On the decay of localized vibrational states in glasses: a one-dimensional example

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Abstract

The interaction between three localized vibrational modes is shown to be as relevant for the lifetimes of localized modes as the interaction involving two localized and one extended, and one localized and two extended modes. This contrasts with previous views. I support my arguments by a numerical study of a strongly disordered linear atomic chain.

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Lack of a periodic structure makes the character of the vibrational states (VS) in glasses rather complex. Their spatio-temporal properties depend on the spectral region of interest. The lowest frequency VS are propagons (p, terminology from [1]) - sound waves with a wavevector (of magnitude $2\pi/\lambda$), polarization and velocity. They elastically scatter off local structural imperfections, and as long as their mean free path $\ell > \lambda$, the scattering does not destroy their wave character. What happens at the Ioffe-Regel frequency $\omega_{IR}$, where $\ell \approx \lambda$ (and the wavevector concept becomes invalid) is still a matter of debate. Since $\omega_{IR}$ is very low (less than 5 meV for amorphous silicon (a-Si) where the maximum frequency is $\approx 80$ meV), VS with $\omega > \sim \omega_{IR}$ are responsible for thermodynamic and transport properties of glasses at temperatures ranging from several Kelvins up to the melting point.

Presently there are two different scenarios for what happens beyond the Ioffe-Regel limit. (i) The “fracton” model [3] postulates that all VS with $\omega > \sim \omega_{IR}$ are “locons” ($l$), localized modes which are anderson-localized [4] because of topological disorder. The relation to “fractal” geometry is an historical accident. (ii) In the “diffuson” model [5,6] the majority of modes are “diffusons”, extended and non-propagating modes, carrying energy diffusively. Numerical simulations on realistic models [6,7] show that locons form only a small portion ($\sim 3\%$ in a-Si) of the highest-frequency modes.

In harmonic approximation the VS will never equilibrate, but anharmonic interactions are always present. What is the rate of equilibration in glasses? Experiment [8] indicates that at a liquid He temperature in a-Si (a) high frequency ($\omega \gtrsim 10$ meV) VS live longer as frequency increases, and (b) the VS live on nanosecond timescales. In contrast, phonons in crystals decay faster as frequency increases (the number of decay channels increases), and lifetimes are picoseconds. Model (i) seems to agree with the experiment in the following way. Locons $l$ can decay via three processes: (1) $l \rightarrow p'p''$, (2) $l \rightarrow p'l''$ or $lp' \rightarrow l''$, and (3) $l \rightarrow ll''$ or $ll' \rightarrow l''$. Process (1) is kinematically forbidden for high frequency locons with $\omega_l > 2\omega_{IR}$. Processes (2) represent “propagon-assisted hopping” between locons $l$ and $l''$. Because $\omega_{p'} \ll (\omega_l, \omega_p)$, $\omega_l \approx \omega_{p''}$, and the locons $l$ and $l''$ spatially repel each other [9]. As the frequency and the inverse localization legth $L^{-1}$ increases, the overlap between
the locons decreases and so does the hopping rate. Processes (3) have been neglected by model (i) on the grounds of a small probability of three locon overlap. Hopping is therefore assumed to be the only locon decay process, and because it has property (a) and can be “adjusted” to fit (b), the experiment [8] seems to be rationalized.

Recent numerical calculations on models (i) and (ii) [1] have shown that decay properties of both diffusons and locons are similar to those of crystalline phonons. It has been argued [1] that the assumption of the negligibility of (3) is incorrect and is the reason behind the discrepancy between the numerical results and model (i) predictions. The experiment [8] therefore remains to be explained. In this paper I show, using a one-dimensional (1d) numerical realization of model (i), that extended and localized modes decay on the same time scale and that the three locon interaction is dominant in the decay of locons with \( \omega \gtrsim 2\omega_c \).

Consider a linear chain of \( N \) uniformly spaced atoms connected by random springs [10,11]. Periodic boundary conditions are assumed. In terms of displacements \( u_a \) (\( u_{N+1} \equiv u_1 \)) of atoms \( a \) from equilibrium, the potential energy of the system can be expressed as

\[
V = \frac{1}{2} \sum_{a=1}^{N} K_a (u_a - u_{a+1})^2. \tag{1}
\]

Here \( K_a = K_0 (1 + \xi_a) \), where \( \xi_a \) are random numbers uniformly distributed in the interval \((-b,b)\), and \( K_0 = 10.6 \text{ eV/Å}^2 \) is chosen to simulate a hypothetical linear silicon chain (the silicon atom mass \( M \)) with maximum vibrational frequency \( \omega_{\text{max}} = 2\sqrt{K_0/M} \approx 80 \text{ meV} \) in the case of no disorder (\( b=0 \)). The length scale is the interatomic spacing, the exact value of which is not important here. For the case \( N=3000 \), vibrational eigenstates and frequencies were found by exact numerical diagonalization. This number is sufficiently large to ensure that the conclusions will not be affected by finite size effects. The goal is to numerically realize the “fracton model” scenario with majority of VS localized. As shown later, the strong disorder value \( b=0.7 \) suits this purpose and will be used from now on. The “clean” case, \( b=0 \), will serve as a reference.

Figure [1] shows vibrational density of states (DOS) for the above model and for the case
b=0. The latter can be trivially solved by introducing wavevectors. Consequently its DOS has the \(1/\sqrt{\omega_{\text{max}}^2 - \omega^2}\) dependence and a Van Hove singularity at \(\omega_{\text{max}}\). As the disorder increases, the description of VS in terms of wavevectors is less and less valid. For b=0.7, the Van Hove singularity is washed out and DOS becomes nearly flat.

In a 1d infinite system any disorder causes all VS to be localized \[12\], with a trivial exemption for the zero frequency mode corresponding to a translation of the system as a whole. However, if N is finite there are always low frequency modes with L larger than the system size. They appear to be extended, sound wave-like modes, although on intermediate timescales they behave as diffusons \[13\]. Only after the system size is increased, they become manifestly localized. A good measure of the number of atoms participating in the vibration of mode \(i\) is the participation ratio \(P_i\). Its inverse is defined as

\[
1/P_i = \sum_{a=1}^{N} (e_a^i)^4, \tag{2}
\]

where \(e_a^i\) are normalized \((\sum_a (e_a^i)^2 = 1)\) vibrational eigenstates with frequencies \(\omega_i\). I plot \(1/P\) in Fig. 2. \(1/P\) grows monotonously with frequency, the dependence being close to quadratic for \(\omega \gtrsim 10\) meV. At lower frequencies \(1/P \approx 1/N\), since \(L \gtrsim\) system size (for infinite \(N\) it has been predicted \[14\] that \(1/P \sim \omega^2\) as a result of 1d elastic sound wave scattering). To accurately locate the mobility edge \(\omega_c\) I use the Thouless criterion \[15\], that \(\Delta\omega/\delta\omega\) should exhibit a sharp drop at \(\omega_c\). For a given mode, \(\Delta\omega\) is the locally averaged change of the mode frequency under the change of boundary conditions from periodic to antiperiodic, and \(\delta\omega\) is the local average level spacing. I find it convenient to consider the cumulative quantity \(\tau(\omega) = \int_0^\omega d\omega' \Delta\omega(\omega')/\delta\omega(\omega')\), which is plotted in the inset of Fig. 2. The mobility edge (vertical line) is \(\omega_c \approx 9\) meV and \(\Delta\omega/\delta\omega\) at \(\omega_c\) is less than 1%. Locons with \(\omega \gtrsim \omega_c\) form \(\approx 90\%\) of the spectrum, enough to simulate model (i).

To get a qualitative understanding of how VS decay, consider a simplified version of zero-temperature decay rate formula (full formula see e.g. \[10\]):

\[
\tilde{\Gamma}_i = \sum_{j,k} (J_{ijk})^2 \delta(\omega_i - \omega_j - \omega_k), \tag{3}
\]
where the sum is over all modes $j$ and $k$, and

$$J_{ijk} = \sum_a \eta_a e_a^i e_a^j e_a^k$$

measures the overlap between modes $i$, $j$, and $k$. $\eta_a$ is a binary random variable having values 1 or -1. Only “fission” decay processes ($i \to jk$) are allowed at zero-temperature. “Fusion” processes ($ij \to k$) appearing at finite temperatures will not be considered here, since they can be handled analogously. In 1d VS have a partial memory of a wavevector, and therefore a partial phase coherence. The reason is that the $n$-th vibrational eigenstate has $n-1$ nodes (if the zero-frequency mode is $n=1$). $\eta_a$ is inserted into Eq. 4 to eliminate this coherence, so that the results will not be substantially affected by dimensionality (apart from the DOS effects). Imagine VS with amplitudes independent of frequency. Then an expression similar to Eq. 3 can be obtained for the decay of the VS caused by a set of nonlinear springs with potentials $\sim \eta_a u_a^3 + O(u_a^4)$, externally attached to each atom.

The spectral dependence of $\tilde{\Gamma}$ is shown in Fig. 3. The case $b=0$, plotted in (b), does not represent the decay of 1d crystalline phonons, since the phases of these are randomized by $\eta_a$ in Eq. 4. It, however, shows the hypothetical behavior of random-phase extended modes, such as occur below the mobility edge in 3d. The $\omega$-variation of $\tilde{\Gamma}$ follows the joint density of states (JDOS), $\sum_{jk} \delta(\omega_i - \omega_j - \omega_k)$, and provides a benchmark when discussing the decay behavior of locons. Data in (a) at $\omega \gtrsim \omega_c$ are much more scattered than in (b), since the locon decay involves some overlap statistics (explained below). Therefore local averages are used to represent the data. $\tilde{\Gamma}$ in (a) increases with frequency and follows JDOS, having similar magnitudes as in (b). According to model (i) there would be a sharp decrease in $\tilde{\Gamma}$ beyond $2\omega_c$. This certainly does not happen.

The reason why the behavior of $\tilde{\Gamma}$ differs from the one predicted by model (i) is that processes (3) can not be neglected. In fact they are dominant decay processes for high frequency locons. Consider a decay $i \to jk$, where $i$, $j$, and $k$ are locons with corresponding participation numbers $P_i$, $P_j$, and $P_k$. Eq. 3 can be rewritten as

$$\tilde{\Gamma}_i = \int_0^\infty \int_0^\infty \int_0^\infty N(\omega_j)N(\omega_k)J_i^2(\omega_j, \omega_k)\delta(\omega_i - \omega_j - \omega_k),$$

where $N(\omega_j)$ is the total number
of states at $\omega_j$, and $J^2_i(\omega_j, \omega_k)$ is $J^2_{ijk}$ averaged over frequency shells of $\omega_j$ and $\omega_k$. A simple scaling argument \[1\] shows that $J^2_i(\omega_j, \omega_k)$ does not explicitly depend on $P_i$, $P_j$, or $P_k$. It goes as follows. The maximum decay magnitude among $J^2_{ijk}$ is found when $i$ lies entirely within an overlap of $j$ and $k$, and scales as $1/P_jP_k$ ($e^a \sim 1/\sqrt{P_i}$, etc., and the sum in Eq. 4 is over $P_i$ random-sign numbers and scales as $\sqrt{P_i}$). The probability for this overlap to occur scales as $P_jP_k/N^2$. An important assumption used here is that locons $i$, $j$, and $k$, having different frequencies, are spatially uncorrelated (illustrated below). The average $J^2_i(\omega_j, \omega_k)$ therefore scales as $1/N^2$ and $\tilde{\Gamma}$, is $N$-independent (and follows the JDOS). The small probability of the three-locon overlap is exactly compensated by the large magnitudes of decay matrix elements, when the overlap occurs. As for hopping processes $l \rightarrow p'l''$, one has to distinguish between two cases. If $\omega_{p'} \ll \omega_l, \omega_l'$ (as in model (i) and this paper), the two locons $l$ and $l''$ spatially repel each other \[3\] and the decay rates decrease as $L$ of $l$ and $l''$ decreases. On the other hand, if $\omega_{p'}$ is not small, one can apply the above scaling arguments, since majority of $l$ and $l''$ are not spatially correlated. Decay rates of this “uncorrelated” hopping are therefore independent of the localization character of VS involved and follow the JDOS. When a low frequency ($\omega_l \lesssim 2\omega_c$) locon decays into two propagons, $l \rightarrow p'l''$, the overlap probability is one ($P_j, P_k = N$) and the case is trivial, similar in behavior to three locon and “uncorrelated” hopping processes. These arguments do not depend on $d$ and worked, as far as one could tell, in 3d also \[4\].

Figure 4 illustrates the foregoing discussion. Data are represented by running averages. Decay $l \rightarrow l''$ in (a), obtained by considering only $j$ and $k$ with $\omega_j, \omega_k \geq \omega_c$ in Eq. 3, dominates the high frequency region and grows monotonously with frequency. When, on the other hand, restricting, say modes $j$ to be propagons ($\omega_j \leq \omega_c$), one gets hopping $l \rightarrow p'l''$ which decreases as frequency is increased. Since $\omega_c$ is large enough to allow hopping between locons from different spectral regions, the decrease is quite moderate. The graph (b) in Fig. 4 shows hopping when propagon frequency is restricted by $\omega_p \leq 1$ meV. Computed rates are scattered over several magnitudes. In a sample of decay rates of locons confined into a small frequency band (of the width, say, 1 meV), majority have negligible values, but there
are \( \lesssim 10\% \) of large magnitude rates giving rise to large local averages (they come from a residual overlap between locons with similar frequencies). As the graph shows, local medians sharply decrease with frequency and deviate strongly from local averages. Since the local distribution of decay rates is found to have a strong central tendency around small values (the area in the tails is small), the medians better represent computed data. For other decay types local medians and averages are similar.

Finally, I present figure 5 to support the argument that locons from different spectral regions are spatially uncorrelated, as opposed to locons with similar frequencies, which repel each other. \( N=1000 \) is used for this purpose. The spectrum is divided into \( \delta=1 \) meV wide bands, each band containing 10 VS on the average. For every mode \( i \) in a band only atoms \( a \) having \( (e^i_a)^2 \geq 0.2 \) \( (e^i_{\text{max}})^2 \) are shown, \( e^i_{\text{max}} \) being the largest magnitude found among the coordinates of the vector \( e^i \). Localization starts somewhere beyond 15 meV, where fewer than \( P \approx 200 \) atoms participate in the vibration. There is clearly no statistically relevant overlap between the locons lying within a frequency band. On the other hand, locons from different spectral regions show no sign of repulsion, their spatial positions are essentially uncorrelated.

In summary, I have used a 1d random spring atomic model to demonstrate that the interaction including three locons is important for the locon decay processes. I have shown that extended mode and locon decay rates have similar magnitudes, increasing with increasing frequency and closely following JDOS. Hopping decay rates decrease as frequency increases, the decrease is sharper as the frequency difference of hopping locons gets smaller.

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FIG. 1. Calculated DOS for the 3000 atom linear chain with the spring constant disorder of b=0.7 (solid line) and b=0 (dashed line).

FIG. 2. Inverse participation ratio 1/P and the cumulative Thouless number τ (inset) as a function of frequency. The vertical line in the inset is the mobility edge $\omega_c \approx 9$ meV.

FIG. 3. Calculated $\bar{\Gamma}$ (circles represent uniformly sampled 1/10-th of data) for b=0.7 (a) and b=0 (b). Scattered data in (a) are represented by their running averages (solid line). Vertical lines indicate $\omega_c$ and $2\omega_c$, and dashed lines are JDOS. Units are arbitrary, though the same in (a) and (b).

FIG. 4. Calculated $\bar{\Gamma}$ (a) for the three locon (solid) and hopping (dashed) decay processes, and (b) for hopping between locons from the same frequency region (circles - see caption to Fig. 3). Vertical line in (a) indicates $\omega_c$. Units are the same as in Fig. 3.

FIG. 5. Localization of VS in the 1000 atom linear random spring model. For each mode only the atoms vibrating with amplitudes higher than a certain value (see text) are shown. The solid line is the participation ratio P.
Figure 1: 

(a) 

\[ \Gamma \text{ (arb. units)} \]

\[ (l \rightarrow l'l'') \]

\[ (l \rightarrow p'l'') \]

(b) 

\[ \Gamma \text{ (arb. units)} \]

\[ 10^{-10} \]

\[ 10^{-8} \]

\[ 10^{-6} \]

\[ 10^{-4} \]

\[ 10^{-2} \]

\[ 10^0 \]

\[ 10^2 \]

\[ 10^4 \]

\[ 10^6 \]

\[ 10^8 \]

\[ 10^{10} \]

\[ 0 \]

\[ 20 \]

\[ 40 \]

\[ 60 \]

\[ 80 \]

\[ 100 \]

FREQUENCY (meV)
This figure "fig5.gif" is available in "gif" format from:

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