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Gaussian density fluctuations and mode coupling theory for supercooled liquids

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Abstract. – The equations of motion for the density modes of a fluid, derived from Newton's equations, are written as a linear generalized Langevin equation. The constraint imposed by the fluctuation-dissipation theorem is used to derive an exact form for the memory function. The resulting equations, solved under the assumption that the noise, and consequently density fluctuations, of the liquid are Gaussian distributed, are equivalent to the random phase approximation for the static structure factor and to the well-known ideal mode coupling theory (MCT) equations for the dynamics. This finding suggests that MCT is a theory of fluid dynamics that becomes exact in a mean-field limit.

The description of the dynamics of supercooled liquids is one of the most intriguing goals in condensed-matter physics. Different approaches have been pursued in the last decades [1–7], originating both from the physics of liquids and from the physics of disordered systems. Many of these approaches strongly suggest that two different mechanisms for the decay of fluctuations are active in two different temperature ranges. A crossover temperature T_c separates the region of "weak" supercooling from the region of "strong" supercooling.

The description of the long-time dynamics and the associated evaluation of the transport coefficients in the strong supercooling region (below T_c) has proved to be an extremely difficult task [2, 4, 8]. The thermodynamic description of the liquid state between T_c and the calorimetric glass transition has been attempted, but no well-defined connection between thermodynamics and dynamics has been achieved as yet. In the weak supercooling regime, detailed predictions for the space and time dependence of the long-time decay of density correlations have been formulated using the ideal mode coupling theory (MCT) [1], one of the first approaches to identify the existence of the crossover temperature. The agreement of MCT predictions with experimental findings [9, 10] and molecular dynamics simulations [11–13] —both for atomic and molecular models— supports the view that MCT is indeed able to describe the slow dynamics in weak supercooled states.

Despite its remarkable practical success, the presence of apparently uncontrolled approximations in the derivation of the MCT equations makes it difficult to gain insights into possible improvements of the theory. The aim of this letter is to present a new derivation of the ideal MCT equations, starting from the microscopic equations for the evolution of the density (Newton's equations) and writing them as a linear generalized Langevin equation. A formally exact expression for the memory kernel is derived and, on making the approximation that the noise in the Langevin equation is Gaussian, the standard MCT equations are obtained. Note that the proposition of Gaussian noise implies that the density fluctuations are also Gaussian [14]. The outcome of the present approach offers a route to MCT that involves clear and known approximations for statics and dynamics. At the same time it suggests how MCT equations can be developed for more complex systems such as molecules and polymers.

The density of a system composed of N particles located at positions $\boldsymbol{r}_j$ is $\rho(\boldsymbol{r},t) \equiv \sum_{j=1}^{N} \delta(\boldsymbol{r}-\boldsymbol{r}_j(t))$. The Fourier transform of $\rho(\boldsymbol{r},t)$ is $\rho_{\boldsymbol{k}(t)} \equiv \sum_{j=1}^{N} e^{i\boldsymbol{k}\cdot\boldsymbol{r}_j(t)}$. In the case of a pairwise additive potential, the time evolution of $\rho_{\boldsymbol{k}(t)}$ can be written as [15]

$$\ddot{\rho}_{\boldsymbol{k}(t)} = -\sum_j (\boldsymbol{k}\cdot\dot{\boldsymbol{r}}_j(t))^2 e^{i\boldsymbol{k}\cdot\boldsymbol{r}_j(t)} - \frac{1}{mV}\sum_{\boldsymbol{k}'} v_{k'}(\boldsymbol{k}\cdot\boldsymbol{k}')\rho_{\boldsymbol{k}-\boldsymbol{k}'}(t)\rho_{\boldsymbol{k}'}(t)\,, \tag{1}$$

where v_k is the Fourier transform of the pair potential. Equation (1) is the Newton equation for the variables $\rho_{\boldsymbol{k}}(t)$.

The equation of motion for the density (eq. (1)) can be rewritten as

$$\ddot{\rho}_{\boldsymbol{k}}(t) + \omega_k^2 \rho_{\boldsymbol{k}}(t) = \mathcal{F}_{\boldsymbol{k}}(t)\,, \tag{2}$$

where the linear term in $\rho_{\boldsymbol{k}}(t)$, that represents the elementary excitations of the system, has been explicitly isolated. These excitations can be interpreted as "phonons", that would oscillate with frequencies ω_k^2 if no interaction force amongst them were present. Combining eqs. (1) and (2) the residual force $\mathcal{F}_{\boldsymbol{k}}(t)$ has the formal expression

$$\mathcal{F}_{\boldsymbol{k}}(t) = \omega_k^2\rho_{\boldsymbol{k}}(t) - \sum_j (\boldsymbol{k}\cdot\dot{\boldsymbol{r}}_j(t))^2 e^{i\boldsymbol{k}\cdot\boldsymbol{r}_j(t)} - \frac{1}{mV}\sum_{\boldsymbol{k}'} v_{k'}(\boldsymbol{k}\cdot\boldsymbol{k}')\rho_{\boldsymbol{k}-\boldsymbol{k}'}(t)\rho_{\boldsymbol{k}'}(t)\,. \tag{3}$$

The value of ω_k^2 that results in the least residual interaction between phonons, *i.e.* the one for which $\partial\langle|\mathcal{F}_{\boldsymbol{k}}|^2\rangle/\partial\omega_k^2 = 0$, is [15,16]

$$\omega_k^2 = \frac{k^2}{\beta m S_k}\,. \tag{4}$$

This choice for ω_k^2 is also imposed by requiring the correct short-time limit of eq. (1) [17]. The minimization principle produces an orthogonality condition between the force and the density variables at all times [15],

$$\langle \rho_{-\boldsymbol{k}}(t)\mathcal{F}_{\boldsymbol{k}}(t)\rangle = 0\,. \tag{5}$$

Substituting the expression for $\mathcal{F}_{\boldsymbol{k}}(t)$ (eq. (3)) in this orthogonality condition leads to the exact Yvon-Born-Green (YBG) equation [18]

$$S_k = 1 - \frac{\beta}{Nk^2V}\sum_{\boldsymbol{k}'} v_{\boldsymbol{k}'}(\boldsymbol{k}\cdot\boldsymbol{k}')\langle\rho_{-\boldsymbol{k}}(t)\rho_{\boldsymbol{k}-\boldsymbol{k}'}(t)\rho_{\boldsymbol{k}'}(t)\rangle\,, \tag{6}$$

where $S_k \equiv \langle \rho_{-\boldsymbol{k}} \rho_{\boldsymbol{k}} \rangle / N$ is the static structure factor, $\beta \equiv 1/k_{\rm B}T$, $k_{\rm B}$ is the Boltzmann constant, T is the temperature and the symbols $\langle \cdots \rangle$ indicate equilibrium averages.

Following the spirit of the Zwanzig-Mori (ZM) formalism [19], the residual force can be written as the sum of two contributions: i) a term of dissipative origin, containing a memory function dependent on time; ii) a general random noise term. Hence,

$$\mathcal{F}_{\boldsymbol{k}}(t) = -\int_0^t \gamma_{\boldsymbol{k}}(t-t') \partial_{t'} \rho_{\boldsymbol{k}}(t') \mathrm{d}t' + f_{\boldsymbol{k}}(t)\,, \tag{7}$$

where $\gamma_{\boldsymbol{k}}(t)$ acts as memory function of the system and $f_{\boldsymbol{k}}(t)$ as fluctuating force. Note that the choice in eq. (7) for the residual force satisfies the orthogonality condition, eq. (5). The functions $f_{\boldsymbol{k}}(t)$ and $\gamma_{\boldsymbol{k}}(t)$ are not independent. Indeed, they must satisfy the fluctuation-dissipation theorem (FDT) to guarantee that the long-time evolution of the system is consistent with the correct Boltzmann equilibrium distribution. This requires that the autocorrelation function of $f_{\boldsymbol{k}}(t)$ is proportional to the memory function $\gamma_{\boldsymbol{k}}(t)$. Also, the average over the noise is zero. Thus, following [20]

$$\langle f_{\boldsymbol{k}}(t) \rangle = 0 \tag{8}$$

$$\frac{\langle f_{-\boldsymbol{k}}(t) f_{\boldsymbol{k}}(t') \rangle}{\langle |\dot{\rho}_{\boldsymbol{k}}|^2 \rangle} = \gamma_{\boldsymbol{k}}(t-t')\,. \tag{9}$$

As a result of all these formal assignments Newton's equations for the density fluctuations are rewritten in a form similar to a linear generalized Langevin equation as

$$\ddot{\rho}_{\boldsymbol{k}}(t) + \omega_k^2\ \rho_{\boldsymbol{k}}(t) + \int_0^t \gamma_{\boldsymbol{k}}(t-t') \partial_{t'} \rho_{\boldsymbol{k}}(t') \mathrm{d}t' = f_{\boldsymbol{k}}(t)\,. \tag{10}$$

Note that the transition from the Newton's equations to the generalized Langevin equation is associated with a transition from averages over the initial conditions to averages over the realization of the noise (defined in eqs. (8)-(10)). FDT guarantees that the dynamics generated by this stochastic process leads to thermodynamic equilibrium. Using eq. (9), we calculate the explicit exact form for the memory function [21] as

$$\begin{aligned}
\gamma_{\boldsymbol{k}}(t) \;=\; & \frac{\beta m}{N k^2} \left[\left(\frac{k^2}{\beta m} \right)^2 (n^2 c_k^2 - 1) N S_k(t) + \left\langle \sum_l \sum_m (\boldsymbol{k} \cdot \dot{\boldsymbol{r}}_l(0))^2 e^{-i\boldsymbol{k}\cdot\boldsymbol{r}_l(0)} (\boldsymbol{k} \cdot \dot{\boldsymbol{r}}_m(t))^2 e^{i\boldsymbol{k}\cdot\boldsymbol{r}_m(t)} \right\rangle + \right. \\
& + \frac{1}{(mV)^2} \sum_{\boldsymbol{k}'} \sum_{\boldsymbol{k}''} v_{k'} v_{k''} (\boldsymbol{k}\cdot\boldsymbol{k}')(-\boldsymbol{k}\cdot\boldsymbol{k}'') \langle \rho_{-\boldsymbol{k}-\boldsymbol{k}''}(0) \rho_{\boldsymbol{k}''}(0) \rho_{\boldsymbol{k}-\boldsymbol{k}'}(t) \rho_{\boldsymbol{k}'}(t) \rangle + \\
& + \frac{nk^2}{\beta m^2 V} c_k \sum_{\boldsymbol{k}'} v_{k'} (\boldsymbol{k}\cdot\boldsymbol{k}') \langle \rho_{-\boldsymbol{k}}(0) \rho_{\boldsymbol{k}-\boldsymbol{k}'}(t) \rho_{\boldsymbol{k}'}(t) \rangle + \\
& + \frac{nk^2}{\beta m^2 V} c_k \sum_{\boldsymbol{k}''} v_{k''} (-\boldsymbol{k}\cdot\boldsymbol{k}'') \langle \rho_{-\boldsymbol{k}-\boldsymbol{k}''}(0) \rho_{\boldsymbol{k}''}(0) \rho_{\boldsymbol{k}}(t) \rangle - \\
& - \frac{k^2}{\beta m} n N c_k \int_0^t \mathrm{d}t' \gamma_{\boldsymbol{k}}(t-t') \partial_{t'} S_k(t') - \\
& \left. - \frac{1}{mV} \sum_{\boldsymbol{k}'} v_{k'} (-\boldsymbol{k}\cdot\boldsymbol{k}') \int_0^t \mathrm{d}t' \gamma_{\boldsymbol{k}}(t-t') \langle \rho_{-\boldsymbol{k}-\boldsymbol{k}'}(0) \rho_{\boldsymbol{k}'}(0) \dot{\rho}_{\boldsymbol{k}}(t') \rangle \right].
\end{aligned} \tag{11}$$

Equations (8)-(11) constitute an exact definition of the noise process and have been obtained by using only FDT and the causality relation $\langle \rho_{-\boldsymbol{k}}(0) f_{\boldsymbol{k}}(t) \rangle = 0$. These equations have the same content as the equations derived in the ZM formalism [19]. To explicitly evaluate the $\gamma_{\boldsymbol{k}}(t)$ function, approximations have to be made on the stochastic processes $\rho_{\boldsymbol{k}}(t)$ and $f_{\boldsymbol{k}}(t)$, *i.e.* the density and the noise [22].

In this letter, a simple approximation is considered. We assume that $f_{\boldsymbol{k}}(t)$ is an additive Gaussian process. This is the major approximation, since the assumption of a Gaussian process and the linearity of eq. (10) imply that $\rho_{\boldsymbol{k}}(t)$ is also a Gaussian process. As shown in eqs. (3) and (7), the true $f_{\boldsymbol{k}}(t)$ is a non-linear function of $\rho_{\boldsymbol{k}}(t)$. This indicates that in an exact theory both quantities cannot be simultaneously Gaussian distributed [4].

A less significant approximation consists in averaging over the velocities neglecting the fluctuations in the single-particle kinetic energy, *i.e.* assuming

$$\left\langle \sum_l \sum_m (\boldsymbol{k}\cdot\dot{\boldsymbol{r}}_l(0))^2 e^{-i\boldsymbol{k}\cdot\boldsymbol{r}_l(0)} (\boldsymbol{k}\cdot\dot{\boldsymbol{r}}_m(t))^2 e^{i\boldsymbol{k}\cdot\boldsymbol{r}_m(t)} \right\rangle \approx \frac{k^4}{\beta^2 m^2} N S_k(t)\,. \tag{12}$$

The Gaussian nature of the noise ensures certain simplifications in the properties of $\rho_{\boldsymbol{k}}(t)$ and this allows to calculate the multiple averages in eq. (11). Together with the approximation in eq. (12), this allows to derive an expression for $\gamma_{\boldsymbol{k}}(t)$ which requires as input the density-density correlation functions. As a result, the Gaussian approximation in the density gives

$$\begin{aligned} \gamma_{\boldsymbol{k}}(t) \;=\; & \frac{n\beta}{mVk^2} \sum_{\boldsymbol{k}'\neq\boldsymbol{k}} \{ v_{k'}^2 (\boldsymbol{k}\cdot\boldsymbol{k}')^2 + v_{k'} v_{k-k'} (\boldsymbol{k}\cdot\boldsymbol{k}')(\boldsymbol{k}\cdot(\boldsymbol{k}-\boldsymbol{k}')) \} S_{|\boldsymbol{k}-\boldsymbol{k}'|}(t) S_{k'}(t) + \\ & + \frac{k^2 n^2}{\beta m} (c_k + \beta v_k)^2 S_k(t) - n(c_k+\beta v_k) \int_0^t \mathrm{d}t' \gamma_{\boldsymbol{k}}(t-t') \partial_{t'} S_k(t')\,. \end{aligned} \tag{13}$$

The previous expression still contains the Fourier transform of the pair potential v_k. As is well known [17, 18], once the Gaussian approximation for the density has been made, the YBG equation (eq. (6)) can be consistently solved [23], providing the so-called random phase approximation (RPA) [17, 24], $c_k = -\beta v_k$, between the direct correlation function $c_k \equiv (1 - 1/S(k))/n$ and the potential v_k. Eliminating in eq. (13) v_k in favor of c_k, one obtains

$$\gamma_{\boldsymbol{k}}(t)^{\mathrm{MCT}} = \frac{n}{\beta m k^2 V} \sum_{\boldsymbol{k}'\neq\boldsymbol{k}} \{ (\boldsymbol{k}\cdot\boldsymbol{k}')^2 c_{\boldsymbol{k}'}^2 + (\boldsymbol{k}\cdot\boldsymbol{k}')(\boldsymbol{k}\cdot(\boldsymbol{k}-\boldsymbol{k}')) c_{\boldsymbol{k}'} c_{\boldsymbol{k}-\boldsymbol{k}'} \} S(|\boldsymbol{k}-\boldsymbol{k}'|,t) S(k',t)\,. \tag{14}$$

$\gamma_{\boldsymbol{k}}(t)^{\mathrm{MCT}}$ coincides with the memory function that has been calculated originally in ref. [1] within the ideal MCT framework.

The reader may note that the choice of a Gaussian process for $\rho_{\boldsymbol{k}}(t)$, if used directly in eqs. (2)-(3), implies that the time evolution of the density-density correlation function is described by undamped harmonic modes [24, 25] with frequency ω_k. In this approximation there is no interaction between the modes and hence, strictly speaking, $\gamma_{\boldsymbol{k}}(t) = 0$. In the present derivation, the memory kernel is not zero, since the assumption that the noise is Gaussian is introduced only after the residual interactions are constrained to be a noise and dissipation and a formally correct expression for $\gamma_{\boldsymbol{k}}(t)$ (eq. (11)) is derived. This apparent contradiction is a consequence of the fact that the same approximations made before constraining phase

space to have the Mori-type properties are not equivalent to those made after this fundamental constraint is forced upon the system. Therefore, the division of the residual forces between density waves into a dissipative and noise term is fundamental. This step, whilst not always acceptable, is a conventional approach to dealing with complex many-body forces, and the fact that the integrable motions have been clearly removed from the interactions prior to this division is quite satisfying.

The present calculations show the following.

i) The single assumption of Gaussian properties for $\rho_{\boldsymbol{k}}(t)$ and $f_{\boldsymbol{k}}(t)$ allows one to derive MCT. All approximations used in the conventional MCT are exact in this limit. Hence, MCT can be seen as a minimum, as an exact theory in the RPA limit and a fully consistent mean-field approximation to the dynamics of a complex system. Thus, the MCT dynamics is in the same class of universality as mean-field dynamics. This possibility was suggested some time ago on the basis of the analogies between the equations describing the schematic MCT models and the dynamics of the order parameter in disordered p-spin models, solved under strict mean-field approximation [5, 26].

ii) The present approach offers a more direct interpretation to the uncontrolled approximation intrinsic in the conventional MCT. Indeed, as clearly stated in ref. [1], *a priori* estimations of the quality of the conventional MCT are not known. The derivation of the basic equations does not include any systematic expansion scheme. In the present approach, the more transparent approximations suggest that improvement over the MCT predictions is feasible. In particular, it is possible to eliminate v_k in eq. (11) by implementing higher-order approximations for the triplet correlation function. For example, it is possible to implement in the present scheme the Singwi-Sjölander (STLS) closure [27], which introduces controlled corrections to the Gaussian statistics. Work is in progress on this topic.

iii) In the RPA, the equation for the memory kernel, eq. (11), simplifies and the integral contributions cancel out. This simplification does not occur in general. It may be shown [28] that these terms provide a renormalization of the memory at infinite time. It can also be shown that these terms are dropped in the conventional MCT, when projecting onto the density pair subspace. In the present approach, these terms can be added to the conventional MCT result. This will provide a consistent expression for the short-time memory function.

iv) An interesting aspect of the Götze MCT derivation is that it does not enforce the consistency conditions between static and dynamics and therefore the input structure factors do not have to be RPA in origin (and indeed, "exact" direct correlation functions calculated from simulation data are often used as input in the theoretical calculations [11, 13]). In the conventional MCT approach, v_k disappears because of the Gaussian approximation in calculating the normalization matrix of the projection [1]. If consistency between statics and dynamics were enforced, the RPA would have followed, as discussed above. Moreover, Kawasaki [4] has recently presented a derivation of MCT based on the (quadratic) density functional Ramakrishnan-Yussouf free energy of a liquid, where the effective interaction between the density pairs is exactly $-c(k)/\beta$, suggesting that it is possible to correct the theory, without fundamentally affecting the universality class of its dynamics. Perhaps the answer to the intriguing question of why this is successful lies in formulating higher corrections to the YBG and memory kernel equations.

v) Finally, the present calculation opens a new route to MCT, and may provide an approach to develop an improved MCT description of the dynamics of deep supercooled liquids as well as extensions in order to deal with more complex systems, like molecules [13, 29–31] and polymers [32]. This is a strategy actively pursued, and it will be most interesting to see the outcome.

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