Origin of reversible and irreversible atomic-scale rearrangements in a model 2D network glass

Firaz Ebrahem∗, Franz Bamer, and Bernd Markert
Institute of General Mechanics, RWTH Aachen University, 52062 Aachen, Germany
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In this contribution, we investigate the fundamental mechanism of plasticity in a model 2D network glass. The glass is generated using a Monte Carlo bond-switching algorithm and subjected to athermal simple shear deformation, followed by subsequent unloading at selected deformation states. This enables us to investigate the topological origin of reversible and irreversible atomic-scale rearrangements. It is shown that some events that are triggered during loading recover during unloading, while some do not. Thus, two kinds of elementary plastic events are observed, which can be linked to the network topology of the model glass.

It is generally accepted that plastic deformation in glasses is triggered by atomic-scale rearrangements, referred to as shear transformations [1][3]. These inelastic events are localised in space and they involve only a small number of atoms. In recent years, much effort has been devoted to achieving a deeper understanding of the atomic phenomena associated with the plastic deformation of glasses. In this regard, numerical studies have extensively been performed on two-dimensional binary glass forming systems [4][11]. In order to investigate how pure structural disorder correlates with plastic response, the athermal quasistatic (AQS) deformation protocol has proved useful. The glass system is subjected to small shear increments, each followed by energy minimisation. Thus, the AQS protocol is used to explore the potential energy landscape (PEL) [11][13]. The AQS deformation allows the system to overcome the energy barrier to jump from one local minimum of the potential energy to another neighbouring minimum. According to the PEL, the atomic-scale rearrangements can be considered as instabilities of the glass system. The mechanical stability is governed by the Hessian matrix, a matrix of the second-order partial derivatives of the total potential energy of the system $U(r_1, r_2, \ldots, r_N)$, which is a function of the particle positions $\{r\}_N^N$. The elements of the Hessian matrix are given as:

$$H_{ij} = \frac{\partial^2 U(r_1, r_2, \ldots, r_N)}{\partial r_i \partial r_j}.$$  \hspace{1cm} (1)

As long as the system is mechanically stable, the Hessian matrix $H$ is symmetric and positive semi-definite. It has zero eigenvalues associated with the Goldstone modes, while all the remaining eigenvalues are real and positive [14][15]. The plastic instability under applied shear strain $\gamma$ occurs via a saddle-node bifurcation, with the lowest positive eigenvalue $\lambda_{\text{min}}$ going to zero as

$$\lambda_{\text{min}} \propto \sqrt{\gamma_c - \gamma},$$  \hspace{1cm} (2)

where $\gamma_c$ it the critical shear strain at the bifurcation. As the lowest eigenvalue $\lambda_{\text{min}}$ of the Hessian matrix $H$ approaches zero the corresponding eigenfunction localises on the typical quadrupolar-like structure. This quadrupolar symmetry is similar to an Eshelby inclusion embedded in an elastic surrounding [16][19]. It was shown that the non-affine irreversible displacement field at the plastic event scales with the eigenfunction whose eigenvalue vanishes at the instability [12][20][22].

The plastic events are identified from the stress-strain curves during AQS shear deformation of glass samples. The stress-strain curves are made of elastic segments intersected by sudden stress drops, corresponding to the localised rearrangements of a subset of atoms. Shear loading and unloading simulations of metallic glasses revealed that some events are recovered during the unloading process, while other events are not [23][24]. From these loading unloading investigations Falk et al. concluded that in glasses, at least two kinds of plastic events can be identified, as originally proposed by Argon and Shi [25]. The local atomic rearrangements that recover upon unloading have been referred to as local “anelastic” transformations. The other local rearrangements that can not recover upon unloading were identified as “viscoplastic” transformations. However, the fundamental mechanism of these two types of elementary plastic events in glasses is, to this date, not fully resolved. In particular, for network glasses, such as vitreous silica, there is a lack of knowledge about the microscopic deformation mechanism, although these types of glasses can be well defined through their network topology, which is, in two dimensions, governed by corner-sharing SiO$_3$ triangles [26][28].

Thus, in this contribution, we study the origin of reversible and irreversible atomic rearrangements in a model 2D network glass. The model glass is numerically prepared by applying a series of topological flip transformations to an initially hexagonal lattice. The network structure is generated based on statistical data extracted from real 2D silica [29]. We investigate the response of the model glass under AQS simple shear deformation and demonstrate that the two kinds of elementary plastic events (reversible and irreversible) can directly be linked to the network topology of the glass.

In order to generate the model 2D network glass, we

∗ ebrahem@iam.rwth-aachen.de
use a recently developed potential function that has been used for the description of silica glass in a two-dimensional framework \[50, 54\]. Thus, the model glass used in this study meets Zachariasen’s requirements of network glasses consisting of covalently bonded corner-sharing SiO\(_2\) triangles \[55\]. The interatomic interactions are described by a Yukawa-type potential function

\[
U_{ij} = \left(\frac{\sigma_{ij}}{r_{ij}}\right)^{12} + \left(\frac{q_{ij}}{r_{ij}}\right) \exp\left(-\kappa r_{ij}\right). \tag{3}
\]

Hereby, the parameters \(\sigma_{ij}, r_{ij}, q_{ij}\) and \(\kappa\) denote the repulsive strength, the interatomic distance, the ionic charge and the screening parameter, respectively. The parameter values can be taken from Roy et al. \[30\]. This potential function was shown to successfully reproduce both the short-range order and the ring statistics of real 2D silica \[26, 28\]. The potential is dimensionless. Thus, all quantities except the length scales are expressed in dimensionless units. Furthermore, the potential is truncated at 11 Å and shifted in order to ensure that both the potential energy and the corresponding interatomic forces vanish smoothly at the cut-off distance.

To generate the glass samples, we start from the hexagonal lattice, as observed for crystalline 2D silica \[36\]. The hexagonal lattice considered in this study consists of 1350 atoms (540 Si and 810 O atoms) that form 270 six-membered rings in a periodic simulation cell with a size of about 8 × 8 nm\(^2\). The hexagonal lattice is then transformed into the underlying dual lattice that is subjected to a Monte Carlo bond-switching algorithm by applying in-plane flips of selected dual bonds \[26, 28\]. After each flip, the system energy is minimised using a harmonic dual potential describing ring-ring interactions \[37\]. This operation affects a set of four neighbouring rings in the network by increasing the size of two rings and decreasing the size of the other two rings. After a sequence of random flips, the dual is transformed back to the overlying network and minimised using the Yukawa-type potential function \[3\].

Furthermore, in order to avoid local ring clusters of similar sizes in the glass samples, we use the empirical Aboav-Weaire law \[38–40\] that controls the ring neighbourhood statistics:

\[
m_n = \mu (1 - \alpha) + \frac{1}{n} (\mu^2 \alpha + s^2), \tag{4}
\]

where \(m_n\) denotes the mean ring size around a ring of size \(n\), and \(\mu\) and \(s^2\) are the mean and the variance of the ring size distributions, respectively. The ring neighbourhood is then identified by only one Aboav-Weaire parameter \(\alpha_n\). Thus, proper values of \(\alpha\) lead to physically meaningful ring neighbourhood distributions and, therefore, prevent an unphysical clustering effect of small or large rings.

The glass samples obtained using this strategy are then subjected to AQS simple shear loading and unloading, applying Lees-Edwards boundary conditions. The deformation is applied in sufficiently small shear increments of \(\Delta \gamma = 4 \times 10^{-6}\). Each shear increment is followed by energy minimisation with a tolerance of \(10^{-16}\), using the conjugate gradient method. The findings presented in this letter are derived from the shear responses of a set of 20 glass samples. In what follows, one representative sample will be discussed in detail.

Figure 1 shows the stress response of a model 2D network glass under athermal quasistatic simple shear deformation. The black curve corresponds to the simple shear loading up to a strain of about 18 %, where three stress drops \(E_1, E_2\) and \(E_3\) are identified. The red curve corresponds to the stress response of the shear unloading after the event \(E_1\). The blue curve corresponds to the stress response of the shear unloading after the event \(E_2\).
This observation is in line with other atomic-scale studies in Figures 3d and 3e, reveals that this event corresponds to atomic configuration before and after the event $E_1$ orientation is barely influenced. The inspection of the formation of a void and dangling bonds, while the bond the form of a bond-breaking event that results in the to a localised rearrangement in the micro-structure in configuration before the event $E_1$ after the plastic instabilities. For this purpose, the atomic structure of the model 2D network glass before and right through a saddle-node bifurcation [12]. However, we are plastic instability under applied shear strain amorphous solids, both events are clearly identified as Equation 2. Thus, following the deformation theory of approaches zero via a square-root singularity, given in the Hessian matrix $H$ as a function of the shear strain $\gamma$. Figure 2 depicts the evolution of the lowest positive eigenvalue $\lambda_{\text{min}}$ of the Hessian matrix $H$ with increasing strain for the events $E_1$ and $E_2$. From this figure, it can clearly be seen that for both events the lowest eigenvalues $\lambda_{\text{min}}$ approaches zero via a square-root singularity, given in Equation [2]. Thus, following the deformation theory of amorphous solids, both events are clearly identified as plastic instability under applied shear strain $\gamma$, occurring through a saddle-node bifurcation [12]. However, we are still curious about the fundamental difference between the two kinds of elementary plastic events.

In order to further study the observed two kinds of events in the model 2D network glass, we, next, investigate the Hessian matrix $H$ of the system. To this end, the eigenvalues $\lambda_i$ of the Hessian matrix $H$ are calculated as function of the shear strain $\gamma$. The elements of the Hessian matrix $H_{ij}$, given in Equation [1], are computed by use of the finite difference method. We follow the procedure presented in the supplemental material of [11], applying a displacement increment of $10^{-7}$Å. Figure 2a depicts the evolution of the lowest positive eigenvalue $\lambda_{\text{min}}$ of the Hessian matrix $H$ with increasing strain for the events $E_1$ and $E_2$. From this figure, it can clearly be seen that for both events the lowest eigenvalues $\lambda_{\text{min}}$ approaches zero via a square-root singularity, given in Equation [2]. Thus, following the deformation theory of amorphous solids, both events are clearly identified as plastic instability under applied shear strain $\gamma$, occurring through a saddle-node bifurcation [12]. However, we are still curious about the fundamental difference between the two kinds of elementary plastic events.

The picture becomes clear by observing the network structure of the model 2D network glass before and right after the plastic instabilities. For this purpose, the atomic configuration before the event $E_1$ is depicted in Figure 3a and after the event $E_1$ in Figure 3b. From these two figures it becomes clear that the first event $E_1$ corresponds to a localised rearrangement in the micro-structure in the form of a bond-breaking event that results in the formation of a void and dangling bonds, while the bond orientation is barely influenced. The inspection of the atomic configuration before and after the event $E_2$, shown in Figures 3c and 3d, reveals that this event corresponds to a rearrangement in the micro-structure that changes the local network topology, i.e. bonding configuration. Unlike in the case of the event $E_1$, here, no voids or dangling bonds are observed after the plastic deformation. Instead, the bond orientations change to a significant degree. It is observed that the network of the model 2D glass is still fully coordinated, while the local ring neighbourhood and size distribution is changed during this plastic event, as illustrated by the circled area in Figures 3e and 3f.

These intriguing observations explain why the first event $E_1$ is recovered upon unloading, while the second event $E_2$ is not. In case of the event $E_1$, the broken bond retains its orientation when the void is formed. However, it can be reformed during the shear unloading process, such that the initial local ring structure is recovered. In case of event $E_2$, bonds that are broken and new bonds that are formed in the perpendicular direction. The local ring transformation is irreversible and cannot recover upon unloading. The non-affine displacement fields regarding the event $E_1$, shown in Figure 3c, and regarding the event $E_2$, shown in Figure 3e, indicate the localisation of the eigenfunctions associated with the lowest eigenvalues $\lambda_{\text{min}}$ vanishing at the plastic instabilities. The irreversible event $E_2$, observed here, can directly be linked to an experimental study on real 2D silica glass. Huang et al. were able to excite and image atomic rearrangements in 2D silica glass, revealing that plastic deformation is accompanied by local ring transformations [12].

A recent numerical study has reported two elementary plastic events in bulk silica glass undergoing AQS shear deformation, one event associated with a localised rearrangement without any change in the atomic coordination, the other resulting from an observed bond breaking [11]. However, in this study, we show that reversing the simple shear deformation is indispensable to reveal the fundamental mechanisms that underlie elementary plastic events in network glasses. Other studies revealed that the directional covalent bonds between atoms are of particular importance in plasticity of network glasses [13,14]. Our results confirm that the network structure plays a key role in understanding the fundamental mechanism of elementary plastic events in covalent glasses.

In summary, we have investigated the origin of plastic response of a model 2D network glass subjected to athermal quasistatic simple shear deformation. Shear loading of the network glass revealed several stress drops, some that can be recovered and some not, when the shear deformation is reversed right after the stress drops. The events have been identified as the elementary building blocks in glassy solids, showing that, for each event, the lowest eigenvalue of the Hessian matrix approaches zero via a square-root singularity. Careful inspection of the atomic configurations before and after the plastic events revealed their underlying topological origin. During the reversible event, a bond is broken, while retaining its orientation and forming a void in the network. During the shear unloading process, the broken bond has been shown to reform, such that the initial local ring structure
is recovered. In case of the irreversible event, local ring transformations have been observed, while the network remains fully coordinated. Bonds break and new bonds are formed in the perpendicular direction, i.e. the local bond orientations change to a significant degree. This event cannot recover upon unloading. We have shown that the two-dimensional framework of a network glass, composed of rings and ring neighbourhoods, constitutes a promising tool that can provide further insights into the elementary mechanisms of plastic deformation in disordered solids.

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FIG. 3. The atomic configuration before the elementary plastic event $E_1$ in (a), and after $E_1$ in (b). The non-affine displacement field at $E_1$ is depicted in (c). The atomic configuration before the elementary plastic event $E_2$ in (d), and after $E_2$ in (e). The non-affine displacement field at $E_2$ is depicted in (f). The blue spheres represent silicon atoms, while the red spheres represent oxygen atoms. The circled areas illustrate the local changes in the network structure due to plastic deformation. The size of arrows in (c) and (f) is proportional to the magnitude of atomic displacement vectors.