Fluids with quenched disorder: scaling of the free energy barrier near critical points

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Abstract
In the context of Monte Carlo simulations, the analysis of the probability distribution \( P_L(m) \) of the order parameter \( m \), as obtained in simulation boxes of finite linear extension \( L \), allows for an easy estimation of the location of the critical point and the critical exponents. For Ising-like systems without quenched disorder, \( P_L(m) \) becomes scale-invariant at the critical point, where it assumes a characteristic bimodal shape featuring two overlapping peaks. In particular, the ratio between the value of \( P_L(m) \) at the peaks \( (P_L)_{\text{max}} \) and the value at the minimum in between \( (P_L)_{\text{min}} \) becomes \( L \)-independent at criticality. However, for Ising-like systems with quenched random fields, we argue that instead

\[
\Delta F_L := \ln\left(\frac{P_L_{\text{max}}}{P_L_{\text{min}}}\right) \propto L^\theta
\]

should be observed, where \( \theta > 0 \) is the ‘violation of hyperscaling’ exponent. Since \( \theta \) is substantially non-zero, the scaling of \( \Delta F_L \) with system size should be easily detectable in simulations. For two fluid models with quenched disorder, \( \Delta F_L \) versus \( L \) was measured and the expected scaling was confirmed. This provides further evidence that fluids with quenched disorder belong to the universality class of the random field Ising model.

(Some figures in this article are in colour only in the electronic version)

1. Introduction
It was postulated by de Gennes that liquid–gas-type transitions in fluids confined to quenched random porous media belong to the universality class of the random field Ising model (RFIM) [1]. The unambiguous experimental verification of this conjecture remains elusive to this day [2], but there is growing numerical evidence from computer simulations [3–5] of quenched–annealed mixtures [6]. In quenched–annealed mixtures, a fluid of mobile particles is confined to a configuration of immobile (quenched) particles. The quenched particles induce a random field provided (1) their positions are sufficiently random and (2) they display a preferred affinity to one of the phases formed by the mobile particles. When either one of these conditions is not met, the conjecture of de Gennes does not apply [2, 7].

Computer simulations of quenched–annealed mixtures remain complicated. One problem is that for systems exhibiting quenched disorder an additional average over many samples drawn from the distribution characterizing the disorder needs to be taken. In addition to the thermal averaging, a disorder averaging over the different samples must be taken, and hence the computational effort is orders of magnitude larger. The convergence of disorder-averaged quantities with the number of samples is typically slow (random field systems at criticality do not self-average [8–13]) and so the disorder averaging must comprise many samples. A second problem is that the critical exponents of the three-dimensional RFIM are not known very precisely. In particular, estimates for the correlation length exponent \( \nu \) range from 1.1 to 2.25 [14]. Due to the large uncertainty in \( \nu \), standard finite-size scaling (FSS) of simulation data is problematic. The only critical exponent of the RFIM on which there is some consensus is the ‘violation of hyperscaling’ exponent \( \theta \). In pure systems, meaning systems without quenched disorder, it holds that \( \theta = 0 \), but in the RFIM \( \theta \sim 1.5 \), i.e. distinctly non-zero. In order to detect RFIM universality, it follows that FSS of a quantity ‘somehow involving’ the violation of hyperscaling exponent \( \theta \) is a promising approach. In this paper, we present one such strategy, based on the free energy cost of interface formation. In the RFIM at its critical point, the barrier diverges with the system size as

\[
\Delta F_L \propto L^\theta
\]

where \( L \) is the lateral extension of the simulation box [5]. In the pure Ising model where \( \theta = 0 \), \( \Delta F_L \) is \( L \)-independent at criticality [5, 15, 16]. Since \( \theta \) is large in the RFIM,
divergence of $\Delta F_L$ should be easily detectable in random field systems.

The outline of this paper is as follows. We first describe how the scaling of the free energy barrier at criticality can be derived and exploited in the context of FSS. We then illustrate the approach for the RFIM, the Widom–Rowlinson (WR) model [17] with quenched obstacles and the Asakura–Oosawa (AO) model [18, 19] of colloid–polymer mixtures inside a porous medium. In all cases $\theta > 0$ is observed, but the precise value varies. For the RFIM and WR mixture $\theta \sim 1.5$ is obtained, while colloid–polymer mixtures yield a somewhat lower value $\theta \sim 1.0$; possible origins for this discrepancy are discussed in section 4.

2. Finite-size scaling of the interface free energy

2.1. Theoretical background

We assume that the RFIM in $d = 3$ dimensions undergoes a continuous phase transition, at critical temperature $T_c$, from a disordered phase at high temperature to an ordered phase with finite magnetization at low temperature. The existence of a transition at non-zero temperature was controversial until a proof for the existence of a spontaneous magnetization settled this issue [20]; rigorous results of the order of the transition remain elusive, but numerical studies [21, 22] indicate that the transition is continuous. Following convention, we define the relative distance from the critical point as

$$ t := T/T_c - 1. $$

In the vicinity of the critical point, $t = 0$, the correlation length diverges as a power law

$$ \xi \propto |t|^{-\nu}, $$

while in the ordered phase a finite interfacial tension (excess free energy per unit of interface) develops:

$$ \sigma \propto |t|^{2-\alpha - \nu} \quad (t < 0), $$

where $\alpha$ is the critical exponent of the specific heat. Near the critical point, the correlation length is the only relevant length scale, and so a simple dimensional argument implies that the total interface free energy must scale as

$$ \Delta F_\xi \propto \sigma \xi^{d-1} \propto |t|^{2-\alpha - \nu} \xi^{d-1} \propto \xi^{(\alpha + d\nu - 2)/\nu}, $$

where in the last step $t$ was eliminated using equation (3). If we now use the FSS ‘ansatz’ $\xi \propto L$, the above equation reduces to

$$ \Delta F_L \propto L^{(\alpha + d\nu - 2)/\nu}, $$

which upon substitution in equation (6) yields

$$ \Delta F_L \propto L^n, $$

and so we have derived equation (1). In the pure Ising model $\theta = 0$, in which case the barrier does not depend on $L$ at the critical point (as is well known [15]). In the RFIM $\theta \sim 1.5$, implying a strong divergence with system size.

2.2. Order parameter distributions

We now explain how the barrier $\Delta F_L$ is measured in computer simulations of liquid–gas-type transitions. For simplicity, we first consider a single-component fluid in the absence of quenched disorder. The key quantity is the order parameter distribution (OPD):

$$ P_L(\rho|T, \mu) $$

defined as the probability to observe a state with particle density $\rho$. The OPD is measured in the grand canonical ensemble, at fixed temperature $T$ and chemical potential $\mu$, using a periodic $L \times L \times L$ simulation box [26, 27]. Below the critical temperature $T_c$ and at the coexistence chemical potential $\mu_{cx}$, the OPD becomes bimodal; the peak at low (high) density corresponds to the gas (liquid) phase. The coexistence chemical potential is obtained by maximizing the derivative of the average density with respect to $\mu$:

$$ \mu_{cx} : \partial \langle \rho \rangle / \partial \mu \rightarrow \text{max,} $$

with $\langle \rho \rangle = \int \rho P_L(\rho|T, \mu) d\rho$. Note that other choices to define $\mu_{cx}$ are also possible [28], most notably the ‘equal-weight’ rule [29], but equation (10) has the advantage that it remains well defined also when the peaks in the OPD overlap. The free energy barrier $\Delta F_L$ is encoded in the logarithm of the OPD; it corresponds to the average peak height of $\ln P_L(\rho)$, measured from the minimum ‘in between’ the peaks [30].

In athermal binary mixtures with mobile species A and B (but still without disorder), the analogue of equation (9) becomes

$$ P_L(\rho_A|z_A, z_B) $$

where $\rho_A$ is the density of the A particles, and where $z_A$ and $z_B$ are the fugacities of the A and B particles, respectively. By athermal we mean that the particle interactions are either hard-core or ideal, as is the case in the WR and AO models. Those mixtures correspond to a single-component fluid if one identifies $\rho \leftrightarrow \rho_A$, $T \leftrightarrow 1/z_B$ and $\mu \leftrightarrow \ln z_A$. The transition between a ‘gas phase’ with low $\rho_A$ and a ‘liquid phase’ with high $\rho_A$ is now driven by the fugacities. It occurs on the coexistence curve $z_A = z_{cx}(z_B)$, provided the fugacity of the B particles exceeds the critical fugacity $z_B > z_c$. On the coexistence curve, which again can be found using equation (10), the OPD becomes bimodal and allows the extraction of a free energy barrier $\Delta F_L$, see figure 1(a).

We now consider an athermal binary mixture in the presence of quenched disorder. We thus have mobile particles A and B as before, but also quenched obstacles which are
distributed at random locations at the start of each simulation after which they remain frozen. The A and B particles then diffuse through the quenched environment. From the set of all possible configurations of quenched disorder, we consider a finite number of $N$ configurations, and for each of them the OPD is measured. Instead of a single OPD as in equation (11), we thus have a set of distributions

$$P_{L,i}(\rho_A | z_B, z_A), \quad (12)$$

where $i = 1, \ldots, N$ labels the different configurations. Even though we are interested in the OPDs for different fugacities, it is sufficient to simulate only a few combinations; having simulated a system at some $(z_A, z_B)$, the OPDs for nearby fugacities can be obtained via histogram reweighting [31]. To facilitate an efficient storage of the data, the approach of [3, 5] is used. Meaningful results around the critical point require $N \sim 10^{3-4}$, so the computational effort is much larger compared to the pure systems.

### 2.3. Extracting the free energy barrier

To obtain the quenched-averaged free energy barrier, we extract the free energy barrier $\Delta F_{L,i}$ for each configuration separately, as in figure 1(a), and then average these values. To be precise: for a given $z_B$, each OPD is tuned to coexistence via equation (10), which implies that $z_{c3}$ varies between disorder configurations [3, 5]. By tuning the distributions separately, we ensure that the majority of them become bimodal, such that a barrier can indeed be ‘read-off’. From the individual barriers, the quenched-averaged barrier is obtained as a simple average:

$$\Delta F_L = (1 / N) \sum_{i=1}^{N} \Delta F_{L,i}. \quad (13)$$

The finite number of disorder configurations used in calculating the barrier gives rise to a statistical deviation:

$$u_L^2 := \frac{1}{\sqrt{N(N-1)}} \sum_{i=1}^{N} (\Delta F_{L,i} - \Delta F_L)^2 \quad (14)$$

which represents the expected deviation of our result from the result we were to obtain for $N \rightarrow \infty$.

In practice, obtaining the barrier $\Delta F_{L,i}$ can be problematic. For some disorder configurations, the OPD does not feature a clear liquid and gas peak under the criterion of equation (10). In these cases, an intermediate peak is seen at a density roughly between that of the liquid and gas. This extra peak can occur as a replacement for either the liquid or gas peak, or as an additional third peak (figure 1(b)). The existence of these ‘problematic’ distributions is due to cavities formed by the obstacles. In some disorder configurations, the obstacles create large regions with a local preference for either A or B particles. One can easily show that such regions induce an intermediate peak (figure 2). We emphasize that around the critical point such problematic distributions are scarce in the full ensemble of disorder configurations (although modified hyperscaling does allow a finite fraction of them [5]).

### 2.4. Analysis of finite-size effects: scaling plots

Near the critical point, we expect $\Delta F_L$ to scale according to equation (1). Following standard FSS practice [32, 33], we test equation (1) using scaling plots. That is, we plot $y_L := L^{-\theta} \Delta F_L$ versus $\tau := t L^{1/\nu}$, $t$ given by equation (2). Then, if the correct values for $\theta$, $\nu$ and $T_c$ are used, the curves $y_L(\tau)$ for different system sizes $L$ should collapse on top of each other. The practical difficulty with this method is assessing the quality of the collapse: seemingly good data collapses are obtained over a range of parameter values, and simple ‘eye gauging’ becomes unreliable. To obtain the parameters of best collapse,
as well as a measure of the reliability of the results, a numerical procedure was therefore used.

Given a candidate tuple \((\theta, \nu, T_c)\), equation (14) directly yields the statistical uncertainty in \(y_L(\tau)\), namely \(\delta y_L(\tau) = L^{-\theta} u_L\). We average \(\delta y_L(\tau)\) over the different system sizes to obtain the typical statistical uncertainty \(u(\tau)\) of the curves at a value \(\tau\). For a given \(\tau\), the curves \(y_L(\tau)\) for the different system sizes spread around some average value with a root-mean-square width \(\sigma(\tau)\). We then compare \(\sigma(\tau)\) to the typical statistical uncertainty \(u(\tau)\) via the ratio \(\sigma(\tau)/u(\tau)\). This ratio measures how strong the curves differ compared to the random scatter induced by the finite number \(N\) of obstacle configurations. To assess the quality of the collapse, we then compute the average of the ratio over some region of \(\tau\) around the critical point:

\[
R(\theta, \nu, T_c) := \frac{1}{\tau_1 - \tau_0} \int_{\tau_0}^{\tau_1} \frac{\sigma(\tau)}{u(\tau)} \, d\tau.
\]

Values \(R < 1\) are interpreted as fully consistent with a perfect collapse of the curves, while \(R > 1\) is considered less consistent. The integration range is chosen as large as possible, but small enough to stay within the critical region. We used \(\tau_0 = -0.2 \times 10^{-1/\nu}\) and \(\tau_1 = 0.2 \times 10^{-1/\nu}\), which assumes a typical system size \(L \sim 10\) and restricts the temperature range to \(|\tau| < 0.2\).

3. Results

3.1. RFIM

To provide a benchmark point for our analysis, we first apply our scaling test to data for the RFIM model. These data were taken from our previous work [5] and obtained for the RFIM in three dimensions using a random field drawn from a Gaussian distribution. The total number of disorder configurations per system size equals \(N \sim 10^4\). For system sizes \(L = 8, 10, 14\) and 16, we examine \(R(\theta, \nu, T_c)\) in the plane of critical exponents \((\theta, \nu)\). For each point in the plane, the critical temperature \(T_c\) is tuned such that \(R(\theta, \nu, T_c)\) is minimized; the value at the minimum is denoted \(R_{\text{min}}(\theta, \nu)\). The results are collected in figure 3 as contours. The region where \(R_{\text{min}}(\theta, \nu) \leq 1\), i.e. where the collapse is fully consistent, agrees with the expected value \(\theta \sim 1.5\). Also of interest is the region of good collapse features a tail extending towards the pure 3D Ising values \((\theta = 0, \nu \sim 0.63)\) [34]. The cause of this tail is not entirely clear, but we suspect it is due to crossover effects [35]. However, at the pure Ising values, the deviation between the curves \(y_L\) exceeds the statistical fluctuation due to the finite number of disorder configurations by a factor of more than three. We conclude that the pure Ising exponents are effectively excluded by our analysis. Note that
of a first-order transition, are effectively excluded.

The WR model [17] consists of unit-diameter spheres of species A and B. The only interaction is a hard-core repulsion between particles of the opposite species. We consider the WR model in the presence of quenched obstacles; the spatial interaction with mobile B (A) particles. In this work, systems of size $L = 7, 8, \ldots, 12$ are used, which on average contain 14, 20, \ldots, 69 obstacles, respectively. For each system size, OPDs for $N \sim 10^4$ disorder configurations were generated at fugacities $z_A = z_B = 1.4$, which is the approximate location of the critical point as anticipated from our previous simulations [5]; all details regarding the simulation method can also be found in that reference.

To obtain $\Delta F_L$ via equation (13) over a range of $z_B$ requires extracting the free energy barrier from several million(!) distributions, which obviously cannot be done by hand. To deal with the ‘problematic’ distributions of figure 1(b) automatically, a numerical filter was therefore applied: if one of the two dominating peaks in the OPD is centered in the intermediate density range $0.20z_B < \rho_A < 0.75z_B$, the distribution is considered to not feature a distinct liquid and gas peak. For these distributions we set $\Delta F_{L,i} = 0$. Since for the values of $z_B$ considered here no more than $\sim 0.1\%$ of the distributions failed this criterion, our results should not significantly depend on the filter. The results for $R_{\min}(\theta, \nu)$, as well as some estimates for the critical fugacity $z_c$ obtained from minimizing $R$, are shown in figure 4. Note that, for the WR model, the analogue of equation (2) becomes $t := z_c/z_B - 1$. As in the RFIM, figure 4 features a clear preference for $\theta \sim 1.5$ and the competing values $\theta = 0$ and $\theta = 2$ of, respectively, the pure Ising model and a first-order transition are effectively excluded. We interpret this finding as a strong sign that the WR model with quenched obstacles indeed belongs to the universality class of the RFIM. Note, however, that we cannot provide high-precision estimates of the critical exponents, nor of the critical ‘inverse temperature’ $z_c$. There is a substantial range of possible values $(\theta, \nu, z_c)$ in the region $R_{\min}(\theta, \nu) \leq 2$, i.e. the region where our results for the WR model and RFIM become quantitatively compatible.

### 3.3. Colloid–polymer mixtures in porous media

We now consider a colloid–polymer mixture confined to a porous medium. The colloids and polymers are unit-diameter spheres, with only a hard-core repulsion between colloid–colloid and colloid–polymer pairs (this is just the AO model [18, 19] with equally sized colloids and polymers). As the porous medium we use a quenched configuration of polymers, which are distributed randomly in a periodic cube of edge $L$ at the start of each simulation run; the average packing fraction of the quenched configuration $\eta_Q = 0.05$. We consider system sizes $L = 10–12$, which implies that the actual number of obstacles ranges from $N_Q = 95$ for the $L = 10$ system to $N_Q = 165$ for the $L = 12$ system. The OPD for the AO model is the probability distribution $P_{L,i}(\eta_{\text{col}}, \eta_{\text{pol}})$ [27], where $i$ denotes the quenched polymer configuration for which the OPD was measured, $\eta_{\text{col}}$ the colloid packing fraction, $\eta_{\text{pol}}$ the colloid fugacity and $\eta_{\text{pol}}^0$ the polymer reservoir packing fraction. Note that, for the AO model, $\eta_{\text{pol}}^0$ is just the polymer fugacity multiplied by the volume of a single polymer and thus plays the role of inverse temperature. Consequently, the analogue of equation (2) becomes $t := \eta_{\text{pol}}^0/\eta_{\text{pol}}^0 - 1$. One of us (RV) recently studied the above model using the same type of
quenched disorder. We now extend the analysis of that previous work [3] to the scaling of the free energy barrier.

One practical problem is that the number of disorder configurations in our AO dataset is ‘only’ $N \sim 10^5$, i.e. one order of magnitude below the value used in the analysis of the RFIM and WR models. Also the quality of individual distributions was worse compared to that of the latter two models. A further complication is that the AO model is asymmetric: the coexistence colloid fugacity is not trivially related to $\eta_c$. In contrast, for the WR model, it holds that $z_A = z_B$ at the critical point due to symmetry. Because of all these problems, the construction of contour plots, such as figures 3 and 4, was not feasible for the AO model, and so a modified analysis is presented. To this end, we extract the barrier $\Delta F_L$ from the quenched-averaged OPD, following the method outlined in the appendix of [5]. This procedure yields a result similar to equation (13), but it is less susceptible to statistical uncertainties in individual distributions. Since the barrier is extracted from the quenched-averaged OPD, we lose information about the fluctuations between disorder configurations. In particular, equation (14) can no longer be used to calculate the typical statistical uncertainty $u(\tau)$ defined in section 2.4. Hence, in the construction of the scaling plot $y_L(\tau)$, we cannot compare the deviation between the curves $\sigma(\tau)$ to the statistical uncertainty $u(\tau)$. Only best estimates can be provided, which are obtained by minimizing the relative deviations between the curves. That is, for a given $\tau$ we minimize the value $\sigma(\tau)/y(\tau)$, where $y(\tau)$ is the average value of the $y_L(\tau)$. For $-0.05 < \tau < 0.05$ and system sizes $L = 10–12$ we computed these best estimates: their variation with $\tau$ is shown in figure 5. If the collapse was perfect, the best estimates should not depend on $\tau$. Regarding $\eta_{pcr}$ and $\theta$ this is confirmed, and from the average of the curves we conclude that $\eta_{pcr} \sim 1.3$ and $\theta \sim 1.0$ for the AO model. However, it is clear from figure 5 that no meaningful estimate of $\nu$ can be provided. We emphasize that $\eta_{pcr} \sim 1.3$ deviates by about 10% from previous estimates. The reason is that, in [3], we assumed $\nu_{RFIM} = 1.1$. However, from the analysis of the RFIM (figure 3) it transpires that $\nu_{RFIM}$ may well be different. This is furthermore corroborated by the literature [14], where large variations in $\nu_{RFIM}$ are also reported.

4. Conclusions
The aim of this paper was to test the relation $\Delta F_L \propto L^{\nu}$ in fluids with quenched disorder. This relation applies when the universality class is that of the RFIM [5]. For the WR and the AO model with quenched obstacles, our data confirm that $\Delta F_L$ diverges as a power law with system size at the critical point. This is an important qualitative indication that the universality class is no longer that of the pure Ising model, since then $\Delta F_L$ would be constant at criticality. For the WR model, $\theta$ is consistent with the RFIM value $\theta_{RFIM} \sim 1.5$. For the AO model, $\theta \sim 1.0$ is found, which is somewhat below the RFIM value. Possible reasons for this discrepancy are (1) crossover effects [35] (in this case from pure Ising to RFIM universality), (2) corrections to scaling due to asymmetry and (3) insufficient disorder averaging. We believe that the crossover scenario is the most likely explanation, since $\theta \sim 1.0$ is ‘in between’ the pure Ising value $\theta = 0$ and the RFIM value.

The finite-size scaling analysis presented in this paper follows the standard ‘ansatz’ $\xi \propto L$, i.e. that close to the critical point a diverging correlation length is prevented by the finite box size. In the presence of quenched obstacles, a second source of finite-size effects is to be expected, which stems from the structures that can be formed by the random obstacles. Obviously, a single obstacle cannot induce any randomness. Also, for two obstacles, the variety of created structures is limited, to say the least. A proper treatment of corrections due to finite obstacle numbers poses an interesting theoretical challenge. However, such an analysis is beyond the scope of this work and all we can do is verify the consistency of our results. For the WR model, a large increase in the critical fugacity (from $z_c = 0.9379(4)$ for the pure model [36] to $z_c \sim 1.42$) and a de facto exclusion of the critical exponents of the Ising model are seen. Since in our analysis of the AO model even more obstacles were used, we believe that the number of obstacles used here is sufficient to demonstrate the change towards random field Ising universality. Another important issue in relation to finite-size scaling is that one has to take care that correlations between obstacles do not extend beyond the size of the box. Since, in this work, the obstacles are distributed completely randomly, spatial correlations are trivially absent and so this problem does not occur here. If spatial correlations in the obstacle configuration are long-ranged, for instance by placing them on the sites of a regular lattice, the random field hypothesis does not apply [37].

Of course, all these problems could be reduced by simulating larger systems and more disorder configurations, but the cost in CPU time is still prohibitively large. Our data also make it clear that high-precision estimates of critical exponents and temperatures in random field systems remain difficult to obtain in simulations. The data for the RFIM and the WR model reveal large variations, see figures 3 and 4, in particular for the correlation length critical exponent $\nu$.

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