Dynamical Heterogeneities close to the glass transition probed through nonlinear responses.

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In the last 20 years, it was established [1] that relaxation in supercooled liquids occurs via groups of dynamically correlated molecules called Dynamical Heterogeneities (DH). These groups of typically N_{corr} molecules are extremely difficult to evidence both because N_{corr} is not macroscopic, and because their lifetime is finite. However, several experimental breakthroughs [1,2] have unveiled DH's. In particular, since 1996, Non Resonant Hole Burning experiments (NRHB) have convincingly shown [2] that the dynamics is heterogeneous at least in time : the various DH's do not have the same relaxation time τ, there is instead a whole distribution of τ around the typical value τ_α. In brief, NRHB experiments measure the nonlinear response produced by applying a strong field during a short time.

In this poster, we argue that nonlinear experiments can give more than what was originally expected in the late nineties, since they can yield informations about N_{corr} itself and not only reveal the existence of a distribution of τ's. The interpretation of nonlinear experiments was indeed renewed in 2005 by two of us [3], who predicted that the cubic nonlinear response χ_3 is directly proportional to N_{corr}. The core of the argument is that χ_3 is related, through a Fluctuation Dissipation relation, to the four point correlation function which had just been recognised as being the suitable correlation function capturing the subtle correlations constituting the DH's. We shall review the results [4,5,6,7] that we have obtained on glycerol by using a specially designed experimental setup where χ_3 can be accurately measured. Our results were obtained close to the glass transition temperature T_g. We shall show results for two cubic nonlinear susceptibilities, namely $\chi_3^{(1)}$ (first harmonics) and $\chi_3^{(3)}$ (third harmonics). We shall see that :

• the modulii $|\chi_3^{(1or3)}|$ have a humped shape [4,5], and reach their maximum for an angular frequency of the order of $1/\tau_\alpha$. This shape had been predicted in [3], and is at odds of the monotonous behavior of the modulus $|\chi_{lin}|$ of the linear susceptibility.

• At equilibrium above the glass transition temperature, $\chi_3^{(1)}$ and $\chi_3^{(3)}$ have the same temperature dependance [5], which we interpret as being directly that of $N_{corr}(T)$.

• In the aging regime, where observables depend on the age t_a elapsed since the glass transition temperature T_g was crossed, one can extract the age dependance of N_{corr} from that of $|\chi_3^{(3)}|$. This yields the first direct observation [6] that N_{corr} increases with t_a, before reaching its equilibrium value when aging is over. By putting together our results above and below T_g, we follow the increase of N_{corr} with τ_α, when τ_α increases by 8 decades. This allows a stringent test of the theoretical approaches of the glass transition, such as RFOT.

Finally, we shall evoke a phenomenological toy model [7] that we built recently : it accounts fairly well for both the modulii and for the phases of $\chi_3^{(1or3)}$ with reasonable values of N_{corr}.

Références

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