A simple non-linear equation for structural relaxation in glasses

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When a liquid is rapidly cooled, structural degrees of freedom decouple from vibrational ones. In the glass transition region, structural changes become observable over experimental timescales, which vary from seconds to years on a range of several degrees. One way to observe these changes is to measure the volume relaxation of the glass following a temperature step \[2\]. The resulting evolution is highly nonlinear, roughly logarithmic in time and exhibits very long time memory effects. This behavior appears to be quite universal in complex disordered systems. It is found in aging of contact area between dry solids \[3\], vibration induced compaction of granular materials \[4\], the crumpling of thin sheets under stress \[5\] and the relaxation of conductivity in electron glasses \[6\].

In the present work \[1\] we present a novel analysis of volume relaxation experiments. We show that the glass carries memory of its initial state throughout the relaxation (see Fig. 1). In this context we propose an equation to model the relaxation \(\dot{\delta} = a(1 - e^{b\delta})\), where \(\delta\) is the normalized deviation of volume from equilibrium, and \(a, b\) are state-dependent parameters. This equation is solvable and it reproduces the salient features of both positive (heating) and negative (cooling) temperature steps. When tested against experiments it provides an excellent description of an extensive body of data. Its parameters show a systematic dependence on initial temperature \(T_0\) and

![Figure 1](image_url)

\textbf{Figure 1:} (a) Temperature jumps in PVAc with target temperature \(T=30\text{C}\). Legend details the initial temperatures. (b) Same data time-shifted onto the 60\text{C} curve. Failure to collapse indicate initial state memory.
target temperature $T$. In particular, the basic relaxation rate $a$ has a strong exponential dependence on $T$.

The exponential dependencies of relaxation rate on $T$ and $\delta$ motivate us to offer a rationalization of our model equation in terms of a two-state model. In this model, volume evolves through activated hops over volume-dependent barriers. The above equation is then obtained as an approximation of this model for small volume changes. While it cannot account for the memory of the initial state, this model allows us to extract basic energy and volume scales which are inherent to structural relaxation.

References: