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Calculation of 4-point functions

Definitions

The 4-point correlation function:

$$G_4(\mathbf{r},t) \overset{\text{def}}{=} \langle \rho(\mathbf{r_0},t_0)\rho(\mathbf{r_0},t_0+t)\rho(\mathbf{r_0}+\mathbf{r},t_0)\rho(\mathbf{r_0}+\mathbf{r},t_0+t)\rangle \\ -\langle \rho(\mathbf{r_0},t_0)\rho(\mathbf{r_0},t_0+t)\rangle\langle \rho(\mathbf{r_0}+\mathbf{r},t_0)\rho(\mathbf{r_0}+\mathbf{r},t_0+t)\rangle$$

where <…> denotes ensemble average, and the density field

$$\rho(\mathbf{r},t) \overset{\text{def}}{=} \sum_{i=1}^{N} \delta(\mathbf{r}-\mathbf{r}_i)$$

The 4-point susceptibility:

$$\chi_4(t) \overset{\text{def}}{=} \int_V G_4 d\mathbf{r}$$

$$= \int_V \langle \rho(\mathbf{r_0},t_0)\rho(\mathbf{r_0},t_0+t)\rho(\mathbf{r_0}+\mathbf{r},t_0)\rho(\mathbf{r_0}+\mathbf{r},t_0+t)\rangle d\mathbf{r}$$

$$+ \int_V \langle \rho(\mathbf{r_0},t_0)\rho(\mathbf{r_0},t_0+t)\rangle d\mathbf{r} \int_V \langle \rho(\mathbf{r_0}+\mathbf{r},t_0)\rho(\mathbf{r_0}+\mathbf{r},t_0+t)\rangle d\mathbf{r}$$

$$= \iint_V \rho(\mathbf{r_1},t_0)\rho(\mathbf{r_1},t_0+t)\rho(\mathbf{r_2},t_0)\rho(\mathbf{r_2},t_0+t)d\mathbf{r_1}d\mathbf{r_2}$$

$$+ \int_V \langle \rho(\mathbf{r_1},t_0)\rho(\mathbf{r_1},t_0+t)\rangle d\mathbf{r_1} \int_V \langle \rho(\mathbf{r_2},t_0)\rho(\mathbf{r_2},t_0+t)\rangle d\mathbf{r_2}$$

$$= \left\langle \iint_V \rho(\mathbf{r_1},t_0)\rho(\mathbf{r_1},t_0+t)\rho(\mathbf{r_2},t_0)\rho(\mathbf{r_2},t_0+t)d\mathbf{r_1}d\mathbf{r_2} \right\rangle + \left[\left\langle \int_V \rho(\mathbf{r},t_0)\rho(\mathbf{r},t_0+t)d\mathbf{r} \right\rangle\right]^2$$

Calculation of $\chi_4(t)$

- Calculation of the integral $\iint_V \rho(\mathbf{r_1}, t_0)\rho(\mathbf{r_1}, t_0 + t)\rho(\mathbf{r_2}, t_0)\rho(\mathbf{r_2}, t_0 + t)d\mathbf{r_1}d\mathbf{r_2}$ by the definition of density field $\rho(\mathbf{r}, t) \stackrel{\text{def}}{=} \sum_{i=1}^{N} \delta(\mathbf{r} - \mathbf{r}_i)$
  - For a set of N particles in volume V with positions $\mathbf{r}_i$, $i = 1, 2, \ldots, N$, by definition, $\rho(\mathbf{r}, t) = 1$ for $\mathbf{r} = \mathbf{r}_i$ and 0 otherwise. So $\int_V \rho(\mathbf{r}, t)d\mathbf{r}$ is equivalent to counting all particles, *i.e.* $\int_V \rho(r, t)dr = \left\langle \int_V \rho(r, t)dr \right\rangle = N$ for canonical ensemble, and the volume average of the density field $V^{-1} \int_V \rho(\mathbf{r}, t)d\mathbf{r} = \frac{N}{V} \stackrel{\text{def}}{=} \rho$, the bulk density.
  - The problem of the integral is equivalent to counting all instances of $\rho(\mathbf{r_1}, t_0) = \rho(\mathbf{r_1}, t_0 + t) = \rho(\mathbf{r_2}, t_0) = \rho(\mathbf{r_2}, t_0 + t) = 1$ in the volume V.
  - Starting from the raw data of particle tracking, we have all positions $\mathbf{r}_m(t_0)$ and $\mathbf{r}_n(t_0 + t)$ of particles at t_0 and $t_0 + t$, respectively, so that $\rho[\mathbf{r}_m, t_0] = \rho[\boldsymbol{r}_n, t_0 + t] = 1$. Note that assuming canonical ensemble, the set of particles of t_0 and $t_0 + t$ should be the same, so $m = n = N$.
  - Find all distinct pairs from the N positions $\mathbf{r}_N(t_0)$ (C_N^2) into $[\mathbf{r}_i(t_0), \mathbf{r}_j(t_0)]$. Do the same thing to $\mathbf{r}_N(t_0 + t)$ into $[\mathbf{r}_k(t_0 + t), \mathbf{r}_l(t_0 + t)]$, so that $\rho(\mathbf{r}_i, t_0) = \rho(\mathbf{r}_j, t_0) = 1$, $\rho(\mathbf{r}_k, t_0 + t) = \rho(\mathbf{r}_l, t_0 + t) = 1$. Note that from the last point, we already have $\rho(\mathbf{r}_{i,j}, t_0)$=$\rho(\mathbf{r}_{k,l}, t_0 + t) = 1$.

Calculation of $\chi_4(t)$

- Calculation of the integral $\iint_V \rho(\mathbf{r_1},t_0)\rho(\mathbf{r_1},t_0+t)\rho(\mathbf{r_2},t_0)\rho(\mathbf{r_2},t_0+t)d\mathbf{r_1}d\mathbf{r_2}$ by the definition of density field $\rho(\mathbf{r},t) \stackrel{\text{def}}{=} \sum_{i=1}^{N} \delta(\mathbf{r}-\mathbf{r}_i)$
  - Now to calculate the above integral we only need to find the overlapping pairs from $\mathbf{r}_k(t_0 + t)$ and $\mathbf{r}_i(t)$, and also the overlapping pairs from $\mathbf{r}_{l=k}(t_0 + t)$ and $\mathbf{r}_{j=i}(t)$. The condition of "overlapping" is described by a Boolean function $w(|\mathbf{r}_1\text{-}\mathbf{r}_2|)$ so that $w = 1$ for $\mathbf{r}_1 = \mathbf{r}_2$ and $w = 0$ for $\mathbf{r}_1 \neq \mathbf{r}_2$. In practice, however, no two positions of particles at two space-times can be mathematically identical. It is only possible to defined an overlap function $q\,(|\mathbf{r}_1\text{-}\mathbf{r}_2|)$ that approaches 1 for $\mathbf{r}_1$ close enough to $\mathbf{r}_2$, and 0 for $\mathbf{r}_1$ far enough from $\mathbf{r}_2$, gauged by a probe length a, *e.g.*

    $$q_a(|\mathbf{r}_1-\mathbf{r}_2|) \stackrel{\text{def}}{=} \exp\left[-\frac{|\mathbf{r}_1-\mathbf{r}_2|^2}{2a^2}\right]$$

    So counting the instance of overlapping is equivalent to summing the overlapping function of all distinct pairs of positions $(\mathbf{r}_1, \mathbf{r}_2)$, *i.e.*

    $$N_{r_1=r_2} = \sum_{i}^{N}\sum_{j>i}^{N} q_a\left(\left|\mathbf{r}_i-\mathbf{r}_j\right|\right)$$

  - The integral is equal to

    $$\sum_{i}^{N}\sum_{k}^{N} q_a(|\mathbf{r}_i(t_0)-\mathbf{r}_k(t_0+t)|)\sum_{j=i}^{N}\sum_{l=k}^{N} q_a\left(\left|\mathbf{r}_j(t_0)-\mathbf{r}_l(t_0+t)\right|\right) = Q(a,t_0,t)^2$$

Calculation of $\chi_4(t)$

- Calculation of the integral $\iint_V \rho(\mathbf{r_1},t_0)\rho(\mathbf{r_1},t_0+t)\rho(\mathbf{r_2},t_0)\rho(\mathbf{r_2},t_0+t)d\mathbf{r_1}d\mathbf{r_2}$ by the definition of density field $\rho(\mathbf{r},t) \stackrel{\text{def}}{=} \sum_{i=1}^{N}\delta(\mathbf{r}-\mathbf{r}_i)$:

$$\iint_V \rho(\mathbf{r_1},t_0)\rho(\mathbf{r_1},t_0+t)\rho(\mathbf{r_2},t_0)\rho(\mathbf{r_2},t_0+t)d\mathbf{r_1}d\mathbf{r_2} = Q(a,t_0,t)^2$$

- Calculation of $\chi_4(t)$

$$\chi_4(t) \stackrel{\text{def}}{=} \left\langle \iint_V \rho(\mathbf{r_1},t_0)\rho(\mathbf{r_1},t_0+t)\rho(\mathbf{r_2},t_0)\rho(\mathbf{r_2},t_0+t)d\mathbf{r_1}d\mathbf{r_2} \right\rangle + \left[\left\langle \int_V \rho(\mathbf{r},t_0)\rho(\mathbf{r},t_0+t)d\mathbf{r} \right\rangle\right]^2 = \langle Q(a,t_0,t)^2\rangle - \langle Q(a,t_0,t)\rangle^2$$

The function $Q(a, t)$ and $F(\mathbf{k}, t)$

- The intermediate scattering function $F(\mathbf{k}, t)$ is

$$F(k,t) \stackrel{\text{def}}{=} \frac{1}{N}\langle \rho_{\mathbf{k}}(t_0)\rho_{-\mathbf{k}}(t_0+t)\rangle$$
$$= \frac{1}{N}\sum_i^N \sum_j^N \left\langle \exp\left[\mathbb{i}\mathbf{k}\cdot\left(\mathbf{r}_j(t_0+t)-\mathbf{r}_i(t_0)\right)\right]\right\rangle$$

- The overlap function $Q(a, t)$ is

$$Q(a,t) \stackrel{\text{def}}{=} \sum_i^N \sum_j^N \left\langle \exp\left[-\frac{\left|\mathbf{r}_j(t_0+t)-\mathbf{r}_i(t_0)\right|^2}{2a^2}\right]\right\rangle$$

- So $Q(a, t)$ is similar to the inverse Fourier transform of $F(\mathbf{k}, t)$ with $a \sim |\mathbf{k}|^{-1}$, having the same property that Q or $F \rightarrow 1$ when $\mathbf{r}_j(t_0+t)-\mathbf{r}_i(t_0) \sim a \sim |\mathbf{k}|^{-1}$ and 0 otherwise. For this reason, $\chi_4(t)$ is also defined as

$$\chi_4 \stackrel{\text{def}}{=} \langle F(\mathbf{k},t)^2\rangle - \langle F(\mathbf{k},t)\rangle^2$$

The self- and distinct-part of $\chi_4(t)$

- The summation in $Q(a, t_0, t) = \sum_i^N \sum_k^N q_a(|\mathbf{r}_i(t_0) - \mathbf{r}_k(t_0 + t)|)$ does not require the non-/equality between *i* and *k*. Requiring $i = k$ leads to the self-part of $\chi_4(t)$, in which case,

$$Q^{\mathrm{S}}(a, t_0, t) = \sum_i^N q_a(|\mathbf{r}_i(t_0) - \mathbf{r}_i(t_0 + t)|)$$
$$= \sum_i^N q_a\big(\Delta\mathbf{r}_i(t_0, t)\big)$$

  and $\chi_4^{\mathrm{S}} \stackrel{\mathrm{def}}{=} \left\langle {Q^{\mathrm{S}}}^2 \right\rangle - \langle Q^{s} \rangle^2$. In most literatures only the self-part of $\chi_4(t)$ was calculated.

- The distinct-part is simply calculated by requiring $k \neq i$.

The characteristic probe length $a^*(\tau)$

- $a^*(\tau)$ simply echoes the MSD(τ). Only the value of $\chi_4(a^*, \tau^*)$ reflects the correlation strength

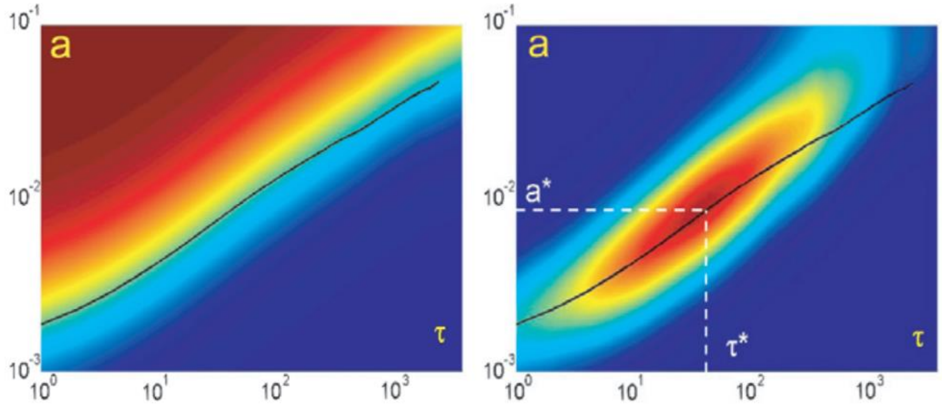

Fig. 6: Left: $Q_a(\tau)$ and right: $\chi_{4,a}(\tau)$ as a function of probing length a and delay time τ for $\phi = 0.8413 < \phi_J$ in a log-log color plot. The bold line represents $0.613\sigma_\phi(\tau)$.

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Calculation of $G_4(\mathbf{r}, t)$

- Calculation of the average $\langle \rho(\mathbf{r_0}, t_0)\rho(\mathbf{r_0}, t_0 + t)\rho(\mathbf{r_0} + \mathbf{r}, t_0)\rho(\mathbf{r_0} + \mathbf{r}, t_0 + t)\rangle$
  - The average is written as
$$\left\langle \int_V \rho(\mathbf{r_0}, t_0)\rho(\mathbf{r_0}, t_0 + t)\rho(\mathbf{r_0} + \mathbf{r}, t_0)\rho(\mathbf{r_0} + \mathbf{r}, t_0 + t) d\mathbf{r_0} \right\rangle$$
  - Starting from the raw data of particle tracking, we have all positions $\mathbf{r}_m(t_0)$ and $\mathbf{r}_n(t_0 + t)$ of particles at t_0 and $t_0 + t$, respectively, so that $\rho[\mathbf{r}_m, t_0] = \rho[\boldsymbol{r}_n, t_0 + t] = 1$. Note that assuming canonical ensemble, the set of particles of t_0 and $t_0 + t$ should be the same, so $m = n = N$.
  - Find all distinct pairs from the N positions $\mathbf{r}_N(t_0)$ (C_N^2) into $[\mathbf{r}_i(t_0), \mathbf{r}_j(t_0)]$. Do the same thing to $\mathbf{r}_N(t_0 + t)$ into $[\mathbf{r}_k(t_0 + t), \mathbf{r}_l(t_0 + t)]$, so that $\rho(\mathbf{r}_i, t_0) = \rho\left(\mathbf{r}_j, t_0\right) = 1$, $\rho(\mathbf{r}_k, t_0 + t) = \rho(\mathbf{r}_l, t_0 + t) = 1$. Note that from the last point, we already have $\rho\left(\mathbf{r}_{i,j}, t_0\right)=\rho\left(\mathbf{r}_{k,l}, t_0 + t\right) = 1$.
  - Select distinct pairs from two sequence $A_m = [\mathbf{r}_i(t_0), \mathbf{r}_j(t_0)]$ and $B_m = [\mathbf{r}_k(t_0 + t), \mathbf{r}_l(t_0 + t)]$ (pairs of pairs), where $m = C_N^2$. Construct a histogram of n bins for $\mathbf{r} = \mathbf{r}_k - \mathbf{r}_i$ for all pairs of pairs $[A, B]$ (there are C_m^2 pairs of pairs). The bins of $\mathbf{r}$ are denoted $\mathbf{r}_b$, $b = 1, 2, \ldots, n$.
  - For the N_b pairs of $[A, B]$ belonging to a bin $\mathbf{r}_b$, calculate the 4-point averaged overlap
$$\left\langle \sum_{i}^{N_b} \sum_{k}^{N_b} q_a(|\mathbf{r}_i(t_0) - \mathbf{r}_k(t_0 + t)|) \sum_{j=i}^{N_b} \sum_{l=k}^{N_b} q_a\left(\left|\mathbf{r}_j(t_0) - \mathbf{r}_l(t_0 + t)\right|\right) \right\rangle = \langle Q(a; \mathbf{r}_b, t_0, t)^2 \rangle$$
  which is the value of the initial average for $\mathbf{r} = \mathbf{r}_b$.

Calculation of $G_4(\mathbf{r}, t)$

- Calculation of the averages $\langle \rho(\mathbf{r_0}, t_0)\rho(\mathbf{r_0}, t_0 + t)\rangle$ and $\langle \rho(\mathbf{r_0} + \mathbf{r}, t_0)\rho(\mathbf{r_0} + \mathbf{r}, t_0 + t)\rangle$
  - The two averages are equal.

$$\langle \rho(\mathbf{r_0}, t_0)\rho(\mathbf{r_0}, t_0 + t)\rangle = \left\langle \int_V d\mathbf{r_0}\rho(\mathbf{r_0}, t_0)\rho(\mathbf{r_0}, t_0 + t) \right\rangle = \langle Q^s \rangle$$

- Calculation of $G_4(\mathbf{r}, t)$
  - $\mathbf{r}$ is discretized into $\mathbf{r}_b$, and G_4 is probe length dependent:

$$G_4(a; \mathbf{r}_b, t) = \langle Q(a; \mathbf{r}_b, t_0 , t)^2 \rangle - \langle Q^s \rangle^2$$

  - For isotropic systems, $\mathbf{r}_b$ can be replaced by $r_b = |\mathbf{r}_b|$.

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