

Binary fluids in Vycor: Anticorrelated random fields

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Binary fluids in Vycor are a realization of anticorrelated random-field systems. The change from uncorrelated to anticorrelated fields has a profound effect at zero temperature. At any finite temperatures an effective uncorrelated random field of entropic origin is generated and thus the large scale behavior is that of uncorrelated systems. The possible crossovers from the pure-system behavior to the eventual uncorrelated behavior and from an effective anticorrelated to uncorrelated behavior are discussed.

I. INTRODUCTION

The effects of quenched disorder on the thermodynamics properties of condensed systems have been at the focus of intensive research activity in recent years. From all possible disorder agents, random fields have particularly drastic effects due to the linear coupling between the disorder and the order parameter of the underlying system. As such they have been at the center of both experimental and theoretical investigations.¹⁻⁴ Relative to the large volume of these investigations the results so far are quite modest and most important problems are still open to heated debates. Undoubtedly these difficulties are associated with the exceptionally strong effect of the random fields. Therefore the study of systems in which the disorder still couples linearly to the order parameters but in which its effects may be weaker, will be especially interesting, since they have the potential to be more tractable theoretically and better understood experimentally.

This paper is devoted to the study of such systems. As we outline below these expectations are partly fulfilled although we find that the drastic effects of regular random fields are subtly generated in unexpected ways.

Let us begin by analyzing where such systems may be looked at experimentally. The major realizations of random-field systems originate in random antiferromagnets in an external magnetic field.⁵ However, other random field realizations have been investigated: binary fluids in presence of a gel or in porous media (like Vycor). Binary fluids are described in an Ising model in which the order parameter is the concentration difference between the two components in the fluid.^{6,7} As was pointed out by de Gennes, if there is a difference in the affinities of these two components to the gel (or to the Vycor) then

the latter will generate a local change in the energy which will be proportional to the density difference, namely there will be linear coupling between the gel (Vycor) and the order parameter.⁶ We are going to denote this coupling as a "field" because of its analogy to the magnetic field in the magnetic systems.

The important question is of course: Is this field "random"? The answer to this question depends on the properties of the quenched configurations of the gel or the Vycor. For the gel, de Gennes has argued^{6,7} that its structure is basically random beyond a finite scale. Below this scale the gel structure has some "persistence" and is not random. These small-scale correlations cannot change the behavior on the large scales and will thus be that typical to regular random-field (RF) systems.

The answer for the Vycor systems is totally different. Indeed the Vycor is formed from a deep-quenched phase separation in which the silica is phase separated from boron oxide. Before the phase separation is complete the melt is quenched to a lower temperature below the glass transition. The boron is leached out leaving a spongelike SiO₂ structure. As has been pointed out by Weichmann and Fisher⁸ (in their study of helium in Vycor) such a phase-separation process is controlled by the conservation of the silica (and the boron). Because silica cannot be created or annihilated but only exchange its location with the boron, the "conserved" spinodal decomposition leads to random structure with peculiar correlations exhibited by its structure factor $S(\mathbf{q})$ (the Fourier transform of its real-space correlations) in the limit $|\mathbf{q}| \rightarrow 0$. For conserved processes it always holds that⁹ $S(\mathbf{q}) \sim |\mathbf{q}|^2$. By contrast a completely random structure (which would have given rise to a regular RF) would have $S(\mathbf{q}) = \text{const}$ for small $|\mathbf{q}|$. So the behavior of $S(\mathbf{q})$ for $|\mathbf{q}| \rightarrow 0$ is very

different for both cases. Randomness with a correlation function which vanishes as $|\mathbf{q}| \rightarrow 0$ is expected to be weaker than that for which the correlation at small $|\mathbf{q}|$ are finite. This effect, which will be explained in detail below, is at the origin of our present theoretical explorations of such systems. Because diverging correlations arise from long-range correlation in the disorder, we call this situation in which $S(\mathbf{q})$ vanishes as $|\mathbf{q}| \rightarrow 0$ as “anticorrelated” random fields. Experimentally these systems have been studied by Wiltzius and co-workers^{10,11} as well as by Coh *et al.*¹² The analysis of these experiments was all based on the “random-field” paradigm¹³ and the essential differences that may arise due to $S(\mathbf{q}) \rightarrow 0$ have not been analyzed heretofore.

In the case of helium in Vycor⁸ the coupling of the Vycor is to the superfluid density which is quadratic in the order parameter (the superfluid amplitude $|\psi|^2$). Thus in this case, studied by Weichmann and Fisher,⁸ the disorder is equivalent to “random bonds” in the magnetic terminology and is expected to have weaker effects (in comparison with a binary fluid for which, as discussed above, the order parameter is the excess density itself).

Similar effective correlation functions in the randomness were found earlier in another type of system in which the randomness couples to the gradient of the order parameter: the so-called random vector field model.¹⁴ Realizations of such models include impurities effects on charge-density waves and directed models of percolation or transport in which the preferred direction changes randomly in space. Because the disorder couples linearly to the gradients, there is an extra q factor in the Fourier transform of the random coupling and an extra $|\mathbf{q}|^2$ for the two-point correlations of the disorder (compared with the regular random-field case). This model was partially analyzed in the past,¹⁴ and the results obtained agree, as particular cases, with the more general results derived here.

The organization of this paper is as follows: In Sec. II, we discuss the anticorrelated random field and the zero-temperature behavior. We find that the lower critical dimension and domain-wall roughening are drastically affected by the anticorrelation tuning parameter. To understand the low-temperature behavior, we apply a real-space, low-temperature, decimation procedure in Sec. III. We find that a weak uncorrelated random field of entropic origin is generated, thus obliterating the zero-temperature behavior and replacing it by the behavior typical to uncorrelated random-field systems. In Sec. IV, we discuss the crossover into the eventual uncorrelated random-field behavior. Section V discusses the crossover from the unattainable anticorrelated critical behavior to the uncorrelated behavior. Conclusions for the potential realizations are summarized in Sec. VI.

II. ANTICORRELATED RANDOM-FIELDS—ZERO-TEMPERATURE BEHAVIOR

The anticorrelated random field (ACRF) model has algebraic correlations as $q \rightarrow 0$ with a variable power $\mu > 0$ defined by

$$[h_q h_{-q}] \sim q^{2\mu} \quad (1)$$

for $q \rightarrow 0$. To generate such correlations in momentum space we choose the real-space correlations of the ACRF to be of the form

$$[h(\mathbf{r}')h(\mathbf{r}'+\mathbf{r})] = G(\mathbf{r}) = \begin{cases} -Kh^2/r^{d+2\mu} & |\mathbf{r}| \neq 0, \\ +h^2|\mathbf{r}|=0. \end{cases} \quad (2)$$

d is the spatial dimension and K is a constant determined by the anticorrelation condition $\sum_r G(r) = 0$. If this condition were not satisfied the properties would be the same as for uncorrelated fields. K vanishes as μ approaches zero. Therefore, the limit $\mu=0$ corresponds to uncorrelated random fields. Equation (2) implies for R much larger than the interatomic distance l

$$\sum_{r < R} G(r) \sim - \sum_{r > R} G(r) \sim \frac{h^2}{R^{2\mu}}, \quad (3)$$

and in Fourier space if $\mu < 1$,

$$[h_q h_{-q}] \sim h^2 q^{2\mu}. \quad (4)$$

A. The lower critical dimension

d_c may be obtained as in the pure case¹ by comparing the exchange and Zeeman energies due to flipping a bubble of linear size R relative to its surroundings. The average Zeeman energy is zero but the typical excess energy is given according to (3), by

$$[W_z^2]^{1/2} \sim \left[\sum_r \sum_{r'} h(r')h(r'+r) \right]^{1/2} \sim h[R^{d-2\mu} + R^{d-1}]^{1/2}, \quad (5)$$

where the second term between brackets is a surface contribution. The domain-wall energy¹ to be compared with (5) is Jr^{d-1} , thus

$$d_c = 2(1-\mu) \quad \text{if } \mu < \frac{1}{2}. \quad (6a)$$

For $\mu > \frac{1}{2}$ the random field does not affect the lower critical dimension d_c which is related to thermal solitons as in the case $h=0$. The result is

$$d_c = 1 \quad \text{if } \mu > \frac{1}{2}. \quad (6b)$$

Note that the d_c found here is related to the stability of the ferromagnetic ground state. We shall see that this is not the finite temperature d_c .

B. Domain-wall roughness at $T=0$

This question has been addressed in Refs. 2, 3, and 15–17 for uncorrelated fields. The simplest method¹⁵ is to treat a problem with a single degree of freedom, namely a bump of radius ρ and height h on a domain wall. The balance between the random field or Zeeman energy W_z (which tends to make h large) and the exchange energy W_{ex} (which keeps h small) determines the fluctuation of h , $\langle h^2 \rangle = h_1^2(\rho)$. If h_1 diverges with ρ , the wall is rough.

In the case of uncorrelated fields one uses the fact that the random-field energy change δW_z is the sum of a large

number of random variables, namely the random fields inside the bump. One therefore, expects that the absolute value of δW_z for a given value of h has, with a large probability, the order of magnitude $(\delta W_z^2)^{1/2}$ and this can actually be checked by an exact or a numerical calculation.¹⁶ If we accept this principle (to be discussed below) for anticorrelated fields too, one finds $\langle \delta W_z^2 \rangle = \sum_{rr'} G(\mathbf{r}-\mathbf{r}')$, where the sum is over \mathbf{r} and \mathbf{r}' , within the bump. For $\mu < 1/2$ the dominant contribution to this sum comes from points \mathbf{r}, \mathbf{r}' far from the wall and one obtains, using (3)

$$W_z^2 \sim \rho^{d-1} \int_0^{h_1} dz \int_{r' > z} d^d r' G(\mathbf{r}) \sim \rho^{d-1} \int_0^{h_1} dz \frac{h^2}{z^{2\mu}}, \quad (7)$$

$$W_z^2 \sim h^2 \rho^{d-1} h_1^{1-2\mu} \quad (\mu < \frac{1}{2}).$$

This bump-favoring energy is equilibrated by a bump-opposing exchange energy δW_{ex} . The latter will be evaluated in the continuum version for the Ising model. In that model^{2,3,15}

$$\delta W_{\text{ex}} \sim J \rho^{d-1} (h_1/\rho)^2. \quad (8)$$

Minimization of $(\delta W_z + \delta W_{\text{ex}})$ with respect to h_1 yields for large ρ

$$h_1^{3+2\mu} \sim (h/J)^2 \rho^{5-d} \quad (\mu < \frac{1}{2}). \quad (9)$$

The uncorrelated analog of the above simple argument^{15,17} has been also substantiated numerically.¹⁶

In $d=2$ dimensions the domain-wall roughness in the present model was considered by a one-loop renormalization-group study via the Burger's equation.^{18,19} It turned out, that the result (9) is correct even for $\mu < \mu_c(d=2) = \frac{3}{4}$. For $\mu > \mu_c$, $h_1 \sim \rho^\xi$ with $\xi = \frac{2}{3}$, independent of μ . If this scenario applies also for higher dimensions, one expects that the critical value $\mu_c(d)$ increases slightly from 2 to 5 dimensions with $\mu_c(d=5) \approx 0.9$. Here we used the result of the functional renormalization group²⁰ for the roughness exponent $\xi \approx 0.2083$ ($5-d$) of the random bond model, which is believed to be valid for $\mu \geq \mu_c(d)$ and $d \leq 5$. For $\mu < \mu_c(d)$, the result (9) is correct. The critical dimensions $d_R=5$, below which domain walls are rough, is independent of μ .

III. LOW-TEMPERATURE RENORMALIZATION: GENERATION OF NONCORRELATED RF

In this section we show how the thermal fluctuations of the spin degrees of freedom at finite temperature lead to the generation of regular random fields on larger scales. To see how the anticorrelated field is renormalized let us consider an explicit two-dimensional example.

Consider the system described by the following anticorrelated random-field Hamiltonian:

$$H = H_0 + \sum_{i \in \Omega} h_i \left[\sigma_i - \frac{1}{4} \sum_{n_i} \sigma_{n_i} \right], \quad (10)$$

where Ω is a partial set of the square lattice of points scattered at some low density and the summation over n_i is over the neighbors of i , the h_i 's are taken to be random and uncorrelated. H_0 is the usual ferromagnetic Hamiltonian. We apply now a decimation procedure based on integrating over the spins on the sites belonging to Ω .

The renormalized dimensionless Hamiltonian is

$$H' = H'_0 + \Delta H - \frac{1}{4} \sum_{i \in \Omega} h_i \sum_{n_i} \sigma_{n_i}, \quad (11)$$

where H'_0 is that part of H_0 that is independent on the spins belonging to Ω and

$$\Delta H = - \sum_{i \in \Omega} \ln \text{tr}_{(\sigma_i)} e^{J \sigma_i (\sum_{n_i} \sigma_{n_i} - h_i)} \equiv - \sum_i \Delta H^i. \quad (12)$$

The last relation stems from the fact that the density of points belonging to Ω is low. [In fact they can be chosen from the beginning in such a way that any two sites belonging to Ω are beyond next-nearest neighbors and then the last equality in (12) is exact.] We choose a given i denote it by 0 and its neighbors by 1,2,3,4 and calculate

$$\begin{aligned} \Delta H^0 &= - \ln \text{tr}_{(\sigma_0)} e^{J \sigma_0 (\sigma_1 + \sigma_2 + \sigma_3 + \sigma_4 - h_0)} \\ &= A + B [\sigma_1 \sigma_2 + \sigma_1 \sigma_3 + \dots \sigma_3 \sigma_4] \\ &\quad + C \sigma_1 \sigma_2 \sigma_3 \sigma_4 + F_1 (\sigma_1 + \sigma_2 + \sigma_3 + \sigma_4) \\ &\quad + F_2 \sigma_1 \sigma_2 \sigma_3 \sigma_4 (\sigma_1 + \sigma_2 + \sigma_3 + \sigma_4), \end{aligned} \quad (13)$$

where A, B, C, F_1 , and F_2 are explicit functions of J and h . We present here only F_1 and F_2 that break the inversion symmetry.

$$\begin{aligned} F_1 &= -\frac{1}{16} \{ [\ln \cosh(4J - h_0) - \ln \cosh(4J + h_0)] \\ &\quad + 2[\ln \cosh(2J - h_0) - \ln \cosh(2J + h_0)] \}, \end{aligned} \quad (14)$$

$$\begin{aligned} F_2 &= -\frac{1}{16} \{ [\ln \cosh(4J - h_0) - \ln \cosh(4J + h_0)] \\ &\quad - 2[\ln \cosh(2J - h_0) - \ln \cosh(2J + h_0)] \}. \end{aligned} \quad (15)$$

At low temperature $\sigma_1 \sigma_2 \sigma_3 \sigma_4$ may be taken as 1. (In fact we have calculated the correction to that and it does not affect the leading order of the final result.) We are therefore left with a field on the points 1,2,3,4, h_{eff} , given by

$$\begin{aligned} h_{\text{eff}} &= -\frac{1}{8} \{ \ln \cosh(4J - h_0) - \ln \cosh(4J + h_0) \} - \frac{1}{4} h_0 \\ &\simeq -\frac{1}{4} e^{-8J} \sinh(2h_0) \simeq -\text{sign}(h_0) \frac{1}{8} e^{-8J} e^{2|h_0|} \end{aligned} \quad (16)$$

for $J \gg h_0 \gg 1$. We see the appearance of the same field h_{eff} on the sites 1,2,3,4 and this field is uncorrelated with the fields generated on sites surrounding any other site in Ω . For $|h_i| \ll J$, the effect of the fields on the couplings A, B, C is negligible and therefore we could have obtained H' by starting with

$$\tilde{H} = H_0 + \sum_{i \in \Omega} h_{\text{eff}}^i \sum_{n_i} \sigma_{n_i}, \quad (17)$$

namely, a system of correlated (rather than anticorrelated) fields with short-range correlation.

The origin of this random uncorrelated field is purely entropic as may be seen from the fact that for the fixed ratio $|h|/J$ it approaches zero as J tends to infinity (the temperature tends to zero). Our particular system has a ferromagnetic ground state, while it is disordered at any finite temperature, due to the effective uncorrelated field. (Note that we consider here a two-dimensional system.)

$$\Delta'(q) = \Delta(q) + Au^2 \int \int \int dq_1 dq_2 dq_3 \delta(q - q_1 - q_2 - q_3) \Delta(q_1) G_0^2(q_1) \Delta(q_2) G_0^2(q_2) \Delta(q_3) G_0^2(q_3). \quad (18)$$

where $G_0(q) \sim 1/(r + q^2)$ is the unperturbed propagator.

The small-scale fluctuations are integrated out by the explicit integration over q_1, q_2 , and q_3 in a momentum shell $\pi/a < q_i < \pi/2a$. Clearly for $q=0$ there will be a nonzero contribution from this integral. The dependence on the rescaling factor is also easily estimated.

For the first (trivial term) $\Delta(q) = \Delta_0 q^{2\mu}$ the rescaling $q \rightarrow bq$ yields a dependence of $b^{2\mu}$. In the second term the power counting for $q=0$ yields a dependence of $u^2 \Delta_0^3 b^{2d-12+6\mu}$. Since the upper critical dimension is $d^* = 6 - 2\mu$ this may be expressed as $u^2 \Delta_0^3 b^{2\mu} b^{2\epsilon}$ ($\epsilon = d - d^*$). So in total we have to this order

$$\Delta(q) = b^{2\mu} [Au^2 \Delta_0^3 b^{2\epsilon} + \Delta_0 q^{2\mu}]. \quad (19)$$

The q -independent term is proportional to Δ_0^3 . This means that the effective uncorrelated field is proportional to the third power of the amplitude of the original correlated field. In particular the crossover variable to the usual RF behavior is $[h^2]t^{-\gamma}$ (γ is the susceptibility exponent of the pure system) and, since $h^2 \sim \Delta_0^3$, the crossover variable in all thermodynamic functions will be $\Delta_0 t^{-\gamma/3}$ namely, the crossover exponent is $\gamma/3$. This is the crossover exponent from the pure to eventual random-field exponent.

V. CROSSOVER FROM ANTICORRELATED TO UNCORRELATED CRITICAL BEHAVIOR

As seen from both renormalization procedures an uncorrelated random field is generated from the anticorrelated field and therefore the critical behavior to be expected is that of the uncorrelated field.²⁰⁻²² The effective uncorrelated field is much weaker than h that measures the strength of the anticorrelated field. Therefore it may be expected that the critical behavior observable near the transition line in the (h, J) plane, as a function of the distance to the line will first be that of the unattainable anticorrelated behavior and only closer to the line will it crossover to the behavior of the uncorrelated random field. The condition for the existence of this "intermediate phase" is that the anticorrelated fields as such are a relevant perturbation when added to the pure system.

IV. CROSSOVER TO THE UNCORRELATED RF BEHAVIOR

We have seen by real-space decimation that due to thermal fluctuations an uncorrelated RF is generated. To study the crossover behavior due to this effect we explore it in momentum space. We follow standard renormalization-group procedures to find the lowest order in u and Δ at which this RF is generated. (u is the standard ϕ^4 coupling constant.)

The lowest order (besides the trivial term) is given by the following expression ($\Delta(q) = [h_q h_{-q}]$),

Otherwise, if this perturbation is irrelevant there will be no intermediate phase and the crossover will be directly to the uncorrelated RF dominated behavior. The crossover exponent from the pure system due to the anticorrelated fields is $\phi = 2 - 2\mu - \eta_0$, where η_0 is spin-spin correlation exponent of the pure system. The intermediate phase will exist for $\phi > 0$. Note that for the Vycor system with $\mu = 1$ $\phi = -\eta$ and, since $\eta > 0$ for Ising systems, the transition will be directly to RF behavior without this intermediate phase. For the case $\phi > 0$ we define the effective exponents $\eta, \bar{\eta}$ and σ by the behavior of the susceptibility, correlation function, and Zeeman energy as a function of the correlation length ξ , where ξ can still be made very large.

$$\chi \sim \xi^{2-\eta}, \quad (20)$$

$$\langle m(0)m(\xi) \rangle \sim \xi^{d-4+2\mu+\bar{\eta}}, \quad (21)$$

and

$$W_z^2 \sim h^2 \xi^{d-2\mu-2\sigma} \quad \text{for } \mu \text{ not too large.} \quad (22)$$

The Arrhenius law yields for relaxation times

$$\begin{aligned} \tau &= \tau_0 \exp[a \xi^{d/2-\mu-\sigma}] \\ &= \tau_0 \exp[C(T - T_c)^{-\nu(d/2-\mu-\sigma)}]. \end{aligned}$$

Going through the same arguments discussed by Villain¹⁷ and Fisher²⁰ for uncorrelated fields we obtain the following scaling relations for the effective exponents:

$$\gamma = (2 - \eta)\nu, \quad (23)$$

$$\eta = \frac{d}{2} + \bar{\eta} + \mu - 2 - \sigma, \quad (24)$$

and

$$2 - \alpha = \left[\frac{d}{2} + \sigma + \mu \right] \nu. \quad (25)$$

Following Ref. (21) we also obtain that $\bar{\eta}$ and η are not independent but

$$\bar{\eta} = 2\eta. \quad (26)$$

(For the uncorrelated system the same result has been obtained to all orders in ϵ in a $2+\epsilon$ expansion.²²) Modified hyperscaling follows:

$$2-\alpha=(d-2+\eta+2\mu)\nu. \quad (27)$$

(For $\mu=0$ the same result was already obtained by Nattermann²³ and in a different language by Grinstein a long time ago.²⁴) Because of the μ dependence of the scaling relations it is clear that indeed the effective exponents are different from the true exponents.

When can we expect the crossover to occur? A crude answer may be obtained by equating the Zeeman energies of domains of size ξ of the anticorrelated field with that of the effective correlated field. Taking into account that $h_{\text{eff}} \sim h^3$ we obtain

$$h\xi^{d/2-\mu-\sigma_\mu} \sim h^3\xi^{d/2-\sigma_0}, \quad (28)$$

so that the correlation length at crossover obeys

$$\xi \sim (h)^{-2/(\mu+\sigma_\mu-\sigma_0)}. \quad (29)$$

Taking into account, that usually the σ 's are small $\xi \sim (h)^{-2/\mu}$. This is the size of the correlation when the systems starts to see the effective uncorrelated field.

VI. CONCLUSIONS

In this work we have explored the effects of anticorrelated random fields which couple linearly to the order pa-

rameter but are weaker than noncorrelated random fields. Our intention was to learn more on general random fields by the study of a controllable weaker perturbation. This study led us, however, to the conclusion that anticorrelated random fields generate noncorrelated RF at any finite temperature. So the anticorrelated RF, strictly speaking, is exhibited only at $T=0$. At finite temperatures the noncorrelated RF generated may be very weak and an "effective" behavior may be observed on relatively large length scales. Eventually the regular RF will dominate. The crossover exponent to this behavior is $\gamma/3$, namely third of the RF crossover exponent.

For binary fluids in Vycor the observed behavior must be that of regular (noncorrelated random fields) but with very strong corrections to scaling due to the fact that these noncorrelated fields are generated on large scales from coupling of the local fields with $S(q) \propto q^2$ due to the thermal fluctuations of the underlying order parameter. The effects should be seen as described above in the crossover from pure system behavior and are therefore to be expected not much below the critical temperature of the bulk (pure) system. The actual details would of course depend on the properties of the fluids and the porous medium.

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