Local Density Approximation for Superconductors

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A universal LDA-type density functional describing the electronic correlations in superconductors is developed from first principles. The functional is constructed from the exchange-correlation free-energy density, $f_{\rm xc}^{\rm hom}$, of a homogeneous electron gas exposed to an external translationally invariant pairing field. The quantity $f_{\rm xc}^{\rm hom}$, which is a function of the density and a functional of the induced order parameter, is calculated by many-body perturbation theory.

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Density functional theory (DFT) [1] is a powerful tool in electronic-structure calculations of atoms, molecules, and solids. Conventional DFT, however, is not able to describe the superconducting phase of matter. In 1988, triggered by the discovery of the high-temperature superconductors, Oliveira, Gross, and Kohn [2] developed the formal framework of a DFT for superconductors. In this formalism the exchange-correlation (xc) energy is a functional of two quantities, the ordinary density, $n(\mathbf{r}) = \sum_\sigma \langle \hat{\psi}_\sigma^\dagger(\mathbf{r}) \hat{\psi}_\sigma(\mathbf{r}) \rangle$, and the superconducting order parameter $\chi(\mathbf{r}, \mathbf{r}') = \langle \hat{\psi}_\uparrow(\mathbf{r}) \hat{\psi}_\downarrow(\mathbf{r}') \rangle$. The corresponding Kohn-Sham (KS) equations have the form of the Bogoliubov–de Gennes equations (atomic units are used throughout)

$$\left(-\frac{\nabla^2}{2} + v_s(\mathbf{r}) - \mu\right) u_k(\mathbf{r}) + \int d^3r' \, \Delta_s(\mathbf{r}, \mathbf{r}') \, v_k(\mathbf{r}') = E_k u_k(\mathbf{r}) , \tag{1}$$

$$-\left(-\frac{\nabla^2}{2} + v_s(\mathbf{r}) - \mu\right) v_k(\mathbf{r}) + \int d^3r' \, \Delta_s^*(\mathbf{r}, \mathbf{r}') \, u_k(\mathbf{r}') = E_k v_k(\mathbf{r}) , \tag{2}$$

where μ is the chemical potential of the superconductor, and the effective electrostatic and pairing potentials, $v_s(\mathbf{r})$ and $\Delta_s(\mathbf{r}, \mathbf{r}')$, are given by

$$v_s[n, \chi](\mathbf{r}) = v_0(\mathbf{r}) + \int d^3r' \, \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + v_{\rm xc}[n, \chi](\mathbf{r}) , \tag{3}$$

$$\Delta_s[n, \chi](\mathbf{r}, \mathbf{r}') = \Delta_0(\mathbf{r}, \mathbf{r}') + \frac{\chi(\mathbf{r}, \mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} + \Delta_{\rm xc}[n, \chi](\mathbf{r}, \mathbf{r}') . \tag{4}$$

v_0 represents the Coulomb potential of the lattice and Δ_0 is an external pairing potential produced, e.g., by the proximity effect of an adjacent superconductor. The xc potentials are formally defined as functional derivatives of the xc-free-energy functional $F_{\rm xc}[n, \chi]$:

$$v_{\rm xc}[n, \chi](\mathbf{r}) = \frac{\delta F_{\rm xc}[n, \chi]}{\delta n(\mathbf{r})} , \tag{5}$$

$$\Delta_{\rm xc}[n, \chi](\mathbf{r}, \mathbf{r}') = -\frac{\delta F_{\rm xc}[n, \chi]}{\delta \chi^*(\mathbf{r}, \mathbf{r}')} . \tag{6}$$

The first numerical solution of these KS equations was achieved in 1993 for niobium [3]. Recently, the first attempts to tackle the high-T_c superconductors within the above DFT framework have appeared [4,5]. In this work, the xc functional was modeled by a phenomenological interaction kernel which was expanded in the linear muffin-tin orbitals of a recently proposed eight-band model for YBCO [6,7]. The comparison of various scenarios pointed to the conclusion that the pairing mechanism operates between electrons of opposite spins on nearest-neighbor Cu sites.

While it is certainly fruitful to study the xc potential of a particular system, the charm and the power of DFT derives from the *universality* of the xc functional: One and the same functional of n and χ should predict the specific properties of *all* materials. The present Letter represents the first attempt to construct such a universal functional for superconductors. The proposed functional can be viewed as the superconducting analog of the local spin-density approximation (LSDA). To explain the nature of our construction we first take a step back and briefly review the basic idea behind the LSDA as it is commonly used in the calculation of magnetic properties: To construct the LSDA, the homogeneous electron gas is exposed to a constant magnetic field (in z direction) which produces a finite spin polarization m. The corresponding xc energy per unit volume then becomes a function $e_{\rm xc}^{\rm hom}(n, m)$ of the density n and the spin polarization m. Once this function is known, the LSDA functional for inhomogeneous systems with density $n(\mathbf{r})$ and magnetization $m(\mathbf{r})$ is defined by $E_{\rm xc}^{\rm LSDA}[n, m] = \int d^3r \, e_{\rm xc}^{\rm hom}(n(\mathbf{r}), m(\mathbf{r}))$. It is well known that this functional provides a rather successful description of magnetic properties. The functional itself is universal, i.e., its dependence on n and m is the same for all systems. The fact that the homogeneous electron gas (without external magnetic fields) becomes spin polarized only at unphysically low densities is not relevant. What is used in the

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LSDA is the function $e_{\rm xc}^{\rm hom}(n,m)$ produced by *finite* external magnetic fields.

We apply the same philosophy to superconductors: The homogeneous electron gas is exposed to an external pairing field Δ_0 which induces a finite order parameter χ. Consequently, the xc energy depends on the density n and on χ. To preserve translational invariance, the external pairing field $\Delta_0(\mathbf{r},\mathbf{r}')$ is chosen to depend on $(\mathbf{r}-\mathbf{r}')$ only, so that its Fourier transform is given by $\Delta_0(\mathbf{k}) = \int d^3(r-r')e^{i\mathbf{k}(\mathbf{r}-\mathbf{r}')}\Delta_0(\mathbf{r},\mathbf{r}')$. As a consequence, the induced order parameter $\chi(\mathbf{r},\mathbf{r}')$ is translationally invariant as well; its Fourier transform will be denoted by $\chi(\mathbf{k})$. Considering a homogeneous superconductor at finite temperature, we have to determine the xc free energy per unit volume, $f_{\rm xc}^{\rm hom}[n,\chi(\mathbf{k})]$, which is a *function* of the (constant) density n and a *functional* of $\chi(\mathbf{k})$. In analogy to the LSDA functional discussed above, we then define the LDA for superconductors by

$$F_{\rm xc}^{\rm LDA}[n(\mathbf{R}),\chi_W(\mathbf{R},\mathbf{k})] = \int d^3R\, f_{\rm xc}^{\rm hom}[n,\chi]\Big|_{\substack{n=n(\mathbf{R})\\ \chi=\chi_W(\mathbf{R},\mathbf{k})}}, \tag{7}$$

where $\chi_W(\mathbf{R},\mathbf{k})$ is the Wigner transform

$$\chi_W(\mathbf{R},\mathbf{k}) = \int d^3s\, e^{i\mathbf{k}\mathbf{s}}\chi\left(\mathbf{R}+\frac{\mathbf{s}}{2},\mathbf{R}-\frac{\mathbf{s}}{2}\right) \tag{8}$$

of the anomalous density $\chi(\mathbf{r},\mathbf{r}')$ of the inhomogeneous systems to be treated. It is obvious that this definition correctly reduces to the LDA of nonsuperconducting systems in the limit $\chi \to 0$. At first sight, other definitions of an LDA for superconductors with the correct nonsuperconducting limit might be conceivable. However, a systematic gradient expansion of the total-energy functional shows that Eq. (7) is in fact *the only* correct LDA for superconductors. This follows from a diagrammatic expansion of the total energy of the *inhomogeneous* system in terms of the normal and anomalous Green's functions and the particle-particle interaction. A subsequent $\hbar$ expansion of the Green's functions [8] results in a semiclassical expansion of the total energy. From this expansion one readily concludes [9] that the lowest order terms in $\hbar$ are identical with the LDA, leading to Eq. (7).

The LDA requires $f_{\rm xc}^{\rm hom}$ as an input. To determine this functional we have to calculate the free energy associated with the Hamiltonian $\hat{H} = \hat{H}_0 + \hat{U}$, where

$$\hat{H}_0 = \sum_\sigma \sum_{\mathbf{k}} (k^2/2 - \mu_0)\hat{c}_{\mathbf{k}\sigma}^\dagger \hat{c}_{\mathbf{k}\sigma} - \sum_{\mathbf{k}} \left[\Delta_0^*(\mathbf{k})\hat{c}_{\mathbf{k}\uparrow}\hat{c}_{-\mathbf{k}\downarrow} + \text{H.c.}\right] \tag{9}$$

describes a noninteracting gas exposed to the external pairing field $\Delta_0(\mathbf{k})$. μ_0 is a shorthand for the constant $(\mu - v_0)$ and $\hat{U}$ represents the bare Coulomb interaction

$$\hat{U} = \frac{1}{2}\sum_{\sigma,\sigma'}\int d^3r \int d^3r'\, \hat{\psi}_\sigma^\dagger(\mathbf{r})\hat{\psi}_{\sigma'}^\dagger(\mathbf{r}')\frac{1}{|\mathbf{r}-\mathbf{r}'|} \times \hat{\psi}_{\sigma'}(\mathbf{r}')\hat{\psi}_\sigma(\mathbf{r}). \tag{10}$$

In standard many-body perturbation theory one would take $\hat{H}_0$ as the unperturbed Hamiltonian and $\hat{U}$ as the perturbation. For reasons explained below we choose, for our diagrammatic analysis, the Kohn-Sham Hamiltonian

$$\hat{H}_s = \sum_\sigma \sum_{\mathbf{k}} (k^2/2 - \mu_s)\hat{c}_{\mathbf{k}\sigma}^\dagger \hat{c}_{\mathbf{k}\sigma} - \sum_{\mathbf{k}} \left[\Delta_s^*(\mathbf{k})\hat{c}_{\mathbf{k}\uparrow}\hat{c}_{-\mathbf{k}\downarrow} + \text{H.c.}\right], \tag{11}$$

as the "unperturbed" Hamiltonian. μ_s is a shorthand for the constant $(\mu - v_s)$ and $\Delta_s(\mathbf{k})$ is the KS pairing potential (4) of a homogeneous system. The full Hamiltonian can then be written as $\hat{H} = \hat{H}_s + \hat{H}_1$ with the perturbation $\hat{H}_1 = \hat{U} - \hat{H}_s + \hat{H}_0$. Because of the presence of the pairing field $\Delta_s(\mathbf{k})$ in the unperturbed Hamiltonian $\hat{H}_s$, the diagrammatic analysis not only involves the normal KS Green's function G_s but also the anomalous KS propagators F_s and $F_s^\dagger$. The latter are represented by lines with arrows pointing in opposite directions. The three diagrams contributing to the free energy to first order in $\hat{H}_1$ are shown in Fig. 1. However, only the third diagram, Fig. 1c, contributes to the xc free energy $F_{\rm xc}$. Figure 1a corresponds to the classical electrostatic energy of the charge distribution. This energy contribution is not approximated within the LDA but rather treated exactly, leading to the Hartree potential in Eq. (3). Likewise, the "anomalous Hartree energy" depicted in Fig. 1b is not included in $F_{\rm xc}$. This term leads to the anomalous Hartree potential, the second term on the right-hand side of Eq. (4). The only contribution to the xc free energy per unit volume in first order is the exchange diagram of Fig. 1c yielding

$$\tilde{f}_x^{\rm hom}[\mu_s,\Delta_s] = -\frac{1}{4}\int\frac{d^3k}{(2\pi)^3}\frac{d^3k'}{(2\pi)^3}\left[1-\frac{\varepsilon_{\mathbf{k}}-\mu_s}{R_{\mathbf{k}}}\tanh\left(\frac{\beta}{2}R_{\mathbf{k}}\right)\right]\frac{4\pi}{|\mathbf{k}-\mathbf{k}'|^2}\left[1-\frac{\varepsilon_{\mathbf{k}'}-\mu_s}{R_{\mathbf{k}'}}\tanh\left(\frac{\beta}{2}R_{\mathbf{k}'}\right)\right], \tag{12}$$

where $R_{\mathbf{k}} = \sqrt{(\varepsilon_{\mathbf{k}}-\mu_s)^2 + |\Delta_s(\mathbf{k})|^2}$ and $\varepsilon_{\mathbf{k}} = k^2/2$. The wiggly lines in Figs. 1a–1c represent the bare Coulomb interaction $\hat{U}$. The first-order contributions resulting from the one-body operators in $\hat{H}_1$ cancel with terms in the free energy of the unperturbed system described by the Hamiltonian $\hat{H}_s$.

In second order, as in the normal electron gas, some of the diagrams are divergent due to the long range of the

Coulomb interaction. To obtain a finite result a partial resummation of an infinite subset of diagrams needs to be performed. The simplest resummation of this kind is the random phase approximation (RPA) [10], which includes the most divergent terms in every order. The RPA for superconductors includes all the normal and anomalous bubble diagrams as indicated in Fig. 2. The resummation leads to

$$\tilde{f}_c^{\mathrm{RPA}}[\mu_s, \Delta_s] = \frac{1}{2\beta} \sum_{\nu_n} \int \frac{d^3q}{(2\pi)^3} \left\{ \log\left[1 - \Pi_s(\mathbf{q}, \nu_n) \frac{4\pi}{q^2} \right] + \Pi_s(\mathbf{q}, \nu_n) \frac{4\pi}{q^2} \right\} \tag{13}$$

with the even Matsubara frequencies $\nu_n = 2n\pi/\beta$. Π_s is the irreducible KS polarization given by

$$\Pi_s(\mathbf{q}, \nu_n) = \frac{2}{\beta} \sum_{\omega_n} \int \frac{d^3k}{(2\pi)^3} \{ G_s(\mathbf{k}, \omega_n) G_s(\mathbf{k} + \mathbf{q}, \omega_n + \nu_n) + F_s(\mathbf{k}, \omega_n) F_s^{\dagger}(\mathbf{k} + \mathbf{q}, \omega_n + \nu_n) \} \tag{14}$$

with the odd Matsubara frequencies $\omega_n = (2n + 1)\pi/\beta$.

Equations (12) and (13) represent the xc free energy as a functional $\tilde{f}_{\mathrm{xc}}^{\mathrm{hom}}[\mu_s, \Delta_s(\mathbf{k})]$ of the potentials μ_s and $\Delta_s(\mathbf{k})$ appearing in the KS Hamiltonian (11). The DFT for superconductors, however, requires the xc energy as a functional of the densities $\{n, \chi(\mathbf{k})\}$. By virtue of the Hohenberg-Kohn theorem for superconductors [2], applied to the noninteracting case, the potentials $\{\mu_s, \Delta_s(\mathbf{k})\}$ and the densities $\{n, \chi(\mathbf{k})\}$ are in 1-1 correspondence, i.e., μ_s and $\Delta_s(\mathbf{k})$ can be written as functionals of n and $\chi(\mathbf{k})$,

$$\mu_s = \mu_s[n, \chi(\mathbf{k})], \qquad \Delta_s(\mathbf{q}) = \Delta_s[n, \chi(\mathbf{k})](\mathbf{q}), \tag{15}$$

so that the desired density functional $f_{\mathrm{xc}}^{\mathrm{hom}}[n, \chi(\mathbf{k})]$ is obtained from

$$f_{\mathrm{xc}}^{\mathrm{hom}}[n, \chi(\mathbf{k})] = \tilde{f}_{\mathrm{xc}}^{\mathrm{hom}}[\mu_s[n, \chi(\mathbf{k})], \Delta_s[n, \chi(\mathbf{k})]]. \tag{16}$$

The functionals (15) corresponding to noninteracting systems can be constructed explicitly by inverting the well-known relations

$$n = \int \frac{d^3k}{(2\pi)^3} \left[1 - \frac{\varepsilon_{\mathbf{k}} - \mu_s}{R_{\mathbf{k}}} \tanh\left(\frac{\beta}{2} R_{\mathbf{k}} \right) \right], \tag{17}$$

$$\chi(\mathbf{k}) = \frac{1}{2} \frac{\Delta_s(\mathbf{k})}{R_{\mathbf{k}}} \tanh\left(\frac{\beta}{2} R_{\mathbf{k}} \right). \tag{18}$$

Given some densities $\{\bar{n}, \bar{\chi}(\mathbf{k})\}$, the corresponding potentials $\bar{\mu}_s = \mu_s[\bar{n}, \bar{\chi}(\mathbf{k})]$ and $\bar{\Delta}_s = \Delta_s[\bar{n}, \bar{\chi}(\mathbf{k})]$ are determined by the following steps: (a) for fixed β, Eq. (18) is inverted, leading to $\Delta_s(\mathbf{k}) = D(\mu_s, \bar{\chi}(\mathbf{k}))$. At zero temperature, the inverse function D is easily constructed analytically, while at finite temperature, D has to be evaluated numerically from Eq. (18). (b) Insert the result of step (a) in Eq. (17), yielding the density as a function $n(\mu_s)$. (c) Find $\bar{\mu}_s$ such that $\bar{n} = n(\bar{\mu}_s)$. (d) Insert this $\bar{\mu}_s$ in the inverse function D to get $\bar{\Delta}_s(\mathbf{k}) = D(\bar{\mu}_s, \bar{\chi}(\mathbf{k}))$. With this procedure we have explicitly constructed the functionals (15) and thereby the desired density functional (16) for the xc energy. Had we chosen $\hat{H}_0$ as unperturbed Hamiltonian and $\hat{U}$ as the perturbation, the diagrammatic analysis would have given the xc free energy as a functional $\tilde{\tilde{f}}_{\mathrm{xc}}^{\mathrm{hom}}[\mu_0, \Delta_0(\mathbf{k})]$ of the external potentials μ_0 and $\Delta_0(\mathbf{k})$. By virtue of the Hohenberg-Kohn theorem for superconductors [2], the latter can, in principle, be eliminated in favor of the densities $\{n, \chi(\mathbf{k})\}$. In practice, however, the required interacting functionals $\mu_0[n, \chi(\mathbf{k})]$ and $\Delta_0[n, \chi(\mathbf{k})]$ are not known while the noninteracting KS relations (15) can be constructed explicitly as shown above. For this reason we performed the RPA resummation in terms of the KS propagators.

To gain some insight in the relative importance of the anomalous Hartree, the exchange, and the correlation contributions we have evaluated these energies for the simple model pair potential $\Delta(\mathbf{k}) = \delta \exp(-\frac{(k-k_F)^2}{\sigma^2})$, where k_F is the Fermi wave vector and δ and σ are parameters. In Fig. 3 we show the difference of exchange energies, $f_x^S - f_x^N$, in the superconducting (S) and normal conducting (N) states, the negative difference, $-(f_c^{\mathrm{RPA},S} - f_c^{\mathrm{RPA},N})$, of the corresponding RPA correlation energies, and the anomalous Hartree energy, f_{AH}. The plotted values are the energy densities corresponding to zero temperature and $r_s = 1$. Their dependence on the parameters δ and σ turns out to be rather smooth. The exchange part is positive and roughly 1 order of magnitude smaller than the other two terms. The anomalous Hartree term gives rise to a large positive contribution. The RPA correlation energy difference $(f_c^{\mathrm{RPA},S} - f_c^{\mathrm{RPA},N})$, on the other hand, leads to a large negative contribution which nearly cancels the positive Hartree term; the sum of the three terms is positive everywhere. The same statement holds

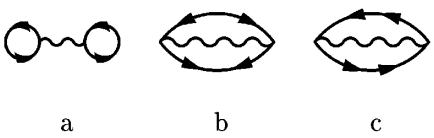

FIG. 1. First order diagrams contributing to the free energy.

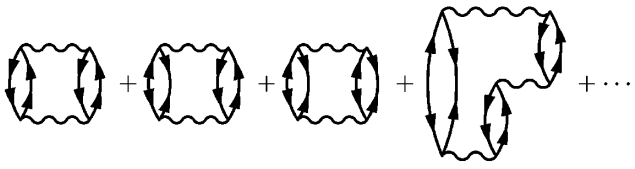

FIG. 2. The RPA diagrams.

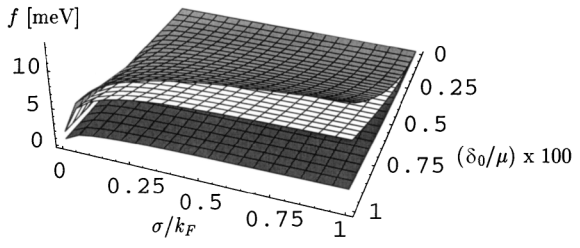

FIG. 3. Numerical results for $f_x^S - f_x^N$ (lower sheet), $f_c^{\mathrm{RPA},N} - f_c^{\mathrm{RPA},S}$ (middle sheet), and f_H (upper sheet) ($r_s = 1$).

true for $r_s = 0.1$, 1, 2, 3, 4, and 5. In the conventional s-wave superconductors these Coulombic positive-energy contributions are overcome by negative contributions due to the electron-phonon coupling. We emphasize that for the prediction of material-specific properties, such as T_c, it is indispensible, even for the conventional s-wave superconductors, to treat both the electronic correlations and the electron-phonon coupling from first principles. While the former are taken care of by the proposed LDA functional, the latter can be accounted for by an appropriate "external" pairing field Δ_0 in Eq. (4). In the weak-coupling regime, Δ_0 is well represented by the ordinary mean-field potential associated with the Bardeen-Pines [11] interaction. Strong electron-phonon coupling can also be treated within the DFT for superconductors. This, however, is a separate matter which will be discussed elsewhere [12].

In this work we have constructed an LDA-type functional to be employed in the description of real (in particular, inhomogeneous) materials. The functional is obtained from the xc free energy of a homogeneous electron gas exposed to an external pairing field. Although it is not the main purpose of this Letter, we finally address the question whether the homogeneous electron gas itself, i.e., without external pairing field, has a superconducting phase. This is a completely different and rather subtle issue. It should be remembered that the LSDA as it is commonly applied in ordinary DFT is not able to describe realistically the spin-polarized phases of the homogeneous electron gas *without* external magnetic field. These phases have been reliably calculated only very recently [13]. The question whether the electron gas has a superconducting phase has been discussed in a number of publications: In a classical paper, Kohn and Luttinger [14] predicted a superconducting phase at very low T_c with an order parameter of high angular momentum. A different mechanism, due to plasmon exchange, was suggested later by Takada [15]. Sham and co-workers [16] solved the Eliashberg equations with an RPA-screened interaction and found superconductivity at unrealistically high critical temperature. The inclusion of vertex and other corrections [17] lowers T_c considerably. To investigate this issue within the present DFT context, we have to solve the KS equations (1) and (2) for a homogeneous system with vanishing external pairing field. Clearly, in the case of a uniform gas, the solutions for the particle and hole amplitudes are plane waves. The self-consistency condition (4) for the KS pairing potential is equivalent to a gap equation that contains both mean-field (Hartree) and xc contributions. We found that this gap equation has no nonvanishing solutions with s-wave symmetry. Whether or not the proposed RPA functional allows solutions of the gap equation with higher angular momentum remains to be investigated. Work along these lines is in progress.

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