in which we have neglected the terms higher than the second order of $|\Delta|$ or $(\mathbf{k}\cdot\mathbf{v}_s)$.

Considering the large Fermi energy, one can neglect ε_{p_s} term in Eq. (B6) and obtains

$$\frac{\mathbf{k}\cdot\nabla|\boldsymbol{\Delta}|^2}{m}(\mathbf{k}\cdot\mathbf{v}_s)\left[\frac{m^+_{\mathbf{k}}+m^-_{\mathbf{k}}}{2}-\frac{\partial_{E_k}}{E_k}\left(\frac{\rho^{q0}_{k3}}{E_k}\right)-\frac{\rho^q_{k3}}{\xi_kE_k}\frac{1}{4\varepsilon_k}\right]-\frac{\mathbf{v}_s\cdot\nabla|\Delta|^2}{2\xi_k}\left\{\partial_{E_k}\rho^{q0}_{k3}+\frac{a^+_{\mathbf{k}}+a^-_{\mathbf{k}}}{2}+(\mathbf{k}\cdot\mathbf{v}_s)\right.$$
$$\left.\times\left[\partial_{E_k}\left(\frac{a^+_{\mathbf{k}}-a^-_{\mathbf{k}}}{2}\right)+\frac{b^+_{\mathbf{k}}+b^-_{\mathbf{k}}}{2}+\frac{\partial_{E_k}\rho^{q0}_{k0}}{E_k}\right]\right\} = 0. \quad \text{(B7)}$$

It is noted that Eq. (B7) holds in the entire momentum space. Consequently, one has

$$a^\pm_{\mathbf{k}} = \mp\partial_{E_k}f(E^\pm_{\mathbf{k}}), \quad \text{(B8)}$$

$$b^\pm_{\mathbf{k}} = \partial^2_{E_k}f(E^\pm_{\mathbf{k}})+\frac{\partial_{E_k}f(E^\pm_{\mathbf{k}})}{E_k}, \quad \text{(B9)}$$

$$m^\pm_{\mathbf{k}} = \pm\left[\frac{1}{E_k}\partial_{E_k}+\frac{1}{4\xi_k\varepsilon_k}\right]\frac{f(E^\pm_{\mathbf{k}})}{E_k}. \quad \text{(B10)}$$

As for the scattering contribution $\delta\rho_{\mathbf{k}}^{qs}$, one has

$$\frac{\mathbf{k}\cdot\nabla}{m}\delta\rho_{\mathbf{k}0}^{qs} = \frac{\xi_k}{E_k}\{\partial_t\rho_{\mathbf{k}}^{q}|_{\rm sc}\}_3. \tag{B11}$$

In the weak-scattering limit, the scattering only causes the momentum (current) relaxation. Therefore, by keeping the linear-order terms of $(\mathbf{k}\cdot\mathbf{v}_s)$ in $\rho_{\mathbf{k}}^{q}$, from Eq. (18), Eq. (B11) becomes

$$\frac{\mathbf{k}\cdot\nabla}{m}\delta\rho_{\mathbf{k}0}^{qs} = -\theta\left(\frac{kv_s}{E_k}\right)\frac{\xi_k}{E_k\tau_k}|\Delta|^2(\mathbf{k}\cdot\mathbf{v_s})\frac{m_{\mathbf{k}}^{+}-m_{\mathbf{k}}^{-}}{2}, \tag{B12}$$

from which, one obtains

$$\delta\rho_{\mathbf{k}}^{qs} = |\Delta|^2(\mathbf{k}\cdot\mathbf{v_s})\begin{pmatrix}\delta m_{\mathbf{k}}^{+} & 0\\ 0 & \delta m_{\mathbf{k}}^{-}\end{pmatrix}, \tag{B13}$$

with $\delta m_{\mathbf{k}}^{\pm} = \mp\frac{\xi}{\tau_k v_F}\theta(\frac{kv_s}{E_k})\frac{\xi_k}{E_k}m_{\mathbf{k}}^{\pm}$. Here, we have used $|\Delta|^2/(\nabla|\Delta|^2) = \xi$. Consequently, Eq. (22) is obtained.

APPENDIX C: DERIVATION OF GINZBURG-LANDAU EQUATION

In this part, we derive the Ginzburg-Landau equation. By using Eq. (20) to substitute $\rho_{\mathbf{k}+}$ into the gap equation [Eq. (9)], one has

$$-\frac{\Delta}{V} = \sum_{\mathbf{k}}{}'\left[\frac{\rho_{\mathbf{k}3}}{\xi_k} - \frac{i\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}0}\cdot(\nabla-2ie\mathbf{A})}{2\xi_k} - \frac{\varepsilon_{\mathbf{p}-2e\mathbf{A}}}{4\xi_k\Delta}\rho_{\mathbf{k}+} - \frac{\boldsymbol{\partial}_{\mathbf{k}}\boldsymbol{\partial}_{\mathbf{k}}\rho_{\mathbf{k}3}:(\nabla-2ie\mathbf{A})(\nabla-2ie\mathbf{A})}{8\xi_k}\right]\Delta. \tag{C1}$$

At $v_s < v_L$ with only the nonviscous superfluid, the superconducting state behaves like the BCS one. Consequently, the density matrix in the quasiparticle space reads

$$\rho_{\mathbf{k}}^{q} = \begin{pmatrix} f(E_k) & 0\\ 0 & 1-f(E_k)\end{pmatrix}. \tag{C2}$$

With this BCS-state density matrix in the quasiparticle space, by treating Δ as a small quantity near the critical temperature, Eq. (C1) becomes

$$\begin{aligned}\frac{\Delta}{D_0V} &= \int_{-\omega_D}^{\omega_D} d\xi_k\left[\partial_k^2\left(\frac{\xi_k\rho_{\mathbf{k}3}^{q}}{E_k}\right)\frac{(\nabla-2ie\mathbf{A})^2}{24\xi_k} - \frac{\rho_{\mathbf{k}3}^{q}}{E_k}\right]\Delta \approx \int_{-\omega_D}^{\omega_D} d\xi_k\left\{\frac{1-2f(|\xi_k|)}{2|\xi_k|} + \frac{|\Delta|^2\partial_{|\xi_k|}}{2|\xi_k|}\left[\frac{1-2f(|\xi_k|)}{2|\xi_k|}\right]\right.\\ &\quad\left.+\left(\frac{2\partial_{E_k}}{E_k}+\partial_{E_k}^2\right)\left(\frac{2f(E_k)-1}{2E_k}\right)\frac{k_F^2}{2m}\frac{(\nabla-2ie\mathbf{A})^2}{12m}\right\}\Delta.\end{aligned} \tag{C3}$$

With $\frac{1}{D_0V} = \ln(\frac{2\gamma}{\pi}\frac{\omega_D}{T_c})$ in the BCS theory [48], from Eq. (C3), one obtains

$$\left\{\frac{(\nabla-2ie\mathbf{A})^2}{4m} + \frac{1}{\lambda}[\alpha-\beta|\Delta|^2]\right\}\Delta = 0, \tag{C4}$$

with

$$\alpha = \int_{-\omega_D}^{\omega_D} d\xi_k\frac{1-2f(|\xi_k|)}{2|\xi_k|} - \frac{1}{D_0V} = \ln\left(\frac{2\gamma}{\pi}\frac{\omega_D}{T}\right) - \ln\left(\frac{2\gamma}{\pi}\frac{\omega_D}{T_c}\right) = \ln\left(\frac{T}{T_c}\right), \tag{C5}$$

$$\beta = \int_{-\omega_D}^{\omega_D} d\xi_k\frac{1}{2|\xi_k|}\partial_{|\xi_k|}\left[\frac{2f(|\xi_k|)-1}{2|\xi_k|}\right] = T\sum_n\int_{-\omega_D}^{\omega_D} d\xi_k\frac{1}{2|\xi_k|}\partial_{|\xi_k|}\left[\frac{1}{(i\omega_n)^2-\xi_k^2}\right] \approx T\sum_n\int_{-\infty}^{\infty} d\xi_k\frac{1}{\left[(\omega_n)^2+\xi_k^2\right]^2} = \frac{7R(3)}{8(\pi T)^2}, \tag{C6}$$

$$\begin{aligned}\lambda &= \frac{\varepsilon_{k_F}}{3}\int_{-\omega_D}^{\omega_D} d\xi_k\left(\frac{2}{E_k}+\partial_{E_k}\right)\partial_{E_k}\left(\frac{2f(E_k)-1}{2E_k}\right) \approx \frac{\varepsilon_{k_F}}{3}\int_{-\infty}^{\infty} d\xi_k\frac{\partial_{|\xi_k|}^2 f(|\xi_k|)}{|\xi_k|} = T\sum_n\frac{2\varepsilon_{k_F}}{3}\int_{-\infty}^{\infty} d\xi_k\frac{1}{(i\omega_n-|\xi_k|)^3|\xi_k|}\\ &= T\sum_{n>0}\frac{4\varepsilon_{k_F}}{3}\int_{-\infty}^{\infty} d\xi_k\frac{3\omega_n^2-\xi_k^2}{(\xi_k^2+\omega_n^2)^3} = \varepsilon_{k_F}\frac{7R(3)}{6(\pi T)^2}.\end{aligned} \tag{C7}$$

Here, $\omega_n = (2n+1)\pi T$ represents the Matsubara frequency [54]. Consequently, the Ginzburg-Landau equation [12,53,54] is exactly derived in Eq. (C4).

APPENDIX D: DERIVATION OF EQ. (46)

We give the derivation of Eq. (46) in this section. Following the derivation of the density matrix in the magnetic response, in the optical response, at the weak-scattering limit, the solution of density matrix reads

$$\rho^q_{\mathbf{k}} = \rho^{q0}_{\mathbf{k}} + \delta\rho^q_{\mathbf{k}} + \delta\rho^{qs}_{\mathbf{k}}, \tag{D1}$$

with

$$\rho^{q0}_{\mathbf{k}} = \begin{pmatrix} f(E^+_{\mathbf{k}}) & 0 \\ 0 & f(E^-_{\mathbf{k}}) \end{pmatrix}. \tag{D2}$$

Here, $\rho^{q0}_{\mathbf{k}}$ is the quasiparticle distribution of the FFLO-like state with $E^{\pm}_{\mathbf{k}} = \mathbf{k}\cdot\mathbf{v}_s \pm E_k$; $\delta\rho^q_{\mathbf{k}}$ denotes the disturbance from the FFLO-like state in the optical response in the absence of the scattering; $\delta\rho^{qs}_{\mathbf{k}}$ represents the scattering contribution.

In Eq. (45), one has $t_3 = (u_k^2 - v_k^2)\tau_3 - 2u_kv_k\tau_1$, $t_2 = \tau_2$, $t_1 = (u_k^2 - v_k^2)\tau_1 + 2u_kv_k\tau_3$, and $U_k^\dagger \boldsymbol{\partial}_{\mathbf{k}} U_k = \frac{i}{2}\frac{\mathbf{k}}{m}\frac{|\Delta|}{E_k^2}\tau_2$ as well as $U_k^\dagger \boldsymbol{\partial}_{\mathbf{k}}\boldsymbol{\partial}_{\mathbf{k}} U_k = -\frac{\mathbf{k}}{m}\frac{\mathbf{k}}{m}(\frac{|\Delta|^2}{4E_k^4}\tau_0 - ib_k\tau_2)$ with $b_k = \frac{\Delta}{4\varepsilon_k E_k^2} - \frac{\xi_k\Delta}{E_k^4}$. As revealed in the previous work [60–62], in the optical response, the effective chemical potential $\mu_{\rm eff}$, determined from the charge neutrality condition [60], is excited and then involved in the kinetic equation as a feedback. Considering the large Fermi energy in conventional superconductors, for a relatively weak optical field, we neglect the feedback of $\mu_{\rm eff}$ in Eq. (45). Then, by substituting the density matrix [Eq. (B1)] into Eq. (45), one can construct $\delta\rho^q_{\mathbf{k}}$ as

$$\delta\rho^q_{\mathbf{k}} = -(\mathbf{k}\cdot\mathbf{v}_s)\rho^{q1}_{\mathbf{k}} + \frac{(\mathbf{k}\cdot\mathbf{v}_s)^2}{2}\rho^{q2}_{\mathbf{k}} + mv_s^2\rho^{q3}_{\mathbf{k}}, \tag{D3}$$

from which, the linear-order terms of $(\mathbf{k}\cdot\mathbf{v}_s)$ in Eq. (45) become

$$i(\mathbf{k}\cdot\mathbf{v}_s)\big(\omega\rho^{q1}_{\mathbf{k}} + 2E_k\rho^{q1}_{\mathbf{k}+}\tau_+ - 2E_k\rho^{q1}_{\mathbf{k}-}\tau_-\big) = e(\mathbf{k}\cdot\mathbf{E})\frac{|\Delta|^2}{E_k^2}\left(\frac{1}{E_k} - \partial_{E_k}\right)\rho^{q0}_{\mathbf{k}3}\tau_0 + \mathbf{k}\cdot\left(i\omega\mathbf{v}_s + \frac{e\mathbf{E}}{m}\right)\partial_{E_k}\rho^{q0}_{\mathbf{k}3}\tau_0. \tag{D4}$$

Here, we have neglected the $\partial_{E_k}\rho^{q0}_{\mathbf{k}0}$ term, which is zero in either pairing or unpairing regions. From Eq. (D4), one has $\mathbf{v}_s = -\frac{e\mathbf{E}}{i\omega m}$ and $\rho^{q1}_{\mathbf{k}} = \frac{\rho_{m\mathbf{k}}\tau_0}{4\varepsilon_{k_F}}$.

By using $e\mathbf{E}/m = -i\omega\mathbf{v_s}$, the nonlinear-order terms of $(\mathbf{k}\cdot\mathbf{v}_s)^2$ in Eq. (45) reads

$$\begin{aligned} i(\mathbf{k}\cdot\mathbf{v}_s)^2\big(\omega\rho^{q2}_{\mathbf{k}} + E_k\rho^{q2}_{\mathbf{k}+}\tau_+ - E_k\rho^{q2}_{\mathbf{k}-}\tau_-\big) = {}& i(\mathbf{k}\cdot\mathbf{v}_s)^2\sum_{j=\pm}\bigg[\omega\frac{\xi_k^2}{2E_k^2}\partial_{E_k}\rho^{q1}_{\mathbf{k}0}\tau_3 - (\omega + 2jE_k)\frac{\xi_k|\Delta|}{E_k^2}\partial_{E_k}\rho^{q1}_{\mathbf{k}0}\tau_j \\ &+ j\tau_j\frac{|\Delta|}{E_k^2}\left(\xi_k^2\partial^2_{E_k}\rho^{q0}_{\mathbf{k}3} - 3\xi_k\rho^{q1}_{\mathbf{k}0} + E_k^2\frac{\partial_{E_k}\rho^{q0}_{\mathbf{k}3} + \rho^{q1}_{\mathbf{k}0}}{2\varepsilon_k}\right)\bigg], \end{aligned} \tag{D5}$$

from which one has

$$\rho^{q2}_{\mathbf{k}} = \frac{\xi_k^2}{E_k^2}\partial_{E_k}\rho^{q1}_{\mathbf{k}0}\tau_3 - \sum_{j=\pm}\left[\frac{\omega + 2jE_k}{\omega + jE_k}\frac{\xi_k|\Delta|}{E_k^2}\partial_{E_k}\rho^{q1}_{\mathbf{k}0}\tau_j + \frac{j|\Delta|\tau_j}{\omega + jE_k}\left(\frac{\xi_k^2\partial^2_{E_k}\rho^{q0}_{\mathbf{k}3} - 3\xi_k\rho^{q1}_{\mathbf{k}0}}{E_k^2} + \frac{\partial_{E_k}\rho^{q0}_{\mathbf{k}3} + \rho^{q1}_{\mathbf{k}0}}{2\varepsilon_k}\right)\right]. \tag{D6}$$

For the nonlinear-order terms of v_s^2 in Eq. (45), one obtains

$$2iv_s^2\big(\omega\rho^{q3}_{\mathbf{k}} + E_k\rho^{q3}_{\mathbf{k}+}\tau_+ - E_k\rho^{q3}_{\mathbf{k}-}\tau_-\big) = v_s^2\left[\frac{|\Delta|}{E_k}\rho^{q0}_{\mathbf{k}3}\tau_2 + i\omega\left(\frac{\xi_k}{E_k}\tau_3 - \frac{|\Delta|}{E_k}\tau_1\right)\rho^{q1}_{\mathbf{k}0} + 2\Delta\rho^{q1}_{\mathbf{k}0}\tau_2\right], \tag{D7}$$

from which, $\rho^{q3}_{\mathbf{k}}$ reads

$$\rho^{q3}_{\mathbf{k}} = \frac{\xi_k}{2E_k}\rho^{q1}_{\mathbf{k}0}\tau_3 - \frac{|\Delta|}{2E_k}\sum_{j=\pm}\frac{\big[j\rho^{q0}_{\mathbf{k}3} + \rho^{q1}_{\mathbf{k}0}(\omega + 2jE_k)\big]\tau_j}{\omega + jE_k}. \tag{D8}$$

As for the scattering contribution $\delta\rho^{qs}_{\mathbf{k}}$, one has

$$\partial_T\delta\rho^{qs}_{\mathbf{k}} = \big\{\partial_t\rho^q_{\mathbf{k}}\big|_{\rm sc}\big\}. \tag{D9}$$

In the weak-scattering limit, the scattering only causes the momentum (current) relaxation. Therefore, by keeping the linear-order terms of $(\mathbf{k}\cdot\mathbf{v}_s)$ in $\rho^q_{\mathbf{k}}$, from Eq. (18), Eq. (D9) becomes

$$\partial_T\delta\rho^{qs}_{\mathbf{k}0} = -\frac{(\mathbf{k}\cdot\mathbf{v}_s)}{\tau_k}\theta\left(\frac{kv_s}{E_k}\right)(\partial_{E_k}\rho^{q0}_{\mathbf{k}3} + \hat{O}_kf_{\mathbf{k}}), \tag{D10}$$

where $\hat{O}_k = 4u_k^2 v_k^2 (1/E_k - \partial_{E_k})$ and $f_{\mathbf{k}} = [3f(E_{\mathbf{k}}^+) - 3f(E_{\mathbf{k}}^-) - f(E_{\mathbf{k}}^+ + 2E_k) + f(E_{\mathbf{k}}^- - 2E_k)]/8$. Thus $\delta\rho_{\mathbf{k}}^{qs}$ is obtained as

$$\delta\rho_{\mathbf{k}0}^{qs} = -\frac{(\mathbf{k}\cdot\mathbf{v}_s)}{i\omega\tau_k}\theta\left(\frac{kv_s}{E_k}\right)(\partial_{E_k}\rho_{\mathbf{k}3}^{q0} + \hat{O}_k f_{\mathbf{k}})\tau_0. \tag{D11}$$

Consequently, Eq. (46) is obtained.

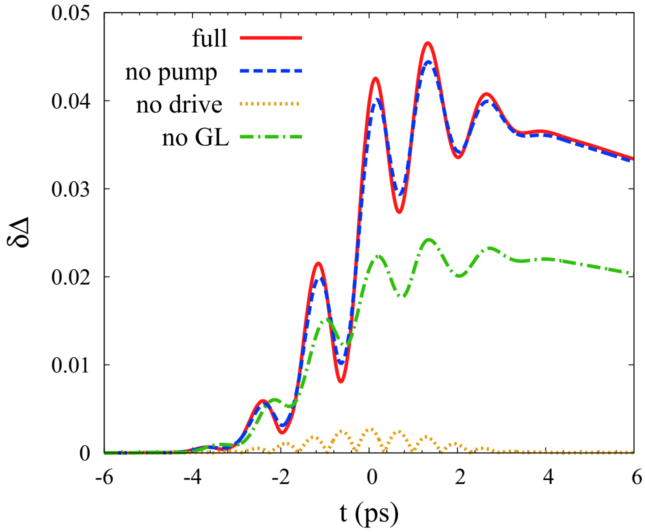

FIG. 5. Temporal evolutions of the Higgs mode from the numerical calculation of Eq. (44). Blue dashed curve: without the pump effect by setting $p_s^2/(2m) = 0$ in Eq. (44). Brown dotted curve: without the drive effect by setting $\partial_{\mathbf{k}}\rho_{\mathbf{k}} = 0$ in Eq. (44). Green chain curve: without the Ginzburg-Landau kinetic effect by removing the last two terms on the left-hand side of Eq. (44). In the calculation, we used a THz linear-polarized optical pulse: $\mathbf{p_s} = (e/\omega)E_0\mathbf{e_x}\sin(\omega t)\exp[-t^2/(2\sigma_t^2)]$ and $\partial_t\mathbf{p_s} = eE_0\mathbf{e_x}\cos(\omega t)\exp[-t^2/(2\sigma_t^2)]$ with σ_t being the width of the optical pulse. The used parameter [60] in the calculation includes $m = 0.067m_e$, $\Delta_0 = 0.8$ meV, $E_0 = 0.1$ kv/cm, $\sigma_t = 2$ ps, $n_i = 0.2N_0$, $\kappa = 12.9$, $N_0 = 5\times10^{11}$ cm^{-2}, $V = 0.1788$ eV nm^3, $\omega = 2\Delta_0$, and $T = 0.02$ K. For comparison, we take the same scattering terms as those in the previous theory by Yu and Wu [60].

APPENDIX E: COMPARISON BETWEEN DRIVE AND PUMP EFFECTS

We compare the drive and pump effects in the excitation of the Higgs mode by performing a numerical calculation of Eq. (44) in the presence of a THz linear-polarized optical pulse. As seen from the numerical results plotted in Fig. 5, a plateau of the superconducting gap is observed after a THz pulse as a consequence of the thermal effect and the excitation of the Higgs mode (red solid curve) is dominated by the drive effect (blue dashed curve) whereas the pump effect (brown dotted curve) is marginal, in consistency with our analytical analysis in Sec. III D 3.

In addition, we also calculate the case without the Ginzburg-Landau kinetic effect [last two terms on the left-hand side of Eq. (44)], which exactly reduces to the previous theory by Yu and Wu [60]. As seen from Fig. 5, in comparison to the full results (red solid curve), the absence of the Ginzburg-Landau kinetic effect, represented by green chain curve, leads to a quantitative reduction in the excitation of the Higgs mode. In order to compare the excitation of the Higgs mode between our theory and Ref. [60], we separate the drive effect as

$$\delta\Delta^{\text{drive}} = \delta\Delta^{\text{drive}}_{\text{no GL}} + \delta\Delta^{\text{drive}}_{\text{GL}}, \tag{E1}$$

with $\delta\Delta^{\text{drive}}_{\text{no GL}}$ denoting the pure drive effect [from forth term on the left-hand side of Eq. (44)] in the absence of the Ginzburg-Landau kinetic effect and $\delta\Delta^{\text{drive}}_{\text{GL}}$ representing the contribution exactly from the Ginzburg-Landau kinetic effect. By using the same technique in Appendix D to derive the nonlinear response, one has

$$\begin{aligned}\frac{\delta\Delta^{\text{drive}}_{\text{GL}}}{\Delta_0 V} = \varepsilon_{p_s}\sum_{\mathbf{k}\in P}\Bigg\{&\Bigg[2\varepsilon_{k_F}\frac{\xi_k^2\cos^2\theta_{\mathbf{k}}\partial_{E_k}^2 f(E_k)}{E_k(\omega^2-E_k^2)}\\ &\times\left(1-\frac{\omega|\Delta_0|^2}{E_k^3}\right) + a_k\frac{|\Delta|^2}{E_k^3}\frac{E_k\cos^2\theta_{\mathbf{k}}-\omega}{\omega^2-E_k^2}\Bigg]\\ &-2\left[\frac{\varepsilon_{k_F}\cos\theta_{\mathbf{k}}^2\xi_k^2}{E_k(\omega^2-E_k^2)}\partial_{E_k} - \frac{\omega}{\omega^2-E_k^2}\right]\rho_{\mathbf{k}0}^{q1}\Bigg\},\end{aligned} \tag{E2}$$

$$\frac{\delta\Delta^{\text{drive}}_{\text{no GL}}}{\Delta_0 V} = \varepsilon_{p_s}\sum_{\mathbf{k}\in P}2\left[\frac{\varepsilon_{k_F}\cos\theta_{\mathbf{k}}^2\xi_k^2}{E_k(\omega^2-E_k^2)}\partial_{E_k} - \frac{\omega}{\omega^2-E_k^2}\right]\rho_{\mathbf{k}0}^{q1}. \tag{E3}$$

As seen from above, in the complete contribution $\delta\Delta^{\text{drive}}$ [Eq. (E1)], the pure drive effect $\delta\Delta^{\text{drive}}_{\text{no GL}}$ in the previous work [60] is canceled by the second term in $\delta\Delta^{\text{drive}}_{\text{GL}}$ [Eq. (E2)], leaving only the contribution from the first term in $\delta\Delta^{\text{drive}}_{\text{GL}}$ [Eq. (E2)].

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