

sult of a better distribution of the pressure across the end surfaces of the sample. A higher component of p along [010] for curve 2 explains the increased dT_c/dp coefficient, since dT_c/dp is higher for p along [110]. Curve 1 was measured first, starting at $p=0$. Then the points above $p=0.35$ kbar on curve 2 were taken. Then, having kept the sample at room temperature at $p=0$ for several days, we measured the points below $p=0.3$ kbar on curve 2. For the sake of clarity, the quantities A , $T_d - T_0$, and ΔT_c , corresponding to points on curve 2 above $p=0.20$ kbar, were adjusted to correct for a shift caused by the imperfect pressure distribution.⁹

We see that both A and $T_d - T_0$ fall off in the second-order region of Fig. 3 and just below the TCP's in Fig. 4, which supports our conclusions that the system has different properties here. This change of Landau parameters, we believe, is connected to an increased influence from critical fluctuations, which must be expected to be larger in a second-order region and close to a TCP. Also, the fact that dT_c/dp is higher for p along [110] than for p along [100] indicates influence of fluctuations. According to Landau theory, it should be the same in these two cases.¹⁰ Note that $T_d - T_0 = 0$ at a Landau tricritical point. Our measurements, which give $T_d - T_0 > 0$ even close to the TCP's, confirm that the change of order of the transition is caused by critical fluctuations and is not a result of pressure-dependent Landau parameters. Landau theory explains the specific-heat curve shape outside the temperature range of 0.5 K around T_c . Close to T_c , critical fluctuations are important for the C_p curves,

and they determine the order of the phase transition and influence the shape of the phase diagrams and the values of the Landau parameters.

In conclusion, our measurements of $T_c(p)$ have led us to believe that KMnF_3 for p along [110] has a TCP near $p=0.45$ kbar, $T=109.4$ K, and two new consecutive TCP's joined by a second-order line near $p=0.25$ kbar and $T=188.0$ K for p along [100]. The specific-heat curves fit Landau theory with parameters showing a change with pressure which is correlated with the change of the order of the transitions. More details will be published elsewhere.⁹

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Droplet Theory of Low-Dimensional Ising Models

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A droplet theory of Ising systems is constructed, valid near space dimension $d=1$, and based on a model incorporating only the configurational energy of surface tension. The droplet distribution function is calculated explicitly, with use of renormalization-group methods to control the droplet-shape fluctuation effects essential to its scaling form. Universal quantities are calculated; those (e.g., β) which reflect the droplet concentration have an essential singularity in $\epsilon = d - 1$.

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Droplet models have been studied for many years with a view to obtaining a theoretical description of a two-phase (Ising-like) thermody-

amic system.¹ Two problems have attracted particular attention. On the one hand, the existence of large droplets of one phase embedded

in a background of the other phase has been recognized as the source of an essential singularity on the coexistence curve.² In this case the relevant droplets have scale size R large compared with the correlation length ξ ; the theory is tractable since these droplets are geometrically well defined (ξ measures the intrinsic thickness of the droplet boundary) and surface tension is manifestly controlling the droplet distribution. On the other hand, droplet models have formed the basis of numerous theories of behavior near the critical point.³ In this case it is recognized that droplets with R small compared with ξ (though still large compared with the lattice spacing L_0) also play an essential role. To date no computational framework has existed for this range of droplet sizes; accordingly, droplet theories of critical-point behavior have been largely phenomenological.³

In this paper we present a theory which provides a unified description of droplets of *all* scale sizes. The theory is based on the statistical

mechanics of droplet configurations controlled entirely by surface tension for the zero field case which we consider in this paper; the description of the regime $R \leq \xi$ is achieved with use of renormalization-group (RG) methods, which are analytically controllable for sufficiently low space dimensionality d , technically for $d = 1 + \epsilon$. Universal quantities, such as droplet density, critical exponents, correlation functions, and the probability density function (pdf) for block coordinates, are calculated explicitly for small ϵ ; those quantities (e.g., the order parameter exponent β) which reflect the droplet density are found to have no perturbative expansion in ϵ . In the $\epsilon \rightarrow 0$ limit our results recapture the essential predictions of kink-based studies of one dimension.⁴ Like these, our own study illuminates the universal configurational physics underlying critical-point behavior.

Our first task is to study the statistical mechanics of the effective Hamiltonian for a *single* ("spin-down") droplet embedded in an otherwise homogeneously ordered ("spin-up") phase:

$$\mathcal{K} = T_0^{-1} \int d\Omega (R+f)^{d-1} \left[1 + \frac{1}{2}(R+f)^{-2} \sum_{i,j} (L_{ij} f)^2 \right]^{1/2}. \quad (1)$$

The integral is the surface area of a droplet of scale size R which deviates from spherical by an amount $f(\eta)$, depending on the direction η ; $L_{ij} \equiv x_i \partial/\partial x_j - x_j \partial/\partial x_i$ are the angular momentum derivatives. This is the generic surface-tension Hamiltonian which emerges⁵ from a general Landau-Ginzburg model in the "thin-wall limit" when the mass parameter of the model is taken to infinity. Our standard tool for calculating the trace over $f(\eta)$ is perturbation expansion of \mathcal{K} in f , which has a natural decomposition in spherical harmonics. Both the $l=0$ and $l=1$ components must be handled by collective-coordinate methods. The collective coordinate for the $l=1$ modes is the coordinate of the center of the droplet, and gives a factor extensive in the volume available to the droplet. The $l=0$ collective coordinate can be taken as the scale size R of the droplet. The modes with $l \geq 2$ (corresponding to droplets of a given position and scale size but varying shape) can be handled perturbatively. The resulting single-droplet partition function may be utilized to yield the mean fraction of available space occupied by *all* droplets of scale size $R \rightarrow R + dR$, in the dilute limit (realized, we shall see, for small ϵ):

$$\psi(R) dR = A (T_0 R^{-\epsilon})^{-(2+\epsilon)/2} \exp \left[\left(\frac{S_d R^\epsilon}{T_0} + \frac{2}{\epsilon \Gamma(d)} \right) \frac{dR}{R} \right] [1 + O(\epsilon, T_0 R^{-\epsilon})], \quad (2)$$

where $A = S_d^{(d+3)/2} d^{(d+2)/2} (2\pi)^{-(d+1)/2} = 2/\pi [1 + O(\epsilon)]$. Here S_d is the surface area of the unit sphere and $\epsilon = d - 1$.

For *large* R the R dependence of this result agrees with coexistence-curve calculations, even the prefactor structure being consistent with series expansions for the two-dimensional Ising model.⁶ The failure of the perturbation theory for *small* R , signaled also by the ϵ^{-1} in the exponential, may be circumvented, as in the case of the planar interface,⁷ by introducing an appropriate renormalized dimensionless temperature $T(R) = T_0 R^{-\epsilon} [1 + O(T_0 R^{-\epsilon})]$ obeying the RG equation

$$\left(\frac{R}{\xi} \right)^\epsilon = \frac{|1 - T(R)/T_c|^\nu}{T(R)} [1 + O(\epsilon T(R))], \quad (3)$$

where $\nu^{-1} = \epsilon + \frac{1}{2}\epsilon^2 + O(\epsilon^3)$ and $T_c = \epsilon + O(\epsilon^2)$. The RG-invariant length ξ locates a crossover between large- R [$T(R) \sim (R/\xi)^{-\epsilon}$] and small- R [$T(R) \rightarrow T_c$] behavior, and exhibits the critical behavior $\xi \propto [T_c - T(L_0)]^{-\nu}$; as the fundamental length scale of our theory it is identified as a measure of the true corre-

lation length. When reexpressed in terms of $T(R)$ Eq. (2) becomes

$$\psi(R) dR = A(T(R))^{-(2+\epsilon)/2} R^{-1} \exp\left[-\left(\frac{S_d}{T(R)} - \frac{\hat{S}_d \epsilon \nu}{T_c} + \frac{2}{\epsilon \Gamma(d)}\right)\right] dR [1 + O(\epsilon, T(R))]. \quad (4)$$

In conjunction with (3), this is our first key result. It is universal, being independent of the renormalization scheme; we have also derived it directly from limiting cases of a class of Landau-Ginzburg models. Its scaling form $\psi(R) = R^{-1} \tilde{\psi}(R/\xi)$ originates in the renormalization effects of fluctuations in droplet shape.⁸ For $\epsilon \rightarrow 0$, (3) and (4) correctly reproduce the results appropriate to a one-dimensional distribution of kink-antikink pairs. For $R \rightarrow 0$ we find $\psi(R) = \psi_0 R^{-1}$, where

$$\psi_0 = 2\pi^{-1} \epsilon^{-(2+\epsilon)/2} \exp\{-[1 + 2C + (2/\epsilon)]\} [1 + O(\epsilon)] \quad (5)$$

and $C = 0.577\dots$ is Euler's constant.

In utilizing Eq. (4) to characterize actual multi-droplet configurations one must build in two essential refinements. Firstly, even in a noninteracting droplet picture, one must account for excluded-volume effects: A spin-down droplet cannot meaningfully be embedded in a region occupied by a larger spin-down droplet. Secondly, it is clear that, any spin-down droplet will in general contain smaller spin-up droplets, which may themselves contain still more-deeply nested droplets.⁹ Configurations consistent with these requirements, and with Eq. (4), may be constructed by decorating a hyperspherical volume $S_d d^{-1} L^d$ of homogeneously ordered spin-up phase with spin-down *and* spin-up droplets of successively smaller scale sizes ranging from L down to L_0 . At each stage of the decoration procedure the down- (up-) spin droplets utilized are such as to cover the requisite fraction (4) of the volume occupied, *at that stage*, by up- (down-) spin phase. It is easily established that, with this prescription, the fraction of space occupied by the *outermost* of the hierarchy of spin-down droplets (i.e., droplets which are not themselves nested within larger spin-down droplets) is $\Psi_0(L, L_0) = 1 - \exp[-\int_{L_0}^L \psi(R) dR]$, while the *total* fraction of space occupied by spin-down phase is $\Psi(L, L_0) = \frac{1}{2}\{1 - \exp[-2\int_{L_0}^L \psi(R) dR]\}$. The mean number $N_-(R) dR$ of spin-down droplets with scale size $R \rightarrow R + dR$, and the *actual* volume of spin-down phase of which each consists, $M_-(R)$, then follow as

$$N_-(R) = [1 - \Psi(L, R)] L^d R^{-d} \psi(R), \quad (6a)$$

$$M_-(R) = [1 - \Psi_0(R, L_0)] S_d d^{-1} R^d. \quad (6b)$$

These key results have three immediate consequences. Firstly, the critical ($\xi \gg L_0$) droplet "density" $\rho_c \equiv (L_0/L)^d \int_{L_0}^L N_-(R) dR \approx \psi_0/2d$, revealing the physical significance of the parameter ψ_0 , whose smallness makes legitimate the dilute approximation implicit in the neglect of partial

droplet overlap.¹⁰ Secondly, the magnetization

$$\begin{aligned} m &\equiv 1 - 2(L^d S_d/d)^{-1} \int_{L_0}^L N_-(R) M_-(R) dR \\ &= 1 - 2\Psi(L, L_0) \end{aligned}$$

has the critical behavior

$$m \sim \exp\left(-\int_{L_0}^{\xi} 2\psi_0 R^{-1} dR\right) \sim (\xi/L_0)^{-2\psi_0},$$

permitting the identification $\beta = 2\psi_0 \nu$, and revealing that β does not have a perturbative expansion in ϵ . Setting $\epsilon = 1$ in the small- ϵ expansion of ψ_0 [Eq. (5)] gives $\psi_0 \approx 0.01$, in poor accord with the exact $d=2$ result $\beta/2\nu = 0.0625$; the discrepancy is scarcely surprising in view of the extreme sensitivity of the result to the manner in which Eq. (4) is extrapolated to $d=2$. Thirdly, the second moment of the droplet distribution $\int_{L_0}^L N_-(R) \times M_-^2(R) dR$ diverges with a "cluster size exponent" $\theta = (d - 2\psi_0)\nu = d\nu - \beta$. This conclusion is consistent with series-expansion studies¹¹ in $d=2$ and bears out a conjecture based on a phenomenological droplet theory.¹² We remark, however, that the general scaling form for the droplet distribution,¹² which forms the core of the phenomenological theory, is not borne out by the explicit scaling form (6a); the discrepancy can be traced to a sum rule satisfied by (6a) but not anticipated in the phenomenological theory.

The arguments developed above can be extended to account for coordinate fluctuations by allowing for a Poisson distribution of the number of each hierarchy of dressing droplets. The pair correlation function at criticality and the order parameter susceptibility have been calculated, and the associated exponents $d - 2 + \eta = 4\psi_0 = 2\beta/\nu$ and $\gamma = (d - 4\psi_0)\nu = (2 - \eta)\nu$ are consistent with two-exponent scaling theory. We have also calculated the probability density function for block ("coarse-grained") coordinates, characterizing the instantaneous spatial average of the ordering coordinate within a region of radius L_B . The predicted¹³

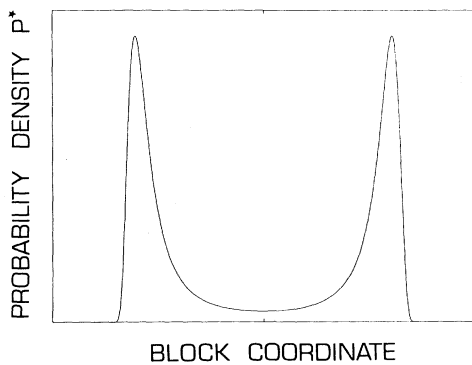


FIG. 1. Droplet-based calculation of the block-coordinate pdf at criticality, with $d = 2$ and $\psi_0 = \frac{1}{16}$.

universality of this pdf, in the range $\xi \gg L_B \gg L_0$ is borne out by the result which depends solely upon $\beta/\nu = 2\psi_0$ and d . Setting $d = 2$ and $\psi_0 = \beta/2\nu = \frac{1}{16}$ gives the form shown in Fig. 1. The existence of two distinct markedly asymmetric peaks is in accord with calculations of similar pdf's, based on the approximate recursion formula¹³ and Monte Carlo simulations^{14,15} in $d = 2$. The width of the peaks (relative to the standard deviation of the pdf) is proportional to $\psi_0^{1/2}$, and vanishes as $\epsilon \rightarrow 0$, when the pdf assumes the double- δ -function form appropriate¹³ in $d = 1$. This observation reemphasizes the connection (already apparent in the identification $\beta/\nu = 2\psi_0$) between the smallness of β/ν and the high degree of short-range order at length scales small compared to ξ , characteristic of low- d systems.

In assessing the potential for further developments, we foresee no problems of principle in extending, to higher order in ϵ , the calculation of the fundamental parameter ψ_0 ; the incorporation of the $O(\psi_0)$ corrections necessary to refine our procedure for constructing multidroplet configurations seems considerably more problematic. One may also question the relevance of the entire droplet picture to three-dimensional systems, where additional prefactors appear in the calculation of (2), and connectivity properties can lead to ambiguities in identifying localized droplets.¹² It seems clear, nevertheless, that this explicit and analytically tractable realization of droplet phenomenology does much to illuminate the configurational physics controlling exponent values: It appears that, in low enough dimension, the fractal dimension of the droplet surface¹⁶ controls ν while the critical droplet concentration controls β .

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