

Surface Magnetism in a Thin Film of Heisenberg Ferrimagnets

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Magnetization of every spin layer is calculated self-consistently in thin films of two-sublattice Heisenberg ferrimagnets by means of the retarded Green's function equation of motion. Both lattice structures of CsCl and NaCl types are considered and results from slabs with different surface spins and different thickness are presented and discussed. The surface-induced quantum fluctuation is found to be absent in simple cubic lattices. Spin wave spectra are also obtained and differences from previous results are noted. Our formulation includes antiferromagnetic films as a special case in which the mean spins of the two sublattices are equal but opposite.

I. INTRODUCTION

There has been growing interest in spin waves in ferrimagnetic systems of confined geometry in recent years. A ferrimagnet has, in general, multilattice structures and the sublattices do not have their mean magnetization vectors adding up to zero. On the other hand, the mean magnetizations of the two sublattices in antiferromagnets add up to zero. Simple two-sublattice models have been constructed to study surface spin waves (SSW) in semi-infinite ferrimagnetic systems by assuming cubic sublattice structures [1-3]. Although ferrimagnets are actually much more complicated, basic features of magnetic properties such as the layer magnetization and spin wave spectra obtained in these models are qualitatively correct. Very recently, interface spin waves (ISW) are investigated for a system of two different semi-infinite ferrimagnets [4] and for a heterostructure of two ferrimagnetic thin films [5].

For the geometry of a slab, most studies have thus far been limited to ferromagnets and antiferromagnets [6-10]. Only recently, spin wave spectra for a ferrimagnetic slab have been calculated in the uniform magnetization approximation (UMA) [11]. Since the surfaces reduce the symmetry of the system under consideration, physical quantities in the vicinity of surfaces generally deviate from their bulk values. Surface magnetization, in particular, has attracted much attention in recent years, both experimentally [12-16] and theoretically [17,18] because of the controversial dead layer.

The spin waves and magnetic properties of bulk ferrimagnets have been treated on the two-sublattice Heisenberg model by the method of retarded Green's function equation of motion [19]. The method introduced in Ref. 19 simplifies greatly the algebraic procedure and is particularly useful in the treatment of SSW [2] and ISW [4,5] in ferrimagnetic systems. We shall make liberal use of the results of Ref. 19 which will be referred to as I from now on.

We consider, in this paper, ferrimagnetic slabs with (001) free surfaces. The slab has a total of N atomic layers. Two different kinds of magnetic ions a and b with spins \vec{S}_a and \vec{S}_b occupy the alternative layers forming the a -sublattice and b -sublattice respectively. Thus, every ion except for those on surfaces has eight nearest neighbors of the other kind in CsCl structure and six in NaCl structure. A schematic diagram of the spin arrangement in these structures is illustrated in Fig. 1. The NaCl structure is actually a simple cubic crystal with a face-centered cubic Bravais lattice for each sub-lattice. The CsCl structure,

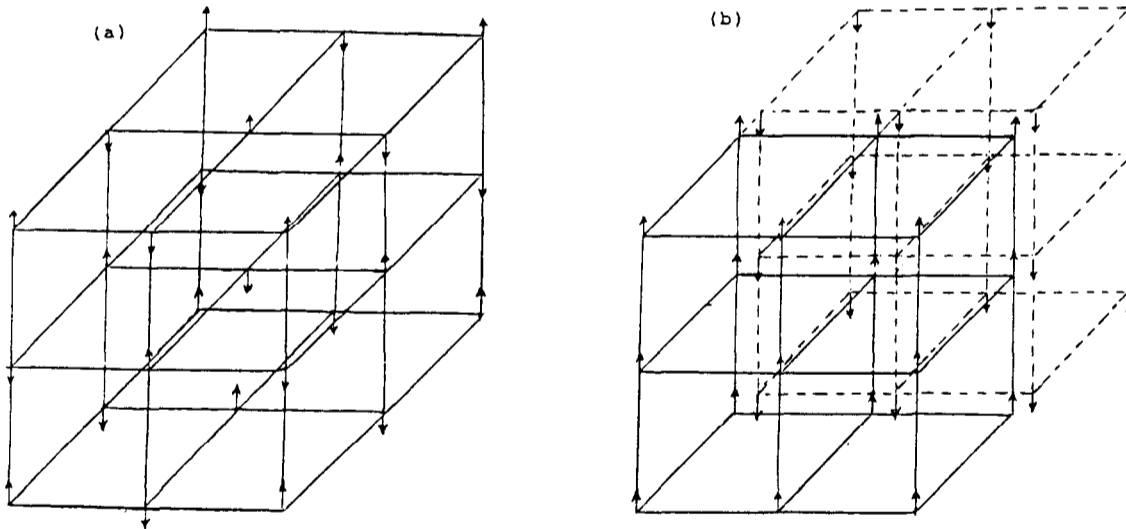


FIG. 1. Arrangement of spins in a ferrimagnet of (a) NaCl structure and (b) CsCl structure. The spins in the two sublattices are opposite in direction, and are in general not equal in size.

on the other hand, is a body-centered cubic crystal with each sublattice being a simple cubic Bravais lattice. For definiteness, we label the atomic layers from the left by $1, 2, 3, \dots, N$. Unless otherwise defined, we follow the notation of I.

II. OUTLINE OF THE THEORY

The Heisenberg model Hamiltonian for a two-sublattice ferrimagnet with nearest-neighbor exchange interactions is given by

$$H = J \sum_{(\vec{a}, \vec{b})} \vec{S}_{\vec{a}} \cdot \vec{S}_{\vec{b}} \quad (1)$$

where $\vec{S}_{\vec{a}}(\vec{S}_{\vec{b}})$ is the spin vector located at the lattice site \vec{a} (\vec{b}) in the a- (b-) sublattice, $\sum_{(\vec{a}, \vec{b})}$ means the summation over each pair only once, and J denotes the exchange integral and is assumed to be constant.

For convenience, we make use of the translational symmetry in the plane of surfaces and introduce the Bloch-wannier representation in which the Bloch function is used in the xy-plane and the Wannier function is used in the z direction. The two-dimensional wave vector \vec{k} is defined by $\vec{k} = (\vec{k}, q) = (k_x, k_y, q)$. We denote the layer magnetization in the CsCl structure by μ_m and in the NaCl structure by μ_{2m-1} and μ_{2m} for the two sublattices with m labelling the layer, where $\mu = \langle S^z \rangle$.

Equations of motion for operators $\vec{S}_{\vec{a}}$ and $\vec{S}_{\vec{b}}$ are obtained by the random phase approximation (RPA) decoupling procedure [20,21] in terms of the retarded Green's function. By introducing the two-dimensional Fourier transform in the plane parallel to surfaces, the equations of motion can be expressed in terms of the Fourier components of the retarded Green's function, namely, in terms of the Green's function $g_{ij}(\vec{k}, E; m, n)$ where i, j label the sublattices and m, n label the spin layers in the film.

For the CsCl-structure film with the surface layer belonging to the a-sublattice, we find the Green's function equations of motion

where $\xi(\vec{\kappa}) = \frac{1}{2}(\cos k_x d + \cos k_y d)$.

The layer magnetization can be calculated from the Green's functions found in Eqs. (2) and (3). For this purpose, we need the auxiliary functions [19-21]

$$\Phi_\ell^m(T) = \frac{i}{2\pi N_0} \int_{-\infty}^{\infty} \frac{dE}{e^{E/k_B T} - 1} \sum_{\vec{\kappa}} \left(g_{\ell\ell}(\vec{\kappa}, \vec{E} + i0^+; m, m) - g_{\ell\ell}(\vec{\kappa}, \vec{E} - i0^+; m, m) \right) \quad (4)$$

where N_0 denotes the total number of unit cells, k_B is the Boltzmann constant and ℓ indicates the sublattice to which the layer m belongs. The sum is over the first Brillouin zone. The magnetization μ_ℓ^m for the layer m in the sublattice ℓ is given by

$$\mu_\ell^m \approx \frac{[S_\ell - \Phi_\ell^m(T)][1 + \Phi_\ell^m(T)]^{2S_\ell+1} + [S_\ell + 1 + \Phi_\ell^m(T)]^{2S_\ell+1}}{[1 + \Phi_\ell^m(T)]^{2S_\ell+1} - [\Phi_\ell^m(T)]^{2S_\ell+1}} \quad (5)$$

where the lattice spin S_ℓ may be arbitrary.

The Green's functions are obtained by solving Eqs. (2) and (3), each of which can be written as two sets of coupled matrix equations. When the eigenvalues of the coefficient matrix X are nondegenerate, the Green's function takes according to the Gramer rules the following form

$$g_{\ell\ell}(\vec{\kappa}, E; m, m) = \frac{\det |X_m(E)|}{\det |X(E)|} = \sum_i \frac{\det |X_m(E_i)|}{\prod_{j \neq i} (E_i - E_j)(E - E_i)} \quad (6)$$

where E_i and E_j are the nondegenerate eigenvalues of X unless $\eta(\vec{\kappa}) = 0$ or $\xi(\vec{\kappa}) = 0$, and X_m is the matrix X with the m th column replaced by the column of constant on the right-hand side of the linear equation set. When $\eta(\vec{\kappa}) = 0$, the matrix X for CsCl structure becomes diagonal and the spins are effectively decoupled. Therefore the Green's function in this case takes the form

$$g_{\ell\ell}(\vec{\kappa}, E; m, m) = \frac{1}{X_{mm}}. \quad (7)$$

After the layer magnetizations are calculated self-consistently, we find a twofold degeneracy in energy for films of odd number layers but no degeneracy for films of even number layers. In the case of NaCl structure, two-fold degeneracy is found only in films of even number layers when $\xi(\vec{\kappa}) = 0$. As a matter of fact, the a-sublattice and b-sublattice become effectively decoupled in this case. And Eq. (6) can still be applied to calculate the Green's functions.

III. RESULTS AND DISCUSSION

We start with a given set of either single particle spins \vec{S}_a and \vec{S}_b , or mean layer magnetizations in the bulk as computed in I. We then solve Eqs. (2) and (3) for Green's functions for CsCl and NaCl structures, respectively. These Green's functions are substituted in Eq. (4) to obtain the auxiliary function Φ_ℓ^m from which we finally find a new set of magnetizations. The same procedure is repeated until self-consistency is achieved. The spin wave energies are determined by the poles of Green's functions.

In the case of CsCl structure, the surface layers belong to the same or different sublattices if the film has an odd or even number of layers. There is no such difference in the NaCl structure. Numerical computation has been carried out for all cases with various choices of parameters S_a, S_b and N . Spin wave spectra obtained this way are qualitatively the same as those from the GMA [11], and therefore are not reproduced here. However, a few comments about surface spin waves are in order at this point. In general, there are two sets of SSW corresponding to the two surfaces. They approach to each other as N increases and eventually become degenerate, indicating the mutual influence between the two surfaces of a thin film. In the limit of large N , say, $N \geq 20$, the spectra are shown in Fig. 2. In the case of NaCl structure, there is no qualitative difference from the semi-infinite results [2] except for the SSW energy separations from the corresponding bulk spectra. In the case of CsCl structure, however, qualitative difference is observable. Both the acoustic and optical branches of the SSW's are localized in the whole Brillouin zone. In addition, there appear two more SSW's in the small η regime as shown in Fig. 2(a). These SSW's disappear into the bulk spectrum as κ decreases and no solution can be found when $\kappa \rightarrow 0$. It is therefore not possible to label them as the optical or acoustic branch.

Surface effects on the magnetization are obviously seen in our numerical studies. The deviations of layer magnetizations from the bulk value are calculated for both structures. In the case of CsCl structure, some typical results are illustrated in Fig. 3 in which deviations from the corresponding bulk magnetizations [19] are plotted for every layer in various thin films.

It is observed that the surface magnetization is greater than the second layer magnetization which is in turn smaller than the third layer magnetization. This unusual oscillating behavior appears near the surfaces for every film of CsCl structure we have considered. It does not matter whether the film has the left-right symmetry or not. The phenomenon has been noted in a recent publication [10] in which the layer magnetizations in an antiferromagnetic film with body-centered cubic structure have been calculated.

Since there is no such phenomenon in classical theory for either ferromagnetic or antiferromagnetic thin films, and since Monte Carlo simulations with classical spins do not show this behavior at low temperatures, it is interpreted as a result of quantum fluctuation.

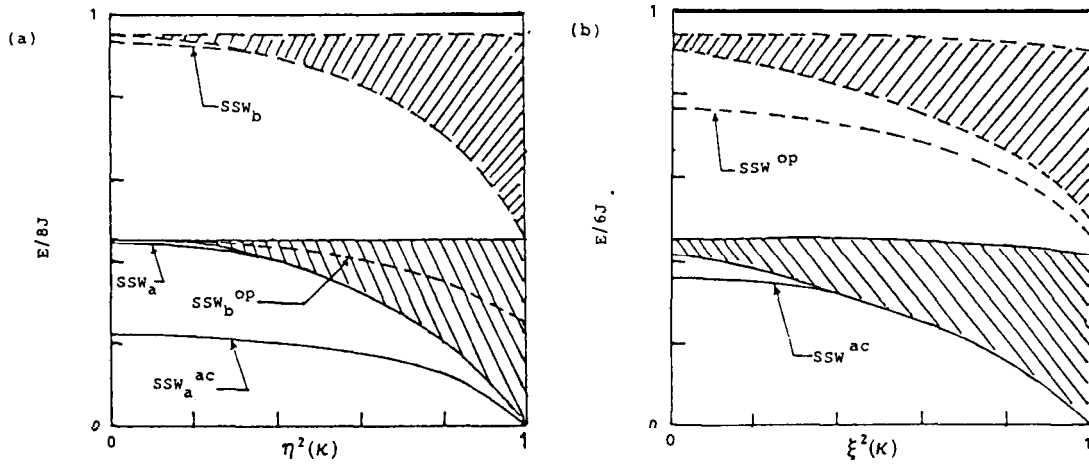


FIG. 2. Spin wave spectra for slabs in the large N limit. (a) CsCl structure and (b) NaCl structure. $S_A = 1, S_b = 0.5$ and $\langle S^z \rangle_a = \mu_{bulk}^a = 0.958$ and $\langle S^z \rangle_b = \mu_{bulk}^b = 0.461$. Solid (dashed) lines represent positive (negative) frequency branches of the spectra. The shaded area show the corresponding bulk spectrum

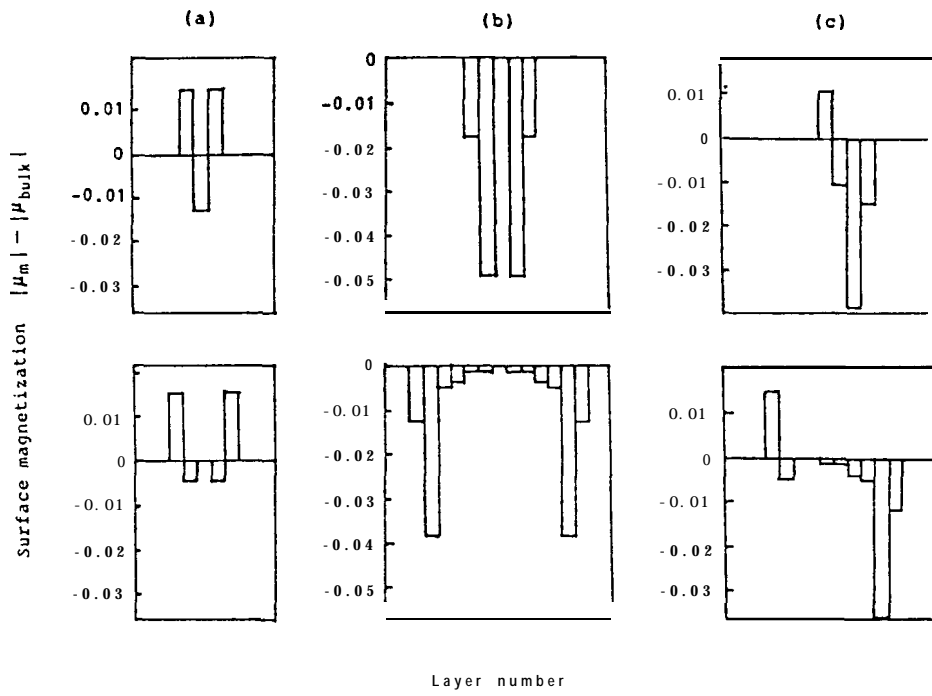


FIG. 3. Magnetization of spin layers in films of various thickness. The lattice structure is of CsCl type and $S_a = 1, S_b = 0.5$ (a) a-sublattice surfaces, (b) b-sublattice surfaces and (c) a-sublattice surface on the left and b-sublattice on the right.

It is convenient to introduce the range λ of the surface effect into the bulk. We find in our study that λ depends sensitively on the relative surface magnetizations of the two sublattices. If we define $\alpha = \mu_1/\mu_2$, i. e., the ratio between magnetizations of layer 1 and layer 2, the range λ is found to increase with increasing α for $\alpha < 1$, and decrease with increasing α for $\alpha > 1$. Thus, λ is largest for $\alpha = 1$. In other words, effects of the surface reconstruction penetrate deepest in antiferromagnetic films.

For the case of NaCl structure, the situation is much simpler. Fig. 4 depicts the surface reconstruction for two typical cases. First of all, we note that there is no anomalous phenomenon of oscillating behavior near the surfaces. Thus the interpretation of quantum fluctuation effect as suggested in Ref. 10 may not be valid in general. At least there is no sign of such effect in films of simple-cubic lattice structure. Why the phenomenon exists only in ferrimagnetic or antiferromagnetic films of body-centered cubic structure remains to be understood.

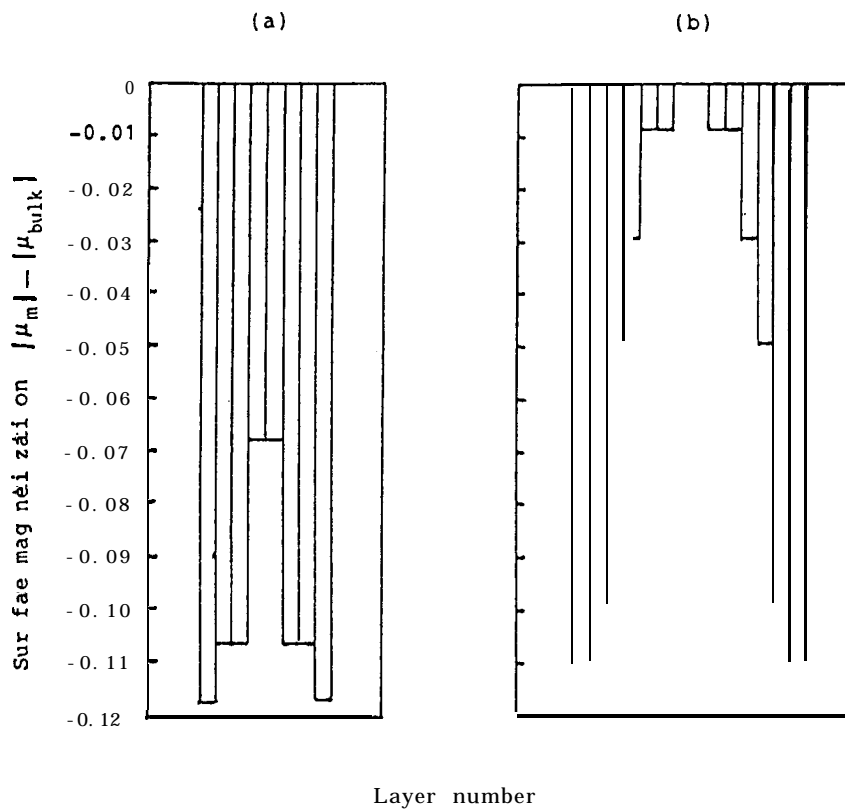


FIG. 4. Magnetization of spin layers in films with NaCl structure. $S_a=1, S_b=0.5, \mu_{bulk}^a=0.944$ and $\mu_{bulk}^b=0.449$. (a) $N=4$ and (b) $N=7$.

It is also seen that the deviation from the bulk magnetization is generally much smaller than the previous case except for the surface layer. This is not difficult to understand because an individual spin on the surface layer has only one out six nearest neighbors missing in this case while 4 out of 8 nearest neighbors of a surface spin are cut off in the case of CsCl structure. As the final remark, we emphasize that our results apply directly to antiferromagnetic films if we take the mean magnetizations of the two sublattices to be equal and opposite in direction.

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