

Probe independence of hyperfine critical exponents*

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The critical exponent β for magnetically ordered materials has been obtained from a variety of hyperfine experiments, such as nuclear magnetic resonance, perturbed angular correlations, and Mössbauer effect. In this paper we discuss probe disturbance effects on hyperfine measurements of β . We consider both chemically pure substances and materials into which the hyperfine probe has been introduced as a dilute impurity, and emphasize the latter. From a theoretical point of view, we investigate several molecular-field models and present results of new calculations for an isolated nonmagnetic impurity in a three-dimensional Ising model. It is found that the disturbance produced by the impurity is substantially smaller in the latter case than for the corresponding molecular-field model. From an experimental point of view we present a survey of cases for which bulk and hyperfine measurements have been made on the same substance. We also report on a reanalysis of data in Ni, and present the results of power-law fits made for various ranges of reduced temperature. The cases studied include $Ni^{100}Rh$, $Ni^{111}Cd$, $Ni^{57}Fe$, and $Ni^{181}Ta$, and are restricted to samples produced by diffusion or melting. On the available theoretical and experimental evidence, we conclude that values of β determined from hyperfine measurements involve probe-disturbance errors that are certainly smaller than 2% if the reduced temperature is restricted to $t < 10^{-2}$ and care is taken to avoid source inhomogeneities and temperature gradients.

I. INTRODUCTION

The universality hypothesis^{1,2} states that static critical exponents depend on only a few relevant variables. At one time it might have been hoped that for interactions that are not long range, only lattice dimensionality and effective spin dimensionality are relevant. More recent work, particularly by the renormalization-group method,³ shows that other factors such as dipolar or cubic interactions affect critical exponents, generally by small amounts.

One desires, therefore, reliable detection of small differences in static exponents to an accuracy of (1–5)%. Discrepancies between experiment and present theory may in this way lead to the discovery of as yet unanticipated relevant variables. The challenge to the experimentalist is twofold: (a) to look for small differences in measured exponents; (b) to be concerned more than ever with the validity of the method used. In this paper we concern ourselves with the second problem as it applies to critical exponents measured by hyperfine fields.

In particular, we consider available theoretical and experimental evidence relating to possible probe disturbance of hyperfine exponents. We restrict ourselves to cases for which the impurity concentration is sufficiently small to avoid significant impurity-impurity interactions. Theoretically, we shall treat isolated impurities. Experi-

mentally we shall show that for impurity concentrations less than 10^{-4} the critical exponent β does not exhibit disturbance for measurements in the reduced temperature range $10^{-4} \leq t \leq 10^{-2}$.

Because available data are largely limited to the exponent β , we focus our discussion accordingly. Reduced hyperfine fields $h(T) = H_{\text{hf}}(T)/H_{\text{hf}}(0)$, as measured by nuclear magnetic resonance, perturbed angular correlations, and Mössbauer effect, have been used to determine β since the work of Heller and Benedek.⁴ Fundamentally, β is defined by the asymptotic expression

$$\lim_{H \rightarrow 0^+} \sigma(t, H) \equiv \sigma_s(t) \approx B t^\beta, \quad (1)$$

where σ , H , and $t = 1 - T/T_c$ are the reduced magnetization, magnetic field, and reduced temperature, respectively, and T_c is the critical temperature. If the reduced hyperfine field is fitted by

$$h \approx B' t^{\beta'}, \quad (2)$$

then the absence of probe disturbance requires $\beta = \beta'$ and is expressible by the condition

$$\mu(t) \equiv h(t)/\sigma_s(t) \approx B'/B = \text{const.} \quad (3)$$

The advantages of hyperfine techniques are well known: For antiferromagnets they make accessible the staggered magnetization; for ferromagnets, they permit determination of the spontaneous magnetization without assumptions about the magnetic equation of state near T_c . Hyperfine mea-

measurements have been criticized because probe-disturbance effects have not been ruled out. In particular, Eq. (3) has not been proven near T_c and is, in fact, known to fail in many cases well below T_c . The most dramatic failures occur for certain impurities dissolved in ferromagnets,^{5,6} but failures also occur in many other cases, including quite possibly chemically pure systems.⁷ For antiferromagnets, failure of Eq. (3) is difficult to investigate since h and σ_s (the staggered magnetization) can not be independently determined.

II. THEORETICAL EVIDENCE

An impurity disturbs the local magnetic environment in a magnetically ordered host. This suggests that one might expect a different temperature dependence of the local magnetization than in an undisturbed host. Since h at an impurity nucleus is determined by the local magnetization, h should have a similarly altered temperature dependence. We consider this problem here for the case of an isolated impurity in a ferromagnetic host.

A. Mean-field theories

Several models have been proposed in the past^{5,8-13} to describe $\mu(t)$ for isolated impurities in ferromagnetic hosts well below T_c . In these theories, the impurity induced deviations of the local host magnetization are treated through mean-field assumptions, and the exact $\sigma_s(T)$ is put in as a parameter. One test of the degree of probe disturbance consists of extrapolating these theories into the critical region. A brief report of this has been given previously.¹⁴

The results of such extrapolations are shown in Fig. 1. For magnetic impurities in ferromagnets, the models of Shirley, Rosenblum, and Matthias⁵ and the model of Campbell¹¹ are evaluated, both for the case of $Ni^{99}Ru$. This was chosen because it is one of the well-known cases showing maximum deviation of h from σ_s well below T_c . For diamagnetic impurities, we have used the model of Lovesey and Marshall¹⁰ as applied to Ni. We conclude from Fig. 1 that compared to σ_s , all three models show relatively weak temperature variation in $\mu(t)$. For nonmagnetic impurities, in a realistic computer fit to data, a positive systematic error ($\beta' > \beta$) of $\sim 5\%$ would be made for any measurement in the range $10^{-4} \leq t \leq 10^{-1}$. For magnetic impurities, the systematic error would be less than 1% for $10^{-4} \leq t \leq 10^{-2}$ and $\sim 2\%$ for $10^{-3} \leq t \leq 10^{-1}$.

These calculations leave much to be desired since there is considerable doubt that any mean-

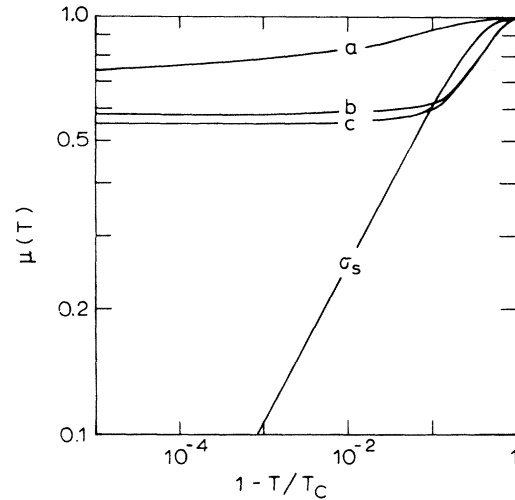


FIG. 1. Behavior of $\mu(t)$ as calculated for three mean-field models, and comparison to $\sigma_s(t)$, the magnetization of Ni. (a) Lovesey-Marshall model, fcc lattice, (Ref. 10): The behavior of $\mu(t)$ represents the maximum possible effect, and occurs only if h is produced by nearest-neighbor host atoms. (b) Shirley, Rosenblum, and Matthias (Ref. 5) extension of the model of Jaccarino, Walker, and Wertheim, with parameters chosen to fit the case $NiRu$. (c) Campbell's model (Ref. 11), with parameters chosen to fit the case of $NiRu$.

field theory has validity in the critical region even if augmented by input of $\sigma_s(T)$. Also, the expected systematic error for a nonmagnetic impurity is undesirably large if one wishes to determine critical exponents unambiguously to an accuracy of 1%. For these reasons we are led to investigate a model which does not make use of a mean-field approximation, with results as described below.

B. Three-dimensional Ising model

We model the ferromagnetic host by an Ising Hamiltonian for a simple cubic lattice, nearest-neighbor exchange, and spin $\frac{1}{2}$. This is the most realistic model Hamiltonian that was found calculable in a relatively straightforward manner. An isolated, nonmagnetic, substitutional impurity is placed at one lattice site. The major determinant of the zero-frequency component of $h(t)$ at the impurity nucleus is the statistically averaged net spin of the nearest-neighbor host atoms, which we denote by $\sigma_1(t)$. We assume $h(t) \propto \sigma_1(t)$ so that

$$\mu(t) = \sigma_1(t) / \sigma_s(t). \quad (4)$$

We then determine the temperature dependence of $\mu(t)$ from a calculation of $\sigma_1(t)$.

The essential ingredient of the calculation is to relate statistical averages in the lattice with defect to the perfect lattice correlation functions.

In broad outline, this is analogous to the simpler and well known relation of the susceptibility to the two-point correlation functions. To obtain the necessary perfect lattice correlation functions, the existing literature on low-temperature series expansions in the critical region has been used.

Details of the above described calculations are being published separately.¹⁵ To illustrate the results, we present $\mu(t)$ in Fig. 2. For comparison, we also show sc and fcc lattice results for the mean-field model of Lovesey and Marshall.¹⁰ We draw conclusions as follows: (i) The mean-field calculation is qualitatively correct in that $\mu(t)$ is slowly varying and its limit as t approaches zero is not zero. (ii) The mean-field calculation overestimates the depressing effect of the impurity on the neighboring spins. (iii) Most importantly, the mean-field calculation overestimates the lack of constancy of $\mu(t)$ in the critical region.

Though model dependent, the results of Fig. 2 suggest that probe disturbance of hyperfine critical exponents is generally less than estimated from molecular-field calculations. For $10^{-4} \leq t \leq 10^{-2}$, the systematic error in β arising in a realistic computer fit is less than 1% and even for $10^{-3} \leq t \leq 10^{-1}$ it is small. As will be seen in more detail below for $10^{-2} \leq t \leq 10^{-1}$, the power law for σ_s will begin to fail significantly, and hence in this region, departure of h from power-law behavior involves more than probe-disturbance effects.

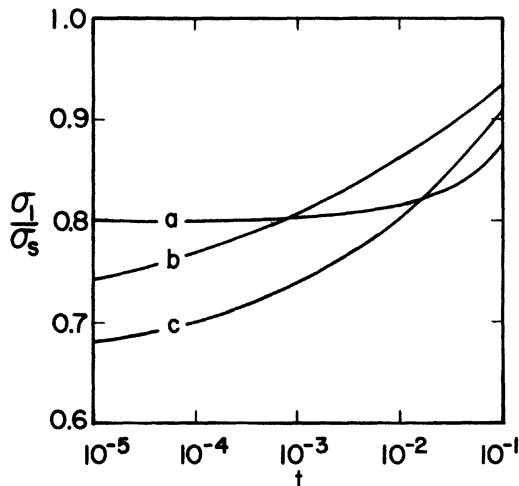


FIG. 2. Model calculation of probe disturbance. (a) Spin- $\frac{1}{2}$ Ising model, nonmagnetic impurity, sc lattice, as described in the text; (b) and (c) molecular field, nonmagnetic impurity, fcc and sc lattice as described in Ref. 10.

III. EXPERIMENTAL EVIDENCE

The various models of h that we have discussed, in particular the Ising model, suggest the following hypothesis. For chemically pure materials and materials that are probed by an isolated impurity, hyperfine exponents are, within experimental error, equal for different probes and equal to bulk exponents for the host. The word "equal" is meant in the asymptotic sense. Implicitly this requires an investigation of the range of t over which the asymptotic region extends.

A. Relevant experiments

That isolated impurities do not disturb critical behavior was first suggested by the work of Wertheim, Guggenheim, and Buchanan,¹⁶ who showed by Mössbauer measurements that h at dilute Fe^{2+} in MnF_2 displays the same critical behavior as the F resonance measured earlier by Heller and Benedek with nuclear magnetic resonance.⁴ Results for this case, as well as all other cases for which, to our knowledge, two or more measurements of β are available, appear in Table I. Experiments on chemically pure materials as well as impurity hyperfine probes are included. In regard to the latter we attempt to restrict ourselves to cases for which probe atoms are free of impurity-impurity interactions and free of disturbance from nearby lattice defects. This implies restriction to well-annealed dilute probe-host systems produced by diffusion or melting, and excludes sources made of nondilute alloys, or those produced by in-beam techniques¹⁷ and by ion implantation. Certainly the first, and probably the second and third source techniques, violate the assumptions made in the theoretical treatment of probe disturbance discussed above, and thus will not be a good test of our hypothesis.

In addition to the comment on MnF_2 above, we may make the following observations based on Table I:

(a) For CrBr_3 , EuO , and EuS , all isotropic insulating ferromagnets, bulk and hyperfine measurements agree very well. (in this case, as in others below, we regard Faraday rotation as a bulk technique.) The possible exception to agreement is the slightly low Mössbauer result on EuO . These data were characterized by severe inhomogeneous broadening and an extremely small reduced temperature range.

(b) For FeF_2 , an insulating antiferromagnetic, hyperfine measurements on two different probes are in excellent agreement. Bulk measurements are, of course, difficult to obtain directly.

(c) For YIG, a nearly isotropic ferromagnet, two bulk measurements and Mössbauer results are

TABLE I. Comparison of hyperfine and bulk results.

Material	Method*/probe	β	B	Range of $1-T/T_c$					Ref.
				10^{-5}	10^{-4}	10^{-3}	10^{-2}	10^{-1}	
MnF ₂	NMR/ ¹⁹ F	0.333 (3)	1.200 (4)						a
	ME/ ⁵⁷ Fe	0.327 (12)	1.49 (5)						b
CrBr ₃	FR	0.368 (5)		c					
	NMR/ ⁷⁹ Br	0.365 (5)	1.32 (7)	d					
EuO	KP	0.368 (5)		e					
	ME/ ¹⁵¹ Eu	0.34 (2)	1.14 (4)	f					
EuS	FR	0.335 (10)	1.13	g					
	NMR/ ¹⁵³ Eu	0.330 (15)	1.145 (20)	h					
FeF ₂	ME/ ⁵⁷ Fe	0.325 (5)	1.36 (3)	i					
	NMR/ ¹⁹ F	0.320 (1)	1.341 (5)	j					
YIG	FR	0.370 (5)	0.875	g					
	bulk	0.380 (5)		k					
	ME/ ⁵⁷ Fe	complex behavior		l					
Ni	bulk	0.378 (4)	1.422	m					
	bulk	0.385		n					
	ND	0.385 (10)		o					
	KP	0.398 (10)		k					
	KP	0.346 (7)		p					
	PAC/ ¹⁰⁰ Rh	0.385 (5)	1.28 (2)	r					
	ME/ ⁵⁷ Fe	0.38 (1)		s					
	ME/ ⁵⁷ Fe	0.378 (10)	1.56 (6)	t					
	PAC/ ¹⁸¹ Ta	0.417 (10)	1.35 (5)	u					
PAC/ ¹¹¹ Cd	0.383 (4)	1.31 (1)	v						

*Abbreviations used: FR—Faraday rotation; NMR—nuclear magnetic resonance; ME—Mössbauer effect; PAC—perturbed angular correlations; ND—neutron depolarization; KP—kink-point method of bulk magnetization.

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^v See Ref. 22.

available. The latter show different β values for the two sublattices as well as a transition from $\frac{1}{3}$ to a significantly higher value at $t=10^{-2}$ in each case. Because of the presence of quadrupole interactions in addition to two hyperfine interactions, the data present a formidable interpretational problem. Neither of the bulk data show evidence of a double power law, and the Faraday rotation result (Table I, Ref. g) explicitly demonstrates that the two sublattices have the same critical behavior.

(d) For Ni, a metallic ferromagnet, five bulk measurements (we include neutron depolarization) and five hyperfine measurements involving isolated impurity probes exist. Agreement among the ten numbers is excellent with two exceptions: one of the kink-point results (Table I, Ref. p), and the ^{181}Ta perturbed-angular-correlation result (Table I, Ref. u). Given the extrapolation difficulties discussed in Table I, Ref. p, and other bulk data to the contrary, we feel justified in discounting the former. The case of Ni^{181}Ta will be discussed further below; it may well suffer from impurity-impurity interactions and, hence, may not belong in Table I at all.

(e) The range of t over which β has been measured varies substantially. If our model calculations can be a guide, data that are predominantly in the range $t \geq 10^{-2}$ must be regarded with some caution; i.e., they may produce erroneous values of β because the asymptotic region is not reached.

(f) All systems in Table I are characterized by lattice dimensionality $d=3$, but may have different effective-spin dimensionality n . If EuS is excluded, the results fall into two classes: (i) more or less isotropic materials (CrBr_3 , EuO , YIG, and Ni) with $\beta \approx 0.37$; (ii) strongly anisotropic materials with spin alignment along a single crystallographic axis (MnF_2 and FeF_2) with $\beta \approx 0.33$. The results (i) and (ii) are in rough agreement with theoretical predictions for universality classes $(d, n) = (3, 3)$ and $(d, n) = (3, 1)$ for which β is predicted to be 0.38 and 0.31, respectively.³ The case EuS, an isotropic system, remains a puzzle. It is possible that β is disturbed because the data is not sufficiently asymptotic.

B. Reanalysis of Ni hyperfine data

As a further test of the effect of impurities, we present below a detailed reanalysis of data on Ni^{100}Rh , Ni^{57}Fe , Ni^{181}Ta , and Ni^{111}Cd . Results on all cases have been published previously, as indicated in Table I. In reanalysis we were particularly interested in understanding the apparently deviant β value for Ni^{181}Ta , which stands in apparent clear violation of our central hypothesis.

To determine the asymptotic region empirically, three parameter least-squares fits were made to Eq. (2) in which β' , B' , and T_c were treated as free parameters. For each material several fits were made in which points were successively removed on the low end of the temperature scale. In all cases, the data were weighted with the inverse square of the statistical error in the hyperfine field.

If our hypothesis of probe independence is correct, the above method should yield constant values of β' , B' , and T_c independent of the range of t , provided the range is sufficiently restricted, and should show deviation of effective values of all three critical parameters as the asymptotic region is exceeded.

Determining deviations from power-law behavior by the method described may be compared to the method of the misfit parameter introduced by Heller.⁴ In the latter, deviations of individual data points from the best power-law fit are plotted against $\log_{10} t$. It is our finding that the method of Heller, which was actually used in some of the previously published analyses of the Ni data (Table I, Refs. r and t), is roughly equivalent to the present method, but that it is more unwieldy when one is interested in testing ten or more ranges of t . In addition we have found that varying the range of fit is capable of elucidating deviations from power-law behavior very close to T_c which were obscured by the Heller misfit analysis used previously.

Unlike the work on MnF_2 ,^{4,16} all experiments discussed below were done without absolute calibration of the temperature scale. Hence, to within 2 K, no significance should be placed on absolute values of T_c . The impurity concentration for all four cases to be discussed has been reported to be less than 1×10^{-4} , and for one Ni^{181}Ta source is estimated at 3 ppm.

1. Ni^{100}Rh

The system Ni^{100}Rh represents an example of an impurity probe which probably possesses a local magnetic moment in the ferromagnetic state. This can be concluded from the magnitude of the hyperfine field, which is far larger than the conduction-electron polarization would predict.⁵ A room-temperature measurement was first made by Matthias *et al.*¹⁸ and some data above T_c have been reported by Rosenblum.¹⁹ Data for $77 \leq T \leq T_c$ were obtained by Reno.²⁰ A selection of these are reproduced in Fig. 3(a) and show that $h(t)$ deviates from $\sigma_s(t)$ by $\sim 10\%$ at $t=0.1$. Data in the critical region have been reported by Reno and Hohenemser²¹ (Table I, Ref. r).

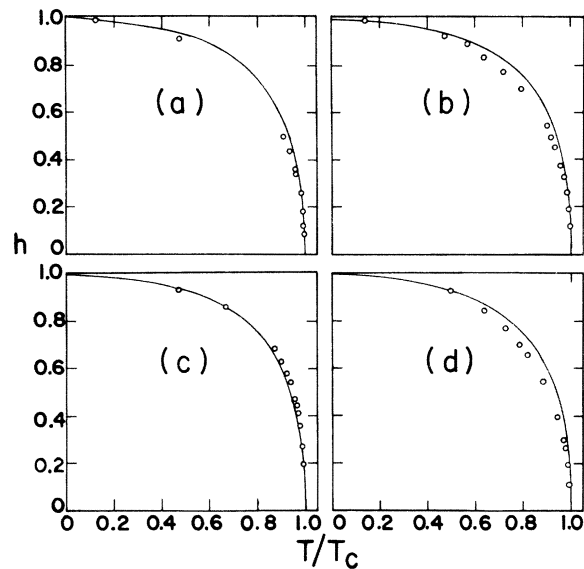


FIG. 3. Reduced hyperfine field (circles) below the critical point for various hyperfine probes in Ni. (a) $Ni^{100}Rh$ (Ref. 20); (b) $Ni^{111}Cd$ (this work); (c) $Ni^{57}Fe$ (Ref. 24); (d) $Ni^{181}Ta$ (Ref. 34). Solid line is magnetization of pure Ni.

Table II and Fig. 4(a) summarize the critical-region data as reanalyzed for various temperature ranges. The values of β' , B' , and T_c are to within statistics independent of the range of t ; and the observed increase in errors of critical parameters as t_{\max} is reduced is a consequence of fitting over a reduced range. The result for β' agrees with bulk values, and the result for the full range agrees to within 1% with the result previously quoted.²¹ The difference is attributable to difference in method of analysis.

2. $Ni^{111}Cd$

The system $Ni^{111}Cd$ is a case of a diamagnetic-impurity probe for which nearly all of the hyperfine field is accounted for by conduction-electron polarization. The noncritical region was first studied by Shirley, Rosenblum and Matthias⁵ and described by the molecular-field model of Lovesey and Marshall.¹⁰ More recently the system has been studied by Kachnowski, Gottlieb and Hohenemser, below, near, and above the Curie temperature. A preliminary account of this work has appeared previously.²² Data for the noncriti-

TABLE II. Analysis of $Ni^{100}Rh$.^a

Source ^b	T_c (K)	$10^3 t$ ^d	ω_L (rad/sec)	β' ^e	T_c ^e	B' ^e
1	575.60	87.8	1153.0 (110)	0.383 (2)	630.92 (1)	1.26 (1)
1	591.47	62.5	1006.9 (81)	0.382 (2)	630.92 (1)	1.26 (1)
1	606.58	38.6	836.6 (56)	0.382 (2)	630.92 (1)	1.26 (1)
1	610.33	32.6	786.4 (90)	0.382 (2)	630.92 (1)	1.26 (1)
2	619.16	18.6	634.7 (26)	0.382 (3)	630.92 (1)	1.26 (2)
1	620.66	16.3	599.5 (52)	0.381 (3)	630.91 (1)	1.25 (2)
2	622.22	13.8	563.0 (20)	0.381 (4)	630.91 (1)	1.25 (2)
2	625.30	8.91	477.4 (15)	0.381 (5)	630.91 (1)	1.25 (3)
1	626.84	6.47	420.8 (26)	0.376 (6)	630.91 (1)	1.22 (3)
2	627.43	5.33	396.2 (15)	0.376 (8)	630.90 (2)	1.22 (5)
2	629.49	2.27	281.8 (20)			
2	630.19	1.16	216.7 (12)			
2	630.40	0.82	192.7 (10)			
2	630.77	0.24	115.9 (35)			

^a An analysis of these data has been previously published in Ref. 21. Values of ω_L and T_c have been conservatively selected from Ref. 20, so as to avoid all possibility of disturbances of external magnetizing fields.

^b Source 1 data were measured without magnetizing field, source 2 had magnetizing fields of 2 and 6 G. These fields were shown in Ref. 20 to have negligible effect on the observed critical behavior.

^c For a particular source, temperatures were reproducible to 0.03 K. The measurement sensitivity was 0.01 K. From one source to the next, the absolute temperature scale differed somewhat, presumably because of differences in thermocouple calibration. For sources 2 and 1, T_c was determined to be 630.92 (2) and 631.06 (4) K, respectively, using separate power-law fits. A single temperature scale is obtained for the two sources by subtracting 0.14 K from source 1 temperatures.

^d The reduced temperatures given are based on $T_c = 630.92$ K.

^e Values of β' , T_c , and B' are deduced from a three-parameter fit in which all points with the given T or larger are included. Points have been weighted according to the inverse square of the ω_L error.

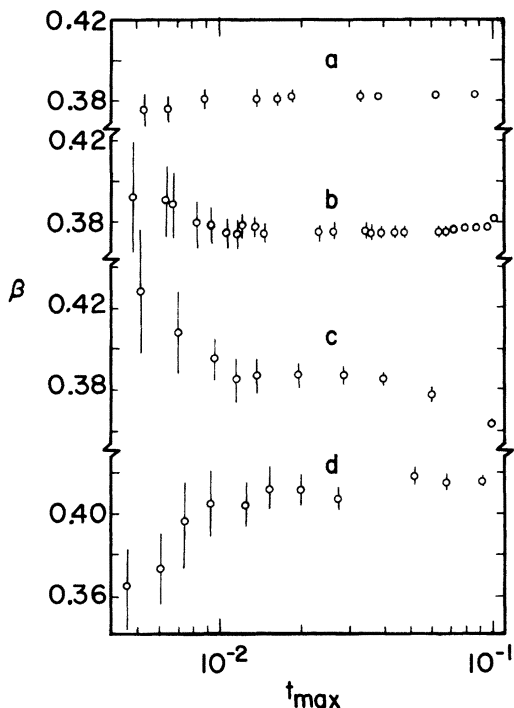


FIG. 4. Variation of the hyperfine exponent β' with the temperature range fitted. For each point the fit extends from the smallest value of t to t_{\max} . (a) $Ni^{100}Rh$; (b) $Ni^{111}Cd$; (c) $Ni^{57}Fe$; (d) $Ni^{181}Ta$.

cal region below T_c appear in Fig. 3(b), and show that $h(t)$ deviates from $\sigma_s(t)$ by 10% at $t=0.1$.

Table III and Fig. 4(b) summarize the critical-region data as reanalyzed. As in the ^{100}Rh case, the values of β' , B' , and T_c are essentially independent of the range of t , and the result for the full range agrees well with that quoted previously²² as well as the results on bulk Ni. The somewhat larger errors near T_c for $Ni^{111}Cd$ reflect the reduced sensitivity of the ^{111}Cd probe in relation to the ^{100}Rh probe.

3. $Ni^{57}Fe$

$Ni^{57}Fe$, like $Ni^{100}Rh$, involves an impurity with a local moment. The noncritical region was first studied by Dash, Dunlap, and Howard²³ and fitted with the molecular-field model of Jaccarino, Walker, and Wertheim.⁸ A later study by Benski²⁴ confirmed these results. The data of Benski are reproduced in Fig. 3(c), and show that $h(t)$ deviates from $\sigma_s(t)$ by $\sim 5\%$ at $T/T_c=0.1$. [The curve $h(t)$ shows less deviation than in the work of Dash *et al.*²³ because a number of corrections made by the latter were not applied.]

Data in the critical region were first reported by Howard, Dunlap, and Dash,²⁵ later by Gumprecht, Steiner, Crecelius, and Hufner,²⁶ and

most recently by Benski, Reno, Hohenemser, Lyons, and Abeledo.²⁷ Howard *et al.* obtained a transition from $\beta' = \frac{1}{3}$ to $\beta' = \frac{1}{2}$ at $1 - T/T_c = 9 \times 10^{-3}$, with the higher value of β' found closer to T_c . Gumprecht *et al.*²⁶ report $\beta' = 0.38 \pm 0.01$ without further comment. The data of Benski *et al.* reproduced the latter result and are used here for reanalysis because they extend closer to T_c than any other.

Table IV and Fig. 4(c) summarize the critical-region data of Benski *et al.*²⁷ as reanalyzed for various temperature ranges. The contrast to $Ni^{100}Rh$ and $Ni^{111}Cd$ is striking. As the range of fitting is successively reduced, the effective value of β' increases at first, remains constant at about the value previously quoted, and then continues to increase further, albeit with increasing statistical error. The values of B' and T_c vary correspondingly with range. Taken together, the above findings are interpreted as a failure of the power law both close to and far from T_c .

The falling off of β' for $t_{\max} > 6 \times 10^{-2}$ was previously noted²⁷ and interpreted as the limit of the asymptotic region for $Ni^{57}Fe$. The fact that the effective asymptotic region for $Ni^{57}Fe$ is smaller than for $Ni^{100}Rh$ and $Ni^{111}Cd$ is not surprising and may in fact be understood in terms of the magnitude of B' (see Sec. IIIC below).

More disturbing is the apparent increase of β' and variation of T_c and B' in the region $t_{\max} < 10^{-2}$. This effect was, in fact, missed in the previous analysis of the data over the full range of t using the Heller misfit parameter. It is comparable to the effect reported by Howard *et al.*²⁵ As seen in Fig. 5, the apparent increase in β' appears on a logarithmic plot only if T_c is determined by a three-parameter fit to nine points closest to T_c .

We attribute the apparent increase in β' not to probe disturbance but to residual temperature gradients previously thought insignificant. These produce line broadening and effectively round $h(t)$ near T_c . Observed line broadening both below²⁷ and above²⁴ T_c was far greater than in a subsequent study of critical fluctuations by Kobeissi *et al.*²⁸ in which the effect of temperature gradients on linewidth was explicitly demonstrated.

Nevertheless, we regard the value of β' as quoted by Benski *et al.*²⁷ (Table I, Ref. t) as close to correct for the following reasons. In the Kobeissi study,²⁸ T_c was determined in three ways: Through thermal scanning, through analysis of the linewidth divergence above T_c , and through the use of $\beta' = 0.378$ in a two-parameter fit (B' and T_c variable) to new data below T_c . The remarkable finding was that all three values of T_c agreed to within 0.05 K. This can only be if $\beta' = 0.378$ is close to correct for $t \leq 10^{-2}$ as well as farther from T_c .

TABLE III. Analysis of $Ni^{111}Cd$.^a

Source ^b	T_c ^c (K)	$10^3 t$ ^d	ω_L ^e (rad/sec)	β' ^f	T_c ^f (K)	B' ^f
2	568.49	100.7	57.66 (16)	0.382 (1)	632.16 (2)	1.30 (1)
1	571.73	95.5	57.74 (10)	0.378 (2)	632.13 (2)	1.27 (1)
3	577.50	86.4	53.76 (21)	0.378 (2)	632.13 (2)	1.28 (1)
3	582.16	79.0	52.31 (18)	0.378 (2)	632.13 (2)	1.28 (1)
3	586.89	71.6	50.34 (12)	0.377 (2)	632.13 (2)	1.27 (1)
1	589.56	67.3	48.78 (10)	0.376 (2)	632.13 (2)	1.27 (1)
3	592.14	63.2	47.83 (14)	0.376 (3)	632.13 (2)	1.27 (1)
3	602.40	47.0	42.95 (11)	0.376 (3)	632.12 (2)	1.26 (2)
3	604.78	43.2	41.29 (16)	0.375 (3)	632.12 (2)	1.26 (2)
3	607.56	38.8	39.83 (10)	0.375 (3)	632.12 (2)	1.26 (2)
1	609.61	35.6	38.12 (10)	0.375 (4)	632.12 (3)	1.26 (2)
3	610.60	34.0	37.50 (16)	0.376 (4)	632.12 (3)	1.26 (2)
3	615.61	26.1	34.41 (15)	0.376 (4)	632.12 (3)	1.27 (2)
3	617.67	22.8	32.42 (17)	0.375 (4)	632.12 (3)	1.26 (2)
3	622.86	14.6	27.32 (14)	0.375 (5)	632.12 (3)	1.26 (2)
2	622.98	14.4	27.28 (9)	0.375 (5)	632.12 (3)	1.26 (3)
2	623.79	13.2	26.43 (12)	0.378 (5)	632.13 (3)	1.28 (3)
2	624.51	12.0	25.62 (9)	0.379 (6)	632.13 (3)	1.28 (3)
2	624.75	11.7	25.06 (12)	0.375 (7)	632.12 (4)	1.26 (4)
2	625.44	10.6	24.23 (12)	0.375 (7)	632.12 (4)	1.26 (4)
2	626.26	9.27	23.16 (10)	0.379 (9)	632.13 (4)	1.28 (5)
2	626.94	8.19	22.04 (13)	0.380 (12)	632.14 (5)	1.29 (7)
2	627.85	6.76	20.49 (13)	0.389 (16)	632.16 (6)	1.35 (10)
2	628.08	6.39	20.23 (11)	0.391 (17)	632.16 (6)	1.36 (12)
2	629.07	4.82	18.24 (13)	0.393 (26)	632.17 (8)	1.38 (18)
2	629.44	4.24	17.24 (11)	0.384 (30)	632.15 (9)	1.32 (20)
2	630.02	3.32	15.46 (15)	0.376 (47)	632.14 (11)	1.26 (32)
2	630.48	2.59	14.46 (11)	0.447 (64)	632.26 (14)	1.88 (64)
2	631.02	1.74	12.48 (19)			
2	631.21	1.44	11.12 (21)			
2	631.50	0.98	9.94 (17)			
2	631.81	0.49	7.83 (33)			
2	632.01	0.18	5.80 (70)			

^a A preliminary report on these data was given in Ref. 22.

^b Source 2 data were taken in zero applied field. Source 1 and 3 data were taken with applied fields of 14–50 G, in a temperature region where applied fields of this magnitude do not disturb the observed frequency.

^c For each source, temperatures were reproducible to 0.03 K; the measurement sensitivity of the thermocouples was 0.01 K. From one source to the next the absolute temperature scale differed by 1–2 K near T_c , presumably because of differences in thermocouple calibration and physical clamping of the sample. Thus for source 1, 2, and 3, T_c was determined to be 633.55 (20), 632.12 (03), and 629.42 (12) K, respectively, using separate power-law fits for each run. In order to establish the single temperature scale given here, source 1 and 3 temperatures were renormalized to source 2 by the addition of –1.43 and +2.70 K, respectively, to the measured temperatures.

^d The reduced temperature t reported in this column is calculated on the basis of $T_c = 632.12$ K, the value obtained by fitting the entire range of data.

^e Errors in ω_L indicate statistical uncertainty of fitting PAC spectra only. In all cases, the effects of combining three runs, particularly uncertainty in T_c renormalization and time calibration, produced an error in ω_L of about 1%. For fitting the combined data to power laws, the latter uncertainty was employed. For calculating h , $\omega_L(0) = 106.3 \times 10^6$ rad/sec was used. This differs somewhat from the result of Rosenblum (Ref. 19), who obtained $\omega_L(0) = 104.4 \times 10^6$ rad/sec.

^f Values of β' , T_c , and B' are deduced by fitting all data with the indicated reduced temperature or smaller.

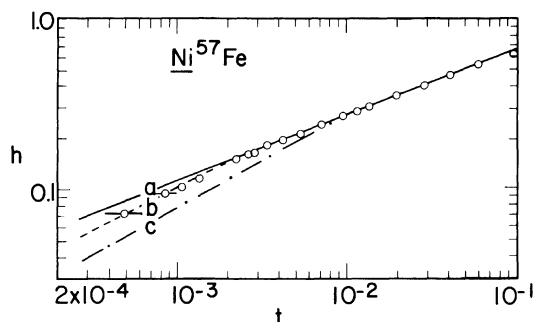


FIG. 5. Double power law in $Ni^{57}Fe$. (a) Data of Benski *et al.* as previously published (Ref. 27); (b) Ref. 27 data with T_c from fit to the nine points closest to T_c ; (c) data of Howard *et al.* (Ref. 25).

4. $Ni^{181}Ta$

Like $Ni^{111}Cd$, $Ni^{181}Ta$ represents a case of a nonmagnetic probe. Some perturbed-angular-correlation experiments have led to anomalous results involving several frequencies and much reduced anisotropies.^{29,30} Simple diffusion has not worked, and hence ion implantation has been used.^{31,32} Oddou, Berthier, and Perretto³³ have succeeded in making well annealed samples by drawing single-crystal sources from a melt. The temperature dependence of h obtained from these sources is shown³⁴ in Fig. 3(d) and indicates that $h(t)$ deviates from $\sigma_s(t)$ by 20% at $t=10^{-1}$.

Results for the critical region have been reported by Oddou *et al.*³⁵ for two samples with quoted impurity content of 3 ppm and 30 ppm. The derived β' value is 0.417 ± 0.010 for both concentrations, i.e., significantly different than other data on Ni. In addition, Oddou, Moulin, and Perretto³⁶ have also reported bulk measurements using the same samples as used in the hyperfine experiments. By scaling equation of state analysis, they obtain $\beta = 0.380 \pm 0.005$, with the exponent γ fixed at 1.32. This is in agreement with most other hyperfine and bulk results on Ni, and it is for this reason that Oddou *et al.* argue that the deviant hyperfine results are evidence for probe-disturbance effects.

Oddou has kindly sent us detailed tables of $H_{hf}(T)$ for both the 3- and 30-ppm sources.³⁴ For the 30-ppm source, we conclude that T_c is undefined and undefinable. An illustration of this appears in Fig. 6. Evidently something has destroyed the sharpness of the transition.

Table V and Fig. 4(d) summarize the 3-ppm data, as reanalyzed for various temperature ranges. Though there is some scatter in the data it is clear that within statistical error β' , B' , and T_c are independent of the range of t , and that the

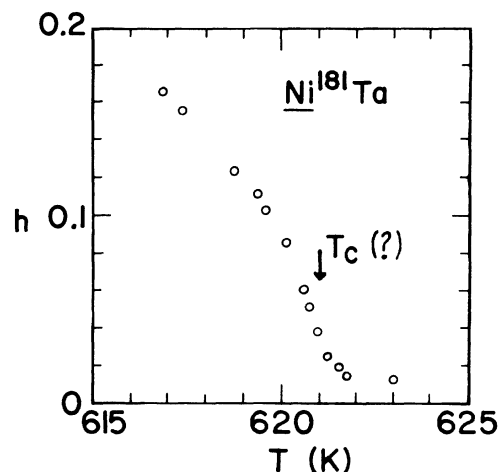


FIG. 6. $h(T)$ near T_c for 30-ppm $Ni^{181}Ta$ data of Oddou *et al.* (Ref. 35).

value of β' previously reported is essentially correct. It remains to be explained why the exponent is different than in the other three cases. Two recent developments bear on this question.

(i) Oddou has recently reported that the originally estimated impurity concentration for his sources was in error, and that the "3-ppm" source in fact contained $\sim 0.3\%$ nonmagnetic impurities.³⁷ This finding is also consistent with the shift $\Delta T_c \approx 7$ K toward lower temperature found in the source.

(ii) Suter of our laboratory has shown that, for $Ni_{99.5}Cu_{0.5}$ with dilute ^{111}Cd impurities, perturbed-angular-correlation measurements yield $\Delta T_c \approx 10$ K and $\beta' = 0.42$ in agreement with Oddou's finding. An account of this work will be published shortly.³⁸

We must conclude, therefore, that the results for $Ni^{181}Ta$ given in Table I does not affect our hypothesis, since the samples do not contain isolated impurities. On the other hand, one desires an explanation for the occurrence of an anomalous value of β' when the impurity concentration is $\sim 1\%$.

Three possible explanations may be envisioned: (a) the effect arises as a fitting artifact for data that are strongly inhomogeneous; (b) the anomalous exponent arises from a probe-disturbance effect; and (c) the anomalous exponent arises from an impurity-induced change in σ_s , which in turn affects h . The first possibility is not very interesting and would merely serve to illustrate the limitation of the hyperfine method. The second is possible, though unlikely. The third has been suggested on theoretical grounds³⁹ and would be highly interesting. To deal with either proposition (b) or (c), proposition (a) must first be considered. This has not been done; hence no choice between the three possibilities is presently possible.

TABLE IV. Analysis of $Ni^{57}Fe$.^a

T ^b (K)	$10^3 t$ ^c	h_{ht}	β' ^d (T_c free)	T_c ^d (K)	B' ^d
573.17	98.7	0.630 (2)	0.363 (4)	635.87 (4)	1.48 (2)
597.69	60.1	0.538 (2)	0.377 (3)	635.91 (3)	1.56 (2)
610.27	40.4	0.469 (2)	0.385 (3)	635.94 (2)	1.62 (2)
617.83	28.5	0.411 (2)	0.387 (4)	635.95 (2)	1.63 (3)
623.54	19.5	0.355 (2)	0.387 (6)	635.95 (3)	1.63 (4)
627.32	13.6	0.309 (2)	0.387 (8)	635.95 (3)	1.63 (6)
628.59	11.6	0.288 (2)	0.385 (10)	635.94 (3)	1.61 (7)
629.84	9.60	0.271 (2)	0.395 (11)	635.96 (4)	1.70 (9)
631.46	7.06	0.240 (5)	0.408 (20)	635.98 (5)	1.82 (19)
632.64	5.21	0.214 (5)	0.429 (30)	636.02 (6)	2.04 (32)
633.35	4.08	0.196 (5)	0.460 (43)	636.07 (9)	2.43 (56)
633.86	3.29	0.183 (5)	0.496 (68)	636.12 (13)	2.97 (108)
634.23	2.71	0.161 (5)			
634.38	2.47	0.161 (5)			
634.63	2.07	0.151 (5)			
635.20	1.18	0.115 (5)			
635.39	0.91	0.102 (5)			
635.53	0.66	0.096 (5)			
635.76	0.30	0.073 (5)			

^a These data were originally reported in Ref. 27.

^b Temperatures listed here were **reproducible** to 0.05 K; the relative sensitivity of the thermocouples was 0.02 K. The absolute temperature scale is believed to be accurate to 2 K.

^c The reduced temperature t reported in this column is calculated on the basis of $T_c = 635.95$ K.

^d Values of β' , T_c , and B' are deduced from a three-parameter fit in which all points with the given T or larger are included. Points have been weighted according to the inverse square of the field error.

C. Range of power-law behavior

Examination of Figs. 4(a) and 4(b) indicates that the range of t over which power-law behavior is observed is considerably larger than the region free of probe disturbance in the Ising-model calculation discussed in Sec. II. The latter would lead one to expect observable deviations for $t \geq 10^{-2}$.

We may explain this observation if we recognize that deviation from power-law behavior depends on more than probe-disturbance effects. To characterize the deviation of h from power-law behavior, consider the quantity $h/B't^{\beta'}$. It will be unity as long as h follows Eq. (2) and will deviate from unity as h deviates from Eq. (2). Since in our model $h = \sigma_1$, we may write the identity (with $\beta = \beta'$)

$$h/B't^{\beta} = (\sigma_s/Bt^{\beta})(\sigma_1/\sigma_s)(B/B'). \quad (5)$$

Thus, the deviation of $h/B't^{\beta'}$ from unity depends on a product of three terms, as follows: (i) σ_s/Bt^{β} , describing the deviation of the bulk magnetization from power-law behavior; (ii) σ_1/σ_s , describing the probe disturbance; and (iii) B/B' , a constant. It is therefore, in principle, possible that bulk deviation and probe-disturbance effects

approximately cancel and yield a fortuitously large region of power-law behavior for h . It is also possible that bulk deviation and probe-disturbance effects reinforce each other, and in this way result in an unusually small region of power-law behavior for h . For $Ni^{111}Cd$ and $Ni^{100}Rh$, we believe the former to be true, for $Ni^{57}Fe$ the latter. In Fig. 7, the effect is illustrated for the Ising model discussed in Sec. II. Figure 7 also makes plausible why beyond $t = 3 \times 10^{-2}$ a slight increase in the effective value of β' is seen for the cases of $Ni^{100}Rh$ and $Ni^{111}Cd$: It is in this region that $h/B't^{\beta'}$ in the Ising model experiences a slight maximum, before decreasing precipitously.

Finally, we make the empirical observation that the region of qualitatively good power-law fit is related to the magnitude of the coefficient B or B' . The closer B and B' are to unity, the larger will be the region of good fit. An illustration for the cases we have discussed appears in Fig. 8.

D. Best value of β for Ni-based impurity-probe systems

Given our observations about the four systems we have discussed, including the size of the asymptotic region in each case, we arrive at the

TABLE V. Analysis of $Ni^{181}Ta$, 3 ppm. ^a

T^b (K)	$10^3 t^c$	H_{hf} (kG)	β'^d	T_c^d (K)	B'^d
553.20	116.0	52.30 (15)	0.413 (2)	625.82 (3)	1.33 (1)
568.20	92.0	47.89 (15)	0.415 (2)	625.84 (3)	1.34 (1)
583.25	68.0	42.33 (15)	0.415 (3)	625.84 (3)	1.34 (1)
583.30	67.9	42.01 (15)	0.414 (3)	625.83 (3)	1.33 (1)
593.25	52.0	38.08 (15)	0.418 (4)	625.85 (4)	1.35 (2)
608.65	27.4	28.75 (13)	0.410 (6)	625.83 (4)	1.31 (3)
613.35	19.9	25.34 (13)	0.417 (8)	625.86 (5)	1.35 (5)
616.20	15.3	22.93 (15)	0.422 (12)	625.88 (6)	1.38 (7)
618.00	12.5	20.85 (13)	0.416 (15)	625.86 (7)	1.34 (9)
620.05	9.19	18.51 (15)	0.427 (24)	625.90 (10)	1.41 (16)
621.15	7.43	16.90 (15)	0.424 (31)	625.88 (12)	1.39 (20)
621.15	7.43	17.10 (21)	0.423 (39)	625.88 (14)	1.39 (26)
622.00	6.07	15.43 (23)	0.383 (40)	625.78 (12)	1.13 (23)
622.92	4.60	13.55 (19)	0.359 (50)	625.73 (13)	0.98 (26)
623.88	3.07	11.89 (23)	0.448 (143)	625.90 (32)	1.62 (125)
624.48	2.11	10.34 (19)			
624.82	1.57	8.77 (24)			
625.18	0.99	7.70 (19)			
625.32	0.76	6.56 (23)			
625.44	0.57	6.45 (40)			

^a An analysis of these data has been previously published in Ref. 35. The numerical data presented here for H_{hf} and T were supplied by J. L. Oddou (unpublished).

^b Temperature uncertainties average ± 0.12 K.

^c Reduced temperatures given are calculated on the basis of $T_c = 625.80$ K.

^d Values of β' , T_c , and B' are deduced from a three-parameter fit in which all points with the given T or larger are included. Points have been weighted according to the inverse square of the field error.

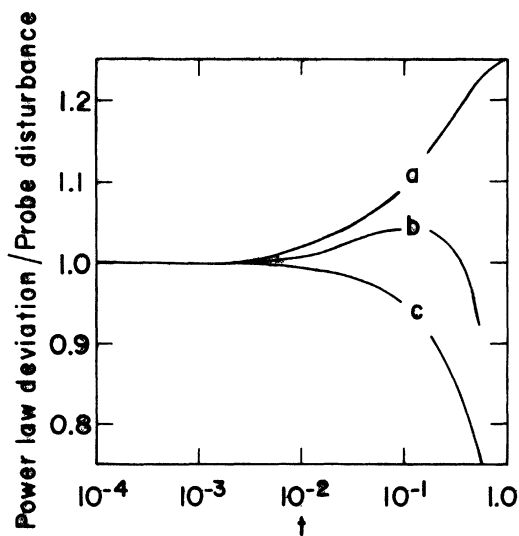


FIG. 7. Functions describing power-law deviation for the hyperfine exponent β' in the Ising model discussed in the text. (a) $(B/B')(\sigma_1/\sigma_s)$; (b) $h/B' t^{\beta'}$; (c) $\sigma_s/B t^{\beta'}$.

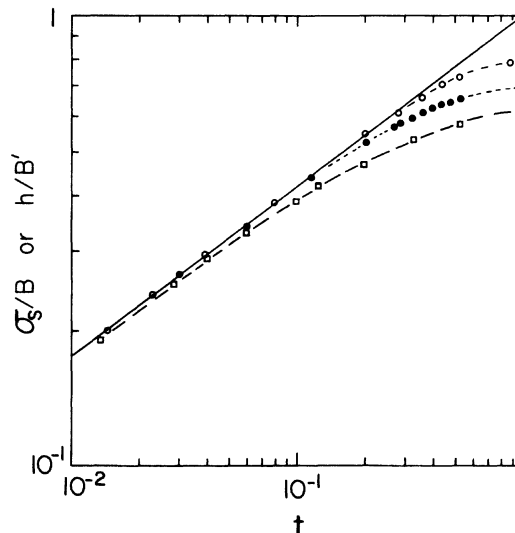


FIG. 8. Relation of range of power law to coefficient B or B' . Open circles, $Ni^{111}Cd$ ($B' = 1.26$); solid circles, pure Ni ($B = 1.42$); squares, $Ni^{57}Fe$ ($B' = 1.63$). Solid line is $t^{0.378}$.

TABLE VI. Best values of critical parameters for Ni-based systems.

Material	β'	T_c	B'	t
$Ni^{100}Rh$	0.382 (3)	630.91 (1)	1.26 (2)	$2 \times 10^{-4} < t < 2 \times 10^{-2}$
$Ni^{114}Cd$	0.375 (4)	630.12 (3)	1.26 (3)	$2 \times 10^{-4} < t < 2 \times 10^{-2}$
$Ni^{57}Fe$	0.387 (4)	635.95 (3)	1.63 (4)	$3 \times 10^{-4} < t < 2 \times 10^{-2}$

results given in Table VI. The $Ni^{57}Fe$ β' , even though taken from the "plateau" region of Fig. 4(c), is undoubtedly somewhat disturbed by the temperature inhomogeneity we have discussed and can be expected to decrease somewhat in a more careful experiment. The $Ni^{181}Ta$ value is not included because it is unlikely to be a case in which the probe atoms are free of impurity-impurity interactions. We can see that, with the stated reservations, Table VI confirms our central hypothesis. Probe-disturbance effects for $t \leq 10^{-2}$ are small. Specifically, Table VI leaves the possibility of $\sim 2\%$ probe-disturbance effect but is consistent with no effect.

IV. SUMMARY AND CONCLUSIONS

Probe disturbance of the hyperfine critical exponent β has been discussed for various magnetically ordered systems, both from a theoretical and an experimental point of view.

From a theoretical point of view, the question whether Eq. (3) is correct in the critical region has been investigated, and if so, for what range of reduced temperature t . For molecular-field models of an isolated nonmagnetic impurity, systematic errors as high as $\sim 5\%$ in β are possible. For a three-dimensional Ising model the impurity-induced disturbance is substantially less and for $10^{-4} \leq t \leq 10^{-2}$ produces a systematic error in β of less than 1% . This surprising and pleasant result suggests that hyperfine measurements using isolated impurity probes are essentially free of systematic error.

From an experimental point of view, we have examined all systems for which, to our knowledge, two or more measurements have been made on the same substance. The results appear in Table I, as reported by their authors. They look generally

encouraging and support the hypothesis that probe-disturbance effects, if any, are small. To investigate the variation of the effective β with temperature range of fitting, we have reanalyzed all available hyperfine data on Ni for which impurity probes have been introduced by diffusion or melting. Despite the problematic nature of the results on $Ni^{57}Fe$ and $Ni^{181}Ta$, we find no evidence that the critical behavior of impurity hyperfine fields at isolated impurities is disturbed by anything other than instrumental problems when $t \leq 2 \times 10^{-2}$. If anything, the asymptotic region is in some cases markedly expanded because of the partial cancellation of bulk-deviation and probe-disturbance effects.

Both on theoretical and experimental grounds we conclude, therefore, that for $t \leq 10^{-2}$ probe-disturbance effects are no more than 2% in β and probably are considerably less. At the same time, it is now clear that great care must be taken in interpreting experimental results. Without analysis over various temperature ranges, it is possible to obscure crossover due to inhomogeneities, temperature gradients, and for that matter, changes in σ_s brought on by the presence of appreciable impurity concentration.

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