A Rigorous Formalism of Unconventional Symmetry Breaking in Fermi Liquid Theory and Its Application to Nematicity in FeSe

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Unconventional symmetry breaking due to nonlocal order parameters has attracted considerable attention in many strongly correlated metals. Famous examples are the nematic order in Fe-based superconductors and the star-of-David charge density order in kagome metals. Such exotic symmetry breaking in metals is a central issue of modern condensed matter physics, while its theoretical foundation is still unclear in comparison with the well-established theory of superconductivity. To overcome this difficulty, here we introduce the “form factor” that generalizes the nonlocal order parameter into the Luttinger-Ward (LW) Fermi liquid theory. We then construct a rigorous formalism of the “density-wave equation” that gives the thermodynamically stable form factor, similarly to the superconducting-gap equation. In addition, a rigorous expression of the Ginzburg-Landau free-energy for the unconventional order is presented to calculate various thermodynamic properties. In the next stage, we apply the derived formalism to a typical Fe-based superconductor FeSe, by using the one-loop LW function that represents the free-energy gain due to the interference among paramagnons. The following key experiments are naturally explained: (i) Lifshitz transition (=disappearance of an electron-pocket) due to the bond+orbital order below $T_c$. (ii) Curie-Weiss behavior of the nematic susceptibility at higher $T$, and the deviation from the Curie-Weiss behavior at lower $T$ near the nematic quantum-critical-point. (iii) Scaling relation of the specific heat jump at $T_c$, $\Delta C/T_c \propto T_c^3$ with $b \approx 3$. (Note that $b = 0$ in the BCS theory.) These results lead to a conclusion that the nematicity in FeSe is the bond-orbital order due to the “paramagnon interference mechanism”. The present theory paves the way for solving various unconventional phase transition systems.

I. INTRODUCTION

Recently, rich symmetry breaking phenomena due to unconventional order parameters have attracted considerable attention in various electron systems. Famous examples are the $C_2$-symmetric nematic order in various Fe-based superconductors [1–4]. It has been established that the nematic state is driven by the electron correlation, thanks to the “electronic nematic susceptibility” measurements performed by the shear-modulus analysis [5–7], the Raman spectroscopy [8–10], and the elastoresistivity measurements [11–14]. Similar nematic orders are also observed in magic-angle-twisted-bilayer-graphene [15, 16], titanium pnictide oxide [17], and cuprate superconductors [18–21]. Also, correlation-driven density-wave (DW) with nonzero wavevector ($\mathbf{q} \neq 0$) has attracted increasing attention recently. Famous examples are the star-of-David DW state in kagome metal CsV$_3$Sb$_5$ [22–25] the CDW states in cuprate superconductors [20, 21, 26–30], and the multopole DW states in heavy fermion systems [31–33]. Furthermore, more exotic odd-parity DW orders that accompany the charge or spin loop current have been discovered in kagome metals [34, 35], iridates [36], and cuprates [37].

We call these DW states “unconventional” because they have non-local and non-s-wave order parameters, in analogy to unconventional (non-s-wave) superconductivity. For example, the order parameter of the $d$-wave bond order is $O_{i,j} = \overline{O}(\delta_{x-x'}\delta_{y-y'},0-\delta_{x-x',0}\delta_{y-y'|1})$, where $(x, y)$ and $(x', y')$ are the integer coordinates of $i$ and $j$ sites [20, 21, 26, 38–41]. (In high contrast, the conventional magnetic order $m_i = n_{i\uparrow} - n_{i\downarrow}$ is local.) However, simple local spin-density-wave (SDW) is inevitably derived within the mean-field-level approximations for the Hubbard models with screened Coulomb interactions [42, 43]. Therefore, beyond-mean-field many-electron theories are necessary to understand the unconventional DW states. This is a difficult but very interesting theoretical problem, and this is a central issue of modern condensed matter physics. On the other hand, these unconventional DW states we are interested in are metallic, so the itinerant picture will be fruitful. In addition, the $U(1)$ gauge symmetry is preserved. Thus, it is promising to construct the formalism of the DW states on the basis of the microscopic Fermi liquid theory [44–47].

In general, the DW state at wavevector $\mathbf{q}$ originates from the particle-hole (p-h) pairing condensation, $D_{i}^\mathbf{q} = (1 - P_0)\langle c_{\mathbf{k} + \mathbf{q}, \sigma} c_{\mathbf{k}, \sigma} \rangle$, where $c_{\mathbf{k}, \sigma}$ is the electron annihilation operator, $\mathbf{k}$ is the momentum, and $\sigma (\pm 1)$ is the spin index [38, 42, 43]. $P_0$ represents the projection onto the totally-symmetric state with respect to the space-group and the time-reversal [42, 43]. Rich classes of the DW states are determined by the symmetry of the p-h condensation $D_{i}^\mathbf{q}$. For example, a simple spin-DW state is given as the $\hat{k}$-independent function $D_{i}^\mathbf{q} = m_\mathbf{q}$. The realized DW state $D_{i}^\mathbf{q}$ should be uniquely determined as the extremum of the free energy.
of the self-energy [42, 43]:

$$\delta t_{i,j}^{\sigma} \equiv (1 - P_0) \Sigma_{i,j}^{\sigma}. \quad (1)$$

In the absence of the DW order ($T \geq T_c$), we obtain $\delta t_{i,j} = 0$ by definition. When the DW order emerges ($T \leq T_c$), $\delta t_{i,j}$ becomes finite due to nonzero $D_{i,j}^{kq}$. Thus, $\delta t_{i,j}^{\sigma}$ is the energy-dimension order parameter of the DW state.

Here, we consider the DW state at a constant wavevector $q$. For convenience, we introduce the “form factor” $\delta t_{k}^{q\sigma}$, that is the Fourier transform of $\delta t_{i,j}^{\sigma}$ [42, 43]:

$$\delta t_{k}^{q\sigma} \equiv \frac{1}{N} \sum_{ij} \delta t_{i,j}^{\sigma} e^{-ik \cdot (r_i - r_j)} e^{-iq \cdot r_i}, \quad (2)$$

where $r_i$ is the position of site $i$. The classification of the symmetry of the form factor is presented in Sect. 1A.

The theoretical way to derive the form factor $\delta t_{k}^{q\sigma}$ has not been established yet. The aim of the present study is to establish an exact framework to derive $\delta t_{k}^{q\sigma}$, based on which we can construct reliable and useful approximate theories. In the statistical mechanics, the symmetry breaking with $\delta t_{k}^{q\sigma}$ occurs when the grand potential $\Omega$ is minimized. In other words, $\delta t_{k}^{q\sigma}$ is uniquely determined as the stationary condition with the minimum grand potential. In strongly correlated Fermi liquids, a rigorous formalism of the grand potential $\Omega$ is given by the Luttinger-Ward (LW) theory [45]. In the LW theory, the LW function $\Phi[G]$, which is the functional of electron Green function $G$, plays a central role. The self-energy $\Sigma$ and the irreducible two-particle interaction $I$ are uniquely derived from the functional derivatives of $\Phi[G]$ [45].

In this paper, we first introduced the “form factor” $\delta t_{k}^{q\sigma}$ into the LW theory to analyze the symmetry breaking at wavevector $q$. Its $(k - q/2)$-dependence represents the symmetry of the DW. We next derived the “DW equation” to obtain the form factor that minimize the LW ground potential below $T_c$. In this theory, the optimized $\delta t_{k}^{q\sigma}$ is uniquely obtained because the DW equation is equivalent to the thermodynamic stationary condition. This formalism enables us to study rich variety of electron-correlation-driven DW states ($\delta t_{k}^{q\sigma}$) without assuming any symmetry, like the analysis of the superconducting (SC) states ($\Delta_{k}^{\sigma\tau}$) based on the SC gap equation. In addition, we derive an exact expression of the Ginzburg-Landau (GL) free energy, $F \propto a_\delta \phi^2$, where $\phi$ is the amplitude of the DW order. The coefficient $a_\delta$ is uniquely related to the eigenvalue of the DW equation $\lambda_\delta$. Thus, we can calculate the thermodynamic properties and the stability of the DW state.

In the next stage, we apply the derived DW equation to FeSe by using the one-loop LW function $\Phi_{\text{FL}}[G]$ that represents the quantum interference among paramagnons. This theory naturally explains the following essential experimental reports: (i) Nematic Fermi surface (FS) and the Lifshitz transition due to bond+orbital order [48–51]. (ii) Curie-Weiss (CW) behavior of the nematic susceptibility $\chi_{\text{nem}}$ at higher temperatures, and the deviation from the CW behavior at lower temperatures near the nematic quantum-critical-point (QCP). [12–14, 52]. (iii) Scaling relation of the specific heat jump at $T_c$, $\Delta C/C \propto T_c^b$ $(b \sim 3)$. This relation naturally explains the smallness of (or undetected) $\Delta C/C$ reported in several nematic systems, such as RbFe$_2$As$_2$ ($T_c \sim 40$K) [53] and cuprate superconductors ($T_c \sim 200$K) [18, 19]. In cuprates, the nematic transition occurs at the pseudogap temperature $T^*$ [18, 19], while no anomaly in specific heat at $T = T^*$ has been reported previously.

Interestingly, recent experiments have revealed that the nematic QCP is clearly separated from the magnetic QCP in Fe(Se,S) and Fe(Se,Te) [54] in addition to Na(Fe,Co)As [55]. Such clear separation between two QCPs in addition to the key points (i)-(iii) are naturally understood by the present theory. Therefore, the nematicity in FeSe is the bond+orbital order due to the “paramagnon interference mechanism” [2, 3, 42, 56].

Here, we construct the DW equation based on the LW theory. The LW theory is expected to be valid for various strongly-correlated metals except for the vicinity of the localized Mott states. Thus, the present theory is expected to pave the path to understanding the useful concept of the “vestigial nematic order” from the itinerant picture.

A. Form Factor

Here, we discuss the rich variety of unconventional DW states [38, 42, 43, 57, 58] by classifying the symmetry of the form factor. The exotic DW states are represented by the non-local order parameter $\delta t_{ij}^{\sigma}$, which is parameterized by different site indices $i$ and $j$. Then, the effective hopping integral is $t_{ij}^{\sigma} = t_{ij}^{0\sigma} + \delta t_{ij}^{\sigma}$, where $t_{ij}^{0\sigma}$ is the original hopping integral with $A_{ij}$ symmetry. In conventional charge (spin) orders, the order parameter is expressed as $\delta t_{ij}^{\sigma} = +(-) \delta t_{ij}^{\sigma}$ with $i = j$, respectively.

In recent years, in contrast, unconventional non-local orders given by $\delta t_{ij}^{\sigma}$ with $i \neq j$ have been discovered and attracted increasing attentions. Here, we assume the Hermitian order parameter [42, 43]:

$$\delta t_{ij}^{\sigma} = (\delta t_{ij}^{\sigma})^*, \quad (3)$$

$$\delta t_{k}^{q\sigma} = (\delta t_{k}^{q\sigma})^*, \quad (4)$$

$\delta t_{ij}^{\sigma}$ is classified into four channels in terms of parity symmetry ($P = \pm 1$) and time-reversal symmetry ($T = \pm 1$) as shown in Fig.1 (a). Below, we discuss the case of $q = 0$. First, we consider the case of $\delta t_{ij}^{\sigma} = \delta t_{ij}^{\sigma}$. When $\delta t_{ij}^{\sigma}$ is real, the bond order with $(P, T) = (+1, +1)$ is realized. As an example, the $d$-wave bond order in square lattice model is shown in Fig. 1 (b). When $\delta t_{ij}^{\sigma}$ is pure imaginary, the charge-loop current (cLC) with $(P, T) = (-1, -1)$ is realized. Its form factor in $k$-space is $\delta t_k \propto k_x - k_y$. An example of the cLC order in anisotropic triangular lattice model is shown in Fig. 1 (c).
Next, we consider the case of $\delta t_{ij} = -\delta t_{ji}$. When $\delta t_{ij}$ is real, the spin-bond order with $(P, T) = (+1, -1)$ appears. When $\delta t_{ij}$ is pure imaginary, the spin-loop current (sLC) order with $(P, T) = (-1, +1)$ appears. An example of the sLC order is shown in Fig. 1 (d).

In addition, the translational symmetry is violated when the DW wavevector $q$ is nonzero. Furthermore, orbital orders [50], valley orders [16], and multipole orders [31] emerge when the Wannier functions possess multiple degrees of freedom. These rich non-local DW states are applicable for strongly correlated metals unless the perturbation treatment is violated. In the first half part of this paper, we consider the GL free energy for the magnetic transition:

$$F_{GL} = aM^2 + \frac{1}{2}bM^4.$$  

Here, $F_{GL} = \Omega_{MF} + \mu N$, where $\mu$ is the chemical potential. Based on Eq. (7), the coefficient $a$ is expressed as

$$a = \chi^0(0) \left( -1 + \frac{1}{\alpha_S} \right).$$  

This equation is consistent with the Stoner theory [60].

Here, we briefly review the mean-field theory of the ferromagnetic (FM) transition and derive a simplified DW equation based on the LW theory in later sections. It is also understood that the mean-field theory is insufficient to explain the nematic order. Below, we consider the following Hubbard model,

$$H = \sum_{i,j,\sigma} t_{ij}^0 c_{i\sigma}^\dagger c_{j\sigma} + H_1, \quad (5)$$

$$H_1 = \sum_{i} U c_{i\sigma}^\dagger c_{i\sigma}^\dagger c_{i\sigma} c_{i\sigma} = \frac{U}{4} \sum_{i} (n_i^2 - m_i^2), \quad (6)$$

where $n_i = n_{i\uparrow} + n_{i\downarrow}$ and $m_i = n_{i\uparrow} - n_{i\downarrow}$. The conduction electron energy is $\epsilon_k = \frac{1}{N} \sum_{j} t_{ij}^0 e^{i(r_i - r_j) \cdot k}$. In the mean-field approximation, the magnetic order is expressed as the spin-dependent $\delta t$: $\delta t_{ij}^0 = -\delta t_{ji}^0 = -\frac{U}{2} m_i$. Here, we set $M_i = U m_i/2$. In the case of the FM ($q = 0$) order, $M_i = M$. Thus, the grand potential is given by

$$\Omega_{MF} = \frac{T}{N} \sum_{k,\sigma} \ln G_{k\sigma}^{MF} + \frac{1}{U} M^2$$

$$= -\frac{1}{N} \sum_{k,\sigma} \ln (1 + e^{-\beta (\epsilon_k - \mu)}) + \frac{1}{U} M^2, \quad (7)$$

where $k \equiv (k, \epsilon_n = (2n + 1) \pi T)$, $\epsilon_{k\sigma} = \epsilon_k + M \sigma$, and $G_{k\sigma}^{MF} = (i\epsilon_n - \epsilon_{k\sigma} + \mu)^{-1}$ is the electron Green function. From the stationary condition of the $\Omega_{MF}$, which is given by $\partial \Omega_{MF}/\partial M = 0$, the mean-field equation for $M$ is obtained as

$$M = -\frac{U}{2N} \sum_{k\sigma} f(\epsilon_{k\sigma}) \sigma. \quad (8)$$

Next, we derive the linearized mean-field equation by linearizing the right-hand side of Eq. (8). It is given as

$$\alpha_S M = \frac{U}{N} \sum_{k} \left( -\frac{\partial n(\epsilon_k)}{\partial k} \right) M = U \chi^0(q = 0) M, \quad (9)$$

where $\alpha_S$ is the eigenvalue, which reaches unity at the critical temperature. $n(\epsilon)$ is the Fermi distribution function. In the mean-field approximation, $\alpha_S = U \chi^0(0)$, where $\chi^0(0)$ is the irreducible susceptibility at $q = 0$. $\alpha_S$ is called the spin Stoner factor. Equation (9) is the simplest example of the spin-channel DW equation with $I_{k'kq} = U$.

Here, we consider the GL free energy for the magnetic transition:

$$F_{GL} = aM^2 + \frac{1}{2}bM^4.$$  

This equation is consistent with the Stoner theory [60].

For the magnetic state at finite wavevector $q$, the eigenvalue of Eq. (9) is given as $\alpha_S(q) = U \chi^0(q, 0)$, and the $q$-dependent coefficient of the GL free energy becomes $a_q = \chi^0(q, 0) \left( -1 + \frac{1}{\alpha_S(q)} \right)$. Note that $a_q \approx a_q = 0 + \frac{1}{2} \sum_{\mu} c_{\mu} q_\mu^2$, where $c_{\mu} = \partial^2 \alpha_q / \partial q^2 |_{q=0}$. Here, $\chi^0(q, 0) = \frac{1}{N} \sum_{k} \frac{n(\epsilon_{k+q}) - n(\epsilon_k)}{\epsilon_k - \epsilon_{k+q}}$ is the irreducible susceptibility for general $q$. The relation $\alpha_S > \alpha_S(q)$ holds for $q \neq 0$ in ferromagnetic metals.

Considering the $T$-dependence of $\chi^0(0)$, the coefficient $a_q = 0$ at $T = 0$ is given as

$$a_q = 0 (T = 0) \approx \frac{\pi^2}{3} BT^2 c,$$  

where $B \equiv D''(0) + (D'(0))^2 / D(0)$, and $D(0)$ is the density of states at the Fermi energy. Thus, the necessary condition for the FM transition ($T_c > 0$) is given as $B < 0$ in the mean-field approximation.

However, recent experiments have revealed that the ferro-DW order appears even in the case of $B > 0$ in several strongly correlated electron systems such as Fe-based superconductors. Thus, to understand the ferro-DW transition, we have to go beyond the mean field theory. In addition, exotic nonlocal DW orders ($\delta t_{ij}$ with $i \neq j$) summarized in Fig. 1 cannot be explained within the mean-field approximation. To solve these difficulties, in this paper, we study the DW phase transition on the bases of the LW theory.

II. DERIVATION OF DW-EQUATION FROM LUTTINGER-WARD THEORY

The Luttinger-Ward (LW) theory in Ref. [45] provides an exact expression of the grand potential $\Omega$, which is applicable for strongly correlated metals unless the perturbation treatment is violated. In the first half part of this
In Eq. (14), $\Phi[G]$ is the Luttinger-Ward function which is given by calculating the all closed linked skeleton diagrams. Based on Eq. (14), we can define $\Omega$ as a functional of $\Sigma$ [61].

$$\Omega[\Sigma] = -\frac{T}{N} \sum_{k\sigma} \ln(-G_{k\sigma}^{\text{free}})^{-1} + \Sigma_{k\sigma}) + P[\Sigma],$$

(16)

where $P[\Sigma]$ is considered as the Legendre-transformation of $\Phi[G]$ introduced by Potthoff [61].

$$P[\Sigma] \equiv -\frac{T}{N} \sum_{k\sigma} G_{k\sigma} \Sigma_{k\sigma} + \Phi[G].$$

(17)

In deriving the GL free energy, we have to analyze $\Omega[\Sigma]$ in Eq. (16). Using the Luttinger-Ward function $\Phi$ and Potthoff function $F$ [61], the self-energy and Green function are respectively expressed as

$$\frac{\delta\Phi[G]}{\delta G_{k\sigma}} = \Sigma_{k\sigma}[G],$$

(18)

$$\frac{\delta P[\Sigma]}{\delta \Sigma_{k\sigma}} = -G_{k\sigma}[\Sigma].$$

(19)

Then, the functional derivations of $\Omega[G]$ and $\Omega[\Sigma]$ are
respectively given by
\[
\frac{\delta \Omega[G]}{\delta G_{\sigma\sigma'}} = G^{-1}_{\sigma\sigma'} - (G_{\sigma\sigma}')^{-1} + \Sigma_{\sigma\sigma'}[G], \tag{20}
\]
\[
\frac{\delta \Omega[\Sigma]}{\delta \Sigma_{\sigma\sigma'}} = \frac{1}{(G_{\sigma\sigma}')^{-1} - \Sigma_{\sigma\sigma'}} - G_{\sigma\sigma'}[\Sigma]. \tag{21}
\]

When $\Omega$ is stationary, the following Dyson equation is satisfied,
\[
\Sigma_{\sigma\sigma'}[G] = (G_{\sigma\sigma}')^{-1} - G_{\sigma\sigma'} \quad \text{(from Eq.}(20) = 0), \tag{22}
\]
\[
G_{\sigma\sigma'}[\Sigma] = \frac{1}{(G_{\sigma\sigma}')^{-1} - \Sigma_{\sigma\sigma'}} \quad \text{(from Eq.}(21) = 0). \tag{23}
\]

Based on the Luttinger-Ward theory, the ferro ($q = 0$) DW transitions are naturally described by the self-consistent equation (We call this the DW equation).

Here, we introduce the irreducible 4-point vertex $I_{kk'q}^{\sigma\sigma'}$ shown in Fig. 2 (a). It is a Jacobian connecting $\Sigma$ and $G$ as [61]
\[
\frac{\delta \Sigma_{k\sigma}}{\delta G_{k'\sigma'}} = I_{kk'q}^{\sigma\sigma'}, \tag{24}
\]
\[
\frac{\delta G_{k\sigma}}{\delta \Sigma_{k'\sigma'}} = \left(I_{kk'q}^{\sigma\sigma'}\right)^{-1}. \tag{25}
\]

Therefore, the following analytical relation [61] is obtained:
\[
\frac{T}{N} \sum_{k'q} \frac{\delta \Sigma_{k\sigma}[G]}{\delta G_{k'\sigma'}} \frac{\delta G_{k'\sigma'}[\Sigma]}{\delta \Sigma_{k'\sigma'}} = \delta_{kk'} \delta_{\sigma\sigma'}, \tag{26}
\]
which is exactly satisfied when $G$ is uniquely determined from $\Sigma$ via one-to-one correspondence.

The DW equation is derived from the following stationary conditions
\[
\frac{\delta \Omega_{\Sigma}[G]}{\delta \Sigma_{k\sigma}} \bigg|_{\Sigma^0} = 0, \quad \text{(at any } T) \tag{27}
\]
\[
\frac{\delta \Omega_{\Sigma}[G]}{\delta \Sigma_{k\sigma}} \bigg|_{\Sigma} = 0 \quad (T < T_c), \tag{28}
\]
where $\Sigma^0$ is the self-energy without any symmetry breaking, and $\Sigma$ is the stationary self-energy after the symmetry breaking. For $T > T_c$, the thermodynamic state is given by Eq. (27), which corresponds to the minimum of the free energy shown in Fig. 3 (a). For $T < T_c$, Eq. (28) gives the symmetry breaking state shown in Fig. 3 (b). (Eq. (28) corresponds to the unstable extremum.)

By using Eq. (28) and (21),
\[
\frac{\delta \Omega_{\Sigma}[G]}{\delta \Sigma_{k\sigma}} \bigg|_{\Sigma^0} = -G_{k\sigma}^2 \Sigma_{k\sigma} + \frac{\delta \Phi[G]}{\delta \Sigma_{k\sigma}} = -G_{k\sigma}^2 \Sigma_{k\sigma} + \frac{T}{N} \sum_{k'q} \delta G_{k'\sigma'}[\Sigma] \delta G_{k'\sigma'}[\Sigma][29]
\]
where $\delta G_{k'\sigma'}[\Sigma] = G^2_{k\sigma} \delta_{kk'}$. Thus, the stationary condition of Eq. (28) is rewritten as
\[
\frac{\delta \Phi[G]}{\delta G_{k\sigma}} \bigg|_{\Sigma^0} = \Sigma^0_{k\sigma}, \tag{30}
\]
\[
\frac{\delta \Phi[G]}{\delta G_{k\sigma}} \bigg|_{\Sigma} = \Sigma_{k\sigma}, \tag{31}
\]
where $\Sigma^0 = \left(G_{\sigma\sigma}' - \Sigma^0_{\sigma\sigma}'\right)^{-1}$. Equations (30) and (31) compose the “exact DW equation” that describes the DW state below $T_c$. Then, the order parameter is $\delta t_{k\sigma} = \Sigma_{k\sigma} - \Sigma^0_{k\sigma}$.

It is noteworthy that superconducting gap equation is derived from the stationary condition of the LW function in the SC state $\delta \Phi[G,F]$, where $F$ is the anomalous (normal) Green function [62]. Thus, the derived DW equation for the form factor, Eqs. (30) and (31), is well-founded comparable with the well-known superconducting-gap equation.

Next, we derive the linearized DW equation with respect to $\delta t$. By subtracting Eq. (30) from (31), we obtain
\[
\delta t_{k\sigma} = \frac{T}{N} \sum_{k'q} \delta^2 \Phi[G]_{k\sigma} \delta G_{k'\sigma'} + O(\delta G^2), \tag{32}
\]
where $\delta G \equiv \bar{G} - G^0$. Here, we rewrite $\delta t_{k\sigma}$ as
\[
\delta t_{k\sigma} = \phi \cdot f_{k\sigma}, \tag{33}
\]
where $\phi$ is a real parameter, and $f^x_k$ is the normalized order parameter that belongs to one of the irreducible representations in non-$A_1_g$ symmetry. It is convenient to set $\max_k |f_{k\sigma}| = 1$ because the relation $\phi = \max_k |\delta t_{k\sigma}|$ holds. Thus, we derive the following “linearize DW equation for $q = 0$” by introducing the eigenvalue $\lambda$ to the left-hand side of Eq. (32):

$$\lambda f_{k\sigma} = \frac{T}{N} \sum_{k', \sigma'} I^x_{kk'} (G^0_{k'\sigma'}) f_{k'\sigma'},$$

(34)

where we denote $I^x_{kk'} \equiv I^x_{kk'}/\Sigma_0$ to simplify the notation. In Eq. (34), the largest eigenvalue $\lambda$ reaches unity at $T = T_c$, and its eigenvector gives the form factor of the DW state.

The linearized DW equation can be generalized for finite $q$ orders as follows. First, we consider the DW order with the wavevector $q \equiv g \cdot m/n$ where $g$ is the reciprocal lattice vector, and $m, n$ are integers ($0 \leq m < n$). Then, we can introduce the $n \times n$-matrix Green function $G_{lm}^q = \langle k + q \mid \overline{G}(k + mq) \rangle$ where $l, m = 0 \sim n - 1$. In this case, Eq. (31) becomes

$$\delta \Phi[G] |_{G^m}^{\Sigma_m} = \Sigma_{l\sigma\sigma}. \quad \text{(35)}$$

Hereafter, we drop the overlines of $\bar{G}$ and $\bar{\Sigma}$ to simplify the notation.

Here, we adopt the extended Brillouin zone scheme for $\overline{k}$ to simplify the explanation. After that, the DW equation can be linearized with respect to the $q$-linear term in $\delta t$ by $\delta t_{m+1,m} \equiv \delta t_{m}^{q}$. By introducing the $q$-dependent eigenvalue, we obtain the following “linearize DW equation for general $q$”,

$$\lambda_q f^{q}_{k\sigma} = \frac{T}{N} \sum_{k', \sigma'} I^x_{kk'} q G^0_{k'\sigma'} G^0_{k'+q\sigma'} f^{q}_{k'\sigma'}. \quad \text{(36)}$$

where $q \equiv (q_x, q_y) = (q_x, q_y)$ is the wavevector and $\omega = 2\pi T$ is the boson Matsubara frequency. The condition $\lambda_q = 1$ brings the DW transition temperature $T_c$ with wavevector $q$, which can be interpreted as the particle-hole (p-h) gap equation. The form factor of the eigenvalue equation (36) contains the uncertainty of the phase factor $e^{iq\cdot x}$. The correct phase $\theta$ is uniquely fixed by following the Hermitian condition in Eq. (4).

The DW equation (36) is further simplified by introducing the spin ($s$) and charge ($c$) channel functions in the absence of the spin-orbit interaction [43]:

$$f^{c(s)} = f^s + (-) f^s. \quad \text{(37)}$$

$$f^{c(s)} = I^{s\uparrow} + (-) I^{s\downarrow}. \quad \text{(38)}$$

Finally, we derive the simplified “linearize DW equation for $x = s, c$ channel form factor at $q$” from Eq. (36) as follows:

$$\lambda^x_q f^x_k = \frac{T}{N} \sum_{k'} I^x_{kk'} G^0_k G^0_{k'+q} f^x_{k'}. \quad \text{(39)}$$

The right-hand side of Eq. (39) is shown in Fig. 2 (b). Therefore, we derived the exact expression of the DW equation in Eq. (36) or Eq. (39) composed of the true irreducible four-point vertex $I$ and the self-energy.

We can derive the expression of $\lambda^x_q$ from Eq. (39) as

$$\lambda^x_q = X^x_q / \chi^{0f}(q), \quad \text{(40)}$$

where $X$ is given as

$$X^x_q = \frac{T^2}{N} \sum_{k,k'} (f^x_{k})^* G^0_{k} G^0_{k+q} I^x_{kk'} q G^0_{k'+q} f^x_{k'}. \quad \text{(41)}$$

and $\chi^{0f}(q)$ is the irreducible susceptibility with the form factor $f^q_{k\sigma}$:

$$\chi^{0f}(q) = - \frac{T}{N} \sum_{k\sigma} f^q_{k\sigma} G^0_{k+q\sigma} G^0_{k-\sigma} f^{-q}_{k\sigma}. \quad \text{(42)}$$

From Eq. (40), the relation $\lambda^x_q \propto X^x_q$ is obtained because $\chi^{0f}(q)$ is nearly $T$-independent.

It is noteworthy that the DW equation introduced by Onari and Kontani et al. in Refs. [3, 20, 21, 42], which has been applied to iron-based and cuprate superconductors, is derived from the exact DW equation given in Eq. (36). The detailed derivation is shown in Appendix A.

Before closing this section, we reproduce the Stoner theory by applying the mean-field approximation to Eq. (39). In the mean-field theory,

$$I^s_{kk'q} = -U, \quad \delta \Sigma^q = M \delta q_0. \quad \text{(43)}$$

Therefore, the Eq. (39) is given by

$$\lambda^s M = - \frac{T}{N} \sum_{k} G^0_{k} G^0_{k} U M = U \chi_0 M. \quad \text{(44)}$$

Thus, the eigenvalue for the ferromagnetic transition corresponds to the Stoner factor $\alpha_S$:

$$\lambda^s = U \chi_0 \equiv \alpha_S. \quad \text{(45)}$$

### III. DERIVATION OF GL FREE ENERGY BASED ON THE LINEARIZED DW EQUATION

#### A. GL free energy for DW state with form factor $f$

Here, we derive the expression of the Ginzburg-Landau (GL) free energy based on the DW-equation. The GL free energy due to the ferro-DW transition ($\delta t_{k\sigma} = \phi f_{k\sigma}$) is expressed as

$$\Omega_{DW}(T, \mu, \phi) = a(T) \phi^2 + \frac{1}{2} b \phi^4. \quad \text{(46)}$$

Its schematic picture above $T_c$ ($a(T) > 0$) and that below $T_c$ ($a(T) < 0$) are shown in Figs. 3 (a) and (b), respectively.
Note that the coefficients $a$ and $b$ are functional of the form factor $f_{k\sigma}$. By using the $\Omega[\Sigma]$ defined in Eq. (16), the coefficient $a$ is calculated from the second functional derivation of $\Omega[\Sigma]$ as

$$
\Omega[\Sigma^0 + \delta t] - \Omega[\Sigma^0] \approx \frac{T}{N} \sum_{k\sigma k'\sigma'} \frac{\delta^2 \Omega[\Sigma]}{\delta \Sigma_{k\sigma} \delta \Sigma_{k'\sigma'}} \delta t_{k'\sigma'} \delta t_{kk'\sigma}. \tag{47}
$$

By using Eq. (21), we obtain

$$
\frac{\delta^2 \Omega[\Sigma]}{\delta \Sigma_{k'\sigma} \delta \Sigma_{kk'} \sigma} = \frac{\delta}{\delta \Sigma_{k'\sigma} \sigma} \left\{ \frac{1}{(G_{k\sigma}^{\text{free}})^{-1} - \Sigma_{kk'}} \right. - \frac{\delta G_{k\sigma}[\Sigma]}{\delta \Sigma_{k'\sigma}} \left. \right\} = G_{k\sigma}^2 \delta_{kk'} - \frac{ \left\{ I_{kk'}^\sigma \right\}^{-1} }{ \delta t_{kk'\sigma} \delta t_{kk'\sigma} } \tag{48}
$$

where we used the relation in Eq. (25). Therefore, the Eq. (47) is rewritten as

$$
\Omega[\Sigma^0 + \delta t] - \Omega[\Sigma^0] \approx \frac{T}{N} \sum_{k\sigma k'\sigma'} (G_{k\sigma}^0)^2 \left( \delta t_{kk'\sigma} \right)^2 \tag{49}
$$

Here, we recall that the order parameter $\delta t_{kk'\sigma} = \phi \cdot f_{k\sigma}$ is determined by using the DW equation (36). By using Eq. (36) together with Eqs. (24)-(26), Eq. (49) is rewritten as

$$
\Omega[\Sigma^0 + \delta t] - \Omega[\Sigma^0] \approx \left(1 - \frac{1}{\lambda}\right) \frac{T}{N} \sum_{k\sigma} (G_{k\sigma}^0)^2 f_{kk\sigma}^2 \tag{50}
$$

where the factor 2 originates from the spin degeneracy.

We can derive the GL free energy for the order parameter at nonzero wavevector $q$ by considering the large unit cell as we discussed in Sect. II. Thus, the coefficient $a$ defined in the Gibbs free energy in Eq. (46) is obtained as

$$
a_q(T) = -2 \chi_{qf}^0(0) \left( 1 - \frac{1}{\lambda_q} \right). \tag{51}
$$

As a result, we obtain the exact expression for coefficient $a_q$ by using the eigenvalue $\lambda_q$ in the DW equation. The obtained general expression in Eqs. (42) and (51) are meaningful to discuss the DW transition.

Finally, we stress that the Pottshoff’s Legendre-transformation of the LW formalism [61] is necessary to derive the correct GL free energy expression. In Appendix B, we explain that the expansion of (14) with respect to $\delta t$ leads to an inaccurate expression.

**B. GL free energy for BCS Superconductivity**

Here, we consider the GL equation for the spin-singlet superconductivity. Here, we express the spin-singlet SC gap function as $\Delta_k = \psi \cdot f_k$, where $f_k$ is the normalized form factor. Based on the LW theory for the superconducting states, we can derive that the second order GL parameter is given by

$$
a(T) = -2 \chi_{pp}^0(0) \left( 1 - \frac{1}{\lambda_{ac}} \right), \tag{52}
$$

$$
\chi_{pp}^0(0) = \frac{T}{N} \sum_k |\psi_k|^2 G_{k\sigma}^0 G_{-k\sigma}, \tag{53}
$$

where $\lambda$ is the eigenvalue of the linearized gap equation given by

$$
\lambda_{ac} f_k = \frac{T}{N} \sum_{k'} V_{kk'} G_{kk'}^0 G_{-k'\sigma}, \tag{54}
$$

where $V_{kk'} = \frac{\delta^2 f}{\delta \sigma_i \delta \sigma_j} |_{\Delta=0}$. Here, $F$ and $F^\dagger$ are anomalous Green functions. The derivation of Eq. (52) is essentially the same as that for the DW transition given in previous sections. We can show that the relationship (52) is also valid for the spin-triplet superconductivity.

**IV. NUMERICAL ANALYSIS OF NEMATIC STATE IN FeSe**

In this section, we explain the important unsolved problems in FeSe, which is one of the most famous Fe-based superconductors. We try to understand the following key topic on the nematicity, for both above and below the nematic transition temperature $T_c$, based on a unified theory. (i) Lifshitz transition below $T_c$. (ii) Nematic susceptibility above and below $T_c$. (iii) Specific heat jump at $T = T_c$. In FeSe, $T_c$ corresponds to the structure transition temperature $T_S$.

In previous sections, we derived the exact expressions of the linearized DW equation in Eq. (39) for $T > T_c$ and the full DW equation in Eqs. (30)-(31) for $T < T_c$. Here, we solve these equations for FeSe based on the one-loop approximation for the LW function, $\Phi_{\text{FLEX}}$, derived in Appendix A. We include the normal state (=without order parameter) self-energy $\Sigma^0$ into the DW equations because it is necessary to satisfy the stationary condition (30)-(31), although it was dropped in previous studies [3].

We study a realistic $d + p$ orbital Hubbard model with on-site multiorbital Coulomb interaction $U$ for FeSe:

$$
H = H_0 + v H_U, \tag{55}
$$

where $H_0$ is the $d + p$ orbital tight-binding model for FeSe, and $H_U$ is the $d$-orbital Coulomb interaction for $d + p$ orbital model given by the constrained-random-phase-approximation (crPFA) method. The matrix elements in
$H_U$ are composed of the intra-orbital Coulomb repulsion $U_{i,i}$, the inter-orbital Coulomb repulsion $U_{i,m}$ ($i \neq m$), and the exchange term $J_{i,m}$, as we explain in Appendix C.

In Eq. (55), $r(<1)$ is the reduction factor of $H_U$ that represents the screening due to p orbitals. According to Ref. [63], the averaged intra-orbital $U_{av} \sim 7eV$ is reduced to $\sim 4eV$ due to the screening effect by p-orbitals. In the present study, we set $r = 0.3 \sim 0.4$, where $T_c$ increases with $r$. In contrast, $T_c$ slowly decreases with $r$ for $r > 0.5$. Thus, obtained $T_c$ depends on $r$, while the symmetry and the form factor of the nematic order is sensitive to the choice of $r$. We note that the relation $\alpha_S < 1$ is satisfied for any $r$ in the present two-dimensional FeSe model because the FLEX approximation satisfies the Mermin-Wagner theorem [64]. This fact is favorable for realizing the nonmagnetic nematic state ($\lambda > 1$ and $\alpha_S < 1$).

In the present numerical study for FeSe, we use $64 \times 64 k$-meshes, and 4096 or 8192 Matsubara frequencies. Figure 4 (a) represents the Fermi surface (FS) of FeSe model. We derive the normal state self-energy $\Sigma^0$ by applying the FLEX approximation. In the case of $r = 0.36$, the obtained orbital-dependent mass-enhancement factors at $T = 10meV$ are about $z^{-1}_{xy} \sim 5$ and $z^{-1}_{xz,yz} \sim 3$, respectively. The Stoner factor is about 0.9 and its temperature dependence is very weak.

A. Above $T_c$: linearized DW equation analysis

First, we analyze the multi-orbital Hubbard model for FeSe based on the linearized DW equation in Eq. (39), with the kernel function in Eq. (A3). Here, we incorporate the normal state self-energy $\Sigma^0$ given in Eq. (A2) into the DW equation to perform the conserving approximation. $\Sigma^0$ is significant to derive realistic $T_c$ and beautiful CW/non-CW behaviors of $\chi_{nem}$, although it has been dropped in our previous analyses.

Here, we discuss the kernel function in Eq. (A3). The first line in Eq. (A3) gives the Hartree term, Maki-Thompson (MT) term, and the second and the third lines in Eq. (A3) give the Aslamazov-Larkin (AL) terms. Both MT and AL terms cause important “fluctuation-induced interaction for the DW”. In Fe-based SCs, the nematic order mainly originates from the AL terms, which represent the “interference between paramagnons” [2, 3, 56, 65]. The MT term is also important to induce the characteristic sign-reversing in the form factor in $k$-space [1, 3].

On the other hand, the cLC orders in geometrically frustrated Hubbard models mainly originate from the the MT terms [59]. Note that the MT terms induce striking non-Fermi-liquid transport phenomena near the QCPs [66].

Figure 4 (b) shows the $q$-dependence of the largest charge-channel eigenvalue $\lambda_q^0$ at $r = 0.40$ and $T = 5meV$ derived from the DW equation. (Below, we drop the superscript $c$ of $\lambda_q^0$ for simplicity.) The obtained $\lambda_q^0$ exhibit the maximum at $q = 0$ because the convolution of two $\lambda_q^0$s, $C_q^0 = \sum_p \lambda_p^0 \lambda_p^{q+q'}$, included in the AL-type VCs includes is largest at $q = 0$. Here, $\lambda_{q=0}$ exceeds unity, and typical transition temperature $T_c$ ($\sim 100K$) in Fe-based SCs is reproduced by including $\Sigma^0$. We note that similar results were obtained in Fig. S4 (a) in Ref. [67] by taking $\Sigma^0$ into account.

The obtained form factors belong to $B_{1g}$ symmetry, which is shown in Fig.4 (c): The $xz,yz$-orbital form factors express the $k$-dependent orbital polarization, which has been reported by previous DW equation studies without $\Sigma^0$ [3]. The obtained $xz,yz$-orbital polarization elongates the hole-pocket along the $k_y$-axis as experimentally reported in Refs. [48, 68]. In addition, the $xy$-orbital form factor the represents the $d$-orbital bond order $f \propto \cos k_x - \cos k_y$ emerges at the same $T_c$. This $d$-wave order leads to the disappearance of an electron-pocket around Y-point [49–51]. Thus, experimentally-observed ferro-nematic order in FeSe is naturally obtained. We stress that the coexistence of the $xz,yz$-orbital order and the $d_{2-x^2+y^2}$-wave bond order on $xy$-orbital was already reported in Fig. S3 (a)-(d) in Ref. [67]. In addition, the $d_{xy}$-wave bond order on $xy$-orbital has been studied in RbFe$_2$As$_2$ in Ref. [69].

We comment that a simple $A_{1g}$ symmetry order that accompanies the net charge order is suppressed by the Hartree term in the kernel function. In contrast, the $B_{2g}$ symmetry order in Fig. 4 (c) is free from the suppression by the Hartree term due to sign reversal in the form factor.

Figure 4 (d) shows the temperature-dependence of $\lambda_{q=0}(T)$ for $r = 0.40 \sim 0.32$. At higher temperatures ($T \gtrsim 10meV$), $\lambda_{q=0}(T)$ exhibits the $T$-linear behavior. The nematic susceptibility above $T_c$ is $\chi_{nem} = \lambda_{q=0}(T) - 1$, as proved theoretically in Ref. [69]. Therefore, it is confirmed that the experimental CW behavior of $\chi_{nem}$ at higher temperature is naturally explained in the present theory. The deviation from the CW behavior at lower temperatures will be discussed in Sect. VB.

Figure 4 (e) shows the obtained $R = (-\dot{\lambda}T_c)/\lambda$ as a function of $T_c$. Here, $\dot{\lambda} = \frac{d\lambda}{dT}$, $R$ approximately corresponds to $\lambda_{q=0}(T) = 0$, and $R$ becomes very small when $T_c \ll 10$meV. The reason is the recovery of the Fermi-liquid behavior $\lambda(0) - \lambda(T) \propto T^2$ because the system is far from the magnetic QCP. Also, the relation $R \propto T_c^2$ ($b \sim 3$) is satisfied in the present study. In Sect. VA, we will explain that $R$ is proportional to the jump in the heat capacity at $T_c$.

B. Below $T_c$: full DW equation analysis

Next, we analyze the full DW equation given as Eqs. (30)-(31) for $T < T_c$. We safely assume the uniform ($q = 0$) order parameter because $\lambda_q$ takes the largest value at $q = 0$ as found in Fig. 4 (b). The aim of this subsection is to explain the essential properties of the
derive the symmetry breaking part \( \Sigma \) given by Eq. (A2). Here, we calculate \( \sum \) in detail: The total self-energy is given in Eq. (15), and \( \delta t \) means the emergence of the ferro-DW order. The obtained eigenvalue \( \lambda \) at \( q = 0 \) in the BCS superconductivity. The inset shows the original band-dispersion is shown in Fig. 10 in Appendix C. The hole-pocket is elongated along the \( k \)-axis due to the \( d_{x^2-y^2} \)-wave form factor on the \( xz, yz \) orbitals. Interest-

nematic state \( T < T_c \) in FeSe based on the paramagnon interference mechanism.

Now, we explain the procedure of the numerical study in detail: The total self-energy is given in Eq. (15), where \( \Sigma^0 \) is the normal self-energy without any symmetry breaking given by Eq. (A2). Here, we calculate \( \Sigma^0 \) at each \( T \) by subtracting its static and Hermitian part, \( \Sigma^0, H(k) = (\Sigma^0(k, +i\delta) + \Sigma^0(k, -i\delta))/2 \), in order to fix the shape of the FS [70]. Next, we derive the symmetry breaking part \( \delta t \) self-consistently based on the following procedure: (a) We first calculate \( S_k \) as a function of the total self-energy. (b) Next, we derive \( \delta t \)

\[
\delta t_k = (1 - P_0)S_k, \tag{56}
\]

where \( P_0 \) is the projection operator for the totally-symmetric (\( A_{1g} \)) channel. (c) The total self-energy is given as \( \Sigma = \Sigma_0 + \delta t \). We repeat (a)-(c) till \( \delta t \) converges.

It is easy to show that the \( \delta t \)-linear term of \( S_k \) gives the right-hand side of the linearized DW equation (39) with the kernel function in Eq. (A3). Thus, the full-DW equation is equivalent to the linearized-DW equation when \( \delta t \) is very small.

Figure 5 (a) represents the obtained renormalized order parameter \( \delta t^*_{xy}(k) = \delta t_{m,m}(k) \) for \( m = xz \) and \( m = xy \) at \( Y \)-point. The obtained \( T_c = 12 \text{meV} \) completely coincides with that given by the linearized DW equation. The nematic order occurs as the second-order, and the averaged order parameter \( \delta t_{xy} \equiv (|\delta t_{xz}| + |\delta t_{yz}|)/2 \) at \( Y \)-point is about \( 2T_c \) at \( T = 5 \text{meV} \). Thus, the present theory gives the ratio \( \delta t^{\text{GW}}/T_c \sim 2 \), which is similar to the ratio \( \Delta_0^*/T_c^\text{SC} \sim 2 \) in the BCS theory. Thus, both the development of \( \chi_{\text{em}} \) above \( T_c \) and the nematic order parameter below \( T_c \) are well explained by the present theory.

The relation \( 2|\delta t^*_{x|y} \approx |\delta t^*_{xy} \) in Fig. 5 (a) indicates that both the \( (xz, yz) \)-orbitals and \( xy \)-orbital equally contribute to the nematic order. In Appendix D, we explain the relative phase between \( xz \)-orbital and \( xy \)-orbital form factors, \( \delta t_{xz,xz}(0, \pi) \delta t_{xy,xy}(0, \pi) < 0 \), on the basis of the Ginzburg-Landau analysis. This relation is significant for the Lifshitz transition below \( T_c \) as we will explain below.

Due to the nematic order parameter, the FS with \( C_4 \)-symmetry in Fig. 4 (a) is deformed to the \( C_4 \)-symmetry FS depicted in Fig. 5 (b) at \( r = 0.40 \) and \( T = 5 \text{meV} \). The corresponding band-dispersions in the normal state and that in the nematic state are shown in Figs. 5 (c) and (d), respectively. They are renormalized by the factor \( z \sim 5 \) for \( xy \)-orbital and \( z \sim 3 \) for \( xz, yz \) orbitals. The original band-dispersion is shown in Fig. 10 in Appendix C. The hole-pocket is elongated along the \( k_y \)-axis due to the \( d_{xz, yz} \)-orbital polarization [3, 48, 68]. Interestingly, the electron-pocket around \( Y \)-point disappears in the nematic state due to the \( d_{x^2-y^2} \)-wave form factor on the \( xy \)-orbital. This nematic Lifshitz transition has been confirmed by many angle-resolved photoemission spectroscopy (ARPES) studies [49, 50]. The relative phase between two form factors, \( \delta t_{xz,xz}(0, \pi) \delta t_{xy,xy}(0, \pi) < 0 \), originates from the kinetic energy gain due to the pseudogap formation by the Lifshitz transition.

In addition, in the nematic phase, the hole-pocket is elongated along the \( k_y \)-axis due to the \( xz, yz \)-orbital polarization with the sign-reversal in \( k \)-space. This has been also confirmed by ARPES studies [48, 68]. Thus, experimental key findings in the nematic states are satisfactorily reproduced by the present “paramagnon interference mechanism” [2, 3, 42, 56].

### C. Connection between above and below \( T_c \)

In the previous subsection, we derived the nematic self-energy \( \Sigma = \Sigma^0 + \delta t \) based on the full DW equation. The
Figure 5: (a) Obtained renormalized symmetry breaking self-energy $\delta t_{m,m}^*(0,k)$ for $m = xx, xy$ at Y-point, derived from the full DW equation at $r = 0.40$. The second-order transition occurs at $T_c = 12\text{meV}$, which is consistent with the linearized DW equation analysis in Fig. 4 (d). $T_L (= 9\text{meV})$ is the Lifshitz transition temperature. (b) FS in the nematic state at $T = 5\text{meV}$ ($< T_L$). The electron-pocket around Y-point disappears due to the $xy$-orbital form factor. In addition, the hole-pocket is elongated along the $k_y$-axis due to the $xz, yz$-orbital polarization with the sign-reversal in $k$-space. (c) Renormalized band structure in the normal state. (d) Renormalized band structure in the nematic state.

The derived nematic state corresponds to the stationary point of the LW grand potential $\Omega_{\text{LW}}[\Sigma]$, as proved in Sect. II. Because the obtained nematic state is thermodynamically stable, we can calculate the nematic susceptibility below $T_c$ on the bases of the linearized DW equation with the nematic self-energy.

Figure 6 exhibits the eigenvalue of the linearized DW equation in the nematic state ($\Sigma \equiv \Sigma^0 + \delta t$), $\lambda_{\text{full-DW}}$, in the case of $r = 0.40$. (We also show the DW equation eigenvalue with the normal self-energy $\Sigma^0$, $\lambda'(T)$, for reference.) We see that $\lambda(T)$ reaches unity at $T = T_c$, while it monotonically decreases for $T < T_c$. We find that $1 - \lambda(T) \approx |1 - \lambda'(T)|$ for $T \leq T_c$, as naturally expected in the GL theory. The beautiful numerical result in Fig. 6 means that the nematic-state derived from the full-DW method corresponds to the stationary point of $\Sigma$ very accurately. Thus, electronic states of FeSe both above and below $T_c$ are understood in a unified way based on the paramagnon interference mechanism.

The nematic susceptibility in the nematic state $T < T_c$ is $\bar{\chi}_{\text{nem}}(T = 0) = \chi^0(0)/(1 - \lambda'(T = 0))$, where $1 - \lambda(T = 0) \approx R$. Since $R (\propto T^3)$ is much smaller than unity as shown in Fig. 4 (e), the present study clarified that sizable nematic fluctuations remain in the nematic phase in Fe-based superconductors. This is an important information to understand the pairing mechanism in FeSe.

V. DISCUSSIONS

In this section, we discuss important unsolved properties in Fe-based SCs based on the present theory. We analyze the specific heat jump at $T = T_c$ in Sect. V A,
and calculate the $T$-dependence of $\chi_{\text{nem}}$ near the nematic QCP in Sect. V B.

### A. Jump in the specific heat at $T_c$ in FeSe

From the stationary point of the free energy $(\partial \Omega_{\text{DW}}(T, \mu, \phi)/\partial \phi|_{T, \mu} = 0)$, we obtain $\phi = 0$ and $\phi = \sqrt{-\frac{a(T)}{b}}$ above and below $T_c$, respectively. Here, we assume a simple $T$-dependence of $a$; $a(T) = \hat{a}(T - T_c)$, where $\hat{a} = -\frac{d a}{dT}|_{T_c} (> 0)$. Then, we obtain the BCS-like order parameter for $T < T_c$

$$\phi = \sqrt{\frac{\hat{a}}{b}}(T_c - T).$$

(57)

Then, the order parameter at $T = 0$ is $\phi_0 \approx \sqrt{(\hat{a}T_c)/b}$.

Based on the GL free energy, we discuss the jump of the heat capacity $\Delta C$ due to the DW transition, which is calculated by

$$\frac{\Delta C_{\text{DW}}}{T_c} = -\frac{d^2 \Omega_{\text{DW}}}{dT^2}|_{T=T_c} = (\hat{a}T_c) \left( \frac{\phi_0}{T_c} \right)^2.$$

(58)

Now, we calculate $\hat{a}$ based on Eq. (50). In FeSe, $\chi_{\text{eff}}(0)$ is almost independent of $T$, while $-\lambda \equiv -\frac{d \lambda}{dT}|_{T_c}$ takes a large positive value as shown in Fig. 4 (d), due to the AL-type VCs in the kernel function $I$. Then, we obtain

$$\langle \hat{a}T_c \rangle = 2\chi_{\text{eff}}(0)(-\hat{\lambda}T_c).$$

(59)

Note that $\chi_{\text{eff}}(0)$ is equal to the DOS projected by the form factor $f$, $D_f(0) \equiv \frac{1}{T} \sum_{k} \delta(\epsilon(k) - \mu) f_k^2$, in the absence of the self-energy.

Hereafter, we explicitly consider the mass-enhancement factor due to the self-energy, $z^{-1} \equiv 1 - \partial \text{Re} \Sigma / \partial \epsilon|_{\epsilon=\mu}$. The relation $z^{-1} \approx 1$ holds in general strongly correlated metals. In the Fermi liquid theory, the Green function is given as $G_k = z/(i\epsilon_n - \epsilon_k - \mu)$. Then, the DOS is changed to $\chi_{\text{eff}}(0) = z\chi_f(0) = zD_f(0)$, and the observed renormalized order parameter is $\phi_0^* = z\phi_0$. Thus, $\Delta C_{\text{DW}}/T_c$ due to the nematic transition is given by

$$\frac{\Delta C_{\text{DW}}}{T_c} = 2z^{-1}D_f(0)R \left( \frac{\phi_0^*}{T_c} \right)^2,$$

(60)

where $R = (-\hat{\lambda}T_c)$. As we summarized in Fig. 4 (e), $R \sim 0.3$ for $r = 0.40$, and $R \sim 0.1$ for $r = 0.36$. In contrast, $\Delta C_{\text{SC}}/T_c^{\text{SC}}$ due to the BCS superconductivity is $\Delta C_{\text{SC}}/T_c^{\text{SC}} = 2z^{-1}D_f(0)\left( \psi_0^*/T_c^{\text{SC}} \right)^2$ with $\Delta = \psi \cdot f$, which corresponds to $R = 1$ in Eq. (60). ($\psi_0^* \equiv z\psi_0$ is the observed gap function.) Because $\psi_0^*/T_c^{\text{SC}} \approx 2$, we obtain $\Delta C_{\text{SC}}/T_c^{\text{SC}} \approx 8z^{-1}D_f(0)$, which is close to $9.4z^{-1}D_f(0)$ in the BCS theory. Because $\phi_0^*/T_c^{\text{SC}} \approx 2$ in the present numerical study, we obtain the relation

$$\frac{\Delta C_{\text{DW}}}{T_c} \sim R \frac{\Delta C_{\text{SC}}}{T_c^{\text{SC}}}.$$

(61)

In the present theory, $R \propto T_c^b$ with $b \sim 3$ for $r = 0.40 \sim 0.34 (T_c = 12 \sim 6\text{meV})$ as shown in Fig. 4 (e). Thus, the relation $\Delta C_{\text{DW}}/T_c \propto T_c^b$ is predicted by the present theory.

![Figure 7](image-url)

**FIG. 7:** (a) Obtained eigenvalue $\lambda_q$ for RbFe$_2$As$_2$ model at $r = 0.30$ [69]. The obtained form factor at $q = 0$ is the $d_{xy}$-wave bond order on $xy$-orbital. Here, we take the renormalization factor $z = 1/2$ into account by following Ref. [2]. (b) $T$-dependence of $\lambda_q = 0$ in RbFe$_2$As$_2$ model.

Next, we discuss the nematic state in RbFe$_2$As$_2$, which is a heavily hole-doped Fe-based superconductor. This system exhibits the uniform ($q = 0$) nematic order. Interestingly, the observed nematicity possesses the $d_{xy}$-wave symmetry, whose director is 45-degrees rotated from the $d_{xy}$-$y^2$-wave nematicity in FeSe. Figure 7 (a) shows the DW equation eigenvalue in the RbFe$_2$As$_2$ Hubbard model, which was introduced in Ref. [69]. The obtained form factor at $q = 0$ is the $d_{xy}$-wave bond order on $xy$-orbital, as we revealed in Ref. [69]. Here, we set $\Sigma^0 = (1 - z^{-1})(i\epsilon_n - \mu)$ by following Ref. [2], instead of calculating the FLEX self-energy. We set the renormalization factor $z = 1/2 (= m/m^*)$. Then, $T_c$ is renormalized to be $T_c' = zT_c$, and $\lambda_{q=0}(T)$ is equal to $\lambda_q^0(zT)$ [2].

Figure 7 (b) exhibits the $T$-dependence of $\lambda_{q=0}$. Here, the relations $R \sim 0.1$ when $T_c \sim 10\text{meV}$ and $R \propto T_c^b$ ($b \sim 2.5$) are obtained in the RbFe$_2$As$_2$ model. This result indicates that the relation $R \sim 0.01$ is satisfied at $T_c \sim 40\text{K}$. In fact, in FeSe model, we obtained the
relation \( R \propto T_c^b \) (\( b \approx 3 \)) at low \( T_c \) (= \( 6 \sim 12 \) meV) by using fine \( k \)-meshes (64\(^2\)) and many Matsubara frequencies (8192) to obtain reliable results at low \( T \); see Fig. 4 (e). Similar relation is expected to be realized in other Fe-based superconductor models within the same paramagnon interference mechanism. Therefore, the present theory gives a natural explanation why \( \Delta C_{\mathrm{DW}} \) in RbFe\(_2\)As\(_2\) (\( T_c \approx 40 \)K) reported in Ref. [53] is much smaller than that in FeSe (\( T_c \approx 90 \)K) in Ref. [71].

### B. CW/non-CW behavior in Nematic susceptibility

Here, we discuss the nematic susceptibility \( \chi_{\mathrm{nem}} \) due to the electron correlation. According to Ref. [67, 69], the nematic susceptibility is given as

\[
\chi_{\mathrm{nem}} = zDf(0) \frac{1}{1 - \lambda(T)},
\]

where \( \lambda(T) \) is the eigenvalue of the DW equation with the “normal self-energy” \( \Sigma^0 \). Figure 8 (a) shows the normalized susceptibility \( \bar{\chi}_{\mathrm{nem}} \equiv \chi_{\mathrm{nem}}/\chi_{\mathrm{nem}}^0 = 1/(1 - \lambda(T)) \) for \( r = 0.40 \) and 0.34, which corresponds to \( T_c = 12 \)meV and 6.2meV respectively. In both cases, \( \bar{\chi}_{\mathrm{nem}} \) follows the CW behavior at higher temperatures (\( T > T^* \approx 10 \)meV). In contrast, at lower temperatures (\( T < T^* \) for \( r = 0.34 \), \( \bar{\chi}_{\mathrm{nem}} \) exhibits a clear deviation from the CW behavior. The Weiss temperature \( T_0 \) is derived from Fig. 8 (b): We see that \( \lambda(T) \) changes from T-linear to \( T^2 \)-like at \( T \approx T^* \approx 8 \)meV. Similar Fermi-liquid behavior in \( \lambda(T) \) is also recognized in Fig. 4 (d). This result is natural because the system is far from the magnetic QCP. Thus, the present theory provides a natural explanation for the non-CW \( \chi_{\mathrm{nem}} \) near the nematic QCP with \( T_c \approx 0 \) reported in Refs. [12].

Here, we discuss the reason why \( \chi_{\mathrm{nem}} \) exhibits the CW/non-CW behavior depending on \( r \). According to Eq. (40), the eigenvalue \( \lambda_q \) at \( q = 0 \) is proportional to \( X \) in Eq. (41) because \( \chi^{\phi \phi}(q = 0) \) is almost constant. In FeSe, the nematic state is mainly caused by the “paramagnon interference AL term” in \( X_q \). It is approximately given as

\[
X_{q=0} = T \sum_{Q,m} 3|A_{Q,q=0}^{f,m}|^2(U^2 \chi^2_{Q}).
\]

where \( \chi^{s,m} \) is the spin susceptibility for \( d \)-orbital \( m \), and \( A_{Q,q=0}^{f,m} \) is the three-point vertex composed of three Green functions:

\[
A_{Q,q=0}^{f,m} = -T \sum_p \{ \hat{G}_p^{0} f_q^{0} \hat{G}_p^{0} \}_{m,m} \{ \hat{G}_p^{0} \}_{m,m},
\]

where \( \hat{G}_p^{0} \) is the matrix representation of the multiorbital Green function with FLEX self-energy. Their expressions are shown in Fig. 8 (c). In a simple single-orbital model, the analytic expression of Eq. (64) is given as \( A_{Q,q=0}^{f,m} = \frac{1}{N} \sum_k \left( -\frac{\partial n(\epsilon_k)}{\partial \epsilon_k} \right) \frac{1}{\epsilon_k - Q - \epsilon_k} f_{q=0}^{0} \), where \( n(\epsilon) \) is the Fermi distribution function [72]. When \( Q \approx Q_{\mathrm{nesting}} \), \( A_{Q,q=0}^{f,m} \) exhibits strong enhancement at low temperatures due to \( (-\partial n(\epsilon_k)/\partial \epsilon_k) \) [2, 72].

In FeSe, the spin Stoner factor \( \alpha_S \) is nearly constant for \( T > T_c \). Then, the strong \( T \)-dependence of \( \lambda(T) \) originates from \( A_{Q,q=0}^{f} \), not from \( V_q^{\phi} \), as we explained in Ref. [2]. The paramagnon interference magnifies the nematic susceptibility, but its magnification is nearly constant. To confirm this fact, we introduce a simplification of \( X_{q=0} \) in Eq. (63) as follows:

\[
X_{q=0} = T \sum_{Q,m} |A_{Q,q=0}^{f,m}|^2.
\]

Figure 8 (d) shows the numerical result of \( X_{q=0} \) at \( r = 0.34 \). For \( T \gtrsim 10 \)meV, \( X_{q=0} \) exhibit almost perfect \( T \)-linear behavior. Its increment at low \( T \) originates from the FS nesting. In contrast, at lower temperatures, \( X_{q=0} \) starts to saturate when \( T \) is smaller than the nesting energy scale [2]. This saturation gives rise to the non-CW behavior of \( \chi_{\mathrm{nem}} \) near the nematic QCP (\( T_c \approx 0 \)) as shown in Fig. 8 (a).

Thus, the paramagnon interference mechanism satisfactorily explains both the CW behavior of \( \chi_{\mathrm{nem}} \) above \( T^* \approx 10 \)meV and its non-CW behavior below \( T^* \). These behaviors are actually observed in various Fe-based superconductors near the nematic QCP: Ba(Fe,T)\(_2\)As\(_2\) with T\(_c \)\( \approx \)Co,Ni, (Ba,A)Fe\(_2\)As\(_2\) with A\( = \)K,Rb, and Fe(Se,\( \phi \)) with Pn\( = \)Te,S [14, 52, 54]. In this mechanism, \( A_{Q,q=0}^{f} \) is the coupling constant between the nematicity and the paramagnons, and its increment leads to large \( \chi_{\mathrm{nem}} \) at low temperatures. Once the nematic order is established below \( T_c \), the spin Stoner factor \( \alpha_S \) increases as we discussed in [3, 73].

In the present mechanism, the deviation from the CW behavior of \( \chi_{\mathrm{nem}} \) below \( T^* \) is equal to the Fermi-liquid behavior \( \lambda(0) - \lambda(T) \propto T^2 \). This deviation is naturally expected when the nematic QCP is well separated from the magnetic QCP, even in the absence of impurities. We will discuss this point in the Summary section.

### VI. SUMMARY

In this paper, we derived a formally exact “density-wave (DW) equation”, by introducing the form factor of the DW state \( \delta_q^{\phi \phi} \) into the LW theory. Its solution automatically satisfies the extremum condition of the grand potential. By solving the DW equation, the optimized form factor and its wavevector are uniquely obtained for both above and below \( T_c \). This formalism enables us to perform the Baym-Kadanoff conserving approximation that is essential to obtain thermodynamic stable states. In addition, we derive an exact expression of the Ginzburg-Landau (GL) free energy, \( F \propto \alpha q \phi^2 \), where \( \phi \)
is the amplitude of the DW order at wavevector \( q \). The coefficient \( a_q \) is uniquely related to the eigenvalue of the DW equation \( \lambda_q \). This formalism enables us to calculate various thermodynamic properties of the DW state.

In the second part, we analyzed the nematic state in FeSe based on the derived DW equation based on a realistic multiorbital Hubbard model with one single parameter \( r \). We explained the following key experiments in Fe-based SCs: (i) Lifshitz transition due to bond+orbital order \([49, 50]\). (ii-1) The CW behavior of \( \chi_{\text{nem}} \propto 1/(1 - \lambda(T)) \) at higher-temperatures; 1 - \( \lambda(T) \propto T_0 - T \). (ii-2) Deviation from the CW behavior of \( \chi_{\text{nem}} \) at low temperatures near the nematic QCP without magnetic criticality; \( \lambda(0) - \lambda(T) \propto T^2 \) \([12, 13, 52]\). (iii) A scaling relation \( \Delta C/T_c \propto T_c^b \) \((b \sim 3)\) that naturally explains the smallness of \( \Delta C/T_c \) reported in several nematic systems \([18, 19, 53]\). This is because the gain of the free energy in the nematic transition is much smaller than that in the SC state. In addition, we explain the (iv) Nematic QCP away from the magnetic QCP observed in Fe(Se,S), Fe(Se,Te), \([54]\) and Na(Fe,Co)As \([55]\).

The present theory naturally explains the essential points (i)-(iv). Thus, it is concluded that the nematicity in FeSe is the bond+orbital order due to the “paramagnon interference mechanism” depicted in Fig. 8 (c) \([2, 3, 42, 56]\).

The behavior (ii-2) has been observed in various Fe-based superconductors near the nematic QCP: Ba(Fe,T)\(_2\)As\(_2\) with \( T = \text{CoNi}, (\text{Ba,A})\text{Fe}_2\text{As}_2\) with \( \text{A}=\text{K,Rb, and Fe(Se,Pn)} \) with Pn=Te,S \([14, 54]\). This behavior is frequently ascribed to the impurity-induced Griffiths phase, while it is widely observed insensitively to the impurity potential strength. (For example, the quantum oscillation is observed in Te- and S-doped FeSe.) In the present theory, the behavior (ii-2) is naturally explained when the nematic QCP is well separated from the magnetic QCP, even in the absence of impurities. It is useful to verify the relation \( \lambda(0) - \lambda(T) \propto T^2 \) experimentally. In the present mechanism, the increment of \( \chi_{\text{nem}} \) at low \( T \) originates from the \( T \)-dependence of \( \Lambda_Q, q = 0 \) in Eq. (64) \([2]\), and the self-energy due to thermal spin fluctuations \([66, 74]\) is also important to derive a perfect CW behavior of \( \chi_{\text{nem}} \). The self-energy due to nematic fluctuations will be also important (\( |\Sigma_{\text{nem}}| \geq |\Sigma_{\text{FLEX}}| \)) adjacent to the nematic QCP, unless the dynamical nematic fluctuations are suppressed by the acoustic phonons. This is an important future issue. It is considered that a perfect

![Diagram](image_url)

FIG. 8: (a) Nematic susceptibility \( \bar{\chi}_{\text{nem}} = 1/(1 - \lambda(T)) \) for \( r = 0.34 \) and 0.40 as a function of \( T \). \( \bar{\chi}_{\text{nem}} \) follows the CW behavior \((C/(T - T_0)) \) at higher temperatures. However, it deviates from the CW behavior at low temperatures. (b) Derivation of the Weiss temperature \( T_0 \) from \( \lambda(T) \). (c) The “paramagnon interference AL term” \( X_{q=0} \) that is proportional to \( \lambda_q \). \( \Lambda_{q=0}(Q) \) is the three-point vertex. (d) Obtained \( X_{q=0} \) as a function of \( T \). \( X_{q=0} \) starts to saturate at low temperatures, consistently with the deviation from the CW law in \( \bar{\chi}_{\text{nem}} \).
CW behavior for 30-250K observed in BaFe$_2$(As$_{0.3}$P$_{0.7}$)$_2$ [14] is ascribed to the magnetic criticality due to the coincidence of the nematic and magnetic QCPs.

The present theory paves the way for understanding various unconventional phase transition systems for both above and below $T_c$. For example, the analysis of the odd-parity DW order accompanying spontaneous current, which has been reported in cuprates and kagome metals recently, is an important future problem [43, 59]. The (local and/or non-local) multipole order physics in metals recently, is an important future problem [43, 59].

It is important to develop the numerical method beyond the one-loop approximation, the functional-renormalization-group method [42, 59, 75–77], which is equivalent to the one-loop approximation. the functional-renormalization-group method [42, 59] is ascribed to the magnetic criticality due to the coexistence of the nematic and magnetic QCPs.

The exotic superconductivity mediated by the DW fluctuations [22, 78–80] would be a very interesting future problem.

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Appendix A: Justification of Onari-Kontani approximation in the DW equation

Here, we derive the DW equation introduced by Onari and Kontani et al. in Refs. [3, 20, 21, 42], which has been applied to iron-based and cuprate superconductors, from the exact DW equation given in Eq. (36). In the present calculation, we apply the fluctuation-exchange (FLEX) approximation for $\Phi_{FLEX}[G]$. It is given as [62]

$$\Phi_{FLEX} = T \sum_q \text{Tr} \left\{ \frac{3}{2} \ln(1 - U^s \chi^0_{q}) + \frac{1}{2} \ln(1 - U^c \chi^0_{q}) \right\}$$

$$+ \frac{T}{4} \sum_q \text{Tr} \left\{ (U^s \chi^0_{q})^2 + (U^c \chi^0_{q})^2 \right\}$$

$$+ T \sum_q \text{Tr} \left\{ \frac{3}{2} U^s \chi^0_{q} + \frac{1}{2} U^c \chi^0_{q} \right\}. \quad (A1)$$

which is expressed in Fig.9 (a). Here, $U^s(c)$ is the spin-channel (charge-channel) Coulomb interaction, and $U^s = -U^c = U$ in the single-orbital Hubbard model. Their matrix expressions in multiorbital systems are introduced in the next section.

In the framework of the conserving approximation, the first-order derivative of $\Phi_{FLEX}[G]$ gives the self-energy $\Sigma$.

It is expressed as [81, 82]

$$\Sigma_k^0 = \frac{T}{N} \sum_q G_{k+q}^0 W_q^0, \quad (A2)$$

which is expressed in Fig.9 (b). Here, $W_q^0 = \left( \frac{3}{2} V_q^s + \frac{1}{2} V_q^c \right)$, $V_q = U^x + U^x \chi^x_q U^x$ ($x = s, c$), and $\chi^x_q = \chi^x_0(q)/(1 - U^x \chi^0_0(q))$.

Finally, we derive the irreducible four-point vertex $I$ from the second derive of $\Phi_{FLEX}[G]$. The derived charge-channel kernel function in the DW equation (39) for $x = c$ is given by [20, 43]

$$I_{kk'=q}^c = -\frac{3}{2} V_{k-k'}^c \frac{1}{2} V_{k-k'}^c$$

$$+ \frac{T}{N} \sum_p \left[ \frac{3}{2} V_{p+q}^c V_{p+q}^c \right] G_{k-p}^0 G_{k'-p}^0$$

$$+ \frac{T}{N} \sum_p \left[ \frac{3}{2} V_{p+q}^c V_{p+q}^c \right] G_{k-p}^0 G_{k'-p}^0, \quad (A3)$$

which is depicted in Fig.9 (c). Note that the double-counting $U^2$-terms in Eqs. (A3) and (A2) should be subtracted properly.

(a) $\Phi[G] = \quad (b) \Sigma_k = \quad (c) \lambda_q f_q^s = \quad$

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{fig9.pdf}
\caption{(a) Diagrammatic expressions of $\Phi_{FLEX}[G]$ in the one-loop approximation. For simplicity, diagrams in the single-orbital Hubbard models are shown, while we study multi-orbital Hubbard models in this paper. (b) Self-energy $\Sigma_k = \delta \Phi_{FLEX}/\delta G_k$. (c) Linearized DW equation with the kernel function $I$ derived from the second derivative of $\Phi_{FLEX}[G]$ with respect to $G$. The Maki-Thompson (MT) term and the Aslamazov-Larkin (AL) terms give the “fluctuation-induced interaction for the DW”.

Although the DW equation with the kernel in Eq. (A3) is an approximation, it satisfies the Baym-Kadanoff’s conserving laws by introducing $\Sigma_{FLEX}$. That is, the solution of the DW equation is the thermal equilibrium state derived from the stationary condition of $\Omega_{FLEX}$. Thus,
the Onari-Kontani type DW equation [3, 20, 21, 42] is given by dropping $\Sigma_{\text{FLEX}}$ from Eq. (A3). Furthermore, the present exact DW equation is useful to go beyond the Onari-Kontani’s approximation.

**Appendix B: GL free energy from $\Omega[G]$**

In the main text, we derive the GL free energy based on the grand potential $\Omega[\Sigma]$. Here, we show a different way to obtain GL free energy by using $\Omega[G]$ starting from Eq. (14) in the main text. The coefficient $a$ is derived from the second functional derivation of $\Omega[G]$ as

$$
\frac{\delta^2 \Omega[G]}{\delta G_{k_\sigma} \delta G_{k_\sigma'}} \bigg|_{G=G^0} = - (G_0^0)^{-2} \delta_{k_\sigma k_\sigma'} - I_G^{\sigma \sigma'}.
$$

Thus, we obtain the following results

$$
\Omega[G] - \Omega[G^0] = \sum_{k} \frac{1}{N} \sum_{\sigma,\sigma'} G_{k_\sigma}^{-2} \delta_{G_{k_\sigma} G_{k_\sigma'}}^2 - \frac{T}{N} \sum_{k} I_G^{\sigma \sigma'} \delta_{G_{k_\sigma} G_{k_\sigma'}}
$$

We also explain the multiorbital Coulomb interaction term $H_U$. The multiorbital Coulomb interaction term is expressed as $H_U = \frac{1}{2} \sum_{\sigma, \sigma'} \sum_{i,m} U^{\sigma \sigma'} U^{\sigma \sigma'}_{m,m'} U^{\sigma \sigma'}_{i,m,i'\sigma'} U^{\sigma \sigma'}_{i',m',i'\sigma} d_i^{\sigma} d_i^{\sigma'} d_i^{m} d_{i'}^{m'}$, where $l, m$ represent the orbital indices, in $\sigma = +1 (-1)$ represents the $\uparrow$ ($\downarrow$) spin, $i$ is the site index, and $U^{\sigma \sigma'} = -U^{\sigma} - \sigma^{\sigma'} U^{\sigma}$. The matrix elements of $U^\sigma$ is given by

$$
(U^\sigma)_{i,l,i,l} = \begin{cases} U_{l,l}, & l_1 = l_2 = l_3 = 4 \\
U_{l,l}, & l_1 = l_3 \neq l_2 = 4 \\
J_{l,l}, & l_1 = l_2 \neq l_3 = 4 \\
J_{l,l}, & l_1 = l_4 \neq l_2 = 4 \\
0, & \text{otherwise}.
\end{cases}
$$

Also, the bare Coulomb interaction for the charge channel is

$$
(U_c)_{i,l,i,l} = \begin{cases} -U_{l,l}, & l_1 = l_2 = l_3 = 4 \\
U_{l,l} - 2J_{l,l}, & l_1 = l_3 \neq l_2 = 4 \\
-2U_{l,l} + J_{l,l}, & l_1 = l_2 \neq l_3 = 4 \\
-J_{l,l}, & l_1 = l_4 \neq l_2 = 3 \\
0, & \text{otherwise}.
\end{cases}
$$

Here, $U_{l,l}$, $U_{l,l}'$ and $J_{l,l}$ are the first-principles Coulomb interaction terms given in Ref. [63].
In the main text, we omit the orbital indices of the Green functions and the Coulomb interactions to simplify the expressions. It is straightforward to write the orbital indices of these expressions in multiorbital models by using Eqs. (C1)-(C3).

Appendix D: Relative phase between $xz$-orbital and $xy$-orbital form factors

As we discussed in the main text, the relation $2|\delta_{xz}| \approx |\delta_{xy}|$ in Fig. 5 (a) indicates that both the $(xz + yz)$-orbitals and $xy$-orbital equally contribute to the nematic order in FeSe. The $(xz + yz)$-orbital polarization $[xy$-orbital bond order] originates from the spin fluctuations on $(xz + yz)$-orbitals $[xy$-orbital]. Here, we discuss the relative phase between $xz$-orbital and $xy$-orbital form factors, $\delta_{xz,xy}(0, \pi) \times \delta_{xy,xy}(0, \pi) < 0$, based on the Ginzburg-Landau analysis.

According to Eq. (48) or (49), the second-order inter-orbital free-energy is $E^{(2)}_{\phi} = -T \sum \delta_{n', n} \phi_{n'} \phi_n$, where $\delta_{n', n} = -T \sum G_{\phi}(k)G_{\phi}(k) f_{n'}(k) f_n(k)$ and $\delta_{m, m}(k) = f_{m, m}(k) \phi_m$. We verified numerically that $\delta_{xy,xy} > 0$, which is consistent with the relations $\chi_{2,4}(0) < 0$ in FeSe and $\delta_{\phi, \phi}(0) < 0$ around Y-point shown in Fig. 4 (c). Thus, the relation $\phi_{xy,xy} > 0$ is realized, and therefore the Lifshitz transition occurs in FeSe. In other words, the relation $\phi_{xy,xy} > 0$ (i.e., $\delta_{\phi, \phi}(0, \pi) \delta_{\phi, \phi}(0, \pi) < 0$) is a direct consequence of the multiorbital band structure of Fe-based superconductors.

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