

Bulk viscosity of liquid noble gases

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ABSTRACT

An equation of state for the bulk viscosity of liquid noble gases is proposed. On the basis of dedicated equilibrium molecular dynamics simulations, a multi-mode relaxation ansatz is used to obtain precise bulk viscosity data over a wide range of liquid states. From this dataset, the equation of state emerges as a two-parametric power function with both parameters showing a conspicuous saturation behavior over temperature. After passing a temperature threshold, the bulk viscosity is found to vary significantly over density, a behavior that resembles the frequency response of a one pole low-pass filter. The proposed equation of state is in good agreement with available experimental sound attenuation data.

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I. INTRODUCTION

In contrast to prevailing opinion, bulk viscous effects have widely been explored in fluid mechanics and even share a somewhat controversial history. In 1845, Stokes¹ had argued geometrically that Cauchy's stress tensor field,²

$$\Pi_{ij} = \left(\mu_b - \frac{2}{3} \mu_s \right) \partial_n v_n \delta_{ij} + \mu_s (\partial_j v_i + \partial_i v_j), \quad (1)$$

entails not only the well-known shear viscosity μ_s but also the bulk viscosity μ_b , which he concurrently postulated to be zero,

$$\mu_b \stackrel{!}{=} 0. \quad (2)$$

Stokes, whose proof can hardly be considered rigorous, had limited his argument to incompressible fluids and remained skeptical that his finding (2) would emerge as universally valid. For more than a century, this hypothesis was afflicted with misconceptions until the Royal Society hosted "a discussion on the first and second viscosities of fluids,"^{3–18} reinvigorating investigations of bulk viscous effects on a wide variety of fluid mechanical problems.

Riemann's solution¹⁹ of the Euler equation²⁰ was among the first. Retaining the bulk viscosity in the stress tensor field²¹ conclusively solved the problem of insensibly small wave thicknesses that were predicted^{22,23} in descriptions limited to shear viscosity.²⁴ The conjectured increase in wave thickness was qualitatively confirmed in successive atomistic simulations of non-ideal liquids^{25,26} and in

numerical investigations of rarefied gases,^{27–29} given that the shock structure is symmetric.³⁰

Bulk viscous effects have long been considered mere higher-order contributions.³¹ However, including the bulk viscosity in the acoustic approximation^{32,33} straightforwardly explained the observed second-order fields associated with ultrasonic waves. The vorticity that is generated across a propagating wave's free surface induces a counter-oriented circulatory flow^{34–39} that is also known as acoustic streaming or quartz-wind. In hypersonic boundary layer approximations, bulk viscous effects are promoted to second-order in the pressure distribution⁴⁰ and even first-order in the outer and inner flow velocities of high Reynolds number flows.⁴¹

In fluids confined to capillaries, the bulk viscosity contributes to first-order in radial pressure and to second-order in the axial velocity.^{42–47}

Bulk viscous behavior was also investigated for more complex scenarios. An increased shock wave thickness affects the outer flow's adverse pressure gradient and consequently suppresses the shock induced boundary layer separation,⁴⁸ while additionally the shock wave's location and strength are much more accurately predicted when bulk viscous effects are included.⁴⁹ Likewise, bulk viscous damping has been observed in compressible turbulent flows.⁵⁰ A non-zero bulk viscosity enhances kinetic energy dissipation while additionally inhibiting the energy transfer between translational and configurational energy, thus rendering the flow effectively incompressible.⁵¹

Each of the aforementioned investigations, however, has suffered from missing, incomplete, or unreliable bulk viscosity data and was consequently restricted to predominantly qualitative results.

II. SOUND ATTENUATION

Since its introduction, the bulk viscosity was closely associated with linear acoustics. Stokes had originally proposed that sound attenuation measurements could confirm his hypothesis (2) experimentally. A propagating wave's linear momentum is dissipated by a surrounding atmosphere, which leads to the exponential decay of its amplitude $A(z)$ over traveled distance $\Delta z = z_2 - z_1$ and is measured by an attenuation coefficient α ,

$$\frac{A(z_2)}{A(z_1)} = \exp\left(-\alpha_\lambda \frac{\Delta z}{\lambda}\right), \quad (3)$$

given here per wavelength λ , i.e., $\alpha_\lambda = \alpha\lambda$. In a first estimate of α_λ for one-dimensional motion, limited to shear viscosity μ_s , Stokes found that the attenuation factorizes into frequency $\omega = 2\pi f$ and a transport function ξ ,

$$\alpha_\lambda = \omega \xi^{\text{Stokes}} = \omega \frac{\pi}{K} \frac{4}{3} \mu_s, \quad (4)$$

where $K = \rho c^2$ is the fluid's low-frequency modulus of compression, ρ its density, and c its thermodynamic speed of sound. Kirchhoff⁵² advanced the discussion by including heat conduction and established that both effects superimpose linearly in the transport function,

$$\begin{aligned} \alpha_\lambda^{\text{classical}} &= \omega \left(\xi^{\text{Stokes}} + \xi^{\text{Kirchhoff}} \right) \\ &= \omega \frac{\pi}{K} \left(\frac{4}{3} \mu_s + (\kappa - 1) \frac{\gamma}{c_p} \right), \end{aligned} \quad (5)$$

where $\kappa = c_p/c_v$ is the ratio of specific heats and γ is the thermal conductivity. The attenuation predicted by this classical theory corresponds ostensibly well with low frequency acoustic measurements.

Subsequently performed ultrasonic measurements,^{53–57} however, disclosed large deviations from the classical description (5) and motivated Herzfeld⁵⁸ and Kneser^{59,60} to introduce an additional mechanism,

$$\alpha_\lambda = \alpha_\lambda^{\text{classical}} + \alpha_\lambda^{\text{excess}}. \quad (6)$$

Both connected classical hydrodynamics to relaxation theory and attributed the excess attenuation $\alpha_\lambda^{\text{excess}}$ to a time lag that occurs during the transfer of energy between the molecules' translational and internal degrees of freedom. In contrast, Tisza⁶¹ refrained from any physical interpretation and incorporated the Herzfeld mechanism into the transport function ξ by force fitting the complex mechanism of relaxation into a scalar valued bulk viscosity,⁶²

$$\begin{aligned} \alpha_\lambda &= \omega \left(\xi^{\text{Stokes}} + \xi^{\text{Kirchhoff}} + \xi^{\text{Tisza}} \right) \\ &= \omega \frac{\pi}{K} \left(\frac{4}{3} \mu_s + (\kappa - 1) \frac{\gamma}{c_p} + \mu_b \right). \end{aligned} \quad (7)$$

This extended theory has been validated experimentally up to moderate ultrasonic frequencies, i.e., $f \leq 280$ MHz,⁶³ over a wide range of thermodynamic states.

In the extended vicinity of the critical point, however, an anomalously high attenuation was observed^{64–66} that was conclusively attributed to long-range density correlations,^{67–73} which are alterations of structural relaxation effects.^{74–77} The total attenuation partitions into the already established background part (7) and a critical contribution,^{78,79}

$$\alpha_\lambda^{\text{total}} = \alpha_\lambda + \alpha_\lambda^{\text{critical}}. \quad (8)$$

Acoustic dispersion, i.e., the frequency dependent speed of sound $c(\omega)$, confines this critical attenuation to an extended critical region in the thermodynamic state space, cf. [supplementary material](#),

$$\alpha_\lambda^{\text{critical}} = \pi \frac{I(\tilde{\omega})}{J(\tilde{\omega})} \left(\left[\frac{c(\omega)}{c} \right]^2 - 1 \right), \quad (9)$$

where $I(\tilde{\omega}), J(\tilde{\omega})$ are improper integrals of a characteristic frequency $\tilde{\omega}$.

III. EXPERIMENTAL DATA

At present, the bulk viscosity is determined experimentally either by non-resonant Rayleigh–Brillouin scattering^{80–90} or ultrasonic attenuation measurements.^{91–121} Both techniques have successfully been applied to a variety of substances, yet each measurement series was restricted to selective thermodynamic states.

A substantially larger dataset, however, can be obtained for liquid noble gases by utilizing their self-similar behavior. While all available sound attenuation measurements for neon, argon, krypton, and xenon were evaluated, data subject to critical attenuation

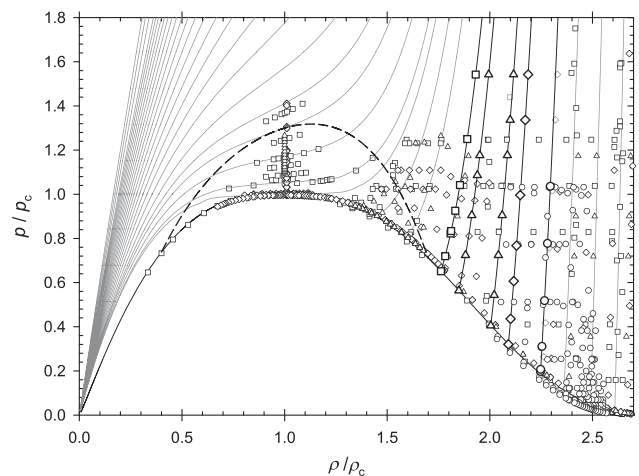


FIG. 1. Overview of thermodynamic states at which experimental sound attenuation data are available for neon¹⁰⁶ (circles), argon^{98–101,110,112,113,115,116} (squares), krypton^{115–117,119} (triangles), and xenon^{79,115,116,118,119,122} (diamonds). *Highlighted:* Seven isotherms were selected $T/T_c = 0.759, 0.76, 0.83, 0.86, 0.863, 0.91,$ and 0.93 , while $T/T_c = 0.759$ and 0.863 were omitted in the plot for visibility reasons, along which atomistic simulations were performed to complement and extend the dataset to higher pressures $p/p_c \leq 2.1$. All thermodynamic states were reduced with the respective fluid's critical pressure p_c and density ρ_c . The extended critical region was constructed on the basis of sound dispersion measurements from the literature and is delimited by the dashed line.

TABLE I. The following parameters for the full, i.e., untruncated Lennard-Jones potential σ , ϵ , and atomic mass m were used in the canonical transformation¹³⁰ to reduce the bulk viscosity and time $t^* = t\sqrt{\epsilon/(m\sigma^2)}$, where k_B is Boltzmann's constant.

| | ϵ/k_B (K) | σ (Å) | m (u) |
|----|--------------------|--------------|---------|
| Ne | 33.92 | 2.801 | 20.180 |
| Ar | 116.79 | 3.395 | 39.948 |
| Kr | 162.58 | 3.627 | 83.798 |
| Xe | 226.14 | 3.949 | 131.293 |

were identified and discarded, cf. Fig. 1. The bulk viscosity was calculated on the basis of the extended classical theory (7) and in contrast to previous works, more accurate thermodynamic data were available by resorting to most recent equations of state.^{123–127} Subsequently, μ_b was reduced by a canonical transformation resting on the Lennard-Jones potential, i.e., $\mu_b^* = \mu_b \sigma^2 / \sqrt{m\epsilon}$, with the required set of parameters specified in Table I.

Each contribution to bulk viscosity is afflicted by a different uncertainty. While the uncertainty of most recent thermodynamic data are well established to range from $\Delta\rho/\rho = 0.005$ to $\Delta\mu_s/\mu_s = 0.3$ depending on substance and state, the attenuation coefficient α_λ has been determined by single measurements at the respective state point, and thus, only its absolute maximum errors $\Delta\alpha_\lambda$ have been estimated. Moreover, it is not assured that systematic measurement errors in the literature data, specifically diffraction effects,¹²⁸ were properly accounted for. Consequently, the reduced bulk viscosity's uncertainty $\Delta\mu_b$ was determined to be linearly affected¹²⁹ by its various contributions $\Delta\alpha_\lambda$, Δc , $\Delta\mu_s$, $\Delta\gamma$, Δc_v , Δc_p ,

$$\Delta\mu_b = \left| \frac{K}{\omega\pi} \Delta\alpha_\lambda + \dots + \left| \frac{(\kappa - 1)\gamma}{c_p^2} \right| \Delta c_p \right| \quad (10)$$

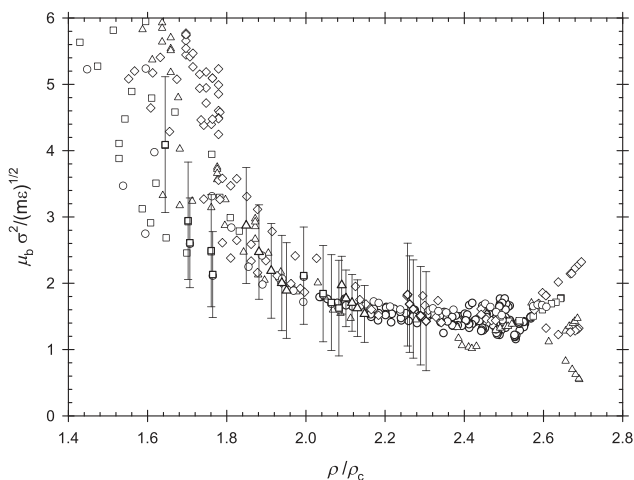


FIG. 2. Overview of bulk viscosity data calculated from the extended classical theory on the basis of experimental sound attenuation data, taken from the literature at the state points indicated in Fig. 1. **Highlighted:** Experimental data at the selected isotherms $T/T_c = 0.76, 0.83, 0.86, 0.91, 0.96$ with their respective uncertainties, calculated from Eq. (10), indicating a monotonic decline of μ_b as a function of density.

After selecting and evaluating the experimental data, a decline of the bulk viscosity along each isotherm, i.e., from saturation line toward higher density, can qualitatively be inferred. The effect tends to increase with temperature, however, due to large errors neither the function's gradient nor the curvature can accurately be specified, cf. Fig. 2.

IV. MOLECULAR DYNAMICS SIMULATION

To interpret and extend the experimental dataset to higher densities, additional bulk viscosity data were sampled by equilibrium molecular dynamics (EMD) simulations along seven selected isotherms $T/T_c = 0.759, 0.76, 0.83, 0.86, 0.863, 0.91, 0.93$, cf. Fig. 1. The bulk viscosity was determined microscopically by time-autocorrelation functions of local small-scale, transient pressure fluctuations^{131,132} that are intrinsic in any fluid under equilibrium.¹³³ The Lennard-Jones interaction potential was used, which has demonstrated to resolve such small-scale dynamics adequately and hence successfully describes macroscopic transport in liquid noble gases.^{134–138}

In the present work, the bulk viscosity's autocorrelation function B_A was sampled in the microcanonical (NVE) ensemble, utilizing the fully open source program *ms2*.¹³⁹ The necessary average energies $\langle E \rangle$ were determined by preceding canonical (NVT) ensemble simulations for the given pair of temperature and density. Finite size effects were minimized by placing $N = 4096$ particles in cubic volumes with periodic boundary conditions and choosing a sufficiently large cutoff radius $r_c \geq 5.5\sigma$.^{140–142} The employed particle number is well chosen, as simulations containing $N = 12\,000$ particles yielded virtually identical results, cf. [supplementary material](#). In order to adequately resolve both the existing small-scale dynamics and also the slowly decaying pressure fluctuations,¹⁴³ a reduced integrator time step $\Delta t^* = 5 \cdot 10^{-4}$ was specified and each autocorrelation function B_A was sampled over a reduced time period of at least $t^* \geq 14.6$.

A. Relaxation ansatz

The fluid's intrinsic small-scale pressure fluctuations have conclusively been established to relax in different modes.^{144–146} Each mode decays exponentially over time following a Kohlrausch–Williams–Watts function.^{147,148} For all of the investigated state points, three superimposing relaxation modes were found to be present, leading to the relaxation model's analytical form,

$$B_R(t) = C_f \exp\left(-\left(\frac{t}{\delta_f}\right)^{\beta_f}\right) + C_m \exp\left(-\left(\frac{t}{\delta_m}\right)^{\beta_m}\right) + C_s \exp\left(-\left(\frac{t}{\delta_s}\right)^{\beta_s}\right). \quad (11)$$

The first term describes the fast, and the subsequent terms describe the intermediate and slow modes, respectively. The weighting factors are constraint $C_f + C_m + C_s = 1$, and the Kohlrausch parameters δ_i , β_i are a measure of relaxation time scale and distortion from the exponential function, respectively. The eight independent parameters of Eq. (11) were determined by fitting the relaxation model B_R to the sampled autocorrelation function B_A at each state point independently.

Each mode's average relaxation time τ_i is properly defined as integral mean value of its respective contribution $B_{R,i}$ to the relaxation model,¹⁴⁹

$$\tau_i = \frac{1}{C_i} \lim_{t \rightarrow \infty} \int_0^t dt (B_{R,i}(t)). \quad (12)$$

As originally proposed by Maxwell, the bulk viscosity μ_b is proportional to the cumulative averaged relaxation time,

$$\mu_b = K_r \sum_i C_i \tau_i, \quad (13)$$

with the proportionality constant K_r being the fluid's relaxation modulus.¹⁵⁰

V. RESULTS

A. Relaxation times

The present results are exemplarily discussed for states along the isotherm $T/T_c = 0.86$ yet are qualitatively similar for all other investigated state points as disclosed in the [supplementary material](#).

The sampled autocorrelation function B_A partitions into three segments, with each segment being dominated by a different mode, cf. Fig. 3. The two dents in the sampled signal readily indicate the fast and intermediate mode's decay. Accompanying the latter, incipiently small scale oscillations are amplified, giving rise to substantial noise contributions in the slow mode. After reconstructing its signal by post-processing, the good agreement between the sampled autocorrelation function B_A and the proposed relaxation model B_R becomes apparent.

More importantly, in contrast to the sampled autocorrelation function that is plagued by noise, the employed relaxation model's time integral properly converges to a definite value, thus allowing to determine the bulk viscosity unambiguously at each state point, cf. Fig. 4.

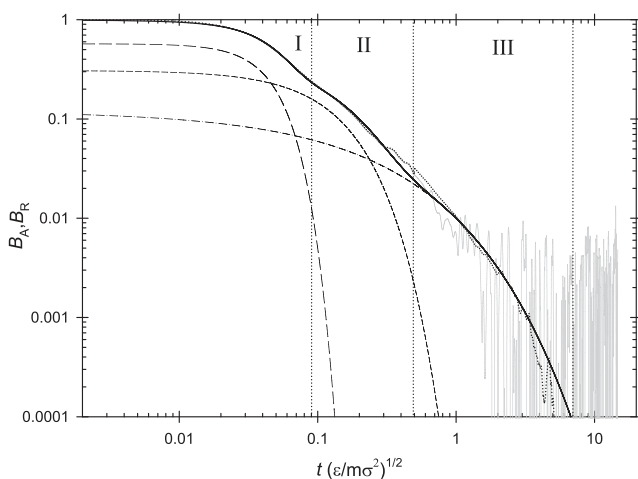


FIG. 3. Comparison of sampled autocorrelation function and relaxation model including all three relaxation modes. The gray line constitutes B_A sampled at $T/T_c = 0.86$ and $\rho/\rho_c = 2.06$, while the solid black line represents the relaxation model (11), and its fast, intermediate, and slow modes are depicted by dashed, short-dashed, and dashed-dotted lines, respectively. The post-processed simulation signal (dotted line) is in good agreement with the relaxation model.

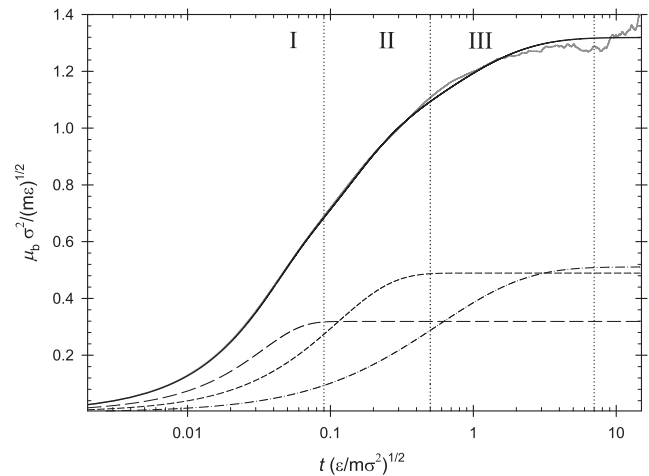


FIG. 4. Comparison of the integrated autocorrelation function B_A with the relaxation model's (11) integral at $T/T_c = 0.86$ and $\rho/\rho_c = 2.06$. Due to noise contributions to B_A , the bulk viscosity μ_b is difficult to determine precisely by molecular dynamics simulation (gray line). In contrast, the proposed relaxation model (solid black line) converges toward an unambiguous value at finite times. The dashed, short-dashed, and dotted-dashed lines represent the fast, intermediate, and slow modes, respectively.

The average reduced relaxation times τ_i were found to decline exponentially with density for each mode and to differ roughly by one order of magnitude among the modes up to $\rho/\rho_c \leq 2.2$. While all relaxation modes are increasingly damped with rising density, facilitating shorter relaxation times, the slow mode is damped disproportionately, cf. Fig. 5.

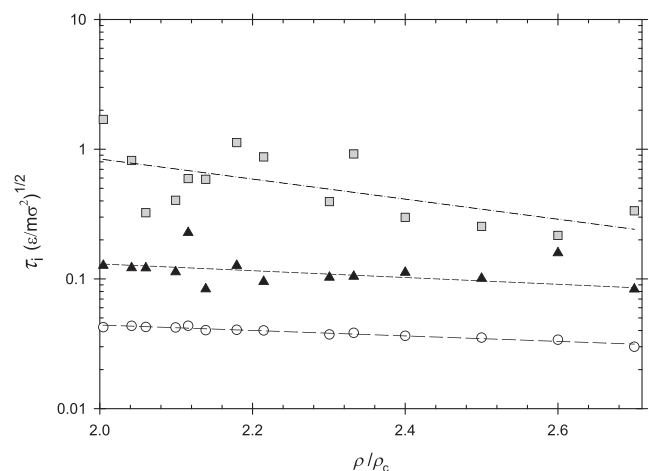


FIG. 5. Distribution of average reduced relaxation times τ_i for all three modes along the isotherm $T/T_c = 0.86$. While each mode is identified to relax exponentially, the slow mode is predominantly affected by increasing density. The symbols represent the relaxation times of the fast τ_s (white circles), the intermediate τ_m (black triangles), and the slow mode τ_s (gray squares), respectively.

B. Equation of state

The most recognized equation of state, of the very few that have actually been discussed in the literature,^{151,152} relates the bulk viscosity to a power function of density,

$$\mu_b \propto \rho^\alpha, \quad (14)$$

for which special case solutions with fixed exponents $2\alpha = -1, 0, 1, 3$ exist.^{153–156} This proportionality, which was originally proposed in the context of viscous cosmological fluids, is much more universal and also applies to liquid noble gases if the observed temperature dependence is included.

After consolidating all relaxation results, a threshold temperature $T_t/T_c \sim 0.74$ emerges below which the reduced bulk viscosity indicates virtually no variation with density.

In contrast, at higher temperatures, μ_b increases progressively toward saturated liquid states, an effect that intensifies with rising temperature, cf. Fig. 6.

TABLE II. Best fit parameters a_i, b_i, c_i of Eq. (16).

| | a_i | b_i | c_i |
|------------|-------|-------|-------|
| α_1 | −0.93 | 5.91 | 8.67 |
| α_2 | −0.53 | −1.49 | 7.86 |

While any physically sound equation of state must necessarily establish a unique one-to-one relation between μ_b and each state point, i.e., being bijective, entropy constraints additionally restrict this equation to be non-negative^{157,158} and the present results further specify it to be convex with monotonically decreasing gradient and curvature. Satisfying all conditions, a reduced two-parametric power function is proposed

$$\mu_b^* = \left(\frac{\rho}{\rho_c} - 1 \right)^{\alpha_1} + \alpha_2, \quad (15)$$

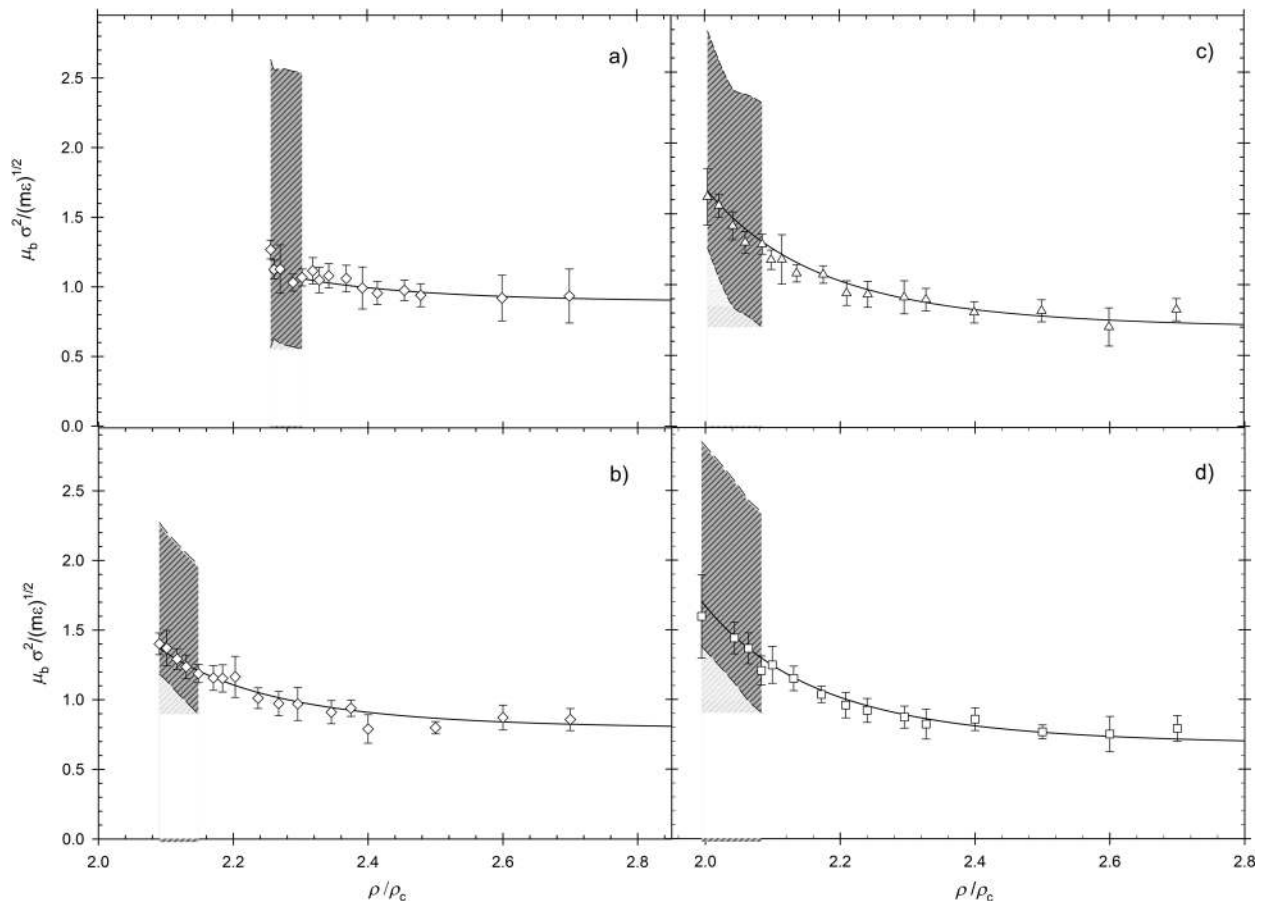


FIG. 6. Outline of bulk viscosity variations from the saturation line toward high density along four isotherms (a) $T/T_c = 0.759$, (b) $T/T_c = 0.83$, (c) $T/T_c = 0.86$, and (d) $T/T_c = 0.863$. The gray shaded areas represent the experimentally determined bulk viscosity, according to Eq. (7), including its absolute maximum error, i.e., $\mu_b \pm \Delta\mu_b$, according to Eq. (10). The symbols indicate bulk viscosity data obtained by the employed relaxation ansatz (11) on the basis of the present EMD simulations. The solid line constitutes the present equation of state (15).

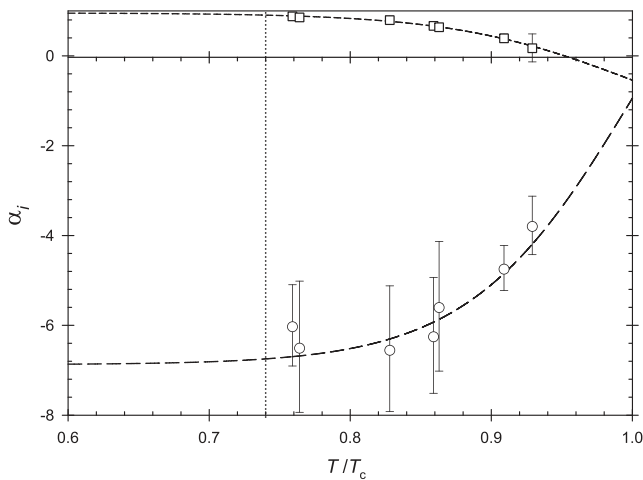


FIG. 7. Variation of parameters α_1 (circles) and α_2 (squares) over temperature. Above the threshold temperature $T/T_c \sim 0.74$ indicated by the vertical line, the bulk viscosity shows a substantial density dependence.

with parameters α_1, α_2 depending exclusively on temperature,

$$\alpha_i = a_i + b_i \cdot \tanh\left(c_i \left(\frac{T}{T_c} - 1\right)\right). \quad (16)$$

Both parameters, as specified in Table II, offer a salient saturation behavior that closely resembles the frequency response of a one

TABLE III. Comparison of the bulk viscosity from the present equation of state (15) with literature data that have either been obtained by molecular dynamics simulation^{140,159} or by theoretical calculations.¹⁶⁰

| T/T_c | p/p_c | ρ/ρ_c | $\mu_b \sigma^2 / \sqrt{m\epsilon}$ Eq. (15) | $\mu_b \sigma^2 / \sqrt{m\epsilon}$ literature |
|---------|---------|---------------|-------------------------------------------------|---------------------------------------------------|
| 0.795 | 0.545 | 2.196 | 1.17 | 1.20 ± 0.37 ¹⁴⁰ |
| ... | 3.616 | 2.353 | 0.99 | 0.93 ± 0.05 |
| ... | 8.727 | 2.510 | 0.92 | 0.87 ± 0.05 |
| ... | 16.558 | 2.667 | 0.88 | 0.89 ± 0.05 |
| 0.875 | 1.118 | 2.039 | 1.42 | 1.10 ± 0.36 |
| ... | 3.274 | 2.196 | 0.98 | 1.03 ± 0.05 |
| ... | 6.966 | 2.353 | 0.79 | 0.79 ± 0.04 |
| 0.674 | 0.473 | 2.448 | 1.00 | 0.97 ± 0.10 ¹⁵⁹ |
| 0.679 | 0.707 | ... | 1.00 | 0.98 ± 0.10 |
| 0.826 | 7.875 | ... | 0.89 | 0.75 ± 0.08 |
| 0.699 | 17.065 | 2.773 | 0.94 | 1.10 ± 0.11 |
| 0.763 | 21.558 | ... | 0.91 | 1.00 ± 0.11 |
| 0.802 | 24.239 | ... | 0.87 | 0.96 ± 0.10 |
| 0.605 | 3.117 | 2.640 | 0.96 | 1.05 ¹⁶⁰ |
| 0.652 | 6.149 | ... | 0.96 | 0.93 |
| 0.667 | 7.126 | ... | 0.96 | 1.01 |
| 0.678 | 7.800 | ... | 0.96 | 1.00 |
| 0.747 | 12.086 | ... | 0.93 | 0.86 |
| 0.792 | 14.845 | ... | 0.89 | 0.86 |

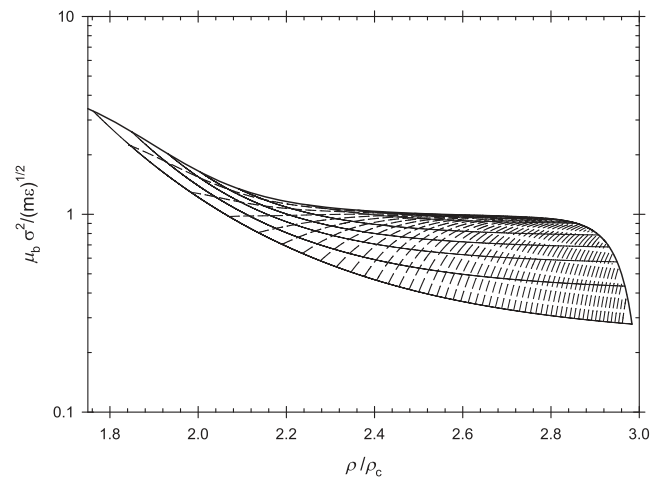


FIG. 8. Illustration of the present equation of state (15). The solid lines constitute the bulk viscosity's variation over density at constant temperature, and the dashed lines constitute its variation at constant pressure. The present results straightforwardly explain the experimental results in Fig. 2, which already suggested the bulk viscosity to be a monotonically declining function of density with increasing gradient for higher temperatures.

pole low-pass filter, cf. Fig. 7, causing the fluid to become increasingly more bulk viscous after passing the threshold temperature. This observed temperature dependence is caused by a transition from short-range order, that is present at low temperatures,¹⁵⁰ to the long-range density correlations that are intrinsic within the extended critical region. The present equation of state is in good agreement not only with experimental sound attenuation data but also with concurrent MD simulations, cf. Table III.

VI. CONCLUSION

An equation of state for the bulk viscosity of liquid noble gases is proposed. The bulk viscosity originates microscopically from relaxations of small-scale pressure fluctuations which were found to decay in three different modes following stretched exponential functions. The slow mode was observed to be disproportionately affected by high density and the average relaxation times τ_i between the modes to differ roughly by one order of magnitude. Each mode was determined on the basis of an autocorrelation function that was straightforwardly sampled by EMD simulations at the respective state point. In order to adequately resolve the slow mode, considerably long autocorrelation functions were necessary.

The equation of state emerges as a two-parametric power function with parameters depending exclusively on temperature, cf. Fig. 8. This temperature dependence is attributed to a transition from short-range order that is present at high densities to long-range density correlations that arise when approaching the extended critical region. After a threshold $T/T_c \sim 0.74$ is passed, the fluid becomes increasingly more bulk viscous. This effect causes sound attenuation to rise progressively with temperature, closely resembling the frequency response of a one pole low-pass filter. In addition, both bulk viscosity coefficients were observed to exhibit opposing behavior, i.e., an increase in bulk viscosity corresponds to a decline of shear viscosity and vice versa, causing the viscosities' ratio to peak

$\mu_b/\mu_s \sim 5$ at the highest investigated temperature close to the saturation line.

SUPPLEMENTARY MATERIAL

See the [supplementary material](#) for the construction of the critical region, as well as for a full disclosure of all data associated with this manuscript.

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