PHYSICAL REVIEW E **69**, 031501 (2004)

# Relation between the activation energy of the Johari-Goldstein $\beta$ relaxation and $T_g$ of glass formers

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(Received 8 August 2003; published 30 March 2004)

For glass-forming substances, we show that the ratio $E_\beta/RT_g$ can be predicted quantitatively from the coupling model. Here $E_\beta$ is the glassy state activation enthalpy of the Johari-Goldstein $\beta$ relaxation, $T_g$ is the glass transition temperature of the $\alpha$ relaxation, and $R$ is the gas constant. The calculated value is in good agreement with the experimental value in many glass formers. The results locate the origin of this cross correlation between $E_\beta$ of the Johari-Goldstein $\beta$ relaxation and $T_g$ of the $\alpha$ relaxation, although there are some notable exceptions to this cross correlation.

DOI: 10.1103/PhysRevE.69.031501 PACS number(s): 64.70.Pf, 77.22.Gm

## I. INTRODUCTION

Among secondary relaxations in glass formers, the intriguing ones are those that involve the motion of essentially all parts of the molecule and not just an isolated part of the molecule. Evidences of the existence of such secondary relaxation are provided in the works of Johari and Goldstein (JG) [1,2]. They found secondary relaxation even in rigid molecular glass formers, any motion of which must involve the entire molecule. Thus it is appropriate to refer to a secondary relaxation as JG $\beta$ relaxation if it is originating from motion of essentially all parts of the molecule. Naturally, the JG relaxation so defined is not intramolecular but intermolecular in origin, a phrase commonly used in the literature to define JG $\beta$ relaxation. Since the primary $\alpha$ relaxation also involves motion of the entire molecule, albeit cooperatively with other molecules, there is reason to expect that the dynamic properties of JG $\beta$ relaxation defined here may bear some correlation with that of the $\alpha$ relaxation. In fact, based on such correlations, more precise criteria for identification of JG $\beta$ relaxation are given in a recent work [3]. Since the independent relaxation of the coupling model (CM) [4–6] also involve the local motion of the entire molecule, one such criterion is the correspondence between the most probable JG $\beta$-relaxation time $\tau_{\mathrm{JG}}$ and the independent relaxation time $\tau_0$ [7–10], i.e.,

$$\tau_{\mathrm{JG}} \approx \tau_0 . \tag{1}$$

The correspondence has been shown to hold for genuine JG $\beta$ relaxation in a number of glass formers at temperatures above the glass transition temperature $T_g$ [7–10]. This is an indication of the possible fundamental role played by the JG $\beta$ relaxation in glass transition because, via $\tau_0$, it relates $\tau_{\mathrm{JG}}$ to parameters that characterize the $\alpha$ relaxation, namely, the relaxation time $\tau_\alpha$ and exponent $n$ in its correlation function given by the Kohlrausch-Williams-Watts function

$$\phi(t) = \exp[-(t/\tau_\alpha)]^{1-n}. \tag{2}$$

Explicitly, the relation from the CM that relates $\tau_0$ to $\tau_\alpha$ is

$$\tau_0 = (\tau_\alpha)^{1-n} t_c^n , \tag{3}$$

where $t_c$ is the crossover time in the coupling model which is about 2 ps for small molecular and polymeric glass formers [4–6].

Another possible connection of the JG $\beta$ relaxation to glass transition is suggested by the empirical relation between $T_g$ and the activation enthalpy $E_\beta$ of $\tau_{\mathrm{JG}}$

$$E_\beta = 24RT_g , \tag{4}$$

found by Kudlik *et al.* [11–13]. Although the relation is only approximate and there are deviations, it is a remarkable finding. The purpose of this work is to derive a relation between $E_\beta$ and $T_g$ from Eqs. (1) and (3) of the coupling model, and compared it with experimental data as well as with the empirical relation (4).

## II. RELATION BETWEEN $E_\beta$ AND $T_g$

At temperatures below $T_g$, the most probable relaxation times of all secondary relaxations have Arrhenius temperature dependence. In particular, for the JG relaxations, we have

$$\tau_{\mathrm{JG}}(T) = \tau_\infty \exp(E_\beta/RT), \quad T \leqslant T_g , \tag{5}$$

where $\tau_\infty$ is the prefactor and $R$ the gas constant. This expression for $\tau_{\mathrm{JG}}$ together with Eqs. (1) and (3) lead us to the relation

$$E_\beta/RT = 2.303[(1-n)\log_{10}\tau_\alpha(T) + n\log_{10} t_c - \ln\tau_\infty], \quad T \leqslant T_g , \tag{6}$$

At $T = T_g$, Eq. (6) is reduced to a relation between $E_\beta$ and $T_g$ given by

$$E_\beta/RT_g = 2.303[(1-n)\log_{10}\tau_\alpha(T_g) + n\log_{10} t_c - \log_{10}\tau_\infty]. \tag{7}$$

Most measurements of secondary relaxations are obtained by means of dielectric relaxation spectroscopy, where $T_g$ is conveniently defined as the temperature at which the dielectric relaxation time $\tau_\alpha$ reaches an arbitrarily long time, typically $10^2$ s. Following this convention, on substituting $\tau_\alpha(T_g)=10^2$ s and $t_c=2$ ps into Eq. (7), we arrive at the expression

$$E_\beta/RT_g=2.303(2-13.7n-\log_{10}\tau_\infty). \qquad (8)$$

The ratio $E_\beta/RT_g$ for JG $\beta$ relaxation depends on the exponent $n$ of the $\alpha$ relaxation and the prefactor $\tau_\infty$ of the JG $\beta$ relaxation, but it is not immediately clear why the ratio stays close to 24 for JG $\beta$ relaxation in many glass formers examined by Kudlik *et al.* We hasten to mention here that not all secondary relaxations examined by Kudlik *et al.* are genuine JG $\beta$ relaxation [3]. Most glass formers that exhibit well-resolved JG $\beta$ relaxation have $n\geqslant 0.40$. This condition was found empirically [7–9]. It is also required theoretically by Eqs. (1) and (3) for sufficient separation of the JG $\beta$ relaxation from the $\alpha$ relaxation. Otherwise, if $n$ is too small, the separation is not sufficient and the JG $\beta$ relaxation cannot be resolved. Instead an excess wing is observed [10]. With very few exceptions, most of these glass formers have $n<0.70$. The majority has $n$ lying within the approximate range of $0.65\geqslant n\geqslant 0.40$. The prefactor $\tau_\infty$ varies but most are within the range of $10^{-13}>\tau_\infty>10^{-18}$ s. There is also a correlation between $n$ and $\tau_\infty$. Smaller $n$ is associated with longer $\tau_\infty$. These bounds of $n$ and $\tau_\infty$, as well as the correlation between $n$ and $\tau_\infty$, hold for genuine JG $\beta$ relaxations in most of the glass-formers considered by Kudlik *et al.* and by us in this work (Table I). For several representative values of $\tau_\infty$, the ratio is calculated as a function of $n$ according to Eq. (8). Thus, for the majority of glass formers, it is sufficient to display the calculated $E_\beta/RT_g$ within the established bounds of $n$ (the abscissa) and $\tau_\infty$ (the parameter) as shown by the straight lines in Fig. 1. Indeed the calculated values of the ratio $(E_\beta/RT_g)_{\rm cal}$ are in the neighborhood of the value 24 (horizontal line in Fig. 1) found for many glass formers by Kudlik *et al.*

The experimental data of glass formers considered in this work are introduced and further discussed in the following paragraphs. But before that, we enter into Fig. 1 the experimental values of the ratio $(E_\beta/RT_g)_{\rm expt}$ for the majority of glass formers that obey the imposed bounds of $n$ and $\tau_\infty$. Many glass formers indeed have $(E_\beta/RT_g)_{\rm expt}$ (symbols in Fig. 1) in the neighborhood of empirical value 24. Overall $(E_\beta/RT_g)_{\rm expt}$ has a spread but is matched by the calculated $(E_\beta/RT_g)_{\rm cal}$ (straight lines).

To see if the ratio $(E_\beta/RT_g)_{\rm expt}$ can be adequately accounted for by Eq. (8) of the coupling model (CM), we have to examine individually the experimental data of a number of glass formers that show genuine JG $\beta$ relaxation. For each glass former, we calculate the ratio according to the right-hand side of Eq. (8) using $n$ and $\tau_\infty$ from experimental data, and compare the ratio obtained directly from $E_\beta$ and $T_g$. The results together with the parameters used are shown in Table I. Several small molecular glass formers have two secondary relaxations. They are triphenylolmethane triglicidyl ether (TPMTGE) [14], diglycidyl ether of bisphenyl-A (DGEBA) [15], poly[(phenyl glycidyl ether)-co-formaldehyde] (PPGE) [15], dipropyleneglycol dibenzoate (DiPGDiB) [16], and benzoin isobutylether (BIBE) [17] (see Table I). The different dynamic properties of the two secondary relaxations have been used to tell which one is a JG $\beta$ relaxation [3]. Without exception, the slower one is the JG $\beta$ relaxation. This is intuitively obvious because JG $\beta$ relaxation involves motion of essentially all parts of the molecule must be slower than the other secondary relaxation. For DGEBA, PPGE, and DiPGDiB, the experimental values of $E_\beta/RT_g$ of the JG $\beta$ relaxation is not far from the value of 24 proposed by Kudlik *et al.* TMPTGE has notably a smaller value of 17.8. In contrast, BIBE has a larger value of 30.1. Nevertheless, in spite of such variations, for each of these glass-formers there is good agreement between the experimental value of ratio $(E_\beta/RT_g)_{\rm expt}$ and the calculated value $(E_\beta/RT_g)_{\rm cal}$ for the JG $\beta$ relaxation (see Table I).

The calculation by Eq. (8) does not apply to non-JG relaxations, and therefore no calculated value of $E_\beta/RT_g$ is supplied for them in Table I. The experimental values $(E_\beta/RT_g)_{\rm expt}$ of the faster non-JG secondary relaxations in TMPTGE, DGEBA, and PPGE are all about 13, which is significantly smaller than the proposed values of 24. There are more trivial non-JG secondary relaxations in other glass formers that have even smaller values of $(E_\beta/RT_g)_{\rm expt}$. An extreme example is the rotational motion of a pendant methyl group in poly(vinyl methyl ether), which has a small $E_\beta$ of about 8.4 kJ/mol and $T_g=250$ K [18], and thus $(E_\beta/RT_g)_{\rm expt}=4.0$. This falls way outside the empirical correlation of $E_\beta$ with $RT_g$. Hence non-JG secondary relaxations are to be excluded in order to preserve any correlation.

The polymeric glass-formers, poly(ethylene terephthalate) (PET) and poly(ethylene 2,6-naththalene dicarboxylate) (PEN), also have more than one secondary relaxations [19–22]. Again the slowest one (commonly called $\beta^*$) is likely the JG $\beta$ relaxation, particularly since it involves motion of the bond linking the aromatic ring carbon to the ester carbon. The value of $(E_\beta/RT_g)_{\rm expt}$ for PET is 24.3, nearly the same as 24. But for PEN, it takes a much larger value of 41.4. We draw attention to the very large value of $E_\beta$ compensated by a corresponding large value of $-\log_{10}\tau_\infty$ of the $\beta^*$ relaxation [21,22], which we identified as the JG $\beta$ relaxation here. These large values have led others to believe there exists some degree of cooperative of the naphthalene groups [20,22]. Thus the $\beta^*$ relaxation in PEN is a special case and the assumption that it is a JG $\beta$ relaxation may not be valid. Nevertheless, there is good agreement between the experimental and calculated values of the ratio for the slowest secondary relaxation for both PET and PEN. The faster non-JG secondary relaxations in PET show up more prominently in the dielectric spectrum. They are the motions of the ester ether oxygen to the aliphatic carbon bond and the aliphatic carbon-carbon bond, and have larger dielectric strength than the slowest JG $\beta$ relaxation. The values of $(E_\beta/RT_g)_{\rm expt}$ for the fastest non-JG secondary relaxation in PET and PEN are appreciably smaller than 24 (see Table I), just as in the cases of TMPTGE, DGEBA, and PPGE. Again, inclusion of these

TABLE I. Comparison of the ratio $(E_\beta/RT_g)_{\mathrm{expt}}$ obtained directly with $E_\beta$ and $T_g$ taken from experimental data and $(E_\beta/RT_g)_{\mathrm{cal}}$ calculated according to Eq. (8) with $n$ and $\tau_\infty$ from experimental data. All secondary relaxations are genuine JG $\beta$ relaxation unless otherwise stipulated. No value of $(E_\beta/RT_g)_{\mathrm{cal}}$ is given for non-JG secondary relaxations because Eq. (8) only applies to JG $\beta$ relaxations.

| Glass former | $T_g$ (K) | $n$ | $-\log_{10}\tau_\infty$ | $E_\beta$ (kJ/mol) | $(E_\beta/RT_g)_{\mathrm{expt}}$ $p$ | $(E_\beta/RT_g)_{\mathrm{cal}}$ | Refs. |
|---|---|---|---|---|---|---|---|
| TMPTGE | 287 | 0.54 | 12.54 | 41.9 | 17.8 | 16.4 | [14], [16] |
| TMPTGE (non-JG) | 287 | | 14.3 | 30.6 | 13 | | [14], [16] |
| DGEBA | 253.7 | 0.47 | 14.78 | 47.6 | 22.6 | 23.8 | [15], [16] |
| DGEBA (non-JG) | 253.7 | | 14.35 | 27.6 | 13.1 | | [15], [16] |
| PPGE | 258.4 | 0.54 | 14.6 | 47.3 | 22.0 | 21.2 | [15], [16] |
| PPGE (non-JG) | 258.4 | | 14.7 | 27.9 | 13.0 | | [15], [16] |
| DiPGDiB | 220 | 0.38 | 14.7 | 49.6 | 26.8 | 26.2 | [16] |
| DiPGDiB (non-JG) | 220 | | 13.7 | 32.8 | 18 | | [16] |
| BIBE (JG) | 220 | 0.35 | 16.3 | 55 | 30.1 | 31.1 | [17] |
| BIBE (non-JG) | 220 | | 14.3 | 28 | 15.3 | | [17] |
| PET *amorphous* | 353 | 0.52 | 17.4 | 63.8 | 24.3 | 28.3 | [20], [35] |
| PET *amorphous* (non-JG $\beta_1$) | 353 | | | 31.4 | 13.3 | | [20] |
| PEN amorphous | 389 | 0.52 | 22.3 | 133.9 | 41.4 | 39.9 | [19], [20] |
| | 389 | 0.52 | 24 | 147 | 45.5 | 43.5 | [21] |
| PEN (non-JG $\beta_1$) | 389 | | | 31.4 | 9.7 | | [19], [20] |
| | 389 | | 12.2 | 36 | 11.1 | | [21] |
| Sorbitol | 268 | 0.52 | 15.2 | 51.96 | 23.3 | 23 | [8], [23–25] |
| Xylitol | 246.7 | 0.46 | 13.7 | 44.73 | 21.8 | 21.6 | [8], [9], [26] |
| 5-methyl-2-hexanol | 152.7 | 0.46 | 14.2 | 26.0 | 20.5 | 22.8 | [8], [27] |
| 2-ethyl-1-hexanol | 148 | 0.46 | 11.82 | 19.2 | 15.6 | 17.3 | [28] |
| 1-Propanol | 95 | 0.40 | 15.5 | 23.1 | 29.3 | 27.7 | [7], [36] |
| Toluene | 116 | 0.46 | 17.0 | 25.3 | 28.5 | 29.2 | [8], [11] |
| OTP | 245 | 0.50 | 16.8 | 53.0 | 26.0 | 27.5 | [11], [24] |
| 1,4 Polybutadiene | 176 | 0.56 | 15.3 | 35.7 | 24.4 | 22.2 | [8], [11–13] |
| Polyvinylchloride | 350 | 0.73 | 19.5 | 57.5 | 19.8 | 26.4 | [7], [29] |
| Heterocyclic polymer Network (linear to Network ratio $L/N$) | | | | | | | |
| HPN($L/N$=100/0) | 291.6 | 0.57 | 15.53 | 54.0 | 22.3 | 22.4 | [30] |
| HPN($L/N$=75/25) | 311.9 | 0.61 | 15.78 | 55.3 | 21.3 | 21.7 | [30] |
| HPN($L/N$=60/40) | 322.3 | 0.63 | 15.75 | 55.2 | 20.6 | 21.0 | [30] |
| HPN($L/N$=43/57) | 333.0 | 0.63 | 16.33 | 57.4 | 20.7 | 22.3 | [30] |
| HPN($L/N$=0/100) | 351.3 | 0.71 | 16.47 | 58.3 | 20.0 | 20.1 | [30] |
| Maltitol (dielectric) | 313 | 0.60 | 16.0 | 56.51 | 21.7 | 22.52 | [32] |
| Maltitol (mechanical) | 310 | 0.60 | 15.52 | 61.7 | 23.9 | 21.5 | [33] |
| Glucose | 309 | 0.66 | 16.79 | 51.83 | 20.18 | 22.5 | [34] |
| Fructose | 277.2 | 0.66 | 15.62 | 48.03 | 20.8 | 19.8 | [34] |

non-JG secondary relaxations would further undermine the already not-so-perfect correlation between $E_\beta$ and $RT_g$ of JG $\beta$ relaxation.

Toluene, ortho-terphenyl (OTP), 1-propanol and 1,4 polybutadiene are among the glass formers considered by Kudlik *et al.* in obtaining the near constancy of $(E_\beta/RT_g)_{\mathrm{expt}}$ given by Eq. (3). They are included in Table I together with the newcomers, sorbitol [23–25], xylitol [26], 5-methyl-2-hexanol [27], 2-ethyl-1-hexanol [28], polyvinylchloride [29], and several heterocyclic polymer networks systems with different linear to network ratios [30]. The secondary relaxation in all these glass-formers are genuine JG $\beta$ relaxations. The values of $(E_\beta/RT_g)_{\mathrm{expt}}$ straddle about the suggested value of 24. In all cases, except polyvinylchloride, as with the other glass formers discussed above, the calculated value $(E_\beta/RT_g)_{\mathrm{cal}}$, matches the experimental value $(E_\beta/RT_g)_{\mathrm{expt}}$. Some glass formers in Kudlik *et al.* are not considered in this work. Isothermal dielectric relaxation spectra of the $\alpha$-relaxation in these glass-formers are either incomplete or unavailable, making the determination of $n$ and hence the calculation of $(E_\beta/RT_g)_{\mathrm{cal}}$ via Eq. (8) impossible. Since one of the main goals of this work is to compare $(E_\beta/RT_g)_{\mathrm{cal}}$ with $(E_\beta/RT_g)_{\mathrm{expt}}$, the absence of $(E_\beta/RT_g)_{\mathrm{cal}}$ for these glass formers is the reason for not including them

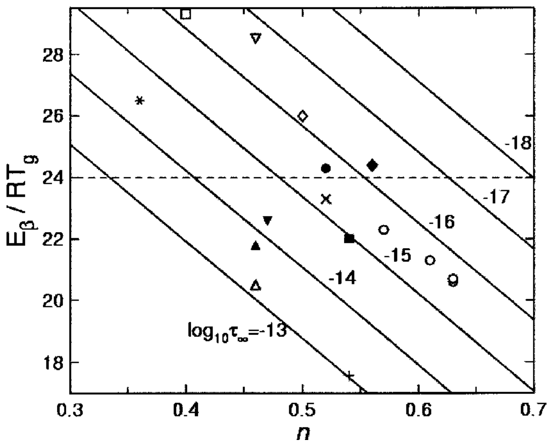

FIG. 1. The straight lines are the ratios $E_\beta/RT_g$ calculated as a function of $n$ according to Eq. (8) for several representative values of $\tau_\infty$ as indicated. The figure shows the results within the bounds of $n$ and $\tau_\infty$ established (see text). The experimental values of the ratio $E_\beta/RT_g$ for the glass formers considered in this work having $n$ and $\tau_\infty$ within the bounds are shown by symbols. TMPTGE (+), DGEBA (▼), PPGE (■), DiPGDiB (*), PET (○), Sorbitol (x), Xylitol (▲), 5-methyl-2-hexanol (△), 1-propanol (□), toluene (▽), OTP (◇), 1,4 polybutadiene (◆), HPN [$L/N$=100/0, 75/25,60/40,43/57] (○).

in this work. We also exclude glass formers that are binary mixtures because concentration fluctuations introduce a distribution of environments and coupling parameters, causing extraneous broadening to the $\alpha$ relaxation and even the most probable $n$ cannot be determined without large uncertainty [31]. Lastly, we include maltitol [32,33], fructose [34], and glucose [34] in Table I, but we caution that there are larger uncertainties in the values of $n$ given there for these glass formers, because of the lack of complete data.

In this work we have confined our consideration of the JG $\beta$-relaxation below $T_g$, where the relaxation time $\tau_{\mathrm{JG}}$ has Arrhenius temperature dependence. However, the temperature of $\tau_{\mathrm{JG}}$ above $T_g$ is less certain because of the overlap of the JG $\beta$ relaxation with the $\alpha$ relaxation. Some procedure with assumption has to be used to resolve the JG $\beta$ relaxation in the isothermal spectra at ambient temperature and determine $\tau_{\mathrm{JG}}$. Some workers found that the Arrhenius temperature dependence does not continue to hold at temperatures above $T_g$ [23–25,37], while the results of others [13,36] seem to indicate otherwise. The situation becomes even more confusing when non-JG secondary relaxations and genuine JG $\beta$ relaxation are both included in the discussion of temperature dependence above $T_g$. Non-JG secondary relaxations are more local, occur at higher frequencies, and bear little or no relation to the $\alpha$ relaxation. Hence its Arrhenius temperature dependence below $T_g$ can continue to above $T_g$. Examples of such behavior are found in the $\gamma$ relaxation of TPMTGE [14], DGEBA [15], and PPGE [15]. Other examples are the well-resolved secondary relaxation in BMPC [36,38,39], diethyl phthalate (DEP) [40], and fluroaniline (FAN) [13]. None of these well-resolved secondary relaxations in the latter group of glass formers with narrow $\alpha$-loss peaks (smaller $n$) are genuine JG $\beta$ relaxation [3]. An example of the evidence is the total lack of pressure dependence of the relaxation time [39], in stark contrast to the $\alpha$-relaxation time. This difference shows that they are unrelated to the $\alpha$-relaxation and hence not genuine JG $\beta$ relaxations according to our definition. The genuine JG $\beta$ relaxations are not resolved but appear as part of the excess wing. For FAN, there is evidence of hydrogen-bond induced clusters [41] from neutron scattering and computer simulations. The clearly observed secondary relaxation in FAN [13] possibly arises from some motion associated with hydrogen-bond induced clusters [41]. If so, then it is definitely not a genuine JG $\beta$ relaxation. The apparent persistence of the Arrhenius temperature dependence of the non-JG secondary relaxation in the aforementioned glass-formers give an impression may cause many to jump to the conclusion that all secondary relaxations, including the genuine JG $\beta$ relaxations, behave in the same way. Thus it is of paramount importance to separate out genuine JG $\beta$ relaxations from non-JG secondary relaxation in discussing properties. Recently indisputable evidence has been acquired [42] to show that genuine JG $\beta$ relaxations do not have the Arrhenius temperature dependence of its relaxation time below $T_g$ continued to temperatures above $T_g$. At elevated pressure, the separation between the $\alpha$- and $\beta$-relaxation peaks is larger than at ambient pressure, enabling the JG $\beta$-relaxation times to be directly and unambiguously determined without using any arbitrary procedure. Taking advantage of this, it was proven that the Arrhenius temperature dependence of the JG $\beta$-relaxation time $\nu_\beta$ for temperatures below $T_g$ does not persist for temperatures above $T_g$ [42].

## III. CONCLUSION

Any relation between the parameters that characterize the Johari-Goldstein (JG) $\beta$ relaxation and the $\alpha$ relaxation is interesting because it links the two relaxations together and indicate the possibility that the former is a ''local step'' or the precursor of the latter. Thus the correlation of $E_\beta$ with $RT_g$ found by Kudlik *et al.* has drawn attention to workers in glass transition, including us. By examining additional glass formers, we confirm the findings of Kudlik *et al.* that the ratio $(E_\beta/RT_g)_{\mathrm{expt}}$ for the JG $\beta$ relaxation in many glass formers straddles the value of 24. However, there are notable large deviations in a few glass formers. Finally, the values of $(E_\beta/RT_g)_{\mathrm{expt}}$ for non-JG secondary relaxations examined in this work are significantly smaller than 24.

For JG $\beta$ relaxations, the ratio $E_\beta/RT_g$ can be computed by the extended coupling model, which relates the JG $\beta$-relaxation time to the $\alpha$-relaxation time. There is good correspondence between the calculated and the experimental values for all glass formers considered with one minor exception. The computation of the exact value of $E_\beta/RT_g$ requires the knowledge of the prefactor $\tau_\infty$ of the JG $\beta$ relaxation. Even without knowing $\tau_\infty$ but locating it within a broad range of $10^{-13} \leqslant \tau_\infty \leqslant 10^{-18}$ s, it is sufficient to show that the computed values of $E_\beta/RT_g$ fall within a broad neighborhood about 24 for most glass formers (Fig. 1), an empirical result first found by Kudlik *et al.* Thus our work provides a rationale for the empirical relation between $E_\beta$ and $T_g$ of the $\alpha$ relaxation. Such a relation is just one example of several relations found to exist [3,7–10,42] between dynamics of the JG $\beta$ relaxation, on the one hand, and dynamics of the $\alpha$ relaxation, on the other. These cross relations all have the physical meaning that the JG $\beta$ relaxation is not only a ''universal'' feature in glass formers but also has fundamental implications for the mechanism of glass transition. The good correspondence of the JG $\beta$ relaxation time with the primitive relaxation time of the coupling model implies that both relaxations can be considered as a ''local step'' or the precursor of the cooperative $\alpha$ relaxation.

## ACKNOWLEDGMENTS

The research at NRL was supported by the Office of Naval Research, and at Pisa by I.N.F.M.

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