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High-speed observation of the piston effect near the gas-liquid critical point

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We investigated adiabatic changes in a near-critical fluid on acoustic time scales using an ultrasensitive interferometer. A sound emitted by very weak continuous heating caused a stepwise adiabatic change at its front with a density change of order 10^{-7} g/cm^3 and a temperature change of order 10^{-5} K. Very small heat inputs at a heater produced short acoustic pulses with width of order 10 μs, which were broadened as they moved through the cell and interacted with the boundaries. The pulse broadening became enhanced near the critical point. We also examined theoretically how sounds are emitted from a heater and how applied heat is transformed into mechanical work. Our predictions agree well with our data.

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Adiabatic changes are ubiquitous in nature and fundamental in thermodynamics. In confined fluids, they are usually caused by a mechanical piston, but they can also be achieved by a heat input through a boundary. In the second case, the fluid contained within the thermal diffusion layer at the boundary expands and plays the role of a *thermal piston* [1–11]. This method is efficient near the gas-liquid critical point, where the thermal expansion of the layer grows [2,3]. Previous experiments on the thermal piston effect [1,4–6] have detected temperature and density changes on slow time scales ($\sim$1 s). However, a mechanical or thermal piston should emit sounds, which cause adiabatic compression and heating in the interior as they traverse the container. This acoustic process has been found by solving hydrodynamic equations of compressible fluids [5,7,8], but it has never been observed experimentally because it takes place on the fast time scale of $t_a=L/c$ ($\sim 10^{-4}$ s for $L=1$ cm). Here L is the cell length and c is the sound velocity. Though it is a general phenomenon, there is still little understanding of this process. The aim of this Rapid Communication is to report ultrasensitive, high-speed observation of adiabatic changes.

Before presenting our acoustics research, we elucidate the slow time evolution after a slight increase of the boundary temperature by T_1 at time $t=0$. An acoustic mass flow is then induced from the layer to the interior and the mass change in the layer is $M_b=(\partial\rho/\partial T)_p T_1 \ell(t)$ per unit area, where the layer expansion is nearly isobaric [2]. Here $\ell(t)=(Dt)^{1/2}$ is the layer thickness at time t with D being the thermal diffusion constant. It is much shorter than L for $t \ll L^2/D$. The interior region is adiabatically compressed with density change $-M_b/L$ from mass conservation, leading to the early stage increase of the interior temperature,

$$T_{\rm in} = -(\partial T/\partial\rho)_s M_b/L = T_1(\gamma-1)\,\ell\,(t)/L, \tag{1}$$

where $\gamma=C_p/C_V$ is the specific-heat ratio growing near the critical point [12]. Use has been made of the thermodynamic identity $-(\partial T/\partial\rho)_s/(\partial T/\partial\rho)_p=\gamma-1$. We set $T_{\rm in}=T_1/2$ to find the relaxation (piston) time [13]

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

which is much shorter than the diffusion time $L^2/4D$ by $(\gamma-1)^{-2}$. Thus, on approaching criticality, thermal equilibration occurs increasingly faster at fixed volume, despite the fact that D tends to zero [11,14].

We developed an experimental setup to measure density changes of order 10^{-8} g/cm^3 on a time scale of 1 μs in a cell filled with CO_2 on the critical isochore close to the critical point $T_c=304.12$ K. It is displayed in Fig. 1, where the fluid temperature was controlled with precision of ± 1 mK using a Pt thermometer and a four-wire resistance bridge [15]. The upper and lower plates have area 1 cm^2 and are made of Cu

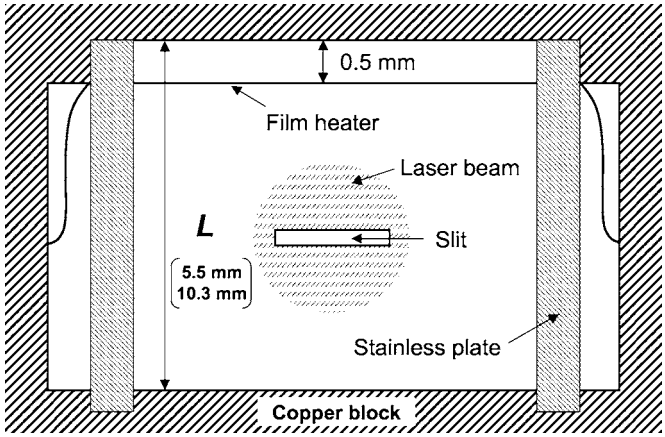


FIG. 1. Front view of experimental setup. Cell length L is either 10.3 mm or 5.5 mm. A dc current is sent to a film heater placed 0.5 mm below the top boundary. Light passing through the slit of width 0.25 mm is used to detect small density changes using interferometry. Gravity is downward.

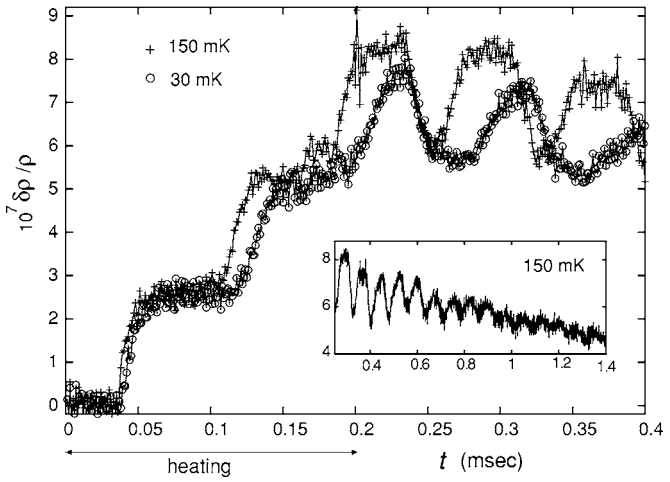


FIG. 2. Normalized density change $\delta\rho(t)/\rho$ at the cell center in the time region $0<t<0.4$ ms for $T-T_c=150$ and 30 mK, produced by continuous heating in $0<t<0.2$ ms in a cell of $L=1.03$ cm. Inset: Long-time behavior for $T-T_c=150$ mK in the time region $0.2<t<1.4$ ms.

with high thermal conductivity $\lambda_{Cu}/k_B=2.8\times10^{23}$/cm sc. The cell length L was either 1.03 cm or 5.5 mm. A thin NiCr-foil heater with thickness 3 μm was placed $d=0.5$ mm below the top plate. The heat capacity of the foil is very small and the generated heat was almost all released to the fluid [16]. The sidewalls are made of stainless steel, whose thermal conductivity is 3.9% that of Cu. We thus neglect heat flow to the sidewalls. A laser beam was sent to the cell center and a Twymann-Green interferometer was used to detect small density changes in a slit region of width 0.25 mm. Individual signals were very noisy and the data points in the following figures are the averages over 418 heat pulses successively generated in 2 s. Using sound pulses we first measured the sound velocity c. For both $L=1.03$ cm and 5.5 mm, our data on c closely agree with previous ones in the range $T/T_c-1\gtrsim10^{-4}$ [17], but are independent of $T-T_c$ closer to the critical point because of the frequency (inherent to short pulses) and/or gravity effect [18].

In Fig. 2 we display acoustic density variations $\delta\rho(t)$ at the cell center $x=L/2$ in a cell of $L=1.03$ cm at $T-T_c=150$ and 30 mK, where $c\cong140$ and 123 m/s, respectively. Continuous heating was applied in the time region $0<t<0.2$ ms and the supplied heat to the fluid was 367 erg. Figure 3 illustrates spatial profiles of the density deviation $\delta\rho(x,t)$, where the behavior inside the thermal diffusion

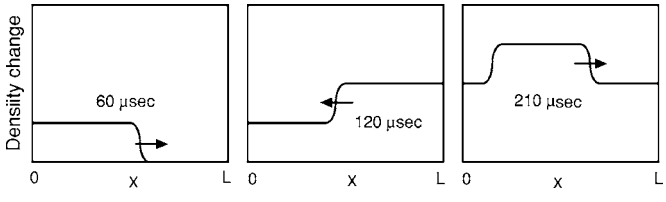
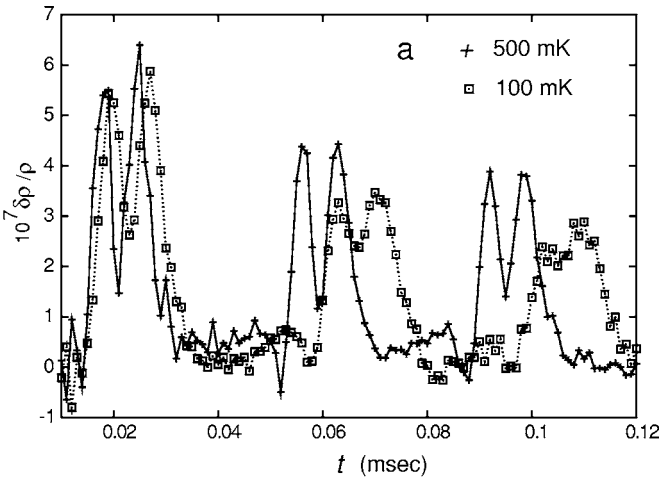


FIG. 3. Schematic profiles of density deviations moving at the sound velocity at $T-T_c=150$ mK. Heating is stopped at $t=200$ μs, which results in a trapezoidal pulse (right).



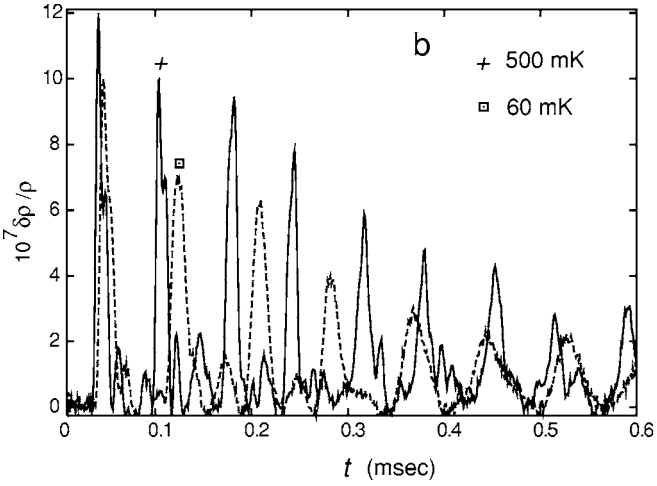


FIG. 4. $\delta\rho(t)/\rho$ at the cell center, where (a) $0<t<0.12$ ms in a cell of $L=5.5$ mm and (b) $0<t<0.6$ ms in a cell of $L=1.03$ cm. Heating time was (a) 4.5 and (b) 7 μs.

layers is not shown. The layer thickness is so thin ($\sim2.6\times10^{-5}$ cm at $t=0.2$ ms for $T-T_c=150$ mK). Steplike sounds were emitted on both sides of the heater placed 0.5 mm below the top. The one moving upward was reflected at the top at $t\cong d/c\sim5$ μs. The reflected one and the one directly going downward merged for $t>2d/c\sim10$ μs. The sound thus formed swept through the cell, producing a stepwise increase in the density, which was observed at $t\cong L/2c$, $3L/2c$, and $5L/2c$ at the center. After the switching off at $t=0.2$ ms, a trapezoidal pulse appeared and moved back and forth in the cell. Comparison of the two curves in Fig. 2 indicates that the trapezoidal part decreases when the heating time is close to a multiple of $2L/c$. The interior temperature was adiabatically increased by $\delta T=(\partial T/\partial\rho)_s\delta\rho$ and a heat flow was induced through the boundaries. In the present case $\delta T\sim0.2T\delta\rho/\rho\sim10^{-5}$ K. The inset of Fig. 2 shows the subsequent relaxation at the center for $T-T_c=150$ mK on a rapid time scale of 1 ms [$\sim t_1$ in Eq. (2)]. This speeding up is a (reverse) piston effect, caused by contraction of the thermal diffusion layers.

In Fig. 4 we display $\delta\rho(t)$ at the cell center after very short heating at $T-T_c=500$ and 100 mK in a cell of $L=5.5$ mm in (a) and at $T-T_c=500$ and 60 mK in the cell of

1.03 cm in (b). A dc electric current passed through the heater with duration time 4.5 μs in (a) and 7 μs in (b), where the pulse shape was determined by the relaxation time 2 μs of the power amplifier. The supplied heat was 53.6 erg in (a) and 129 erg in (b). In the case (a), the pulse directly leaving downward and that reflected at the top can be distinguished since their peaks are separated by $2d/c$ (which is 6.7 μs for $T-T_c=500$ mK). Afterward, a two-peak pulse was formed. In the case (b), the pulses are singly peaked because of the longer pulse-duration time 7 μs ($>2d/c$). In both (a) and (b), the pulse was gradually flattened as it moved in the cell. The broadening became more enhanced on approaching the critical point. Using these data we calculated the adiabatically increased energy $E_{\rm ad}$ ($=p\Delta x$) in the pulse region per unit area. To linear order it is expressed as

$$E_{\rm ad}=\frac{p}{\rho}\int dx\ \delta\rho(x,t)=\frac{pc}{\rho}\int dt\ \delta\rho(x,t). \tag{3}$$

The ratio of $E_{\rm ad}$ to the supplied heat $Q=\int dt\ \dot{Q}(t)$ is the efficiency of transforming applied heat to mechanical work. Over wide ranges of $T-T_c$ and Q, $E_{\rm ad}/Q$ was in the range 0.11–0.12 at the first arrival at the cell center. However, each pulse has a tail persisting in later times and there remains ambiguity of order 10% in the calculation of $E_{\rm ad}$.

In our experiment the fluid velocity in the pulse region was of order $v=c\delta\rho/\rho\sim 10^{-2}$ cm/s. The fluid displacement is then estimated by $\Delta x=v\Delta t$ where Δt is the pulse-duration time. For $\Delta t\sim 10$ μs we find $\Delta x\sim 1$ nm. For such extremely small displacements the film heater did not disturb the sound propagation [19].

We give a simple theory of sound emission from a heater to interpret our data, neglecting the effect of the bulk viscosity. Let a small amount of heat be supplied at a rate $\dot{Q}(t)$ per unit area for $t>0$ in a one-dimensional geometry. The volume expansion rate near the heater is $\dot{V}_1=(\partial\rho^{-1}/\partial s)_p\dot{Q}(t)/T$ per unit area, where s is the entropy per unit mass. If the heater is attached to the upper plate [20], this volume change produces a sound wave propagating downward as $\delta\rho_{\rm out}(t-x/c)$, where x is the distance from the heater and $\delta\rho_{\rm out}(t)$ is the density increase in the sound emitted at the heater at time t. Since acoustic disturbances leave with the sound velocity c, the mass conservation gives the sound amplitude

$$\delta\rho_{\rm out}(t)=\frac{\rho}{c}\dot{V}_1=\frac{\rho}{cT}\left(\frac{\partial T}{\partial p}\right)_s\dot{Q}(t), \tag{4}$$

where the Maxwell relation $(\partial\rho^{-1}/\partial s)_p=(\partial T/\partial p)_s$ has been used. The adiabatic coefficient $(\partial T/\partial p)_s$ is nearly equal to the derivative along the coexistence curve $(\partial T/\partial p)_{\rm cx}$ near the critical point [11]. For CO_2 it is equal to $T_c/6.98p_c$ [18] and the above relation becomes $\delta\rho_{\rm out}/\rho=1.38\times 10^{-13}\dot{Q}$ with $\dot{Q}$ in erg/cm^2 s. The heating rate used for our data in Fig. 2 was $\dot{Q}=0.183\times 10^7$ and then $\delta\rho_{\rm out}/\rho=2.55\times 10^{-7}$. This theoretical value fairly agrees with the observed height of the first step $\sim 2.2\times 10^{-7}$ in Fig. 2. The efficiency of energy transformation discussed below Eq. (3) becomes [2]

$$\frac{E_{\rm ad}}{Q}=\frac{p}{T}\left(\frac{\partial T}{\partial p}\right)_s, \tag{5}$$

which is $1/6.98=0.14$ for near-critical CO_2. The experimental efficiency values calculated from Eq. (3) were in the range 0.11–0.12.

We clarify the relationship of the formula (4) and the assumption in the theory of the piston effect [2]. Let the heat input $\dot{Q}(t)$ change slowly compared to the acoustic time $t_a=L/c$. We suppose a time interval with width $\delta t\gg t_a$, in which $\dot{Q}(t)$ is almost unchanged. After many sound traversals, the adiabatic pressure and density increases in the interior region are given by

$$\delta p=c^2\delta\rho=c^2\frac{\delta t}{t_a}\frac{\rho}{cT}\left(\frac{\partial T}{\partial p}\right)_s\dot{Q}, \tag{6}$$

as a result of superposition of many steps. In terms of the incremental heat supply $\delta Q=\dot{Q}\delta t$ we find

$$\delta p=\frac{\rho}{LT}\left(\frac{\partial T}{\partial p}\right)_s\delta Q=\left(\frac{\partial p}{\partial s}\right)_\rho\frac{\delta Q}{\rho TL}, \tag{7}$$

where the Maxwell relation $(\partial p/\partial s)_\rho=\rho^2(\partial T/\partial\rho)_s$ has been used. In the original theory [2], δp was assumed to be homogeneous in the whole cell without gravity; then Eq. (7) follows from the space average of $\delta p=(\partial p/\partial s)_\rho\delta s+(\partial p/\partial\rho)_s\delta\rho$, since the space integral of δs is $\delta Q/\rho T$ and that of $\delta\rho$ vanishes. The interior temperature deviation $T_{\rm in}=(\partial T/\partial p)_s\delta p$ may be expressed as [11]

$$T_{\rm in}=(\gamma-1)\delta Q/C_pL, \tag{8}$$

where $C_p=\rho T(\partial s/\partial T)_p$ is the isobaric specific heat. If the boundary temperature is raised by T_1 at $t=0$, Eq. (1) is reproduced since $\delta Q\sim\rho C_p\ell(t)T_1$ in the early stage.

In summary, we have measured acoustic emission and propagation caused by extremely small heat inputs in near-critical CO_2. Our setup has allowed measurements of adiabatic changes on the time scale of the acoustic time L/c. We have also presented the theoretical formulas (4) and (5). The height of stepwise sound from Eq. (4) and the efficiency from Eq. (5) agree fairly with the experimental values. Differences of 20–30 % remain, probably because a fraction of heat escaped to the stainless part and the pulse shape was considerably broadened even at the first arrival at the cell center.

There are a number of future problems. (i) We should understand how a sound pulse is reflected at a boundary wall and how it is damped in the bulk region [7]. Figure 4 suggests that the change of the pulse shape on reflection is more marked than that due to the bulk damping, as can be inferred from comparison of the results for $L=5.5$ mm in (a) and those for $L=1.03$ cm in (b). This is still the case even very close to the critical point. (ii) The bulk viscosity exhibits strong critical anomaly as $\zeta\cong 0.03\rho c^2t_\xi$ [11], where t_ξ is the order parameter relaxation time (18 μs at $T-T_c=30$ mK for CO_2) [14]. It gives a bulk acoustic damping, but it also affects the thickness of the thermal diffusion layer [9,10].

The effects of the bulk viscosity on heat transport are not well understood. (iii) We can produce a pulse with width shorter than t_ξ near the critical point. There is no study in this regime. (iv) Upon heat exchange, the wall temperature is not fixed when the effusivity ratio $a_w=(\lambda_w C_w/\lambda C_p)^{1/2}$ is small [3,9]. Here λ_w, λ, C_w, and C_p are the thermal conductivities and the specific heats (per unit volume) of the solid and the fluid, respectively. The asssumption of fixed boundary temperature leading to Eqs. (1) and (2) is valid only in the limit $a_w \gg 1$. For small a_w, the equilibration time becomes nearly a constant [3]. In our case $a_w=3\times 10^3(T/T_c-1)^{0.92}$ between Cu and CO_2 and $a_w<1$ is reached for $T-T_c<50$ mK. This crossover should be studied.

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[12] For CO_2 on the critical isochore, $\gamma\sim 0.1(T/T_c-1)^{-1.14}$. For example, $\gamma=590$ for $T-T_c=150$ mK.

[13] Equation (2) is valid for $t_1 \gg t_a$. Moreover, the assumption of fixed boundary temperature does not hold very close to the criticality. See the summary of this Rapid Communication.

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[16] We compare the heating rate $C_h\partial T/\partial t$ of the foil and the heat flux. Here C_h is the specific heat of the foil multiplied by its thickness. In our case $C_h/k_B=8.4\times 10^{19}/\mathrm{cm}^2$. If ω is the typical frequency, the ratio of the former to the latter is $0.02(T/T_c-1)^{1.24}(\omega/D)^{1/2}\ll 1$ in cgs units.

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[19] L. D. Landau and E. M. Lifshitz, *Theory of Elasticity* (Pergamon, New York, 1973). The mechanical stress needed to displace the center of the film plate by $\Delta x=10^{-7}$ cm is of order 10^{-5} dyn/cm^2, where the Young's modulus of the plate is 2.2×10^{12} dyn/cm^2. This value is much smaller than the acoustic pressure deviation $\sim c^2\delta\rho$.

[20] In our setup the heater was separated by $d=0.5$ mm from the top. Using the volume expansion rate at the top plate $\dot{V}_2$, we modify Eq. (4) to $c\delta\rho_{\mathrm{out}}(t)/\rho=\dot{V}_1+\dot{V}_2$. If ω is the typical frequency, we find $\dot{V}_2/\dot{V}_1\sim(\omega t_2)^{1/2}$ with $t_2\equiv L^2/c^2t_1$. In our case, t_2 was very short and $\dot{V}_2$ was negligible, so Eq. (4) is applicable.