PHYSICAL REVIEW B VOLUME 48, NUMBER 5 1 AUGUST 1993-I

Phase transitions in a model porous medium

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(Received 19 January 1993; revised manuscript received 26 February 1993)

An exact solution is presented for Ising-like transitions in a decorated lattice model of a porous medium. The model is solved by decimation of the spins, leading to a space-filling lattice with renormalized parameters. The critical temperature is found to vary as $1/\ln L$, where L is the number of sites between intersections of the spin chains. Some of the critical exponents differ from those of the ordinary Ising problem. We have also studied the case of a single, infinitely long pore, using both exact and approximate methods. An exploration of finite-width effects reveals surprisingly small (quantitative) deviations from mean-field theory.

The behavior of fluids confined in porous media has received much current attention[1] and yet several phenomena have eluded even qualitative understanding. For example, the liquid-vapor transition of helium in aerogel[2] surprisingly exhibits critical exponents quite different from the random-field Ising model,[3] while the onset of superfluidity shows a marked lack of universality.[4] Many different theoretical models have been proposed to explain certain aspects of the observed results; some emphasize single, regular, one-dimensional (1D) pores and the related wetting phenomena of the fluid in the confined system,[5] while others invoke randomness in the substrate geometry to explain the global behavior of the fluid on long length scales.[6]

It is clear that connectivity between different pore-space regions is required for the fluid to have a phase transition. However, such a geometrical constraint may not be sufficient to ensure and characterize singular behavior of the confined fluid, as the wetting properties of the substrate may also play an important role. In this paper we investigate the combined effects of confinement, connectivity and adsorption, all present in porous media. We shall first consider a simple model system having these attributes which yields interesting exact results: an interesting form of finite-size scaling, a phase diagram topology reminiscent of the random-field Ising model, and some critical exponents which differ from those of the pure Ising system. We then investigate in more detail a model involving a single isolated (but infinitely long) pore; this exhibits a rich phase diagram for intrapore phenomena, such as wetting and capillary condensation.[5]

Our models use the lattice-gas approach commonly applied to address phase transition phenomena. We consider first an Ising model on a decorated hypercubic lattice[7,8] with Hamiltonian

$$H=-J\sum_{\langle i,j\rangle}S_iS_j-h\sum_{i\in O}S_i-(h+h_1)\sum_{i\in X}S_i\ , \tag{1}$$

where the spins interact with nearest neighbors ferromagnetically with energy J. A portion of the lattice is shown in Fig. 1. There are two types of sites: node sites (denoted by O) which experience a field h, and pore sites (denoted by X) which experience a field $(h+h_1)$. The difference is due to the (assumed attractive) adsorption potential experienced by the pore sites; the value of h is determined by the chemical potential, or pressure, of the coexisting vapor external to the system. The lattice is characterized by a single length scale, L, equal to the number of pore sites between nodes. $L=0$ is the ordinary Ising model.

The partition function may be evaluated in two steps; the first is a partial trace over the intermediate pore sites. As these sites constitute isolated chains of spins connected only by node sites, this partial trace can be performed exactly. The result is an effective Hamiltonian for the remaining node-site spins:

$$-\frac{H_{\mathrm{eff}}}{T}=G+K'\sum_{\langle i,j\rangle;i,j\in O}S_iS_j+H'\sum_{i\in O}S_i\ . \tag{2}$$

Second, we need to evaluate the trace over these remaining node sites. Equation (2) is a uniform, space-filling Ising Hamiltonian. The new parameters K' and H' are explicit, nonsingular functions of the original variables J, T, h, and h_1 and of the pore length L. Thus we know that the only singularities of the free energy occur at singular points of the ordinary, space-filling Ising model. Since the

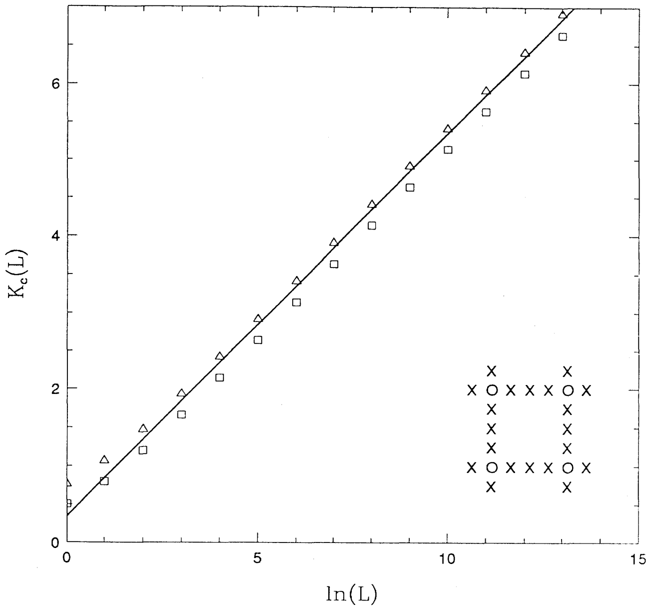

FIG. 1. Lower right inset depicts a fragment of the model lattice in two dimensions for $L=3$. Triangles (squares) represent $K_c=J/T_c$ as a function of L for $d=2$ (3). The straight line is defined by Eq. (7) the point at which the 1D correlation length $=L$.

latter has been much studied (and even solved exactly in two dimensions), our original problem can now be considered well understood by this mapping. For example, criticality will occur in the original decorated Ising lattice when the effective coupling K' and field H' satisfy the conditions for criticality in the regular Ising lattice. Letting the value of K' at the regular Ising critical point to be $K_c(0)$, the conditions for criticality in the decorated lattice become

$$K'(J,T,h,h_1,L)=K_c(0) , \tag{3}$$

$$H'(J,T,h,h_1,L)=0 . \tag{4}$$

These equations determine the critical points of our problem implicitly.

We consider first the case of no pore-site field, $h_1=0$. By symmetry, the critical point must occur at $h=0$. In this case, the partial trace over pore sites generates a simple, analytic relation between the original coupling in the decorated lattice, $K=J/T$, and the effective coupling K'. For pores of length L,

$$\tanh K'=[\tanh K]^{L+1} . \tag{5}$$

Thus, the condition for criticality in the decorated lattice, Eq. (3), becomes

$$K_c(L)=\tanh^{-1}[(\tanh K_c(0))^{1/(L+1)}] , \tag{6}$$

where $K_c(L)$ is the critical coupling of the decorated lattice with pore length L. The behavior of this critical coupling constant as a function of $\ln L$ is shown in Fig. 1. Note that the reduction of the corresponding critical temperature $T_c(L)=J/K_c(L)$ relative to the value for the space-filling lattice $T_c(0)$ is quite dramatic: For $L=1$ the ratio of these temperatures is 0.438 (0.576) in three dimensions (two dimensions). As L diverges, the 2D and 3D values of $T_c(L)$ decrease, but have the same asymptotic dependence on L. The explanation of this interesting behavior is the following. A line of spins has no ordering transition (except at zero temperature). At a temperature T_L, the 1D correlation length $\xi\sim\frac{1}{2}e^{2J/T}$ becomes equal to the spacing $\sim L$ between nodes. The interconnections then permit an ordering transition. This heuristic argument leads to

$$T_c(L)\sim T_L\sim 2J/\ln(2L) . \tag{7}$$

This result is independent of dimensionality and agrees asymptotically with the exact data in Fig. 1.

The preceding may be extended to the case of interconnected pores of finite cross section $L_\perp$. As each individual pore has a cross-sectional area $L_\perp^{d-1}$, one would expect correlated regions along the pore of characteristic length

$$l\sim e^{\sigma L_\perp^{d-1}/T} , \tag{8}$$

where σ is the surface tension of the pure d-dimensional system. As in our single site ($L_\perp=1$) model, we postulate that criticality will occur when $l\sim L$. Assuming $\sigma\sim\sigma_0[T_c(0)-T]^{\mu}$ with $\mu=(d-1)\nu$, we find an implicit equation for $T_c(L)$ of the connected structure

$$\frac{[T_c(0)-T_c(L)]^{(d-1)\nu}}{T_c(L)}\sim\frac{\ln L}{\sigma_0 L_\perp^{d-1}} . \tag{9}$$

Equation (9) contains both standard finite-size scaling ($L_\perp\rightarrow\infty$, L finite) and 1D scaling ($L\rightarrow\infty$, $L_\perp$ finite). For the special case of $L_\perp=1$, for which the exact solution exists, Eq. (6), Eq. (9) is valid only in the limit $L\rightarrow\infty$. This is not surprising in view of the simplicity of the heuristic argument invoked. However, Eq. (9) does indicate that the relevant combination of length scales governing the asymptotic behavior is $(\ln L)/L_\perp^{d-1}$. Thus, only in the limit of long, narrow pores would we expect the 1D scaling form to hold.

We have observed similar correlation effects for Potts spins on a decorated lattice (see Appendix A). In particular, the absence of any transitions in the decorated antiferromagnetic Potts model ($q\geq 3$) can be traced to the lack of 1D ordering at $T=0$.

We now turn to consider the decorated Ising lattice in the presence of a substrate field h_1. In this case, the transition occurs at $h\neq 0$ and the corresponding magnetization at the critical point is nonzero. This is consistent with the symmetry-breaking effect of the surface field h_1; the average field over the whole sample is also nonzero at the transition. We find from the exact solution that $T_c(L)$ is now driven to zero for sufficiently large h_1 as shown in Fig. 2. This behavior can be understood qualitatively by considering the $T=0$ phase diagram for the original decorated lattice in the $(-h,h_1)$ parameter space. There exist three possible ground states: all spins up [all sites occupied] or down [empty lattice], and a $+/-$ phase; pore spins up but node spins down [all but the

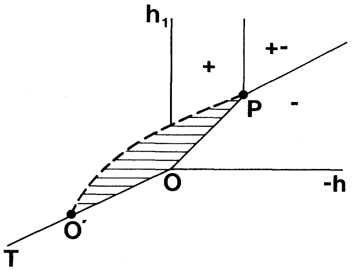

FIG. 2. Schematic phase diagram in $(-h,h_1,T)$ space. Shaded surface is the porous medium's only nonzero T transition. At $T=0$, there is also a $+/-$ phase. P is a $T=0$ triple point and O' is the critical point for $h_1=0$ at a temperature $T_c(L)$.

node sites occupied]. These states coexist at a triple point denoted by P. At finite T, however, only the transition associated with the line OP remains; there is thus a threshold value of the substrate field, corresponding to the value of h_1 at P. The absence of the $+/-$ phase at nonzero T may be understood in terms of the role of 1D fluctuations; the $+/-$ phase corresponds to isolated clusters of spins which cannot order for $T>0$.

While the present calculation exploits properties of the space-filling $(L=0)$ Ising lattice, the critical exponents will not be the same, in general, as those of the latter problem. The reason for this possibly surprising behavior is that the mapping, while nonsingular, introduces a new dependence into the pertinent quantities. The usual symmetries associated with the conventional Ising transition have been broken by the substrate field h_1. This results in a nonzero magnetization at the transition and effective critical exponents. The critical behavior can be determined from the known scaling properties of the Ising model. The singular part of the latter's free energy[9] satisfies

$$F_{\text{sing}}=f(t',h')=\lambda^d f(\lambda^{-y_t}t',\lambda^{-y_h}h') , \tag{10}$$

where the reduced temperature $t'=T'/T_c(L)-1$ and field h' are, in this case, functions of h and h_1, as well as T. Because of these dependences, derivatives of the free energy with respect to the physical temperature or external field will generate linear combinations of the specific heat and susceptibility (see Appendix B). However, as y_h is the larger of the two scaling dimensions, we obtain effective critical exponents at specified nonzero h and h_1 (Ref. 10) for the heat capacity and susceptibility/compressibility:

$$\alpha'=\gamma'=1-1/\delta . \tag{11}$$

Here δ is the magnetic exponent and we have used known relations between Ising exponents. The value of δ is 15 (4.82) in two dimensions (three dimensions). These effective exponents are not equal to either α or γ of the pure system (which would apply only to the special case of a thermodynamic path for which $h'\equiv 0$).

In contrast, the order-parameter exponent β does not differ from its Ising value because it is measured along the system's coexistence curve, by definition; this automatically includes the h dependence on T in a nonsingular way. Interestingly, the density coexistence region associated with the transition is narrowed by a fraction $f=(dL+1)^{-1}$ relative to the pure Ising model because the singularity at the ordering transition arises only from the node sites, which are a fraction f of the total. Such a shrinking of the coexistence region, as well as a reduction of T_c, have been observed in recent experiments.[2]

While the present model takes into account connectivity in the pore structure, it ignores the nonuniform adsorption potential within the pore. For a planar substrate, a spatially varying adsorption potential gives rise to wetting and layering transitions and it is interesting to consider the possibility of such transitions (as well as capillary condensation) in the more restricted geometry of a 1D pore.[5]

We have investigated a single pore with square cross section, containing $L_\perp^2$ adsorption sites, and of length $L_\parallel$. The adsorption potential at $\mathbf{r}$ within the pore is approximated by summing the van der Waals interaction, $-C_6/|\mathbf{r}-\mathbf{r}'|^6$, over all sites of a lattice in the space external to the pore; the value at a given site, V_i, depends on its position within the pore. We thus have the lattice-gas Hamiltonian describing the adsorbate-pore system,

$$H=-J\sum_{\langle i,j\rangle} n_i n_j-\sum_i n_i(\mu-V_i) , \tag{12}$$

where $n_i=0$, 1 and $\mu=\Delta\mu-3J$ being the external chemical potential, $\Delta\mu=0$ corresponding to coexistence of the pure three-dimensional fluid. Our study is complementary to the work of Liu *et al.*, who considered similar 1D pores in a canonical ensemble, thus emphasizing the effects of wetting.[11]

To determine the thermodynamic behavior of this model, we have compared the results from three different calculational techniques; a mean-field (MF) approximation, exact enumeration of the free energy using numerical transfer-matrix (TM) methods[12] and direct TM diagonalization, which is exact in the limit $L_\parallel\to\infty$.

The mean-field equations can be derived in the usual way starting from the Bogoliubov inequality. The true free energy of the system, F, is approximated by the free energy F_0 calculated with a trial Hamiltonian H_0, where

$$F\le F_0+\langle H-H_0\rangle_0 . \tag{13}$$

We ignore thermal fluctuations along the pore but allow the average occupancy per site $\langle n_i\rangle$ to depend on the transverse position of the site within the pore. Thus, the trial Hamiltonian becomes

$$H_0=-\sum_i \hat{\mu}_i n_i , \tag{14}$$

where the sum over i is restricted to the $L_\perp^2$ sites in the transverse direction and $\hat{\mu}_i$ is the mean field at site i. Minimizing F with respect to the variational parameters $\{\hat{\mu}_i\}$ results in the set of coupled mean-field equations:

$$\langle n_i\rangle=\frac{1}{1+e^{-\beta\hat{\mu}_i}} , \tag{15}$$

$$\hat{\mu}_i=\mu-V_i+J\sum_k\langle n_k\rangle , \tag{16}$$

where the sum in the Eq. (16) is over the nearest neighbors of site i in the full three-dimensional space within the pore. Finally, the square symmetry of the infinite pore reduces the number of coupled equations [Eq. (15)] to be solved numerically. By allowing $\langle n_i \rangle$ to depend on the transverse position within the pore, the mean-field solution becomes exact in the limit $T \to 0$.

For such quasi-one-dimensional systems, the transfer-matrix formalism allows an exact evaluation of the free energy. We have employed this technique to calculate numerically, but otherwise exactly, the free energy and hence adsorption isotherms for pores with small cross-sectional areas. For a pore with $L_\perp^2$ transverse adsorption sites, we represent the state of a single layer by a m-dimensional vector, where $m = 2^{L_\perp^2}$. The free energy can be evaluated by numerically updating this vector layer by layer along the pore[12] or by direct diagonalization of the resulting $m \times m$ transfer matrix T_m; including interactions between two neighboring layers along the pore, as well as in-layer interactions in the usual way. Due to numerical limitations, we were able to consider pores with $L_\perp \leq 4$ by the former technique but only $L_\perp \leq 3$ by matrix diagonalization. However, the latter approach is exact for infinite-length pores and allows longitudinal correlation functions to be determined.

Our findings are summarized below:

(1) The ground-state phase diagram can be determined exactly by evaluating the energies of the many possible states of the system. A representative example is shown for $L_\perp = 3$ in Fig. 3. There are four possible $T = 0$ phases: empty, corner adsorption, monolayer adsorption, and full. The sequence of states through which the system passes on approach to saturation, $\Delta\mu \to 0$, depends on the adsorption potential strength C_6. For very small C_6, the pore remains empty even at saturation. For pores of larger cross-sectional areas, these same general features were also observed.

(2) At $T > 0$, as seen in Fig. 4, the transitions become rounded due to thermal fluctuations. This rounding is captured exactly by the TM calculations but is not by the MF approximation. We note that there is surprisingly good agreement between the two sets of results, except within the very narrow regions where MF predicts a spurious transition. This good agreement occurs because at low temperatures the ground-state properties of the system dominate, and these have been preserved within the MF approximation.

(3) We have investigated the dependence of the adsorption isotherms on the length of the pore ($L_\parallel$) by comparing results from both TM methods for infinite pores and finite $L_\parallel$ pores with periodic boundary conditions. In general, we find the adsorption isotherms to be very insensitive to pore length $L_\parallel$, even down to $L_\parallel \simeq L_\perp$, shown in Fig. 4.

(4) Even though the transitions are rounded, the correlation length $\xi_\parallel$ along the pore shows marked peaks at values of $\Delta\mu/J$ corresponding to the transitions predicted by MF, also shown in Fig. 4. It is for this reason that the isotherms are insensitive to $L_\parallel$; only if $\xi_\parallel \geq L_\parallel$ does this finite-length scale play an important role. In particular, in a connected system of pores as in the former model, it is plausible that these correlations could propagate throughout the system giving rise to true thermodynamic singularities.

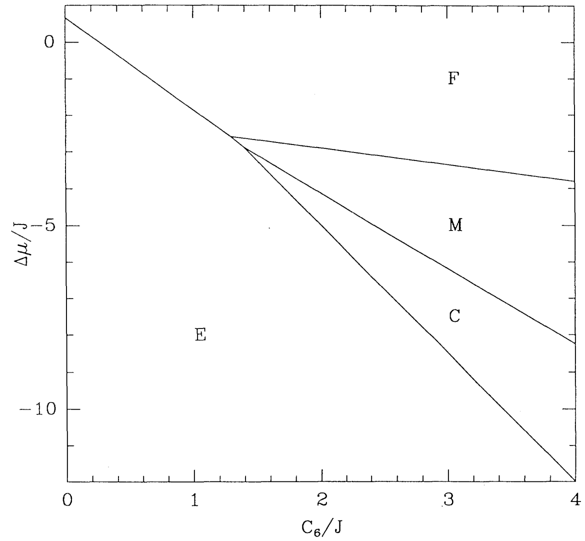

FIG. 3. The ground-state phase diagram for an infinite pore with $L = 3$. There are four possible phases; empty (E), corner adsorption (C), monolayer adsorption (M) and full (F). Note that for small C_6/J the pore remains empty even at saturation, $\Delta\mu/J = 0$.

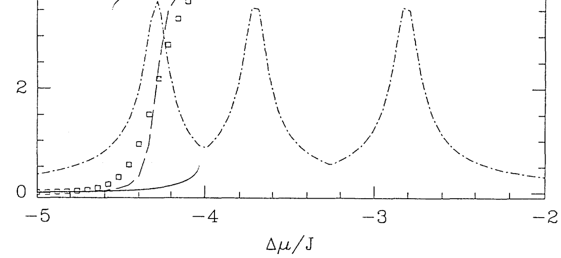

FIG. 4. Adsorption isotherms and correlation length $\xi_\parallel$ for a square pore with $L_\perp = 3$, $C_6/J = 1.8$, and $T/J = 0.25$. The dashed curve shows the average occupancy per layer N as a function of $\Delta\mu/J$, calculated exactly by TM diagonalization. The squares show the corresponding results for a finite pore with $L_\parallel = 3$ and periodic boundary conditions. The stable branches of solutions generated by the MF approximation are also plotted. The spurious MF transitions occur at the maxima of the longitudinal correlation length, $\xi_\parallel$ shown on the same scale by the dot-dash curve, where N now denotes the number of correlated sites along the pore.

While our models are somewhat simplified, they do include the fundamental symmetry aspects of many porous media, namely, interconnected quasi-one-dimensional pores in 3D space. It is the presence of both 1D and 3D scaling properties that lead to many of the features captured here by our model: the reduction of T_c and the role of 1D correlation, the shrinking of the coexistence region, and the symmetry-breaking effect of the substrate field giving rise to critical exponents. These general results should be applicable to a wide class of porous systems.

This research has been supported by the ONR, NSF, NASA, and the Petroleum Research Fund administered by the American Chemical Society. We are grateful to Moses Chan, Marek Cieplak, David Huse, Amos Maritan, and especially Attilio Stella for helpful discussions. We also are grateful to the Center for Academic Computing at Penn State. Parts of the numerical calculations have been performed on Ohio Supercomputer Center Cray Y-MP8/864.

APPENDIX A: DECORATED POTTS LATTICE

The role of 1D correlations in the decorated lattice can be investigated further by considering Potts spins occupying the lattice sites shown in Fig. 1. The Hamiltonian for the system is

$$\frac{H}{T}=-K\sum_{\langle i,j\rangle}\delta_{\sigma_i\sigma_j}\ , \tag{A1}$$

where $\sigma_i=1,2,\ldots,q$ and the sum is over nearest neighbors in the decorated lattice. As in the Ising case, the partial trace over pore sites (X) can be performed exactly (in the absence of any symmetry-breaking field), giving a renormalized coupling K' in a uniform d-dimensional Potts lattice. For q-state Potts spins, one finds[13]

$$K'=\ln\left[1+\frac{q}{(1+q/e^{K}-1)^{L+1}-1}\right]\ , \tag{A2}$$

where K is the coupling parameter in the original decorated Potts lattice. The variation of K' with K and L is shown in Fig. 5 for the three-state Potts model and both ferro- and antiferromagnetic ordering. Criticality in the decorated lattice again requires the renormalized coupling K' to equal the critical coupling of the regular lattice in d dimensions, $K_c^{(d)}$. These critical couplings are shown on the ordinate for $d=2$ and 3.[13]

For $K>0$ and any L, we can always reach criticality in the decorated lattice. However, for $K<0$ and $L>2$, no critical transitions will occur in the decorated model as seen in Fig. 5. This is a direct consequence of the fact that for $q\geq 3$ and antiferromagnetic ordering, the 1D correlation length remains finite even at $T=0$. Thus, correlations cannot propagate between node sites (which requires $\xi_{\parallel}\geq L$) and the system exhibits 1D behavior.

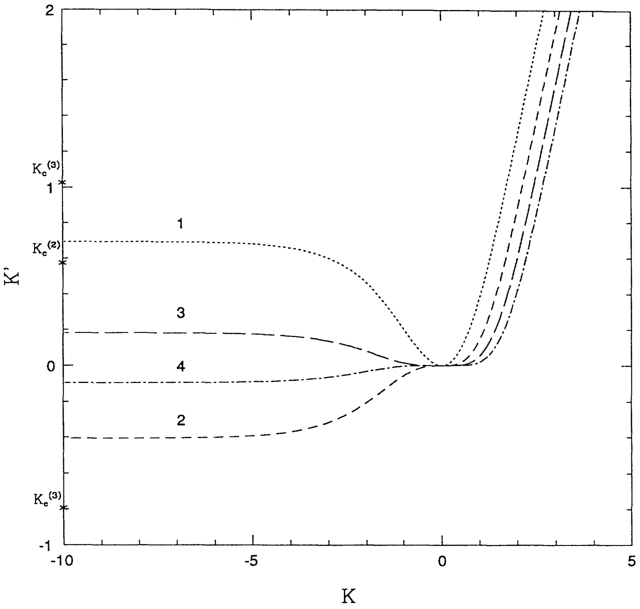

FIG. 5. Renormalized couplings K' as a function of original coupling K, for the three-state Potts model on the decorated lattice. The curves correspond to lattices with $L=2$, 3, 4, and 5. The critical coupling for the pure system in both two and three dimensions, $K_c^{(d)}$, is marked on the abscissa; criticality in the decorated lattice requires $K=K_c^{(d)}$.

APPENDIX B: CRITICAL EXPONENTS

Let h' and t' be the critical point parameters of the space-filling Ising lattice. They can be expanded in terms of the external field, h, and physical temperature, t, of the decorated lattice, scaled such that criticality occurs for $h'=t'=h=t=0$. In a linear regime close to criticality, we may write

$$t'=at+bh\ , \tag{B1}$$

$$h'=ct+dh\ . \tag{B2}$$

In the critical region, the singular part of the free energy obeys the scaling form

$$f(t',h')=\lambda^d f(\lambda^{-y_t}t',\lambda^{-y_h}h')\ , \tag{B3}$$

with y_t and y_h the standard Ising exponents. If the external field is chosen such that $h=0$, the free energy as a function of the physical temperature is

$$F(t)\sim\lambda^d f(\lambda^{-y_t}at,\lambda^{-y_h}ct)\ . \tag{B4}$$

The specific heat follows by taking derivatives with respect to t,

$$\begin{aligned}C(t)\sim{}&a^2\lambda^{d-2y_t}f_{xx}(\lambda^{-y_t}at,\lambda^{-y_h}ct)\\&+ac\lambda^{d-y_t-y_h}f_{xy}(h^{-y_t}at,\lambda^{-y_h}ct)\\&+c^2\lambda^{d-2y_h}f_{yy}(\lambda^{-y_t}at,\lambda^{-y_h}ct)\ ,\end{aligned} \tag{B5}$$

where $f_{xx}\equiv(\partial^2/\partial x^2)f(x,y)$, etc.

The important point is that because $y_h>y_t$, λ must be chosen to scale with t such that

$$\lambda\sim t^{1/y_h}\ . \tag{B6}$$

As $t\to 0$, the dominant singularity arises from the last term of Eq. (B5),

$$C(t)\sim c^2 t^{(d-2y_h)/y_h} f_{yy}(0,c)\ , \tag{B7}$$

giving the new α' exponent

$$\alpha'=\frac{2y_h-d}{y_h}\ . \tag{B8}$$

In terms of the exponent δ, $y_h/d=\delta/(1+\delta)$,

$$\alpha'=1-\frac{1}{\delta}\ . \tag{B9}$$

Note that this is not equal to the standard exponent

$$\gamma=\frac{2y_h-d}{y_t} \tag{B10}$$

for the pure Ising system, but is the exponent characterizing the divergence of the susceptibility at T_c as the external field goes to zero.

Similarly, the susceptibility for the decorated lattice may be calculated by taking derivatives of Eq. (B3) with respect to h. Again, the dominant singularity is controlled by y_h giving rise to the effective susceptibility exponent

$$\gamma'=\frac{2y_h-d}{y_h}=1-\frac{1}{\delta}\ . \tag{B11}$$

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[1] *Physics and Chemistry of Porous Media-II,* Proceedings of the Second International Symposium on the Physics and Chemistry of Porous Media, edited by J. R. Banavar, J. Koplik, and K. Winkler, AIP Conf. Proc. No. 154 (AIP, New York, 1986); *Molecular Dynamics in Restricted Geometries,* edited by J. Klafter and J. M.Drake (Wiley, New York, 1989), and references therein.

[2] A. Wong and M. H. W. Chan, Phys. Rev. Lett. **65**, 2567 (1990); see also S. B. Dierker and P. Wiltzius, *ibid.* **58**, 1865 (1987); P. Wiltzius, S. B. Dierker, and B. S. Dennis, *ibid.* **62**, 804 (1989); B. J. Frisken, F. Ferri, and D. S. Cannell, *ibid.* **66**, 2754 (1991).

[3] P. G. de Gennes, J. Phys. Chem. **88**, 6469 (1984).

[4] M. H. W. Chan, K. I. Blum, S. Q. Murphy, G. K. S. Wong, and J. D. Reppy, Phys. Rev. Lett. **61**, 1950 (1988); G. K. S. Wong, P. A. Crowell, H. A. Cho, and J. D. Reppy, *ibid.* **65**, 2410 (1990); R. A. Guyer and J. Machta, *ibid.* **60**, 2054 (1988).

[5] R. C. Ball and R. Evans, Langmuir **5**, 714 (1989); R. Evans, in *Liquids at Interfaces,* edited by J. Charvolin, J. F. Joanny, and J. Zinn-Justin (Elsevier, Amsterdam, 1990); S. Dietrich, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and J. L. Lebowitz (Academic, New York, 1989), Vol. 12.

[6] For a recent review, see D. S. Fisher, G. M. Grinstein, and A. Khurana, Phys. Today **41** (12), 56 (1988); A. Maritan, M. R. Swift, M. Cieplak, M. H. W. Chan, M. W. Cole, and J. R. Banavar, Phys. Rev. Lett. **67**, 1821 (1991).

[7] M. E. Fisher, Phys. Rev. **113**, 969 (1959); I. Syozi, in *Phase Transitions and Critical Phenomena,* edited by C. Domb and M. S. Green (Academic, New York, 1972), Vol. 1, p. 291.

[8] S. Fishman and T. A. L. Ziman, Phys. Rev. B **26**, 1258 (1982).

[9] M. E. Fisher, Rev. Prog. Phys. **30**, 615 (1967).

[10] This dependence on h introduces a bias which is absent in the case of "Fisher renormalization" [M. E. Fisher, Phys. Rev. **176**, 257 (1968)].

[11] A. J. Liu, O. J. Durian, E. Herbolzheimer, and S. A. Safran, Phys. Rev. Lett. **65**, 1897 (1990); see also M. R. Swift, A. L. Owczarek, and J. O. Indekeu, Europhys. Lett. **14**, 475 (1991).

[12] I. Morgenstern and K. Binder, Phys. Rev. B **22**, 288 (1980); H. W. J. Blote and M. P. Nightingale, Physica A **112**, 405 (1982); E. Cheng, J. R. Banavar, M. W. Cole, and F. Toigo, Surf. Sci. **216**, 389 (1992).

[13] F. Y. Wu, Rev. Mod. Phys. **54**, 235 (1982).