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Brownian dynamics : from glassy to trivial

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We endow a system of interacting particles with two distinct, local, Markovian and reversible microscopic dynamics. We find that while the first, standard one, leads to glassy behavior, the other one leads to a simple exponential relaxation towards equilibrium. This finding questions the intrinsic link that exists between the underlying, thermodynamical, energy landscape, and the dynamical rules with which this landscape is explored by the system.

The microscopic phenomena driving the dynamical arrest in supercooled liquids and in glasses are still controversial. One line of thought, which originates in the work of Adam and Gibbs, pictures a glass-forming liquid as a system whose energy landscape complexity accounts for the slowing down of its dynamics. The original Adam-Gibbs [1] theory relates viscosity –the momentum transport coefficient– to configurational entropy. The more recent Kirkpatrick, Thirumalai, and Wolynes [2] scenario is based on the study of the metastable configurations of the system and on the concept of nucleating entropic droplets. This is the Random-First-Order Theory (RFOT). The idea is that metastability arises from the many valleys of the energy landscape the system can be trapped in. RFOT gives a large set of quantitative predictions including non-mean field particle models [3]. These aspects of the physics of glasses were reviewed by Debenedetti and Stillinger [4] and more recently by Biroli and Bouchaud [5].

In another line of thought no complex energy landscape needs to be invoked, and dynamically induced metastability alone is held responsible for the dynamics slowing down. This is a phenomenological approach in which at a coarse-grained scale local patches of activity are the only ingredients of the dynamics. This has led to the development of kinetically-constrained models (see [6] for a review), which have the advantage of lending themselves with greater ease than molecular models to both numerical experiments and analytical treatment. The hallmark of the corresponding lattice models is the absence of any structure in the energy landscape (the equilibrium distribution is that of independent degrees of freedom). These somewhat simplistic descriptions are not directly built from the microscopics, and it is often argued that their glassy-like properties, like the existence of dynamical heterogeneities, are almost tautological, but recent works [7] have tried to bridge the gap from the microscopics to dynamic facilitation.

A central concept in both approaches is that of metastability. A metastable state can be viewed as an eigenstate of the evolution operator with a nonzero but small relaxation rate. The existence of such states can be induced by the dynamical evolution rules alone, as in KCM's, but physical intuition dictates that, in realistic systems, these originate from the deepest structures of the energy landscape. The method for catching and counting metastable structures from the statics alone was coined by Mézard and Parisi, thus providing the first first-principle calculation of some of the properties of

glasses. Their idea is to help polarize two attractively coupled copies the system into one of these deep valleys and to investigate whether thermal fluctuations are enough to wash out any overlap between them. By contrast, catching dynamically induced metastable structures is done by examining the overlap of the system properties in the course of a long time interval. Implementing this program was done by exploiting Ruelle's thermodynamic formalism first on KCM and then in realistic systems [8].

Our purpose in this work is to show that a minute modification of the evolution rules of a glass-forming liquid can lead to dramatic differences in the dynamics. We will propose two possible dynamics for our system of interacting particles, which are both time-reversible and Markovian. However, one is known to induce dynamical arrest and glassy-like phenomenology, while we show that the other one is characterized by straight, one-step, exponential relaxation, thus bypassing any slowing down. We emphasize that time reversibility ensures that the equilibrium distribution is the same in both systems. They share the exact same energy landscape. In both cases, the complex energy landscape is sampled by means of local moves. A central difference with existing works is that ours focuses on interacting degrees of freedom, while KCM's, the system of oscillators studied earlier by Kawasaki and Kim [9], or the single oscillator of Whitelam and Garrahan [10], are noninteracting degrees of freedom.

Our system is made of identical particles interacting via a two-body potential $v(\mathbf{x} - \mathbf{y})$ and we choose to describe its properties in terms of collective density modes $\rho(\mathbf{x}, t)$ with average density ρ_0 . In terms of these, the equilibrium distribution is the standard Gibbs weight $P_{\text{eq}}[\rho] \propto e^{-\beta\mathcal{F}[\rho]}$, where the density dependent effective free energy has the well-known expression

$$\beta\mathcal{F}[\rho] = \int_{\mathbf{x}} \left[\rho(\mathbf{x}, t) \ln \frac{\rho(\mathbf{x}, t)}{\rho_0} - \rho(\mathbf{x}, t) \right] + \frac{1}{2} \int_{\mathbf{x}, \mathbf{y}} \rho(\mathbf{x}, t) \beta v(\mathbf{x} - \mathbf{y}) \rho(\mathbf{y}, t), \quad (1)$$

in which the first term in the rhs is of entropic origin, and the second one is the potential interaction energy. From a dynamical point of view, the particle density must locally be conserved, which constrains it to evolve according to a continuity equation

$$\partial_t \rho + \nabla \cdot \mathbf{j} = 0 \quad (2)$$

Our two choices of dynamics lie in the specific density dependence of the particle current \mathbf{j} . We begin with a standard choice in which the particle current is given by

$$\mathbf{j}_L = -\rho \nabla \frac{\delta \mathcal{F}[\rho]}{\delta \rho} + \sqrt{2T\rho} \boldsymbol{\xi} \quad (3)$$

where $\boldsymbol{\xi}$ is a vector whose components are independent white noises with unit correlations $\langle \xi^\alpha(\mathbf{x}, t) \xi^\beta(\mathbf{x}', t') \rangle = \delta^{\alpha\beta} \delta^{(d)}(\mathbf{x} - \mathbf{x}') \delta(t - t')$, where d is the space dimension. In an expanded form, the deterministic part of \mathbf{j}_L features a Fick diffusion term $-T \nabla \rho$ and a driving term $\rho \mathbf{F}$, where $\mathbf{F}(\mathbf{x}, t) = -\int_{\mathbf{y}} \nabla v(\mathbf{x} - \mathbf{y}) \rho(\mathbf{y}, t)$ is the local fluctuating force field acting on particles located around \mathbf{x} . We index the particle current with the letter L because this very expression of \mathbf{j}_L was shown by Dean [11] to exactly encode the dynamics of a fluid of interacting particles evolving through an overdamped Langevin equation. The parameter $\beta = T^{-1}$ is the inverse temperature of the thermostat. Once the deterministic part of \mathbf{j}_L is set, time-reversibility (or detailed balance) forces the specific $\sqrt{2T\rho}$ density dependence of the noise strength. Our second choice of dynamics for the collective modes is defined by another expression for the particle current, namely

$$\mathbf{j}_B = -\rho_0 \nabla \frac{\delta \mathcal{F}[\rho]}{\delta \rho} + \sqrt{2T\rho_0} \boldsymbol{\xi} \quad (4)$$

where the index B is reminiscent of model B dynamics of the Hohenberg and Halperin classification [12], as already defined for discussion purposes in [11]. Note that in (4) the coupling to the thermal bath is now independent of the density (noise is additive instead of being multiplicative as in (3)). Dynamics B can be obtained from dynamics L by thinking the fluctuations of the coupling to the thermostat have been turned off. We emphasize that the detailed balance property is satisfied also in this alternative dynamics, which ensures that in principle both \mathbf{j}_L and \mathbf{j}_B drive the system towards the same Gibbs equilibrium state $P_{\text{eq}}[\rho]$.

Our goal here is not to come up with a new approximation for the Langevin dynamics. A huge body of numerical and analytical literature is devoted to extracting signatures of glassiness from these dynamical rules [13]. We take them for granted. Instead, our purpose is to conduct an analytical study of our model B dynamics in which it will appear that no sign of dynamical arrest can be found. We search for an evolution equation for the density-density correlation function (otherwise called the intermediate scattering function). If a glassy behavior is to be found, then we must obtain a non vanishing long-time limit of this correlation function, indicating ergodicity breaking, or, if not, we should at least expect a multi-step slow relaxation. As we shall now see, model B dynamics leads to a simple exponential relaxation for the density-density correlations $G_{\rho\rho}$, as we conclude in (20). In order to demonstrate this property, we resort to a set of techniques coined by Kim and Kawasaki (KK) [14], originally designed to cope with Langevin dynamics, which we adapt to our model

B evolution. The central idea of their work is a refined way to cast the dynamical rules in such a way that the detailed balance property is a manifest and linearly-realized symmetry of the evolution equation. While it was not clear whether and how time-reversibility and ergodicity breaking are related, KK showed that a certain type of (reversibility preserving) approximation on the dynamics, when applied to the two point density correlations, was fully equivalent to the well known Mode-Coupling approximation [15] (alternative methods were recently found [16]). Their work follows a number of earlier attempts [17] to deal with collective coordinates within a field-theoretic framework.

The field theory that describes the evolution of the density is written in terms of the fluctuations of the density field $\delta\rho$ around its mean value ρ_0 and a response field $\bar{\rho}$:

$$Z = \int \mathcal{D}\rho \mathcal{D}\bar{\rho} e^{-S[\rho, \bar{\rho}]}, \quad (5)$$

$$S[\rho, \bar{\rho}] = \int_{\mathbf{x}, t} i\bar{\rho} \partial_t \delta\rho - i\bar{\rho} \rho_0 \Delta \frac{\delta \mathcal{F}[\rho]}{\delta \rho} - T\rho_0 (\nabla \bar{\rho})^2 \quad (6)$$

In this theory, time-reversal (TR) is encoded in the following symmetry [18]:

$$\begin{cases} \delta\rho(t) & \rightarrow \delta\rho(-t) \\ \bar{\rho}(t) & \rightarrow -\bar{\rho}(-t) + i\beta \frac{\delta \mathcal{F}[\rho]}{\delta \rho(-t)} \end{cases} \quad (7)$$

We parallel the KK approach, and define $\theta = \beta \frac{\delta \mathcal{F}}{\delta \rho} - K \star \delta\rho$, with $K \star \delta\rho(\mathbf{x}, t) = \frac{\delta\rho(\mathbf{x}, t)}{\rho_0} + \int_{\mathbf{y}} \beta v(\mathbf{x} - \mathbf{y}) \delta\rho(\mathbf{y}, t)$ and $\bar{\theta}$ is an auxiliary Lagrange multiplier field that enforces this expression for θ .

The introduction of the θ field was originally intended to deal with the non-linearity of the time-reversal symmetry in the Langevin dynamics described by the current given in (3). In our case, the time-reversal symmetry is a linearly realized symmetry, and use of an additional field is thus not necessary in that matter. However, introducing the θ field was shown by Kawasaki and Kim to be useful to properly take into account, non-perturbatively, the renormalized static properties of the system, and lead to simplifications that would be otherwise hard to detect in a standard Martin-Siggia-Rose field theory.

Grouping all four fields in a vector $\Phi = (\delta\rho, \theta, \bar{\rho}, \bar{\theta})$, and introducing $\Omega(\mathbf{k}) = \rho_0 T k^2 K(\mathbf{k})$, the action now reads:

$$S[\Phi] = S_g[\Phi] + S_{ng}[\Phi], \quad (8)$$

$$S_g[\Phi] = \frac{1}{2} \int_{\mathbf{k}, \omega} \Phi(-\mathbf{k}, -\omega) G_0^{-1}(\mathbf{k}, \omega) \Phi(\mathbf{k}, \omega), \quad (9)$$

$$S_{ng}[\Phi] = \int_{\mathbf{x}, t} i\Phi_4(\mathbf{x}, t) f(\Phi_1(\mathbf{x}, t)), \quad (10)$$

where we have split the quadratic part S_g from the higher order terms S_{ng} , and f is a function of $\delta\rho$ given by $f(x) =$

$\ln(1 + x/\rho_0) - x/\rho_0$. The bare propagator G_0^{-1} is given by :

$$G_0^{-1}(\mathbf{k}, \omega) = \begin{pmatrix} 0 & 0 & -\omega - i\Omega(\mathbf{k}) & 0 \\ 0 & 0 & -i\rho_0 T k^2 & -i \\ \omega - i\Omega(\mathbf{k}) & -i\rho_0 T k^2 & 2\rho_0 T k^2 & 0 \\ 0 & -i & 0 & 0 \end{pmatrix} \quad (11)$$

The TR invariance (7) now reads

$$\begin{cases} \delta\rho(t) & \rightarrow \delta\rho(-t) \\ \theta(t) & \rightarrow \theta(-t) \\ \bar{\rho}(t) & \rightarrow -\bar{\rho}(t) + iK \star \delta\rho(-t) + i\theta(-t) \\ \bar{\theta}(t) & \rightarrow \bar{\theta}(-t) - i\partial_t \rho(-t) \end{cases} \quad (12)$$

As was the case in KK, both S_g and S_{ng} are separately invariant under (12). We define the vertex functions $\Sigma[G]$ as the functional inverse of the correlator matrix $G = \langle \Phi \Phi \rangle$ as :

$$G^{-1} = G_0^{-1} - \Sigma[G], \quad (13)$$

We note in passing that the standard strategy to derive an approximation for G is based on performing an approximation on $\Sigma[G]$ and inserting it back into (13), thus yielding a self-consistent equation for G alone. Here, however, no approximation is involved. TR symmetry, along with causality and time and space translational invariance, impose severe constraints on the form of the correlation functions of the theory, as well as on the vertex functions. The only difference between KK's field theory and ours lies in the non-Gaussian term S_{ng} of the action in (10). Since our bare propagator and form of the time-reversal coincide with those in KK's work [14], it is easy to verify that all non-perturbative results that follow from the symmetries of the action coincide between the two theories. Differences will be found when determining the vertex functionals $\Sigma[G]$.

Writing down the inverse relation that exist between Σ and G , and combining them with the constraints arising from the time-reversal symmetry and causality, we can write exact non-perturbative evolution equations for $G_{\rho\rho}$, $G_{\theta\rho}$ and $G_{\theta\theta}$, the 11, 12 and 22 elements of the matrix G . Inspection of the equal-time singularities of the Schwinger-Dyson equations (13) also shows that :

$$\Sigma_{\bar{\theta}\bar{\theta}}(\mathbf{k}, 0) = \frac{1}{G_{\rho\rho}(\mathbf{k}, 0)} - K(\mathbf{k}), \quad (14)$$

which give a renormalized value of the bare diffusion coefficient, in a way that is reminiscent of the renormalization-group approach to model B dynamics of magnetic systems [12] : the initial time value of $G_{\rho\rho}$ is by definition $\rho_0 S(\mathbf{k})$, where $S(\mathbf{k})$ is the static structure factor of the liquid, thus we have, using the expression of K given after (7) :

$$\Sigma_{\bar{\theta}\bar{\theta}}(\mathbf{k}, 0) = \frac{1}{\rho_0} \frac{1}{S(\mathbf{k})} - \frac{1}{\rho_0} - \beta v(\mathbf{k}) = -c(\mathbf{k}) - \beta v(\mathbf{k}), \quad (15)$$

where c is the full two-body direct correlation function of the liquid, related to S by $S(\mathbf{k}) = (1 - \rho_0 c(\mathbf{k}))^{-1}$.

By Laplace transforming the four relevant members of the Schwinger-Dyson equations (13), one can deduce that the correlation functions $G_{\rho\theta}$ and $G_{\theta\theta}$ are simply related to $G_{\rho\rho}$:

$$G_{\rho\theta}(\mathbf{k}, \tau) = \left(\frac{1}{G_{\rho\rho}(\mathbf{k}, 0)} - K(\mathbf{k}) \right) G_{\rho\rho}(\mathbf{k}, \tau), \quad (16)$$

$$G_{\theta\theta}(\mathbf{k}, \tau) = \left(\frac{1}{G_{\rho\rho}(\mathbf{k}, 0)} - K(\mathbf{k}) \right)^2 G_{\rho\rho}(\mathbf{k}, \tau), \quad (17)$$

Substitution of all these results in the Schwinger-Dyson equation (13) lead to a closed equation on the density-density correlation function :

$$\partial_\tau G_{\rho\rho}(\mathbf{k}, \tau) + \frac{T k^2}{S(\mathbf{k})} G_{\rho\rho}(\mathbf{k}, \tau) = - \int_0^\tau dt \left[\Sigma_{\bar{\rho}\bar{\rho}}(\mathbf{k}, \tau - t) \frac{G_{\rho\rho}(\mathbf{k}, t)}{\rho_0 S(\mathbf{k})} + \Sigma_{\bar{\rho}\bar{\theta}}(\mathbf{k}, \tau - t) \partial_t G_{\rho\rho}(\mathbf{k}, t) \right] \quad (18)$$

This final non-perturbative result is identical to that obtained by Kawasaki and Kim, which therefore also applies to our modified dynamics. Starting from this closed equation, we now exploit the fact that our theory has only one non-quadratic term, that involves the field $\bar{\theta}$ and any (greater or equal to two) number of $\delta\rho$ fields. Thus, when calculating perturbatively the functions Σ , we are bound to find, *to all orders of perturbation* :

$$\Sigma_{\bar{\rho}\bar{\rho}} = \Sigma_{\bar{\rho}\bar{\theta}} = 0 \quad (19)$$

which shows that we have

$$\partial_\tau G_{\rho\rho}(\mathbf{k}, \tau) = - \frac{T k^2}{S(\mathbf{k})} G_{\rho\rho}(\mathbf{k}, \tau). \quad (20)$$

The physical meaning of (20) is clear : relaxation towards equilibrium occurs in a single exponential step. This is the central analytical result of this letter. The relaxation rate $\frac{T k^2}{S(\mathbf{k})}$ is proportional to k^2 (due to the local conservation of particles) and only involves the additional knowledge of the static structure factor. Away from any thermodynamic phase transition, the structure factor displays no specific singularity and thus the relaxation rate has no reason to vanish.

In retrospect, the two dynamical rules considered so far are very similar to each-other. Only local moves are involved, they both preserve particle conservation and for both time-reversibility is encoded in the dynamics. Closer inspection, however, shows potentially relevant differences. We denote by $r[\rho]$ the interaction-dependent component of the rate at which the system leaves a given configuration of the density landscape (we leave out the component expressing independent local diffusive hops). For Langevin dynamics, this rate reads

$$r_L[\rho] = \frac{\beta^2}{4} \int_{\mathbf{x}, \mathbf{y}, \mathbf{z}} \nabla v(\mathbf{x} - \mathbf{y}) \nabla v(\mathbf{x} - \mathbf{z}) \rho(\mathbf{x}, t) \rho(\mathbf{y}, t) \rho(\mathbf{z}, t) - \frac{\beta}{2} \int_{\mathbf{x}, \mathbf{y}} \rho(\mathbf{x}, t) \nabla^2 v(\mathbf{x} - \mathbf{y}) \rho(\mathbf{y}, t) \quad (21)$$

The average rate, as can be seen in the first term in the right hand side of (21), involves two and, more importantly three-body correlations. The knowledge of these correlations is necessary for the system to decide to which location of phase space it is going to evolve. Turning now to the model B dynamics, we find that

$$r_B[\rho] = \frac{\beta^2 \rho_0}{4} \int_{\mathbf{x}, \mathbf{y}, \mathbf{z}} \nabla v(\mathbf{x} - \mathbf{y}) \rho(\mathbf{y}, t) \nabla v(\mathbf{x} - \mathbf{z}) \rho(\mathbf{z}, t) + \beta \rho_0 \int_{\mathbf{x}, \mathbf{y}} \frac{\nabla \rho}{\rho}(\mathbf{x}, t) \nabla v(\mathbf{x} - \mathbf{y}) \rho(\mathbf{y}, t) \quad (22)$$

so that on average the system requires no more than two-point correlations to proceed with its evolution. The Langevin dynamics requires a more refined knowledge of the local organization than model B dynamics. We speculate that we may relate this mathematical observation to the physical picture of local caging. It is a well-known fact that the pair correlation function of a glass is no different from that of a liquid, but recent works [19, 20] suggest that triplet correlations (and higher order correlations) may behave differently.

Our result stands out as one in which the dynamics of a system of interacting particles can exactly be solved at the level of two-point functions. Yet is based on a coarse-grained formulation in which the collective density modes are treated as smoothly varying function of space. For softly repulsive potentials, the system will fall into a jammed state as the density is increased. Our approach is thus bound to reach its limitations at high densities. However, in the usual glassy regime where ergodicity is maintained, a standard two-step relaxation will never be observed with our choice of dynamics.

Furthermore, we have insisted on characterizing the dynamics via its density modes, but relating the dynamics of the modes to that of the particles is a non-trivial task. We can see the two dynamics, described by the currents \mathbf{j}_L and \mathbf{j}_B , as the coarse-grained continuum versions of two dynamics for interacting particles on a lattice. Denoting by i the lattice sites and n_i the occupation number on this site, a particle on site i may hop on an adjacent site j with rate

$$\begin{cases} n_i e^{-\beta \Delta E_p / 2} & \text{for Langevin dynamics} \\ n_0 \sqrt{\frac{n_i}{n_j + 1}} e^{-\beta \Delta E_p / 2} & \text{for model } B \text{ dynamics} \end{cases} \quad (23)$$

where $E_p = \frac{1}{2} \sum_{i \neq j} n_i v(i - j) n_j$ and ΔE_p is the potential energy variation due to the particle hopping. For Langevin dynamics, the reader is referred to [21] for a more complete discussion (with slightly different rates). Starting from these lattice dynamics, standard manipulations [22] allow one to write down a path integral representation for the two dynamics. In the coarse-grained limit, they coincide with the path integral representations of the stochastic equations Eq.(3) and (4), obtained via standard procedures [23]. Going back and forth between particles and modes is simple in Langevin dynamics, but what would an effective particle system described by model B dynamics look like? Would this effective particle

dynamics still be local? Answering that question would allow for the design of algorithms able to bypass metastable traps.

From a broader perspective, our calculation points to the necessity of establishing criteria permitting to relate static energy-landscape-based considerations, to kinetic properties. A naive way to ask the question would be: What are the generic conditions on the effective diffusion constant coupling the energy gradient to the thermal bath that allow for a reliable correspondence between minima of the energy landscape, and metastable states? This question echoes a recent work [24] in which it is shown that one could map a subclass of KCM models onto a subclass of spin-glasses, uncovering a non-trivial behavior of appropriately defined static quantities in these KCMs, which could mirror non-trivial dynamical transitions in the corresponding spin-glass systems. Once again, the traditional separation between static and dynamic framework does thus not seem to hold anymore, calling for a more refined treatment of the interplay between the two.

A quantitative characterization of the dynamic complexity, as expressed for example by the Lyapunov spectrum of our two dynamics could uncover the fundamental difference that exist between the two. More generally, what is the signature of an ergodicity breaking transition on the Lyapunov spectrum? These are ongoing investigations.

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