

On the way to a Lattice Boltzmann Model for glassy systems?

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1. – Introduction

The behavior of glassy materials and related disordered systems is one of the most debated and still open problems in modern condensed matter physics. Glasses are undercooled liquids which show a spectacular increase in viscosity (14 orders of magnitude) under rapid cooling below a given temperature threshold T_g , known as the glass-transition temperature. In particular, they exhibit anomalous relaxation, namely long-lived departures from local equilibria, which result from the trapping of atoms around fixed positions in space according to a disordered pattern. The numerical simulation of glassy behavior relies mainly on Molecular Dynamics and Monte Carlo techniques [1, 2]. Both methods incorporate details of microscopic behavior (intermolecular potentials) and, as a result, they fall short of reaching spatio-temporal scales of macroscopic interest. Coarse-grained lattice gas models represent a step ahead in reaching larger scales: On the one hand there are spin-glasses [3, 4] and on the other their modern lattice glass variants [5, 6]. Clearly, these lattice models cannot reach the same degree of physical fidelity as truly atomistic methods.

It is in the spirit of mesoscopic models that the lattice Boltzmann equation (LBE) was

originally introduced for simulating hydrodynamic behavior in ordinary fluids [7]. Over the years, LBE has proved quite flexible in describing situations beyond the mere fluid dynamics framework [8]. It appears therefore reasonable to explore whether LBE can be extended in such a way to cover glassy behavior as well. We present in the following a lattice Boltzmann model which allows to catch some features of glassy systems [9].

2. – The Model

We use a lattice Boltzmann equation with a single relaxation parameter [10] which reads as follows:

$$(1) \quad f_i(\mathbf{r} + \mathbf{c}_i, t + 1) - f_i(\mathbf{r}, t) = -\omega (f_i(\mathbf{r}, t) - f_i^{eq}(\mathbf{r}, t))$$

where $f_i(\mathbf{r}, t) \equiv f(\mathbf{r}, \mathbf{v} = \mathbf{c}_i, t)$ is a discrete distribution function of particles moving along the direction i with discrete speed \mathbf{c}_i . We use a nine-velocity model on a square lattice with $c_i = (\pm 1, 0), (0, \pm 1), (\pm 1, \pm 1)$. The right-hand side represents the relaxation to a local equilibrium f_i^{eq} in a time lapse of the order of ω^{-1} . This local equilibrium is taken in the form of a quadratic expansion of a local Maxwellian:

$$(2) \quad f_i^{eq} = \rho w_i \left[1 + \frac{\mathbf{u} \cdot \mathbf{c}_i}{c_s^2} + \frac{\mathbf{u}\mathbf{u} \cdot (\mathbf{c}_i \mathbf{c}_i - c_s^2 I)}{2c_s^4} \right]$$

where w_i is a set of weights normalized to unity, c_s the lattice sound speed (a constant equal to $1/\sqrt{3}$ in our case) and, finally, I stands for the unit tensor. The distribution functions are related to the fluid density ρ and to the fluid momentum $\rho \mathbf{u}$ by

$$(3) \quad \rho = \sum_i f_i, \quad \rho \mathbf{u} = \sum_i f_i \mathbf{c}_i.$$

In order to describe light and heavy phases with a single species fluid we follow the approach of Shan and Chen [11] and introduce a non ideal-gas behavior within the LBE formalism by means of a phenomenological pseudo-potential which describes the effects of potential energy interactions.

We introduce a repulsive Lennard-Jones potential interaction, which takes the following expression on the lattice:

$$(4) \quad V(r) = \begin{cases} G\Psi(\rho)r^{-12} & \text{if } 1 \leq r \leq \sqrt{2} \\ 0 & \text{otherwise} \end{cases}$$

where G is a positive parameter controlling the strength of the interaction. The structure of $\Psi(\rho)$ is in the form of nested polynomials, whose density zeroes are distributed according to a binary tree whose depth depends on the number of generations N_g :

$$(5) \quad \Psi(\rho) = (\rho - \rho_0) \prod_{g=2}^{N_g} \frac{-1}{\left(1 + \left(\frac{\rho - \rho_0}{\rho_0/2}\right)^{2^{g-1}}\right)^2} \prod_{k=1}^{2^{g-1}} \frac{(\rho - \rho_{gk})}{(\rho_{gk} - \rho_0)}$$

where $\bar{\rho}_0$ controls the density gap. This hierarchical potential implements the presence of $N_\rho = 2^{N_g} - 1$ competing density extrema.

The denominator in Eq. (4) is such to let $\Psi \rightarrow 0$ outside the hierarchical range of zeroes ρ_{gk} . The result is an effective potential alternating stable repulsive regions ($\Psi' = \frac{\partial \Psi}{\partial \rho} > 0$) with unstable attractive ones ($\Psi' < 0$), distributed approximately in the density range $\rho_{\mp} = \rho_0(1 \mp \bar{\rho}_0(1 - 2^{-N_g}))$. The presence of unstable regions causes the appearance of phase transitions which determine the separation of light and heavy phases inside the fluid. In this case we should not observe the typical patterns of phase separation in binary fluids where coherent regions of the two phases grow in time until they span completely the system. In a mode-coupling perspective [12], we expect to be able to generate a disordered short-range coexistence of two competing species: "voids" (regions with $\rho < \rho_0$) and "cages" (regions with $\rho > \rho_0$). In real glasses, the dense phase is responsible for cage-effects which ultimately lead to mechanical arrest of the system with an extraordinary increase of the viscosity. The glassy LBE takes the final form

$$(6) \quad f_i(\mathbf{r} + \mathbf{c}_i, t + 1) - f_i(\mathbf{r}, t) = -\omega(f_i(\mathbf{r}, t) - f_i^{eq}(\mathbf{r}, t)) + F_i(\mathbf{r}, t)$$

where $F_i = -G \frac{\rho}{\rho_0} \nabla \Psi(\rho) \cdot \mathbf{c}_i$.

3. – Numerical results

We have simulated the fluid on a 2D lattice of size 256×256 with the following parameters: $\omega = 1.0$, $\rho_0 = 1.0$, $N_g = 3$, $\bar{\rho}_0 = 0.5$, G is varied in the range $[0, 1]$. The initial condition is $\rho(\mathbf{r}) = \rho_0(1 + \xi)$ where ξ is a random perturbation uniformly distributed in the range $[-0.01, 0.01]$. The effects due to propagation (left hand side of Eq. (6)) between neighboring sites and to the non-linear potential $\Psi(\rho)$ make the density distribution to spread out in time, so that an instability is triggered as soon as G exceeds the critical threshold G_c . Such a value can be found by solving the systems of equations

$$(7) \quad \begin{cases} \frac{\partial p'}{\partial \rho} = 0 \\ \frac{\partial^2 p'}{\partial \rho^2} = 0 \end{cases}$$

where $p' = c_s^2 \rho + V$ is the equation of state when the potential (4) is switched on.

Solving the system (7) we find $G_c \sim 0.089$. Numerically we can verify this prediction by monitoring the value of G for which the quantity $\Delta\rho \equiv \rho_{max} - \rho_{min}$ becomes non zero, ρ_{max} and ρ_{min} being the maximum and minimum values of ρ , respectively. The density gap $\Delta\rho$ starts up at $G = G_c \sim 0.09$ in good agreement with the analytical result and rapidly fills the available range of states between ρ_- and ρ_+ , as the interaction strength is increased. The density ρ distributes in time within the available range of values according to a Gaussian distribution, characterized by decreasing fluctuations around the mean value as time goes on.

In order to check whether we are simulating a phase-separating fluid or a glassy system we looked at the spatial distribution of the density field $\rho(x, y)$. In Fig. 1 we present

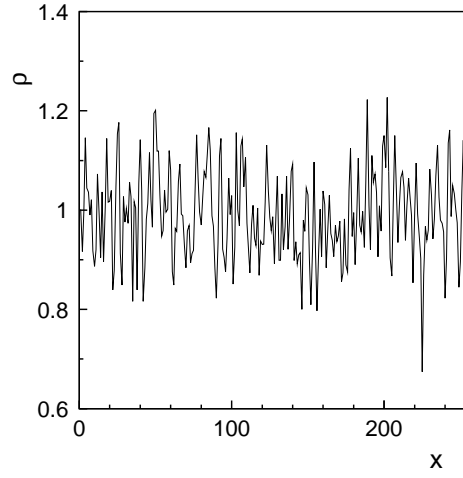


Fig. 1. – The density profile along the x -direction at $y = 128$ at $t = 200$ for the case $G = 0.45$.

a cross section of the density field ρ taken at $y = 128$ at time $t = 200$ (steady state). The pictures clearly indicate a disordered coexistence of voids and cages. This confirms that the glassy potential fulfils the minimal requisite for glassy-behavior: sharp density contrasts over a disordered spatial pattern.

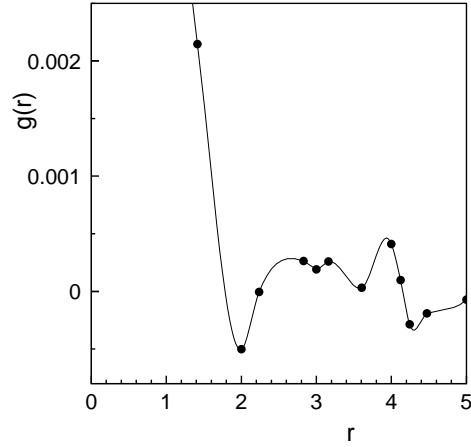


Fig. 2. – Density-density correlation function at $t = 200$.

As a further observable in Fig. 2 we plot the density-density correlation:

$$(8) \quad g(r) = \frac{\langle \delta\rho(x+r_x, y+r_y)\delta\rho(x, y) \rangle}{\langle \rho(x, y)\rho(x, y) \rangle},$$

where $\delta\rho \equiv \rho - \langle \rho \rangle$ is the density fluctuation around its spatial average and $r^2 = r_x^2 + r_y^2$. This function shows an oscillating-decay behavior similar to the case of real glasses. Being in a lattice, it is clear that data are only available at discrete positions $1, \sqrt{2}, 2, \dots$. However, a spline interpolation (solid line) indicates that the observed oscillations are not a numerical artifact. In the case of no glassy potential ($G = 0$), $g(r)$ is virtually zero as a consequence of the uniform and constant density distribution. In Fig. 3 we present the density-density time-correlation function $h(\tau)$, taken at $x = 128, y = 128$. Here, the analogy with glasses is weaker. Instead of anomalous slow relaxation, we find persistent oscillations around zero, possibly reflecting an everlasting competition between the various density minima. This is at odd with the typical signature of glassy behaviour,

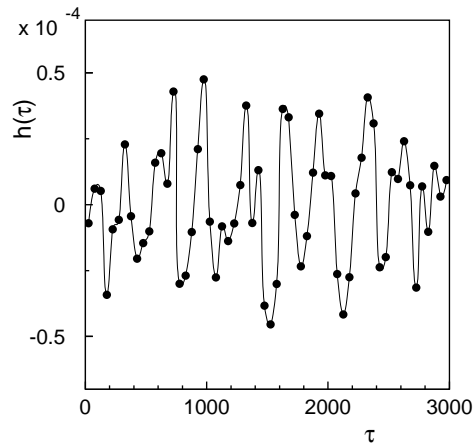


Fig. 3. – Density-density time correlation function.

namely a plateau in the time correlation function. To this purpose, further research work is required.

4. – Conclusion

The proposed glassy LBE produces a number of encouraging results. Geometrical frustration is qualitatively captured by a hierarchical density-dependent potential, alternating repulsive and attractive interactions. This potential gives rise to disordered patterns with short-range, sharp density contrasts. It also yields qualitatively reasonable spatial density-density correlation functions. However, anomalous time relaxation is not captured. We believe that the introduction of a conditional propagation is a route to be explored in future work: possible strategies include a density-dependent relaxation frequency ω and/or density-dependent site-to-site hopping rates [13].

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