Isolated ferromagnetic bonds in the two-dimensional spin-$\frac{1}{2}$ Heisenberg antiferromagnet

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An isolated ferromagnetic bond replacing an antiferromagnetic link in the spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a square lattice is investigated within the linearized spin-wave theory. As a function of the ferromagnetic coupling K, quantum fluctuations on neighboring sites are first reduced, which increases the sublattice magnetization. Larger couplings suppress the sublattice magnetization and increase the transversal correlations between nearest neighbors close to the impurity link. Our simple approach breaks down for $|K| > |J|$ (J being the antiferromagnetic coupling). The interaction between two such ferromagnetic links depends on their relative position and its sign oscillates as a function of distance.

I. INTRODUCTION

The discovery of numerous high-temperature superconductors has renewed the interest in the two-dimensional Heisenberg antiferromagnet. Several mechanisms for high-T_c superconductivity invoke properties of the two-dimensional Hubbard model,[1-3] in particular the strong antiferromagnetic correlations within the CuO planes. Properties of the high-T_c compounds are believed to be related to defects in the planes, e.g., static vacancies, ferromagnetic bonds, and mobile holes. It is then important to investigate the effects of isolated defects on the magnetic properties of the two-dimensional antiferromagnet.

Previous studies were devoted to static vacancies[4,5] and dynamic holes.[3] In this paper we consider the effects of isolated ferromagnetic links on an otherwise antiferromagnetic square lattice. The addition of holes in La_2CuO_4, e.g., by doping with Sr, introduces a local ferromagnetic exchange coupling between Cu spins.[6] The resulting frustration affects the antiferromagnetic correlations in the neighborhood of the ferromagnetic link. In Ref. 6 it is argued that these ferromagnetic defects are the origin of the spin-glass[7] and superconducting phases of $La_{2-x}Sr_xCuO_4$.

In two dimensions the quantum fluctuations prevent the long-range magnetic order of the spin-$\frac{1}{2}$ Heisenberg antiferromagnet at nonzero temperature. The nature of the ground state, i.e., if nonmagnetic of the resonant-valence-bond-type[8] or magnetic with broken symmetry as in the Néel state,[9-11] for a square lattice, however, is still controversial. Based on the numerical results presented in Refs. 9–11 we adopt the Néel ground state as our working basis. Quantum fluctuations reduce the sublattice magnetization to an ordered moment of about 0.3. Surprisingly, the linear spin-wave approximation[12,13] is remarkably successful in reproducing quantitatively exact results for finite-size systems and those of more elaborated approaches (see, e.g., Ref. 14).

We assume that the simple linearized spin-wave theory provides a reasonable description of the two-dimensional square lattice spin-$\frac{1}{2}$ Heisenberg antiferromagnet. The problem of a single ferromagnetic bond embedded in an otherwise antiferromagnetic square lattice (nearest-neighbor coupling only) can be solved exactly within the spin-wave approximation. The scattering potential arising from the ferromagnetic bond is local and hence factorizable. The procedure is similar to the one employed in Refs. 4 and 5.

Our main results are the following. There are two competing interactions affecting the sublattice magnetization: The longitudinal terms, involving S_z, tend to enhance the ordered staggered magnetic moment, while the transverse terms, involving S_x and S_y, represent the quantum fluctuations that suppress the sublattice magnetization. We have analyzed the interplay between these two effects as a function of the ferromagnetic coupling strength K. For small K the quantum fluctuations are reduced in the neighborhood of the impurity link and the local magnetic moment is enhanced in agreement with results by Bulut *et al.*[4] for a static vacancy. This process is reversed with increasing K, i.e., the staggered magnetization is gradually suppressed close to the impurity link. The perturbation caused by the ferromagnetic bond falls off as R^{-3} with the distance from the defect. We also calculated the correlation $\langle S_i^x S_j^x \rangle$ for neighboring sites and the interaction energy of two distant ferromagnetic bonds.

We noticed the formation of a localized mode that is due to the impurity link. Within our formulation this mode gets soft as $K \to |J|$ and our approach breaks down. Because of the relatively strong local perturbation caused by K, interactions between spin waves should be taken into account (beyond the linear spin-wave approximation), which then would prevent the softening of the local mode and stabilize the staggered magnetization.

The rest of the paper is organized as follows. In Sec. II we introduce the model and present its exact solution within the linear spin-wave approximation. Our results are discussed in Sec. III, followed by concluding remarks in Sec. IV.

II. MODEL AND CALCULATION

We consider the two-dimensional spin-$\frac{1}{2}$ Heisenberg antiferromagnet on a square lattice with nearest-neighbor coupling only. One antiferromagnetic link in the lattice is replaced by a ferromagnetic one of strength K. The Hamiltonian then has the following form:

$$H=\frac{J}{2}\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j-\frac{J+K}{2}\mathbf{S}_0\cdot\mathbf{S}_1 , \tag{1}$$

with $J>0$ and the impurity bond links the neighboring sites labeled 0 and 1. For $K=0$ Eq. (1) corresponds to a missing bond. Here we adopt the usual convention of $\langle ij\rangle$ meaning a single term for each pair.

Within the spin-wave theory the square lattice is divided into two interpenetrating sublattices denoted by $\underline{a}$ and $\underline{b}$, respectively, and the spin operators are replaced by two sets of boson operators by means of the Holstein-Primakoff transformation.[12,13] We denote these operators $a_i,a_i^\dagger$ and $b_j,b_j^\dagger$, respectively. In this way the Hamiltonian contains products of operators to all orders. Keeping only up to the bilinear terms we obtain the so-called linear spin-wave approximation (LSWA)

$$H_0=J\sum_{\mathbf{k}}[\gamma_{\mathbf{k}}(a_{\mathbf{k}}b_{\mathbf{k}}+b_{\mathbf{k}}^\dagger a_{\mathbf{k}}^\dagger)+(a_{\mathbf{k}}^\dagger a_{\mathbf{k}}+b_{\mathbf{k}}^\dagger b_{\mathbf{k}})]-\frac{J}{2}N , \tag{2}$$

where N is the number of sites of one sublattice, the $\mathbf{k}$ summation is over the N points of the reduced Brillouin zone and

$$\gamma_{\mathbf{k}}=\tfrac{1}{2}[\cos(k_x d)+\cos(k_y d)] , \tag{3}$$

with d being the lattice parameter. The perturbation due to the ferromagnetic link oriented in the x direction is given by

$$H_1=-\frac{J+K}{4N}\sum_{\mathbf{k},\mathbf{k}'}(a_{\mathbf{k}}^\dagger a_{\mathbf{k}'}+e^{-i(k_x-k_x')d}b_{\mathbf{k}}^\dagger b_{\mathbf{k}'}+e^{ik_x'd}a_{\mathbf{k}}b_{\mathbf{k}'}+e^{-ik_x d}a_{\mathbf{k}}^\dagger b_{\mathbf{k}'})+\tfrac{1}{8}(J+K) . \tag{4}$$

The spin operators in terms of the magnon creation and annihilation operators are given by (LSWA)

$$\begin{aligned}
&S_{aj}^+=\frac{1}{\sqrt{N}}\sum_{\mathbf{k}}e^{-i\mathbf{k}\cdot\mathbf{x}_j}a_{\mathbf{k}},\quad S_{bl}^+=\frac{1}{\sqrt{N}}\sum_{\mathbf{k}}e^{-i\mathbf{k}\cdot\mathbf{x}_l}b_{\mathbf{k}}^\dagger ,\\
&S_{aj}^-=\frac{1}{\sqrt{N}}\sum_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{x}_j}a_{\mathbf{k}}^\dagger,\quad S_{bl}^-=\frac{1}{\sqrt{N}}\sum_{\mathbf{k}}e^{i\mathbf{k}\cdot\mathbf{x}_l}b_{\mathbf{k}} ,\\
&S_{aj}^z=\tfrac{1}{2}-\frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'}e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{x}_j}a_{\mathbf{k}}^\dagger a_{\mathbf{k}'} ,\\
&S_{bl}^z=-\tfrac{1}{2}+\frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'}e^{-i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{x}_l}b_{\mathbf{k}}^\dagger b_{\mathbf{k}'} .
\end{aligned} \tag{5}$$

We introduce the standard advanced and retarded one-particle Green's functions in matrix form

$$\hat{G}_{\mathbf{k},\mathbf{k}'}(z)\equiv\begin{bmatrix}\langle\langle a_{\mathbf{k}}^\dagger;a_{\mathbf{k}'}\rangle\rangle_z & \langle\langle a_{\mathbf{k}}^\dagger;b_{\mathbf{k}'}^\dagger\rangle\rangle_z\\ \langle\langle b_{\mathbf{k}};a_{\mathbf{k}'}\rangle\rangle_z & \langle\langle b_{\mathbf{k}};b_{\mathbf{k}'}^\dagger\rangle\rangle_z\end{bmatrix} . \tag{6}$$

The standard equation of motion for $\hat{G}_{\mathbf{k},\mathbf{k}'}(z)$ yields

$$\hat{G}_{\mathbf{k},\mathbf{k}'}(z)=\hat{G}_{\mathbf{k}}^{0}(z)\delta_{\mathbf{k},\mathbf{k}'}-\frac{J+K}{4N}\hat{G}_{\mathbf{k}}^{0}(z)\sum_{\mathbf{k}''}\hat{T}_{\mathbf{k},\mathbf{k}''}\hat{G}_{\mathbf{k}'',\mathbf{k}'}(z) , \tag{7}$$

where $\hat{G}_{\mathbf{k}}^{0}(z)$ is the Green's function for the lattice in the absence of impurity bonds

$$\hat{G}_{\mathbf{k}}^{0}(z)=[J\hat{I}-J\gamma_{\mathbf{k}}\hat{\sigma}_x-z\hat{\sigma}_z]/[z^2-J^2(1-\gamma_{\mathbf{k}}^2)] \tag{8}$$

and $\hat{T}_{\mathbf{k},\mathbf{k}'}$ is the reduced scattering matrix given by

$$\hat{T}_{\mathbf{k},\mathbf{k}'}=\begin{bmatrix}1 & 0\\ e^{-ikxd} & 0\end{bmatrix}\begin{bmatrix}1 & e^{ik_x'd}\\ 0 & 0\end{bmatrix} . \tag{9}$$

Here $\hat{I},\hat{\sigma}_x$ and $\hat{\sigma}_z$ denote the identity and the respective Pauli matrices. Since $\hat{T}_{\mathbf{k},\mathbf{k}'}$ factorizes [see Eq. (9)], Eq. (7) is not a real integral equation and can be solved analytically,

$$\hat{G}_{\mathbf{k},\mathbf{k}'}(z)=\hat{G}_{\mathbf{k}}^{0}(z)\delta_{\mathbf{k},\mathbf{k}'}-\frac{J+K}{4N}\frac{1}{\chi(z)}\hat{G}_{\mathbf{k}}^{0}(z)\hat{T}_{\mathbf{k},\mathbf{k}'}\hat{G}_{\mathbf{k}'}^{0}(z) , \tag{10}$$

where

$$\chi(z)=1-\frac{J+K}{2J}+\frac{J+K}{2JN}\sum_{\mathbf{k}}\frac{z^2}{z^2-J^2(1-\gamma_{\mathbf{k}}^2)} . \tag{11}$$

Due to the $\mathbf{k}$ *and* $\mathbf{k}'$ dependence in Eq. (10) the local magnetization is a function of the relative position of the site with respect to the ferromagnetic bond. On the a sublattice, the magnetization is obtained via [from Eq. (5)]

$$\begin{aligned}\langle S_{aj}^z\rangle&=\tfrac{1}{2}-\langle a_j^\dagger a_j\rangle\\&=\tfrac{1}{2}+\frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'}e^{i(\mathbf{k}-\mathbf{k}')\cdot\mathbf{x}_j}\int_0^\infty\frac{d\omega}{\pi}\mathrm{Im}\langle\langle a_{\mathbf{k}}^\dagger;a_{\mathbf{k}'}\rangle\rangle_{\omega+i\epsilon} ,\end{aligned} \tag{12}$$

where Im denotes the imaginary part and the Green's function is given by Eq. (10). In the absence of impurity bonds we obtain

$$\langle S_{aj}^z\rangle=\tfrac{1}{2}-\frac{2}{\pi^2}\int_0^1 dx\left[\frac{1-x}{1+x}\right]^{1/2}K(x)=0.3034 , \tag{13}$$

in agreement with previous LSWA results.[12,13] Here $K(x)$ is the elliptical integral of the first kind. The site dependence is introduced by the last term in Eq. (10). The magnetization involves then two $\mathbf{k}$ sums and a frequency integration, i.e., a fivefold integration. By means of a contour integral the real-frequency integration is transformed into one along the imaginary axis by rescaling $\omega=iyJ$. The following identity is useful to decouple the k_x and k_y integrations:

$$(y^2+1-\gamma_{\mathbf{k}}^2)^{-1}=\tfrac{1}{2}(1+y^2)^{-1/2}\int_0^\infty dt e^{-t(1+y^2)^{1/2}}\times(e^{t\gamma_{\mathbf{k}}}+e^{-t\gamma_{\mathbf{k}}}) , \tag{14}$$

so that they can be carried out giving rise to modified Bessel functions of integer order I_n. The position of the lattice site $\mathbf{X}_j$, relative to the left corner of the impurity bond, is denoted by $\mathbf{X}_j = d(n_x\hat{\mathbf{x}} + n_y\hat{\mathbf{y}})$, where $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ are unit vectors. In this way the ferromagnetic link is given by the vector $d\hat{\mathbf{x}}$. Asymptotically for $|\mathbf{X}_j| \to \infty$ the magnetization on the sublattice a reaches the value 0.3034 [see Eq. (13)]. The deviations from that value at finite $\mathbf{X}_j$ can be expressed as

$$\delta\langle S^z_{aj}\rangle = -\delta\langle a_j^\dagger a_j\rangle = -\frac{J+K}{64\pi}\int_0^\infty dy \times \left[(3\tilde{I}^{|n_y|}_{|n_x|} - \tilde{I}^{|n_y|}_{|n_x-2|} - \tilde{I}^{|n_y+1|}_{|n_x-1|} - \tilde{I}^{|n_y-1|}_{|n_x-1|})^2 - (4y\tilde{I}^{|n_y|}_{|n_x|})^2\right] / \left[\tfrac{1}{2}(J-K)(1+y^2) + \tfrac{1}{2}(J+K)y^2(1+y^2)^{1/2}\tilde{I}^0_0(y)\right], \tag{15}$$

where

$$\tilde{I}^m_n(y) = \int_0^\infty dt\; I_n\left(\frac{t}{2}\right) I_n\left(\frac{t}{2}\right)\exp[-t(1+y^2)^{1/2}]\,. \tag{16}$$

In a similar fashion the nearest-neighbor spin correlations $\langle S_i^x S_j^x\rangle$ can be calculated using the Green's function, Eq. (10),

$$\langle S_i^x S_j^x\rangle = \tfrac{1}{2}\langle a_i^\dagger b_j^\dagger\rangle = -\frac{1}{N}\sum_{\mathbf{k},\mathbf{k}'}\exp[i(\mathbf{k}\cdot\mathbf{x}_i - \mathbf{k}'\cdot\mathbf{x}_j)] \times \int_0^\infty \frac{d\omega}{2\pi}\,\mathrm{Im}\langle\langle a_{\mathbf{k}}^\dagger; b_{\mathbf{k}'}^\dagger\rangle\rangle_{\omega+i\epsilon}\,. \tag{17}$$

The first term in Eq. (10) yields the correlation in the absence of impurity link, i.e.,

$$\langle S_i^x S_j^x\rangle^0 = -\frac{1}{\pi^2}\int_0^1 dx(1-x^2)^{1/2}K(x) = -0.1378\,. \tag{18}$$

The second term in Eq. (10) again yields the position-dependent deviations from the asymptotic value, which can be brought into a form similar to expression (15). The t and y integrations are then performed numerically. The results are presented in Sec. III.

III. RESULTS

Our results for the magnetization and the spin-spin correlation function are summarized in Figs. 1(a)–1(d). From the symmetry of the problem only the sector $n_x > 0$ and $n_y \geq 0$ needs to be considered. If $K + J = 0$ the impurity link has the same antiferromagnetic coupling strength as all other bonds, so that it is actually not an impurity. Under these circumstances all spin-spin correlations between neighbors are equal to -0.1378 and the staggered sublattice magnetization has an ordered moment of 0.3034.

When K is increased, two competing interactions affect the neighborhood of the impurity link. The modified longitudinal term tends to reduce the magnetic moment, while the change in the transversal terms suppresses quantum fluctuations; this latter effect should lead to an increase of the magnetization. The case $K = 0$ just corresponds to a missing link. Here the sites with the unsatisfied bond have a larger magnetic moment than the asymptotic value, i.e., the reduction of the quantum fluctuations is more important than the missing Ising interaction. This is similar to the findings by Bulut *et al.*,[4] who solved the vacancy problem within the LSWA, i.e., four missing bonds with one common vertex. This trend is reversed for most of the other sites, where the local moment is reduced (with respect to the one of the pure antiferromagnet). This general increase in the transversal fluctuations is consistent with the enhanced $\langle S_1^x S_j^x\rangle$ values we obtained from our calculation. As expected the correlation across the missing bond is strongly reduced.

The trend towards a planar antiferromagnet in the neighborhood of the impurity bond increases with growing ferromagnetic coupling K. This development can be seen by comparing the sequence of Figs. 1(a) through 1(d), where K monotonically increases from 0 to $3J/4$. Almost all the magnetic moments are now smaller than the one of the pure antiferromagnet and the transverse correlation is systematically enhanced with K (except for those bonds corresponding to a translation of the impurity link into the y direction). Not unexpectedly the strongest reduction of the magnetic moment occurs at the sites linked by the ferromagnetic coupling. Note that for $K = 3J/4$ the transversal correlation across the ferromagnetic bond has already inverted its sign, i.e., the spins are weakly ferromagnetically correlated.

The magnetic moment of the sites joined by the impurity bond is shown in Fig. 2 as a function of K/J. For $K = 0$ we notice the slightly increased value already discussed above, but for $K > 0$ (ferromagnetic coupling) $\langle S_z\rangle$ drops monotonically. It is evident from this figure that the magnetic moment within our approach breaks

down around $K \sim 0.85J$. The mathematical reason for this breakdown can be understood from the integral in Eq. (15). As $K \to J$ the weight in the denominator is shifted towards the second term, which for small y is proportional to y^2. The numerator, on the other hand, is finite as $y \to 0$, so that if $K \to J$ the integral diverges. This argument is independent of n_x and n_y, so that for $K = J$ the Néel state for the entire lattice breaks down. Similarly the corresponding integrals giving the transversal correlation $\langle S_i^x S_j^x \rangle$ also diverge. This result is of course unphysical and an artifact of our treatment. The denominator in Eq. (15), which arises from the scattering of spin waves off the impurity link, corresponds to a localized mode. This mode becomes soft as $K \to |J|$. Due to the strong local perturbation a large number of spin-waves is generated and interaction effects among the spin waves have to be taken into account, which probably prevent the mode from becoming soft. In other words, if K is large the two spins linked by the impurity bond are likely to form a triplet state, which is not adequately treated within our approach and leads to a frustration of the spins.

Inserting Eq. (10) into Eq. (12) it can be shown that asymptotically for large distances ($K < |J|$) the perturbation due to the impurity bond falls off with distance as R^{-3}, i.e., $\delta\langle S_j^z \rangle \sim R^{-3}$. With increasing K the amplitude of the perturbation grows and the asymptotic region is shifted to larger R. The amplitude diverges as $K \to |J|$.

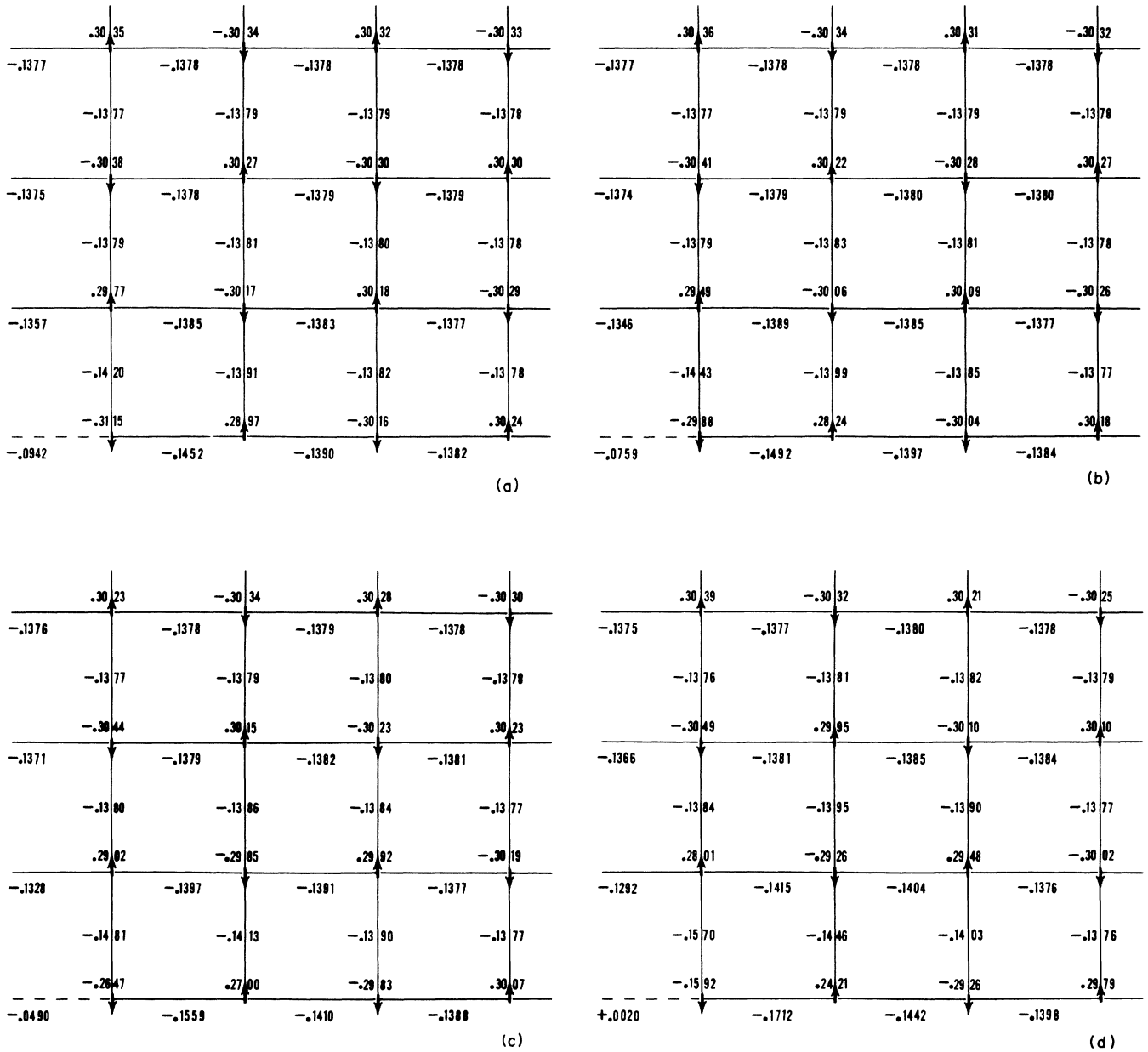

FIG. 1. Magnetic moment $\langle S_j^z \rangle$ and transversal correlation $\langle S_i^x S_j^x \rangle$ around an isolated impurity bond of coupling strength K in the lower left corner as given by the LSWA. For the pure lattice $|\langle S_j^z \rangle^0| = 0.3034$ and $\langle S_i^x S_j^x \rangle^0 = -0.1378$. The values of the ferromagnetic coupling are, respectively (a) $K = 0$, (b) $K = J/4$; (c) $k = J/2$, and (d) $K = 3J/4$.

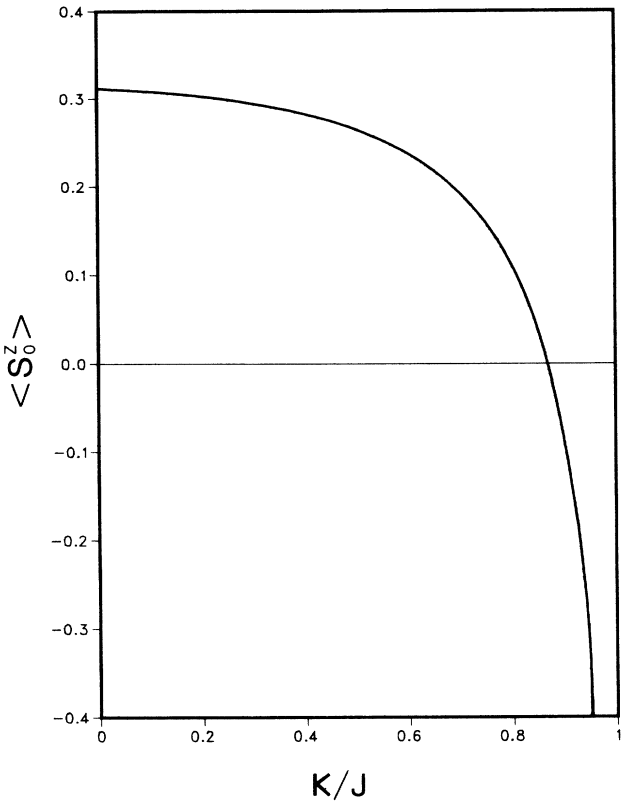

FIG. 2. Magnetization at the sites joined by the ferromagnetic bond as a function of K. As $K \to |J|$ our approximation scheme breaks down. The local magnetization at the impurity bond disappears at about $K \sim 0.9J$.

IV. CONCLUDING REMARKS

The solution of a ferromagnetic link embedded in a two-dimensional antiferromagnet on a square lattice within the LSWA has been presented. Two competing interactions affect the ordered magnetic moment locally, namely the z component of the interaction (Ising terms) and the transversal terms (fluctuations). With increasing coupling K, the transversal interaction becomes more important and our approximation scheme breaks down as K approaches $|J|$. For $K > |J|$, the two spins linked by K are expected to form a triplet state. A similarly profound change in the nature of the ground state has been found for the frustrated x-y model in two dimensions.[15] The quantitative changes of the magnetic moment and the transversal correlations in the neighborhood of the impurity link are presented in Figs. (1a)–(1d).

Finally we would like to summarize our results for the interaction energy of two distant ferromagnetic bonds. Without loss of generality we place one ferromagnetic bond parallel to the x axis. We denote with **R** the vector joining the two links and call θ the angle **R** forms with the x axis. The second bond can be parallel to the x axis or to the y axis, i.e., the bonds are parallel or perpendicular to each other. We calculated the asymptotic interaction energy for large R from second-order perturbation in $J+K$ within the Debye approximation for the spin-wave spectrum

$$E_{\rm int} \propto (3\cos^2\theta - 1) R^{-9/2} \cos(q_D R) ,$$

$$E_{\rm int} \propto |\sin 2\theta| R^{-9/2} \cos(q_D R) ,$$

where $q_D \simeq \sqrt{2\pi}/d$ is the Debye cutoff. The amplitude depends on the relative position of the bonds, falls off as $R^{-9/2}$ with distance and the sign of the interaction oscillates with the distance.

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