Corrections to the Kelvin equation for long-range boundary fields

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(Received 18 January 2005; published 18 May 2005)

The properties of a simple fluid, or Ising magnet, confined in an $L\times\infty$ geometry, are studied by means of numerical density-matrix renormalization-group techniques. Whereas the particle-particle interactions are short ranged, the wall-particle interactions can be long ranged. Using a few complementary criteria the wetting transition line has been found for boundary fields with different ranges (including a marginal case) and its scaling form has been analyzed. A main goal was to analyze the influence of wetting on the scaling of the Kelvin equation, focusing on the leading correction term. We have found that at complete wetting the larger the range of the boundary fields, the smaller the region of anomalous corrections of type $1/L^{4/3}$.

DOI: 10.1103/PhysRevE.71.056110 PACS number(s): 05.50.+q, 68.35.Rh, 68.08.Bc

I. INTRODUCTION

Wetting phenomena are very common in nature [1–3]; the most familiar situation is a liquid-vapor system in a contact with a solid wall. Usually one considers a semi-infinite geometry with a solid planar wall that preferentially adsorbs one of the phases of a system in thermodynamic equilibrium. Below the bulk critical temperature T_c, the adsorbed phase forms either isolated droplets (a dry regime) or a thick macroscopic layer (a wet regime). The first case, known as partial wetting, occurs for temperatures below the wetting temperature T_w, while the second case occurs for $T_w \leqslant T < T_c$ and it is referred to as complete wetting (see Fig. 1). Here, as a model system, the Ising model is considered in a two-dimensional ($d=2$) geometry and both phases, a liquid one and a vapor one, correspond to two phases with opposite magnetization. The fact that a wall can favor one of the phases corresponds, in Ising language, to introducing surface magnetic field h_1. The bulk magnetic field h refers to the chemical potential of the liquid-vapor system. In the $d=2$ semi-infinite Ising model the wetting temperature is known exactly [4] and decreases monotonically with the surface field h_1 (the solid line in Fig. 2).

For $d=2$ Ising strips of a finite width L (with opposing surface fields $h_2=-h_1$) a partial wetting is restricted to temperatures below the so-called interface delocalization (ID) temperature $T_d(L)$ [case (2) in Fig. 1]. For the short-range boundary fields, when L grows to infinity $T_d(L)$ scales to the wetting temperature T_w as $T_d(L)-T_w \approx L^{-1/\beta_s}$, where β_s is the exponent describing the divergence of the thickness of the wetting layer for a semi-infinite system [5].

In this paper we analyze the effect of wetting on the thermodynamics of the Ising model confined in an $L\times\infty$ geometry with identical boundaries. For $L\to\infty$, i.e., in the bulk, phase coexistence occurs for temperatures $T<T_c$ and for vanishing bulk magnetic field $h=0$. It is well known that the combined effect of identical boundary fields and confinement shifts phase coexistence to a finite value of the bulk magnetic field $h=h_0(L)\neq 0$ [case (1) in Fig. 1] [6,7]. For short-range boundary fields this line scales for large L, according to the Kelvin equation [8], as

$$h_0(L) = \frac{\sigma_0 \cos\theta}{m_b}\frac{1}{L}, \tag{1}$$

where σ_0, m_b, and θ are the surface tension, bulk spontaneous magnetization, and contact angle, respectively. This phenomenon is analogous to the capillary condensation for a fluid confined between parallel surfaces, where the gas-liquid transition occurs at a lower pressure than in the bulk. Moreover, the finite size scaling predicts that the capillary critical point $[h_c(L), T_c(L)]$ (see Fig. 1) scales as

$$T_c(L) - T_c \sim L^{-1/\nu} \quad \text{and} \quad h_c(L) \sim L^{-(d-\beta/\nu)}, \tag{2}$$

where d is the dimensionality and ν and β are the correlation length and magnetization exponents [9].

Capillary condensation with long-range boundary fields in an $L\times\infty$ geometry was analyzed by Albano *et al.* [10]. Using Monte Carlo simulations they confirmed also here the dominant role of the $1/L$ contribution.

The next order correction term to the Kelvin equation was studied by Albano *et al.* [6] and by Parry and Evans [7]. Both studies, using scaling and thermodynamics arguments, concluded that at partial wetting, the leading correction to scaling term is of type $1/L^2$. In the case of complete wetting the correction is expected to be nonanalytic due to a singularity of the surface free energy. For the $d=2$ Ising model [6,7] the predicted correction term is proportional to $1/L^{5/3}$. The Monte Carlo data [10] for long-range boundary fields were not accurate enough to convincingly test the type of corrections to scaling.

In a subsequent investigation, using density-matrix renormalization-group (DMRG) techniques, it was found [11] that for a wide regime of short-range boundary fields and temperatures, higher order corrections are of type $1/L^{4/3}$. This apparent disagreement was due to the fact that even for the large sizes considered ($L\sim 150$) the wetting layer had a limited thickness, so that the singular part of the surface free energy which determines the correction to scaling behavior was dominated by the contacts with the walls. In this case a simple random walk argument indeed predicts the leading correction term of the type $1/L^{4/3}$ for a thin wetting layer [11].

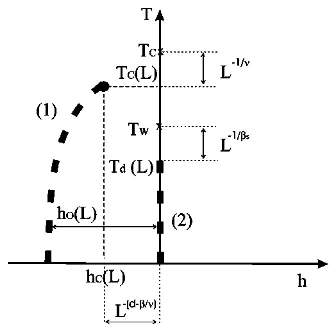

FIG. 1. Phase diagram of the d-dimensional Ising model for a bulk system in the (h,T) plane (h in units of J, T in units of J/k_B). The dashed lines are the phase diagrams of confined systems with identical (1) and opposing (2) surface fields.

In order to complete the picture, it is natural to extend the considerations to the long-range case, especially since that it is known [2,3] that long-range forces modify the wetting behavior significantly. Therefore, in the present paper, we study an Ising model with long-range boundary fields. It is reasonable to expect that for such a case the wet layer should be thicker with respect to the short-range case, which is likely to shrink the region with anomalous corrections (of the type $1/L^{4/3}$) to the Kelvin equation.

II. MODEL

In spite of the name, the DMRG has only some analogies with the traditional renormalization group being essentially a numerical, iterative basis, truncation method. It was proposed by White in 1992 as a new tool for the diagonalization of quantum chain spin Hamiltonians [12]. Later, it was adopted by Nishino for $d=2$ classical systems at nonzero temperatures [13]. The DMRG method allows to study much larger systems (up to $L=690$ in this paper) than it is possible with standard exact diagonalization method (up to $L=20-30$ for Ising strips) and provides data with remarkable accuracy. In the application of the DMRG method for classical $d=2$ spin systems, symmetric transfer matrices are used. Comparisons with exact results for the case of vanishing bulk magnetic field and boundary fields acting only on spins in the surface layers, show that this technique gives very accurate results in a wide range of temperatures [14]. Recently the method has been also applied to an Ising film subject to long-range boundary fields to study the solvation force behavior [15].

Our results refer to the $d=2$ strip defined on the square lattice of the size $M\times L$, $M\to\infty$. The lattice consists of L parallel columns at spacing $a=1$, so that the width of the strip is $La=L$. We label successive columns by the index l. At each site, labeled (k,l), there is an Ising spin variable taking the value $\sigma_{kl}=\pm 1$. We assume nearest-neighbor interactions of strength J and Hamiltonian of the form

$$\mathcal{H}=-J\left\{\sum_{\langle kl,k'l'\rangle}\sigma_{kl}\sigma_{k'l'}+h\sum_{kl}\sigma_{kl}+\sum_{l=1}^{L}H_l\sum_{k}\sigma_{kl}\right\}, \quad (3)$$

where h and H_l are in units of the coupling constant J. The first term in Eq. (3) is a sum over all nearest-neighbor pairs of sites, while in the second term h is the bulk magnetic field acting on all sites. The value $H_l=H_l^s+H_{L+1-l}^s$ is the total boundary magnetic field experienced by a spin in column l. The single-boundary field H_l^s is assumed to have a form $H_l^s=h_1/l^p$ with $p>0$, and the reduced amplitude of the boundary field $h_1\geqslant 0$.

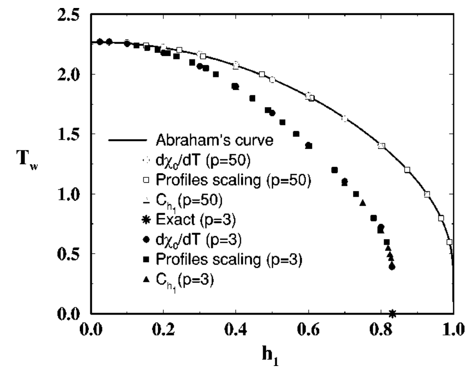

FIG. 2. Phase diagram of the interface delocalization transition for $p=50$ (short-range boundary fields) and $p=3$ (long-range boundary fields) in the $L\to\infty$ limit (h_1 in units of J, T_w in units of J/k_B). For $p=3$ the limiting $T\to 0$ value is known exactly $(\Sigma_{l=1}^{\infty} l^{-3})^{-1}$.

In order to study how the long-range interactions between walls and spins modify the corrections to the Kelvin equation we analyze first their influence on the wetting temperature.

III. THE WETTING TRANSITION

In order to determine a location of the ID transition we have applied three criteria. For all cases the finite system calculations were done first (at fixed L) and then extrapolated to infinity to recover the wetting transition line. To compare our results with the short-range boundary fields case, where the exact results exist [4], we put $p=50$. As we have checked the H_l value for $l=2,\ldots,L-1$ is here so small that internal spins are practically not affected by the boundary fields at all. In order to analyze the long-range case we perform our studies for $p=3$. That is a marginal case, where both energy and entropy of relevant degrees of freedom scale with L in the same way [2,10,16–18]. There is no wetting transition for fields which drop off more slowly than $1/l^3$. In this case the interface remains pinned to the wall at all finite temperature. For $p>3$ the entropic contribution dominates and critical behavior is the same as for the short-range case.

A. Magnetic susceptibility

The singularity (or a maximum) of the magnetic susceptibility χ is one of the most popular criteria for localization of a phase transition (or a pseudophase transition for a finite system). The magnetic susceptibility can be calculated directly from the fluctuations of the total magnetization. It is used, e.g., in Monte Carlo simulations [10]. This is less convenient for the DMRG method, where the free energy is calculated straightforwardly with very high accuracy. Therefore it is natural to use an alternative expression, known from thermodynamics [19], relating the magnetic susceptibility to

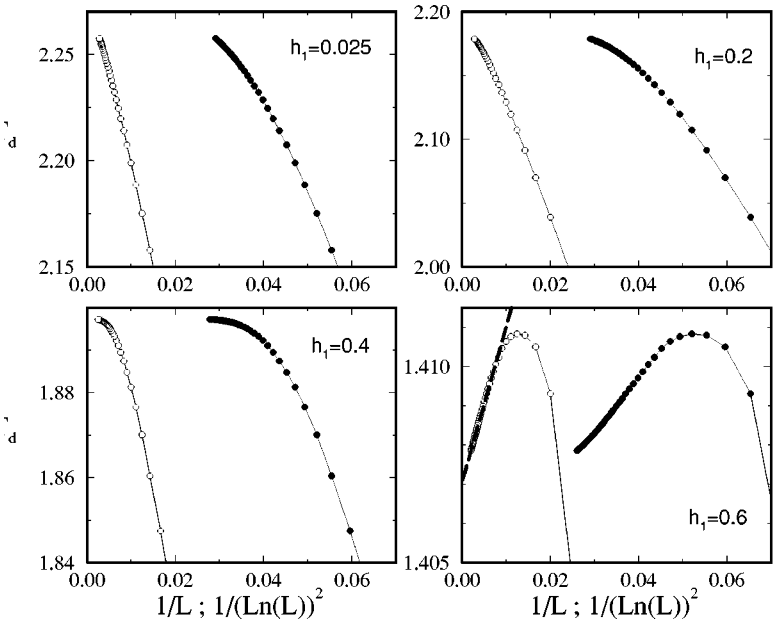

FIG. 3. Plots of $T_d(h_1;L)$ versus $1/L$ (open circles) and versus $(\ln L)^{-2}$ (filled circles) for different values of the reduced amplitude of the boundary field h_1 at $p=3$ (L in units of lattice constant; T_d in units of J/k_B). The long-dashed line is a guide for the eye.

the second derivative of the free energy f with respect to the bulk magnetic field h.

Nevertheless our case is a bit special, because we want to determine the transition line at zero bulk field (see Fig. 2), where in the dry regime (below the wetting transition line) there is a first order transition region. For infinite system there is a coexistence of phases with opposing magnetizations. So, there is a discontinuity of the first derivative of the free energy (a jump of the magnetization $m=-df/dh$), when the bulk magnetic field changes sign. That is the reason why, in order to calculate χ (a reaction of a system for a change of the bulk magnetic field) here, one should calculate the derivatives for small nonzero bulk fields and then let h go to zero. In the wet regime (above the wetting transition line) the most likely are the configurations, where an interface meanders freely between walls and where there is no discontinuity of the free energy derivatives, when the $h=0$ plane is crossed.

For numerical calculations (as for the DMRG method) the necessity of performing an extra limit ($h\to 0$ in this case) is troublesome. Therefore we decided to use another quantity χ_0 instead of χ, which is also the second derivative of the free energy at fixed T and h_1, but calculated in a symmetrical way with respect to the $h=0$ plane (by means of the free energy values taken for five equidistant points: $-2\Delta h$, $-\Delta h$, 0, Δh or $2\Delta h$; we used $\Delta h=10^{-5}$ typically). Because our calculations are always carried out for finite L, there is not anymore a discontinuity of the magnetization in the dry regime. They are replaced by rounded, but very steep, functions when the $h=0$ plane is crossed.

In order to determine the ID transition we scan the phase diagram at fixed h_1. The higher the temperature is, the less steep are magnetizations and the values of their derivative χ_0 are smaller. Above the wetting transition line, where a discontinuity never exists, χ_0 saturates to the zero, when T grows (χ_0 is equivalent to χ here). Therefore, at fixed L, the ID transition can be indicated by the maximal slope of the χ_0 or the minimum of its derivative with respect to temperature. Although all derivatives have been performed in a numerical way, the DMRG method accuracy guarantees very precise results.

In contrast to the $d=2$ Ising strip with short-range boundary fields or long-range boundary fields with $p>3$ where the scaling is $T_d(L)-T_w\approx L^{-1}$, Albano *et al.* [10] have proposed an alternative formula for the long-range case at $p=3$. They argued that the ID transition may be shifted away from the wetting transition temperature T_w according to the rule $T_d(L)-T_w\approx(\ln L)^{-2}$. It would mean that $T_d(L)$ would be more strongly depressed from T_w with decreasing L than in the short-range case. Because their data did not allow them to distinguish between these two possibilities we have considered this problem again.

The plots of $T_d(L)$ in Fig. 3 show that the scaling behavior is more complicated than it was expected before. However, it is worth recalling that Albano *et al.* [10] analyzed the scaling of the χ maxima, whereas we analyze the scaling of the χ_0 maximal slope. For both extrapolations (against $1/L$ and against $1/(\ln L)^2$) and various values of the reduced amplitude of the boundary field h_1 there are higher order corrections that seem to be opposite to the leading term. The most likely scenario is that due to them there are maxima for some values of L and only for larger L the data apparently follow the leading term. The smaller h_1 is the larger L is neccessary to reach a maximum. Therefore we are not able to present them for $h_1=0.025$ and $h_1=0.2$, although our calculations have been done up to $L=300-400$.

In order to distinguish between both extrapolations we compare the data at $h_1=0.6$ for L larger than the maximum position (up to $L=490$). Though there is no strong difference for both curves we have checked that the better linear approximation is for the $1/L$ case. Therefore it is very likely that the scaling for long-range $p=3$ boundary fields has the same asymptotic form as for short-range ones.

B. Scaling of profiles

Our second criterion is based on a shape of a profile. It is known [20] that for the short-range case for infinite Ising

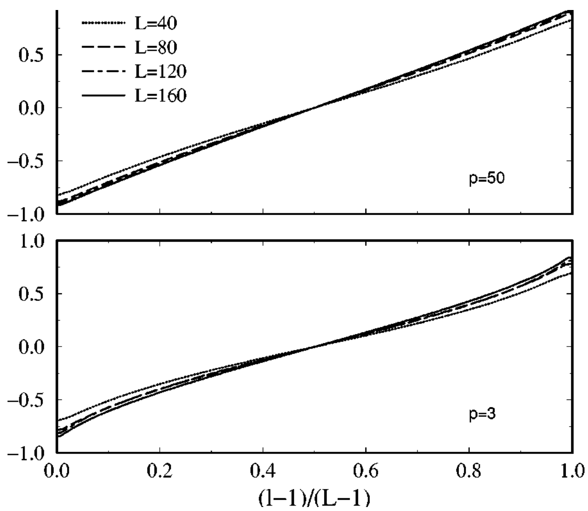

FIG. 4. Magnetization profiles $m(l)$ versus $(l-1)/(L-1)$ at $T=1.8$ [$(l-1)/(L-1)$ and magnetization are dimensionless].

strips with opposing boundary fields at the ID transition the interface meanders in such a way that the magnetization gradient is constant over the whole width L. Therefore the magnetization profile is characterized by the scaling function of a linear form

$$m(l) = m_b\left(1 - \frac{2l}{L}\right). \tag{4}$$

In order to localize the ID transition $T_d(h_1;L)$ at a certain temperature T, boundary field scans can be performed for lengths L and $L+2$. The profiles are compared according to the quantity

$$d(L) = \left[\frac{1}{L}\sum_{l=1}^{L}\left(\frac{m(l)}{m_b}\right)^2\right]^{1/2} \tag{5}$$

which measures a deviation from the bulk magnetization. To compare the data for L and $L+2$ we rescale l to value $(l-1)/(L-1)$ which varies between 0 and 1. Then at the value of h_1, where $d(L)$ and $d(L+2)$ cross each other as functions of h_1, both profiles are the most similar. This optimal scaling corresponds to the ID transition. The procedure can be applied to the long-range case as well. As one can see in Fig. 2 the limiting $L\rightarrow\infty$ results are in an excellent agreement with those from the first criterion.

It is worth stressing that although profiles for subsequent L and $L+2$ are of the same shape [$d(L)=d(L+2)$ on the ID transition line] they change when L grows. Figure 4 collects the data for both p at $T=1.8$. For $p=50$, which means the short-range boundary fields, the profiles collapse rapidly to the straight line. The larger L is the smaller are the minor deviations at ends. A similar scenario holds at $p=3$. Although generally plots are here less linear, they seem also to converge towards the universal shape predicted for the case of short-range boundary fields.

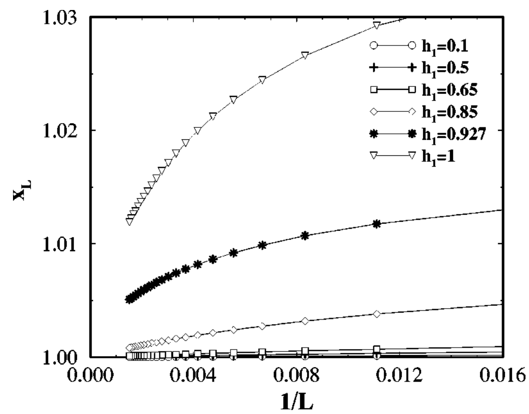

FIG. 5. The plots of x_L versus $1/L$ for the short-range boundary fields ($p=9$) (L in units of lattice constant; x_L dimensionless).

C. Specific heat

The third criterion is based on the behavior of the specific heat C_{h_1}, which is calculated as the second derivative of the free energy with respect to temperature at fixed h_1. It is known [21] that there are two maxima of C_{h_1}. Whereas the stronger one is related to T_c [22], the weaker one corresponds to the wetting. When h_1 goes down the second maximum merges in the first one, which prevents one from calculating $T_d(h_1;L)$ for small h_1.

As one can see in Fig. 2 the third criterion results coincide with the previous ones.

IV. KELVIN EQUATION

In order to find the corrections to the Kelvin equation it is preferable to consider only temperatures not too close to T_c, where DMRG iterations converge very quickly. Moreover, when one is far below T_c, one avoids the effect of crossover from an initial scaling (for small L) according to Eq. (2) towards the final scaling (for enough large L) according to the Kelvin equation. In the present study we have calculated a series of values of $h_0(L)$ up to $L=690$ with different values of boundary fields at $T=1$.

Our calculations have been carried out for two ranges of boundary fields: $p=3$ and $p=9$. Whereas the first case is the marginal long-range case for wetting, the second one is to show that here we practically meet short-range behavior.

We assume the following expansion for the bulk magnetic field h_0, which restores the phase coexistence:

$$h_0(L) = \frac{A}{L^\alpha} + \frac{B}{L^\gamma} + \cdots , \tag{6}$$

where we expect $\alpha=1$ and A to be given by the Kelvin equation [Eq. 1]. In order to calculate the exponents α and γ we define the logarithmic derivatives:

$$x_L \equiv -\frac{\ln[h_0(L)] - \ln[h_0(L+30)]}{\ln L - \ln(L+30)}, \tag{7}$$

for $L=30,60,\ldots,690$ (in steps of $L=30$). Then introducing the expansion (6) into the definition (7) one has to the lowest orders in $1/L$

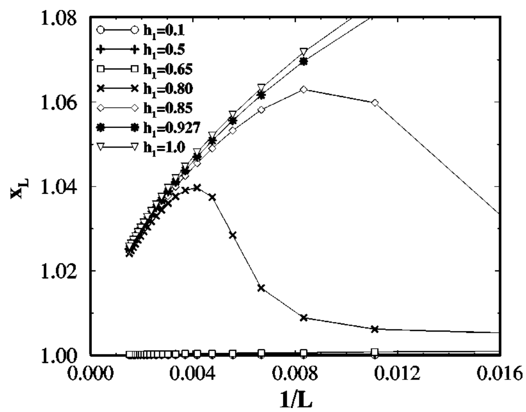

FIG. 6. The plots of x_L versus $1/L$ for the long-range boundary fields ($p=3$) (L in units of lattice constant; x_L dimensionless).

$$x_L = \alpha\left(1 - \frac{B}{A}\frac{1}{L^{\gamma-\alpha}} + \cdots\right). \tag{8}$$

Figures 5 and 6 show plots of x_L versus $1/L$ for different values of boundary fields at $p=9$ and 3, respectively. For the first case the wall-spin interactions decays so fast that there we expect short-ranged boundary behavior. From the exact solution of Abraham one knows that the surface field corresponding to the wetting temperature $T_w=1$, is equal to $h_1 \approx 0.927$. Therefore it is clear that the plots in Fig. 5 for $h_1 < 0.927$ correspond to the case of partial wetting, where the scaling behavior of x_L should be linear in $1/L$ since one expects $\gamma=2$ and $\alpha=1$ in Eq. (6). The fact that the data follow straight lines confirms the behavior predicted by the theory for the short-range case ($p=9$). It is worth noticing that at $h_1=0.85$, the data follow a straight line only for large L. Therefore, we can conclude that the closer h_1 is to the coexistence line value (at $h_1 \approx 0.927$ here), the larger L is necessary to reach an asymptotic regime.

A qualitatively similar behavior is observed for the long-range boundary fields in Fig. 6 (at $p=3$), but the linear dependence occurs here only below $h_1=0.80$. It is in perfect agreement with our data for the $p=3$ wetting transition (see Fig. 2), where $T_w=1$ for $h_1 \sim 0.73$.

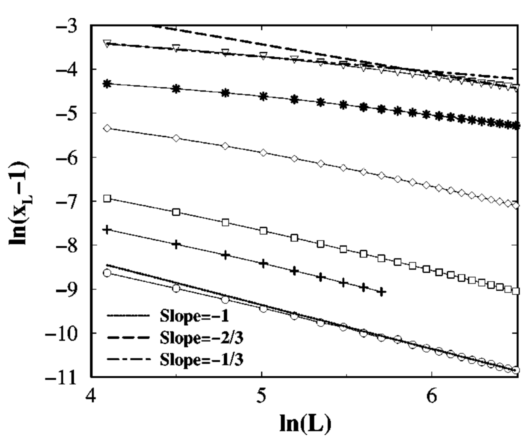

FIG. 7. The plots of $\ln(x_L-1)$ versus $\ln L$ for the short-range boundary fields ($p=9$). The meaning of the symbols is the same as in Fig. 5 (L in units of lattice constant; x_L dimensionless).

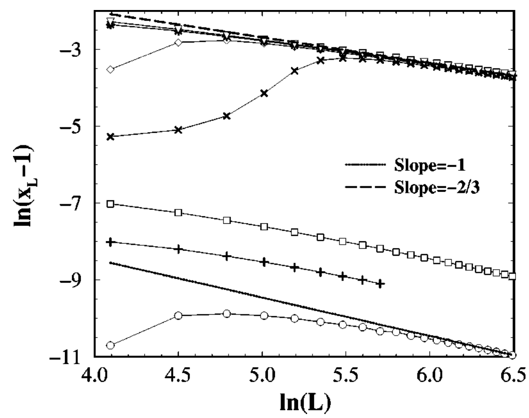

FIG. 8. The plots of $\ln(x_L-1)$ versus $\ln L$ for the long-range boundary fields ($p=3$). The meaning of the symbols is the same as in Fig. 6 (L in units of lattice constant; x_L dimensionless).

As one can see in Figs. 5 and 6 the values of the leading scaling exponent, obtained from the DMRG converge very well to the expected value $\alpha=1$ [11]. Thus the Kelvin equation dependence is of fairly general character and it does not depend on the range of the boundary fields, which is in an agreement with the Monte Carlo results [10]. To consider higher-order corrections (setting $\alpha=1$) one obtains from Eq. (8)

$$\ln(x_L - 1) = \ln\left|\frac{B}{A}\right| - (\gamma - 1)\ln L + \cdots . \tag{9}$$

Figures 7 and 8 show plots of $\ln(x_L-1)$ versus $\ln L$ for different values of surface fields at $p=9$ and 3, respectively. In both cases the behavior at partial and complete wetting can be clearly distinguished.

At partial wetting the straight dotted lines with slope -1 fit very well the asymptotic behavior, so $\gamma=2$ here. At complete wetting for large L the plots are well fitted with dashed lines with slope $-2/3$ ($\gamma=5/3$) in agreement with the current theory. However, one can observe that the smaller the range of the boundary fields is, the larger L is necessary to reach

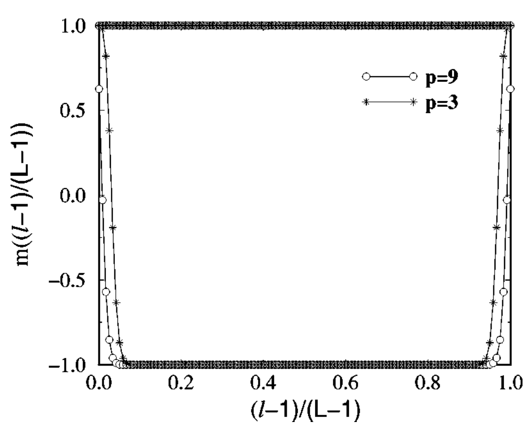

FIG. 9. The magnetization profiles of the two coexisting phases at $T=1$ and $h_1=1$ (for $L=120$), approaching complete wetting, for different boundary fields [$(l-1)/(L-1)$ and magnetization are dimensionless].

the asymptotic regime. Therefore, in agreement with the previous results [11], in Fig. 7 (a short-range case practically) the data for a large interval of L (more or less up to $L=250$) are well fitted by straight lines with slope $-1/3$ ($\gamma=4/3$). It is consistent with the previous explanation that even for large system sizes, like $L=100-200$, the wetting layer has a limited thickness, so the singular part of the surface free energy that determines the leading correction term behavior, is dominated by the contacts with the walls. In this case a simple random walk argument predicts the correction of the type $\gamma=4/3$ for a thin wetting layer [11]. When the range of the boundary fields grows, a wetting layer is getting thicker and for smaller L the asymptotic limit ($\gamma=5/3$) is reached (see Fig. 8).

The above arguments are illustrated in Fig. 9, where the magnetization profiles for two coexisting phases are presented. The magnetizations are plotted as a function of the scaled variable $(l-1)/(L-1)$ at $T=1$ and $h_1=1$; this corresponds to a regime of complete wetting for both p. We should also stress that the profiles refer to bulk fields slightly lower and higher than the coexistence field $h_0(L)$. For bulk field exactly equal to $h_0(L)$ the magnetizations of the two coexisting phases are averaged and it is not possible to distinguish between them. The negative bulk field favors a bulk phase with negative magnetization (vapor) but the positive boundary fields favor the adsorption of positive spins (liquid) at the boundaries. Since for $T \geqslant T_w$ the positive spins form a layer that wets the walls. Obviously for a long-range boundary fields ($p=3$) the wetting layer is thicker than for the short-range case ($p=9$).

V. CONCLUSIONS

We have analyzed the influence of long-range boundary fields on the thermodynamics of an Ising model in a strip geometry comparing the results to corresponding studies with short-range boundary fields.

In the case of opposing fields at the walls the transition line of wetting was found for various values of the fields. In order to determinate its location three criteria have been applied [based on the (pseudo) magnetic susceptibility, magnetization profiles and specific heat] resulting in the data which coincide with one another in a perfect way. The data for the short-range wall-particle interactions are in a full agreement with the exact result. For the long-range wall-particle interactions the transition line was found for the marginal case ($p=3$), where both energy and entropy of relevant degrees of freedom scale with L in the same way. It is worth noticing that this line is in agreement with the shift of the coexistence line at fixed temperature found for the identical boundary fields.

In the case of identical boundary fields we have confirmed that the bulk coexistence field scales always according to the Kelvin equation and have verified the previous results for leading correction terms at partial and complete wetting. We have found that for the long-range case the wet layer is thicker with respect to the short-range case which results in shrinking of the region with the leading correction term of the type $1/L^{4/3}$. This confirms the previously proposed scenario to explain the origin of the anomalous corrections to the Kelvin equation at complete wetting.

Generally, our studies have shown that a range of boundary fields is not relevant at the marginal case $p=3$ in the asymptotic regime of large L.

ACKNOWLEDGMENTS

A.D. wants to thank A. Maciołek and A. O. Parry for stimulating discussions. We would also like to thank A. Maciołek and J. M. J. van Leeuwen for a critical reading of the manuscript.

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