Nature of the "Griffiths" Singularity in Dilute Magnets

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Nature of the "Griffiths" Singularity in Dilute Magnets

Abstract
The nature of the singular behavior pointed out by Griffiths for H=0 in dilute magnets is investigated. It is argued that for concentration p less than that for formation of an infinite cluster, all derivatives of M(H) are finite. The nonanalyticity in M(H) is due to a branch cut along the imaginary H axis having weight \( \exp[-(\text{const})/|H|] \) for \( |H| \to 0 \), and is thus too weak to be experimentally observable. Some numerical and exact analytic results for the dilute magnet on a Bethe lattice are presented.

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Nature of the “Griffiths” singularity in dilute magnets

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The nature of the singular behavior pointed out by Griffiths for \( H = 0 \) in dilute magnets is investigated. It is argued that for concentration \( p \) less than that for formation of an infinite cluster, all derivatives of \( M(H) \) are finite. The nonanalyticity in \( M(H) \) is due to a branch cut along the imaginary \( H \) axis having weight \( \exp[-(\text{const})/|H|] \) for \(|H| \to 0\), and is thus too weak to be experimentally observable. Some numerical and exact analytic results for the dilute magnet on a Bethe lattice are presented.

I. INTRODUCTION

Recently there has been great interest in the precise nature of the singularity in the critical behavior of dilute magnets.\(^1\)\(^2\) Some years ago Griffiths\(^3\) showed that the free energy of a dilute ferromagnet is a nonanalytic function of magnetic field \( H \) at \( H = 0 \) for all temperatures below the transition temperature \( T_c^0 \) of the undiluted system. As yet, this singularity has not been detected either by high-temperature expansions\(^3\)\(^4\) or by renormalization-group methods.\(^3\)\(^4\)

Lifshitz\(^7\) has studied a related problem, namely, the density of states \( g(\omega) \) of an electron in a random potential. If the potential at each site \( V \) is distributed uniformly over an interval \( 0 \leq V \leq V_0 \), then the density of states near the lower band edge \( \omega_0 \) is of the form

\[
g(\omega) \sim e^{(\omega-\omega_0)^2} ,
\]

where \( \xi \) was determined by a single dimensional argument to be \( \frac{\xi}{2} \) for a three-dimensional system. This form results from the relatively infrequent occurrence of large regions having arbitrarily small values of \( V \).

For a ferromagnet it is known\(^8\)\(^9\) that the singularities in the free energy occur at imaginary \( H \), with a density at \( H = 0 \) which is proportional to the spontaneous magnetization.\(^8\) As we shall see, an argument similar to that of Lifshitz shows that the magnetization of a randomly dilute ferromagnet has a singularity of the form

\[
M(H) \sim \int_{-\infty}^{\infty} \frac{\rho(z)}{H-z} dz ,
\]

where \( \rho(z) = e^{-A|x|} \) for small \( z \) and \( A \) is nonzero for \( T < T_c^0 \).\(^10\) In contrast, Domb\(^11\) has recently proposed that the Griffiths singularity is a much stronger one, leading to a discontinuity in \( d^2 M/dH^2 \) at \( H = 0 \), whereas the arguments we give suggest that all derivatives of \( M(H) \) are smooth at \( H = 0 \) for \( p < p_c \), where \( p_c \) is the critical concentration for the formation of an infinitely large cluster. Our analysis and conclusions are very similar to those given by Fisher\(^12\) in his treatment of the cluster theory of condensation.

II. ANALYTIC PROPERTIES

Domb’s approach, which we follow here, is to write the magnetization for \( p < p_c \) as that of an assembly of separate finite clusters,

\[
M(H) = \sum_n W_n(p) M_n(H) ,
\]

where \( W_n(p) \) is the probability per site that a cluster of size \( n \) is formed and \( M_n(H) \) is the corresponding magnetization. Initially we will treat the case \( kT \ll J \), where \( 2J \) is the energy difference between parallel and antiparallel alignment of a pair of spins. Then \( M_n(H) \) depends only on \( n \) and not on the shape of the cluster. For an Ising system of spin \( \frac{1}{2} \) one has

\[
M_n(H) = \frac{1}{2} n \tanh(nH/2kT) ,
\]

whereas for a Heisenberg system of spin \( \frac{1}{2} \)

\[
M_n(H) = \frac{1}{2} \left( (n+1) \coth \frac{(n+1)H}{2kT} - \coth \frac{H}{2kT} \right) .
\]

Thus, for an Ising system one has

\[
M(H) = \frac{1}{2} \sum_{n=1}^{\infty} W_n(p) n \tanh \frac{nH}{2kT} .
\]

Clearly, the terms in Eq. (6) with \( n \) finite are analytic for \( H = 0 \), so it is only the arbitrarily large clusters which produce the Griffiths’ singularity. To study the nature of the singularity at \( H = 0 \) we need only to know the asymptotic form of \( W_n(n) \) for large \( n \). One can write \( W_n(p) \) in the form\(^11\)\(^12\)

\[
W_n(p) = \sum_s g(n, s) p^n (1-p)^s ,
\]

where \( g(n, s) \) is the number of cluster configurations per site having \( n \) sites and \( s \) bounding-surface sites. It is clear that \( \sum_s g(n, s) \) is less than the corresponding quantity for a Bethe lattice (Cayley tree) having the same number of bonds entering
a vertex. Thus it seems clear that
\[ W_n(p) = e^{-An} \]  
(8)

In three dimensions one would expect \( s \) in Eq. (7) to be of order \( n^{2/3} \), producing a factor \( e^{-n^{2/3}} \) in Eq. (8) which we drop, since the factor in Eq. (8) gives correctly the dominant behavior.

In fact, Fisher and Essam\(^\text{15}\) give the exact result for the Bethe lattice as
\[ W_n(p) = \frac{\sigma(n + 1)}{(n - 1)[\sigma(n + 1) + 1]} p \left( 1 - p \right) e^{-\sigma(n - 1)} \]  
(9)

where \( \sigma(n + 1) \) is the number of bonds which meet at each site. For large \( n \) one obtains Eq. (8) with
\[ A = (1 - \sigma) \ln \left( \frac{1 - p}{1 - \sigma} \right) - \ln p \sigma \]  
(10)

so that \( A > 0 \) for \( p \neq p_c \), where \( p_c = 1/\sigma \).

Thus the analytic properties of \( M(H) \) are determined by using Eq. (8) in Eq. (6). A convergent power series for \( M(H) \) at \( H = 0 \) does not exist, because \( M(H) \) has a branch cut along the imaginary \( H \) axis caused by the poles in \( \tan(nH/2kT) \) at rational values of \( H/\pi kT \). However, an asymptotic expansion for \( M(H) \) at \( H = 0 \) can be generated by expanding \( \tanh(nH/2kT) \) in powers of \( H/kT \). To proceed further we replace the sum over \( n \) in Eq. (6) by an integral over \( n \) from \( n = 0 \) to \( n = \infty \). This replacement will not affect the nonanalytic contribution from large \( n \). By suitable changes of variables one then obtains Eq. (1) with
\[ \rho(z) = \pi(kT)^2 Q(z)^2 \left( 1 + Q(z) \right) \]  
(11)

where \( Q(z) = e^{-\Delta xz/(2\pi kT)} \), with \( \Delta x \) given in Eq. (10). This result again shows that all derivatives of \( M(H) \) are finite at \( H = 0 \). In the Appendix we show the error in the analysis of Ref. 11 which leads to a different result.

A crude analysis of Eq. (6) can be made by recognizing that \( n \tanh(nH/2kT) \) is proportional to \( n^2 H \) for \( n H \ll kT \) and to \( n H \gg kT \). No matter how small \( H \) may be, this crossover behavior creates an anomalous variation in \( M(H) \), thus causing a singularity. At finite temperatures for sufficiently large \( n \) one will still have \( M_n(H) \approx n^2 H \) for \( n H \ll kT \) and \( n H \gg J \) providing \( T < T^0 \). Thus, for large \( n \) we set
\[ M_n(H) \sim n^2 H \left[ M_0(T)/M_0(0) \right] \]  
(12)

where \( M_0(T) \) is the spontaneous magnetization of the infinite system. Since \( M_0(T) \) is only nonzero for \( kT < J \), the condition \( n H \ll J \) is redundant in Eq. (12). Equation (12) remains valid for the Heisenberg model, so we suggest that the form of the singularity in Eqs. (2) and (11) is appropriate for both Heisenberg and Ising models for \( T < T^0 \).

III. RESULTS FOR THE BETHE LATTICE

In this section we present several analytic and numerical results for the Bethe lattice. While the Bethe lattice does have some properties uncharacteristic of three-dimensional lattices, the general trend of the results we obtain seems appropriate for three-dimensional systems.

Since \( W_n(p) \) is known exactly, the zero-temperature value of any order derivative of \( M(H) \) at \( H = 0 \) can in principle be evaluated in closed form. We have calculated \( \chi \) and \( d^2 \chi/dH^2 \) at \( H = 0 \) for the Ising (I) and Heisenberg (H) systems, using Eqs. (4) and (5), respectively. We write results as
\[ kT \chi = \frac{1}{2} \left\langle n^2 \right\rangle \]  
(13)
\[ kT \frac{d^2 \chi}{dH^2} = -\frac{1}{8} \left\langle n^4 \right\rangle \]  
(14)
\[ kT \frac{d^2 \chi}{dH^2} = -\frac{1}{160} \left[ \left\langle n^4 \right\rangle + 4 \left\langle n^2 \right\rangle + 4 \left\langle n^2 \right\rangle \right] \]  
(15)

where \( \left\langle n^r \right\rangle = \sum_n W_n(p) n^r \). Using Eq. (9) for \( W_n(p) \) one finds
\[ \left\langle n \right\rangle = p \]  
(16)
\[ \left\langle n^2 \right\rangle = p(1 + p)/(1 - \sigma p) \]  
(17)
\[ \left\langle n^2 \right\rangle = p(1 + 3p - 3\sigma p^2 - \sigma p^3)/(1 - \sigma p) \]  
(18)
\[ \left\langle n^4 \right\rangle = p(1 + p)/(1 - \sigma p) \]  
(19)
\[ \left\langle n^5 \right\rangle = p(1 + p)/(1 - \sigma p) \]  
(20)

[To obtain these results it is convenient to evaluate derivatives of \( K^2(x, y) \) given in Ref. 13.] The values of \( \chi \) and \( \chi'' \) at \( H = 0 \) are shown as a function of \( p + 1 = 6 \) in Fig. 1.

There one sees the striking divergence in \( d^2 \chi/dH^2 \) as \( p \rightarrow p_c \). In fact, from Eq. (10) one sees that \( A \sim |p_c - p|^2 \), so that \( \left\langle n^r \right\rangle /\left\langle n \right\rangle \sim |p_c - p|^2 \). We have explicitly verified that \( \left\langle n^r \right\rangle /\left\langle n \right\rangle \sim |p_c - p|^2 \) as \( p \rightarrow p_c \) for \( r \geq 2 \). Thus, succeeding even-order derivatives of \( \chi \) diverge increasingly strongly as \( p \rightarrow p_c \). Even for \( p \) fixed the zero-field derivatives for large \( r \) can be estimated to obey \( d^r \chi(dH^2)^r \) in view of the asymptotic form \( \left\langle n^r \right\rangle \sim A^r r! \) in the slope of \( M(H) \).

These results are illustrated by the numerical evaluations shown in Figs. 2 and 3. There one sees that \( -kT^3 \frac{d \chi}{dH} \) is a function of \( H \) is very much larger than \( kT \) in the region of \( M(H) \). Also, the region in which \( M \) is nearly a linear function of \( H \) is very much larger than the corresponding lattice region for \( \chi \). This effect becomes more pronounced as \( p \) approaches \( p_c \). Still higher derivatives will be larger and have
smaller linear regimes. So, if measurements were taken for \( H > H_0 \), then for some \( r \) depending on the size of \( H_0 \) one would find an apparent discontinuity in \( d^r \mu / dH^r \). Nonetheless, the true analytic behavior is that all derivatives of \( \mu(H) \) are continuous and the even-order ones vanish as \( H \to 0 \). Also, in conformity with Eqs. (13)-(16) one sees from Figs. 2 and 3 that \( \mu \) and its derivatives are noticeably smaller for the Heisenberg model than for the Ising model.

Finally, we conclude this section by giving some exact analytic results for finite temperatures. The following discussion will be confined to the paramagnetic region, i.e., for \( p \leq p_c = \sigma^{-1} \), which, for \( p < p_c = \sigma^{-1} \), includes all temperatures. In Ref. 15 we give the exact result for \( \chi(T, H=0) \) as

\[
p \sigma \tanh(J/kT) < 1,
\]

(21)

which, for \( p < p_c = \sigma^{-1} \), includes all temperatures. In the units of the present paper, we have

\[
4kT\chi(T, H=0) = p(1 + pt)/(1 - pt)
\]

(22)

in the units of the present paper, where \( t = \tanh(J/kT) \).

We now evaluate \( d^2\chi/dH^2 \) at \( H = 0 \) as a function of

\[
\frac{1}{1 + pt}
\]

FIG. 1. Zero-field and zero-temperature values of the reduced susceptibility \( \tilde{\chi} = kT\chi \), solid line, and \( \tilde{\chi}'' = - (kT)^3 d^2\chi/dH^2 \), broken line, for the Ising (I) and Heisenberg (H) models as a function of concentration for a Bethe lattice with \( \sigma + 1 = 6 \). Note the difference in scales; that for \( \tilde{\chi}'' \) for the Ising model is on the right; that for the other curves is on the left.

FIG. 2. Zero-temperature values of \( \mu(H) \), full line, and \( \mu''(H) = - (kT)^3 d^2\mu/dH^2 \), broken line, versus \( H \) for the Ising model on a Bethe lattice with \( \sigma + 1 = 6 \) for \( p = 0, 10 \) and \( p = 0, 14 \) \((p_c = 0, 2)\). The scale for \( \mu(H) \) is on the left; that for \( \mu''(H) \) is on the right.

FIG. 3. As in Fig. 2, but for the Heisenberg model.
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\[
16(kT)^3 \frac{d^3X}{dH^3} = \sum_{ijkl} \left[ (\langle \sigma_i \sigma_j \sigma_k \rangle - \langle \sigma_i \rangle \langle \sigma_j \rangle \langle \sigma_k \rangle) - (\langle \sigma_i \sigma_k \rangle - \langle \sigma_i \rangle \langle \sigma_k \rangle) \right] (v_i v_j v_k v_l)
\]

(23)

since all odd-power averages of \( \sigma \)'s vanish. We classify the terms on the right-hand side of Eq. (23) into 11 topologically distinct classes, as shown in Fig. 4. We use the result for the Bethe lattice for \( p=1 \),

\[
\langle \sigma_i \sigma_j \rangle = t^{d_{ij}}
\]

(24)

where \( d_{ij} \) is the distance between sites \( i \) and \( j \). For averages of four \( \sigma \)'s we have similar results. For terms having the topological structure of diagrams shown in Figs. 4(d)–4(g) we have, respectively,

\[
\begin{align}
\langle \sigma_i \sigma_j \sigma_k \sigma_l \rangle &= t^{d_{ijk}} \\
&= t^{d_{ijk}} t^{d_{ikt}} \\
&= t^{d_{ijkt}} \\
&= t^{d_{ijkt}}
\end{align}
\]

To evaluate Eq. (23) we combine these results for the averages at \( p=1 \) with a factor of \( p \) for each site in the diagram. The counting of diagrams then proceeds as usual for a high-temperature expansion. Since we sum all terms in the high-temperature expansion our results are valid throughout the paramagnetic region. We find that

\[
\begin{align}
-16(kT)^3 \frac{d^3X}{dH^3} &= 2p + \frac{8p^2(t^3+1)}{1-\sigma pt} + \frac{6p^3t^4}{1-\sigma pt^3} + 12p^5t^4(\sigma+1) \left[ \frac{1}{1-\sigma pt} + \frac{2t}{1-\sigma pt^3} \right] \\
&+ \frac{24p^7t^5(\sigma^2-1)}{(1-\sigma pt^3)(1-\sigma pt^5)} \left[ 11 - \sigma pt^2 + \frac{\sigma}{1-\sigma pt} - \frac{1}{2} \right] + \frac{2p^5t^5(\sigma^2-1)}{(1-\sigma pt^3)(1-\sigma pt^5)} \left[ 12\sigma - 2 \right] + \frac{6p^7t^5(\sigma^2-1)(\sigma+1)}{(1-\sigma pt)(1-\sigma pt^3)(1-\sigma pt^5)}
\end{align}
\]

(26)

These terms represent the contributions of diagrams 4(a)–4(k), respectively. Some numerical evaluations of Eqs. (22) and (26) are shown in Fig. 5. As expected, both \( kT X \) and \( (kT)^3 \frac{d^3X}{dH^3} \) are monotonic functions of both \( p \) and \( T \).

IV. CONCLUSION

We conclude that \( M(H) \) has a branch cut along the imaginary axis with exponentially small weight near \( H=0 \). All derivatives of \( M(H) \) are finite at \( H=0 \). Numerically, the higher-order derivatives become large, particularly near \( p=p_c \), so that an experimental determination of the exact nature of the singularity at \( H=0 \) is probably impossible.

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APPENDIX: ANALYSIS OF REF. 11

In Ref. 11 Domb writes a set of equations, Eq. (1) and (4), equivalent to

\[
kT \frac{dM}{dH} = \sum_n W_n(p)n^2 \text{sech}^2 nH/kT,
\]

(A1)
which he approximates as

$$kT \frac{\partial M}{\partial H} = \sum_n W_n(\theta) n^2 e^{2n\theta_1/kT}. \quad (A2)$$

We claim that this approximation is inappropriate for analyzing the singularity for $H \to 0$. To see this, differentiate Eq. (A2):

$$-(kT)^2 \frac{\partial^2 M}{\partial H^2} = \theta(H) \sum_n W_n(\theta) n^3 e^{2n\theta_1/kT}, \quad (A3)$$

where $\theta(H) = H/|H|$. According to Eq. (A3) $\partial^2 M/\partial H^2$ has a discontinuity for $H \to 0$ given by

$$\frac{\partial^2 M}{\partial H^2} \bigg|_{H \to 0} = \frac{1}{(kT)^2} \sum_n W_n(\theta)n^3. \quad (A4)$$

On general grounds we know that the contribution to the left-hand side of Eq. (A4) from finite-sized clusters is zero. However, the right-hand side of Eq. (A4) incorrectly has nonvanishing contributions from finite-sized clusters. Thus we conclude that Eq. (A2) does not correctly represent the low-field singularity contained in Eq. (A1).

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Footnotes:

*This work supported by the U. S. Atomic Energy Commission.
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©In the range of temperature and concentration for which the spontaneous magnetization $M_s(T)$ is nonzero, i.e., for $T < T_c(\phi)$, there will of course be a singular contribution from the infinite cluster of the same form as for the pure system, viz., $M(H, T) \sim H/|H| M_c(T)$ for $H \to 0$. For an Ising system for $T = T_c(\phi)$, $\chi(H)$ is probably finite as $H \to 0$, and one can separate the singular contributions of the infinite cluster from those of the finite clusters considered in the text. In contrast, for a Heisenberg model $\chi(H)$ diverges as $H \to 0$, so such a separation may be impossible.
∥C. Domb, J. Phys. C 7, 2677 (1974). In the note added in proof Domb cites an observation by Fisher that the approximation Domb used is inadequate for a discussion of the Griffiths singularity and that a more careful treatment indicates no discontinuity but rather an essential singularity in $dM/dH$. Technically speaking, the singularity we find in Eqs. (2) and (11) is not an essential singularity inasmuch as it is not an isolated one. In a very recent paper M. Wortis obtained conclusions similar to those obtained here. See M. Wortis, Phys. Rev. B 10, 4665 (1974).
‡Note that since $\tanh(H/2kT)$ has a pole at $H = \pm kT/n$, we may estimate that for $H = 0$, $d/dH \chi^{(\pm)} \sim (2r+1)! (n/|kT|)^{2r+1}$ for large $r$. Thus $d/dH \chi^{(\pm)} \sim (2r+1)! (n/|kT|)^{2r+1}$. For $r \to 1$, $H = 0$.