Comment on "Models of Hierarchically Constrained Dynamics for Glassy Relaxation"

Palmer, Stein, Abrahams, and Anderson[1] (PSAA) have described some dynamical models for glassy relaxation. They were concerned in particular with "stretched exponential" decay of the form

$$q(t)=q(0)\exp[-(t/\tau)^{\beta}], \quad 0<\beta<1. \tag{1}$$

They proposed as a decay function the infinite series

$$q(t)=\sum_{n=0}^{\infty} w_n \exp(-t/\tau_n), \tag{2}$$

where the weights w_n and relaxation times τ_n are given below in Eq. (3). On converting the sum in Eq. (2) to an integral and then evaluating the integral by steepest descent, they found asymptotic decay of the form of Eq. (1), along with explicit expressions for β and τ.

I am concerned here with the relevance of this theory to the experimental observations. What is the evidence? For example, Douglas[2] analyzed data of Kurkjian on stress relaxation in a glass, Chamberlin, Mozurkewich, and Ohrbach[3] studied the decay of magnetization in a spin-glass, and Williams and Watts[4] studied dielectric relaxation. It is useful to divide the experimental time scale into three segments. For "short" times, the observed quantity decays to 0.99 of its initial value. (The exact value is not important; I use 0.99 for convenience.) At short times one does not necessarily expect stretched exponential decay. For "intermediate" times, the observed quantity decays from 0.99 of its initial value to 0.01 of its initial value. (Again, the exact value is not important, and 0.01 is used for convenience.) *The experimental data fall in this intermediate time range.* At "long" times the observed quantity is less than 0.01 of its initial value, and is hard to measure.

A successful theory must explain stretched exponential decay at intermediate times, where it is actually seen. How does PSAA's procedure actually test out? There is no question that they obtain asymptotic stretched exponential decay. But, as is well known, asymptotic calculations have limited validity.

The specific weights and relaxation times used by PSAA are

$$w_n = w_0/\lambda^n, \quad n \geq 0,$$
$$\tau_{n+1}=2^{\mu_n}\tau_n, \quad n\geq 0, \tag{3}$$
$$\mu_n=\mu_0/n, \quad n\geq 1.$$

The decay is characterized by the four parameters w_0, λ, τ_0, and μ_0. These determine the exponent β and the effective relaxation time τ. [Their Eq. (14) for τ is rather complicated and has some misprints.]

I have computed $q(t)$ directly by summing its defining series, for the parameters $w_0=0.5$, $\lambda=2$, $\tau_0=1$, and $\mu_0=2$. These lead to the asymptotic estimates $\beta=0.419$ and $\tau=2.92$. For times in the interval $10^{-3}<t<10^4$, the first 100 terms of the sum were sufficient.

At time $t=0.1$, I find $q_{\rm sum}=0.95$, while $q_{\rm asy}=0.78$. ($q_{\rm asy}$ was determined from the stretched exponential with $\beta=0.419$ and $\tau=2.92$.) At $t=1.0$, $q_{\rm sum}=0.62$, and $q_{\rm asy}=0.53$. For $t>10$, $q_{\rm sum}$ and $q_{\rm asy}$ have values that would be hard to distinguish experimentally, but the relative error is still large. At $t=100$, $q_{\rm sum}=0.018$ and $q_{\rm asy}=0.012$.

Another test is to compute an effective $\beta(t)$ by evaluation of

$$\beta(t)=[\ln\ln 1/q(t)]/\ln(t/\tau) \tag{4}$$

with use of $\tau=2.92$. I find $\beta(0.1)=0.85$, $\beta(1.6)=0.62$, and $\beta(4)=0.48$. These times are all in the intermediate range. The effective β drops from an initial value of 1 to a minimum of about 0.39 (at $t=40$) and then begins a very slow rise to the asymptotic value 0.419. Even at $t=10^4$, where $q_{\rm sum}=2\times10^{-13}$, the effective β has only reached 0.414.

These results show clearly that stretched exponential decay is not predicted by PSAA's model *at intermediate times.* Their model works only at long times, where $q(t)$ is very much less than one percent of its initial value.

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