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Surface-induced phenomena in uncompensated collinear antiferromagnets

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Abstract

The net spontaneous magnetization of antiferromagnets with modified surfaces was computed using mean-field theory. For ordinary phase transitions the net magnetization of uncompensated AFM is smaller than the surface magnetization and the Néel vector, whereas for extraordinary phase transitions the net magnetization is larger than the surface magnetization and the Néel vector at finite temperature. Moreover, the temperature dependence of these three observable internal parameters changes drastically with the surface properties, i.e. the surface exchange coupling J_S. Based on these findings, contour plots showing different regions of magnetization and Néel vector behavior as functions of temperature and surface exchange strength are proposed.

Keywords: antiferromagnet, surface, order parameter

(Some figures may appear in colour only in the online journal)

1. Introduction

The broken symmetry at surfaces or interfaces in magnetic materials generates phenomena that do not exist in 3D solids. A prominent example is the occurrence of a non-zero net spontaneous magnetization in antiferromagnets (AFM) with uncompensated surfaces, such as the (1 1 1) surface of archetypal Mott insulators like CoO and NiO. The Mott physics can have drastic effects on the surface state and lead to a modified pairwise exchange between surface spins, e.g. due to changes in the crystal fields [1], spin–orbit coupling [2], or Coulomb repulsion [3].

The effects of the surface state on magnetic systems have been investigated extensively over the past decades [4–13]. When spins on the surface have the same pairwise exchange interaction as the core of the film, the system undergoes an *ordinary* phase transition [14, 15] and the magnetization is weaker on the surface [4, 8, 10], but when the surface exchange is enhanced the system exhibits an *extraordinary* phase transition [14, 15], where the surface orders at a temperature higher than the ordering temperature of the bulk [17] and the magnetization is stronger at the surface [4, 8, 10]. To be precise, in semi-infinite extraordinary systems two transitions are predicted [17], where the surface undergoes a phase transition at $T_N^{surf} > T_N^{bulk}$, separate from that of the rest of the system. The surface exerts an effective field on atomic planes close to the surface, causing them also to order above T_N^{bulk}. If the film is sufficiently thin, the transition at T_N^{bulk} cannot be observed because the intervening atomic planes are forced into the ordered state at a temperature $T_N^{bulk} < T < T_N^{surf}$, i.e. the ordering temperature of the intervening atomic planes also depends on the thickness [16]. With increasing thickness, the effective field of the ordered surface will only affect the atomic planes close to the surface, and a separate bulk phase transition will then be evidenced at T_N^{bulk}.

The modification of the pairwise exchange on the surface also affects spins beneath the surface, and a non-trivial magnetization profile is produced through the film thickness in ferromagnets (FM) at finite T [5, 8]. For ordinary phase transitions the magnetization increases away from the surface, whereas for extraordinary transitions it decreases. For FM with semi-infinite thickness, the effect of the surface on M is minimal as the core dominates the magnetic behavior. In AFM, however, the net magnetization $M(T)$ exists solely because of the uncompensated surface (provided the number of planes is odd), and therefore $M(T)$ of AFM will be dominated by the

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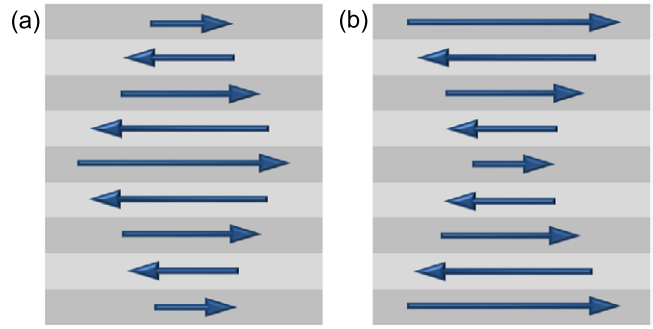

Figure 1. Illustration of the magnetization profile in AFM films with the Néel vector (staggered magnetization) perpendicular to the film plane. The arrows show the direction of the magnetic moment in each atomic plane, whereas we distinguish between two cases: (*a*) the magnetization on the surface is weaker due to lesser coordination, and (*b*) the magnetization on the surface is stronger due to enhanced pair-wise exchange coupling. The size of the arrows corresponds to the magnetization of each atomic plane, and their relative size is to scale for $T = 0.95T_N$. The net magnetization, i.e. the sum of all moments, is not equal to the surface moment due to the magnetization profile.

surface even for systems with semi-infinite thickness because of cancellations of the core [18, 19]. Note that cancellations of the magnetic moments are not one-to-one (see figure 1), so that the net M is not equal to the surface m, despite arising due to the surface.

The T-dependence of M in AFM, unlike FM, is therefore strongly dependent on the surface. Despite the importance of AFM in exchange-bias systems [20, 21] and spintronics [22, 23], the thermodynamic behavior of the net magnetization is largely unknown, mainly due to its small value (on the order of 10^{-6} emu cm^{-2}) [24, 25].

In this work we show, using mean-field theory (MFT), that the net $M(T)$ of AFM with uncompensated surfaces exhibits exotic behavior, which changes dramatically with the surface exchange coupling J_S. AFM with reduced J_S (compared to that of the bulk) exhibit negative values for the net magnetization and a compensation point (i.e. where the net magnetization changes sign at a finite temperature value), whereas enhanced J_S produces a non-monotonic $M(T)$ that has a peak, at which the net M exceeds the surface magnetization. In the former case, the net M is less than the surface m at all finite T, where in the latter, it is greater. The $J_S - T$ phase diagram of AFM with surfaces is therefore much more complex and richer than that of FM.

2. Thoretical model

Considering a collinear (type G) AFM where spins in each atomic plane are ferromagnetically coupled and spins in adjacent planes are antiferromagnetically coupled (see figure 1), the Heisenberg Hamiltonian, counting only nearest-neighbor interactions, is $\mathcal{H} = -\sum J_{ij}\mathbf{S}_i\mathbf{S}_j$, where J_{ij} is the exchange coupling between spins i and j, which is positive (FM) for inter-plane neighbors and negative (AFM) for intra-plane neighbors. The number of nearest neighbors in monoxide AFM, such as CoO and NiO, in the NaCl structure is 12. For $T \geqslant T_N$ (with T_N the Néel temperature) all 12 neighbors are equivalent, but below T_N the cubic crystal symmetry breaks into a rhombohedral symmetry and there are 6 neighbors with positive exchange and 6 with negative exchange [26]. We thus set the intra-plane exchange constant $J = 1$ and the inter-plane exchange constant as $J^* = -1$, to obtain a general description for collinear AFM. The intra-plane pair-wise exchange constant at the surface is defined as J_S, and will be scaled to J. The inter-plane exchange between the surface plane and the second-to-surface plane is taken to be unaffected by the modification of the surface.

We used mean-field theory (MFT) to calculate the magnetization because it is a convenient technique to model the thermodynamic behavior of insulating magnetic systems, and provides a clear picture of the physics involved in interacting many-body systems by reducing the problem to a one-body system. The self-consistent equation of state within MFT for the magnetization of each atomic plane m_d is the Brillouin function $m_d = B_S(S\ b_d\ m_d)$ with b_d the mean field due to interactions within the same atomic plane and interactions with spins in adjacent atomic planes. The mean field is $b_d = zJm_d + z^*J^*(m_{d-1}+m_{d+1})$, with $z = 6$ the coordination number inside the (1 1 1) plane and $z^* = 3$ the coordination number in the adjacent planes. In order to minimize the total free energy of the system, the coupled set of non-linear equations needs to be solved iteratively by changing the value m_d for each atomic plane and checking which set of values corresponds to the free energy minimum. For a film with D atomic planes, we need to solve D equations simultaneously to obtain the ground state by minimizing $\sum_d^D [m_d - B_S(S\ b_d\ m_d)]^2 = 0$ (for a more detailed description of the iteration see [18]).

In this work we calculated the thermodynamic properties of uncompensated AFM with a thickness of $D = 7$ and $D = 11$ (corresponding to 1.6 nm and 2.5 nm in transition-metal oxides) with $S = 1/2$ in order to see how the thermodynamic behavior of the AFM order changes with the surface state.

The thermodynamic *global* observable internal parameters for an uncompensated AFM film are the atomic plane magnetization (m), the net magnetization (M), and the Néel vector or staggered magnetization (N). The plane magnetization of the dth plane, containing n spins, at any given temperature is the normalized sum of all spins in each atomic plane

$$m_d = \frac{1}{|S|n}\sum_i^n S_i\ , \tag{1}$$

which can take any value in the range $-1 \leqslant m \leqslant +1$ (positive/negative corresponding to the oscillation of the magnetization in the AFM). The net magnetization, at any given temperature, is the sum of the plane magnetizations

$$M = \sum_{d=1}^{D} m_d\ , \tag{2}$$

and the Néel vector (the staggered magnetization), at any given temperature, is the sum of the absolute plane magnetizations

$$N = \sum_{d=1}^{D} |m_d|\ . \tag{3}$$

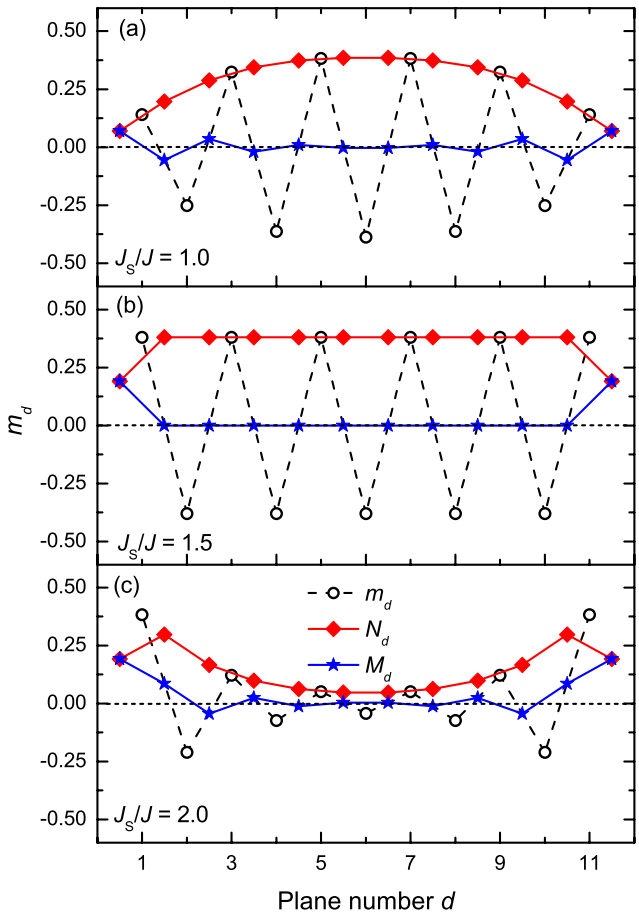

Figure 2. Magnetization profile for an AFM with $D = 11$ at $T/T_N = 0.95$, showing a comparison between the atomic plane magnetization m_d, the local net magnetization M_d, and the local Néel vector N_d (staggered magnetization) for (a) ordinary ($J_S/J = 1.0$), (b) special ($J_S/J = 1.5$), and (c) extraordinary ($J_S/J = 2.0$) systems. Lines are a guide to the eye.

The conventional order parameter for the antiferromagnet is N, but all three observables M, N, and m_S couple to external fields (magnetic field for M, neutrons for N, and surface effective fields, such as that exerted by a ferromagnetic overlayer, for the surface atomic plane magnetization m_S). Moreover, all three observables describe the same symmetry change at the ordering temperature and exhibit critical behavior near T_N. The critical behavior of net M is the same as that of m_S, whereas the critical behavior of N is the same as that of FM M, as shown e.g. by Monte Carlo simulations [19].

Further, we define a depth dependence for M and N at any given temperature, by computing the *local* observable internal parameter of each bi-plane, i.e. between each two adjacent atomic planes, which is $M_d = (m_d + m_{d+1})/2$, and $N_d = (-1)^d(m_{d+1} - m_d)/2$, respectively. At the surfaces, and considering the vacuum state as having $m_0 = 0$ and $m_{D+1} = 0$, the boundary conditions are $M_0 = (m_0 + m_1)/2 = m_1/2$ and $M_D = (m_{D+1}+m_D)/2 = m_D/2$, and similarly $N_0 = m_1/2$ and $N_D = (-1)^D m_D/2$. Note that we define the positive direction of the magnetic moments to be that of M at saturation, i.e. at $T = 0$.

For the surface state, we considered systems with ordinary phase transitions, i.e. with homogeneous exchange $J_S/J = 1$, and extraordinary phase transitions, i.e. with enhanced surface exchange $J_S/J = 2$, as well as what we call sub-ordinary where $J_S/J < 1$. For the discussion, we focus on an AFM with a thickness of $D = 7$ and $D = 11$ atomic planes, since in an earlier work [18] we showed that for $D \geqslant 7$ the magnetization profile, and thus the thermodynamic state, of AFM with surfaces does not change significantly. Hence, the results shown here are valid even for very thick AFM with surfaces.

3. Results and discussion

We begin by showing in figure 2 the magnetization profile as a function of the atomic plane number (as shown qualitatively in figure 1) for $D = 11$. The profile changes with temperature, because the near-surface magnetization changes differently than that in the core of the film [18]. The difference between m near the surface and the core, thus the dependence of m on d, is maximal close to the phase transition, and therefore we plot the local parameters close to the Néel temperature, at $T/T_N = 0.95$. Figure 2 shows a comparison between m_d (open circles), M_d (stars), and N_d (diamonds) for a system with $D = 11$, for AFM with (a) ordinary, (b) special, and (c) extraordinary phase transitions. The plane magnetization in both cases oscillates between positive and negative values. For $J_S/J = 1$ m_d is stronger in the core of the film and decays as we approach the surface. This behavior is reflected by the local Néel vector, which has a maximum in the center of the film and a minimum at the surface. The local net magnetization, on the other hand, has maxima at the AFM surfaces, and oscillates in sign but with rapidly decreasing magnitude as we move away from the surface. The first bi-plane has positive moment, whereas the second bi-plane has a negative moment, because the magnetization at the surface is much weaker than the magnetization in the second-to-surface plane. At a depth of more than 3 atomic planes, the net magnetization is nearly zero, because the values of the atomic plane magnetizations are very close in magnitude with opposite signs, and they cancel out. This shows that the magnetization profile is only important in the outer 3 atomic planes, and that the net $M(T)$ does not depend on the thickness of the AFM for $D \geqslant 7$, since all intervening magnetic moments cancel out. Note that this is valid for any temperature away from the phase transition, where the correlation length is on the order of a few lattice spacings [8]. Close to the critical temperature T_N, the effect of the surface (which depends on the perpendicular correlation length) is much stronger because the correlation length diverges as T approaches T_N, and hence becomes larger than the film thickness for some sufficiently small reduced temperature $(T - T_N)/T_N$. Therefore $M(T)$ in the limit of T approaching T_N of all films with finite thickness belongs in the 2D universality class. Below T_N, however, the correlation length decreases wth decreasing T, and when it becomes smaller than the film thickness the character of the $M(T)$ behavior changes from 2D to 3D, as shown e.g. by Monte Carlo simulations [19].

If the surface exchange coupling is enhanced, compared to that of the core of the film, the magnetization profile changes,

because the magnetization at the surface becomes stronger, but the system is still characterized as ordinary, as long as m on the surface is smaller than that in the core. When, however, J_S is enhanced enough to compensate for the missing neighbors of the surface plane, in this case the 3 missing neighbors can be compensated when $J_S/J = 1.5$, the system will exhibit a *special* transition, where all atomic planes are equivalent. As seen in figure 2(*b*), the atomic plane magnetization oscillates between positive and negative values, as it did for $J_S/J = 1$, but m_d is exactly the same in all atomic planes and the magnetization profile is flat. The local magnetization is zero for every d, and it has a finite value only at the surface, and the local Néel vector is constant for all d, except at the surface.

For $J_S/J = 2$ a different behavior is observed for m_d, M_d, and N_d (see figure 2(*c*)). The atomic plane magnetization now has *maxima* at the AFM surfaces and a *minimum* at the center of the film. The Néel vector follows the same trend, with the difference that the values of N_d at the surfaces are smaller than the neighboring $d-1$ (this is associated with the definition of $N_0 = (m_1 + 0)/2$). The local net magnetization oscillates between positive and negative values, as it did for $J_S/J = 1$, with pronounced maxima at the AFM surfaces. An important difference here is that while the net magnetic moment of the first bi-plane is positive, that of the second bi-plane is also positive, and only for the third bi-plane is M_d negative, in contrast to the ordinary case. This occurs because the magnetization at the surface is much stronger than that in the second-to-surface plane. As we move away from the surface, at a depth of more than 3 atomic planes the net magnetization vanishes and it is nearly zero, as it is in the $J_S/J = 1$ case.

Turning to the temperature dependence of the *global* parameters in AFM, in figure 3 we compare the thermodynamic behavior of the surface magnetization $m_S(T)$ (which is equal to m_1 and m_D), global $M(T)$, and global $N(T)$ for a film with $D = 7$. We studied 6 different scenarios of surface exchange, which include the ordinary ($J_S/J < 1.5$), special ($J_S/J = 1.5$), and extraordinary ($J_S/J > 1.5$) phase transitions.

We begin the discussion by considering the scenario that the surface exchange is reduced compared to that of the bulk. For $J_S/J < 0.5$, including the extreme case where $J_S/J = 0$, i.e. the spins on the surface are not coupled to each other, but only interact with the spins of the second-to-surface plane, the magnetization increases with negative values with decreasing temperature below T_N (with T_N the Néel temperature of the film) and exhibits a non-monotonic behavior with a compensation point at $T/T_N \approx 0.5$, below which it increases with decreasing T and saturates close to $T = 0$ at $M = 1$, the magnetic moment of a single atomic plane (note that we define the positive values of M to be that at $T = 0$). This occurs because the magnetic moment at the surface is much weaker than the rest of the AFM. At the compensation point, the magnetization at each surface is equal to that of the rest of the film and with decreasing temperature it becomes stronger.

For $J_S/J = 0.5$, $M(T)$ exhibits a nearly linear behavior with a flattened regime near T_N, and there is no compensation point. The criterion for the occurrence of the negative $M(T)$

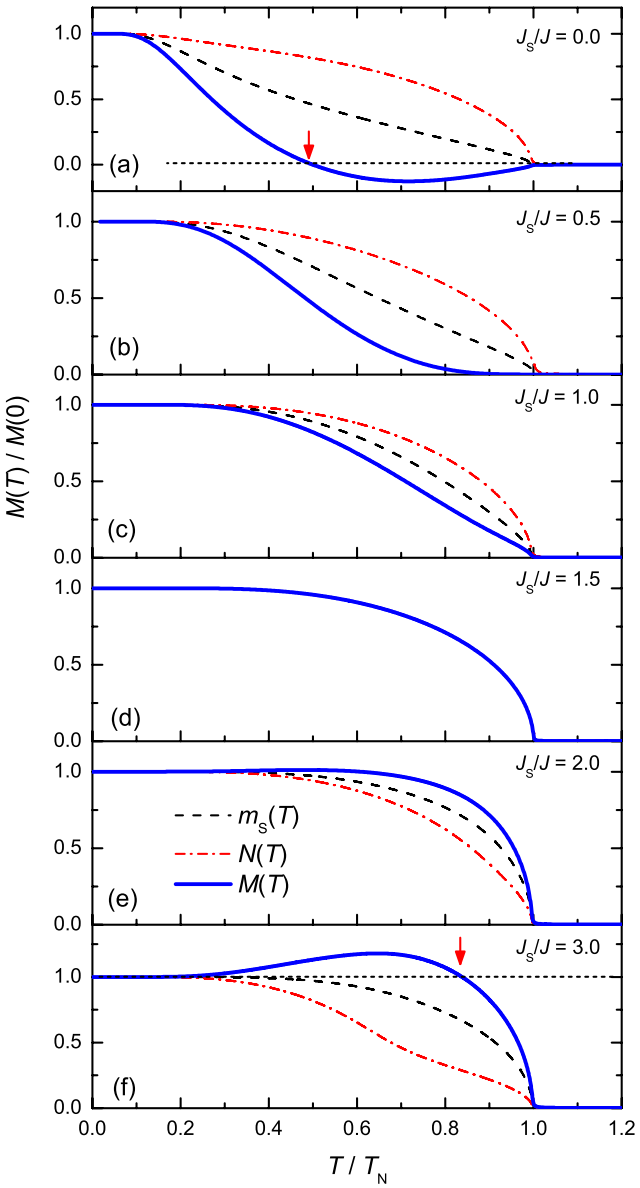

Figure 3. Temperature dependence of the surface magnetization (dashed line), global net magnetization (solid line), and magnitude of the global Néel vector (dash–dot line) for AFM with $D = 7$ and a surface exchange of (*a*) $J_S/J = 0$, (*b*) 0.5, (*c*) 1.0, (*d*) 1.5, (*e*) 2.0 and (*f*) 3.0.

and the compensation point for the surface exchange is $J_S/J \leqslant 1 - |z^*J^*|/|zJ|$ (with z the intra-plane coordination and z^* the inter-plane coordination), which in this case is 0.5. Note that reduced J_S/J could be an intrinsic electronic property of the surface, but would also emerge due to incomplete occupation of surface sites, causing little to no direct coupling between spins on the surface.

When $J_S/J = 1.0$, corresponding to an ordinary phase transition, the global net magnetization $M(T)$ increases monotonically with decreasing T with nearly linear T-dependence, distinctly non-Brillouin-like, down to $T/T_N = 0.25$ where it saturates. This unique $M(T)$ is due to the fact that the magnetization of the atomic planes close to the surface have a different temperature dependence than those in the core of the film, and the magnetization profile, and in turn

the cancellation of magnetic moments, changes with T (see figure 1(a) for a graphical explanation, and [18] for a more detailed description).

Note that for all $J_S/J < 1.5$ at intermediate T ($0.25 < T/T_N < 1$), the magnitude of the global Néel vector $N(T)$ (staggered magnetization) is larger than both $m_S(T)$ and $M(T)$, and the surface magnetization is larger than the net magnetization (see figures 3(a)–(c)). At the multicritical point ($J_S/J = 1.5$), all three observable internal parameters have the same T-dependence and exhibit a Brillouin-like $M(T)$ curve (figure 3(d)).

When the surface exchange is enhanced compared to that of the bulk ($J_S/J > 1.5$), the comparison of the observables is reversed, with $M(T)$ larger than $m_S(T)$ and $N(T)$. With decreasing T, M increases and reaches a maximum at around $T/T_N = 0.75$, exceeding the saturation value of 1. Upon further decreasing the temperature, M decreases and reaches $M = 1$ at $T = 0$. This non-monotonic behavior of $M(T)$ was also seen in Monte Carlo simulations of Ising AFM [19], and is due to the fact that the surface has a stronger magnetization than the core of the AFM, and therefore M becomes larger than m of any single atomic plane (see figure 1(b) for graphical explanation).

On increasing J_S/J to 3, the behavior of the $M(T)$ is amplified, and M exhibits a strong peak, and the disparity between $M(T)$, $N(T)$, and $m_S(T)$ becomes larger. By comparing figures 3(e) and (f) we see that with increasing J_S/J the maximum value of $M(T)$ increases; in the limit of large J_S/J it will equal 2, resulting from the addition of the magnetic moments of the two surface planes. The maximum $M(T)$ lies between T_N^{surf} and T_N^{bulk}, i.e. where the surface has long-range order but the center of the film is still paramagnetic, and it shifts to higher T with increasing J_S/J.

The reason that M is not equal to m_S for all values of J_S (except for $J_S/J = 1.5$), even though M originates from the uncompensated surface, is that the cancellations between the individual atomic planes are not one-to-one. For ordinary transitions the second-to-surface planes have m_{SS} such that $|m_S| - |m_{SS}| < 0$, whereas for extraordinary phase transitions, the surface moments are stronger than the rest of the system, resulting in $|m_S| - |m_{SS}| > 0$, causing $M < m_S$ for $J_S/J < 1.5$ and $M > m_S$ for $J_S/J > 1.5$ at finite T. At the multicritical point, when $J_S/J = 1.5$, all atomic planes have exactly the same T-dependence $|m_1| = |m_2| = \ldots = |m_D|$ and consequently $M(T) = m_S(T) = |m(T)|$ for all T.

The net magnetization is a *topological* property, i.e. it does not depend on the thickness of the AFM for $D \geqslant 7$. The form of $M(T)$ of an AFM is unique for all J_S, and quite unlike the Brillouin function of FM with similar thickness. Figure 4 shows a $J_S - T$ diagram for the thermodynamic observable internal parameters of AFM with uncompensated surfaces and surface exchange, and compares the net magnetization M (figure 4(a)) with the magnitude of the Néel vector (figure 4(b)). Note that the magnitude of the Néel vector is arithmetically identical to the normalized net magnetization of an equivalent FM system, since all magnetic moments in an FM are parallel, i.e. $N = |m_1| + |m_2| + \ldots + |m_D| = m_1 + m_2 + \ldots + m_D = M_{FM}$.

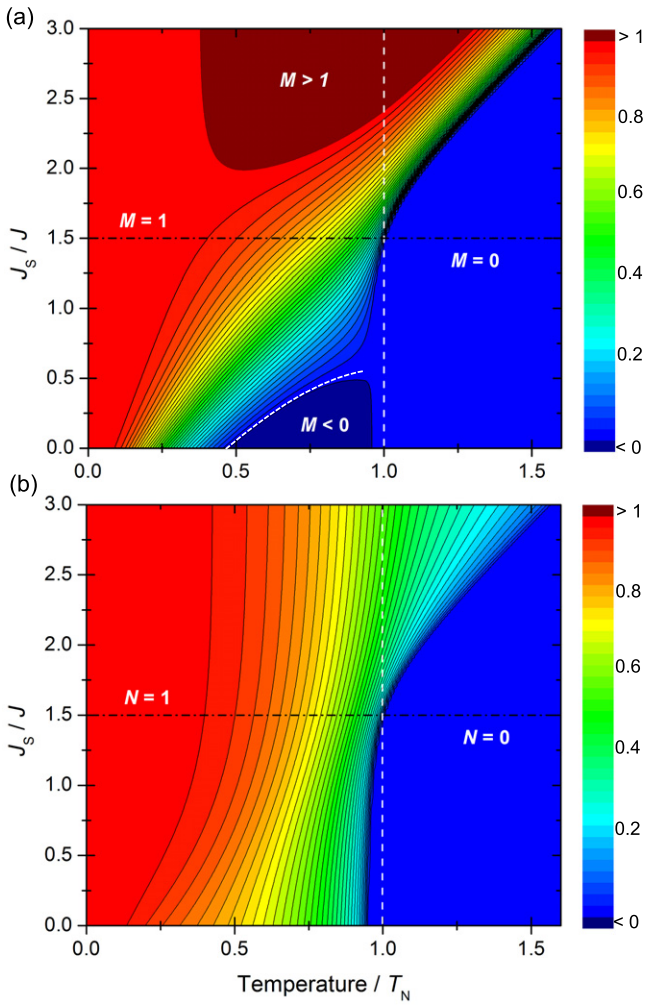

Figure 4. Contour plot of (a) the net magnetization M and (b) the Néel vector N as a function of temperature, normalized to the bulk ordering temperature (T_N^{bulk}), and normalized surface exchange J_S/J. The horizontal dot–dashed black lines mark the special transition and indicate the border between ordinary and extraordinary phase transitions. The vertical dashed white lines indicate the ordering temperature of the bulk. In panel (a) we observe three different regimes: (i) $M(T)$ has negative values (deep blue region) and exhibits compensation points (shown with a white short dashed line) for $J_S/J < 0.5$; (ii) $M(T)$ is monotonic for $0.5 \leqslant J_S/J \leqslant 2.0$; and (iii) $M(T)$ has a peak and exceeds the magnetization of any one atomic plane for $J_S/J > 2.0$ (deep red region).

As seen in figure 4(a), the AFM exhibits a very rich set of phenomena, which includes negative values and compensation points for small J_S/J, an over-saturation (when M is larger than the m of a single atomic plane) for high J_S/J, and a Brillouin-like character in between. The border between ordinary and extraordinary phase transitions is shown in the figure by horizontal dot–dashed lines, whereas the vertical dashed lines show the bulk ordering temperature (T_N). The compensation effect for sub-ordinary transitions occurs when the surface exchange is smaller than $J_S/J \leqslant 0.5$, and at temperatures around $T = 0.75T_N$ when the bulk dominates the surface. The over-compensation effect for extraordinary transitions occurs when $J_S/J \geqslant 2$ in a wide temperature range around T_N when the surface dominates the bulk. In the

phase space between the ordinary and the extraordinary phase transition, M has a Brillouin-like character, with an onset of M at T_N and monotonic increase towards $M = 1$ with decreasing temperature. Note that with increasing surface exchange, the ordering temperature of the film increases monotonically, as seen in the upper right area of figure 4(a).

The diagram for the magnitude of the Néel vector (equivalent to that of a FM film), is much simpler, and exhibits a Brillouin-like character at all J_S/J. For $J_S/J \geqslant 1.5$ the ordering temperature increases monotonically with increasing surface exchange, in the same way as in figure 4(a), but the magnitude of N for T larger than the bulk ordering temperature is very small (compared to the saturation value), because it corresponds to that of the surface, and will become increasingly smaller (compared to the saturation value) with increasing D. The significance of the surface is thus the most important difference between FM and AFM, since m_S becomes less important with increasing thickness for FM, whereas it dominates the $M(T)$ of the AFM regardless of the thickness, even though M describes the order of the entire AFM.

The above findings can be tested experimentally by comparing magnetometry $[M(T)]$ and neutron diffraction intensity $[I(T) = |m(T)|]$, where for AFM with an ordinary transition $M(T) \leqslant I(T)$, and with an extraordinary transition $M(T) \geqslant I(T)$. Also, the character of $M(T)$ should be different, either a nearly linear slope for $J_S/J = 1$, or a peak at finite T for $J_S/J \geqslant 2$.

Note that throughout our calculations we have neglected thermally-activated spin waves, which would reduce the plane magnetization, and in turn M at all non-zero T [27]. Even at $T = 0$, quantum fluctuations will affect the magnetization [28]. In a translation-invariant system the reduction of the magnetic moments by quantum fluctuations is homogeneous, but at the surface, the fluctuations are stronger [29], and this results in uneven cancellations of the magnetic moments, similarly to the finite-T case.

Despite these limitations of mean-field theory, i.e. the fact that fluctuations are not considered, the findings of this study for the temperature dependence of the net magnetization in AFM are in very good qualitative agreement with Monte Carlo simulations [19], in which fluctuations are intrinsic, and reveal the distinct temperature dependence of the net M, m_S, and N of antiferromagnets with surfaces. Moreover, the agreement between MFT and Monte Carlo leads to the conclusion that these findings are not dependent on the theoretical technique, but are intrinsic to AFM with surfaces.

4. Conclusions

In conclusion, the net magnetization M of collinear Heisenberg antiferromagnets with modified surfaces exhibits a rich set of phenomena that depend on the surface exchange coupling. For ordinary phase transitions with surface exchange coupling equal to that of the bulk interactions, M is nearly linear in T, whereas for extraordinary phase transitions with enhanced surface exchange coupling, M is non-monotonic and exhibits a peak. In addition, M is smaller (larger) than the magnetization of a single atomic plane for AFM with ordinary (extraordinary) phase transitions, respectively, despite arising from the uncompensated magnetization of the surface spins. A surface exchange—temperature contour plot shows significant differences between the net magnetization and the Néel vector of AFM, the latter being similar to M of FM. These findings offer a deeper understanding of the effect of surfaces on the magnetism of antiferromagnets.

Acknowledgments

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