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Anomalous magnetic thermodynamics in uncompensated collinear antiferromagnets

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Abstract – Monte Carlo simulations show that the net magnetization of collinear antiferromagnets (AFM) with uncompensated surfaces exhibits a unique thermodynamic behavior, with a temperature dependence unlike that of ferromagnets or of the Néel vector. The magnetization of AFM is not equal to that of its surface, even though it results from surface effects. This phenomenon appears in thin AFM films but is valid even in the limit of semi-infinite systems. The net magnetization of AFM therefore corresponds to a distinct topological thermodynamic state due to the free surface.

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Introduction. – Since their discovery [1,2], antiferromagnets have attracted immense attention due to their unique magnetization properties. Like ferromagnets (FM), antiferromagnets (AFM) exhibit a spontaneous long-range ordering of the atomic spins below the Néel temperature (T_N), but unlike FM, the spins order antiparallel, and cancel each other out, and the net magnetization of a perfect AFM is zero. If, however, there are defects, such as vacancies, grain boundaries, or surfaces, AFM can have a small uncompensated magnetization. As a specific manifestation of this effect, when an AFM film has an odd number of atomic planes along the Néel vector, *i.e.*, the axis of antiparallel alignment, the spins are not fully compensated and the AFM will exhibit a non-zero net magnetization because of the uncompensated surface [1]. In this paper we will show, using Monte Carlo simulations (MC), that the magnetization of AFM as a function of temperature shows a behavior which is strongly different from that of the Néel vector, that of a ferromagnet, or even that of the surface, despite the fact that the AFM magnetization only exists because of the uncompensated surface.

Numerical methods, such as Monte Carlo simulations, allow a unique and straight-forward approach to model magnetic systems. The thermodynamic behavior, in particular the critical region of phase transitions, of FM thin films has been studied extensively (see for example refs. [3–6]). Pioneering works of Binder, Hohenberg, and Landau showed how the modification of exchange

interactions at surfaces can strongly affect the magnetic ordering in systems with free surfaces [7–13]. When the pairwise exchange interaction between spins on the surface is the same as that between spins in the core of the film, the system undergoes an *ordinary phase transition* [14,15] at the critical temperature and the magnetization on the surface is weaker because spins at the surface have fewer neighbors. When, however, the exchange at the surface is stronger, possibly due to the reduction of the crystal field [16] or the enhanced of single-site spin-orbit coupling [17], the thermodynamic behavior changes drastically [18]. When the surface exchange is strong enough to overcompensate for the missing neighbors, then the system undergoes an *extraordinary phase transition* [14,15], where the surface orders at a higher temperature and has a magnetization which is stronger than in the core of the film.

The properties of the surface are therefore crucial for the thermodynamic behavior of low-dimensional systems. For example, EuTe(111) films exhibit strongly reduced magnetization near the surface of the film [19], whereas the magnetization of NiO(111) and NiO(100) films is stronger at the surfaces, and the surface order persists even above the T_N of bulk NiO [17,20]. Moreover, KMnF₃(110) [21] and MnO(001) [22,23] surfaces exhibit ordering at temperatures that are twice as high as the T_N of the bulk counterparts. Monte Carlo simulations used to calculate spin-spin correlation functions showed this is possible

when the pairwise exchange at the surface is much stronger than the bulk exchange [24].

The critical behavior of FM and AFM has been studied in detail, but the possibility of a net moment of an AFM and the effect of the free surfaces on the net magnetization M of AFM over a wide temperature range, *i.e.*, $0 \leq T \leq T_N$, have not been previously considered and is experimentally elusive because the direct measurement of the net spontaneous magnetization of uncompensated AFM is a challenging task due to its small value [25,26]. M has been studied only indirectly with exchange-biased systems, *i.e.*, when an AFM film is exchange-coupled to an adjacent FM film, both experimentally [27–29] and numerically [30,31].

While in FM systems the effect of the surface magnetization becomes less important as the thickness increases, the magnetization of AFM only exists because of the free surfaces, and the thermodynamic state of a free AFM system will thus be dominated by the state of the surface, even in the limit of infinite thickness. It is therefore of fundamental importance and interest to investigate the *intrinsic net spontaneous magnetization* of uncompensated AFM at finite temperature $M(T)$ under the circumstances of both ordinary and extraordinary phase transitions.

Model and computation. – Transition-metal oxides like CoO and NiO have a NaCl crystal structure, where each magnetic ion has 12 nearest neighbors in the fcc structure. Above the ordering 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 [32]. Thus, spins within each of the (111) planes are ferromagnetically coupled with the 6 nearest neighbors, and antiferromagnetically coupled to three nearest neighbors in each of the adjacent planes. The distortion of the crystal generates an on-site anisotropy, which is along the $\langle 11\bar{7} \rangle$ axis in CoO [33] and the $\langle 11\bar{2} \rangle$ axis in NiO [34].

In our simulations we used an Ising model to simulate oxide AFM, in order to capture the general thermodynamic behavior of AFM with surfaces. Even though an Ising model, which implies infinite uniaxial anisotropy, cannot reproduce the distortion-induced anisotropy in CoO and NiO, these oxides have shown Ising characteristics [35–39], and therefore our study is focused on a qualitative description of the long-range order, and not on a quantitative analysis of the exchange mechanisms or the anisotropy.

The classical Ising-type Hamiltonian is

$$\mathcal{H} = -J \sum_{\text{nn}} S_i S_j - J^* \sum_{\text{nn}} S_i S_k^*, \quad (1)$$

where S represents the local spin at each lattice site, J is the ferromagnetic exchange coupling in the (111) planes, and J^* is the exchange coupling between spins in adjacent planes, which is antiferromagnetic. Spins S_i and S_j are thus in the same plane, and spins S_i and S_k^* are

in adjacent planes. The exchange constants were set as $J = 1$ and $J^* = -1$, to obtain a general description of an AFM system, and the spins were allowed to take the values $S = \pm 1$. Spins on the surface interact with each other via J_S , where for ordinary transitions $J_S/J = 1$ and for extraordinary transitions $J_S/J > 1.5$ [9,12]. We assume that J^* between the surface and the second-to-surface atomic plane is not affected by the surface enhancement, since that occurs only due to the broken symmetry of the surface.

The simulations were performed using the Metropolis algorithm, *i.e.*, by updating a single spin each time until the system comes to quasi-equilibrium. 1000 Monte Carlo steps per site were run to reach equilibrium, and then an additional 10^8 steps were taken to obtain the averages of the observables, which are: the *magnetization of each atomic plane*

$$m_d = \frac{1}{N_d} \sum_{i=0}^{N_d} S_{d,i}, \quad (2)$$

where N_d is the number of spins S_d in each atomic plane, the *net magnetization* on the bond between each two atomic planes

$$M_d = \frac{1}{2}(m_{d+1} + m_d), \quad (3)$$

and the Néel vector, *i.e.*, the *staggered magnetization*, on the bond between each two atomic planes

$$|m|_d = (-1)^d \frac{1}{2}(m_{d+1} - m_d). \quad (4)$$

Using eqs. (3) and (4) we can compute the local value of the net magnetization and the Néel vector. To obtain the global values we add the d components, and end up with the global net magnetization $M = (m_1 + m_2 + \dots + m_D)/D$, and the global Néel vector $|m| = (|m_1| + |m_2| + \dots + |m_D|)/D$, which we will plot as a function of temperature.

The lateral size of the systems was $L \times L = 120 \times 120$ and periodic boundary conditions were used to eliminate finite-size effects. Occasional checks were performed with system sizes $L = 200$ to verify the validity of the results. The thickness of the films was varied between $D = 1$ and 15 atomic planes, which for films of transition-metal monoxides corresponds to $0.24 \leq D \leq 3.6$ nm. For the analysis of the critical behavior we performed finite-size scaling with thicknesses of 11, 21, 31, 41, and 51 atomic planes.

Results and discussion. – We begin by simulating AFM films with an ordinary phase transition ($J_S/J = 1$). Figure 1(a) shows the results of Monte Carlo simulations for AFM with increasing thickness, starting from a single atomic plane, *i.e.*, a 2D FM system. Figure 1 only shows results for odd values of D , since for even D numbers $M = 0$. The most drastic changes in the thermodynamic behavior occur during the first few monolayers, as seen by the shift of T_N , starting at $T_N \approx 3.7 J$ for $D = 1$

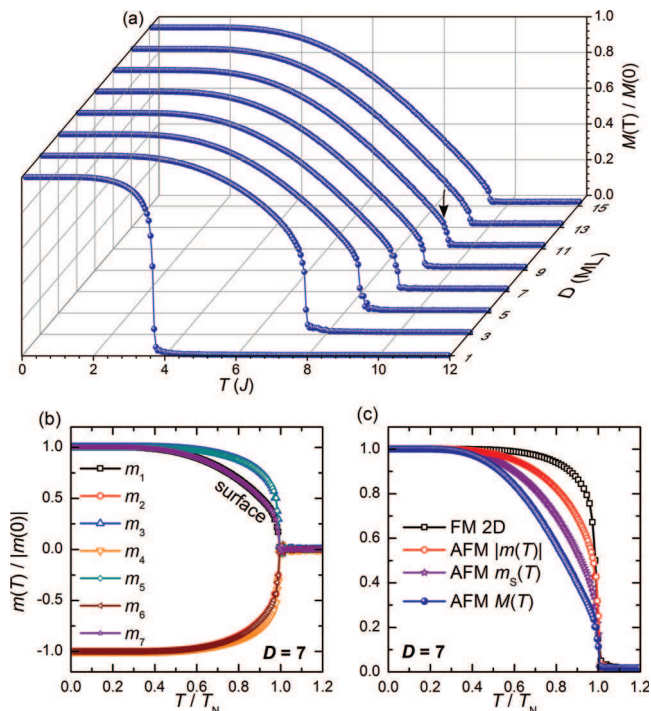


Fig. 1: (Colour on-line) Results for AFM with ordinary phase transitions ($J_S/J = 1$). (a) $M(T)$ (the temperature is given in units of J) for uncompensated AFM with increasing thickness D ; (b) T -dependence of the individual plane magnetization m for $D = 7$; and (c) comparison between M , the surface magnetization m_S , the Néel vector $|m|$, and the magnetization of a 2D ferromagnet as a function of the normalized temperature. The arrow in (a) indicates the kink in $M(T)$ which marks the crossover from 2D to 3D behavior. The kink shifts closer to T_N with increasing thickness.

and increasing monotonically as it approaches the bulk value $T_N(\infty) \approx 9.9J$, following $T_N(D) \propto 1 - D^{-\lambda}$, where $\lambda \sim 1$ [10].

The most striking observation is the gradual modification of the AFM $M(T)$ curve with increasing thickness, which does not resemble any thermodynamic behavior observed in FM systems. Starting from $D = 1$, the $M(T)$ curves exhibit the well-known Brillouin-like shape of the 2D magnet, but as the thickness is increased the curve changes drastically and for a wide temperature range ($0.5T_N \leq T \leq T_N$) it becomes nearly linear with T . Close to T_N the $M(T)$ exhibits a kink (see example arrow in fig. 1(a) for $D = 9$), where the dimensionality of the system changes from 3D to 2D (with increasing T) as the correlation length ξ diverges and becomes larger than the film thickness [40,41], and therefore as D increases, the kink moves closer to T_N .

To elucidate the T -dependence of M , we investigate the magnetization of each atomic plane (see fig. 1(b)) and compare M to that of the Néel vector $|m(T)|$ (which is proportional to M of a FM film of the same thickness), that of the surface atomic plane $m_S(T)$, and that of a 2D

FM for a system with the same D . Figure 1(c) shows the differences between these order parameters for $D = 7$ and one clearly sees that $M(T)$ is lower than $|m(T)|$ and m_S at all $T \geq 0.25T_N$. The fact that $M \neq m_S$ is surprising, considering that M exists only *because* of the uncompensated surface magnetization. The reason for this deviation is the inhomogeneous magnetization profile through the thickness of the AFM and the fact that the magnetic moments near the surfaces have a different T -dependence. The outer planes order at T_N together with the core of the film, but because the surfaces have smaller coordination, their magnetic moment is less than that of the core at finite temperature [$m_1(T) \leq |m(T)|$] (see fig. 1(b)). The magnetization of the AFM changes direction every atomic plane, but the cancellations are unequal and M is therefore lower than m_S [42]. For example, at $T = 0.75T_N$ the magnetic moments for a system with $D = 5$ are: $m_1 = 0.81$, $m_2 = -0.92$, $m_3 = 0.94$, $m_4 = -0.92$, and $m_5 = 0.81$. Adding these values results to $M = 0.72$, which is lower than the surface moment of m_1 (or m_5). Even though $M \neq m_1$ (or m_5), the nearly linear T -dependence of M is associated with the linear T -dependence of the surface magnetization predicted by mean-field theory [18,43].

At low temperature ($T \leq 0.25T_N$) the magnetization saturates and becomes equal to 1, *i.e.*, the magnetization of the surface plane. When, however, quantum fluctuations emerge at low temperature they will act in a way as to reduce the magnetic moments [44,45], especially at surfaces or interfaces [46], and the spin configuration at $T = 0$ will resemble the finite-temperature scenario, producing $M(0) < m_S(0)$, as it does for finite T (this effect was not included in this study).

The unusual behavior of the AFM $M(T)$ remains even in the limit of $D \rightarrow \infty$, as long as there is a free surface, which means that this unique $M(T)$ behavior is not an exclusive property of thin films, but it is an intrinsic property of AFM with free surfaces.

The above discussion is based on the ordinary transition, for which J is the same throughout the system. We turn now to the extraordinary transition, for which J_S is enhanced compared to J . When $J_S/J = 1.5$, *i.e.*, enhanced just enough to compensate for the missing neighbors, then the system will exhibit a *special* transition [9,12], where for thicknesses of $D \geq 2$ the magnetization profile is flat, and in which case $M = m_S = |m|$ at all T . This scenario corresponds to a singularity. We now consider systems with $J_S/J = 2$.

As before, we calculated $M(T)$ of AFM films with increasing thickness, and show the results in fig. 2. The ordering temperature of a single atomic plane ($D = 1$) is $7.4J$, double what it was for $J_S/J = 1$. The evolution of T_N with increasing thickness exhibits a peak ($T_N = 1.08T_N(\infty)$) at $D = 2$ (not shown in fig. 2 since $M = 0$ for even numbers of atomic planes) after which it shifts to lower values ($T_N(\infty) \rightarrow \sim 9.9J$, similar to the infinite D limit of the ordinary transition in fig. 1), because the intermediate atomic planes reduce

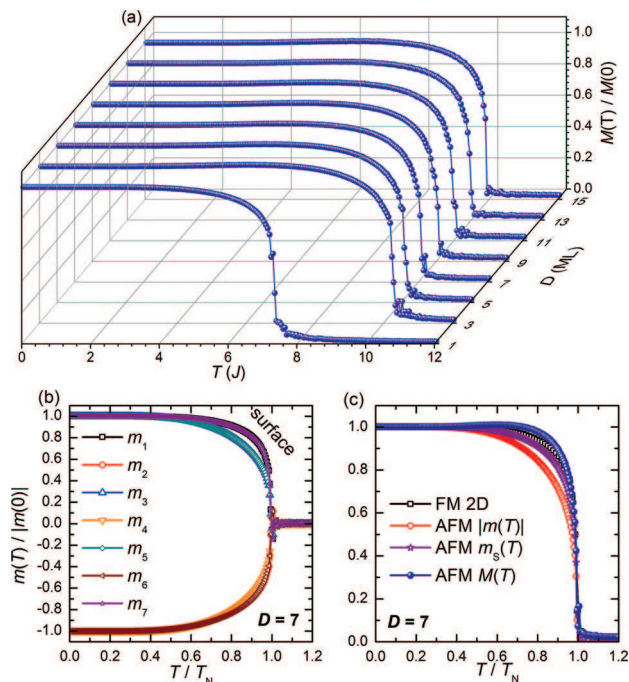


Fig. 2: (Colour on-line) Results for AFM with extraordinary phase transitions ($J_S/J = 2$). (a) M as a function of T (given in units of J) for uncompensated AFM with increasing thickness D ; (b) magnetization of each atomic plane m for $D = 7$; and (c) T -dependence of M , m_S , and $|m|$.

the ordering temperature due to their weaker exchange coupling [6,47,48].

Mean-field theory predicts that in a semi-infinite extraordinary system with surfaces, two transitions should be observed, *i.e.*, one for the surface and one for the bulk, with $T_N^{\text{surf}} > T_N^{\text{bulk}}$ [18]. Here only one phase transition is observed because of the finite thickness. As the two surfaces order at T_N , they force the rest of the AFM into the ordered state, and the surface and bulk phase transitions are mixed.

The T -dependence of the net moment $M(T)$ is strikingly different from that seen in fig. 1. As seen in fig. 2, the magnetization is saturated at $T = 0$, but with increasing temperature ($T \geq 0.5 T_N$) rather than the linearly decreasing (T) behavior seen in fig. 1, M increases and exceeds $M(0)$, *i.e.*, the saturation magnetization of a single atomic plane (by 1% for $J_S/J = 2$), after which it decreases rather abruptly and vanishes at T_N . This non-monotonic behavior of $M(T)$ occurs for all systems with $D \geq 3$, and does not change significantly with increasing thickness; there is only a slight shifting of T_N to lower values.

Figure 2(c) compares the different order parameters as a function of temperature. The net magnetization M is higher than the surface magnetization m_S , the Néel vector, and the magnetization of a 2D FM, all the opposite of what was found for $J_S/J = 1$. The magnetization on the two surfaces m_S is higher than $|m|$, *i.e.*, the magnetization profile is reversed (see fig. 2(b)), explaining the peak in $M(T)$ since the cancellation of the surface moments

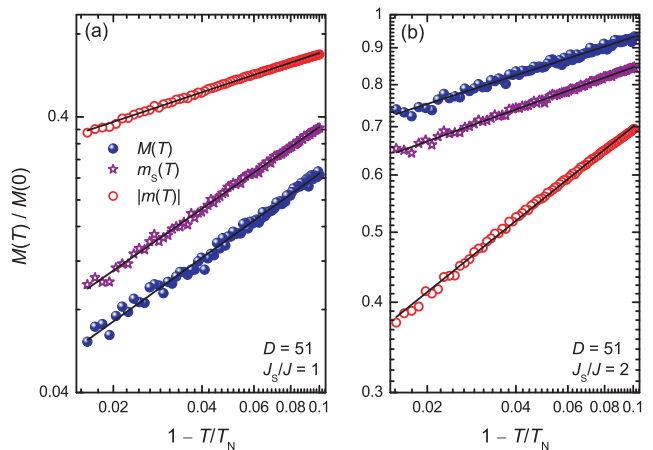


Fig. 3: (Colour on-line) Log-log plot of the normalized magnetization as a function of reduced temperature $t = 1 - T/T_N$ for AFM with $D = 21$, with (a) an ordinary phase transition $J_S/J = 1$ and (b) an extraordinary phase transition $J_S/J = 2$. The solid lines are fits with $M \propto t^\beta$ used to obtain the critical exponent β for m_S , $|m|$, and M .

(m_S) by the second-to-surface (m_{SS}) moments is weak ($2|m_S| - 2|m_{SS}| \geq 1$).

We note that the results regarding the form of $M(T)$, have also been obtained from mean-field theory. For $J_S/J = 1$ the net magnetization is lower than the surface magnetization and the Néel vector, and at intermediate temperatures $M(T)$ is nearly linear [42]. For $J_S/J = 2$, $M(T)$ is higher than m_S and $|m(T)|$, and it increases with decreasing temperature before it collapses at the phase transition (not shown here). Mean-field theory (MFT), however, does not distinguish the dimensionality of the system and cannot provide insight into the critical region, near T_N , so we have not included MFT results in this work.

Turning to the criticality of the AFM net magnetization, we have simulated systems with $L \times L = 120 \times 120$ and thicknesses $D = 11, 21, 31, 41$, and 51 to estimate the critical exponent β ($M \propto |T_N - T|^\beta$) for both ordinary and extraordinary phase transitions in the limit of infinite thickness. In fig. 3 we show the log-log plot of the magnetization as a function of reduced temperature for an AFM with $D = 51$.

For $D = 51$ and $J_S/J = 1$, the critical exponent of the net magnetization $\beta_M = 0.77(5)$ is within error bars of that of the surface magnetization $\beta_S = 0.75(5)$, which is in agreement to the known value of 0.78 for the free surface of a semi-infinite system [9]. The exponent of the Néel vector ($\beta_N = 0.35(5)$) is equal (within error bars) to the value for the 3D Ising magnet ($\beta_{3D} = 0.327$) [49,50]. The errors (given in parentheses) arise from the fit and from the uncertainty in the value of T_N , which was extracted from the data by comparing the Binder cumulant [51] for different system sizes.

For $J_S/J = 2$ the criticality changes dramatically and the exponent of M and m_S is $\beta_M \approx 0.13(5)$ and

$\beta_S = 0.14(5)$, respectively, within error bars of 0.125 of the 2D Ising magnet, and significantly different than those found for the ordinary transition. The critical exponent of the Néel vector is $0.33(5)$, *i.e.*, within error bars equal to that of the 3D system, since for a thickness of $D = 51$ the core of the film is not affected by the dimensionality of the surface.

By performing finite-size scaling we also estimated β for an AFM with infinite thickness. For ordinary phase transitions, $\beta = 0.79(2)$, whereas for extraordinary transitions it is $\beta = 0.12(1)$, which are within error bars of the values for the system with $D = 51$. Therefore, a thickness of 51 atomic planes is sufficiently large for simulating semi-infinite systems with surfaces, based on ref. [9], and considering that our simulations reproduce the results for the criticality of the surface magnetization m_1 and the staggered magnetization $|m|$, which are well known.

The fact that the net magnetization M and the surface magnetization m_S have the *same* critical behavior as each other for both the ordinary and the extraordinary transition, is an important distinction between AFM and FM systems. In a FM film, the value of m_S is different from the average M , as it is in the AFM, but in contrast to AFM, where the behavior is dominated by the surface, the magnetization of an FM depends on the entire film, as does the Néel vector of an AFM, which is the average absolute magnetization of the atomic planes. Hence the critical exponent for M of a FM system will be the same as that of the Néel vector of an AFM with the same thickness, but M of an AFM will exhibit the same criticality as the surface because the occurrence of M depends solely on the uncompensated surface, even though $M \neq m_S$ at finite T .

This unique thermodynamic state of uncompensated AFM can be investigated experimentally. The difference between the Néel vector and the net magnetization will be seen in the T -dependence of neutron diffraction intensity and magnetometry, respectively. In systems with ordinary transitions, such as CoO and FeO, the normalized neutron intensity $I(T)/I(0)$, which corresponds to $|m|$, should be stronger than M at finite temperatures, whereas in systems with extraordinary transitions, such as NiO and MnO, one would find the opposite ($M \geq |m|$) in addition to the anomalous behavior of $M(T)$ for $J_S/J = 2$, and the nearly linear $M(T)$ for $J_S/J = 1$.

Conclusions. – We conclude that the net magnetization $M(T)$ of collinear antiferromagnets with uncompensated surfaces exhibits a unique T -dependence, and that it is not equal to the surface magnetization, despite the fact that it exists due to the free surface. In ordinary phase transitions, $M(T)$ is lower than the surface magnetization and the Néel vector, with an unusual linear T -dependence, whereas in extraordinary phase transitions it exceeds both. In both cases, however, M follows the criticality of the surface m_S near T_N . This topological thermodynamic state persists with increasing thickness, and is valid even for a semi-infinite system with free surfaces.

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