Order in glassy systems

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Abstract

A directly measurable correlation length may be defined for systems having a two-step relaxation, based on the geometric properties of a density profile that remains after averaging out the fast motion. We argue that the length diverges if and when the slow timescale diverges, whatever the microscopic mechanism at the origin of the slowing down. Measuring the length amounts to determining explicitly the complexity from the observed particle configurations. One may compute in the same way the Renyi complexities $K_q$; their relative behavior for different $q$ characterizes the mechanism underlying the transition. In particular, the ‘random first order’ scenario predicts that in the glass phase $K_q = 0$ for $q > x$, and $K_q > 0$ for $q < x$, with $x$ the Parisi parameter. The hypothesis of a nonequilibrium effective temperature may also be directly tested from configurations.

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(Some figures in this article are in colour only in the electronic version)

1. Introduction

A solid is an arrangement of particles with permanent density modulations. While commonplace, it is nevertheless remarkable that a system of soft particles may condense into a crystalline state, since due to thermal activation, no particle is strictly confined to a specific region of space. These notwithstanding, permanent density modulations persist forever in an infinite sample. The reason for this is that in order for spontaneous thermal fluctuations to completely erase the density modulations in the solid phase, rearrangements involving a divergent number of particles are required, and this in turn requires infinite free energies, and as such is an infinitely rare event.
Glasses also appear solid, in the sense that they have long-lived density modulations, although it is still not known whether these are truly permanent as for crystals. Similarly, relaxation in some supercooled liquids occurs on a timescale that appears to diverge faster than Arrhenius as the temperature is lowered. Both these features of slow relaxation, if they hold rigorously even for a system of soft particles, may only be explained if there is an ever increasing number of particles having correlated evolutions, implying that there is a growing coherence length [1] as a function of decreasing temperature. The central puzzle of glasses is that there does not appear to be any recognizable form of spatial order—in contrast, the configurations seem definitely liquid-like, so it is not clear from whence they derive their solidity.

There are several possible resolutions to this: the simplest is to conclude that glasses are not truly solids. That is, their relaxation would become purely Arrhenius at sufficiently low temperatures, as is expected for a system comprising independent parts. In this case, an amorphous solid would strictly speaking exist only in the trivial case of hard particles immobilized by contact with others, for which all activation is absent. Another possibility is that (at least some) glasses indeed have a truly super-Arrhenius behavior as $T \to 0$ or even as $T \to T_K$ for some finite temperature $T_K$, termed the ‘Kauzmann temperature’ [3]. This assumption is implicit in several theories of fragile glasses. For example, density functional theory expresses the liquid properties in terms of a free energy functional of the density and exhibits, for low temperatures and high pressures, not only periodic ‘crystalline’ configurations but also non-periodic, amorphous states [4]. A related, more complete version of this is the ‘random first order’ scenario [5, 6], which posits an ‘ideal glass’ state to which the system would equilibrate given an infinitely slow annealing from high temperature to $T < T_K$. Such an ideal state would presumably be characterized by permanent non-periodic spatial density modulations. In order for the solids postulated in these theories to exist in finite dimensions beyond the mean-field approximation, they require coherence at diverging lengths. This leads to a central question: can this cooperativity be observed directly from configurations?

In this paper we shall discuss a family of lengths [2] that, we argue, should diverge in any system having a timescale that diverges at some temperature (or equivalently shows a super-Arrhenius behavior as $T \to 0$). The lengths are geometric in nature, in the sense that they do not require information on either microscopic dynamics, the history of the sample or even whether the system is in or out of equilibrium. The various scenarios for the glass transition are distinguished by the relative manner in which the different lengths behave.

The reasoning proceeds as follows. First, we recognize that order concerns the time-averaged density, which is insensitive to fast excitations such as vibrations, particle-vacancy excitations or isolated spin flips. In fact, this is true as well for crystals, which are only truly periodic once such excitations are averaged out. In a non-crystalline system, these average density profiles (which we shall term ‘states’) are not periodic, so a multitude of possibilities for the density profiles are obtained simply by translations. This leads naturally to the question: How many possible states are there? The logarithm of this number is the complexity (we reserve the word ‘entropy’ for quantities based on counting individual configurations, and ‘complexity’ for those associated with classes of configurations). We note that care must be

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4 Here we always refer to coherence in the particle positions, rather than in their velocities. Coherence in the motion over long distances is not in itself a hallmark of a solid, as the example of a sound wave travelling through a fluid makes clear.

5 In the case of crystals, we have the alternative of spatial, rather than temporal, averaging, which is automatically done by the Fourier transform. We do not know how to do this passage from time to space averaging in non-periodic systems.
taken here, because density distributions are continuous objects, so that counting them is much like counting the number of trajectories of a dynamical system. We shall exploit this analogy.

At this point we make the following observation: if we wish to concentrate on a truly solid phase, in which a density profile lasts forever, the number of different possible density profiles cannot be exponential in the volume, or, to be more precise, the complexity cannot be extensive. This is because if there are exponentially many metastable states in competition, a simple nucleation argument suggests that they cannot all have infinite lifetime; starting from any particular state, some other state will nucleate in finite time. (This is of course only true for short range, finite interactions in finite dimensions.) As we shall argue, the reason is that if the number of possible finite-size motifs the system may nucleate is exponential in their volume, the increase in the probability of nucleating a new motif overcomes the cost, in terms of probability, of creating an interface.

We now consider a sample of very large size $V$, and 'patches' of volume $V \ll V$ belonging to it (see figures 1 and 2). The fact that the number of density profiles in a solid is subexponential immediately implies that patches of configurations of volume $V$ repeat more often than exponentially rarely in $V$, so that their number $N(V)$ obeys $\ln(N(V)) < O(V)$. In contrast, within the liquid phase, or in a defective (nonequilibrium) glass, the system breaks into uncorrelated regions, and we have, for large $V$, $\ln(N(V)) \sim V K_1^{V}$, with $K_1$ being the complexity per unit volume in the large $V$ limit ($\lim_{V \to \infty} K_1^{V}$), with the statistics taken over patches belonging to the (much larger) volume $V$.

The fact that patterns of all sizes repeat less often than exponentially, so that there is no (extensive) complexity, directly implies the existence of a divergent correlation length. We may see this most easily as follows. Suppose we have a region of volume $V$ with a configuration $A$. To what extent does $A$ determine the configuration (say, $B$) of a neighboring region, also of size $V$? The typical number of repeats of configurations of volume $V$ (or rather, its typical logarithm) is $\ln(N(A)) \sim \ln(N(B)) \sim -K_1^{V}$. Clearly, if $A$ and $B$ appeared independently, the repetition probability of the pair $A+B$ would be $\ln(N(A+B)) = \ln(N(A)) + \ln(N(B)) \sim -2V K_1^{V}$, while in general it will be some number $\ln(N(A + B)) \sim -V K_2^{V}$. The patches will then not be independent if $K_2^{V} < 2K_1^{V}$, or, in other words, if the entropy of patches of size $V$ is subextensive. In that case, the knowledge of the configuration in a volume $V$ gives information on that of an adjoining patch. Note that we have assumed here that for large $V$, the most likely patch entropy (based on the logarithm of the frequency of appearance of a patch) is, to leading order in $V$, independent of the shape. If we have an extensive patch entropy $K_1$, and we assume that for large volumes the system may be considered as being composed of a number of independent components of volume $V_1 \sim \ell_1^{D}$, then $N \sim e^{V K_1} \sim e^{V/V_1}$, and we identify a correlation volume as $V_1 \sim 1/K_1$.

A system with subextensive patch entropy $K_1$ will then have an infinite length—in this sense, it is ordered. Now let us start from such an ordered system and fracture it into pieces of typical size $V_c$. Let us now reassemble these pieces at random to form a new system of large size. In this case, we expect that patches smaller than $V_c$ will be found often, and those larger than $V_c$ will be exponentially rare, as in figure 1. This leads us then to the definition of the following types of length for an amorphous system:

- **Complexity length**: $\ell_1 = K_1^{\frac{1}{D}}$, measures the frequency with which a typical patch repeats.
- **Renyi lengths**. In order to distinguish different glass transition scenarios, it is useful to consider patches that repeat more or less often than is typical. This leads to the definition of lengths based on the Renyi complexities, or, equivalently, the large deviation function for patch repetition frequencies.
Figure 1. Two patterns, one smaller and one larger than the crystallite length. The small pattern will recur with high frequency, but the large one will be rare, because it requires recurrence of the crystallite boundaries within a patch, which is a random event. Patches that are very large with respect to the crystallite volume occur with multiplicity exponential in the number of defects per volume, of the order of the number of microcrystallites contained.

Figure 2. Looking for a patch that is the same, up to a certain precision, and allowing for rotations.

- **Crossover length**: patches with volumes smaller than $V \ll V_c$ repeat often, while larger ones $V_c \ll V$ are exponentially rare. The crossover length is $\ell \sim V_c^{\frac{1}{D}}$. A related measure is the following: suppose the complexity patch of volume $V$ is given, with finite $V$ corrections, by: $V K_1 - A(V) K_1^{(1)} + \ldots$, where $A(V)$ is subleading in $V$, for example $A(V) \sim V^\nu$. The next to leading term $A(V) K_1^{(1)}$ describes correlations, and is hence referred to as the 'synergy' [7]. $K_1^{(1)}$ might diverge at the critical point (see footnote 9). If $A(V)$ is not $O(1)$ in $V$, $K_1^{(1)}$ provides a new length.

These measures may be understood easily in the case of a microcrystalline system, as sketched in figure 1. However, one of the central points of our proposal is that it allows us to define a correlation length for systems whose ground states are not only non-periodic but even conventionally thought of as amorphous, in the sense that their scattering patterns have
no Bragg peaks. In what follows, we discuss in detail how the counting and identification of patterns may be made, as well as the implications to current glass scenarios.

2. Scrambled sequences and imperfect tilings

2.1. Perfect deterministic sequences

It is important to note that sub-extensive complexity of patches does not imply that the infinite system is either periodic or quasiperiodic, as characterized by a diffraction spectrum consisting of \( \delta \) function Bragg peaks. To make this clear, we review a particularly well understood class of systems, namely those which possess ‘recursive symmetry’. For ease, we shall consider one-dimensional sequences of two symbols (1 and 0) which may be generated by means of a substitution rule, wherein a symbol is replaced by a specific word (composed of 0’s and 1’s) everywhere it occurs, and then iterating this process. Generalization to higher dimensions, for which building blocks are subdivided, is immediate. We consider three prototypes which characterize three types of spatial structure:

Rule 1: 
\[ 1 \rightarrow 10 \quad 0 \rightarrow 10. \]  
(1)

Starting from (say) 0, we generate sequences of length \( 2^n \) by iterating this rule \( n \) times; the result is clearly periodic:

\[ 0101010101010101010101010101. \]

The second possibility is illustrated by:

Rule 2: 
\[ 1 \rightarrow 10 \quad 0 \rightarrow 1. \]  
(2)

If we start from 0 as before, we generate strings of length \( F_n \), where \( F_n \) is the \( n \)th Fibonacci number:

\[ 1011010111011101101110111011101110111011101110111011101110111011. \]

This rule produces the Fibonacci sequence, which is quasiperiodic, with a discrete Fourier transform consisting of a dense set of \( \delta \)-functions. Finally, we consider:

Rule 3:
\[
\begin{align*}
11 & \rightarrow 110 \\
10 & \rightarrow 1101 \\
01 & \rightarrow 0010 \\
00 & \rightarrow 0001
\end{align*}
\]

Iterating this rule \( n \) times produces a string of length \( 2^n \):

\[ 11101111000100011111011110111011110001011101111011110111101111011110111101111011110111101111011110111101. \]  
(3)

This sequence, known as the Rudin–Shapiro sequence, has a discrete Fourier transform with a flat modulus \([8–11]\), and

Rule 3’:
\[ 1 \rightarrow 10 \quad 0 \rightarrow 01 \]  
(4)

which produces

\[ 011010011001011001100101100110010110011001100101. \]

This sequence, known as the Thue–Morse sequence, also has no \( \delta \)-functions in its Fourier transform \([9, 10]\). The Rudin–Shapiro and the Thue–Morse sequences are examples of ‘non-Pisot sequences’\([8]\).

Thus, the three examples have different kinds of orders.
Figure 3. Probability of overlap of two Fibonacci sequences $P(q)$.

(1) **Periodic.** The Fourier spectrum peaked at the period and its harmonics. Degeneracy $= 2$ (entropy $= \ln 2$) (and more generally the logarithm of the cell size).

(2) **Quasiperiodic.** The Fourier spectrum is a dense set of $\delta$-functions. Two sequences taken at random have an overlap $Q$ distributed according to $P(Q)$ consisting of a $\delta$-function and a uniform tail, as in figure 3: the Parisi function $P(Q)$ is non-trivial. The number of subsequences of length $\ell$ (that is, with $\ell$ symbols) appearing in a large sequence is proportional to $\ell$ (actually $\ell + 1$) as is easy to see from the rules above. All patches of size $\ell$ repeat within a distance of $O(\ell)$. The probability that any two subsequences taken at random have a Hamming distance per unit length $< \epsilon$ is proportional to $\epsilon$, and independent of their size $\ell$.

(3) **Non-Pisot sequence.** For the example of the Rudin–Shapiro sequence, the modulus of the discrete Fourier transform is flat, independent of $\omega$. The Parisi function $P(Q)$ is peaked: two (asymptotically large) subsequences taken at random from the infinite sequence have an overlap that is, with high probability, the smallest value possible (one-half, in this case). Here again, the number of subsequences of length $\ell$ is also $\propto \ell$, and all patches of size $\ell$ repeat within a distance of $O(\ell)$.

In going from (i) to (iii), some of the conventional (scattering) manifestations of order are lost, despite the fact that the entropies of sequences of types 2 and 3 are still logarithmic, and patches of size $\ell$ repeat much more often than $2^\ell$, as they typically would for a random system. To understand this frequency of repetition, note that a subsequence of size $\ell$ is completely included within the ‘descendants’ of a single site after $k \propto \log(\ell)$ substitution steps; the subsequence repeats in the descendant of a neighboring ancestor—its ‘cousin $k$-times removed’. This argument is applicable to all systems obtained by substitutions [13, 14].

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6 Are there further intermediate forms of order between these and random sequences? The answer is yes, although we may not have characterized all of them. A strategy to construct some [11] is to use the successive 1’s and 0’s of an auxiliary sequence to decide which one out of two sets of substitution rules we use each time to construct the main sequence. The two sets of rules for the main sequence, as well as the one determining the auxiliary one, may be one of the types above. This way one obtains sequences that are deterministic, and have entropies larger than $\ln N$, but still non-extensive.
Figure 4. (a) A set of Wang tiles with matching rules indicated by numbered edges. The rule for laying the tiles is that only edges of the same number may abut. (b) States obtained from a Monte Carlo simulation of a Wang tiling quenched from high to low temperature are left with only ‘hard’ defects (i.e. mismatches of adjacent sides), as indicated by the red lines.

We conclude that both the crossover length $\ell_c$ and complexity length $\ell_1$ diverge in all these deterministic sequences—even though there may be no Bragg peaks in the diffraction spectrum, so that by the conventional measure of scattering such systems would be considered amorphous.

These considerations may be generalized to higher dimensions as well. Perhaps the best-known 2D quasiperiodic tiling is the Penrose tiling, but a system with similar properties is a Wang tiling [15]. In constructing a Wang tiling one is required to follow matching rules. One set of tiles with their associated matching rules is shown in figure 4(a): every interface between tiles has to have the same number on both tiles, a two-dimensional domino.

An example of a system that is statistically isotropic is a tiling of identical triangles [16] called the ‘Pinwheel’ tiling (figure 6). Pinwheel tiling can be constructed by recursive substitution rules, so patterns of size $\ell$ repeat (modulo rotation) every $D \sim \ell$ in a perfect tiling, as for the 1D sequences discussed above.

Significantly, it is known that any packing which can be generated by a recursive substitution rule admits a set of matching rules which only allows tilings consistent with the recursive symmetry [17]; the set of matching rules for the Pinwheel tiling is known explicitly [16].

Two-dimensional tilings of the non-Pisot type have also been constructed [18]. This means that an experimental system having this kind of order would go unnoticed, if the only way to detect quasicrystals order is a diffraction pattern! The strategy we outline in the following sections may be used to detect this form of order.

2.2. Imperfect sequences: finite lengths

The examples of the previous subsection are all systems with infinite correlation lengths. What does an imperfect quasiperiodic system, or an imperfect non-Pisot system look like? The question begins to be tricky, as there is no notion of periodicity in the latter case. In analogy to a system which has not achieved the ideal glass state, let us consider a sequence constructed by concatenating unrelated subsequences of varying lengths $n$, drawn from a
Figure 5. The average distance $D(\ell)$ between words of length $\ell$ in a perfect Thue–Morse sequence of length 1000 (blue) and a sequence of length 1000 composed by concatenating strings of average length 50 randomly drawn from a perfect Thue–Morse sequence (red).

Figure 6. The Pinwheel tiling, with infinite correlation length.

distribution $P(n)$, with average $\bar{n}$. For example, the scrambled Thue–Morse sequence shown below:

$$0110101101001001100011001$$

is the analog of a polycrystallite, but with micro/non-Pisot pieces. It is easy to see that strings of length $\ell \ll \bar{n}$ will repeat often, because even in independent subsequences the same string will appear. However, if $\ell \gg \bar{n}$, the likelihood of overlap is exponentially small, by the central limit theorem. This is seen in figure 5, where the average distance between strings of length $\ell$ is plotted as a function of $\ell$ for a perfect and a randomly concatenated Thue–Morse sequence. The two lengths $\ell_c$ and $\ell_1$ are then estimates of the coherence length $\bar{n}$.

Consider next the example of the Wang tiling, which has the virtue that it can be expressed as a lattice model of spins [19] (taking 16 values in the example of [19]), assigning an energy
corresponding to a mismatched edge—that is to say, the matching rules may be enforced by a suitable Hamiltonian. Consequently, such a system can be simulated at finite temperature (see figure 4(b)) to measure the growth of the coherence length. At finite temperature, the Wang tile system has energetic point defects—local violations of the matching rules. Starting from a high temperature state and annealing down to low temperatures, many easy-to-cure ‘benign’ defects disappear, leaving the system in a state with isolated ‘hard’ defects (figure 4(b)) that can only be removed by large-scale rearrangements of the order of the interdefect distance. Such behavior is also present in the Penrose tilings, where there are benign matching rule violations due to local tile flips, and non-trivial configurations of the phason field [20] which require large-scale rearrangements to eradicate. In this case, as we have verified numerically for the Wang tile system, patches repeat often when much smaller than the distance between serious defects, and rarely when they are larger.

3. Particle systems: defining and counting density profiles

3.1. Density profiles

Particle systems are more subtle to compare than number sequences, since they have continuous positions and thermal vibrations. We now present a method that allows us to distinguish and count configurations in principle. Recalling our motivation from glasses, we proceed in two steps: first we average out rapid fluctuations to obtain a continuous density profile (a state), and then we count the number of profiles.

Consider first a snapshot of a crystal at finite temperature. It is clear that, strictly speaking, no two patches of the crystal coincide exactly, because of thermal fluctuations in the atomic positions. Therefore, we consider a density profile averaged over a long (in principle infinite) time:

\[
\rho(r) = \lim_{\tau \to \infty} \frac{1}{\tau} \int_0^{\tau} \rho(r, t) \, dt
\]

(7)

with

\[
\rho(r, t) = \sum_a \delta(r - r_a(t)).
\]

In the long-time limit (in this paper, this always means taken after the thermodynamic limit), the density tends to a smooth function with a well-defined crystal modulation, as sketched in figure 7. The fact that there are permanent density modulations is reflected in the fact that autocorrelation function

\[
C(t, t') = \frac{1}{V} \int \rho(r, t) \rho(r, t') \, dr \sim q_{EA}
\]

(9)

has a plateau \( q_{EA} \) as \( t - t' \to \infty \):

\[
\frac{1}{V} \int (\rho(r) - \bar{\rho})^2 = \frac{1}{\tau_p} \int \rho(r) \, dr \sim q_{EA} > 0
\]

(10)

where

\[
\bar{\rho} = \frac{1}{V} \int \rho(r)
\]

and \( \sim \) denotes ‘in the large \( \tau_p \) limit’. This defines the Edwards–Anderson parameter \( q_{EA} \).

An ideal amorphous solid should have permanent, non-periodic density modulations. By this we mean that, if we define a density profile as in equation (7), we obtain a function that
may be neither constant nor periodic nor quasiperiodic. Such solids might or might not strictly speaking exist. What we know in practice is that a supercooled liquid has a relaxation time \( \tau_a \), much larger than the microscopic time \( \tau_m \), during which an amorphous density profile remains stable, so that the autocorrelation function (9) has a plateau as in figure 8. Only if \( \tau_a \to \infty \) do we have a true amorphous solid.

For finite \( \tau_a \), we may define a density profile for a supercooled liquid configuration as in equation (7), but with the average taken over a finite time \( \tau_\rho \) such that \( \tau_m \ll \tau_\rho \ll \tau_a \). This brings about an essential limitation on the precision with which both \( \rho(r) \) and \( r \) are defined. The density resolution is of the order of, but inevitably not better than,

\[
\epsilon(\tau_\rho) \sim \left[ \frac{1}{V\tau_\rho^2} \int dr \int dt (\rho^2(r, t) - \rho^2(r)) \right]^\frac{1}{2} = \left[ \frac{1}{\tau_\rho^2} \int_0^{\tau_\rho} dt' \int dr' C(t', t') - q_{EA} \right]^\frac{1}{2}.
\]  

(11)
The precise manner in which the resolution $\epsilon(\tau_\rho)$ goes to zero depends on the way that $[C(t - t') - qEA]$ goes to zero on approaching the plateau: if it does so in an integrable form, then $\epsilon(\tau_\rho) \sim (\tau_\rho)^{-\frac{1}{2}}$.

The resolution of $r$ is in turn limited by the fact that the density is generated by time-dependent $\delta$-functions, corresponding to particle positions. Suppose we divide the total volume $V$ in cubes of side $\Delta r$. A particle traveling with velocity $v$ will visit $v\tau_\rho/\Delta r$ cubes in time $\tau_\rho$. Estimating $v \sim \sqrt{k_B T}$, and considering that there are $N$ particles, a total of order $N\sqrt{k_B T} \tau_\rho / \Delta r$ is visited. If we want this to be larger than the total number $V / [\Delta r]^D$ of cubes, we get the condition

$$\Delta r(\tau_\rho) \sim \left[\sqrt{k_B T \tau_\rho \rho}\right]^{-\frac{1}{D}}. \quad (12)$$

For large $\tau_\rho$, $\Delta r(\tau_\rho)$ is exceedingly small, and we shall assume throughout that we have discretized space or smoothed the average density functions up to a distance of this order. In what follows we shall, in addition, sample $\rho$ at even larger spatial intervals, but this we will do after having discretized space at the scale (12).

In realistic situations, the above time-averaging procedure may be impractical. Two alternatives to this procedure are:

- Identifying positions modulo some fixed tolerance that we know a priori to be appropriate, for example, a fraction of the inter-particle spacing;
- Instead of comparing the averaged configurations, we may omit the time-averaging and compare the inherent structures of configurations, that is, the positions reached after a quench to $T = 0$. Using inherent structures, however, is not free of artifacts and ambiguities: two inherent structures that differ in buckling modes that flip-flop on a timescale $<\tau_\alpha$ should be treated as being the same, while a naive counting will treat them as different. If the density of such localized flip-flops is finite, one gets an entropy that may be absent in a correctly time-averaged procedure.

### 3.2. Correlation lengths

The preceding procedure yields a density profile that has averaged out—to the extent possible—all rapid degrees of freedom, such as particle-vacancy pairs, phonons, particle rattlings. Our problem now is how to count the resulting continuous profiles or, alternatively, how to identify congruent patches. This situation is already familiar in a different context: it arises when we wish to determine the entropy of trajectories of a dynamical system, in which setting it has been well discussed both in principle and in practice. In the case of dynamical systems, we first discretize phase space in cubes of size $\epsilon$ and time in intervals $\tau$. We consider pieces of trajectories $d$ time-intervals long, and define the Kolmogorov entropy on the basis of the probability

$$P_\epsilon(i_1, \ldots, i_d)$$

that at time $\tau$ the trajectory lies in the cube $i_1$, at time $2\tau$ it lies in the cube $i_2$, . . . and at time $d\tau$ it lies in cube $i_d$, see figure 9, as

$$K_1 = -\lim_{\tau \to 0} \lim_{\epsilon \to 0} \lim_{d \to \infty} \frac{1}{\tau d} \sum_{i_1, \ldots, i_d} P_\epsilon(i_1, \ldots, i_d) \ln P_\epsilon(i_1, \ldots, i_d). \quad (13)$$

More generally, we may define all the Renyi complexities:

$$K_q = -\lim_{\tau \to 0} \lim_{\epsilon \to 0} \lim_{d \to \infty} \frac{1}{\tau d(q - 1)} \ln \left( \sum_{i_1, \ldots, i_d} P_\epsilon(i_1, \ldots, i_d)^q \right). \quad (14)$$

Note that $d$ goes to infinity before $\epsilon$ and $\tau$ go to zero, a fact that shall turn out to be important.

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7 Here we follow the presentation and notation of [21].
In practice, one may proceed as proposed by Grassberger and Procaccia [22]: we consider a very long sequence of $M$ times, and for every patch of length $d$ labeled by its starting point $i$ (figure 10), we count the number of patches $n_i^{(d)}(\epsilon)$ that coincide with it—up to precision $\epsilon$. The entropies are estimated by

$$K_q \sim -\lim_{\tau \to 0} \lim_{\epsilon \to 0} \lim_{d \to \infty} \frac{1}{\tau d (q-1)} \ln \left\{ \frac{1}{M} \sum_i \left[ n_i^{(d)} \right]^{(q-1)} \right\}$$

and in particular

$$K_1 \sim -\lim_{\tau \to 0} \lim_{\epsilon \to 0} \lim_{d \to \infty} \frac{1}{\tau d} \left\{ \frac{1}{M} \sum_i \ln \left[ n_i^{(d)} \right] \right\}.$$  

This way of putting the problem is practical, because we may extrapolate the results from finite $\epsilon$ (for which we have many coincidences) to smaller $\epsilon$. This is illustrated in figure 12 (a sketch).

The fact that an entropy may be defined and computed may be understood if we assume that (see footnote 7) the dependence on $\epsilon$ (for small $\epsilon$) is subdominant in $d$:

$$\frac{1}{M} \sum_i \left[ n_i^{(d)} \right]^q \sim e^{q \tau d K_1 + \phi(q)} \ln \epsilon$$

for some $\phi(q)$. As we shall discuss later, this means that $d$ goes to infinity keeping $|\ln \epsilon|/d$ small.

We may easily adapt this procedure to count density profiles. We consider average profiles $\rho(r)$ in $D$-dimensional space (figure 11). We discretize space in some manner, with a typical
Figure 11. The analog of a trajectory is the density profile. The density plays the role of the position in a dynamical system, and space the role of time.

grid spacing $\Delta$ playing the role of the time interval $\tau$ in a dynamic system$^8$, and we also discretize $\rho$ in intervals of size $\epsilon$. Here again, we may proceed as follows. We consider a large sample containing $M$ different patches $a_1, \ldots, a_M$ each of volume $V$. We look for the number of coincidences between a patch and all others, centered about any site and allowing for rotations. Two patches coincide if the sum of all the differences in the value of $\rho$ between corresponding sites is smaller than $\epsilon$. Denoting by $n_a^{(V)}(\epsilon)$ the number of patches that coincide in this way with the patch $a$, we have

$$K_q \sim - \lim_{\Delta \to 0} \lim_{\epsilon \to 0} \lim_{V \to \infty} \frac{1}{(q-1)V} \ln \left\{ \frac{1}{M} \sum_a \left[ n_a^{(V)}(\epsilon) \right]^{q-1} \right\} \quad (18)$$

and in particular

$$K_1 \sim - \lim_{\Delta \to 0} \lim_{\epsilon \to 0} \lim_{V \to \infty} \frac{1}{V} \left\{ \frac{1}{M} \sum_i \ln \left[ n_i^{(V)}(\epsilon) \right] \right\} \quad (19)$$

Here and in what follows we shall for (conceptual, though not practical) simplicity keep the limits $\Delta \to 0$ and $\epsilon \to 0$, although they are probably unnecessary provided that one takes $V \to \infty$.

The Renyi complexities are at most extensive ($K_q = O(1)$ as $V \to \infty$), or subextensive, and then $K_q \to 0$. Writing equation (18) in the form

$$e^{-qV K_{q+1}} \sim \frac{1}{M} \sum_a \left[ n_a^{(V)}(\epsilon) \right]^q = \frac{1}{M} \sum_a e^{qV \left( \frac{\ln n_a}{V} \right)} \quad (20)$$

(where the symbol ‘$\sim$’ means ‘in the limit $\Delta \to 0$ $\epsilon \to 0$ $V \to \infty$’) suggests that a large deviation function be defined for the variable $\beta Y = -\frac{\ln n_a}{V}$ (the sign and the factor $\beta$ entering the definition of $Y$ are purely conventional). Introducing the large-deviation function $G$ through

$$P \left( \beta Y = -\frac{\ln n_a}{V} \right) \sim e^{V G(Y)} = \frac{1}{M} \sum_a \delta \left( V \beta Y + \ln n_a \right)$$

$$= \int dq \frac{1}{M} \left[ \sum_a e^{qV \left( \frac{\ln n_a}{V} \right) + \beta V} \right]$$

$$= \int dq \ e^{qV \left( \beta Y - K_{q+1} \right)} \quad (21)$$

$^8$ Note that to obtain a continuous $\rho$ we have already smoothened by a much smaller scale equation (12).
Figure 12. A sketch of the plots of the logarithm of patch frequency versus precision $\epsilon$ for different lengths $d$ (see [22]). The Grassberger–Procaccia procedure takes advantage of the scaling with $\epsilon$ to extrapolate results from relatively large $\epsilon$ to small $\epsilon$, a limit in which no coincidences would in practice be found.

Thus, the Renyi complexities may be thought of as (a form of) a Legendre transform of the large-deviation function $G(Y)$. We may evaluate the integral in (21) by the saddle point

$$G(Y) = q^* [\beta Y - K_{q^*}]$$

with $\beta Y = \frac{d}{dq} [g K_{q^*}]_{q^*}$. (22)

Before proceeding, let us be more precise about the dependence on $\epsilon$. If we assume, as in (17), that (20) may be written in the limit of small but non-zero $(\Delta, \epsilon)$ and large but not infinite $V$ as

$$\frac{1}{M} \sum_a \left[ n_a^{(V)}(\epsilon) \right]^q \sim e^{-qVK_{q^*} + 1} + \epsilon \phi(q)$$

(23)

for small $\epsilon$. Note that there might be a correction intermediate in order $V$ between the leading one and the one dependent on $\epsilon$. If we work under the condition

$$\frac{\phi[\ln \epsilon]}{VK_{q^*}} < 1 \rightarrow \epsilon \gg e^{-VK_{q^*}/\phi},$$

(24)

we may refine equation (22) to read

$$\ln P(Y) = V \Delta \ G(Y) + \ln \epsilon \ G_o(Y) \quad \text{with} \quad G_o = \phi(q^*(Y))$$

(25)

with $q^*(Y)$ given in (22). From this equation we read the approach as $\epsilon e^{K_{q^*} + 1} \gg 0$ of the large-deviation function $G$ to its large-$V$ limit. Let us stress that we do not know exactly what the dependence of entropy on $\epsilon$ might be like in a true glassy system, and this one of the interesting questions that an experimental/numerical investigation should answer.

3.3. Direct measurement of the complexity

The (patch, or block) complexity $\Sigma^V$ is defined as the log of the number of different kinds of states for a given patch size $V$, per unit volume. (Here, and in what follows, $\Sigma$ without
supraindex stands for the large $V$ limit). Because each state $\alpha$ is repeated $n_\alpha$ times, we have to divide by $n_\alpha$ to count each type once

$$
\Sigma(Y) = \lim_{\Delta \to 0} \lim_{V \to \infty} \frac{1}{V} \ln \left\{ \frac{P[n(\epsilon)]}{n(\epsilon)} \right\} = \beta Y + G(Y) \quad \text{and} \quad \beta Y = -\frac{\ln n}{V}. \quad (26)
$$

From equations (26) and (21) we obtain the useful expression

$$
e^{-(q-1)VK_\epsilon} = \int \text{d}Y \, e^{\Sigma(Y)-\beta qY}. \quad (27)
$$

In practice, one would work at different values of $\Delta$ and $V$ and make histograms of the probability that patterns repeat with frequency $n(\epsilon)$ at given precision $\epsilon$. Plotting in terms of the normalized logarithm of this frequency $\beta Y = \ln \left( \frac{P[n(\epsilon)]}{n(\epsilon)} \right)$ one should obtain a set of curves that scale correctly as $\Sigma(\epsilon, V) = \Sigma(Y) + \ln \epsilon \, G_\epsilon(Y)$.

The content of the functions $G(Y)$, $\Sigma(Y)$ and $K_\epsilon$’s is the same. They encode important information to distinguish different scenarios for the amorphous state.

4. Long-range order

4.1. Amorphous order versus weak periodicity

The property of weak periodicity, introduced by Aubry, is defined as follows [24]: for patches of size $V$, there is a distance $R(V, \epsilon)$ along the sample such that in an infinite sample every patch will recur—up to precision $\epsilon$—at a distance closer than $R(V, \epsilon)$. Note that nothing is said about the dependence of $R(V, \epsilon)$ with $V$. What this definition excludes is the possibility that for any given type of patch $a$, there might be, in an infinite sample, arbitrarily large $a$-free regions, something that would clearly occur in a random sample. Aubry [24] showed that, under reasonable assumptions, the ground-state configuration of a particle system with short-range interactions is weakly periodic.

In this paper, we are defining a length $\ell_1$ for amorphous order also based on patch repetition which is, although conceptually related, not equivalent. Central to our argument is the necessity of time-averaging, which allows us to compare average-density profiles. We are measuring complexities—based on the multiplicities of these averaged profiles—rather than entropies, which are related to the multiplicities of configurations. Time averaging allows us to work at non-zero temperature, a situation in which weak periodicity of configurations necessarily breaks down.

The definition we follow in this paper of perfect order, implying an infinite correlation length, is a situation in which almost every patch recurs often, the recurrence distance being less than exponential in the patch’s size. This is related to the vanishing of the complexity, and it does not exclude the possibility that there will be arbitrarily large regions where some patch is absent.

Is it possible that we could use the notion of weak periodicity applied to time-averaged density profiles, rather than to configurations? The answer is that, even in that case, the two conditions (vanishing complexity and weak periodicity) are distinct. To understand this, consider first the case of a quasicrystal at low but non-zero temperature, for example the Wang tile system discussed above. In equilibrium, defects will appear and disappear continuously, just as in a crystal. These ‘benign’ defects will be averaged out in our procedure, so we may safely ignore them. However, there are also ‘serious’ defects having low energies that cannot be eliminated with a finite number of rearrangements (the best known being the ‘decapods’ of the Penrose tilings). These are long-lived defects, and will therefore show up in a time-averaged density function. Moreover, they have, in equilibrium, finite probability if their energy is finite, as is the case in the Wang-tile and Penrose system above.
Denote by \( f_\alpha \) the free energy density of the metastable state \( \alpha \) (measured with respect to the one \( f_0 \) of a perfectly ordered system) defined as having a fixed set of ‘serious’ defects, and including all rapid thermal fluctuations. \( f_\alpha \) is computed from the partial Gibbs–Boltzmann distribution, keeping ‘serious’ defects fixed. Denote \( N = e^{\chi_1(f)} \) as the number of different such metastable states of free energy density \( f \) for a sample of total volume \( V \). We may write the partition function at low temperature as

\[
Z = e^{-\beta V f_0} \int df e^{V(\chi_1(f) - \beta f)}.
\]

The integral may be evaluated for large \( V \). If \( \frac{\partial \chi_1}{\partial f} \bigg|_{f=0} > \frac{1}{T} \) a saddle point with \( f > 0 \) dominates, and there is a finite density of ‘serious’ defects in equilibrium. (As we shall see below, a finite density of defects brings the system back to the liquid phase, in this case because defects are so close that they may be mutually annihilated with finite rearrangements, and have no longer infinite life.) If, on the other hand \( \frac{\partial \chi_1}{\partial f} \bigg|_{f=0} < \frac{1}{T} \), the system is frozen in \( f \sim 0 \); the order is not destroyed because the density of defects tends to zero, although the total number present may still be large. In the latter case we have an ordered system with infinite complexity-related correlation length, but no weak periodicity (even for a time-averaged configuration) since the ‘serious’ defects spoil it.

Although we have chosen to discuss this situation in terms of the Wang tile system, it is also relevant for the (more hypothetical) ‘ideal glass’ scenario at finite temperature, as we shall see.

4.2. Infinite lifetime and diverging lengths

We shall now argue that if a system of particles with short-range smooth interactions has a state with permanent density modulations, then these density profiles necessarily have subextensive complexity, and hence there is, by our definition, a long-range amorphous order. This is also true for the zero-temperature limit of a system with super-Arrhenius timescale behavior. Our reasoning is not rigorous, but we believe it may be made so. The argument is based on an elementary observation: it is not possible to have exponentially many metastable states, all of which have a divergent lifetime.

A situation that arises in supercooled liquids is that a system has many options of phases for nucleating. The question then is: does this multiplicity increase the probability of nucleation

\[ 9 \text{ This is what happens at the critical temperature of the Random Energy Model, a toy model of the glass transition (see [12]).} \]
into a different phase? In other words, is there an ‘entropic drive’ for nucleation away from a state, leading the system to another state which, though individually not necessarily more favorable, belongs to an exponential family of states with free energies comparable to the original one? The argument against an entropic drive goes as follows: While a system in a given phase is nucleating another phase \( A_0 \), it needs to follow a certain number of steps. Why should this process be accelerated by the fact that there were many options of phases \( A_1, A_2, \ldots \) given that these themselves require different sets of steps to nucleate? To clarify the point, let us consider a simple situation—a system at very low temperature, activating its escape out of the spherical crater \( V(r) \) of dimension \( d \), in figure 13. Starting at the bottom, the particle follows (say) a Langevin process, and the probability evolves via a Fokker–Planck equation:

\[
\dot{P} = \nabla [T \nabla + \nabla V] P. \tag{29}
\]

The probability distribution was spherically symmetric at the start, and it will remain so, so we may go to spherical coordinates:

\[
\dot{\tilde{P}} = \frac{1}{r^{d-1}} \left[ T \frac{d}{dr} \left( r^{d-1} \frac{dP}{dr} \right) + P \frac{d}{dr} \left( r^{d-1} \frac{dV}{dr} \right) + \frac{dV}{dr} \frac{dP}{dr} \right]. \tag{30}
\]

Putting \( \tilde{P} \equiv r^{d-1} P \) we get the radial diffusion equation

\[
\dot{\tilde{P}} = \left[ T \frac{d^2}{dr^2} + \frac{d}{dr} \left( V(r) - T (d - 1) \ln r \right) \right] \tilde{P}. \tag{31}
\]

Thus, the system evolves in an effective potential \( V(r) - T (d - 1) \ln r \). The second term in this effective potential derives from the multiplicity of excited states: \((d - 1) \ln r\) is the entropy of excited states, being the logarithm of the volume of a shell of radius \( r \). We thus conclude that the multiplicity of excited states have the effect of lowering the barrier against nucleation. In particular, with very small \( T \), but with \( T d = O(1) \) such that \( V(r) - T (d - 1) \ln r \) is monotonically decreasing, the particle will escape in finite time.

An intuitive way of understanding this result is as follows. At low temperatures, for every trajectory that escapes, there are many unsuccessful trials which fall just short of escape, and return to the center. Although still extremely rare, trials reaching, say, 90% of the barrier height are exponentially more common than those which actually escape. Their frequency is, however, proportional to the surface of the 90% energy level. The situation for one out of an exponential number of metastable states is very similar. The nucleation of a droplet of any other state of volume \( V \) will cost a free energy at most proportional to the surface. However, there are \( e^{V(\Sigma,f)} \) options of different droplets of free energy density \( f \), and they will be accessed with total frequency \( \int df \ e^{V(\Sigma,f)} \). These need not be grown simultaneously, but the many unsuccessful attempts in which a droplet grows and collapses will be this number of times as frequent as if there were only one other option. The existence of many paths (in the particle language) or metastable states (in the system language) does not make it easier to nucleate into any particular state, but it makes it easier to nucleate out of the initial deep well. In other words, if we lump the ensemble of other states as a single entity (the liquid state, in fact), the probability of nucleating one of this ensemble (that is, of escaping a specific state) is just as if there were an extra term in the free energy of the liquid equal to \(-T \ln \left[ \int df \ e^{V(\Sigma,f)-\beta f} \right]\). Therefore, the usual nucleation argument states that this volume term will sooner or later overcome the surface term, and the system will leave the original state: this competition leads to escape only if there is an extensive complexity.

Going back to the example of escaping a crater in large dimensions (such that \( T d \) is of order 1), we may describe this at low temperatures either by an ensemble of extremal trajectories with different angles (the low temperature treatment of (29) or by a single escape trajectory for the radius (i.e. the low temperature limit as applied to equation (31)).
ensemble of escape routes at different angles in the original space and the single escape trajectory in radial coordinates play respectively the role played in an extended system by the nucleation of many phases and the nucleation of the ‘liquid phase’ that encompasses all of them.

Technically, activation out of a state may be calculated by considering the extremal trajectory, starting in the state and moving away from it by nucleating another phase. The logarithm of the escape time is given by the action of the trajectory. In ordinary nucleation, the trajectory goes through a free energy maximum at the critical droplet radius, and the total action of the escape trajectory is of the order of the free energy barrier at the critical droplet. The situation here is different, because we have an exponential number of different trajectories moving out of the state, none of which needs to have a finite critical droplet radius. When we sum over all trajectories, it is the multiplicity of trajectories that decreases the action, in a manner entirely analogous to the way that multiplicity of states decreases the total free energy (cf equation (28)). That is to say, we have a trajectory complexity that decreases the values of the dynamic action, and hence the logarithm of the escape time. If the action of an individual nucleation keeps growing with the surface of the droplet size, but the number of trajectories is proportional to the exponential of the droplet volume, then escaping will sooner or later win—even without a critical radius for an individual passage.

We thus conclude that the state will be destabilized by the presence of exponentially many others, even if the passage to any individual one is extremely improbable. Note that this argument also implies that it is impossible to have an exponential number of states with super-Arrhenius lifetimes all the way down to zero temperature, because the Arrhenius escape probability will sooner or later dominate.

The argument we outlined above is close in spirit to the mathematical results in [1] and to the construction in [35–37] related to the point-to-set correlation function. What has been proven there, at least for lattice systems with discrete variables, is as follows: if one considers a closed volume, and asks if a fixed configuration on the surface is correlated with the one in the center (a point-to-set correlation), the maximal radius $r_{\text{max}}$ at which this happens diverges if there is a divergent timescale. Translated into density profiles, this might seem to suggest that for $r_{\text{max}} \to \infty$ the surface configuration determines the time-averaged configuration inside, and this would imply that the complexity is necessarily subextensive, since the number of different configurations of the boundary is proportional to the exponential of an area. There is a problem with this argument, as the simple example of classical mechanics shows. In that case, giving the configuration at two times determines fully a trajectory in between, and yet the number of possible trajectories may be exponential (the Kolmogorov–Sinai entropy need not be zero). The reason for this is that, because of the Lyapunov instability, to know the configuration in the whole interval one needs to know those of the boundaries to greater and greater precision—the number of digits necessary being proportional to the interval of time considered times the Lyapunov exponent. In conclusion, the entropy of the bulk is not necessarily proportional to the boundary surface, even in the case that the bulk configuration is fully determined by the boundary one, but may be—in the case of exponential sensitivity to boundary conditions—also proportional to the enclosed volume.

5. The Kauzmann/random first order scenario

Assume that there is a temperature at which there is an equilibrium situation with permanent density modulations. We now ask: are there other excited states with infinite lifetimes that we may define in a time-averaged way? For example, a ferromagnet with periodic boundary conditions may be composed of two domains of positive and negative magnetization,
separated by an essentially flat (at sufficiently high dimensions) interface. Another example is a crystal with a single straight dislocation. A third example was already mentioned above: in quasicrystals one may have isolated defects, the most famous being the ‘decapod’ defect in a Penrose tiling. In addition, in any non-periodic system, new states may be obtained from translations, if this does not violate the boundary conditions. We call these states because we are considering the ensemble of configurations visited in infinite times (the thermodynamic limit has been taken first) by a system that started with any one configuration with a given permanent defect. In other words, we are studying the problem modulo short-lived (such as particle-vacancy) excitations.

Next we ask about the free energy of these states. In many cases, such as the macroscopic dislocations or domain walls mentioned above, their free energy is infinitely (though non-extensively in the system’s volume $V$) higher than the ground state. Their contribution to the equilibrium distribution hence vanishes. In other cases, it could conceivably occur, and the Wang tile example cited above is an example, that an excited state of this kind has a free-energy difference of order 1 with respect to the ground state. In that case, the conclusion is that it will have a finite probability in equilibrium, and we are forced to conclude that the equilibrium distribution contains several ‘ergodic components’, mutually inaccessible (for a macroscopic system) in finite times. Note that the state with a single ‘serious’ defect will typically have many short-lived activated ‘benign’ defects.

We now ask about the number of these states, in terms of their free energy, at a given temperature. If we count how many of these states are there, regardless of their probability of appearing in an equilibrium sample, the logarithm of this number is the complexity $\Sigma(T)$ (only weakly dependent on $T$). The probability of each state is proportional to $e^{-\beta V f + \alpha}$ for a sample of volume $V$. The definition of $\Sigma$ is extremely tricky: as soon as $\Sigma(f)$ is of order 1, the simple ‘entropic drive’ argument above states that the states cannot be stable. This is most clearly seen in the example of the Wang tile system: as soon as we have a finite density of defects, and hence an extensive $\Sigma$, they are separated by finite distances and may be annihilated with local rearrangements. Strictly speaking then, $\Sigma$ is only well defined when it is subextensive, and it otherwise counts states whose definition is timescale dependent.

As mentioned above (cf equation (28)), the whole question is whether $\frac{\partial \Sigma}{\partial f} \bigg|_{f=0} < \frac{1}{T}$ or $\frac{\partial \Sigma}{\partial f} \bigg|_{f=0} > \frac{1}{T}$. Only in the former case will a distribution frozen in the lower states be dominant. In the latter case entropy wins, states with higher free energy density dominate and since these are exponential in number, they are necessarily unstable: we are in the liquid phase. Suppose then that $\frac{\partial \Sigma}{\partial f} \bigg|_{f=0} < \frac{1}{T}$ holds, and we have a solid with permanent density modulations. As the temperature is increased, the system eventually becomes a liquid. How is the transition made? There are several possibilities:

- **Second order.** The density modulations diminish with increasing temperature, and eventually vanish completely at a temperature $T_*$. This means that the level $q_{E_A}$ of the plateau in the autocorrelation decreases continuously and vanishes at a given temperature $T_*$. This is what would happen in a ferromagnet, with the plateau value being the magnetization squared.

- **First order.** A set of exponentially many (i.e. necessarily unstable) states suddenly begins to dominate the Gibbs distribution above a temperature $T_m$. Such a transition is like melting. Note that if $\Sigma$ suddenly becomes large, the states will necessarily be individually short lived, and the system will jump to a liquid without high viscosity.

- The $\Sigma$ versus $f$ curve reaches zero with a finite derivative $\beta_K$ (figure 14). Because, as mentioned above, $\Sigma$ is in general weakly dependent upon temperature, we must state that this has to happen at temperature $T_K$. At $T > T_K$ the saddle point $\frac{\partial \Sigma}{\partial f} \bigg|_{f=0} = \frac{1}{T}$ begins to
5.1. A crucial hypothesis: from global to patch quantities

A standard result in statistical mechanics is that the entropy of a system is calculable, to leading order in the volume, as the limit of the patch (or ‘block’) entropy. This is because, even though the boundary conditions of each block are not those of the total system, one assumes that for short-range correlated systems the effect is of the order of the boundary surface, and hence subdominant in the large volume limit. If we assume that the same holds for the complexity and, furthermore, for all the Renyi complexities of finite $q$, then we may translate the predictions of theoretical scenarios for the entire probability distributions to observable statements concerning the distribution for patches. In other words, we are led to the assumption that the large-deviation function $G = \frac{1}{V} \ln P$, expressed in terms of the normalized logarithmic frequencies $\ln n/V$, is the same, to leading order in $V$, as the one obtained for the whole system with volume $V$, and given boundary conditions. Note that this does not mean that individual profiles dominating patches of a given volume $V$ are the same as the state that would dominate a single patch with, say, periodic boundary conditions.

The random first order scenario is strictly speaking, at least for the moment, a mean-field notion, and any extrapolation to finite dimensions is made at the price of phenomenological thinking (see, however, [23] for a recent incursion into finite dimension). The argument above allows us to make the reasonable assumption that the predictions of RFOT for the number of states of the entire system may be applied to patches embedded in an infinite system as well. The RFOT theory predicts, at least for mean-field models, a series of metastable states with internal free-energy $f_\alpha$. For a given $f$, there is a quantity $N(f)$ of these states, with $\ln N \sim V^{\gamma(f)}$, which defines the complexity $\Sigma^{\gamma(f)}$—a function that depends only weakly on temperature, because states retain their integrity when temperature changes. The assumption that all Renyi complexities of the whole system may be computed as a limit of the Renyi
complexities for large patches means that we may identify $\Sigma^{\text{eq}}$ and $\Sigma$. Thus

$$
\rho^{(\text{equil})} = \frac{e^{-\beta \bar{f}}}{\sum_{\alpha} e^{-\beta f_{\alpha}}}
$$

$$
K_q \sim \frac{1}{V(q - 1)} \left\{ \ln \left[ \sum_{\alpha} e^{-V q f_{\alpha}} \right] - q \ln \left[ \sum_{\alpha} e^{-V f_{\alpha}} \right] \right\}
$$

$$
= \frac{1}{V(q - 1)} \left\{ \ln \int df e^{V(\Sigma(f) - \beta f)} - q \ln \int df e^{V(\Sigma(f) - \beta f)} \right\}
$$

$$
= \frac{1}{V(q - 1)} \ln \int df e^{V(\Sigma(f) - \beta \bar{f})}
$$

(32)

where

$$
\beta \bar{f} \equiv -\frac{1}{V} \ln \int df e^{V(\Sigma(f) - \beta f)}. \quad (33)
$$

Comparison with equation (27) yields, and we recognize $q$ as the $m$ parameter of replica theory [34]. Varying the Renyi parameter $q$ is then exactly the procedure introduced by Monasson [25] to analyze these models (see figure 15). For example, this means that if the system were a one-step replica symmetry-breaking solution in the low temperature phase, with the Parisi parameter $x$, then the Renyi complexities would be (see figure 14)

$$
K_q = 0 \quad \text{for} \quad q < x
$$

$$
K_q > 0 \quad \text{for} \quad q > x. \quad (34)
$$

This is directly measurable if an equilibrium configuration is available (figure 14).

Experimentally, we thus have to proceed as follows. We first define an averaged profile, over a time $\tau_\rho$ of the order of $\tau_x$. Note that $\tau_x$ is not the same for all patches with the rarer ones expected to have shorter lifetimes. Hence, we must be careful of the fact that the number of rare states (i.e. the Renyi complexities for large $q$) may be dependent on the averaging time $\tau_\rho$.

Next, we either compute the Renyi complexities in the manner of Grassberger–Procaccia by scaling the results for different $V$ and different $\epsilon$, or, alternatively, we set up a large deviation function plotting the logarithm of the probability of having a number of repetitions versus the logarithm of the number of repetitions, both normalized by $V$.
A fingerprint of the RFOT scenario is then a non-zero crossover Renyi parameter $q = x$, implying that there are always rare patches that repeat exponentially infrequently. On approaching the transition from below $x \to 1$, and this means that non-repeating patches become more and more frequent, and eventually typical at $T = T_K$. Conversely, in the liquid phase just above $T_K$, most patches repeat exponentially rarely. However, there are a few that repeat frequently, but their number is so small that even their high repetition rate does not allow them to dominate the measure.

Let us mention here that we may take the point of view of patches, rather than the whole system, for more complicated (and informative) large-deviation functions, depending on the patch overlap $Q$, defined, for example as

$$Q_{ab} = \frac{1}{V} \int d\mathbf{r} \left( \rho_a(\mathbf{r}) - \bar{\rho} \right) \left( \rho_b(\mathbf{r}) - \bar{\rho} \right).$$

(35)

One takes a patch at random, and counts the average complexity $K_1(Q)$ as in equation (19), but restricting the sum to patches at distance $Q$. Finally, one averages over initial patches. $K_1(Q)$ is the patch version of the Franz–Parisi effective potential [26], a function closely related to the Parisi ansatz for which there are also suggestions within the RFOT scenario.

6. Out of equilibrium: effective temperatures

In practice, glasses are out of equilibrium, and the concept of temperature does not apply in principle to them. An old idea [27] has been to assume that the structure somehow ‘remembers’ the situation of the liquid at the temperature it had before it fell out of equilibrium. The system would have one (or, in later versions, several) ‘fictive temperatures’ governing the slow evolution. On the other hand, when analytic solutions for the glassy dynamics of mean-field models became available [28], it turned out that these systems possessed an effective temperature $T_{\text{eff}}$ relating fluctuation and dissipation (of any observable) at low frequencies [29]. Indeed, $T_{\text{eff}}$ would be measured by applying a low-frequency thermometer to the system out of equilibrium, an experiment that has been performed [30]. The interpretation for the appearance of this temperature is not yet fully clear, but it is not related to structures that are frozen in the form they had when the system crossed the transition, but rather to structures that form again and again. We know this because effective temperatures exist at times which are long compared to the autocorrelation time, so the system had more than enough time to restructure.

A somewhat strong assumption is to suppose that this temperature arises because a system out of equilibrium explores metastable states with a probability at $T_{\text{eff}}$—a proposal that agrees with a hypothesis that had been made by Edwards for granular matter [31–33]—and that $T_{\text{eff}}$ slowly evolves in a much longer (perhaps infinite) timescale toward the equilibrium value $T$. In this very strong form, the assumption is that if the system has metastable states with free energy $f_\alpha$, so that the equilibrium probability would be proportional to $e^{-f_\alpha/T}$, out of equilibrium the probabilities would be those associated with a probability $\propto e^{-f_\alpha/T_{\text{eff}}}$. In other words, if the probability of a certain configuration in equilibrium is $p^{(\text{equil})}_\alpha$, the probability out of equilibrium should be

$$p^{(\text{out of equil})}_\alpha \propto \left[ p^{(\text{equil})}_\alpha \right]^{T_{\text{eff}}/T}. $$

(36)

Again, if we assume that what has been said for $V$ can also be said for large $V$, then we should have

$$V(q - 1)K_q = \ln \left( \sum_\alpha e^{-q\beta_\alpha f_\alpha} \right) - q \ln \left( \sum_\alpha e^{-\beta_\alpha f_\alpha} \right)$$

(37)
which means that, in every physical situation, the dependence on $q$ may be written as

$$
\frac{(q - 1)}{q} K_q = \beta_{\text{eff}} \left[ U(q \beta_{\text{eff}}) - U(\beta_{\text{eff}}) \right]
$$

(38)

where $U(b) \equiv -\frac{1}{b} \ln \sum_\alpha e^{-bf_\alpha}$ is a function that depends, upon this assumption, on the final parameters (temperature, pressure). All the history of the sample would manifest itself, under this hypothesis, only through the single parameter $T_{\text{eff}} = 1/\beta_{\text{eff}}$. In the limit $q \to 1$ (38) reduces to $K_1 = \Sigma |\beta_{\text{eff}}|$, the complexity evaluated in the point where $\Sigma' = \beta_{\text{eff}}$.

In order to check the hypothesis of an effective temperature of this kind, we should repeat different protocols, and for each determine the value of $T_{\text{eff}}$ that will put the results in the form (38), with a single protocol-dependent $T_{\text{eff}}$ for all $q$’s. This should be possible in principle, and if this is so, one could check that the patterns that dominate the measure at a given $T_{\text{eff}}$ are the same, irrespective of the protocol by which it has been reached.

7. Conclusion

In this paper we have discussed how to render observable several quantities from glass theory, in particular those that have their origin in mean-field-like theories. We have discussed a correlation length that, we argue, should diverge for any solid, defined as a system in which, in spite of thermal fluctuations, there are permanent spatial density modulations. The construction has the strength of not being tied to any specific phenomenological scenario. Once we define states, order and lengths in a concrete and experimentally measurable manner, we are led to the conclusion that the mere existence of a timescale that goes to infinity faster than an Arrhenius law implies, for finite, short-range interactions, that the lengthscale diverges.

Defining further quantities associated with patches embedded within the system, we may answer the question of how, on the basis of the data from an ideal glass configuration, we could recognize if it corresponds to a random first order/Kauzmann scenario. Perhaps more relevant for realistic systems, we may also use the technique to decide, using the experimental out-of-equilibrium data, whether there is a flat exploration of metastable states, leading to an effective temperature.

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