Elementary excitations and local spectral distributions in nonhomogeneous antiferromagnets

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Elementary excitations in antiferromagnets which lack translational symmetry are studied. The Green's-function formalism is used to describe the local character of the excitations in terms of spectral distribution functions which are related to the local magnetic moment of the excitations. Two types of excitations are found: one associated with the "up" sublattice and with total magnetic moment $-g\mu_B$, and another associated with the "down" sublattice and with total magnetic moment $g\mu_B$. It is argued that in systems where the up and down sublattices are not equivalent a net ground-state magnetization should appear. After the general formalism is discussed two specific examples are given: a semi-infinite chain and an infinite chain with a single impurity. Both of these systems display nonuniform ground-state magnetization and in the case of the semi-infinite chain, for which the up and down sublattices are not equivalent, a net magnetization does appear.

I. INTRODUCTION

Double-time Green's functions[1] provide a powerful tool for the theoretical description of systems which lack translational symmetry.[2,3] Their use has proven particularly suited to the study of the local behavior of physical quantities such as densities of states, charge densities, and magnetization densities. In most cases there exists a straightforward connection between diagonal matrix elements of the appropriate Green's function and local values of the corresponding physical quantities. Difficulties have been encountered, however, in the application of the method to the study of the low-level excitations of antiferromagnetic surfaces.[4] Attempts to obtain a local density of states from the Green's function[5] have been shown to lead to inconsistencies.[4]

In this paper we present a way to circumvent these difficulties. We argue that no local density of states can be defined because the number-of-excitations operator cannot be decomposed into local number operators. On the other hand, we find that the total density of states can be represented as a sum of local terms, each of which is not a local density of states in the usual sense but can be given a straightforward physical interpretation in terms of the magnetic moment carried by the excitations. Moreover, intrinsically local quantities, such as the ground-state magnetization and the magnetic moment of the excitations, can be readily extracted from the formalism.

To illustrate these points we report calculations for two simple models: A semi-infinite antiferromagnetic chain and a single impurity in an infinite antiferromagnetic chain.

This paper is organized as follows: in Sec. II the general formalism is discussed; in Sec. III the calculations for the systems mentioned above are presented; and finally in Sec. IV our results are summarized and some concluding remarks are presented.

II. GENERAL FORMALISM

We consider an antiferromagnet which can be divided into two equivalent sublattices, α and β, of opposite magnetization. To a point $\mathbf{R}_i$ in the α sublattice there corresponds a point $\mathbf{R}_i+\boldsymbol{\Delta}$ in the β sublattice. The constant vector $\boldsymbol{\Delta}$ is so chosen that the point $\mathbf{R}_i+\boldsymbol{\Delta}$ is one of the nearest neighbors of $\mathbf{R}_i$. The spin operator of the ion at $\mathbf{R}_i$ is denoted by $\mathbf{S}_{i,a}$ and that of the ion at $\mathbf{R}_i+\boldsymbol{\Delta}$ by $\mathbf{S}_{i,b}$. The Heisenberg Hamiltonian for this antiferromagnet can then be written as

$$\mathscr{H}=J\sum_{i,\delta}\mathbf{S}_{i,a}\cdot\mathbf{S}_{i+\delta,b}-g\mu_B H_A\sum_i(S^z_{i,a}-S^z_{i,b})\ , \tag{1}$$

where $J>0$, δ runs over nearest neighbors, μ_B is the Bohr magneton, g is the gyromagnetic ratio, and H_A is an anisotropy field introduced to fix the direction of the magnetization in each sublattice.

We now introduce the Holstein-Primakoff transformation[6]

$$S^z_{i,a}=S-a_i^\dagger a_i\ , \tag{2}$$

$$S^+_{i,a}=(2S-a_i^\dagger a_i)^{1/2}a_i\ , \tag{3}$$

and

$$S^z_{i,b}=-S+b_i^\dagger b_i\ , \tag{4}$$

$$S^-_{i,b}=(2S-b_i^\dagger b_i)^{1/2}b_i\ . \tag{5}$$

When Eqs. (2)—(5) are introduced into Eq. (1) one obtains

$$\begin{aligned}\mathscr{H}_0=&NzJS^2-2NSg\mu_B H_A\\&+(JSz+g\mu_B H_A)\sum_i(a_i^\dagger a_i+b_i^\dagger b_i)\\&+JS\sum_{i,\delta}(a_i b_{i+\delta}+b_{i+\delta}^\dagger a_i^\dagger)\ ,\end{aligned} \tag{6}$$

where N is the total number of atoms in each sublattice, z

is the number of nearest neighbors, and only bilinear terms have been kept.

The above Hamiltonian may be diagonalized by a Bogoliubov transformation of the form

$$\alpha_n = \sum_i u_{ni}^{\alpha} a_i - v_{ni}^{\alpha} b_i^{\dagger} , \tag{7}$$

$$\beta_n = \sum_i -v_{ni}^{\beta} a_i^{\dagger} + u_{ni}^{\beta} b_i , \tag{8}$$

where it is required that

$$[\alpha_n, \alpha_{n'}^{\dagger}] = \delta_{nn'} , \tag{9}$$

$$[\beta_n, \beta_{n'}^{\dagger}] = \delta_{nn'} , \tag{10}$$

$$[\alpha_n, \mathscr{H}] = \epsilon_n^{\alpha} \alpha_n , \tag{11}$$

$$[\beta_n, \mathscr{H}] = \epsilon_n^{\beta} \beta_n , \tag{12}$$

and all other commutators equal zero.

The operator $\alpha_n^{\dagger}$ creates an excitation of energy ϵ_n^{α} and the operator $\beta_n^{\dagger}$ creates an excitation of energy ϵ_n^{β}. The commutation relations can be used to find the inverse transformation

$$a_i = \sum_m (u_{mi}^{\alpha})^* \alpha_m + v_{mi}^{\beta} \beta_m^{\dagger} , \tag{13}$$

$$b_i = \sum_m v_{mi}^{\alpha} \alpha_m^{\dagger} + (u_{mi}^{\beta})^* \beta_m , \tag{14}$$

as well as the orthogonality conditions

$$\sum_i u_{ni}^{\alpha} (u_{n'i}^{\alpha})^* - v_{ni}^{\alpha} (v_{n'i}^{\alpha})^* = \delta_{nn'} , \tag{15a}$$

$$\sum_i u_{ni}^{\beta} (u_{n'i}^{\beta})^* - v_{ni}^{\beta} (v_{n'i}^{\beta})^* = \delta_{nn'} , \tag{15b}$$

$$\sum_m u_{mi}^{\alpha} (u_{mj}^{\alpha})^* - v_{mi}^{\beta} (v_{mj}^{\beta})^* = \delta_{ij} , \tag{16a}$$

$$\sum_m u_{mi}^{\beta} (u_{mj}^{\beta})^* - v_{mi}^{\alpha} (v_{mj}^{\alpha})^* = \delta_{ij} . \tag{16b}$$

From the above equations one can show that the ground-state magnetizations at i are given in units of $g\mu_B$ by

$$\langle S_{i,a}^z \rangle = S - \sum_n | v_{ni}^{\beta} |^2 \tag{17}$$

and

$$\langle S_{i,b}^z \rangle = -S + \sum_n | v_{ni}^{\alpha} |^2 , \tag{18}$$

where the angular brackets denote expectation values. For nonhomogeneous systems these magnetizations will be, in general, nonuniform.

When an excitation $\alpha_n^{\dagger}$ (α excitation) is created from the ground state, the variations in magnetization with respect to the ground state are given by

$$\delta\langle S_{i,a}^z \rangle = - | u_{ni}^{\alpha} |^2 \tag{19}$$

and

$$\delta\langle S_{i,b}^z \rangle = | v_{ni}^{\alpha} |^2 . \tag{20}$$

The corresponding relations for an excitation $\beta_n^{\dagger}$ (β excitation) are

$$\delta\langle S_{i,a}^z \rangle = - | v_{ni}^{\beta} |^2 \tag{21}$$

and

$$\delta\langle S_{i,b}^z \rangle = | u_{ni}^{\beta} |^2 . \tag{22}$$

Since one expects that in most cases $| u_{ni} |^2 > | v_{ni} |^2$, it follows that α excitations tend to be localized in the α sublattice while β excitations tend to be localized in the β sublattice. Moreover the net magnetization change at the ith cell is

$$\delta S_{i,a}^z + \delta S_{i,b}^z = -(| u_{ni}^{\alpha} |^2 - | v_{ni}^{\alpha} |^2) \tag{23}$$

for an α excitation and

$$\delta S_{i,a}^z + \delta S_{i,b}^z = (| u_{ni}^{\beta} |^2 - v_{ni}^{\beta} |^2) \tag{24}$$

for a β excitation.

From the orthogonality relation (15) one sees that the total magnetization change due to an α excitation is -1, while that due to a β excitation is 1. In antiferromagnets in which the α and β sublattices are equivalent, α and β excitations are degenerate and no net magnetization appears. When the symmetry is broken by, for example, a surface, one expects a net magnetization. In the next section we shall see an example of where such net magnetization is found.

The global density of states may be written as

$$N(\omega) = \sum_n [\delta(\hbar\omega - \epsilon_n^{\alpha}) + \delta(\hbar\omega - \epsilon_n^{\beta})] . \tag{25a}$$

With the help of Eq. (15) one finds that

$$N(\omega) = \sum_i [J_i^{\alpha}(\omega) + J_i^{\beta}(\omega)] , \tag{25b}$$

where

$$J_i^{\alpha}(\omega) = -\sum_n \delta S_{i,\alpha}^z \delta(\hbar\omega - \epsilon_n^{\alpha}) , \tag{26a}$$

$$J_i^{\beta}(\omega) = \sum_n \delta S_{i,\beta}^z \delta(\hbar\omega - \epsilon_n^{\beta}) , \tag{26b}$$

and $\delta S_{i,\alpha}^z$ and $\delta S_{i,\beta}^z$ are the net magnetization changes in the ith cell due to α and β excitations, respectively. We thus see that $J_i^{\beta}(\omega) d\omega$ gives the net magnetization change at the ith cell due to the β excitations in the range $(\omega, \omega + d\omega)$. The interpretation of J_i^{α} is the same except that a sign change is introduced so that both spectral distributions are normalized to unity.

We see from the above considerations that the spectral distributions J_i^{α} and J_i^{β} provide a meaningful representation of the global density of states as a sum of local terms. Is it possible to find a projected density of states in the usual sense? In order to be able to locally project the density of states one must be able to define a local particle density for the excitations. However, the number operator

$$\hat{N} \equiv \sum_n \alpha_n^{\dagger} \alpha_n + \beta_n^{\dagger} \beta_n \tag{27}$$

cannot be represented as a sum of local number operators and therefore no single-particle density can be defined. On the other hand, from Eqs. (2) and (4) we see that the z component of the total spin, $\mathscr{S}_z$, may be written as

$$\mathcal{S}_z=\sum_i b_i^\dagger b_i - a_i^\dagger a_i \ , \tag{28}$$

i.e., $\mathcal{S}_z$ does have a representation as a sum of local number operators. Since $\hat{N}$, $\mathcal{S}_z$, and $\mathcal{H}$ mutually commute one may classify the energy eigenstates according to the eigenvalues of $\hat{N}$ and $\mathcal{S}_z$. (As shown above, α and β excitations are one-particle excitations with $\mathcal{S}_z=-1$ and $\mathcal{S}_z=1$.) Thus, it is appropriate to use the local contributions to the total magnetization to describe the local character of the excitations.

In systems with translational invariance the explicit form of the coefficients of the Bogoliubov transformation can be found by a Fourier transformation to the k-space representation.[6] In systems without translational invariance it is convenient to introduce the Green's functions

$$G_{ij}^{aa}(\omega)=-\frac{i}{\hbar}\int_{-\infty}^{\infty} dt\, e^{i\omega t}\theta(t)\langle\psi_0 \mid [a_i(t),a_j^\dagger(0)] \mid \psi_0\rangle \tag{29}$$

and

$$G_{ij}^{bb}(\omega)=-\frac{i}{\hbar}\int_{-\infty}^{\infty} dt\, e^{i\omega t}\theta(t)\langle\psi_0 \mid [b_i(t),b_j^\dagger(0)] \mid \psi_0\rangle \ . \tag{30}$$

The diagonal elements of these Green's functions have the spectral representations

$$G_{ii}^{aa}(\omega)=\sum_n \frac{\mid u_{ni}^\alpha \mid^2}{\hbar\omega-\epsilon_n^\alpha+i\hbar\eta}-\frac{\mid v_{ni}^\beta \mid^2}{\hbar\omega+\epsilon_n^\beta+i\hbar\eta} \tag{31}$$

and

$$G_{ii}^{bb}(\omega)=\sum_n \frac{\mid u_{ni}^\beta \mid^2}{\hbar\omega-\epsilon_n^\beta+i\hbar\eta}-\frac{\mid v_{ni}^\alpha \mid^2}{\hbar\omega+\epsilon_n^\alpha+i\hbar\eta} \ . \tag{32}$$

From the above equations we see that G_{ii}^{aa} becomes singular when $\hbar\omega$ belongs to the spectrum of α excitations and also when $-\hbar\omega$ belongs to the spectrum of β excitations. The behavior of G_{ii}^{bb} is similar except that the roles of α and β excitations are interchanged. We may define the spectral distributions

$$\hat{J}_{ii}^{aa}(\omega)=\frac{1}{\pi}\mathrm{Im}G_{ii}^{aa}(\omega) \tag{33a}$$

and

$$\hat{J}_{ii}^{bb}(\omega)=\frac{1}{\pi}\mathrm{Im}G_{ii}^{bb}(\omega) \ . \tag{33b}$$

These can be expressed as

$$\hat{J}_{ii}^{aa}(\omega)=\sum_n \mid u_{ni}^\alpha \mid^2\delta(\hbar\omega-\epsilon_n^\alpha)-\sum_n \mid v_{ni}^\beta \mid^2\delta(\hbar\omega+\epsilon_n^\beta) \tag{34a}$$

and

$$\hat{J}_{ii}^{bb}(\omega)=\sum_n \mid u_{ni}^\beta \mid^2\delta(\hbar\omega-\epsilon_n^\beta)-\sum_n \mid v_{ni}^\alpha \mid^2\delta(\hbar\omega+\epsilon_n^\alpha) \ . \tag{34b}$$

We can now express the spectral distributions J_i^α and J_i^β as

$$J_i^\alpha(\omega)=\hat{J}_{ii}^{aa}(\omega)+\hat{J}_{ii}^{bb}(-\omega) \tag{35}$$

and

$$J_i^\beta(\omega)=\hat{J}_{ii}^{bb}(\omega)+\hat{J}_{ii}^{aa}(-\omega) \ , \tag{36}$$

where ω is now restricted to positive values.

For a uniform antiferromagnet all of the above spectral distributions are independent of both the lattice site i and the sublattice (α or β) and therefore all indices can be omitted. In Fig. 1 we display $\hat{J}(\omega)$ (solid line) and $J(\omega)$ (dashed line) for a uniform linear antiferromagnet. In this case $J(\omega)$ coincides with the global density of states per site.

The ground-state magnetizations at i, given by Eqs. (17) and (18), may now be expressed as

$$\langle S_{i,a}^z\rangle=S-\int_{-\infty}^{0} d\omega\,\hat{J}_{ii}^{aa}(\omega) \tag{37}$$

and

$$\langle S_{i,b}^z\rangle=-S+\int_{-\infty}^{0} d\omega\,\hat{J}_{ii}^{bb}(\omega) \ . \tag{38}$$

III. ONE-DIMENSIONAL ANTIFERROMAGNETS

In this section we present the results of calculations for two specific systems: a semi-infinite antiferromagnetic chain with constant J ("surface" case) and an infinite antiferromagnetic chain in which all J's are equal except for that at a single bond, which takes the value J_0 (impurity case). All spins S are taken to be $\frac{1}{2}$.

One-dimensional antiferromagnets do not sustain spontaneous magnetization in the absence of a magnetic field.[7] So that antiferromagnetic order appears we assume an anisotropy field, H_A equal to 0.1 (in units of $g\mu_B/zJS$). The Green's functions $G_{ij}^{aa}(\omega)$ and $G_{ij}^{bb}(\omega)$ are found by means of the transfer-matrix technique.[8] The formalism of the previous section is then applied to obtain the results that follow.

In Figs. 2 and 3 we display $J_i^\alpha(\omega)$ and $J_i^\beta(\omega)$ for several values of i in the semi-infinite chain. A surface mode appears below the band edge. This mode is not present in $J_i^\beta(\omega)$ and is, therefore, an α excitation. At $i=4$ both spectral distributions resemble the bulk density of states,

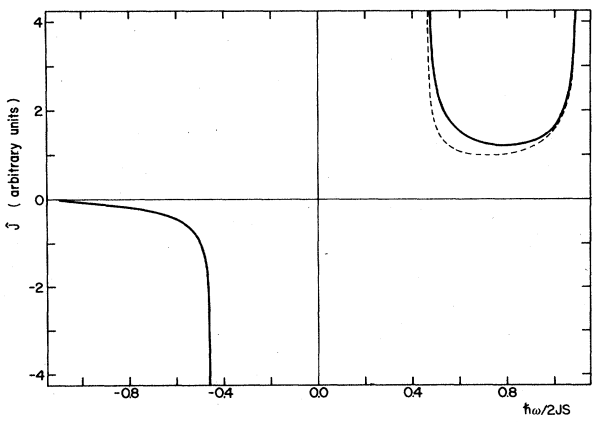

FIG. 1. Local spectral distributions $\hat{J}(\omega)$ (solid line) and $J(\omega)$ (dashed line), for a uniform linear antiferromagnet.

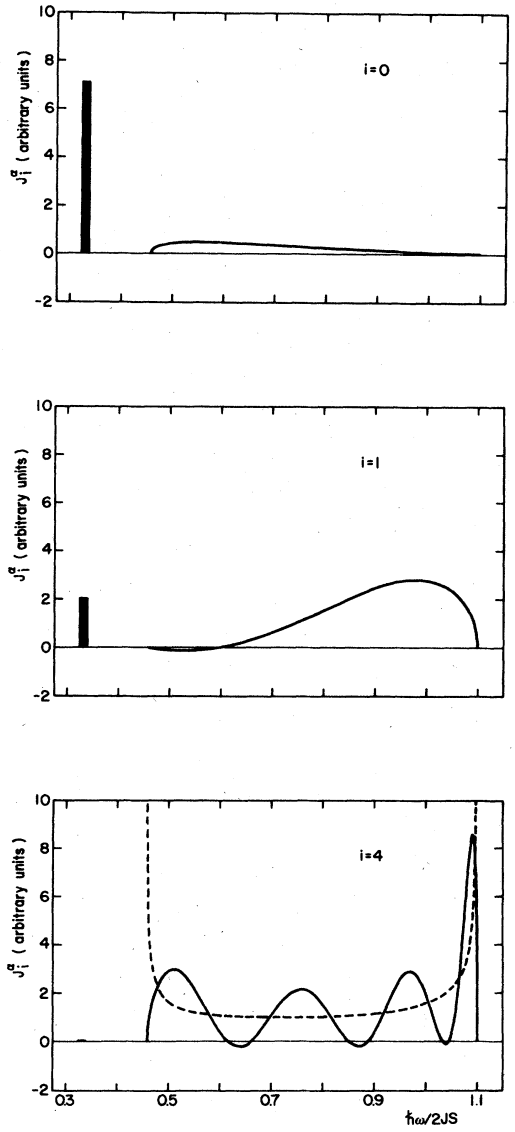

FIG. 2. Local spectral distribution of α excitations, $J_i^\alpha(\omega)$, for $i=0$, 1, and 4 in a semi-infinite antiferromagnetic chain. The solid bars represent a localized state. The height of the bar is equal to ten times the weight of the state at the given site. The vertical scale for the continuum is the same for all values of i.

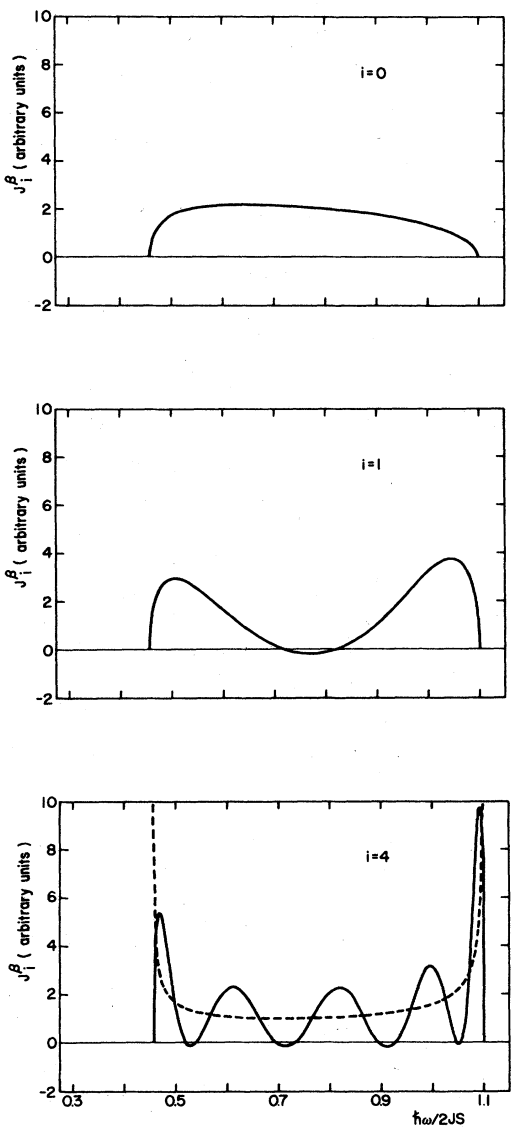

FIG. 3. Local spectral distribution of β excitations, $J_i^\beta(\omega)$, for $i=0$, 1, and 4 in a semi-infinite antiferromagnetic chain. The solid bars represent a localized state. The height of the bar is equal to ten times the weight of the state at the given site. The vertical scale for the continuum is the same for all values of i.

except for oscillations which become more rapid as one goes into the bulk. This is characteristic of one-dimensional systems, where local densities of states converge to the bulk density of states in the distribution sense, but not pointwise. For a given i, the spectral distributions may become negative for some values of ω. This is no cause for concern since it simply means that the local cell-magnetization variations due to the excitations corresponding to such ω's change sign. For example, an α excitation, which produces an overall magnetization change of -1, could actually *increase* the magnetization of some particular cells.

The spectral distributions for the impurity systems behave similarly. For $J_0<J$ there appear two degenerate local modes below the band: an α excitation with most of its weight to the right of the impurity, and a β excitation with most of its weight to the left. When $J_0=0$ these modes correspond to the surface modes of two uncoupled semi-infinite chains. When $J_0>J$ the local modes appear above the band. As the ratio J_0/J is made larger the size of the fluctuations (i.e., the value of $\langle a_i^\dagger a_i \rangle$) at the sites adjacent to the impurity grows without bound. At some value of J_0/J (about 4 for the anisotropy field chosen here) the fluctuations are so large that the one-magnon

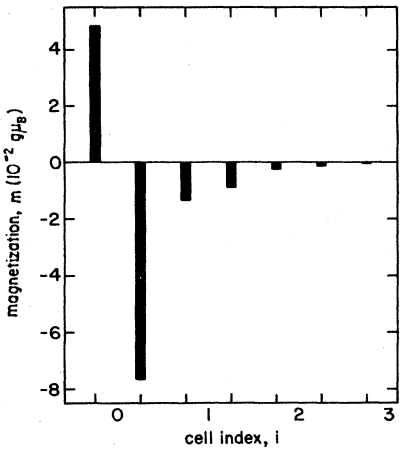

FIG. 4. Local magnetization in the vicinity of the end point of a semi-infinite antiferromagnetic chain. The heights of the bars are given by the difference between the absolute value of the local magnetization and that of the corresponding bulk magnetization. The bars to the left and right of a cell index i correspond to a and b sites, respectively.

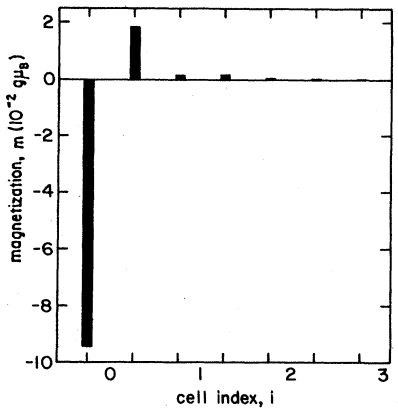

FIG. 6. Local magnetization to the right of an impurity bond in an infinite antiferromagnetic chain. The impurity exchange integral is given by $J_0=2J$. The heights of the bars are given by the difference between the absolute value of the local magnetization and that of the corresponding bulk value. The bars to the left and right of a cell index i correspond to a and b sites, respectively.

approximation breaks down.

The ground-state magnetization of both systems considered can be obtained from Eqs. (37) and (38). For comparison it is useful to define a local magnetization relative to the bulk magnetization as follows:

$$m_i^a = g\mu_B(\,|\langle S_{i,a}^z\rangle| - |\langle S_{\text{bulk}}^z\rangle|\,) \tag{39}$$

and

$$m_i^b = g\mu_B(\,|\langle S_{i,b}^z\rangle| - |\langle S_{\text{bulk}}^z\rangle|\,)\,. \tag{40}$$

Thus, if m_i^a (m_i^b) is positive it indicates that the absolute value of the magnetization at the a site (b site) of lattice point i is larger than the absolute value of the bulk magnetization, and vice versa.

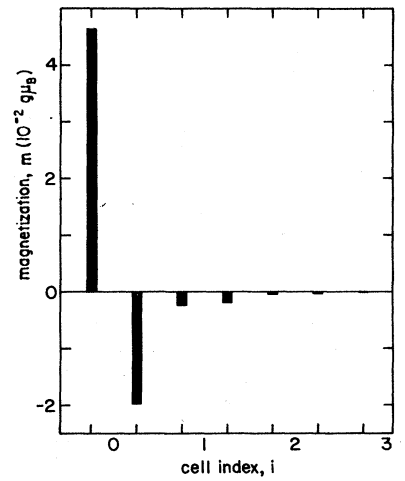

FIG. 5. Local magnetization to the right of an impurity bond in an infinite antiferromagnetic chain. The impurity exchange integral is given by $J_0=0.5J$. The heights of the bars are given by the difference between the absolute value of the local magnetization and that of the corresponding bulk value. The bars to the left and right of a cell index i correspond to a and b sites, respectively.

In Figs. 4, 5, and 6 we display m_i^a and m_i^b for the semi-infinite chain, the infinite chain with $J_0/J=0.5$, and the infinite chain with $J_0/J=2.0$. Only sites to the right of the impurity bond are shown for the infinite chains, as the magnetizations are antisymmetric with respect to reflections about the impurity. From Fig. 3 we see that the absolute value of the magnetization of the surface ion increases, while that of neighboring ions decreases. Furthermore, a net magnetization appears at the surface, as we expect from the lack of symmetry between the α and β sublattices. For the infinite chain with $J_0/J=0.5$ the behavior of the magnetization is similar to that of the surface except that, due to symmetry, no net magnetization appears. When $J_0/J=2$ (and in general when $J_0/J>1$) the situation is reversed: the absolute value of the magnetization of the ion nearest to the impurity decreases, while that of the neighboring ions increases.

IV. SUMMARY AND CONCLUSIONS

We have considered the local description of excitations in a nonhomogeneous antiferromagnet. We have shown that a physically consistent picture can be given in terms of two types of excitations which correspond to changes in the z component of total spin of 1 and -1, respectively. The relevant local "density" associated with a given excitation and a given lattice point, turns out to be the net magnetization of the two ions corresponding to the lattice point. Once this point of view is taken all local quantities are readily extracted from the appropriate Green's functions.

We have also studied two one-dimensional systems: an infinite chain with a single impurity and a semi-infinite chain. The loss of translational symmetry leads to a nonuniform local magnetization in both systems. In the

semi-infinite chain a net total magnetization also appears, a fact that can be thought of as the result of the loss of the equivalence between the spin-up and spin-down sublattices. (When these sublattices are equivalent the two types of excitations are degenerate and their contributions to the magnetization cancel out.) In three-dimensional systems fluctuations tend to be smaller. Nonetheless, since the two effects above are related to the loss of symmetries and not to the specific properties of one-dimensional systems, one expects them to be present in three-dimensional systems as well, although they should be relatively weaker. Thus, in particular, the surface of an antiferromagnet could have a net magnetization, even at $T=0$.

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