Interfacial adsorption in the two-dimensional Blume–Capel model

Walter Selke†, David A Huse‡, and Daniel M Kroll†

† Institut für Festkörperforschung der Kernforschungsanlage Jülich, Postfach 1913, 5170 Jülich 1, Federal Republic of Germany
‡ Bell Laboratories, Murray Hill, New Jersey 07974, USA

Abstract. New Monte Carlo (MC) results on the interfacial adsorption in the vicinity of the tricritical point of the two-dimensional Blume–Capel model are reported. The net adsorption, W_0, is found to diverge as $W_0 \sim t_c^{-\omega}$ with $\omega = 0.44 \pm 0.02$ as the tricritical point is approached along a path asymptotically not parallel to the phase boundary. This behaviour as well as previous MC results on the interfacial adsorption at the critical line and the first-order transition of the model are explained using analytic and scaling arguments. In addition, finite size effects *at* the first-order transition are analysed, and it is shown that the critical adsorption scales as $L_\perp f(L_\parallel / L_\perp^2)$ where $L_\parallel$ and $L_\perp$ are the lengths of the system in the directions parallel and perpendicular to the interface. The scaling function f is evaluated by performing MC simulations on systems with various length to width ratios.

1. Introduction

Most of the studies on interfaces have been confined to two phase systems. A more complex situation may be encountered at interfaces between two phases (α, β) in the presence of one or more additional phases $(\gamma, \ldots)$. For example, in a two-component fluid system in equilibrium with its vapour phase a thin film of the heavier fluid may intervene and 'wet' the interface between the lighter fluid and the vapour (Moldover and Cahn 1980). This wetting effect which is caused by an interplay of interface tensions, repulsion between interfaces, and the gravitational field has been described within the classical, phenomenological framework (Cahn 1977, Widom 1978, Teubner 1983).

A similar effect, 'interfacial adsorption' (Selke and Huse 1983), has recently been found (Selke and Pesch 1982) in multi-state models, i.e. lattice models where the variables, S_i, on lattice sites i, can have at least three values, $S_i = \alpha, \beta, \gamma, \ldots$. In particular, two-dimensional models which have been studied include the q-state Potts models, $q = 3, 4, \ldots$ (Selke and Pesch 1982, Selke and Huse 1983, Selke 1984b), the three-state chiral clock model (Huse and Fisher 1982, 1984, Huse *et al* 1983, Selke 1984a, b), the Blume–Capel model (Selke and Yeomans 1983) as well as lattice gas models for adsorbates like H/Fe(110) and O/Pd(110) (Sega *et al* 1984, Selke 1984b). In all cases an interface is introduced by fixing variables on opposite boundaries in two different states, say α and β. At the interface an excess of *non-boundary* states,

$\gamma, \ldots$, occurs, either in the form of droplets, e.g., in the Potts models, or in a layer-like fashion. To study this effect quantitatively the 'net adsorption of non-boundary states at the interface', W, is defined by comparing systems with and without interfaces. The adsorption is found to exhibit interesting new critical behaviour as one approaches either the bulk transition temperature, T_c, or in some cases a distinct 'wetting temperature' below T_c (for a review see Selke 1984b).

In this article the previous Monte Carlo (MC) work of Selke and Yeomans (1983) on the interfacial adsorption, W_0, in the two-dimensional Blume–Capel (BC) model is extended. The model is known to display a second-order transition, a tricritical point, and a first-order transition. Selke and Yeomans (1983) found different types of critical behaviour for W_0 at the various transitions. We extend the previous MC calculations in two ways: the vicinity of the tricritical point is explored in much more detail, because crossover effects may have been overlooked previously. Indeed, the exponent for the temperature dependence of W_0 on approaching tricriticality is found to be larger than reported previously. In addition, finite size effects at the first-order transition are studied by performing MC calculations on systems with various length to width ratios.

The outline of the article is as follows: in § 2 the previous MC findings (Selke and Yeomans 1983) and our new results in the vicinity of the tricritical point are described. As shown in § 3, all these results can be explained using scaling arguments as well as analytic approaches employing, for example, the collective coordinates method (Diehl *et al* 1980, Lipowsky *et al* 1983). In addition, the finite size scaling behaviour at the first-order transition is discussed in that section. The critical adsorption is found to scale as $L_\perp f(L_\parallel / L_\perp^2)$ where $L_\parallel$ and $L_\perp$ are the lengths of the system parallel and perpendicular to the interface. A brief summary and an outlook to possible experimental applications conclude the article.

2. Monte Carlo results

The Blume–Capel model (Capel 1966, Blume 1966) is described by the spin-1 Ising Hamiltonian

$$\mathcal{H} = -J \sum_{\langle ij \rangle} S_i S_j + D \sum_i S_i^2 \qquad S_i = -1, 0, 1 \tag{1}$$

where $\langle ij \rangle$ denotes summation over nearest neighbours on a square lattice of size $L \times L$, and $J, D > 0$. The model is known to display a second-order transition for $0 \leq D/J \leq (D/J)_t$, a tricritical point at $(D/J)_t$ (close to 1.95; the exact position is open to question, see below) and a first-order transition in the range $(D/J)_t < D/J < 2$. The phase diagram, based on MC results for finite lattices, but extrapolated to $L \to \infty$, is depicted in figure 1 (see Selke and Yeomans 1983).

To introduce an interface, the spins on two opposite boundaries were fixed in the two different non-zero states ± 1. Two types of boundary conditions were considered in the lateral direction. Most data were taken with fixed spin boundary conditions: on one half of the systems the boundary spins were fixed to be $+1$, on the other -1. This pins the ends of the interface each at half way across the system (see, e.g., figure 1 of Selke and Yeomans 1983 or Selke 1984b). A few data were taken with periodic boundary conditions in the lateral direction. However, since the behaviour of the interfacial adsorption was found to be essentially the same in both cases, the results discussed here are all only for the fixed spin case.
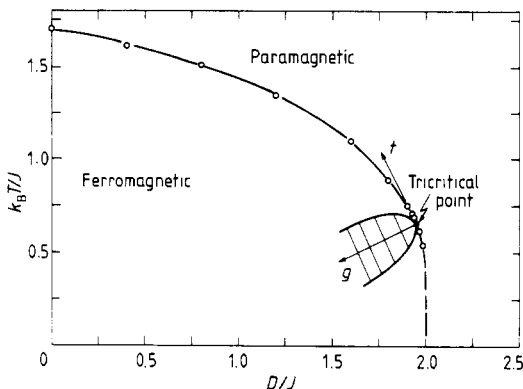
Figure 1. Phase diagram of the two-dimensional Blume–Capel model. The crossover region of the tricritical point below the phase boundary is sketched.

By examining typical equilibrium MC configurations it is seen that an excess of the non-boundary state, $S_i = 0$, is generated at the interface (see, e.g. figure 1 of Selke and Yeomans 1983). This phenomenon is described quantitatively by the net adsorption per unit length of the interface (Selke and Yeomans 1983)

$$W_0(T, L) = \frac{1}{L} \sum_i (\langle \delta_{0,s_i} \rangle_{1:-1} - \langle \delta_{0,s_i} \rangle_{1:1}) \tag{2}$$

where the angular brackets denote thermal averages and the subscripts $1:-1$ and $1:1$ refer to systems with and without interfaces, respectively. The (possibly) critical behaviour of W_0 can be characterised by two critical exponents, ω and a:

$$W_0(t_c, L = \infty) \sim t_c^{-\omega} \qquad \text{for small } t_c \tag{3}$$

and

$$W_0(T = T_c, L) \sim L^a \qquad \text{for large } L \tag{4}$$

where $t_c = (T_c - T)/T_c$, and T_c is the bulk transition temperature. Instead of $W_0(T = T_c, L)$, one may also consider the maximum adsorption at fixed D/J, $W_0^{\max}(L)$, to define a.

In a previous Monte Carlo study (Selke and Yeomans 1983) it has been shown that W_0 does not diverge at the critical line, $D/J < (D/J)_t$. At the first-order line ω was estimated to be $\omega = 0.33 \pm 0.03$. Indeed, as is discussed below, $\omega = \frac{1}{3}$ is expected. An exponent $a = 0.7 \pm 0.05$ has been found at the first-order line, but it will be shown in the next section that this is just an apparent exponent. At the tricritical point, W_0 appeared to diverge weakly, possibly in a logarithmic fashion (Selke and Yeomans 1983). However, these data were taken at constant (D/J), i.e. along a path almost parallel to the transition line, see figure 1. This may have lead to quite complicated crossover phenomena, and the data have to be interpreted with great care. In order to avoid seeing such phenomena one must approach the tricritical point along one of the two relevant scaling axes, $t = 0$ or $g = 0$ (e.g. Fisher 1974), see figure 1. The t-axis, on which $g = 0$, is tangent to the transition line. The orientation of the g-axis is not

known, but one would expect it to be approximately perpendicular to the *t*-axis. Thus, in order to avoid possible misleading crossover effects we have chosen to approach the tricritical point along a line perpendicular to the phase boundary in the belief that t/g is small on that line. Indeed, MC runs along that path indicate a divergence of the net adsorption, W_0, as depicted in figure 2. Typically we took 2–3×10^4 Monte
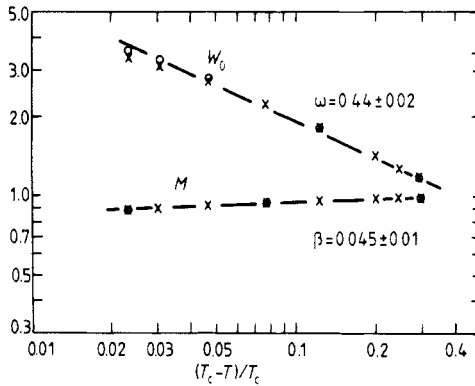

Figure 2. Log–log plot of the interfacial adsorption, W_0, and the order parameter, M, against temperature on approaching the tricritical point along a path perpendicular to the phase boundary. The crosses denote $L = 40$ and the circles denote $L = 60$.

Carlo steps per site (MCS/site). The sizes of the systems were 40×40 and 60×60 (only a few points are shown for the latter case). In the range $2 \times 10^{-2} \lesssim t_c \lesssim 3 \times 10^{-1}$ we obtain, using linear regression, $\omega = 0.44 \pm 0.02$ (note that here $t_c \sim g$) where we took $(D/J)_t = 1.945$ and $k_B T_t/J = 0.65$, see below. MC runs along slightly different paths, e.g. assuming $(D/J)_t$ to be 1.931 (Selke and Yeomans 1983), gave estimates for ω within the error bars quoted above, reflecting the existence of a broad crossover region. To check the reliability of the estimate we determined the tricritical exponent, β, of the order parameter, $M \sim \Sigma_i \langle S_i \rangle$, from the same MC runs. For $(D/J)_t = 1.945$ we found $\beta = 0.04 \pm 0.01$, see figure 2, in very good agreement with the exact value $\beta = \frac{1}{24}$ (Nienhuis 1982, den Nijs 1983). Assuming $(D/J)_t$ to be 1.931 the effective exponent β, determined in the same range of temperatures as ω, becomes somewhat larger, $\beta \approx 0.06$. This suggests that the tricritical point is located closer to $(D/J) = 1.945$ than 1.931. Additional evidence for this position of the tricritical point stems from finite size analyses, as shown in figure 3. There we monitored the maximum adsorption, $W_0^{max}(W_0(T_c)$ at the first-order line) at fixed values of D/J as a function of the linear dimension L. Log–log plots in the vicinity of the tricritical point showed a pronounced curvature. We therefore determined effective exponents $a_{eff}(L_1, L_2, L_3)$ from fitting a slope to the data for systems of sizes $L_1 \times L_1$, $L_2 \times L_2$, and $L_3 \times L_3$ with $L_3 > L_2 > L_1$. Of course, for large values of L this quantity should approach the exponent of equation (4). The results are displayed in figure 3, taking 2×10^4 to 10^5MCS/site, the larger number for the larger systems, especially $L = 100$. They are consistent with a vanishing exponent, a, at the critical line and a value of $a \approx 0.7 \pm 0.05$ at the first-order line (see below). The change between these two types of behaviour occurs at $D/J = 1.945 \pm 0.01$, presumably at the tricritical point. However, one must be cautious here because the expected value of $a = \frac{4}{5}$ at the tricritical point is not seen, as discussed in §3.
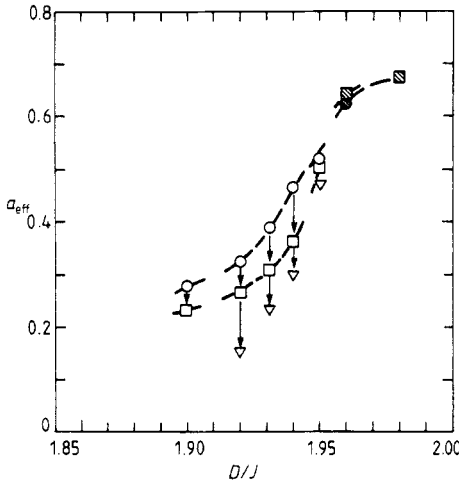
Figure 3. Finite size behaviour of the apparent exponent a_{eff} (see text) at various values of D/J. The open symbols refer to the maximum adsorption, W_0^{max}, while the shaded symbols denote Monte Carlo results taken at the bulk critical temperature. The circles denote $L = 10, 20, 40$; the squares denote $L = 20, 40, 60$ and the triangles denote $L = 40, 60, 100$.

3. Analytic and scaling arguments

In order to understand the MC results, it is useful to express W_0 as a derivative of the interface tension σ. Noting that $\langle \delta_{0, S_i} \rangle = 1 - \langle S_i^2 \rangle$, W_0 may be rewritten as

$$W_0 = \frac{1}{L} \sum_i [\langle S_i^2 \rangle_{1:1} - \langle S_i^2 \rangle_{1:-1}] \tag{5}$$

Denoting the total free energy for $1:1(1:-1)$ boundary conditions by $F_{1:1}(F_{1:-1})$, W_0 can then be expressed in terms of the interface tension of the system $\sigma = (1/L)(F_{1:1} - F_{1:-1})$ as

$$W_0 = \beta^{-1} \partial \sigma / \partial D \tag{6}$$

where $\beta = 1/k_B T$.

For the spin-$\frac{1}{2}$ Ising model it is known that $\sigma \sim t_c$ for $t_c = (T_c - T)/T_c \to 0^+$ at the critical point (Onsager 1944). The BC model belongs to the same universality class for $D/J < (D/J)_t$, so that we expect $\sigma \sim t_c$, where t_c is a measure of distance from the critical curve of this model. Since D is a non-ordering field (Riedel 1972) one has

$$W_0 \sim \partial \sigma / \partial t_c \sim \text{constant} \tag{7}$$

on the critical line, in agreement with the MC results.

At the tricritical point there are two relevant scaling fields, t and g, as discussed above. When $t \to 0$ with $g = 0$, the tricritical point is approached tangentially to the phase boundary, whereas an increase in g results in departure from the tricritical point and the phase boundary. Near the tricritical point the bulk free energy density has the scaling form (Fisher 1974, Lawrie and Sarbach 1984)

$$F = g^{2-\alpha} \Phi(t/g^\phi) \tag{8}$$

where $\alpha = \frac{8}{9}$ and $\phi = \frac{4}{9}$ are the expected exact exponents in two dimensions (Nienhuis 1982). Similarly, the interface tension σ should scale as (Widom 1974)

$$\sigma = g^{\mu} \sum (t/g^{\phi}) \tag{9}$$

where $\mu = 2 - \alpha - \nu = (d-1)\nu = \frac{5}{9}$. D has a projection on both g and t so that using equation (6), the leading singularity in W_0 is found to be

$$W_0 = g^{-4/9}\Omega(t/g^{4/9}). \tag{10}$$

The exponent $\frac{4}{9}$ thus describes the divergence of W_0 for paths asymptotically *not* parallel to the transition line. For paths asymptotically parallel to the phase boundary (e.g. along the critical line) $W_0 \sim t^{-1}$. Because of the steepness of the phase boundary at the tricritical point, see figure 1, MC data taken for fixed value of D/J may well be in the crossover region of large $(t/g^{4/9})$. Furthermore, the interpretation of the data in this case is very sensitive to the exact value of $(D/J)_t$. On the other hand, for paths more or less perpendicular to the phase boundary, the scaling region is very wide and the scaling behaviour should be rather insensitive to the exact value taken for $(D/J)_t$. Indeed, this is confirmed by the MC data reported above. The exponent $\omega = 0.44 \pm 0.02$ is obviously in agreement with the expected exact result.

The finite-size behaviour observed at the tricritical point, on the other hand, does not give an apparent exponent consistent with the exact exponents. Since the correlation length at the tricritical point scales as

$$\xi_t = g^{-5/9}\Xi(t/g^{4/9}), \tag{11}$$

finite size scaling implies $W_0^{max} \sim L^{4/5}$ *at the tricritical point*. In figure 3 it is clear that we do not observe this behaviour. Presumably much larger systems sizes as well as a precise knowledge of the tricritical coordinates would be necessary to observe the expected $a = \frac{4}{5}$. (A MC study of Landau and Swendsen suggests $(D/J)_t$ to be close to 1.96.) A careful MC study at the tricritical point of hard squares with diagonal interactions (Huse 1982, Baxter and Pearce 1983), whose position is exactly known, might clarify this problem.

At the critical and tricritical points, the singularities in the adsorption, W_0, are induced by bulk critical fluctuations. On the other hand, at the first-order transition there are no bulk critical fluctuations and the divergence of W_0 arises from an interface delocalisation transition (Abraham and Smith 1982, Kroll and Lipowsky 1983). Near the first-order transition, the interface between the '+1' rich or [+] phase and the '−1' rich or [−] phase may be viewed as having a compound structure, consisting of a microscopic layer of '0' rich or [0] phase bounded above and below by an interface. The widths of the basic [+/0] and [0/−] interfaces do not diverge at the transition; the divergence of the width of the [+/−] interface is simply an unbinding of the two basic interfaces. This transition is most easily studied using a model in which the dynamical variables are simply the coordinates of the [+/0] and [0/−] interfaces, $y_+(x)$ and $y_-(x)$, respectively, where the x-axis is chosen parallel to the interface and $y_+(x) > y_-(x)$. It is straightforward to derive the Lagrangian of this model using the collective coordinates method described elsewhere (Diehl *et al* 1980, Lipowsky *et al* 1983). At sufficiently large length scales the free energy functional is well approximated by

$$F\{y_+, y_-\} = \int dx \left\{ \tfrac{1}{2}\sigma_c \left[\left(\frac{dy_+}{dx}\right)^2 + \left(\frac{dy_-}{dx}\right)^2 \right] + V(y_+(x) - y_-(x)) \right\} \tag{12}$$

where $V(w) \sim e^{-w/\xi_c} + (f_0 - f_+)w$. The bulk free energy densities of the [0] and [+] phases, f_0 and f_+, respectively, are equal at the transition. The mean field interface tension is σ_c, and ξ_c is the mean field bulk correlation length at the transition. To simplify this model, make the change of variables $y(x) = \frac{1}{2}(y_+(x) + y_-(x))$ and $z(x) = \frac{1}{2}(y_+(x) - y_-(x))$. The fluctuations in $y(x)$ may then be integrated out (they do not contribute any singularities to W_0) leaving

$$F\{z(x)\} = \int dx\{\sigma_c(dz/dx)^2 + V(2z)\}. \tag{13}$$

This Lagrangian has been previously considered; it also arises in the problem of interfacial unbinding from a rigid free surface (Lipowsky *et al* 1983). The adsorption W_0 is simply proportional to the average thickness of the [0] phase layer $\langle 2z \rangle$. In two dimensions we then expect (Lipowsky *et al* 1983)

$$W_0 \sim t_c^{-1/3} \tag{14}$$

where $t_c \sim (T_c - T) \sim (f_0 - f_+)$. This critical exponent, $\omega = \frac{1}{3}$, which is certainly consistent with the MC data (Selke and Yeomans 1983) was originally found in a lattice model with interface unbinding (Abraham and Smith 1982), but should hold quite generally at first-order transitions in two-dimensional systems (Kroll and Lipowsky 1983, Fisher 1984).

It should be noted that there is an additional diverging length scale at this wetting transition, namely $\xi_\parallel$, the longitudinal correlation length, describing composition correlations parallel to the interface. $\xi_\parallel$ is found to diverge with the exponent $\nu_\parallel = 2\omega = \frac{2}{3}$ (Abraham and Smith 1982, Huse and Fisher 1982, Lipowsky *et al* 1983).

The problem of analysing finite size effects at the first-order transition is complicated by the fact that there are several diverging lengths in the problem. Let $L_\parallel$ denote the length of the system in the direction parallel to the interface and $L_\perp$ the length in the perpendicular direction. As $t_c \to 0$, both $W_0 \sim t_c^{-1/3}$ and $\xi_\parallel \sim t_c^{-2/3}$ diverge. The finite size of the system plays a role when either $W_0 \sim L_\perp$ and/or $\xi_\parallel \sim L_\parallel$. Since $\nu_\parallel = 2\omega$ we therefore expect $W_0(t_c = 0) \sim L_\parallel^{1/2}$ for $x = L_\parallel / L_\perp^2 \to 0$ and $W_0(t_c = 0) \sim L_\perp$ for $x \to \infty$. This motivates the scaling ansatz

$$W_0(t_c = 0) = L_\perp f(L_\parallel / L_\perp^2) \tag{15}$$

where $f(x) \to x^{1/2}$ in the limit $x \to 0$ and $f(x) \to \text{constant}$ for $x \to \infty$. The scaling function can be evaluated by performing MC simulations on systems with various length to width ratios. This was done at T_c for $D/J = 1.96$ and 1.98 (T_c was obtained considering systems without an interface with full periodic boundary conditions). Some results are shown in figure 4 where it may be seen that the data fall reasonably well on one curve. In particular, in the range $10^{-3} \leq x \leq 10^{-2}$ we find $f(x) \sim x^{1/2}$, in agreement with the above argument. For $L_\parallel = L_\perp = L$ and $10 \leq L \leq 80$ all data can be fitted relatively well by a power law $f(x) \sim x^{0.3}$ (implying $a_{\text{eff}} = 0.7$ (Selke and Yeomans 1983)). However, because these data lie in a region of intermediate values of x, this is only an apparent exponent.

One problem with examining in detail the finite size scaling behaviour at a particular D/J and $T(= T_c)$ is that one cannot be certain of T_c and can get spurious results if one's estimate of T_c is incorrect. Another measure for testing finite size scaling at the first-order transition is W_0^{max} which does not rely on any knowledge of T_c. While there should be little boundary-condition dependent shift in the effective T_c in the system
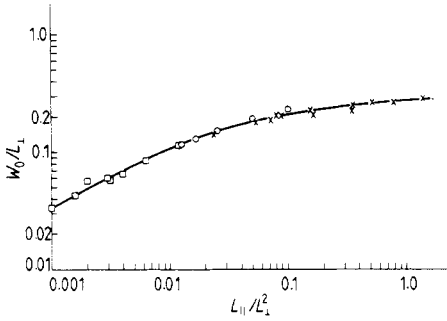
Figure 4. Interfacial adsorption at the bulk transition temperature of the first-order transition $(D/J = 1.96, k_B T/J = 0.6275)$. The Monte Carlo data for various length to width ratios $(L_\parallel, L_\perp)$ ($\bigcirc$, $L_\perp = L_\parallel$; $\times$, $L_\perp < L_\parallel$; $\square$, $L_\perp > L_\parallel$) fall on one scaling curve, see equation (15).

with $(1:-1)$ boundary conditions, for the $(1:1)$ system the first-order melting should be delayed by an amount proportional to L^{-1} due to the need to produce two new interfaces (Fisher and Berker 1982, Lipowsky and Gompper 1984). There should thus be a temperature range of order L^{-1} in which the $(1:1)$ system is fully ordered (no [0] phase present) but the two interfaces in the $(1:-1)$ system have fully unbound. Thus we may postulate

$$W_0^{max} = L_\perp \tilde{f}(L_\parallel/L_\perp^2) \tag{16}$$

where we expect $W_0^{max} \sim L_\perp$. This should be more or less independent of the system shape $L_\parallel/L_\perp^2$. Thus we expect $\tilde{f}(x) \approx$ constant, so that for $L \times L$ systems

$$W_0^{max} \sim L. \tag{17}$$

However, because of pronounced metastability effects at the first-order transition it is very difficult to determine W_0^{max} reliably, and we did not perform a detailed MC analysis. Nevertheless, at $D/J = 1.98$ we did find for large $L \times L$ systems, $40 \le L \le 100$, an almost linear increase of W_0 with L in a small range of temperatures above T_c.

4. Summary

In this paper we have extended and refined previous (Selke and Yeomans 1983) Monte Carlo work on interfacial adsorption in the two-dimensional Blume–Capel model. In particular, the net adsorption, W_0, is found to diverge as $W_0 \sim t_c^{-\omega}$ with $\omega = 0.44 \pm 0.02$ as the *tricritical point* is approached along a path asymptotically not parallel to the phase boundary. This behaviour is explained using a scaling argument which leads to $\omega = \frac{4}{9}$. The non-critical behaviour at the *continuous transition* is also explained by a scaling argument. Using the collective coordinates method the exponent which describes the temperature dependence of W_0 as one approaches the *first-order transition* is expected to be $\omega = \frac{1}{3}$, in excellent agreement with the previous (Selke and Yeomans) Monte Carlo result. By performing new MC calculations on systems with various length to width ratios we find that the critical adsorption at the first-order transition scales as $L_\perp f(L_\parallel/L_\perp^2)$, where $L_\parallel$ and $L_\perp$ are the lengths of the systems parallel and perpendicular to the interface.

It is hoped that the results may provide a framework for understanding experimentally observable two-dimensional interface adsorption phenomena which are expected to occur, e.g., at domain boundaries in adsorbed monolayers. Related work is in progress (Selke 1984b, Sega *et al* 1984).

References

Abraham D B and Smith E R 1982 *Phys. Rev.* B **26** 1480
Baxter R J and Pearce P A 1983 *J. Phys. A: Math. Gen.* **16** 2239
Blume M 1966 *Phys. Rev.* **141** 517
Cahn J 1977 *J. Chem. Phys.* **66** 3667
Capel H W 1966 *Physica* **32** 966
den Nijs M 1983 *Phys. Rev.* B **27** 1674
Diehl H W, Kroll D M, and Wagner H 1980 *Z. Phys.* B **36** 329
Fisher M E 1974 *Proc. Nobel Symp.* **24** 16
—— 1984 *J. Stat. Phys.* **34** 667
Fisher M E and Berker A N 1982 *Phys. Rev.* B **26** 2507
Huse D A 1982 *Phys. Rev. Lett.* **49** 1121
Huse D A and Fisher M E 1982 *Phys. Rev. Lett.* **49** 793
—— 1984 *Phys. Rev.* B **29** 239
Huse D A, Szpilka A M and Fisher M E 1983 *Physica* **121**A 363
Kroll D M and Lipowsky R 1983 *Phys. Rev.* B **28** 6435
Landau D P and Swendsen R H 1984 Private communication
Lawrie I and Sarbach S 1984 *Theory of Tricritical Points, vol 10 of Phase Transitions and Critical Phenomena,* ed by C Domb and J L Lebowitz (New York: Academic)
Lipowsky R and Gompper G 1984 *Phys. Rev.* B **29** 5213
Lipowsky R, Kroll D M and Zia RKP 1983 *Phys. Rev.* B **27** 4499
Moldover M and Cahn J 1980 *Science* **207** 1073
Nienhuis B 1982 *J. Phys. A: Math. Gen.* **15** 199
Riedel E K 1972 *Phys. Rev. Lett.* **28** 675
Onsager L 1944 *Phys. Rev.* **65** 117
Sega I, Selke W and Binder K 1984 in preparation
Selke W 1984a *Surface Sci.* **144** 176
—— 1984b *XXth Karpacz Winter School, Lecture Notes in Physics* vol 206 ed A Pekalski and J Sznajol (Heidelberg: Springer) p 191
Selke W and Huse D A 1983 *Z. Phys.* B **50** 113
Selke W and Pesch W 1982 *Z. Phys.* B **47** 335
Selke W and Yeomans J 1983 *J. Phys. A: Math. Gen.* **16** 2789
Teubner M 1983 *Preprint*
Widom B 1974 *J. Chem. Phys.* **62** 1332
—— 1978 *J. Chem. Phys.* **68** 3878