PHYSICAL REVIEW B

COMMENTS AND ADDENDA

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Heat capacity of FeF₂ near the antiferromagnetic transition

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Already existing results for C_p of the uniaxial antiferromagnet FeF₂ near the transition temperature are shown to yield the exponents $\alpha = \alpha' = 0.16$ and the amplitude ratio A/A' = 0.53. As expected from theory, these results are very different from those for the isotropic antiferromagnet RbMnF₃, but they are consistent with the exponents and amplitude ratio obtained for liquid-gas critical points, and thereby support the theoretically predicted universality of critical-point parameters.

The concept of universality¹ implies that critical exponents² and certain other dimensionless parameters which describe the behavior of systems near critical points will be identical for apparently quite different physical systems. It is expected that these parameters depend only upon such general properties as the physical dimensionality d of the system and the number of degrees of freedom n of the order parameter. This universality principle has received strong support recently from explicit calculations of exponents 3,4 and of the equation of state.^{5,6} There also appears to be some verification of this principle from experiments, 7-9 but quantitative experimental confirmation is still incomplete, and a violation of universality seems to exist near the superfluid transition in liquid He⁴.⁹ In this note, we present the results of a new analysis of specific-heat measurements¹⁰ for the uniaxial antiferromagnet FeF2. For this system the physical dimensionality is d=3, the order parameter effectively has only one degree of freedom because of the anisotropy, and the forces in antiferromagnets are short range.¹¹ In every respect known to be important for the values of the critical-point

parameters, FeF_2 is therefore identical to the Ising model or the liquid-gas critical point, and these three systems should have identical parameters. Examination of critical exponents for Ising-like systems seems particularly warranted in view of certain differences which seem to exist⁸ between experimental results for liquid-gas critical points¹²⁻¹⁴ and numerical calculations for Ising systems.¹⁵⁻¹⁷

We obtained values for the specific-heat exponents α and α' from previously existing experimental results¹⁰ for C_{ρ} by using a method of analysis which has been described in detail elsewhere.^{18,19} We used all available data for which $|t| > 10^{-3}$, where

$$t = T/T_c - 1 , \qquad (1)$$

and where T_c is the transition temperature. Results with smaller |t| were discarded because we believed them to be excessively affected by inhomogeneities in the sample. The available measurements spanned the range $-0.1 \le t \le 0.05$. They were fitted to the function

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TABLE I. Parameters of Eq. (2) for FeF₂. The resulting units of C_p are cal mole⁻¹ K⁻¹. The uncertainties are standard errors and do not reflect possible systematic errors in the original data. Numbers in column 2 are for the seven-parameter fit ($\alpha = \alpha'$, D = D' = 0, and E = E'). Numbers in column 3 are for the nine-parameter fit ($D, D' \neq 0, x = 0.5$, and $\alpha = \alpha'$). See text for further description of the analysis.

Parameters	without singular correction terms	with singular correction terms
$\overline{\alpha} = \alpha'$	0.157 ± 0.016	0.112 ± 0.044
A/A'	0.528 ± 0.049	0.65 ± 0.43
A'	0.684 ± 0.054	1.13 ± 0.35
$B - A/\alpha$	2.72 ± 0.55	-6.0 ± 2.3
$B' - A'/\alpha'$	4.03 ± 0.95	-5.9 ± 4.9
$T_c = T_c'$	78.257 ± 0.007	78.261 ± 0.018
$\vec{E} = \vec{E'}$	13.0 ± 1.2	9.4 ± 2.6
D	0ª.	5.9 ± 2.4
D'	0 ^a	3.0 ± 2.0
x	• • •	0, 5ª

^aFixed.

$$C_{\mathbf{a}} = (A/\alpha)(|t|^{-\alpha} - 1)(1 + D|t|^{\mathbf{x}}) + B + Et \qquad (2)$$

for t > 0, and to the same function with primed coefficients for t < 0 (for details, see Ref. 19), with various constraints upon the parameters.

For the specific heat of liquid He⁴ near the superfluid transition⁹ it was necessary to invoke the higher-order singular correction terms represented by the contribution $D | t |^x$ in Eq. (2) in order to obtain consistency between the measurements and the scaling prediction^{2,20,21}

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

We wished to explore first whether it was necessary to consider these terms also in the case of FeF₂. For that purpose, we initially forced D=D'=0. In addition, the constraint E=E' was imposed, assuring that the term *Et* represents the temperature dependence of any regular contributions to C_p .¹⁹ Further, we used the physically reasonable constraint $T_c = T'_c$ and permitted A, α , and B to be different from A', α' , and B'. The resulting eight-parameter fit to the data yielded

$$\alpha = 0.12 \pm 0.11$$
; $\alpha' = 0.19 \pm 0.09$. (4)

This result is consistent with Eq. (3) and allows the constraint $\alpha = \alpha'$. It is therefore not necessary to invoke singular corrections to the asymptotically dominant contribution to C_p in order to obtain consistency between scaling and the measurements.

Since there appears to be little point in inquiring into the validity of universality if scaling fails, the scaling constraint Eq. (3) was next imposed upon the data analysis. There remained seven parameters and the fit yielded

and

$$\alpha = \alpha' = 0.157 \pm 0.02 \tag{5}$$

 $A/A' = 0.53 \pm 0.05$ (6)

The value for the exponents agrees with the earlier estimate¹⁰ of α , but differs appreciably from the previous¹⁰ $\alpha' = 0$. All the parameters of Eq. (2) with the constraints of this analysis are collected in Table I, column 2.

Although it was not necessary to invoke singular correction terms in order to obtain consistency with scaling, these terms may in general be expected to be present, 9,22,23 but their amplitudes D and D' will depend upon the particular system. We therefore next permitted D and D' to be nonzero: but for x we used the theoretically²² and experimentally^{9,23} suggested value x = 0.5. This analysis therefore involved nine parameters. Under these circumstances it was difficult to obtain convergence of the iterative nonlinear least-squares calculation, but we believe the parameters in column 3 of Table I to be reasonably good estimates of the least-squares values. We find that the values of $\alpha = \alpha'$ and of A/A' do not differ much from Eqs. (5) and (6), but the uncertainties are larger.

For liquid-gas critical points the experimental values of α and α' lie in the range $0.06-0.14^{-0.14}$ and those for A/A' are rather close to $0.5.^{12-14}$. The spread in these exponents is probably attributable to differences in the data analysis and to experimental errors rather than to nonuniversal behavior. Equations (5) and (6) are therefore in good agreement with the results for liquid-gas critical points. They are very different, however, from those for the *isotropic* antiferromagnet RbMnF₃.^{18,19} This difference is expected, because for the isotropic system the number of degrees of freedom n of the order parameter is three, whereas for the uniaxial FeF₂, n is equal to one. For the liquid-gas critical points we also have n = 1, and there-

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fore expect the same α , α' , and A/A' as for FeF₂. The dependence of the specific-heat exponent and amplitude ratio upon n has been discussed in detail elsewhere.²⁴

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