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Electronic Raman scattering of two-band superconductors: a time-dependent Landau–Ginzburg theory approach

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Abstract
Electronic Raman scattering of two-band superconductors is studied based on the time-dependent Landau–Ginzburg theory. The focus is on the possible features of the π phase shift between the two superconducting order parameters which may be realized in the Fe-pnictides. The Raman response was computed up to the Gaussian fluctuations in the functional integral formalism including the long range Coulomb interaction with the four channels of symmetric and antisymmetric combinations of the phases and amplitudes of the two order parameters. The Raman spectra is found to be composed of the quasiparticle and the phase collective mode contributions without mixing between them. The contributions from the quasiparticle and the symmetric phase collective mode (the Anderson–Bogolyubov mode) are similar to the two-band superconductors without the π phase shift. The antisymmetric phase mode (the Leggett mode) originates from the fluctuations of the relative phase of the two order parameters. It lies between twice the smaller gap and twice the larger gap and is damped by the quasiparticles. However, this mode is eliminated by the long range Coulomb interaction in the zero-wavenumber limit.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Electronic Raman scattering is a very useful probe in studying the symmetry of the order parameter and the excitations (in the zero-wavenumber limit) of superconductors [1]. It plays an especially important role in the study of unconventional and exotic superconductors owing to its strong dependence on the symmetry of superconducting order parameters. It has been instrumental in understanding and clarifying various excitations in the cuprate high temperature superconductors [2]. Currently the iron-based pnictide superconductors are being studied very actively with a prospect of opening a key window to understanding the mechanism of high temperature superconductivity. A leading proposal for the orbital pairing symmetry is the sign-reversed full-gap state [3, 4]. It is the ground state of a two-band superconductor where both pairing order parameters, Δ_1 and Δ_2, on the two bands have full gaps while acquiring the π phase shift between them. A repulsive interband interaction is turned to induce pairing by generating the sign reversal between the two order parameters. It seems to be able to explain the experimental observations indicating the full gap as well as a gap with nodes [5, 6].

In this paper, the Raman response of the $s_\pm$ or $s\pi$ pairing state is studied in the framework of time-dependent Landau–Ginzburg theory. This is because the physical properties, in particular the low energy excitations in the ordered state (the superconducting state), are most clearly described and also understood in terms of order parameters. The thermodynamic

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properties are very succinctly expressed by the free energy of the time-independent (static) configuration of the order parameters. This free energy can be derived from the microscopic BCS theory [7].

To understand the Raman response of superconductors in terms of the order parameters, we have to consider the dynamics, namely the time dependence of the order parameters. This time-dependent Landau–Ginzburg theory has been widely used in the context of the quantum phase transition [8]. Reference [8] employed the method of the functional integral, and the partial reason for this is that the order parameters exhibit themselves manifestly from the early stage of development. We adapt the approach of [8] to the problems of two-band superconductors.

The excitations of the *one*-band superconductor consist of the quasiparticle excitation and the collective excitation. In the case of the conventional s-wave superconductors, these excitations appear in the Raman response at energy $\omega = 2\Delta$ (where Δ is the superconducting gap) and $\omega = v_s q$ (v_s is a velocity), respectively. The collective excitation is a soft sound-like wave mode, and is often called the Anderson–Bogolyubov mode [9, 10]. However, the long range Coulomb interaction lifts the Anderson–Bogolyubov mode to plasmon excitation of a finite energy at zero wavenumber [11].

In the case of *two*-band superconductors, in addition to the excitations mentioned above, there can be other excitations due to the presence of more degrees of freedom. In particular, Leggett suggested a collective mode which stems from the *out-of-phase* oscillation of two phases of two superconducting order parameters with energy below twice the smaller gap energy [12]. The Leggett mode was reported to be observed in a two-band MgB_2 superconductor [13]. We note a few theoretical works on the Raman response of Fe-pnictides [14, 15]. The work by Chubukov *et al* [14] considered the response in the A_{1g} geometry and the existence of a resonance peak below twice the gap for the pairing gap is claimed. The long range Coulomb interaction is argued not to contribute to the Raman response in A_{1g} geometry if particle–hole symmetry is present. The work by Boyd *et al* [15] mainly focused on the case with the anisotropic gap amplitudes and considered only the quasiparticle contributions dressed by the long range Coulomb interaction.

In our study we have formulated a time-dependent Landau–Ginzburg effective action (in imaginary time) of the two-band superconductor, which also enables us to compute general correlation functions by introducing source fields. Our results may be summarized as: (1) one collective mode exists with energy *between* twice the smaller and twice the larger gap energy and this mode stems from the interaction of relative superconducting phases (thus it is essentially the Leggett mode). (2) The screening effect of the long range Coulomb interaction eliminates the above-mentioned collective mode. If, on the other hand, the long range Coulomb interaction is not taken into account, the collective mode indeed shows up in the Raman response. (3) The π-phase shift is not *directly* observable in the Raman response, since basically it is squared to one.

This paper is organized as follows: in sections 2–6, the model, its functional formulation and the derivation of the effective action (including the long range Coulomb interaction) are presented. The collective excitations and the Raman response in the presence of the long range Coulomb interaction are investigated in section 7 and 8. We close this paper with a summary in section 9. Diverse polarization functions appearing in the main text are collected and computed in the appendices. For completeness, a general proof of the gauge invariance and the charge conservation of the electromagnetic response of two-band superconductors based on the functional approach is presented in appendix C. To present the results in a compact way we have introduced various notations. For the reader's convenience we have tried to remind them in appropriate places in the text.

2. Model

To avoid the complexities of the most general models we choose a specific model of the $s\pi$ pairing state [3, 6] of the iron-based pnictide superconductors. This model is described by the following two-band Hamiltonian with a *repulsive interband* pairing interaction. This model can be motivated by the unbiased renormalization group approach [16]. More concretely, starting from the bare Hamiltonian which also includes an intraband pairing interaction, the renormalization group trajectory flows into a region where the interband pairing interaction dominates (see equation (14) and figure 6(a) of [16]). The model Hamiltonian is given by

$$\hat{H} = \hat{H}_{\rm K} + \hat{H}_{\rm P} + \hat{H}_{\rm C} + \mu \sum_{\mathbf{k},\sigma} \left(c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + f^\dagger_{\mathbf{k}\sigma} f_{\mathbf{k}\sigma} \right), \qquad (1)$$

where $c_{\mathbf{k}\sigma}$ and $f_{\mathbf{k}\sigma}$ are the hole-like band and the electron-like band operator, respectively. These bands are believed to play a key role in the pnictide superconductors. $\sigma = \uparrow, \downarrow$ is a spin index and μ is the chemical potential which will be taken to be zero hereafter. The kinetic energy Hamiltonian $\hat{H}_{\rm K}$ is given by

$$\hat{H}_{\rm K} = \sum_{\mathbf{k},\sigma} (\epsilon_{c\mathbf{k}} c^\dagger_{\mathbf{k}\sigma} c_{\mathbf{k}\sigma} + \epsilon_{f\mathbf{k}} f^\dagger_{\mathbf{k}\sigma} f_{\mathbf{k}\sigma}), \qquad (2)$$

where $\epsilon_{c\mathbf{k}}$ and $\epsilon_{f\mathbf{k}}$ are the hole-like and electron-like energy band, respectively. These bands have tetragonal symmetry. The hole-like energy band $\epsilon_{c\mathbf{k}}$ is centred at $\mathbf{k} = (0, 0)$, while the electron-like energy band $\epsilon_{f\mathbf{k}}$ is centred at $\mathbf{k} = (\pi/a, \pi/a)$. a is a lattice spacing in the x–y plane. The explicit form of each band is

$$\begin{aligned} \epsilon_{c\mathbf{k}} &= \epsilon_{0c} - t_c (ka)^2 + t'_c (ka)^4 \cos(4\phi), \\ \epsilon_{f\mathbf{k}+(\frac{\pi}{a},\frac{\pi}{a})} &= -\epsilon_{0f} + t_f (ka)^2 + t'_f (ka)^4 \cos(4\phi), \end{aligned} \qquad (3)$$

where $k^2 = k_x^2 + k_y^2$ and $\phi = \tan^{-1}(k_y/k_x)$. $t_c, t_f > 0$, $\epsilon_{0c}, \epsilon_{0f} > 0$, $|t'_c| \ll t_c$, $|t'_f| \ll t_f$ are assumed. The dispersion along the z direction is ignored, making the model effectively two-dimensional.

The interband pairing Hamiltonian $\hat{H}_{\rm P}$ is given by

$$\hat{H}_{\rm P} = U \sum_{\mathbf{x}} [c^\dagger_{\mathbf{x}\uparrow} c^\dagger_{\mathbf{x}\downarrow} f_{\mathbf{x}\downarrow} f_{\mathbf{x}\uparrow} + {\rm h.c.}], \qquad (4)$$

where $\mathbf{x}$ denotes the direct lattice sites in the x–y plane. U is the interband pairing interaction, which is implicitly assumed to vanish above an energy cutoff ω_D, namely for $|\epsilon_{c/f\,\mathbf{k}}| > \omega_D$. For the $s\pi$ state U is positive, which is the case to be considered in this paper, while for the conventional s-wave state with no relative phase it should be taken to be negative.

$\hat{H}_C$ is the Hamiltonian of the long range Coulomb interaction:

$$\hat{H}_C = \frac{1}{2}\sum_{\mathbf{x},\mathbf{y}} \frac{e^2}{|\mathbf{x}-\mathbf{y}|}(\rho_{\mathbf{x}}-\rho_0)(\rho_{\mathbf{y}}-\rho_0), \tag{5}$$

where $\rho_{\mathbf{x}} = \rho_{c\mathbf{x}} + \rho_{f\mathbf{x}}$ with $\rho_{c\mathbf{x}} = \sum_\sigma c^\dagger_{\mathbf{x}\sigma} c_{\mathbf{x}\sigma}$ and $\rho_{f\mathbf{x}} = \sum_\sigma f^\dagger_{\mathbf{x}\sigma} f_{\mathbf{x}\sigma}$. ρ_0 is the uniform positive charge background required for the charge neutrality of the whole system.

3. The functional integral formulation

The partition function of the Hamiltonian (1) in the form of a functional integral is given by [17] (τ is an imaginary time and $\beta = 1/k_B T$)

$$Z[J] = \int D[c,f]\mathrm{e}^{-\int_0^\beta \mathrm{d}\tau\,[\sum_{\mathbf{x}}(c^\dagger\partial_\tau c + f^\dagger\partial_\tau f) + H(\tau)] - S_J}, \tag{6}$$

where we have inserted a source term ($\mathbf{q}$ is a wavenumber)

$$S_J = -\int_0^\beta \mathrm{d}\tau \sum_{\mathbf{q}} \tilde{\rho}_{\mathbf{q}}(\tau) J_{-\mathbf{q}}(\tau) \tag{7}$$

which is necessary in computing the correlation functions of the generalized density operator of the (non-resonant part of) Raman scattering. The generalized density operator $\tilde{\rho}_{\mathbf{q}}$ is defined by

$$\tilde{\rho}_{\mathbf{q}} = \sum_{\mathbf{k},\sigma}(\gamma^c_{\mathbf{k}} c^\dagger_{\mathbf{k}+\mathbf{q}/2\sigma} c_{\mathbf{k}-\mathbf{q}/2\sigma} + \gamma^f_{\mathbf{k}} f^\dagger_{\mathbf{k}+\mathbf{q}/2\sigma} f_{\mathbf{k}-\mathbf{q}/2\sigma}). \tag{8}$$

For the non-resonant electronic Raman scattering, the coefficients $\gamma^{c/f}_{\mathbf{k}}$ are given by

$$\gamma^{c/f}_{\mathbf{k}} = \sum_{\alpha,\beta} \hat{e}^{\mathrm{I}}_\alpha \frac{\partial^2 \epsilon_{c/f\,\mathbf{k}}}{\partial k_\alpha \partial k_\beta} \hat{e}^{\mathrm{F}}_\beta, \tag{9}$$

where $\hat{e}^{\mathrm{I}}$ and $\hat{e}^{\mathrm{F}}$ are the polarization vector of the incoming and outgoing photon, respectively. α and β label the coordinates perpendicular to the photon momentum which is taken to be along the z axis in our case. Note that c and f appearing in the functional integral are now *anticommuting Grassman* variables [17].

The correlation function of the generalized density operator $\chi_{\tilde{\rho}\tilde{\rho}}$ is defined by

$$\chi_{\tilde{\rho}\tilde{\rho}}(\tau-\tau',\mathbf{q}) = -\langle T_\tau \tilde{\rho}_{\mathbf{q}}(\tau)\tilde{\rho}_{-\mathbf{q}}(\tau')\rangle = -\frac{\delta^2 \ln Z[J]}{\delta J_{-\mathbf{q}}(\tau) J_{\mathbf{q}}(\tau')}\bigg|_{J\to 0}. \tag{10}$$

The electronic Raman cross section is proportional to the dynamical structure factor $S(\omega, \mathbf{q} \to 0)$ which is related to $\chi_{\tilde{\rho}\tilde{\rho}}$ in the following way:

$$S(\omega,\mathbf{q}) = [1 + n_B(\omega)]\Big[-\frac{1}{\pi}\,\mathrm{Im}\,\chi^R_{\tilde{\rho}\tilde{\rho}}(\omega,\mathbf{q})\Big], \tag{11}$$

where $n_B(\omega)$ is the Bose distribution function and $\chi^R_{\tilde{\rho}\tilde{\rho}}$ denotes the retarded correlation function [18–20]. The formalisms for the electronic Raman scattering of superconductors (including the generalizations to unconventional superconductors) are well expounded in [18–21].

The next step is to employ the Hubbard–Stratonovich transformations [17] to decouple the electron–electron interactions: $\hat{H}_P$ and $\hat{H}_C$.

To decompose $\hat{H}_P$ into the pairing channel it is necessary to change the order of either the c or f operator (but not both) since U is of positive sign. Flipping the order of the c operator the pairing Hamiltonian becomes

$$H_P = -U\sum_{\mathbf{x}}(c^\dagger_{\downarrow\mathbf{x}} c^\dagger_{\uparrow\mathbf{x}} f_{\downarrow\mathbf{x}} f_{\uparrow\mathbf{x}} + f^\dagger_{\uparrow\mathbf{x}} f^\dagger_{\downarrow\mathbf{x}} c_{\uparrow\mathbf{x}} c_{\downarrow\mathbf{x}}). \tag{12}$$

Applying the Hubbard–Stratonovich transformation in the pairing channel, we obtain

$$\begin{aligned} &\mathrm{e}^{-\int_0^\beta \mathrm{d}\tau\, H_P(\tau)} = \int D[\Delta_c,\Delta_f]\mathrm{e}^{-S_P}, \\ &S_P = \int \mathrm{d}\tau \sum_{\mathbf{x}}\Big[\frac{\Delta^*_{c\mathbf{x}}(\tau)\Delta_{f\mathbf{x}}(\tau)}{U} + \frac{\Delta^*_{f\mathbf{x}}(\tau)\Delta_{c\mathbf{x}}(\tau)}{U} \\ &\quad - c^\dagger_{\mathbf{x}\downarrow}(\tau)c^\dagger_{\mathbf{x}\uparrow}(\tau)\Delta_{c\mathbf{x}}(\tau) - f^\dagger_{\mathbf{x}\uparrow}(\tau)f^\dagger_{\mathbf{x}\downarrow}(\tau)\Delta_{f\mathbf{x}}(\tau) \\ &\quad - \Delta^*_{c\mathbf{x}}(\tau)c_{\mathbf{x}\uparrow}(\tau)c_{\mathbf{x}\downarrow}(\tau) - \Delta^*_{f\mathbf{x}}(\tau)f_{\mathbf{x}\downarrow}(\tau)f_{\mathbf{x}\uparrow}(\tau)\Big]. \end{aligned} \tag{13}$$

$\Delta_{c/f\mathbf{x}}(\tau)$ are nothing but the (fluctuating) superconducting order parameters.

The Coulomb interaction can also be decomposed in a similar way:

$$\begin{aligned} &\mathrm{e}^{-\int_0^\beta \mathrm{d}\tau\, H_C(\tau)} = \int D[\phi]\,\mathrm{e}^{-S_C}, \\ &S_C = \int_0^\beta \mathrm{d}\tau \sum_{\mathbf{q}}[\tfrac{1}{2}V^{-1}_{\mathbf{q}}\phi_{-\mathbf{q}}(\tau)\phi_{\mathbf{q}}(\tau) \\ &\quad - \mathrm{i}\phi_{-\mathbf{q}}(\tau)\rho_{\mathbf{q}}(\tau)], \end{aligned} \tag{14}$$

where $V_{\mathbf{q}}$ is the Coulomb matrix element:

$$V^{-1}_{\mathbf{q}} = \begin{cases} \dfrac{|\mathbf{q}|^2}{4\pi e^2}, & \text{3-dimension,} \\ \dfrac{|\mathbf{q}|}{2\pi e^2}, & \text{2-dimension.} \end{cases} \tag{15}$$

The boson field $\phi(\tau)$ plays the role of the scalar potential of electromagnetic fields. Now the partition function Z becomes

$$Z = \int D[\Delta_c,\Delta_f,\phi]\int D[c,f]\mathrm{e}^{-S_K - S_C - S_P - S_J}, \tag{16}$$

where S_K is the action for the kinetic term $\hat{H}_K$:

$$S_K = \int_0^\beta \mathrm{d}\tau\Big[\sum_{\mathbf{x},\sigma}(c^\dagger_{\mathbf{x}\sigma}\partial_\tau c_{\mathbf{x}\sigma} + f^\dagger_{\mathbf{x}\sigma}\partial_\tau f_{\mathbf{x}\sigma}) + H_K(\tau)\Big]. \tag{17}$$

Note that the fermion variables c, f appear in (16) quadratically, so that they can be integrated out exactly using the Gaussian integration formula of Grassman variables [17]. Let us introduce the following Nambu spinors:

$$\Psi_{\mathbf{k}} = \begin{pmatrix} \Psi_{c\mathbf{k}} \\ \Psi_{f\mathbf{k}} \end{pmatrix}, \qquad \Psi_{c\mathbf{k}} = \begin{pmatrix} c_{\mathbf{k}\uparrow} \\ c^{\dagger}_{-\mathbf{k}\downarrow} \end{pmatrix}, \qquad \Psi_{f\mathbf{k}} = \begin{pmatrix} f_{\mathbf{k}\uparrow} \\ f^{\dagger}_{-\mathbf{k}\downarrow} \end{pmatrix}. \tag{18}$$

In terms of the Nambu spinors the generalized density operator $\tilde{\rho}_{\mathbf{q}}$, (8), can be re-expressed as

$$\tilde{\rho}_{\mathbf{q}} = \sum_{\mathbf{k}} \Psi^{\dagger}_{\mathbf{k}+\mathbf{q}/2} \begin{pmatrix} \gamma^{c}_{\mathbf{k}} \tau_3 & 0 \\ 0 & \gamma^{f}_{\mathbf{k}} \tau_3 \end{pmatrix} \Psi_{\mathbf{k}-\mathbf{q}/2}, \tag{19}$$

where $\tau_{1,2,3}$ are the Pauli matrices acting on the particle–hole space. The parts containing the Nambu spinors in the actions of S_{C} and S_{P} can be written in the following ways:

$$S^{\mathrm{Nambu}}_{\mathrm{C}} = \int_0^{\beta} \mathrm{d}\tau \sum_{\mathbf{k},\mathbf{q}} \Psi^{\dagger}_{\mathbf{k}+\mathbf{q}/2}(\tau) \hat{M}_{\mathrm{C}} \Psi_{\mathbf{k}-\mathbf{q}/2}(\tau), \qquad \hat{M}_{\mathrm{C}} = (-\mathrm{i}) \mathrm{I}_2 \otimes \tau_3 \frac{\phi_{-\mathbf{q}}(\tau)}{\sqrt{N}}, \tag{20}$$

where I_2 is a 2×2 unit matrix acting on the space of c, f species. N is the number of lattice sites.

The sum $S_{\mathrm{K}} + S^{\mathrm{Nambu}}_{\mathrm{P}}$ can be expressed as

$$S_{\mathrm{K}} + S^{\mathrm{Nambu}}_{\mathrm{P}} = \int_0^{\beta} \mathrm{d}\tau \sum_{\mathbf{x}} \Psi^{\dagger}_{\mathbf{x}}(\tau) \hat{M}_{\mathrm{P}} \Psi_{\mathbf{x}}(\tau), \qquad \hat{M}_{\mathrm{P}} = \begin{pmatrix} \hat{K}_c & 0 \\ 0 & \hat{K}_f \end{pmatrix}, \tag{21}$$

where the kernel matrices are given by

$$\hat{K}_c = \begin{pmatrix} \partial_\tau + \epsilon_c(-\mathrm{i}\nabla) & \Delta_c \\ \Delta^*_c & \partial_\tau - \epsilon_c(-\mathrm{i}\nabla) \end{pmatrix}, \qquad \hat{K}_f = \begin{pmatrix} \partial_\tau + \epsilon_f(-\mathrm{i}\nabla) & \zeta \Delta_f \\ \zeta \Delta^*_f & \partial_\tau - \epsilon_f(-\mathrm{i}\nabla) \end{pmatrix}, \tag{22}$$

where $\zeta = -1$ is a factor which keeps the track of the π phase difference of the $s\pi$ pairing.

The source term can also be expressed as

$$S_J = \int_0^{\beta} \mathrm{d}\tau \sum_{\mathbf{k},\mathbf{q}} \Psi^{\dagger}_{\mathbf{k}+\mathbf{q}/2}(\tau) \hat{M}_J \Psi_{\mathbf{k}-\mathbf{q}/2}(\tau), \qquad \hat{M}_J = -J_{-\mathbf{q}}(\tau) \begin{pmatrix} \gamma^{c}_{\mathbf{k}} \tau_3 & 0 \\ 0 & \gamma^{f}_{\mathbf{k}} \tau_3 \end{pmatrix}. \tag{23}$$

Integrating out fermions (this means doing the c, f integral in (16)) we arrive at

$$Z[J] = \int D[\Delta_c, \Delta_f, \phi] \mathrm{e}^{-S_{\mathrm{B}}},$$

$$S_{\mathrm{B}} = \int_0^{\beta} \mathrm{d}\tau \left[\sum_{\mathbf{x}} \frac{1}{U} [\Delta^*_{c\mathbf{x}}(\tau) \Delta_{f\mathbf{x}}(\tau) + \Delta^*_{f\mathbf{x}}(\tau) \Delta_{c\mathbf{x}}(\tau)] + \sum_{\mathbf{q}} \tfrac{1}{2} V^{-1}_{\mathbf{q}} \phi_{-\mathbf{q}}(\tau) \phi_{\mathbf{q}}(\tau) \right] - \ln\det(\hat{M}_{\mathrm{P}} + \hat{M}_{\mathrm{C}} + \hat{M}_J) \tag{24}$$

S_{B} is the effective action of bosonic excitations: $\Delta_{c/f}$ and ϕ. Equation (24) is a formally exact result. The difficult part is to compute the determinant factor $\ln\det(\hat{M}_{\mathrm{P}} + \hat{M}_{\mathrm{C}} + \hat{M}_J)$ in a closed form.

We will compute the partition function (24) employing a saddle point approximation (plus incorporating Gaussian fluctuations around the saddle point). The saddle point condition, as expected, is nothing other than the gap equations for the two-band superconductors. Furthermore, the Gaussian fluctuations will determine the dynamics of the collective excitations of the two-band superconductors.

4. The saddle point solution: the gap equations

The saddle point solution is determined by the condition that the first-order functional derivatives of S_{B} with respect to Δ_c, Δ_f, and ϕ vanish. The source field J should be put to zero in the determination of a saddle point, since it is just a (infinitesimally small) formal device introduced for the computation of correlation functions:

$$\left. \frac{\delta S_{\mathrm{B}}}{\delta \phi} \right|_{\Delta_{c0}, \Delta_{f0}, \phi_0} = \left. \frac{\delta S_{\mathrm{B}}}{\delta \Delta_c} \right|_{\Delta_{c0}, \Delta_{f0}, \phi_0} = \left. \frac{\delta S_{\mathrm{B}}}{\delta \Delta_f} \right|_{\Delta_{c0}, \Delta_{f0}, \phi_0} = 0, \tag{25}$$

where $\Delta_{c0}, \Delta_{f0}, \phi_0$ denote the saddle point values. These functional derivatives can be computed using the following matrix identity:

$$\delta \ln\det M = \delta\, \mathrm{Tr} \ln M = \mathrm{Tr}(M^{-1} \delta M). \tag{26}$$

The ϕ equation is easily shown to give $\phi_{\mathbf{q}\neq 0,0} = 0$, and $\phi_{\mathbf{q}=0,0}$ enforces the overall charge neutrality condition. The $\Delta_{c/f}$ equations yield the gap equations:

$$\Delta_{c/f\,0}(\mathbf{k}) = \frac{U}{N} \sum_{\mathbf{k}'} \frac{\tanh(\beta E_{f/c\mathbf{k}'}/2)}{2 E_{f/c\mathbf{k}'}} \Delta_{f/c\,0}(\mathbf{k}'), \tag{27}$$

where (from now on, $T = 0$ will be assumed)

$$E_{c/f\mathbf{k}} = \sqrt{\epsilon^2_{c/f\mathbf{k}} + |\Delta_{c/f0}(\mathbf{k})|^2} \tag{28}$$

is the (bare) quasiparticle energy. The gap equation (27) admits the *real* s-wave solution of Δ_{c0} and Δ_{f0} with the *same* sign. This solution represents the relative π-phase between two gaps due the phase factor ζ.

Using the explicit form of the energy bands of (3) the gap equations reduce to

$$\Delta_{c0} = \Delta_{f0} \lambda_f \ln \frac{2\omega_D}{\Delta_{f0}}, \qquad \Delta_{f0} = \Delta_{c0} \lambda_c \ln \frac{2\omega_D}{\Delta_{c0}}, \tag{29}$$

where $\lambda_{c/f}$ are the dimensionless coupling constants:

$$\lambda_{c/f} = U D_{c/f}, \qquad D_{c/f} = \frac{1}{4\pi t_{c/f}}, \tag{30}$$

where $D_{c/f}$ is the density of states at the Fermi energy. Choosing $\omega_D = 100$ meV, $\lambda_c = 0.2171$ and $\lambda_f = 0.6676$, we obtain that $\Delta_c = 20$ meV and $\Delta_f = 10$ meV, which are compatible with experimental data. These will be taken as the representative input for the presentation of our results.

5. Gaussian fluctuations

Once a saddle point is determined, we can incorporate the fluctuations around the saddle point exactly in quadratic order.

The fluctuations of the superconducting order parameters can be separated from the saddle point value up to quadratic order in the following way (let us use 4-vector notation $x = (\mathbf{x}, \tau)$):

$$\Delta_{c/f}(x) = [\Delta_{c/f0} + \delta\Delta_{c/f}(x)]e^{i\theta_{c/f}(x)} \approx \Delta_{c/f0} + \delta\Delta_{c/f}(x) + \Delta_{c/f0}[i\theta_{c/f}(x)] - \tfrac{1}{2}\Delta_{c/f0}\theta^2_{c/f}(x) + \delta\Delta_{c/f}[i\theta_{c/f}(x)], \quad (31)$$

where $\delta\Delta_{c/f}(x)$ and $\theta_{c/f}(x)$ are the amplitude fluctuation and the phase fluctuation, respectively. This expansion represents a spin-wave approximation (along with amplitude fluctuation) where the vortex excitations are ignored. Plugging (31) into (22), the kernel matrices $\hat{K}_{c/f}$ can be decomposed into the saddle point part, the first-order and the second-order fluctuation parts as follows:

$$\hat{K}_c = \hat{K}_{c0} + \delta\hat{K}^{(1)}_c + \delta\hat{K}^{(2)}_c,$$
$$\hat{K}_{c0} = \begin{pmatrix} \partial_\tau + E_c & \Delta_{c0} \\ \Delta_{c0} & \partial_\tau - E_c \end{pmatrix}, \quad (32)$$
$$\delta\hat{K}^{(1)}_c = \tau_1\delta\Delta_c(x) - \tau_2\Delta_{c0}\theta_c(x),$$
$$\delta\hat{K}^{(2)}_c = -\tfrac{1}{2}\tau_1\Delta_{c0}\theta^2_c(x) - \tau_2\delta\Delta_c(x)\theta_c(x).$$

$$\hat{K}_f = \hat{K}_{f0} + \delta\hat{K}^{(1)}_f + \delta\hat{K}^{(2)}_f,$$
$$\hat{K}_{f0} = \begin{pmatrix} \partial_\tau + E_f & \zeta\Delta_{f0} \\ \zeta\Delta_{f0} & \partial_\tau - E_f \end{pmatrix}, \quad (33)$$
$$\delta\hat{K}^{(1)}_f = \zeta\tau_1\delta\Delta_f(x) - \zeta\tau_2\Delta_{f0}\theta_f(x),$$
$$\delta\hat{K}^{(2)}_f = -\zeta\tfrac{1}{2}\tau_1\Delta_{f0}\theta^2_f(x) - \zeta\tau_2\delta\Delta_f(x)\theta_f(x).$$

As for the fluctuation of $\phi_{\mathbf{q}\neq 0}$, its saddle point value is null, so that $\phi_{\mathbf{q}\neq 0}$ itself can be treated as a fluctuation.

The inverse of $\hat{K}_{c/f0}$ is the (bare) one-particle Green's function in the superconducting state:

$$-\hat{K}^{-1}_{c0} = \hat{G}_c(i\epsilon, \mathbf{k}) = \frac{i\epsilon\tau_0 + \epsilon_{c\mathbf{k}}\tau_3 + \Delta_{c0}\tau_1}{(i\epsilon)^2 - E^2_{c\mathbf{k}}},$$
$$-\hat{K}^{-1}_{f0} = \hat{G}_f(i\epsilon, \mathbf{k}) = \frac{i\epsilon\tau_0 + \epsilon_{f\mathbf{k}}\tau_3 + \zeta\Delta_{f0}\tau_1}{(i\epsilon)^2 - E^2_{f\mathbf{k}}}, \quad (34)$$

where $E_{c/f\mathbf{k}}$ is the bare quasiparticle energy defined in (28). The one-particle Green function in a 4×4 matrix form is

$$\hat{G}(i\epsilon, \mathbf{k}) = \begin{pmatrix} \hat{G}_c(i\epsilon, \mathbf{k}) & 0 \\ 0 & \hat{G}_f(i\epsilon, \mathbf{k}) \end{pmatrix}. \quad (35)$$

Plugging in (32) and (33) into (24) and expanding it with respect to $\delta\hat{K}^{(1,2)}_{c/f}$ and ϕ up to the second order, we obtain the Gaussian effective action. The terms which are of first order in fluctuations vanish owing to the saddle point condition. Then the partition function can be approximated by

$$Z[J] \approx e^{-S_{\text{sad}}}\int D[\delta\Delta_{c/f}, \theta_{c/f}, \phi]e^{-S_{\text{Gau}}}, \quad (36)$$

where S_{sad} is the saddle point contribution whose explicit form is not necessary in our discussion and S_{Gau} is the second-order Gaussian effective action $[x = (\mathbf{x}, \tau)]$:

$$S_{\text{Gau}} = \int_0^\beta d\tau\Big\{\sum_{\mathbf{x}}\frac{2}{U}\delta\Delta_c(x)\delta\Delta_f(x) - \sum_{\mathbf{x}}\frac{1}{U}\Delta_{c0}\Delta_{f0}[\theta_c(x) - \theta_f(x)]^2 + \sum_{\mathbf{q}}\tfrac{1}{2}V^{-1}_{\mathbf{q}}\phi_{-\mathbf{q}}(\tau)\phi_{\mathbf{q}}(\tau)\Big\} + \int_0^\beta d\tau\sum_{\mathbf{x}}\text{tr}[\hat{G}_c(0)\hat{K}^{(2)}_c(x) + \hat{G}_f(0)\hat{K}^{(2)}_f(x)] + \tfrac{1}{2}\sum_{k,q}\text{tr}[\hat{G}_c(k+q)\hat{V}_c(q)\hat{G}_c(k)\hat{V}_c(-q)] + \tfrac{1}{2}\sum_{k,q}\text{tr}[\hat{G}_f(k+q)\hat{V}_f(q)\hat{G}_f(k)\hat{V}_f(-q)], \quad (37)$$

where more 4-vector notations are introduced: $k = (i\epsilon, \mathbf{k})$ and $q = (i\omega, \mathbf{q})$. The traces are over the Pauli matrices. The vertex factors $V_{c/f}(q)$ are given by

$$\hat{V}_c(q) = \Big[-\gamma^c_{\mathbf{k}+\mathbf{q}/2}J_q + (-i)\frac{\phi_q}{\sqrt{N}}\Big]\tau_3 + \delta\Delta_c(q)\tau_1 - \Delta_{c0}\theta_c(q)\tau_2,$$
$$\hat{V}_f(q) = \Big[-\gamma^f_{\mathbf{k}+\mathbf{q}/2}J_q + (-i)\frac{\phi_q}{\sqrt{N}}\Big]\tau_3 + \zeta\delta\Delta_f(q)\tau_1 - (\zeta\Delta_{f0})\theta_f(q)\tau_2. \quad (38)$$

The Gaussian action (37) can be decomposed into three pieces: S_{J2} which is quadratic in J_q, S_{J1} which is linear in J_q, and S_{J0}, which does not depend on J_q. The explicit form of S_{J2} is

$$S_{J2} = \tfrac{1}{2}\sum_q[\pi^c_{33,\gamma\gamma}(q) + \pi^f_{33,\gamma\gamma}(q)]J_qJ_{-q}, \quad (39)$$

where the polarization functions $\pi^{c/f}_{33,\gamma\gamma}$ are defined in appendix A. In fact, (39) (being plugged into (10)) yields the correlation function which is identical with that obtained from the BCS mean-field theory (without Coulomb correction).

To simplify notations let us introduce the following (column) vector (T is a matrix transpose):

$$X_q = [\delta\Delta_c(q), \delta\Delta_f(q), \Delta_{c0}\theta_c(q), \Delta_{f0}\theta_f(q)]^{\text{T}}. \quad (40)$$

Then S_{J1} is given by

$$S_{J1} = \frac{1}{2}\sum_q\Big\{i[\pi^c_{33,\gamma 1}(q) + \pi^f_{33,\gamma 1}(q)]\frac{J_q\phi_{-q}}{\sqrt{N}} + i[\pi^c_{33,1\gamma}(q) + \pi^f_{33,1\gamma}(q)]\frac{J_{-q}\phi_q}{\sqrt{N}}\Big\} + \tfrac{1}{2}\sum_q[J_q Z_q\cdot X_{-q} + J_{-q}\tilde{Z}_{-q}\cdot X_q], \quad (41)$$

where (the dot between X_{-q} and Z_q denotes a matrix product and all of the polarization functions are defined in appendix A)

$$Z_q = [-\pi^c_{31,\gamma 1}(q), -\zeta\pi^f_{31,\gamma 1}(q), \pi^c_{32,\gamma 1}(q), \zeta\pi^f_{32,\gamma 1}(q)],$$
$$\tilde{Z}_{-q} = [-\pi^c_{13,1\gamma}(q), -\zeta\pi^f_{13,1\gamma}(q), \pi^c_{23,1\gamma}(q), \zeta\pi^f_{23,1\gamma}(q)]. \quad (42)$$

Note that, in spite of their appearances, Z_q and $\tilde{Z}_{-q}$ are, in fact, independent of the phase factor ζ, since the polarization functions $\pi^f_{13,1\gamma}, \pi^f_{31,\gamma 1}, \pi^f_{32,\gamma 1}$ and $\pi^f_{23,1\gamma}$ are proportional to the phase factor ζ. Namely the phase factor is squared to one, $\zeta^2 = 1$.

Let us also define a 4×4 matrix $\Gamma(q)$ (the argument q of the polarization functions is suppressed below for notational simplicity):

$$\Gamma(q) = \begin{bmatrix} \pi^c_{11} & \frac{2}{U} & -\pi^c_{12} & 0 \\ \frac{2}{U} & \pi^f_{11} & 0 & -\pi^f_{12} \\ -\pi^c_{21} & 0 & -\frac{\Delta_{f0}}{\Delta_{c0}}\frac{2}{U} + \tilde{\pi}^c_{22} & \frac{2}{U} \\ 0 & -\pi^f_{21} & \frac{2}{U} & -\frac{\Delta_{c0}}{\Delta_{f0}}\frac{2}{U} + \tilde{\pi}^f_{22} \end{bmatrix}, \tag{43}$$

where

$$\tilde{\pi}^{c/f}_{22}(q) = \pi^{c/f}_{22}(q) + \frac{1}{N}\sum_{\mathbf{k}} \frac{1}{E_{c/f\mathbf{k}}}. \tag{44}$$

A simple calculation shows that

$$\frac{1}{N}\sum_{\mathbf{k}} \frac{1}{E_{c/f\mathbf{k}}} = D_{c/f} \ln \frac{2\omega_D}{\Delta_{c/f0}}. \tag{45}$$

Now S_{J0} takes the following form:

$$S_{J0} = \tfrac{1}{2}\sum_q \Big[V^{-1}_{\mathbf{q}} - (\pi^c_{33} + \pi^f_{33})\Big]\phi_q\phi_{-q} + \mathrm{i}\frac{1}{2}\frac{1}{\sqrt{N}}\sum_q [\phi_q(Y_q\cdot X_{-q}) + \phi_{-q}(\tilde{Y}_{-q}\cdot X_q)] + \tfrac{1}{2}\sum_q X^{\mathrm{T}}_{-q}\Gamma(q)X_q, \tag{46}$$

where

$$\begin{aligned} Y_q &= [-\pi^c_{31}(q), -\zeta\pi^f_{31}(q), \pi^c_{32}(q), \zeta\pi^f_{32}(q)] \\ \tilde{Y}_{-q} &= [-\pi^c_{13}(q), -\zeta\pi^f_{13}(q), \pi^c_{23}(q), \zeta\pi^f_{23}(q)]. \end{aligned} \tag{47}$$

Note that Y_q and $\tilde{Y}_{-q}$ are also independent of ζ by the same reason as Z_q and $\tilde{Z}_{-q}$. From the explicit form of the polarization functions one can easily check that the matrix $\Gamma(q)$ is also independent of the phase factor ζ. Thus we find that the presence of the π-phase shift is *not manifest*, at least in the present approach of the saddle point approximation plus Gaussian fluctuations.

At this stage the functional integral takes the following form:

$$Z[J] \approx \mathrm{e}^{-S_{\mathrm{sad}} - S_{J2}} \int D[\delta\Delta_{c/f}, \theta_{c/f}, \phi]\mathrm{e}^{-S_{J1} - S_{J0}}. \tag{48}$$

In section 6 we perform the functional integral of (48) over ϕ.

6. Influences of long range Coulomb interaction

The functional integral over ϕ amounts to incorporating the long range Coulomb interaction. The functional integral of (48) over ϕ can be done exactly, since it is a Gaussian integral. The action S_{J0} has a piece which is quadratic in ϕ:

$$\begin{aligned} S^{(2)}_\phi &= \tfrac{1}{2}\sum_q D^{-1}(q)\phi_q\phi_{-q}, \\ D^{-1}(q) &\equiv \frac{1}{V(\mathbf{q})} - [\pi^c_{33}(q) + \pi^f_{33}(q)]. \end{aligned} \tag{49}$$

Collecting the terms linear in ϕ from S_{J0} and S_{J1}, we find

$$S^{(1)}_\phi = \frac{\mathrm{i}}{2}\frac{1}{\sqrt{N}}\sum_q \{\phi_q[J_{-q}(\pi^c_{33,1\gamma} + \pi^f_{33,1\gamma}) + Y_q\cdot X_{-q}] + \phi_{-q}[J_q(\pi^c_{33,\gamma 1} + \pi^f_{33,\gamma 1}) + \tilde{Y}_{-q}\cdot X_q]\}, \tag{50}$$

where X_q and Y_q are defined in (40) and (47), respectively. The functional integral to be done is

$$\int D[\phi]\mathrm{e}^{-S^{(2)}_\phi - S^{(1)}_\phi}. \tag{51}$$

Now the integral is straightforward and the result is

$$\exp\Big\{-\tfrac{1}{2}\sum_q D(q)[J_{-q}(\pi^c_{33,1\gamma} + \pi^f_{33,1\gamma}) + Y_q\cdot X_{-q}] \times [J_q(\pi^c_{33,\gamma 1} + \pi^f_{33,\gamma 1}) + \tilde{Y}_{-q}\cdot X_q]\Big\}. \tag{52}$$

From the exponent of (52), we again obtain the terms quadratic and linear in J_q, and independent of J_q. These terms can be combined with the results obtained in the previous section, namely S_{J2}, S_{J1} and S_{J0}. These terms coming from the integration over ϕ represent the renormalization by long range Coulomb interaction. The action which is quadratic in J becomes (the superscript r indicates that the renormalization by Coulomb interaction is incorporated)

$$S^{\mathrm{r}}_{J2} = \tfrac{1}{2}\sum_q J_{-q}J_q\{\pi^c_{33,\gamma\gamma} + \pi^f_{33,\gamma\gamma} + D(q)(\pi^c_{33,\gamma 1} + \pi^f_{33,\gamma 1})(\pi^c_{33,1\gamma} + \pi^f_{33,1\gamma})\}, \tag{53}$$

where the last piece proportional to $D(q)$ comes from the renormalization by Coulomb interaction. The action which is independent of J becomes (see (40) for the definition of X_q)

$$S^{\mathrm{r}}_{J0} = \tfrac{1}{2}\sum_q X^{\mathrm{T}}_{-q}\Gamma^{\mathrm{r}}(q)X_q, \tag{54}$$

where the elements of the renormalized matrix Γ^{r} are given by ($i, j = 1, 2, 3, 4$, and see (47) for the definition of Y_q)

$$[\Gamma^{\mathrm{r}}(q)]_{ij} = [\Gamma(q)]_{ij} + D(q)Y_{q,i}\tilde{Y}_{-q,j}. \tag{55}$$

The action which is linear in J_q becomes

$$S^{\mathrm{r}}_{J1} = \tfrac{1}{2}\sum_q (J_q W_q\cdot X_{-q} + J_{-q}\tilde{W}_{-q}\cdot X_q), \tag{56}$$

where (see (42) for the definition of Z_q)

$$\begin{aligned} W_q &= Z_q + D(q)[\pi^c_{33,\gamma 1}(q) + \pi^f_{33,\gamma 1}(q)]Y_q, \\ \tilde{W}_{-q} &= \tilde{Z}_{-q} + D(q)[\pi^c_{33,1\gamma}(q) + \pi^f_{33,1\gamma}(q)]\tilde{Y}_{-q}. \end{aligned} \tag{57}$$

Again the renormalization by Coulomb interaction is reflected in the terms proportional to $D(q)$ in (57).

At this step the functional integral has been reduced to

$$Z[J] \approx \mathrm{e}^{-S_{\mathrm{sad}} - S^{\mathrm{r}}_{J2}} \int D[X]\mathrm{e}^{-S^{\mathrm{r}}_{J0} - S^{\mathrm{r}}_{J1}}. \tag{58}$$

Noting that S^{r}_{J0} is quadratic in X_q and that S^{r}_{J1} is linear in X_q, the functional integral of (58) can be done exactly to yield the desired correlation functions. This will be done in section 8. The source field J_q is just a formal device which was introduced to facilitate the computation of correlation functions, and it has nothing to do with the intrinsic properties of the physical system under consideration. If J_q is put to zero, we have

$$Z[J=0] \sim \text{constant} \times \int D[X] e^{-S^{r}_{J0}}. \tag{59}$$

Thus the dynamics of the fluctuations of order parameters is encapsulated in the action S^{r}_{J0}. In particular, it determines the collective excitations, and these are discussed in section 7.

7. Collective excitations

To reveal the structure of the action S^{r}_{J0} it is convenient to change the basis of fluctuating order parameters in the following way. First define

$$\Delta_0 \equiv \sqrt{\Delta_{c0}\Delta_{f0}}. \tag{60}$$

Then introduce the following *symmetric* and *antisymmetric* amplitude and phase fluctuations $\delta\Delta_{\pm}, \theta_{\pm}$:

$$\delta\Delta_{\pm} = \frac{1}{\sqrt{2}}(\pm\delta\Delta_c + \delta\Delta_f),$$
$$\begin{pmatrix} \Delta_0\theta_+ \\ \Delta_0\theta_- \end{pmatrix} = \begin{pmatrix} \sqrt{\frac{\Delta_{f0}}{2\Delta_{c0}}} & \sqrt{\frac{\Delta_{c0}}{2\Delta_{f0}}} \\ -\sqrt{\frac{\Delta_{f0}}{2\Delta_{c0}}} & \sqrt{\frac{\Delta_{c0}}{2\Delta_{f0}}} \end{pmatrix} \begin{pmatrix} \Delta_{c0}\theta_c \\ \Delta_{f0}\theta_f \end{pmatrix}. \tag{61}$$

The symmetric phase θ_+ corresponds to the Goldstone boson of spontaneously broken electromagnetic $U(1)$ symmetry (see appendix C for more details).

Then the fluctuating order parameters X_q (40) are transformed into

$$X'_q = \begin{bmatrix} \delta\Delta_+ \\ \delta\Delta_- \\ \Delta_0\theta_+ \\ \Delta_0\theta_- \end{bmatrix} = S X_q, \tag{62}$$

where the 4×4 transformation matrix S is given by

$$S = \begin{pmatrix} \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 & 0 \\ -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 & 0 \\ 0 & 0 & \sqrt{\frac{\Delta_{f0}}{2\Delta_{c0}}} & \sqrt{\frac{\Delta_{c0}}{2\Delta_{f0}}} \\ 0 & 0 & -\sqrt{\frac{\Delta_{f0}}{2\Delta_{c0}}} & \sqrt{\frac{\Delta_{c0}}{2\Delta_{f0}}} \end{pmatrix} \tag{63}$$

Then the action S^{r}_{J0} (54) becomes

$$S^{r}_{J0} = \tfrac{1}{2}\sum_q (X'_{-q})^{\mathrm{T}}\Gamma^{r'}(q)X'_q, \tag{64}$$

where

$$\Gamma^{r'}(q) = (S^{-1})^{\mathrm{T}}\Gamma^{r}(q)S^{-1}. \tag{65}$$

Note that $\det\Gamma^{r'} = \det\Gamma^{r}$. The vanishing condition of the real part of the determinant of Γ^{r} determines the spectra of the collective excitations which are renormalized by the Coulomb interaction, since it corresponds to the poles of the Green's functions of collective excitations.

The Raman scattering probes the excitations with very small wavenumber $\mathbf{q} \to 0$. In the limit $\mathbf{q} \to 0$, the following polarization functions, $\pi^{c/f}_{12}(\mathrm{i}\omega, \mathbf{q}=0)$ and $\pi^{c/f}_{13}(\mathrm{i}\omega, \mathbf{q}=0)$, vanish for the energy bands with a particle–hole symmetry (see appendix A), which we assume in this paper. Due to the vanishing of the above correlation functions, the 4×4 matrix Γ^{r} is block-diagonalized into the amplitude block and the phase block. Namely, the amplitude fluctuations and the phase fluctuations decouple in the $\mathbf{q} \to 0$ limit:

$$\Gamma^{r}(\mathrm{i}\omega, \mathbf{q}\to 0) = \begin{bmatrix} \Gamma^{\Delta}(\mathrm{i}\omega, \mathbf{q}\to 0) & 0 \\ 0 & \Gamma^{\theta}(\mathrm{i}\omega, \mathbf{q}\to 0) \end{bmatrix}, \tag{66}$$

where $\Gamma^{\Delta,\theta}$ are the 2×2 matrices in the amplitude and the phase block, respectively. The explicit form of each matrix is

$$\Gamma^{\Delta} = \begin{bmatrix} \pi^{c}_{11} & \frac{2}{U} \\ \frac{2}{U} & \pi^{f}_{11} \end{bmatrix}. \tag{67}$$

From the above form one can see that the amplitude sector is free of Coulomb corrections ($D(q)$ terms) in the $\mathbf{q} \to 0$ limit:

$$\Gamma^{\theta} = \begin{bmatrix} -\frac{\Delta_{f0}}{\Delta_{c0}}\frac{2}{U} + \tilde{\pi}^{c}_{22} - D(q)[\pi^{c}_{23}]^2 & \frac{2}{U} - D(q)\pi^{c}_{23}[\zeta\pi^{f}_{23}] \\ \frac{2}{U} - D(q)\pi^{c}_{23}[\zeta\pi^{f}_{23}] & -\frac{\Delta_{c0}}{\Delta_{f0}}\frac{2}{U} + \tilde{\pi}^{f}_{22} - D(q)[\pi^{f}_{23}]^2 \end{bmatrix}. \tag{68}$$

In the basis of the symmetric (+) and the antisymmetric (−) fluctuations (via the relation (65)), the matrices become

$$\Gamma^{r'}(\mathrm{i}\omega, \mathbf{q}\to 0) = \begin{bmatrix} \Gamma^{\Delta'}(\mathrm{i}\omega, \mathbf{q}\to 0) & 0 \\ 0 & \Gamma^{\theta'}(\mathrm{i}\omega, \mathbf{q}\to 0) \end{bmatrix}. \tag{69}$$

First note that $\det\Gamma^{\Delta/\theta} = \det\Gamma^{\Delta/\theta'}$. The matrix elements of $\Gamma^{\theta'}$ are given by (the explicit form of $\Gamma^{\Delta'}$ is not necessary)

$$\begin{aligned} \Gamma^{\theta'}_{++} &= \frac{1}{2}\frac{\Delta_{c0}}{\Delta_{f0}}(\tilde{\pi}^{c}_{22} - D(q)[\pi^{c}_{23}]^2) \\ &\quad + \frac{1}{2}\frac{\Delta_{f0}}{\Delta_{c0}}(\tilde{\pi}^{f}_{22} - D(q)[\pi^{f}_{23}]^2) - D(q)\pi^{c}_{23}(\zeta\pi^{f}_{23}), \\ \Gamma^{\theta'}_{--} &= \frac{1}{2}\frac{\Delta_{c0}}{\Delta_{f0}}(\tilde{\pi}^{c}_{22} - D(q)[\pi^{c}_{23}]^2) \\ &\quad + \frac{1}{2}\frac{\Delta_{f0}}{\Delta_{c0}}(\tilde{\pi}^{f}_{22} - D(q)[\pi^{f}_{23}]^2) - \frac{4}{U} + D(q)\pi^{c}_{23}(\zeta\pi^{f}_{23}), \\ \Gamma^{\theta'}_{+-} &= \Gamma^{\theta'}_{-+} = -\frac{1}{2}\frac{\Delta_{c0}}{\Delta_{f0}}(\tilde{\pi}^{c}_{22} - D(q)[\pi^{c}_{23}]^2) \\ &\quad + \frac{1}{2}\frac{\Delta_{f0}}{\Delta_{c0}}(\tilde{\pi}^{f}_{22} - D(q)[\pi^{f}_{23}]^2). \end{aligned} \tag{70}$$

Using the results obtained for $\pi^{c/f}_{ij}(\mathrm{i}\omega, \mathbf{q}=0)$ in appendix B and that (see (49))

$$D(\mathrm{i}\omega, \mathbf{q}=0) = -\frac{1}{\pi^{c}_{33}(\mathrm{i}\omega, \mathbf{q}=0) + \pi^{f}_{33}(\mathrm{i}\omega, \mathbf{q}=0)}. \tag{71}$$

Using the explicit expressions of polarization functions presented in appendix B it can be shown that (all functions

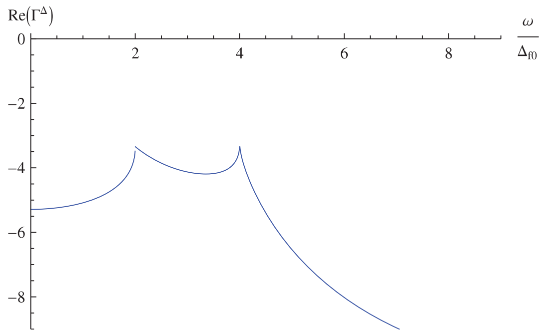

Figure 1. The plot of the real part of det $\Gamma^{\Delta}(\omega + i0^+)$ in arbitrary units. The input data are $\omega_D = 100$ meV, $\lambda_c = 0.2171$ and $\lambda_f = 0.6676$, which give $\Delta_{c0} = 20$ meV and $\Delta_{f0} = 10$ meV.

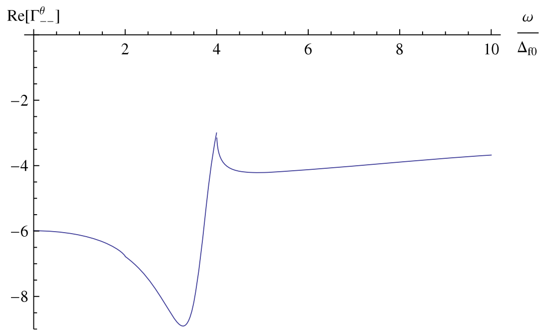

Figure 2. The plot of the real part of $\Gamma^{\theta'}_{--}(\omega + i0^+)$ in arbitrary units. The input data are identical with those of figure 1.

below are to be evaluated at $\mathbf{q} = 0$)

$$\frac{1}{2}\frac{\Delta_{c0}}{\Delta_{f0}}(\tilde{\pi}^c_{22} - D(q)[\pi^c_{23}]^2) = \frac{1}{2}\left(\frac{\omega}{2\Delta_0}\right)^2 \frac{D_c F_c D_f F_f}{D_c F_c + D_f F_f},$$
$$\frac{1}{2}\frac{\Delta_{f0}}{\Delta_{c0}}(\tilde{\pi}^f_{22} - D(q)[\pi^f_{23}]^2) = \frac{1}{2}\left(\frac{\omega}{2\Delta_0}\right)^2 \frac{D_c F_c D_f F_f}{D_c F_c + D_f F_f},$$
$$D(q)\pi^c_{23}(\zeta\pi^f_{23}) = \left(\frac{\omega}{2\Delta_0}\right)^2 \frac{D_c F_c D_f F_f}{D_c F_c + D_f F_f}. \tag{72}$$

The functions $F_{c/f}(i\omega)$ are defined by (B.4). Then from (72) it follows that

$$\Gamma^{\theta'}_{++}(i\omega, \mathbf{q} = 0) = 0, \qquad \Gamma^{\theta'}_{+-}(i\omega, \mathbf{q} = 0) = 0. \tag{73}$$

We also note that

$$\frac{\Gamma^{\theta'}_{++}(i\omega, \mathbf{q} \to 0)}{\Gamma^{\theta'}_{+-}(i\omega, \mathbf{q} \to 0)} = 2, \tag{74}$$

which will be useful later. To obtain the above results it is absolutely essential to take the screening effect by long range Coulomb interaction into account. This implies that the symmetric phase mode decouples and the collective mode in the phase sector is determined by the condition of Re $\Gamma^{\theta'}_{--}(\omega + i0^+, \mathbf{q} = 0) = 0$.

The collective excitations (of $\mathbf{q} = 0$) coming from the amplitude fluctuations are determined by

$$\mathrm{Re}[\det \Gamma^{\Delta}(i\omega \to \omega + i0^+, \mathbf{q} = 0)] = 0,$$
$$\det \Gamma^{\Delta}(i\omega, \mathbf{q} = 0) = \pi^c_{11}(i\omega, \mathbf{q} = 0) \times \pi^f_{11}(i\omega, \mathbf{q} = 0) - \left(\frac{2}{U}\right)^2. \tag{75}$$

And thanks to (73) the collective excitations (of $\mathbf{q} = 0$) from the phase fluctuations are determined by

$$\mathrm{Re}[\Gamma^{\theta'}_{--}(i\omega \to \omega + i0^+, \mathbf{q} = 0)] = 0. \tag{76}$$

Using (72), (76) can be rephrased as

$$2\left(\frac{-i\omega}{2\Delta_0}\right)^2 \mathrm{Re}\left(\frac{D_c F_c D_f F_f}{D_c F_c + D_f F_f}\right) - \frac{4}{U} = 0. \tag{77}$$

The spectrum of the collective excitations are to be obtained in the real frequency domain, so that analytic continuations ($i\omega \to \omega + i\delta$) are made in (75) and (76). Due to the complicated forms of the polarization functions, analytic solutions are not feasible. Instead, the equations are solved graphically using the representative input proposed in section 4.

Figure 1 shows that there are no zeros, which in turn implies that there are no collective excitations in the amplitude sector. The cusp structure at $\omega = 2\Delta_{f0}, 2\Delta_{c0}$ are to be expected.

Figure 2 shows that there collective mode does *not* exist in the phase sector, either. Thus we do not expect a sharp resonance behaviour coming from the collective mode in Raman response.

Now we turn to the discussion of the Anderson–Bogolyubov mode and the plasmon excitation. Since the Anderson–Bogolyubov mode and the plasmon excitation emerge from the phase fluctuation in the one-band superconductor case, it is natural to expect the similar will happen in the *symmetric* phase mode of two-band superconductors. Thus let us take a close look at $\Gamma^{\theta'}_{++}(q)$. Recall that $\Gamma^{\theta'}_{++}$ vanishes for $\mathbf{q} = 0$ (73). It turns out that the next leading terms in $|\mathbf{q}|$ come from the Taylor expansion of $\tilde{\pi}^{c/f}_{22}(i\omega, \mathbf{q})$ in $|\mathbf{q}|^2$, and from

$$\tilde{D}(q) = D(q) - D(i\omega, \mathbf{q} = 0) \approx \frac{V^{-1}_{\mathbf{q}}}{(\pi^c_{33} + \pi^f_{33})[V^{-1}_{\mathbf{q}} - (\pi^c_{33} + \pi^f_{33})]}, \tag{78}$$

where $\pi^{c/f}_{33}$ in the last line are to be evaluated at $\mathbf{q} = 0$. Now, recalling that $V^{-1}_{\mathbf{q},3D} \propto |\mathbf{q}|^2/e^2$, $V^{-1}_{\mathbf{q},2D} \propto |\mathbf{q}|/e^2$ and that $\pi^{c/f}_{23} \propto \omega$ (see (B.9)), we find

$$\Gamma^{\theta'}_{++}(i\omega, \mathbf{q}) \sim \begin{cases} |\mathbf{q}|^2\left[A_{3D}(i\omega) + \dfrac{\omega^2}{e^2}B_{3D}(i\omega)\right], & \text{3-dim,} \\ |\mathbf{q}|\left[|\mathbf{q}|A_{2D}(i\omega) + \dfrac{\omega^2}{e^2}B_{2D}(i\omega)\right], & \text{2-dim,} \end{cases} \tag{79}$$

where $A_{2D,3D}$ and $B_{2D,3D}$ are some functions of frequency, whose detailed forms do not concern us. In the three-dimensional case, the existence of the plasmon excitation with energy proportional to e^2 is evident from the expression in the bracket. The two-dimensional case does not seem to support the plasmon excitation, but that is due to the extremely two-dimensional band structure. For more realistic anisotropic three-dimensional band structure the plasmon excitation would show up.

In any case, in the $\mathbf{q} \to 0$ limit, the plasmon excitation is washed away by the $|\mathbf{q}|^2$ factor in front. However, this is just due to the structure of the density–density type correlation function. In terms of dielectric functions, we will have one more factor of $V_{\mathbf{q}}$ multiplied by the density–density correlation function, which cancels the factor of $|\mathbf{q}|^2$ in front. Then the plasmon excitation will be clearly visible.

To assess the importance of the screening effect of long range Coulomb interaction, it is useful to consider the collective excitations *in the absence of* the Coulomb interaction. At small $\omega, \mathbf{q}$ we find an Anderson–Bogolyubov mode with a velocity ($\omega = v_s|\mathbf{q}|$)

$$v_s = \left(\frac{D_c v_c^2 + D_f v_f^2}{D_c + D_f}\right)^{1/2}. \tag{80}$$

Note that this velocity coincides with the one obtained by Leggett ((3.18) in [12]). Apart from the Anderson–Bogolyubov mode, we do find one additional collective mode between $2\Delta_{f0}$ and $2\Delta_{c0}$ which is contrary to the previous case with Coulomb interaction included. Clearly this collective mode originates from the fluctuation of the relative phases, so that it belongs to the same category as that of Leggett's mode, and it clearly shows up in the Raman response. This mode between $2\Delta_{f0}$ and $2\Delta_{c0}$ originates from the interaction between the phases of two superconducting order parameters, but it behaves differently from the relative phase mode (Leggett mode) of *intraband* pairing two-band superconductors [12]. Leggett's collective mode is expected to appear *below* $2\Delta_{f0}$ (twice the smaller gap) [12]. However, our result is not in contradiction with that of Leggett since the condition for the existence of Leggett's collective mode is

$$\det V = V_{cc}V_{cf} - V_{cf}^2 > 0, \tag{81}$$

where V_{cc} and V_{ff} are the *intraband* pairing interactions, and V_{cf} is the *interband* pairing interaction. Our case corresponds to the one with $V_{cc} = V_{ff} = 0$ and $V_{cf} = U$, so that Leggett's condition is evidently violated. Thus we conclude that the collective excitation we have found belongs to the same category as that of Leggett's mode but in a different parameter regime. We also note that the condition (81) is independent of the sign of the interband pairing, which implies that we will obtain the same result for the *attractive* interband pairing interaction which does not have the relative phase between two gaps.

8. Raman scattering intensity

The functional integral over X_q of (58) can be done exactly, which yields (recall S^{r}_{J2}, W_q are given by (53) and (57), respectively)

$$Z[J] \approx \mathrm{e}^{-S_{\mathrm{sad}} - S^{\mathrm{r}}_{J2} + \frac{1}{2}\sum_q J_{-q}J_q(\sum_{i,j}\tilde{W}_{-q,i}[\Gamma^{\mathrm{r}}]^{-1}_{ij}W_{q,j})}. \tag{82}$$

Now the partition function has been expressed solely in terms of the source field J_q, and then from the definition of correlation function (10), the Raman response correlation function can be read off ($q = (\mathrm{i}\omega, \mathbf{q})$):

$$\begin{aligned}\chi_{\tilde{\rho}\tilde{\rho}}(q) &= \pi^c_{33,\gamma\gamma}(q) + \pi^f_{33,\gamma\gamma}(q) \\ &\quad + D(q)[\pi^c_{33,\gamma 1}(q) + \pi^f_{33,\gamma 1}(q)][\pi^c_{33,1\gamma}(q) + \pi^f_{33,1\gamma}(q)] \\ &\quad - \sum_{i,j}\tilde{W}_{q,i}[\Gamma^{\mathrm{r}}]^{-1}_{ij}W_{q,j}.\end{aligned} \tag{83}$$

The first two terms of (83) which come from S^{r}_{J2} represent the *quasiparticle contribution* and the $D(q)$ term from the Coulomb screening correction to it. The last term is the contribution from the collective excitations. Thus in our approach (within the saddle point plus Gaussian approximation) the contributions from the quasiparticles and the collective excitations are distinguished from each other. Evidently there should be processes contributing to the Raman response which mix quasiparticle and collective excitation, but these turn out to be subleading, at least in our approach. Whether this character of the separate contribution is true of the diagrammatic approach of [18, 19] is not clear. Note also that (83) is expressed entirely in terms of the (one-loop) polarization functions only, which are evaluated explicitly at $\mathbf{q} = 0$ in appendix B.

The Raman response requires the limit $\mathbf{q} \to 0$. The limit can be taken straightforwardly for the quasiparticle contribution, while for the collective excitation a careful treatment is necessary as will be shown below. In the limit $\mathbf{q} \to 0$, using the results of appendix B (especially (B.15)), it can be shown that ($\alpha_{c/f}$ are parameters characterizing the band anisotropy, which are defined by (B.16))

$$\begin{aligned}W_{\mathbf{q}=0}(\mathrm{i}\omega) &= (\alpha_f\gamma^f - \alpha_c\gamma^c) \\ &\quad\times\left[0, 0, \pi^c_{23}\frac{\pi^f_{33}}{\pi^c_{33}+\pi^f_{33}}, (\zeta\pi^f_{23})\frac{(-\pi^c_{33})}{\pi^c_{33}+\pi^f_{33}}\right], \\ \tilde{W}_{\mathbf{q}=0} &= -W_{\mathbf{q}=0},\end{aligned} \tag{84}$$

where all of the polarization functions are to be evaluated at $\mathbf{q} = 0$. Recall that $W_{\mathbf{q}}(\mathrm{i}\omega)$ is defined by (57). The first two entries corresponding to the amplitude block vanish since $\pi^{c/f}_{13}(\mathrm{i}\omega, \mathbf{q} = 0) = 0$, $\pi^{c/f}_{13,\gamma 1}(\mathrm{i}\omega, \mathbf{q} = 0) = 0$. Thus only the phase block contributes to the Raman response. However, this is a feature special to the $\mathbf{q} = 0$ limit, and at finite $\mathbf{q}$ the amplitude fluctuations do contribute.

Now the collective mode contribution (which entirely comes from the phase block) can be recast as

$$\begin{aligned}\chi_{\theta,\tilde{\rho}\tilde{\rho}}(\mathrm{i}\omega, \mathbf{q} = 0) &= +\sum_{i,j=c,f}W^\theta_{\mathbf{q}=0,i}[\Gamma^\theta]^{-1}_{ij}W^\theta_{\mathbf{q}=0,j}, \\ [W^\theta_{\mathbf{q}=0}]^{\mathrm{T}} &= (\alpha_f\gamma^f - \alpha_c\gamma^c) \\ &\quad\times\left[\frac{\pi^c_{23}\pi^f_{33}}{\pi^c_{33}+\pi^f_{33}}, \frac{(\zeta\pi^f_{23})(-\pi^c_{33})}{\pi^c_{33}+\pi^f_{33}}\right].\end{aligned} \tag{85}$$

Let us re-express (85) in the symmetric and the antisymmetric phase basis using the transformation (65). The result is (see (66), (69))

$$\chi_{\theta,\tilde{\rho}\tilde{\rho}}(\mathrm{i}\omega, \mathbf{q}=0) = \sum_{i,j=+,-} \tilde{W}^{\theta}_{\mathbf{q}=0,i}[\Gamma^{\theta'}]^{-1}_{ij}\tilde{W}^{\theta}_{\mathbf{q}=0,j},$$
$$\tilde{W}^{\theta}_{\mathbf{q}=0,i} = \sum_j (s^{\mathrm{T}})^{-1}_{ij} W^{\theta}_{\mathbf{q}=0,j}, \tag{86}$$

where the matrix s is the phase block (lower right corner) of the matrix S of (63). Using the results on the polarization functions in the appendices it can be shown easily that

$$\tilde{W}^{\theta}_{\mathbf{q}\to 0}(\mathrm{i}\omega) = (\alpha_f\gamma^f - \alpha_c\gamma^c) \times \left[\frac{\sqrt{2}}{(\omega/2\Delta_0)}\Gamma^{\theta'}_{+-}(\mathrm{i}\omega, \mathbf{q}\to 0), \frac{\sqrt{2}\omega}{2\Delta_0}\frac{D_cF_c\cdot D_fF_f}{D_cF_c + D_fF_f}\right]. \tag{87}$$

The first entry of (87) is actually zero as $\Gamma^{\theta'}_{+-}(\mathrm{i}\omega, \mathbf{q}\to 0) = 0$, but it is presented in this form since it should be multiplied by the matrix elements involving the singular limit of $\Gamma^{\theta'}_{+-}$ (see below). The inverse matrix $[\Gamma^{\theta'}]^{-1}$ in the limit $\mathbf{q}\to 0$ can be expressed as (arguments of matrix elements suppressed)

$$[\Gamma^{\theta'}]^{-1} = \frac{1}{\Gamma^{\theta'}_{++}\Gamma^{\theta'}_{--} - (\Gamma^{\theta'}_{+-})^2}\begin{bmatrix}\Gamma^{\theta'}_{--} & -\Gamma^{\theta'}_{+-}\\ -\Gamma^{\theta'}_{+-} & \Gamma^{\theta'}_{++}\end{bmatrix}$$
$$= \frac{1}{(\frac{\Gamma^{\theta'}_{++}}{\Gamma^{\theta'}_{+-}})\Gamma^{\theta'}_{--} - \Gamma^{\theta'}_{+-}}\begin{bmatrix}\frac{\Gamma^{\theta'}_{--}}{\Gamma^{\theta'}_{+-}} & -1\\ -1 & \frac{\Gamma^{\theta'}_{++}}{\Gamma^{\theta'}_{+-}}\end{bmatrix}$$
$$= \frac{1}{2\Gamma^{\theta'}_{--}}\begin{bmatrix}\frac{\Gamma^{\theta'}_{--}}{\Gamma^{\theta'}_{+-}} & -1\\ -1 & 2\end{bmatrix}. \tag{88}$$

In the last line of the above equation, we have used (74) and taken the limit $\Gamma^{\theta'}_{+-} \to 0$ where the limit is well defined. Plugging the result of (88) into (86) and again using $\Gamma^{\theta'}_{+-}(\mathrm{i}\omega, \mathbf{q}\to 0) = 0$, we find that

$$\chi_{\theta,\tilde{\rho}\tilde{\rho}}(\mathrm{i}\omega, \mathbf{q}\to 0) = \frac{2(\alpha_f\gamma_f - \alpha_c\gamma_c)^2}{\Gamma^{\theta'}_{--}(\mathrm{i}\omega, \mathbf{q}=0)} \times \left(\frac{\omega}{2\Delta_0}\right)^2 \left(\frac{D_cF_cD_fF_f}{D_cF_c + D_fF_f}\right)^2,$$
$$= (\alpha_f\gamma_f - \alpha_c\gamma_c)^2 \frac{(\frac{\omega}{2\Delta_0})^2(\frac{D_cF_cD_fF_f}{D_cF_c+D_fF_f})^2}{(\frac{\omega}{2\Delta_0})^2\frac{D_cF_cD_fF_f}{D_cF_c+D_fF_f} - \frac{2}{U}} \tag{89}$$

The quasiparticle contribution of the Raman response at $\mathbf{q}=0$ is

$$\chi_{\mathrm{qp},\tilde{\rho}\tilde{\rho}}(\mathrm{i}\omega, \mathbf{q}=0) = \pi^c_{33,\gamma\gamma} + \pi^f_{33,\gamma\gamma} - \frac{[\pi^c_{33,\gamma 1} + \pi^f_{33,\gamma 1}][\pi^c_{33,1\gamma} + \pi^f_{33,1\gamma}]}{\pi^c_{33} + \pi^f_{33}}. \tag{90}$$

The exact same result as (90) can be obtained by the conventional perturbative diagram summation method [18, 19].

The Raman vertex factor $\gamma_{\mathbf{k}}$ for the lattice with tetragonal symmetry in various scattering geometries is given by [20]

$$\gamma_{\mathrm{A_{1g}}}(\phi) = 1 + \gamma_A\cos 4\phi,$$
$$\gamma_{\mathrm{B_{1g}}}(\phi) = \gamma_{B_1}\cos 2\phi, \qquad \gamma_{\mathrm{B_{2g}}}(\phi) = \gamma_{B_2}\sin 2\phi. \tag{91}$$

The screening corrections vanish for $\mathrm{B_{1g}}$ and $\mathrm{B_{2g}}$ symmetry since the polarization functions $\pi^{c/f}_{33,\gamma 1}(\mathrm{i}\omega, \mathbf{q}=0) = \pi^{c/f}_{33,1\gamma}(\mathrm{i}\omega, \mathbf{q}=0)$ vanish. It is because all other integrands appearing in the evaluation of the polarization functions have $\cos(4\phi)$ dependence except for the $\mathrm{B_{1g,2g}}$ scattering vertices, $\cos(2\phi)$ and $\sin(2\phi)$, and these angular functions are orthogonal to each other. Therefore

$$\chi_{\mathrm{B_{1g},B_{2g}}} = \pi^c_{33,\gamma\gamma} + \pi^f_{33,\gamma\gamma}, \qquad \gamma = \gamma_{\mathrm{B_{1g},B_{2g}}}. \tag{92}$$

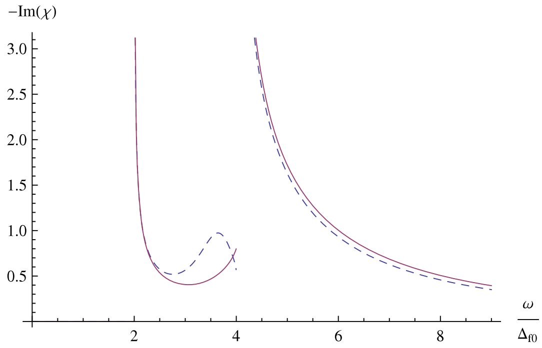

Figure 3. The Raman response function in $\mathrm{A_{1g}}$ scattering geometry in the unit of $D_c(\gamma^c_A)^2$. The anisotropy factors are chosen to be $\alpha_c = 0.3$, $\alpha_f = -0.27$, and the rest of the input data are identical with those of figure 1. The solid line is for the response function with both the quasiparticle contribution with Coulomb screening correction (90) and the phase fluctuation contributions (89) included, while the dashed line is for the quasiparticle contribution with Coulomb screening correction.

As for the $\mathrm{A_{1g}}$ geometry the factor of 1 for the $\mathrm{A_{1g}}$ geometry case is seen to be cancelled by the Coulomb correction, therefore, effectively $\gamma_{\mathrm{A_{1g}}}(\phi) \to \gamma_A\cos 4\phi$. The matrix element γ_A is determined by band structure. From (9) one can estimate $\gamma^{c/f}_A \sim t_{c/f}$.

The Raman scattering intensity for the $\mathrm{A_{1g}}$ geometry is plotted in figure 3. The inverse square root singularities at $\omega = 2\Delta_{f0}, 2\Delta_{c0}$ are well expected. The Coulomb correction is responsible for the peak structure between $2\Delta_{f0}$ and $2\Delta_{c0}$ in the quasiparticle contribution, while the contribution from phase fluctuation eventually suppresses the peak structure. The detailed behaviour depends sensitively on the band structure through the anisotropy $\alpha_{c/f}$ (B.16).

9. Summary and concluding remarks

We have developed a time-dependent Landau–Ginzburg theory for the Raman response of the two-band superconductors following the approach by Hertz [8]. Our approach can be generalized to a much wider class of problems of superconductivity. Unlike the standard diagrammatic approach, the (fluctuations of) order parameters appear explicitly throughout the course of development, which makes the understanding of the nature of collective excitations very clear.

Using the Hubbard–Stratonovich transformations the pairing interaction and the long range Coulomb interaction have been expressed in terms of the superconducting order parameter and the scalar potential, respectively. The functional integral technique enables us to integrate out the electrons completely, leaving us with the effective action of the order parameters. All of the interesting physical properties are encapsulated in the effective action (or time-dependent Landau–Ginzburg free energy). The spectrum of the collective excitations and the correlation functions (including Raman response) can be obtained from the effective action in a straightforward way.

This approach has been applied to the $s\pi$ pairing model of the Fe-pnictides. The Raman response was computed up to the Gaussian fluctuations with the four channels of symmetric and antisymmetric combinations of the phases and amplitudes of the two order parameters. The Raman spectra is composed of the quasiparticle and the phase collective mode contributions without mixing between them. There are no contributions from the symmetric or antisymmetric amplitude modes. The antisymmetric phase mode (Leggett mode) originates from the fluctuations of the relative phase of the two order parameters. It lies between twice the smaller gap and twice the larger gap. It is therefore damped by quasiparticles and its contribution to the Raman spectra is weak. It turns out that the long range Coulomb interaction eliminates the Leggett mode.

The weak Raman response of the Leggett mode can, of course, be expected. Consider the two cases of two-band superconductivity: interband-dominant and intraband-dominant pairings. In the former case, one solution gives pairing but the other does not. In the latter case, both solutions give pairing, albeit one is of lower T_c. Physically, it is the existence of this metastable solution that gives rise to the Leggett mode below twice the smaller gap [12]. For the $s_\pm$ state there is no metastable solution and we do not expect the Leggett mode response. The long range Coulomb interaction suppresses it completely by increasing the quasiparticle damping.

We are currently applying the present formalism to the spin–spin correlation function at the finite wavevector to elucidate this point and the distinct features of the (π, π) resonance mode in the $s\pi$ pairing state [22–24]. The result will be reported in a separate publication.

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Appendix A. Polarization functions

The polarization functions in the main text are defined as follows. $\gamma_{\mathbf{k}}^{c/f}$ are the Raman vertices defined in (9) and the Green functions $\hat{G}_{c/f}$ are defined in (34). τ_i with $i = 1, 2, 3$ are the Pauli matrices and the trace below is done over them. $\zeta_{c/f}$ is 1 for the c-band case and $-1 = \zeta$ for the f-band case:

$$\pi_{ij,\gamma\gamma}^{c/f}(\mathrm{i}\omega, \mathbf{q}) = \frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \gamma_{\mathbf{k}+\frac{\mathbf{q}}{2}}^{c/f}\gamma_{\mathbf{k}-\frac{\mathbf{q}}{2}}^{c/f} \times \mathrm{tr}(\hat{G}_{c/f}(\mathrm{i}\epsilon, \mathbf{k})\tau_i \hat{G}_{c/f}(\mathrm{i}\epsilon + i\omega, \mathbf{k}+\mathbf{q})\tau_j). \quad (A.1)$$

$$\pi_{ij,\gamma 1}^{c/f}(\mathrm{i}\omega, \mathbf{q}) = \frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \gamma_{\mathbf{k}+\frac{\mathbf{q}}{2}}^{c/f} \times \mathrm{tr}(\hat{G}_{c/f}(\mathrm{i}\epsilon, \mathbf{k})\tau_i \hat{G}_{c/f}(\mathrm{i}\epsilon + \mathrm{i}\omega, \mathbf{k}+\mathbf{q})\tau_j). \quad (A.2)$$

$$\pi_{ij,1\gamma}^{c/f}(\mathrm{i}\omega, \mathbf{q}) = \frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \gamma_{\mathbf{k}-\frac{\mathbf{q}}{2}}^{c/f} \times \mathrm{tr}(\hat{G}_{c/f}(\mathrm{i}\epsilon, \mathbf{k})\tau_i \hat{G}_{c/f}(\mathrm{i}\epsilon + \mathrm{i}\omega, \mathbf{k}+\mathbf{q})\tau_j). \quad (A.3)$$

$$\pi_{ij}^{c/f}(\mathrm{i}\omega, \mathbf{q}) = \frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \mathrm{tr}(\hat{G}_{c/f}(\mathrm{i}\epsilon, \mathbf{k})\tau_i \times \hat{G}_{c/f}(\mathrm{i}\epsilon + \mathrm{i}\omega, \mathbf{k}+\mathbf{q})\tau_j). \quad (A.4)$$

Below the trace over the Pauli matrices and the frequency summation are done at $T = 0$ for the polarization functions $\pi_{ij}^{c/f}(\mathrm{i}\omega, \mathbf{q})$, (A.4). The results will be identical for other polarization functions except for the insertion of Raman vertices since the Raman vertices $\gamma_{\mathbf{k}}^{c/f}$ do not depend on the Pauli matrices and the frequency. (Below $q = (\mathbf{q}, \mathrm{i}\omega)$.)

$$\pi_{11}^{c/f}(q) = 2\frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \frac{\mathrm{i}\epsilon(\mathrm{i}\epsilon+\mathrm{i}\omega) + \Delta_{c/f0}^2 - \epsilon_{c/f\mathbf{k}}\epsilon_{c/f\mathbf{k}+\mathbf{q}}}{[(\mathrm{i}\epsilon)^2 - E_{c/f\mathbf{k}}^2][(\mathrm{i}\epsilon+\mathrm{i}\omega)^2 - E_{c/f\mathbf{k}+\mathbf{q}}^2]} = \frac{1}{N}\sum_{\mathbf{k}} \frac{E_{c/f\mathbf{k}} + E_{c/f,\mathbf{k}+\mathbf{q}}}{E_{c/f\mathbf{k}}E_{c/f\mathbf{k}+\mathbf{q}}} \times \frac{-E_{c/f\mathbf{k}}E_{c/f\mathbf{k}+\mathbf{q}} + \Delta_{c/f0}^2 - \epsilon_{c/f\mathbf{k}}\epsilon_{c/f\mathbf{k}+\mathbf{q}}}{\omega^2 + (E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}})^2} \quad (A.5)$$

$$\pi_{22}^{c/f}(q) = 2\frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \frac{\mathrm{i}\epsilon(\mathrm{i}\epsilon+\mathrm{i}\omega) - \Delta_{c/f0}^2 - \epsilon_{c/f\mathbf{k}}\epsilon_{c/f\mathbf{k}+\mathbf{q}}}{[(\mathrm{i}\epsilon)^2 - E_{c/f\mathbf{k}}^2][(\mathrm{i}\epsilon+\mathrm{i}\omega)^2 - E_{c/f\mathbf{k}+\mathbf{q}}^2]} = \frac{1}{N}\sum_{\mathbf{k}} \frac{E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}}}{E_{c/f\mathbf{k}}E_{c/f\mathbf{k}+\mathbf{q}}} \times \frac{-E_{c/f\mathbf{k}}E_{c/f,\mathbf{k}+\mathbf{q}} - \Delta_{c/f0}^2 - \epsilon_{c/f\mathbf{k}}\epsilon_{c/f\mathbf{k}+\mathbf{q}}}{\omega^2 + (E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}})^2} \quad (A.6)$$

$$\pi_{33}^{c/f}(q) = 2\frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \frac{\mathrm{i}\epsilon(\mathrm{i}\epsilon+\mathrm{i}\omega) - \Delta_{c/f0}^2 + \epsilon_{c/f\mathbf{k}}\epsilon_{c/f\mathbf{k}+\mathbf{q}}}{[(\mathrm{i}\epsilon)^2 - E_{c/f,\mathbf{k}}^2][(\mathrm{i}\epsilon+\mathrm{i}\omega)^2 - E_{c/f\mathbf{k}+\mathbf{q}}^2]} = \frac{1}{N}\sum_{\mathbf{k}} \frac{E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}}}{E_{c/f\mathbf{k}}E_{c/f\mathbf{k}+\mathbf{q}}} \times \frac{-E_{c/f\mathbf{k}}E_{c/f\mathbf{k}+\mathbf{q}} - \Delta_{c/f0}^2 + \epsilon_{c/f\mathbf{k}}\epsilon_{c/f\mathbf{k}+\mathbf{q}}}{\omega^2 + (E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}})^2} \quad (A.7)$$

$$\pi_{12}^{c/f}(q) = 2\frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \frac{\mathrm{i}\epsilon_{c/f\mathbf{k}}(\mathrm{i}\epsilon+\mathrm{i}\omega) + \mathrm{i}\epsilon(-\mathrm{i}\epsilon_{c/f\mathbf{k}+\mathbf{q}})}{[(\mathrm{i}\epsilon)^2 - E_{c/f\mathbf{k}}^2][(\mathrm{i}\epsilon+\mathrm{i}\omega)^2 - E_{c/f\mathbf{k}+\mathbf{q}}^2]} = \frac{1}{N}\sum_{\mathbf{k}} \frac{\mathrm{i}\omega}{E_{c/f\mathbf{k}}E_{c/f\mathbf{k}+\mathbf{q}}} \times \frac{\mathrm{i}\epsilon_{c/f\mathbf{k}}E_{c/f\mathbf{k}+\mathbf{q}} + \mathrm{i}\epsilon_{c/f\mathbf{k}+\mathbf{q}}E_{c/f\mathbf{k}}}{\omega^2 + (E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}})^2} \quad (A.8)$$

$$\pi_{13}^{c/f}(q) = 2\frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \frac{\zeta_{c/f}\Delta_{c/f0}(\epsilon_{c/f\mathbf{k}} + \epsilon_{c/f\mathbf{k}+\mathbf{q}})}{[(\mathrm{i}\epsilon)^2 - E_{c/f\mathbf{k}}^2][(\mathrm{i}\epsilon+\mathrm{i}\omega)^2 - E_{c/f\mathbf{k}+\mathbf{q}}^2]} = \frac{1}{N}\sum_{\mathbf{k}} \frac{E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}}}{E_{c/f\mathbf{k}}E_{c/f\mathbf{k}+\mathbf{q}}} \times \frac{\zeta_{c/f}\Delta_{c/f0}(\epsilon_{c/f\mathbf{k}} + \epsilon_{c/f\mathbf{k}+\mathbf{q}})}{\omega^2 + (E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}})^2} \quad (A.9)$$

$$\pi_{23}^{c/f}(q) = 2\frac{T}{N}\sum_{\mathrm{i}\epsilon,\mathbf{k}} \frac{\mathrm{i}(\mathrm{i}\omega)\zeta_{c/f}\Delta_{c/f0}}{[(\mathrm{i}\epsilon)^2 - E_{c/f\mathbf{k}}^2][(\mathrm{i}\epsilon+\mathrm{i}\omega)^2 - E_{c/f\mathbf{k}+\mathbf{q}}^2]}$$
$$= \frac{1}{N}\sum_{\mathbf{k}} \frac{E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}}}{E_{c/f\mathbf{k}}E_{c/f\mathbf{k}+\mathbf{q}}} \times \frac{\mathrm{i}(\mathrm{i}\omega)\zeta_{c/f}\Delta_{c/f0}}{\omega^2 + (E_{c/f\mathbf{k}} + E_{c/f\mathbf{k}+\mathbf{q}})^2}. \quad \text{(A.10)}$$

After taking the trace over the Pauli matrices, the following relations among the polarization functions are easily seen:

$$\pi_{21}^{c/f}(\mathrm{i}\omega,\mathbf{q}) = -\pi_{12}^{c/f}(\mathrm{i}\omega,\mathbf{q}),$$
$$\pi_{31}^{c/f}(\mathrm{i}\omega,\mathbf{q}) = \pi_{13}^{c/f}(\mathrm{i}\omega,\mathbf{q}), \quad \text{(A.11)}$$
$$\pi_{32}^{c/f}(\mathrm{i}\omega,\mathbf{q}) = -\pi_{23}^{c/f}(\mathrm{i}\omega,\mathbf{q}).$$

Note that $\pi_{13}^{f} = \pi_{31}^{f}$ and $\pi_{23}^{f} = -\pi_{32}^{f}$ are proportional to the phase factor $\zeta = -1$.

Appendix B. Polarization functions at $\mathbf{q} = 0$

At $\mathbf{q} = 0$, the summands of the wavenumber summation depend on the wavenumber only through the energy bands. Introducing the density of states as follows:

$$D_{c/f}(\epsilon) = \frac{1}{N}\sum_{\mathbf{k}} \delta(\epsilon - \epsilon_{c/f\mathbf{k}}) \quad \text{(B.1)}$$

the wavenumber summation can be expressed as (h is a general function)

$$\frac{1}{N}\sum_{\mathbf{k}} h(\epsilon_{c/f\mathbf{k}}) = \int_{-\infty}^{\infty} \mathrm{d}\epsilon\, h(\epsilon) D_{c/f}(\epsilon)$$
$$\approx D_{c/f}(\epsilon = 0)\int_{-\omega_D}^{\omega_D} \mathrm{d}\epsilon\, h(\epsilon), \quad \text{(B.2)}$$

where $D_{c/f}(\epsilon = 0) \equiv D_{c/f}$ is the density of states at the Fermi energy and ω_D is a cutoff energy.

From the results of the previous section it can be easily shown

$$\pi_{11}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = -\left(\frac{1}{N}\sum_{\mathbf{k}}\frac{1}{E_{c/f\mathbf{k}}}\right) + \left(\Delta_{c/f0}^2 + \frac{\omega^2}{4}\right)\frac{1}{N}\sum_{\mathbf{k}}\frac{1}{E_{c/f\mathbf{k}}}\frac{1}{\omega^2/4 + E_{c/f\mathbf{k}}^2}. \quad \text{(B.3)}$$

Defining a dimensionless function $F_{c/f}(\mathrm{i}\omega)$ as follows:

$$F_{c/f}(\mathrm{i}\omega) = \int_{-\frac{\omega_D}{\Delta_{c/f0}}}^{\frac{\omega_D}{\Delta_{c/f0}}} \mathrm{d}x\, \frac{1}{\sqrt{x^2+1}} \frac{1}{\left(\frac{\omega}{2\Delta_{c/f0}}\right)^2 + x^2 + 1}. \quad \text{(B.4)}$$

Equation (B.3) can be expressed as

$$\pi_{11}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = -\left(\frac{1}{N}\sum_{\mathbf{k}}\frac{1}{E_{c/f\mathbf{k}}}\right) + \left(1 + \left(\frac{\omega}{2\Delta_{c/f0}}\right)^2\right) D_{c/f}F_{c/f}(\mathrm{i}\omega). \quad \text{(B.5)}$$

A simple integral shows that

$$\frac{1}{N}\sum_{\mathbf{k}}\frac{1}{E_{c/f\mathbf{k}}} = 2D_{c/f}\ln\frac{2\omega_D}{\Delta_{c/f0}} = \frac{\Delta_{f/c0}}{\Delta_{c/f0}}\frac{2}{U}, \quad \text{(B.6)}$$

where the last equality comes from the gap equation (27). Similarly, it can be shown that

$$\pi_{22}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = -\frac{1}{N}\sum_{\mathbf{k}}\frac{1}{E_{c/f\mathbf{k}}} + \left(\frac{\omega}{2\Delta_{c/f0}}\right)^2 D_{c/f}F_{c/f}(\mathrm{i}\omega). \quad \text{(B.7)}$$

Then we have (see (44))

$$\tilde{\pi}_{22}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = \left(\frac{\omega}{2\Delta_{c/f0}}\right)^2 D_{c/f}F_{c/f}(\mathrm{i}\omega). \quad \text{(B.8)}$$

Similarly, it is easily shown that

$$\pi_{33}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = -D_{c/f}F_{c/f}(\mathrm{i}\omega),$$
$$\pi_{23}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = \mathrm{i}\left(\frac{\mathrm{i}\omega}{2\Delta_{c/f0}}\right)\zeta_{c/f}D_{c/f}F_{c/f}(\mathrm{i}\omega), \quad \text{(B.9)}$$
$$\pi_{32}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = -\pi_{23}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0).$$

At $\mathbf{q} = 0$, the integrands of $\pi_{12}^{c/f}$ and $\pi_{13}^{c/f}$ are proportional to ϵ, making the integrands an odd function, so that their integral vanishes:

$$\pi_{12}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = \pi_{21}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = 0,$$
$$\pi_{13}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = \pi_{31}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = 0. \quad \text{(B.10)}$$

For the polarization functions with Raman vertices $\gamma_A^{c/f}\cos(4\phi)$ inserted, we have to take the average of the vertices over the Fermi surface [20]. This averaging has nothing to do with the particle–hole symmetry, so that the following hold (along with $(1 \leftrightarrow 2, 1 \leftrightarrow 3)$ counterparts)):

$$\pi_{12,\gamma\gamma}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = \pi_{12,\gamma 1}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = 0,$$
$$\pi_{13,\gamma\gamma}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = \pi_{13,\gamma 1}^{c/f}(\mathrm{i}\omega,\mathbf{q}=0) = 0. \quad \text{(B.11)}$$

In the case of $\pi_{33,\gamma_{A1g}\gamma_{A1g}}^{c/f}$ of A_{1g} scattering geometry, the relevant density of states is

$$D_{c/f,\gamma\gamma} = \frac{1}{N}\sum_{\mathbf{k}}(\gamma_A^{c/f})^2\cos^2(4\phi)\delta(\epsilon - \epsilon_{c/f\mathbf{k}}). \quad \text{(B.12)}$$

The average of $\cos^2(4\phi)$ provides just a factor of $1/2$, and we obtain

$$\pi_{33,\gamma_{A1g}\gamma_{A1g}}^{c/f}(\mathrm{i}\omega) = \frac{(\gamma_A^{c/f})^2}{2}\pi_{33}^{c/f}(\mathrm{i}\omega). \quad \text{(B.13)}$$

For B_{1g} and B_{2g} scattering geometries, $\cos^2(2\phi)$ and $\sin^2(2\phi)$ should be inserted instead, yielding the same result as (B.13).

In the case of $\pi_{33,\gamma_{A1g}1}^{c/f}$ and $\pi_{23,\gamma_{A1g}1}^{c/f}$, the relevant density of states is

$$D_{c/f,\gamma 1} = \frac{1}{N}\sum_{\mathbf{k}}(\gamma_A^{c/f})\cos(4\phi)\delta(\epsilon - \epsilon_{c/f\mathbf{k}}). \quad \text{(B.14)}$$

This density of states would vanish if it were not for the anisotropy of the energy bands. Using the energy band of (3) and the assumption of small anisotropy, we obtain

$$\pi^{c/f}_{33,\gamma_{A1g}1}(i\omega) = \alpha_{c/f}\gamma_A^{c/f}\,\pi^{c/f}_{31}(i\omega),\qquad \pi^{c/f}_{23,\gamma_{A1g}1}(i\omega) = \alpha_{c/f}\gamma_A^{c/f}\,\pi^{c/f}_{23}(i\omega), \tag{B.15}$$

where $\alpha_{c/f}$ is a dimensionless parameter characterizing the anisotropy of the energy band:

$$\alpha_{c/f} = \frac{t'_{c/f}\epsilon_{0c/f}}{t^2_{c/f}}, \qquad |\alpha_{c/f}| \ll 1. \tag{B.16}$$

Since the integral (B.4) is convergent in the limit $|x| \to \infty$, one can perform the integral with $\omega_D/\Delta_{c/f0} \to \infty$. To obtain the results in the real frequency domain we have to make an analytic continuation $i\omega \to \omega + i0^+$. Define the following complex functions $g(x)$:

$$\begin{aligned} g(x) &= g_R(x) + i g_I(x),\\ g_R(x) &= -\Theta(x-1)\frac{2\ln(x+\sqrt{x^2-1})}{x\sqrt{x^2-1}} + 2\Theta(1-x)\frac{\tan^{-1}\frac{x}{\sqrt{1-x^2}}}{x\sqrt{1-x^2}},\\ g_I(x) &= \pi\frac{\Theta(x-1)}{x\sqrt{x^2-1}}, \end{aligned} \tag{B.17}$$

where $\Theta(x)$ is a step function. Then a straightforward integral shows

$$F_{c/f}(i\omega \to \omega + i0^+) = g\left(\frac{\omega}{2\Delta_{c/f0}}\right). \tag{B.18}$$

Now all of the polarization functions at $\mathbf{q} = 0$ have been obtained in terms of the function $F_{c/f}$.

Appendix C. The gauge invariance and the conservation law of electromagnetic responses in two-band superconductors

In this appendix we show that the induced current of two-band superconductors in the presence of an external electromagnetic field satisfies the charge conservation law by generalizing a proof by Weinberg as presented in [25]. This implies, in particular, all of the electromagnetic response functions satisfy the charge conservation law (or, equivalently, Ward identities) since the response functions are obtained by taking the derivatives of the current with respect to the external electromagnetic fields.

We first note that in our model the spontaneously broken symmetry is the *diagonal* $U(1)$ symmetry:

$$c \to e^{i\alpha}c, \qquad f \to e^{i\alpha} f, \tag{C.1}$$

and the associated electric charge is

$$Q = e\sum_{\mathbf{x},\sigma}(c^\dagger_{\sigma\mathbf{x}}c_{\sigma\mathbf{x}} + f^\dagger_{\sigma\mathbf{x}}f_{\sigma\mathbf{x}}). \tag{C.2}$$

Each charge $e\sum_{\mathbf{x}}(c^\dagger c)$ and $e\sum_{\mathbf{x}}(f^\dagger f)$ is not conserved separately due to the interband pairing interaction. The general phase rotation:

$$c \to e^{i\alpha_c}c, \qquad f \to e^{i\alpha_f} f \tag{C.3}$$

is not a symmetry due to the interband pairing interaction. By writing

$$\alpha_{c/f} = \frac{\alpha_c + \alpha_f}{2} \pm \frac{\alpha_c - \alpha_f}{2} \tag{C.4}$$

one can see that the *symmetric* phase is the Goldstone field associated with the spontaneously broken $U(1)$ symmetry, while the antisymmetric phase is neutral.

In our approach, the fermions (c, f) are integrated out exactly via Hubbard–Stratonovich transformation, and we are left with the effective field theory of the bosonic fluctuating complex order parameters (Δ_c, Δ_f) interacting with electromagnetic fields. Introducing an external electromagnetic field A_μ, the partition function becomes

$$\begin{aligned} Z[A_\mu] &= \int D[\Delta_c, \Delta_f, \phi]\, e^{-S_B},\\ S_B &= \int_0^\beta d\tau \left[\sum_{\mathbf{x}} \frac{1}{U}[\Delta^*_{c\mathbf{x}}(\tau)\Delta_{f\mathbf{x}}(\tau) + \Delta^*_{f\mathbf{x}}(\tau)\Delta_{c\mathbf{x}}(\tau)] + \sum_{\mathbf{q}} \tfrac{1}{2}V_{\mathbf{q}}^{-1}\,\phi_{-\mathbf{q}}(\tau)\phi_{\mathbf{q}}(\tau)\right]\\ &\quad - \ln\det(\hat{M}_P(A_\mu) + \hat{M}_C(A_\mu)). \end{aligned} \tag{C.5}$$

Equation (C.5) is an exact result. Expressing the order parameters $\Delta_{c,f}$ in terms of amplitude and phase

$$\Delta_{c/f}(x) = |\Delta_{c/f}(x)|\, e^{i\theta_{c/f}(x)}, \tag{C.6}$$

it is clear that the amplitudes $|\Delta_{c/f}(x)|$ and the relative (antisymmetric combination) phase $\theta_c - \theta_f(x)$ are gauge-invariant quantities. Thus $\Delta^*_{c\mathbf{x}}(\tau)\Delta_{f\mathbf{x}}(\tau) + \Delta^*_{f\mathbf{x}}(\tau)\Delta_{c\mathbf{x}}(\tau)$ is also gauge-invariant. The external electromagnetic field is coupled via the kernel $\hat{M}_{P,C}$:

$$\hat{M}_P = \begin{pmatrix} \hat{K}_c & 0 \\ 0 & \hat{K}_f \end{pmatrix}, \tag{C.7}$$

where the kernel matrices are given by

$$\begin{aligned} \hat{K}_c &= \begin{pmatrix} \partial_\tau + iA_0 + \epsilon_c(-i\nabla - e\vec{A}) & \Delta_c \\ \Delta_c^* & \partial_\tau + iA_0 - \epsilon_c(-i\nabla - e\vec{A}) \end{pmatrix},\\ \hat{K}_f &= \begin{pmatrix} \partial_\tau + iA_0 + \epsilon_f(-i\nabla - e\vec{A}) & \zeta\Delta_f \\ \zeta\Delta_f^* & \partial_\tau + iA_0 - \epsilon_f(-i\nabla - e\vec{A}) \end{pmatrix}. \end{aligned} \tag{C.8}$$

Now perform the gauge transformations on $\Delta_{c/f}$ by $e^{i(\theta_c+\theta_f)/2}$ in the functional integral. Then the complex order parameters $\Delta_{c/f}$ are left only with the amplitude and the relative phase, which are gauge-invariant. Note that we do not apply gauge transformations on the external electromagnetic field. By the minimal coupling of gauge fields (covariant derivative), the gauge transformation modifies the derivative in the following way ($\theta_+ = (\theta_c + \theta_f)/2$):

$$\partial_\mu \to \partial_\mu - \mathrm{i}\partial_\mu \theta_+. \tag{C.9}$$

Of course, θ_+ is the Goldstone field associated with the spontaneously broken $U(1)$ symmetry and it generates a supercurrent. From this result one can see that the *longitudinal* electromagnetic response couples to the (longitudinal) supercurrent. After the gauge transformation the effective action takes the form

$$S_\mathrm{B} = S_\mathrm{B}[A_\mu - \mathrm{i}\partial_\mu \theta_+, |\Delta_{c/f}|, \theta_-]. \tag{C.10}$$

Then the induced current is given by

$$J_\mu = \left\langle \frac{\delta S}{\delta A^\mu} \right\rangle = \left\langle \mathrm{i} \frac{\delta S}{\delta \partial_\mu \theta_+} \right\rangle, \tag{C.11}$$

where the average is taken with respect to the partition functions $Z[A_\mu]$.

Now the equation of motion of θ_+ is

$$\left\langle \partial_\mu \frac{\delta S}{\delta \partial_\mu \theta_+} \right\rangle = \left\langle \frac{\delta S}{\delta \theta_+} \right\rangle = 0, \tag{C.12}$$

where the right-hand side becomes zero since the θ_+ appears in the action only as a derivative (recall θ_+ is a Goldstone boson). By (C.11) the equation of motion implies the current conservation:

$$\partial_\tau J^0 + \nabla \cdot \vec{J} = 0. \tag{C.13}$$

One can verify various Ward identities of response functions by taking the functional derivative of (C.13) with respect to A_μ (recall that J_μ is a functional of A_μ). Our study is equivalent to the computation of the induced current J_μ in the Gaussian approximation.

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