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Nonlinear response functions in an exponential trap model

Gregor Diezemann

Institut für Physikalische Chemie, Universität Mainz, Duesbergweg 10-14, 55128 Mainz, FRG, Germany

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ABSTRACT

The nonlinear response to an oscillating field is calculated for a kinetic trap model with an exponential density of states above the glass transition temperature T_0. For temperatures not too close to T_0, the results are similar to those obtained for the model with a Gaussian density of states. The choice of the dynamical variable that couples to the field and in particular its dependence on the trap energies generally has a strong impact on the shape of the dynamic response. The modulus of the frequency dependent third-order response either shows a peak or exhibits a monotonous decay from a finite low-frequency limit to a vanishing response at high frequencies depending on the dynamical variable. If a peak is observed, its height can show different temperature dependencies with the common feature of a scaling behavior near T_0. Additionally, in some but not all cases the static nonlinear susceptibility diverges at T_0. A recently proposed approximation that relates the cubic response to a four-time correlation function does not give reliable results due to a wrong estimate of the low-frequency limit of the response.

1. Introduction

There have been many attempts to understand the heterogeneous dynamics in supercooled liquids and glasses, cf. the reviews [1,2] and references therein. In particular, the development of experimental techniques to probe higher-order time correlation functions has played a pivotal role in the development of our understanding of the nature of the heterogeneities [3–8] and also the study of computer models allowed one to gain insight into the structural and dynamical properties of these correlation functions [9,10]. Also a length scale associated with the heterogeneities has been extracted from specially designed NMR experiments [11,12].

In addition to these approaches an additional way to extract a length scale from a special four-point correlation function $\chi_4(t)$ has been introduced and discussed in detail [13–16].

By relating the nonlinear (cubic) response $\chi_3(\omega, T)$ to a four-point correlation function, Bouchaud and Biroli showed how to extract a length scale or equivalently the number of correlated particles, N_{corr}, from measured nonlinear response functions [17]. The modulus of the cubic response function, $|\chi_3(\omega, T)|$, was found to exhibit a hump-like structure which is assumed to be a distinctive feature of glassy correlations [18,19]. It is found that the maximum of $|\chi_3(\omega, T)|$ decreases with increasing temperature and it is assumed to be proportional to N_{corr}. If glassy correlations are absent, a 'trivial' behavior is expected, i.e. a smooth decay of $|\chi_3(\omega, T)|$ as a function of frequency.

It should be mentioned that nonlinear dielectric experiments on supercooled liquids have also been interpreted in a slightly different way with a stronger emphasis on the heterogeneous nature of the dynamics [20–22].

A nonlinear response theory for Markov processes has been presented in Ref. [23], to be denoted as I in the following. The theory was applied to the model of dipole reorientations in an asymmetric double well potential (ADWP-model) [24,25]. For this model, $|\chi_3(\omega, T)|$ exhibits a trivial behavior except for a small temperature range in the vicinity of vanishing low-frequency limit $\chi_3(0, T)$ for finite asymmetry. In addition, model calculations were presented for the well-studied trap model with a Gaussian density of states [26–31] showing both, a peak or a trivial behavior, depending on the variable chosen and on temperature.

Furthermore, for some specific choice of the dynamical variable used to probe the dynamics, the peak-maximum increases as a function of temperature and for other choices it decreases. The results of the model calculations suggest that a direct relation between the cubic response function and some type of glassy correlations cannot be shown to exist in these mean-field models. Also other calculations employing specific models show a similar behavior, i.e. either the existence of a hump or a trivial decay [32,33].

In addition, in Ref. [34], denoted as II in what follows, I considered various four-time correlation functions and a particular approximation for the cubic response for the Gaussian trap model. According to the approximations employed by Bouchaud and Biroli [17], the most dominant contribution to the cubic response in the vicinity of a phase transition is related to a four-time correlation function. For the Gaussian trap model, it was found in II that the corresponding relation does not give a sound description of $|\chi_3(\omega, T)|$ due to a wrong estimate of the low-frequency behavior.

In the present paper, I will calculate the nonlinear response for the trap model with an exponential density of states (DOS) instead of a

E-mail address: diezeman@uni-mainz.de.

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Gaussian DOS. The most prominent difference between the two models is that the exponential DOS gives rise to a glass transition at a temperature T_0 below which the system cannot equilibrate. Thus, there is a critical point in this model and one can investigate the nonlinear response in the vicinity of this point. In the present paper, I will only consider the high temperature phase where equilibrium is always reached. In the next section, I will briefly recall the properties of the model and discuss the modifications in the linear response resulting from a specific choice of the dynamic variables. Section 3 is devoted to the discussion of the nonlinear response and the paper closes with some conclusions.

2. Trap model with an exponential density of states

In a simple picture the complex dynamics of a glassforming system can be described by collective transitions between inherent structures or meta-basins in the energy landscape of the system [28,30,35]. Neglecting the correlations among the various energy minima, a very simple stochastic dynamics can be defined in the following way [27]. The escape from a minimum or trap of energy ϵ_i takes place with a rate $\kappa(\epsilon_i)$ and the destination trap of the transition, determined by ϵ_f, is chosen at random, i.e. according to the density $\rho(\epsilon_f)$. Therefore, the conditional probability to find the system in the trap characterized by the trap energy ϵ at time t provided it occupied trap ϵ_0 at t_0, $G(\epsilon, t+t_0|\epsilon_0, t_0) = G(\epsilon, t|\epsilon_0, 0) \equiv G(\epsilon, t|\epsilon_0)$ obeys the following master equation (ME):

$$\dot{G}(\epsilon, t|\epsilon_0) = -\kappa(\epsilon)G(\epsilon, t|\epsilon_0) + \rho(\epsilon)\int d\epsilon' \kappa(\epsilon')G(\epsilon', t|\epsilon_0). \tag{1}$$

Here, the first term describes the loss of probability due to the escape from trap ϵ and the second term is responsible for the gain of probability due to all transitions from ϵ' to ϵ. A simple choice for the escape rate is provided by an Arrhenius-like dependence on the trap energy,

$$\kappa(\epsilon) = \kappa_\infty e^{-\beta\epsilon} \tag{2}$$

where a common activation energy is absorbed in the attempt frequency κ_∞. The model with an exponential density of states (DOS) is defined by [27]

$$\rho(\epsilon) = \beta x e^{-\beta x \epsilon} \quad \text{with} \quad x = T/T_0 \tag{3}$$

where $\beta = 1/T$ and the Boltzmann constant is set to unity. The motivation for this particular choice has its origin in the typical behavior of the tails of the energy distributions in models for disordered systems, in particular the random energy model, cf. Ref. [36]. The model so defined gives rise to non-exponential waiting time distributions and the relation of these to glassy dynamics has also been discussed earlier [37–39]. In Eq. (3), T_0 denotes the characteristic temperature of the model, below which a stationary distribution $p^{\mathrm{eq}}(\epsilon) = \lim_{t\to\infty} G(\epsilon, t|\epsilon_0)$ does not exist. The reason is that the normalization of the distribution is given by the integral $\int d\epsilon \rho(\epsilon) e^{\beta\epsilon}$ which diverges for $x < 1$ when $\rho(\epsilon)$ is given by Eq. (3). Above T_0, one has

$$p^{\mathrm{eq}}(\epsilon) = \beta(x-1)e^{-\beta(x-1)\epsilon} \tag{4}$$

and below T_0 the system ages for all times. The model exhibits a number of features that are reminiscent of what is observed in glassy systems above and below T_0. In particular, the aging dynamics below T_0 has been investigated in detail [27,40]. In the present paper, I will solely consider temperatures above T_0, i.e. $x > 1$, where equilibrium is always reached and aging is unimportant. The results can be compared with the model with a Gaussian DOS, $\rho(\epsilon) = \left(1/\sqrt{2\sigma}\right)\exp\left(-\epsilon^2/\left(2\sigma^2\right)\right)$, that does not show a glass transition but slow dynamics and non-exponential relaxation.

The two-time correlation function (2t-CF) of a variable $M(t)$ in general is given by:

$$\langle M(t)M(t_0)\rangle = \int d\epsilon \int d\epsilon_0 M(\epsilon)M(\epsilon_0)G(\epsilon, t-t_0|\epsilon_0)p^{\mathrm{eq}}(\epsilon_0). \tag{5}$$

The quantity $M(t)$ might for example represent a magnetization or a dipole moment. Generally, $M(t)$ is time-dependent due to its dependence on the trap energy, $M(t) = M(\epsilon(t))$ and the time-dependence of ϵ is governed by Eq. (1). Thus, one has to determine the ϵ-dependence of M. In a naive picture one could for instance assume that high energy regions correspond to low density regions and that the dipole moment varies with the latter. As in papers I and II [23,34], a Gaussian approximation for the correlations of the dynamical variables $M(\epsilon)$ will be used,

$$\langle M(\epsilon)\rangle = 0 \quad \text{and} \quad \langle M(\epsilon)M(\epsilon_0)\rangle = \delta(\epsilon-\epsilon_0)\left\langle M(\epsilon)^2\right\rangle. \tag{6}$$

For a fully connected trap model one has (changing to the common notation [27])

$$\Pi(t) = \int d\epsilon \left\langle M(\epsilon)^2\right\rangle p^{\mathrm{eq}}(\epsilon)e^{-\kappa(\epsilon)t}. \tag{7}$$

This function has a simple interpretation. Each transition out of the trap with energy ϵ completely decorrelates the variable and gives rise to a decay. For $\langle M(\epsilon)^2\rangle = 1$, it has been shown that the long-time behavior of $\Pi(t)$ is given by $\Pi(t) \sim t^{-(x-1)}$ [27].

Throughout the present paper, I will use an Arrhenius-like energy dependence of $\langle M(\epsilon)^2\rangle$ that first has been considered by Fielding and Sollich [40] in their treatment of the violations of the fluctuation dissipation theorem for the trap model and that I have used also in papers I and II [23,34]:

$$\left\langle M(\epsilon)^2\right\rangle = e^{-n\beta\epsilon} \tag{8}$$

where n is an arbitrary real constant. In particular, the choice $n = 0$ represents a variable that is independent of the trap energy. If M is viewed as a dipole moment, this might be interpreted as structure- or density-independent. If $n = 1$, a strong dependence of M on the trap energy and thus on the particular realization of the system is assumed. The particular form chosen is compatible with the result for an ADWP, where $\langle M^2\rangle \sim e^{-\beta\Delta}$ with Δ denoting the asymmetry. Additionally, it has been shown in Ref. [40] that weaker dependences give results equivalent to $n = 0$.

The linear susceptibility, which is the Fourier transform of $\Pi(t)$, is given by

$$\chi_1(\omega) = \beta\int_0^\infty d\epsilon \left\langle M(\epsilon)^2\right\rangle p^{\mathrm{eq}}(\epsilon)\frac{\kappa(\epsilon)}{\kappa(\epsilon)-i\omega}. \tag{9}$$

The resulting static susceptibility, i.e. the low-frequency limit, is

$$\Delta\chi_1 = \chi_1(0) = \beta\frac{x-1}{x-1+n} \tag{10}$$

which diverges at a temperature $T = (1-n)T_0$. Furthermore, the integral relaxation time

$$\tau_{\mathrm{eq}}^{(n)} = \int_0^\infty dt \Pi(t) = \frac{1}{\kappa_\infty}\frac{x-1}{x-2+n} \tag{11}$$

diverges at $T = (2-n)T_0$ which reduces to the well-known result $2T_0$ for $n = 0$ [27]. In Fig. 1a), $\Pi(t)$ is shown for various values of n.

It is obvious that the asymptotic behavior is given by $\Pi(t) \sim t^{-(x-1+n)}$. This means that for the model with an exponential DOS the choice of the dynamical variables used here has a strong impact on the temperature dependent dynamical properties as has been discussed earlier [40]. In addition, the relaxation time τ that is