Mössbauer study of static and dynamic critical behavior in Fe

M. A. Kobeissi*
Department of Physics, Yarmouk University, Irbid, Jordan
and Department of Physics, Clark University, Worcester, Massachusetts 01610
(Received 9 January 1981)

In this paper we present the results of detailed experimental studies on the critical behavior of Fe near its critical temperature T_c (here also Curie temperature T_C) using the Mössbauer effect. Using a highly controlled two-stage Mössbauer furnace, we made a careful measurement of the critical exponent β and the critical exponent z. The value of β_{eff} was obtained in both source and absorber experiments and was found to be 0.379(4) for the reduced-temperature range $10^{-3} \leq t \leq 2\times10^{-2}$ and 0.371(8) for $10^{-4} \leq t \leq 2\times10^{-2}$, respectively. We have used correction-to-scaling form in the critical region and have obtained the universal β as $\beta = 0.367(5)$ and the correction amplitude $A = -0.458(22)$. The results on β agree well with theory, bulk measurements on Fe, and most data on other ferromagnets in the universality class $(n,d) = (3,3)$. The value z was measured as $z = 1.93(19)$, which is inconsistent with the theory of short-range exchange interactions and neutron scattering results in Fe, but in agreement with previous hyperfine studies on Ni and Fe, and a recent hypothesis on the dynamic behavior in the asymptotic critical region. We have also investigated the possible magnetization dependence of the isomer shift in Fe and obtained negative results, both within 2 K of the Curie point, and in a wide region above and below the Curie point. These findings, like the β measurements, are based on both source and absorber experiments. They disagree with Preston's observations near T_c. By studying the Mössbauer line intensities we observed a magnetostrictive reorientation of ferromagnetic domains as a function of temperature. This behavior is similar to that observed in some Fe alloys under the influence of compressive strain as well as temperature variation.

I. INTRODUCTION

In the last few years there has been intensive activity in both theoretical and experimental study of static and dynamic critical phenomena with special emphasis on ferromagnets and antiferromagnets.[1–17] The critical behavior of the magnetization M and the spin fluctuations has been characterized in terms of critical exponents.

According to the renormalization-group approach,[1–3] static exponents describing singularities in thermal average properties depend primarily on effective-spin dimensionality, n, and lattice dimensionality d. Renormalization-group methods[4] yield highly accurate predicted values for several physically interesting pairs of (n,d). These results agree reasonably well with previously derived high-temperature-series results and incorporate fully the earlier generalizations of universality and scaling. In the case of dynamics, critical behavior depends in addition on conservation laws, and leads to the division of a single static universality class into several dynamic subclasses.[5]

From an experimental point of view one would like to test theoretical predictions for both static and dynamic behavior. The experimental results must be very precise, since the theoretical differences between classes are small as is illustrated for example in Table I for the exponent β. In this paper we are concerned with Fe, an isotropic ferromagnet with $(n,d) = (3,3)$, for which the isotropic Heisenberg model is thought to be an adequate description. Using the Mössbauer effect in ^{57}Fe we have made: (a) careful measurements of the critical exponent β; (b) a determination of the dynamic critical exponent z just above T_c; (c) measurements of the energy shift within a wide range of temperature around T_c; and (d) an observation of magnetic domain reorientation with temperature. Previous reports have given a preliminary account of some of this work.[6–8] The present paper contains our results and interpretations in complete form.

TABLE I. Variation of β with the spin dimensionality n for three-dimensional lattice $(d=3)$ as given by the theory (Ref. 4).

n	1	2	3
β	0.325(1)	0.346(1)	0.365(1)

II. THEORETICAL PREDICTION

In the magnetic solids the critical behavior of the magnetization M has been characterized in terms of the critical exponent β. The individual atomic moments in these solids fluctuate with frequencies of 10^{15} Hz far above and below the critical temperature T_c. These fluctuations slow down as the temperature approaches T_c from above or below. Hyperfine experiments provide methods of measuring β and may exhibit signals that reflect the time-dependent behavior of the spins. Here we summarize salient aspects of the theory and indicate what types of observables exist for various experimental techniques.

A. Static behavior: The static exponent β

For ferromagnets, the temperature dependence of the spontaneous magnetizations $M(T)$ in the vicinity of the critical temperature T_c can be described by the asymptotic relation of the form[17]

$$\lim_{T\to T_c} \sigma(T) = B'(1 - T/T_c)^{\beta_{\text{eff}}} , \tag{1}$$

where σ is the reduced magnetization, $\sigma = M(T)/M(0)$, T is the temperature, B' is a constant, and β_{eff} is an effective critical exponent. For measurements including data far away from T_c, the above equation must be modified.[18] If we assume that the hyperfine field $H(T)$ is proportional to magnetization, as is proven in this work (see below), then Eq. (1) can be written in terms of hyperfine field and must include a "correction-to-scaling" term as follows[18,19]:

$$h(t) = Bt^{\beta}(1 + At^{\Delta}) . \tag{2}$$

Here $t = (T_c - T)/T_c$ is the reduced temperature, $h = H(T)/H(0)$ is the reduced hyperfine field, B is a constant, and A is a "correction-to-scaling" amplitude. β and Δ are universal static critical exponents of the ferromagnets. For measurements very close to T_c the correction term becomes very small and Eq. (2) can be approximated by a pure power-law form as[19]

$$h(t) = Bt^{\beta_{\text{eff}}} \tag{3}$$

with β_{eff} given by

$$\beta_{\text{eff}} = \beta - A\Delta|t|^{\Delta} . \tag{4}$$

In this case experimentally determined values of β_{eff} from pure power law may differ appreciably from the universal value β and comparison with theory becomes difficult. For the prediction of β and other static critical exponents, some theories have been put forward.[4] For $(n,d) = (3,3)$ the theoretical value of Δ is 0.55. The best theoretical results of β, obtained by renormalization-group methods, are given in Table I.

In recent publications, Aharony and Ahlers,[19] and Chang and Houghton[20] have derived theoretical expressions for the correction amplitudes and their ratios. According to these authors the ratios among the correction amplitudes of the same material are universal, but not the amplitudes themselves. For two critical exponents λ_i and λ_j and their corresponding correction amplitudes A, Aharony derived the relation

$$\frac{A_i}{A_j} = \frac{\lambda_{i,\text{eff}} - \lambda_i}{\lambda_{j,\text{eff}} - \lambda_j} , \tag{5}$$

where $\lambda_{i,\text{eff}}$ and λ_i are the effective and the universal exponents, respectively. In ferromagnets experimental information on the ratios in Eq. (5) are scarce. Thus, when data are fitted to a single power law close to T_c and to correction-to-scaling form in a wide range of the reduced temperature, the universal exponent β, β_{eff}, and the correction amplitude A may be obtained. Theoretical predictions for the universal ratios in Eq. (5) can also be tested, provided experimental values on other critical exponents are available. The range of the asymptotic behavior in Eq. (3) must be determined experimentally in order to determine a single-valued critical exponent β_{eff}.

B. Critical fluctuations: The exponent z

Dynamic properties of magnetic critical phenomena are related to critical spin fluctuations. Above and close to the critical temperature T_c individual spin fluctuations are correlated to those of neighbors and regional spin clusters of size ξ form. As T_c is approached, the lifetime τ_c and the correlation length ξ of the spin clusters diverge. In what follows we will give a brief summary of the theory on this subject which is closely related to our work.

The fundamental property of interest in the study of spin fluctuations is the spin-correlation function defined by

$$G(r,t) = \langle \vec{S}(r,t) \cdot \vec{S}(0,0) \rangle . \tag{6}$$

This describes the space-time correlation between a spin $\vec{S}(r,t)$ situated at r and t, and the spin $\vec{S}(0,0)$ situated at the origin $r = 0$, $t = 0$.

In a recent review article Hohenberg and Halperin have given a detailed discussion on critical dynamics.[5] These authors find that, for isotropic spin systems, Eq. (6) has a Fourier transform given by

$$S(\vec{q},\omega) = 2\pi\omega^{-1}(\vec{q})S(\vec{q})f_{q/\kappa}(\omega/\omega(\vec{q})) , \tag{7}$$

where ω and $\vec{q}$ are the energy and the wave number of the fluctuations, respectively, $S(\vec{q})$ is the static scaling function, $\omega(\vec{q})$ is the energy linewidth of the critical mode describing the dynamics of the spin fluctuations, $f_{q/\kappa}$ represents the shape function of the energy line, and κ is the inverse correlation length of

the fluctuations. According to static and dynamic scaling hypotheses, $S(\vec{q})$ and $\omega(\vec{q})$ are assumed to be homogeneous functions of $\vec{q}$ and κ, and can be written therefore as

$$S(\vec{q}) = q^{-2+\eta} g(\vec{q}/\kappa) \ , \tag{8}$$

$$\omega(\vec{q}) = q^z \Omega(\vec{q}/\kappa) \ , \tag{9}$$

$$\omega(\vec{q}) = \kappa^z \Omega'(\vec{q}/\kappa) \ , \tag{10}$$

where z is the dynamic critical exponent and Ω and Ω' are two different shape functions, and η is a static exponent whose value is close to zero. The shape function in Eq. (7) must satisfy the following normalization conditions as

$$\int_{-1}^{+1} f\left(\frac{\omega}{\omega(\vec{q})}\right) \omega^{-1}(\vec{q})\, d\omega = \tfrac{1}{2} \ ,$$

$$\int_{-\infty}^{\infty} f\left(\frac{\omega}{\omega(\vec{q})}\right) \omega^{-1}(\vec{q})\, d\omega = 1 \ .$$

From the above equations it can be seen that the shape of $S(\vec{q},\omega)$ does not depend on $\vec{q}$ and κ separately but on their ratio $\vec{q}/\kappa$.

The determination of the dynamic exponent z can be achieved by neutron scattering; the energy linewidth $\omega(\vec{q})$ is measured at $T = T_c$ as a function of the wave vector $\vec{q}$. In this case $\kappa \to 0$ and Eq. (9) follows a power-law form given by

$$\omega(\vec{q}) = \text{const}\ q^z \ . \tag{11}$$

The linewidth measurements are then fitted to Eq. (11) and z can be obtained. The problem with this method is that in ferromagnets one is limited to a wave number $q \geq 0.05$ Å^{-1}, in order to avoid the effects of elastic scattering at $q = 0$. To overcome this experimental difficulty, hyperfine interaction methods, which measure fluctuation times for all values of $\vec{q}$, are used. To relate the isotropic spin autocorrelation time τ_c to the function $S(\vec{q},\omega)$, we use Eq. (6) and the definition

$$\tau_c \equiv \tfrac{1}{2} \int_{-\infty}^{\infty} \frac{G(0,t)}{G(0,0)} dt \ ,$$

from which it follows that[11]

$$\tau_c = B \int_{V_q} S(\vec{q},0)\, d\vec{q} \ , \tag{12}$$

where V_q is the Brillouin-zone volume. The temperature dependence of τ_c can be introduced via the inverse correlation length κ given by

$$\kappa = \kappa_0 t^{\nu} \ , \tag{13}$$

where $t = (T - T_c)/T_c$ is the reduced temperature and ν is a static exponent. From Eqs. (6)–(13) it can be shown that[11]

$$\tau_c = C_0 t^{-w}; \quad w = \nu(z + 2 - d - \eta) \ . \tag{14}$$

Thus the correlation time follows a power-law form and diverges as T_c is approached. For a lattice dimensionality $d = 3$, we get $w = \nu(z - 1 - \eta)$. Hence measurements of correlation times via hyperfine interactions allow the determination of w; consequently the value of z can be obtained if ν and η are known. With $\nu = \frac{2}{3}$, $\eta = 0$, and the predicted $z = 2.5$, the exponent w acquires the value $w = 1$.

To test whether these predictions are correct, it is necessary to relate τ_c to Mössbauer observables. This has been done by a number of authors.[21–24] Here we make use of the theory of Bradford and Marshall,[22] who give an explicit relation between τ_c and the excess linewidth $\Delta\Gamma$. Bradford and Marshall begin with the Hamiltonian

$$H = A_\alpha \vec{I} \cdot \vec{S}(t), \quad \alpha = e,g \ , \tag{15}$$

where e and g denote the excited and ground state, respectively, and $\vec{I}$ and $\vec{S}(t)$ are the nuclear and electronic spins. A is a coupling constant given by

$$A_\alpha = \gamma_\alpha H_0 / S \ . \tag{16}$$

γ is the gyromagnetic ratio, S is the atomic spin, and H_0 is the hyperfine field at zero temperature. The use of this Hamiltonian assumes that the hyperfine field at the ^{57}Fe nucleus is produced entirely by an electronic spin $S(t)$ and that it is proportional to the time-averaged spin. That this is at least approximately true follows from the study of hyperfine-field systematics in ferromagnetic hosts.[25]

The basic assumptions that restrict the applicability of the theory are as follows: (a) the fluctuations are assumed to be isotropic; (b) the inequalities $\tau_c \omega_L \ll 1$, $\Delta\Gamma \tau_c \ll 1$, $\Delta\Gamma/\omega_L \ll 1$ are assumed to hold, where ω_L is the Larmor frequency; and (c) the correlation function is taken to be exponential in time. The validity of the theory subject to these conditions has been shown by Gottlieb and Hohenemser,[26] and by us in a previous work[27] on Ni; we will also use it in our present work.

With these restrictions the line shape is a Lorentzian of the form

$$I(\omega) = \frac{\Gamma_n + b\tau_c}{\omega^2 + (\Gamma_n + b\tau_c)^2} \ . \tag{17}$$

Here Γ_n is the natural linewidth, and

$$\Delta\Gamma = b\tau_c \tag{18}$$

is the line broadening with

$$b = \frac{S(S+1)}{3\hbar^2} \left(\tfrac{15}{4} A_e^2 - \tfrac{5}{2} A_e A_g + \tfrac{3}{4} A_g^2\right) \ . \tag{19}$$

To relate the line broadening to temperature we use Eq. (14) with the result

$$\Delta\Gamma = E t^{-w} \ , \tag{20}$$

with $\Delta\Gamma$ expressed in units of $\sec^{-1}$:

$$E = B\left[\frac{2S(S+1)}{3h^2}\left(\frac{15}{2}A_e^2 - \frac{5}{2}A_eA_g + \frac{3}{4}A_g^2\right)\right], \quad (21)$$

where B is a constant.

Thus, $\Delta\Gamma$ diverges with the exponent $w=1$, and an amplitude E that is quantitatively specified by the hyperfine interaction strength. This prediction offers an alternative method of testing the theory of critical spin dynamics. This method is promising in that it is not subject to the experimental and physical limitation imposed on neutron scattering in ferromagnets[28–31]; i.e., we are able in principle to measure arbitrarily close to T_c and thus obtain results in a wide temperature range not accessible to neutron scattering.

III. EXPERIMENTAL PROCEDURE

As we have mentioned above, experimental determination of critical exponents in ferromagnets requires precise measurements so that we may compare experimental results with highly precise theoretical values. This demands measurements very close to T_c with good resolution of Mössbauer lines. In addition the sample in both absorber and source experiments must be free of thermal gradient.[27] In source experiments the source must be homogeneous in order to minimize linewidth broadening. While it is easier to achieve a source experiment nearly free of thermal gradient, an absorber experiment has larger sample area and is therefore more susceptible to small thermal gradients. This may cause unreliable measurements of critical fluctuations just above T_c,[27] although linewidths in this case are narrower and make regions just below T_c more accessible for the measurement of the critical exponent β. In Fig. 1 we compare source and absorber Mössbauer spectra at almost the same reduced temperature. It can be seen that the absorber spectrum shows much better resolved lines.

A source of spurious results might also be the Mössbauer drive, which must maintain a very good linearity and high stability so that line broadening and apparent energy shift can be avoided. The parabolic motion of the drive also produces a slightly larger uncertainty in the Mössbauer line positions when an absorber experiment is employed.

For the above reasons, we found it appropriate in the present work on Fe to choose a source experiment for the determination of the exponent z. As for β measurement, in addition to the source experiment, we used also an absorber experiment in order to assure β determination in pure Fe so that comparison between both cases can be made.

The measurement of critical exponents in Fe is more difficult than in other ferromagnets with lower

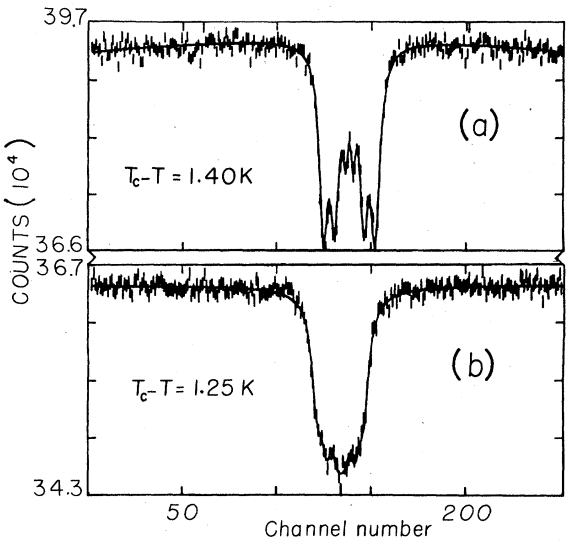

FIG. 1. Close-in Mössbauer spectra in an (a) absorber experiment and (b) source experiment at the same reduced temperature. It can be seen that better resolvable spectra are obtained in the absorber experiment.

T_c. First, in Fe, $T_c = 1043$ K, making temperature control and stability a greater challenge to the experimentalist. Secondly, the recoil-free fraction f is reduced to about one-third of its room-temperature value, thus markedly decreasing the signal-to-noise ratio. Thirdly, Fe oxidizes very easily, and therefore great care must be taken to keep it in a clean environment. In the following we will give a description of the experimental procedure by which we avoided problems in these areas.

A. Mössbauer furnace

Our specially designed vacuum furnace, full details of which can be found elsewhere,[32] consisted of a tubular outer heater and a disk-shaped inner heater, both made of boron nitrate with bifilar windings. The furnace servo system maintained a rapid response due to the good thermal conductivity of the ceramics used and produced a long-term stability better than 0.02 K per day at the Fe Curie point. At that temperature, the thermal gradient across the source was found to be less than 0.05 K. For the absorber experiment the thermal gradient was less than 0.10 K across the sample as can be shown by the sharpness of the Curie-point transition obtained in our Mössbauer data (see below).

To create a clean surface for Fe, the sample was sandwiched between two thin BeO disks. We achieved an environment nearly free of magnetic fields by shielding the furnace with μ–metal. Stray fields from the Mössbauer drive were thereby reduced to less that 0.15 G at the source.

We used Chromel-Alumel thermocouples for temperature measurements. We did not make absolute calibration of the thermocouples because in our measurements only the reduced temperature enters into our results. However, the error in the absolute temperature scale is about 2 K. The power consumed by the furnace at 1100 K was about 60 W. With the help of a forepump and a diffusion pump equipped with a liquid-nitrogen trap, the vacuum of the furnace was maintained at about 10^{-6} torr throughout the experiment.

B. Source and absorber

Unlike the Mössbauer-effect (ME) results of Preston,[9,12,13] who did an absorber experiment, our results were obtained with both an Fe source and Fe absorber mounted inside a furnace. Our choice of a source experiment for β and z measurements, as stated above, was dictated by our judgement that temperature control and uniformity are the most important requirements for an improved experiment, and that success in this could be more easily achieved for a relatively small source "spot" than for a much larger absorber surface.

The source was made by electroplating 2.8-m Ci ^{57}Co on a less than 3-mm diameter area of 99.99% pure Fe foil, and diffusing the activity in an ultrapure H_2 atmosphere for 15 h. This produced a nearly uniform distribution in the 15×10^{-4}-cm thick foil with an estimated fraction of radioactive ^{57}Co of less than 4×10^{-4}. The homogeneity of the source distribution was checked by the equal count rate of the x rays as well as the 14.4-keV gamma rays from both sides of the Fe foil. The fact that the probe distribution was homogeneous follows from the absence of line broadening for $t\geq10^{-3}$.

As an absorber for the source experiment we used a 25-mm-diameter matrix of $K_4Fe(CN)_6\cdot3H_2O$ enriched to 90% in ^{57}Fe and containing 0.25 mg/cm^2 of ^{57}Fe.

The absorber, which was at room temperature, was driven by a constant-acceleration drive. For an emission spectrum of natural width an experimental linewidth of 0.35 mm/sec is expected.[33,34] The difference between this value and our observed linewidth of 0.398(5) mm/sec at room temperature can be accounted for by a small amount of resonant self-absorption in the source.

In the absorber experiment, for an additional measurement of β and the energy shift near T_c in pure iron, a 99.99% pure Fe foil was used. The exposed area for the transmission was a circle with 7-mm diameter. The moving source was an 18-m Ci ^{57}Co diffused in paladium foil and maintained at controlled room temperature. For this arrangement the transmission Mössbauer spectrum showed an experimental linewidth of 0.266(9) mm/sec as compared to 0.398(5) in the source experiment. Thus absorber experiments produce better Mössbauer line resolution than source experiments and provide more precise measurements in regions very close to T_c, as can be seen in Fig. 1. The only problem with this method is that thermal gradients across the sample might be larger than in the case of source experiments. This effect is undesirable when measuring magnetic fluctuations above T_c.[27] Furthermore, the errors in the linewidths and positions are slightly larger due to the parabolic form of the spectrum background.

IV. DATA ANALYSIS

Mössbauer study of Fe near its Curie point was done first by Preston, who obtained values of $\beta_{\rm eff}=0.342(4)$ and $\beta_{\rm eff}=0.37(2)$ in the temperature ranges $10^{-3}\leq t\leq10^{-2}$ and $10^{-4}\leq t\leq10^{-2}$, respectively,[9] but no investigation was made on the critical fluctuations. Shaham *et al.*[10] have recently studied critical behavior in pure Fe by NMR and determined $\beta_{\rm eff}=0.320(4)$ in the range $4\times10^{-3}\leq t\leq5\times10^{-2}$. The results of the above works on $\beta_{\rm eff}$ are not consistent with other ferromagnets of the $(n,d)=(3,3)$ class and are in disagreement with recent theoretical results in the same class. Shaham studied also the dynamics of Fe just below T_c and found the dynamic critical exponent $w=0.63(12)$, $[z=1.95(18)]$, within $4\times10^{-3}<t<1.5\times10^{-2}$, in disagreement with the theory on dynamics, but consistent with a recent hypothesis by Suter and Hohenemser[11] and with the theoretical treatment of the non-spin-conserving forces.[5,35] In a later publication Lee Chow *et al.*[36] reported a study on the dynamics in Fe near T_c using the perturbed angular correlation (PAC) method and a ^{100}Rh probe. They observed a crossover of z ($z=2.5\rightarrow z=2.0$) thus confirming the above-mentioned hypothesis. It is of interest to measure z in probe-free Fe and to compare with NMR results and theory. In the following we present our measurements of $\beta_{\rm eff}$, universal β obtained through correction-to-scaling form, the correction amplitude A in Eq. (2) and the exponent z in Fe where probe and sample are the same.

A. Universal β and $\beta_{\rm eff}$

To extract the value of β in our experiment, Mössbauer positive and negative velocity spectra were recorded in halves of a multichannel analyzer, thus providing a useful consistency check on our results. Typical spectra appear in the following sections (see Fig. 14). To fit the spectra a standard multiparameter nonlinear least-squares program was used, in which line intensity, position, and width were all treated as free parameters. In this way the hyperfine field was determined to an accuracy of better than 0.5

kG for $10^{-2} \leq t \leq 0.7$, and to an accuracy of better than 1 kG for $10^{-3} \leq t \leq 10^{-2}$. Although data in the source experiments were obtained for $t \leq 10^{-3}$, spectra from this region were excluded because they showed no discernible structure other than a single broad line. The hyperfine-field values in both source and absorber experiments were the same as those obtained by Preston at the corresponding temperatures. Figure 2 summarizes a plot of reduced field $h(t)$ vs T/T_c and is in agreement with previous ME work.[12] The solid line in Fig. 2 represents bulk measurements by Potter[37] and confirms the relation $h(t) = \text{const}\ \sigma(t)$, which justifies Eqs. (2) and (3) in this work.

Hyperfine fields from both source and absorber experiments were fitted separately to a single power law, Eq. (3), with B, T_c, and β_{eff} as free parameters. In order to examine the extent of the critical region, a series of fits was made in which the maximum value of t was successively reduced. The result is that for all values of $t_{\max} \leq 2 \times 10^{-2}$, the values of B, T_c, and β_{eff} obtained as a function of $t_{\max}$ remain constant. This is illustrated in Fig. 3 for both experiments. We conclude that the asymptotic region for Fe is defined by $t \leq 2 \times 10^{-2}$, in good agreement with expectation.[38,39] For this region our best estimate of all three parameters in the source experiment is: $\beta_{\text{eff}} = 0.379(4)$, $B = 1.66(3)$, $T_c = 1042.91(4)$ for $10^{-3} \leq t \leq 2 \times 10^{-2}$. For the absorber experiment we obtained: $\beta_{\text{eff}} = 0.371(8)$, $B = 1.62(5)$, $T_c = 1043.05(8)$ for $3 \times 10^{-4} \leq t \leq 2 \times 10^{-2}$.

As a check on the above procedure, T_c was also determined by thermal scanning: i.e., the transmission at the centroid of the spectra was determined in the neighborhood of the transition, with the result illustrated in Fig. 4. From this it is concluded that T_c, for source and absorber experiments, is equal to:

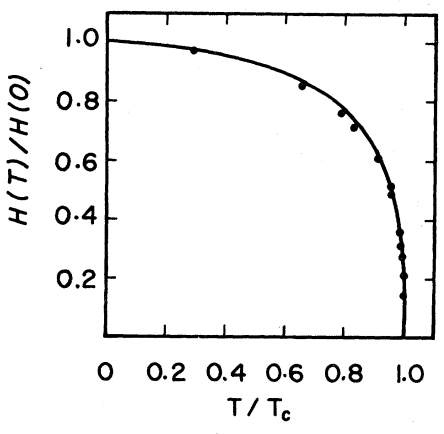

FIG. 2. Reduced hyperfine field $h(t) = H(T)/H(0)$ as measured by ME in $Fe^{57}Fe$ below T_c. The solid line is the reduced bulk magnetization as measured by Potter in pure Fe.

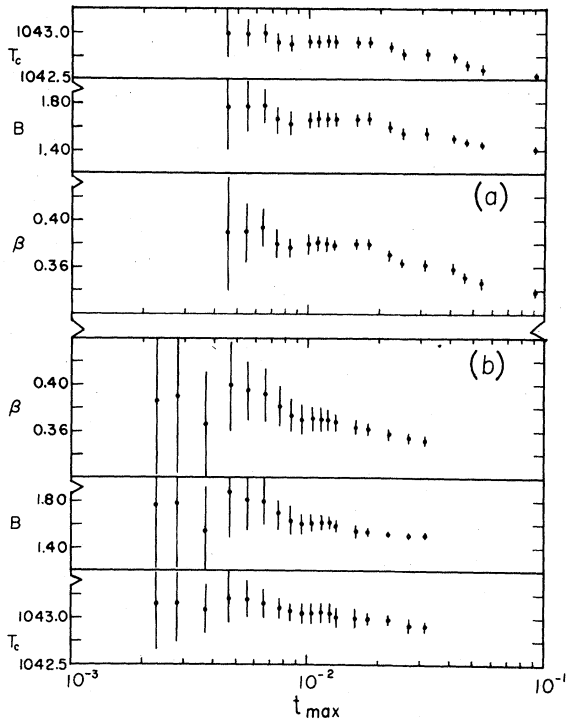

FIG. 3. β_{eff} measurement in $Fe^{57}Fe$. The variation of the exponent β_{eff}, the constant B and T_c in $Fe^{57}Fe$ with maximum reduced temperature in both (a) source experiment and (b) absorber experiment. Notice the range of the critical region where β_{eff}, B, and T_c remain constant.

$T_c = 1043.0(1)$ and $T_c = 1043.1(1)$ K, respectively, and in good agreement with the above. Also shown in Fig. 4 is a plot of H_{hf} raised to the $1/\beta_{\text{eff}}$ power, for $\beta_{\text{eff}} = 0.379$ and $\beta_{\text{eff}} = 0.371$. This illustrates graphically the degree of agreement between the two T_c estimates in each experiment.

A conventional logarithmic plot of $h(t)$ appears in Fig. 5 showing the range of the critical region in Fe.

To check if linewidth problems affected the present work, we made a careful determination of the linewidth predictions for thick absorbers,[34] with the results $\Gamma = 0.389(3)$ mm/sec as shown in Fig. 6. It is seen that our work is not affected by unexplained line broadening in the region attained by our resolvable spectra in the source experiment. In the absorber experiment the linewidth was smaller and we were able to obtain resolvable spectra closer to T_c than in the source experiment as shown in Fig. 1. Figure 6 shows the small increase in linewidth at $t \leq 10^{-3}$ in the absorber experiment. This is due mainly to two effects: the first is the spin fluctuations where linewidth increase is very small (see below) and the second and more influential[27] is the possible small thermal gradient due to the relatively large transmission area in the iron foil. Therefore

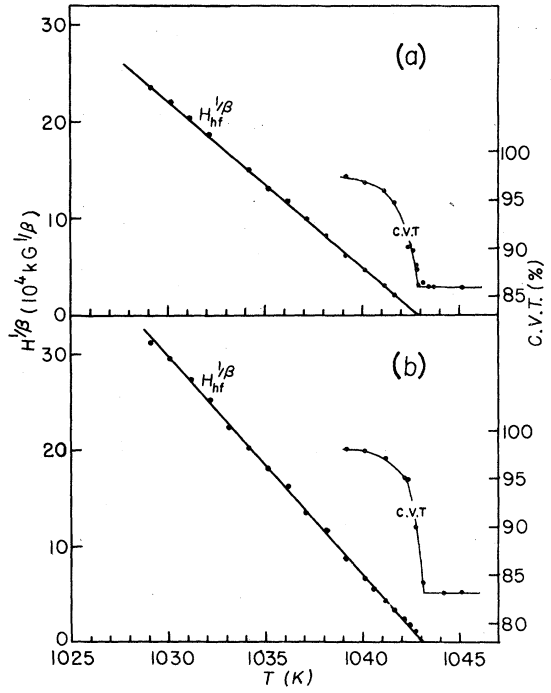

FIG. 4. Determination of T_c for the ME spectra via the single power law and thermal scan in (a) source and (b) absorber experiment. Each graph shows both the temperature variation of $H^{1/\beta}$ and the centroid velocity transmission (c.v.t.). In each case T_c is determined independently. For each case the corresponding β_{eff} was used.

source experiments with small active spots are more appropriate for spin fluctuations than are absorber experiments.

For the determination of the universal β, our data, in a wider temperature range, must be fitted to the correction-to-scaling form of Eq. (2) with B, β, T_c, and A as free parameters and fixed theoretical[4] $\Delta=0.55$. The results are $B=1.67(3)$, $\beta=0.367(5)$,

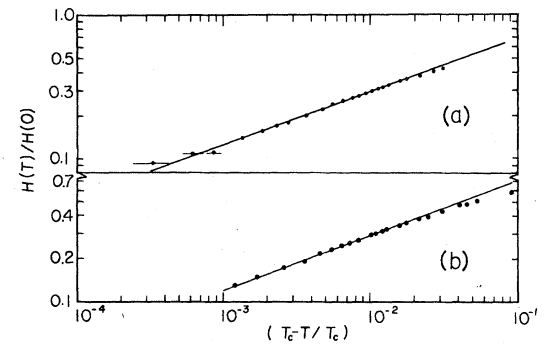

FIG. 5. Logarithmic plot of the reduced hyperfine field for (a) absorber and (b) source experiment. The breakdown of the power law at $t=2\times10^{-2}$ can be seen in both cases.

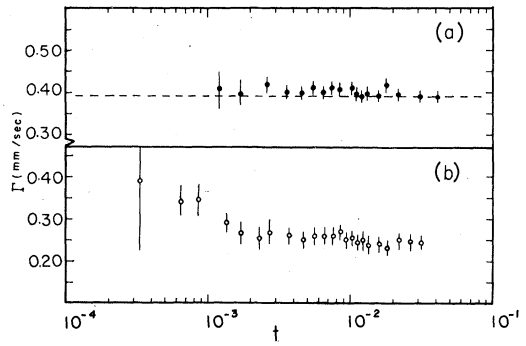

FIG. 6. Variation of the linewidth with reduced temperature below T_c. (a) The width in the source experiment remains constant and agrees with $\Gamma=0.389(3)$ mm/sec calculated from the theory of Margulies and Ehrmann (Refs. 33 and 34) (dashed line). (b) The increase in the width for $t<10^{-3}$ in the absorber experiment is due to fluctuations and possible small thermal gradient across the foil (Ref. 27).

$A=-0.458(22)$, and $T_c=1042.43(13)$ K for $10^{-3}\leq t\leq 3.4\times10^{-1}$ with a T_c shift of $\Delta T_c\approx0.5$ K from the independently determined T_c in the thermal-scan method. To prevent this variability in T_c we decrease the number of free parameters by fixing, in addition to Δ, the value of A obtained in the above procedure and we get $B=1.67(2)$, $\beta=0.367(3)$, $T_c=1042.82(25)$ K with T_c consistent with the thermal-scan value but with a larger error. It should be observed here that the value of the leading amplitude B is in perfect agreement, as it should be, with that obtained previously from the asymptotic behavior, where B remained constant. This gives more confidence in our determination of the universal β value obtained from the correction-to-scaling form. This value of β is in excellent agreement with the theory.[4] It is different from β_{eff} in accordance with Eq. (4). By using this equation and $t\simeq2\times10^{-2}$ at which β_{eff} begins to become constant when it approaches T_c, we get $\beta_{\text{eff}}=0.396(5)$, which is very close to β_{eff} obtained from the single power law near T_c (see Fig. 3). As a further check on our β measurement, we have made a range of fits to Eq. (2) with fixed $A=-0.458$ and $\Delta=0.55$, and with $t_{\max}$ successively reduced. This is similar to what we did in the case of the single power law. The results are shown in Fig. 7. It can be seen that β remains constant as long as correction to scaling is appreciable and tends toward β_{eff} when corrections become very small.

A comparison of the β value in this work to theory and of β_{eff} to other $(n,d)=(3,3)$ ferromagnets[40–48] appears in Table II. In constructing the table we have selected only those results with significant data in the region $t\leq10^{-2}$. The exclusion of results based only on data in $t\geq10^{-2}$ is justified by the strong likelihood that asymptotic power-law behavior

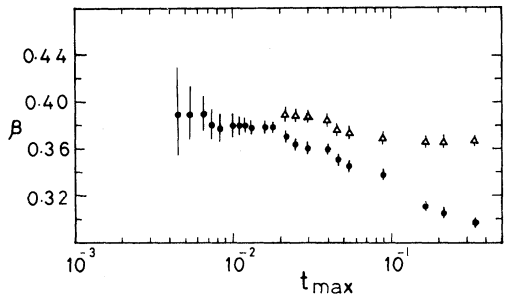

FIG. 7. The variation of the critical exponent β (triangle points) with maximum reduced temperature t_{max} as obtained from the correction-to-scaling form compared with the variation of β_{eff} (circle points) with t_{max} as obtained from single power law. Notice how β converges toward β_{eff} as the correction term starts to become very small at $t \simeq 10^{-2}$.

then fails[38,39] as we have shown above. Under the constraint applied in constructing Table II, we conclude that our value of β_{eff} in Fe is in good agreement with other $(n,d)=(3,3)$ ferromagnets, with the exception of EuS. The consistency of our β values obtained from the single power law and the correction-to-scaling form also indicates the correctness of our method of analysis. The significance of the difference between our result and bulk measurements of Arrot *et al.*[48] is difficult to evaluate since no error is given for the bulk value. The disagreement between our results and recent NMR measurements by Shaham *et al.*[10] (see Table II) in pure Fe is due to the fact that NMR measurements are not close enough to T_c and that the data were fitted to a single power law outside the asymptotic region. To show this we have fitted our data to a single power law within the same range used by Shaham *et al.* and we obtained $\beta_{eff}=0.320(4)$, $B=1.35(5)$ for $4.5\times10^{-3} \leq t \leq 5.3\times10^{-2}$ as compared to Shaham's results $\beta=0.320(4)$, $B=1.35(5)$ for $4\times10^{-3} \leq t \leq 5\times10^{-2}$. This illustrates the fact that spurious results occur when one overlooks the appropriate use of the single power law and the correction-to-scaling form when investigations of critical phenomena in magnetic solids are made. The discrepancy between our results and Preston's can be explained within the same context.

For the determination of the universal ratios among the correction amplitudes in Fe, one needs, in

TABLE II. β_{eff} values for (n,d) = Heisenberg ferromagnets.

Material	Method [a]/probe	Range in t (10^{-4} 10^{-3} 10^{-2} 10^{-1})	β_{eff}	Reference
EuO	NS/bulk		0.36(1)	40
	Kp/bulk		0.368(5)	41
$CrBr_3$	FR/SES		0.368(5)	42
EuS	FR/SES		0.335(10)	43
Ni	ME/^{57}Fe		0.378(10)	44
	PAC/^{100}Rh		0.385(5)	45
	PAC/^{111}Cd		0.383(4)	38
	NMR/^{61}Ni		0.354(14)	46
	Bulk/SES		0.378(4)	47
Fe	ME/^{57}Fe		0.342(4)	9
	ME/^{57}Fe		0.37(2)	9
	NMR/^{57}Fe		0.320(4)	10
	Bulk		0.368	48
	ME/^{57}Fe		0.371(8)[b]	This work
	ME/^{57}Fe		0.379(4)[c]	This work
	ME/^{57}Fe		0.367(5)[d]	This work
Theory			0.365(1)	4

[a] Abbreviations: bulk/SES = bulk measurements analyzed by equation of state, FR/SES = Faraday rotation of light analyzed by scaling equation of state, KP = kink point measurements, ME = Mössbauer effect, NMR = nuclear magnetic resonance, PAC = perturbed angular correlation of gamma rays, NS = neutron scattering.

[b] Obtained from absorber experiment.

[c] Obtained from source experiment.

[d] Obtained from source experiment and correction-to-scaling form [see Eq. (2)].

addition to our present results, further work on another critical exponent $\lambda_{j\,\mathrm{eff}}$ and λ_j in Fe with the corresponding correction amplitude A_j. We have shown that β_{eff} in Fe is single valued with $10^{-4} \leq t \leq 2\times10^{-2}$ and is probe independent as was reported in recent theoretical work.[39] Hence measurements of critical fluctuations in our source experiment can be made with no effect on the dynamic critical exponent z.

B. Measurement of the exponent z

In previous experiments on Ni conducted in our laboratory at Clark University,[26,27] it was shown that PAC relaxation in $Ni^{100}Rh$ and ME line broadening in $Ni^{57}Fe$ both satisfy the preconditions of Eq. (20) for $t > 10^{-4}$ above T_c. It was also shown that τ_c values deduced via explicit forms of Eq. (18) agree well in the two cases, but yield a temperature dependence describable by $w = 0.7$ instead of $w = 1$ ($z = 2.5$). The latter value is predicted by the theory and supported experimentally by neutron scattering measurements as shown in Table III. In a recent work at Clark University by Lee Chow *et al.* in Ni and Fe, a crossover of z from 2.5 to 2.0 has been observed as $T \rightarrow T_c$. These findings have naturally cast doubt on the applicability of the theory leading to Eq. (20). This discrepancy was resolved by a recent hypothesis by Suter and Hohenemser stating that the exponent z crosses from 2.5 to 2.0 as the wave number $\vec{q}$ of the fluctuations goes to zero. In this case the interactions in different ranges of $\vec{q}$ are different, thus producing different z values. Additional doubt on the results might be raised also by the fact that the above hyperfine interaction experiments have used impurity probes in the host materials. Measurements of z in pure Fe have been done recently by Shaham *et al.*,[10] who used NMR methods, and obtained $z = 1.95(18)$ in the temperature range $4\times10^{-3} \leq t \leq 1.5\times10^{-2}$ below T_c and in agreement with other hyperfine measurements. Furthermore, for the dynamic critical exponent in Fe, experiments have been done using neutron scattering and results do not agree either with theory or with values obtained for other ferromagnets[49–53] (see Table III). In order to address the above concerns and to compare with previous results for z, we have studied the fluctuations in the $Fe^{57}Fe$ system where probe and host are the same.

Despite many previous ME studies of Fe this is the first case in which evidence of critical fluctuations is seen and interpreted via ME method, perhaps be-

TABLE III. The critical exponent z as measured in different ferromagnets.

Material	Method[a]/probe	z	Range in t	Range in q	Reference
EuO	ME/^{151}Eu	2.80(45)	2×10^{-2}–10^{-1}	[b]	49
	NS/bulk	2.29(3)	0	0.12–0.48	50
	ESR/bulk	2.04(7)	3×10^{-4}–10^{-1}	0	51
Ni	NS/bulk	2.46(25)	0	0.04–0.2	29
	ME/^{57}Fe	2.50(15)	5×10^{-4}–5×10^{-3}	[b]	52
	PAC/^{100}Rh	2.05(10)	10^{-4}–10^{-2}	[b]	26
	ME/^{57}Fe	2.06(30)	10^{-4}–5×10^{-2}	[b]	27
	PAC/^{100}Rh	2.0–2.5[c]	10^{-4}–10^{-1}	[b]	36
	NMR/^{61}Ni	2.01(12)	2×10^{-3}–10^{-1}	[b]	10,46
Fe	NS/bulk	2.7(2)	0	0.05–0.2	30
	ME/^{57}Fe	1.93(18)	10^{-4}–5×10^{-2}	[b]	This work
	PAC/^{100}Rh	2.0–2.5[c]	10^{-3}–10^{-1}	[b]	36
	NMR/^{57}Fe	1.95(18)[c]	4×10^{-3}–10^{-2}	[b]	10
Co	NS/bulk	2.4(2)	0	0.04–0.09	31
	NMR/^{59}Co	2.44(11)	7×10^{-3}–6×10^{-3}	[b]	10,53
Theory		2.0			5,35
		2.5			5

[a] ME = Mössbauer effect, NS = neutron scattering, ESR = electron spin resonance, PAC = perturbed angular correlation, NMR = nuclear magnetic resonance.
[b] Methods measure an integral over all values of the wave vector q.
[c] Crossover was observed.

cause ^{57}Fe has small nuclear g factors and in terms of Eq. (18) is a relatively insensitive probe. For fluctuation measurements we used a source experiment with activity in Fe in the order of 1.5 mCi. The source and the absorber arrangement for this experiment are the same as described above.

The critical temperature was determined by a fit to the hyperfine field in the ferromagnetic region and by thermal scanning, both as discussed above. Results for B, T_c, and β_{eff} averaged over both halves of the MCA were $\beta_{\text{eff}}=0.376(5)$, $B=1.65(4)$, and $T_c=1041.04$ K, and $T_c=1041.0(1)$ K from the thermal-scan method. As in our previous work[27] on Ni^{57}Fe, the phase transition was also signaled by the non-Lorentzian character of the line as T passes through T_c from above as seen in Fig. 8. The agreement between β_{eff} and B with the results of the above section as well as the agreement between the two determinations of T_c gives confidence in our temperature-control technique. The absolute difference in T_c value is due to the use of different thermocouples in the two cases.

Above T_c we defined the line broadening by

$$\Delta\Gamma(T)=\Gamma(T)-\Gamma_0 \ , \tag{22}$$

where Γ_0 is the linewidth measured 10 K or more above T_c. From the average of several observations we obtained $\Gamma_0=0.391(3)$ mm/sec; this compares well with $\Gamma_0=0.389(3)$ mm/sec calculated from resonant thickness broadening theory.[33,34]

Results for $\Delta\Gamma$ vs t are plotted in Fig. 9. A least-squares fit to

$$\Delta\Gamma=Dt^{-w} \tag{23}$$

with T_c fixed at $T_c=1041.04$ and where $t=(T-T_c)/T_c$, and D is a constant, yields the results shown in Table III. As a check we removed points from both extremes of reduced temperature as well as points with large scatter and observed no significant change in w. Our final results are: $w=0.62\pm0.13$, $D=(7.8\pm8.8)\times10^{-5}$ mm/sec. For three-dimensional Heisenberg ferromagnets, $d=3$, $\nu=\frac{2}{3}$, and $\eta=0$, and from Eq. (14), we arrive at $z=1.93(19)$ which is in good agreement with NMR results in pure Fe and Ni by Shaham *et al.*,[10,46] and the PAC measurements in Fe and Ni by Lee Chow, Suter, and Hohenemser,[36] who used ^{100}Rh as a probe. z agrees well also with our previous ME work on Ni using ^{57}Fe probe.[27] In Table III we list available values of the critical exponent z as measured and predicted in various cases, along with the corresponding temperature range of investigation. To obtain quantitative conversion to correlation time we have used the Brandford-Marshall theory,[22] which in the case of Fe^{57}Fe, implies

$$\tau_c=2.19\times10^{-10}\Delta\Gamma \ , \tag{24}$$

with τ_c in units of sec and $\Delta\Gamma$ in mm/sec. Here we used $H(0)=340$ kG for the Fe system. With this the results for Fe may be compared to results for

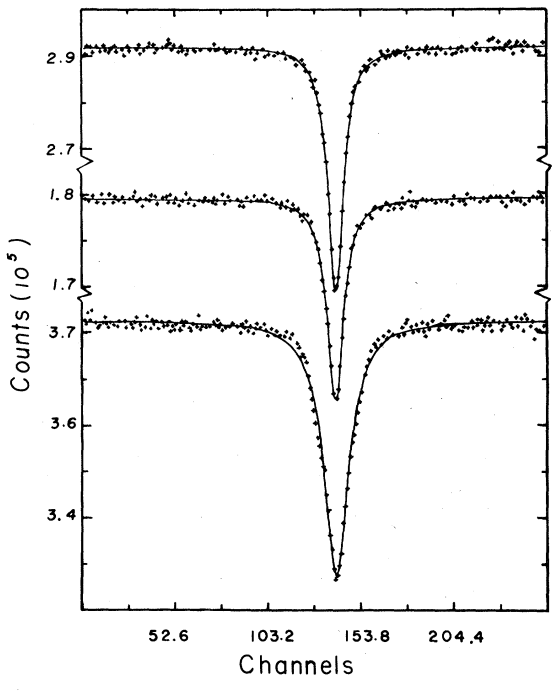

FIG. 8. Typical single-line spectra near T_c in Fe^{57}Fe. All three lines were fitted with Lorentzians. Top: $T-T_c=0.41$ K. Middle: $T-T_c=0.06$ K. Bottom: $T-T_c=-0.29$ K. Observe the failure of the Lorentzian fit below T_c.

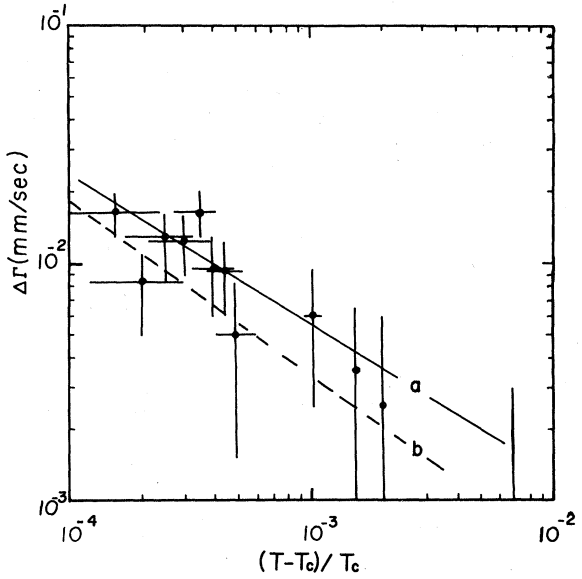

FIG. 9. (a) Line broadening in the Fe^{57}Fe system obtained in this work and (b) line broadening measured in the Ni^{57}Fe system by ME in our previous work (Ref. 27).

$Ni^{100}Rh$ and $Ni^{57}Fe$ as shown in Fig. 10. We notice that the correlation times obtained by ME for both ferromagnets are in good agreement within the error range. On physical grounds this is to be expected because the coupling constants for both systems are very close.

From the above results we conclude that the critical exponent z obtained for Fe via ME is consistent with previous hyperfine exponents measured in Ni and Fe and that autocorrelation times at a given reduced temperature are remarkably close for these cases. This means that deviation of experimental values of z from the theoretically expected result $z=2.5$ is unlikely to be caused by probe disturbance. From Table III we see that only one of the experimental results obtained by ME in Ni with close-in measurements appear to agree with theory; this measurement was seriously disturbed by temperature gradient.[52] The ME value in EuO was measured far from T_c and is strongly affected by inhomogeneous broadening just below T_c.[49] Thus the hyperfine results give $z=2.0$ which is in disagreement with neutron results in Ni, Fe, and Co. From Table III we notice that hyperfine results are sampled in the region $10^{-4} \leq t \leq 10^{-2}$, while neutron results are sampled in the region $10^{-2} \leq t \leq 10^{-1}$. The Co results by NMR are also not sufficiently close to T_c.

To remove the contradiction in the above values for z, Suter and Hohenemser[11] suggested that the observation of z will depend on the size of the asymptotic region in $\vec{q}$ space as we have mentioned above. Thus the hyperfine interaction sampled in the temperature range $10^{-4} \leq t \leq 10^{-2}$ is dominated by sufficiently small $\vec{q}$ and yields asymptotic behavior in accordance with the modified theory on dynamics, where non-spin-conserving forces are included, in addition to the isotropic Heisenberg coupling. Furthermore, neutron scattering experiments for the wavelength range sampled ($0.05 \leq q \leq 0.5$ Å^{-1}) are dominated by nonasymptotic values of $\vec{q}$ and thus give effective values of z that are an indication of Heisenberg or crossover behavior. This hypothesis was confirmed by a recent PAC experiment on Fe and Ni at Clark University[36] and, as in Shaham's NMR measurement in Fe, a crossover was observed.

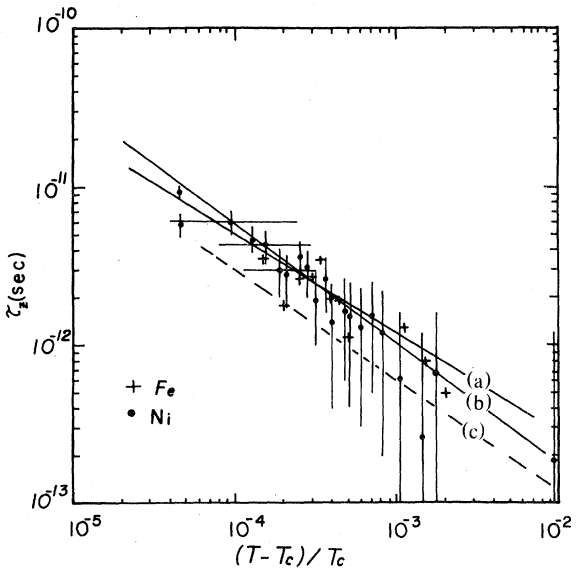

FIG. 10. Autocorrelation times calculated via the theory of Bradford-Marshall. (a) $Fe^{57}Fe$ data (cross points) as obtained in this work, (b) Correlation times (circles) obtained from (ME) in $Ni^{57}Fe$ system (Ref. 27). (c) Dashed line represents the data measured in the $Ni^{100}Rh$ system via PAC (Ref. 26).

C. Energy shift in Fe

Prior to the present work, experimental information on the energy shift in Fe has been obtained by several authors. We have given a detailed report on the shift in Fe in a previous publication.[7] In the following we will give a brief account on our findings with additional quantitative information on the data obtained.

The first quantitative study was done by Preston[12,13] who measured the shift from zero temperature to the Fe γ-phase structural transition. He observed a 0.04-mm/sec step anomaly[13] at T_c and a 0.03-mm/sec discontinuity at the γ-phase transition. The anomaly observed at T_c suggests that the transition is of the first order, which would be at variance with the usual characterization of the Curie point of Fe. Kovats and Walker[54] repeated Preston's measurements in a subsequent experiment and extended the measurements to the δ-phase transition at $T=1673$ K where a discontinuity of 0.07 mm/sec was observed. Kovats and Walker's work was insufficiently detailed to determine whether a shift anomaly exists at T_c. To date, several qualitative efforts have been made to explain the Preston shift anomaly in terms of band changes which in turn affect the isomer shift.[55,56] Prior to this work, attempts to observe the effect of magnetic ordering in Fe on the isomer shift were made by Housely and Hess[57] and subsequently by Nandwani and Puri.[58] The approach in both cases was to calculate theoretically the second-order Doppler shift, subtract it from the total measured shift, and obtain the temperature dependence of the isomer shift. Both works relied on the experimental data of Preston *et al.*[12] In both cases the results were inconclusive because of apparent smallness of the effect and the scatter of the data near T_c.

To investigate the effect of the magnetization on the isomer shift in a wide range of temperature, separate values for the isomer-shift dependence on T must be obtained. The energy shift consists of two parts,[12] the second-order Doppler shift (SOD) and

the isomer shift (IS). Purely on experimental grounds it is impossible to deduce values for each separately. One has to rely on theory to obtain either part from experiment.

For high temperature, as in our measurements, and in the Debye model, the theory gives an expression of SOD as[59]

$$\Delta_D(T) = \left(\frac{3k_BT}{2mc}\right)\left(1 + \frac{1}{20}\frac{(\theta')^2}{T^2}\right), \quad (25)$$

where θ' is the effective Debye temperature given by[60]

$$\theta' = \theta_0(1 + B_0\sigma^2)^{1/2} \quad (26)$$

and depends on the magnetization of the system. B_0 in the above equation is a constant, $0 < B_0 < 1$, θ_0 is the Debye temperature, and σ is the reduced magnetization. Thus the SOD should vary more rapidly with magnetic ordering near T_c. From Eq. (25) it can be seen that for $T > \theta'$, a condition that is reasonable in our case, the effect of the second-order term on $\Delta_D(T)$ is too small to detect. We conclude therefore that any observable effect of the magnetization on the energy shift must come from the isomer shift.

To check on the shift anomaly observed by Preston in the vicinity of T_c, we carefully made measurements with both source and absorber experiments. T_c was determined as in previous sections of this work. We analyzed our data in terms of line positions. The data of the source experiment included two runs with the same foil. Both gave the same results. Values of $\Delta(T)$ near T_c are listed in Table IV. To check on instrumental drifts in the Mössbauer spectrometer, we monitored the drive with a $Ni^{57}Fe$ source during the experiment. By this method it was shown that the drive system was linear to 0.5%. In Fig. 11 we show results for energy shift as obtained by Preston in absorber experiments, and by us in source and absorber experiments. The variable plotted in this figure represents the shift with respect to room temperature. It is obvious from Fig. 11 that the variation of $\Delta(T)$ near T_c can be explained by the slow variation of $\Delta(T)$ over a wide range above and below T_c. We conclude, therefore, that the Preston anomaly cannot be reproduced in either source or absorber experiments.

TABLE IV. Energy shift $\Delta(T)$ near $T_c = 1042.9$ K in $Fe^{57}Fe$ source experiment.

T (K)	Shift (mm/sec)
1040.15	0.524(11)
1041.15	0.524(11)
1041.65	0.522(11)
1042.15	0.524(5)
1042.27	0.525(5)
1042.65	0.523(5)
1042.77	0.524(5)
1042.85	0.527(5)
1042.90 ← T_c	0.531(5)
1043.10	0.528(5)
1043.40	0.531(5)
1043.65	0.522(5)
1045.15	0.528(5)

If the energy shift depends to any extent on the magnetization, it is possible that this dependence is observable over a wide range of temperature and not just at T_c. Although this possibility has been investigated previously with negative results,[57,58] our more extensive (source) data, shown in Table V, permits a new look at the problem.

At high temperature ($T > \theta'$) the SOD is calculable from the first term of Eq. 25. In this way we deduce

$$\Delta_D^{\text{theor}}(T) = (7.29 \times 10^{-4} T - 0.2135) \quad (27)$$

with Δ_D in units of (mm/sec) and T in K, with results shown in Fig. 12. To test whether the high-temperature approximation is correct, we have made least-squares fits to our data below and above T_c us-

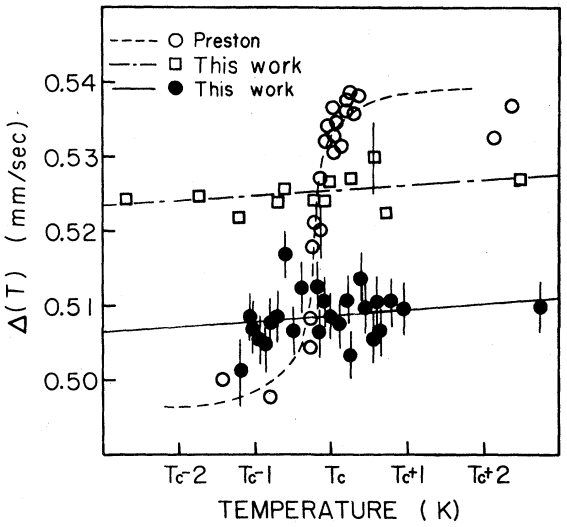

FIG. 11. ME energy-shift measurements near T_c as observed by Preston and in this work. Square points are source experiment results using a K_4 Fe $(CN)_6 \cdot 3H_2O$ absorber at 293 K. Circle points are the results of the absorber experiment using ^{57}Co *pd* source at 293 K. The systematic uncertainty in absolute shifts is around 0.015 mm/sec. Near-horizontal lines indicate shift variation over 100 K on either side of T_c. The dashed line through Preston's data is for visual clarity.

ing the form

$$\delta_t = aT + b \quad . \tag{28}$$

The results obtained for $T < T_c$ and $T > T_c$ were very close. Thus for $820 < T < 1096$ K we obtain

$$\delta_t = (7.06 \pm 0.18) \times 10^{-4} T - (0.21 \pm 0.02) \quad , \tag{29}$$

with δ_t in units of mm/sec , which is very close to Eq. (27). Our confidence in Eq. (27) allows point by point subtraction of $\Delta B^{\text{theor}}(T)$ from $\Delta(T)$ to obtain the value of the isomer shift shown in Fig. 13. We

TABLE V. Energy shift in Fe. The shift is given for $293 \leq T \leq 1096$ K. The results are compared with those obtained in absorber experiments by Preston.

	This work		Preston's work[a]
T(K)	$\Delta(T)$ (mm/sec)	T(K)	$\Delta(T)$ (mm/sec)
293.0	0	293.0	0
684.9	0.271(7)		
		717.6	0.285
		816.0	0.363
820.77	0.367(7)		
867.52	0.401(7)		
		912.5	0.429
947.77	0.455(7)		
978.40	0.475(7)		
986.90	0.486(7)		
994.40	0.489(7)		
1000.15	0.494(7)		
1010.15	0.500(5)		
1016.73	0.496(5)		
1020.15	0.509(5)	1019	0.511
1031.15	0.516(5)		
1038.15	0.516(5)		
1038.55	0.522(7)		
1038.95	0.519(7)	1039.4	0.523
1041.15	0.524(5)		
1041.55	0.524(5)		
1042.15	0.522(5)	1042.1	0.524
1042.65	0.525(5)	1042.8	0.531
1043.10	0.528(5)	1042.4	0.520
1043.40	0.531(5)		
1043.65	0.522(5)		
		1046.1	0.535
1048.15	0.525(3)		
1053.00	0.527(3)	1054.6	0.549
1059.15	0.532(3)		
1060.27	0.534(3)		
1066.15	0.536(3)		
1073.15	0.545(3)		
1079.89	0.552(3)		
		1084.6	0.566
1096.90	0.563(3)		
		1104.7	0.580

[a] Average error in Preston's work is given as 0.003 mm/sec.

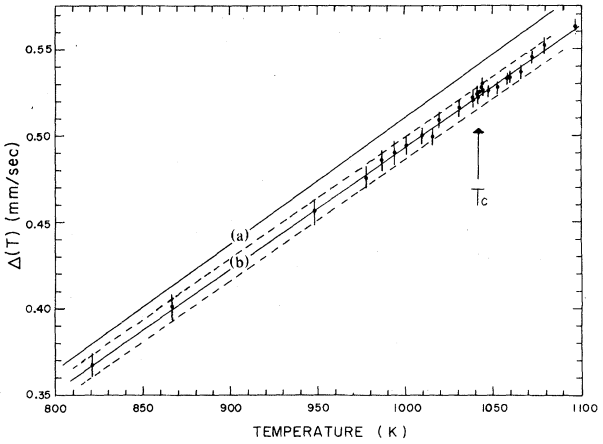

FIG. 12. Energy shift in $Fe^{57}Fe$ in the neighborhood of T_c. Curve (a) indicates the theoretical SOD calculated via the first term of Eq. (27). Curve (b) illustrates a linear fit to total shift measured. The dashed lines show the systematic error.

conclude from this that the effect of magnetic ordering on the isomer shift is unobservable, and that it must be smaller than deducible from similar previous analyses.[57,58] Also we notice from Fig. 12 that the isomer shift is opposite in sign to the SOD. This indicates an increase in charge density at the ^{57}Fe nucleus as the temperature increases. This might result from decreased screening of $4s$ electrons when $3d$ electrons transfer to the $4s$ band via phonon excitation.

Our conclusion is that:

(a) No Preston-like energy shift anomaly can be observed within 2 K of T_c, either via source or absorber experiments.

(b) Using the high-temperature approximation for the second-order Doppler shift we obtain a predicted temperature dependence that is independent of magnetization to within better than 10^{-4} near T_c.

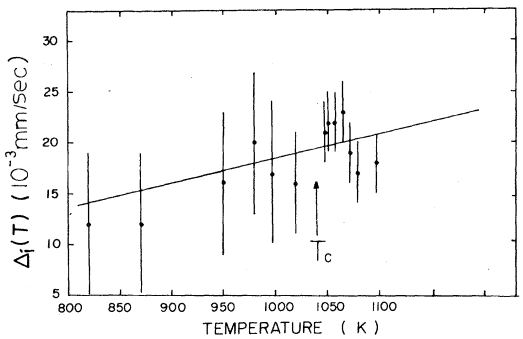

FIG. 13. Isomer shift variation with temperature as obtained by subtraction of the second-order Doppler shift (SOD) from the total shift measured.

(c) Subtraction of the theoretical second-order Doppler shift from the data yields explicit values of the isomer shift which for a broad region around T_c shows no effect of the magnetization to within 0.005 mm/sec.

D. Domain rotations with temperature

Recent Mössbauer studies on amorphous metallic solids have shown thermally induced rotation of the axis of magnetization. The interesting effect was the abrupt change in the domain directions at certain temperatures. The work by Chien[15] on the amorphous $Fe_{80}B_{20}$ and subsequently by Dwynn Lafleur[16] has shown a sudden change of domain direction at low temperature ($T < 230$ K) and at high temperature ($T > 550$ K). The large amount of change at low temperature was related to compressive stress on the foil sample, and after removing such stress the amount of the domain orientation decreased but did not disappear. In his work Chien noticed that demagnetizing effect was not the only factor determining the magnetic-spin structure. Dwynn Lafleur has shown that domain orientation depends on the external stress on the sample as well as on the magnitude and direction of the internal stress in the sample. He derived a model which allows one to calculate the magnitude of the internal stress and its direction with respect to an applied external stress. In this work we have also observed the rotation of the magnetization axis in pure Fe and we report on it in the following.

The splittings of the ground and 14.4-keV states of the ^{57}Fe in a magnetic environment result in six magnetic-dipole transitions. The transition probability for each of the six allowed lines depends on the angle between the quantization direction (i.e., field direction) and the emitted gamma rays. For angle θ between H and $\vec{K}_\gamma$, the intensities for the six lines of ^{57}Fe Zeeman spectrum are[12]

$$\begin{aligned} I_1 &= I_6 = 3(1+\cos^2\theta) \ , \\ I_2 &= I_5 = 4\sin^2\theta \ , \\ I_3 &= I_4 = (1+\cos^2\theta) \ . \end{aligned} \tag{30}$$

The dependence of the intensities on the magnetic orientation is a useful property which can be employed to study the magnetization orientation of the domains containing the Mössbauer probe. Thus from the above equation we obtain

$$I_1/I_2 = \tfrac{3}{4}(1+\cos^2\bar{\theta})/\sin^2\bar{\theta} \ , \tag{31}$$

where the angle $\bar{\theta}$ characterizes the average domain direction. The quantity I_1/I_2 is a measure of domain alignment in ferromagnets, provided that the hyperfine field at the ^{57}Fe nucleus is proportional to the magnetization vector of the domain in which the nucleus is located. This was confirmed in a previous

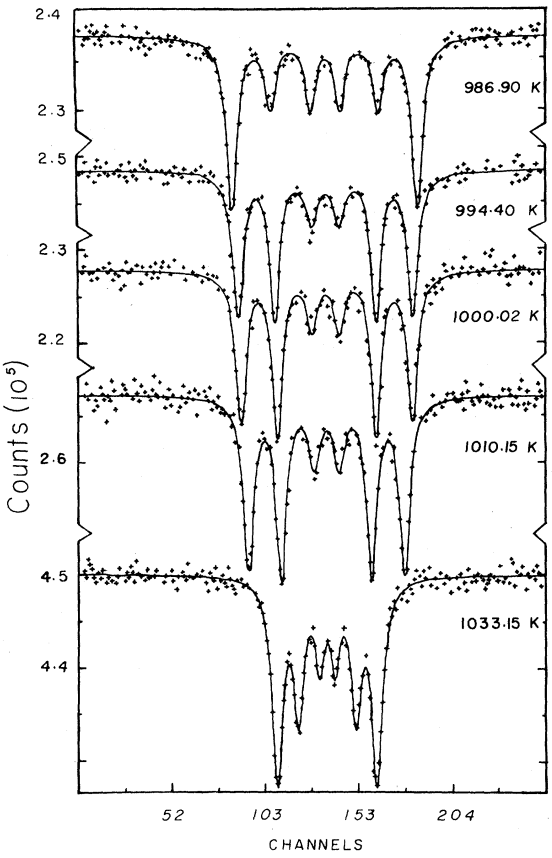

FIG. 14. Variation of line intensity I_2 for an Fe source. ME spectra between 300 and 685 K were similar to $T = 1000.02$ K. Notice the intensity change of the second line at 986.90 K.

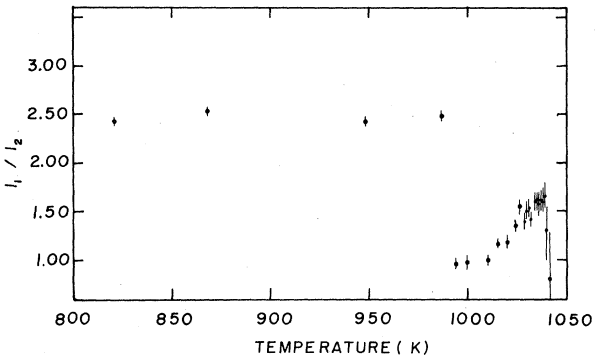

FIG. 15. Behavior of I_1/I_2 as a function of temperature. For 800 K $< T <$ 986.9 K the value I_1/I_2 remained 2.5 and dropped to 1 outside this range. Notice the abrupt change at about 990 K. $I_1/I_2 = 1.5$ corresponds to random domain alignment. $I_1/I_2 \to \infty$ corresponds to domain alignment in a direction parallel to gamma ray. $I_1/I_2 = \frac{3}{4}$ corresponds to domain alignment in a direction perpendicular to the gamma ray.

section of this work. Clearly an observed value of I_1/I_2 does not uniquely specify the domain configuration. On the other hand changes in domain configuration are observable as changes in $\bar{\theta}$ or I_1/I_2.

In this work we have observed variation of I_1/I_2 with temperature as indicated in Figs. 14 and 15. Numerical results for I_1/I_2 and $\bar{\theta}$ are given in Table VI. The remarkable thing is not that I_1/I_2 varies, but that it appears to change abruptly at about 984 K although the foil was annealed before starting the experiment.

This phenomenon is similar to the one observed by Chien in $Fe_{80}B_{20}$ alloy. In our work we did not apply stress on the sample as was done by Dwynn Lafleur in $Fe_{80}B_{20}$. The domain orientation change at $T < 820$ and $T > 994$ K is still puzzling. One plausible explanation of the effect is that the entire source is spanned by one or a very small number of domains, or it may have been subject to an internal stress and that effect involves a sudden reorientation of a single domain or sudden change in the internal stress involving magnetization in the sample. Why internal stress or domain orientation should behave in the manner shown in Fig. 15 remains unexplained.

TABLE VI. Line intensity ratios I_1/I_2 and average domain orientations $\bar{\theta}$ for an Fe source as a function of temperature.

T (K)	I_1/I_2	$\bar{\theta}$
684.90	0.94(4)	72
820.77	2.42(4)	43
867.52	2.54(4)	42
947.77	2.43(4)	43
986.90	2.62(4)	41
994.40	0.95(5)	72
1000.02	0.93(5)	72
1010.15	0.98(5)	70
1016.73	1.11(5)	66
1020.27	1.14(7)	65
1024.15	1.42(7)	56
1026.15	1.57(7)	53
1029.15	1.36(7)	57
1030.15	1.47(7)	56
1031.15	1.50(7)	55
1032.15	1.44(7)	56
1033.15	1.53(7)	54
1034.15	1.58(7)	53
1035.15	1.62(7)	53
1036.15	1.57(7)	53
1037.15	1.66(7)	52
1038.15	1.61(10)	52
1039.15	1.62(14)	52
1040.15	1.40(23)	57
1041.65	0.83(50)	

V. CONCLUSION

We have studied the static and dynamic critical behavior of iron near its Curie temperature and have measured the energy shift over a wide range of temperature. In addition we have observed magnetic-domain reorientation with temperature. Our conclusions are as follows:

(i) The effective critical exponent β_{eff} in the source experiment was found from a single power law to be $\beta_{\text{eff}}=0.379(4)$ for $10^{-3} \leq t \leq 2\times10^{-2}$, which agrees very well with the value obtained in the absorber experiment as $\beta_{\text{eff}}=0.371(8)$ for $10^{-4} \leq t \leq 2\times10^{-2}$, in a wider range of reduced temperature. The amplitude B in both cases was also extracted and found to have the same value $B=1.66(3)$. The above results show that the small ^{57}Co impurity introduced in the source experiment has no effect on our measurements. The difference between our value for β_{eff} and Preston's results and those of Shaham *et al.* is due to our careful determination of the asymptotic critical region near T_c.

We have used in addition the correction-to-scaling form for the reduced magnetization near T_c and obtained the universal exponent β, the leading amplitude B, and the correction amplitude A as $\beta=0.367(5)$, $B=1.67(2)$, $A=-0.458(22)$ for $10^{-3} \leq t \leq 3.4\times10^{-1}$. The fact that B is the same in both cases provides high confidence in the correctness of our results. The values of β_{eff} and β are in excellent agreement with the renormalization-group predictions for $(n,d)=(3,3)$ as well as high-quality experiments on other materials in this class.

(ii) We observed critical fluctuations above T_c and measured the dynamic critical exponent z as $z=1.93(19)$, which agrees very well with Shaham's NMR results in Fe and Ni and the PAC results of Chow, Suter, and Hohenemser in Fe and Ni and our work on Ni using ME and ^{57}Fe. The agreement in the latter case extends beyond the value of z to numerical values of correlation times as a function of temperature. Our results do not agree with neutron data nor with the theory on dynamics with conserved order parameters, but agree with the modified theory where nonconserving forces are included in the spin interactions,[5,35] and with recent hypothesis by Suter and Hohenemser.[11]

(iii) No energy shift discontinuity can be observed within 2 K of T_c either via source or absorber experiments.

Explicit values of the isomer shift were obtained and for a broad region around T_c show no effect of the magnetization to within 0.005 mm/sec. The search for a magnetization dependence of the Mössbauer energy shift in Fe therefore yields negative results in all respects.

(iv) The domain-reorientation dependence on the temperature showed a jump behavior at $T \simeq 684$ K

and at around $T \simeq 994$ K. This jump behavior may be attributed to an internal strain in the sample (magnetostrictive behavior). Just below T_c the domains remained randomly distributed.

ACKNOWLEDGMENTS

I would like to express my appreciation to Dr. C. Hohenemser for encouraging this work and for the helpful comments on this manuscript. I have benefitted from discussions with Dr. A. Gottlieb, Dr. R. Suter, Dr. T. Kachnowski, Dr. L. Chow, and Dr. H. Gould. The assistance of Mr. A. R. Chowdhury in the absorber experiment and the comments on the manuscript of Dr. John Van Zytveld are appreciated. My thanks go to the National Science Foundation Grant No. DMR 76-12168 and No. DMR 77-01250, the Research Corporation, and Yarmouk University for their financial support.

---

*Present address: Department of Physics, Yarmouk University, Irbid, Jordan.

[1]R. B. Griffiths, Phys. Rev. Lett. 24, 1479 (1970).
[2]A. D. Bruce and A. Aharony, Phys. Rev. B 10, 2078 (1974).
[3]S. Ma, *Modern Theory of Critical Phenomena* (Benjamin, New York, 1976).
[4]L. C. LeGuillou and J. Zinn-Justin, Phys. Rev. Lett. 39, 95 (1977).
[5]P. C. Hohenberg and B. I. Halperin, Rev. Mod. Phys. 49, 435 (1977).
[6]M. A. Kobeissi and C. Hohenemser, Hyper. Inter. 4, 480 (1978).
[7]M. A. Kobeissi, L. Chow, and C. Hohenemser, Hyper. Inter. 4, 485 (1978).
[8]M. A. Kobeissi and C. Hohenemser, in *Magnetism and Magnetic Materials—1975,* edited by J. J. Becker, G. H. Lander, and J. J. Rhyne, AIP Conf. Proc. No. 29 (AIP, New York, 1976), p. 497.
[9]R. S. Preston, J. Appl. Phys. 39, 1231 (1968).
[10]M. Shaham, J. Barak, M. El-Hanany, and W. W. Warren, Jr. (unpublished).
[11]R. Suter and C. Hohenemser, Phys. Rev. Lett. 41, 705 (1978).
[12]R. S. Preston, S. S. Hanna, and H. Heberle, Phys. Rev. 128, 2207 (1962).
[13]R. S. Preston, Phys. Rev. Lett. 19, 75 (1967).
[14]R. Rabinovich and Hanan Schechter, J. Appl. Phys. 39, 2464 (1968).
[15]C. L. Chien, Phys. Rev. B 18, 1003 (1978).
[16]L. D. Dwynn Lafleur, Phys. Rev. B 20, 2581 (1978).
[17]H. E. Stanley, *Introduction to Phase Transitions and Critical Phenomena* (Oxford University Press, New York, 1971).
[18]W. J. Camp, D. M. Saul, J. P. Van Dyke, and M. Wortis, Phys. Rev. B 14, 3990 (1976).
[19]A. Aharony and G. Ahlers, Phys. Rev. Lett. 44, 782 (1980).
[20]M. Chang and A. Houghton, Phys. Rev. Lett. 44, 785 (1980).
[21]H. Wegner, Z. Phys. 186, 498 (1965).
[22]E. Bradford and W. Marshall, Proc. Phys. Soc. London 87, 731 (1966).
[23]M. B. Blume and Tjon, Phys. Rev. 165, 446 (1968).
[24]A. M. Afanas'eve and V. D. Gorobschenko, Sov. Phys. JETP 39, 690 (1974).
[25]D. A. Shirley, S. S. Rosenblum, and E. Matthias, Phys. Rev. 170, 363 (1968).
[26]A. M. Gottlieb and C. Hohenemser, Phys. Rev. Lett. 31, 1222 (1973).
[27]M. A. Kobeissi, R. Suter, A. M. Gottlieb, and C. Hohenemser, Phys. Rev. B 11, 2455 (1975).
[28]M. F. Collins, V. J. Minkiewicz, R. Nathans, L. Passel, and G. Shirane, Phys. Rev. 179, 417 (1969).
[29]V. J. Minkiewicz, M. F. Collins, R. Nathans, L. Passel, and G. Shirane, Phys. Rev. 182, 624 (1969).
[30]V. J. Minkiewicz, Int. J. Magn. 1, 149 (1971).
[31]C. J. Glinka, V. J. Minkiewicz, and L. Passel, Phys. Rev. B 16, 4084 (1977).
[32]M. A. Kobeissi and C. Hohenemser, Rev. Sci. Instrum. 49, 601 (1978).
[33]S. Margulies and J. R. Ehrman, Nucl. Instrum. Methods 12, 131 (1961).
[34]S. Margulies, P. Debrunner, and H. Frauenfelder, Nucl. Instrum. Methods 21, 217 (1963).
[35]G. B. Teitelbaum, JETP Lett. 21, 154 (1975).
[36]L. Chow, C. Hohenemser, and R. Suter, Phys. Rev. Lett. 45, 908 (1980).
[37]H. H. Potter, Proc. R. Soc. London Ser. A 140, 362 (1934).
[38]C. Hohenemser, T. Kachnowski, and T. Bergstresser, Phys. Rev. B 13, 3154 (1976).
[39]T. K. Bergstresser and H. Gould, J. Phys. C 12, 2611 (1979).
[40]J. Als-Nielsen, O. W. Dietrich, and L. Passel, Phys. Rev. B 14, 4908 (1976).
[41]N. Menyuk, K. Dwight, and T. B. Reed, Phys. Rev. B 3, 1689 (1971).
[42]J. T. Ho and J. D. Litster, Phys. Rev. Lett. 22, 603 (1969).
[43]D. D. Berkner and J. D. Litster, Phys. Rev. Lett. A 54, 396 (1975).
[44]H. C. Benski, R. C. Reno, C. Hohenemser, R. Lyons, and C. Abelado, Phys. Rev. B 6, 4266 (1972).
[45]R. C. Reno and C. Hohenemser, Phys. Rev. Lett. 25, 1007 (1970).
[46]M. Shaham, J. Barak, U. El-Hanany, and W. W. Warren, Jr., Phys. Rev. Lett. 39, 570 (1977).
[47]J. S. Kouvel and J. B. Comely, Phys. Rev. Lett. 20, 1237 (1968).
[48]A. S. Arrott, B. Heinrich, and D. S. Bloomberg, in *Magnetism and Magnetic Materials—1972,* edited by C. D. Graham and J. J. Rhyne, AIP Conf. Proc. No. 10 (AIP, New York, 1973), p. 941.
[49]G. Groll, Z. Phys. 243, 60 (1971).
[50]O. W. Dietrich, J. Als-Nielsen, and L. Passel, Phys. Rev. B 14, 4923 (1976).
[51]R. A. Dunlap and A. M. Gottlieb, Phys. Rev. B 22, 3422 (1980). See also R. A. Dunlap, Ph.D thesis (Clark

University, 1980) (unpublished).
[52]D. Gumprecht, P. Steiner, G. Crecelius, and S. Hufner, Phys. Lett. A 34, 79 (1971).
[53]M. Shaham, J. Barak, U. El-Hanany, and W. W. Warren, Jr., Solid State Commun. 29, 835 (1979).
[54]T. A. Kovats and J. C. Walker, Phys. Rev. 181, 610 (1969).
[55]S. Alexander and D. Treves, Phys. Lett. 20, 134 (1966).
[56]R. Ingalls, Phys. Rev. 155, 157 (1967).
[57]R. M. Housely and F. Hess, Phys. Rev. 164, 340 (1967).
[58]S. S. Nandwani and S. P. Puri, J. Phys. Chem. Solids 34, 711 (1973).
[59]G. K. Wertheim, D. N. Buchanan, and H. J. Guggenheim, Phys. Rev. B 2, 1392 (1970).
[60]Sh. Sh. Bashkirov and G. Ya Selyutin, Phys. Status Solidi 26, 253 (1968).