Interface Localization-Delocalization in a Double Wedge: A New Universality Class with Strong Fluctuations and Anisotropic Scaling

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Using Monte Carlo simulations and finite-size scaling methods we study "wetting" in Ising systems in a $L \times L \times L_y$ pore with quadratic cross section. Antisymmetric surface fields H_s act on the free $L \times L_y$ surfaces of the opposing wedges, and periodic boundary conditions are applied along the y direction. In the limit $L \to \infty$, $L_y/L^3 =$ const, the system exhibits a new type of phase transition, which is the analog of the "filling transition" that occurs in a single wedge. It is characterized by critical exponents $\alpha = 3/4$, $\beta = 0$, and $\gamma = 5/4$ for the specific heat, order parameter, and susceptibility, respectively.

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The presence of external boundaries may have profound consequences on coexisting (fluid) phases: in a semi-infinite system bounded by one wall, wetting transitions of first or second order may occur. Understanding these transitions has been a challenging problem [1–3]. If the system is a thin film, it is no longer possible to form (infinitely thick) wetting layers. Depending on the character of the boundary conditions at the walls, interesting phase transitions may occur: if both walls favor the same phase, phase coexistence is shifted relative to the value of the control parameter at which it occurs in the bulk ("capillary condensation" [3–5]). If both walls favor different phases, an interface is stabilized between the coexisting phases in the film. This interface runs parallel to the walls. It may be bound either to the left or to the right wall, and the two states coexist laterally. Upon increasing the temperature an interface localization-delocalization transition occurs at T_c to a state where the interface freely fluctuates around the center of the film and the system is laterally homogeneous. The temperature T_c depends on the strength of the surface interactions H_s and it converges towards the wetting transition temperature $T_w(H_s)$ as the film thickness L_y diverges. The critical behavior of this interface localization-delocalization [6–9] and the general aspects of the interplay between phase separation and wetting in thin films [3,10] have found abiding interest. In both cases—capillary condensation and interface localization-delocalization—the transition belongs to the two dimensional Ising universality class.

A particularly intriguing variation of these phenomena is found when one considers wetting in a wedge, where two surfaces meet under an angle $\pi - 2\alpha$. Then, a "wedge filling" transition [11–14] occurs at a temperature $T_f(H_s)$ where a planar surface ($\alpha = 0$) still would be nonwet, namely, when the contact angle of a droplet $\Theta(T) = \alpha$. Based on an approximate treatment of this problem using an effective interface Hamiltonian, Parry *et al.* [15] have proposed that this transition is generically not described by mean field theory: the filling transition in a wedge is related to the strong fluctuation regime of critical wetting, for surface potentials $W(l)$ that decay sufficiently fast with the distance l of the interface from the surface [$W(l) \sim l^{-p}$ with $p > 4$ or short range forces]. Parry *et al.* [15] also suggest that critical filling can occur even if the associated wetting transition of a planar surface is first order. This opens the way to observe critical wetting behavior—a phenomenon long sought and controversial [16–18]. Specifically, Parry and co-workers predicted the distance l_0 of the interface from the bottom of a wedge to diverge as $l_0 \sim (T_f - T)^{-\beta_s}$ with $\beta_s = 1/4$. Correlations along the wedge and in the other two directions are characterized by diverging correlation lengths $\xi_y \sim (T_f - T)^{-\nu_y}$ and $\xi_x \sim \xi_\perp \sim (T_f - T)^{-\nu_\perp}$ with exponents $\nu_y = 3/4$ and $\nu_\perp = 1/4$, respectively. Experiments [19] on complete filling provided evidence for the unusual effects induced by the wedge geometry [15], but the intriguing predictions for the strong critical fluctuations at the filling transition have not been confirmed by experiments or simulations.

We shall present a test of these predictions by Parry *et al.* [15] using Monte Carlo simulations of an Ising model in the double wedge geometry sketched in Fig. 1. We use a simple cubic lattice with an $L \times L \times L_y$ geometry. There are four free $L \times L_y$ surfaces and periodic boundary conditions are applied along the third direction. Thus, the two opposed wedges create a pore with a square cross section. We use, however, a special boundary condition in the simulations. As for the study of the wetting and interface localization-delocalization transition it is advantageous to employ antisymmetric surface fields: We choose fields $+H_s$ on two neighboring $L \times L_y$ free surfaces and $-H_s$ on the other two $L \times L_y$ surfaces. The interesting and practically relevant case of a single

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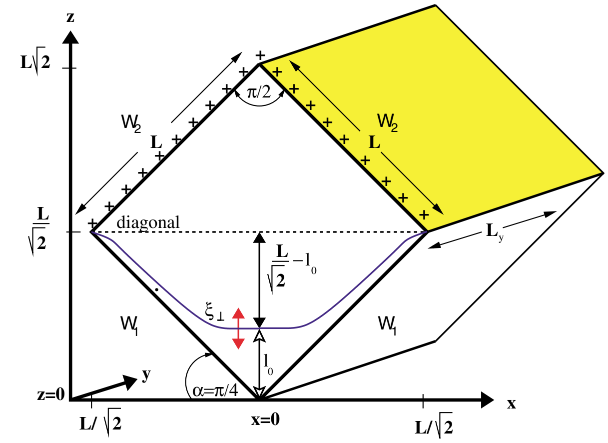

FIG. 1 (color online). Sketch of the antisymmetric double wedge Ising lattice composed of the two opposing wedges W_1 and W_2. The sign of the surface magnetic fields $\pm H_s$ along the boundaries is indicated. l_0 denotes the position of the interface from one corner.

wedge formed by two different surfaces (corresponding to the left and right corners of Fig. 1) has been studied [20], but we do not expect details of the interactions at those two corners to influence the universal behavior in the double wedge geometry.

The Ising model in this antisymmetric double wedge geometry is described by the Hamiltonian:

$$\mathcal{H} = -J \sum_{\langle i,j \rangle_{\rm bulk}} S_i S_j - J_s \sum_{\langle i,j \rangle \in W_1 \cup W_2} S_i S_j - H_s \sum_{i \in W_1} S_i + H_s \sum_{i \in W_2} S_i, \quad (1)$$

where J denotes the exchange constant of the Ising model in the bulk and the spin variables S_i can take values ± 1. The interactions in the surface planes are weakened ($J_s = J/2$) to avoid the proximity of tricritical wetting behavior [16]. Under these conditions the semi-infinite system exhibits a second order wetting transition. Rather than varying the temperature T, we bring about the filling of the wedge by increasing the strength of the surface field H_s. This has the advantage that all bulk quantities (e.g., magnetization m_b or bulk correlation length) remain constant. The temperature is fixed to $k_B T/J = 4$ (i.e., $T \approx 0.887 T_c^{\rm bulk}$). It is high enough to avoid layering [21]. We vary the linear dimensions L, L_y but we keep the ratio $L_y/L^{\nu_y/\nu_\perp} = L_y/L^3 \approx 0.0031$ approximately constant. If we measure L and L_y in units of the correlation lengths $\xi_\perp$ and ξ_y, respectively, the "generalized aspect ratio" will remain constant and finite-size rounding will set in simultaneously in all directions [22]. We shall verify *a posteriori* the ratio of the critical exponents for the correlation lengths.

We demonstrate that in this model the predictions of Parry *et al.* [15] can indeed be confirmed. Moreover, keeping the generalized aspect ratio constant we obtain a new type of "bulk" transition of the Ising model, although the filling transition in a macroscopic wedge amounts to a singularity of the surface excess free energy of the system only: At this wedge localization-delocalization transition, occurring at the critical surface field H_{sc}, the average total magnetization $m = \sum_i S_i/(L^2 L_y)$ jumps from zero [for $H_s > H_{sc}(T)$ or $T > T_f(H_s)$] to a finite value which is of the order of the spontaneous magnetization $\pm m_b$ in the bulk. This transition goes along with a divergence of the total susceptibility, and we argue that it constitutes a new universality class of second order transitions and relates the concomitant critical exponents to the critical exponents of wedge filling. Our findings exemplify the general principle that in circumstances where a broken symmetry occurs the state of the system is sensitive to boundary conditions, even if the boundaries are an infinite distance apart.

Figure 2 presents the absolute value of the average total magnetization $\langle |m| \rangle$. For small values of H_s a single wedge is not filled. In a double wedge, the interface is bound to one of the two wedges and the magnetization adopts large positive or negative values. Upon increasing the surface fields we encounter a wedge interface localization-delocalization transition at which the magnetization in a finite-size system rapidly decreases. At large values of the surface fields the interface fluctuates around the diagonal which joins the two wedges and the magnetization is small. The larger the system the steeper the decrease of the magnetization at the transition. In sharp contrast to a transition of the Ising universality class, the magnetization curves for different system sizes intersect at a rather well-defined value of the surface field

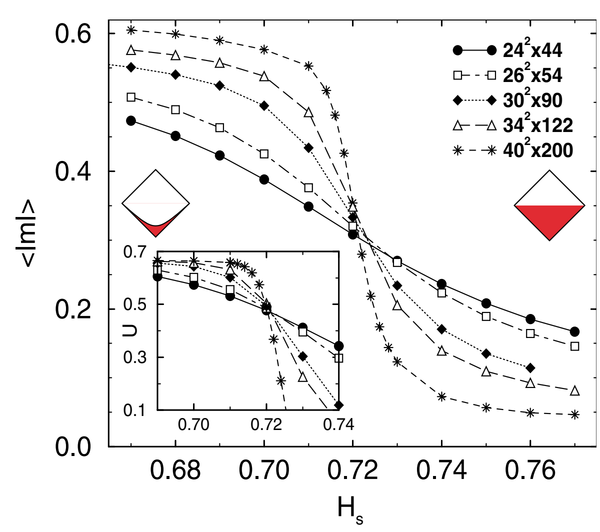

FIG. 2 (color online). Absolute value of the magnetization plotted versus surface field H_s and several choices of L and L_y, keeping the "aspect ratio" L_y/L^3 approximately constant. The inset shows the cumulant U_{L,L_y}. Typical configurations are sketched.

$H_{sc} = 0.72(1)$. In the inset we locate the surface field at which the transition occurs using the intersection of the cumulant $U_{L,L_y} = 1 - \langle m^4\rangle/[3\langle m^2\rangle^2]$. The intersection of the cumulants locates the transition at a surface field which agrees (within the statistical error) with the intersection point of the magnetization curves.

In Fig. 3 we show the fluctuation $\langle m^2\rangle - \langle|m|\rangle^2$ of the magnetization. At H_{sc} the fluctuation has a peak, which distinctly sharpens up when $L \to \infty$. The half-width Γ behaves as $\Gamma \sim L_y^{-1/\nu} = L^{-4/3}$, cf. inset of Fig. 3(a).

Phenomenologically, we can relate this unusual critical behavior to the strong fluctuation effects of the filling transition in a single wedge: The interface localization-delocalization transition in a double wedge occurs when a single wedge is filled to a height l_0 such that the fluctuations of the height δl_0 are comparable to the distance of the interface from the diagonal that divides the two wedges, i.e., $L/\sqrt{2} - l_0 \sim \delta l_0$. Parry and co-workers [15]

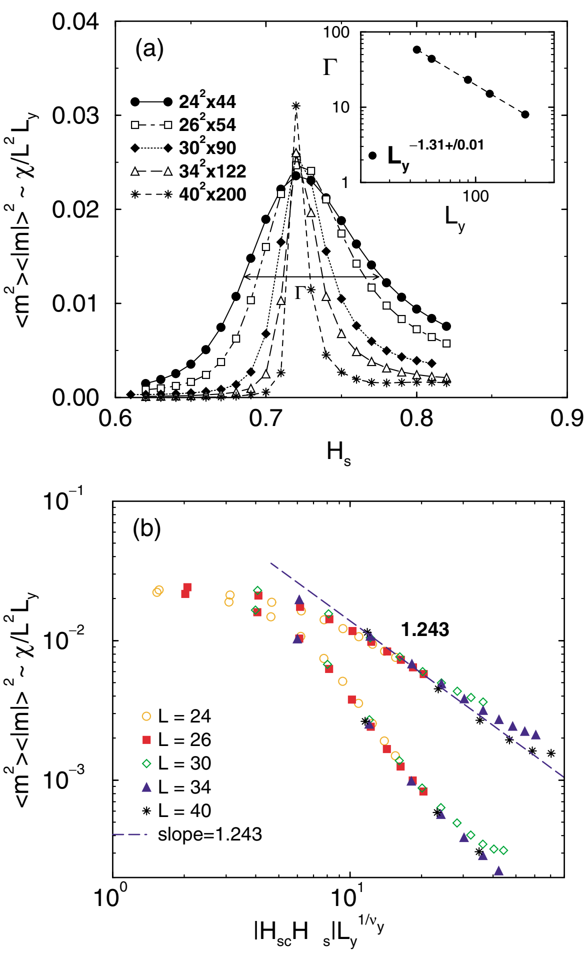

FIG. 3 (color online). (a) Magnetization fluctuation plotted versus H_s for the same choice of parameters as in Fig. 2. The inset shows a test of the power law for the half-width $\Gamma \sim L_y^{-1/\nu_y} = L_y^{-4/3}$. (b) Scaling plot of the magnetization fluctuation versus $|H_{sc} - H_s|L_y^{1/\nu_y}$. The broken line has a slope of -1.243, close to the predicted value $-\gamma = -5/4$.

predict $l_0 \sim t^{-\beta_s}$ and $\delta l_0 \sim \xi_\perp \sim t^{-\nu_\perp}$. The fact that $\nu_\perp = \beta_s$ implies that l_0 and δl_0 are of order L at the transition, i.e., neither the position nor the widths of the peaks of the distribution of the scaled distance l_0/L of the interface from one corner depend on the system size L. Assuming that magnetization fluctuations at H_{sc} are predominantly caused by fluctuations in the location of the interface, the peaks of the distribution $P(m)$ of the magnetization also do not depend on the system size L. More generally, we predict for the anisotropic scaling behavior of the probability distribution of the magnetization:

$$P(m) \sim \tilde{P}(m, L_y/\xi_y, L/\xi_\perp) \sim \tilde{P}(m, L_y t^{\nu_y}, L t^{\nu_\perp}) \sim \tilde{P}(m, L_y^{1/\nu_y} t, L_y/L^{\nu_y/\nu_\perp}), \quad (2)$$

where $\tilde{P}$ is a scaling function, $t \equiv |H_{sc} - H_s|/H_s$ denotes the reduced distance from the critical point, and amplitude prefactors of order unity are ignored. Note that for $\beta > 0$ we would have an argument $L_y^{\beta/\nu_y} m$ instead of m and a power law prefactor L_y^{β/ν_y} in Eq. (2).

At fixed generalized aspect ratio $L_y/L^{\nu_y/\nu_\perp}$, Eq. (2) implies that all moments of the magnetization exhibit a scaling behavior of the form

$$\langle|m|^k\rangle \sim \tilde{f}_k(L_y^{1/\nu_y} t) = \tilde{f}_k(L_y^{4/3} t) \quad \forall\ k, \quad (3)$$

where $\tilde{f}_k$ are scaling functions, and the same scaling behavior holds also for the cumulant U_{L,L_y}. The special case $k = 1$ explains the crossing of the magnetization curves at $t = 0$. As this intersection involves lower moments of the magnetization than the cumulant, it yields an accurate estimate of the transition point for this specific universality class. Figure 3 shows a direct verification of the scaling behavior suggested in Eq. (3) for $k = 2$.

The temperature dependence of the susceptibility χ can be obtained as follows:

$$k_B T\chi = \frac{\sum_{i,j}\langle S_i S_j\rangle}{L^2 L_y} \sim m_b^2 \xi_y \xi_x \xi_\perp \sim t^{-\nu_y - 2\nu_\perp}. \quad (4)$$

Note that spins S_j inside the volume $\xi_y\xi_x\xi_\perp \sim \xi_y\xi_\perp^2$ around spin S_i are correlated. All spins S_i of the wedge contribute equally (in a scaling sense), because the fluctuations of the interface position are on the scale of the wedge extension L itself. Thus, the critical exponent of the susceptibility is $\gamma = \nu_y + 2\nu_\perp = 5/4$. This critical exponent is straightforwardly read off from the slope of the broken line in Fig. 3(b). This value is also consistent with the scaling of the moments of the magnetization:

$$k_B T\chi = L_y L^2(\langle m^2\rangle - \langle|m|\rangle^2) \sim L_y L^2 \tilde{f}_2(L_y t^{\nu_y}, L t^{\nu_\perp}) \sim t^{-\nu_y - 2\nu_\perp} = t^{-\gamma}, \quad (5)$$

where we have chosen the powers of the anisotropic scaling variables $L_y t^{\nu_y}$ and $L t^{\nu_\perp}$ such that they cancel

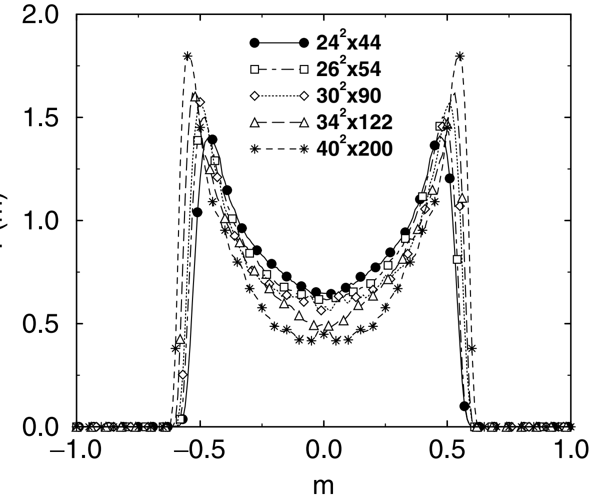

FIG. 4. Probability distribution of the magnetization at the critical field as a function of the system size.

the system size dependence. These critical exponents, $\beta = 0$ and $\gamma = 5/4$, comply with the anisotropic hyperscaling relation [23] $\gamma + 2\beta = (d = 1)\nu_\perp + \nu_y = 2 \cdot (1/4) + 3/4 = 5/4$. The thermodynamic scaling $2 - \alpha = \gamma + 2\beta$ then implies $\alpha = 3/4$ for the specific heat exponent.

A direct test of the scaling of the distribution function in Eq. (2) at the transition is presented in Fig. 4. Without any L-dependent prefactor the distributions for several system sizes approach a single master curve. Because of corrections to scaling the peaks of $P(m)$ for the small system sizes shift outwards (which would correspond formally to $\beta < 0$), but for $L_y = 200$ the peak positions have almost reached $\pm m_b$; they cannot shift much farther outwards and $P(m)$ is close to the scaling limit.

This scaling behavior strongly supports our predictions. It is important to note that the shape of the distribution depends on the generalized aspect ratio $L_y/L^{\nu_y/\nu_\perp}$; consequently the scaling behavior not only confirms the value $\beta = 0$ but also the ratio $\nu_y/\nu_\perp = 3$. Unlike second order transitions in the Ising universality class ($\beta > 0$) the peaks of the distribution at the transition do not approach each other as we increase the system size; however, the width of the peaks does not become narrower as they would at a first order transition.

In summary, we have carried out the first studies of the phase transition of Ising models in a double wedge geometry and have shown that a very unconventional critical behavior results. The critical exponents of this transition can be related to those of the filling transition with the help of a few plausible phenomenological arguments. This model seems to be the rare case where for a strongly fluctuating system in $d = 3$ dimensions the critical exponents can be predicted exactly.

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[1] J.W. Cahn, J. Chem. Phys. **66**, 3667 (1977).
[2] S. Dietrich, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, New York, 1988), Vol. XII, p. 1.
[3] K. Binder *et al.*, J. Stat. Phys. **110**, 1411 (2003).
[4] M. E. Fisher and H. Nakanishi, J. Chem. Phys. **75**, 5875 (1981).
[5] R. Evans, J. Phys. Condens. Matter **2**, 8989 (1990).
[6] A. O. Parry and R. Evans, Physica (Amsterdam) **181A**, 250 (1992); Phys. Rev. Lett. **64**, 439 (1990).
[7] K. Binder, D. P. Landau, and A. M. Ferrenberg, Phys. Rev. Lett. **74**, 298 (1995); Phys. Rev. E **51**, 2823 (1995).
[8] K. Binder *et al.*, Phys. Rev. E **53**, 5023 (1996).
[9] M. Müller and K. Binder, Phys. Rev. E **63**, 021602 (2001).
[10] M. Müller, K. Binder, and E.V. Albano, Physica (Amsterdam) **279A**, 188 (2000); Europhys. Lett. **50**, 724 (2000).
[11] P. Concus and R. Finn, Proc. Natl. Acad. Sci. U.S.A. **63**, 292 (1969).
[12] Y. Pomeau, J. Colloid Interface Sci. **113**, 5 (1986).
[13] E. H. Hauge, Phys. Rev. A **46**, 4994 (1992).
[14] K. Rejmer, S. Dietrich, and M. Napiórkowski, Phys. Rev. E **60**, 4027 (1999).
[15] A. O. Parry *et al.*, Phys. Rev. Lett. **85**, 345 (2000); C. Rascón and A. O. Parry, J. Chem. Phys. **112**, 5175 (2000); A. O. Parry *et al.* J. Phys. Condens. Matter **12**, 7671 (2000); A. O. Parry *et al.*, J. Phys. Condens. Matter **13**, 4591 (2001); A. O. Parry *et al.*, Phys. Rev. Lett. **87**, 196103 (2001); C. Rascón and A. O. Parry, Nature (London) **407**, 986 (2000).
[16] K. Binder and D. P. Landau, Phys. Rev. B **37**, 1745 (1988); K. Binder, D. P. Landau, and S. Wansleben, Phys. Rev. B **40**, 6971 (1989).
[17] D. Ross *et al.*, Phys. Rev. Lett. **87**, 176103 (2001).
[18] A. I. Posazhennikova *et al.*, J. Stat. Phys. **110**, 611 (2003).
[19] L. Bruschi, A. Carlin, and G. Mistura, Phys. Rev. Lett. **89**, 166101 (2002); J. Chem. Phys. **115**, 6200 (2001).
[20] P. Jakubczyk and M. Napiórkowski, Phys. Rev. E **66**, 041107 (2002).
[21] L. Bahmand *et al.*, Phys. Rev. E **66**, 056117 (2002).
[22] In the limit L fixed, $L_y \to \infty$ the system is quasi-one-dimensional and no phase transition occurs at finite temperature. The limits L_y fixed, $L \to \infty$ or L/L_y fixed, $L \to \infty$ correspond to quasi-two-dimensional systems, where one observes corner filling.
[23] K. Binder and J. S. Wang, J. Stat. Phys. **55**, 87 (1989).