## Zero-frequency critical bulk viscosity: Is the amplitude ratio truly universal?

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It was shown by Onuki that the zero-frequency bulk viscosity is associated with a universal amplitude ratio that was calculated to be around 0.10. We show that the sound attenuation data can be used to extract a value for this universal number and we find this number to be around 0.18, reasonably close to Onuki's estimate. However, we argue that a reconsideration of this amplitude ratio shows that this ratio is not truly universal. It has a logarithmic correction instead.

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It was shown by Onuki [1,2] that the zero-frequency bulk viscosity  $\eta_B$  diverges near the liquid gas critical point. Further, it was established that if the critical point fluctuations decay with a relaxation rate  $\Gamma = \Gamma_0 \xi^{-(3+x_\eta)}$ , where  $\xi$  is the correlation length,  $x_\eta$  is the small viscosity exponent, and  $C_s$  is the zero-frequency sound velocity, then there is a universal combination given by

$$\frac{\eta_B}{\rho} \frac{\Gamma_0}{C_s^2 \xi^{(3+x_\eta)}} = R_B.$$
<sup>(1)</sup>

From an  $\epsilon$  expansion Onuki found that  $R_B$  is to be 0.087 and from a three-dimensional calculation he found it to be 0.13. The points that we want to make in this note are as follows.

(i) The ultrasound attenuation data of Roe and Meyer [3] yield a value of  $R_B$  (with  $x_{\eta}=0$ ) which is fairly close to the value predicted by Onuki [12].

(ii) The bulk viscosity is related to the frequencydependent specific heat and using an exponentiation due to Nicoll [4] for the scaling function of the specific heat, we show that  $R_B$  is not truly universal. There is a logarithmic correction and we show that the data of Roe and Meyer offer some support for this logarithmic factor. From the data it is impossible to say at present whether it favors the universality of  $R_B$  or the logarithmic scale breaking that we believe exists. Accurate measurements of the zero-frequency bulk viscosity would consequently be very helpful to resolve the issue.

Since bulk viscosity is a slightly nonstandard concept, we will explain our point of view in a somewhat detailed manner. We claim that bulk viscosity will be generated from the pressure gradient term when the relaxation between pressure and density fluctuations is correctly handled [5]. It is the delayed response of the density fluctuation [6] to an imposed pressure fluctuation that causes the damping of sound (apart from the purely Stokes damping). This means that apart from the Stokes damping, all other damping should emerge from the two linear equations

$$\frac{\partial}{\partial t}\delta\rho + \rho_0(\vec{\nabla}\cdot\vec{v}) = 0 \tag{2}$$

and

$$\frac{\partial \vec{v}}{\partial t} = -\frac{\vec{\nabla}\,\delta P}{\rho_0},\tag{3}$$

where  $\delta \rho$  and  $\delta P$  are fluctuations in density and pressure about a constant background. In terms of the density and entropy fluctuations

$$\delta P = \left(\frac{\partial P}{\partial \rho}\right)_{S} \delta \rho + \left(\frac{\partial P}{\partial S}\right)_{\rho} \delta S = \kappa_{S} \delta \rho + \left(\frac{\partial P}{\partial S}\right)_{\rho} \delta S, \quad (4)$$

where  $\kappa_S = (\partial P / \partial \rho)_S$  is the isentropic bulk modulus.

A time derivative of Eq. (3) yields

$$\frac{\partial^2 \vec{v}}{\partial t^2} = \vec{\nabla} (\kappa_S(\vec{\nabla} \cdot \vec{v})) - \vec{\nabla} \left(\frac{\partial P}{\partial S}\right)_0 \frac{1}{\rho_0} \delta^2 S.$$
(5)

The second term on the right-hand side of the above equation is a damping term and can be shown to give rise to the Kirchoff damping [7], which we will ignore here. The first term on the right-hand side is the sound speed if  $\kappa_s$  is a constant. This is generally true except when the system is very close to its critical point. Near the critical point the relaxation of the density fluctuations is very slow [8,9] and the frequency of the sound can resonate [10] with the decay rate of the density fluctuations. This gives rise to a frequency dependence in  $\kappa_s$  that now has real and imaginary parts and can be written as

$$\kappa_S = \kappa_S^r + i\,\omega\,\kappa_S^{im}\,.\tag{6}$$

With the above discussion in mind, we can now write Eqs. (2) and (3) as

$$\frac{\partial^2 \rho}{\partial t^2} = -\rho_0 \vec{\nabla} \cdot \dot{\vec{v}} = \vec{\nabla} \cdot (\vec{\nabla} \,\delta P) = \kappa_S \nabla^2 \,\delta \rho. \tag{7}$$

The term in the right-hand side of Eq. (7) leads to damping because of the  $i\omega$  factor in Eq. (6) and defines the bulk viscosity as

$$\eta_B(\omega) = \rho \kappa_S^{im}(\omega). \tag{8}$$



FIG. 1. The plot of attenuation per wavelength  $\alpha_{\lambda}$  vs reduced frequency  $\Omega = \omega/2\Gamma_0 \kappa^3$  for  $\Omega < 1$ . We have ignored the small exponent  $x_\eta$  in the relaxation rate. The slope is seen to be about 0.18.

The dispersion relation that we get from Eq. (7) is

$$\omega^2 = C_S^2 k^2 + i\omega \kappa_S^{im} k^2, \qquad (9)$$

where  $C_s^2 = \kappa_s^r(\omega)$ . Writing  $\omega = C_s(k_1 + ik_2)$ , equating real and imaginary parts in Eq. (9), and remembering  $k_2 \ll k_1$ , we find,  $\omega^2 \simeq C_s^2 k_1^2 + i\omega \kappa_s^{im} = C_s^2 (k_1^2 - k_2^2 + 2ik_1k_2) \simeq C_s^2 k_1^2$  $+ 2ik_1k_2C_s^2$ , which leads to  $k_2/k_1 = \omega \kappa_s^{im}/2C_s^2$ . The attenuation per unit wavelength is  $k_2$  and hence the attenuation per wavelength is

$$\alpha_{\lambda} = \frac{k_2}{k_1} = \frac{\omega \kappa_s^{\iota m}}{2C_s^2}.$$
 (10)

This attenuation comes entirely from critical fluctuations.

For very low frequency, i.e.,  $\omega \ll \Gamma_0 \kappa^z$ , we expect the attenuation to be linear in the reduced frequency  $\omega/\Gamma_0 \kappa^z$  and hence

$$\alpha_{\lambda} = \alpha_0 \frac{\omega}{2\Gamma_0 \kappa^z},\tag{11}$$

where  $\alpha_0$  is a constant,  $\kappa = \xi^{-1}$ , and  $z = 3 + x_\eta$  is the dynamic scaling exponent. As a first approximation (correct to lowest order in  $\epsilon = 4 - D$ , where D is the dimensionality of space), we drop  $x_\eta$  and plot the low-frequency data of Roe and Meyer in the form  $\alpha_\lambda$  vs  $\Omega = \omega/2\Gamma_0\kappa^z$ . The result is shown in Fig. 1. The plot is quite linear and bears out our expectation. From equations Eqs. (11), (10), (8), and (1),

$$R_B = \alpha_0 \,. \tag{12}$$

The slope of Fig. 1 yields  $\alpha_0 = 0.18$ . The lowest-order  $\epsilon$  expansion of Onuki yields 0.087 for  $R_B$ .

We now note that near the critical point  $T_c$ , the sound speed can be written as

$$C_{S}^{2} = T \frac{\left(\frac{\partial P}{\partial T}\right)^{2}}{\rho^{2} C_{V}},$$
(13)

where  $C_V$  is the constant volume specific heat. This is the only critical quantity in the above expression. Both  $\rho$  (one phase region) and  $(\partial P/\partial T)_V$  are noncritical. The response function which is the specific heat exhibits a lagging effect near the critical point and hence  $C_V$  is frequency dependent. Consequently, we can write

$$C_V = C_V^r + i C_V^{im} \tag{14}$$

and the use of Eqs. (13) and (14) leads to

$$C_{S}^{2} = T \frac{\left(\frac{\partial P}{\partial T}\right)_{V}^{2}}{\rho^{2}} \frac{C_{V}^{r} - iC_{V}^{im}}{(C_{V}^{r})^{2} + (C_{V}^{im})^{2}}$$

since  $C_V^{im} \ll C_V^r$ ,

$$C_{S}^{2} \approx T \frac{\left(\frac{\partial P}{\partial T}\right)_{V}^{2}}{\rho^{2}} \frac{C_{V}^{r} - iC_{V}^{im}}{(C_{V}^{r})^{2}}.$$

Now  $C_s^2 = \omega^2 / k^2 \simeq (\omega^2 / k_1^2 (1 - 2ik_2 / k_1)]$  and comparing with the previous form, we get

$$\alpha_{\lambda} = \frac{k_2}{k_1} = \frac{1}{2} \frac{C_V^{im}}{C_V^r}.$$
 (15)

We now need to discuss the scaling behavior of  $C_V$ . At zero frequency the specific heat diverges near the critical point as  $\xi^{\alpha/\nu}$ , where  $\alpha$  is a small exponent (for the gas liquid critical point  $\alpha \approx 0.11$ ). Thus,

$$C_V(\omega=0,\kappa) = C_0 \frac{\kappa^{-\alpha/\nu} - 1}{\frac{\alpha}{\nu}}.$$
 (16)

At finite frequency, the response is limited by the finite frequency as one approaches the critical point. As we lower the value of  $\kappa$ , at a finite  $\omega$ , there comes a temperature at which  $\Gamma_0 \kappa^z \sim \omega$  and if  $\kappa$  (temperature) is further lowered, the response cannot change any more and we have the dynamic scaling result [11]

$$C_V(\omega,\kappa=0) = C_0 \frac{(-i\omega)^{-\alpha/z\nu} - 1}{\frac{\alpha}{\nu}}.$$
 (17)

The passage from Eq. (16) to Eq. (17) is determined by the scaling function  $F(\omega/\Gamma_0 \kappa^z)$ , such that  $C_V(\kappa, \omega)$  can be written as

$$C_{V}(\kappa,\omega) = C_{0} \frac{\kappa^{-\alpha/\nu} F(\Omega) - 1}{\frac{\alpha}{\nu}}, \qquad (18)$$

where  $\Omega = \omega/2\Gamma_0 \kappa^z$  and  $\Omega \ge 1$ ,  $F(\Omega) \sim (-i\Omega)^{-\alpha/z\nu}$  to reproduce Eq. (17). These results are general. Similar arguments give rise to the frequency dependence of shear viscosity [12–16], which has been experimentally verified.

The simplest form of  $F(\Omega)$  which is inspired by an exponentiation scheme due to Nicoll [4] for the wave-numberdependent specific heat is

$$C_{V}(\kappa,\omega) = C_{0} \frac{\kappa^{-\alpha/\nu} [1-i\beta\Omega]^{-\alpha/\nu} - 1}{\frac{\alpha}{\nu}}, \qquad (19)$$

where  $\beta$  is a number of order unity, which can be fixed from the low-frequency (linear) behavior in  $\Omega$  or the highfrequency behavior. In this case, we imagine  $\beta$  determined by the low-frequency expansion of the diagrammatic perturbation series for  $C_V(\kappa, \omega)$ . Expanding Eq. (19) for  $\Omega \ll 1$ , we find

$$\alpha_{\lambda} = \beta \frac{\alpha}{\nu} \frac{\omega}{2\Gamma_{0}\kappa^{z}} \frac{\frac{\alpha}{\nu}}{\kappa - \frac{\alpha}{\nu} - 1} \approx \beta \frac{\alpha}{\nu} \frac{\omega}{2\Gamma_{0}\kappa^{z}} \frac{1}{\ln \frac{1}{\kappa}} \quad \text{for } \frac{\alpha}{\nu} \ll 1.$$
(20)

From a one-loop calculation  $\beta(\alpha/\nu) = 4/3\pi$ . Comparing with Eqs. (1), (11), and (12), we find

$$R_B = \frac{4}{3\pi} \frac{1}{\ln \frac{\kappa_0}{\kappa}}.$$
 (21)

This is the principal result that we wanted to show.  $R_B$  is not a strict constant, but has a weak dependence on  $\kappa$  through the logarithmic factor.

Does the data of Roe and Meyer support this factor? In Fig. 2 we explore this issue. What is shown in Fig. 2(a) is the plot of  $\alpha_{\lambda}$  vs  $\omega/\Gamma_0 \kappa^z$ , with  $z=3+x_{\eta}$  (Fig. 1 had  $x_{\eta}=0$ ). In



FIG. 2. Attenuation per wavelength  $\alpha_{\lambda}$  vs the reduced frequency in the "hydrodynamic" region. In (a) we show  $\alpha_{\lambda}$  vs  $\Omega$  $= \omega/2\Gamma_0 \kappa^{3+x_\eta}$  using the same data points that were used in Fig. 1. In (b) we show  $\alpha_{\lambda}$  vs  $\Omega = \omega/2\Gamma_0 \kappa^{3+x_\eta} \ln(\kappa_0/\kappa)$  for the same set of data points as in (a). The quality of the straight-line fit is improved over that in (a).

Fig. 2(b) we show the same set of data points, now plotted against  $\omega/\Gamma_0 \kappa^z \ln(\kappa_0/\kappa)$ . The improvement in the quality of the straight line is visually obvious. While this is not conclusive in any way, it is certainly an issue that can be probed in future measurements.

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