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Ab initio theory of superconductivity in a magnetic field. I. Spin density functional theory for superconductors and Eliashberg equations

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We present a first-principles approach to describe magnetic and superconducting systems and the phenomena of competition between these electronic effects. We develop a density functional theory SpinSCDFT by extending the Hohenberg-Kohn theorem and constructing the noninteracting Kohn-Sham system. An exchange-correlation functional for SpinSCDFT is derived from the Sham-Schlüter connection between the SpinSCDFT Kohn-Sham and a self-energy in Eliashberg approximation. The reference Eliashberg equations for superconductors in the presence of magnetism are also derived and discussed.

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I. INTRODUCTION

In this work, we present how magnetic and superconducting (SC) properties can be computed on the same footing and from first principles by extending the density functional theory (DFT) framework. In developing this spin DFT for SC (SpinSCDFT), we will restrict ourselves to situations where currents are negligible and only consider the effect of the Zeeman term of the Hamiltonian. Under this assumption, we can exclude the occurrence of the Abrikosov vortex state [1] that, having a mesoscopic characteristic length scale, would be beyond the present computational power for a fully *ab initio* method.

The expulsion of static magnetic fields from the bulk [2] is one of the most spectacular properties of SC materials and illustrates the profound competition between magnetic and SC behavior. The SC-magnetic interaction generates, in fact, a large number of interesting phenomena on which the scientific community has focused attention. Some of the most investigated are the Abrikosov vortices [1] and the variety of fascinating effects occurring in heterostructures [3], such as stacked layers of magnetic with SC material (see Ref. [4] for a review). Among these effects is also the FFLO state, named after Fulde, Ferrel [5], Larkin, and Ovchinnikov [6], where strong exchange fields induce a SC state with a finite momentum pairing. Evidently, this state was recently observed experimentally [7,8] in heavy-fermion SC, many years after its prediction. In addition, triplet SC has been observed in several systems [9–15], and is usually associated to ferromagnetism.

Among the many effects generated by the interplay of magnetism and superconductivity, some have an intrinsic microscopic nature and could be accessible to first-principles calculations, in particular, we refer to the sharp suppression of the critical temperature due to paramagnetic impurities [16], and the surprising evidence of coexisting phases between singlet SC and local magnetism, in particular, close to a magnetic phase boundary [14,17–19] where high-T_c SC occurs [20,21]. We devote this work to set the ground for an *ab initio* theory to describe these physical effects.

We will start our formulation from the Pauli Hamiltonian in Sec. II. In Sec. III, we formulate a density functional theory (DFT), proving that the electronic density $n(\boldsymbol{r})$, the spin magnetization $\boldsymbol{m}(\boldsymbol{r})$, the diagonal of the nuclear N-body density matrix, and the singlet and triplet SC order parameters $\boldsymbol{\chi}(\boldsymbol{r},\boldsymbol{r}')$ are uniquely connected with their respective external potentials. With this extension of the Hohenberg-Kohn theorem [22] we lay the foundation of the DFT for magnetic and SC systems: SpinSCDFT. In Sec. III A, we introduce the formally noninteracting Kohn Sham (KS) system that reproduces the exact densities of the interacting system. Similar to every DFT, SpinSCDFT relies on the construction of an exchange-correlation (xc) functional that connects the KS with the interacting system. In this work, this is achieved by establishing, in Sec. III B, a Sham-Schlüter connection [23] via the Dyson equation of the interacting system.

The interacting system is also being investigated directly by means of a magnetic extension of the Eliashberg method [24–29]. A derivation[1] of this alternative approach in the present notation is given in Sec. IV. Advantages and disadvantages of these two theoretical schemes, SpinSCDFT and Eliashberg, will be discussed in the conclusions.

II. HAMILTONIAN

We assume that the interacting system is governed by the Pauli Hamiltonian (we use Hartree atomic units throughout)

$$\hat{H} = \hat{T}_{\mathrm{e}} + \hat{T}_{\mathrm{n}} + \hat{V}_{\mathrm{e}} + \hat{U}_{\mathrm{ee}} + \hat{U}_{\mathrm{en}} + \hat{U}_{\mathrm{nn}}\,, \tag{1}$$

where $\hat{T}_{\mathrm{e}}$ ($\hat{T}_{\mathrm{n}}$) is the kinetic energy operator of the electron (nuclei) and $\hat{U}_{\mathrm{ee}}(\hat{U}_{\mathrm{nn}})$ is the electron-electron (nuclei-nuclei) interaction, i.e., usually the Coulomb potential. $\hat{U}_{\mathrm{en}}$ is the Coulomb potential between electrons and nuclei. To break the

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[1]Schossmann and Schachinger have derived Eliashberg equations including the vector potential [61]. However, they set out from a self-energy that is taken to be local in real space with an empirical electron phonon coupling. It is not straightforward to generalize their approach to the case of *ab initio* calculations, where the pairing interactions are usually taken to be local in the space of normal-state quasiparticles. Vonsovsky *et al.* [24] have derived Eliashberg equations, treating the magnetic field perturbatively except for an onsite splitting parameter. They require the self-energy to be diagonal with respect to normal-state electronic orbitals which is similar to the main results in this work.

respective symmetries and allow the corresponding densities to adopt nonzero values in a thermal average, we include an external vector potential $\boldsymbol{A}_{\rm ext}(\boldsymbol{r})$ and an external singlet/triplet pair potential $\boldsymbol{\Delta}^{\rm ext}(\boldsymbol{r},\boldsymbol{r}')$ in the Hamiltonian. These external fields will be set to zero at the end of the derivation. Because we do not consider currents, the only term in the Pauli Hamiltonian containing $\boldsymbol{A}_{\rm ext}(\boldsymbol{r})$ is

$$\hat{T}_{\rm e} = \int d\boldsymbol{r}\, \hat{\psi}^\dagger(\boldsymbol{r}) \cdot \left(-\sigma_0 \frac{\boldsymbol{\nabla}^2}{2} + \mathbf{S} \cdot \boldsymbol{B}_{\rm ext}(\boldsymbol{r}) \right) \cdot \hat{\psi}(\boldsymbol{r}) \qquad (2)$$

with $\boldsymbol{B}_{\rm ext}(\boldsymbol{r}) = \boldsymbol{\nabla} \times \boldsymbol{A}_{\rm ext}(\boldsymbol{r})$ and $\mathbf{S} = \frac{1}{2}(\sigma_x \quad \sigma_y \quad \sigma_z)^{\rm T}$, $\sigma_{x,y,z}$ being the Pauli matrices. We use the notation $\hat{\psi}^\dagger(\boldsymbol{r}) = (\hat{\psi}^\dagger(\boldsymbol{r}\uparrow) \quad \hat{\psi}^\dagger(\boldsymbol{r}\downarrow))$ for the field operator where $\hat{\psi}^\dagger(\boldsymbol{r}\uparrow)$ creates an electron at location $\boldsymbol{r}$ with spin up. The scalar potential part of $\hat{H}$ reads as

$$\hat{V}_{\rm e} = \int d\boldsymbol{r}\, \hat{\psi}^\dagger(\boldsymbol{r}) \cdot \sigma_0 \cdot \hat{\psi}(\boldsymbol{r}) v_{\rm ext}(\boldsymbol{r}) - \frac{1}{2} \int d\boldsymbol{r} \int d\boldsymbol{r}' (\hat{\boldsymbol{\chi}}(\boldsymbol{r},\boldsymbol{r}') \cdot \boldsymbol{\Delta}^{\rm ext*}(\boldsymbol{r},\boldsymbol{r}') + \text{H.c.}). \qquad (3)$$

Here, the anomalous density operator is defined by

$$\hat{\boldsymbol{\chi}}(\boldsymbol{r},\boldsymbol{r}') = \hat{\psi}(\boldsymbol{r}) \cdot \boldsymbol{\Phi} \cdot \hat{\psi}(\boldsymbol{r}')\,. \qquad (4)$$

$\hat{\boldsymbol{\chi}}(\boldsymbol{r},\boldsymbol{r}')$ is a 4-vector of which the first component (proportional to Φ_1) is the singlet part of the order parameter, while the other components (related to Φ_2, Φ_3, and Φ_4) are the triplet part. The four components of the singlet/triplet vector $\boldsymbol{\Phi} = (i\sigma_y, -\sigma_z, \sigma_0, \sigma_x)^{\rm T}$ are 2×2 spin matrices similar to the components of $\mathbf{S}$. Similarly, the anomalous external potential

$$\boldsymbol{\Delta}^{\rm ext}(\boldsymbol{r},\boldsymbol{r}') = \begin{pmatrix} \Delta_{\rm s}^{\rm ext}(\boldsymbol{r},\boldsymbol{r}') \\ \Delta_{\rm tx}^{\rm ext}(\boldsymbol{r},\boldsymbol{r}') \\ \Delta_{\rm ty}^{\rm ext}(\boldsymbol{r},\boldsymbol{r}') \\ \Delta_{\rm tz}^{\rm ext}(\boldsymbol{r},\boldsymbol{r}') \end{pmatrix} \qquad (5)$$

is assumed to have singlet and triplet components.

III. SPIN SCDFT

The conventional density functional approach to the many-body problem [22,30–32] consists of two steps: *first* establishing the Hohenberg-Kohn (HK) theorem, i.e., realize that a chosen set of densities is uniquely connected with a set of external potentials; *second*, construct an auxiliary, noninteracting KS system to reproduce the densities of the interacting system.

We follow Ref. [33] and consider a multicomponent DFT with the normal $n(\boldsymbol{r})$, the SC order parameter as the anomalous density $\boldsymbol{\chi}(\boldsymbol{r},\boldsymbol{r}')$, that describes the electrons condensed into singlet and triplet states, and $\Gamma(\boldsymbol{R}_1 \ldots \boldsymbol{R}_N)$ the diagonal of the nuclear N-body density matrix. In addition, we introduce the magnetization $\boldsymbol{m}(\boldsymbol{r})$ as another electronic density.

The HK proof $[n(\boldsymbol{r}),\boldsymbol{m}(\boldsymbol{r}),\boldsymbol{\chi}(\boldsymbol{r},\boldsymbol{r}'),\Gamma(\boldsymbol{R}_1 \ldots \boldsymbol{R}_N)] \leftrightarrow [v_{\rm ext}(\boldsymbol{r}),\boldsymbol{B}_{\rm ext}(\boldsymbol{r}),\boldsymbol{\Delta}^{\rm ext}(\boldsymbol{r},\boldsymbol{r}'),W_{\rm ext}(\boldsymbol{R}_1 \ldots \boldsymbol{R}_N)]$ is a straightforward generalization of Mermin's HK proof in a finite-temperature ensemble [34]. For this reason, we will not repeat it here. On the other hand, the construction of the KS system is done assuming that densities are always v representable, i.e., we assume the existence of the KS system. Being noninteracting it consists of independent equations for nuclei and electrons, coupled only via the xc potentials. Our focus will be on the electronic system, discussed in detail in Sec. III A 2. The nuclear part will be addressed in Sec. III A 1, briefly, since it is usually enough to approximate the nuclear KS system with its non-SC counterpart [33,35]. The construction of the xc potentials will be discussed in Secs. III B and III C.

A. Kohn-Sham system

In this work, we are mainly interested in the influence of a magnetic field on the SC state. We briefly review the approximation steps to arrive at the Fröhlich Hamiltonian starting from the formally exact multicomponent DFT. The reader may refer to the existing literature for further details [32,33]. We introduce the KS Hamiltonian

$$\hat{H}_{\rm KS} = \hat{H}_{\rm KS}^{\rm e} + \hat{H}_{\rm KS}^{\rm n}\,, \qquad (6)$$

where we have separated the electronic $\hat{H}_{\rm KS}^{\rm e}$,

$$\hat{H}_{\rm KS}^{\rm e} = \int d\boldsymbol{r}\, \hat{\psi}^\dagger(\boldsymbol{r}) \cdot \sigma_0 \left(-\frac{\boldsymbol{\nabla}^2}{2} + v_{\rm s}(\boldsymbol{r}) - \mu \right) \cdot \hat{\psi}(\boldsymbol{r}) - \frac{1}{2} \int d\boldsymbol{r} \int d\boldsymbol{r}' (\hat{\boldsymbol{\chi}}(\boldsymbol{r},\boldsymbol{r}') \cdot \boldsymbol{\Delta}^{\rm s*}(\boldsymbol{r},\boldsymbol{r}') + \text{H.c.}) + \int d\boldsymbol{r}\, \hat{\boldsymbol{m}}(\boldsymbol{r}) \cdot \boldsymbol{B}_{\rm s}(\boldsymbol{r}), \qquad (7)$$

from the nuclear $\hat{H}_{\rm KS}^{\rm n}$,

$$\hat{H}_{\rm KS}^{\rm n} = -\int d\boldsymbol{R}\, \hat{\zeta}^\dagger(\boldsymbol{R}) \frac{\boldsymbol{\nabla}_{\boldsymbol{R}}^2}{2M} \hat{\zeta}(\boldsymbol{R}) + \int \cdots \int d\boldsymbol{R}_1 \cdots d\boldsymbol{R}_{N_n} \hat{\zeta}^\dagger(\boldsymbol{R}_1) \cdots \hat{\zeta}^\dagger(\boldsymbol{R}_{N_n}) \times W_{\rm s}(\boldsymbol{R}_1, \ldots, \boldsymbol{R}_{N_n}) \hat{\zeta}(\boldsymbol{R}_1) \cdots \hat{\zeta}(\boldsymbol{R}_{N_n}). \qquad (8)$$

We write $v_{\rm s}(\boldsymbol{r}) = v_{\rm ext}(\boldsymbol{r}) + v_{\rm xc}(\boldsymbol{r})$ with $v_{\rm xc}(\boldsymbol{r})$ being the scalar xc potential [similar for $\boldsymbol{B}_{\rm s}(\boldsymbol{r})$ and $\boldsymbol{\Delta}^{\rm s}(\boldsymbol{r},\boldsymbol{r}')$]. $\hat{\boldsymbol{m}}(\boldsymbol{r}) = \hat{\psi}^\dagger(\boldsymbol{r}) \cdot \mathbf{S} \cdot \hat{\psi}(\boldsymbol{r})$ is the operator of the magnetic density. In the nuclear description, $\hat{\zeta}^\dagger(\boldsymbol{R})$ creates the nuclear field at location $\boldsymbol{R}$. Following Lüders *et al.* [33] and Marques *et al.* [35], we use the N-body potential $W_{\rm s}(\boldsymbol{R}_1, \ldots, \boldsymbol{R}_{N_n})$ because in this way the nuclear KS system can be easily related to the standard Born-Oppenheimer approximation. M refers to the ionic mass. Here, we neglect the spin of the nuclei and consider only one atomic type (the generalization is straightforward).

1. Nuclear part

Since SC occurs in the solid phase, we assume that ions can only perform small oscillations about their equilibrium position. A discussion that goes beyond this simple picture can be found in Refs. [32,36]. We expand $W_{\rm s}(\boldsymbol{R}_1, \ldots, \boldsymbol{R}_{N_n})$ in $\boldsymbol{u}_i = \boldsymbol{R}_i - \boldsymbol{R}_{0i}$ around the equilibrium positions $\boldsymbol{R}_{0i}$. The nuclear degrees of freedom (up to harmonic order) are described by the Hamiltonian $\hat{H}_{\rm KS}^{\rm ph}$ with $\hat{H}_{\rm KS}^{\rm n} = \hat{H}_{\rm KS}^{\rm ph} + \mathcal{O}(\boldsymbol{u}^3)$ in second quantization

$$\hat{H}_{\rm KS}^{\rm ph} = \sum_q \Omega_q \left(\hat{b}_q^\dagger \hat{b}_q + \frac{1}{2} \right). \qquad (9)$$

We use the notation $q = \boldsymbol{q},\lambda$ with Bloch vector $\boldsymbol{q}$ and mode number λ. We further use the notation $-q = -\boldsymbol{q},\lambda$ for all Bloch vector and band or mode combinations. We point out that via the functional dependence of $W_s[n,\boldsymbol{m},\boldsymbol{\chi},\Gamma]$, the KS phonon frequencies Ω_q are in principle functionals of the densities as well. $\hat{b}_q^\dagger$ creates a bosonic KS phonon with quantum numbers q. Usually, approximating W_s with the Born-Oppenheimer energy surface leads to phonon frequencies in excellent agreement with experiment [37,38].

The electron phonon scattering should be formally constructed from the bare Coulomb interaction [36]. However, in order to have a proper description of the electronic screening, this is not feasible in practice. The solution is the substitution of the many-body electron-phonon interaction with its KS counterpart $\hat{U}_{\text{en}} \to \hat{H}_{\text{KS}}^{\text{e-ph}}$:

$$\hat{H}_{\text{KS}}^{\text{e-ph}} = \sum_{q\,m} \int d\boldsymbol{r}\, g_q^a(\boldsymbol{r}) \hat{\psi}^\dagger(\boldsymbol{r}) \cdot \sigma_m \cdot \hat{\psi}(\boldsymbol{r})(\hat{b}_q + \hat{b}_{-q}^\dagger), \quad (10)$$

where $m = 0,z$ and $g_q^0(\boldsymbol{r}) = \frac{\delta v_s(\boldsymbol{r})}{\delta \boldsymbol{u}_q}$, $g_q^z(\boldsymbol{r}) = \frac{\delta \boldsymbol{B}_{sz}(\boldsymbol{r})}{\delta \boldsymbol{u}_q}$, $\boldsymbol{u}$ being the phononic displacement vectors [37,38]. This form incorporates most of the electronic influence on the bare Coulomb interaction between electrons and nuclei. We consider this as a good approximation for the dressed phonon vertex in the non-SC state (see also Ref. [36] for a further discussion). Note that $\langle \hat{H}_{\text{KS}}^{\text{e-ph}} \rangle$ is part of the xc functional of the electronic KS system and will be added later in our approximate functional using perturbation theory. For later use in the derivation of the xc potential, we define the propagator of the noninteracting system of KS phonons

$$D_{q,q'}^0(\tau) = \langle \mathrm{T}[\hat{b}_q(\tau) + \hat{b}_{-q}^\dagger(\tau)][\hat{b}_{q'}(0) + \hat{b}_{-q'}^\dagger(0)] \rangle_{\text{ph}}, \quad (11)$$

$$D_{q,q'}^0(\nu_n) = \delta_{q,-q'} \left(\frac{1}{i\nu_n + \Omega_q} - \frac{1}{i\nu_n - \Omega_q} \right). \quad (12)$$

Here, T is the usual time (τ) ordering symbol of operators $\hat{b}_q(\tau) + \hat{b}_{-q}^\dagger(\tau)$ in the Heisenberg picture and $\langle \cdots \rangle_{\text{ph}}$ means to evaluate the thermal average using the Hamiltonian $\hat{H}_{\text{KS}}^{\text{ph}}$ of Eq. (9). The bosonic Matsubara frequency is $\nu_n = \frac{2\pi n}{\beta}$.

2. *Electronic part*

The electronic KS Hamiltonian $\hat{H}_{\text{KS}}^{\text{e}}$ is not diagonal in the electronic field operator $\hat{\psi}(\boldsymbol{r})$ because Eq. (7) involves terms proportional to $\psi\psi$ and $\psi^\dagger\psi^\dagger$. Being a Hermitian operator, we can find an orthonormal set of eigenvectors of $\hat{H}_{\text{KS}}^{\text{e}}$ in which it is diagonal. Let $\hat{\gamma}_k^\dagger$ create such a two-component vector in spin space (the Hamiltonian is not diagonal in spin so the spin degrees of freedom is in the set $\{k\}$), then the SC KS system will take the form

$$\hat{H}_{\text{KS}}^{\text{e}} = E_0 + \sum_k E_k \hat{\gamma}_k^\dagger \hat{\gamma}_k, \quad E_k \geqslant 0 \quad (13)$$

where E_0 is the ground-state energy and the E_k are all positive. This form can be achieved [39] by commuting the operators $\hat{H}_{\text{KS}}^{\text{e}} = \sum_k \tilde{E}_k \hat{a}_k^\dagger \hat{a}_k = \sum_{k|\tilde{E}_k<0} + \sum_{k|\tilde{E}_k\geqslant 0} \tilde{E}_k \hat{a}_k^\dagger \hat{a}_k + \sum_{k|\tilde{E}_k<0} |\tilde{E}_k| \hat{a}_k \hat{a}_k^\dagger$ and then redefining the negative-energy particle operators as holes $\hat{a}_k = \hat{\gamma}_k^\dagger$. We use a notation based on the original Refs. [24,40,41] and introduce

$$\hat{\Psi}(\boldsymbol{r}) = \begin{pmatrix} \hat{\psi}(\boldsymbol{r}\uparrow) \\ \hat{\psi}(\boldsymbol{r}\downarrow) \\ \hat{\psi}^\dagger(\boldsymbol{r}\uparrow) \\ \hat{\psi}^\dagger(\boldsymbol{r}\downarrow) \end{pmatrix}. \quad (14)$$

Using this Nambu field operator $\hat{\Psi}(\boldsymbol{r})$, the KS Hamiltonian reads as

$$\hat{H}_{\text{KS}}^{\text{e}} = \int d\boldsymbol{r} \int d\boldsymbol{r}' \hat{\Psi}^\dagger(\boldsymbol{r}) \cdot \frac{1}{2} \bar{H}_{\text{KS}}(\boldsymbol{r},\boldsymbol{r}') \cdot \hat{\Psi}(\boldsymbol{r}'), \quad (15)$$

where the KS Hamiltonian (first quantization Nambu form) is given by

$$\bar{H}_{\text{KS}}(\boldsymbol{r},\boldsymbol{r}') = \begin{pmatrix} \delta(\boldsymbol{r}-\boldsymbol{r}') H_{\text{KS}}^{\text{NS}}(\boldsymbol{r}) & \boldsymbol{\Phi} \cdot \boldsymbol{\Delta}^{\text{s}}(\boldsymbol{r},\boldsymbol{r}') \\ -[\boldsymbol{\Phi} \cdot \boldsymbol{\Delta}^{\text{s}}(\boldsymbol{r},\boldsymbol{r}')]^* & -\delta(\boldsymbol{r}-\boldsymbol{r}') \big[H_{\text{KS}}^{\text{NS}}(\boldsymbol{r})\big]^{\text{T}_s} \end{pmatrix}, \quad (16)$$

with

$$H_{\text{KS}}^{\text{NS}}(\boldsymbol{r}) = \big(-\tfrac{1}{2}\nabla^2 + v_s(\boldsymbol{r}) - \mu\big)\sigma_0 - \mathbf{S} \cdot \mathbf{B}_s(\boldsymbol{r}). \quad (17)$$

Note that the changed order of the electronic field operator implies a transposition in spin space in the $(-1,-1)$ component that is equivalent to using $\mathbf{S}^*$. In a similar transformation, the diagonal KS Hamiltonian (13) becomes

$$\hat{H}_{\text{KS}}^{\text{e}} = \sum_k \hat{\Phi}_k^\dagger \cdot \frac{1}{2} \begin{pmatrix} E_k & 0 \\ 0 & -E_k \end{pmatrix} \cdot \hat{\Phi}_k \quad (18)$$

with $(\gamma\gamma)^T = \binom{\hat{\gamma}_k}{\hat{\gamma}_k^\dagger}$. As a consequence of the rearrangement of the operators to the Nambu-Anderson form of Eq. (18), there should appear the trace of the Hamiltonian $\hat{H}_{\text{KS}}^{\text{e}}$. However, not being an operator, this trace cancels from thermal averages and has been disregarded. $\hat{\Phi}_k$ is a two- (not four-) component vector because the spin may not be a good quantum number in the SC KS system. We can diagonalize $\hat{H}_{\text{KS}}^{\text{e}}$ in the form of Eq. (15) resulting in Eq. (18) by introducing a unitary transformation that we parametrize generically with four complex spinor functions. This connection between $\hat{\Psi}(\boldsymbol{r})$ and $\hat{\Phi}_k$ is known as the Bogoliubov-Valatin transformation [42,43]. We write it in the form

$$\hat{\Psi}(\boldsymbol{r}) = \sum_k \begin{pmatrix} \vec{u}_k(\boldsymbol{r}) & \vec{v}_k^*(\boldsymbol{r}) \\ \vec{v}_k(\boldsymbol{r}) & \vec{u}_k^*(\boldsymbol{r}) \end{pmatrix} \cdot \hat{\Phi}_k,$$

$$\hat{\Phi}_k = \int d\boldsymbol{r} \begin{pmatrix} \vec{u}_k^*(\boldsymbol{r}) & \vec{v}_k^*(\boldsymbol{r}) \\ \vec{v}_k(\boldsymbol{r}) & \vec{u}_k(\boldsymbol{r}) \end{pmatrix} \cdot \hat{\Psi}(\boldsymbol{r}). \quad (19)$$

Note that in the first case the matrix is 4×2 dimensional, and in the second 2×4 because of the spinor property of the $\vec{u}_k(\boldsymbol{r}), \vec{v}_k(\boldsymbol{r})$. In going from Eq. (15) to (18), we identify

$$\int d\boldsymbol{r} \int d\boldsymbol{r}' \begin{pmatrix} \vec{u}_k^*(\boldsymbol{r}) & \vec{v}_k^*(\boldsymbol{r}) \\ \vec{v}_k(\boldsymbol{r}) & \vec{u}_k(\boldsymbol{r}) \end{pmatrix} \cdot \bar{H}_{\text{KS}}(\boldsymbol{r},\boldsymbol{r}') \cdot \begin{pmatrix} \vec{u}_{k'}(\boldsymbol{r}') & \vec{v}_{k'}^*(\boldsymbol{r}') \\ \vec{v}_{k'}(\boldsymbol{r}') & \vec{u}_{k'}^*(\boldsymbol{r}') \end{pmatrix}$$

$$= \begin{pmatrix} E_k & 0 \\ 0 & -E_k \end{pmatrix} \delta_{kk'}, \quad (20)$$

which are the KS Bogoliubov–de Gennes (KSBdG) equations for magnetic system. Applying the inverse Bogoliubov-Valatin transformation from the left we obtain two redundant vector

equations of which we usually consider the first for the positive eigenvalues E_k:

$$\int d\boldsymbol{r}'\,\bar{H}_{\mathrm{KS}}(\boldsymbol{r},\boldsymbol{r}')\cdot\begin{pmatrix}\vec{u}_k(\boldsymbol{r}')\\ \vec{v}_k(\boldsymbol{r}')\end{pmatrix}=E_k\begin{pmatrix}\vec{u}_k(\boldsymbol{r})\\ \vec{v}_k(\boldsymbol{r})\end{pmatrix}. \tag{21}$$

This is the usual form of the KSBdG equations which generalize those of Refs. [33,44]. The equation in $(\vec{v}_k^*(\boldsymbol{r})\quad \vec{u}_k^*(\boldsymbol{r}))^{\mathrm{T}}$ leads to the equivalent negative eigenvalue $-E_k$ which reflects the additional degrees of freedom that we have created in going to the 2×2 Nambu formalism.

a. Normal-state KS basis expansion. The KSBdG equations (21) pose a challenging integrodifferential problem. Sensible approximations can be obtained by first performing an expansion into a basis set that is accessible in practice and resembles closely to the true quasiparticle structure of the nonsuperconducting phase of the material under consideration. With this in mind, we consider the non-SC KS single-particle equation

$$\varepsilon_{i\sigma}\vec{\varphi}_{i\sigma}(\boldsymbol{r})=\left[\left(-\frac{\boldsymbol{\nabla}^2}{2}+v_{\mathrm{s}}^0(\boldsymbol{r})-\mu\right)\sigma_0-\boldsymbol{B}_{\mathrm{s}z}^0(\boldsymbol{r})\frac{\sigma_z}{2}\right]\cdot\vec{\varphi}_{i\sigma}(\boldsymbol{r}). \tag{22}$$

$v_{\mathrm{s}}^0(\boldsymbol{r})$ and $\boldsymbol{B}_{\mathrm{s}z}^0(\boldsymbol{r})$ are known functionals, like the local spin density approximation (LSDA) [45]. We also assume that $\boldsymbol{B}_{\mathrm{s}}^0$ is collinear and has components in σ_z only. We use a pure spinor notation for the orbitals, i.e., $\vec{\varphi}_{i\sigma}(\boldsymbol{r})$ has only one nonvanishing component, e.g., $\vec{\varphi}_{i\uparrow}(\boldsymbol{r})=\binom{\varphi_i(\boldsymbol{r}\uparrow)}{0}$. We use the indices i,j for the quantum numbers of the basis and thus distinguish from the quantum number k of the SC KS system. Later, in the spin decoupling approximation (Sec. III A 2 c) when we assume the expansion coefficients to have only one nonvanishing component each, this distinction will not be made. As a next step, we expand the Bogoliubov-Valatin transformations in these solutions[2] $\{\vec{\varphi}_{i\sigma}(\boldsymbol{r})\}$:

$$\vec{u}_k(\boldsymbol{r})=\sum_{i\sigma}u_k^{i\sigma}\vec{\varphi}_{i\sigma}(\boldsymbol{r}),\quad \vec{v}_k(\boldsymbol{r})=\sum_{i\sigma}v_k^{i\sigma}\vec{\varphi}_{i\sigma}^*(\boldsymbol{r}). \tag{23}$$

Defining the matrix elements

$$\mathcal{R}_{ij}^{\sigma\sigma'}=\int d\boldsymbol{r}\,\vec{\varphi}_{i\sigma}^*\big(\sigma_0\big[v_{\mathrm{s}}(\boldsymbol{r})-v_{\mathrm{s}}^0(\boldsymbol{r})\big]-\mathbf{S}\cdot\big[\boldsymbol{B}_{\mathrm{s}}(\boldsymbol{r})-\boldsymbol{B}_{\mathrm{s}}^0(\boldsymbol{r})\big]\big)\vec{\varphi}_{j\sigma'}, \tag{24}$$

$$\Delta_{ij}^{\mathrm{s}\sigma\sigma'}=\int d\boldsymbol{r}\int d\boldsymbol{r}'\vec{\varphi}_{i\sigma}^*(\boldsymbol{r})\cdot\big[\boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}}(\boldsymbol{r},\boldsymbol{r}')\big]\cdot\vec{\varphi}_{j\sigma'}^*(\boldsymbol{r}'), \tag{25}$$

$$\mathcal{E}_{ij}^{\sigma\sigma'}=\varepsilon_{i\sigma}\delta_{ij}\delta_{\sigma\sigma'}+\mathcal{R}_{ij}^{\sigma\sigma'}, \tag{26}$$

and the singlet/triplet parts of the pair-potential expansion coefficient matrix

$$\Delta_{\mathrm{s}ij}^{\mathrm{s}}=\tfrac{1}{2}\big(\Delta_{ij}^{\mathrm{s}\uparrow\downarrow}-\Delta_{ij}^{\mathrm{s}\downarrow\uparrow}\big),\quad \Delta_{\mathrm{tx}ij}^{\mathrm{s}}=\tfrac{1}{2}\big(\Delta_{ij}^{\mathrm{s}\downarrow\downarrow}-\Delta_{ij}^{\mathrm{s}\uparrow\uparrow}\big),$$
$$\Delta_{\mathrm{ty}ij}^{\mathrm{s}}=\tfrac{1}{2}\big(\Delta_{ij}^{\mathrm{s}\downarrow\downarrow}+\Delta_{ij}^{\mathrm{s}\uparrow\uparrow}\big),\quad \Delta_{\mathrm{tz}ij}^{\mathrm{s}}=\tfrac{1}{2}\big(\Delta_{ij}^{\mathrm{s}\uparrow\downarrow}+\Delta_{ij}^{\mathrm{s}\downarrow\uparrow}\big), \tag{27}$$

we can cast Eq. (20) into a convenient form

$$(g_k^+\quad g_k^-)^\dagger\cdot\begin{pmatrix}\mathcal{E} & \boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}}\\ (\boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}})^\dagger & -\mathcal{E}^{\mathrm{T}}\end{pmatrix}\cdot(g_{k'}^+\quad g_{k'}^-)=E_k\delta_{kk'}\tau_z\,, \tag{28}$$

with

$$g_k^+=\big(u_k^{1\uparrow}\;\;u_k^{1\downarrow}\;\;u_k^{2\uparrow}\;\;\cdots\;\;v_k^{1\uparrow}\;\;v_k^{1\downarrow}\;\;v_k^{2\uparrow}\;\;\ldots\big)^{\mathrm{T}}, \tag{29}$$

$$g_k^-=\big(v_k^{1\uparrow *}\;\;v_k^{1\downarrow *}\;\;v_k^{2\uparrow *}\;\;\cdots\;\;u_k^{1\uparrow *}\;\;u_k^{1\downarrow *}\;\;u_k^{2\uparrow *}\;\;\ldots\big)^{\mathrm{T}}. \tag{30}$$

The superscript $1,2,\ldots$ means we have ordered the Bloch vectors and bands in some way. The precise way of ordering is unimportant. The $2N$-dimensional vectors $\{g_k^\pm\}$ have an N-dimensional part that refers to the particle $(\ldots,1)$ components (the u's for g_k^+) and a second part that refers to the hole $(\ldots,-1)$ components (the v's for g_k^+) when multiplied from the right to a Nambu matrix. Note that the set of $\{g_k^-\}$ solves the eigenvalue equation similar to Eq. (21) with the negative eigenvalues $-E_k$, while the set $\{g_k^+\}$ corresponds to the eigenvectors with positive eigenvalues E_k. The elements of the set $\{g_k^+\}$ are the SC KS orbitals of SpinSCDFT, represented in the normal-state KS orbital basis. We may easily represent the densities using the normal-state KS orbital basis $\{\vec{\varphi}_{i\sigma}(\boldsymbol{r})\}$, for example,

$$n(\boldsymbol{r})=\sum_{i\sigma j\sigma'}\vec{\varphi}_i^*(\boldsymbol{r}\sigma)(n_{ij})_{\sigma\sigma'}\vec{\varphi}_j(\boldsymbol{r}\sigma'), \tag{31}$$

and similar for $\boldsymbol{m}(\boldsymbol{r})$ and $\boldsymbol{\chi}(\boldsymbol{r},\boldsymbol{r}')$ where $\boldsymbol{\chi}(\boldsymbol{r},\boldsymbol{r}')$ is expanded in $\vec{\varphi}_i^*(\boldsymbol{r}\sigma)$ and $\vec{\varphi}_j^*(\boldsymbol{r}'\sigma')$. The coefficients read as

$$(n_{ij})_{\sigma\sigma'}=(\sigma_0)_{\sigma\sigma'}\sum_k\big[\big(u_k^{i\sigma}\big)^*u_k^{j\sigma'}f_\beta(E_k)+v_k^{i\sigma}\big(v_k^{j\sigma'}\big)^*f_\beta(-E_k)\big], \tag{32}$$

$$(\boldsymbol{m}_{ij})_{\sigma\sigma'}=(\mathbf{S})_{\sigma\sigma'}\sum_k\big[\big(u_k^{i\sigma}\big)^*u_k^{j\sigma'}f_\beta(E_k)+v_k^{i\sigma}\big(v_k^{j\sigma'}\big)^*f_\beta(-E_k)\big], \tag{33}$$

$$(\boldsymbol{\chi}_{ij})_{\sigma\sigma'}=(\boldsymbol{\Phi})_{\sigma\sigma'}\sum_k\big[u_k^{j\sigma'}\big(v_k^{i\sigma}\big)^*f_\beta(E_k)+u_k^{i\sigma}\big(v_k^{j\sigma'}\big)^*f_\beta(-E_k)\big]. \tag{34}$$

We want to stress that we have not performed any approximations so far and the SC KS system reproduces the exact interacting densities of the Hamiltonian of Eq. (1).

b. Singlet superconductivity. Due to the antisymmetric structure of the fermionic wave function and the effectively attractive interaction, in absence of magnetism, the singlet solution usually leads to a more stable SC state. Known SC that feature a triplet pairing all share a very low critical temperature less than a few Kelvin [9–15]. In presence of magnetism, as we have seen, the spin is not a good quantum number and singlet/triplet components mix. Since the triplet pairing channel seems to be rather unimportant for many systems, it is of use to define a singlet approximation, in which it is completely disregarded.

We therefore make the assumption that our pairing potential has only the singlet component (marked as a subscript S in the KS potential). In addition, we assume a collinear spin structure

[2]Note that the $-1,-1$ component of the SC KS Hamiltonian Eq. (16) is the complex conjugated of the 1,1. This comes from the property $[H_{\mathrm{KS}}^{\mathrm{NS}}(\boldsymbol{r})]^{\mathrm{T}_{\mathrm{s}}}=[H_{\mathrm{KS}}^{\mathrm{NS}}(\boldsymbol{r})]^*$ of the Hamiltonian, T_{s} being a transposition in spin space.

in the normal-state part of the Hamiltonian

$$\mathbf{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}} \approx \Phi_1 \Delta^{\mathrm{s}}_{\mathrm{s}}, \quad \mathcal{E}^{\sigma\sigma'}_{ij} \approx \mathcal{E}^{\sigma}_{ij}\delta_{\sigma\sigma'}. \tag{35}$$

We observe that spin becomes a good quantum number in the SC KS system. This follows because the KS Hamiltonian matrix elements can be brought to a block-diagonal structure in Nambu and spin space with two kinds of eigenfunctions to each individual block. Consequently, we relabel the eigenvectors with $k \to k,\mu$ where the size of the set of k is reduced to half. Each block μ is diagonalized as

$$\begin{aligned}(g^{+}_{k\mu} \quad g^{-}_{k,-\mu})^{\dagger} \begin{pmatrix} \mathcal{E}^{\mu} & \mathrm{sign}(\mu)\Delta^{\mathrm{s}}_{\mathrm{s}} \\ \mathrm{sign}(\mu)\Delta^{\mathrm{s}\dagger}_{\mathrm{s}} & -\mathcal{E}^{-\mu\mathrm{T}} \end{pmatrix} (g^{+}_{k'\mu} \quad g^{-}_{k',-\mu}) \\ = \delta_{kk'} \begin{pmatrix} E^{+}_{k\mu} & 0 \\ 0 & E^{-}_{k\mu} \end{pmatrix}\end{aligned} \tag{36}$$

with

$$g^{+}_{k\mu} = \begin{pmatrix} u^{1\mu}_{k\mu} & u^{2\mu}_{k\mu} & \cdots & v^{1-\mu}_{k\mu} & \cdots \end{pmatrix}^{\mathrm{T}}, \tag{37}$$

$$g^{-}_{k\mu} = \begin{pmatrix} v^{1,-\mu *}_{k\mu} & v^{2,-\mu *}_{k\mu} & \cdots & u^{1\mu *}_{k\mu} & \cdots \end{pmatrix}^{\mathrm{T}}. \tag{38}$$

$E^{+}_{k\mu}$ is an eigenvalue that may or may not be positive. However, we have introduced the SC KS particles in Eq. (13) with a positive excitation energy $E_{k\mu}$ so this fact requires further commenting. In the present situation where the matrix elements of the SC KS Hamiltonian are block diagonal in Nambu and spin space, we can show that if $g^{+}_{k\mu}$ has the eigenvalue $E^{+}_{k\mu}$ the "negative" labeled eigenfunction $g^{-}_{k,-\mu}$ has the eigenvalue $-E^{+}_{k,-\mu}$.[3] Thus, we still have the original redundancy in the eigenvalue spectrum but not in the same spin channel μ. Instead,

$$E^{\pm}_{k\mu} = -E^{\mp}_{k,-\mu}. \tag{39}$$

We conclude that to every k we have four eigenvalues of which two are positive. These positive eigenvalues are identified with $E_{k\mu}$. In Sec. III A 2 c after introducing the decoupling approximation we will be able to compute these eigenvalues explicitly, and continue this discussion. Note that the correspondence of the spin μ with the spin of the underlying electrons σ is not one to one. The pair potential is spin singlet and thus couples the σ channel of a particle with the $-\sigma$ channel of the hole state. As we shall see later using the decoupling approximation in the non-SC limit, a given eigenvalue $E^{-}_{k\mu}$, for example, changes its spin character in terms of the non-SC eigenvalues $\varepsilon_{k\sigma}$ from μ to $-\mu$ upon crossing the Fermi level. In general, the pairing will mix different spin channels. On the other hand, for states far away from the Fermi level where the pairing is negligible, one finds that the mixing of opposite spin hole states is negligible and the spin μ of the SC KS states equals the NS KS state spin.

c. Spin decoupling approximation. It is desirable to reduce the effort to solve the KSBdG equation (36) further. A substantial simplification is the decoupling approximation [33,35] (or Anderson approximation [46]). There, one considers only singlet SC and pairing between a quasiparticle state $(i\sigma)$ and its time-reversed hole state $(-i,-\sigma)$. Furthermore, it is assumed that the basis $\{\vec{\varphi}_{i\sigma}\}$ approximates the true non-SC quasiparticle structure well enough. In the language of the KSBdG equation (28), this reads as

$$\mathcal{E}^{\sigma\sigma'}_{ij} \approx \varepsilon_{i\sigma}\delta_{\sigma\sigma'}\delta_{ij}, \quad (\mathbf{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}})^{\sigma\sigma'}_{ij} \approx \Phi^{\sigma\sigma'}_{1}\Delta^{\mathrm{s}}_{\mathrm{s}i,-i}\delta_{i,-j}. \tag{40}$$

This type of approximation is inherent in the Eliashberg equations as well as SCDFT functionals. It is also straightforward to include a diagonal correction $\mathcal{R}^{\sigma\sigma}_{ii}$. In the form presented here, we will call it spin decoupling approximation (SDA). For each k and μ, Eq. (28) reduces to the 2×2 equation

$$\begin{aligned}\begin{pmatrix} u^{k}_{k\sigma} & v^{k*}_{-k\sigma} \\ v^{-k}_{k-\sigma} & u^{-k*}_{-k-\sigma} \end{pmatrix}^{\dagger} \begin{pmatrix} \varepsilon_{k\sigma} & \mathrm{sign}(\sigma)\Delta^{\mathrm{s}}_{\mathrm{s}k,-k} \\ \mathrm{sign}(\sigma)\Delta^{\mathrm{s}*}_{\mathrm{s}-k,k} & -\varepsilon_{-k-\sigma} \end{pmatrix} \\ \times \begin{pmatrix} u^{k}_{k\sigma} & v^{k*}_{-k\sigma} \\ v^{-k}_{k-\sigma} & u^{-k*}_{-k-\sigma} \end{pmatrix} = \begin{pmatrix} E^{+}_{k\sigma} & 0 \\ 0 & E^{-}_{k\sigma} \end{pmatrix}.\end{aligned} \tag{41}$$

Here, we have introduced a single spin notation $v^{-k-\sigma}_{k\sigma} = v^{-k}_{k-\sigma}$ and $u^{k\sigma}_{k\sigma} = u^{k}_{k\sigma}$. The spin label on the coefficients of the Bogoliubov transformation always refers to the normal-state KS basis spin label and thus we use the spin notation $\mu \to \sigma$. As mentioned before, the spin label cannot be strictly identified with the spin of a SC KS particle. We will come back to this point later. From now on, we will use the notation $\Delta^{\mathrm{s}}_{\mathrm{s}k} = \Delta^{\mathrm{s}}_{\mathrm{s}k,-k} = \Delta^{\mathrm{s}}_{\mathrm{s}-k,k}$. We may compute the two eigenvalues and eigenvectors analytically. From the high-energy limit $\varepsilon_{k\sigma} + \varepsilon_{-k,-\sigma} \gg \varepsilon_{k\sigma} - \varepsilon_{-k,-\sigma}$, we identify the name $\pm$ for the two branches. The eigenvalues are

$$E^{-}_{k\sigma} = \frac{\varepsilon_{k\sigma} - \varepsilon_{-k-\sigma}}{2} - \sqrt{\left(\frac{\varepsilon_{k\sigma} + \varepsilon_{-k-\sigma}}{2}\right)^2 + \left|\Delta^{\mathrm{s}}_{\mathrm{s}k}\right|^2}, \tag{42}$$

$$E^{+}_{k\sigma} = \frac{\varepsilon_{k\sigma} - \varepsilon_{-k-\sigma}}{2} + \sqrt{\left(\frac{\varepsilon_{k\sigma} + \varepsilon_{-k-\sigma}}{2}\right)^2 + \left|\Delta^{\mathrm{s}}_{\mathrm{s}k}\right|^2}. \tag{43}$$

In the spin degenerate limit, the $+$ branch has always positive eigenvalues $E^{+}_{k\sigma}$ and it is clear which of the eigenvectors belong to the first column of the Bogoliubov-Valatin transformation. In the spin-polarized case, the situation is more complicated. Again, because $E^{\pm}_{k\sigma} = -E^{\mp}_{-k,-\sigma}$, two of the four Bogoliubov eigenvalues to a given k are positive, but without knowledge of $\varepsilon_{k\sigma}$ and $\Delta^{\mathrm{s}}_{\mathrm{s}k}$ one can not tell in advance which ones these are. The general situation is sketched in Fig. 1 for a constant $\Delta^{\mathrm{s}}_{\mathrm{s}k}$ and homogeneously splitting free-electron gas. In the next paragraph, we give a more detailed discussion of the Bogoliubov eigenvalues $E^{\pm}_{k\sigma}$.

d. Eigenvalues in the SDA. Our first concern is how to interpret the spin quantum number σ of $E^{\pm}_{k\sigma}$ in connection with the underlying normal states $\varepsilon_{k\sigma}$. First, consider the non-SC limit where

$$\Delta^{\mathrm{s}}_{\mathrm{s}k} = 0: \; 2E^{\pm}_{k\sigma} = \varepsilon_{k\sigma} - \varepsilon_{-k-\sigma} \pm |\varepsilon_{k\sigma} + \varepsilon_{-k-\sigma}|. \tag{44}$$

This situation is plotted in Fig. 1(b). Note that if $\varepsilon_{k\sigma} + \varepsilon_{-k-\sigma} > 0$, then $E^{-}_{k\sigma} = -\varepsilon_{-k-\sigma}$ and if $\varepsilon_{k\sigma} + \varepsilon_{-k-\sigma} < 0$ we conclude $E^{-}_{k\sigma} = \varepsilon_{k\sigma}$.

Second, consider the following case that occurs at any k_0 where $\varepsilon_{k_0\uparrow} + \varepsilon_{-k_0\downarrow} = 0$. Given that we have an energy splitting $\varepsilon_{k_0\uparrow} - \varepsilon_{-k_0\downarrow} > 2|\Delta^{\mathrm{s}}_{\mathrm{s}k}|$, we find that both $E^{\pm}_{-k_0,\downarrow}$ are negative.

[3]The explicit calculation uses the fact that $\mathcal{E}$ is Hermitian and thus $\mathcal{E}^{*} = \mathcal{E}^{\mathrm{T}}$ and further that $\mathbf{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}}$ is totally antisymmetric $(\mathbf{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}})^{\dagger} = -(\mathbf{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}})^{*}$.

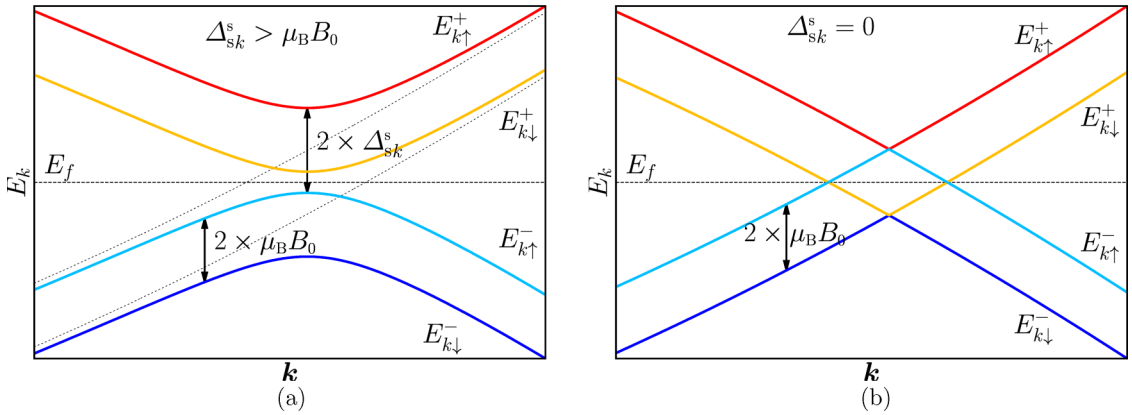


FIG. 1. (Color online) Sketch of the Bogoliubov eigenvalues $E^{\pm}_{k\sigma}$ for a free-electron gas with a homogeneous splitting $\varepsilon_{k\sigma} = \frac{1}{2}\boldsymbol{k}^2 + \mathrm{sign}(\sigma)\mu_{\mathrm{B}} B_0$. We choose a constant $\Delta^{\mathrm{s}}_{sk} > \mu_{\mathrm{B}} B_0$ in (a) and $\Delta^{\mathrm{s}}_{sk} = 0$ in (b). We plot the + Bogoliubov branch in red and orange for $\uparrow$ and $\downarrow$ and the − branch in light blue and dark blue for $\uparrow$ and $\downarrow$, respectively. We indicate the $\varepsilon_{k\sigma}$ in (a) as thin dashed lines. In (a), the + branches are strictly larger than the Fermi energy E_f and thus constitute the SC KS particle excitations. On the other hand, for $\Delta^{\mathrm{s}}_{sk} < \mu_{\mathrm{B}} B_0$ as in (b), the + and − branches partly swap their order. When $E^{-}_{k\uparrow} > E_f$, the SC KS particle excitations are from the − branch as well.

This means that according to the definition in Eq. (13) to take the positive eigenvalues, both KS particles are from the $\sigma = \uparrow$ branch. It is not possible to construct the Bogoliubov transformations in this case and in any case the $\hat{\gamma}^{\dagger}_{k\uparrow}$ state cannot be occupied twice. It is, however, possible to give up the requirement that all SC KS particles are positive and simply always take the + branch. Then, we can say that $\hat{\gamma}^{\dagger}_{k\sigma}$ creates a negative-energy excitation which will be occupied in the ground state. By analogy with BCS, $\hat{\gamma}^{\dagger}_{k\sigma}$ creates an electronlike single-particle state on the SC vacuum, this leads to the interpretation that, in the ground state, this k-space region is occupied by unpaired electrons. A similar discussion can be found (still in the context of BCS theory) in the work of Sarma [47]. Similar to Eq. (13), we can redefine electron to hole operators at the price of changing the ground-state energy. Because the ground-state energy, in turn, cancels from the thermal averages, the expectation values computed with this theory do not depend on this interpretation. We want to point out that this discussion only applies when the splitting is larger than the pair potential.

e. Eigenvectors in the SDA. Furthermore, we can analytically compute the normalized eigenvectors $g^{\alpha}_{k\mu}$ to the eigenvalues $E^{\alpha}_{k\mu}$ ($\alpha = \pm$). As discussed before, it is not known *a priori* if an eigenvalue $E^{\alpha}_{k\mu}$ is positive and, thus, we cannot determine if the components of $g^{\alpha}_{k\mu}$ have to be labeled according to Eq. (30) or (29). We introduce a combined notation for the right-hand side of Eqs. (29) and (30) with an additional index $\alpha = \pm$. Due to the SDA, as compared to Eqs. (29) and (30), only one of the entries $u^{i\sigma}_{k}$ and $v^{i\sigma}_{k}$, respectively, is nonzero. We may thus only denote these two components in our combined notation

$$g^{\alpha}_{k\sigma} = \begin{pmatrix} u^{k\alpha}_{k\sigma} \\ v^{-k\alpha}_{k-\sigma} \end{pmatrix}. \tag{45}$$

When $E^{\alpha}_{k\mu}$ is positive, $u^{k\alpha}_{k\mu}$ ($v^{-k\alpha}_{k-\mu}$) corresponds to the nonvanishing $u^{k\mu}_{k}$ ($v^{-k-\mu}_{k}$); if on the other hand $E^{\alpha}_{k\mu}$ is negative, $u^{k\alpha}_{k\mu}$ ($v^{-k\alpha}_{k-\mu}$) corresponds to $v^{-k-\mu *}_{k}$ ($u^{k\mu *}_{k}$). The components $u^{k\alpha}_{k\sigma}$ and $v^{-k\alpha}_{k-\sigma}$ are given in terms of the eigenvalues and components of the matrix that are diagonalized in Eq. (41) by

$$v^{-k\alpha}_{k-\sigma} = \sqrt{\frac{\left|E^{\alpha}_{k\mu} - \varepsilon_{k\sigma}\right|}{\left|E^{+}_{k\sigma} + E^{+}_{-k-\sigma}\right|}}, \tag{46}$$

$$u^{k\alpha}_{k\sigma} = \frac{\mathrm{sign}(\sigma)}{\mathrm{sign}(\alpha)} \frac{\Delta^{\mathrm{s}}_{sk}}{\left|\Delta^{\mathrm{s}}_{sk}\right|} \sqrt{\frac{\left|\varepsilon_{-k-\sigma} + E^{\alpha}_{k\sigma}\right|}{\left|E^{\alpha}_{k\sigma} + E^{\alpha}_{-k-\sigma}\right|}}. \tag{47}$$

Starting from a converged zero-temperature normal-state calculation, within the SDA the only remaining variable is thus the matrix elements of the pair potential Δ^{s}_{sk} because the SC KS wave functions as well as the Bogoliubov eigenvalues are explicitly given in terms of it.

It is important to point out that within the SDA, Δ^{s}_{sk} can be chosen to be real [40,41]. This can be proven by exploiting the gauge symmetry of Eq. (41) under rotation about the τ_z axis. If the rotation is applied with a k-dependent angle θ_k of

$$\theta_k = \arctan\left(\frac{\mathrm{Im}\Delta^{\mathrm{s}}_{sk}}{\mathrm{Re}\Delta^{\mathrm{s}}_{sk}}\right), \tag{48}$$

we get

$$\begin{aligned} & e^{-i\tau_z \frac{\theta_k}{2}} \begin{pmatrix} \varepsilon_{k\sigma} & \mathrm{sign}(\sigma)\Delta^{\mathrm{s}}_{sk} \\ \mathrm{sign}(\sigma)\Delta^{\mathrm{s}*}_{sk} & -\varepsilon_{-k-\sigma} \end{pmatrix} e^{i\tau_z \frac{\theta_k}{2}} \\ & = \begin{pmatrix} \varepsilon_{k\sigma} & \mathrm{sign}(\sigma)\tilde{\Delta}^{\mathrm{s}}_{sk} \\ \mathrm{sign}(\sigma)\tilde{\Delta}^{\mathrm{s}}_{sk} & -\varepsilon_{-k-\sigma} \end{pmatrix}, \end{aligned} \tag{49}$$

where $\tilde{\Delta}^{\mathrm{s}}_{sk} = \mathrm{sign}(\mathrm{Re}\Delta^{\mathrm{s}}_{sk})|\Delta^{\mathrm{s}}_{sk}| \in \mathbb{R}$. Thus, the $(k,-k)$ matrix elements of our general complex decoupled pair potential are gauge equivalent to purely real ones. We still keep a general complex notation for Δ^{s}_{sk} first to investigate explicitly if self-energy corrections affect this conclusion and, second, to make it easier to extend the formalism to the case where the gauge symmetry does not have enough freedom to make all matrix elements real.

3. Competition between SC and magnetism in the SDA

The SDA, as introduced so far, assumes that we compute SC on top of a (magnetic) quasiparticle structure. Thus, for example, it does not allow magnetism to be suppressed when a weakly magnetic system becomes SC. In conventional SCDFT [33,35], this type of feedback can be safely neglected because SC changes the dispersion only for states very close to the Fermi level. The effect on the electronic density is thus negligible and so is the change in the normal-state xc potential. However, since the contributions to $\boldsymbol{m}(\boldsymbol{r})$ are in general more localized at the Fermi level, assuming quasiparticle energies $\varepsilon_{i\sigma}$ to be unaffected when SC sets in may not be reasonable for magnetic systems.

We want to point out that it is also possible to keep the simple form of the SDA and include competition of SC and magnetism at the same time, by means of the following iterative scheme:

(1) Take the normal KS states $\{\vec{\varphi}_{i\sigma}\}$ and eigenvalues $\varepsilon_{k\sigma}$ as starting orbitals.

(2) Solve the KS–BdG equations in the SDA.

(3) Recompute the densities $n(\boldsymbol{r})$ and $\boldsymbol{m}(\boldsymbol{r})$ according to the Eqs. (32) and (33).

(4) Rediagonalize the normal-state KS equations with the updated densities [in particular, changes in $\boldsymbol{m}(\boldsymbol{r})$ may be of relevance].

(5) Iterate from point 2 until self-consistency is reached.

This procedure changes the meaning of the SDA during the iteration because we are self-consistently updating the orbitals $\{\vec{\varphi}_{i\sigma}\}$ it refers to.

B. Sham-Schlüter equation of SpinSCDFT

So far we have presented the structure of SpinSCDFT with the focus on the electronic SC KS system. The derivation of the approximations for the xc potentials presented in this section generalizes one originally proposed by Marques [48] and later used by Sanna and Gross [49] in SCDFT and uses the Sham-Schlüter equation of SpinSCDFT. This equation is based on the observation that the parts of the KS Green's function (GF) and the interaction GF that correspond to the densities must be equal. Using the Dyson equation for a SC in a magnetic field starting from the SC KS system as the formally noninteraction one, we can relate the xc potentials to an approximation for the self-energy.

We introduce the GF with the τ ordering symbol $\bar{\mathrm{T}}$ and the field operators in the Heisenberg picture

$$\bar{G}(\boldsymbol{r}\tau,\boldsymbol{r}'\tau') = -\langle\bar{\mathrm{T}}\hat{\Psi}(\boldsymbol{r}\tau)\otimes\hat{\Psi}^{\dagger}(\boldsymbol{r}'\tau')\rangle. \tag{50}$$

The imaginary-time ordering symbol in Nambu space $\bar{\mathrm{T}}$ is defined to act on every one of the (4×4) components individually which can be achieved by transposing in Nambu spin space

$$\begin{aligned}\bar{\mathrm{T}}\hat{\Psi}(\boldsymbol{r}\tau)\otimes\hat{\Psi}^{\dagger}(\boldsymbol{r}'\tau') = {}&\theta(\tau-\tau')\hat{\Psi}(\boldsymbol{r}\tau)\otimes\hat{\Psi}^{\dagger}(\boldsymbol{r}'\tau')\\ &-\theta(\tau'-\tau)[\hat{\Psi}^{\dagger}(\boldsymbol{r}'\tau')\otimes\hat{\Psi}(\boldsymbol{r}\tau)]^{\mathrm{T_{sn}}}.\end{aligned} \tag{51}$$

We define the equal-time limit in the $-1,-1$ component different as compared to the usual one in that we take the annihilator operator infinitesimally before the creation operator. The equal-time limit of the time-ordering symbol should be defined to recover the density matrix operator, but the usual rule where the creation operator is taken infinitesimally before the annihilator would lead to the form $\psi\psi^{\dagger}$ in the $-1,-1$ component.

From the equation of motion we derive the Dyson equation starting from the SC KS system as a formally noninteracting system

$$\bar{G}(\boldsymbol{r},\boldsymbol{r}',\omega_n) = \bar{G}^{\mathrm{KS}}(\boldsymbol{r},\boldsymbol{r}',\omega_n) + \int d\boldsymbol{r}_1\int d\boldsymbol{r}'_1\,\bar{G}^{\mathrm{KS}}(\boldsymbol{r},\boldsymbol{r}_1,\omega_n)\cdot\bar{\Sigma}^{\mathrm{s}}(\boldsymbol{r}_1,\boldsymbol{r}'_1,\omega_n)\cdot\bar{G}(\boldsymbol{r}'_1,\boldsymbol{r}',\omega_n), \tag{52}$$

with

$$\bar{\Sigma}^{\mathrm{s}}(\boldsymbol{r},\boldsymbol{r}',\omega_n) = \bar{\Sigma}(\boldsymbol{r},\boldsymbol{r}',\omega_n) - \bar{v}_{\mathrm{xc}}(\boldsymbol{r},\boldsymbol{r}'). \tag{53}$$

Here, $\bar{\Sigma}$ is the irreducible Nambu self-energy, where the electronic Hartree diagram was subtracted, and $\bar{v}_{\mathrm{xc}}$ is the Nambu xc potential

$$\bar{v}_{\mathrm{xc}}(\boldsymbol{r},\boldsymbol{r}') = \begin{pmatrix}\delta(\boldsymbol{r}-\boldsymbol{r}')[\sigma_0 v_{\mathrm{xc}}(\boldsymbol{r}) - \mathbf{S}\cdot\boldsymbol{B}_{\mathrm{xc}}(\boldsymbol{r})] & \boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{xc}}(\boldsymbol{r},\boldsymbol{r}')\\ -\boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{xc}*}(\boldsymbol{r},\boldsymbol{r}') & -\delta(\boldsymbol{r}-\boldsymbol{r}')[\sigma_0 v_{\mathrm{xc}}(\boldsymbol{r}) - \mathbf{S}^{*}\cdot\boldsymbol{B}_{\mathrm{xc}}(\boldsymbol{r})]\end{pmatrix}. \tag{54}$$

The SC KS GF satisfies

$$\int d\boldsymbol{r}_1(i\omega_n\delta(\boldsymbol{r}-\boldsymbol{r}_1)\tau_0\sigma_0 - \bar{H}_{\mathrm{KS}}(\boldsymbol{r},\boldsymbol{r}_1))\cdot\bar{G}^{\mathrm{KS}}(\boldsymbol{r}_1,\boldsymbol{r}',\omega_n) = \delta(\boldsymbol{r}-\boldsymbol{r}')\tau_0\sigma_0. \tag{55}$$

From the equation of motion we can compute the SC KS GF. Because by construction the SC KS GF yields the same densities as the interacting system, we can cancel the respective parts of the GFs in the Dyson equation (52) that correspond to the densities. The result is the Sham-Schlüter equation

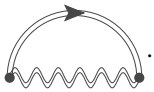
$$\frac{1}{\beta}\sum_n\int d\boldsymbol{r}_1\int d\boldsymbol{r}'_1[\bar{G}^{\mathrm{KS}}(\boldsymbol{r},\boldsymbol{r}_1,\omega_n)\cdot\bar{\Sigma}^{\mathrm{s}}(\boldsymbol{r}_1,\boldsymbol{r}'_1,\omega_n)\cdot\bar{G}(\boldsymbol{r}'_1,\boldsymbol{r}',\omega_n)]_{\alpha,-\alpha} = 0, \tag{56}$$

$$\frac{1}{\beta}\sum_n\int d\boldsymbol{r}_1\int d\boldsymbol{r}'_1[\bar{G}^{\mathrm{KS}}(\boldsymbol{r},\boldsymbol{r}_1,\omega_n)\cdot\bar{\Sigma}^{\mathrm{s}}(\boldsymbol{r}_1,\boldsymbol{r}'_1,\omega_n)\cdot\bar{G}(\boldsymbol{r}'_1,\boldsymbol{r},\omega_n)]_{\alpha,\alpha} = 0. \tag{57}$$

For convenience, the self-energy is decomposed in a phononic part $\bar{\Sigma}_{\rm ph}(\omega_n)$ and a Coulomb part $\bar{\Sigma}_{\rm C}(\omega_n)$:

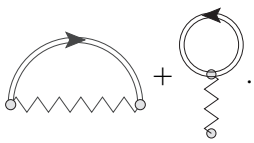
$$\bar{\Sigma}(\omega_n) = \bar{\Sigma}_{\rm ph}(\omega_n) + \bar{\Sigma}_{\rm C}(\omega_n). \tag{58}$$

$\bar{\Sigma}(\omega_n)$ has a diagrammatic expansion in terms of $\bar{G}(\omega_n)$ [24] and can be even viewed as part of a Hedin cycle for a SC including phononic and Coulomb interactions [50]. For simplicity, we do not consider vertex corrections, thus the Coulomb self-energy part $\bar{\Sigma}_{\rm C}$ is the electronic GW diagram

$\bar{\Sigma}_{\rm C}(\omega_n) \approx$  . (59)

As an interesting extension, we could include parts of the vertex corrections that lead to spin fluctuations. These, in the form of an effective spin interaction, are discussed by Essenberger *et al.* [51] and the extension to the present spin-dependent formalism is straightforward.

As compared to the polarization corrections of the same order, the phononic vertex corrections are negligible [52]. Moreover, due to the quality of the phonon spectra one obtains with density functional perturbation theory [37,38] we do not consider further diagrammatic electronic screening and treat the phononic interaction in the Hartree-Fock approximation

$\bar{\Sigma}_{\rm ph}(\omega_n) \approx$  . (60)

It has been observed that computing the GW quasiparticle band structure in a metal gives usually small corrections to the KS bands (compare Ref. [53], Fig. 2), also densities result to be almost identical. Thus, at least in the spin degenerate case, the GW corrections on a KS band structure of a metal are usually negligible. For convenience, we use a similar assumption for the spin part. This way, we can drop the construction of the Nambu diagonal part of $\bar{v}_{\rm xc}$ from the Sham-Schlüter equation. We represent $\bar{G}^{\rm KS}(\boldsymbol{r},\boldsymbol{r}',\omega_n)$ and $\bar{G}(\boldsymbol{r},\boldsymbol{r}',\omega_n)$ in the same basis as the Bogoliubov-Valatin transformations, i.e., the normal-state KS orbitals $\{\Psi^{\rm KS}_{i\sigma\alpha}(\boldsymbol{r})\}$

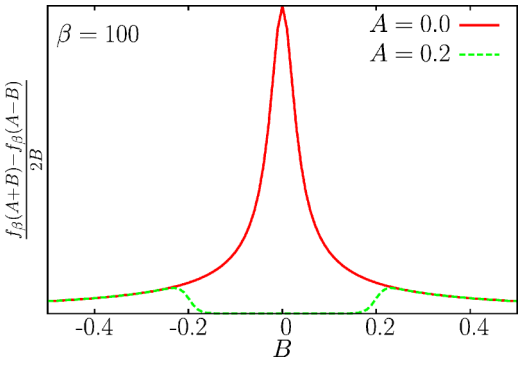


FIG. 2. (Color online) Sketch of the function $\beta a_k^{\sigma,-\sigma}(\Delta^{\rm s})$ multiplying $\Delta^{\rm s}_{sk}$ into the coefficients for $\chi_{\rm s}(\boldsymbol{r},\boldsymbol{r}')$. While being always nonzero, the coefficient function at $B = 0$ behaves as $-(\beta/2)/\cosh[\beta(E^+_{k\sigma} + E^-_{k\sigma})/2]$ and thus becomes exponentially small with decreasing temperature.

with the pure Nambu and spin spinor wave functions

$$\Psi^{\rm KS}_{i\sigma\alpha}(\boldsymbol{r}) = \begin{pmatrix} \delta_{\alpha,1}\vec{\varphi}_{i\sigma}(\boldsymbol{r}) \\ \delta_{\alpha,-1}\vec{\varphi}^*_{i\sigma}(\boldsymbol{r}) \end{pmatrix}. \tag{61}$$

Sorting the expansion coefficients of $\bar{G}^{\rm KS}(\boldsymbol{r},\boldsymbol{r}',\omega_n) = \sum_{\alpha\alpha'\sigma\sigma' ij} \bar{G}^{\rm KS}_{i\alpha\sigma j\alpha'\sigma'}(\omega_n)\Psi^{\rm KS}_{i\alpha\sigma}(\boldsymbol{r}) \otimes \Psi^{\rm KS}_{j\alpha'\sigma'}(\boldsymbol{r}')$ in a similar Nambu and spin matrix form, we obtain the $4N \times 4N$ matrix equation

$$\begin{aligned} &\frac{1}{\beta}\sum_n \bar{G}^{\rm KS}(\omega_n) \cdot \begin{pmatrix} 0 & \boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\rm xc} \\ (\boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\rm xc})^\dagger & 0 \end{pmatrix} \cdot \bar{G}(\omega_n) \\ &= \frac{1}{\beta}\sum_n \bar{G}^{\rm KS}(\omega_n)\Bigg[\begin{pmatrix} 0 & \bar{\Sigma}^{1,-1}_{\rm C}(\omega_n) \\ \bar{\Sigma}^{-1,1}_{\rm C}(\omega_n) & 0 \end{pmatrix} \\ &\quad + \bar{\Sigma}_{\rm ph}(\omega_n)\Bigg]\bar{G}(\omega_n) \end{aligned} \tag{62}$$

that we need to solve for $\boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\rm s}$. From here on, we use $\boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\rm s}$ and $\boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\rm xc}$ synonymously, i.e., the external pair potential is assumed to be infinitesimal.

In the next section, we reduce the problem to the singlet case and employ the SDA. Because in this case we can solve the KSBdG equations analytically, we obtain a potential functional theory and arrive at a functional form that is formally similar to the BCS gap equation. We stress that the methods presented here and in the next section could also be applied without the restriction to the SDA. However, in that case the equations would have an implicit form and require a numerical solution of the KSBdG equations. Such a general form would be of importance to account for pairings beyond the usual one of time-reversed states as would be needed, for example, to describe the FFLO state [5,6]. A further discussion how to go beyond the SDA can be found in Ref. [54].

C. Derivation xc potential

The Sham-Schlüter equation (62) involves the interacting GF which is usually only available after solving the Dyson equation. In an approximate scheme, this step can be avoided. The straightforward way is to replace the matrix $\bar{G}(\omega_n)$ with $\bar{G}^{\rm KS}(\omega_n)$ on all occurrences. As was realized before [33], this approximation violates Migdal's theorem because there a vertex diagram is compared with the polarization diagram of the same order. Thus, the phonon vertex corrections are only negligible as compared to the Hartree exchange diagram with the full GF. To circumvent this problem, some of the authors have introduced a procedure to construct a self-energy that does satisfy Migdal's theorem [49]. Starting from an electron gas model with a phononic Hartree exchange diagram, this leads to excellent agreement with experiment while still retaining the numerically simple form of the Sham-Schlüter equation that is independent on $\bar{G}(\omega_n)$ and involves only Matsubara sums that can be evaluated analytically. The self-energy $\Sigma^{\rm KS}(\omega_n)$, where $G(\omega_n)$ is replaced by $\bar{G}^{\rm KS}(\omega_n)$, so far has been the basis of all further improvements in SCDFT. In this work, as a first step, we will not investigate the parametrization procedure of Ref. [49] or improve the comparison of $T_{\rm c}$ as in Ref. [33]. Instead, we will limit the complexity of the derivation by assuming $\bar{\Sigma}(\omega_n) \approx \bar{\Sigma}^{\rm KS}(\omega_n)$.

This will give inaccurate critical temperatures but qualitatively correct results. Thus, we are left to solve the equation

$$\frac{1}{\beta}\sum_n \bar{G}^{\mathrm{KS}}(\omega_n)\cdot\begin{pmatrix}0 & \boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}}\\ (\boldsymbol{\Phi}\cdot\boldsymbol{\Delta}^{\mathrm{s}})^{\dagger} & 0\end{pmatrix}\cdot\bar{G}^{\mathrm{KS}}(\omega_n)=\frac{1}{\beta}\sum_n \bar{G}^{\mathrm{KS}}(\omega_n)\left[\begin{pmatrix}0 & \bar{\Sigma}_{\mathrm{C}}^{\mathrm{KS}1,-1}(\omega_n)\\ \bar{\Sigma}_{\mathrm{C}}^{\mathrm{KS}-1,1}(\omega_n) & 0\end{pmatrix}+\bar{\Sigma}_{\mathrm{ph}}^{\mathrm{KS}}(\omega_n)\right]\bar{G}^{\mathrm{KS}}(\omega_n). \tag{63}$$

In this form, the matrix elements of the SC KS GF in the normal-state KS basis are given by

$$\bar{G}_{ij}^{\mathrm{KS}}(\omega_n)=\sum_k\frac{1}{i\omega_n-E_k}\begin{pmatrix}\vec{u}_k^i\otimes\vec{u}_k^{j*} & \vec{u}_k^i\otimes\vec{v}_k^{j*}\\ \vec{v}_k^i\otimes\vec{u}_k^{j*} & \vec{v}_k^i\otimes\vec{v}_k^{j*}\end{pmatrix}+\sum_k\frac{1}{i\omega_n+E_k}\begin{pmatrix}\vec{v}_k^{i*}\otimes\vec{v}_k^{j} & \vec{v}_k^{i*}\otimes\vec{u}_k^{j}\\ \vec{u}_k^{i*}\otimes\vec{v}_k^{j} & \vec{u}_k^{i*}\otimes\vec{u}_k^{j}\end{pmatrix}. \tag{64}$$

We use $\vec{u}_k^i=(u_k^{i\uparrow}\quad u_k^{i\downarrow})^{\mathrm{T}}$ with the expansion coefficients $u_k^{i\sigma}$ of $\vec{u}_k(\mathbf{r})$ in $\vec{\varphi}_{i\sigma}(\mathbf{r})$ given in Eq. (23). Similar definitions apply for $\vec{v}_k^i$. Further, we assume the SDA for the rest of this paper. Results beyond the SDA are discussed in Ref. [54]. In the SDA, the SC KS GF simplifies to

$$\bar{G}_{ij}^{\mathrm{KS}}(\omega_n)=\sum_\alpha\begin{pmatrix}\frac{|u_{i\uparrow}^{i\alpha}|^2\delta_{ij}}{i\omega_n-E_{i\uparrow}^{\alpha}} & 0 & 0 & \frac{u_{i\uparrow}^{i\alpha}(v_{i\downarrow}^{-i\alpha})^*\delta_{i,-j}}{i\omega_n-E_{i\uparrow}^{\alpha}}\\ 0 & \frac{|u_{i\downarrow}^{i\alpha}|^2\delta_{ij}}{i\omega_n-E_{i\downarrow}^{\alpha}} & \frac{u_{i\downarrow}^{i\alpha}(v_{i\uparrow}^{-i\alpha})^*\delta_{i,-j}}{i\omega_n-E_{i\downarrow}^{\alpha}} & 0\\ 0 & \frac{(u_{i\uparrow}^{i\alpha})^*v_{i\downarrow}^{-i\alpha}\delta_{i,-j}}{i\omega_n+E_{i\uparrow}^{\alpha}} & \frac{|u_{i\uparrow}^{i\alpha}|^2\delta_{ij}}{i\omega_n+E_{i\uparrow}^{\alpha}} & 0\\ \frac{(u_{i\downarrow}^{i\alpha})^*v_{i\uparrow}^{-i\alpha}\delta_{i,-j}}{i\omega_n+E_{i\downarrow}^{\alpha}} & 0 & 0 & \frac{|u_{i\downarrow}^{i\alpha}|^2\delta_{ij}}{i\omega_n+E_{i\downarrow}^{\alpha}}\end{pmatrix}. \tag{65}$$

This form and any further formula based on it use the components of the SC KS wave function as given in Eqs. (46) and (47). In the Dyson equation $\bar{G}^{-1}=\bar{G}^{\mathrm{KS}^{-1}}-\bar{\Sigma}$ we see that we need to compare the self-energy contributions with the inverse SC KS GF. Inverting $\bar{G}_{ij}^{\mathrm{KS}}(\omega_n)$ we obtain

$$(\bar{G}^{\mathrm{KS}})_{ij}^{-1}(\omega_n)=\delta_{ij}\left[i\omega_n\tau_0\sigma_0-\left(\frac{\varepsilon_{i\uparrow}+\varepsilon_{-i\downarrow}}{2}\right)\tau_z\sigma_0-\left(\frac{\varepsilon_{i\uparrow}-\varepsilon_{-i\downarrow}}{2}\right)\tau_z\sigma_z\right]+\delta_{i,-j}\left[(i\tau_y)(i\sigma_y)\mathrm{Re}\Delta_{\mathrm{s}i}^{\mathrm{s}}+\tau_x(i\sigma_y)i\,\mathrm{Im}\Delta_{\mathrm{s}i}^{\mathrm{s}}\right]. \tag{66}$$

Here, we see that self-energy contributions $\propto\tau_z\sigma_0$ change the average spin Fermi level $\frac{\varepsilon_{i\uparrow}+\varepsilon_{-i\downarrow}}{2}=0$. Similarly, contributions $\propto\tau_z\sigma_z$ change the splitting of single-particle levels. It has to be understood that these are global properties of the band structure, meaning that the full $\varepsilon_{i\sigma}$ dispersion has to be integrated to obtain N electrons per unit cell. If the interaction changes dispersion and occupations far away from the Fermi level, this may still cause a shift of the original Fermi level. A clear-cut example is the following: In the context of SC one often employs the Eliashberg function $\alpha^2F(\Omega)$, which is the Fermi-surface average of the electron-phonon interaction [25,28,29], to describe the electron-phonon interaction. This function is assumed to apply equally to all states, also those away from the Fermi level. This is a good approximation only if corrections of the Fermi level are excluded *a priori* (electron-hole symmetry), otherwise under this assumption the correction to the Fermi level $\frac{\varepsilon_{i\uparrow}+\varepsilon_{-i\downarrow}}{2}$ and the splitting $\frac{\varepsilon_{i\uparrow}-\varepsilon_{-i\downarrow}}{2}$ would show a logarithmic divergence. As commonly done in Eliashberg theory, where the same effect occurs, one then excludes self-energy contributions $\propto\tau_z$. We will assume the same approximation.

Because the Hartree diagram is proportional to τ_z, it is thus not considered. While the expected Fermi energy shift is negligible, corrections to the spin splitting $\frac{\varepsilon_{i\uparrow}-\varepsilon_{-i\downarrow}}{2}$ could be of relevance. However, due to the extreme additional numerical complexity of considering the true full electronic state dependence of the electron-phonon interaction, we leave this to a future project. From now on, we use g exclusively for the electron-phonon coupling. The solutions to the KSBdG equations $g_{k\sigma}^{\pm}$ are referenced only via their components u and v. We compute the self-energy matrix elements in the SDA from Eq. (60):

$$\bar{\Sigma}_{\mathrm{ph}\ i\sigma j\sigma'}^{\mathrm{KS}\,1,1}(\omega_n)=\delta_{\sigma\sigma'}\sum_{qk\alpha}g_{ik\sigma}^{q}g_{kj\sigma}^{-q}\left|u_{k\sigma}^{k\alpha}\right|^2M_{\mathrm{ph}}\left(\Omega_q,E_{k\sigma}^{\alpha},\omega_n\right), \tag{67}$$

$$\bar{\Sigma}_{\mathrm{ph}\ i\sigma j\sigma'}^{\mathrm{KS}\,1,-1}(\omega_n)=-\delta_{\sigma,-\sigma'}\sum_{qk\alpha}g_{ik\sigma}^{q}g_{j,-k,-\sigma}^{-q}u_{k\sigma}^{k\alpha}\left(v_{k-\sigma}^{-k\alpha}\right)^*M_{\mathrm{ph}}\left(\Omega_q,E_{k\sigma}^{\alpha},\omega_n\right), \tag{68}$$

$$\bar{\Sigma}_{\mathrm{ph}\ i\sigma j\sigma'}^{\mathrm{KS}-1,1}(\omega_n)=-\delta_{\sigma,-\sigma'}\sum_{qk\alpha}g_{ki\sigma}^{q}g_{-k,j,-\sigma}^{-q}\left(u_{k\sigma}^{k\alpha}\right)^*v_{k-\sigma}^{-k\alpha}M_{\mathrm{ph}}\left(\Omega_q,-E_{k\sigma}^{\alpha},\omega_n\right), \tag{69}$$

$$\bar{\Sigma}_{\mathrm{ph}\ i\sigma j\sigma'}^{\mathrm{KS}-1,-1}(\omega_n)=\delta_{\sigma\sigma'}\sum_{ak\alpha}g_{ki\sigma}^{q}g_{jk\sigma}^{-q}\left|u_{k\sigma}^{k\alpha}\right|^2M_{\mathrm{ph}}\left(\Omega_q,-E_{k\sigma}^{\alpha},\omega_n\right). \tag{70}$$

From the Hermiticity of $\hat{H}_{\mathrm{KS}}^{\mathrm{e\text{-}ph}}$ of Eq. (10) comes $g_{-q}^{a}(\boldsymbol{r})=[g_q^a(\boldsymbol{r})]^*$ and thus the electron-phonon interaction matrix elements

$$g_{ij\sigma}^{q}=\int d\boldsymbol{r}\sum_{a=0,z}\vec{\varphi}_{i\sigma}^{*}(\boldsymbol{r})\cdot\sigma_a\cdot\vec{\varphi}_{j\sigma}(\boldsymbol{r})g_q^a(\boldsymbol{r}) \tag{71}$$

have the property $g^{q}_{ij\sigma} = g^{-q\,*}_{ji\sigma}$. Moreover, $g^{q}_{ij\sigma} \propto \delta_{\boldsymbol{k}_i,\boldsymbol{k}_j+\boldsymbol{q}}$ which is expected from the lattice translational symmetry [37]. The Matsubara summation $M_{\rm ph}(\Omega,E,\omega_n)$ is evaluated with the result

$$Y_{\rm ph}(\Omega,E,\omega_n) = \frac{1}{\beta}\sum_{n'}\frac{1}{i\omega_{n'}-E}\frac{1}{i(\omega_n-\omega_{n'})+\Omega} \tag{72}$$

$$= \frac{n_\beta(\Omega)+f_\beta(E)}{\Omega-E+i\omega_n}, \tag{73}$$

$$M_{\rm ph}(\Omega,E,\omega_n) = \frac{n_\beta(\Omega)+f_\beta(E)}{\Omega-E+i\omega_n} + \frac{f_\beta(E)+n_\beta(-\Omega)}{\Omega+E-i\omega_n} \tag{74}$$

$$= Y_{\rm ph}(\Omega,E,\omega_n) - Y^{*}_{\rm ph}(\Omega,-E,\omega_n), \tag{75}$$

where $f_\beta(E)$ and $n_\beta(\Omega)$ are Fermi and Bose functions, respectively. The Coulomb self-energy parts on the Nambu off diagonal with the diagram of Eq. (59) are

$$\bar{\Sigma}^{{\rm KS}\,1,-1}_{{\rm C}\;i\sigma j\sigma'}(\omega_n) = -\delta_{\sigma,-\sigma'}\sum_{k\alpha} W^{\rm stat}_{ikj,-k_{\sigma,-\sigma}} u^{k\alpha}_{k\sigma} v^{-k\alpha *}_{k-\sigma} f_\beta\left(E^{\alpha}_{k\sigma}\right), \tag{76}$$

$$\bar{\Sigma}^{{\rm KS}-1,1}_{{\rm C}\;i\sigma j\sigma'}(\omega_n) = -\delta_{\sigma,-\sigma'}\sum_{k\alpha} W^{\rm stat}_{ki,-k,j_{\sigma,-\sigma}} u^{k\alpha *}_{k\sigma} v^{-k\alpha}_{k-\sigma} f_\beta\left(-E^{\alpha}_{k\sigma}\right) \tag{77}$$

with the static screened Coulomb matrix elements

$$W^{\rm stat}_{k_1k_2k_3k_4\sigma\sigma'} = \int d\boldsymbol{r}\int d\boldsymbol{r}'\vec{\varphi}^{*}_{k_1\sigma}(\boldsymbol{r})\cdot\vec{\varphi}_{k_2\sigma}(\boldsymbol{r})\frac{\epsilon^{-1}(\boldsymbol{r},\boldsymbol{r}',0)}{|\boldsymbol{r}-\boldsymbol{r}'|}\vec{\varphi}^{*}_{k_3\sigma'}(\boldsymbol{r}')\cdot\vec{\varphi}_{k_4\sigma'}(\boldsymbol{r}'), \tag{78}$$

with the inverse dielectric function $\epsilon^{-1}(\boldsymbol{r},\boldsymbol{r}',0)$ that is accessible in many electronic-structure codes [55,56]. $\epsilon^{-1}(\boldsymbol{r},\boldsymbol{r}',0)$ is often calculated within the random phase approximation (RPA) which yields very good results for metals in general. As we have pointed out, terms proportional to τ_z, i.e., contributions $(\bar{\Sigma}^{{\rm KS}\,1,1}_{\rm ph} - \bar{\Sigma}^{{\rm KS}-1,-1}_{\rm ph})$ are dropped from the functional construction.

Because of the gauge symmetry discussed in Sec. III A 2 c, we expect the equations for $\Delta^{\rm s}_{sk}$ and $\Delta^{\rm s*}_{sk}$ to be similar. Thus, we proceed and evaluate only the $1,-1$ component of the Sham-Schlüter equation (62) in SDA and arrive at

$$M^{k,-k}_{k,-k}\Delta^{\rm s}_{sk} + M'^{k,-k}_{-k,k}\Delta^{\rm s*}_{sk} = \mathfrak{D}_{k,-k} + \mathfrak{C}_{k,-k}\,. \tag{79}$$

Here, $\mathfrak{D}_{k,-k}$ are the purely phononic contributions due to the Nambu diagonal self-energy parts $\tau_0(\bar{\Sigma}^{{\rm KS}\,1,1}_{\rm ph} + \bar{\Sigma}^{{\rm KS}-1,-1}_{\rm ph})$. $\mathfrak{C}_{k,-k}$ is due to the Nambu off-diagonal self-energy contributions and contains the phononic interaction along with the Coulomb potential on the same footing. The coefficients

$$M^{k,-k}_{k,-k} = \frac{1}{\beta}\sum_{n\sigma}\bar{G}^{{\rm KS}1,1}_{k\sigma,k\sigma}(\omega_n)\bar{G}^{{\rm KS}-1,-1}_{-k,-\sigma,-k,-\sigma}(\omega_n), \tag{80}$$

$$M'^{k,-k}_{-k,k} = \frac{1}{\beta}\sum_{n\sigma}\bar{G}^{{\rm KS}1,-1}_{k\sigma,-k,-\sigma}(\omega_n)\bar{G}^{{\rm KS}-1,1}_{k\sigma,-k,-\sigma}(\omega_n) \tag{81}$$

are the Matsubara summed SC KS GF parts. Note that $M'^{k,-k}_{-k,k}\Delta^{\rm s*}_{sk} \propto |\Delta^{\rm s}_{sk}|^2\Delta^{\rm s}_{sk}$ so the Sham-Schlüter equation in the SDA is unaffected by the phase of $\Delta^{\rm s}_{sk}$, as expected from the gauge symmetry.

$\mathfrak{D}_{kk'}$ and $\mathfrak{C}_{kk'}$ have nonvanishing matrix elements apart from $k,-k$. These are not included in the SDA. Other SC theories such as Eliashberg and spin degenerate SCDFT are built on similar approximations and from the quality of the results one obtains, we conclude that such corrections are in general not important.

Another interesting aspect of the functional construction to observe is that a self-energy part showing tx triplet symmetry appears, that means the spin-inverted Nambu off-diagonal components are not equal and of opposite sign

$$\bar{\Sigma}^{{\rm KS}\,\alpha,-\alpha}_{k\uparrow k'\downarrow} + \bar{\Sigma}^{{\rm KS}\,\alpha,-\alpha}_{k\downarrow k'\uparrow} \neq 0\,. \tag{82}$$

This self-energy part leads to nonvanishing functional contributions in $\mathfrak{C}_{k,-k}$ in the singlet channel. We call these contributions intermediate triplet contributions. We have investigated the effect of removing them and found that this has essentially no consequence in the numerical calculation for a spin-independent coupling (see Paper II [57]). In addition, we note that similar to the matrix elements $k' \neq -k$, the diagrams generate triplet contributions that cannot be incorporated into the SDA. This also means that the terms

$$\sum_{\sigma}\left(\frac{1}{\beta}\sum_{n}\bar{G}^{{\rm KS}1,1}(\omega_n)\bar{\Sigma}^{{\rm KS}1,-1}(\omega_n)\bar{G}^{{\rm KS}-1,-1}(\omega_n)\right)_{\sigma,-\sigma} \neq 0, \tag{83}$$

$$\sum_{\sigma}\left(\frac{1}{\beta}\sum_{n}\bar{G}^{{\rm KS}1,-1}(\omega_n)\bar{\Sigma}^{{\rm KS}-1,1}(\omega_n)\bar{G}^{{\rm KS}1,-1}(\omega_n)\right)_{\sigma,-\sigma} \neq 0 \tag{84}$$

are not zero as, on the other hand, one would expect for a singlet SC. This fact simply means that ignoring the triplet components from the external potential is not consistent, in presence of a magnetic field, because a triplet-singlet coupling exists at the level of the xc potential. As discussed earlier (Sec. III A 2 b), it is not clear in which cases triplet effects become relevant. In this work, we focus on purely singlet SC and will not consider triplet SC further.

Within the SDA, the SC KS wave function components $v^{-k\alpha}_{k-\sigma}, u^{k\alpha}_{k\sigma}$ are explicit functionals of the potential $\Delta^{\rm s}_{\rm s}$.

Thus, the left- and right-hand sides of the Sham-Schlüter equation (79) are equally nonlinear functionals of the potential $\Delta_{\mathrm{s}}^{\mathrm{s}}$. We interpret the Sham-Schlüter condition (79) as

$$S_\beta\big[\Delta_{\mathrm{s}}^{\mathrm{s}}\big]\Delta_{\mathrm{s}}^{\mathrm{s}} = 0, \quad S_\beta = S_\beta^{\mathrm{M}} + S_{\mathrm{ph}\beta}^{\mathfrak{C}} + S_{\mathrm{C}\beta}^{\mathfrak{C}} + S_\beta^{\mathfrak{D}}\,. \tag{85}$$

Here, $S_\beta^{\mathrm{M}}\Delta_{\mathrm{s}}^{\mathrm{s}}$ is equivalent to $-(\Delta_{\mathrm{s}k}^{\mathrm{s}} M_{k,-k}^{k,-k} + M'^{k,-k}_{-k,k}\Delta_{\mathrm{s}k}^{\mathrm{s}*})$, $S_\beta^{\mathfrak{D}}\Delta_{\mathrm{s}}^{\mathrm{s}} = \mathfrak{D}_{k,-k}$, and $(S_{\mathrm{ph}\beta}^{\mathfrak{C}} + S_{\mathrm{C}\beta}^{\mathfrak{C}})\Delta_{\mathrm{s}}^{\mathrm{s}} = \mathfrak{C}_{k,-k}$. The nonlinear Sham-Schlüter operator contributions are given by

$$S_{\beta\,kk'}^{\mathrm{M}} = -\delta_{kk'}\sum_\sigma\left(\frac{(\varepsilon_{k\sigma}+\varepsilon_{-k-\sigma})^2}{|E_{i\sigma}^+ - E_{i\sigma}^-|^2}P_{\mathrm{s}}(E_{k\sigma}^+, E_{k\sigma}^-) + 2\big|u_{k\sigma}^{k+}\big|^2\big|v_{k-\sigma}^{-k+}\big|^2\sum_\alpha P_{\mathrm{s}}\big(E_{k\sigma}^\alpha, E_{k\sigma}^\alpha\big)\right) \tag{86}$$

and

$$\begin{aligned} S_{\beta\,kk'}^{\mathfrak{D}} = \frac{1}{2}\delta_{kk'}\sum_{qk_2\sigma}\sum_{\alpha_1\alpha_2\alpha_3}\frac{\mathrm{sign}(\alpha_3)}{|E_{k\sigma}^+ - E_{k\sigma}^-|}\Big(\big(\big|v_{k_2-\sigma}^{-k_2\alpha_2}\big|^2\big|v_{k-\sigma}^{-k\alpha_1}\big|^2\big|g_{-k_2,-k,-\sigma}^q\big|^2 + \big|u_{k\sigma}^{k\alpha_1}\big|^2\big|u_{k_2\sigma}^{k_2\alpha_2}\big|^2\big|g_{kk_2\sigma}^q\big|^2\big)L\big(\Omega_q, E_{k\sigma}^{\alpha_1}, E_{k_2\sigma}^{\alpha_2}, E_{k\sigma}^{\alpha_3}\big) \\ + \big(\big|v_{k_2-\sigma}^{-k_2\alpha_2}\big|^2\big|v_{k-\sigma}^{-k\alpha_1}\big|^2\big|g_{-k,-k_2,-\sigma}^q\big|^2 + \big|u_{k\sigma}^{k\alpha_1}\big|^2\big|u_{k_2\sigma}^{k_2\alpha_2}\big|^2\big|g_{k_2k\sigma}^q\big|^2\big)L\big(\Omega_q, E_{k\sigma}^{\alpha_1}, -E_{k_2\sigma}^{\alpha_2}, E_{k\sigma}^{\alpha_3}\big)\Big). \end{aligned} \tag{87}$$

The functions P_{s} and L come from analytic Matsubara summations and are given in the Appendix together with various limiting cases. The term $S_{\beta\,kk'}^{\mathfrak{D}}$ due to the Nambu diagonal acts to reduce the critical temperature. In Refs. [33,35], this term was reduced to approximately $\frac{1}{2}$ by subtracting a temperature-sensitive extra term for the purpose of compensating for a systematic underestimation as compared to the Eliashberg critical temperature in the phonon-only case. In Ref. [49], a SCDFT functional is constructed by using a proper interacting GF in the exchange self-energy of Eq. (60), therefore removing the necessity to reduce the repulsive $S_{\beta\,kk'}^{\mathfrak{D}}$. Having in mind to generalize this functional to SpinSCDFT, in this work we decided not to use the scale factor. In Paper II [57], we find further indications that this scaling may also effect the robustness of the SC state against a magnetic splitting. The predicted critical temperature will be too low as compared to experiment, but the correctness of the qualitative behavior of the theory will be preserved. The Nambu off-diagonal contributions that derive from the phonon interaction then read as

$$S_{\mathrm{ph}\beta\,kk'}^{\mathfrak{C}} = -\sum_{q\sigma}\sum_{\alpha_1\alpha_2\alpha_3}\frac{g_{kk'\sigma}^q g_{-k,-k',-\sigma}^{-q}}{\mathrm{sign}(\alpha_2)|E_{k'\sigma}^+ - E_{k'\sigma}^-|}\left(\big|u_{k\sigma}^{k\alpha_1}\big|^2\big|v_{k-\sigma}^{-k\alpha_3}\big|^2 + u_{k\sigma}^{k\alpha_1}v_{k-\sigma}^{-k\alpha_1 *}u_{k\sigma}^{k\alpha_3}v_{k-\sigma}^{-k\alpha_3 *}\frac{\Delta_{\mathrm{s}k'}^{\mathrm{s}*}}{\Delta_{\mathrm{s}k'}^{\mathrm{s}}}\right)L\big(\Omega_q, E_{k\sigma}^{\alpha_1}, E_{k'\sigma}^{\alpha_2}, E_{k\sigma}^{\alpha_3}\big)\,, \tag{88}$$

and the contribution that derives from the static Coulomb interaction reads as

$$\begin{aligned} S_{\mathrm{C}\beta\,kk'}^{\mathfrak{C}} =& -\sum_\sigma\sum_{\alpha_1\alpha_2\alpha_3}\frac{\mathrm{sign}(\alpha_2)}{|E_{k'\sigma}^+ - E_{k'\sigma}^-|}\left(W_{kk',-k,-k'\sigma,-\sigma}^{\mathrm{stat}}\big|u_{k\sigma}^{k\alpha_1}\big|^2\big|v_{k-\sigma}^{-k\alpha_3}\big|^2 + u_{k\sigma}^{k\alpha_1}v_{k-\sigma}^{-k\alpha_1 *}u_{k\sigma}^{k\alpha_3}v_{k-\sigma}^{-k\alpha_3 *}\frac{\Delta_{\mathrm{s}k'}^{\mathrm{s}*}}{\Delta_{\mathrm{s}k'}^{\mathrm{s}}}W_{kk',-k,-k'\sigma,-\sigma}^{\mathrm{stat}\,*}\right) \\ & \times L_{\mathrm{C}}\big(E_{k\sigma}^{\alpha_1}, E_{k'\sigma}^{\alpha_2}, E_{k\sigma}^{\alpha_3}\big). \end{aligned} \tag{89}$$

The function L_{C} from an analytic Matsubara summation is also given in the Appendix. The relation to previous work on SCDFT presented in Ref. [33] is summarized in Sec. III D.

1. Description of the second-order phase transition

If the SC transition to the normal state is of second order, $\chi(\boldsymbol{r},\boldsymbol{r}')$ is assumed to go to zero continuously upon approaching the critical temperature. From earlier work [47] in the BCS framework, we expect this to be the case in the low magnetic field part of the phase diagram. The formalism in the SDA is built on the potential $\Delta_{\mathrm{s}}^{\mathrm{s}}$ not the order parameter χ. We thus need to prove that a second-order phase transition implies also a continuous vanishing of the potential $\Delta_{\mathrm{s}}^{\mathrm{s}}$. We note that in the SDA it is sufficient to show that the expansion coefficients of χ and $\Delta_{\mathrm{s}}^{\mathrm{s}}$ in our normal-state basis are of the form

$$\chi_{\mathrm{s}k\sigma,-k,-\sigma} = a_k^{\sigma,-\sigma}\Delta_{\mathrm{s}k}^{\mathrm{s}}\,, \tag{90}$$

where $a_k^{\sigma,-\sigma}$ is some function of $\Delta_{\mathrm{s}}^{\mathrm{s}}$ and show that $\lim_{|\Delta_{\mathrm{s}}^{\mathrm{s}}|\to 0} a_k^{\sigma,-\sigma}(\Delta_{\mathrm{s}}^{\mathrm{s}}) \neq 0$. Given that this is the case, in the limit $|\Delta_{\mathrm{s}}^{\mathrm{s}}| \to 0$ only linear order terms in the Sham-Schlüter equation are relevant. Then, at a second-order phase transition, T_{c} can be computed from the condition that the matrix $\lim_{|\Delta_{\mathrm{s}}^{\mathrm{s}}|\to 0} S_\beta[\Delta_{\mathrm{s}}^{\mathrm{s}}]$ is singular.

Coming back to Eq. (90) and using the SDA together with Eq. (34) we see

$$a_k^{\sigma,-\sigma} = \frac{f_\beta(E_{k\sigma}^+) - f_\beta(E_{k\sigma}^-)}{|E_{k\sigma}^+ - E_{k\sigma}^-|} \equiv P_{\mathrm{S}}(E_{k\sigma}^+, E_{k\sigma}^-). \tag{91}$$

Clearly, at $T > 0$, $a_k^{\sigma,-\sigma}$ can only be zero if $E_{k\sigma}^+ - E_{k\sigma}^- \to 0$. Taking the respective limit

$$\lim_{E_{k\sigma}^+ - E_{k\sigma}^-\to 0} a_k^{\sigma,-\sigma} = -\frac{\beta}{2}\frac{1}{\cosh[\beta(E_{k\sigma}^+ + E_{k\sigma}^-)/2]} \tag{92}$$

$$< 0, \tag{93}$$

which is the desired result. We may thus use $|\Delta_{\mathrm{s}}^{\mathrm{s}}| \to 0$ instead of $|\chi_{\mathrm{s}}| \to 0$ at the point of a second-order phase transition. We sketch the function $a_k^{\sigma,-\sigma}$ using $A = \beta(E_{k\sigma}^+ + E_{k\sigma}^-)/2$ and $B = \beta(E_{k\sigma}^+ - E_{k\sigma}^-)/2$ in Fig. 2. Note that while $a_k^{\sigma,-\sigma}$ is strictly nonzero if $\beta(E_{k\sigma}^+ + E_{k\sigma}^-)/2 \gg 1$, then $a_k^{\sigma,-\sigma}$ is exponentially small in the range $|B| \ll |A|$.

We thus observe that the order parameter $\chi_s(\boldsymbol{r},\boldsymbol{r}')$ is only weakly dependent on the potential matrix elements Δ^s_{si} that correspond to states below the splitting energy. Still, this does not invalidate the conclusion that at any finite temperature a continuously vanishing order parameter implies a continuously vanishing pair potential. We thus expect that (at low splitting) we can use the linearized Sham-Schlüter equation (85). In the following, we use a breve on top of linearized entities such as $\breve{S}_{\beta_c} = \lim_{|\Delta^s|\to 0} S_\beta[\Delta^s]$ and Eq. (85) can be solved from the condition

$$\det \breve{S}_{\beta_c} = 0, \tag{94}$$

where $\beta_c = 1/T_c$. The right eigenvector of $\breve{S}_{\beta_c}$ is proportional to Δ^s. To compute the small Δ^s limit of $\breve{S}_{\beta_c}$ we first investigate the behavior of $|u^{k\alpha}_{k\sigma}|^2$, $|v^{-k\alpha}_{k-\sigma}|^2$, and $E^\alpha_{k\sigma}$ separately where we find

$$\lim_{|\Delta^s|\to 0} \left|u^{k\alpha}_{k\sigma}\right|^2 = \delta_{\alpha,\mathrm{sign}(\varepsilon_{k\sigma}+\varepsilon_{-k-\sigma})}, \tag{95}$$

$$\lim_{|\Delta^s|\to 0} \left|v^{-k\alpha}_{k-\sigma}\right|^2 = \delta_{\alpha,-\mathrm{sign}(\varepsilon_{k\sigma}+\varepsilon_{-k-\sigma})}, \tag{96}$$

$$\lim_{|\Delta^s|\to 0} E^\alpha_{k\sigma} = \delta_{\alpha,\mathrm{sign}(\varepsilon_{k\sigma}+\varepsilon_{-k-\sigma})}\varepsilon_{k\sigma} - \delta_{\alpha,-\mathrm{sign}(\varepsilon_{k\sigma}+\varepsilon_{-k-\sigma})}\varepsilon_{-k,-\sigma}. \tag{97}$$

Also, we see that

$$\lim_{|\Delta^s|\to 0} \left|u^{k\alpha}_{k\sigma}\right|^2 \left|v^{-k\alpha}_{k-\sigma}\right|^2 = \lim_{|\Delta^s|\to 0} u^{k\alpha}_{k\sigma} v^{-k\alpha *}_{k-\sigma} = 0. \tag{98}$$

Thus, it is straightforward to arrive at

$$\breve{S}^{\mathrm{M}}_{\beta\, kk'} = -2\delta_{kk'} P_s(\varepsilon_{k\uparrow}, -\varepsilon_{-k\downarrow}) \tag{99}$$

and

$$\begin{aligned}\breve{S}^{\mathfrak{D}}_{\beta\, kk'} = \frac{\delta_{kk'}}{\varepsilon_{k\uparrow}+\varepsilon_{-k,\downarrow}} \sum_{ql} \Big(&\left|g^q_{kl\uparrow}\right|^2 [L(\Omega_q,\varepsilon_{k\uparrow},\varepsilon_{l\uparrow},\varepsilon_{k\uparrow}) \\ &+ L(\Omega_q,\varepsilon_{k\uparrow},-\varepsilon_{l\uparrow},\varepsilon_{k\uparrow}) - L(\Omega_q,\varepsilon_{k\uparrow},\varepsilon_{l\uparrow},-\varepsilon_{-k,\downarrow}) \\ &- L(\Omega_q,\varepsilon_{k\uparrow},-\varepsilon_{l\uparrow},-\varepsilon_{-k,\downarrow})] \\ &+ \left|g^q_{-k,-l\downarrow}\right|^2 [L(\Omega_q,\varepsilon_{-k\downarrow},\varepsilon_{-l\downarrow},\varepsilon_{-k\downarrow}) \\ &+ L(\Omega_q,\varepsilon_{-k\downarrow},-\varepsilon_{-l\downarrow},\varepsilon_{-k\downarrow}) - L(\Omega_q,\varepsilon_{-k\downarrow},\varepsilon_{-l\downarrow},-\varepsilon_{k\uparrow}) \\ &- L(\Omega_q,\varepsilon_{-k\downarrow},-\varepsilon_{-l\downarrow},-\varepsilon_{k\uparrow})]\Big).\end{aligned} \tag{100}$$

Moreover,

$$\begin{aligned}\breve{S}^{\mathfrak{C}}_{\mathrm{ph}\beta\, kk'} = -2\sum_q \frac{g^q_{kk'\uparrow} g^{-q}_{-k,-k',\downarrow}}{|\varepsilon_{k'\uparrow}+\varepsilon_{-k',\downarrow}|} &[L(\Omega_q,\varepsilon_{k\uparrow},\breve{E}^+_{k'\uparrow},-\varepsilon_{-k\downarrow}) \\ &+ L(\Omega_q,\varepsilon_{-k\downarrow},\breve{E}^+_{-k'\downarrow},-\varepsilon_{k\uparrow})]\end{aligned} \tag{101}$$

and

$$\begin{aligned}&\breve{S}^{\mathfrak{C}}_{\mathrm{C}\beta\, kk'} \\ &= -2W^{\mathrm{stat}}_{kk',-k,-k'\uparrow,\downarrow} P_s(\breve{E}^+_{k'\uparrow},-\breve{E}^+_{-k'\downarrow}) P_s(\varepsilon_{k\uparrow},-\varepsilon_{-k\downarrow}).\end{aligned} \tag{102}$$

2. Nonlinear gap equation

Far from T_c or in those parts of the phase diagram where the SC transition is of first order we need to use the nonlinear Sham-Schlüter equation, because a solution with small $|\Delta^s_{sk}|$ may not exist. The most common method to solve an equation of type Eq. (85) is to use an invertible splitting matrix $\mathcal{S}$ and cast $S_\beta[\Delta^s]\Delta^s = 0$ into a fixed-point problem

$$\Delta^s = \mathcal{K}_\mathcal{S}[\Delta^s]\Delta^s, \quad \mathcal{K}_\mathcal{S} = \mathcal{S}^{-1}(S_\beta + \mathcal{S}). \tag{103}$$

This is the gap equation of SpinSCDFT. In the spin degenerate limit, the choice $\mathcal{S} = -\breve{S}^{\mathrm{M}}_\beta$ leads to the SCDFT gap equation given in Ref. [33]. We point out that while we can show that all $\breve{S}^{\mathrm{M}}_{\beta k,k} < 0$ at $\varepsilon_{k\sigma}+\varepsilon_{-k,-\sigma} = 0$ $\breve{S}^{\mathrm{M}}_{\beta k,k} \sim \exp[-\frac{1}{2}\beta(\varepsilon_{k\sigma} - \varepsilon_{-k,-\sigma})]$ and is thus a numerically problematic object. In the implementation that we describe in detail in Paper II [57], we find that a good choice is $\mathcal{S} = -S^{\mathrm{M}}_\beta(\varepsilon_{k\sigma} = \varepsilon_{-k,-\sigma})$. Obviously, in the spin degenerate limit we recover the formulas given in Ref. [33]. In Paper II, we will also discuss the properties of the splitting versus temperature diagram for a simple system in detail.

D. Relation to earlier work on SCDFT

Previous work on SCDFT in Ref. [33] led to a similar gap equation as Eq. (103) using KS perturbation theory. While a nonlinear functional is derived in Ref. [33] as well, in most applications only the linear or partially linearized equation has been used. In the partially linearized gap equation, the temperature behavior of the gap below T_c is modeled using the BCS hyperbolic tangent [33]. On the other hand, in the fully linearized approach, KS perturbation theory and the derivation via the Sham-Schlüter equation lead to the same results [48]. Thus, we focus on the comparison of results form a linearization. In Ref. [48], the Sham-Schlüter equation is kept as in Eq. (63) also within the decoupling approximation in the form where the susceptibilitylike terms $M^{k,-k}_{k,-k}$ and $M'^{k,-k}_{-k,k}$ times the potential Δ^s is on the left-hand side of the equation while the self-energy term is on the right-hand side. Note that M' is of higher than linear order.

To solve the resulting equation for the potential Δ^s, it seems natural to divide by the function $M^{k,-k}_{k,-k} = \frac{1}{\beta}\sum_{n\sigma} \bar{G}^{\mathrm{KS}1,1}_{k\sigma,k\sigma}(\omega_n)\bar{G}^{\mathrm{KS}-1,-1}_{-k,-\sigma,-k,-\sigma}(\omega_n)$. Doing so, we arrive at the analog of Eq. (35) in Ref. [33] that reads as in our notation

$$\Delta^s_{sk} = -\mathcal{Z}_k\Delta^s_{sk} - \frac{1}{2}\sum_{k'} \mathcal{K}_{k,k'} \frac{\tanh\left(\frac{\beta}{2}\varepsilon_{k'}\right)}{\varepsilon_{k'}} \Delta^s_{sk'}, \tag{104}$$

where, following the derivation of this work, we have

$$\mathcal{Z}_k = -\breve{S}^{\mathfrak{D}}_{\beta\, k,k} / \breve{M}^{k,-k}_{k,-k}, \quad \mathcal{K}_{k,k'} = -\breve{S}^{\mathfrak{C}}_{\beta\, kk'} / \breve{M}^{k,-k}_{k,-k}. \tag{105}$$

In the spin degenerate limit, this is the linear result in Ref. [33]. Note that there is a problem with $\breve{M}^{k,-k}_{k,-k}$ if the splitting is not zero as we discuss in the following, which is why the more general approach of this work has been introduced.

We show the function a in Fig. 2 which is equivalent to the function P_s given in the Appendix. From this plot and Eq. (99) can be understood that $(\breve{M}^{k,-k}_{k,-k})^{-1} = (\breve{S}^{\mathrm{M}}_{\beta kk'})^{-1}$ is numerically not well behaved at finite splitting and low temperatures. Since $\breve{M}^{k,-k}_{k,-k}$ represents the connection between χ and Δ to linear order, the application of the functional derivative $\frac{\delta\Delta}{\delta\chi}$ as in Ref. [33] is equally problematic in this limit.

Within the decoupling approximation, the right-hand side of the Sham-Schlüter equation equally depends on Δ^s. So, in this work we use the entire equation directly without first

separating the potential term. The solution can be found using a fixed-point iteration with a more general splitting matrix. This directly leads to the fixed-point equation (103).

As compared to the previous work on SCDFT, the Matsubara summations in $G^{\mathrm{KS}}\Sigma G^{\mathrm{KS}}$ that result in the functions $L(\Omega,E_1,E_2,E_3)$ and $L_{\mathrm{C}}(E_1,E_2,E_3)$ given in the Appendix are complicated by the fact that E_3 is not necessarily equal or opposite equal to E_1. Thus, L does not reduce to I, I', J, and J' of Ref. [33] that depend only on two energy variables.

Most importantly, we need to solve a nonlinear gap equation since the high-field part of the phase diagram in a magnetic field shows a transition that is first order. Also, a partial linearization is thus not straightforward.

IV. ELIASHBERG EQUATIONS

In the KS-SpinSCDFT formalism, interaction effects are mimicked by the xc potential that is an (implicit) functional of the densities. While the functional construction and the additional complications of the SC KS system pose additional algebraic complexity, the result is a numerically cheaper computational scheme. This is owed to the fact that Matsubara summations in the self-energy are not computed numerically but absorbed into the analytic structure of the xc potential. Likely, the knowledge of the interacting self-energy is essential to a future improvement of the presented functional. The self-energy (53) in turn is constructed via diagrammatic perturbation theory using the electronic and phononic GF similar to Sec. III C, and involving the solution of a Dyson equation. In this section, we develop this direct many-body scheme to obtain the electronic GF. The final set of equations generalize the ones of Eliashberg [29] and we refer to them with the same name. Reference [24] discusses similar equations in a different notation with a limitation to isotropic system with a homogeneous splitting parameter.

A. Solving the Dyson equation

The starting point of the derivation of the Eliashberg equations is the Dyson equation of a SC equation (52). We represent it in the basis of normal-state, zero-temperature KS orbitals $\{\vec{\varphi}_{i\sigma}(\boldsymbol{r})\}$ defined in Eq. (22). We use the Nambu-Anderson [40,41] notation similar to that used in the functional derivation and in Eq. (62). The Dyson equation reads as

$$\bar{G}_{ij}(\omega_n)=\bar{G}^{\mathrm{KS}}_{ij}(\omega_n)+\sum_{kl}\bar{G}^{\mathrm{KS}}_{ik}(\omega_n)\bar{\Sigma}^{\mathrm{s}}_{kl}(\omega_n)\bar{G}_{lj}(\omega_n), \quad (106)$$

where $\bar{G}^{\mathrm{KS}}_{ij}$ is the SC KS GF and $\bar{\Sigma}^{\mathrm{s}}_{ij}(\omega_n)=\bar{\Sigma}_{\mathrm{xc}ij}(\omega_n)-\bar{v}_{\mathrm{xc}ij}$ where $\bar{\Sigma}_{\mathrm{xc}ij}(\omega_n)$ is the Nambu exchange and correlation self-energy that also includes the phononic Hartree diagram [50]. $\bar{v}_{\mathrm{xc}ij}$ are the matrix elements of the xc potential of the SC KS system. Note that the SC KS GF is not diagonal in the space of $\{\vec{\varphi}_{i\sigma}(\boldsymbol{r})\}$. Similar to our approach in SpinSCDFT of Sec. III, we assume that $\{\vec{\varphi}_{i\sigma}(\boldsymbol{r})\}$ is a good approximation to the quasiparticle state,[4] i.e., $\bar{\Sigma}^{\mathrm{s}}_{kl}(\omega_n)$ and $\bar{G}_{ij}(\omega_n)$ are essentially diagonal. We use similar diagrams [Eqs. (59) and (60)] as for the functional construction of SpinSCDFT in Sec. III C, namely, the phononic and Coulomb exchange diagram. Again similarly (compare Sec. III C) we drop the Coulomb corrections on the Nambu diagonal that add to the xc potential. Further, we assume, as in the SDA of Sec. III A 2 c, that the pairing occurs only between time-reversed states [46]. This means we only consider singlet SC. Starting from Eq. (106) in the form $\bar{G}_{ij}(\omega_n)=[\bar{G}^{\mathrm{KS}^{-1}}_{ij}(\omega_n)-\bar{\Sigma}^{\mathrm{s}}_{ij}(\omega_n)]^{-1}$, under the mentioned approximations, the Dyson equation is a 4×4 matrix equation that can be solved analytically. Note that here we do not substitute the SC KS GF for the interaction GF in the self-energy (as was done in the functional construction of SpinSCDFT of Sec. III C). This is the main difference in the two approaches so far.

TABLE I. Self-energy contributions, the variable of the inverse SC KS GF which they add to, and the basis vector, e.g., along the $\tau_0\sigma_0$ direction in spin and Nambu space $i\omega_n+\Sigma^{\omega}_k(\omega_n)$. In the last column, we give the related Eliashberg property. Note that $\Delta^{\mathrm{E}}_k\sim\Sigma^{\mathrm{Re}\Delta}_k+i\,\Sigma^{\mathrm{Im}\Delta}_k$ and $\Delta^{\mathrm{E}\star}_k(\omega_n)\sim\Sigma^{\mathrm{Re}\Delta}_k-i\,\Sigma^{\mathrm{Im}\Delta}_k$.

SE part	$\bar{G}^{\mathrm{KS}^{-1}}$ part	Basis vector	Eliashberg
$\Sigma^{\omega}_k(\omega_n)$	$i\omega_n$	$\tau_0\sigma_0$	$Z_k(\omega_n)$
$A^{\omega}_k(\omega_n)$		$\tau_0\sigma_z$	$\tilde{A}^{\omega}_k(\omega_n)$
$\Sigma^{\varepsilon}_k(\omega_n)$	$(\varepsilon_{k\uparrow}+\varepsilon_{-k\downarrow})/2$	$\tau_z\sigma_0$	$\tilde{\varepsilon}_k(\omega_n)$
$\Sigma^{J}_k(\omega_n)$	$(\varepsilon_{k\uparrow}-\varepsilon_{-k\downarrow})/2$	$\tau_z\sigma_z$	$\tilde{J}_k(\omega_n)$
$\Sigma^{\mathrm{Re}\Delta}_k(\omega_n)$	$\mathrm{Re}\Delta^{\mathrm{s}}_{sk}$	$(i\tau_y)(i\sigma_y)$	$\Delta^{\mathrm{E}}_k(\omega_n)$
$\mathrm{i}\Sigma^{\mathrm{Im}\Delta}_k(\omega_n)$	$i\,\mathrm{Im}\Delta^{\mathrm{s}}_{sk}$	$\tau_x(i\sigma_y)$	$\Delta^{\mathrm{E}\star}_k(\omega_n)$

1. *Analytic inversion of the Dyson equation*

The easiest way to invert the right-hand side of the Dyson equation

$$\bar{G}_{ij}(\omega_n)=\left[\bar{G}^{\mathrm{KS}^{-1}}_{ij}(\omega_n)-\bar{\Sigma}^{\mathrm{s}}_{ij}(\omega_n)\right]^{-1} \quad (107)$$

is to identify contributions of the self-energy that add to a given variable of the inverse SC KS GF $\bar{G}^{\mathrm{KS}^{-1}}_{ij}(\omega_n)$ of Eq. (66). We summarize these self-energy contributions in Table I. This means we decompose the Nambu and spin matrix $\bar{\Sigma}^{\mathrm{s}}_{kl}(\omega_n)$ along the vectors $\tau_0\sigma_0$, $\tau_z\sigma_0$, and so on. Then, we name the self-energy contributions according to the property of the SC KS GF they add to in Eq. (107) and indicate the property in the superscript. For example, the Matsubara frequency variable of the inverse SC KS GF points along the $\tau_0\sigma_0$ axis in spin and Nambu space. Correspondingly, the self-energy part along basis vector is referred to as $\Sigma^{\omega}_k(\omega_n)$. In the following, we use $|g^{q}_{kk'\sigma}|^2=|g^{-q}_{k'k\sigma}|^2$, $D^0_{q,-q}=D^0_{-q,q}$, and $W^{\mathrm{stat}}_{kk'k'k\sigma\sigma}=W^{\mathrm{stat}}_{k'kkk'\sigma\sigma}$. Then, the equations for the corresponding scalar self-energy components read as

$$\Sigma^{\omega}_k(\omega_n)=\frac{1}{4}\sum_{\sigma\alpha k'q}\frac{1}{\beta}\sum_{n'}(\tau_0)_{\alpha\alpha}D^0_{q,-q}(\omega_{n'}-\omega_n)\times\left|g^{q}_{kk'\sigma}\right|^2\bar{G}^{\alpha,\alpha}_{k'\sigma,k'\sigma}(\omega_{n'}), \quad (108)$$

[4]The same scheme for going beyond the decoupling approximation presented in Sec. III A 3 can be used in this Eliashberg approach: the KS orbital basis could, in principle, be self-consistently updated with modified densities in the SC state.

$$A_k^{\omega}(\omega_n) = \frac{1}{4}\sum_{\sigma\alpha k' q}\frac{1}{\beta}\sum_{n'}\frac{(\tau_0)_{\alpha\alpha}}{\mathrm{sign}(\sigma)} D^0_{q,-q}(\omega_{n'}-\omega_n) \times \left|g^q_{kk'\sigma}\right|^2 \bar{G}^{\alpha,\alpha}_{k'\sigma,k'\sigma}(\omega_{n'}), \tag{109}$$

$$\Sigma_k^{\varepsilon}(\omega_n) = \frac{1}{4}\sum_{\sigma\alpha k' q}\frac{1}{\beta}\sum_{n'}(\tau_z)_{\alpha\alpha} D^0_{q,-q}(\omega_{n'}-\omega_n) \times \left|g^q_{kk'\sigma}\right|^2 \bar{G}^{\alpha,\alpha}_{k'\sigma,k'\sigma}(\omega_{n'}), \tag{110}$$

$$\Sigma_k^{J}(\omega_n) = \frac{1}{4}\sum_{\sigma\alpha k' q}\frac{1}{\beta}\sum_{n'}\frac{(\tau_z)_{\alpha\alpha}}{\mathrm{sign}(\sigma)} D^0_{q,-q}(\omega_{n'}-\omega_n) \times \left|g^q_{kk'\sigma}\right|^2 \bar{G}^{\alpha,\alpha}_{k'\sigma,k'\sigma}(\omega_{n'}). \tag{111}$$

Note that $A_k^{\omega}(\omega_n)$ stands out in the sense that the SC KS GF has no contribution along this direction in Nambu and spin space. On the Nambu off-diagonal we similarly introduce

$$\Sigma_k^{\mathrm{Im}\Delta}(\omega_n) = -\sum_{\sigma k'}\frac{1}{\beta}\sum_{n'}\frac{\mathrm{sign}(\sigma)}{4\mathrm{i}}\left(\sum_q D^0_{q,-q}(\omega_{n'}-\omega_n) \times g^q_{kk'\sigma} g^{-q}_{-k,-k',-\sigma} + W^{\mathrm{stat}}_{kk',-k,-k'\sigma,-\sigma}\right) \times \sum_{\alpha}(\tau_x)_{\alpha\alpha}\bar{G}^{\alpha,-\alpha}_{k'\sigma,-k',-\sigma}(\omega_{n'}), \tag{112}$$

$$\Sigma_k^{\mathrm{Re}\Delta}(\omega_n) = -\sum_{\sigma k'}\frac{1}{\beta}\sum_{n'}\frac{\mathrm{sign}(\sigma)}{4}\left(\sum_q D^0_{q,-q}(\omega_{n'}-\omega_n) \times g^q_{k'k\sigma} g^{-q}_{-k',-k,-\sigma} + W^{\mathrm{stat}}_{k'k,-k',-k_{\sigma,-\sigma}}\right) \times \sum_{\alpha}(i\tau_y)_{\alpha\alpha}\bar{G}^{\alpha,-\alpha}_{k'\sigma,-k',-\sigma}(\omega_{n'})\,. \tag{113}$$

Here, we introduce $B_k(\omega_n) = \Sigma_k^{\mathrm{Re}\Delta}(\omega_n) + i\,\Sigma_k^{\mathrm{Im}\Delta}(\omega_n)$ and $B_k^{\star}(\omega_n) = \Sigma_k^{\mathrm{Re}\Delta}(\omega_n) - i\,\Sigma_k^{\mathrm{Im}\Delta}(\omega_n)$:

$$B_k(\omega_n) = -\sum_{\sigma k'}\frac{1}{\beta}\sum_{n'}\frac{\mathrm{sign}(\sigma)}{2}\left(\sum_q D^0_{q,-q}(\omega_{n'}-\omega_n) \times g^q_{kk'\sigma} g^{-q}_{-k,-k',-\sigma} + W^{\mathrm{stat}}_{kk',-k,-k'\sigma,-\sigma}\right) \times \bar{G}^{1,-1}_{k'\sigma,-k',-\sigma}(\omega_{n'}), \tag{114}$$

$$B_k^{\star}(\omega_n) = \sum_{\sigma k'}\frac{1}{\beta}\sum_{n'}\frac{\mathrm{sign}(\sigma)}{2}\left(\sum_q D^0_{q,-q}(\omega_{n'}-\omega_n) \times g^q_{k'k\sigma} g^{-q}_{-k',-k,-\sigma} + W^{\mathrm{stat}}_{k'k,-k',-k_{\sigma,-\sigma}}\right) \times \bar{G}^{-1,1}_{k'\sigma,-k',-\sigma}(\omega_{n'}). \tag{115}$$

If both $\Sigma_k^{\mathrm{Re}\Delta}$ and $\Sigma_k^{\mathrm{Im}\Delta}$ are real, $B_k^{\star}$ is the complex conjugate of B_k. Further, for the same reasons discussed in Sec. III A 2 b, we do not consider the possibility that triplet self-energy contributions appear. It is important to remark that, just as in the usual derivation of the spin degenerate Eliashberg equations, the k dependence of all self-energy parts is generated via the k dependence of the couplings $|g^q_{kk'\sigma}|^2$ and in addition $W^{\mathrm{stat}}_{k'k,-k',-k_{\sigma,-\sigma}}$ on the Nambu off diagonal.

Introducing the mass renormalization function $Z_k(\omega_n)$ as

$$Z_k(\omega_n) = 1 + i\,\Sigma_k^{\omega}(\omega_n)\big/\omega_n\,, \tag{116}$$

we can rewrite some of the above equations by including $Z_k^{\mathrm{E}}(\omega_n)$ into the self-energy parts

$$\Delta_k^{\mathrm{E}}(\omega_n) = B_k(\omega_n)/Z_k(\omega_n), \tag{117}$$

$$\Delta_k^{\mathrm{E}\star}(\omega_n) = B_k^{\star}(\omega_n)/Z_k(\omega_n), \tag{118}$$

$$\tilde{\varepsilon}_k(\omega_n) = \left[(\varepsilon_{k\uparrow}+\varepsilon_{-k\downarrow})/2 + \Sigma_k^{\varepsilon}(\omega_n)\right]\big/Z_k(\omega_n), \tag{119}$$

$$\tilde{J}_k(\omega_n) = \left[(\varepsilon_{k\uparrow}-\varepsilon_{-k\downarrow})/2 + \Sigma_k^{J}(\omega_n)\right]\big/Z_k(\omega_n), \tag{120}$$

$$\tilde{A}_k^{\omega}(\omega_n) = A_k^{\omega}(\omega_n)\big/Z_k(\omega_n). \tag{121}$$

Then, by introducing the abbreviation

$$\mathfrak{F}_{k\sigma}(\omega_n) = \left(\left[\tilde{\varepsilon}_k(\omega_n) + \mathrm{sign}(\sigma)\tilde{A}_k^{\omega}(\omega_n)\right]^2 + \Delta_k^{\mathrm{E}}(\omega_n)\Delta_k^{\mathrm{E}\star}(\omega_n)\right)^{\frac{1}{2}}, \tag{122}$$

and suppressing the arguments ω_n, we arrive at the formulas for nonvanishing SC GF components:

$$\bar{G}^{1,1}_{k\sigma,k\sigma} = \frac{1}{2\mathfrak{F}_{k\sigma}Z_k}\sum_{\alpha}\frac{\mathfrak{F}_{k\sigma} + \alpha\left[\tilde{\varepsilon}_k + \mathrm{sign}(\sigma)\tilde{A}_k^{\omega}\right]}{i\omega_n - \mathrm{sign}(\sigma)\tilde{J}_k - \alpha\mathfrak{F}_{k\sigma}}, \tag{123}$$

$$\bar{G}^{-1,-1}_{k\sigma,k\sigma} = \frac{1}{2\mathfrak{F}_{k,-\sigma}Z_k}\sum_{\alpha}\frac{\mathfrak{F}_{k,-\sigma} + \alpha\left[\tilde{\varepsilon}_k - \mathrm{sign}(\sigma)\tilde{A}_k^{\omega}\right]}{i\omega_n + \mathrm{sign}(\sigma)\tilde{J}_k + \alpha\mathfrak{F}_{k,-\sigma}}, \tag{124}$$

$$\bar{G}^{1,-1}_{k\sigma,-k,-\sigma} = \frac{1}{2\mathfrak{F}_{k\sigma}Z_k}\sum_{\alpha}\frac{\mathrm{sign}(\sigma)\alpha\Delta_k^{\mathrm{E}}}{i\omega_n - \mathrm{sign}(\sigma)\tilde{J}_k - \alpha\mathfrak{F}_{k\sigma}}, \tag{125}$$

$$\bar{G}^{-1,1}_{k\sigma,-k,-\sigma} = \frac{1}{2\mathfrak{F}_{k,-\sigma}Z_k}\sum_{\alpha}\frac{\mathrm{sign}(\sigma)\alpha\Delta_k^{\mathrm{E}\star}}{i\omega_n + \mathrm{sign}(\sigma)\tilde{J}_k + \alpha\mathfrak{F}_{k,-\sigma}}. \tag{126}$$

We have thus expressed the GF in terms of the self-energy components [Eqs. (117) to (121)] explicitly. The coupled set of equations (117) to (121) are the Eliashberg equations and have to be solved according to the following scheme:

(1) Start with the coupling matrix elements $g^q_{k'k\sigma}$ and $W^{\mathrm{stat}}_{k'k,-k',-k_{\sigma,-\sigma}}$ and an initial guess for the self-energy components Δ_k^{E}, $\Delta_k^{\mathrm{E}\star}$, $\tilde{\varepsilon}_k$, $\tilde{J}_k$, and $\tilde{A}_k^{\omega}$.

(2) Evaluate Eqs. (117) to (121). They are closed in the sense that inserting the equations of this section Δ_k^{E}, $\Delta_k^{\mathrm{E}\star}$, $\tilde{\varepsilon}_k$, $\tilde{J}_k$, and $\tilde{A}_k^{\omega}$ are only dependent on each other and the coupling matrix elements $g^q_{k'k\sigma}$ and $W^{\mathrm{stat}}_{k'k,-k',-k_{\sigma,-\sigma}}$.

(3) Construct a new self-energy and iterate from point 2, up to self-consistency.

One may insert the expression for the GF into the self-energy components [Eqs. (117) to (121)] which would provide us with the magnetic generalization to the equations derived in Ref. [26]. $\tilde{A}_k^\omega$ is a peculiar object because it generates a spin imbalance in the particle as compared to the hole channel. To understand the effect of $\tilde{A}_k^\omega$, consider the following self-consistent cycle. We start the iteration of these equations with $\tilde{A}_k^\omega = 0$ and $\Sigma_k^{\mathrm{Im}\Delta} = 0$. Then follows $\bar{G}^{1,-1}_{k\sigma,-k,-\sigma} = \bar{G}^{-1,1}_{k,-\sigma,-k\sigma}$ which results in $B_k^\star = B_k$ and no self-energy part $\Sigma_k^{\mathrm{Im}\Delta}$ is generated. Further, because $\bar{G}^{-1,-1}_{k\sigma,k\sigma} = \bar{G}^{1,1}_{k,-\sigma k,-\sigma}$ we find then that $\tilde{A}_k^\omega$ is proportional to the difference of the interaction in the spin channels $\tilde{A}_k^\omega \propto |g^q_{kk'\uparrow}|^2 - |g^q_{kk'\downarrow}|^2$. If now the interaction is independent on the spin channel $|g^q_{kk'\uparrow}|^2 - |g^q_{kk'\downarrow}|^2 = 0$, then $\tilde{A}_k^\omega$ also remains zero and we are at our starting point. Thus, we conclude that for spin-independent couplings $\Sigma_k^{\mathrm{Im}\Delta}$ and $\tilde{A}_k^\omega$ remain zero during iteration. If the interaction *is* spin dependent $|g^q_{kk'\uparrow}|^2 - |g^q_{kk'\downarrow}|^2 \neq 0$ the self-consistency iteration will generate a spin imbalance in the GF. This is not surprising because the up and down single-particle spectrum is altered in a different way by the interaction. Then, a nonvanishing $\Sigma_k^{\mathrm{Im}\Delta}$ cannot be excluded.

For future reference, we extract the renormalized energy dependence $\tilde{\varepsilon}_k$ of the GF as it appears in the self-energy equations (108) to (111) and (114) and (115). With the abbreviation

$$a_k(\omega_n) = \left(\tilde{A}_k^{\omega z}\right)^2 + \Delta_k^{\mathrm{E}}\Delta_k^{\mathrm{E}\star} + \omega_n^2 - (\tilde{J}_k)^2 \tag{127}$$

we obtain ($b = 0,z$)

$$\sum_\alpha (\tau_b)_{\alpha\alpha}\bar{G}^{\alpha,\alpha}_{k\sigma,k\sigma} = \sum_\alpha \frac{(\tau_b)_{\alpha\alpha}}{Z_k}\frac{\alpha\left(\tilde{\varepsilon}_k - \mathrm{sign}(\sigma)\left(\tilde{J}_k + \alpha\tilde{A}_k^\omega\right)\right) - i\omega_n}{a_k - \frac{2\alpha}{\mathrm{sign}(\sigma)}\left(i\omega_n\tilde{J}_k + \tilde{A}_k^\omega\tilde{\varepsilon}_k\right) + \tilde{\varepsilon}_k^2} \tag{128}$$

and

$$\sum_\sigma \bar{G}^{1,-1}_{k\sigma,-k,-\sigma}(\omega_n) = -\frac{1}{Z_k}\sum_\alpha \frac{\Delta_k^{\mathrm{E}}}{a_k - \alpha\left(i\omega_n\tilde{J}_k + \tilde{A}_k^\omega\tilde{\varepsilon}_k\right) + \tilde{\varepsilon}_k^2}. \tag{129}$$

B. Analytic integration of the energy

In a numerical solution, Eqs. (117) to (121) have to be iterated until self-consistency is reached. Each self-consistent step requires to perform Matsubara summations in addition to the k-space summations which will be numerically demanding.

Note, however, that the k-space summations can be avoided using an approximation that is very common in the context of Eliashberg theory which is essentially to replace the couplings with their value at $\tilde{\varepsilon}_k(\omega_n) = 0$. The reason why this is sensible can be understood from the GF. From the above equation (128) one can easily see that $\bar{G}^{1,1}_{k\sigma,k\sigma}(\omega_n) - \bar{G}^{-1,-1}_{k\sigma,k\sigma}(\omega_n)$ behaves as $[\tilde{\varepsilon}_k(\omega_n)]^{-1}$ for large $\tilde{\varepsilon}_k(\omega_n)$. In turn, $\bar{G}^{1,1}_{k\sigma k\sigma}(\omega_n) + \bar{G}^{-1,-1}_{k\sigma k\sigma}(\omega_n)$ and the Nambu off-diagonal parts $\bar{G}^{\alpha,-\alpha}_{k\sigma,-k,-\sigma}(\omega_n)$ behave as $[\tilde{\varepsilon}_k(\omega_n)]^{-2}$ for large $\tilde{\varepsilon}_k(\omega_n)$. Using this insight, we see from the Eqs. (108), (109), (114), and (115) that $Z_k(\omega_n)$, $\tilde{A}_k^\omega(\omega_n)$, $\Delta_k^{\mathrm{E}}(\omega_n)$, and $\Delta_k^{\mathrm{E}\star}(\omega_n)$ are almost independent on the space k belonging to large $\tilde{\varepsilon}_k$ because its contributions are suppressed by a factor $[\tilde{\varepsilon}_k(\omega_n)]^{-2}$. Thus, these quantities can be computed replacing the couplings with their value at $\tilde{\varepsilon}_k(\omega_n) = 0$.

With the integrand behaving as $[\tilde{\varepsilon}_k(\omega_n)]^{-1}$, the convergence of the Brillouin zone integrals in $\Sigma_k^\varepsilon(\omega_n)$ and $\Sigma_k^J(\omega_n)$ depend on the k dependence of the couplings in an essential way, even on k that correspond to a large $\tilde{\varepsilon}_k$. In particular, in absence of any k dependence of the couplings $\Sigma_k^\varepsilon(\omega_n)$ and $\Sigma_k^J(\omega_n)$ diverge logarithmically. From the physical point of view, $\Sigma_k^\varepsilon(\omega_n)$ shifts the position of the Fermi energy and $\Sigma_k^J(\omega_n)$ the magnetic splitting of quasiparticle states due to many-body interactions. These terms are zero if the system shows particle-hole symmetry and small in general (see also the discussion in Sec. III C). Therefore, we will discard these contributions completely and replace the couplings with their value at $\tilde{\varepsilon}_k(\omega_n) = 0$ entirely, reducing the computational costs significantly.

Another very effective simplification of the formalism comes from assuming the system to be isotropic in k. This means that the couplings will depend on k only via the quasiparticle energy $\varepsilon_{k\sigma}$. Here, we introduce the averaging operation on a generic function $F_{k\sigma}$ on equal center of energy and equal splitting surfaces according to

$$F_\sigma(\varepsilon,J) = \hat{I}_{k\sigma}(\varepsilon,J)F_{k\sigma} \tag{130}$$

$$= \frac{1}{\varrho(\varepsilon,J)}\sum_k \delta\left(\frac{\varepsilon_{\mathrm{sign}(\sigma)k\uparrow} + \varepsilon_{-\mathrm{sign}(\sigma)k\downarrow}}{2} - \varepsilon\right) \times \delta\left(\frac{\varepsilon_{\mathrm{sign}(\sigma)k\uparrow} - \varepsilon_{-\mathrm{sign}(\sigma)k\downarrow}}{2} - J\right)F_{k\sigma}, \tag{131}$$

where the number of states on center of energy and splitting surfaces is given by $\varrho(\varepsilon,J) = \hat{I}_{k\sigma}(\varepsilon,J)\,1$. The subscript indices "$k\sigma''$ on $\hat{I}_{k\sigma}(\varepsilon,J)$ indicate the variables that are averaged. Note that we invert the sign of $\boldsymbol{k}$ for the $\sigma = \downarrow$ part which makes $\hat{I}_{k\sigma}(\varepsilon,J)F_{k\sigma} = \hat{I}_{-k,-\sigma}(\varepsilon,J)F_{-k,-\sigma}$. Now, we define the analog of the Eliashberg function $\alpha^2F(\omega)$ [25,28]. We are going to keep the state dependence k for a little longer, and eventually take only those k such that $\tilde{\varepsilon}_k(\omega_n) = 0$. On the Nambu diagonal, it appears the coupling function

$$\alpha^2F^{\mathrm{D}}_\sigma(\varepsilon,J,\varepsilon',J',\Omega) = \varrho(\varepsilon',J')\hat{I}_{k'\sigma}(\varepsilon',J')\hat{I}_{k\sigma}(\varepsilon,J)\sum_q \left|g^q_{kk'\sigma}\right|^2\delta(\Omega - \Omega_q) \tag{132}$$

and on the Nambu off diagonal

$$\alpha^2F(\varepsilon,J,\varepsilon',J',\Omega) = \varrho(\varepsilon',J')\hat{I}_{k'\sigma}(\varepsilon',J')\hat{I}_{k\sigma}(\varepsilon,J) \times \sum_q g^q_{kk'\sigma}g^{-q}_{-k,-k',-\sigma}\delta(\Omega - \Omega_q), \tag{133}$$

$$C^{\mathrm{stat}}(\varepsilon,J,\varepsilon',J') = \varrho(\varepsilon',J')\hat{I}_{k'\sigma}(\varepsilon',J')\hat{I}_{k\sigma}(\varepsilon,J)W^{\mathrm{stat}}_{k'k,-k',-k_{\sigma,-\sigma}}. \tag{134}$$

Note that in the above equations (133) and (134), the left-hand side does not depend on σ because the averaging leads to the same result for $\sigma = \uparrow$ or $\downarrow$. The summation over k' and q in

the self-energy equations (108) to (115) are then transformed to integrals over ε', J', and Ω, respectively. However, if the couplings lose their center-of-energy dependence ε, the following functions only depend on the Matsubara frequency ω_n (that we now indicate as the index n) and the splitting: $Z_n(J)$, $\tilde{A}^{\omega}_n(J)$, $\Delta^{\mathrm{E}}_n(J)$, and $\Delta^{\mathrm{E}\star}_n(J)$. With $\tilde{\varepsilon}_k(\omega_n) \equiv \varepsilon/Z_n$ and $\tilde{J}_k(\omega_n) = J/Z_n$ we can compute analytically the integral over the center of energy ε of Eq. (128). Because the integrand decays faster than ε^{-1} for large ε, we may compute the integral

$$\mathfrak{M}_{n\sigma}(J) = \int d\varepsilon \sum_{\alpha} \frac{\alpha\big(\varepsilon - \mathrm{sign}(\sigma)\big(J + \tilde{A}^{\omega}_n Z_n\big)\big) - i\omega_n Z_n}{a_n(J)Z_n^2 - \frac{2\alpha}{\mathrm{sign}(\sigma)}\big(i\omega_n Z_n J + \tilde{A}^{\omega}_n Z_n \varepsilon\big) + \varepsilon^2} \tag{135}$$

as the sum of residues in the upper complex half-plane. Since it is not clear which of the four poles will be in the upper half, we compute all residues. Adding those, we obtain the energy integral in Eqs. (108) and (109) with

$$\mathfrak{S}_{n,\sigma}(J) = \sqrt{-\big[Z_n^2 \Delta^{\mathrm{E}}_n \Delta^{\mathrm{E}\star}_n - (i\omega_n Z_n - \mathrm{sign}(\sigma)J)^2\big]} \tag{136}$$

as

$$\begin{aligned}\mathfrak{M}_{n\sigma}(J) = {} & \pi i\left(\frac{i\omega_n Z_n - \mathrm{sign}(\sigma)J}{\mathfrak{S}_{n,\sigma}} - 1\right)\theta\left[\mathrm{Im}\left(-\frac{\tilde{A}^{\omega}_n Z_n}{\mathrm{sign}(\sigma)} - \mathfrak{S}_{n,\sigma}\right)\right] - \pi i\left(\frac{i\omega_n Z_n - \mathrm{sign}(\sigma)J}{\mathfrak{S}_{n,\sigma}} + 1\right) \\ & \times \theta\left[\mathrm{Im}\left(-\frac{\tilde{A}^{\omega}_n Z_n}{\mathrm{sign}(\sigma)} + \mathfrak{S}_{n,\sigma}\right)\right] + \pi i\left(\frac{i\omega_n Z_n + \mathrm{sign}(\sigma)J}{\mathfrak{S}_{n,-\sigma}} + 1\right)\theta\left[\mathrm{Im}\left(\frac{\tilde{A}^{\omega}_n Z_n}{\mathrm{sign}(\sigma)} - \mathfrak{S}_{n,-\sigma}\right)\right] \\ & - \pi i\left(\frac{i\omega_n Z_n + \mathrm{sign}(\sigma)J}{\mathfrak{S}_{n,-\sigma}} - 1\right)\theta\left[\mathrm{Im}\left(\frac{\tilde{A}^{\omega}_n Z_n}{\mathrm{sign}(\sigma)} + \mathfrak{S}_{n,-\sigma}\right)\right].\end{aligned} \tag{137}$$

Further, for Eqs. (114) and (115), we integrate Eq. (129) in center of energy ε. We define

$$\mathfrak{N}_n(J) = \sum_{\alpha} \int \frac{d\varepsilon}{a_n Z_n^2 - \alpha\big(i\omega_n Z_n J + \varepsilon \tilde{A}^{\omega}_n\big) + \varepsilon^2}, \tag{138}$$

that is evaluated to

$$\begin{aligned}\mathfrak{N}_n(J) = {} & \pi i\big(\mathfrak{S}^{-1}_{n,\uparrow}\theta\big[\mathrm{Im}\big(-\tilde{A}^{\omega}_n Z_n - \mathfrak{S}_{n,\uparrow}\big)\big] - \mathfrak{S}^{-1}_{n,\uparrow}\theta\big[\mathrm{Im}\big(-\tilde{A}^{\omega}_n Z_n + \mathfrak{S}_{n,\uparrow}\big)\big] \\ & + \mathfrak{S}^{-1}_{n,\downarrow}\theta\big[\mathrm{Im}\big(\tilde{A}^{\omega}_n Z_n - \mathfrak{S}_{n,\downarrow}\big)\big] - \mathfrak{S}^{-1}_{n,\downarrow}\theta\big[\mathrm{Im}\big(\tilde{A}^{\omega}_n Z_n + \mathfrak{S}_{n,\downarrow}\big)\big]\big).\end{aligned} \tag{139}$$

We obtain the Eliashberg equations similar to their usual, spin degenerate form [26,27,58], that only refer to the GF implicitly

$$Z_n(J) = 1 + \frac{\mathrm{i}}{4\omega_n}\int dJ' \frac{1}{\beta}\sum_{n'\sigma} \mathfrak{K}^{\sigma}_{n,n'}(J,J')\mathfrak{M}_{n'\sigma}(J'), \tag{140}$$

$$\tilde{A}^{\omega}_n(J) = \frac{1}{4Z_n(J)}\int dJ' \frac{1}{\beta}\sum_{n'\sigma} \frac{\mathfrak{K}^{\sigma}_{n,n'}(J,J')}{\mathrm{sign}(\sigma)}\mathfrak{M}_{n'\sigma}(J'), \tag{141}$$

$$\begin{aligned}\Delta^{\mathrm{E}}_n(J) = {} & -\frac{1}{2Z_n(J)}\int dJ' \frac{1}{\beta}\sum_{n'} \mathfrak{L}_{n,n'}(J,J') \\ & \times Z_{n'}(J')\Delta^{\mathrm{E}}_{n'}(J')\mathfrak{N}_{n'}(J'),\end{aligned} \tag{142}$$

$$\begin{aligned}\Delta^{\mathrm{E}\star}_n(J) = {} & -\frac{1}{2Z_n(J)}\int dJ' \frac{1}{\beta}\sum_{n'} \mathfrak{L}_{n,n'}(J,J') \\ & \times Z_{n'}(J')\Delta^{\mathrm{E}\star}_{n'}(J')\mathfrak{N}_{n'}(J'),\end{aligned} \tag{143}$$

where

$$\mathfrak{K}^{\sigma}_{n,n'}(J,J') = \int d\Omega \frac{2\Omega\,\alpha^2 F^{\mathrm{D}}_{\sigma}(0,J,0,J',\Omega)}{(\omega_n - \omega_{n'})^2 + \Omega^2}, \tag{144}$$

$$\begin{aligned}\mathfrak{L}_{n,n'}(J,J') = {} & \int d\Omega \frac{2\Omega\,\alpha^2 F(0,J,0,J',\Omega)}{(\omega_n - \omega_{n'})^2 + \Omega^2} \\ & + C^{\mathrm{stat}}(0,J,0,J').\end{aligned} \tag{145}$$

We point out that the Coulomb interaction is not well suited for the k-constant coupling approximation. The reason is that the function $\mathfrak{N}_n(J)$ behaves as $1/n$ for large n while $Z_n(J)$ goes to 1 and thus the Matsubara integral shows a logarithmic divergence due to $C^{\mathrm{stat}}(0,J,0,J')$ if $\Delta^{\mathrm{E}}_n(J)$ does not cut off the integral. Often, the effect of the Coulomb potential is mimicked by replacing C^{stat} with $\mu^{\star}\theta(\omega_c - |\omega_n|)$ where $\mu^{\star} = \frac{C^{\mathrm{stat}}}{1 + C^{\mathrm{stat}}\ln(\mathcal{E}/\omega_c)}$ with $\mathcal{E}$, a parameter of the electronic band structure and ω_c a phonon frequency cutoff [59,60]. Usually, the so-called Morel-Anderson pseudopotential $\mu^{\star}$ is fitted so that the calculated T_c matches the experimental one. $\mu^{\star}$ usually ranges between 0.1 and 0.16 for conventional SC [28]. The above equations imply that the coupling is isotropic in the sense that all states with equal center of energy and equal splitting share the same coupling matrix elements. Sometimes as in the well-known case of MgB_2 there are significant differences in the couplings and it is important to group states into bands for the isotropic approximation to hold. We refer to this case as the multiband approximation which simply means that all isotropic variables obtain another index for the band they correspond to.

Comparing the equations for the SC KS GF of Eq. (65) [noting $u^{k\alpha}_{k\sigma}(v^{-k\alpha}_{k-\sigma})^* = \alpha\,\mathrm{sign}(\sigma)\Delta^{\mathrm{s}}_{sk}/F_k$ where $F_k = \sqrt{\frac{\varepsilon_{k\uparrow} + \varepsilon_{-k\downarrow}}{2} + |\Delta^{\mathrm{s}}_{sk}|^2}$] with the interacting GF Eq. (125) we note that Δ^{s}_{sk} takes the role of $\Delta^{\mathrm{E}}_k(\omega_n)$ so the similar name is not accidental. However, as we have seen $\Delta^{\mathrm{E}}_k(\omega_n)$ takes its

significant shape in Matsubara space while Δ^{s}_{sk} does not have such a ω_n dependence and mimics the SC pairing in its k dependence in a way that densities of the interacting system are reproduced.

V. SUMMARY AND CONCLUSION

In this work, we have developed fully *ab initio* methods to compute the SC phase of a material in a magnetic field Zeeman coupled to the spin magnetization. In a unified notation we present a purely GF based (the Eliashberg approach) and a density functional based scheme.

In our DFT we have employed a SC KS system to reproduce the interacting densities $n(\boldsymbol{r})$, $\boldsymbol{m}(\boldsymbol{r})$, $\boldsymbol{\chi}(\boldsymbol{r},\boldsymbol{r}')$, and $\Gamma(\boldsymbol{R}_1 \ldots \boldsymbol{R}_N)$. The SC KS system can be solved analytically using the SDA where we only consider the singlet pairing of time-reversed basis states. We have derived xc potentials in this case that include the electron-nuclear interaction on the level of KS phonons and treat the Coulomb interaction in the same footing without the need for any adjustable parameter.

As a second step, we have applied similar approximations to the Dyson equation starting from the SC KS system as a formally noninteracting system. This procedure leads to the Eliashberg equations of a SC in a magnetic field similar to those discussed in Ref. [24].

While SpinSCDFT allows to include the full Coulomb potential and promises numerically efficient calculations for real materials, the direct GF approach is, instead, valuable to get direct physical insights to develop approximations and further improve the SpinSCDFT scheme.

The theoretical framework presented in this work allows to compute the phenomenon of coexistence and competition of SC with magnetism from first principles.

In the subsequent Paper II [57], we will discuss a detailed numerical implementation of the equations presented in this work, i.e., the linear and nonlinear functionals and the Eliashberg equations without Coulomb interactions. Further, we will introduce a G_0W_0 scheme to obtain the excitation spectrum starting from a SpinSCDFT calculation.

APPENDIX: FORMULAS FOR THE MATSUBARA SUMS

In the potential terms, it appears the Matsubara summation

$$P_s(E,E') = \frac{1}{\beta}\sum_n \frac{1}{(i\omega_n - E)(i\omega_n - E')}. \tag{A1}$$

This is analytically evaluated with the result

$$P_s(E,E') = \frac{f_\beta(E) - f_\beta(E')}{E - E'}, \tag{A2}$$

$$\lim_{E'\to E} P_s(E,E') = \partial_E f_\beta(E) = -\beta f_\beta(E) f_\beta(-E), \tag{A3}$$

where the symmetries $P_s(E,E') = P_s(-E,-E')$ and $P_s(E,E') = P_s(E',E)$ hold. The Matsubara frequency summation

$$I(\Omega,E_1,E_2,E_3) = \frac{1}{\beta^2}\sum_{nn'} \frac{1}{i\omega_n - E_1}\frac{1}{i(\omega_n - \omega_{n'}) - \Omega} \times \frac{1}{i\omega_{n'} - E_2}\frac{1}{i\omega_n - E_3}, \tag{A4}$$

$$L(\Omega,E_1,E_2,E_3) = I(-\Omega,E_1,E_2,E_3) - I(\Omega,E_1,E_2,E_3) \tag{A5}$$

is also in principle straightforward. However, the resulting formulas are rather large and computer algebra becomes essential for the evaluation of residues and limiting behaviors, necessary for a numerical implementation. Note that a partial summation leads to

$$L(\Omega,E_1,E_2,E_3) = \frac{1}{\beta}\sum_n \frac{M_{\mathrm{ph}}(\Omega,E_2,\omega_n)}{(i\omega_n - E_1)(i\omega_n - E_3)}. \tag{A6}$$

From the definition we observe the following symmetry relations:

$$L(\Omega,E_1,E_2,E_3) = L(\Omega,E_3,E_2,E_1), \tag{A7}$$

$$L(-\Omega,E_1,E_2,E_3) = -L(\Omega,E_3,E_2,E_1), \tag{A8}$$

$$[L(\Omega,E_1,E_2,E_3)]^* = -L(\Omega,-E_1,-E_2,-E_3). \tag{A9}$$

Evaluation of the Coulomb diagram requires the following summation:

$$L_{\mathrm{C}}(E_1,E_2,E_3) = \frac{1}{\beta^2}\sum_{nn'} \frac{1}{i\omega_{n'} - E_2}\frac{1}{i\omega_n - E_1}\frac{1}{i\omega_n - E_3} \tag{A10}$$

$$= f_\beta(E_2) P_s(E_1,E_3). \tag{A11}$$

Using *Mathematica*, we evaluate the sums Eqs. (A4) and (A5) to

$$L(\Omega,E_1,E_2,E_3) = \left(\frac{f_\beta(E_2)n_\beta(\Omega)}{(E_2 - E_1 + \Omega)(E_2 - E_3 + \Omega)} + \frac{f_\beta(E_2)[1 + n_\beta(\Omega)]}{(E_1 - E_2 + \Omega)(E_3 - E_2 + \Omega)} + \frac{f_\beta(E_1)[1 - f_\beta(E_2) + n_\beta(\Omega)]}{(E_1 - E_3)(E_1 - E_2 - \Omega)} + \frac{f_\beta(E_3)[1 - f_\beta(E_2) + n_\beta(\Omega)]}{(E_1 - E_3)(E_2 - E_3 + \Omega)} + \frac{f_\beta(E_1)[f_\beta(E_2) + n_\beta(\Omega)]}{(E_1 - E_3)(E_1 - E_2 + \Omega)} + \frac{f_\beta(E_3)[f_\beta(E_2) + n_\beta(\Omega)]}{(E_3 - E_1)(E_3 - E_2 + \Omega)}\right). \tag{A12}$$

Clearly, some points, e.g., $E_1 = E_3$ are numerically problematic, so whenever $E_1 \approx E_3$ we may have to evaluate the limiting formula instead. In general, the various limits where the denominators are zero all exist and can be computed explicitly, again

using *Mathematica*. The results are

$$\lim_{E_1\to E_3} L(\Omega,E_1,E_2,E_3) = f_\beta(E_2)\left(\frac{n_\beta(\Omega)}{(E_2-E_3+\Omega)^2}+\frac{1+n_\beta(\Omega)}{(E_2-E_3-\Omega)^2}\right) - f_\beta(E_3)\left(\frac{f_\beta(-E_2)+n_\beta(\Omega)}{(E_2-E_3+\Omega)^2}\frac{f_\beta(E_2)+n_\beta(\Omega)}{(E_2-E_3-\Omega)^2} - \frac{\beta f_\beta(-E_3)}{(E_2-E_3)^2-(\Omega)^2}([f_\beta(E_2)-f_\beta(-E_2)]\Omega+[2n_\beta(\Omega)+1](E_2-E_3))\right), \tag{A13}$$

$$\lim_{\Omega\to E_3-E_2}\lim_{E_1\to E_3} L(\Omega,E_1,E_2,E_3) = \beta\frac{[1+f_\beta(E_2)+n_\beta(E_3-E_2)]f_\beta(-E_3)f_\beta(E_3)}{2(E_2-E_3)} + \frac{f_\beta(E_2)+f_\beta(E_3)[1-2f_\beta(E_2)]}{4(E_2-E_3)^2} + \beta^2 f_\beta(-E_2)[2+n_\beta(E_3-E_2)]f_\beta(E_3)\left(\frac{1}{2}-f_\beta(E_3)\right), \tag{A14}$$

$$\lim_{\Omega\to E_1-E_2} L(\Omega,E_1,E_2,E_3) = \frac{f_\beta(E_1)[f_\beta(E_2)+n_\beta(E_1-E_2)]}{2(E_1-E_2)(E_1-E_3)} + \frac{f_\beta(E_2)[1+n_\beta(E_1-E_2)]}{2(E_1-E_2)(E_1-2E_2+E_3)} + \frac{f_\beta(E_3)[n_\beta(E_1-E_2)+f_\beta(-E_2)]-f_\beta(E_2)n_\beta(E_1-E_2)}{(E_1-E_3)^2} + \frac{f_\beta(E_3)[f_\beta(E_2)+n_\beta(E_1-E_2)]}{2(E_3-E_1)(E_1-2E_2+E_3)} + \beta\frac{f_\beta(-E_1)f_\beta(E_2)n_\beta(E_1-E_2)}{E_3-E_1}, \tag{A15}$$

$$\lim_{E_1\to 2E_2-E_3}\lim_{\Omega\to E_1-E_2} L(\Omega,E_1,E_2,E_3) = \frac{f_\beta(E_2)[1+n_\beta(E_2-E_3)]}{2(E_2-E_3)}\left(\beta f_\beta(-E_3)-\frac{1}{2(E_2-E_3)}\right). \tag{A16}$$

We point out here that the limit $\Omega_{q\lambda}\to 0$ does not exist. It is, however, unimportant as the $g_{ij}^{\lambda q}$ go to zero in the limit $\Omega\to 0$ faster than L diverges.

[1] A. Abrikosov, J. Phys. Chem. Solids **2**, 199 (1957).
[2] R. O. Walther Meissner, Naturwissenschaften **21**, 787 (1933).
[3] D. K. Satapathy, M. A. Uribe-Laverde, I. Marozau, V. K. Malik, S. Das, T. Wagner, C. Marcelot, J. Stahn, S. Brück, A. Rühm, S. Macke, T. Tietze, E. Goering, A. Frañó, J. H. Kim, M. Wu, E. Benckiser, B. Keimer, A. Devishvili, B. P. Toperverg, M. Merz, P. Nagel, S. Schuppler, and C. Bernhard, Phys. Rev. Lett. **108**, 197201 (2012).
[4] A. I. Buzdin, Rev. Mod. Phys. **77**, 935 (2005).
[5] P. Fulde and R. A. Ferrell, Phys. Rev. **135**, A550 (1964).
[6] A. I. Larkin and Yu. N. Ovchinnikov, Zh. Eksp. Teor. Fiz. **47**, 1136 (1964) [Sov. Phys. JETP **20**, 762 (1965)].
[7] A. Bianchi, R. Movshovich, C. Capan, P. G. Pagliuso, and J. L. Sarrao, Phys. Rev. Lett. **91**, 187004 (2003).
[8] R. Lortz, Y. Wang, A. Demuer, P. H. M. Böttger, B. Bergk, G. Zwicknagl, Y. Nakazawa, and J. Wosnitza, Phys. Rev. Lett. **99**, 187002 (2007).
[9] M. Sigrist and K. Ueda, Rev. Mod. Phys. **63**, 239 (1991).
[10] D. Jérome, A. Mazaud, M. Ribault, and K. Bechgaard, J. Phys. Lett. **41**, L95 (1980).
[11] T. M. Rice and M. Sigrist, J. Phys.: Condens. Matter **7**, L643 (1995).
[12] G. M. Luke, Y. Fudamoto, K. M. Kojima, M. I. Larkin, J. Merrin, B. Nachumi, Y. J. Uemura, Y. Maeno, Z. Q. Mao, Y. Mori, H. Nakamura, and M. Sigrist, Nature (London) **394**, 558 (1998).
[13] K. Ishida, H. Mukuda, Y. Kitaoka, K. Asayama, Z. Q. Mao, Y. Mori, and Y. Maeno, Nature (London) **396**, 658 (1998).
[14] S. S. Saxena, P. Agarwal, K. Ahilan, F. M. Grosche, R. K. W. Haselwimmer, M. J. Steiner, E. Pugh, I. R. Walker, S. R. Julian, P. Monthoux, G. G. Lonzarich, A. Huxley, I. Sheikin, D. Braithwaite, and J. Flouquet, Nature (London) **406**, 587 (2000).
[15] E. Bauer, G. Hilscher, H. Michor, C. Paul, E. W. Scheidt, A. Gribanov, Y. Seropegin, H. Noël, M. Sigrist, and P. Rogl, Phys. Rev. Lett. **92**, 027003 (2004).
[16] A. Abrikosov and L. Gor'kov, Zh. Eksp. Teor. Fiz. **39**, 1781 (1960) [Sov. Phys. JETP **12**, 1243 (1961)].
[17] I. Felner, U. Asaf, Y. Levi, and O. Millo, Phys. Rev. B **55**, R3374 (1997).
[18] D. A. Dikin, M. Mehta, C. W. Bark, C. M. Folkman, C. B. Eom, and V. Chandrasekhar, Phys. Rev. Lett. **107**, 056802 (2011).
[19] D. Aoki, A. Huxley, E. Ressouche, D. Braithwaite, J. Flouquet, J.-P. Brison, E. Lhotel, and C. Paulsen, Nature (London) **413**, 613 (2001).
[20] D. Manske, *Theory of Unconventional Superconductors: Cooper-Pairing Mediated by Spin Excitations*, Physics and Astronomy Online Library No. 202 (Springer, Berlin, 2004).
[21] P. A. Lee, N. Nagaosa, and X.-G. Wen, Rev. Mod. Phys. **78**, 17 (2006).
[22] P. Hohenberg and W. Kohn, Phys. Rev. **136**, B864 (1964).
[23] L. J. Sham and M. Schlüter, Phys. Rev. Lett. **51**, 1888 (1983).
[24] S. Vonsovsky, Y. Izyumov, E. Kurmaev, E. Brandt, and A. Zavarnitsyn, *Superconductivity of Transition Metals: Their Alloys and Compounds*, Springer Series in Solid-State Sciences Series (Springer, London, 1982).
[25] P. B. Allen and B. Mitrović, in *Theory of Superconducting T_c*, edited by F. S. Henry Ehrenreich and D. Turnbull, Solid State Physics (Academic, New York, 1983), Vol. 37, pp. 1–92.
[26] E. R. Margine and F. Giustino, Phys. Rev. B **87**, 024505 (2013).

[27] A. Sanna, S. Pittalis, J. K. Dewhurst, M. Monni, S. Sharma, G. Ummarino, S. Massidda, and E. K. U. Gross, Phys. Rev. B **85**, 184514 (2012).
[28] J. P. Carbotte, Rev. Mod. Phys. **62**, 1027 (1990).
[29] G. M. Eliashberg, Zh. Eksp. Teor. Fiz. **38**, 966 (1960) [Sov. Phys. JETP **11**, 696 (1960)].
[30] W. Kohn and L. J. Sham, Phys. Rev. **140**, A1133 (1965).
[31] E. Runge and E. K. U. Gross, Phys. Rev. Lett. **52**, 997 (1984).
[32] T. Kreibich and E. K. U. Gross, Phys. Rev. Lett. **86**, 2984 (2001).
[33] M. Lüders, M. A. L. Marques, N. N. Lathiotakis, A. Floris, G. Profeta, L. Fast, A. Continenza, S. Massidda, and E. K. U. Gross, Phys. Rev. B **72**, 024545 (2005).
[34] N. D. Mermin, Phys. Rev. **137**, A1441 (1965).
[35] M. A. L. Marques, M. Lüders, N. N. Lathiotakis, G. Profeta, A. Floris, L. Fast, A. Continenza, E. K. U. Gross, and S. Massidda, Phys. Rev. B **72**, 024546 (2005).
[36] R. van Leeuwen, Phys. Rev. B **69**, 115110 (2004).
[37] S. Baroni, S. de Gironcoli, A. Dal Corso, and P. Giannozzi, Rev. Mod. Phys. **73**, 515 (2001).
[38] P. Giannozzi, S. de Gironcoli, P. Pavone, and S. Baroni, Phys. Rev. B **43**, 7231 (1991).
[39] W. Nolting, *Grundkurs Theoretische Physik 7: Viel-Teilchen Theorie*, Grundkurs Theoretische Physik (Springer, Berlin, 2005).
[40] Y. Nambu, Phys. Rev. **117**, 648 (1960).
[41] P. W. Anderson, Phys. Rev. **112**, 1900 (1958).
[42] J. Valatin, Nuovo Cimento **7**, 843 (1958).
[43] N. N. Bogoliubov, Zh. Eksp. Teor. Fiz. **34**, 58 (1958) [Sov. Phys. JETP **34**, 51 (1958)].
[44] L. N. Oliveira, E. K. U. Gross, and W. Kohn, Phys. Rev. Lett. **60**, 2430 (1988).
[45] U. von Barth and L. Hedin, J. Phys. C: Solid State Phys. **5**, 1629 (1972).
[46] P. Anderson, J. Phys. Chem. Solids **11**, 26 (1959).
[47] G. Sarma, J. Phys. Chem. Solids **24**, 1029 (1963).
[48] M. Marques, Ph.D. thesis, University of Würzburg, 2000.
[49] A. Sanna and E. K. U. Gross (unpublished).
[50] A. Linscheid and F. Essenberger, arXiv:1503.00970.
[51] F. Essenberger, A. Sanna, A. Linscheid, F. Tandetzky, G. Profeta, P. Cudazzo, and E. K. U. Gross, Phys. Rev. B **90**, 214504 (2014).
[52] A. B. Migdal, Zh. Eksp. Teor. Fiz. **34**, 1438 (1958) [Sov. Phys. JETP **7**, 996 (1958)].
[53] A. Marini, G. Onida, and R. Del Sole, Phys. Rev. Lett. **88**, 016403 (2001).
[54] A. Linscheid, Ph.D. thesis, Martin-Luther-Universität Halle-Wittenberg, 2014.
[55] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. Dal Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussis, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Sbraccia, S. Scandolo, G. Sclauzero, A. P. Seitsonen, A. Smogunov, P. Umari, and R. M. Wentzcovitch, J. Phys.: Condens. Matter **21**, 395502 (2009).
[56] K. Dewhurst, S. Sharma, L. Nordström, F. Cricchio, F. Bultmark, O. Granäs, H. Gross, C. Ambrosch-Draxl, C. Persson, C. Brouder, R. Armiento, A. Chizmeshya, P. Anderson, I. Nekrasov, F. Wagner, F. Kalarasse, J. Spitaler, S. Pittalis, N. Lathiotakis, T. Burnus, S. Sagmeister, C. Meisenbichler, S. Lebègue, Y. Zhang, F. Körmann, A. Baranov, A. Kozhevnikov, S. Suehara, F. Essenberger, A. Sanna, T. McQueen, T. Baldsiefen, M. Blaber, A. Filanovich, T. Björkman, M. Stankovski, J. Goraus, M. Meinert, D. Rohr, V. Nazarov, K. Krieger, P. Floyd, A. Davydov, F. Eich, A. R. Castro, K. Kitahara, J. Glasbrenner, K. Bussmann, I. Mazin, M. Verstraete, D. Ernsting, S. Dugdale, and P. Elliott, http://elk.sourceforge.net
[57] A. Linscheid, A. Sanna, and E. K. U. Gross, Phys. Rev. B **92**, 024506 (2015).
[58] A. Sanna, Ph.D. thesis, Università degli Studi di Cagliari, 2007.
[59] P. Morel and P. W. Anderson, Phys. Rev. **125**, 1263 (1962).
[60] D. J. Scalapino, J. R. Schrieffer, and J. W. Wilkins, Phys. Rev. **148**, 263 (1966).
[61] M. Schossmann and E. Schachinger, Phys. Rev. B **33**, 6123 (1986).