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Trends in the temperature dependence of dynamical heterogeneity in strong and fragile supercooled liquids

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ABSTRACT

The temperature dependence of the Kohlrausch-Williams-Watts (KWW) stretching exponent β of shear relaxation kinetics is determined for a wide variety of deeply supercooled glass-forming liquids with fragility index m ranging between ~ 30 and 90 using small amplitude oscillatory shear parallel plate rheometry. Intriguingly, and contrary to conventional wisdom, β and m are observed to be uncorrelated at temperatures close to the glass transition T_g. However, a clear pattern emerges when the variation of β is considered as a function of the α-relaxation timescale τ_α. In particular, β is observed to increase rapidly (slowly) for relatively strong (fragile) liquids with decreasing τ_α upon increasing temperature above T_g. Consequently $d\beta/d(\log \tau_\alpha)$ is found to be negatively correlated with m near and above T_g. A possible origin of this intriguing trend is discussed within the frameworks of the energy landscape and the elastic facilitation models of relaxation of supercooled liquids.

1. Introduction

From windows and containers to photonics and telecommunication, innovative developments in glass science and technology have been key enablers of civilization throughout history [1-4]. The compositional and processing engineering of these functional glasses require a fundamental understanding of the underlying relationship between the atomic structure, chemistry and the dynamical processes in the parent supercooled liquids from which these glasses are derived. The transport properties such as diffusivity and viscosity of deeply supercooled liquids frequently display a non-Arrhenius behavior with the corresponding activation energy being dependent on temperature [5]. The degree of non-Arrhenius behavior of viscosity η of a supercooled liquid is often parameterized in the literature by the fragility index m proposed by Angell, which is defined as: $m = \frac{d(\log_{10}\eta)}{d(T_g/T)}|_{T=T_g}$, where T_g represents the glass transition temperature [6]. The unitless parameter m has been shown to be useful for classification of a wide range of supercooled liquids into “strong” and “fragile”, the former (latter) being characterized by a low (high) value of m associated with a weaker (stronger) departure from the Arrhenius behavior. Typically, network glass-forming liquids such as SiO_2 and GeO_2 show strong behavior with m as low as ~ 18–20 [6]. On the other hand, many molecular and polymeric glass-forming liquids display fragile behavior with m well above 100 [7].

In addition to the non-Arrhenius behavior of viscosity, the primary- or α- relaxation of deeply supercooled liquids approaching glass transition, as measured using a wide variety of techniques including dielectric, mechanical or heat capacity spectroscopy and dynamic light scattering, typically exhibits a non-exponential decay of the relaxation function [8-11]. This decay can be described empirically using a stretched exponential function $\exp\left[-\left(\frac{t}{\tau_K}\right)^\beta\right]$, which was originally proposed independently by Kohlrausch and by Williams and Watts (KWW) [12,13]. In this expression τ_K is the KWW relaxation timescale and $0 \leq \beta \leq 1$ is the stretching exponent. While the atomistic origin of a stretched exponential relaxation in deeply supercooled liquids remains a highly debated topic in the literature, it is now well-established that the stretching results from a spatio-temporally *heterogeneous* relaxation with a distribution of relaxation times τ. In this case the stretching parameter β is a measure of the width of this distribution, where a lower value of β corresponds to a wider distribution and vice versa [14,15].

A question that naturally arises in light of the discussion above is whether m and β are related in any fundamental way, which may be central in our understanding of the physics of glass transition. It is particularly interesting therefore to note that nearly three decades ago Böhmer et al. reported an interesting observation of a negative correlation between m and β in a wide variety of supercooled glass-forming

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liquids near glass transition [16]. This correlation has recently been challenged by Dyre, Blochowicz and coworkers, who analyzed broadband dielectric spectroscopy data for more than 50 molecular liquids to show that an overwhelming majority of these liquids display no correlation between m and β; instead β for these liquids with a relatively wide range of m is ~ 0.5 near T_g [17,18].

However, the systematics of the temperature dependence of β in supercooled liquids with a wide range of fragility index m remains poorly known in the literature to date. Here we report the temperature dependence of β for shear relaxation in a variety of inorganic glass-forming liquids with a wide range of fragility index m, using shear mechanical spectroscopy at temperatures in the glass transition range. Our results, in combination with the data previously reported in the literature, show intriguing patterns in the dependence of β on the primary or α -relaxation timescale τ_α as a function of m.

2. Experimental details

2.1. Glass synthesis

All glass compositions investigated in this study were prepared via conventional melt quenching. The highly fragile ($m \sim 90$) ionic glass $Ca_{0.4}K_{0.6}NO_3$ (CKN) was prepared by melting appropriate mixture of the as-received high-purity $Ca(NO_3)_2{\cdot}4H_2O$ and KNO_3 chemicals in an evacuated and sealed borosilicate glass tube following the procedure outlined by Luo et al. [19]. The mixture was heated slowly to 523 K and the resulting melt was held there for 48 h before quenching. The tube was then transferred to a glovebox and cut open to recover the CKN glass, which was stored in the glovebox until further use for rheometry experiments. The network glasses included a highly fragile composition ($[3Li_2O\text{-}2Na_2O\text{-}3K_2O]_{40}\text{-}[WO_3]_{60}$, $m \sim 90$), two rather strong ($m \sim$ 30–32) compositions (As_2S_3 and As_3Se_7), one moderately strong composition (B_2O_3; $m \sim 40$) and one moderately fragile composition ($[PbO]_{60}[SiO_2]_{40}$; $m \sim 57$) [20-22]. The As_2S_3 and As_3Se_7 glasses were prepared from the constituent elements ($\geq$99.999 % purity, metal basis), that were loaded into quartz ampoules that were evacuated to 10^{-4} Torr and sealed. The ampoules were then loaded in a rocking furnace and melted at 650 °C for 24 h to ensure melt homogeneity [23,24]. The melts were subsequently quenched to form glass by dipping the ampoules in water. The Pb-silicate and B_2O_3 glasses were prepared in platinum crucibles, while the Li,Na,K-tungstate glass (designated, henceforth, as ($[R_2O]_{40}\text{-}[WO_3]_{60}$; R= Li,Na,K) was prepared in a silica crucible. Details of their synthesis procedure can be found in our previous publications [20,21,25]. The thermophysical properties of B_2O_3 are known to be critically dependent on the structural water content. Therefore, special precaution was taken in preparing a dry B_2O_3 glass [21]. The high-purity oxide reagent was calcined at 773 K for 24 h in a Pt crucible and subsequently melted at 1473 K for 24 h followed by quenching in air. Following their synthesis all glasses were promptly transferred to and stored in a glovebox to avoid exposure to atmospheric moisture until they were used for parallel plate rheometry.

2.2. Shear-mechanical spectroscopy

All shear-mechanical spectroscopic measurements were carried out on an Anton-Paar MCR 302 parallel plate rheometer equipped with a convection oven (up to 600 °C) under constant nitrogen gas flow. All samples were first heated inside the oven above their softening point, then trimmed into a disc geometry with $\sim$ 1 mm thickness, which was sandwiched between the oscillating upper plate (4 mm diameter) and the stationary lower plate. At each temperature small-amplitude oscillatory shear measurements were carried out by applying an oscillatory strain within the linear viscoelastic region with angular frequency ω varying between 0.01 and 600 rad/s, and the induced torque was recorded to obtain the storage and loss moduli G′and G″as a function of ω (Fig. 1). The α -relaxation timescale τ_α was determined at each temperature from the frequency location ω_{max} of the maximum in the loss spectrum G(ω) using the relation: $\tau_\alpha = (2\pi / \omega_{max})$.

3. Results and discussion

The shear relaxation behavior of supercooled liquids and glasses is described well in the time domain by the KWW stretched exponential function such that: $G(t) = G_\infty \exp\left[-\left(\frac{t}{\tau_K}\right)^\beta\right]$ where G_∞represents the glassy modulus. Therefore, the loss spectra G(ω) obtained in this study are fitted in the frequency domain with the Fourier Transform of the same function to obtain the β values [26]. It may be noted that any change in β with temperature is expected to result in a violation of time-temperature superposition (TTS) in the form of a change in the width of the loss peak in the G(ω) spectra as well as a change in the slope on the high-frequency side of the peak. For example, an increase in β would result in a narrowing of the G(ω) peak and a steepening of its high-frequency slope. The instrumental limitation of frequency ω of the shear mechanical spectroscopic technique allows for the determination of G′(ω)-G(ω) data suitable for obtaining β over a rather limited range of τ_α between 6×10^{-2} s and 10^2s, as shown in Fig. 1. The validity of TTS in these data is tested by the standard procedure of shifting the normalized G(ω) loss spectra along the frequency axis to check for superposition as shown in Fig. 2. It is clear from Fig. 2 that within this relatively narrow range of τ_α the TTS is indeed obeyed by the moderately and highly fragile liquids such as Pb-silicate ($m \sim 57$) and CKN ($m \sim 90$), while the strongest liquids As_2S_3 and As_3Se_7 ($m \sim 30$) show a clear violation of TTS. On the other hand, the moderately strong liquid B_2O_3 ($m \sim 40$) shows a rather subtle violation of TTS. However, in all cases of violation of TTS, a narrowing of the G(ω) peak and a steepening of its high-frequency slope (Fig. 2) are observed with increasing temperature indicating an increase in β with decreasing τ_α. The τ_α dependence of β values as obtained from the fits of the KWW relation to the G(ω) data (Fig. 2) are shown in Fig. 3. The β values for all liquids vary within a relatively narrow range of $\sim$ 0.45–0.57 near T_g, where $\tau_\alpha \sim 10^2$ s. It is important to note that the β values thus obtained for B_2O_3 and CKN liquids are in good agreement with those obtained in the time domain by Sidebottom and coworkers using photon correlation spectroscopy [27, 28]. At temperatures above T_g the strong liquids with $m \leq 40$ display an approximately linear increase in β with decreasing log τ_α, while β remains nearly unchanged for relatively fragile liquids over the entire range of τ_α accessible to the shear mechanical spectroscopic technique utilized in the present study (Fig. 3). The quantity $d\beta/d(\log \tau_\alpha)$ is found to be negatively correlated with m for liquids with $m \leq 40$, and to be zero for liquids with $m \geq 50$ in this range of τ_α near and above T_g (Fig. 4).

It is noteworthy that the lack of any significant temperature dependence of β observed for the relatively fragile liquids with $50 \leq m \leq 90$ studied here, is completely consistent with the results reported by Dyre and coworkers for several simple organic molecular liquids characterized by similar m values [17,18,29]. The mechanical loss spectra of these liquids were shown to obey TTS, i.e. the β for these liquids were found to be temperature-independent, over a large variation in the α-relaxation timescale ranging from $\sim 10^2$ s to up to $\sim 10^{-3}$ s [29]. Our results are also in good qualitative agreement with the β (T) data for the binary $Na_2O\text{-}GeO_2$ liquids, obtained using dynamical light scattering in a recent systematic study by Sidebottom, over a wide τ_α range from $\sim 10^1$ s to up to $\sim 10^{-5}$ s [30]. These β (T) data of Sidebottom are significantly noisier than the shear mechanical spectroscopic data in the present study. However, β was observed to increase approximately linearly with decreasing log τ_α for all liquids with $m \leq 47$. The corresponding slopes $d\beta/d(\log \tau_\alpha)$ were obtained from these data and are shown in Fig. 4 as a function of m, along with the results obtained in the present study. When taken together, these data provide an intriguing trend where $d\beta/d(\log \tau_\alpha) = 0$ for liquids with $m \geq 50$ and increases rapidly upon lowering of m down to a value of $\sim$ 17 for the GeO_2 liquid [30] characterized by a

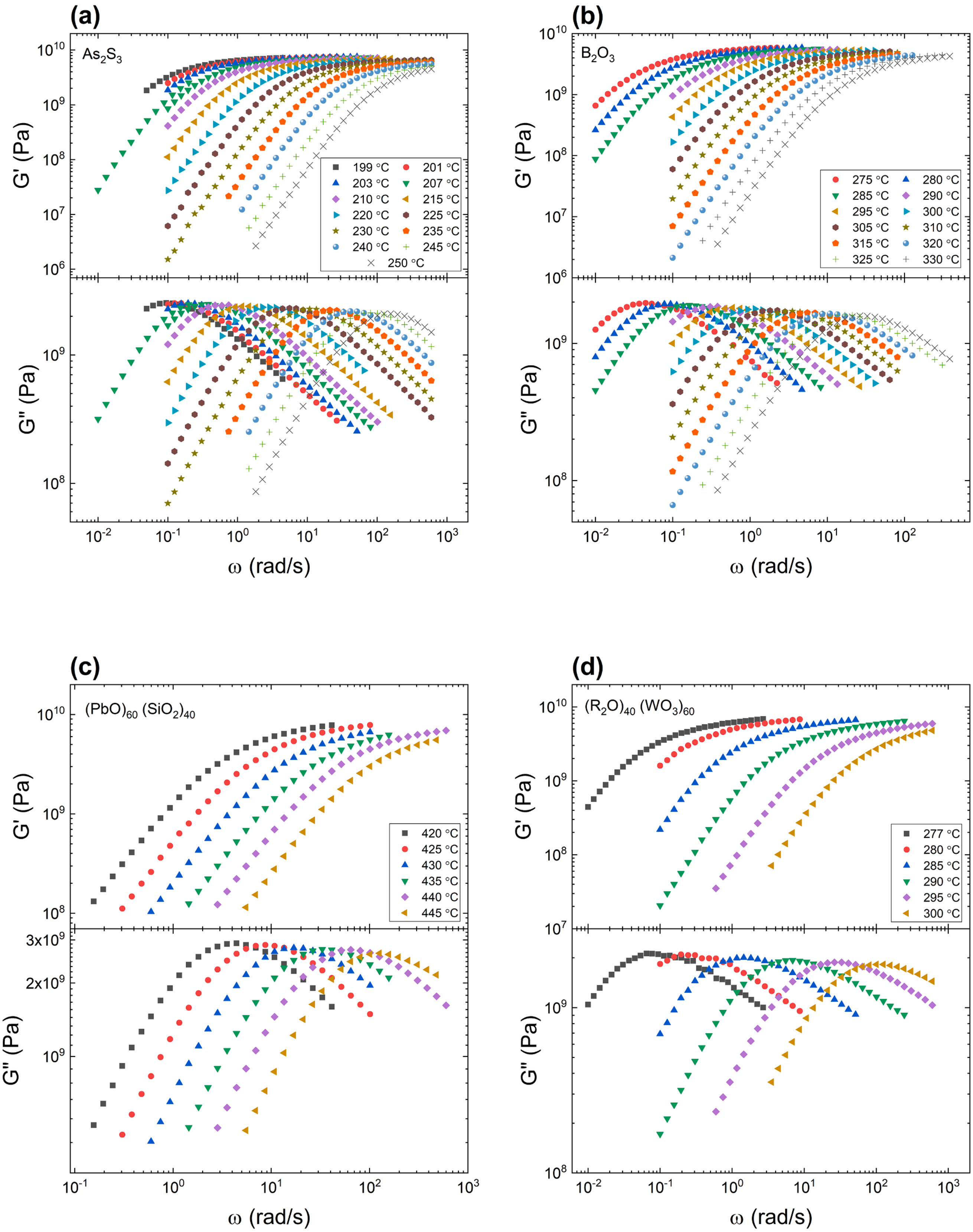

Fig. 1. Representative shear-mechanical spectra of (a) As_2S_3, (b) B_2O_3, (c) $([PbO]_{60}[SiO_2]_{40}$, and (d) $([R_2O]_{40}[WO_3]_{60}$ liquids with m ranging between ~ 30 and 90, at select temperatures showing frequency dependence of G′ (top panels) and G″ (bottom panels).

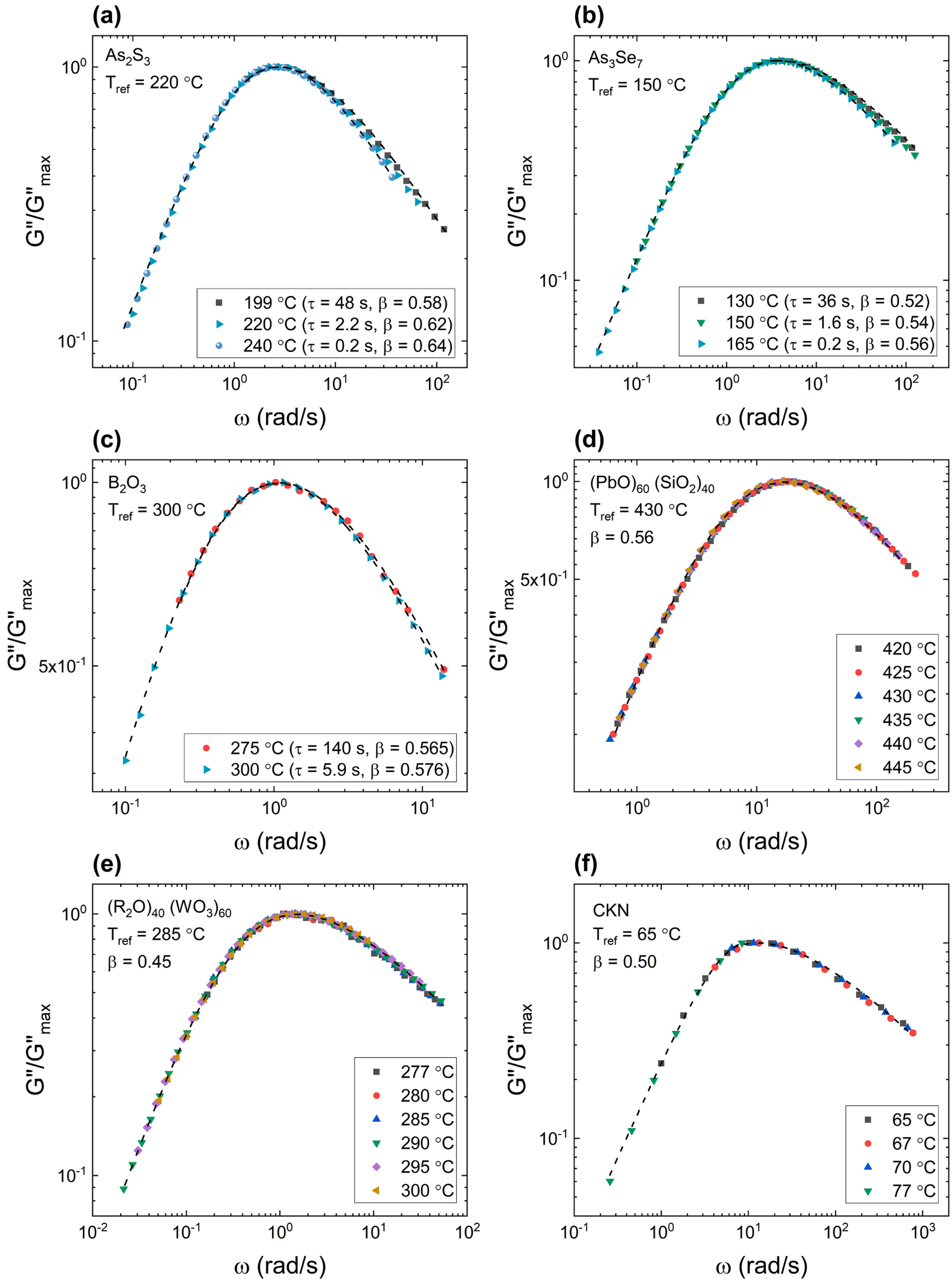

Fig. 2. G(ω) spectra normalized to the value at the maximum for all liquids investigated in the present study at various temperatures. For each liquid normalized spectra at different temperatures are shifted along the frequency axis to line up with the spectrum at the reference temperature T_{ref} (listed in the inset) to test the validity of TTS. Note clear violation of TTS in (a) and (b), subtle violation in (c) and no violation in (d)-(f). Dashed lines through the data points are representative fits of KWW equation (see text for details) at various temperatures. Corresponding values of τ_α and β are listed in the inset.

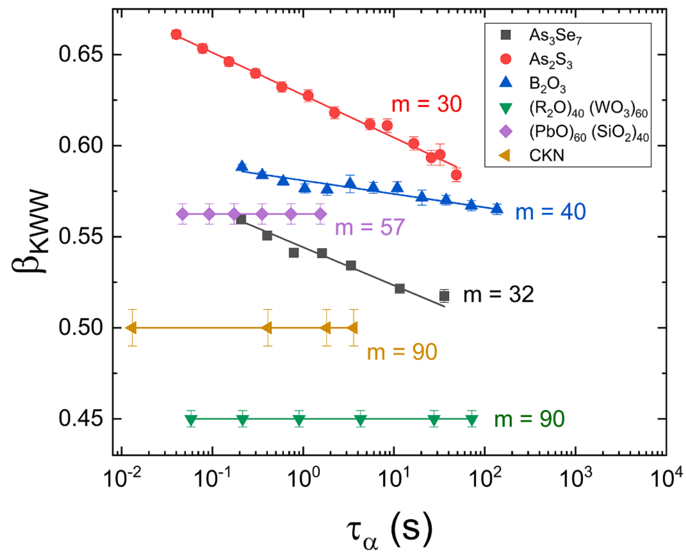

Fig. 3. Variation of β with τ_α, as obtained from fits of KWW equation to G(ω) spectra in Fig. 2, for all supercooled liquids investigated in the present study.

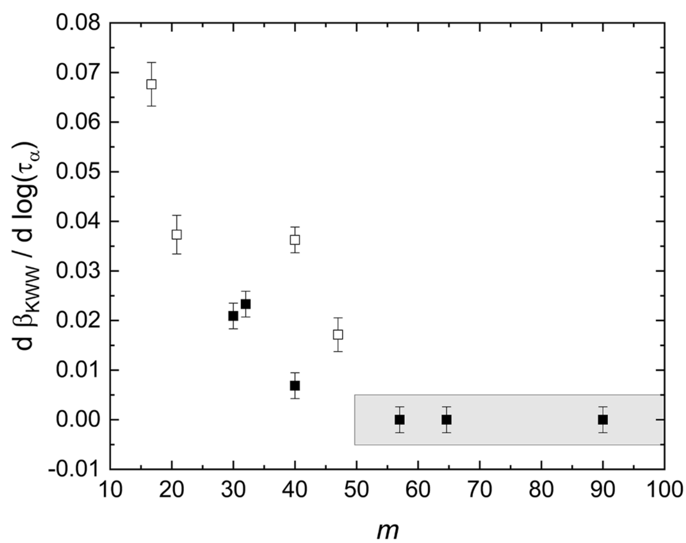

Fig. 4. *dβ/d(log τ_α)* obtained from analysis of G(ω) spectra for all supercooled liquids investigated in the present study (filled symbols) as a function of their fragility index *m*. Open symbols are for supercooled Na_2O-GeO_2 liquids from previous dynamical light scattering studies [30].

3-dimensional network of corner-shared $GeO_{4/2}$ tetrahedral units (Fig. 4). It may be noted at the outset that the observed variation of *dβ/d (log τ_α)* with *m* in Fig. 4 is in contradiction with the prediction of the coupling model of Ngai [31,32]. In this model a dynamical coupling between the neighboring cooperatively rearranging regions (CRR) is introduced to modify the original Adam-Gibbs configurational entropy model, as in the latter, the CRRs relax independently [33]. In Ngai's model the strength of this coupling n increases with decreasing temperature, where $n = 1-\beta$, and gives rise to a stretched exponential kinetics of the α-relaxation in conjunction with a non-Arrhenius temperature dependence of $\langle\tau\rangle$. The governing relations in this model give rise to a positive correlation between the fragility index *m* and the temperature dependence of β, while the data in Fig. 4 indicates that β is nearly independent of temperature for moderately and highly fragile liquids. Rather it is the strong liquids that are characterized by a pronounced temperature dependence of β !

As noted above, the data for fragile liquids in Fig. 4 are consistent with the observation of a number of recent studies based on dielectric and shear mechanical spectroscopy and light scattering [17,18,28-30], that the loss spectra of a variety of network, molecular and ionic liquids with $50 \le m \le 125$ obey TTS over a wide τ_α range ($\sim 10^2$ to10^{-5} s) with $\beta \sim 0.5$ i.e. G(ω) $\sim \omega^{-1/2}$ at frequencies higher than the loss peak frequency. This "generic" stretched exponential behavior of the α-relaxation has been hypothesized by Dyre to result from long wavelength fluctuations dominating the dynamics in deeply supercooled liquids [34]. It is clear from Fig. 4 that such a generic non-exponentiality of the α-relaxation in supercooled glass-forming liquids breaks down for strong liquids with $17 \le m \le 50$. It is important to note that previous dielectric relaxation measurements by Wang and Richert [35] on a variety of fragile molecular organic liquids with $m \ge 50$ indicated that while *dβ/d (log τ_α)* ~ 0 for these liquids when $10^{-6}\,s \le \tau_\alpha \le 10^2$ s, further lowering of τ_α with increasing temperature resulted in a rapid rise in β, which approached a value of $\sim$1 near $\tau_\alpha \sim 10^{-9}$ s. A similar temperature dependence of β was also reported for the fragile ionic liquid CKN ($m \sim 90$) by Sidebottom and Sorensen [28] in a compilation of data obtained using a variety of probes ranging from enthalpy relaxation and photon correlation spectroscopy to ultrasonic absorption, neutron spin echo and Brillouin scattering. It may be noted, however, that a direct comparison between the β values obtained by different spectroscopic techniques (e.g. mechanical, dielectric and dynamic light scattering) is complicated by their different degrees of sensitivity towards self- vs. cross- correlations in the dynamics [18]. Additionally, these techniques probe fluctuations in different properties associated with a dynamical process. For example, the dielectric loss spectra have recently been shown to contain contributions from both higher-frequency self-correlation and lower-frequency cross-correlation in molecular reorientation dynamics, whereas the mechanical spectra are dominated by the translational part of the α-relaxation dynamics [18]. This issue is not necessarily problematic if the temperature dependence of β and τ for the self and collective dynamics is the same. However, problems may arise if these dynamics decouple from each other with temperature. Nevertheless, keeping these caveats in mind, it is tempting to suggest that β will eventually have to reach unity for the α-relaxation of all supercooled liquids at some sufficiently high temperature where the activated jumps of the structure between minima in the potential energy landscape disappears and the relaxation becomes exponential [36]. When taken together, these results suggest a strong vs. fragile pattern of variation of β vs. τ_α, which is shown schematically in Fig. 5. While on lowering of temperature both strong and fragile liquids may have a crossover from a region of temperature dependent β to a temperature independent region, the τ_α for this crossover depends on *m*. Below we propose a possible

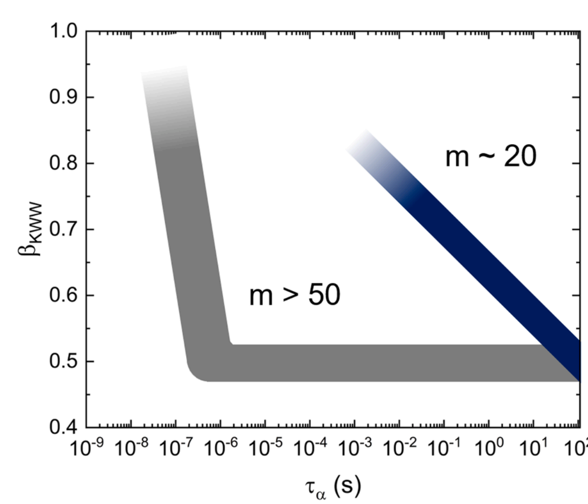

Fig. 5. Schematic representation of pattern of variation of β with τ_α for strong ($m \sim 20$) and fragile ($m > 50$) liquids.

scenario to qualitatively explain this pattern in Fig. 5 within the framework of the free-energy landscape model of dynamics of supercooled liquids.

In this model we consider the dynamics in two extreme cases: (i) for a very strong network liquid (m ~20) and (ii) for a rather fragile molecular liquid (m ~ 90). Previous high-temperature ^{29}Si and ^{17}O nuclear magnetic resonance spectroscopic studies on supercooled network liquids such as silicates indicated that the shear relaxation process in these liquids is controlled by local Si-O bond flipping [37]. In this case the local free-energy barrier for bond flipping also controls the global relaxation rate. In this scenario the dynamical heterogeneity arises from an elastic facilitation: a flow event triggered by a local bond flipping induces a long-range stress field via elastic interactions that facilitates another flow event nearby [38]. Cooling would result in a continuous lowering in the damping of these elastic interactions and a concomitant increase in the length scale of the stress field, which would lead to increased cooperativity. Consequently, one would expect β to decrease steadily with decreasing temperature in these liquids, as is indeed shown to be the case in the present study (Fig. 4). On the other hand, for a fragile molecular liquid the free-energy landscape is extremely rugged and temperature dependent, which gives rise to a highly cooperative dynamics with a broad distribution of timescales [39]. As a result, once a highly fragile liquid descends into this landscape upon cooling below the mode coupling temperature T_c, β rapidly decreases signifying caging of molecules by their neighbors that becomes increasingly effective on lowering of temperature. It may be noted here that Goldstein originally estimated the corresponding τ_α at T_c to be approximately 10^{-9} s [40]. At some stage further cooling results in a width of the barrier height distribution that is broader than the thermal energy k_BT, which causes τ_α to be controlled by the percolation of slow domains with a long-time cutoff [38,41]. At this point the elastic facilitation and long wavelength fluctuation dominated dynamics come into effect at frequencies higher than the loss-peak frequency, which results in the "generic" stretched exponential behavior of the α-relaxation with a temperature-independent $\beta \sim 0.5$ [34,38].

4. Conclusions

In summary, the stretching exponent β for shear relaxation in a wide variety of supercooled liquids shows distinct patterns in its temperature dependence as a function of the fragility index *m*. The value of β for moderately and highly fragile liquids with $m \geq 50$ appears to be nearly independent of temperature, i.e. TTS is obeyed, in the deeply supercooled regime where $10^{-5}\ s \leq \tau_\alpha \leq 10^2$ s. We speculate that β for these liquids becomes strongly temperature dependent, but only at relatively high temperatures where $\tau_\alpha < 10^{-6}$ s. In contrast, β steadily increases with temperature on heating above T_g for relatively strong liquids with $m \leq 40$, i.e. TTS is violated even in the deeply supercooled regime, and $d\beta/d(\log \tau_\alpha)$ is found to increase monotonically with lowering of *m*. This strong vs. fragile pattern of the temperature dependence of β is hypothesized to be linked to the free energy landscape of these liquids.

CRediT authorship contribution statement

Sabyasachi Sen: Writing – review & editing, Writing – original draft, Visualization, Validation, Resources, Project administration, Methodology, Funding acquisition, Formal analysis, Conceptualization. **Jacob Lovi:** Writing – review & editing, Visualization, Methodology, Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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