

$= 0) = 3200$ G and $T_c = 5.33^\circ\text{K}$. The sample resistance ratio between 300 and 10°K is about 80, which corresponds to a three-to-one ratio of the electron mean free path to the coherence length.⁵

We have studied the temperature dependence of the absorption spectrum ranging from $0.788T_c$ to $0.131T_c$ at several applied fields. In Fig. 1 we show a typical series of data for temperature ranging from $0.6T_c$ down to $0.187T_c$ at $H_e = 1072$ G. As the temperature is lowered, the linewidth increases, but the triangular lattice structure is apparently preserved. We have also studied the sample at applied fields from 2410 down to 536 G and find the same general temperature dependence as that shown in Fig. 1.

In Fig. 2 we plot h_v , h_s , and B for temperatures ranging from $0.7T_c$ down to $0.1T_c$ at $H_e = 804$ and 1608 G. Both of these sets of curves have the same general behavior, in that B , as mentioned before, is almost equal to H_e , while h_s decreases as the temperature is lowered and becomes less sensitive to temperature changes at lower temperatures. These two fields, h_s and B , have almost the same temperature dependence because the spins are predominantly located in and around the saddle point h_s . The decrease of B below the transition temperature is due to the diamagnetic nature of the superconducting state.

The field h_v was found to be linear in temperature (see Fig. 2) down to the lowest temperature attainable by the apparatus ($\sim 0.1T_c$) for all applied fields we have studied ($H_e > 536$ G). Since

the flux is quantized, at low temperatures, this linear behavior can only be due to the change in the local field distribution near the vortex core, while h_s stays constant. For temperatures below $0.1T_c$, it is possible that h_v departs from linearity. However, that is unlikely because our observations have covered more than 80% of the superconducting temperature range.

In conclusion, we summarize our findings. For $H_e < H_{c2}$, $\partial h_v / \partial T$ is always less than zero and is a constant for a given applied field. The extrapolated value at $H_e = 0$ is -355 G/ $^\circ\text{K}$. To obtain the value of h_v of an isolated vortex ($B = 0$) at $T = 0$, we use extrapolation and find h_v to be 1780 ± 240 G.

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PERTURBED ANGULAR CORRELATION MEASUREMENT ON ¹⁰⁰Rh IN A Ni HOST: CRITICAL EXPONENT β FOR Ni \uparrow *

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We have measured the hyperfine field at ¹⁰⁰Rh impurity nuclei in a ferromagnetic Ni host in the region just below the Curie temperature, using time-differential perturbed $\gamma\gamma$ angular correlations. We obtain a single-valued critical exponent of $\beta = 0.385 \pm 0.005$.

For comparison with theory¹ the object of most experiments on static magnetic critical phenomena has been to determine the exponents β , γ , and δ in the relations

$$\lim_{H \rightarrow 0, T \rightarrow T_c^-} M(T, H) \propto (1 - T/T_c)^\beta, \quad T < T_c; \quad (1)$$

$$\lim_{H \rightarrow 0, T \rightarrow T_c^+} \chi(T, H) \propto (T/T_c - 1)^\gamma, \quad T > T_c; \quad (2)$$

$$H \propto M^\delta, \quad T = T_c. \quad (3)$$

Here, M is the magnetization, χ the magnetic susceptibility, H the magnetic field in the sample, T the absolute, and T_c the critical temperature. Experiments through 1967 have been reviewed by Heller.² Recently, the above relations have been combined into a single scaling equation of state involving β , γ , and δ as parameters.

In this note, we report new experiments on β for Ni. The first measurements of $M(T, H)$ near

Table I. Determination of critical indices for Ni.

β	γ	δ	H_{ext} range kG	Ref. - date	Comments (SES = scaling equation of state)
	1.35 ± .02		0.05 - 18	5 (1964)	Reanalysis of Ref. 4 bulk data.
	1.29 ± .03		0.04 - 0.36	8 (1965)	bulk data.
1/2 ^a			0	9 (1965)	Hf field in ⁵⁷ Fe/Ni by Mössbauer Effect.
1/3 ^b					
0.41 ± .04	1.30 ± .05	4.22 ± .03	0.05 - 18	6 (1967)	SES reanalysis of Ref. 4 data.
0.3865	1.31 ± .01	4.39 ± .03	0.05 - 18	7 (1967)	SES analysis of Ref. 4 & new data.
0.378 ± 0.004	1.34 ± .01	4.58 ± .05	0.5 - 25	10 (1968)	SES analysis of new bulk data
0.50 ± 0.02 ^c			0	13 (1968)	neutron depolarization
0.375 ± 0.013	1.31	4.48 ± .14	0.05 - 18	3 (1970)	SES reanalysis of Ref. 4 bulk data.
0.373 ± 0.016	1.28	4.44 ± .8	0.5 - 25	3 (1970)	SES reanalysis of Ref. 10 data.
0.50 ± 0.01 ^a	1.31 ± .01	4.17 ± .05	0.008 - 1.1	11 (1970)	bulk data; β obtained by kink point method.
0.34 ± 0.01 ^b					
0.398 ± 0.01				12 (1970)	bulk data; kink point method.
0.385 ± 0.005			0 - 0.015	this work	Hf field in ¹⁰⁰ Rh/Ni by TDPAC

^aFor $(1-T/T_c) < 9 \times 10^{-3}$.^bFor $(1-T/T_c) > 9 \times 10^{-3}$.^cFor $(1-T/T_c) < 3 \times 10^{-3}$.

T_c were done in 1926 by Weiss and Forrer.³ These data have been extensively reanalyzed to extract critical indices.⁴⁻⁷ New measurements have been made by others,⁸⁻¹³ all but two of these being bulk measurements. A summary of critical index determinations for Ni appears in Table I.

Though fitting by a scaling equation of state yields an analytical form that is of considerable theoretical interest, the values of β and γ derived are not directly obtained in the limits indicated above, but instead involve considerable applied fields (see Table I), as well as experimental problems discussed by Heller.² Thus, β and γ may eventually approach values at variance with the scaling equation analysis.

The possibility was first experimentally suggested by the Mössbauer data of Howard, Dunlap, and Dash,⁹ who found in zero field that $\beta = \frac{1}{2}$ for $(1-T/T_c) < 9 \times 10^{-3}$, and $\beta = \frac{1}{3}$ above that. Unfortunately these data are inherently ambiguous in the region of greatest interest due to unresolved spectra.

More recently, a new technique known as the "kink point" method¹⁴ has allowed an indirect determination of β without reference to a scaling equation of state, and hence the other exponents. With this method, Arajcs *et al.*¹¹ obtain results

nearly identical with the Mössbauer data; yet Miyatani and Yoshikawa,¹² using the same technique, observe a single value of β . The reasons for this discrepancy are unclear. Finally, Bakker, Rekveldt, and van Loef¹³ have used neutron depolarization to measure the magnetization of Ni, with the result that $\beta = 0.50$ for $(1-T/T_c) < 3 \times 10^{-3}$ and "no unique fit to a power law" above this.

In the work on Ni reported here we introduce time-differential perturbed angular correlations (TDPAC) as a tool in the study of magnetic critical phenomena. This technique, like the Mössbauer effect and nuclear magnetic resonance, probes the hyperfine interaction of individual nuclei, and thus avoids a principal difficulty of bulk measurements. In undertaking our work we hoped to avoid as well the major disadvantage of the Mössbauer data,⁹ i.e., their lack of resolution near T_c .

We used the 84-75 keV $\gamma\gamma$ cascade of ¹⁰⁰Rh, reached via the 4-day ¹⁰⁰Pd parent. Carrier-free ¹⁰⁰Pd was plated onto 99.998% pure Ni foil, and then diffused at 1150°C. We determined the effective hyperfine field at the ¹⁰⁰Rh nuclei by observing the Larmor precession of the angular correlation.

General aspects of the TDPAC technique have been discussed in several reviews.^{15,16} In our work, we particularly depend on optimum delayed coincidence time resolution for NaI(Tl) scintillation counters, a problem which has been discussed by us in detail elsewhere.^{17,18}

Our choice of ¹⁰⁰Rh was indicated by two facts: (1) The 84-75 keV cascade, because of a long intermediate state lifetime, large intermediate state moment, and large anisotropy ($T_{1/2} = 235$ nsec, $g = 2.15$, $A_2 = 0.174$),^{19,20} is one of the most sensitive TDPAC probes known, Alonso and Grodzins²¹ having detected 180 G, or about 100 times smaller than the resolution limit of ⁵⁷Fe Mössbauer experiments. (2) The ¹⁰⁰Rh/Ni impurity host system had been previously studied by the Berkeley group both below²² and above²³ T_c , and had been shown to have a large hyperfine field ($H_{eff} \approx 200$ kG at $T = 295^\circ\text{K}$).

Two types of experiments were performed, as follows: (1) The source foil was polarized along its plane and perpendicular to the plane of the counters, as described previously.¹⁸ With the counters at an angle of $5\pi/4$, the ratio

$$R(t) = [C_{\uparrow}(t) - C_{\downarrow}(t)] / [C_{\uparrow}(t) + C_{\downarrow}(t) - 2B] \quad (4)$$

was measured, where \uparrow and \downarrow denote source foil magnetization, $C(t)$ denotes the time dependent 84-75 keV coincidence rate, and B the time-independent background. (2) With zero applied fields and the counters at an angle π , the rate $C(t)$ was measured.

Assuming only static hyperfine interactions and neglecting the anisotropy coefficient A_4 , we expect for the above two cases, respectively,²⁴

$$R(t) = [3A_2 / (4 + A_2)] \sin 2\omega_L t, \quad (5)$$

$$C(t) - B \propto \exp(-t/\tau) [1 - (A_2/5)(2 \cos 2\omega_L t + 2 \cos \omega_L t + 1)], \quad (6)$$

where $\omega_L = \mu H_{eff} / \hbar$ is the Larmor frequency of the excited-state moment in the effective hyperfine field, and τ is the intermediate state lifetime. The frequency ω_L was obtained in both cases by Fourier analysis of the data.

Temperature control of the sample was achieved with a thermocouple-controlled dc feedback system having a stability of $\pm 0.02^\circ\text{C}$ over 24 h. Temperature gradients between the sample and measuring thermocouple were less than 0.25°C , and absolute accuracy was $\pm 2^\circ\text{C}$.

For $(1 - T/T_c) > 10^{-2}$ we find that the TDPAC precession shows no loss of coherence in time, or equivalently, broadening of the Fourier transform. At room temperature, using Pb loaded

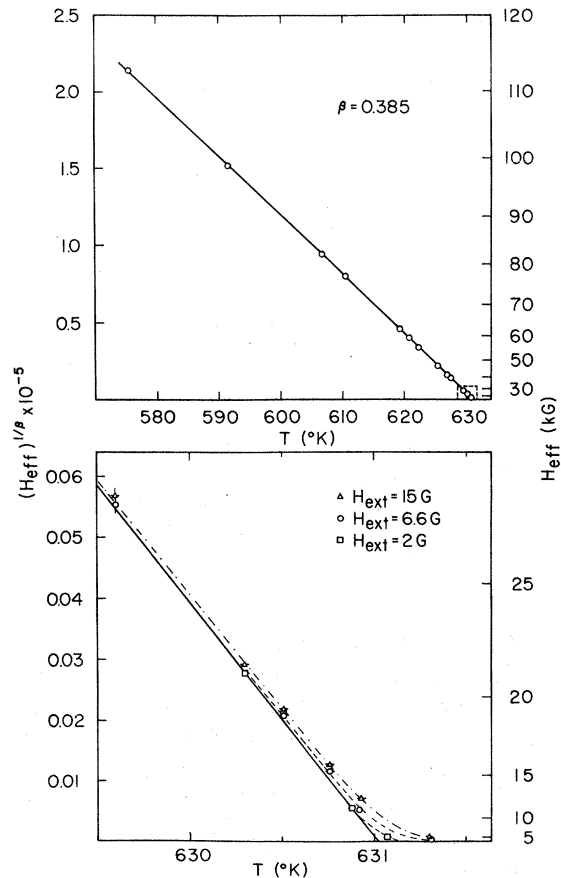


FIG. 1. Plot of $H_{eff}^{1/0.385}$ vs T . Data from two sources have been combined by normalizing the temperature scale to the same Curie point. The upper graph contains data with 2 and 0 G applied fields only, and shows no deviations from a straight line. The lower graph shows, on an expanded scale, the effect of various external fields near T_c . The solid line in both graphs is a least squares fit to the 2 and 0 G applied field data.

plastic scintillators, a Fourier transform with $\Delta\omega/\omega \approx 1.5 \times 10^{-2}$ was observed, this being the natural width for the range of Fourier analysis chosen.²⁵

As T_c is approached, a loss of coherence sets in until at $(1 - T/T_c) \approx 10^{-4}$ the effective relaxation time becomes shorter than the precession period. Since such behavior is expected because of slowing field fluctuations near T_c we might attribute our observations to this cause. Yet a distribution of temperature of order 0.08°K over the source can completely explain the observed loss of coherence near T_c . No unique interpretation is therefore possible, though this does not

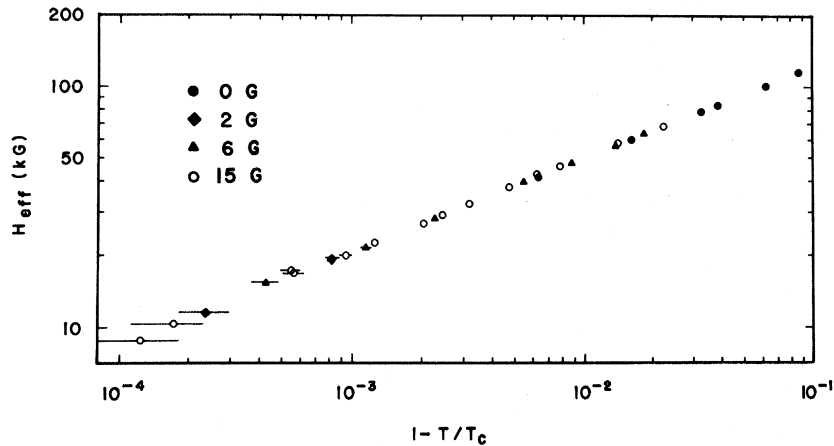


FIG. 2. Combination of all data from the three sources into a log-log plot. The evidence for a single power law over the entire range is clearly discernible. Data taken at 15 and 6.6 G applied fields have been corrected in the region of $(1-T/T_c) < 10^{-3}$ because here the effects of applied fields are substantial (see Fig. 1). The error bars reflect the uncertainty in T_c .

prevent measurement of fields down to 3 kG.

Measurements were made on three sources with external polarizing fields of 15, 6.6, 2.0, and 0 G. Since the presence of fields as low as 6.6 G caused visible departure from a single power law for values of $(1-T/T_c) < 10^{-3}$, only the 0 and 2 G applied field data were used to determine β .

To derive the value of β we used two complementary methods as follows:

(1) For each of the three sources studied, a plot of $H_{\text{eff}}^{1/\beta}$ vs T was made for various values of β . The correct exponent was selected by noting which value of β produces a straight line. This method has the advantage of not requiring prior knowledge of T_c . If T_c is defined as the T intercept of the best straight line through the data for the optimum choice of β , the three sources yield T_c values of 631.02, 630.91, and 630.30°K, with an error of $\pm 0.04^\circ\text{K}$ in each case. The observed variations in T_c may be due to possible real differences in the three Ni samples, or alternatively they may reflect various instrumental problems. In Fig. 1, results from two sources are shown for 2 and 0 G applied fields.

(2) Given the values of T_c defined above, a plot of $\ln H_{\text{eff}}$ vs $\ln(1-T/T_c)$ was made. This serves to expand the temperature region near T_c and can also be used to determine β . Figure 2 shows the combination of all data on a plot of this type.

It can be seen at once that the change in exponent form $\frac{1}{3}$ to $\frac{1}{2}$ at $(1-T/T_c) = 9 \times 10^{-3}$, which is

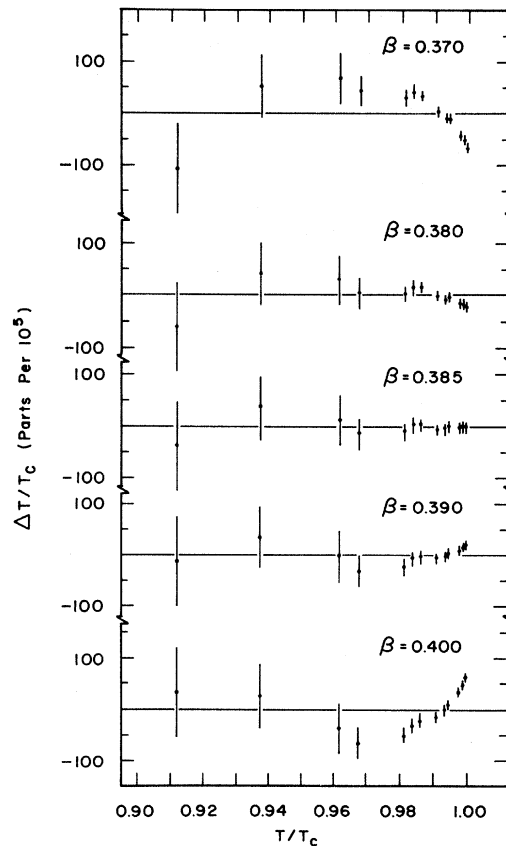


FIG. 3. In graphs of $H_{\text{eff}}^{1/\beta}$ vs T , straight line fits were made for various values of β . The above shows the fractional temperature deviation, $\Delta T/T$, plotted against T for five choices of β . It is seen from this that one may definitely distinguish $\beta = 0.385$ as a better fit than either $\beta = 0.380$ or $\beta = 0.390$. (The error analysis shown here is similar to that given in Ref. 2.)

reported by Howard, Dunlap, and Dash⁹ and Arajs *et al.*,¹¹ does not appear. It is also clear from the lower part of Fig. 1 that the disturbance of straight-line behavior due to a 15 G external field is noticeable for $(1-T/T_c) < 10^{-3}$, but that as the external field goes from 15 to 2 G, the data approach the zero field line as expected.

Based on the error analysis shown in Fig. 3, it is concluded that

$$\beta = 0.385 \pm 0.005, \quad 10^{-1} > (1-T/T_c) > 10^{-4}. \quad (7)$$

The above result bears comparison to the bulk data if $H_{\text{eff}} \propto M$. We have investigated $H_{\text{eff}}(T)$ down to 300°K, and have found that if H_{eff} and M are normalized there, the deviation of H_{eff} from M above 300°K is nowhere greater than 7.5%. Insofar as this behavior is attributable to a local moment on the Rh atom,²⁶ it may be fitted by a suitable Brillouin function,²³ which has the property of scaling as the bulk magnetization as T approaches T_c .

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