PHYSICAL REVIEW B, VOLUME 64, 214420

Testing renormalization group theory at the critical dimension in $LiHoF_4$

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(Received 2 July 2001; published 13 November 2001)

We have performed high-precision specific heat measurements on the Ising dipolar magnet $LiHoF_4$ in the critical regime (reduced temperature $|t| \lesssim 0.02$). Combining these results with existing magnetization M and susceptibility χ data, we test renormalization group predictions at the critical dimension. In particular, the nontrivial prediction that $t^2 \chi C_P T_C / M^2 = \frac{1}{3}$ is well verified.

DOI: 10.1103/PhysRevB.64.214420

PACS number(s): 75.40.Cx

I. INTRODUCTION

Since the ground-breaking theoretical work of Larkin and Khmel'nitskii,[1] it has been recognized that dipolar-coupled Ising magnets, with the physically realizable critical dimensionality $d^* = 3$, constitute a powerful testing ground for the theory of critical behavior and particularly of renormalization group theory (RGT). In particular, while systems above or below d^* should manifest power-law critical behavior, a variety of logarithmic corrections are predicted for magnets at d^*. Very close to T_C, magnetization, specific heat, susceptibility, and other quantities are predicted to vary as $t^a \log^{1/3}(t)$ [$t \equiv$ the reduced temperature, $(T - T_C)/T_C$], where a is the standard mean-field exponent. Among the first supporting evidence were the beautiful specific heat (C_P) data of Ahlers, Kornblit, and Guggenheim[2] on the dipolar pseudo-Ising system $LiTbF_4$. (Specific heat displays a particularly clear signature of logarithmic corrections since the mean-field exponent $\alpha = 0$.) Other work on the critical behavior of the magnetization[3] M and susceptibility[4] χ on this and similar systems followed, all of which were consistent with the predictions of RGT. However, the degree of incisiveness with which these results uniquely identify the theoretical analysis as the correct description of the results varies widely.

Along with straightforward predictions of logarithmic corrections, RGT also predict several *universal relations* between various physical quantities. For example, it is predicted that[5]

$$\xi^2 \xi_{||} C_P t^2 / k_B = \frac{3}{32\pi} |\ln(t)|. \quad (1)$$

The confirmation[6] in $LiTbF_4$ of this relationship between the longitudinal and transverse correlation lengths $\xi_{||}$ and ξ and the specific heat was strong evidence for the existence and magnitude of anomalous corrections to mean-field behavior at d^*. Another important universal formula[5] for the specific heat, magnetization, and susceptibility for $T < T_C$ is given by

$$R \equiv t^2 C_P \chi T_C / M^2 = \frac{1}{3} \quad (t < 0). \quad (2)$$

Note that, like Eq. (1), this equation implies more than the prediction that all three quantities contain a term that $\sim \log^{1/3}(t)$. Rather, it is a precise statement about the magnitudes of the respective quantities in the critical regime. Involving as it does three quantities generally measured in separate experiments, Eq. (2) has only been roughly tested to date. We will show, using three independent data sets, that this universal prediction is well realized in a true Ising dipolar-coupled magnet.

II. MATERIAL AND EXPERIMENTAL DETAILS

The nature of ferromagnetism in $LiHoF_4$ has been the subject of substantial study.[7] The lowest ($^5I_8$) spin-orbit manifold of the Ho^{3+} ions is split by crystal fields into a ground-state doublet, a singlet approximately 9.4 K higher in energy,[8] and 15 higher energy states. At the ferromagnetic Curie temperature $T_C \approx 1.54$ K, only the ground state is appreciably occupied. Each member of the doublet is itself split by strong hyperfine interactions ($I = 7/2$) into eight levels spaced by 205 mK.[9] The system is truly Ising like:[10] the doublet g factor along the tetragonal c axis is $g_{||} \approx 14$ while $g_\perp = 0$. This is in contrast to the pseudodoublet found in $LiTbF_4$, making the Ho system more attractive from the standpoint of testing theories of Ising critical behavior at d^*. The spins are coupled primarily by dipole-dipole interactions, with the nearest-neighbor superexchange interaction[11] contributing an antiferromagnetic coupling $J \approx -0.34$ K. As discussed in Ref. 12, exchange interactions in three dimensions influence critical behavior when[14]

$$t^\phi \gtrsim \frac{(g\mu_B)^2}{Ja^3} \quad (\phi = 7/6). \quad (3)$$

Here a is a characteristic lattice parameter. Conservatively using[11] the c-axis lattice parameter of 10.75 Å and $|J| = 0.34$ K, we find that exchange coupling dominates when $t \gtrsim 0.34$. This is a factor of 30 greater than the range of reduced temperature used in the present analysis, implying that we may consider $LiHoF_4$ to be a strictly dipole-coupled Ising system for the purpose of this paper.

We have measured the specific heat of a 0.1295g sample[15] of $LiHoF_4$ in the region of the critical point using a semiadiabatic technique. The sample and single-crystal quartz substrate (containing a RuO_2 thermometer and film heater) were thermally equilibrated with a ^{3}He refrigerator via a mechanical heat switch at various temperatures. At each point the substrate thermometer was calibrated against a Ge resistor. The switch was then opened at a relatively low temperature

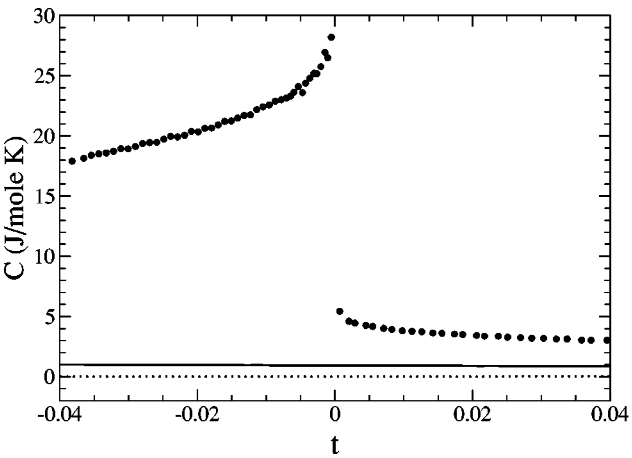

FIG. 1. Specific heat data for $LiHoF_4$ vs reduced temperature. The solid line is the hyperfine contribution, and the dashed line indicates the phonon contribution.

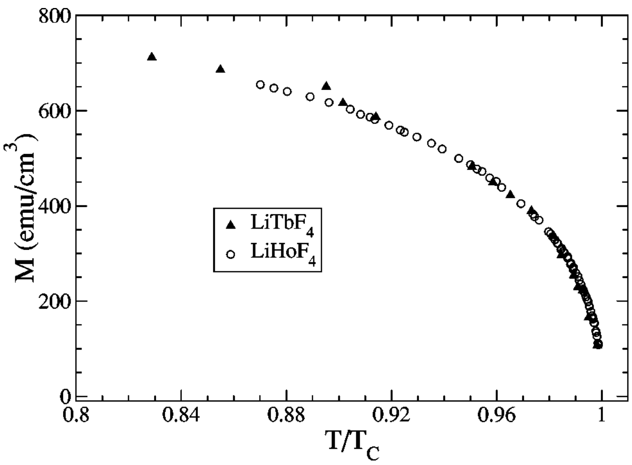

FIG. 2. Magnetization of $LiTbF_4$ and $LiHoF_4$ vs temperature, normalized to $LiHoF_4$'s saturation magnetization. Data from Refs. 6 and 3, respectively.

and a series of small heat pulses was introduced, raising the sample temperature an amount proportional to its specific heat at each substrate temperature. The results, over a fairly wide range of reduced temperature, are shown in Fig. 1. We find a critical temperature $T_C = 1.5384$ K, consistent with previous results (e.g., 1.5383 K in Ref. 3).

III. ANALYSIS AND RESULTS

We used the magnetization data of Griffin, Huster, and Folweiler,[3] measured using an elastic light scattering technique. Unfortunately, their experiment did not reach sufficiently low temperatures to saturate the magnetization. Furthermore, the data are given in arbitrary units, while Eq. (2) perforce refers to absolute units. Taking advantage of the fact that the saturation magnetization of $LiHoF_4$ is known[10] accurately, we therefore applied the following procedure to find the multiplicative factor needed to convert the data of Ref. 3 into physical units.

(a) We normalized the absolute temperature-dependent magnetization data[6] $M(T)$ from neutron scattering measurements of $LiTbF_4$, so that the saturation magnetization was equal to that of $LiHoF_4$ (892 ± 5 emu/g).[10,16]

(b) We assume that the shapes of the Tb and Ho compound $M(T)$ functions are similar, at least at high temperatures close to T_C where the splitting of the Tb system's ground state is comparatively unimportant.

(c) We scaled the data of Ref. 3 to lie on top of the rescaled $LiTbF_4$ results as a function of T/T_C. This gives the desired factor.

The results of this procedure are shown in Fig. 2.

The scaling is excellent near T_C. However, the Ho and Tb data deviate slightly at lower T. Whether this is due to the aforementioned splitting, shortcomings of the normalization procedure or other intrinsic differences in the two systems is unknown.[17] This uncertainty could be remedied by careful measurements of the absolute magnetization of $LiHoF_4$ at one or more temperatures in the range of Ref. 3 and at sufficiently low T to saturate the magnetic moment.

We have used the susceptibility data of Beauvillain *et al.*[4] One manipulation required in this case was to apply a demagnetization correction. Calling the measured susceptibility in the ferromagnetic state χ_{max}, we compute

$$\chi(t) = [\chi_{raw}^{-1}(t) - \chi_{max}^{-1}]^{-1}. \tag{4}$$

The value used for χ_{max} is from an RGT fit.[4] The data go sufficiently low in temperature, however, that this quantity may be read off directly without recourse to a fit. Changes in χ_{max} within the scatter of the $\chi(T<T_C)$ data have negligible effect on our analysis. The result is shown in Fig. 3. Notably, the relation that is being tested, Eq. (2), only holds *below* T_C while the susceptibility data are only meaningful *above* the transition. To extract values for $t<0$ we will use a basic result of the RGT critical behavior analysis,[1] namely, the "law of two": $\chi(T<T_C) = 0.5\chi(T>T_C)$.

To utilize the specific heat data of Fig. 1, we first subtracted a hyperfine contribution calculated using basic statistical mechanics and the hyperfine coupling constant of Ref. 13. (The raw data used in the analysis are contained in Table I.) This contribution, shown in Fig. 1 as a solid line, is fairly

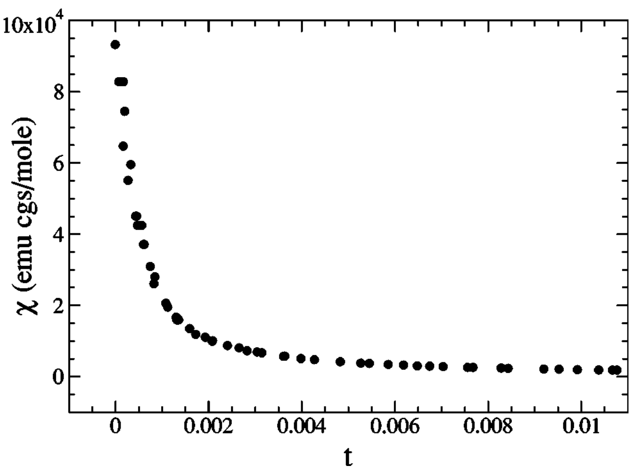

FIG. 3. Demagnetization-corrected magnetic susceptibility data for $LiHoF_4$ vs reduced temperature from Beauvillain *et al.* (Ref. 4).

TABLE I. Raw specific heat data (no subtractions) in $J/(\mathrm{mol\,K})$ as a function of reduced temperature with $T_C = 1.5384$ K.

t	$C_P(t)$	t	$C_P(t)$
-9.642×10^{-3}	22.48	5.295×10^{-4}	4.763
-9.185×10^{-3}	22.26	9.440×10^{-4}	4.796
-8.729×10^{-3}	22.58	1.367×10^{-3}	4.540
-8.281×10^{-3}	23.13	1.812×10^{-3}	4.290
-7.839×10^{-3}	23.23	2.273×10^{-3}	4.203
-7.399×10^{-3}	23.33	2.741×10^{-3}	4.145
-6.959×10^{-3}	23.26	3.223×10^{-3}	3.951
-6.521×10^{-3}	23.40	3.714×10^{-3}	3.989
-6.084×10^{-3}	23.54	4.202×10^{-3}	4.014
-5.650×10^{-3}	23.64	4.703×10^{-3}	3.783
-5.218×10^{-3}	23.76	5.210×10^{-3}	3.802
-4.792×10^{-3}	24.33	5.729×10^{-3}	3.715
-4.370×10^{-3}	24.30	6.252×10^{-3}	3.649
-3.950×10^{-3}	24.60	6.777×10^{-3}	3.699
-3.533×10^{-3}	24.56	7.308×10^{-3}	3.562
-3.120×10^{-3}	25.19	7.839×10^{-3}	3.727
-2.716×10^{-3}	25.55	8.370×10^{-3}	3.583
-2.317×10^{-3}	25.99	8.914×10^{-3}	3.513
-1.922×10^{-3}	26.01	9.464×10^{-3}	3.541
-1.533×10^{-3}	26.78		
-1.149×10^{-3}	26.80		
-7.707×10^{-4}	27.57		

small and almost constant over the critical temperature interval. Thus, this term does not play a major role in testing RG theory by direct fits of logarithmic corrections predicted by RGT to the $C_P(T)$ data. However, it is essential to properly account for it when testing the more constraining condition implied by Eq. (2). Similarly, the phonon contribution βT^3 should also be subtracted. The result (using a literature value[11] for β) is shown in Fig. 1 as a dashed line. In this case, the contribution is so small that it is neglected.

Since Eq. (2) pertains to temperatures below T_C (i.e., $t<0$), we must also subtract the mean-field jump experienced by the specific heat at T_C. Furthermore, the universal condition applies to what might be called the *fully critical* regime, i.e., where $|t|$ is sufficiently small that $C_P \propto \log^{1/3}(t) + \mathrm{const}$. To our knowledge, no specific heat experiment to date has accessed this regime (indeed, rounding of the transition due to sample defects may make it impossible to do so). To extract this behavior from our results, we have fit the data for $0.0005 \leqslant |t| \leqslant 0.01$ to the form predicted by Ref. 1,

$$C_P(t) = C_1\left[\begin{pmatrix}1\\4\end{pmatrix}[1 + C_2 \ln(C_3/|t|)]^{1/3}\right]. \tag{5}$$

The upper values in the parentheses apply when $t>0$ while the lower ones apply when $t<0$. This form (which is identical to that used by Ahlers, Kornblit, and Guggenheim[2]) is expected to interpolate between relatively large t and the asymptotically critical region of small t. The result of a simultaneous fit of Eq. (5) above and below T_C is shown in Fig. 4 with the fit parameters C_i found to be $C_1 = 3.818\,03$ J/mol K, $C_2 = 1.380\,91$, and $C_3 = 0.129\,688$. Note that, as required by RGT, the values of the C_i are constrained to be the same above and below the Curie point. From Eq. (5), very near T_C, $C_P = 4C_1 \log^{1/3}(C_3/t)$. It is this asymptotic function, solely composed of lowest-order corrections to mean-field behavior, which will be used in testing the universal relation, Eq. (2). While one could do something similar for the succeptibility and magnetization, we choose not to. This will be discussed below.

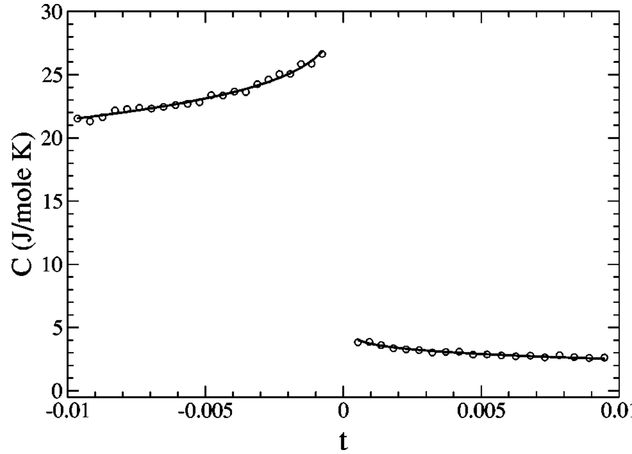

FIG. 4. Fit of the Larkin-Khmel'nitskii (Ref. 1) theory of Ising dipolar critical behavior to the C_p data near T_C.

We are now in a position to test the RG prediction. The calculated values of R are shown in Fig. 5 as a function of reduced temperature. The temperatures used are those from the $M(T)$ data, and we have fit the χ data to a high-order polynomial to interpolate to these points. The precise polynomial order used was found to be unimportant in the analysis. The scatter in the points predominantly reflects that in the $M(T)$ data, since the specific heat and susceptibility components are represented by the aforementioned smooth RGT (to extract asymptotic behavior) and polynomial (interpolation) fits. The value of T_C used in Eq. (2) was that from the specific heat, $T_C = 1.5384$ K. The data in Fig. 5 are quite close to the predicted value of 1/3: the average of R over the

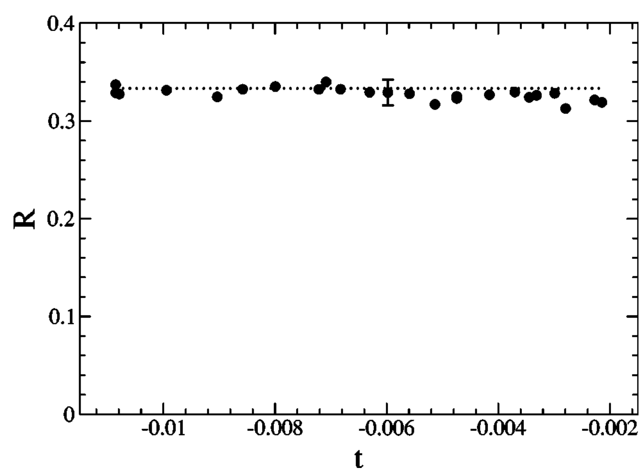

FIG. 5. The quantity $R \equiv t^2 C_P \chi T_C / M^2$. The dashed line is the RGT predicted value of 1/3.

temperature range shown is $\overline{R}=0.327$, with a standard deviation of 1.9%.[18] While gratifying, we conservatively estimate systematic uncertainties [mainly connected with the normalization of $M(T)$] of $\pm 4\%$. Since we have used the χ and M data ''as is,'' i.e., using the data themselves rather than RGT fits, one can also see that R is essentially constant out to a reduced temperature of -0.01, implying that the magnetization and susceptibility data are essentially ''asymptotic'' out to this temperature. This is consistent with the relatively successful fits[3,4] to $\log^{1/3}(t)$ terms in the original works. The same may not be said of the specific heat, where the ''interpolative'' form Eq. (5) is definitely required to fit the data. The reason for this difference is not known, though it may be related to the fact that the logarithmic term is the leading behavior for specific heat while it is multiplied by the appropriate mean-field power of t for M and χ (i.e., $\chi \sim t^{-1}$ and $M \sim t^{0.5}$, respectively). It would be very interesting to extend Fig. 5 to lower temperatures to observe deviations from the asymptotic value of 1/3. The present analysis is limited by range over which published susceptibility data are available.

In conclusion, we have tested a universal relation predicted by renormalization group theory at the critical dimension. We find excellent agreement with this prediction. Improvements in the data, particularly reliably normalized magnetization data and susceptibility data over a wider temperature range, would make this test even more powerful.

ACKNOWLEDGMENTS

This work was supported by NSF DMR Award No. 9874930 and a Research Corporation Cottrell Scholars Award No. CS0769.

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[14] It is unclear to us whether this is formally true below T_C. In any case, we use Eq. (3) as a rough guide to the applicability of the theory.

[15] Sanders Inc., now BAE Information and Electronic Warfare Systems, Nashua, NH.

[16] In particular, we normalized the single ion $LiTbF_4$ magnetization data at low T assuming a g factor of 17.7 and then multiplied the result by the saturation magnetization of $LiHoF_4$.

[17] Recent ultrasound data over a wide range of temperatures below T_c (unpublished) give similar values for the scaling factor when using the above value for the saturation magnetization. The phase shift for bulk ultrasound waves is proportional to $M^2(T)$.

[18] Of course, this measure of scatter should not be taken too seriously since the analysis uses smooth fits of the χ and C_p data.