Relating the Poisson's ratio to molecular dynamics of glass-formers

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ABSTRACT

Despite the importance of Poisson's ratio in materials science and engineering, its connection to molecular dynamics and relaxation processes in glass-forming systems remains unclear. The existence of such connection made the family of metallic glasses in our previous study [K.L. Ngai et al. J. Chem. Phys. 140 (2014) 044511], where it was also justified by molecular dynamics simulation, physics of glass-forming materials, and theoretical considerations. Here we extend the consideration to molecular glass-formers and amorphous polymers, and demonstrate that the Poisson's ratio correlates with several microscopic dynamic properties, and explained theoretically in exactly the same way as given in our previous publication.

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1. Introduction

The dynamics of molecular liquids and the relation to glass transition are still unsolved problems in condensed matter physics. Despite the intensive research in the past decades by various experimental techniques and theoretical approaches, there is still no consensus on the key issue, i.e. the fundamental physics that govern the dynamics and explain the salient experimental facts. In this state of affair, the search of strong correlations between different aspects or properties of the dynamics is a worthwhile undertaking. Success in establishing such strong correlations can provide deeper insight into the problem, and clues for possible breakthrough in solving the problem.

There are many outstanding aspects of the dynamics of glass-forming molecular liquids found all over the spectrum from THz phonon frequencies down to 10^{-6} Hz by quasielastic neutron scattering and dynamic light scattering, nuclear magnetic resonance, dielectric and mechanical relaxation and other techniques. Examples include (1) the intensity of the Boson peak measured by the ratio between the maximum and the minimum of the peak in the Raman- and neutron-scattering spectra [1]; (2) the degree of non-exponentiality of the correlation function of the structural α-relaxation, or the fractional exponent of the Kohlrausch stretched exponential function that fits it, at the glass transition temperature T_g [2]; (3) the steepness index m or fragility characterizing the rapidity of the increase in the structural α-relaxation time, $\tau_\alpha(T)$, on lowering temperature to approach T_g from the supercooled liquid side [3,4]; (4) the mean squared displacement,

$<r^2(T)>$, measured by quasielastic neutron scattering in the glassy state, as well as at T_g and above T_g in the liquid state [5,6]; (5) the effective Debye–Waller factor (i.e., the non-ergodicity parameter), f_0 [7–10], (6) the ratio, $\tau_\alpha(T_g)/\tau_\beta(T_g)$, where $\tau_\beta(T_g)$ is the relaxation time of a secondary relaxation belonging to a special class referred to as the Johari–Goldstein β-relaxation [11,12]; and (7) the Poisson's ratio ν [13–22], or K/G, the ratio of the elastic bulk modulus K to the shear modulus G.

While the first six items in the above are associated with one property to the other of the molecular dynamics (obtainable by spectroscopy in the time or frequency domain), the last item on the Poisson's ratio is an exception because it is from an elastic property of a material when compressed in one direction that it tends to expand in the two other directions The Poisson's ratio ν is defined as the ratio between the fraction of expansion divided by the fraction of compression. If the material is stretched instead of compressed, the Poisson's ratio is the ratio of the relative contraction to relative stretching, the same value as for compression. For isotropic material, the Poisson's ratio is related to other elastic constants K and G by [19–21]

$$\nu = (3K-2G)/2(3K+G). \tag{1}$$

Since K and G are positive numbers, the Poisson's ratio can vary between −1 (if $K = 0$) and 0.5 (if $G = 0$). The Poisson's ratio ν is an important parameter for elastic properties of materials and shown to critical for other macroscopic properties of materials of different kinds including glass-formers [19–21]. It is intriguing as a macroscopic parameter that it can bear relation or correlation at all to any of the microscopic molecular dynamic properties. Nonetheless, via its connection to the longitudinal sound waves governed by the longitudinal modulus, M, a relation has been found between ν and the effective Debye–Waller

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factor [14], which is the plateau of time-dependent density–density correlation function $F(Q,t)$ normalized by the static structure factor $S(Q)$ in the long wavelength limit, i.e. $Q \rightarrow 0$. The plateau appears at times longer than the vibration times but shorter than the secondary β-relaxation and the α-relaxation time. The height of the plateau, $f_0(T)$, is also known as the nonergodicity parameter in the context of Mode Coupling Theory (MCT) [23], but we shall continue referring to it as the effectively the Debye–Waller factor because no connection is made with MCT in this paper. The quantity, $1 - f_0(T)$, represents the decorrelation introduced by the vibrational dynamics, and it gives the part of the density fluctuations that is frozen deep in the glassy state. It also characterizes the combined amplitude of the β- and α-processes.

From the relation of the longitudinal sound waves to the long wavelength limit of the dynamical structure factor, it has been shown [23,24] that

$$f_0 = 1 - M_0/M_\infty \tag{2}$$

where M_0 and M_∞ are the zero and infinite frequency longitudinal modulus respectively. Using the identity, $M = K + \left(\frac{4}{3}\right)G$, the expression for f_0 can be rewritten as

$$f_0 = 1 - [K_0 + (4/3)G_0]/[K_\infty + (4/3)G_\infty]. \tag{3}$$

In Ref. [14], the approximation was made in the glassy state by putting the zero frequency shear modulus $G_0 = 0$, and neglecting the softening of the bulk modulus on decreasing frequency, i.e., putting $K_0 \cong K_\infty$. The final result on the relation between f_0 and K_∞/G_∞ is given by [14,25]

$$f_0 = \frac{1}{\frac{3}{4}\left(\frac{K_\infty}{G_\infty}\right) + 1}. \tag{4}$$

Thus, glass-former with larger K_∞/G_∞ has a smaller effective Debye–Waller factor f_0. In other words, K_∞/G_∞ correlates with $1/f_0$ or $(1 - f_0)$. Eq. (4) in conjunction with the exact relation,

$$\nu = \frac{1}{2} - \frac{3}{6K_\infty/G_\infty + 2}, \tag{5}$$

leads to the correlation of the Poisson's ratio ν with $1/f_0$ and $(1 - f_0)$, or the anti correlation with f_0. A plot of ν against $(1 - f_0)$ to show the correlation quantitatively is given in Fig. 1.

Despite of the fact that the correlation of ν with $1/f_0$, or $(1 - f_0)$ can be derived formally, the connection of the Poisson's ratio molecular dynamics of glass-forming liquids is limited. This is because the derivation of Eq. (4) linking the effective Debye–Waller factor, f_0, to the elastic
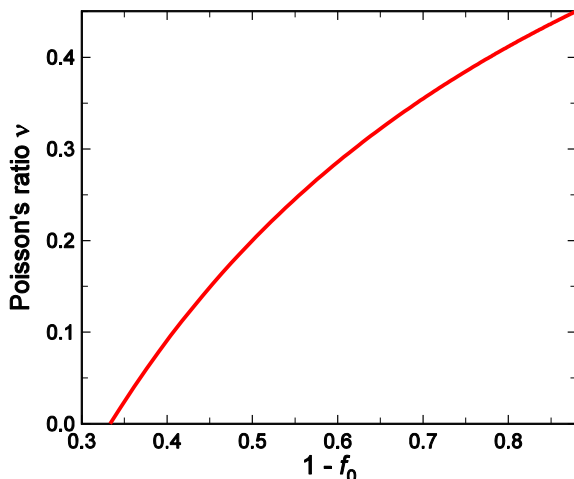
constants involves the longitudinal sound waves but none of the relaxation dynamics of the glass-forming liquid or glass. Therefore, insight of the connection of Poisson's ratio to molecular dynamics gained by Eqs. (4) and (5) is limited. Unclear is the question of whether and how the Poisson's ratio is related to the structural α-relaxation, which is central to the problem of glass transition. Advance was made in a recent paper [25] where a connection was made between $(1 - f_0)$ and the breadth of the time/frequency dispersion of the α-relaxation. The latter is proportional to the degree of non-exponentiality of the α-relaxation correlation function represented by n appearing in the fractional exponent, $1 - n$, of the Kohlrausch stretched exponential function,

$$\phi(t) = \exp\left[-(t/\tau_\alpha)^{1-n}\right]. \tag{6}$$

commonly used to fit the correlation function. Results from molecular dynamics simulations [26,27], quasielastic neutron and dynamic light scattering, and dielectric relaxation [25] together with the interpretations by the Coupling Model [28–31] were employed in establishing the correlation between $(1 - f_0)$ and n [25]. Also shown is that both quantities, f_0 and n, ultimately are determined by the anharmonicity of the inter-molecular potential, which governs the dynamics at all times ranging from vibrations including the Boson peak, the dissipation of caged molecules related to f_0 at short times, and Johari–Goldstein β-relaxation [11,12,32] at intermediate times, and the terminal α-relaxation at long times. On combining the two correlations, $(1 - f_0)$ with n, and ν with $(1 - f_0)$, we are able to reach the connection that the Poisson's ratio is having with the fundamental physics governing the structural α-relaxation and glass transition.

In applying these predictions to real glass-formers, the previous work [25] is totally focused on the family of metallic glasses. Although by and large the predictions have been verified in the special class of metallic glasses, naturally the task needed to follow up is to test if they hold also for glass-formers of other classes. This step is critical because most known glass-forming liquids are not metallic. The present paper reports the results of such an investigation.

The paper is organized as follows. We first briefly summarize the relevant molecular dynamics simulations results, the quasielastic neutron scattering data, and the theoretical basis and predictions of the Coupling Model, all of which have led to the correlation between $(1 - f_0)$ and n as well as the fundamental connection to anharmonicity of the potential. Application of the prediction to other glass-formers constituting the central part of the paper is presented next, and followed by discussion and conclusion. The correlation of the Poisson's ratio to molecular dynamic properties of glass-forming materials had been rationalized and explained theoretically by fundamental physics governing the structural α-relaxation in the previous publication [25], where the focus is on metallic glasses. Nevertheless, the same theoretical considerations apply verbatim to the molecular and polymeric glass-formers discussed in this paper. There is no need to duplicate it here because readers interested in the theoretical justification can consult Ref. [25].

2. Connecting f_0 to anharmonicity of potential and the coupling parameter n

In our previous publication [25], we made use of results from molecular dynamics simulations, various experiments, and the Coupling Model (CM) [28–31] to establish connection between f_0 and the relaxation processes in glass-forming liquids. A more fundamental connection was ultimately established between all quantities and parameters, including f_0, and the anharmonicity of the inter-molecular potential. On combining with the correlation between f_0 and ν shown in Fig. 1, the goal of linking the Poisson's ratio to molecular dynamics and glass transition is accomplished. In order to ensure that the paper is self contained and readable without having to go back to the previous work, we give

Fig. 1. Plot of Poisson's ratio against $(1 - f_0)$.

an abbreviated version of the essential connections or correlations before testing them for glass-formers that are not metallic glasses.

2.1. Correlation of $(1 - f_0)$ with n

Molecular dynamics simulations of binary Lennard–Jones (LJ) systems by Bordat et al. [26,27] in which the potential, $V(r)$, was varied systematically to increase its anharmonicity. To compare the dynamics, temperature in each case is scaled by T_{ref} at which τ_α is equal to an arbitrarily chosen long time. For all T/T_{ref}, increasing anharmonicity of the interparticle potential is accompanied by (i) increasing non-exponentiality of the α-relaxation or the coupling parameter $n(T/T_{ref})$ of the CM, (ii) decreasing effective Debye–Waller factor $f(Q_0, T/T_{ref})$ or equivalently increasing value of $(1 - f_0)$, and (iii) increasing steepness index or fragility parameter, m. Thus, correlations were established between four quantities, anharmonicity of potential, n, $1/f(Q_0,T)$, and m, in the supercooled liquid state. In the glassy state, the results of the simulations again show $f(Q \rightarrow 0, T/T_{ref}) \equiv f_0(T/T_{ref})$ anti-correlates with anharmonicity, and is able to reproduce the correlation found by Scopigno and coworkers [7,10]. Hence, we have in the glassy state as well the correlations between anharmonicity, n, $1/f_0(T/T_{ref})$ or $(1 - f_0)$, and m. Applying the simulation results to real glass-formers, T_{ref} can be taken as the glass transition temperature T_g. The increase of n with anharmonicity of the interparticle potential found by simulations is consistent with the CM based on nonlinear Hamiltonian dynamics (i.e., classical chaos) of systems governed by anharmonic interaction potential. Exact solutions of simplified model systems have demonstrated the increase of n with anharmonicity or nonlinearity of the potential [29–31].

Molecular dynamics simulations of the binary L–J particles with different potentials provide the best evidence of existence of correlation of $(1 - f_0)$ with n. Notwithstanding, it is necessary to see if such correlation holds for real glass-formers. For this purpose, we obtain $f_0(T_g)$ at $T = T_g$ from the its temperature dependence in the glass state,

$$f_0(T) = \frac{1}{1 + \alpha \left(\dfrac{T}{T_g} \right)}, \tag{7}$$

determined by means of inelastic X-ray scattering [7,11] and other means [8,9], and using the published values of α. Lesson has been learned before [2] in demonstrating correlations of properties that it is better not to mix glass-formers of different classes. This is because glass-formers of widely different chemical and physical structures have dissimilar intermolecular potentials, which can contribute variations beyond the correlation sought after. Therefore we consider the data of $f_0(T_g)$ from amorphous polymers separately. The rest are inorganic and organic glass-formers. Despite not from the same class, we consider all of them together in Fig. 2 because of limited number of data of $f_0(T_g)$ available if we further sort them out into different classes. Plotting $f_0(T_g)$ against $n(T_g)$ in Figs. 2 and 3 for the non-polymeric glass-formers and polymers respectively, existence of anti-correlation between $f_0(T_g)$ and $n(T_g)$, or correlation between $(1 - f_0(T_g))$ and $n(T_g)$, has support from the experimental data.

From Eqs. (4) and (5) we can see that the Poisson's ratio ν anti-correlates with $f_0(T_g)$, or ν correlates between $(1 - f_0(T_g))$. Thus, on combining the latter as shown quantitatively in Fig. 1 with the experimentally supported correlation between $(1 - f_0(T_g))$ and $n(T_g)$ in Figs. 2 and 3, we arrive at the important result that the Poisson's ratio ν is connected to n and through n the dynamics of the glass-former. Moreover, since n is determined by the potential, so is the Poisson's ratio related to the potential.

It would be nice to show the correlation between ν and $(1 - f_0(T_g))$ by experimental data. However, very few glass-formers have both ν and $(1 - f_0(T_g))$ been measured. Despite it is most interesting to verify
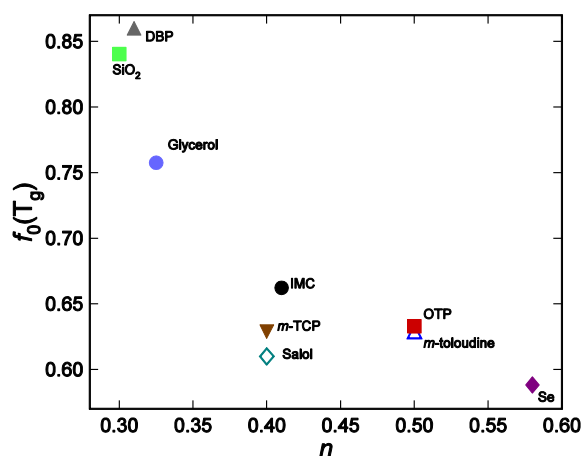

Fig. 2. Plot of $f_0(T_g)$ against the non-exponentiality or coupling parameter n. The data of $f_0(T_g)$ are deduced from Refs. [7–10] as described in the text. The n values are obtained from Refs. [5,6,31].

directly the relation of ν to $(1 - f_0(T_g))$ by real data, we have to wait for measurements in the future. There is only the Ge_xSe_{1-x} systems of several compositions ($x = 0.0, 0.10, 0.15$, and 0.25) in which both ν at room temperature in the glassy state [19] and $n(T_g)$ are available from experiments [33]. The average coordination number $<r>$ of Ge_xSe_{1-x} is computed from the formula $<r> = 4x + 2(1 - x)$. The plot of ν against $n(T_g)$ in Fig. 4 shows presence of the strong correlation on restricting the glass-formers within the same family.

2.2. Correlation of the mean-square-displacement, $<u^2>$, with n and ν at T_g

From quasielastic neutron scattering experiment at wavevector Q, the Debye–Waller factor of the glass-former is given in terms of the mean square displacement (MSD), $<u^2>$, by [3,34,35]

$$f_0 = \exp \left[-<u^2>Q^2/3 \right]. \tag{8}$$

Hence, by this relation, another way to obtain f_0 is from $<u^2>$. More than ten years ago, the data of $<u^2>$ from different glass-formers studied by quasielastic neutron scattering were collected and compared, resulting in the establishment of a correlation between n and $<u^2>$ [35]. Since different glass-formers have different T_g, comparison of $<u^2>$ is made after temperature has been scaled by T_g. Glass-former with larger n has a larger $<u^2(T/T_g)>$ at the same value of $T/T_g \leq 1$
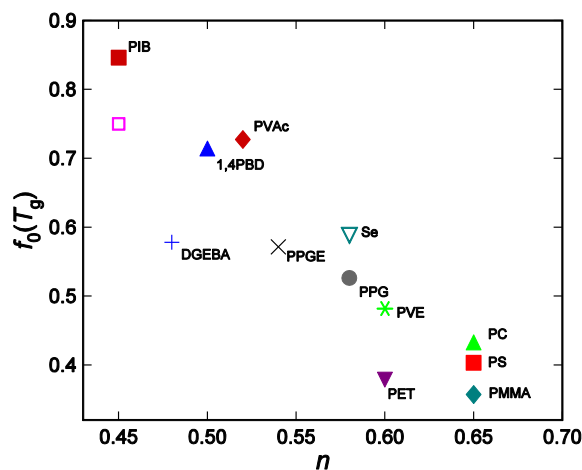

Fig. 3. Plot of $f_0(T_g)$ against the non-exponentiality or coupling parameter n. The data of $f_0(T_g)$ are deduced from Refs. [7,10] as described in the text. The n values are obtained from Refs. [5,6,31].
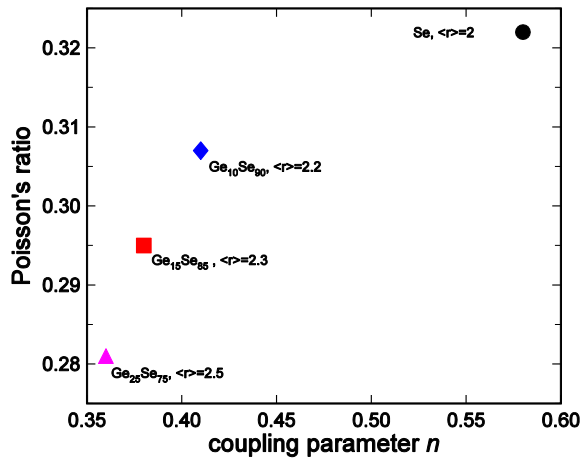
Fig. 4. Plot of Poisson's ratio against the non-exponentiality or coupling parameter n for the Ge_xSe_{1-x} glasses.

including $T/T_g = 1$. It increases more rapidly as a function of T/T_g. The T_g-scaled temperature dependence of $<u^2(T/T_g)>$ changes on crossing T_g to become stronger. The change is also more marked for glass-former with larger n. We show as examples the comparison between ortho-terphenyl ($n(T_g) = 0.50$), and glycerol ($n(T_g) = 0.30$–0.35) of their $<u^2(T/T_g)>$ in Fig. 5, and the comparison of PS ($n(T_g) = 0.65$) versus 1,4 PBD ($n(T_g) = 0.50$) in Fig. 6. The insets in each figure show the glass-former with larger $<u^2(T/T_g)>$ and $n(T_g)$, has larger Poisson's ratio ν. It would be nice to show more examples like in Figs. 5 and 6. Unfortunately, there are very few glass-formers that have all three quantities, $<u^2(T/T_g)>$, $n(T_g)$, and ν, determined by experiments. Furthermore, the MSD $<u^2(T/T_g)>$ depends on the energy resolution of the neutron scattering spectrometer. In Fig. 5, the data were obtained with spectrometer with higher energy resolution to include fast processes with frequencies higher than 0.24 GHz, while lower energy resolution in Fig. 6 includes processes higher than 48 GHz [35]. Hence meaningful comparison of $<u^2(T/T_g)>$ between different glass-formers can be made if the data are taken with the same energy resolution. This further restricts the number of cases we can consider.

There are a few glass-formers for which the Poisson's ratio ν has been measured as a function of temperature below and above T_g [19,20], and at the same time the microscopic dynamic parameters, $<u^2(T/T_g)>$ and $n(T_g)$, are also available. This gives us an opportunity to correlate the T_g-scaled temperature dependence of ν with $<u^2(T/T_g)>$ and $n(T_g)$ in Fig. 7 even though the number of glass-formers is limited. In this figure, $\nu(T/T_g)$ of PS ($T/T_g = 373$ K), Se
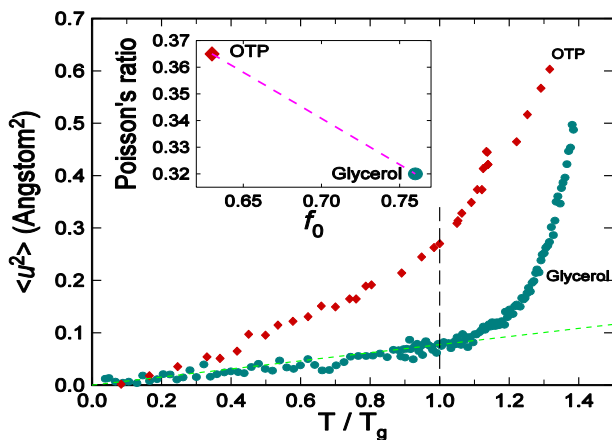
$(T/T_g = 313$ K), glycerol ($T/T_g = 186$ K), and B_2O_3 ($T/T_g = 541$ K) taken from Ref. [19] can be compared with their $<u^2(T/T_g)>$ shown in the inset. For Se, there is only a single data point of $\nu(T/T_g)$ taken at $T = 293$ K and is shown at $T/T_g = 1.07$. For amorphous water ($T_g = 136$ K), there is no data of $<u^2(T/T_g)>$ available to present in the inset. By inspection, the magnitudes as well as the T_g-scaled temperature dependence of $\nu(T/T_g)$ and $<u^2(T/T_g)>$ of PS, Se, glycerol and B_2O_3 correlate quite well. In addition, the magnitude of both quantities as well as the steepness of the rise on crossing T_g to the 'liquid' state correlate also with $n(T_g)$, 0.65 for PS, 0.58 for Se, 0.35–0.30 for glycerol, 0.40 for B_2O_3, and close to zero value for amorphous water [36]. Since the steepness or fragility index m has the values of 139, 87, 53, and 32 respectively for PS, Se, glycerol, and B_2O_3, it also correlates with $<u^2(T/T_g)>$, $n(T_g)$, and especially $\nu(T/T_g)$, the focus of this paper.

3. Discussion and conclusion

The Poisson's ratio ν has a long history in materials science and engineering in providing an important measure of the elastic performance of any material under mechanical strain. In recent years, it has been linked to other properties beyond elasticity such as ductility and plasticity of materials, and others summarized in reviews [19,20,37]. These broad range developments indicate that somehow ν has connection to the microscopic molecular dynamics and relaxation processes. One way to demonstrate the existence of the connection is to derive from

Fig. 6. Plot of mean-square-displacement, $<u^2(T/T_g)>$ versus T/T_g for polystyrene (PS) and 1,4 polybutadiene (1,4 PBD). The inset shows the observance of the anticorrelation between the Poisson's ratio and $f_0(T_g)$.

Fig. 5. Plot of mean-square-displacement, $<u^2(T/T_g)>$ versus T/T_g for OTP and glycerol. The inset shows the observance of the anticorrelation between the Poisson's ratio and $f_0(T_g)$.

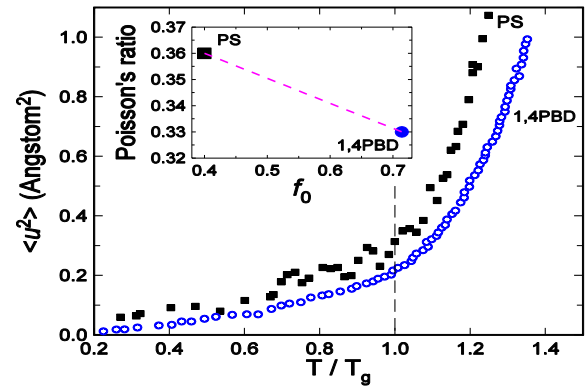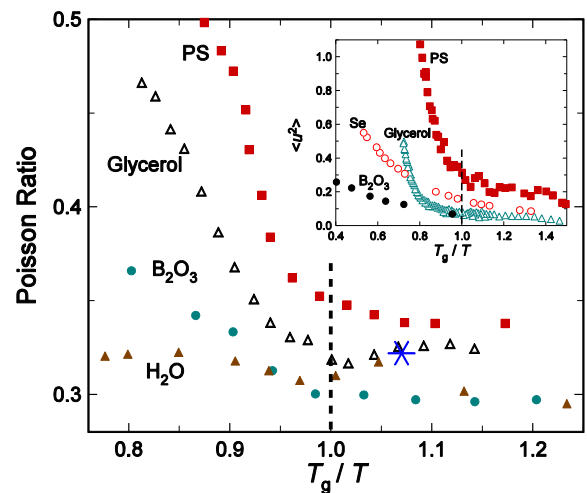**Fig. 7.** Plotting against T/T_g the Poisson's ratio, $\nu(T/T_g)$, in the main figure, and $<u^2(T/T_g)>$ in the inset. For $\nu(T/T_g)$ data are available only for PS, glycerol, B_2O_3, and H_2O, the single blue star is the Poisson's ratio of Se at 297 K. All are taken from Ref. [19]. The inset shows the data of PS, Se, glycerol, and B_2O_3 taken from Ref. [3].

theoretical consideration a relation between ν and some dynamic quantity. In fact there is a relation between ν and the effective Debye–Waller factor, f_0, given by Eqs. (4) and (5) combined, and plotted out in Fig. 1. Notwithstanding, f_0 is related to vibrational properties, and the relation of the Poisson's ratio to molecular dynamics and relaxation is still not in hand. To reach this goal we have utilized the results from molecular dynamics simulations and the precept of the Coupling Model to establish the relation between f_0 and the non-exponentiality or coupling parameter, n, of the structural α-relaxation. Ultimately it is the anharmonicity of the intermolecular potential that determines both f_0 and n, and hence their relation. By stringing together the two relations, ν with f_0 and f_0 with the structural α-relaxation, we have obtained a connection between ν and microscopic dynamics. In support of this result, experimental data of f_0, n, and ν from many glass-forming liquids are collected together to show that there is a correlation. Mean square displacement $<u^2>$ obtained from quasielastic neutron scattering experiments is related to f_0, and there is also a correlation of $<u^2>$ with ν and n, as well as with fragility index m. In order to establish the correlations, we need ν, f_0, $<u^2>$, n, and m, all determined for each of the glass-formers. There are not many glass-formers satisfying this requirement, and more are needed in the future to confirm these correlations. With the caveat that it is based on the data available so far, the conclusion we make is that the goal of relating the Poisson's ratio to microscopic dynamics of glass-formers has been achieved.

Acknowledgment

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