Mean-field theory for vacancies in a quantum antiferromagnet

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(Received 31 May 1989)

The vacancies in a quantum antiferromagnet have dipolar interactions mediated by antiferromagnetic (AF) spin waves. An effective Hamiltonian incorporating this effect is studied within a mean-field approximation. The ground state is found to be an s- or d-wave superconductor. In the strong-coupling regime the superconductivity coexists with the incommensurate spiral AF order. The analysis is limited to the regime where the AF correlation length is larger than the distance between vacancies.

The observation of antiferromagnetic correlations[1] and superconductivity[2] in CuO-based compounds has made it important to learn more about the effect of antiferromagnetic (AF) correlations on the charge carriers in CuO planes. Starting with a Hubbard model for electrons with on-site repulsion at the density close to one per site,[3] one arrives at the problem of mobile vacancies in a quantum antiferromagnet described by a "t-J" Hamiltonian

$$H_0=-t\sum_{r,a}\bar{c}_{r+a}c_r+J\sum_{r,a}\mathbf{s}_r\cdot\mathbf{s}_{r+a}\ ,$$

where $a=\hat{x},\hat{y}$, the spin-$\frac{1}{2}$ fermion operator $c_{r,\sigma}$ is restricted to single occupancy, and $\mathbf{s}_r=\bar{c}_r\hat{\tau}c_r$ is the local spin.

Our analysis here is based on the assumption that the AF order, which is known[4] to be present in the ground state of H_0 at zero vacancy concentration $n=0$, persists for small $n\ll 1$ at least within a finite AF correlation length ξ. We assume that ξ is larger than the intervacancy distance and hence approach the problem from the AF-ordered side. Since the AF correlation length (at T not too high) is determined by n, our assumption can be verified *a posteriori*; in Ref. 5 we have argued that $\xi\sim n^{-1}$, and hence in the $n\to 0$ limit is much larger than the vacancy Fermi wavelength $k_F^{-1}\sim n^{-1/2}$.

From earlier work[6–9] we know that the ground state of a single vacancy in an antiferromagnet lies at the center of the magnetic zone face and involves a dipolar distortion[7] of the staggered magnetization. The latter leads to long-range dipolar interactions between the vacancies. In Ref. 5 we have argued that for $0<n\ll 1$, this interaction leads to the ordering of the AF dipole moments associated with the vacancies, which in turn leads to incommensurate spiral AF order of the background spins described by

$$\langle\hat{\Omega}\times\partial_a\hat{\Omega}\rangle=\mathbf{P}_a\neq 0$$

with $|\mathbf{P}_a|\sim n$.

Another consequence of the dipolar interactions is superconducting pair correlation, which we explore in the following. We shall do that on the basis of a phenomenological Hamiltonian for the vacancies with four-fermion interactions. This H_{eff} incorporates the low-frequency part of the spin-wave mediated interaction between vacancies and can be derived from the "t-J" Hamiltonian H_0 (at least in the perturbative parameter range, $t\ll J$) by integrating out the spin waves. (We emphasize that the interaction originates from the fluctuations of the direction, rather than the magnitude, of the staggered magnetization.) For this effective Hamiltonian we find, within the mean-field approximation, that the ground state is an s- or d-wave superconductor with the gap Δ scaling with the Fermi energy (which for $t/J\gg 1$ and low vacancy density $n\ll 1$ is of order nJ). The s- and d-wave channels are degenerate in the low-density limit because of the structure of the Fermi sea (which consists of four valleys). We shall define a phenomenological coupling constant g characterizing dipolar interactions in the following. The weak coupling limit, $g\ll g_c\approx 1$, is BCS-like with $\Delta\sim nJ\exp(-2/g)$. For $g>g_c$ the ground state acquires a dipole polarization corresponding to the spiral AF state as expected on the basis of the one-particle picture of Ref. 5. The superconductivity, however, persists in the polarized phase as well.

In the following we shall introduce the effective Hamiltonian and present the mean-field theory results. To derive H_{eff} we factorize[7] the electron operator (constrained to single occupancy): $c_{r,\sigma}=\bar{\psi}^{\alpha}_r z^{\alpha}_{r,\sigma}$, where $\bar{\psi}^{\alpha}_r$ creates a vacancy on site r which belongs to sublattice $\alpha=A,B$ and $z^{\alpha}_{r,\sigma}$ is the Schwinger spin boson. The explicit sublattice index is introduced because in the spin-wave ground state the unit cell is doubled; more generally it can be argued that α labels the spin of the vacancy. (The ground state of a vacancy has the total spin $\frac{1}{2}$.) In the spin-wave approximation we take

$$z_r^A=(1-\tfrac{1}{2}a_r^{\dagger}a_r,a_r)$$

and

$$z_r^B=(b_r,\ 1-\tfrac{1}{2}b_r^\dagger b_r) .$$

Substituting into H_0 one obtains

$$H_{\rm SW}=-4tN^{-1}\sum_{k,q}\bar\psi_k^A\psi_{k+q}^B(\gamma_k a_q^\dagger+\gamma_{k+q}b_{-q})+\text{H.c.}$$
$$+4J\sum_k[a_k^\dagger a_k+b_k b_k^\dagger+\gamma_k(a_k b_{-k}+\text{H.c.})] , \qquad (1)$$

with

$$\gamma_k\equiv\tfrac{1}{2}(\cos k_x+\cos k_y) .$$

Integrating out the spin waves yields a four-fermion interaction, the static part of which is

$$H_{\rm int}=-g_0N^{-2}\sum_{k,k',q}V(k,k',q)\bar\psi_k^A\psi_{k+q}^B\bar\psi_{k'+q}^B\psi_{k'}^A , \qquad (2)$$

where $g_0=2t^2/J$ is the coupling constant, and the interaction has the form

$$V(k,k',q)=(1-\gamma_q)^{-1}(\gamma_k-\gamma_{k+q})(\gamma_{k'}-\gamma_{k'+q})$$
$$+(1+\gamma_q)^{-1}(\gamma_{k+q}\gamma_{k'}+\gamma_{k'+q}\gamma_k) . \qquad (3)$$

In the effective Hamiltonian it is convenient to *explicitly* include a one-particle dispersion term $\varepsilon_k\bar\psi_k^\alpha\psi_k^\alpha$. This band energy term arises as a zero frequency part of the fermion self-energy[10,8] as derived from Eq. (1); alternatively it appears in the Hartree-Fock factorization of Eq. (2). It has been observed[6–9] that the minimum of ε_k occurs at the face centers of the magnetic zone boundary (see Fig. 1). Although various approximations[6–9] indicate that the band is strongly anisotropic, being relatively flat along the zone face, in the following, for the sake of simplicity, we will take $\varepsilon_k\approx\frac{1}{2}m^{-1}(k-k_\nu)^2$ in each of the four valleys $k_\nu=(\pm\pi/2,\pm\pi/2)$. In the perturbative regime $t/J\ll1$, the bandwidth m^{-1} is of order t^2/J. (In the strong-coupling limit $t/J\gg1$ it has been argued[8] that $m^{-1}\sim J$; one also expects the same scaling for the renormalized coupling constant, i.e., $g_0\sim J$.) Next we observe that for low density of vacancies only the states close to the zone face centers are important, and therefore we can approximate $V(k,k',q)$ assuming that $k,k',k+q,k'+q$ are all close to k_ν. Since γ_k vanishes on the zone boundary, the second term in Eq. (3) is small compared to the first,[11] and the interaction potential is effectively

$$V(k,k',q)\approx\tilde V(k,k',q)=\frac{[\sin(\frac{1}{2}q_a)\sin(k_a+\frac{1}{2}q_a)][\sin(\frac{1}{2}q_{a'})\sin(k'_{a'}+\frac{1}{2}q_{a'})]}{(\sin\frac{1}{2}q_a)^2(\cos\frac{1}{2}q_{a'})^2} , \qquad (4)$$

which has the dipolar character. The latter is not surprising in view of the dipolar nature of the coupling of the vacancy to the transverse distortions of the staggered magnetization.[5] Finally, with all the aforementioned remarks, the effective Hamiltonian takes the form

$$H_{\rm eff}=\sum_{k,\alpha=A,B}(\varepsilon_k-\mu)\bar\psi_k^\alpha\psi_k^\alpha-gN^{-2}\sum_{k,k',q}\tilde V(k,k',q)\psi_k^{\bar A}\psi_{k+q}^B\bar\psi_{k'+q}^B\psi_{k'}^A , \qquad (5)$$

where we have chosen m^{-1} as the energy unit, defined a dimensionless coupling constant $g\equiv mg_0$, and introduced the chemical potential μ. Although our derivation implies $g\sim O(1)$ in both weak and strong coupling, we shall take a broader view, interpreting Eq. (5) as a phenomenological Hamiltonian and exploring the behavior it describes as a function of $g>0$. Our analysis will be the straightforward mean-field (MF) approximation,[12] and will differ from the BCS theory only by virtue of the appearance of the dipolar polarization[5] (or spiral) order parameter.

Let us proceed by introducing the superconducting order parameter

$$\Delta_k\equiv g\sum_{k'}{}'\Gamma_{kk'}\zeta_{k'}\equiv g\sum_{k'}{}'\tilde V\left[\frac{k+k'}{2},\frac{k+k'}{2},k-k'\right]\langle\bar\psi_{k'}^A\bar\psi_{-k'}^B\rangle \qquad (6)$$

(where $\zeta_k\equiv\langle\bar\psi_k^A\bar\psi_{-k}^B\rangle$ and the prime on the sums denotes that they are normalized by the number of sites N). The dipolar polarization order parameter is

$$Q_k\equiv-g\sin(k_a)Q_a\equiv-g\sin k_a\sum_{k'}{}'\sin k'_a\langle\bar\psi_{k'}^A\psi_{k'}^B\rangle . \qquad (7)$$

The MF Hamiltonian then has the form

$$H_{\rm MF}=\sum_{k,\alpha}\varepsilon_k\bar\psi_k^\alpha\psi_k^\alpha+\sum_k Q_k\bar\psi_k^B\psi_k^A-\sum_k\Delta_k\psi_k^B\psi_{-k}^A+\text{H.c.}+g^{-1}|Q_a|^2+\tfrac{1}{2}\sum_k(\zeta_k\bar\Delta_k+\text{H.c.}) . \qquad (8)$$

It is convenient to write $H_{\rm MF}$ in the form

$$H_{\rm MF}=\tfrac{1}{2}\bar\Phi M\Phi+\text{const}$$

in terms of a four-component operator

$$\Phi_k=(\psi_k^A,\ \psi_k^B,\ \bar\psi_{-k}^A,\ \bar\psi_{-k}^B)$$

with the definition

$$M\equiv\begin{bmatrix}\varepsilon_k & \bar{Q}_k & 0 & -\bar{\Delta}_{-k}\\ Q_k & \varepsilon_k & \bar{\Delta}_k & 0\\ 0 & \Delta_k & -\varepsilon_k & Q_k\\ -\Delta_{-k} & 0 & \bar{Q}_k & -\varepsilon_k\end{bmatrix}. \quad (9)$$

The matrix M has two pairs of eigenfrequencies:

$$\omega_\pm^2(k)=\{[\varepsilon_k^2+|\Delta_k^t|^2+|Q_k|^{-2}(\mathrm{Re}\Delta_k^s\bar{\Delta}_k^t)^2]^{1/2}\pm|Q_k|\}^2+|\Delta_k^s|^2-|Q_k|^{-2}(\mathrm{Re}\Delta_k^s\bar{\Delta}_k^t)^2 , \quad (10)$$

where $\Delta_k^s\equiv(\Delta_k+\Delta_{-k})/2$ is the s- or d-wave (depending on the assumed symmetry with respect to rotation of k) singlet superconducting parameter, while $\Delta_k^t\equiv(\Delta_k-\Delta_{-k})/2$ is the p-wave triplet. Although we have carried out the analysis of the general case, since in the end we found that $\Delta^t=0$ for all g, to simplify the presentation, we restrict ourselves to the case of the pure singlet right away. In the latter case the form of $\omega_\pm(k)$ is greatly simplified.

The quasiparticle operators χ_k^σ (with $\sigma=\pm$) corresponding to the positive frequencies have the form

$$\chi_k^\sigma=\frac{1}{\sqrt{2}}u_\sigma(k)(\psi_k^A-\sigma\psi_k^B)+\frac{\sigma}{\sqrt{2}}v_\sigma(k)(\bar{\psi}_{-k}^A+\sigma\bar{\psi}_{-k}^B) , \quad (11)$$

where

$$u_\sigma(k)=(2)^{-1/2}[1+\omega_\sigma^{-1}(\varepsilon_k+\sigma|Q_k|)]^{1/2}$$

and

$$v_\sigma^2(k)=1-u_\sigma^2(k) .$$

We have

$$H_{\mathrm{MF}}=\sum_{k,\sigma}\omega_\sigma(k)(\bar{\chi}_k^\sigma\chi_k^\sigma-\tfrac{1}{2})+\sum_k\varepsilon_k+g^{-1}|Q_a|^2+\tfrac{1}{2}\sum_k(\zeta_k\bar{\Delta}_k+\mathrm{H.c.}) . \quad (12)$$

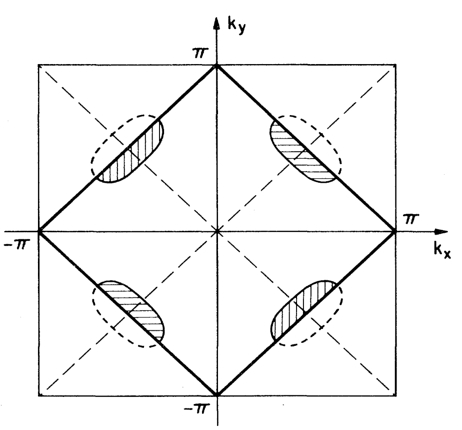

FIG. 1. The structure of the reduced Brillouin zone and the four valleys of the Fermi sea.

The MF ground state is annihilated by the quasiparticle operator $\chi_k^\sigma|0\rangle=0$. The self-consistent values of the order parameters are obtained by minimizing $\langle 0|H_{\mathrm{MF}}|0\rangle$ with respect to Q_a and ζ_k. The MF equations are

$$\Delta_{k'}=\frac{g}{4}\sum_k\Gamma_{kk'}\Delta_k[\omega_+^{-1}(k)+\omega_-^{-1}(k)] , \quad (13)$$

$$Q_{k'}=\tfrac{1}{4}\sin k_a'\sum_k\sin k_a\frac{Q_k}{|Q_k|}\left[\frac{\varepsilon_k-|Q_k|}{\omega_-(k)}-\frac{\varepsilon_k+|Q_k|}{\omega_+(k)}\right] . \quad (14)$$

The chemical potential μ, which has been absorbed into a shift of ε_k, is fixed by the density constraint

$$n=\tfrac{1}{2}\sum_k\left[2-\frac{\varepsilon_k-|Q_k|}{\omega_-(k)}-\frac{\varepsilon_k+|Q_k|}{\omega_+(k)}\right] . \quad (15)$$

To further simplify the analysis we neglect the dependence of the order parameters on k within the valleys defining $\Delta_\nu\approx\Delta_{k_\nu}$ and $Q_\nu\approx Q_{k_\nu}$, which is reasonable when the number of vacancies is small. Assuming $\Delta_\nu=0$ yields exactly the result of Ref. 5, i.e., the dipolar polarization appears[11] for $g\geq 1$ and $Q_\nu=\hat{e}_\nu(\pi g/2)n$ where $\hat{e}=\hat{x}$ or $\hat{y}$. More generally one finds in the weak-coupling regime $g<g_c$ the unpolarized state $|Q_a|=0$, and the BCS-like superconducting gap

$$\Delta_\nu=\Delta=\sqrt{2\pi n E_{\mathrm{co}}}\,e^{-g^{-1}} , \quad (16)$$

where s-wave symmetry was assumed and $E_{\mathrm{co}}\sim O(\mu)$ is some cutoff energy (since on physical grounds only the states not too far from the Fermi surface can contribute). Note that because in the low-density limit $\Gamma_{kk'}$ couples only the *opposite* valleys of the Fermi sea, one is free to choose the phase of Δ_ν in the two orthogonal directions. Hence the result of Eq. (16) also holds for d-wave symmetry of the gap $\Delta_{x+y}=-\Delta_{x-y}=\Delta$. (The degeneracy of s and d channels should be lifted in the next order in n where the coupling between valleys rotated by $\pi/2$, in the Brillouin zone appears.) The transition to the polarized state occurs for $g=g_c$,

$$g_c=1+2(\pi n)^{-1}E_{\mathrm{co}}\exp(-2g_c^{-1}) .$$

For $g > g_c$ one finds

$$\Delta^2 = \frac{4E_{\text{co}}^2}{g-1} e^{-4g^{-1}} \tag{17}$$

and

$$Q_a = \hat{e}_a \frac{\pi g}{s} n \left[1 - \frac{\Delta^2}{(\pi n)^2 (g-1)} \right] . \tag{18}$$

As shown in Fig. 2, there is a discontinuity of $d\Delta/dg$ at $g = g_c$. The chemical potential for $g > g_c$ is given by $\mu = \pi n(1 - \frac{1}{2}g)$. Note that the chemical potential becomes negative for $g > 2$, which signals an instability towards phase separation of holes. The latter, however, is unlikely because of the long-range Coulomb interactions; instead we expect a transition from the state with spiral order (at $T = 0$) to a disordered state.

It is straightforward to redo the calculation for the case of finite temperature; however, it is evident that the transition into the "spiral" state ($Q \neq 0$) occurs at $T_S \sim |Q_a| \sim nm^{-1}$, while the transition to the superconductor occurs at $T_{\text{SC}} \sim m^{-1}\Delta$; hence Fig. 2 is also a caricature of the g, T phase diagram. We emphasize that the relevant energy scale for both transitions is just the Fermi energy $\mu \sim nm^{-1}$ (i.e., nJ in the $t/J \gg 1$ limit). Our crude analysis, however, did not include effects due to the finiteness of the AF correlation length ξ, which decreases as the density of vacancies increases (e.g., in Ref. 5 we argued that for the spiral state $\xi \sim n^{-1}$). The finite correlation length acts to cut off the dipolar interaction and therefore reduces Δ, so that within the present mechanism superconductivity would disappear for larger n when $\xi^{-1}(n)$ becomes smaller than k_F. The competition between the effect of increasing μ and decreasing ξ, which occurs as n increases, is clearly crucial to understand. Unfortunately, the study of the disordered AF limit is clearly beyond the simple spin-wave approach taken up in this paper. Another unresolved issue involves the energy scales. If the spin-wave integration is limited to $k > \xi^{-1} \sim n$, the corresponding energy scale is crudely $\omega > \Gamma \sim J\xi^{-1}$, and therefore leaves little phase space with energy below $\varepsilon_F \sim nJ$. The Fermi energy, however, might in reality be larger than the "t-J" model estimate used earlier, because of the contribution of the direct hopping processes.

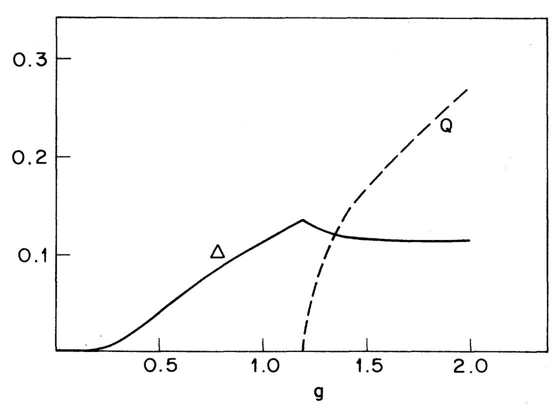

FIG. 2. The superconducting order parameter Δ and the spiral order parameter Q as a function of the phenomenological coupling constant g for the vacancy density $n = 0.1$.

In summary, we have demonstrated that spin-wave mediated dipolar interactions may lead to s-wave as well as d-wave superconductivity of vacancies in a quantum AF magnet. While the competition between the two is resolved by the finite density effects which were not presently considered, we note that in either case because of the multivalley structure of the vacancy Fermi sea there are no nodes of the gap on the Fermi surface. Our conclusion is to be contrasted with the notion that the repulsive interaction in the Hubbard-like model of correlated electrons may lead to d-wave pairing only. The latter is based on an analysis[13,14] which considers the interaction via AF *amplitude* fluctuations of the paramagnetic state which are diffusive as opposed to the propagating collective modes of the *ordered* AF state which were studied here. The idea of this paper is closer in spirit to that of Schrieffer, Wen, and Zhang,[15] who also consider the AF-ordered state. The crucial difference, however, is that we find the vacancies to interact through the long-ranged *transverse* spin waves rather than short-range amplitude modes as in the "spin-bag" scenario of Ref. 15. The short-range interaction (which may be thought of as the result of vacancies sharing "strings" of overturned spins,[16] and can be studied in the Ising limit), however, may also be important, especially since we are interested in a two-dimensional model. Superconductivity due to dipolar interactions was previously discussed qualitatively by Aharony *et al.*[17] in the context of the extended Hubbard model with holes in the σO orbital band. Finally, our analysis suggests that vacancy superconductivity may coexist with AF order (provided the carriers stay mobile) and should be enhanced if for fixed carrier density (and ε_F) the AF correlation length could be increased. On the other hand, our discussion obviously fails to explain the experimental correlation between the appearance of superconductivity and the *disappearance* of the long-range AF order in CuO-based materials as well as the apparent *nonexistence* of superconductivity in doped AF systems of higher spin. Clearly, a better understanding of the disordered state of the doped AF is needed.

ACKNOWLEDGMENTS

It is a pleasure to acknowledge discussions with W. Brinkman, P. Hohenberg, P. Lee, and S. Shastry, and the stimulating environment of the Aspen Workshop on Correlated Electron Systems where this work was initiated. E. D. S. acknowledges support of the Guggenheim Foundation, National Science Foundation (NSF) Grant No. DMR8314625, and the hospitality of AT&T Bell Laboratories.

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