Tests of nonuniversality and finite-size scaling for two-dimensional wetting with long-ranged forces

A. Drzewiński,[1] A. O. Parry,[2] and K. Szota[1]
[1]*Institute of Mathematics and Computer Science, Czestochowa University of Technology, ulica Dabrowskiego 73, 42-200 Czestochowa, Poland*
[2]*Department of Mathematics, Imperial College London, London SW7 2BZ, United Kingdom*
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Effective Hamiltonian models predict nonuniversal critical singularities for two-dimensional wetting transitions with marginal long-ranged forces. We test these predictions by studying interfacial delocalization transitions in an infinitely long Ising strip, of width L (lattice spacings), with external fields that are long ranged and have opposite signs at each surface. Finite-size scaling suggests that the shift of the delocalization temperature $T_c(L)$ below the (semi-infinite) wetting temperature T_w scales as L^{-1/β_s} with β_s the adsorption critical exponent. Density-matrix renormalization-group methods allow us to study the behavior of $T_c(L)$ for L up to several hundred lattice spacings. For short-ranged forces the method recovers the universal value of $\beta_s=1$ known from the exact solution. While marginal long-ranged forces strongly influence the finite-size scaling of $T_c(L)$, the extrapolated asymptotic value for the exponent β_s does not appear to confirm the predicted nonuniversality, but instead approaches the same universal value representative of systems with short-ranged forces.

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I. INTRODUCTION

While the presence of marginal forces or a marginal (upper critical) dimension has a relatively minor influence on critical singularities at bulk (order-disorder) phase transitions [1], they are believed to dramatically influence fluctuation effects at interfacial unbinding (wetting) transitions in both two [2–4] and three dimensions [5–12]. These predictions arise from analysis of simple interfacial Hamiltonian models, which are supposed to describe the relevant physics at length scales much greater than the bulk correlation length. It is, of course, crucially important to test these ideas against more microscopic theories for which the Ising model is often ideally suited. Indeed, the nature of wetting in the three-dimensional (3D) Ising model with short-ranged surface forces has long been the subject of controversy and debate in the literature. In the present paper, we use density-matrix renormalization-group (DMRG) methods [13–17] to numerically investigate predictions of nonuniversality, and also finite-size scaling [18–20], occurring at critical wetting transitions in a (bulk) two-dimensional (2D) Ising model but with long-ranged surface interactions. The DMRG generates essentially exact numerical results for thermodynamic quantities, magnetization, and energy-density profiles and their correlations, as defined for infinitely long strips with widths up to several hundred lattice spacings. The accuracy is considerably better than that attainable using Monte Carlo simulation in this dimension and for striplike geometries, and allows us to test the aforementioned predictions of the mesoscopic, interfacial approaches. As we shall show, there is clear evidence from our numerics that the finite-size scaling shift of $T_c(L)$, at fixed L, is sensitive to the presence of marginal forces. These certainly have a more pronounced influence on the shift compared to irrelevant long-ranged forces (which decay with a faster power law). However, much to our surprise, the extrapolated, $L\to\infty$, values for the exponent β_s (for different amplitudes of the marginal interaction) all appear to converge to the same universal value $\beta_s=1$ and do not take the anticipated nonuniversal values. Some reasons for this discrepancy are discussed, and possible further lines of inquiry are suggested.

II. INTERFACIAL HAMILTONIAN PREDICTIONS

We begin by reviewing some necessary background material. For systems with purely short-ranged intermolecular and surface interactions the 2D critical wetting transition is very well understood. Abraham's [21,22] celebrated exact solution of the square lattice 2D Ising model with a contact surface field (for both [10,11] lattice orientations) shows universal critical behavior for the pertinent diverging length scales. It is important that the values of the corresponding critical exponents are in perfect agreement with the predictions of effective interfacial Hamiltonian studies [4,23–26] and also rather general random-walk arguments [27,28]. These allow one to classify the critical behaviors into a number of scaling regimes dependent on the range of the intermolecular forces, and also assess the role of interfacial fluctuations arising from impurity-induced disorder [4,29,30].

The nature of the critical wetting transition in 3D systems with short-ranged forces has proven to be much less transparent. Here progress is considerably more difficult for two reasons. First, in the absence of an exact solution for a truly microscopic model, analytical approaches are essentially limited to the study of interfacial Hamiltonians. Second, for systems with short-ranged forces, 3D is marginal for critical wetting and the predicted critical behavior is highly sensitive to the structure of the interfacial Hamiltonian. The simplest interfacial model is a local Hamiltonian characterized by a binding potential function $W(\ell)$, with $\ell(\mathbf{x})$ the collective coordinate representing the local interfacial thickness, and a stiffness coefficient Σ [4–6]. RG analysis of this model predict strongly nonuniversal critical singularities. As is well documented, however, these predictions are not supported by extensive Monte Carlo simulation studies of critical wetting in the Ising model, which instead are broadly consistent with the singularities (and phase diagram) of mean-field theory [7,8]. Tweaking the interfacial model to allow for a necessary position-dependent stiffness coefficient $\Sigma(\ell)$ [9,10] worsens the comparison with the Ising simulations, since the interfacial model predicts fluctuation-induced first-order (and second-order) transitions which would effectively alter the structure of the surface phase diagram altogether, an effect not observed in the simulations. It seems likely that this potential pluralistic situation and also the original discrepancy with the Ising model simulations are resolved by allowing for the full nonlocal structure of the interfacial Hamiltonian, which does not exhibit stiffness instabilities [11,12,31]. Nonlocal effects restore the original topology of the surface diagram, consistent with the scaling theory of Nakanishi and Fisher [32], but also dramatically reduce the size of the asymptotic critical regime in line with the Ising model simulation results.

Given the aforementioned subtleties concerning the nature of marginal critical wetting transitions in 3D, it is natural to enquire whether there are also issues concerning the predictions of marginal behavior in 2D due to the presence of long-ranged forces. The universality classes and fluctuation regimes for 2D critical wetting were studied in depth during the 1980s in a series of beautiful articles by Lipowsky and co-workers based largely on analysis of the simple interfacial Hamiltonian

$$H[\ell] = \int dx \left[\frac{\Sigma}{2} \left(\frac{d\ell}{dx} \right)^2 + W(\ell) \right]. \tag{1}$$

Here $\ell(x) > 0$ represents the local height of the unbinding fluid interface which separates a bulk fluid (β, say) from a coexisting phase (α) that intrudes between the bulk and wall. The wetting transition refers to the unbinding of the equilibrium interface position from the wall as the temperature or strength of the binding potential is varied. The interfacial model can be studied exactly using transfer-matrix methods, which, in the limit of infinite momentum cutoff, reduce the eigenvalue problem to a Schrödinger-like equation from which a complete description of the allowed critical behavior emerges. For binding potentials of the type $W(\ell) \sim -A\ell^{-p} + B\ell^{-q}$ containing long-ranged attractive and repulsive tails, there are three scaling regimes [3], depending on whether the exponents p, q are greater than or less than a marginal value $\tau = 2$. The three regimes were classified as the strong-fluctuation (SFL), weak-fluctuation (WFL), and mean-field regimes, respectively, with the SFL regime representing the universality classes of wetting with short-ranged forces that correspond to $q > p > 2$. The borderline between the regimes involves either the attractive or repulsive part of $W(\ell)$ being a marginal interaction (in the RG sense). The most subtle case is the intermediate-fluctuation (IFL) case, borderline between the SFL and WFL regimes, in which $W(\ell)$ has a short-ranged contribution and a long-ranged tail $W(\ell) \sim \ell^{-2}$. This was studied in detail by Lipowsky and Nieuwenhuizen [2] using the binding potential

$$W(\ell) = -u[1 - \Theta(\ell - R)] - \frac{w}{\ell^2}\Theta(\ell - R), \tag{2}$$

together with a hard wall repulsion at the origin. Here $\Theta(x)$ is the usual Heaviside step function with $\Theta(x>0)=1$, $\Theta(x<0)=0$. The surface phase diagram is described by the separatrix $u_c(w)$, which distinguishes the bound and unbound interfacial regimes corresponding to partial and complete wetting, respectively. The possible critical behavior in this IFL regime is extraordinarily rich and exhibits three further subregimes labeled A, B, and C, all of which show peculiar features. Regime A describes wetting transitions with essential singularities, while in regime C different moments of the interfacial height distribution function show discontinuous (first-order-like) and continuous behavior. In regime B the exponents are nonuniversal, depending on the value of the long-ranged interaction. For example, the divergence of the mean interfacial height $\langle \ell \rangle \sim (u-u_c)^{-\beta_s}$ is characterized by the critical exponent

$$\frac{1}{\beta_s} = \sqrt{1 - 8\Sigma w}, \tag{3}$$

where we have absorbed factors of k_BT into the stiffness and the Hamaker constant. Note that, for purely short-ranged forces ($w=0$), this reduces to the universal result $\beta_s^{SFL}=1$ representative of the SFL regime supported by the exact Ising model calculations of Abraham. In general, within subregime B we have $\frac{1}{2} < \beta_s < \infty$.

III. MICROSCOPIC MODEL AND FINITE-SIZE SCALING

We now turn to the microscopic model used to investigate the IFL regime and the methodology used to extract the critical exponents. First, microscopic studies of long-ranged critical wetting are somewhat easier in 2D than in 3D. In 3D it would be necessary to have both long-ranged fluid-fluid and substrate-fluid interactions in order to induce the requisite vanishing of Hamaker constant A (and also leave $B>0$) [33]. This fine tuning is one of the reasons that critical wetting is so rare in 3D systems. In 2D, however, this sensitive balancing of surface- and fluid-fluid forces is not required, since the wetting phase boundary occurs at a nonzero value of the Hamaker constant A. Following the usual (sharp-kink) method [33] for constructing binding potentials, the potential (2) pertinent to the IFL regime can be induced in a model that has purely short-ranged fluid-fluid interactions but with surface forces that have a long-ranged tail. This means that a semi-infinite Ising model with the usual nearest-neighbor spin-spin interactions but with a one-body external potential that contains (a) a contact surface field h_1 at the "wall" and (b) a long-ranged tail decaying as $h_{LR}l^{-3}$, with l the distance from the surface, should generate the required marginal interactions.

To extract a value for the critical exponent β_s, we rely on finite-size scaling methods [18,19], which are known to work well for systems with short-ranged forces in 2D and 3D [35,36]. Consider a 3D Ising capillary slit or 2D Ising strip with surfaces that preferentially adsorb different bulk phases.

That is, the external potentials arising from each wall are of opposite signs. Further, we suppose each semi-infinite surface exhibits a critical wetting transition at temperature (or, equivalently, surface field strength) T_{wet}. In the confined geometry phase coexistence (or pseudo-phase-coexistence) between net up-spin and down-spin phases is restricted to temperatures $T < T_c(L)$. Rather general finite-size scaling arguments suggest that the capillary-critical temperature (or delocalization point) is shifted below the semi-infinite wetting temperature by an amount [18]

$$T_{wet} - T_c(L) \approx L^{-1/\beta_s}. \tag{4}$$

Alternatively, at fixed T the same scaling holds for the shifted surface field $h_1(L)$ below its wetting value (see below). As mentioned above, the finite-size shift of the delocalization temperature (and the associated behavior of the transverse correlation length) has been well studied in 2D and 3D for short-ranged forces and allows one to identify the critical wetting exponent β_s as well as further information concerning critical amplitudes [34,35]. In the IFL regime we anticipate that this type of scaling should work well within subregime B, where both the film thickness and roughness are similar. The multiscaling and pseudo-first-order behavior of subregime C is potentially more complicated. Recall that in higher dimensions first-order wetting does not lead to the same finite-size behavior for the confined interface. Similarly, in subregime A, where there is an essential singularity ($\beta_s = \infty$), it is unlikely that one would be able to extract the (presumably) logarithmic dependence on L for the finite-size shift of $T_c(L)$. Thus, the question we concentrate on is the following: Does the finite-size shift of $T_c(L)$ conform to the finite-size law (4) with a nonuniversal exponent characteristic of the IFL regime?

In 2D strips the temperature $T_c(L)$ is the point at which pseudo-phase-coexistence, associated with the asymptotic degeneracy of the transfer-matrix spectrum, ends [19,36,37]. Thus for $T < T_c(L)$ the correlation length $\xi_\parallel$ (measured along the strip) is exponentially large, $\xi_\parallel \sim e^{\sigma(\sin\theta)L}$, where σ is the (reduced) surface tension and θ the contact angle. In contrast, for $T > T_c(L)$ (but below the bulk critical temperature T_c^{bulk}), we have $\xi_\parallel \approx L^2$. The latter is characteristic of the soft-mode fluctuations of an up-spin–down-spin interface or domain wall that wanders freely between the confining walls. The distinct high- and low-temperature phases are depicted schematically in Fig. 1. While in 2D the pseudo-capillary-critical point is not associated with the divergence of any thermodynamic quantities, its location can be reliably determined using a number of criteria. Within the subregime B, which is the main topic of our study, these all yield similar results. For example, one could define $T_c(L)$ as the temperature at which the gradient of the magnetization at the midpoint is maximal. Alternatively, one can calculate the temperature at which the susceptibility is maximal, which gives similar results to the first method. The approach we have found most straightforward to adopt, however, is one involving the calculation of the two-point spin-spin correlation function $c_{L/2} = \langle \sigma_{i,L/2}\sigma_{i,L/2+1}\rangle$, where $\sigma_{i,j}$ is the usual Ising spin variable defined on a square lattice. This quantity is independent of the index i denoting the position along the strip, and measures the correlation between a nearest-neighbor pair centered at the middle and directed across the strip. In the pseudocoexistence (partial wetting) regime $c_{L/2}$ is large and positive since the two spins tend to align. On the other hand, if an interface forms at the center of the strip the spins tend to have opposite sign and $c_{L/2}$ is negative. We identify $T_c(L)$ as the maximum of the derivative $\partial c_{L/2}/\partial h_1$ at fixed T, L, and h_{LR}.

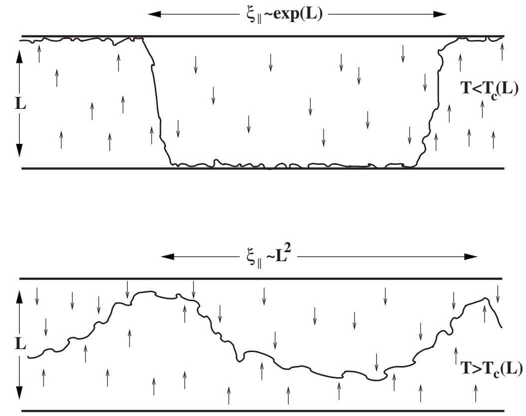

FIG. 1. Schematic illustration of low- $[T < T_c(L)]$ and high-temperature $[T > T_c(L)]$ phases in a strip with opposing wall fields. At low temperatures there is pseudo-phase-coexistence in which the interface is bound to a wall over a distance exponentially large in the strip width. Above the finite-size delocalization temperature, the interface wanders freely between the two walls with a correlation length $\xi_\parallel \propto L^2$, representing a soft mode.

Our model Hamiltonian for the infinitely long strip is written

$$H = -J\left(\sum \sigma_{\kappa,\ell}\sigma_{\kappa',\ell'} - h_1\sum_{\kappa}\sigma_{\kappa,1} + h_1\sum_{\kappa}\sigma_{\kappa,L} + \sum_{\ell=2}^{L-1} V_{ext}(\ell,L)\sum_{\kappa}\sigma_{\kappa,\ell}\right), \tag{5}$$

where the first sum is over all nearest-neighbor pairs, and we have measured the surface field h_1 and external potential V_{ext} in units of J. The walls are located in the $\ell = 1$ and $\ell = L$ lines, which are the source of the surface fields h_1 and the long-ranged potential $V_{ext}(\ell, L)$. The latter is assumed to arise from the sum of the two independent wall contributions

$$V_{ext}(\ell,L) = h_{LR}\left(\frac{1}{\ell^r} - \frac{1}{(L+1-\ell)^r}\right), \tag{6}$$

where $r=3$ is chosen to generate a marginal long-ranged interaction in the binding potential. The contact surface field h_1 is related to the (inverse) of the short-ranged contribution to the binding potential u. The precise relation between these two parameters is not important. The relationship between the parameter h_{LR} appearing in the external field and w appearing in the binding potential has been studied by Dietrich and Napiorkowski [33] in the context of density functional theory. For the present Ising system and with $r=3$, the bind-

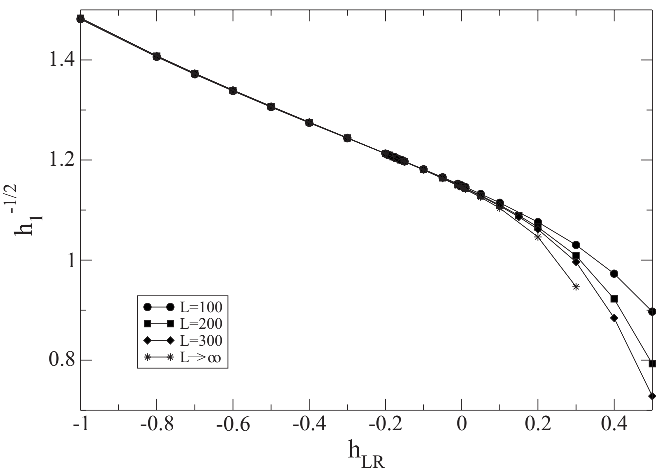

FIG. 2. Numerically determined phase diagram for the marginal case ($r=3$), showing the line of pseudocritical points separating the pseudo-phase-coexisting (localized) and soft-mode (delocalized) interfacial states, for various strip widths L. The finite-size shift of the delocalization temperature is much smaller for negative h_{LR}, suggesting that the asymptotic finite-size scaling law (4) is more reliable in this region.

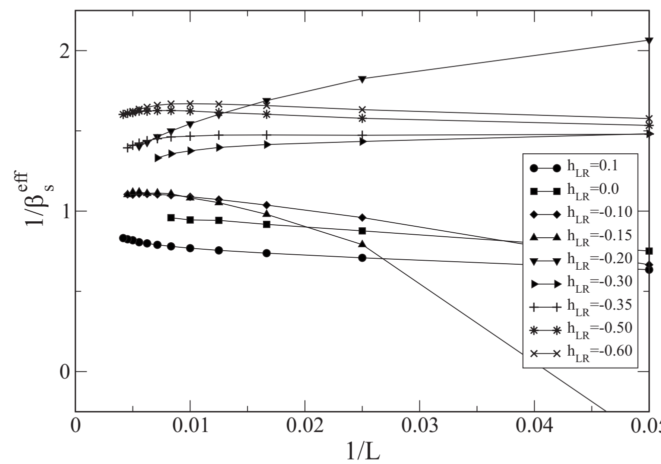

FIG. 3. Numerically determined value of the effective critical exponent as a function of the inverse strip widths $1/L$ for various values of the long-ranged field h_{LR}.

ing potential decays as the inverse square of the distance from the wall, with strength $w=m_0Kh_{LR}$, where m_0 is the spontaneous magnetization and $K=J/k_BT$. The values for m_0 and Σ are of course known for the Ising model, so one can trivially translate the effective Hamiltonian result (3) into the pertinent Ising expression. In particular, we can identify

$$8\Sigma w = 4Kh_{LR}(1-\sinh^4 2K)^{1/8} \sinh 2\left(K - \frac{1}{2}\ln \coth K\right), \tag{7}$$

from which we can readily determine the predicted numerical value for the microscopic model.

IV. NUMERICAL RESULTS AND DISCUSSION

To numerically determine the shifted delocalization temperature $T_c(L)$ in the finite-size strip, we use the DMRG method. Despite its name, the method has only some analogies with the traditional RG, being essentially a numerical, iterative basis, truncation method. It was originally proposed by White [13] as a new tool for diagonalization of quantum spin chains and was later adapted to classical 2D equilibrium statistical mechanics by Nishino [14]. The DMRG allows one to study much larger systems (up to $L=220$ in the present paper) than is possible with standard exact diagonalization methods (typically $L\approx 50$) and provides data with remarkable accuracy. Comparison with exact results for the case of vanishing bulk and contact boundary fields show that this gives very accurate results in a wide range of temperatures [17].

Some representative numerical results are shown in Figs. 2–4. Figures 2 and 3 refer to the marginal case $r=3$ corresponding to the IFL regime in the effective Hamiltonian description. Figure 2 shows the section of the phase diagram separating the localized and delocalized interfacial regimes, obtained from measuring the maximum in the slope of the $c_{L/2}$ correlation function, for different strip widths. The spin-spin coupling strength is fixed at $K=2/3$ corresponding to a temperature far below the bulk critical point $K_c=0.44$. The spontaneous magnetization m_0 is very close to unity at this temperature and the bulk correlation length ξ_b is of order a lattice spacing. This is the region where we anticipate that the effective Hamiltonian accurately describes the interfacial physics at length scales much bigger than ξ_b. It is clear that the finite-size effects for the shift of the surface field at the delocalization point are much more pronounced, even for these quite large strips, in the positive h_{LR} region, i.e., as one moves toward the predicted subregime A. This is indeed to be anticipated qualitatively, because the exponent determining the shift, $1/\beta_s$, is vanishing. It does mean, however, that it is harder to determine the exponent quantitatively. The data for negative h_{LR}, on the other hand, appear to be converging to the appropriate semi-infinite phase diagram considerably faster. The use of the asymptotic finite-size scaling law (4) is therefore more reliable in this regime.

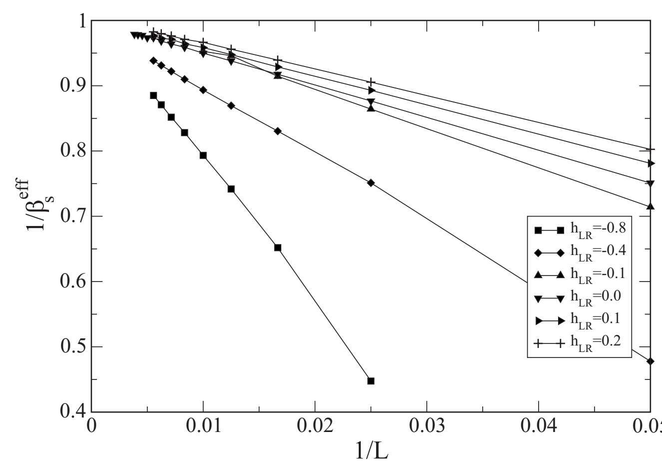

FIG. 4. As for Fig. 3 but for shorter-ranged algebraically decaying forces with $r=4$. The effective exponent can be seen to converge to the universal value $\beta_s=1$ representative of the SFL regime.

Figure 3 shows the numerical results obtained for the critical exponent $1/\beta_s$ using the finite-size scaling law (4) as applied to the surface field $h_1(L)$. The effective exponent $1/\beta_s^{eff}$ is defined using the usual logarithmic derivative and should converge to the asymptotic results as $L\to\infty$. For short-ranged forces $h_{LR}=0$, the exponent can indeed be seen to converge to the exact value $\beta_s=1$ as L increases. As discussed earlier this exponent is representative of the SFL regime. Therefore, there are no problems related to the size of the asymptotic critical regime, which have made observations of fluctuation effects for 3D wetting so difficult. This gives us some confidence that, at least for small negative values of h_{LR}, where the finite-size effects appear to be less extreme, the present method should be able to determine the presence of any nonuniversality. Our numerical results show some quite unexpected features. Let us begin with a point of agreement with theoretical predictions. For negative values of h_{LR} and widths up to $L=100$ lattice spacings, there appears no doubt that the effective exponent $\beta_s^{eff}(L)$ is strongly influenced by the presence of the marginal long-ranged interactions. Indeed, for any fixed value of L up to $L=100$, the value of $\beta_s^{eff}(L)$ for each amplitude h_{LR} is in reasonable agreement with the expected theoretical, nonuniversal value for β_s. This (partial) agreement breaks down when $1/\beta_s \approx 2$, where there is more discrepancy. This itself is also (partially) consistent with the effective Hamiltonian predictions, since this region is close to the crossover to the anomalous subregime C, where the finite-size scaling is certain to be more involved. We may also remark that, for widths up to $L=100$, the graph of $1/\beta_s^{eff}(L)$ vs L is quite flat. If one had access only to these slit widths one might reasonably conclude that the effective Hamiltonian predictions of nonuniversality are well founded. However, it is clear that, at larger values of L, the graphs of $1/\beta_s^{eff}$ each show crossover behavior, such that the asymptotic, extrapolated value is close to unity, regardless of the strength of the interaction h_{LR}. This is obviously quite at odds with the theoretical expectations and was a surprising finding for the authors. We know of no theoretical justification for this behavior.

Several possible explanations that do not invalidate the nonuniversality of β_s are as follows.

(a) Numerical errors associated with the DMRG. While these cannot be completely ruled out they appear very unlikely. There are two reasons for this. First (as shown in Fig. 4) the DMRG shows excellent agreement with theoretical predictions of universal critical behavior for the case of irrelevant long-ranged forces. The numerical results for the largest system sizes show no unexpected behavior. If there were problems with the DMRG they would also have shown for this system. Second, we have made extensive investigations of the number of states kept in the numerical iterations and find no evidence that any problem arises as the system size increases. All indications are that our numerics remain highly accurate even for the largest values of L. Of course, there is always numerical uncertainty in extracting asymptotic exponents from the extrapolation of data pertaining to finite-size systems, but there appears no systematic trend to suggest that errors incurred in this way are the reason behind the mismatch with theory.

(b) There are nonsingular $1/L$ corrections to the finite-size scaling law (4) associated with the lattice nature of the Ising model which are absent in the continuum limit. Recall that in 2D there is only a pseudocritical point at $T_c(L)$, so that the definition of the critical point is open to question. One possible test of this would be to study the solid-on-solid limit of the Ising model, equivalent to a discrete version of the continuum model. One would be able to access far larger systems sizes L in this case.

Our final figure (Fig. 4) is a plot of the effective exponent versus strip widths for faster decaying long-ranged forces corresponding to $r=4$. This serves as a check on application of the DMRG for long-ranged systems that are away from marginality. According to the general RG of wetting the surface force is now irrelevant and critical exponent β_s should belong to the SFL regime universality class. The data are completely consistent with this anticipated behavior. Regardless of the sign of the surface forces, the effective exponent rapidly converges to the universal value as $L\to\infty$. It is noticeable that the convergence to the expected, asymptotic, universal value, is far more clear cut than that for the troublesome case of marginal forces shown in Fig. 3.

In summary we have investigated predictions of nonuniversality for 2D critical wetting in an Ising model with marginal long-ranged interactions. A numerical DMRG study only partially supports the predictions of effective Hamiltonian theory. Specifically for slit widths up to $L=100$, the finite-size scaling shift of the delocalization temperature $T_c(L)$ is described by an effective exponent that appears to be weakly dependent on L and strongly dependent on the amplitude of the interaction h_{LR}. However, the data from larger slit widths appear to show that the extrapolated asymptotic values for the critical exponents all appear to converge toward a universal value (of unity) at odds with theoretical expectations. Further studies are required to ascertain the reasons for this discrepancy and clarify the status of effective Hamiltonian theories of wetting with marginal forces in 2D.

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