PHYSICAL REVIEW E **106**, 054150 (2022)

Quantum mechanical interpretation of the minimum viscosity of metallic liquids

A. K. Gangopadhyay,[1,*] Z. Nussinov,[1,2] and K. F. Kelton[1]
[1]*Department of Physics, Washington University in St. Louis, One Brookings Drive, St. Louis, Missouri 63130, USA*
[2]*Rudolf Peierls Centre for Theoretical Physics, University of Oxford, Oxford OX1 3PU, United Kingdom*

(Received 16 September 2022; accepted 2 November 2022; published 22 November 2022)

Possible fundamental quantum bounds for viscosity and many other physical properties have drawn serious considerations recently from diverse communities encompassing those studying quantum gravity, high-energy physics, condensed matter physics, strongly correlated electron systems, and "strange metals," to name a few. However, little attention has been paid by materials scientists and the fluid dynamics community, perhaps because of the general belief that quantum mechanics is of little consequence for classical fluid dynamics. Here, considering the extrapolated high-temperature viscosity of 32 metallic alloy liquids as representative of minimum viscosity, experimental results are presented and evaluated in terms of a number of quantum- and statistical-mechanics-based theories. The surprising result is that the experimental data are within one order of magnitude of estimates from those theories. That quantum mechanics could be of importance at such high temperatures in conventional classical fluids is quite interesting. Another surprise is that the minimum viscosities of metallic alloy liquids are not too different from an archetypal quantum liquid, such as He.

DOI: 10.1103/PhysRevE.106.054150

I. INTRODUCTION

Among the hydrodynamic properties of a liquid, the shear viscosity is of utmost importance for glass formation. It is a property that reflects the timescale for momentum dissipation and structural relaxation. In glass-forming systems, it changes by 12−16 orders of magnitude between the liquidus (T_l) and the glass transition temperatures (T_g). An important question that has attracted much attention in recent years is whether a fundamental lower bound constrains the viscosity of all materials. Interestingly, this has been strongly debated not among the glass community, but among other communities such as string theorists examining the viable consequence of the powerful conjectured gauge/gravity duality [1], high-energy and nuclear physicists probing quark-gluon plasmas [2], and condensed matter physicists studying strongly correlated electronic systems, especially high-T_c superconductors [3]. Intriguingly, hydrodynamic models are finding increasing applications for charge conduction and dissipation timescales in strongly correlated electron systems [4], similar to momentum dissipation in liquids. Naturally, quantum gravity and quantum field theories are the basic starting points of such theories [1–4]. Although quantum mechanics provides the foundation for all fundamental theories of material properties, classical and statistical mechanics have remained the cornerstone of fluid dynamics; quantum mechanics is considered to be of little consequence. Therefore, it might appear a little strange that such disparate exotic theories have a connection with ordinary fluid flow properties. Here, we present experimental results for the minimum viscosity of metallic liquids at temperatures far above T_l and compare them with theoretical predictions. In general, the experimental results are roughly within an order of magnitude agreement with different theoretical predictions. These results, therefore, are expected to be of considerable interest to theorists active in many areas of physics since the main focus of these theories is on the lower bound of viscosity.

Motivated by notions from reaction rate theory, the temperature dependence of liquid shear viscosity is generally described by the simple relationship,

$$\eta = \eta_o \exp[E(T)/k_B T], \tag{1}$$

where η_o is a constant, $E(T)$ the temperature-dependent activation energy of a particular material, and k_B the Boltzmann constant. However, this simplicity is lost when the functional form of $E(T)$ over the entire temperature range, encompassing the equilibrium and supercooled liquids (below T_l), becomes the subject of first principle theories or empirical models. Predicting $E(T)$ over the entire temperature range, particularly for supercooled liquids, is a challenging endeavor for theorists [5–9]. In contrast, by assuming the activated form of Eq. (1) it has been much easier to suggest a possible material specific value for η_o [10]. Almost a century ago, inspired by notions of reaction rate theory, Eyring proposed a theory for the viscosity of liquids based on reaction kinetics and propagation of "holes" of the size of the molecules in the liquid [10]. After several approximations, he arrived at a simple, but intriguing link between Planck's constant h (which appeared in the quantum attempt rate for overcoming an energy barrier in the reaction rate theory), and the prefactor in Eq. (1), η_o, via a geometric factor containing the atomic or molecular volume number density n, as $\eta_o = nh$. Although the century-old Eyring's description of fluid dynamics under shear as a sequence of reactions associated with "hole hopping" between equilibrium states is rather simplistic, with

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[*] anup@wuphys.wustl.edu

hindsight, it appears to be quite illuminating in positing a connection between a seemingly classical quantity such as the viscosity of a fluid at noncryogenic temperatures and Planck's constant of quantum mechanics. Since the activation energy in Eq. (1) is positive, Eyring's expression for the viscosity suggests that it cannot be smaller than nh. In a recent publication [11], rigorous proofs have been presented that, under quite modest conditions, the product nh sets the scale for a lower bound for the viscosity. By introducing the "local" Heisenberg uncertainty principle in many-body systems and invoking other considerations, various bounds (both upper and lower) for many other dynamic and thermodynamic properties were suggested [11].

Originating from an entirely different perspective on the hydrodynamic behavior of a special type of black hole, called a black brane, it was suggested sometime back [1] that the ratio of the shear viscosity and the entropy density (s) has a universal lower bound, determined by the fundamental constants h and k_B.

$$\frac{\eta}{s} \geqslant \frac{h}{8\pi^2 k_B} = 6.08 \times 10^{-13}\,\mathrm{K\,s}. \tag{2}$$

This will be referred to as the KSS prediction. It is consistent with measurements of viscosity of relativistic quark-gluon plasmas at extremely high temperatures in relativistic heavy-ion collider experiments at CERN [2]. It was also argued that this limit may apply even in strongly interacting Bose-Einstein condensates (BECs) at low temperatures and nonrelativistic liquids such as water and He. After much initial controversy, this conjecture has been largely accepted [12], although the prefactor may differ from $1/8\pi^2$ in different systems. It is quite noteworthy that fundamental constants may determine properties such as the viscosity! If μ is the entropy per atom, then $s = n\mu$, and Eq. (2) becomes

$$\eta \geqslant \left(\frac{\mu}{8k_B\pi^2}\right) nh, \tag{3}$$

It should be noted that the lower bound on the viscosity that Eyring's relation suggests and the estimate from Eq. (3) are very similar, although they originated from entirely different theoretical perspectives. Since μ/k_B is dimensionless, the prefactor is just a numerical quantity differentiating the two estimates.

In another recent development, Trachenko and Brazhkin (hereafter referred to as TB) [13] argued for a minimum value of the kinematic viscosity, $\nu_m = \eta/\rho$ (ρ is the mass density), of liquids that can be expressed in terms of the electron (m_e) and molecular or atomic (m) masses,

$$\nu_m = \frac{h}{8\pi^2 (m_e m)^{1/2}}, \tag{4}$$

Since the liquid viscosity decreases with increasing temperature [following Eq. (1)] and the gas viscosity increases with increasing temperature, at sufficiently high temperatures the viscosity must pass through a minimum. Such a minimum has been observed in many inert elemental liquids and vapor, water, and CO_2 [1,13,14]. For the liquid, the shear viscosity is connected to the high-frequency shear modulus $G_\propto$ by Maxwell's relation, $\eta = G_\propto \tau = c^2\rho\tau$, where τ is the structural relaxation time and c is the transverse sound velocity. Assuming that the minimum viscosity is attained when the mean free path becomes comparable to the atomic separation a (the Mott-Ioffe-Regel limit [15]), and approximating $c = a/\tau$ and $\tau = 2\pi/w_D$, where w_D is the Debye frequency, the kinematic minimum viscosity for the liquid is

$$\nu_m = \eta/\rho = a^2/\tau = a^2 w_D/2\pi, \tag{5}$$

The kinetic theory of gases predicts that $\eta = \rho v L/3$ for the gas viscosity, where v and L are, respectively, the average velocity and the mean free path of the molecules. Then, taking $L = a$, and $\tau = a/v = 2\pi/w_D$, the kinematic minimum viscosity for the gas is expressed as

$$\nu_m = \eta/\rho = a^2 w_D/6\pi. \tag{6}$$

Equations (5) and (6) differ by a numerical factor of 1/3. From the relation between the cohesive energy E_{coh} and the Debye frequency of solids, $w_D = (E_{\mathrm{coh}}/ma^2)^{1/2}$, Eq. (5) takes the final form

$$\eta/\rho = a/2\pi (E_{\mathrm{coh}}/m)^{1/2}. \tag{7}$$

Equation (4) was obtained when E_{coh} was approximated by the Rydberg energy. TB claimed fair agreement between the experimental data and the theoretical predictions for many elemental inert gas liquids, as well as water, CO_2 and CH_4. To the best of our knowledge, we are not aware of any such study on metallic liquids. The main focus of this investigation is, therefore, to evaluate the experimental data for metallic liquids in terms of the three theories mentioned previously [1,10,13].

II. EXPERIMENTS

The temperature-dependent viscosities of 32 metallic alloy liquids were measured over a few hundred degrees above T_l in the equilibrium and deeply supercooled metastable liquids below T_l, until crystallization. Measurements on levitated (container-free) liquids under high vacuum ($\sim 10^{-7}$ Torr) using the electrostatic levitation technique [16,17] allow the liquid to be probed into the supercooled state in a contamination-free environment. For the present discussion, however, only viscosities far above T_l are of interest for the reasons already outlined. The viscosities were determined using the established oscillation technique [18], where near-resonant frequency oscillations are induced in, typically 50–60 mg, levitated liquid droplets by applying a sinusoidal electric field. The exponential decay time τ_{dec} of the induced oscillations is related to the viscosity by [18]

$$\eta = \frac{\rho R}{(l-1)(2l+1)\tau_{\mathrm{dec}}}, \tag{8}$$

where R is the sample radius and l the order of the harmonic, which is 2 under the experimental conditions. The sample radius and its temperature dependence were measured from two-dimensional video images of the liquid drops to evaluate density and viscosity.

III. RESULTS AND DISCUSSION

Before presenting the experimental data, it should be noted that the minimum viscosity for the metallic liquids cannot be

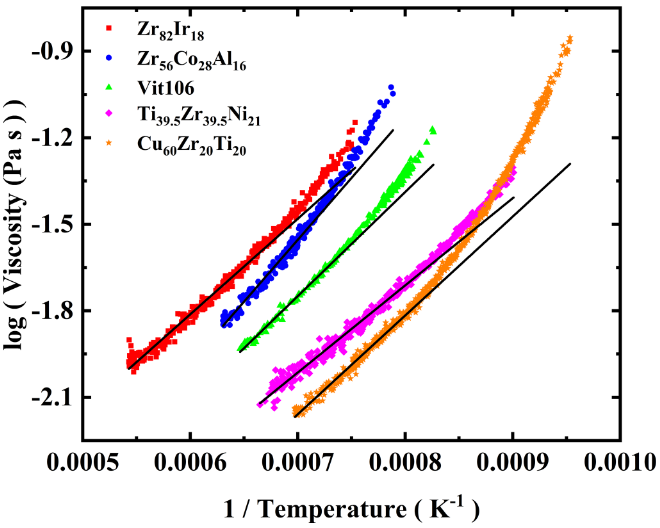

FIG. 1. Viscosities at high temperatures for a few representative liquids. The lines are Arrhenius fits to the experimental data above T_A, which is the temperature at which deviation from Arrhenius fits starts.

measured directly in most cases because such minima appear at extremely high temperatures where measurements cannot be performed due to excessive evaporative mass losses from the liquids. In principle, such measurements may be possible under high pressure, but difficult to do so because of the reactivity of most metals with containers at such high temperatures; containerless technique, such as the one used here, has not been developed for such high pressures. Therefore, the prefactor in viscosity, η_o, from the highest-temperature experimental data is taken as an approximate measure of the minimum viscosity.

To determine η_o from the experimental data over a finite temperature interval, a knowledge of the temperature- and material-dependent parameter $E(T)$ is necessary. Interestingly, above a characteristic crossover temperature T_A, $E(T)$ becomes approximately temperature independent and Eq. (1) becomes effectively Arrhenius. From both experimental and molecular dynamics simulation studies, T_A has been identified as the temperature below which the liquid dynamics becomes cooperative [19–22]. Therefore, η_o can be determined from fits of the experimental data to Eq. (1) above T_A, as shown in Fig. 1; if Eq. (1) is not strictly Arrhenius even at such high temperatures as reported in [23], the possible consequence of this would be an underestimation of η_o. This, and the previous approximation (taking η_0 as a measure of minimum viscosity), may make the present estimates smaller, but unlikely to be larger than the minimum viscosity.

We have investigated possible consequences of those approximations to the present results using available kinematic viscosity data over an extended temperature range from the NIST database [14] for four *representative* liquids and gases with different types of chemical interactions. Distinct minima in the kinematic viscosity were observed for those liquids under pressure in the supercritical states. We selected two inert systems, helium and argon (van der Waals bond) under 100 MPa pressure, water (hydrogen bond) under 100 MPa pressure, and methane (organic, covalent bond) under 20 MPa pressure. The viscosity data were analyzed from T_l up to temperatures where the temperature dependence of viscosity deviated significantly from Arrhenius-type behavior. The upper limits for the Arrhenius-type relation were found to be about 26 K (helium), 200 K (argon), 370 K (water), and 200 K (methane). The minimum viscosities for the corresponding liquids were observed near 56, 250, 710, and 250 K, respectively. The η_o and $\eta_{\min}$ for these liquids are 2.01×10^{-8} and 7.4×10^{-8} m^2/s for He, 3.4×10^{-8} and 7.7×10^{-8} m^2/s for Ar, 0.58×10^{-8} and 12.1×10^{-8} m^2/s for H_2O, and 3.83×10^{-8} and 11.0×10^{-8} m^2/s for CH_4. Therefore $\eta_{\min}$ are larger than η_o by factors of 3.7, 2.26, 21, and 2.9, respectively for He, Ar, H_2O, and CH_4. The ratio for $\eta_{\min}/\eta_o$ is anomalously large for H_2O, most likely because of its known anomalous properties, such as a maximum in the density with temperature. Assuming similar behavior for the metallic liquids, $\eta_{\min}$ may, therefore, be larger than η_o by a factor of 2−4 if we exclude H_2O and about 7 if we take an average for all four liquids. To keep the narrative simple, we will consider the upper limit of 4 as the appropriate correction factor for all metallic liquids studied in the present investigation.

The number density n is required to make a comparison with Eyring's relation, which can be determined from the experimental density data. While, to be precise, the number density at very high temperature is needed, a useful approximation is the value at T_l. Since the thermal expansion coefficient of these liquids is of the order of 10^{-5} [24], if the minimum appears at a temperature that is 1000 K above T_l, this may introduce an error of a few percent in n, which is much smaller compared to the effects of other approximations to the experimental data, as mentioned above. Figure 2 shows the ratios of the experimentally determined values of η_o and estimates from Eyring's relation as a function of fragility, where the fragility is defined as T_A/T_g [19,21]. It should be noted that this measure of fragility differs from the conventional fragility parameter introduced by Angell [25], $m = d\log\eta/d(T_g/T)$ at T_g, which measures how fast or slow the viscosity changes with temperature as T_g is approached. Liquids with smaller m values are termed *strong* and larger ones as *fragile.* Instead, the present measure of fragility was used since many of the liquids examined are marginal or poor glass formers; making measurements of traditional fragility parameters is difficult because of rapid crystallization near $T \geqslant T_g$. According to this scheme, T_A/T_g is larger for *stronger* liquids and smaller for *fragile* liquids, just the opposite of m. It has been demonstrated [19,22] that the two measures of fragility are equivalent, especially when relative changes in fragilities are being considered. It is relevant to add that deviation from Arrhenius-type behavior is much stronger for the fragile liquids. Therefore, the difference between η_o and $\eta_{\min}$ is expected to be different for different liquids, with larger differences for the more fragile liquids.

All relevant data used here and later for comparison with other theories are presented in Table I. Important points to note from comparison with Eyring's estimate and experimental data (Fig. 2 and Table I) are (a) most of the experimental values of η_o are larger (ratio is larger) or smaller (ratio is smaller) than Eyring's estimate (nh), up to an order of magnitude, with only a few in agreement; (b) the value

TABLE I. Experimental data and the theoretical estimates of the minimum viscosities of 32 metallic liquids. T_g and T_A are the experimental glass transition and crossover temperatures. T_gs with asterisks are estimated values using a method described in Ref. [31] since those liquids either do not form a glass by conventional melt-quench technique, or when they do, there is no distinct feature in the DSC data near T_g. TB1 and TB2 are the kinematic viscosities, η_o/ρ, estimated from Eqs. (4) and (7), respectively.

Alloy number	Alloy composition	T_g (K)	T_A (K)	η_o (10^{-5} Pa s) Experiment	η_o (10^{-5} Pa s) Eyring	η_o/ρ (10^{-8} m^2 s^{-1}) Experiment	η_o/ρ (TB1) (10^{-8} m^2 s^{-1})	η_o/ρ (TB2) (10^{-8} m^2 s^{-1})
1	$Cu_{64}Zr_{36}$	739	1249	1.03	4.07	0.133	2.52	5.41
2	Vit101 ($Ti_{34}Zr_{11}Cu_{47}Ni_8$)	685	1190	2.2	4.35	0.332	2.77	5.99
3	$Cu_{47}Zr_{47}Al_6$	695	1222	2.0	3.67	0.293	2.50	5.91
4	$Cu_{60}Zr_{20}Ti_{20}$	693	1219	2.76	4.20	0.397	2.66	5.63
5	$Cu_{50}Zr_{50}$	677	1196	3.2	3.67	0.450	2.45	5.84
6	$Cu_{50}Zr_{45}Al_5$	695	1241	3.66	3.71	0.530	2.51	5.85
7	$Cu_{50}Zr_{42.5}Ti_{7.5}$	669	1181	4.0	3.72	0.579	2.51	5.85
8	$Cu_{46}Zr_{54}$	646	1128	5.4	3.54	0.775	2.44	5.92
9	$Ti_{40}Zr_{10}Cu_{30}Pd_{20}$	670	1235	3.23	3.98	0.472	2.61	5.98
10	$Cu_{50}Zr_{40}Ti_{10}$	661	1218	3.24	3.75	0.472	2.52	5.85
11	$Cu_{43}Zr_{45}Al_{12}$	724	1352	4.07	3.65	0.622	2.55	6.00
12	$Zr_{64}Ni_{25}Al_{11}$	681	1290	2.68	3.34	0.421	2.47	6.63
13	$Zr_{56}Co_{28}Al_{16}$	738	1420	2.8	3.49	0.446	2.54	6.51
14	Vit105 ($Zr_{52.5}Cu_{17.9}Ni_{14.6}Al_{10}Ti_5$)	672	1305	2.5	3.52	0.388	2.53	6.24
15	$Ti_{40}Zr_{10}Cu_{36}Pd_{14}$	642	1239	2.36	4.04	0.353	2.66	6.06
16	Vit106a ($Zr_{58.5}Nb_{2.8}Cu_{15.6}Ni_{12.8}Al_{10.3}$)	668	1310	2.96	3.41	0.455	2.47	6.27
17	$Zr_{60}Ni_{25}Al_{15}$	688	1355	3.07	3.39	0.492	2.52	6.68
18	$Zr_{65}Ni_{10}Cu_{17.5}Al_{7.5}$	635	1230	3.7	3.31	0.570	2.44	6.28
19	$Cu_{47}Zr_{45}Al_8$	677	1335	5.0	3.70	0.737	2.52	5.91
20	$Zr_{70}Pd_{30}$	646*	1285	5.2	3.12	0.694	2.20	6.27
21	Vit106 ($Zr_{57}Nb_5Cu_{15.4}Ni_{12.6}Al_{10}$)	671	1349	5.28	3.41	0.807	2.47	6.30
22	$Zr_{59}Ti_3Ni_8Cu_{20}Al_{10}$	653	1282	6.9	3.39	1.078	2.49	6.28
23	$Zr_{75.5}Pd_{24.5}$	641*	1260	8.6	3.04	1.19	2.21	6.33
24	LM601 ($Zr_{51}Cu_{36}Al_9Ni_4$)	694	1320	10	3.58	1.50	2.51	6.04
25	$Zr_{74}Rh_{26}$	677*	1345	8.9	3.12	1.21	2.22	6.43
26	$Zr_{76}Ni_{24}$	585*	1170	7.6	3.21	1.13	2.36	6.47
27	$Zr_{62}Ni_8Cu_{20}Al_{10}$	655	1314	7.4	3.35	1.15	2.46	6.24
28	$Zr_{64}Ni_{36}$	623*	1265	8.7	3.44	1.27	2.42	6.43
29	$Ti_{39.5}Zr_{39.5}Ni_{21}$	584*	1223	7.4	3.51	1.25	2.63	6.61
30	$Y_{68.9}Co_{31.1}$	560*	1208	5.8	3.30	0.88	2.42	6.12
31	$Zr_{82}Ir_{18}$	682*	1495	15.8	2.97	1.94	2.06	6.20
32	$Zr_{80}Pt_{20}$	732*	1660	22	2.98	2.63	2.04	6.06

of η_o qualitatively tracks the fragility. Considering that the minimum viscosities may be 4 (or 7 in the extreme case) times larger than experimental η_o, all experimental data would become larger than Eyring's η_o by factors of 2–30. There may be other corrections necessary to Eyring's estimate as suggested recently [11], which will be discussed later.

For a consistent comparison with TB's theory, the kinematic viscosities are considered here instead of η_o. Focusing on the predictions by TB [13], Fig. 3(a) shows the ratios of the experimental data and estimates (TB1 in Table I) made using Eq. (4), where the Rydberg energy of atoms was chosen as the defining energy scale. Alternatively, the cohesive energy was used for theoretical estimates (TB2 in Table I) in Fig. 3(b), following Eq. (7). Interestingly, a rigorous estimate of the lower bound of viscosity from local uncertainty relations in Ref. [11] involves the variance of the local energy density. If the local energy density is bounded by the cohesive energy, it provides more theoretical justification for this choice. The cohesive energies were estimated from those of the elemental solids [26] using proper weighting factors [21,24] reflecting the compositions and the heats of mixing of the constituents from the binary alloys [27]. Since the cohesive energies of elemental liquids are usually smaller by about $10\% - 20\%$ from the corresponding solids [28], assuming similar differences for these alloys, the theoretical estimates of minimum viscosities may be smaller by only about 5% [square root of $E_{\rm coh}$ in Eq. (7)] than the data presented here. This is not significant considering that the other approximations made for the experimental data make much larger contributions.

Without applying any correction to η_o, there are some differences between the experimental data and the two theoretical estimates TB1 and TB2 (Fig. 3 and the last three columns of Table I). However, if a correction factor of 4 (or

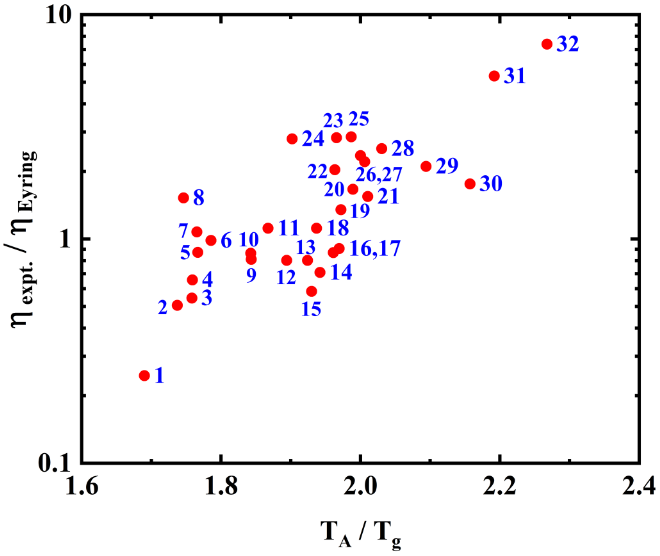

FIG. 2. The ratio of the experimental minimum viscosity and the estimated viscosity following Eyring's relationship for 32 metallic liquids as a function of fragility parameter (see text). The numbers corresponding to different alloy liquids are (see Table I), $Cu_{64}Zr_{36}$ (1), Vit101 (2), $Cu_{47}Zr_{47}Al_6$ (3), $Cu_{60}Zr_{20}Ti_{20}$ (4), $Cu_{50}Zr_{50}$ (5), $Cu_{50}Zr_{45}Al_5$ (6), $Cu_{50}Zr_{42.5}Ti_{7.5}$ (7), $Cu_{46}Zr_{54}$ (8), $Ti_{40}Zr_{10}Cu_{30}Pd_{20}$ (9), $Cu_{50}Zr_{40}Ti_{10}$ (10), $Cu_{43}Zr_{45}Al_{12}$ (11), $Zr_{64}Ni_{25}Al_{11}$ (12), $Zr_{56}Co_{28}Al_{16}$ (13), Vit105 (14), $Ti_{40}Zr_{10}Cu_{36}Pd_{14}$ (15), Vit106a (16), $Zr_{60}Ni_{25}Al_{15}$ (17), $Zr_{65}Ni_{10}Cu_{17.5}Al_{7.5}$ (18), $Cu_{47}Zr_{45}Al_8$ (19), $Zr_{70}Pd_{30}$ (20), Vit106 (21), $Zr_{59}Ti_3Ni_8Cu_{20}Al_{10}$ (22), $Zr_{75.5}Pd_{24.5}$ (23), LM601 (24), $Zr_{76}Rh_{24}$ (25), $Zr_{76}Ni_{24}$ (26), $Zr_{62}Ni_8Cu_{20}Al_{10}$ (27), $Zr_{64}Ni_{36}$ (28), $Ti_{39.5}Zr_{39.5}Ni_{21}$ (29), $Y_{68.9}Co_{31.1}$ (30), $Zr_{82}Ir_{18}$ (31), and $Zr_{80}Pt_{20}$ (32).

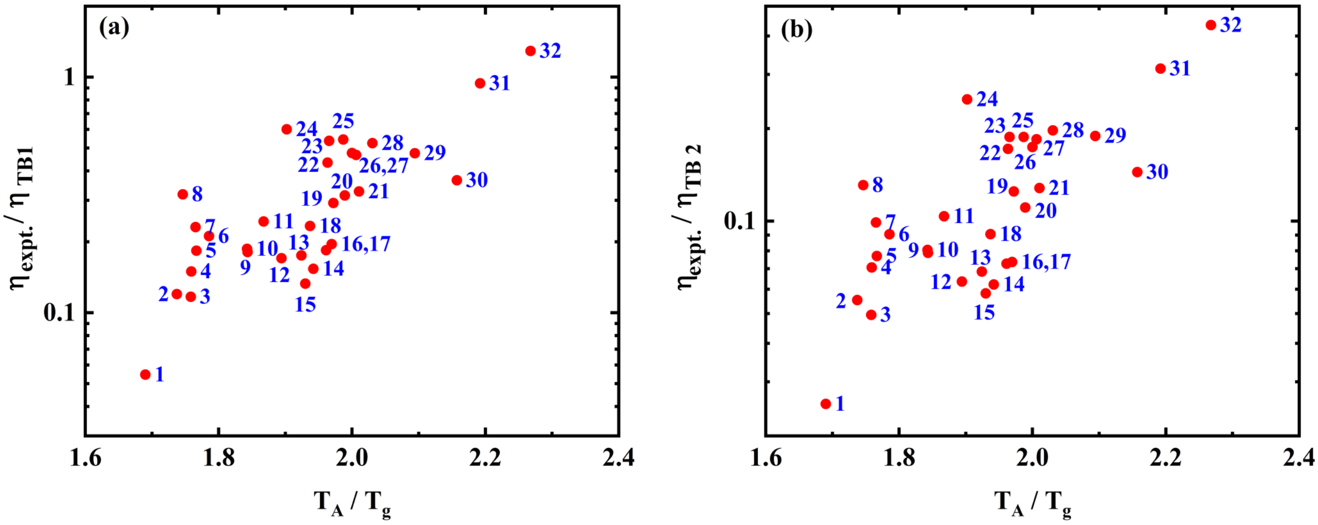

FIG. 3. The ratio of the experimental minimum viscosity and the estimated viscosity in (a) following Eq. (4) and (b) following Eq. (7) for 32 metallic liquids as a function of fragility parameter (see text). The numbers corresponding to different alloy liquid are (see Table I), $Cu_{64}Zr_{36}$ (1), Vit101 (2), $Cu_{47}Zr_{47}Al_6$ (3), $Cu_{60}Zr_{20}Ti_{20}$ (4), $Cu_{50}Zr_{50}$ (5), $Cu_{50}Zr_{45}Al_5$ (6), $Cu_{50}Zr_{42.5}Ti_{7.5}$ (7), $Cu_{46}Zr_{54}$ (8), $Ti_{40}Zr_{10}Cu_{30}Pd_{20}$ (9), $Cu_{50}Zr_{40}Ti_{10}$ (10), $Cu_{43}Zr_{45}Al_{11}$ (11), $Zr_{64}Ni_{25}Al_{11}$ (12), $Zr_{56}Co_{28}Al_{16}$ (13), Vit105 (14), $Ti_{40}Zr_{10}Cu_{36}Pd_{14}$ (15), Vit106a (16), $Zr_{60}Ni_{25}Al_{15}$ (17), $Zr_{65}Ni_{10}Cu_{17.5}Al_{7.5}$ (18), $Cu_{47}Zr_{45}Al_8$ (19), $Zr_{70}Pd_{30}$ (20), Vit106 (21), $Zr_{59}Ti_3Ni_8Cu_{20}Al_{10}$ (22), $Zr_{75.5}Pd_{24.5}$ (23), LM601 (24), $Zr_{76}Rh_{24}$ (25), $Zr_{76}Ni_{24}$ (26), $Zr_{62}Ni_8Cu_{20}Al_{10}$ (27), $Zr_{64}Ni_{36}$ (28), $Ti_{39.5}Zr_{39.5}Ni_{21}$ (29), $Y_{68.9}Co_{31.1}$ (30), $Zr_{82}Ir_{18}$ (31), and $Zr_{80}Pt_{20}$ (32).

7 in the extreme case) is applied to increase experimental η_o, they would be closer to estimates from TB1, which considers the Rydberg energy as the appropriate energy scale [13]. The TB1 estimates would lie within factors of 0.2 ($Cu_{64}Zr_{36}$) to 5 ($Zr_{80}Pt_{20}$, two extreme cases) to the corrected experimental data. Interestingly, the experimental minimum viscosities of a few elemental and molecular liquids in Ref. [13] are also *larger* than the theoretical estimates (TB1) by factors of 1.3–3; however, for He, the experimental values are about 2 times *smaller* under 20 MPa and 1.4 times *smaller* under 100 MPa pressure than TB1. Instead, if cohesive energies are used in the TB theory (TB2), the theoretical estimates would lie within factors of 0.1 and 1.7 to the corrected experimental η_o for the various liquids.

To make a comparison between the experimental data and the KSS estimate [Eq. (2)], the entropy density of the liquid is required. We have calculated the entropy density of two liquids, Vit 106 ($Zr_{57}Nb_5Cu_{15.4}Ni_{12.6}Al_{10}$) and $Ti_{39.5}Zr_{39.5}Ni_{21}$ from the measured specific heat. The former alloy is a bulk metallic glass former [29], and the latter crystallizes into an icosahedral quasicrystal [30]. It is to be noted that $Ti_{39.5}Zr_{39.5}Ni_{21}$ cannot be quenched into a glass even by fast cooling ($\sim 10^6$ K/s) melt-quench technique. The quoted T_g in Table I for this alloy is an estimated value [31] if it were at all possible to quench it into a glass. The specific heat was measured from 2 to 1500 K for Vit106 and from 2 to 1600 K for $Ti_{39.5}Zr_{39.5}Ni_{21}$ using three different techniques. The solid specific heat was measured below 373 K by a commercial Physical Properties Measurement System (Quantum Design, San Diego, USA), and by a commercial differential scanning calorimeter (DSC, PerkinElmer, USA, model 7) for the higher temperatures. Data for the equilibrium and supercooled liquids were obtained using the modulation calorimetry technique [32] on electromagnetically levitated liquid droplets on the International Space Station. In both cases, no experimental data could be obtained in the supercooled liquids over about 200 K due to rapid crystallizations.

As shown in Fig. 4, the total entropy is 113 J mol^{-1} K^{-1} for Vit 106 up to the highest temperature of 1500 K and 89 J mol^{-1} K^{-1} for $Ti_{39.5}Zr_{39.5}Ni_{21}$ to 1600 K. Using the experimentally measured values for η_o (5.28×10^{-5} Pa s for Vit 106 and 7.4×10^{-5} Pa s for $Ti_{39.5}Zr_{39.5}Ni_{21}$) and the experimental molar volumes at T_l (11.7×10^{-6} for Vit106 and 11.4×10^{-6} m^3 for $Ti_{39.5}Zr_{39.5}Ni_{21}$), the ratio of the viscosity to entropy density is 5.4×10^{-12}~K s for Vit106 and 9.5×10^{-12}~K s for $Ti_{39.5}Zr_{39.5}Ni_{21}$. These values are, respectively, 9 and 16 times larger than the minimum estimates from Eq. (2); the data for Vit 106 are surprisingly close to those of liquid He [1]. However, if a correction factor of 4 is applied to η_o to bring it closer to $\eta_{\min}$ and the entropy density is extrapolated to even higher temperatures, the experimental data for the two liquids would likely exceed the KSS limit by about two orders of magnitude, but still remain smaller than that for water (340 times larger [1]).

IV. CONCLUSIONS

After a factor of 4 correction applied to the experimental data, they are larger than Eyring's estimate for the viscosity prefactor η_o by factors of 2–30. The important point to note, however, is that Eyring's estimate for η_o ($= nh$) nonetheless continues to be a lower bound on the viscosity. This bound is indeed anticipated if, at the point where the viscosity minimum occurs, the ionic mean free path is larger than its de Broglie wavelength [11], which is empirically expected to hold for conventional fluids. More general rigorous bounds require further incorporation of the thermal equilibrium moments involving the system interactions and other additional numerical factors (including those associated with the replacement of h by quantum uncertainty derived inequalities) that render the value of the resulting lower bound smaller than nh [11]. With those considerations, the estimates are about one to two orders of magnitude (see Table I) smaller than the experimental data. This may suggest that a material specific correction factor is needed to reconcile theory with experiment. However, what that factor would be is not clear at the moment. TB's estimates appear to be much better when possible corrections (about a factor of 4) are applied to the experimental η_o. With this correction, TB1 estimates are, at the most, a factor of $0.2-5$ times larger than the experimental data. TB2 estimates are slightly better; they are in agreement with the experimental data within a factor of $0.1 - 1.7$.

For the two liquids where specific heat and entropy data are available, the KSS prediction appears reasonable. While data for more metallic liquids are required to make a critical assessment, since the molar entropy and molar volumes are unlikely to change much among metallic liquids (a factor of 2 at most), they will likely be similar to what is reported here for these two liquids. It then appears that the minimum viscosities of classical metallic liquids are about one to two orders of magnitude larger than the KSS estimates [1], but smaller than that of water.

When the limitations of the experimental data are kept in mind, the estimates from all three disparate theories appear to be roughly in agreement with the experimental data within one to two orders of magnitude. Implications of these

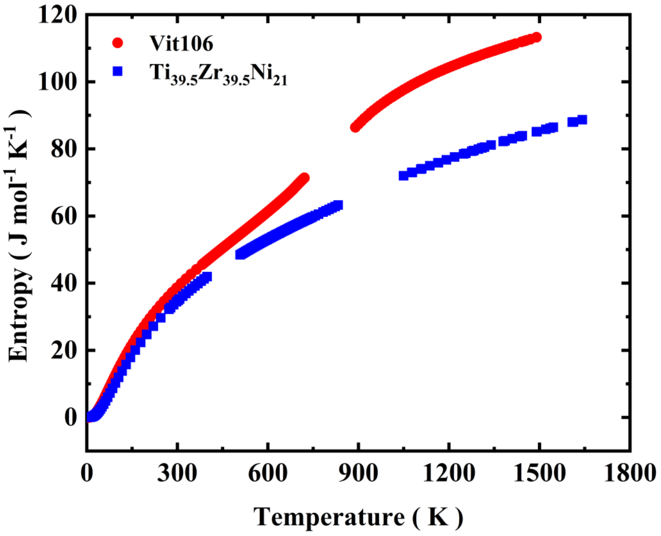

FIG. 4. Calculated entropies of Vit 106 ($Zr_{57}Nb_5Cu_{15.4}Ni_{12.6}Al_{10}$) and $Ti_{39.5}Zr_{39.5}Ni_{21}$ solids and liquids from the specific heat data.

results will remain under future considerations. A connection between minimum viscosity and cohesive energy appears to be consistent with some theoretical [11] and experimental [21] reports. Since a semiquantitative relation between cohesive energy and fragility was reported earlier [21], the relation between minimum viscosity and cohesive energy is consistent with those results. Although not a central point of discussion, the observed qualitative correlation between fragility and minimum viscosity is still somewhat surprising since η_o is an extreme high-temperature property and fragility is usually defined near T_g, or in this case near T_l (T_A is usually close to T_l [19,22]).

ACKNOWLEDGMENTS

A.K.G. and K.F.K. are supported by NASA under Grants No. NNX21AU19G and No. 80NSSC21K1649. Any opinions, findings, and conclusions or recommendations expressed in this material are those of the author(s) and do not necessarily reflect the views of NASA.

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