Scaling Laws for Dynamic Critical Phenomena

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The usual static scaling laws are generalized to nonequilibrium phenomena by making assumptions on the behavior of time-dependent correlation functions near the critical point of second-order phase transitions. At any temperature different from T_c, the correlation functions are assumed to reflect the hydrodynamic behavior of the system, for sufficiently long wavelengths and low frequencies. As the critical temperature is approached, however, the range of spatial correlations in the system diverges, and the domain of applicability of hydrodynamics is reduced to a vanishingly small region of wavelengths and frequencies. The dynamic-scaling assumptions lead to predictions for the behavior of the hydrodynamic parameters near T_c, as well as for the form of the correlation functions for macroscopic distances and times, outside the hydrodynamic range. In particular, singularities are predicted to occur in the temperature dependence of transport coefficients, and anomalies are expected in the frequency spectrum of certain operators, which are observable by inelastic scattering of neutrons or light. A distinction is made between the restricted dynamic-scaling hypothesis, which refers to the order parameter only, and extended dynamic scaling, which applies to other operators and involves stronger assumptions. Applications are discussed to antiferromagnets, ferromagnets, the gas-liquid critical point, and the λ transition in superfluid helium. Specific experiments are suggested to test the scaling assumptions, and existing experimental evidence is briefly reviewed. Finally, a comparison is made with other theories of dynamical behavior near critical points.

I. INTRODUCTION

IN recent years there has been considerable activity, both theoretical and experimental, in the field of critical phenomena.[1–3] The primary aim of this research has been to determine the precise form of the singularities which occur in equilibrium[4] properties at the critical point of a "second-order" phase transition. Although this problem has not been solved theoretically for any realistic system, the solution of the two-dimensional Ising model[5] paved the way to an extensive series of numerical and phenomenological investigations, as well as many experiments for various systems.[1–3] From this work, it has become clear that the "classical" or "mean-field" theories[6,7] of second-order phase transitions are not quantitatively correct.[8] One attempt to find a phenomenological description of the Ising model which could be generalized to other systems was made by Kadanoff,[9] who proposed a "scaling theory," leading to relations between the various critical exponents.[10] This approach turned out to be equivalent to an earlier theory of Widom,[11] who made assumptions about the form of the equation of state and the correlation function of a fluid near its critical point. For various systems a number of other authors[12] have proposed similar relations between the critical exponents, based either on empirical observations, or heuristic arguments. These relations, which have come to be known as

[1] L. P. Kadanoff, W. Götze, D. Hamblen, R. Hecht, E. A. S. Lewis, V. V. Palciauskas, M. Rayl, J. Swift, D. Aspnes, and J. Kane, Rev. Mod. Phys. **39**, 395 (1967).

[2] M. E. Fisher, Rept. Progr. Phys. **30**, 615 (1967).

[3] P. Heller, Rept. Progr. Phys. **30**, 731 (1967).

[4] The equilibrium properties of interest near the critical point include the magnetization and susceptibility for a magnetic system, the density and compressibility for a gas-liquid transition, the superfluid density ρ_s for liquid helium, and the specific heat in all cases. Another "equilibrium" property of interest is the spatial dependence of the equal-time autocorrelation function for spin or density fluctuations.

[5] L. Onsager, Phys. Rev. **65**, 117 (1944); also S. G. Brush, Rev. Mod. Phys. **39**, 883 (1967). The historical importance of Onsager's solution cannot be overemphasized, since it provided the most solid evidence that phase transitions are indeed describable by equilibrium statistical mechanics. Moreover, the mathematical singularities in this exact solution are analogous to the "apparent singularities" which are observed in real finite systems at temperatures approaching the critical point.

[6] See, e.g., R. Brout, *Phase Transitions* (W. A. Benjamin, Inc., New York, 1965).

[7] L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Addison-Wesley Publishing Co., Inc., Reading, Mass., 1958), Sec. 135.

[8] In many cases mean-field theory is a reasonable approximation far from the critical point, but breaks down for $\epsilon \equiv |T-T_c|/T_c \ll 1$. It is precisely the behavior in this latter region, generally called the critical region, which is the concern of the present paper. For some systems, however, such as superconductors, there is an intrinsic small parameter $\theta \ll 1$ such that mean-field behavior breaks down, and critical behavior sets in only for $\epsilon \ll \theta$. [For the pure superconductor $\theta = (k_B T_c/E_F)^4$.] The critical region in bulk superconductors is so small as to be out of reach experimentally, and we shall not discuss these systems in the present paper. A general discussion of the breakdown of mean-field theory may be found in P. C. Hohenberg, in Proceedings of the Conference on Fluctuations in Superconductors, Asilomar, California, 1968 (unpublished; obtainable from Stanford Research Institute, Palo Alto, Calif.). Other systems with a small parameter may have two "critical" regions of different behavior: $\theta \ll \epsilon \ll 1$ and $\epsilon \ll \theta$. Some examples will be discussed below.

[9] L. P. Kadanoff, Physics **2**, 263 (1966).

[10] The critical exponents are assumed to specify completely the nature of the thermodynamic singularity. They are defined and discussed in Refs. 1–3. When a small parameter θ exists in a system (Ref. 8), the true critical exponents are only defined for $\epsilon \ll \theta$, but "apparent" exponents may often be discussed in the range $\theta \ll \epsilon \ll 1$, since they correspond to true critical exponents for a model in which $\theta = 0$.

[11] B. Widom, J. Chem. Phys. **43**, 3892 (1965); **43**, 3898 (1965).

[12] J. W. Essam and M. E. Fisher, J. Chem. Phys. **39**, 842 (1963); M. E. Fisher, J. Appl. Phys. **38**, 981 (1967); C. Domb and D. L. Hunter, Proc. Phys. Soc. (London) **86**, 1147 (1965); E. Helfand, paper presented at a meeting of the American Physical Society, 1965 (unpublished); G. E. Uhlenbeck and P. C. Hemmer, in *Proceedings of the International Symposium on Statistical Mechanics and Thermodynamics, Aachen, Germany* (North-Holland Publishing Co., Amsterdam, 1965); A. Z. Patashinskii and V. L. Pokrovskii, Zh. Eksperim. i Teor. Fiz. **50**, 439 (1966) [English transl.: Soviet Phys.—JETP **23**, 292 (1966)].

"scaling laws," have been extensively compared to experiments and to numerical calculations on model systems.[1-3] The over-all agreement is quite satisfactory and certainly much better than is obtained from the "classical exponents" of mean-field theory, although a number of persistent discrepancies remain primarily with numerical work on the three-dimensional Ising model.[2]

For dynamic[13] phenomena, on the other hand, there is no exactly soluble model which exhibits interesting properties at the critical point, nor even a well-defined mean-field theory, which would lead to unambiguous general predictions. This is largely because dynamical properties depend much more crucially on such details of the Hamiltonian as the conservation laws and the interparticle potential than do static properties. In consequence, much less is known about dynamic critical behavior, and previous theories have at best provided a qualitative guide to experimental observations.

The first general prediction was made by Van Hove[14] in his discussion of "critical slowing down," of density or spin fluctuations, which could be detected by neutron-scattering experiments. In the case of a Heisenberg ferromagnet in the paramagnetic phase ($T>T_c$, $H=0$) Van Hove's argument may be sketched by writing down the macroscopic equation which governs the slow variations of the magnetization in space and time,

$$\partial\mathbf{M}(\mathbf{r},t)/\partial t=D\nabla^2\mathbf{M}(\mathbf{r},t). \tag{1.1}$$

The spin-diffusion coefficient D satisfies the Einstein relation

$$D=\Lambda/\chi^{\mathrm{M}} \tag{1.2}$$

in terms of the transport (Onsager) coefficient Λ, and the magnetic susceptibility χ^{M}. Van Hove argued that the transport coefficient Λ depends primarily on the short-range behavior of the system, and should therefore remain finite at the critical point; (subsequent approximate calculations, based largely on mean-field theory, supported this assertion[15]). The susceptibility, on the other hand, was known to diverge at the critical point, so that D was predicted to vanish at T_c. When the mean-field temperature dependence was used for χ^{M}, the diffusion constant was predicted to be linear in $T-T_c$. As Van Hove pointed out, this slowing down of the spin diffusion would show up as a reduction of the inelasticity of neutron scattering by magnetic systems, as T approached T_c. Attempts to observe this reduction were not entirely successful,[16] and it is only recently that the reasons for this failure have begun to be clarified. In particular, Marshall[17] pointed out that most neutron experiments are not done at wavelengths large compared to the correlation range near T_c, and that therefore the Van Hove theory does not apply. The alternative theory that he outlined was not very specific, but it did suggest that "some remnant of spin-wave motion should exist above T_c."

A slightly different line of investigation was initiated by Fixman,[18] who attempted to calculate various transport coefficients at the critical point of simple fluids and fluid mixtures. The main physical idea contained in Fixman's calculations is that the long-range spatial correlations predicted by the Ornstein-Zernike[19] theory should lead to enhanced fluctuations and anomalous transport properties. The quantitative predictions which followed from this interesting idea could not be expected to be correct, since the temperature dependence of the Ornstein-Zernike correlation function is known to be wrong. Furthermore, the attempt by Fixman[18] and others[20] to apply the same idea to a calculation of the anomaly in specific heat at T_c overestimates considerably the effect of critical fluctuations. Nevertheless, the notion that long-range spatial correlations can affect the transport coefficients is an attractive one, and has led Kawasaki and co-workers,[21] and also Kadanoff and Swift,[22] to develop a more general semiphenomenological approach which is less tied to the mean-field and Ornstein-Zernike theories. These authors have made specific predictions concerning the singularities of transport coefficients in a number of systems.

From a purely phenomenological point of view, it is natural to ask whether the scaling laws of Widom,[11] Kadanoff,[9] and others[12] can give any information about dynamic properties which could serve to test their validity or to broaden their range of applicability. Such an approach was followed by Ferrell and co-workers,[23] in their study of the λ point of He^4. These authors predicted an anomalous damping of second sound below T_λ and a singular thermal conductivity above T_λ.

In this paper we present, in somewhat greater detail, a theory of dynamic scaling which was previously intro-

[13] The "dynamic" phenomena of interest include the time-dependent correlation functions and various transport coefficients and relaxation rates, such as spin-diffusion constants, thermal and electrical conductivities, acoustic attenuation coefficients, viscosities, etc.

[14] L. Van Hove, Phys. Rev. **93**, 1374 (1954).

[15] P. G. de Gennes, J. Phys. Chem. Solids **4**, 223 (1958); P. G. de Gennes and J. Villain, *ibid.* **13**, 10 (1960); H. Mori and K. Kawasaki, Progr. Theoret. Phys. (Kyoto) **27**, 529 (1962).

[16] B. Jacrot, J. Konstantinovic, G. Parette, and D. Cribier, *Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1963), p. 317; L. Passell, K. Blinowski, T. Brun, and P. Nielsen, Phys. Rev. **139**, A1866 (1965).

[17] W. Marshall, Natl. Bur. Std. (U. S.) Misc. Publ. **273**, 135 (1966).

[18] M. Fixman, J. Chem. Phys. **47**, 2808 (1967), and references therein.

[19] L. S. Ornstein and F. Zernike, Proc. Acad. Sci. Amsterdam **17**, 793 (1914).

[20] M. Fixman, J. Chem. Phys. **36**, 1597 (1962); W. Botch and M. Fixman, *ibid.* **42**, 196 (1965); I. A. Kvasnikov, Dokl. Akad. Nauk SSSR **119**, 475 (1958) [English transl.: Soviet Phys.—Doklady **3**, 329 (1958)]; see also Ref. 6.

[21] K. Kawasaki, Phys. Rev. **150**, 291 (1966); K. Kawasaki and M. Tanaka, Proc. Phys. Soc. (London) **90**, 791 (1967).

[22] L. P. Kadanoff and J. Swift, Phys. Rev. **166**, 89 (1968); Ann Phys. (N. Y.) (to be published).

[23] R. A. Ferrell, N. Menyhárd, H. Schmidt, F. Schwabl, and P. Szépfalusy, Phys. Rev. Letters **18**, 891 (1967); Ann. Phys. (N. Y.) **47**, 565 (1968).

duced as a reformulation and generalization of the theory of Ferrell *et al.*,[23] and applied to magnetic systems.[24] We distinguish here between a restricted (weak) dynamic-scaling hypothesis, and an extended (strong) hypothesis, which is necessary to reproduce some of the previously derived results.[23,24] In both cases we attempt to make our assumptions explicit, and to avoid the use of macroscopic concepts outside their range of applicability. In certain systems our results agree with the quasimicroscopic calculations of Kadanoff and Swift,[22] although their theory is more detailed and therefore allows a calculation of additional transport coefficients which do not follow from scaling. On the other hand, our approach is not restricted to the hydrodynamic limit, and so we make predictions which fall outside the "long-wavelength" region, where transport coefficients are defined. In particular, we are interested in the results of critical scattering of neutrons or light which do not involve purely macroscopic quantities near T_c. Whenever possible we attempt to suggest experimental tests of the assumptions of the theory and to evaluate their practical observability.

In Sec. II, the macroscopic information contained in the correlation functions is reviewed, and related to the phenomenon of phase transitions. In Sec. III, a qualitative description is presented of the main physical ideas contained in the scaling theories. Section IV is devoted to a more precise formulation of the scaling hypotheses, both static and dynamic, and restricted and extended. In Sec. V, applications to specific systems are discussed; these include the antiferromagnet, the ferromagnet, the gas-liquid critical point of a simple fluid, and finally the λ point of superfluid helium. A number of experiments are suggested and a few existing ones very briefly discussed. In Sec. VI, a comparison is made with other theories.

II. CORRELATION FUNCTIONS FOR "MACROSCOPIC" OPERATORS

In this section, we wish to summarize the important information contained in the correlation functions of observable operators in any macroscopic system. Although the facts stated here are well known,[25,26] it is probably useful to reiterate them in a unified notation in order to make the subsequent discussion clearer.[27]

Let us consider a system which is described by thermodynamic variables, whose densities satisfy a set of macroscopic (hydrodynamic) equations, valid at long wavelengths and low frequencies.[28] These variables may be the usual thermodynamic parameters such as the density n, the entropy S, the pressure p, or the magnetization $\mathbf{M}$. In many cases the macroscopic description also involves "order parameters" or "quasiconstants" of the motion, such as the fluxoid in a superconductor, the superfluid velocity $\mathbf{v}_s$ in helium, or the *staggered* magnetization $\mathbf{N}$ in an antiferromagnet. In general, the hydrodynamic equations may be linearized, for small departures from equilibrium, and various long-lived transport modes identified, with frequencies $\omega_i(\mathbf{k})$ and decay rates $\Gamma_i(\mathbf{k})$. If $\omega_i(k)$ and $\Gamma_i(k)/\omega_i(k)$ approach zero as $k\rightarrow 0$, then the frequency $\omega_i(\mathbf{k})$ may be related to purely thermodynamic quantities, whereas the decay rate $\Gamma_i(k)$ is given in terms of thermodynamic parameters *and* transport coefficients.

An alternative description starts from the *microscopically* defined[29] densities of conserved quantities and quasiconstants of the motion, and their correlation functions in the equilibrium ensemble. In the long-wavelength low-frequency limit,[28] these correlation functions contain all the information inherent in the macroscopic description, whenever the latter is applicable. The correlation functions are more general, however, since they may be defined and often measured outside of the hydrodynamic domain. Specifically, given a Hermitian operator $A(\mathbf{r},t)$, the dynamic and static correlation functions are defined, respectively, as[30]

$$\hat{C}^A(\mathbf{r},t)\equiv\tfrac{1}{2}\langle\{(A(\mathbf{r},t)-\langle A(\mathbf{r},t)\rangle),\,(A(0,0)-\langle A(0,0)\rangle)\}\rangle$$

$$\equiv\int\frac{d^3k}{(2\pi)^3}\int\frac{d\omega}{2\pi}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)}C^A(\mathbf{k},\omega)\quad(2.1)$$

and

$$\hat{C}^A(\mathbf{r})\equiv\hat{C}^A(\mathbf{r},\,t=0)\equiv\int\frac{d^3k}{(2\pi)^3}e^{i\mathbf{k}\cdot\mathbf{r}}C^A(\mathbf{k})\,,\quad(2.2)$$

where the angular bracket is an equilibrium expectation value, and the curly bracket is an anticommutator. If $\hat{C}^A(\mathbf{r})$ decays exponentially at large r, then we may

[24] B. I. Halperin and P. C. Hohenberg, Phys. Rev. Letters **19**, 700 (1967).

[25] L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Addison-Wesley Publishing Co., Inc., Reading, Mass.), Chap. XVII.

[26] L. P. Kadanoff and P. C. Martin, Ann. Phys. (N. Y.) **24**, 419 (1963).

[27] Some of the assumptions needed to derive the relation between macroscopic laws and correlation functions are discussed in Ref. 37.

[28] By this we mean wavelengths long compared to all microscopic lengths such as mean free paths, interparticle spacings, force ranges, or "correlation ranges," and frequencies small compared to microscopic ones, such as collision or internal excitation frequencies of molecules, as well as frequencies small compared to $k_BT/\hbar$.

[29] An operator is "microscopically defined" if it is expressible in terms of the field operators and interaction constants of the microscopic theory. In a fluid, for example, the energy density is microscopic, whereas the entropy density is not. As discussed below, there exists an entropy operator which is a linear combination of energy density and particle density, and whose fluctuations reduce to those of the entropy in a suitable long-wavelength limit. Outside of this limit, however, the identification of this operator with entropy is meaningless.

[30] In order to be able to treat a wide variety of systems in a unified language, we have departed from the notation of Ref. 24 in a number of significant ways. Thus, for example, the static correlation function is $C_\xi(\mathbf{k})$ rather than $N_\xi(\mathbf{k})$; the total and staggered magnetizations are $\mathbf{M}$ and $\mathbf{N}$, respectively, rather than $\mathbf{S}$ and $\mathbf{M}$; and the correlation length ξ is positive for all temperatures, rather than having the sign of $T-T_c$; in addition the correlation function [Eq. (2.1)] is *one-half* the anticommutator in the present paper.

define the *range* r_A of $\hat{C}^A(\mathbf{r})$ by the relation

$$\lim_{r\to\infty} \hat{C}^A(\mathbf{r}) = G^A(\mathbf{r})e^{-r/r_A}, \tag{2.3}$$

where $G^A(\mathbf{r})$ varies more slowly than the exponential at large distances. The range is in general a microscopic length.

If $\hat{C}^A(\mathbf{r})$ does not decay exponentially at large distances, but according to a power law p, we shall define r_A by[30a]

$$\hat{C}^A(\mathbf{r}) \sim |\langle A\rangle|^2(r_A/r)^p \quad \text{as} \quad r\to\infty. \tag{2.4}$$

Definition (2.4) will only make sense, of course, if the expectation value $\langle A\rangle$ is nonzero.

The macroscopic information is contained in the correlation functions in the following way:

(i) *Static correlations.* Classically, the long-wavelength limit of the correlation function is proportional to a static susceptibility χ^A,

$$\lim_{k\to 0} C^A(\mathbf{k}) = k_BT\chi^A, \tag{2.5}$$

where χ^A is the derivative of the equilibrium value of $\langle A\rangle$ with respect to an appropriately defined conjugate field. In a fluid, for instance,

$$\chi^n = n\left(\frac{\partial n}{\partial p}\right)_T = (k_BT)^{-1}\int d^3r\; C^n(\mathbf{r}),$$

where n is the density and $n^{-2}\chi^n$ is the compressibility. Equation (2.5) also holds for a quantum system insofar as $C^A(\mathbf{k},\omega)$ is dominated at long wavelengths by frequencies satisfying the condition $\hbar\omega \ll k_BT$. We believe this to be the case, at least near T_c, for all the functions that we shall consider.

(ii) *Dynamic correlations.* The correlation function $C^A(\mathbf{k},\omega)$, for fixed (small[28]) k, has poles at the complex frequencies $\omega_i(k)\pm i\Gamma_i(k)$ corresponding to the modes of linearized hydrodynamics. The residue is determined by the coupling strength of the given mode to the particular operator A, and is proportional in general to a thermodynamic derivative. Since, as mentioned earlier, the quantities ω_i and Γ_i may be related to thermodynamic derivatives and transport coefficients, we expect certain exact relations to hold between these quantities and the functions $C^A(\mathbf{k},\omega)$. Such relations are the sum rules and Kubo formulas[31] which have been studied in great detail for certain systems.[26]

(iii) *Phase transitions.* In a second-order phase transition, there is an operator, which we call Ψ, the order parameter, whose average value is zero for $T>T_c$, is nonzero for $T<T_c$, and approaches zero continuously as $T\to T_c^-$. The corresponding susceptibility χ^Ψ, which describes the response to an "external field,"[32] is infinite at $T=T_c$. Since

$$\chi^\Psi = (k_BT)^{-1}\int \hat{C}^\Psi(\mathbf{r})d^3r, \tag{2.6}$$

and $\hat{C}^\Psi(\mathbf{r})$ is finite for finite r, the singularity in χ^Ψ suggests that the range r_Ψ becomes infinite at T_c. When $\hat{C}^\Psi(\mathbf{r})$ decays with a power law as in Eq. (2.4) (with $p\leq 3$), the susceptibility χ^Ψ remains infinite for all $T<T_c$. The range r_Ψ is finite for $T<T_c$ but also diverges as $T\to T_c^-$. For finite external field H, it is assumed that no phase transition takes place; consequently the range r_Ψ remains finite for $T=T_c$, $H\neq 0$, and diverges as $H\to 0$. The range r_Ψ plays an important role in the theory of phase transitions. We shall generally call this length simply the "correlation length" and shall denote it by the symbol ξ.

III. SCALING HYPOTHESIS: QUALITATIVE PICTURE

The scaling hypothesis can be formulated in rather precise mathematical terms,[33] from which certain specific predictions follow rigorously, as will in part be shown in the next section. Since, on the one hand, this formulation is not altogether transparent and, on the other hand, it is no more than a *conjecture* based at best on heuristic arguments, it seems useful to present a simple qualitative picture of the main physical ideas involved. These ideas might in part survive if it turns out that the more precise formulation is not entirely correct.

A. Static Scaling

It is assumed that the order-parameter correlation length $r_\Psi\equiv\xi$, which diverges at $T=T_c$, $H=0$, contains the most important effects of critical fluctuations. In the domain of large r and ξ (compared to microscopic lengths), the correlation function $\hat{C}^\Psi(\mathbf{r})$ is assumed to depend critically on the ratio r/ξ, with quite different behavior for $r\ll\xi$ and $r\gg\xi$. The divergence of the susceptibility χ^Ψ is characterized by its dependence on ξ, which becomes infinite at T_c. The significance of the correlation length can best be illustrated by the graph in Fig. 1, where the wave number k is plotted on the ordinate and the inverse length ξ^{-1} on the abscissa. The origin $\xi^{-1}=0$ is the critical point $T=T_c$, and the disordered phase is on the right ($T>T_c$), whereas the ordered phase is on the left ($T<T_c$). (We restrict ourselves to $H=0$.) Three asymptotic regions may be

[30a] This is the definition employed by B. D. Josephson, Phys. Letters **21**, 608 (1966); J. A. Tyson and D. H. Douglass, Jr., Phys. Rev. Letters **17**, 472 (1966); J. W. Kane and L. P. Kadanoff, Phys. Rev. **155**, 80 (1967); and in Ref. 23.

[31] R. Kubo, *Lectures in Theoretical Physics* (Interscience Publishers, Inc., New York, 1959), Vol. I, Chap. 4.

[32] An external field is one which couples directly to the order parameter, such as a uniform magnetic field for a ferromagnet, or a "staggered" field for an antiferromagnet. For a fluid near its critical point the analogous quantity is roughly a pressure different from its critical value p_c. In superfluid helium the corresponding uniform Bose field is impossible to realize experimentally, but it plays the same formal role as a magnetic field in a spin system.

[33] See, e.g., R. B. Griffiths, Phys. Rev. **158**, 176 (1967).

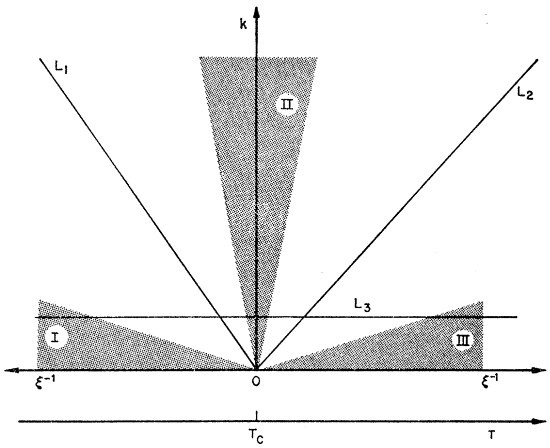

FIG. 1. The macroscopic domain of wave vector k and coherence length ξ, defined by the conditions $k\bar{a}\ll 1$, $\xi/\bar{a}\gg 1$. In the three shaded regions the correlation functions have different characteristic behaviors. These regions are defined by ($k\xi\ll 1$, $T<T_c$), ($k\xi\gg 1$, $T\approx T_c$), and ($k\xi\ll 1$, $T>T_c$), respectively. The asymptotic forms for these regions merge when extrapolated to the lines L_1 or L_2 ($k\xi=1$ for $T<T_c$ and $T>T_c$, respectively). An experiment done at constant k (line L_3) will pass through all three regions as the temperature is varied.

identified in the (k,ξ^{-1}) plane, in each of which the correlation function $C_T^{\Psi}(\mathbf{k})$ has different characteristic behavior.[34]

The shaded region marked I, corresponding to $k\xi\ll 1$, $T<T_c$, is the macroscopic region in the ordered phase; it refers to phenomena occurring over distances r large compared to ξ. Similarly there is a macroscopic region for $T>T_c$ denoted by III on Fig. 1. The region marked II, in which $k\xi\gg 1$ for either $T\geq T_c$ or $T\leq T_c$, is the so-called critical region,[35] which describes phenomena occurring over distances small compared to ξ, but large compared to all other relevant lengths. Since $\xi\to\infty$ as $T\to T_c$, region II is a "macroscopic" region, except insofar as critical fluctuations are concerned.

The function $C^{\Psi}(\mathbf{k})$ diverges at the origin ($k=0$, $\xi^{-1}=0$) and remains finite for finite k at $T=T_c$. The scaling hypothesis rests on the assumption that $C^{\Psi}(\mathbf{k})$ varies smoothly throughout the (k,ξ^{-1}) plane, except for the singularity at the origin. Furthermore, the function is assumed to be essentially determined by its limiting behavior in the three shaded asymptotic regions. Thus, if the forms valid in regions I and II are separately extrapolated to the line $L_1(k\xi=1, T<T_c)$, then the two resulting expressions must coincide, up to a possible factor of order unity. This means that there is no other dividing line between macroscopic and critical behavior than that provided by the length ξ. Moreover, a *single* function describes the correlations in the whole (k,ξ^{-1}) plane, with a characteristic dependence on the parameter $\xi/r\sim k\xi$.[36] The susceptibility $\chi_T^{\Psi}=(k_BT)^{-1}C_T^{\Psi}(0)$, defined as a function of temperature along the abscissa of Fig. 1, has a singularity at the *point* $\xi^{-1}=0$. For finite k, the correlation function $C_T^{\Psi}(\mathbf{k})$ passes smoothly (along L_3, say) from "below T_c" (region I), through a critical region whose size depends on k, to "above T_c" (region III) with no divergence.

For finite external field the situation is more complicated, but it is natural to assume that the above picture remains essentially unchanged, except that the length ξ is a function of both the field and $T-T_c$.

B. Dynamic Scaling

As mentioned earlier, the hydrodynamic analysis, which determines the form of $C^{\Psi}(\mathbf{k},\omega)$ for long wavelengths and low frequencies, is based on the concept of local thermodynamic equilibrium, which permits an analytic expansion of the currents of the conserved quantities in powers of $\mathbf{k}$.[37] At the critical point the length ξ becomes infinite, so that the range of applicability of hydrodynamics vanishes. This is because the long-range fluctuations of the order parameter destroy local equilibrium over increasingly large regions as T approaches T_c. On the other hand, from the definition of the correlation functions, Eqs. (2.1) and (2.2), we have the sum rule

$$C_T^{\Psi}(\mathbf{k})=\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}C_T^{\Psi}(\mathbf{k},\omega)\,. \tag{3.1}$$

It is natural to ask how the above-mentioned breakdown of hydrodynamics comes about, and in particular how the scaling hypothesis in the static function $C_T^{\Psi}(\mathbf{k})$ reflects itself in the dynamic function $C_T^{\Psi}(\mathbf{k},\omega)$, via the constraint (3.1).

In analogy with static scaling, the dynamic-scaling hypothesis rests on the assumption that the form of $C_T^{\Psi}(\mathbf{k},\omega)$ is essentially characterized by its behavior in the three limiting regions of Fig. 1. One consequence of this assumption is that if a certain long-wavelength mode dominates the sum rule (3.1) for T far from T_c, where hydrodynamics is surely valid (say, for $T<T_c$), then it will continue to do so for T arbitrarily close to T_c, so long as $k\xi\ll 1$. The frequency of the mode may of course depend on ξ, but it will still be related to thermodynamics in the way specified by the hydrodynamic analysis, up to corrections of order $k\xi$; these corrections

[34] In this section, the subscript T will sometimes be used to indicate the dependence of a quantity on $T-T_c$.

[35] The term "critical region" is generally used to denote the temperature interval in which critical phenomena dominate the behavior of the correlation functions, and corresponds to *all* portions of the (k,ξ^{-1}) plane depicted in Fig. 1, in which both k^{-1} and ξ are macroscopic lengths. We shall often refer to region II alone as the critical region, however, in order to contrast it with the hydrodynamic or macroscopic regions (I and III); in region II the system displays behavior which is characteristic of the critical temperature and is qualitatively different from the behavior "away from T_c." In practice it will be clear from the context whether we are using the term "critical region" in this restricted sense, or in its more usual general meaning.

[36] The precise meaning of these qualitative statements will be given in Sec. IV in terms of *homogeneous* functions.

[37] B. I. Halperin and P. C. Hohenberg (to be published).

may be made arbitrarily small (at fixed $T<T_c$) by going to long enough wavelengths. At fixed k, as T approaches T_c, the terms of order $k\xi$ become significant and modify the hydrodynamic behavior. When T reaches T_c (region II), ξ is infinite and $C_T^{\Psi}(k,\omega)$ depends on k in a way which is again largely determined by the scaling hypothesis, as shown in Sec. IV. When T is raised above T_c, the parameter $k\xi$ decreases and in the limit $k\xi\ll 1$ (region III) the system again follows hydrodynamics. The form of the hydrodynamic laws may of course be different in regions I and III.

IV. SCALING HYPOTHESIS: PRECISE FORMULATION

A. Static Scaling

We are concerned with the asymptotic form of correlation functions $\hat{C}_T^A(\mathbf{r})$ for large r, and T near T_c, which means that both r and ξ are large compared to all microscopic lengths.[38] In zero external field, the correlation length $\xi=r_\Psi$ is assumed to diverge at T_c as[39]

$$\begin{aligned}\xi&=\xi_0'\epsilon^{-\nu'}, \quad T<T_c\\ \xi&=\xi_0\epsilon^{-\nu}, \quad T>T_c\end{aligned} \tag{4.1}$$

where

$$\epsilon\equiv|T-T_c|/T_c. \tag{4.2}$$

According to the static scaling hypothesis, applied to the operator A, the function $\hat{C}_T^A(\mathbf{r})$ is a homogeneous function of r and ξ, which means it has the form

$$\begin{aligned}\hat{C}_T^A(\mathbf{r})&=r^x\hat{g}^+(r/\xi)\,, \quad T>T_c\\ \hat{C}_T^A(\mathbf{r})&=r^{x'}\hat{g}^-(r/\xi)\,, \quad T<T_c.\end{aligned} \tag{4.3}$$

It is assumed, furthermore, that for r finite there is no discontinuity at T_c, so that for any r

$$\lim_{T\to T_c^+}\hat{C}_T^A(\mathbf{r})=\lim_{T\to T_c^-}\hat{C}_T^A(\mathbf{r})\,, \tag{4.4}$$

and thus

$$\hat{g}^+(0)=\hat{g}^-(0)\,, \tag{4.5}$$

$$x=x'. \tag{4.6}$$

In Fourier transform we have

$$C_T^A(\mathbf{k})=k^y g^+(k\xi)\,, \quad T>T_c \tag{4.7a}$$

$$C_T^A(\mathbf{k})=k^y g^-(k\xi)\,, \quad T<T_c \tag{4.7b}$$

$$g^+(\infty)=g^-(\infty)\,, \tag{4.7c}$$

where $y=-(s+x)$, s being the dimensionality of the system. The functions $g^\pm$ and the exponent y of course depend on the operator A. The subscript T on the correlation function denotes its dependence on the parameter $T-T_c$, which may be considered to be a dependence on both ξ, and the sign of $T-T_c$. In many cases we shall suppress this last dependence and merely write $C_\xi^A(\mathbf{k})$, where it is understood that in general the correlation function may be different for $T<T_c$ and $T>T_c$. We shall similarly drop the superscript on the functions $g^\pm$ of Eq. (4.7), and merely state, whenever the distinction is necessary, whether we are referring to $T>T_c$ or $T<T_c$. In writing Eq. (4.7) we have assumed that for small k, the main contribution to the Fourier transform comes from the homogeneous (large r) part of $\hat{C}(\mathbf{r})$. This will be the case if $y<0$ (i.e., $x>-s$), and the remaining part gives a finite contribution. If $y=0$ there will be logarithmic terms in Eq. (4.7), in which case $C(\mathbf{k})$ cannot, strictly speaking, be considered a homogeneous function. In most of the following we shall ignore the possibility of such logarithms, although they explicitly occur in the two-dimensional Ising model (see Ref. 41 below), and in superfluid helium (Sec. V D).

We may find an immediate consequence of the static scaling assumption for $C_\xi^\Psi(\mathbf{k})$. Let us define critical exponents[2] γ and η such that for $T>T_c$,

$$C_\xi^\Psi(\mathbf{k}=0)=(k_BT)\chi^\Psi=C_0\epsilon^{-\gamma}=[C_0\xi_0^{-\gamma/\nu}]\xi^{\gamma/\nu} \tag{4.8}$$

and for $T=T_c$,

$$C_\infty^\Psi(\mathbf{k})=C_0'k^{-2+\eta}\,, \tag{4.9}$$

where C_0 and C_0' are constants. If $C_\xi^\Psi(\mathbf{k}=0)$ is finite for $\xi^{-1}\neq 0$, we must have, by Eq. (4.7a),

$$C_\xi^\Psi(\mathbf{k})\propto\xi^{-y}[1+\cdots]\,, \quad k\xi\ll 1\,, \quad \text{region III}. \tag{4.10}$$

Similarly, if $C_\infty^\Psi(\mathbf{k})$ is finite for $\mathbf{k}\neq 0$, we have

$$C_\xi^\Psi(\mathbf{k})\propto k^y[1+\cdots]\,, \quad k\xi\gg 1\,, \quad \text{region II}. \tag{4.11}$$

The dots in Eqs. (4.10) and (4.11) refer to higher-order terms in $k\xi$ and $(k\xi)^{-1}$, respectively. Comparing Eqs. (4.8)–(4.11), we find the *scaling law*

$$y=-2+\eta=-\gamma/\nu\,, \tag{4.12}$$

relating the exponents η and γ which characterize the order-parameter correlations in regions II and III, respectively.

For finite external field H, the correlation length remains finite even at $T=T_c$, and the structure of $C_{T,H}^\Psi(k)$ becomes more complicated. By making certain assumptions about this structure[9] it is possible to derive other scaling laws into which we shall not enter here.

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[38] As mentioned in Ref. 8, for systems possessing an intrinsic small parameter θ, there may be two different regions of critical behavior, $\epsilon\ll\theta$ and $\theta\ll\epsilon\ll 1$. The first inequality defines the true critical temperature interval, determined by the conditions $r\gg\bar{a}\bar{\theta}^{-1}$, $\xi\gg\bar{a}\bar{\theta}^{-1}$, where $\bar{a}$ will henceforth denote a general microscopic length, and $\bar{\theta}$ is a small parameter related to θ. The second inequality corresponds to the conditions $\bar{a}\ll r\ll\bar{a}\bar{\theta}^{-1}$, $\bar{a}\ll\xi\ll\bar{a}\bar{\theta}^{-1}$.

[39] Although most existing scaling laws (Refs. 9 and 11) include the symmetry relations $\gamma=\gamma'$, $\nu=\nu'$, and $\alpha=\alpha'$, we shall distinguish, at least in our notation, between the primed and unprimed indices. Stell [Phys. Rev. **173**, 314 (1968)] has recently proposed a variant of Widom's homogeneity assumptions, in which one may have $\gamma\neq\gamma'$ and $\nu\neq\nu'$.

B. Dynamic Scaling

A dynamic correlation function may in general be written in the form[30]

$$C_\xi^A(\mathbf{k},\omega)=2\pi[\omega_\xi^A(\mathbf{k})]^{-1}C_\xi^A(\mathbf{k})f_{\mathbf{k},\xi}^A(\omega/\omega_\xi^A(\mathbf{k}))\,, \quad (4.13)$$

where Eq. (3.1) implies that

$$\int_{-\infty}^{\infty} f_{\mathbf{k},\xi}^A(x)dx=1\,, \quad (4.14)$$

and where the *characteristic frequency* $\omega_\xi^A(\mathbf{k})$ is to be determined by the constraint

$$\int_{-1}^{1} f_{\mathbf{k},\xi}^A(x)dx=\tfrac{1}{2}\,. \quad (4.15)$$

It is easy to show that if the correlation function $C_\xi^A(\mathbf{k},\omega)$ consists of a pair of δ functions at $\omega=\pm\omega_1$, then the characteristic frequency will be $\omega_\xi^A(\mathbf{k})=\omega_1$. Similarly, if $C_\xi^A(\mathbf{k},\omega)$ is a Lorentzian of width Γ_1, centered about zero frequency, then $\omega_\xi^A(\mathbf{k})=\Gamma_1$. In many systems the spectrum of $C_\xi^A(\mathbf{k},\omega)$ in the hydrodynamic region consists of various transport modes with differing frequencies, strengths, and decay rates. In that case $\omega_\xi^A(\mathbf{k})$ will be some combination of the frequencies of the various modes; to the extend that the strength of one mode dominates, however, the characteristic frequency tends to the frequency of that particular mode. Examples of such behavior will be given below. Outside of the hydrodynamic domain, where the concept of a transport mode is not well defined, Eq. (4.15) provides a precise, if somewhat arbitrary, definition of the characteristic frequency for fluctuations of the operator A. This arbitrariness does not affect the critical exponents of $\omega_\xi^A(\mathbf{k})$.

The dynamic-scaling assumption is a generalization of Eqs. (4.7) to the frequency dependence of $C_\xi^A(\mathbf{k},\omega)$. In addition to Eqs. (4.7), we assume that $\omega_\xi^A(\mathbf{k})$ is a homogeneous function of k and ξ^{-1},

$$\omega_\xi^A(\mathbf{k})=k^z\Omega(k\xi)\,. \quad (4.16)$$

Furthermore we assume that the dimensionless function f^A, whose total weight and "frequency spread" are determined by Eqs. (4.14) and (4.15), depends only on the product $k\xi$ and not on $\mathbf{k}$ and ξ separately,[40]

$$f_{\mathbf{k},\xi}^A(x)=f_{k\xi}^A(x)\,. \quad (4.17)$$

Let us note an immediate consequence of the above scaling assumptions, which may be tested experimentally whenever the function $C_\xi^A(\mathbf{k},\omega)$ can be measured, as for instance in a scattering experiment. It follows from Eq. (4.17) that if the frequency dependence of $C_\xi^A(\mathbf{k},\omega)$ is measured for different values of k and $T-T_c$, then, apart from a change of scale determined by Eqs. (4.7) and (4.16), the shape of $C_\xi^A(\mathbf{k},\omega)$ will be the same along any straight line through the origin in the (k,ξ^{-1}) plane (Fig. 1).

There exists another formulation of the scaling hypothesis which is somewhat weaker, but leads to many of the same results. Let us define the asymptotic forms of $C_\xi(\mathbf{k})$ in the different regions. We have $C^{\mathrm{II}}(\mathbf{k})\equiv C_{\xi=\infty}(\mathbf{k})$ while $C_\xi^{\mathrm{I}}(\mathbf{k})$ and $C_\xi^{\mathrm{III}}(\mathbf{k})$ are the asymptotic forms of $C_\xi(\mathbf{k})$ as $k\rightarrow 0$ for fixed ξ with $T<T_c$ and $T>T_c$, respectively. The scaling hypothesis is a matching condition when the asymptotic forms are extrapolated to $k\xi=1$:

$$C_{\xi=k^{-1}}^{\mathrm{III}}(\mathbf{k})=a'C^{\mathrm{II}}(\mathbf{k})\,, \quad (4.7a')$$

$$C_{\xi=k^{-1}}^{\mathrm{I}}(\mathbf{k})=aC^{\mathrm{II}}(\mathbf{k})\,, \quad (4.7b')$$

where a and a' are numerical constants of order unity. Similar expressions may be assumed for the characteristic frequency $\omega_\xi(\mathbf{k})$ and for the shape function $f_{k,\xi}(x)$. This weaker formulation, which is the one adopted in Ref. 23, has the advantage that it is applicable in cases where the functions contain logarithmic factors.

C. Restricted and Extended Scaling

If the dynamic-scaling assumptions [Eqs. (4.16) and (4.17)] are ever valid, they ought to hold for the case where the operator A is the order parameter Ψ. This operator is the one whose behavior is most singular at the phase transition and whose fluctuations are most intimately connected with the nature of the critical point. The assumption that the *order-parameter* correlation function obeys the dynamic-scaling laws at the critical point will be referred to as the "restricted dynamic-scaling assumption," and is the minimal assumption that we make. It also seems likely that the dynamic-scaling assumptions will hold for various other operators A, as, for example, the static scaling law has been verified to apply to the energy correlation function in the two-dimensional Ising model.[41] When applied to operators other than the order parameter, Eqs. (4.16) and (4.17) will be considered as extensions of the dynamic-scaling hypothesis. It is entirely possible that if restricted scaling holds, certain extensions will be valid but others will not, or even that no extensions are possible. Although a general discussion of the hierarchy of possible extensions is not particularly instructive, many of them may be tested in specific cases, and are therefore worth identifying. Roughly speaking, an extension of dynamic scaling to some operator $A\neq\Psi$ will become less likely the weaker the singularity in the static susceptibility χ_ξ^A. On the other hand, if the characteristic frequency $\omega_\xi^A(\mathbf{k})$ is the same as $\omega_\xi^\Psi(k)$ in some region, then an extension to the operator

[40] Once again, we remind the reader that in all the expressions of this section the subscript ξ also implies an additional index f referring to the sign of $T-T_c$. Indeed, the functions $\Omega(k\xi)$ and $_{k\xi}$ may be different functions ($\Omega_\pm$ and $f^\pm$) of $k\xi$ for $T>T_c$ and $T<T_c$; by continuity at $T=T_c$ for $\omega\neq0$, $k\neq0$, we must, however, have $\Omega_+(\infty)=\Omega_-(\infty)$, $f_\infty^+=f_\infty^-$, which implies the same exponent z in Eq. (4.16), for $T>T_c$ and $T<T_c$.

[41] R. Hecht, Phys. Rev. 158, 557 (1967).

A might hold even if χ_ξ^A is only weakly singular. Alternatively, if dynamic scaling fails for some function $C_\xi^A(\mathbf{k},\omega)$ it is likely that the static function $C_\xi^A(\mathbf{k})$ will also fail to obey static scaling laws.

It must be stressed that we are not attempting to *predict* which one of the large number of distinguishable extensions will in fact hold. We wish merely to point out that many of them may be tested experimentally in specific systems. We shall only consider extensions to "legitimate" microscopic[29] operators which retain their meaning outside of the thermodynamic and hydrodynamic domains. In particular, we restrict ourselves to operators which may be expressed as combinations of field operators with constant (e.g., temperature-independent) coefficients.

V. APPLICATIONS TO SPECIFIC SYSTEMS

A. The Antiferromagnet

1. *Hydrodynamics*

To explore the consequences of the dynamic-scaling assumption for any particular system it is important to analyze the hydrodynamics in order to calculate the corresponding order-parameter correlation function $C_\xi^\Psi(\mathbf{k},\omega)$. For the "isotropic" antiferromagnet,[42] such an analysis has been carried out,[37] in analogy with the derivation of two-fluid hydrodynamics for superfluid helium. We shall summarize the main results of this analysis.

The order parameter for the isotropic antiferromagnet is the staggered magnetization $\mathbf{N}$,[30,43] which is a vector quantity; the correlation function analogous to (2.1) must be defined by

$$\hat{C}^{\mathbf{N}}(\mathbf{r},t) \equiv \tfrac{1}{2}\sum_i \langle\{(N_i(\mathbf{r},t)-\langle N_i(r,t)\rangle),\ (N_i(0,0)-\langle N_i(0,0)\rangle)\}\rangle = \hat{C}^{N_x}(\mathbf{r},t)+\hat{C}^{N_y}(\mathbf{r},t)+\hat{C}^{N_z}(\mathbf{r},t), \quad (5.1)$$

where the summation is over the three coordinate directions, so that $\hat{C}^{\mathbf{N}}$ is the expectation value of the symmetrized scalar product of the fluctuations in $\mathbf{N}$.

Static correlation functions. For $T>T_N$, the system is isotropic and $\hat{C}^{\mathbf{N}}=3\hat{C}^{N_z}$. Moreover, the function $\hat{C}^{\mathbf{N}}(\mathbf{r})$ decays exponentially at large r, with a range satisfying the usual relations (4.1).

For $T<T_N$, let us suppose the order parameter at equilibrium to be lined up in the z direction uniformly over the whole system. Then we have

$$\hat{C}^{\mathbf{N}}(\mathbf{r},t)=\hat{C}^{N_z}(\mathbf{r},t)+2\hat{C}^{N_x}(\mathbf{r},t), \quad (5.2)$$

which expresses $C^{\mathbf{N}}$ as the sum of a parallel and a perpendicular correlation function. At large r the perpendicular function $\hat{C}^{N_x}(\mathbf{r})$ dominates,[44] and falls off as r^{-1} [as in Eq. (2.4) with $p=1$]. In Fourier transform this means that for small k,

$$C^{\mathbf{N}}(\mathbf{k})=bk^{-2}+O(k^{-1})\approx 2C^{N_x}(\mathbf{k}), \quad (5.3)$$

where the constant b may be identified in terms of Eq. (2.4) as $b=4\pi\langle N_z\rangle^2\xi$.

The quantity

$$\rho_s\equiv 2k_BTb^{-1}\langle N_z\rangle^2 \propto \xi^{-1} \quad (5.4)$$

plays the role of a "stiffness constant" for fluctuations in the direction of $\mathbf{N}$, the notation being intended to suggest the analogy with helium II, where the superfluid density ρ_s is the stiffness constant for phase fluctuations.

The *total* spin[43] $\mathbf{M}$ can also be separated into parallel and perpendicular correlation functions,

$$C^{\mathbf{M}}(\mathbf{k},\omega)=C^{M_z}(\mathbf{k},\omega)+2C^{M_x}(\mathbf{k},\omega). \quad (5.5)$$

The susceptibility

$$\chi^{\mathbf{M}}=(k_BT)^{-1}C^{\mathbf{M}}(\mathbf{k}=0) \quad (5.6)$$

remains finite at all temperatures. Below T_c, the perpendicular susceptibility χ^{M_x} is larger than χ^{M_z}, but the latter is also believed to be nonzero.

Dynamic correlation functions. The main result of the hydrodynamic analysis[37] is that, for any $T<T_N$, spin waves exist as well-defined excitations and exhaust the function $C_\xi^{\mathbf{N}}(\mathbf{k},\omega)$ in the long-wavelength limit. The spin-wave frequency is given by

$$\omega(\mathbf{k})=ck, \quad (5.7)$$

$$c^2=\rho_s/\chi^{M_x}, \quad (5.8)$$

and the damping by

$$\Gamma(\mathbf{k})=\tfrac{1}{2}D_sk^2, \quad (5.9)$$

where D_s is some (unknown) function of temperature. The above result, of course, depends crucially on the isotropy of the starting Hamiltonian.

It follows from Eqs. (5.4), (5.7), and (5.8) that the characteristic frequency for $\mathbf{N}$ is

$$\omega_\xi^{\mathbf{N}}(\mathbf{k})=ck\propto \xi^{-1/2}k. \quad (5.10)$$

Furthermore, since the spin waves exhaust the sum rule, the function $f^{\mathbf{N}}$ is, to lowest order in k, a pair of δ functions

$$f_0^{\mathbf{N}}(x)=\tfrac{1}{2}[\delta(x-1)+\delta(x+1)], \quad (5.11)$$

and to order k it is a two-peaked function

$$f_{k,\xi}^{\mathbf{N}}(x)=\frac{1}{\pi}\frac{\eta_k}{(x^2-1)^2+\eta_k^2}, \quad (5.12)$$

[42] An isotropic magnetic system is one whose Hamiltonian is invariant under rotation of the axis of spin quantization.

[43] We measure magnetization in units of spin angular momentum per unit volume. If $\mathbf{A}$ and $\mathbf{B}$ denote the magnetizations on the two sublattices, then the staggered magnetization is $\mathbf{N}\equiv\mathbf{A}-\mathbf{B}$ while the total magnetization is $\mathbf{M}=\mathbf{A}+\mathbf{B}$.

[44] The asymptotic behavior of the parallel function $\hat{C}^{N_z}(\mathbf{r})$ is not known precisely; the spin-wave approximation predicts $\hat{C}^{N_z}(\mathbf{r})\sim r^{-2}$ for the ferromagnet and is thought to be the same for the antiferromagnet. See K. Kawasaki and H. Mori, Progr. Theoret Phys. (Kyoto) **25**, 1043 (1961); **38**, 1052 (1967); Vaks *et al.*, Ref. 51, below; for the antiferromagnet see K. Tani and H. Tanaka, Phys. Letters **26A**, 68 (1967).

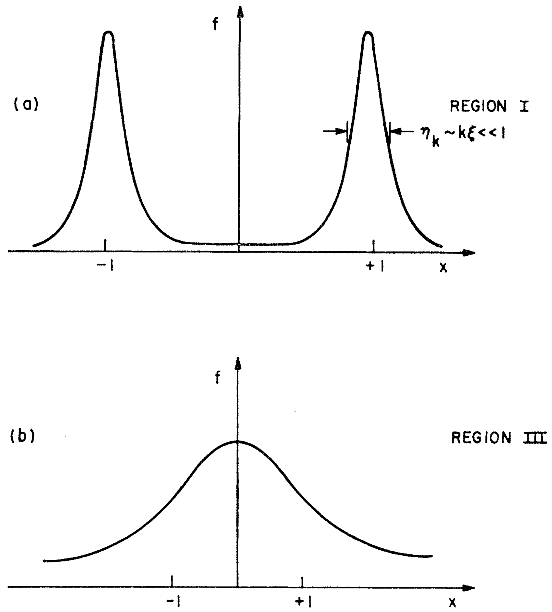

FIG. 2. Schematic representation of the shape of the frequency spectrum for order-parameter fluctuations at long wavelengths in an antiferromagnet. The function $f^{\mathbf{N}}(x)$ is depicted in (a) the spin-wave region $T<T_N$, and (b) the paramagnetic region $T>T_N$. For the ferromagnet, the function $f^{\mathbf{M}}$ has similar behavior, except that in region I the width parameter η_k is roughly proportional to $(k\xi)^2$.

where

$$\eta_k = D_s k^2/ck \ll 1. \tag{5.13}$$

For $T>T_N$, there is no propagating mode at long wavelengths. The function $C^{\mathbf{N}}(\mathbf{k},\omega)$ is peaked about $\omega=0$. Its width $\Gamma^{\mathbf{N}}(\mathbf{k})$ has a nonzero limit $\Gamma^{\mathbf{N}}(\mathbf{k}=0)$, because $\mathbf{N}$ is not a constant of the motion. From the definition of the characteristic frequency it is clear that for $T>T_N$ and $k\to 0$ (region III)

$$\omega^{\mathbf{N}}(\mathbf{k}) = \Gamma^{\mathbf{N}}(\mathbf{k}). \tag{5.14}$$

Turning now to the total magnetization $\mathbf{M}$, the characteristic frequency for transverse fluctuations $\omega^{M_x}(\mathbf{k})$ is identical to the spin-wave frequency (5.7) for $T<T_N$ (region I), and the damping is also described by Eq. (5.9). For $T>T_N$, on the other hand, the relaxation rate $\Gamma^{M_x}(\mathbf{k})$ has the diffusion form

$$\omega^{M_x}(\mathbf{k}) = \Gamma^{M_x}(\mathbf{k}) \approx Dk^2 \tag{5.15}$$

because $\mathbf{M}$ is a constant of the motion. The frequency $\omega^{M_z}(\mathbf{k})$, describing longitudinal fluctuations, is not known precisely for $T<T_N$ but we shall assume it to be also of the diffusion form

$$\omega^{M_z}(\mathbf{k}) = D_z k^2. \tag{5.16}$$

Above T_N, of course, $\omega^{M_z}(\mathbf{k})$ is identical to $\omega^{M_x}(\mathbf{k})$.

2. Scaling Predictions

Restricted dynamic scaling. The characteristic frequency $\omega_\xi^{\mathbf{N}}(k)$ is assumed to have the general form given in Eq. (4.16) and to reduce to (5.10) in region I. This implies that the exponent z of Eq. (4.16) is $\frac{3}{2}$. Consequently, in region II we have

$$\omega^{\mathbf{N}}(\mathbf{k}) = Bk^{3/2} + \cdots = k^{3/2}\Omega(k\xi), \quad k\xi \gg 1. \tag{5.17}$$

Because $\omega_\xi^{\mathbf{N}}(\mathbf{k}=0) = \Gamma_\xi^{\mathbf{N}}(0)$ is nonzero in region III, we find

$$\omega_\xi^{\mathbf{N}}(\mathbf{k}) = B'\xi^{-3/2} + \cdots \tag{5.18}$$

for $k\xi \ll 1$ and $T>T_c$. In the above equations B and B' are constants and the dots represent corrections which are higher order in $(k\xi)^{-1}$ and $k\xi$, in regions II and III, respectively.

From assumption (4.17) on the form of $f^{\mathbf{N}}(x)$, it follows that the damping correction $\eta_k = (D_s k^2/ck)$ of Eq. (5.13) cannot depend on any other parameter than $k\xi$, and since it is linear in k it must be of the form

$$\eta_k = B'' k\xi. \tag{5.19}$$

It follows that

$$D_s \propto \xi^{1/2}. \tag{5.20}$$

The shape function $f_{k\xi}^{\mathbf{N}}(x)$ has the spin-wave and relaxation forms depicted in Figs. 2(a) and 2(b) in the hydrodynamic regions I and III. In region II, the scaling hypotheses do not specify the shape function; examples of possible behavior are shown in Fig. 3.

The above predictions also follow from the "asymptotic matching conditions" for the frequency and shape function, analogous to Eqs. (4.7′). The frequency $\omega_\xi^{\mathbf{N}}(\mathbf{k})$ has the asymptotic forms $\omega_\xi^{\mathrm{I}}(\mathbf{k}) \propto \xi^{-1/2}k$ [Eq. (5.10)] and $\omega_\xi^{\mathrm{III}}(\mathbf{k}) \propto \xi^{-z}$. It follows that $\omega^{\mathrm{II}}(k) \propto \omega_{k^{-1}}^{\mathrm{I}}(\mathbf{k}) \propto k^{3/2}$, and $\omega_\xi^{\mathrm{III}}(\mathbf{k}) \propto \xi^{-3/2}$. Similarly, the damping frequency is $\Gamma(\mathbf{k}) = \frac{1}{2}D_s k^2$, which extrapolates to $k^{3/2}$ if $D_s \propto \xi^{1/2}$. A similar argument on the scaling of frequencies was first advanced by Ferrell *et al.*[23] for the λ transition of superfluid helium.

As pointed out by Heller,[45] the correlation function $C_\xi^{\mathbf{N}}(\mathbf{k},\omega)$ determines the linewidth for NMR at temperatures near T_N. For $T>T_N$, the NMR linewidth is given by

$$\Delta \propto A^2 \int_0^\infty dt\, \hat{C}_\xi^{\mathbf{N}}(\mathbf{r}=0,t) \tag{5.21}$$

$$\propto A^2 \int d^3k\, C_\xi^{\mathbf{N}}(\mathbf{k},\omega=0), \tag{5.22}$$

where A is the hyperfine coupling constant.

Inserting the scaling form [(4.7), (4.13), (4.16), and (4.17)] into Eq. (5.22), we find

$$\Delta \propto \int d^3k \frac{C_\xi(\mathbf{k})}{\omega_\xi(\mathbf{k})} f_{k\xi}(0),$$

$$\Delta \propto \int_0^\infty k^2 dk \frac{k^{\eta-2} g(k\xi) f_{k\xi}(0)}{k^{3/2}\Omega(k\xi)}, \tag{5.23}$$

$$\Delta \propto \xi^{\frac{1}{2}-\eta} \int_0^\infty d(k\xi) F(k\xi) \propto \xi^{\frac{1}{2}-\eta},$$

[45] P. Heller, Natl. Bur. Std. (U. S.) Misc. Publ. 273, 58 (1966).

where Eqs. (4.12) and (5.17) have been used. The integral in Eq. (5.23) is found to converge, with the major contribution coming from $k\xi$ of order 1. The NMR linewidth of the isotropic antiferromagnet is thus predicted to diverge for $T>T_N$, as $\epsilon^{-\nu(\frac{1}{2}-\eta)}\approx\epsilon^{-1/3}$; this divergence is slightly weaker than the $\epsilon^{-1/2}$ behavior predicted by Heller[45] on the basis of the Van Hove[14] theory. Similar results are obtained for the NMR linewidth below T_c.

Extended dynamic scaling. The above predictions all refer to the time dependence of the order parameter **N**, and therefore follow from the restricted dynamic-scaling hypothesis. We may also discuss the extension to the total magnetization **M**, whose hydrodynamic behavior is known. This analysis is not of immediate practical interest, since $C^{\mathbf{M}}(\mathbf{k},\omega)$ is difficult to measure, but we present it here as an illustration of extended dynamic scaling.

As mentioned above, for $T<T_N$ (region I) the frequency $\omega^{M_x}(\mathbf{k})$ and the shape function f^{M_x} coincide with those of the order parameter, Eqs. (5.7)–(5.9) and (5.12), whereas above T_N (region III) the total spin satisfies a diffusion equation with a frequency given by Eq. (5.15). The susceptibility χ^{M_x} is believed to remain finite and continuous at T_N, even though its temperature derivative probably has some singularity. The damping of the spin waves of $C_\xi{}^{M_x}(\mathbf{k},\omega)$ in region I follows from the *restricted* scaling hypothesis, and is given by (5.20), since f^{M_x} and $f^{\mathbf{N}}$ agree to order $(k\xi)^2$. The spin-diffusion coefficient in region III, on the other hand [Eq. (5.15)], is only determined by the extended hypothesis as

$$D\propto\xi^{1/2}. \tag{5.24}$$

This divergence in D corresponds to a "critical speeding up" of (total) spin fluctuations at fixed k, as $T\rightarrow T_N{}^+$.[46] In region II, the frequency ω^{M_x} is given by Eq. (4.16), with $z=\frac{3}{2}$, this result being again a consequence of extended rather than restricted scaling. The characteristic frequency for longitudinal total spin fluctuations $\omega^{M_z}(\mathbf{k})$ is identical to $\omega^{M_x}(\mathbf{k})$ in regions II and III, and should be proportional to $\xi^{1/2}k^2$ in region I.

3. Experimental Tests

The antiferromagnet is a very favorable system for testing the dynamic-scaling hypothesis, since the order-parameter correlation function is directly measurable by neutron-scattering experiments. Furthermore there exists at least one material, $RbMnF_3$, which seems to conform quite closely to the isotropic Heisenberg model.[47] In principle, one test of the applicability of this model is the verification of the spin-wave dispersion relation Eqs. (5.7) and (5.10), both its k dependence at

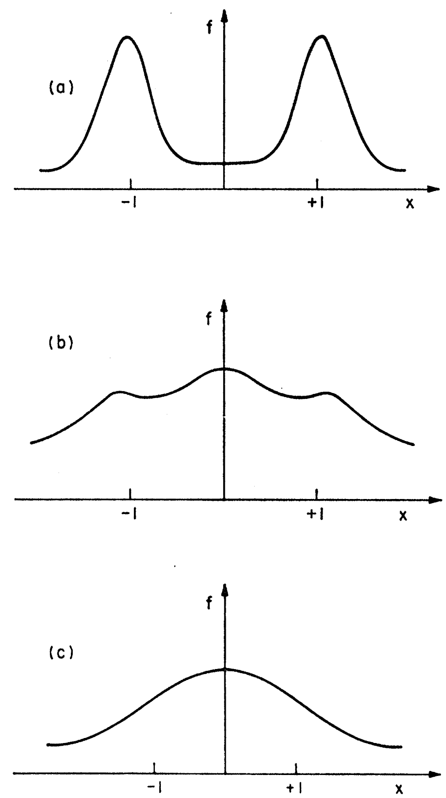

FIG. 3. Some possible forms for the shape function $f^{\mathbf{N}}(x)$ in the antiferromagnet or $f^{\mathbf{M}}(x)$ in the ferromagnet at T_c (region II), which are all consistent with scaling. Only the shape shown in (a) can be interpreted in terms of propagating spin waves.

low temperatures and its ξ dependence near T_N. If the model turns out to be applicable far from T_N, then it is reasonable to test the scaling predictions which apply near T_N.

Static correlations. The static function can be determined from the quasi-elastic scattering cross section[48]

$$d\sigma/d\Omega\propto C^{\mathbf{N}}(\mathbf{k})+(2\pi)^3|\langle\mathbf{N}\rangle|^2\delta(\mathbf{k}), \tag{5.25}$$

for scattering at wave vectors $\mathbf{q}=\mathbf{K}+\mathbf{k}$ near the magnetic lattice vector $\mathbf{K}$. For $T>T_N$, and $k\rightarrow 0$, we thus expect a cross section of the form

$$d\sigma/d\Omega=A'/(k^2+\kappa^2), \tag{5.26}$$

from which the correlation length

$$\xi=\kappa^{-1} \tag{5.27}$$

can be determined (A' is a constant if $\eta=0$). For $T<T_N$ the "Bragg" peak proportional to the order parameter $|\langle\mathbf{N}\rangle|^2$ appears at $\mathbf{k}\equiv\mathbf{q}-\mathbf{K}=0$, and from our definition of the correlation length, Eq. (2.4), it follows that

$$d\sigma/d\Omega\propto|\langle\mathbf{N}\rangle|^2[(2\pi)^3\delta(\mathbf{k})+4\pi\xi/k^2+\cdots], \tag{5.28}$$

[46] The kinematic slowing down still exists since $\omega^{\mathbf{M}}(\mathbf{k})\propto k^2$ for $k\xi\ll1$.

[47] D. T. Teany, M. J. Freiser, and R. W. H. Stevenson, Phys. Rev. Letters **9**, 212 (1962); R. Nathans, F. Menzinger, and S. J. Pickart, J. Appl. Phys. **39**, 1237 (1968).

[48] We assume that below T_c the crystal is made up of a number of domains of random spin orientation, so that the scattering cross section represents an average over the x, y, and z directions relative to the alignment of the order parameter.

where the dots refer to terms of higher order in k. In principle, the quasi-elastic experiments might also be used to verify the more specific scaling form (4.7) postulated for $C_\xi^{\mathbf{N}}(\mathbf{k})$.

Dynamic correlations: region II. Having determined the correlation length ξ, it is possible to test the dynamic-scaling prediction for the k dependence of the characteristic frequency. In practice, region II is the easiest to study with neutrons, since it is relatively easy to fix the temperature at T_N and to make measurements at different scattering angles. In particular, in $RbMnF_3$, where $T_N \approx 82°K$, we expect ξ to be several hundred angstroms when T is within 10 mdeg of T_N. The interesting range of k values lies between the minimum practical with neutrons (roughly 0.02 Å^{-1}) and a maximum k_m above which the macroscopic theory is invalid. We estimate that $k_m = \frac{1}{10}\pi\bar{a}^{-1} \approx 0.1$ Å^{-1}, where $\bar{a}$ denotes a microscopic length, such as the lattice spacing or the force range. The observable part of region II corresponds roughly to a decade of k values, $\xi^{-1} < k < k_m$. The experiments of Nathans, Menzinger, and Pickart[47] are consistent with a $k^{3/2}$ dependence of the characteristic frequency in region II.

Dynamic correlations: hydrodynamic regions. In order to test the assertions referring to regions I and III, namely, the spin-wave damping, Eq. (5.20), and the spin relaxation, Eq. (5.18), it is necessary to go to very small k where the accuracy of the neutron experiments decreases, since the resolution corrections which must be applied to the observed cross sections become large. In order to be in a "critical" region it is necessary that ξ be larger than $\bar{a}$ (say, $\xi \gtrsim 10\bar{a}/\pi$), which means roughly $\epsilon < 0.1$, or $82°K < T < 90°K$ in $RbMnF_3$, for $T \geq T_c$. Thus the interesting hydrodynamic regions are restricted to scattering angles for which $k \ll \xi^{-1} \lesssim k_m$, where experiments are difficult. One method of meeting these difficulties is to assume a form for $\omega_\xi^{\mathbf{N}}(\mathbf{k})$ containing a few adjustable parameters, and satisfying the scaling hypothesis (4.16). The parameters are then fitted from the data for $k\xi > \frac{1}{2}$, say, and the function extrapolated to the region $k\xi < \frac{1}{2}$.[49] The resulting expression may be convoluted with the resolution function and compared to the measured cross section. This procedure would provide a consistency check rather than an absolute measurement, but is probably a more realistic test of Eqs. (5.18) and (5.20) in the hydrodynamic regions.

[49] For example, take

$$\omega_\xi^{\mathbf{N}}(\mathbf{k}) = (Ak^4 - Bk^3\xi^{-1} + Ck^2\xi^{-2} + D\xi^{-4})^{3/8} \quad \text{for} \quad T > T_N. \quad (1)$$

This has the asymptotic forms

$$\omega_\xi^{\mathbf{N}}(\mathbf{k}) = A^{3/8}k^{3/2} - \tfrac{3}{8}BA^{-5/8}k^{1/2}\xi^{-1} + \cdots \quad \text{for} \quad k\xi \gg 1, \quad (2)$$

$$\omega_\xi^{\mathbf{N}}(\mathbf{k}) = D^{3/8}\xi^{-3/2} + \tfrac{3}{8}CD^{-5/8}k^2\xi^{1/2} + \cdots \quad \text{for} \quad k\xi \ll 1. \quad (3)$$

The parameter A may be fitted from data at $T = T_N$, 0.05 Å$^{-1} \leq k \leq 0.10$ Å^{-1} using the first term of Eq. (2). To determine C and D use data at fixed $T > T_N$, with $k\xi < 1$, say, at $T = 88°K$, for $0 \leq k \leq 0.1$ Å^{-1}, using Eq. (3). Then B may be found by adjusting the minimum in $\omega_\xi^{\mathbf{N}}(\mathbf{k})$ as a function of T for fixed $k > 0.05$ Å^{-1}. Once a consistent set of constants A, B, C, D is found, the expression for $\omega_\xi^{\mathbf{N}}(k)$ can be convoluted with the resolution function in order to predict the experimental temperature dependence of $\omega_\xi^{\mathbf{N}}(0)$.

The prediction for the NMR linewidth, Eq. (5.23), may be verified on $RnMnF_3$, by an experiment similar to Heller's[45] measurements on the anisotropic material MnF_2.

4. *Effect of Anisotropy*

The presence of anisotropy will have a marked effect on the dynamic properties of the antiferromagnet. Let us first consider the case of a strongly anisotropic model which has cubic symmetry above T_N.[50] The correlation function $C^{\mathbf{N}}(\mathbf{k},\omega)$ is still equal to $3C^{N_z}(\mathbf{k},\omega)$ for $T \geq T_N$, and the characteristic relaxation rate $\omega_\xi^{\mathbf{N}}(\mathbf{k})$ approaches a finite limit $\omega_\xi^{\mathbf{N}}(0) \propto \xi^{-z}$ as $k \to 0$, for fixed $T > T_N$. Below T_N, we must distinguish between fluctuations in $\mathbf{N}$ which are parallel and those which are perpendicular to the average staggered magnetization $\langle \mathbf{N} \rangle$. As contrasted with the isotropic case, the perpendicular fluctuations do not now diverge at $k=0$, and do not necessarily dominate $C^{\mathbf{N}}(\mathbf{k})$ at long wavelengths. Let us again suppose $\langle \mathbf{N} \rangle$ is in the z direction. The correlation functions $C^{N_x}(\mathbf{k},\omega)$ and $C^{N_y}(\mathbf{k},\omega)$ are dominated by well-defined spin-wave modes at low temperatures, and it seems likely that these modes exist at long wavelengths for any temperature below T_N. Unlike the isotropic case, however, there is a gap in the spin-wave spectrum. The value of the spin-wave frequency at long wavelengths is not exactly relatable to thermodynamic properties of the system. As $T \to T_N$, the spin-wave gap is believed to approach zero; if dynamic scaling is obeyed, the gap must approach zero with the same power of ξ as the relaxation rate above T_N, namely, ξ^{-z}. The parallel component N_z is believed to have a finite relaxation rate at $k=0$ below T_c as well as above T_c. According to the restricted dynamic-scaling hypothesis, we must have $\omega_\xi^{N_z}(\mathbf{k}) \propto \xi^{-z}$ in region I. In region II, of course, we should have $\omega^{\mathbf{N}}(\mathbf{k}) \propto k^z$.

For the anisotropic case we cannot determine the exponent z from thermodynamic considerations. Nonetheless, an experimental test of the scaling hypotheses should be feasible. The gap in the spin-wave spectrum in region I can be determined quite accurately by an antiferromagnetic resonance experiment. The dispersion relation in region II can be measured by neutron scattering, and the exponents may be compared for the two regions. The relaxation rates in regions I and III may also be accessible to neutrons.

Let us next consider a strongly anisotropic model which does not have cubic symmetry above T_c. Here, the order parameter is the scalar quantity N_z. The predictions of the restricted dynamic-scaling hypothesis for ω^{N_z} are the same as for the cubic case: a relaxation frequency proportional to ξ^{-z} in regions I and III and a characteristic frequency Bk^z in region II, where z is an unknown exponent. In the present case, however,

[50] The anisotropy might result from a spin-spin interaction such as a dipole-dipole interaction, which is not invariant under rotation of the spins, or it might result from the combined effect of spin-orbit coupling and crystal-field splitting on the individual atomic levels.

restricted dynamic scaling can say nothing about fluctuations in N_x and N_y. Unlike $C^{N_z}(k)$, the static correlation functions $C^{N_x}(\mathbf{k})$ and $C^{N_y}(\mathbf{k})$ either do not diverge, or diverge only weakly as $k\to 0$ and $T\to T_N$. Presumably ω^{N_x} will also have a different behavior than ω^{N_z} near the critical point.

A weakly anisotropic antiferromagnet will have an energy gap at low temperatures proportional to $(H_A H_{\mathrm{ex}})^{1/2}\equiv H_{\mathrm{ex}}\delta$, where H_A is the "anisotropy field" and H_{ex} is the exchange field (we assume $\delta\ll 1$). The spin-wave spectrum at low temperatures is linear for $\delta\ll k\bar{a}\ll 1$, and deviates from linearity for $k\bar{a}\lesssim\delta$. The existence of the small parameter δ raises the possibility of two different temperature regions in which critical behavior occurs.[8,38] We expect to find a region $\theta\ll\epsilon\ll 1$ or $\bar{\theta}\bar{a}^{-1}\ll\xi^{-1}\ll\bar{a}^{-1}$, in which the system behaves like an isotropic antiferromagnet, for k values in the "quasi-macroscopic" range $\bar{\theta}\bar{a}^{-1}\ll k\ll a^{-1}$. The small parameter θ is proportional to an unknown power of δ, while $\bar{\theta}$ is proportional to $\theta^{2/3}$. We may speculate that the true critical behavior is reached when the conditions $\xi^{-1}\ll\bar{\theta}\bar{a}^{-1}$, $k\ll\bar{\theta}\bar{a}^{-1}$ are satisfied. Here the system behaves like an anisotropic model, with a gap in the long-wavelength spin-wave spectrum. This change of behavior could be interpreted, in terms of the scaling picture, as a dependence of the characteristic frequency on the parameter $k\bar{a}\bar{\theta}^{-1}$, rather than $k\xi$, and thus as a violation of dynamic scaling. In fact, it merely results from the existence of an anomalously long "microscopic" (i.e., temperature-independent) length $\bar{\theta}^{-1}\bar{a}$, which is inherent in the model. The asymptotic critical behavior is not reached until the relation $\xi^{-1}\ll\bar{\theta}\bar{a}^{-1}$ is satisfied, even though most thermodynamic functions may be insensitive to the parameter $\bar{\theta}\xi/\bar{a}$.

B. Ferromagnet

1. Hydrodynamics

As is well known, isotropic ferromagnets (Heisenberg or itinerant electron model) have spin waves at low temperatures with a dispersion relation proportional to k^2, for small k. Just as for the isotropic antiferromagnet, it is believed that at any fixed temperature below T_c the spin-wave damping rate is negligible compared to the real part of the frequency in the long-wavelength limit. Indeed, the microscopic theory of the Heisenberg model predicts a spin-wave decay rate proportional to $k^4(\ln k)^2$ in an appropriate low-temperature and long-wavelength limit.[51]

The purely macroscopic spin-wave theory,[37] which may not be as well founded for the ferromagnet as it is for the antiferromagnet, predicts a decay rate proportional to k^4 in the long-wavelength limit at all temperatures below T_c. In any case, the spin-wave frequency is given by the usual Landau-Lifshitz formula[52]

$$\omega(\mathbf{k})=\lambda k^2\,, \tag{5.29}$$

where

$$\lambda=\rho_s/|\langle\mathbf{M}\rangle|\,, \tag{5.30}$$

and the stiffness constant ρ_s is related to the static correlation function $C_\xi{}^{\mathbf{M}}(\mathbf{k})$ by equations analogous to (5.3) and (5.4). Neglecting the possible logarithmic factor, we find that the function $f^{\mathbf{M}}$ has the form (5.12) in region I, with a width parameter

$$\eta_k=(\zeta k^4/\lambda k^2)\,, \tag{5.31}$$

which depends on the "spin-wave damping coefficient" ζ.

For $T>T_c$, the hydrodynamic form of $C_\xi{}^{\mathbf{M}}(\mathbf{k},\omega)$ is the same for the ferromagnet as for the antiferromagnet [Eq. (5.15)], since both are in the paramagnetic phase. The susceptibility $\chi^{\mathbf{M}}$ diverges at T_c for the ferromagnet, however, whereas it remains finite for the antiferromagnet.

2. Scaling Predictions

The analysis of the scaling hypotheses is similar to the case of the antiferromagnet. Let the temperature dependence of $\langle\mathbf{M}\rangle$ at the critical point be characterized by the exponent β. The stiffness constant ρ_s is proportional to ξ^{-1}, just as for the antiferromagnet.

In region I, we find[39]

$$\lambda=\rho_s/\langle\mathbf{M}\rangle\propto\xi^{-1}\epsilon^{-\beta}=\xi^{-1+\beta/\nu'}\approx\xi^{-1/2}\approx\epsilon^{1/3}\,, \tag{5.32}$$

$$\omega_\xi{}^{\mathbf{M}}(\mathbf{k})=\lambda k^2\propto k^{3-\beta/\nu'}(k\xi)^{-1+\beta/\nu'}\approx k^{5/2}(k\xi)^{-1/2}\,, \tag{5.33}$$

$$\eta_k=(\zeta/\lambda)k^2\propto(k\xi)^2\,, \tag{5.34}$$

$$\zeta\propto\xi^2\lambda\propto\xi^{1+\beta/\nu'}\approx\xi^{3/2}. \tag{5.35}$$

We have inserted the (approximate) exponent values $\beta=\frac{1}{2}$, $\nu'=\nu=\frac{2}{3}$; alternatively, the static scaling laws[9,11] may be used to express the exponent z of $\omega_\xi{}^{\mathbf{M}}(k)$ directly as $3-\beta/\nu'=\frac{1}{2}(5-\eta)\approx\frac{5}{2}$. From Eq. (5.34) we see that the damping is small in region I. The idea that spin waves will exist arbitrarily close to T_c in ferromagnets so long as $k\xi\lesssim 1$ and $T<T_c$ was put forward independently by Vaks *et al.*[51]

In region II, the characteristic frequency is given by

$$\omega_\xi{}^{\mathbf{M}}(\mathbf{k})=Bk^{3-\beta/\nu'}=Bk^{(5-\eta)/2}. \tag{5.36}$$

For finite $k\xi(T\neq T_c)$ there are corrections which will be of higher order in $(k\xi)^{-1}$.

In region III, the relaxation frequency is

$$\omega_\xi{}^{\mathbf{M}}(\mathbf{k})=Dk^2=\xi^{-1+\beta/\nu'}k^2\,, \tag{5.37}$$

which leads to a spin-diffusion constant going as

$$D\propto\epsilon^{\nu-\beta\nu/\nu'}\approx\epsilon^{1/3}. \tag{5.38}$$

[51] V. N. Kashcheev and M. A. Krivoglaz, Fiz. Tverd. Tela 3, 1541 (1961) [English transl.: Soviet Phys.—Solid State 3, 1117 (1961)]; A. B. Harris, Phys. Rev. 175, 674 (1968). Recently, V. G. Vaks, A. I. Larkin, and S. A. Pikin, Zh. Eksperim. i Teor. Fiz. 53, 1089 (1967) [English transl.: Soviet Phys.—JETP 26, 647 (1968)] have derived a decay rate of this form at all $T<T_c$ in the limit of a long-range spin-spin interaction, and have argued that it is probably valid for short-range forces as well.

[52] L. D. Landau and E. M. Lifshitz, Physik Z. Sowjetunion 8, 153 (1935).

Using the static scaling laws,[9,11] we may write the exponent $\nu-\beta\nu/\nu'$ in Eq. (5.38) as $\frac{1}{4}\gamma(1-\eta)(1-\frac{1}{2}\eta)^{-1}$. For $\eta=0$, the diffusion constant is proportional to $(\chi^M)^{-1/4}$, which is the behavior first suggested by Bennett and Martin,[53] and later by Kawasaki.[54]

The expression in Eq. (5.22) for the NMR linewidth may be applied to the ferromagnet, with **N** replaced by **M**, yielding[55]

$$\Delta \propto \int_0^\infty k^2dk\frac{k^{\eta-2}}{k^{3-\beta/\nu'}}\frac{g(k\xi)}{\Omega(k\xi)}f_{k\xi}(0) \propto \xi^{2-\eta-\beta/\nu'}\approx\epsilon^{-1}. \tag{5.39}$$

3. *Experimental Tests*

The ferromagnet is not as appropriate a system for testing the scaling predictions as the antiferromagnet, since there does not seem to exist any material which approximates the isotropic Heisenberg model. A fundamental difference with the antiferromagnet lies in the importance of the (anisotropic) dipolar part of the Hamiltonian, which is always important at long wavelengths in the ferromagnet, and thus must surely influence the critical phenomena. The situation might be analogous to the slightly anisotropic antiferromagnet discussed above, where there is an "apparent" critical region in one temperature range, and a change of behavior very close to T_c. In addition many ferromagnets are metals, and the presence of conduction electrons may have an effect on the critical phenomena. In any case, an understanding of dynamical critical behavior in real ferromagnets requires much more analysis than has been given here, and the scaling predictions presented above can only be used as a qualitative guide. At the present time, it is probably more useful to compare the dynamic-scaling laws to *theoretical* work on the Heisenberg ferromagnet, either approximate microscopic or numerical calculations, or other phenomenological theories. The experimental evidence on spin diffusion which exists at present[16] is inconclusive, since the $k\xi$ corrections are probably large near T_c, and the predicted temperature dependence of D [Eq. (5.38)] is very gradual. The fact that the k dependence of the relaxation frequency appears quadratic does not prove that diffusion is occurring (and that one is still in region III), since the exponent of k is always close to 2, and has a maximum value $\frac{5}{2}$ at T_c (region II).

C. Gas-Liquid Transition

1. *Hydrodynamics*

The hydrodynamic theory of fluctuation in a simple fluid is, of course, well known, and was first worked out in connection with light scattering by Landau and Placzek.[56] A particularly convenient formulation in terms of correlation functions was developed by Kadanoff and Martin.[26]

The order parameter is the density n (or more precisely $n-n_c$, where n_c is the critical density) and its correlation function at long wavelengths is

$$C^n(\mathbf{k},\omega)=C^n(\mathbf{k})\Bigg[\left(1-\frac{c_v}{c_p}\right)\left(\frac{2D_Tk^2}{\omega^2+(D_Tk^2)^2}\right)+\frac{c_v}{c_p}\left(\frac{2c^2k^2D_sk^2}{(\omega^2-c^2k^2)^2+(\omega D_sk^2)^2}\right)-\frac{2D_Tk^2}{ck}\left(1-\frac{c_v}{c_p}\right)\times\left(\frac{(\omega^2-c^2k^2)ck}{(\omega^2-c^2k^2)^2+(\omega D_sk^2)^2}\right)\Bigg], \tag{5.40}$$

where

$$C^n(0)=k_BT\chi^n=k_BTn\left(\frac{\partial n}{\partial p}\right)_T = n^3mk_B(c_p-c_v)\left[\left(\frac{\partial p}{\partial T}\right)_n\right]^{-2}, \tag{5.41a}$$

$$D_T=\kappa/mnc_p, \tag{5.41b}$$

$$D_s=\frac{\frac{4}{3}\zeta+\eta}{mn}+\frac{\kappa}{mnc_p}\left(\frac{c_p}{c_v}-1\right), \tag{5.41c}$$

$$c^2=\frac{1}{m}\left(\frac{\partial p}{\partial n}\right)_S=\frac{1}{m}\frac{c_p}{c_v}\left(\frac{\partial p}{\partial n}\right)_T =\frac{T(\partial p/\partial T)_n^2}{n^2m^2c_v(1-c_v/c_p)}. \tag{5.41d}$$

In Eqs. (5.40) and (5.41), m is the atomic or molecular mass, c_p and c_v are the heat capacities per unit mass at constant pressure and constant volume, κ is the thermal conductivity, and η and ζ are the shear and bulk viscosities, respectively. These equations hold for the gas and liquid separately below T_c as well as in the single fluid above T_c. To illustrate the use of the reduced variables of Eqs. (4.13)–(4.15), we calculate the characteristic frequency and shape function, using Eq. (5.40), in the long-wavelength limit ($D_sk^2\ll ck$, $D_Tk^2\ll ck$) and for $\gamma\equiv c_p/c_v>2$. We find

$$\omega^n(\mathbf{k})=D_Tk^2(1+d)+O(k^4), \tag{5.42}$$

$$f^n(x)\approx\frac{1}{\pi}(1-\gamma^{-1})\left(\frac{(1+d)^{-1}}{x^2+(1+d)^{-2}}\right)+\tfrac{1}{2}\gamma^{-1}[\hat{\delta}(x+b)+\hat{\delta}(x-b)], \tag{5.43}$$

with

$$(1+d)=\tan[\pi/4(1-\gamma^{-1})], \quad b=ck/D_Tk^2(1+d)\gg1, \tag{5.44}$$

[53] H. S. Bennett and P. C. Martin, Phys. Rev. **138**, A608 (1965).

[54] K. Kawasaki, J. Phys. Chem. Solids **28**, 1277 (1967).

[55] Due to a misprint in Ref. 24 the exponent of Eq. (5.39) was mistakenly written as ϵ^{+1} rather than ϵ^{-1}.

[56] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Addison-Wesley Publishing Co., Inc., Reading, Mass., 1960), Chap. XIV.

and where $\hat{\delta}$ is a Lorentzian of unit area and with a width small compared to b. Note that when $\gamma^{-1}\rightarrow 0$, $d\approx\frac{1}{2}\pi\gamma^{-1}\rightarrow 0$.

At the critical point the quantity $(\partial p/\partial T)_n$ remains finite, so that the specific heat c_p has the strong singularity of the compressibility χ^n [see Eq. (5.41a)],

$$c_p=c_p{}^0\epsilon^{-\gamma}, \quad T>T_c \quad \text{(critical isochore)} \tag{5.45a}$$

$$c_p=c_p{}^{0\prime}\epsilon^{-\gamma'}, \quad T<T_c \quad \text{(coexistence curve)}. \tag{5.45b}$$

(If scaling laws are to make sense, it is necessary that all critical exponents below T_c be the same for the liquid as for the gas.) The specific heat c_v is much more weakly singular[57] than c_p,

$$c_v=c_v{}^0\epsilon^{-\alpha}, \quad T>T_c \tag{5.46a}$$

$$c_v=c_v{}^{0\prime}\epsilon^{-\alpha'}, \quad T<T_c \tag{5.46b}$$

and from Eq. (5.41d) it follows that the sound velocity varies as $(c_v)^{-1/2}$. Near T_c the parameters $\gamma=c_v/c_p$ and d [Eq. (5.44)] go to zero and Eqs. (5.42) and (5.43) reduce to

$$\omega^n(k)=D_Tk^2, \tag{5.47a}$$

$$f^n(x)=\pi^{-1}/(x^2+1). \tag{5.47b}$$

These formulas show that in the fluid the sound-wave contribution to the order-parameter correlation function vanishes relative to the thermal diffusion contribution as $T\rightarrow T_c$ in the *hydrodynamic domain*.

Finally, let us introduce the (unknown) exponent describing the singularity of the thermal conductivity

$$\kappa=\kappa_0\epsilon^{-l}, \quad T>T_c \tag{5.48a}$$

$$\kappa=\kappa_0{}'\epsilon^{-l'}, \quad T<T_c. \tag{5.48b}$$

2. Scaling Predictions

The characteristic frequency may be found in region I from Eqs. (5.47a) and (5.41b):

$$\omega_\xi{}^n(\mathbf{k})=D_Tk^2\propto\xi^{(l'-\gamma')/\nu'}k^2 = k^{2-(l'-\gamma')/\nu'}(k\xi)^{(l'-\gamma')/\nu'}. \tag{5.49}$$

This determines the exponent of Eq. (4.16) to be

$$z=2+(\gamma'-l')/\nu'. \tag{5.50}$$

A similar evaluation is possible in region III. The restricted dynamic-scaling hypothesis predicts[39]

$$z=2+(\gamma-l)/\nu=2+(\gamma'-l')/\nu'. \tag{5.51}$$

If we have $\gamma=\gamma'$ and $\nu=\nu'$, as predicted by static scaling, then we must have $l=l'$. (The semiphenomenological calculation by Kadanoff and Swift[22] predicts in addition $l'=\nu'$, $l=\nu$.) In region II we have

$$\omega_\infty{}^n(\mathbf{k})=Bk^z, \tag{5.52}$$

with z again given by Eq. (5.51). For finite but small $k\xi$ (in region III, say) the general form (4.16) implies

$$\omega_\xi{}^n(\mathbf{k})=D_Tk^2[1+B'(k\xi)^2+\cdots], \tag{5.53}$$

where B' is a constant. A similar temperature-dependent correction to the thermal conduction frequency was first proposed by Fixman[18]; in the present case, however, the parameter ξ does not have the mean-field[6,7] temperature dependence $\xi\sim\epsilon^{-1}$.

In order to make predictions concerning the sound mode, an operator must be found which has this mode as its characteristic frequency in region I or III. Such an operator is the longitudinal part of the momentum-density operator

$$\mathbf{g}(\mathbf{r},t)=\sum_i \mathbf{p}_i\delta[\mathbf{r}-\mathbf{r}_i(t)], \tag{5.54}$$

where $\mathbf{p}_i$ and $\mathbf{r}_i(t)$ are the momentum and position of the ith particle of the system, respectively. The longitudinal part $C^{gl}(\mathbf{k},\omega)$ of the correlation function $C^g(\mathbf{k},\omega)$ may be obtained from $C^n(\mathbf{k},\omega)$ by applying the equation of continuity.[26] In the long-wavelength limit it turns out that $C^{gl}(\mathbf{k},\omega)$ is dominated by the sound mode. An extension of the dynamic-scaling assumption to the momentum density would imply, among other things, that the sound-damping constant would be given by

$$\tfrac{1}{2}D_sk^2\propto ck\times k\xi, \tag{5.55}$$

$$D_s\propto c\xi. \tag{5.56}$$

Kadanoff and Swift[22] show that this is not the correct behavior as $k\rightarrow 0$, even though Eq. (5.56) seems to apply for slightly larger k values.[58] The failure of extended dynamic scaling for the momentum-density operator suggests that the scaling hypothesis may be generally inapplicable to operators whose characteristic frequency $\omega_\xi{}^A(\mathbf{k})$ is different from that of the order parameter, in both hydrodynamic regions.

For a classical system, the equal-time momentum-density correlation function has the form

$$C^{gl}(\mathbf{k})\equiv mnk_BT. \tag{5.57}$$

Thus the static scaling law is trivially satisfied for the momentum density, but dynamic scaling does not hold.

[57] There is evidence (Ref. 3) that c_v is logarithmically infinite both above and below $T_c(\alpha\approx\alpha'\approx 0)$. We shall assume that this is so in what follows, although a finite cusp $(\alpha,\alpha'<0)$ or a stronger singularity $(\alpha,\alpha'>0)$ are also possible.

[58] The anomalous damping of sound found by Kadanoff and Swift (Ref. 22) at long wavelengths does not come from a $k\xi$ correction to the dispersion relation, as in Eq. (5.55). It is interesting to note, however, that they find D_sk^2/ck proportional to $\omega_1\tau$, where ω_1 is the sound frequency and τ is the characteristic time for fluctuations of the *order parameter*, $\tau=[\omega_\xi{}^n(k=\xi^{-1})]^{-1}\approx D_T{}^{-1}\xi^2$. Specifically, they predict that the thermal conductivity κ is proportional to ξ, and that $D_s=Ac^2\xi^2mnc_p\kappa^{-1}=A'\xi c_p\propto\epsilon^{-2}$, where A and A' are constant or very weakly divergent. This long-wavelength damping correction to the sound mode has the form $D_sk^2/ck=Ak\xi(c\xi^{-1}/D_T\xi^{-2})$, which means that the function f^{gl} depends not only on $k\xi$ but also on the temperature-dependent small parameter $(D_T\xi^{-2}/c\xi^{-1})$ which is a ratio of characteristic times for momentum and density fluctuations, respectively. It is worth pointing out that the three frequency regions defined by Kadanoff and Swift are all contained in our region I, and merely reflect the fact that for the sound mode, hydrodynamics breaks down long before the condition $k\xi\approx 1$ is reached.

3. *Experimental Tests*

Since the critical mode (5.42) is nonpropagating both above and below T_c, its temperature dependence cannot be related to the thermodynamic exponents alone, but involves also the exponent l [Eq. (5.48)]. A test of dynamic scaling thus must involve the comparison of measured dynamical behavior in different regions, namely, a test of Eqs. (5.51) and (5.52).

The symmetry of D_T above and below T_c has been verified by light-scattering experiments on CO_2[59] but has been found not to hold in SF_6.[60] Although there is a possibility that the discrepancy in SF_6 is caused by an asymmetry in ξ (i.e., $\nu\neq\nu'$), the mere fact that two seemingly simple fluids should behave differently casts serious doubt on the whole notion of a unique phase transition phenomenon in fluids. Until this discrepancy between the two substances has been explained, there seems little point in speculating on the possible breakdown or verification of dynamic scaling.

For those substances in which $D_T(\xi)$ behaves symmetrically, a number of tests of dynamic scaling appear feasible. The $(k\xi)^2$ correction to the dispersion relation (5.53) has been observed by light-scattering techniques in Xe,[61] and is of the right order of magnitude. The measurement is not precise enough to determine the temperature dependence of this correction, but could hopefully be improved. Another promising idea is to do both light scattering and neutron diffraction on the same substance, for instance, argon. By increasing the range of application of both techniques, it might be possible to measure overlapping regions of $k\xi$, and to check the consistency of the two sets of measurements of the density correlation function. Even in the absence of such an overlap, the light-scattering experiments can measure D_T accurately in region III, and thus determine the exponent z of Eq. (5.50). This would then completely fix the k dependence in region II, Eq. (5.52), which is relatively easily accessible to neutrons. This experiment would be a detailed test of dynamic scaling, which might be considerably more rigorous than the experiments in antiferromagnets, since the $k\xi\ll 1$ limit may be measured with the more precise technique of light scattering. In addition, the hydrodynamic formulas (5.40) and (5.41) are exact for real fluids, whereas the corresponding expressions in antiferromagnets apply only to a simple isotropic model.

It is worth pointing out that the critical point of binary-fluid mixtures has properties which are very similar to those of the simple-fluid critical point. Critical scattering of light once again can measure the width of the central diffusive peak,[62] and it is probably also possible to compare these measurements with neutron-scattering results in region II.

D. λ Transition of Superfluid Helium

1. *Hydrodynamics*

The two-fluid hydrodynamics of Landau and Khalatnikov[63] leads to correlation-function expressions which were evaluated earlier.[64] The order parameter $\Psi(\mathbf{r},t)$ is equal to the quantum field operator $\psi(\mathbf{r},t)$ for annihilation of a helium atom. Since the field operators are non-Hermitian, the average order parameter is a complex number, analogous to a two-dimensional vector. The gauge invariance of the helium Hamiltonian corresponds to rotational invariance in a spin system; for the superfluid it is an exact symmetry of the real system, however, rather than a property of an approximate model. By treating the Hermitian and non-Hermitian parts of ψ as the components of a two-dimensional vector, we find that the order-parameter correlation function analogous to (5.1) is

$$\hat{C}^{\psi}(\mathbf{r},t)$$
$$=\tfrac{1}{2}\mathrm{Re}\langle\{(\psi(\mathbf{r},t)-\langle\psi(\mathbf{r},t)\rangle),(\psi^{\dagger}(0,0)-\langle\psi^{\dagger}(0,0)\rangle)\}\rangle. \quad (5.58)$$

For $T<T_\lambda$ the angular brackets denote an average in a "reduced ensemble" in which the phase of the order parameter has a definite value.[65] In the normal fluid $(T>T_\lambda)$ the equilibrium ensemble is identical to the grand-canonical ensemble; the correlation function is isotropic in the complex plane, and decays exponentially at large distances. For $T<T_\lambda$, let us suppose the order parameter to be "lined up" along the real axis,

$$\mathrm{Im}\langle\psi(\mathbf{r},t)\rangle=0. \quad (5.59)$$

The correlation function splits into a parallel part and a perpendicular part, corresponding to fluctuations of the real and imaginary parts of ψ ("magnitude" and "phase" fluctuations), respectively:

$$\hat{C}^{\psi}(\mathbf{r},t)=\hat{C}^{R}(\mathbf{r},t)+\hat{C}^{I}(\mathbf{r},t)\,. \quad (5.60)$$

According to two-fluid hydrodynamics the perpendicular component $\hat{C}^{I}(\mathbf{r},t)$ dominates at large r,[64] just as for antiferromagnets. The Fourier transform of $\hat{C}^{\psi}(\mathbf{r})$ at small k is

$$C^{\psi}(\mathbf{k})=a/k^2+\cdots=k_BTn_0m^2/\hbar^2\rho_sk^2+\cdots, \quad (5.61)$$

where $n_0=|\langle\psi\rangle|^2$, and the "superfluid density" ρ_s is a

[59] H. L. Swinney and H. Z. Cummins, Phys. Rev. **171**, 152 (1968); B. Maccabee and J. A. White, Bull. Am. Phys. Soc. **13**, 182 (1968).

[60] N. C. Ford, Jr., and G. B. Benedek, Phys. Rev. Letters **15**, 649 (1965).

[61] Y. Yeh, Phys. Rev. Letters **18**, 1043 (1967).

[62] B. Chu, invited talk delivered at meeting of the American Physical Society, Boston, 1968 (unpublished); Phys. Rev. Letters **18**, 200 (1967).

[63] I. M. Khalatnikov, *Introduction to the Theory of Superfluidity* (W. A. Benjamin, Inc., New York, 1965), Chaps. 8–10.

[64] P. C. Hohenberg and P. C. Martin, Ann. Phys. (N. Y.) 34, 291 (1965).

[65] It is convenient theoretically to consider the λ transition at constant chemical potential μ. We can then add a constant potential to the Hamiltonian, so that $\mu=0$, and the time derivative of the phase is zero in equilibrium. All singularities will be unchanged, however, if the experiments are done at constant pressure.

stiffness constant for fluctuations of the phase φ, i.e.,

$$\rho_s=\frac{\partial^2\tilde{\epsilon}}{\partial v_s^2}=\left(\frac{m}{\hbar}\right)^2\frac{\partial^2\tilde{\epsilon}}{\partial(\nabla\varphi)^2}, \tag{5.62}$$

where $\tilde{\epsilon}$ is the energy density.[66] Near T_λ, in the superfluid state ($T<T_\lambda$), the correlation length is given by Eqs. (2.4) and (5.61) as

$$\xi=m^2k_BT/4\pi\hbar^2\rho_s. \tag{5.63}$$

For $T>T_\lambda$, the correlation length is deduced from the long-wavelength form of $C^\psi(\mathbf{k})$,

$$C^\psi(\mathbf{k})=A'/(k^2+\xi^{-2}),\quad k\rightarrow 0,\quad T>T_\lambda \tag{5.64}$$

where A' is temperature-independent if the Ornstein-Zernike theory is valid ($\eta=0$), but depends on ξ for $\eta\neq 0$.

Below T_λ, the dynamic correlation function $C^\psi(\mathbf{k},\omega)$ is dominated by second sound, whose frequency is

$$\omega_2(k)=c_2k, \tag{5.65}$$

with

$$c_2^2=T\rho_s s^2/\rho_n c_p, \tag{5.66}$$

where $\rho_n\equiv nm-\rho_s$, and $s=S/mN$ is the entropy per unit mass. The attenuation of second sound is determined from the decay rate

$$\Gamma_2(\mathbf{k})=\tfrac{1}{2}D_2k^2, \tag{5.67}$$

where the damping constant D_2 is expressible in terms of the transport coefficients [see Eq. (4.22) of Ref. 64].

The above formulas are valid[67] to lowest order in small parameter $\gamma^{-1}\equiv c_p/c_v-1$, which varies from $\approx 10^{-4}$ at 1°K to $\approx 10^{-2}$ at $T-T_\lambda\approx 10^{-5}$ °K. For finite $\gamma-1$, the first-sound pole appears in $C^\psi(\mathbf{k},\omega)$, and the relations determining the second-sound velocity and damping, Eqs. (5.66) and (5.67), must be modified. The complete formulas are given in Ref. 64, Eqs. (4.14)–(4.18).

Above T_λ the frequency dependence of the order-parameter correlation function cannot be determined from hydrodynamics since the operator ψ is not a constant of the motion, nor is it coupled to constants of the motion in the macroscopic limit. This situation is analogous to the antiferromagnet, where $\mathbf{N}$ does not commute with the Hamiltonian. The function $C^\psi(\mathbf{k},\omega)$ is presumably peaked about $\omega=0$, with a width $\Gamma(\mathbf{k})$ which in general goes to a nonzero limit as $k\rightarrow 0$.

The order-parameter correlation function is not directly measurable either above or below T_λ, and all verifications of the predictions of two-fluid hydrodynamics must involve other operators, which couple to ψ in the long-wavelength limit. The operator

$$q(\mathbf{r},t)=\tilde{\epsilon}(\mathbf{r},t)+[\langle\tilde{\epsilon}+p\rangle/\langle n\rangle]n(\mathbf{r},t) \tag{5.68}$$

plays the role of a heat operator.[68] [In (5.68), p is the pressure and the angular brackets denote an equilibrium expectation value.] The correlation function $C^q(\mathbf{k},\omega)$ may be evaluated in the hydrodynamic domain[64] in the superfluid phase, where it is exhausted by the second-sound mode (5.64), corresponding to the fact that second sound is a pure temperature wave in the limit $\gamma-1\rightarrow 0$. In the normal fluid ($T>T_\lambda$), the heat correlation function is exhausted at long wavelengths by the thermal conduction mode, whose frequency was given in Eqs. (5.40) and (5.41b). The shape function f^q has the form given in Eq. (5.47b).

The definition of the heat operator of Eq. (5.68) depends on temperature through $\langle\tilde{\epsilon}+p\rangle$ and $\langle n\rangle$, and q is thus not microscopic[29] in the sense of Sec. II. Since the expectation values and their derivatives are relatively slowly varying functions of temperature near T_λ, however, the scaling predictions may be derived for q itself, as if it were a microscopic operator.[69]

The density correlation function $C^n(\mathbf{k},\omega)$ is exhausted by the first-sound mode at long wavelengths in the limit $\gamma-1=0$. Thus, for the usually accessible range of temperature, $C^n(\mathbf{k},\omega)$ is not expected to obey the dynamic-scaling laws. [See the discussion of $C^{gl}(\mathbf{k},\omega)$ in Sec. V C.] On the other hand, for T *very* close to T_c, the parameter $\gamma-1$ is expected to become larger than 1, if c_p diverges logarithmically, because c_v cannot actually be infinite at T_c.[70] The temperature interval for which $\gamma-1\geq 1$ is unattainably small for helium at its vapor pressure, but may be increased to $\epsilon\leq\theta\approx 10^{-4}$ at 27 atm.[23,71] For $\epsilon\ll\theta$, $C^n(\mathbf{k},\omega)$ is dominated by a second-sound pole in region I, and by thermal diffusion in region III. Thus there may be a small region of temperatures and wave vectors where $C^n(\mathbf{k},\omega)$ obeys dynamic scaling.

[66] In Ref. 64, the energy density E/V was denoted by ϵ, but here we use $\tilde{\epsilon}$, to distinguish it from $\epsilon\equiv|T-T_c|/T_c$.

[67] The essential assumption needed to prove these formulas is the validity of an expansion of the currents in powers of k^2 at long wavelengths [see Eq. (4.3) of Ref. 64, and Refs. 63 and 37]. This expansion depends on the existence of local equilibrium, characterized by local values of the density, the energy density, the momentum density, and the superfluid velocity. A test of the validity of the expansion away from T_λ is the verification of the linear and quadratic powers of k in $\omega_2(\mathbf{k})$ and $\Gamma_2(\mathbf{k})$, respectively, in a real second-sound wave for sufficiently small k. Although such a verification has been carried out in the temperature range between 1 and 1.8°K [see W. B. Hanson and J. R. Pellam, Phys. Rev. **95**, 321 (1954)], further, more accurate experiments would be desirable, especially near T_λ where any effects associated with the breakdown of hydrodynamics inside region I might be expected to be observable.

[68] More precisely, for long-wavelength fluctuations, $q(\mathbf{r},t)-\langle q\rangle$ is equal to the change in the local equilibrium value of the entropy *per particle*, multiplied by the average density and temperature of the system as a whole. The statements made in Ref. 26, p. 441 and in Ref. 22, p. 90 that the change in q represents T times the change in entropy *density* are not strictly correct.

[69] Indeed, an operator $Q\equiv\tilde{\epsilon}(\mathbf{r},t)+h_\lambda n(\mathbf{r},t)$ may be defined, where $h_\lambda=\langle\tilde{\epsilon}+p\rangle/\langle n\rangle|_{T=T_\lambda}$; this operator is microscopic and has correlation functions which agree with those of q up to terms which vanish as $c_p(T-T_\lambda)^2$ near T_λ. The scaling predictions are thus identical for Q and q.

[70] M. J. Buckingham and W. M. Fairbank, in *Progress in Low Temperature Physics*, edited by C. J. Gorter (North-Holland Publishing Co., Amsterdam, 1961), Vol. III, p. 80.

[71] V. Korenman, University of Maryland Technical Report, Department of Physics, 1967 (unpublished); G. Ahlers (private communication).

2. Scaling Predictions

The restricted dynamic-scaling hypothesis implies that

$$\omega^{\psi}(k)=c_2k+\cdots\propto\xi^{-1/2}k+\cdots=k^{3/2}[(k\xi)^{-1/2}+\cdots],$$
$$=k^{3/2}\Omega(k\xi)\,, \tag{5.69}$$

where we have used Eqs. (5.63) and (5.66) and have neglected the relatively weak divergence of c_p. As shown in Ref. 64, the poles of $C^{\psi}(\mathbf{k},\omega)$ [it was called $\mathcal{G}^{(0)}(\mathbf{k},\omega)$] are identical to those of the *observable* heat correlation function $C^q(\mathbf{k},\omega)$ in region I, where $\omega_\xi{}^q(\mathbf{k})$ is the frequency of second sound. (For $\gamma-1\ll1$, the heat correlation function is measured at long wavelengths by experiments on the propagation of temperature fluctuations.) The width of the peak in the spectrum of ψ is identical to the width of the second-sound peak in $C^q(\mathbf{k},\omega)$, to lowest order in k. From Eq. (5.67) and the *restricted* scaling hypothesis (4.17) we thus obtain

$$D_2\propto c_2\rho_s{}^{-1}\,, \tag{5.70}$$

a result which was first derived by Ferrell *et al.*[23] Restricted scaling predicts in addition that $\omega^{\psi}(\mathbf{k})$ is proportional to $k^{3/2}$ in region II, while in region III the Bose field should have a relaxation rate proportional to $\xi^{-3/2}$, similar to the staggered magnetization of the antiferromagnet. Outside of region I, however, the order-parameter correlation function is not simply related to observable quantities and the restricted scaling hypothesis cannot be tested. The remaining predictions must follow from extended scaling, applied to the operator q. In region II, we have

$$\omega^q(\mathbf{k})=Bk^{3/2}\,, \tag{5.71}$$

and in region III [see Eq. (5.47)]

$$\omega^q(k)=D_Tk^2=k^{3/2}\Omega(k\xi)\propto\xi^{1/2}k^2\,, \tag{5.72}$$

which implies $D_T\propto\xi^{1/2}$, and using Eq. (5.41b), $\kappa\propto\xi^{1/2}$. In order to obtain a more precise prediction we may employ the asymptotic matching condition, Eq. (4.7′). The thermal conductivity turns out to be proportional to $\xi^{1/2}c_p{}^+/(c_p{}^-)^{1/2}$, where $c_p{}^+$ and $c_p{}^-$ are the heat capacities at the given value of $|T-T_c|$, above and below T_λ, respectively. This result was also obtained originally by Ferrell *et al.*[23]

Just as for the gas-liquid transition, the first-sound mode contributes a negligible weight to order-parameter fluctuations. Also, extended dynamic scaling (applied to the current or the density operator) does not predict the correct critical behavior. The more detailed theory of Kadanoff and Swift[22] permits a calculation of the damping of first sound, which agrees with experiment[72] for $T<T_\lambda$, but disagrees for $T>T_\lambda$. In order to calculate the damping of first sound below T_λ, Ferrell and coworkers[23] *assumed* that the second viscosity ζ_2 is the transport coefficient which leads to the large damping of second sound given in Eq. (5.70). They then inserted the temperature dependence of ζ_2 into the hydrodynamic expression for the damping of first sound. Their additional assumption does not follow from dynamic scaling, and does not serve to explain the anomalous damping of first sound above T_λ.[72]

[72] M. Barmatz and I. Rudnick, Phys. Rev. **170**, 224 (1968).

3. Experimental Tests

The predicted damping of second sound, which follows from the restricted hypothesis, may be tested by a direct measurement on a macroscopic second-sound wave below T_λ. It is important to note, however, that very near T_λ the nonlinear corrections to the hydrodynamic equations may become quite large in practice, and prevent a macroscopic measurement of the properties of second sound. In the linear regime for which the scaling prediction applies, the damping coefficient must be independent of the amplitude of the wave. The divergence of the thermal conductivity above T_λ may be tested in a conventional static experiment for $T>T_\lambda$, and has already been qualitatively verified by Kerrisk and Keller.[73] Unlike the second-sound damping experiment, the measurement of thermal conductivity is a test of extended rather than restricted scaling. As mentioned above, for temperatures very close to T_λ, the density-correlation function becomes coupled to second sound. Since $C^n(\mathbf{k},\omega)$ can be measured by neutron or light-scattering techniques, this offers the possibility of observing the critical fluctuations in a scattering experiment, and thus of observing nonhydrodynamic behavior (e.g., region II). In practice, however, the temperature interval required is so small ($\epsilon<\theta$) that the experiment is unfeasible, except possibly at high pressures.[23,73]

Note added in proof. A verification of the scaling prediction for the damping of second sound [Eq. (5.70)] was recently reported by J. A. Tyson.[73a] The divergence of the thermal conductivity was measured by G. A. Ahlers,[73b] who verified the detailed behavior predicted by the matching condition (4.7′), $\kappa\propto\xi^{1/2}c_p{}^+/(c_p{}^-)^{1/2}$. A departure from the dynamic scaling prediction was reported by Archibald *et al.*[73c] for very short samples (<0.2 mm).

4. Mixtures of He^3 in He^4

When small amounts of He^3 are introduced into He^4, the λ temperature decreases and the character of the hydrodynamic modes is modified. As shown by Pomeranchuk[74] and by Khalatnikov,[75] the He^3 becomes part

[73] J. F. Kerrisk and W. E. Keller, Bull. Am. Phys. Soc. **12**, 550 (1967); see also Ref. 23.
[73a] J. A. Tyson, Phys. Rev. Letters **21**, 1235 (1968).
[73b] G. Ahlers, Phys. Rev. Letters **21**, 1159 (1968).
[73c] M. Archibald, J. M. Mochel, and L. Weaver, Phys. Rev. Letters **21**, 1156 (1968).
[74] I. Pomeranchuk, Zh. Eksperim. i Teor. Fiz. **19**, 42 (1949).
[75] I. M. Khalatnikov, Zh. Eksperim. i Teor. Fiz. **23**, 265 (1952); see also Ref. 63, part IV.

of the normal fluid, and therefore oscillates with respect to the superfluid in a second-sound wave. This means that second sound involves both temperature and concentration fluctuations, and will therefore appear in the spectrum of the concentration correlation function. Its strength is $(\rho/c)\partial c/\partial\rho$, where ρ is the total *mass* density and c is the concentration of He^3. As shown by Gor'kov and Pitaevskii,[76] and in somewhat more detail by Ganguly and Griffin,[77] the spectrum of light scattered from this system will include the concentration fluctuations, and will therefore have a peak at the second-sound frequency. This offers the possibility to test the prediction of Eq. (5.70), which follows from restricted dynamic scaling, by a scattering experiment. On the other hand, a traditional second-sound propagation experiment is also possible in the mixtures, and a comparison of the results of the two techniques may give information on the relationship between correlation functions and macroscopic hydrodynamics. In particular, any differences between the properties of a mode containing a small number of quanta and one that has been macroscopically excited might show up near T_λ in such experiments.[78] Of course, the behavior of the concentration correlation function outside of region I will not give information about the order parameter, and only extended scaling can be tested. Furthermore, it seems more likely that $C^c(\mathbf{k},\omega)$ violates the extended-scaling hypothesis than was the case for $C^q(\mathbf{k},\omega)$ in pure helium. Indeed, in the mixtures, neither $C^c(\mathbf{k},\omega)$ nor $C^q(\mathbf{k},\omega)$ will be exhausted by the critical mode for the order parameter, second sound, in region I, since these functions also contain a diffusive central peak[76,77] whose strength increases with increasing concentration c. It may be possible, nevertheless, to apply dynamic scaling to an operator A which is an appropriate linear combination of q and c, and whose correlation function is exhausted by second sound in region I. The characteristic frequency $\omega^A(\mathbf{k})$ has the form (5.67) in region III, with a diffusion coefficient D^A whose temperature dependence can be determined by extended dynamic scaling. The result is

$$D^A \propto c_2 \rho_s^{-1}, \qquad (5.73)$$

where the left and right sides of Eq. (5.73) are measured at the same distance from T_c, above and below, respectively. The thermal diffusion constant D_T and the mass diffusion[79] D are linearly related to D^A and to another diffusion constant which we expect to be less singular than D^A. It follows that the constants D_T and D will have the same divergence as D^A. We therefore find

$$\kappa/C_{p,c} \propto c_2/\rho_s, \qquad (5.74)$$

where κ is the thermal conductivity, and $C_{p,c}$ is the specific heat at constant pressure and concentration, measured above T_c. The specific heat is not expected to diverge at T_c for finite concentrations,[80] so that the matching condition (4.7′) predicts a slightly different divergence of κ in mixtures and in pure He^4. It would be extremely interesting to measure κ, $C_{p,c}$, c_2, and ρ_s at a particular concentration and to test the detailed relation (5.74) predicted by extended dynamic scaling. In order to observe deviations from the behavior in pure He^4, it will be necessary to study mixtures with He^3 concentrations sufficient to depress the transition temperature well below the λ point of pure He^4.

VI. COMPARISON WITH OTHER THEORIES AND CONCLUSIONS

The phenomenon of critical slowing down of fluctuations, which was predicted by Van Hove[14] for the ferromagnet and fluid, is seen to be a common feature of second-order phase transitions, and is closely connected to the divergence of the spatial range of correlations. Critical slowing down is limited to the hydrodynamic regions I and III, however, since at fixed k the characteristic fluctuation frequency will not vanish as $T\to T_c$, but rather go to its finite limiting value $\omega(k)=Bk^z$ in region II. Consequently, as pointed out by Marshall,[17] in a typical neutron-scattering experiment where relatively large k values are employed, the Van Hove ("conventional") theory will fail to account for even the qualitative behavior of the relaxation phenomena at T_c. In light scattering from a fluid on the other hand, the $k\xi$ corrections to the conventional theory are small ($k\xi\approx 0.1$ for $\epsilon=10^{-4}$ and $k^{-1}=5000$ Å), and critical slowing down is in fact observed,[59,60] in accordance with Van Hove's qualitative predictions. The quantitative result, that $D\propto\chi^{-1}$, is not found in the scaling theory, since the transport coefficient Λ of Eq. (1.2) in general diverges.[81]

In discussing the failure of the conventional theory in a ferromagnet, Marshall[17] states that the observed behavior[16] of the spin-diffusion coefficient suggests that "spin-wave motion exists above T_c." Such an inference does not follow from the scaling theory. It is, of course, only in region II that spin-wave motion could occur at long wavelengths for $T>T_c$, since it is known not to be present in the region where hydrodynamics is valid. Of the various possible forms for the shape function f in region II, depicted in Fig. 3, only the one in part (a) may reasonably be interpreted in terms of "spin-wave

[76] L. P. Gor'kov and L. P. Pitaevskii, Zh. Eksperim. i Teor Fiz. **33**, 634 (1957) [English transl.: Soviet Phys.—JETP **6**, 486 (1958)].
[77] B. N. Ganguly and A. Griffin, Can. J. Phys. **46**, 1895 (1968).
[78] This general question is discussed in further detail in Ref. 37.
[79] Reference 25, p. 224.

[80] J. C. Wheeler and R. B. Griffiths, Phys. Rev. **170**, 249 (1968).
[81] In the antiferromagnet, for instance, we have $\Gamma^N(0)\propto\xi^{-3/2}$ and $\Lambda^N\equiv\Gamma^N(0)\chi^N\sim\epsilon^{-\nu(1+\eta)/2}$. In certain cases, such as thermal diffusion in liquid helium or *total* spin diffusion in the antiferromagnet, the divergence of Λ is predicted to be stronger than that of χ, and the ratio $D=\Lambda/\chi$ *diverges* at T_c, corresponding to a critical speeding up of fluctuations. Note, however, that these predictions do not refer to the order parameter, and thus follow from extended rather than restricted dynamic scaling. See also footnote 46.

motion," yet all the forms are consistent with the same behavior in region III.[82]

Recently, Kawasaki[83] and Villain[84] have independently presented theories of critical relaxation in ferromagnets and antiferromagnets for $t \gtrsim t_c$, using a random-phase approximation, or an RPA for decoupling correlation functions, and assuming the validity of the Ornstein-Zernike[19] theory ($\eta=0$). The resulting expressions for the dynamic correlation functions, when expressed in terms of the static exponents ν and β, are equivalent to the results of the scaling analysis for $\eta=0$. Subsequently, Villain[85] was able to modify his theory to include the possibility of a finite η, and found agreement with the results of Ref. 24, by making scaling assumptions which are similar to ours, and claimed to be in fact weaker. Similarly, Kawasaki[86] reformulated his theory, and derived the dynamic-scaling laws, using Mori's theory of Brownian motion[87] and the static-scaling assumptions.[9,11] The fact that the same results are obtained from such seemingly different approaches seems to us to lend support to our assumptions.

A calculation of the dynamic correlation function in a ferromagnet was performed recently by Resibois and de Leener[88] also using a random-phase approximation, or an RPA. These authors find agreement with the scaling laws (for $T>T_c$ and $\eta=0$), and in addition predict the existence of propagating spin waves in region II, in accordance with Marshall's[17] suggestion. This result must follow from the details of their model, rather than general scaling arguments. Recently, Mori and Okamoto[89] applied the continued-fraction representation of Mori[90] to a calculation of relaxation in ferromagnets, and derived results similar but not identical to ours. They find a damping rate for $\mathbf{M}$ which is homogeneous in k and ξ^{-1} with exponent $z=\frac{1}{2}(5+\eta)$, rather than $z=\frac{1}{2}(5-\eta)$. In addition, if $\eta\neq 0$, this damping rate does not scale with the real part of the spin-wave frequency predicted by hydrodynamics. Since η is in practice very small, it is unlikely that this feature can be tested experimentally.

[82] It must be remembered that region II is macroscopic, in the sense that k^{-1} and ξ are both large compared to microscopic lengths. For shorter wavelengths ($k\bar{a}\sim 1$), there might very well be approximate spin-wave excitations for $T\gtrsim T_c$, associated with short-range order or with other properties of the system which are not singular at T_c. These approximate spin waves, which have been discussed in the literature, and seem to have been observed experimentally, need not have any relation to the spectrum in region II. See, e.g., the theoretical discussions of R. Brout, Phys. Letters **24A**, 117 (1967); J. L. Beeby and J. Hubbard, *ibid.* **26A**, 376 (1968); see also the experiments of T. Riste, J. Phys. Soc. Japan Suppl. **17**, 60 (1962); and of Nathans *et al.* (Ref. 47).

[83] K. Kawasaki, Progr. Theoret. Phys. (Kyoto) **39**, 285 (1968).

[84] J. Villain, J. Phys. (Paris) **29**, 321 (1968); **29**, 687 (1968).

[85] J. Villain (to be published).

[86] K. Kawasaki, Progr. Theoret. Phys. (Kyoto) **39**, 1133 (1968); **40**, 11 (1968).

[87] H. Mori, Progr. Theoret. Phys. (Kyoto) **33**, 423 (1965).

[88] P. Resibois and M. de Leener, Phys. Letters **25A**, 65 (1967).

[89] H. Mori and H. Okamoto, Phys. Letters **26A**, 249 (1968); and (to be published).

[90] H. Mori, Progr. Theoret. Phys. (Kyoto) **34**, 399 (1965).

As mentioned earlier, the semiphenomenological theory of Kadanoff and Swift[22] for the critical point of a fluid is more detailed than the scaling theory, since it allows a direct calculation of the anomaly in transport coefficients. On the other hand, it also contains a certain number of unverified scaling assumptions, and moreover makes use of heuristic concepts and qualitative arguments in order to estimate the divergences in various quantities. It is hoped that the present more deductive approach may illuminate the significance of the semiphenomenological calculation by distinguishing between those properties which may be predicted by general scaling arguments, and those for which a detailed analysis is necessary. In particular, Kadanoff and Swift find a divergence of the sound-attenuation coefficient at long wavelengths that disagrees with the prediction that one would obtain from extending dynamic scaling to the current operator. This example suggests that the scaling theory does not apply to all "microscopically defined" operators.

At the λ point of superfluid helium, we have rederived the prior results of Ferrell and co-workers,[23] but have made a distinction between predictions referring to the superfluid phase, which follow from restricted scaling, and those referring to the normal phase, which depend on an extension.

Finally, we wish to comment on the anology which is sometimes drawn between spin waves and the Brillouin doublet in a fluid.[91] As the critical point is approached from below, the spin-wave peaks broaden and their position moves in toward $\omega=0$. It is possible that a central peak will also emerge, and become predominant at T_c [see Fig. 3(b)]. The analogy to the thermal conduction peak in the density-correlation function of a fluid seems to us misleading, however, since in the fluid the phenomenon is entirely describable by hydrodynamics. Indeed, at long wavelengths the weight of the thermal conduction peak is a fraction $(1-c_v/c_p)$ of the total weight of density fluctuations, independent of the behavior of the damping constants[92] [see Eq. (5.40)]. Near T_c, the total weight diverges as c_p, and the Rayleigh peak dominates the frequency spectrum in the hydrodynamic regions ($k\xi\ll 1$) [see Eqs. (5.43) and (5.48)]. In the magnetic systems, on the other hand, if our hydrodynamic analysis is correct, the spin waves alone exhaust the frequency spectrum of $\omega^{\mathbf{M}}(\mathbf{k},\omega)$ at long wavelengths for all $T<T_c$. Any additional modes must come from corrections to hydrodynamics, which are of higher order in $k\xi$, and may also contribute to spin-wave damping. The spin waves are *not* analogous to sound waves in a fluid, since the sound waves are

[91] See, e.g., the discussion in Ref. 17.

[92] The statement made in Ref. 17, p. 141, that the diffuse (Rayleigh) mode appears on the imaginary axis as soon as the sound waves are damped, is incorrect. The relative strengths of the Rayleigh and Brillouin modes are determined by thermodynamics alone; only the positions of the poles in the complex plane depend on damping coefficients.

not the characteristic mode of the order parameter in region I. In contrast to the magnetic system, the gas-liquid transition does not have a propagating critical mode $\omega^{\Psi}(\mathbf{k})$, either above or below the transition. The fluid system which is most closely analogous to magnetic systems (in particular, antiferromagnets) is superfluid helium, where the critical mode is *second* sound (analogous to spin waves) below T_λ, and a nonhydrodynamic relaxation mode $[\omega^{\Psi}(0)\neq 0]$ above T_λ.

The scaling viewpoint gives a unified description of a wide variety of critical phenomena, both static and dynamic. Although it is too early to tell whether any of the new predictions of the dynamic theory are exactly correct, it is hoped that the qualitative picture can already be a useful guide to further experimental investigation. As more accurate experimental results become available, and more detailed microscopic and phenomenological theories are developed, their results can be compared to those of the scaling theory. The specific mathematical assumptions of dynamic scaling can then be put to a test, and the reasons for their success or failure investigated.

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Ultrasonic Velocity and Attenuation in KH_2PO_4†

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The velocity and attenuation of ultrasonic shear waves have been investigated at temperatures above $T_C=121.8_2$°K in single-crystal KDP. An elastic Curie-Weiss law, $(s_{66}^E-s_{66}^P)=D/(T-T_C)$, is obtained with an elastic Curie constant D equal to 6.31×10^{-11} dyn^{-1} cm^2 deg. The attenuation data are consistent with a cooperative relaxation time at constant stress τ which varies as $\tau=24\times10^{-12}/(T-T_C)$ sec.

INTRODUCTION

At a Curie temperature of ~122°K, potassium dihydrogen phosphate (KDP) undergoes a cooperative transition from a paraelectric to a ferroelectric phase.[1,2] There has been a great deal of current interest, both theoretical[3–7] and experimental,[8–11] in this transition and the analogous transition at ~223°K in KD_2PO_4. The present ultrasonic investigation involves the measurement of the anomalous shear velocity and attenuation at temperatures above T_C.

In its paraelectric phase, KDP is tetragonal $(m)\bar{4}2$ and the x_y mechanical strain is coupled to the polarization along the ferroelectric z axis. Therefore, a transverse ultrasonic wave propagating in the [100] direction with its polarization in the [010] direction is the shear wave of interest. The elastic constant related to this shear is called c_{66}, but there are two limiting values of this constant depending on the electrical boundary conditions. One can specify the elastic properties at constant dielectric displacement (c^D) or at constant electric field (c^E). Mason's low-frequency resonance measurements[12] on bare and on plated crystals show that c_{66}^D exhibits a normal linear temperature dependence above T_C, whereas c_{66}^E drops toward zero at the Curie point. Elastic shear constants of KDP measured at ultrasonic frequencies have been shown by Jona[13] to correspond to c^E; thus, we have determined $c_{66}^E=\rho U^2$, where ρ is the density and U is the ultrasonic velocity.

In addition to the rapid decrease in velocity, there is an anomalous increase in the attenuation of this shear wave as the temperature approaches the Curie point. Since KDP is still piezoelectric in its paraelectric phase, there is a strong coupling between the elastic wave and the polarization even above T_C. Thus, attenuation measurements provide a convenient way to determine the polarization relaxation time. As in other cases

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