Critical Fluctuations in Ni above the Curie Point*

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Spin-correlation times in Ni are derived independently from perturbed angular correlation and neutron scattering experiments just above the Curie point. A comparison of these times shows that the usual form of the van Hove function does not apply for reduced temperatures $\epsilon < 10^{-2}$.

Critical fluctuations in Ni as observed by neutron scattering[1] have been a puzzle for some time. For reduced temperatures $10^{-2} < \epsilon < 10^{-1}$ above the Curie point, T_c, the correlation length, spin diffusion constant, and static susceptibility appear to violate static and dynamic scaling. Perturbed angular correlation (PAC) experiments[2] and several studies of transport coefficients[3] also depart from expected behavior. Here we report an extension of previous PAC experiments on very dilute (<0.01 at.%) *Ni*Rh in the region $10^{-4} < \epsilon < 10^{-2}$ and, in terms of spin correlation times, obtain a quantitative comparison to the neutron data. We conclude that the form of the van Hove function used to fit the neutron data [see Eqs. (14) and (15)] does not apply for $\epsilon < 10^{-2}$, and may be incorrect for $\epsilon > 10^{-2}$.

We first ask, how reliable is a hyperfine study of dilute *Ni*Rh as a probe of Ni critical behavior? The ions Rh^{3+} and Ni^{2+} are electronically similar and both have radii of 0.70 Å. Just below T_c the average hyperfine field $\langle \vec{H} \rangle$ at Rh in Ni scales precisely like the bulk magnetization.[4] Hyperfine-field systematics[5] indicate that the atomic moment of Rh in ferromagnetic Ni is very close to that of Ni itself (i.e., $0.6\mu_B$). *Ni*Rh shows no incoherent magnetic neutron scattering, indicating that neutrons do not "distinguish" the magnetic structure of the Rh impurities from the Ni host.[6] Thus, short of Ni itself (for which no useful hyperfine probe exists) the choice of Rh is ideal.

With diffused sources as described earlier[2] we observed time-differential perturbations of the 75–84-keV $\gamma\gamma$ cascade of ^{100}Rh in zero external field. We formed the ratio

$$R(t) = [C(\pi, t) - C(\pi/2, t)]/[C(\pi, t) + 2C(\pi/2, t) - 3B]. \tag{1}$$

In the notation of Steffen and Frauenfelder[7] the coincidence rate is

$$C(\theta, t) = C_0 \exp(-t/\tau_N)[1 + \gamma_a A_{22} G_2(t) P_2(\cos\theta)] + B, \tag{2}$$

where γ_a describes the finite solid-angle attenuation. Putting Eq. (2) into (1), and using[8] $A_{22} = 0.173 \pm 0.004$ and $\gamma_a = 0.80 \pm 0.02$, we have

$$R(t) = (0.069 \pm 0.002) G_2(t). \tag{3}$$

The function $R(t)$ thus affords a direct measure of $G_2(t)$. Gabriel[9] has discussed the effective Hamiltonian

$$\mathcal{H}(t) = \gamma \vec{I} \cdot \vec{H}(t) = \gamma I \cdot [\langle \vec{H} \rangle + h(t)], \tag{4}$$

where $\langle H \rangle$ and $h(t)$ are the average and time-varying hyperfine fields, respectively. With Gabriel's Eq. (61) we obtain above T_c that

$$G_2(t) = 0.20[\exp(-\lambda_{20} t) + 2\exp(-\lambda_{21} t) + 2\exp(-\lambda_{22} t)]. \tag{5}$$

If in addition the fluctuations are isotropic, we have

$$G_2(t) = \exp(-t/\tau_2), \qquad 1/\tau_2 \equiv \lambda_{20} = \lambda_{21} = \lambda_{22}. \tag{6}$$

As a test of isotropy, we have made careful measurements of $R(t)$ at $\epsilon = 5.9 \times 10^{-4}$, with results as shown in Fig. 1. From this we conclude that Eq. (6) is satisfied. As in all measurements re-

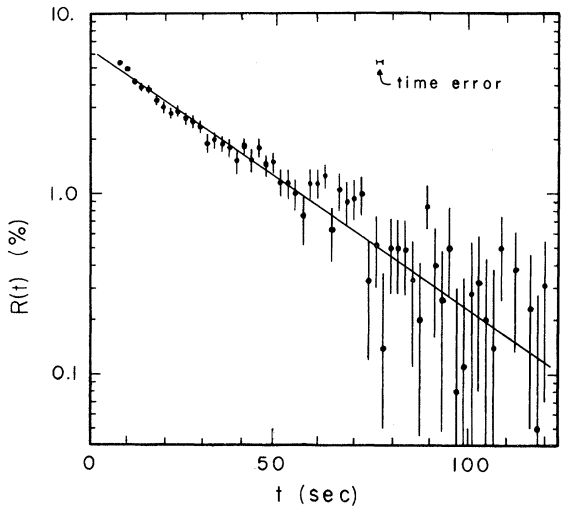

FIG. 1. Angular-correlation relaxation at reduced temperature of 5.9×10^{-4}.

ported here, the value of T_c was found to 0.05 K from study of the ferromagnetic region.[4]

For an atomic spin component $S_z(t)$, the spin correlation time is

$$\tau_z = \tfrac{1}{2}\int_{-\infty}^{\infty}[G^{zz}(t)/G^{zz}(0)]\,dt \tag{7}$$

where $G^{zz}(t) = \tfrac{1}{2}\langle[S_z(t), S_z(0)]_+\rangle$ is the spin autocorrelation function.

We view $h(t)$ both below and immediately above T_c as predominantly due to core polarization produced by localized Rh d-shell spins, $S(t)$. With this assumption, Eq. (4) becomes

$$\mathcal{H}(t) = AI\cdot S(t), \tag{8}$$

and by the use of the isotropy condition $\langle S_x{}^2\rangle = \langle S_y{}^2\rangle = \langle S_z{}^2\rangle = S(S+1)/3$ the relaxation rate becomes

$$\tau_2 = 6A^2\tau_z S_z{}^2 = 2A^2\tau_z S(S+1). \tag{9}$$

In the local-moment model of the field, $A = \gamma H_0/S$, where H_0 is the measured saturation average field at $T=0$ K. According to Reno,[8] $H_0 = 225\pm3$ kG and $\gamma = 1.026\pm0.007$, so that

$$1/\tau_2 = 1.06\times10^{19}\tau_z(S+1)/S\ \text{sec}^{-1}. \tag{10}$$

To justify the predominantly local origin of $h(t)$, we note the following.

(1) Below T_c, the magnitude of H requires the existence of core polarization, for in the phenomenological model of Shirley, Rosenblum, and Matthias,[5] the saturation field due to conduction-electron polarization is only $0.3H_0$.

(2) For our experiment range, $T_c < T < T_c + 6$ K, the magnetic coupling of Rh in Ni should have the same character as below T_c. This follows because Ni exists in fluctuating, quasiferromagnetic clusters having a size ranging from ∞ at T_c down to 50 Å at our highest temperature. The size of these clusters is thus always much greater than any estimate of the range of the magnetic interaction.

(3) As a test of the above, we have taken extensive new data[10] at a number of applied fields, and have re-examined Reno's data[8] for $T_c < T < T_c + 8$ K. Both data sets show that $H(T) = \alpha\mu(T)$, where $\alpha = 35$ is a constant of proportionality observed everywhere in the ferromagnetic region, and $M(T)$ is calculated from the equation of state of Arrott and Noakes.[11,12]

In addition, Eqs. (6)–(8) are subject to the conditions $\tau_z/\tau_N \ll 1$, $\tau_z\gamma H_0 \ll 1$, and $\tau_z/t \ll 1$. For the largest τ_z values observed, $\tau_z/\tau_N = 1.1\times10^{-5}$ and $\tau_z\gamma H_0 = 7.6\times10^{-3}$. The condition $\tau_z/t \ll 1$ is more problematical. It arises from the assumption that the limits of integration in Eq. (7), which are strictly $\pm t$, may be taken as $\pm\infty$. For critical fluctuations $G^{zz}(t)$ is *not* exponential but has a long tail. This means that t must exceed τ_z by a *large margin.* Having no reliable estimate of this margin, we looked for a reduced decay rate, i.e., rounding, in $R(t)$ near $t=0$, following a suggestion of Blume.[13] As evidence against rounding we cite measurements at $\epsilon = 5.9\times10^{-4}$ and 2.7×10^{-4}, the first of which is shown in Fig. 1. If slight rounding were present for $8 < t < 100$ nsec, one should expect severe rounding for experimentally inaccessible times $t < 8$ nsec. Extrapolation of the data to $t=0$ leads to $R(0) = 0.064 \pm 0.006$ and $R(0) = 0.068 \pm 0.008$, respectively, in agreement with Eq. (3). We conclude that severe rounding for $t < 8$ nsec does not occur, and that $\tau_z/t \ll 1$ is satisfied for our data.

Equation (10) thus provides a basis for calculating τ_z absolutely, albeit with some uncertainty. Even with nonlocal contributions to the field, however, the power law for τ_z will be unaltered if, as one would expect, the nonlocal spins have the same autocorrelation function as the local spins. Table I shows all available results for τ_2; derived τ_z values appear in Fig. 2 on the assumption that $S = \frac{1}{2}$.

In neutron scattering, one measures the van Hove function $S^{zz}(\vec{q}, \omega)$. It is the Fourier trans-

TABLE I. *Ni*Rh data.

Data of Ref. 2		Data of this work	
ϵ (10^{-4})	τ_2 (10^{-8} sec)	ϵ (10^{-4})	τ_2 (10^{-8} sec)
1.58	2.3 ± 0.9	1.11	0.9 ± 0.2
3.33	2.8 ± 0.5	2.23	2.1 ± 0.3
4.76	3.4 ± 0.7	2.70[a]	2.1 ± 0.2
7.30	4.2 ± 1.1	5.90[a]	3.0 ± 0.5
9.68	7.1 ± 1.7	6.48	4.0 ± 0.7
12.7	6.7 ± 1.0	9.00	4.8 ± 0.8
16.0	6.2 ± 1.3	40.0	10.5 ± 2.5
20.3	6.7 ± 1.4	105.	32.0 ± 6.7
26.3	9.6 ± 1.8	110.	29.2 ± 8.8
34.4	12.7 ± 3.2	134.	32.2 ± 6.7
46.5	18.9 ± 4.7		
71.7	14.3 ± 3.4		
108.	40.1 ± 13.2		

[a]Error in ϵ is $\pm 1.5\times10^{-4}$, all other points $\pm 0.9\times10^{-4}$.

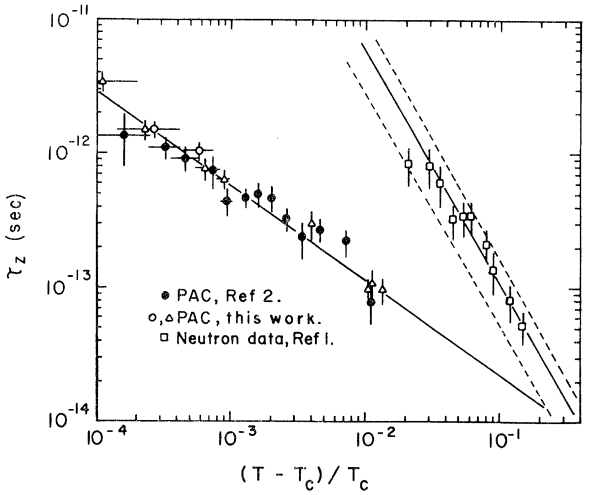

FIG. 2. Derived spin correlation times. Dotted lines around neutron data indicate systematic error.

form of the spin correlation function[14] $G^{zz}(\vec{r}, t)$:

$$G^{zz}(\vec{r}, t) = (1/\pi v_q)\int_{v_q} \exp(i\vec{q}\cdot\vec{r})\, d\vec{q} \int_{-\infty}^{\infty} \exp(i\omega t) S^{zz}(q, \omega)\, d\omega. \tag{11}$$

Using the definition of τ_z [Eq. (7)], and writing $G^{zz}(0, t) = G^{zz}(t)$, we have

$$\tau_z = 3J^{zz}(0)/S(S+1), \tag{12}$$

where

$$J^{zz}(\omega) = \tfrac{1}{2}\int_{-\infty}^{\infty} \exp(i\omega t) G^{zz}(t)\, dt = (\pi/v_q)\int_{v_q} S^{zz}(\vec{q}, \omega)\, d\vec{q}. \tag{13}$$

Thus, τ_z may be obtained by evaluating Eq. (13) for $\omega = 0$.

The neutron data have been analyzed using[14]

$$S^{zz}(q, \omega) = [S(S+1)/3\pi]\, \hat{\chi}(q)\, \Gamma(q)/[\omega^2 + \Gamma^2(q)], \tag{14}$$

where

$$\hat{\chi}(q) = (1/B\kappa^2)/[1 + (q/\kappa)^2] \text{ and } \Gamma(q) = cq^{5/2} f(\kappa/q) \tag{15}$$

are the reduced susceptibility and linewidth, respectively, and $f(\kappa/q)$ is a scaling function to be specified. The temperature enters the problem through the inverse correlation length κ. The use of a single length κ^{-1} to describe $\Gamma(q)$ and $\hat{\chi}(q)$ is in effect equivalent to dynamical scaling.[15]

For the Heisenberg model, with $x = \kappa/q$, Resibois and Piette[16] obtain

$$f(x) = c_1 x^{1/2} \text{ for } 2 < x < \infty, \quad f(x) = c_2 \text{ for } 0 < x < 2. \tag{16}$$

With this we obtain

$$\tau_z = (1/Bcv_q)\hat{J}(0)\kappa^{-3/2}, \tag{17}$$

where $\hat{J}(\omega)$ is a reduced form of $J^{zz}(\omega)$, and

$$\hat{J}(0) = 4\pi\{c_1^{-1}\int_0^{1/2}(1+x^2)^{-1}dx + c_2^{-1}\int_{1/2}^{\infty} x^{1/2}(1+x^2)^{-1}dx\}. \tag{18}$$

If $\kappa \propto \epsilon^{\nu}$ and $\chi(0) \propto \epsilon^{-\gamma}$, Eq. (15) implies $\gamma = 2\nu$, so that $\tau_z = D\epsilon^{-n}$ where $n = 1.5\nu = 0.75\gamma$. Since bulk measurements[17] show $\gamma = 1.34 \pm 0.01$, we expect that $n = 1.00 \pm 0.01$. The constants needed for Eq. (17) are given in Table II, calculated τ_z values in Fig. 2.

TABLE II. Constants for neutron determination of τ_2.

Constant	Value	Error	Source
B	1.5 Å	50%	Heller,[a] Ref. 1
c	8.2×10^{14} sec	20%	Refs. 1 and 16
v_a	2.9 $Å^{-3}$	1%	
$\hat{J}(0)$	27	10%	Ref. 16

[a]P. Heller, Rep. Progr. Phys. 30, 731 (1967).

Figure 2 shows that neutron scattering yields $n = 1.44 \pm 0.37$, PAC $n = 0.70 \pm 0.03$. Neither is consistent with the prediction $n = 1.00 \pm 0.01$. It is also seen that far from T_c, where the neutron data are expected to be most reliable, agreement between neutron and extrapolated PAC results is approached; and that near T_c a factor of 10^3 appears to separate the two.

We emphasize: PAC τ_2 values measure an integral property of $S^{zz}(q, \omega)$, and are thus indeon the other hand, are dependent on the form chosen for $S^{zz}(q, \omega)$. We therefore conclude that (a) the functional form of $S^{zz}(q, \omega)$ given by Eqs. (14) and (15) may be correct for $\epsilon \gtrsim 10^{-1}$, where neutron and PAC results appear to be consistent; (b) for $\epsilon \lesssim 10^{-2}$ the given form of $S^{zz}(q, \omega)$ must be incorrect, not only because neutron and PAC results diverge, but independently, because the PAC results violate the predicted temperature dependence.

Conclusion (b) may arise from failure of dynamical scaling, or the particular scaling function given in Eq. (16). It may also be that the neutron data are nonhydrodynamic, and have been incorrectly fitted by the hydrodynamic form of $S^{zz}(q, \omega)$.

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