Thermoacoustic effects in supercritical fluids near the critical point: Resonance, piston effect, and acoustic emission and reflection

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We present a general theory of thermoacoustic phenomena in supercritical fluids near the critical point in a one-dimensional cell. We take into account the effects of the heat conduction in the boundary walls and the bulk viscosity near the critical point. We introduce a coefficient $Z(\omega)$ characterizing sound with frequency $\omega$ at the boundary. As applications, we examine the acoustic eigenmodes in the cell, the response to time-dependent perturbations, sound emission and reflection at the boundary. Resonance and rapid adiabatic changes are noteworthy. In these processes, the role of the thermal diffusion layer is enhanced near the critical point because of the strong critical divergence of the thermal expansion.

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I. INTRODUCTION

In highly compressible fluids, adiabatic changes take place with propagation of sounds and are much faster than the thermal diffusion [1]. When a one-component fluid is heated or cooled at a boundary, a thermal diffusion layer expands or shrinks to emit sounds, which then cause adiabatic changes in the interior (the thermal piston effect). The density change in the boundary layer is enhanced near the gas-liquid critical point because of the strong critical growth of the isobaric thermal expansion. As a result, thermal equilibration times become shorter near the critical point at fixed volume, despite the fact that the thermal diffusion constant $D$ tends to zero at the criticality $C_p/C_V$ is the specific-heat ratio growing around a heater and by rapid adiabatic density and traversal of sound pulses with width of order $10^{-7}$ g/cm$^3$ in near-critical CO$_2$ on the acoustic time scale using an ultra-sensitive interferometer. They detected emission and traversal of sound pulses with width of order 10µsec, which were broadened as they moved through the cell and interacted with the boundary walls. Some of their data agreed with predictions, but most data remain unexplained. Afterwords, part of the measured time-evolution of the density was numerically reproduced by Carlès, neglecting the bulk viscosity [21].

Recently, Carlès and Dadzie [14,15] found that the bulk viscosity, which grows strongly near the critical point, can affect the hydrodynamics in the thermal diffusion layer. Gillis et al. [16] performed experiments of acoustic resonance in xenon, where the frequency and attenuation of the resonating modes were measured. For such long wavelength sounds, the heat conduction at the boundary is the dominant damping mechanism relatively far from the critical point, while the viscous effect in the bulk becomes more important closer to the critical point. They also presented thorough theoretical analysis of their data. The critical growth of the effusivity and the bulk viscosity of the fluid both serve to suppress the boundary damping, as confirmed experimentally and theoretically. Very recently, Miura et al. [19,20] measured acoustic density changes with precision of order $10^{-7}$g/cm$^3$ in near-critical CO$_2$ on the acoustic time scale using an ultra-sensitive interferometer. They detected emission and traversal of sound pulses with width of order 10µsec, which were broadened as they moved through the cell and interacted with the boundary walls. Some of their data agreed with predictions, but most data remain unexplained. Afterwords, part of the measured time-evolution of the density was numerically reproduced by Carlès, neglecting the bulk viscosity [21].

Some unique aspects of the supercritical hydrodynamics have been revealed by experiments [22,23,24] and by simulations [18,19,25,26,27,28,29]. In Rayleigh-Bénard convection, overall temperature changes are induced by plume arrivals at the boundary walls due to the piston effect, leading to overshoot behavior observed in experiments of $^3$He near its critical point [22,26,27]. Significant noises of the adiabatic temperature changes were predicted in turbulent convective states [26], though not yet measured systematically. Recently three-dimensional simulations were performed [29]. Due to large thermal expansion in supercritical fluids, jet-like fluid flow has been observed around a heated boundary [23,24]. In these processes, the plume motions governed by the shear viscosity are strongly influenced by large thermal expansion around a heater and by rapid adiabatic density and
transport coefficients may be treated to be independent
\cite{13} and the growing bulk viscosity. In Section II, we will decompose fluid motions into sound modes and thermal diffusion modes with frequency $\omega$. These two modes are mixed at the boundary under given boundary conditions, leading to various thermoacoustic phenomena. In Section III, we will study the acoustic eigenmodes determined to various thermoacoustic phenomena. In Section II, we will decompose fluid motions into sound modes and thermal diffusion modes with frequency $\omega$. These two modes are mixed at the boundary under given boundary conditions, leading to various thermoacoustic phenomena. In Section III, we will study the acoustic eigenmodes determined to various thermoacoustic phenomena.

We will also examine the response of the fluid to various time-dependent perturbations. Resonance is induced when the frequency of the perturbation is close to one of the eigenfrequencies, while nearly uniform adiabatic changes are caused in the interior due to the piston effect at much lower frequencies. We will also examine sound emission and reflection at the boundary. In Appendix A, we will present a simple theory of the piston effect, which can be a starting point to understand the complicated calculations in the text. In Appendix B, the critical behavior of one-component fluids used in the text will be summarized.

II. THEORETICAL BACKGROUND

A. Linear hydrodynamics

Near the critical point, we treat the hydrodynamic deviations with spatial scales much exceeding the thermal correlation length $\xi$, but the typical frequency $\omega$ can be higher than the relaxation rate of the critical fluctuations $t_\xi^{-1} \propto e^{1.89} \xi \rho$ \cite{31, 32, 33}. For such high frequencies $\omega$, the bulk viscosity $\zeta$ behaves as $1/\omega$, while $\zeta \propto \omega^{-1.67}$ for $\omega t_\xi < 1$ \cite{31, 32, 33}. See Appendix B for more details. The other transport coefficients may be treated to be independent of $\omega$ \cite{1}. The critical singularity of the shear viscosity $\eta$ is negligible small, while the critical growth of the thermal conductivity $\lambda$ arises from the convective motions of the critical fluctuations taking place on a short time scale of order $\rho \xi^2/\eta$. Here we assume $\omega \ll \eta/\rho \xi^2$.

The mass density, the temperature, the entropy (per unit mass), and the pressure are written as $\rho$, $T$, $s$, and $p$, respectively, with their small deviations being $\delta\rho$, $\delta T$, $\delta s$, and $\delta p$. The velocity in the $x$ direction is written as $v$. These deviations depend on time $t$ as $\exp(i\omega t)$ and vary in space along the $x$ axis. We may assume $\omega > 0$ without loss of generality (see Eq.(3.1)). These deviations may be regarded as the Fourier transformations of the space-time dependent deviations with respect to time ($= \int dt \exp(-i\omega t (\cdot \cdot \cdot))$). They obey the linear equations \cite{34},

\[
\begin{align*}
\text{i} \omega \delta \rho &= -\rho v', \\
\text{i} \omega \rho v &= -\delta \rho + \rho \nu v'', \\
\text{i} \omega \rho T \delta s &= \lambda \delta T''.
\end{align*}
\]

Here the prime denotes the differentiation with respect to $x$. We have two dissipative coefficients; one is the thermal conductivity $\lambda$ and the other is $\nu_t = (\zeta + 4\eta/3)/\rho$, \eqref{eq:2.4}

where $\zeta$ and $\eta$ are the bulk and shear viscosities, respectively. Using the thermodynamic derivatives we may express $\delta s$ and $\delta p$ in terms of $\delta \rho$ and $\delta T$ as

\[
\begin{align*}
\rho T \delta s &= C_V [\delta T - b^{-1} \delta p], \\
\delta p &= \gamma^{-1} c^2 \delta T + (1 - \gamma^{-1}) a_s \delta T,
\end{align*}
\]

where $c^2 = \partial p/\partial \rho$ is the square of the sound velocity,

\[
\gamma = C_p/C_V
\]

is the specific-heat ratio with $C_p = \rho T (\partial s/\partial T)_p$ and $C_V = \rho T (\partial s/\partial T)_p$ being the isobaric and constant-volume specific heat (per unit volume), respectively. To avoid cumbersome notation, we write

\[
\begin{align*}
a_s &= \left( \frac{\partial p}{\partial T} \right)_s, \\
b_s &= \left( \frac{\partial p}{\partial T} \right)_s = c^{-2} a_s.
\end{align*}
\]

For low-frequency sounds, the adiabatic relations $\delta p \cong a_s \delta T$ and $\delta \rho \cong b_s \delta T$ should hold. We use the following thermodynamic identities \cite{1},

\[
\left( \frac{\partial p}{\partial T} \right)_\rho = (1 - \gamma^{-1}) a_s = \rho c^2 C_V / T a_s.
\]

Next we consider small hydrodynamic deviations by assuming the space-dependence in the sinusoidal form $\exp(iq x)$. From Eqs.(2.3) and (2.5) $\delta \rho$ and $\delta T$ are related by

\[
\delta T = \frac{\text{i} \omega^2}{(\text{i} \omega + \gamma D q^2) a_s} \delta p,
\]

where $D = \lambda / C_p$ is the thermal diffusion constant. Equations Eq.(2.1)-(2.3) give the dispersion equation between $q$ and $\omega$,

\[
[\omega^2 - (\text{i} \omega \nu_t + \gamma^{-1} c^2)^2] (\text{i} \omega + \gamma D q^2)
= \text{i} \omega c^2 (1 - \gamma^{-1}) q^2.
\]

If we set $g = \omega / c \sqrt{X}$ or $X = (\omega / c g)^2$, the dimensionless quantity $X$ obeys the quadratic equation,

\[
X^2 - (1 + \Delta_v + \gamma \Delta_T) X + (1 + \gamma \Delta_v) \Delta_T = 0,
\]

where we introduce two dimensionless coefficients representing the dissipation strength \cite{18},

\[
\begin{align*}
\Delta_v &= \text{i} \omega \nu_t / c^2, \\
\Delta_T &= \text{i} \omega D / c^2.
\end{align*}
\]

If $\omega > 0$, $\Delta_v$ and $\Delta_T$ are purely imaginary. The ratio $\Delta_v / \Delta_T = \nu_t / D$ grow strongly near the critical point (see Appendix B).
For given \( \omega \), Eq. (2.11) or Eq. (2.12) yields four solutions \( q = \pm q_+ = \pm \omega / c \sqrt{X_\pm} \), where \( X_+ \) and \( X_- \) are the solutions of Eq. (2.12) written as

\[
X_\pm = \frac{1}{2} (1 + \Delta_v + \gamma \Delta_T \mp \Xi),
\]

\[
= \frac{2(1 + \gamma \Delta_v) \Delta_T}{1 + \Delta_v + \gamma \Delta_T \pm \Xi}, \tag{2.15}
\]

where we define

\[
\Xi = [(1 + \Delta_v - \gamma \Delta_T)^2 + 4(\gamma - 1) \Delta_T]^{1/2}, \tag{2.16}
\]

with \( \text{Re} \Xi > 0 \). The second line of Eq. (2.15) follows from \( X_+ X_- = (1 + \gamma \Delta_v) \Delta_T \). The modes with \( q = \pm q_- \) represent the sound, while those with \( q = \pm q_+ \) the thermal diffusion. We may define \( q_- \) and \( q_+ \) such that \( \text{Re}(q_-/\omega) > 0 \) and \( \text{Im} q_+ < 0 \) hold. It is convenient to introduce \( k \) and \( \kappa \) by

\[
k = q_- = \frac{\omega}{c \sqrt{X_-}}, \quad \kappa = q_+ = \frac{\omega}{c \sqrt{X_+}}. \tag{2.17}
\]

The argument of \( X_- \) is in the range \([0, \pi/2]\) for \( \omega > 0 \), leading to \( \text{Im} k < 0 \), which implies that sound waves propagating in the positive \( x \) direction \( (\propto e^{-i k x}) \) are damped with increasing \( x \).

As \( \omega \to 0 \), we may treat \( \Delta_v \) and \( \Delta_T \) as small quantities. To their first order we find \( X_+ \simeq \Delta_T \) and \( X_- \simeq 1 + \Delta_v + (\gamma - 1) \Delta_T \) so that \( \kappa \simeq \sqrt{\omega / D} \) and \( k \simeq \omega / c - i \Gamma \omega^2 / 2 c^3 \), where \( \Gamma \)

\[
\Gamma_s = (\zeta + 4 \eta / 3) / \rho + (\gamma - 1) D. \tag{2.18}
\]

is the attenuation constant in the long wavelength limit. We have \( |\kappa| \gg |k| \) at low frequencies. For example, \( |\kappa| \sim 10^5 \text{ cm}^{-1} \) and \( |k| \sim 10^{-2} \text{ cm}^{-1} \) for \( \omega = 10^4 \text{ s}^{-1}, \ D = 10^{-6} \text{ cm}^{-1} \text{s}^{-1} \), and \( c = 10^5 \text{ cm/s} \). In a cell with length \( L \), the strength of the bulk dissipation of sounds is represented by the damping factor \( \exp(-\beta B \ell) \) with

\[
\delta_B = -\text{Im} k \\
\cong \Gamma \omega^2 / 2 \rho c^3, \tag{2.19}
\]

where the second line is the low-frequency expression. Mathematically, we may consider the high frequency limit \( |\Delta_v| \gg 1 \) and \( |\Delta_T| \gg 1 \) neglecting the frequency-dependence of the transport coefficients to derive the limiting behavior \( k \to (i \omega / \nu_0)^{1/2} \) and \( \kappa \to (i \omega / \gamma D)^{1/2} \), though this limit is unrealistic. In this paper, we will assume \( |\Delta_T| \ll \gamma^{-1} \) in Eq. (2.34), because it is satisfied in realistic experimental conditions, as will be discussed.

B. Solutions in a finite cell

We consider small hydrodynamic perturbations behaving as \( e^{\nu t} \) in a fluid in a finite cell with length \( L \). The density deviation can be expressed in the following linear combination,

\[
\delta \rho = ae^{-\kappa x} + be^{\kappa (x-L)} + \alpha e^{ikx} + \beta e^{-ikx}. \tag{2.20}
\]

The coefficients \( a, b, \alpha, \) and \( \beta \) depend on time as \( e^{\nu t} \). The first and second terms represent the deviations in the thermal diffusion layers. The thickness of the layers is given by \( 1/|\kappa| \), which is assumed to be much shorter than the cell length \( L \), so

\[
|\kappa| \gg 1/L. \tag{2.21}
\]

Then the second (first) term is virtually zero near \( x = 0 \) \( (x = L) \). The third term in Eq. (2.20) represents a sound propagating in the negative \( x \) direction, while the fourth term a sound propagating in the positive \( x \) direction. From Eq. (2.1) the velocity is expressed as

\[
v = \frac{i \omega}{\rho k} [a e^{-\kappa x} - b e^{\kappa (x-L)}] - \frac{\omega}{\rho k} [\alpha e^{ikx} - \beta e^{-ikx}]. \tag{2.22}
\]

If the boundary walls are fixed in time, we should require \( v = 0 \) at \( x = 0 \) and \( L \) to obtain

\[
a = (\kappa / ik)(\alpha - \beta), \tag{2.23}
\]

\[
b = -(\kappa / ik)(\alpha e^{ikL} - \beta e^{-ikL}). \tag{2.24}
\]

Note that the mass change in the thermal diffusion layers is \( (a + b) / \kappa \) and that in the interior is \( \alpha (e^{ikL} - 1) / ik \) \( + \beta (1 - e^{-ikL}) / ik \) per unit area. From Eqs. (2.23) and (2.24) these two changes cancel, ensuring the mass conservation. Use of Eq. (2.10) gives the temperature deviation in the following linear combination,

\[
\delta T = \frac{i \omega}{b_s} \frac{\alpha e^{-\kappa x} + \beta e^{\kappa (x-L)} + \alpha e^{ikx} + \beta e^{-ikx}}{i \omega - \gamma D \kappa^2} - \frac{i \omega}{i \omega + \gamma D k^2}. \tag{2.25}
\]

Let \( Q_0 \) and \( Q_L \) be the heat flux \(-\lambda \delta T^* \) at \( x = 0 \) and \( L \), respectively. Use of Eqs. (2.23)-(2.25) gives

\[
\frac{\beta - \alpha}{Q_0} = \frac{\beta - \alpha e^{2ikL}}{e^{ikL} Q_L} = \frac{b_s (\gamma - 1) ik}{\lambda (1 + \gamma \Delta_v) k^2 + \kappa^2}, \tag{2.26}
\]

where \( b_s (\gamma - 1) / \lambda = \rho / Ta_c D \) with the aid of Eq. (2.9). The above quantities tend to the constant \( \rho / Ta_c D \) in the low frequency limit. We may use Eq. (2.26) when \( Q_0 \) is a control parameter or when \( Q_L \) is measurable.

The coefficients \( a, b, \alpha, \) and \( \beta \) can be determined if we specify the boundary conditions at \( x = 0 \) and \( L \). Hereafter we assume no temperature discontinuity at the boundaries. In most theoretical calculations the boundary temperatures are fixed, but in some papers the bottom heat flux \( Q_0 \) is fixed \([26]\). In this paper, we consider a more realistic boundary condition of the temperature accounting for the thermal conduction in the boundary wall regions \([13, 18]\). Here we assume that \( \delta T \) tends to zero in the solid far from the boundaries without heat input. In the solid region \( (x < 0) \), the temperature deviation then decays as \( \delta T^* (0) e^{\kappa w x} \) with

\[
\kappa_w = (i \omega C_w / \lambda_w)^{1/2}, \tag{2.27}
\]
where $\lambda_w$ and $C_w$ are the thermal conductivity and the heat capacity (per unit volume) of the solid, respectively. The $1/|\kappa_w|$ is the thickness of the thermal diffusion layer in the solid and is assumed to be shorter than the thickness of the wall. Without temperature discontinuity at the boundary, the energy balance at $x = 0$ yields

$$\delta T' = \lambda_w \kappa_w \delta T / \lambda = a_w (i\omega / D)^{1/2} \delta T,$$  

(2.28)

where $\delta T$ and $\delta T'$ are the values at $x = 0$. In the second line, the coefficient $a_w$ is the effusivity ratio $\frac{C_w}{C_s} \frac{\lambda}{\lambda}$.

For CO$_2$ in a Cu cell [20] we have $a_w = 3 \times 10^3 \psi$ where $\psi$. The boundary temperature at $x = 0$ is fixed or $\delta T(0) = 0$ for $a_w \rightarrow \infty$, while the boundary is thermally insulating or $(d\delta T/dx)_{x=0} = 0$ as $a_w \rightarrow 0$. On the other hand, if the other boundary wall in the region $x > L$ is made of the same material, the boundary condition at $x = L$ reads

$$\delta T' = -a_w (i\omega / D)^{1/2} \delta T,$$  

(2.30)

with the same $a_w$ as in Eq.(2.28), where $\delta T$ and $\delta T'$ are the values at $x = L$.

The boundary conditions at $x = 0$ give Eqs.(2.23) and (2.28), from which we may readily calculate the reflection factor $Z \equiv \beta / \alpha$ between the outgoing and incoming sound waves. It is convenient to introduce the combination,

$$W = \frac{\alpha - \beta}{\alpha + \beta} = \frac{1 - Z}{1 + Z},$$  

(2.31)

because $W$ is a small quantity in our system. Some calculations yield a general expression,

$$W = \frac{-ik(i\omega - \gamma D k^2) / \kappa}{i\omega + \gamma D J^2 + \sqrt{i\omega D (k^2 + k^2) / a_w \kappa}},$$  

(2.32)

in terms of $k$ and $\kappa$. In the case of a thermally insulating boundary, we have $W = 0$ and $Z = 1$ by setting $a_w \rightarrow 0$ in Eq.(2.32). The interaction of sounds and the boundary wall is characterized by $Z$ or $W$, where the wall properties appear only through the effusivity ratio $a_w$ and the system length $L$ does not appear.

C. Adiabatic condition in the interior

We will clarify an upper bound of the frequency, below which the sound motions in the interior are adiabatic or without entropy deviations. Under this adiabatic condition, the results from the linear hydrodynamic equations can be much simplified.

Far from the boundary walls or outside the thermal diffusion layers, we may neglect the localized modes to obtain the interior hydrodynamic deviations. From Eqs.(2.5), (2.6), and (2.25), those of the density, temperature, and pressure are related by

$$\delta \rho = |1 + \gamma D k^2 / i\omega| b, \delta T,$$

$$\delta \rho = |1 + D k^2 / i\omega| a, \delta T.$$

(2.33)

Here $x$ and $L - x$ are much longer than $1/|\kappa|$. The second term in the brackets arise from a small entropy deviation in the interior. Since $\gamma > 1$, the usual adiabatic relations hold in the interior under the condition,

$$|\gamma D k^2 / i\omega| \sim \gamma |\Delta T| \ll 1 \quad \text{or} \quad \omega \ll c^2 / \gamma D,$$

(2.34)

where $\Delta T$ is defined by Eq.(2.14). This condition is well satisfied in the usual hydrodynamic processes. Even near the critical point, the time $t_{ad} \equiv \gamma D / c^2$ remains very short. For example, $t_{ad} \approx 7.6 \times 10^{-14} \epsilon^{-0.62}$ sec for CO$_2$.

Under Eq.(2.34) we have $\Xi \equiv 1 + \Delta_v$ so that $X_+$ and $X_-$ in Eq.(2.15) are approximated as

$$X_+ = \frac{1 + \gamma \Delta_v}{1 + \Delta_v} \Delta T, \quad X_- = 1 + \Delta_v.$$

(2.35)

The wave numbers $k$ and $\kappa$ are expressed as

$$\kappa = \left( \frac{i\omega}{D} \right)^{1/2} \left( \frac{1 + \Delta_v}{1 + \gamma \Delta_v} \right)^{1/2}, \quad k = \frac{\omega / c}{\sqrt{1 + \Delta_v}}.$$

(2.36)

We retain $\Delta_v$, since it becomes appreciable near the critical point because of the strong critical divergence of $\zeta$. As will be discussed in Appendix B, $\zeta \equiv \rho c^2 R_B t_\xi$ for $\omega t_\xi < 1$, where $R_B \equiv 0.03$ is a universal number and $t_\xi = \xi^2 / D$ is the characteristic time of the critical fluctuations with $\xi$ being the correlation length. Carlès found the dependence of $\kappa$ on the singular combination $\zeta$ as in Eq.(2.36) [16, 17]. By setting $\gamma \Delta_v = i\omega t_B$, we introduce a new characteristic time $t_B$ as

$$t_B = \gamma / \rho c^2 = R_B \gamma t_\xi.$$

(2.37)

Then $t_B \gg t_\xi$ once $R_B \gg 1$. For CO$_2$, $t_B = 1.9 \times 10^{-15} \epsilon^{-3.6}$ sec. See Table 1 and Fig. 1 for the characteristic times with $L = 1$ cm, where $t_B$ exceeds the acoustic time $L/c$ for $\epsilon < 3 \times 10^{-4}$ and the modified piston time $t_1$ (to be introduced in Eq.(3.16)) for $\epsilon < 3 \times 10^{-5}$. There can be a sizable frequency range with $t_B^{-1} < \omega < t_B^{-1}$ at small $\epsilon$, where $\kappa$ becomes independent of $\omega$ as

$$\kappa \equiv (\rho c^2 / \gamma D \zeta)^{1/2} \equiv (R_B \gamma)^{-1/2} \xi^{-1}.$$

(2.38)

The thickness of the thermal diffusion layer $1/|\kappa|$ remains longer than $\xi$ by $(R_B \gamma)^{1/2}$. Also from the expression of $k$ in Eq.(2.36) we write the sound dispersion relation as $k = \omega / c^*(\omega)$, where we define the complex sound velocity [1],

$$c^*(\omega) = c \sqrt{1 + \Delta_v},$$

(2.39)

whose critical behavior will be discussed in Appendix B.

As $\epsilon$ is decreased, we first encounter the regime where $W$ grows but $a_w \gg 1$ and $\omega t_B < 1$ still hold. However,
the critical growth of $W$ is eventually suppressed by the growing $a_w^{-1}$ and $\zeta$. If we use Eqs. (2.35) and (2.36) under Eq. (2.34), we approximate $W$ in Eq. (2.32) as

$$W = \frac{(\gamma - 1)\sqrt{\Delta_T}}{(1 + \Delta_v)X_v},$$  

(2.40)

where we define

$$X_v = \sqrt{1 + \gamma \Delta_v + a_w^{-1}\sqrt{1 + \Delta_v}}.$$  

(2.41)

The limiting behaviors of $X_v$ are as follows: $X_v \approx 1 + a_w^{-1}$ for $\omega \ll t_B^{-1}$ and $X_v \approx (i\omega t_B)^{1/2}$ for $\omega \gg t_B^{-1}(1 + a_w^{-1})^2$.

For $\omega \ll t_B^{-1}$, it follows the classical expression valid far from the critical point,

$$W = (\gamma - 1)\sqrt{\Delta_T/(1 + a_w^{-1})} = \sqrt{\Delta_2},$$  

(2.42)

which is the result without the viscous effect and under the isothermal boundary condition. We may introduce a characteristic time $t_2$ defined by

$$t_2 = [a_w/(1 + a_w)]^2(\gamma - 1)^2D/c^2,$$  

(2.43)

which includes the effect of the heat conduction in the wall. As shown in Table 1 and Fig. 1, $t_2$ is very short even compared with $t_\xi$. In the literature (see Section 77 of Ref. [84]), it is argued that the amplitude of a plane wave sound is decreased by the factor $(\gamma - 1)\sqrt{2\Delta/\epsilon}$ upon reflection at an isothermal boundary wall. This factor is obviously equal to $1 - |Z| \approx 2\text{Re}W$ if use is made of Eq. (2.42).

![TABLE I: Parameters of CO$_2$ in a Cu cell with $L = 1$ cm for $\epsilon = 10^{-3}$ (first line), $10^{-4}$ (second line), and $10^{-5}$ (third line). Times are in sec.](image)

| $\gamma$ | $a_w$ | $t_\xi \times 10^6$ | $t_B \times 10^6$ | $L/c \times 10^4$ | $t_1 \times 10^6$ | $t_2 \times 10^6$ |
|----------|-------|---------------------|---------------------|---------------------|---------------------|---------------------|
| 260      | 5.0   | 0.24                | 1.9                 | 0.71                | 1.0                 | 0.12                |
| 3600     | 0.63  | 18                  | 1900                | 0.83                | 0.1                 | 1.7                 |
| 5 x 10$^2$ | 0.075 | 1300                | $1.7 \times 10^9$   | 0.98                | 0.08                | 3.0                 |

![FIG. 1: Characteristic times $t_\xi$ in Eq. (B3), $t_B$ in Eq. (2.37), $t_2$ in Eq. (2.43), $t_1$ in Eq. (3.16), and $L/c$ vs $\epsilon = T/T_c - 1$ for CO$_2$ in a Cu cell with $L=1$ cm.](image)

![FIG. 2: ReW, ImW, and $|\Delta_v| = \omega \zeta/\rho c^2 \omega L/\pi c$ at $\epsilon = 10^{-3}$ (left) and $10^{-4}$ (right) for CO$_2$ in a Cu cell with $L=1$ cm. As $\omega$ exceeds $t_B^{-1}$, ReW tends to saturate and ImW decreases, due to the growing bulk viscosity and the decreasing effusivity ratio.](image)

D. Hydrodynamic variables in the adiabatic condition

Under Eq. (2.34) we obtain simple expressions of the deviations of the temperature, the pressure, and the entropy including $\Delta_v$. From Eqs. (2.5), (2.6), (2.9), and (2.20) we find

$$\delta T = \left(\frac{\partial T}{\partial \rho}\right)_p (1 + \Delta_v)[\delta \rho]_b + b_s^{-1}[\delta \rho]_{in}$$  

(2.46)

$$\delta s = \left(\frac{\partial s}{\partial \rho}\right)_p (1 + \Delta_v)[\delta \rho]_b,$$  

(2.47)

$$\delta p = -c^2\Delta_v[\delta \rho]_b + c^2[\delta \rho]_{in},$$  

(2.48)

where $[\delta \rho]_b = a\epsilon^{kx} + b\epsilon^{-kx}$ is the density deviation localized near the boundaries and $[\delta \rho]_{in} = \alpha e^{ikx} + \beta e^{-ikx}$ is the interior density deviation. In deriving Eq. (2.46) use has been made of the thermodynamic relation $(1 - \gamma)[\delta \rho]_b = (\partial \rho/\partial T)_p$. Under Eq. (2.34), the entropy deviation $\delta s$ is localized near the boundaries, while the localized part of the pressure deviation $\delta p$ is nonvanishing to satisfy...
\[ \omega \rho v_0 = -\delta p' + \rho \nu v_0' = -\delta p' - i\omega \nu \delta p' \rightarrow 0 \text{ as } x \rightarrow 0 \text{ and } L \] in Eq. (2.2).

We examine the deviations close to the boundary at \( x = 0 \) by assuming Eqs. (2.23) and (2.28) and setting \( Z_0 = \beta \). In this case the density ratio \( |\delta \rho|/|\delta \rho|_0 \) tends to \((\gamma - 1)/X_v \sqrt{1 + \gamma \Delta_v} \) as \( x \rightarrow 0 \), so that

\[ b_0 \delta T = (\alpha + \beta) \left[ 1 - \frac{1}{X_v} \sqrt{1 + \gamma \Delta_v} e^{-\alpha x} \right], \quad (2.49) \]

\[ \frac{\delta \rho}{c^2} = (\alpha + \beta) \left[ 1 - \frac{1}{X_v} \sqrt{1 + \gamma \Delta_v} e^{-\alpha x} \right], \quad (2.50) \]

where \( 0 < x \ll 1/|k| \). Note that the second terms in the brackets in Eqs. (2.49) and (2.50) tend to unity for \( \omega t_B \gg (1 + a_w^{-1})^{-2} \), and can be the case of very large \( \omega \).

In the original work \[3\], the pressure homogeneity and the isobaric relations among the hydrodynamic variables were assumed in the thermal diffusion layers. We recognize that the pressure homogeneity and the isobaric condition hold only in the low frequency limit \( \omega \ll t_B^{-1}(1 + a_w^{-1})^{-2} \).

### III. APPLICATIONS

#### A. Acoustic modes in a cell

Gillis \textit{et al.} \[18\] calculated the acoustic eigenmodes for their experimental geometry, taking into account the growing \( a_w^{-1} \) and \( \zeta \). In the following, we will present a simpler version in a one-dimensional cell, \( 0 < x < L \), taking into account these two ingredients. In this case \( \omega \) is treated as one of the eigenvalues and is complex, while we have assumed \( \omega > 0 \) in the previous section. Then \( \omega \) should have a positive imaginary part for the stability of the system. Here \( \sqrt{\omega} = (1 + i) \sqrt{\omega}/2 \) for \( \omega > 0 \), while \( \sqrt{\omega} = (1 - i) \sqrt{\omega}/2 \) for \( \omega < 0 \). The latter follows from the requirement that the real part of \( \omega \) in Eq. (2.17) should be positive. For general complex \( \omega \), all the quantities introduced so far should be functions of \( \omega \) analytic for \( \Re(\omega) > 0 \) or for \( \Im(\omega) < 0 \). Therefore, \( X_\pm(\omega) = X_\pm(\omega^*)^* \) and

\[ Z(-\omega^*) = Z(\omega^*), \quad W(-\omega^*) = W(\omega^*), \quad (3.1) \]

where \( \omega^* \) is the complex conjugate of \( \omega \).

Under the boundary conditions (2.28) and (2.30), the interior density deviation is expressed as

\[ \delta \rho = \alpha (e^{ikx} + Ze^{-ikx}), \]

\[ = \alpha'(e^{ik(L-x)} + Ze^{-ik(L-x)}), \quad (3.2) \]

in terms of \( Z = \beta / \alpha \). The first and second lines follow from Eqs. (2.28) and (2.30), respectively, and should coincide so that \( \alpha'(L-x) = \beta \) and \( \alpha' Ze^{-ikL} = \alpha \), leading to \( Ze^{-ikL} = Z^{-1} e^{ikL} \). We now find the condition of the eigenmodes,

\[ Z = \pm e^{ikL}, \quad (3.3) \]

FIG. 3: Normalized damping constants \( \alpha_\lambda, \alpha_\zeta, \) and \( \alpha_\lambda + \alpha_\zeta \) defined by (3.3) vs \( \epsilon = T/T_c - 1 \) for \( n = 1 \) (left) and \( n = 4 \) (right). The resonant frequencies are close to \( \omega = n\pi c/L \approx n 	imes 4 	imes 10^4 \text{sec}^{-1} \), which are exceeded by \( t_B^{-1} \) close to the critical point as marked by the arrows.

where \( + \) corresponds to even modes and \( - \) to odd modes. Namely, the density and temperature deviations are even close to the critical point as marked by the arrows.

In Fig. 3, we show \( \alpha_\lambda, \alpha_\zeta, \) and \( \alpha_\lambda + \alpha_\zeta \) functions of \( n = 1, 2, 3 \cdots \) for the even (odd) modes. For \( W = (1 - Z)/(1 + Z) \) calculated in Eq. (2.32) or Eq. (2.40), we obtain

\[ W = -i \tan(\kappa L/2) \] (even modes)

\[ = i \cot(\kappa L/2) \] (odd modes). \quad (3.4)

Since \( W \) is small, the eigenfrequencies \( \omega_n \) \( (n = 1, 2, 3 \cdots) \) are nearly equal to \( n\pi c/L \) where \( c^* \) is defined by Eq. (2.39). In the case \( |W| \ll 1 \) the leading correction from \( W \) can be written as

\[ \omega_n = \left( n\pi + 2iW + \cdots \right)c^*/L, \]

\[ = \left[ 1 + i\alpha_\lambda + i\alpha_\zeta + \cdots \right] \Re(\omega_n), \quad (3.5) \]

where \( n = 2, 4, \cdots \) for the even modes and \( n = 1, 3, \cdots \) for the odd modes. We assume small bulk damping \( |\Delta_v| \ll 1 \) and define

\[ \alpha_\lambda = 2n\Re(\omega), \quad \alpha_\zeta = \frac{1}{2} |\Delta_v| = n\pi \frac{\zeta}{2\rho c L}, \quad (3.6) \]

where \( \alpha_\lambda \) represents the boundary damping and \( \alpha_\zeta \) the bulk damping. The resonance quality factor \( Q^{-1} \) is equal to \( 2(\alpha_\lambda + \alpha_\zeta) \) in our notation. The resonance frequency including the shift is given by the real part,

\[ \Re(\omega_n) = (n\pi - 2i\Re(\omega) = \Re(\omega_n). \quad (3.7) \]

The frequency \( \omega \) in \( c^* \) and \( W \) may be equated with \( \Re(\omega_n) \approx n\pi c/L \).

In Fig. 3, we show \( \alpha_\lambda, \alpha_\zeta, \) and the sum \( \alpha_\lambda + \alpha_\zeta \) as functions of \( \epsilon \) in the regime \( \omega t_B < 1 \) for the odd mode of \( n = 1 \) and the even mode of \( n = 4 \) for \( \text{CO}_2 \) in a \( \text{Cu} \) cell with \( L = 1 \text{cm} \). We notice the following. (i) For such long wavelength sounds, the boundary damping is relevant far from the critical point, but the bulk damping eventually dominates close to the critical point. (ii) According with the discussion around Eq. (2.44), \( \alpha_\lambda \)
decreases on approaching the criticality in the region of $t_B > \omega^{-1} \sim L/n\pi c$, with a maximum at $\omega t_B \sim 1$. As a result, the curve of the sum $\alpha_\lambda + \alpha_\zeta$ is flattened considerably around $t_B \sim \omega^{-1}$.

These theoretical results are consistent with the experimental data by Gillis et al. They performed the resonance experiment over a wide range of $\omega t_\xi$ (up to about 200) to measure the frequency-dependent bulk viscosity. In agreement with the theory, $\alpha_\zeta$ or $\omega_\zeta/pc^2$ became independent of $\epsilon$ in the high-frequency regime $\omega t_\xi > 1$ (see Eq.(B5) in Appendix B).

B. Periodic perturbations

Periodic perturbations may be applied to a fluid in a cell in various manners. Resonance can occur when the frequency $\omega$ is close to $\text{Re} \omega_n$. It is sharp for small $\text{Im} \omega_n$. We will give three boundary conditions at $x = 0$ leading to resonance. We assume the boundary condition Eq.(2.30) at $x = L$. Then use of Eq.(3.2) yields $\alpha = \beta Z e^{-2ikL}$. The interior density deviation is of the form,

$$\delta \rho = \beta e^{-ikx} + \beta Z e^{ikx-2ikL},$$

where the term proportional to $Z$ arises from the reflection at $x = L$.

The bulk damping of the reflected waves is represented by $|e^{-ikL}| = e^{-\delta_B L}$. From Eq.(2.19) $\delta_B$ is expressed as

$$\delta_B = A_B (\omega/\pi c)^2,$$

in the low frequency regime $\omega t_\xi < 1$. We find $A_B = 0.5 \times 10^{-3}$ cm and 0.03 cm at $\epsilon = 10^{-3}$ and $10^{-4}$, respectively, for CO$_2$. In the relatively high frequency range $\omega > (2A_B)^{-1/2} \pi c/L$, the factor $e^{-2ikL} (\propto e^{-2\delta_B L})$ becomes negligibly small. Then, near the boundary, $\delta \rho$ consists of the outgoing wave only, resulting in no resonance. On the other hand, in the high frequency regime $\omega t_\xi > 1$, Eq.(B6) gives

$$\delta_B \approx \omega \text{Im} \Delta_\nu /2c \approx 0.27 \omega/\pi c,$$

which means that a sound emitted at $x = 0$ reaches the other end with the damping factor $e^{-0.27}$ for the first resonance frequency $\omega \approx \pi c/L$.

1. Temperature oscillation

In the first example, the temperature in the wall region $x < 0$ is oscillated, while the boundary walls are mechanically fixed. More precisely, we require $\delta T \rightarrow T_w \propto e^{i\omega t}$ as $x \rightarrow -\infty$; then, $\delta T(x) = e^{i\omega x} [\delta T(0) - T_w] + T_w$ in the region $x < 0$. The thermal boundary condition at $x = 0$ is then given by

$$\delta T' = a_w (i\omega/D)^{1/2} (\delta T - T_w),$$

as a generalization of Eq.(2.28). Some calculations using Eqs.(2.23) and (2.25) give the response function defined by $R_T \equiv \beta/b_s T_w$ in the form,

$$R_T = \frac{1}{2} (1 + \gamma Dk^2/\omega) \frac{1 - Z}{1 - Z^2 e^{-2ikL}}.$$

Notice that $R_T$ diverges as $R_T \equiv cW/2iL(\omega - \omega_n)$ for $\omega = \omega_n (n = 1, 2, \cdots)$ in the complex $\omega$ plane from Eq.(3.3). Under the adiabatic condition Eq.(2.34), the interior temperature deviation is expressed as

$$\delta T = b^{-1}_s \delta \rho = (e^{-ikx} + Ze^{ikx-2ikL}) R_T T_w.$$

Furthermore, neglecting $\gamma Dk^2/\omega$ in Eq.(3.12) and using $|W| \ll 1$ (see Fig. 2), we obtain

$$R_T \approx W/[1 - (1 - 4W)e^{-2ikL}].$$

In Fig. 4, we plot the absolute value $|R_T|$ calculated from Eq.(3.12) vs the normalized frequency $\omega L/\pi c$ at $\epsilon = 10^{-3}$ and $10^{-4}$, using the data for CO$_2$ in a Cu cell with $L = 1$ cm [20]. It exhibits peaks at $\omega \approx n\pi c/L$ as expected, but its peak heights do not exceeds 1/2 due to the small factor $1 - Z \approx 2W$ in the numerator in Eq.(3.12)

As discussed below Eq.(3.9), the resonant peaks should disappear for $\omega L/\pi c > (2A_B)^{-1/2}$, where we may neglect $e^{-2ikL}$ in $R_T$ to obtain $R_T \approx W$. These results are in accord with Fig. 4, since $(2A_B)^{-1/2} \approx 30$ and 4 for $\epsilon = 10^{-3}$ and $10^{-4}$, respectively.

In the low frequency case $\omega \ll c/L$, the interior deviations become nearly homogeneous. Figure 1 indicates that $t_B$ can much exceed $L/c$ very close to the critical
point, while $|\Delta_v| \ll 1$ holds. Thus, retaining $\gamma \Delta_v$, we set $e^{ikL} \approx 1 + ikL$ and $1 + \Delta_v \approx 1$ and use Eqs. (2.40) and Eq. (3.12) to find

$$R_T \approx \frac{1}{4\sqrt{\omega t_1 X_v + 1}}^{-1},$$

(3.15)

where $t_1$ is defined by Eq. (1.1) and $X_v$ by Eq. (2.41). If $\omega \ll t_1^{-1}$, we further have $R_T \approx (1 + \omega t_1)/4\sqrt{\omega t_1 X_v + 1}$ with

$$t_1' = (1 + a_w^{-1})^2 t_1 = (1 + a_w^{-1})^2 L^2/4(\gamma - 1)^2 D,$$

(3.16)

which is related to $t_2$ in Eq. (2.43) by $t_1' t_2 = L^2/4c^2$. The $t_1'$ first decreases as $t_1 \sim \epsilon^{-2.26}$ for $a_w \gg 1$ but finally weakly increases as $t_1 a_w^{2} \equiv \epsilon^{-0.22}$ for $a_w \ll 1$. See Fig. 1 for the curve of $t_1'$. As will be discussed in Subsection III C, $t_1'$ is the piston time including the effect of the wall heat conduction [14].

We need to know when $|\sqrt{\omega t_1 X_v}| \gg 1$ holds. It holds for $\omega \gg 1/t_1'$ under the condition,

$$(1 + a_w^{-1})^{-2} t_1' = (1 + a_w^{-1})^{-4} t_1 \ll 1.$$  

(3.17)

If $a_w < 1$ for CO$_2$ in a Cu cell, the above condition becomes $a_w^4 t_1' t_1 = 2 \times 10^{-6} \epsilon^{-0.97}/L^2 \ll 1$ with $L$ in cm, which is well satisfied for $\epsilon \gg 10^{-6}$ with $L = 1$cm. If $\omega t_1' \ll 1$ under Eq. (3.17), we find

$$\delta T \approx T_w/2,$$

(3.18)

in the interior. Note that the reverse condition of Eq. (3.17), $a_w^4 t_1' t_1 > 1$, holds extremely close to the critical point, where $|\sqrt{\omega t_1 X_v}| > 1$ and $R_T \approx 1/4i\omega \sqrt{t_1 t_2}$ are obtained for $\omega \gg (t_1 t_2)^{-1/2}$. See the discussion below Eq. (3.32) for the relaxation behavior in this ultimate regime.

Zhang et al. [11] measured a density change induced by boundary temperature oscillation in near-critical $^3$He, where the frequency was very low ($\omega/2\pi < 2$Hz) and the bulk viscosity was not important. However, they could measure in-phase and out-of-phase response in agreement with the original theory [3].

2. Mechanical oscillation

In the second example, the boundary wall at $x = 0$ is mechanically oscillated without heat input from outside. This is the case in the usual acoustic experiments using a piezoelectric transducer [12]. Let $u_w(\propto e^{i\omega t})$ be the applied displacement amplitude; then,

$$v = i\omega u_w$$

(3.19)

at $x = 0$ in Eq. (2.12). Assuming Eq. (2.30) and using Eq. (3.8) we obtain

$$\beta = -\left[1 + \frac{\kappa}{a_w} \frac{D}{i\omega} \right] \frac{\kappa R_T}{1 - \gamma DN^2/i\omega \rho u_w}$$

$$= (\gamma - 1)^{-1} \frac{\sqrt{i\omega}}{DX_v R_T \rho u_w}$$

(3.20)

where the first line is general and the second line is the approximation under the adiabatic condition Eq. (2.34). Since the response is proportional to $R_T$, resonance occurs as in the previous case of temperature oscillation.

In the low frequency case $\omega \ll c/L$, the interior density change is nearly homogeneous and

$$\delta \rho \approx 2\beta \approx \left[1 - \frac{1}{\sqrt{i\omega t_1 X_v + 1}}\right] \frac{\rho u_w}{L},$$

(3.21)

which is the counterpart of Eq. (3.14). As discussed below Eq. (3.16), $\sqrt{i\omega t_1 X_v}$ is large in the relatively high-frequency range $\omega \gg 1/t_1'$ under Eq. (3.17). Thus the interior density deviation behaves as

$$\delta \rho \approx \rho u_w/L \quad (1/t_1' \ll \omega \ll c/L)$$

$$\approx \sqrt{i\omega t_1 X_v \rho u_w}/L \quad (\omega \ll 1/t_1'),$$

(3.22)

under Eq. (3.17). The volume change mostly occurs in the bulk region for $1/t_1' \ll \omega \ll c/L$ and in the thermal diffusion layers for $\omega t_1' \ll 1$.

3. Heat flux oscillation

![FIG. 5: Absolute value of the response function $R_Q(\omega)$ in Eq. (3.24) vs $\omega L/\pi$ on a semi-logarithmic scale for $\epsilon = 10^{-3}$ (upper panel) and $10^{-4}$ (lower panel) applicable for CO$_2$ in a Cu cell with $L = 1$ cm.](image)

In the third example, we apply a heat flux $\dot{Q}_0 = -\lambda (dT/dx)_{x=0} \propto e^{i\omega t}$ at $x = 0$ assuming the boundary condition (2.30). It is convenient to introduce a dimensionless response function $R_Q$ by

$$\beta = \frac{\rho}{cT} \left( \frac{\partial T}{\partial p} \right) R_Q \dot{Q}_0$$

(3.23)
Under Eq.(3.17) we find that the boundary temperatures were held fixed for small with increasing $\delta$. From the discussion below Eq.(3.16), we find $\dot{Q} \approx 1/(2ikL + 2W)$ to obtain the counterpart of Eq.(3.15),

$$R_Q \approx \frac{X_v}{2\sqrt{i\omega t_1}}[1 + 2\sqrt{i\omega t_1 X_v}]^{-1}. \quad (3.25)$$

Under Eq.(3.17) we find that $R_Q \approx (1 + a_w^{-1})/2\sqrt{i\omega t_2}$ for $\omega \ll 1/t'_1$ and $R_Q \approx 1/4i\omega \sqrt{t_1 t_2}$ for $1/t'_1 \ll \omega \ll c/L$.

In this situation we may calculate the heat flux $Q_L$ at $x = L$. From Eq.(2.26) it is written as

$$\dot{Q}_L = \frac{(1 - Z)e^{-ikL}}{1 - Ze^{-2ikL}} \dot{Q}_0, \quad (3.26)$$

which vanishes for $Z = 1$ (or for $a_w = 0$) and becomes small with increasing $\delta B$. Near the resonance frequency $\pi \Delta c/L$, the ratio $Q_L/Q_0$ behaves as $(-1)^n W/[i(\omega L/c - n\pi) + n\pi \zeta/2\rho c^2 + W]$. The low frequency behavior for $\omega \ll c/L$ is given by

$$\dot{Q}_L = (1 + 2\sqrt{i\omega t_1 X_v})^{-1} \dot{Q}_0. \quad (3.27)$$

From the discussion below Eq.(3.16), we find $Q_L \approx \dot{Q}_0$ for $\omega t'_1 \ll 1$ under Eq.(3.17). That is, an applied heat flux passes through a near-critical fluid on the time scale of $t'_1$ under Eq.(3.17), due to the piston effect.

### C. Thermal and mechanical piston effects

#### 1. Boundary temperature change

In the original papers of the piston effect [3], the boundary temperatures at $x = 0$ and $L$ were both raised by a common small amount $T_1$ at $t = 0$. Subsequently, the boundary temperatures were held fixed for $t > 0$. In this paper, we examine the effects of finite $a_w^{-1}$ [14, 18] and large $\zeta$ [16, 17]. We suppose that the system was in equilibrium for $t < 0$ and the temperature in the wall region $x < 0$ was instantaneously raised by $T_1$ at $t = 0$ without external heat input in the other wall region $x > L$. The boundary conditions are then given by

$$\delta T(x, t) \rightarrow T_1 \text{ as } x \rightarrow -\infty \text{ and } \delta T(x, t) \rightarrow 0 \text{ as } x - L \rightarrow \infty \text{ for } t > 0.$$ 

All the deviations vanish for $t < 0$.

The Fourier transformation of the interior temperature deviation $\delta T(x, t)$ with respect to $t$ is given by Eq.(3.13) with $T_w = T_1 e^{i\omega t}/i\omega$ (since $\int_0^\infty dt e^{-i\omega t} = 1/i\omega$). The inverse Fourier transformation gives

$$\frac{\delta T(x, t)}{T_1} = \int \frac{d\omega e^{i\omega t}}{2\pi i\omega} (e^{-ikx} + Ze^{ikx - 2ikL}) R_T, \quad (3.28)$$

where the integration is in the range $[-\infty, \infty]$. Under Eq.(2.34), $W = W(\omega)$ and $R_T = R_T(\omega)$ are given by Eqs.(2.40) and (3.12), respectively. The integrand is analytic (without singularities) in the lower half plane $\text{Im} \omega < 0$ and hence the integral is nonvanishing only for $t > 0$.

In the time region $t \gg L/c$ we may neglect the space dependence of $\delta T(x, t)$ in the interior and use the simple expression (3.15) for $R_T(t)$. It then follows

$$\delta T(t) = T_1 \Psi(t)/2, \quad (3.29)$$

where we introduce the dimensionless relaxation function $\Psi(t)$. Its Fourier transformation reads

$$\int_0^\infty dt e^{-i\omega t} \Psi(t) = \frac{1}{i\omega(\sqrt{i\omega t_1 X_v} + 1)}. \quad (3.30)$$

The inverse Fourier transformation of the right hand side of Eq.(3.30) may be transformed into an integral along the positive imaginary axis $\text{Im} \omega > 0$. With $X_v$ being defined by Eq.(2.41), we generally find $\Psi(t) > 0$ for $t > 0$, $\Psi(t) \approx t/\sqrt{t_1 B}$ as $t \rightarrow 0$, and $\Psi(t) = 1 - (t_1/\pi t)^{1/2} + \cdots$ as $t \rightarrow \infty$. In particular, not very close to the critical point, we may neglect the bulk viscosity and take the limit $t_B \rightarrow 0$; then, $X_v \rightarrow 1 + a_w^{-1}$ and $\Psi(t) \rightarrow \Psi_0(s)$.
where $\Psi_0(s)$ is a universal function of $s = t/t_1'$ expressed as

$$
\Psi_0(s) = 1 - \int_0^\infty \frac{du}{\pi \sqrt{u}} \frac{e^{-us}}{1 + u} = 1 - e^s \text{erf}(\sqrt{s}),
$$

(3.31)

where $\text{erf}$ is the complementary error function and $\Psi_0 \approx 2(s/\pi)^{1/2}$ for $s \ll 1$ and $\Psi_0 \approx 1 - 2(\pi s)^{-1/2}$ for $s \gg 1$.

In Fig. 6, we display $\Psi(t)$ as a function of $t/t_1'$ at $\epsilon = 10^{-3}$, $10^{-4}$, and $10^{-5}$ for CO$_2$ in a Cu cell with $L = 1$ cm. For $\epsilon = 10^{-3}$ we can see $\Psi(t) \approx \Psi_0(t/t_1')$, where $t_B/t_1' \approx 0.02$ from Table 1. The discussion below Eq.(3.16) indicates that $\Psi(t)$ approaches unity on the time scale of $t_1'$ as long as Eq.(3.17) is satisfied. This is the case even for $\epsilon = 10^{-5}$, where $t_B/t_1' \approx 21$ from Table 1. In fact, if $t_B/t_1' \gg 1$ and $a_w \ll 1$, we may set $\sqrt{i\omega_{1}X_c} \approx i\omega_{1}t_B + a_w^{-1}\sqrt{i\omega_{1}}$, where the second term is relevant in $\Psi(t)$ under Eq.(3.17), again leading to $\Psi(t) \approx \Psi_0(t/t_1')$ for $t > a_w^{-2}t_B$. However, the reverse condition of Eq.(3.17) holds extremely close to the critical point, where $R_T \approx 1/[\sqrt{i\omega_{1}}t_B + 1]$ holds yielding [17]

$$
\Psi(t) \approx 1 - \exp(-t/\sqrt{i\omega_{1}t_B}),
$$

(3.32)

The new relaxation time $\sqrt{i\omega_{1}t_B}$ here grows as $\sqrt{i\omega_{1}t_B} \approx 1.0 \times 10^{-4} e^{-0.64t}$ for near-critical CO$_2$.

Assuming the isothermal boundary ($a_w = \infty$), Carles and Dadzic examined the bulk viscosity effect in the thermal equilibration [17]. Their relaxation function is obtained if we set $X_c = (1 + i\omega t_B)^{1/2}$ in Eq.(3.30). Then a new viscous regime appears for $t_1 \gg t_B$ with $\Psi(t)$ being given by Eq.(3.32), while the usual piston regime is encountered for $t_1 \ll t_B$. For CO$_2$ we have $t_B/t_1 \cong 2.7 \times 10^{-20} L^{-2} \approx 4.6$ so $t_1 = t_B$ holds at $\epsilon \approx 0.6 \times 10^{-4}$ with $L = 1$ cm. In our calculations based on Eq.(3.17), the different predictions have arisen from the reduced temperature dependence of $a_w$ or the crossover of the boundary condition into the insulating one.

2. Volume change

We suppose a volume change by moving the boundary wall at $x = 0$ by a small length $u_1$ instantaneously at $t = 0$ [1]. We assume the thermal boundary conditions (2.28) and (2.30) at $x = 0$ and $L$. As in Eq.(3.26), the complete interior density deviation is the inverse Fourier transformation of Eq.(3.8), where $\beta$ is given by Eq.(3.20) with $u_w = u_1 e^{-i\omega t}/i\omega$.

Here we are interested in the late stage $t \gg L/c$, where the interior deviations depend only on $t$. The inverse Fourier transformation of Eq.(3.21) gives the interior deviations,

$$
\delta\rho(t) = b_s \delta T(t) = [1 - \Psi(t)] \rho u_1/L,
$$

(3.33)

where $\Psi(t)$ defined by Eq.(3.30) represents the effect of the thermal diffusion layers at $x = 0$ and $L$. The above form with $\Psi = \Psi_0$ was derived in Ref.[1]. If $u_1 > 0$, the interior is adiabatically heated by $b_s^{-1} \rho u_1/L$ on the acoustic time scale $L/c$ after the volume change, while the boundary wall temperature is almost unchanged. Subsequently, the thermal diffusion layers become effective as reverse pistons and the interior temperature deviation decays as $(t_1'/t)^{1/2}$.

The reverse piston effect itself generally occurs on the time scale of $t_1'$ after a near-critical fluid was adiabatically heated or cooled. Miura et al. observed such a process after a pulse-like heat input (see Fig. 2 in Ref.[20]).

D. Emission of sound

We examine sound emission at the boundary at $x = 0$. We neglect the incoming wave reflected at the other end $x = L$ and consider the semi-infinite limit $L \to \infty$.

The problem is simple in the case of boundary wall motion. An emitted sound propagates with the velocity $c$ and integration of the continuity equation gives the density deviation,

$$
\delta \rho(x, t) \cong \rho v_1 (t - x/c)/c,
$$

(3.34)

where $v_1(t)$ is the velocity of the boundary. The localized part of the density deviation (the term proportional to $a$ in Eq.(2.20)) should be small when differentiated with respect to time. In fact, under the adiabatic condition (3.24), Eqs.(3.12) and (3.20) lead to

$$
\beta \cong \frac{1 + Z}{2(1 + \Delta_w/c)} i\omega \rho u_w,
$$

(3.35)

for $e^{-2ikL} \to 0$. If we set $1 + \Delta_w \cong 1$ and $Z \cong 1$, the above relation becomes $\beta \cong i\omega \rho u_w/c$, leading to Eq.(3.34). Here $i\omega \rho u_w$ is the Fourier transformation of $v_1(t)$ multiplied by $e^{i\omega t}$. Thus Eq.(3.34) holds on time scales longer than $t_c$ (even when the time scale of $v_1(t)$ is shorter than $t_B$).

A sound is also emitted when a time-dependent heat flux $Q_0(t)$ is supplied at the boundary at $x = 0$. From Eq.(3.8) the Fourier transformation of the interior density deviation $\delta \rho(x, t)$ is of the form $\beta e^{i\kappa x}$ with $\beta$ being given by Eq.(3.23). Under the adiabatic condition Eq.(3.24) we may use the second line of Eq.(3.24) to find the convolution relation,

$$
\delta \rho(x, t) = \frac{\rho}{cT} \left( \frac{\partial T}{\partial \rho} \right)_x - \int_{-\infty}^t d\tau \Phi(x, t - \tau) Q_0(\tau).
$$

(3.36)

The memory function $\Phi(x, t)$ is defined for $t > 0$ as

$$
\Phi(x, t) = \int \frac{d\omega}{2\pi} e^{i\omega t - \omega^2 x} (1 + \Delta_w)^{-3/2},
$$

(3.37)

where $\Delta_w = i\omega \zeta / \rho c^2 = i\omega R_B t_c$ with $R_B \cong 0.03$. The time integration of this function is normalized as $\int_0^\infty d\Phi(x, t) = 1$. From the integration in the region
\[ \omega < t_c^{-1} \] we obtain the long-time behavior \( \Phi(0, t) \cong (4t/\pi \xi^3)_{\xi}^{1/2} e^{-t/t_c} \) with \( t_c \equiv R_B t_c \) at \( x = 0 \). Since this relaxation is rapid, we may set \( \Phi(0, t) \cong \delta(t) \) (\( \delta \)-function) at \( x = 0 \) on time scales longer than \( t_c \) or when \( Q_0(t) \) varies slower than \( t_c \). Furthermore, if the distance \( x \) is not large such that the bulk damping is negligible in the region \( 0 < x < L \), we may set \( \Phi(x, t) \cong \delta(t - x/c) \) to find the simple formula for the emitted sound,

\[
\delta p(x, t) = \frac{\rho}{c^2 T} \left( \frac{\partial T}{\partial p} \right)_s Q_0(t - x/c), \tag{3.38}
\]

as the counterpart of Eq.(3.34). On the other hand, use of Eq.(B6) for \( \Delta \nu \) gives the short-time behavior,

\[
\Phi(0, t) = (\dot{\alpha}/2\nu)(t/t_c)^{\dot{\alpha}/2\nu}/t, \tag{3.39}
\]
valid in the time region \( t \ll t_c \) with \( \dot{\alpha}/2\nu \cong 0.088 \) (see Appendix B). This behavior is detectable only for an increase of \( Q_0 \) within a time shorter than \( t_c \).

Miura et al. applied a stepwise heat flux with \( \dot{Q}_0 = 0.183 \times 10^7 \) to find a stepwise outgoing sound with \( \delta p/\rho \cong 2.2 \times 10^{-7} \) for CO\(_2\), where \( Q_0 \) is in cgs units (erg/cm\(^2\)sec) \[20\]. Our theoretical expression (3.38) becomes \( \delta p/\rho = 1.38 \times 10^{-13} \dot{Q}_0 \) with the aid of \( (\partial T/\partial p)_s \cong T_c/6.98 \rho_c \) for CO\(_2\) \[22\]. For their experimental \( \dot{Q}_0 \) our theory gives \( \delta p/\rho = 2.55 \times 10^{-7} \) in fair agreement with the observed density change. Furthermore, they could generate sound pulses with duration of order 10\( \mu \)sec by applying short-time heat input. They were interested in the adiabatically increased energy \( E_{\text{ad}} = p \int dx \delta p(x, t)/\rho \) in the pulse region per unit area. Here Eq.(3.38) yields \[3\]

\[
E_{\text{ad}} = \frac{\rho}{T} \left( \frac{\partial T}{\partial p} \right)_s Q, \tag{3.40}
\]

where \( Q = \int dt \dot{Q}_0(t) \) is the total heat supplied. The ratio \( E_{\text{ad}}/Q \) represents the efficiency of transforming applied heat to mechanical work. Theoretically, it is given by \( (\partial T/\partial p)_s/\rho T \) as in Eq.(3.40) and is equal to 1/6.98 = 0.14 for near-critical CO\(_2\) \[22\]. The measured values of the ratio \( E_{\text{ad}}/Q \) were in the range 0.11 – 0.12 again in fair agreement with our theory.

### E. Reflection of sound

Reflection of plane wave sounds is discussed for an isothermal boundary in the textbook of Landau-Lifshitz \[34\]. Miura et al. \[20\] observed reflected pulses passing through a detector in the cell. Their shapes gradually flattened after many traversals within the cell, resulting in the interior temperature homogenization. At present, it is not clear how to understand their data. Here, as a first step, we will derive some fundamental relations on sound reflection.

We consider a pulse approaching to the boundary at \( x = 0 \) in the semi-infinite limit \( L \to \infty \). Reflection takes place upon its encounter with the wall. The density deviations of the incoming and outgoing pulses are obtained as the inverse Fourier transformation of Eq.(2.20). Neglecting the bulk damping in the neighborhood of the boundary, we may express them as \( \rho_i(t + x/c) \) and \( \rho_o(t - x/c) \), respectively. Using \( \alpha(\omega) = e^{i\omega t} \int d\tau e^{-i\omega \tau} \rho_i(\tau) \) and \( \beta = Z\alpha \), we obtain

\[
\rho_o(t) = \int \frac{d\omega}{2\pi} \int dt' Z(\omega)e^{i\omega(t'-t)} \rho_i(t'). \tag{3.41}
\]

The interior density deviation is the sum \( \delta p(x, t) = \rho_i(t + x/c) + \rho_o(t - x/c) \). Since \( Z(0) = 1 \), the excess mass is invariant upon reflection as

\[
\Delta M = \int dt \rho_i(t) = \int dt \rho_o(t). \tag{3.42}
\]

This relation holds if we integrate a long tail of the reflected pulse \( \rho_o(t) \) at large \( t \) (see Eq.(3.47)).

If \( \rho_i(t) \) changes much slower than \( t_c \), we may set \( Z = 1 - 2W \) with \( W = (\gamma - 1)\sqrt{\Delta_T}/X_c \) from Eq.(2.40).

In this approximation we may rewrite Eq.(3.40) in the following convolution form,

\[
\rho_o(t) = \rho_i(t) - \int_0^\infty dt' \chi(\tau) \rho_i(t - \tau - \tau) \tag{3.43}
\]

From Eq.(3.41) the function \( \chi(t) \) is the inverse Fourier transformation of \( (1 - Z)/i\omega \cong 2W/i\omega \). Some calculations (in the complex \( \omega \) plane) give \( \chi(t) \) in the integral form,

\[
\chi(t) = \varepsilon_r \int_0^\infty \frac{d\Omega}{\pi \sqrt{\Omega}} \Re \left[ \frac{e^{-i\Omega s}}{a_w^{-1} + \Omega^{-1}} \right], \tag{3.44}
\]

where \( s = t/t_B \) is the scaled time, \( \Re[\cdot] \) denotes taking the real part, and \( \sqrt{1 - \Omega} = i\sqrt{\Omega - 1} \) for \( \Omega > 1 \). The dimensionless parameter \( \varepsilon_r \) is defined by

\[
\varepsilon_r = 2(\gamma - 1)\sqrt{D/c^2 t_B} \tag{3.45}
\]

which decreases near the critical point as \( \varepsilon_r \cong 4.3e^{0.75} \) for CO\(_2\). The function \( \chi(t) \) depends only on \( s \) and \( a_w \). For \( a_w \gg 1 \) we have \( \chi(t)/\varepsilon_r \equiv e^{-s/2I_0(s/2)} \) with \( I_0 \) being the modified Bessel function, while for \( a_w \ll 1 \) we have \( \chi(t)/\varepsilon_r \equiv 1 - \Phi_0(s/a_w^2) \) with \( \Phi_0 \) being defined in Eq.(3.31). Thus \( \chi(t) \) changes on the scale of \( t_B(1 + a_w^{-2})^{-1} \) and its limiting behaviors are as follows:

\[
\frac{\chi(t)}{\varepsilon_r} = (1 + a_w^{-2})^{-1}(\pi s)^{-1/2} + \cdots \quad (s \to \infty),
\]

\[
= 1 - 2a_w^{-1}(s/\pi)^{1/2} + \cdots \quad (s \to 0). \tag{3.46}
\]

In addition, the second term of Eq.(3.43) representing the distortion is negative (positive) when \( \rho_i(t) \) is increasing (decreasing). This initial drop is because of heating and expansion of the pulse at the boundary.
From the first line we obtain \( \chi(t) \propto -(t_2/\pi)^{1/2}t^{-3/2} \) for \( t \gg t'_B \). (i) Let \( \rho_1(t) \) is peaked in the region \( |t| < t_0 \); then, for \( t \gg t'_B \) and \( t_0 \), the first line of Eq.(3.43) gives a long-time tail of the reflected wave,

\[
[\rho(t)]_{\text{tail}} = \Delta M (t_2/\pi)^{1/2}t^{-3/2}
\]  \hspace{1cm} (3.47)

where \( \Delta M \) is defined by Eq.(3.42). If \( t_0 > t'_B \), the total mass behind the peak is given by the time integral of the tail Eq.(3.47) in the region \( [t_0, \infty] \). Thus the mass fraction behind the peak is \( (4t_2/\pi t_0)^{1/2} \). For CO\(_2\) this quantity is estimated as \( 10^{-7}\epsilon - 0.75t_0^{-1/2} \) with \( t_0 \) in sec for \( a_w \gg 1 \). (ii) As another example, we consider a stepwise change, where \( \rho_1(t) \) is equal to 0 for \( t < 0 \) and to a constant \( \rho_1 \) for \( t > t_0 \) with \( t_0 \) the transient time. Then, for \( t \gg t'_B \) and \( t_0 \), the second line of Eq.(3.43) gives a longer tail,

\[
[\rho(t)]_{\text{tail}} = \rho_1 (4t_2/\pi)^{1/2}t^{-1/2}.
\]  \hspace{1cm} (3.48)

The bulk viscosity does not appear in these tails.

When \( \rho_1(t) \) changes much slower than \( t'_B \), only the long time behavior of \( \chi(t) \) is relevant in \( \rho_0(t) \). From Eq.(3.43) we find the following convolution relations,

\[
\rho_0(t) = \rho_1(t) + \sqrt{\frac{t_2}{\pi}} \int_0^\infty \frac{d\tau}{\tau^{3/2}} [\rho_1(t-\tau) - \rho_1(t)]
\]

\[
= \rho_1(t) - \sqrt{\frac{4t_2}{\pi}} \int_0^\infty \frac{d\tau}{\sqrt{\tau}} \dot{\rho}_1(t-\tau),
\]  \hspace{1cm} (3.49)

from which the long-time tails (3.47) and (3.48) readily follow. The above expressions contain only \( t_2 \) in Eq.(2.43) and not \( t_B \). They are widely applicable far from the critical point (where \( t_B \) becomes short). With decreasing \( \epsilon \) for the isothermal boundary (\( a_w > 1 \), \( t_2 \) grows and the distortion of the reflected pulse increases as long as the pulse width is longer than \( t_B \). However, if the pulse width is shorter than \( t'_B \), the distortion decreases on approaching the critical point since \( \varepsilon_r \) in Eq.(3.45) decreases.

As a simple illustration, let us consider a Gaussian pulse \( \rho_0(t) = \rho_1 \exp(-t^2/2t'_B^2) \), where \( \rho_1 \) is the pulse height and \( t_B \) is the pulse width. Since its Fourier transformation is \( (2\pi)^{1/2} \rho_1 t_B e^{-\omega^2 t'_B^2/2} \), we may readily calculate \( \rho_0(t) \). In Fig. 7, we plot the normalized pulse deformation defined by

\[
F_C(t) = [\rho_0(t) - \rho_0(t)]/(\rho_1 \sqrt{t_B/t_0}).
\]  \hspace{1cm} (3.50)

The curve (a) is for the limiting case \( t_B/t_0 \to 0 \) and \( a_w = \infty \), while \( t_B/t_0 = 10 \) and \( a_w = 0.63 \) in (b), and \( t_B/t_0 = 50 \) and \( a_w = 0.63 \) in (c). In Table 1 we have \( a_w = 0.63 \) and \( t_B = 1.9 \) msec at \( \epsilon = 10^{-4} \) for CO\(_2\) in a Cu cell, where pulses with \( t_B < t_B \) are well possible [21]. We recognize that the distortion is negative for \( t < t_0 \) and is positive for \( t > t_0 \) (in accord with the comment below Eq.(3.43)) and that the distortion is decreased as \( t_B/t_0 \) is increased or for shorter pulses due to the bulk viscosity growth.

IV. SUMMARY AND REMARKS

In summary, we have examined various thermoacoustic effects in one-component supercritical fluids in a one-dimensional geometry. We summarize our main results.

(i) In the linear hydrodynamics, sound modes and thermal diffusion modes are both present as in Eq.(2.20), depending on given boundary conditions. The latter modes can be absent only for the insulating boundary condition \( a_w = 0 \). The calculations are straightforward and the final expressions are much simplified under the adiabatic condition (2.34) or for low frequencies \( \omega \ll c^2/\gamma D \). It is remarkable that the bulk viscosity \( \zeta \) appears in the combination \( \omega \gamma \zeta/\rho c^2 = \omega t_B \) as first pointed out by Carless [16, 17]. The resultant characteristic time \( t_B \) grows as \( \epsilon^{-3.0} \), while the life time of the critical fluctuations \( \xi \) grows as \( \epsilon^{-1.9} \).

(ii) We have introduced the reflection factor \( Z(\omega) \) as the ratio between outgoing and incoming sounds. Using \( Z \) or \( W = (1-Z)/(1+Z) \) we have examined the acoustic eigenmodes, the response of the fluid to applied oscillation of the boundary temperature, the boundary heat flux, and the boundary position. To these thermal and mechanical perturbations, resonance is induced when the frequency of the perturbation is close to one of the eigenfrequencies, while nearly uniform adiabatic changes are caused in the interior at much lower frequencies owing to the piston effect.

(iii) We have also examined the response to a stepwise change of the boundary temperature and the boundary position. The relaxation time is given by the modified piston time \( t'_1 \) in Eq.(3.16) first introduced by Ferrell and Hao [14]. It is equal to the original piston time \( t_1 \) in Eq.(1.1) for the isothermal boundary \( a_w \gg 1 \) and to
The volume expansion of the thermal diffusion layers is
ary per unit area in the one dimensional geometry.
and the pressure change

δQ

for viscous regime predicted by Carlès and Dadzie emerges
bulk viscosity effect in the thermal diffusion layers as
ζ of the bulk viscosity
becomes increasing stronger. The bulk viscosity effect
in the thermal diffusion layer is thus masked by its
enhanced effect in the bulk.

(v) For CO₂ in a Cu cell, the boundary becomes
thermally insulating for ϵ ∝ 10⁻⁴. This suppresses the
bulk viscosity effect in the thermal diffusion layers as
long as Eq.(3.17) holds or for ϵ > 10⁻⁶. In this case, the
viscous regime predicted by Carlès and Dadzie emerges
for ϵ < 10⁻⁶ [14, 17]. To increase this crossover reduced
temperature, the cell length L needs to be shorter. For
the wall materials in Ref.[18], this crossover occurs much
closer to the critical point [15].

(vi) We have also examined sound emission and reflection
at the boundary, which are elementary hydrodynamic
processes but seem to have not been well examined [34]. For emission, the formulas (3.34) and (3.38)
are valid for a mechanical piston and a thermal heat
input on time scales longer than t_c. For reflection,
Eq.(3.43) with Eq.(3.44) holds on time scales longer
than t_c. The formula (3.49) is the classical one valid on
time scales much longer than t_B, where the distortion
of the outgoing pulse increases on approaching the
critical point. For pulses shorter than t_B, the distortion
of the outgoing pulse is decreased as can be seen in Fig. 7.

In this paper, we have treated near-critical fluids in
one phase states. However, more challenging are hydro-
dynamic effects in in two phase states, where latent heat
transport, wetting dynamics, and Marangoni convection
come into play in addition to the piston effect [30, 37, 38].

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Appendix A: Simple theory of the piston effect

Here we give a simple derivation of t_1 in Eq.(1.1). Let
us apply a small heat δQ to a fluid from the bound-
ary per unit area in the one dimensional geometry. The
volume expansion of the thermal diffusion layers is
given by (δT/δp)_AδQ(t)/T, where A is the area of the
heater surface and use is made of the Maxwell relation
(δρ/δs)_p = (δT/δp)_s. The interior density change
and the pressure change δp are nearly homogeneous in
the interior and are given by

δρ = δp/ε² = ρ/T (δT/δp)_s δQ/L. 

(A1)

where L = V/A is the cell length. The interior tem-
perature deviation is caused adiabatically as δT =
(δT/δp)_p dp and is written as

δT = (γ − 1)δQ/C_p L, 

(A2)

where C_p = ρ(T(δs/δT)_p is the isobaric specific heat and
use is made of Eq.(2.9). If the boundary temperature is
raised by

δt = 0, we have δQ ∼ C_p(δt)T_1 in the
early stage, where δt(t) = √Bt is the thickness of the
thermal diffusion layer. If we set δT = T_1/2, Eq.(1.1) is
reproduced.

The relation (A1) also follows from our formula
Eq.(3.38). Let the heat input rate Q(t) from the bound-
ary to the fluid change slowly compared to the acoustic
time t_a = L/c. We suppose a time interval with width
δt ≫ t_a, in which Q(t) is almost unchanged. Since δt/t_a
is the traversal number much larger than unity, the adia-
batic pressure and density increases in the interior region
are given by

δρ = δp/ε² = δt/ε(t_a)C_p T δQ/L, 

(A3)

as a result of superposition of many steps. In terms
of the incremental heat supply δQ = Qδt we reproduce
Eq.(A1).

Appendix B: Summary of critical behavior

Let a one-component fluid be on the critical isochore
(ρ = ρ_c) with small positive ϵ = T/T_c − 1 near the gas-
liquid critical point. The physical parameters used in
the table 1 and the figures are given below. Hereafter
ν(≈ 0.63), γ(≈ 1.24), and α(≈ 0.10) are the usual critical
exponents. Data of near-critical CO₂ can be found in
Refs. 30, 35.

Our hydrodynamic description is valid when the spa-
tial scale under investigation is longer than the correla-
tion length ξ = ξ_0 e^−α, where ξ_0 = 1.5Å for CO₂. The
constant-volume specific heat C_V = ρ(T(δs/δT)_p and the
isobaric specific heat C_p = ρ(T(δs/δT)_p are expressed as

C_V = A_V[ε^−α + B], C_p = A_p ε^−γ. 

(B1)

For CO₂ on the critical isochore, the coefficients are given
by A_V = 26.3k_Bn^∗, B = 0.9, and A_p = 2.58k_Bn^∗, where
n^∗ = p_c/k_BT_c ≈ 1.76 × 10^{21} cm^{-3}. The specific-heat ratio
γ grows strongly as γ_0 e^−α if the background (α B) is
neglected, where γ_0 = 0.1 for CO₂. The sound velocity
and the constant-volume specific heat are weakly singular
as c^2 ∝ ε^α (if the background is neglected [1]). We have
set c = 2.3 × 10^{4} cm sec^{-1} for CO₂ [26].
The thermal conductivity $\lambda$ grows such that the thermal diffusion constant $D$ behaves as

$$D = \lambda / C_p = k_B T / 6\pi \xi = D_0 e^{\xi^0}, \quad (B2)$$

where $D_0 = 4.0 \times 10^{-4} \text{cm}^2\text{sec}^{-1}$ for CO$_2$. Thus $\lambda \propto e^{\xi^0}$. The relaxation time of the critical fluctuations with size $\xi$ increases as

$$t_\xi = \xi^2 / D = t_0 e^{-3\xi^0}, \quad (B3)$$

where $t_0 = 0.56 \times 10^{-12}\text{sec}$ for CO$_2$. The shear viscosity $\eta$ is only weakly singular and may be treated as a constant independent of $\epsilon$ and $\omega$. However, the zero-frequency bulk viscosity $\zeta$ grows very strongly as

$$\zeta = \rho c^2 R_B t_\xi, \quad (B4)$$

where $R_B$ is a universal number estimated to be about 0.03 [1, 32]. For CO$_2$, $\zeta / \rho \approx 0.9 \times 10^{-5} \epsilon - 2^{-2/3} \text{cm}^2\text{sec}^{-1}$, so $\zeta^0 / \rho D = \Delta_\nu / \Delta T \approx 0.02 e^{-2^{1/3} \epsilon}$ (see Eqs.(3.13) and (3.14)). In the high frequency regime $\omega t_\xi \gg 1$, the complex sound velocity $c^*(\omega)$ in Eq.(2.39) becomes asymptotically independent of $\epsilon$ [31, 33, 34]. Thus,

$$c^*(\omega) \equiv c(i \omega t_\xi)^{\hat{\alpha}/6\hat{\nu}}. \quad (B5)$$

Since the exponent $\hat{\alpha}/6\hat{\nu}$ is small, we may set $\Delta_\nu = i \omega c^2 / \rho \approx (\hat{\alpha}/3\hat{\nu}) \ln(i \omega t_\xi)$. Thus, in this high frequency regime, $\text{Im} \Delta_\nu$ tends to the following universal number,

$$\text{Im} \Delta_\nu = \pi \hat{\alpha}/6\hat{\nu} \approx 0.27 \times 2/\pi \quad (B6)$$

In the high frequency regime $\omega t_\xi > 1$, $\Delta_\nu$ remains to be as a small quantity and the frequency-dependent bulk viscosity defined by $\zeta(\omega) \equiv \rho c^2 \Delta_\nu / \omega$ decays roughly as $1/\omega$ with increasing $\omega$.

Furthermore, in our thermoacoustic problems, we have introduced the time $t_B$ in Eq.(2.37), which behaves as

$$t_B = t_0^0 e^{-3\xi^0 + \eta^0}, \quad (B7)$$

where $t_0^0 = 1.7 \times 10^{-12}\text{sec}$ for CO$_2$. The effusivity ratio $a_w$ in Eq.(2.29) decreases as

$$a_w = a_w^0 e^{-\xi^0 / 2}. \quad (B8)$$

For $a_w^0 \gg 1$ the boundary wall crosses over from an isothermal one to an thermally insulating one on approaching the critical point. For example, between Cu and CO$_2$, we have $a_w^0 = 3 \times 10^9 \text{cm}^2\text{sec}^{-1/2}$, where $a_w < 1$ is reached for $\epsilon < 1.6 \times 10^{-4}$. The $a_w^0$ was smaller for the walls used in Ref.[15, 18].

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