Instantaneous normal modes of a glass-forming liquid during the relaxation process

Masanari Shimada,1,∗ Kumpei Shiraishi,1 Hideyuki Mizuno,1 and Atsushi Ikeda1,2

1Graduate School of Arts and Sciences, The University of Tokyo, Tokyo 153-8902, Japan
2Research Center for Complex Systems Biology, Universal Biology Institute, The University of Tokyo, Tokyo 153-8902, Japan
(Dated: November 24, 2021)

Understanding glass formation by quenching remains a challenge in soft condensed matter physics. Recent numerical studies on steepest descent dynamics, which is one of the simplest models of quenching, revealed that quenched liquids undergo slow relaxation with a power law towards mechanical equilibrium and that local rearrangements of particles govern the late stage of the process. These advances motivate the detailed study of instantaneous normal modes during the relaxation process because they are significant in the dynamics governed by stationary points of the potential energy landscape. Here, we performed a normal mode analysis of configurations during relaxation and found that unstable localized modes dominate the dynamics. We also observed power-law relations between several fundamental observables and a stretched exponential law in the most unstable mode of a configuration. These findings substantiate our naive expectation about the relaxation dynamics based on quantitative analysis.

I. INTRODUCTION

In order to avoid crystallization, glasses are typically made from liquids by quenching [1, 2]. This is contrasted with the quasistatic processes, through which the system remains in thermal equilibrium. The non-equilibrium relaxation dynamics after quenching is highly non-trivial and has attracted much interest in the field of glassy systems because their equilibration time exceeds the experimental timescale below the glass transition temperature $T_g$; in other words, they are always out of equilibrium. The complexity of the relaxation dynamics of glasses is most vividly demonstrated in a phenomenon called aging [3–5]. Below $T_g$, the two-time correlation functions depend on the waiting time after the system is quenched into a target temperature. This behavior is never observed in the equilibrium systems and is a characteristic of non-equilibrium glassy systems.

To further understand the complicated process of relaxation, recent numerical studies have focused on steepest descent dynamics [6–9]. This is one of the simplest models of quenching to zero temperature; the Langevin equation is applied with the noise term suppressed, i.e., the velocity of a particle is proportional to the force acting on it. Particularly in Ref. [6], packings of athermal particles slightly above jamming were studied in both two and three spatial dimensions. It was found that the root mean squared particle speed, which is directly connected to the time derivative of the total potential energy, decays with a power law $\propto t^{-\beta}$. The exponent $\beta$ depends on the spatial dimension: $\beta = 0.92$ for two dimensions and $\beta = 0.85$ for three dimensions. This power-law relaxation of liquids into the underlying local minima of the potential energy landscape, called inherent structures, is governed by temporally intermittent and spatially localized spots of non-affine displacements, which are called hot spots in Ref. [6]. The hot spots are surrounded by backgrounds exhibiting swirling motion and are becoming more prominent and sparser with time, thus resembling local plastic events in glasses under shear, known as shear transformation zones [10–13].

Soon after the research reported in Ref. [6], Nishikawa et al. investigated the steepest descent dynamics in several glass-forming models over a broad range of temperatures and spatial dimensions [7]. The results showed that a mean-field Mari-Kurchan model exhibits a transition of the relaxation dynamics. The root mean squared particle speed follows a power-law decay at high temperatures and an exponential decay at low temperatures. Such behavior was predicted by a recent theoretical study of a mean-field spin glass model, the mixed $p$-spin model [14]. In contrast, in infinite dimensional models, the relaxation dynamics does not show a transition even much below the mode-coupling transition temperature $T_{MCT}$. In all the models considered, the root mean squared particle speed decays algebraically with the same exponent $\beta$ reported in Ref. [6] at high temperatures. Thus, this exponent is universal for systems with different pairwise interactions and depends only on the spatial dimension. These relaxation processes are dominated by spatially localized hot spots as observed in Ref. [6]. The density of hot spots rapidly decreases with decreasing temperature, but it remains finite and does not exhibit singular behavior. This means that the hot spots dominate the relaxation dynamics at all temperatures in the finite dimensional models.

∗ masanari-shimada444@g.ecc.u-tokyo.ac.jp

In Ref. [7], hot spots are called defects using a different but equivalent definition.
In view of this progress, it is essential to explore the spatial structure of the hot spots in depth. Unlike equilibrium liquids, it is predicted that configurations during relaxation are getting close to stationary points [4]. Thus, investigating instantaneous normal modes is a reasonable approach to study hot spots. Such regions with high mobility will be linked to unstable normal modes of instantaneous configurations. Although several studies have investigated instantaneous normal modes of equilibrium liquids [1, 15–20] and glasses under finite-rate shear [21], there is no study to our knowledge that has thoroughly explored instantaneous modes during the relaxation process after quenching.

Contrary to instantaneous normal modes, there have been a large number of studies on normal modes of inherent structures and saddles. Because the low-frequency modes of inherent structures have been extensively studied [22–24], it is important to compare them with unstable normal modes during relaxation. In the lowest-frequency tail of the spectrum of inherent structures, spatially localized modes whose cores are surrounded by algebraically decaying far fields are observed for a broad class of glasses, in addition to acoustic phonons [22]. Such modes, known as quasi-localized modes, are attributed to several anomalous low-energy properties of glasses, including plastic events under shear [25–27] and low-temperature thermal properties [28, 29]. Recently, comparing the quasi-localized modes and unstable modes found at saddles, it was also established that these two types of modes are deeply interlinked in terms of their structures [30]. Along these lines, it is meaningful to analyze instantaneous modes during relaxation using the methods adopted to analyze the quasi-localized modes.

Pertinent to the normal mode analysis of glasses, the notion of marginal stability [31–33] assumes an increasingly important role in understanding anomalous behavior of glasses. The marginal stability of amorphous materials is characterized by the flat free energy landscape predicted by replica theory as full replica symmetry breaking [32, 33], which is related to the ultrametricity or fractality of the landscape. In terms of mechanical stability, it is considered to lead to low-energy excitations with a gapless spectrum and extreme susceptibility to perturbations [31]. Marginal stability is now considered to be one of the fundamental properties of disordered materials [31]. If the geometry of the potential energy landscape is investigated through a relaxation process with a normal mode analysis, it is possible to evaluate its flatness directly [4]. For example, the theoretical analysis of marginal stability using a normal mode analysis during relaxation dynamics was performed in Ref. [4].

Based on the above considerations and results, we have explored instantaneous normal modes of configurations during relaxation using a typical glass-forming liquid. Instead of the ensemble average at a fixed time, we considered the average at a fixed mean squared force, which is defined shortly. Since many observables exhibit rapid time dependences in the final stage of the dynamics, the latter average is easier to treat. Using numerical methods that have been developed in studies of inherent structures, we have investigated unstable instantaneous normal modes, which can be seen as “bare” hot spots [6, 7]. We have found power-law relations between several fundamental observables and found that the unstable localized modes dominate the dynamics, particularly in the late stage in accordance with the previous observations [6, 7]. Furthermore, focusing on the most unstable mode of each sample, we revealed that its core is left to be unstable while the overall system rapidly stabilizes following a non-trivial functional form. These findings can help us understand the steepest descent dynamics by bridging the two different descriptions: one based on the real space and another one based on the potential energy landscape.

In Sec. II, we delineate the model and numerical methods. In Sec. III A, we investigate the time evolution of the mean squared force and several statistical properties of instantaneous normal modes. In Sec. III B, we focus on the most unstable mode and analyze its spatial structure. In Sec. IV, we conclude this paper. In Appendix A, we display the data that suggest that the most unstable mode of a configuration has only a single core. In Appendix B, we examine the system size dependence of several quantities in Sec. III B. In Appendix C, we discuss asymptotic behavior of the energy profile defined in Sec. III B. In Appendix D, we present data at very low temperatures generated by the replica exchange molecular dynamics (MD) method [34, 35].

II. MODEL AND METHODS

Our model is based on the three-dimensional Kob-Andersen binary Lennard-Jones (KALJ) model [36, 37], which is widely used in the context of the glass transition because of its particularly high glass-forming ability. In this model, there are two types of particles, A and B, with a composition ratio of 80:20. Both types of particles have the same mass $m$, and the particles interact with each other via a pair potential

$$v_{0,ab}(r) = 4\epsilon_{ab} \left[ \left( \frac{\sigma_{ab}}{r} \right)^{12} - \left( \frac{\sigma_{ab}}{r} \right)^6 \right],$$

where $r$ is the distance between a pair and the parameters are as follows: $\epsilon_{AB} = 1.5\epsilon_{AA}, \epsilon_{BB} = 0.5\epsilon_{AA}, \sigma_{AB} = 0.8\sigma_{AA}$, and $\sigma_{BB} = 0.88\sigma_{AA}$. In this paper, length, mass, and time are reported in units of $\sigma_{AA}, m$, and $\sqrt{m\sigma_{AA}^2/\epsilon_{AA}}$, respectively. The total number density is fixed at $\rho = 1000/(9.4)^3 \approx 1.204$ [36, 37]. In the KALJ model, Eq. (1) is
TABLE I. The number of samples for each $N$. The number of samples used to plot Figs. 9(b) and 10 are given in parentheses for $N = 1000$ and 32000. See the text for details.

| $N$    | #samples   |
|--------|------------|
| 1000   | 1000 (10000) |
| 2000   | 1000       |
| 4000   | 1000       |
| 8000   | 500        |
| 32000  | 498 (10000) |
| 128000 | 495        |

cut at a distance $r_{\text{cut},ab} = 2.5\sigma_{ab}$. However, the discontinuity of the pair force $v'_{0,ab}$ at the cutoff distance strongly affects the low-frequency modes [38]. Thus, we modified Eq. (1) as

$$v_{ab}(r) = \begin{cases} v_{0,ab}(r) - v_{0,ab}(r_{\text{cut},ab}) - (r - r_{\text{cut},ab})v'_{0,ab}(r_{\text{cut},ab}) & (r < r_{\text{cut},ab}), \\ 0 & (r > r_{\text{cut},ab}) \end{cases}. \quad (2)$$

Using Eq. (2), we carried out MD simulations. First, the system was equilibrated at a sufficiently high temperature of $T = 2.0$ in the NVT ensemble for a time interval of $t = 500$, and then we performed a simulation in the NVE ensemble for the same time interval. Independent liquid configurations were sampled in the subsequent NVE simulation. The time interval between two configurations is $t = 50$.

Starting from these liquid states, we performed simulations of the steepest descent dynamics [6, 7]; namely, we numerically solved the following equation of motion:

$$\gamma \frac{dr_i}{dt} = -\frac{\partial V}{\partial r_i}, \quad (3)$$

where $r_i$ is the position of particle $i$, and $V = \sum_{i<j} v_{ab}(r_{ij})$ is the total potential energy. The ratio of the discretized time step to the friction coefficient $\Delta t/\gamma$ was set to 0.0001. The process stops at the time that the system reaches an inherent structure. The number of configurations used for each $N$ is listed in Tab. I. To plot Figs 9(b) and 10, we needed a particularly large sample size and the number of configurations is given in the parentheses in Tab. I.

We have performed a normal mode analysis [39] of instantaneous configurations during the steepest descent dynamics. The dynamical matrix, being the Hessian matrix of the total potential $V$, was diagonalized, to obtain its eigenvalues $\lambda_\alpha$ and eigenvectors $e_\alpha = (e_{\alpha,1}, \ldots, e_{\alpha,N})$, where $\alpha = 1, 2, \ldots, 3N - 3$. The three modes corresponding to the global translations were excluded. The eigenvalues are sorted in ascending order: $\lambda_1 < \lambda_2 < \cdots < \lambda_{3N-3}$, and the eigenvectors are normalized as $|e_\alpha| = 1$.

### III. RESULTS

#### A. Mean squared force and statistics of unstable modes

Before considering the averaged quantities, we show raw data for $N = 2000$ in Fig. 1. Figure 1(a) shows the mean squared force $W := (1/N) \sum_{i=1}^{N} f_i^2$ as a function of the time and Fig. 1(b) is a parametric plot of $W$ versus the fraction of the unstable modes

$$f_u := \int_{-\infty}^{0} d\lambda D(\lambda), \quad (4)$$

where $D(\lambda)$ is the spectrum, or the density of states. In Fig. 1(c) and (d), we present the same plots for a different sample, to be compared in the next paragraph. For the steepest descent dynamics in Eq. (3), the mean squared force is translated into the potential energy as follows:

$$W = \frac{1}{N} \sum_{i=1}^{N} f_i^2 = -\frac{\gamma}{N} \sum_{i=1}^{N} \frac{\partial V}{\partial r_i} \frac{dr_i}{dt} = \frac{\gamma}{N} \left| \frac{dV}{dt} \right|. \quad (5)$$
Note that the time derivative of the potential energy is always negative. Because $W$ vanishes at all inherent structures, it is more useful than the potential energy itself and has been a central focus of recent related studies [6, 7]. In addition to the steepest descent dynamics, this function is often called the $W$-function and has been used to locate saddle points in the potential energy landscape [40]. Although the data is noisy, one can see that the mean squared force first exhibits an almost algebraic decay followed by a precipitous drop at a particular time point. The system finds an inherent structure and the dynamics stops immediately after the drop. Following this behavior, the fraction of the unstable modes decreases with decreasing $W$. However, the decay is not monotonic and the value of $f_u$ oscillates randomly before the mean squared force drops and the unstable modes disappear.

Figure 1 indicates the difficulty in carrying out an ensemble average; because of the strong fluctuations of the time at which $W$ drops, the ensemble average at a fixed time will suffer from significant uncertainties in the late stage. To avoid this issue, we averaged the data at fixed $W$. During simulations, we have picked up a configuration every time $W$ reaches a specific value and have averaged observables over the configurations with the same $W$.

Figure 2 shows the same data as in Fig. 1, which are averaged with $W$ fixed. The mean squared force in Fig. 2(a) decays algebraically and drops more mildly than in Fig. 1(a) and (c) at a time depending on the system size $N$. The algebraic decay in the middle stage is fitted by a power law $W \propto t^{-1.7}$ [6, 7], as shown by the solid line. Note that this is twice the exponent $\beta = 0.85$ reported in Refs. [6, 7] because $\sqrt{W}$ was measured in these studies. In addition, Fig. 2(b) shows another power law, $W \propto f_u^2$, which holds down to $W \sim 10^{-8}$. Physically, this is quite reasonable. The system approaches a stable inherent structure $W \rightarrow 0$ as the fraction of the unstable modes $f_u$ becomes smaller. Related to these results, we note that similar scaling relations were reported for the spherical Sherrington-Kirkpatrick (SK) model [4]. This simple mean-field model does not show a glass transition, but it is characterized by slow relaxation similar to that of the spherical $p$-spin model with $p > 2$, which has a spin-glass phase. In Ref. [4], it was shown that $W \sim t^{-2}$ and $f_u \sim t^{-3/2}$, which leads to $W \sim f_u^{4/3}$ in the spherical SK model.

From these two functions, it is possible to obtain the dependence of the potential energy $V$ on $f_u$ [1, 16, 18, 41].
FIG. 2. (a) Time evolution of mean squared force. The solid line represents a power law $W \propto t^{-1.7}$ [6, 7]. Note that $W$ is controlled in this plot. See the text for details. The errors are smaller than the symbols and thus not shown. (b) Mean squared force versus the fraction of the unstable modes. The solid line represents a power law $W \propto f_u^2$.

The $f_u$-derivative of the potential energy, $\partial V/\partial f_u$, gives an estimation of the height of the potential energy barrier compared to stationary points and has been well studied in terms of saddle modes [1, 16, 18, 41]. Equation (5) gives

$$-\frac{d(V/N)}{dt} = \frac{1}{\gamma} W \propto f_u^2,$$

which leads us to find

$$f_u \propto \sqrt{W(t)},$$

$$\Delta V/N := V/N - \lim_{t \to \infty} V/N \propto \int_t^\infty dt' W(t').$$

If one assumes $W(t) \sim t^{-a}$, these are

$$f_u \sim t^{-a/2},$$

$$\Delta V/N \sim t^{-a+1}.$$

Thus, $(1/N)\partial V/\partial f_u \to \infty$ as $t \to \infty$ for $a < 2$ while $(1/N)\partial V/\partial f_u \to 0$ for $a > 2$. The latter result is also reproduced if, for example, one assumes $W(t) \sim e^{-t}$. As shown in Fig. 2(a), the mean squared force finally decays to zero much faster than $t^{-1.7}$. Thus, the latter would be the case. The fact that the $f_u$-derivative of the potential energy tends to zero in the final stage of the dynamics indicates that the potential energy landscape becomes flatter as the system approaches an inherent structure [1]. Although the specific exponents are different, the spherical SK model [4] shares an important property: $(1/N)\partial V/\partial f_u \to 0$ as $t \to \infty$.

The spectrum is a fundamental quantity for characterizing the stationary points of the potential energy landscape. Though Figs. 1 and 2 show an integrated version of it, $f_u$, direct investigation of the spectrum gives more insight into the dynamics. Figure 3 shows the unstable part of the spectrum at several values of $W$. At each value of $W$, the spectrum hardly depends on the system size and shows an exponential tail. To explore these tails, we have fitted an exponential function $\propto e^{\lambda/\lambda_0}$, where the constant $\lambda_0$ can be viewed as a characteristic energy scale of unstable modes, to the spectrum of $N = 8000$, as shown in Fig. 4. Figure 4(a) shows the spectrum of $N = 8000$ for $\log_{10} W = -4.5, -4.0, -3.5, -3.0, -2.5, -2.0, \text{ and } -1.5$. The spectrum of $\log_{10} W = -1.5$ is fitted with an exponential function, as shown by the solid line. Likewise, we performed the fitting of all the data in Fig. 4(a), and the estimated values of $\lambda_0$ are presented in Fig. 4(b) as a function of $W$. These data agree well with a power law $\lambda_0 \propto W^{0.22}$. The existence of the simple scaling relations as shown in Fig. 2(b) and Fig. 4 shows that the mean squared force $W$ is a useful parameter to characterize the system during the steepest descent dynamics.

The exponential tail of the spectrum is characteristic of localized states. This motivated us to perform a finite-size scaling analysis of the participation ratio [42-44]

$$P_\alpha := \frac{1}{N} \left( \sum_{i=1}^{N} |e_{\alpha,i}|^4 \right)^{-1},$$

(11)
which measures the degree of localization of a given mode $\alpha$. If all particles vibrate equally, $P_\alpha = 1$ and if only a single particle vibrates, $P_\alpha = 1/N$. Thus, $NP_\alpha$ gives the number of particles participating in the mode, enabling us to determine the mobility edge $\lambda_e$, which separates localized and delocalized modes. Specifically, the average participation ratio $P := \langle P_\alpha \rangle_\lambda$ at $\lambda$ for delocalized modes grows at least linearly with $L$, where $L$ is the linear size of the system, whereas, for localized modes, it is independent of $L$ [40, 45]. Thus, the mobility edge, the border between these two species of modes, is defined as the fixed point of $NP/L$, which is proportional to $NP/N^{1/3} = N^{2/3}P$ if the density is fixed in three dimensions, by changing $N$ [40, 45]. Figure 5 shows $N^{2/3}P$ as a function of the eigenvalues.
\( \lambda \) at the same values of \( W \) as in Fig. 3. The mobility edges, estimated as the mean intersection of pairs of data for different \( N \), are shown by the solid lines in panels (a), (b), (c), and (d). It can be seen that the mobility edge converges to zero, which means that localized modes dominate the late stage of the dynamics. Note that the mobility edge cannot be positive because of the phonons that are always delocalized. To further corroborate this result, it is helpful to compute the level spacing distribution \([17, 19, 40]\), although the number of samples used in this study is too small to obtain precise data, particularly for small \( W \).

B. The most unstable mode

In the previous subsection, we found that unstable localized modes dominate the steepest descent dynamics. Here we focus on the most unstable mode, which is the most localized. First, we consider the decay profile \( d(r) \) \([22, 46]\). It quantifies how an eigenvector decays from its localized core, defined as the particle with the largest norm; for the detailed definition, see Refs. \([22, 46]\). Figure 6 shows the decay profile of the most unstable modes for \( N = 128000 \) at several values of \( W \). We present the corresponding data for \( N = 8000 \) and 32000 in Appendix B to examine the system size dependence. Figure 6(a) shows the decay profile averaged over the samples. In the early stage of the dynamics, or the large-\( W \) regime, it decays rapidly with distance \( r \) from the core. This behavior resembles the unstable localized modes found at saddle points \([30]\). As \( W \) decreases with time, this decay becomes slower. In the case of the lowest-frequency quasi-localized mode of an inherent structure, the decay profile exhibits a power law, \( d(r) \propto r^{-2} \); the same law describes the response of an elastic medium to a local dipolar force \([47, 48]\). Thus, we plot \( d(r)r^2 \) versus \( r \) in Fig. 6(b). One can see that the decay profile becomes flatter as \( W \) decreases far from the origin, which means that the unstable instantaneous modes during the steepest descent dynamics indeed converge to the quasi-localized modes.

In contrast, \( d(r)r^2 \) appears to decay exponentially for large values of \( W \) far from the origin. Thus, we fitted an exponential function \( \propto e^{-r/r_*} \) to the data in Fig. 6(b) and plotted the logarithm of the characteristic length \( \ln r_* \) as a function of the mean squared force \( W \) for \( N = 8000, 32000, \) and 128000 in Fig. 7. The data depend on the system size, but appear to converge rapidly as \( N \to \infty \). For all \( N \), the characteristic length \( r_* \) diverges as \( W \to 0 \), which is consistent with the results in Fig. 6, and it obeys a stretched exponential law \( \ln r_* \propto W^{-0.11} \) over four decades in
FIG. 6. (a) The decay profile for \( N = 128000 \). The values of \( W \) are \( \log_{10} W = -7, -6, -5, -4, -3, \) and \(-2 \) from top to bottom, as indicated by the colors and arrow. The errors are smaller than the symbols and thus not shown. (b) The decay profile multiplied by \( r^2 \). The data of \( \log_{10} W = -6, -5, -4, -3, \) and \(-2 \) are fitted with exponential functions as shown by the solid lines.

FIG. 7. The logarithm of the characteristic length \( \ln r_* \) as a function of the mean squared force \( W \). The solid line represents a stretched exponential law \( \ln r_* \propto W^{-0.11} \).

\( W \) for \( N = 128000 \). This non-trivial functional form has not been reported in the context of relaxation dynamics, at least to our knowledge, and its underlying mechanism remains to be explored in future work.

Considering the spatial distribution of the eigenenergies of unstable modes, it is possible to investigate the local mechanical stability. Thus, we next study the energy profile \( \Lambda(r) \) [30, 49, 50] of the most unstable mode. The energy profile is a function of the distance \( r \) from the particle that has the most negative local harmonic energy \( \delta E_i \) for a given mode [49]. Such a particle often has the largest norm of the mode and thus it can also be understood as a core. The energy profile \( \Lambda(r) \) is the partial harmonic energy of the mode integrated from the core to the distance \( r \). In other words, \( \Lambda(r) \) is the harmonic energy that the mode would have if the system were cut at a distance \( r \) from the core, and thus \( \Lambda(r \to \infty) \) converges to the eigenvalue of the mode, see Refs. [30, 49, 50] for the complete definition. Figure 8 shows the results for \( N = 128000 \) at the same values of \( W \) as in Fig. 6. In Fig. 8(a), the energy profile of the most unstable modes averaged over the samples is plotted. The profile shifts upward as a whole with decreasing \( W \). Roughly speaking, its overall shape does not change within this range of \( W \). It starts from a negative value by definition, reaches the minimum \( \Lambda_{\text{min}} \), shows an upturn, and finally converges to the average eigenenergy. The length at which the energy profile reaches its minimum \( \Lambda_{\text{min}} \) is a useful definition of the localization length. Figure 8(a) indicates that the localization length of the most unstable mode is determined in the early stage of the dynamics. This behavior should be compared with the results in Ref. [6], that is, the size of the hot spots increases with time.
Probably non-affine displacements in a hot spot involve surrounding particles and thus the size appears to grow in the late stage because the surrounding far field develops as shown in Fig. 6. In contrast to this short-length behavior, one can see several important time dependences far from the origin by looking into the detailed shape of the energy profile. Figure 8(b) shows the energy profile normalized by the minimum value |Λ\text{min}|. In panel (b), the data are normalized by the minima Λ\text{min}, and in panel (c), the final values are subtracted.

![Figure 8](image)

**FIG. 8.** (a) The energy profile for N = 128000. The values of W are \( \log_{10} W = -7, -6, -5, -4, -3, \) and \(-2\) from top to bottom, as indicated by the colors and arrow. In panel (b), the data are normalized by the minima Λ\text{min}, and in panel (c), the final values are subtracted.

![Figure 9](image)

**FIG. 9.** (a) Minimum of the energy profile as a function of the mean squared force. The solid line represents a power law \(-Λ\text{min} \propto W^{0.14}\). The final value \(-Λ_\infty\) is also plotted by the dashed lines. (b) The PDF of Λ\text{min} shown by the black squares for \(N = 1000\). The values of W are \( \log_{10} W = -9, -7.5, -6, -4.5, -3, \) and \(-1.5\) from right to left as indicated by the colors and arrow. The shifted spectrum \(100D(λ + 15.5)\) for \(N = 1000\) and \(\log_{10} W = -1.5\) is also shown by the pink circles.

Finally, we analyze the characteristic energy scale of the core Λ\text{min} in detail. In Fig. 9(a), \(-Λ\text{min}\) is plotted as a function of W. For comparison, the final value \(Λ_\infty := \lim_{r \to \infty} Λ(r)\), which is equal to the average eigenvalue, is also plotted by the dashed lines. For large values of W, the data show a power law \(-Λ\text{min} \propto W^{0.14}\). This decay is much slower than that of \(Λ_\infty\) and thus, the core stabilizes more slowly than the overall mode. For small values of W, it appears to converge to a finite value depending on the system size. Furthermore, Fig. 9(b) shows the probability density function (PDF) of Λ\text{min} for \(N = 1000\) by the black squares. The values of W are \( \log_{10} W = -9, -7.5, -6, -4.5, -3, \) and \(-1.5\). For large values of W, the function shows a clear exponential tail and the explanation is obvious: the far fields provide only minor contributions and the cores determine the total eigenenergies of the localized modes. As a proof of this, the shifted spectrum \(100D(λ + 15.5)\) for \(\log_{10} W = -1.5\), indicated by the pink circles, almost
collapses onto the corresponding data. For small values of $W$, the PDF still appears to decay exponentially. If this is the case, it should be contrasted with the PDF of the eigenvalues of the lowest-frequency quasi-localized modes of inherent structures, which perfectly obeys a Weibull distribution with a power law $\propto \omega^4$, where $\omega = \sqrt{\lambda}$ is the frequency, in its left tail [22–24].

To precisely determine the functional form of the PDF of the characteristic energy scale of cores $\Lambda_{\text{min}}$ for small $W$, we computed its cumulative distribution function (CDF), which is free from the binning error unlike the PDF. Figure 10 shows the CDF of $\Lambda_{\text{min}}$ for (a) $N = 1000$ and (b) $N = 32000$. The values of $W$ are $10^{-9}$ and $10^{-16}$, as indicated by the arrows and colors. These are very small and the system of $W = 10^{-16}$ has no unstable modes, i.e., it is an inherent structure. For any $N$ and $W$, the CDF exhibits an exponential tail. Thus, the distribution of $\Lambda_{\text{min}}$ remains exponential, whereas that of the eigenenergy $\lambda = \Lambda_\infty$ gradually develops an algebraic tail.

IV. SUMMARY AND CONCLUSION

In this study, we have numerically investigated the steepest descent dynamics as one of the simplest models of quenching. In particular, we have focused on unstable instantaneous normal modes during the dynamics and their relation with the mean squared force $W$, which is essential to characterize the dynamics [6, 7]. In Sec. III A, we have examined the statistics of the unstable modes and found the following two main results. First, we found a power law between the mean squared force and the fraction of the unstable modes, $W \propto f_u^2$. This leads to the vanishing $f_u$-derivative of the total potential energy in combination with the time dependence of the mean squared force. This fact indicates that the paths in the potential energy landscape passed by quenched liquids become flatter as the dynamics proceeds, which supports the marginal stability of glasses [31–33]. Second, the spectrum of unstable normal modes exhibits an exponential tail, a known characteristic of localized modes. Performing a finite-size scaling analysis, we have located the mobility edge, which appears to converge to zero. Thus, in the late stage of the dynamics, the motion of the particles is mainly governed by localized modes. This finding is consistent with the previous studies [6, 7].

In Sec III B, we have focused on the most unstable mode of an instantaneous configuration and investigated how its structure develops during the relaxation process. By computing the decay and energy profiles of the modes, we have found that the core length is already determined in the early stage of the dynamics. In contrast, the surrounding far field significantly develops during dynamics with a non-trivial stretched exponential law. Moreover, the core energy distribution $\Lambda_{\text{min}}$ remains exponential in contrast to the power-law distribution of the smallest eigenvalues of inherent structures [22–24]. This means that the cores of the quasi-localized modes are hot spots left to be unstable during the dynamics, in which the overall system rapidly stabilizes.

To the best of our knowledge, the power-law relations and the stretched exponential law reported in this paper have not been observed in previous studies of the relaxation dynamics and also no theory predicts them. Therefore, it would be worth investigating the extent to which our findings are universal by, for example, using different models.

---

Three out of 10000 configurations of $N = 32000$ were stuck at saddles of $W \sim 10^{-15}, 10^{-12}$. Because each saddle configuration had a single unstable mode, we slightly pushed the configuration along the mode and then the system reached $W = 10^{-16}$. 

or varying the initial temperature. It would also be interesting to study whether several theoretical models such as the elastoplastic model discussed in Refs. [51–54] can reproduce them.

ACKNOWLEDGMENTS

We thank P. Chaudhuri, D. Coslovich, K. Hukushima, Y. Nishikawa, and N. Oyama for useful exchanges. This work was supported by JSPS KAKENHI Grant Numbers 18H05225, 19H01812, 19J20036, 20H00128, 20H01868, and 21J10021. Numerical simulations were performed using the Fujitsu PRIMERGY CX400M1/CX2550M5 (Oakbridge-CX) in the Information Technology Center, The University of Tokyo.

Appendix A: The number of cores

In this section, we show data that suggest that on average, there is a single core in the most unstable mode of each configuration. By the term “core”, we designate the particle with the most negative harmonic energy $\delta E_i$. In Fig. 11, the average distance from the core $r_i$ is plotted against the average harmonic energy $\delta E_i$ for several values of $W$. Each panel shows only the data for the ten particles with the most negative harmonic energies for each $W$. This figure indicates that these unstable particles are close to the core particles and thus, it is unlikely that they constitute other independent cores.

![Figure 11: The average distance from the core versus the harmonic energy for (a) $N = 8000$, (b) 32000, and (c) 128000. The error bar indicates the standard deviation of each data point and not the error itself. The values of $W$ are $\log_{10} W = -2, -3, -4, -5, -6, \text{ and } -7$ indicated by the colors.](image)

Appendix B: The system size dependence

Here, we show the decay and energy profiles for $N = 8000$ and 32000 for comparison with Figs. 6 and 8, respectively. From these plots, it is evident that the data do not qualitatively depend on the system size.
FIG. 12. (a) The decay profile for \( N = 8000 \). The values of \( W \) are \( \log_{10} W = -7, -6, -5, -4, -3, \) and \(-2\), indicated by the color and arrow. The errors are smaller than the symbols and thus not shown. (b) The decay profile multiplied by \( r^2 \). The data of \( \log_{10} W = -4, -3, \) and \(-2\) are fitted with exponential functions, as shown by the solid lines.

FIG. 13. (a) The decay profile for \( N = 32000 \). The values of \( W \) are \( \log_{10} W = -7, -6, -5, -4, -3, \) and \(-2\), indicated by the color and arrow. The errors are smaller than the symbols and thus not shown. (b) The decay profile multiplied by \( r^2 \). The data of \( \log_{10} W = -6, -5, -4, -3, \) and \(-2\) are fitted with exponential functions as shown by the solid lines.

FIG. 14. (a) The energy profile for \( N = 8000 \). The values of \( W \) are \( \log_{10} W = -7, -6, -5, -4, -3, \) and \(-2\), indicated by the color and arrow. In panel (b), the data are normalized by the minima \( \Lambda_{\text{min}} \) and in panel (c), the final values are subtracted.
Appendix C: Asymptotic behavior of the energy profile

Here, we discuss the long-distance behavior of the energy profile in an infinite stable system. Namely, we consider the energy profile of the lowest-frequency quasi-localized mode. To this end, we assume that the shape of the quasi-localized modes far from the cores is the same as the response of an elastic continuum to a local dipolar force, which is supported by recent studies [47, 48]. By approximating the system as a spring network, which is the simplest elastic continuum, the energy profile is proportional to the spatial integral of the first response function defined in Ref. [55]. Thus, the energy profile obeys $|\Delta \Lambda(r)| := |\Lambda(r) - \Lambda_\infty| \sim r^{-3}$, where $\Lambda_\infty := \lim_{r \to \infty} \Lambda(r) \sim r^{-3}$, and $r_{\text{core}}$ is the size of the core. This scaling relation is not inconsistent with the data for $N = 128000$ and $W = 10^{-7}$ in Fig. 16, although they are noisy because of the finite-size effect. We also note that the modes shown in Fig. 16 are still slightly unstable.

Appendix D: Low-temperature results

Here, we present the results at low temperatures, which correspond to those in Fig. 23. To sample the equilibrium configurations at low temperatures, we used the replica exchange method [34, 35]. In our MD simulations, 18 replicas were used and each replica corresponded to a temperature ranging from 0.7800 to 0.3971. The MD simulations were

---

3 In this section, we set the total number density to $\rho = 1.204$. Thus, the linear size of the system is $L \approx 9.3999$, which is slightly different from the original value $L = 9.4$ in Refs. [56, 57].
performed in the NVT ensemble using the Nosé-Hoover thermostat \cite{56} with a time step of 0.005. Exchange trials were performed every 2800 MD steps using the Metropolis criterion. We checked that all replicas were equilibrated after 350000 exchange trials and sampling was performed every 10000 trials thereafter. All calculations were carried out in an in-house code that uses MPI to handle parallel computations. Because of the high computational cost, we were only able to prepare 45 samples of $N = 1000$.

Using these low-temperature configurations, we performed the same analysis as described in Sec. III A. Figure 17 corresponds to Fig. 2. The solid line in Fig. 17(a) represents a power law $W \propto t^{-2.5}$, which was observed in Ref. \cite{7}. This is consistent with the data, although the power-law region is small because of the small system size.

It is possible to generalize the discussion of $\partial \Delta V/\partial f_u$ in Sec. III A. Setting $W \sim t^{-a}$ and $W \sim f_u^b$, where $a > 1$ and $b > 0$, yields

\begin{equation}
 f_u \sim t^{-a/b},
\end{equation}

\begin{equation}
 \Delta V/N \sim t^{-a+1}.
\end{equation}

Thus, one obtains

\begin{equation}
 \frac{1}{N} \frac{\partial \Delta V}{\partial f_u} \sim t^{-a+1+a/b}.
\end{equation}

As a result, if $-a + 1 + a/b < 0$; namely, $a/(a-1) < b$, this derivative vanishes as $t \to \infty$, which is observed in the case of $T = 2.0$ in Sec. III A and in the spherical SK model \cite{4}. According to Ref. \cite{7}, $a = 2.5$ is the maximum value in the lowest-temperature regime and in this case, the above inequality becomes $5/3 < b$. As shown in Fig. 17(b), low-temperature data for $f_u$ roughly follow the same power law $W \propto f_u^2$ as in the case of $T = 2.0$. Thus, this inequality always seems to hold. Larger systems are necessary to establish this result.

![Figure 17](image.png)

**FIG. 17.** (a) Time evolution of mean squared force. The solid line represents a power law $W \propto t^{-2.5}$. (b) Mean squared force versus fraction of the unstable modes. The solid line represents a power law $W \propto f_u^2$.
