# Heat capacity of EuO near the Curie temperature

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Precision measurements of the heat capacity $C_p$ of the Heisenberg ferromagnet EuO near the Curie temperature $T_c \approx 69.3$ K are reported. The data are for $64 \leq T \leq 73$ K. When they are analyzed in terms of pure-power-law singularities over the range $0.003 \leq |t| \equiv |1 - T/T_c| \leq 0.07$, they permit equal exponents $\alpha$ and $\alpha'$ above and below the transition, and a continuous specific heat at $T_c$. They yield $\alpha = \alpha' = -0.044 \pm 0.01$ and an amplitude ratio $A/A' = 1.22 \pm 0.06$ when $C_p$ is assumed to be continuous. Although these results are consistent with scaling predictions, they disagree with estimates based on renormalization-group calculations for isotropic ferromagnets with dipolar interactions. When, in addition to the leading power-law term, singular correction terms are also included in the analysis and $C_p$ is assumed to be continous at $T_c$, one finds $\alpha = \alpha' = -0.10 \pm 0.05$, $A/A' = 1.50 \pm 0.2$, and a correction exponent $x = 0.56 \pm 0.2$. These latter results are consistent with all theoretical predictions.

## I. INTRODUCTION

In a previous publication[1] we have reported in detail about measurements and interpretations of the specific heat $C_p$ of the isotropic antiferromagnet $RbMnF_3$. This material was expected to serve as a model substance for systems with short-range forces and three degrees of freedom in the order parameter (spin dimensionality $n = 3$). Indeed, those measurements yielded a specific-heat exponent and an amplitude ratio which agreed very well with theoretical predictions.[2-7] We now wish to report on our measurements for the isotropic ferromagnet EuO. In ferromagnets, there is an appreciable contribution to the interactions from dipolar forces, and for that reason it is expected from theory that the critical-point parameters should differ from those which describe the behavior of short-range force systems.[5,6] Unfortunately, the predicted difference in the parameters is only small,[6] and therefore difficult to detect by experiment. We find, however, that our specific-heat data, when fitted to a pure power law, yield a critical exponent which differs considerably from that of $RbMnF_3$, and from calculations for either short range or dipolar interactions. Only when we invoke singular correction terms to the asymptotically dominant term do we obtain reasonable consistency with the theoretical predictions.

Throughout the remainder of this paper we have retained the same structure that was adopted in Ref. 1, and therefore the two papers easily can be read in parallel. Section II, pertaining to the apparatus, is kept extremely short since the details of the experiment have been described before.[1] Section III contains our results, and in Sec. IV these data are analyzed in order to obtain the critical-point parameters. In Sec. IV B2, we also discuss a method of analysis which has been employed recently by others, and which has yielded critical-point parameters for EuO which are rather different from ours. In Sec. V we compare our results with those obtained elsewhere, and Sec. VI provides a brief comparison with pertinent theoretical predictions. A short summary is given in Sec. VII.

A brief report on part of this work already has been presented.[8]

## II. APPARATUS AND METHODS

The calorimeter used in this work was the same as that employed for previously reported measurements on $RbMnF_3$, and the reader is referred for details to Sec. II A of Ref. 1. We used the temperature scale based on a calibration of a platinum thermometer by Cryocal, Inc. (Sec. II B of Ref. 1).

In the work described here two samples of EuO were used. They were both grown from the melt by E. Buehler, using the Bridgman technique. Sample I was used also by others for thermal expansion and acoustic measurements,[9,10] and was cut from the same main crystal which yielded the sample used for specific-heat measurements by Salamon.[11] Sample II consisted of a few single crystals. Although it was grown by the same technique as sample I, higher-purity europium was used in the process. EuO I weighed 5.284 g and EuO II weighed 4.196 g. A molecular weight of 167.96 g was used in the conversion of the data to a molar basis.

## III. RESULTS

The results for EuO I and EuO II are shown in Figs. 1 and 2. Those for sample I were reported briefly previously.[8] The data for EuO I show a somewhat sharper transition with a very slightly higher maximum than that observed for sample II. In order to represent the results conveniently in closed form, the data were fitted to the function[12]

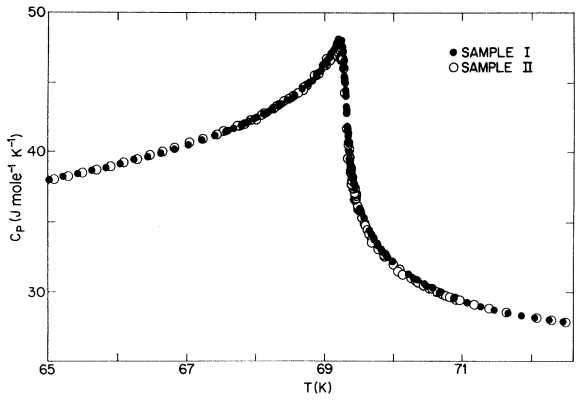


FIG. 1. Specific heat of EuO (samples I and II) as a function of $T$.

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

with $t \equiv (T - T_c)/T_c$ for $T > T_c$, and to the same function with primed parameters for $T < T_c$. In our analysis we found that $E$ and $T_c$ could be forced to be equal to $E'$ and $T_c'$, respectively, without any significant sacrifice in the quality of the fit, but $\alpha$ could be forced to be equal to $\alpha'$ only if the fitting was limited to the range $|t| \gtrsim 3\times10^{-3}$ (see Sec. IV C). The parameters obtained with those constraints are listed in Table I. We would like to emphasize that these parameters are given primarily as a closed-form representation of the data, and that they do not necessarily provide a description of $C_p$ in the limit as $|t|$ vanishes. A discussion of the asymptotic behavior of $C_p$ is given below in Sec. IV C. For EuO I the deviations in percent of the data from Eq. (1) with the parameters in Table I are shown in Fig. 3. The function represents the data quite well for $|t| \gtrsim 3\times10^{-3}$, but for $|t| \lesssim 2\times10^{-3}$ the deviations are over 1%. These deviations are attributed primarily to sample inhomogeneities which cause the transition to be "rounded," and to a lesser extent to the earth's magnetic field which becomes important for $|t| \lesssim 10^{-3}$.[13] The data very near the transition are shown on an expanded linear scale in Fig. 4, where the "rounding" is also evident.

## IV. ANALYSIS

### A. Function

For a comparison of our measurements with theoretical predictions, it is necessary to extract from the data estimates of parameters which describe the behavior of $C_p$ in the limit as $|t|$ vanishes. This very difficult problem has been discussed in detail in Sec. IV of Ref. 1, and we shall not elaborate upon it much further here. We shall

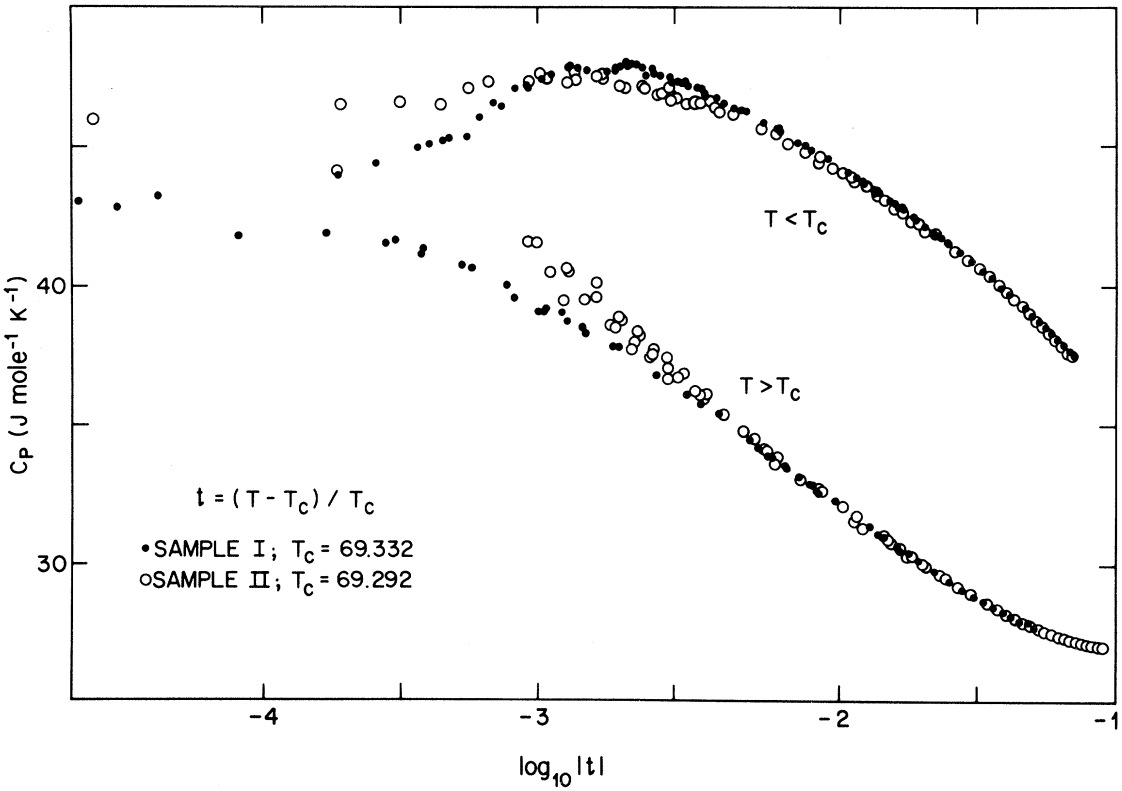


FIG. 2. Specific heat of EuO (samples I and II) as a function of $\log_{10}|T/T_c - 1|$.

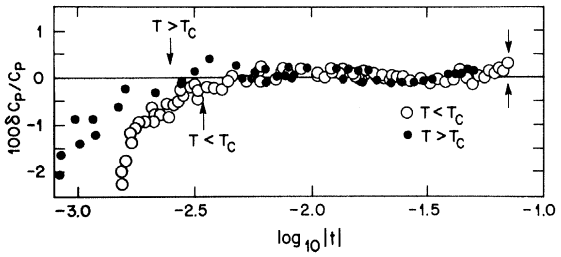


FIG. 3. Deviation of the data from the best fit. Solid circles: $T > T_c$; open circles: $T < T_c$. The arrows mark $t_{min}$ and $t_{max}$.

fit our results to the equation

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

for $T > T_c$, and to the same equation with primed parameters for $T < T_c$. When $D = D' = 0$, Eq. (2) is identical to Eq. (1). During the analysis, various reasonable constraints, partly suggested by theory, will be imposed upon the parameters. We refer the reader to Sec IV A of Ref. 1 for a more detailed discussion of Eq. (2).

### B. Methods

#### *1. Method used here*

The method used here was the same as that employed in our analysis of the $RbMnF_3$ specific-heat data,[1,8] and the reader is referred to Sec. IV B, Ref. 1 for more details. All the parameters above and below the transition were determined simultaneously by a least-squares fit with explicitly specified constraints imposed upon the parameters. All data points were given weights $W_i$ which were inversely proportional to the square of the estimated random probable error. We used the smaller of

$$W_i = (3\times 10^{-5} C_p/\Delta T)^{-2} \qquad (3a)$$

or

$$W_i = (10^{-3} C_p)^{-2} \qquad (3b)$$

for the $i$th data point. The number $3\times 10^{-5}$ in Eq. (3a) is the estimated temperature resolution in K, and $10^{-3}C_p$ in Eq. (3b) corresponds to a random error of 0.1% which is assumed when temperature resolution does not limit the accuracy of the data.

Since the transition region was "rounded," points close to the transition had to be excluded. We defined a reference temperature $T_0$ (see Sec. IV B of Ref. 1) and fitted data for which $|t| \geq t_{min}$ where

$$t_{min} \equiv |T' - T_0|/T_0$$

for several arbitrarily chosen $t_{min}$. The values of $T_0$ were 69.30 and 69.33 K for samples I and II, respectively. Similarly, we defined $t_{max}$ and fitted the data for $|t| \leq t_{max}$ for several arbitrarily chosen $t_{max}$. This was done to test our assumption that the term $Et$ adequately represents the temperature dependence of regular contributions to the specific heat. Clearly, close to $T_c$, this assumption is correct, but far from the transition higher-order regular terms must be considered.

TABLE I. Parameters for Eq. (1). The resulting units of $C_p$ are J mole$^{-1}$ K$^{-1}$.

| Parameter | EuO I ($0.003 \leq \|t\| \leq 0.07$) | EuO II ($0.005 \leq \|t\| \leq 0.07$) |
|---|---|---|
| $\alpha = \alpha'$ | −0.0418 | −0.0473 |
| $A$ | 4.189 | 4.350 |
| $A'$ | 2.907 | 2.803 |
| $B$ | 114.32 | 105.55 |
| $B'$ | 101.97 | 92.11 |
| $E = E'$ | 33.545 | 37.073 |
| $T_c$(K) | 69.332 | 69.292 |

#### *2. Other methods*

A variety of other methods of analysis of specific-heat data near critical points has been devised by other authors. Most of them are designed to avoid the difficulties inherent in least-squares analyses involving functions which are nonlinear in their parameters. They often are a compromise between computational ease and objectivity, frequently involve some subjective judgment, and sometimes lead to unrealistic estimates of statistical errors for the relevant parameters.

We would like to discuss here specifically a new technique which has been introduced recently[11] for the analysis of $C_p$ of EuO,[11] $RbMnF_3$, Fe, Ni, and certain copper salts.[14] We wish to discuss this method in detail because it has been used to obtain

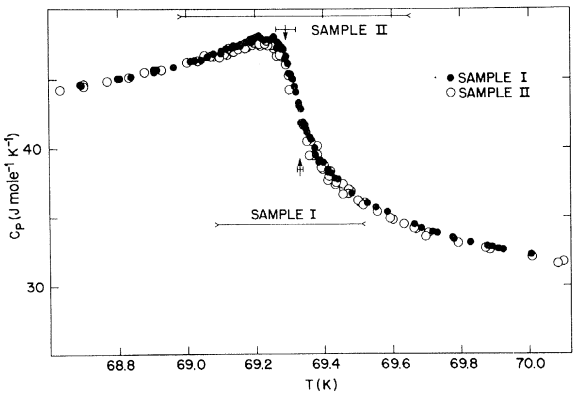


FIG. 4. Specific heat of EuO near the transition. The arrows indicate $T_c$ with its uncertainty as obtained by least-squares fits for the two samples. The horizontal bars indicate the ranges of data that were excluded from the corresponding fits.

parameters for EuO which are different from ours.[11] In particular, the method was believed to yield a so-called "crossover" from short-range force to dipolar behavior for which we find no evidence. In this analysis, $C_p$ is written as

$$C_p^+ = (A/\alpha)(|t|^{-\alpha}-1)+B+Et \tag{4a}$$

for $T > T_c$, and

$$C_p^- = (A'/\alpha')(|t|^{-\alpha'}-1)+B'+Et \tag{4b}$$

for $T < T_c$. By combining Eqs. (4a) and (4b), one can obtain

$$\tilde{C}_p^- = (A'/\alpha')\{[(\alpha/A)(\tilde{C}_p^+ - B)+1]^{\alpha'/\alpha}-1\}+B'. \tag{5}$$

Here

$$\tilde{C}_p^- = C_p^- - Et \tag{6a}$$

and

$$\tilde{C}_p^+ = C_p^+ - Et. \tag{6b}$$

If $\alpha = \alpha'$, Eq. (5) reduces to

$$\tilde{C}_p^- = (A'/A)\tilde{C}_p^+ + (B' - A'B/A). \tag{7}$$

Therefore a test of the equality $\alpha = \alpha'$ is expected to be provided by the linearity of the plot of $\tilde{C}_p^-$ vs $\tilde{C}_p^+$ for equal values of $|t|$. If the plot is linear, $\tilde{C}_p^+$ can be "transformed" to $\tilde{C}_p^-$ using Eq. (7). The method of analysis then consists of fitting all the data to the three-parameter equation

$$C_p^{\text{merged}} = (A'/\alpha)(|t|^{-\alpha}-1)+B' \tag{8}$$

by a least-square procedure. Once these parameters are found, $A$ and $B$ can be determined from Eq. (7). The technique has produced rather small estimates of statistical errors for the parameters of the specific heat of EuO,[11] iron, nickel, and copper salts.[14] Sometimes it has led to different results[11] than the so-called "conventional" least-squares analysis. It has been claimed[11,14] that this method has the advantage over a conventional least-squares fit of testing the equality $\alpha = \alpha'$ without any commitment to specific values for the other parameters.

We would like to point out that the method described above has certain shortcomings. First, in order to plot $\tilde{C}_p^-$ vs $\tilde{C}_p^+$ for equal $|t|$, $T_c$ must be known in advance. Although for large $|t|$ the plot of $\tilde{C}_p^-$ vs $\tilde{C}_p^+$ is relatively insensitive to the choice of $T_c$, the behavior of data near $T_c$ will be strongly influenced. A second arbitrary decision involves the choice of the constant $E$. The value of this constant will not strongly affect the behavior close to the transition, but will be significant far from $T_c$. In order to illustrate this point, we will use as an example the $RbMnF_3$ results which can be represented by Eq. (4), with the parameters listed in Table I of Ref. 1. The constant $E$ was found[1] to be 76.638 J mole$^{-1}$K$^{-1}$. After subtracting the term $Et$ from $C_p^-$ and $C_p^+$ to obtain $\tilde{C}_p^-$ and $\tilde{C}_p^+$, respectively, one can get a straight line with a slope (equal to $A'/A$) of 0.71 by plotting $\tilde{C}_p^-$ as a function of $\tilde{C}_p^+$. This line is shown in Fig. 5. The constant $E$ was found by a least-squares procedure; but on the basis of a less-objective inspection of the data one could conceivably also choose the value $E$ = 70 J mole$^{-1}$K$^{-1}$. If $Et = 70t$ is subtracted from $C_p^-$ and $C_p^+$, one obtains the dashed line plotted in Fig. 5. Clearly, this line is not straight. In that case one could conclude that $\alpha \neq \alpha'$, and analyze the data accordingly. Indeed this is a possibility considered in Ref. 11 when the EuO specific-heat data were analyzed. Alternately, one could assume the validity of the theoretical prediction $\alpha = \alpha'$, and divide the data into two regions. An "outer region" with $0.06 \leq |t| \leq 0.2$ has a slope (equal to $A'/A$) of approximately 1.06, and an "inner region" with $0.001 \leq t \leq 0.06$ has a slope of about 0.75. It can be seen that the wrong choice of $E$ did not strongly affect the value of $A/A'$ in the region near $T_c$. However, it artificially produced an apparent "crossover" effect from an "inner" to an "outer" region. We shall return to this effect later in Sec. V A when we discuss the crossover behavior which has been claimed by Salamon[11] for EuO.

In a more recent version of the above method of analysis[14] the data are analyzed in terms of Eq. (7) by maximizing the linear-correlation coefficient $r$ by varying both $T_c$ and $E$. The attempt to linearize $\tilde{C}_p^-$ vs $\tilde{C}_p^+$ is equivalent to trying to get as close as possible to the equality $\alpha = \alpha'$. The values obtained for $E$ and $T_c$ by maximizing $r$ are not necessarily the values that would have been obtained by least squares adjusting all the parameters, however. They will be equal to the over-all least-squares values only if the complete least-squares

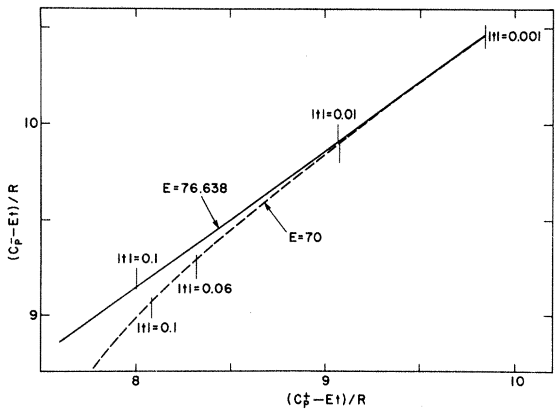


FIG. 5. $C_p^- - Et$ vs $C_p^+ - Et$ for $RbMnF_3$ (using the fitted function). Solid line: $E = 76.638$ J mole$^{-1}$K$^{-1}$; broken line: $E = 70.0$ J mole$^{-1}$K$^{-1}$.

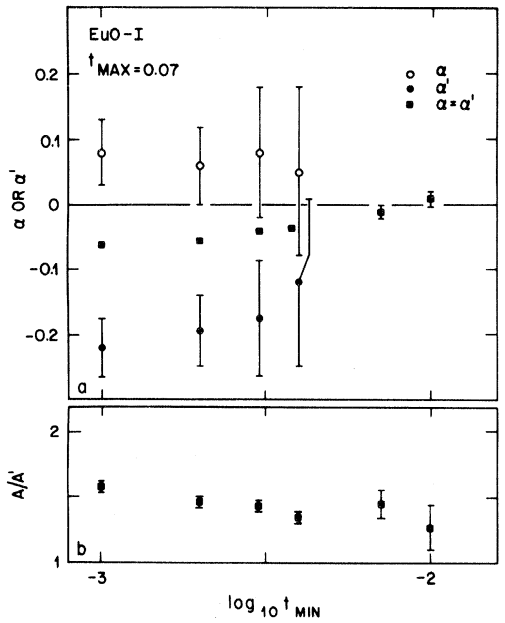


FIG. 6. $\alpha$, $\alpha'$, and $A/A'$ obtained from a fit of the data to Eq. (1) for various $t_{min}$, with the constraint $E=E'$. When error bars are omitted, they are comparable to or smaller than the size of the point.

analysis, with $\alpha$ and $\alpha'$ independent of each other, yields exactly $\alpha=\alpha'$.

Another shortcoming of the method[11,14] is the unrealistically small statistical errors for the parameters which result from the *a priori* constraints inherent in it. By holding $E$ and $T_c$ at their fixed values, for instance, while $A'/A$ and $B'-A'B/A$ are least-squares adjusted to yield a fit to Eq. (7), the correlation between the parameters is not fully taken into account. This results in artificially small statistical errors. These small errors are one of the "advantages" claimed for the new technique over the "conventional" least-squares analysis. However, by imposing equivalent unrealistic constraints, identical errors are obtained by the "conventional" method.

### C. Results

We fitted our data to Eq. (2) with certain constraints, using the method described in Sec. IV B 1. Many of the constraints have been discussed in detail in Sec. IV C of Ref. 1. First, we assume that $E=E'$ so that the term $Et$ will be regular at $T_c$. Initially we permitted $T_c$ to be different from $T_c'$. This resulted in rather large errors for the parameters, mainly because data close to $T_c$, which primarily determine the transition temperature, had to be discarded due to the "rounding" (which was discussed in Sec. III and is shown in Fig. 4). The errors for $T_c$ and $T_c'$ were large enough to permit $T_c=T_c'$, and this constraint was imposed from then on.

#### 1. *Single-power-law analysis*

At first, we assumed that the contribution of singular higher-order correction terms is negligible, i.e., $D=D'=0$. We let the eight parameters $T_c$, $A$, $A'$, $B$, $B'$, $\alpha$, $\alpha'$, and $E$ be determined by a least-squares fit of the data over the range $t_{min} \le |t| \le 0.07$. The exponents $\alpha$ and $\alpha'$ obtained for sample I are shown in Fig. 6. It can be seen that $\alpha \neq \alpha'$ for $t_{min}=10^{-3}$; but Fig. 4 suggests that the analysis probably included data too close to the "rounded" region. For $t_{min}=2\times10^{-3}$ the difference between $\alpha$ and $\alpha'$ is only twice the sum of the standard errors and therefore we do not feel that there is evidence for a violation of the scaling relation $\alpha=\alpha'$. Next, the constraint $\alpha=\alpha'$ was imposed and the data were refitted for various $t_{min}$. The results are shown in Fig. 6 as solid squares. Although for $t_{min}=10^{-3}$ the uncertainties do not permit $\alpha=\alpha'$, the constraint was imposed anyway, in order to show the dependence of $\alpha=\alpha'$ upon $t_{min}$. The results reveal that $\alpha=\alpha'$ becomes more positive when $t_{min}$ becomes larger. Sample II exhibits similar behavior except that the values for $\alpha=\alpha'$ are slightly more negative. The values of $\alpha=\alpha'$ for the two samples are compared with each other in Fig. 7. Since the "rounding" of the transition in the case of EuO II is slightly worse, Fig. 7 might suggest that the variation of $\alpha=\alpha'$ is a result of sample inhomogeneities. This tends to be supported by the analysis made for $3\times10^{-3} \le |t| \le t_{max}$ for EuO I for various $t_{max}$. Those exponents $\alpha=\alpha'$ are shown in Fig. 8(a). They become more negative as $t_{max}$ is decreased. The corresponding values of $A/A'$ are shown in Fig. 8(b). On the basis of the above analysis we feel that $\alpha=\alpha'$ and $A/A'$ probably lie in the ranges $-0.04\pm0.03$ and $1.42\pm0.1$, respectively, if singular correction terms are omitted from the

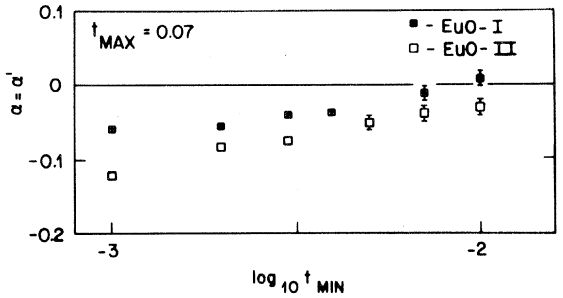


FIG. 7. Comparison of $\alpha=\alpha'$ obtained from a fit for the two samples as a function of $t_{min}$. When error bars are omitted, they are comparable to or smaller than the size of the point.

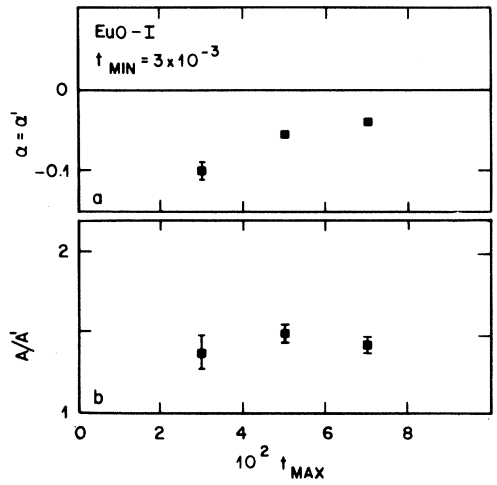


FIG. 8. $\alpha = \alpha'$ and $A/A'$ obtained from a fit of the data to Eq. (1) for various $t_{max}$. When error bars are omitted, they are comparable to or smaller than the size of the point.

analysis. The uncertainties are not standard errors, but are somewhat subjective estimates based upon the dependence of $\alpha = \alpha'$ and $A/A'$ on the range of $|t|$ over which data were used in the least-squares fit. Because of the range dependence of the parameters, the standard errors cannot be regarded as significant; but they were considerably smaller than the estimates given above.

So far we have considered the data analysis in terms of Eq. (2), with the constraints $D = D' = 0$, $\alpha = \alpha'$, $E = E'$, and $T_c = T_c'$. In addition, however, we also expect that

$$B = B'. \tag{9}$$

When $\alpha < 0$, it is easy to see that Eq. (9) must be valid; for in that case $C_p$ is finite at $T_c$, and $B \neq B'$ implies that $C_p$ is discontinuous. A discontinuity corresponds to an exponent of zero, and would therefore have to be regarded as the leading singularity. When $\alpha > 0$, the validity of Eq. (9) is not as obvious, and follows only from more detailed calculations based upon renormalization-group theory. For that case, the exponent of the first correction term to the asymptotically dominant contribution has been calculated. This has been done both by expansion in

$$\epsilon = 4 - d, \tag{10}$$

where $d$ is the dimensionality of the system,[6,15] and by numerical techniques based upon approximate recursion relations.[16] It is clear from these calculations that $x > \alpha$, where $x$ is the exponent of the first correction term as defined by our Eq. (2). For that case, all higher-order singular contributions to $C_p$ vanish at $T_c$, and Eq. (9) is valid.

In the analysis of our data for sample I and with $0.003 \leq |t| \leq 0.07$ we find $B - B' = 12.5 \pm 5.9$ J/mole K, where the uncertainty is the standard error. Although this result does not seem to quite agree with Eq. (9), restricting the range of data to $0.005 \leq |t| \leq 0.07$ yields errors which permit a continuous $C_p$. We consider it possible that the apparent failure by a small margin of Eq. (9) for the wider range of $|t|$ is associated with the inhomogeneities which cause the transition to be "rounded" near $T_c$.

In order to obtain the best estimates, based on a pure-power-law analysis, for $A/A'$ and $\alpha = \alpha'$, we imposed Eq. (9) as a constraint in the least-squares analysis of the data for EuO I. For $0.005 \leq |t| \leq 0.07$, this yielded

$$\alpha = \alpha' = -0.044 \pm 0.004, \tag{11a}$$

$$A/A' = 1.22 \pm 0.03, \tag{11b}$$

and

$$B = B' = 104 \pm 6 \text{ J/mole K}, \tag{11c}$$

where the uncertainties are standard errors. When only data over the very narrow range $0.01 \leq |t| \leq 0.07$ were used in this analysis, the same values were obtained for the parameters; but the standard errors increased by about a factor of 2. We therefore believe that the continuity constraint Eq. (9) has helped considerably to overcome the adverse effects due to sample inhomogeneities. Still somewhat subjectively, we choose $\alpha = \alpha' = -0.044 \pm 0.01$ and $A/A' = 1.22 \pm 0.06$ for the best estimates of the parameters pertinent to a pure-power-law analysis.

### 2. *Analysis with singular correction terms*

The analysis given above was based upon the assumption that higher-order singular terms do not contribute appreciably to $C_p$. Although this assumption has often been made in the analysis of measurements near critical points, there is theoretical evidence for the existence of singular correction terms in general,[6,15,16] and experimental evidence for their existence in specific cases.[17,18] We therefore included them in our analysis by permitting $D$ and $D'$ in Eq. (2) to be different from zero. We fitted the data for the range $0.003 \leq |t| \leq 0.07$ to Eq. (2) with various values of $x = x'$, but were unable to let $x = x'$ be least-squares adjusted since the strong correlation between the large number of parameters resulted in very slow convergence of the iterates. Even when $x = x'$ was chosen independently, for instance at a value equal to 0.5 as suggested by theory[15] and experiments on other systems,[17,18] there was still too much lattitude due to the high correlation of the remaining parameters for the analysis to yield meaningful results for $\alpha = \alpha'$ and $A/A'$. Therefore, we also imposed the

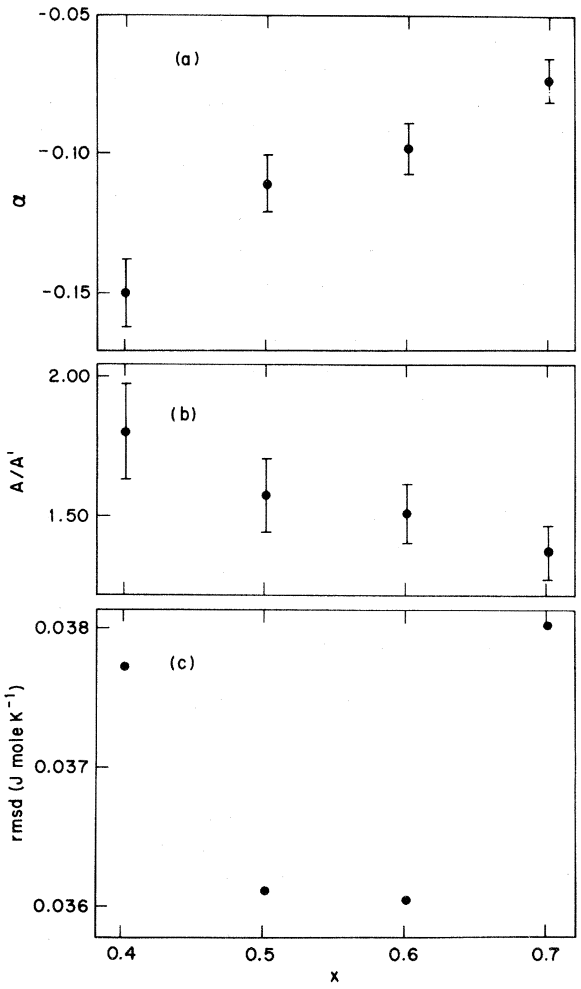


FIG. 9. Parameters $\alpha = \alpha'$ and $A/A'$, and the root-mean-square deviation, as obtained from a fit to Eq. (2) for various values of $x = x'$.

additional constraint Eq. (9). The fit with the eight parameters $T_c = T_c'$, $A$, $A'$, $\alpha = \alpha'$, $B = B'$, $E = E'$, $D$, and $D'$ least squares adjusted yielded $\alpha = \alpha' = -0.17 \pm 0.013$, $A/A' = 2.20 \pm 0.16$, $D = -0.91 \pm 0.16$, $D' = 0.73 \pm 0.28$, and $B = B' = 57.9 \pm 1.3$ J mole$^{-1}$K$^{-1}$. However, it also gave the unrealistic value $E = E' = -12.6 \pm 11.3$ J mole$^{-1}$K$^{-1}$. We therefore permitted $E = E'$ to vary only over a physically reasonable range, and initially fixed this parameter at 32 J mole$^{-1}$K$^{-1}$. The results for $\alpha = \alpha'$ and $A/A'$ which were obtained with this choice for $E = E'$ are shown in Fig. 9 as a function of $x$. Also shown is the root-mean-square deviation. Clearly, the best fit is obtained when $x = x'$ is between 0.5 and 0.6, with $\alpha = \alpha' \cong -0.10$ and $A/A' \cong 1.50$. Similar analyses, but with the reasonable guesses 22 and 42 J mole$^{-1}$K$^{-1}$ for the extreme values of $E = E'$, yielded essentially the same result for $\alpha = \alpha'$, $A/A'$, and $x = x'$. The only difference was that the values of the root-mean-square deviation were shifted downward or upward depending on the choice of $E = E'$. On the basis of this analysis, we estimate, somewhat subjectively, that

$$\alpha = \alpha' = -0.10 \pm 0.05, \tag{12a}$$

$$A/A' = 1.51 \pm 0.2, \tag{12b}$$

$$B = B' = 69 \text{ J mole}^{-1}K^{-1}, \tag{12c}$$

$$x = x' = 0.56 \pm 0.20, \tag{12d}$$

$$D = -0.2 \pm 0.2, \tag{12e}$$

$$D' = -0.1 \pm 0.1. \tag{12f}$$

Last, we remark that the inclusion of the correction terms in the analysis did not substantially improve the quality of the fit. To illustrate this we quote the root-mean-square deviations of the data from the functions. For the data with $0.003 \leq |t| \leq 0.07$, they were (in the units J mole$^{-1}$K$^{-1}$) 0.0378 for the pure power law with $B$ permitted to be different from $B'$, 0.0410 for the pure power law with the constraint $B = B'$, 0.0359 when correction terms were included and the constraints $B = B'$ and $E = E' = 32$ J mole$^{-1}$K$^{-1}$ were imposed and 0.0319 when correction terms were included and $E = E'$ had their least-squares adjusted value $-12.6$ J mole$^{-1}$ K$^{-1}$.

## V. COMPARISON WITH OTHER EXPERIMENTS

### A. Specific heat

The specific heat of EuO was measured previously by Teaney,[19] but his sample was less homogeneous and his data exhibited much more severe rounding than ours. No exponents were reported.

More recent measurements were made by Salamon[11] on a sample that was cut from the same single crystal that yielded one of ours (see Sec. II). Salamon reported two sets of parameters for pure power laws, one each for an "inner region" ($|t| < 0.02$) and an "outer region" ($0.02 < |t| < 0.2$). His exponents and amplitude ratios were

$$\alpha = \alpha' = -0.026; A/A' = 2.03$$

for the inner region, and

$$\alpha = \alpha' = -0.09; A/A' = 1.00$$

for the outer region. The exponents for the inner region seem to be in agreement with ours, but the amplitude ratio is different. The exponents for the outer region are in agreement with the theoret-

TABLE II. Parameters obtained by least-squares fits of the data in Ref. 11 to Eq. (1). The resulting units of $C_p$ are J mole$^{-1}$ K$^{-1}$.

| Parameter | $0.003 \leq \lvert t \rvert \leq 0.2$ | $0.003 \leq \lvert t \rvert \leq 0.07$ |
|---|---|---|
| $\alpha = \alpha'$ | −0.0320 | 0.0220 |
| $A$ | 4.936 | 4.570 |
| $A'$ | 2.911 | 1.895 |
| $B$ | 167.90 | −194.89 |
| $B'$ | 125.26 | −48.935 |
| $E = E'$ | 36.662 | 48.355 |
| $T_c$(K) | 69.396 | 69.318 |

ical[2-6] and experimental[1,8,14,20] values for Heisenberg systems, but the value of $A/A'$ is too low. Salamon also observed that his crossover effect is quite sharp and occurs at a rather well-defined value of $\lvert t \rvert$. This seems rather surprising; for true crossover effects would be expected to occur gradually, perhaps over several decades in $\lvert t \rvert$. Because of these unusual results, and because of the reservation about Salamon's method of analysis which we already expressed in Sec. IV B 2, we shall reexamine the data.

In his analysis, Salamon chose the values $T_c = 69.330$ K and $E = 2.9 \times R = 24.1$ J mole$^{-1}$ K$^{-1}$. Our least-squares fit to a pure power law of his data in the range $3 \times 10^{-3} \leq \lvert t \rvert \leq 0.2$ yielded $T_c = 69.396$ K and $E = 36.66$ J mole$^{-1}$ K$^{-1}$. These latter values compare reasonably well with those for our samples, as quoted, for instance, in Table I. When these two coefficients are used to obtain Salamon's graph of $\tilde{C}_p^-$ vs $\tilde{C}_p^+$ [see Sec. IV B 2 and Eq. (6)], the crossover effect is removed entirely. This is shown by the open circles in Fig. 10. The solid circles in the figure correspond to Salamon's analysis. The good fit by a straight line to the open circles demonstrates that there is no evidence for crossover behavior obtainable from the data. The measurements yield a unique $A/A' = 1.70$ and $\alpha = \alpha' = -0.032$. The value for the exponents is in agreement with the value obtained by us, while $A/A'$ is somewhat high. However, a detailed comparison should be based upon data analyses restricted to the same range of $\lvert t \rvert$.

In order to provide a more direct comparison with Salamon's results, we fitted his data to Eq. (1) with the constraints $E = E'$, $\alpha = \alpha'$, and $T_c = T_c'$, and obtained the parameters listed in Table II. The results are not in particularly good agreement with ours. The difference between the two sets of data also is readily apparent if they are compared directly on linear scales. We have done this in Fig. 11. Although Salamon's specific-heat data are higher than ours, they may be reduced by a multiplicative factor because the original data were relative and had to be normalized to other absolute measurements. This normalization will not affect $\alpha = \alpha'$ or $A/A'$. If the two sets of data are made to agree at 72.6 K, agreement is poor over nearly the entire range of $T$. However, if the data are normalized at 65 K, good agreement is achieved below the transition, while above $T_c$, Salamon's data are still higher than ours, for instance by 0.8 J mole$^{-1}$ K$^{-1}$ at 71 K.

### B. Other experiments

The properties of EuO near the critical temperature have been investigated by various methods,[21-24] and the results are summarized in Table III. All these results are based upon analyses in terms of pure power laws, often including data for rather large values of $\lvert t \rvert$, and should therefore be viewed with some reservation. The values reported for $\beta$ and [25]$\delta$ are all consistent with each other. The value $\gamma \cong 1.4$ obtained by neutron diffraction differs

TABLE III. Critical exponents for EuO.

| Method | Vibrating-coil magnetometer | | Neutron diffraction | Faraday magnetic balance | High-temperature-series short-range interaction | | $\epsilon$-expansion short-range interaction | $\epsilon$-expansion dipolar interaction |
|---|---|---|---|---|---|---|---|---|
| Ref. | 21 | 22 | 23 | 24 | 2 | 3 | 5,6 | 6 |
| $\beta$ | $0.368 \pm 0.005$ | $0.385^{+0.008}_{-0.028}$ | $0.367 \pm 0.008$ | ... | $0.373 \pm 0.014$[a] | ... | 0.380[a] | 0.381[a] |
| $\gamma$ | $1.29 \pm 0.01$ | $1.315 \pm 0.015$ | $1.396 \pm 0.030$ | 1.29 | $1.405 \pm 0.020$ | $1.375^{+0.02}_{-0.01}$ | 1.365[a] | 1.372[a] |
| $\delta$ | $4.46 \pm 0.1$ | $4.35 \pm 0.1$ | ... | ... | $4.8 \pm 0.4$[a] | $4.8 \pm 0.3$[a] | 4.458[a] | 4.454[a] |
| $\nu$ | ... | ... | $0.690 \pm 0.023$ | ... | $0.717 \pm 0.007$ | $0.703^{+0.01}_{-0.005}$ | 0.688 | 0.692 |
| $\alpha$ | ... | ... | ... | ... | $-0.14 \pm 0.06$ | $-0.09 \pm 0.04$[a] | −0.125[a] | −0.135[a] |
| $\alpha = 2 - 2\beta - \gamma$ | $-0.026 \pm 0.015$ | $-0.085^{+0.07}_{-0.03}$ | $-0.13 \pm 0.03$ | ... | | | | |
| $\alpha = 2 - \beta(\delta + 1)$ | $-0.01 \pm 0.04$ | $-0.06^{+0.19}_{-0.08}$ | ... | ... | | | | |
| $\alpha = 2 - 3\nu$ | ... | ... | $-0.07 \pm 0.07$ | ... | | | | |

[a]Scaling relations were used.

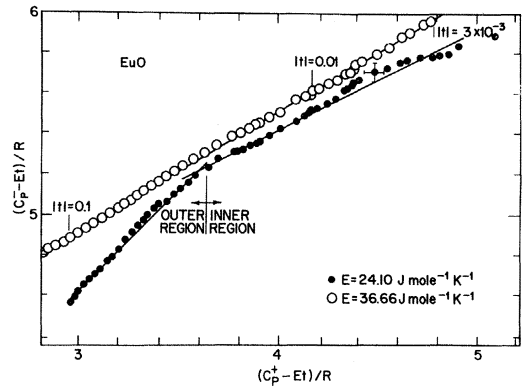


FIG. 10. Examination of the "crossover" effect reported in Ref. 11. The effect, which manifests itself by departures from linearity in this graph, can be removed by the right choice of $E$ and $T_c$.

considerably from the others; but it is in agreement with theoretical predictions for Heisenberg systems.[2-6] However, a value of $\gamma$ near 1.30 is consistent with our $\alpha = -0.04$, the experimental $\beta$, and scaling.[25] This is shown in the seventh row of Table III. Scaling and the larger value for $\gamma$ obtained by neutron diffraction, together with the experimental $\beta$, would require $\alpha \cong -0.13$. This would be allowed by our data only if singular correction terms contribute appreciably to $C_p$.

### C. Other Heisenberg systems

We saw in Sec. IV C that the value of $\alpha = \alpha'$ for EuO which is obtained with the assumption that higher-order correction terms are negligible differs significantly from the values obtained with the same assumption for $RbMnF_3$,[1,8] Ni,[14,20] and Fe.[14,26] For these materials the original authors obtained for pure-power-law exponents $-0.14$, $-0.10$, and $-0.12$, respectively. A universal value of $\alpha = \alpha'$ in this range, and of $A/A'$ near 1.4, can be obtained only if significant contributions to the specific heat of EuO from singular correction terms exist and are considered in the data analysis.

TABLE IV. Exponents and amplitude ratios for several materials with $n = 3$. All analyses are based upon a pure power law with a continuous $C_p$ at $T_c$, on a regular contribution which is linear in $t$, on the assumption that $\alpha = \alpha'$, and on the range $0.005 \leq |t| \leq 0.07$. The errors are standard errors, and do not include possible systematic uncertainties.

| Material | $\alpha = \alpha'$ | $A/A'$ | Ref. |
|---|---|---|---|
| EuO | $-0.044 \pm 0.004$ | $1.22 \pm 0.03$ | This work |
| $RbMnF_3$ | $-0.137 \pm 0.004$ | $1.463 \pm 0.017$ | 1 |
| Ni | $-0.089 \pm 0.004$ | $1.382 \pm 0.022$ | 20 |
| Fe | $-0.096 \pm 0.023$ | $1.30 \pm 0.08$ | 14 |

We consider it worth noting that in the case of EuO appreciable singular correction terms have to be invoked in order to obtain universal parameters which agree with theory (see also Sec. VI below), whereas in all other systems with $n = 3$ which have been examined these terms could be much smaller. It might be conjectured that the apparently greater importance of the singular corrections for EuO is in fact attributable to a more limited range of $t$ over which usable data exist. For that reason we have reanalyzed[27] the measurements for $RbMnF_3$,[1] Ni,[20] and Fe,[14] using the range $0.005 \leq |t| \leq 0.07$ with a pure power law and the continuity constraint. These results should be directly comparable to Eq. (11). They are summarized in Table IV. It can be seen that even completely equivalent pure-power-law analyses, restricted to a range of $|t|$ where sample inhomogeneities are believed unimportant, yield results for EuO which differ significantly from those for other systems with $n = 3$. A more detailed comparison will be given elsewhere.[27]

## VI. COMPARISON WITH THEORY

Europium oxide is an isotropic ferromagnet.[28] Its critical behavior sufficiently near $T_c$ should thus be characteristic of systems with dipolar interactions and three degrees of freedom ($n = 3$) for the order parameter.[5] Further away from $T_c$, the critical behavior should be that appropriate to isotropic systems with short-range forces and $n = 3$. A crossover from isotropic short-range to dipolar behavior has been predicted to occur near $t \simeq 10^{-2}$.[5,6] For the dipolar case, the specific-heat exponent has been calculated for[6] $n = d$ (which for $d = 3$ should pertain here), and to second order in $\epsilon$ is given by

$$\alpha = -\epsilon/34 - (6223/58956)\epsilon^2 + \cdots$$

$$\cong -0.135.$$

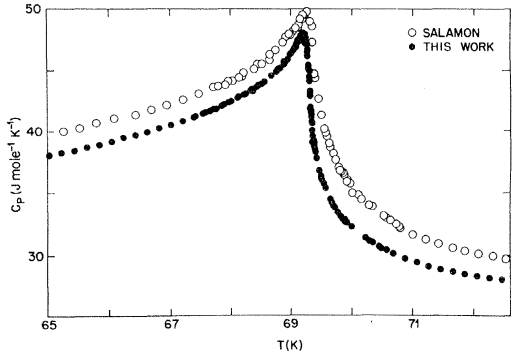


FIG. 11. Comparison of the specific heat of EuO I with the measurements reported in Ref. 11.

Although this result differs in principle from the prediction[5,6]

$$\alpha = -\epsilon^2/8 + \cdots \cong -0.125$$

which pertains to $n = d$ and isotropic short-range interactions, it is not reasonable to hope to distinguish between these cases on the basis of the type of experimental data available for phase transitions in solid systems. There is, of course, even less hope of seeing the crossover from short-range-force behavior far from $T_c$ to dipolar behavior near $T_c$ which in principle would be expected from theory. Although the expansion to second order in $\epsilon$ does not necessarily yield accurate numerical values for the exponents, the qualitative prediction of only a small difference in $\alpha$ between systems with dipolar and isotropic short-range interactions can probably be regarded as reliable. The actual value of $\alpha$ for the short-range-force case is supported by high-temperature series expansions,[2] as well as by experiment.[1,14,20,26,27]

A fit of our data to a pure power law (Sec. IV C 1) yielded $\alpha = \alpha' = -0.04$, which is inconsistent with the theoretical prediction. However, when we invoked singular correction terms in our data analysis (Sec. IV C 2), we obtained $\alpha = \alpha' = -0.10 \pm 0.05$, in agreement with theory. We therefore conclude that consistency between theory and experiment is possible only if singular correction terms make an appreciable contribution to $C_p$. Our correction exponent $x = 0.56 \pm 0.20$ is also in good agreement with theoretical estimates[6]; but again the data are not good enough to distinguish between the dipolar and the short range prediction.

## VII. SUMMARY

In this paper we presented heat-capacity measurements for EuO near the ferromagnetic transition temperature 69.3 K. Two sets of results obtained for two different samples over the temperature range 64–74 K were found to be consistent with each other. Although the transition region exhibited some "rounding," it was possible to fit the data to a pure power law. This yielded the parameters $\alpha = \alpha' = -0.044 \pm 0.01$ and $A/A' = 1.22 \pm 0.06$. The value for the exponents is in disagreement with the theoretical prediction for Heisenberg ferromagnets with dipolar interactions. However, when singular correction terms are included in the data analysis, then $\alpha = \alpha'$ becomes more negative and its value less certain. The result is then consistent with the theoretical value.

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[28]The ratio $u$ between the anisotropy energy [J. F. Dillon, Jr. and C. E. Olsen, Phys. Rev. 135, A434 (1964)] and the exchange energy [L. Passell, O. W. Dietrich, and J. Als-Nielsen, in *AIP Conference Proceedings No.* 5, *Magnetism and Magnetic Materials*-1971, edited by C. D. Graham, Jr. and J. J. Rhyne (AIP, New York, 1972), p. 1251] for EuO is of the order of $10^{-3}$. However, because of the symmetry of the crystal the anisotropy is cubic. Crossover to behavior characteristic of the cubic fixed point for systems which exhibit isotropic short-range Heisenberg behavior far from $T_c$ has been examined theoretically by Aharony [Phys. Rev. B 8, 4270 (1973)]. This crossover is expected to occur when $|t| \approx u^{1/\varphi}$, where $1/\varphi$ was estimated to be about equal to 18. The cubic behavior therefore is not expected to dominate until $|t|$ is much smaller than the experimentally accessible range. Furthermore, the cubic exponents, although different in principle, were estimated by Aharony to be numerically very similar to the short-range Heisenberg 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 (Ref. 6). They find $1/\varphi \cong 5.6$ for this case. Although this value is much smaller than the corresponding one for crossover from the isotropic short-range fixed point, it is still much too large to permit experimental measurements at values of $t$ where cubic behavior should dominate. The exponents corresponding to or the existence of a cubic fixed point in the presence of dipolar forces apparently has not yet been investigated, however (Ref. 6).