Frequency-domain study of $\alpha$-relaxation in the Random Orthogonal Model

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printout: 15.01.2003

Abstract

The time-dependent susceptibility for the finite-size mean-field Random Orthogonal model (ROM) is studied numerically for temperatures above the mode-coupling temperature. The results show that the imaginary part of the susceptibility $\chi''(\nu)$ obeys the scaling form proposed for glass-forming liquids with the peak frequency decreasing as the temperature is lowered consistently with the Vogel-Fulcher law with a critical temperature remarkably close to the known critical temperature $T_c$ of the model where the configurational entropy vanishes.

PACS: 64.40.-i, 64.60.Cn, 75.10.Nr

The spectral properties of the primary or $\alpha$-relaxation in supercooled liquids has been largely studied by means of dielectric spectroscopy (Dixon et al. 1990, Chamberlin 1991, Dixon et al. 1991, Schönhals et al. 1991, Menon and Nagel 1993, Schönhals et al. 1993, Kuldlik et al. 1995, Leheny et al. 1996, Leheny and Nagel 1997) finding that the data for the imaginary part $\epsilon''(\nu)$ of the dielectric susceptibility $\epsilon(\nu)$ at different temperatures and for several glass-forming liquids can be collapsed onto a master curve using
a three-parameter scaling function. The master plot is able to reproduce the $\varepsilon''(\nu)$ data around the relaxation peak $\nu_p$ and also at higher frequencies. The bad collapse in the low frequency part has been object of some debates (Schönhals et al. 1991, Menon and Nagel 93, Schönhals et al. 1991, Kuldlik et al. 1995, Leheny et al. 1996), however there is no dispute above $\nu_p$. The frequency $\nu_p$ has a very strong temperature dependence, commonly fitted by a Vogel-Fulcher form $\log_{10}(\nu_p) = \log_{10}(\nu_0) - A/(T - T_0)$, where $T_0$ is close to the Kauzmann temperature (Kauzmann 1948) where the configurational entropy vanishes (see e.g., Angell 1988).

In this contribution we compare the frequency-domain analysis of the finite-size Random Orthogonal Model (ROM) above the mode coupling temperature with the above scenario. The main motivation for this study was to make a stringent test on the ROM as a possible toy-model for the fragile-glass scenario. Model Hamiltonians capable of describing relaxation processes in supercooled liquids and structural glasses are difficult to obtain. However, starting with the work of Kirkpatrick, Thirumalai and Wolynes (Kirkpatrick and Thirumalai 1987a, b, Kirkpatrick and Wolynes 1987) in the late 80’s, it is now clear that there is a close analogy between some mean-field spin-glass models and structural glasses (Bouchaud et al. 1998). The basic simplification occurring in mean-field models is that in the limit of a very large ($N \rightarrow \infty$) number of spin one is left with a closed set of equations for the two-time correlation and response functions which, above a critical temperature $T_D$, are equivalent to the schematic mode coupling equations introduced by Leutheusser, Götze and others (Bengtzelius et al. 1984, Leutheusser 1984, Götze 1991) as a model for the ideal glass transition.

In mean-field models the barrier separating different ergodic components diverges in the mean-field limit, hence at the critical temperature $T_D$ a real ergodic to non-ergodic transition takes place with diverging relaxation times. The critical temperature $T_D$ coincides with the critical temperature $T_{MCT}$ derived in the Mode-Coupling Theory (MCT) and in what follows we will use only the notation $T_{MCT}$. In a real system, however, barriers are of finite height and the glass transition appears at $T_g < T_{MCT}$ where the typical activation time over barriers is of the same order of the observation time. In these systems $T_{MCT}$ represents the temperature below which the dynamics is dominated by activated hopping over energy barriers. Therefore to go beyond mean-field it is necessary to include activated processes, a very difficult task since it implies the knowledge of excitations involved in the dynamics. Recent studies have shown that activated processes in mean-field models could be included just keeping $N$ finite (Crisanti and Ritort 2000a, b), giving support to the scenario of the fragile glass transition developed from spin-glass models. This is not a trivial assumption since it is not a priori clear why excitations in mean-field spin glass models should have similar properties to those of supercooled liquids. This, for example, seems to be the case for the Random Orthogonal model (ROM) (Marinari et al.
1994), but not for the mean-field Potts-Glass model (Bragian et al. 2001, Bragian et al. 2002).

To compare the results from the finite-size ROM with the experimental one (Dixon et al. 1990, Chamberlin 1991, Schönhals et al. 1991, Dixon et al. 1991, Menon and Nagel 1993, Schönhals et al. 1993, Kuldlik et al. 1995, Menon and Nagel 1995, Leheny et al. 1996, Leheny and Nagel 1997) in this contribution we shall consider only temperatures above $T_{MCT}$. Since the range of temperatures we explore are all above the mode-coupling transition $T_{MCT}$, we do not expect to find diverging timescales in the large $N$ limit.

The ROM (Marinari et al. 1994) is defined by the Hamiltonian

$$H = -2 \sum_{ij} J_{ij} \sigma_i \sigma_j - h \sum_i \sigma_i$$

where $\sigma_i = \pm 1$ are $N$ Ising spin variables, and $J_{ij}$ is a $N \times N$ random symmetric orthogonal matrix with $J_{ii} = 0$. For $N \to \infty$ and $h = 0$ this model has a dynamical transition at $T_{MCT} = 0.536$, and a static transition at $T_c = 0.256...$ (Marinari et al. 1994). Numerical simulations are performed using the Monte Carlo (MC) method with the Glauber algorithm for temperatures in the range $0.6$ up to $2.0$. To study the frequency response we considered a time-dependent field of the form $h(t) = h_0 \cos(2 \pi \nu t)$, where the time is measured in MC steps and $h_0 = 0.2$ small enough to be within the linear response regime. In our simulations the typical range of $\nu$ was $10^{-6} - 10^{-1}$. For each frequency $\nu$ the complex susceptibility $\chi(\nu) = \chi'(\nu) + i\chi''(\nu)$ is given by

$$\chi'(\nu) = \frac{1}{NM} \sum_{t=1}^{M} \sum_{j=1}^{N} \sigma_j(t) \cos(2 \pi \nu t),$$

$$\chi''(\nu) = \frac{1}{NM} \sum_{t=1}^{M} \sum_{j=1}^{N} \sigma_j(t) \sin(2 \pi \nu t).$$

The number of MC steps $M$ after equilibration was 100 for the largest $\nu$ and up to $10^7$ for the shortest $\nu$. As system size we used $N = 300$ which is a good compromise between small sample-to-sample fluctuations and small barriers height.

Figure 1 shows the real and imaginary parts of the susceptibility over the available range of frequency. Not all temperatures are reported for a better drawing. The relaxation peak in the imaginary part can be fitted with a log-normal form (Wu and Nagel 1992):

$$\chi''(\nu) = \frac{\Delta \chi}{\sqrt{\pi \Sigma}} \exp \left[ -\left( \log_{10} \nu - \log_{10} \nu_p \right)^2 / \Sigma^2 \right]$$

where $\nu_p$ is the frequency of the peak, $\Sigma$ the width, and $\Delta \chi = \chi'_0 - \chi'_\infty$, where $\chi'_0$ and $\chi'_\infty$ are, respectively, the low and high frequency limit of $\chi'(\nu)$. 3
As the temperature is lowered the peak frequency $\nu_p$ decreases, and the width $\Sigma$ broadens. The behavior of $\nu_p$ is consistent with the Vogel-Fulcher law $\exp[-A/(T - T_0)]$ (Menon and Nagel 1995). The fit of the frequency peak $\nu_p$ for the ROM with the Vogel-Fulcher formula is rather good, see Fig. 2 and gives $A = 0.89 \pm 0.06$ and $\ln \nu_0 = 0.64 \pm 0.02$ $T_0 = 0.28 \pm 0.02$, a value in agreement with the critical value $T_c = 0.256...$. We note, however, that data can also be fitted using different expressions such as $\exp[-A/T^2]$ or the Adam-Gibbs formula $\exp[-A/(TS_c(T))]$ where $S_c$ is the configurational entropy, and in particular with the formula (CR) $\nu_p = \nu_0 \exp[-A\beta_{\text{eff}}(T)/T]$ where $\beta_{\text{eff}}(T) = \partial S_c(e_{\text{is}})/\partial e_{\text{is}}|_{e_{\text{is}} = e_{\text{is}}(T)}$ derived from a cooperative scenario of relaxation (Crisanti and Ritort 2002), see Fig. 2. The CR formula predicts a crossover form fragile to strong behaviors as the temperature is lowered, however, differences among all these expressions can be appreciated only for very low values of $\nu_p$ which are out of our measurements range.

The analysis of the response for glass-former liquids reveals three power
Figure 2: $\log_{10} \nu_p$ as function of $T$ for the ROM with $N = 300$. The full line is the Vogel-Fulcher law $\nu_p = \nu_0 \exp[-A/(T - T_0)]$ while the dashed line is the formula $\nu_p = \nu_0 \exp[-A\beta_{\text{eff}}(T)/T]$ of Ref. (Crisanti and Ritort 2002) with $\beta_{\text{eff}}(T) = \partial S_c(e_{\text{is}})/\partial e_{\text{is}}|_{e_{\text{is}}=e_{\text{is}}(T)}$ evaluated using the results of Refs. (Crisanti and Ritort 2000a, b). The discrepancy at high temperature is probably due to a poor numerical estimation of the configurational entropy, indeed a similar deviation is found using the Adam-Gibbs formula (not reported).

laws for $\chi''$ (Leheny and Nagel 1997):

$$\chi''(\nu) \sim \begin{cases} 
\nu^m & \nu < \nu_p \\
\nu^{-\beta} & \nu > \nu_p \\
\nu^{-\sigma} & \nu \gg \nu_p
\end{cases}$$

(5)

The discrete nature of Monte Carlo dynamics time step prevents us from resolving the last, nevertheless the first two regimes are clearly seen, as shown in Figure 3. At higher temperatures $m = \beta = 1$ and the relaxation is Debye-like with exponential decaying correlations. As the temperature is lowered the value of $\beta$ decreases below 1 and decay becomes stretched-exponential. It is known that for glass-former liquids $\beta$ and $\sigma$ are related by $(\sigma + 1)/(\beta + 1) = \gamma$, where $\gamma$ is a constant (Leheny and Nagel 1997). Furthermore $\sigma$ varies linearly with temperature: $\sigma = B(T - T_0)$ with $T_0 \simeq T_0$ (Leheny and Nagel 1997). This implies that $\beta = B'(T - T_0) + (1 - \gamma)/\gamma$. Inserting into this formula the values of $\beta$ obtained for the ROM at various temperatures and the value of $T_0$ computed from $\nu_p$ we find $\gamma = 0.72 \pm 0.02$ the same value found for real liquids (Menon and Nagel 1995, Leheny and Nagel 1997).

The analyticity of $\chi(\nu)$ and linearity of absorption at asymptotically low frequencies implies that $\chi''(\nu) \propto \nu$ for $\nu \ll \nu_p$ (Schönhals et al. 1991, Menon and Nagel 1993, Schönhals et al. 1993). For the ROM with $N = 300$ we find $m \simeq 1$ for temperatures down to about $T = 0.8$ while below significant
deviations with $m < 1$ are observed. Similar deviations have been observed in data from glass-forming liquids and generated some controversy (Dixon et al. 1990, Chamberlin 1991, Schönhals et al. 1991, Dixon et al. 1991, Menon and Nagel 1993, Schönhals et al. 1993, Kuldlik et al. 1995, Leheny et al. 1996) on the reliability of the scaling form proposed by Dixon et al. (Dixon et al. 1990, Chamberlin 1991, Dixon et al. 1991). Many liquids possess secondary relaxations which overlap the primary response broadening the peak and leading to deviation from linearity (Kuldlik et al. 1995, Leheny et al. 1996). In the case of ROM these secondary relaxations are related to the fact that the barriers separating the low states sampled as the temperature is decreased toward $T_{MCT}$ are not well separated for not too large $N$. Indeed studies of mean-field spin-glass models for the structural glass transition shows that in the thermodynamic limit there is no gap between saddles separating local minima with energy above the threshold energy associated with the dynamical transition (Cavagna et al. 1997). This is a situation more reminiscent of spin-glasses rather than glasses for which both experimental (Bitko et al. 1996) and numerical simulations (Bitko et al. 1996, Rao 2001) show a broader shape of $\chi''(\nu)$ near the peak.

This scenario is supported by a finite-size scaling analysis of the ROM. Indeed we find that for a fixed temperature while $\beta$ is independent on $N$, the value of $m$, when less than 1, increases toward 1 as $N$ is increased.

**Figure 3:** $\chi''(\nu)$ for the ROM with $N = 300$ for temperatures $T = 0.7$ and $1.1$. The exponents are $m = 1, \beta = 1.$ for $T = 1.1$ and $m = .96, \beta = .71$ for $T = 0.7$. Inset: $\chi''(\nu)$ at temperature $T = 0.6$ for $N = 64$ (filled circles) and $300$ (empty circles). The lines have slope $m = 0.3$ for $N = 64$ and $m = 0.5$ for $N = 300$. The increase of $m$ toward 1 as $N$ grows is clearly seen.
In the inset of Figure 3 we show $\chi''(\nu)$ at $T = 0.6$ for systems of size $N = 64$ and $N = 300$. The increase of $m$ going from 64 to 300 spins is clear. Finally we address the goodness of the Nagel scaling. Dixon et al. (Dixon et al. 1990, Chamberlin 1991, Dixon et al. 1991) have shown that all data for the dielectric susceptibility $\epsilon(\nu)$ of different glass-forming liquids and temperatures can be collapsed onto a single master curve by plotting $(1/w)\log_{10}(\epsilon''(\nu)\nu_p/\nu \Delta\epsilon)$ versus $(1/w)(1/w + 1)\log_{10}(\nu/\nu_p)$, where $\Delta\epsilon = \epsilon'(0) - \epsilon'(\infty)$, $\nu_p$ is the peak frequency and $w$ is the half-maximum width of the $\epsilon''(\nu)$ peak normalized to the corresponding width of the Debye peak: $w_D \approx 1.14$ decades.

Figure 4: The Nagel's scaling for the ROM with $N = 300$ and temperatures $T = 2.0, 1.3, 1.2, 1.1, 1.0, 0.9, 0.8, 0.7$ and 0.6.

Figure 4 shows the Nagel plot for the ROM with $N = 300$. The data are from temperatures ranging from 0.6 up to 2.0. For each curve the parameters $\Delta\chi$, $w = 2\sqrt{2\Sigma}/w_d$ and $\nu_p$ have been obtained from the log-normal fit of $\chi''$ using (4). We see that while the collapse for $\nu > \nu_p$ is good for all temperatures, for $\nu < \nu_p$ only data with $m = 1$ do collapse. As noted in Refs. (Kuldlik et al. 1995, Leheny et al. 1996) the optimization of the three parameters through the fitting is essential to have a good collapse of data.

In conclusion, we have shown that the primary relaxation in the finite size mean-field ROM obeys the scaling form typical of glass-forming liquids. Furthermore, the frequency peak of the imaginary part of the complex susceptibility follows the Vogel-Fulcher law with critical temperature $T_0 = 0.28 \pm 0.02$ very close to the critical temperature $T_c = 0.256...$, the Kauzmann temperature of the model. All system sizes studied (up to $N = 300$) lead to this value for $T_0$. Because we used Monte Carlo dynamics there is a maximum value for the frequency $\sim N$ determined by the discreteness of the elementary time step. As a consequence we are not able to resolve the second
scaling behaviors of the imaginary part of the susceptibility \( \chi''(\nu) \sim \nu^{-\sigma} \) for \( \nu \gg \nu_p \). Nevertheless by assuming that the exponent \( \sigma \) vanishes linearly at \( T_0 \), we obtained for the constant \( \gamma \) relating the exponents \( \beta \) and \( \sigma \) the same value \( \gamma = 0.72 \pm 0.02 \), found for real glass-forming liquids (Menon and Nagel 1995, Leheny and Nagel 1997). This value does not depend on the system size for the sizes we studied. Overall, the present results show that finite-size mean-field spin glasses capture the cooperative effects responsible for the relaxational processes observed in glass forming liquids when approaching the mode-coupling temperature from above. The extension of this analysis to the region below \( T_{MCT} \) where strong finite \( N \) effects are to be observed remains an interesting open problem.

We acknowledge F. Sciortino and P.Tartaglia for a critical reading of the manuscript. A.C. acknowledges support from the INFM-SMC center. F.R has been supported by project the Spanish Ministerio de Ciencia y Tecnología Grant number BFM2001-3525 and Generalitat de Catalunya. A.C and F.R have also benefited from the Acciones Integradas España-Italia HI2000-0087.

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