Localized superconductivity

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From a self-consistent solution of the mean-field theory describing a negative-U impurity embedded in a tight-binding band of fermions, we derive the conditions necessary for the formation of an off-diagonally ordered local moment in contact with a continuum of unpaired electrons. We calculate the electron removal and addition spectra and show that the spectrum is gapless, although in the ordered state some spectral weight is shifted away from the region around the Fermi level. We discuss the temperature dependence and the proximity-induced distance dependence of the order parameter and discuss the coupling between local moments as a function of distance and temperature. We predict that for a sufficiently strong attractive interaction the superconducting phase transition occurs between a state with off-diagonal long-range order below T_c and off-diagonal short-range order above T_c.

I. INTRODUCTION

In the Bardeen-Cooper-Schrieffer theory[1] of superconductivity the ground state is formed by a condensate of fermions bound in Cooper pairs. Although the operators describing Cooper pairs generally do not obey Bose commutation rules, many of the physical properties of a condensate of bosons and the BCS ground state are the same. The difference between the BCS state and a system of condensed bosons appears if one compares the behavior at and above the critical temperature. Whereas the phase transition in the former is driven by breaking up Cooper-pairs, in the latter the transition takes place through thermal excitation of individual bosons out of the condensate. As a result the normal state is a liquid of interacting fermions in the former and a gas of bosons in the latter case.[2–7] Cooper pairs do not exist as individual particles outside the condensate due to the low-energy scale of pair formation compared to the kinetic-energy scale of pairs of electrons. As a result the pairs cease to exist at a critical temperature much lower than the temperature where they would start to move individually. Usually one argues that the coherence length $\xi_0 = \frac{\hbar v_F}{\pi \Delta_0}$ is much larger than the average separation between neighboring pairs $\beta \lambda_F = \frac{\pi \beta \hbar v_F}{E_F}$, where $\beta = 0.50$, 0.56, 0.62 for a dimensionality of 1, 2, and 3, respectively. The crossover therefore occurs near $\pi^2 \beta \Delta_0 / E_F = 1$. In "classical" BCS superconductors such as Al one finds Δ_0 typically of the order of a few meV, and E_F of several eV, so that Cooper pairs do not exist outside a condensate.

With the advent of superconductivity in systems characterized by a short coherence length of the order of a few lattice spacings and a relatively dilute gas or fluid of electrons or holes the debate about the stability of local pairs has gained new momentum. In the high-T_c cuprates a Δ of about 20–30 meV has been found with far infrared,[8,9] energy loss,[10] photoelectron spectroscopy,[11] tunneling,[12,13] inelastic light scattering,[14] and phonon frequency shifts and widths,[15] whereas the Fermi energy is usually estimated to be of the order of 100–200 meV.[16] Hence the parameter $\pi^2 \beta \Delta_0 / E_F$ is somewhere between 0.6 and 1.8. There are several experimental hints that the phase transition could be unusual: (1) non-Fermi-liquid-like properties of the "normal" state,[17] (2) two-fluid phenomenology[18,19] of the superconducting phase transition, and (3) classification of several types of "exotic" superconductors close to—but not at—the Bose condensation line in the T_c versus T_F phase diagram.[20]

Usually a distinction is made between BCS superconductivity and Bose condensation of preexisting pairs. In this paper we follow a different approach by studying the off-diagonal short-range order (ODSRO) in the absence of bulk superconductivity characterized by off-diagonal long-range order (ODLRO). The possibility that such a situation could exist has already been pointed out by White and Geballe,[21] who stated that "...it is interesting to note that the BCS theory imposes long-range order when pairing occurs by requiring that the pairs all have the same phase ϕ. We are not aware of any situation where 'incoherent' pairing occurs." In this paper we present a study of the stability of a system with a single pairing site against the formation of a state with ODSRO. This situation is very close to the formation of a local pair in a narrow band system.[22–28]

The organization of the paper is as follows: In Sec. II we derive the self-consistent equations for an off-diagonally ordered local moment. In Sec. III A we present analytical expressions for the self-consistency condition. We discuss the free energy and the temperature dependence of the local order parameter. In Sec. III B we give numerical results of the single electron removal and addition density of states (or the photoelectron and inverse-photoelectron spectra) for the locally ordered system, showing that transfer of spectral weight away from the Fermi level occurs without the formation of a true energy gap. In Sec. III C we discuss the similarity to the Kondo

problem, and show that the phase of the local off-diagonal order is not a good quantum number. In Sec. III D we discuss the proximity-induced order parameter and the coupling between centers as a function of distance, and vector potential. We furthermore show that for a system of electrons with a sufficiently strong attractive interaction, the superconducting phase transition takes place from a state with ODLRO at low temperatures to a state with ODSRO at higher temperatures. In Sec. III E we consider the possibility of a staggered order parameter and discuss some of the consequences of this for the statistical properties of the many-body wave function.

II. MEAN-FIELD SOLUTION OF THE NEGATIVE-U IMPURITY PROBLEM

We consider a system described by the following tight-binding Hamiltonian:

$$H = \sum_{k,\sigma}(\epsilon_k - \mu)c^\dagger_{k\sigma}c_{k\sigma} + \sum_{R,\sigma} V_R c^\dagger_{R\sigma}c_{R\sigma} + \sum_R U_R n_{R\uparrow}n_{R\downarrow}, \tag{1}$$

where R refers to coordinates in the tight-binding lattice, and U_R is the site-dependent electron-electron attraction. The interaction between particles is assumed to be zero, except at a number of sites R where there is an attractive interaction between up and down spins. Hence we consider the situation where $U_R < 0$ at a number of sites and $U_R = 0$ elsewhere. The potential V_R at each negative-U site controls the average occupation. We will treat this many-body problem within the pairing ansatz of the BCS mean-field approximation. The method is complementary to the recent work by Gyorffy, Staunton, and Stocks,[29] who used the coherent-potential approximation in their treatment of the homogeneous negative-U Hamiltonian (i.e., with the same value of U at every site). Within this approximation one replaces the product over four operators with two operators multiplied with an average over the other two. In this particular case, where we concentrate on $U_R < 0$ we anticipate a finite expectation value of terms of the form

$$\Psi(R) \equiv < c_{R\uparrow}c_{R\downarrow} >,$$

whereas we assume terms of the type $< c^\dagger_{R\uparrow}c_{R\uparrow} - c^\dagger_{R\downarrow}c_{R\downarrow} >$ to remain zero. We therefore introduce the order parameter

$$\Delta_R \equiv -U_R\Psi(R) \tag{2}$$

with the help of which we linearize the Hamiltonian:

$$H = \sum_{k,\sigma}(\epsilon_k - \mu)c^\dagger_{k\sigma}c_{k\sigma} + \sum_{R,\sigma} V^{\text{eff}}_R c^\dagger_{R\sigma}c_{R\sigma} - \sum_R \Delta_R c^\dagger_{R\downarrow}c^\dagger_{R\uparrow} - \sum_R \Delta^*_R c_{R\uparrow}c_{R\downarrow}. \tag{3}$$

Here V^{eff}_R absorbs the diagonal Hartree-terms of the third term in Eq. (1). We now introduce the single-particle and anomalous Green's functions:

$$G^{R'\sigma'}_{R\sigma}(\tau,\tau') \equiv - < T_\tau[c_{R\sigma}(\tau)c^\dagger_{R\sigma'}(\tau')] >,$$
$$\bar{F}^{R'}_R(\tau,\tau') \equiv - < T_\tau[c^\dagger_{R\uparrow}(\tau)c^\dagger_{R\downarrow}(\tau')] > . \tag{4}$$

These Green's functions obey the following standard equations of motion:

$$\left(-\hbar\frac{\partial}{\partial\tau} - H' + \mu\right)G(\tau,\tau') - \Delta\bar{F}(\tau,\tau') = \hbar\delta(\tau-\tau'),$$
$$\left(\hbar\frac{\partial}{\partial\tau} - H' + \mu\right)\bar{F}(\tau,\tau') - \Delta^*G^\dagger(\tau,\tau') = 0, \tag{5}$$

where H' is the single-particle Hamiltonian consisting of the kinetic-energy part and the local potentials V^{eff}_R. We now express the equations of motion in terms of the thermal Green's functions:

$$G(\tau,\tau') \equiv k_BT\sum_n e^{-i\omega_n(\tau-\tau')}G(i\hbar\omega_n) \tag{6}$$

and a similar expression for $F(\tau,\tau')$. Here $\omega_n = (2n+1)\pi k_BT/\hbar$ are fermion Matsubara frequencies. The coupled equations of motion have to be solved together with the self-consistency condition, which follows directly from the definition of the anomalous Green's function:

$$\Delta^*_R = -Uk_BT\sum\bar{F}(i\hbar\omega_n). \tag{7}$$

It is easy to show that the coupled equations can also be cast in the following (Dyson) form:

$$G(i\hbar\omega_n) = g(i\hbar\omega_n) - g(i\hbar\omega_n)\Delta\bar{F}(i\hbar\omega_n),$$
$$\bar{F}(i\hbar\omega_n) = g(-i\hbar\omega_n)\Delta^*G(i\hbar\omega_n), \tag{8}$$

where $g(i\hbar\omega_n)$ is the solution of the single-particle problem, i.e., the solution for $\Delta = 0$ including the local potentials V^{eff}_R. We are interested in the physics of a single negative-U impurity. Therefore we set $\Delta_R = 0$ everywhere, except at $R = 0$. In that case the equations of motion can be inverted directly resulting in the following expressions for the on-site anomalous Green's function:

$$\bar{F}^0_0(i\hbar\omega_n) = \frac{g^0_0(-i\hbar\omega_n)\Delta^*(T)g^0_0(i\hbar\omega_n)}{1 + g^0_0(-i\hbar\omega_n)\Delta^*(T)g^0_0(i\hbar\omega_n)\Delta(T)}, \tag{9}$$

from which we derive with Eq. (7) the following self-consistency condition:

$$1 = \sum_n \frac{-Uk_BT}{|\Delta(T)|^2 + [g^0_0(i\hbar\omega_n)g^0_0(-i\hbar\omega_n)]^{-1}}. \tag{10}$$

The Green's function g^0_0 for $\Delta = 0$ can be easily expressed in terms of the free-particle Green's function f^0_0 and the on-site potential V^{eff}_0, using standard techniques which are often employed in the literature on localized

states:[30–34]

$$g_0^0(i\hbar\omega_n) = \frac{f_0^0(i\hbar\omega_n)}{1 - V_R f_0^0(i\hbar\omega_n)},$$
$$f_0^0(i\hbar\omega_n) = \frac{1}{N}\sum_k \frac{1}{i\hbar\omega_n + \mu - \epsilon_k}, \tag{11}$$

where N is the number of lattice sites. If we take $T \to 0$ the summation over Matsubara frequencies becomes an integral, resulting in

$$1 = \int_0^\infty \frac{-\pi^{-1} U \hbar d\omega}{|\Delta(0)|^2 + [g_0^0(i\hbar\omega) g_0^0(-i\hbar\omega_n)]^{-1}}, \tag{12}$$

from which we can solve $\Delta(0)$.

The local density of states at the site of the impurity is obtained by analytic continuation to the real energy axis of the single-particle Green's function $G_0^0(i\hbar\omega_n)$, which is obtained from the coupled Dyson equations:

$$\rho(E) = \pi^{-1}\Im G_0^0(E),$$
$$G_0^0(i\hbar\omega_n) = \frac{g_0^0(i\hbar\omega_n)}{1 + g_0^0(-i\hbar\omega_n)\Delta^*(T) g_0^0(i\hbar\omega_n)\Delta(T)}. \tag{13}$$

We will be interested in the spatial dependency of the proximity-induced superconductivity at neighboring sites. This has to be calculated from the anomalous Green's function at those sites, which again follows from the solution of Eq. (8):

$$G_R^R(i\hbar\omega_n) = g_R^R(i\hbar\omega_n) - \frac{g_R^0(i\hbar\omega_n) g_0^R(i\hbar\omega_n)|\Delta(T)|^2 g_0^0(-i\hbar\omega_n),}{1 + g_0^0(-i\hbar\omega_n) g_0^0(i\hbar\omega_n)|\Delta(T)|^2},$$
$$\bar{F}_R^R(i\hbar\omega_n) = \frac{g_R^0(-i\hbar\omega_n)\Delta^*(T) g_0^R(i\hbar\omega_n)}{1 + g_0^0(-i\hbar\omega_n) g_0^0(i\hbar\omega_n)|\Delta(T)|^2}. \tag{14}$$

From this expression we immediately see that the phase of the order parameter does not depend on R and is equal to the phase of Δ at the origin. As we will point out in more detail in the discussion, the coupling between the local superconducting moments is therefore always such that the phases are equal, unless an additional gauge field rotates the phase. As we can see the numerical evaluation of this expression requires knowledge of the site off-diagonal noninteracting Green's function g_0^R, which for the present system with a perturbation potential V_0^{eff} at the origin is of the following form:[30–33]

$$g_0^R(i\hbar\omega_n) = \frac{f_0^R(i\hbar\omega_n)}{1 - V_R f_0^0(i\hbar\omega_n)},$$
$$f_0^R(i\hbar\omega_n) = \frac{1}{N}\sum_k \frac{e^{-ikR}}{i\hbar\omega_n + \mu - \epsilon_k}. \tag{15}$$

For a general band structure the analytical form of f_0^R can be quite complicated. In this study we will restrict the discussion to some examples where the band structure is very simple, allowing a straightforward analytical continuation in the complex plane.

The free energy of the superconducting and the normal state at $T = 0$ is the expectation value of the Hamiltonian. Hence the free energy is

$$\Omega(\Delta, T \to 0) = \left\langle\left\langle \sum_{k,\sigma}(\epsilon_k - \mu) c_{k\sigma}^\dagger c_{k\sigma} + \sum_{R,\sigma} V_R^{\text{eff}} c_{R\sigma}^\dagger c_{R\sigma} \right\rangle\right\rangle_{\Delta, T\to 0} + U_R \sum_R < |\Psi(R)|^2 >_{\Delta, T\to 0} . \tag{16}$$

With the help of the definition of the Green's functions [Eqs. (4) and (6)] and Δ [Eq. (7)] the free-energy difference with the normal state can now be cast in the following form:

$$\Omega(\Delta, 0) - \Omega(0, 0) = \sum_R U_R |\Psi(R)|^2 + \left(\sum_{R,n} 2k_B T [< R|H' G(i\hbar\omega_n)|R > - < R|H' g(i\hbar\omega_n)|R >] \right)_{T\to 0}, \tag{17}$$

where H' is again the first two terms of Eq. (3). With the help of the relation $H' = i\hbar\omega_n - g^{-1}(i\hbar\omega_n)$ and using Eq. (8) we rewrite this as

$$\Omega(\Delta, 0) - \Omega(0, 0) = \sum_R U_R |\Psi(R)|^2 + \left(\sum_{R,n} 2k_B T < R|i\hbar\omega_n [G(i\hbar\omega_n) - g(i\hbar\omega_n)] + \Delta F^\dagger(i\hbar\omega_n)|R > \right)_{T\to 0}, \tag{18}$$

which, using Eq. (14) for the case of a single negative-U impurity and after making the change of variables $x = (2n+1)\pi k_B T$, becomes

$$\Omega(\Delta, 0) - \Omega(0, 0) = -U|\Psi(0)|^2 + \pi^{-1}|\Delta|^2 \int_{-\infty}^{\infty} \frac{1 - ix\sum_R g_R^0(ix) g_0^0(-ix) g_0^R(ix)}{1 + |\Delta|^2 g_R^0(ix) g_0^R(ix)} dx. \tag{19}$$

From the definition of a single-particle Green's functions it is easy to prove that

$$\sum_R g_0^R(ix) g_R^0(ix) = -\frac{\partial}{\partial ix} g_0^0(ix). \tag{20}$$

We are now ready to integrate Eq. (19), which by means of partial integration becomes

$$\Omega(\Delta,0) - \Omega(0,0) = -\pi^{-1} \int_0^\infty dx \ln\left[1 + |\Delta|^2 g_0^0(ix) g_0^0(-ix)\right] - \frac{|\Delta|^2}{U}. \tag{21}$$

As the minimum of the free energy as a function of Δ should correspond to the self-consistency condition [Eq. (12)], we can check our result by differentiating Eq. (21) with respect to $|\Delta|^2$. Indeed one easily obtains Eq. (12) with this procedure.

III. DISCUSSION

A. Self-consistent solution of the order parameter

We first derive the criterion for the existence of ODSRO. The easiest example, which can also be solved analytically, is the Anderson impurity model. In this model extra impurity levels are added to the Hilbert space. The Hamiltonian is

$$H = \sum_{k,\sigma} (\epsilon_k - \mu) c_{k\sigma}^\dagger c_{k\sigma} + \sum_\sigma \epsilon_0 \psi_{0\sigma}^\dagger \psi_{0\sigma} + \sum_{k\sigma} V_k (c_{k\sigma}^\dagger \psi_{0\sigma} + \psi_{0\sigma}^\dagger c_{k\sigma}) + U_0 n_{0\uparrow} n_{0\downarrow}. \tag{22}$$

The site-projected single-particle Green's function at the impurity is calculated without the last term, and becomes

$$g_0^0(\epsilon) = \frac{1}{\epsilon - \epsilon_0 - \sigma(\epsilon)}, \qquad \sigma(\epsilon) = \sum_k \frac{|V_k|^2}{\epsilon - \epsilon_k}. \tag{23}$$

If we make the simplifying assumption that the product of the density of states with $|V_k|^2$ is constant, we can replace $\sigma(\epsilon)$ with a constant decay rate (-)$i\Gamma$ for ϵ in the (upper) lower half of the complex plane. Together with Eq. (12) we arrive at the following self-consistency condition for the local order parameter:

$$1 = \int_0^\infty \frac{-\pi^{-1} U \hbar d\omega}{\epsilon_0^2 + |\Delta(0)|^2 + (\hbar\omega + \Gamma)^2}. \tag{24}$$

Direct integration leads to the condition

$$\sqrt{|\Delta|^2 + \epsilon_0^2} = \Gamma \tan\left(\frac{\pi\sqrt{|\Delta|^2 + \epsilon_0^2}}{-U}\right). \tag{25}$$

It is easy to prove that a solution for Δ exists if and only if

$$-U \frac{\arctan(\epsilon_0/\Gamma)}{\pi\epsilon_0} > 1, \tag{26}$$

which now looks very similar to the condition derived by Anderson for the single-magnetic-impurity problem.[35] A similar criterion was found recently by Gyorffy, Staunton, and Stocks using a numerical calculation based on the coherent-potential approximation.[29] It can also be seen immediately that for $-U \gg \Gamma$ the order parameter Δ is approximately equal to $-U/2$.

We now consider the dependency Δ on $-U$ in the Wolff model. We first treat the example of a quasi-two-dimensional (2D) density of states (DOS), i.e., a square density of states with a bandwidth W. We will scale all expressions to this bandwidth. The Green's function is in that case

$$f_0^0(i\hbar\omega_n/W) = \ln\left(\frac{2\mu/W + 1 + 2i\hbar\omega_n/W}{2\mu/W - 1 + 2i\hbar\omega_n/W}\right). \tag{27}$$

In Fig. 1(a) we display the order parameter as a function of interaction potential for a band which is precisely half filled, i.e., for $\mu = W/2$, using three different values for $V_0^{\rm eff}$, i.e., $V_0^{\rm eff}/W = 0$, 0.5, and 1. We see that in all three cases the order parameter is zero until a critical value is reached which depends on $V_0^{\rm eff}$. The lower this critical value is, the easier ODSRO will occur. The most favorable case for off-diagonal ordering occurs for this band filling if $V_0^{\rm eff} = 0$, i.e., if the average number of electrons at the negative-U site equals 1. In Fig. 1(b) we plot the same set of curves, but now for a band that has

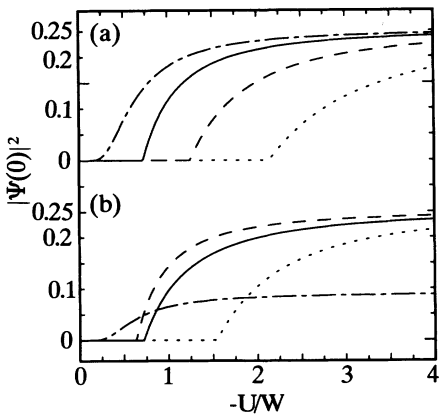

FIG. 1. $|\Psi(0)|^2$ vs $-U/W$ for the square band model. (a) $k_B T_F/W = 0.5$, and $V_0^{\rm eff}/W = 0$ (solid), 0.5 (dashed), 1.0 (dotted curve). Dash-dotted curve: The same for the negative-U lattice with $k_B T_F/W = 0.5$. (b) $k_B T_F/W = 0.9$, and $V_0^{\rm eff}/W = 0$ (solid), 0.4 (dashed), 1.0 (dotted curve). Dash-dotted curve: The same for the negative-U lattice with $k_B T_F/W = 0.9$.

90% filling. Here we see that the most favorable case for ODSRO occurs for $V_0^{\rm eff}/W \simeq 0.4$, which as we will see later, again corresponds to a situation where the average occupation of the impurity site is close to 1. We can see from these plots that all curves asymptotically approach the behavior $\Delta = U/2$. This is the expected behavior of a set of unhybridized negative-U centers in mutual equilibrium (i.e., pairs of electrons can be swapped between centers without a cost of energy). As has been explained by Anderson[38] both the energy necessary to remove and to add an electron are exactly $-U/2$ for such a system. Also indicated in the same figure is the solution for the gap using the negative-U Hubbard model, i.e., assuming the same value of U at each site. The solution for the square band model is[39]

$$\Delta/W = \frac{\sqrt{k_B T_F/W(1-k_B T_F/W)}}{\sinh(-W/U)}, \tag{28}$$

where T_F is the Fermi temperature. We see that at exact half filling this equation has the proper limiting behavior for $U \rightarrow \infty$. However, away from half filling this limiting behavior is no longer found. This result probably reflects some of the limitations of the mean-field Hartree approach to the negative-U Hubbard lattice, as for $U \gg W$ the role of hybridization is suppressed and the solid essentially behaves as a set of unhybridized atoms in electrical equilibrium, for which we expect $\Delta \rightarrow -U/2$. Apparently the *impurity* model treated with mean-field theory works much better and shows the correct limiting behavior also in the limit of $U \gg W$. To demonstrate the effect of average occupation at the impurity level we display in Fig. 2 the critical value of U where ODSRO sets in versus $V_0^{\rm eff}$ for a number of different values for the band filling. Again we see that U_c becomes very low for a low density of electrons or holes, but with $V_0^{\rm eff}$ adjusted such that the average occupancy at the negative-U impurity is close to half filling.

To see that our self-consistency condition Eq. (12) corresponds to a minimum of the free energy, we plot in Fig. 3 the free energy at T =0 for a number of different parameters, using Eq. (21). The value at the minimum of the free-energy curve is displayed as a function of $-U|\Psi(0)|^2$ in Fig. 4. As we can see from these curves, the free energy is $U|\Psi(0)|^2$ for strong coupling, and proportional to $-U^2|\Psi(0)|^4$ for U close to the critical value where ordering sets in. We checked numerically that in the whole range of parameters the free energy behaves approximately according to the formula

$$\Omega_s - \Omega_n \simeq -\frac{U^2|\Psi(0)|^4}{W/2 - U|\Psi(0)|^2}. \tag{29}$$

In Fig. 5 we show the temperature dependence of the order parameter calculated with Eq. (7) for a number of parameters. We have to add here that this critical behavior is an artifact from our mean-field approximation to the single-impurity coupled to an infinite bath

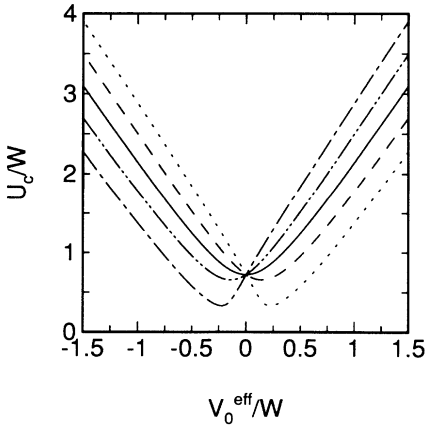

FIG. 2. $-U_c/W$ vs $V_0^{\rm eff}/W$ for $k_B T_F/W = 0.01$ (dash-dotted), 0.25 (dash double-dotted), 0.5 (solid), 0.75 (dashed), and 0.99 (dotted).

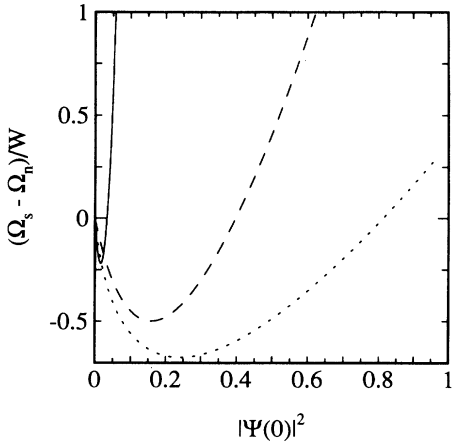

FIG. 3. Free energy vs $|\Psi(0)|^2$ at $T = 0$ for the square band model using the following parameters: $k_B T_F/W = 0.9$, $V_0^{\rm eff} = 0.5$, and $-U/W = 0.78$ (solid), 1.25 (dashed), and 4.08 (dotted curve). The vertical scale of the solid (dashed) curve is 1000-fold (10-fold) expanded.

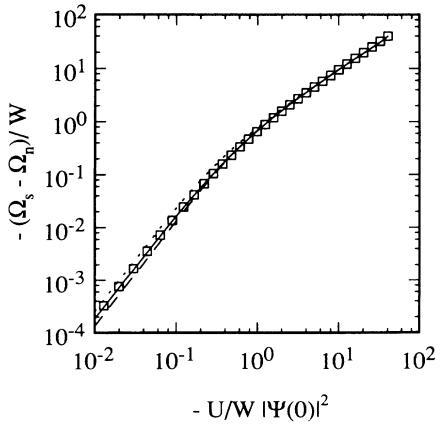

FIG. 4. Value of minimum of the free energy at $T = 0$ vs the corresponding value of $-U|\Psi(0)|^2$ for the following parameters: $k_B T_F/W = 0.9$ and $V_0^{\rm eff}/W = 0.0$ (solid), $k_B T_F/W = 0.9$, $V_0^{\rm eff}/W = 0.5$ (dashed), $k_B T_F/W = 0.99$, $V_0^{\rm eff}/W = 0.4$ (dotted curve). The open squares are the approximation formula given in Eq. (29).

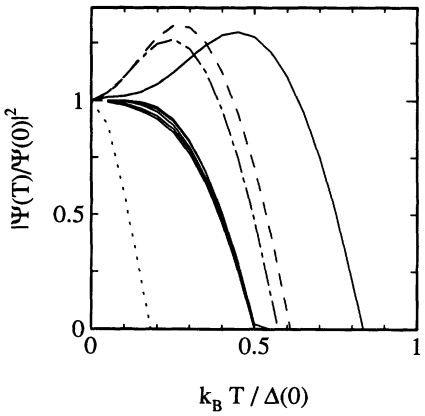

FIG. 5. Temperature dependence of the order parameter. Solid, dashed, and dash-dotted curves: $k_BT_F/W = 0.9$, 0.99, and 0.999, respectively, with $V_0^{\rm eff}/W = 0.4$ and $\Delta/W = 0.1$. Dotted curve: $k_BT_F/W = 0.99$, $V_0^{\rm eff}/W = 0$, and $\Delta/W = 0.1$. Other solid curves: $k_BT_F/W = 0.9$ and 0.99, $V_0^{\rm eff}/W = 0$ and 0.4, and $\Delta/W = 1.0$, and 10.

of electrons, which should disappear if we include many-body corrections. We should therefore be careful and regard the resulting "critical temperature" as a typical temperature where ordering vanishes, not as a temperature where a phase transition takes place. We notice that for a wide range of different parameters the temperature dependence close to T_c behaves BCS-like, i.e., $|\Psi|^2$ approaches zero linearly when the temperature approaches T_c, with $2\Delta/T_c$ close to 4. In the regions of parameter space where $\Delta(0)/|U|$ is small, T_c differs strongly from this value in either direction. In particular for choices of parameters where Δ is small, close to an average occupation of one electron at the impurity, the order parameter first *increases* on increasing T and becomes zero at a relatively high temperature. In fact in a narrow region of U values *below* U_c reentrant behavior exists, where the order parameter is zero at zero temperature, has a second-order phase transition at a lower critical temperature to a finite value, and becomes zero again at a higher critical temperature. If the average occupation at the impurity is close to two electrons we see that for small Δ the transition temperature is strongly supressed.

It is perhaps illustrative to compare the off-diagonally ordered local moment to a local pair (e.g., a bipolaron). The usual way to think about local pairs is as a stable pair of electrons. An off-diagonally ordered local moment, on the other hand, is a mixture of states with two and zero electrons at the impurity site, which is in equilibrium with its *unpaired* environment. The latter is an improvement over the local-pair picture, especially in the region of parameter space where the pairs overlap, or where they exist in equilibrium with unpaired electrons. In a recent study Bar-Yam considered singlet pairing in a model of a local-pair band which hybridizes with a separate wide band.[23] A similar situation has been considered for triplet pairing.[40] In these studies it is assumed that in spite of the fact that the local-pair band hybridizes with a wide band, local pairing is conserved. In the present study of a single negative-U center and a many-electron system we show that the pair binding energy has to be at least of the order of the bandwidth. In studies of bipolaronic superconductivity a similar condition follows from considering two interacting electrons in an empty lattice.[25]

B. The density of states at a negative-U impurity

In Fig. 6 we display the calculated density of states for a band which is 90% filled, both for $V_0^{\rm eff} = 0$ and $V_0^{\rm eff}/W = 0.4$, using a number of different values of Δ. All curves have been given an artificial lifetime broadening of $0.1W$ to remove singularities in the spectra. We make a number of observations here: For $V_0^{\rm eff} = 0$ [Fig. 6(a)] the local density of states (LDOS) with $\Delta = 0$ is just the (broadened) DOS of our square band model. Upon increasing U, and therewith Δ, some spectral weight is transferred away from the Fermi energy towards higher excitation energies. Most of the intensity is built up at sharp points in the DOS, which in this case is at the top and bottom of the band. On further increasing the local order parameter a quasigap develops with a sharp single peak in the electron removal and addition spectra, both at $E = U/2$. The weight of the two peaks is not the same, and reflects the average number of electrons at the negative-U impurity site. One can see this in the following way: The energy of putting two electrons at the same site (with opposite spin) is twice the single-electron addition energy plus the interaction energy U (which we assume is negative). In order to obtain a ground state which has an occupancy between 0 and 2 the zero-electron and two-electron energy levels have to be degenerate. In other words $2[E(1)-E(0)]+U = 0$. As a result the single-electron addition energy equals $-U/2$. The ground-state wave function is in this case

$$|\psi_g> = u|\psi(0)> + ve^{i\phi}|\psi(2)> \qquad (30)$$

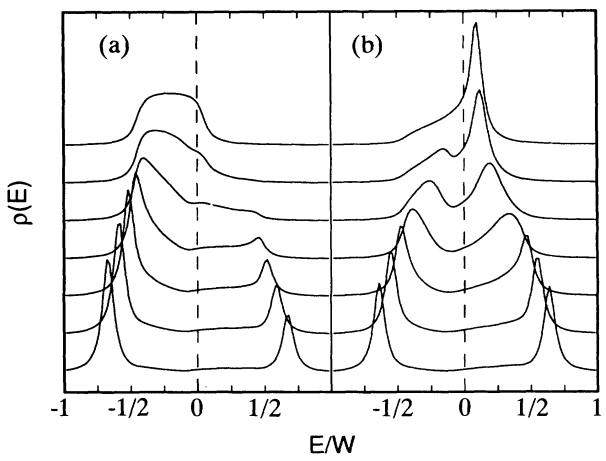

FIG. 6. Local density of states at the negative-U impurity. The vertical dashed curve is the Fermi energy. The DOS on the left (right) -hand side of E_F corresponds to the single electron removal (addition) density of states. The parameters used in the calculation are $k_BT_F/W = 0.9$, with (a) $V_0^{\rm eff}/W = 0$, and (b) 0.4. From top to bottom: $\Delta/W = 0$, 0.2, 0.4, 0.6, 0.8, 1.0, and 1.2.

and the intensity of the one-electron removal and addition peaks is just the weight of the doubly and unoccupied character in the ground state. Hence the one-electron removal peak has a weight of v^2 and the one-electron addition peak has a weight of u^2. Note that the above ground state is doubly degenerate. This degeneracy is lifted if the negative-U impurity couples to a band of electrons. The splitting in that case reflects a low-energy scale which is related to the Kondo problem, as has been pointed out by Haldane.[41]

We also see in In Fig. 6(a) that for very high values of U the difference in weight of the electron removal and electron addition peaks becomes smaller. In Fig. 6(b) we study the same band filling, but with $V_0^{\text{eff}} = 0.4$ at the impurity site. As we can see in Fig. 1(b) this value of the potential corresponds to having a lower critical value of U where ODSRO occurs. From the plot of the LDOS with $\Delta = 0$ we see that this corresponds to having one electron on average at the impurity site. Note that although the *average* occupancy is 1, the ground state is in fact a mixture of zero and double occupancy with a *reduced* amount of singly occupied character. We see that like in Fig. 6(a) spectral weight is transferred away from the Fermi energy upon increasing Δ, without forming a true gap. We also note that in this case the relative weight of the peaks on both sides of E_F is approximately 0.5 for all values of Δ.

C. Similarities to the Kondo problem

The physical picture that emerges from this has many similarities to the magnetic impurity problem, as has also been discussed recently by Taraphder and Coleman.[42] Among many other issues these authors discussed the formal similarites to the Kondo problem, and introduced the term "isospin" for the zero- or double-occupation doublet at the impurity site. In their discussion they approached the problem from the strongly coupled limit, whereas we start from the weakly coupled regime, treating correlations with mean-field theory. Schematically the Kondo singlet state of a positive-U impurity can be written in the following way:

$$|\Psi_g(\phi)> = u\, \tilde{c}^{\dagger}_{0\downarrow} |\psi_{\uparrow}> + v e^{i\phi}\, \tilde{c}^{\dagger}_{0\uparrow} |\psi_{\downarrow}> . \qquad (31)$$

Here $|\psi_\sigma>$ indicates a state of the electron gas surrounding the magnetic impurity with $s_z = \sigma$, and $\tilde{c}^{\dagger}_{0\sigma}$ creates an electron with spin σ at site 0. The $\sim$ indicates that these are renormalized operators with some admixture of valence band character of the surrounding electrons. The Schrieffer-Wolff transformation projects out the spin-zero state at the impurity (i.e., zero or double occupation with an extra electron or hole in the valence band) and renormalizes the $c^{\dagger}_{\sigma}$ operators.[43] An external magnetic field orients the local moment and moves u^2 away from 0.5. Without a magnetic field the phase ϕ equals 0 and π for the triplet and singlet state, respectively. In the uncorrelated state the phase fluctuates randomly, thereby destroying the subtle correlation of spins which is responsible for the formation of a singlet state. In the Hartree-Fock description, such as used in the early papers on the magnetic impurity problem by Anderson, Wolff, and Clogston,[35–37] the formation of a singlet ground state is not included. Due to this lack of correlation the ground state of the local moment is doubly degenerate, i.e., the orientation of the spin can be either up or down. The equivalent wave function for the ground state of the off-diagonally ordered local moment is

$$|\Psi_g(\phi)> = u|\Psi(N)> + v e^{i\phi}\, \tilde{c}^{\dagger}_{0\uparrow}\, \tilde{c}^{\dagger}_{0\downarrow} |\Psi(N-2)>, \qquad (32)$$

where $|\Psi(N)>$ indicates the wave function of the surrounding electron gas containing N electrons. Again the $\sim$ indicates that the operators are renormalized by means of a canonical transformation, which projects out the singly occupied character at the impurity. This canonical transformation was given in Ref. 40. The weight factor u is now determined by the chemical potential and the local fields at the impurity, which fixes the average occupation. As in the Hartree-Fock theory of the magnetic impurity problem, the Hartree ground state of the ordered negative-U impurity has an essential degeneracy, which exists in this case with respect to the choice of phase of the order parameter Δ. This is the phase factor $e^{i\phi}$ in the above expression. Indeed it is a well-known result of BCS theory that phase is a broken symmetry of the ground state of a superconductor. Due to the fact that the ground state is degenerate with respect to the choice of phase one is allowed to take linear combinations of different values of ϕ, by means of which one can construct a wave function where the number of particles is a good quantum number and the phase is undefined. In the above example of a negative-U impurity one is tempted to assume that it is allowed to take even and odd combinations of $|\Psi_g(\pi)>$ and $|\Psi_g(0)>$, resulting in two degenerate solutions with zero and two electrons at the local moment. Although this is true in the limit of $-U/W \rightarrow \infty$, it is no longer correct if the moment is coupled to the surrounding metallic state. The anomalous scattering of the conduction electrons on the off-diagonally ordered local moment (which is in this case Andreev scattering[44] rather than spin-flip scattering) correlates the isospin of the surrounding electron gas with that of the negative-U site and removes the ambiguity in ϕ. In other words: *Although the Hartree theory of the negative-U impurity problem allows a free choice of phase of the order parameter, this is an artifact of the mean-field treatment. The true ground state has no degeneracy with respect to the choice of phase.* As in the magnetic analogon one expects the phase coherence to be lost gradually at elevated temperatures *before* the off-diagonal order itself disappears. Taraphder and Coleman showed that the temperature scale is in a similar way as in the Kondo problem given by the relation $T_{\psi} = |U|(\pi J\rho)^{1/2} \exp[-1/2J\rho]$, where J is the isospin-exchange parameter $2V^2/|U|$, and V is the hopping parameter between the impurity and the surrounding electron gas. In terms of our model parameters $V \simeq W/2$ and $\rho = 1/W$. Hence

$$k_B T_\psi / W \simeq \sqrt{\frac{\pi}{2}|U|/W} e^{-|U|/W}. \tag{33}$$

Hence the highest scaling temperature occurs near $U = -W$. For larger values of $-U$ the isospin starts to behave as an unhybridized negative-U impurity which swaps pairs of electrons to and from the surrounding electron gas incoherently. In that case coherence sets in at exponentially low temperatures.

D. Proximity coupling between negative-U centers

We now come to the discussion of the dependency of the order parameter on distance to the off-diagonally ordered local moment. For that purpose we consider a tight-binding linear chain model with lattice constant a, for which the Green's function is[34]

$$f_0^R(E) = \frac{2\rho_1^{R/a}}{2E/W - 1},$$
$$\rho_1(E) \equiv 2E/W - 1 - \sqrt{(2E/W-1)^2 - 1}\,. \tag{34}$$

With the help of Eqs. (7) and (14) we calculate the distance dependency of the order parameter

$$t_{1,0} e^{i\eta_{1,0}} \equiv \Psi_0(R_1)/\Psi_0. \tag{35}$$

Here $\eta_{i,j}$ is the phase factor of $\Psi_i(R_j)/\Psi_i$, which is 0 in the present case, as we can see by combining Eq. (14) with Eqs. (4) and (6). The result is presented in Fig. 7. We took $V_0^{\rm eff} = 0$, $k_B T_F/W = 0.9$, and $\Delta/W = 0.1, 0.5$, and 1.0. The order parameter decreases algebraically as a function of distance with a $1/R$ behavior. We see that for large Δ and $-U$ the proximity-induced order becomes smaller. In fact, for $|U| \gg W$ the proximity-induced order is proportional to $W/|U|$.

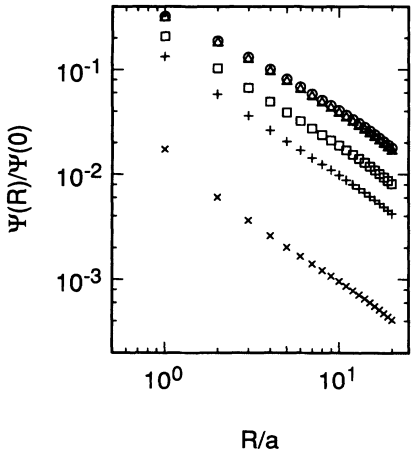

FIG. 7. The order parameter $< \Psi(R)/\Psi(0) >$ vs R for a negative-U impurity in a 1D tight-binding model. $k_B T_F/W = 0.9$, and $V_0^{\rm eff} = 0$. From top to bottom: 1/R, Δ/W=0.01 (open circles), 0.1 (closed circles), 0.5 (open squares), 1.0 (closed squares), and 10 (crosses).

We now consider the coupling between negative-U impurities at positions R_0 and R_1. Center one has a local on-site order parameter Ψ_0 and induces an order parameter $t_{1,0} e^{i(\phi_1+\eta_{1,0})}\Psi_0$ at site R_1 and vice versa for center two. Due to this one adds additional terms

$$-U t_{1,0} e^{i(\phi_1+\eta_{1,0})}\Psi_0 c_{1,\downarrow}^\dagger c_{1,\uparrow}^\dagger - U t_{1,0} e^{-i(\phi_1+\eta_{1,0})}\Psi_0^* c_{1,\uparrow} c_{1,\downarrow} \tag{36}$$

and similar terms for site R_1 to the Hamiltonian. Here ϕ_i denotes the phase of order parameter Ψ_i at site R_i. In the limit of small Ψ the energy due to these additional terms adds linearly to the the Hamiltonian of Eq. (3), and one obtains the following cross terms:

$$\Omega_X = -J_{1,0}\cos(\phi_1 - \phi_0 + \eta_{1,0}) - J_{0,1}\cos(\phi_0 - \phi_1 + \eta_{0,1}), \tag{37}$$

where (in the limit of small Ψ) the coupling parameter is

$$J_{i,j} = -2U t_{i,j} |\Psi_i \Psi_j|. \tag{38}$$

In the presence of a vector potential the single-particle Green's functions g_0^R occurring in the solution of $\Psi(R) =< c_{R\uparrow} c_{R\downarrow} >$ [see Eq. (14)] acquire a phase factor $\eta = e/\hbar c \int \mathbf{A}\cdot d\mathbf{s}$. As two of these functions occur in the numerator of Eq. (14) this phase factor enters with a factor of 2 in the distance dependency of the proximity-induced order parameter. Hence $\eta_{2,1} = -\eta_{1,2} = 2e/\hbar c \int \mathbf{A}\cdot d\mathbf{s}$ in the presence of an external magnetic field. This rotates the phases of the two impurities with respect to each other. If the negative-U centers form a regular array, the free energy describing the coupling of the system in the presence of a magnetic field becomes

$$\Omega_X = -J_{i,j}\sum_{i,j}\cos\left(\phi_i - \phi_j - 2e/\hbar c \int_{R(i)}^{R(j)} \mathbf{A}\cdot d\mathbf{s}\right), \tag{39}$$

which is similar to the free-energy expression for an array of Josephson junctions.[45,46] A similar expression has been recently derived by Doniach using the mapping of the local-pair picture on the x-y model.[47] We should point out here, that from our analysis we find that the sign of $J_{i,j}$ is always positive, which implies that in the absence of a field $\mathbf{A}$ the coupling between sites is ferromagnetic.

In Eq. (39) the energy scale of the long-range ordering temperature is determined by the coupling parameter $J_{i,j}$ between sites, which as we have seen above, can be drastically reduced from the on-site free energy. To be more precise: In a 2D system such an arrangement gives rise to a Kosterlitz-Thouless transition. If we assume that the occupation of the electron gas is away from half filling, or that the negative-U impurities occur at every second site, the energy scale of the intermoment coupling $J_{i,j}$ between neighboring isospins is in the limit of small Ψ about $0.2\Delta^2/U \simeq 0.1\Delta$. The corresponding Kosterlitz-Thouless temperature[48] is in that case of the same order as the coupling energy, i.e., $T_c \simeq J \simeq 0.1\Delta$. In a simple cubic 3D lattice the classical XY model has a phase

transition at $T_c \simeq 2.2J \simeq 0.2\Delta$.[49] These values should be compared to the ODSRO temperature of $\sim 0.5\Delta$. We see that the bulk ODLRO T_c can be drastically reduced compared to the onset of local off-diagonal order. If $|U| \gg W$ the interaction terms in Eq. (3) are no longer the small parameter in the problem, and problems with convergence occur if one simply takes the linear superposition which we used in Eq. (38). In fact the change of energy resulting from the extra terms in Eq. (36) is now controlled by the hopping of pairs on and off the negative-U sites, which is proportional to $W^2/|U|$. Hence in that limit the correct expression for J is

$$J_{i,j} \propto \frac{W^2}{-U} t_{i,j}. \tag{40}$$

As both factors are proportional to $|U|^{-1}$ for large $|U|$, the coupling constant varies as U^{-2}. For neighboring negative-U centers the coupling is proportional to $|U|^{-1}$.

In the limit of $|U| \gg W$ one can regard the above system of negative-U impurities as a system of pairs hopping between negative-U sites. The kinetic-energy scale is in that case determined by the bandwidth of the collective modes, which is $J_{i,j}$, and for a 3D system the phase transition is a Bose-Einstein condensation of the preexisting pairs into the state at $\mathbf{k} = \mathbf{0}$. A different approach to the same problem is to consider the proximity-induced pairing between electrons in the regions between the negative-U impurities. Here electrons are paired due to a mechanism where they enter and leave the negative-U sites,[40,50] as indicated in Fig. 9(e) of Ref. 23(a). Due to the reverse process [see Fig. 9(d) of Ref. 23(a)] the paired states located at the negative-U sites acquire a finite bandwidth V^2/W, where V is the hopping parameter of a negative-U site to the wide electronic band with a width W, and is proportional to $W^2/|U|$. Note that the same factor occurs in Eq. (40). As was shown by Bar-Yam the phase transition is triggered by the Bose-Einstein condensation of pairs inside the narrow band, with a condensation temperature proportional to the bandwidth (apart from a logarithmic factor). There is a close relation between this approach, and our analysis where we find J for the scale of the transition to ODLRO. An important difference is that our analysis can be applied more easily to a disordered system of negative-U centers.

The local-pair approach is applicable to a situation where these pairs are stable and have a narrow bandwidth. On the other hand, if $|U| \ll W$ local pairs are unstable and the transition becomes more BCS-like. If there is a negative-U center at every site, the transition can be approximated with the Hartree approach to the negative-U Hubbard model.[39] If the negative-U centers have a larger separation, one effectively has a situation which can also be described with a negative-U Hubbard model with a larger unit cell, and with a value of U which is effectively reduced due to hybridization.

As T_c scales roughly with the order parameter Δ at $T = 0$ one can also regard the vertical axis in Fig. 1 as an approximate measure of the critical temperature. One then observes that in a system with a negative-U center at each site, the off-diagonal long-range ordering temperature (T_{ODLRO}) at half filling is larger than the temperature scale for ODSRO (T_{ODSRO}) for individual sites. There are several mechanisms which suppress T_{ODLRO} compared to T_{ODSRO}: Even in the case of a lattice with a negative-U center at each site a reduced filling of the band reduces the scale of the gap with a factor $2\sqrt{n(1-n)}$, where n is the filling of the band. As we have seen in the above discussion this reduction does not occur for the short-range order parameter, which asymptotically approaches $U/2$. Moreover, if $|U|/W$ is large, $J_{i,j}$ decreases with increasing $|U|$, leading to a strong suppression of long-range order. Also if the negative-U impurities are not on nearest-neighbor sites (say, on every second atom), the coupling between the negative-U centers is reduced, leading to a reduction of T_{ODLRO}.

In Fig. 8 the phase diagram is drawn for illustrative purposes. In the limit of weak coupling a phase transition occurs between the superconducting state with ODLRO and the normal state without any order, whereas for sufficiently strong coupling there exists a region above the bulk superconducting T_c where ODSRO exists without ODLRO. A maximum T_{ODLRO} occurs for $1 < -U/W < 2$. A similar optimum has been recently reported by Denteneer, An, and van Leeuwen using a fluctuation approach to the same problem.[51] The curve separating the nonordered phase from the ODSRO state is calculated for a single negative-U impurity; it could be shifted due to the influence of a neighboring negative-U site on T_{ODSRO}. In fact the solid curve on the left-hand side of Fig. 8 marks the onset of ODLRO (which implies ODSRO) according to the weak-coupling formula Eq. (39), outside the region of U where a single negative-U center exhibits off-diagonal order. Hence the presence of neighboring

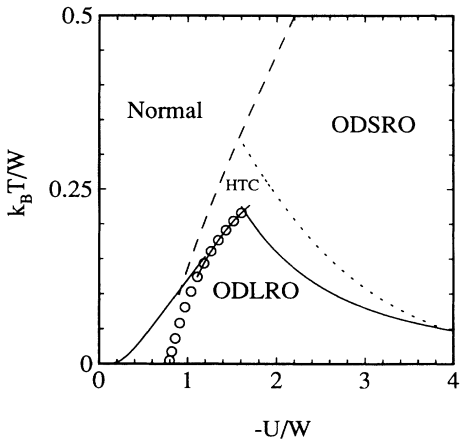

FIG. 8. Phase diagram (Temperature vs coupling parameter) for a system of negative-U impurities. ODLRO indicates the bulk superconducting phase, ODSRO the local off-diagonally ordered state, and HTC the Haldane-Taraphder-Coleman state. The curves were calculated as follows: Eq. (10) (dashed), Eq. (33) (dotted), T_c using the Hartree approximation for the negative-U Hubbard model (Ref. 39) (left-hand section of solid curve), the combination of $k_B T_c = 2.2J$ with Eq. (38) (circles), and Eq. (40) (right-hand section of solid curve). The parameters used are $k_B T_F = 0.9$ and $V_0^{\mathrm{eff}} = 0$.

negative-U impurities tends to enhance ODSRO. On the other hand, at high enough temperatures where there is no long-range order, the contributions to Ψ of the various neighboring impurites as expressed in Eq. (35) tend to cancel as the phases add up randomly. Hence in lowest order the influence of neigboring negative-U impurities on T_{ODSRO} tend to cancel. Also, for large $-U/W$ the proximity-induced order parameter due to neigboring impurities is proportional to $1/|U|$. Therefore on the right-hand side of Fig. 8 the dashed curve is a good estimate of T_{ODSRO}. On the left-hand side T_{ODSRO} is never reached, because it is *smaller* than T_{ODLRO}. As long-range order implies short-range order, we anticipate that, if a full calculation could be carried out, the dashed curve would merge into the solid curve rather than crossing it.

Also indicated in this plot is the Haldane-Taraphder-Coleman state. The boundary between the Haldane-Taraphder-Coleman state and ordinary ODSRO is not sharp, as in the magnetic impurity problem where a gradual crossover from the Kondo state to spin fluctuations occurs. The ODSRO phase is rather peculiar, as it should exhibit large deviations from ordinary Fermi-liquid behavior: As part of the charge carriers is in a paired state, the statistical properties are no longer purely fermionic. In particular one expects anomalous scattering from the localized isospins, which should strongly affect the transport properties, however, without resulting in a superfluid state. In fact, it has been shown by Alascio *et al.*[52] using an expression for the imaginary part of the self energy obtained by Eliashberg,[53] that a system of electrons in contact with a narrow band of local pairs provides the form of the self energy such as was postulated by Varma *et al.*[17] As has been shown by Bar-Yam,[23] the decay rate of the local-pair subsystem also has a linear frequency and temperature dependency. In addition, one expects an anomalous contribution to the resistivity due to Andreev scattering[44] at the localized isospins. Upon increasing the temperature the local order decreases, due to which one expects this anomalous scattering to become smaller.

E. η Pairing

In a recent paper Singh and Scalettar argued that *in the presence of spin asymmetry* for bipartite negative-U lattices in arbitrary dimensions $d \geq 2$ one obtains an η-paired superconducting ground state at exact half filling.[54] This was shown by combining two sources of information: (1) the existence of an exact mapping of the negative-U Hubbard model on the repulsive large-U Hubbard model[55–59] and (2) the result by Nagaoka and Thouless,[60,61] that the latter model gives rise to a fully aligned ferromagnetic ground state slightly away from half filling. In the case of the high-T_c cuprates the underlying electronic structure of a doped Mott-Hubbard insulator provides a mechanism for antiferromagnetic correlations,[62–65] and hence for spin asymmetry between the two sublattices. Anderson argued that the Hamiltonian of a doped Mott-Hubbard insulator also gives rise to an electronic pairing mechanism,[66] resulting in a fairly complete microscopic framework for the situation considered by Scalettar and Singh. The η pairing corresponds to a staggered order parameter with opposite phases on the two sublattices, similar to an XY antiferromagnet. Our analysis does not include a spin asymmetry, and moreover treats the problem on the mean-field level, which may not be sufficient to obtain the antiferromagnetic coupling between neighboring sites. In a phenomenological way one could also *define* a gauge field, which rotates the phases such as to provide the antiferromagnetic coupling. In principle this could give rise to anomalous statistics, both of the Cooper pairs and of the quasiparticles as has been argued for the normal state of a doped Mott-Hubbard insulator,[66–69] however, for a half-filled band we obtain no statistical anomaly using this concept.

IV. CONCLUSIONS

We conclude that in the regime of a strong local effective attraction between electrons off-diagonal short-range ordering can occur reminiscent of a local pair. The interaction has to exceed a certain critical value before this phenomenon occurs. A fascinating phase diagram occurs, which bears a resemblance to the phase diagram of magnetic metals. A lattice of negative-U impurities effectively behaves as an array of proximity coupled Josephson junctions, but has a number of additional features remiscent of the Kondo problem. The latter Haldane-Taraphder-Coleman state is characterized by a mixture of paired and unpaired electrons. The anomalous scattering of unpaired electrons at off-diagonally ordered local moments can give rise to strong deviations from ordinary Fermi-liquid behavior.

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