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Universality of the dynamic crossover in glass-forming liquids: A “magic” relaxation time

V. N. Novikov* and A. P. Sokolov

Department of Polymer Science, The University of Akron, Akron, Ohio 44325-3909

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Analysis of experimental data on the structural relaxation time τ_α in various glass formers revealed its universality at the critical temperature T_c of the mode-coupling theory. In most glass formers studied $\ln \tau_\alpha(T_c) = -(6.5-7.5)$. Possible reasons for such a universality are discussed.

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The nature of the glass transition phenomenon still remains poorly understood. Research in the field of the glass transition for many decades has been concentrated in the temperature range around and below the conventional glass transition temperature T_g [1,2]. The main question asked was what kind of transition would occur if the glass-forming system was cooled extremely slowly.

However, already in the 1960s, a couple of groups proposed [1,3,4] that something might be happening with the glass-forming liquids at temperatures much above T_g. In 1963, Boyer and co-workers proposed the existence of a liquid-liquid transition in polymeric systems at $T \sim 1.2\ T_g$. They collected a body of experimental evidence [5] (a portion of which appears to be wrong [6]) and ascribed the transition to a third order thermodynamic phase transition. In 1969, Martin Goldstein proposed [3] that a crossover from liquidlike dynamics to a viscous flow driven by over-barrier relaxation should occur at some temperature much above T_g. In other words, Goldstein proposed a dynamic crossover in glass-forming liquids that, according to his estimates [3], should occur when the structural relaxation time $\tau \sim 10^{-9}$ s.

Developments of the mode-coupling theory (MCT) of the glass transition in the mid 1980s [7,8] attracted significant attention of researchers to the temperature range much above T_g. The applicability of the MCT approximation to a description of the dynamics of glass-forming systems still remains a subject of heated debate [9,10]. Regardless of that, it is the theory responsible for shifting attention from the region around T_g to the temperature range above it. The theory predicts the existence of a dynamic crossover, i.e., a transition from a liquidlike to a solidlike dynamics on a molecular level, at some critical temperature, T_c, above T_g. The theory relates the transition to a qualitative change in a caging phenomenon (the motion of a particle in a cage formed by its neighbors) and provides clear predictions for the dynamics of the glass-forming liquids at temperatures around T_c. Neutron and light scattering spectroscopy, dielectric relaxation [8], time-domain measurements by the optical Kerr effect [11], and computer simulations [12–14] were all used for tests of the MCT predictions. Experiments and simulations have been done on many different glass-forming systems including small molecule [15–60], polymeric [61–68], ionic [69–84], covalent [85–87], orientational [88], and colloidal systems [89], as well as water [90] and biopolymers [91]. It has been found that at high temperatures ($T > T_c$) the dynamics of these widely different systems follow reasonably well the scenario suggested by MCT [7]. However, it fails (at least in the idealized approximation) to describe the dynamics at temperatures below T_c.

T_c has been estimated for large variety of materials. More importantly, it has been found that many dynamic properties of glass-forming liquids exhibit qualitative changes at temperatures around T_c. Decoupling of the α relaxation (the main structural relaxation) and slow β relaxation (a secondary relaxation) processes happens for most glass-forming systems at $T \sim T_c$, although counterexamples exist [92]. Decoupling of rotational and translational diffusion also occurs at $T \sim T_c$ [59]. Analysis of the Debye-Waller factor also demonstrates some change at $T \sim T_c$ [87,93], although one should note that this result depends strongly on the data analysis. A particular derivative analysis of temperature variations of the structural relaxation time τ_α, proposed by Stickel *et al.* [16,94], clearly demonstrates qualitative changes in the relaxation behavior at some temperature $T_b \sim T_c$. Recent analysis of temperature variations of positron annihilation data also demonstrate qualitative changes at temperatures around T_c [95]. It has been also found that temperature variations of dielectric relaxation strength change around T_b [96]. Thus, at present, there is a long list of evidences that some qualitative changes occur in dynamics of glass forming-systems in a particular temperature range above T_g.

The present paper analyzes literature data on the α-relaxation time in different glass-forming systems at the critical temperature of MCT. We carefully searched the literature for all papers where estimates of the crossover temperature T_c (using MCT approximation) has been done for various glass-forming systems. Recently, Beiner *et al.* collected estimates of crossover temperature and a crossover frequency for a large number of glasses. Most estimates have been done using the temperature of the decoupling of α and β processes. Also MCT’s T_c and Stickel’s T_b were used for many systems, and the temperature of the rotational-translational diffusion decoupling has been used in a few cases. We want to stress that the temperature of the α-β decoupling may depend strongly on an approximation used for the spectra analysis (see, e.g., Ref. [97], where two different approximations have been used). Also, decoupling of the rotational and translational diffusions does not provide a

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*On leave from Institute of Automation & Electrometry, Russian Academy of Sciences, Novosibirsk 630090, Russia.

TABLE I. The values of glass transition temperature T_g, critical temperature T_c, ratio T_c/T_g, and fragility m.

	T_g	T_c	T_c/T_g	m [106]	$-\ln \tau_c$
Small molecules					
Propylen glycol	167	251 [15]	1.50	52	7.5 [16,17]
Propylen carbonate	158	176 [18], 187 [19,20], 196 [21]	1.11–1.24	104	5.2–7.7 [16,20,22]
Orthoterphenyl (OTP)	243	285 [23], 290 [24], 293 [25]	1.17–1.19	81	7–7.8 [16]
Salol	218	256 [26], 263 [27], 266 [28], 275 [29]	1.17–1.26	73	6.6–8.1 [16]
Glycerol	186	225 [30], 223–233 [31], 262 [32], 288 [33], 300 [34]	1.18–1.61	53	4.4–9 [16,35–38]
Sorbitol	264	309 [39]	1.17	93	6.5 [40,41]
Toluene	118	143 [42], 153 [43,44]	1.21–1.30	107	7–8 [45]
m-fluoroaniline	173	212 [46]	1.22	109 [47]	7.3 [46], 7.8 [40,47,48]
picoline	133	162 [49]	1.22		7.5 [49]
n-butylbenzene	128	150 [50], 160 [18]	1.17–1.25		6–7.2 [18]
m-tricresyl phosphate	210	260 [51]	1.24	63	7 [16]
dibuthylphthalate	170	227 [52]	1.34	69	6.8 [53]
isopropylenbenzene	125	150 [54]	1.20		5.8 [55]
2,4,6-trimethylheptane	125	150 [56]	1.20		6.3 [56]
$\alpha\alpha\beta$-tris-naphtylbenzene (TNB)	345	407 [57], 410 [58], 415 [59]	1.18–1.19	86 [60]	6.6–7 [60]
Polymers					
PB	180	216 [61,62]	1.20	59 [63]	7.1 [61]
PPG	200	250 [64]	1.25	117	7.1 [64,65]
PIB	200	270 [66]	1.35	46	6.5 [67]
PS	370	420 [66]	1.14	139	6 [68]
Ionic systems					
$[Ca(NO_3)_2]_{0.4}[KNO_3]_{0.6}$ (CKN)	333	368 [69–71], 375 [72,73], 378 [74–76], 388 [77]	1.10–1.14	93	6.3–7.5 [78,79]
$[Ca(NO_3)_2]_{0.4}[RbNO_3]_{0.6}$ (CRN)	333	365 [73], 378 [80]	1.10–1.13	97 [79]	6.6–7.5 [79]
$ZnCl_2$	375	563 [81]	1.50	30	7.9 [81]
$Na_{05}Li_{05}PO_3$	515	620 [82]	1.20		6.5 [82]
$LiCl/H_2O$, 13 m%	135	164 [83]	1.21	67 [84]	6 [84]
Covalent systems					
B_2O_3	526	800 [85], 800–900 [86]	1.52–1.71	32	6.2–6.7 [87]
Orientationally disordered crystals					
$(NPA_{0.7}NPG_{0.3})$ [88]	156	227	1.45	30	6

very accurate estimate of the crossover temperature and may depend on the guest molecule used in some of these measurements. That is the main reason why here we choose to focus on a better defined temperature obtained using the MCT analysis (T_c). Table I presents values of T_g, T_c, and the structural relaxation time τ_α at T_c (τ_c). The data are collected for 26 different systems that we were able to find in the literature. These include molecular and polymeric glass formers, covalent, hydrogen-bonded, ionic, van der Waals, and orientationally disordered systems. For many systems Table I includes data from various methods and groups. Only glycerol data show significant scattering of estimates of T_c and τ_c. We think that the lowest estimates of T_c obtained in Refs. [30,31] are not correct due to wrong light scattering data used (this problem of the light scattering has been discussed in Refs. [98–100] and new light scattering spectra of glycerol [101] differ strongly from the data presented in Refs. [30,31]), and also the data from Ref. [34] overestimate T_c due to very limited frequency range analyzed. Data for other systems have much smaller scattering (Table I).

In many papers one can find a claim that usually $T_c \sim 1.2T_g$. Analysis of the data presented in Table I clearly shows that this is not a general case: T_c scatters from $\sim 1.14T_g$ in the case of CKN up to $\sim 1.6T_g$ in the case of B_2O_3. It has been already noticed in Refs. [51,102] (see, also, Refs. [103,104]) that the ratio T_c/T_g correlates with the fragility of glass-forming systems: the higher the fragility, the lower the value of T_c/T_g. Fragility is a characteristic of glass-forming systems based on temperature variation of the structural relaxation time τ_α. Systems with slow variations in τ_α with T/T_g are called ‘‘strong,’’ while those with steeper variations in τ_α are called ‘‘fragile’’ [103].

The most interesting and unexpected result presented in Table I is the universal value of τ_α at T_c: It appears that for all systems presented in Table I, $-\ln[\tau_\alpha(T_c)]\sim 7\pm 1$. Analysis of the data presented by Beiner *et al.* [105] shows that most of their data also points to values in the same range, $-\ln \tau_c = 6\text{–}8$. In particular, a majority of the points in Fig. 2 of that paper are concentrated in this region. However, a few strong deviations are obvious. The crossover temperature obtained by different means is expected to be close to the critical temperature T_c of the MCT, but there are clear exceptions. This is especially related to the α-β-splitting region and characteristic relaxation time at this point $\tau_{\alpha\beta}$. For example, in polymers with long side chains $\tau_{\alpha\beta}$ depends on the length of the side chains, and increases up to $\tau_{\alpha\beta}\sim 0.1$ s in poly (n-hexyl methacrylate). This unusual value for $\tau_{\alpha\beta}$ might suggest that in the particular case of these polymers the observed decoupling of two relaxations is not related to the glass transition, but rather to a particular relaxation of the long side chains.

The observed universality of τ_c (Table I) is surprising because the relaxation time in glass-forming liquids varies between $\tau_\alpha\sim 10^{-12}$ s and $\tau_\alpha\sim 10^{3}$ s. Why do the values of $\tau_\alpha(T_c)$ appear to lie in such a narrow universal time interval in different systems, including polymeric and molecular liquids? We want to note that in 1997 Roessler *et al.* proposed a universal scaling of viscosity between T_g and T_x for various molecular glass-forming systems [107]. T_x for all systems analyzed in Ref. [107] appears to be $\sim T_c$. This universal scaling suggests that the viscosity of low-weight molecular systems has a universal value at T_c. These data support our observation of universality of τ_α at T_c. The observed universality agrees with the idea put forward by Goldstein that at some particular relaxation time a crossover from liquidlike to a solidlike dynamics should occur. He predicted $-\ln \tau_c = 9$ (Ref. [3]), a value that deviates by roughly two orders of magnitude from the value found here.

How can one explain this "magic" relaxation time that marks a crossover in dynamics of glass-forming liquids from liquidlike to solidlike? There is no theory currently that predicts this universality. The traditional approximation for temperature variations of τ_α is the Vogel-Fulcher-Tamman equation [108–110]:

$$\tau = A\exp[DT_0/(T-T_0)]. \tag{1}$$

Here $A\sim 10^{-16}$ s, D is related to fragility of the system and T_0 is a temperature below T_g. Since $\tau_\alpha(T_g)\sim 10^2$ s, from Eq. (1) it follows that $DT_0 = 18(T_g-T_0)\ln 10$. Using this relationship, one can rewrite Eq. (1) at T_c as

$$\tau_c = A\exp\left[\frac{18(T_g-T_0)\ln 10}{T_c-T_0}\right] = 10^{-16+18(T_g-T_0)/(T_c-T_0)}. \tag{2}$$

Thus, universality of τ_α thought of in this way suggests universality of $(T_g-T_0)/(T_c-T_0)$. Indeed, it was found in a few papers that $(T_c-T_0)/(T_c-T_g)\sim 2$ [104,111]. This observation leads to the relationship $(T_g-T_0)/(T_c-T_0)\sim 1/2$ and gives an estimate

$$\tau_c \sim 10^{-7}\ \text{s},$$

i.e., the value found in our analysis. We should stress that the quantitative coincidence is rather fortunate, because our estimate [Eq. (2)] is very rough and an average value of the ratio $(T_c-T_0)/(T_c-T_g)$ for various systems was found to be ~ 1.7 [111]. Nevertheless, this simple consideration based on an empirical relationship among T_c, T_g, and T_0 gives strong support to the contention that a "magic" relaxation time exists at T_c. This consideration, however, does not explain the observed universality.

Another idea may be related to an analysis of a process by which a particle (an atom) escapes from a cage formed by its neighbors. The time of escape is directly related to the structural relaxation time τ_α. The probability of a particle to escape from the cage depends on the motion of its neighbors. At high enough temperatures, all particles are moving with large mean square amplitude r. As a result, on a time scale of τ_α, a particle can always find an open space and move between its neighbors, i.e., to find a "doorway" and escape from the cage. This escape can be considered as a simple jump over a rather small, temperature independent, energy barrier, E. The mean square amplitude of atomic motion decreases with decreasing temperature, lowering the probability of a particle finding the doorway. At some temperature r becomes so small that the particle cannot escape without pushing its neighbors out of the way. In order to do that, the particle needs a higher energy to overcome the repulsion forces. This leads to an effective increase in the barrier height for the relaxation. This simplified picture with a "doorway" was analyzed in Ref. [112]. It suggests that temperature variations of τ_α will be nearly of an Arrhenius form at high temperatures and will vary much steeper below T_c.

The universality of τ_α at T_c in this picture can be connected to another universal parameter—the Lindemann ratio [113,114]. According to the Lindemann criterion of melting, the mean square atomic displacement, r, has a universal value in units of interatomic distance, a, at the melting point in a crystal, $r\sim(0.12\text{–}0.15)a$. In supercooled liquids, the critical temperature T_c may play a role of the melting temperature [7], because a liquid is frozen to a solid at this point within the framework of the idealized MCT. Indeed, it has been shown recently that in hard sphere systems at the critical temperature, $r = 0.15d$, where d is the diameter of the sphere [115]. Also, computer simulations for binary Lennard-Jones (LJ) system [116] predict $r/a = 0.14$ at T_c, in good agreement with the Lindemann ratio. Thus, we assume that at $T = T_c$, $r = ba$ where parameter b has a value that is close to the Lindemann ratio $b_L = 0.13\text{–}0.16$ for many glass formers. It is clear that this assumption cannot be applied to the glass formers with $T_c > T_m$, such as silica: in these cases the value of $r(T_c)$ should be higher than that expected from the Lindemann ratio. Actually, $r(T_c)/a = 0.22\text{–}0.24$ was found for silica in computer simulations [13]. It is interesting that according to Ref. [117], this amplitude of the mean-square displacement corresponds to a vanishing of the shear modules in a LJ crystal, or to the so-called Born criterion of melting.

We use the following simple model to connect the Lindemann criterion with the relaxation time at the critical temperature. We assume that near and above T_c collective effects became unimportant for the α relaxation and the latter can be described by an activation process in which a single atom jumps over a barrier of height E in a harmonic potential well. Thus, at the critical temperature the relaxation time may be approximated by an activation exponential with some activation energy E, i.e., $\tau_c=\tau_0\exp(E/T_c)$. Assuming that the typical jump length is of the order of the interatomic distance, one has the following connection between the activation energy and the interatomic distance

$$k(pa/2)^2/2\sim E, \tag{3}$$

where k is a force constant and $p\sim 1$ is the numerical coefficient that characterizes the typical length of the atomic displacement in this process. When a particle is trapped in a cage, at a given temperature T the average value of the mean square displacement amplitude $\overline{u^2}$ in this harmonic potential is connected with temperature by the equation

$$k\overline{u^2}/2\sim T/2. \tag{4}$$

From Eq. (4) taken at $T=T_c$ (where $\overline{u^2}=b^2a^2$) and Eq. (3) it follows that

$$\frac{E}{T_c}=\frac{3p^2}{8b^2}, \tag{5}$$

and hence

$$\tau_c=\tau_0\exp(3p^2/8b^2). \tag{6}$$

With b equal to the typical Lindemann value, $b\sim 0.13–0.15$, one has $E/T_c\sim(17-22)p^2$, or

$$\ln\tau_c=\ln\tau_0+(7–9)p^2. \tag{7}$$

If one assumes that τ_0 is a characteristic vibration time, $\tau_0\sim 10^{-13}$ s, then the value of $p\simeq 0.9$ will lead to the observed universal value $-\ln\tau_c=6–8$. This shows that if the Lindemann criterion is fulfilled at the critical temperature, the relaxation time τ_c is fixed with the same degree of universality as the Lindemann ratio. Some small additional scattering may come from τ_0 values and the specific local structure of the glass former. Let us note that the expression for $\tau(T)$ in this model can be written in the form

$$\tau(T)=\tau_0\exp[u_0^2/\overline{u^2}(T)], \tag{8}$$

where u_0 is the jump length. Recently, such an expression for the relaxation time was confirmed in a computer simulation study of a glass-forming polymer melt [118]. Equation (8) is also similar to the expression proposed by Buchenau and Zorn in Ref. [119], where instead of $\overline{u^2}$, a difference between $\overline{u^2}$ and the value of the mean square displacements in the respective crystal has been used. The expression that we are using is in agreement with the Vogel-Fulcher law. Indeed, the experimental data on many glass formers show that at $T>T_g$, $\overline{u^2}(T)\simeq(T-T_0)/k_L$, where k_L characterizes the slope of the $\overline{u^2}(T)$ dependence for $T\gg T_g$, and T_0 is a constant, close to the Vogel-Fulcher temperature, $T_0<T_g$. Although this model is simple enough, it can shed some light on the origin of the universality of τ_c in various glass formers, at least in the cases of high and intermediate fragility.

Our analysis demonstrates that the structural relaxation time τ_α at T_c appears to be the same in most systems, including those that are covalent and hydrogen bonded, ionic, molecular, and polymeric. In other words, there is a ‘‘magic relaxation time’’ at which the dynamics of most glass-forming systems change qualitatively. Some ideas that might explain this universal τ_α have been presented. The question remains open if this universality can be explained by some universal properties of the free energy landscapes of glass formers.

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