Universality of the specific heat of Heisenberg magnets near the critical temperature

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Specific-heat measurements for the Heisenberg magnets $RbMnF_3$, Fe, Ni, and EuO near their magnetic-phase-transition temperatures T_c are reanalyzed. We use the theoretically predicted constraints that the specific-heat exponents α and α' above and below T_c be equal to each other and that C_p be continuous at T_c. When these constraints result in systematic deviations of the data from the fitting function, we assume that the deviations are caused by sample inhomogeneities. For those cases we discard as much of data near T_c as is necessary to obtain a statistically satisfactory fit. An analysis in terms of a pure power law yields values for the amplitude ratio A/A' which cover a considerably smaller range than some previous analyses by others had indicated. However, there remain significant, systematic trends in A/A' and in α which can be correlated with the relative strength of the dipolar contribution to the interaction. These trends are inconsistent with estimates of these parameters based on renormalization-group theory. We consider the possibility that the inconsistency is caused by crossover effects associated with the presence of both isotropic short-range and dipolar interactions. These effects may result in effective values for α and A/A' which are not representative of either of the pure systems. We attempt to take crossover behavior into consideration by fitting the data to a function which, in addition to the leading singularity, includes a confluent singularity. With this function, a fairly wide range for the value of the leading exponent is allowed by the data for each material. We choose $\alpha = -0.14$. This value is based on measurements and a pure-power-law fit for the short-range-force system $RbMnF_3$, but theoretical estimates indicate that the leading exponent is numerically very similar for the dipolar case. The resulting amplitude ratios for the dipolar materials EuO and Ni are equal to each other within their uncertainties. They differ significantly from the value characteristic of short-range-force systems as represented by $RbMnF_3$. This difference agrees with renormalization-group estimates for the difference between the values of A/A' for the two cases.

I. INTRODUCTION

Critical exponents, and certain other dimensionless parameters which describe the singularities in various properties near critical points, have been predicted to be universal in the sense that they depend only upon a small number of very general symmetry properties of the system.[1] This prediction was fully supported, and the universality classes were defined in more detail, by the recently developed renormalization-group theory of critical phenomena.[2] A particular consequence of these theories is that isotropic antiferromagnets should exhibit the same critical behavior as isotropic ferromagnets, provided that isotropic short-range forces dominate the interactions for both types of systems. However, dipolar forces, which may become important for some ferromagnets but are negligible for antiferromagnets, are expected to modify the critical behavior.[3,4] The objectives of the present paper are, on the one hand, to determine whether there exists any experimental evidence for this modification and, on the other, to examine whether the available data can be explained solely in terms of these two universality classes. We shall restrict the present discussion to systems with spin dimensionality n (number of degrees of freedom of the order parameter) equal to 3.

Contrary to the prediction of universality, it was stated by Lederman, Salamon, and Shacklette[5] (LSS) that pertinent experimentally determined parameters for the specific heat C_p indicate that ferromagnets belong to one universality class, and differ significantly and systematically in their behavior from that of antiferromagnets. This difference between ferromagnets and antiferromagnets was asserted to exist even in the absence of dipolar forces. We have reexamined the experimental data which led to this statement, and have arrived at a different conclusion. Our fit of the measurements to a pure power law also yields apparently nonuniversal amplitude ratios and exponents, although the range over which particularly the amplitude ratios vary is considerably smaller than that claimed by LSS. We believe that our analysis of the measurements yields parameters which are consistent with the predictions of universality when the existence of crossover effects due to dipolar forces is taken into account by including singular contributions of higher order than the leading power-law singularity in the data analysis.

The wider range of values covered by the parameters of LSS is in part attributable to the fact that certain predictions of scaling[6,7] and renormalization-group theory,[2] which are in some sense more general than the specific values of critical-point parameters, were not utilized in the data analysis of LSS. Specifically, they did not impose the continuity constraint which we shall discuss below. We believe that the predicted relations between criti-

cal-point parameters should be imposed as constraints in the analysis when the determination of the "best" values of the exponents or amplitude ratios is the object. Only these "best" values should then be used to examine the universality of different critical points within a given symmetry class. In this paper we report on new analyses of already existing experimental results for C_p which utilize the pertinent constraints predicted by theory.

None of our new exponents differ very much from those of the original authors,[5,8-12] but some of the amplitude ratios have changed considerably. Although our amplitude ratios for all materials are much closer to each other than earlier estimates had indicated, the differences, both in the exponents and in the amplitude ratios, are still larger than any reasonable error estimates which result from pure-power-law fits. However, these differences do not necessarily indicate a breakdown of universality. They are sufficiently small that instead they can be interpreted as evidence for crossover effects associated with the existence of dipolar interactions particularly in the ferromagnets with low transition temperatures. These effects can be represented by singular higher-order contributions to C_p which were neglected in a pure-power-law analysis. With the large range for the amplitude ratio reported by LSS[5] such an interpretation would have been difficult to accept. When higher-order singular contributions were included in our least-squares fit, all statistical errors became sufficiently large to permit universal values of either the exponents or the amplitude ratios. Theoretical estimates indicate that the leading exponent remains numerically very similar to the short-range-force value even in the presence of dipolar forces. Since we do not have an experimental value of α for a pure dipolar system, we therefore fixed the leading exponents for all materials at the value corresponding to our best experimental estimate for the pure Heisenberg-like systems with short-range forces. We then found a significant difference in the amplitude ratios for dipolar and short-range-force systems which is consistent with an estimate based on renormalization-group theory for the change in the amplitude ratio due to dipolar forces.

II. ANALYSIS

The details of our least-squares method have been discussed adequately before,[11,12] but we would like to examine some more general aspects of the analysis of specific-heat data pertinent to critical points in this section.

Although elsewhere[11] we have already expressed certain other reservations about the method of analysis used by LSS,[5] we believe that an additional serious shortcoming of this method is that it permits C_p to be discontinuous at T_c. Therefore, we would like to discuss the continuity constraint in some detail here. In order to extract the parameters which describe C_p in the limit as $t \equiv T/T_c - 1$ vanishes, the experimental data must be fitted to a function of the type

$$C_p = (A/\alpha)|t|^{-\alpha}[1+g(t)] + B + Et \tag{1}$$

for $T > T_c$, and to the same function with primed coefficients for $T < T_c$. Here we will always impose the constraint $E = E'$ so that the term Et represents the temperature dependent part of a regular contribution. The function $g(t)$ vanishes as $|t|$ goes to zero. When $\alpha < 0$, C_p is finite at T_c and has its maximum value, equal to B, at T_c. For that case it is easy to see that C_p must be continuous, i.e.,

$$B = B'; \tag{2}$$

for a discontinuity in C_p at T_c corresponds to an exponent of zero and would therefore have to be regarded as the leading singularity. Even when $\alpha > 0$, Eq. (2) is expected to be valid, but its validity is not as obvious and follows only from more detailed calculations based upon renormalization-group theory. From this theory, the exponent of the first correction term to the asymptotically dominant contribution has been calculated by two independent techniques.[4,13,14] It is clear from these calculations that corrections to the leading power-law singularity vanish at T_c. For a positive leading exponent, a value of B different from B' corresponds to a singular higher-order contribution which remains nonzero even at T_c. Equation (2) is therefore expected to be valid also for $\alpha > 0$. It is of course possible that a discontinuity at T_c does indeed represent the leading singularity of C_p for a particular type of system. This is the behavior predicted by the Landau theory of phase transitions, and corresponds to $\alpha = 0$. In three-dimensional materials, this case is closely approximated by Ising systems with dipolar interactions; and for that case a fit of C_p to a power law will lead to $B < B'$.[15] However, if it is found on the basis of experimental data that Eq. (2) is not satisfied for a particular case, it must also be concluded that the leading exponent is zero.

In addition to the constraint Eq. (2), it is predicted by scaling[6,7] that the exponents α and α' are equal to each other. Therefore, the constraint

$$\alpha = \alpha' \tag{3}$$

also should be imposed in order to obtain the best values for the parameters.

The data should of course first be fitted to Eq. (1) with all parameters, including α, α', B, and B', independently least-squares adjusted in order to determine whether the statistical errors permit the use of Eqs. (2) and (3) as constraints. When the differences between the resulting values of α and α' or B and B' are considerably larger than the sum

of the corresponding standard errors, then the imposition of Eqs. (2) or (3) as constraints does not appear justified. In that case, it is necessary to decide whether the difficulty is attributable to a departure from theoretical predictions, or whether it is due to experimental factors. Whereas it is usually not possible to make this choice on purely objective grounds, we have assumed for the data under consideration here that the problem, when present, is due to sample inhomogeneities which cause "rounding" of the specific heat near T_c. We have attempted to overcome this shortcoming by omitting data very near T_c from the analysis. Data analyses based on a pure power law, equivalent to Eq. (1) with $g(t) = 0$ even for $|t| > 0$, usually yielded results for α and α' which permitted Eq. (3) to be used as a constraint even when data over a fairly wide range of t were included. However, in some cases Eq. (2) was not consistent with the data unless the range of t was severely restricted. We therefore imposed Eq. (3) as a constraint in all the analyses which we will present in detail in this paper, and we will discuss the applicability of Eq. (2) for each individual material in Sec. III.

III. RESULTS

A. Pure-power-law analysis

1. Nickel

In the first row of Table I we quote the parameters which were obtained for nickel by Connelly *et al.*[8] from their own data. Before the analysis they subtracted a regular contribution from C_p in an attempt to obtain a singular remainder characteristic of the magnetic degrees of freedom. We have not adopted this procedure in our analysis, and have discussed these corrections for the so-called "lattice specific heat" in detail in Sec. IV A of Ref. 12. However, both approaches should yield similar values for the amplitude ratios and exponents. Obviously, B, B', and E will be changed by subtracting a regular term. It is evident that the results of Connelly *et al.* imply that C_p is discontinuous at T_c, contrary to the prediction Eq. (2). Their exponent is of about the right size for Heisenberg systems (see, for instance, Fig. 28 of Ref. 16), but their amplitude ratio is rather low. The low value of A/A' becomes more apparent when one considers the universal parameter

$$\mathcal{P} \equiv (1 - A/A')/\alpha \ . \tag{4}$$

It is known from experiment[16] and theory[17] that $\mathcal{P}$ is only mildly dependent even upon such *relevant* properties of the system as the spin dimensionality. The results quoted by Connelly *et al.* yield $\mathcal{P} = 1.4$, whereas the most reliable measurements for other systems yield $\mathcal{P} \cong 4$ (see Fig. 28 of Ref. 16).

The data of Connelly *et al.* also have been ana-

TABLE I. Comparison of the parameters obtained for several Heisenberg magnets. The resulting units of the specific heat are J mole^{-1} K^{-1}.

Material	Ref.	Range $x = \log_{10}\|t\|$	T_c (K)	$\alpha = \alpha'$	A/A'	A'	B	B'	E	RMSD
Ni	8[a]	$-3.2 \le x \le -1.6$	631.58	-0.10 ± 0.03	1.136	1.416 ± 0.005	13.49 ± 0.03	15.67 ± 0.03	...	...
	10	$-3.5 \le x \le -1.0$	631.52	-0.089 ± 0.002	1.264 ± 0.03	1.46	46.73	47.82	13.0	0.0302
		$-3.5 \le x \le -1.0$	631.581 ± 0.006	-0.095 ± 0.002	1.189 ± 0.013	1.541 ± 0.017	45.43 ± 0.24	47.45 ± 0.18	12.10 ± 0.25	0.0186
		$-3.0 \le x \le -1.0$	631.479 ± 0.017	-0.095 ± 0.002	1.310 ± 0.021	1.461 ± 0.018	46.04 ± 0.29	46.94 ± 0.28	13.74 ± 0.22	0.0137
		$-3.0 \le x \le -1.0$	631.415 ± 0.010	-0.091 ± 0.002	1.396 ± 0.010	1.385 ± 0.006	47.02 ± 0.27		14.83 ± 0.17	0.0155
Fe	10	$-3.8 \le x \le -1.3$	1041.32	-0.120 ± 0.01	1.036 ± 0.015	7.242	84.59	95.56	100.0	0.431
		$-3.8 \le x \le -1.3$	1041.48 ± 0.01	-0.140 ± 0.004	0.929 ± 0.03	8.362 ± 0.202	77.22 ± 0.85	91.78 ± 0.78	97.07 ± 3.62	0.0722
		$-3.8 \le x \le -1.3$	1041.25 ± 0.03	-0.103 ± 0.011	1.406 ± 0.05	5.628 ± 0.23	95.21 ± 3.9		158.42 ± 8.6	0.248
EuO		$-2.3 \le x \le -1.3$	69.372 ± 0.005	-0.044 ± 0.004	1.22 ± 0.03	3.222 ± 0.02	103.7 ± 5.9		27.54 ± 0.8	0.0340
$RbMnF_3$	9	$-3.6 \le x \le -1.2$	83.082 ± 0.001	-0.142 ± 0.002	1.402 ± 0.018	4.496 ± 0.066	97.78 ± 0.43	98.87 ± 0.26	76.64 ± 0.96	0.0761
		$-3.6 \le x \le -1.2$	83.0787 ± 0.0005	-0.135 ± 0.0002	1.462 ± 0.010	4.265 ± 0.024	99.21 ± 0.26		79.09 ± 0.60	0.0806

[a]The data were converted to magnetic specific heat.

lyzed by LSS,[5] who used their own method of analysis which we have discussed in Sec. IV B2 of Ref. 11. These authors used the total specific heat, and obtained the parameters given in the second row of Table I. Our analysis with the same set of data yielded the parameters in the third row. We note that the root-mean-square deviation (RMSD) from the function with the parameters of LSS is a factor of 1.7 larger than the RMSD obtained with our own least-squares-adjusted parameters, indicating that the fit of LSS is far from being a best fit in the least-squares sense. From our analysis it appears that C_p is discontinuous at T_c, and our $B - B'$ has very nearly the same value as that indicated by the parameters of Connelly *et al.* We find $\Phi = 2.0$, which is still rather low.

In order to overcome the problem of an apparently discontinuous C_p, we discarded measurement which were near T_c on the assumption that these results might be influenced by sample imperfections. As more and more data near T_c were eliminated, B and B' became more nearly equal to each other and A/A' increased. We show the results for $|t| \geq 10^{-3}$ in the fourth row of Table I. We obtain $\Phi = 3.3$, which is much closer to the expected range of values. The uncertainties and values of B and B' now reasonably permit the constraint Eq. (2), and when the specific heat was assumed continuous, the values quoted in the fifth row of the table were obtained as the best estimates for Ni. The best value of the exponent is not significantly different from that of Connelly *et al.*, but the amplitude ratio has changed quite a lot. We now have $\Phi = 4.35$, which is in good agreement with values obtained for many other materials.[16]

2. Iron

The parameters reported by LSS[5] for iron are given in the sixth row of Table I. They indicate that C_p is discontinuous and that Φ has the unlikely value 0.3. We analyzed the same data, using our method, and obtained the parameters in the seventh row. Our fit yields a RMSD which is a factor of 6.0 smaller than that obtained by LSS. Our analysis implies a discontinuous C_p, and our parameters yield the very unlikely result $\Phi < 0$.

The data for iron are not as precise and plentiful as those for nickel, and discarding much of the data near T_c would have left us with very little experimental information. We therefore first retained all data with $|t| \geq 10^{-3.8}$ and imposed the continuity constraint Eq. (2), although Eq. (2) is not really permitted as a constraint by the random errors in the data. This approach yielded the parameters in the eighth row of Table I. We found an exponent very similar to that for nickel, and obtained the very reasonable value $\Phi = 3.9$ from the amplitude ratio. It is difficult to guess at systematic errors in the parameters, however, although some feeling for their uncertainties can be obtained from their variation when data near T_c are discarded. We found that discarding all data with $|t| \leq 10^{-2.3}$ yielded $A/A' = 1.3$, $\alpha = \alpha' = -0.10$, and $\Phi = 3.1 \pm 1.3$. These results are consistent with those in row 8 of Table I.

3. Europium oxide

The results of detailed analyses of our specific-heat measurements for EuO have been reported in a separate recent publication.[11] There they have also been compared with the results and the analysis obtained by Salamon[18] who claimed to have observed a crossover from isotropic short-range-force behavior to behavior characteristic of systems with appreciable dipolar interactions. We found no evidence for such a crossover in our or Salamon's data, and refer the interested reader to Sec. V A of Ref. 11 for a detailed discussion of this problem. A power-law analysis of our data suggested that sample inhomogeneities were appreciable, and a continuous C_p was obtained only when all data with $|t| \lesssim 0.005$ were discarded. The parameters obtained from our measurements over the range $0.005 \leq |t| \leq 0.07$, with Eq. (2) as a constraint, are given in the ninth row of Table I. We found that the exponent differed considerably from those for Fe and Ni, and the result $\Phi = 5.0 \pm 0.3$, although not very much out of line, seems somewhat higher than the values for most materials.

4. $RbMnF_3$

The results of detailed analyses of our specific-heat measurements for the isotropic antiferromagnet $RbMnF_3$ have been published elsewhere.[12] We quote in row 10 of Table I the values of the parameters which were obtained when data rather near the transition were included in the fit. These results permit a continuous C_p. Row 11 of Table I contains the parameters which were obtained when Eq. (2) was used as a constraint. For this material we find an exponent which is consistent with theoretical estimates for Heisenberg systems,[19,20] although it differs somewhat from the result for iron and nickel. The amplitude ratio also is reasonable. The result $\Phi = 3.4$, although a little low compared to results for some other materials,[16] cannot really be regarded as seriously out of line.

5. Summary and comparison

In Table I the last row for each material represents our best estimate of the parameters which correspond to a pure-power-law analysis of the specific-heat data. The exponents and amplitude ratios cover ranges which exceed the statistical errors by a wide margin, and they do not appear to fall into one or two groups or universality classes.

TABLE II. Parameters based on a pure-power-law analysis with the assumption of a continuous C_p and the existence of a regular contribution which is linear in t, and with the constraint $\alpha = \alpha'$, over the range $0.005 \leq |t| \leq 0.07$. The quoted errors are standard errors and do not include possible systematic uncertainties.

Material	$\alpha = \alpha'$	A/A'	φ
EuO	-0.044 ± 0.004	1.22 ± 0.03	5.02 ± 0.29
Ni	-0.089 ± 0.004	1.38 ± 0.02	4.30 ± 0.22
Fe	-0.096 ± 0.023	1.30 ± 0.08	3.1 ± 1.3
$RbMnF_3$	-0.137 ± 0.004	1.46 ± 0.02	3.38 ± 0.16

One can argue that the parameters in Table I are effective parameters which do not represent the limiting behavior of C_p as $|t|$ vanishes because the contribution $g(t)$ to Eq. (1) has been neglected in the analysis. Although $g(t)$ contains nonuniversal parameters, one might have conjectured that $g(t)$ should be of about the same size for different materials provided $|t|$ is similar. The pure-power-law fits represented in Table I cover various ranges of t however, and the variation in the resulting parameters could be due to range-dependent contributions from $g(t)$. In order to obtain sets of pure-power-law parameters which can be compared with each other more readily, we therefore fitted the data for each substance over the very restricted range $0.005 \leq |t| \leq 0.07$. Over this range, data which are presumably not influenced by sample inhomogeneities are available for each material. The results for α and A/A' are given in Table II. We would like to base our further discussion on these parameters and the associated statistical errors, although the errors are of course larger than they would be if a wider range of $|t|$ were used. It can be seen that using identical ranges of t for all materials did not greatly alter the parameters, and yielded values which are far from universal. Universality can therefore pertain to these substances only if contributions from $g(t)$ are appreciable at least for some of the substances, and vary considerably in size for different materials.

As an attempt towards finding an explanation for the variation of A/A' and α, we considered the possible influence of dipolar forces on the critical behavior[3,4] in somewhat more detail. For systems with dipolar contributions to the interaction, it has been shown by Fisher and Aharony[3] that the critical-point parameters which describe the behavior of the system as $|t|$ vanishes are different from those characteristic of isotropic short-range-force (SR) systems. Dipolar forces are negligible for antiferromagnets,[3] and therefore we might expect to measure the specific heat parameters characteristic of SR systems in the case of $RbMnF_3$. Dipolar forces are expected to dominate for isotropic ferromagnets with low transition temperatures,[3] but as yet we have no very quantitative measurements for such a material. The ferromagnet EuO occupies an intermediate position[3,4] where dipolar interactions are appreciable, but where purely dipolar behavior would be observed only rather near the transition. In this case we expect to deduce from a pure-power-law fit effective values for A/A' and α which are not representative of either of the pure systems. The values of these effective parameters will reflect the extent to which the crossover from SR behavior towards dipolar behavior has progressed upon approaching T_c through the experimentally accessible range of $|t|$. Both Ni and Fe are ferromagnets with much higher transition temperatures than EuO, and therefore one would expect the dipolar contribution to the interactions to be relatively less important and the critical-point parameters to be nearer those for the SR system.

In Fig. 1 we arranged the four sets of parameters from Table II in order of decreasing dipolar contribution, as judged for the ferromagnets by their transition temperatures and with the antiferromagnet on the extreme right, but we have not felt it warranted at this point to attach a quantitative scale to the separation between successive entries. With-

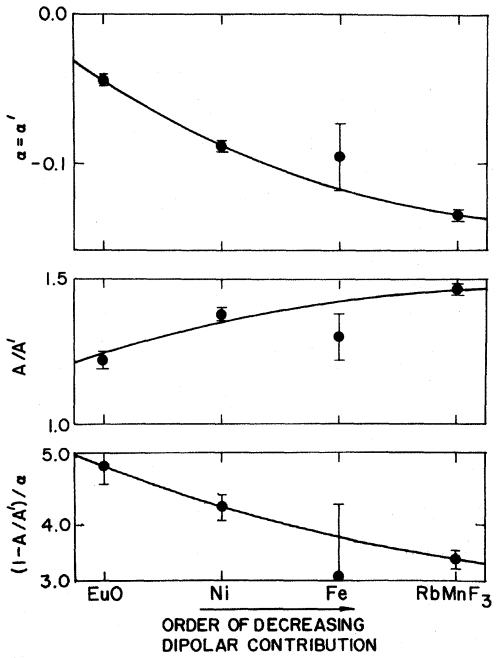

FIG. 1. Parameters obtained from a fit to a pure power law. The four materials are arranged in order of decreasing dipolar contribution to the interaction. No quantitative value should be attached to the spacing between successive material. The solid lines are drawn only to emphasize the monotonic trend of the data, and have no further significance.

in the statistical errors indicated in the figure, it is apparent that there exists a smooth trend for α, A/A', and φ. This observation tends to support the concept of effective parameters which reflect the extent of crossover from one type of behavior to another. Figure 1 suggests that the SR systems with $n=3$ have

$$\alpha_{SR} \cong -0.14 , \tag{5a}$$

$$(A/A')_{SR} \cong 1.46 , \tag{5b}$$

and

$$P_{SR} \cong 3.4 . \tag{5c}$$

It is extremely difficult, however, to estimate the parameters for the pure dipolar case from the data in Fig. 1. The assumption of a monotonic variation of the effective parameters with the strength of the dipolar interaction leads to

$$\alpha_D > -0.04 , \tag{6a}$$

$$(A/A')_D < 1.22 , \tag{6b}$$

and

$$P_D > 5.0 . \tag{6c}$$

We do not regard the monotonicity assumption as reliable, however. Theoretical estimates of effective exponents and amplitude ratios as a function of the relative strength of the dipolar interaction could provide a most useful guidance for the interpretation of the experimental data represented in Fig. 1. The SR results given by Eq. (5) are consistent with those obtained by high-temperature series expansions which have yielded $\alpha = -0.14 \pm 0.06$[19] and $\alpha = -0.09 \pm 0.03$,[20] and with an expansion in $4-d$ (d is the dimensionality of the system) based on renormalization-group theory[2] which yielded $\alpha = -0.135$ and $A/A' = 1.36$.[21] However, the dipolar parameters given by Eq. (6) disagree rather severely with renormalization-group estimates from an expansion in $4d$ which yield $\alpha_D \approx \alpha_{SR}$[4] and $(A/A')_D > (A/A')_{SR}$.[22]

In an attempt to find an explanation of the results in Fig. 1 we also considered the possible influence of anisotropy upon the specific-heat parameters. Because of the symmetry of the crystals, any anisotropy would be cubic. Crossover to behavior characteristic of the cubic universality class for systems which exhibit SR behavior far from T_c has been examined theoretically by Aharony.[23] This crossover is expected to occur when $|t| \approx u^{1/\varphi}$, where u is the ratio of the anisotropy energy to the exchange energy, and where $1/\varphi$ was estimated to be about equal to 18.[24] The cubic behavior, therefore, is not expected to dominate until $|t|$ is much smaller than the experimentally accessible range for any material with $u \ll 1$. Furthermore the cubic exponents, although different in principle, were estimated by Aharony[23] to be numerically very similar to the SR exponents, and probably could not be distinguished from them by experiment. The crossover to cubic behavior in the presence of dipolar forces has been treated by renormalization-group theory by Bruce and Aharony.[4] They find $1/\varphi \cong 5.6$ for this case. Although this value is smaller than the corresponding one for crossover from SR behavior, it is still much too large to permit experimental measurements at values of t where cubic behavior should dominate. The exponents corresponding to and the existence of a cubic universality class in the presence of dipolar forces apparently have not yet been investigated.[4] In any event, it is not expected that the small cubic anisotropy in the materials under consideration here would have a noticeable influence on the measured parameters.

B. Analysis with correction terms

In Section III A 5 we indicated that the apparently nonuniversal behavior of the specific-heat parameters could be interpreted in terms of crossover behavior resulting in effective parameters which deviate from those characteristic of the SR systems by various amounts. We now would like to present an interpretation in which such crossover effects are taken into consideration explicitly by including the term $g(t)$ of Eq. (1) in the data analysis. We shall approximate this term by

$$g(t) = D|t|^x \tag{7}$$

for $T > T_c$, and by a similar expression with primed parameters for $T < T_c$. The exponent x describes the first correction term to C_p. For SR systems, x has been estimated from renormalization-group theory both by an expansion in $4-d$,[4,13] and by using the approximate recursion relations.[14] Although the $4-d$ expansion yields results which depend upon the order to which terms are retained, all these calculations are consistent with $x \cong 0.5 \pm 0.2$. They indicate that x is only very mildly dependent upon the spin dimensionality n. Experimental measurements for the superfluid transition in liquid helium (SR, $n=2$) yielded $x = 0.5 \pm 0.1$.[25] A very similar value has been obtained also from high-temperature series expansions for the Ising model ($n=1$).[26] However, in Eq. (7) we need to use the value of x characteristic of dipolar systems with $n=3$. For this case, an expansion in $4-d$ to second order[4] indicates that $x_D - x_{SR} \approx 0.1$. But none of our results are very sensitive to the choice of x, and we therefore analyzed the data in terms of the function

$$C_p = (A/\alpha)|t|^{-\alpha}(1 + D|t|^x) + B , \tag{8}$$

with

$$x = x' = 0.5 , \tag{9}$$

where this value of x is based largely on experi-

TABLE III. Amplitude ratios A/A' of the leading singularity, and the parameter $\mathcal{P} = (1 - A/A')/\alpha$, obtained by fitting to Eq. 8 with two choices for the fixed leading exponent α.

	$\alpha = -0.14$		$\alpha = -0.10$	
	A/A'	$\mathcal{P}$	A/A'	$\mathcal{P}$
EuO	1.812 ± 0.036	5.80 ± 0.26	1.516 ± 0.022	5.16 ± 0.22
Ni	1.759 ± 0.032	5.42 ± 0.23	1.489 ± 0.019	4.89 ± 0.19
$RbMnF_3$	1.420 ± 0.024	3.00 ± 0.17	1.281 ± 0.017	2.81 ± 0.17

mental measurements for liquid helium (SR, $n = 2$).[25] We retained the constraints Eqs. (2) and (3). In addition, we wanted to fix the leading exponent at the value appropriate to the dipolar universality class. We do not know this value from experiment, but the $4 - d$ expansions indicate that α_D is not much different from α_{SR}. We therefore used our best experimental estimate $\alpha = -0.14$ for the SR system [Eq. (5a)] and assumed that it was also representative of α_D. We also performed the analysis with $\alpha = \alpha' = -0.10$, however, because we wanted to see whether any of the conclusions are sensitive to the choice of α. In order to make the comparison of results for the several materials more reasonable, we again restricted the range of t to $0.005 \leq |t| \leq 0.07$ in all cases, even though reliable data nearer T_c were available for some. We did not find it useful to analyze the data for Fe in this manner because the statistical errors would have been too large. The results for A/A' and $\mathcal{P}$ are given in Table III. In Fig. 2, A/A' is shown graphically, again with the materials arranged in order of decreasing dipolar interactions as was done in Fig. 1.

The results of the data analysis in terms of Eq. (8) are consistent with all existing theoretical predictions. From calculations based on renormalization-group theory it is expected that the leading dipolar specific-heat exponent, although different in principle from α_{SR}, remains numerically almost unaltered by the dipolar forces.[4] This result is of course consistent with the use of the same fixed value of α for all three materials represented in Table III. Further, it has been estimated that $(A/A')_D > (A/A')_{SR}$.[22] Unfortunately, the calculation for the dipolar case has been done only to zeroth order in $4 - d$, but to that order the calculation yields $(A/A')_D - (A/A')_{SR} = 0.2$. We see from the data in Fig. 2 that Ni and EuO have the same value of A/A'. This value presumably is characteristic of the $n = 3$ dipolar universality class. The experimental value for the SR system $RbMnF_3$ is smaller than the experimental $(A/A')_D$ by 0.4 or 0.2, depending somewhat on the choice of α_D. This difference in A/A' is in remarkably good agreement with the calculation.

IV. SUMMARY AND CONCLUSION

In this paper we presented the results of a reanalysis of specific-heat data for magnetic materials with spin dimensionality $n = 3$. It was our objective to determine what evidence exists for a difference in critical behavior between systems with only isotropic short-range (i.e., exchange) interactions, and systems with an appreciable dipolar contribution to the interaction. In addition, we wanted to determine whether all available experimental data could be explained in terms of these two universality classes. In our analysis we imposed certain theoretically predicted relations between specific-heat parameters as constraints. Specifically, we used the scaling law $\alpha = \alpha'$ and insisted that the specific heat be continuous at T_c (note that $\alpha < 0$ for $n = 3$ and therefore C_p is finite at T_c). The purpose of using these constraints was to obtain, separately for each material, the best set of parameters which, taken by itself, was consistent with theoretical predictions. We then proceeded to compare the parameters for the various materials with each other in order to see whether they are consistent with the predicted universal behavior of materials whose Hamiltonians belong to the same universality class.

We first analyzed the measurements by fitting them to a pure power law. We obtained values for the specific-heat exponents α which were rather similar to earlier estimates by others, but the use of the continuity constraint resulted in amplitude ratios A/A' which differed far less for different

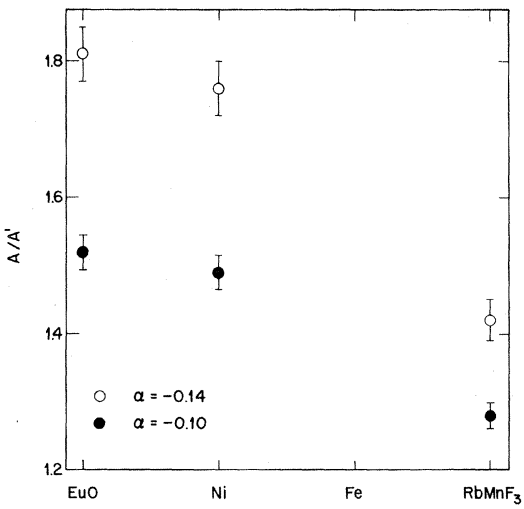

FIG. 2. Amplitude ratios for the leading singularity obtained from fitting to a function which includes a confluent singularity in addition to the leading power-law term. The leading exponent α was fixed at the two values given in the figure. As in Fig. 1, the materials are arranged in order of decreasing dipolar contribution to the interaction.

materials than previous analyses had indicated. Even though our exponents and amplitude ratios covered only fairly narrow ranges of values, the differences between parameters for different materials was well outside of the statistical errors. A monotonic trend in $\alpha = \alpha'$, A/A', and $(1 - A/A')/\alpha$ with the relative strength of the dipolar contribution to the interaction was evident (Fig. 1). Although this trend is consistent with the existence of different critical-point parameters for dipolar and isotropic short-range-force systems, there is no assurance that any of the materials represent the pure dipolar case. We do expect, however, that the antiferromagnet $RbMnF_3$ represents the pure isotropic short-range case because dipolar forces are believed to be absent in antiferromagnets. Except for $RbMnF_3$, it is likely that the parameters in Fig. 1 are effective parameters which do not describe the limiting behavior of C_p as t vanishes, but rather are determined by the extent to which the experimental range of t coincides with a rather wide crossover region.

Next, we analyzed the measurements in terms of a power law for the leading singularity, but included also a correction to this leading term in the form of a confluent singularity. The additional parameters introduced enough flexibility to permit the assumption of a universal leading exponent for all materials under investigation. We fixed $\alpha = \alpha'$ at the value -0.14. This is our best experimental estimate for the isotropic short-range case, but calculations based on renormalization-group theory indicate that the leading exponent, although changed in principle, remains numerically very similar to the short-range value even when dipolar forces are present. For EuO and Ni, we obtained the same amplitude ratios for the leading singularity. We presume that this value is characteristic of dipolar systems. The value of A/A' for $RbMnF_3$ was considerably smaller than the one for EuO and Ni, and we assume it to be representative of SR systems with $n = 3$. The difference in A/A' between the two classes is consistent with calculations based on renormalization-group theory, which yielded the prediction $(A/A')_D > (A/A')_{SR}$. However, the calculations are available only to zeroth order in $4 - d$, where d is the dimensionality of the system. Obviously they must be regarded with great caution when applied to the case $d = 3$, but it is not unreasonable to expect that they might yield correctly the relative size of $(A/A')_D$ and $(A/A')_{SR}$. In any event, they are consistent in this respect with our analyses for the real materials.

Our analysis of specific-heat data has revealed no evidence for a systematic difference in critical behavior between ferromagnets and antiferromagnets which cannot be attributed to the presence of dipolar forces in the case of the ferromagnets. This conclusion differs from that of Lederman, Salamon, and Shacklette.[5]

We believe that the pure-power-law parameters for the antiferromagnet $RbMnF_3$ are representative of Heisenberg systems with isotropic short-range interactions. There appear to be no measurements at this time, however, which are representative of the strong dipolar case with spin dimensionality $n = 3$. For all ferromagnets examined so far the experimental range of t appears to lie in a region in which the pure-power-law parameters have effective values which are not representative of either the dipolar or the short-range case. It would be interesting to obtain precise measurements on a good sample of an isotropic ferromagnet with a lower transition temperature than that of EuO.

Note added in proof: Very recent measurements [A. Kornblit and G. Ahlers (unpublished)] of the specific heat of EuS have yielded $\alpha = \alpha' = -0.130 \pm 0.003$ and $A/A' = 1.54 \pm 0.02$ from a pure-power-law fit over the range $0.005 \leq |t| \leq 0.07$ (to be compared with Table II). For EuS, $T_c = 16.51$ K and dipolar forces are thus expected to be more important than for EuO. As expected from theory (Ref. 4) for pure dipolar systems, the exponent agrees well with the $n = 3$ SR value obtained for $RbMnF_3$. The amplitude ratio is not as large as had been estimated for the pure dipolar case by using Eq. (8) and the data for EuO and Ni. The EuS data and Eq. (8) yield $A/A' = 1.63$ if $\alpha = -0.14$, and $A/A' = 1.41$ if $\alpha = -0.10$, to be compared with Table III. Consistent with the prediction for pure dipolar systems (Ref. 22), these ratios are still larger than the SR values based on the measurements for $RbMnF_3$.

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