Stable phase separation and heterogeneity away from the coexistence curve

T. R. Kirkpatrick and D. Belitz

Phys. Rev. B **93**, 144203 — Published 19 April 2016

DOI: 10.1103/PhysRevB.93.144203

Stable phase separation and heterogeneity away from the coexistence curve

T.R. Kirkpatrick[1] and D. Belitz[2]
[1]*Institute for Physical Science and Technology, University of Maryland, College Park, MD 20742*
[2]*Department of Physics, Institute of Theoretical Science,*
and Materials Science Institute, University of Oregon, Eugene, OR 97403

(Dated: February 21, 2016)

Phase separation, i.e., the coexistence of two different phases, is observed in many systems away from the coexistence curve of a first-order transition, leading to a stable heterogeneous phase or region. Examples include various quantum ferromagnets, heavy-fermion systems, rare-earth nickelates, and others. These observations seem to violate basic notions of equilibrium thermodynamics, which state that phase separation can occur only on the coexistence curve. We show theoretically that quenched disorder allows for phase separation away from the coexistence curve even in equilibrium due to the existence of stable minority-phase droplets within the majority phase. Our scenario also answers a related question: How can a first-order transition remain sharp in the presence of quenched disorder without violating the rigorous lower bound $\nu \geq 2/d$ for the correlation-length exponent? We discuss this scenario in the context of experimental results for a large variety of systems.

PACS numbers: 05.30.Rt; 64.75.-g

I. INTRODUCTION

Phase separation, i.e., the coexistence of two different phases in thermodynamic equilibrium, is a hallmark of first-order transitions. It follows from basic thermodynamics that this phenomenon can occur only on the coexistence curve, where the two phases have the same free energy.[1] However, in many solid-state systems phase separation is observed by a variety of techniques – muon spin rotation (μSR), nuclear magnetic resonance (NMR), nuclear quadrupole resonance (NQR), neutron depolarization imaging, and neutron Larmor diffraction – even away from a coexistence curve. Examples include quantum ferromagnets and helimagnets such as MnSi,[2,3] $\mathrm{Sr}_{1-x}\mathrm{Ca}_x\mathrm{RuO}_3$,[2,4] and UGe_2,[5,6] heavy-fermion systems such as $\mathrm{CeCu}_{2.2}\mathrm{Si}_2$,[7] high-$\mathrm{T_c}$ superconductors such as Europium-doped $\mathrm{La}_{1.85}\mathrm{Sr}_{0.15}\mathrm{CuO}_4$,[8] and systems displaying Mott transitions such as the rare-earth nickelates.[9,10] In some of these systems the first-order transition is from an ordered phase to a disordered phase (e.g., the ferromagnet-to-paramagnet transition in $\mathrm{Sr}_{1-x}\mathrm{Ca}_x\mathrm{RuO}_3$,[4] or the transition from an antiferromagnetic insulator to a paramagnetic metal in the nickelates[9]), in others, it is between two phases with the same order parameter (e.g., the FM1-FM2 transition between two ferromagnetic phases in UGe_2.[6]) Some phase diagrams contain a tricritical point, in others the transition is first order for all accessible parameter values, and in still others a line of first-order transitions ends in a critical point; schematic observed phase diagrams are shown in Fig. 1. Phase separation is observed on either side of the transition, but the experimental evidence is clearer in the ordered phase, and in some cases no phase separation has been observed so far in the disordered phase. In some of these systems there is independent evidence for the transition being first order, e.g., in the helical magnet MnSi,[11] in others the observed phase separation is used as *prima facie* evidence for the first-order nature of a nearby phase transition.

These experiments and their interpretations raise a fundamental question: How can stable phase separation occur away from the coexistence curve, where the two phases necessarily have different free energies? Surprisingly, this question does not seem to have been addressed so far, although it is crucial for an understanding and in-

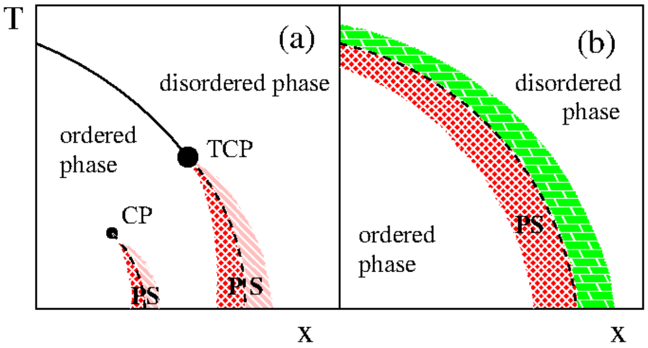

FIG. 1: Schematic phase diagrams in the temperature (T) - control parameter (x) plane. (a) A tricritical point (TCP) separates a line of second-order transitions (solid line) from a line of first-order transitions (dashed line). Phase separation (PS) is observed in both the cross-hatched (red) and hatched (pink) regions, although some experiments show PS clearly only in the ordered phase. Examples are the helical magnet MnSi[2,12] and the ferromagnet UGe_2[5], with hydrostatic pressure as the control parameter. Inside the ordered phase another first-order transition and associated PS may be present, with the line of first-order transitions ending in a critical point (CP). An example is UGe_2[6]. (b) The transition is first order for all values of the control parameter. An example is the rare-earth nickelate $\mathrm{Nd}_{1-x}\mathrm{La}_x\mathrm{NiO}_3$, with the dopant concentration as the control parameter[9,13]. PS in the tiled (green) region in the disordered phase is expected, but has so far not been observed.

terpretation of the experiments mentioned above. We can think of only two possible explanations: Either these systems are not in true thermodynamic equilibrium,[14] or they contain quenched disorder that couples to the order parameter and leads to the existence of static droplets of the minority phase within the majority phase. The first option would require a non-equilibrium state with a very long relaxation time (at least years), since phase separation has been observed in samples for which a first-order transition had been reported much earlier.[2,11] While this is not inconceivable, it seems implausible that such a state would not also lead to other observable consequences, e.g., glass-like features. We therefore focus on the second possibility. We will show that a disorder-induced droplet scenario, namely, the existence of static minority-phase droplets within the majority phase, leads to a consistent theoretical picture that is in agreement with the experimental observations for a large class of systems.[15] In particular, it can explain the deviation of the ordered-volume fraction from unity away from the coexistence curve, and the observed asymmetry between the ordered and disordered phases. It assumes the existence of quenched disorder that couples to the order parameter, but is not necessarily reflected in transport experiments, as some of the systems in question are rather good metals. We will come back to this assumption in the discussion.

The organization of this paper, and its main achievements, are as follows. In Sec. II we use established results about first- and second-order transitions in the presence of quenched disorder to show that disorder-induced droplets can exist near first-order transitions, but not near second-order ones. In Sec. III we establish criteria for the stability of minority-phase droplets. We show that they are stable in a sizable region of the phase diagram with reasonable parameter values, and we estimate the ordered-volume fraction in the ordered phase as a function of the distance from the coexistence curve. In Sec. IV we discuss our results and their relation to experiments that have already observed the phenomena for which this paper provides a physical explanation.

II. PHASE TRANSITIONS IN THE PRESENCE OF QUENCHED DISORDER

We start with some general considerations regarding second- and first-order transitions in the presence of quenched disorder that couples to the order parameter. Let us first recall the Harris criterion for the critical behavior at a second-order transition to be unaffected by quenched disorder. Let t be the dimensionless distance from the critical point, and ν the correlation-length exponent. Then the correlation length ξ scales as $\xi \sim t^{-1/\nu}$, or $t \sim \xi^{-1/\nu}$.[1,16] Quenched disorder leads to an uncertainty in t. By the law of large numbers, this uncertainty will fall off as the inverse square root of the system volume. Over a correlation volume, it thus obeys $(\Delta t)_{\text{dis}} \sim \xi^{-d/2}$, where d is the spatial dimensionality. In order for the transition to not be affected by the disorder, the uncertainty Δt must be smaller than t itself. This leads to the condition[17]

$$\nu \geq 2/d \ . \tag{2.1}$$

In Harris's original argument[17] this condition referred to the renormalization-group fixed point that describes the critical point in a clean system. If it is violated, i.e., if $\nu < 2/d$ in the *clean* system, then disorder either modifies the critical behavior or destroys the transition. No statement could be made at the time about ν at the new fixed point, if any, that describes the transition in the presence of disorder. Later, Chayes et al.[18] proved rigorously that Eq. (2.1) must hold at any fixed point that describes the physical critical behavior in the presence of quenched disorder.

Now consider a second-order transition in a finite system of linear size L (which can be a subsystem of a larger system) that is large compared to the microscopic length scale a, $L \gg a$. In the vicinity of the transition, the order-parameter susceptibility χ (among other observables) obeys a homogeneity law[19]

$$\chi(t,L) = b^{\gamma/\nu}\, F_\chi(t\, b^{1/\nu}, L\, b^{-1}) = t^{-\gamma}\, F_\chi(1, L\, t^{\nu}) \ . \tag{2.2}$$

Here b is an arbitrary scale factor, and γ is the susceptibility exponent. The phase transition thus gets rounded on a scale $(\Delta t)_{\text{rounding}} \sim L^{-1/\nu}$, while disorder fluctuations lead to an uncertainty (see above) $(\Delta t)_{\text{dis}} \sim L^{-d/2}$. We now ask whether it is possible to form a droplet of size L that contains a distinguishable phase different from the majority phase. This requires $(\Delta t)_{\text{rounding}} < (\Delta t)_{\text{dis}}$, or

$$\nu < 2/d \ . \tag{2.3}$$

This means that the existence of droplets is incompatible with the lower bound on ν given by Eq. (2.1). We conclude that disorder-induced droplets cannot exist in the vicinity of a second-order transition.

Near a first-order transition, the situation is qualitatively different. As was shown by Fisher and Berker (Ref. 20, see also Ref. 21), finite-size scaling considerations yield

$$\nu = 1/d \tag{2.4a}$$

for the correlation-length exponent at *any* thermal first-order transition. This has been generalized to quantum phase transitions.[22,23] It was shown that at $T = 0$ Eq. (2.4a) gets generalized to

$$\nu = 1/(d + z_{\text{OP}}) \ , \tag{2.4b}$$

where $z_{\text{OP}} > 0$ is the dynamical critical exponent that governs the temperature scaling of the order parameter.[23] The rounding scale thus crosses over from $(\Delta t)_{\text{rounding}} \sim L^{-d} \ll L^{-d/2}$ in the classical case to $(\Delta t)_{\text{rounding}} \sim L^{-(d+z_{\text{OP}})} \ll L^{-d/2}$ at $T = 0$, and rounding does not preclude the existence of droplets at any temperature. This striking difference between first- and second-order transitions gives a first indication of the physics behind the observed phase diagrams.

To avoid misunderstandings we emphasize that the droplets we consider are not rare regions. The length scale we consider is set by the correlation length, so we are considering typical fluctuations rather then rare ones. We also stress that we are not discussing nucleation phenomena; the droplets we consider are static in nature.

Let us now consider the energies relevant for the stability of droplets near a first-order phase transition. Consider a (sub)system of linear size L and volume L^d. Consider two states with free-energy densities f_1 and f_2, respectively. Suppose a disorder fluctuation causes the lowest free-energy state in half of the system to be state 1, and in the other half, state 2. The system can take advantage of this fluctuation by going into a phase-separated state, but this will incur an energy cost in the form of a interface energy. For an order parameter with Ising symmetry, or for any n-vector model where the coupling of the order parameter to the underlying lattice breaks the $O(n)$ symmetry, however weakly, the interface energy is proportional to the surface area L^{d-1}. With σ the surface tension, the free energy of the phase-separated state is

$$F_{\rm ps} = f_1\, L^d/2 + f_2\, L^d/2 + \sigma\, L^{d-1} \ , \tag{2.5a}$$

whereas the homogeneous state has a free energy

$$F_{\rm hom} = f_1\, L^d \ . \tag{2.5b}$$

Now the free energy in a system with quenched disorder is a random variable, and hence its root-mean square deviation is $\langle(\Delta F)^2\rangle^{1/2} \propto L^{d/2}$. If we take this to be representative of the free-energy difference $(f_1 - f_2)L^d$, then we have $(f_1 - f_2)L^d = 2\Delta\, L^{d/2}$, with Δ a measure of the quenched disorder.[24] The free-energy difference $\Delta F = F_{\rm ps} - F_{\rm hom}$ between the two phases then becomes

$$\Delta F = -\Delta\, L^{d/2} + \sigma\, L^{d-1} \ . \tag{2.6}$$

If $d < 2$, we have $\Delta F < 0$ for any sufficiently large L. There thus is no energy barrier precluding the existence of arbitrarily many droplets, and the first-order transition will be smeared. However, if $d > 2$, then the phase-separated state has a lower free energy than the homogeneous one only if L is smaller than a critical value L_c given by

$$L_c = (\Delta/\sigma)^{2/(d-2)} \tag{2.7}$$

For $d > 2$, the largest possible linear droplet size is thus given by L_c.

The arguments related to Eqs. (2.5) - (2.7) are very similar to those given by Imry and Wortis,[25] which in turn relied on a study of the random-field problem by Imry and Ma.[26] These authors, and many others that used their ideas, focused on instabilities of phases with long-range order, and on the existence or otherwise of a sharp phase transition. While we will comment on the latter aspect below, our main focus is on a different aspect of the same arguments, namely, the idea that droplets of the "wrong" phase be energetically stabilized inside the "right" phase by disorder fluctuations as long as their size does not exceed a critical value. In this sense the two phases will coexist away from the coexistence curve. The arguments say nothing about the region of the phase diagram where droplets can be expected, and *a priori* it is not clear whether the allowable droplet size makes them realizable. In what follows we will explore this scenario in more detail. Specifically, we will explore how L_c depends on the distance from the coexistence curve, what other length scales are relevant for the problem, and what the resulting volume fraction of the majority phase is expected to be. For definiteness, we will consider $d = 3$, and we will consider droplets with no order inside the ordered phase. We will discuss the issue of ordered droplets within the disordered phase in Sec. IV.

III. CONDITIONS FOR THE EXISTENCE OF DROPLETS

A. Length scales, and probabilities

There are various length scales in addition to the microscopic length a that enter the problem, viz.: (1) The largest linear size a region or "droplet" favoring the minority phase can have and still occur with a probability that is not exponentially small. This we will denote by L^*; it is the largest size of typical regions, as opposed to rare regions. (2) The largest size a droplet can have and still be energetically favorable, taking into account both the energy gain due to the disorder fluctuation and the surface energy cost. This we will denote by L_c, as in Eq. (2.7). (3) The minimum size a droplet must have in order to support an identifiable distinct phase. This we will denote by L_0. (4) The thickness of the droplet wall, which is important for determining the surface tension σ. This we will denote by $L_{\rm dw}$. (5) In systems with a weakly broken continuous symmetry there are Goldstone modes. Let their frequency-momentum relation in the long-wavelength limit be $\Omega(k) = s\, k^\omega$, with s a stiffness parameter. The breaking of the symmetry gives the Goldstone modes a gap Ω_g, which corresponds to a length scale $L_g = (s/\Omega_g)^{1/\omega}$.[27] In an Ising-like system, $L_g \approx a$.

In order to discuss and estimate these characteristic length scales, consider a field theory with a random-mass term, i.e., an action whose Gaussian part reads[28]

$$\mathcal{A}^{(2)} = \int_V d\boldsymbol{x}\, \phi(\boldsymbol{x}) \left[r + \delta r(\boldsymbol{x}) - c\boldsymbol{\nabla}^2\right] \phi(\boldsymbol{x}) \ . \tag{3.1}$$

Here $\phi(\boldsymbol{x})$ is the order-parameter field. For simplicity, we consider a scalar order parameter, we will consider the n-vector case below. V is the system volume, and r and c are parameters of the Landau-Ginzburg-Wilson functional $\mathcal{A}^{(2)}$. $\delta r(\boldsymbol{x})$ is a random variable governed by a distribution with zero mean and second moment

$$\langle \delta r(\boldsymbol{x})\, \delta r(\boldsymbol{y}) \rangle = \rho\, \delta(\boldsymbol{x} - \boldsymbol{y}) \ , \tag{3.2}$$

which defines ρ. For simplicity, we assume δr to be delta-correlated; we will come back to this assumption below. If we take the order parameter to be dimensionless, then r will be an energy density. Let J be the energy scale relevant for the order described (at a thermal phase transition, J will be on the order of the transition temperature T_{c}). Then we expect, up to dimensionless factors, $c \approx J/a$, $r \propto J/a^3$, and

$$\rho \approx \delta\, J^2/a^3 \ , \tag{3.3}$$

where δ is a dimensionless measure of the disorder. Weak disorder corresponds to $\delta \ll 1$, and very strong disorder corresponds to $\delta \approx 1$. Here, and throughout our discussion, we ignore dimensionless factors that qualitative arguments give no control over.[29]

1. *The length scale L^**

In order to estimate the length scale L^*, we consider δr coarse-grained over a volume L^3 by defining

$$\delta r_L := \frac{1}{L^3} \int_{L^3} \delta r(\boldsymbol{x}) \ . \tag{3.4}$$

δr_L is an average of independent random variables, so by the central limit theorem it is Gaussian distributed,

$$\mathcal{P}(\delta r_L) = \frac{1}{\sqrt{2\pi} s}\, e^{-(\delta r_L)^2/2s^2} \tag{3.5a}$$

with second moment

$$s^2 = \frac{\rho}{L^6} \int_{L^3} d\boldsymbol{x}\, d\boldsymbol{y}\ \delta(\boldsymbol{x} - \boldsymbol{y}) = \rho/L^3 \ . \tag{3.5b}$$

Suppose the system undergoes a first-order transition at $r = r_1$. Let the system be in the ordered phase at $r < r_1$, and let $t = (r_1 - r)a^3/J$ be the dimensionless distance from the coexistence curve. Then the probability of finding a region of size L around any given point that favors the disordered phase, i.e., where locally $\delta r_L > t$, is

$$P_{\delta r_L > t} = \int_t^\infty dr_L\, \mathcal{P}(\delta r_L) = \frac{1}{2}\ \text{erfc}\left((L/L^*)^{3/2} \right) \ . \tag{3.6a}$$

Here $\text{erfc}(x) = 1 - \text{erf}(x)$ is the complementary error function, and

$$L^* \approx a\, \delta^{1/3}/t^{2/3} \ . \tag{3.6b}$$

where we have omitted a factor of $O(1)$. L^* is the largest linear size a region or "droplet" favoring the disordered phase can have and still be found with a probability that is not exponentially small.

2. *The length scale L_c*

Now consider the disorder-induced contribution to the free energy. With φ a characteristic value of the dimensionless order parameter it is expected to be $F_{\text{dis}} \approx \overline{\delta r_L}\, \varphi^2 L^3$, with $\overline{\delta r_L}$ a characteristic value of δr_L. For the latter we take

$$\overline{\delta r_L} = \int_t^\infty d(\delta r_L)\, \delta r_L\, \mathcal{P}(\delta r_L) \approx \rho\, L^{-3/2}\, e^{-\frac{1}{2}(L/L^*)^3} \ , \tag{3.7}$$

which is the average of δr_L under the probability distribution $\mathcal{P}$ times the probability of finding a droplet of this size. We thus have

$$F_{\text{dis}} \approx \Delta(L)\, L^{3/2} \tag{3.8a}$$

where

$$\Delta(L) = \Delta_0\, e^{-\frac{1}{2}(L/L^*)^3} \tag{3.8b}$$

with

$$\Delta_0 = \delta^{1/2}\, J\, \varphi^2/a^{3/2} \ . \tag{3.8c}$$

This is a generalization of the first term in Eq. (2.6).

In addition to F_{dis}, we need to consider the surface energy cost. Suppose the order, upon approaching a droplet, is gradually destroyed over a length L_{dw}, and the droplet wall is locally in the y-z plane. Then the surface energy from the gradient-squared term in Eq. (3.1) is $F_{\text{dw}} \approx L_y L_z c \varphi^2/L_{\text{dw}}$. The LGW coefficient c is roughly $c \approx J/a$, and hence the surface tension $\sigma = F_{\text{dw}}/L_y L_z$ is

$$\sigma \approx J\varphi^2/aL_{\text{dw}} \ . \tag{3.9}$$

The free-energy difference from Eq. (2.6) now becomes

$$\Delta F = -F_{\text{dis}} + F_{\text{dw}} = -\Delta(L) L^{3/2} + \sigma\, L^2 \ , \tag{3.10}$$

and the largest possible droplet size L_c is the solution of the transcendental equation

$$L_c = L_c^0\, e^{-(L_c/L^*)^3} \ , \tag{3.11a}$$

which generalizes Eq. (2.7). Here

$$L_c^0 = (\Delta_0/\sigma)^2 \approx \delta L_{\text{dw}}^2/a \ . \tag{3.11b}$$

Apart from logarithmic corrections in the case $L_c^0 \gg L^*$, the solution of Eq. (3.11a) is

$$L_c \approx \text{Min}(L_c^0, L^*) \ . \tag{3.11c}$$

3. *The length scale L_0*

The length scale L_0 is determined by the condition that the energy gain F_{dis} from the disorder fluctuation be larger than the characteristic energy J:[30]

$$\Delta(L_0)\, L_0^{3/2} = J \ . \tag{3.12a}$$

Using Eqs. (3.8) we have a transcendental equation for L_0,

$$L_0 = L_0^0 \, e^{(L_0/L^*)^3/3} \ , \tag{3.12b}$$

where

$$L_0^0 = a/\delta^{1/3} \, \varphi^{4/3} \ . \tag{3.12c}$$

This has a solution only if $L_0^0 \ll L^*$, in which case

$$L_0 \approx L_0^0 \ . \tag{3.12d}$$

4. *The length scale L_{dw}*

Of the length scales discussed so far, L^* depends explicitly on the distance t from the coexistence curve, L_0 depends on t only via φ, and L_c^0 depends on t via L_{dw}. In order to estimate the latter, we must distinguish between scalar and vector order parameters. Let us first discuss the latter case.

A detailed determination of the droplet-wall thickness L_{dw} is a very hard problem, but we can gain sufficient insight from the case of a domain wall between ordered domains, say, ferromagnetic domains for the sake of definiteness.[31,32] In the case of a truly isotropic order parameter the domain-wall width is equal to the system size. The reason is that an order-parameter modulation with an infinite wavelength does not cost any energy; i.e., it is a consequence of the existence of gapless Goldstone modes. In any real solid the underlying lattice couples to the order parameter and produces a small gap $\Omega_g \ll J$, which corresponds to a length scale $L_g \gg a$, see Sec. III A. The domain-wall width in an infinite system is then given by

$$L_{\text{dw}} \approx L_g/\varphi \ . \tag{3.13}$$

In the case of a ferromagnet, $L_g \approx a/g_{\text{so}}$,[31,32] with $g_{\text{so}} \ll 1$ the dimensionless spin-orbit coupling.

In our case we are interested in a wall between the ordered bulk and a disordered droplet, rather than one between two ordered domains. This makes the problem more complicated, since the suppression of the long-ranged order can occur via a loss of angular correlation, or via a modulation of the modulus of the order parameter, or both, but we expect Eq. (3.13) to still give the correct order of magnitude.[33]

We stress that the above considerations apply to an infinite system, or one whose linear size L is large compared to L_g. For $L < L_g$ one has $L_{\text{dw}} \approx L/\varphi$, the surface tension σ, Eq. (3.9), is proportional to $1/L$, and the corresponding contribution to the free energy scales as $F_{\text{dw}} \sim L^{d-2}$ rather than L^{d-1}.[25] For our phase-separation problem this implies an intrinsic and profound asymmetry between the ordered and disordered phases, as in the latter the size of the ordered "system" is the droplet size. This implies that it is harder to form droplets in the disordered phase. We will come back to this point in Sec. IV.

Now consider making the anisotropy stronger and stronger. Ω_g will increase, and L_g will decrease, with the limiting case being $\Omega_g \approx J$, $L_g \approx a$, for a strongly broken rotational symmetry. This limiting case describes an Ising system, as can be checked by solving the saddle-point equation for an Ising-type action, see Appendix A. We conclude that Eq. (3.13) is generally valid. Deep inside the ordered phase, where $\varphi \approx 1$, we have $L_{\text{dw}} \approx a$ for Ising systems and $L_{\text{dw}} \approx a/g \gg 1$, with g the symmetry-breaking parameter, for systems with a weakly broken rotational symmetry. (In the case of a ferromagnet, $g = g_{\text{so}}$.) Near the coexistence curve, L_{dw} gets enhanced by a factor of $1/\varphi_1$, with φ_1 the discontinuity of the order parameter at the first-order transition.

5. *Conditions for the existence of droplets*

We now are in a position to discuss the region in the phase diagram where droplets can exist. The basic requirement is that the largest droplets that are energetically allowed are large enough to host a distinguishable minority phase, i.e., we must have $L_c > L_0$. Since L_c is the smaller of L_c^0 and L^*, see Eq. (3.11c), we need to distinguish two cases:

(i) If $L^* > L_c^0$, which is equivalent to $t < t^*$, with

$$t^* = (a/L_g)^3 \varphi^3/\delta \ , \tag{3.14}$$

we have $L_c = L_c^0$, which leads to

$$\delta > \varphi_1^{1/2} (a/L_g)^{3/2} \equiv \delta_c \ . \tag{3.15a}$$

Here we have replaced φ by φ_1, the value of the order parameter on the coexistence curve, since our considerations are valid close to a first-order transition.

(ii) If $L_c^0 > L^*$, which is equivalent to $t > t^*$, we have $L_c = L^*$, which leads to

$$t < \delta \, \varphi_1^2 \equiv t_c \tag{3.15b}$$

and we have again replaced φ by φ_1. In this second case droplet thus can exist in a window of t-values, $t^* < t < t_c$, and in order for this window to exists the inequality (3.15a) must again hold.

The Eqs. (3.15) are the two conditions for the existence of droplets containing the disordered phase within the ordered phase.[34] Equation (3.15a) sets a threshold for the disorder strength required for droplets to exist. It is smaller for more isotropic systems (larger L_g), and for more weakly first-order transitions (smaller φ_1). For a given disorder strength, Eq. (3.15b) determines the region in the phase diagram where droplets can exist. The size of this region goes to zero for $\varphi_1 \to 0$, consistent with our conclusion in Sec. II that no droplets can exist in the vicinity of a continuous transition. Now consider a phase diagram that includes a tricritical point, such as in Fig. 1(a), and consider a point on the coexistence curve. Let us assume that Eq. (3.15a) is satisfied. This will require a very small amount of disorder for systems where $L_g \gg a$, but is more difficult to fulfill in Ising-type systems; for an estimate within a very simple model, see Appendix B. Close to the tricritical point φ_1 is small, and droplets can exist only in a very small t-region according to Eq. (3.15b). As we move to lower temperatures, φ_1 increases, and reaches its maximum at $T = 0$. Equation (3.15b) thus predicts that droplets can exist in a wedge-shaped region that emanates from the tricritical point. Analogous considerations hold for phase diagrams where the tricritical point is not accessible (Fig. 1(b)), or where the coexistence line ends in a critical point, Fig. 1(a). All of this is qualitatively consistent with the observations, see the schematic phase diagrams shown in Fig. 1.

B. The ordered-volume fraction

We now return to the probability considerations of Sec. III A 1 in order to obtain an expression for the ordered-volume fraction. We start by considering the following conditional probability: Given a disorder fluctuation that favors the paramagnetic phase, the probability density of that region having a volume V is, according to Eq. (3.6a),

$$\mathcal{P}_{\delta r_L > t} = \text{erfc}(\sqrt{V/V^*}) / \int_0^\infty dV\ \text{erfc}(\sqrt{V/V^*})\ , \quad (3.16)$$

with $V^* = (L^*)^3$. Of these regions, only those with volumes between $V_0 = (L_0)^3$ and $V_c = (L_c)^3$ have a lower free energy than the ordered phase once the surface energy is taken into account. The probability that a given disorder fluctuation forms a minority-phase droplet is thus

$$\begin{aligned} P_d &= \int_{V_0}^{V_c} dV\, \mathcal{P}_{\delta r_L > t}(V) \\ &= p_d(\sqrt{V_c/V^*}) - p_d(\sqrt{V_0/V^*})\ , \quad (3.17a) \end{aligned}$$

where we have defined

$$p_d(x) := \text{erf}(x) + 2x^2\, \text{erfc}(x) - 2x e^{-x^2}/\sqrt{\pi}\ . \quad (3.17b)$$

In the region $L^* < L_c^0$, where each disorder fluctuation can support only one droplet, this leads to an ordered-volume fraction $F_{\text{OV}} \approx 1 - \frac{1}{2}\, P_d$. The factor of 1/2 accounts for the conditional-probability nature of our starting point, Eq. (3.16): On the coexistence curve half of all disorder fluctuations will favor the disordered phase, and the other half will favor the ordered phase. In the region $L^* < L_c^0$, which corresponds to $t > t^*$, we have, from Eq. (3.11c), $L_c \approx L^*$, and hence

$$F_{\text{OV}} \approx 1 - \frac{1}{2}\left[p_d(1) - p_d(\sqrt{V_0/V^*})\right]\ , \quad (L^* < L_c^0)\ . \quad (3.18a)$$

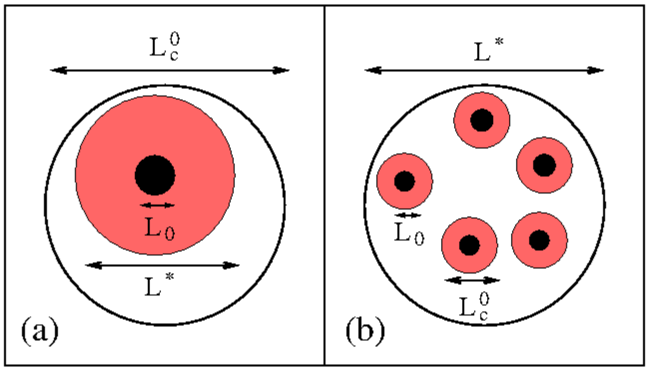

FIG. 2: (a) For $L_c = L^* < L_c^0$ each disorder fluctuation of size L^* can support one droplet (orange (grey) circle) of size $L_c = L^*$. The minimum droplet size is L_0 (black circle), and droplets are expected to take up a volume fraction $1 - V_0/V^*$. (b) For $L^* \gg L_c^0 = L_c$ a disorder fluctuation can support on the order of $N_d \approx C_d V^*/V_c$ droplets, with C_d a number that does not exceed the close-packing fraction for droplets. Droplets are expected to take up a volume fraction $C_d[1 - V_c^0/V_0]$.

For $L^* > L_c^0$ the relation between F_{OV} and P_d changes. This is obvious for $L^* \gg L_c^0$, when a single disorder fluctuation can contain many droplets. We roughly expect $F_{\text{OV}} \approx 1 - \frac{1}{2}\, P_d\, N_d$, where N_d is the number of droplets per volume V^*. Ignoring droplet-droplet interactions and a factor of $O(1)$, we have $N_d \approx C_d\, V^*/V_c$, where C_d is a number that is bounded above by the close-packing volume fraction of the droplets within the volume V^*. The precise value of C_d depends on many details; we expect it to be on the order of 1/2. Since $L_c = L_c^0$ in this region, this leads to an ordered-volume fraction

$$F_{\text{OV}} \approx 1 - \frac{C_d}{2}\left[p_d(\sqrt{V_c^0/V^*}) - p_d(\sqrt{V_0/V^*})\right] \frac{V^*}{V_c^0}\ , \quad (L_c^0 < L^*)\ . \quad (3.18b)$$

We note again that these simple arguments ignore any interaction between the droplets as well as correlations between the disorder fluctuations and are expected to give only a very rough estimate of the ordered-volume fraction. With this in mind, let us consider the behavior of F_{OV} as a function of t as the coexistence curve is approached:

(1) $t > t_c$. In this region $L_c = L^* < L_0$. Equation (3.15b) is violated, droplets cannot exist, and $F_{\text{OV}} = 1$.

(2) $t_c > t > t^*$. In this region $L_0 < L_c = L^*$. Droplets can exist, and P_d increases from 0 at $t = t_c$, where $V^* = V_0$, to $p_d(1) - p_d((\delta_c/\delta)^2)$ for $t = t^*$, with δ_c from Eq. (3.15a). $p_d(1) \approx 0.74$, and $p_d((\delta_c/\delta)^2) \ll 1$ unless the disorder strength is close to the threshold value δ_c. More specifically, $p_d(x \to 0) = 2x^2$, so for $V_0 \ll V^*$ we have $P_d = p_d(1) - 2V_0/V^* = p_d(1) - 2(\delta_c/\delta)^2$, consistent with what one would expect from a simple geometric argument, viz., $P_d \approx 1 - V_0/V^*$, see Fig. 2(a). For $t = t^*$ we thus expect F_{OV} to be on the order of 2/3.

(3) $t^* > t$. In this region L^* continues to increase and eventually multiple droplets will form within each disorder fluctuation. In the limit $L^* \gg L_c^0$, asymptotically close to the coexistence curve, Eq. (3.18b) yields $F_{\rm OV} \approx 1 - C_d[1 - V_0/V_c^0] = 1 - C_d[1 - (\delta_c/\delta)^4]$. This is again consistent with a the expectation from a simple geometric consideration, see Fig. 2(b).

In summary, for $t \gtrsim t^*$ our mechanism results in behavior that is qualitatively consistent with the experimental observations: Droplets can exist within certain distance t_c from the coexistence curve, and the ordered-volume fraction decreases from unity as the first-order transition is approached from within the ordered phase. We will discuss the corresponding behavior in the disordered phase in Sec. IV.

IV. DISCUSSION

We conclude by discussing our results and their underlying assumptions in more detail, and also add some remarks about aspects of the problem that we have not covered so far.

Before going into details, let us first reiterate how widespread the observations are. Phase separation is observed near phase transitions that are known or suspected to be first order in a wide variety of materials with an equally wide variety of types of order; examples were given in the introduction. In addition to transitions from an ordered phase to a disordered one it also is observed near first-order transitions from one ordered phase to another; an example is $\rm UGe_2$, where it is observed both near the FM1-PM transition[5] and near the FM1-FM2 transition within the FM phase.[6] Furthermore, the observations are the same for systems with a strongly uniaxial or Ising-like order parameter, such as $\rm UGe_2$, and systems with an order parameter that is approximately rotationally invariant or Heisenberg-like, such as MnSi.

As we pointed out in Sec. I, these observations of the coexistence of two phases *away from the coexistence curve* are very surprising and require an explanation. As we have shown, disorder fluctuations provide a plausible scenario. We reiterate that the conditions for this scenario to work require less disorder for systems with an approximate rotational symmetry than for Ising-like ones, see the discussion after Eqs. (3.15), and Appendix B. At the same time, there is some experimental evidence for inhomogeneities that do not necessarily show in transport experiments and therefore can be present even in nominally rather clean systems.[12]

A. The role of disorder

We emphasize that our scenario requires a substantial amount of disorder that couples to the order parameter. This is not to say that the disorder is necessarily visible in the transport properties of metallic systems. If it were, then a rough estimate for our disorder parameter δ would be $1/k_{\rm F}\ell$, with $k_{\rm F}$ the Fermi wave number and ℓ the elastic mean-free path. In clean samples of, e.g., MnSi, $k_{\rm F}\ell$ can exceed $1,000$.[35] However, Yu et al. have reported evidence for substantial pressure inhomogeneities in MnSi.[12] The latter do indeed couple to the order parameter, as is evidenced by the strong dependence of the transition temperature on applied hydrostatic pressure. Pfleiderer et al. have emphasized the sensitivity of systems near quantum phase transitions in general to disorder.[3] This can be illustrated as follows.

Consider a thermal transition with a transition temperature $T_{\rm c}$ that depends on a defect concentration $n(\boldsymbol{x}) = n + \delta n(\boldsymbol{x})$:

$$T_{\rm c}(\boldsymbol{x}) \approx T_{\rm c}(n) + \delta n\, dT_{\rm c}/dn\ . \tag{4.1}$$

Let the defect concentration fluctuations be randomly distributed with a second moment $\langle \delta n(\boldsymbol{x})\, \delta n(\boldsymbol{y})\rangle \approx n\, \delta(\boldsymbol{x} - \boldsymbol{y})$. The definition of the dimensionless disorder δ in Eqs. (3.2, 3.3) then leads to the estimate $\delta \approx (n/a^3 T_{\rm c}^2)(dT_{\rm c}/dn)^2$. Now model the n-dependence of $T_{\rm c}$ by $T_{\rm c} \approx T_{\rm c}^0[1 - (n/n_c)^2]$, which is roughly the shape of the phase diagram in, for instance, many quantum ferromagnets that display a quantum phase transition triggered by chemical composition.[35]. This leads to an estimate

$$\delta \approx \frac{1}{a^3 n_c}\, (n/n_c)^3\ . \tag{4.2}$$

For n a sizable fraction of n_c this is of $O(1)$. The observed strong dependence of $T_{\rm c}$ on a dopant concentration in many systems thus translates into a rather large value of δ. At least in some materials is therefore would be misleading to conclude, from the fact that they are good metals, that disorder is irrelevant. We also note that weak-localization effects are not commonly observed in quantum ferromagnets with a ferromagnet-to-paramagnet quantum phase transition driven by composition.[35] This is another example of disorder that couples to the order parameter, but is not easily observed in the transport properties.

We also mention that there was no a priori guarantee that the energetic considerations in Sec. II would lead to an observable size of the inhomogeneity region for reasonable values of the disorder, as we saw in Sec. III is indeed the case. The fact that we found the wedge-shaped region where minority-phase droplets are stable to be of an observable size is nontrivial and consistent with our interpretation of the existing experiments.

B. The ordered-volume fraction in the ordered and disordered phases

In Sec III B we have discussed the ordered-volume fraction $F_{\rm OV}$ as the first-order transition is approached from the ordered phase. We found that $F_{\rm OV}$ drops below unity due to the existence of droplet in a certain region bounded by a dimensionless distance t_c from the coexistence curve. t_c depends on the strength of the disorder and the strength of the first-order transition, see Eq. (3.15b). $F_{\rm OV}$ then decreases monotonically with decreasing t. In the asymptotic region $t < t^*$, with t^* given by Eq. (3.14), some of our simple assumptions become questionable. For instance, the assumption of noninteracting droplet will certainly break down with increasing droplet density. It is likely that at some point a percolation transition will lead to droplets merging, and the precise behavior on or very close to the coexistence curve is a very hard problem. For instance, it is not obvious whether $F_{\rm OV}$ is continuous or discontinuous across the coexistence curve, and more detailed considerations are necessary to determine this.

Another issue is the behavior on the disordered side of the phase transition; we have considered the existence of minority-phase droplets in an ordered majority phase only. At first sight one might think that the behavior should be roughly symmetric with respect to the phase boundary; however, this is likely not the case. For instance, our estimate of the droplet-wall width $L_{\rm dw}$ in Sec. III A 4 assumes that most of the surface free-energy cost of forming the droplet is paid within the bulk ordered phase. This is plausible at least at a saddle-point level, as the saddle-point differential equation for a droplet field configuration is very similar to, say, a square-well problem in quantum mechanics. If the same is true in the disordered phase, then this introduces an intrinsic asymmetry into the problem: In the disordered bulk there are no Goldstone modes, the problem is always Ising-like, and $L_{\rm dw}$ is likely substantially smaller than in the ordered phase. This will make droplet energetically less favorably in the disordered phase than in the ordered one. If there are solutions for which this is not the case, then one needs to add the requirement $L_c > L_{\rm dw}$ to the conditions for droplet existence, as we have noted above.[34] In either case, the conditions for droplet existence are more stringent in the disordered phase. While speculative, these considerations are consistent with the experimental observations, which generally see much weaker indications of phase separation in the disordered phase than in the ordered one. Another contributing factor may be experimental limitations: If droplets are intrinsically smaller in the disordered phase, then the spatial resolution limit of any experimental technique will lead to an underestimation of the ordered-volume fraction.

C. First-order transitions in the presence of disorder

The fate of a first-order transition in the presence of disorder is a problem with a long history, going back to Imry and Ma, and Imry and Wortis.[25,26] The relevant arguments have been made rigorous in Refs. 36,37 for classical systems, and in Refs. 38,39 for the quantum case.[40] The conclusion of these studies is that no sharp first-order phase transition is possible in dimensions $d \leq 2$ for systems with a discrete symmetry, and in $d \leq 4$ for systems with a continuous symmetry. Consistent with this, Fernández et al found a sharp first-order transition in a 3-d disordered Potts model.[41] In the present context these rigorous results provide, strictly speaking, no constraints: We have considered only systems with a discrete symmetry (as is always the case for any system on a lattice), so in $d = 3$ a sharp first-order phase transition is possible. One might wonder, however, if in the case of a weakly broken continuous symmetry any sharp transition must be weakly first order. To see whether our droplet scenario is consistent with this hypothesis would require a determination of the ordered-volume fraction on the coexistence curve, where our arguments are not reliable. We can imagine four scenarios: (1) The non-overlapping droplet picture remains valid up to the transition, and $F_{\rm OV}$ (and hence the magnetization) has a discontinuity on the coexistence curve. This would mean that there still is a sharp first-order transition. (2) The droplets merge at some critical droplet density, and the resulting percolation transition is first order,[42] leading again to a discontinuous $F_{\rm OV}$. (3) $F_{\rm OV}$ changes continuously from unity in the ordered phase to exponentially small values in the disordered phase. This would mean no sharp transition, i.e., the original sharp first-order transition has been smeared due to the existence of droplets. (4) A second-order percolation transition occurs in the droplet system. This would mean there still is a sharp transition, but it is continuous rather than first order.

More detailed work is needed in order to determine which of these possibilities is realized. We note that our scenario is entirely consistent with possibilities (1) - (3). If (4) is realized, then some of our arguments will have to be reconsidered, at least close to the transition, in order to ensure consistency.

We finally comment on the relation between scaling descriptions of first-order phase transitions[20,21] and the rigorous bound for the correlation-length exponent, Eq. (2.1). The scaling description leads a relevant operator with scale dimension $\lambda = 1/\nu = d$. Naively, this violates Eq. (2.1) and thus seems to preclude a sharp first-order transition in any dimension for any nonzero amount of disorder. This apparent contradiction is resolved by the realization that there are two distinct correlation-length exponents: One is related to the rounding of the transition in system of finite size L and is equal to $\nu_{\rm rounding} = 1/d$ at any regular first-order transition.[20,43] The other is related to the effects of the disorder, which shifts the transition point relative to the clean one. For uncorrelated disorder, this exponent is equal to $\nu_{\rm shift} = 2/d$. The theorem proven by Chayes et al.[18], Eq. (2.1) applies to the latter, so there is no contraction. The presence of two positive exponents is not at odds with the usual notion of only one relevant operator at a fixed point describing a phase transition, since the tuning parameter for a first-order transition is $r(L)L^d$, with $r(L) = r_\infty + r_1/L^{d/2}$ the scale-dependent mass parameter. The tuning parameter is thus $r_\infty L^d + r_1 L^{d/2} = r_\infty L^d[1 + (r_1/r_\infty)/L^{d/2}]$, and the operator with scale dimension $d/2$ describes corrections to scaling in a well-defined sense.

Acknowledgments

We thank John Toner and Yasutomo Uemura for discussions. This work was supported by the NSF under grant Nos. DMR-1401410 and DMR-1401449.

Appendix A: The length scale $L_{\rm dw}$ in an Ising model

Consider an action

$$\mathcal{A} = \mathcal{A}^{(2)} + \int_V d\boldsymbol{x}\ \left[v\,\phi^3(\boldsymbol{x}) + u\,\phi^4(\boldsymbol{x})\right] \tag{A1}$$

with $\mathcal{A}^{(2)}$ from Eq. (3.1), which allows for a first-order transition by virtue of the cubic term with coupling constant v. In natural units, $u = (J/a^3)\hat{u}$, $v = (J/a^3)\hat{v}$, with $\hat{u}$ and $\hat{v}$ dimensionless. In a mean-field approximation, this model has a first-order transition at $r = r_1 = 2v^2/9u$ where the order parameter changes discontinuously from $\phi = \phi_1 = 2v/3u$ to zero. Now consider a situation where the mass parameter $r(x,y,z)$ changes discontinuously from $r < r_1$ for $x > 0$ to $r > r_1$ for $x < 0$, which models a plane droplet wall, and look for a variational solution of the saddle-point equation

$$c\,\phi''(x) = r\,\phi(x) - v\,\phi^2(x) + u\,\phi^3(x) \tag{A2}$$

that obeys the boundary conditions $\phi(x=0) = 0$ and $\phi(x \to \infty) = \text{const}$. Deep inside the ordered phase, for r negative and large, the v-term is negligible and the solution will vary on the microscopic length scale a. For $r = 0$ the characteristic length scale, which determines the droplet wall thickness, is $a\,\hat{u}^{1/2}/\hat{v}$, and for $r \to r_1$ it is $3a\,\hat{u}^{1/2}/\sqrt{2}\,\hat{v}$. The former result is obtained by suitable scaling of the ODE; the latter, by linearizing it about ϕ_1. We conclude that upon approaching the coexistence curve, $L_{\rm dw}$ increases from a $L_{\rm dw} \approx a$ deep inside the ordered phase to a value on the order of $L_{\rm dw} \approx a\,\hat{u}^{1/2}/\hat{v}$ in the vicinity of the coexistence curve. If $\hat{u} = O(1)$, we can write this as

$$L_{\rm dw} \approx a/\varphi\ , \tag{A3}$$

which recovers Eq. (3.13) specialized to the Ising case (Ł$_g \approx a$). For typical parameter values one expects $L_{\rm dw}$ to saturate at a few times the microscopic length.

Appendix B: Conditions for the existence of droplets within a ϕ^4-theory

Here we discuss the disorder threshold requirement expressed by Eq. (3.15a) in the framework of the Ising model action given by Eq. (A1). In the region $t \lesssim \hat{r}_1 \equiv 2\hat{v}^2/9\hat{u}$, which corresponds to $r \gtrsim 0$, we have $\phi \approx \varphi_1 = \sqrt{2\hat{r}_1/\hat{u}}$. With $L_{\rm dw} \approx a\,\hat{u}^{1/2}/\hat{v}$ from Appendix A, Eq. (3.15a) yields

$$\delta_c \approx \hat{u}^{3/4}\,\varphi_1^{1/2}\ . \tag{B1}$$

Deep inside the ordered phase, $\hat{r} \approx -1$, we have $\phi = \sqrt{-\hat{r}/\hat{u}} \approx 1/\hat{u}^{1/2}$, and at a moderately strongly first-order transition φ_1 is, say, one-tenth of that value. The remaining question is the value of $\hat{u}$. Within Hertz's model for itinerant quantum ferromagnets,[44] $\hat{u} = 1/12$, which leads to $\delta_c \approx 0.1$. A dimensionless disorder strength on this order seems realistic in the light of the discussion in Sec. IV A. For systems with an approximate continuous symmetry, where $L_{\rm dw}$ is larger, the disorder threshold will be correspondingly lower.

---

[1] L. D. Landau and E. M. Lifshitz, *Statistical Physics Part 1* (Butterworth Heinemann, Oxford, 1980).

[2] Y. J. Uemura, T. Goko, I. M. Gat-Maleruanu, J. P. Carlo, P. L. Russo, A. T. Savici, A. Aczel, G. J. MacDougall, J. A. Rodriguez, G. M. Luke, et al., Nature Physics **3**, 29 (2007).

[3] C. Pfleiderer, P. Böni, C. Franz, T. Keller, A. Neugebauer, P. G. Niklowitz, P. Schmakat, M. Schulz, Y. K. Huang, J. A. Mydosh, et al., J. Low Temp. Phys. **161**, 167 (2010).

[4] I. M. Gat-Maleruanu, J. P. Carlo, T. Goko, A. Fukaya, T. Ito, P. P. Kyriakou, M. I. Larkin, G. M. Luke, P. L. Russo, A. T. Savici, et al., Phys. Rev. B **84**, 224415 (2011).

[5] A. Harada, S. Kawasaki, H. Kotegawa, Y. Kitaoka, Y. Haga, E. Yamamoto, Y. Onuki, K. M. Itoh, E. Haller, and H. Harima, J. Phys. Soc. Japan **74**, 2675 (2005).

[6] H. Kotegawa, A. Harada, S. Kawasaki, Y. Kawasaki, Y.Kitaoka, Y. Haga, E. Yamamoto, Y. Onuki, K. M. Itoh, E. E. Haller, et al., J. Phys. Soc. Japan **74**, 705 (2005).

[7] G. M. Luke, A. Keren, L. P. Le, B. J. Sternlieb, W. D. Wu, Y. J. Uemura, Y. Onuki, and T. Komatsubara, Phys. Rev. Lett. **73**, 71853 (1994).

[8] K. M.Kojima, S. Uchida, Y. Fudamoto, I. M. Gat, M. I. Larkin, Y. J. Uemura, and G. M. Luke, Physica B **326**, 316 (2003).

[9] B. Frandsen, L. Liu, S. Cheung, Y. J. Uemura, T. Munsie, M. Wilson, A. Hallas, G. M. Luke, B. Chen, C. Jin, et al. (2015), http://meetings.aps.org/link/BAPS.2015.MAR.D21.7.

[10] B. A. Frandsen, L. Liu, S. C. Cheung, Z. Guguchia, R. Khasanov, E. Morenzoni, T. J. S. Munsie, A. M. Hallas, M. N. Wilson, Y. Cai, et al. (2016), unpublished results.

[11] C. Pfleiderer, G. J. McMullan, S. R. Julian, and G. G.

Lonzarich, Phys. Rev. B **55**, 8330 (1997).

[12] W. Yu, F. Zamborszky, J. D. Thompson, J. L. Sarrao, M. E. Torelli, Z. Fisk, and S. E. Brown, Phys. Rev. Lett. **92**, 086403 (2004).

[13] Y. J. Uemura (2015), private communcation.

[14] J. D. Gunton, M. San Miguel, and P. S. Sahni, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. Lebowitz (Academic, New York, 1983), vol. 8, p. 269.

[15] Doiron-Leyraud et al. (Ref. 45) have speculated that droplet formation might occur in the paramagnetic phase of MnSi. We stress that these ideas pertained to dynamical phenomena, whereas we are discussing static droplets in either phase.

[16] In general there may be different correlation lengths, and different associated exponents ν. For instance, Eq. (2.1) has been proven for an appropriately defined finite-size correlation length exponent.[18] We will come back to this point in Sec. IV.

[17] A. B. Harris, J. Phys. C **7**, 1671 (1974).

[18] J. T. Chayes, L. Chayes, D. S. Fisher, and T. Spencer, Phys. Rev. Lett. **57**, 2999 (1986).

[19] M. N. Barber, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, New York, 1983), vol. 8, p. 145.

[20] M. E. Fisher and A. N. Berker, Phys. Rev. B **26**, 2507 (1982).

[21] V. Privman and M. E. Fisher, J. Stat. Phys. **33**, 385 (1983).

[22] M. A. Continentino and A. S. Ferreira, Physica A **339**, 461 (2004).

[23] T. R. Kirkpatrick and D. Belitz, Phys. Rev. B **91**, 214407 (2015).

[24] Here we assume that a root-mean-square deviation is an adequate representation of an average value. Imry[46] makes the same assumption in his estimates of the finite-size rounding of a first-order transition.

[25] Y. Imry and M. Wortis, Phys. Rev. B **19**, 3580 (1979).

[26] Y. Imry and S. K. Ma, Phys. Rev. Lett. **35**, 1399 (1975).

[27] Whether or not one introduces the notion of a frequency here is a matter of taste. Alternatively, one can define L_g as the length scale that cuts off the long-range nature of the correlations that characterized the Goldstone modes, without any reference to dynamics.

[28] Our arguments apply equally to thermal and quantum phase transitions, even though Eq. (3.1) represents a classical action. Generalization to a quantum mechanical action does not affect the following discussion.

[29] We note that the dimensionless factors that are left undetermined in these purely dimensional arguments are not necessarily close to unity. For instance, one can argue that ρ should be proportional to J^2 times the microscopic wave number $2\pi/a$ cubed, which leads to a factor of $(2\pi)^3$ in Eq. (3.3).

[30] This is a generalization of the argument given by Privman and Fisher[21] for thermal first-order phase transitions. They approximated the partition function as $Z \approx e^{-F_1/T} + e^{-F_2/T}$, see Eq. (2.17) in Ref. 21, with $F_{1,2}$ the free energies of the two phases. Writing this as $Z \approx e^{-F_1/T}\left(1 + e^{(F_1-F_2)/T}\right)$ makes it clear that in order to have a distinct phase in a droplet one must have $F_1 - F_2 < T_c \approx J$, and the latter inequality will be equally valid at a quantum phase transition as at a thermal one.

[31] C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1996), chap. 15.

[32] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford, 1984).

[33] There is disagreement in the literature about the scaling of the domain-wall energy with the system size in systems with a continuous symmetry.[47,48] This is a complicated issue since in the case of a continuous symmetry loss of order can be achieved by either the modulus of the order parameter going to zero, or by phase decoherence, or both. We only use the argument that L_{dw} is larger in a weakly anisotropic system than in a strongly anisotropic one.

[34] We need to ask whether $L_c > L_{\text{dw}}$ is also necessary for the existence of well-defined droplets. In the ordered phase this condition is *not* necessary, since a typical order-parameter configuration will decrease gradually as the droplet is approached from the outside, and then decrease exponentially inside the droplet. However, for droplets to exist in the *disordered* phase it may be necessary if the order parameter is exponentially small outside the droplet and needs to build gradually inside. This is one possible reason for the observed asymmetry between the ordered and disordered phases, see the discussion.

[35] M. Brando, D. Belitz, F. M. Grosche, and T. R. Kirkpatrick (2016), Rev. Mod. Phys., in press, arXiv:1502.02898.

[36] M. Aizenman and J. Wehr, Phys. Rev. Lett. **62**, 2503 (1989).

[37] M. Aizenman and J. Wehr, Commun. Math. Phys. **130**, 489 (1990).

[38] R. L. Greenblatt, M. Aizenman, and J. L. Lebowitz, Phys. Rev. Lett. **103**, 197201 (2009).

[39] M. Aizenman, R. L. Greenblatt, and J. L. Lebowitz, J. Math. Phys. **53**, 023301 (2012).

[40] The original Imry-Ma argument was for a random-field problem. It was later extended to other types of disorder.[37,49].

[41] L. A. Fernández, A. Gordillo-Guerrero, V. Martín-Mayor, and J. J. Ruiz-Lorenzo, Phys. Rev. Lett. **100**, 057201 (2008).

[42] Y. S. Cho and B. Kahng, Nature Sci Rep **5**, 11905 (2015).

[43] In glasses, random first-order transitions have been considered where there is an intrinsic randomness to the order parameter and $\nu_{\text{rounding}} = \nu_{\text{shift}} = 2/d$, see Refs. 50,51.

[44] J. Hertz, Phys. Rev. B **14**, 1165 (1976).

[45] N. Doiron-Leyraud, I. Walker, L. Taillefer, M. J. Steiner, S. R. Julian, and G. G. Lonzarich, Nature (London) **425**, 595 (2003).

[46] Y. Imry, Phys. Rev. B **21**, 2042 (1980).

[47] A. N. Berker, Physica **194A**, 72 (1993).

[48] P. Goswami, D. Schwab, and S. Chakravarty, Phys. Rev. Lett. **100**, 015703 (2008).

[49] K. Hui and A. N. Berker, Phys. Rev. Lett. **62**, 2507 (1989).

[50] T. R. Kirkpatrick, D. Thirumalai, and P. G. Wolynes, Phys. Rev. A **40**, 1045 (1989).

[51] P. Charbonneau, J. Kurchan, G. Parisi, P. Urbani, and F. Zamponi, Nature Commun. **5**, 3725 (2014).