

$$\text{or } \underline{s} = \begin{pmatrix} 0 \\ 0 \\ \frac{2kc^2}{\omega_p^2} \\ \frac{i2kc^2}{\omega_p^2} \end{pmatrix}. \quad (\text{A10})$$

Now we go to order  $\epsilon^3$ . Substituting Eqs. (A4) and (A9) into (A3) and multiplying the resulting equations by  $\underline{l}$  from left we obtain

$$\underline{l}(W_2 - v_g W_3) \underline{s} \frac{\partial^2 \varphi_1}{\partial \xi^2} + \underline{l} W_4 r \varphi_1 + \underline{l} W_5 r \frac{\partial^2 \varphi_1}{\partial \xi^2}$$

$$+ \underline{l} W_6 r \frac{\partial^2 \varphi_1}{\partial \eta^2} + \underline{l} W_7 r \frac{\partial \varphi_1}{\partial \tau} = 0,$$

which, after some manipulation, reduces to the expression shown as Eq. (14) in the text.

Although the ion dynamics is ignored here it is not difficult to include it. In this case, the dispersion relation changes, but it can be shown that if a suitable group velocity and wave number are used, the final expression [Eq. (14)] is still valid.

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<sup>2</sup>R. Y. Chiao, E. Garmire, and C. H. Townes, *Phys. Rev. Letters* **13**, 479 (1964).

<sup>3</sup>T. Taniuti and H. Washimi, *Phys. Rev. Letters* **21**, 209 (1968).

<sup>4</sup>C. K. W. Tam, *Phys. Fluids* **12**, 1028 (1969).

<sup>5</sup>For example, V. I. Karpman and E. M. Krushkal', *Zh. Eksperim. i Teor. Fiz.* **55**, 530 (1968) [*Soviet Phys. JETP* **28**, 277 (1969)], where a derivation of nonlinear Schrödinger equation is made assuming an amplitude-dependent frequency of the modulated wave.

<sup>6</sup>V. N. Oraevskii and R. Z. Sagdeev, *Zh. Tekhn. Fiz.* **32**, 1291 (1962) [*Soviet Phys. Tech. Phys.* **7**, 955 (1963)].

## Sound Attenuation and Dispersion near the Liquid-Gas Critical Point

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The attenuation and dispersion of sound near the gas-liquid critical point are studied theoretically using the author's extended mode-mode coupling theory. The results differ in the different regions of the sound-wave frequency  $f$  expressed in a dimensionless unit and of  $\epsilon$ , the dimensionless temperature distance from the critical point. The attenuation behaves as  $f^2 \epsilon^{-3\nu-\alpha/2}$  for  $0 \leq f \ll \epsilon^{3\nu}$ , and as  $f^{2-2p/3} \epsilon^{-3\alpha/2}$  for  $\epsilon^{3\nu} \ll f \ll \epsilon^\nu$ , where  $p$  is the exponent which appears in the wave-number ( $\vec{k}$ )-dependent correlation of the order parameter expressed as  $A_1 k^{-2+\eta} + A_2 \epsilon^{1-\alpha} k^{-2+\eta-\rho}$ , when  $k$  is much greater than the inverse correlation range of critical fluctuations. The relative sound-velocity change with  $f$  behaves as  $f^{3/2} \epsilon^{-2\nu/2}$  for  $0 \leq f \ll \epsilon^{3\nu}$ , as  $f^{1-2p/3} \epsilon^{-\alpha}$  if  $p \leq \frac{3}{2}$ , and as  $f^0 \epsilon^0$  if  $p > \frac{3}{2}$  for  $\epsilon^{3\nu} \ll f \ll \epsilon^{\nu+\alpha/2}$ . The explicit expressions for the attenuation and dispersion are given for  $f \sim \epsilon^{3\nu}$ .

### I. INTRODUCTION

In recent years the sound attenuation and dispersion near the critical points have attracted an increasing amount of attention as a means of studying dynamics of critical fluctuations.<sup>1,2</sup> In particular, the first successful theoretical study of sound attenuation and dispersion near the liquid-gas critical points was carried out in 1965 by Botch and Fixman.<sup>3</sup>

After 1965, there has been a considerable prog-

ress in our understanding of the dynamics of critical fluctuations.<sup>4</sup> In particular, Kadanoff and Swift's brilliant application<sup>5</sup> of the mode-mode coupling theory<sup>6</sup> to the liquid-gas transition yielded valuable information on the divergences of various transport coefficients near the transition. This progress made it necessary to reconsider the problem of the sound attenuation and dispersion near the liquid-gas transition. Thus, it is the purpose of the present paper to study this problem in some detail using the extended version of the

author's mode-mode coupling theory<sup>7</sup> which is valid in the nonhydrodynamical regime as well.

In Sec. II we study the dynamics of the order-parameter fluctuation (the entropy fluctuation in this case), and obtain the decay rate of the fluctuation which is valid in the nonhydrodynamical regime as well. In Sec. III the results of Sec. II are used to find the frequency-dependent complex transport coefficients that enter the sound attenuation and dispersion. In Secs. IV and V the behaviors of the sound attenuation and dispersion are studied in various frequency and temperature regions near the critical point, which restricted us to the cases where the sound-wave length is very much greater than the correlation range of critical fluctuations. Since the work is closely related to that of Kadanoff and Swift,<sup>5</sup> we shall often quote their work simply as KS in the following.

## II. ORDER PARAMETER DYNAMICS

Although the order parameter associated with the liquid-gas transition is the density, the entropy fluctuation dominates the dynamics of the density fluctuation near the critical point.<sup>8</sup> Thus, in this section we study the dynamical behavior of the entropy fluctuations employing our general theory.<sup>7</sup> For this purpose we need the kinetic equations for the entropy fluctuation  $\delta S_{\vec{q}}$  and the transverse component of the local velocity  $v_{\vec{q}}^{\sigma}$  where  $\sigma = x, y, z$ . The forms of nonlinear terms in these kinetic equations are derived in Appendix A, and we have

$$\dot{S}_{\vec{q}} = -\frac{q^2 \lambda_0}{C_p^q} \delta S_{\vec{q}} - \sum_{\sigma} i q^{\sigma} V^{-1/2} \sum_{\vec{k}}' \delta S_{\vec{k}} v_{\vec{q}-\vec{k}}^{\sigma} + f_{\vec{q}}^S, \quad (2.1)$$

$$\dot{v}_{\vec{q}}^{\sigma} = -\frac{\eta_0}{\rho} q^2 v_{\vec{q}}^{\sigma} - \frac{T}{2\rho V^{1/2}} \sum_{\vec{k}}' i \left( k^{\sigma} - \frac{\vec{q} \cdot \vec{k}}{q^2} q^{\sigma} \right) \times \left( \frac{1}{C_p^k} - \frac{1}{C_p^{\vec{q}-\vec{k}}} \right) \delta S_{\vec{k}} \delta S_{\vec{q}-\vec{k}} + f_{\vec{q}}^v, \quad (2.2)$$

where  $\lambda_0$  and  $\eta_0$  are the nonanomalous parts of thermal conductivity and shear viscosity, respectively.  $C_k^p$  is the  $k$ -dependent heat capacity per unit volume at constant pressure,  $\rho$  is the mass density, and  $V$  is the volume of the system. The  $f$ 's are the random forces acting upon  $S_{\vec{q}}$  and  $v_{\vec{q}}^{\sigma}$ , and are related to  $\lambda_0$  and  $\eta_0$  by the familiar fluctuation-dissipation theorem.<sup>7,9</sup>  $\sum_{\vec{k}}'$  designates the sum over  $k$  which is smaller than some cutoff wave number, so that the hydrodynamical concepts employed remain valid.

The pair of equations (2.1) and (2.2) for  $\delta S_{\vec{q}}$  and  $v_{\vec{q}}^{\sigma}$  is essentially identical to the corresponding pair of equations for the concentration fluctuation  $\delta c_{\vec{q}}$  and  $v_{\vec{q}}^{\sigma}$  of the binary solution [see Eqs. (3.27) of Ref. 7]. The analogy between the two problems has been noted by Swift<sup>10</sup> previously, and we can

thus transcribe the results obtained for the binary mixture<sup>7</sup> to the present problem. We thus conclude that  $\langle \delta S_{\vec{q}}(t) \delta S_{-\vec{q}}(0) \rangle / \langle |\delta S_{\vec{q}}|^2 \rangle = e^{-\Gamma_{\vec{q}} t}$  ( $t > 0$ ) with

$$\Gamma_{\vec{q}} = \frac{k_B T}{\eta} \frac{1}{(2\pi)^3} \int d\vec{k} \left[ \left( \frac{q}{k} \right)^2 - \left( \frac{\vec{q} \cdot \vec{k}}{k^2} \right)^2 \right] \frac{C_p^{\vec{q}-\vec{k}}}{C_p^{\vec{k}}}, \quad (2.3)$$

where the shear viscosity  $\eta$  remains finite and is insensitive to  $\omega$  and  $k$ . If we use the Ornstein-Zernike form for  $C_k^p \propto (k^2 + \kappa^2)^{-1}$  with  $\kappa$  as the inverse correlation range of density fluctuations, (2.3) reduces to<sup>11</sup>

$$\Gamma_{\vec{q}} = D \kappa^3 K(q/\kappa), \quad (2.4)$$

where  $D \equiv k_B T / 6\pi\eta$

$$\text{and } K(x) \equiv \frac{3}{4} [1 + x^2 + (x^3 - x^{-1}) \tan^{-1} x]. \quad (2.5)$$

For  $x \ll 1$ ,  $K(x) = x^2 + \dots$ , and thus for  $q \ll \kappa$ ,  $\Gamma_{\vec{q}} = D \kappa q^2$ . Therefore  $D \kappa$  is identical to the thermal diffusion constant  $\lambda / C_p$ , which can be directly determined by the inelastic-light-scattering experiment.<sup>12</sup> Since the shear viscosity is expected to be finite, so is  $D$ . This behavior of  $\lambda$  was first found by KS.

## III. FREQUENCY-DEPENDENT COMPLEX TRANSPORT COEFFICIENTS

In Sec. II, we have studied the heat diffusion mode. Other hydrodynamic modes that couple to the sound wave are the viscous mode and the sound-propagation mode,<sup>5</sup> both of which are, at most, only weakly critical. Thus we are ready to study the sound-wave damping, or equivalently, the frequency-dependent transport coefficients entering the sound-wave damping. These are  $\theta(\omega) = \zeta(\omega) + \frac{4}{3}\eta(\omega)$ , where  $\zeta$  and  $\eta$  are the bulk and shear viscosities, respectively, and the thermal conductivity  $\lambda(\omega)$ .

The sound absorption coefficient  $\hat{\alpha}(\omega)$  and the dispersion  $\Delta c(\omega) = c(\omega) - c$  [where  $c(\omega)$  is the sound velocity at the frequency  $\omega$ , and  $c$  is the zero-frequency sound velocity] are expressed in terms of these transport coefficients as

$$\hat{\alpha}(\omega) = \frac{\omega^2}{c^3} \left[ \frac{1}{\rho} \text{Re}\theta(\omega) + \left( \frac{1}{C_V} - \frac{1}{C_P} \right) \text{Re}\lambda(\omega) \right], \quad (3.1)$$

$$\frac{\Delta c(\omega)}{c} = \frac{\omega}{2c^2} \left[ \frac{1}{\rho} \text{Im}\theta(\omega) + \left( \frac{1}{C_V} - \frac{1}{C_P} \right) \text{Im}\lambda(\omega) \right], \quad (3.2)$$

where  $C_V$  and  $C_P$  are the specific heats per unit volume at constant volume and pressure, respectively. Since the sound-wave frequency of our interest  $\omega$  satisfies  $\omega/c \ll \kappa$ , we only have to take into account the frequency dependence in (3.1) and (3.2).

In the following we consider the two heat-modes contribution  $\theta_T(\omega)$ , the two sound-waves contribution  $\theta_p(\omega)$ , the two viscous-modes contribution  $\theta_v(\omega)$ , and the contribution of the viscous mode-

heat mode,  $\lambda_{\eta T}(\omega)$ .

According to KS (3.23), we have

$$\theta_T(\omega) = \frac{1}{2k_B T} \int \frac{d\vec{k}}{(2\pi)^3} \frac{|L_{\vec{q}, \vec{k}}/q|^2}{\Gamma_{\vec{k}} + \Gamma_{\vec{q}-\vec{k}} - i\omega} \quad (3.3)$$

$$\text{with } L_{\vec{q}, \vec{k}} \cong -\rho \langle v_{-\vec{q}}^z \hat{a}_2(\vec{q}) \rangle \langle a_2(-\vec{q}) a_1(\vec{k}) a_1(\vec{q}-\vec{k}) \rangle, \quad (3.4)$$

where  $\vec{q}$  is the wave vector of the sound wave and is taken to be along the  $z$  direction.  $a_1$  and  $a_2$  are defined in KS and are related to entropy and pressure fluctuations  $\delta S_{\vec{k}}$  and  $\delta P_{\vec{k}}$ , respectively, as (see also Ref. 13)

$$\begin{aligned} a_1(\vec{k}) &= \delta S_{\vec{k}} / (k_B C_{\vec{k}}^P)^{1/2}, \quad (3.5) \\ a_2(\vec{k}) &= \frac{(\rho/k_B T)^{1/2}}{n} c \delta n_{\vec{k}} + \left[ \frac{1}{k_B} \left( \frac{1}{C_k^V} - \frac{1}{C_k^P} \right) \right]^{1/2} \delta S_{\vec{k}} \\ &= c \chi_S (\rho/k_B T)^{1/2} \delta P_{\vec{k}}, \quad (3.6) \end{aligned}$$

where  $C_k^V$  is the  $k$ -dependent specific heat at constant volume,  $\chi_S$  is the adiabatic compressibility, and  $n$  is the number density.

Since we can take the limit  $q \rightarrow 0$  in (3.3), we have<sup>14</sup>

$$\begin{aligned} \langle a_2(-\vec{q}) a_1(\vec{k}) a_1(\vec{q}-\vec{k}) \rangle \\ \cong (\rho k_B T)^{1/2} \frac{c \chi_S T}{C_V} \left( \frac{\partial P}{\partial T} \right)_V \left( \frac{\partial \ln C_{\vec{k}}^P}{\partial T} \right)_S, \quad (3.7) \end{aligned}$$

and thus, using  $\langle v_{-\vec{q}}^z \hat{a}_2(\vec{q}) \rangle = -iqc(k_B T/\rho)^{1/2}$ , we obtain

$$L_{\vec{q}, \vec{k}} \cong iq \frac{k_B T^2}{C_V} \left( \frac{\partial P}{\partial T} \right)_V \left( \frac{\partial \ln C_{\vec{k}}^P}{\partial T} \right)_S. \quad (3.8)$$

Using the Ornstein-Zernike form for  $C_{\vec{k}}^P \propto (k^2 + \kappa^2)^{-1}$  we have finally<sup>15</sup>

$$\begin{aligned} \theta_T(\omega) &= \frac{2k_B T^3}{C_V^2} \left( \frac{\partial P}{\partial T} \right)_V^2 \kappa^2 \left( \frac{\partial \kappa}{\partial T} \right)_S^2 \\ &\times \int \frac{d\vec{k}}{(2\pi)^3} \frac{1}{(\kappa^2 + k^2)^2} \frac{1}{2\Gamma_{\vec{k}} - i\omega}. \quad (3.9) \end{aligned}$$

Substituting the result (2.4) into (3.9), we obtain the following form for  $\theta_T(\omega)$ , which is more convenient for numerical computation,

$$\begin{aligned} \theta_T(\omega) &= \frac{k_B T^3}{\pi^2 C_V^2} \left( \frac{\partial P}{\partial T} \right)_V^2 \kappa \left( \frac{\partial \kappa}{\partial T} \right)_S^2 \\ &\times \int_0^\infty dx \frac{x^2}{(1+x^2)^2} \frac{1}{2D\kappa^3 K(x) - i\omega}. \quad (3.10) \end{aligned}$$

Next we consider the two sound-waves contribution  $\theta_p(\omega)$ . According to KS, Sec. III G, we find

$$\begin{aligned} \theta_p(\omega) &= \frac{1}{2k_B T} \sum_{s, s' = \pm 1} \int \frac{d\vec{k}}{(2\pi)^3} \\ &\times \frac{|H_{\vec{q}, \vec{k}}/q|^2}{icsk + ics'|\vec{q}-\vec{k}| + \gamma_{\vec{k}} + \gamma_{\vec{q}-\vec{k}} - i\omega}, \quad (3.11) \end{aligned}$$

where we have added the sound-wave damping constants  $\gamma_{\vec{k}}$  and  $\gamma_{\vec{q}-\vec{k}}$  in the denominator and

$$\begin{aligned} H_{\vec{q}, \vec{k}} &\cong \frac{1}{2} \rho k_B T \langle \{v_{-\vec{q}}^z, a_2(\vec{q})\} \rangle \\ &\times \langle a_2(-\vec{q}) a_2(\vec{k}) a_2(\vec{q}-\vec{k}) \rangle. \quad (3.12) \end{aligned}$$

For  $q \ll k$ , we may set  $a_2(-\vec{q}) = (\rho/k_B T)^{1/2} c \chi_S \Delta P$  by (3.3) with  $\Delta P$ , the pressure fluctuation. Thus we have, as for (3.8),

$$H_{\vec{q}, \vec{k}} \cong iq \frac{\chi_S}{2C_V} k_B T^2 \left( \frac{\partial P}{\partial T} \right)_V \left( \frac{\partial [\chi_S(\vec{k})]^{-1}}{\partial T} \right)_S, \quad (3.13)$$

where  $\chi_S(\vec{k})$  is the  $\vec{k}$ -dependent adiabatic compressibility which is related to Fourier-transformed local-pressure fluctuations  $\delta P_{\vec{k}}$  by  $k_B T / \langle |\delta P_{\vec{k}}|^2 \rangle$ . The real and imaginary parts of  $\theta_p(\omega)$  have been analyzed in detail in Appendix B, where the damping of intermediate-state sound waves was found out to be important.

Next, we also considered the process in which two viscous modes contribute to  $\theta$ . Essentially the same type of analysis as before yields a contribution  $k_B T \rho \kappa / \eta$  to  $\theta$  and is always negligible.

Finally, we turn our attention to  $\lambda_{\eta T}(\omega)$ , which is very closely related to  $\Gamma_{\vec{q}}$ . In fact, we have

$$\lambda_{\eta T}(\omega) = \frac{k_B T}{\rho} \int \frac{d\vec{k}}{(2\pi)^3} C_{\vec{q}-\vec{k}}^P \frac{1 - (k_z/k)^2}{(\eta/\rho)k^2 - i\omega}, \quad (3.14)$$

$$\text{and } \Gamma_{\vec{q}} = q^2 \lambda_{\eta T}(0) / C_{\vec{q}}^P.$$

#### IV. SOUND ATTENUATION

We first examine the sound attenuation arising from  $\theta_T(\omega)$  which we denote as  $\hat{\alpha}_T(\omega)$ . From (3.1) and (3.10) we immediately find

$$\begin{aligned} \hat{\alpha}_T(\omega) &= \frac{k_B T^3}{\pi^2 \rho c^3 C_V^2} \left( \frac{\partial P}{\partial T} \right)_V^2 D \kappa^4 \left( \frac{\partial \kappa}{\partial T} \right)_S^2 \omega^2 \\ &\times \int_0^\infty dx \frac{x^2}{(1+x^2)^2} \frac{K(x)}{\omega^2 + 4D^2 \kappa^6 K^2(x)}. \quad (4.1) \end{aligned}$$

For  $\omega \ll D\kappa^3$ , we find<sup>16</sup>

$$\hat{\alpha}_T(\omega) \propto \frac{1}{c^3 C_V^2} \left( \frac{\partial \ln \kappa}{\partial T} \right)_S^2 f^2 \propto \epsilon^{(\alpha/2) - 2} f^2, \quad (4.2)$$

where  $\epsilon \equiv |T - T_c|/T_c$  and we introduced the dimensionless frequency  $f = \omega/\omega_0$ , with  $\omega_0$  as some microscopic frequency  $\sim 1/\eta a^3$ ,  $a$  being the average intermolecular distance. If the scaling-law relation  $2 = 3\nu + \alpha$  is valid,

$$\hat{\alpha}_T(\omega) \propto \epsilon^{-3\nu - \alpha/2} f^2. \quad (4.3)$$

This result is essentially the same as that of KS in the low-frequency region  $0 \ll f \ll \epsilon^{3\nu}$ .

For higher frequencies  $f \gg \epsilon^{3\nu}$ , it may appear that  $4D^2 \kappa^6 K^2(x)$  in the denominator of (4.1) is negligible compared with  $\omega^2$ . This, however, is not the case. Since  $K(x) \approx (\frac{3}{8}\pi)x^3$  for  $x \gg 1$ , if we neg-

lect  $4D^2\kappa^6K^2(x)$ , the integral over  $x$  diverges at large  $x$ . Namely, here the fluctuations with  $k \gg \kappa$  give major contributions to  $\alpha_T(\omega)$ . This contribution was not considered in the earlier work of the author.<sup>13</sup>

Now, for  $k \gg \kappa$ , Ferer, Moore, and Wortis<sup>17</sup> have shown recently that the order-parameter correlation, or  $C_E^P$  in our case, has the following form,

$$C_E^P = A_1 k^{-2+\eta} + A_2 \epsilon^{1-\alpha} k^{-2+\eta-p}, \quad (4.4)$$

where  $A_1$  and  $A_2$  are some constants, and  $p = (1-\alpha)/\nu$  if the strong scaling holds for  $C_E^P$ . For the three-dimensional Ising ferromagnet,  $p \cong 1.51$ .<sup>17</sup> Hence, we have for  $k \gg \kappa$ ,

$$T_c \left( \frac{\partial \ln C_E^P}{\partial T} \right)_S = \frac{A_2}{A_1} \epsilon^{-\alpha} k^{-p}. \quad (4.5)$$

Note that this is quite different from  $\kappa(\partial\kappa/\partial T)_S k^{-2} \sim \epsilon^{2\nu-1} k^{-2}$  that follows from the Ornstein-Zernike form for  $C_E^P$ , and  $(\partial C_E^P/\partial T)_S$  is the only quantity we need which is sensitive to the form of  $C_E^P$ , [(4.4)]. We thus find

$$\theta_T(\omega) \sim \int_{\kappa}^{\infty} \frac{dk}{k^{2p-2}} \frac{1}{2\Gamma_k - i\omega}, \quad (4.6)$$

provided that  $k \gg \kappa$  gives main contributions to the integral, and

$$\hat{\alpha}_T(\omega) \sim \epsilon^{-3\alpha/2} \omega^2 \int_{\kappa}^{\infty} \frac{dk}{k^{2p-2}} \frac{2\Gamma_k}{(2\Gamma_k)^2 + \omega^2}. \quad (4.7)$$

Indeed, as in (4.1) the main contributions to (4.7) come from  $k \gg \kappa$  which can be estimated by introducing the frequency-dependent cutoff  $k_f \sim f^{1/3}$  defined by  $2\Gamma_{k_f} = \omega$ . In this way we obtain

$$\hat{\alpha}_T(\omega) \propto \epsilon^{-3\alpha/2} f^{2-2p/3}, \quad (\epsilon^{\nu+\alpha/2} \gg f \gg \epsilon^{3\nu}). \quad (4.8)$$

If we assume strong scaling  $p = (1-\alpha)/\nu$ ,

$$\hat{\alpha}_T(\omega) \propto \epsilon^{-3\alpha/2} f^{2-2(1-\alpha)/3\nu}, \quad (\epsilon^{\nu+\alpha/2} \gg f \gg \epsilon^{3\nu}) \quad (4.8')$$

or for  $\alpha = 0$  (hence  $\nu = \frac{2}{3}$ )<sup>18</sup>

$$\hat{\alpha}_T(\omega) \propto \epsilon^0 f, \quad (\epsilon^{2/3} \gg f \gg \epsilon^2). \quad (4.8'')$$

For  $f \sim \epsilon^{3\nu}$ , we may find  $\hat{\alpha}_T(\omega)$  by integrating (4.1) numerically. Unfortunately, the closed expression (4.1) does not continue smoothly into the high-frequency result (4.8) due to the fact that the Ornstein-Zernike form for  $C_E^P$  does not go into (4.4) for  $k \gg \kappa$ .

Here it is appropriate to remark about our ignoring the frequency dependence of  $\Gamma_k$  which is valid as long as the condition

$$\omega \ll (\eta/\rho) l^2 \quad (4.9)$$

is satisfied where  $l$  is the wave number of fluctu-

ations that give major contributions to  $\Gamma_k$  [see (3.14)]. In the low-frequency region  $f \ll \epsilon^{3\nu}$ ,  $l \sim \kappa$  and (4.6) is satisfied. In the high-frequency region  $f \gg \epsilon^{3\nu}$ ,  $l \gtrsim k_f = \kappa x_{\omega} \sim \alpha^{-1} f^{1/3}$  and the condition (4.9) is satisfied if  $f \ll f^{2/3}$ , which is again satisfied since  $f \ll 1$ .

The contribution of  $\theta_p(\omega)$  to the attenuation  $\hat{\alpha}_p(\omega)$  is obtained using the result of Appendix B and is for  $f \ll \epsilon^{\nu}$ ,

$$\hat{\alpha}_p(\omega) \sim \epsilon^{-\nu-\alpha} f^2, \quad (4.10)$$

in agreement with KS.

The  $\lambda_{\eta T}(\omega)$  gives the following contribution  $\hat{\alpha}_{\lambda}(\omega)$  to the sound attenuation:

$$\hat{\alpha}_{\lambda}(\omega) = \omega^2 (k_B T / \rho c^3 C_V) \times \int \frac{d\vec{k}}{(2\pi)^3} C_{\vec{k}-\vec{k}}^P \left[ 1 - \left( \frac{k_x}{k} \right)^2 \right] \frac{(\eta k^2 / \rho)}{\omega^2 + (\eta k^2 / \rho)^2}. \quad (4.11)$$

In the low-frequency region  $f \ll \epsilon^{2\nu}$ , we have

$$\hat{\alpha}_{\lambda}(\omega) \sim f^2 \epsilon^{-(\alpha/2) - (1-\eta)\nu}, \quad (4.12)$$

and in the high-frequency region  $\epsilon^{2\nu} \ll f \ll \epsilon^{(\alpha/2) + \nu}$ , we obtain

$$\hat{\alpha}_{\lambda}(\omega) \sim f^{(3+\eta)/2} \epsilon^{-\alpha/2}. \quad (4.13)$$

We summarize the results obtained in this section in Table I. Comparing the sizes of various contributions to  $\hat{\alpha}(\omega)$ , we find that  $\hat{\alpha}_T(\omega)$  dominates practically in all the frequency regions considered here, namely,  $0 < f < \epsilon^{(3/2)(\nu-\alpha/2)}$ ,  $p \approx \epsilon^{\nu}$ . The overall behavior of  $\hat{\alpha}(\omega)$  is given in Table II.

## V. SOUND-WAVE DISPERSION

We first consider the contribution of  $\theta_T(\omega)$  to the sound-wave dispersion  $\Delta c_T(\omega)$ . Equations (3.2) and (3.10) immediately yield

$$\frac{\Delta c_T(\omega)}{c} = \frac{k_B T^3}{2\pi^2 \rho c^2 C_V^2} \left( \frac{\partial P}{\partial T} \right)_V^2 \kappa \left( \frac{\partial \kappa}{\partial T} \right)_S^2 \omega^2 \times \int_0^{\infty} dx \frac{x^2}{(1+x^2)^2} \frac{1}{\omega^2 + 4D^2 \kappa^6 K^2(x)}. \quad (5.1)$$

In the low-frequency region  $\omega \ll D\kappa^3$ , we cannot ignore  $\omega^2$  in the denominator since this would cause the integral to diverge at small  $x$  because  $K(x) \approx x^2$  for  $x \ll 1$ . The small- $\omega$  behavior of (5.1) is found

TABLE I. Behaviors of contributions to sound attenuation from  $\theta_T$ ,  $\theta_p$ , and  $\lambda_{\eta T}$  defined in Sec. III in various frequency regions.

| $f$                              | 0                               | $\longleftrightarrow$ | $\epsilon^{3\nu}$                      | $\longleftrightarrow$ | $\epsilon^{2\nu}$ | $\longleftrightarrow$ | $\epsilon^{\nu+\alpha/2}$             |
|----------------------------------|---------------------------------|-----------------------|--|-----------------------|-------------------|-----------------------|---------------------------------------|
| Frequency regions                | I                               |                       | II                                     |                       | III               |                       |                                       |
| $\hat{\alpha}_T(\omega)$         | $f^2 \epsilon^{-3\nu-\alpha/2}$ |                       | $f^{2-2p/3} \epsilon^{-3\alpha/2}$     |                       |                   |                       |                                       |
| $\hat{\alpha}_p(\omega)$         |                                 |                       | $f^2 \epsilon^{-\nu-\alpha}$           |                       |                   |                       |                                       |
| $\hat{\alpha}_{\lambda}(\omega)$ |                                 |                       | $f^2 \epsilon^{-(1-\eta)\nu-\alpha/2}$ |                       |                   |                       | $f^{(3+\eta)/2} \epsilon^{-\alpha/2}$ |

TABLE II. Behaviors of dominant contributions to sound attenuation in various frequency regions.

| $f$                    | 0 | $\longleftrightarrow$                      | $\epsilon^{3\nu}$ | $\longleftrightarrow$   | $\epsilon^{\nu+\alpha/2}$ |
|------------------------|---|--|-------------------|---|---------------------------|
| $\hat{\alpha}(\omega)$ |   | $\hat{\alpha}_T(\omega)$<br>given by (4.1) |                   | $\hat{\alpha}_T(\omega)$<br>$\sim f^{2-2p/3} \epsilon^{-3\alpha/2}$ |                           |

by introducing again the frequency-dependent cut-off at  $x = x_\omega \sim f^{1/2} \epsilon^{-3\nu/2} \ll 1$ . Thus we find<sup>19</sup>

$$f \ll \epsilon^{3\nu}, \quad \Delta c_T(\omega)/c \sim \epsilon^{\alpha - (3/2)\nu - 2} f^{3/2}. \quad (5.2)$$

Or, using the scaling-law relation,

$$\Delta c_T(\omega)/c \sim \epsilon^{-9\nu/2} f^{3/2}. \quad (5.3)$$

For higher frequencies  $f \gg \epsilon^{3\nu}$  the two cases  $2p - 3 > 0$  and  $2p - 3 \leq 0$  must be distinguished. This becomes apparent if we consider the sound-velocity change using (4.6),

$$\frac{\Delta c_T(\omega)}{c} \sim \epsilon^{-\alpha} \omega^2 \int_{\kappa}^{\infty} \frac{dk}{k^{2p-2}} \frac{1}{(2\Gamma_k)^2 + \omega^2}. \quad (5.4)$$

Now, for  $2p - 3 \leq 0$ , the integral  $\int_{\kappa}^{\infty} dk k^{2-2p}$  diverges at infinite  $k$ , and hence major contributions to (5.4) come from  $k \gg \kappa$ . On the other hand, for  $2p - 3 > 0$ , the contributions from  $k \gg \kappa$  to (5.4) are not significant, and we should use (5.1) where the main contributions to the integral come from  $x \leq 1$ . For  $2p - 3 \leq 0$  the integral (5.4) is again estimated by introducing the same frequency-dependent cutoff  $k_f$  as for (4.7). Thus we find for  $2p - 3 \leq 0$ ,

$$\Delta c_T(\omega)/c \sim \epsilon^{-\alpha} f^{1-2p/3}, \quad (\epsilon^{3\nu} \ll f \ll \epsilon^{\nu+\alpha/2}) \quad (5.5)$$

and for  $2p - 3 > 0$ ,

$$\Delta c_T(\omega)/c \sim \epsilon^{\alpha+3\nu-2} f^0 \sim \epsilon^0 f^0, \quad (\epsilon^{3\nu} \ll f \ll \epsilon^{\nu+\alpha/2}) \quad (5.6)$$

where the scaling-law relation has been used in the second line of (5.6). The result (5.6) is identical to our earlier result.<sup>13</sup>

For  $f \sim \epsilon^{3\nu}$ , we can evaluate (5.1) numerically. Here again the same remark made for the attenuation applies for the interpolation of (5.1) and (5.5).

The contribution to the dispersion  $\Delta c_p(\omega)$  from  $\theta_p(\omega)$  is found from the results of Appendix B. There are two kinds of contributions to  $\Delta c_p(\omega)$ .

TABLE III. Behaviors of contributions to sound dispersion from  $\theta_T$ ,  $\theta_p$ , and  $\lambda_{\eta T}$  defined in Sec. III in various frequency regions.  $\Delta c_p'$  arises from fluctuations with  $k \sim \kappa$  and  $\Delta c_p''$  from those with  $k \ll \kappa$ .

| $f$                          | 0 | $\longleftrightarrow$                  | $\epsilon^{3\nu}$                 | $\longleftrightarrow$                     | $\epsilon^{2\nu+\alpha/4}$ | $\longleftrightarrow$ | $\epsilon^{2\nu}$                      | $\longleftrightarrow$ | $\epsilon^{\nu+\alpha/2}$     |
|------------------------------|---|--|-----------------------------------|---|----------------------------|-----------------------|--|-----------------------|-------------------------------|
| $\Delta c_T(\omega)/c$       |   | $f^{3/2} \epsilon^{-9\nu/2}$           |                                   |   |                            |                       |  |                       |                               |
| $\Delta c_p'(\omega)/c$      |   |  |                                   |   |                            |                       |  |                       | $f^2 \epsilon^{-2\nu-\alpha}$ |
| $\Delta c_p''(\omega)/c$     |   | $f^{3/2} \epsilon^{(3/2)(\nu-\alpha)}$ |                                   | $f^{3/2} \epsilon^{-(15\nu/2)-3\alpha/2}$ |                            |                       | $f^{3/2} \epsilon^{-3\nu/2-3\alpha/4}$ |                       |                               |
| $\Delta c_\lambda(\omega)/c$ |   |  | $f^{3/2} \epsilon^{-(2-\eta)\nu}$ |   |                            |                       |  |                       | $f^{(1+\eta)/2} \epsilon^0$   |

One from the fluctuations with  $k \sim \kappa$  denoted by  $\Delta c_p'(\omega)$ , and another from the fluctuations with  $k \ll \kappa$  denoted by  $\Delta c_p''(\omega)$ . Namely,

$$\Delta c_p'(\omega)/c \sim f^2 \epsilon^{-\alpha-2\nu}, \quad \text{for } 0 \leq f \ll \epsilon^{\nu+\alpha/2} \\ \Delta c_p''(\omega)/c \sim f^{3/2} \epsilon^{1-2\alpha}, \quad (5.7)$$

$$\Delta c_p'(\omega)/c \sim f^{9/2} \epsilon^{-(15/2)\nu - (3/2)\alpha}, \quad \text{for } \epsilon^{3\nu} \ll f \ll \epsilon^{2\nu+\alpha/4} \\ \Delta c_p''(\omega)/c \sim f^{3/2} \epsilon^{-(3/2)\nu - (3/4)\alpha}, \quad \text{for } \epsilon^{2\nu+\alpha/4} \ll f \ll \epsilon^{\nu+\alpha/2}. \quad (5.8)$$

Finally, the contribution from  $\lambda_{\eta T}$  denoted by  $\Delta c_\lambda(\omega)$  is found in a similar manner, and the result is,

$$\Delta c_\lambda(\omega)/c \sim f^{3/2} \epsilon^{-(2-\eta)\nu}, \quad \text{for } 0 \leq f \ll \epsilon^{2\nu} \\ \Delta c_\lambda(\omega)/c \sim f^{(1+\eta)/2} \epsilon^0, \quad \text{for } \epsilon^{2\nu} \ll f \ll \epsilon^{\nu+\alpha/2}. \quad (5.9)$$

We summarize the results obtained in this section in Table III. Comparison of magnitudes of various  $\Delta c(\omega)$  shows that  $\Delta c_T(\omega)$  always dominates in the frequency range considered, that is,  $0 \leq f \ll \epsilon^{\nu+\alpha/2}$ . This is shown in Table IV.

## VI. CONCLUDING REMARKS

The sound attenuation and dispersion near the liquid-gas critical point are studied for the sound-wave frequency much less than  $c\kappa$ , and the results are summarized in Tables II and IV. In particular, we considered in detail the frequency dependence of the attenuation and the sound-velocity change arising from the very important process of a sound wave breaking up into two heat modes.<sup>5</sup> These theoretical results can be tested with the recent accurate experiments,<sup>1,20</sup> which will be deferred to another occasion.

So far our study has been restricted to the cases where the sound-wave frequencies are much less than  $c\kappa$ . We now briefly discuss what is expected

TABLE IV. Behavior of the dominant contribution to sound dispersion.

| $f$                  | 0                      | $\longleftrightarrow$ | $\epsilon^{3\nu}$              | $\longleftrightarrow$ | $\epsilon^{\nu+\alpha/2}$ |
|----------------------|------------------------|-----------------------|--------------------------------|-----------------------|---------------------------|
| $\Delta c(\omega)/c$ | $\Delta c_T(\omega)/c$ |                       | $f^{1-2p/3}\epsilon^{-\alpha}$ |                       | for $2p-3 \leq 0$         |
|                      | given by (5.1)         |                       | $f^0 \epsilon^0$               |                       | for $2p-3 > 0$            |

when the sound-wave frequency becomes comparable to  $c\kappa$ . Here  $L_{\vec{q},\vec{k}}$  and  $H_{\vec{q},\vec{k}}$  are no longer expected to have the simple forms of (3.8) and (3.13), respectively. Nevertheless, the static scaling ideas<sup>5</sup> allow us to estimate the various contributions to the attenuation and dispersion. Namely, for  $\omega \sim c\kappa$ , we have, apart from some dimensionless factors which are functions of  $q/\kappa$ ,

$$\hat{\alpha}_T \sim \epsilon^{2(\nu+\alpha/2)(1-p/3)-3\alpha/2} \sim \epsilon^\nu, \quad \text{if } \alpha=0 \text{ and } p=1/\nu \quad (6.1)$$

$$\hat{\alpha}_p \sim \epsilon^\nu, \quad (6.2)$$

$$\Delta c_T/c \sim \epsilon^{(\nu+\alpha/2)(1-2p/3)-\alpha}, \quad \text{if } 2p-3 \leq 0 \quad (6.3)$$

$$\Delta c_T/c \sim \epsilon^0, \quad \text{if } 2p-3 > 0$$

$$\Delta c_p/c \sim \epsilon^0, \quad (6.4)$$

$\hat{\alpha}_\lambda$  and  $\Delta c_\lambda/c$  are much smaller and are ignored.

The contributions (6.1)–(6.4) are roughly of the same order of magnitudes and satisfy the dynamical scaling in the extended sense<sup>5,13,21</sup> when  $\alpha=0$ . That is, the sound-wave damping and the frequency shift become comparable to and scale as the sound-wave frequency itself. This is an interesting future problem to study both experimentally and theoretically. The proper theoretical treatment, however, requires the consideration of the frequency dependences of  $\gamma_{\vec{k}}$  and  $\gamma_{\vec{q}-\vec{k}}$  in (3.11) in a self-consistent manner.

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#### APPENDIX A

Here we derive the kinetic equation for the entropy density  $\vec{S}(\vec{r})$ , restricting ourselves to the terms quadratic in the deviations from equilibrium.<sup>21a</sup> Regarding  $\vec{S}(\vec{r})$  as a function of the particle number density  $\vec{n}(\vec{r})$  and the internal energy density  $\vec{H}(\vec{r})$ , we have for the entropy density fluctuation  $\delta\vec{S}(\vec{r})$ ,

$$\delta\vec{S}(\vec{r}) = \delta\vec{S}'(\vec{r}) + \frac{1}{2} s_{NE} \delta\vec{n}(\vec{r})^2$$

$$+ s_{NE} \delta\vec{n}(\vec{r})\delta\vec{H}(\vec{r}) + \frac{1}{2} s_{EE} \delta\vec{H}(\vec{r})^2 + \dots, \quad (A1)$$

$$\text{where } \delta\vec{S}'(\vec{r}) = (1/T) \delta\vec{H}(\vec{r}) - (h/T) \delta\vec{n}(\vec{r}), \quad (A2)$$

and  $h$  is the equilibrium value of enthalpy per atom, and the  $s$ 's are appropriate numerical coefficients.

If we take the local pressure, the entropy density and the three components of the local velocity as the five macroscopic variables, the local pressure and the longitudinal velocity change more rapidly in time with the frequency of sound wave than the entropy and the transverse components of the local-velocity change. Thus, for the dynamics of entropy fluctuations, we can retain only the entropy density and the transverse components of the local velocity as the macroscopic variables  $\vec{a}$ 's entering the kinetic equation of Ref. 7. However, as in the case of binary mixtures, we temporarily include the longitudinal component of the local velocity and drop it later on.

Let us first consider  $\delta\vec{S}'(\vec{r})$  or its Fourier transform  $\delta\vec{S}'_{\vec{q}}$ . The kinetic equation aside from the random force and the damping term is written as<sup>7</sup>

$$\delta\dot{\vec{S}}'_{\vec{q}} = \frac{k_B T}{2} \sum_{\vec{k}} \sum_{\alpha\beta} \frac{\langle \{ \vec{S}'_{\vec{q}}, [\vec{a}_{\vec{k}}^{\alpha*}, \vec{a}_{\vec{q}-\vec{k}}^{\beta*}] \} \rangle}{\langle |[\vec{a}_{\vec{k}}^{\alpha}, \vec{a}_{\vec{q}-\vec{k}}^{\beta}]|^2 \rangle} [\vec{a}_{\vec{k}}^{\alpha}, \vec{a}_{\vec{q}-\vec{k}}^{\beta}], \quad (A3)$$

where the curly bracket denotes the Poisson bracket and

$$[\vec{a}_{\vec{k}}^{\alpha}, \vec{a}_{\vec{q}-\vec{k}}^{\beta}] \equiv \vec{a}_{\vec{k}}^{\alpha} \vec{a}_{\vec{q}-\vec{k}}^{\beta} - \sum_{\gamma} \frac{\langle \vec{a}_{\vec{k}}^{\alpha} \vec{a}_{\vec{q}-\vec{k}}^{\beta} \vec{a}_{\vec{q}}^{\gamma*} \rangle}{\langle |\vec{a}_{\vec{q}}^{\gamma}|^2 \rangle} \vec{a}_{\vec{q}}^{\gamma}, \quad (A4)$$

where we subtracted the orthogonalizing term from  $\vec{a}_{\vec{k}}^{\alpha} \vec{a}_{\vec{q}-\vec{k}}^{\beta}$  to ensure the proper orthogonalization of (A4) to  $\vec{a}_{\vec{q}}^{\gamma}$ . By the time-reversal argument, the only allowed combination on the right-hand side of (A3) is  $[\vec{S}'_{\vec{q}}, \vec{v}_{\vec{q}-\vec{k}}^{\sigma}]$ , ( $\sigma=x, y, z$ ). The coefficients in (A3) and (A4) are obtained by first noting the following relations:

$$\langle |\vec{v}_{\vec{k}}^{\sigma}|^2 \rangle = k_B T / \rho, \quad (A5)$$

$$\langle |\delta\vec{S}'_{\vec{k}}|^2 \rangle = k_B C_{\vec{k}}^E, \quad (A6)$$

$$\langle \delta\vec{S}'_{\vec{k}} \vec{v}_{\vec{q}-\vec{k}}^{\sigma} \vec{v}_{\vec{q}}^{\sigma} \rangle = [(k_B T)^2 / \rho n V^{1/2}] \alpha_{\vec{k}}^E \delta_{\sigma\tau}, \quad (A7)$$

where  $C_{\vec{k}}^E$  is the  $\vec{k}$ -dependent specific heat per unit volume at constant pressure,  $\alpha_{\vec{k}}^E$  is the  $\vec{k}$ -dependent thermal expansion coefficient at constant pressure given by  $\alpha_{\vec{k}}^E = \langle \delta\vec{S}'_{\vec{k}} \delta\vec{n}_{-\vec{k}} \rangle / k_B T$ , and  $\rho$  is the mass density. This gives

$$[\vec{S}'_{\vec{k}}, \vec{v}_{\vec{q}-\vec{k}}^{\sigma}] = \delta\vec{S}'_{\vec{k}} \vec{v}_{\vec{q}-\vec{k}}^{\sigma} - (T \alpha_{\vec{k}}^E / n V^{1/2} C_{\vec{k}}^E) \vec{v}_{\vec{q}}^{\sigma}. \quad (A8)$$

Use of the relations<sup>13</sup>

$$\{\delta\vec{S}'_{\vec{k}}, \delta\vec{S}'_{\vec{k}'}\} = iV^{-1/2} T^{-2} (\vec{k}' - \vec{k}) \cdot \vec{J}_{\vec{k}+\vec{k}'}, \quad (A9)$$

$$\{\vec{v}_{\vec{k}}^{\sigma}, \delta\vec{S}'_{\vec{k}'}\} = \frac{ik'^{\sigma}}{\rho V^{1/2}} \vec{S}_{\vec{k}+\vec{k}'} - \frac{i}{\rho T V^{1/2}} \sum_{\tau} k^{\tau} \vec{P}_{\vec{k}+\vec{k}'}^{\sigma\tau}, \quad (A10)$$

where  $\vec{P}_{\mathbf{k}}^{\sigma}$  is the  $\tau\sigma$ -component of stress tensor and  $\vec{J}_{\mathbf{k}}^{\tau}$  is the heat current related to the energy current  $\vec{J}_{\mathbf{k}}^{\tau}$  by

$$\vec{J}_{\mathbf{k}}^{\tau} = \vec{J}_{\mathbf{k}}^{\tau} - h\vec{v}_{\mathbf{k}}^{\tau}, \quad (\text{A11})$$

yields

$$\begin{aligned} & \langle \{ \delta\vec{S}_{\mathbf{q}}^{\sigma}, [\vec{S}'_{\mathbf{k}}, \vec{v}_{\mathbf{k}-\mathbf{q}}^{\sigma}] \} \rangle \\ &= -i \sum_{\tau} V^{-1/2} T^{-2} (q^{\tau} + k^{\tau}) \langle \vec{J}_{\mathbf{q}-\mathbf{k}}^{\tau} \vec{v}_{\mathbf{k}-\mathbf{q}}^{\sigma} \rangle \\ & - \frac{i}{\rho V^{1/2}} \langle \delta\vec{S}'_{\mathbf{k}} [q^{\sigma} \delta\vec{S}'_{\mathbf{k}} + T^{-1} \sum_{\tau} (k^{\tau} - q^{\tau}) P_{\mathbf{k}}^{\tau\sigma}] \rangle, \end{aligned} \quad (\text{A12})$$

where we have used that  $\langle \{ \delta\vec{S}_{\mathbf{q}}^{\sigma}, \vec{v}_{\mathbf{q}}^{\sigma} \} \rangle = 0$ . Near the critical point the major contribution comes from  $\langle |\delta\vec{S}_{\mathbf{k}}^{\sigma}|^2 \rangle$ , and thus,

$$\langle \{ \delta\vec{S}_{\mathbf{q}}^{\sigma}, [\vec{S}'_{\mathbf{k}}, \vec{v}_{\mathbf{k}-\mathbf{q}}^{\sigma}] \} \rangle = -(ik_B/\rho V^{1/2}) q^{\sigma} C_{\mathbf{k}}^{\sigma}. \quad (\text{A13})$$

Noting that  $\langle |\delta\vec{S}'_{\mathbf{k}}, \vec{v}_{\mathbf{k}-\mathbf{q}}^{\sigma}|^2 \rangle = k_B^2 T C_{\mathbf{k}}^{\sigma}/\rho + O(N^{-1/2})$ , we then find from (A3) and (A13)

$$\delta\dot{S}'_{\mathbf{q}} = -iV^{-1/2} \sum_{\sigma} q^{\sigma} \sum_{\mathbf{k}}' [S_{\mathbf{k}}^{\sigma}, v_{\mathbf{k}-\mathbf{q}}^{\sigma}]. \quad (\text{A14})$$

Next, the contributions to  $\delta\dot{S}$  from the second, third, ... terms in (A1) either involve  $\delta\dot{n}$  or  $\delta H$ .  $\delta\dot{n}$  is the longitudinal component of local velocity and averages out, and  $\delta H = T\delta S' + h\delta\dot{n}$  where  $\delta S'$  starts from the quantities quadratic in  $\delta S'$  and  $\vec{v}$ . Therefore,  $\delta\dot{S} = \delta\dot{S}' +$  (the quantities that average out, or that are cubic, quartic, ... in  $\delta S'$  and  $\delta v$ ), and for our purpose we can take  $\delta\dot{S} = \delta\dot{S}'$ . Thus, finally we obtain

$$\delta\dot{S}_{\mathbf{q}} = -V^{-1/2} \sum_{\sigma} iq^{\sigma} \sum_{\mathbf{k}}' [\delta S_{\mathbf{k}}^{\sigma}, v_{\mathbf{k}-\mathbf{q}}^{\sigma}], \quad (\text{A15})$$

where  $v_{\mathbf{k}-\mathbf{q}}^{\sigma}$  is supposed to contain only the transverse component.

Next, consider the kinetic equation for  $v_{\mathbf{q}}^{\sigma}$ , which is written as

$$\dot{v}_{\mathbf{q}}^{\sigma} = \frac{k_B T}{2} \sum_{\mathbf{k}}' \frac{\langle \{ \vec{v}_{\mathbf{q}}^{\sigma}, [\vec{S}'_{\mathbf{k}}, \vec{S}'_{\mathbf{k}-\mathbf{q}}] \} \rangle}{\langle |\vec{S}'_{\mathbf{k}}, \vec{S}'_{\mathbf{k}-\mathbf{q}}|^2 \rangle} [S_{\mathbf{k}}^{\sigma}, S'_{\mathbf{k}-\mathbf{q}}]. \quad (\text{A16})$$

Retaining only the most important terms near the critical point in the coefficient of (A16) as before, we find

$$\dot{v}_{\mathbf{q}}^{\sigma} = -\frac{T}{2\rho V^{1/2}} \sum_{\mathbf{k}}' i \left( \frac{k^{\sigma}}{C_{\mathbf{k}}^{\sigma}} + \frac{q^{\sigma} - k^{\sigma}}{C_{\mathbf{k}-\mathbf{q}}^{\sigma}} \right) [S_{\mathbf{k}}^{\sigma}, S'_{\mathbf{k}-\mathbf{q}}], \quad (\text{A17})$$

$$[S_{\mathbf{k}}^{\sigma}, S'_{\mathbf{k}-\mathbf{q}}] = \delta S_{\mathbf{k}}^{\sigma} \delta S'_{\mathbf{k}-\mathbf{q}} - \frac{\langle \delta\vec{S}_{\mathbf{k}}^{\sigma} \delta\vec{S}'_{\mathbf{k}-\mathbf{q}} \delta\vec{S}'_{\mathbf{q}} \rangle}{\langle |\delta S_{\mathbf{q}}^{\sigma}|^2 \rangle} \delta S_{\mathbf{q}}^{\sigma}. \quad (\text{A18})$$

Furthermore, if we note that only the transverse components of  $\dot{v}_{\mathbf{q}}^{\sigma}$  are to be retained, (A17) reduces to

$$\begin{aligned} \dot{v}_{\mathbf{q}}^{\sigma} &= -(T/2\rho V^{1/2}) \sum_{\mathbf{k}}' i(k^{\sigma} - (\vec{q} \cdot \vec{k}/q^2) q^{\sigma}) \\ & \times \left( \frac{1}{C_{\mathbf{k}}^{\sigma}} - \frac{1}{C_{\mathbf{k}-\mathbf{q}}^{\sigma}} \right) [S_{\mathbf{k}}^{\sigma}, S'_{\mathbf{k}-\mathbf{q}}]. \end{aligned} \quad (\text{A19})$$

## APPENDIX B

Here we analyze  $\theta_p(\omega)$  given by (3.11) and (3.13) in detail. Since  $\chi_S$  behaves as  $C_V$  as far as the critical anomaly is concerned, we have, assuming the strong scaling for  $\chi_S(\vec{k})$ ,<sup>17,22</sup>

$$[\chi_S(\vec{k})]^{-1} = k^{\alpha/\nu} F(\kappa/k). \quad (\text{B1})$$

$$\begin{aligned} \text{Hence } & \left( \frac{\partial[\chi_S(\vec{k})]^{-1}}{\partial T} \right)_S \\ &= \kappa^{-1+\alpha/\nu} \left( \frac{\partial \kappa}{\partial T} \right)_S (\kappa/k)^{1-\alpha/\nu} F'(\kappa/k) \\ & \sim \epsilon^{-1+\alpha} F_1(\kappa/k), \end{aligned} \quad (\text{B2})$$

where  $F_1(x) = x^{1-\alpha/\nu} F'(x)$  apart from a finite numerical coefficient.

Therefore,

$$\begin{aligned} \theta_p(\omega) & \sim \epsilon^{2(\alpha-1)} \sum_{s,s'} \int d\vec{k} \\ & \times \frac{|F_1(\kappa/k)|^2}{ic[sk+s'|\vec{q}-\vec{k}|] + \gamma_{\mathbf{k}} + \gamma_{\mathbf{q}-\mathbf{k}} - i\omega}. \end{aligned} \quad (\text{B3})$$

If  $F_1(x) \sim x^g$  for  $x \ll 1$  with  $g > 1$ , the integral converges at large  $k$  provided that  $\gamma_k$  increases at least as fast as  $k$  at large  $k$ .<sup>23</sup> We thus assume the convergence of the integral at large  $k$ . Now, if  $\gamma_k = 0$ , there is no significant contribution to  $\text{Re}\theta_p(\omega)$  from the terms with  $s = s'$ . The terms with  $s - s'$  behave like the contribution from the three-phonon processes at low temperature solids or superfluid helium<sup>24</sup> and  $\text{Re}\theta_p(\omega) \sim 1/q$ . Thus here the lifetime effects<sup>24</sup> are important contrary to the calculation of KS. Assuming the integral for  $\text{Re}\theta(\omega)$  to converge at small  $k$  as well<sup>23</sup> and noting that for  $k \sim \kappa$ ,  $\gamma_k \sim c\kappa$  [see the discussion of Sec. VI], we find for  $\omega \ll c\kappa$  that

$$\text{Re}\theta_p(\omega) \sim \epsilon^{2\nu-2+3\alpha/2} f^0 = \epsilon^{-\nu+\alpha/2} f^0$$

using  $2 = 3\nu + \alpha$ , recovering KS's result.

Turning now to  $\text{Im}\theta_p(\omega)$ , we have

$$\begin{aligned} \text{Im}\theta_p(\omega) & \sim \epsilon^{2(\alpha-1)} \sum_{ss'} \int d\vec{k} |F_1(\kappa/k)|^2 \\ & \times \frac{\omega}{c^2[sk+s'|\vec{q}-\vec{k}|-q]^2 + (\gamma_{\mathbf{k}} + \gamma_{\mathbf{q}-\mathbf{k}})^2}, \end{aligned} \quad (\text{B4})$$

since  $\text{Im}\theta_p(\omega)$  vanishes for  $\omega = 0$ . For definiteness let us suppose that  $F_1(x) \sim x^0$  for  $x \gg 1$ .<sup>23</sup> The terms with  $s = s'$  in (B4) give no problem and behave as  $\epsilon^{-2+\nu+\alpha} f = \epsilon^{-2\nu} f$ . For  $s = -s'$ , (B4) behaves something like the following quantity<sup>25</sup>

$$I(\epsilon, f) = \epsilon^{2(\alpha-1)} f \int d\vec{k} (f^2 + \gamma_k^2)^{-1}. \quad (\text{B5})$$

Since for very small  $k$ ,  $\gamma_k \propto k^2$ , the integral diverges at small  $k$  if we set  $f = 0$  in the denominator.

This is the cause of some complications here. The integral can be estimated by introducing the frequency-dependent cutoff at  $k_f$  given by  $f = \gamma k_f$  as

$$I(\epsilon, f) \sim \epsilon^{2(\alpha-1)} f^{-1} k_f^3, \quad (\text{B6})$$

where the correct most dominant  $\gamma_{k_f}$  of Table II must be used in each frequency regions  $ck_f$ . The

results of somewhat tedious analysis are

$$\begin{aligned} 0 \leq f \ll \epsilon^{3\nu}, & \quad I(\epsilon, f) \sim \epsilon^{1-\alpha} f^{1/2}, \\ \epsilon^{3\nu} \ll f \ll \epsilon^{2\nu+\alpha/4}, & \quad I(\epsilon, f) \sim \epsilon^{-15\nu/2-\alpha/4} f^{7/2}, \\ \epsilon^{2\nu+\alpha/4} \ll f \ll \epsilon^{\nu+\alpha/2}, & \quad I(\epsilon, f) \sim \epsilon^{-3\nu/2+\alpha/4} f^{1/2}. \end{aligned} \quad (\text{B7})$$

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<sup>1</sup>C. W. Garland, in *Physical Acoustics* (Academic, New York, to be published), Vol. VII.

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<sup>6</sup>K. Kawasaki, *Phys. Rev.* **150**, 291 (1966).

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<sup>9</sup>We have omitted the orthogonalizing terms from the bilinear terms in (2.1) and (2.2). That is, we set  $[a_{\vec{k}}^\alpha, a_{\vec{q}-\vec{k}}^\beta]$  of (A4) equal to  $\delta a_{\vec{k}}^\alpha \delta a_{\vec{q}-\vec{k}}^\beta$ . The effects of the orthogonalizing terms in the final results are  $O(V^{-1/2})$ , and thus vanish in the thermodynamic limit.

<sup>10</sup>J. Swift, *Phys. Rev.* **173**, 257 (1968).

<sup>11</sup>For a binary solution this decay rate has been compared with the recent light-scattering experiment with satisfactory agreement. See K. Kawasaki, *Phys. Letters* **30A**, 325 (1969).

<sup>12</sup>G. B. Benedek, in *Statistical Physics, Phase Transition and Superfluidity*, edited by M. Chrétien, E. P. Gross and S. Deser (Gordon and Breach, New York, 1968), Vol. 2.

<sup>13</sup>K. Kawasaki, *Progr. Theoret. Phys. (Kyoto)* **40**, 930 (1968).

<sup>14</sup>K. Kawasaki and M. Tanaka, *Proc. Phys. Soc. (London)* **90**, 791 (1967).

<sup>15</sup>Use of more plausible form  $C_k^p \propto (k^2 + \kappa^2)^{-1+\eta/2}$  only introduces a factor  $(1 - \frac{1}{2}\eta)^2$  which is close to 1 due to the smallness of  $\eta$ . For  $k \gg \kappa$ , however, a quite different form of  $C_k^p$  (4.4) below, is more appropriate.

<sup>16</sup>Here and after we use the conventional notation for the critical exponents. See, e.g., M. E. Fisher, *Rept. Progr. Phys.* **30**, 615 (1967).

<sup>17</sup>M. Ferer, M. Moore, and M. Wortis, *Phys. Rev. Letters* **22**, 940 (1969); **22**, 1382 (1969).

<sup>18</sup>This behavior of the high-frequency sound attenuation is true also for magnetic systems and near the  $\lambda$  transition in helium [K. Kawasaki, *J. Appl. Phys.* (to be published); *Phys. Letters* (to be published)]. In fact one can demonstrate that this is true under a rather general condition.

<sup>19</sup>The  $f^{3/2}$  dependence which differs from the  $f^2$  dependence inferred from the single-relaxation-time model is due to the large contribution of the long wavelength fluctuations. The situation is somewhat similar to that encountered in the nonlinear shear viscosity. See W. Botch and M. Fixman, *J. Chem. Phys.* **36**, 3100 (1962); W. Botch and M. Fixman (report of work prior to publication); T. Yamada and K. Kawasaki, *Progr. Theoret. Phys. (Kyoto)* **38**, 1031 (1967).

<sup>20</sup>M. Barmatz (private communication).

<sup>21</sup>B. I. Halperin and P. C. Hohenberg, *Phys. Rev.* **177**, 952 (1969).

<sup>21a</sup>In this Appendix symbols  $\vec{S}$ ,  $\vec{n}$ ,  $\vec{H}$ , etc., stand for the molecular expressions for the variables  $S$ ,  $n$ ,  $H$ , etc.

<sup>22</sup>This may not be always true. See Ref. 17 for the case of spin correlations.

<sup>23</sup>If

$$\chi_S(\vec{k}) \sim (\kappa^2 + k^2)^{-\alpha/2\nu}, \quad F_1(x) \sim x^{2-\alpha/\nu} (1+x^2)^{\alpha/2\nu-1},$$

and  $g = 2 - \alpha/\nu$ . The behavior of  $\gamma_k$  at large  $k$  can be found in Table II. See also Sec. VI.

<sup>24</sup>K. Kawasaki, *Progr. Theoret. Phys. (Kyoto)* **26**, 795 (1961).

<sup>25</sup>The corresponding integral for  $\text{Re}\theta_p(\omega)$  behaves like

$$\epsilon^{2(\alpha-1)} \int d\vec{k} \gamma_k / (f^2 + \gamma_k^2).$$