Renormalization-group treatment of the critical dynamics of the binary-fluid and gas-liquid transitions

E. D. Siggia*
Department of Physics, Harvard University, Cambridge, Massachusetts 02138
and Bell Laboratories, Murray Hill, New Jersey 07974

B. I. Halperin
Bell Laboratories, Murray Hill, New Jersey 07974

P. C. Hohenberg
Bell Laboratories, Murray Hill, New Jersey 07974
and Physik Department, Technische Universität Munchen, 8046, Garching, West Germany
(Received 28 October 1975)

A simplified model for the dynamics of the gas-liquid and binary-fluid transitions is studied with the renormalization group. An exact scaling law is found, connecting the exponents for the diverging transport coefficients to static exponents for arbitrary value of the dimensionality $d < 4$. This scaling law had been anticipated by Kadanoff and Swift, and Kawasaki, on the basis of approximate mode-coupling arguments. The "Kawasaki-Stokes" relation between the diffusivity, the shear viscosity, and the correlation length is shown to hold exactly, but with a universal amplitude which differs slightly from its mode-coupling value. Critical exponents for the transport coefficients are evaluated to second order in $\epsilon = 4 - d$, and lead to the prediction of a weak divergence of the shear viscosity $[\bar{\eta}(T) \propto (T - T_c)^{-0.04}]$ in three dimensions. The weakness of this divergence reflects the existence of a small parameter in the theory, which explains the excellent agreement between Kawasaki's evaluation of the Rayleigh linewidth and experiment. Corrections to the simple Kawasaki theory carried out by various authors are reviewed, and a number of suggestions are made for refining these calculations. The simple model studied in this paper is shown to have the same dynamic properties as real fluids, sufficiently close to the critical point.

I. INTRODUCTION

The dynamical properties of binary-fluid and gas-liquid phase transitions have been studied extensively, both experimentally and theoretically, in recent years.[1] Of primary interest are the temperature dependence of the transport coefficients (e.g., the thermal conductivity or the shear viscosity) and the temperature and wave-vector dependence of the characteristic frequency of fluctuations of the order parameter, as measured, for instance, by inelastic light scattering ("Rayleigh linewidth"). These properties were calculated some years ago using the mode-coupling approach,[2,3] with results which were in remarkably good agreement with experiment,[4] as regards both the critical exponents and amplitudes and also the scaling function for the Rayleigh linewidth. Thus the critical dynamics of the gas-liquid and binary-fluid transitions is a case where a simple diagrammatic treatment yields accurate exponents and scaling functions in three dimensions ($d = 3$), which differ significantly from their mean-field or conventional values.

The renormalization-group method[5] provides a systematic means of proving scaling relations for general d, and of calculating exponents and scaling functions in expansions near $d = 4$. In the present paper, we apply this method to a simple model of the binary-fluid and gas-liquid transitions, for $T \geq T_c$, on the critical isochore.[6] We derive a number of exact scaling results which were in part anticipated by the mode-coupling[2,3] and dynamic-scaling[7] approaches. In addition, we find that some of the results of the Kawasaki theory,[3] in particular the numerical values of the critical exponents and scaling functions, are only approximately correct for any dimensionality. The success of the simple Kawasaki approximation,[8] which neglects the critical variation of the shear viscosity in lowest order, is traced to the existence of a small parameter, of order 0.05, in the theory. This parameter is of kinematic origin, and arises from the transverse nature of current fluctuations which couple to the order parameter.

A principal result of the present work is an exact scaling relation between the exponents x_λ and $x_{\bar{\eta}}$ for the transport coefficient λ of the order parameter (e.g., the thermal conductivity in a simple fluid) and the shear viscosity $\bar{\eta}$. This relation reads

$$x_\lambda + x_{\bar{\eta}} = \epsilon - \eta, \tag{1.1}$$

where $\epsilon \equiv 4 - d$ and η is the usual static exponent.[5] An alternative and physically more suggestive

statement of the above result, is in terms of the "Kawasaki-Stokes" relation,[3,9] which in three dimensions is

$$D \equiv \lambda/\chi_\psi = Rk_B T/\bar{\eta}\xi, \tag{1.2}$$

where χ_ψ is the static susceptibility of the order parameter ($\chi_\psi \propto C_p$ in a pure fluid) and ξ is the static correlation length. Equation (1.1) is an exact result, which differs from Kawasaki's original relation[3,8] only in that the value of the *universal* constant R is estimated from the ϵ expansion to be $R = 0.064$ rather than $(6\pi)^{-1} = 0.053$. Note that according to Eqs. (1.1) and (1.2), it is only the combination $\lambda\bar{\eta}$ whose exponent is determined by an exact scaling law, and not the transport coefficients λ and $\bar{\eta}$ separately. The individual exponents x_λ and $x_{\bar{\eta}}$ may also be obtained in an ϵ expansion,[6] and we find, to second order,

$$x_\lambda = \tfrac{18}{19}\epsilon[1 - 0.033\epsilon + O(\epsilon^2)] \simeq 0.916, \tag{1.3a}$$

$$x_{\bar{\eta}} = \tfrac{1}{19}\epsilon[1 + 0.238\epsilon + O(\epsilon^2)] \simeq 0.065, \tag{1.3b}$$

which satisfies Eq. (1.1) since $\eta = \frac{1}{54}\epsilon^2 + O(\epsilon^3)$. The ratio $x_{\bar{\eta}}/x_\lambda$ is the small parameter referred to above, which is responsible for the unusually good accuracy of the earlier mode-coupling calculation.[3]

Another exact result of our work is the dynamic scaling relation[7,10]

$$\omega_\psi(k) = Dk^2\bar{\Omega}(k\xi) = D_0\xi^{2-z}k^2\bar{\Omega}(k\xi) \tag{1.4}$$

for the characteristic frequency of the order parameter, valid in the limit $\xi \to \infty$, $k \to 0$, $k\xi$ arbitrary [the function $\bar{\Omega}(x)$ is defined so that $\bar{\Omega}(0) = 1$]. It must be noted, however, that since the order-parameter correlation function is non-Lorenztian in the "critical region" $k\xi \gg 1$, the scaling function $\bar{\Omega}(k\xi)$ will depend on the precise definition chosen for the characteristic frequency $\omega_\psi(k)$. On the other hand, any reasonable definition of $\omega_\psi(k)$ will lead to the form (1.4), with an appropriate function $\bar{\Omega}(k\xi)$. From the preceding discussion it also follows that the critical exponent z is not precisely equal to 3 as in the Kawasaki theory,[3] but rather to $z = 4 - \eta - x_\lambda$; our estimate based on Eq. (1.3) yields $z = 3.065$ when extrapolated to $d = 3$.

Many features of the above results were in fact already contained in the various improved mode-coupling calculations,[10-14] which appeared after the original work of Kadanoff and Swift[2] and of Kawasaki.[3,8] In particular, the self-consistent approximation of Kawasaki[3,11] satisfies Eqs. (1.1) and (1.4) (with $\eta = 0$), and can yield estimates of the parameters in those equations, including the exponents x_λ and $x_{\bar{\eta}}$. Our examination of the ϵ expansion to second order leads us to expect that an accurate solution of these self-consistent equations should yield parameters which are correct to better than 5% in three dimensions.

The simple model of a fluid employed in most treatments of the critical dynamics[2,3] omits a number of features of real fluids, such as the longitudinal current and pressure fluctuations and the asymmetry of the free energy with respect to a change in sign of the order parameter. These properties may be studied using the renormalization group,[15,16] and it is confirmed that they are irrelevant, asymptotically close to the critical point. Thus, provided the ϵ expansion is a reliable guide to the behavior for $d = 3$, the scaling relations found for the simple model should also apply exactly to real fluids. Moreover, a concrete result of our study of more realistic models of the binary fluid is the statement that the transport coefficient Dk_T should remain finite at T_c, in agreement with the conclusions of Swift[17] and others[18] which had been questioned by Papoular.[19]

In Sec. II our model for the critical dynamics of a fluid is defined,[6] and the lowest-order recursion relations are discussed in detail. Generalizations to higher orders are then considered, in order to prove the scaling relation (1.1) to all orders in ϵ and to calculate the second-order values (1.3). Details of the computations are given in the Appendix. Section III justifies the application of our simplified model to the binary-fluid and gas-liquid transitions. Thermodynamics and linearized hydrodynamics establish the connection between the physical parameters of real fluids and those of our model. It is also argued that the simplified model provides a complete description of the critical point. In Sec. IV we compare our results with previous work, and comment on the experimental situation.

II. ANALYSIS OF A SIMPLE MODEL

The model we consider is very close to the one introduced earlier in the mode-coupling theories.[2,3,17] It was denoted as model H in Refs. 20 and 21, and is defined by the following equations, in d dimensions:

$$W = \int d^d x\,[\tfrac{1}{2}r_0\psi^2 + \tfrac{1}{2}(\nabla\psi)^2 + u_0\psi^4 + \tfrac{1}{2}\vec{j}^2], \tag{2.1a}$$

$$\frac{\partial\psi}{\partial t} = \lambda_0\nabla^2\frac{\delta W}{\delta\psi} - g_0\vec{\nabla}\psi\cdot\frac{\delta W}{\delta\vec{j}} + \theta, \tag{2.1b}$$

$$\frac{\partial\vec{j}}{\partial t} = \mathcal{T}\left(\bar{\eta}_0\nabla^2\frac{\delta W}{\delta\vec{j}} + g_0\vec{\nabla}\psi\frac{\delta W}{\delta\psi} + \vec{\zeta}\right), \tag{2.1c}$$

$$\langle\theta(xt)\theta(x't')\rangle = -2\lambda_0\nabla^2\delta(x-x')\delta(t-t'), \tag{2.1d}$$

$$\langle\zeta_\alpha(xt)\zeta_\beta(x't')\rangle = -2\bar{\eta}_0\nabla^2\delta(x-x')\delta(t-t')\delta_{\alpha\beta}. \tag{2.1e}$$

The scalar field ψ is the order parameter, which represents the deviations of the concentration or density from its critical value. The transverse vector field $\vec{j}$ in d dimensions corresponds to the momentum density or velocity in an incompressible fluid. The matrix $\mathcal{T}$ is a projection operator which selects the transverse part of the vector in brackets ($\mathcal{T}_k^{\alpha\beta} = \delta_{\alpha\beta} - k_\alpha k_\beta / k^2$), and θ and ζ are Gaussian noise sources. The background or bare viscosity and the bare order-parameter transport coefficient are given by $\bar{\eta}_0$ and λ_0, respectively. The parameters in the thermodynamic potential W have their usual meaning, and we have dropped certain unnecessary constants. A value of g_0 different from unity has been allowed for, since we will define scale changes that leave the correlation functions invariant but cause trivial rescalings of g_0. Our model contains both dissipative and reversible interactions. The former arise from quartic terms in the effective enthalpy for the order parameter [Eq. (2.1a)], with the coupling constant u_0. The effects of dissipative couplings on critical dynamics were first studied with the renormalization group by Halperin, Hohenberg, and Ma.[22] The reversible interactions (proportional to g_0) are needed to make the theory Galilean invariant. It may be seen from Eqs. (2.1a)–(2.1e) that there is a dimensionless bare coupling constant f_0 for the nondissipative interactions,

$$f_0 = K_d g_0^2 \Lambda^{d-4} / \lambda_0 \bar{\eta}_0. \tag{2.2}$$

We use Λ to denote the ultraviolet cutoff and define K_d as the geometric factor[5] $(2\pi)^{-d} 2\pi^{d/2} \times [\Gamma(\frac{1}{2}d)]^{-1}$; note also that W is here defined to be dimensionless (i.e., $k_B T_c = 1$). Because $\vec{j}$ is transverse, ψ is conserved. The equilibrium distribution of ψ and $\vec{j}$ is proportional to e^{-W} and the static critical properties of ψ are clearly the same as in the Ginzburg-Landau-Wilson model.[5]

We define a renormalization group on the equations of motion by thinning the degrees of freedom and rescaling,[20,22] with scale factors chosen to preserve the form of the correlation functions at T_c. The scaling transformation is

$$x \to x' = x/b, \tag{2.3a}$$

$$\omega \to \omega' = b^z \omega, \tag{2.3b}$$

$$\psi \to \psi' = b^{(d-2+\eta)/2} \psi, \tag{2.3c}$$

$$j \to j' = b^{d/2} j. \tag{2.3d}$$

The recursion formulas for the static parameters r_l and u_l are the same as found by Wilson and Fisher.[23] Dynamic effects, in particular the renormalization of λ and $\bar{\eta}$, are calculated by means of the Martin, Siggia, and Rose[24] formalism which is summarized in the Appendix. For dimension $d = 4 - \epsilon$, we find a fixed point at which the vertex f_∞ is of order ϵ. (Above four dimensions the Van Hove theory[25,26] is correct.) We can then order the perturbation expansion in powers of the small parameter f_0 and show that it is the only dimensionless coupling constant required to calculate the critical contributions to the transport coefficients to order ϵ.

At sufficiently high order in ϵ, the dissipative coupling of the order parameter to itself via the vertex u_0 in Eq. (2.1) will also affect the critical exponents and scaling functions. The lowest-order effect of u_0, which influences exponents in order ϵ^2, may be included in the mode-coupling formalism of Kawasaki[3] by using the correct static susceptibility $\chi_\psi(k)$ rather than the Ornstein-Zernike form. In our present calculations, these effects are included when we insert the value of the static exponent η, to order ϵ^2, in Eq. (A25) of the Appendix. It should be noted, however, that there are additional affects of u_0 which are not included in the Kawasaki formalism.[3] These first occur in the exponents at order ϵ^3 and in the Rayleigh lineshape for large $k\xi$ at order ϵ^2, orders which are not reached in the explicit calculations of the present paper.

By applying the rescalings to the equations of motion and calculating the simplest mode-coupling diagrams [Figs. 1(a) and 2(a)] in four dimensions, we find

$$\lambda_{l+1} = b^{z+\eta-4} \lambda_l (1 + \tfrac{3}{4} f_l \ln b), \tag{2.4a}$$

$$\bar{\eta}_{l+1} = b^{z-2} \bar{\eta}_l (1 + \tfrac{1}{24} f_l \ln b), \tag{2.4b}$$

$$g_{l+1} = b^{z-3+\epsilon/2} g_l. \tag{2.4c}$$

We have retained the static exponent η in the scaling factors although it is of higher order in ϵ. The equation for f_l follows:

$$f_{l+1} = b^{\epsilon-\eta} f_l (1 - \tfrac{19}{24} f_l \ln b). \tag{2.4d}$$

We see that f is a relevant variable below four dimensions, and that a stable fixed point exists with $f_\infty = \frac{24}{19}\epsilon + O(\epsilon^2)$.

After iterating the recursion formulas L times, where $L = \ln(\xi\Lambda)/\ln b$, the effective correlation length is of order unity and further renormalizations are regular. It follows that the physical temperature-dependent transport coefficients $\lambda(T)$ and $\bar{\eta}(T)$ are

$$\lambda(T) \approx \lambda_L (\xi\Lambda)^{4-\eta-z}, \tag{2.5}$$

$$\bar{\eta}(T) \approx \bar{\eta}_L (\xi\Lambda)^{2-z}. \tag{2.6}$$

(These equations are exact to lowest order in ϵ. More generally, these relations are correct up to a multiplicative constant of order unity, pro-

vided the exact renormalization group is used for λ_l and $\bar{\eta}_l$.[20] As $\xi \to \infty$ we define

$$\lambda(T) \simeq \xi^{x_\lambda}, \tag{2.7}$$

$$\bar{\eta}(T) \simeq \xi^{x_{\bar{\eta}}}. \tag{2.8}$$

If we choose z such that λ_l approaches a finite fixed point, then z is the exponent of the characteristic frequency of the order-parameter relaxation. From Eq. (2.5) we have

$$z = 4 - \eta - x_\lambda, \tag{2.9}$$

$$x_\lambda = \tfrac{18}{19}\epsilon + O(\epsilon^2). \tag{2.10}$$

Similarly we find

$$x_{\bar{\eta}} = \tfrac{1}{19}\epsilon + O(\epsilon^2). \tag{2.11}$$

Note that $\bar{\eta}_l$ and g_l do *not* approach finite constants as $l \to \infty$, if we adjust the frequency scale to make λ_∞ finite. The dimensionless coupling constant f_l does remain finite, however. The frequency for momentum relaxation scales as $k^{2-x_{\bar{\eta}}}$, which is much faster than k^z [cf. Eqs. (2.9) and (2.11)].

A. Analysis of higher-order terms

The analysis of higher-order terms is very similar to that outlined in Ref. 20. First of all, the fact that the recursion relation for g_l contains only the trivial scaling factor $b^{z-3+\epsilon/2}$ is not a coincidence. The absence of renormalizations of the convective vertex arising from the interactions is a consequence of Galilean invariance, and is correct to all orders in ϵ (see the discussion of the corresponding interaction vertex in the models of Ref. 20). We will show in the Appendix that all higher-order contributions to Eq. (2.4d) can be expressed as a power series in f_l and u_l with finite coefficients, so that the vertex f_l approaches a finite fixed-point value

$$f_\infty = \tfrac{24}{19}\epsilon + O(\epsilon^2). \tag{2.12}$$

[The vertex u_l approaches its static value $u_\infty = \epsilon/4K_4(n+8) + O(\epsilon^2)$.] From Eq. (2.4d) it follows that the physical temperature-dependent vertex $f(T) \equiv K_d g_0^2 \Lambda^{-\epsilon}/\bar{\eta}(T)\lambda(T)$ varies as

$$f(T) \sim f_\infty(1 + a_1\epsilon)(\Lambda\xi)^{\eta-\epsilon}, \tag{2.13}$$

where a_1 is a universal constant of order unity. Using Eqs. (2.7), (2.8), and (2.13), we thus find the exact scaling law

$$x_\lambda + x_{\bar{\eta}} = \epsilon - \eta. \tag{2.14}$$

B. Universal amplitudes and scaling functions

Associated with the exponent relation (2.14) there is also a universal amplitude ratio[20]

$$R \equiv g_0^{-2}\lambda(T)\bar{\eta}(T)\chi_\psi^{-1}(T)\xi^{d-2}, \tag{2.15}$$

or

$$R = (k_B T_c)^{-1}\lambda(T)\bar{\eta}(T)\chi_\psi^{-1}(T)\xi^{d-2}, \tag{2.16}$$

in the more usual units where $g_0 = 1$ and the free energy W has units of $k_B T$. Equation (2.16) may be rewritten in the Kawasaki-Stokes form[3,9] in terms of the diffusion constant

$$D = \lambda\chi_\psi^{-1} \tag{2.17}$$

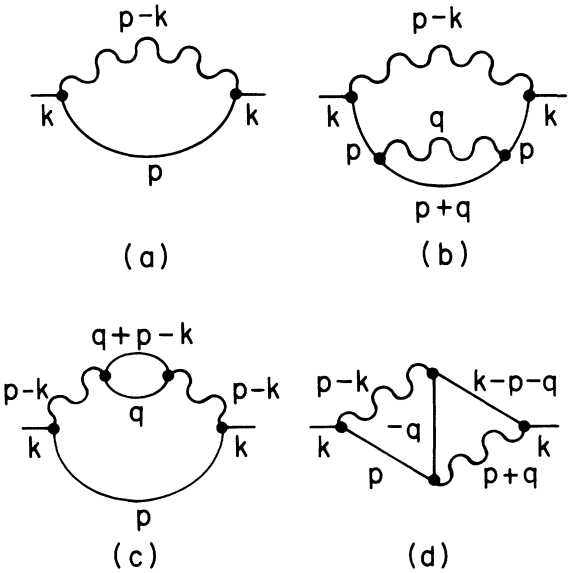

FIG. 1. Self-energy Σ_2 that renormalizes λ. The wavy lines represent momentum propagators and the solid lines denote the order parameter. The vertex g is a dot.

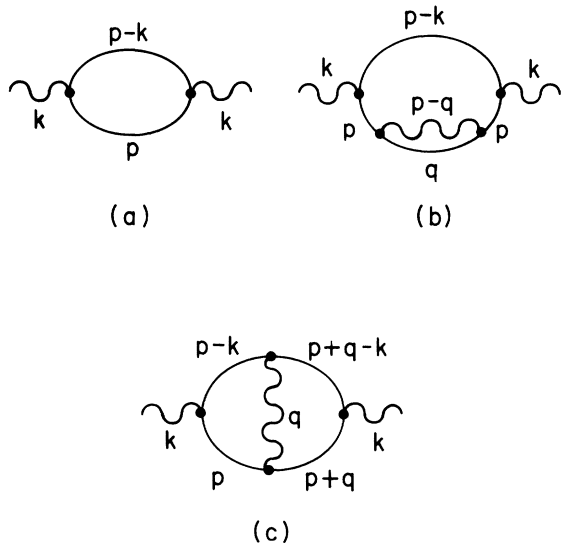

FIG. 2. Self-energy Π_2 renormalizing η. The lines have the same meaning as in Fig. 1.

as

$$D = Rk_B T_c/\overline{\eta}\xi^{d-2}. \tag{2.18}$$

From the second-order calculation described in the Appendix we find

$$R = K_d \tfrac{19}{24}\epsilon^{-1}[1 + 0.06\epsilon + O(\epsilon^2)]. \tag{2.19}$$

If we simply set $d=3$ and $\epsilon = 1$ we obtain

$$R = 0.042 = 0.79(6\pi)^{-1}. \tag{2.20}$$

In Sec. IV we shall discuss a different method of extrapolating Eq. (2.19), which we consider to be more reliable and which yields the value $R = 0.064 = 1.2(6\pi)^{-1}$ for $d=3$, quoted in Sec. I.

In obtaining both the above result and the next term in the ϵ expansion of the critical exponents we have used Wilson's Feynman-graph expansion method,[27] rather than the recursion relations. The calculation is outlined in the Appendix. The main results are the exponent and amplitude values in Eqs. (1.3) and (2.19), and the expression for the order-parameter decay rate,

$$\omega_\psi(k,\xi) = (Rk_B T/\overline{\eta}(T)\xi^{2-\epsilon})k^2\overline{\Omega}(k\xi) \equiv k^z\Omega(k\xi), \tag{2.21}$$

$$\overline{\Omega}(x) = (1+x^2)\left[1 + \frac{24\epsilon}{19K_4}\int \frac{d^4p}{(2\pi)^4}\left(\frac{\hat{x}\cdot\mathcal{T}_p\cdot\hat{x}}{p^2[1+(\vec{x}+\vec{p})^2]} - \frac{\frac{3}{4}}{p^2(1+p^2)}\right)\right]. \tag{2.22}$$

We shall compare these results with previous work in Sec. IV.

From the calculated scaling function we may obtain another interesting universal amplitude ratio, namely,

$$\tilde{R} \equiv [\omega_\psi(k, \xi=\infty)/\omega_\psi(k=0,\xi)](k\xi)^{2-z}. \tag{2.23}$$

The calculation of $\tilde{R}$ using Eqs. (2.21) and (2.22) is straightforward, and we find

$$\tilde{R} = 1 + \tfrac{9}{38}\epsilon + O(\epsilon^2). \tag{2.24}$$

As was mentioned in Sec. I the definition of the characteristic frequency $\omega_\psi(k,\xi)$ is not unique for $k\xi \gg 1$, owing to the non-Lorentzian form of the correlation function. The definition which is most convenient computationally is in terms of the low-frequency response function $\chi_\psi(k,\omega)$,

$$\omega_\psi(k,\xi) \equiv \lambda(k,\xi)k^2/\chi_\psi(k), \tag{2.25}$$

$$\lambda^{-1}(k,\xi) \equiv k^2 \lim_{\omega\to 0}\frac{\partial\chi_\psi^{-1}(k,\omega)}{-i\partial\omega}, \tag{2.26}$$

where $\chi_\psi(k) \equiv \chi_\psi(k,\omega=0)$. Alternatively, one can attempt to fit the frequency dependence of the correlation function to a Lorentzian and define $\omega_\psi(k,\xi)$ as the appropriate half-width, or employ the definition of Ref. 7 in terms of the median frequency of the correlation function. The latter methods are more difficult to calculate, but correspond more closely to an experimental determination of ω_ψ. It may be shown, however, that to zeroth and first order in ϵ, the correlation function is a Lorentzian for any value of $k\xi$, so that Eqs. (2.25) and (2.26) are correct for any of the above definitions.

III. REAL FLUIDS

In order to compare the results of Sec. II to experiment, it is necessary to establish the precise correspondence between the parameters of the simple model and those of a real fluid, and also to show that the features which have been omitted from the model are irrelevant near the critical point.

The choice of thermodynamic variables in a fluid requires some care, in order that their static fluctuations be independent.[28] For the liquid-gas transition in a pure fluid, it is convenient to use as the order parameter a quantity $q(x)$, which is equal to the fluctuations of the entropy per unit mass, s, times the critical density ρ_c times the temperature T_c, i.e.,

$$q(x) = [\epsilon(x) - (\overline{\mu} + T_c\overline{s})\rho(x)] \equiv \rho_c T_c s(x), \tag{3.1}$$

where $\epsilon(x)$ and $\rho(x)$ are the fluctuations in the energy density and the mass density, and $\overline{s}$ and $\overline{\mu}$ are the equilibrium entropy and chemical potential (per unit mass), respectively. Thus $q(x)$ corresponds to $\psi(x)$ in Eq. (2.1). For the vector $\vec{j}(x)$ we use the momentum density of the fluid, also denoted $\vec{j}(x)$. The fluctuations in the pressure are not thermodynamically coupled to the other variables under these definitions, and appear only through the sound waves in the dynamics. The sound-wave frequency is always much faster than the diffusive modes,[2,3] and is irrelevant at the critical point. We will systematically neglect both the pressure and the longitudinal component of the momentum density. The critical temperature is taken as unity.

At equilibrium near the gas-liquid transition, the probability of fluctuations $q(r)$ and $\vec{j}(r)$, at wavelengths long compared to the coherence length, is given by

$$\mathcal{P} = e^{-W}, \tag{3.2}$$

$$W = \frac{1}{2}\int d^d x \left(\rho_c^{-1}\left.\frac{\partial T}{\partial s}\right|_\rho q^2 + \rho_c^{-1}\vec{j}^2\right), \tag{3.3}$$

where W represents the enthalpy, and the specific heat $C_p = \rho_c (\partial s/\partial T)_p$ plays the role of the static susceptibility r^{-1}. (We have returned to units where $k_B T_c = 1$.) The linearized hydrodynamic equations are

$$\frac{\partial q}{\partial t} = \kappa \rho_c^{-1} \nabla^2 T \equiv \kappa \nabla^2 \frac{\delta W}{\delta q}, \tag{3.4}$$

$$\frac{\partial \vec{j}}{\partial t} = \bar{\eta} \rho_c^{-1} \nabla^2 \vec{j} = \bar{\eta} \nabla^2 \frac{\delta W}{\delta \vec{j}}. \tag{3.5}$$

They define the transport coefficients κ (thermal conductivity) and $\bar{\eta}$ (viscosity).

As is well known,[5] in order to use the renormalization group to calculate *static* properties, it is necessary to supplement the Gaussian free energy (3.3) with nonlinear and gradient terms which describe the interactions of fluctuations with wave vectors of the order of ξ. Similarly, we must add appropriate coupling terms to the hydrodynamic equations (3.4) and (3.5) in order to obtain the dynamic critical behavior. It is of course important that the dynamic coupling be chosen in such a way as to preserve the important conservation laws of the fluid, just as the static couplings are chosen to preserve the relevant symmetries. The system of equations (2.1), with the variables interpreted as noted above, are the simplest equations which preserve Galilean invariance and the conservation of q, appropriate to a simple fluid. In principle, we must allow for all possible couplings and nonlocal effects, consistent with the symmetries and conservation laws of the fluid, but it is rather easy, at least within an ϵ expansion, to show that these additional terms are irrelevant near T_c.

As an example, let us consider a free energy which is asymmetric with respect to a change of sign of the order parameter,

$$W_a = \int d^d x \left[\tfrac{1}{2} r_0 \psi^2 + \tfrac{1}{2} (\nabla \psi)^2 + v_0 \psi^3 + u_0 \psi^4 + \tfrac{1}{2} \rho_0^{-1} \vec{j}^2 + \Delta_0 \psi \vec{j}^2 - \mu_0 \psi \right]. \tag{3.6}$$

It has been shown[15] within the ϵ-expansion near $d = 4$, and also[16] using Wilson's approximate recursion relations for $d = 3$ (Ref. 29), that the freedom to adjust the additional parameter μ_0 to achieve criticality permits one to shift ψ in such a way as to eliminate v_0. The static critical properties are those of the symmetric model, since Δ_0 is an irrelevant field, and it is clear that the same conclusion holds for the dynamics, when W_a is inserted into (2.1) in place of W. (Of course, our considerations are valid only so long as the ϵ expansion or the approximate recursion relations are reliable guides to the behavior of a real fluid in three dimensions, and it is not definitely established that the asymmetry is irrelevant in the latter case.)

For the binary fluid we choose the fluctuations of the concentration $c(x)$ as the order parameter, and the total momentum density as $\vec{j}$. There is now another scalar field $s(x)$, which has diffusive behavior; the correct linear combination of $s(x)$ and $c(x)$ whose static fluctuations are orthogonal to those of $c(x)$ is[17,28]

$$q(x) \equiv \rho_c s(x) + \rho_c \left(\frac{\partial \mu}{\partial T} \right)_{c,p} c(x). \tag{3.7}$$

The long-wavelength static fluctuations are described by the enthalpy

$$W = \frac{1}{2} \int d^d x \left(\rho_c^{-1} \frac{\partial T}{\partial s}\bigg|_{c,p} q^2 + \rho_c \frac{\partial \mu}{\partial c}\bigg|_{T,p} c^2 + \rho_c^{-1} \vec{j}^2 \right). \tag{3.8}$$

Note that the fluctuations in the chemical potential and the temperature are

$$\mu(r) = \rho_c^{-1} \frac{\delta W}{\delta c(r)}, \quad T(r) = \frac{\delta W}{\delta q(r)}. \tag{3.9}$$

Following Landau and Lifshitz[28] we define transport coefficients from the equations of motion

$$\frac{\partial c}{\partial t} = \alpha \rho_c^{-1} \nabla^2 \mu + \beta \rho_c^{-1} \nabla^2 T \tag{3.10a}$$

$$= \alpha \rho_c^{-2} \nabla^2 \frac{\delta W}{\delta c} + \alpha k_T \rho_c^{-1} \frac{\partial \mu}{\partial c}\bigg|_{T,p} \nabla^2 \frac{\delta W}{\delta q}, \tag{3.10b}$$

$$\frac{\partial q}{\partial t} = \left(\beta + \alpha \frac{\partial \mu}{\partial T}\bigg|_{c,p} \right) \nabla^2 \mu + \left(\gamma + \beta \frac{\partial \mu}{\partial T}\bigg|_{c,p} \right) \nabla^2 T \tag{3.11a}$$

$$\equiv \left(\kappa + \alpha k_T^2 \frac{\partial \mu}{\partial c}\bigg|_{T,p}^2 \right) \nabla^2 \frac{\delta W}{\delta q} + \left(\alpha k_T \rho_c^{-1} \frac{\partial \mu}{\partial c}\bigg|_{T,p} \right) \nabla^2 \frac{\delta W}{\delta c}, \tag{3.11b}$$

$$\frac{\partial \vec{j}}{\partial t} = \bar{\eta} \rho_c^{-1} \nabla^2 \vec{j} = \bar{\eta} \nabla^2 \frac{\delta W}{\delta \vec{j}}. \tag{3.12}$$

As in the case of the simple fluid, we assume in all of the above equations that the pressure and longitudinal current are held constant, an assumption which is valid for variation on a time scale long compared to the sound-wave period.

The introduction of nonlinear couplings into Eqs. (3.10)–(3.12) is complicated by the existence of the extra field $q(x)$, in addition to $\psi(x) \equiv c(x)$ and

$\vec{j}(x)$. If we assume that q is decoupled from c and $\vec{j}$, we again obtain Eqs. (2.1) as the simplest nonlinear model consistent with the conservation laws of a binary fluid. In order to study the possible effect of the field $q(x)$, we have examined a three-field model (which one might denote as H', in the notation of Ref. 20). Its defining equations are

$$F = \int d^d x \left[\tfrac{1}{2} r_0 \psi^2 + \tfrac{1}{2} (\nabla \psi)^2 + u_0 \psi^4 + \tfrac{1}{2} \chi_0^{-1} q^2 + \gamma_0 q \psi^2 + \tfrac{1}{2} \rho_0^{-1} \vec{j}^2 - Tq \right], \tag{3.13}$$

$$\frac{\partial \psi}{\partial t} = \lambda_0 \nabla^2 \frac{\delta F}{\delta \psi} + L_0 \nabla^2 \frac{\delta F}{\delta q} - g_0 \vec{\nabla} \psi \cdot \frac{\delta F}{\delta \vec{j}} + \theta, \tag{3.14}$$

$$\frac{\partial q}{\partial t} = K_0 \nabla^2 \frac{\delta F}{\delta q} + L_0 \nabla^2 \frac{\delta F}{\delta \psi} - g_0 \vec{\nabla} q \cdot \frac{\delta F}{\delta \vec{j}} + \varphi, \tag{3.15}$$

$$\frac{\partial \vec{j}}{\partial t} = \mathcal{T} \left(\bar{\eta}_0 \nabla^2 \frac{\delta F}{\delta \vec{j}} + g_0 \vec{\nabla} \psi \frac{\delta F}{\delta \psi} + g_0 \vec{\nabla} q \frac{\delta F}{\delta q} + \vec{\zeta} \right). \tag{3.16}$$

The noise sources θ and φ satisfy

$$\langle \theta(1)\, \theta(1') \rangle = -2\lambda_0 \nabla^2 \delta(1 - 1'), \tag{3.17a}$$

$$\langle \varphi(1)\, \varphi(1') \rangle = -2K_0 \nabla^2 \delta(1 - 1'), \tag{3.17b}$$

$$\langle \varphi(1)\, \theta(1') \rangle = -2L_0 \nabla^2 \delta(1 - 1'), \tag{3.17c}$$

$$\langle \zeta_\alpha(1)\, \zeta_\beta(1') \rangle = -2\bar{\eta}_0 \nabla^2 \delta(1 - 1')\, \delta_{\alpha\beta}. \tag{3.17d}$$

From a comparison of the linearized form of (3.13)–(3.16) with Eqs. (3.10)–(3.12), we may identify the dressed (or physical) transport coefficients as

$$\lambda = \alpha \rho_c^{-2}, \tag{3.18}$$

$$L = \alpha k_T \rho_c^{-1} \frac{\partial \mu}{\partial c}\bigg|_{T,\rho}, \tag{3.19}$$

$$K = \kappa + \alpha k_T^2 \frac{\partial \mu}{\partial c}\bigg|_{T,\rho}^2, \tag{3.20}$$

while $\bar{\eta}$ is the same in both systems. Note that the free energy F may be chosen to be symmetric in ψ, since any asymmetry is shown to be irrelevant by the same argument as for the simple fluid.

If g_0 is neglected, Eqs. (3.13)–(3.17) are just those defining model D of Ref. 22, except that the terms proportional to L_0 have been added. It was found for model D, and is equally true here, that there is no renormalization of the transport coefficients by the dissipative interactions. The diagramatic rules of the appendix may be generalized to Eqs. (3.13)–(3.17) with a convective coupling. One finds, at least for $L_0^2 \ll \lambda_0 K_0$, that there are no critical renormalizations of the transport coefficients other than those already calculated in the absence of q and L_0. The type of diagram which renormalizes λ_0 vanishes identically for K_0 and L_0. Specifically, L is finite at T_c, which implies that $\alpha k_T (\partial \mu / \partial c)_{T,\rho}$ is finite [Eq. (3.19)]. This quantity was denoted as DK_T by Swift,[17] who reached conclusions similar to our own. We therefore disagree with the assertions of Papoular,[19] which contradicted Swift. The transport coefficients λ and $\bar{\eta}$ diverge with the same exponents x_λ and $x_{\bar{\eta}}$ as in the absence of q. Finally, we find that the true thermal conductivity $\kappa = K - L^2/\lambda$ [see Eq. (3.20)] remains finite at T_c, though with a weak cusp from the divergence of λ.

IV. DISCUSSION AND CONCLUSIONS

The present paper contains the exact ϵ expansion for the critical dynamics of fluids, carried out to second order for the critical exponents and for certain amplitudes. In order to assess the relevance of the results for real systems, it is useful to compare them with previous approximate three-dimensional mode-coupling calculations, which have been quite successful in describing experiments.[1]

The simplest approximation, that of Kadanoff and Swift[2] and especially of Kawasaki,[3] neglects completely the critical behavior of the viscosity and leads to the following expression for $\lambda(k, \xi)$ (in units where $g_0 = 1$, but $k_B T \neq 1$):

$$\lambda(k, \xi) = \frac{k_B T}{\bar{\eta}} \int \frac{d^d p}{(2\pi)^d} \left(\frac{\hat{k} \cdot \mathcal{T}_p \cdot \hat{k}}{p^2 [\xi^{-2} + (p - k)^2]} \right). \tag{4.1}$$

The exponents are therefore

$$x_\lambda = \epsilon, \quad x_{\bar{\eta}} = 0, \tag{4.2}$$

and they satisfy Eq. (2.14) with $\eta = 0$. Within this approximation we may define the characteristic frequency

$$\omega_\psi(k) \equiv \lambda(k, \xi) k^2 / \chi_\psi(k) = (R_K k_B T / \bar{\eta} \xi)\, k^2 \bar{\Omega}_K(k\xi) \tag{4.3}$$

and evaluate the quantities R_K and $\bar{\Omega}_K$ for $d = 3$, with $\chi_\psi(k) = (k^2 + \xi^{-2})^{-1}$. The result is[3]

$$R_K = (6\pi)^{-1}, \tag{4.4}$$

$$\bar{\Omega}_K(x) \equiv x^{-2} K_0(x) = \tfrac{3}{4} x^{-2} [1 + x^2 + (x^3 - x^{-1}) \arctan x], \tag{4.5}$$

which is the original "simple" Kawasaki approximation.[3,8] From Eqs. (4.3) and (4.5) we see that the corresponding approximate value of the coefficients $\tilde{R}$, defined in Eq. (2.23), is

$$\tilde{R}_K = \tfrac{3}{8}\pi. \tag{4.6}$$

In d dimensions Eqs. (4.1) and (4.3) for $k = 0$ yield

$$R_K = \pi^{(1-d)/2}\Gamma^2\left(\frac{1+d}{2}\right)\left[\Gamma\left(\frac{d-1}{2}\right)\Gamma(1+d)\times\sin\left(\frac{\pi}{2}(d-4)\right)\right]^{-1}, \quad (4.7)$$

which has the ϵ expansion

$$R_K = K_d(\tfrac{19}{24}\epsilon^{-1})^{\frac{18}{19}}\,(1-\tfrac{1}{12}\epsilon). \quad (4.8)$$

Comparing with the exact expansion (2.19), we see that the simple Kawasaki approximation is incorrect even in the lowest order in ϵ, but only by the factor $\frac{18}{19}$.

It has been remarked by several authors[3,9] that the Kawasaki-Stokes expression (1.2) with R given by (4.4) corresponds to the Brownian-motion formula for the diffusion constant of a sphere of radius ξ. From our general expression (2.18) it may be seen that a similar result holds for arbitrary dimensionality d, but that the radius of the sphere is precisely equal to ξ only for $d=3$ and in the simple Kawasaki approximation (4.4). More generally the radius is only proportional to ξ; indeed, near four dimensions the Stokes law for a sphere of radius r may be shown to yield $D = \frac{3}{16}k_BT/\pi^2\bar{\eta}r^2$, whereas the exact result (2.18), (2.19) is $D = 19k_BT/192\pi^2\epsilon\bar{\eta}\xi^2$, so we have $r = (6/\sqrt{19})\epsilon^{1/2}\xi = 1.38\epsilon^{1/2}\xi$.

An improved approximation is the *self-consistent* scheme of Kawasaki,[3] which consists of two coupled integral equations for $\lambda(k,\omega)$ and $\bar{\eta}(k,\omega)$ [or equivalently $\Sigma(k,\omega)$ and $\Pi(k,\omega)$], obtained by retaining the diagrams in Figs. 1(a) and 2(a) and solving self-consistently for the self-energies. This approximation differs from the exact solution by the neglect of proper diagrams *("vertex corrections")* and by the use of the Ornstein-Zernike form for the static susceptibility $\chi_\psi(k)$. We shall examine below the validity of these two approximations in second order in ϵ, but we first wish to discuss the results obtained from the self-consistent scheme in three dimensions. Up to now no one has solved the full self-consistent equations, but a number of authors have obtained approximate solutions, and argued that these were rather close to the exact ones.

Kawasaki and Lo[11] neglected the frequency dependence of λ and $\bar{\eta}$ and obtained integral equations for $\lambda(k)$ and $\bar{\eta}(k)$ which they then solved numerically. The result may be cast in the form of Eq. (2.21) with $\bar{\Omega}$ and R now given by

$$\bar{\Omega}_s(x) = x^{-2}K(x)/K(0), \quad (4.9)$$

$$R_s = (6\pi)^{-1}K(0) = (6\pi)^{-1}(1.027) = 0.053, \quad (4.10)$$

where $K(x)$ is defined in Eq. (7) of Ref. 11. [Note, however, that due to a factor-of-2 error in Eq. (5) of Ref. 11, pointed out in Ref. 13, the values of $K/K_0 - 1$ have been divided by 2 in writing Eq. (4.10).] It follows from Eq. (4.10) that the self-consistency hardly changes the value of R from that in Eq. (4.4). Moreover, since the self-consistent approximation obeys the exact scaling relations (2.21) and (2.14) with $\eta = 0$, we must have

$$\bar{\Omega}(y) \underset{y\to\infty}{\sim} b y^{2-\epsilon+x_{\bar{\eta}}}, \quad (4.11)$$

that is,

$$K_0^{-1}(y)\,K(y) \underset{y\to\infty}{\sim} b\,y^{x_{\bar{\eta}}}. \quad (4.12)$$

Unfortunately, it is not possible to extract the exponent $x_{\bar{\eta}}$ from the published results, since $\bar{\Omega}$ may not have reached its large-y behavior in Fig. 3 of Ref. 11. Nevertheless, it seems clear that $x_{\bar{\eta}}$ remains rather small, and it would be interesting to extract a value of $x_{\bar{\eta}}$ and $\tilde{R}$ [Eq. (2.23)] from the equations of Kawasaki and Lo.[30] Kawasaki and Lo have also examined the validity of neglecting the frequency dependence of the transport coefficients, and they concluded[31] that the error was at most a few percent for large $k\xi$, and negligible for small $k\xi$.

It is interesting to ask why the *simple* Kawasaki approximation (4.1)–(4.5) is so successful[1] both near $d=4$ and at $d=3$, i.e., why $x_{\bar{\eta}}$ seems to remain small relative to x_λ. We believe that the answer lies in the geometric factors appearing in the numerators of Eqs. (A6) and (A7) of the Appendix. The extra factor of $\vec{p}\cdot\vec{k}$ in Eq. (A6) leads to a factor-of-18 difference in the coupling terms of the recursion relations (2.4a) and (2.4b) near four dimensions, and to the corresponding difference in exponents. A similar effect occurs for all values of d in Eqs. (A6) and (A7) and in higher diagrams as well (see the Appendix). Thus all corrections to the simple Kawasaki approximation (4.1)–(4.5) will contain the small parameter $x_{\bar{\eta}}/x_\lambda$, and the exact answer for $d=3$ is expected to be rather close to the lowest approximation.

The theory of Perl and Ferrell[12] is in principle equivalent to the self-consistent scheme of Kawasaki,[3] but slightly different approximations were made in obtaining quantitative results. Perl and Ferrell[12] used an iterative method of solution, with the simple Kawasaki solution (4.5) used in lowest order. In the next approximation a (weak) ξ dependence of $\bar{\eta}$ was obtained in the form

$$\bar{\eta}(\xi) = \bar{\eta}_0 + (8/15\pi^2)\ln(q_D\xi), \quad (4.13)$$

with q_D an adjustable parameter, and a scaling expression for $\bar{\eta}(\xi, q, \omega)$ inserted into the equation for $\lambda(q,\xi)$. The resulting expression for $\omega_\psi(q,\xi)$ was then found to be rather close to the original form

(4.3)–(4.5) as a result of a cancellation between the q and ω dependences of $\bar{\eta}$. This feature differs in detail from the estimates of Kawasaki and Lo,[11] but in any case the effects are all rather small. Equation (4.13) may be exponentiated to yield

$$x_{\bar{\eta}} \approx 8/15\pi^2 = 0.054, \tag{4.14}$$

and from the evaluation of ω_ψ made in Refs. 12 and 1 we estimate the Perl-Ferrell value of R to be

$$R = (6\pi)^{-1}(1.05). \tag{4.15}$$

It is difficult to extract $\tilde{R}$ from the published results,[12] but according to Swinney and Henry,[1] its value is slightly larger in Ref. 12 than in Ref. 11.

In an interesting extension of the work of Kawasaki and Lo,[11] Oxtoby and Gelbart[13] have calculated $\bar{\eta}(T)$ numerically, with no adjustable parameters, by including background terms λ_B and $\bar{\eta}_B$ in the self-consistent equations (see below). The results agree rather well with experiments on xenon,[32] but the approximations made are such that the authors cannot extract the asymptotic exponent $x_{\bar{\eta}}$ from their analysis.[33]

In order to assess the validity of the self-consistent approximation, a number of authors have made direct estimates in three dimensions of the omitted terms. The effect of vertex corrections was studied by Lo and Kawasaki,[34] and by Garisto and Kapral,[14] and although the quantitative estimates differ, the qualitative conclusion of both groups is that these effects introduce errors of less than 3% for $\omega_\psi(k, \xi)$. [For $\bar{\eta}(T)$ Garisto and Kapral[14] found somewhat larger effects, which improved the agreement with experiment.] The Ornstein-Zernike approximation was considered by Swinney and Saleh,[35] who replaced χ_ψ by the Fisher-Burford form, and found a modification of the scaling function which corresponds to[1]

$$R = (6\pi)^{-1}(1.02) = 0.054, \tag{4.16}$$

$$\tilde{R} = \tfrac{3}{8}\pi\,(1.03). \tag{4.17}$$

There is also a modification of $x_{\bar{\eta}}$ owing to the exponent η, but this was not explicitly considered in Ref. 35.

Another way to test the validity of the self-consistent approximation is to examine the ϵ expansion in second order. Indeed, as emphasized recently by Gunton and Kawasaki,[36] the self-consistent scheme is exact in first order in ϵ, and it is reasonable to ask how well it compares with our second-order results. The approximation consists in omitting the proper diagrams in Figs. 1(d) and 2(c) (vertex corrections), and setting $\eta = 0$ in the scaling relation (2.14) (the other corrections to Ornstein-Zernike do not affect R or the exponents in this order). If we repeat the calculation of the Appendix with these omissions, we find the ϵ expansion for the self-consistent approximation,

$$f_0 = \tfrac{24}{19}\epsilon\,[1 + 0.062\epsilon + O(\epsilon^2)], \tag{4.18}$$

$$x_\lambda = \tfrac{18}{19}\epsilon\,[1 - 0.013\epsilon + O(\epsilon^2)] \simeq 0.935, \tag{4.19}$$

$$x_{\bar{\eta}} = \tfrac{1}{19}\epsilon\,[1 + 0.242\epsilon + O(\epsilon^2)] \simeq 0.065, \tag{4.20}$$

$$R \equiv R_s = K_d(\tfrac{19}{24}\epsilon^{-1})\,[1 - 0.023\epsilon + O(\epsilon^2)] \simeq 0.039. \tag{4.21}$$

These results are indeed very close to the exact second-order expansion (1.3) and (2.19). We may therefore rewrite the exact coefficient R in an alternate form,

$$R = (R/R_s)(R_s/R_K)R_K, \tag{4.22}$$

where R_K is the simple Kawasaki approximation (4.7) and R_s is the self-consistent answer (4.21). According to the arguments given earlier, we expect the factors R/R_s and R_s/R_K to be close to unity for all d, so that the primary d dependence of R will come from the known d dependence of R_K. Thus a reasonable way to extrapolate R to $d = 3$ is to expand the small quantities $R/R_s - 1$ and $R_s/R_K - 1$ in powers of ϵ, and to evaluate R_K exactly for $d = 3$. The extrapolations of Eq. (4.8), (4.21), and (2.19) yield

$$R_s/R_K = 1 + \tfrac{1}{18} + \tfrac{1}{12}\epsilon - 0.023\epsilon \approx 1.116, \tag{4.23}$$

$$R/R_s = 1 + 0.083\epsilon \approx 1.083, \tag{4.24}$$

so that

$$R = 1.20R_K = 1.2/6\pi = 0.064. \tag{4.25}$$

This last estimate differs from the direct extrapolation $R = 0.042$ in (2.20), but is expected to be more reliable. We may also compare the result in Eq. (4.23) with the numerical estimate by Kawasaki and Lo, Eq. (4.10), and we find a rather large discrepancy whose origin is unclear to us at this point (note that we have divided the result of Fig. 3 in Ref. 11 by 2 because of the error mentioned above.)

A. Correction terms and "background subtractions"

Thus far we have discussed only the universal aspects of the problem, which should be applicable very near the critical point. In practice, however, owing to the weak divergence of the viscosity $\bar{\eta}(T)$, the region of strict universality is unattainable, since it corresponds to those temperatures where the singular part of $\bar{\eta}(T)$ dominates its background. In all experiments to date, $\bar{\eta}(T)$ never rises by more than 20%–30%, so that it is essential to treat the "correction" and "background" terms properly. This point has been emphasized by many authors,[1,13,37] but there remains some ambiguity in

deciding precisely how to perform the background subtractions.

The simplest scheme, which is in the spirit of the lowest-order Kawasaki approximation and would be correct for $x_{\bar{\eta}}=0$, is to replace $\lambda(T)$ by $\lambda(T)-\lambda_B$ and to keep the full $\bar{\eta}(T)$ in the mode-coupling equations. This is the method used by Kawasaki and Lo,[11] and it leads to Eqs. (4.3) and (4.9) with $\lambda(T)$ replaced by $\lambda(T)-\lambda_B$. (λ_B and $\bar{\eta}_B$ are the background values, which we may identify with λ_0 and $\bar{\eta}_0$, respectively.) A similar scheme was discussed in Ref. 20 for the thermal conductivity of helium at the λ point, where the specific heat has a very weak singularity. It was seen there that there are slow transients which appear as nonuniversal corrections to the leading singularities, and whose importance is measured by the difference between the effective exponent $\alpha_{\rm eff}$ and the true exponents $\tilde{\alpha}=\max(\alpha,0)$ of the specific heat. In the present case a similar analysis is possible, and singular corrections appear which are proportional to $x_{\bar{\eta}}^{\rm eff}-x_{\bar{\eta}}$ (here $x_{\bar{\eta}}$ remains positive). There is, however, a more practical way to take into account the background terms and to generate the appropriate corrections, namely, by solving the self-consistent equations for the full $\lambda(k,\xi,\omega)$ and $\bar{\eta}(k,\xi,\omega)$, *including* the background terms. This scheme was used by Oxtoby and Gelbart[13] and by Garisto and Kapral,[14] and it did lead to nonuniversal behavior, generated by the terms λ_B and $\bar{\eta}_B$. The statement by Oxtoby and Gelbart[13] that the nonuniversality persists right up to the critical point is misleading, however, since this is only true at finite q, and the nonuniversal corrections vanish in the scaling limit, i.e., when q and $\xi^{-1}\to 0$, for any fixed $q\xi$. It is nevertheless interesting that according to the estimates of these authors[13,14] the nonuniversal corrections to $\omega_\psi(k,\xi)$ which arise out of the self-consistency are of order 3%–5% in the experimental range.

From the preceding discussion we conclude that the most accurate way to obtain $\bar{\eta}(T)$ and $\omega_\psi(k,\xi)$ is by solving the following self-consistent equations in three dimensions:

$$\Sigma_2(k,\omega)=-k_BT\chi_\psi^{-1}(k)\int\frac{d^3p}{(2\pi)^3}\frac{d\omega'}{2\pi}\times\left(2\chi_\psi(p_+)(\vec{k}\cdot\mathcal{T}_{p_-}\cdot\vec{k})\frac{\mathrm{Re}[-i\omega'+\lambda_0p_+^2\chi_\psi^{-1}(p_+)-\Sigma_2(p_+,\omega')]^{-1}}{-i(\omega-\omega')+\bar{\eta}_0p_-^2-\Pi_2(p_-,\omega-\omega')}\right),\tag{4.26}$$

$$\Pi_2(k,\omega)=-2k_BT(d-1)^{-1}\int\frac{d^3p}{(2\pi)^3}\frac{d\omega'}{2\pi}\times\left(2\chi_\psi(p_-)(\vec{p}\cdot\vec{k})(\vec{p}\cdot\mathcal{T}_k\cdot\vec{p})\frac{\mathrm{Re}[-i\omega'+\lambda_0p_-^2\chi_\psi^{-1}(p_-)-\Sigma_2(p_-,\omega')]^{-1}}{-i(\omega-\omega')+\lambda_0p_+^2\chi_\psi^{-1}(p_+)-\Sigma_2(p_+,\omega-\omega')}\right),\tag{4.27}$$

with $p_\pm=p\pm\frac{1}{2}k$. Together with Eqs. (A11), (A23), and (A24) of the Appendix and Eqs. (2.25) and (2.26), the above equations fully determine the Rayleigh linewidth and transport coefficients if the Fisher-Burford approximant[35,38] is used for $\chi_\psi(k)$ and the background values λ_0 and $\bar{\eta}_0$ are replaced by smooth function $\lambda_B(T)$ and $\bar{\eta}_B(T)$, extrapolated from experiments far from T_c.[1] The universal limit of the solution should be rather close to that obtained by Kawasaki and Lo,[11] with a factor of 2 corrected and the "correlation function" modification of Swinney and Saleh[35] also taken into account (note, however, that in contrast to previous work, this scheme will satisfy the scaling relation $x_\lambda+x_{\bar{\eta}}=\epsilon-\eta$ with *finite* η). The most important difference with the solution of Kawasaki and Lo will be in the nonuniversal corrections, which are now included in the theory in a complicated way. These will not be negligible and they must be considered in any comparison with experiment. The main feature missing from Eqs. (4.26) and (4.27), apart from the approximate nature of the static $\chi_\psi(k,\xi)$, is the vertex correction, which can be taken into account approximately, as was done by a number of authors,[14,34] but in any case is not expected to change the values of $\omega_\psi(k,\xi)$ significantly. In this way the theory may be improved to an expected accuracy of roughly 3%–5%, which is quite remarkable for a first-principles calculation with no adjustable parameters. Indeed, the above scheme has in large measure already been implemented for the static viscosity $\bar{\eta}(T)$,[14] and to a considerable extent for the Rayleigh linewidth $\omega_\psi(k,\xi)$.[11–13] It is hoped that the refinements mentioned above will bring about further improvement in the agreement with experiment.

APPENDIX

A. Formalism

The systematic perturbation theory we use to implement the renormalization group has been published some time ago[24] and reviewed in the context of helium and the antiferromagnet[20] recently. We will say very little of a general nature here and

merely list a few of the intermediate steps in our calculations. In order to have Feynman-like rules it was necessary to enlarge the set of fields ψ and j by two others, $\hat{\psi}$ and $\hat{j}$, which act as their formal adjoints. In addition to the correlation functions $\langle\psi\psi^*\rangle(k,\omega)$ and $\langle jj^*\rangle(k,\omega)$, there are two nonphysical response functions to calculate. A dressed perturbation theory exists if one introduces two self-energies. For the model of Eqs. (2.1) the physical correlation functions are

$$\langle\psi\psi^*\rangle = \frac{2\lambda_0 k^2 + \Sigma_1}{|-i\omega + \lambda_0 k^2(r_0 + k^2) - \Sigma_2|^2}, \tag{A1}$$

$$\langle j_\alpha j_\beta^*\rangle = \mathcal{T}_k^{\alpha\beta}\frac{2\bar{\eta}_0 k^2 + \Pi_1}{|-i\omega + \bar{\eta}_0 k^2 - \Pi_2|^2}, \tag{A2}$$

where

$$\mathcal{T}_k^{\alpha\beta} \equiv \delta_{\alpha\beta} - k_\alpha k_\beta/k^2. \tag{A3}$$

The self-energies Σ and Π may be expressed as expansions in the vertices g_0 and u_0 of Eqs. (2.1a)–(2.1c). From the fluctuation dissipation relation it follows that Σ_2 is proportional to $k^2(r_0 + k^2)$, while Σ_1, Π_1, and Π_2 are proportional to k^2 because the corresponding fields are conserved. We assert that at each stage of renormalization the frequency integral of the correlation function is just the static susceptibility. In the limit of small k and ω the wave-number prefactors of the self-energies match those in the bare theory and the correlation functions may be written

$$\langle\psi\psi^*\rangle_l = \frac{2\lambda_l k^2}{|-i\omega + \lambda_l k^2(r_l + k^2)|^2}, \tag{A4}$$

$$\langle jj^*\rangle_l = \frac{2\bar{\eta}_l k^2}{|-i\omega + \bar{\eta}_l k^2|^2}. \tag{A5}$$

In other words, one may use either self-energy to renormalize the transport coefficients. We will analyze only Σ_2 and Π_2 in what follows.

B. Lowest-order recursion relations

In Ref. 22 it was shown that in model B [$g_0 = 0$ in Eq. (2.1)] the transport coefficients are unaffected by interactions. In our more general model, the four-point interaction u_0 renormalizes only static parameters, up to order ϵ^2. At any step l in the renormalization procedure our equations properly reduce to the equations for the free energy of Wilson and Kogut.[5] The transport coefficients are therefore renormalized only by g_0, at least up to second order. To first order we find in d dimensions

$$\Pi_2^l = \frac{-2g_l^2}{d-1}\int\frac{d^dp}{(2\pi)^d}\frac{(\vec{p}\cdot\mathcal{T}_k\cdot\vec{p})(\vec{p}\cdot\vec{k})}{(r_l + p_-^2)[-i\omega + \lambda_l p_+^2(r_l + p_+^2) + \lambda_l p_-^2(r_l + p_-^2)]}, \tag{A6}$$

$$\Sigma_2^l = -(r_l + k^2)g_l^2\int\frac{d^dp}{(2\pi)^d}\frac{(\vec{k}\cdot\mathcal{T}_{p_-}\cdot\vec{k})}{(r_l + p_+^2)[-i\omega + \bar{\eta}_l p_-^2 + \lambda_l p_+^2(r_l + p_+^2)]}, \tag{A7}$$

with $p_\pm = p \pm \frac{1}{2}k$, the integrals being over a momentum shell $\Lambda/b \leq p \leq \Lambda$. The recursion relations (2.4a) and (2.4b) then follow from the equations

$$\lambda_{l+1} = b^{z+\eta-4}\left(\lambda_l - \lim_{k,\omega\to 0}(k^2 r_l)^{-1}\Sigma_2^l(k,\omega)\right), \tag{A8}$$

$$\bar{\eta}_{l+1} = b^{z-2}\left(\bar{\eta}_l - \lim_{k,\omega\to 0} k^{-2}\Pi_2^l(k,\omega)\right), \tag{A9}$$

with the integrals in (A6) and (A7) calculated for $d = 4$. Our self-energies are similar to those found from mode-coupling theory. It should be noted that $\lambda_l p_+^2(r_l + p_+^2)$ may be omitted in the denominator of Eq. (A7) because it is small compared to $\bar{\eta}_l p_-^2$. The integrals are logarithmically divergent in four dimensions. Had we retained higher-order terms in k in Eqs. (A8) and (A9), we would generate contributions to $\bar{\eta}$ and λ proportional to k^2, k^4, etc., of order $g_l^2/\lambda_l\bar{\eta}_l$. If such terms are reinserted into Eqs. (A6) and (A7) the convergence properties of the integrals are unchanged and terms of order ϵ^2 appear in the recursion formulas. Thus terms in the equations of motion which are of higher order in wave number than those involving λ and $\bar{\eta}$ are irrelevant.

C. Higher-order terms

We shall now outline the argument which shows that higher-order diagrams will not modify the recursion formulas to order ϵ. In addition, for the ϵ expansion, we must show that the dimensionless constant which measures the strength of the nondissipative coupling is simply $f_l = g_l^2/\lambda_l\bar{\eta}_l$, in the limit $\bar{\eta}_l/\lambda_l \to \infty$, which occurs when $l \to \infty$. Consider the proper fourth-order self-energy diagrams in Figs. 1(d) and 2(c). (Improper diagrams may be treated inductively.) In Fig. 1(d) which contributes to λ_l there are two loops and five propagators. Performing the internal frequency integral will leave just three denominators, each one being the sum of some subset of the five characteristic frequencies in the diagram. The most divergent term occurs when both internal frequency scales are set by the order parameter, since it is of order $\lambda_l p^4$ while $\vec{j}$ relaxes as $\bar{\eta}_l p^2$. But there are two momen-

tum propagators, each relaxing as $\overline{\eta}_l p^2$, which carry through the internal frequency integrals. The final result has at most one denominator out of the three which can be of order $\lambda_l p^4$. The others are of order $\overline{\eta}_l p^2$. If we remove $\overline{\eta}_l^{-2}\lambda_l^{-1}\chi_\psi^{-1}(k)k^2$ from the expression for Σ_2, simple dimensional analysis implies that the remainder of the diagram in Fig. 1(d) is dimensionless for $d=4$, and behaves as $\ln b$ in the limit $l\to\infty$, $\lambda_l/\overline{\eta}_l\to 0$. The result is quite general; the singular part of the diagram of order g_l^{2n} is $\overline{\eta}_l^{-n}\lambda_l^{-n+1}$, and when written in the form of Eq. (2.4d) it is proportional to f_l^n. For diagrams beyond second order in ϵ, there occur terms proportional to $u_l^m g_l^{2n}$. It may be shown that these contribute a correction to λ_{l+1} which is proportional to $u_l^m f_l^n \lambda_l$ in the limit $\eta_l/\lambda_l\to\infty$. There are analogous results for Π_2^l; the most divergent diagrams are of order $\ln b$ and behave as $\overline{\eta}_l^{-1}\lambda_l^{-2}$.

D. Scaling function

In order to evaluate the scaling function for the characteristic frequency $\omega_\psi(k,\xi)$ and the exponents in second order, we shall use Wilson's Feynman-graph expansion.[27] In this method momentum integrals are performed from 0 to Λ, as in ordinary perturbation theory (rather than from Λ/b to Λ), and the relevant parameters are fixed at particular values which ensure the exponentiation of the series (see the discussion in Appendix D of Ref. 20).

The parameter which must be fixed in the present case is the vertex f_0 defined in Eq. (2.2), and it may be shown in the usual way that the proper choice is

$$f_0(\epsilon)=\tfrac{24}{19}\epsilon+O(\epsilon^2). \tag{A10}$$

In order to calculate the characteristic frequency ω_ψ, defined in (2.25) and (2.26), we need the response function $\chi_\psi(k,\omega)$. Although the general expression for $\chi_\psi(k,\omega)$ in terms Σ_1 and Σ_2 is rather complicated when both u_0 and g_0 are nonzero (see Appendix B of Ref. 20), the equations simplify in first and second order in ϵ. We have

$$\chi_\psi(k,\omega)=[\lambda_0 k^2-\chi_\psi(k)\Sigma_2(k,\omega)]\times[-i\omega+\lambda_0 k^2\chi_\psi^{-1}(k)-\Sigma_2(k,\omega)]^{-1}, \tag{A11}$$

and using Eq. (2.26),

$$\lambda(k,\xi)=\lambda_0(\xi\Lambda)^{18\epsilon/19}\left[1+(24\epsilon/19K_4)\int\frac{d^4p}{(2\pi)^4}\left(\frac{\hat{k}\cdot\Gamma_p\cdot\hat{k}}{p^2[\xi^{-2}+(p+k)^2]}-\frac{\frac{3}{4}}{p^2(\xi^2+p^2)}\right)+O(\epsilon^2)\right]. \tag{A12}$$

In obtaining Eq. (A12) we have made a subtraction in the integral, and have exponentiated the order-ϵ term which is independent of k. Inserting (A12) into (2.25) yields Eq. (2.22), when the expressions $\chi_\psi^{-1}(k)=r+k^2=\xi^{-2}+k^2$, $\overline{\eta}(T)=\eta_0(\xi\Lambda)^{\epsilon/19}$, Eq. (2.14), and Eq. (2.2) with $g_0^2=k_BT=1$ are used.

As mentioned in Sec. II, to first order in ϵ the definition of λ in Eq. (2.26) agrees with that obtained from a Lorentzian fit, since the corrections coming from $\partial\Sigma_2/\partial(-i\omega)$ are of higher order in ϵ or k. It may be shown quite generally that for $k\to 0$ the definitions (A8) and (2.26) are identical, but for finite k and in higher order in ϵ this is not the case.

E. Second-order calculation

The computation of x_λ and $x_{\overline{\eta}}$ to second order in ϵ is rather complicated and we will summarize only the principal steps. (It is very similar to the corresponding calculation in Appendix D of Ref. 20.) Contributions to Σ_2 and Π_2 come from three sources: the first-order diagrams [Figs. 1(a) and 2(a)] in $4-\epsilon$ dimensions, the second-order improper diagrams in four dimensions [Figs. 1(b), 1(c), and 2(b)], and the second-order proper diagrams also in four dimensions [Figs. 1(d), 2(c)]. The diagram in Σ_2 of order u_0^2 does not contribute to the renormalization of λ, as was found previously in model B of Ref. 22. The diagram of order $u_0 f_0$ also gives no contribution. We assume that a mass renormalization[27] has been made and the integrals which now run over all momenta are functions of the inverse susceptibility r. The improper and first-order diagrams are of the form $A\ln^2 r+B\ln r+C$, while the proper contributions do not contain a $\ln^2 r$ term. For our purposes we need only the $\ln r$ terms and the constants to order ϵ. The $\ln^2 r$ terms serve as a check that the perturbation series exponentiate. From its definition we calculate the renormalized f which must scale as $r^{(\epsilon-\eta)/(2-\eta)}$. Matching against the perturbative result gives f_0 to second order. We then return to Σ_2 and Π_2 and exponentiate to find x_λ and $x_{\overline{\eta}}$.

We label the three contributions to the self-energy in conformity with Figs. 1 and 2,

$$\Sigma_2^a=\lambda_0 k^2(r+k^2)f_0[\tfrac{3}{8}(1-\tfrac{1}{12}\epsilon)\ln r-\tfrac{3}{32}\epsilon\ln^2 r], \tag{A13}$$

$$\Sigma_2^b+\Sigma_2^c=(r+k^2)g_0^4\overline{\eta}_0^{-2}\lambda_0^{-1}\frac{1}{3}\int\frac{d^4p}{(2\pi)^4}\frac{d^4q}{(2\pi)^4}\left(\frac{(\vec{k}\cdot\Gamma_p\cdot\vec{k})(\vec{q}\cdot\Gamma_p\cdot\vec{q})[(q+p)^2-q^2]}{(r+p^2)(r+q^2)p^4\{p^2(r+p^2)+q^2(r+q^2)+(p+q)^2[r+(p+q)^2]\}}\right) \tag{A14}$$

$$=\lambda_0 k^2(r+k^2)f_0^2(\tfrac{1}{256}\ln^2 r+(0.00231)\ln r), \tag{A15}$$

$$\Sigma_2^d = (r+k^2)\, g_0^4 \bar{\eta}_0^{-2} \lambda_0^{-1} \int \frac{d^4p}{(2\pi)^4} \frac{d^4q}{(2\pi)^4}$$

$$\times \frac{(\vec{k}\cdot\mathcal{T}_p\cdot\vec{q})\,(\vec{p}\cdot\mathcal{T}_q\cdot\vec{k})[(p+q)^2-q^2][(p+q)^2-p^2]}{(r+p^2)(r+q^2)[r+(p+q)^2]p^2q^2\{p^2(r+p^2)+q^2(r+q^2)+(p+q)^2[r+(p+q)^2]\}} \tag{A16}$$

$$= \lambda_0 k^2 (r+k^2) f_0^2 (0.0426) \ln r, \tag{A17}$$

$$\Pi_2^a = \bar{\eta}_0 k^2 f_0 (\tfrac{1}{48})[\tfrac{3}{2} + (1-\tfrac{1}{3}\epsilon)\ln r - \tfrac{1}{4}\epsilon \ln^2 r], \tag{A18}$$

$$\Pi_2^b = g_0^4 \lambda_0^{-2} \bar{\eta}_0^{-1} \int \frac{d^4p}{(2\pi)^4} \frac{d^4q}{(2\pi)^4} \left(\frac{(\vec{p}\cdot\vec{k})^2 (\vec{p}\cdot\mathcal{T}_k\cdot\vec{p})\,(\vec{p}\cdot\mathcal{T}_{p+q}\cdot\vec{p})}{p^4(r+p^2)^3(r+q^2)(p+q)^2} \right) \tag{A19}$$

$$= \bar{\eta}_0 k^2 f_0^2 (\tfrac{1}{128})\,(\tfrac{1}{2}\ln^2 r + \ln r), \tag{A20}$$

$$\Pi_2^c = -g_0^4 \lambda_0^{-2} \bar{\eta}_0^{-1} \frac{1}{3} \int \frac{d^4p}{(2\pi)^4} \frac{d^4q}{(2\pi)^4} \left(\frac{(\vec{k}\cdot\vec{p})\,(\vec{k}\cdot\vec{q})(\vec{p}\cdot\mathcal{T}_k\cdot\vec{q})\,(\vec{q}\cdot\mathcal{T}_{p+q}\cdot\vec{q})}{(r+p^2)^2(r+q^2)^2(p+q)^2p^2q^2} \right), \tag{A21}$$

$$= \bar{\eta}_0 k^2 f_0^2 (\tfrac{1}{384}) \ln r. \tag{A22}$$

The two integrals in $\Sigma_2^b + \Sigma_2^c$ and Σ_2^d were solved numerically after reduction to integrals in one variable.

The above expressions are inserted into the equations for λ and $\bar{\eta}$ analogous to (A8) and (A9),

$$\lambda = \lambda_0 - \lim_{k,\omega\to 0} (k^2 r)^{-1} \Sigma_2(k,\omega), \tag{A23}$$

$$\bar{\eta} = \bar{\eta}_0 - \lim_{k,\omega\to 0} k^{-2} \Pi_2(k,\omega), \tag{A24}$$

and f_0 is adjusted order by order in ϵ, in order to satisfy the scaling relation (2.14),

$$f(T) \propto (\lambda\bar{\eta})^{-1} \propto r^{(\epsilon-\eta)/(2-\eta)}. \tag{A25}$$

The appropriate value is

$$f_0 = \tfrac{24}{19}\epsilon - 0.127\epsilon^2 + O(\epsilon^3), \tag{A26}$$

where we have used the static value[5] $\eta = \frac{1}{54}\epsilon^2 + O(\epsilon^3)$. With the above value of f_0 the individual series for λ and $\bar{\eta}$ may be compared with $1 - \frac{1}{2}x_\lambda \ln r$ and $1 - \frac{1}{2}x_{\bar{\eta}} \ln r$, respectively, to find the exponents given in (1.3).

In order to determine the amplitude R [Eq. (2.15)], we note that to order ϵ the series for $\bar{\eta}$ and λ have the form

$$\bar{\eta} = \bar{\eta}_0 (1 - \tfrac{1}{32} f_0)[1 - \tfrac{1}{2} x_{\bar{\eta}} \ln(r/\Lambda^2)], \tag{A27a}$$

$$\lambda = \lambda_0 [1 - \tfrac{1}{2} x_\lambda \ln(r/\Lambda^2)], \tag{A27b}$$

so that R may be written, to order ϵ^2, as

$$R = (K_d/f_0)\,(1 - \tfrac{1}{32} f_0), \tag{A28}$$

where we have used Eq. (2.14). Inserting Eq. (A26) into (A28) yields (2.19).

*Present address: Department of Physics, University of Pennsylvania, Philadelphia, 19174.

[1]An excellent review of the experimental and theoretical situation may be found in H. L. Swinney and D. L. Henry, Phys. Rev. A 8, 2586 (1973).
[2]L. P. Kadanoff and J. Swift, Phys. Rev. 166, 89 (1968).
[3]K. Kawasaki, Ann. Phys. (N.Y.) 61, 1 (1970).
[4]P. Bergé, P. Calmettes, C. Laj, M. Tournarie, and B. Volochine, Phys. Rev. Lett. 24, 1223 (1970).
[5]K. G. Wilson and J. Kogut, Phys. Rep. C 12, 95 (1974).
[6]A preliminary account of this work appeared in B. I. Halperin, P. C. Hohenberg, and E. D. Siggia, Phys. Rev. Lett. 32, 1289 (1974).
[7]B. I. Halperin and P. C. Hohenberg, Phys. Rev. 177, 952 (1969).
[8]K. Kawasaki, Phys. Lett. A 30, 325 (1969).
[9]G. Arcovito, C. Faloci, M. Roberti, and L. Mistura, Phys. Rev. Lett. 22, 1040 (1969); R. A. Ferrell, Phys. Rev. Lett. 24, 1169 (1970); B. J. Ackerson, C. M. Sorensen, R. C. Mockler, and W. J. O'Sullivan, Phys. Rev. Lett. 34, 1371 (1975).
[10]R. A. Ferrell, N. Menyhàrd, H. Schmidt, F. Schwabl, and P. Szépfalusy, Ann. Phys. (N.Y.) 565 (1968).
[11]K. Kawasaki and S. M. Lo, Phys. Rev. Lett. 29, 48 (1972).
[12]R. Perl and R. A. Ferrell, Phys. Rev. Lett. 29, 51 (1972).
[13]D. W. Oxtoby and W. M. Gelbart, J. Chem. Phys. 61, 2957 (1974).
[14]F. Garisto and R. Kapral, report (unpublished).
[15]J. Hubbard and P. Schofield, Phys. Lett. A 40, 245 (1972).
[16]M. Grover and J. Swift (unpublished); J. van Leeuven (unpublished).
[17]J. Swift, Phys. Rev. 173, 257 (1968).
[18]M. S. Giterman and E. E. Gorodetskii, Zh. Eksp. Teor. Fiz. 57, 637 (1969) [Sov. Phys.-JETP 30, 348 (1970)]; L. Mistura, J. Chem. Phys. 62, 4571 (1975).
[19]M. Papoular, J. Chem. Phys. 60, 86 (1974).
[20]B. I. Halperin, P. C. Hohenberg, and E. D. Siggia, Phys. Rev. B (to be published).
[21]B. I. Halperin, in Proceedings of the International Conference on Statistical Physics, Budapest, 1975 (unpublished).

[22]B. I. Halperin, P. C. Hohenberg, and S. K. Ma, Phys. Rev. B 10, 139 (1974).
[23]K. G. Wilson and M. E. Fisher, Phys. Rev. Lett. 28, 240 (1972).
[24]P. C. Martin, E. D. Siggia, and H. A. Rose, Phys. Rev. A 8, 423 (1973).
[25]L. Van Hove, Phys. Rev. 93, 1374 (1954).
[26]B. I. Halperin, in *Collective Properties of Physical Systems, Nobel Symposium 24,* edited by B. Lundqvist and S. Lundqvist (Academic, New York, 1973), p. 54.
[27]K. G. Wilson, Phys. Rev. Lett. 28, 548 (1972).
[28]L. D. Landau and E. M. Lifshitz, *Fluid Mechanisms* (Pergamon, London, 1959), Chap. VI.
[29]K. G. Wilson, Phys. Rev. B 4, 3174, 3184 (1971).
[30]An approximate estimate of the exponent x_η has recently been made by T. Ohta (unpublished), starting from the equations of Ref. 11.
[31]K. Kawasaki and S. M. Lo, Phys. Rev. A 8, 2176 (1973).
[32]H. J. Strumpf, A. F. Collings, and C. J. Pings, J. Chem. Phys. 60, 3109 (1974); see also J. V. Sengers, AIP Conf. Proc. 11, 229 (1973).
[33]K. Kawasaki and J. D. Gunton, Phys. Lett. A 53, 119 (1975).
[34]S. M. Lo and K. Kawasaki, Phys. Rev. A 5, 421 (1972); T. Ohta and K. Kawasaki (unpublished).
[35]H. L. Swinney and B. E. A. Saleh, Phys. Rev. A 7, 747 (1973).
[36]J. D. Gunton and K. Kawasaki, J. Phys. A 8, L9 (1975), and unpublished.
[37]J. V. Sengers and P. H. Keyes, Phys. Rev. Lett. 26, 70 (1971).
[38]M. E. Fisher and R. J. Burford, Phys. Rev. 156, 583 (1967).