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Effects of surface exchange anisotropy in Heisenberg ferromagnetic insulators

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We consider an fcc semi-infinite ferromagnetic insulator displaying an anisotropic exchange interaction between spins on the (111) surface plane of the form $J_{||}(S_i^xS_j^x+S_i^yS_j^y+\eta S_i^zS_j^z)$. We assume all other interactions isotropic. A self-consistent random-phase-approximation calculation is performed, with a Green's-function method valid for any spin S, up to the bulk transition temperature T_c^b, by the assumption that the magnetization of the third layer equals the bulk value. For η sufficiently large, the surface magnetization is nonzero for $T>T_c^b$, up to a transition temperature $T_c^s(\eta)$ whenever $\eta\geq\eta_c>1$, where $T_c^s(\eta_c)=T_c^b$. For $T>T_c^b$ the system is equivalent to a film of three layers, where the magnetization of the third one is identically zero as a boundary condition. A discontinuity of the derivative in the curve of the magnetization of the first two layers versus temperature is found at T_c^b. The results show clearly a crossover from Heisenberg to Ising behavior at the surface.

I. INTRODUCTION

In a previous paper we have obtained the layer magnetization and the spectrum of excitations of a Heisenberg semi-infinite ferromagnet with isotropic exchange interactions. There it was found, through the use of the random-phase approximation (RPA) for evaluating the one-spin Green's functions, that the bulk behavior drives the surface magnetic for all values of the exchange coupling $J_\perp$ of surface to bulk.[1] It was also found, however, that when $J_\perp$ decreases the surface magnetization σ_0 decreases as well, and the results indicate that $\lim_{J_\perp\to 0}\sigma_0\to 0$ at all finite temperatures, showing clearly a dimensional crossover from three- to two-dimensional behavior as $J_\perp\to 0$. In other words, it is possible to realize, by adequate parameters, a system where a surface region of a few atomic layers is almost paramagnetic, that is where the surface region has a very small magnetization at all finite temperatures for which the bulk is ferromagnetic.

It is natural to ask whether it is also possible to realize the opposite case, i.e., that of a ferromagnetic surface film over a paramagnetic bulk. We know that a completely isotropic exchange interaction between surface spins will not sustain long-range order at a finite temperature,[2] but that a purely uniaxial interaction, the Ising model, orders for $T>0$ in two dimensions. Therefore, a necessary condition for the surface to be ferromagnetic at a finite temperature is the introduction of uniaxial anisotropy in such a way that the exchange interaction of two spins i and j on the surface is of the form

$$J_{||}(S_i^xS_j^x+S_i^yS_j^y+\eta S_i^zS_j^z)\ .$$

In this paper we show that, in effect, as η becomes larger than one the surface layers tend to decouple from the bulk, their magnetization increasing accordingly. We also find that, as η increases, more and more layers can sustain spontaneous magnetization for $T>T_c^b$, these layers displaying the proximity effect of a ferromagnetic film on an underlying paramagnetic bulk.[3] Similar results were found by Sarmento *et al.*[4] for a semi-infinite Ising ferromagnetic ($S=\frac{1}{2}$). In Sec. II we develop the theoretical framework including a short summary of the main point of Ref. 1, and we state the present results. In Sec. III we discuss the results obtained.

II. METHOD OF CALCULATION AND RESULTS

The Hamiltonian

$$H=-\sum_{i\neq j}I_{ij}\{\tfrac{1}{2}(S_i^+S_j^-+S_i^-S_j^+)+\eta_{ij}S_i^zS_j^z\}\ , \tag{1}$$

where $\eta_{ij}=1$ if point i or j is not on the surface, and $\eta_{ij}=\eta>1$ if both points i and j are near neighbors on the surface. We consider, in what follows, an fcc ferromagnet with a (111) surface and we define the parameters as in Ref. 1. The equations of motion for the Green's functions Fourier transformed over time and the coordinates parallel to the surface are[1]

$$\sum_m \left[\left[\nu - 2\sum_l \sigma_l \eta_{il}\epsilon_{il}(\vec{k}_{||}=0) \right] \delta_{im} + 2\sigma_i \sum_l \epsilon_{lm}(\vec{k}_{||})\delta_{il} \right] \times g_{mj}(\omega,\vec{k}_{||}) = \frac{\sigma_i}{\pi}\delta_{ij} , \quad (2)$$

$$\sigma_i = \frac{\langle S_i^z \rangle}{h}, \quad \epsilon_{il}(\vec{k}_{||}) = \frac{I_{il}(\vec{k}_{||})}{I}, \quad \nu = \frac{\omega}{I} , \quad (3)$$

where

$$I_{ll}(\vec{k}_{||}) = \begin{cases} 3I_{||}(3\phi^2-1), & l=0 \text{ (surface layer)} \\ 3I(3\phi^2-1), & l>0 \end{cases}$$

$$I_{l,l+1}(\vec{k}_{||}) = \begin{cases} 3I_{\perp}\phi, & l=0 \\ 3I\phi, & l>0 \end{cases} \quad (4)$$

$$\phi = \tfrac{1}{3}\{3+2[\cos 2\pi(k_1-k_2) + \cos 2\pi k_1 + \cos 2\pi k_2]\}^{1/2} .$$

We define the matrix

$$\Omega_{im} = \left[\nu - 2\sum_l \sigma_l \eta_{il}\epsilon_{il}(\vec{k}_{||}=0) \right] \delta_{im} + 2\sigma_i \sum_l \epsilon_{lm}(\vec{k}_{||})\delta_{il} . \quad (5)$$

In the particular case where only σ_0 and σ_1 are different from the bulk magnetization σ, Ω reduces to the following:

$$\Omega = \begin{vmatrix} \nu-a_{00} & b_{01} & & & \\ b_{10} & \nu-a_{11} & b_{12} & & \\ & b & \nu-a_{22} & b & \\ & & b & \nu & b\cdots \\ & & & \vdots & \vdots \end{vmatrix} , \quad (6)$$

where

$$\begin{aligned} a_{00} &= 6\{\epsilon_{||}\sigma_0[2(\eta-1)+4\Lambda(\vec{k}_{||})]+\epsilon_{\perp}\sigma_1\} , \\ a_{11} &= 6[\epsilon_{\perp}\sigma_0+4\Lambda(\vec{k}_{||})\sigma_1] , \\ a_{22} &= 6[\sigma_1+4\sigma\Lambda(\vec{k}_{||})] , \\ b_{01} &= -6\phi\epsilon_{\perp}\sigma_0, \quad b_{10}=-6\phi\epsilon_{\perp}\sigma_1 , \\ b_{12} &= -6\phi\sigma_1, \quad b=-6\phi\sigma , \\ \Lambda &= \tfrac{3}{4}[1-\phi^2(\vec{k}_{||})] . \end{aligned} \quad (7)$$

As in our previous work,[1] the magnetization of a given plane is obtained through the application of the relations derived by Hewson and Ter-Haar,[5]

$$\langle S_l^z \rangle = \frac{[S-\psi_l(S)][1+\psi_l(S)]^{2S+1}+[S+1+\psi_l(S)][\psi_l(S)]^{2S+1}}{[1+\psi_l(S)]^{2S+1}-[\psi_l(S)]^{2S+1}} , \quad (8)$$

where

$$\psi_l(S) = -\lim_{\epsilon\to 0+} \frac{1}{\pi}\int_{-\infty}^{\infty} d\omega \frac{a^2}{4\pi^2}\int \frac{d^2\vec{k}_{||}\,\mathrm{Im}[\Omega^{-1}(\omega+i\epsilon)]_{ll}}{e^{\beta\omega}-1} . \quad (9)$$

As can be seen from Fig. 1, the results in this case, when the surface anisotropy η is sufficiently large, are quite new in respect to what has been found in Ref. 1. We measure temperature in units of $12\hbar^2 I/k_B$.

Let us call T_c^b the transition temperature of the infinite system, which can be calculated as in the paper of Tahir-Kheli and Ter-Haar,[6] and also as in the RPA. We find that as $T\to T_c^b$, the magnetization of the first and second layers, σ_0 and σ_1, respectively, *does not vanish.*

In order to obtain σ_0 and σ_1 for $T>T_c^b$, we refer to Eq. (6), where we see that if $\sigma=0$, the matrix Ω

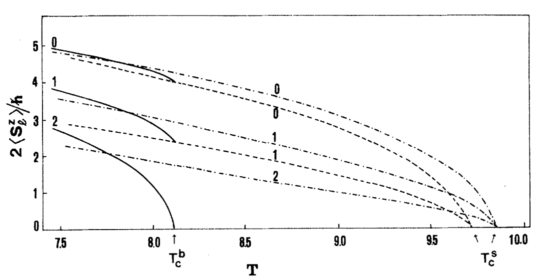

FIG. 1. Magnetization of the first two layers and the bulk vs T for $\eta=1.8$, $\epsilon_{||}=\epsilon_{\perp}=1$. Temperature in units of $12\hbar^2 I/k_B$. Case a: continuous curves. Case b: four layers (dash-dotted lines); three layers (dashed lines).

simplifies, and one has only to deal now with a 3×3 submatrix of Ω, namely

$$\begin{bmatrix} \nu-a_{00} & b_{01} & \\ b_{10} & \nu-a_{11} & b_{12} \\ & 0 & \nu-a_{22} \end{bmatrix} . \quad (10)$$

It is now immediate to calculate Ω^{-1}. It is found that

$$(\Omega^{-1})_{00}=\frac{\nu-a_{11}}{(\nu-a_{11})(\nu-a_{00})-b_{10}b_{01}} , \quad (11)$$

$$(\Omega^{-1})_{11}=\frac{\nu-a_{00}}{(\nu-a_{11})(\nu-a_{00})-b_{10}b_{01}} . \quad (12)$$

These expressions are substituted in Eq. (9) to evaluate $\psi_l(S)$, $(l=0,1)$, and the two self-consistent equations (8) are solved as before for the unknowns σ_0 and σ_1. Above T_c^b the whole continuum has reduced to a single zero frequency that is infinitely degenerate, while the local surface modes remain finite, up to a certain higher temperature which we call T_c^s.

In Fig. 1 we have plotted the results of the calculation with $\sigma\equiv 0$ above T_c^b. It must be remarked that one can just as well maintain $\sigma\equiv 0$ as a boundary condition on the third layer of a thin film of three layers, and calculate the variable σ_0 and σ_1 for all T down to $T=0$. At $T=0$, of course, $(\sigma_0,\sigma_1)\to S$. The film model $(\sigma\equiv 0)$ is related to previous calculations of the properties of thin films with surface and/or bulk anisotropy by Diep-The-Hung *et al.*[7] These authors also find for a film of four layers in the bcc and simple cubic structures, and where $s=\frac{1}{2}$, a transition temperature $T_c^s>T_c^b$ whenever η is sufficiently large. The consideration of the film $(\sigma\equiv 0)$ enables us to study the dependence of T_c^s upon η.

In this way we define a critical anisotropy η_c, such that $T_c^s(\eta_c)=T_c^b$. For $\eta\geq\eta_c$ the surface magnetization becomes larger than the bulk magnetization $\sigma(T)$ for $T<T_c^b$, and it does not vanish when σ does at T_c^b. The temperature T_c^s as a function of η is plotted in Fig. 2. The case $\sigma\equiv 0$ we call "case b", while the calculations for $T<T_c^b$ for the semi-infinite system we denote "case a". It turns out to be instructive to consider a film with *three* varied layers, and correspondingly $(\sigma_0,\sigma_1,\sigma_2)\neq 0$, while $\sigma_3\equiv\sigma\equiv 0$ for all T. The self-consistent equations for σ_l $(l=0,1,2)$ can be written and solved just as before. The respective results are also plotted in Figs. 1 and 2. We find that T_c^s for $\eta=1.8$ varies by $\sim 1.5\%$ when the number of perturbed planes n varies from two to three.

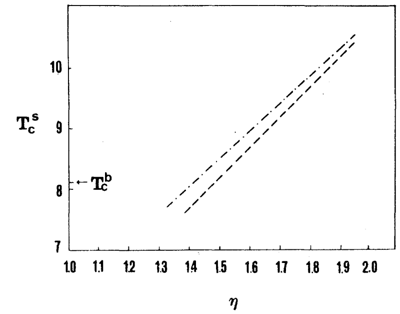

FIG. 2. Critical temperature of the two- and three-layer films as a function of η. Same temperature units as in Fig. 1.

In order to assess approximately the convergence of the results with increasing n, we plot in Fig. 3 the relative layer magnetization σ_l/σ_0 for the two cases considered, namely, $n=2$ and 3 at $T=8.2>T_c^b$ and $\eta=1.8>\eta_c$. If one assumes that the space variation of σ_l is exponential, from these results it can be estimated that σ_l vanishes for $l\gtrsim 5$, so that a five-layer calculation should give a better matching of cases *a* and *b*. This work is already in progress.

We have plotted in Fig. 4, for $T=8.0$, the dispersion relation of the film eigenvalues $\omega_l[\Lambda(\vec{k}_{||})]$, and those of the corresponding surface localized modes obtained for the semi-infinite system (case a), which in this case are the three optical surface magnon branches $\omega_\alpha[\Lambda(\vec{k}_{||})]$ $(\alpha=1,2,3)$. We observe that the latter are comprised, for all $\vec{k}_{||}$, between the eigenfrequencies of the three- and four-layer films. In the same figure we have indicated for each surface mode of case *a*, the eigenvalue of the transfer matrix $\xi[\Lambda(\vec{k}_{||})$ for $\Lambda=0.6$, which measures the wave-amplitude decrease of the corresponding mode between the successive (111) layers towards the interior of the crystal. It is clear that the higher the energy, the greater the degree of localization of the mode near the surface, and this is consistent with the results of case *b*.

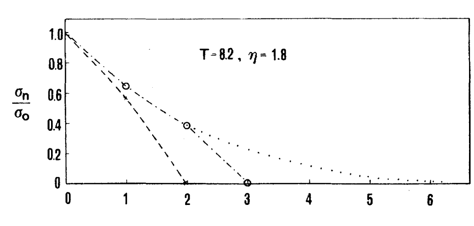

FIG. 3. Profile of the magnetization at $T=8.2$ and $\eta=1.8$ for the two films considered in Fig. 2: three layers (dashed line); four layers (dash-dotted line). Dots indicate assumed extrapolation. n is the layer index.

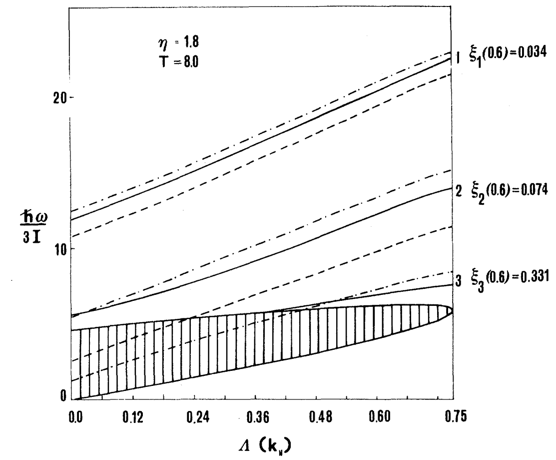

FIG. 4. Dispersion relations of localized magnon branches (case a, continuous curves) and film eigenfrequencies (case b) for three layers (dashed lines) and four layers (dash-dotted lines) for $T=8.0$. For case a, the numbers $1-3$ identify the optical magnon branches and for each of these, values of ξ_i $(i=1,2,3)$ for $\Lambda=0.6$ are also indicated. The shaded region is the bulk continuum.

III. CONCLUSIONS

The main conclusions from the preceding sections can be stated as follows:

(1) In the presence of surface exchange anisotropy the first layers of an insulating ferromagnet with isotropic exchange interactions in the bulk can stay ferromagnetic, that is, they show a nonzero spontaneous magnetization at temperatures greater than the bulk transition temperature T_c^b, if the surface anisotropy parameter $\eta > \eta_c > 1$, where the critical parameter η_c is defined by the condition

$$T_c^s(\eta_c) = T_c^b \, .$$

(2) From the magnetization curves shown in Fig. 1 we conclude that one should expect a discontinuity of the derivative of the magnetization curves of the different layers at the bulk transition temperature T_c^b, when $\eta > \eta_c$.

(3) For $\eta \le \eta_c$, the critical behavior of the surface is driven by the bulk. It is hoped that the techniques now available[8] will allow the study of the surface magnetization in samples where $\eta > \eta_c$.

On the other hand, theoretical results similar to ours have been obtained for an Ising semi-infinite ferromagnet.[4] Also in this case, when $T \to T_c^b$ from below, the magnetization of the surface layer tends to a finite limit when the surface coupling constant is larger than that of the bulk, which, on the basis of the present work, is to be expected, since the Ising Hamiltonian can be considered as the limit of an anisotropic exchange Heisenberg Hamiltonian, for infinite uniaxial anisotropy. From what has been said above, a similar behavior is found even with a finite uniaxial anisotropy at the surface.

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