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LETTER TO THE EDITOR

Avalanche-like behaviour of a model liquid: how a dense liquid flows

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Abstract. Samplings of potential energy minima of a two-dimensional model of a binary random alloy ($A_{64}B_{36}$) are performed in two distinct ways. The first method uses successive local changes (nearest-neighbour AB switches) to produce a series of energy minima. The second method uses successive global changes (shear strain) to produce a series of minima. This material is amorphous and it is argued that each of the two methods samples a region of configuration space large enough to allow the material to flow. That is, each procedure samples states of a model liquid. For each of these two sampling methods the statistics of the changes in atomic configuration are similar to the avalanche statistics of models of self-organized critical phenomena. This leads naturally to a simple picture of how a dense liquid may flow.

The dynamics of liquids [1] is complicated by the existence of two distinct time scales. Over short times the motion of the atoms in liquids is comparable to that of atoms in crystalline solids. The atoms rattle around in ‘cages’ created by their neighbours but do not move from one neighbourhood to another. This caged motion apparently includes most of the degrees of freedom of the liquid so that it leads to the well known result that molar specific heats of simple liquids are comparable to the Dulong–Petit specific heats of simple solids. On the other hand, the atomic motion in liquids over long time scales is quite complex, as atoms move from one neighbourhood to another. These changes in the topology of the atoms in a liquid are arguably the distinguishing characteristic of a liquid, since they lead to the macroscopic feature which distinguishes liquids from solids, the ability to flow.

Most of the progress in the theory of liquids, both analytically [2] and with simulations [3, 4], has come from studies which include both of these time scales at once. In particular, the simulations may allow one to understand the detailed atomic motion which takes place during a change from one topologically distinct atomic configuration to another. For instance, recent molecular dynamics (MD) simulations [4] have shown that, for liquids at low temperatures, changes in atomic configuration are accompanied by one dimensional (string-like) regions of mobile atoms with correlated motion. Computer simulations are well characterized methods for sampling the atomic configuration space but are perhaps most important for the fact that they yield predictions for thermodynamic quantities. In this Letter, I want to discuss two other samplings of the space of atomic configurations of a liquid and, in the process, relate the configuration changes to the so-called self-organized critical (SOC) systems [5]. In particular, in this Letter I report the statistics of the sizes of the atomic rearrangements and show that they follow a power law.

A two-dimensional model is chosen for computational and viewing ease and because liquids seem to be as common in two-dimensional matter as in three dimensions. The particular

model studied here is a two-component random alloy similar to models previously used to study glasses [6]. For the purposes of this Letter I will distinguish liquids from glasses using Kauzmann's [7] point that a real glass is simply a liquid which is limited, because of its low temperature, in the range of atomic configurations it may sample during the time scale of an experiment. This makes a glass unable to flow, and so it is solid. In this study the material apparently samples a large enough set of atomic configurations to allow it to flow, so I consider the sampling to be of the configuration space of a liquid.

The model [8] used is a two-dimensional random alloy of large A atoms and small B atoms with atomic size ratio = (radius of B)/(radius of A) = 0.75. The i th atom is bound to the j th atom by the Lennard-Jones pair potential

$$E_{ij} = 4(\varepsilon)((\sigma_{ij}/r_{ij})^{12} - (\sigma_{ij}/r_{ij})^6)$$

where r_{ij} is the distance between the atoms, $\sigma_{ij} = \sigma_{AA} = 1$ if both i and j are A atoms, similarly $\sigma_{BB} = 0.75$ and $\sigma_{AB} = \sigma_{BA} = (\sigma_{AA} + \sigma_{BB})/2$. All energies are given in units of ε and distances in units of σ_{AA} . To speed up calculations the pair potential is cut off at $r_{ij} = 3.5$. The system consists of either 1024 atoms or 4096 atoms[†] with A and B atoms originally placed randomly on a hexagonal lattice. The concentration of B atoms is c . To minimize the effects of sample size, periodic boundary conditions are used. The simulation cell is chosen to be initially rectangular (shear angle = $\theta = 0$) with $R = (\text{cell height}/\text{cell width}) = \sin(\pi/3)$. The energy of the system, $E = \sum_i \sum_{j>i} E_{ij}$, is then minimized (with respect to all atomic position and cell size and shape parameters[‡]) to one part in 10^7 using standard techniques [9]. The energy minimization allows the atoms to adjust their positions away from their initial lattice points. For small concentrations of B atoms (c less than 20–25%) the resulting material has the same type of long-range bond-orientational order and quasi-long-range translational order (power-law decay in real space) [10] as a single component two-dimensional crystal at low T [11]. However, for c greater than about 25–30% the atoms move around sufficiently in the initial relaxation so that no long-range translational or bond-orientational order is left [8, 10]. That is, these materials are amorphous. In this Letter I will focus on $c = 36\%$, which is well into the amorphous regime and is the concentration I have studied the most. However, I have examined the configuration states of a series of samples with compositions $3\% \leq c \leq 42\%$ and have found that all of the amorphous samples behave in the same fashion as $c = 36\%$. The crystalline samples will be the subject of another paper [12].

The point of this study is to examine the statistics of changes in atomic configuration which occur when the material moves from one local potential energy minimum to a nearby minimum. I have chosen two distinct methods to produce these successive energy minima. For the first method the material is driven from one local energy minimum to another by (i) randomly choosing a nearest-neighbour AB pair of atoms and turning the A atom into a B and the B atom into an A, and (ii) minimizing E again with respect to all of the atomic position and cell shape parameters. I change the radii of the atoms rather than switching them so that *all atomic motion takes place in the relaxation process* and no atoms are moved arbitrarily. Throughout the rest of this Letter, reference to AB switches will imply the subsequent energy minimization. In the second method the configuration space is sampled by (i) shearing the material by changing the ratio, R , of the height of the cell to the width, and (ii) minimizing the energy with respect to all atomic position and cell shape parameters except R . Again,

[†] There do not seem to be qualitative differences in the two different size samples so I will generally use the largest lattice which allows sampling of a large enough set of configuration states in a reasonable time.

[‡] Minimizing the energy with respect to simulation cell size and shape parameters implies that one is working at zero pressure and zero stress. I have checked that the resulting statistics are the same if one instead leaves the simulation cell shape parameters fixed.

throughout the rest of this Letter, unless otherwise noted, any discussion of steps in R will imply the subsequent energy minimization. This second method is essentially changing the shape of the container holding the sample. These two methods have obvious analogues in the sandpile models. In sandpile models [5] (as opposed to actual sandpiles) one considers a lattice of sandpile 'heights' where the pile at a lattice point 'topples' when it is larger than some fixed value. One can drive the system into a critical state either by increasing the pile heights at random lattice positions (analogous to the local AB switches) or by increasing the pile height at every lattice position simultaneously (analogous to the macroscopic strain) [13]. For sandpile models both methods lead to power-law distributions of avalanche sizes.

First I will discuss results from the local method on simulation samples with 1024 atoms. First, 100 AB switches are performed to verify that the system is in a stable state[†] with respect to these switches. Then 2000 switches are carried out while monitoring E , θ , R and the cell volume. It is found that all of these macroscopic quantities fluctuate around well defined averages. For instance, the energy per atom for $c = 36\%$ is -3.113 ± 0.005 . In analogy with sandpile models [4, 13], one can define an avalanche by counting the number of atoms, n , which move more than a given cut-off distance. One needs to define an avalanche in terms of a cut-off distance because all of the atoms move a finite amount in each rearrangement. If the system is in a critical state then there is no natural length scale in the problem and the shape of the resulting distribution, $P(n)$, follows a power law $P(n) \propto (n)^{-\nu}$ where ν is of order one. In figure 1 one sees that such a power-law tail is consistent with the avalanche results.

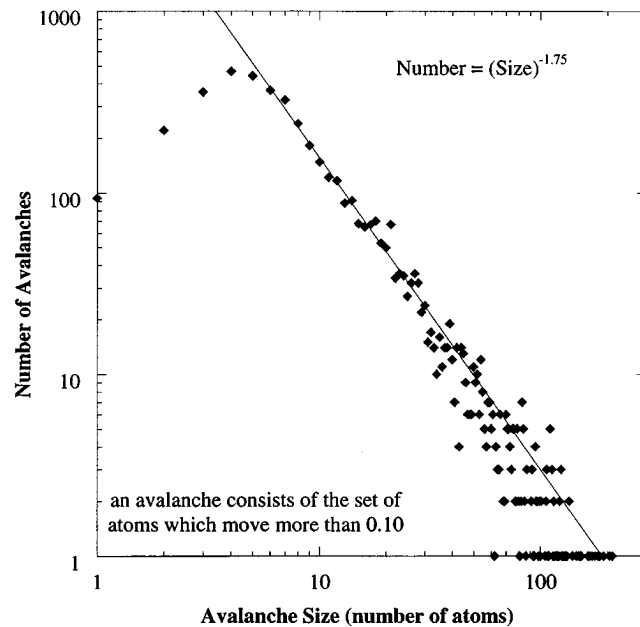


Figure 1. Power-law distribution of avalanche sizes for amorphous $A_{64}B_{36}$ under AB switches. The line is a power-law fit to the data for avalanche sizes between 6 atoms and 200 atoms. Included are data for 4500 steps (three separate runs of 2000, 2000, and 500 steps).

The energy states sampled by straining the material can be analysed in a similar fashion in terms of avalanches. However, since R must be changed in finite steps, one must first decide

[†] There are very small bond-orientational correlations (calculated as described in [8]) for $c = 36\%$ which decay away in the first 50 switches.

what size steps in R to take. One of the sampled states from the AB switches is chosen to be the starting state. Then the ratio of cell height to cell width, R , is changed from about 0.86 to about 0.93 in six different ways; (i) 7 steps of $\Delta R = 0.01$, (ii) 21 steps of 0.003, (iii) 70 steps of 0.001, (iv) 210 steps of 0.0003, (v) 700 steps of 0.0001, and (vi) 2100 steps of 0.00003. The resulting energies are shown in figure 2. For the largest steps the energy increases quadratically (as an elastic material) up to a strain, $\Delta R/R$ of about 0.05, after which the response is clearly plastic. As the step size of R is decreased (smaller strain rate) the response at low strain becomes less and less elastic-like as sharp drops in energy are seen superimposed on the mainly quadratic background. The states sampled at the three smallest strain rates show no quadratic region, but the energy versus strain curves are similar, leading one to the idea that these strain rates will yield approximately the same avalanche distributions.

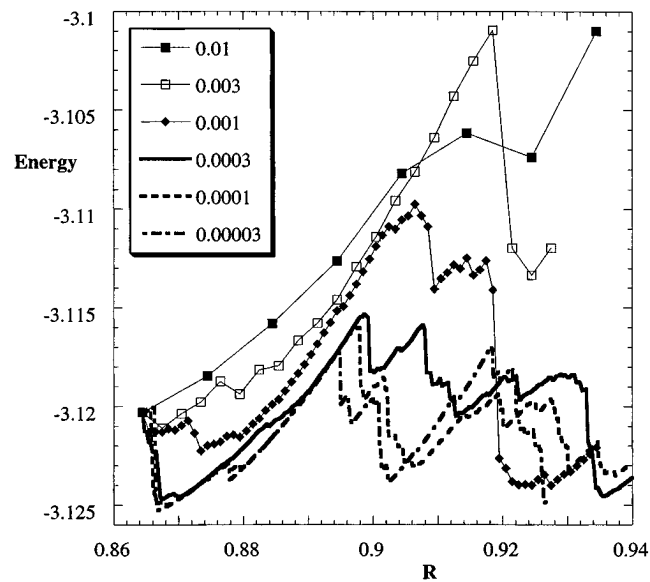


Figure 2. Energy of the amorphous alloy as a function of R , the ratio of height to width of the simulation cell. The ratio, R , was changed from small R to larger R in steps of 0.01, 0.003, 0.001, 0.0003, 0.0001, and 0.00003 as shown in the figure.

In order to sample a large enough set of states to get an idea of the statistics of the avalanches, 2800 steps of $\Delta R = 0.0001$ are taken for each of three distinct model materials with 1024 atoms, and one set of 2800 steps is taken for a sample with 4096 atoms. The avalanche size is, again, the number of atoms which move more than a given cut-off distance. However, as for the AB switches I only consider the distance an atom moves in the relaxation process. The shift of atoms which is put in by hand (shearing of the simulation cell) is not considered in measuring the avalanche size. The resulting size distribution is shown in figure 3. Again, the resulting distribution follows a power law.

Before proceeding with a discussion of these results I will address the question of how this sampling of states is related to the configuration states of a liquid. A liquid at a finite temperature certainly cannot sample all of configuration space in a finite time. However, as discussed above, a liquid is able to sample states over a large enough volume of configuration space, Ω_{sample} , that it can flow (change its shape). One might suppose that there is a roughly defined minimum configuration space volume, Ω_{liquid} , such that for $\Omega_{sample} > \Omega_{liquid}$ the material behaves as a liquid and for $\Omega_{sample} < \Omega_{liquid}$ the material behaves as a glass. Do the

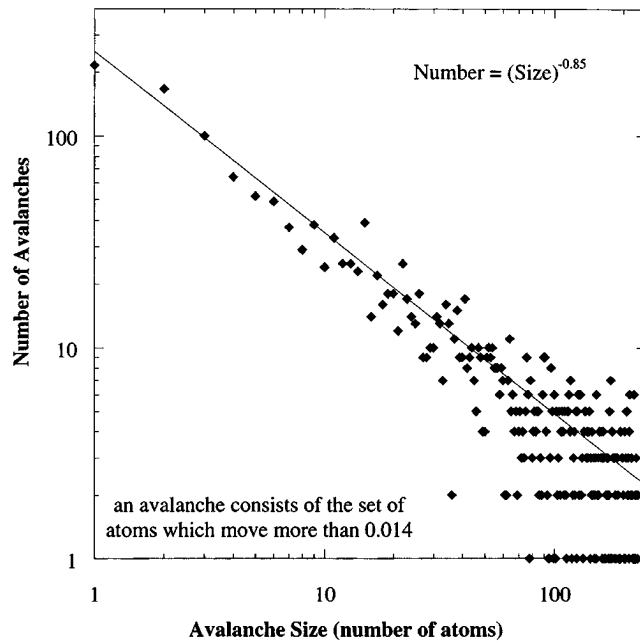


Figure 3. Power-law distribution of avalanche sizes for amorphous $A_{64}B_{36}$ under steps in R (the height to width ratio of the simulation cell) of $\Delta R = 0.0001$ per step. The line is a power-law fit to the data for avalanche sizes between 1 atom and 200 atoms.

two methods that I have used sample states over a volume of configuration space, Ω_{sample} , such that $\Omega_{sample} > \Omega_{liquid}$? For the strain method of sampling states I am, in fact, changing the shape of the container so the sample does flow, and Ω_{sample} must be larger than Ω_{liquid} . There are three reasons why I think that the same holds true for the AB switch sampling method. First, the atomic diffusion is just as large with this method as with the strain method. By that I mean that when I calculate the mean square atomic deviation, $\langle \delta r^2 \rangle$ (which equals the square of the distance an atom has moved from its initial position, averaged over all atoms), I find that $\langle \delta r^2 \rangle \sim 1.8$ after 2000 AB switches but $\langle \delta r^2 \rangle \sim 0.5$ after the 2800 steps of $\Delta R = 0.0001$. Large diffusion implies small viscosity. Second, the range of state energies spanned by this method is at least as large as for the strain method. Finally, if a sample is rapidly strained to a high energy and then AB switches are made with the sample shape held constant, the energy of the sample relaxes back to the unstrained energy. That is, the switches allow the sample to flow. So it seems that $\Omega_{sample} > \Omega_{liquid}$ for both sampling methods and both of them are sampling states of a liquid.

The results of this sampling of liquid configuration states suggest that, at the atomic level, the flow of liquids is an intermittent process. At low temperatures liquids behave as SOC systems with highly correlated, essentially irreversible, atomic movements (avalanches) triggered by thermal fluctuations. The backbones of the largest avalanches, consisting of the atoms which move the most, are one-dimensional in nature [10]. This leads to the possible identification of these avalanche backbones with the correlated strings of atoms seen in MD simulations. At very high temperatures, the thermally triggered avalanches overlap each other sufficiently that the atomic motion is essentially uncorrelated Brownian motion as in a gas.

Finally, I should add the caveat that the potential energies used in this model do not include the angular constraints of covalent bonding, so it is not an appropriate phenomenological model for covalently-bonded materials even if they also turn out to be SOC systems.

I would like to thank W L Johnson and M Li for introducing me to the subject of alloy glasses and for very useful discussions.

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