# Critical exponents, amplitudes, and correction to scaling in nickel measured by neutron depolarization

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The magnetic induction B was measured with a precision better than  $2 \times 10^{-4}$  T around the phase transition in a temperature region from  $T_c - 20$  K to  $T_c + 15$  K in fields below 1550 A/m. An asymptotic value equal to 0.390(3) was found for the critical exponent  $\beta$ . An effective critical exponent  $\gamma_{\text{eff}} = 1.315(15)$  is given in the range  $5 \times 10^{-4} \le t \le 2 \times 10^{-3}$ . Four critical amplitudes and one correction to the scaling amplitude are reported.

## I. INTRODUCTION

The ferromagnetic-paramagnetic phase transition in nickel has been studied by several authors since the first measurement of the magnetic induction *B* around the critical temperature  $T_c$  by Weiss and Forrer in 1926.<sup>1-11</sup> Critical exponents derived on the one hand by fitting experimental data to asymptotic power laws were assumed to be valid near  $T_c$  along suitable thermodynamic paths. On the other hand, the analysis of (B,H,T) data (*H* is the magnetic field, *T* is the temperature) to "scaled equations of state" have been performed to yield the critical exponents. The reported values for the exponents  $\beta$  and  $\gamma$ , defined by

$$B_s(T) \approx t^{\beta}, \quad \chi \approx t^{-\gamma} \tag{1}$$

(B<sub>s</sub> is the spontaneous magnetic induction,  $\chi$  the susceptibility, and  $t = |T - T_c| / T_c$  is the reduced temperature), are in most cases located around 0.37–0.39 and 1.28–1.35, respectively. These values deviate slightly from  $\beta$ =0.365 and  $\gamma$ =1.387 calculated for a three-dimensional (3D) Heisenberg system.<sup>12</sup>

Since the experimental data are outside the asymptotic critical region, correction-to-scaling terms should be considered in the analysis.<sup>13,14</sup> The existence of correction terms has been seen experimentally in the phase transitions of fluids.<sup>15</sup> The observation of these small correction terms in magnetic systems is hampered by the fact that most experimental methods for measuring  $\beta$  are not precise enough. Only Mössbauer experiments in iron gave some information about the correction term.<sup>16</sup> However, the reported universal  $\beta$  and  $\beta_{\text{eff}}$ 's show some inconsistencies.<sup>17</sup>

Recently, it was demonstrated that the neutron depolarization technique, applied to ring-shaped samples with vanishingly small demagnetization factors, is a very sensitive method for measuring *B* in ferromagnets.<sup>17</sup> In former neutron depolarization experiments on nickel, a value for the exponent  $\beta$  could be derived from the measured depolarization  $B_s^{2\delta}$  ( $B_s$  is the spontaneous magnetic induction) within 1 degree close to  $T_c$ , where the mean domain size  $\delta$  was shown to be constant in that temperature range.<sup>18,19</sup> The analysis of the field-dependent measurements was hampered by the depolarization caused by demagnetizing fields.<sup>20</sup> The high resolution in *B* below  $2 \times 10^{-4}$  T, achieved in the experiments presented here, permits the measurement of *B* in the close vicinity of  $T_c$  independent of the external field *H*. The fields applied are on the order of a few hundred A/m.

In this paper we aim to determine the asymptotic value of the exponent  $\beta$  and to give a close estimate of the correction terms to scaling in the asymptotic power law which describes the temperature dependence of the spontaneous magnetic induction  $B_s$  just below  $T_c$ .

Secondly, we aim to check the increase of the exponent  $\gamma_{\rm eff}$  while approaching  $T_c$ , observed from analyzed data of a "perturbed angular correlation" experiment within  $t \leq 4 \times 10^{-3.9}$  The latter experiment gives, so far, the only data available in this small reduced temperature range just above  $T_c$ .

Thirdly, the lack of experimentally determined values for critical amplitudes, which are needed to test predicted universal amplitude ratios, has motivated us to derive amplitudes from our data without using any equation-ofstate analysis.

#### **II. EXPERIMENT**

The neutron depolarization experiments were performed on a pure (0.99997) polycrystalline nickel ring (outer diameter of 16 mm, inner diameter of 10 mm, d=3 mm), which had been annealed for 60 h at 1050 K to reduce present internal stresses.

The magnetic induction B was derived from the total angle of Larmor precession  $\varphi_t$  that a polarized neutron beam experiences during traversal of the sample. Figure 1(a) shows the geometry of the neutron depolarization set up used.<sup>18</sup> The magnetic induction B is given by  $B = \varphi_t/cd$  with  $c = \gamma/v$ , where  $\gamma$  is the gyromagnetic ratio of the neutron, v is the velocity of neutrons. The angle  $\varphi_t$  is determined from the directly measured x and z components  $D_{xx}$  and  $D_{zx}$ , respectively, of an incident beam polarized parallel to the x axis by

$$\varphi_t = \tan^{-1}\left(\frac{D_{zx}}{D_{xx}}\right) + 2\pi n$$
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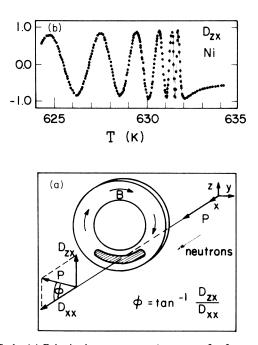


FIG. 1. (a) Principal geometry and system of reference in the neutron depolarization setup. The shaded area indicates that part of the ring traversed by the neutrons. **P** is the polarization vector of the neutron beam. (b) Example of a measured depolarization element  $D_{zx}$  versus temperature. The physical meaning of  $D_{zx}$  is visible in Fig. 1(a).

*n* counts the multiple rotations of the polarization vector **P** [see Fig. 1(b)]. The *B* resolution achieved in our experiments on nickel was better than  $2 \times 10^{-4}$  T.

The temperature of the sample was measured within 0.01 K. The long-term drifts in temperature were below 0.025 K/24 h. More details are reported elsewhere.<sup>17</sup> The maximal field strength that could be applied was about 1550 A/m. Stray fields at the sample position were below 10 A/m. Measuring series at a given field strength were performed by continuously decreasing and successively increasing the temperature with a constant change of T versus time. Each data point B(T) equals an averaged value over a temperature interval  $\Delta T$ , determined by the chosen temperature change with time and the required resolution in B. The latter is determined by the counting statistics.

The quantity  $\Delta T$  amounts from 0.01 to 0.05 K in our measuring series. Typical cooling (heating) rates in a series were about a few 0.01 K/min. All data points of the decreasing and increasing temperature run were consistent within 0.02 K.

## III. EXPERIMENTAL RESULTS AND DATA ANALYSIS

## A. Critical temperature

The magnetic induction B was measured at different field strengths between H=1 and 1550 A/m in a temperature region from  $T_c-20$  K to  $T_c+15$  K. In the measurements with  $H \le 20$  A/m the point of inflection in

B(T) could be determined within 0.01 K and it was assumed to correspond to the value of  $T_c$ . Figure 2 shows a measurement at H=8 A/m. Adjacent data points in that measurement differ by about 0.01 K in temperature. In larger fields the phase transition smears out and the "definition" of  $T_c$  becomes more difficult. One way to obtain  $T_c$  is to fit the data both below and above the phase transition to asymptotic power laws (1) with  $T_c$  as an adjustable parameter. The comparison of the  $T_c$ 's obtained from such fits gives some indication of the precision in the value of  $T_c$ . This standard method was applied by us to neutron depolarization data for iron.<sup>17</sup> However, as is well known, slightly different values of  $T_c$  give fits of nearly the same quality with somewhat different values of the critical exponent and amplitude. To avoid this problem measurements of B(H) along isotherms were performed at a few temperatures around  $T_c$  (Fig. 3). In these experiments a magnetic field of triangular shape in time with a frequency of one cycle per sec was applied periodically to the sample and the neutron intensity, which determines  $D_{xx}$  and  $D_{zx}$ , was counted in time channels synchronized to the applied field.<sup>21</sup> This sort of measurement will be referred to as "quasistatic". It will be noted that each data point B(H) represents an average value in an interval  $\Delta H$ . In this way the temperature in the experiments with larger fields was calibrated to those in lower fields and  $T_c$  could be determined with an error below  $\pm 0.01$  K in all the measurements up to 1550 A/m.

#### B. Paramagnetic phase

The singularity of the initial susceptibility above  $T_c$  is characterized by the exponent  $\gamma$ :

$$\chi = G^+ t^{-\gamma_{\rm eff}} , \qquad (2)$$

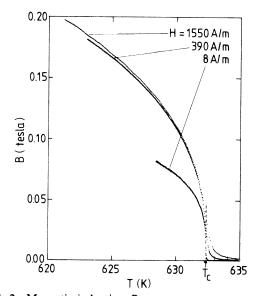


FIG. 2. Magnetic induction B versus temperature measured at different field strengths. The arrow indicates  $T_c$  and was determined from the inflection point in the measuring series at H=8 A/m.

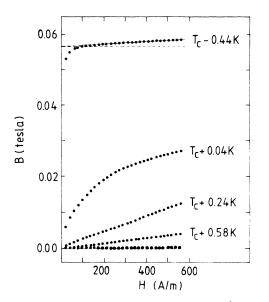


FIG. 3. Magnetic induction B versus magnetic field H at different temperatures around  $T_c$ .

where  $G^+$  is a critical amplitude. The susceptibility is defined by  $\chi = \partial m/\partial h$  with *m* identical to the magnetic induction normalized to the spontaneous magnetic induction  $B_s$  at zero temperature  $[B_s(T=0)=0.65 \text{ T}]$ , and *h* identical to the magnetic field *H* normalized to  $k_B T_c / n_B = 1.21 \times 10^9 \text{ A/m}$  for nickel  $(n_B$  is the magnetic moment per atom).

First, isotherms B(H) were measured at different temperatures close to  $T_c$  (Fig. 3). From these measurements a lower bound in the reduced temperature of about  $5 \times 10^{-4}$  was found, above which *m* is linearly dependent on *h* for  $H \le 1550$  A/m and *m/h* is a good measure of the *initial* susceptibility. Least-squares fits to Eq. (2) were performed on the data in the reduced temperature range  $5 \times 10^{-4}$  to  $2 \times 10^{-3}$  at a few field strengths *H* between 390 and 1550 A/m. The upper bound  $t=2 \times 10^{-3}$  in temperature and the minimum field strength of 390 A/m were chosen to keep the relative error in *m* below a few percent.

All fits within the above T region yielded values for  $\gamma_{eff}$  between 1.30 and 1.34 with a mean value about 1.315. The quantity  $G_{eff}^{+}$  amounts to 1.40(3).

#### C. Ferromagnetic phase

Including the first-order correction to the scaling, the temperature dependence of the ordering parameter m near the phase transition is given by

$$m = bt^{\beta}(1 + at^{\Delta}) , \qquad (3)$$

with b,a and  $\beta,\Delta$  as two critical amplitudes and two critical exponents, respectively. Effective critical exponents obtained by fits of data B(T) in a finite T region near  $T_c$  to a simple power law

$$m = b_{\rm eff} t^{\rho_{\rm eff}} \tag{4}$$

are related to the asymptotic value  $\beta$  by

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$$\beta_{\rm eff} = \beta + a \,\Delta \bar{t}^{\,\Delta} \,, \tag{5}$$

with t a mean reduced temperature of the covered T region.

To derive the spontaneous magnetic induction  $B_s(T)$ from the measured B(T,H) one has to consider two corrections. First, a field induced induction B of paramagnetic origin has to be taken into account. Secondly, the presence of magnetic anisotropy causes the measured B to be below  $B_s$ , with B approaching  $B_s$  only in fields much larger than these anisotropy fields.

From the constant slope of B versus the external field plot at  $H \ge 150$  A/m, with the data along the  $(T_c - 0.44$ K) isothem (Fig. 3), we estimate a critical amplitude  $G_{\text{eff}} \sim 0.41(4)$ . The critical exponent  $\gamma$  below  $T_c$  was assumed to be identical with the one found above  $T_c$ . The measured m was corrected for this paraeffect by subtracting  $G_{\text{eff}}t^{-\gamma}h$ . Despite this correction, the data used to fit Eq. (4) were limited to those obtained below  $T_c - 0.7$  K  $(t=1.1\times10^{-3})$  in order to restrict the correction in B below 2% at the maximum temperature in the highest external field used. Moreover B(H) looses its linearity with H upon approaching  $T_c$  so that the applied correction becomes invalid.

The strength of anisotropy fields present was derived from a study of the law of "approach to saturation" and will be discussed in detail elsewhere.<sup>22</sup> In this way we could extrapolate the measured B at the different field strength to obtain a value of  $B_s$  with an estimated error below 2% at the lowest temperature. It is evident from Fig. 2 that B(T,H=390 A/m) is already close to saturation ( $\Delta B/B_s \leq 2\%$ ) above  $T_c - 12$  K. Within 1 K below  $T_c$ , the anisotropy fields are much smaller than 150 A/m, and this justifies attributing the change in B with H at  $H \geq 150$  A/m to paraeffects. On the other hand, paraeffects are negligible below  $T_c - 2$  K at fields below 550 A/m, and B(H) is dominated by anisotropy fields.

The data from different measuring series with field strengths between 390 and 1550 A/m were fitted to Eq. (5). The least-squares fits were performed in different Tregions to obtain the change of  $\beta_{eff}$  with  $\overline{t}$  [Fig. 4(a)]. It is evident that  $\beta_{\rm eff}$  increases on approaching  $T_c$ . The rather similar change of  $\beta_{eff}$  with  $\overline{t}$  analyzed from measuring series at different field strengths, gives support to the proper choice of the applied corrections to B. The best least-squares fit of  $\beta_{eff}(\bar{t})$  to Eq. (5) yields an asymptotic  $\beta$ of 0.390(3) and a correction to the scaling amplitude a = -0.42(4). The errors in  $\beta$  and in the quantity result from the uncertainty in  $T_c$ , possible errors in the applied corrections to B, and the standard deviation obtained in the fit. The value  $\Delta$  was kept at 0.55 calculated for the three-dimensional Heisenberg system.<sup>12</sup> The change of  $\Delta$ with the dimension of the order parameter is small for three-dimensional systems<sup>12</sup> and the effect on the value of  $\beta$  is within the quoted error.

The asymptotic value of b was derived from<sup>17</sup>

$$b = b_{\rm eff} \left\langle \frac{t a \,\Delta \overline{t}^{\,\Delta}}{1 + \alpha t^{\,\Delta}} \right\rangle_{\rm av} \tag{6}$$

to be 1.52(2) [see Fig. 4(b)].  $\langle \rangle_{av}$  denotes an averaging in the reduced temperature range under investigation.

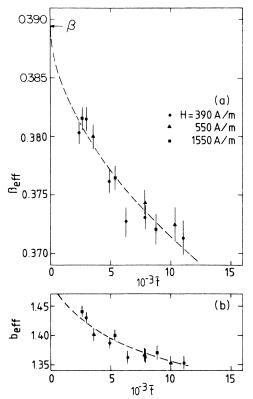


FIG. 4. (a) Effective critical exponent  $\beta_{eff}$  versus reduced temperature  $\overline{t}$  (see text). The errors in the data account for the uncertainty in  $T_c$  and possible drifts in temperature. [The same is valid in Fig. 4(b)]. Dashed lines denote the result of least-squares fit to Eq. (5). (b) Effective amplitude  $b_{eff}$  versus  $\overline{t}$ . The symbols have the same meaning as in Fig. 4(a). Dashed lines denote the result of least-squares fit to Eq. (6) with fixed parameters  $\Delta = 0.55$ , a = -0.415, and only one fittable parameter b.

### D. Data near the critical isotherm

On the critical isotherm, h(m) is given by

$$h = Dm \mid m \mid^{1-\delta}, \tag{7}$$

with D and  $\delta$  as a critical amplitude and critical exponent, respectively. The critical exponent can be calculated from the found  $\beta$  and  $\gamma$  to be 4.38(6), using the scaling relation  $\delta = 1 + \gamma/\beta$ . Data of m and h with  $t \le 1.5 \times 10^{-5}$  were obtained from the temperature series. From these data a value of D=0.32(10) was estimated for  $H \ge 390$  A/m. The error is caused by the uncertainty in both  $\delta$  as well as in  $T_c$ . To get reliable data of h(m) on the critical isotherm by a "quasistatic" measurement was impossible because of small temperature drifts during the measuring time of a few hours needed to perform such a measurement.

The data above  $T_c + 0.06$  K, including those outside the "linear" h(m) region, were transformed to the reduced quantities  $\tilde{m} = m/t^{\beta}$  and  $\tilde{h} = h/t^{(\beta+\gamma)}$ . The nearly linear relation between  $\tilde{m}^2$  and  $\tilde{h}/\tilde{m}$  (Fig. 5) shows that the scaling function is well approximated by  $\tilde{h} = f(\tilde{m}) = c_1 \tilde{m} + c_2 \tilde{m}^3$ , with  $c_1 = 0.72(3)$  and  $c_2$ = 0.92(10). It will be noted that in comparison with data obtained at fields about 1000 times larger,<sup>4</sup> the value of  $c_1$ 

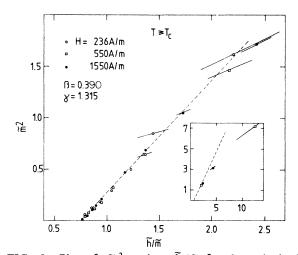


FIG. 5. Plot of  $\tilde{m}^2$  against  $\tilde{h}/\tilde{m}$  for data obtained in measuring series at three different field strengths.  $\tilde{m} = m/t^{\beta}$  and  $\tilde{h} = h/t^{\beta+\gamma}$  are the reduced order parameter and the reduced field. The error bars of data points indicate the effect of a shift of 5 mK in the value of  $T_c$ .

agrees quite well while the value of  $c_2$  found here is about a factor of 2 larger. Data were used from measuring series at *H* above 236 A/m. Data obtained at smaller fields were excluded because *m* can be measured with a small relative error only very close to  $T_c$  at these small field strengths and the reduced quantities  $\tilde{m}$  and  $\tilde{h}$  become very sensitive to the choice of  $T_c$  in that case.

# **IV. SUMMARY AND DISCUSSION**

All critical quantities obtained in this study are summarized in Table I and compared with theoretical calculations for the 3D Heisenberg system, which is assumed to be the universality class that describes best the critical behavior in nickel.

The value of 0.390(3) found for  $\beta$  in nickel is well above that calculated for the 3D Heisenberg system, 0.365(1).<sup>12</sup> The effective exponents  $\beta_{eff}$ 's found in our study are in agreement with most values obtained from the analysis of other data in corresponding temperature ranges.<sup>6,7,8,11</sup> Our analysis shows that correction terms to the scaling are present. These correction terms cause the experimentally found  $\beta_{eff}$ 's to be below the asymptotic value  $\beta$  in the case of nickel.

Due to the small absolute values of B above  $T_c$  for fields  $H \le 1550$  A/m, the exponent  $\gamma$  could only be derived with a relatively large uncertainty. The possibility of obtaining data in very small external fields makes an extrapolation of m(h) data to zero field unnecessary for yielding a measure of the initial susceptibility.<sup>10,11</sup> The value for  $\gamma_{eff} = 1.315(15)$  is based upon data within a reduced temperature range  $t \le 2 \times 10^{-3}$ , which was accessible up to now only by a perturbed angular correlation experiment in the small-field region.<sup>9</sup> The data of the latter experiment indicated an increase of  $\gamma_{eff}$  upon approaching  $T_c$  at  $t \le 4 \times 10^{-3}$  to perhaps a value of 1.387(1) for the 3D Heisenberg system. Our data do not corroborate this effect, and the value of 1.315 is within the limits of error, in excellent agreement with the analyses of data in a wider

	This work	Theory	Results of scaled equation of state analysis on nickel taken from Ref. 21	
			Linear mode	Modified MLSG
β	0.390(4)	0.365(1) <sup>a</sup>	0.38	0.38
γ	1.315(15)	1.387(1) <sup>a</sup>	1.35	1.33
b	1.52(2)		1.5	1.4
а	-0.42(4)			
G +	1.40(3)		1.5	1.3
G -	0.41(4)		0.38	1.1
D	0.32(10)		0.29	0.29
$G^+b^{\delta-1}D$	1.8(5)	1.23,1.33 <sup>b</sup>	1.7	1.4
$\frac{G^+b^{\delta-1}D}{G^+/G^-}$	3.4(4)	0°	3.9	1.3

TABLE I. Critical exponents, amplitudes, and amplitude ratios in nickel. Explanations and definitions of the symbols used are given in the text. All amplitudes are based on a magnetic induction normalized to 0.65 T and a magnetic field normalized to  $1.21 \times 10^9$  A/m.

\*Reference 12.

<sup>b</sup>First value is calculated from a high-temperature series; second value by  $\epsilon$  expansion (Ref. 21). <sup>c</sup>Reference 23.

temperature region and at much larger fields  $(\langle \gamma \rangle \approx 1.29 - 1.35)$ .<sup>2,4,8,10,11</sup>

Using zero-order results for universal ratios of correction amplitudes,<sup>14</sup> a correction amplitude of about -0.5can be estimated which results in a difference of 0.006 between  $\gamma_{\text{eff}}$  at  $\overline{t} = 1 \times 10^{-3}$  and the asymptotic value and, hence, cannot be resolved within the low-field region.

The four derived critical amplitudes b,  $G^+$ ,  $G^-$ , and Dallow one to calculate two universal ratios  $G^+/G^-$  and  $R_{\chi}=G^+b^{\delta-1}D$ . The values quoted in Table I agree well with those obtained by a scaled equation-of-state analysis of the Weiss-Forrer data for the linear model, however, they depart from the values obtained by the application of a modified Missori, Levelt Sengers, and Green (MLSG) equation of state to the Weiss-Forrer data.<sup>23,24</sup>

The calculated values of  $R_{\chi} = 1.23$  (high-temperature series) and 1.33 ( $\epsilon$  expansion) do not contradict the experimental value of 1.8(5). The amplitude ratio  $G^+/G^-$  is theoretically predicted to be zero for systems with  $n \ge 2$ 

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(*n* is the dimension of the order parameter), but the ratio found experimentally is significantly different from zero; this may be due to the effect of dipolar interactions.<sup>25</sup>

# **V. CONCLUSIONS**

The (B,H,T) data presented have a higher precision than those reported by other methods so far and moreover, they are located closer to the asymptotic region. The accessibility of data in the region of low fields allows one to get reliable data of the initial susceptibility without applying nonlinear extrapolations to zero field.

Our experimental data give clear evidence for the existence of a correction term to the simple power law  $m = bt^{\beta}$  close to  $T_c$  in nickel. An increase of  $\gamma_{\text{eff}}$  to the 3D Heisenberg value with  $T \rightarrow T_c$  is not observed.

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