R lab simulation seminar
Chapter 1: Simulations of glasses

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Agenda

Sec. 1

Glass simulations

1.1 Set up

Consider a two-dimensional binary particle mixture, composed of two species of particles. Set the particle size ratio to be $a_0 : a_1 = 1 : 1.4$, the particle number ratio to be $N_1 : N_2 = 1 : 1$, the particle number to be $N = N_1 + N_2 = 4000$, and the particle number density to be $\rho = \frac{N}{L^2} = 0.8$. The motion of the particle j is driven by the following Langevin equation:

$$m \frac{d\mathbf{v}_j(t)}{dt} = -\zeta \mathbf{v}_j(t) + \mathbf{F}_j^{\mathrm{I}}(t) + \mathbf{F}_j^{\mathrm{B}}(t).$$

Here, the thermal fluctuation   force acting on a particle satisfies the following fluctuation dissipation theorem :

$$\langle \mathbf{F}_{\mathrm{B}}(t)\mathbf{F}_{\mathrm{B}}(t')\rangle = 2k_B T\zeta\delta(t - t')\mathbf{1}.$$

As the interparticle potential, use the following Bernu-Hansen-Hiwatari-Pastore (BHHP) potential,

$$U(r_{jk}) = \epsilon\left(\frac{a_{jk}}{r_{jk}}\right)^{12} + C_{jk} \quad (r_{jk} < a_{\mathrm{cut}}).$$

The cut-off length is set to be $a_{\mathrm{cut}} = 3.0a_0$. The interaction force is $\mathbf{F}_j^{\mathrm{I}}(t) = -\sum_k \frac{\partial U(r_{jk})}{\partial \mathbf{r}_j}$. Now the time unit is given as $t_0 = \sqrt{ma_0^2/\epsilon}$, the length scale as a_0, and the temperature as $k_B T/\epsilon = 1$. By following the setting, evaluate the physical properties when the temperature is changed. In particular compute the radial distribution function, the static structure factor, the mean-square displacement, and the intermediate scattering function at temperatures $T = 1.0,\ 0.72,\ 0.64,\ 0.56$.

1.2 Radial distribution function

- The radial distribution function is the most frequently used analytical function for investigating the structure of liquids and solids.
- It is a two bodies density correlation function and is expressed as follows

$$g(\mathbf{r}) = \frac{1}{\rho N} \left\langle \sum_{j=1}^{N} \sum_{k=1}^{N} {}' \delta(\mathbf{r} - \mathbf{r}_j + \mathbf{r}_k) \right\rangle$$

$\sum_{k=1}^{N} {}'$ means $k = j$ is excluded.

- Now let us suppose the system is homogeneous and isotropic.
- Here, when $N_j(r)$ is defined to be the number of other particles contained on a spherical shell at a distance r from the position of the particle j, the radial distribution function becomes as follows.

$$g(r) = \frac{1}{N} \left\langle \sum_{j=1}^{N} \frac{\Delta N_j(r)}{4\pi r^2 \Delta r \rho} \right\rangle$$

where ρ is the number density N/V.
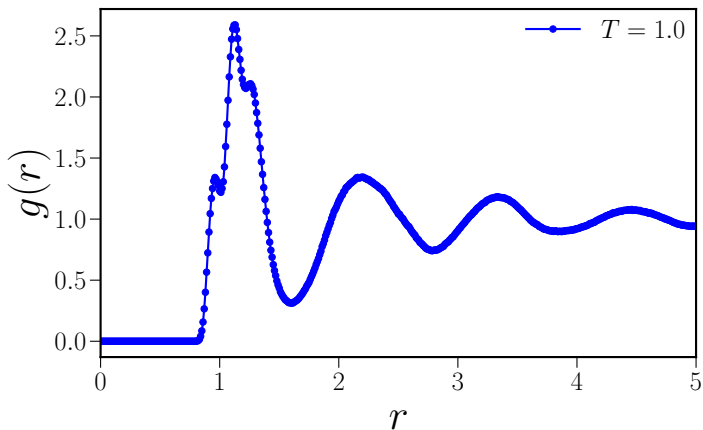
1.2 Radial distribution function (2)

FIG 1: A radial distribution function $g(r)$ obtained from the HW3 data at $T = 1.0$.

1.3 Static structure factor

- When examining the static structure of particles by scattering experiments, etc., Fourier transformed quantities are often used.
- The most basic function is the static structure factor

$$S(q) = \frac{1}{N} \left\langle \tilde{\rho}(q)\tilde{\rho}(-q) \right\rangle.$$

- $S(q)$ is the Fourier transform of $g(\mathbf{r}) - 1$ in space.
- When calculating $S(q)$ numerically, Fourier transformation of the density field $\rho(\mathbf{r}) = \sum_{j=1}^{N} \delta(\mathbf{r} - \mathbf{r}_j)$ is treated as follows:

$$\tilde{\rho}(q) = \int d\mathbf{r}\rho(\mathbf{r})e^{-i\mathbf{q}\cdot\mathbf{r}} = \sum_{j=1}^{N} e^{-i\mathbf{q}\cdot\mathbf{r}_j}$$

- And then we obtain $S(q)$ as

$$S(q) = \frac{1}{N} \left\langle \sum_{j=1}^{N} e^{-i\mathbf{q}\cdot\mathbf{r}_j} \sum_{j=1}^{N} e^{i\mathbf{q}\cdot\mathbf{r}_j} \right\rangle = \frac{1}{N} \left\langle \left(\sum_{j=1}^{N} \cos{(\mathbf{q}\cdot\mathbf{r}_j)}\right)^2 + \left(\sum_{j=1}^{N} \sin{(\mathbf{q}\cdot\mathbf{r}_j)}\right)^2 \right\rangle$$
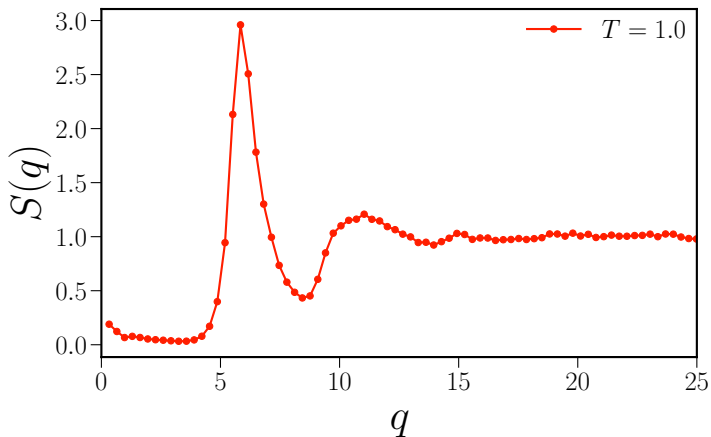
1.3 Static structure factor (2)

FIG 2: A static structure factor $S(q)$ obtained from the HW3 data at $T = 1.0$.

1.4 Intermediate scattering function

- Next, we introduce the method for measuring the relaxation of the particle structure.
- The density field at time t is represented as $\rho(\mathbf{r}) = \sum_{j=1}^{N} \delta(\mathbf{r} - \mathbf{r}_j)$.
- As with the static structure factor, if we Fourier-transform the density field in space, we obtain $\tilde{\rho}(q,t) = \int d\mathbf{r} \rho(\mathbf{r}) e^{-i\mathbf{q}\cdot\mathbf{r}(t)} = \sum_{j=1}^{N} e^{-i\mathbf{q}\cdot\mathbf{r}_j(t)}$.
- Here, the characteristic time in which the correlation of the Fourier transformed density field decreases with time is the structure relaxation time corresponding to the wave number q.
- To measure the structural relaxation time, we introduce the following intermediate scattering function, $F(q,t)$ such that

$$
\begin{aligned}
F(q,t) &= \frac{1}{N} \left\langle \sum_{j=1}^{N} e^{-i\mathbf{q}\cdot\mathbf{r}_j(t)} \sum_{j=1}^{N} e^{i\mathbf{q}\cdot\mathbf{r}_j(0)} \right\rangle \\
&= \frac{1}{N} \left\langle \sum_{j=1}^{N} \cos\left(\mathbf{q}\cdot\mathbf{r}_j(t)\right) \sum_{j=1}^{N} \cos\left(\mathbf{q}\cdot\mathbf{r}_j(0)\right) + \sum_{j=1}^{N} \sin\left(\mathbf{q}\cdot\mathbf{r}_j(t)\right) \sum_{j=1}^{N} \sin\left(\mathbf{q}\cdot\mathbf{r}_j(0)\right) \right\rangle.
\end{aligned}
$$

- Here as with $S(q)$, Fourier transforms can be performed analytically.

1.4 Intermediate scattering function (2)

- The following self-intermediate scattering functions, i.e., taking only the self term of the intermediate scattering function, are commonly used:

$$
\begin{aligned}
F_s(q,t) &= \frac{1}{N}\left\langle \sum_{j=1}^{N} e^{-i\mathbf{q}\cdot(\mathbf{r}_j(t)-\mathbf{r}_j(0))}\right\rangle \\
&= \frac{1}{N}\left\langle \sum_{j=1}^{N} \cos\left(\mathbf{q}\cdot\mathbf{r}_j(t)\right)\cos\left(\mathbf{q}\cdot\mathbf{r}_j(0)\right) + \sum_{j=1}^{N}\sin\left(\mathbf{q}\cdot\mathbf{r}_j(t)\right)\sin\left(\mathbf{q}\cdot\mathbf{r}_j(0)\right)\right\rangle
\end{aligned}
$$

- Especially, in the simple liquid in which the cooperativity of particle motion, etc. is not often observed, the result is almost the same as the whole intermediate scattering function. The first peak of the static structure factor $q = 2\pi/a$ (a is the diameter of the particle) is usually used as the wavenumber to study the structural relaxation of liquids.

1.5 Logarithmic sampling

- When calculating the time correlation function，it is better to store the data by the time interval in geometric series (Useful for analysis over a wide range of time).
- After sampling for a fixed time, the same sampling is repeated again, and the time average can be taken.

LIST 1: logarithmic sampling

```
1    // add in the time development loop:
2    if(int(t/dt) == int((sampling_time + time_stamp)/dt)){
3       output_t(x,y,avU,avK,Np,t,time_stamp,x_corr,y_corr);
4       sampling_time*=pow(10.,0.1);
5       sampling_time=int(sampling_time/dt)*dt;
6       if(sampling_time > sampling_time_max/pow(10.,0.1)){
7       time_stamp=t;
8       sampling_time=5.*dt;
9       }
10    }
```

1.6 Center of mass correction

- Under the periodic boundary condition of finite size, a motion called Goldstone mode occurs in which **all** particles move together.
- Such motion disappears in the thermodynamic limit of infinite system size.
- It affects the calculation of the density relaxation of the particles.
- Even if the structure does not change much, the particles appear to be relaxed since they move all together.
- By subtracting the center of mass motion, "genuine structural relaxation" can be captured.

LIST 2: Center of mass correction

```
1   int com_correction(double *x,double *y,double *x_corr,double *y_corr,int Np,double L){
2   int i;
3   double dx,dy;
4   static double x0[Npm],y0[Npm];
5   static bool IsFirst = true;
6   if(IsFirst){
7     for(i=0;i<Np;i++){
8       x0[i]=x[i];
9       y0[i]=y[i];
10    }
11    IsFirst = false;
12  }
13
14  for(i=0;i<Np;i++){
```

1.6 Center of mass correction (2)

```
15      dx=x[i]-x0[i];
16      if(dx>0.5*L) dx-=L;
17      else if(dx<-0.5*L)dx+=L;
18
19      dy=y[i]-y0[i];
20      if(dy>0.5*L) dy-=L;
21      else if(dy<-0.5*L)dy+=L;
22
23      *x_corr+=dx/Np; //center of mass displacement.x
24      *y_corr+=dy/Np;
25
26      x0[i]=x[i];
27      y0[i]=y[i];
28   }
29   return 0;
30 }
```
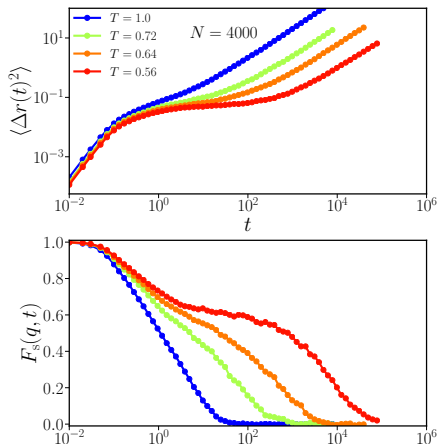
1.6 Center of mass correction (3)

FIG 3: The mean-square displacement, and the intermédiate scattering function at temperatures $T = 1.0, \ 0.72, \ 0.64, \ 0.56$ at $N = 4000$.