Determination of the critical exponents β and γ in iron by neutron depolarization

N. Stüsser, M. Th. Rekveldt, and T. Spruijt
Interuniversitair Reactor Instituut, 2629 JB Delft, The Netherlands
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By measuring the total angle of the Larmor precession that a polarized neutron beam experiences during the perpendicular transmission of a magnetically saturated ring of polycrystalline iron, the magnetic induction B can be determined with a precision of $1-2\times10^{-4}$ T. Measurements have been carried out in a reduced temperature range $t=5\times10^{-4}$ to 5×10^{-3} and an effective critical exponent $\beta_{\text{eff}}=0.363\pm0.004$ has been derived. The paramagnetic region just above T_c has been studied at very low fields of about 100 A/m. The critical exponent $\gamma=1.33\pm0.02$ is consistent with the results obtained by other techniques in much larger fields.

I. INTRODUCTION

Renormalization-group methods predict very accurately values of the universal critical exponents which relate thermodynamic quantities at phase transitions.[1] To test the predicted values one has to measure the thermodynamic quantities with a high degree of precision in the close vicinity of the phase transition. Measurements carried out in scaling fields (i.e., reduced temperature, the effective magnetic field in a ferromagnet, etc.), which deviate significantly from zero should involve a "correction to scaling" in their analysis.[2,3]

The main purpose of this paper is to demonstrate the possibilities of using the neutron depolarization technique to study the static critical behavior of a ferromagnet. The measurements presented here have been performed on pure iron (0.999975% pure), whose critical behavior is believed to be described by a three-dimensional Heisenberg model. With neutron depolarization, the magnetic induction can be resolved within 1 to 2×10^{-4} T, which is considered to be equal to the precision.

A neutron depolarization analysis has been carried out already on a disk-shaped single crystal of iron around T_c.[4] The demagnetizing field of that sample, however, prevented a quantitative analysis of the critical behavior. The measurements presented here were carried out on an annular polycrystalline iron ring, where demagnetizing fields are very small and hence can be neglected.

II. EXPERIMENTAL DETAILS

A neutron beam with a wavelength of $\lambda=1.6$ Å and a degree of polarization $P=0.91$ can be obtained by Bragg reflection in a magnetized Fe_3Si single crystal. The polarization $\mathbf{P}$ can be adjusted parallel to any one of the three orthogonal directions x, y, z (Fig. 1) by means of a polarization turner consisting of two coils wound perpendicularly with respect to each other.[5,6] In the polarization turner a homogeneous field is built up with a well-defined magnitude and direction, thus causing the polarization $\mathbf{P}$ to rotate by Larmor precession to the desired x, y, or z direction. During transmission through the ferromagnetic sample, which is positioned inside a soft iron box to exclude external magnetic fields, the neutron beam polarization changes by Larmor precession around the local magnetization.

The depolarization, i.e., the reduction in the degree of polarization and the change in the direction of $\mathbf{P}$ relative to the incoming beam, is obtained from a successive polarization analysis along the three orthogonal directions. The analyzer (which consists of a second polarization turner plus a magnetized Fe_3Si single crystal) mirrors the polarizer. Hence, a three-dimensional depolarization analysis delivers a (3×3) matrix $\underset{\sim}{D}$ defined by

$$\mathbf{P}_{fj}=D_{ji}\mathbf{P}_{0i}\ , \tag{1}$$

where i and j refer to the principal axes x, y, or z of the laboratory system, $\mathbf{P}_{0i}$ is the polarization vector of the incident beam along the i axis, and $\mathbf{P}_{fj}$ defines the direction of analysis. The right-hand side of Eq. (1) has to be summed over i.

Since the experiments were carried out on a polycrystalline sample in the close vicinity of the phase transition at T_c, the angle of rotation for $\mathbf{P}$ in one domain is small. The total rotation angle of P in the 2-mm-thick sample may still be appreciable. Thus a small-angle approximation is applicable[5–7] which allows one to determine the mean magnetic induction B and $B_s^2\delta(1-m^2)$ from a measured depolarization matrix D. Here, δ is a measure of the mean domain size in the propagation direction of the neutrons and B_s and $m=B/B_s$ denote the spontaneous and normalized mean magnetic induction, respectively. Actually, in our measurements with an applied field, B is proportional to the total angle of rotation φ_t, of the polarization vector $\mathbf{P}$. This angle is directly obtained from the imaginary arguments of the eigenvalues of D.[6,7] This angle is determined modulo 2π. The number of multiple rotations of 2π is found by measuring the angle, starting with $0\le\varphi_t<2\pi$ in the paramagnetic region, and successively counting the 2π rotations below T_c while decreasing the temperature continuously. A rotation angle of 1° corresponds to about 1.2×10^{-4} T in our experiments. The resolution in φ is 1 to 2°. A more extensive description of the neutron depolarization apparatus and the derivations of the basis equations of the neutron depolari-

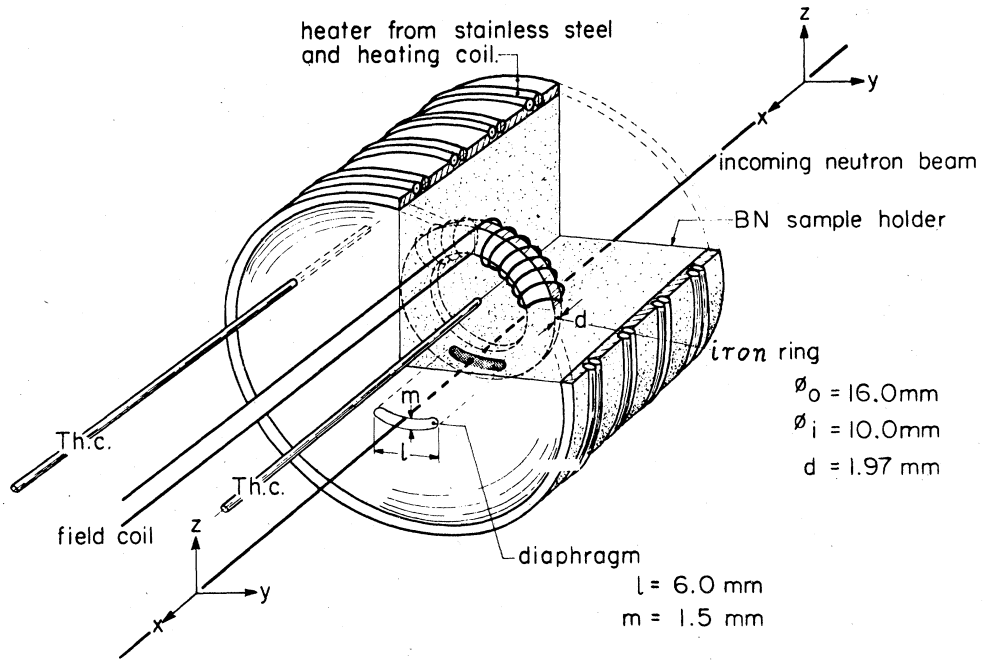

FIG. 1. Sketch of the sample holder and the system of reference used in the depolarization analysis. A short description is given in the text. (Th.c denotes thermocouple.)

zation analysis has been given in Refs. 5–7.

The experiments were carried out on a ring of polycrystalline pure iron (0.999975% pure). The outer (φ_o) and inner (φ_i) diameter of the ring were 16.0 and 10.0 mm, respectively, and the thickness $d=1.97$ mm (Fig. 1). Both surfaces of the ring were mechanically polished so that the maximum variation in the thickness of the sample was within 2 μm. Hence, the macroscopic demagnetization factor will be very small and the influence of demagnetizing fields can be neglected. The magnetic field was generated by a toroidal coil wound around the ring with 783 turns/m measured at the average circumference of the ring. The part of the sample indicated by the shaded area was transmitted by the neutrons.

The sample was mounted in a vacuum furnace ($p \lesssim 10^{-5}$ Torr) whose mantle temperature was controlled within 100 mK by water cooling. Two boron nitride (BN) disks symmetrically sandwiched the ring and they were positioned in the cylindric heater of the furnace. The heater was surrounded by three radiation shields. Both BN disks had a sickle-shaped opening which provided a diaphragm of 6.0×1.5 mm^2 (Fig. 1) for the neutrons. Chromel alumel thermocouples were used as temperature sensors. The thermovoltage was measured with an accuracy of 0.1 μV and a proportional-integral feedback circuit stabilized the temperature within ±3 mK. The long-term stability was found to be better than 100 mK during one day. This should be compared with the measuring time of 10 min at one temperature and with the 40 h normally needed to carry out one measuring series with successive increasing and decreasing temperature steps. We could reasonably well correct for small drifts in temperature by comparing the results from the increasing and decreasing temperature runs.

In order to check for temperature gradients in the sample holder which are introduced by heat dissipation of the current in the field coil, one thermocouple was positioned at the center of the BN disks and the other near the outer side of the field coil. A careful analysis revealed that only very small temperature gradients ($\Delta T \lesssim 25$ mK) were introduced by currents below 0.2 A ($H \lesssim 160$ A/m). Above 0.2 A, the gradient increased almost quadratically with the current. All results presented in this paper are limited to this very-low-field region. In the future we will modify the sample holder such as to minimize the gradient caused by larger currents in the field coil.

III. EXPERIMENTAL RESULTS AND DISCUSSION

A. Zero-field measurements

To test the sharpness of the phase transition, a neutron depolarization analysis was performed on the unmagnetized sample ($m,H=0$). Figure 2 shows $B_s^2\delta$ versus temperature very close to T_c. From 0.5 K below T_c to the near vicinity of T_c, the convex curvature reflects the critical behavior of B_s, expressed by a power law with a critical exponent β. With $\beta=0.37$, a mean domain size $\delta\approx15$ μm was determined which appears to be constant in the temperature region $T=T_c-0.5$ K to $T_c-0.1$ K. This indicates that no significant change in the domain structure takes place on approaching T_c, consistent with re-

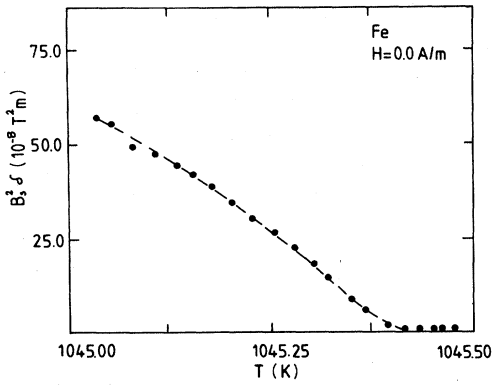

FIG. 2. $B_s^2\delta$ versus temperature T, analyzed from a depolarization analysis in zero field.

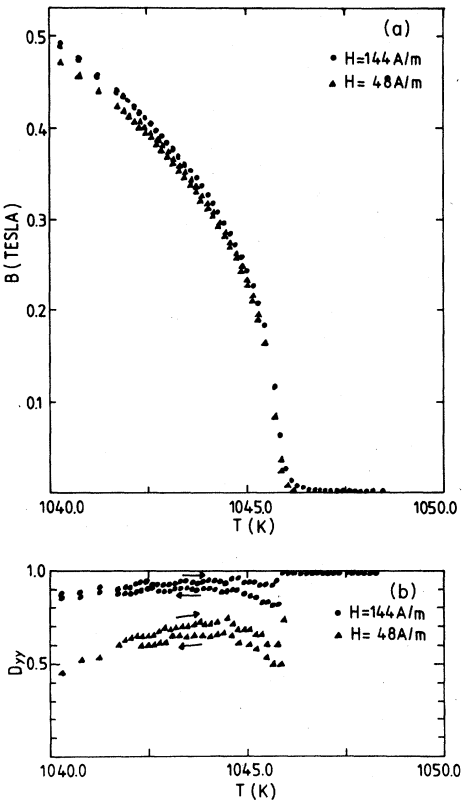

FIG. 3. Experimental data of the mean magnetic induction B and the depolarization matrix element D_{yy} versus temperature T measured at two different field strengths. The smaller value of B in the lower field $H=48$ A/m relative to the measurement in $H=144$ A/m is related to the presence of crystalline anisotropy (see text). The arrows in (b) indicate the direction of the temperature change during the measuring run.

sults obtained formerly in nickel.[5] Due to this stability in the domain structure, it should be possible to derive a value of β in the reduced temperature range $t=1\times10^{-4}$ to 5×10^{-4}. However, to do this with a small error in the value of β, the temperature has to be sensed with better precision.

Very near to T_c, a small tail in $B_s^2\delta$ versus T is observed, the origin of which is not quite clear; it might be caused by a still existing small temperature gradient less than 50 mK across the sample exposed. The presence of a tail may also arise from depolarization due to short-range order fluctuations around T_c. Assuming that the magnetization in one fluctuation cluster is comparable to the spontaneous magnetization at zero temperature, the cluster size δ is estimated to be a few 10th nanometers in a region 30 mK around T_c. These values are of the same order of magnitude as found from neutron scattering.[8] Similar results in the neutron depolarization have also been obtained in nickel.[5]

B. Measurements in low field

These measurements were performed with successive increasing and decreasing temperature steps. In all the series the sample was cooled down below T_c to the starting temperature without applying a field. Thereafter, the magnetic field was turned on. The mean magnetic induction $B(T)$ and the depolarization matrix element $D_{yy}(T)$ measured at two field strengths $H=48$ A/m and $H=144$ A/m are shown in Figs. 3(a) and 3(b). The difference in the mean magnetic induction B observed in the measurements at the two field strengths below T_c [Fig. 3(a)] has to be attributed to the presence of anisotropy fields. The strength of the anisotropy field has been tested by the field dependence of the mean magnetization B. Measurements that were carried out in a reduced temperature range $t=5\times10^{-4}$ to 5×10^{-3} at different field strengths between $H=144$ A/m and $H=500$ A/m revealed no field dependence in B. Hence it has been concluded that the sample is nearly magnetically saturated at $H=144$ A/m. On the other hand, B measured at a field $H=48$ A/m has a somewhat lower value [$\Delta B/B$ about 3% at the reduced $t=5\times10^{-3}$, see Fig. 3(a)]. The latter field strength should then be of the same order of magnitude as the anisotropy field. Its value has to be compared with an anisotropy field of $H=23$ A/m at $t=5\times10^{-3}$ obtained from the analysis of the field-dependent ac susceptibility in iron whiskers.[9] The deviation of the mean magnetization B from its saturated value is also visible from the yy element of the depolarization matrix $\underset{\sim}{D}$ [Fig. 3(b)], where D_{yy} is related to the mean square of the component of the domain magnetization $m_\perp$ (normalized to B_s) that is perpendicular to the applied field direction[6] by

$$\ln(D_{yy}) \sim -B_s^2\delta m_\perp^2 \ . \tag{2}$$

It is evident from Fig. 3(b) that D_{yy} approaches 1, i.e., $m_\perp^2 \to 0$ [Eq. (2)] with increasing field strength at the same temperature. At a constant field strength, D_{yy} gradually decreases with decreasing temperature below $T=1044$ K. Such a behavior can be expected on the basis of an increase in both the magnetic anisotropy and in B_s. On approaching T_c, D_{yy} decreases with temperature and shows a minimum about T_c. In addition a pronounced hysteresis is observed between the runs with increasing

and decreasing temperature steps. The hysteresis in temperature indicates the presence of different domain structures at T_c, dependent upon whether the sample has been warmed up in a field from a temperature well below T_c or cooled down in the same field from the paramagnetic phase. This feature is probably related to the presence of anisotropy in the critical region[4] and will be discussed elsewhere.[10]

C. Critical behavior below T_c

To determine the spontaneous magnetization B_s from the measured B, one has to apply a magnetic field sufficiently large to overcome the anisotropy field (see preceding section), and in addition, one has to correct for a field-induced magnetization of paramagnetic origin. This paraeffect becomes significant very near to T_c and is expected to be a factor of about 4 smaller in the ferromagnetic phase compared with the paramagnetic phase.[11] Because the fits are based on data points with $t \geq 5\times10^{-4}$, the contribution of a paraeffect to B is only relevant at the highest-temperature data considered and then at most 1% at fields $H \leq 144$ A/m. Therefore, this effect is neglected in the analysis. It is evident from the above discussion that the mean magnetic induction B, measured at reduced temperatures t in the range $t=5\times10^{-4}$ to 5×10^{-3} with $H\approx144$ A/m, should be closely identical to B_s. Therefore, B must obey the same critical behavior versus temperature near the phase transition as predicted for B_s, i.e.,

$$m=\frac{B(T)}{B_s(0)}\approx\frac{B_s(T)}{B_s(0)}=bt^{\beta}(1+at^{\Delta}) , \tag{3}$$

β and b are the critical exponent and the critical amplitude, respectively. Equation (3) includes a correction to the scaling term defined by an universal critical exponent Δ and a correction to the scaling amplitude a. This last term is expected to be important when data cover a much wider temperature range. In the analysis of our data within the range $t=5\times10^{-4}$ to 5×10^{-3}, a three-parameter least-squares fit in $b_{\rm eff}$, T_c, and $\beta_{\rm eff}$ to a simple power law has been made:

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

which approximates Eq. (3) for small values of t. Initial values of $b_{\rm eff}$, T_c, and $\beta_{\rm eff}$ have been obtained from a $\ln(m)-\ln(t)$ plot, using that value of T_c, which produced the best straight line. Those values in $b_{\rm eff}$, T_c, and $\beta_{\rm eff}$ that gave the minimum in the sum of the least squares are given in Table I. The fit to the experimental data at $H=144$ A/m is shown in Fig. 4. The excellent quality of the fit indicates that the data are well described by the power law (4) and possible deviations due to the correction to scaling are not resolved in the reduced temperature range $t=5\times10^{-4}$ to 5×10^{-3}. The mean value of $\beta_{\rm eff}$ and $b_{\rm eff}$ at $H=144$ A/m from both runs (decreasing T steps and increasing T steps, see Table I) leads to $\beta_{\rm eff}=0.363\pm0.004$ and $b_{\rm eff}=1.525\pm0.02$. The errors are mainly due to the small drift in temperature during the measuring series. The difference between $\beta_{\rm eff}$ and the universal β can be estimated by evaluating $d\ln(m)/d\ln(t)$ in Eqs. (3) and (4) which results in

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

$\bar{t}$ is some mean reduced temperature of the temperature range covered in the fit. With $\Delta=0.55$, predicted theoretically for a three-dimensional Heisenberg model,[1] and $a=-0.5$ for iron,[11] a value of $\beta=0.373\pm0.004$ can be estimated using $\bar{t}=2.5\times10^{-3}$. This β is close to 0.365 ± 0.001, derived from renormalization theory for a three-dimensional Heisenberg model. The relation between $b_{\rm eff}$ and b is obtained from a comparison of Eq. (3) and Eq. (4) with (5) substituted into (4):

$$b_{\rm eff}=b\left\langle\frac{(1+at^{\Delta})}{t^{a\Delta\bar{t}^{\Delta}}}\right\rangle_{\rm av} . \tag{6}$$

$\langle\ \rangle_{\rm av}$ denotes an averaging in the reduced temperature range under investigation. From Eq. (6) the critical amplitude $b=1.57\pm0.02$ is estimated. It is difficult to compare our present results with those obtained from most other techniques, where a value of $\beta_{\rm eff}$ is derived from fitting the spontaneous magnetization, measured in a larger temperature range, to the power law (4). Mainly, Mössbauer studies on iron have been performed at temperatures corresponding with reduced temperatures $t\leq5\times10^{-3}$.[12,13] The hyperfine field has been resolved in those experiments within 0.5 to 1 kG, which corresponds to a precision in the magnetic induction B of $30-60\times10^{-4}$ T. The precision in B is about a factor of 20 worse than that obtained by neutron depolarization. The lower resolution in B by Mössbauer experiments becomes evident from the relatively large error $\Delta\beta_{\rm eff}=0.01$ to 0.02 reported by these authors.

TABLE I. Critical exponents of iron. $\beta_{\rm eff}$, $b_{\rm eff}$ result from fits in a reduced temperature range $t=5\times10^{-4}$ to 5×10^{-3}. The numbers in parentheses are the uncertainty in the last digit. The uncertainty does not include the small drifts in the temperature (see text).

$\beta_{\rm eff}$	$b_{\rm eff}$	T_c (K)	γ	$H_{\rm appl}$ (A/m)
0.360(2)	1.51	1045.84	1.330(15)	144[a]
0.366(2)	1.54	1045.88	1.330(15)	144[b]
			1.35(3)	48[a]
			1.34(3)	48[b]

[a]Measuring run with increasing T steps.
[b]Measuring run with decreasing T steps.

The resolution in the determination of B, achieved by the neutron depolarization technique, should be sufficiently high to derive the first-order correction to scaling term in iron and to determine the critical exponent Δ with significant accuracy. To do this a measurement of B in a wider temperature range from about T_c-50 K up to T_c is needed. Such a measurement has to be performed in larger fields ($H\approx 1500$ A/m) to saturate the polycrystalline iron even at the lowest temperature in this temperature range. Preliminary results show a continuous variation in β_{eff} with temperature [see Eq. (5)]. However, the temperature gradients, introduced by the field current (see experimental details), prohibit at the moment a reasonably accurate determination of the first-order correction to scaling term in the critical exponent Δ. It should be noted that the correction to scaling in iron has been recently analyzed from Mössbauer experiments.[12] From that analysis a value of the correction-to-scaling amplitude $a=-0.46$ and a universal $\beta=0.367$ has been derived from a multiparameter fit in four variables (b, β, T_c, and a) of the hyperfine field to Eq. (3). The fit has been performed in a reduced temperature range $t=10^{-3}$ to 3.4×10^{-1}, assuming that the theoretically predicted value of $\Delta=0.55$ is valid in iron. Due to the negative value of a, β_{eff} should always be below the universal β [see Eq. (5)]. Values of β_{eff}, derived by this author from the power law (4) for $t\leq 2\times 10^{-2}$ are between 0.371 and 0.379 and hence located above $\beta=0.367$. We can estimate from this β an expected value of $\beta_{\text{eff}}=0.34$ using the correction parameters found by this author and a mean reduced temperature $\bar{t}=10^{-2}$ for the temperature range $t=10^{-3}$ to 2×10^{-2}, where this author fitted a value of $\beta_{\text{eff}}=0.371$ (absorber experiment) or 0.379 (source experiment). From the latter calculations, an inconsistency of about 0.03—0.04 in β_{eff} or β may be concluded.

D. Critical behavior above T_c

The static critical behavior just above T_c is described by the relation

$$m=\frac{B(T)}{B(T=0)}=G^{+}t^{-\gamma}H\ . \tag{7}$$

m/H is assumed to be identical with the initial paramagnetic susceptibility χ_0 and t, γ, and G^{+} are the reduced temperature, the critical exponent, and the critical amplitude, respectively. From a theoretical point of view, Eq. (7) is strictly valid in the limiting case $H\rightarrow 0$. A least-squares fit was performed to fit our data to Eq. (7). Initial values of T_c, γ, and G^{+} were obtained by the same procedure, used in the fit of a similar power law in the ferromagnetic region. Due to the small drift in temperature, the data of the runs with increasing and decreasing temperature have been fitted separately. The values of γ, which produced the best fits to the individual measuring runs are also summarized in Table I. In Fig. 4 the best fit, using the parameters given in the first column of Table I, to the experimental data is shown for one measurement, which was carried out with increasing temperature steps at $H=144$ A/m. The fit is excellent. The values of γ, obtained from the fits, are consistent with

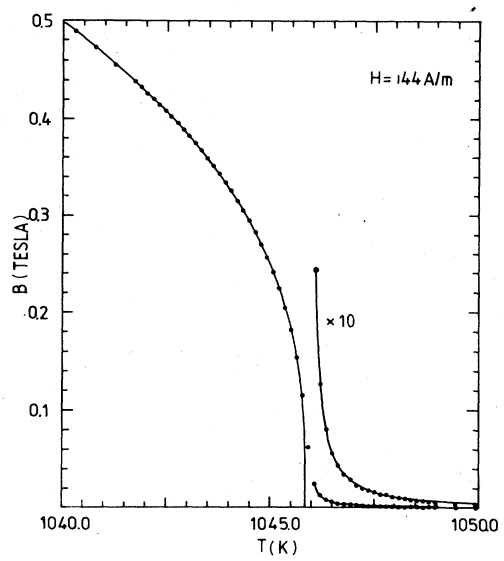

FIG. 4. Results of our best fits (——) based on simple scaling laws (see text) to the experimental data (•••••) of the mean magnetic induction B for the measuring run, performed with increasing temperature steps in the presence of a field of $H=144$ A/m.

those obtained by other methods such as the Faraday balance [$\gamma=1.33$ with $H\geq 1400$ A/m (Ref. 14)] or induction experiments [$\gamma=1.33$ with $H\geq 2000$ A/m (Ref. 15)]. It will be noted that the accuracy of γ increases when using larger fields. In addition, it should be mentioned that the values of T_c, fitted in one run from scaling laws above and below T_c, are in all cases consistent within $\Delta T=20$ mK.

IV. SUMMARY AND CONCLUSION

The results obtained from a neutron depolarization analysis on a ring-shaped iron sample show that the neutron depolarization technique is a promising method to study the critical behavior of a ferromagnet around the phase transition. The magnetic induction B, derived from the Larmor precession of a polarized neutron beam in a ring of iron, magnetized to saturation, can be determined with a precision of about 1 to 2×10^{-4} T. In the ferromagnetic region this exceeds by more than a factor of 20 the relative precision of the best-known method used up to now, which is the determination of the hyperfine field by means of the Mössbauer effect.[12,13]

Fits of the magnetic induction B versus T in a reduced temperature range ($t=5\times 10^{-4}$ to 5×10^{-3}) to a simple power law yield an effective critical exponent $\beta_{\text{eff}}=0.363\pm 0.004$. Taking into account a correction to scaling, this results in an universal $\beta\approx 0.373\pm 0.004$, a value which is slightly above the predicted $\beta=0.365$ for the three-dimensional Heisenberg system.[1] The critical exponent $\gamma=1.33\pm 0.02$, derived from a fit of B versus T in the paramagnetic phase with applied fields H below 150 A/m, agrees well with those values obtained from

other techniques,[14,15] where much larger fields have been used.

The high resolution in B obtained by the neutron depolarization technique in combination with an elaborate temperature sensing should allow us to analyze data, measured in an extended reduced temperature range $t \leq 10^{-1}$, to determine the "correction to scaling" and to determine in an experimental way values of both the critical exponents β and Δ.

The neutron depolarization in zero field indicates that the domain structure is stable within 0.2 K just below T_c, a fact that can, in principle, be used to derive β_{eff} within $t \leq 5 \times 10^{-4}$ from the analyzed $B_s^2 \delta$.

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