Critical Behavior of Griffiths Ferromagnets

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From a heuristic calculation of the leading order essential singularity in the distribution of Yang-Lee zeroes, we obtain new scaling relations near the ferromagnetic-Griffiths transition, including the prediction of a discontinuity on the analogue of the critical isotherm. We show that experimental data for the magnetization and heat capacity of $La_{0.7}Ca_{0.3}MnO_3$ are consistent with these predictions, thus supporting its identification as a Griffiths ferromagnet.

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The influence of disorder on ferromagnets remains, after more than 30 years of effort, a complex and poorly-understood phenomenon. In its simplest form, disorder can be represented as a random spatial variation of the exchange interaction J in the bonds between neighboring sites on a regular lattice. If a great enough fraction $p > p_c$ of the bonds have $J = 0$, then one would expect that there is a vanishingly small probability of finding a percolating pathway of bonds throughout the system, and the cooperative ferromagnetic phase would cease to exist. For smaller values of p, we would expect that the ferromagnetic phase will exist in a form weakened by the shortage of percolating paths; hence, thermal fluctuations will destroy the ferromagnetic phase at a temperature T_c which is *lower* than the critical temperature T_G of the pure ferromagnet. However, as Griffiths showed , it is not the case that the phase for $T_c < T < T_G$ is purely paramagnetic, because in the thermodynamic limit, there can exist arbitrarily large volumes of the system that are devoid of disorder, with a probability exponentially sensitive to the volume. As a result, the free energy is nonanalytic in external field, h, throughout the whole Griffiths phase. The effect of disorder is to partition the pure system into small ferromagnetic clusters. Depending on the size, each cluster has a different value of T_c, so that the system as a whole exhibits a spectrum of T_c, spanning from the critical temperature of the pure system, T_G, due to arbitrarily large clusters, to some value of T_c, contributed by smaller clusters.

Here we are concerned with the phase transition between the ferromagnetic and Griffiths phases. Just as in the case of a pure ferromagnet, one would like to predict the critical phenomena, but the nonanalytic nature of the Griffiths phase makes it difficult to apply off-the-shelf renormalization group techniques [2–4] or to posit simple scaling laws, despite recent theoretical progress [5–10]. Indeed, it is currently controversial whether or not there is clear experimental evidence [11–14] supporting the existence of the Griffiths phase. From the practical perspective, perhaps the most unsatisfactory aspect of efforts to relate theory to experiment is that critical exponents derived from conventional scaling laws are unrealistically large:, for example, the critical isotherm exponent was recently [15] estimated as $\delta = 17$. The breakdown of conventional scaling strongly suggests that the functional form of the scaling relations actually reflects the essential singularities intrinsic to the Griffiths phase, and that some form of exponential scaling, rather than algebraic scaling, is appropriate.

The purpose of this Letter is to address these problems by exploring the expected form of scaling relations that would follow from a leading essential singularity contribution to the statistical mechanics in the Griffiths phase. Such a contribution can be conveniently represented using the Yang-Lee theory of phase transitions [16] to derive the scaling behavior in the Griffiths phase from a simple, physically-motivated ansatz for the distribution of partition function zeros, following arguments originally due to Bray and Huifang [5] for the case of long-ranged ferromagnets. We demonstrate that the leading singularities in the thermodynamics can be deduced, and that our predictions consistently describe high quality magnetic and thermodynamic data [11,15] on the disordered Heisenberg ferromagnet $La_{0.7}Ca_{0.3}MnO_3$. We emphasize that our purpose is only to identify the leading essential singularities, and that it is beyond the scope of our work to provide a full description valid outside of the asymptotic critical regime. Nevertheless, the experimental data have sufficient resolution that we have strong support for our scaling predictions in this asymptotic regime.

Magnetic properties are well accounted for by our approach, but heat capacity data are not expected to follow a simple scaling form—and indeed do not—due to the complex form of the theoretical predictions which arise from even the simplest Yang-Lee zero distribution function that we use. The nonanalyticity of magnetization in external field—a signature of the Griffiths phase—is also explicitly demonstrated. Our results provide strong evidence for a Griffiths singularity and highlight the need for a more systematic renormalization group approach to understanding the singularities in such disordered systems.

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Yang-Lee zeroes and critical phenomena.—In 1952, Lee and Yang [16] developed a theory of phase transitions based upon the density g of zeroes of the grand partition function as a function of the complex fugacity and showed that the zeroes lie on a unit circle in the complex plane, parameterized below in terms of the angle θ. The distribution $g(\theta)$ varies with temperature T and dictates the functional form of thermodynamics. Near a critical point, it is expected that $g(\theta, T)$ exhibits behavior which reflects the nonanalyticity of the thermodynamics [17,18], and we provide this connection explicitly here, for both the case of conventional ferromagnetic critical point scaling, and then for the scaling near the Griffiths point.

We begin with the scaling of the magnetization per spin, $M(H, t)$, where H is the external magnetic field, and $t \equiv (T - T_c)/T_c$ is the reduced temperature for a regular Ising ferromagnet. The exact relationship between $M(H, t)$ and $g(\theta, t)$ can be written as [16]

$$M(H, t) = 2\mu \int_0^{\pi} d\theta \frac{g(\theta, t)\tanh(\mu H/k_B T)[1 + \cot^2(\theta/2)]}{1 + [\tanh(\mu H/k_B T)\cot(\theta/2)]^2}, \tag{1}$$

where μ is the magnetic moment of individual spins. To extract the scaling behavior, we proceed by expanding around the critical point $H = 0$ and $t = 0$. Because the singular behavior arises from the limit $\theta \to 0$, we also expand the integrand about $\theta = 0$, resulting in

$$M(H, t) = 2\mu \frac{\mu H}{k_B T} \int_0^{\pi} d\theta \frac{g(\theta, t)}{(\mu H/k_B T)^2 + \theta^2/4}, \tag{2}$$

which is valid near the critical point, up to corrections reflecting a smooth background. A change of variables, $\theta = \mu H \phi / k_B T$, gives

$$M(H, t) = 4\mu \int_0^{\pi k_B T/2\mu H} d\phi \frac{g(2\mu H\phi/k_B T, t)}{1 + \phi^2} \tag{3}$$

where the upper limit is replaced by ∞ as $H \to 0$. This is the primary relation between $M(H, t)$ and $g(\theta, t)$ near the critical point. The scaling form of the magnetization,

$$M(H, t) = |t|^{\beta} f_M(H/|t|^{\beta\delta}), \tag{4}$$

where $f_M(x)$ is an unknown scaling function and β and δ are two critical exponents, implies a scaling form for $g(\theta, t)$. Substituting Eqn. (4) into Eqn. (3), Mellin transforming the expression and using the corresponding convolution relation, we arrive at the scaling form of $g(\theta, t)$ as $\theta \to 0$

$$g(\theta, t) = |t|^{\beta} G(\theta/|t|^{\beta\delta}), \tag{5}$$

where $G(x)$ is a scaling function for $g(\theta, t)$.

Apart from exhibiting the scaling form of $g(\theta, t)$ for normal ferromagnets, this exercise also shows that knowledge of $g(\theta, t)$ can be gained by studying the scaling form of $M(H, t)$, and vice versa. In the following section, we study the scaling behavior of $M(H, t)$ in the Griffiths phase using a heuristically-derived form for $g(\theta)$. We will see that the result is different from that of the pure case, reflecting the intrinsic essential singularity that characterizes Griffiths phases.

Density of zeros for a disordered ferromagnet.— We start with the scaling form of $g(\theta, t)$ derived on the basis of heuristic arguments by Bray and Huifang [5] for disordered ferromagnets with short-ranged interactions:

$$g(\theta, t) = \frac{1}{\pi} \mathrm{Re} \sum_{r=1}^{\infty} \exp[-A(t)r] \tanh[r(i\theta + \epsilon)] \tag{6}$$

where $\epsilon \to 0^+$ and $A(T) \sim (T - T_c)^{2-\beta_r}$ as $T \to T_c$ and $A(T) \to \infty$ as $T \to T_G$. The exponent β_r is the order parameter exponent for the random case, and its value will reflect the universality class of the magnet, be it Ising, Heisenberg, O(n), etc. In the limit that $\theta \to 0$ and $A \to 0$, the summand is dominated by peaks at values of $r = (2n + 1)\pi/2\theta$ for all non-negative values of n, whose height is given by $[2\theta/\epsilon(2n + 1)\pi]\exp[-(2n + 1)\pi A/2\theta]$. The width of these peaks therefore scales in the same way as their separation, both being proportional to $1/\theta$. Thus, the expression takes the form

$$g(\theta, t) = g_0 \exp(-A(t)/|\theta|), \tag{7}$$

where g_0 is a constant. The essential singularity in Eqn. (7) reflects the Griffiths phase of disordered ferromagnets, not present in the pure case. Disordered magnets with long-range interactions have a power-law prefactor to the essential singularity [5], but this is not present in the short-range case. A disordered ferromagnet can be thought of as an ensemble of weakly interacting, finite-sized ferromagnetic clusters. When $T \sim T_G^-$, only large clusters contribute to the overall magnetization. For each large cluster of linear size L, the smallest Yang-Lee zero is of the order of $\theta \sim 1/mL^d$, where $m \sim (T_G - T)^{\beta}$ is the magnetization per spin of the cluster and d is the spatial dimension of the system. The probability of a spin belonging to a cluster of size L follows the Poisson distribution, i.e. prob $\sim \exp(-cL^d)$. As a result, large clusters contribute to $g(\theta, t)$ in the form of Eqn. (7), where $A(t) \sim (T_G - T)^{-\beta}$ and β is the usual exponent for pure ferromagnets.

For $T < T_c$, the system is in its ferromagnetic phase with nonzero magnetization, implying a nonzero value of $g(\theta = 0, t)$ [16]. This requires $A(t) = 0$ at $T = T_c$ to counteract the essential singularity at $\theta = 0$. For $T \sim T_c^+$, we can expand $A(t)$ and approximate it by $A(t) \sim t^{2-\beta_r}$. This accounts for the asymptotic behavior of $A(t)$.

With this form of $g(\theta, t)$, the scaling behavior of $M(h, t)$ can be obtained by substituting Eqn. (7) into Eqn. (2) and making a change of variables, $y = A(t)/\theta$, yielding

$$\frac{M(h, t)}{\mu} = \frac{g_0 A(t)}{2h} \int_{A(t)/\pi}^{\infty} dy \frac{\exp(-y)}{y^2 + [A(t)/2h]^2}, \tag{8}$$

where we defined $h \equiv \mu H/k_B T_c$. Eqn. (8) can be written in terms of the exponential integral, $E_1(x) \equiv \int_x^{\infty} e^{-t}/t dt$, as

$$\frac{M(h,t)}{\mu} = -g_0 \,\mathrm{Im}\{\exp[iA(t)/2h]E_1[A(t)/\pi + iA(t)/2h]\}. \tag{9}$$

We expand Eqn. (9) about $A = 0$, because $A(t) = A_0 t^{2-\beta_r}$ is asymptotically small in the critical region, resulting in

$$\frac{M(h,t)}{\mu} = -g_0 e^{-A(t)/\pi}\,\mathrm{Im}[e^{iA(t)/2h}E_1(iA(t)/2h)]. \tag{10}$$

This implies an approximate scaling form

$$\frac{M(h,t)}{\mu} = \exp[-A(t)/\pi][f_M(A(t)/h) + O(h)], \tag{11}$$

where

$$f_M(x) = -g_0 \,\mathrm{Im}[\exp(ix/2)E_1(ix/2)], \tag{12}$$

and the corrections of $O(h)$ involve exponential integrals of A/h. This scaling prediction is valid in the limits $A \to 0$, $h \to 0$, but the ratio A/h has not been fixed by the analysis so far.

Analysis of magnetization data.— We now analyze the experimental data on $La_{0.7}Ca_{0.3}MnO_3$ [11,15], using the above results, to see if the data are consistent with the presence of a Griffiths phase. Figure 1 shows the predicted data collapse of the magnetization with fitted values $T_c = 218$ K, $g_0 = 0.5 \pm 0.05$, $A_0 = 0.5 \pm 0.05$, and $\beta_r = 0.8 \pm 0.05$. The error bars were obtained by estimating the best fit visually. The figure also shows the agreement between the theoretically-predicted universal scaling function and the collapsed data. Overall, the data scale quite well, and the scaling function of the collapsed data is close to that of the theory, except near the turning point of the curve, where corrections to the leading order ansatz we have used for Eqn. (11) become important, and the data are no longer in the asymptotic limits $A \to 0$ and $h \to 0$. In order to show this clearly, we calculate the asymptotics of the scaling function, in the limits $A/h \to 0$ and $A/h \to \infty$, where the leading terms in the magnetization can be calculated systematically.

Asymptotic behavior.— From Eqn. (9), the asymptotic behavior of $M(h,t)$, in the limit $A/h \to \infty$, is given by

$$\frac{M(h,t)}{\mu g_0} \mathrm{sim} \frac{2h}{A} e^{-A/\pi}\left[1 - \frac{4h^2}{\pi^2} - \frac{8h^2}{\pi A} + O\left(\frac{h^2}{A^2}\right)\right]. \tag{13}$$

showing that the small field susceptibility, M/h, in the limit $A/h \to \infty$, depends linearly on $\exp(-A/\pi)/A$. This prediction recovers Curie-Weiss-like behavior, as verified by the experimental data shown in Fig. 2. The experimental data show a slope of 0.93, consistent with the fitted range of values for $0.9 < 2g_0 < 1.1$.

A profound difference between conventional and Griffiths ferromagnets is found in the limit $A/h \to 0$. It is well understood, for conventional ferromagnets, that $M(h,t) \sim h^{1/\delta} \to 0$ as $h \to 0$; this is, however, not true in Griffiths ferromagnets, as we can see by calculating the asymptotic behavior of $M(h,t)$

$$\frac{M(h,t)}{\mu g_0} \sim \frac{\pi}{2} + \left[\ln\left(\frac{A}{2h}\right) + \gamma - 1\right]\frac{A}{2h} - \frac{2h}{\pi} + \frac{Ah}{\pi^2} + O\left(\frac{A^2}{h^2}\right) \tag{14}$$

as $A/h \to 0$, where $\gamma \sim 0.57722\ldots$ is the Euler-

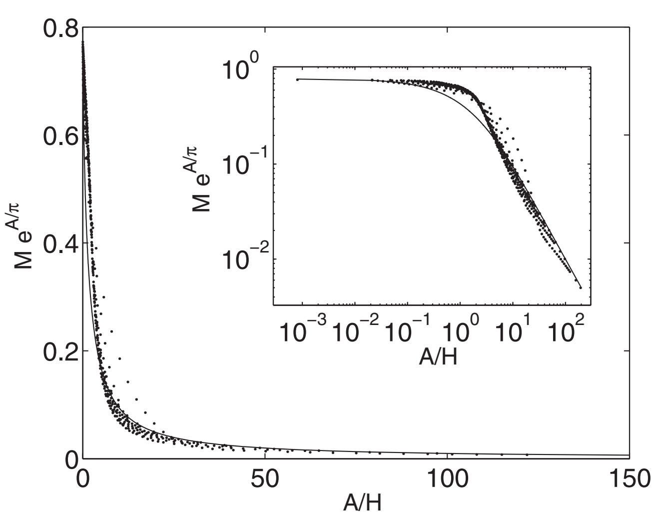

FIG. 1. Data collapse of magnetization, $M(h,t)$. Dots are experimental data and the line is the theoretical prediction for the universal scaling function, with $g_0 = 0.5$, $A_0 = 0.5$, and $\beta_r = 0.8$. Data shown are in the range of $t < 0.35$ and $h \equiv \mu H/k_B T_c < 0.39$. The insert shows the data on logarithmic scales.

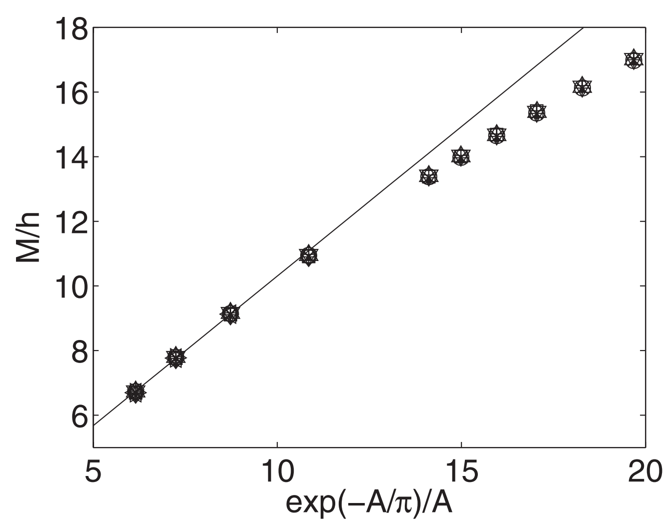

FIG. 2. Experimental verification of the asymptotic behavior of the magnetization for $A/h \to \infty$. The data show a linear dependence of M/h on $\exp(-A/\pi)/A$ with slope 0.93 as predicted by Eqn. (13). The range of values of A plotted is $0.05 < A < 0.155$. Legend: $\cdot\, h = 0.0006$, $\times\ h = 0.0011$, $\bigcirc\ h = 0.0017$, $\triangle\ h = 0.0023$, $\nabla\ h = 0.0028$, $+\ h = 0.0034$, $*\ h = 0.0045$, $\square\ h = 0.0057$, $\diamond\ h = 0.0072$, $\triangleleft\ h = 0.0090$, $\triangleright\ h = 0.0122$, $\star h = 0.0141$, and the solid line shows the linear prediction of Eqn. (13).

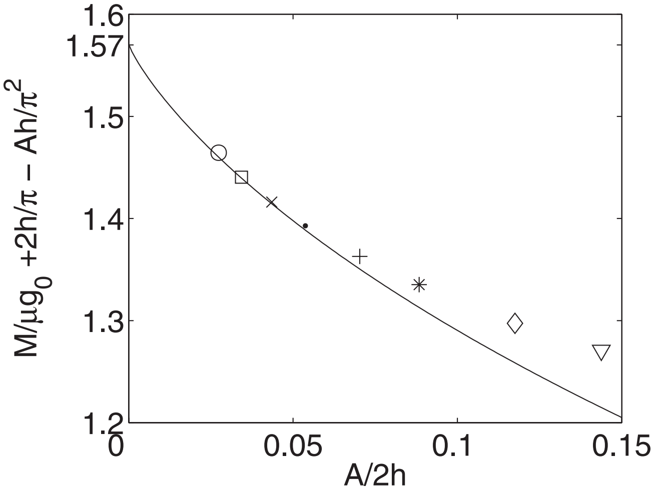

FIG. 3. Asymptotic behavior of the magnetization for $A/h \to 0$, showing that the data follow Eq. (14) and exhibit a discontinuity $M \to \mu g_0 \pi/2$ in the limit $h \to 0$. The data shown here are within the limit $A/h < 0.3$ and $h < 0.0072$. Legend: ▽ $h = 0.0028$, ◇ $h = 0.0034$, * $h = 0.0045$, + $h = 0.0057$, · $h = 0.0072$, × $h = 0.0090$, □ $h = 0.0112$, ○ $h = 0.0141$. Solid line: theoretical prediction of Eq. (14).

Mascheroni constant. This implies that $M(h, t) \to \mu g_0 \pi/2$ as h, $A/h \to 0$, i.e. a discontinuity of $M(h, t)$ at $h = 0$ on the critical isotherm. Experimental support for this surprising result is present in Fig. 3 of Ref. [15], which documents the increase of the exponent δ fitted to data assuming the conventional scaling applies. As the transition temperature is lowered by increasing disorder, the inferred value of δ rises to as much as 16.9, inconsistent with any known universality class, but indicative of a very rapid and dramatic rise in magnetization. In order to test the precise predictions made here, we show in Fig. 3 the experimental verification of Eqn. (14), where the function $M/\mu g_0 + 2h/\pi - Ah/\pi^2$ is plotted against $A/2h$. The data points satisfying the criteria $h < 0.0072$ and $A/h < 0.3$ are shown in Fig. 3. All the parameters were determined previously according to the data collapse in Fig. 1, so that we have not made any additional fitting. The experimental data approach the theoretical curve as $A/h \to 0$, and moreover tend to the universal number $\pi/2$ as dictated by Eqn. (14). We conclude that the data are consistent with the prediction that $M(h, t)$ is discontinuous at $h = 0$ in the limit $A/h \ll 1$, a prediction which follows from the essential singularity characterizing the Griffiths transition [1].

Heat Capacity.— We conclude with a brief discussion of the heat capacity, $C(h, t)$. We can integrate Eqn. (11) to obtain the free energy, $F(h, t)$ and then an expression for $C(h, t)$. There is no prediction of data collapse, due to the interference from the exponential terms, in agreement with our failure to obtain data collapse from the data. This form contains singular terms of the form

$$\int d\theta e^{-A/\theta} \ln(4h^2 + \theta^2) \sim \exp(-A/h), \qquad (15)$$

where the integral is restricted to the neighborhood of the origin where Eqn. (7) is valid, and the upper limit is assumed to scale with h, leading to the estimate of the essential singularity. Similar terms have also been predicted in Ref. [10], but more than the leading term must be retained in order to consistently compute the magnetization.

In conclusion, we have argued that the essential singularity of the Griffiths phase leads to novel features in the critical behavior, including a discontinuity of magnetization in external field. These features are reproduced to the accuracy expected from our lowest order theoretical predictions by high quality experimental data from $La_{0.7}Ca_{0.3}MnO_3$, and lead to a consistent description of its critical behavior, supporting the identification of this material as a Griffiths ferromagnet.

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