

# Simulation of Plastic Deformation in a Two-Dimensional Atomic Glass by Molecular Dynamics IV

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*Phil. Trans. R. Soc. Lond. A* 1989 **329**, 613-640

doi: 10.1098/rsta.1989.0092

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# SIMULATION OF PLASTIC DEFORMATION IN A TWO-DIMENSIONAL ATOMIC GLASS BY MOLECULAR DYNAMICS IV

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(Communicated by M. F. Ashby, F.R.S. – Received 22 September 1988)

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Plastic deformation in a structurally well-relaxed two-dimensional atomic glass was simulated by a computer molecular dynamics approach. The simulation, which was carried through yielding and to substantial plastic strains, demonstrated that the principal mechanism of plastic strain production is by local partly dilatant shear transformations nucleated preferentially in the boundaries of liquid-like material separating the small quasi-ordered domains that form when the glass is well relaxed. Under imposed forward shear-strain increments, local shear transformations in atomic clusters were found to be mostly in the same direction as the applied stress. There were, however, substantial levels of shear transformations in other random directions, including many opposed to the applied stress. In all instances, however, nucleation of shear transformations reduced the Gibbs free energy monotonically, which is governed largely by the locked-in excess enthalpies of the glassy state. At shear strains above 15%, localization of shear into bands was observed to begin. This steadily intensified and formed well-defined sharp shear bands into which all the shear strain became concentrated by the end of the simulation at a strain of 27%. A strong correlation was found between the tendency for shear localization and retained shear-induced dilatation.

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## 1. INTRODUCTION

Amorphous solids make up a large group ranging from intrinsically brittle inorganic ones, such as the familiar oxide glasses, to glassy metallic alloys and polymers, which exhibit extensive ductility in the unaged form and are embrittled when aged. Although it is very difficult to initiate plastic flow in the intrinsically brittle family of glasses short of superposing very large components of pressure to suppress fracture, plastic flow to large strains is readily achievable in most unaged glassy metallic alloys and polymers.

The mechanisms of deformation in these materials have long been a source of controversy. While some investigators have sought to explain the plasticity of glasses by generalized dislocations (Gilman 1969; Li 1976), others have attributed it to the same cluster relaxations that give rise to viscous behaviour (Orowan 1951; Spaepen 1977; Argon 1979). As drawing conclusions from observations of external features such as shear localization zones can be risky, and because the usual powerful methods of direct observations of defects based on diffraction contrast are inoperative, simulation of the deformation by various techniques have proved to be most fruitful. In one of the first such attempts, Argon and co-workers (Argon & Kuo 1979; Argon & Shi 1982) have performed shearing experiments on disordered Bragg bubble rafts as two-dimensional analogues of a real atomic glass, and analysed the kinematics and energies of this deformation by means of an inter-bubble pair potential (Shi & Argon 1982). These analogue simulations have demonstrated convincingly that the mechanism of plastic flow in simple disordered solids indeed involves very localized atomic cluster relaxations, and that dislocation glide, when present at all, plays only a minor role. These results largely confirm the very limited amount of the earlier three-dimensional computer simulations (Srolovitz *et al.* 1983). Here we present the results of a new two-dimensional computer simulation of the plastic shearing of the same two-component model amorphous solid discussed in the three accompanying communications, to be referred to here as I–III (Deng *et al.* 1989*a, b, c*).

## 2. DETAILS OF SIMULATION

### 2.1. *The simulation cell*

A two-component material composed of equal numbers of Cu and Zr atoms was chosen for the simulation, because single-component materials proved to crystallize too rapidly below the melting point (part I). As visualization of the kinematic details of the local deformation processes were of primary interest, the simulation was carried out in two-dimensional cells subjected to periodic boundary conditions, to avoid spurious end effects. When a preliminary simulation cell containing 144 atoms on which detailed simulations of melting and quenching (parts I and II) had been carried out, gave indications of being too small for the shearing experiments, a larger two-dimensional simulation cell containing 780 atoms was chosen for most of the results to be presented here. This cell, which was nearly perfectly square, contained in its initial crystalline form 26 close-packed rows of 30 atoms. As we have discussed in connection with our simulations for melting, quenching, and structural relaxations, there are certain limitations of two-dimensional models when compared with three-dimensional models. Nevertheless, on the basis of our findings, if two-dimensional cells containing 780 atoms are necessary to have minimal border effects and constraints, then a proper three-dimensional model should have over  $2 \times 10^4$  atoms. This would make the simulations themselves, as well as

the graphical representation and analysis of the deformation, exceedingly costly. We are of the opinion that the results of our two-dimensional simulation have captured the most important qualitative details, and have further given quantitative measures that compare quite well with experimental information on amorphous metallic alloys. Thus, the majority of the results that we present relate to the large 780 atom cell. We have, however, included a number of additional results derived from the smaller simulation cell of only 144 atoms.

The details of the Cu–Zr pair potential, as well as the details of the procedures in carrying out the molecular dynamics simulation with it were presented in part I, which also contains nomenclature, and normalization quantities. The most important of these normalization quantities are:  $E_0$  ( $= 0.15$  eV), the binding energy for the Cu–Cu pair potential;  $r_0$  ( $= 2.556$  Å†), the critical atomic separation in the Cu–Cu potential where the potential becomes zero; for two-dimensional stresses,  $E_0/r_0^2$  ( $= 0.367$  N m<sup>-1</sup>); for time ( $r_0^2 m_0/E_0$ )<sup>1/2</sup>, the fundamental atomic period for Cu ( $= 5.38 \times 10^{-13}$  s), where  $m_0 = 10.63 \times 10^{-23}$  g is the atomic mass of Cu, and for temperatures,  $E_0/k$  ( $= 1740$  K). As the simulation mats are two-dimensional, to obtain meaningful stresses in three-dimensions the non-dimensional stress components and pressures must be multiplied by  $E_0/r_0^3$  ( $= 1.44$  GPa). In the simulations, the fundamental time steps were taken as  $\frac{1}{20}$  of the above atomic fluctuation period.

## 2.2. State parameters and inelastic strains

Of fundamental interest in this simulation are the forms of inelastic strain production and the local environments in which increments of inelastic strain develop under externally-imposed conditions of stress or cell distortion. The most meaningful state parameters that describe the local conditions have been defined in part II, in connection with the topological features of isothermal structural relaxations obtained in a preparatory simulation to the present one of plastic deformation. These local parameters of state are:

- (1) the volume per atom  $\Omega(i)$  (the area of the Voronoi polygon constructed around each atom);
- (2) the free volume  $v_f(i)$  of an atomic site (i.e., the excess volume per atom averaged over the atom and its immediate neighbours – over and above the average volume per atom in the entire simulation field);
- (3) the atomic level stress tensor  $\sigma_{\alpha\beta}(i)$  (defined for each site by the procedure of Born & Huang (1954));
- (4) atomic level elastic constants  $C_{\alpha\beta\gamma\delta}(i)$  (representing the local stiffnesses of each atomic site);
- (5) the net atomic site distortion parameter  $w'(i)$  defined as the ratio of the perimeter of a Voronoi polygon of a site to the circumference of a circle having the same area. Of the atomic level stresses, two scalar invariants are of particular interest: the atomic site two-dimensional pressure, and the maximum (Mohr circle) shear stress.

When the simulation cell is deformed by imposed shear increments, the local internal increments of deformation are of interest. Most investigators who have performed similar simulations have provided only atomic displacement fields, which in our opinion offer, at best, only a confusing image of the local processes, as they need a reference origin, and are not free of rigid body rotations, which are of no fundamental importance. For that reason, Argon and

†  $1 \text{ \AA} = 10^{-10} \text{ m}$ .

co-workers (Argon & Kuo 1979; Argon & Shi 1982) have given the local deformations as atomic site shear-strain increments and dilatations, which need no reference origin and are free of rigid body rotations. The tensor components of the strain increments have been defined as weighted area integrals of the local deformation gradients as follows:

$$\Delta\epsilon_{\alpha\beta}(i, \Delta t) = \frac{1}{2s_i} \int_{s_i} \left( \frac{\partial \Delta u_\alpha}{\partial \beta} + \frac{\partial \Delta u_\beta}{\partial \alpha} \right) ds \quad (1)$$

$$= \frac{1}{2s_i} \sum_{j=1}^n \left[ \frac{\Delta u_\alpha(j) - \Delta u_\alpha(i)}{\Delta \beta} + \frac{\Delta u_\beta(j) - \Delta u_\beta(i)}{\Delta \alpha} \right] \Delta s_j, \quad (1a)$$

where  $\Delta u_\alpha(j)$  and  $\Delta u_\alpha(i)$  are the displacement increments for atoms  $j$  and  $i$  in the  $\alpha$  direction occurring in the time increment between  $t$  and  $t + \Delta t$ , i.e. between the start and finish of an imposed strain increment on the simulation cell;  $\Delta \alpha$  and  $\Delta \beta$  are initial relative separations of atoms  $j$  and  $i$  in the  $\alpha$  and  $\beta$  directions at time  $t$ . The term  $\Delta s_j$  represents the triangular area of the Voronoi polygon (VP) of atom  $i$  that views the neighbouring atom  $j$ ;  $s_i$  is the total area of the VP of atom  $i$ , and the sum on  $j$  is over all neighbours of  $i$  that enter into to description of the VP of the atom  $i$ . These definitions of strain increments can also be transformed by the divergence theorem into contour integrals around the periphery  $l_i$  of the VP of each atom  $i$ , when they become (in  $xy$  coordinates):

$$\Delta\epsilon_{xy}(i, \Delta t) = \frac{1}{2s_i} \left( -\oint_{l_i} \Delta u_x dx + \oint_{l_i} \Delta u_y dy \right) \quad (2)$$

$$= \frac{1}{2s_i} \sum_{j=1}^n \left[ -\frac{(\Delta u_x(j) + \Delta u_x(i))}{2} \Delta x_j + \frac{(\Delta u_y(j) + \Delta u_y(i))}{2} \Delta y_j \right], \quad (2a)$$

where  $\Delta u_x(j)$  and  $\Delta u_x(i)$  again represent the displacement increments for atoms  $j$  and  $i$  in the  $x$  direction occurring in the time increment between  $t$  and  $t + \Delta t$  between the start and finish of an imposed strain increment on the simulation cell, and  $\Delta x_j$  represents the component in the  $x$  direction of the edge of the VP viewing the neighbouring atom  $j$  of the central atom  $i$ , and again, the sum on  $j$  is taken over all neighbours of  $i$  that enter into the description of the VP of the atom  $i$ .

As with the local atomic level stresses, two scalar invariants of the local strain increments are of special interest, the local dilatation  $\Delta\epsilon(i)$ , and the local increment of the (Mohr circle) maximum shear strain  $\Delta\gamma_{\max}(i)$ , which are defined as:

$$\Delta\epsilon(i) = \Delta\epsilon_{xx}(i) + \Delta\epsilon_{yy}(i), \quad (3)$$

$$\Delta\gamma_{\max}(i) = 2((\Delta\epsilon_{xx}(i) - \Delta\epsilon_{yy}(i))^2 + (\Delta\epsilon_{xy}(i))^2)^{\frac{1}{2}}. \quad (4)$$

In what follows, we will present our results primarily as atomic site information of the current state and increment of inelastic strain.

### 2.3. The shearing simulation

The shearing simulations were carried out on two different sizes of periodic cells. In both instances, the material was a two-dimensional mat composed of equal numbers of Cu and Zr atoms, bound together by three different pair potentials, to take into account the Cu–Cu, Zr–Zr, and Cu–Zr interactions. In both cases, the initial state of the material was a hexagonal

two-dimensional crystal solid solution, in which the individual Cu and Zr atoms were placed randomly. Both mats were melted in the computer and quenched in steps, at an average rate of about  $2 \times 10^9 \text{ K s}^{-1}$ , to a low temperature, followed by a period of structural relaxation of the order of 4000 time steps. The details of the potentials, the melting and quenching simulations, and the descriptions of the molten state, as well as the low temperature quenched states, were described in detail in parts I and II. The small simulation cell, which contained 144 atoms in 12 rows of 12 close-packed atoms, was rectangular in shape, with the edge parallel to the  $x$  axis being 15% longer than that parallel to the  $y$  axis. This cell, on which nearly all the earlier melting, quenching and structural relaxations were performed (reported in parts I and II) proved to give asymmetrical results upon being subjected to simple shear in the  $x$  and the  $y$  directions. To rectify this asymmetry and also to incorporate a larger field to sample more configurations, the second simulation cell was selected to be square in shape, with 30 vertical stacks of close-packed rows containing 26 atoms each, and thus, consisting of 780 atoms on the whole. To check the thermo-physical properties of this material, and relate them to those of the smaller mat, the simulation for the large mat was also started with a melting and quenching sequence identical to those for the smaller mat. The melting temperature and the glass transition temperature for the larger mat were identical to those of the smaller mat. The only difference between the two mats was a constant coefficient of expansion for the larger mat in the crystalline region before melting, rather than the somewhat more curved expansion curve that had been obtained for the small mat. The areal expansion of the large system with changing temperature is shown in figure 1. Figure 2*a, b* show the structure of the large mat

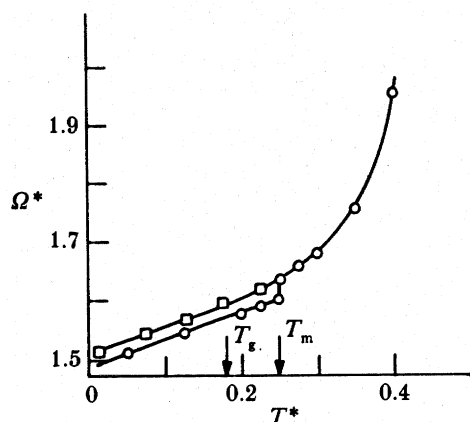


FIGURE 1. Change of volume per atom in simulation cell with temperature in the large system of 780 atoms:  $\circ$ , increasing temperature;  $\square$ , decreasing temperature.

at the melting point of  $T^* = 0.25$  and at  $T^* = 10^{-3}$  respectively, after 4000 time steps (200 fluctuation periods) of structural relaxation. As can be seen, the structure of the relaxed glass is composed of quasi-ordered domains, separated by boundaries of liquid-like material composed of ordered sequences of five- and seven-sided polygons, which in pairs represent edge dislocation cores. Figure 3 shows the radial distribution function (RDF) of the relaxed glass in the large mat before the shearing simulation at  $T^* = 10^{-3}$ . This compares well with the RDF of the relaxed smaller mat, shown in figure 11*d* of part I. (RDF not shown.)

In the shearing simulation, the external shear-strain increments were kept quite small at

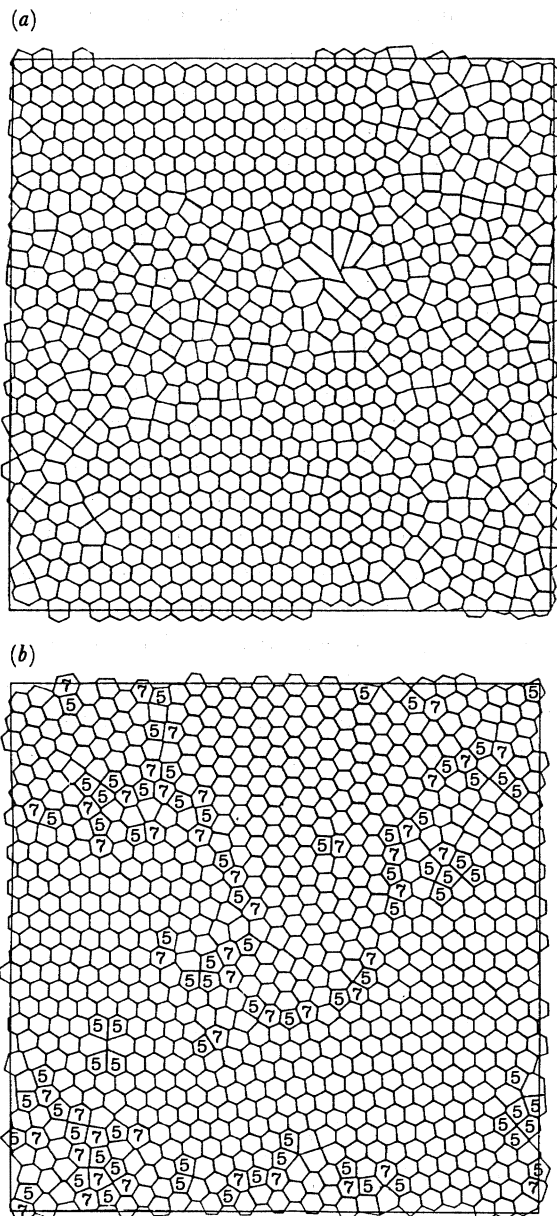


FIGURE 2. (a) Distorted Voronoi polygons in the two-dimensional two-component material at its melting point of  $T^* = 0.25$  on a normalized temperature scale, (b) the atomic structure of the same material shown in (a) after quenching to  $T^* = 10^{-3}$  and relaxing for 4000 time steps. Note the liquid-like material of 5–7-sided polygons separating quasi-ordered regions.

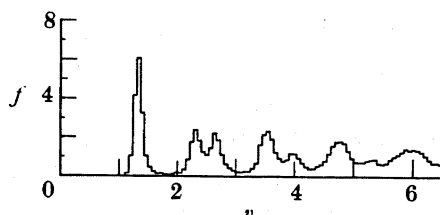


FIGURE 3. Radial distribution function of atom positions in the as-quenched glass shown in figure 2b.

$5 \times 10^{-4}$ , so as to be able to resolve elementary processes in the interior. These shear increments are much smaller than those imposed by Srolovitz *et al.* (1983), or by Maeda & Takeuchi (1982). The shearing was done deliberately at constant volume to stabilize the dilatancy effects producing early shear localization. To monitor the expected effects of shear localization, however, the change in external pressure in response to the imposed shear was calculated for each increment of shear strain. As the starting state of the material for the shearing simulation was obtained by quenching and relaxing a previously melted mat, all at an imposed external pressure of  $p^* = 1.0$ , the shear simulation at constant volume also started with this superimposed pressure.

The increments of shear strain were applied by shearing all atoms into new positions by an affine simple shear transformation in the positive  $y$  direction. To test for possible asymmetries in the behaviour of the mat, shearing was also done parallel to the  $x$  direction, and resulting in very similar results, without any sign of asymmetry. Whether the asymmetry in the behaviour of the small mat resulted from its small size, or its rectangular shape, was not explored further. In any event, the majority of the results to be presented here are related to the large mats, which had symmetrical and isotropic behaviour. Upon the application of a shear-strain increment, the atom positions were relaxed by the MD procedures. The time dependent relaxations of the average enthalpy per atom after an affine shear-strain increment are given in figure 4, which shows that the most unnatural rise in enthalpy due to the imposed

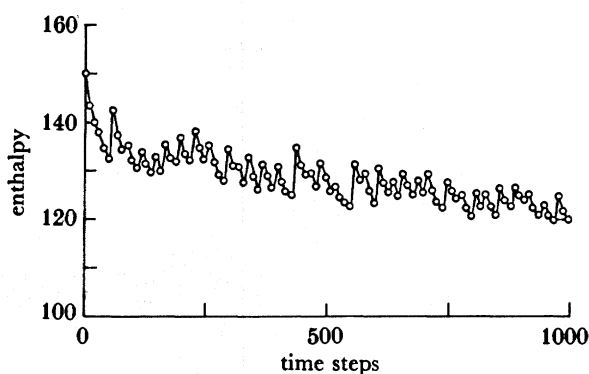


FIGURE 4. Sequence of enthalpy relaxation in the simulation mat of the two-dimensional material shown in figure 2*b*, following a small shear strain increment of  $5 \times 10^{-4}$ . Note that the most rapid early relaxation is complete, after 190 time steps.

affine re-positioning of the atoms is eliminated after about 190 time steps, or about 10 fluctuation periods. Relaxations continue more slowly after this time, and would eventually eliminate any metastable states associated with production of dilatancy and related effects that were of interest. Therefore, the relaxations were stopped after 190 time steps, followed by a new increment of shear strain, and so on. Parenthetically, such rates of shear amount to  $10^8 \text{ s}^{-1}$  in real time. The shear induced intensive properties associated with each atom were calculated after each shear increment. Both the incremental changes in the intensive parameters, and the accumulated totals were obtained at certain strain levels in the stress strain curve, and will be presented in detail in what follows.



## 3. RESULTS

3.1. *Elastic bulk properties*

The elastic properties of the small mat in bulk response were investigated by changing the cell area systematically, in small increments, in both directions to determine the pressure response. The resulting volume average pressure–volume curve is shown in figure 5*a*, from which the small-strain, two-dimensional bulk modulus  $K_{2D}$  under a pressure of 1.0 can be determined readily around this initial pressure, i.e. at  $v/v_0 = 1.0$ , which was 48 in dimensionless stress. This will be compared below with the apparent shear modulus obtained from a shear-stress, shear-strain simulation curve. The atomic displacements and their character that accompanied this volumetric deformation were not studied. It is expected that some inelastic volumetric relaxation had accompanied this deformation. Weaire *et al.* (1971), found in a similar three-dimensional study, that the inelastic relaxations in volumetric deformation were much less extensive than the corresponding shear relaxations accompanying an apparent elastic response. We expect this to be the case here also. As an added feature to this volumetric response, the shear stress changes that result on the borders of the mat were also monitored, and are shown in figure 5*b*. As expected, there were no shear tractions evoked by the volumetric strains (beyond random fluctuations).

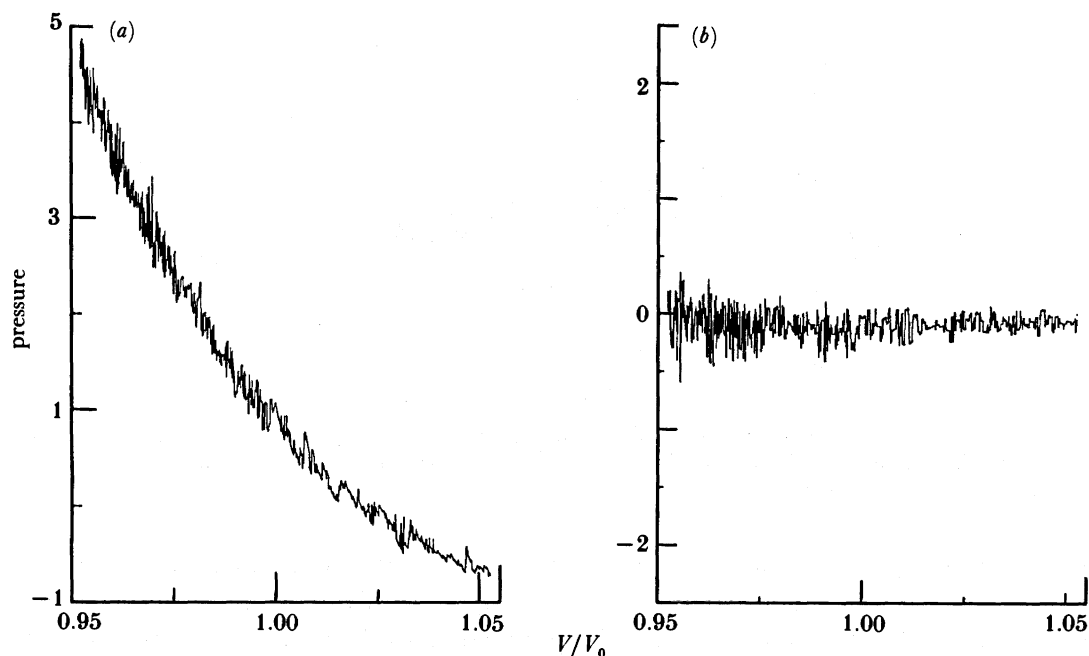


FIGURE 5. (a) Dependence of external cell pressure on volume change (area change); (b) dependence of shear stress on the cell, on volume change.

From the curvature of the pressure–volume curve of figure 5*a*, the pressure dependence of the two-dimensional bulk modulus can be readily determined, and is shown in figure 6. It is found to be quite constant over a wide range at a value of

$$dK/dp = 19. \quad (5)$$

Because of the non-dimensional nature of the information, we expect that the value given in (5) also represents the pressure dependence of the three-dimensional bulk modulus.

By extrapolation of the line in figure 6 to zero bulk modulus, we find that the biaxial cohesive strength  $\sigma_t$  of the mat is 1.55 in dimensionless stress units. The ratio of this to the two-dimensional bulk modulus of 18.8 at zero pressure gives a value of  $8.2 \times 10^{-2}$ , which we find quite reasonable. Extrapolation of the curve in figure 5*a* to zero slope gives the critical areal strain for two-dimensional decohesion as 0.175.

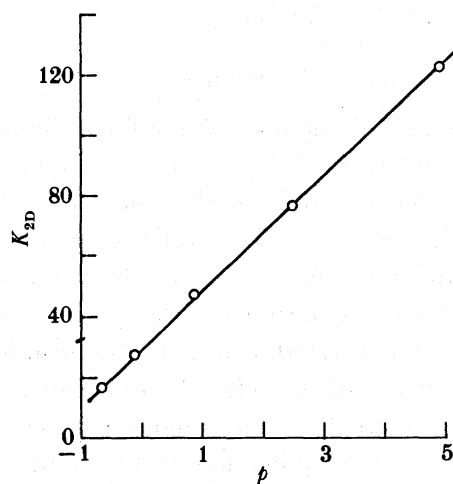


FIGURE 6. Pressure dependence of the two-dimensional bulk modulus derived from the information in figure 5.

### 3.2. Shear response

The elasto-plastic shear response of the large mat up to a total shear strain of 0.27, is shown in figure 7. A clear initial elastic region is visible with a gradual elasto-plastic transition that is complete at a shear strain of around 0.08, where the boundary shear tractions level off at a value slightly above 1.0 in dimensionless stress units. There are considerable variations in the shear resistance between increasing increments of shear strain. This is to be expected from the relatively small nature of the simulation cell, which even with 780 atoms contains only quite finite numbers of strain producing configurations. The apparent two-dimensional shear modulus  $\mu_{2D}$  obtained from the average slope of the initial linear region was found to be 16 in dimensionless stress units under a pressure of  $p^* = 1.0$ . From this shear modulus and the two-

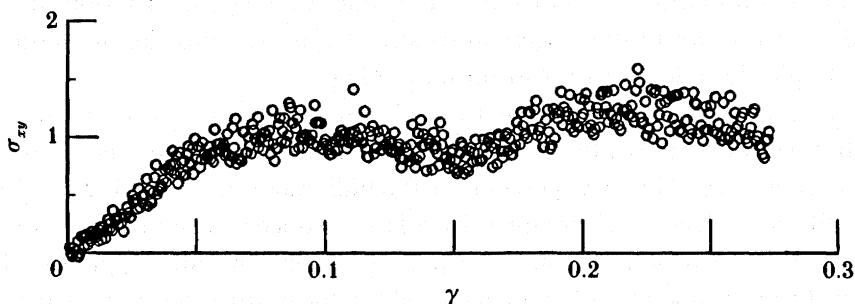


FIGURE 7. The shear stress-shear strain curve of the model atomic glass sheared isothermally at  $T^* = 10^{-3}$ .

dimensional bulk modulus presented in §3.1 above, the fundamental elastic properties of the three-dimensional reference material under a pressure of  $p^* = 1.0$  can be determined through some well-known relations:

$$K_{2D} = \frac{1}{2}s_{11}/(s_{11}^2 + s_{11}s_{12} - 2s_{12}^2), \quad (6a)$$

$$K_{3D} = \frac{1}{3}(s_{11} + 2s_{12})^{-1}, \quad (6b)$$

$$\mu_{2D} = \mu_{3D} = \frac{1}{2}(s_{11} - s_{12})^{-1} = 1/s_{44}, \quad (6c)$$

$$E = 1/s_{11}, \quad (6d)$$

$$\nu = -s_{12}/s_{11}, \quad (6e)$$

where  $K_{3D}$  is the three-dimensional bulk modulus,  $E$  the three-dimensional Young's modulus,  $\nu$  the Poisson's ratio,  $s_{11}$  is the tensile compliance,  $s_{12}$  the transverse compliance,  $s_{44}$  the shear compliance, and  $\mu_{3D} = \mu_{2D}$  is the shear modulus of the amorphous alloy. In the determination of  $K_{2D}$ , a three-dimensional solid was considered stretched in only two dimensions under a constraint of no deformation in the third direction. From the simulation results of  $\mu^* = 16$  and  $K_{2D}^* = 48$ , the following values were determined from the relations given above:  $s_{11}^* = 2.35 \times 10^{-2}$ ,  $-s_{12}^* = 7.80 \times 10^{-3}$  in reciprocal dimensionless stress units;  $K_{3D}^* = 42.2$ ,  $E^* = 42.6$  in dimensionless stress units; and  $\nu = 0.332$ . The absolute values of the three-dimensional moduli are lower by about 25% than the values reported by Davis (1978) for amorphous metals. The calculated Poisson's ratio is quite close to the values reported for close packed metals, but considerably lower than the very high value of 0.41 reported by Davis (1978). We take these results as quite representative, considering that two-dimensional solids usually entrap higher levels of free volume in the amorphous state, and should therefore, have lower moduli.

We note that the ratio of the yield stress in shear to the shear modulus is  $6.25 \times 10^{-2}$ . This is about a factor of 2 higher than the values exhibited by amorphous metals, back extrapolated to 0 K (Argon 1982, 1986), but less than one third of the value obtained in the three-dimensional computer simulation of Srolovitz *et al.* (1983). The latter simulation was also performed by shearing at constant volume, but was performed on a simulation cell so small that it was likely to be too confined to represent a realistic sampling of the possible shearing configurations. Figure 7 shows a dip in the plastic resistance at a total shear strain of 0.15. Although the exact cause of this dip is uncertain, the behaviour could be associated with a large increase in the concentration of liquid-like material in the mat observed at this same shear strain, which we will discuss in §4 below. If this is indeed the case, the dip should be attributed to a size effect in the simulation cell. Outside of this long wavelength variation, the flow stress remains roughly constant up to the largest shear strain of 0.27. Thus the simulation material acts as an ideally plastic solid, with no strain hardening.

Simulations of amorphous media by Weaire *et al.* (1971) have shown that when compared with their crystalline counterparts, the elastic moduli of the amorphous materials are lower, but that while this reduction is only a few percent for the bulk modulus, it is 15–20% for the shear modulus. This difference has been explained by a larger concentration of anelastic non-affine changes in the apparent elastic range in shear processes, than in bulk expansion. To test this possibility, as well as to probe the inhomogeneity of the local processes, the shearing direction was reversed after a forward strain of 0.025, as well as after 0.075 and 0.125, until in each case

unrestrained plastic deformation in the reverse direction occurred. This is seen in figure 8, which shows that the unloading in the apparent elastic range (the upright crosses) after a forward strain of 0.025 does not follow the loading curve, but rather shows a substantial anelastic hysteresis behaviour. The total inelastic residual strain, when the reversed shear stress is zero, is around 0.01, or quite close to 40% of the forward strain. The reverse loading

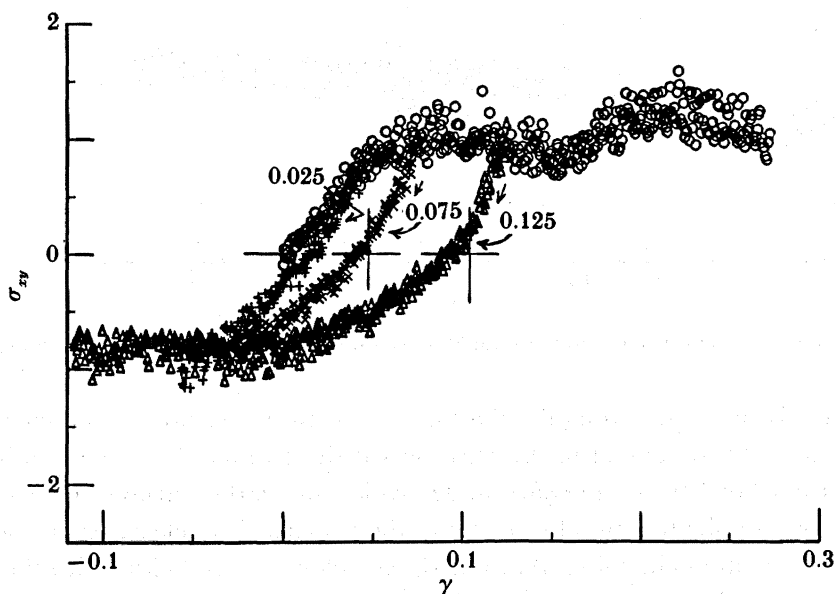


FIGURE 8. Portion of the forward shear-stress–shear-strain curve and three curves for reverse deformation, initiated at forward shear strains of 0.025, 0.075 and 0.125. Note the very prominent Bauschinger effects.

behaviour at the border of the elasto-plastic transition at 0.075 strain (the inclined crosses) and in the unrestricted plastic flow region at 0.125 strain (the upright triangles) show very pronounced Bauschinger effects. The total inelastic residual strain upon complete unloading of the stress was 0.04 and 0.09 respectively, for the two reverse loading paths after 0.075 and 0.125 forward strain. These residual strain magnitudes and inspection of the unloading curves show directly that the initial unloading slopes are considerably higher than the loading slope. Direct determination of these initial unloading shear moduli from figure 8 gave a value of 39 in dimensionless stress units. In an uncomplicated elastic–plastic solid, with no hardening and no rate effect, such an apparent increase in the unloading modulus would be difficult to explain. In our simulation material, however, where considerable anelastic behaviour is exhibited, such increases in the unloading modulus signify continued forward deformation under stress during the early portion of the unloading. Thus more rapid unloading should result in a lower unloading modulus, and vice-versa for a slower rate of unloading.

### 3.3. Pressure response to shearing

It is well known that plastic shearing of a structurally relaxed disordered material increases the disorder, because of an intrinsic dilatancy effect. Such deformation induced dilatations have been measured by Deng & Lu (1983) and Argon *et al.* (1985) in amorphous  $\text{Pd}_{77.5}\text{Cu}_6\text{Si}_{16.5}$  alloy. As our shear simulations were carried out at constant volume, the

developing dilatancy was expected to increase the pressure on the simulation cell. This was found to be the case in both the small simulation cell as well as in the large one. Figure 9 shows the developing pressure upon shearing in the large system. In the beginning, there is a sudden and unexplained rise in the pressure, which decays slightly in the strain range of  $0-2.5 \times 10^{-2}$ , but then rises monotonically to an asymptotic level of 1.6 in dimensionless stress units at a strain

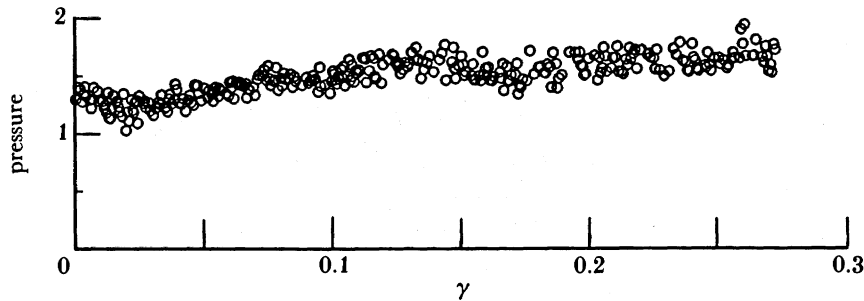


FIGURE 9. Increase in evoked pressure on the simulation cell due to deformation induced volume increase.

of 0.13 (beyond this it remains roughly constant). The rising portion of the curve between 0.025 and 0.13, when extrapolated backwards toward the ordinate line, gives an intersection roughly at a pressure of 1.0, the pressure under which the earlier structural relaxation was carried out. The reason for the initial step rise in the response is unclear. After about 0.13 of strain, conditions remain unchanging. As we shall see below, this coincides with the beginning of perceptible shear localization. It is expected that shear localization due to dilatancy should begin very early in the deformation process, but that the conditions of deformation at constant volume have been responsible for its delay to a strain as large as 0.13.

To explore the changes in the shear induced pressure response, the pressure on the system during the two reverse loading histories, starting at  $\gamma = 0.075$  and  $0.125$ , was also monitored and is shown in figure 10. Reversal of deformation results in a rapid and dramatic decrease in the previously built-up pressure, reaching a minimum of  $p^* = 1.0$  at the point where the reverse stress-strain curve crosses through the point of zero shear stress. Clearly, the decrease in the pressure must be due to reversals of many of the shear transformations just accomplished before the reversal of deformation. Such transformations are expected to have nearly identical

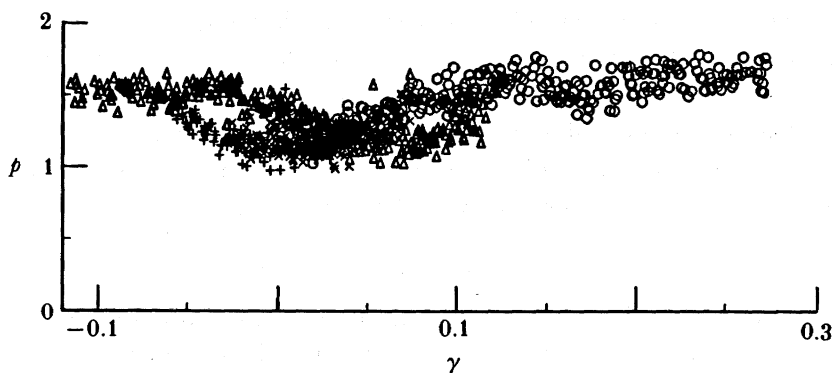


FIGURE 10. Effect of deformation reversals on the evoked pressure. Note dips in evoked pressure with reverse deformations initiated at forward shear strains of 0.075 and 0.125.

environments of residual back stress and associated pressure, which could be systematically reversed as these transformations are reversed. Once the supply of these most recent transformations is exhausted in reverse deformation, the early history is wiped out, and new reverse transformations have to be initiated in fertile but neutral material. Finally, from the shear induced increase in pressure of  $\Delta p = 0.6$  between 0–0.13 strain, and the two-dimensional bulk modulus of 59 at the final pressure of 1.6, where steady conditions of flow are reached, we calculate an overall change of dilatancy effect of  $(\Delta v/v)/\Delta\gamma^p = 0.151$  for a total plastic strain increment of  $6.75 \times 10^{-2}$  between 0 and 0.13 total strain.

### 3.4. Change in enthalpy and structure due to plastic shearing

The dilatancy of the shearing process serves to make the material more amorphous and raises its excess enthalpy. At the very low temperature, where the simulation was performed, the major contribution to the enthalpy of the system is through its change of potential energy (see Appendix 1). Figure 11*a, b* show the change in potential energy with forward shear and the two special paths of reverse shear respectively. The potential energy changes are parallel to the changes in deformation induced pressure resulting from the dilatancy, which itself is a direct cause of the rise in enthalpy.

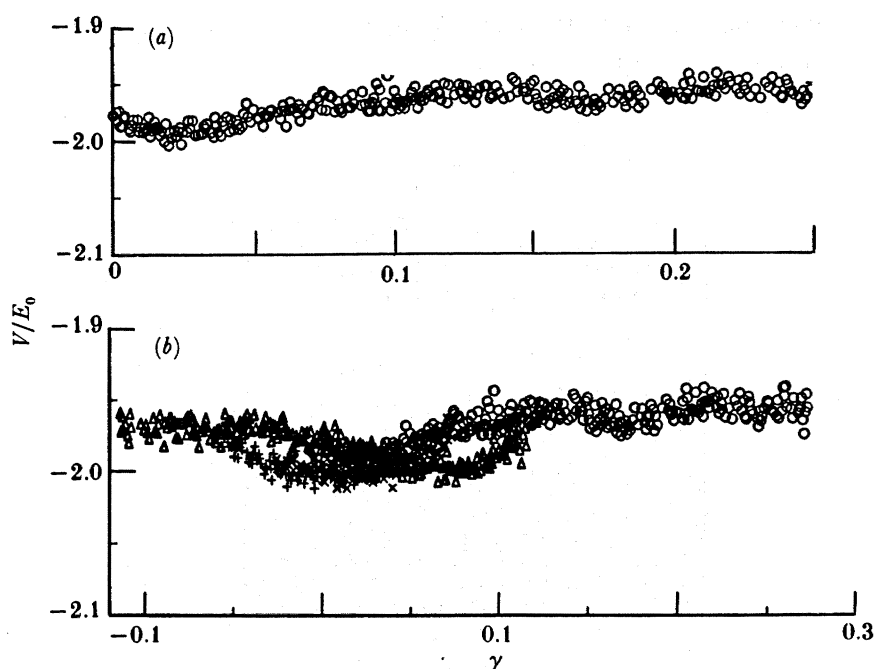


FIGURE 11. (a) Change in potential energy per atom (enthalpy per atom) in dimensionless units of energy as a function of shear strain. The close parallel of this behaviour to that of the pressure shown in figure 9 indicates that the increases in enthalpy are due to the dilatancy effect; (b) changes in the potential energy upon deformation reversals.

The corresponding changes in the structure due to dilatancy induced amorphization are also apparent in the RDFs. Figure 12 shows the RDF of the material at a strain of 0.10. Comparison of this with the RDF in figure 3 shows a very clear broadening of the distribution with decreases in the peak heights and filling-in of the regions between the peaks. This change in the RDFs due

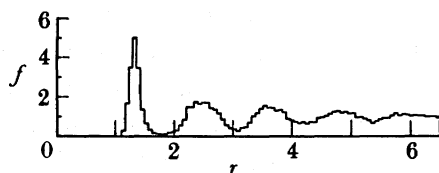


FIGURE 12. Radial distribution function of atom positions of the simulation cell after a shear strain of 0.1.

to reamorphization proceed most rapidly in the very early straining range between 0 and 0.08, where fully established plastic flow conditions are achieved, but continues more slowly up to 0.13, where the rise in shear induced pressure ceases. After about 0.10 (the state in figure 12), the further changes in the RDF with shearing are less dramatic than in the early phases. The observed amorphization is a consequence of plastic shearing, even in the apparent elastic range, where as we shall see below, inelastic transformations occur with sufficient frequency.

### 3.5. The topological features of local plastic flow

#### 3.5.1. Plastic flow at yield

To establish the nature of plastic flow in the simulation, we present first the distribution of local shear transformations and describe the environments in which they occur for a small plastic-strain increment of  $\Delta\gamma^p = 5 \times 10^{-4}$  near the macroscopic yield region of the elastic to plastic transition, at a total strain of  $\gamma = 0.05$ .

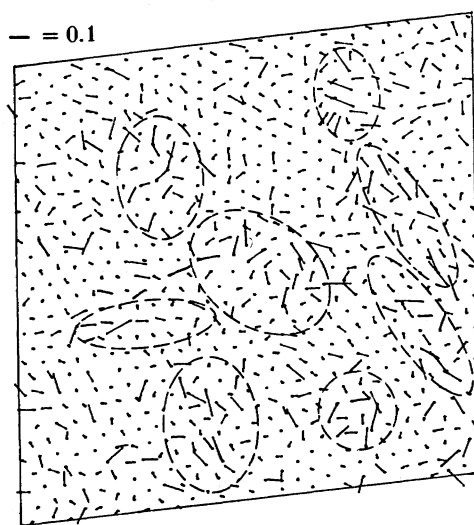


FIGURE 13. Incremental displacement field of atoms due to a shear strain of  $5 \times 10^{-4}$  at a total shear strain of 0.051. Note several of the clusters undergoing cooperative deformation. ( $\Delta\gamma = 0.0515$ – $0.052$ .)

Figure 13 shows the incremental displacement field of the atoms in units of the fundamental length  $r_0$ , during the plastic increment. Each displacement vector emanates from the atom with which it is associated. Such a displacement field, presented by most investigators in the past, as the only feature of the plastic deformation is too confusing to provide much enlightenment. Nevertheless, it is possible to outline in it regions where the most intense activity is concentrated, such as the eight regions that have been delineated by closed contours. They

resemble to some extent the zones of concentrated (C) and diffuse (D) shear transformations identified by Argon & Kuo (1979) in sheared Bragg soap bubble rafts. Here, we will not pursue details of displacement fields further, but rather concentrate our attention on the field of local strain spikes introduced first also by Argon & Kuo (1979). Thus, figure 14*a* shows the incremental maximum shear strain spikes (regardless of their principal axes) that have resulted during the specific macroscopic plastic strain increment under consideration. The accompanying local dilatation spikes (upright triangles represent positive quantities) are shown in figure 14*b*. As the simulation has been performed under constant volume conditions, the volume average sum over all the dilatation spikes should give zero. Clearly, many dilatancy events have been either suppressed or perhaps even reversed by this condition, or in the least have affected their surroundings somewhat by pressurizing the background.

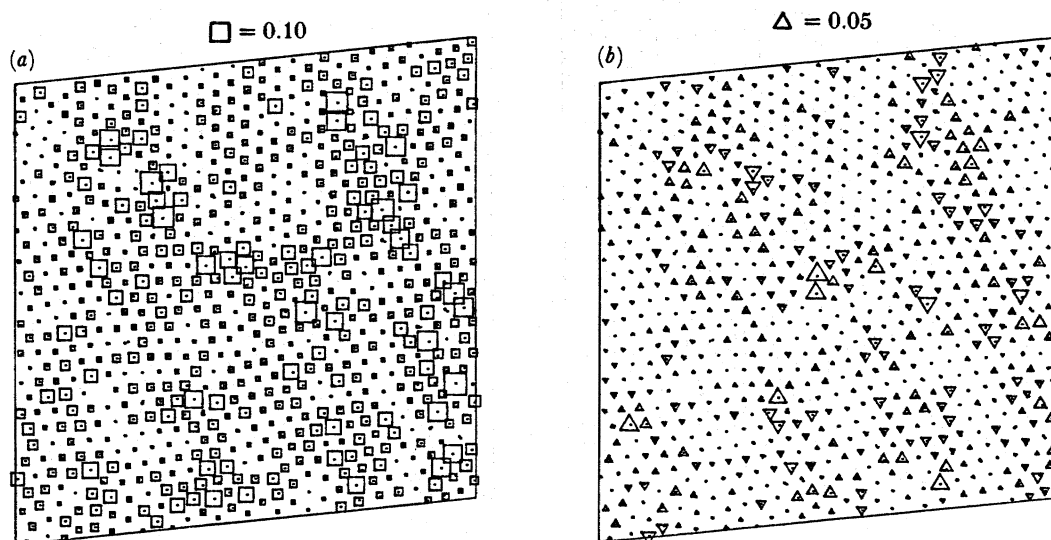


FIGURE 14. (a) Local atomic level maximum shear spikes, regardless of principal axis orientation in the simulation field for an external imposed shear strain increment of  $5 \times 10^{-4}$  at a total shear strain level of 0.051; (b) the corresponding atomic site dilatations associated with the shear strain increment of (a). ( $\Delta\gamma = 0.051-0.0515$ .)

Inspection of figure 14*a* shows that the local shear-strain spikes are quite large, with many of them being in the range of 0.05–0.10, and some are somewhat larger. This occurs during an external total strain increment of only  $5 \times 10^{-4}$ . Thus, clearly, the local plasticity is in the form of shear transformations, with transformation shear strains of the order of 0.05–0.10 in local volume elements. This is quite similar to the kinematics of plastic flow studied by Argon & Shi (1982) in the Bragg soap bubble rafts, where transformation shear strains of these magnitudes were also observed.

A point of primary interest in this simulation is the description of the most relevant state parameter of the local environment, in which the local shear spikes develop. Four primary candidates that must be considered are shown in figure 15*a-d*. They are:

- (1) the local atomic level maximum shear stress (regardless of orientation of principal axes);
- (2) the atomic level pressure and negative pressure (upright triangles represent pressure);
- (3) the local free volume;
- (4) the local atomic site distortion parameter;



all evaluated at the beginning of the specific, small plastic-strain increment, which is the subject of discussion.

Comparison of figure 14*a* with 14*b* shows a reasonable visual correlation between the local dilatations and the local shear strain spikes everywhere in the field, but especially in the upper right quadrant and along the diagonal zone radiating from the centre of the figure into the upper left quadrant. Correlation of the shear spike distribution with the distributions of the atomic level maximum shear stress and atomic level pressure, shown in figure 15*a, b*, is weak at best. The best evidence of any such correlation is in the centre of the cell, in small portions of the lower left-hand quadrant and lower right-hand corner of the lower right-hand quadrant for the maximum shear stress. For the pressure, the correlation is roughly at the same places,

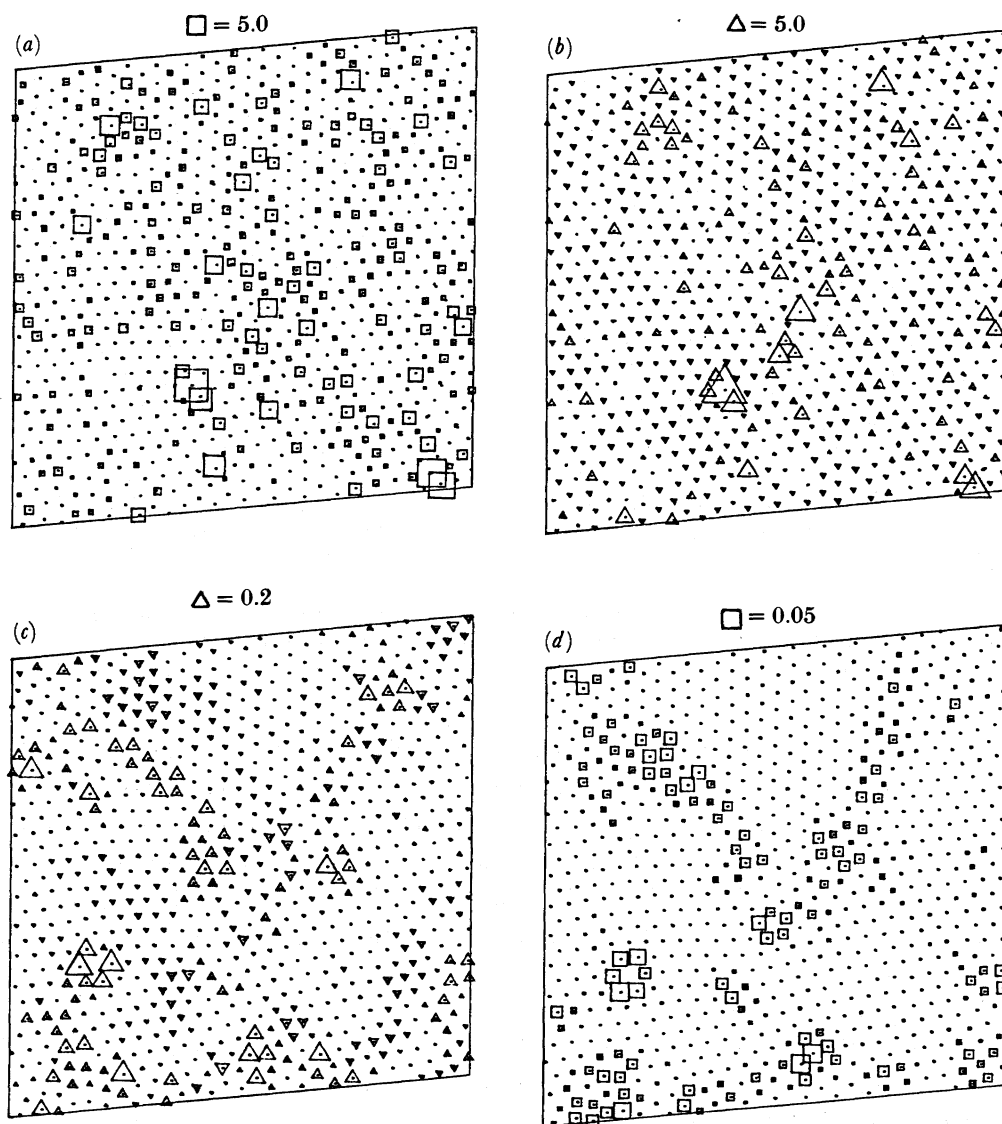


FIGURE 15. (a) Distribution of atomic site maximum shear stress, (b) distribution of atomic site pressure ( $\Delta$ ) and negative pressure ( $\nabla$ ), (c) distribution of free volume among atom sites, (d) distribution of atomic site distortion parameter; all at a total shear strain of 0.051 considered as description of the initial state for a subsequent increment of shear strain of  $5 \times 10^{-4}$ .

but even weaker. On the other hand, the correlation between the sites of the maximum shear strain and both the free volume (figure 15*c*) and the atomic site distortion parameter (figure 15*d*) is quite striking. The latter two are, of course, specific quantitative characterizations of the boundaries between the quasi-ordered domains, where the liquid-like material concentrates, as can be seen by comparing figure 15*c, d* with figure 2*b*. That this is bound to be the case must be clear from the fact that in the liquid-like material there is a concentration of excess free volume and the local distortion. This has produced a significant reduction of cohesive interaction, as we have discussed in detail in parts I and II, and as can be deduced also from the discussion in §3.1 above.

### 3.5.2. Character of shear transformations

As the plastic shear events are enforced by the imposed  $\Delta\gamma_{xy}$  shear (or alternatively by the applied shear stress  $\sigma_{xy}$ ), it is of interest how well the local shear spikes are polarized by these driving forces. To probe this, only the  $xy$  components of the shear spikes were determined, and their magnitudes are plotted in figure 16. A cursory check of the integrated magnitudes of these

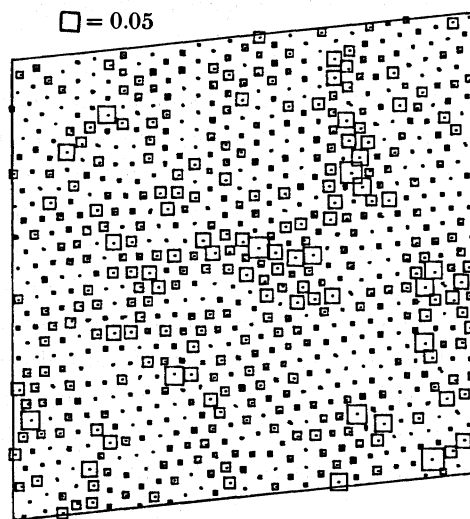


FIGURE 16. Distribution of local atomic  $xy$  shear strain increments during an externally imposed shear strain increment of  $5 \times 10^{-4}$ , at a total shear strain of 0.051. ( $\Delta\gamma = 0.051 - 0.0515$ .)

shear strain spikes has shown that this total integral over the field exceeds considerably the applied shear increment of  $5 \times 10^{-4}$ . This observation disclosed that a significant fraction of the total shear events shown in figure 16 are actually of a negative sign, and occur against the applied stress. Figure 17 shows the distribution of both the positive and negative shear spikes during the specific plastic strain increment. This startling behaviour of reverse local shear against the applied shear stress is readily explained when it is recognized that the root mean square level of excess enthalpy associated with the structural disorder is many times larger than the work done by the applied stresses interacting with the local strain increments. Thus, as primary shear transformations occur in and around the liquid-like regions, they momentarily destroy local mechanical equilibrium and permit some surrounding material to lower its Gibbs free energy, even at the expense of doing work against the applied stress. This fact was confirmed by studying the Gibbs free energy change for a number of local cluster systems as

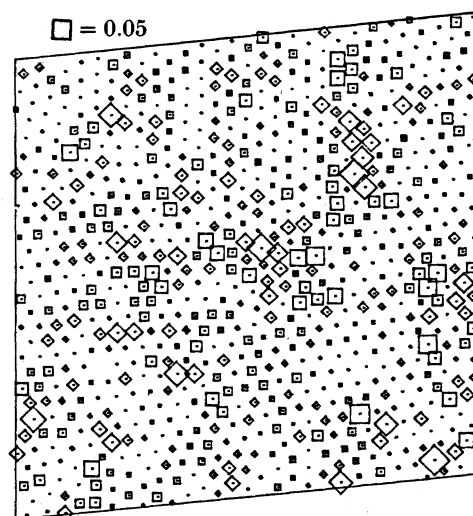


FIGURE 17. The distribution of  $xy$  shear strains of positive and negative sign occurring during an externally imposed shear strain increment of  $5 \times 10^{-4}$  at a total shear strain of 0.051, the squares are strain spikes, and are in the same direction as the applied strain increment, the diamonds are of negative sign. ( $\Delta\gamma = 0.051-0.0515$ .)

a function of strain in which shear transformations had been observed, regardless of sign. In all instances, the Gibbs free energy was found to decrease monotonically (see Appendix 1), as a function of increasing average strain. Two typical cases are shown in figure 18*a, b*. Of these, figure 18*a* pertains to a case of positive local shear strain, while 18*b* pertains to a case of negative local shear strain. These figures also show that the monotonic decline of the free energy is interrupted at many places by energy barriers of varying height. These are interpreted as barriers that can be overcome by thermal activation under a constant applied stress at higher temperature. Accordingly, the frequency distribution of these barrier heights was determined over the history paths of six separate clusters as an indication of the distributed nature of the shear relaxation processes. This distribution is shown in figure 19. It demonstrates that while the majority of the energy barriers are less than  $E_0$  (i.e. 0.15 eV), some are nearly

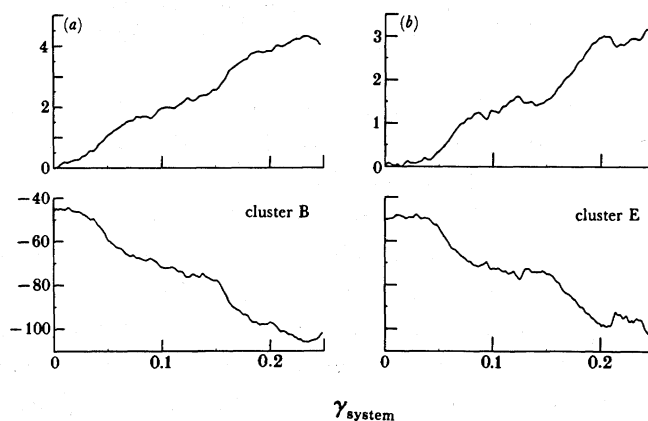


FIGURE 18. The specific history of plots of two separate clusters showing the monotonic decrease in Gibbs free energy accompanying a monotonic rise in shear strain: (a) cluster shearing in the same direction as external shear increment; (b) cluster shearing in the opposite direction.

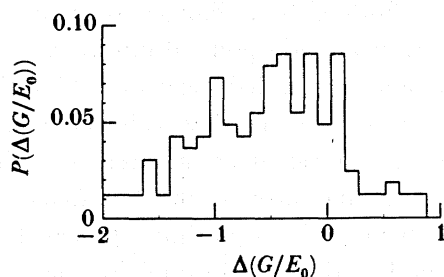


FIGURE 19. Frequency distribution of free energy barriers on the monotonic decrease of the Gibbs free energies in deforming clusters, as obtained from history plots of the type shown in figure 18*a, b*.

a factor of 10 larger. The information derived from these observations, however, is too incomplete for purposes of quantitative comparisons with experimental distributions of the type studied in detail by Deng & Argon (1986*a, b*), from internal friction measurements.

Examination of figure 17 also shows that once the forward and reverse shear processes are separated, a significant amount of clustering of shear processes of either type becomes more apparent. Among these clusters, those that are of elongated and concentrated shear type, as classified by Argon & Kuo (1979), immediately stand out. The diffuse shear transformations, as usual, are more difficult to identify. Part of this difficulty is associated with the lack of time resolution in the chronology of appearance of individual processes. Even in the span of a very small plastic strain increment, such as the one on which the information in figures 14–17 is based, many transformation events, actually separated in time, have become superimposed.

The proximity of the clusters producing positive and negative shear to each other also attests to a cause-effect relation between the two. The change in local atomic configurations during the development of a forward shear cluster upsets mechanical equilibrium in the immediate surrounding regions, and can trigger a reverse shear-producing cluster if the initial misfit incorporated in that region is conducive for such a reverse transformation.

A similar form of clustering is also perceivable in the distribution of the dilatation spikes, shown in figure 14*b*. In many regions where upright triangles are clustered, indicating dilatation increments, the immediate local surroundings show rings of downright triangles

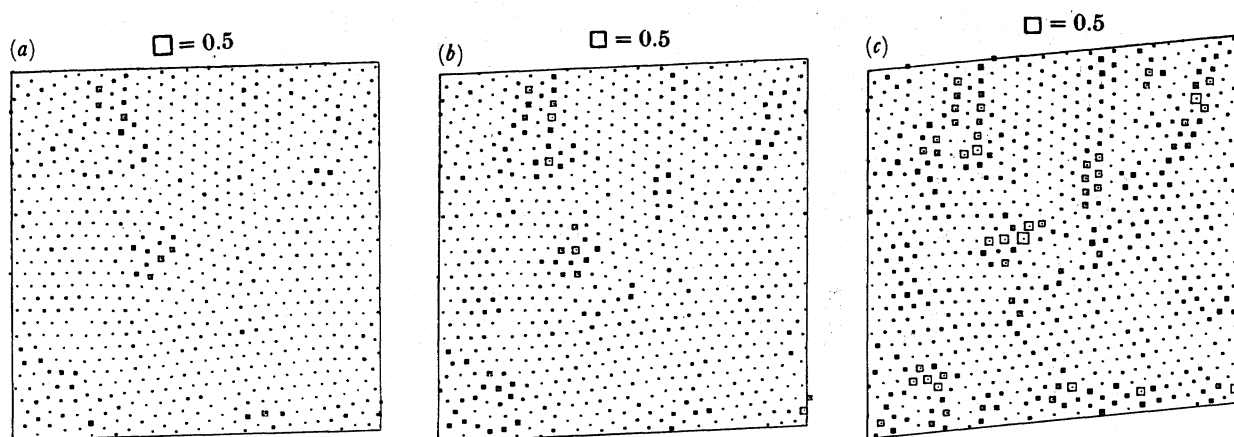


FIGURE 20. Distribution of local total  $xy$  shear strains in the initial apparent elastic response range: (a) at a total shear strain of 0.0125; (b) at a strain of 0.025; (c) at a strain of 0.05.

indicating inelastic local compaction. Both of these changes correlate reasonably well in spatial distribution with the shear processes, as already pointed out above.

Examination of the distributions of local coupled shear and dilatational transformations in different regions of the stress–strain curve indicated that regions in which transformations were found in earlier stages of straining could transform again, later. As in the vast majority of cases, the shear transformations occurred in and around the boundaries of liquid-like material separating quasi-ordered domains; this repetition, in time, of local shear activity in the fertile boundary regions, attests to the true liquid-like nature of this material. As these boundaries

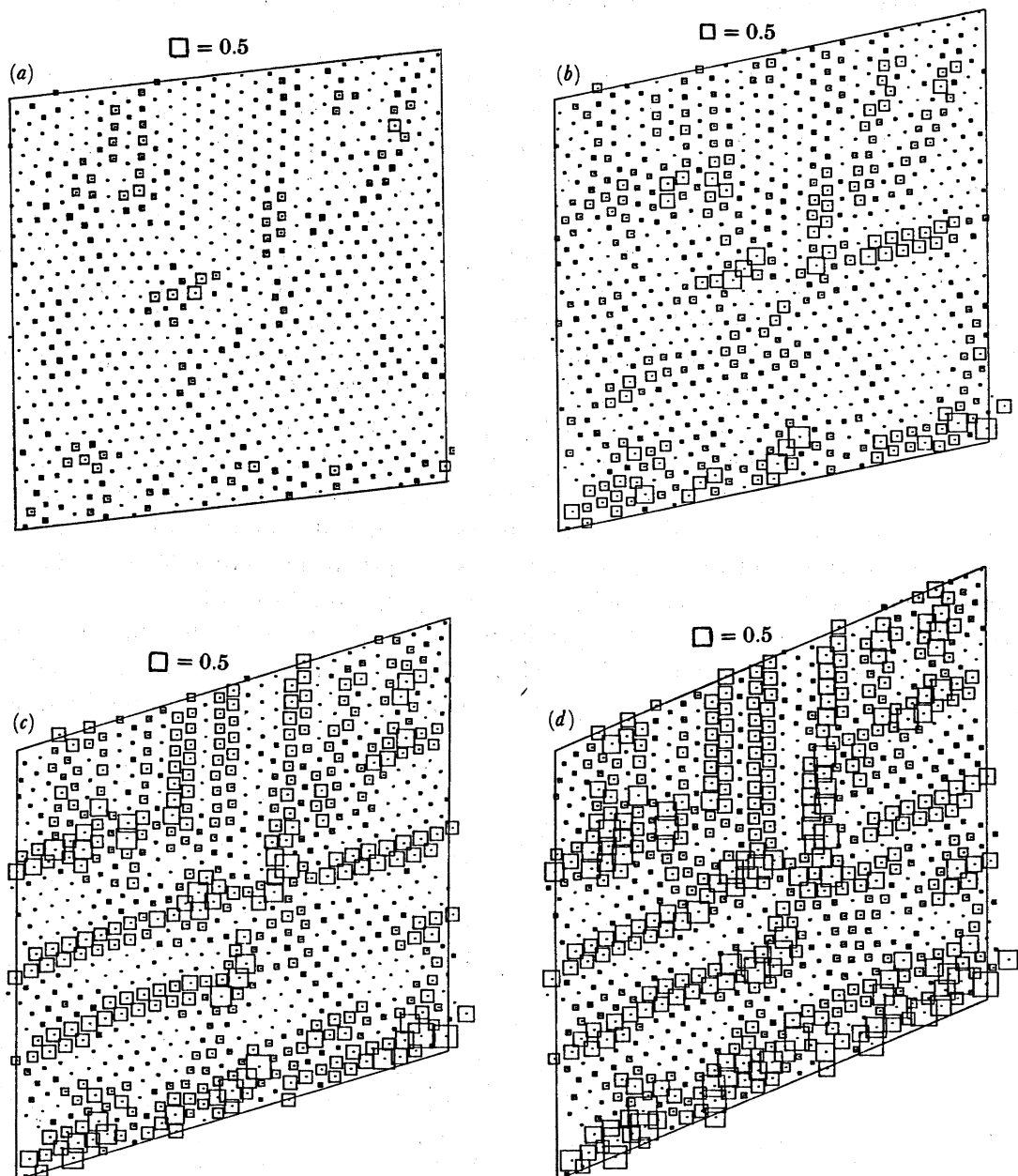


FIGURE 21. Sequence of development of shear localization in the simulation cell due to the accumulated effect of deformation induced dilatations. (a)  $\gamma = 0.05$ , (b)  $\gamma = 0.10$ , (c)  $\gamma = 0.15$ , (d)  $\gamma = 0.70$ .

were not planar but intricately tortuous, increasing strain must shift them around, so as not to build up large plastic misfit stresses in the quasi-ordered domains. That this is indeed the case can be seen from the set of frames shown in figure 24 *a-d*.

### 3.5.3. The elastic-to-plastic transition

In §3.2 it was demonstrated, by means of reverse loading within the apparent elastic range, that inelastic non-affine processes must be occurring even in this range of response of the material. That this is indeed so, is shown in figure 20 *a-c*, which shows the total accumulated

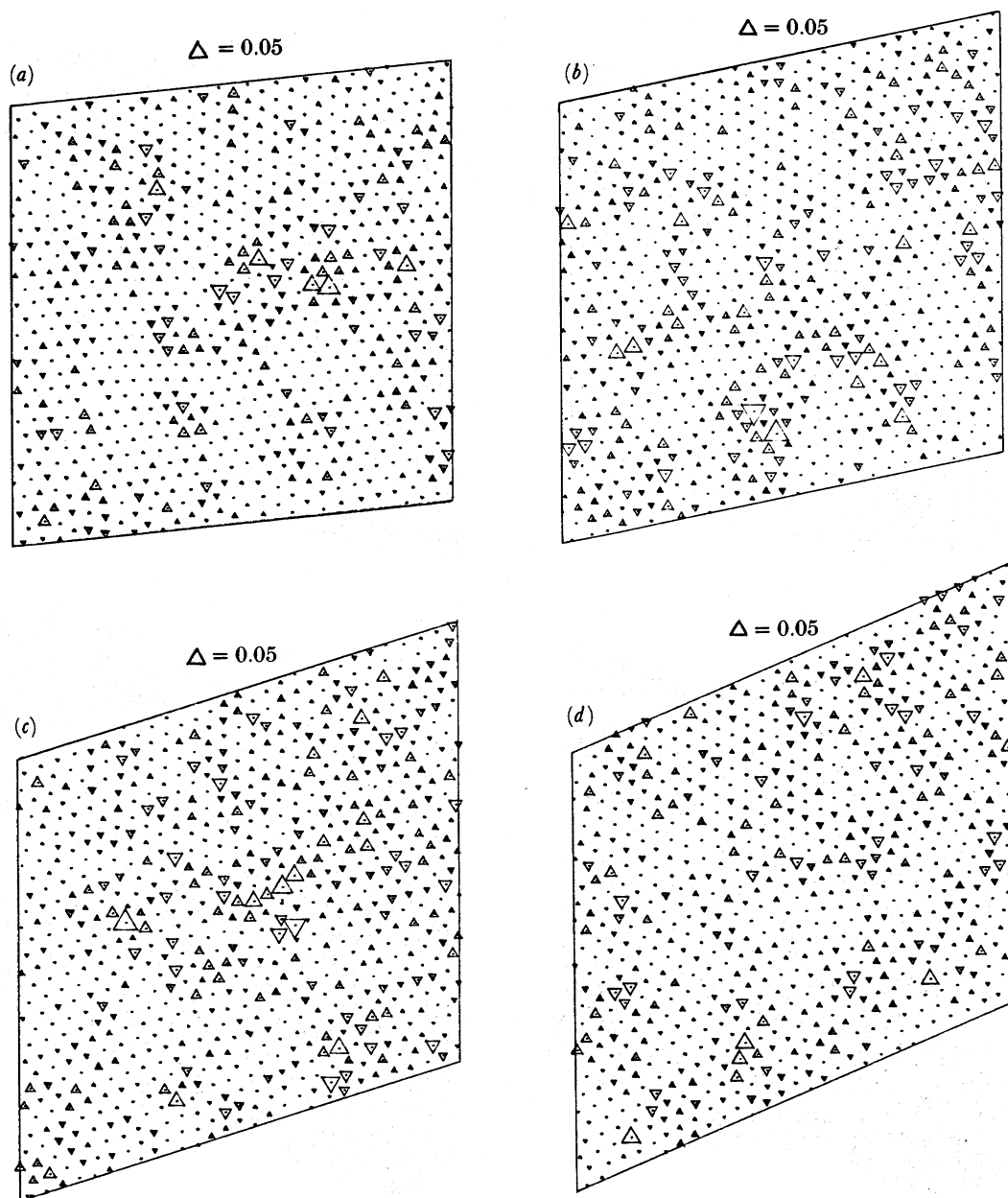


FIGURE 22. Distribution of deformation induced local volume changes during externally imposed strain increments of  $5 \times 10^{-4}$  at total strains of: (a)  $\gamma = 0.05$  ( $\Delta\gamma = 0.0505-0.051$ ); (b)  $\gamma = 0.10$  ( $\Delta\gamma = 0.1005-0.101$ ); (c)  $\gamma = 0.15$  ( $\Delta\gamma = 0.1505-0.151$ ); and (d)  $\gamma = 0.20$  ( $\Delta\gamma = 0.2005-0.201$ ).

$\gamma_{xy}$  strain in the apparent elastic range at total average strains of 0.0125, 0.025, and finally at 0.05, where the most rapid transition to fully developed plastic flow begins. Examination of these three plots shows that inelastic strains in this range do not spring-up as shear transformations with the same full strength as they have during fully developed plastic flow. They rather grow gradually and incrementally in fertile regions that lie in or near to the liquid-like material. Thus they are more of an anelastic nature, permitting them to return to nearly the undistorted state of the material when the stress is removed (albeit accompanied by a hysteresis effect).

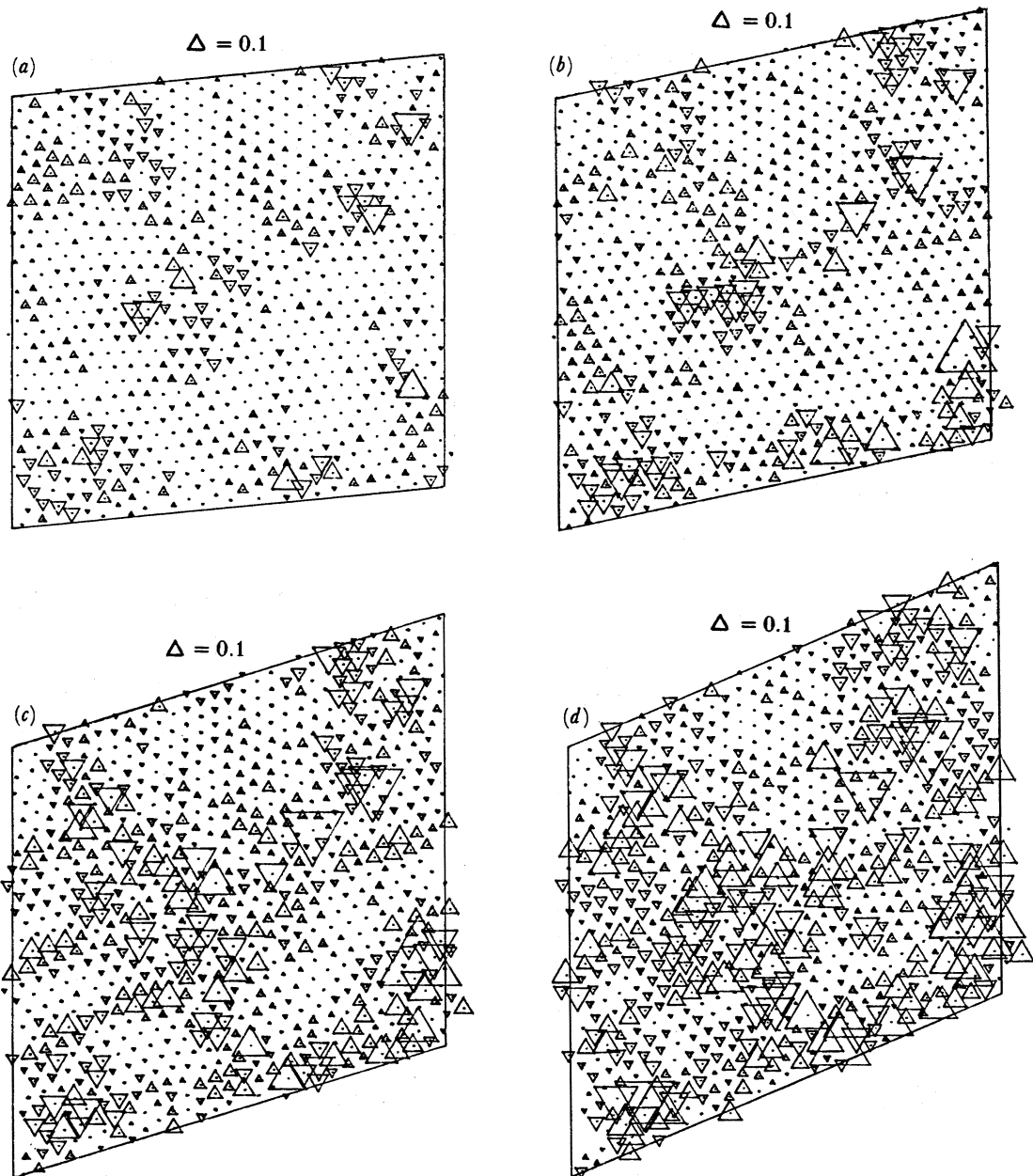


FIGURE 23. Distribution of total accumulated local dilations at atom sites at total strains of (a)  $\gamma = 0.05$ , (b)  $\gamma = 0.10$ , (c)  $\gamma = 0.15$  and (d)  $\gamma = 0.20$ .

### 3.5.4. Shear strain localization

The dilatant nature of plastic flow in ductile amorphous media has been proposed to be the source of strong shear localization in these materials at low temperatures and high strain rates (Spaepen 1977; Argon 1979; Argon *et al.* 1985; Steif *et al.* 1982), where the shear induced free volume or newly formed liquid-like regions do not decay fast enough by thermally controlled structural relaxations. This should be particularly the case at the low temperature of the present simulation. These regions then catalyse further shear transformations and progressively localize the deformation into intense planar shear zones (Argon 1979; Steif *et al.* 1982). This phenomenon has been observed directly in the simulation cell. Figure 21 *a–d* show a sequence of cumulative shear activity in the strain range of 0.05–0.2. The localization of shear into planar zones in the simulation cell is apparent. To establish the connection of this localization to dilatancy, both the incremental changes in local volume (figure 22 *a–d*), as well as the total accumulated dilatation (figure 23 *a–d*) was determined for these four states of total strain. It needs to be kept in mind that the simulation was carried out under conditions of constant volume. Therefore, the total integrated volume changes must be zero for all cases, and the total dilatation must be the same as that in the initial state of the simulation. Thus the intrinsic dilatancy of the flow, which clearly manifests itself in the shear induced pressure rise, shown in figure 9, must produce local inelastic dilatation spikes in regions of flow concentration as well as some spikes of material compaction in the immediate neighbourhood. Clearly, in this process, the inelastic dilatations must dominate, because they elastically pressurize the entire simulation cell. The sequence of figure 22 *a–d* shows that incremental dilatancy activity does indeed fall into the zones of the apparent shear localization. The correlation is much more striking when viewed as the total accumulated dilatations, as shown in figure 23 *a–d*, where the correlation of the localized accumulated dilatations with the localized shears, shown in figure 21 *a–d*, is clear.

## 4. DISCUSSION

As stated in §1 above, the description of the mechanism of plastic flow in amorphous media has created much controversy. Gilman (1968), Li (1976), and others (Chaudhari *et al.* 1983; Shi & Chaudhari 1983) have drawn specific analogies between the plastic flow mechanisms in nearly perfect crystals, where long range mobility of crystal dislocations produces strain, and the plastic flow in amorphous metals, where shear localization is frequently observed at low temperatures and high strain rates (Spaepen 1977; Argon *et al.* 1985; Megusar *et al.* 1979). Two compelling points in support for this view are the observed intense shear localization process, as already mentioned, and an observed second-order kinetics in the thermal recovery behaviour of deformed samples (Ahn & Li 1983), not to mention the great attraction to immediately capitalize on many other explanations of crystal plasticity for allied phenomena observed in amorphous metals. Many adherents of this point of view have extended the analogy also to glassy amorphous polymers, even though there the chain molecules would introduce important constraints to any concentrated shear process. As there are important differences even in the phenomenology of the deformation of polymeric materials, we do not pursue their discussion here further either for or against the above point of view.

A separate compelling reason for taking a dislocation point of view for the behaviour of



amorphous atomic solids is the many thought-provoking theoretical studies of a topological nature of transforming a crystalline solid into an amorphous one by progressively dislocating it (Koizumi & Ninomiya 1978, 1980). A further stimulus in this direction comes from equally interesting studies on the melting of crystalline solids by introducing increments of disorder through dislocations, and demonstrating that this process can give rise to sharp melting behaviour (for a general review of the early work see Nabarro (1967)). When carried out with care (Kuhlmann-Wilsdorf 1971; Kotze & Kuhlmann-Wilsdorf 1971), both the latter and the former theoretical studies do indeed lead to relaxed structures that exhibit most of the quantitative signatures of the glassy state. On the basis of this, it could then be argued that the dislocations that have led to the end state might have vestigial properties that can be mobilized for the processes of plastic deformation, in spite of the fact that the amorphous condition can be reached also by melting and severe radiation damage (Cherns *et al.* 1980). Nevertheless, the attraction of this indirect reasoning should be clear in the light of what we have presented in this paper and in parts I and II. In these studies we see, however, that the role of the dislocations and their vestigial properties are not to provide mobile configurations, but to introduce fertile matter that makes possible plasticity by shear transformations, which have no mobile properties, but are initiation controlled.

The alternative mechanism to explain plastic flow in amorphous atomic media is based on shear transformations in local small atom clusters proposed first by Orowan (1951) to explain viscous deformation. This was later generalized by Argon and co-workers (Argon 1979, 1982; Argon & Kuo 1979; Argon & Shi 1982) to explain most observed inelastic phenomena in atomic glasses, including the rate process of deformation, the temperature dependence of the flow stress over its entire range from 0 K to  $T_g$ , and the distributed nature of the rate controlling process. Separately, Spaepen and co-workers (Spaepen 1977; Taub & Spaepen 1980) have developed a point of view of shear relaxations in a region of free volume where enhanced mobility of atoms should be preferred because of the lowered level of cohesive interactions. This mechanism of plasticity by local shear transformations has found support from many other investigations, including those based on computer simulations (Srolovitz *et al.* 1983; Maeda & Takeuchi 1978). These latter investigations and the simulations based on the disordered Bragg bubble model (Argon & Kuo 1979; Argon & Shi 1982) have provided important information on the particular fertile neighbourhoods, in which shear transformations develop.

Thus, our simulations on melting, structural relaxations, and on plastic flow, presented in parts I and II, and in this paper, have given a certain unification to many of these diverse points of view. The simulations on melting and structural relaxations have demonstrated that the disorder in the amorphous material can be divided into quasi-ordered regions of very small dimensions of 3–10 atoms across and internally disordered boundaries, surrounding these quasi-ordered regions. These boundaries, which percolate through the entire structure, exhibit topological signatures of dislocation cores that are apparent in the form of ordered sequences of five- and seven-sided polygons. Here, contact is made with the dislocation theories of melting and amorphization (Koizumi & Ninomiya 1978, 1980; Kuhlmann-Wilsdorf 1965; Kotze & Kuhlmann-Wilsdorf 1971), and other analogue simulations using vibratory agitation of Bragg bubble rafts (Fukushima & Ookawa 1955). The present simulation on plastic flow on the other hand, has reinforced rather strongly the points of view of deformation by nucleation of shear transformations in fertile regions, which have turned out to be along the boundaries of disordered material composed of dislocation cores. Here, contact has been made with

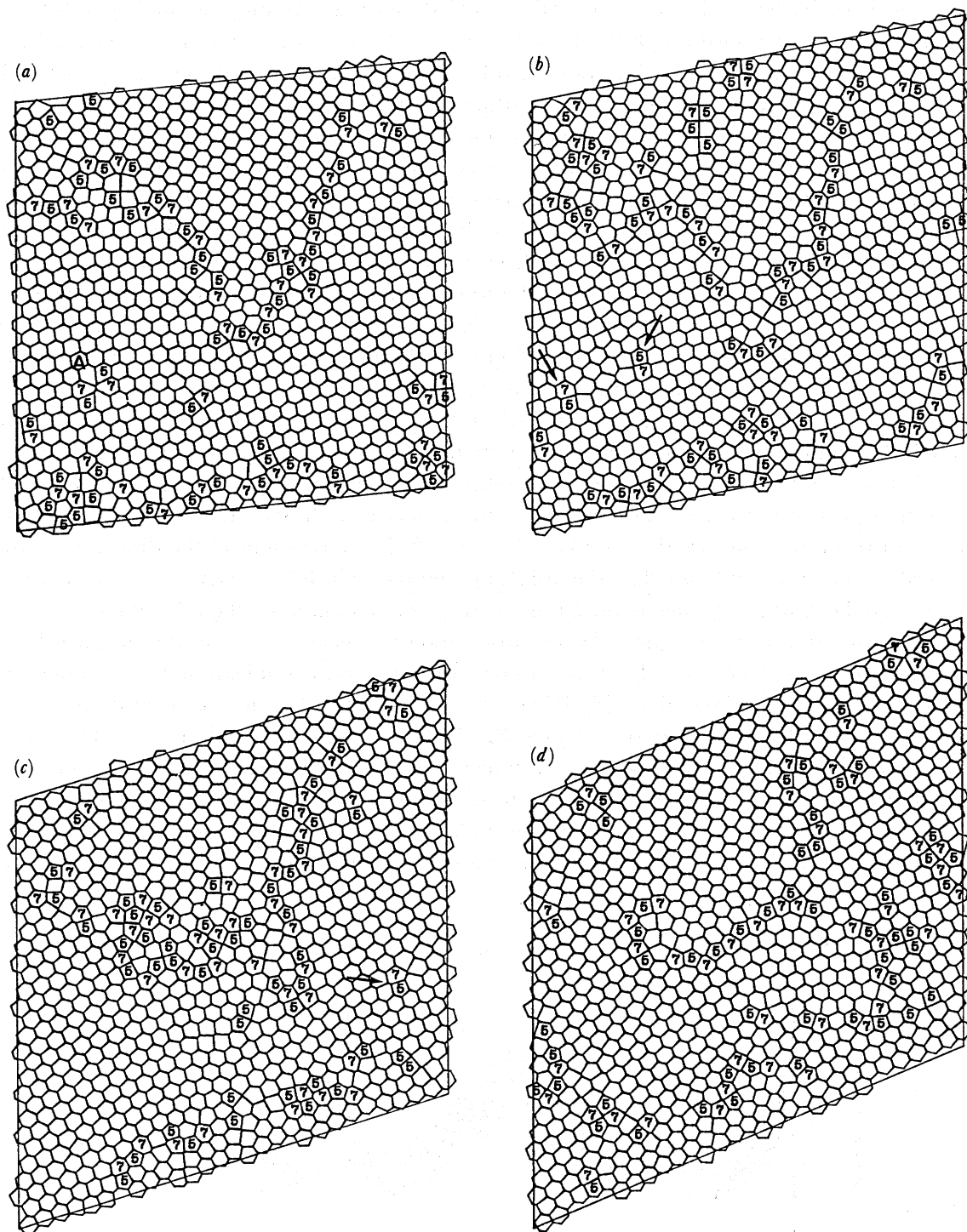


FIGURE 24. Deformation induced changes in the distribution of liquid-like material represented by the boundaries of 5–7-sided polygons: (a) after a total strain of  $\gamma = 0.05$ , note a dipole of two-edge dislocations at site marked A; (b) after  $\gamma = 0.10$ , note the separation of the two-edge dislocations from the dipole configuration; (c) after  $\gamma = 0.15$ , note that the left-hand dislocation has now entered the cell from the right side; (d) after  $\gamma = 0.20$ .

theoretical studies on fluidity in melts and subcooled melts, attributing the fluidity to liquid-like material percolating through the structure (Cohen & Grest 1979; Grest & Cohen 1981).

We recognize that the simulations presented here, and in the associated studies of part I and part II, are two-dimensional, and that three-dimensional material will have another degree of freedom in accomplishing the topologically complex redistribution of structural disorder and repositioning of atoms in shear transformations. We maintain, however, that our simulations have captured the qualitative essence of these complex processes, which differ in three-dimensional matter only quantitatively. On the other hand, the powerful capability of visualization of processes in a plane, have been in our view, of vital importance.

As in our simulation mats of well-relaxed glasses small quasi-ordered regions exist, the lack of a contribution to the overall strain by dislocation glide is surprising. Actually, some evidence for such isolated glide events has also been found. In addition to the short-range glide motion under the mutual interaction of two  $60^\circ$  dislocations during structural relaxation that has already been pointed out in part II (see figure 3*a–e* in part II), a rare instance of stress initiated glide is shown in the deformation sequence of figure 24*a–d*, in which the stages in the mat are seen between a total shear strain of 0.05–0.20. In figure 24*a*, a closely spaced dipole of dislocations is seen in the region marked as A. In the successive frames, the dipole is separated, and the two dislocations are driven apart. In figure 24*b*, the positions of the dislocations are marked with arrows. In figure 24*c*, the dislocation on the right has already been incorporated into a boundary, while the one on the left has entered the simulation cell on the other side, and is shown with an arrow. In figure 24*d*, this dislocation too seemed to have been trapped by a boundary that has reached it. The shear strain contributed to the total strain in the simulation cell can readily be estimated as  $bx/A$ , where  $x$  is the total distance of separation of the dislocations, i.e. about 20 Voronoi polygons,  $b$  is the Burgers vector, and  $A$  the area of the mat, i.e. 780 Voronoi polygons. This gives a strain contribution of  $20/780 = 0.0256$ , which makes up about 11% of the total inelastic strain in the simulation, and thus is not a negligible amount. There were, however, no other unambiguous glide events that could be found. There are two reasons why such events are not more numerous. First, the dislocation glide appears to be

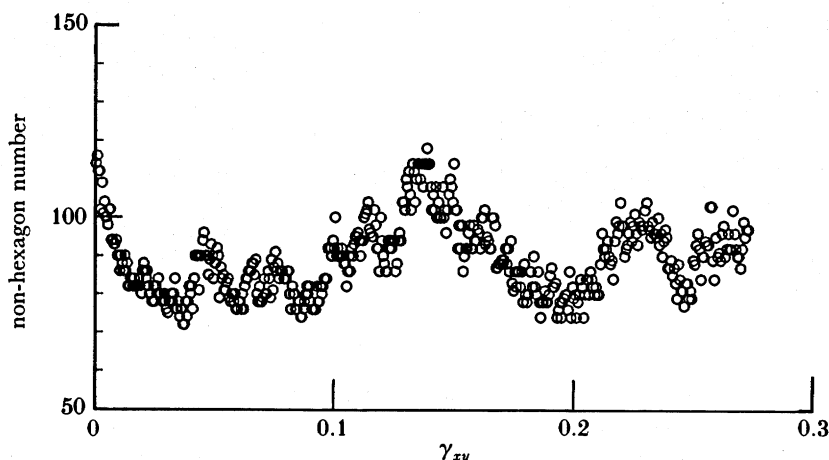


FIGURE 25. Variation with strain of the total number of liquid-like material polygons with edges different than six.

subject to a significant lattice drag, indicated by the many metastable configurations of two adjoining polygons of five sides, and second, the near impossibility of operating dislocation sources at the lower stress levels within the confines of the very small quasi-ordered regions. Thus, the shear transformations dominate the deformation even though they have no mobile character, but have the overwhelming advantage of a large volume fraction of fertile material, where they can be initiated.

Figure 24*a–d* also shows that as the deformation proceeds, the liquid-like material itself deforms and reconstitutes itself in a non-affine manner. In fact, the overall fraction of the liquid-like material itself varies in a fluctuating way, as the shearing continues. This is shown in figure 25, where the total numbers of non-hexagonal Voronoi polygons are plotted as a function of strain. The figure shows that the initial high concentration of this fertile material is depleted with shear in the apparent elastic range of deformation. Further shearing produces relatively large amplitude variations due to the shear induced dilatations and compactions. Around a strain of 0.15, a significant increase in the fraction of this material takes place. This coincides with the dip in the flow stress, shown in figure 7, and is most likely the explanation for it.

This research has been supported by several sources. The early phases of the simulation on the smaller periodic cells were supported by the NSF through Grant No. 85-17224-DMR, while the latter stages of the simulation on the large simulation cells requiring greatly increased levels of computations, became possible through the support of the Defense Advanced Research Projects Agency under Contract N00014-86-K-0768. Additional salary support for D. D. was provided by the Allied Signal Corporation through a fellowship, for which we are grateful to Dr Lance Davis. We also acknowledge a number of useful discussions with Professor D. M. Parks on presentation of strain spikes. S. Y. also acknowledges the support of NSF under grants CHE-84-15078 and CHE-88-06767.

#### APPENDIX 1. FREE ENERGY CHANGES DUE TO TRANSFORMATIONS

A shear transformation changes the Gibbs free energy of the simulation cell in a well-known way (Kocks *et al.* 1975), as

$$\Delta G = \Delta F - \Delta W, \quad (\text{A } 1)$$

where  $\Delta F$  is the Helmholtz free energy change in the transformation, and  $\Delta W$  the work done as the external tractions are displaced due to the local transformation. Expansion of the expression for the Helmholtz free energy in (A 1) gives

$$\Delta F = \Delta U - T\Delta S = \Delta E_k + \Delta V - T\Delta S, \quad (\text{A } 2)$$

where  $\Delta U$  is the internal energy change which can be divided further into a change in potential energy  $\Delta V$ , and kinetic energy change  $\Delta E_k$  of the atoms affected by the transformation. The entropy change  $\Delta S$  is primarily of a vibrational nature. In the MD method, the  $\Delta E_k$  is given by

$$\Delta E_k = \frac{1}{2}m \sum_{i=1}^n \frac{1}{N\tau} \int_0^{N\tau} v_i^2(t) dt. \quad (\text{A } 3)$$

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