Heat capacity of EuS near the ferromagnetic transition

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We report precision measurements of the heat capacity of EuS over the temperature range $1.5 \leq T \leq 33$K, with particular emphasis on the region near the Curie temperature $T_C = 16.51$ K. Our data yield $\alpha = \alpha' = -0.13 \pm 0.02$ and $A/A' = 1.54 \pm 0.1$ for the exponents and the amplitude ratio of the specific heat. These results agree with those obtained previously for the isotropic antiferromagnet $RbMnF_3$, and with theoretical predictions. The previously reported anomalous behavior of EuO is discussed in terms of effects associated with crossover from isotropic short-range force to dipolar behavior.

I. INTRODUCTION

It is expected from the renormalization-group (RG) theory of phase transitions that critical exponents, and certain dimensionless combinations of amplitudes, which describe the singularities in various properties near critical points should be universal in the sense that they depend only upon a small number of very general symmetry properties of the system.[1] In particular, isotropic Heisenberg antiferromagnets should exhibit the same critical behavior as isotropic Heisenberg ferromagnets, provided that in each case the interactions consist only of isotropic short-range forces. However, in most magnetic materials there are also contributions to the interactions from dipolar forces. For the behavior of antiferromagnets near the Néel temperature T_N, it has been predicted on the basis of the RG theory that dipolar interactions do not modify the critical behavior.[2,3] Contrary to this, the behavior of isotropic ferromagnets near the Curie temperature T_C is expected to be altered by the dipolar forces.[2,4-7] However, estimates of the exponents based on the $\epsilon \equiv 4 - d$ expansion[5] (d is the dimensionality of the system) indicate that the parameters characterizing the short-range-force and the dipolar systems, although different in principle, are numerically so similar that experiment probably could not distinguish between them. Nonetheless, specific-heat measurements for the isotropic Heisenberg ferromagnet EuO,[8,9] for which dipolar interactions are expected to be appreciable,[2,3] have yielded values for the exponent α and the amplitude ratio A/A' which differ significantly from the values obtained for the isotropic antiferromagnet $RbMnF_3$.[8,10] Because of this apparent disagreement between experiment and the ϵ-expansion estimates of α and A/A' for the two systems, it is desirable to have specific heat measurements also for the isotropic Heisenberg ferromagnet EuS. For EuS, dipolar forces are expected to be more appreciable than for EuO because the exchange interactions are weaker.[2,3] Therefore it is expected that the behavior characteristic of the pure dipolar system should be observable further away from T_C. Although there exist previous experimental results for C_p of EuS,[11,12] these data have not been published in detail and do not yield the values of α and A/A' which are needed for comparing different systems. We report in this paper new measurements of the specific heat of EuS. On the basis of these data, we find values of α and A/A' very similar to those reported previously for $RbMnF_3$.[8,10] Therefore, it appears that agreement between experiment and the ϵ-expansion estimates[5] exists provided that the dipolar forces contribute sufficiently to the interactions.

In view of the results for $RbMnF_3$ and EuS, the different behavior of EuO must be regarded as anomalous. A possible explanation of the anomaly may be found by assuming that the dipolar forces in EuO are not sufficiently strong to yield pure dipolar behavior in the experimentally accessible temperature range, but not so weak as to be negligible. In that case, the experiment would yield *effective* exponents and amplitude ratios characteristic of a crossover region which need not correspond to the behavior of either the pure dipolar or the pure short-range-force system. This interpretation of the results would lead to effective parameters $\overline{\alpha}$ and $\overline{A}/\overline{A}'$ which vary in a nonmonotonic manner as one passes from one pure system through the crossover region to the other pure system. In the absence of a detailed theoretical prediction, one might have expected a monotonic variation because it is simpler. In that case, crossover effects between isotropic short-range-force and dipolar systems would have been expected to be unobservable because the pure systems are predicted to have very similar parameters. However, recent detailed calculations[13-15] of the scaling function have demonstrated that effective exponents vary in a *non*monotonic manner and by amounts comparable to the difference between the experimental parameters for EuO on the one hand and EuS and $RbMnF_3$ on the other. This qualitative agreement between experiment and detailed theoretical calculations tends to lend support to the explanation of the anomalous behavior of EuO in terms of crossover effects.

In this paper we report the results of specific-heat measurements for three different samples of EuS. The samples were of varying quality, and the measurements permitted us to demonstrate that our conclusions are not influenced by sample imperfections. For one of the samples, measurements were made over the wide temperature range $1.4 \leq T \leq 33$ K. The low-temperature data may be compared with previous results by others[16,17] and yield useful information about the exchange interactions.

In Sec. II, we describe the experimental apparatus. This section is short and contains only the modifications made to the apparatus described in detail in a previous publication.[10] The results are reported in Sec. III, and in Sec. IV the data are analyzed. In Sec. V our results are compared with other experiments. Section VI summarizes our conclusions.

II. APPARATUS AND SAMPLE PREPARATION

A. Apparatus

The calorimeter used for the work presented in this paper was described previously in Sec. II A of Ref. 10. Only the sample holder and associated thermometer and heater were changed in order to make possible the measurements in the temperature range of the present work.

The sample holder consisted of a 1.75-cm-diam $\times$ 0.030-cm-thick circular copper disk to which a 11-cm-long $\times$ 0.10-cm-diam copper wire was soldered. It was suspended in the calorimeter by nylon strings, and could be thermally attached to the isothermal shield by closing the jaws of a mechanical heat switch upon the 0.10-cm-diam copper wire. A Honeywell germanium thermometer was attached to the bottom of the sample holder. Two reference resistors made of 0.0038-cm-diam Karma wire and with a resitance of nominally 100 and 3500 Ω, respectively were wound noninductively around the copper wire. Either of them could be used in conjunction with the thermometer. The availability of the two reference resistors made it possible to cover the temperature ranges $1.3 \lesssim T \lesssim 6$K and $4.5 \lesssim T \lesssim 35$K without warming up the calorimeter to room temperature. An ac bridge technique was used for the temperature measurement.[18] The bridge was calibrated against another calibrated germanium reference thermometer which was located on the isothermal platform and whose resistance could be measured by a four-lead dc potentiometric technique. The temperature scale of the reference thermometer was the same as that used previously for measurements of the heat capacity of copper[19] and silver.[20]

Also wound noninductively on the copper wire were two heaters of nominally 5800 and 2900 Ω resistance. They were made of 0.0038-cm-diam Karma wire. One of them could be used when necessary to balance any heat loss to the isothermal shield, and the other was used for the heat-capacity measurement. The latter had a resistance whose temperature dependence could be described within 0.02% by the function

$$R_H = 5823.2(1 + 1.54 \times 10^{-3} T^{-2} + 5.3 \times 10^{-6} T) \quad .$$

The weight of the sample holder was 3.54 g, and its heat capacities at 10 and 20 K were 1.7×10^{-3} J/K and 7.6×10^{-2} J/K, respectively.

B. Sample preparation

The EuS samples were prepared by first reacting 99.9% pure resublimed Eu and 99.9999% pure sulfur in a quartz tube mounted in a two-zone furnace. Reaction with the quartz envelope was avoided by controlling the temperature of the sulfur side of the furnace. The synthesized EuS was then sealed in a $\frac{1}{2}$-in.-diam heavy wall (0.050-in.-thick) tungsten Bridgman-type crucible and rf heated in an argon atmosphere to 2600 ℃. Gradient freeze cooling, beginning at the pointed lower end of the crucible, was then applied at a rate of 20°/h. This produced both melt-grown and vapor-grown crystals of EuS. The vapor-grown crystals were found in the space above the melt, growing inwardly from the wall, and also on the surface of the melt.

Three EuS samples were used. EuS-I was a single piece consisting of a few single crystals grown from the melt, EuS-II was a single crystal grown from the vapor, and EuS-III was a single crystal grown from the melt. EuS-I, -II, and -III weighed 7.706, 2.332, and 2.327 g, respectively. The molecular weight of 184.024 g was used to convert the data to a molar basis.

III. RESULTS

The results for EuS-I are shown in Fig. 1 on a linear scale, and the results for EuS-II in Fig. 2 on a logarithmic scale. For the sake of clarity, the remainder of the data were excluded from these figures. The results for the three samples differ significantly from each other only in the transition region, and agree within experimental scatter both at higher and lower temperatures. The data near T_C are shown for all three samples in Fig. 3. EuS-II exhibits the sharpest transition, while EuS-III, somewhat surprisingly, the most rounded. The transition temperatures of the

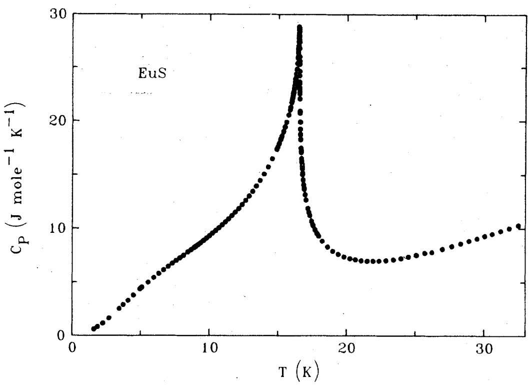

FIG. 1. Specific heat of EuS-I as a function of the temperature.

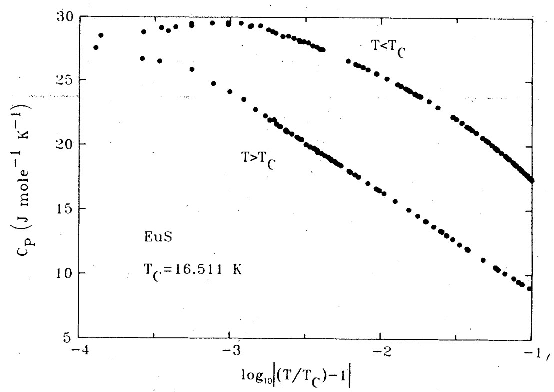

FIG. 2. Specific heat of EuS-II as a function of $\log_{10}|T/T_C-1|$.

three samples also differed from each other, and estimates are given in Fig. 4. Figure 4 shows the data near T_C of all three samples on a logarithmic scale.

The data of all three samples were fitted to the function

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

for $T > T_C$, and to the same function with primed parameters for $T < T_C$. Here $t \equiv (T-T_C)/T_C$. No significant increase in the standard error was realized when the constraints $E \equiv E'$ and $T \equiv T_C'$ were imposed. Furthermore when the "rounded" region was excluded from the analysis, the constraint $\alpha \equiv \alpha'$ could be imposed too. The results for EuS-II with those constraints are listed in Table I, column 1. The additional constraint $B \equiv B'$ would require more data to be excluded from the least-squares fit, and will be discussed in Sec. IV.

The heat-capacity data of EuS-I in the range 1.4–5 K are shown in Fig. 5. They agree well with previously published data[16,17] obtained with powdered samples in this range.

All the data are available in numerical form elsewhere.[21]

IV. ANALYSIS

A. Low-temperature data

At sufficiently low temperatures, the specific heat of EuS can be attributed primarily to the thermal excitation of spin waves.[22] For a purely exchange-coupled ferromagnet, this contribution is expected to have the temperature dependence given by

TABLE I. Parameters for Eq. (3) for EuS-II. The resulting units of C_p and σ are J mole^{-1}K^{-1}. The uncertainties are standard errors and do not reflect possible systematic errors.

Parameter	Without correction terms		With correction terms
t_{min} ^{a}	0.002	0.005	0.003
t_{max} ^{a}	0.07	0.07	0.07
$\alpha=\alpha'$	-0.133 ± 0.002	-0.131 ± 0.003	-0.124 ± 0.016
A/A'	1.392 ± 0.018	1.536 ± 0.018	1.540 ± 0.091
A'	4.467 ± 0.053	4.192 ± 0.022	3.981 ± 0.25
$B'-B$	2.47 ± 0.34	0^b	0^b
B	41.35 ± 0.48	43.08 ± 0.58	43.93 ± 2.44
x	...	...	0.5^b
D	0^b	0^b	-0.042 ± 0.12
D'	0^b	0^b	0.135 ± 0.081
T_C (K)	16.5183 ± 0.0007	16.5112 ± 0.0006	16.5154 ± 0.0010
E	18.20 ± 0.60	21.68 ± 0.61	15.06 ± 5.23
σ	0.0220	0.0201	0.0206

^{a}Defined with respect to $T_0 = 16.515$ K (see text). ^{b}Fixed.

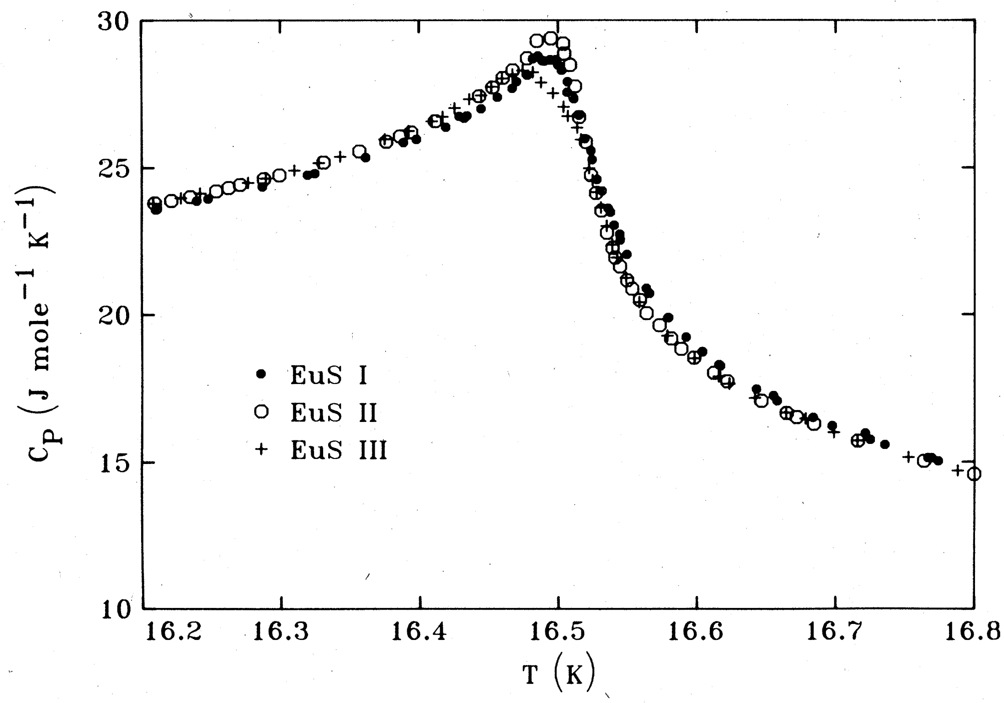

FIG. 3. Specific heat of all three samples of EuS near T_C as a function of the temperature.

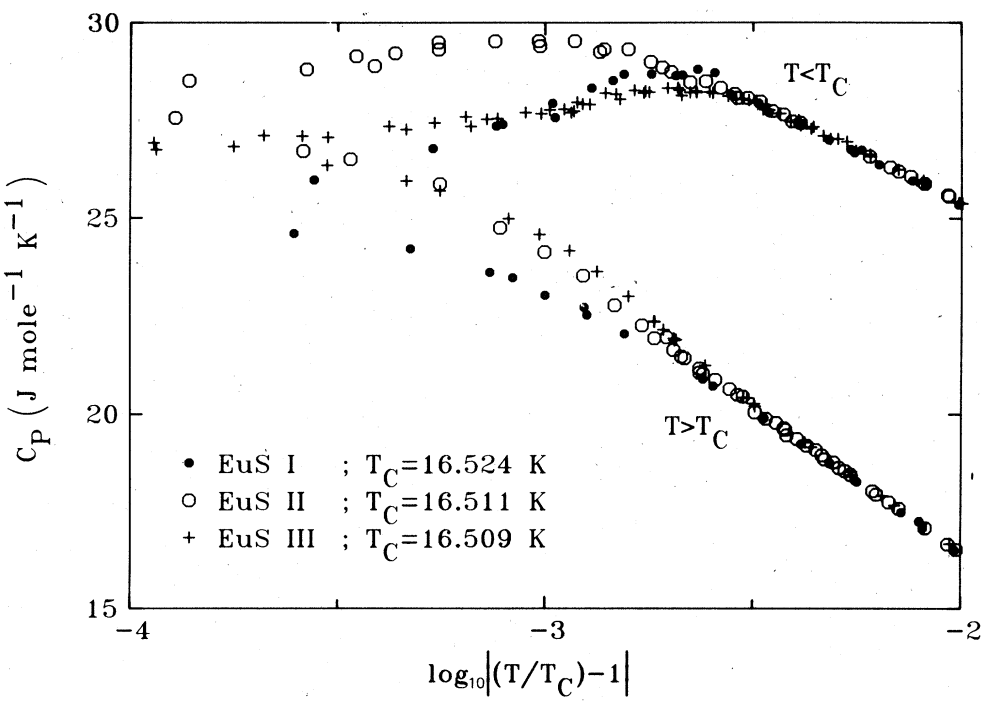

FIG. 4. Specific heat of all three samples of EuS near T_C as a function of $\log_{10}|T/T_C-1|$, with T_C obtained separately for each sample from a least-squares fit.

$$C_p/R = aT^{3/2} + bT^{5/2} + \cdots \quad . \tag{2}$$

Equation (2) suggests that $C_p T^{-3/2}$ should be well approximated by a linear function of T at small T. We therefore plotted $C_p/RT^{3/2}$ vs T in Fig. 5. Also shown in Fig. 5 are the data obtained by Passenheim *et al.*[16] and by Dietrich *et al.*[17] The agreement of our data with those of Passenheim *et al.* is remarkably good. The small difference in the range 1.8–2.8 K between our data and those of Dietric *et al.* does not exceed 4% of C_p, and probably is within reasonable estimates of the combined systematic errors in the two sets of data.

The previous measurements[16,17] in this temperature range have been analyzed recently[17] in great detail; and the good agreement particularly between our data and those of Passenheim *et al.*[16] makes a detailed analysis of our data superfluous since the results for the exchange constants would be virtually identical. We do remark, however, that our data indicate that the values of the exchange constants derived from the data of Passenheim *et al.*[16] and given in Eq. (21) of Ref. 17 should be regarded as more accurate than those derived from the data of Dietrich *et al.*[17] and given in Eq. (22) of Ref. 17. Coincidentally, the values given by Eq. (21) of Ref. 17 also agree exactly with the best values estimated by Dietrich *et al.* from the calorimetric and neutron evidence together and given by them in their Table IV.[17]

B. Data near the phase transition

1. Method

We have discussed our method of analysis of specific-heat data near phase transitions in several previous publications[9,10,23] to which we refer the reader who is interested in the details. We fitted the results over various temperature ranges to the function

$$C_p = (A/\alpha)|t|^{-\alpha}(1 + D|t|^x) + B + Et \tag{3}$$

for $T > T_C$, and to the same function with primed coefficients for $T < T_C$. Equation (3) reduces to Eq. (1) when $D = D' = 0$.

For the purpose of eliminating data outside a desired temperature range, it is convenient to define a fixed reference temperature T_0 which is approximately equal to T_C. In terms of T_0, we define the parameters

$$t_{\min} \equiv |T' - T_0|/T_0 \tag{4}$$

and

$$t_{\max} \equiv |T'' - T_0|/T_0 \ , \tag{5}$$

and analyze data for which $t_{\min} \leq t \leq t_{\max}$ for various values of $t_{\min}$ and $t_{\max}$.

2. Results

a. Test of the scaling prediction $\alpha = \alpha'$. Initially, we fitted the data to pure power laws by setting

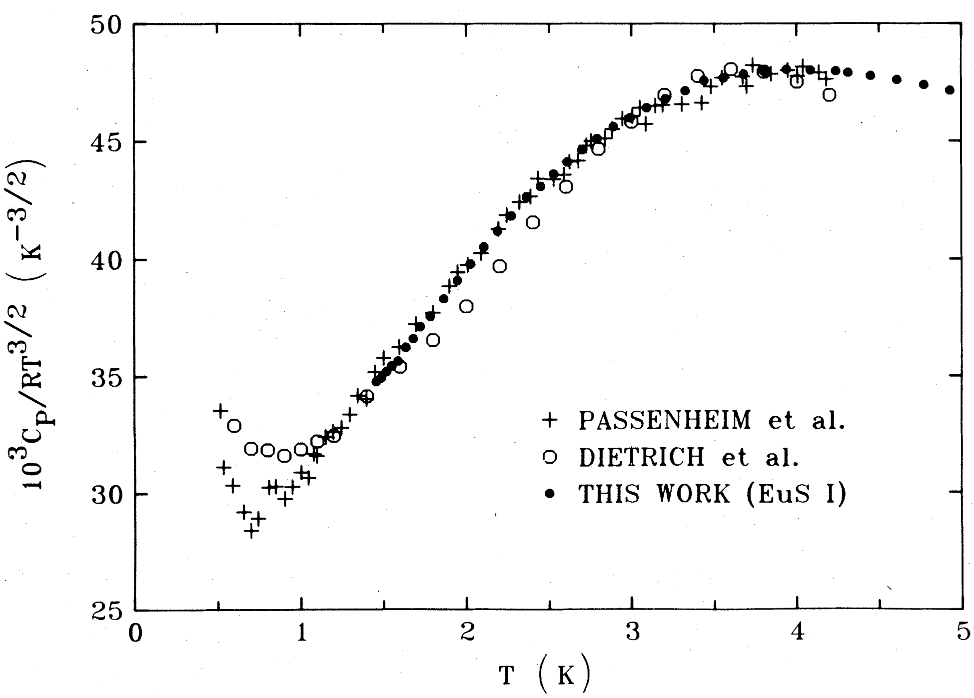

FIG. 5. Comparison of the specific heat of EuS-I at low temperature with the data of Passenheim *et al.* (Ref. 16), and with the smoothed results of Dietrich *et al.* (Ref. 17).

$D = D' = 0$. The data always permitted us to also impose the constraints $T_C = T_C'$ and $E = E'$ without significant increase in the deviations from the fit. The constraint $E = E'$ was imposed in order to assure that the term Et represents the temperature dependence of any regular contribution to C_p.

In order to test the validity of the scaling law[24] $\alpha = \alpha'$, we initially permitted α to be different from α' and fitted the data for EuS-II with $t_{max} = 0.07$ for various t_{miin}. The results are shown in Fig. 6. In each case, the standard errors for α and α' which are indicated in the figure overlap and thus permit $\alpha = \alpha'$, consistent with the scaling prediction. Similar results were obtained for the other two samples. Since the data permits $\alpha = \alpha'$, this additional constraint was imposed in all further analyses.

b. Test of the RG prediction $B = B'$. The next point of interest is to see whether the data also permit the constraint $B = B'$ which is predicted by the RG theory.[25] First, we will examine this question within the context of a pure power law analysis (i.e., by retaining the constraint $D = D' = 0$). Of course we must recognize that a failue of the data to permit equality between B and B' may actually be an indication for the necessity of including the confluent singular terms $D|t|^x$ and $D'|t|^{x'}$ in the analysis. Recently, this point has been demonstrated by a detailed analysis of thermal-expansion measurements near the superfluid transition in liquid ^{4}He.[26] We show as open circles in Fig. 7 the results of an analysis for EuO-II with $t_{max} = 0.07$ for several values of t_{min}. It is clear that the data for $B' - B$ differ significantly from zero unless only the measurements rather far from T_C are

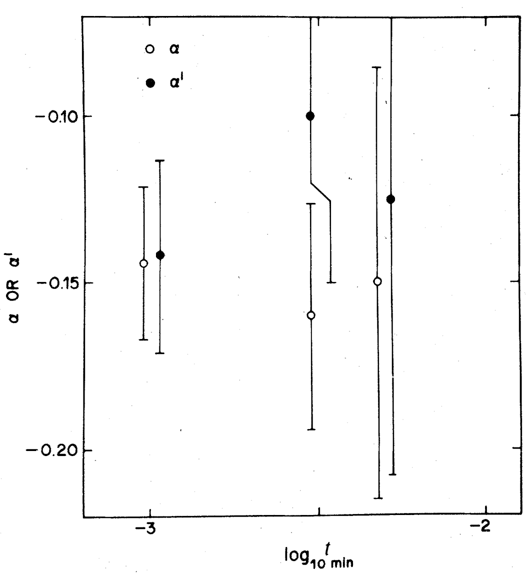

FIG. 6. Exponents α and α' obtained from fits of the specific heat of EuS-II to Eq. (3) with the constraints $T_C = T_C'$, $E = E'$, and $D = D' = 0$, for various values of t_{min} with $t_{max} = 0.07$.

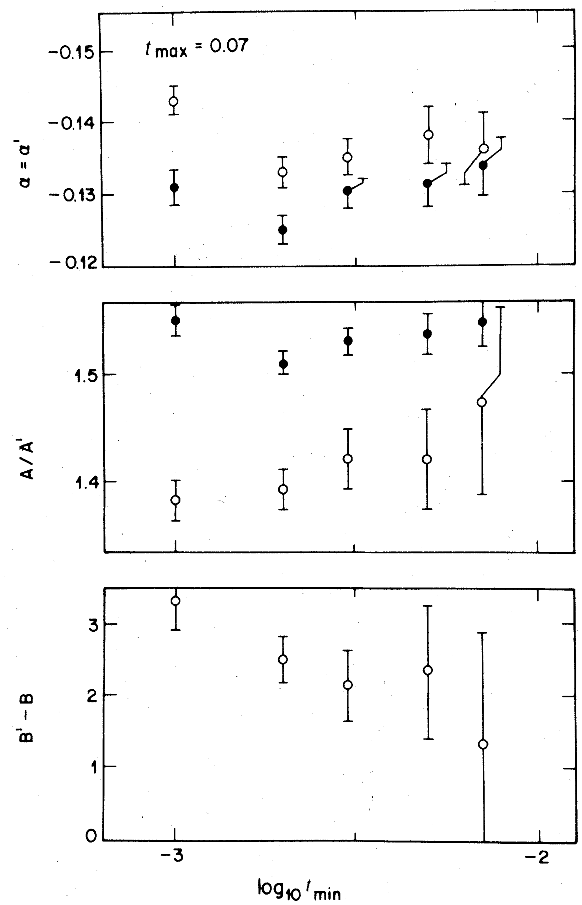

FIG. 7. Parameters $\alpha = \alpha'$ and A/A' obtained from fits of the specific heat of EuS-II to Eq. (3) with the constraints $T_C = T_C'$, $\alpha = \alpha'$, $E = E'$, and $D = D' = 0$. Open circles: B allowed to be different from B'. Solid circles: B constrained to be equal to B'.

analyzed. We do not regard this observation as being in conflict with the RG theory, however. Instead, we take it as an indication for significant contributions from confluent singular terms.

c. Best pure power-law values for $\alpha = \alpha'$ and A/A'. Instead of proceeding immediately with an analysis of our data in terms of Eq. (3) with D and D' different from zero, we first used a different approach. We retained the pure power-law analysis, and imposed the constraint $B = B'$ even though the statistics of the data does not really permit this constraint. The motivation for this is based on the experience gained in analyzing data near the superfluid transition of ^{4}He where it is known that confluent singular terms are important at the higher pressures.[26,27] We call the reader's attention to Figs. 10 and 12 of Ref. 26. The data shown there demonstrate that imposing the constraint $B = B'$, even when it is not statistically allowed, yields values for α and A/A' which are very close to the presumably correct ones which are obtained by permitting D and D' to be different from zero (those "correct" values are given in Fig. 14 of Ref. 26). The results of our analysis with $B=B'$ are shown as solid circles in Fig. 7. This analysis yields $\alpha=\alpha'=-0.130\pm0.01$ and $A/A'=1.54\pm0.05$, where the errors are somewhat subjective estimates based on the scatter in $\alpha=\alpha'$ and A/A'. Numerical values of the parameters for Eq. (3) with $t_{\min}=0.005$ and $t_{\max}=0.07$ are given in the second column of Table I.

d. Effect of sample inhomogeneities. Figure 4 demonstrates that the three samples are of varying quality. So far we have presented results of analyses for sample EuS-II which has the sharpest phase transition. We now wish to demonstrate that the varying extent of the "rounding" of the specific heat in the three samples does not influence $\alpha=\alpha'$ and A/A', provided that data in the obviously rounded region are excluded. Figure 4 indicates that the effect of sample imperfections is small for all three samples provided that $|t|\gtrsim0.003$. We therefore fitted the data with $0.005\leq|t|\leq0.07$ for each sample to Eq. (3) using $T_C=T_C'$, $E=E'$, $\alpha=\alpha'$, $B=B'$, and $D=D'=0$ as constraints. The parameters obtained from these fits are given in Table II. It is clear that this method of analysis yields critical point parameters which are independent of the quality of our samples.

e. Analysis with confluent singular terms. The data for EuS-II were also analyzed by fitting to Eq. (3) with D and D' permitted to be different from zero. We retained the constraints $T_C=T_C'$, $\alpha=\alpha'$, $E=E'$, and $B=B'$. The increase in the correlation between all parameters due to the inclusion of D and D' as least-squares adjusted parameters resulted in larger standard errors for B and B', and permitted the constraint $B=B'$ without a significant increase in the deviations from the fitted function. We fixed $x=x'=0.5$, consistent with experimental results for other systems[26,27] and theoretical predictions.[28] Because of the large remaining number of parameters that had to be determined from the data, it was not possible to perform an analysis over restricted temperature ranges. Thus we fitted the data only over the rather large range $0.003\leq|t|\leq0.07$. The results are given in the third column of Table I. It can be seen that these parameters do not differ significantly from those obtained with a pure power law fit and the constraint $B=B'$ (Sec. IV B 2 *c*). We therfore retain the values for $\alpha=\alpha'$ and A/A' obtained there; but we are inclined to increase somewhat our estimates of the probable errors. We quote

$$\alpha=\alpha'=-0.13\pm0.02 \tag{6a}$$

and

$$A/A'=1.54\pm0.1 \tag{6b}$$

as our best estimates of the specific-heat parameters and their errors.

f. Crossover effects. The values of $\alpha=\alpha'$ and A/A' which were derived above from the specific-heat data for EuS differ negligibly from those pertinent to the short-range-force Heisenberg system $RbMnF_3$.[10] As discussed in Sec. I, this is consistent with ϵ-expansion estimates for the values of the exponents of the dipolar and short-range-force systems. It remains to understand the anomalous behavior of EuO which has $\alpha=\alpha'\simeq-0.04$ and $A/A'\cong1.2$.[9] If we assume that the exponents and the amplitude ratio for EuO are effective parameters $\bar{\alpha}$ and $\bar{A}/\bar{A}'$, the values of which reflect the extent to which crossover from dipolar to short-range-force behavior has progressed in the temperature range of the data, then $\bar{\alpha}$ and $\bar{A}/\bar{A}'$ should, in principle, depend upon the range of t over which data are analyzed. If the crossover is sufficiently slow, however, this trend in $\bar{\alpha}$ or $\bar{A}/\bar{A}'$ might not be detectable over the experimentally accessible range. In order to search for a range dependence of $\bar{\alpha}$ and $\bar{A}/\bar{A}'$, we reanalyzed the EuO data[9] by fitting several segments of data to a pure power law with $T_C=T_C'$, $\alpha=\alpha'$, $B=B'$, and $E=E'$. Each segment

TABLE II. Parameters for Eq. (3) for samples I, II, and III. The constraints $T_C=T_C'$, $E=E'$, $\alpha=\alpha'$, $B=B'$, and $D=D'=0$ were imposed. The temperature range was determined by $t_{\min}=0.005$ and $t_{\max}=0.07$. The units of C_p and σ are J mole^{-1}K^{-1}. The uncertainties are standard errors and do not reflect possible systematic errors.

Sample	I	II	III
T_0 (K)	16.520	16.515	16.515
T_C (K)	16.5237 ± 0.0009	16.5112 ± 0.0006	16.5085 ± 0.0009
$\alpha=\alpha'$	-0.1272 ± 0.0043	-0.1311 ± 0.0031	-0.1335 ± 0.0040
A/A'	1.509 ± 0.023	1.536 ± 0.018	1.541 ± 0.022
$B=B'$	43.74 ± 0.84	43.08 ± 0.58	42.76 ± 0.69
$E=E'$	20.67 ± 0.88	21.68 ± 0.61	20.66 ± 0.86
A'	4.163 ± 0.034	4.192 ± 0.022	4.243 ± 0.034
σ	0.0303	0.0201	0.0259

consisted only of the data spanning one or one-half decade in t. As a control, we did similar analyses with the results for $RbMnF_3$ (Ref. 10) and EuS. The parameters are shown in Fig. 8 as a function of $\langle \log_{10}|t|\rangle$ were

$$\langle \log_{10}|t|\rangle \equiv \tfrac{1}{2}(\log_{10}|t_{max}| + \log_{10}|t_{min}|) \quad . \tag{7}$$

We find that the data for $RbMnF_3$ and EuS do not reveal any range dependence of α or A/A'. However, for EuO there appears to be a trend both in $\bar{\alpha}$ and in $\bar{A}/\bar{A}'$. Although this trend is only slightly larger than the standard errors (which are particularly large when data over only half a decade are used), it does tend to support the explanation of the anomalous behavior of EuO in terms of crossover effects.

The effective specific-heat exponent in the dipolar to isotropic short-range-force crossover region was calculated by Bruce *et al.*[13] from an expansion of the scaling function to first order in ϵ. Their theoretical result does indeed suggest that $\bar{\alpha}$ should be a nonmonotonic function of t, initially increasing by about 0.1 from the pure dipolar value as t is increased. This increase in $\bar{\alpha}$ is consistent with the difference in the experimental exponents -0.13 and -0.04 determined for EuS and EuO,[9] respectively.

We have attempted to obtain a best estimate of the behavior of $\bar{\alpha}$ and $\bar{A}/\bar{A}'$ by taking the experimental EuS result as the pure dipolar limit, and drawing a smooth curve with this limit at small $|t|$ through the EuO parameters in Fig. 8. This estimate is shown in the figure as a solid line. Also shown is the temperature t_x in the general vicinity of which crossover effects should occur.[2] The observed crossover effects occur over one decade below t_x, and presumably would continue to occur for perhaps another decade above t_x before the effective exponents reach the short-range-force limit. Of course at those temperatures far from T_C the behavior of the system is complicated by other phenomena such as crossover to mean-field behavior, and therfore one cannot expect to observe the pure short-range-force behavior in the real system. The width of perhaps two decades for the crossover region of the real system is somewhat narrower than the theoretical result,[13] which indicates a width of about four decades; but it is not clear that this feature is displayed sufficiently accurately by an expansion of the scaling function to first order in ϵ.

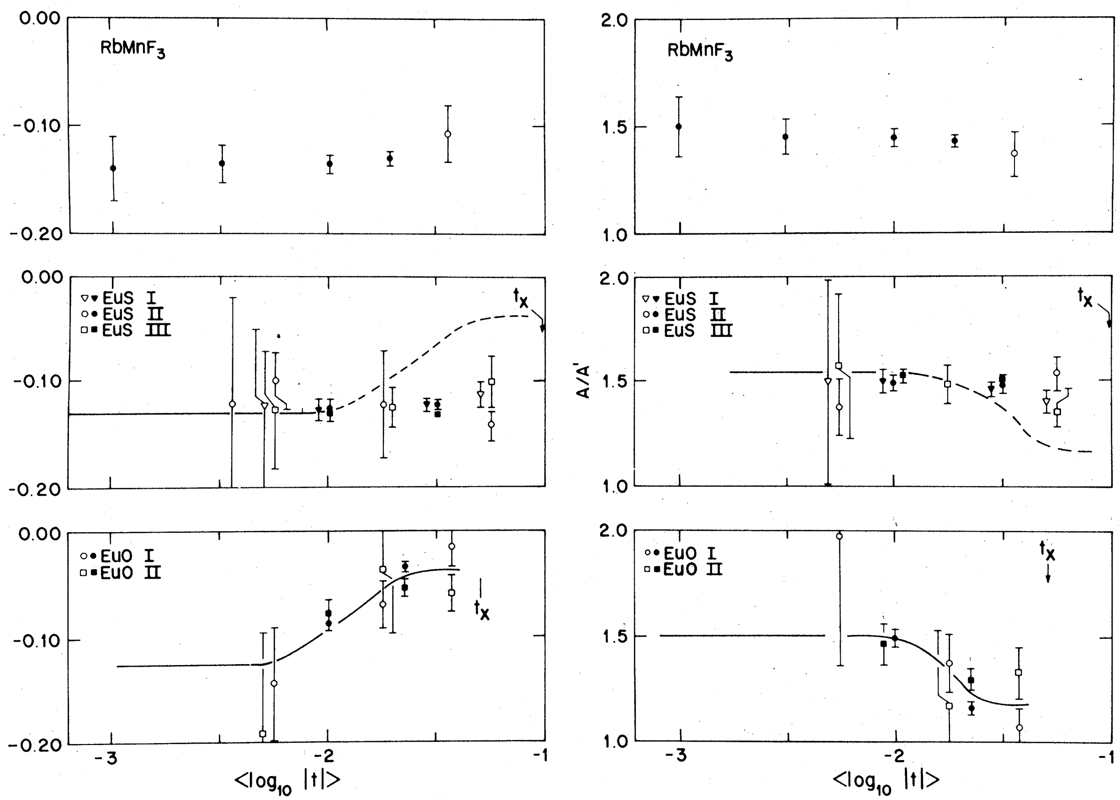

FIG. 8. Parameters $\alpha = \alpha'$ and A/A' obtained for $RbMnF_3$, EuS, and EuO by fitting data over limited range of $|t|$ (solid circles: one decade; open circles: half a decade) to Eq. (3) with the constraints $T_C = T_C'$, $\alpha = \alpha'$, $B = B'$, $E = E'$, and $D = D' = 0$. The solid line and dashed lines are explained in the text.

Although the above qualitative considerations provide a reasonable explanation for the behavior of EuO, they also imply that it is surprising that no crossover effects are observed in the case of EuS. For EuS, t_x is estimated to be only a factor of 2 larger than for EuO (Ref. 2); and we would thus expect that a simple translation of the solid lines through the EuO data to the right by $\log_{10}(2)$ should fit the effective parameters for EuS. This translated curve is reproduced in the middle sections of Fig. 8 as a dashed line. It is clearly inconsistent with the EuS data at large $|t|$. An explanation for this discrepancy might be sought in an incorrect estimate of t_x for EuS. Alternately, it is possible that additional complications exist in the case of EuS for $|t| \gtrsim 0.02$, perhaps due to crossover to mean-field behavior, which happen to cancel the dipolar to short-range-force crossover effects.

In this section we have presented some experimental evidence for the existence of crossover effects in the specific heat of EuO; but we do not suggest that this evidence, taken by itself, is conclusive. Only the comparison of measurements on the three materials $RbMnF_3$, EuO, and EuS, and their comparison with explicit theoretical calculations of the crossover scaling function, present a reasonably convincing picture. On the other hand, it has been claimed by Salamon[29] that his own measurements of C_p for EuO, taken by themselves, provide conclusive evidence for crossover from dipolar to short-range-force behavior. We do not believe that his claim is justified. Rather, we feel that his "crossover effects" are the result of an improper analysis of his data. We have discussed this point in detail elsewhere.[9] Here we only point out that his amplitude ratios $A/A'=2.03$ for the dipolar region and $A/A'=1.00$ for the short-range-force region do not agree with the experimental values 1.54 and 1.46 for EuS and $RbMnF_3$, respectively.

V. COMPARISON WITH OTHER EXPERIMENTS

A. Specific heat

The specific heat of EuS near T_C was measured previously by Van der Hoeven *et al.*[11] These authors subtracted an estimate of the lattice contribution from their data and reported only the remaining magnetic specific heat. Direct comparison is therefore not possible. Their analysis yielded $\alpha = 0.00 \pm 0.03$ and $\alpha' = -0.25 \pm 0.03$ which disagrees with our results. Another set of measurements of C_p for EuS was reported by Teaney and Moruzzi.[12] In this case, both the magnetic specific heat and the estimate for the lattice contribution were reported, and the total C_p can be reconstructed. The result differs from our C_p being 20% higher than ours at 10 K and 10% lower at 20 K. We have no explanation for these large differences.

A comparison of our data below 5 K with those of Passenheim *et al.*[16] and Dietrich *et al.*[17] was made already in Fig. 5. The agreement is very good.

B. Neutron scattering and scaling

Extensive neutron-scattering experiments for EuS have been performed by Als-Nielsen *et al.*[30] These experiments yield the exponents β, ν, and γ which are related to the specific-heat exponent α via the scaling laws[24]

$$\alpha = 2 - 2\beta - \gamma \tag{8}$$

and

$$\alpha = 2 - 3\nu \ . \tag{9}$$

The neutron results[30] are

$$\beta = 0.36 \pm 0.01 \ , \tag{10a}$$

$$\nu = 0.70 \pm 0.02 \ , \tag{10b}$$

$$\gamma = 1.40 \pm 0.04 \ . \tag{10c}$$

These parameters yield

$$2 - 2\beta - \gamma = -0.12 \pm 0.04 \tag{11}$$

and

$$2 - 3\nu = -0.11 \pm 0.07 \ , \tag{12}$$

in good agreement with our measured value $\alpha = \alpha' = -0.13 \pm 0.02$.

C. Other experiments and scaling

If we assume the validity of scaling,[24,31] then our value of α can be used to obtain the correlation length exponent ν from the relation $3\nu = 2 - \alpha$. In order to derive the remaining exponents, one additional exponent must be known. Consistent with theoretical estimates for the short-range-force system[5,32–34] and for the dipolar system,[5] we will use $\eta = 0.03$ and assign to it the rather generous uncertainty of ± 0.02. This enables us to derive the susceptibility exponent γ from $\gamma = \frac{1}{3}(2-\eta)(2-\alpha)$ and the magnetization exponent β from $\beta = \frac{1}{6}(1+\eta)(2-\alpha)$. We obtain

$$\alpha = -0.13 \pm 0.02 \ , \tag{13a}$$

$$\beta = 0.366 \pm 0.01 \ , \tag{13b}$$

$$\gamma = 1.40 \pm 0.03 \ , \tag{13c}$$

$$\nu = 0.710 \pm 0.006 \tag{13d}$$

as our best estimates for the exponents. These can be compared with other experiments. The agreement with the neutron results quoted above is obviously excellent. The result $\beta = 0.33 \pm 0.015$ was obtained by Heller and Benedek,[35] and is rather low compared to our estimate. Közler *et al.*[36] found $\gamma = 1.35 \pm 0.03$. Although this value differs somewhat from our value, the errors overlap. The values $\gamma = 1.06 \pm 0.05$ and $\beta = 0.335 \pm 0.01$ were obtained by Berkner.[37] They differ significantly from our estimates.

VI. SUMMARY AND CONCLUSIONS

We reported in this paper the results of specific-heat measurements for the isotropic ferromagnetic EuS over the temperature range $1.5 \lesssim T \lesssim 35$ K.

For $T \lesssim 5$ K, our measurements are in good agreement with previous results by others,[16,17] and tend to confirm the values of the exchange parameters derived by Dietrich *et al.*[17] from the measurements by Passenheim *et al.*[16]

Near the Curie temperature T_C, the measurements were made on three different samples of varying quality. We were able to demonstrate that the sample quality did not influence our conclusions regarding the critical point parameters.

The data near T_C were analyzed first by fitting them to a pure power law. They were found to be consistent with the scaling prediction[24] $\alpha = \alpha'$, but did not permit the constraint $B = B'$ which is predicted by the RG theory.[25] We interpret the apparent inequality between B and B' as an indication for the need to include confluent singular terms in the data analysis.

During the analysis of thermal-expansion data near the superfluid transition in ^{4}He it was found that the imposition of the constraint $B = B'$, even when it is not statistically allowed by the data, will tend to result in the correct values for $\alpha = \alpha'$ and A/A'.[26] We therfore imposed the constraint $B = B'$ in the pure power-law analysis, and obtained $\alpha = \alpha' = -0.130 \pm 0.01$ and $A/A' = 1.54 \pm 0.05$. As an alternate interpretation, we also fitted the results to a function which, in addition to the leading power-law term, contained confluent singular terms [Eq. (3)]. This analysis yielded $\alpha = \alpha' = -0.124 \pm 0.015$ and $A/A' = 1.54 \pm 0.09$, consistent with the pure power-law analysis with $B = B'$.

The values obtained here for $\alpha = \alpha'$ and A/A' agree within experimental error with the results derived previously for the isotropic Heisenberg antiferromagnet $RbMnF_3$.[10] From the RG theory it is expected that the critical behavior of ferromagnets should be altered by the presence of dipolar forces, whereas for antiferromagnets dipolar forces are believed not to affect the critical behavior. However, theoretical estimates of the exponents based on the ϵ expansion to second order for the two types of critical points yield the values $\alpha = -0.125$ and -0.135 (Ref. 5); and these results are so similar as to be indistinguishable by our experiments. We therfore conclude that our data are consistent with the theoretical predictions.

Although the present results for EuS are in agreement with theory, it remained to explain the anomalous behavior of EuO which was reported previously.[9] The experimental values $\alpha = \alpha' = -0.04$ and $A/A' = 1.2$ which had been obtained for EuO are distinctly different from those for EuS and $RbMnF_3$. In Sec. IV B 2 *f* we have presented some evidence which tends to support an explanation of the anomalous behavior of EuO in terms of effects associated with crossover from isotropic short-range-force to dipolar behavior. We find that the experimental estimates of effective exponents associated with this crossover are consistent with calculations of the crossover scaling function by Bruce *et al.*[13]

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[1]Early statements of the hypothesis of universality may be found in M. E. Fisher, Phys. Rev. 16, 11 (1966); P. G. Watson, J. Phys. C 2, 1883 (1969); 2, 2158 (1969); D. Jasnow and M. Wortis, Phys. Rev. 176, 739 (1968). More recent references are L. P. Kadanoff, in Critical Phenomena, *Proceedings of the International School "Enrico Fermi,"* edited by M. S. Green, (Academic, New York, 1971); R. B. Griffiths, Phys. Rev. Lett. 24, 1479 (1970); D. D. Betts, A. J. Guttmann, and G. S. Joyce, J. Phys. C 4, 1994 (1971); D. Stauffer, M. Ferer, and M. Wortis, Phys. Rev. Lett. 29, 345 (1972); K. G. Wilson and J. Kogut, Phys. Rep. C 12, 76 (1974). For recent reviews of the RG theory, see also M. E. Fisher, Rev. Mod. Phys. 46, 597 (1974); and S. K. Ma, *Modern Theory of Critical Phenomena* (Benjamin, Reading, Mass., 1976). For a more general introduction to the field of critical phenomena, and for the definition of critical exponents, see H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford U. P., Oxford/New York, 1971).

[2]M. E. Fisher and A. Aharony, Phys. Rev. Lett. 30, 559 (1973).

[3]A. Aharony, Phys. Rev. B 8, 3349 (1973).

[4]A. Aharony and M. E. Fisher, Phys. Rev. B. 8, 3323 (1973).

[5]A. D. Bruce and A. Aharony, Phys. Rev. B 10, 2078 (1974).

[6]A. Aharony and A. D. Bruce, Phys. Rev. B 10, 2973 (1974).

[7]Th. Nattermann and S. Trimper, Phys. Lett. A 50, 307 (1974).

[8]A. Kornblit, G. Ahlers, and E. Buehler, Phys. Lett. A 43, 531 (1973).

[9]A. Kornblit and G. Ahlers, Phys. Rev. B 11, 2678 (1975).

[10]A. Kornblit and G. Ahlers, Phys. Rev. B 8, 5163 (1973).

[11]B. J. Van der Hoeven, D. T. Teaney, and V. L. Moruzzi, Phys. Rev. Lett. 20, 719 (1968).

[12]D. T. Teaney and V. L. Moruzzi, in *Colloques Internationaux du Centre National de la Recherche Scientifique No. 180: Les Elements des Terres Rares, Paris-Grenoble, May, 1969* (Centre Nat. Rech. Sci., Paris, 1970), Tome II, p. 131.

[13]A. D. Bruce, J. M. Kosterlitz, and D. R. Nelson, J. Phys. C 9, 825 (1976).

[14]T. Nattermann and S. Trimper (unpublished).

[15]A. D. Bruce, J. M. Kosterlitz, and D. R. Nelson, AIP Conf. Proc. 24, 321 (1975).

[16]B. C. Passenheim, D. C. McCollum, Jr., and J. Callaway, Phys. Lett. 23, 634 (1966).

[17]O. W. Dietrich, A. J. Henderson, and H. Meyer, Phys. Rev. B 12, 2844 (1975).

[18]G. Ahlers, Phys. Rev. 171, 275 (1968). A more detailed description is given by K. H. Mueller, G. Ahlers, and F. Pobell, Phys. Rev. B 14, 2096 (1976).

[19]G. Ahlers, Rev. Sci. Instrum. 37, 477 (1966).

[20]G. Ahlers, J. Phys. Chem. Solids 28, 525 (1966).

[21]See AIP document No. PAPS PLRBA-17- 282-7 for 7 pages of tabulated data. Order by PAPS number and journal reference from American Institute of Physics, Physics Auxiliary Publication Service, 335 East 45th St., New York, N. Y. 10017. The price is $1.50 for each microfiche (98 pages), or $5.00 for photocopies of up to 30 pages with $0.15 for each additional page over 30 pages. Airmail additional. Make checks payable to the American Institute of Physics. This material also appears in Current Physics Microfilm, the monthly microfilm edition of the complete set of journals published by AIP, on the frames immediately following this journal article.

[22]We neglect here the contribution from the hyperfine splitting of the Eu^{151} and Eu^{153} nuclei which is important for $T \lesssim 1$ K but negligible in our temperature range.

[23]G. Ahlers and A. Kornblit, Phys. Rev. B 12, 1938 (1975).

[24]B. Widom, J. Chem. Phys. 43, 3898 (1965); C. Domb and D. L. Hunter, Proc. Phys. Soc. Lond. 86, 1147 (1965); L. P. Kadanoff, Physics (N.Y.) 2, 263 (1966); A. Z. Patashinskii and V. L. Pokrovskii, Zh. Eksp. Teor. Fiz. 50, 439 (1966) [Sov. Phys.-JETP 23, 292 (1966)]; R. B. Griffiths, Phys. Rev. 158, 176 (1967); J. W. Essam and M. E. Fisher, J. Chem. Phys. 39, 842 (1963).

[25]The constraint $B = B'$ has been discussed in detail in Refs. 9 and 23, and by M. Barmatz, P. C. Hohenberg and A. Kornblit, Phys. Rev. B 12, 1947 (1975).

[26]K. H. Mueller, G. Ahlers, and F. Pobell, in Ref. 18.

[27]D. S. Greywall and G. Ahlers, Phys. Rev. A 7, 2145 (1973).

[28]The choice $x = x' = 0.5$ has been discussed in Refs. 9, 23, 26, and 27. The ϵ-expansion result of Ref. 5 for x (ω in Table I of Ref. 5) in the dipolar case does not differ appreciably from the isotropic short-range-force case.

[29]M. B. Salamon, Solid State Commun. 13, 1741 (1973).

[30]J. Als-Nielsen, O. W. Dietrich, W. Kunnmann, and L. Passell, Phys. Rev. Lett. 27, 741 (1971); and J. Als-Nielsen, O. W. Dietrich, and L. Passell, Phys. Rev. B 14, 4908 (1976).

[31]The scaling laws have also been derived from the RG theory (see, for instance, K. G. Wilson and J. Kogut, in Ref. 1; M. E. Fisher, in Ref. 1; and S. K. Ma, in Ref. 1). This lends very strong support to the assumption of their validity.

[32]K. G. Wilson, Phys. Rev. Lett. 28, 548 (1972); E. Brezin, J. C. LeGuillou, J. Zinn-Justin, and B. G. Nickel, Phys. Lett. A 44, 227 (1973).

[33]G. R. Golner and E. K. Riedel, Phys. Lett. A 58, 11 (1976); G. A. Baker, Jr., B. G. Nickel, M. S. Green, and D. I. Meiron, Phys. Rev. Lett. 36, 1351 (1976); J. C. LeGuillou and J. Zinn-Justin, *ibid.* 39, 95 (1977).

[34]D. S. Ritchie and M. E. Fisher, Phys. Rev. B 5, 2668 (1972).

[35]P. Heller and G. Benedek, Phys. Rev. Lett. 14, 71 (1965).

[36]J. Kotzler, G. Kamleiter, and G. Weber, J. Phys. C 9, L361 (1976); and private communications.

[37]D. D. Berkner, Phys. Lett. A 54, 396 (1975).