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Change from diverging to finite susceptibility below T_c of the Heisenberg ferromagnet EuS

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Approaching the coexistence line, magnetization data recorded with a superconducting quantum interference device reveal the nonlinear decay of the scaled field H conjectured by early series-expansion and renormalization-group theories. The associated divergence of the longitudinal ferromagnetic susceptibility, $\chi_L \propto H^{-1/3}$, appearing in a scaling representation of existing data, changes to $\chi_L \sim (T_c - T)^{-\gamma'}$ very close to the coexistence curve. This crossover is associated with a linear, nonscaling behavior of H, which explains $\gamma' \simeq \gamma$, reported previously also for EuO.

I. INTRODUCTION

Despite the long-standing debate on the field dependence of the magnetization near the coexistence curve of Heisenberg ferromagnets, the present situation is controversial. Classical spin-wave calculations predicted a square-root dependence of $\Delta M \equiv M(T,H) - M(T,O)$ on internal field H.[1] Close to T_c, renormalization-group (RG) work by Brezin and others[2-4] provided strong arguments that due to the divergent transverse susceptibilities (massless Goldstone modes) similar nonlinear behavior, $\Delta M \propto H^{1/p}$ with $p \simeq \frac{3}{2}$, should occur.

Experimental evidence for the most outstanding consequence of the theory, i.e., the divergence of the longitudinal susceptibility at *all* temperatures below T_c, $\chi_L(T,H) \propto H^{1/p-1}$, is still lacking. In early work on EuO (Ref. 5) a finite susceptibility, $\chi_L(T,O) \propto (T_c - T)^{-\gamma'}$ with $\gamma' \simeq \gamma = 1.3$, was derived from magnetization data employing an extended version of the kink-point method. More recently, $\chi_L(T,H)$ has been evaluated from the slope of magnetization isotherms of EuS at *finite* fields.[6] The scaling representation of this susceptibility showed a trend of $\chi_L(T_c - T)^{\gamma}$ to flatten at small scaled fields, which appeared to sustain the result for EuO. An independent search for anomalous longitudinal fluctuations at small q and H below T_c of Pd+10 at.% Fe (Ref. 7) by using polarized neutrons was unsuccessful, too. The present work investigates the magnetic equation of state of EuS below T_c with the aim to resolve the existing fundamental difference between experiment and theory.

II. EXPERIMENTAL RESULTS

The magnetization of a single-crystalline sphere has been measured by a home-made variable-temperature magnetometer[8] using superconducting quantum interference device detection (SHE-330). A double-walled chamber of Araldit F separated the sample from the He bath, while thermal contact to the carbon-glass thermometer was provided by a 99.999%-pure Cu holder. In order to minimize the background signal from Fe impurities, the Cu was subjected to 10^{-4} mbar of oxygen at 800 °C for 30 h. $M(T,H_{ext})$ scans were performed from 5.5 to 90 K in external magnetic fields between 0.1 and 6.1 kOe, frozen by a superconducting cylinder. The absolute values were determined at $T \gg T_c$ and $T \ll T_c$ by using $M = \chi(T)H_{ext}$ and $M = H_{ext}/N$, respectively, where $N = \frac{1}{3}$ is the demagnetization coefficient.

Our main results in the vicinity of the critical point, 10 K $\lesssim T \lesssim$ 23 K, are shown in Fig. 1 in terms of the scaled field $y \equiv h/m^{\delta}$ and the scaled temperature $x \equiv t/m^{1/\beta}$. The Curie temperature and $\gamma = \beta(\delta - 1) = 1.35(1)$ were obtained by a careful analysis of the low-field ($H_{ext} = 0.1$ kOe) susceptibility in agreement with a previous ac measurement at $H_{ext} = 0$.[9] The exponent of the spontaneous magnetization, $m_s = B(-t)^{\beta}$, $\beta = 0.350(5)$, was deter-

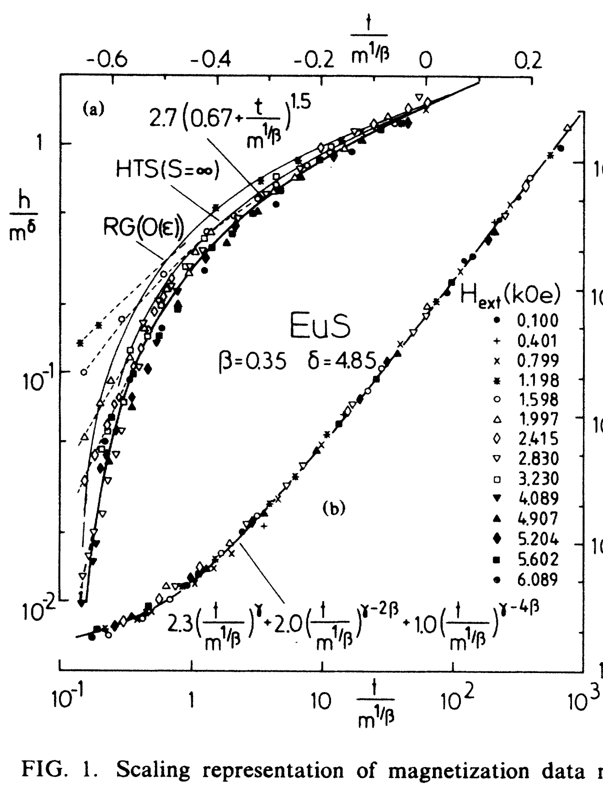

FIG. 1. Scaling representation of magnetization data measured on an EuS sphere as a function of temperature at the quoted external magnetic fields: (a) below and (b) above $T_c = 16.56$ K. Standard normalizations of M and of the internal field $H \equiv H_{ext} - NM$ to $M_0 = 15.4$ kOe and $H_c = kT_c/\mu = 35.4$ kOe have been used: $m \equiv M/M_0$, $h \equiv H/H_c$, and $t \equiv T/T_c - 1$. Model curves are explained in the text.

mined from the "best" scaling of the data (Fig. 1). This number is consistent with $\beta=0.36(1)$ evaluated by Als-Nielsen, Dietrich, and Passell[10] from neutron scattering. Close to the coexistence curve, a violation of scaling appears, which will be discussed in Sec. IV.

III. SCALING BEHAVIOR

A. Equation of state

Above T_c, the scaling function $y(x)$ can be well parametrized using Griffith's[11] expansion around the critical isochore ($x=\infty$):

$$y_\infty(x)=C_1x^\gamma+C_2x^{\gamma-2\beta}+C_3x^{\gamma-4\beta}+\dots , \tag{1}$$

as illustrated in Fig. 1(b). The coefficients $C_1=2.3(1)$, $C_2=2.0(2)$, and $C_3=1.0(3)$ are close to the corresponding values 2.5(1), 2.5(2), and 0.96(24) reported by Huang and Ho[12] for the related ferromagnet EuO. Moreover, they do not differ very much from a RG calculation to $O(\epsilon\equiv 4-d)$ for the dipolar Heisenberg ferromagnet[4] predicting $C_1=2.0$, $C_2=2.1$, and $C_3=0.9$.[13]

For the other extreme region, the same theory argued that h/m^δ vanishes nonlinearly in terms of the distance of $x=t/m^{1/\beta}$ from the coexistence line $x_0\equiv t/m_s^{1/\beta}=B^{-1/\beta}$:

$$y_0(x)=A(x_0+x)^p , \tag{2}$$

with $p>1$ (Refs. 2-4). In fact, our numerical evaluation of the explicit expressions for $y_0(x)$ published to $O(\epsilon)$ for the pure[2] and the dipolar[4] Heisenberg ferromagnets proved Eq. (2) to be valid from $x\simeq 0$ down to the experimentally inaccessible value of $x_0+x\simeq 10^{-3}$. We found the exponent $p=1.13$ for both the pure and the dipolar case.

To adjust our scaled data to the proposed power law, we used $B=1.15$ as critical amplitude for the spontaneous magnetization, which follows from the neutron results[10] if the exponent is fixed to our value, $\beta=0.35$. In fact, Fig. 1(a) demonstrates that Eq. (2) with $p=1.5(1)$ nicely describes all experimental data below T_c within their uncertainty, with the exception of the nonscaling region near the coexistence curve. We note, that the characteristic exponent is closer to the value predicted for the Gaussian model $p=1+\epsilon/2$,[2,4] than to those obtained above for the pure and dipolar Heisenberg ferromagnets. Due to the first order approximation in ϵ of the underlying RG results, the differences should not be taken too seriously. Moreover, Fig. 1(a) shows that numerical values from high-temperature series expansions for the fcc Heisenberg model with $S=\infty$, available for $|x|>0.125B^{-1/\beta}$,[14] are also fairly close to the data. A more direct impression of the good quality of the fit to the power law, is provided by Fig. 2 in which the measured isofield magnetizations are compared to the calculated values. In the scaling region ($10\ \mathrm{K}\lesssim T\lesssim T_c$), the rms of the relative deviation is smaller than 0.005 and stays below the overall experimental accuracy of about 0.008.

B. Longitudinal susceptibility

The previously published results[6] are reproduced on the scaling plot of Fig. 3 showing somewhat larger errors in the ferromagnetic region due to the high magnetizations involved. From the excellent agreement between the *paramagnetic* susceptibilities and the full curve, obtained by differentiating the corresponding equation of state, Eq. (1), we infer that $\chi_L\equiv(\delta m/\delta h)_t$ was correctly evaluated from the magnetization isotherms of Ref. 8.

For the *ferromagnetic* susceptibilities we expect according to the power-law equation of state [Eq. (2)],

$$\chi_L(t<0,h)=\frac{m}{h}\frac{\beta}{\delta\beta-px/(x_0+x)} . \tag{3}$$

Figure 3 clearly shows, that within the experimental un-

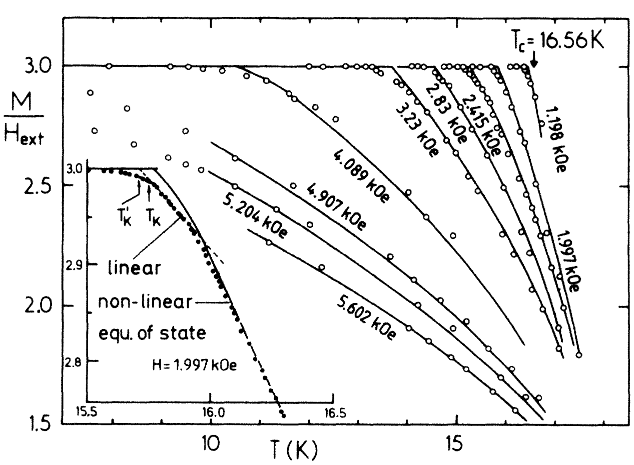

FIG. 2. Experimental M/H_{ext} ratios (random 1% sample of all data) compared to calculations (full curves) based on the power law, Eq. (2). Inset exemplifies the general shape very close to the coexistence line: nonlinear scaling followed by linear [dotted line, calculated from Eq. (6)] and rounding behavior.

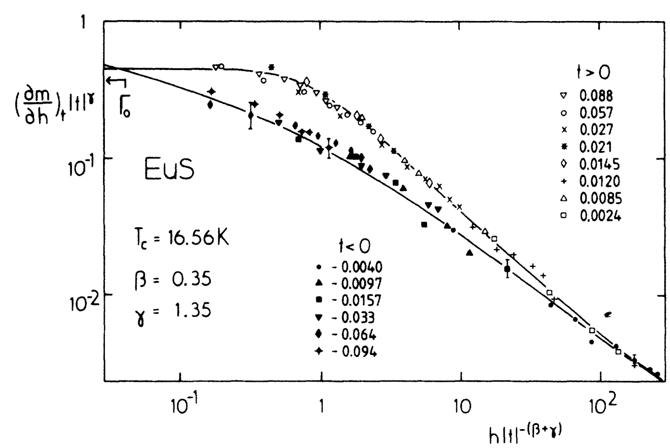

FIG. 3. Scaling representation of the paramagnetic (open) and ferromagnetic (closed symbols) susceptibilities (from Ref. 6). Full lines correspond to calculations based on the equations of state displayed in Fig. 1, Γ_0 labels the limiting behavior due to crossover to linear behavior [Eq. (7)].

certainty, the data are well described by Eq. (3). Of particular interest is the vicinity of the coexistence line, $x \rightarrow -x_0$, where the second term in the denominator of Eq. (3) dominates the first one, so that χ_L reduces to

$$\chi_L(t<0,h\rightarrow 0)=\frac{\beta x_0}{pA^{1/p}}\frac{m^{1-\delta/p}}{h^{1-1/p}} . \quad (4)$$

Because of the finite magnetization $m \rightarrow m_s$, χ_L is expected to diverge proportional to $h^{1/p-1}$. To see whether this prediction is borne out by our scaled data we put χ_L into the scaling form

$$\chi_L(t<0,h\rightarrow 0)=\Gamma_-(-t)^{-\gamma}\left(\frac{h}{t^{\beta+\gamma}}\right)^{1/p-1} , \quad (5)$$

with $\Gamma_- = (B^{1-\beta(\delta-p)}/A)^{1/p}\beta/p$. Obviously, $\chi_L(-t)^\gamma$ exhibits the same divergence in terms of the scaled field as does $\chi_L(h)$. In fact, the ferromagnetic susceptibilities, calculated from Eq. (3) for EuS, display this power law below $h/|t|^{\beta+\gamma} \simeq 0.1$. The trend of the data to this behavior (Fig. 3) is to our knowledge the first indication for the divergence of χ_L.

IV. LINEAR REGION

From the scaling plot in Fig. 1(a), one notices that in the vicinity of the coexistence curve (dashed lines) the isofield (H_{ext} = const) magnetization curves can be approximately described by an empirical equation of state that is linear in x ($\simeq -x_0$):

$$y_1(x,H_{ext})=C(x_0+x)+\left(\frac{H_a}{H_{ext}}\right)^{1/\beta} , \quad (6)$$

with $C=0.8(2)$. $H_a=0.60(5)$ kOe may be considered as an effective anisotropy field causing the deviations from the Heisenberg scaling function, Eq. (2), to be discussed below.

As the most important consequence of the linear term in $y(x)$ one expects from Eq. (5) a crossover to a finite susceptibility at the coexistence curve, $\chi_L \propto (-t)^{-\gamma'}$, with $\gamma'=\gamma$. More precisely, Eq. (6) yields for $x \rightarrow -x_0$,

$$\chi_L(t<0,h=0)=\Gamma_a(-t)^{-\gamma} . \quad (7)$$

Taking $H_{ext}=NM_s(T)$ near the phase boundary, the amplitude turns out to depend on temperature, $\Gamma_a=\Gamma_0/(1-t_a/t)$, with $\Gamma_0 \equiv \beta B^{(1-\gamma)/\beta}/C$, while $t_a \equiv \delta\beta(H_a/NM_0)^{1/\beta}/C$ is affected by H_a.

Actually we found such "apparent" critical behavior using the thermodynamic relation,

$$\chi_L(t,H=0)=\frac{\chi_0}{N}\left[\left(\frac{\partial M}{\partial T}\right)_{H=0}\left(\frac{\partial M}{\partial T}\right)^{-1}_{H_{ext}\approx NM_s}-1\right] \quad (8)$$

($\chi_0=M_0/H_0$ due to the normalization). As is illustrated by the inset to Fig. 2 there are well-defined slopes of the measuring curves $(\partial M/\partial T)_{H_{ext}}$ in the small linear region. At $(-t)>0.01$, the resulting susceptibility can be described by the power law $\chi_L=0.33(5)(-t)^{-1.29(5)}$ showing some tendency to flatten closer to T_c. This is fully consistent with the predictions of Eq. (7) yielding for the amplitude $\Gamma_0=0.38(9)$ and for the anisotropy parameter $t_a=0.0046$. Moreover, as seen in Fig. 3 this behavior seems to limit the increase of the scaled susceptibilities.

By the same procedure, Høg and Johansson determined $\chi_L(t,0)$ parallel to the easy [111] axis of EuO and obtained similar behavior: the power-law equation (7) with $\gamma'=1.3(1)$ and $\Gamma_a=0.3(1)$ and the trend to flatten below $|t|=0.01$.[5] Taking $\gamma'=\gamma$, they interpreted their results within the linear model by Schofield[15] in terms of uniform Heisenberg scaling. In addition to the theoretical proviso against the applicability of the Schofield model for non-Ising systems,[2] the present experimental findings on the related ferromagnet EuS suggest that one check whether the finite zero-field susceptibility of EuO below T_c also can be traced to a crossover to linear behavior $y \sim (x_0+x)$ near the coexistence line. It should perhaps be pointed out, that according to Eq. (7a), the presence of the (scaling-violating) anisotropy field H_a is not a necessary condition for the occurrence of the power-law equation (7).

Another consequence of our findings near the phase boundary is associated with the so-called kink-point method, widely used to determine the spontaneous magnetization at the kink-point temperature T_k by extrapolation of experimental $M(T,H_{ext}=\text{const})$ curves to $H_{ext}/N=M_s(T_k)$ (see, e.g., Ref. 16). Obviously, this method relies on the existence of a constant finite slope $(\partial M/\partial T)_{H_{ext}}$ close to T_k, which for an ideal Heisenberg ferromagnet with $\chi_L \sim H^{-1+1/p}$ is not available, as follows, e.g., from the thermodynamic relation, Eq. (8). Therefore, if the kink-point analysis has identified a truly linear region, i.e. $(\partial M/\partial T)_{H_{ext}}$ = const, on some Heisenberg ferromagnet, this may indicate a crossover to linear behavior of $y(x)$ and to a finite susceptibility at $H=0$. We should stress, that in this case the extrapolation is leading to an apparent kink-point $T_k' < T_k$ and, therefore, to erroneous values for $M_s(T)$. For EuS, this is illustrated by the inset to Fig. 2.

This inset also shows that for $H_{ext}/M - N \lesssim 0.002$, i.e., around T_k' extremely close to the coexistence curve, a new nonlinear region appears. Since these data do not obey a uniform power law in H and, moreover, the inverse zero-field susceptibility $(\partial H_{ext}/\partial M - N)$ starts to fall off from $t^{-\gamma}$ at just about the same value, 0.002 (corresponding to $t \approx 0.003$), we tentatively associate this behavior with "rounding" effects. It seems that it is a general feature of real Heisenberg and Ising ferromagnets (see, for example, Ref. 16) related to random anisotropies, like deviations from ellipsoidal sample shape, grain boundaries, dislocations, and other imperfections. However, detailed information on the effect near the coexistence curve is not known to us. Thus, we cannot exclude that linear (Ising-like) behavior also arises from these local anisotropy fields which, as soon as H decreases to their size, first freeze the transverse (Goldstone) modes, M/H, before in the rounding region, the remaining divergence of the longitudinal susceptibility, $\partial M/\partial H$ is affected.

On the other hand, attempts to explain the linearity of $y(x)$ and the associated finite susceptibilities of EuS (and also EuO) should also consider the small single-ion anisotropy, $\varepsilon_A \approx 0.03k_BT_c$, following, for example, from Ref. 17. Together with magnetostriction and exchange striction (unknown for EuS), ε_A may give rise to rhombohedrally (i.e., parallel to the easy ⟨111⟩ directions) distorted crystallographic domains, the uniaxial anisotropy fields of which prevent the transverse modes from divergence as soon as they become larger than the internal field H.

Very recently, Chudnovsky, Saslow, and Serota[18] predicted a linear behavior of the low-field and low-temperature ($T \ll T_c$) magnetization $M = \chi H$ for Heisenberg ferromagnets with random and coherent anisotropies as well. Following their suggestion to extend the results to the critical region, we find for the susceptibility $\chi \sim M_s^{-4\lambda} \sim (-t)^{-4\beta\lambda}$. Identifying this exponent with our result $\gamma = 1.29(5)$, we obtain $\lambda \approx 1$. By the definition, $\lambda = p(p+1)/2 - 2$,[18] this corresponds to a ($p = 2$)-fold axis of the anisotropy which cannot arise from the (coherent) magnetostriction considered above.

V. SUMMARY

The scaling analysis of the magnetic equation of state confirms the critical exponents $\beta = 0.35$ and $\gamma = 1.35$ obtained from previous neutron work[10] and paramagnetic susceptibility measurements.[9] Below T_c, the internal magnetic field disappears nonlinearly, as a function of the distance from the coexistence curve, i.e., $H \propto (M^{1/\beta} - M_s^{1/\beta})^p$ in fair agreement with numerical results of high-temperature series[14] and renormalization-group[2-4] expansions. An indication for the divergence of the ferromagnetic susceptibility, $\chi_L \sim H^{1/p-1}$, implied by the nonlinear equation of state has been detected by a scaling analysis of existing data. Close to the phase boundary, a crossover of unidentified origin to linear, non-Heisenberg behavior is observed, which prevents χ_L from further divergence. The apparent zero-field susceptibility $\chi_L \sim (T_c - T)^{-\gamma'}$, with $\gamma = \gamma'$ also reported for EuO (Ref. 5) could be explained by an empirical, linear equation of state. We have shown that both nonlinear and linear behavior make an accurate application of the conventional kink-point method impossible.

Further systematic work, combining local methods (NMR, Mössbauer, perturbed angular correlation), neutron-scattering and bulk magnetization measurements on well-defined samples seems to be necessary to establish the magnetic behavior, in particular the divergence of χ_L and its saturation near the phase boundary of Heisenberg ferromagnets.

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