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Interface localization transition in Ising films with competing walls: Ginzburg criterion and crossover scaling

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In a simple fluid or Ising magnet in a thin film geometry confined between walls a distance D apart that exert opposing surface fields, an interface parallel to the walls is stabilized below the bulk critical temperature $T_{\rm cb}$. While this interface is ''delocalized'' (i.e., freely fluctuating in the center of the film) for $T_{\rm cb} > T > T_c(D)$, below the ''interface localization transition'' temperature $T_c(D)$ the interface is bound to one of the walls. Using the mean field description of Parry and Evans [Physica A **181**, 250 (1992)], we develop a Ginzburg criterion to show that the Ginzburg number scales exponentially with thickness, $\mathrm{Gi} \propto \exp(-\kappa D/2)$, κ^{-1} being the appropriate transverse length scale associated with the interface. Therefore, mean field theory is self-consistent for large D, thus explaining why recent Monte Carlo simulations observed Ising criticality only in a very close neighborhood of $T_c(D)$. A crossover scaling description is used to work out the thickness dependence of the critical amplitudes in the Ising critical regime. Extending these concepts to consider finite size effects associated with the lateral linear dimension L, we reanalyze the Monte Carlo results of Binder, Landau, and Ferrenberg [Phys. Rev. B **51**, 2823 (1995)]. The data are in reasonable agreement with the theory, provided one accepts the suggestion of Parry *et al.* [Physica A **218**, 77 (1995); **218**, 109 (1995)] that the length scale $\kappa^{-1} = \xi_b(1+\omega/2)$, where ξ_b is the true correlation range in the bulk, and ω is the universal amplitude associated with the interfacial stiffness.

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I. INTRODUCTION

In thin films of simple fluids or fluid mixtures it is rather natural to have two different walls confining the system (e.g., one solid substrate on the bottom and air on top, two inequivalent solid surfaces forming a slit, etc.). Recently it has been found that the competition between surface effects and finite size effects leads to interesting phase behavior with interesting types of phase transitions [1–9]. Figure 1 sketches the situation for the generic model, the Ising lattice model (remember the equivalence between Ising ferromagnets and lattice gas models of fluids) with opposite surface magnetic fields $H_D = -H_1$ [10]. For any finite value of D, irrespective how large D is, the transition that would occur in the bulk at $T_{\rm cb}$ is rounded in the thin film geometry with competing fields. The local order near the walls, already induced by the surface fields at temperatures T far above the bulk critical temperature $T_{\rm cb}$, spreads out over the whole film as the correlation length grows. But this ordering is a smooth process, and no transition temperature can be identified where the interface between positively and negatively magnetized regions in the ferromagnetic Ising film appears. As long as this interface occurs with the highest probability in the center of the film, no spontaneous order parameter can be identified for the film, although there would already be a spontaneous magnetization in the bulk. However, in order to decrease the configurational entropy of the interface at low temperatures, a transition takes place at $T_c(D)$ where the interface becomes localized near one of the walls. For $|H_D| = |H_1|$, both situations shown in Fig. 1 for $T < T_c(D)$ are equally likely; there is a symmetry breaking of two-dimensional (2D) Ising character at $T_c(D)$. But as $D \to \infty$, $T_c(D) \to T_w(H_1)$, which is the wetting transition temperature [1,5], and the situation becomes equivalent to the problem of an interface bound to one wall in a semi-infinite geometry [11–15]. Thus the limit $D \to \infty$ for a film with competing walls is fairly special. While for any finite D there is a *single* transition at $T_c(D)$, presumably belonging to the two-dimensional Ising universality class [16], in the limit of $D \to \infty$ *two distinct* transitions appear in a discontinuous fashion: the wetting transition at $T_w(H_1)$ and the bulk transition at $T_{\rm cb}$ (now belonging to the three-dimensional Ising model universality class). Although $T_c(D)$ is a singularity of the total free energy of the thin film for any finite D, its limit

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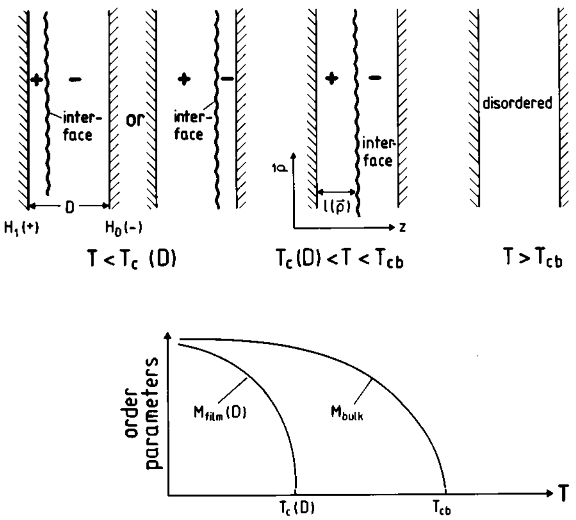

FIG. 1. Schematic description of the interface localization-delocalization transition of an Ising model confined between two walls a distance D apart, where at one wall (left side) a positive field H_1 acts, while on the other wall (right side) a negative field H_D acts. For $T<T_c(D)$ the interface is bound either to the left or the right wall, and then the average magnetization $M_{\text{film}}(D)$ of the film is nonzero. For $T_c(D)<T<T_{\text{cb}}$, however, the interface fluctuates delocalized in the center of the film, and thus $M_{\text{film}}(D)=0$, although there is still a nonzero bulk magnetization $\pm M_{\text{bulk}}$ in an infinite system, as well as locally in the film away from the interface. For $T>T_{\text{cb}}$, however, the film is disordered (apart from the response to the surface fields near the walls). The description of the interface in terms of a coordinate $z=1(\vec{\rho})$, with $\vec{\rho}$ being the x,y coordinates in the plane formed by the left wall, is also indicated.

$T_w(H_1)$ for $D\to\infty$ is not a singularity of the bulk free energy of the system, but only of the surface free energy [11–15].

It is clear that the subtleties of the associated critical behavior cannot be fully accounted for by mean field theory [1,5], and since three distinct types of criticality compete (2D Ising, 3D Ising, and critical wetting [17,18]), analytical approaches to the understanding of the critical behavior seem difficult. Hence three of us [7–9] have tried to verify the above picture by Monte Carlo simulations. While the general features of the scenario sketched above are certainly in agreement with the numerical findings of this work [7–9], it came as a surprise that the presumed two-dimensional Ising character could only be verified for extremely thin films (such as $D=6$ and 8, measuring lengths in units of the lattice spacing). Even for films as thin as $D=12$, very strong deviations from two-dimensional Ising criticality were found. Another problem was that the length scale κ^{-1}, associated with the interface in the transverse direction, was not identifiable with the bulk correlation length ξ_b, as suggested by the mean field theory [5], but was distinctly larger [8]. This discrepancy has also stimulated extensive theoretical activity, which in turn improved on the theoretical description of complete wetting phenomena [19–22].

In the present paper, we address these problems first by formulating a Ginzburg criterion [23–26], by which the self-consistency of the mean field description of Parry and Evans [1,5] for this interface delocalization transition is investigated (Sec. II). We find that the Ginzburg number Gi, which rules the crossover from mean field to two-dimensional Ising behavior, scales as $\exp(-\kappa D/2)$. From a crossover [27] scaling type description analogous to [28–31], predictions for the critical amplitudes of various quantities in the Ising regime also are obtained. The consequences of this crossover scaling description in the context of finite size effects associated with the finite extension of the linear dimension L parallel to the walls are also spelled out.

Section III then presents additional analyses of the Monte Carlo data of Binder, Landau, and Ferrenberg [8] in order to estimate critical amplitudes and study their dependence on film thickness D. Unfortunately, for the available range of D the crossover scaling variable $\exp(-\kappa D/2)$ is not really small enough to allow for a stringent test of the theory. However, we do find that the numerical results are compatible with the theoretical predictions, provided we use the prediction of Parry and co-workers [19–22],

$$\kappa^{-1}=\xi_b(1+\omega/2), \tag{1}$$

as the transverse length scale κ^{-1}.

Here ξ_b is the true correlation range in a lattice direction in the bulk, which is known from independent estimates [32,33], and ω is the universal constant entering the theory of critical wetting [34–39],

$$\omega=k_BT/(4\pi\xi_b^2\sigma), \tag{2}$$

σ being the interfacial stiffness constant. The direct numerical estimates for κ^{-1} [8] are available only at temperatures distinctly higher than $T_c(D)$, but when extrapolated to the temperatures of interest they are roughly compatible with Eq. (1) (see also Ref. [22]). Thus, our analysis has at least an indirect bearing on problems with critical wetting, since unlike other simulation studies [40–42] the present analysis is compatible with recent estimates of ω [39]. Finally, Sec. IV briefly summarizes some conclusions.

II. CRITICAL BEHAVIOR OF INTERFACE LOCALIZATION TRANSITIONS: A PHENOMENOLOGICAL THEORY

A. Mean field theory

Treating the interface between coexisting phases in the sharp kink approximation [5,11–14], we write the effective Hamiltonian in terms of a collective coordinate $l(\vec{\rho})$, namely the local distance of the interface from the left wall at position $\vec{\rho}$ in the xy plane (which coincides with this wall for $z=0$ while the right wall is at position $z=D$; cf. Fig. 1). In terms of the interfacial stiffness constant σ already alluded to in Eq. (2), this effective Hamiltonian then is

$$\mathcal{H}_{\text{eff}}\{l\}=\int d\vec{\rho}\left[\frac{\sigma}{2}(\nabla l)^2+\sum\{l(\vec{\rho})\}\right], \tag{3}$$

where the potential $\Sigma(l)$ in the presence of a bulk field H is (see [5])

$$\Sigma(l)=-\delta\varepsilon a_0[e^{-\kappa l}+e^{-\kappa(D-l)}]+b_0[e^{-2\kappa l}+e^{-2\kappa(D-l)}] -h(l-D/2), \tag{4}$$

where $h \equiv 2m_b H$. Here $a_0, b_0 > 0$ are constants, $\delta\varepsilon \equiv (T_w - T)/T_w$ is the normalized distance from the wetting transition temperature (in the corresponding semi-infinite geometry), and κ^{-1} is the transverse length scale alluded to in Eq. (1). As the binding potential Eq. (4) is constructed from a mean field theory of wetting films, it follows that κ^{-1} is simply $\kappa_{\rm MF}^{-1} = \xi_b$, the bulk correlation length. Note that Eq. (4) explicitly assumes short range forces due to the walls at $z=0$ and $z=D$. This is appropriate for the lattice gas simulations of Refs. [7–9,40–42], but would not be suitable for real fluids where long range van der Waals forces act [13,14]. We also note that Eqs. (3) and (4) describe the problem in terms of a *single* collective coordinate l, although recently it has been suggested that additional collective coordinates (describing the order parameter profile close to the walls at $z=0$ and $z=D$) should be included [19–21]. However, at this point we aim only at a description of the most simple mean field theory of the interface delocalization transition.

In this spirit, the fluctuations of the interface position, included in Eq. (3) via the $(\nabla l)^2$ term, are neglected altogether, and hence $\Sigma(l)$ is treated as an effective free energy function which simply needs to be minimized in order to find the average position of the interface $l_{\rm eq}$ in thermal equilibrium. For zero field $h=0$ and $T > T_w$ it is clear that all terms in Eq. (4) are positive, and assume their minimum for $l_{\rm eq} = D/2$, i.e., we are in the delocalized phase of the film. For $T < T_w$, however, $\delta\varepsilon > 0$ in Eq. (4), so that the terms with the square bracket in Eq. (4) compete against each other, and a solution $l_{\rm eq} \neq D/2$ (i.e., a nonzero order parameter) is expected.

For $h=0$ the equilibrium condition hence yields

$$0 = \left.\frac{\partial \Sigma(l)}{\partial l}\right|_{l=l_{\rm eq}} = \delta\varepsilon a_0 \kappa [e^{-\kappa l} - e^{-\kappa(D-l)}] - 2b_0\kappa[e^{-2\kappa l} - e^{-2\kappa(D-l)}], \quad (5)$$

and setting $l_{\rm eq} = D/2 + \Delta$, with $\Delta \to 0$, we obtain an equation for the order parameter Δ,

$$0 = -\delta\varepsilon a_0 \sinh(\kappa\Delta) + 2b_0 \exp(-\kappa D/2)\sinh(2\kappa\Delta). \quad (6)$$

Expanding the sinh in terms of $X = \kappa\Delta$ for $X \ll 1$ yields ($\sinh x \approx x + x^3/3$)

$$X = \pm \frac{\exp(\kappa D/4)}{\sqrt{2b_0/a_0}} t^{1/2}, \quad (7)$$

where $t \equiv [T_c(D) - T]/T_w$ is the reduced distance from the transition temperature in the thin film,

$$\delta\varepsilon = t + \frac{4b_0}{a_0}\exp(-\kappa D/2) \equiv t + \delta\varepsilon_c(D). \quad (8)$$

Equation (8) shows that $T_c(D)$ differs from T_w only by terms exponentially small in $\kappa D/2$ [5]. From Eq. (7) we now obtain the order parameter of the film as (in mean field theory)

$$M_{\rm MF} \equiv 2m_b \Delta/D \approx \pm 2m_b \exp(\kappa D/4)/(\kappa D) t^{1/2}/\sqrt{2b_0/a_0}, \quad t \to 0, \quad (9)$$

and defining a mean field critical amplitude $\hat{B}_{\rm MF}$ of the order parameter from the equation $M_{\rm MF} = \hat{B}_{\rm MF} t^{\beta_{\rm MF}}$, with $\beta_{\rm MF} = \frac{1}{2}$, we find that $\hat{B}_{\rm MF}$ scales with film thickness as

$$\hat{B}_{\rm MF} \propto \exp(\kappa D/4)/D. \quad (10)$$

Next we turn to the response function (susceptibility) at temperatures above $T_c(D)$, using

$$\left.\frac{\partial^2 \Sigma}{\partial l^2}\right|_{\rm eq} = 2\exp[-\kappa D/2]\kappa^2[-a_0\delta\varepsilon + 4b_0\exp(-\kappa D/2)], \quad (11)$$

where on the right hand side $l_{\rm eq} = D/2$ was already inserted. As expected, $\partial^2\Sigma/\partial l^2|_{l_{\rm eq}}$ vanishes at $\delta\varepsilon = \delta\varepsilon_c(D)$ [Eq. (8)], as it should for a second order transition from $M_{\rm MF} \equiv 0$ for $T > T_c(D)$ to $M_{\rm MF} \neq 0$ for $T < T_c(D)$.

Defining the response function for nonzero h as

$$\overline{\chi} \equiv (\partial l_{\rm eq}/\partial h)_{T,D}, \quad (12)$$

Eqs. (4), (5), and (11) yield

$$\left(\frac{\partial l_{\rm eq}}{\partial h}\right)_{T,D}\left(\frac{\partial^2\Sigma}{\partial l^2}\right)_{l_{\rm eq}} = 1, \quad \overline{\chi} = \left(\frac{\partial^2\Sigma}{\partial l^2}\right)_{l_{\rm eq}}^{-1}, \quad (13)$$

and hence

$$\overline{\chi} = \frac{\exp[\kappa D/2](-t)^{-1}}{2\kappa^2 a_0}. \quad (14)$$

The susceptibility $\chi_{\rm MF} = \partial M_{\rm MF}/\partial H$ then becomes $\chi_{\rm MF} = 4m_b^2 \overline{\chi}/D$, and, defining a critical amplitude $\hat{C}_{\rm MF}^+$ in the mean field critical regime from

$$\chi_{\rm MF} = \hat{C}_{\rm MF}^+ (-t)^{-\gamma_{\rm MF}}, \quad \gamma_{\rm MF} = 1, \quad (15)$$

we conclude that

$$\hat{C}_{\rm MF}^+ = \frac{2m_b^2}{\kappa^2 a_0}\exp[\kappa D/2]/D \quad (16)$$

[the plus sign in Eqs. (15) and (16) refers to $T > T_c(D)$, and the minus sign to $T < T_c(D)$].

It is also interesting to note the magnitude of the susceptibility at the wetting temperature T_w of the semi-infinite system, where $\delta\varepsilon = 0$, and hence [Eq. (11)] $\partial^2\Sigma/\partial l^2|_{l_{\rm eq}} \propto \exp(-\kappa D)$, is

$$\chi_{\rm MF}(T = T_w) \propto \exp(\kappa D).$$

The response function for $T < T_c(D)$ can also be worked out easily; as always, in Landau-type theories it is of the same form as Eq. (15), $\chi_{\rm MF} = \hat{C}_{\rm MF}^- t^{-1}$, with $\hat{C}_{\rm MF}^+/\hat{C}_{\rm MF}^- = 2$ being the universal critical amplitude ratio in the mean field critical regime.

Finally we are interested in the correlation length near the transition. For that purpose we define the deviation of the interface position from its mean value $\delta l(\vec{\rho}) \equiv l(\vec{\rho}) - l_{\text{eq}}$. We wish to consider a correlation function of fluctuations in the direction parallel to the walls,

$$G_{\parallel}(\vec{\rho}-\vec{\rho}^{1}) \equiv \langle \delta l(\vec{\rho}) \delta l(\vec{\rho}^{1}) \rangle. \tag{17}$$

From a quadratic expansion of Eq. (3) around l_{eq} it is clear that the problem is analogous to the treatment of correlations in standard Ginzburg-Landau theory; hence one concludes $G_{\parallel}(\vec{\rho}-\vec{\rho}^{1})$ is of Ornstein-Zernike form, and the correlation length is related to the response function in the standard way,

$$\xi_{\parallel}^{-2} = \frac{1}{\sigma} (\partial^2 \Sigma / \partial l^2)_{l_{\text{eq}}} = (\overline{\chi\sigma})^{-1}. \tag{18}$$

From Eqs. (14) and (18) we conclude that the critical amplitude $\hat{\xi}_{\text{MF}}^{+}$ defined from $\xi_{\parallel}^{+} = \hat{\xi}_{\text{MF}}^{+}(-t)^{-\nu_{\text{MF}}}$, with $\nu_{\text{MF}} = \frac{1}{2}$, so that

$$\hat{\xi}_{\text{MF}}^{+} = \kappa^{-1} \left(\frac{\sigma}{2a_0} \right)^{1/2} \exp[\kappa D/4] \tag{19}$$

and $\hat{\xi}_{\text{MF}}^{+} / \hat{\xi}_{\text{MF}}^{-} = \sqrt{2}$, as usual.

B. Ginzburg criterion

As is well known [23–26], mean field theory is self-consistent if the order parameter fluctuations in a correlation volume are small in comparison to the square of the mean order parameter. For our geometry, with $\vec{r} = (\vec{\rho}, z)$, we obtain

$$\int_0^D dz \int_{\rho < \xi_{\parallel}} d^{d-1}\rho [\langle M(\vec{r}^{1}) M(\vec{r}^{1}+\vec{r}) \rangle - M_{\text{MF}}^2] \ll D \xi_{\parallel}^{d-1} M_{\text{MF}}^2. \tag{20}$$

Near the transition, $\xi_{\parallel}$ is very much larger than D, and the inhomogeneity of $M(\vec{r})$ in the pure phases [for $z < l(\vec{\rho})$ or $z > l(\vec{\rho})$, respectively] can be ignored; it contributes only prefactors of order unity. One concludes that Eq. (20) is equivalent to the standard form of the Ginzburg criterion, expressed by the condition that the Ginzburg number Gi is small, with [25,26,30]

$$\text{Gi} \equiv \hat{C}_{\text{MF}}^{+} \hat{B}_{\text{MF}}^{-2} k_B T / (\hat{\xi}_{+}^{\text{MF}})^2 D = \exp(-\kappa D/2) \; \frac{2 b_0 k_B T \kappa^2}{a_0 \sigma}, \tag{21}$$

$$\text{Gi} \ll |t|. \tag{22}$$

In Eq. (21), we have specialized to $d = 3$ in Eq. (20); i.e., we consider a two-dimensional criticality in the thin film. Using Eqs. (10), (16), and (19), we conclude that the mean field theory of the interface localization-delocalization transition (as derived by Parry and Evans [1,5]) is self-consistent if

$$\text{Gi} \propto \exp(-\kappa D/2) \ll |t|. \tag{23}$$

We see that *the Ginzburg number decreases exponentially fast* with film thickness, and hence for large D mean field theory should be an excellent description even though we deal with a two-dimensional Ising-type criticality in a system with short range forces. This conclusion, which was surprising and unexpected, is one of the central results of the present paper.

C. Crossover scaling

From the fact that the Ginzburg number is given by Eq. (23), we can immediately derive scaling predictions for the Ising critical regime as well, using a phenomenological crossover scaling description [27]. Note that the crossover variable is (for large D, in leading order)

$$\mathscr{T} = t \, \exp(\kappa D/2); \tag{24}$$

i.e., one simply uses the ratio between the temperature distance and the Ginzburg number; for large $\mathscr{T}$ one has mean field critical behavior, for small $\mathscr{T}$ one has (two-dimensional) Ising criticality. Following similar routes as in related crossover problems [28–31], we find

$$M = \frac{1}{D} \exp(\kappa D/4) t^{1/2} \widetilde{M}(\mathscr{T}). \tag{25}$$

For large $\mathscr{T}$ the crossover scaling function tends to a constant, recovering Eq. (9), while for small $\mathscr{T}$ we must have

$$\widetilde{M}(\mathscr{T} \ll 1) \propto \mathscr{T}^{\beta - 1/2}, \quad \beta = \tfrac{1}{8} \tag{26}$$

in order to have the correct critical behavior of the order parameter, in agreement with the Ising model. From Eqs. (25) and (26) we obtain

$$M(t \to 0) = \hat{B}(D) t^{\beta},$$

$$\hat{B}(D) \propto \frac{1}{D} \exp(\kappa D \beta/2) = \frac{1}{D} \exp(\kappa D/16). \tag{27}$$

For an explicit calculation of crossover scaling functions such as $\widetilde{M}(\mathscr{T})$, renormalization group methods need to be used [25].

Next we consider the susceptibility for $T > T_c(D)$,

$$\chi = \frac{1}{D} \exp(\kappa D/2)(-t)^{-1} \widetilde{\chi}(\mathscr{T}), \tag{28}$$

where again we conclude that $\widetilde{\chi}(\mathscr{T} \to -\infty)$ tends to a constant, and Eq. (28) becomes equivalent to Eqs. (15) and (16), while for $-\mathscr{T} \ll 1$ we have

$$\widetilde{\chi}(-\mathscr{T} \ll 1) \propto (-\mathscr{T})^{-(\gamma-1)}, \quad \gamma = \tfrac{7}{4}. \tag{29}$$

This yields the critical behavior of the susceptibility in the Ising regime as follows:

$$\chi = \hat{\chi}^{+}(-t)^{-\gamma},$$

$$\hat{\chi}^{+} \propto \frac{1}{D} \exp[\kappa D(2-\gamma)/2] = \frac{1}{D} \exp(\kappa D/8). \tag{30}$$

Finally we consider the correlation length $\xi_{\parallel}$, Eq. (18),

$$\xi_{\parallel} = \exp(\kappa D/4)(-t)^{-1/2} \widetilde{\xi}(\mathscr{T}), \tag{31}$$

where Eq. (19) is reproduced if $\tilde{\xi}(\mathcal{T}\to-\infty)$ tends to a constant, while for small $(-\mathcal{T})$ we have

$$\tilde{\xi}(-\mathcal{T}\ll 1)\propto(-\mathcal{T})^{(1/2)-\nu}, \quad \nu=1. \tag{32}$$

Hence the asymptotic critical behavior of the correlation length is

$$\xi_\parallel=\hat{\xi}^+(-t)^{-\nu}, \quad \hat{\xi}^+\propto\exp[\kappa D(1-\nu)/2]\propto \text{ const.} \tag{33}$$

There is no anomalous thickness dependence in the critical amplitude of the correlation length.

Finally we consider the Ginzburg combination $D\xi_\parallel^2\phi^2\chi^{-1}$: this quantity is independent of both t and D in the Ising critical region, as it should be: in the Ising critical region this quantity reduces to the universal amplitude combination $\hat{\xi}^{+2}\hat{B}^2(\hat{\chi}^+)^{-1}D$, which using Eqs. (27), (30), and (33) is independent of D.

D. Finite size scaling

We now wish to extend the above considerations to the case where the finiteness of the parallel linear dimension L competes with the growth of the correlation length $\xi_\parallel$. For this purpose we simply have to add an additional argument $L^{1/\nu}t$ in the scaling functions, remembering that $\hat{\xi}^+$ does not have any significant (i.e., exponential) dependence on the thickness of the film. Thus the order parameter becomes a function of two scaling variables $t\ \exp(\kappa D/2), L^{1/\nu}t$:

$$M=\frac{1}{D}\exp(\kappa D/4)t^{1/2}\tilde{\tilde{M}}(t\exp(\kappa D/2),L^{1/\nu}t)\underset{\text{small } t,\text{large } L}{\longrightarrow}$$
$$\to\frac{1}{D}\exp(\kappa D/16)t^{1/8}\bar{\bar{M}}(L^{1/\nu}t)\underset{t=0}{\to}\frac{1}{D}\exp(\kappa D/16)L^{-1/8}. \tag{34}$$

An alternative consideration concerns the crossover at $t=0$, where we rewrite the first line of Eq. (34) as follows, using $\nu=1$:

$$M=\frac{1}{D}\exp(\kappa D/16)t^{1/8}\tilde{\tilde{M}}(\exp(\kappa D/2)/L,Lt)$$
$$\underset{t\to 0}{\to}\frac{1}{D}\exp(\kappa D/16)L^{-1/8}\tilde{M}_c(\exp(\kappa D/2)/L). \tag{35}$$

A similar result follows for the susceptibility:

$$\chi=\frac{1}{D}\exp(\kappa D/2)(-t)^{-1}\tilde{\tilde{\chi}}(-t\ \exp(\kappa D/2),L^{1/\nu}t)$$
$$\to\frac{1}{D}e^{\kappa D/8}(-t)^{-7/4}\tilde{\tilde{\chi}}(\exp(\kappa D/2)/L,Lt)$$
$$\underset{t\to 0}{\to}\frac{1}{D}e^{\kappa D/8}L^{7/4}\tilde{\chi}_c(\exp(\kappa D/2)/L), \tag{36}$$

where $\tilde{\tilde{\chi}}$, $\tilde{\tilde{\chi}}$, and $\tilde{\chi}_c$ are the scaling functions in obvious limits.

Finally, the fourth order cumulant [43,44]

$$U_L=1-\langle M^4\rangle/3\langle M^2\rangle^2 \tag{37}$$

becomes

$$U_L=\tilde{\tilde{U}}\{t\ \exp(\kappa D/2),L^{1/\nu}t\}=\tilde{\tilde{U}}(\exp(\kappa D/2)/L,Lt)$$
$$\underset{t\to 0}{\to}\tilde{U}_c\{\exp(\kappa D/2)/L\}. \tag{38}$$

Equation (38) shows rather explicitly that, in the present problem, due to the mean field to Ising crossover, there is no unique cumulant intersection point at criticality $(t=0)$. This is compatible with the simulations [7–9], and clearly made the precise estimation of $T_c(D)$ difficult [8]. Of course, the problem is qualitatively similar to other crossover scaling problems [28–31].

III. REANALYSIS OF THE MONTE CARLO DATA

A. Model and ''raw data''

In Sec. II, a wealth of predictions was obtained which was not available when corresponding Monte Carlo simulations were published [7–9]. Thus it is very interesting to return to these simulations, where no critical amplitudes had yet been extracted, and look more closely at the critical behavior.

The model considered is a nearest neighbor Ising Hamiltonian on the simple cubic lattice, with exchange constant J,

$$\mathcal{H}=-J\sum_{\langle i,j\rangle}S_iS_j-H\sum_i S_i-H_1\sum_{i\in\text{layer } n=1}S_i$$
$$-H_D\sum_{i\in\text{layer } n=D}S_i,\ \ S_i=\pm 1. \tag{39}$$

Typically we use zero bulk field H, while the surface fields H_1 and H_D acting on the two free $L\times L$ surfaces are chosen as $H_1/J=+0.55$ and $H_D/J=-0.55$. The wetting transition in the semi-infinite geometry would then be estimated to occur at [41] $k_BT_w/J\approx 4.00$ which is sufficiently below the bulk critical temperature $T_{\rm cb}$ ($k_BT_{\rm cb}/J\cong 4.511\,42 \pm 0.000\,05$ [45]) that the correlation length ξ_b near T_w is only of the order of about one lattice spacing. This temperature range is far away from the roughening transition temperature T_R ($k_BT_R/J\approx 2.445$ [46]), so we are safely in the regime of wetting transitions, avoiding any problems due to crossover toward layering transitions [47]. We choose a $L\times L\times D$ geometry with $D=6$, 8, and 12, and parallel linear dimensions L in the range from $L=16$ to 256, with periodic boundary conditions in both x and y directions. More details about the simulation technique can be found in Ref. [8].

The quantities that are reanalyzed here are the average absolute value $\langle|M|\rangle$ of the magnetization of the film,

$$M=(L^2D)^{-1}\sum_i S_i, \tag{40}$$

the corresponding susceptibility χ^1 [48],

$$\chi^1=L^2D(\langle M^2\rangle-\langle|M|\rangle^2)/k_BT, \tag{41}$$

and the fourth order cumulant U_L [Eq. (37)].

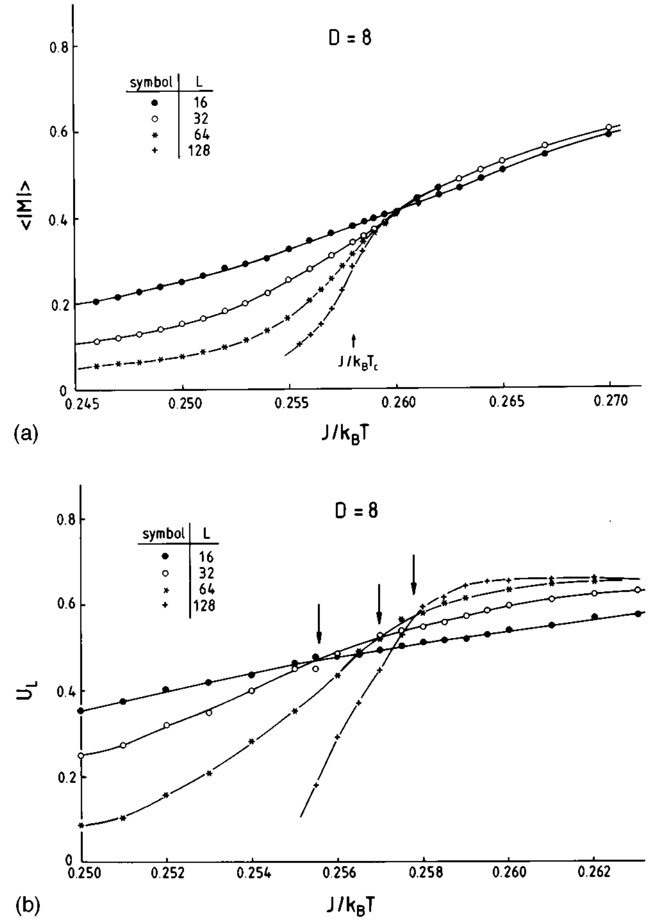

FIG. 2. Order parameter (a) and cumulant (b) plotted vs inverse temperature, for $D=8$. Four linear dimensions L are included, as indicated. The arrow in part (a) shows the final estimate for $J/k_BT_c(D)$, while arrows in part (b) show the distinct cumulant intersections between U_L and U_{2L}. Note that lengths are measured in units of the lattice spacing throughout. From Ref. [8].

Figure 2 shows typical data [8]. The temperature dependence of the order parameter has little similarity with corresponding data for the two-dimensional Ising model [49], since $\langle |M| \rangle$ at $J/k_BT_c(D)$ is rather small, and increases only slowly with decreasing temperature. Unlike either two- or three-dimensional simple Ising models which have a well-defined unique point U^* at which the cumulants $U_L(T)$ intersect at T_c [43,44], the cumulant intersections are spread out over a significant temperature range, making the precise estimation of $T_c(D)$ difficult [7–9].

In previous work the estimates [8] $J/k_BT_c(D=6)=0.2655\pm0.0002$, $J/k_BT_c(D=8)=0.2578\pm0.0002$, and $J/k_BT_c(D=12),=0.2497\pm0.0003$ were quoted. However, noting that the values of the cumulants at the crossing points for $D=12$ are far from their asymptotic two-dimensional value [50] $U^*\approx0.615\pm0.003$ [see Fig. 3 (upper part)], we now feel that the estimate for $J/k_BT_c(D=12)$ was slightly too low. Figure 3 (lower part) shows that a fit of straight lines to the data on characteristic temperatures versus L^{-1} is compatible with values slightly larger than 0.250, if data for $L=32$ are not included. In fact, the slight but systematic curvature of the plot indicates that even a value as large as 0.251 cannot be excluded. Therefore, we use

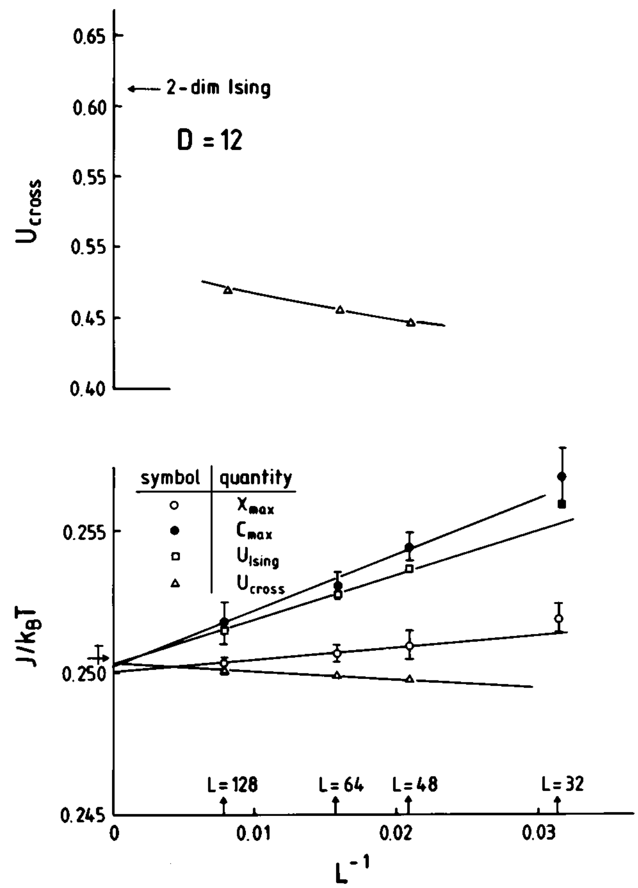

FIG. 3. Extrapolation of the cumulant crossings U_{cross} [defined from $U_{\text{cross}}=U_L(T)=U_{L'}(T)$, with L and L' being two neighboring linear dimensions) vs $1/L$ (upper part), and extrapolation of inverse temperatures $J/k_B T_{\text{cross}}(L)$, where this crossing occurs (triangles) vs $1/L$ (lower part). The inverse temperatures $J/k_BT_{\max}(L)$ of the specific heat $C_{\max}$ (full dots) and susceptibility χ' ($\chi_{\max}$, open circles) are also included. The inverse temperatures $J/k_BT_c^{\text{Ising}}(L)$ are obtained from the condition $U_L(T_c^{\text{Ising}})=U^*=0.615$ (squares). The arrow with the error bar shows the final estimate for J/k_BT_c $(D=12)$.

$J/k_BT_c(D=12)=0.2505\pm0.0005$ as our final estimate for the present analysis.

B. Finite size scaling

Given the hypothesis that the asymptotic critical behavior is that of the two-dimensional Ising model, one should have data collapsing such as in Fig. 2 according to the finite size scaling hypothesis [43,44,48,49]

$$\langle |M| \rangle L^{1/8}=\widetilde{M}(Lt), \quad U_L(T)=\widetilde{U}(Lt), \tag{42}$$

if we use only the largest values of L such that the crossover scaling variable $\mathcal{T}=\exp(\kappa D/2)L$ in Eqs. (34)–(38) is small enough, so that we are in the Ising critical regime. Note that, using the above estimates for $J/k_BT_c(D)$, there are no adjustable parameters in such a finite size scaling plot, and thus the success (or failure, respectively) of data collapsing is a significant test of whether the asymptotic Ising critical region is reached. Figures 4–6 present such data for $D=6$, 8, and

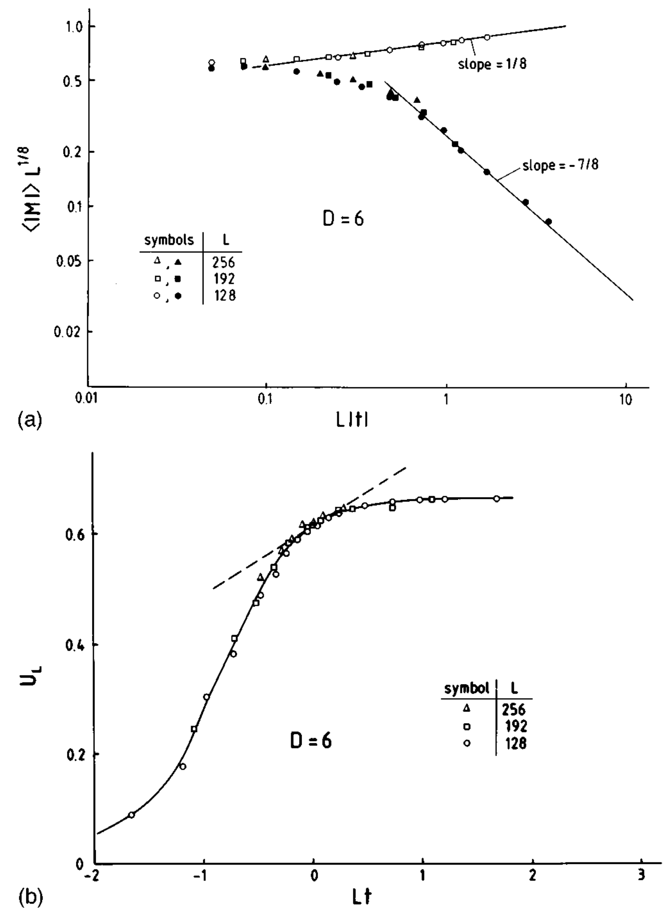

FIG. 4. (a) Log-log plot of $\langle |M| \rangle L^{1/8}$ vs $L|t|$ for $D=6$. Three choices of L are included. Data for $T<T_c(D)$ are shown by open symbols, data for $T>T_c(D)$ by full symbols. The straight lines through the data were fitted using the correct exponents for order parameter ($\beta=\frac{1}{8}$) and susceptibility ($\gamma=\frac{7}{4}$). [Recall that for $T>T_c(D)$ we have [43,48] $\langle |M| \rangle \propto \sqrt{\langle M^2 \rangle} \propto \sqrt{k_B T \chi / L^2}$ in $d=2$ dimensions, and hence $L^{1/8} \langle |M| \rangle \propto (-tL)^{-7/8}$.] (b) Cumulant $U_L(T)$ plotted vs Lt for $D=6$, including three choices of L. Broken straight line shows estimation of the slope of the scaling function at $T=T_c(D)$ (i.e., at $Lt=0$).

12, always including only the largest values of L that were studied. We see that the scaling works reasonably well for $D=6$ and 8, while for $D=12$ there is no good data collapse at all for the previous choice [8] $J/k_B T_c = 0.2497$. Even for the largest present estimate [$J/k_B T_c(D=12)=0.251$ is our present upper bound, cf. Fig. 3] there are still systematic deviations from finite size scaling. However, in view of the crossover scaling description [Sec. II D] such a difficulty was clearly expected: with the finite size crossover scaling variable being $\exp(\kappa D/2)L$, it is clear that with increasing D much larger parallel linear dimensions L are needed than for small D. However, since the problem of critical slowing down becomes much more severe for larger D, smaller values of L were actually used for the simulations for $D=12$ (Fig. 6) than for $D=6$ (Fig. 4). Thus the breakdown of finite size scaling seen in Fig. 6 for $D=12$ is certainly a consequence of our use of linear dimensions L which were too small. This difficulty prevents us from obtaining reliable estimates of critical amplitudes, unlike the case of $D=6$ and 8 (Figs. 4 and 5). We note that the scaling function $\widetilde{M}(\mathscr{T})$ for large $\mathscr{T}$ must behave as

$$\widetilde{M}(\mathscr{T} \to \infty) = \hat{B} \mathscr{T}^{\beta}, \quad \beta = \frac{1}{8}, \tag{43}$$

thus reproducing the power law $\langle |M| \rangle = \hat{B} t^{\beta}$ from Eq. (42) in the thermodynamic limit. Reading off the constant $\hat{B}$ (intercept of the straight line with slope $\frac{1}{8}$ at $Lt=1$) from the plot we find

$$\hat{B}(D=6) \cong 0.83, \quad \hat{B}(D=8) = 0.72, \tag{44}$$

while the amplitude of $\langle |M| \rangle^2 L^{1/4}$ (which is proportional to the amplitude of the susceptibility) becomes

$$\hat{\Gamma}_+^{\parallel}(D=6) \cong 0.060, \quad \hat{\Gamma}_+^{\parallel}(D=8) \cong 0.049. \tag{45}$$

While we have predicted that the critical amplitudes $\hat{B}(D)$ [Eq. (27)] and $\hat{\chi}^+(D)$ [Eq. (30)] increase exponentially with D for large D, we actually find a decrease of the amplitudes from $D=6$ to $D=8$. As will be shown in Sec. III C, this finding is *not* in contradiction with Eqs. (27) and (30), but actually due to the preexponential correction $1/D$ which is more important for $D=6$ and 8 than the factor $\exp(\kappa D/16)$ or $\exp(\kappa D/8)$ in Eqs. (27) and (30).

Noting that the finite size scaling of the cumulant U_L is a simple dependence on the ratio L/ξ,

$$U_L(T) = \widetilde{U}(L/\xi) \underset{t \to 0}{\approx} U^* + c^*(L/\hat{\xi}^+)(-t)^1 + \cdots, \tag{46}$$

where c^* is another universal constant, we should be able to extract the variation of the critical amplitude $\hat{\xi}^+$ from the slope of the cumulant at U^* in the scaling plot [Figs. 4(b) and 5(b)]. Unfortunately, the function $\widetilde{U}$ near $\mathscr{T}=Lt=0$ seems to have distinct curvature; and given the considerable scatter of our data $\hat{\xi}^+$ cannot yet be estimated with sufficient accuracy. As always, estimation of critical amplitudes from noisy simulation data is notoriously difficult.

C. Crossover scaling

An alternative test of the theory of Sec. II is possible by studying the size dependence of the magnetization $\langle |M| \rangle_c$ at the critical point $T_c(D)$ [Eq. (35)] or the cumulant [Eq. (38)] or susceptibility [Eq. (36)]. Equivalently, the maximum value $\chi^1_{\max}$ of the susceptibility χ^1 defined in Eq. (41) can be studied, and should also scale like Eq. (36); the latter quantity has the advantage that the uncertainty about $T_c(D)$ does not enter.

For this analysis, of course, estimates for the parameter κ (the inverse of the transverse interfacial length scale) are of crucial importance. This problem is addressed in Fig. 7. According to mean field theory, which neglects all types of critical fluctuations, we should simply have [5] $\kappa = \kappa_{\rm MF} = 1/\xi_b$, and the temperature dependence of ξ_b in the regime of interest is shown in Fig. 7, using Refs. [32,33]. However, the values of κ extracted from plotting the Monte Carlo results [8] for the maxima of the layer susceptibility $\chi_{\rm nn}$ at $T>T_W>T_c(D)$ are substantially smaller than ξ_b^{-1}. It

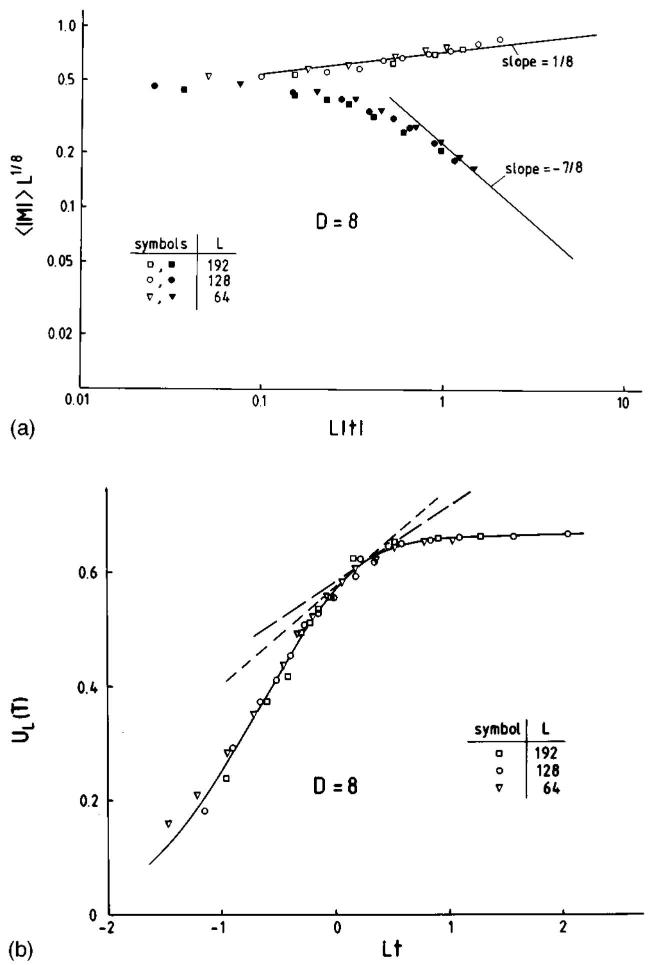

FIG. 5. Same as Fig. 4 but for $D=8$. Note that the scatter of the data points leads to a considerable uncertainty in the slope of the cumulant at $t=0$, as indicated by two possible estimates.

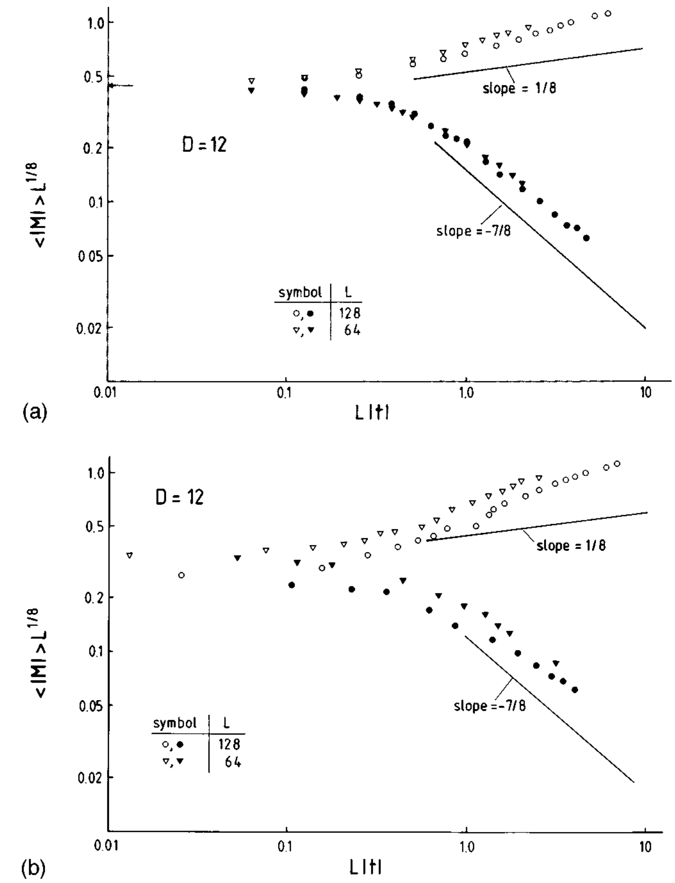

FIG. 6. Log-log plot of $\langle|M|\rangle L^{1/8}$ vs $L|t|$ for $D=12$ and two choices of L, using either $J/k_BT_c=0.2497$ (a) or $J/k_BT_c=0.2510$ (b). Straight lines indicate the slopes that the two branches of the scaling function should develop.

has been suggested [8,19–21] that capillary wave fluctuations act to renormalize the length scale in that temperature range. The direct Monte Carlo estimates for $\kappa/2$ [that were extracted from the maximum value of the layer susceptibility χ_{nn}^{max}, using [8] $\kappa/2=\ln(\chi_{nn}^{max})/D$ for large D] also are included in Fig. 7, but do not extend in the temperature regime of interest for our present analysis. Since their dependence on inverse temperature is quite smooth, a tentative estimation of κ_c at $T_c(D)$ for $D=12$, 8, and 6 could simply use a linear extrapolation, as shown by the broken line. Of course, this is not necessarily quantitatively reliable, but should give at least an order of magnitude estimation. Using this simple recipe, would yield [κ_c is the value of κ at $T_c(D)$]

$$\kappa_c/2\approx 0.364(D=12),\ \ 0.44(D=8),\ \text{and}\ \ 0.526(D=6). \tag{47}$$

These values of κ_c incorporate, albeit empirically, some capillary wave renormalization. We now assume that the crossover scaling variable $\mathscr{T}$ is still given by Eq. (24), but with κ taking into account this renormalization. The corresponding exponential factors $\exp(\kappa_c D/2)$ entering the crossover scaling variable become

$$\exp(\kappa_c D/2)\approx 78.9(D=12),\ \ 33.8(D=8),$$
$$\text{and}\ \ 23.5(D=6). \tag{48}$$

These numbers already show that our choices of L do satisfy the condition $L\gg\exp(\kappa_c D/2)$ necessary to have two-dimensional Ising critical behavior for the thinner films $D=6$ and 8, but do not for $D=12$ where the largest values of L and $\exp(\kappa_c D/2)$ are comparable. Thus, as observed, no Ising-type finite size scaling should work.

When we work out the ratio $\exp(\kappa_c D/16)D$, which describes the D dependence of the order parameter critical amplitude $\hat{B}(D)$, we find, from Eq. (47),

$$\exp(\kappa_c D/16)/D\approx 0.247(D=6),\ \ 0.194(D=8),$$
$$\text{and}\ \ 0.144(D=12). \tag{49}$$

Indeed, these amplitude factors decrease with increasing D, as observed. The same fact holds for the susceptibility amplitudes

$$\exp(\kappa_c D/8)/D\approx 0.367(D=6),\ \ 0.301(D=8),$$
$$\text{and}\ \ 0.248(D=12). \tag{50}$$

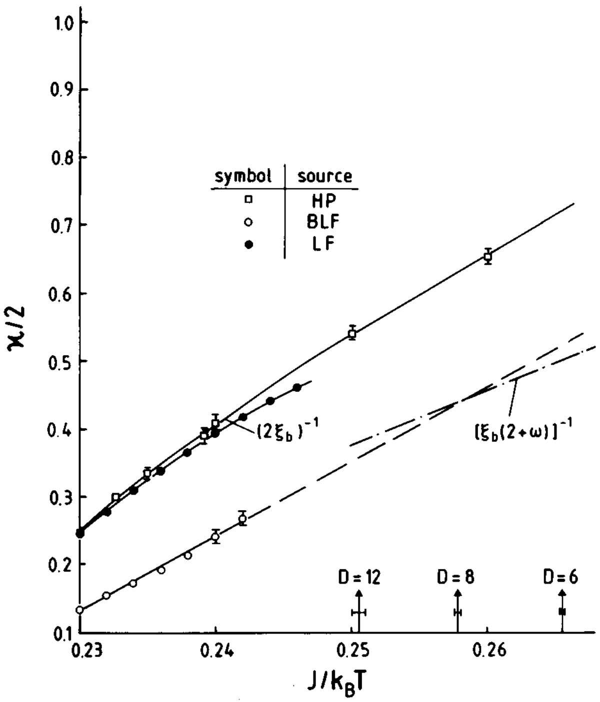

FIG. 7. Plot of the inverse length scale $\kappa/2$ vs J/k_BT. Here full dots represent estimates of $(2\xi_b)^{-1}$, ξ_b being the true correlation range in a lattice direction, obtained from the leading term of the Padé approximant to the low temperature series analysis of Liu and Fisher (LF) [32]. Open squares are corresponding Monte Carlo estimates of Hasenbusch and Pinn (HP) [33]. Open circles are the direct estimates of $\kappa/2$ from interfacial response functions, extracted by Binder, Landau, and Ferrenberg (BLF) from Monte Carlo simulations [8]. The dash-dotted curve shows the suggestion of Parry and co-workers [19–22] that $\kappa/2=[\xi_b(2+\omega)]^{-1}$, using $\omega\approx0.86$ [39] in the temperature region of interest. Arrows (with error bars) at the abscissa show location of $T_c(D)$ for $D=12$, 8, and 6, respectively.

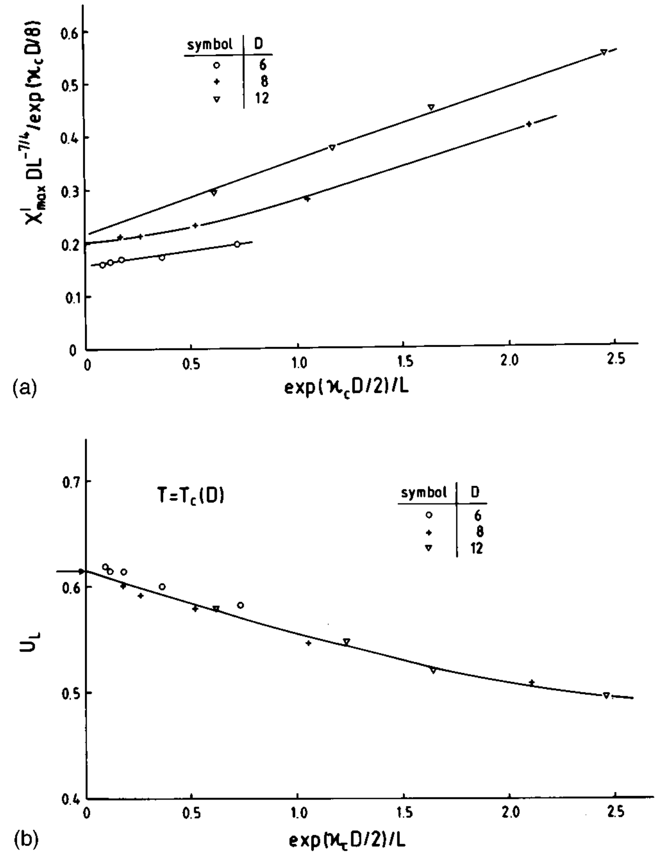

FIG. 8. Plot of the scaled susceptibility maximum $\chi^1_{\max}$ [Eq. (36)] (a) and of the cumulant [Eq. (38)] (b) vs the crossover scaling variable $\exp(\kappa_c D/2)/L$, using the estimates Eq. (48) and all values of L that are available. Curves are guides to the eye only. Arrow on ordinate in (b) shows $U^*=0.615$.

The ratios between the numbers for $D=6$ and 8 are actually compatible with the ratios of the corresponding observed amplitudes, within their numerical errors (however, we estimate that our amplitude estimates quoted above are at best accurate to 10%).

It is also interesting to test the prediction Eq. (1) suggested in Refs. [19–22]. Boulter and Parry [19,21] showed that a renormalization group treatment of the (standard) effective Hamiltonian Eq. (3) in the complete wetting regime $T>T_w$ leads to the susceptibility varying as $\exp(D/[2\xi_b(1+\omega/2)])$ rather than the mean field result $\exp[D/2\xi_b]$. They pointed out that this renormalization, Eq. (1), is insufficient to account for the Monte Carlo results [8], and introduced a two-field Hamiltonian. Renormalization of this yields a susceptibility with the same exponential form but with the factor $1+\omega/2$ replaced by $1+\omega_{\mathrm{eff}}/2$, where $\omega_{\mathrm{eff}}=\omega+\Delta\omega$. Including the additional contribution $\Delta\omega$ (>0), which arises from fluctuations at the wall coupling to those of the unbinding interfaces, improves the agreement with the Monte Carlo results [19,21]. Subsequently, Parry, Boulter, and Swain [22] argued that at lower temperatures, as $T\to T_w$, $\Delta\omega\to0$ so that for $T_w\sim T\geqslant T_c(D)$ Eq. (1) should describe the capillary wave renormalization of the transverse length scale. Figure 7 shows that the resulting numbers are numerically close to the linear extrapolation of the direct Monte Carlo estimates (see also the figure in [22]), although the increase with J/k_BT is somewhat less steep. While the resulting numbers for $\kappa_c/2$ would differ relatively little from Eq. (47),

$$\kappa_c/2\approx0.387(D=12),\ 0.444(D=8),\ \text{and}\ 0.507(D=6), \tag{51}$$

the resulting differences in the crossover scaling variable $\exp(\kappa D/2)$ seem to be significant [cf. Eq. (48)],

$$\exp(\kappa_c D/2)\approx104(D=12),\ 34.9(D=8),\ \text{and}\ 21(D=6). \tag{52}$$

Figures 8 and 9 now use the predictions Eqs. (48) and (52) in the crossover scaling description suggested in Eqs. (35)–(38). While both choices of $\exp(\kappa_c D/2)$ yield comparably good data collapse for the cumulant, we find that the data for $\chi^1_{\max}$ definitely fail to scale with the choice Eq. (48), but do scale rather well with the choice Eq. (52), resulting from Eq. (1). Given the fact that in Fig. 9 we simply use results from the literature for ω [39] and ξ_b [33], as well as the data for U_L (T_c) and $\chi^1_{\max}$ published in [8], we feel the agreement between simulation and theory is rather satisfactory, as there is no adjustment of parameters whatsoever.

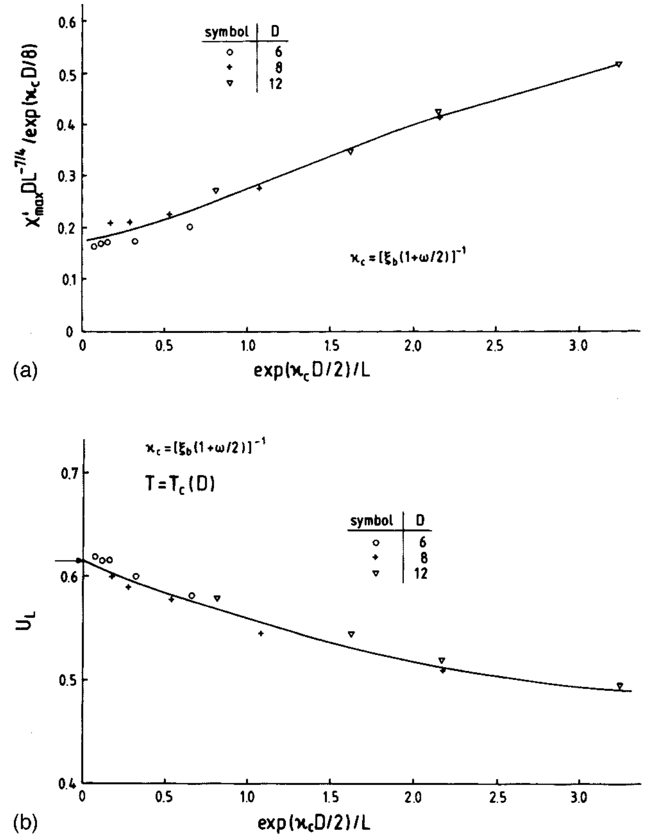

FIG. 9. Same as Fig. 8, but with κ_c estimated from Eq. (1) [as quoted in Eq. (52)].

IV. DISCUSSION

Ising thin films with competing surface fields of equal strength but opposite sign exhibit an unusual phase transition [1–9]: the bulk phase transition at T_{cb} only shows up as a very smooth formation of two ordered domains induced by the fields at the walls, separated by an interface between them. While for $T > T_c(D)$ this interface fluctuates freely and its average position is in the center of the film, for $T < T_c(D)$ this interface is localized at one of the walls, and a spontaneous magnetization of the film as a whole develops. As the film thickness $D \to \infty$, $T_c(D)$ does not converge toward T_{cb}, but rather toward the wetting transition temperature T_w of a semi-infinite system.

While general considerations about phase transitions put this interface delocalization-localization transition into the Ising universality class for $d=2$ dimensions, Monte Carlo results have shown [7–9] that the critical regime is extremely narrow and shrinks with increasing film thickness D. In fact, the Monte Carlo data [8] led to the speculation that for $D \to \infty$ the Ising regime completely disappears, but to crossover to some other universality class.

While this transition can be described as a wetting transition, when one considers surface excess properties of the system approaching $T_c(D)$ from below, this description does not answer questions about the nature of the bulk critical behavior of the thin film as a whole as one approaches $T_c(D)$ (from either side). This problem is studied in the present paper in detail. The first step elaborates the mean field theory of Parry and Evans [5], extracting the various mean field critical amplitudes. It is shown that they vary exponentially with the thickness of the film, containing factors such as $\exp(\kappa D/4)$ [cf. Eq. (10) for the order parameter amplitude in mean field]. From these mean field critical amplitudes, the Ginzburg number Gi is then constructed [Eq. (21)], which rules the self-consistency of the mean field description as well as the crossover to the Ising universality class. We show [Eq. (23)] that Gi contains a factor $\exp(-\kappa D/2)$; i.e., Gi becomes very small as $D \to \infty$, and hence mean field theory should become accurate. From a crossover scaling theory the D dependence of critical amplitudes in the Ising regime is also obtained [there are also exponential factors such as $\exp(\kappa D/16)$, cf. Eq. (27)], and consequences for finite size scaling (needed for the analysis of the Monte Carlo data) are worked out.

The Monte Carlo data allow an estimation of critical amplitudes in the Ising regime only for two neighboring small thicknesses, $D=6$ and 8, and thus these results are not very conclusive about the validity of the above theory, although at least there is also no contradiction. A more stringent test turns out to be possible through examination of finite size dependencies at $T_c(D)$, where one considers the crossover from Ising behavior [for $L \gg \exp(\kappa_c D/2)$] to mean field behavior [for $L \ll \exp(\kappa_c D/2)$]. Here all three thicknesses, $D=6$, 8, and 12, and a wide range of linear dimensions L parallel to the surfaces are included, and reasonable agreement with the crossover scaling description is obtained, provided one uses the prediction of Parry and co-workers [19–22] to calculate the appropriate transverse length scale $2/\kappa_c = \xi_b(2+\omega)$ [Eq. (1)]. Comparison between Figs. 8(a) and 9(a) shows that this test is rather sensitive, and using other plausible estimates for the temperature dependence of κ_c works considerably less well.

It is important to recognize that the crossover described here is considerably more complex than that treated in earlier work [28–31]. We are dealing with interfacial (capillary wave) as well as two-dimensional bulklike critical fluctuations. The scheme we have presented, which accounts for the Monte Carlo data, assumes that capillary wave fluctuations simply act to renormalize the transverse length scale κ^{-1}. This length is then employed in the crossover scaling variable $\mathscr{T}$ which describes the crossover to two-dimensional Ising criticality. Although the scheme is plausible (and does appear to work!) we are not aware of any formal justification for it. Trying to treat both types of fluctuations simultaneously would be very difficult.

This treatment fully explains the difficulties observed in Refs. [7–9], since it shows that for $D=12$ one is just in the middle of a crossover regime, and no simple critical behavior is to be expected. However, for films as thin as $D=20$ layers we would predict from Eq. (51) [κ_c for $D=20$ and 12 can be taken as approximately equal] that the crossover length scale $\exp(\kappa_c D/2) \approx 2300$. This implies that for all practical possible choices of L we would have $L \ll \exp(\kappa_c D/2)$, and hence a simple mean field description applies.

Of course, this finding does have interesting consequences for the interpretation of the experiment. We expect that this transition should be observable in thin films of fluid binary mixtures which undergo liquid-liquid phase separation in the bulk, if one chooses a geometry where one surface is a solid substrate while the other surface is just the interface against gas. Choosing the substrate such that it energetically prefers the species other than that preferred by the gas-fluid interface, one qualitatively realizes a situation as-modeled in the present paper. Of course, one never expects that real systems have exactly the antisymmetry property of the walls ($H_1=-H_D$) assumed here, but arguments have been provided [8] that this interface localization-delocalization transition also persists in less symmetric situations. While the formation of an interface in a thin film of a binary polymer mixture running parallel to the substrate surface has indeed already been observed (these studies were then used to estimate the compositions of the two bulk phases coexisting with each other, from the local compositions near the two walls [51]), we are not aware of an experimental observation of the transition at $T_c(D)$. Presumably the data of Ref. [51] are all taken in the regime $T_c(D)<T<T_{\rm cb}$, since in partially compatible polymer blends T_w may be far from $T_{\rm cb}$ [52]. In any case, typical experimental systems will most likely satisfy the condition $\kappa_c D \gg 1$, and then mean field behavior at $T_c(D)$ should hold.

ACKNOWLEDGMENTS

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