# DROPLET MODEL FOR SOUND ABSORPTION IN FLUIDS NEAR THE CRITICAL POINT★

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The dynamic droplet model of critical fluids is applied to understanding critical fluid sound absorption. The resonance frequency of an oscillating droplet is used to explain the scaling on frequency and temperature observed in sound absorption data.

A number of experimental studies of sound absorption in single component or binary fluid systems near the critical point have been performed in the last several years [1–5]. These studies include both ultrasonic and Brillouin light-scattering techniques. These studies show that the sound absorption increases as the critical point is approached, indicating that this increased absorption may be related to the onset of large scale fluctuations in density or concentration characteristic of the critical region.

Perhaps the most interesting result of the experimental studies is that the critical sound absorption per wavelength, $\alpha_\lambda$, at any given frequency, $\omega$, or reduced temperature, $t = (T - T_C)/T_C$, can be expressed as a single function of the reduced frequency $\omega^*$ [2] where

$$\omega^* = \omega/\omega_D, \tag{1}$$

and where the characteristic frequency $\omega_D$ is

$$\omega_D = (2\Lambda/\rho C_p)\xi^{-2} = \omega_0 t^x. \tag{2}$$

In eq. (2) $\Lambda/\rho C_p$ is the thermal diffusivity and $\xi$ is the Ornstein–Zernike correlation length, $\xi = \xi_0 t^{-\nu}$, $\nu \approx 0.64$. The value of the exponent $x$ can be found from the sound absorption data to be $1.8 < x < 2.0$ [2, 4, 5]. The $\alpha_\lambda$ versus $\omega^*$ curve is rather sigmoidal in shape with $\alpha_\lambda$ increasing very slowly for $\omega^* \ll 1$, then increasing quickly in the region $\omega^* \approx 1$, and then leveling off or decreasing somewhat for $\omega^* \gg 1$ [2, 4, 5].

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Two theories in the mode coupling spirit have been proposed to explain this sound absorption phenomenon [2, 6, 7]. While it can be shown that these theories are not identical [8], they are both reasonably successful in describing the data in at least a qualitative manner, the theory by Kawasaki [6] not working well for $\omega^* > 10$. Mistura's theory [2, 7] predicts the value of $\omega_D$ given in eq. (2) and appears to be more successful in understanding both the absorption and dispersion data [8].

Recently a new model to explain critical fluid light scattering behavior has been proposed [9]. This very physical model is called the dynamic droplet model. It assumes the fluctuations in density or concentration in a critical fluid can be thought of as droplets with a given shape and size distribution which diffuse under the laws of random Brownian motion in the "background" fluid. In addition to describing the light-scattering data, the model has also been successful in describing the anomalous shear viscosity of a critical fluid [10]. In this note we shall show how the model can be applied to understand the magnitude and temperature dependence of the characteristic frequency $\omega_D$ on which the sound absorption data scale.

The dynamic droplet model gives us the physical picture of critical fluid as a fluid with a suspension of individual diffusing droplets of finite size. While the droplet sizes are given by a size distribution, we may take the Ornstein–Zernike correlation length $\xi$, as some sort of "average" size. Furthermore, we may infer from the form of the size distribution [9] that the surface tension $\sigma$ of the droplets is given by

$$\sigma \propto k_B T/\xi^2, \tag{3}$$

where $k_B$ is Boltzmann's constant and the proportionality constant in eq. (3) is of the order of unity. Eq. (3) is very reasonable as it is also the form for the interface surface tension for the fluid for temperatures less than the critical temperature [11].

We shall take the fluid in *one* droplet to be incompressible. This is a good assumption near the critical point for a single component system as well as for a binary system because while macroscopically the single component system is very compressible, this compressibility is a result of the *system* of droplets. The individual droplets will still be described on a microscopic level by the background properties of the fluid for which the incompressibility assumption is good. For such an incompressible, finite sized droplet with a surface tension we may expect the possibility of surface waves on the surface of the droplet.

To calculate the resonance frequency of a droplet, we shall follow the procedure of Rayleigh [12]. We assume irrotational flow, $\nabla \times \mathbf{v} = 0$, implying potential flow, $\mathbf{v} = -\nabla\phi$. With incompressibility this gives us the Laplace equation

$$\nabla^2\phi = 0, \tag{4}$$

which is easily solved. Rayleigh finds for surface oscillations of a spherical droplet of fluid with the condition of eq. (4) a resonance frequency of

$$\omega_R^2 = l(l-1)(l+2)\,\sigma/\rho a_0^3, \tag{5}$$

where $a_0$ is the equilibrium droplet radius and $l = 1, 2, 3 \ldots$.

We now present a model of sound absorption in a critical fluid. The fluid is described by the dynamic droplet model and so is composed of droplets of various sizes. These droplets have resonant surface oscillation frequencies given by eq. (5). For an incident sound wave of frequency $\omega \ll \omega_R$ the response of the droplets is small and little absorption occurs. However, for $\omega \approx \omega_R$ the droplets respond greatly and absorption processes inherent in the motion of the fluid are enhanced. As $\omega$ increases, successively higher-order modes of droplet oscillation are excited and the absorption remains fairly constant. Thus, with such a simple, physical picture we can very qualitatively explain the rather sigmoidal shape of the sound absorption data as described at the beginning of this note.

The point we wish to make here is that the absorption versus frequency curve should scale as the resonance frequency of the droplets. Thus we wish to identify $\omega_R$ of our model with $\omega_D$ of the data. To see if this is reasonable we find $\omega_R$ for an average sized droplet with $a_0 = \xi$. From eqs. (3) and (5) for the lowest mode ($l = 2$) we find

$$\omega_R \propto \frac{\sqrt{k_B T}}{\rho \xi^{5/2}}. \tag{6}$$

We can calculate $\omega_R$ for xenon from data for $\xi$ and $\rho$ above $T_C$ [13] and the surface tension *below* $T_C$ [14] and deduce

$$\omega_R = 7.6 \times 10^{12} t^{1.60}\ \mathrm{sec}^{-1}, \tag{7}$$

whereas from sound absorption data for xenon one finds (e.g. [4]),

$$\omega_D = 14.6 \times 10^{12} t^{1.87}\ \mathrm{sec}^{-1}, \tag{8}$$

though the data could be fitted by a characteristic $\omega_0$ in the range $2.5 \times 10^{12}\ \mathrm{sec}^{-1} < \omega_0 < 15 \times 10^{12}\ \mathrm{sec}^{-1}$. Thus our simple argument gives order of magnitude agreement for both $\omega_0$ and the critical exponent $x$.

Finally, we point out that our calculation here is only an indication of the physics behind the sound absorption mechanism. We have left out effects of the finite size distribution of the droplets by using an average size, and we have not included the effects of damping of the droplet oscillations. These considerations would not affect the conclusion that the absorption should be a universal function of frequency, but would affect merely the form of that function. We cannot allow for viscous damping without dropping the assumption of irrotational motion implied in the derivation of eq. (5), but the purely viscous absorption is probably small in the critical region.

We feel the argument presented above not only lends insight to the process of sound absorption in critical fluids, but also strengthens the concept of physical droplets in the critical fluid.

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