

Decay of localized vibrational states in glasses: A one-dimensional example

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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. [S0163-1829(97)50606-0]

Lack of a periodic structure makes the character of the vibrational states (VS) in glasses rather complex. Their spatiotemporal properties depend on the spectral region of interest. The lowest frequency VS are propagons (p , terminology from Ref. 1)—sound waves with a wave vector (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 frequency² ω_{IR} , where $\ell \approx \lambda$ (and the wave-vector concept becomes invalid) is still a matter of debate. Since ω_{IR} is very low [less than 5 meV for amorphous silicon (a -Si) where the maximum frequency is ≈ 80 meV], VS with $\omega \gtrsim \omega_{\text{IR}}$ 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” model³ postulates that all VS with $\omega \gtrsim \omega_{\text{IR}}$ are “locons” (l),¹ localized modes which are Anderson localized⁴ because of topological disorder. The relation to fractal geometry is an historical accident. (ii) In the “diffuson” model^{1,5,6} the majority of modes are “diffusons,” extended and nonpropagating 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? Experiment⁸ 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 time scales. 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) $l \rightarrow l'l''$ or $ll' \rightarrow l''$. Process (1) is kinematically forbidden for high-frequency locons with $\omega_l > 2\omega_{\text{IR}}$. Processes (2) represent “propagon-assisted hopping” between locons l and l'' . Because $\omega_p \ll (\omega_l, \omega_{l''})$, $\omega_l \approx \omega_{l''}$, and the locons l and l'' spatially repel each other.⁹ As the frequency and the inverse localization length 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 experiment⁸ seems to be rationalized.

Recent numerical calculations on models (i) and (ii) (Ref. 1) have shown that decay properties of both diffusons and locons are similar to those of crystalline phonons. It has been argued¹ 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 experiment⁸ 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

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

Here $K_a = K_0(1 + \xi_a)$, where ξ_a are random numbers uniformly distributed in the interval $\langle -b, b \rangle$, and $K_0 = 10.6 \text{ eV/\AA}^2$ is chosen to simulate a hypothetical linear silicon chain (the silicon atom mass M) with maximum vibrational frequency $\omega_{\text{max}} = 2\sqrt{K_0/M} \approx 80$ 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 a 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 the vibrational density of states (DOS) for the above model and for the case $b=0$. The latter can be trivially solved by introducing wave vectors. Consequently its DOS has the $1/\sqrt{\omega_{\text{max}}^2 - \omega^2}$ dependence and a van Hove singularity at ω_{max} . As the disorder increases, the description of VS in terms of wave vectors is less and less valid. For $b=0.7$, the van Hove singularity is washed out and the DOS becomes nearly flat.

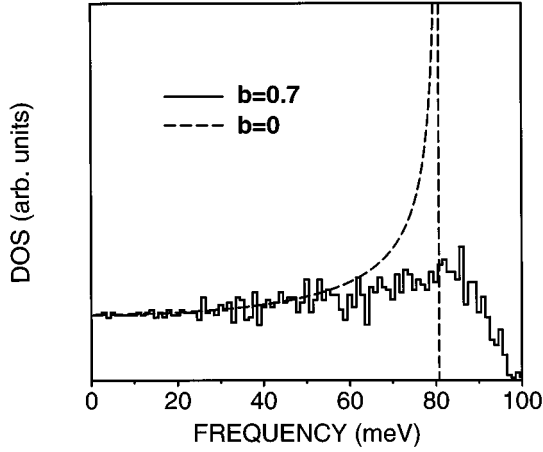


FIG. 1. Calculated DOS for the 3000-atom linear chain with the spring constant disorder of $b=0.7$ (solid line) and $b=0$ (dashed line).

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 time scales 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

$$1/P_i = \sum_{a=1}^N (e_a^i)^4, \quad (2)$$

where e_a^i are normalized [$\sum_a (e_a^i)^2 = 1$] vibrational eigenstates with frequencies ω_i . I plot $1/P$ in Fig. 2. $1/P$ grows monotonously with frequency, the dependence being close to quadratic for $\omega \geq 10$ meV. At lower frequencies $1/P \approx 1/N$, since $L \geq$ system size (for infinite N it has been predicted¹⁴ that $1/P \sim \omega^2$ as a result of 1D elastic sound-wave scatter-

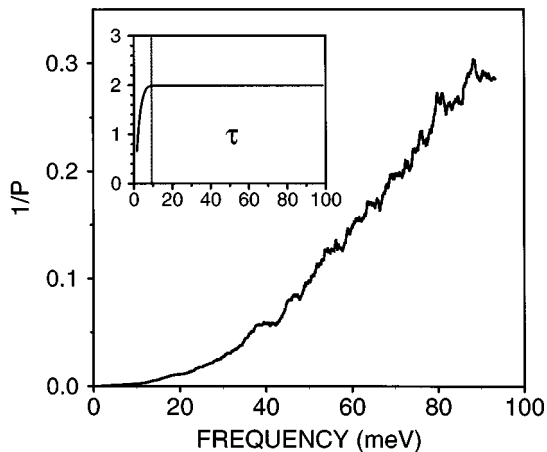


FIG. 2. Inverse participation ratio $1/P$ and the cumulative Thouless number τ (inset) as a function of frequency. The vertical line in the inset is the mobility edge $\omega_c \approx 9$ meV.

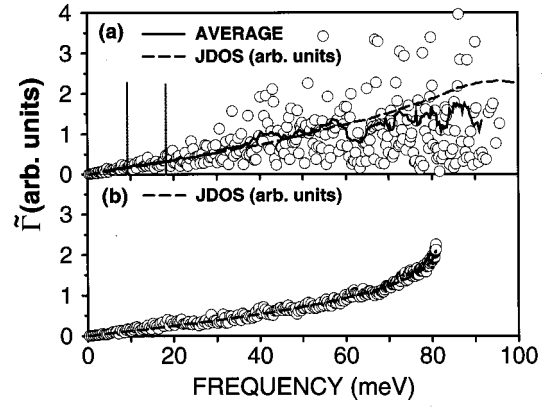


FIG. 3. Calculated $\tilde{\Gamma}$ (circles represent uniformly sampled one-tenth of data) for $b=0.7$ (a) and $b=0$ (b). Scattered data in (a) are represented by their running averages (solid line). Vertical lines indicate ω_c and $2\omega_c$, and dashed lines are JDOS. Units are arbitrary, though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

ing). To accurately locate the mobility edge ω_c , I use the Thouless criterion¹⁵ that $\Delta\omega/\delta\omega$ should exhibit a sharp drop at ω_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 ω_c is less than 1%. Locons with $\omega \geq \omega_c$ 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 the zero-temperature decay rate formula (for the full formula see, e.g., Ref. 16),

$$\tilde{\Gamma}_i = \sum_{j,k} (J_{ijk})^2 \delta(\omega_i - \omega_j - \omega_k), \quad (3)$$

where the sum is over all modes j and k , and

$$J_{ijk} = \sum_a \eta_a e_a^i e_a^j e_a^k \quad (4)$$

measures the overlap between modes i , j , and k . η_a is a binary random variable having values 1 or -1 . Only ‘‘fission’’ decay processes ($i \rightarrow jk$) are allowed at zero temperature. ‘‘Fusion’’ processes ($ij \rightarrow k$) appearing at finite temperatures will not be considered here, since they can be handled analogously. In 1D, VS have a partial memory of a wave vector, 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 $n=1$).¹⁰ η_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 η_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 ω 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 $\omega \geq \omega_c$ 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) cannot be neglected. In fact they are dominant decay processes for high-frequency locons. Consider a decay $i \rightarrow jk$, where i , j , and k are locons with corresponding participation numbers P_i , P_j , and P_k . Equation (3) can be rewritten as $\tilde{\Gamma}_i = \int \int d\omega_j d\omega_k N(\omega_j) N(\omega_k) J_i^2(\omega_j, \omega_k) \delta(\omega_i - \omega_j - \omega_k)$, where $N(\omega_j)$ is the total number of states at ω_j , and $J_i^2(\omega_j, \omega_k)$ is J_{ijk}^2 averaged over frequency shells of ω_j and ω_k . A simple scaling argument¹ shows that $J_i^2(\omega_j, \omega_k)$ does not explicitly depend on P_i , P_j , or P_k . It goes as follows. The maximum decay magnitude among J_{ijk}^2 is found when i lies entirely within an overlap of j and k , and scales as $1/P_j P_k$ [$e_a^i \sim 1/\sqrt{P_i}$, 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 $P_j P_k / N^2$. An important assumption used here is that locons i , j , and k , having different frequencies, are spatially uncorrelated (illustrated below). The average $J_i^2(\omega_j, \omega_k)$ therefore scales as $1/N^2$ and $\tilde{\Gamma}_i$ 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 $\omega_p \ll \omega_l, \omega_{p''}$ [as in model (i) and this paper], the two locons l and l'' spatially repel each other⁹ and the decay rates decrease as L of l and l'' decreases. On the other hand, if ω_p is not small, one can apply the above scaling arguments, since, the 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, $l \rightarrow p'p''$, the overlap probability is 1 ($P_j, P_k = N$) and the case is trivial, similar in behavior to three locon and uncorrelated hopping processes. These arguments do not depend on dimension and worked, as far as one could tell, in 3D also.¹

Figure 4 illustrates the foregoing discussion. Data are represented by running averages. Decay $l \rightarrow l'l''$ 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 ω_c is large enough to allow hopping between locons from different spectral regions, the decrease is quite moderate. The

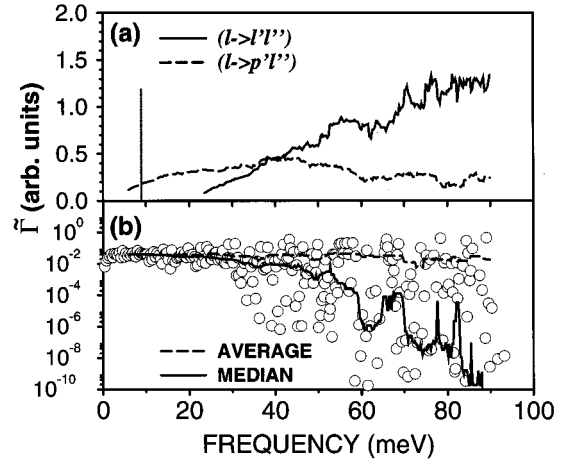


FIG. 4. Calculated $\tilde{\Gamma}$ (a) for the three-locon (solid) and hopping (dashed) decay processes, and (b) for hopping between locons from the same frequency region (circles—see caption to Fig. 3). Vertical line in (a) indicates ω_c . Units are the same as in Fig. 3.

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), the majority have negligible values, but there are $\leq 10\%$ 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 Fig. 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

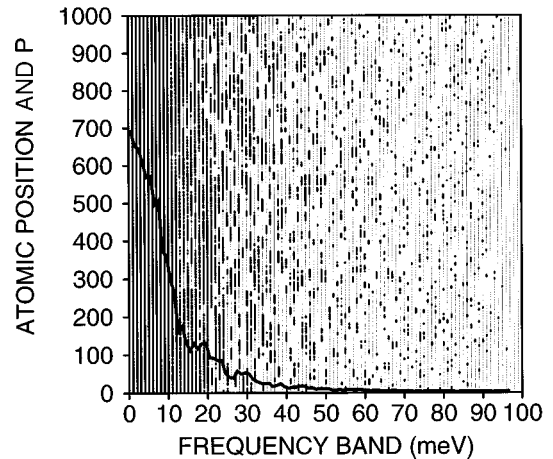


FIG. 5. Localization of VS in the 1000-atom linear random spring model. For each mode only the atoms vibrating with amplitudes higher than a certain value (see text) are shown. The solid line is the participation ratio P .

containing ten VS on the average. For every mode i in a band only atoms a having $(e_a^i)^2 \geq 0.2 (e_{\max}^i)^2$ are shown, e_{\max}^i 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.

One may ask how the above results depend on the degree of disorder. I have made calculations similar to those above, with different values of b (up to almost $b = 1$). I found the foregoing conclusions valid for all the studied cases, the only effect of the increase in b being the increase of the spread of

the calculated $\tilde{\Gamma}$. This happens because by increasing b , L decreases, as does the number of the decay events, and the overlap statistics gets worse.

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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