Scaling Theory for Finite-Size Effects in the Critical Region*

Michael E. Fisher and Michael N. Barber
Baker Laboratory, Cornell University, Ithaca, New York 14850
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Critical phenomena in films of finite thickness are considered. A detailed scaling theory, with allowance for distinct exponents λ and $\theta = 1/\nu$ for the critical-point shift and rounding, respectively, is confirmed by exact calculations on d-dimensional ferromagnetic spherical models and ideal Bose fluids with various boundary conditions. Ising-model results and existing data on real helium films are consistent with the theory.

Improvements in experimental technique should soon allow the detailed and accurate study of critical phenomena in systems with one or more finite, even though microscopically large, dimensions. Both experimentally and conceptually, the simplest systems to consider are films of finite thickness $L = na$ (where a is an atomic length or lattice spacing), and essentially infinite extent in the remaining $d' = d - 1$ dimensions (with $d = 3$ for real films). In this note we discuss such experiments theoretically.[1]

At the outset one should[2] generally distinguish a fractional *shift* $\epsilon(n)$ in critical temperature (or quasicritical temperature[3]) from a fractional *rounding* $\delta(n)$. If $T_c^{\tau}(n)$ is the critical temperature of the finite-thickness film under boundary conditions denoted by τ (see below), and $T_c = T_c^{\tau}(\infty)$ is the corresponding bulk critical temperature, the fractional shift is defined by

$$\epsilon^{\tau}(n) = [T_c - T_c^{\tau}(n)]/T_c \approx b^{\tau}/n^{\lambda} \text{ as } n \to \infty, \quad (1)$$

where the expected asymptotic behavior for thick films ($n \to \infty$) is characterized by a shift exponent λ. To define the rounding, consider an intensive property Y (such as the specific heat C, or the reduced susceptibility, $\bar{\chi} = k_B T \chi$) which in the bulk system has a critical-point divergence

$$Y_{\infty}(T) \approx A t^{-\psi} \text{ as } T \to T_c+, \quad (2)$$

where $t = (T - T_c)/T_c$ is the reduced temperature deviation (and $\psi = \alpha_d$ or γ_d, if $Y = C$ or $\bar{\chi}$, respectively[4]). In terms of the corresponding finite-thickness property $Y^{\tau}(n, T)$, the rounding may be defined, albeit a little loosely, by

$$\delta^{\tau}(n) = \Delta T^{*\tau}(n)/T_c \approx c^{\tau}/n^{\theta} \text{ as } n \to \infty, \quad (3)$$

where $T^{*\tau}(n) = T_c^{\tau}(n) + \Delta T^{*\tau}(n)$ is the temperature at which $Y^{\tau}(n, T)$ first shows significant (relative order unity) deviations from the bulk limit $Y_{\infty}(T)$. Note that this "rounding" is measured relative to the *shifted* critical temperature $T_c^{\tau}(n)$ and, in effect, measures the region of "crossover" from the bulk behavior (2) to the characteristic $d' = (d-1)$-dimensional film behavior

$$Y^{\tau}(n, T) \approx \dot{A}^{\tau}(n) \dot{t}^{-\dot{\psi}} \text{ as } T \to T_c^{\tau}(n), \quad (4)$$

where the *shifted* reduced temperature deviation is

$$\dot{t} = [T - T_c^{\tau}(n)]/T_c = t + \epsilon^{\tau}(n). \quad (5)$$

The d'-dimensional exponent satisfies $\dot{\psi} = \alpha_{d-1}$ or γ_{d-1} if $Y = C$ or $\bar{\chi}$, respectively.

Fisher and Ferdinand[2] suggested that, in general, the rounding exponent θ in (3) might be larger than λ so that *the shift is asymptotically larger than the rounding.* They argued for the relation

$$\theta = 1/\nu_d, \quad (6)$$

which follows from the hypothesis that rounding should set in when the correlation length $\xi(T) \approx \xi_0/\dot{t}^{\nu_d}$ matches the thickness $L = na$.[5] On the other hand, for free boundaries, they conjectured that λ equals 1, which is generally less than $1/\nu_d$. An exact analysis of the plane-square Ising lattice[2,6] confirmed (6), but could not distinguish the conjecture for λ since $\nu_d = 1$ in that case.

We now report calculations[7] for d-dimensional ferromagnetic spherical models[8] and ideal Bose fluids[5] which confirm the possibility $\theta > \lambda$. Three types of boundary conditions have been considered:

($\tau = 0$) *periodic* (or cyclic), in which the first and nth lattice layers are coupled ferromagnetically as nearest neighbors,[8] or the Bose wave functions satisfy $\Psi_N(x_j + L) = \Psi_N(x_j)$ (all $j = 1, \cdots, N$);

($\tau = \frac{1}{2}$) *antiperiodic*,[9] in which the first and nth lattice layers are coupled *anti*ferromagnetically with the interaction $-J$,[8] or $\Psi_N(x_j + L) = -\Psi_N(x_j)$; and

($\tau = 1$) *free surfaces* (or hard walls), where the first and nth lattice layer each couple to only one adjacent layer, or $\Psi_N(x_j = 0) = \Psi_N(x_j = L) = 0$.

The resulting exact asymptotic behavior of $\epsilon^{\tau}(n)$ and $\delta^{\tau}(n)$ is exhibited in Table I.[10] By comparison with the second column, which lists the

TABLE I. Asymptotic behavior of critical-point shift and rounding for spherical models and ideal Bose fluids. (Note there is no sharp transition when $d=2$.)

Dimension d	Correlation exponent ν_d	Rounding $\delta^\tau(n)$	Shifts $\epsilon^\tau(n)$ $\tau=0$	$\tau=\frac{1}{2}$	$\tau=1$	Surface exponent $\gamma_d^\times$
3	1	$\sim n^{-1}$	$b_3^{\,0}/n$	$b_3^{\,1/2}/n$	$-\lvert b_3^{\,1}\rvert(\ln n)/n$	3 (×log)
4	$\frac{1}{2}(\times\log^{1/2})$	$\sim(\ln n)/n^2$	$b_4^{\,0}/n^2$	$b_4^{\,1/2}(\ln n)/n^2$	$-\lvert b_4^{\,1}\rvert/n$	2 (×log)
5	$\frac{1}{2}$	$\sim n^{-2}$	$b_5^{\,0}/n^3$	$b_5^{\,1/2}/n^2$	$-\lvert b_5^{\,1}\rvert/n$	2
≥6	$\frac{1}{2}$	$\sim n^{-2}$	$b_d^{\,0}/n^{d-2}$	$b_d^{\,1/2}/n^2$	$-\lvert b_d^{\,1}\rvert/n$	2

correlation length exponents ν_d,[8] we see that in all cases the rounding exponent satsifies the matching relation (6). The behavior of the shift is more subtle: For antiperiodic conditions we have $\epsilon^{1/2}(n)\sim\delta^{1/2}(n)$ and $\lambda=\theta=1/\nu_d$, in accord with the analogous matching hypothesis for the shift. Under periodic boundary conditions, however, we find $\lambda=d-2$, so that, for $d\geq 4$, the shift is asymptotically much smaller than the rounding. We know of no simple heuristic argument for this effect, although it has also been noticed in series calculations for Ising[11] and Heisenberg[12] ferromagnetic films for $d=3$, where $\lambda\approx 2.0$. Finally, for the more realistic free-surface conditions we find

$$\lambda=1\ (\tau=1,\ \text{all } d), \tag{7}$$

although there is an additional logarithmic factor for $d=3$; the shift $\epsilon^1(n)$ is thus always asymptotically *larger* than the rounding $\delta^1(n)$, as anticipated. Indeed, the value (7) is in accord with the conjecture[2] mentioned, although for $d\geq 4$ the *sign* of the shift, which implies $T_c^{\,1}(n)>T_c$ for $n\gg 1$, is in direct contradiction with the "naive mean-field arguments"[2] originally adduced. The rather surprising enhancement of T_c for large n seems to be related to the constraint (imposed for all n) of constant particle density in the ideal Bose fluid, and of constant mean-square spin magnitude in the spherical model. The value $\lambda=1$ may thus not apply to fluid films observed under constant pressure[13] or to more realistic "fixed-spin" models; indeed, numerical evidence for the three-dimensional Ising model indicates $\lambda=1/\nu_3\simeq 1.56$ $(\tau=1)$.[1,11]

More detailed predictions and more precise definitions of $\delta(n)$ and $\epsilon(n)$ can be made on the basis of the explicit scaling postulate[1,14]

$$Y^\tau(n,T)\approx n^\omega X^\tau(n^\theta\dot{t})\ \text{as}\ n\to\infty,\ \dot{t}\to 0, \tag{8}$$

where we will accept the matching argument leading to (6). Note that the shifted temperature variable $\dot{t}$ appears; if this were replaced by t, we would be forced to conclude $\lambda=\theta$. The asymptotic behavior of the scaling functions $X^\tau(x)$ follows from the requirements that (8) reproduce both (2) and (4). This yields

$$X^\tau(x)\approx X_\infty x^{-\psi}\ \text{as}\ x\to\infty, \tag{9}$$

$$\approx X_0^{\,\tau}x^{-\dot{\psi}}\ \text{as}\ x\to 0, \tag{10}$$

where $X_\infty=A$, and one must have

$$\omega=\theta\psi=\psi/\nu \tag{11}$$

(so that $\omega=2-\eta$ when $Y=\overline{\chi}$). Evidently $X^\tau(x)$ describes the "shape" of the crossover from d-dimensional to d'-dimensional critical behavior as $T\to T_c^{\,\tau}(n)$. We can now make the amplitude prediction

$$\dot{A}^\tau(n)\approx X_0^{\,\tau}n^{(\psi-\dot{\psi})/\nu}\ \text{as}\ n\to\infty. \tag{12}$$

Note that when $\dot{\psi}=0$ the amplitude $\dot{A}^\tau(n)$ is just the critical-point value $Y_c^{\,\tau}(n)$.

The case $\psi=0$ usually corresponds to a bulk logarithmic singularity

$$Y_\infty(T)=A\ln(t^{-1})+\cdots\ \text{as}\ t\to 0. \tag{13}$$

To accommodate such a weak singularity, the "background" terms

$$-n^\omega X^\tau(n^\theta\dot{t}_0)+Y^\tau(n,T_0) \tag{14}$$

should be added to the postulate (8). Here T_0 is a fixed noncritical reference temperature. Then (13) requires $\omega=0$ and the replacement of (9) by $X^\tau(x)\approx A\ln x^{-1}$ $(x\to\infty)$. An interesting application can then be made to the specific heat ($Y=C$, $\psi=\alpha=0$) of a planar Ising model[5] and a helium film.[13] In these cases $\dot{\psi}$ vanishes and we conclude from (8) and (14) that

$$Y_c(n)=Y_{\max}(n)\approx(A/\nu)\ln n+\cdots. \tag{15}$$

This prediction for the specific-heat maximum is confirmed by the Ising-model results[5] $(\nu_2=1)$. It is also consistent with the helium data[13] and the theoretically expected value $\nu_3\simeq\frac{2}{3}$, as shown by Moore.[15] However, more extensive experiments are desirable: Indeed, accepting (15) for helium would allow one to determine directly the otherwise inaccessible correlation exponent ν_3.

As $n\to\infty$ at fixed T in a film with free boundaries, a surface contribution can be defined by

$$Y^{\times}(T)\approx \tfrac{1}{2}n[Y^1(n,T)-Y_\infty(T)], \tag{16}$$

with corresponding exponent $\psi^{\times}$ and amplitude $A^{\times}$. This generally entails a higher-order term $X_\infty{}^{\times}x^{-\varphi}$ in (9), which then leads to[1] the alternative predictions[16]

$$\psi^{\times}=\psi+1,\quad A^{\times}=-\tfrac{1}{2}\psi b^1 A,\quad \text{if } \lambda=1 \text{ and } \nu_d<1, \tag{17}$$

$$\psi^{\times}=\psi+\nu_d,\quad A^{\times}=\tfrac{1}{2}X_\infty{}^{\times},\ \text{if } \lambda>1. \tag{18}$$

We have checked the predictions (17) in detail for the spherical and ideal Bose gases by direct calculation[6] of the surface susceptibilities $\bar{\chi}^{\times}(T)$: See the last column of Table I where $\gamma^{\times}$ is listed. An appropriately adapted scaling theory even accounts correctly for the observed logarithmic factors for $d\leq 4$ and for the leading corrections to $\bar{\chi}^{\times}(T)$ as $T\to T_c$ [which are generally in accord with (18)]. The surface specific heat of the plane-square Ising model[2,5] also verifies the theory.

Finally, we have been able to check the full scaling postulate (8) by calculating asymptotically the detailed critical behavior in d dimensions as $n\to\infty$. For $d=3$ the postulate applies precisely and the explicit susceptibility scaling functions for the spherical model ($\gamma_3=2$, $\gamma_2=0$)[8] can be given as

$$2JX_3{}^0(x)=\{2\sinh^{-1}[\tfrac{1}{2}\exp(4\pi K_c x)]\}^{-2}, \tag{19}$$

with $b_3{}^0=0$, and, parametrically for free surfaces,

$$\begin{aligned} 2JX_3{}^1(x)&=y^{-2}[1-2y^{-1}\tanh(\tfrac{1}{2}y)],\\ 8\pi K_c x&=\ln[(\sinh y)/y], \end{aligned} \tag{20}$$

with $b_3{}^1=-1/4\pi K_c$, where $K_c=J/k_BT_c$. When $x\to\infty$ these expressions reproduce (9). Furthermore, in the case of free surfaces ($\tau=1$) the leading correction term is in exact accord with the scaling argument sketched above and the calculated surface susceptibility. Conversely, for periodic boundary conditions the corrections to (9) are exponentially small (for all d). This means, as in the Ising model,[5] that for fixed $T>T_c$, the function $Y^0(n,T)$ approaches its limit $Y_\infty(T)$ as $\exp(-cnt^{\nu})$, i.e., *exponentially fast.* This is probably a rather general result for systems with periodic boundary conditions.

A finite Bose or spherical model film in three dimensions has no phase transition (for $T>0$) so $\dot{\psi}=0$. However, as $x\to-\infty$ the scaling functions (19) and (20) give the correct behavior of $\bar{\chi}(n,T)$ for $n\gg 1$ as $T\to 0$. In this sense the "critical region" of the film extends from $T=T_c$ down to $T=0$. Explicit expressions similar to (19) and (20) have also been found[6] for the scaling functions for the specific heat of ideal Bose films.

In five or more dimensions the detailed scaling predictions are again confirmed. In four dimensions a scaling form based on the variable $x=L/\xi(\dot{t})$ describes the rounding and crossover region correctly. However, the same form fails, by logarithmic corrections, to reproduce precisely the limits $n\to\infty$ at fixed $T>T_c$, or $T\to T_c{}^{\tau}(n)$ at fixed n. In view of similar difficulties with thermodynamic scaling[5] for $d=4$ this is not so surprising. In summary, the overall agreement of the exact calculations with the scaling hypotheses is most gratifying.

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[1]A survey of the previous theoretical situation and more detailed discussion of some of the results reported here will appear in M. E. Fisher, in Proceedings of the Enrico Fermi International School of Physics, Course No. 51 (to be published).

[2]M. E. Fisher and A. E. Ferdinand, Phys. Rev. Lett. 19, 169 (1967). Note the sign of the exponent is in error in Eq. (14).

[3]In the case where, for finite L or n, the system has no sharp transition, one can normally define a pseudocritical point: See also Eq. (20) below.

[4]We use the standard exponent definitions: See, e.g., M. E. Fisher, Rep. Progr. Phys. 30, 615 (1967).

[5]See also A. E. Ferdinand and M. E. Fisher, Phys. Rev. 185, 832 (1969).

[6]M. N. Barber and M. E. Fisher, to be published.

[7]We consider d-dimensional hypercubical lattices with nearest-neighbor ferromagnetic interactions of strength $J>0$: See G. S. Joyce, Phys. Rev. 146, 349 (1966).

[8]For an ideal Bose fluid we may take $a=(h^2/2\pi m k_BT)^{1/2}$; see also J. D. Gunton and M. J. Buckingham, Phys. Rev. 166, 152 (1968).

[9]As will be described elsewhere, antiperiodic conditions are particularly useful for calculating the "helicity modulus" or, for a Bose fluid, the superfluid density $\rho_s(T)$.

[10]These results were announced by the authors in Proceedings of the IUPAP Conference on Statistical Mechanics, Chicago, April 1971 (to be published).

[11]G. A. T. Allan, Phys. Rev. B 1, 352 (1970); for further developments, see Ref. 1. For ideal Bose gas films the asymptotic behavior of $\epsilon^T(n)$ has also been investigated more recently by R. K. Pathria, Phys. Lett. 35A, 351 (1971).

[12]D. S. Ritchie and M. E. Fisher, in *Magnetism and Magnetic Materials—1971*, AIP Conference Proceedings No. 5 (American Institute of Physics, New York, 1972).

[13]D. F. Brewer, J. Low Temp. Phys. 3, 205 (1970).

[14]M. A. Moore, Phys. Lett. 37A, 345 (1971).

[15]We assume a zero ordering field ζ. Nonzero values can be included by allowing X^T to depend also on the standard thermodynamic scaling variable $y=\zeta/t^\Delta$, where $\Delta=\beta+\gamma=\beta\delta$.

[16]The proposal (18), but without the condition $\lambda>1$, was first advanced by P. G. Watson [J. Phys. C: Proc. Phys. Soc., London 1, 268 (1968)], on less general grounds. Watson (private communication) has since withdrawn his claim that it is correct for a spherical model.

X-Ray Brillouin Scattering

P. Eisenberger, N. G. Alexandropoulos, and P. M. Platzman
Bell Laboratories, Murray Hill, New Jersey 07974
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An investigation of x-ray scattering, when the Bragg condition is nearly satisfied, reveals the possibility of studying phonon phenomena at momenta as small as 10^4 cm^{-1}. A closer study of elastic Bragg scattering in that region provides an explanation for the previously observed "coherent crystal radiation."

In the past, x-ray and neutron investigations[1-3] of phonon phenomena have only been extended to momentum transfers as small as 10^6 to 10^7 cm^{-1}, which is about 100 times larger than that measured by laser techniques. In the x-ray case, it was expected that a large background produced by elastic Bragg scattering, which was known to be experimentally indistinguishable on the basis of energy considerations from the inelastic thermal diffuse scattering (TDS), would obscure the signal. In the neutron case, the finite energy and angular resolution typically employed in neutron scattering together with the large specimens required also restricted the range of investigation to relatively large values.

In this work, we show how a closer study of elastic and TDS components when the Bragg condition is nearly satisfied (i.e., when the momentum transfer is almost equal to a reciprocal-lattice vector $\vec{G}$) shows that the two components have different phase-matching conditions. Experimentally, when we investigated phenomena at $\vec{G}\pm\vec{q}$, where $\vec{q}$ was on the order of 10^4 cm^{-1}, the two components were easily separated by using a triple-crystal spectrometer. The region of q space that can be investigated depends primarily upon the perfection of the crystals, the degree of input collimation, and not on the intrinsic width of the Bragg peak. In addition, these studies provide an explanation for the previously reported "coherent crystal radiation"[4] and have ramifications for the general field of x-ray diffraction.

We consider the situation where a well-defined (infinitely collimated) beam $\vec{K}_{in}$ is incident on a perfect crystal. The crystal has its surface cut perpendicular to a reciprocal-lattice vector $\vec{G}$. Neglecting the small phonon energy and refraction effects, the basic equations governing both Bragg scattering and TDS are

$$\vec{K}_{out}-\vec{K}_{in}=\vec{G}+\vec{q}, \quad |\vec{K}_{in}|=|\vec{K}_{out}|=K. \tag{1}$$

Here $\vec{K}_{out}$ is the scattered-photon's wave vector and $\vec{q}$ is either the spread in G associated with the interaction effects described by dynamical[5,6] theory, or a phonon wave vector. We are interested in the region where the angular deviation of the input δ_{in} and the output δ_{out} beams from the Bragg angle θ_B are small. The angles are defined by the following relations[7]:

$$\begin{aligned}|\vec{G}|&=2|K|\sin\theta_B,\\ \vec{K}_{in}\cdot\vec{G}&=-|K||\vec{G}|\sin(\theta_B+\delta_{in}),\\ \vec{K}_{out}\cdot\vec{G}&=|K||\vec{G}|\sin(\theta_B+\delta_{out}),\\ \vec{G}\cdot\vec{q}&=|\vec{G}||\vec{q}|\cos\theta_q.\end{aligned} \tag{2}$$

All the vectors are in the same plane (Fig. 1).

As a result of the strong interaction of the x