Susceptibility Singularities at First-Order Phase Transitions

D. B. Abraham and P. J. Upton
Department of Theoretical Physics, 1, Keble Road, Oxford, OX1 3NP, England
(Received 6 April 1992)

The bubble picture for pair-spin correlation functions is applied to two-dimensional Ising-like models, at subcritical temperatures, in an external field h, from which we show, via the fluctuation sum, that the susceptibility $\chi(h)$ has an *essential singularity* at $h=0$. This is studied further using a solid-on-solid bubble model for (a) a *restricted* ensemble corresponding to metastability, where $h=0$ is found to be the limit point of an infinite number of poles of χ along the negative real axis, and (b) an *unrestricted* ensemble, where a Yang-Lee circle theorem is found.

PACS numbers: 05.50.+q, 05.70.Fh, 64.60.−i, 75.10.Hk

The droplet model of condensation, as well as a theory of the critical point, received crucial impetus some years ago: Andreev [1] and Fisher [2] used it to confirm Mayer's conjecture that the condensation point is a mathematical singularity of a suitable free energy (in the thermodynamic or infinite-volume limit), although a weak one of essential type in the fugacity variable. At that time it had already become clear, in the work of Peierls [3] and then Onsager [4] on the planar Ising model free energy followed by that of Yang and Lee [5,6], that a substantial theory of phase transitions could be constructed based on partition functions without *ad hoc* restrictions of the ensemble but with the essential requirements of the infinite-volume limit being clearly identified [7].

The work of Andreev [1] and Fisher [2] focused attention on the heresy of van der Waals loops—the Maxwell construction is needed to make a proper theory which avoids a negative compressibility. Nevertheless, when mean-field theory is formulated correctly (in one dimension) [8] it does indeed contain a Maxwell construction. But there was still a misplaced desire to attribute significance, even virtue, to loops as smooth extensions into a metastable region, possibly requiring a phase restriction in the ensemble of the partition function to do so. The droplet singularity makes this impossible; any continuation must be in the complex plane.

As a bonus, Fisher's [2] work gave a derivation of some scaling laws between critical exponents, even though the excluded-volume interactions between different droplets had to be neglected for technical reasons. The treatment was also in a thermodynamic spirit—the correlation-function fluctuation-sum route not followed. In this Letter we reexamine a bubble model of correlation functions [9,10] for the planar Ising and related models with an external magnetic field. We obtain a new closed form expression for the magnetic susceptibility and we use it to extract a Yang-Lee circle theorem and the associated density of zeros [6(b)]. We also examine the analog of the droplet singularity [1,2] in this system.

Consider a planar Ising model with a bulk plus phase. Throughout this paper we shall keep the temperature T below its critical value T_c. As has been explained elsewhere, if we perform a coarse graining of the system on a scale of the bulk correlation length [9], then the Peierls contours [3] (or equivalently the low-temperature series in the graphical representation of spin configurations) contributing to the two-point correlation function reduce asymptotically to a simple loop Γ which passes through the two points being correlated, (0,0) and (x,y); Γ also partitions the plane into regions of opposite magnetization, the value of which is determined by the small contours which are scaled away and the boundary condition. The loop Γ has a Boltzmann statistical weight with energy $E(\Gamma)$ where

$$\beta E(\Gamma)=\tau L(\Gamma)+2mhA(\Gamma). \qquad (1)$$

Here $L(\Gamma)$ and $A(\Gamma)$ are the length and area of the loop, m is the magnetization outside Γ, τ is the surface tension, and h is the external field in units of $k_BT=1/\beta$. The magnetization m is usually assigned its *spontaneous* value m^* [5]; this should be valid for small enough $h\geq 0$, but other choices will be considered below. Since $T<T_c$, $h=0$ defines the first-order phase boundary. The truncated pair-spin correlation function is then

$$u_2(x,y)=m^2\sum_{\Gamma}\exp[-\beta E(\Gamma)], \qquad (2)$$

where the sum is over all loops passing through (0,0) and (x,y). The susceptibility, $\chi(h)\equiv\partial m/\partial h$, is determined from the fluctuation sum,

$$\chi(h)=\sum_{x=-\infty}^{\infty}\sum_{y=-\infty}^{\infty}u_2(x,y). \qquad (3)$$

The kth derivative of u_2 with respect to h can be expressed in terms of moments of $A(\Gamma)$:

$$\frac{\partial^k u_2}{\partial h^k}=(-2m^*)^k\langle A^k(\Gamma)\rangle u_2(x,y) \qquad (4)$$

assuming that $m=m^*$ (independent of $h\geq 0$). Here, $\langle\cdots\rangle$ denotes the ensemble average with respect to the Boltzmann distribution given from (1). From convexity inequalities, we have $\langle A^k\rangle\geq\langle A\rangle^k$. For large $r=(x^2+y^2)^{1/2}$ and $h=0$, u_2 takes the Kadanoff-Wu form

$u_2 \sim e^{-2\tau r}/r^2$ [11]. Random-walk ideas imply that the mean bubble shapes are elliptical when $h=0$ [10] giving $\langle A\rangle \sim r^{3/2}$ for large r. Hence, from (3) and (4), it follows that

$$(-1)^k \frac{\partial^k \chi}{\partial h^k}\bigg|_{h=0+} \geq c_0 \left[\frac{2c_1 m^*}{(2\tau)^{3/2}}\right]^k \Gamma(3k/2), \qquad (5)$$

where c_0 and c_1 are positive and independent of k and $\Gamma(z)$ is the gamma function. This implies that the Taylor expansion about $h=0$ has a zero radius of convergence and therefore that $h=0$ is a nonanalytic point of $\chi(h)$. Below, we shall investigate the detailed nature of this singular point by considering specific implementations of the bubble picture. For the nearest-neighbor planar Ising model, Isakov [12] proved that the right-hand side of (5) diverges with k as $(k!)^2$, for *sufficiently low* temperatures. This result was previously anticipated by low-temperature series expansions [13]. We therefore suspect that the convexity inequality used above is rather weak. For the bubble model, general arguments [14] imply that $\chi(h)$ is analytic for all $\mathrm{Re}(mh)>0$. In this case, the gamma function in (5) would be replaced by $\Gamma(k)$ since here $\langle A\rangle \sim r$ for large r because the field would tend to pull opposite sides of the bubble together [10].

To evaluate (2) the configurations of the bubble are simplified by making the solid-on-solid restriction with respect to the (1,0) direction. We perform a coarse graining on a grid $l\times l$ with $x=Nl$, $y=Ml$, and require that the bubble intersect each vertical line $x=jl$ ($j=0, 1, \ldots, N$) just twice, with intercepts $y_j^>$ and $y_j^<$ satisfying $y_j^> \geq y_j^<$. Then

$$\beta E(\Gamma) = E^> + E^< + 2mhl \sum_{j=0}^{N} (y_j^> - y_j^<) \qquad (6)$$

with

$$E^> = \tau \sum_{j=1}^{N} \Phi(y_j^> - y_{j-1}^>) \qquad (7)$$

and $\Phi(y) = (l^2+y^2)^{1/2}$. Assuming l is large enough—we expect it to be proportional to the bulk correlation length—so that y/l is typically small, a quadratic approximation to $\Phi(y)$ gives a transfer-integral problem for evaluation of (2) which can be separated by going to center-of-mass and relative coordinates. The former gives an elementary, field-independent problem. For the latter, a further simplification is needed: Take $\Phi(y) = a + b|y|$. The parameters a, b, and l (we expect and confirm that l is proportional to the bulk correlation length ξ) are then chosen by fitting the $h=0$ result to the exact two-fermion sector contribution to $u_2(x,y)$. The solution of the $h>0$ problem is given elsewhere [9,14]:

$$u_2(Nl,Ml) = \frac{m^2}{2^{1/3}\pi a} \int_{-\infty}^{\infty} \frac{\exp(iM\omega)d\omega}{(1+2^{-4/3}\omega^2)^N} \sum_{j=1}^{\infty} \left(\frac{c}{v_j}\right)^{2N}, \qquad (8)$$

where $a = 2\tau^2/mh$, $l^{-1} = 2^{2/3}\tau$, $c = \exp(-2^{-2/3})$, and v_j are solutions of $J_{a-1}(av_j)=0$ with $J_a(z)$ being the Bessel function of order a in standard notation. Note that a is a *scaling* variable and its dependence on h through mh is important. For large x, $u_2(x,0) \sim hx^{-1/2} \times \sum_{j=1}^{\infty} \exp(-m_j x)$ so that it has a "mass" spectrum given by $m_j = 2\tau(1+2^{2/3}\ln v_j)$.

Some remarks are in order.

(i) The bubble can be described by a functional integral [14]; the occurrence of Airy functions is natural because the associated Schrödinger equation [in the quadratic approximation to $\Phi(y)$] has a linear potential. This approach agrees with (8) in the $a\to\infty$ limit and the masses also agree with McCoy and Wu [15]. The problems with the functional-integral approach are the lack of convergence of the fluctuation sum of $u_2(x,y)$ without a cutoff (which then occurs explicitly in the susceptibility) and the implied requirement to take $l\to 0$ which is unphysical.

(ii) If $u_2(x,y)$ is written as a dispersion series from the exact solution (in fermions) [16] the leading term to contribute is the two-particle one. For $(x^2+y^2)^{1/2} > \xi$ it bounds the series sum so closely that the critical amplitude of the susceptibility [17] is given to better than 1%. Examination of the excess-energy-density distribution in the two-particle sector agrees with the bubble picture [14].

(iii) A realization of the Weeks columnar picture [18] allows the construction of $\Phi(y)$ from first principles [14,19] and gives the weighting of the integration variables *a priori*.

(iv) As stated above, we choose a, b, and the coarse-graining length l to get $u_2(x,0)$ for the bubble picture to *agree exactly* with the asymptotic behavior of the Ising two-fermion sector result as $|x|\to\infty$. This agreement is not related to the exact reproduction of the Ising model surface tension by the solid-on-solid model; in that case, $l=1$ and $\tau a = \tau b = 2\beta J$ where J is the spin-spin interaction energy in the Ising model, whereas ours has $l \propto \xi$. The correct asymptotic decay of $u_2(x,0)$ is not recaptured in the $l=1$ case.

Since N takes positive integral values and M is taken to be continuous, the fluctuation sum (3) becomes

$$\chi(h) = 2l^2 \sum_{N=1}^{\infty} \int_{-\infty}^{\infty} dM\, u_2(Nl,Ml). \qquad (9)$$

Substituting (8) into (9) and applying the Mittag-Leffler theorem [20] leads to the following closed-form expression for χ:

$$\frac{\tau^2\chi(h)}{m^2} = \frac{cJ_a(ca)}{2^{2/3}J_{a-1}(ca)}, \quad a = \frac{2\tau^2}{mh}. \qquad (10)$$

Note that χ has the expected *scaling* form $\chi \sim t^{-\gamma} X(h/t^{\Delta})$ where $t \equiv (T_c - T)/T_c$ and h are small with critical exponents $\gamma = \frac{7}{4}$ and $\Delta = \frac{15}{8}$. The analytic character of the right-hand side of (10) in the complex a plane can immediately be seen by noting that it can be written as a quotient of two uniformly convergent series for all $\alpha \in \mathbb{C}$ [20]. It is therefore a meromorphic function of α with simple poles at the zeros of $J_{\alpha-1}(c\alpha)$. Since we know that $\chi(h)$ is analytic for $\mathrm{Re}(mh)>0$ then $J_{\alpha-1}(c\alpha)$ should have no zeros for $\mathrm{Re}\alpha>0$. For α real positive, this was already known to be true provided $c<1$ [20]. However, there are zeros for $\mathrm{Re}\alpha<0$. These can be located by first writing

$$J_{\alpha-1}(c\alpha)=e^{i\pi\alpha}[\cos(\pi\alpha)J_{1-\alpha}(-c\alpha)+\sin(\pi\alpha)Y_{1-\alpha}(-c\alpha)], \tag{11}$$

which relates the Bessel functions $J_\alpha(z)$ and $Y_\alpha(z)$. We now examine (11) for large $|\alpha|$ with $\mathrm{Re}\alpha<0$. Since $c<1$, as $|\alpha|$ increases $J_{1-\alpha}(-c\alpha)$ vanishes exponentially while $Y_{1-\alpha}(-c\alpha)$ grows exponentially and both are monotonic [21]. The second term of (11), which dominates for large $|\alpha|$, is zero for $\alpha=-n$ where $n\in\mathbb{Z}_+$. Hence, by Rouché's theorem, the zeros of $J_{\alpha-1}(c\alpha)$ occur on the real axis near $\alpha\sim -n$ when $|\alpha|$ is large. Thus the origin of the $1/\alpha$ plane is a limit point of poles along the negative real axis and any neighborhood of the origin contains an infinite number of poles and hence this point is an essential singularity. We now explain how the above considerations relate to singularities at first-order phase transitions. To do this, we consider the following two ways of analytically continuing m off the real axis of the h plane.

Case (a).— Here we *restrict* the ensemble so that the magnetization stays positive when h takes negative real values. This facilitates a discussion of metastability. To do this we construct the bubble model so that $m=m^*$ for all $h\in\mathbb{C}$, in which case we can have $\mathrm{Re}\alpha<0$. Hence $\chi(h)$ has simple poles along the negative real axis of the h plane when

$$m^*h/2\tau^2\sim -1/n \tag{12}$$

for large $n\in\mathbb{Z}_+$. Thus, $h=0$ is the limit point of an infinite number of poles along the negative real axis and is therefore an essential singularity. This is reminiscent of singular behavior found through numerical studies of the two-dimensional Ising transfer matrix and theoretical work on two-level models [22]. It is also analogous to the singularity found in the droplet model [1,2] and those found in field-theoretical models of condensation [23] as predicted by approximate instantonlike solutions—in these cases one finds that the negative real axis forms a cut rather than a line of poles.

Case (b).— We now work with an *unrestricted* ensemble for which one can show in the Ising case that $m(-h)=-m(h)$ and $\chi(h)=\chi(-h)$ for all $h\in\mathbb{C}\backslash\mathrm{Im}h$ on a finite lattice and also on the infinite lattice provided the limits exist [6(b)]. The bubble model is constructed first in the half plane $\mathrm{Re}h>0$ by analytic continuation from the real axis (on which $m=m^*$) to the entire half plane. In the half plane $\mathrm{Re}h<0$ we put $m=-m^*$ on $\mathrm{Im}h=0$ and then continue to the entire left half plane. Thus, since now $\mathrm{Re}\alpha>0$ for all $h\in\mathbb{C}\backslash\mathrm{Im}h$, the only singularities can be on $\mathrm{Re}h=0$, equivalent to a circle theorem. There is a jump discontinuity on this line given by $\delta\chi(ih)=\lim_{\epsilon\to 0}[\chi(ih+\epsilon)-\chi(-ih+\epsilon)]$ using the even character of $\chi(h)$, so that the asymptotics given by Olver for Bessel functions [21] can be applied, giving

$$\frac{2^{2/3}\tau^2}{(m^*)^2}\delta\chi(ih)\sim\frac{(1-c^2)^{1/2}}{2i\zeta^{1/2}\tilde{\alpha}^{1/3}}\,\mathrm{Im}\left[\frac{e^{i\pi/6}\mathrm{Ai}'(\tilde{\alpha}^{2/3}\zeta e^{-i\pi/3})}{\mathrm{Ai}(\tilde{\alpha}^{2/3}\zeta e^{-i\pi/3})}\right],\quad \tilde{\alpha}=\frac{2\tau^2}{m^*h} \tag{13a}$$

for large $\tilde{\alpha}\to\infty$ where $\mathrm{Ai}(z)$ is the Airy function and ζ is a number given by

$$2\zeta^{3/2}/3=\ln\{[1+(1-c^2)^{1/2}]/c\}-(1-c^2)^{1/2}. \tag{13b}$$

The Yang-Lee circle theorem [6(b)], for a finite lattice, states that partition-function zeros (and therefore free-energy singularities) are found only along the imaginary axis of h. Following Yang and Lee [6(b)], in the thermodynamic limit, these zeros form a distribution characterized by a density $g(\theta)$ where $g(\theta)d\theta$ gives the number of zeros per spin site for $-2h$ between $i\theta$ and $i(\theta+d\theta)$. This can be obtained from $\chi(h)$ through

$$4\pi g'(\theta)=\mathrm{Im}\chi(-i\theta/2+0) \tag{14}$$

with the boundary condition $g(0)=m^*/2\pi$. Substituting (10) into (14) leads to an expression for $dg/d\theta$ which can be expressed in terms of Airy functions as $m^*|\theta|/\tau^2\to 0$ [21]. Note that $g(\theta)$ has itself an essential singularity at $\theta=0$ reflecting the fact that the origin of the $1/\alpha$ plane is a limit point of poles. Moreover, we know on rigorous grounds [14], from the theorem of Isakov [12], that $g(\theta)$ has an essential singularity at $\theta=0$ for the d-dimensional Ising model for low enough temperature.

Observe that $m=m(h)$ can be determined in principle for larger values of h by *boot-strapping* equation (10). This involves writing $\chi\equiv\partial m/\partial h$ and solving the resulting differential equation for $m=m(h)$ [14].

We conclude by emphasizing that for the planar Ising model, with $T<T_c$, the bubble picture gives a very good description of two-point correlation functions at $h=0$; indeed it can even give *exact* results [24]. Furthermore, when $h\neq 0$, it predicts a mass spectrum which, as $h\to 0$, coincides *exactly* with that found by McCoy and Wu [15]. Therefore, while the bubble model is not expected to work so well for large h, it appears to be very reliable close to $h=0$. This is fortunate because it is the thermodynamics close to this point, at the first-order phase boundary, that has been the main focus of this paper. We have found, via the fluctuation sum applied to the bubble model, that $\chi(h)$ has an *essential singularity* at

$h=0$. Furthermore, the bubble model for an *unrestricted* ensemble leads to a Yang-Lee circle theorem, whereas *restricting* the ensemble in a way corresponding to metastability implies that $h=0$ is a limit point of an infinite number of poles [of $\chi(h)$] along the negative real axis—an analog of the droplet singularity. Finally, we stress that these results are specific to *two-dimensional* systems. Higher-dimensional models may well behave differently, but it is hoped that the ideas presented here remain applicable.

We wish to thank M. E. Fisher, C. M. Newman, and V. Privman for constructive comments. We acknowledge support from the Science and Engineering Research Council (United Kingdom) under Grant No. GR/G44017 and the Forschungszentrums Jülich for kind hospitality where part of this work was performed.

[1] A. F. Andreev, Zh. Eksp. Teor. Fiz. **45**, 2068 (1963) [Sov. Phys. JETP **18**, 1415 (1964)].
[2] M. E. Fisher, Physics (Long Island City, N.Y.) **3**, 255 (1967); in *Critical Phenomena*, International School of Physics "Enrico Fermi," Course LI, edited by M. S. Green (Academic, New York, 1971), p. 1.
[3] R. Peierls, Proc. Cambridge Philos. Soc. **32**, 477 (1936).
[4] L. Onsager, Phys. Rev. **65**, 117 (1944).
[5] C. N. Yang, Phys. Rev. **85**, 808 (1952).
[6] (a) C. N. Yang and T. D. Lee, Phys. Rev. **87**, 404 (1952); (b) T. D. Lee and C. N. Yang, Phys. Rev. **87**, 410 (1952).
[7] See D. Ruelle, *Statistical Mechanics: Rigorous Results* (Benjamin, Reading, MA, 1969); R. B. Griffiths, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and M. S. Green (Academic, New York, 1971), Vol. 1, and references therein.
[8] M. Kac, G. E. Uhlenbeck, and P. C. Hemmer, J. Math. Phys. **4**, 216 (1963).
[9] D. B. Abraham, Phys. Rev. Lett. **50**, 291 (1983).
[10] M. E. Fisher, J. Stat. Phys. **34**, 667 (1984).
[11] T. T. Wu, Phys. Rev. **149**, 380 (1966).
[12] S. N. Isakov, Commun. Math. Phys. **95**, 427 (1984); actually, the results here cover all dimensions greater than one.
[13] G. A. Baker, Jr., and D. Kim, J. Phys. A **13**, L103 (1980).
[14] D. B. Abraham and P. J. Upton (to be published).
[15] B. M. McCoy and T. T. Wu, Phys. Rev. D **18**, 1259 (1978).
[16] D. B. Abraham, Phys. Lett. **61A**, 217 (1977); Commun. Math. Phys. **59**, 17 (1978).
[17] T. T. Wu, B. M. McCoy, C. A. Tracy, and E. Barouch, Phys. Rev. B **13**, 316 (1976).
[18] J. D. Weeks, J. Chem. Phys. **67**, 3106 (1977).
[19] D. B. Abraham, Phys. Rev. Lett. **47**, 545 (1981).
[20] G. N. Watson, *A Treatise on the Theory of Bessel Functions* (Cambridge Univ. Press, London, 1952), 2nd ed.
[21] F. W. J. Olver, Philos. Trans. R. Soc. London A **247**, 328 (1954); see also *Handbook of Mathematical Functions*, edited by M. Abromowitz and I. A. Stegun (Dover, New York, 1965), Chap. 9.
[22] C. M. Newman and L. S. Schulman, J. Math. Phys. **18**, 23 (1977); R. J. McCraw and L. S. Schulman, J. Stat. Phys. **18**, 293 (1978).
[23] J. S. Langer, Ann. Phys. (N.Y.) **41**, 108 (1967); N. J. Günther, D. A. Nicole, and D. J. Wallace, J. Phys. A **13**, 1755 (1980).
[24] D. B. Abraham, N. M. Švrakić, and P. J. Upton, Phys. Rev. Lett. **68**, 423 (1992).