

Specific heat of $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ near the ferromagnetic phase transition

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High-resolution measurements of the specific heat, C , near the ferromagnetic phase transition of the diluted ferromagnetic system $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ are reported. Samples with four different concentrations ($x=1$, $x=0.9$, $x=0.7$ and $x=0.5$) have been studied and the expected phase diagram, i.e. the linear decrease of T_c with decreasing x , is confirmed. Our specific-heat data of the pure EuO samples yield a critical exponent $\alpha = -0.12 \pm 0.02$ in contrast to literature results, but in agreement with the value expected theoretically for this three-dimensional Heisenberg system. The origin of the discrepancies is traced down to differences in data analysis.

EuO is a well-known ferromagnet where the critical behavior at the magnetic phase transition, $T_c \approx 69$ K, has been extensively investigated. It is a text-book example for a three-dimensional (3D) Heisenberg system. Nearly all static critical exponents are in very good agreement with values calculated by renormalization group theory [1]. Up to now, the only discrepancy (beyond the error bars) between the predicted and measured values was found for α , the critical exponent of the specific heat. Experimentally, $\alpha = -0.04$ was obtained [2], which differs substantially from $\alpha = -0.116$ expected theoretically [3]. As a possible explanation of this disagreement dipole-dipole interactions have been proposed [4]. It is known that these interactions have a strong influence on the dynamic critical behavior of EuO and other cubic ferromagnets, as seen, e.g., in a crossover in the dynamic susceptibility close to T_c [5]. Also, the static exponents might be affected, yielding effective exponents in the crossover region [4]. Nevertheless, if α would be influenced by dipolar interactions it is by no means clear why no effect is seen in the magnetic critical exponents γ , β and δ . In addition, as pointed out already by the authors of [2] the scaling relations connecting the different exponents, e.g., the Rushbrooke relation $\alpha = 2(1 - \beta) - \gamma$ would be violated with $\alpha = -0.04$. On the other hand, correction terms to the leading power law of the specific

heat at T_c lead to $\alpha \approx -0.10$ [2]. An additional ambiguity arises from the data analysis, in particular in the presence of unavoidable sample inhomogeneities.

In order to clarify this problem, we investigated the specific heat C of ten different EuO samples from six different charges. C near the Curie temperature T_c was measured with high precision allowing us to get reliable results of the critical behavior and to analyze the data close to T_c with the necessary accuracy. In addition, EuO was diluted with diamagnetic SrO. $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ samples with $x=0.9$, $x=0.7$ and $x=0.5$ were measured to gain more information about the phase diagram and the concentration dependence of the critical behavior.

EuO is an isotropic insulating ferromagnet with both ferromagnetic nearest neighbor ($J_1/k_B = 0.55 \pm 0.05$ K) and next-nearest neighbor ($J_2/k_B = 0.15 \pm 0.1$ K) exchange interactions [6]. With random replacement of EuO by SrO, the phase boundary para- to ferromagnetic is expected to decrease monotonically down to the percolation limit for the fcc structure with nearest and next-nearest neighbor exchange interactions, $x_p = 0.136$. Below x_p no long-range ordering should be present.

The single crystals of EuO are prepared from a nominal composition of approximately $\text{Eu}_{1.050}\text{O}$ consisting either of Eu_2O_3 powder (Johnson Matthey, specpure for run #513-1, and Metals research 4N pure for the other runs) plus Eu metal (Ames Laboratory, Iowa State University), or from oxygen-rich inhomogeneous EuO pieces plus Eu metal. Growth was performed in sealed molybdenum crucibles in a tungsten-mesh heated vacuum furnace. According to the phase diagram [7] directional freezing from melt or flux would yield inhomogeneous crystals.

To obtain homogeneous EuO single crystals a near-equilibrium growth run had to be performed. This was accomplished by growth at $T_g \approx \text{const}$ with a small temperature gradient applied over the charge volume. At the growth temperature the charge consists of a solid phase, a metal-rich liquid (flux), and a gas phase. The fraction of the charge acting as flux depends on the nominal composition. In the case of $\text{Eu}_{1.050}\text{O}$ and $T_g = 1780$ °C for the

growth of 1:1 stoichiometric crystals [7], the flux has a composition of approximately $\text{Eu}_{1.27}\text{O}$ and the solid to flux ratio is 4:1. Oxygen-rich and metal-rich crystals from the same nominal composition have also been grown at $T_g = 1820^\circ\text{C}$ and 1740°C , respectively.

With this technique, charges of 15 to 35 grams transformed within 15 to 25 h into cm^3 -sized single crystals. The ampoule was cooled fast by 'power off' down to approximately 900°C and then within 8 h to room temperature. The crystal was located at the bottom of the ampoule embedded in a thin frozen flux layer with most of the flux on top.

For the growth of $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ crystals, the starting materials were EuO crystal pieces and SrO powder obtained by calcinating SrCO_3 . The SrO powder and the EuO crystal pieces were ground together in a mortar in dry atmosphere and sealed into tungsten crucibles. The starting composition was varied between $x=0.9$ and $x=0.5$. With decreasing x the growth temperature was raised from 1850°C to 2150°C . It was found that with increasing Sr content stoichiometric charges (with respect to the metal-oxygen ratio) attacked the tungsten crucible and tungsten was precipitated in the crystals or on the surface of the material. Therefore crystals were grown from metal-rich compositions which suppressed the tungsten attack. Crystal growth occurred below the melting temperatures by grain growth in the presence of 10 to 20 mol% flux. The bigger grains could be cleaved into samples.

The specific heat was measured between 20 and 80 K by the continuous method in a ^4He cryostat. The samples with masses between 18 and 228 mg were fixed with Apiezon N grease on top of a sapphire plate. On the back of this plate an evaporated manganin film of approximately 1800 Ohm served as heater and a platinum resistor (PT 100) as thermometer. The heat capacity of this whole assembly was always less than 20% of the total capacity including the sample. The sample holder was surrounded by two separate copper shields. To reduce additional heating effects by radiation the temperature of these shields was controlled by a computer as close to the temperature of the sample as possible, typically to less than 1 mK temperature difference between sample and inner shield. The overall accuracy of the absolute value of C was checked by measuring the specific heat of high-purity copper (residual resistance ratio 7000) yielding an agreement with literature data [8] within 1% in the temperature range of the present investigation. Besides C , the dc -magnetization in a magnetic field of ~ 1 mT was measured in the vicinity of T_c .

Figure 1 shows C of four investigated $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ samples with nominal concentrations of $x=1$, $x=0.9$, $x=0.7$ and $x=0.5$ close to the ferromagnetic transition temperatures. Our measurements show that the specific heat of the two samples with large Eu concentrations ($x=1$ and $x=0.9$) have both a λ -type anomaly at T_c . The sample with $x=0.9$ shows a slightly broader and smeared anomaly. With increasing Sr dilution the maximum of C at T_c broadens more and more, the sample with $x=0.7$ shows a curvature change already below T_c . For the most diluted sample investigated here ($x=0.5$) the phase tran-

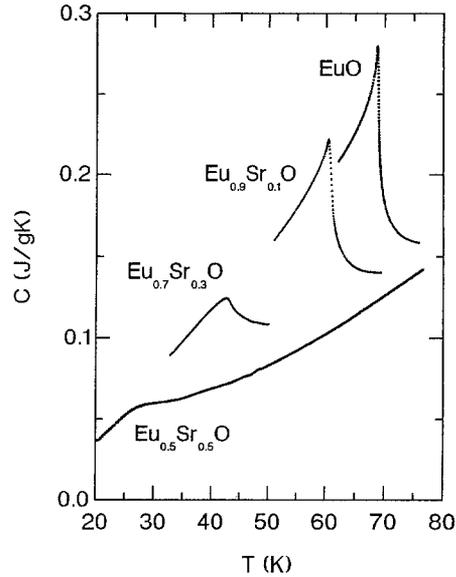


Fig. 1. Specific heat C vs. temperature T of $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ with $x=1$, $x=0.9$, $x=0.7$ and $x=0.5$

sition is smeared out and only a shallow maximum around 28 K is found. Both samples with $x=0.7$ and $x=0.5$ have an additional small feature in C at approximately 48 K (barely visible in Fig. 1) showing clearly the presence of sample inhomogeneities in these crystals.

The decreasing quality of the samples with increasing Sr content can also be seen in the difference of T_c determined by specific heat and dc -magnetization measurements, respectively. Figure 2 shows T_c vs. x for the investigated $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ samples. In addition to the values extracted from our measurements of the maximum of the anomaly in C vs. T (filled circles) and of the extrapolated onset of the constant magnetization below T_c for the same samples (open squares), we also plotted data points of the $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ phase diagram from the only previous study [9], obtained by susceptibility measurements of polycrystalline samples. The latter values show considerable

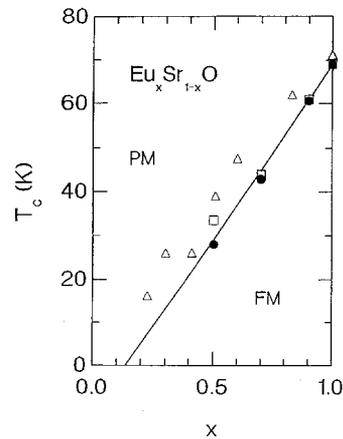


Fig. 2. Magnetic phase diagram T_c vs. x of $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ (circles: specific heat; squares: dc -magnetization (this work); triangles: magnetic susceptibility [9]). The results of the specific-heat measurements and the dc -magnetization measurements coincide for $x=1.0$. Solid line extrapolates to the percolation threshold $x_p=0.136$

scatter and are, in addition, always higher than the critical temperatures determined by our measurements. Overall, T_c decreases nearly linearly with increasing Sr content, cf. Fig. 2. The solid line extrapolates very well to the percolation threshold $x_p = 0.136$ for a fcc-crystal with both ferromagnetic nearest and next-nearest neighbor exchange interactions [10].

As already mentioned, we measured ten different EuO-samples of six different charges. Crystals from a metal-rich charge show only a very broad featureless maximum in C . Therefore, these measurements will not be discussed further. The specific heat of four samples of the remaining charges is shown in Fig. 3. The temperature dependence and the shape of the anomaly of these samples is nearly identical. The averaged value of $T_c = 69.0 \pm 0.2$ K for our investigated EuO samples is in very good agreement with most of the published values [6]. The relative deviation of T_c for the different charges is less than 0.5%, the maximum values of C are up to 5% apart from each other. This latter effect is mainly attributed to the different sample qualities and will be discussed in more detail later.

To check the macroscopic sample homogeneity of the individual charges we measured four pieces cut from different positions of the charge #513-1. We found no significant differences in C and obtained a very good reproducibility of our results and the critical parameters.

A comparison of our specific-heat results with those of Kornblit and Ahlers [2] shows a very good agreement of these two data sets. The temperature of the maximum in C is less than 0.5% apart from each other and the absolute C values agree within 2%.

To obtain the critical exponent α we analyzed our specific-heat data close to T_c with the usual formula [2]

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

for $T > T_c$ with $t = (T - T_c)/T_c$ and the same function with primed parameters for $T < T_c$. A more detailed description of this function and the parameters involved

has been given previously [2, 11, 12]. Equation (1) does not include higher-order singular corrections. These corrections appeared not to be necessary for a fit, as already found for EuS [11]. The point up to where the critical behavior can be represented by (1) is determined by the upper limit of the fit range $|t_{\max}|$ which was always fixed to a constant value around $|t_{\max}| = 0.09$. The lower limit closer to T_c , $|t_{\min}|$, was reduced successively until the fit curve deviated considerably from the data points. In most cases this procedure yielded different values for the data sets for $T > T_c(t_{\min})$ and for $T < T_c(t'_{\min})$.

An important parameter affecting all other fit parameters in (1) is T_c . To find the correct T_c we used the following iterative procedure: the rounding close to T_c seen in the deviation of the fit from the data points is attributed to small sample inhomogeneities producing a distribution of slightly different transition temperatures within the sample. With the assumption of a Gaussian distribution of the volume of subsystems with critical temperatures deviating with variance δT_c from T_c of a perfectly homogeneous sample, we are able to describe the measured specific heat over the whole temperature range, but only if we choose the appropriate T_c [12, 13]. Therefore, we iteratively fitted the data with (1) for a fixed $T_c = T'_c$ without inclusion of the rounding parameter δT_c . Then we numerically calculated with fixed parameters $\alpha = \alpha'$, $A, A', B = B'$ and $E = E'$ the complete specific heat from (1) taking into account the Gaussian distribution of subsystems by adjusting δT_c for a good match of the resulting fit curve with the data. This procedure was repeated with a different choice of T_c until an optimum fit resulted. With this method we always obtained a T_c which was higher than the temperature where C has its maximum value. A typical result of this fit procedure is shown for one EuO sample in Fig. 4 where C is plotted vs. $\log |t|$. The solid lines represent the fitting without rounding according to (1). At approximately $t < t_{\min} = 0.004$ and $|t'| < |t'_{\min}| = 0.0025$ a clear deviation of the fit from the measured data points is visible. With the inclusion of a smearing of $\delta T_c/T_c = 0.0013$, we

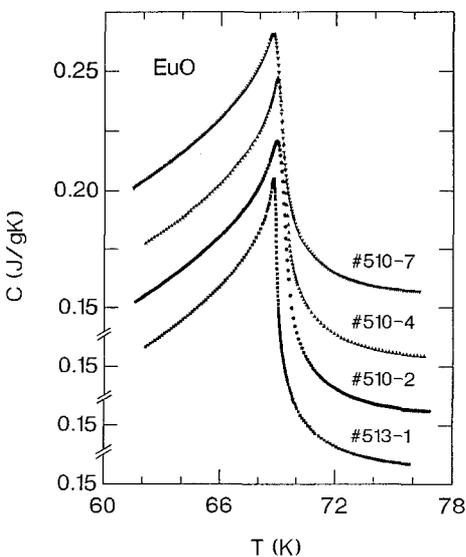


Fig. 3. Specific heat C vs. temperature T of four different EuO samples

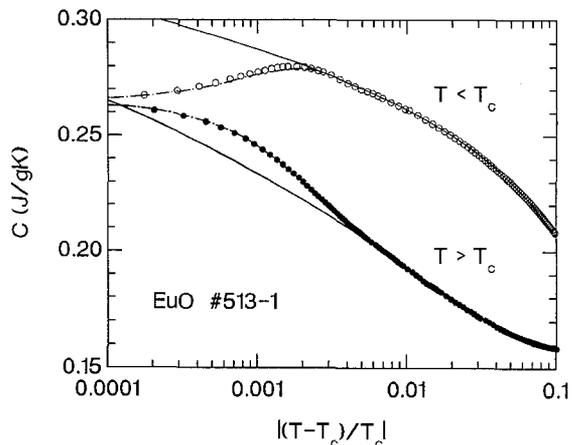


Fig. 4. Specific heat C vs. the reduced temperature $|T - T_c/T_c|$ of EuO (sample #513-1) with fits after (1) (solid curves) and fits with a Gaussian distribution of transition temperatures (dash-dotted curves). The fitting parameters are given in Table 1

obtain perfect agreement between measured and calculated C vs. T dependence (dash-dotted lines in Fig. 4). In particular, not only the maximum of C for the data set $T < T_c$, but also the enhancement of C for the data set $T > T_c$ close to T_c as compared to the behavior of ideal samples described by (1) is now easily explicable if $A \neq A'$.

For the sample #513-1 (Fig. 4) we find the critical exponent $\alpha = -0.129 \pm 0.02$ and the amplitude ratio $A/A' = 1.79 \pm 0.15$, α agrees very well with the theoretical value, while A/A' is somewhat larger than predicted [14]. Within the error bars mentioned in Table 1, the quality of the fit measured by the RMS error did not change significantly. The mean value taken over five EuO samples for which the specific heat was analyzed in detail (see Table 1 for the fit parameters) are $\alpha = -0.118 \pm 0.02$ and $A/A' = 1.73 \pm 0.2$. In the following we compare our results which are summarized for all charges in Table 1, with the parameters obtained from other experiments. The determination of the critical exponent α by Salamon [15] yielded an apparent crossover and, therefore, two different values: $\alpha = -0.026$ for the data points $|t| < 0.02$ and $\alpha = -0.09$ for $0.02 < |t| < 0.2$. However, as already pointed out by the authors of [2] the method of data analysis used in [15] is ambiguous and the crossover effect can be entirely removed by an appropriate choice of the relevant parameters T_c and E .

Comparing our result of $\alpha = -0.12 \pm 0.02$ with the value of $\alpha = -0.044 \pm 0.01$ reported by Kornblit and Ahlers [2] it is somewhat surprising to find such a large difference in spite of the fact that the overall specific-heat data are almost identical. A closer look at the data analysis of [2] shows the origin of this discrepancy. The lower boundary for the data set $T > T_c$ in [2] was chosen to $t_{\min} = 0.0025$ (for the sample shown in Fig. 4 we limited the fit to $t_{\min} = 0.004$). This results in a T_c appreciably above the maximum temperature of C for the best fit according to (1). Therefore, in a plot of C vs. $\log |t|$ like Fig. 4 no enhancement of the measured data closely above T_c is visible (see Fig. 2 of [2]). Nevertheless, this enhancement is a real sample effect and, as mentioned above, can be easily explained by the special form of (1) with $A/A' \neq 1$ and by a statistical distribution of T_c 's including sample inhomogeneities as for $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$ [12] and for diluted Ising antiferromagnets as, e.g., $\text{Mn}_x\text{Zn}_{1-x}\text{F}_2$ by Jaccarino and King [13]. This effect has also been em-

phasized by the authors of [2] to explain their observed rounding close to T_c without, however, analyzing this rounding quantitatively. Indeed, reanalysis of the scanned data of sample I of [2] with T_c adjusted to $T_c = 69.25$ K instead of 69.332 K yield a satisfactory description of the data with $\alpha = -0.11$ in agreement with our results and as expected from theory. The obtained rounding parameter $\delta T_c/T_c \approx 0.0013$ reveals the comparable quality of the investigated crystals [16].

The above analysis in terms of a distribution of T_c 's assumes that the macroscopic sample is divided into subsystems each having a sharp transition, but with different T_c 's. The different T_c 's could result from tiny differences in the average concentrations. However, according to the Harris criterion [17], the diverging correlation length ξ for $T > T_c$ should average over these concentration fluctuations for $\alpha < 0$, yielding a sharp T_c . Therefore, the subsystems have to be *independent*, preventing ξ to grow to infinity as T approaches T_c . This break-up into different subsystems might be due to extended lattice defects such as mosaic grain boundaries of very small angle, dislocations, or macroscopic concentration fluctuations. Of course, the finite size of the subsystems may also contribute to the rounding.

We now briefly discuss a possible effect of the dipolar dynamic crossover on the specific-heat exponent. Theoretically, this crossover is expected to occur for EuO around a reduced temperature $t_x \approx 5 \cdot 10^{-2}$ [4] while experimentally $t_x \approx 1.2 \cdot 10^{-1}$ [5]. In this crossover region the effective exponent α_{eff} can differ widely from the asymptotic exponent α_0 . Here the effective exponent $\alpha_{\text{eff}}(t_x)$ is defined by (1) with the fit range limited to one decade on either side of t_x . Since $|t_x|$ is at least one order of magnitude larger than the $|t|$ range where rounding sets in (cf. Fig. 4), the dipolar dynamic crossover does not seem to affect α to any measurable extent. The predicted differences between the asymptotic exponents for Heisenberg and dipolar ferromagnets [18] are too small to be detected by experiment. For EuS, t_x is a factor of two larger [4, 5] hence the experiments which yielded $\alpha = -0.13$ [11, 19] may have in fact probed the dipolar region more deeply than for EuO. For EuS no influence of the upper limit of the fit range $|t_{\max}|$ on the result of the fit procedure of the critical exponent is observed [11], similar to the present findings. This suggests that no sig-

Table 1. Experimental parameters according to (1) for $\text{Eu}_x\text{Sr}_{1-x}\text{O}$. The units of A , B and E in (1) are J/gK

Sample	T_c (K)	α	$A \cdot 10^2$	A/A'	B	E	$\delta T_c/T_c$
EuO # 510-2	69.21 ± 0.02	-0.10 ± 0.03	3.305 ± 0.4	1.57 ± 0.2	0.398 ± 0.02	0.233 ± 0.02	0.0032
EuO # 514-4	69.20 ± 0.01	-0.095 ± 0.03	3.19 ± 0.4	1.52 ± 0.2	0.407 ± 0.02	0.228 ± 0.03	0.0025
EuO # 510-7	69.00 ± 0.02	-0.128 ± 0.03	3.71 ± 0.4	1.82 ± 0.2	0.348 ± 0.02	0.255 ± 0.02	0.0029
EuO # 513-1	68.859 ± 0.01	-0.129 ± 0.02	3.88 ± 0.3	1.79 ± 0.15	0.357 ± 0.02	0.249 ± 0.02	0.0013
EuO # 513-2	68.878 ± 0.01	-0.14 ± 0.02	3.95 ± 0.3	1.93 ± 0.2	0.34 ± 0.02	0.267 ± 0.02	0.0023
$\text{EuO}_{0.9}\text{Sr}_{0.1}\text{O}$ # 509-4	60.685 ± 0.01	-0.23 ± 0.04	5.024 ± 0.5	2.58 ± 0.3	0.25 ± 0.02	0.209 ± 0.02	0.0036

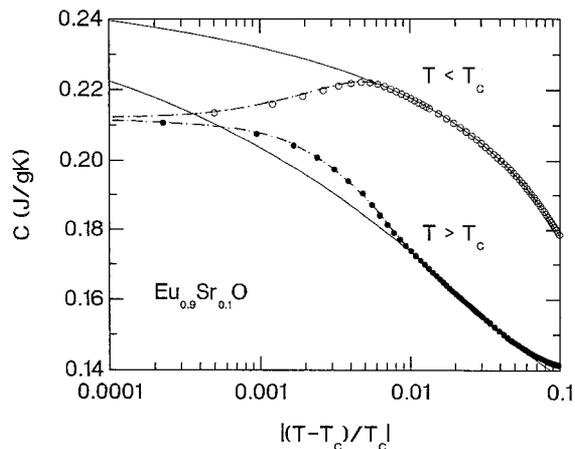


Fig. 5. Specific heat C vs. the reduced temperature $|T - T_c|/T_c$ of $\text{Eu}_{0.9}\text{Sr}_{0.1}\text{O}$ with fits after (1) (solid curves) and fits with a Gaussian distribution of transition temperatures (dash-dotted curves). The parameters are given in Table 1

nificant change of α occurs in the crossover region, in contrast to the behavior expected for the critical exponent γ of the magnetic susceptibility [20].

In the Sr-diluted samples inhomogeneities increasing with dilution severely hampered a similarly reliable determination of critical exponents. A meaningful analysis of the specific heat close enough to T_c was possible only for $\text{Eu}_{0.9}\text{Sr}_{0.1}\text{O}$. Figure 5 shows the data and fits in the same way as for EuO (Fig. 4). The resulting critical exponent for this concentration is $\alpha = -0.23 \pm 0.04$. The rounding close to T_c again can be explained remarkably well with a rounding parameter of $\delta T_c/T_c = 0.0036$ (see also Table 1).

The slight decrease of α with increasing dilution resembles the behavior found in the systems $\text{Eu}_x\text{Sr}_{1-x}\text{S}$ [11] and $\text{Eu}_x\text{Sr}_{1-x}\text{Te}$ [12] in apparent contradiction to the idea based on the Harris criterion [17] that the critical behavior should not change when introducing disorder, as long as $\alpha < 0$. However, before drawing a definite conclusion concerning the concentration dependence of the critical exponent for $\text{Eu}_x\text{Sr}_{1-x}\text{O}$, more diluted samples which are of comparable quality to the EuO crystals have to be prepared and investigated.

In conclusion, we have presented a systematic specific-heat study of the system $\text{Eu}_x\text{Sr}_{1-x}\text{O}$ for $x \geq 0.5$. The obtained phase diagram is in accordance with the expected x dependence for this system with only ferromagnetic exchange interactions. With our measurements and the careful analysis of C close to the phase transition the

existing discrepancy between the experimental and theoretically expected critical exponent could be resolved. The obtained value of $\alpha = -0.12 \pm 0.02$ is in every respect consistent with the predicted value for 3D Heisenberg systems.

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