The phase diagram is modified in an important way when fluctuations are taken into account. For a rotationally symmetric liquid the fluctuations in $\rho_{\vec{q}}$ above the transition depend only on the magnitude of q and thus have a one-dimensional dispersion. They depend only on $|q| - Q$. As shown by Brazovskii,[11] it follows that the continuous transition (predicted at C or in two dimensions) does not occur, because it would imply diverging fluctuations $\langle\rho^2\rangle$ as the transition is approached from above. As a result the transition is shifted downwards and is always first order, independent of dimensionality, provided that there is a divergent susceptibility for the field conjugate to the order parameter. The possibility of a liquid-solid critical point is thus eliminated. We have drawn the shifted phase boundary (P') schematically as a dash-dotted line in Fig. 1. Note that the single point C is now replaced by a segment (along this line P') of weakly first order L-S transitions. Below this line the bcc phase is essentially stable, compared to the liquid (but of course less stable than S). If the downward shift is sufficiently large the bcc pockets would completely disappear as thermodynamically stable phases. Thus one could have a segment of the phase boundary where one can nucleate metastable bcc structures even where there are no stable bcc phases and without drastic supercooling.

We would like to thank P. Pincus for encouragement, illuminating discussions, and help in bringing the authors together. One of us (S.A.) thanks both R. Orbach for bringing Ref. 2 to his attention, and the patient students of a seminar for giving him the opportunity to sort out the Landau theory for this problem. One of us (J.P.M.) thanks A. Rahman for sharing his insight and enthusiasm. This work was supported in part by the National Science Foundation, Grants No. CHE76-21293 and No. DMR75-19544 and the U. S. Office of Naval Research, Contract No. N0014-75-C-0245 P3.

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Crossover in the Dynamic Exponent z for Three-Dimensional Ferromagnets

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For isotropic three-dimensional ferromagnets, neutron-scattering and hyperfine-interaction experiments yield 2.5 and 2.0, respectively, for the dynamical critical exponent z. This apparent contradiction is removed by two hypotheses. (1) The effective value of z depends on the wave-vector region sampled, and for small q approaches $z = 2.0$. (2) Hyperfine-interaction experiments are asymptotic in q, while present neutron-scattering experiments are not.

Near a magnetic critical point, the spin-spin correlation function for the ith spin component, $S^{ii}(\vec{q}, \omega)$, depends on three critical exponents, η, ν, and z. According to current theory, the static exponents η and ν are universal within a class of systems having the same lattice dimensionality, d, and spin dimensionality, N; the dynamic exponent, z, depends in addition on the conservation laws describing the motions of the spins.[1] Since $S^{ii}(\vec{q}, \omega)$ is proportional to the cross section for inelastic neutron scattering, critical fluctuations in ferromagnets and antiferromagnets were initially probed by that technique. Results largely confirm the predicted forms for $S^{ii}(\vec{q}, \omega)$ and yield the universal exponents expected for various classes.[2]

More recently critical fluctuations have also been probed by hyperfine interactions (nuclear magnetic resonance, perturbed angular correlations, and Mössbauer effect). For antiferromagnets, results are consistent with theory and neutron scattering.[3] For $(d, N) = (3, 3)$ ferromagnets, however, the divergence of the spin autocorrelation time, which depends on the exponent $n = \nu(z - \eta - 1)$, has consistently yielded $n \simeq 0.7$, whereas neutron scattering and theory for the Heisenberg model imply $n \simeq 1$. This means either that hyperfine-interaction experiments are flawed or that the model which successfully describes neutron scattering does not apply to hyperfine-interaction measurements.

In this Letter we sketch how scaling-theory predictions for $S^{ii}(\vec{q}, \omega)$ are tested by neutron and hyperfine-interaction experiments, present new evidence on the latter's validity, and conclude that the simple model for ferromagnets must be modified. Guided by recent developments in theory,[1,4] and the fact that the two kinds of experiments sample q space differently, we obtain a plausible and consistent picture on the assumption that the expoent z crosses from an effective value of 2.5 to an asymptotic value of 2.0 as q goes to zero.

Tests of theory.—According to scaling theory, for temperatures $T > T_c$,[1]

$$S^{ii}(\vec{q}, \omega) = 2\pi[\omega^{ii}(\vec{q})]^{-1} S^{ii}(\vec{q}) f_{\vec{q}}[\omega/\omega^{ii}(\vec{q})], \quad (1)$$

where in the limits $q \to 0$ and $T \to T_c$, $S^{ii}(\vec{q})$ and $\omega^{ii}(\vec{q})$ are homogeneous functions of q, related by the same correlation length $1/\kappa$:

$$S^{ii}(\vec{q}) = q^{-2+\eta} g^{ii}(\kappa/q); \quad (2)$$

$$\omega^{ii}(\vec{q}) = q^z \Omega^{ii}(\kappa/q). \quad (3)$$

These relations define η and z. A temperature dependence is carried by κ, which has the asymptotic form

$$\kappa = \kappa_0 t^\nu, \quad t \equiv |T - T_c|/T_c. \quad (4)$$

Neutron scattering can, in principle, determine all exponents and scaling functions. To find z the energy linewidth at T_c is fitted to the power law

$$\omega^{ii}(\vec{q}) = q^z \Omega^{ii}(0). \quad (5)$$

To find η and ν, data for $S^{ii}(\vec{q}, \omega)$ are integrated according to

$$S^{ii}(\vec{q}) = \tfrac{1}{2}\pi \int_{-\infty}^{+\infty} d\omega\, S^{ii}(\vec{q}, \omega). \quad (6)$$

and fitted to the modified Ornstein-Zernike function

$$S^{ii}(\vec{q}) = B^{ii} q^{-2+\eta}[1 + (\kappa/q)^2]^{-1}. \quad (7)$$

Spectrometer geometry and Bragg-scattering widths limit experimentally accessible momentum transfer to $q > 0.05$ Å^{-1}.[2]

Hyperfine-interaction experiments yield the spin autocorrelation time τ_i, defined as the integral of the space-time correlation function:

$$\begin{aligned} \tau_i &\equiv \int_0^\infty [G^{ii}(\vec{r}, t)/G^{ii}(0, 0)]dt \\ &= C_0 \int_{v_q} d^d q\, S^{ii}(\vec{q}, 0) \\ &= C_0 \int_{v_q} d^d q\, q^{-2+\eta-z} g^{ii}(\kappa/q)/\Omega^{ii}(\kappa/q). \end{aligned} \quad (8)$$

Here, C_0 is a constant and v_q is the Brillouin-zone volume. With Eq. (4), the temperature dependence may be removed from the integral, yielding

$$\tau_i = C_0' t^{-n} \int_0^\infty dx\, x^{d-3+\eta-z}[g^{ii}(x)/\Omega^{ii}(x)], \quad (9)$$

$$n = \nu(z + 2 - d - \eta). \quad (10)$$

Thus, the nature of the divergence of τ_i at T_c involves all three critical exponents but is independent of the forms of the scaling functions.

Deduction of n from hyperfine-interaction experiments requires use of nuclear relaxation theory to relate observed nuclear relaxation effects to τ_i. For an isotropic hyperfine interaction $A\vec{S}\cdot\vec{I}$, there is one spin correlation time, $\tau_c = \tau_i$, $i = x, y, z$. Mössbauer line broadening, $\Delta\Gamma$ (for spin $\frac{3}{2}$-$\frac{1}{2}$ transitions), and angular correlation or resonance relaxation times τ_r are then given by[5,6]

$$\begin{aligned} \Delta\Gamma &= \tau_c S(S+1)[\tfrac{15}{4}A_e^2 - \tfrac{5}{2}A_e A_g + \tfrac{3}{4}A_g^2], \\ 1/\tau_r &= \tau_c 2A^2 S(S+1), \end{aligned} \quad (11)$$

provided τ_c is the shortest time scale in the problem. As shown in previous work,[7-9] both the condition of isotropy and the limitations on τ_c are well satisfied for accessible reduced temperatures in Fe and Ni. Additional confirmation of isotropy for Ni comes from recent NMR experiments[10] which show that $T_1 = T_2$ near T_c (using the usual notation).

Experimental results.—Experimental[8-13] and theoretical[4,14-17] results for $(d, N) = (3, 3)$ ferromagnets are compared in Table I. As can be

TABLE I. Theory and experiment for $(d,N)=(3,3)$ ferromagnets.

	ν	z	n	Ref.
		Theory		
Heisenberg	0.705(1)	2.5	1.03[a]	14, 15
Heisenberg + dipole	0.69	2.0	0.7[a]	4, 16
Heisenberg + dipole	0.69	1.0	0.0[a]	16, 17
		Neutron scattering		
Co	0.65(4)	2.4(2)	0.98(15)[a]	11
Fe	0.69(2)	2.7(2)	1.1(1)[a]	12
Ni	0.96(25)	2.46(25)	1.0(2)[a]	12
EuO	0.70(2)	2.29(3)	0.90(2)[a]	13
EuS	0.70(2)	...	...	13
		Hyperfine interactions[b]		
$Ni\,^{100}$Rh/PAC	...	2.03(3)[a]	0.70(3)	7
$Ni\,^{57}$Fe/ME	...	2.0(3)[a]	0.71(24)	8
$Ni\,^{61}$Ni/NMR[c]	...	2.0(1)[a]	0.67(8)	10
$Fe\,^{57}$Fe/ME	...	1.9(2)[a]	0.62(13)	9

[a]Using $n=(z-\eta-1)$, with $\eta=0$ and $\nu=0.7$.

[b]Techniques are: PAC, perturbed angular correlations; ME, Mössbauer effect; NMR, nuclear magnetic resonance.

[c]Ni NMR data were taken below T_c and may require alternative interpretation to that given in the text.

seen, neutron-scattering results[11-13] agree with predictions based on the Heisenberg model, while hyperfine-interaction experiments[7-10] are consistent with one of the calculations including the effects of dipole interactions.

How reliable are the hyperfine-interaction results? The reduced temperature region in which critical fluctuations are visible via nuclear relaxation is limited, and for available probes lies quite close to T_c, as shown in Fig. 1. Under these conditions a spread of T_c's or temperature gradients can easily mimic nuclear relaxation and cause erroneous conclusions about the temperature dependence of τ_c. To rule out significant temperature-gradient distrubances we have in past work shown that relaxation effects are *irreducible*, i.e., they do not get smaller when improvements in temperature uniformity are made.

As an additional test, we have conducted a new and decisive null experiment. As can be seen from Fig. 1, the system $Ni\,^{111}$Cd has hyperfine coupling so weak that critical fluctuations induced nuclear relaxation lies too close to T_c to be observable. Hence if $Ni\,^{111}$Cd were to show "relaxation," the cause must be temperature gradients or a spread of T_c's. Other results based on similar experimental technique would then be in doubt. However, we have taken data at $t \simeq 8\times10^{-5}$ above T_c and have observed no relaxation; we are therefore confident that relaxation observed in previous experiments[7-9] was due only to critical fluctuations.

Interpretation.—Why do neutron-scattering experiments agree with one model of dynamics while hyperfine-interaction measurements agree with another? To begin with, we note that in any physical system, some spin-nonconserving forces will be present, for example, dipolar or spin-

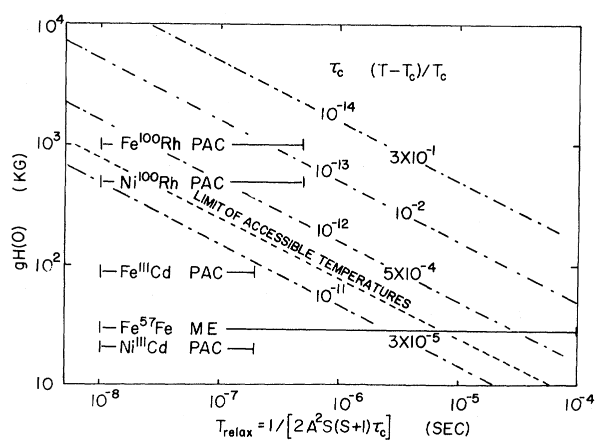

FIG. 1. Nuclear relaxation time as a function of hyperfine-interaction strength for various values of the spin autocorrelation time or reduced temperature. The temperature scale was taken from Ref. 7. Horizontal bars indicate the range that may be studied for various probe-host combinations, and shows that some cases lie outside the limit of accessible reduced temperature.

lattice forces. Even if these are small, we expect them to modify the dynamics at long wavelengths.[1,4,17] What is observed in actual experiments will depend on the size of the asymptotic region in q space. With this insight, we explain Table I as follows:

(1) Hyperfine-interaction experiments for the temperature range sampled ($10^{-4} < t < 10^{-2}$) are dominated by sufficiently small q so as to yield asymptotic behavior in agreement with the proper treatment of dipolar interactions.[4]

(2) Neutron scattering experiments for the wavelength range sampled ($0.05 < q < 0.5$ Å^{-1}) are dominated by nonasymptotic values of q and hence yield effective values of z that are indicative of Heisenberg or crossover behavior, i.e., $2.3 < z < 2.9$.

Hyperfine-interaction results are consistent with electron-spin-resonance studies of the uniform mode ($q=0$) relaxation rate. The weak temperature dependence predicted by Teitelbaum[4] for small t has been seen in $GdCl_3$,[18] EuS,[18] and EuO.[19] The predictions due to Maleev[17] appear to be incorrect, possibly because of the neglect of demagnetization effects.[4]

Additional support for the above hypotheses comes from explicit evaluation of the integral in Eq. (9), using approximate forms for the two scaling functions for a Heisenberg system.[20] As shown in Fig. 2, for the range of t sampled in hyperfine-interaction experiments the predominant fraction of the integral lies below the minimum value sampled by neutron scattering experiments.

From an experimental point of view, it would

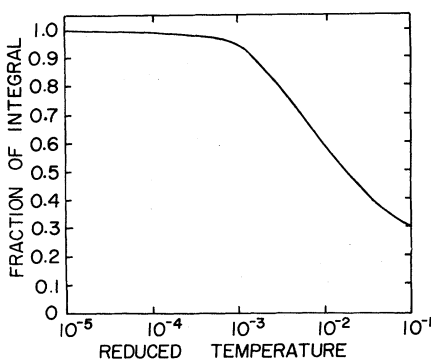

FIG. 2. Fraction of the integral in Eq. (9) contributed by the wave-vector region $q < 0.05$ Å^{-1}, the minimum q accessible in neutron-scattering experiments. The calculation is based on Heisenberg scaling functions. Qualitatively similar behavior is expected in real ferromagnets.

be interesting to observe crossover in n or z directly. For hyperfine-interaction experiments, this can be achieved if a broader range of t is available (Fig. 2), as in Fe^{100}Rh (Fig. 1), or if non-Heisenberg distribances are smaller, as may be the case in some Heusler alloys.[21] For neutron scattering, crossover might be seen if strongly dipolar materials such as EuS ($T_c = 17$ K) are carefully studied. It appears, however, that there are serious resolution problems involved in such experiments.[22]

From a theoretical point of view, calculations that describe $S^{ii}(\vec{q}, 0)$ in the crossover region are highly desirable. Such information would allow quantitative comparison of the wave-vector ranges sampled in neutron and hyperfine-interaction work.

We thank R. Birgeneau, H. Gould, P. Hohenberg, S. Ma, G. Mazenko, and L. Passell for helpful comments on an earlier version of the manuscript. This work was supported in part by National Scientific Foundation Grant No. DMR77-01250.

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[22]L. Passell (private communication) has pointed out to us the difficulty in resolving the narrow energy widths in low-temperature systems like EuS.

Influence of the Electron-Phonon Interaction on the de Haas–van Alphen Effect in Mercury

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Accurate measurements of de Haas–van Alphen effect amplitudes in mercury show, for the first time, departures from quasiparticle behavior. The departures arise from the influence of the electron-phonon interaction.

The controversy relating to the influence of the electron-phonon interaction on the de Haas–van Alphen (dHvA) effect has an interesting history. Wilkins and Woo[1] first demonstrated that the cyclotron effective mass which appears in the thermal damping factor will be renormalized by the electron-phonon interaction. In addition to this, however, Palin[2] anticipated that the Dingle factor,[3] which embraces the effect of electron collisions, might also contain a term proportional to T^3, such as affects the line shape in Azbel-Kaner cyclotron resonance,[4] and that such a term would lead to significant departures from the original theory of Lifshitz and Kosevich[5] (LK) in the amplitude of dHvA-effect oscillations. Surprisingly, however, even for the most favorable metal in this respect, namely mercury with its low-lying phonon mode ($\Theta_D \sim 21$ K) and strong electron-phonon coupling, no such departures were observed.[2] This unexpected result was explained by Engelsberg and Simpson[6] (ES), who showed that the effect of electron-phonon interactions could be incorporated in the oscillatory part of the thermodynamic potential by replacing, at a certain state, the noninteracting single-particle electron energies by the noninteracting energy plus full electron-phonon self-energy. Their result, which is an extension of earlier work by Fowler and Prange,[7] indicates that, with regard to the amplitude of the dHvA effect under accessible laboratory conditions, the departures from the LK theory are expected to be small, even for mercury, and are not inconsistent with the experiments of Palin.[2] This result is sometimes interpreted as being due to a cancellation between changes in the mass-renormalization and electron-phonon scattering rate. However, Engelsberg[8] has shown that this description is not completely valid and has interpreted departures from LK as being the result of a failure of the quasiparticle approximation. The same problem, however, has also been discussed by Gantmakher[9] who claims that Palin's apparently null result follows from a "classical" calculation of electron scattering in a magnetic field in the presence of electron-phonon interactions. The purpose of this Letter is to present new experiments on dHvA amplitudes in mercury, under conditions rather more favorable than those of Palin, in which, for the first time, we have observed departures from quasiparticle behavior. Aside from underpinning the theory of the electron-phonon interaction, these results, we believe, have implications for the experimental investigation of other many-body effects in metals using the dHvA effect.

Prior to discussing the experimental results we shall comment briefly on the theory of ES in order that the optimum experimental conditions for observing the effects might be appreciated and also that, in what follows, we may compare theory and experiment. According to ES, the entire effect of electron-phonon interactions is contained in a term, A_r, for the amplitude of the rth harmonic of the dHvA effect so that

$$A_r = \sum_{n=0}^{\infty} \exp\{(-2\pi r/\hbar\omega_c)[\omega_n + \zeta(\omega_n)]\}, \qquad (1)$$

in which ω_c is the cyclotron frequency $(= eB/m_c)$,

