Universal aspects of the structural glass transition from density functional theory

T.R. Kirkpatrick\(^1\) and D. Thirumalai\(^2\)

\(^1\)Institute for Physical Science and Technology, University of Maryland, College Park, MD 20742
\(^2\)Department of Chemistry, University of Texas at Austin, Austin, TX 78712

(Dated: September 23, 2024)

The random first order transition (RFOT) of the structural glass transition (SGT) can be formulated as a density functional theory (DFT). An important feature of the complete RFOT theory is that it has at least two distinct transition temperatures, one of which is a dynamical transition signaling loss of effective ergodicity, and the other is an equilibrium ideal glass transition. The dynamical transition exists only in mean-field-like theories and becomes a rounded transition when activated transport processes become prominent. Using scaling and renormalization group (RG) ideas, universal aspects of both of these transitions are discussed. Particular attention is paid to the coherence, or correlation, length associated with these transitions in the RFOT. We also derive universal scaling relations for several experimentally measurable quantities that are valid near the ideal glass transition. In particular, activated scaling ideas are used to obtain a scaling equation for the non-linear structural glass susceptibility as the ideal glass transition is approached. Important finite corrections to the ideal glass transition temperature are also discussed. Our work provides additional criteria for assessing the validity of RFOT theory of the liquid to glass transition, and provides a firm theoretical foundation for analyzing experimental data on the temperature dependence of the relaxation times.

I. INTRODUCTION

When undercooled rapidly almost all liquids undergo a transition to a glassy state, characterized by a dramatic increase in viscosity over a narrow temperature range. Because of the central importance of this phenomenon in condensed matter physics, material science, and biology there has been considerable interest in understanding the characteristics of the structural glass transition (SGT)\(^1\)–\(^6\). In the late 1980s, we along with Wolynes developed the random first order phase transition (RFOT)\(^7\) theory of the glass transition\(^8\)–\(^12\) to explain many of the unique aspects of the structural glass transition (SGT) problem. Although initially motivated by precise solutions of a class of infinite range spin glass models (see\(^1\) for a short review), we subsequently used a density functional approach and established that the generic aspects of RFOT can be found in systems even without quenched disorder\(^13\). It was further argued that the structural glass transition is an example of a RFOT. The emergence of RFOT ideas in a theory without quenched disorder is an important development because it showed that liquid state theories could form the basis for describing glassy states. Indeed, the RFOT has been much discussed in this context and in many ways clarified and further extended\(^14\)–\(^17\).

There are necessarily at least two\(^8\),\(^9\) distinct transitions within the RFOT theory in the mean-field limit. When the temperature is decreased, the liquid undergoes the so-called dynamical transition at a temperature, \(T_d\). This transition is, in general, not a true equilibrium phase transition, although it is related to the topology of the state space of an equilibrium system in that at \(T_d\) there is a breakdown in effective ergodicity. Technically, it can be described by using either an equilibrium approach\(^13\),\(^18\),\(^19\), or a dynamical approach\(^11\),\(^20\)–\(^22\). The dynamical approach is consistent with the mode coupling theory glass transition\(^20\),\(^21\),\(^23\) in that both the theories show that the nature of relaxation dynamics changes at \(T_d\). The dynamical order parameter of the theory is a freezing of the density fluctuations To further characterize the nature of the transition at \(T_d\), we introduced\(^24\) a non-linear susceptibility or four point correlation function \(\chi(t)\) where \(t\) is time), which is the natural susceptibility for this transition\(^17\),\(^24\),\(^25\). We showed that \(\chi(t)\) at \(T_d\) is also a measure of broken ergodicity - a finding that has been confirmed in computer simulation studies\(^26\). A number of studies have also shown that \(\chi(t)\) also gives information on the dynamical heterogeneity in glassy liquids\(^27\)–\(^29\).

The second transition within RFOT theory is a true equilibrium phase transition occurring at a lower temperature denoted by \(T_K\), for historical reasons. The transition at \(T_K\) is a new type of equilibrium phase transition, where the order parameter is both random and, in a technical sense, discontinuous at \(T_K\), and one that has a unique and different coherence or correlation length exponent associated with it compared to other well-studied phase transitions.

In\(^7\) (see also\(^10\)) it was argued that in realistic, non-mean-field, systems the transport for \(T < T_d\) is activated and that the driving force is entropic. The eventual freezing at the ideal glass transition occurs because the entropic driving force vanishes at \(T_K\). This and various scaling arguments were the final ideas in the original\(^7\) RFOT theory.

More recently\(^30\)–\(^32\) there has been an enormous amount of work on liquid systems that can be exactly solved in the high-dimension limit \((d \to \infty)\) and that undergo a SGT. As in earlier work, these theories find two transitions; (i) an ergodicity breaking dynamical transition and (ii) a true equilibrium ideal glass transition. The universal features of these transitions are the same.
as in the original RFOT of the SGT. It is interesting, and possibly relevant, to note that the fundamental variables in these theories are all related to particle displacements, and not directly to density fluctuations (see however\textsuperscript{33}).

In recent years there has also been a lot of work on the thermodynamics of metastable or non-equilibrium states within RFOT. This is motivated by the fact that on a restricted time scale, that gets longer and longer further into the glassy phase, these states are almost like equilibrium states. This enlarges the parts of phase space that can be probed by theoretically compressing a fluid. Among other things, this has led to a connection between the glass transition and the so-called jamming transition\textsuperscript{32,33}. This work has emphasized that the existence of a Kauzmann temperature is not central to what is referred to as the Gardner transition\textsuperscript{35,36} and the subsequent jamming transition.

The importance of $T_d$ was already anticipated by Goldstein\textsuperscript{37} (see also\textsuperscript{38}), who argued that below a certain temperature transport in liquids is determined by activated transitions. Since then there is overwhelming evidence from theoretical, simulation\textsuperscript{26}, and experimental\textsuperscript{39} studies that there is a material-dependent special temperature, $T_d$, or temperature region, below which ergodicity breaking, dynamic heterogeneity, aging effects play an important role. This temperature, $T_d$, is generally well above that which is conventionally referred to as the Gardner transition and the so-called jamming transition\textsuperscript{32,33}.

The contents of this paper are as follows. In Section II we discuss the important characteristics of a RFOT, with an eye towards the DFT of the SGT and on the two distinct (dynamical and equilibrium) transitions within RFOT. Using scaling and RG ideas we obtain previously unnoticed scaling relations and scaling predictions for the SGT. Activated dynamical scaling ideas are used to predict a number of effects that should occur as $T_K$ is approached. Important finite size corrections to the ideal glass transition temperature are also discussed. In Section III, we compare a RFOT transition with more conventional phase transition theories for the structural glass transition. In this section we also make a general argument for a correlation exponent inequality. We conclude in Section IV with a discussion.

II. CHARACTERISTICS OF THE RFOT THEORY OF THE SGT

A. The order parameter and state degeneracy

The glassy state may be visualized as a frozen liquid state with elastic properties. To describe a glassy state, we introduce two key ideas. First, we imagine an order parameter description in terms of frozen density fluctuations, $\delta n = n - n_l$, where $n$ is a local number density, and $n_l$ is the spatially average density that is identical to the liquid state density. Other order parameters can be imagined, but frozen density fluctuations are the simplest, and are most directly related to an important characteristic of a solid: elastic properties and a nonzero Debye-Waller factor. Because the glassy phase is amorphous or has random characteristics, the frozen density order parameter is specified by a functional probability measure $DP[\delta n]\textsuperscript{13,14,40}$.

The second key idea in the formulation of RFOT is that, in general, one expects a large number of distinct metastable glassy states as the dynamics starts to become sluggish. If the number of low free energy metastable states is large enough, this in turn leads to two distinct transitions. The physical arguments leading to this conclusion proceed as follows. We denote a particular glassy state by the label $s$, with the frozen density in that state, $n_s = n_l + \delta_n$, and the free energy in that state being equal to $F_s$ [the density fields in Eqs. (1) and (2) should also be labelled by $s$]. The first two moments of the order parameter are,

$$\overline{\delta n_s(x)} = \int DP[\delta n] \delta n_s(x) = \frac{1}{V} \int dx \delta n_s(x) = 0,$$  \hspace{1cm} (2.1)

and

$$q_{ss'} = \int DP[\delta n] \delta n_s(x) \delta n_{s'}(x) = \frac{1}{V} \int dx \delta n_s(x) \delta n_{s'}(x).$$

The final equalities in these equations assume self-averaging. From Eq.(2.2) it follows that the zero in Eq.(2.1) is a term of $O(V^{-1/2})$ with $V$ the system volume and the bulk limit is always taken.

Exact calculation for some infinite range spin-glass models as well as for infinite dimensional liquid models\textsuperscript{32} show that below a temperature, which we denote by $T_d$, there are an extensive number (the number of states scales like $\exp[\alpha N]$ for a $N$ particle system) of global statistically similar incongruent metastable glassy states\textsuperscript{41}. In the RFOT, it is assumed that, in a restricted sense (cf below), this feature also holds in realistic structural glass systems. Statistically similar states have the same spatially averaged correlation functions, and incongruent states have zero overlap,

$$q_{ss'} = \delta_{ss'} q.$$

Because the states are statistically similar one cannot simply use an external field to pick out a particular state,
as could be done in a regular Ising model for example. The canonical free energy, $F_c$, is then given by the partition function via

$$Z = \exp[-\beta F_c] = Tr \exp[-\beta H] = \sum_s \exp[-\beta F_s]. \quad (2.4)$$

In the STG, there are two important cases when $F_c$ is not the physical free energy. First, if the barrier between states is actually infinite then $F_c$ cannot be a physically meaningful free energy. Second, if the barriers are finite but the experimental time scale is too short for fluctuations to probe the various states, then it is also not a physical free energy. These considerations also apply to cases where there is a multiplicity of long-lived metastable states in any system.

A component averaged free energy can be defined by

$$\bar{F} = \sum_s P_s F_s \quad (2.5)$$

with $P_s$ the probability to be in the state $s$,

$$P_s = \frac{1}{Z} \exp[-\beta F_s] \quad (2.6)$$

$F_c$ and $\bar{F}$ are related by

$$F_c = \bar{F} + T \sum_s P_s \ln P_s = \bar{F} - TS_s. \quad (2.7)$$

Here, $S_s$ is the state entropy (sometimes called the complexity, $I$), which is bounded from above by what is usually meant by the configurational entropy, $S_c$, in non-mean-field models where there will be transitions between the various states. In general, $S_s$ is related to the solution degeneracy and is extensive (and $F_c \neq \bar{F}$) if there are an exponentially large number of states. Note that in infinite range models with a RFOT and a nonzero $S_s$ the physical free energy is $\bar{F}$ because the $S_s$ in Eq. (2.7) is an entropy term which is a measure of parts of state space not explored in a finite amount of time. Since a physical entropy should only be associated with accessible configurations, it follows that $F_c$ is not a physically meaningful free energy.

The scenario for the two transitions in the RFOT theory is obtained as follows. For $T > T_d$ transport is not collective, and the topology of state space is trivial. However, as $T \to T_d^+$ the dynamics slows down because at $T_d$ an extensive number of statistically similar, incongruent globally glassy metastable states emerge. If activated transport is neglected, these states are infinitely long lived. The liquid state free energy, $F_l$ is lower than the physical glassy state free energy, $\bar{F}$, but it is equal to the canonical free energy, $F_c$. Because there is a multiplicity of glassy states, a liquid with unit probability will get stuck in one of the metastable glassy states at temperatures below $T_d$. If activated transport is ignored, it will remain in that state for all times. For infinite range models with an RFOT, exact dynamical calculations shows a continuous slowing down and freezing occurs as $T \to T_d^+$. The same result is also found for some approximate, mean-field like, calculations of dynamics in realistic liquid state models even in the absence of quenched randomness\textsuperscript{13}. The transition at $T_d$ is also closely related to the temperature at which mode coupling theory of the glass transition predicts a power law divergence of the relaxation times.

In realistic systems, with particles interacting with short-range interactions, activated transport does take place at $T < T_d$. The change from diffusive to sluggish dynamics, which occurs at surprisingly low viscosities, is the reason $T_d$ is associated with a dynamical transition. It is a sharp transition only in infinite range models, but in general it sets a temperature at which the dynamics becomes glassy like. In support of this interpretation, whose origins in retrospect can be traced to the insightful arguments given by Goldstein\textsuperscript{37}, there is considerable experimental evidence that at $T_d$ the nature of transport changes from being diffusive to activated\textsuperscript{39}. As a result, it is our view, that theories (for example\textsuperscript{42}) in which the analogue of $T_d$ does not naturally emerge cannot describe the STG.

The driving force for the activated transport in the RFOT theory for temperatures less than $T_d$ is entropic, and is given by the state or configurational entropy, $S_s$, Eq. (2.7). At a lower temperature denoted by $T_K$, after the so-called Kauzmann temperature, the configurational entropy vanishes, and transport ceases. In other words, the second transition at $T_K$ is the ideal or equilibrium glass transition temperature. Fits to viscosity data for an extremely wide class of materials point to the existence of non-zero $T_K$ (for a compilation of experimental data see Ref.\textsuperscript{43}).

**B. The dynamic transition and the associated correlation length**

We characterize the dynamical transition as follows. Consider the dynamical order parameter,

$$q(x-y,t_1-t_2) = \langle \delta n(x,t_1) \delta n(y,t_2) \rangle, \quad (2.8)$$

Above $T_d$, this correlation decays as $t \to \infty$ but as $T \to T_d^+$ the decay gets slower and slower in a power law fashion. At $T_d$ it no longer decays, (except in non-mean-field models) on the longest time scale. Effectively, it is the Edwards-Anderson order parameter for the glass transition:

$$q_{EA} = \lim_{t \to \infty} q(0,t). \quad (2.9)$$

Because the order parameter involves the square of the density fluctuations it is clear that the susceptibility will be non-linear involving higher order correlations\textsuperscript{24},

$$\chi_{NL}(x-y,t_1-t_3,t_4-t_2,t = t_4 - t_1) = \langle \delta n(x,t_1) \delta n(y,x,t_3,t_4) \rangle. \quad (2.10)$$

Where $\langle \rangle_c$ denotes cumulant average where all pairwise density correlations are subtracted out. In an exactly
soluble spin-glass model we have previously shown\textsuperscript{24} that the 'static' susceptibility for the glass transition at $T_d$ is

$$\chi_{NL}(k, t < \tau_c) = \int d(t_1 - t_3) d(t_4 - t_2) d(x - y) \exp(-i k \cdot (x - y))$$

$$\chi_{NL}(x - y, t_1 - t_3, t_4 - t_2, t = t_4 - t_1 < \tau_c),$$

(2.11)

where $\tau_c$ is the time that the plateau exist in $q(t)$ (it diverges at $T_d$ in the mean-field limit). As $r = T/T_d - 1 \to 0^+$, the homogeneous static susceptibility, $\chi_{NL} = \chi_{NL}(k \to 0)$, in a mean-field like theory, diverges as\textsuperscript{24},

$$\chi_{NL} \sim 1/\sqrt{r},$$

(2.12)

and at finite and small wavenumber,

$$\chi_{NL}(k) \sim 1/[k^2 + \xi_o^{-2} \sqrt{r}]$$

(2.13)

with $\xi_o$ a microscopic (correlation) length. This in turn defines a divergent length scale as $T \to T_d^+$ given by,

$$\xi \sim \xi_o/r^{1/4}.$$  

(2.14)

This is the same divergence seen at a mean-field spinodal point. These same results with $r \to |r|$ were obtained\textsuperscript{10} in an exactly soluble spin-glass model for $r = T/T_d - 1 \to 0^-$.

The limiting procedure implied by Eq.(2.11) is crucial to obtain the correct diverging non-linear susceptibility result\textsuperscript{24}. In\textsuperscript{24} it was suggested that this is a reasonable definition of an ergodic to non-ergodic transition.

\section*{C. The equilibrium transition and its correlation length}

According to RFOT, the ideal glass transition at $T_K$ is a discontinuous transition. Hence, care must be taken in defining and interpreting a correlation or coherence length. For ordinary first order phase transitions (liquid to a periodic crystal for example) Fisher and Berker (FB)\textsuperscript{14} have formally described the meaning of such a length scale. They conclude that there are two distinct interpretations of a divergent length, referred to as coherence or persistent length for a first order transition. The first, using a scaling analysis and the observation that the definition of long range order is consistent with a correlation length exponent given by $\nu_H = 1/d$, with $d$ being the spatial dimension. The second is via a finite-size scaling analysis and the divergent length is the length scale where the sharp first order phase transition becomes rounded in a finite system. This correlation length exponent is also given by $\bar{\nu} = 1/d$.

Here, we generalize the arguments of FB to a RFOT, further elaborating on our previous study\textsuperscript{7}. We use the density order parameter discussed in Section II.A. For the conjugate field we use a chemical potential $h = \mu(T, p) - \mu(T_K, p_K)$, allowing for the location of the glass transition to depend on both temperature and pressure.

We then give two further distinct arguments for the exponent characterizing the divergence of the correlation length. The first is an exact calculation for an infinite range spin-glass model with a RFOT that gives an identical result. The final argument that gives the same exponent is a partly dynamical/nucleation-like one and is presented in Sec II.E. All together there are four separate arguments that give the same result.

\subsection*{1. Scaling analysis}

If we define a renormalization group (RG) length rescaling factor by $b$ then the conjugate field $h$ should be associated with the largest RG eigenvalue, $b^\lambda$, and therefore should scale as,

$$h' = b^\lambda h$$

(2.15)

while the free energy density will scale like,

$$f(h) = b^{-d} f(h').$$

(2.16)

In addition, the scaling of the correlation length should by definition follow,

$$\xi(h) = b\xi(h').$$

(2.17)

If we choose $b = h^{-1/\lambda}$ then we formally obtain,

$$f(h) = h^{d/\lambda} f(1),$$

(2.18)

and

$$\xi(h) = h^{-1/\lambda} \xi(1).$$

(2.19)

Now, the order parameter $q$ is related to the second derivative of $f$ with respect to $h$ or,

$$q \sim h^{d/\lambda - 2}.$$  

(2.20)

At the RFOT, $q$ is discontinuous which gives $\lambda = d/2$. Equation (2.19), on the other hand implies that the correlation length exponent is $\nu_h = 1/\lambda$, or

$$\nu_h = 2/d.$$  

(2.21)

These considerations establish, rather rigorously, that as long as long as FB type scaling holds near a first order transition it is inevitable that the correlation length should diverge at $T_K$ with an exponent $\xi$ at RFOT. The difference in the values of the correlation length exponent is due to the differing nature of the order parameter describing RFOT and first order transitions. In the first order transition density itself is discontinuous whereas in RFOT at $T_K$ it is the square of the density (EA parameter) that changes abruptly.
2. Finite size effects

Next we consider a system with finite size, \( L \), and do a finite size scaling argument. We postulate that the scaling part of the free energy behaves as,

\[
f(h, L) \sim L^{-\zeta} f(L/\xi) = L^{-\zeta} Y(h L^{1/\nu})
\]  

(2.22)

where \( \xi \sim h^{-\bar{\nu}} \) is the (finite-size correlation or coherence length) length scale where finite size rounding of the RFOT takes place at \( T_K \). By noting that the order parameter \( q \) is related to the second derivative of \( f \) with respect to \( h \), we find,

\[
q \sim L^{2/\bar{\nu} - \zeta} Y''(h L^{1/\nu})
\]  

(2.23)

The discontinuous nature of the transition gives,

\[
\zeta = 2/\bar{\nu}
\]  

(2.24)

Note that if we used dimensional analysis to obtain \( \zeta = d \) then it immediately follows that

\[
\bar{\nu} = 2/d
\]  

(2.25)

As an alternative, we follow the FB treatment, and examine the susceptibility, \( \chi \), for this transition. Because the order parameter for the transition already involves two density fluctuations, the susceptibility, as used in Section I.C, will involve four density fields. Accordingly, it is related to the fourth derivative of \( f \) with respect to \( h \). The finite size scaling relation will then be something like,

\[
\chi(h, L) \sim L^{2/\bar{\nu} - \zeta} Y'''(h L^{1/\nu})
\]  

(2.26)

One can then argue that \( \chi \) will attain a maximum value at \( h = 0 \), and will scale as \( \sim L^d \). This in turn again gives \( \bar{\nu} = 2/d \). The inescapable conclusion is that the arguments leading to \( \bar{\nu} = 2/d \) are robust, and theoretically well-founded.

3. Exact results for an infinite range model

Finally, there is an exact calculation for an infinite range p-state (\( p > 4 \)) Potts spin glass model with a RFOT\(^{10,11} \) that gives a length scale that diverges with a correlation length exponent of \( \nu = 2/d \) as the transition is approached. In particular, it has been shown that as \( T_K \) is approached from above the complexity or state entropy (which here and below we denote by the configurational entropy, \( S_c \)) behaves as,

\[
S_c \sim \exp[N\epsilon]
\]  

(2.27)

with \( N \) the number of lattice sites (\( N \to \infty \)) and \( \epsilon = 1 - T/T_K \) the dimensionless distance from the RFOT. On the other hand, as \( T_K \) is approached from below one finds\(^{10} \) that \( S_c \) diverges as,

\[
S_c \sim \exp[1/|\epsilon|]
\]  

(2.28)

If we identify \( N \) with a system volume \( \sim L^d \) then these two equations give a length scale where even below \( T_K \) the system behaves as though it was not frozen. Defining a correlation length, \( \xi \), in this way gives,

\[
\xi \sim 1/|\epsilon|^{2/d}
\]  

(2.29)

or,

\[
\nu = 2/d.
\]  

(2.30)

Notice that this definition of a correlation length is closely related to a finite size correlation length.

4. Finite size shift in the glass transition temperature

Understanding glassy physics in a finite geometry is technologically important, and is needed to analyze results from computer simulations. From a phase transition perspective, many authors starting with the pioneering studies by Ferdinand and Fisher\(^{45} \) have stressed that there are two important finite size effects at any phase transition. The first is the rounding of the transition as described in Sec II.D.2 above. The second distinct effect is the finite size shift in the transition temperature. Naturally, we expect them both to be relevant near RFOTs.

Physically, the \( \epsilon \) (or \( h \)) in the above expressions should be replaced by \( \epsilon = \epsilon - \Delta \epsilon(L) \), where \( \Delta \epsilon(L) \) measures the shift in the glass transition temperature due to the finite size. We give two distinct arguments for the \( L \) dependence of \( \Delta \epsilon(L) \). The first uses that the free energy difference between two coexisting states at the RFOT is of order \( \sim T_K L^{d/2} \). The temperature difference, \( \Delta T \), where the system cannot distinguish between the two states is the total free energy difference, which is proportional to \( \sim L^d \Delta T \). Equating these two free energies we obtain,

\[
\Delta \epsilon(L) \sim \frac{1}{L^{d/2}}
\]  

(2.31)

The second argument hinges on an old result of Imry\(^{46} \) who suggested that at an ordinary first order phase transition the finite size shift in the transition temperature is given by,

\[
\Delta T \sim \frac{1}{L^d s}
\]  

(2.32)

where \( s \) is the latent heat associated with the transition. At an RFOT, though, the latent heat vanishes as the configurational entropy or, \( s \sim \xi^{-1/\nu} \sim L^{-d/2} \) in a finite system. Using this we recover the first argument for \( \Delta \epsilon(L) \).

If the predicted \( L \)-dependent shift in the ideal glass transition temperature can be systematically studied then measuring the above exponent would be an important verification of the RFOT theory. Recent important developments in computer simulations probing the equilibrium glass transition by random pinning of a fraction of particles\(^{47} \) as \( L \) is varied could be a way of verifying Eq. (2.31).
D. The specific heat at the equilibrium RFOT transition

First order phase transitions are associated with strong coupling RG fixed points. As a result, dangerous irrelevant variables are not expected to play any role, which in turn implies that hyperscaling is valid in all dimensions. If the same applies to random first order phase transitions, then the standard hyperscaling relation for the specific heat exponent,
\[ \alpha = 2 - \nu d \]

should always hold. With \( \nu = 2/d \) one then finds that \( \alpha = 0 \) at a RFOT. This scaling result for the specific heat is also consistent with exact calculations for infinite range models with an RFOT. For structural glasses, this result presumably implies a discontinuity in the specific heat, as is always observed experimentally.

E. Transport near the equilibrium RFOT

An important characteristic of a glass transition is the occurrence of extremely long time scales associated with transport below \( T_d \). The critical slowing down at an ordinary transition means that the time scale grows as a power of the correlation length, \( \tau \sim \xi^z \) with \( z \) being the dynamical scaling exponent. In sharp contrast, at a glass transition the critical time scale grows exponentially with \( \xi \),
\[ \ln(\tau/\tau_m) \sim \xi^\psi \]

with \( \tau_m \) a microscopic time scale, and \( \psi \) a generalized dynamical scaling exponent. Effectively, this implies \( z = \infty \). As a result of such extreme slowing down, the equilibrium behavior of the system near \( T_K \) becomes inaccessible for all practical purposes. In other words, the experimentally accessible time scales are not sufficient for the system to reach equilibrium. It is in this sense the glassy system falls out of equilibrium.

Activated scaling, as described by Eq. 2.34, follows from a barrier crossing picture of the system’s free energy landscape. In the context of the STG, it is related to cooperative motions involving particles within a length scale that continues to grow as \( T \rightarrow T_K \). In the temperature range, \( T_d > T > T_K \), there is an entropic driving force that causes a, compact, glassy state of size \( \xi^d \) to make a transition to a different glassy state, with approximately the same free energy, also of size \( \xi^d \). The physical picture that results is a system that looks like a patchwork, of different glassy regions separated by diffuse or fuzzy interfaces slowly making a transition to yet other glassy states. This is also called the mosaic state.

For the uncorrelated states that exist above \( T_K \), the law of large numbers is consistent with a barrier that scale like \( \sim \xi^{d/2} \). This is also consistent with scaling and an entropic driving force \( \epsilon \xi^d \sim \xi^{d-1/\nu} \), if \( \nu = 2/d \). Thus, the driving force and the free energy barrier scale in the same way, if and only if \( \nu = 2/d \). Activated transport with the barriers scaling as \( \sim \xi^{d/2} \) was also recently argued by Langer.

In the original RFOT paper, we modified a wetting argument first given by Villain for the random field Ising model and applied it to the structural glass problem which also led to barriers scaling like \( \xi^{d/2} \). Here, we give a different but related argument. We first imagine a putative compact glassy state of size \( L^d \). There will in general two distinct driving forces that can cause it to make a transition to another glassy state: An entropic one scaling like \( \sim \epsilon L^d \) and a free energy one \( \sim L^{d/2} \) because there are states that typically have a lower free energy by this amount. Opposing the transition is a barrier that naively scales like a surface tension term \( \sim \sigma L^{d-1} \). Now assume a bump (or an excitation) of scale also \( \sim L \) (another scale can be introduced but the final result is not altered) forms on the interface because of the free energy driving force. The free energy change due to the creation of the bump is,
\[ \delta F \sim -L^{d/2} + \sigma L^{d-1} \]

The physical effect of the bump forming is that the surface tension is reduced compared to its naive value. The scale dependent surface tension can be obtained by minimizing \( \delta F \) with respect to \( r \) with the result,
\[ \sigma(L) \sim L^{-(d-2)/2} \]

The barriers to transport therefore scale as \( \sim \sigma(L) L^{d-1} = L^{d/2} \).

All of these arguments in turn imply a Vogel-Fulcher law for relaxation as the glass transition is approached,
\[ \tau \sim \tau_m \exp\left[\frac{D}{T/T_K - 1}\right] \]

with \( D \) a positive constant.

Note that the barriers scaling like \( \xi^{d/2} \) implies there is really no interface between two of the statistical similar glassy states. Rather, to go from one state to another, roughly \( N^{1/2} \) of the \( N \) particles in a correlated volume must be rearranged, as noted earlier by Bouchaud and Biroli.

F. Rare regions near the glass transition

The existence of DH suggests that in a very viscous liquid the longest time decay of any time correlation function will be determined by the large rare region or anomalous clusters of particles of some linear dimension \( L^d \). These large clusters are fluidized and can relax to a
more typical configuration of particles in some characteristic time $\tau(L)$. For this argument to be sensible $L$ must be larger than a molecular scale. To estimate the effect of these large rare regions on a typical time correlation function an average over $L$ must be performed.

Since the large clusters are rare, we assume that their probability distribution is controlled by Poisson statistics so that the tail probability of an unusual cluster of size $L$ is

$$P(L) \propto \exp(-cL^d).$$

We consider a diffusive correlation function, $C(k,t) \propto \exp(-Dk^2t)$, and we assume the relevant wavenumber scales as $k \propto 1/L$, and a scale dependent diffusion coefficient $D(L)$. That is, we consider,

$$C(L,t) \propto \exp(-t/\tau_D(L)),$$

with $\tau_D(L) = L^2/D(L)$ the diffusive time scale. Since $D(L)$ has units of $\text{length}^2/\text{time}$ we take $\tau_D$ to be proportional to the RFOT time scale,

$$\tau_D(L) \propto \tau_{\text{RFOT}}(L) = \tau_m \exp(aL^{d/2}),$$

with $\tau_m$ a microscopic time and $a$ a positive constant.

Using all this the average correlation function deep in the supercooled region then decays for long times as

$$C(t) \propto \int dLP(L) \exp(-t/\tau_m) e^{-aL^{d/2}} \propto \exp[-A(\ln t/\tau_m)^2],$$

with $A$ a positive constant. We conclude that for long times $C(t)$ decays faster than any power law, but slower than any exponential. Note that the saddle point evaluation gives the characteristic length scale $L(t) \propto (\ln t)^{2/d}$.

Finally if we define a distribution of relaxation times, $P(\tau)$, by

$$C(t) \propto \int d\tau P(\tau) \exp(-t/\tau),$$

then,

$$P(\tau \to \infty) \propto \exp[-c(\ln t/\tau_m)^2],$$

again, slower than any exponential.

G. Activated scaling as T approaches $T_K$

Activated scaling was developed to understand finite dimensional (i.e., three dimensions) spin glasses and random field magnets where the dynamics is controlled by large, possibly divergent, free energy barriers. Similar ideas can be applied to the structural glass problem, also in three-dimensions.

Here we examine the behavior of the glass transition susceptibility, introduced in Sec II.C using activated scaling ideas as the ideal glass transition is approached.

First we define a slightly different non-linear susceptibility (see Eqs.(2.10) and (2.11)),

$$\tilde{\chi}_{NL}(k,t) = \int d(x-y) \exp(-ik \cdot (x-y))$$

$$\chi_{NL}(x-y,t,t,0),$$

we then start with the observation, discussed in Sec II.D, that the scale dimension of $g(x,t)$ is zero. This and the activated scaling ansatz implies that the wavenumber and time dependent glass transition susceptibility will satisfy the scaling law:

$$\tilde{\chi}_{NL}(\epsilon,k,t) = b^d F_{\chi} [\epsilon b^{1/\nu}, bk, \frac{b^{d/2}}{\ln(1/\tau_m)}]$$

where $\epsilon = T/T_K - 1$ is the dimensionless distance from the ideal glass transition, $\tau_m$ is some microscopic time scale, and $F_{\chi}$ is a scaling function. Note that we have used here that the barrier height scales as $b^{d/2} \sim \xi^{d/2}$. This equation implies a number of non-trivial results. For example, at zero wavenumber, and at the ideal glass transition temperature we can choose $b = [\ln(t/\tau_m)]^{2/d}$ to obtain,

$$\tilde{\chi}_{NL}(0,0,t \to \infty) \sim [\ln(t/\tau_m)]^2.$$  

This dynamic scaling result is valid as long as $\epsilon \ln(t/\tau_m) < 1$. This in also defines a dynamic crossover $\epsilon$ being given by,

$$\epsilon_x \sim 1/\ln(t/\tau_m).$$

Physically this means that the large correlations that exist at $T_K$ can be measured by examining the slow growth in time of the glass transition susceptibility around $k = 0$. This should be experimentally relevant. If the exponent of 2 in Eq.(2.46) can be experimentally demonstrated then that would be very strong evidence for the validity of the RFOT theory of the SGT.

The frequency dependent glass transition susceptibility defined by Eq.(2.46) can similarly be expressed as a scaling function. In general the $\epsilon_x$ given by Eq.(2.47) will give the scale distinguishing static critical behavior from dynamical critical behavior for all quantities as $T \to T_K$.

Although not as rigorously founded as the scaling law for $\tilde{\chi}_{NL}$, we can also give a scaling law for the frequency dependent shear viscosity, $\eta(\epsilon,\omega)$, if we assume that because it is related to a time integral of a time correlation function its static value is proportional to $\tau$ given by Eq. (38). We then obtain,

$$\eta(\epsilon,\omega) = \exp(b^{d/2} F_{\eta} [\epsilon b^{1/\nu}, \frac{b^{d/2}}{\ln(1/\tau_m\omega)}])$$

with $F_{\eta}$ a scaling function. The static or zero frequency shear viscosity then behaves as $\tau$ but for $\epsilon < 1/\ln(1/\tau_m\omega)$ it behaves as

$$\eta|\epsilon \ln(1/\tau_m\omega) < 1| \sim \frac{1}{\tau_m\omega}.$$
Again, the important physical and experimental point is that \( \tau \) given by Eq.(2.47) sets the crossover scale in either time or frequency (\( t \rightarrow 1/\omega \)) space. Note that \( \eta \) being simply proportional to \( \tau \) in Eq.(2.48) is needed to obtain Eq.(2.49), which in turn is required for the proper stress/strain relation in the glassy phase. Also note that Eq.(2.49) in the Naiver-Stokes equation will lead to shear waves in this frequency range, which is certainly the correct physical result for frequencies not too small near the glass transition. The crucial result from activated scaling is that \( \varepsilon < 1/\ln(1/\tau \omega) \) sets the frequency scale where shear waves should be observed in the supercooled liquid phase.

III. COMPARISON TO OTHER PHASE TRANSITION THEORIES FOR THE GLASS TRANSITION AND AN INEQUALITY

In this Section, we compare and contrast the RFOT theory with conventional first and second order phase transitions, with particular focus on the SGT.

A. First order phase transition theory for the glass transition

An earlier DFT of the SGT, advanced in the in the mid-eighties\(^{51,52} \), was based on the approximate Ramakrishnan-Yussouff\(^{53} \) theory of the liquid to crystal transition and modified it for the SGT. By generating random sites of a large system to describe the amorphous state (viewed as an aperiodic solid instead of periodic crystal characterizing an ordered solid) it was shown numerically that the RY free energy functional has a metastable solution, which presumably corresponds to a static description of the glassy state. The transition to such an aperiodic state was deemed to be first order, occurring at a density below the putative crystallization density.

Although such a description might capture aspects of the amorphous state it differs qualitatively from the RFOT in two important aspects: (1) The transition to the aperiodic state is first order with density itself exhibiting a discontinuity where as in RFOT it is the analogue of the EA parameter describing density fluctuations (Eq. 2) that has a jump at \( T_K \). A regular first order transition would also have a latent heat, and exhibit something that resembles a discontinuity in the specific heat. In contrast, RFOT theory predicts absence of latent heat at \( T_K \). It is also unclear if the identified freezing density is close to the analogue of \( T_K \). (2) The RY theory was not used to identify a dynamical transition temperature or density. More importantly, the link between \( T_d \) and \( T_K \) requiring the emergence of exponentially large number of low free energy metastable glassy states, which is the most important aspect of the RFOT theory was not demonstrated. (3) Finally, if one were to associate a divergent length scale or exponent with an ordinary first order phase transition then it would be the Berker-Fisher value \( \nu = 1/d \) discussed in Sec II.D. Such a value is inconsistent with the equality found using the RFOT theory (further discussed in Sec III.C), nor is it in accord with recent simulation results\(^{54,55} \).

B. Second order phase transition theory for the SGT

Over the years there have been numerous theories of the SGT that can be classified as standard second order or continuous phase transitions. The earliest ones were the Adam-Gibbs (AG)\(^{42} \) and Gibbs-DiMarzio (GDM) and DiMarzio of the SGT and rubber transitions in polymer systems\(^{56} \). A scenario along these lines has been postulated by Tanaka\(^{55,57} \) based on computer simulations, which has been further discussed by Langer\(^{48} \). It is worth remarking that these theories do not consider consequences of the change in the nature of transport at \( T \approx T_d \).

We first remark that it is hard to imagine how any equilibrium order parameter description of a liquid to glass transition would not generically have a discontinuous character, at least according to Landau theory. At isolated critical points, a continuous transition is possible. In fact, within RFOT this has been discussed by others, and related to a critical point for a random field problem\(^{58} \). Second, in a second order transition the concept of a metastable state is not obvious. As a consequence it is difficult to see how such transitions can have a temperature crossover scale (in RFOT, it is \( T_d \)) and also a true equilibrium transition temperature (in RFOT, it is \( T_K \)) at lower temperatures. The existence of two distinct temperatures in supercooled liquids seems to be well confirmed both numerically, as well as experimentally. The existence of the temperature scale \( T_d \) at which loss of ergodicity starts to manifest itself also tidily explains dynamic heterogeneity and aging. Technically, the problem with a second order phase transition scenario is that by definition there is only one temperature where the system becomes unstable to infinitesimal fluctuations everywhere.

C. Self-generated quenched disorder implies \( \nu \geq 2/d \)

For systems with quenched disorder, the correlation length exponent at any phase transition must satisfy the inequality \( \nu \geq 2/d \). For the structural glass problem, there is effectively quenched disorder that is self generated. Here, we argue the self generated quenched randomness in the structural glass problem also leads to the inequality \( \nu \geq 2/d \).

We begin by noting that below \( T_d \) (technically close to \( T_K \)) the glassy state can be partitioned into cluster of particles of size \( \xi \). Because the observation time is much greater than the relaxation time for particles in two clus-
ters to interchange, the motion of particles in one cluster is essentially isolated from another. Since $\xi$ is sufficiently large each cooperatively rearranging (CRR) region will have differing thermodynamic properties, which clearly is a violation of large numbers. In particular, we expect fluctuations of the ideal glass transition temperature, $\delta T_K$, in the various clusters to scale as $\sqrt{(\delta T_K)^2 \sim 1/\xi^{d/2}}$. In order for the ideal glass transition to exist, this fluctuation must be smaller than the distance to the ideal glass transition, $\epsilon \sim 1/\xi^{1/\nu}$. Consequently, $\nu$ must satisfy $\nu \geq 2/d$.

IV. DISCUSSION

The RFOT theory provides a coherent picture of the liquid state as it is cooled and supercooled. We have given physical arguments, rooted in theory, that there are two relevant temperatures, one attributable to change in transport at $T_d$ and the other in which a genuine equilibrium transition occurs at $T_K$ with both first and second order characteristics. Both these temperatures emerge from a single static theory. By expanding on our prior order characteristics. Both these temperatures emerge from a single static theory. By expanding on our prior work, we obtained a number of results. (1) We give a variety of arguments to show that a characteristic length diverges at $T_K$ with an exponent $\nu = 2/d$. In many ways this makes rigorous our earlier predictions, and this finding is an important cornerstone of RFOT. (2) We predict that there are large finite size effects at $T_K$. The theory predicts a specific rounding of the transition, as well as a specific shift in the location of $T_K$, due to finite size effects. This implies that there will be variations in the value of $T_K$ depending on the system size. This prediction is amenable to test by computer simulations using the random pinning method. (3) Using ideas based on activated scaling, we predict that the non-linear susceptibility near $T_K$ on long length scales diverges as $\sim [\ln(t/\tau_0)]^2$. We have also obtained the cross-over scale that distinguishes between dynamical critical effects and static critical effects as $T_K$ is approached. This is especially relevant for the shear viscosity.

The equilibrium transition in the RFOT theory is a novel type of transition and as such, it has certain universal properties. The implication is that most of the features of the ideal glass transition discussed here are independent of the Kauzmann-like interpretation of $T_K$. For example, the order parameter definition given by Eqs. (1) and (2) does not hinge on an entropy or state solution like crisis, rather it is motivated by a glass being characterized by a frozen and random density order parameter. Physically this seems necessary to obtain the elastic behavior of a glass. Any field theory, or Landau theory, for such a transition will necessarily have a first order nature because general symmetry allows for a cubic term in the theory. This implies in general, that if an ideal glass transition exists with such an order parameter it will be a RFOT with some sort of divergent length scale, characterized by an exponent of $\nu = 2/d$. Given this, results like the specific heat behavior discussed in Section II.E will be equally generic. The inequality $\nu \geq 2/d$ also seems generic. To discuss transport near an ideal glass transition we have largely used many state type arguments to obtain free energy barriers that scale like $\xi^{d/2}$. However, this result too seems to go beyond an entropy crisis picture: It only depends on local particle rearrangements to go from two uncorrelated one glassy configurations. The law of large numbers suggests that if the volume in question contains $N$-particles then this sort of fluctuation will scale as $N^{1/2} \sim \xi^{d/2}$.

The RFOT theory provides a coherent picture of the equilibrium transition in the RFOT theory is a specific shift in the location of $T_K$ is approached. This is especially relevant for the shear viscosity.

Historically, the Adam-Gibbs (AG) theory was the first to propose an explanation for the possible divergence of relaxation times at $T_K$ based on vanishing of configurational entropy. The predictions based on the more ad hoc AG mechanism is distinct from the RFOT theory in at least two ways. First, in the AG theory there is no analogue of $T_d$ giving the impression that the character of transport changes only at $T_K$ or at all temperatures. This would be inconsistent with many observations suggesting that at $T_d$, particle transport changes character, and theoretically with numerous exact dynamical solutions in high dimensions. Indeed, there are innumerable computer simulations that experimentally support this picture. More importantly, by analyzing data for a number of glass forming materials Novikov and Sokolov have proposed that there may be a near "universal" value of $10^{-7}$ s (larger than the estimate by Goldstein) for relaxation, which sets the boundary between diffusive transport and onset of activated transitions. Of course, the value of $T_K$ at which relaxation exceeds $10^{-7}$ s is clearly material dependent. Several computer simulations have noted that the nature of transport (non-Gaussian behavior in van Hove correlation functions and evidence for hopping mechanism) changes even before $T_d$ is reached, further supporting the
accepted view that the dynamical transition is avoided. We surmise that theories, such as the AG theory, which cannot describe the dynamical changes at $T_d$ may not be suitable for describing glass transition. Second, RFOT predicts a much stronger growth of the correlation length than the AG theory. Interestingly, recent computer simulation studies support the predictions of RFOT theory first by identifying the existence of $T_K$ (or equivalently density\textsuperscript{65}), and second by confirming that $\nu = \frac{2}{54,55}$.

Universal aspects of mode coupling theory have been remarkably successful in describing the change in the character of relaxation of undercooled liquids in the neighborhood of $T_d$. However, because MCT predicts that relaxation times only grow as power law as temperature is decreased it cannot account for activated transitions. As a result MCT cannot describe transport deep into the supercooled regime.

In our view, currently RFOT theory encompassing both $T_d$ and $T_K$, appears to be the only theory that accounts for changes in phase space structure, break down of effective ergodicity, emergence of dynamic heterogeneity as a consequence of violation of law of large numbers, and growth of $\xi$. Most importantly, both the equilibrium and dynamical aspects at $T_d$ and $T_K$ emerge naturally without having to treat the physics at these temperatures separately as done by MCT or the AG theory. The result of the RFOT theory is that analysis of experimental relaxation data must be done in two steps. For $T \leq T_d$ the relaxation time should be analyzed using a power law. In the temperature range, $T_K \leq T \leq T_d$ the temperature dependence of the relaxation data should follow VFT.

Attempts to fit the data over the entire range by activated dynamics could lead to inconsistent values of the parameters. Finally, we stress that all of this in accord with large dimensionality exact solution of liquid state systems\textsuperscript{30–32}.

Acknowledgements: This work was supported by the National Science Foundation under Grant Nos PHY 17-08128 and CHE 19-00033, and Welch Foundation (F-0019).

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