Static Vacancies on a 2D Heisenberg Spin-1/2 Antiferromagnet

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We study static vacancies on a 2D Heisenberg spin-$\frac{1}{2}$ lattice at $T=0$, using linear spin-wave theory (LSW) and exact diagonalization. Unexpectedly, quantum fluctuations are *reduced* on neighbors of an isolated vacancy. Two vacancies are attractive, with lowest energy as nearest neighbors. We find LSW to be surprisingly accurate relative to exact diagonalization, both done on a 4×4 lattice. However, LSW on larger systems give substantial modification of the 4×4 results for binding and ground-state energies, suggesting the need for larger lattices than previously suspected for reliable numerical estimates.

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There are several reasons for studying the problem of a static vacancy on a 2D Heisenberg square lattice with an antiferromagnetic (AF) nearest-neighbor exchange. In La_2CuO_4, the holes at the planar Cu sites form an AF lattice with a Néel temperature of $T_N \sim 270$ K. There have been suggestions[1] that very small amounts of Sr in La_2CuO_4 lead to almost localized holes in CuO_2 planes. Holes localized around planar copper ions might effectively create static vacancies in the antiferromagnetic lattice. The static-vacancy problem is, then, an important limiting model for understanding the interaction of mobile holes through the antiferromagnetic background. Even more immediately related to this model are recent experiments in which nonmagnetic Zn has been substituted for Cu to probe the magnetic interactions in La_2CuO_4. Finally, the solution of the static-impurity problem contributes to a more complete understanding of quantum Heisenberg antiferromagnets in general. For example, one might have thought that removing a spin from a 2D AF lattice would enhance quantum fluctuations at the sites next to the vacancy. However, the 2D Heisenberg Hamiltonian, as given by

$$H = \tfrac{1}{2} J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j = \tfrac{1}{2} J \sum_{\langle ij \rangle} [S_i^z S_j^z + \tfrac{1}{2}(S_i^- S_j^+ + S_i^+ S_j^-)] , \quad (1)$$

with $J > 0$, has competing interactions in the sense that the longitudinal terms enhance the staggered magnetization (chosen to be in the z direction), while the transverse terms suppress it. Hence, it is not clear which of these predominates when a spin is removed.

In our analysis we employ a linear spin-wave (LSW) theory,[2,3] which uses a bosonic representation of the spins. First, we calculate the exact ground-state properties of the impure infinite lattice within this representation.[4] Concerned by the significant difference between the LSW calculations on an infinite lattice and the exact diagonalization results on a 4×4 lattice, we diagonalized the LSW Hamiltonian for finite lattices. Surprisingly, on a 4×4 lattice we found that the LSW ground-state energy is very close to the exact value, in spite of the low dimensionality and small lattice size, both of which make LSW suspect. As the lattice size is enlarged, the binding and the ground-state energies smoothly converge to those of an infinite lattice, indicating that the finite-size effects in small-cluster calculations might be very important.

In the simple LSW approximation the Heisenberg Hamiltonian, Eq. (1), reduces to

$$H = \tfrac{1}{2} J \sum_{\langle ij \rangle} [-\tfrac{1}{4} + \tfrac{1}{2}(a_i^\dagger a_i + b_j^\dagger b_j) + \tfrac{1}{2}(a_i^\dagger b_j^\dagger - a_i b_j)] , \quad (2)$$

where a ($a^\dagger$) and b ($b^\dagger$) are magnon annihilation (creation) operators, and the site indices i and j refer to A (down) and B (up) sublattices, respectively. The longitudinal spin operators are, then,

$$S_i^z = -\tfrac{1}{2} + a_i^\dagger a_i , \quad S_j^z = \tfrac{1}{2} - b_j^\dagger b_j . \quad (3)$$

For the LSW calculation on an infinite lattice it is convenient to introduce the following time-ordered Green's functions:

$$G_{jj'}(t) \equiv -i\langle T[b_j(t) b_{j'}^\dagger(0)] \rangle , \quad F_{ij}(t) \equiv -i\langle T[a_i^\dagger(t) b_j^\dagger(0)] \rangle . \quad (4)$$

The time Fourier transforms of the pure host Green's functions obey the following equations:

$$G_{jj'}^0(\omega) = \frac{1}{N} \sum_k e^{i\mathbf{k}\cdot(\mathbf{r}_j - \mathbf{r}_{j'})} \frac{\omega + J}{\omega^2 - E_k^2 + i\delta} , \quad (5a)$$

$$F_{ij}^0(\omega) = -\frac{1}{N} \sum_k e^{i\mathbf{k}\cdot(\mathbf{r}_i - \mathbf{r}_j)} \frac{\epsilon_k}{\omega^2 - E_k^2 + i\delta} , \quad (5b)$$

where the spin-wave energy is $E_k = \pm (J^2 - \epsilon_k^2)^{1/2}$, with $\epsilon_k = \frac{1}{2} J(\cos k_x + \cos k_y)$. Here N is the number of sites in a single sublattice and the sum over k is done for the

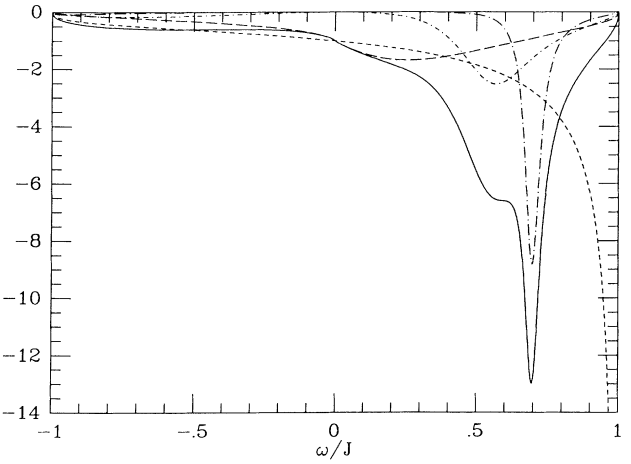

FIG. 1. Imaginary parts of the pure host Green's function $G^0_{jj}(\omega)$ (short-dashed line) and the dressed Green's function $G_{11}(\omega)$ (solid line) and its components $S(\omega)$ (long-dashed line), $P(\omega)$ (dot-short-dashed line), and $D(\omega)$ (dot-long-dashed line). Using $G^0_{ij}(\omega)$ one can calculate $\langle S^z_j\rangle_0=0.3034$. $G_{11}(\omega)$ is given by $G_{11}(\omega)=S(\omega)+2P(\omega)+D(\omega)$. The peaks in the imaginary parts of $S(\omega)$, $P(\omega)$, and $D(\omega)$ are resonances in the magnon excitation spectrum with the corresponding symmetries.

N points in the first (magnetic) Brillouin zone. Taking the limit $N\to\infty$, we convert the sums into integrals, which can be evaluated analytically in terms of elliptic integrals.[5,6] For the particular case of $j=j'$,

$$G^0_{jj}(\omega)=-\frac{2}{\pi}\left(\frac{1+\omega}{1-\omega}\right)^{1/2}[K(1-\omega^2)^{1/2}+iK(|\omega|)], \tag{6}$$

for $-1<\omega<1$ (we set $J=1$), while the imaginary part of $G^0_{jj}(\omega)$ (which is proportional to the density of states) vanishes outside this spin-wave band. The imaginary part of $G^0_{jj}(\omega)$ is graphed in Fig. 1. The average number of magnons per site in the ground state is given by

$$\langle b^\dagger_j b_j\rangle=-\frac{1}{\pi}\int_{-1}^{0}\mathrm{Im}G_{jj}(\omega)\,d\omega. \tag{7}$$

Using Eq. (7), for the pure lattice, we find $\langle b^\dagger_j b_j\rangle_0=0.1966$, which from Eq. (3) is equivalent to the well known result[2] $\langle S^z_j\rangle_0=0.3034$. Similarly, the calculation of $F^0_{ij}(\omega)$ for nearest-neighbor spin pairs yields $\langle S^x_i S^x_j\rangle_0=\langle S^y_i S^y_j\rangle_0=-0.1378$ and the energy of one bond of an infinite lattice, in the LSW approximation, is $\epsilon^0_{ij}=\frac{1}{2}J\times\langle\mathbf{S}_i\cdot\mathbf{S}_j\rangle_0=-0.1645J$.

Now we consider the problem of a static impurity. The effect of removing the down spin at the site we label 0 (see Fig. 2) can be taken into account by the impurity potential

$$V=-\tfrac{1}{2}J\sum_{\delta=1}^{4}\mathbf{S}^B_\delta\cdot\mathbf{S}^A_0, \tag{8}$$

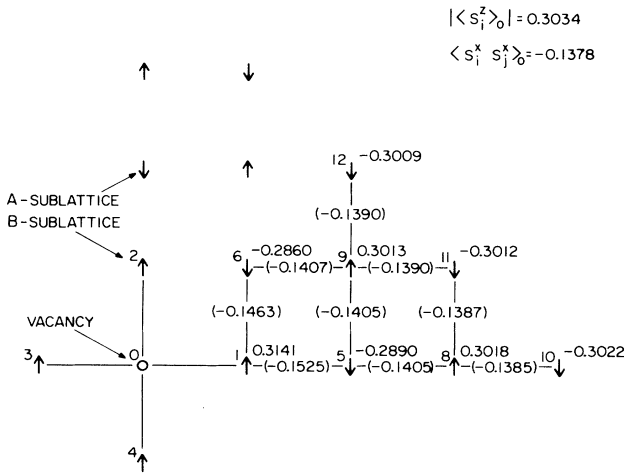

FIG. 2. $\langle S^z_i\rangle$ and $\langle S^x_i S^x_j\rangle$ (in parentheses) around a single vacancy at site 0 on an infinite lattice. Important sites are labeled by integers from 0 through 12.

where δ labels the sites neighboring the vacancy. This potential is local, so it can be treated *exactly*[7] (within the LSW approximation) to yield the following coupled equations for the dressed Green's functions:

$$G_{jj'}(\omega)=G^0_{jj'}(\omega)-\tfrac{1}{4}J\sum_{\delta=1}^{4}[G^0_{j\delta}(\omega)+F^0_{j0}(\omega)]G_{\delta j'}(\omega). \tag{9}$$

This Dyson-type equation can be inverted easily, if we go to a representation based on the point symmetry of the impure lattice. For example, to calculate the corresponding Green's functions for the site next to the vacancy, we introduce

$$S(\omega)\equiv\tfrac{1}{4}\sum_{\delta=1}^{4}G_{1\delta}(\omega), \tag{10a}$$

$$P(\omega)\equiv\tfrac{1}{4}[G_{11}(\omega)-G_{13}(\omega)], \tag{10b}$$

$$D(\omega)\equiv\tfrac{1}{4}\sum_{\delta=1}^{4}(-1)^{\delta+1}G_{1\delta}(\omega). \tag{10c}$$

The imaginary part of the dressed Green's function $G_{11}(\omega)$, and of its components with S-, P-, and D-type symmetries, are plotted in Fig. 1. Using Eq. (8) we find that $\langle S^z_1\rangle=0.3141$ is about 3.5% higher than the pure host value of $\langle S^z_1\rangle_0=0.3034$. This unexpected suppression of the zero-point deviation for the spins next to the vacancy indicates that the *absence* of the transverse terms more than compensates for the missing longitudinal term. This is consistent with the exact results which Wang and Callen[8] obtained for an antiferromagnetically coupled impurity in a spin-$\frac{1}{2}$ Heisenberg ferromagnet. They found that the quantum fluctuations are suppressed as the strength of the AF coupling of the impurity spin to the host spins is reduced. In Fig. 2 site magnetizations $\langle S^z_i\rangle$ and nearest-neighbor transverse spin correla-

TABLE I. Column 2: $\langle S_l^z \rangle$ as a function of the Manhattan distance $l = |x| + |y|$ from the static vacancy on a 4×4 lattice (where $l \leq 4$). Column 3: $\Delta = E(2) - 2E(1)$, the binding energy (in units of J) of two static vacancies on a 4×4 lattice.

l	$\langle S_l^z \rangle$	Δ
1	0.2071	−0.2822
2	−0.1519	0.0037
3	0.1831	−0.1358
4	−0.1491	−0.0112

tions $\langle S_i^x S_j^x \rangle$ are shown for a cluster of 40 sites around the vacancy. We note that the suppression of the quantum fluctuations is observed only for the spins next to the vacancy, whereas on the *average* the quantum fluctuations are enhanced.[9]

The energy cost of removing one spin from an infinite lattice is $E(1) \simeq 0.5776J$. This is calculated by neglecting the vacancy effects outside the 40-site cluster. As the position of the bond moves away from the vacancy, ϵ_{ij}, the bond energy evaluated in the LSW approximation, rapidly approaches its pure host value, which justifies the neglect of energy changes beyond the finite cluster. $E(1)$ is less than the cost of losing four bonds, $4|\epsilon_{ij}^0| = 0.6579J$. The difference of $0.0803J$ is due to the fact that the remaining spins have lowered their energy considerably by readjustment of their correlations with their neighbors. This change in the energy of the background spins is less than ϵ_{ij}^0, the energy of a single bond in the pure lattice; within the LSW approximation the dominant term in the energetics of many vacancies will be the number of bonds missing. We also find that $\langle S_{\rm tot}^z \rangle = \sum_l (\langle S_l^z \rangle - \langle S_l^z \rangle_0) \simeq 0.4825$ for l inside the cluster. Thus, although $\langle S_l^z \rangle$ does not converge as rapidly as the bond energy ϵ_{ij}, we nevertheless have most of the total lattice spin deviation of 0.5 contained within the cluster.[10]

Table I shows exact diagonalization results on a 4×4 lattice with periodic boundary conditions. In column 2, $\langle S_l^z \rangle$ is given for a single vacancy. We note that $|\langle S_l^z \rangle|$ is greatest near the vacancy, which is in agreement with the LSW calculation. The energy cost of a single vacancy is $0.6743J$, which is much higher than what we found in the LSW calculation for an infinite lattice. To investigate this point further, we diagonalized the full LSW Hamiltonian on finite lattices with a Bogoliubov transformation.[6] The results we obtained are given in Table II. The cost of removing a single spin is in column 3. Note that on a 4×4 lattice the LSW and the exact results agree remarkably well, with the background spins lowering their energy by about 4% of $4|\epsilon_{ij}^0|$, the cost of four missing bonds. However, for larger lattices the background spins lower their correlation energy by up to 12% of $4|\epsilon_{ij}^0|$, resulting in a significantly lower ground-state energy. We believe that the difference is due to finite-size effects.

TABLE II. Energetics of the 2D Heisenberg AF lattice with static impurities. Here ϵ_{ij}^0 is the bare bond energy of a pure lattice, $E(1)$ and $E(2)$ are the energy costs of a single and two nearest-neighbor vacancies, respectively, and $\Delta = E(2) - 2E(1)$ is the binding energy.

	ϵ_{ij}^0	$E(1)$	$E(2)$	Δ
Exact (4×4)	−0.1754	0.6743	1.0664	−0.2822
LSW (4×4)	−0.1730	0.6635	1.0686	−0.2584
LSW (6×6)	−0.1669	0.6166	1.0416	−0.1915
LSW (8×8)	−0.1655	0.6000	1.0336	−0.1664
LSW (infinite)	−0.1645	0.5776	1.0218	−0.1333

Next, we investigate the nature of the interaction between two static vacancies through the AF background. The potential that represents two nearest-neighbor vacancies can be written as

$$V' = -\tfrac{1}{2} J \sum_{\delta=1}^{4} \mathbf{S}_\delta^B \cdot \mathbf{S}_0^A - \tfrac{1}{2} J \sum_{\alpha=5}^{7} \mathbf{S}_\alpha^A \cdot \mathbf{S}_1^B , \qquad (11)$$

where α labels the sites 5, 6, and 7. We repeated the calculations above, using V' rather than V. The resulting site magnetization $\langle S_l^z \rangle$ and the nearest-neighbor transverse correlations $\langle S_i^x S_j^x \rangle$ for an infinite lattice will be given elsewhere.[6]

We find that the energy cost of two adjacent vacancies, $E(2)$, on an infinite lattice is $1.0218J$, which is less than twice the cost of a single vacancy. The binding energy is $\Delta = E(2) - 2E(1) \simeq -0.1333J$. This is about a factor of 2 smaller than what we found in the LSW and exact diagonalization calculations on a 4×4 lattice, as shown in Table II. Note that the binding energy Δ is an extremely sensitive function of the lattice size. Another interesting result, as well as an internal check on the numerical calculations, is that $E(1)$ and $E(2)$ vary linearly with L^{-2}, while $|\epsilon_{ij}^0|$ varies linearly with L^{-3}, as the length L of an $L \times L$ lattice is changed.[11]

In conclusion, we have studied the problem of a static vacancy on a 2D AF Heisenberg lattice both analytically, within the LSW approximation, and numerically. We found that at sites next to a single vacancy the zero-point fluctuations are suppressed, leading to an enhanced staggered magnetization. This enhancement might be observable in a Knight-shift experiment in $La_2Cu_{1-x}Zn_xO_4$ or $La_{2-y}Sr_yCuO_4$ for small values of y, where the holes are localized. We also studied the interaction of two static vacancies and found an attractive effective interaction when they are nearest neighbors. Finally, we diagonalized the LSW Hamiltonian and found a remarkable agreement between the LSW and the exact results on a 4×4 lattice. However, LSW on larger lattices yielded significantly different binding and ground-state energies, suggesting that finite-size effects in small-cluster calculations might be very important.

We have recently received a preprint by Nagaosa, Hatsuga, and Imada,[12] who studied the related problem

of an *extra* spin coupled to an infinite 2D AF lattice rather than a vacancy.

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