Quasi-equilibrium in glassy dynamics: a liquid theory approach

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Abstract
We introduce a quasi-equilibrium formalism in the theory of liquids in order to obtain a set of coarse grained dynamical equations for the description of long time glassy relaxation. Our scheme allows to use typical approximations devised for equilibrium to study glassy dynamics. After introducing dynamical Ornstein–Zernike relations, we focus on the hypernetted chain (HNC) approximation and a recent closure scheme developed by Szamel. In both cases we get dynamical equations that have the structure of the mode-coupling theory (MCT) equations in the long time regime. The HNC approach, that was so far used to get equilibrium quantities is thus generalized to a fully consistent scheme where long-time dynamic quantities can also be computed. In the context of this approximation we get an asymptotic description of both equilibrium glassy dynamics at high temperature and of aging dynamics at low temperature. The Szamel approximation on the other hand is shown to lead to the canonical MCT equations obtained by Götze for equilibrium dynamics. We clarify the way phase space is sampled according to MCT during dynamical relaxation.

Keywords: glasses, dynamics, quasi-equilibrium

The relation between dynamical slowing down and equilibrium free-energy landscape is a longly debated issue in the theory of glassy relaxation of low temperature liquids [1, 2]. Free energy landscape [3–5] and pure dynamical approaches [2, 6] provide alternative pictures of...
the glass transition [7]. At the theoretical level, the standard techniques and approximations involved in the study of equilibrium and dynamics of realistic liquid models are considerably different. Here we overcome this difficulty relating the dynamical properties of interest with the ones of an appropriate auxiliary system with Boltzmann–Gibbs distribution.

In dynamics, mode coupling theory (MCT), based on projection operator techniques, provides an approximation for an exact evolution equation of the density-density correlator and requires the static structure function as an input [8]. In equilibrium, the replica method is used in conjunction with approximations like the hypernetted chain (HNC) [9] or the small cage expansion [10]. Both approaches differ on specific predictions, but agree on the same general picture. In recent times there has been considerable effort to reconcile the two approaches in the 'β regime', where, after a first fast relaxation, correlation functions remain close to a plateau value. Szamel [11] has derived the MCT equation for the non-ergodicity parameter using a particular closure of the replicated Yvon–Born–Green (YBG) hierarchy for the density correlation function. Moreover the celebrated exponent parameter of MCT, has been related to amplitude ratios between equilibrium correlation functions [12] and has been computed in the framework of replicated liquid theory [13–15]. One of the difficulties in unifying equilibrium and dynamical approaches lies in the necessity of including conservation laws (mass, momentum and energy) in the dynamical description at short times. However this problem should disappear in the long time ‘α regime’ where correlations decay below their plateau value. Here we show that progress can be achieved using a coarse grained description where the details of the short times are neglected and one concentrates on the slow part of the dynamics. Emerging properties of glassy relaxation, as temperature-time superposition principle [8] or the appearance of effective temperatures [16], can be rationalized through the hypothesis that quasi-equilibrium principles drive the dynamics during the phase space exploration in the long time regime [17]. In a nutshell, the two steps glassy relaxation at low temperature can be described as a first rapid decay inside a metastable state, followed by a very slow relaxation in a complex landscape of metastable states. This relaxation can be thought as a random walk in phase space where the states that are available at a given time are selected according to their Boltzmann weight. This insight has been formalized in [18] through the introduction of an appropriate Markov chain construction [19, 20]. Consider a system in a discrete time dependent configuration specified by $S_t$ and subjected to a Hamiltonian $H(S)$ and an overlap function $Q(S, S') = \int dx dy w(x-y)\rho_{S}(x)\rho_{S'}(y)$ that measures the similarity between density profiles of couples of configurations5. One can define the Boltzmann pseudodynamics with the following discrete time Markov chain

$$M(S_{\tau+1}|S_{\tau}) = \frac{e^{-\beta H(S_{\tau+1})}}{Z[S_{\tau}]}\delta\left(Q(S_{\tau+1}, S_{\tau}) - C(\tau + 1, \tau)\right)$$

which implements the previous idea, assuming that the states available at a given time $\tau + 1$ are the ones that lie at an overlap $C(\tau + 1, \tau)$ from the configuration at time $\tau$. This construction has been employed in mean field spin glasses where Boltzmann pseudodynamics provides an exact coarse grained version of slow dynamics when an infinitely long chain is considered and the value of $C(\tau + 1, \tau)$ is such that the constraints do not affect the free-energy of the system [18].

This result is remarkable at the fundamental and practical levels. It provides a basic probabilistic description of how configuration space is explored during glassy relaxation.

5 The function $w$ is a coarse graining function and its precise form is irrelevant in this context provided it is short ranged.
Moreover it opens the possibility to study long time dynamics through equilibrium techniques.

We want to study the Boltzmann pseudodynamics in glassy liquids, and to discuss its relation with present dynamical theories, in particular, with the MCT of the glass transition and the theory of aging dynamics as is known from schematic spin models. We will need approximations to deal with the strongly interacting character of the systems. We will focus on (1), the well known HNC approximation and (2), the Szamel closure scheme of the replicated YBG hierarchy [11]. Our main results are:

- We introduce a long time dynamical version of the Ornstein–Zernike (OZ) equations. These equations can be used within any selected closure scheme and are quite general MCT-like equations.
- We use the HNC approximation in order to close the OZ dynamical equations. We obtain a complete set of new dynamical equations that describe the evolution of liquids at low temperature. Moreover, starting from an equilibrium initial condition our equations reduce to a single one for the correlation function. This equation displays the universal features of MCT with a singularity at a previously identified dynamical transition point [9]. Relaxation in the early and late \( \beta \) regime is characterized by power laws whose respective exponents \( a \) and \( b \) verify the MCT relation \( \Gamma (1 - a) / \Gamma (1 - 2a) = \Gamma (1 + b) / \Gamma (1 + 2b) = \lambda \). The exponent parameter \( \lambda \) coincides with the one computed in [13, 14]. Differently from MCT which needs the external input of the static structure factor, we get here a complete set of equations determining static and long time dynamical quantities.
- Always within HNC, starting from a strongly off-equilibrium situation, we find an aging regime. Modulo the identification of a soft mode of slow relaxation, the gross picture follows the main lines found in mean field spin glasses [21]. The system falls asymptotically in a non-equilibrium state where time translation invariance and fluctuation dissipation theorem are violated.
- We use Szamel’s scheme that allows to derive the canonical mode coupling equation for the slow part of the momentum dependent correlator in the critical regime. We thus unveil that, within MCT, the relaxation in the whole \( \alpha \) regime consists in a quasi-equilibrium exploration of the configuration space that explains the insensitivity of this regime from the details of dynamics [22, 23].

**Boltzmann pseudodynamics.** In order to study the pseudodynamic process one can start from the probability of a trajectory of length \( L \)

\[
P(S_L, S_{L-1}, \ldots, S_0) = \frac{e^{-\beta_0 H[S_0]}}{Z(\beta)}
\]

\[
\times \prod_{\tau=0}^{L-1} \frac{e^{-\beta H[S_{\tau+1} - \tau]}}{Z[S_{\tau}, C(\tau + 1, \tau)]} \delta(Q(S_{\tau+1}, S_\tau) - C(\tau + 1, \tau))
\]

where \( Z(\beta) = \sum_S e^{-\beta H[S]} \) and \( Z[S, C] = \sum_{S'} e^{-\beta H[S']} \delta(Q(S', S) - C) \) are normalization factors. The choice of \( \beta_0 = \beta \) corresponds to choose an equilibrium initial condition at temperature \( \beta \), while \( \beta_0 = 0 \) is useful to model a high temperature quench and an aging situation. The probability (2) can be used to compute averages of observables or the free-energy of the system as a function of time. One way to deal with this construction is to use the replica method and substitute the partition functions \( Z[S_{\tau}, C(\tau + 1, \tau)] \) in the denominator of
\(Z^{-1}\{S, C(\tau + 1, \tau)\} \rightarrow Z\{S, C(\tau + 1, \tau)\}^{n-1}\). As usual, the numbers of replicas \(n_x\), that here can depend on time, is kept integer for intermediate computations and finally sent to zero. Enforcing the constraint in (1) through a Lagrange multiplier \(\nu(\tau)\), and expliciting the partition functions as sums over configurations, we obtain formally the expressions of the equilibrium of a system made of a mixture of different particle species so that static approximation schemes can be applied. Each system is indexed by a time label that encodes for its position along the chain and a replica index \(a\) that goes from 1 to \(n_x\). We define for notational simplicity Greek indexes that code for both time and replica indexes, e.g. \(\alpha \equiv (\tau, a)\).

**Dynamical Ornstein–Zernike equations.** Let us introduce the basic fields that we study. We start with a chain of systems where each system is composed by \(N\) particles interacting through a pair potential \(\varphi(x)\). The replicated one and two points density functions are

\[
\rho_{ab}(x) = \sum_{i=1}^{N} \delta(x - x_{i}^{(a)}) \tag{3}
\]

\[
\rho_{ab}(x, y) = \left\langle \rho_{a}(x)\rho_{b}(y) \right\rangle - \left\langle \rho_{a}(x) \right\rangle \delta(x - y). \tag{4}
\]

Our starting point are the OZ equations for a mixture of particles \([24, 25]\)

\[
c_{ab}(x, y) = h_{ab}(x, y) - \rho \sum_{\tau} \int dz h_{ab}(x, z)c_{a}(z, y), \tag{5}
\]

where we have supposed that the density \(\left\langle \rho_{a}(x) \right\rangle = \rho\) is constant. Here \(\rho^2 h_{ab}(x, y) = \rho_{ab}(x, y) - \rho^2\), while \(c_{ab}(x, y)\) defines the direct correlation function.

We want to use this equation to study slow dynamics. As explained in \([18]\) this can be done seeking for continuous solutions in the infinite chain limit, where the time variables \(t = \tau/L\) etc. become continuous and \(\nu(t) \rightarrow 0\) for all \(t\). As usual in the replica method, we need to choose a parametrization of the replica matrix \(h_{ab}(x, y)\) that allows the analytic continuation to \(n \rightarrow 0\). In \([18]\) an appropriate form to describe long time dynamics was shown to be

\[
h_{ab}(x, y) = h(x, y; s, u) + \delta_{ab}\delta(s - u)\Delta h(x, y; s, u)
+ \Theta(u - s)\delta_{ab}\Delta h(x, y; u, s)
+ \Theta(s - u)\delta_{ab}\Delta h(x, y; s, u) \tag{6}
\]

where \(a = (s, a)\) and \(b = (u, b)\). The Heaviside function \(\Theta(x)\) is here defined equal to 1 when \(x\) is strictly positive and zero otherwise. The ansatz (6) is consistent with causality and respects the replica symmetry of the problem. It can be shown that this is equivalent to assume the self-averaging of space averaged correlation and response functions with respect to the thermal noise. In the limit \(\{n_x\} \rightarrow 0\), \(h(x, y; s, u)\) represents the normalized density-density space and time dependent correlation function along the chain; remarkably, in the long chain limit \(du \approx 1/L\) and the function

\[
R_{b}(x, y; s, u) = \beta \Theta(s - u)\frac{\Delta h(x, y; s, u)}{du} \tag{7}
\]

for \(s > u\) appears to be the response function that encodes how the density in \(x\) at time \(s\) varies when a small perturbing pressure is applied to the system in \(y\) at time \(u\). Analogous expressions hold for \(c_{ab}(x, y)\) that therefore defines not only a direct time dependent correlation function but also a direct response function. The equal time quantities \(\Delta h(q; s, s)\) and \(\Delta c(q; s, s)\) encode the information on the short time dynamics, namely the jump in correlation from the
instantaneous to the plateau value. Assuming space translational invariance, using (7) and taking the infinite chain limit, the OZ equation (5) in Fourier space becomes

$$0 = W_q[h, c] + \frac{\rho}{\beta} \int_0^u dz \ h(q; s, z) R_e(q; u, z) + \frac{\rho}{\beta} \int_0^r dz \ R_h(q; s, z) c(q; z, u)$$  \hspace{1cm} (8)$$

$$W_q^{\text{eff}}[R_h, R_c] = -\frac{\rho}{\beta} \int_s^u dz R_h(q; z, s) R_c(q; u, z)$$  \hspace{1cm} (9)$$

$$\Delta h(q; s, s) = \Delta c(q; s, s) + \rho \Delta h(q; s, s) \Delta c(q; s, s)$$  \hspace{1cm} (10)$$

where we have defined $$W_q[h, c] = c(q; s, u) - h(q; s, u) + \rho [h(q; s, 0) c(q; u, 0) + h(q; s, u) \Delta c(q; u, u) + c(q; s, u) \Delta h(q; s, s)]$$ and $$W_q^{\text{eff}}[R_h, R_c] = R_e(q; u, s) - R_h(q; u, s) + \rho R_h(q; u, s) \Delta c(q; u, u) + \rho R_e(q; u, s) \Delta h(q; s, s)$$. These equations are valid for $$u \geq s$$; the equivalent ones hold for $$s > u$$. We notice that both (8) and (9) are compatible with equilibrium where time translational invariance (TTI) and fluctuation dissipation theorem $$R_h(q, s - u) = -\beta \Delta h(q, s - u)/ds$$ hold. In this case they reduce to the single equation

$$F_q[h] = \int_0^r dz \frac{dh(q; z)}{dz} [c(q; s - z) - c(q; s)]$$  \hspace{1cm} (11)$$

where $$F_q[h] = c(q; s) - h(q; s) + \rho (h(q; s) \Delta c(q) + c(q; s) \Delta h(q) + c(q; 0) h(q; s) - (h(q; s) - h(q; 0)) c(q; s))$$. This equation has the structure of a long time MCT equation [8] where the time derivative is neglected and the memory kernel is represented by the direct correlation function.

As in the usual static case, the OZ equations are useful in the context of closure approximations that provide a second equation that relates the response and correlation functions. The final equations depend on the closure one uses. We now proceed to the analysis of the HNC approximation and the Szamel closure.

**The HNC closure: equilibrium and aging solution.** We study now the closure provided by the HNC approximation for a particle mixture [26]. This closure has proved to provide a coherent qualitative picture of glassy phenomena on which we focus, despite it fails to give reliable quantitative results. In the present case it can be rewritten as

$$\ln[h_{\text{eff}}(x, y) + 1] + \beta \phi_{\text{eff}}(x, y) = h_{\text{eff}}(x, y) - c_{\text{eff}}(x, y)$$  \hspace{1cm} (12)$$

where for us $$\phi_{\text{eff}}(x, y) \equiv \phi_{\text{eff},\text{loc}}(x, y) = \delta_{x,a} \delta_{y,a} \phi(x - y) + \delta_{x,1} \delta_{y,1} w(x - y) v(x - y)$$ contains the inter particle potential at equal time and replica indexes and the Lagrange multiplier constraining the value of the overlap at consecutive times. Plugging the parametrization (6) into (12) we obtain for $$s \neq u$$

$$\ln[h(x; s, u) + 1] = h(x; s, u) - c(x; s, u)$$  \hspace{1cm} (13)$$

$$R_c(x; s, u) = R_h(x; s, u) \frac{h(x; s, u)}{h(x; s, u) + 1}.$$  \hspace{1cm} (14)$$

Together with (10) we obtain that the quantities $$\Delta h(q; s, s) \equiv \Delta h(q)$$ and $$\Delta c(q; s, s) \equiv \Delta c(q)$$ are s independent. The dynamical equations (8)–(14) provide a complete set of equations that can be solved in time. They are reparametrization invariant [21, 27]: knowing a solution $$h(q; s, u)$$, $$R_h(q; s, u)$$, a whole family is obtained by putting $$h'(q, s, u) = h(q, f(s), f(u))$$ and $$R_h'(q, s, u) = \frac{df}{ds} R_h(q; f(s), f(u))$$ (and the analogous expressions for c and R) provided that f(s) is an increasing function with f(0) = 0. Now we want to study the equilibrium dynamics. For $$T \rightarrow T^+_d$$ a nontrivial solution emerges that
satisfies equilibrium, (13) becomes TTI and trough FDT (14) reduces to the derivative of (13). The phenomenology displayed by the resulting equation is similar to the one of conventional MCT and its solution can be identified with the scaling function determining the correlation function within the temperature-time superposition principle [8]. Equation (11) is invariant under time rescaling $t \to At$ for positive $A$, and the condition for the existence of a decaying solution is that the operator $dF_0(c)/dh(k)$ has a zero mode $\mathcal{K}_0(q)$. By using this fact we can derive an analytic expression for the MCT exponent parameter $\lambda$

$$\lambda = \frac{\int d^3x \mathcal{K}_0^3(x)/(1 + h(x))^2}{2\rho \int \frac{d^3q}{(2\pi)^3} \mathcal{K}_0^3(q)[1 - \rho \Delta c(q)]}.$$  

Remarkably, this expression coincides with the one computed in [13, 14] using a the static HNC approach. Below the dynamical glass transition, the dynamical equations (8) and (9), under the assumption of a high temperature initial condition and loss of memory of the initial condition $h(q; s, 0) = c(q; s, 0) = 0$, admit a solution that describes an aging regime. The resulting equations could be solved in principle using the modified FDT relation $R_0(q; t, s) = \rho \Delta h(q; t, s)/\rho ds$ [21]. The combination $T/X$ is interpreted as an effective temperature [16], and $X$ is independent of $q$. As in [21], the FDT ratio $X \in [0, 1]$ is fixed by the requirement that the equation for the response function in the limit $u \to \infty$ admits a non vanishing solution. This is only possible in presence of a zero mode and it turns out that this is equivalent to the marginal stability condition in spin glass dynamics [21]. The actual computation of $X$ as a function of the control parameters turns out to be numerically quite invoved and we leave it for future works.

A different closure: conventional MCT equations. The crucial point to obtain the dynamical equations (8)–(11) is that, modulo a different interpretation of the replica indexes, there is formal coincidence of pseudodynamic OZ equations with the ordinary equations of liquid mixtures. This coincidence, which can be traced in the symmetry of the pseudodynamic effective Hamiltonian under replica index exchange, should be respected by any consistent approximation scheme. It has been observed in [11] that the MCT equation specifying the non-ergodicity parameter can be derived from replicated OZ equation, through a defned closure of YBG hierarchy leading to the following approximation for the non diagonal elements of the replica directed correlation function $c_{\alpha\beta}(q)$ for $\alpha \neq \beta$

$$c_{\alpha\beta}(k) = \int dq \ V(k, q)h_{\alpha\beta}(q)h_{\alpha\beta}(k - q)$$  

where the $V(k, q)$ is the mode coupling vertex given in terms of the static direct correlation function $c_0(q)$ as

$$V(k, q) = \frac{1}{16\pi k^2} \left[ k \cdot (q c_0(q) + (k - q)c_0(k - q)) \right]^2,$$  

which is independent of the replica indexes. We can then interpret (16) in pseudodynamics with $\alpha \to (a = 1, t), \beta \to (a = 1, s = 0)$ with $t > 0$. Assuming TTI, we get the expression for the dynamical direct correlation function

$$c(k, t) = \int dq \ V(k, q)h(q, t)h(k - q, t).$$  

Inserting the previous equation into (11), after some simple algebra we obtain exactly the Götze mode coupling equation for the slow part of the relaxation. From these equations it can

2 The detailed derivation will be given elsewhere.
be derived the expression for the exponent parameter whose expression coincides with the one derived in [28]. We notice that in [29] MCT has been generalized to describe aging below $T_d$. We leave for future work the analysis of possible generalizations Szamel closure to this case.

Conclusions

We have shown that within liquid theory approximations, under marginal conditions, Boltzmann pseudodynamics gives rise to a dynamical picture strictly following the MCT phenomenology. We have been able to derive dynamical MCT-like equations in a coherent scheme that includes both equilibrium and aging dynamics in the context of the HNC approximation. These equations allow to describe equilibrium dynamics on approaching $T_d$ and aging dynamics below $T_d$. We have computed the zero mode and the exponent parameter $\lambda$ from these equations. In equilibrium both these quantities coincide with the ones computed with completely static techniques in [13]. In aging, the description is coherent with the $p$-spin picture [21], but goes beyond this schematic model. We also found that the conventional MCT equation can be obtained from our replica-dynamic Ornstein–Zernike equations generalizing to the pseudodynamic a closure scheme of the YBG hierarchy proposed by Szamel. Our analysis demonstrates that critical MCT and MCT-like dynamical behavior in equilibrium and aging is equivalent to a quasi-equilibrium sampling of configuration space. We expect a similar picture to hold in other approximation schemes. In particular it will be interesting to study the case of small cage approximation that has been found to give exact result in the limit of high dimensionality [30]. These are mean field like results and the usual caveats apply. It is well known that due to activated processes the sharp transition becomes a dynamical cross-over in real life. One should then address the question if our Boltzmann pseudodynamics description is valid beyond mean field. To our view, quasi-equilibrium exploration of configuration space is a fundamental property of glassy dynamics that should also hold when mean field theory fails. Further work will be needed to test this conjecture. Moreover it could be interesting to see what the predictions of this approach are for the theory of dynamical fluctuations in the $\alpha$ regime. Recently it has been shown in [31] using equilibrium techniques that the dynamical fluctuations in the $\beta$ regime are described by a scalar cubic field theory in a random field. It could be very interesting to test if our approach can be used to test if these results extends to the $\alpha$ regime.

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