

On the ratio of the shear viscosity to the density of entropy of the rare gases and H₂, N₂, CH₄, and CF₄



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ABSTRACT

The ratio κ of the shear viscosity η to the volume density of entropy s , $\kappa = \eta/s$, of the rare gases He, Ne, Ar, Kr, and Xe and the molecules H₂, N₂, CH₄, and CF₄ is studied in the critical region. It is shown that the minimum value κ_m of η/s is observed in the vicinity of the critical point. In addition the universal inequality $\kappa \geq h/(4\pi k)$ proposed by Kovtun et al. (2005) holds for these gases. For the rare gases and ⁶Li κ_m depends linearly on $m^{1/2}\sigma^2$, where m is the mass and σ defined via the potential energy function as $U(\sigma) = 0$. κ_m of the molecules H₂, N₂, CH₄, and CF₄ falls on the same straight line provided that the vibrational and rotational contributions to the entropy density s are removed.

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1. Introduction

In 2005 Kovtun et al. [1] studied the shear viscosity in strongly interacting quantum field theories from black hole physics. In particular they concentrate on the ratio κ of the shear viscosity η to the volume density of entropy s , $\kappa = \eta/s$. They speculate that $\kappa_0 = h/(4\pi k) \approx 6.08 \times 10^{-13}$ Ks ($h = 2\pi\hbar$ is the Planck constant and k is the Boltzmann constant) is a lower bound to κ for all single-component nonrelativistic systems of particles with spin 0 or 1/2 leading to the universal inequality $\kappa \geq \kappa_0$. A fluid with $\kappa = \kappa_0$ is denoted a perfect fluid. Kovtun et al. and Zhou et al. [2] showed that even in the case of strongly interacting systems with very small η like the quark-gluon plasma the inequality $\kappa \geq \kappa_0$ holds. Of course this fundamental inequality gave rise to some controversial discussions. Buchel [3] and others [4,5] pointed out a possible violation of this inequality whereas Cohen tried to give some counterexamples for a system of increasing number of particle species [6,7]. However, the few available theoretical and experimental studies confirm this inequality. As was shown by Fortov and Mintsev [8] κ of a fully ionized strongly coupled plasma comes close to κ_0 but still is in full accordance with the inequality $\kappa > \kappa_0$. Müller et al. [9] studied the ratio η/s of graphene and also found $\kappa > \kappa_0$ in that case. Two-dimensional Fermi gases also obey the inequality $\kappa > \kappa_0$ [10,11]. Csernai et al. [12] pointed out some similarities between the general behavior of κ near the critical points of He, N₂, and H₂O and that of the phase transition from hadrons to the quark-gluon plasma. Of more interest to most chemists seems

to be the behavior of κ of ordinary fluids. A few of them like He, N₂, and H₂O have been casually studied [1,13,12]. A general but more refined analysis was given by March and co-workers [14] who additionally considered NH₃ and dense alkali metals [15], too. They found that also for ordinary systems the inequality $\kappa \geq \kappa_0$ holds and the minimum value κ_m is observed at or very near the critical point. In this paper we present a much more refined study of κ near the critical point of the gases He, Ne, Ar, Kr, Xe, H₂, N₂, CH₄ and CF₄. In our analysis we rely on the data presented in the NIST fluid properties database [16]. In addition experimental results for ⁶Li are taken into consideration.

2. Methodology

In our analysis of $\kappa = \eta/s$ we rely on the thermophysical properties given in the NIST Standard Reference Database Number 69 [16]. The shear viscosity η and the density ρ are used without changes, whereas the entropy S is normalized such as to yield $S = 0$ for $T \rightarrow 0$. For all gases the κ -surface was scanned in the ranges of $p_c \pm \Delta p$ and $T_c \pm \Delta T$, where p_c and T_c are the critical pressure and critical temperature, respectively. As a typical result obtained from the database in Ref. [16] some isobars for neon are shown in Fig. 1. These curves also support the observation made by Csernai et al. [12] that for ordinary gases κ has a minimum at the critical point, whereas for $p < p_c$ a discontinuity occurs and for $p > p_c$ a broad smooth minimum can be observed. In view of the accuracy of the thermophysical input data we can conclude that for all gases considered here a minimal value κ_m of κ is located on the critical isobar in the vicinity of the critical temperature T_c .

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Table 1
Critical constants and fit-parameters according to Eq. (1) of the rare gases.

	He	Ne	Ar	Kr	Xe
$10^{-6}p_c/\text{Pa}$	0.22746	2.6786	4.8630	5.5250	5.8420
T_c/K	5.1953	44.49	150.687	209.48	289.733
$10^{12}P_0/(\text{Ks})$	5.3395 (fixed)	10.214(12)	22.552(17)	34.861(17)	50.816(14)
$10^{12}P_1/(\text{Ks})$	8.865(61)	29.15(24)	43.62(47)	49.70(26)	74.40(40)
$10^{12}P_2/(\text{Ks})$	7.777(31)	27.8(1.1)	47.3(1.5)	88.9(2.6)	106.0(1.5)
P_3	0.8048(26)	0.5592(28)	0.5203(32)	0.4202(15)	0.4424(13)
P_4/K	5.19897(22)	44.5078(15)	150.71000(64)	209.520085(66)	289.74160(70)
P_5	0.5697(15)	0.870(14)	0.6993(89)	0.7734(78)	0.6537(35)
$10^{12}\Delta\kappa/(\text{Ks})$	0.014	0.028	0.032	0.029	0.024

Table 2
Critical constants and fit-parameters according to Eq. (1) of the molecules H_2 , N_2 , CH_4 , and CF_4 . The results for the parameters $P_0, P_1, P_2, P_3, P_4, P_5$, and the standard deviation $\Delta\sigma$ of N_2 , CH_4 , and CF_4 in the second row refer to the fit of the translational part $\kappa_{\text{tr},m}$, see text.

	H_2	N_2	CH_4	CF_4
$10^{-6}p_c/\text{Pa}$	1.2964	3.3958	4.5992	3.7500
T_c/K	33.18	126.192	190.564	227.51
$10^{12}P_0/(\text{Ks})$	5.3257 (fixed)	14.125 (fixed)	12.3486(48)	24.374(26)
$10^{12}P_1/(\text{Ks})$	12.33(13)	19.788(44)	17.609(19)	43.473(56)
$10^{12}P_2/(\text{Ks})$	12.23(31)	26.81(32)	20.71(15)	25.52(27)
P_3		49.7(3.4)	37.25(78)	220(32)
P_4/K		0.5054(34)	0.5666(22)	0.3803(33)
		0.4911(54)	0.5294(29)	0.3456(30)
P_5		126.2244(56)	190.5728(16)	227.320000(29)
		126.2231(66)	190.5976(22)	227.320000(15)
		0.6837(64)	0.5970(31)	0.836(18)
		0.788(23)	0.6571(57)	0.994(37)
$10^{12}\Delta\kappa/(\text{Ks})$	0.019	0.051	0.0088	0.044
		0.093	0.020	0.11

Subsequently $\kappa = \eta/s = \eta/(Sg)$ is calculated with a small resolution of $\delta T < 0.1$ K near the critical temperature T_c along the critical isobar p_c . The range ΔT depends on the gas and is typically in the order of a few Kelvin. The resulting curves are displayed in Fig. 2. In all cases a pronounced minimum is observed at or in the direct vicinity of the critical point. In order to locate the minimal value κ_m the critical isobars are fitted to a piecewise defined six parameter function.

$$\kappa = \begin{cases} P_0 + P_1 \times (-\tau)^{P_3} & \text{for } T < P_4 \\ P_0 + P_2 \times \tau^{P_5} & \text{for } T \geq P_4 \end{cases} \quad (1)$$

with $\tau = (T - P_4)/P_4$. With $P_4 = T_c$ this functional form is often used for an analytic modeling of measurable quantities in the vicinity of the critical point [17]. The minimal value of κ is given by $\kappa_m = P_0$. In Table 1 the fit parameters are listed together with the critical data of the rare gases He, Ne, Ar, Kr, and Xe. In Table 2 the same properties are displayed for the molecules H_2 , N_2 , CH_4 , and CF_4 . In order to get a stable fit in some cases P_0 was fixed to the minimal value κ_m extracted directly from the entries in the database [16]. In all cases a small standard deviation $\Delta\kappa$ of the fit with $\Delta\kappa/P_0 < 0.003$ is observed.

3. Analysis of κ_m

The ratio $\kappa = \eta/s$ can be used as an indicator of the coupling inside a fluid. According to Kovtun et al. [1] a fluid obeying the

relation $\kappa = \kappa_0$ is called a perfect fluid. In this sense the substances studied in this work are not perfect fluids. However in the group of the ordinary fluids studied here helium and hydrogen can be considered as the most perfect ones with nearly equal values of $\kappa_m \approx 8.8\kappa_0$. Whereas the relatively small ratio of κ_m/κ_0 of He has been noticed before [12,18] this observation is new for normal hydrogen. In our further analysis we also include experimental results obtained for the Lithium isotope ${}^6\text{Li}$ which forms a quantum degenerate, strongly interacting Fermi gas at low temperatures [19–21]. Measurements on ${}^6\text{Li}$ yield an approximate minimal value of $\kappa_m = (\eta/s)_{\text{min}} \approx 3.8 \times 10^{-12}$ Ks [19]. Having now experimental material on κ_m of some ordinary gases and the Fermi gas ${}^6\text{Li}$ we seek for a relationship between κ_m and other molecular properties. In the kinetic theory of gases the viscosity depends on the mass m and the collisional cross section $\pi\sigma^2$. On the other hand the entropy S can be obtained from statistical mechanics. For atoms in the ideal gas limit the only molecular property entering S is the mass m as can be seen in the Sackur–Tetrode equation.

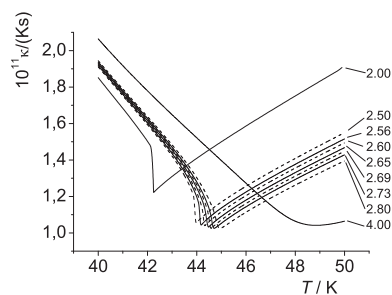


Fig. 1. κ as a function of the temperature at various pressures p for neon. The pressures on the right scale are given in 10^6 Pa.

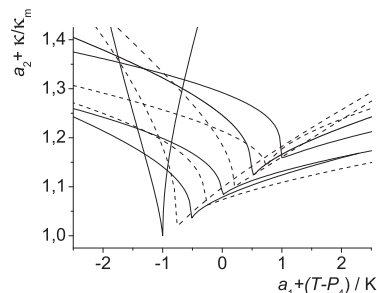


Fig. 2. Reduced representation of the critical isobars of $\kappa = \eta/s$ of the gases He, Ne, Ar, Kr, Xe, H_2 , N_2 , CH_4 , and CF_4 . $\kappa_m = P_0$ is the observed minimal value of κ whereas P_4 in all cases is nearly exactly the critical temperature T_c , see text and Tables 1 and 2. For the sake of clarity the resulting curves are shifted with the offsets a_1 and a_2 . In the above mentioned sequence of gases we have $a_1 = -1.0, -0.75, -0.5, -0.25, 0.0, 0.25, 0.5, 0.75,$ and 1.00 and $a_2 = 0.0, 0.02, 0.04, 0.06, 0.08, 0.10, 0.12, 0.14,$ and 0.16 , respectively.

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