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Frequency-domain study of α relaxation in the random orthogonal model

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ABSTRACT

The time-dependent susceptibility for the finite-size mean-field random orthogonal model is studied numerically for temperatures above the mode-coupling temperature. The results show that the imaginary part $\chi''(\nu)$ of the susceptibility obeys the scaling form proposed for glass-forming liquids with the peak frequency decreasesing 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.

The spectral properties of the primary or α relaxation in supercooled liquids has been largely studied by means of dielectric spectroscopy (Dixon *et al.* 1990, 1991, Chamberlin 1991, Schönhals *et al.* 1991, 1993, Menon and Nagel 1993, Kudlik *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 on to a master curve using a three-parameter scaling function. The master plot is able to reproduce the $\epsilon''(\nu)$ data around the relaxation peak ν_p and also at higher frequencies. The marked collapse in the low-frequency part has been the object of some debates (Schönhals *et al.* 1991, 1993, Menon and Nagel 1993, Kudlik *et al.* 1995, Leheny *et al.* 1996); however, there is no dispute about the data above ν_p. The frequency ν_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 (1948) temperature where the configurational entropy vanishes (for example 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

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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 and Thirumalai (1987a, b) and Kirkpatrick and Wolynes (1987) in the late 1980s, 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 ($N \to \infty$) number of spins, 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 Bengtzelius *et al.* (1984), Leutheusser (1984) and 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 and in what follows we shall use only the notation T_{MCT}. In a real system, however, barriers are of finite height and the glass transition appears at $T_{\mathrm{g}} < T_{\mathrm{MCT}}$ 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 are dominated by activated hopping over energy barries. Therefore to go beyond the mean-field model it is necessary to include activated processes, a very difficult task since it implies knowledge of the 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 ROM (Marinari *et al.* 1994), but not for the mean-field Potts–Glass model (Brangian *et al.* 2001, 2002).

To compare the results from the finite-size ROM with the experimental data (Dixon *et al.* 1990, 1991, Chamberlin 1991, Schönhals *et al.* 1991, 1993, Menon and Nagel 1993, 1995, Kudlik *et al.* 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 that we explore is above the mode-coupling transition temperature T_{MCT}, we do not expect to find diverging time scales in the large-N limit.

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

$$H = -2 \sum_{ij} J_{ij} \sigma_i \sigma_j - h \sum_i \sigma_i, \tag{1}$$

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 dynamic transition at $T_{\mathrm{MCT}} = 0.536$, and a static transition at $T_{\mathrm{c}} = 0.256\ldots$ (Marinari *et al.* 1994). Numerical simulations are performed using the Monte Carlo method with the Glauber algorithm for temperatures in the range from 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 Monte Carlo steps and $h_0 = 0.2$ small enough to be within the linear response regime. In our simulations the typical range

of ν was 10^{-6}–10^{-1}. For each frequency ν the complex susceptibility $\chi(\nu) = \chi'(\nu) + i\chi''(\nu)$ is given by

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

$$\chi''(\nu) = \frac{1}{NM} \sum_{t=1}^{M} \sum_{j=1}^{N} \sigma_j(t) \sin(2\pi\nu t). \tag{3}$$

The number M of Monte Carlo steps after equilibration was 100 for the largest ν and up to 10^7 for the shortest ν. As system size we used $N = 300$ which is a good compromise between small sample-to-sample fluctuations and small barrier heights.

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

$$\chi''(\nu) = \frac{\Delta\chi}{\pi^{1/2}\Sigma} \exp\left(\frac{-(\log_{10}\nu - \log_{10}\nu_p)^2}{\Sigma^2}\right), \tag{4}$$

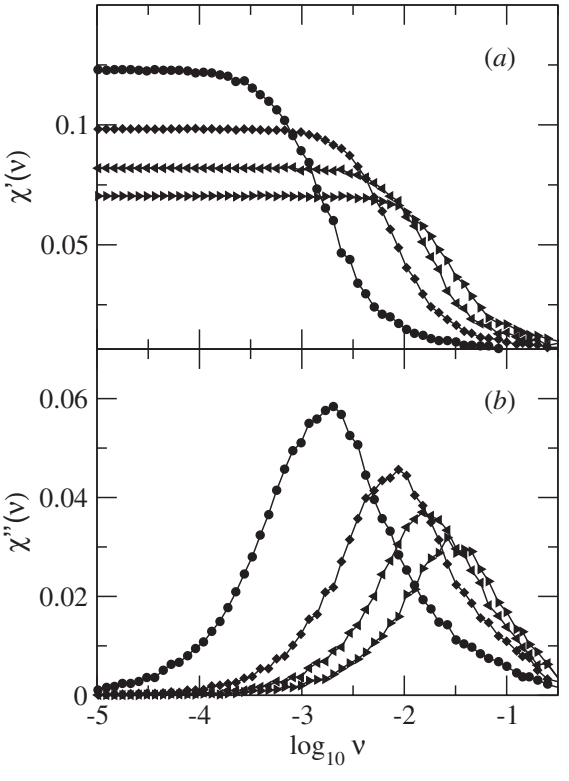

Figure 1. (*a*) The real part χ' of the complex susceptibility and (*b*) the imaginary χ'' of the complex susceptibility as functions of ν for the ROM with $N = 300$ at different temperatures. (*a*) top to bottom, (*b*) left to right, $T = 0.7$, 0.9, 1.1 and 1.3.

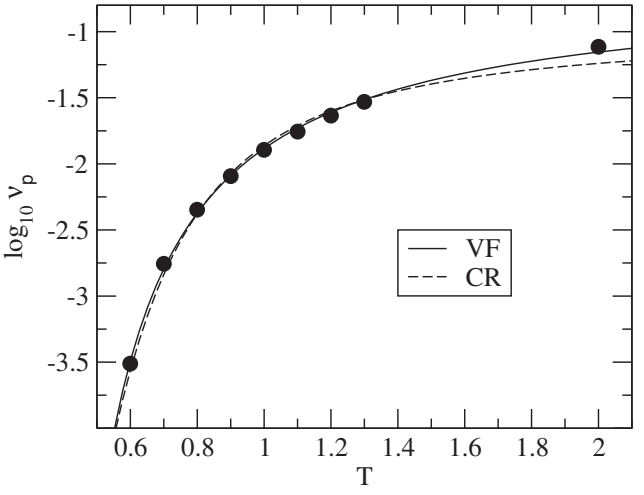

Figure 2. $\log_{10} \nu_p$ as a function of T for the ROM with $N = 300$: (—), is the Vogel–Fulcher law $\nu_p = \nu_0 \exp[-A/(T - T_0)]$; (- - -), the Crisanti–Ritort (2002) formula $\nu_p = \nu_0 \exp[-A\beta_{\text{eff}}(T)/T]$ with $\beta_{\text{eff}}(T) = \partial S_c(e_{is})/\partial e_{is}|_{e_{is}=e_{is}(T)}$ evaluated using the results of Crisanti and Ritort (2000a, b). The discrepancy at high temperatures 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).

where ν_p is the frequency of the peak, Σ is the width and $\Delta\chi = \chi'_0 - \chi'_\infty$, where χ'_0 and χ'_∞ are the low- and high-frequency limits respectively of $\chi'(\nu)$.

As the temperature is lowered, the peak frequency ν_p decreases, and the width Σ broadens. The behaviour of ν_p is consistent with the Vogel–Fulcher law $\exp[-A/(T - T_0)]$ (Menon and Nagel 1995). The fit of the frequency peak ν_p for the ROM with the Vogel–Fulcher formula is rather good (figure 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 $T_c = 0.256\ldots$. 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 $\nu_p = \nu_0 \exp[-A\beta_{\text{eff}}(T)/T]$ where $\beta_{\text{eff}}(T) = \partial S_c(e_{is})/\partial e_{is}|_{e_{is}=e_{is}(T)}$ derived from a cooperative scenario of relaxation (Crisanti and Ritort 2002) (see figure 2). This Crisanti–Ritort formula predicts a crossover form fragile behaviour to strong behaviour as the temperature is lowered; however, differences between all these expressions can be appreciated only for very low values of ν_p which are out of our measurement range.

The analysis of the response for glass-former liquids reveals three power laws for χ'' (Leheny and Nagel 1997):

$$\chi''(\nu) \propto \begin{cases} \nu^m, & \nu < \nu_p, \\ \nu^{-\beta}, & \nu > \nu_p, \\ \nu^{-\sigma}, & \nu \gg \nu_p. \end{cases} \tag{5}$$

The discrete nature of the Monte Carlo dynamics time step prevents us from resolving the last regime; 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,

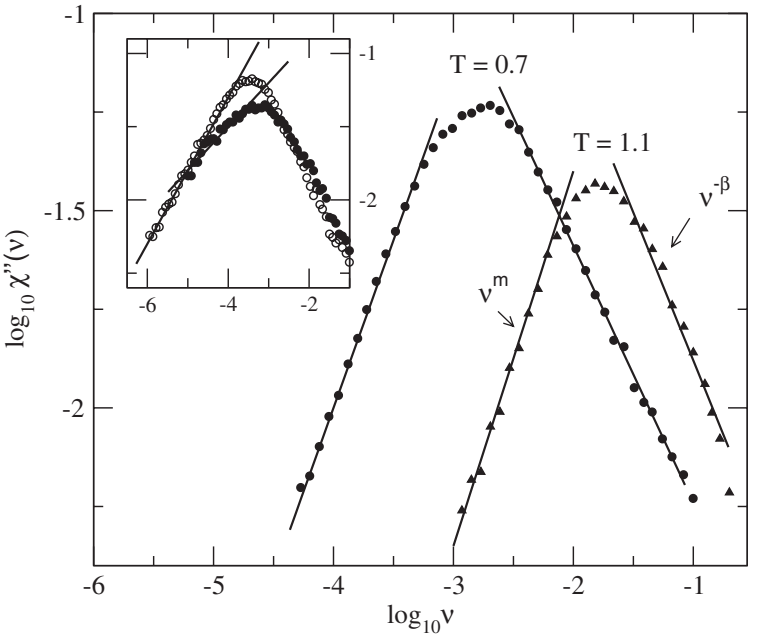

Figure 3. $\chi''(\nu)$ for the ROM with $N = 300$ for temperatures $T = 0.7$ and 1.1. The exponents are $m = 1$ and $\beta = 1$ for $T = 1.1$ and $m = 0.96$ and $\beta = 0.71$ for $T = 0.7$. The inset shows $\chi''(\nu)$ at temperature $T = 0.6$ for $N = 64$ (●) and 300 (○). The lines have slope $m = 0.3$ for $N = 64$ and $m = 0.5$ for $N = 300$. The increase of m towards 1 as N grows is clearly seen.

the value of β decreases below 1 and the decay becomes a stretched exponential. It is known that, for glass-former liquids, β and σ are related by $(\sigma + 1)/(\beta + 1) = \gamma$, where γ is a constant (Leheny and Nagel 1997). Furthermore σ varies linearly with temperature: $\sigma = B(T - T_\sigma)$ with $T_\sigma \approx T_0$ (Leheny and Nagel 1997). This implies that $\beta = B'(T - T_0) + (1 - \gamma)/\gamma$. Inserting into this formula the values of β obtained for the ROM at various temperatures and the value of T_0 computed from ν_p we find that $\gamma = 0.72 \pm 0.02$, 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, 1993, Menon and Nagel 1993). For the ROM with $N = 300$ we find that $m \approx 1$ for temperatures down to about $T = 0.8$ while, below this, significant deviations with $m < 1$ are observed. Similar deviations have been observed in data from glass-forming liquids and have generated some controversy (Dixon *et al.* 1990, 1991, Chamberlin 1991, Schönhals *et al.* 1991, 1993, Menon and Nagel 1993, Kudlik *et al.* 1995, Leheny *et al.* 1996) on the reliability of the scaling form proposed by Dixon *et al.* (1990, 1991) and Chamberlin (1991). Many liquids posses secondary relaxations which overlap the primary response, broadening the peak and leading to deviation from linearity (Kudlik *et al.* 1995, Leheny *et al.* 1996). In the case of the ROM these secondary relaxations are related to the fact that the barriers separating the low states sampled as the temperature is decreased towards T_{MCT} are not well separated for not too large N. Indeed studies of mean-field spin-glass models for the structural glass transition show that in the thermodynamic limit there is no gap between saddle points

separating local minima with energy above the threshold energy associated with the dynamic transition (Cavagna *et al.* 1997). This is a situation more reminiscent of spin glasses rather than glasses for which both experimental data (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 β is independent of N, the value of m, when less than 1, increases towards 1 as N is increased. 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 in m on going from 64 to 300 spins is clear. Finally we address the goodness of the Nagel scaling. Dixon *et al.* (1990, 1991) and Chamberlin (1991) have shown that all data for the dielectric susceptibility $\epsilon(\nu)$ of different glass-forming liquids and temperatures can be collapsed on to 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 $\Delta\epsilon = \epsilon'(0) - \epsilon'(\infty)$, ν_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 shows the Nagel plot for the ROM with $N = 300$. The data are for temperatures ranging from 0.6 up to 2.0. For each curve the parameters $\Delta\chi$, $w = 2^{3/2}\Sigma/w_D$ and ν_p have been obtained from the log-normal fit of χ'' using equation (4). We see that, while the collapse for $\nu > \nu_p$ is good for all temperatures, for $\nu < \nu_p$ only data with $m = 1$ collapse. As noted by Kudlik *et al.* (1995) and Leheny *et al.* (1996) the optimization of the three parameters through the fitting is essential to obtain 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 $T_c = 0.256\ldots$, the Kauzmann temperature of the model. All system sizes studied (up to $N = 300$) lead to this value for T_0. Because we used

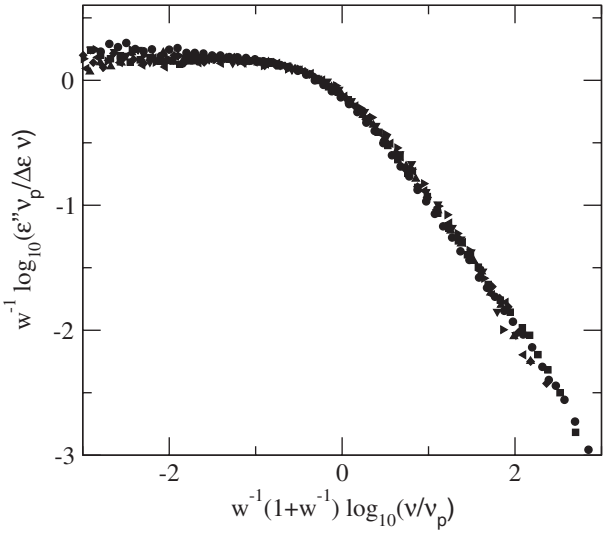

Figure 4. The Nagel scaling for the ROM with $N = 300$ and temperatures $T = 2.0$, 1.3, 1.2, 1.1, 1.0, 0.9, 0.8, 0.7 and 0.6.

Monte Carlo dynamics there is a maximum value for the frequency of about N determined by the discreteness of the elementary time step. As a consequence we are not able to resolve the second scaling behaviours of the imaginary part of the susceptibility $\chi''(\nu) \propto \nu^{-\sigma}$ for $\nu \gg \nu_p$. Nevertheless by assuming that the exponent σ vanishes linearly at T_0, we obtained for the constant γ relating the exponents β and σ 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.

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