Equations of structural relaxation

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Abstract

In the mode coupling theory of the liquid to glass transition the long time structural relaxation follows from equations solely determined by equilibrium structural parameters. The present extension of these structural relaxation equations to arbitrarily short times on the one hand allows calculations unaffected by model assumptions about the microscopic dynamics and on the other hand supplies new starting points for analytical studies. As a first application, power–law like structural relaxation at a glass–transition singularity is explicitly proven for a special schematic MCT model.

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§1. Introduction

Dense liquids exhibit anomalously slow, temperature sensitive and non-exponentional relaxation processes. They are conventionally termed ‘structural relaxation’ because cooperative rearrangements of particles are presumably involved. The mode coupling theory (MCT) suggests that the evolution of the anomalous dynamics and its splitting off from the normal liquid dynamics is caused by bifurcation singularities in non–linear retarded equations of motion for the (normalized) intermediate scattering functions \( \Phi_q(t) = S_q(t)/S_q \) (Leutheusser 1984, Bengtzelius et al. 1984). These functions are some of the most simple ones specifying structural dynamics. The bifurcations of MCT, and the long–time dynamics in their vicinity, are solely determined by the equilibrium (static) structure factor \( S_q \), see (Götze and Sjögren 1989, Franosch et al. 1998), and thus the MCT provides a frame to single out and to define structural relaxation. In this contribution the MCT equations for the structural relaxation are extended to arbitrarily short times thus rendering them well defined even in the limit that the microscopic transient dynamics can be neglected. Whereas in (Franosch et al. 1998) this was achieved by a discrete–dynamics model, here integral equations with explicit initial conditions are formulated. Thus the dependence on the initial conditions is displayed clearly. Additionally, monotone solutions are guaranteed (Fuchs et al. 1991). Furthermore, for the obtained structural dynamics equations non–exponential relaxation can be proven for a special MCT model.

§2. The equations of motion of MCT

The MCT equations of motion for the autocorrelation functions of density fluctuations in simple liquids, \( \Phi_q(t) \), are derived from Newton’s equations with the Zwanzig–Mori formalism leading to:

\[
\partial_t^2 \Phi_q(t) + \Omega_q^2 \Phi_q(t) + \int_0^t dt' M_q(t - t') \partial_{t'} \Phi_q(t') = 0 ,
\]

\[
\Phi_q(t \to 0) = 1 - \frac{1}{2}(\Omega_q t)^2 ,
\]
and subsequent mode coupling approximations for the generalized longitudinal viscosity finally resulting in:

\[ M_q(t) = \Omega_q^2 m_q(t) + M_{\text{reg}}^q(t) , \quad (3) \]

\[ m_q(t) = \sum_{k+p=q} V(q; k, p) \Phi_k(t) \Phi_p(t) . \quad (4) \]

In eqn. (1), the frequency \( \Omega_q \) sets the microscopic time scale for the bare oscillators with wave vector \( q \). In eqn. (3), viscous damping arises from mode–mode coupling, \( m_q(t) \), eqn. (4), and from microscopic short–time dynamics denoted by \( M_{\text{reg}}^q \); see (Götze 1991) for explicit expressions and discussions. A Laplace transformation of eqs. (1) and (3) using \( \hat{\Phi}(s) = \int_0^\infty dt \: e^{-st} \Phi(t) \) leads to

\[ \hat{\Phi}(s) = \frac{1}{[s + 1 / [ \hat{m}(s) + (s + \hat{M}_{\text{reg}}(s))/\Omega_q^2 ]] } . \quad (5) \]

The existence of a unique solution of the MCT equations which has all properties of an autocorrelation function has been shown by Haussmann (1990). Further and more detailed properties could be proven by Götze and Sjögren (1995) upon the assumption of overdamped short time motion. Let us mention two results relevant for this work, namely (i) the existence of bifurcations in the equations derived from eqs. (1) and (4) for the long–time limits, \( f_q = \lim_{t \to \infty} \Phi_q(t) \); here the \( f_q \) are the glass form factors and the bifurcations happen for critical values of the mode coupling vertices, \( V = V^c \), where \( f_q = f_q^c \). (ii) for regular vertices, the \( f_q \) depend smoothly on the \( V \), and there exists a longest relaxation time, \( 1/\Gamma \), cutting off the final relaxation exponentially, \( \Phi_q(t) - f_q = O(e^{-\Gamma t}) \) for \( V \neq V^c \).

One of the most simple models exhibiting the generic features of the MCT bifurcation dynamics in eqs. (1) to (4) is obtained by the simplification to study only one correlator, \( \Phi(t) \), which experiences non–linear feedback via one mode–coupling functional \( m(t) = v_1 \Phi(t) + v_2 \Phi^2(t) \) with \( v_{1,2} \geq 0 \) (Götze 1984). We will start by discussing this so–called \( F_{12} \)–model before generalizing our results to the equations relevant for simple liquids.

§3. Results for the schematic \( F_{12} \)–model
‘Equations of the structural relaxation’ in the $F_{12}$–model are equations of motion determined solely by the vertices $v_1$ and $v_2$ which play the role of the static structural information entering the MCT vertices $V$ in eqn. (4). Bifurcations in the model lie on two lines parametrized by the exponent parameter $\lambda$ (Götze 1984): type–B lines, where $f^c = 1 - \lambda > 0$, at $v^c_1 = (2\lambda - 1)/\lambda^2$ and $v^c_2 = 1/\lambda^2$ with $1/2 \leq \lambda < 1$, and type–A transitions, where $f^c = 0$, at $v^c_1 = 1$ and $v^c_2 = \lambda$ for $0 \leq \lambda \leq 1$. Whereas in eqs. (1) to (4) the microscopic transient is required in order for solutions to exist (Götze 1991), after partial integration the short–time variation can be neglected, $\partial^2_t \Phi(t) + \int_0^t dt' M^{reg.}(t - t') \partial_t \Phi(t') \ll \Omega^2 \Phi(t)$, leading to:

$$\Phi(t) = m(t) - \frac{d}{dt} \int_0^t dt' m(t - t') \Phi(t'),$$

(6)

while the expression for the memory function is not changed:

$$m(t) = v_1 \Phi(t) + v_2 \Phi^2(t).$$

(7)

Demanding the solutions to be of regular variation and that the Laplace transforms $\hat{\Phi}(s)$ and $\hat{m}(s)$ exist, enabling one to use Tauber and Abel theorems, see e. g. (Feller 1971), the equations of structural relaxation have to be completed by specifying the initial variation:

$$\Phi(t) \rightarrow (t/t_*)^{-1/3} \quad \text{for} \quad t \rightarrow 0.$$  

(8)

In this case, the convolution integral, which will be abbreviated as $\int_0^t dt' m(t - t') \Phi(t') = (m * \Phi)(t)$, leads to $(m * \Phi)(t \rightarrow 0) \rightarrow v_2 t_* B(1/3, 2/3)$, where $B(x, y)$ is Euler’s Beta function. The Laplace transform of eqn. (3) for all $s$ then is:

$$(1 + s \hat{m}(s)) \hat{\Phi}(s) = \hat{m}(s) + v_2 t_* B(1/3, 2/3),$$

(9)

It is a straightforward exercise to verify that the last term in eqn. (4) is required for the existence of solutions with the requested properties (Voigtmann 1998), thus leading to eqn. (8).

Note, that the bifurcations to non–ergodic solutions cause $\hat{\Phi}$ and $\hat{m}$ to increase for small frequencies, suggesting to neglect $(s + M^{reg.})\Omega^{-2} \ll \hat{m}(s)$ in eqn. (4) in order to derive
equations for the structural relaxation alone. However, this procedure misses the non-analytic behaviour of \((m \ast \Phi)(t \to 0)\) connected to the limit \(s \to \infty\).

One can extend eqn. (8) by a short-time series expansion, see appendix A,

\[
\Phi(t) = (t/t_\ast)^{-1/3} \left(1 + \sum_{n=1} c_n \left(t/t_\ast\right)^{n/3}\right) \quad \text{for } t < r t_\ast .
\]  

(10)

Figure 1 shows the numerically obtained radii of convergence, \(r\), of eqn. (10) along the line of bifurcations in the \(F_{12}\)-model.

Figure 2 shows numeric solutions of eqs. (6) to (8) which exhibit slow relaxation processes upon approach of a bifurcation singularity at critical values of the coupling vertices. This explains the sensitive dependence of the dynamics on small regular changes in \(\varepsilon\), which measures the distance to the singularity. The algorithm described in (Fuchs et al. 1991) is well suited for the numerical integration. A type-B fold bifurcation at \(\varepsilon = 0\) separates ergodic, \(\Phi(t \to \infty) = f = 0\) for \(\varepsilon < 0\), from non-ergodic dynamics, \(f \geq f^c > 0\) for \(\varepsilon \geq 0\). Asymptotic expansions identify two divergent time scales connected with two power law relaxation windows (Götze 1984). As demonstrated e.g. in (Franosch et al. 1998) the long-time solutions of the complete MCT equations of motion, eqs. (1) to (4), become independent of the microscopic transient. For long times they agree with the solutions of the equations of structural relaxation, eqs. (6) and (7). This is exemplified in figures 2 and 3 using two approximations for the regular microscopics in the \(F_{12}\)-model: undamped oscillation, \(M^{\text{reg}} \equiv 0\), and pure relaxation, \((s + \dot{M}^{\text{reg}}(s))\Omega^{-2} \to \Gamma_0\). It is apparent that oscillatory short-time dynamics masks the structural relaxation more strongly than relaxational.

Assuming regular variation of the solutions of eqs. (1) to (4), at a bifurcation point, power law relaxation could be argued:

\[
\Phi(t) = f^c + h \left(t/t_0\right)^{-a}, \quad \text{for } t \to \infty \quad \text{and } v_{1,2} = v^c_{1,2},
\]  

(11)

where the exponent \(a\) follows from the exponent parameter \(\lambda\), which is a function of \(v^c_{1,2}\), via \(\lambda = B(1 - a, 1 - a)(1 - 2a)\) (Götze 1984). From the equations of structural relaxation, eqs. (3) and (4), and the short-time expansion eqn. (10), one concludes that this asymptote
requires \( t > t_\ast r \) in general. The transient time \( t_0 \) is obtained from matching eqn. (11) to eqn. (14); while the critical amplitude \( h = (1 - f^c) \) for the \( F_{12} \)-model.

In the \( F_{12} \)-model there exist two points where the short-time expansion can be found explicitly and solves the equations of structural relaxation, eqs. (3) to (8), for all times, i.e. \( r \to \infty \) in eqn. (14); see appendix A and figure 1:

\[
\Phi(t) = f^c + (t/t_\ast)^{-1/3}, \quad \text{for} \quad \lambda = \lambda_\ast = \frac{1}{3} B(\frac{2}{3}, \frac{2}{3}) = 0.684 \ldots \quad \text{and} \quad v_{1,2} = v^c_{1,2}. \quad (12)
\]

Both points are bifurcation points, as finite distances from the singularities lead to the existence of long-time relaxation rates as shown in appendix B, see also (Götze 1984, Götze 1991). Furthermore, the initial condition, eqn. (8), requires for the critical exponent \( a = a_\ast = 1/3 \) which is satisfied for \( \lambda = \lambda_\ast \). Thus, eqn. (12) verifies power-law relaxation at a bifurcation singularity of equations describing the structural relaxation of a schematic MCT model.

The existence of a solution of the integral equations for all times at a singular point also allows for these special cases to deduce a long-time solution for small distances from the bifurcation in an expansion which can be expected to have a finite radius of convergence, \( (t/t_0)|\epsilon|^{3/2} < r_\beta \).

\[
\Phi(t) \to (t/t_0)^{-1/3} \left[ 1 + \sum_{n=1} A_n \left( \epsilon (t/t_0)^{2/3} \right)^n \right], \quad \text{for} \quad t \to \infty. \quad (13)
\]

The calculation sketched in appendix B, thus introduces the \( \beta \)-time scale, \( t_\epsilon = t_0|\epsilon|^{-1/2a} \) as \( a = 1/3 \) for \( \lambda = \lambda_\ast \), and agrees with the expansion of the so-called \( \beta \)-correlator for small rescaled times \( \tilde{t} = t/t_\epsilon \); see (Götze 1984). There it is also shown for general \( \lambda \), how to extend this solution to even longer times. Note that the expansion carried through in appendix B does not require a matching to the critical decay.

§4. Extensions to MCT equations for simple liquids

The extension of the analysis of the bifurcation dynamics of schematic models to the full MCT equations for simple liquids, eqs. (11) to (14), has been achieved in (Götze 1985) and
rests upon the central–manifold concept. The generalization of the equations of structural relaxation to coupled, wave vector dependent equations is trivial; eqn. (3) attains $q$–indices and eqn. (7) is replaced by eqn. (4). For quadratic polynomials in eqn. (4), the power law of the initial condition is not changed:

$$\Phi_q(t) \rightarrow x_q \, t^{-1/3} \quad \text{for } t \to 0; \quad x_q \geq 0. \quad (14)$$

It is surprising that a vector $x_q$ has to be specified in order to render the solutions unique; recall that owing to the factorization property of the critical decay, $\Phi_q(t) = f_q^c + h_q(t/t_0)^{-a}$ (Götze 1985), the long–time dynamics of the structural relaxation becomes unique by specifying only the one matching time $t_0$.

From simple generalizations of the calculations presented in appendix A it is obvious that the short–time expansion, eqn. (10), can be obtained in the wave vector dependent case also.

§5. Conclusions

Definitions of equations for structural relaxation not restricted to long times are given within the MCT. These equations, eqs. (4) and (6), together with the initial conditions, eqn. (14), determine smooth but sensitively varying intermediate scattering functions $\Phi_q(t)$ from the equilibrium structure of the liquid; the static structure factor $S_q$ is the only quantity appearing in the equations of motion. The possibility to specify an arbitrary initial non–negative amplitude, $x_q$, is worth stressing and also holds for the discrete dynamics model of (Franosch et al. 1998). The short time divergence of the solutions of the structural relaxation equations renders them unphysical for too short times; they violate the imposed normalization of the $\Phi_q(t)$. This stresses that the physical mechanism (the so–called ‘cage effect’) causing structural relaxation is relevant for long–time dynamics only. Any complete description of the density fluctuations requires a transient to regular microscopic short–time dynamics. Nevertheless, extending the structural relaxation equations to all times on the one hand allows the definition of a lower limit of validity of this long time description —
the time where the solutions increase above unity could be chosen — and on the other hand presents a conceptual simplification as some transient effects are eliminated. Thus for example, analytical proofs of power–law relaxation at bifurcation points become possible for special cases; recall that at regular points of the full MCT equations a longest relaxation time exists. Also, the smaller of the two divergent time scales of MCT could be identified from a perturbation expansion. Moreover, the aspect that MCT equations of motion with different models for the short–time microscopic motion lead to the identical long–time dynamics is again observed.

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**Appendix A**

Entering the ansatz $\Phi(t) = f^c + \phi(t)$ into eqs. (6) and (7) leads to

$$\dot{\bar{\sigma}} = \bar{\sigma} \phi(t) - \bar{\lambda} \phi^2(t) + (1/\lambda' + \bar{\sigma}) (\phi \ast \phi)(t) + (\phi^2 \ast \phi)(t)$$

where a dot denotes a time–derivative and the following abbreviations are used: $\bar{\lambda} = \lambda/\lambda'$, $\lambda' = \lambda/(1 - f^c)$, $\bar{\sigma} = (1 - f^c)^2\sigma/\Delta$, $\sigma = (1 - f^c)(\delta v_1 f^c + \delta v_2 f^c^2)$, $\Delta = (\lambda' + (1 - f^c)^2\delta v_2)$, $1/\lambda' + \bar{\sigma} = [1 + (1 - f^c)^2(\delta v_1 + 2\delta v_2 f^c)]/\Delta$, and $\bar{\sigma} = (1 - f^c)(1 + \bar{\sigma} - (1 + \sigma)/\Delta)$ which are obtained from $v_{1,2} = v_{1,2}^c + \delta v_{1,2}$, where the limit of $v_2$ approaching zero has been excluded.

In the vicinity of a type–B transition, $\lambda' = 1$, $\bar{\lambda} = \lambda$. Close to a type–A transition $\lambda' = \lambda$, and thus $\bar{\lambda} = 1$. From the short–time expansion, eqn. (10), one obtains a recursion relation for $\tilde{c}_n$, where $\tilde{c}_0 = 1$, $\tilde{c}_1 = c_1 - f^c$ and $\tilde{c}_n = c_n$ else, and $\delta_{n,m}$ is a Kronecker symbol.

$$\tilde{c}_{n+1} = \frac{3/(n + 1)}{(2B(\frac{n+2}{3}, \frac{2}{3}) + B(\frac{n+3}{3}, \frac{1}{3}))} \left\{ \sum_{m=0}^{n} \left[ \bar{\lambda} - \frac{n+1}{3} B(\frac{n-m+2}{3}, \frac{m+2}{3})(1/\lambda' + \bar{\sigma}) \right] \tilde{c}_{n-m} \tilde{c}_m + \bar{\sigma} \delta_{n,2} - (1 - \delta_{n,0}) [ \bar{\sigma} \tilde{c}_{n-1} + \frac{n+1}{3} \sum_{m=1}^{n} \sum_{k=0}^{m} B(\frac{n-k+2}{3}, \frac{k+2}{3}) \tilde{c}_{m-k} \tilde{c}_k + B(\frac{n+2}{3}, \frac{2}{3}) \tilde{c}_m) \tilde{c}_{n+1-m} \right\}$$

(A2)
At two singular points, where $\hat{\sigma} = \hat{\sigma} = \bar{\sigma} = 0$ and $\lambda = \lambda_* = B(\frac{2}{3}, \frac{2}{3})/3 = 0.684\ldots$, all $\bar{c}_n$ vanish. This proves eqn. (12). Approaching the $F_1$–model at $(v_1, v_2) = (v_1, 0)$ (Götze 1991), the radius of convergence of eqn. (10) shrinks as for a $m$–th order polynomial mode coupling functional a similar expansion with $\Phi(t) \to (t/t_*)^{-1/(m+1)}$ for $t \to 0$ applies (Voigtmann 1998).

Appendix B

Close to the type–B transition at $\lambda = \lambda_*$, the eqn. (A1) can be expanded around the solution $\phi_0(t) = t^{-1/3}$ at $\delta v_{1,2} = 0$. Using $\phi(t) = \phi_0(t) + \phi_1(t)$, specifying the path such that $\hat{\sigma} = 0$ for simplicity and integrating this leads to:

$$\int_0^t ds \, K(t, s) \, \phi_1(s) = \hat{\sigma} t - \frac{2}{3} \hat{\sigma} t^{2/3} - ((\hat{\sigma} + \phi_1 + 2\phi_0\phi_1) * \phi_1)(t) + ((\lambda_* - \phi_0 - \phi_1) * \phi_2^2)(t),$$

(B1)

where the Volterra kernel is of generalized Abel form:

$$K(t, s) = 2 \, (t - s)^{-1/3} - 2\lambda_* \, s^{-1/3} + (t - s)^{-2/3} + 2 \, (t - s)^{-1/3} \, s^{-1/3}.$$  

(B2)

From the short–time expansion eqn. (10), $\phi_1(t \to 0) \propto t^{1/3}$ can be deduced in this case, ruling out that Kernel $K$ has eigenvalue zero. The expansion $\phi_1(t) = \sum_{n=1}^{\infty} \sigma^n \, g^{(n)}(t)$ thus leads to a recursive set of solvable linear integral equations for the $g^{(n)}$. If their long–time behaviour is searched for, only the first two terms in $K$ need to be considered and by induction one shows that $g^{(n)}(t) \to A_n \, t^{(2n-1)/3}$ for $t \to \infty$, where the $A_n$ obey: $A_1 = 1/(2(B(\frac{2}{3}, \frac{4}{3}) - \lambda_*))$ and

$$A_n = -\frac{1}{2} \left[ \sum_{m=1}^{n-1} A_{n-m} A_m \left( B\left(\frac{2(n-m+1)}{3}, \frac{2m+2}{3}\right) - \lambda_* \frac{2n+1}{3} \right) / \left( B\left(\frac{2}{3}, \frac{2n+2}{3}\right) - \lambda_* \frac{2n+1}{3} \right) \right].$$

(B3)

Replacing $\sigma$ with $\varepsilon$, this coincides with eqn. (13) and the short–time expansion of the $\beta$–correlator for $\lambda = \lambda_*$ (Götze 1984).
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Figure captions

Figure 1: Radius of convergence \( r \) of eqn. (10) numerically obtained using the Cauchy–Hadamard criterion and eqn. (A2) along the line of bifurcations in the \( F_{12} \)-model. The line parametrized by \( \lambda \) is given by \( v_2^c = 1/\lambda^2 \) and \( v_1^c = (2\lambda - 1)/\lambda^2 \) for type–B \((f^c > 0)\), and by \( v_2^c = \lambda \) and \( v_1^c = 1 \) for type–A \((f^c = 0)\) transitions. The vertical dashed lines indicate \( \lambda^* \) where eqn. (10) with \( c_{n>1} \equiv 0 \) holds for all times; i.e. \( r \to \infty \). The dot–dashed line describes the linear decrease, \( r \propto \lambda \), see appendix A, upon approaching the \( F_1 \)-model at \((v_1^c, v_2^c) = (1, 0)\).

Figure 2: Correlators \( \Phi(t) \) obtained from eqs. (6) and (7) for exponent parameter \( \lambda = 0.70 \). The bold line marked by a \( c \) corresponds to the bifurcation point and the others are calculated for \( v_{1,2} = v_{1,2}^c(1 + \varepsilon) \), \( \varepsilon = \pm 1/4^n, n = 0, 1, \ldots \) as labelled. The time scale \( t_0 \) from eqn. (11) is indicated and used in the inset to compare the curve at \( v^c \) to one crossing over to oscillations at short times (dashed line calculated for the \( F_{12} \)-model from eqs. (1) to (4) with \( M_{\text{reg}} \equiv 0 \)) and to another with overdamped short–time dynamics \( ((s + \dot{M}_{\text{reg}}(s))\Omega^{-2} \to \Gamma_0, \text{long dashes}) \).

Figure 3: Comparison of correlators at bifurcation points of the \( F_{12} \)-model for different microscopic transients and shifted using \( f^c \) are plotted versus rescaled time, with \( t_0 \) from eqn. (11). Solutions for exponent parameters \( \lambda_* = 0.684 \) and \( \lambda_\pm = \lambda_* \pm 0.15 \) as labelled are shown obtained from the equations of structural relaxation, eqs. (3) and (7) (solid lines), and from eqs. (1) to (4) with undamped oscillatory \( (M_{\text{reg}} \equiv 0, \text{short dashes; lines terminated for } t \lesssim 10^2 t_0) \) and with overdamped \( ((s + \dot{M}_{\text{reg}}(s))\Omega^{-2} \to \Gamma_0, \text{long dashes}) \) short–time dynamics. Dashed–dotted curves indicate the critical decay law, \((t_0/t)^\alpha \) with \( a_+ = 0.26 \) and \( a_- = 0.39 \) corresponding to the \( \lambda_\pm \). For \( \lambda_* \), where \( a_* = 1/3 \), the structural relaxation follows the critical decay for all times. Black circles (diamonds) indicate where the structural dynamics (critical decay) curves deviate by 20 % in horizontal direction from the solutions with overdamped microscopics.
Fuchs and Voigtmann, Equations of structural ... Fig. 1
Fuchs and Voigtmann, Equations of structural ... Fig. 2
Fuchs and Voigtmann, Equations of structural ... Fig. 3