

Vibrational Modes as a Predictor for Plasticity in a model Glass

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The density of vibrational states in amorphous materials is known to present an unusual shape related as “Boson Peak”, and responsible for the very specific thermal behaviour of these systems. In this letter, we show how the vibrational modes of a model Lennard-Jones glass are affected by a mechanical load. Far from a mechanical instability, vibrational modes can be described at low frequency by weak scattering of acoustic modes. Close to a plastic instability, some of them localize. We show how the shape of the “localized” vibrational modes, just before the plastic instability, is directly related with the spatial organization of the plastic rearrangements. A measurement of the spatial organization of the low-frequency vibrational modes could thus be used as a predictor for plastic activity.

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While the vibrational response of crystals is now very well understood [1], the vibrational response of amorphous materials is still the subject of active debates [2–14]. The description of the vibrational normal modes as plane waves is not possible in disordered materials, even at low frequencies; a feature that is shared by a large number of different materials, ranging from silica glasses[4] to polymers[3], metallic glasses[14], foams[15] or even dense granular packings[6]. This anomalous vibrational behaviour affects the low-temperature dependence of the specific heat and thermal conductivity[16], that are governed by acoustic excitations[1] and it has thus different important applications, not only in terms of sound waves propagation. However, the precise shape of the normal vibrational modes is not well known at present in disordered materials. The departure from plane waves is sometimes related with “soft modes” [8–10] or “quasi-localized modes”[11, 12] and gives rise to Anderson localization at very high frequency [6]. This problem is related with the classical problem of sound waves propagation in a heterogeneous material [13] with intermediate disorder [7, 18, 22], and is generally not tractable analytically. Different models have been proposed: some of them are related with a self-consistent resolution of wave’s scattering[8] in the weak[17] and in the strong[13] disorder approximation. Other models involves the non-harmonicity of the couplings [12]. But none has shown how the disorder strength, and thus the vibration modes, are affected by an external mechanical load. In this paper, we show a numerical resolution of the vibrational eigenmodes of a 2D model glass, based on the exact diagonalization of its Dynamical Matrix. We show how the density and the shape of the eigenmodes is affected by the proximity to a mechanical plastic instability upon mechanical load. In the last part of this letter, we show numerical evidence of the direct relation between the spatial organization of the plastic rearrangements and the shape of the eigenmodes describing the vi-

brational response of the system before the plastic event occurs.

The systems we study here are two-dimensional assemblies of polydisperse beads interacting through a Lennard-Jones potential [18–22] $u_{LJ}(r_{ij}) = 4\epsilon((\sigma/r_{ij})^{12} - (\sigma/r_{ij})^6)$. The corresponding units are $\sigma = 1.00 \pm 0.05LJ \approx 10^{-10}m$, $\epsilon = 1LJ \approx 1eV$, $m_i = 1LJ \approx 10^{-25}kg$ and $\delta t = 1JL = \sqrt{m\sigma^2/\epsilon} \approx 10^{-12}s$. The systems are composed of $N = 10000$ particles with an average density of $\rho = 0.925$ particles per unit volume, corresponding to a very small pressure $P \approx 0.2$ at rest. The different configurations are prepared in an amorphous state by heating an initially ordered and close packed assembly of beads, then kepted in the liquid state during 10^5 molecular dynamics steps corresponding to approx. 10^{-9} seconds, and quenched very quickly from the liquid state into a solid and disordered state corresponding to the nearest equilibrium position (local minimum of the total potential energy)[18]. The system is then submitted to a quasi-static mechanical load, by applying an incremental shear on the borders of the samples. The method used here is the rigorous limit of a deformation at finite shear rate [24]. The upper border is displaced along its direction with an incremental strain $\delta\epsilon_{xy} = 5.10^{-5}$ (this incremental strain can be reduced to 10^{-8} in order to approach closely a plastic instability). After the displacement of the border, the system is then relaxed into its new nearest equilibrium position, and the procedure is repeated up to a total cumulated imposed strain of 170%. Two boundary conditions have been compared: fixed walls [18] or biperiodic Lees-Edwards boundary conditions [19]. For each equilibrium position corresponding to a given strain, the vibration modes are computed through the exact diagonalization of the Dynamical matrix \mathcal{H} that is the matrix of the second

order derivatives of the potential energy \mathcal{U} .

$$\mathcal{U} = \sum_{(i,j)} u_{LJ}(r_{ij}) \approx \mathcal{U}_{eq} + \frac{1}{2}(r - r_{eq}) \cdot \mathcal{H} \cdot (r - r_{eq})$$

where $(r - r_{eq})$ represents the displacement of all the particles from their equilibrium position r_{eq} . For each particle i

$$m_i \frac{d^2 r(i)}{dt^2} = - \sum_j \mathcal{H}_{ij} \cdot (r(j) - r_{eq}(j))$$

The normal vibrational modes and the eigenfrequencies are respectively the eigenvectors, and the squareroot of the eigenfrequencies of \mathcal{H} . The diagonalization of the 20 000 x 20 000 dynamical matrix is performed using the Implicitly Restarted Lanczos method [25]. Note that since \mathcal{H} is defined by the second order expansion of the potential energy, our computation of the normal vibrational modes does not take into account at this stage possible anharmonic contributions. However, the density of vibrational states shows already an anomalous behaviour as compared with the Debye density of states. Figure 1-a shows the total density of states computed by counting the number of modes with eigenfrequency between w and $w + \delta w$ and normalized to 1 for the full spectrum. $\delta w = 0.02$ is chosen in order to let the density of states unchanged as a function of δw . The density of states is then divided by the Debye prediction[1] $g_D(w) = 2w/w_D^2$ where w_D is the 2D Debye frequency, $w_D = (8\pi\rho/(1/C_L^2 + 1/C_T^2))^{1/2}$, $C_L = \sqrt{(\lambda + 2\mu)/\rho}$ is the group velocity of the longitudinal waves and $C_T = \sqrt{\mu/\rho}$ is the velocity of the transverse waves in the ideal homogeneous and isotropic material with the same density ρ and the same macroscopic elastic constants λ and μ . The Debye density corresponds to the number of plane waves of a given frequency in an homogeneous sample. As shown in the figure 1, the normalized density of vibrational states, in the disordered material, shows a peak around $w = 1.6 LJ \approx 1.6 \cdot 10^{12} Hz$. This peak is commonly designed as the ‘‘Boson Peak’’. We show here that it is present even if the anharmonicity of the couplings is not taken into account. Note also that the density of vibrational states, and the boson peak, is displaced to smaller frequencies before a plastic event occurs (red curve in figure 1), in agreement with the general decrease of the effective elastic moduli [21, 22].

The visualization of the normal modes is possible numerically. It shows a progressive departure from plane waves, as the frequency is increased. In order to characterize it, the participation ratio (PR) is computed (see Figure1-b). It is defined as

$$PR = \frac{1}{N} \cdot \frac{\left(\sum_{i=1}^N u_i^2\right)^2}{\sum_{i=1}^N u_i^4}$$

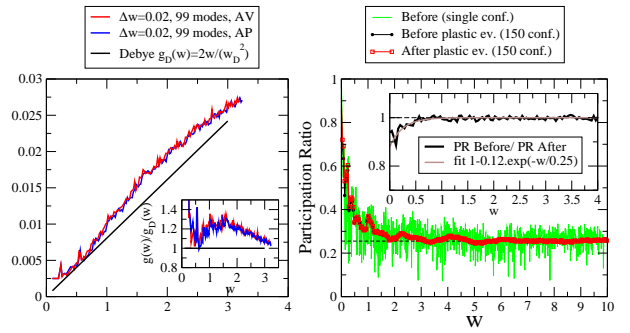


FIG. 1: (a) Density of vibrational states in a 2D Lennard-Jones glass with fixed walls, averaged over 100 configurations, and with $\Delta\omega = 0.02LJ$. Red: for $\delta\epsilon = 5.10^{-5}$ before a plastic rearrangement (AV). Blue: for $\delta\epsilon = 5.10^{-5}$ after a plastic rearrangement (AP). Before the plastic rearrangement (red curve, AV), the density of vibrational states is displaced to smaller frequencies. Here for fixed borders. (b) Participation ratio of the tausend first vibrational modes before (black curve, AV) and after (red curve, AP) a plastic rearrangement, averaged over 150 configurations, with Lees-Edwards boundary conditions. Inset: Ratio between the PR before over after a plastic event. Fit with an exponential decay of characteristic frequency $w_c = 0.25uLJ$.

where u_i is the amplitude of the displacement $r(i) - r_{eq}(i)$ of the particle i . The Participation ratio is equal to 1 when all particles moves together (translational invariance), and would be equal to $1/N$ when one single particle is moving. It gives a percentage of particles that are moving together. The PR shows a progressive convergence from $PR = 1$ (translational invariance), to $PR = 3/8$ characteristic of plane waves, and then to a smaller value $PR \approx 0.255$ related with the increase of the spatial heterogeneity in the particle displacements. Fluctuations are visible around this average value. We will comment later on the detailed evolution of the participation ratio with the proximity to a plastic event. But first, we will focus on the departure from the eigenmodes that would be obtained in a homogeneous material with the same global elastic moduli. In case of a biperiodic system with Lees-Edwards boundary condition, the solutions [26] to the homogeneous problem of sound wave propagation is very simple. They are the longitudinal and transverse sound waves $\vec{u}(\vec{r}_i) = \vec{u}_0 \cdot \exp(-\vec{k} \cdot \vec{r}_i - w \cdot t)$ with $k_x = n_x \cdot 2\pi/L_x$, $k_y = n_y \cdot 2\pi/L_y$ and $w = c_{L,T} \cdot |k|$. Figure 2 shows the projection of the normal modes of the system found by exact diagonalization of the dynamical matrix on the solutions of the homogeneous systems (plane waves) sorted by increasing frequencies. The figure is centered on the frequency of the homogeneous solution of the same order (for example, for the third eigenmode, we trace the amplitude of the projection of this eigenmode on the homogeneous solutions, as a function of the frequency of the homogeneous modes minus the frequency of the third mode of the homogeneous solu-

tion). This figure shows clearly that, below the boson peak, the main projection is given by the homogeneous mode of the same order ($\Delta w = 0$), and that the projection progressively enlarges to homogeneous modes with close frequencies. Beyond the Boson Peak, the projection is displaced to the left, that is to smaller frequencies, and is very noisy. Figure 2-b quantifies the enlargement of the projections below the Boson peak. We subtract here the amplitude of the main projection to the total amplitude of the displacement field. We thus obtain the amplitude of the noise $\Delta u \equiv u - u_h$ where u_h is the solution of the corresponding vibration mode in the homogeneous material. Figure 2 (b) shows a dependence $\Delta u/u \propto (\xi/\lambda)^2$ that is characteristic of a weak Rayleigh Scattering[13] on a heterogeneous structure with a characteristic length $\xi \approx 25 LJ$. At this stage, it is difficult to show evidence of any localization in the vibration modes of the system, and the average behaviour corresponds to “quasi-localized” vibration modes, with a large contribution of the homogeneous mode at low frequency, then a weak scattering up to the Boson Peak, and finally saturation to a spatially correlated noise with an intermediate Participation Ratio of about 0.25. Indeed, we will show now that the proximity to a plastic event affects the shape of the modes, and that few localized eigenmodes are sufficient to predict the spatial organization of the future plastic rearrangements. First, we can show in figure 1-b that before a plastic event, few localized eigenmodes appears at low frequencies. In a previous paper[22], we have shown that a Plastic event can be predicted by the decrease of the local shear modulus at a strain of 0.2% before a plastic rearrangement occurs. The plastic rearrangement is then located around the soft place characterized by a very low local shear modulus[22]. A. Lemaître et al.[7] have also mentioned the fact that before a plastic instability, some eigenfrequencies decrease rapidly. In this letter we are going further, because first, we relate the plastic rearrangement to localized vibrational modes of the system before the plastic event occurs, and second, we compare the case of a local quadrupolar plastic rearrangement (localized spatially), and the case of a large scale plastic event (shear banding).

We show in figure 3 the case of a local, quadrupolar plastic rearrangement. As already shown in [21, 22], the plastic rearrangement described by the arrows standing for the displacement of the particles during the dissipative rearrangement (figure3-c) is located in the soft region (figure3-a). It is entirely described by the first eigenvector with a participation ratio less than the arbitrarily small value of $TP = 0.2$. Other eigenmodes with $TP < 0.2$ are shown in figure 3 (e to i). They do not contribute to the plastic rearrangement that is entirely described by the localized eigenmode (here with $TP \approx 0.1$) that has the smallest frequency ($\omega \ll \omega_{BP}$). The case of an extended plastic rearrangement, also described in a previous paper as “elementary shear band”[20], is very

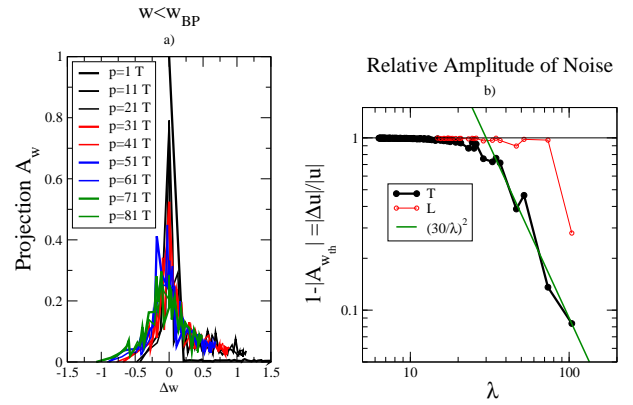


FIG. 2: (a) Projection of the transverse vibration modes on the plane waves corresponding to the vibration modes in a homogeneous and isotropic material. The projection coefficient A_w is averaged over 20 different configurations with Lees-Edwards boundary conditions. (b) Amplitude of the noise $(1 - |A_w|)$ obtained by subtracting the main plane wave from the numerically computed eigenmode (with Lees-Edwards boundary conditions), as a function of the theoretical wavelength $\lambda = L/\sqrt{n_x^2 + n_y^2}$

different. In this case, it is more difficult to identify a local origin to the plastic event, because it spans the entire system and results in a sheared plane (figure4-c the sheared plane - or shear band in a 2D system - is vertical here). It is not clear up to now whether this elementary shear band can be described as resulting from a dynamical cascade of localized quadrupoles [27] or is a collective large scale instability resulting from the percolation of soft zones in the system [22]. From figure4-a, this large plastic event can be related to a spatial structuration in the shear moduli, but the localized eigenmode with the smallest eigenfrequency (Fig.4-d) is not sufficient to predict the position of the shear band. We show in this letter, that such a large event is related to a large number of additional localized eigenmodes of different frequencies (see figure1-b). Surprisingly, and this is one of the main result of this letter, the position of the shear band is even shown clearly in the superposition of the different localized vibration modes, before the plastic event occurs. Figure4-b shows the superposition of the amplitudes of the displacement of the particles involved in the 30 more localized vibrational eigenmodes (with $TP < 0.2$). In contradiction with the case of an isolated quadrupolar rearrangement, a single vibrational eigenmode is not sufficient to recover the spatial structuration of the full plastic rearrangement. But the spatial structuration of the plastic event is already present in the different localized eigenmodes with different eigenfrequencies appearing in the spectrum before the plastic event occurs. The localized eigenmodes are thus directly related with the unstable plastic rearrangements. The different localized eigenmodes appearing before the plas-

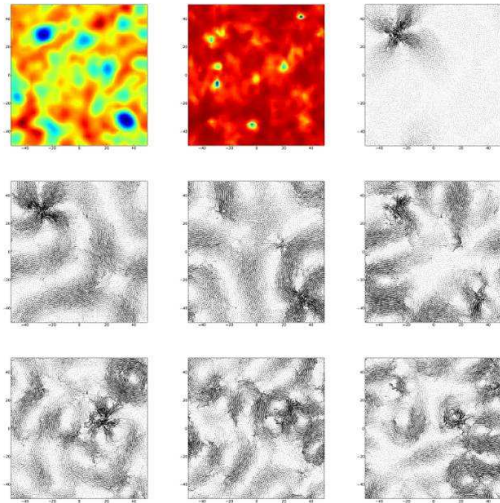


FIG. 3: Case of a local plastic rearrangement. From left to right, and from top to bottom: (a) Map of local shear moduli; (b) Superposition of the amplitudes of the 30 first eigenmodes with $TP_j 0.2$; (c): Plastic rearrangement; (c-d-e-f-g-h-i): First eigenvectors with $TP_j 0.2$.

tic instability seem to couple during the plastic rearrangement. This remark opens new perspectives into the study of the mechanical instability. It would be very interesting to understand the way the microscopic interactions would change, leading to such a coupling between localized vibrational eigenmodes during the non-equilibrium plastic rearrangement. This connection between “soft modes” and dynamical irreversibility has already been mentioned by A. Widmer-Cooper *et al.* [28] in the very different context of supercooled liquids. We show here, that this connection is indeed already present in the low temperature ($T \ll T_g$) mechanical behaviour of the amorphous solid. In our case, the irreversible motion of the particles results only from plastic instability without thermal activation. It underlines the role of the mechanical couplings into the dissipative behaviour of the particles. Moreover, in addition to [28], our systems are sufficiently large to have a large number of delocalized vibration modes. Our results show that the plastic instability is related more specifically with the “localized” eigenmodes ($TP < 0.2$) appearing just before the mechanical instability. This gives, in our opinion, a very concrete definition to the “soft modes” frequently mentioned in the literature.

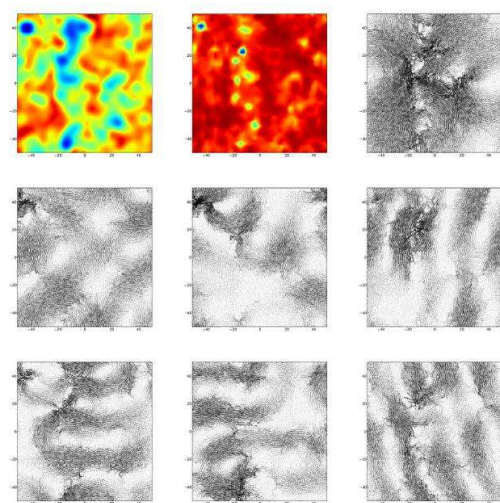


FIG. 4: Case of an elementary shear band. From left to right, and from top to bottom: (a) Map of local shear moduli; (b) Superposition of the amplitudes of the 30 first eigenmodes with $TP_j 0.2$; (c): Plastic rearrangement; (c-d-e-f-g-h-i): First eigenvectors with $TP_j 0.2$.

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