Mobile Vacancies in a Quantum Heisenberg Antiferromagnet

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(Received 5 February 1988)

The ground state of a quantum vacancy in a 2D antiferromagnet is found to involve a long-range dipolar distortion of the staggered magnetization. An effective Hamiltonian of vacancies interacting with long-wavelength spin waves is derived. The implications for the anitferromagnetic long-range order are discussed.

PACS numbers: 75.10.Jm, 71.30.+h, 74.65.+n

The experimental observation of 2D antiferromagnetic (AF) ordering in La_2CuO_4 and related compounds[1,2] has posed a number of puzzling questions concerning the behavior of the correlation length and the dynamical structure function as functions of temperature and doping. While the experimentally observed correlation length for the most pure crystals is consistent with a 2D Heisenberg model,[3] doping or nonstoichiometry has a strong effect which is not understood. It is natural to associate this effect with mobile vacancies introduced into 2D Cu-O planes by doping. The interesting question is to understand how a very small (much below the percolation limit) concentration of holes in the 2D antiferromagnet could limit the correlation length.

Our starting point is the Hamiltonian combining Heisenberg exchange and kinetic energy of the mobile "holes" in a 2D square lattice of spins,

$$H=-t\sum_{\langle ij\rangle}c_{i,\sigma}^{\dagger}c_{j,\sigma}+\frac{J}{2}\sum_{\langle ij\rangle}c_{i,\sigma}^{\dagger}\hat{\tau}_{\sigma,\sigma'}c_{i,\sigma'}\cdot c_{j,\nu}^{\dagger}\hat{\tau}_{\nu,\nu'}c_{j,\nu'}, \qquad (1)$$

where the c_i's are electron creation/annihilation operators constrained to single occupancy and $\hat{\tau}$ is a Pauli matrix, so that the second term is just the Heisenberg spin exchange. Such a Hamiltonian, it may be argued, captures the essential physics of the Hubbard model in the large on-site repulsion limit $J/t \ll 1$ near half filling and is a natural starting point[4] for describing a doped AF insulator. Below we shall study the interaction of the holes with long-wavelength AF spin waves described by the nonlinear σ model.[5] We will show that the spin current of the hole couples to the magnetization current carried by the AF background spins, resulting in the appearance of a nontrivial texture in the AF order parameter. We find that the spin configuration induced by the mobile hole has a dipolar symmetry (is "roton"-like) and the distortion of the direction of the staggered magnetization is long ranged and decays as r^{-1}. This effect contributes to the destruction of the long-range AF order by vacancies. It leads, at zero temperature (and with the assumption of a quenched distribution), to an algebraic decay of spin correlations, $r^{-3\pi p^2 n}$, with the exponent proportional to the density of holes.

A mobile hole interacts strongly with an AF-ordered spin background as is most apparent in the Ising limit $J_\perp=0$, $J_z=J$, where a state consisting of a string of overturned spins is formed.[6-8] Schematically, the hole state can be sought in the form[7]

$$|\psi_\nu(k)\rangle=\sum_{j,\sigma}\chi_{j,\sigma}(k,\nu)\{c_{j,\sigma}+\sum_{\hat{\mathbf{a}},\sigma'}\gamma_{\hat{\mathbf{a}}}(k)c_{j+\hat{\mathbf{a}},\sigma}c_{j+\hat{\mathbf{a}},\sigma'}^{\dagger}c_{j,\sigma'}+\cdots\}|0\rangle, \qquad (2)$$

with $\hat{\mathbf{a}}=\pm\hat{\mathbf{x}},\pm\hat{\mathbf{y}}$. While $\chi_{j,\sigma}(k,\nu),\gamma_{\hat{\mathbf{a}}}(k)$ are simple in the Ising limit,[7] in the isotropic case, $J_\perp=J_z$, Eq. (2) remains a reasonable variational form that can be used in conjunction with the spin-wave theory.[9] The ground state with given momentum k is quite generally twofold degenerate and can be labeled by ν. The staggered magnetization halves the Brillouin zone, but translation by one lattice vector followed by a global spin flip remains a symmetry. Each "spin" state ν clearly resides preferentially on the sublattice with the same spin direction.

The wave function of Eq. (2) also arises perturbatively for t, $J_\perp \ll J_z$.[9,10] The most direct effect of spin fluctuations ($J_\perp\neq 0$) is to give the hole a finite bandwidth $\sim t^2J_\perp/J_z^2$, and, as first noted by Elser and Huse[10] and Trugman,[8] to shift the crystal momentum of the ground state to the edge of the reduced zone. The effective mass is strongly anisotropic and the energy can be roughly approximated along each of the faces of the zone by $E_{\hat{\mathbf{n}}}(\mathbf{k})=E+[\hat{\mathbf{n}}\cdot(\mathbf{k}-\mathbf{k}_0)]^2/2\mu$ where $\hat{\mathbf{n}}=(\pm1,\pm1)/\sqrt{2}$ and $\mathbf{k}_0=(\pm\pi/2,\pm\pi/2)$.

The perturbation theory, however, does not adequately describe the behavior of the continuous and isotropic ($J_z=J_\perp$) spin background at large scales and low energies. The latter can be understood semiclassically and leads to interesting new phenomena. Specifically, the hole, which retains quantum nature, generates a long-ranged distortion in the direction of the staggered magnetization, $\hat{\Omega}$, and a localized canting of the sublattices leading to a (small) magnetization, $\mathbf{m}$. To get some idea about the basic physics, we will consider the hopping term of Eq. (1) in the presence of the classical spin field, which we make explicit by factorizing the electron operator c_σ (constrained to single occupancy) as $c_\sigma = \psi^\dagger_{(a)} w^{(a)}_\sigma$ where $\psi_{(a)}$ creates a spinless fermionic hole residing on sublattice $\alpha = A,B$, and the spinor $w^{(a)}_\sigma$ is a Schwinger spin boson ($\mathbf{s} = \frac{1}{2}\bar{w}\hat{\tau}w$, $\bar{w}w = 1$). The explicit sublattice index α, redundant on the lattice, is essential for a smooth continuum limit, where α acts as the spin label for the hole (N.B. above). We can represent the semiclassical background spin distortion as a slowly varying SU(2) rotation of the Néel state[11] $w^{(a)}(r) = \mathbf{R}_r \hat{\mathbf{e}}_a$ where $\hat{\mathbf{e}}_A = (1,0)$, $\hat{\mathbf{e}}_B = (0,1)$.

The relation of the globally defined $\mathbf{R}_r$ to the local order parameter and magnetization can be understood by the consideration of

$$\mathbf{R}_r \mathbf{R}^\dagger_{r+\hat{a}} = \exp[\tfrac{1}{2} i\hat{\tau}\cdot(\hat{\Omega}\times\Delta_a\hat{\Omega} + A_a\hat{\Omega} + \kappa^{-1}\hat{\Omega}\times\mathbf{m})], \tag{3}$$

where $\Delta_a \approx \hat{\mathbf{a}}\cdot\nabla$ is discrete gradient along $\hat{\mathbf{a}}$, so that the first factor represents the slow twisting of the order parameter $\hat{\Omega}$. The second term is a relative rotation about the local $\hat{\Omega}$ axis[12] accompanying a finite rotation $\mathbf{R}$ (see below). Finally, the $\mathbf{m}\times\hat{\Omega}$ term represents the staggered component of our rotation which generates a small relative canting of the sublattices yielding net magnetization $\mathbf{m}$ (κ is the amplitude of the local staggered moment). We assume slow gradients and small magnetization $\partial\hat{\Omega} \sim m \ll 1$.

The hopping term involves $\mathbf{R}^\dagger_r \mathbf{R}_{r+a}$ which of course is related to Eq. (3) by a rotation, but is more conveniently parametrized by two spinors z_σ and P_σ (constrained by $\bar{z}z = 1$ and $\bar{p}z = 0$) in terms of which $\mathbf{m} = \frac{1}{2}\bar{p}\hat{\tau}z + \text{H.c.}$ and $\hat{\Omega} = \bar{z}\hat{\tau}z$. Passing to the continuum, $\mathbf{R}^\dagger_r \mathbf{R}_{r+\hat{a}} - 1 \approx i\mathbf{A}_a\tau_z + i\mathbf{K}_a + i\kappa^{-1}\mathbf{M}$ with $A_a = iz\,\partial_a\bar{z}$ and

$$\mathbf{K}_a = \epsilon_{\sigma\nu}\begin{bmatrix} 0 & i\bar{z}_\sigma\partial_a\bar{z}_\nu \\ -iz_\sigma\partial_a z_\nu & 0 \end{bmatrix}, \quad \mathbf{M} = \epsilon_{\sigma\nu}\begin{bmatrix} 0 & i\bar{p}_\sigma\bar{z}_\nu \\ -ip_\sigma z_\nu & 0 \end{bmatrix}, \tag{4}$$

where $\epsilon_{12} = -\epsilon_{21} = 1$. This CP^1 parametrization[13] will facilitate later calculations.

To leading order, the hopping term reads

$$H_{ke} = t(i\,\partial_a\bar{\psi}\mathbf{K}_a\psi + \text{H.c.}) + 4t\bar{\psi}\mathbf{M}\psi.$$

Its physical content becomes more familiar if we rotate the hole wave function along the direction of staggered magnetization defining $\Psi = \mathbf{R}\psi$. Noting that $\mathbf{RMR}^\dagger = \hat{\tau}\cdot\mathbf{m}$ and $\mathbf{RK}_a\mathbf{R}^\dagger = \hat{\tau}\cdot\hat{\Omega}\times\partial_a\hat{\Omega}$, one finds

$$H_\psi = (t/\rho J)\mathbf{j}_a\cdot(i\,\partial_a\bar{\Psi}\hat{\tau}\Psi + \text{H.c.}) + 4t\mathbf{m}\cdot\bar{\Psi}\tau\Psi, \tag{5}$$

where $\mathbf{j}_a \equiv \rho J\hat{\Omega}\times\partial_a\hat{\Omega}$ is the magnetization current ($\partial_t\mathbf{m} = \partial_a\mathbf{j}_a$). The second terms couples the spin with background magnetization and is responsible for the familiar[6,14] polaron formation instability. The first term represents the coupling of the spin current carried by the hole to the magnetization current carried by the background and leads to some "new" physics: It favors a twist in Ω (i.e., $\mathbf{j}_a \neq 0$) whenever the hole hops. The distortion of the spin background driven by the linear terms [Eq. (5)] is saturated by the exchange energy which for the long-wavelength limit is given in terms of $\mathbf{m}$, and $\hat{\Omega}$ by the nonlinear σ Hamiltonian[5]

$$H_\sigma = \frac{J}{2}\int d^2r[\chi^{-1}m^2 + \rho j_a^2], \tag{6}$$

where χ is the susceptibility and ρ is the spin-wave stiffness. The dynamics of the long-wavelength spin field is restored by our taking $[\Omega^i_r, m^j_{r'}] = i\delta(r-r')\epsilon_{ijk}\Omega^k_r$, $[m^i_r, m^j_{r'}] = i\delta(r-r')\epsilon_{ijk}m^k_r$, and $[\Omega^i,\Omega^j] = 0$, which yields the correct semiclassical equations of motion.

The essential features of the spin texture which forms around a mobile hole can all be derived from the lattice Hamiltonian evaluated for a semiclassical variational wave function. Let

$$|\psi\rangle = \sum_\alpha \psi_\alpha \sum_{r\in\{r_a\}} e^{ik\cdot r}\Phi_a(\{\sigma\})\,|r,\{\sigma\}\rangle, \tag{7}$$

where $|r,\{\sigma\}\rangle$ is a state with a vacancy at site r and a spin configuration labeled by a set of $\sigma = \pm 1$, while the spin wave function $\Phi_a(\{\sigma\})$ may be defined with the vacancy at the origin, since the spin configuration moves "rigidly" with the hole. In the semiclassical limit Φ can be factorized in terms of single spins and becomes a product of spinors

$$[\Phi_a(\{\sigma\}) = \prod_{\rho\neq 0} w^a_{\sigma,\rho}.$$

Elaborating the earlier definition, we have $w^A_{\sigma,\rho} = z_{\sigma,\rho} + \frac{1}{2}p_{\sigma,\rho}$ if $\rho\in A$ and $w^A_{\sigma,\rho} = \epsilon_{\sigma\nu}(z^*_{\nu,\rho} - \frac{1}{2}p^*_{\nu,\rho})$ if $\rho\in B$ (for $\alpha = B$ the sublattices are interchanged). The hopping matrix element becomes (working to first order in $m \sim \partial\Omega$)

$$E_h = t\,\text{Re}\left\{\psi^*_A\psi_B\left[\sum_{\hat{a}} i\epsilon_{\sigma\sigma'}z_{\sigma,-\hat{a}}z_{\sigma',\hat{a}}\sin\left(\hat{\mathbf{a}}\cdot\mathbf{k} - \int d^2r\,\hat{\mathbf{a}}\cdot\mathbf{A}\right) + \tfrac{1}{2}\sum_{\hat{a}}\epsilon_{\sigma\sigma'}(p_{\sigma,-\hat{a}}z_{\sigma',\hat{a}} - z_{\sigma,-\hat{a}}p_{\sigma',\hat{a}})\cos\left(\hat{\mathbf{a}}\cdot\mathbf{k} - \int d^2r\,\hat{\mathbf{a}}\cdot\mathbf{A}\right)\right]\right\}. \tag{8}$$

The spatial integral over the vector potential $\mathbf{A}$ arises from the expansion of the infinite product. The first term here corresponds to the current coupling in the continuum Hamiltonian [Eq. (5)], leads to a twist in $\hat{\mathbf{\Omega}}$, and is dominant for $k \approx (\pi/2,\pi/2)$, while the second term [the spin coupling of Eq. (5)] induces canting or a local magnetization and is most important near the zone center and corners.

Minimizing $\langle \psi | H | \psi \rangle$ numerically without further approximations we found that the direction of $\mathbf{m}_r$ was along, say, $\hat{\mathbf{z}}$ for all r and that Ω_r was planar ($\Omega^x + i\Omega^y = e^{i\phi}$). Except precisely at $k=0$ and the zone corners, $\hat{\mathbf{\Omega}}$ has a dipolar configuration: $\phi = \mathbf{p}\cdot\mathbf{r}/r^2$. For $t<J$ one readily finds analytically the dipole moment $p_a \sim (2t/\pi J)\sin k_a$ which for k on the zone boundary is normal to the zone face. On the other hand, the magnetization which is present for all k other than $(\pi/2,\pi/2)$ decays exponentially with r since the relevant stabilizing term in the nonlinear σ Hamiltonian is $m^2/2\chi$ with no gradients. For $\mathbf{k}=k(1,1)$ the magnetization on the four sites near the hole is isotropic $\sim (t\chi/J)\cos k$ while for $k=(k,\pi-k)$ it has the same magnitude but is quadrupolar: $m(\hat{\mathbf{x}}) = m(-\hat{\mathbf{x}}) = -m(\pm\hat{\mathbf{y}})$. Finally, we find that the minimum of $E(k)$ occurs at the face center $k=(\pi/2,\pi/2)$ (in agreement with other approximations).

The semiclassical calculation can be extended by our including the spin fluctuations in the variational wave function [Eq. (7)] by taking

$$\Phi(\{\sigma\}) = \exp\left[\sum_j h_j\sigma_j\right]\exp\left[-\sum_{\langle ij\rangle} K_{ij}\sigma_i\sigma_j\right]. \qquad (9)$$

The first factor, with the complex local field h_j, is equivalent to the semiclassical factorization used above, while the second, K_{ij}, term builds in pair correlations as in the Marshall AF state.[15] We have carried out numerical minimization with Eq. (9) and arrived at the same conclusions as listed above. Finally, we have also studied the spin correlations of the numerically obtained exact ground state of the vacancy in a eighteen-site cluster[10] and found them in semiquantitative agreement. The details of these calculations will be presented elsewhere.

It is interesting to note that the form of the spin distortion as a function of k given above is consistent with $t, J_\perp \ll J_z$ perturbation theory, as is the semiclassical contribution to the effective mass $\mu \sim J/t^2$. The latter agrees with a purely classical estimate of the kinetic energy of a moving spin texture, which yields $\mu^{-1} \sim Jp^2$. While the dipole moment p is of order t/J for $t/J \ll 1$, for $t>J$ we expect it to saturate, $p \sim 1$, implying a lower bound on the hole mass in the limit $t \gg J$ which scales as J^{-1}.

It is conceivable that for large t/J, textures other than those described above (ferropolarons[6,7,14] aside) become energetically favorable. One plausible texture is the Belyavin-Polyakov[16] soliton which carries an azimuthal magnetization current and therefore couples to the hole. Unfortunately the careful numerical investigation of this limit is frustrated by the finite-size effects.

The existence of a long-range distortion of the staggered magnetization associated with the hole is liable to have a strong effect on the antiferromagnetic long-range order. At low density, n, one expects the holes to be localized[17] and hence it is not unreasonable to assume in that regime that one has a *quenched* random distribution of dipolar textures.[18] The latter leads to an algebraic decay of the spin-correlation function at $T=0$:

$$S(r) \equiv \langle \hat{\mathbf{\Omega}}(r)\cdot\hat{\mathbf{\Omega}}(0)\rangle = -1 + 2\langle |z_\sigma(r)\bar{z}_\sigma(0)|^2\rangle \sim r^{-3\pi p^2 n}, \qquad (10)$$

as can be found by a straightforward calculation[19] closely following conventional Kosterlitz-Thouless analysis.

The dynamics of holes and long-wavelength spin waves can be further investigated by use of an effective Hamiltonian that generalizes the "microscopic" continuum Hamiltonian of Eq. (5). This H_{eff} describes the coupling of the hole quasiparticles to the spin waves in a Born-Oppenheimer approximation and can be derived perturbatively from Eqs. (1) and (5). It has the form

$$H_{\text{eff}} = (2\mu)^{-1}\partial\bar{\Psi}\partial\Psi + g_1\mathbf{j}_a\cdot(i\partial_a\bar{\Psi}\hat{\boldsymbol{\tau}}\Psi + \text{H.c.}) + g_2\mathbf{m}\cdot\bar{\Psi}\hat{\boldsymbol{\tau}}\Psi - (g_3 j_a^2 + g_4 m^2)\bar{\Psi}\Psi, \qquad (11)$$

where μ is the effective mass and the g's are renormalized [cf. Eq. (5)] coupling constants. As written, H_{eff} is rotationally invariant; however, as discussed earlier, the quasihole state lies at the zone boundary and is anisotropic. This effect can be included by the redefinition of $\mu^{-1}\partial_a$ acting on ψ.

Another potentially important consequence of the long-range structure of the hole state concerns the interaction between the holes. A generalization of the semiclassical analysis described above to the case of two holes shows that they have dipolar interactions and, hence, an attractive channel, leading to a singlet, s-like (but anisotropic) bound state. This clearly has interesting implications for superconductivity, which will be discussed elsewhere.

In conclusion, we have found that the quantum motion of the vacancy results in a long-range dipolar twist of the staggered magnetization in the ground state of the hole. We have also derived an effective Hamiltonian [Eq. (11)] describing the coupling of the hole quasiparticle with long-wavelength spin waves. In this paper we have emphasized the semiclassical aspects of the problem, with the spin fluctuations being added either via the Marshall wave function [Eq. (9)] or spin-wave perturbation theory.[11] It is important to stress that while the existence of AF staggered order in the spin state was an essential assumption, the broken symmetry was not: We find dipolar textures in the Marshall variational state (and the ground state of the small cluster with a vacancy) with no sublattice magnetization (total spin $\frac{1}{2}$ for the exact cluster solution).

Finally, while this article was being prepared for publication we learned of work by Wiegman[20] who has also derived a continuum Hamiltonian similar to Eq. (5).

The authors have benefitted greatly from discussions with D. Huse and V. Elser. It is a pleasure to acknowledge stimulating interactions with G. Aeppli, W. Brinkman, D. Fisher, P. Hohenberg, A. Millis, Stephen Shenker, and R. Shankar. Part of this work was done during the visits of one of us (E.D.S.) to AT&T Bell Labs. One of us (E.D.S.) also acknowledges support from the National Science Foundation under Grant No. DMR8314625, and the U.S. Department of Energy under Contract No. DEAC0283ER13044.

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