Critical viscosity exponent for a classical fluid

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The dynamic renormalization group yields a value for the critical exponent which we find to be too large. We believe that the true value is close to the decoupled mode value of $8/15\pi^2=0.054$. The error in the renormalization-group calculation comes from the two-term truncation of the ϵ expansion. The ϵ^3 term that is neglected is of opposite sign and as large as the ϵ^2 term that is included. By avoiding the ϵ expansion entirely, we eliminate any truncation error and achieve greatly improved accuracy.

I. INTRODUCTION

Some years ago Kawasaki[1] and Perl and Ferrell[2] found to single-loop order a logarithmic divergence in the viscosity of a classical fluid near its critical point, with a coefficient $8/15\pi^2=0.054$. When exponentiated, this lowest-order calculation corresponds to a critical exponent of this same amount, as noted by Ohta and Kawasaki.[3] Subsequently Siggia, Halperin, and Hohenberg[4] reported the larger value of 0.065, on the basis of the dynamic renormalization group (DRG). This DRG value has been generally accepted without much question. Our present purpose is to point out that the DRG value does not bear close scrutiny. We present compelling reasons to believe that the exponent has to be very nearly equal to the "old" decoupled mode value of $8/15\pi^2$. Our goal here is to put forward a convincing case for our assertion of the validity of the old values as a good first-order approximation.

The order-parameter critical fluctuations in a classical fluid near its second-order phase transition are of density and concentration in the single-component and binary liquid, respectively. In each case these fluctuations are carried along by the hydrodynamic shear modes, the fluctuations of which decay because of the viscosity. The coupling between the transverse velocity and the order-parameter fluctuations causes the viscosity to diverge weakly at the critical point. This coupling is sketched in Sec. II where we observe that the DRG[4] result of a very small two-loop effect makes it appropriate to concentrate on the single-loop graphs. Sections II A and II B are devoted to establishing that the exponent and the universal amplitude ratio are close to the single-loop values of 0.054 and 1.00, respectively. Section III shows the inadequacy of a two-term truncation of the single-loop integral in an ϵ expansion. The error in the DRG[4] result can be traced to this truncation. In Sec. IV, we explain the basic reason why low-order ϵ expansions are not reliable. In Sec. V we compare our theoretical values for the exponent and for the universal amplitude ratio with those that can be deduced from the high-precision light scattering experiments of Burstyn and Sengers.[5] Section VI is a brief summary.

II. MODE COUPLING

A. Critical exponent

The magnitude $\phi_{\vec{k}}$ of an order-parameter fluctuation of wave number $\vec{k}$ is given by the critical-point correlation function

$$g(k)=\langle |\phi_{\vec{k}}|^2\rangle=\frac{1}{k^2}\ , \tag{2.1}$$

where we use the Ornstein-Zernike[6] approximation throughout. The rate of relaxation of the fluctuation is expressed in terms of the wave-number-dependent diffusion as

$$\gamma(k)=k^2D(k)=k^2\frac{L(k)}{g(k)}=k^4L(k)\ . \tag{2.2}$$

$L(k)$ is the kinetic coefficient representing the random walk of the inhomogeneities as they are transported by the Brownian motion of the hydrodynamic shear modes. Simple physical considerations[7] give the k dependence

$$L(k)\propto\frac{1}{k^{4-D-z_\eta}}\ , \tag{2.3}$$

which permits us to write Eq. (2.2) in the form

$$\gamma(k)=N_Da_\gamma k^{D+z_\eta}\ , \tag{2.4}$$

where z_η is the small critical viscosity exponent, D is the space dimensionality, and a_γ is the k-independent amplitude. It is convenient to fix the D-dependent normalization factor by

$$N_D^2=\frac{C_D}{(2\pi)^D}\ , \tag{2.5}$$

where C_D is the area of the unit sphere in D-dimensional space. In a similar fashion we can write the k-dependent viscosity as

$$\eta(k)=N_Da_\eta k^{-z_\eta}\ . \tag{2.6}$$

The fluctuations of the stress tensor, which by the Kubo formula determine the viscosity, are proportional to

$$T_{||,\perp} \propto \partial_{||}\phi\partial_{\perp}\phi \propto \mu_{||} p \phi_{\vec{p}} \mu_{\perp} p' \phi_{\vec{p}'} , \tag{2.7}$$

where $\phi_{\vec{p}}$ and $\phi_{\vec{p}'}$ are Fourier components of the order parameter. The wave numbers $\vec{p}$ and $\vec{p}\,'$ satisfy the "conservation of momentum" condition

$$\vec{p}+\vec{p}\,'=\vec{k} , \tag{2.8}$$

where $\vec{k}$ is the wave number of the hydrodynamic shear mode. $\mu_{||}$ and $\mu_{\perp}$ are the direction cosines of $\vec{p}$ and $\vec{p}\,'$ parallel and perpendicular to $\vec{k}$, respectively. As a consequence of Eq. (2.7), the coupled-mode integral for the viscosity in the limit of weak divergence (i.e., $z_\eta \to 0$) is proportional to the angular average

$$\delta_D \equiv \langle \mu_{||}^2 \mu_{\perp}^2 \rangle = \frac{1}{D(D+2)} \tag{2.9}$$

which is of $O(D^{-2})$ and vanishes in the limit $D \to \infty$. This is because the great circles $\mu_{||,\perp}=0$ are nodal lines in the angle integrations of Eq. (2.9) and remove from the sphere belts which make the biggest contribution to C_D. For finite D we have $\delta_4=(24)^{-1}=0.04$ and $\delta_3=(15)^{-1}=0.07$. Thus for the values of D of interest, δ_D provides a convenient small parameter.

The usefulness of δ_D becomes evident upon examining the loop expansion of Siggia *et al.*[4] It will then be noted that their result $z_\eta=0.065$, as quoted above is actually the sum $z_\eta=z_1+z_2$, where $z_1=0.069$ is the single-loop contribution and $z_2=-0.004$ is the total contribution from the two-loop graphs. The fact that $|z_2|/z_1=0.06$ is smaller than unity by an order of magnitude is no accident, and can be traced to the role of δ_D in the two-loop integrals. It is easily seen by examining the integrands of the two-loop integrals of Siggia *et al.*[4] that the additional vertices of the two-loop graphs introduce corresponding additional angular factors of the type shown in Eq. (2.9). It is therefore quite natural that $z_1=O(\delta_D)$ and that $z_2=O(\delta_D^2)$, the general rule being that each additional loop brings in two more nodal lines in the angular integrations and thus another factor of δ_D. This permits us to write the loop expansion for the viscosity amplitude as

$$a_\eta = \frac{\delta_D}{z_\eta a_\gamma} J_D^{(1)}(z_\eta) + \frac{\delta_D^2}{z_\eta a_\gamma^2 a_\eta} J_D^{(2)}(z_\eta) + \cdots , \tag{2.10}$$

where as already noted, the single-loop, or decoupled mode, integral has the limiting value $\delta_D J_D^{(1)}(0)=\delta_D$. The integral is normalized as a function of z_η at $z_\eta=0$ according to

$$J_D^{(1)}(0)=1 . \tag{2.11}$$

The usefulness of a loop expansion in powers of δ_D, such as that of Eq. (2.10), depends, of course, on having $J_D^{(2)}$ and the higher-loop integrals be of $O(1)$. This is supported by the above plausibility argument. But, if the skeptical reader prefers, he can regard this as a *firmly established mathematical fact,* at least for the coefficients of δ_D^2, in consequence of the explicit numerical evaluation of the two-loop integrals by Siggia *et al.*[4] Our general formulation of the problem differs in no essential way from that of Siggia *et al.*[4] When written for general dimensionality D, our integrals are identical to theirs. The only true difference is a computational one. Our point is that, instead of estimating the integrals from a two-term truncation of the Taylor-series expansions in powers of $\epsilon=4-D$, it is more accurate actually to evaluate the integrals by setting $D=3$ at the outset. In other words, in getting numerical results we completely avoid the ϵ expansion. This paper is devoted to carrying this out for z_1 because, as seen in the work of Siggia *et al.*,[4] z_1 is overwhelmingly the dominant contribution to z_η.

The loop expansion for the relaxation rate amplitude is

$$a_\gamma = \frac{1}{a_\eta} I_D^{(1)}(z_\eta) + \frac{\delta_D}{a_\eta^2 a_\gamma} I_D^{(2)}(z_\eta) + \cdots . \tag{2.12}$$

Equations (2.10) and (2.12), assuming that the necessary integrals have been calculated as functions of z_η, suffice to determine both the product $a_\eta a_\gamma$ and the exponent z_η, to any desired order in δ_D. The ratio of the two equations gives z_η implicitly as

$$z_\eta = \delta_D \frac{J_D^{(1)}(z_\eta) + \frac{\delta_D}{a_\eta a_\gamma} J_D^{(2)}(z_\eta) + \cdots}{I_D^{(1)}(z_\eta) + \frac{\delta_D}{a_\eta a_\gamma} I_D^{(2)}(z_\eta) + \cdots}$$

$$= \frac{\delta_D}{I_D} + O(\delta_D^2) = z_1 + z_2 + \cdots , \tag{2.13}$$

where

$$I_D = I_D^{(1)}(0) = \frac{1}{C_D} \int \frac{d^D p \sin^2\theta}{p^2 p'^2} \tag{2.14}$$

with the wave numbers constrained by Eq. (2.8) and scaled to $k=1$. θ is the angle between $\vec{p}$ and $\vec{k}$. For $D=3$ substitution of $\delta_3=\frac{1}{15}$ and

$$I_3 = \frac{\pi^2}{8} = 1.234 \tag{2.15}$$

into Eq. (2.13) gives

$$z_1 = \frac{8}{15\pi^2} = 0.054 . \tag{2.16}$$

The $O(\delta_3^2)$ correction to the first-order value of z_η comes from the two first-order self-consistency corrections to $J_D^{(1)}(z_\eta)$ and $I_D^{(1)}(z_\eta)$ and from the two vertex corrections $J_D^{(2)}(0)$ and $I_D^{(2)}(0)$. Reported[4] computations of these contributions, carried out in the limit $D=4$, are of the expected magnitude $O(\delta_4^2)$. It follows that the true value of z_η has to fall quite close to the first-order estimate of $8/15\pi^2=0.054$. Although they did not emphasize it, this conclusion was also reached by Ohta and Kawasaki.[3] Our purpose here is to go further and to reconcile the DRG and the mode-coupling calculations by demonstrating the error in the former.

In a brief paper Garisto and Kapral[8] advocate a value for z_η between 0.06 and 0.07. This significant deviation from 0.054 is, according to them, due to a large vertex

correction. Their result comes from a numerical integration for $z_\eta^{(1)} J_3^{(2)}(z_\eta)$. They state that they carry out this numerical integration after first making the approximation $z_\eta = 0$ in the integrand. But this procedure produces the divergent integral $\lim_{z_\eta \to 0}[z_\eta^{-1} J_3^{(2)}(z_\eta)]$, which can only be made finite by imposing a Debye cutoff, as was done by Perl and Ferrell[2] for $z_\eta^{-1} J_3^{(1)}(z_\eta)$. With a cutoff the integral acquires a logarithmic dependence on k, which cannot be characterized by a single number, as was done by Garisto and Kapral. The Garisto-Kapral work, moreover, seems not to take into account the other vertex correction $I_3^{(2)}(0)$.

B. Hydrodynamic limit

The fact that the loop expansion is an expansion in the small parameter δ_D makes possible an accurate calculation of the order-parameter decay rate $\gamma(\kappa,k)$ in the hydrodynamic regime $k \ll \kappa$, where κ^{-1} is the correlation length. This is the opposite regime from that studied in Sec. II A, where we were dealing with $\gamma(k) \equiv \gamma(0,k)$. We similarly generalize Eq. (2.6) to $\eta(\kappa,k)$, where $\eta(k) \equiv \eta(\kappa,k)\,|_{\kappa=0}$, and we define the universal amplitude ratio R by

$$\lim_{k\to 0} k^{-2}\gamma(\kappa,k) = R\frac{k_B T \kappa}{6\pi\eta(\kappa,0)} , \tag{2.17}$$

where T and k_B are the temperature and Boltzmann's constant, respectively. From the consideration of Sec. II A we see that the two-loop calculation of R is accurate to $O(\delta_D^2)$. We therefore expect its accuracy for $D=3$ to be better than 1%. In analogy with Eq. (2.12), we have

$$\frac{\pi}{3}R = \tilde{I}_3^{(1)}(z_\eta) + \frac{\delta_3}{a_\eta a_\gamma}\tilde{I}_3^{(2)}(z_\eta) + \frac{\delta_3^2}{a_\eta^2 a_\gamma^2}\tilde{I}_3^{(3)}(z_\eta) + \cdots , \tag{2.18}$$

where $\tilde{I}_D^{(n)}(z_\eta)$ is the integral $I_D^{(n)}(z_\eta)$ in the hydrodynamic limit. For the one-loop integral to zeroth order in z_η, corresponding to Eq. (2.14), we have (with the momentum scaled to $k=1$)

$$I_3 = \frac{1}{C_3}\int \frac{d^3p}{1+p^2}\frac{\sin^2\theta}{p^2} = \langle \sin^2\theta\rangle \int_0^\infty \frac{dp}{1+p^2} = \frac{\pi}{3} , \tag{2.19}$$

yielding the zeroth-order approximation

$$R = 1 + O(\delta_3) . \tag{2.20}$$

The $O(\delta_3)$ term comes from (i) $\tilde{I}_3^{(1)}(z_\eta)$ evaluated to $O(z_\eta)$ by making the appropriate self-energy insertion, and (ii) the vertex correction $\tilde{I}_3^{(2)}(0)$. The effect of space nonlocality in (i) has been treated by Burstyn *et al.*,[9] while the effect of nonlocality in time has been examined in detail by the present authors.[10] These two effects contribute $0.4\delta_3$ to (i). For (ii) we have found $0.2\delta_3$, giving the combined first-order result

$$R = 1 + 0.6\delta_3 \simeq 1.04 . \tag{2.21}$$

As stated before, this answer is expected to be accurate to better than 1%, which renders the value $R=1.2$ obtained by Siggia *et al.*[4] untenable. Experimental evidence for a value of R much closer to 1.0 than to 1.2 has been reported by Burstyn, Sengers, and Esfandiari[11] and by Güttinger and Cannell.[12]

III. ϵ EXPANSION

The value of z_η reported by Siggia *et al.*[4] from their renormalization-group calculation is inconsistent with Eq. (2.16), being some 22% too high. As already mentioned above in Sec. I, this error can be attributed to a too-early truncation of the ϵ expansion, as we will now demonstrate.

The two-term ϵ expansion of the numerator in Eq. (2.15) is

$$\delta_D = \frac{1}{(4-\epsilon)(6-\epsilon)} \simeq \frac{1}{24-10\epsilon} \simeq \frac{1}{24} + \frac{5}{288}\epsilon . \tag{3.1}$$

The denominator has the expansion

$$I_D = \frac{3}{4}\frac{1}{\epsilon} + c_0 + c_1\epsilon + \cdots , \tag{3.2}$$

where for the time being we neglect $c_1\epsilon$ and all higher terms. The leading term comes from applying the high momentum approximation $p' \simeq p$ to Eq. (2.14). The angle average then brings in $\langle \sin^2\theta\rangle = 1 - D^{-1} \simeq \frac{3}{4}$. To find c_0 it is useful to compare I_D with the simpler integral

$$\bar{I}_D = \frac{1}{C_D}\int \frac{d^D P}{p^2(1+p^2)} = \frac{1}{\epsilon} + O(\epsilon) . \tag{3.3}$$

Thus we have

$$I_D = \tfrac{3}{4}\bar{I}_D + (I_D - \tfrac{3}{4}\bar{I}_D) \simeq \tfrac{3}{4}\bar{I}_D + \lim_{D\to 4}(I_D - \tfrac{3}{4}\bar{I}_D) = \frac{3}{4}\frac{1}{\epsilon} + c_0 . \tag{3.4}$$

Evaluation of the subtracted integral gives, therefore,

$$c_0 = \lim_{D\to 4}(I_D - \tfrac{3}{4}\bar{I}_D) = \frac{1}{C_4}\int \frac{d^4p}{p^2}\left[\frac{\sin^2\theta}{p'^2} - \frac{3}{4}\frac{1}{p^2+1}\right] = \frac{1}{8} , \tag{3.5}$$

which yields the two-term truncation of the first-order exponent as

$$z_\eta = \frac{\delta_D}{I_D} \simeq \frac{\epsilon}{18} + \frac{\epsilon^2}{72} . \tag{3.6}$$

Evaluated at $D=3$ or $\epsilon=1$, Eq. (3.6) gives $z_\eta = 0.069$—clearly much too large and a grossly inaccurate approximation to the exact first-order value of 0.054.

The two-term truncation is obviously the source of the error in Eq. (3.6). It is clearly necessary to include at least one more term in the ϵ expansion. As we shall see, a three-term truncation of the ϵ expansion enormously improves the accuracy of the calculation of Siggia *et al.*[4] and brings it into agreement with Eq. (2.16). The computation of the third term in Eq. (3.2) is facilitated by the fact that I_D is a convolution integral which can be written in closed form. This is most conveniently carried out by the mathematical trick[13] of replacing $d^D p \sin^2\theta$ by $d^{D+2}p/p^2$

times an appropriate numerical factor. Thus we find

$$I_D=\frac{\pi}{4}\frac{D-1}{\sin(-\pi D/2)}\frac{\Gamma^2(D/2)}{\Gamma(D-1)}$$
$$=\frac{\pi}{4}\frac{3-\epsilon}{\sin(\pi\epsilon/2)}\frac{\Gamma^2(2-\epsilon/2)}{\Gamma(3-\epsilon)} \tag{3.7}$$

from which it follows

$$c_1=\frac{1}{4}+\frac{\pi^2}{32}-\frac{3}{16}\psi'(1)=\frac{1}{4}\ . \tag{3.8}$$

The last two terms of Eq. (3.8) cancel because the trigamma function is

$$\psi'(1)=\left.\frac{d^2\ln\Gamma(x)}{dx^2}\right|_{x=1}=\xi(2)=\frac{\pi^2}{6}\ . \tag{3.9}$$

Substituting Eq. (3.8) into Eq. (3.2) and carrying the expansion to three-term accuracy gives

$$z_\eta=\frac{\delta_D}{I_D}\simeq\frac{\epsilon}{18}+\frac{\epsilon^2}{72}-\frac{35\epsilon^3}{2592}\ . \tag{3.10}$$

Evaluated at $\epsilon=1$, the last two terms of Eq. (3.10) almost exactly cancel one another (the last one being $-\frac{35}{36}$ times the preceding one), yielding

$$z_\eta=0.056\ . \tag{3.11}$$

This is down 0.013 from the two-term value and now only 4% above the exact value of 0.054. This completes the demonstration that the error in the calculation by Siggia *et al.*[4] resulted from a too-early truncation of the ϵ expansion. A more complete picture is, however, provided by the plot of Eq. (3.10) versus ϵ as shown by the dot-dashed curve in Fig. 1. It should be noted that this has a zero at $\epsilon=2.61$. The solid curve, representing the exact ϵ dependence of z_η that follows from Eq. (3.7), has its zero at $\epsilon=2$. It is the existence of the zero and the attendant curving down of the function which has brought its strength in the vicinity of $\epsilon=1$ down and into good agreement with the solid curve. As discussed in Sec. IV, the occurrence of the zero is not accidental but is a necessary consequence of the "infrared divergence" at $D=2$. Although the actual location of the zero is not correct for the dot-dashed curve, this three-term approximation is in qualitative agreement with this basic requirement. This is not true for the two-term approximation, shown by the dashed curve in Fig. 1, which is the reason that it gives much too high a value at $\epsilon=1$. In other words, the underlying cause of the error in the Siggia *et al.*[4] calculation is that it completely ignores the infrared divergence of I_D.

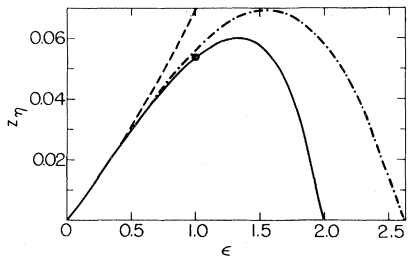

FIG. 1. Critical viscosity exponent z_η vs $\epsilon=4-D$, where D is the space dimensionality, in the single-loop approximation. Solid curve shows the exact function, with the value $z_\eta=8/15\pi^2=0.054$ for $D=3$ indicated by the solid dot. Dashed curve shows the 28% error in the two-term truncation by Siggia, Halperin, and Hohenberg (Ref. 4) of the ϵ expansion. Three-term truncation (dot-dashed curve) is more accurate at $\epsilon=1$ because its zero at 2.6 simulates the $\epsilon=2$ "infrared" zero of the exact function.

But before leaving the ϵ expansion we want to use it to illustrate the loop expansion in powers of δ_D that was discussed above in Sec. II. To show this qualitatively for the self-consistent self-energy insertion in the single-loop relaxation rate integral it will suffice to work to lowest order in the ϵ expansion. In this approximation, Eq. (2.14) is replaced by

$$I_D^{(1)}(z_\eta)=\frac{1}{C_D}\int\frac{d^Dp\sin^2\theta}{p^{2-z_\eta}p'^2}\simeq\frac{3}{4}\frac{1}{\epsilon-z_\eta} \tag{3.12}$$

which substituted into Eq. (2.13) gives

$$z_\eta\simeq\frac{4\delta_4}{3}(\epsilon-z_\eta)=\frac{1}{18}(\epsilon-z_\eta) \tag{3.13}$$

or

$$z_\eta=\frac{\epsilon}{19}\ . \tag{3.14}$$

Thus the first-order insertion has decreased z_η by an amount Δz_η which, evaluated at $\epsilon=1$, is

$$\Delta z_\eta=\frac{1}{18}-\frac{1}{19}=\frac{1}{18\times19}=O(\delta_4^2)\ , \tag{3.15}$$

as expected. This simple example is a prototype for the other two-loop corrections and serves to illustrate the essential point of this paper, which is the rapid convergence of the loop expansion in powers of δ_D. This rapid convergence is qualitatively apparent in Ref. 4. The last sentence of the note added in proof to Ref. 3, however, seems to contradict the rapid convergence of the loop expansion. Unfortunately, no details are given to support the value put forward there for z_η.

Paladin and Peliti[14] have recently put forward the value $z_\eta=\frac{3}{43}=0.070$, based on a one-loop calculation. No estimate is given for the error resulting from the use of the local approximation in this calculation.

IV. MITTAG-LEFFLER EXPANSION

In Sec. III we have demonstrated that the two-term ϵ expansion is grossly in error because it makes no allowance for the infrared divergence. Inspection of Eq. (2.14) reveals that the integral diverges at $D=2$. In the vicinity of $D=2$ its asymptotic behavior is governed by the simple pole

$$I_D\sim\frac{1}{2}\frac{1}{D-2}=\frac{1}{2}\frac{1}{\epsilon'}\ , \tag{4.1}$$

where $\epsilon'\equiv D-2$. Carrying out a subtraction integration,

or appealing to Eq. (3.7), yields the two-term "ϵ' expansion"

$$I_D=\frac{1}{2}\frac{1}{\epsilon'}+\frac{1}{2}\ . \tag{4.2}$$

To develop an appreciation of the extent to which the ϵ expansion can be misleading for $D=3$ problems it is important to recognize that for $D=3$ the infrared pole at $\epsilon'=0$ is as important for I_D as the ultraviolet pole at $\epsilon=0$. In this case $\epsilon=\epsilon'=1$, so that both poles are equally close. For this reason it is necessary to treat both of the neighboring poles on an equal basis and to subtract both of them at the same time, so as to arrive at the function

$$K(\epsilon)=I_D-\frac{3}{4}\frac{1}{\epsilon}-\frac{1}{2}\frac{1}{\epsilon'}\ , \tag{4.3}$$

analytic in the "physical" range $0\leq\epsilon\leq 2$. From Eqs. (3.2), (3.5), and (4.2), the limiting values of $K(\epsilon)$ are

$$K(0)=-\tfrac{1}{8} \tag{4.4a}$$

and

$$K(2)=\tfrac{1}{8}\ , \tag{4.4b}$$

respectively. The two-term ϵ expansion for $K(\epsilon)$ satisfying Eqs. (4.4a) and (4.4b) is

$$K(\epsilon)=-\frac{1}{8}+\frac{\epsilon}{8}\ . \tag{4.5}$$

Solving for I_D from Eq. (4.3) gives then the two-pole Mittag-Leffler expansion

$$I_D=\frac{3}{4}\frac{1}{\epsilon}+\frac{1}{2}\frac{1}{\epsilon'}+K(\epsilon)=\frac{3}{4}\frac{1}{\epsilon}+\frac{1}{2}\frac{1}{2-\epsilon}-\frac{1}{8}+\frac{\epsilon}{8}\ , \tag{4.6}$$

which does not differ appreciably from Eq. (3.7). The latter is shown by the solid curve in Fig. 2. Evaluated at $\epsilon=1$, Eq. (4.6) yields

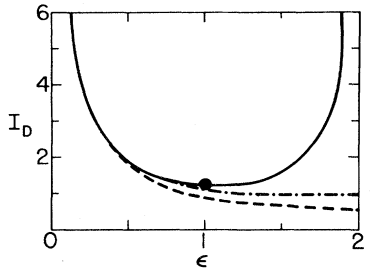

FIG. 2. Single-loop integral I_D vs $\epsilon=4-D$, where D is the space dimensionality. Solid curve shows the exact function, with the value $I_3=\pi^2/8$ indicated by the solid dot. The 30% error of the two-term truncation by Siggia, Halperin, and Hohenberg (Ref. 4) of the ϵ expansion is indicated by the dashed curve. Although the three-term truncation (dot-dashed curve) is better, it also does not take into account the infrared divergence at $\epsilon=2$, a shortcoming that is rectified by the two-pole Mittag-Leffler expansion. The latter agrees with the solid curve, within the accuracy of the plot.

$$I_3=\tfrac{5}{4}=1.25\ , \tag{4.7}$$

only 1.5 % off the exact value of 1.234 of Eq. (2.15). By comparison, the two- and three-term truncations of the ϵ expansion yield, according to Eqs. (3.2), (3.5), and (3.8), the much smaller values of $\frac{7}{8}=0.875$ and $\frac{9}{8}=1.125$, respectively. The latter value comes from setting $\epsilon=1$ in

$$I_D=\frac{3}{4}\frac{1}{\epsilon}+\frac{1}{8}+\frac{\epsilon}{4}\ . \tag{4.8}$$

It is interesting to note that the third term in Eq. (4.8) provides the further constant

$$K'(0)=\tfrac{1}{8}\ , \tag{4.9}$$

where the prime denotes differentiation. This makes it possible to determine one more term in Eq. (4.5). But because the two-term expression for $K(\epsilon)$ already satisfies Eq. (4.9) it follows that the ϵ^2 term in the Taylor series for $K(\epsilon)$ vanishes.

The good accuracy of the truncated Mittag-Leffler expansion of Eq. (4.6) results from the fact that the poles of I_D that have been neglected are relatively far from the physical point $\epsilon=1$. These are simple poles at $\epsilon=-2,-4,-6,\ldots$, and double poles at $\epsilon=4,6,\ldots$. They are sufficiently far removed that they are well represented in the physical region $0\leq\epsilon\leq 2$ by a rapidly convergent Taylor series in ϵ. This advantage of the Mittag-Leffler expansion has been demonstrated by us[15] in detail for the λ transition in liquid helium.

The above considerations can be extended to z_η itself. $z_\eta(\epsilon)$ has no poles along the negative ϵ axis and its first pole for $\epsilon>0$ comes at $\epsilon=3$. As noted in Fig. 1, the infrared pole at $\epsilon=2$, which "spoils" the ϵ expansion for I_D, is converted into a zero of $z_\eta(\epsilon)$. The fact that the nearest pole is farther from $\epsilon=1$ for $z_\eta(\epsilon)$ than for I_D evidently accounts for the fact that the three-term ϵ-expansion error of 4% for $z_\eta(\epsilon)$ is smaller than the corresponding 10% error in the three-term ϵ expansion for I_D. But in any case, it is clear that a truly accurate determination of z_η depends upon a proper evaluation of I_3 in which account is taken of the effect of the infrared pole.

To summarize this section, we have demonstrated from general considerations why the ϵ expansion cannot be relied upon to give accurate results. Although the error in the two-term expansion of Siggia *et al.*[4] is immediately evident from a comparison of Eq. (3.6) with the exact value of Eq. (2.15), our goal here has been to provide a more basic understanding of how this error comes about and why, in a sense, it is inevitable.

V. EXPERIMENTAL SITUATION

Burstyn *et al.*[11] have reviewed the various experimental determinations of the universal amplitude ratio and have emphasized that R is much closer to 1.0, the single-loop value, than it is to 1.2, the value put forward by Siggia *et al.*[4] The measurements of Burstyn *et al.*[11] give $R=1.02\pm0.06$, which is compatible with our result in Eq. (2.22) of $R=1.04\pm0.01$.

Turning now to the critical exponent, the most direct determination of z_η comes, of course, from the tempera-

ture dependence of the hydrodynamic viscosity. But this requires first the subtraction of the noncritical background. Unfortunately, although the latter is not singular at the critical point, it generally has nevertheless a rather strong temperature dependence. It is not always possible to estimate *a priori* the latter in an unambiguous fashion, which renders the subtraction and the ensuing extraction of z_η from the remainder somewhat uncertain. An additional experimental problem is that the hydrodynamic measurements bring the fluid out of equilibrium. The perturbation is more severe the closer the critical point is approached.

Because of the difficulties described above, there is some advantage to inferring z_η from light scattering linewidth measurements. These yield the nonlocal diffusion coefficient

$$D(\kappa,k)=k^{-2}\gamma(\kappa,k) \tag{5.1}$$

as a function of κ and k. Because these measurements can be carried out very close to the critical point, their interpretation is much less sensitive to the background subtraction and nonequilibrium problems associated with the hydrodynamic measurements. At the critical point

$$D(0,k)\propto k^{\tilde{y}} \tag{5.2}$$

with the exponent

$$\tilde{y}=1+z_\eta\ . \tag{5.3}$$

Away from the critical point Burstyn and Sengers[5] were able to determine values of a κ-dependent generalization of $\tilde{y}$ by fitting Eq. (5.2) to the linewidth data from three different angles, for the same value of κ. From general considerations we expect $\tilde{y}$ to be a function of

$$\kappa^2\propto(T-T_c)^{2\nu}\ , \tag{5.4}$$

where T_c is the critical temperature and ν is the correlation length critical exponent. For this reason we have plotted in Fig. 3 the Burstyn-Sengers[5] data versus $(T-T_c)^{2\nu}$, with the temperatures measured in mK and with ν set equal to 0.63. The straight-line fit to the data confirms the expected linear dependence on κ^2 in the vicinity of the critical point. The intercept of $\tilde{y}=1.05\pm0.02$ is, according to Eq. (5.3), consistent with our theoretical expectation and constitutes good verification of Eq. (3.16), within the accuracy of the measurements (as shown by the error bars). It should be noted that the largest value of $\tilde{y}$ actually measured by Burstyn and Sengers[5] was 1.048 at $T-T_c=0.3$ mK. Their inferred value of $\tilde{y}$ at $T=T_c$ was 1.06 ± 0.02 because of an extrapolation which was linear in $T-T_c$ instead of according to Eq. (5.4).

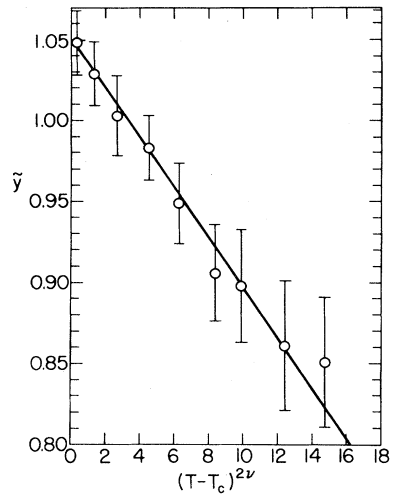

FIG. 3. Effective diffusion exponent data of Burstyn and Sengers (Ref. 5) vs $(T-T_C)^{2\nu}$, where the temperatures are in mK. $\nu=0.63$ is the correlation length critical exponent. Extrapolation of the straight line fit to the critical point gives $\tilde{y}=1+z_\eta=1.051$ in exact agreement with the theoretical prediction, Eq. (3.16).

VI. SUMMARY

The purpose of our paper has been to establish that high-precision calculations can be performed for the critical dynamics of a classical fluid. This involves abandoning the popular ϵ expansion and evaluating the loop integrals directly in three dimensions. The existence of the small expansion parameter δ_D independent of ϵ, which is a special feature of this problem, makes the loop expansion for $D=3$ converge rapidly. A two-loop calculation yields accuracy to $O(\delta_D^2)$. Because $\delta_3=\frac{1}{15}$, the accuracy of a two-loop calculation is of the order of 1%. In this scheme the critical exponent z_η is near $8/15\pi^2=0.054$ and the universal amplitude ratio is $R=1.04$. We contend that the corresponding values advocated by Siggia, Halperin, and Hohenberg,[4] namely, 0.065 and 1.2, are erroneously high. The error in z_η is due to their use of the inaccurate two-term ϵ expansion of the single-loop relaxation rate integral, which overestimates the value of the integral by 28%. Because the second-order contribution to z_η is smaller by more than one order of magnitude, the ϵ expansion error in it is acceptable. Correcting only the first-order contribution would yield a corrected net exponent of $z_\eta=0.050$. If we, on the other hand, assume (as indicated by our own second-order calculations) that the second-order contribution has about the same error as the large first-order term, then we can apply the correction factor of $0.054/0.069=0.78$ to the quoted DRG result[4] to obtain the net corrected exponent as $z_\eta=0.051$. Obviously the way in which the second-order contribution is handled has only the minor effect of 0.001 on z_η. The important thing is to base the calculation on an accurate first-order contribution, since this is by far the predominant part. Once this is done, the best estimate for z_η, within the present theoretical framework, is only slightly below

the old decoupled mode value of $8/15\pi^2$.

In closing, we take note of a so far unexplored source of error in the calculation of z_η. This results from Eq. (2.1) and from the neglect of the anomalous dimension exponent associated with the fourth-order interaction term in the Ginzburg-Landau free-energy functional. Clearly more work needs to be done on this point.

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