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Note: Physical mechanisms for the bulk melting of stable glasses

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In Ref. 1, we discussed recent work on rejuvenating glasses^{2,3} but we incorrectly stated that these papers do not discuss the bulk transformation dynamics of stable glasses. It has been pointed out to us by Peter Wolynes that the nucleation-and-growth scenario that we analyse is related to a discussion in Refs. 2 and 3. Here, we explain this relationship, focusing on the connection between Section II C 2 of Ref. 1 and the “Rejuvenating glasses” Section of Ref. 2.

Both works consider transformation via propagating fronts that move with velocity v . In our work, these fronts are initiated by nucleation events that occur with rate k_{nuc} per unit volume. If we imagine for a moment that there are no propagating fronts and that each nucleation event leads to transformation of a region of fixed volume R^3 , then the fraction f of material that is transformed after time t would satisfy $\partial f/\partial t = -k_{\text{nuc}}R^3f$ leading to $f(t) = \Phi(t)$ with $\Phi(t) = e^{-k_{\text{nuc}}R^3t}$. In the presence of propagating fronts, the transformation is much faster because mobility propagates outwards from each nucleation event. Let us define the transformation time t_{avr} as $f(t_{\text{avr}}) = 1/e$: this occurs when the typical spacing between nucleation events, $(k_{\text{nuc}}t)^{-1/d}$, is of the same order as the distance that the fronts have propagated, vt . Hence

$$(k_{\text{nuc}}t_{\text{avr}})^{-1} \approx (vt_{\text{avr}})^d. \quad (1)$$

Rearranging yields our Eq. (8). To make the connection with Eqs. (8)-(10) of Ref. 2, we should identify $t_{\text{avr}} \rightarrow t_R$, $k_{\text{nuc}} \rightarrow \mu_0$, $\Phi(t) \rightarrow \phi(T, T_F, t)$, and $R \rightarrow \xi_0$. This recovers the special case $\beta = 1$ of that work, due to our assumption of simple exponential nucleation kinetics.

The more general case $\beta < 1$ considered in Ref. 2 accounts for effects of disorder on the nucleation process of the stable glass. The idea is to replace $\Phi(t)$ by $\Phi_\beta(t) = e^{-(kt)^\beta R^3}$ with $0 < \beta < 1$, in which case $\partial\Phi_\beta/\partial t = -\beta t^{\beta-1}k^\beta R^3\Phi_\beta$. We may interpret $\Phi_\beta^{-1}(\partial\Phi_\beta/\partial t)$ as a nucleation rate, and note that this rate depends strongly on time. This assumption encapsulates the fact that the least stable regions of the glass nucleate quickly (large initial rate) after which only the more stable regions are left, and the rate reduces. As noted in Ref. 2, this stretched exponential form for $\Phi_\beta(t)$ is motivated by fitting to empirical data, so the diverging nucleation rate at small time and the details of the functional form are not physically

significant, although the exponent β can be related to material properties.⁴

This approach yields a generalisation of Eq. (6) of Ref. 1, which is

$$\frac{\partial f}{\partial t} = -f \cdot (kt)^\beta \cdot c_d v^d t^{d-1}, \quad (2)$$

leading to

$$f(t) = e^{-(t/\tau_{\text{str}})^{d+\beta}} \quad (3)$$

with

$$\tau_{\text{str}} \approx (k^\beta v^d)^{-1/(d+\beta)}. \quad (4)$$

These results generalise Eqs. (7) and (8) of our work. This formula for τ_{str} mirrors Eq. (10) of Ref. 2, so our nucleation rate k is inversely proportional to the time scale $\tau(T, T_F)$ defined in that work. As anticipated in our article, the effect of the disordered initial glass state leads to a reduction in the Avrami exponent appearing in Eq. (3) above, in this case from $(d+1)$ to $(d+\beta)$.

It is also interesting to contrast the two approaches. Our identification of a first-order phase transition to interpret the melting of stable glasses¹ allows us to put a different perspective on several issues. (1) The approach to front propagation in Ref. 2 is based on an analogy with combustion, whereas we have discussed it in terms of a nucleation-and-growth picture. (2) The existence of a phase transition clarifies the status of the two phases on either side of the propagating front. (3) By varying the coupling field ε , we can control the nucleation rate of these stable glassy states, allowing the relevant phase transition to be investigated directly by numerical simulations.

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