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L.M. Falicov

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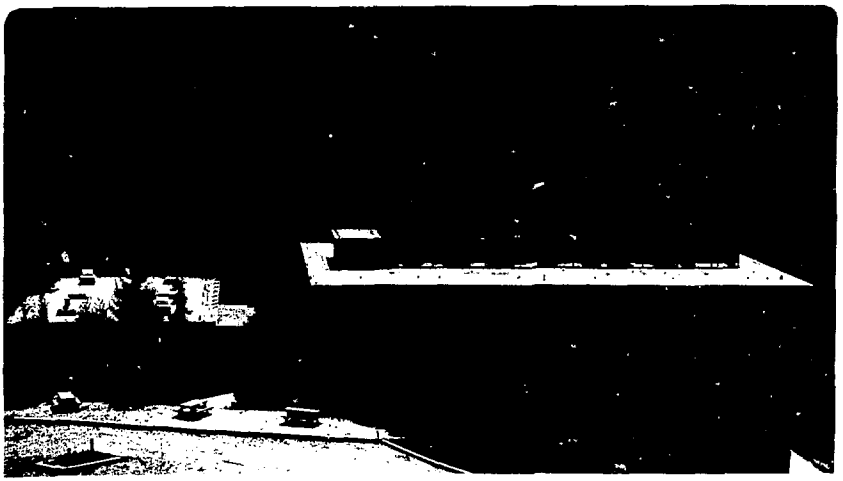

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SPIN WAVES AT SURFACES AND STEPS IN FERROMAGNETS AND ANTIFERROMAGNETS

L.M. Falicov

Materials and Molecular Research Division,
Lawrence Berkeley Laboratory and
Department of Physics, University of California
Berkeley, California 94720

INTRODUCTION

The study of the properties of elementary magnetic excitations--spin waves or magnons--in the neighborhood of surfaces,[1-5] discontinuities, defects,[6] and impurities[7] is an active field of solid-state magnetism. These modes are of interest for understanding relaxation phenomena[8] as well as chemical reactions catalyzed by magnetic substrates.[9]

In this contribution we would like to report two different results: (1) some spectra of the spin-wave modes associated with clean and stepped surfaces of a ferromagnet[10]--the clean and a stepped (001) surface of a face-centered-cubic solid; and (2) the unusual and yet unresolved difficulties encountered when similar procedures are attempted on antiferromagnetic surfaces and steps[11].

MODEL, FORMULATION, AND CALCULATION FOR A FERROMAGNET

The main idea behind our model is to generate in the surface of a solid (a face-centered-cubic solid) steps that repeat periodically but such that, on the average, the surface is parallel to a low-Miller-index plane [the (001) plane in our case]. If these conditions are satisfied, the three-dimensional problem to be solved can be factorized into a two-dimensional periodic part, to be treated in the k representation, and a one-dimensional aperiodic part, which can be treated in the transfer-matrix approach method[12,13] to the Green's-function formalism.[14,15] In addition, since the surface runs, on the average, parallel to a (001) plane, a transfer matrix can be

easily obtained--by means of a simple canonical transformation--from the very simple transfer function for the (001) nickel surface.[12]

Our model for the stepped (001) nickel surface is obtained by removing periodically rows of atoms parallel to the [010] direction from the (001) surface layer. This is shown in Fig. 1. The steps thus formed have a depth of one layer, or half the cubic lattice parameter a, as is the case in real materials.[15,16]

The two-dimensional unit cell describing the stepped surface is a supercell of the primitive (001) cell which contains 12 atoms (see Fig. 1). In the terrace (top) layer, atoms 1 and 7 are symmetrically located at the step edges and have a coordination number of six. Atoms 8-12 are missing. In the second layer, atoms 1 and 7 have a coordination number of 11 (i.e., only one nearest neighbor is missing), atoms 2-6 have a full coordination number of 12, equal to that of a bulk atom, atoms 8 and 12 have a coordination number of nine, and atoms 9, 10, and 11 are once again equivalent to (001) surface atoms with a coordination number of eight. All atoms in the third and subsequent layers are fully coordinated, i.e., have a coordination number of 12.

The Heisenberg Hamiltonian we use in our calculations is of the nearest-neighbor-only type:

$$H = -\sum_{n} \sum_{\Delta} J\vec{S}(n) \cdot \vec{S}(n+\Delta) \ , \tag{1}$$

where J is the nearest-neighbor exchange and $\vec{S}(n)$ is the spin operator at site n. The sum over Δ corresponds to nearest-neighbor positions. The introduction of the raising and lowering operators

$$S^{\pm}(n)=S_x(n)\pm iS_y(n) \tag{2}$$

leads to the more convenient form

$$H = -\sum_{n} \sum_{\Delta} J[S_z(n)S_z(n+\Delta)+S^{+}(n)S^{-}(n+\Delta)] \ . \tag{3}$$

We work only in the one-magnon approximation and for that purpose we introduce a complete set of one-spin-deviation states $|n\rangle$, where $|n\rangle$ denotes a quantum state in which only the spin at the ion site n deviates by one unit from the perfectly aligned ferromagnetic state.

Following the standard Zubarev technique,[13,14,17] the local density of spin-wave states at each atom in the crystal is given by

$$D_{\ell\mu}(\omega)= -\pi^{-1}\,\mathrm{Im} \sum_{\vec{k}} \langle \ell\vec{k}\mu|G(\omega)|\ell\vec{k}\mu\rangle \ , \tag{4}$$

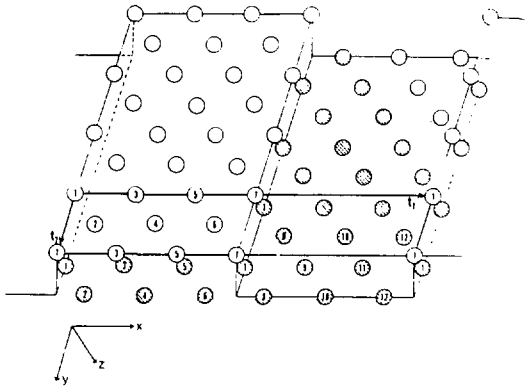

Fig. 1. Model-stepped surface. The surface unit cell contains 12 atoms in each plane, except in the surface layer, where five rows (atoms 8~12) have been removed. The unit cell vectors $\vec{t}_1$ and $\vec{t}_2$ are indicated. Open circles indicate atoms in the terraces (surface layer) and shaded circles atoms in the second layer.

where $\langle \ell\vec{k}\mu|G(\omega)|\ell'\vec{k}\mu'\rangle$ is a matrix element of the Green's function. In (4) ℓ indicates the layer of atoms (ℓ=1 is the surface), $\vec{k}$ is a two-dimensional wave-vector within the corresponding two-dimensional Brillouin zone and μ=1,2,...,12 labels the atoms in the unit cell of Fig. 1. It should be noted that for the stepped surface of Fig. 1 there are no atoms for ℓ=1 and μ=8,9,...,12. If those atoms were present we would obtain an ordinary (001) perfect surface.

If we organize the elements of (4) into a set of (12x12) matrices $\underline{G}_{\ell\ell'}(\omega,\vec{k})$ labeled by ℓ, ℓ', $\vec{k}$, and ω and with rows and columns labeled by μ,μ',=1,...,12, the Green's function satisfies Dyson's equation:

$$\begin{aligned}
\omega\underline{G}_{11} &= \underline{I}+\underline{H}_{11}\underline{G}_{11}+\underline{H}_{12}\underline{G}_{21} \quad , \\
\omega\underline{G}_{21} &= \underline{H}_{21}\underline{G}_{11}+\underline{H}_{22}\underline{G}_{21}+\underline{H}_{23}\underline{G}_{31} \quad , \\
\omega\underline{G}_{n1} &= \underline{H}_{n(n-1)}\underline{G}_{(n-1)1}+\underline{H}_{nn}\underline{G}_{n1}+\underline{H}_{n(n+1)}\underline{G}_{(n+1)1} \quad .
\end{aligned} \tag{5}$$

The solution of the infinite system (5) can be obtained by the transfer matrix technique.[12-14] A matrix $\underline{T}$, independent of n is defined by

$$\underline{G}_{(n+1)n} = \underline{T}\,\underline{G}_{nn}, \quad n\geq 2 \tag{6}$$

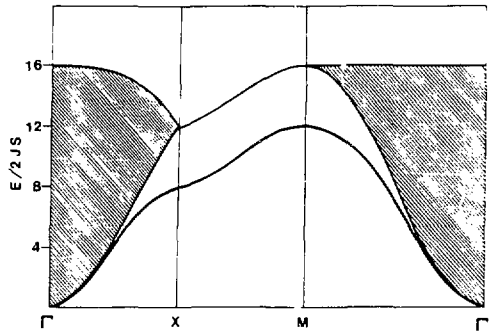

Fig. 2. Bulk and surface states along various lines of the square Brillouin zone of the perfect (001) surface. The shaded area corresponds to the bulk states. Surface states are indicated by the thick line.

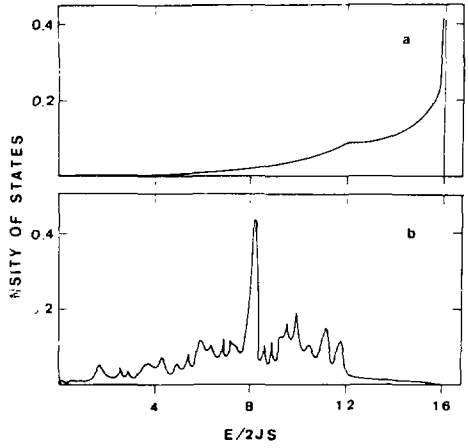

Fig. 3. (a) Local density of states for a bulk atom, integrated over the whole Brillouin zone. (b) Surface local density of states for a perfect (001) surface.

which, substituted in (15), yields

$$\underline{H}_{n(n+1)}\underline{T}\ \underline{T}+(\underline{H}_{nn}-\omega\underline{I})\underline{T}+\underline{H}_{n(n-1)}=0 \quad . \tag{7}$$

If the 12x12 $\underline{T}$ matrix is known, the first two equations in (5) yield

$$\underline{G}_{11}=[\omega\underline{I}-\underline{H}_{11}-\underline{H}_{12}(\omega\underline{I}-\underline{H}_{22}-\underline{H}_{23}\underline{T})^{-1}\underline{H}_{21}]^{-1} \quad , \tag{8}$$

from which the seven nonvanishing local density of states $D_{1\mu}(\omega)$ can be obtained.

With a similar formulation, we obtain

$$\underline{G}_{22}=[\omega\underline{I}-\underline{H}_{22}-\underline{H}_{23}\underline{T}-\underline{H}_{21}(\omega\underline{I}-\underline{H}_{11})^{-1}\underline{H}_{12}]^{-1} \quad , \tag{9}$$

from which the twelve $D_{2\mu}(\omega)$ are easily derived.

Our results for the perfect (001) surface are shown in Figs. 2 and 3.

Results for various local densities of magnon states of the stepped surface (at terrace and step-edge sites) are shown in Figs. 4 and 5.

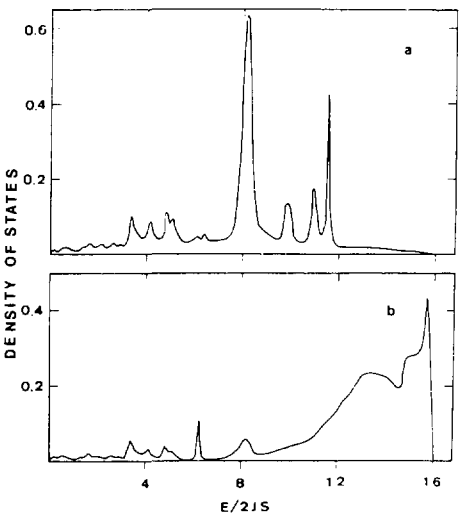

Fig. 4. (a) Local density of states at atom 4 on the surface (terrace) layer of Fig. 1. (b) Local density of states at atom 4 of the second layer.

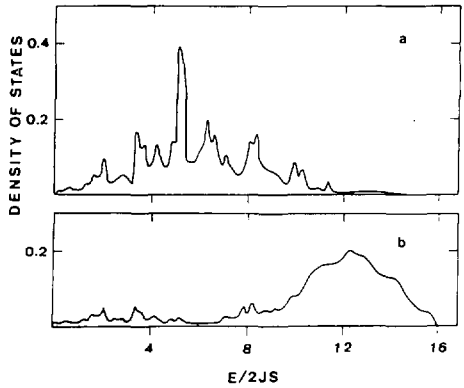

Fig. 5. (a) Local density of states at the edge atom, 7, on the surface (terrace) layer of Fig. 1. (b) Local density of states at the inner edge, atom 7 of the second layer.

The influence of the local spin-wave spectra on the local magnetic properties of the structure can be better understood by looking at the results of Fig. 6. There we have plotted, as a function of temperature, the quantity

$$M=M_0[1-\langle n_{\ell\mu}(T)\rangle] \quad , \tag{10}$$

directly related to the magnetization, where, for each atom (ℓ,μ) in the structure,

$$n_{\ell\mu}(T) = \int_0^\infty \frac{D_{\ell\mu}(\omega)d\omega}{\exp(\omega/kT)-1} \quad . \tag{11}$$

As seen in Fig. 6, all atoms close to the surface are "softer" magnetically than bulk atoms. It is easy to understand that atom 7 in the terrace, the edge atom, is appreciably softer than all others. This is due to its low coordination number, which manifests itself in the spectrum of Fig. 5(a) by the strong dominance of the $\omega\approx 5$ localized mode.

Atoms in the middle of the terrace and in the middle of the inside step, on the other hand, are practically indistinguishable from those on a plain (001) surface.

The results we have found here show how sensitive is the mag-

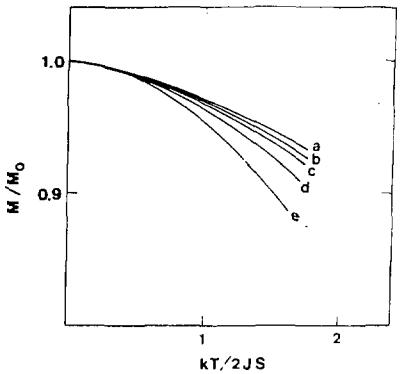

Fig. 6. Low-temperature local magnetization of various atoms as a function of temperature. (a) For bulk atoms. (b) For atom 7 of the second layer (the inner edge). (c) For atom 8 of the second layer (the side of the inner step). (d) For a surface atom in a perfect (001) surface, for atom 4 of the first layer (the middle of the terrace) and for atom 10 of the second layer (the middle of the inner step). (e) For the edge atom, atom 7 on the terrace layer.

netic structure of a solid surface to its atomic structure and geometry. We have to remark that our conclusions are based on a Heisenberg Hamiltonian model, with exchange restricted to nearest neighbors only, and with no inclusion of any reconstruction, pinning, or magnetic anisotropy.[1-3] Our calculation is also restricted to a well-defined model surface and to a reasonable sampling of the Brillouin zone. We believe, however, that we have uncovered some important points and that we have determined qualitatively the salient features of the spin-wave spectrum of stepped magnetic surfaces.

ANTIFERROMAGNETISM, PROJECTED DENSITY OF STATES AND THE BOGOLIUBOV TRANSFORMATION FOR BOSONS

We report now some surprising difficulties which are encountered in the study of an antiferromagnetic surface, when standard Green's functions techniques are used.[11] In fact we show that the procedure described above for ferromagnets, which is usually followed for antiferromagnets,[18] does not yield the wanted information or, if improperly used, may lead to unacceptable negative weights of the local spectral density of magnon states.

The starting point of our calculation is the usual Heisenberg Hamiltonian given by

$$H= \sum_{n} \sum_{\Delta} J\, \vec{S}(n)\cdot\vec{S}(n+\Delta)-2\mu_B H_A \sum_{n} S_z(n) + 2\mu_B H_A \sum_{\ell} S_z(\ell) \tag{12}$$

where μ_B is the Bohr magneton and $H_A>0$ is a fictitious magnetic field, which simulates the crystal anisotropy. The anisotropy field tends to align spins in the predominantly up-sublattice ($n\in$"up") in the (+z) direction and spins in the down-sublattice ($\ell\in$"down") in the (-z) direction. The vector Δ connects each atom with its nearest neighbors on the magnetic lattice.

For the infinite, perfectly periodic case, the standard Holstein-Primakoff and localized-to-running-wave transformations[19] yield for the Hamiltonian (12) the form

$$H= -2Nz\, J\, S^2 - 4N\mu_B H_A S + H_0 + H_1 \quad , \tag{13}$$

where

$$H_0=\varepsilon\sum_{k} (a_k^{\dagger}a_k+b_k^{\dagger}b_k)+ \sum_{k} T_k(a_k^{\dagger}b_k^{\dagger}+b_k a_k) \quad , \tag{14}$$

$$\varepsilon \equiv 2(JzS+\mu_B H_A) \quad , \tag{15}$$

$$T_k \equiv 2J\, S \sum_{\Delta} e^{i\,\vec{k}\cdot\vec{\Delta}} \quad . \tag{16}$$

The vector $\vec{k}$ is in this case a three-dimensional one. From now on we restrict our interest to the one-magnon Hamiltonian H_0, neglecting H_1 which is of fourth-order in the $\{a_k,b_k\}$ operators.

The usual procedures to handle the Hamiltonian H_0 are: a) to perform a *Bogoliubov transformation*, or b) to use Green's functions. The former method is suited only to treat the translationally invariant case, while the latter, more general and more cumbersome, has to be invoked to solve non-uniform problems.

The Bogoliubov transformation

$$\alpha_k \equiv u_k a_k - v_k b_k^{\dagger} \quad , \tag{17}$$

$$\beta_k \equiv u_k b_k - v_k a_k^{\dagger} \quad , \tag{18}$$

where u_k and v_k are real parameters which satisfy

$$u_k^2 - v_k^2 = 1 \quad , \tag{19}$$

yields

$$H_0 = \sum_k E_k(\alpha_k^\dagger \alpha_k + \beta_k^\dagger \beta_k) + \text{const.} \tag{20}$$

where $E_k \equiv \sqrt{\varepsilon^2 - T_k^2} > 0$.

Spectral weights are obtained through the imaginary part of the corresponding Green's functions. The density of antiferromagnetic magnon states in our case is related to

$$\begin{aligned} -\frac{1}{\pi}\,\mathrm{Im}\langle\langle\alpha_k|\alpha_k^\dagger\rangle\rangle\Big|_{\omega+i0^+} &= u_k^2\left[u_k\delta(\omega-E_k)-v_k^2\delta(\omega+E_k)\right] \\ &-2u_k^2v_k^2\left[\delta(\omega-E_k)-\delta(\omega+E_k)\right] \\ &+v_k^2\left[v_k^2\delta(\omega-E_k)-u_k^2\delta(\omega+E_k)\right] \\ &= \delta(\omega-E_k) \quad . \end{aligned} \tag{21}$$

Since the form of H_0 as written in (20) implies that

$$D(\omega) = 2\sum_k \delta(\omega-E_k) \quad , \tag{22}$$

where the factor of 2 takes into account both the $\alpha_k^\dagger|0\rangle$ and $\beta_k^\dagger|0\rangle$ states, we find that the end result of (21) is indeed correct. However, in obtaining this result <u>negative</u> spectral weights related to some terms had to be incorporated, which in itself is a cause of concern.

But moreover, when we related out results with what can be found in the literature, we do find additional difficulties. In fact, the local density of magnon states has been obtained[18] using

$$D_i(\omega) = -\frac{1}{\pi}\,\mathrm{Im}\left[G_{ii}^{+-}(\omega) + G_{ii}^{-+}(\omega)\right] \quad , \tag{23}$$

where

$$G^{\pm\mp}(\omega) \equiv \frac{1}{2\langle S_i^z\rangle}\langle\langle S_i^\pm|S_i^\mp\rangle\rangle_\omega \quad . \tag{24}$$

In the uniform (translationally invariant) case, relation (23) is equivalent to

$$D_i(\omega) = -\frac{1}{\pi}\,\mathrm{Tr}_k\,\mathrm{Im}\left(\langle\langle a_k|a_k^\dagger\rangle\rangle + \langle\langle b_k^\dagger|b_k\rangle\rangle\right) \quad . \tag{25}$$

If, for the sake of clarity and to keep the algebra as simple as possible we restrict ourselves to one dimension, then (24) leads to

$$D_i(\omega) = \frac{1}{\pi}\int \frac{dE}{\sqrt{4V^2-E^2}} \left[\frac{2V+E}{2E}\,\delta(\omega-E) - \frac{2V-E}{2E}\,\delta(\omega+E) + \frac{2V-E}{2E}\,\delta(\omega-E) - \frac{2V+E}{2E}\,\delta(\omega+E) \right] , \tag{26}$$

where we have defined $V \equiv 2JS$.

Carrying out the trivial integration in (26) yields the expression

$$D_i(\omega) = \frac{2}{\pi}\,\frac{2V}{\omega}\,\frac{1}{\sqrt{4V^2-\omega^2}} , \tag{27}$$

which is not the correct density of states. However, if the phase of the square root for the two last terms of the integrand in (26) are chosen to be negative then the correct density of states

$$D(\omega) = \frac{2}{\pi}\,\frac{1}{\sqrt{4V^2-\omega^2}} \tag{28}$$

is obtained. While this is the choice followed by some authors[18] it does not constitute a formally correct procedure to obtain the density of magnon states.

It is important to emphasize that the correct choice of signs is that of (26), which leads to (27) i.e. to the conclusion that $D_i(\omega) \neq D(\omega)$. In other words, $D_i(\omega)$ is not a projected density of states.

The incorrect choice of signs, which produces (28) and appears formally correct, is in fact completely wrong and leads to very serious problems in the non-uniform antiferromagnet. In fact when the spectral density of a non-uniform one-dimensional antiferromagnetic chain is evaluated on the basis of (23), with the incorrect choice of signs which yield (28) in the uniform case, unphysical negative weights of states localized at $\omega=0$ are obtained. In addition, if one carries out an analogous calculation for a Bethe lattice with coordination $z>2$ (the one-dimensional chain can be pictured as a Bethe lattice with $z=2$), then not only localized, but also extended states with negative spectral weight are obtained. This fact violates the fundamental rule[17] that $D_i(\omega)>0$, obtained from hermiticity considerations.

These results [in particular $D_i(\omega) \neq D(\omega)$] constitute a clear indication that relation (24), which exactly or with slight varia-

tions is always used in the literature, does not provide the correct way to project magnon states onto a localized basis set. On the other hand, there seems to be no trivial way to generalize the expression (25) to allow the treatment of non-uniform antiferromagnetic systems.

ACKNOWLEDGEMENTS

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