PROGRESS ON ACOUSTIC MEASUREMENTS OF THE BULK VISCOSITY OF NEAR-CRITICAL XENON (BVX)

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We plan to determine the bulk viscosity of xenon 10 times closer [in reduced temperature $\tau = (T-T_c)/T_c$] to its liquid-vapor critical point than ever before. (T_c is the critical temperature.) To do so, we must measure the dispersion and attenuation of sound at frequencies 1/100 of those used previously. In general, sound attenuation has contributions from the bulk viscosity acting throughout the volume of the xenon as well as contributions from the thermal conductivity and the shear viscosity acting within thin thermoacoustic boundary layers at the interface between the xenon and the solid walls of the resonator. Thus, we can determine the bulk viscosity only when the boundary layer attenuation is small and well understood. We present a comparison of calculations and measurements of sound attenuation in the acoustic boundary layer of xenon near its liquid-vapor critical point.

We used a novel, compact, acoustic resonator designed specifically for these measurements (Fig. 1). We measured the frequency response of this resonator filled with xenon at its critical density ρ_c in the reduced temperature range $10^{-3} < \tau < 10^{-1}$. From the frequency-response data, we obtained the resonance frequency and the attenuation for six resonant modes in the range 0.10 kHz < f < 7.5 kHz. Using the known thermo-physical properties of xenon, we predict that the attenuation at the boundary first increases and then saturates when the effusivity of the xenon exceeds that of the solid. [The effusivity is $\varepsilon \equiv (\rho C_P \lambda_T)^{1/2}$, where C_P is the isobaric specific heat and λ_T is the thermal conductivity.] The model correctly predicts (±1.0 %) the quality factors Q of resonances measured in a steel resonator (ε_{ss} = 6400 kg·K^{-1}·s$^{-5/2}$); it also predicts the observed increase of the Q, by up to a factor of 8, when the resonator is coated with a polymer (ε_{pr} = 370 kg·K^{-1}·s$^{-5/2}$). The measured acoustic dissipation in near-critical xenon shows a prominent plateau for the Helmholtz mode and, to a lesser extent, for the longitudinal modes (Fig. 2). These results are the first direct

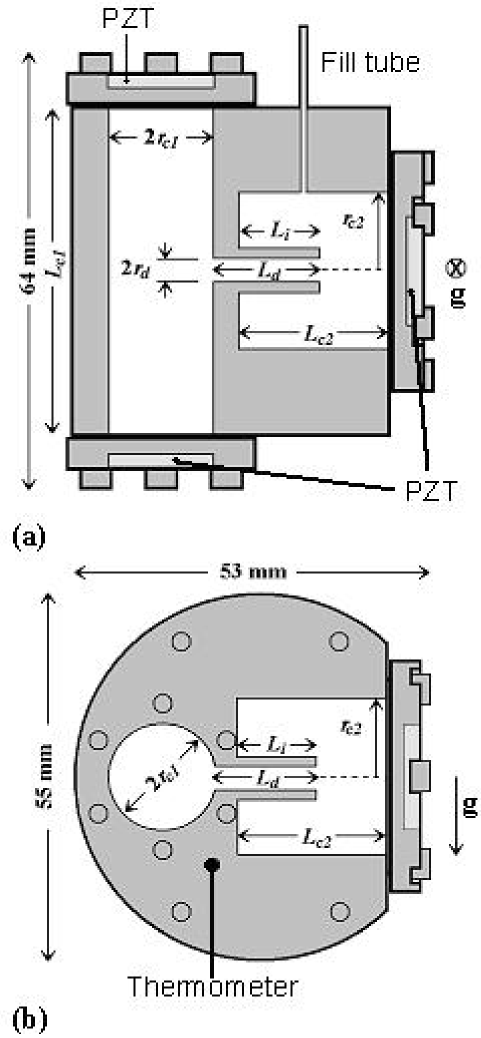

Figure 1. Acoustic resonator

evidence of thermal boundary dissipation being limited by the thermophysical properties of the solid wall.

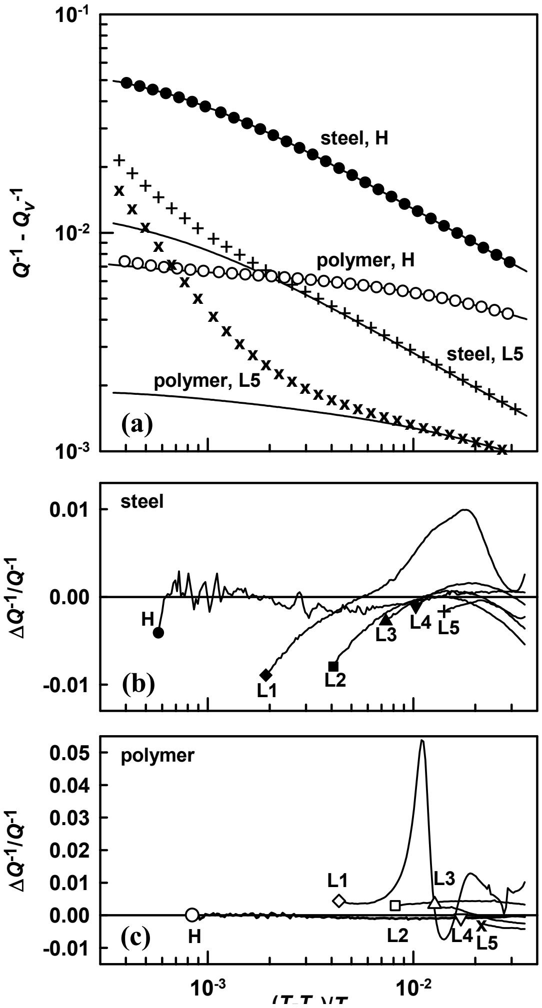

Figure 2. (a) Dissipation due to bulk viscosity and the thermal boundary layer versus reduced temperature. Theory for thermal boundary dissipation (———). The fractional deviations of the measured dissipation from theory versus reduced temperature for all the modes in the steel (b) and polymer-coated (c) resonators. Only data for which the dissipation from bulk viscosity was less than 0.8 % of the total dissipation is shown.

We observed an 8-fold reduction in the thermal boundary dissipation (as predicted) after coating a steel resonator with a low-effusivity polymer. (See Fig. 2a.) Such a reduction in thermal dissipation will be necessary in order to measure the low-frequency dissipation from bulk viscosity close to the critical point in future experiments.

To analyze the data, we formulated a theory for acoustic dissipation in a fluid that is bounded by a rigid wall with the condition that the temperature and heat flux across the boundary be continuous. The theory is valid both near to and far from the critical point; it includes volume and surface dissipation from thermal conduction, shear viscosity, and bulk viscosity.

We measured the speed of sound in xenon as a function of reduced temperature in the range $0.0006 < \tau < 0.03$. Using the measured sound speed and attenuation, the isothermal susceptibility from Ref. [1], and $(\partial P/\partial T)_\rho$ from Ref. [2], we derived C_V, C_P, and the background terms for the thermal conductivity. These derived properties were used to determine the amplitude $A^+ = 18.01$ for the singular part of C_V (with $\alpha = 0.110$) and the correlation length amplitude $\xi_0^+ = 0.1866$ nm. Although the data do not extend far into the asymptotic region, the agreement with other values is remarkable.

The onset of bulk viscosity is evident from the excess dissipation (over the thermal and viscous dissipation) indicated by the upturn at low τ of the measured dissipation shown in Fig. 2a. It is also evident that the dissipation from the bulk viscosity increases as the frequency increases in qualitative agreement with theory. The onset of bulk viscosity is more evident in the polymer-coated resonator because the thermal dissipation is smaller than it is in the bare-steel resonator.

[1] H. Güttinger and D.S. Cannell, Phys. Rev. A **24**, 3188-3201 (1981).
[2] H.L. Swinney and D.L. Henry, Phys Rev A **8**, 2566-2617 (1973).

Progress on acoustic measurements of the bulk viscosity of near-critical xenon (BVX)

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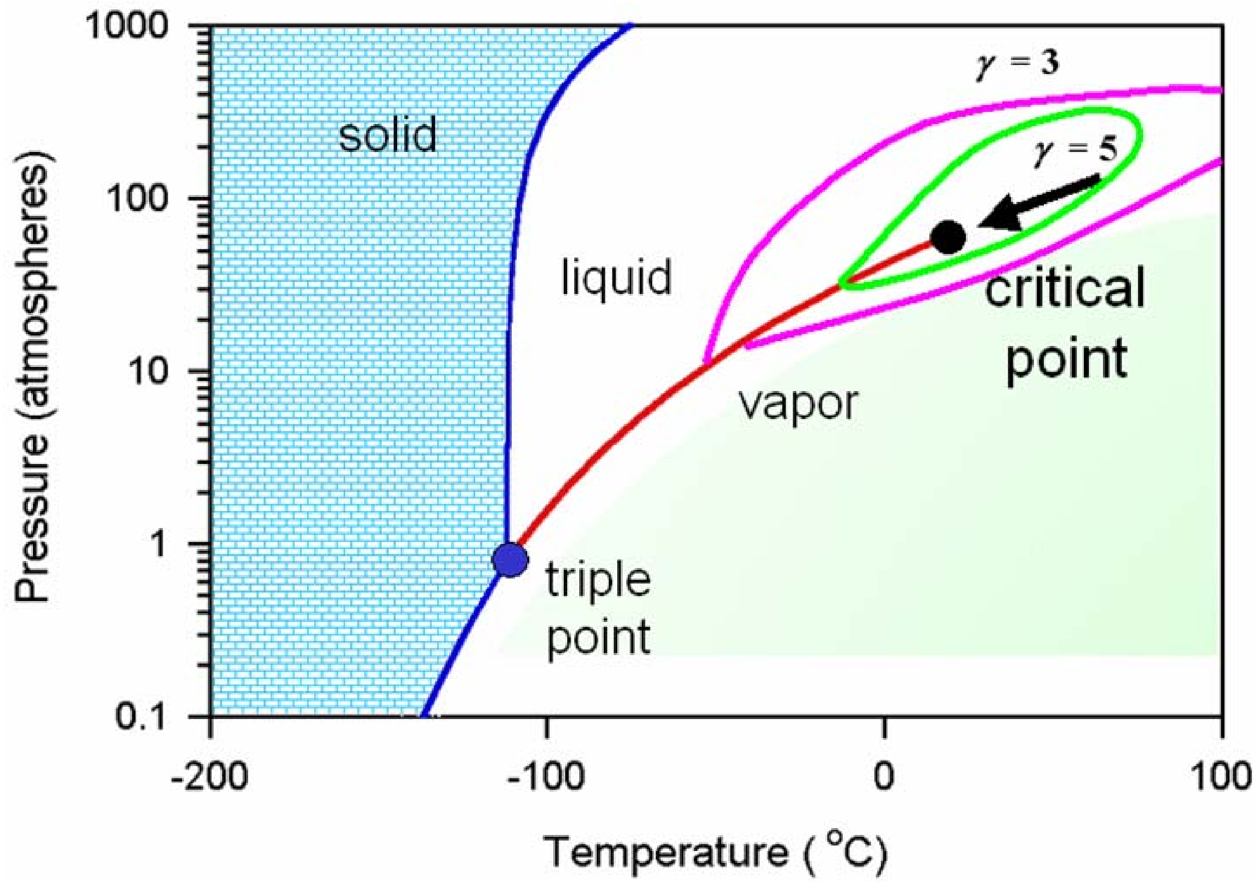
We plan to determine the bulk viscosity of xenon 10 times closer [in reduced temperature $\tau = (T-T_c)/T_c$] to its liquid-vapor critical point than ever before. (T_c is the critical temperature.) To do so, we must measure the dispersion and attenuation of sound at frequencies 1/100 of those used previously. We will determine the bulk viscosity ζ (also "second viscosity" or "dilational viscosity") from measurements of sound attenuation α_λ and dispersion using a novel, compact, acoustic resonator designed specifically for this purpose.

In general, sound attenuation has contributions from the bulk viscosity acting throughout the volume of the xenon and contributions from the thermal conductivity and the shear viscosity acting within thin boundary layers near the resonator wall. Thus, we can best determine the bulk viscosity when the boundary layer attenuation is small and well understood. We present a comparison of calculations and measurements of sound attenuation in the acoustic boundary layer of xenon near its liquid-vapor critical point.

The liquid-vapor critical point

The critical point is the highest temperature and pressure at which liquid and vapor can coexist in the same container. For xenon T_c=16.584 °C, P_c= 5.84 MPa

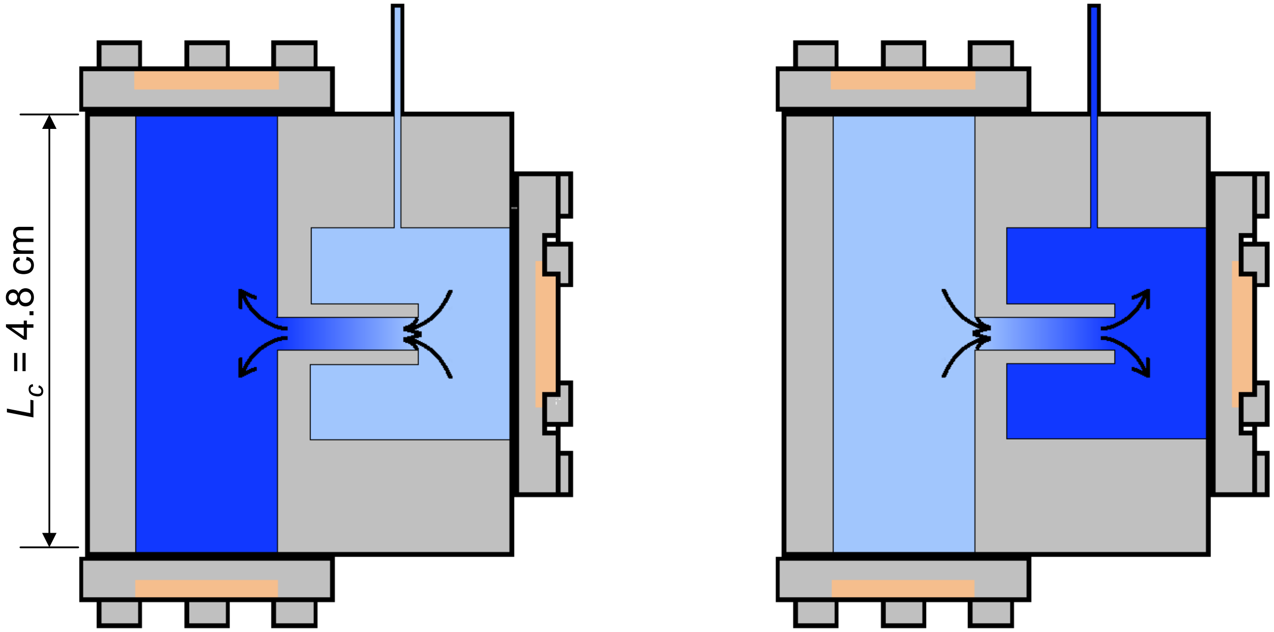


As the critical point is approached: the sound speed → 0; the bulk viscosity ζ diverges; $\gamma = C_p/C_v$ diverges. Attenuation in the thermal boundary layer is proportional to γ and therefore poses a potential problem. Conventional acoustic theory assumes $\gamma \approx 1$ and attenuation is a small perturbation.

Accomplishments

- We extended acoustic theory to include near-critical fluids, heat flow in solid wall, and bulk viscosity.
- We were the first to observe direct evidence of thermal boundary dissipation being limited by thermophysical properties of the solid wall.
- We demonstrated an 8-fold reduction in thermal boundary dissipation after coating steel with a polymer.
- We derived C_V, C_P, and background thermal conductivity from our measured speed of sound.
- We observed the onset of dissipation due to bulk viscosity.

Helmholtz mode

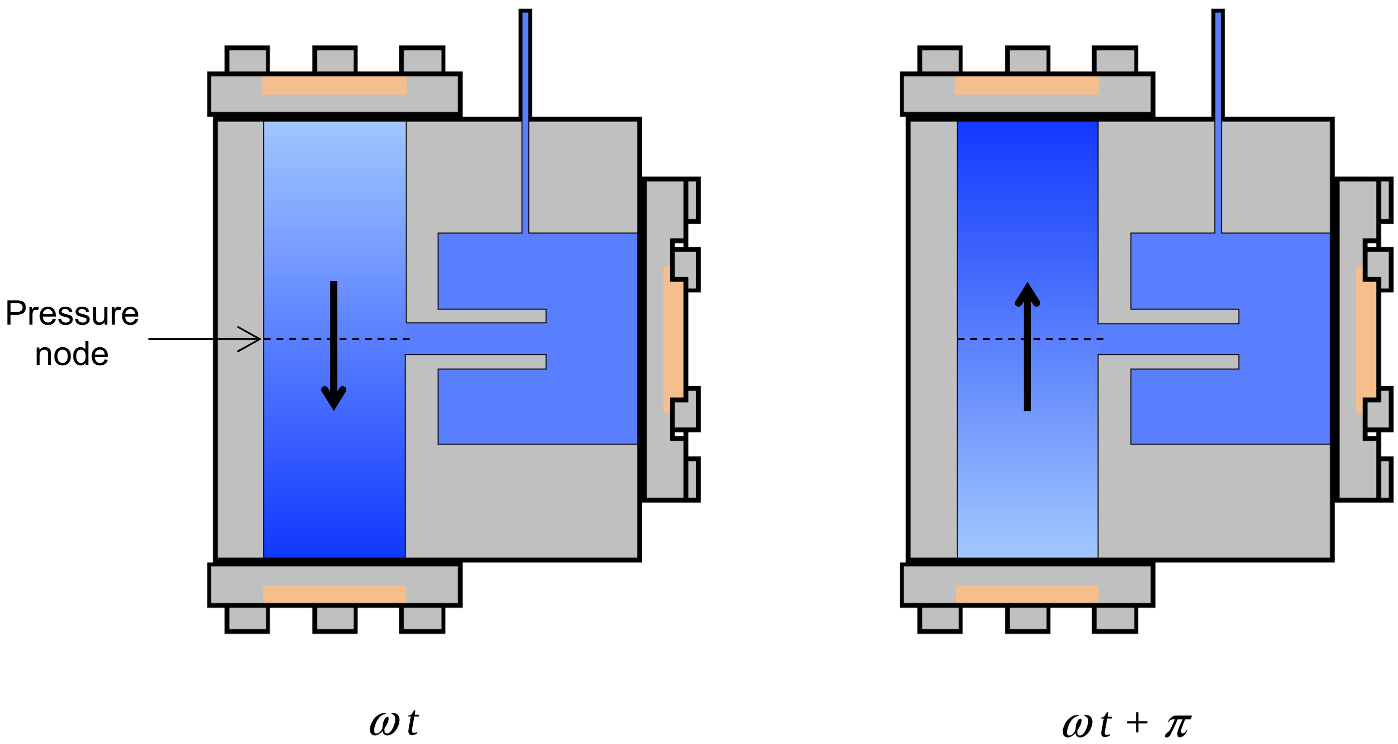


$$k_H = 0.1226\,\mathrm{cm}^{-1} \Rightarrow \lambda = 51.0\,\mathrm{cm} \approx 34L_d$$

In it's fundamental acoustic mode, the gas oscillates between two chambers through a small tube (radius = r_d and length = L_d). The wavelength of this mode is about 10 times the size of the resonator or 34 times the tube length. We measure the response as a function of frequency to determine the resonance frequency and Q. For xenon at its critical density, the frequency of this mode varies from 127 Hz at 3 mK above T_c to 240 Hz at 3 K above T_c.

We use resonators of this type (called double-Helmholtz resonators) to measure the viscosity of dilute gases. See K.A.Gillis, J.B. Mehl, and M.R. Moldover, "Theory of the Greenspan Viscometer," JASA 114, 166-173 (2003)

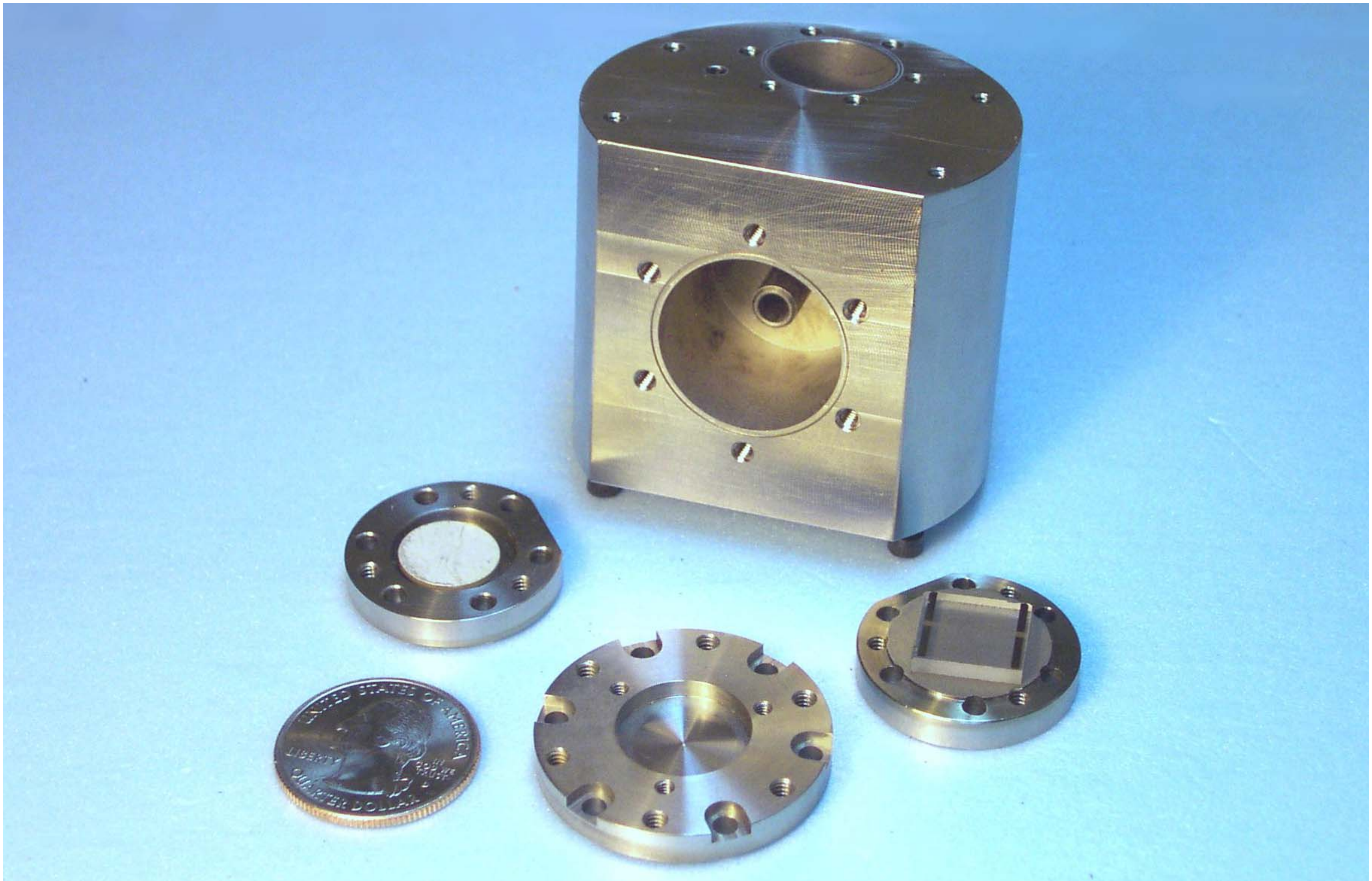
Longitudinal modes



$$k_{L1} = 0.6536\,\mathrm{cm}^{-1} \Rightarrow \lambda = 9.6\,\mathrm{cm} = 2L_c$$

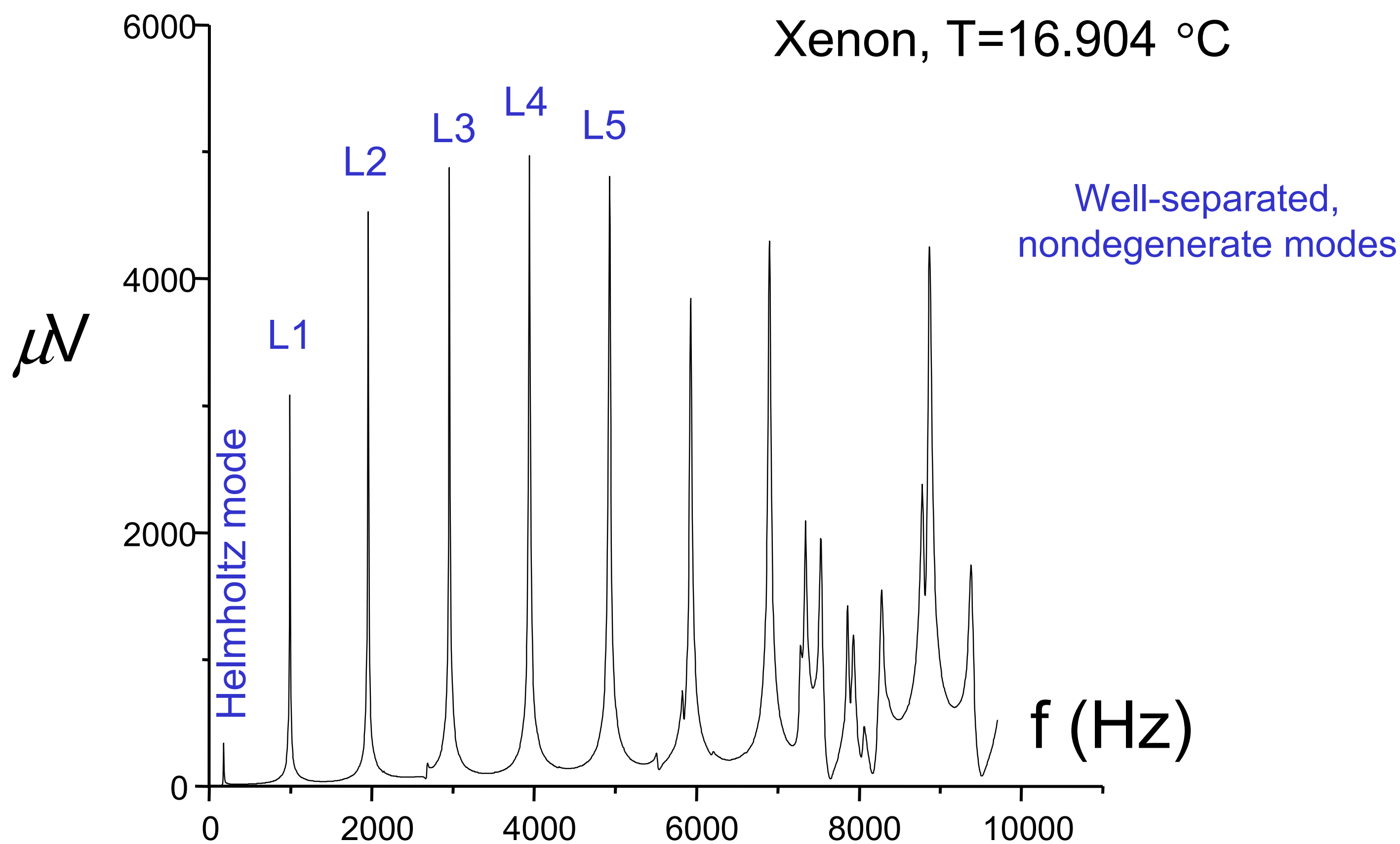
We deduce the sound speed and the acoustic attenuation from the resonance frequency and Q for a given mode.

To measure dispersion, we also study the first 5 longitudinal modes of the longer chamber. Like the Helmholtz mode these modes are non-degenerate and well separated because of the asymmetric resonator design. All 6 modes span a factor of 27 in frequency at a given temperature. The acoustic data span a factor of 60 in frequency over the full temperature range.



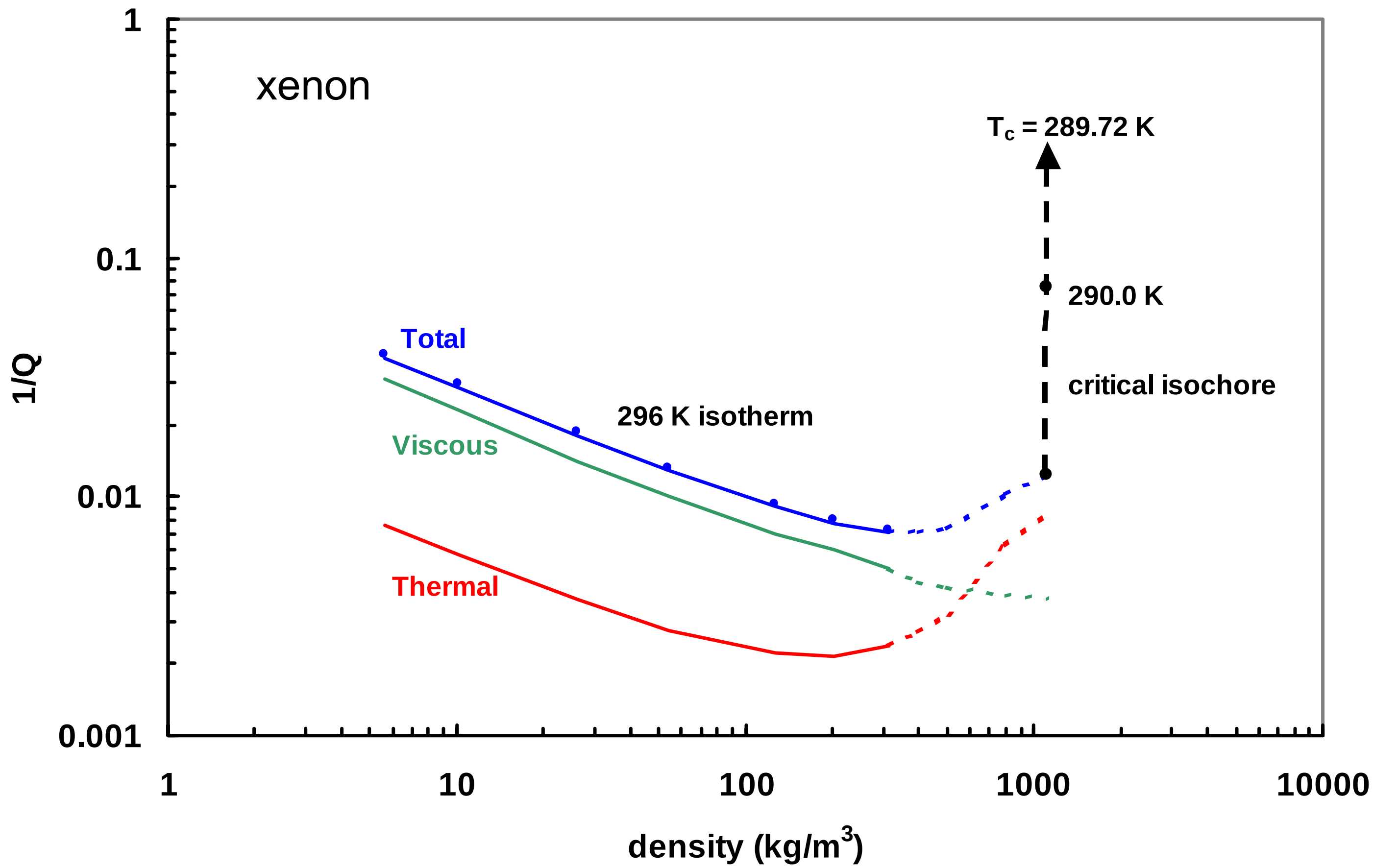
The 2 chambers and the connecting tube were machined as one piece from a block of stainless steel. The high rigidity is crucial for studies of dense fluids. Piezoceramic disks were attached to the outside of the flanges in order to drive and detect sound waves in the fluid. Two similar resonators were used. One resonator was coated with a polymer to reduce the thermal conduction between the xenon and the resonator wall. The other resonator was left as bare steel.

Acoustical Spectrum of Resonator



The graph shows the amplitude of the detector signal as a function of frequency. The Helmholtz mode and the longitudinal modes of the chamber (L1-L5) are clearly visible and well-separated. Isolated, non-degenerate modes are necessary for accurate modeling.

From dilute gas to the critical isochore



This graph shows the measured dissipation (1/Q) as the density increases from ambient conditions to the critical density (1116 kg/m^3) at constant temperature (blue curve). The contributions from viscosity and thermal conductivity within the boundary layers, calculated from the model, are shown in green and red. Viscous dissipation dominates at low density. Thermal dissipation dominates at high density due to the large γ. The black dashed line shows the dramatic increase in 1/Q as the temperature is lowered to T_c along the critical isochore.

Dependence of dissipation on thermal properties of the wall

thermal conduction | **shear viscosity** | **bulk viscosity**

$$Q^{-1} \approx (\gamma - 1)\frac{q_T \delta_T}{1 + \vartheta} + q_v \delta_v + \frac{\omega\zeta}{\rho c^2} + \ldots$$

ε **= effusivity**

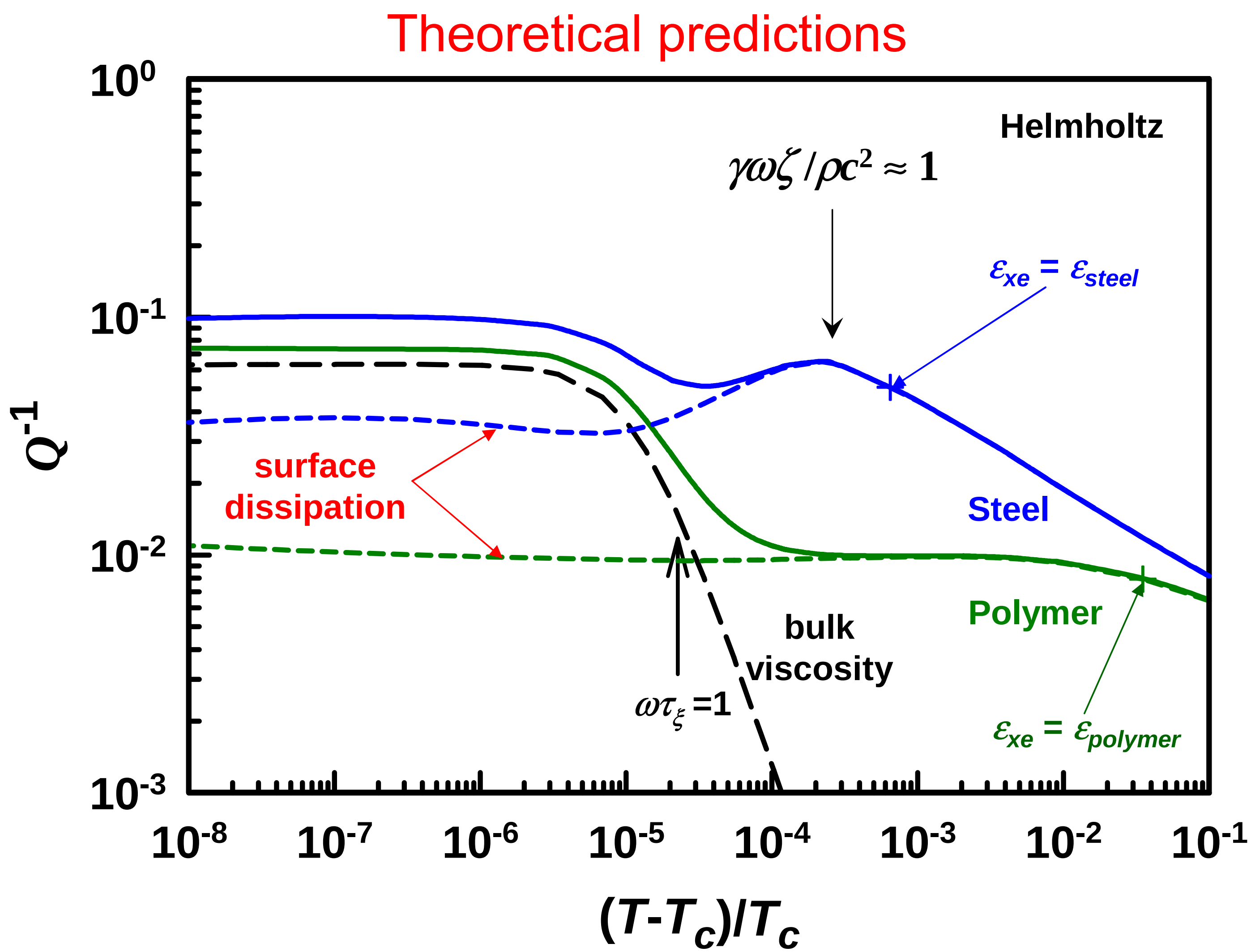
$$\boxed{\vartheta \propto \frac{\varepsilon}{\varepsilon_{\text{solid}}}} \qquad \boxed{\delta_T \approx \sqrt{\frac{2D_T}{\omega}}} \qquad \boxed{\delta_v = \sqrt{\frac{2\eta}{\rho\omega}}}$$

boundary layer thicknesses

$$\frac{\varepsilon}{\varepsilon_{\text{solid}}} = \frac{\left(\rho C_P \lambda_T\right)^{1/2}}{\left(\rho C_P \lambda_T\right)^{1/2}_{\text{solid}}}$$

	$\varepsilon_{\text{solid}}$ $(\text{kg} \cdot \text{K}^{-1} \cdot \text{s}^{-5/2})$
perfect solid	∞
copper	37000
steel	6400
polymer (parylene)	370

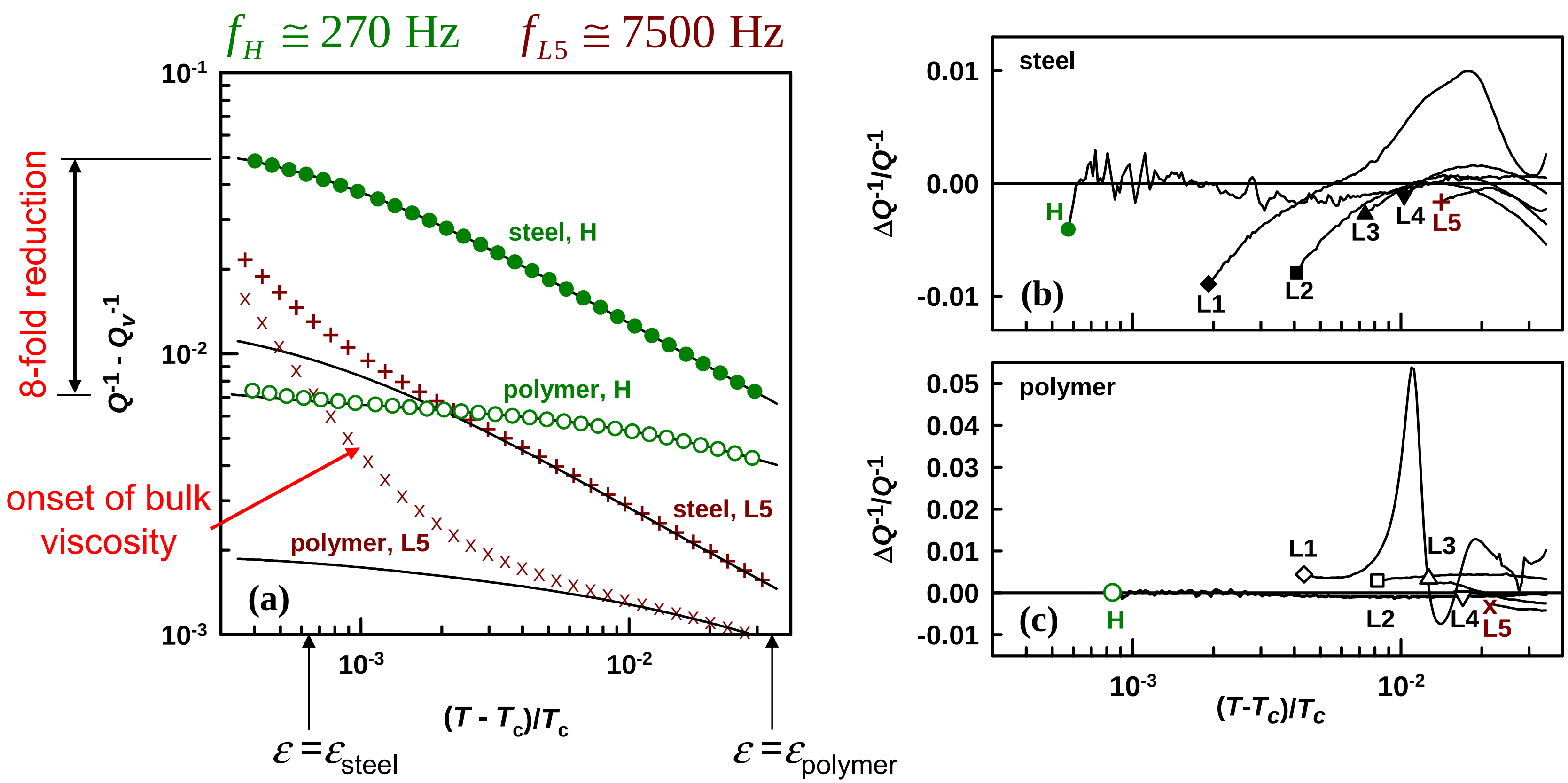
To lowest order, the dissipation is due to the sum of 3 contributions: thermal and viscous dissipation at the interface between xenon and the wall plus a volume dissipation due to bulk viscosity. The thermal dissipation reaches its maximum value when the ratio ϑ of the effusivity of xenon ε to the effusivity of the solid wall ε_{solid} equals one. Lower ε_{solid} means the thermal dissipation saturates at a lower value farther from T_c. See K.A. Gillis, I.I. Shinder, and M.R. Moldover, “Thermoacoustic boundary layers near the liquid-vapor critical point,” (in press Phys. Rev. E, July 2004).



Predicted dissipation as a function of reduced temperature in bare steel and polymer-coated resonators. The crosses indicate where the effusivity ε of xenon equals the effusivity of the solid. The surface dissipation from viscosity and heat conduction is shown with short dashed curves. The contribution from bulk viscosity ζ in the volume is shown with a long dashed curve. The relaxation of fluctuations gives rise to bulk viscosity and has characteristic time τ_ξ. The condition $\gamma\omega\zeta / \rho c^2 \approx 1$ indicates where the boundary layer becomes highly compressible.

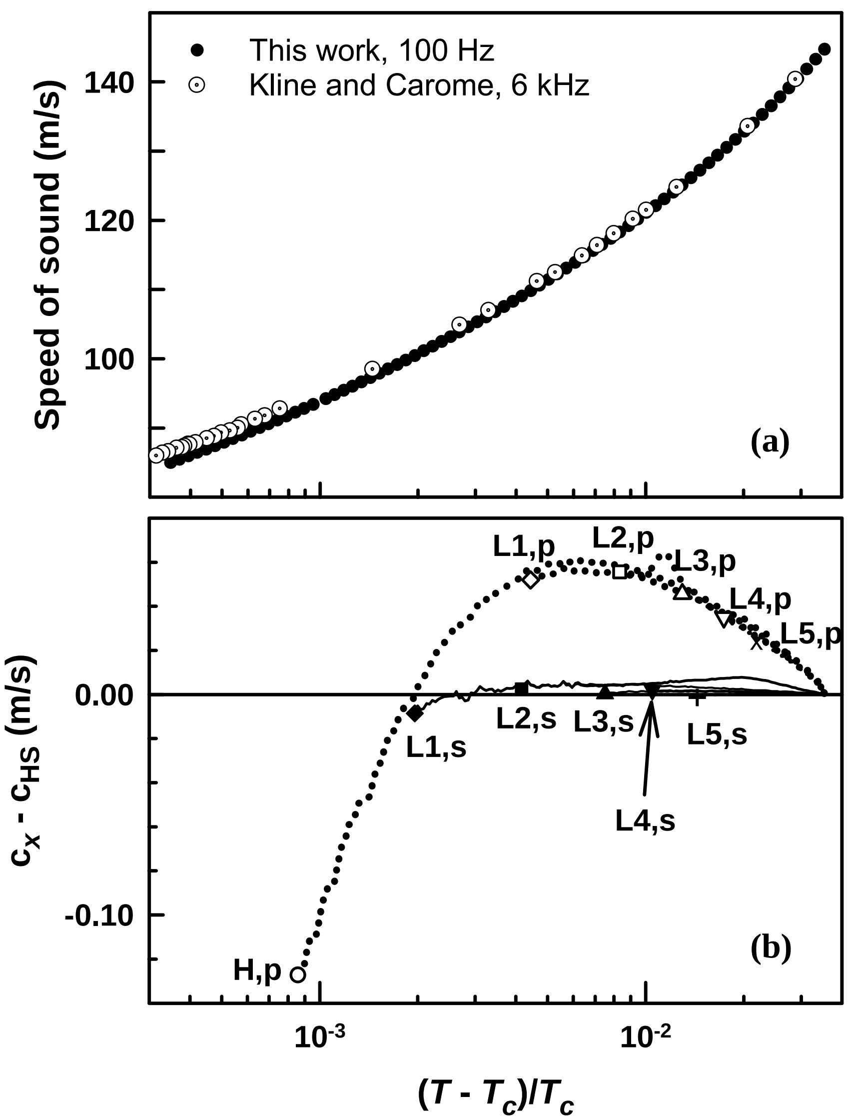
Comparison of measurements with theory

Xenon near its critical point



(a) ***Dissipation due to bulk viscosity and the thermal boundary layer versus reduced temperature. Theory for thermal boundary dissipation (———). The fractional deviations of the measured dissipation from theory versus reduced temperature for all the modes in the steel*** **(b)** ***and polymer-coated*** **(c)** ***resonators. Only data for which the dissipation from bulk viscosity was less than 0.8 % of the total dissipation is shown.***

Speed of sound in xenon near its critical point



(a) ***The speed of sound c_{HS} determined from the Helmholtz mode with bare stainless steel plotted versus the reduced temperature. The speed of sound measured by Kline and Carome at 6 kHz.***

(b) ***Deviations of the speed of sound determined from each mode from c_{HS} shown in the upper graph.***

Consistency among modes ~0.01%

Consistency between resonators ~0.1%