the copper ions, which is directed along the z axis. However, Fig. 2 shows clearly that below T_c the ratio Δ_i/Δ_j for any two resonance lines is no longer constant, as it is above T_c. This experimental result necessitates an extra term in formula (1) so that below T_c

$$\Delta_i = G_i\mu_z(T) + G_i'\mu'(T),$$

where the factors G_i' are different from the G_i. It must be concluded that the function $\mu'(T)$ represents a spontaneous magnetization perpendicular to μ_z. We have repeated the experiment at other fields between 29 and 42 kOe and found the same behavior of the resonance lines, except that the critical temperature is strongly field dependent. The measured values of T_c are indicated by the drawn line in Fig. 3.

The field and temperature region defined by the the temperature independence of the resonance spectrum (shaded area in Fig. 3) is the same as that found by Haseda *et al.*,[3] whose adiabatic magnetization experiments we extended to 50 mK. The proton resonance experiments show that in this region the interpair coupling J' leads only to short-range order, probably in linear chains of spin pairs. Below 0.16 K long-range order sets in, characterized by a spontaneous component of the time-averaged magnetic moment of the Cu^{++} ion perpendicular to the external field. Comparing our results with those of Bonner *et al.*,[5] we are led to the conclusion that the interpair interaction consists of a dominant exchange in one dimension and two weaker components, just strong enough to arrange long-range order below 0.16 K.

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Critical Behavior of the Heisenberg Ferromagnets EuO and EuS†

J. Als-Nielsen* and O. W. Dietrich‡
Research Establishment Risø, Roskilde, Denmark

and

W. Kunnmann and L. Passell
Brookhaven National Laboratory, Upton, New York 11973
(Received 23 July 1971)

Neutron-scattering measurements have been made of the critical parameters of the simple Heisenberg ferromagnets EuO and EuS. Values of the critical exponents β and ν and the amplitudes of B and F describing, respectively, the reduced magnetization and the inverse correlation range (above T_c) are in good accord with theory. The measured values of the exponent γ, describing the static susceptibility, support the recent prediction that $\gamma \approx 1.40$ in a simple nearest-neighbor Heisenberg ferromagnet. The scaling relation between β, ν, and γ is fulfilled.

Although ferromagnets have been extensively studied, most are so complicated that their critical behavior cannot reliably be compared with the simple Heisenberg or Ising models for which theoretical predictions exist.[1] Among the rare exceptions are EuO and EuS. We have used neutron-scattering techniques to investigate the spontaneous magnetization below the Curie temperature T_c and the spatial critical fluctuations above T_c in these two materials, both of which are insulators with almost ideally localized Heisenberg exchange interactions. Since the Eu^{2+} ions are in the $^8S_{7/2}$ spin state and the magnetic structure of both compounds is fcc, the exchange is also nearly isotropic.[2,3] These facts, including the large spin of 7/2, make a comparison with the model calculations for the *classical* Heisenberg ferromagnet extremely relevant.

The reason for studying both compounds is that EuO has a predominantly nearest-neighbor interaction ($J_1/k = 0.75$°K, $J_2/k = -0.10$°K)[4] while EuS has a relatively larger second-nearest-neighbor interaction ($J_1/k = 0.20$°K, $J_2/k = -0.08$°K).[5] Therefore, in addition to providing experimental results on magnetically simple systems, our measurements also illustrate the sensitivity of the critical parameters to the range of the exchange interactions.

The neutron capture cross section of europium is unfortunately so large that even with our material isotopically enriched in Eu^{153}, a sample thickness less than a millimeter was necessary. To achieve sufficient cross-sectional sample area we therefore had to use a powder slab sample rather than a single crystal. Consequently, the critical scattering could only be studied at small scattering angles around the forward direction. However, this technique worked as well as the single-crystal technique (where the scattering is measured around Bragg reflections) because the scattering from both EuO and EuS is essentially isotropic within at least the inner half of the Brillouin zone. Further, when using Bragg scattering to determine magnetization, a powder is actually preferable to a single crystal because the problem of correcting for secondary extinction is eliminated.

The measurements were made with a two-axis crystal spectrometer using a monochromatic neutron wavelength of 1.46 Å. Pyrolytic graphite and quartz filters were used to reduce higher-order contamination. Corrections for the remnant contamination, which was small, were included in the data analysis. Temperatures below 30°K were determined by calibrated cryoresistors; at higher temperatures, platinum resistance thermometers were used. The critical temperature T_c was identified by the usual method of finding the temperature corresponding to the peak of the critical scattering at the smallest possible wave vector, typically $q = 0.05$ Å^{-1}. The observed values of T_c, 69.15 ± 0.05°K for EuO and 16.57 ± 0.02°K for EuS, fall within the range reported in the literature for samples of good purity and stoichiometry. Measurement of the powder diffraction patterns also showed that there were no inclusions of other europium compounds in the samples.[6]

First, let us describe the determination of the spontaneous magnetization below T_c. The magnetic Bragg scattering, which is proportional in intensity to the square of the magnetization M, is superimposed on a "background" of nuclear Bragg scattering from the rock-salt nuclear lattice structure. Because the magnetic scattering is relatively largest compared to the nuclear scattering for the (1, 1, 1) reflection, this reflection was selected for the measurements. In addition to the nuclear scattering, the magnetic inelastic scattering both from critical fluctuations and spin waves is also part of the background.

We assessed the background contributions in the following way. From a study of the *inelastic* neutron scattering in EuO and EuS, the details of which will be described in a subsequent paper, we had already determined the cross section $S(q, \omega)$ both above and below T_c. We integrated $S(q, \omega)$ over ω for those values of q which, in a randomly oriented powder, contribute to the scattering at a fixed angle in a two-axis spectrometer. This scattering accounts for the temperature variation of the intensity observed at the Bragg position immediately above T_c, and enabled us, by scaling to the two-axis data above T_c, to separate the magnetic inelastic contribution from the temperature-independent nuclear scattering (the Debye-Waller factor is almost unity even at room temperature).

The magnetic part of the Bragg intensity normalized to the extrapolated intensity at $T = 0$ is plotted on a log-log scale in Fig. 1 for both EuO and EuS. In the low-temperature region below $T/T_c = 0.5$, the deviation from saturation agrees with simple spin-wave theory.[7] Near T_c the mag-

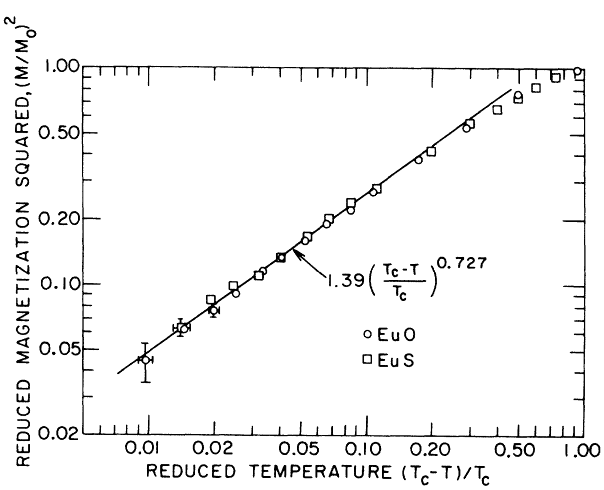

FIG. 1. The temperature dependence of the *square* of the magnetization of EuO and EuS.

TABLE I. Least-squares values for β, B, ν, F, and γ.

Expression	Quantity	EuO	EuS	Theory ($S=\infty$)
$\frac{M}{M_0}=B\left(\frac{T_c-T}{T_c}\right)^\beta$	β	0.367 ± 0.008	0.360 ± 0.012	$0.38\ \pm0.03$[a]
	B	$1.18\ \pm0.03$	$1.18\ \pm0.04$	1.12[a]
$a_{nn}\kappa_1=F\left(\frac{T-T_c}{T_c}\right)^\nu$	ν	0.690 ± 0.023	0.702 ± 0.027	0.717 ± 0.007[b]
	F	$2.4\ \pm0.2$	$2.3\ \pm0.2$	2.54[b]
$\frac{\chi(q=0)}{\chi_1}\propto\left(\frac{T-T_c}{T}\right)^{-\gamma}$	γ	1.396 ± 0.030	1.390 ± 0.039	1.405 ± 0.020[b]

[a] Ref. 8. [b] Ref. 9.

netization obeys a power law,

$$M/M_0=B[(T_c-T)/T_c]^\beta. \tag{1}$$

Least-squares–fitted values of B and β for $(T_c-T)/T_c\leq 0.11$ are given in Table I. We notice that the magnetizations behave identically in EuO and EuS and that the value of β is in good agreement with the results of a recent series expansion calculation by Stephenson and Wood.[8] The value of B is slightly larger than that obtained theoretically from the classical spin model—a reasonable result since the magnetization for $S\to\infty$ has a finite slope at $T=0$.

Above T_c, the intensity of the critical scattering was measured at small angles within the range $0.05\ \text{Å}^{-1}\leq q\leq 0.15\ \text{Å}^{-1}$ at a series of fixed temperatures $0.01\leq (T-T_c)/T_c\leq 0.5$. Critical neutron scattering arises from interactions between neutrons and critical spin fluctuations occurring in both space and time. The scattering cross section integrated over ω at a *fixed q* is proportional to the wave-vector–dependent susceptibility $\chi(q)$, which is the Fourier transform of the spatial spin-correlation function. A two-axis spectrometer performs an instrumental integration over ω, but for a *fixed scattering angle.* We have corrected our data to obtain $\chi(q)$ by using the inelasticity of the critical scattering as determined from our triple-axis spectrometer measurements following a procedure which has previously been described for iron.[10] With 2.5-Å neutrons, the correction was substantially less for EuO than for iron and almost negligible for EuS. Apart from the inelasticity we also corrected for background intensity and for the effect of the finite resolution of the spectrometer. The instrumental resolution function for a two-axis spectrometer at small scattering angles was simply determined from the horizontal and vertical angular divergences of the collimators.

At each temperature the corrected intensity data were analyzed with a Lorentzian form of $\chi(q)$,

$$\frac{\chi(q)}{\chi_1}=\frac{C}{(q/\kappa_1)^2+1}\left(\frac{T-T_c}{T}\right)^{-\gamma}, \tag{2}$$

where the inverse correlation range κ_1 is assumed to vary with temperature as $a_{nn}\kappa_1=F[(T-T_c)/T_c]^\nu$, a_{nn} being the nearest-neighbor distance. In Eq. (2), χ_1 is the susceptibility for a noninteracting spin system. This simple form of $\chi(q)$ is justified by the recent series-expansion calculations for the classical Heisenberg model made by Ferer.[11]

Figure 2 shows the best values of $a_{nn}\kappa_1$ and $\chi(q=0)/\chi_1$ for both EuO and EuS plotted on a log-log scale against the reduced temperatures $(T$

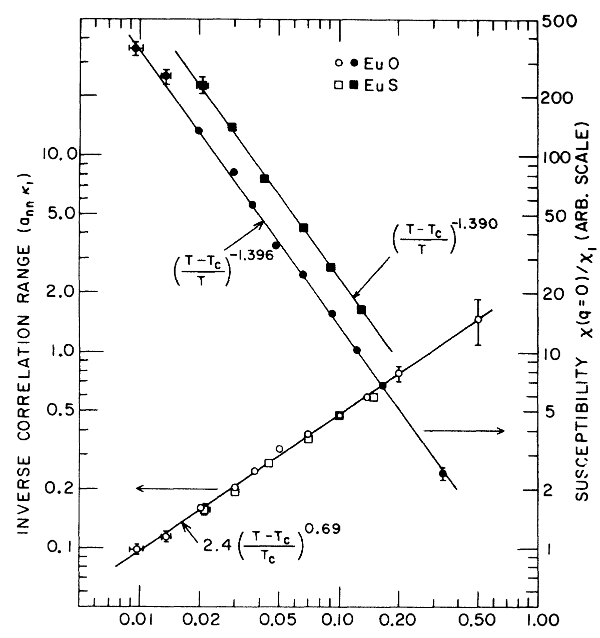

FIG. 2. The temperature dependence of the static susceptibilities $\chi(q=0)/\chi_1$ and of the inverse correlation ranges $a_{nn}\kappa_1$ (a_{nn} is the nearest-neighbor distance) of EuO and EuS. Note that the reduced temperature scales are different.

$-T_c)/T_c$ and $(T-T_c)/T$, respectively; $\chi(q=0)/\chi_1$ is in arbitrary units for the two compounds. Least-squares–fitted values of F, ν, and γ are given in Table I. The theoretical predictions for the classical Heisenberg model with nearest-neighbor interaction are also shown for comparison.[11,9] Agreement is most satisfactory.

The problem of background determination above T_c deserves some additional comment. Background scattering, while small, can be among the most important sources of systematic error in small-angle experiments because it depends on the scattering angle. An upper limit was determined by the intensities obtained at approximately $4T_c$. Part of this, however, is paramagnetic scattering, which at this high temperature is independent of q for $q \lesssim 0.15$ Å^{-1}. To estimate the paramagnetic contribution we extrapolated the power law for the temperature dependence of the intensity at $q=0$ to $4T_c$. This extrapolation is justified by the series-expansion results for $\chi(q=0)/\chi_1$ which generally give a value near 1 for C in Eq. (2). The final background curve was obtained after a few iterations. It is assumed in this procedure that the contributions from competing scattering processes, such as multiple phonon scattering, are either small or independent of temperature up to $4T_c$.

Although we are confident that this evaluation of the background gives reliable results for the critical exponents ν and γ, we do not feel that our measurements can determine a small deviation from the Lorentzian form of $\chi(q)$ such as that proposed by Fisher[1] and Ferer *et al.*[9] in terms of the small critical exponent η. According to scaling theory, $\gamma/\nu = 2-\eta$. We find $\gamma/\nu = 1.98 \pm 0.09$ for EuS and $\gamma/\nu = 2.02 \pm 0.08$ for EuO. The theoretical value of $\eta = 0.04$, proposed by Ferer *et al.*,[9] is not inconsistent with these results.

We are aware of only two other experiments which bear directly on the critical parameters of EuO and EuS. First there are the studies of EuO by Menyuk, Dwight, and Reed[12] made with a vibrating-coil magnetometer. They found $\beta = 0.368 \pm 0.005$, which compares well with our value. From their data we are also able to deduce that $B = 1.22$ by using a moment of $6.8\mu_B$ per Eu ion.[3] Again the agreement is good. Unfortunately, above T_c the agreement is less satisfactory; they find $\gamma = 1.29 \pm 0.01$, which cannot easily be reconciled either with our result $\gamma = 1.40 \pm 0.03$ or with the theoretical prediction of Ferer *et al.*,[9] $\gamma = 1.41 \pm 0.02$.

Second, there are NMR studies of EuS made by Heller and Benedek,[13] who obtained for β and B the values 0.33 ± 0.015 and 1.145 ± 0.020, respectively. Their value of B is in good agreement with our own, while their value of β is lower than ours, slightly outside the range of the statistical uncertainties. Our values for β in both EuO and EuS support the most recent theoretical expectations[8] of a value larger than $\frac{1}{3}$, originally conjectured to be the result for the Heisenberg model.

According to scaling laws, the three critical exponents β, γ, and ν which we have determined should be related through $\beta = (3\nu - \gamma)/2$. Our results are in agreement with this relation:

$$\beta - (3\nu - \gamma)/2 = \begin{cases} +0.03 \pm 0.05 & \text{for EuO} \\ +0.002 \pm 0.05 & \text{for EuS.} \end{cases}$$

It should be noted that the errors in ν and γ are correlated. This has been taken into account in the above estimates of the errors on $\beta - (3\nu - \gamma)/2$.

To summarize, (i) EuO and EuS show identical critical behavior for both the magnetization and the correlation range measured relative to the nearest-neighbor distance. Also, the critical exponents for the susceptibilities are the same. (ii) The critical behavior agrees very closely with the most recent theoretical results for the classical Heisenberg ferromagnet with nearest-neighbor interactions. (iii) The scaling relation between β, ν, and γ is fulfilled.

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