GENERALIZATION OF SCALING LAWS TO DYNAMICAL PROPERTIES OF A SYSTEM NEAR ITS CRITICAL POINT*

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(Received 17 July 1967)

The Widom-Kadanoff scaling laws are generalized to dynamic phenomena, by making assumptions on the structure of time-dependent correlation functions near T_c. The theory is applied to isotropic Heisenberg antiferromagnet and predictions are made, which can be tested by inelastic neutron-scattering and nmr measurements. The ferromagnet is also briefly discussed.

Various systems at their critical points are known to display singularities in their transport coefficients and elementary excitation spectra, in addition to thermodynamic singularities.[1] Although no first-principles theory of critical behavior exists at present, the Widom-Kadanoff scaling laws[2,3] have provided relations between the exponents of various divergent quantities which are in good agreement with experiments and with numerical studies.[4] Until recently, these laws dealt only with time-independent quantities, but it is natural to inquire into the underlying dynamical structure which leads to the static scaling properties. In recent notes, Ferrell and co-workers[5,6] applied the scaling principle to predict anomalous transport properties of helium near the lambda point.

In this Letter we outline a general theory of dynamical properties, which seems to us to be the simplest generalization of the static scaling laws,[2,3] and whose application to the lambda transition reproduces the essential predictions of Ferrell et al.[5,6] An attempt is made to state our assumptions quite explicitly and to avoid the use of hydrodynamic concepts in regions where they do not apply. The theory is then applied to the isotropic Heisenberg antiferromagnet and ferromagnet, where a number of specific predictions can be made about the behavior of time-dependent correlation functions near the transition temperature.

Basic to the theory of static scaling laws is the concept of a unique correlation length ξ which becomes infinite at the transition, and is a measure of the deviation of the temperature from T_c. At a fixed temperature the length ξ defines two domains, the long-wavelength or macroscopic region ($k\xi \ll 1$) and the critical region ($k\xi \gg 1$), the second of which extends down to $k=0$ at T_c. At temperatures far from T_c, when ξ has atomic dimensions, the long-wavelength, low-frequency[7] dynamical properties of a system are often described by an exact macroscopic theory, which we shall always refer to as "hydrodynamics." The length ξ becomes macroscopic as T approaches T_c, and it is expected that hydrodynamics will break down for wavelengths short compared with ξ. The detailed dynamical theory which replaces hydrodynamics in the critical region ($k\xi \gg 1$) is certainly very complicated, and in large part unknown to us. Nevertheless, the present theory represents an attempt to extract some dynamical information about this critical region by using the scaling and homogeneity principles.

We consider a time-dependent correlation function in the equilibrium ensemble,

$$\hat{C}_\xi^A(\vec{r},t) = \int\frac{d\omega}{2\pi}\int\frac{d^3k}{(2\pi)^3}e^{i(\vec{k}\cdot\vec{r}-\omega t)}C_\xi^A(\vec{k},\omega)$$

$$\equiv \langle\{A(\vec{r},t),A^\dagger(0,0)\}\rangle,$$

where A is some operator, such as the particle density, the spin density, the energy density, etc., and the curly bracket denotes an anticommutator. The subscript ξ specifies the temperature dependence which is of interest near T_c,[8] and the superscript A will sometimes be dropped. The function $C_\xi(\vec{k},\omega)$ can be rewritten, with no loss of generality, as

$$C_\xi(k,\omega) = (2\pi\omega_{k,\xi})^{-1}N_{k,\xi}f_{k,\xi}(\omega/\omega_{k,\xi}),$$

with

$$\int_{-\infty}^{\infty} f(x)dx = 1,$$

$$\int_{-1}^{1} f(x)dx = \tfrac{1}{2}.$$

This defines a normalization $N_{k,\xi}{}^A$ and a "width" $\omega_{k,\xi}{}^A$, which are, for the moment, arbitrary functions of K and ξ; the function $f_{k,\xi}{}^A$ depends parametrically on K and ξ. We shall only consider operators A such that $\omega_{k,\xi} \to 0$ when K and ξ^{-1} both go to 0. Since $N_{k,\xi}$ is the equal-time correlation function (proportional to a static susceptibility when $\omega_{k,\xi} \ll k_B T/\hbar$), the usual static scaling hypothesis is the statement that when K^{-1} and ξ are large compared with atomic lengths, (1) $N_{k,\xi}$ is a homogeneous function of K and ξ^{-1}.[9] We now make the following additional dynamic scaling assumptions: (2) $\omega_{k,\xi}$ is also a homogeneous function of k and ξ^{-1}. (3) The parametric dependence of f on k and ξ is a dependence on the product $k\xi$ only.

As an immediate consequence of the above assumptions it is seen that if the frequency dependence of the function $C_\xi(k,\omega)$ is measured for various values of k and ξ with a fixed product $k\xi$, then the various curves of C vs ω can be made to coincide, provided all the abscissas are scaled by some fixed power of k, and all the ordinates are likewise scaled by some power.

The further results of this paper may also be derived, however, if the scaling hypotheses (1)-(3) are replaced by a somewhat weaker statement in terms of the diagram in Fig. 1. It is assumed that the function $C_\xi^A(\vec{k},\omega)$ has different behavior in the three shaded limiting regions, and that the various asymptotic expressions merge on the boundaries $k\xi = \pm 1$. Implicit in this formulation is the notion that there is no dividing line between hydrodynamic and critical behavior other than that provided by the static criterion $k|\xi| \approx 1$.

We have applied the scaling theory to the isotropic Heisenberg antiferromagnet, which is a good model for $RbMnF_3$.[10] For the ordered phase we have developed a hydrodynamic description which is formally quite close to the two-fluid hydrodynamics of He II,[11] both in its derivation and in its predictions.[12] We assume that the equilibrium state of the antiferromagnet below T_c is completely characterized by the parameters energy, total magnetization ($\vec{S}$), and direction of staggered magnetization ($\vec{M}/M$), with the subsidiary condition $\vec{M}\cdot\vec{S}=0$. For departures from uniformity which are sufficiently slowly varying, we assume that the system will suffer only small deviations from local equilibrium, and that the time derivatives of the densities of the thermodynamic parameters may be expanded in powers of the gradient operator and of the magnitudes of the deviations from uniformity.[13] The result of this theory which is of interest here is the existence of a linear mode $\omega_k = ck$ in the spin-spin correlation function, at arbitrary temperatures

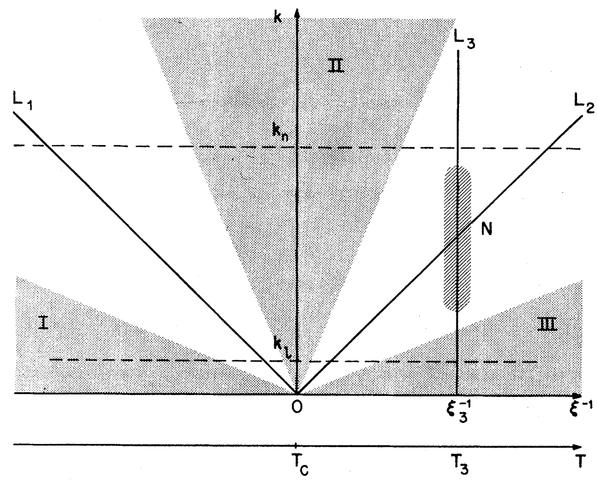

FIG. 1. A schematic plot of the wave number k versus the quantity ξ^{-1} which defines the temperature scale near T_c. The three asymptotic regions of differing dynamical behavior are (I) the hydrodynamic region below T_c [$\xi^{-1}<0, k|\xi| \ll 1$], (II) the critical region near T_c [$k|\xi| \gg 1$], (III) the hydrodynamic region above T_c [$\xi^{-1}>0$, $k\xi \ll 1$]. Both k^{-1} and ξ are large compared with atomic lengths. The asymptotic forms in Regions (I) and (II) are assumed to merge when extrapolated to the line L_1 ($k\xi=-1$), and similarly Regions (II) and (III) merge on L_2. In light-scattering experiments the wave numbers k_l are so small that hydrodynamics applies at most temperatures. Neutrons, on the other hand, probe much larger wave numbers, k_n (not drawn to scale), so that the hydrodynamic regions may only be reached with difficulty. The nmr linewidth depends on an integral over k (along L_3, say, at a fixed temperature T_3) which is dominated by contributions from region N ($k\xi \approx 1$).

below T_c. This hydrodynamic mode, which merges with the spin waves at low temperatures, is arbitrarily well defined at any temperature, in the long-wavelength limit, since the damping can be shown to be quadratic in k. The velocity is given by $c^2 = \rho_s/\chi$ where ρ_s is a stiffness constant and χ is the static susceptibility for uniform fields, which is believed to be finite at all temperatures. As in the helium case,[14] it can be shown[13] that $\rho_s \to 0$ as $|\xi|^{-1}$ when $T \to T_c^-$. If the direction of the sublattice magnetization is taken to be the z axis, then fluctuations in the x and y components of both the local staggered magnetization $\vec{M}(\vec{r})$ and the local total spin density $\vec{S}(\vec{r})$ are exhausted by the spin-wave mode in the $k \to 0$ limit. It follows, therefore, from this hydrodynamic analysis that for $T<T_c$ and $k|\xi| \ll 1$ (Region I of Fig. 1) the spin-spin correlation function $\langle\{M_x(\vec{r},t), M_x(0,0)\}\rangle$ has an f function which is just $f(x)$

$=\delta(x^2-1)$ and a characteristic energy $\omega_{k,\xi} = B|\xi|^{-1/2}k$, where B is a constant. From the scaling assumption (2) it follows that in the critical region ($k|\xi|\gg 1$, Region II of Fig. 1) the characteristic frequency for spin fluctuations is $\omega_k \sim B'k^{3/2}$. Note that we are not predicting the existence of spin waves at T_c, since this would only be true if $f(x)$ remained a δ function, whereas we do not know the form of f for $k|\xi|\gg 1$. Neither do we know the value of B', although presumably B/B' is of order unity.

In the hydrodynamic region above T_c ($k\xi \ll 1$, Region III of Fig. 1), the correlation function C^{M_x} for the staggered magnetization has a characteristic relaxation frequency $\Gamma_k = \omega_{k,\xi}$ which goes to a constant Γ_0 (independent of k) for $k\to 0$. By assumption (2) we thus find $\Gamma_0 \propto \xi^{-3/2}$, which is a manifestation of "critical slowing down" of spin fluctuations as $T\to T_c^+$. Similar arguments show that M_z has a relaxation rate proportional to $\xi^{-3/2}$ in Region I, while the spin-wave damping is proportional to $\xi^{1/2}k^2$ below T_c.

The correlation function $C_\xi^{S_x}(k,\omega)$ for the total spin density, on the other hand, has a characteristic spin-diffusion frequency $\omega_{k,\xi} = Dk^2$ in Region III, since the total spin is a conserved quantity. Below the transition, C^{S_x} also has a spin-wave spectrum for $k|\xi|\ll 1$, so that the scaling arguments yield $\omega_{k,\xi} \propto \xi^{1/2}k^2$ for this function in Region III. This implies an infinite spin-diffusion constant for $T\to T_c^+$ or a critical "speeding up"[15] of total spin fluctuations.[16]

The theory may also be applied to a determination of the temperature dependence of the nmr linewidth Δ, as $T\to T_c$. By substituting the scaling-law form of $C^M(k,\omega=0)$ into Eq. (3) of Heller,[17] we find, using the exponents of Ref. 4,

$$\Delta \propto \xi^{\frac{1}{2}-\eta} \propto |T-T_c|^{-\nu(\frac{1}{2}-\eta)} \approx |T-T_c|^{-\frac{1}{3}}.$$

The temperature dependence thus obtained is to be contrasted with the result $\Delta \propto (T-T_c)^{-1/2}$ found by Heller[17] using a molecular field theory.

The transition from hydrodynamic to critical behavior as the temperature approaches T_c should be apparent in neutron diffraction experiments. It is important to recognize, however, that the temperature at which this transition occurs depends entirely on the wavelength under consideration. For example, the critical slowing down of (staggered) spin fluctuations predicted above ($\Gamma_0 \sim \xi^{-3/2}$) only applies to the hydrodynamic region above T_c, and for any fixed k, the decrease of Γ_k with decreasing temperature will eventually be interrupted (at $k\xi\approx 1$). Very close to T_c ($k|\xi|\gg 1$), the characteristic frequency will be temperature independent ($\omega_k \sim k^{3/2}$).

In the case of the isotropic Heisenberg ferromagnet, the hydrodynamic theory[13] seems more complicated. The phenomenological theory of Landau and Lifshitz,[18] whose validity may be questionable at temperatures of order T_c, predicts spin waves in Region I with a real frequency λk^2, where $k = \rho_s/\langle S\rangle \propto \xi^{-1+\beta/\nu'} \approx |T-T_c|^{\frac{1}{3}}$. Here ρ_s is again a stiffness constant ($\rho_s \propto \xi^{-1}$) and $\langle S\rangle \propto (T_c-T)^\beta$ is the total magnetization. According to the scaling principle, the spin diffusion constant in Region III should also vary as $|T-T_c|^{1/3}$ near the Curie point.[19] At any fixed wave vector, of course, the temperature dependence will stop when $k\xi\approx 1$, and the characteristic frequency will vary as $k^{5/2}$ in the critical region.[20] The nmr linewidth is predicted to go as $|T-T_c|^{(2-\eta)\nu-\beta} \approx |T-T_c|$.

For the gas-liquid critical point the situation is complicated by the fact that sound propagation and thermal diffusion seem rather closely coupled, although some predictions do appear possible.[21] In any case, light-scattering experiments[22] are essentially confined to the hydrodynamic regions ($k|\xi|\ll 1$), so that the possibilities for direct experimental checks on the scaling theory are much more limited in the gas-liquid case, unless neutron-scattering experiments can be performed.

We are indebted to R. Nathans and M. Blume for useful discussions.

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*A preliminary version of this work was presented at the Seminar on Phase Transitions, Western Reserve University, June 1967.

[1]See, for instance, Critical Phenomena, Proceedings of a Conference, Washington, D. C., 1965, edited by M. S. Green and J. V. Sengers, National Bureau of Standards Miscellaneous Publication No. 273 (U. S. Government Printing Office, Washington, D. C., 1966).

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[7]"Long wavelengths" are long compared with "atomic" dimensions such as interparticle spacings or mean free paths, and "low frequencies" are small compared with typical collision frequencies as well as with $k_B T/\hbar$.

[8]We distinguish formally between $T < T_c$ and $T > T_c$ by taking $\xi < 0$ for $T < T_c$.

[9]This means it has the form $N_{k,\xi} = k^a g(k\xi)$.

[10]D. T. Teany, M. J. Freiser, and R. W. H. Stevenson, Phys. Rev. Letters 9, 212 (1962).

[11]I. M. Khalatnikov, Introduction to the Theory of Superfluidity (W. A. Benjamin, Inc., New York, 1965), Pt. II.

[12]More precisely, since there is not momentum conservation for the spin system, the antiferromagnet is analogous to superfluid helium in fine pores.

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[14]J. W. Kane and L. P. Kadanoff, Phys. Rev. 155, 80 (1967); J. A. Tyson and D. H. Douglass, Jr., Phys. Rev. Letters 17, 472, 622(E) (1966).

[15]"Kinematical" slowing down is still present because of the factor k^2 in $\omega_{k,\xi}$.

[16]Note that the magnitude of the fluctuations in $\vec{S}(\vec{r})$ is not divergent for the antiferromagnet. Identical arguments clearly do not go through in the ferromagnet.

[17]P. Heller, Ref. 1, p. 58.

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

[19]This behavior $[D \propto (T-T_c)^{\nu-\beta}]$ is more gradual than the Van Hove prediction, $D \propto (T-T_c)$ [L. Van Hove, Phys. Rev. 95, 1374 (1954)]. The exponent $\nu-\beta$ is equal, by the scaling laws of Refs. 2 and 3, to $\frac{1}{4}\gamma(1-\eta)(1-\frac{1}{2}\eta)^{-1}$, which is very close to the value $\frac{1}{4}\gamma$ proposed recently by K. Kawasaki (to be published).

[20]See also P. Resibois and M. DeLeener, Phys. Letters 25A, 65 (1965). W. Marshall [Ref. 1, p. 135] has discussed critical neutron scattering in the ferromagnet using an approximate renormalized spin-wave theory, and has predicted a modification of the low-temperature behavior at $T \approx 0.8T_c$. It is not clear to us whether this estimate refers only to a specific wave vector $\boldsymbol{k}$ typical of neutron experiments, or whether it holds independent of $\boldsymbol{k}$.

[21]L. P. Kadanoff, lecture delivered at the Seminar on Second Order Phase Transitions, Case Western Reserve University, 1967 (unpublished); L. P. Kadanoff and J. Swift, to be published.

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