Magnetism of thin Ising films with rough surfaces

F. D. A. Aarão Reis
Instituto de Física, Universidade Federal Fluminense, Avenida Litorânea s/n, Campus da Praia Vermelha, 24210-340 Niterói RJ, Brazil
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We study the ferromagnetic Ising model on thin films of random thicknesses using Monte Carlo simulations. The films have a simple cubic lattice structure, length and width N, one flat surface and discretized Gaussian distributions of thicknesses with mean L and rms deviation ΔL. We consider the cases of $\Delta L=\text{const}$ for any L (type I) and $\Delta L/L=\text{const}$ for any L (type II). A decrease of the critical temperature $T_c(L,\Delta L)$ for fixed L and increasing roughness (ΔL) is observed. The specific-heat peak of rough films of finite length is reduced when compared to the uniform films. The susceptibility peak is not reduced for small roughness ($\Delta L \leqslant 1$), and decreases for larger roughness. This type of disorder is shown to be irrelevant for the critical exponents, and two-dimensional finite-size scaling relations (in the lateral length N) do not have remarkable corrections when compared to the uniform films. In films of type I, the critical temperature shift $t_L=T_c(3D)-T_c(L,\Delta L)$ scales with L approximately as in the uniform films, and a small roughness becomes irrelevant for $L>10$, where three-dimensional scaling is attained. In films of type II, t_L decreases slowly with L, in disagreement with both two- and three-dimensional behavior for $L \leqslant 10$. We discuss the possible connections of our results and experiments in magnetic thin films. [S0163-1829(98)01922-5]

I. INTRODUCTION

In recent years there has been much interest in the magnetism of systems with a small number of atoms,[1–5] such as small magnetic clusters and thin films. The magnetic properties depend on several geometric features, which depend on the conditions during the growth. Thus the connection between experimental and theoretical results requires a detailed study of the dependence of physical quantities on the geometry of the systems. Some specific properties have already been considered in theoretical works, such as the size and lattice structure of small clusters,[6,7] the thickness of the films,[8] and the roughness of one-dimensional structures.[9]

Here we will study the influence of surface roughness in the magnetism of thin films, by considering films of Ising spins with random thicknesses. We will address some important questions from both the theoretical and experimental points of view. For instance, this model will provide information on the changes of the magnetic properties that can be attributed solely to the uncorrelated surface roughness. Furthermore, we will analyze the effect of different roughness patterns in the finite-size scaling relations involving the mean thicknesses of the films. Several experimental tests of those relations were already done,[3–5] and the connections to the conditions of growth were analyzed.

The layered Ising systems considered here can be exactly mapped on random spin systems on a square lattice, following the same ideas of the mapping of a two-layer spin-$\frac{1}{2}$ model on a square lattice spin-$\frac{3}{2}$ model.[10] Recently, the effect of dilution or bond disorder on the critical behavior of two-dimensional Ising systems has attracted much interest.[11–14] Although there are evidences that the critical exponents are the same as in the pure system, remarkable corrections to the dominant critical behavior were found. This work is also relevant on the general context of magnetism of disordered systems, particularly for the analysis of the corrections to the critical behavior.

In a recent work we have considered the one-dimensional version of these rough films, i.e., infinitely long strips of random widths.[9] It was shown that finite-size scaling is satisfied, taking the mean width L as the characteristic length of the strip, if the rms deviation ΔL is constant or proportional to L. It was also shown that the corrections to those relations increase with ΔL. However, due to the different dimensionality, most results cannot be extended to thin magnetic films.

We will consider films with dimensions $N\times N\times L$, where the length and width N are fixed throughout the structure, and L represents the mean of a discretized Gaussian distribution of thicknesses, with rms deviation ΔL. One of the surfaces is flat and the other is rough, as shown in Fig. 1. We will consider two possibilities for the rms deviation ΔL: $\Delta L=\text{const}$ for all L (type I) and $\Delta L/L=\text{const}$ for all L (type II). These possibilities represent, respectively, a fixed roughness when the mean thickness L increases and a roughness that increases with the mean thickness. Thermodynamic quantities will be calculated via Monte Carlo simulations.

We limited our study to relatively small thicknesses ($L \leqslant 10$) but, as will be shown below, it reveals interesting properties of rough films (e.g., their critical exponents), and some of these properties can be extrapolated to larger thicknesses, particularly for small roughness. Some cases of large

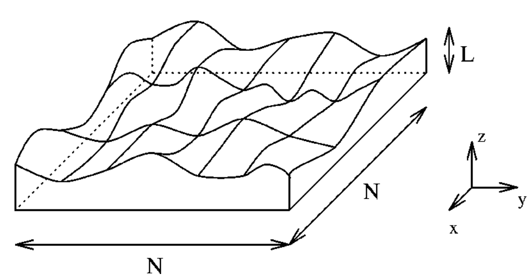

FIG. 1. Thin film with length and width N, one flat surface and one rough surface with mean thickness L.

roughness were considered when we studied the films of type II.

This work is organized as follows. In Sec. II we present the results of Monte Carlo simulations. In Sec. III we analyze the two-dimensional finite-size scaling relations (in the lateral length N) for the rough films. In Sec. IV we analyze the scaling of critical temperatures of films of types I and II. In Sec. V we discuss the possible relations between our results and experiments and summarize our conclusions.

II. NUMERICAL CALCULATIONS AND RESULTS

We considered the nearest-neighbor Ising model with (constant) ferromagnetic interactions

$$\mathcal{H}=-J\sum_{\langle i,j\rangle} S_iS_j-H\sum_i S_i\,, \tag{1}$$

where $\langle i,j\rangle$ denotes a pair of nearest neighbors, $S_i=\pm 1$ is the spin variable at site i, and H is an external magnetic field.

A simple cubic lattice structure is assumed for the films. They have equal lengths N in the x and y directions and variable height (z direction), satisfying a discretized Gaussian distribution with mean L and rms deviation ΔL. We studied films with $N=25$, 50, and 100; $L=2$, 3, 5, and 10. For films of type I we considered $\Delta L=0$ (flat films), $\frac{2}{3}$ and 1. For films of type II we considered $\Delta L/L=\frac{1}{2}$. The maximum acceptable thickness was $2L$ for all distributions. Periodic boundary conditions were considered along directions x and y, and free boundaries along direction z. The geometry of a film is illustrated in Fig. 1.

Monte Carlo simulations were performed using the Metropolis algorithm as usually applied to Ising systems.[15,16] All calculations were done in zero magnetic field. At each temperature, 5×10^4 initial Monte Carlo steps (MCS) per spin were discarded. The averages of thermodynamic quantities were taken from 10^4 spin configurations (5×10^4 near the critical temperatures). In order to reduce the correlations, N_s MCS were skipped between two successive configurations used in the averaging process, with N_s ranging from 10 to 50.

As the films are disordered, physical quantities must be avearaged over different realizations of each distribution of heights (i.e., different films with the same N, L, and ΔL). However, in films with small ΔL (typically $\Delta L\leqslant 1$), the large lengths N and periodic boundaries ensure that a large number of different microscopic environments (characteristic of the distribution) are present in a single realization. Then the differences of the quantities estimated in two different realizations are expected to be very small (this result was obtained in the one-dimensional version of the problem[9]). It was confirmed by calculations in some temperatures near T_c for all films with $\Delta L\leqslant 1$ using three different realizations. For instance, the fluctuations of the magnetic susceptibilities estimated in different realizations were always less than 3%, and this value is within the statistical error bars in the critical region. For other thermodynamic quantities the deviations were smaller. Thus the results presented here for any N, L, and $\Delta L\leqslant 1$ were obtained in a single realization of each distribution, but are sufficiently accurate to represent the averages in this ensemble of films.

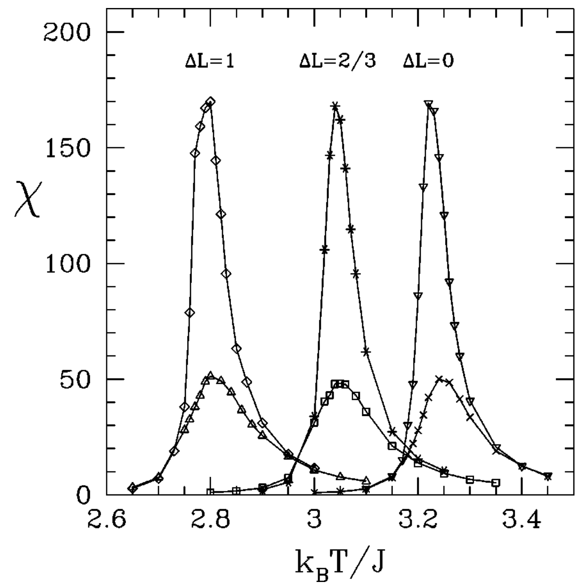

FIG. 2. Magnetic susceptibility per spin for various films with mean thickness $L=2$: $N=100$, $\Delta L=0$ (∇); $N=50$, $\Delta L=0$ ($\times$); $N=100$, $\Delta L=\frac{2}{3}$ (*); $N=50$, $\Delta L=\frac{2}{3}$ ($\square$); $N=100$, $\Delta L=1$ ($\diamond$); $N=50$, $\Delta L=1$ ($\triangle$).

For films with larger ΔL, the differences of the estimates obtained in different realizations are larger. The results presented below for films with $\Delta L>1$ are averages over N_R different realizations, with N_R ranging from 20 to 40, depending on the values of N, L, and ΔL. This number of realizations is sufficient to provide estimates with the same accuracy of the films with small roughness.

In Fig. 2 we show the magnetic susceptibility per spin χ for the films with $L=2$, $N=100$ and 50. It is clear that the critical temperature $T_c(L,\Delta L)$ decreases when the roughness (ΔL) increases. This property is also supported by the specific-heat data for the same films, shown in Fig. 3, and the magnetization per spin, shown in Fig. 4. Although $T_c(L,\Delta L)$ is defined when $N\to\infty$ (when χ and C_H diverge), it is very

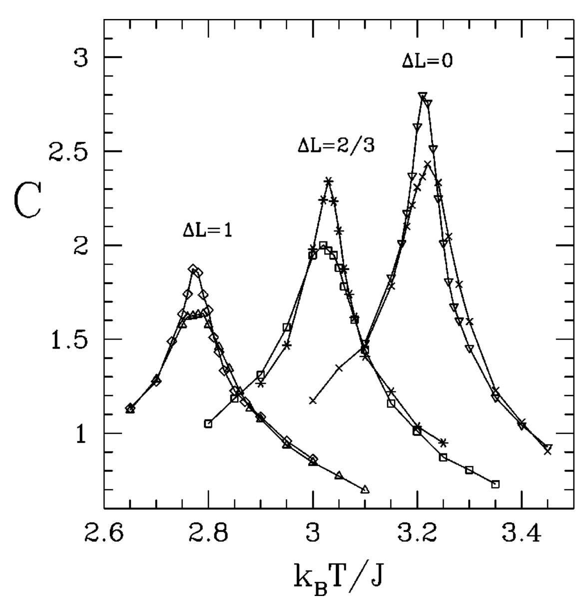

FIG. 3. Specific heat per spin for the same films in Fig. 2, with the same symbols.

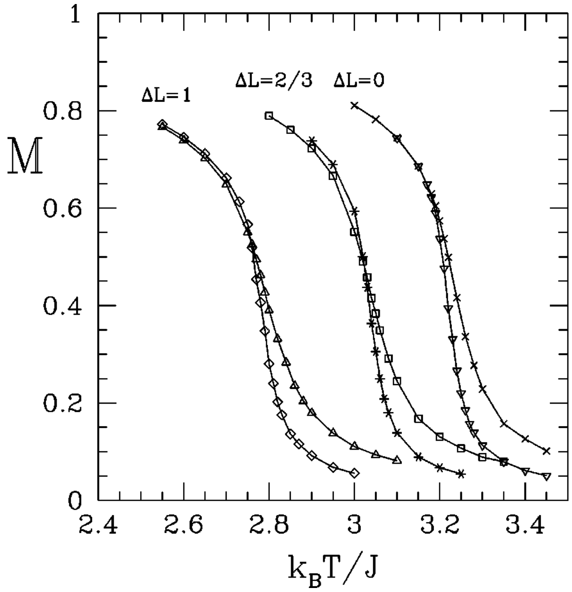

FIG. 4. Magnetization per spin for the same films in Fig. 2, with the same symbols.

near the temperature of the susceptibility peak of the largest films ($N=100$), $T_M(L,\Delta L)$, as will be shown in Sec. III.

The specific-heat peak C_{MAX} decreases when the roughness increases, which is consistent with $\int_0^\infty (C dT/T) = 2k_B$. However, it is interesting that it does not occur with the susceptibility peak χ_{MAX}. We also note that the height and the width of the susceptibility peak have negligible dependence on the roughness for all the other films with small roughness ($\Delta L \leqslant 1$).

For larger roughness, χ_{MAX} starts to decrease. In Fig. 5(a) we show the susceptibility for films with $N=100$, $L=5$ ($\Delta L=0$, 1, and $\frac{5}{2}$) and in Fig. 5(b) we show the susceptibility for films with $N=100$, $L=10$ ($\Delta L=0$ and 5). For films with $L=10$ and $\Delta L=5$, χ_{MAX} is nearly $\frac{2}{3}$ of the uniform film value ($L=10$, $\Delta L=0$).

For small roughness, χ_{MAX} seems to be related only to the lateral length N (the data for films with $N=50$ and $N=25$ support this result), and does not depend on ΔL. Remarkable decreases of χ_{MAX} are observed only in very rough films. Thus it seems that ΔL is the relevant variable to the decrease of χ_{MAX}, because the correlations along the film are more difficult when the roughness increases.

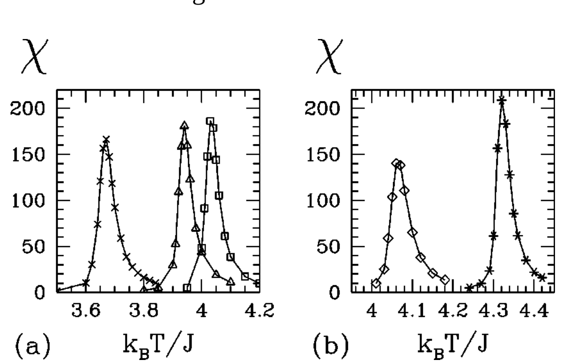

FIG. 5. Magnetic susceptibility per spin for films with: (a) mean thickness $L=5$, length $N=100$ and: $\Delta L=0$ (□); $\Delta L=1$ (△); $\Delta L=\frac{5}{2}$ (×); (b) mean thickness $L=10$, length $N=100$ and: $\Delta L=0$ (*); $\Delta L=5$ (◇).

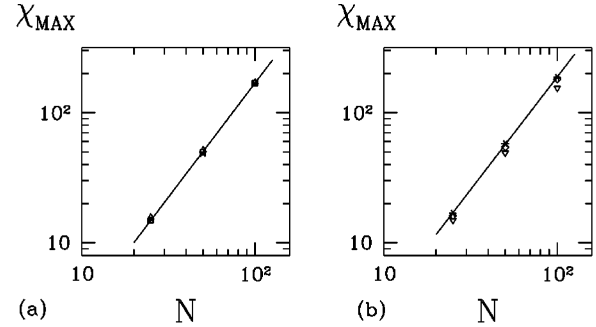

FIG. 6. Maximum susceptibility versus length N for films with: (a) $L=2$ (symbols are superimposed): $\Delta L=0$ (□); $\Delta L=\frac{2}{3}$ (×); $\Delta L=1$ (△); (b) $L=5$: $\Delta L=0$ (*); $\Delta L=1$ (◇); $\Delta L=5/2$ (▽). Straight lines are least-squares fits of the data for flat films.

III. TWO-DIMENSIONAL FINITE-SIZE SCALING ANALYSIS

The thermodynamic quantities of rough films with a fixed distribution of heights ($L,\Delta L$) and various lengths N (25, 50, and 100) scale with N as the corresponding quantities in flat films. It occurs even for large roughness, although the scaling amplitudes are different.

In Figs. 6(a) and 6(b) we show the susceptibility peaks χ_{MAX} versus N for films with $L=2$ and 5, respectively.

The expected scaling[17,18]

$$\chi_{MAX}(L,\Delta L,N) \sim N^{\gamma/\nu}, \tag{2}$$

with $\gamma/\nu = 1.75 \pm 0.03$, is obtained for flat and rough films with $L=2$ and $L=3$. For flat and rough films with $L=5$ [Fig. 6(b)], χ_{MAX} scales as Eq. (2) with $\gamma/\nu \approx 1.70$. For all films with $L=10$, $\gamma/\nu \approx 1.68$. For flat films, it certainly does not represent a change of universality class, but is an effect of the finite sizes N. These effects tend to increase for thicker films with the same lengths, which agrees with the results of a recent numerical study of flat Ising films.[8] The similar behaviors of χ_{MAX} in flat and rough films with the same thickness L indicate that the two-dimensional value $\gamma/\nu = 1.75$ is expected also for the latter, as $N \to \infty$, although the amplitudes of the scaling functions are different.

In Figs. 7(a) and 7(b) we show the specific-heat peaks C_{MAX} versus N for films with $L=2$ and 5, respectively. C_{MAX} scales in the rough films as in the uniform films. The expected scaling[17,18]

$$C_{MAX}(L,\Delta L,N) \sim \ln N \tag{3}$$

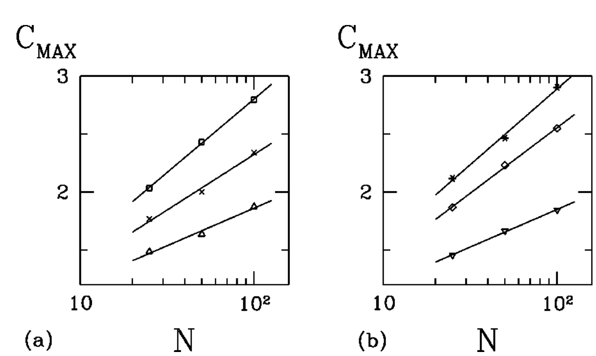

FIG. 7. Maximum specific heat versus length N for the same films in Fig. 6, with the same symbols.

TABLE I. Critical temperature $T_c(L,\Delta L)$, temperature of maximum susceptibility $T_M(L,\Delta L)$ of films with $N=100$, and relative decrease of T_c from the uniform film value, $\epsilon(L,\Delta L)$.

$L,\Delta L$	$T_c(L,\Delta L)$	$T_M(L,\Delta L)$	$\epsilon(L,\Delta L)$ (%)
2 , 0	3.196 ± 0.01	3.225 ± 0.01	
2 , $\frac{2}{3}$	3.022 ± 0.01	3.045 ± 0.01	5.4
2 , 1	2.743 ± 0.02	2.795 ± 0.01	14.2
3 , 0	3.628 ± 0.005	3.65 ± 0.01	
3 , $\frac{2}{3}$	3.528 ± 0.01	3.55 ± 0.01	2.8
3 , 1	3.378 ± 0.015	3.41 ± 0.01	6.9
3 , $\frac{3}{2}$	3.174 ± 0.015	3.23 ± 0.01	12.5
5 , 0	4.01 ± 0.01	4.03 ± 0.01	
5 , $\frac{2}{3}$	3.972 ± 0.005	3.99 ± 0.01	0.9
5 , 1	3.914 ± 0.01	3.94 ± 0.01	2.4
5 , $\frac{5}{2}$	3.634 ± 0.01	3.67 ± 0.01	9.4
10 , 0	4.306 ± 0.01	4.32 ± 0.01	
10 , 5	4.04 ± 0.01	4.065 ± 0.01	6.2

is obtained with good accuracy in the films with small thicknesses ($L=2$ and 3). Small deviations of Eq. (3) are observed in thicker films, but these deviations are also similar for flat and rough films. They can also be attributed to finite-size effects, and the scaling relation (3) must be satisfied in rough films as $N\to\infty$.

The scaling relations (2) and (3) for the disordered films are not evident. In two-dimensional systems with dilution or bond disorder, there is no controversy that Eq. (2) holds with $\gamma/\nu=1.75$, but a weaker divergence in Eq. (3) is expected.[12–14] We then conclude that the type of disorder analyzed here is irrelevant to the two-dimensional Ising critical behavior. Moreover, the corrections to the dominant critical behavior seem to have the same form of the uniform films, differing only in the scaling amplitudes.

In view of the previous results, we considered two-dimensional critical exponents to obtain the critical temperatures $T_c(L,\Delta L)$. We estimated the Binder cumulant[19]

$$U_N=1-\frac{1}{3}\frac{\langle M^4\rangle}{\langle M^2\rangle^2}, \tag{4}$$

where M is the magnetization of the system, and defined the pseudocritical temperature $T^*(N)$ (for fixed L and ΔL) as

$$U_N[T^*(N)]=U_{N/2}[T^*(N/2)]. \tag{5}$$

Then we assumed that

$$T^*(N)-T_c(L,\Delta L)=AN^{-\lambda}, \tag{6}$$

with A constant and $\lambda=1/\nu=1$ (the well-known result in two dimensions[20]). Using the estimates of $T^*(N)$ for $N=50$ and 100, we obtain $T_c(L,\Delta L)$ in Eq. (6).

The estimates of $T_c(L,\Delta L)$ and $T_M(L,\Delta L)$ (temperatures of χ_{MAX} of the largest films, with $N=100$) are shown in Table I. The differences of $T_c(L,\Delta L)$ and $T_M(L,\Delta L)$ are very small, with a maximum relative difference of 2% for $L=2$, $\Delta L=1$.

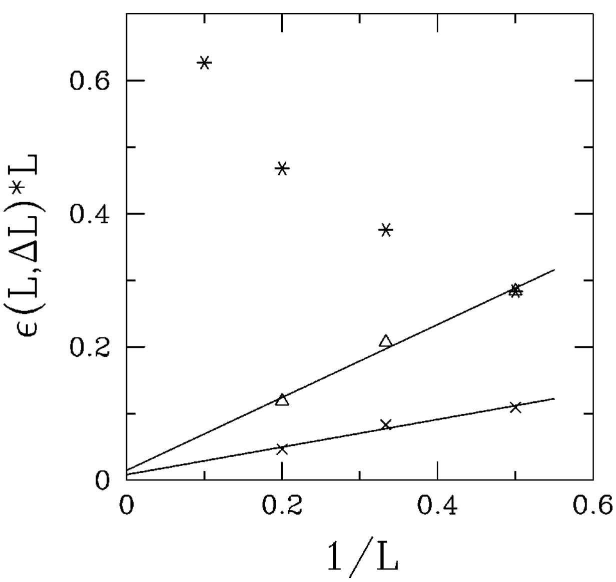

FIG. 8. $\epsilon(L,\Delta L)\times L$ (Eq. 7) versus $1/L$ for some classes of films: $\Delta L=\frac{2}{3}$ ($\times$); $\Delta L=1$ ($\triangle$); $\Delta L=L/2$ (*). Straight lines are least-squares fits of the data for films of type I.

IV. SCALING OF T_c OF ROUGH FILMS

The effect of roughness on films with fixed mean thickness L may be measured by the relative decrease of T_c

$$\epsilon(L,\Delta L)=\frac{T_c(L,0)-T_c(L,\Delta L)}{T_c(L,0)}, \tag{7}$$

which is also shown in Table I. We note that $\epsilon(L,\Delta L)$ increases fast with ΔL, for fixed L, which corresponds to the fast decrease of $T_c(L,\Delta L)$ with increasing roughness.

In Fig. 8 we show $\epsilon(L,\Delta L)\times L$ versus $1/L$. Results for films of type I ($\Delta L=\frac{2}{3}$ and $\Delta L=1$) and films of type II are clearly different. In films of type I, $\epsilon(L,\Delta L)$ decreases approximately with $1/L^2$, indicating that a fixed roughness pattern is irrelevant for quite small thicknesses ($L\approx15$ for $\Delta L\leqslant1$). In films of type II, $\epsilon(L,\Delta L)$ decreases very slowly with L (approximately as $1/\sqrt{L}$), then we expect that only for much thicker films ($L\gg10$) this roughness pattern will become irrelevant.

It is expected that the reduced critical temperatures t_L scale as

$$t_L=\frac{T_c(3D)-T_c(L,\Delta L)}{T_c(3D)}=AL^{-\lambda}, \tag{8}$$

with $T_c(3D)\approx4.51152$ (Ref. 21) and $\lambda=1/\nu$, where ν is the critical exponent for the three-dimensional Ising model [$\nu\approx0.6301$ (Ref. 21)].

For small L, however, that scaling is not obtained even in the flat films. A nearly two-dimensional behavior ($\lambda=1/\nu\approx1$) is obtained for $L<10$, and, as shown in a recent study,[8] the three-dimensional behavior is attained only for $L\gtrsim10$.

In Fig. 9 we show t_L versus L in four classes of films: flat, $\Delta L=\frac{2}{3}$, $\Delta L=1$, and $\Delta L=L/2$. We note that films of type I follow the same trend of the flat films, which is approximately a two-dimensional behavior in this range of thicknesses. A small roughness must become irrelevant for L

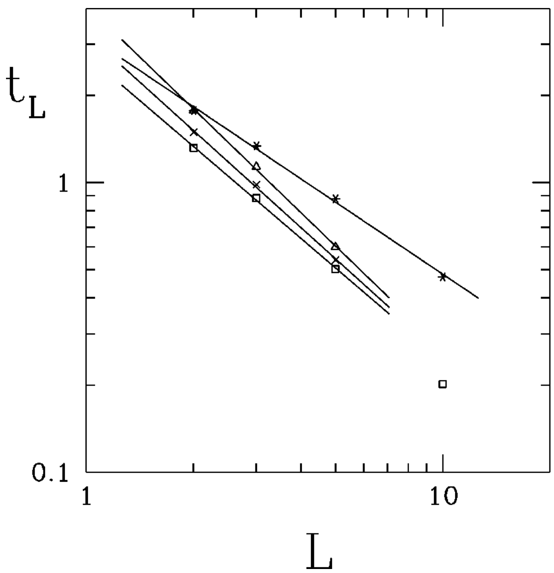

FIG. 9. Reduced critical temperature t_L (Eq. 8) versus mean thickness L for some classes of films: $\Delta L=0$ ($\square$); $\Delta L=\frac{2}{3}$ ($\times$); $\Delta L=1$ ($\triangle$); $\Delta L=L/2$ (*). Straight lines are least-squares fits of the data for all classes. For $\Delta L=0$, $\Delta L=\frac{2}{3}$ and $\Delta L=1$, the fits considered only $L\leqslant 5$, in order to show the similar behavior (nearly two dimensional) in those classes.

≈ 15, but that is exactly the region where three-dimensional behavior is attained. Thus we conclude that a small roughness will not disturb the three-dimensional scaling in the range of thicknesses where it is valid.

On the other hand, a very large roughness will be irrelevant only for larger L, even when it is fixed for all L. Consequently, it will disturb the three-dimensional behavior for $L\approx 10$. For instance, we expect that a fixed $\Delta L=5$ will be irrelevant only for $L\approx 30$, assuming a $1/L^2$ dependence of $\epsilon(L,\Delta L)$, as discussed above.

In films of type II, t_L decreases very slowly with L. The scaling in Fig. 9 is not consistent with two- or three-dimensional behavior, but with $\lambda\approx 0.8$. The previous analysis of $\epsilon(L,\Delta L)$ indicate that these deviations will disappear only for very large L.

The results above are completely different in the strips of random widths studied previously.[9] There, finite-size scaling relations were valid for small lengths ($L\leqslant 12$), but the effects of roughness in the corrections to scaling were remarkable, even for ΔL fixed and small.

V. SUMMARY AND CONCLUSIONS

We have shown that the introduction of uncorrelated roughness on thin Ising films reduces the critical temperature and the height of the specific-heat peak, but only for large roughness ($\Delta L>1$) the susceptibility peak has a considerable decrease. This type of disorder has no effect on the critical exponents, and the corrections to the dominant critical behavior seem to be the same of the flat films. In films of type I, with fixed roughness for all thicknesses L, the reduced critical temperatures t_L (Eq. 8) scale with L approximately as in the flat films, showing a crossover from two- to three-dimensional behavior for small L. A small roughness ($\Delta L\leqslant 1$) has negligible effects on T_c for $L\gtrsim 10$, where the three-dimensional behavior of t_L is observed. In films of type II, where the roughness increases with L, t_L decreases very slowly. Our results may be connected to previous experiments with thin magnetic films and may also help future investigations.

The variations in T_c and χ_{MAX} in different growth conditions are frequently analyzed in experiments with thin magnetic films. As an example, we consider recent experiments with Co films deposited on $W(110)$.[5] In films with mean thickness near 2 monolayers, it was observed that T_c and χ_{MAX} decreased after annealing, but the first layer was thermally stable. It was suggested that an increase in the roughness was one of the reasons for that behavior. According to our results, these properties cannot be explained by uncorrelated roughness alone. A large roughness would be necessary to produce a large decrease of χ_{MAX}, but for large ΔL and $L\approx 2$ the first layer would not be completely filled (for instance, if $\Delta L=2$, 11% of the first layer is not filled). In fact, the decrease of χ_{MAX} in the experiments suggests a decrease of the lateral sizes of the connected regions of the film. Then a model should incorporate other effects, such as formation of islands (also suggested in Ref. 5), in order to describe these experiments.

The dependence of t_L on L is also frequently analyzed in experiments.[2–5] For small roughness, which is present even in the better growth conditions, the three-dimensional behavior of t_L is not affected in the range of thicknesses where it applies ($L\gtrsim 10$). Our results also show that deviations from both two- and three-dimensional behavior for small thicknesses may be a signature of a roughness increasing with the mean thickness (e.g., films of type II).

Another interesting consequence of the large variations of T_c with roughness is that t_L must decrease smoothly with L if there is a smooth relation between L and ΔL. Films of types I and II have these properties (Fig. 9). But if the particular growth conditions lead to a chaotic variation of ΔL when L increases, we also expect a chaotic variation of t_L, which would be clearly displayed in a $t_L\times L$ plot.

Finally, we suggest that simple modifications of our model of rough films, incorporating magnetic inhomogeneities or other types of disorder (e.g., correlated roughness), may give interesting results and explain some features of real systems.

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