Can Surface Magnetic Order Occur?

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It is shown that the surface magnetic ordered state implied by mean field calculations in the paramagnetic temperature region does not lead to a physically inconsistent spin-wave spectrum below the surface ordering temperature, contrary to what would be expected if surface ordering were a purely two-dimensional phenomenon.

Several recent mean-field-theory calculations of the behavior of semi-infinite magnetic systems with one free surface have found that the susceptibility is divergent at the surface in a regime where the bulk system is still paramagnetic, which implies that there has been a phase transition to a state of surface magnetic order. Such results have been found for Heisenberg spin Hamiltonians with a stronger coupling between spins in the surface plane than between other pairs of spins[1-3] and in itinerant electron models.[4-6] These surface magnetically ordered states are presumably essentially two-dimensional (2D) in nature. It has thus been assumed that they are artifacts of an inconsistent 2D mean field theory, since the spin-wave spectrum, in mean field theory, of a purely 2D Heisenberg or itinerant electron magnetic system is physically inconsistent: There is an infinite density of low-energy spin waves.[7] In this paper we shall consider the mean-field-theory spin-wave spectrum of a Heisenberg ferromagnet in the surface ordered state and establish a finite upper bound on the spin-wave density. Hence the physical inconsistency of 2D mean field theories does not apply to these surface magnetic states and they cannot be ruled out on theoretical grounds.

Consider a simple cubic lattice with a free $\{100\}$ surface and nearest-neighbor ferromagnetic Heisenberg coupling between the spins. The exchange coupling is J_s between spins in the surface plane and J between all other pairs of spins. For $J_s > 1.25J$ mean field theory predicts a surface magnetically ordered state for $T_c{}^s > T > T_c$, T_c being the bulk critical temperature.[1-3] An approximate calculation of the spontaneous magnetization in this state yields[2,3,8]

$$S_l = Se^{-\xi(l-1)}, \tag{1}$$

where l is an index labeling the crystal planes in the direction perpendicular to the surface, which is at $l=1$, S_l is the spontaneous magnetization in the lth plane, S the spontaneous magnetization in the surface plane, and ξ is a temperature-dependent and coupling-constant–dependent inverse range.

The spin-wave spectrum for the surface ordered state is obtained by considering the Green's function

$$G_{i,j}(t) = -i\langle\{S_+(\vec{R}_i, t)S_-(\vec{R}_j, 0)\}\rangle. \tag{2}$$

Taking the Fourier transform of G with respect to time and the direction parallel to the surface, one obtains a function $G(l, l')$, with l and l' plane indices (dropping the variables ω and $\vec{k}_\parallel$ from the notation). The mean field equations for G are

$$(\omega - \epsilon_1)G(1, l') + JS_1 G(2, l') = S_1\delta_{1,l'}, \tag{3a}$$

$$(\omega - \epsilon_l)G(l, l') + JS_l[G(l+1, l') + G(l-1, l')] = S_l\delta_{l,l'}, \quad l \geq 2, \tag{3b}$$

where

$$\epsilon_1 = JS(j\epsilon_\parallel + e^{-\xi}), \tag{4a}$$

$$\epsilon_l = JS\epsilon e^{-\xi(l-1)}, \quad l \geq 2, \tag{4b}$$

$$\epsilon = \epsilon_\parallel + 2\cosh\xi, \tag{4c}$$

$$\epsilon_\parallel = 4 - 2\cos k_x - 2\cos k_y, \tag{4d}$$

where $j = J_s/J$, and k_x and k_y are components of $\vec{k}_\parallel$ in units of the inverse lattice spacing. These equations can be solved by an infinite interation: Defining the continued fractions $a_l(\omega)$ and $b_l(\omega)$ in terms of the recursion relations

$$a_l(\omega) = [\omega - \epsilon_l - J^2 S_l S_{l+1} a_{l+1}(\omega)]^{-1}, \tag{5a}$$

$$b_l(\omega) = [\omega - \epsilon_l - J^2 S_l S_{l-1} b_{l-1}(\omega)]^{-1}, \tag{5b}$$

$$b_1(\omega) = (\omega - \epsilon_1)^{-1} \tag{5c}$$

(the a_l's are infinite continued fractions and the b_l's are finite continued fractions), one finds

$$G(l, l') = S_l a_1(\omega) \prod_{m=2}^{l} [a_m(\omega)/b_{m-1}(\omega)][\sum_{n=1}^{l} (-1)^{l-n}\delta_{n,l'} \prod_{m=n}^{l-1} JS_m b_m(\omega) + \sum_{n=l+1}^{\infty} (-1)^{l-n}\delta_{n,l'} \prod_{m=l+1}^{n} JS_m a_m(\omega)], \tag{6}$$

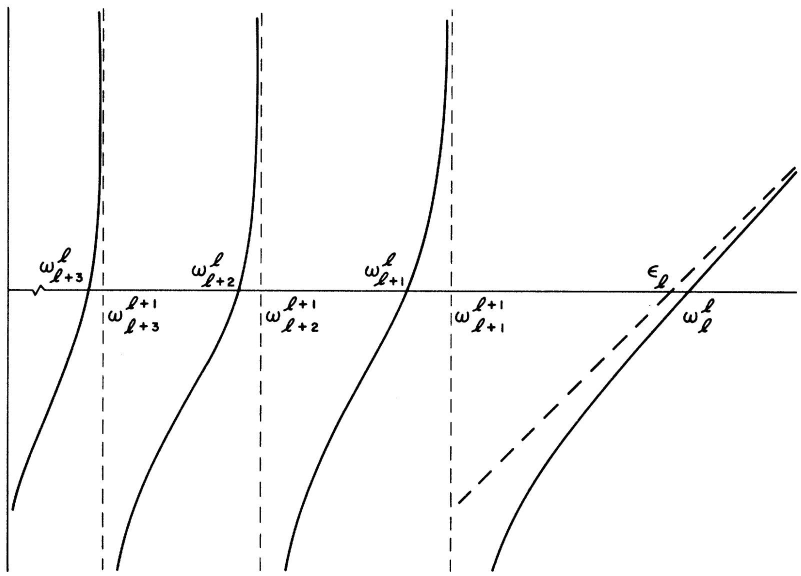

FIG. 1. Solid curves, plot of $a_l^{-1}(\omega)$. The dashed slanted line is $\omega-\epsilon_l$; the dashed vertical lines indicate the poles of $a_{l+1}(\omega)$.

where I use the convention

$$\prod_{i=n}^{n-1} f_i = 1.$$

The spin-wave spectrum for the surface magnetically ordered state is given by the poles of the functions $a_l(\omega)$ and $b_l(\omega)$. Let us concentrate on the spectrum of $a_1(\omega)$, which will give the spin-wave density in the surface plane. Since $\epsilon > 2\cosh\xi > 2$, let us use the approximate inequality $\epsilon^{-2} \ll 1$ in the subsequent analysis. The further assumption that $\exp(-\xi) \ll 1$ enables us to arrive at a reasonably simple form for $a_1(\omega)$. One then finds that

$$a_l(\omega) = \sum_{n=0}^{\infty} R_{l+n}{}^l/(\omega - \omega_{l+n}{}^l),$$
$$\omega_{l+n}{}^l = \epsilon_{l+n}[1 + O(\epsilon^{-2})], \tag{7}$$
$$R_{l+n}{}^l = (e^{-\xi}/\epsilon^2)^n[1 + O(\epsilon^{-2n})],$$

satisfies Eq. (5a) for $l \geq 2$. To demonstrate this, consider

$$a_l^{-1}(\omega) = \omega - \epsilon_l - J^2S^2e^{-\xi(2l-1)}a_{l+1}(\omega), \tag{8}$$

plotted in Fig. 1. This function has simple poles at $\omega = \omega_{l+n}{}^{l+1}$, $n \geq 1$, and the zeros of a_l^{-1} are the poles of a_l. Since $a_{l+1}(\omega)$ is relatively slowly varying for $\omega \simeq \epsilon_l$, one may evaluate it at ϵ_l to find

$$\omega_l{}^l = \epsilon_l[1 + O(e^{-\xi}/\epsilon^2)]. \tag{9}$$

If $\omega_{l+n}{}^l$ is very close to $\omega_{l+n}{}^{l+1}$ the pole in Eq. (8) at $\omega = \omega_{l+n}{}^{l+1}$ will be the dominant term in determining $\omega_{l+n}{}^l$. Approximating the other terms in $a_l^{-1}(\omega)$ by their value at $\omega_{l+n}{}^{l+1}$ one finds, using $\exp(-\xi)$ and ϵ^{-2} small,

$$\omega_{l+n}{}^l = \omega_{l+n}{}^{l+1}(1 - \epsilon^{-2n}) \simeq \epsilon_{l+n}. \tag{10}$$

This result does give $\omega_{l+n}{}^l$ much closer to $\omega_{l+n}{}^{l+1}$ than to any of the other poles in a $a_l^{-1}(\omega)$, justifying the single-pole approximation used to establish Eq. (10). To complete the demonstration that Eq. (7) is an approximate solution of the recursion relation, one must calculate

$$R_{l+n}{}^l = [1 + J^2S^2e^{-\xi(2l-1)} \sum_{m=0}^{\infty} (e^{-\xi}/\epsilon^2)^m(\omega_{l+n}{}^l - \omega_{l+1+m}{}^{l+1})^{-2}]^{-1}. \tag{11}$$

For $n=0$ all the terms in the sum are of order $\exp(-\xi)/\epsilon^2$ or smaller, giving $R_l{}^l = 1$ in agreement with Eq. (7). For $n \geq 1$ the dominant term in Eq. (11) is $m = n-1$, where the frequency denominator is smallest, giving

$$R_{l+n}{}^l = (e^{-\xi}/\epsilon^2)^n[1 + O(\epsilon^{-2n})],$$

again in agreement with Eq. (7).

Equation (7) is valid for $l \geq 2$; a somewhat different result applies to $a_1(\omega)$ because of the possibility that $z = \epsilon_1/JS$ may be less than 1. It is then possible for ϵ_1 to be less than $\omega_2{}^2$. This turns out not to be a serious problem since all it does is confuse the ordering of the high-frequency spin-wave modes in a_1. Since $\epsilon > 2$, no matter what the value of $\epsilon_\parallel$, these modes will give a finite contribution to the surface spin-wave density. I thus concentrate on the poles at ω_n^2, $n \geq N$, where N is at worst a large but finite integer such that $\omega_n^2 < \epsilon_1$. A plot of $a_1^{-1}(\omega)$ in this frequency range will then be very similar to Fig. 1 and an analysis similar to that above yields

$$\omega_n^1 = \omega_n^2[1 - \epsilon^{-2(n-1)}/(z - \epsilon^{-2})], \tag{12}$$

where we have kept an $O(\epsilon^{-2})$ correction. If $\epsilon^{-2(n-1)}/(z - \epsilon^{-2}) \ll 1$, Eq. (12) is consistent with the assumptions used in deriving it and one finds

$$R_n^1 = (e^{-\xi}/\epsilon^2)^{n-1}\epsilon^4/(z\epsilon^2 - 1)^2. \tag{13}$$

The minimum value for $z\epsilon^2 - 1$ is $\exp(-2\xi)$, which occurs when $\epsilon_\parallel = 0$. Since this is finite and positive, Eqs. (12) and (13) will be correct for large enough n.

The contribution of the low-frequency spin-wave modes to the spin-wave density in the surface is proportional to

$$\int d\epsilon_\parallel \, \rho(\epsilon_\parallel) \sum_{n=N}^{\infty} R_n^1(\epsilon_\parallel)/\omega_n^1(\epsilon_\parallel), \tag{14}$$

where $\rho(\epsilon_\parallel)$ is the planar density of states. A conservative upper bound on the sum in Eq. (14) is obtained by using Eqs. (12) and (13), with $\omega_n^1 \simeq \epsilon_n$, all evaluated at $\epsilon_\parallel = 0$. This gives an upper bound proportional to $\exp[-(2N-9)\xi]/JS$ on Eq. (14). Hence the contribution of the low-energy modes to the spin-wave density in the surface plane is finite, contrary to the result which holds for a purely 2D system.

The approximations which have been used in arriving at this finite upper bound on the spin-wave density have principally resulted in a consistent overestimate of the R_{l+n}^l's. For large n this cannot have a serious effect, even if one relaxes the conditions $\exp(-\xi)$ and $\epsilon^{-2} \ll 1$, since the shifts obtained are of order $\epsilon^{-2n} \ll 1$ in any case. The major changes due to keeping all powers of $\exp(-\xi)$ and ϵ^{-2} will occur at small n. There, so long as $\exp(-\xi)$ and ϵ^{-2} are less than one, these changes will be of order unity, i.e., the worst that will happen is $\omega_{l+n}^l = \alpha\epsilon_{l+n}$, α of order unity if n is small. Using this as a starting point and repeating the analysis of the spectrum of $a_l(\omega)$ for ω_{l+n}^l at larger n one arrives at a finite upper bound on the spin-wave density in the surface plane. Furthermore, I have carried out a continuum approximation to Eqs. (3), appropriate to the limit $\xi \ll 1$ or $\exp(-\xi) \lesssim 1$, which yields the same result as the above analysis: a finite density of low-energy spin waves. In addition, I have confirmed that the thermodynamic limit of a true semi-infinite system is essential for the result: The application of the above analysis to a film with a finite number of planes gives an infinite density of low-energy spin waves. Details of these calculations will be given in a future publication.

I have shown in this paper that one of the objections to the mean field theory prediction of surface magnetic order for semi-infinite Heisenberg ferromagnets is not valid: Surface magnetic order is not a purely 2D phenomenon and the physical intuition one has, based on the behavior of purely 2D systems, may not be appropriate for surface phenomena in semi-infinite systems. Whether surface magnetic order can actually occur requires a more exact theoretical treatment than mean field theory, but my conclusion that the fluctuations about the spontaneous magnetization do not have an infinite density should also hold in more exact theories. Hence I expect these more exact theories of critical behavior to give surface order for a sufficiently large value of J_s/J. Note that the spherical model with short-range forces is similar to the Heisenberg model in that it does not order in the bulk unless the thermodynamic limit is taken in all three dimensions; preliminary results of a study of the spherical model with an increased exchange interaction in the surface plane[9] show a surface phase transition before the bulk transition when the thermodynamic limit is taken in all three dimensions.

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