Supplementary Information for

Transport Coefficient Approach for Characterizing Non-equilibrium Dynamics in Soft Matter

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Supporting Information Text

1. Abbreviation

- **Mathematical Operation:**
  - $\mathcal{F}$: Fourier Transform
  - $\mathbb{E}$: Ensemble Average
  - $\mathbb{V}$: Variance of the quantity as $\mathbb{V}[x] = \mathbb{E}\left[x^2\right] - \mathbb{E}[x]^2$
  - Cov: Covariance of two quantities as $\mathrm{Cov}[x, y] = \mathbb{E}[xy] - \mathbb{E}[x]\,\mathbb{E}[y]$
  - $(f * g)[t]$: Convolution of function $f(t)$ and $g(t)$, equivalent to $\int_{-\infty}^{\infty} f(\tau)g(t-\tau)\,\mathrm{d}\tau$
  - $(f \star g)(\tau)$: Correlation between function $f(t)$ and $g(t)$, equivalent to $\int_{-\infty}^{\infty} f(t)g(t+\tau)dt$.
  - $\vec{u} \otimes \vec{v}$: Outer product of vector $\vec{u}$ and $\vec{v}$
  - **Re**: taking the real part from the complex composition given as $\mathbf{Re}\left[\mathrm{e}^{iA}\right] = \mathbf{Re}\left[i\sin(A) + \cos(A)\right] = \cos(A)$
  - $\underline{\mathrm{A}}^{\mathrm{T}}$: Transpose of $\underline{\mathrm{A}}$ tensor.
- **Scattering:**
  - $E(\vec{q}, t)$: Scattered electromagnetic wave profile at time t
  - $I(\vec{q}, t)$: Scattered intensity profile at time t
  - b: Scattering contrast of individual particles in the system.
  - $c(\vec{r}, t_1, t_2)$: Correlation function in real space between t_1 and t_2
  - $c_1(\vec{q}, t_1, t_2)$: The first-order two-time correlation function in the reciprocal space between t_1 and t_2.
  - $c_2(\vec{q}, t_1, t_2)$: The second-order two-time correlation function in the reciprocal space between t_1 and t_2.
  - $g_1(\vec{q}, \tau)$: The first-order one-time correlation function in the reciprocal space
  - $g_2(\vec{q}, \tau)$: The second-order one-time correlation function in the reciprocal space
  - β: Speckle contrast of the incident beam.
  - $x_n(t)$: The fraction of the individual component n in the system at time t. Follow with the relation $1 = \sum_{n=1}^{N} x_n(t)$, with N being the total number of components.
  - ϕ_n: The angle between flow velocity and scattering vector $\vec{q}$ for n.
  - f: the normalization factor in a system with multiple compositions.
- **Probability:**
  - $\Delta\vec{r}(t_1, t_2)$: Displacement of a particle between t_1 and t_2.
  - $\zeta(t)$: The state variable that varies in the random process.
  - $\mathbb{P}(x, t|x_0, v_o)$: The probability density function for position of a particle (x) at time t given the initial conditions $\mathbb{P}(x, v, t = 0) = \delta(x - x_0)\delta(v - v_o)$.
  - $\mathbb{P}(\Delta x, t_1, t_2)$: Transition probability function of a particle making a displacement of Δx as time evolves from t_1 to t_2.
  - $\mathbb{P}(x_2, t_2|x_1, t_1)$: Joint probability function of $\mathbb{P}(x_1, t_1|x_0, v_o)$ and $\mathbb{P}(x_2, t_2|x_0, v_o)$.
  - J: Transport coefficient defined at Equation S-38.
  - $\mathcal{N}(x; \mu, \sigma^2)$: A normalized Gaussian function with respect to x with a mean of μ and a variance of σ^2.
- **Langevin Equation:**
  - γ: The friction coefficient or drift that characterizes the effect of friction on the particle's motion.
  - η: A 'truly random' force or noise independent of the particle's state of motion.
  - Γ: The magnitude of the random force depending on collisions between the particles and the solvent, given as $\Gamma = 2m\gamma k_B T$ for one-dimensional system.
  - ω_o: Force constant for the Brownian oscillator, given as $V(x) = \frac{1}{2}m{\omega_o}^2x^2$.
  - ω_s: The reduced frequency given as $\omega_s = \left({\omega_o}^2 - \frac{1}{4}\gamma^2\right)^{\frac{1}{2}}$.

- γ_s: The reduced drift is given as $\gamma_s = \left(\gamma^2 - 4{\omega_o}^2\right)^{\frac{1}{2}}$.
- D: The classical diffusion constant in the equilibrium state is defined as $D = \frac{k_B T}{m\gamma}$.
- D_o: The intrinsic diffusion constant of the system in the equilibrium state.

- **Rheology:**
  - γ: Strain in response to the applied deformation.
  - $\dot{\gamma}$: Shear rate in a laminar flow system.
  - σ: Stress applied to the system.
  - $\mathcal{J}$: Creep compliance, defined as γ/σ.

- **Acronym:**
  - XPCS: X-ray Photon Correlation Spectroscopy
  - Rheo-XPCS: combined XPCS and *in situ* Rheology
  - MD: Molecular Dynamics

2. Derivation of the Non-equilibrium model

A. Correlation Function in X-ray Photon Correlation Spectroscopy (XPCS). To understand the physical picture of XPCS, we first start with a system composed of N scattered points. The spatial distribution at time t can be described by the function denoted as:

$$\sum_{n=1}^{N} \delta(\vec{r} - \vec{r_n}(t)) \qquad \text{[S-1]}$$

In real space, this position function quantifies the relation between the positions of points at different times.

The profile of its scattered electromagnetic intensity is derived through a Fourier transform ($\mathcal{F}$), expressed as:

$$\begin{aligned} E(\vec{q},t) &= \mathcal{F}\left[\sum_{n=1}^{N} b_n(t)\delta(\vec{r} - \vec{r_n}(t))\right] = \sum_{n=1}^{N} b_n(t) \mathrm{e}^{i\vec{q}\cdot\vec{r}_n(t)} \\ I(\vec{q},t) &= |E(\vec{q},t)|^2 = E(\vec{q},t)E^*(\vec{q},t) \\ &= \sum_{n=1}^{N}\sum_{m=1}^{N} b_n(t)b_m(t)\mathrm{e}^{i\vec{q}\cdot(\vec{r}_n(t)-\vec{r}_m(t))} \end{aligned} \qquad \text{[S-2]}$$

where $b_n(t)$ in the equation S-2 is defined as the unit scattering contrast and $\vec{q}$ is the scattering vector in reciprocal space.

The correlation function $c(\vec{r}, t_1, t_2)$ between t_1 and t_2 in real space could be calculated by following:

$$c(\vec{r}, t_1, t_2) = \mathbb{E}\left[\sum_{n=1}^{N}\sum_{m=1}^{N} \delta\left(\vec{r} - (\vec{r}_n(t_2) - \vec{r}_m(t_1))\right)\right] \qquad \text{[S-3]}$$

where $\vec{r}_n(t_2) - \vec{r}_m(t_1)$ is the spatial correlation of any particle n and m between t_1 and t_2 and $\mathbb{E}$ denotes ensemble average .

During XPCS measurements, the first order two-time intensity autocorrelation function (c_1) is the temporal correlation of electromagnetic fields as the average of equation S-3 in the reciprocal space. To match with the physical picture, a Fourier transform ($\mathcal{F}$) is applied to the correlation function in real space S-3.

$$\mathbb{E}\left[E(\vec{q},t_2)E^*(\vec{q},t_1)\right] = \mathcal{F}\left[\mathbb{E}\left[c(\vec{r},t_1,t_2)\right]\right] = \mathbb{E}\left[\sum_{n=1}^{N}\sum_{m=1}^{N} b_n(t_2)b_m(t_1)\mathrm{e}^{i\vec{q}\cdot(\vec{r}_n(t_2)-\vec{r}_m(t_1))}\right] \qquad \text{[S-4]}$$

For the random dynamical process in the ensemble, each particle is assumed independent. In equation S-4, the terms where $n \neq m$ average out to zero (1, at eq. (1.6)). For the $n = m$ terms, the average intensity and correlation of the electromagnetic field can be expressed as:

$$\begin{aligned} \mathbb{E}\left[I(\vec{q},t)\right] &= \mathbb{E}\left[\sum_{n=1}^{N} b_n(t)^2 \mathrm{e}^{i\vec{q}\cdot(\vec{r}_n(t)-\vec{r}_n(t))}\right] = \mathbb{E}\left[\sum_{n=1}^{N} b_n(t)^2 \mathrm{e}^{i\vec{q}\cdot\vec{0}}\right] = \mathbb{E}\left[\sum_{n=1}^{N} b_n(t)^2\right] \\ \mathbb{E}\left[E(\vec{q},t_2)E^*(\vec{q},t_1)\right] &= \mathbb{E}\left[\sum_{n=1}^{N} b_n(t_1)b_n(t_2)\mathrm{e}^{i\vec{q}\cdot(\vec{r}_n(t_2)-\vec{r}_n(t_1))}\right] = \mathbb{E}\left[\sum_{n=1}^{N} b_n(t_1)b_n(t_2)\mathrm{e}^{i\vec{q}\cdot\Delta\vec{r}(t_1,t_2)}\right] \end{aligned} \qquad \text{[S-5]}$$

where $\Delta\vec{r}(t_1,t_2)$ is defined as the displacement of one particle between t_1 and t_2, given by $\Delta\vec{r}(t_1,t_2)=\vec{r}_n(t_2)-\vec{r}_n(t_1)$.

In the case where the system is composed of monodisperse spheres, with all particles having the same contrast and lacking any time and direction dependence as $b_n(t)=b_n$, this common factor can be extracted from the average.

$$\begin{aligned}\mathbb{E}\left[I(\vec{q},t)\right]&=\mathbb{E}\left[\sum_{n=1}^{N}b_n^2\right]=Nb_n^2\\ \mathbb{E}\left[E(\vec{q},t_2)E^*(\vec{q},t_1)\right]&=\mathbb{E}\left[\sum_{n=1}^{N}b_n^2\mathrm{e}^{i\vec{q}\cdot(\vec{r}_n(t_2)-\vec{r}_n(t_1))}\right]=Nb_n^2\mathbb{E}\left[\mathrm{e}^{i\vec{q}\cdot\Delta\vec{r}(t_1,t_2)}\right]\end{aligned} \tag{S-6}$$

Consequently, $c_1(\vec{q},t_1,t_2)$ is obtained by normalizing the correlation function by $|b_n|^2$. (2, at eq. (3.4.14))

$$\begin{aligned}c_1(\vec{q},t_1,t_2)&=\frac{\mathbb{E}\left[E(\vec{q},t_2)E^*(\vec{q},t_1)\right]}{\left[\mathbb{E}\left[|E(\vec{q},t_1)|^2\right]\mathbb{E}\left[|E(\vec{q},t_2)|^2\right]\right]^{\frac{1}{2}}}=\frac{\mathbb{E}\left[E(\vec{q},t_2)E^*(\vec{q},t_1)\right]}{\left[\mathbb{E}\left[I(\vec{q},t_1)\right]\mathbb{E}\left[I(\vec{q},t_2)\right]\right]^{\frac{1}{2}}}\\ &=\frac{Nb_n^2\mathbb{E}\left[\mathrm{e}^{i\vec{q}\cdot\Delta\vec{r}(t_1,t_2)}\right]}{Nb_n^2}=\mathbb{E}\left[\mathrm{e}^{i\vec{q}\cdot\Delta\vec{r}(t_1,t_2)}\right]\end{aligned} \tag{S-7}$$

To establish a correlation between $c_1(\vec{q},t_1,t_2)$ and the dynamics of particles, we consider a system where the particle distribution at any time t after $t_0=0$ follows a probability density function described by $\mathbb{P}(\vec{r},t|t_0=0)$. We then define $\mathbb{P}(\Delta\vec{r},t_1,t_2)$ as the transition probability of a particle with displacement $(\Delta\vec{r}=\vec{r}_2-\vec{r}_1)$ between time t_1 and time t_2. Then the relation between the probability distribution function at t_1 and t_2 can be connected with the transition probability as follows,

$$\begin{aligned}\mathbb{P}(\vec{r}_2,t_2|t_0=0)&=\int_{-\infty}^{\infty}\mathbb{P}(\vec{r}_1,t_1|t_0=0)\mathbb{P}(\Delta\vec{r},t_1,t_2)\,\mathrm{d}\vec{r}_1\\ &=\int_{-\infty}^{\infty}\mathbb{P}(\vec{r}_1,t_1|t_0=0)\mathbb{P}(\vec{r}_2,t_2|\vec{r}_1,t_1)\,\mathrm{d}\vec{r}_1\end{aligned} \tag{S-8}$$

In a Markov chain, where its behavior depends only on its current state but not on its past, the variable t_1 is excluded from the computation process in Equation (S-8). If $\mathbb{P}(\Delta r,t_1,t_2)$ is known, the c_1 in equation S-7 can be expressed equivalently as follows:

$$c_1(\vec{q},t_1,t_2)=\mathbb{E}\left[\mathrm{e}^{i\vec{q}\cdot\Delta\vec{r}}\right]=\int_{-\infty}^{\infty}\mathbb{P}(\Delta r,t_1,t_2)\mathrm{e}^{i\vec{q}\cdot\Delta\vec{r}}\,\mathrm{d}\Delta\vec{r} \tag{S-9}$$

By converting the $c_1(\vec{q},t_1,t_2)$ to the second order correlation function $c_2(\vec{q},t_1,t_2)$ with Siegert relation, we can obtain the experiment measurement of the XPCS.

$$c_2(\vec{q},t_1,t_2)=1+\beta|c_1(\vec{q},t_1,t_2)|^2=\frac{\mathbb{E}\left[I(\vec{q},t_1)I(\vec{q},t_2)\right]}{\mathbb{E}\left[I(\vec{q},t_1)\right]\mathbb{E}\left[I(\vec{q},t_2)\right]} \tag{S-10}$$

where β is the speckle contrast range from 0-1 depending on the coherence of the incident beam and $I(\vec{q},t)$ is the time-resolved intensity profile at specific $\vec{q}$ and time t.

B. Generalized Model: Markov Chain with Gaussian Random Walk. To gain a preliminary understanding of the dynamics of a complicated non-equilibrium system, it is assumed that the particles undergo a Gaussian random walk in 1-dimension. The step size for each walk varies according to a normal distribution. This dynamic behavior is described as a Markov process, where the evolution at a certain time t for $t>t_0$ does not depend on any of the previous states the system had been in before t. Instead, it relies only on the state of the system at time t. This process represents the simplest form of a system without memory, where the present distribution defines the future distribution.

In this Markov process, the n-time probability distribution for the process, denoted as $\mathbb{P}(\zeta_n,t_n)$, is defined as (3, at eq. (5.3)).

$$\begin{aligned}&n\text{-component general process}:\zeta(t)=(\zeta_0(t),\ldots,\zeta_n(t))\\ &\mathbb{P}(\zeta_n,t_n)=\mathbb{P}(\zeta_n,t_n|\zeta_{n-1},t_{n-1})\ldots\ldots\mathbb{P}(\zeta_1,t_1|\zeta_0,t_0)\mathbb{P}(\zeta_0,t_0)\\ &\mathbb{P}(\zeta_n,t_n)=\int_{-\infty}^{\infty}\cdots\int_{-\infty}^{\infty}\left[\prod_{t=t_0}^{t_n}\mathbb{P}(x_t,t|x_{t-1},t_{t-1})\right]\mathbb{P}(\zeta_0,t_0),\mathrm{d}x_{t_0}\ldots\mathrm{d}x_{t_n}\end{aligned} \tag{S-11}$$

where $\zeta(t)$ is the state variable that varies in the random process and $\mathbb{P}(\zeta_n,t_n|\zeta_{n-1},t_{n-1})$ is defined as the transition probability of states from time t_{n-1} to t_n.

In the Gaussian random walk model in 1-dimension, the state variable $\zeta(t)$ represents the position (x) of the particle, which varies during the random process. The transition probability of states from time t_{n-1} to t_n, is denoted as

$$\begin{aligned}\mathbb{P}(x_n, t_n | x_{n-1}, t_{n-1}) &= \frac{1}{\sqrt{2\pi J(t_n)\Delta t}} \mathrm{e}^{-\frac{\Delta x^2}{2J(t_n)\Delta t}} = \frac{1}{\sqrt{2\pi J(t_n)\Delta t}} \mathrm{e}^{-\frac{(x_n - x_{n-1})^2}{2J(t_n)\Delta t}} \\ &\triangleq \mathcal{N}\left(x_n - x_{n-1}; \mu = 0, \sigma^2 = J(t_n)\Delta t\right) \\ &\triangleq \mathcal{N}\left(x_n; \mu = x_{n-1}, \sigma^2 = J(t_n)\Delta t\right) \\ &\triangleq \mathcal{N}\left(x_{n-1}; \mu = x_n, \sigma^2 = J(t_n)\Delta t\right)\end{aligned} \tag{S-12}$$

This transition probability is a key element in the Gaussian random walk model, where each step follows a Gaussian distribution and the state variable evolves based on these transitions. Δx is the step the particles take from t_{n-1} to t_n, defined as $\Delta x = x_n - x_{n-1}$, and Δt is the time interval between adjacent steps t_n and t_{n-1}, given by $\Delta t = t_n - t_{n-1}$. $J(t_n)\Delta t$ is the standard deviation of the step size. $J(t_n)$ is the transport coefficient corresponding to the transition step in $t_{n-1} \to t_n$. We will define transport coefficient $(J(t))$ physically and mathematically at equation S-38 later.

To obtain the correlation function $c_1(\vec{q}, t_1, t_2)$ for any two states at time t_2 and t_1 with an arbitrary number of steps between them, we need to calculate the transition probability $\mathbb{P}(x_2, t_2 | x_1, t_1)$. According to the Chapman-Kolmogorov Equation, $\mathbb{P}(x_2, t_2 | x_1, t_1)$ can be expressed as the product of the probabilities of propagating from t_1 to all intermediate values $t - t'$ for $t_1 < t' < t_2$, and subsequently to t_2, summed over all possible values of the intermediate state. The expression is given as:

$$\mathbb{P}(x_2, t_2 | x_1, t_1) = \int_{-\infty}^{\infty} \cdots \int_{-\infty}^{\infty} \left[\prod_{t=t_1}^{t_2 - \Delta t} \mathbb{P}(x_{t+\Delta t}, t + \Delta t | x_t, t) \right] \mathbb{P}(x_{t_1}, t_1 | x_1, t_1), \mathrm{d}x_{t_1} \ldots \mathrm{d}x_{t_2 - \Delta t} \tag{S-13}$$

where Δt is the time interval between consecutive states, and the product represents the joint probabilities of propagating from t to $t + \Delta t$ for each intermediate time interval. This integral sums up all possible intermediate states between t_1 and t_2, and allows us to calculate the transition probability from an initial state x_1 at time t_1 to a final state x_2 at t_2 considering all possible paths and intermediate states.

In the transition probability for any two nearby steps $\mathbb{P}(x_c, t_c | x_a, t_a)$ at $t_a \to t_b$ and $t_b \to t_c$, according to equation S-12 and S-13, is

$$\begin{aligned}\mathbb{P}(x_c, t_c | x_a, t_a) &= \int_{-\infty}^{\infty} \mathbb{P}(x_c, t_c | x_b, t_b) \mathbb{P}(x_b, t_b | x_a, t_a) \, \mathrm{d}x_b \\ &= \int_{-\infty}^{\infty} \mathcal{N}\left(x_c - x_b; \mu = 0, \sigma^2 = J(t_c)\Delta t\right) \mathcal{N}\left(x_b - x_a; \mu = 0, \sigma^2 = J(t_b)\Delta t\right) \, \mathrm{d}x_b \\ &= \int_{-\infty}^{\infty} \mathcal{N}\left(x_c - x_b; \mu = 0, \sigma^2 = J(t_c)\Delta t\right) \mathcal{N}\left(x_b; \mu = x_a, \sigma^2 = J(t_b)\Delta t\right) \, \mathrm{d}x_b\end{aligned} \tag{S-14}$$

The equation S-14 is a convolution operation of Gaussian function as $\tau = x_b$ with the property of resulting in a new Gaussian function with a mean of $\mu = \mu_f + \mu_g$ and variance of $\sigma^2 = \sigma_f^2 + \sigma_g^2$ as follow:

$$\begin{aligned}(f * g)[t] &= \int_{-\infty}^{\infty} f(\tau) g(t - \tau) \, \mathrm{d}\tau \\ (\mathcal{N}(\tau; \mu_f, \sigma_f^2) * \mathcal{N}(t - \tau; \mu_g, \sigma_g^2))[t] &= \mathcal{N}(t; \mu_f + \mu_g, \sigma_f^2 + \sigma_g^2)\end{aligned} \tag{S-15}$$

Substituting the relation at S-15 to equation S-14, $\mathbb{P}(x_c, t_c | x_a, t_a)$ is obtained as follow:

$$\begin{aligned}\mathbb{P}(x_c, t_c | x_a, t_a) &= \mathcal{N}\left(x_c - x_b; \mu = 0, \sigma^2 = J(t_c)\Delta t\right) * \mathcal{N}\left(x_b; \mu = x_a, \sigma^2 = J(t_b)\Delta t\right)[x_c] \\ &= \mathcal{N}\left(x_c; \mu = x_a, \sigma^2 = J(t_c)\Delta t + J(t_b)\Delta t\right) \\ &= \mathcal{N}\left(x_c - x_a; \mu = 0, \sigma^2 = (J(t_c) + J(t_b))\Delta t\right)\end{aligned} \tag{S-16}$$

Expanding the result from S-16 to S-13, we can obtain $\mathbb{P}(x_2, t_2 | x_1, t_1)$ as follow:

$$\mathbb{P}(x_2, t_2 | x_1, t_1) = \mathcal{N}\left(x_2 - x_1; \mu = 0, \sigma^2 = \Delta t \sum_{t=t_1}^{t_2} J(t)\right) \tag{S-17}$$

Adapting an assumption that step difference is infinitesimal that there is infinity t' between t_2 and t_1, the summation $\Delta t \sum_{t'=t_1}^{t_2} J(t')$ can be expressed as an integration of a continuous function $J(t)$ as:

$$\begin{aligned} \Delta t \sum_{t=t_1}^{t_2} J(t) &= \int_{t_1}^{t_2} J(t)\,\mathrm{d}t \\ \mathbb{P}(x_2,t_2|x_1,t_1) &= \mathcal{N}\left(x_2-x_1;\mu=0,\sigma^2=\int_{t_1}^{t_2} J(t)\,\mathrm{d}t\right) \end{aligned} \tag{S-18}$$

where $J(t)$ is a continuous function of the change in the distribution standard deviation in $\mathbb{P}(x,t)$ as time evolves.

Considering $x_2 - x_1 = \Delta x$, we in fact obtain the $\mathbb{P}(\Delta x, t_1, t_2)$. The $c_1(q,t_1,t_2)$ is calculated according to the equation S-4 as:

$$\begin{aligned} c_1(q,t_1,t_2) &= \int_{-\infty}^{\infty} \mathbb{P}(\Delta x,t_1,t_2)\mathrm{e}^{iq\Delta x}\,\mathrm{d}\Delta x \\ &= \int_{-\infty}^{\infty} \mathcal{N}\left(\Delta x;\mu=0,\sigma^2=\int_{t_1}^{t_2} J(t)\,\mathrm{d}t\right)\mathrm{e}^{iq\Delta x}\,\mathrm{d}\Delta x \\ &= \frac{1}{\sqrt{2\pi\int_{t_1}^{t_2} J(t)\,\mathrm{d}t}}\int_{-\infty}^{\infty} \mathrm{e}^{-\frac{\Delta x^2}{2\int_{t_1}^{t_2} J(t)\,\mathrm{d}t}+iq\Delta x}\,\mathrm{d}\Delta x \\ &= \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \end{aligned} \tag{S-19}$$

Considering the Siegert relation, the $c_2(q,t_1,t_2)$ obtained from XPCS measurement for the Gaussian random walk process is defined as:

$$c_2(q,t_1,t_2) = 1+\beta|c_1(q,t_1,t_2)|^2 = \beta\mathrm{e}^{-q^2\int_{t_1}^{t_2} J(t)\,\mathrm{d}t}+1 \tag{S-20}$$

This equation describes the $c_2(\vec{q},t_1,t_2)$ of a system constituted of particles undergoing random Gaussian walk.

C. Derivation of $c_1(q,t_1,t_2)$ from Langevin Equation.

Begin with Newton's equation of motion for particles in 1-dimension:

$$m\dot{v} = F(t) = F_{in}(t) + F_{ex}(t) \tag{S-21}$$

where $F(t)$ is the total force on the particles includes internal F_{in} and external F_{ex} force. When the internal force includes a random force $\eta(t)$ and a systematic force $F_{sys}(x,v,t)$ depends on the state of particle motion,

$$F_{in}(t) = F_{sys}(x,v) + \eta(t) \tag{S-22}$$

In a simple system, F_{sys} only contains a drift force $m\gamma v(t)$ act in the opposite direction to its motion to prevent large velocity fluctuations from building up. Therefore, the expression of F_{in} is

$$F_{in} = -m\gamma v(t) + \eta(t) \tag{S-23}$$

We then have the Langevin Equation as:

$$m\dot{v} = -m\gamma v(t) + \eta(t) + F_{ex}(t) \tag{S-24}$$

In this equation, v(t) represents the velocity of the particle, and m denotes its mass. The parameter γ is a positive constant referred to as the friction coefficient or drift, which characterizes the effect of friction on the particle's motion. The term $\eta(t)$ denotes a 'truly random' force or noise independent of the state of motion of the particle. It has a 1st moment of zero and 2nd moment proportional to thermal fluctuations with a factor of Γ, which, according to fluctuation-dissipation theorem, is $\Gamma = 2m\gamma k_B T$,

$$\begin{aligned} \mathbb{E}\left[\eta(t)\right] &= 0 \\ \mathbb{E}\left[\eta(t)\eta(t')\right] &= \Gamma\delta(t-t') \end{aligned} \tag{S-25}$$

Finally, F_{ex} represents the result of all external applied forces acting on individual particles, which we consider identical throughout the paper.

From solving the Langevin Equation, the velocity of individual particle $v(t)$, mean velocity $\mathbb{E}[v(t)]$ and mean position $\mathbb{E}[x(t)]$ of the ensemble can be expressed as: (3, at eq. (2.19, 3.3 and 11.14))

$$\begin{aligned} v(t) &= v_o \mathrm{e}^{-\gamma t} + \frac{1}{m}\int_0^t \mathrm{d}t' \mathrm{e}^{-\gamma(t-t')}\left[\eta(t') + F_{ex}(t')\right] \\ \mathbb{E}[v(t)] &= v_o \mathrm{e}^{-\gamma t} + \mathbb{E}\left[\frac{1}{m}\int_0^t \mathrm{d}t' \mathrm{e}^{-\gamma(t-t')}\left[\eta(t') + F_{ex}(t')\right]\right] \\ &= v_o \mathrm{e}^{-\gamma t} + \frac{1}{m}\int_0^t \mathrm{d}t' \mathrm{e}^{-\gamma(t-t')} F_{ex}(t') \\ \mathbb{E}[x(t)] &= x_0 + \frac{v_o}{\gamma}(1-\mathrm{e}^{-\gamma t}) + \frac{1}{m}\int_0^t \mathrm{d}t' \int_0^{t'} \mathrm{d}t'' \mathrm{e}^{-\gamma(t'-t'')} F_{ex}(t'') \end{aligned} \tag{S-26}$$

From the velocity $v(t)$, the temporal correlation of velocity $\mathbb{E}[v(t)v(t')]$, the mean square average of the position $\mathbb{E}\left[x(t)^2\right]$, and the variance of the position distribution $\mathbb{V}[x(t)]$ exhibit following relations: (3, at eq. (15.2)) (4, at eq. (2.186))

$$\begin{aligned} \mathbb{E}\left[x(t)^2\right] &= x_0^2 + \int_0^t \mathrm{d}t_1 \int_0^t \mathrm{d}t_2\, \mathbb{E}[v(t_1)v(t_2)] \\ \mathbb{V}[x(t)] &= \mathbb{E}\left[x(t)^2\right] - \mathbb{E}[x(t)]^2 \end{aligned} \tag{S-27}$$

For any stochastic process exhibiting the Markov Chain characteristic, and when all particles in the system have the same initial conditions of position $x(t=0) = x_0$ and velocity $v(t=0) = v_0$, as:

$$\mathbb{P}(x, v, t=0) = \delta(x - x_0)\delta(v - v_o) \tag{S-28}$$

the probability distribution function at any given time t can be represented by a Gaussian function. This Gaussian distribution depends on the known mean $\mathbb{E}[x(t)]$ and variance $\mathbb{V}[x(t)]$, for that specific moment.

$$\begin{aligned} \mathbb{P}(x, t|x_0, t=0) &= \int_{-\infty}^{\infty} \mathbb{P}(x, v, t|x_0, v_o, t=0)\,\mathrm{d}v \\ &= \frac{1}{\sqrt{2\pi\mathbb{V}[x(t)]}} \exp\left(-\frac{[x - \mathbb{E}[x(t)]]^2}{2\mathbb{V}[x(t)]}\right) \\ &= \mathcal{N}\left(x; \mu = \mathbb{E}[x(t)], \sigma^2 = \mathbb{V}[x(t)]\right) \end{aligned} \tag{S-29}$$

According to the Equation (S-18), $\mathbb{P}(x, t|x_0, v_o)$ at any time t can be rewritten as:

$$\begin{aligned} \mathbb{E}[x(t)] &= x_0 + \int_0^t \mathbb{E}[v(t)]\,\mathrm{d}t \\ \mathbb{V}[x(t)] &= \int_0^t J(t)\,\mathrm{d}t \\ \mathbb{P}(x, t|x_0, v_o) &= \mathcal{N}\left(x; \mu = x_0 + \int_0^t \mathbb{E}[v(t)]\,\mathrm{d}t, \sigma^2 = \int_0^t J(t)\,\mathrm{d}t\right) \end{aligned} \tag{S-30}$$

with the initial conditions according to Equation (S-28),

$$\begin{aligned} \mathbb{E}[x(0)] &= x_0 \\ \mathbb{V}[x(0)] &= 0 \end{aligned} \tag{S-31}$$

To obtain the transition probability $\mathbb{P}(\Delta x, t_2, t_1)$ between t_1 and t_2, we apply the Fourier transform to both sides of Equation (S-8),

$$\mathcal{F}[\mathbb{P}(x_2, t_2|x_0, t=0)] = \mathcal{F}[\mathbb{P}(x_1, t_1|x_0, t=0)]\,\mathcal{F}[\mathbb{P}(\Delta x, t_2, t_1)] \tag{S-32}$$

so

$$\mathbb{P}(\Delta x, t_2, t_1) = \mathcal{F}^{-1}\left[\frac{\mathcal{F}[\mathbb{P}(x_2, t_2|x_0, t=0)]}{\mathcal{F}[\mathbb{P}(x_1, t_1|x_0, t=0)]}\right] \tag{S-33}$$

Considering the special property of Gaussian-like function in Fourier transform,

$$\mathcal{F}\left[\mathcal{N}\left(x; \mu, \sigma^2\right)\right] = \mathrm{e}^{-\frac{\sigma^2}{2}q^2 - i\mu q} \tag{S-34}$$

$\mathbb{P}(\Delta x, t_2, t_1)$ is obtained as

$$\begin{aligned}\mathbb{P}(\Delta x, t_2, t_1) &= \mathcal{F}^{-1}\left[\frac{\exp\left(-\frac{\mathbb{V}[x(t_2)]}{2}q^2 - i\mathbb{E}\left[x(t_2)\right]q\right)}{\exp\left(-\frac{\mathbb{V}[x(t_1)]}{2}q^2 - i\mathbb{E}\left[x(t_1)\right]q\right)}\right] \\ &= \mathcal{F}^{-1}\left[\exp\left(-\frac{(\mathbb{V}[x(t_2)] - \mathbb{V}[x(t_1)])}{2}q^2 - i\left(\mathbb{E}\left[x(t_2)\right] - \mathbb{E}\left[x(t_1)\right]\right)q\right)\right] \\ &= \mathcal{N}\left(\Delta x; \mu = \mathbb{E}\left[x(t_2)\right] - \mathbb{E}\left[x(t_1)\right], \sigma^2 = \mathbb{V}[x(t_2)] - \mathbb{V}[x(t_1)]\right)\end{aligned} \tag{S-35}$$

The transition probability between two Gaussian-like probability functions preserves the Gaussian shape, and the resulting Gaussian function will have its mean shifted by the difference between the means of the original Gaussian functions ($\mathbb{E}\left[x(t_2)\right] - \mathbb{E}\left[x(t_1)\right]$) and its variance scaled by the difference between the variances of the original Gaussian functions ($\mathbb{V}[x(t_2)] - \mathbb{V}[x(t_1)]$) based on the specific values of the original Gaussian functions.

We then define the change in the mean position of a particle as time evolves from t_1 to t_2, $\mathbb{E}\left[x(t_2)\right] - \mathbb{E}\left[x(t_1)\right]$, as the integration of mean velocity from t_1 to t_2; the change in the variance of the position distribution $\mathbb{V}[x(t_2)] - \mathbb{V}[x(t_1)]$, as the integration of dynamical transport coefficient $J(t)$ from t_1 to t_2:

$$\mathbb{P}(\Delta x, t_1, t_2) = \mathcal{N}\left(\Delta x; \mu = \int_{t_1}^{t_2}\mathbb{E}\left[v(t)\right]\,\mathrm{d}t, \sigma^2 = \int_{t_1}^{t_2} J(t)\,\mathrm{d}t\right) \tag{S-36}$$

so that relation Equation (S-8) is clearly satisfied as follow:

$$\begin{aligned}\mathbb{P}(x_2, t_2|x_0, t=0) &= \int_{-\infty}^{\infty}\mathbb{P}(x_1, t_1|x_0, t=0)\mathbb{P}(\Delta x, t_1, t_2)\,\mathrm{d}x_1 \\ &= \left(\mathcal{N}\left(x_1; \mu = x_0 + \int_0^{t_1}\mathbb{E}\left[v(t)\right]\,\mathrm{d}t, \sigma^2 = \int_0^{t_1} J(t)\,\mathrm{d}t\right) * \mathcal{N}\left(\Delta x; \mu = \int_{t_1}^{t_2}\mathbb{E}\left[v(t)\right]\,\mathrm{d}t, \sigma^2 = \int_{t_1}^{t_2} J(t)\,\mathrm{d}t\right)\right)[x_1] \\ &= \mathcal{N}\left(x_2; \mu = x_0 + \int_0^{t_2}\mathbb{E}\left[v(t)\right]\,\mathrm{d}t, \sigma^2 = \int_0^{t_2} J(t)\,\mathrm{d}t\right)\end{aligned} \tag{S-37}$$

which is consistent with the definition in Equation (S-30).

In Equation (S-36), we define the change in the mean position of a particle as time evolves from t_1 to t_2, $\mathbb{E}\left[x(t_2)\right] - \mathbb{E}\left[x(t_1)\right]$, as the integration of mean velocity from t_1 to t_2; the change in variance of the position distribution $\mathbb{V}[x(t_2)] - \mathbb{V}[x(t_1)]$, as the integration of the dynamical transport coefficient $J(t)$ from t_1 to t_2:

The mathematical and physical definition of the transport coefficient $J(t)$ is shown as:

$$\begin{aligned}J(t) &= \frac{d}{dt}(\mathbb{V}[x(t)]) \\ &= 2\left(\mathbb{E}\left[x(t)v(t)\right] - \mathbb{E}\left[x(t)\right]\mathbb{E}\left[v(t)\right]\right) = 2\mathrm{Cov}\left[x(t), v(t)\right] \\ &= 2\left(\mathbb{E}\left[\int_0^t v(t')v(t)\,\mathrm{d}t'\right] - \mathbb{E}\left[\int_0^t v(t')\,\mathrm{d}t'\right]\mathbb{E}\left[v(t)\right]\right) \\ &= 2\left(\int_0^t \mathbb{E}\left[v(t')v(t)\right]\,\mathrm{d}t' - \int_0^t \mathbb{E}\left[v(t')\right]\mathbb{E}\left[v(t)\right]\,\mathrm{d}t'\right) \\ &= 2\int_0^t \mathrm{Cov}\left[v(t), v(t')\right]\,\mathrm{d}t'\end{aligned} \tag{S-38}$$

where $\mathrm{Cov}\left[x(t), v(t)\right]$ is the covariance of $x(t)$ and $v(t)$ and $\int_0^t \mathrm{Cov}\left[v(t), v(t')\right]\,\mathrm{d}t'$ is a generalized form of Green-Kubo formula (3, at eq. (15.3)).

From eq. (S-36), we have following expression:

$$\mathbb{P}(\Delta x, t_1, t_2) = \frac{1}{\sqrt{2\pi\int_{t_1}^{t_2} J(t)\,\mathrm{d}t}}\exp\left(-\frac{\left[\Delta x - \int_{t_1}^{t_2}\mathbb{E}\left[v(t)\right]\,\mathrm{d}t\right]^2}{2\int_{t_1}^{t_2} J(t)\,\mathrm{d}t}\right) \tag{S-39}$$

At the end, the two-time correlation function $c_1(q,t_1,t_2)$, at specific scattering vector q, can be expressed as:

$$\begin{aligned} c_1(q,t_1,t_2) &= \int_{-\infty}^{\infty} \mathbb{P}(\Delta x,t_1,t_2)\mathrm{e}^{iq\Delta x}\,\mathrm{d}\Delta x \\ &= \frac{1}{\sqrt{2\pi\int_{t_1}^{t_2}J(t)\,\mathrm{d}t}}\int_{-\infty}^{\infty}\exp\left(-\frac{\left[\Delta x-\int_{t_1}^{t_2}\mathbb{E}\left[v(t)\right]\,\mathrm{d}t\right]^2}{2\int_{t_1}^{t_2}J(t)\,\mathrm{d}t}+iq\Delta x\right)\mathrm{d}\Delta x \\ &= \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2}J(t)\,\mathrm{d}t}\,\mathrm{e}^{iq\int_{t_1}^{t_2}\mathbb{E}[v(t)]\,\mathrm{d}t} \end{aligned} \tag{S-40}$$

Compared to Equation (S-35), Equation (S-40) is equivalent to the Fourier transform of the transition probability. In other words, the correlation function measured in XPCS is the ratio between the Fourier transform of the probability function of a particle at t_1 and t_2.

$$c_1(q,t_1,t_2) = \mathcal{F}\left[\mathbb{P}(\Delta x,t_1,t_2)\right] = \frac{\mathcal{F}\left[\mathbb{P}(x_2,t_2|x_0,t=0)\right]}{\mathcal{F}\left[\mathbb{P}(x_1,t_1|x_0,t=0)\right]} \tag{S-41}$$

According to eq. (S-40), the $c_1(q,t_1,t_2)$ could be factorized into two terms:

$$c_1(q,t_1,t_2) = c_{1,in}(q,t_1,t_2)c_{1,ex}(q,t_1,t_2) \tag{S-42}$$

where $c_{1,in}(q,t_1,t_2)$ is the correlation function attributed to internal forces, including the systematic dynamics as well as the thermal fluctuations experienced by particles, defined as:

$$c_{1,in}(q,t_1,t_2) = \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2}J(t)\,\mathrm{d}t} \tag{S-43}$$

and $c_{1,ex}(q,t_1,t_2)$ is the correlation function attributed to external drives defined as:

$$c_{1,ex}(q,t_1,t_2) = \mathrm{e}^{iq\int_{t_1}^{t_2}\mathbb{E}[v(t)]\,\mathrm{d}t} \tag{S-44}$$

In summary, the $c_1(q,t_1,t_2)$ is obtained by following steps:

1. Formalize Langevin equation based force profile of individual particles
2. Calculate $\mathbb{E}\left[v(t)\right]$ and $J(t)$ from Langevin equation.
3. Obtain $c_1(q,t_1,t_2)$ by substituting $\mathbb{E}\left[v(t)\right]$ and $J(t)$ into equation S-40

Equation S-40 is the $c_1(q,t_1,t_2)$ for the model of particles with Langevin Dynamics in 1-dimension as a fluid transports with mean velocity ($\mathbb{E}\left[v(t)\right]$) due to the bulk movement and transport coefficient $J(t)$ due to molecular random motion.

D. $c_1(\vec{q},t_1,t_2)$ in 3-Dimensional Space. To extend the analysis to 3-dimension and derive $c_1(\vec{q},t_1,t_2)$, we start from Langevin equation in 3 dimensions: (3, at eq. (4.9))

$$m\vec{v} = -m\gamma\vec{v}+\eta(t)+\vec{F}_{ex} \tag{S-45}$$

where $\vec{v}$, $\vec{\eta}$ and $\vec{F}_{ex}$ are the vectors consist all Cartesian components. It is noticed that $\vec{\eta}(t)$ is an independent Gaussian white noise. The mean and autocorrelation of the noise are given by the following straightforward generalizations according to S-25:

$$\begin{aligned} \mathbb{E}\left[\vec{\eta}(t)\right] &= \vec{0} \\ \mathbb{E}\left[\vec{\eta}(t)\otimes\vec{\eta}(t')\right]_{ij} &= \Gamma_{ij}\delta(t-t') \end{aligned} \tag{S-46}$$

where the Cartesian indices i , j run over the values (x,y,z) and Γ_{ij} is the noise strength correlated to direction i , j.

In summary, for the case in the Ornstein–Uhlenbeck process where noises in each dimension are uncorrelated, the partial differential equation can be separated and solved independently as one in 1-dimension.

As the first step of solving $c_1(\vec{q},t_1,t_2)$ in 3D, the velocity of individual particle $\vec{v}(t)$, mean velocity $\mathbb{E}\left[\vec{v}(t)\right]$, and mean position $\mathbb{E}\left[\vec{r}(t)\right]$ of the ensemble are solved as:

$$\begin{aligned} \vec{v}(t) &= \vec{v}_o\mathrm{e}^{-\gamma t}+\frac{1}{m}\int_0^t \mathrm{d}t'\mathrm{e}^{-\gamma(t-t')}\left[\vec{\eta}(t')+\vec{F}_{ex}(t')\right] \\ \mathbb{E}\left[\vec{v}(t)\right] &= \vec{v}_o\mathrm{e}^{-\gamma t}+\frac{1}{m}\int_0^t \mathrm{d}t'\mathrm{e}^{-\gamma(t-t')}\vec{F}_{ex}(t') \\ \mathbb{E}\left[\vec{r}(t)\right] &= \vec{r}_o+\frac{\vec{v}_o}{\gamma}(1-\mathrm{e}^{-\gamma t})+\frac{1}{m}\int_0^t \mathrm{d}t'\int_0^{t'}\mathrm{d}t''\mathrm{e}^{-\gamma(t'-t'')}\vec{F}_{ex}(t'') \end{aligned} \tag{S-47}$$

In Ornstein–Uhlenbeck process without external forces, $F_{ex} = 0$, the mean of position at each axis, $\mathbb{E}\left[\vec{r}(t)\right]_i$, and velocity correlation function $\mathbb{E}\left[v_i(t)v_j(t')\right]$ can be written as:

$$\begin{aligned}
\mathbb{E}\left[\vec{r}(t)\right]_i &= r_{o,i} + \frac{v_{o,i}}{\gamma}(1 - \mathrm{e}^{-\gamma t}) \\
\mathbb{E}\left[\vec{v}(t) \otimes \vec{v}(t')\right]_{ij} &= v_{o,i}v_{o,j}\mathrm{e}^{-\gamma(t+t')} + \\
&\frac{1}{m^2}\int_0^t \mathrm{d}t_1 \int_0^{t'} \mathrm{d}t_2\, \mathrm{e}^{-\gamma(t-t_1)-\gamma(t'-t_2)}\mathbb{E}\left[\eta_i(t_1)\eta_j(t_2)\right]
\end{aligned} \tag{S-48}$$

The outer product of mean of velocity $\mathbb{E}\left[\vec{v}(t)\right] \otimes \mathbb{E}\left[\vec{v}(t')\right]$ can be written as:

$$\left[\mathbb{E}\left[\vec{v}(t)\right] \otimes \mathbb{E}\left[\vec{v}(t')\right]\right]_{ij} = v_{o,i}v_{o,j}\mathrm{e}^{-\gamma(t+t')} \tag{S-49}$$

The cross-covariance matrix of velocity at t and t' is calculated as:

$$\begin{aligned}
\mathrm{Cov}\left[\vec{v}(t), \vec{v}(t')\right]_{ij} &= \mathbb{E}\left[\vec{v}(t) \otimes \vec{v}(t')\right]_{ij} - \left[\mathbb{E}\left[\vec{v}(t)\right] \otimes \mathbb{E}\left[\vec{v}(t')\right]\right]_{ij} \\
&= \frac{1}{m^2}\int_0^t \mathrm{d}t_1 \int_0^{t'} \mathrm{d}t_2\, \mathrm{e}^{-\gamma(t-t_1)-\gamma(t'-t_2)}\mathbb{E}\left[\eta_i(t_1)\eta_j(t_2)\right] \\
&= \frac{\Gamma_{ij}}{m^2}\int_0^t \mathrm{d}t_1 \int_0^{t'} \mathrm{d}t_2\, \mathrm{e}^{-\gamma(t-t_1)-\gamma(t'-t_2)}\delta(t_1 - t_2) \\
&= \frac{\Gamma_{ij}}{2m^2\gamma}(\mathrm{e}^{-\gamma|t-t'|} - \mathrm{e}^{-\gamma(t+t')})
\end{aligned} \tag{S-50}$$

The element $J_{ij}(t)$ in transport coefficient tensor $\underline{J}$ is calculated as:

$$\begin{aligned}
J_{ij}(t) &= 2\int_0^t \mathrm{Cov}\left[\vec{v}(t), \vec{v}(t')\right]_{ij}\,\mathrm{d}t' \\
&= \frac{\Gamma_{ij}}{m^2\gamma}\int_0^t \mathrm{e}^{-\gamma|t-t'|} - \mathrm{e}^{-\gamma(t+t')}\,\mathrm{d}t' \\
&= \frac{\Gamma_{ij}}{m^2\gamma^2}(1 - 2\mathrm{e}^{-\gamma t} + \mathrm{e}^{-2\gamma t})
\end{aligned} \tag{S-51}$$

Then, the covariance matrix with elements of $\mathbb{V}\left[\vec{r}(t)\right]_{ij}$ can be calculated from integrating $\mathbb{V}\left[\vec{v}(t) \otimes \vec{v}(t')\right]_{ij}$:

$$\begin{aligned}
\mathbb{V}\left[\vec{r}(t)\right]_{ij} &= \int_0^t \mathrm{d}t_1 \int_0^t \mathrm{d}t_2\, \mathrm{Cov}\left[\vec{v}(t), \vec{v}(t')\right]_{ij} \\
&= \frac{\Gamma_{ij}}{2m^2\gamma^3}(2\gamma t - 3 + 4\mathrm{e}^{-\gamma t} - \mathrm{e}^{-2\gamma t}) \\
\underline{\mathbb{V}\left[\vec{r}(t)\right]} &= \frac{1}{2m^2\gamma^3}(2\gamma t - 3 + 4\mathrm{e}^{-\gamma t} - \mathrm{e}^{-2\gamma t})\left(\underline{\Gamma}_{i=j} + \underline{\Gamma}_{i\neq j}\right) \\
\underline{\Gamma}_{i=j} &= \begin{bmatrix} \Gamma_{xx} & 0 & 0 \\ 0 & \Gamma_{yy} & 0 \\ 0 & 0 & \Gamma_{zz} \end{bmatrix}, \underline{\Gamma}_{i\neq j} = \begin{bmatrix} 0 & \Gamma_{xy} & \Gamma_{xz} \\ \Gamma_{yx} & 0 & \Gamma_{yz} \\ \Gamma_{zx} & \Gamma_{zy} & 0 \end{bmatrix}
\end{aligned} \tag{S-52}$$

where $\left(\underline{\Gamma}_{i=j}\right)$ is the variances of the individual dimension and $\underline{\Gamma}_{i\neq j}$ is the covariances across different dimensions.

If we assume the noise strength Γ in different directions are truly randomized and uncorrelated, the autocorrelation of noise $\mathbb{E}\left[\vec{\eta}(t) \otimes \vec{\eta}(t')\right]_{ij}$ can be written as: (3, at eq. (4.11))

$$\mathbb{E}\left[\vec{\eta}(t) \otimes \vec{\eta}(t')\right]_{ij} = \Gamma_{ij}\delta_{ij}\delta(t - t') \tag{S-53}$$

δ_{ij} is the Kronecker delta.

With the uncorrelated noise assumption, the $\mathbb{V}\left[\vec{r}(t)\right]_{ij}$ can be expressed as follow, according equation (S-52):

$$\begin{aligned}
\mathbb{V}\left[\vec{r}(t)\right]_{ij} &= \frac{1}{2m^2\gamma^3}(2\gamma t - 3 + 4\mathrm{e}^{-\gamma t} - \mathrm{e}^{-2\gamma t})\begin{bmatrix} \Gamma_{xx} & 0 & 0 \\ 0 & \Gamma_{yy} & 0 \\ 0 & 0 & \Gamma_{zz} \end{bmatrix} \\
&= S\left(t\right)\begin{bmatrix} \Gamma_{xx} & 0 & 0 \\ 0 & \Gamma_{yy} & 0 \\ 0 & 0 & \Gamma_{zz} \end{bmatrix} \\
\mathbb{V}\left[\vec{r}(t)\right]_{ii} &= S\left(t\right)\Gamma_{ii}
\end{aligned} \tag{S-54}$$

where $S(t)$ abbreviates for $\frac{1}{2m^2\gamma^3}(2\gamma t - 3 + 4\mathrm{e}^{-\gamma t} - \mathrm{e}^{-2\gamma t})$ in eq. (S-54).

Given the $\mathbb{E}\left[\vec{r}(t)\right]_i$ from eq. (S-48) and $\mathbb{V}\left[\vec{r}(t)\right]_{ii}$ from eq. (S-54), similar to eq. (S-29), the $\mathbb{P}(\vec{r},t)$ can be expressed as follow:

$$\begin{aligned}\mathbb{P}(\vec{r},t) &= \prod_i^{x,y,z} \mathcal{N}\left(r_i; \mu = \mathbb{E}\left[\vec{r}(t)\right]_i, \sigma^2 = \mathbb{V}\left[\vec{r}(t)\right]_{ii}\right) \\ &= \prod_i^{x,y,z} \mathcal{N}\left(r_i; \mu = r_{o,i} + \frac{v_{o,i}}{\gamma}(1-\mathrm{e}^{-\gamma t}), \sigma^2 = \Gamma_{ii} S(t)\right)\end{aligned} \tag{S-55}$$

The transition probability function

$$\mathbb{P}(\Delta\vec{r}, t_2, t_1) = \prod_i^{x,y,z} \mathcal{N}\left(\Delta r_i; \mu = \mathbb{E}\left[r_i(t_2)\right] - \mathbb{E}\left[r_i(t_1)\right], \sigma^2 = \Gamma_{ii}\left(S(t_2) - S(t_1)\right)\right) \tag{S-56}$$

The $J_{ii}(t)$ and $\mathbb{E}\left[v_i(t)\right]$ can be expressed as:

$$\begin{aligned}J_{ii}(t) &= \frac{d}{dt}\left(\Gamma_{ii} S(t)\right) = \frac{\Gamma_{ii}}{m^2\gamma^2}(1-\mathrm{e}^{-\gamma t})^2 \\ \mathbb{E}\left[v_i(t)\right] &= \frac{d}{dt}\left(\mathbb{E}\left[r_i(t_2)\right]\right) = \frac{v_{o,i}}{\gamma}(1-\mathrm{e}^{-\gamma t})\end{aligned} \tag{S-57}$$

Substituting $J_{ii}(t)$ and $\mathbb{E}\left[v_i(t)\right]$ to equation S-56, we have:

$$\mathbb{P}(\Delta\vec{r}, t_2, t_1) = \prod_i^{x,y,z} \mathcal{N}\left(\Delta r_i; \mu = \int_{t_1}^{t_2} \mathbb{E}\left[\vec{v}_i(t)\right]\,\mathrm{d}t, \sigma^2 = \int_{t_1}^{t_2} J_{ii}(t)\,\mathrm{d}t\right) \tag{S-58}$$

The two-time correlation function $c_1(\vec{q}, t_1, t_2)$ obtained through XPCS will be:

$$\begin{aligned}c_1(\vec{q}, t_1, t_2) &= \int_{-\infty}^{\infty} \mathbb{P}(\Delta\vec{r}, t_1, t_2) \mathrm{e}^{i\vec{q}\cdot\Delta\vec{r}}\,\mathrm{d}\Delta\vec{r} \\ &= \prod_i^{x,y,z} \int_{-\infty}^{\infty} \mathcal{N}\left(\Delta r_i; \mu = \int_{t_1}^{t_2} \mathbb{E}\left[v_i(t)\right]\,\mathrm{d}t, \sigma^2 = \int_{t_1}^{t_2} J_{ii}(t)\,\mathrm{d}t\right) \mathrm{e}^{iq_i\Delta r_i}\,\mathrm{d}\Delta r_i \\ &= \prod_i^{x,y,z} \int_{-\infty}^{\infty} \frac{1}{\sqrt{2\pi\int_{t_1}^{t_2} J_{ii}(t)\,\mathrm{d}t}} \exp\left(-\frac{\left[\Delta r_i - \int_{t_1}^{t_2}\mathbb{E}\left[v_i(t)\right]\,\mathrm{d}t\right]^2}{2\int_{t_1}^{t_2} J_{ii}(t)\,\mathrm{d}t} + iq_i\Delta r_i\right)\mathrm{d}\Delta r_i \\ &= \prod_i^{x,y,z} \mathrm{e}^{-\frac{1}{2}q_i^2\int_{t_1}^{t_2} J_{ii}(t)\,\mathrm{d}t} \mathrm{e}^{iq_i\int_{t_1}^{t_2}\mathbb{E}[v_i(t)]\,\mathrm{d}t} \\ &= \mathrm{e}^{-\frac{1}{2}\int_{t_1}^{t_2}\left(q_x^2 J_{xx} + q_y^2 J_{yy} + q_z^2 J_{zz}\right)\mathrm{d}t} \mathrm{e}^{i\int_{t_1}^{t_2} q_x\mathbb{E}[v_x(t)] + q_y\mathbb{E}\left[v_y(t)\right] + q_z\mathbb{E}[v_z(t)]\,\mathrm{d}t} \\ &= \mathrm{e}^{-\frac{1}{2}\int_{t_1}^{t_2}\left(\vec{q}\cdot\underline{J}(t)\cdot\vec{q}^{\mathrm{T}}\right)\mathrm{d}t} \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[\vec{v}(t)]\,\mathrm{d}t}\end{aligned} \tag{S-59}$$

where transport coefficient tensor $\underline{J}$ is defined as:

$$\underline{J}(t) = \begin{bmatrix} J_{xx}(t) & 0 & 0 \\ 0 & J_{yy}(t) & 0 \\ 0 & 0 & J_{zz}(t) \end{bmatrix} \tag{S-60}$$

If we have assumed that the noise strength is isotropic ($\Gamma_{ii} = 2m\gamma k_B T$) and transport coefficient is the same for all the components ($J = 3J_{xx} = 3J_{yy} = 3J_{zz}$) without external drive ($\mathbb{E}\left[v_x(t)\right] = \mathbb{E}\left[v_y(t)\right] = \mathbb{E}\left[v_z(t)\right] = 0$), based on equation S-57, $c_1(\vec{q}, t_1, t_2)$ can be expressed as:

$$c_1(\vec{q}, t_1, t_2) = \mathrm{e}^{-\frac{1}{6}\left(q_x^2+q_y^2+q_z^2\right)\int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \tag{S-61}$$

The term

$$q^2 = |\vec{q}|^2 = q_x^2 + q_y^2 + q_z^2 \tag{S-62}$$

defines an Ewald sphere in the reciprocal space where $c_1(\vec{q}, t_1, t_2)$ measured from any point in the sphere are equivalent in the isotropic system.

In the Ornstein–Uhlenbeck process, assuming noise strength in each direction equals to the case in 1-dimension, $\Gamma_{ii} = 2m\gamma k_B T$, $J(\mathrm{t})$ can be expressed as:

$$J(t) = 3J_{ii}(t) = 6\frac{k_B T}{m\gamma}(1 - \mathrm{e}^{-\gamma t})^2 \tag{S-63}$$

Substituting the diffusion constant defined as $D = \frac{k_B T}{m\gamma}$, the $J(t)$ is obtained as:

$$J(t) = 6D(1 - \mathrm{e}^{-\gamma t})^2 \tag{S-64}$$

As $t \to \infty$ where the system reaches equilibrium, $J(t)$ equals to

$$J(t) = 6D \tag{S-65}$$

Based on the equation, we can conclude that, under the conditions of isotropic and uncorrelated noise, the transport coefficient $J(t)$ in 3-dimensional space is related to the transport coefficient in 1-dimension, $J_{ii}(t) = 2D$, as follows,

$$J(t) = nJ_{ii}(t) \triangleq J_n(t) \tag{S-66}$$

where n stands for the number of dimensions in the system.

Therefore, substituting the equation S-66 to equation S-59, the $c_1(\vec{q}, t_1, t_2)$ in 3 dimensions with isotropic and uncorrelated noise in each direction can be expressed as:

$$\begin{aligned} c_1(\vec{q}, t_1, t_2) &= \mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} (nJ_{ii}(t))\,\mathrm{d}t} \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2} \mathbb{E}[\vec{v}(t)]\,\mathrm{d}t} \\ &= \mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J_{nD}(t)\,\mathrm{d}t} \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2} \mathbb{E}[\vec{v}(t)]\,\mathrm{d}t} \end{aligned} \tag{S-67}$$

Here, we expand the physical picture from 1 dimension to 3 dimensions. The equation S-67 is fundamental for further derivation for the equilibrium systems in complex fluid environments, such as laminar or shear banding flow.

E. $c_1(\vec{q}, t_1, t_2)$ in Homodyne Scattering.

E.1. Generalized Equation for Homodyne Scattering. In the combined XPCS and *in situ* Rheology (Rheo-XPCS) experiment, the laminar flow system can be conceptualized as comprising ensembles of flow layers across the gap ($0 \le y \le h$), each possessing the same transport coefficient $J(t)$ but different mean velocities $\mathbb{E}[\vec{v}(y,t)]$. The correlation function can be expressed as:

$$\begin{aligned} c_1(\vec{q}, t_1, t_2) &= \sum_{y_1=1}^{h}\sum_{y_2=1}^{h} \frac{\Delta y_1}{h}\frac{\Delta y_2}{h} c_{1,y_1-y_2}(\vec{q}, t_1, t_2) \\ &= \frac{1}{h^2}\sum_{y_1=1}^{h}\sum_{y_2=1}^{h} \mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2} \mathbb{E}[\vec{v}(y_1,t)]-\mathbb{E}[\vec{v}(y_2,t)]\,\mathrm{d}t} \Delta y_1 \Delta y_2 \\ &= \frac{1}{h^2}\int_0^h\int_0^h \mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2} \mathbb{E}[\vec{v}(y_1,t)]-\mathbb{E}[\vec{v}(y_2,t)]\,\mathrm{d}t} \mathrm{d}y_1 dy_2 \end{aligned} \tag{S-68}$$

where $c_{1,y_1-y_2}(\vec{q}, t_1, t_2)$ denotes as the correlation function between ensembles at position y_1 and y_2 in the gap.

Depending on its position in the gap (y) between the rotor and stator and taking into account the non-equilibrium nature of the system, where the shear rate changes over time, the velocity can be mathematically expressed as follows:

$$\vec{v}(y,t) = v_x(y,t) = \dot{\gamma}(t)y \tag{S-69}$$

Based on the aforementioned equation, the particle mean displacement in Equation S-30 can be derived as follows:

$$\int_{t_1}^{t_2} \mathbb{E}[\vec{v}(y,t)]\,\mathrm{d}t = \int_{t_1}^{t_2} v_x(t)\,\mathrm{d}t = y\int_{t_1}^{t_2} \dot{\gamma}(t)\,\mathrm{d}t \tag{S-70}$$

In a homogeneous system with laminar flow that produces homodyne scattering, the integration of all the ensemble coordinates in Equation S-68 can be transformed from individual particles to spatial coordinates across the gap in the Couette geometry from 0 to h.

$$\begin{aligned} c_1(\vec{q}, t_1, t_2) &= \frac{1}{h^2}\int_0^h\int_0^h \mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2} \mathbb{E}[\vec{v}(y_1,t)]-\mathbb{E}[\vec{v}(y_2,t)]\,\mathrm{d}t} \mathrm{d}y_1 dy_2 \\ &= \left[\mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t}\right] \frac{1}{h^2}\int_0^h\int_0^h \mathrm{e}^{\left[iq\int_{t_1}^{t_2} \dot{\gamma}(t)\cos(\phi(t))\,\mathrm{d}t\right](y_1-y_2)} \mathrm{d}y_1 dy_2 \\ &= \left[\mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t}\right] \mathrm{sinc}^2\left[\frac{1}{2}qh\int_{t_1}^{t_2} \dot{\gamma}(t)\cos(\phi(t))\,\mathrm{d}t\right] \end{aligned} \tag{S-71}$$

where ϕ is the angle between shear/flow direction and $\vec{q}$.

With Siegert Relation S-10, the $c_2(\vec{q}, t_1.t_2)$ can be expressed as:

$$\begin{aligned} c_2(\vec{q}, t_1, t_2) &= 1 + \beta |c_1(\vec{q}, t_1, t_2)|^2 \\ &= 1 + \beta \left[\mathrm{e}^{-q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \right] \mathrm{sinc}^4 \left[\frac{1}{2} qh \int_{t_1}^{t_2} \dot{\gamma}(t) \cos(\phi(t))\,\mathrm{d}t \right] \end{aligned} \quad \text{[S-72]}$$

E.2. Simplified Equation for Homodyne Scattering. In the Rheo-XPCS experiment for system under strong deformation, the ensembles at different positions exhibit a great difference in velocity. Therefore, eq. (S-68) is simplified by ignoring the cross-correlation between different layers where $y_1 \neq y_2$,

$$\begin{aligned} c_1(\vec{q}, t_1, t_2) &= \sum_{y=y_1=y_2=1}^{h} \frac{\Delta y}{h} c_{1,y}(\vec{q}, t_1, t_2) = \frac{1}{h} \sum_{y=1}^{h} \mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2} \mathbb{E}[\vec{v}(y,t)]\,\mathrm{d}t} \Delta y \\ &= \frac{1}{h} \int_0^h \mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2} \mathbb{E}[\vec{v}(y,t)]\,\mathrm{d}t}\,\mathrm{d}y \\ &= \left[\mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \right] \frac{1}{h} \int_0^h \mathrm{e}^{\left[q \int_{t_1}^{t_2} \dot{\gamma}(t)\cos(\phi(t))\,\mathrm{d}t \right] iy}\,\mathrm{d}y \\ &= \left[\mathrm{e}^{-\frac{1}{2}q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \right] \mathrm{sinc}\left[\frac{1}{2} qh \int_{t_1}^{t_2} \dot{\gamma}(t)\cos(\phi(t))\,\mathrm{d}t \right] \mathrm{e}^{i\frac{1}{2}qh \int_{t_1}^{t_2} \dot{\gamma}(t)\cos(\phi(t))\,\mathrm{d}t} \end{aligned} \quad \text{[S-73]}$$

where ϕ is the angle between shear/flow direction and $\vec{q}$.

With Siegert Relation S-10, the $c_2(\vec{q}, t_1.t_2)$ can be expressed as:

$$\begin{aligned} c_2(\vec{q}, t_1, t_2) &= 1 + \beta |c_1(\vec{q}, t_1, t_2)|^2 \\ &= 1 + \beta \left[\mathrm{e}^{-q^2 \int_{t_1}^{t_2} J(t)\,\mathrm{d}t} \right] \mathrm{sinc}^2 \left[\frac{1}{2} qh \int_{t_1}^{t_2} \dot{\gamma}(t) \cos(\phi(t))\,\mathrm{d}t \right] \end{aligned} \quad \text{[S-74]}$$

To consider the equilibrium condition of standard diffusion under constant shear where shear rate is constant as $\dot{\gamma}(t)\cos(\phi(t)) = \dot{\gamma}\cos(\phi)$ and $J(t) = 6D$, the $c_2(\vec{q}, t_1, t_2)$ can be expressed as:

$$c_2(\vec{q}, t_1, t_2) = 1 + \beta \left[\mathrm{e}^{-6q^2 D(t_2 - t_1)} \right] \mathrm{sinc}^2 \left[\frac{1}{2} qh \cos(\phi) \dot{\gamma}(t_2 - t_1) \right] \quad \text{[S-75]}$$

Substituting the absolute time (t_2, t_1) by delayed time $\tau = t_2 - t_1$, the $c_2(\vec{q}, t_1, t_2)$ is reduced to the one-time correlation function, $g_2(\vec{q}, \tau)$ as:

$$g_2(\vec{q}, \tau) = 1 + \beta \left[\mathrm{e}^{-6q^2 D\tau} \right] \mathrm{sinc}^2 \left[\frac{1}{2} qh \cos(\phi) \dot{\gamma}\tau \right] \quad \text{[S-76]}$$

The eq. (S-76) mentioned above is a commonly used homodyne equation that has been widely applied in various studies.

F. Systems with Multiple-Compositions in Non-Equilibrium.

F.1. $c_2(\vec{q}, t_1, t_2)$ ***for Heterodyne Scattering with*** n***-Compositions.*** We can extend the analysis to include the time-dependent fraction of each composition, $x_n(t)$. The total scattered electromagnetic wave, $E(\vec{q}, t)$, at a given time t can then be expressed as a compositionally weighted contribution from the scattered electromagnetic fields of each composition, $E_n(\vec{q}, t)$, as follows:

$$\begin{aligned} E(\vec{q}, t) &= \sum_{n=1}^{N} x_n(t) E_n(\vec{q}, t) \\ 1 &= \sum_{n=1}^{N} x_n(t) \end{aligned} \quad \text{[S-77]}$$

The ensemble average intensity could be calculated as follows:

$$\begin{aligned} \mathbb{E}\left[I(\vec{q}, t) \right] &= \mathbb{E}\left[E(\vec{q}, t) E^*(\vec{q}, t) \right] \\ &= \mathbb{E}\left[\sum_{n=1}^{N} x_n(t) E_n(\vec{q}, t) \sum_{m=1}^{N} x_m(t) E_m^*(\vec{q}, t) \right] \\ &= \sum_{n=1}^{N} x_n^2(t) \mathbb{E}\left[|E_n(\vec{q}, t)|^2 \right] + \sum_{n=1}^{N} \sum_{m \neq n}^{N} x_n(t) x_m(t) \mathbb{E}\left[E_n(\vec{q}, t) E_m^*(\vec{q}, t) \right] \end{aligned} \quad \text{[S-78]}$$

To simply the equation above, let's consider the following assumptions:

1. The average intensity produced by each scatterer is determined by a consistent unit scattering contrast, as all the scatterers are composed of the same materials. This unit scattering contrast remains constant across all scatterers and does not vary with time.

$$\mathbb{E}\left[I_n(\vec{q},t)\right] = \mathbb{E}\left[\left|E_n(\vec{q},t)\right|^2\right] = \mathbb{E}\left[|b|^2\right] \tag{S-79}$$

2. The electromagnetic field scattered from different compositions does not correlate with each other if the signals come from different compositions:

$$\sum_{n=1}^{N}\sum_{m\neq n}^{N} x_n(t)x_m(t)\mathbb{E}\left[E_n(\vec{q},t)E_m(\vec{q},t)\right] = 0 \tag{S-80}$$

With these two conditions, the average intensity could be written as

$$\mathbb{E}\left[I(\vec{q},t)\right] = \mathbb{E}\left[|b|^2\right]\sum_{n=1}^{N} x_n(t)^2 \tag{S-81}$$

The correlation function between time t_1 and t_2 could be computed as:

$$c_1(\vec{q},t_1,t_2) = \frac{\mathbb{E}\left[E(\vec{q},t_2)E^*(\vec{q},t_1)\right]}{\left[\mathbb{E}\left[|E(\vec{q},t_1)|^2\right]\mathbb{E}\left[|E(\vec{q},t_2)|^2\right]\right]^{\frac{1}{2}}} \tag{S-82}$$

Substituting the fractional relation eq. (S-77) to the previous correlation function:

$$\begin{aligned} c_1(\vec{q},t_1,t_2) &= \frac{\mathbb{E}\left[\sum_{m=1}^{N} x_m(t_2)E_m(\vec{q},t_2)\sum_{n=1}^{N} x_n(t_1)E_n^*(\vec{q},t_1)\right]}{\left[\mathbb{E}\left[I(\vec{q},t_1)\right]\mathbb{E}\left[I(\vec{q},t_2)\right]\right]^{\frac{1}{2}}} \\ &= \frac{\left[\sum_{n=1}^{N} x_n(t_2)x_n(t_1)\mathbb{E}\left[E_n(\vec{q},t_2)E_n^*(\vec{q},t_1)\right] + \sum_{n=1}^{N}\sum_{m\neq n}^{N} x_m(t_2)x_n(t_1)\mathbb{E}\left[E_m(\vec{q},t_2)E_n^*(\vec{q},t_1)\right]\right]}{\left[\sum_{n=1}^{N} x_n(t_1)^2\mathbb{E}\left[I_n(\vec{q},t_1)\right]\sum_{n=1}^{N} x_n(t_2)^2\mathbb{E}\left[I_n(\vec{q},t_2)\right]\right]^{\frac{1}{2}}} \end{aligned} \tag{S-83}$$

The correlation function above could be interpreted as the sum of two components: the self-correlating terms for each component plus their cross-term. To simplify and rationalize the correlation function, we assume no spatial correlation of $E(\vec{q},t)$ between scatterers in different composition ($n \neq m$) at different time:

$$\sum_{n=1}^{N}\sum_{m\neq n}^{N} x_n(t_1)x_m(t_2)\mathbb{E}\left[E_m(\vec{q},t_2)E_n^*(\vec{q},t_1)\right] = 0 \tag{S-84}$$

Now, introducing the compositional temporal correlation function, $c_{1,n}(\vec{q},t_1,t_2)$, with form shown in eq. (S-67),

$$\begin{aligned} c_{1,n}(\vec{q},t_1,t_2) &= \frac{\mathbb{E}\left[E_n(\vec{q},t_2)E_n^*(\vec{q},t_1)\right]}{\left[\mathbb{E}\left[E_n(\vec{q},t_2)\right]^2\mathbb{E}\left[E_n^*(\vec{q},t_1)\right]^2\right]^{\frac{1}{2}}} = \frac{\mathbb{E}\left[E_n(\vec{q},t_2)E_n^*(\vec{q},t_1)\right]}{\mathbb{E}\left[I(\vec{q},t)\right]} \\ &= \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_n(t)\,\mathrm{d}t}\,\mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[\vec{v}_n(t)]\,\mathrm{d}t} \end{aligned} \tag{S-85}$$

the self-correlation part of $c_1(\vec{q},t_1,t_2)$ can be written as follows:

$$\begin{aligned} \sum_{n=1}^{N} x_n(t_1)x_n(t_2)\mathbb{E}\left[E_n(\vec{q},t_2)E_n^*(\vec{q},t_1)\right] &= \mathbb{E}\left[I(\vec{q},t)\right]\sum_{n=1}^{N} x_n(t_1)x_n(t_2)c_{1,n}(\vec{q},t_1,t_2) \\ &= \mathbb{E}\left[|b|^2\right]\sum_{n=1}^{N} x_n(t_1)x_n(t_2)\mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_n(t)\,\mathrm{d}t}\,\mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[\vec{v}_n(t)]\,\mathrm{d}t} \end{aligned} \tag{S-86}$$

The correlation function thus could be simplified as:

$$
\begin{aligned}
c_1(\vec{q},t_1,t_2) &= \frac{\mathbb{E}\left[|b|^2\right]\sum_{n=1}^{N} x_n(t_1)x_n(t_2)c_{1,n}(\vec{q},t_1,t_2)}{\mathbb{E}\left[|b|^2\right]\left[\sum_{n=1}^{N} x_n(t_1)^2\sum_{n=1}^{N} x_n(t_2)^2\right]^{\frac{1}{2}}} \\
&= \frac{\sum_{n=1}^{N} x_n(t_1)x_n(t_2)c_{1,n}(\vec{q},t_1,t_2)}{\left[\sum_{n=1}^{N} x_n(t_1)^2\sum_{n=1}^{N} x_n(t_2)^2\right]^{\frac{1}{2}}} \\
&= \frac{1}{f}\sum_{n=1}^{N} x_n(t_1)x_n(t_2)c_{1,n}(\vec{q},t_1,t_2) \\
&= \frac{1}{f}\sum_{n=1}^{N} x_n(t_1)x_n(t_2)\mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_n(t)\,\mathrm{d}t}\,\mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[\vec{v}_n(t)]\,\mathrm{d}t}
\end{aligned}
\tag{S-87}
$$

Let the normalization factor f define as $f = \left[\sum_{n=1}^{N} x_n(t_1)^2\sum_{n=1}^{N} x_n(t_2)^2\right]^{\frac{1}{2}}$.

We then apply the Siegert relation S-10 to obtain the $c_2(\vec{q},t_1,t_2)$ function measured from the XPCS experiment under speckle contrast of β:

$$
\begin{aligned}
c_2(\vec{q},t_1,t_2) &= 1+\beta|c_1(\vec{q},t_1,t_2)|^2 \\
&= 1+\beta\left[c_1(\vec{q},t_1,t_2)c_1^*(\vec{q},t_1,t_2)\right] \\
&= 1+\beta\left[\sum_{n=1}^{N} x_n(t_1)x_n(t_2)c_{1,n}\sum_{m=1}^{N} x_m(t_1)x_m(t_2)c_{1,m}^*\right] \\
&= 1+\frac{\beta}{f^2}\sum_{n=1}^{N}\sum_{m=1}^{N} x_n(t_1)x_n(t_2)x_m(t_1)x_m(t_2)c_{1,n}c_{1,m}^*
\end{aligned}
\tag{S-88}
$$

Subsequently, $c_2(\vec{q},t_1,t_2)$ is obtained by substituting from equation S-85:

$$
c_2(\vec{q},t_1,t_2) = 1+\frac{\beta}{f^2}\sum_{n=1}^{N}\sum_{m=1}^{N}\begin{bmatrix} x_n(t_1)x_n(t_2)x_m(t_1)x_m(t_2)\times \\ \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_n(t)+J_m(t)\,\mathrm{d}t}\times \\ \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[\vec{v}_n(t)]-\mathbb{E}[\vec{v}_m(t)]\,\mathrm{d}t}\end{bmatrix}
\tag{S-89}
$$

In the XPCS experiment, only the real part in the eq. (S-89) can be observed:

$$
\begin{aligned}
c_2(\vec{q},t_1,t_2) &= 1+\frac{\beta}{f^2}\sum_{n=1}^{N}\sum_{m=1}^{N}\mathbf{Re}\begin{bmatrix} x_n(t_1)x_n(t_2)x_m(t_1)x_m(t_2)\times \\ \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_n(t)+J_m(t)\,\mathrm{d}t}\times \\ \mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[\vec{v}_n(t)]-\mathbb{E}[\vec{v}_m(t)]\,\mathrm{d}t}\end{bmatrix} \\
&= 1+\frac{\beta}{f^2}\sum_{n=1}^{N}\sum_{m=1}^{N}\begin{bmatrix} x_n(t_1)x_n(t_2)x_m(t_1)x_m(t_2)\times \\ \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_n(t)+J_m(t)\,\mathrm{d}t}\times \\ \mathbf{Re}\left[\mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[\vec{v}_n(t)]-\mathbb{E}[\vec{v}(t)_m]}\right]\end{bmatrix}
\end{aligned}
\tag{S-90}
$$

The term **Re** denotes taking the real part from the complex term of $\mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[\vec{v}_n(t)]-\mathbb{E}[\vec{v}(t)_m]}$, the cosine part,

$$
\mathbf{Re}\left[\mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[v_n(t)]-\mathbb{E}[v_m(t)]\,\mathrm{d}t}\right] = \cos\left[\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}\left[\vec{v}_n(t)\right]-\mathbb{E}\left[\vec{v}_m(t)\right]\,\mathrm{d}t\right]
\tag{S-91}
$$

For the self-interference part as $n=m$, this term equalizes to 1:

$$
\begin{aligned}
\mathbf{Re}\left[\mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[v_n(t)]-\mathbb{E}[v_n(t)]\,\mathrm{d}t}\right] &= \cos\left[\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}\left[\vec{v}_n(t)\right]-\mathbb{E}\left[\vec{v}_n(t)\right]\,\mathrm{d}t\right] \\
&= \cos\left[0\right] = 1
\end{aligned}
\tag{S-92}
$$

As we further consider that the displacement of each composition is attributed to the flow along direction $\phi_n(t)$ relative to scattering vector $\vec{q}$, the interference term can be simplified as:

$$\mathbf{Re}\left[\mathrm{e}^{i\vec{q}\cdot\int_{t_1}^{t_2}\mathbb{E}[\vec{v}_n(t)]-\mathbb{E}[\vec{v}_m(t)]\,\mathrm{d}t}\right]=\cos\left[q\int_{t_1}^{t_2}\mathbb{E}\left[v_n(t)\right]\cos\left(\phi_n(t)\right)-\mathbb{E}\left[v_m(t)\right]\cos\left(\phi_m(t)\right)\mathrm{d}t\right] \quad \text{[S-93]}$$

The completed form of the time correlation function with heterodyne scattering under time-dependent change in fraction and relative velocity is written as follows:

$$c_2(\vec{q},t_1,t_2)=1+\frac{\beta}{f^2}\sum_{n=1}^{N}\sum_{m=1}^{N}\begin{bmatrix}x_n(t_1)x_n(t_2)x_m(t_1)x_m(t_2)\times\\ \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2}J_n(t)+J_m(t)\,\mathrm{d}t}\times\\ \cos\left[q\int_{t_1}^{t_2}\mathbb{E}\left[v_n(t)\right]\cos\left(\phi_n(t)\right)-\mathbb{E}\left[v_m(t)\right]\cos\left(\phi_m(t)\right)\mathrm{d}t\right]\end{bmatrix} \quad \text{[S-94]}$$

F.2. Heterodyne Scattering with 2 Compositions. To verify the equation in general form, we consider a simpler case. In this context, the heterodyne equation in the system with 2 compositions, static reference (r) and sample (s), consistently moving with relative speed $\mathbb{E}[v]$ and direction ϕ concerning scattering vector $\vec{q}$, the equation could be simplified as:

$$\begin{aligned}c_2(\vec{q},t_1,t_2)&=1+\frac{\beta}{f^2}\begin{bmatrix}\left[x_r(t_1)x_r(t_2)\right]^2\mathrm{e}^{-q^2\int_{t_1}^{t_2}J_r(t)\,\mathrm{d}t}+\\ \left[x_s(t_1)x_s(t_2)\right]^2\mathrm{e}^{-q^2\int_{t_1}^{t_2}J_s(t)\,\mathrm{d}t}+\\ 2x_r(t_1)x_r(t_2)x_s(t_1)x_s(t_2)\mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2}J_s(t)+J_r(t)\,\mathrm{d}t}\cos\left[q\cos\left(\phi\right)\int_{t_1}^{t_2}\mathbb{E}\left[v\right]\,\mathrm{d}t\right]\end{bmatrix}\\ f^2&=\left[x_s(t_1)^2+x_r(t_1)^2\right]\left[x_s(t_2)^2+x_r(t_2)^2\right]\end{aligned} \quad \text{[S-95]}$$

To further consider the equilibrium conditions in the Wiener process or standard diffusion, in which all parameters, including x_s, x_r and v, are independent of time and $J_n(t)=6D_n$, the equation could be further simplified as:

$$\begin{aligned}c_2(\vec{q},t_1,t_2)&=1+\frac{\beta}{f^2}\begin{bmatrix}x_r^4\mathrm{e}^{-6q^2D_r(t_2-t_1)}+x_s^4\mathrm{e}^{-6q^2D_s(t_2-t_1)}+\\ 2x_r^2x_s^2\mathrm{e}^{-3q^2(D_r+D_s)(t_2-t_1)}\cos\left[q\cos\left(\phi\right)\mathbb{E}\left[v\right](t_2-t_1)\right]\end{bmatrix}\\ f^2&=\left[x_s^2+x_r^2\right]^2\end{aligned} \quad \text{[S-96]}$$

Further apply the condition for the composition (x) and delayed time (τ):

$$\begin{aligned}x&=\frac{I_s}{I_s+I_r}=\frac{x_s^2\mathbb{E}\left[I\right]}{x_s^2\mathbb{E}\left[I\right]+x_r^2\mathbb{E}\left[I\right]}=\frac{x_s^2}{x_s^2+x_r^2}=\frac{x_s^2}{f}\\ \tau&=t_2-t_1\end{aligned} \quad \text{[S-97]}$$

The one-time correlation equation for heterodyne scattering $g_2(\vec{q},\tau)$ is shown as:

$$g_2(\vec{q},\tau)=1+\beta\begin{bmatrix}(1-x)^2\mathrm{e}^{-6q^2D_r\tau}+x^2\mathrm{e}^{-6q^2D_s\tau}+\\ 2x(1-x)\mathrm{e}^{-3q^2(D_r+D_s)\tau}\cos\left[q\cos\left(\phi\right)\mathbb{E}\left[v\right]\tau\right]\end{bmatrix} \quad \text{[S-98]}$$

The equation S-98 above is a commonly used heterodyne equation that has been widely applied in various studies.

F.3. Heterodyne Scattering with 3 compositions. We further consider a system constituted of 3 compositions with 2 flowing components as $(1,2)$ and 1 static component as (r). The flowing composition moves with velocity (v_1,v_2) and direction (ϕ_1,ϕ_2) relative to the $\vec{q}$. The heterodyne equation could be expressed as follows:

$$c_2(\vec{q},t_1,t_2)=1+\frac{\beta}{f(t_1,t_2)^2}[c_{2,self}+c_{2,cross}] \quad \text{[S-99]}$$

with the normalization factor written as:

$$f(t_1,t_2)^2=\left[x_1(t_1)^2+x_2(t_1)^2+x_r(t_1)^2\right]\left[x_1(t_2)^2+x_2(t_2)^2+x_r(t_2)^2\right] \quad \text{[S-100]}$$

and $c_{2,self}$ and $c_{2,cross}$ denoted as the self-correlation and cross-correlation between different compositions, respectively. These correlation functions are defined as follows:

$$
\begin{aligned}
c_{2,self} &= \begin{bmatrix} [x_r(t_1)x_r(t_2)]^2\,\mathrm{e}^{-q^2\int_{t_1}^{t_2} J_r(t)\,\mathrm{d}t} + \\ [x_1(t_1)x_1(t_2)]^2\,\mathrm{e}^{-q^2\int_{t_1}^{t_2} J_1(t)\,\mathrm{d}t} + \\ [x_2(t_1)x_2(t_2)]^2\,\mathrm{e}^{-q^2\int_{t_1}^{t_2} J_2(t)\,\mathrm{d}t} + \end{bmatrix} \\
c_{2,cross} &= \begin{bmatrix} 2x_r(t_1)x_r(t_2)x_1(t_1)x_1(t_2)\mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_r(t)+J_1(t)\,\mathrm{d}t}\times \\ \cos\left[q\int_{t_1}^{t_2}\mathbb{E}\left[v_1(t)\right]\cos\left(\phi_1(t)\right)\mathrm{d}t\right] + \\ 2x_r(t_1)x_r(t_2)x_2(t_1)x_2(t_2)\mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_r(t)+J_2(t)\,\mathrm{d}t}\times \\ \cos\left[q\int_{t_1}^{t_2}\mathbb{E}\left[v_2(t)\right]\cos\left(\phi_2(t)\right)\mathrm{d}t\right] + \\ 2x_1(t_1)x_1(t_2)x_2(t_1)x_2(t_2)\mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_2(t)+J_1(t)\,\mathrm{d}t}\times \\ \cos\left[q\int_{t_1}^{t_2}\mathbb{E}\left[v_1(t)\right]\cos\left(\phi_1(t)\right) - \mathbb{E}\left[v_2(t)\right]\cos\left(\phi_2(t)\right)\mathrm{d}t\right] \end{bmatrix}
\end{aligned}
\tag{S-101}
$$

The equation shown above represents the heterodyne scattering function for a system with three components, and its calculation involves a large number of parameters. Due to the complexity of this calculation, it becomes challenging to directly extract the physical parameters from the measured data using this equation. To overcome this difficulty, it is necessary to employ approximations based on the specific characteristics of the physical system. The following approximations are commonly utilized:

1. The intrinsic dynamics of the reference position is much slower than that of the flowing composition as $J_r = 0$ and, therefore, $c_{1,r} = 1$.
2. The flow direction of each composition is dominated at the horizontal direction and, therefore, $\phi_1 = \phi_2 = \theta$, where θ is the angle between $\vec{q}$ and the horizontal reference of $\theta = 0$.

With all the reasonable assumptions above, the final form of the heterodyne scattering function for 3 flowing bands, which will be utilized for analysis in this paper, is as follows:

$$
\begin{aligned}
c_{2,self} &= \begin{bmatrix} [x_r(t_1)x_r(t_2)]^2 + \\ [x_1(t_1)x_1(t_2)]^2\,\mathrm{e}^{-q^2\int_{t_1}^{t_2} J_1(t)\,\mathrm{d}t} + \\ [x_2(t_1)x_2(t_2)]^2\,\mathrm{e}^{-q^2\int_{t_1}^{t_2} J_2(t)\,\mathrm{d}t} + \end{bmatrix} \\
c_{2,cross} &= \begin{bmatrix} 2x_r(t_1)x_r(t_2)x_1(t_1)x_1(t_2)\mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_1(t)\,\mathrm{d}t}\times \\ \cos\left[q\cos\left(\theta\right)\int_{t_1}^{t_2}\mathbb{E}\left[v_1(t)\right]\,\mathrm{d}t\right] + \\ 2x_r(t_1)x_r(t_2)x_2(t_1)x_2(t_2)\mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_2(t)\,\mathrm{d}t}\times \\ \cos\left[q\cos\left(\theta\right)\int_{t_1}^{t_2}\mathbb{E}\left[v_2(t)\right]\,\mathrm{d}t\right] + \\ 2x_1(t_1)x_1(t_2)x_2(t_1)x_2(t_2)\mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J_2(t)+J_1(t)\,\mathrm{d}t}\times \\ \cos\left[q\cos\left(\theta\right)\int_{t_1}^{t_2}\mathbb{E}\left[v_1(t)\right] - \mathbb{E}\left[v_2(t)\right]\,\mathrm{d}t\right] \end{bmatrix}
\end{aligned}
\tag{S-102}
$$

3. Formalization of Physical Parameters

A. Analysis in Fig. 3 Panels C and D. In an ideal situation of a non-equilibrium model, various time-dependent parameters $p(t)$ within the model, such as $J(t)$, $\dot{\gamma}(t)$ and $\mathbb{E}\left[v(t)\right]$, should be continuous arbitrary where each frame of t corresponds to a specific value. Consequently, to accurately capture the dynamics evolution over time, the fitting process should involve thousands of parameters that correspond to each measured frame, as follows:

$$
p(t) : p(t=0), p(1), p(2) \cdots\cdots p(t) \tag{S-103}
$$

This analysis approach is employed in the results shown in Fig. 3 Panels C and D of the manuscript. However, when practically analyzing extended time procedures, for the sake of streamlining the fitting process and minimizing the parameter count, these time-dependent parameters can be effectively approximated using various functions.

B. Analysis in Fig. 2. In the analysis of our Molecular Dynamics (MD) simulation, where the system is homogeneous and the temperature exhibits a smooth transition from high to low, the trend of the $J(t)$ is represented by the error function (erfc) as follow:

$$
J(t) = J_{scale}\mathrm{erfc}\left(t_{scale}(t - t_{shift})\right) + J_{shift} \tag{S-104}
$$

which J_{scale}, J_{shift}, t_{scale} and t_{shift} are parameters to either stretch or shift the error function in different axis to accommodate the actual change of $J(t)$ along time.

C. Analysis in Fig. 3 Panels A and B. To more effectively model the intrinsic dynamics observed during the relaxation process using a simplified approach, we have represented the trend of $J(t)$ through a power law that incorporates three parameters: the scale factor (J_o), the exponent (b), and a vertical shift (D_o),

$$J(t) = J_o\, t^b + D_o \qquad \text{[S-105]}$$

was utilized to formalize the relation between $J(t)$ and time t. In Equation (S-105), J_o represents the scaling of the transport coefficient, and D_o signifies the intrinsic diffusion rate of the system at equilibrium. The exponent b delineates the power-law behavior.

D. Analysis in Fig. 4. In the analysis of multiple-banding system, the function of the physical parameters for the two flowing components, including mean velocity $v_1(t)$ and $v_2(t)$, transport coefficient $J_1(t)$ and $J_2(t)$, and fraction $x_1(t)$ and $x_2(t)$, are divided into 2 segments at $t = 15$ when the dynamics process changes its trend. This segmentation in the dynamics analysis arises from the non-monotonic behavior observed in the variables $J(t)$,v, and x_n, where each displays a pattern of increase followed by a decrease. Such trends complicate the use of a single function to precisely capture these variations accurately. Consequently, second order two-time intensity autocorrelation function (c_2) was segmented into two parts at the $t = 15s$, each modeled by distinct functions. To describe v_1 and v_2, $J_1(t)$ and $J_2(t)$ at different segments, a power law is utilized as:

$$\begin{aligned}
J_1(t) &= \begin{cases} a_{J,1,1}t^{b_{J,1,1}} + c_{J,1,1} & : t < 15 \\ a_{J,1,2}t^{b_{J,1,2}} + c_{J,1,2} & : t \geq 15 \end{cases} \\
J_2(t) &= \begin{cases} a_{J,2,1}t^{b_{J,2,1}} + c_{J,2,1} & : t < 15 \\ a_{J,2,2}t^{b_{J,2,2}} + c_{J,2,2} & : t \geq 15 \end{cases} \\
v_1(t) &= \begin{cases} a_{v,1,1}t^{b_{v,1,1}} + c_{v,1,1} & : t < 15 \\ a_{v,1,2}t^{b_{v,1,2}} + c_{v,1,2} & : t \geq 15 \end{cases} \\
v_2(t) &= \begin{cases} a_{v,2,1}t^{b_{v,2,1}} + c_{v,2,1} & : t < 15 \\ a_{v,2,2}t^{b_{v,2,2}} + c_{v,2,2} & : t \geq 15 \end{cases}
\end{aligned} \qquad \text{[S-106]}$$

where scale (a), exponent (b), and vertical shift (c) are fitting parameters of the power law and labeled by subscript corresponding to its (Parameters, Segment, Component).

Additional constraints are imposed on the fitting process for $x_1(t)$ and $x_2(t)$, taking into account the inherent limits of fraction growth and reduction, which are confined within the range of 0 and 1.

$$\begin{aligned}
x_1(t) &= \begin{cases} d_{x,1,1} - a_{x,1,1}\mathrm{e}^{b_{x,1,1}(t-c_{x,1,1})} & : t < 15 \\ a_{x,1,2}\mathrm{e}^{b_{x,1,2}(t-c_{x,1,2})} & : t \geq 15 \end{cases} \\
x_2(t) &= \begin{cases} d_{x,2,1} - a_{x,2,1}\mathrm{e}^{b_{x,2,1}(t-c_{x,2,1})} & : t < 15 \\ a_{x,2,2}\mathrm{e}^{b_{x,2,2}(t-c_{x,2,2})} & : t \geq 15 \end{cases}
\end{aligned} \qquad \text{[S-107]}$$

Any negative values in $x_1(t)$ and $x_2(t)$ is substituted by 0.

The interpretation of the physical parameters in these model functions could vary depending on the specific context of their application. For instance, b_n in S-105 could relate to the exponent that characterizes the time dependency of the dynamics rate as $\dot{\gamma} \sim t^{b_n}$ when it is extracted from a system undergoes relaxations or deformations.

4. Application in Model Systems

A. Classical Non-Equilibrium Dynamics Model. In the following section, we apply equation S-67 to several classical processes that match the description of Langevin dynamics, including the Wiener process (standard diffusion), Ornstein–Uhlenbeck process, and Brownian oscillator. By analyzing these dynamic processes, we can estimate the transport coefficients and, hopefully, gain insight into how this physical parameter represents the dynamic behavior of the system.

A.1. Wiener Process / Standard Diffusion. In this case of 1-dimensional Wiener process or standard diffusion, fluctuations ($\zeta(t)$) exist at the velocity level as $\Gamma = 2D$. Under such conditions, the Langevin equation of the Wiener process can be expressed as (3, at eq. (9.2)):

$$\dot{x} = v_o + R(t) = v_o + \sqrt{2D}\zeta(t) \qquad \text{[S-108]}$$

Here, $R(t)$ represents a random fluctuation in velocity attributed to a stationary, Gaussian, δ-correlated, Markov process $\zeta(t)$. It possesses the following properties:

$$\mathbb{E}\left[\zeta(t)\zeta(t')\right] = \delta(t - t') \qquad \text{[S-109]}$$

where v_o and x_0 are the initial velocity and position for the process.

The mean velocity $\mathbb{E}[v(t)]$, mean position $\mathbb{E}[x(t)]$, variance of position $\mathbb{V}[x(t)]$ and transport coefficient $J(t)$ of the ensemble are obtained as:

$$\begin{aligned}
\mathbb{E}[v(t)] &= v_o \\
\mathbb{E}\left[v(t)v(t')\right] &= v_o^2 + 2D\delta(t-t') \\
\mathbb{E}[x(t)] &= x_0 + v_o t \\
\mathbb{V}[x(t)] &= \mathbb{E}\left[x(t)^2\right] - \mathbb{E}[x(t)]^2 = 2Dt \\
J(t) &= \frac{d}{dt}\left(\mathbb{V}[x(t)]\right) = 2D
\end{aligned} \tag{S-110}$$

Substituting the value of $\mathbb{E}[v(t)]$ and $\mathbb{V}[x(t)]$ from equation S-110 to equation S-40, the $c_1(q,t_1,t_2)$ is obtained based on as:

$$c_1(q,t_1,t_2) = \mathrm{e}^{-q^2 D(t_2-t_1)}\mathrm{e}^{iqv_o(t_2-t_1)} \tag{S-111}$$

Equation (S-111) is the $c_1(q,t_1,t_2)$ for the model of Advection-Diffusion in 1-dimension, a fluid transports with external driven bulk movement (v_o) and diffusion (D) of the substance due to molecular random motion.

Considering the case in equilibrium state as changing the absolute time (t_1,t_2) to delayed time τ as $\tau = t_2 - t_1$:

$$g_1(q,\tau) = \mathrm{e}^{-q^2 D\tau}\mathrm{e}^{iqv_o\tau} \tag{S-112}$$

Further considering diffusion is the only dynamics in the system and there is no bulk movement in the system as $v_o = 0$, the equation S-112 can be reduced to the simplest model that commonly applied in XPCS analysis:

$$g_1(q,\tau) = \mathrm{e}^{-q^2 D\tau} \tag{S-113}$$

A.2. Ornstein–Uhlenbeck Process. In **Ornstein–Uhlenbeck Process**, the external force (F_{ex}) is set to zero while the drift coefficient (γ) remains non-zero. This configuration gives rise to an Ornstein–Uhlenbeck process. The Ornstein–Uhlenbeck process exhibits a mean-reverting behavior and is commonly used to model systems with a tendency to return to equilibrium. In such a case, the Langevin equation is given by (3, at eq. (6.1)):

$$m\dot{v} = -m\gamma v(t) + \eta(t) \tag{S-114}$$

In the Ornstein–Uhlenbeck process, the thermal fluctuation noise in velocity is attributed to collisions between particles and solvent with the level of $\Gamma = 2m\gamma k_B T$. Then, the mean velocity $\mathbb{E}[v(t)]$, mean position $\mathbb{E}[x(t)]$ and variance of position $\mathbb{V}[x(t)]$ of the ensemble is solved as:

$$\begin{aligned}
\mathbb{E}[v(t)] &= v_o\mathrm{e}^{-\gamma t} \\
\mathbb{E}[x(t)] &= x_0 + \frac{v_o}{\gamma}(1-\mathrm{e}^{-\gamma t}) \\
\mathbb{V}[x(t)] &= \mathbb{E}\left[x(t)^2\right] - \mathbb{E}[x(t)]^2 = \frac{k_B T}{m\gamma^2}(2\gamma t - 3 + 4\mathrm{e}^{-\gamma t} - \mathrm{e}^{-2\gamma t}) \\
&= \frac{D}{\gamma}(2\gamma t - 3 + 4\mathrm{e}^{-\gamma t} - \mathrm{e}^{-2\gamma t})
\end{aligned} \tag{S-115}$$

where D is defined as the diffusion constant of the system and is equal to $D = \frac{k_B T}{m\gamma}$

The dynamical transport coefficient $J(t)$ is defined as:

$$\begin{aligned}
J(t) = \frac{d}{dt}\left(\mathbb{V}[x(t)]\right) &= \frac{d}{dt}\left(\frac{D}{\gamma}(2\gamma t - 3 + 4\mathrm{e}^{-\gamma t} - \mathrm{e}^{-2\gamma t})\right) \\
&= 2D(1 - 2\mathrm{e}^{-\gamma t} + \mathrm{e}^{-2\gamma t}) \\
&= 2D\left(1 - \mathrm{e}^{-\gamma t}\right)^2
\end{aligned} \tag{S-116}$$

The two-time correlation function $c_1(q,t_1,t_2)$ obtained through XPCS will be:

$$\begin{aligned}
c_1(q,t_1,t_2) &= \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J(t)\,\mathrm{d}t}\,\mathrm{e}^{iq\int_{t_1}^{t_2}\mathbb{E}[v(t)]\,\mathrm{d}t} \\
&= \mathrm{e}^{-q^2 D\int_{t_1}^{t_2}\left(1-\mathrm{e}^{-\gamma t}\right)^2\mathrm{d}t}\,\mathrm{e}^{iqv_o\int_{t_1}^{t_2}\exp(-\gamma t)\,\mathrm{d}t}
\end{aligned} \tag{S-117}$$

A.3. Brownian Oscillator. In a Brownian oscillator with potential of $V(x) = \frac{1}{2}m{\omega_o}^2x^2$, the conservative force in the particle is $m{\omega_o}^2x$ as $-\frac{\partial V(x)}{\partial x} = -m{\omega_o}^2x$. Therefore, the Langevin Equation (LE) for the Brownian oscillator is: (3, at eq. (13.1))

$$m\dot{v} = -m\gamma v + \eta(t) - m{\omega_o}^2x \tag{S-118}$$

For the Brownian oscillator with initial conditions of $x(0) = x_0$ and $v(0) = v_o$, the value of $\mathbb{E}[v(t)]$,$\mathbb{E}[x(t)]$ and $\mathbb{V}[x(t)]$ is solved as: (5, at eq. (214))

$$\begin{aligned}
\mathbb{E}[v(t)] &= \frac{\mathrm{e}^{-\gamma t/2}}{\omega_s}\left[-x_0({\omega_o}^2\sin\omega_s t) + v_o(\omega_s\cos\omega_s t - \frac{1}{2}\gamma\sin\omega_s t)\right] \\
\mathbb{E}[x(t)] &= \frac{\mathrm{e}^{-\gamma t/2}}{\omega_s}\left[x_0(\omega_s\cos\omega_s t + \frac{1}{2}\gamma\sin\omega_s t) + v_o(\sin\omega_s t)\right] \\
\mathbb{V}[x(t)] &= \frac{k_BT}{m{\omega_o}^2}\left[1 - \mathrm{e}^{-\gamma t}(\frac{\gamma^2}{2\omega_s^2}\sin^2\omega_s t + \frac{\gamma}{2\omega_s}\sin 2\omega_s t + 1)\right]
\end{aligned} \tag{S-119}$$

where ω_s is the reduced frequency given as:

$$\omega_s = \left({\omega_o}^2 - \frac{1}{4}\gamma^2\right)^{\frac{1}{2}} \tag{S-120}$$

With the knowledge of $\mathbb{V}[x(t)]$ and $\mathbb{V}[v(t)]$, the transport coefficient $J(t)$ for Brownian oscillator can be written as: (5, at eq. (213))

$$J(t) = \frac{d}{dt}\left(\mathbb{V}[x(t)]\right) = \frac{2D\gamma^2}{{\omega_s}^2}\mathrm{e}^{-\gamma t}\sin^2\omega_s t \tag{S-121}$$

Substituting the value of $\mathbb{E}[v(t)]$ and $J(t)$ from equations S-119 and S-121 to equation S-40, the two-time correlation function for XPCS, $c_1(t_1, t_2)$, becomes,

$$\begin{aligned}
c_1(q, t_1, t_2) &= \mathrm{e}^{-\frac{1}{2}q^2\int_{t_1}^{t_2} J\,\mathrm{d}t}\,\mathrm{e}^{iq\int_{t_1}^{t_2}\mathbb{E}[v(t)]\,\mathrm{d}t} \\
&= \exp\left(-\frac{D\gamma^2q^2}{{\omega_s}^2}\int_{t_1}^{t_2}\mathrm{e}^{-\gamma t}\sin^2\omega_s t\,\mathrm{d}t\right) \\
&\times \exp\left(i\frac{q\mathrm{e}^{-\gamma t/2}}{\omega_s}\int_{t_1}^{t_2} -x_0({\omega_o}^2\sin\omega_s t) + v_o(\omega_s\cos\omega_s t - \frac{1}{2}\gamma\sin\omega_s t)\,\mathrm{d}t\right)
\end{aligned} \tag{S-122}$$

B. Analysis in Transport Coefficient. As delineated in section 3, if the internal and external forces, such as conservative forces or hydrodynamic forces, are described by complicated functions, $J(t)$ and $\mathbb{V}[x(t)]$ are not analytically solvable. Therefore, we propose to extract $J(t)$ with arbitrary profile functions (e.g., eq. (S-104)) to capture non-equilibrium dynamics in such systems. However, the physical interpretation of $J(t)$ and $\mathbb{V}[x(t)]$ are not clear. On the other hand, throughout the derivations in the previous section A, the $J(t)$ and $\mathbb{V}[x(t)]$ are analytically resolved from the Langevin equation for well-defined models of the Wiener process, Ornstein–Uhlenbeck process, and Brownian oscillator. Below, we will show that the estimation of $J(t)$ and $\mathbb{V}[x(t)]$ from these models facilitates a better understanding of their physics.

B.1. Wiener Process / Standard Diffusion. For Wiener process, $J(t)$ is obtained as:

$$J(t) = 2D \tag{S-123}$$

Since there are no drift or external forces applied to the particles, the system is in equilibrium and the dynamics of the particles are independent of time. Therefore, the obtained $J(t)$ correlates with the diffusion constant.

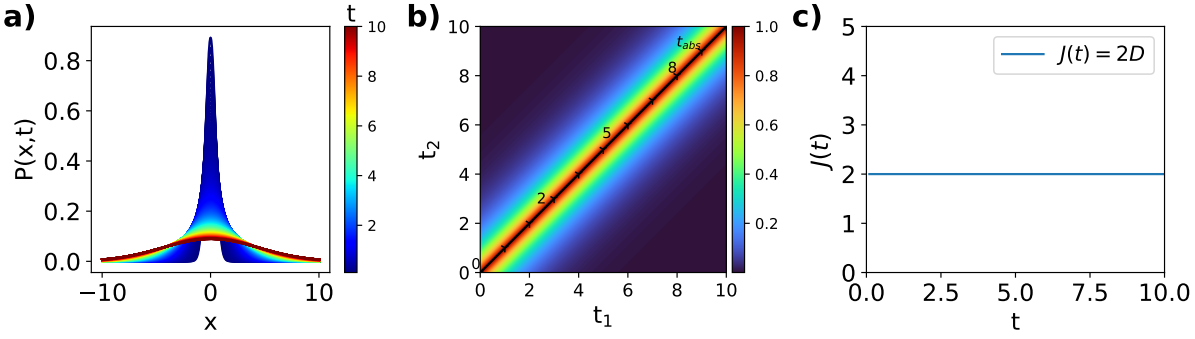

Fig. S1. Simulation for Wiener Process / Standard Diffusion with $D = 1$ and $x_0 = 0$. a) $P(x, t)$ represents the probability density function as time evolves. b) The figure illustrates the two-time correlation function, $c_2(q = 1, t_1, t_2)$. c) $J(t)$ is the transport coefficient described in this work. In such an equilibrium state, the dynamical transport coefficient $J(t) = 2D$ is independent of time.

B.2. Ornstein–Uhlenbeck Process. For Ornstein–Uhlenbeck process, $J(t)$ is obtained as:

$$J(t) = 2D(1 - \mathrm{e}^{-\gamma t})^2 \qquad \text{[S-124]}$$

In the Ornstein–Uhlenbeck process, a drift acts on the particles. At $t = 0$, when the velocity of the particles is at maximum, the drift force is also maximized. $J(t)$ starts at 0, and particles have a uniform bulk movement with v_o. As time evolves, the velocity of the particles and the effect of drift gradually decreases. As the results show, $J(t)$ gradually increases. Eventually, the system reaches equilibrium and the drift effect is minimized after a long time. In this case, $J(t)$ reaches a plateau at $2D$, indicating that the system is at equilibrium just like the result from the Wiener process.

$$J_{eq}(t) = \lim_{t\to\infty} 2D(1 - \mathrm{e}^{-\gamma t})^2 \quad = 2D \qquad \text{[S-125]}$$

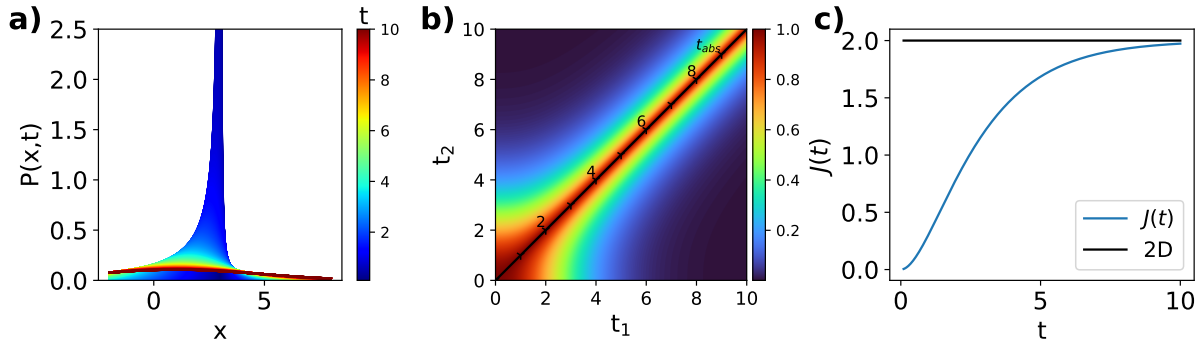

Fig. S2. Simulation for Ornstein–Uhlenbeck Process with $D = 1, \gamma = 0.5, v_o = -1$ and $x_0 = 3$. The dynamical transport coefficient $J(t) = 2D(1 - \mathrm{e}^{-\gamma t})^2$.

B.3. Brownian Oscillator. For Brownian oscillator, $J(t)$ is obtained as:

$$J(t) = \frac{2D\gamma^2}{{\omega_s}^2}\mathrm{e}^{-\gamma t}\sin(\omega_s t)^2 \qquad \text{[S-126]}$$

Recall that ω_s is a reduced frequency defined in Equation S-120 as $\omega_s = \left({\omega_o}^2 - \frac{1}{4}\gamma^2\right)^{\frac{1}{2}}$. In the case of underdamping oscillator as $\omega_o > \frac{1}{2}\gamma$, ω_s is a real and positive value.

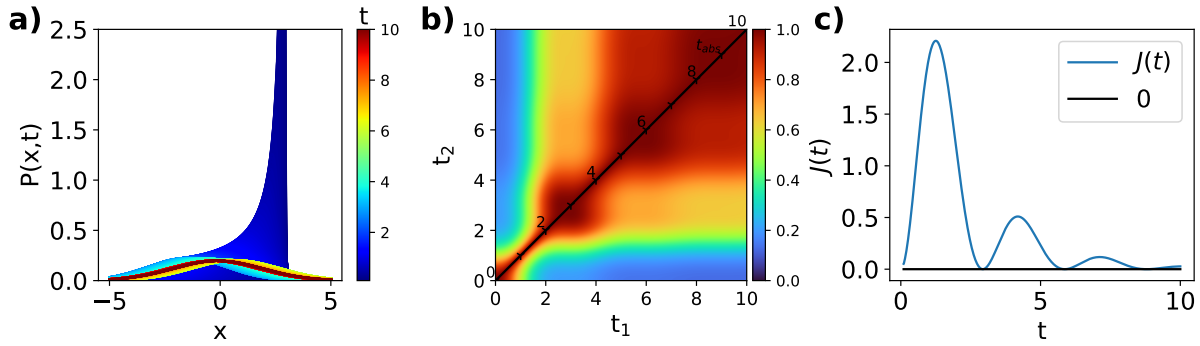

Fig. S3. Simulation for underdamped Brownian Oscillator with $D = 10, \gamma = 0.5, \omega_o = 1.1, v_o = 1$ and $x_0 = 3$.

However, in the case of an overdamped oscillator with $\omega_o < \frac{1}{2}\gamma$, ω_s is an imaginary number, and the trend of equation S-126 is less obvious. Instead, we define a new simplified term of "simplified drift" as $\gamma_s = \left(\gamma^2 - 4{\omega_o}^2\right)^{\frac{1}{2}}$. The term is a real and positive number when $\omega_o < \frac{1}{2}\gamma$ and consists of relations with ω_s as $\gamma_s = i2\omega_s$. Substituting γ_s into equation S-126 and applying a relation $\sin(ix) = i\sinh(x)$, we have an equivalent equation for an overdamped oscillator:

$$J(t) = \frac{8D\gamma^2}{\gamma_s^2}\mathrm{e}^{-\gamma t}\sinh\left(\frac{1}{2}\gamma_s t\right)^2 \qquad \text{[S-127]}$$

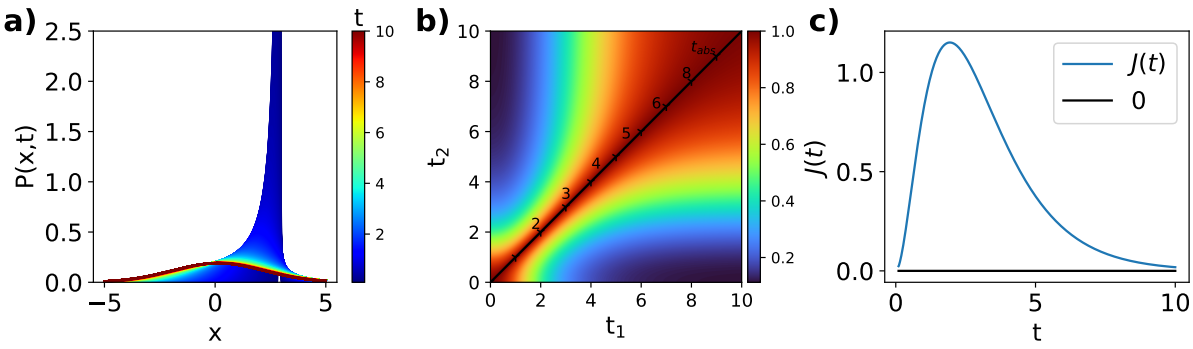

Fig. S4. Simulation for overdamped Brownian Oscillator with $D = 1, \gamma = 1.1, \omega_o = 0.5, v_o = 1$ and $x_0 = 3$.

With eq. (S-126) and eq. (S-127), the dynamics of the Brownian oscillator are obvious. For $t \to \infty$, the term of $\mathrm{e}^{-\gamma t}$ dominates $J(t)$ and $J(t) = 0$. This means that the drift force and elastic force cancel out at equilibrium and the system remains static. For $t \to 0$, the oscillation term $\left(\sin\left(\omega_s t\right)^2\right)$ in eq. (S-126) or $\left(\sinh\left(\frac{1}{2}\gamma_s t\right)^2\right)$ in eq. (S-127) dominates $J(t)$. However, $J(t)$ as shown in fig. S3 and S4 , the oscillation signals vanish much faster for $\sinh^2 \frac{1}{2}\gamma_s t$, as the drift force prevails in the overdamped oscillator.

To reduce the Brownian oscillator to the Ornstein-Uhlenbeck process, we have no external force in the system as

$$\begin{aligned} F_{ex} &= m\omega_o^2 x = \omega_o = 0 \\ \gamma_s &= \left(\gamma^2 - 4{\omega_o}^2\right)^{\frac{1}{2}} = \gamma \end{aligned} \qquad \text{[S-128]}$$

Substituting $\gamma_s = \gamma$, the expression of $J(t)$ can be written as:

$$\begin{aligned} J(t) &= \frac{8D\gamma^2}{\gamma^2}\mathrm{e}^{-\gamma t}\sinh\left(\frac{1}{2}\gamma t\right)^2 \\ &= 8D\mathrm{e}^{-\gamma t}\left(\frac{1-\mathrm{e}^{-\gamma t}}{2\mathrm{e}^{-\frac{1}{2}\gamma t}}\right)^2 \\ &= 2D\left(1-\mathrm{e}^{-\gamma t}\right)^2 \end{aligned} \qquad \text{[S-129]}$$

By setting ω_0 to 0, we simplify the dynamics from a Brownian Oscillator to an Ornstein-Uhlenbeck process. Additionally, if the system reaches equilibrium as t tends to infinity as in equation (S-125), the dynamical process further simplifies from an Ornstein-Uhlenbeck process to a Wiener process, which is the most fundamental stochastic dynamics process.

C. Molecular Dynamics Simulation. To verify the proposed model, we simulate a solution of non-interacting particles (phantom particles) in a non-equilibrium system. The solvent is implicitly modeled by the Langevin thermostat, and the equation of motion of the n-th particle is,

$$\dot{\vec{v}}_n = -\gamma\vec{v}_n + \frac{\vec{\eta}_n(t)}{m} \qquad \text{[S-130]}$$

where the mass, m, is set to unity for all particles, $\vec{v}_n(t)$ is the velocity of the n-th particle. The stochastic force acting on the n-th particle, $\vec{\eta}_n(t)$, satisfies the following conditions,

$$\begin{aligned} \mathbb{E}\left[\vec{\eta}_n(t)\right] &= \vec{0} \\ \mathbb{E}\left[\vec{\eta}_n(t) \otimes \vec{\eta}_n(t')\right]_{ij} &= 6k_B T\gamma\delta_{ij}\delta(t-t') \end{aligned} \qquad \text{[S-131]}$$

The friction coefficient, γ, is set to $\gamma = 1000\frac{m}{\tau}$, where $\tau = \sigma\left(\frac{m}{\epsilon}\right)^{\frac{1}{2}}$ is the reduced time, σ and ϵ are reduced length and energy, respectively. The velocity-Verlet algorithm is used to integrate the equation of motion, with an integration time step $\Delta t = 0.001\tau$. The simulation consists of three stages with different temperatures:

1. The temperature was kept constant at $T = 5\frac{\epsilon}{k_B}$.
2. The temperature linear decreased with time from $T = 5\frac{\epsilon}{k_B}$ to $T = 0.1\frac{\epsilon}{k_B}$.
3. The temperature was maintained constant at $T = 0.1\frac{\epsilon}{k_B}$.

Each stage lasts for 1τ, that is, 1000 simulation steps. All simulations were performed using the Large Scale Atomic / Molecular Massively Parallel Simulator (LAMMPS) (6) under periodic boundary conditions.

In the paper, c_2 and its corresponding analysis for bins at $q_x = 6$, $q_y = q_x = 6$ and $q_x = q_y = q_z = 6$ have been demonstrated. Here, we provide the bins at $q_x = 3$, $q_y = q_x = 3$ and $q_x = q_y = q_z = 3$ to further verify the non-equilibrium analysis approach. Again, $J(t)$ obtained from two respective approaches, derived from analyzing c_2 of XPCS and spatial averaging of the particle positions in the simulation box, shows consistency. This result provides evidence that $J(t)$ accurately reflects the actual physical quantity in the system, validating the extraction approach of $J(t)$.

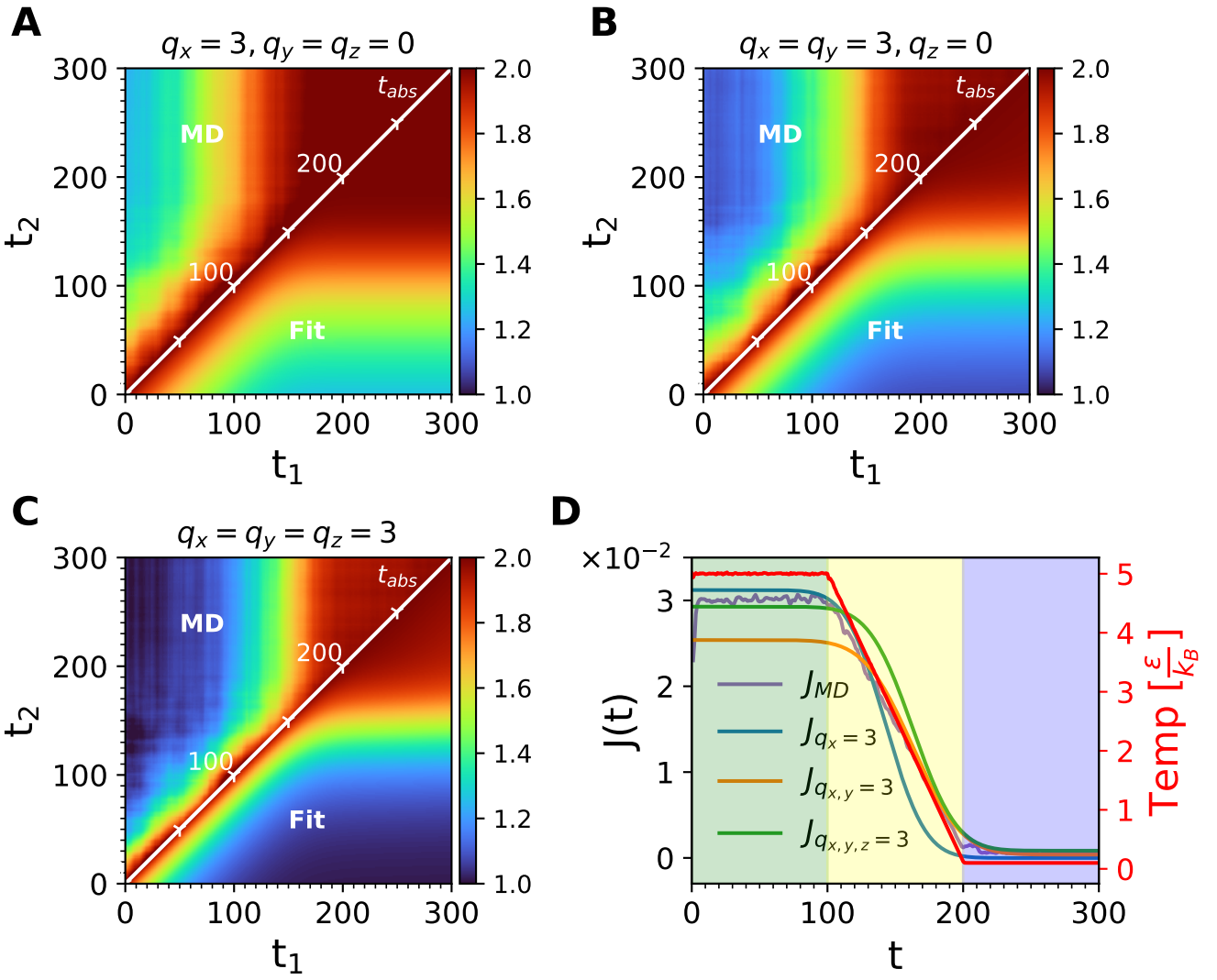

Fig. S5. Application of the model analysis on a simulated non-equilibrium system reveals three distinct stages of temperature change, which are visually represented by the filled-in background color.

5. Correlating Microscopic Dynamics with Macroscopic Rheology through $J(t)$ in Rheo-XPCS

A. Supplementary Results: Recovery following a 10 Pa Creep Test. Beyond the relaxation data for the 100 Pa creep test illustrated in Figures 3A and 3C of the primary manuscript, we further examined the XPCS data derived from a 10 Pa creep test, as detailed in Figure 2A as reference (7). The aggregated findings of the 10 Pa and 100 Pa creep tests are presented in Figure Fig. S6 in this document.

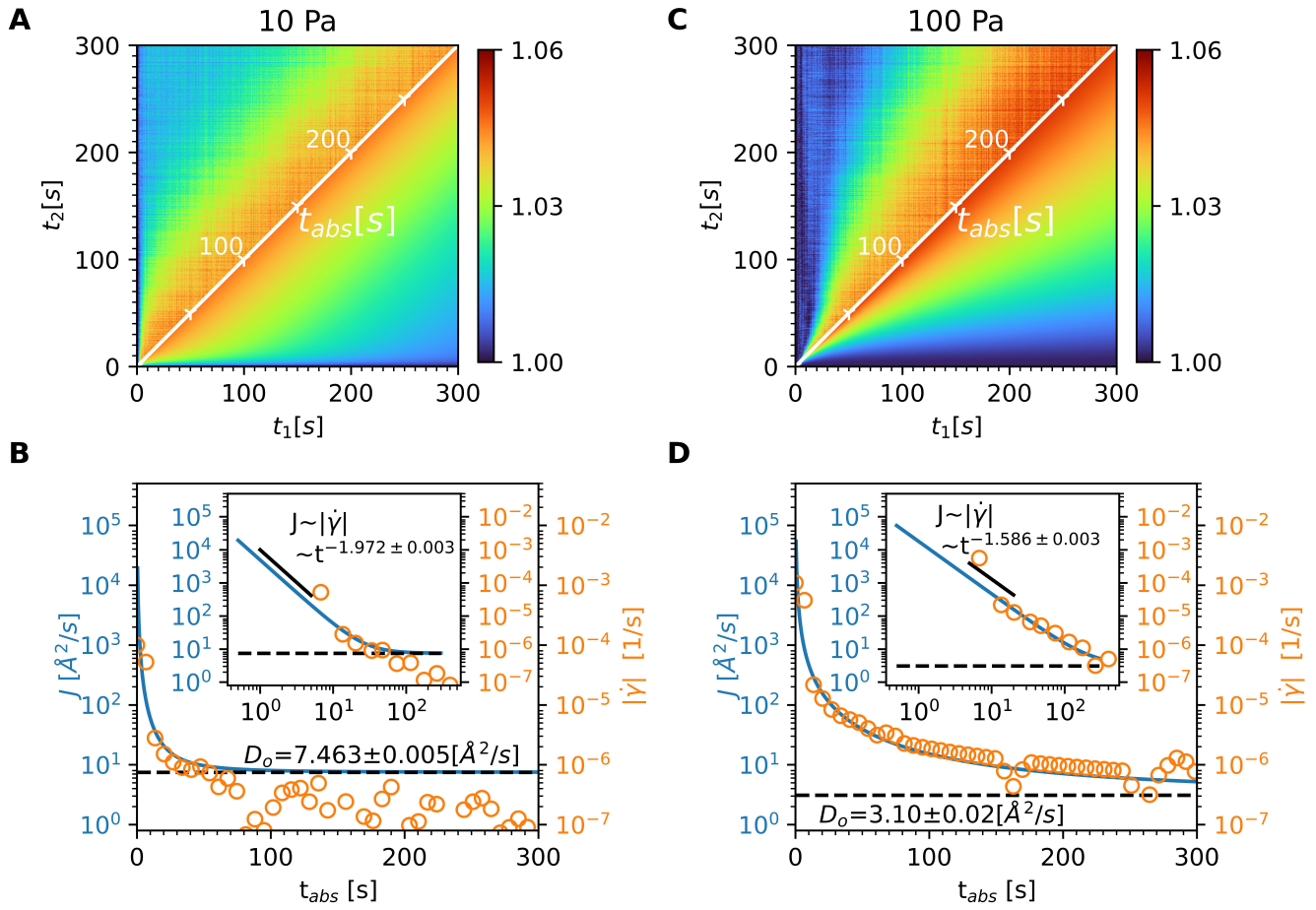

Fig. S6. Comparative Analysis of the $J(t)$ and the shear ($\dot{\gamma}(t)$) During the Recovery Phase of Creep Tests. Panels A and B illustrate the relationship between $J(t)$ and $\dot{\gamma}(t)$ under an applied stress ($\sigma(t)$) of 10 Pa, while Panels C and D show the same under a $\sigma(t)$ of 100 Pa (also shown in Fig 3 A and B in the main texts). From Panels B and D, a consistent scaling relation of $\frac{k_BT}{\pi r}$ is observed between $J(t)$ and $\dot{\gamma}(t)$, independent from the applied $\sigma(t)$ in the creep test.

Analysis of Fig. S6 yields several key insights:

1. During the creep tests, the $J(t)$ diminishes more swiftly at the lower stress level (10 Pa) with an exponent of -1.972, in contrast to the -1.586 exponent observed under the higher stress condition (100 Pa). This trend aligns with the results of both XPCS, evidenced by a pronounced increase in c_2 along the t_{abs} diagonal, and rheological data, which show $\dot{\gamma}(t)$ approaching zero as the strain quickly returns to the baseline level of irrecoverable strain.

2. The estimated values of D_o from both creep tests are similar in magnitude. However, the lack of extended c_2 data for the 100 Pa creep test compromises the precision of D_o, leading to the observed variance in the values D_o between panels B and D.

3. The rapid recovery observed in the 10 Pa creep test results in a rapid decrease in $\dot{\gamma}(t)$, potentially falling below the sensitivity range of the measurement instruments, thus casting doubt on the reliability of the data points with $\dot{\gamma} < 10^{-6}$. Despite this, Fig. S6 generally demonstrates a consistent scaling relation between $J(t)$ and $\dot{\gamma}(t)$.

In the following section, we will concentrate on the third conclusion to further explore the intrinsic relationship between the $J(t)$ and $\dot{\gamma}(t)$.

B. $J(t)$ and $\dot{\gamma}(t)$ in micro-rheology. To elucidate this phenomenon, we used Equation (9) to correlate the variance in position ($\mathbb{V}[x(t)]$) and creep compliance ($\mathcal{J}(t)$) from reference (8), which is presented as follows:

$$\mathbb{V}[x(t)] = \frac{k_BT}{\pi r}\mathcal{J}(t) = \frac{k_BT}{\pi r\sigma}\gamma(t) \qquad \text{[S-132]}$$

where r represents the radius of the particles involved. By differentiating both sides for time, we derive an expression that links $J(t)$ and $\dot{\gamma}(t)$ through a scaling factor of $\frac{k_BT}{\pi r\sigma}$,

$$J(t) = \frac{k_B T}{\pi r \sigma} \dot{\gamma}(t) \quad \text{[S-133]}$$

However, the scaling between $J(t)$ and $\dot{\gamma}(t)$ obtained from reference (7) and as shown in Fig. S6 of our manuscript does not align with eq. (S-133). As indicated in reference (8), the theoretical derivations in micro-rheology suggest a scaling factor of $\frac{k_B T}{\pi r \sigma}$, indicating discrepancies attributable to the recovery-phase measurements of viscoelastic materials, which deviate from the assumed creeping flow conditions.

Interestingly, through our analysis, we observed a power-law decrease of $J(t)$ correlates with the rheometric shear rate via a scaling factor $\frac{k_B T}{\pi r}$, assuming $r = 10$nm and $T = 298$K, as specified in the experiments from reference (7). Ignoring the units for a moment, this leads us to an empirical formulation:

$$J(t) \approx \frac{k_B T}{\pi r} |\dot{\gamma}(t)| \quad \text{[S-134]}$$

This empirical relation, although not theoretically derived, offers a pragmatic approximation under the specific conditions of our experiments.Fig. S7 is corroborated by the results of the creep test performed under a lower stress of 10 Pa, indicating that the observed scaling between $J(t)$ and $\dot{\gamma}(t)$ is not affected by the magnitude of the applied creep stress.

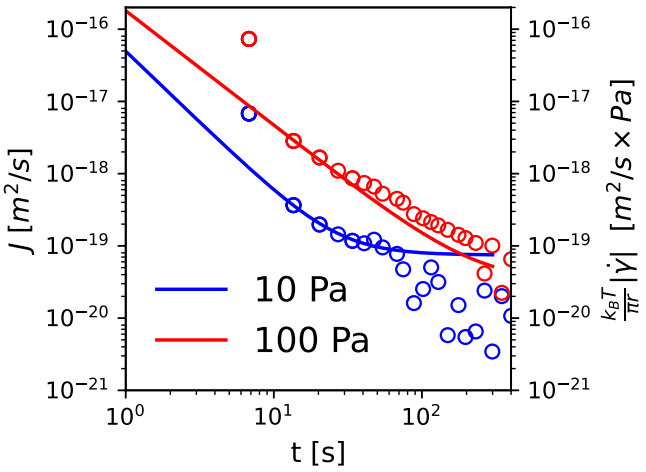

Fig. S7. The connection between microscopic dynamics ($J(t)$) and macroscopic properties ($\dot{\gamma}(t)$) in the recovery phase of creep tests with $\sigma(t)$ of 10Pa (blue) and 100Pa (red). The $J(t)$ (solid line) is derived from non-equilibrium analysis on c_2 measured by XPCS, and the $\dot{\gamma}(t)$ obtained from the rheometer, then scaled by a factor of $\frac{k_B T}{\pi r}$ (scattered dots), is measured from rheometer. For a better comparison, the unit for both $J(t)$ and $\dot{\gamma}(t)$ have been converted to International System of Units (S.I. Units)

Our ongoing research is focused on elucidating these findings and developing a theoretical framework to accurately describe them. In simulations involving phantom particles, we observed that the variations in $J(t)$ and $\dot{\gamma}(t)$ over time are decoupled; the shear rate induces affine transformations that alter the central positioning of the intensity profile $\mathbb{P}(x,t)$ without affecting the distribution breadth and, consequently, $J(t)$. A preliminary hypothesis suggests that external deformation in such systems, characterized by complex potentials, can induce systematic forces, including frictional and interparticle forces, thereby influencing transport dynamics. We are actively working to further explore and define these dynamics and to develop a comprehensive theoretical model. Our goal is to bridge microscopic $J(t)$, as determined by XPCS, with macroscopic rheological properties, such as the complex shear modulus ($G^*(\omega)$) and the stress relaxation modulus ($G_r(t)$), using the principles of microrheology discussed in (9).

6. Microscopically Heterogeneous Dynamics

In reference (7), the microscopically heterogeneous dynamics is quantified using the ratio of the recovery to quiescence phases in c_2, denoted as $c_{\text{ratio}}\,(t_1, t_2 = 0)$ in equation [3] from their paper.

Our methodology introduces an alternative approach for assessing the microscopically heterogeneous dynamics. As delineated in the derivations eqs. (S-4) and (S-5), our model assumes random, homogeneous, and independent particle dynamics. This assumption leads to the simplification where terms involving $n \neq m$ average to zero, yielding our non-equilibrium model. Given this foundation, the correlations we analyzed pertain mainly to diffusive and nonaffine dynamics, denoted $c_{2,\text{Homo}}$, and do not directly correspond to the microscopically heterogeneous dynamics described in the paper (7).

However, our current work has devised a method to discern such complex dynamics. To elucidate the contributions from heterogeneous dynamics ($c_{2,\text{Hetero}}$), we employ the following expression:

$$c_{2,\text{Hetero}} \sim \mathbb{E}\left[\sum_{n=1}^{N}\sum_{m\neq n}^{N} \mathrm{e}^{i\vec{q}\cdot(\vec{r}_n(t_2)-\vec{r}_m(t_1))}\right] = c_{2,\text{exp}} - c_{2,\text{Homo}} \quad \text{[S-135]}$$

This approach involves subtracting the component of homogeneous dynamics from the total experimental correlation, thus isolating the residuals that reflect heterogeneous dynamics, as demonstrated in Fig. S8.

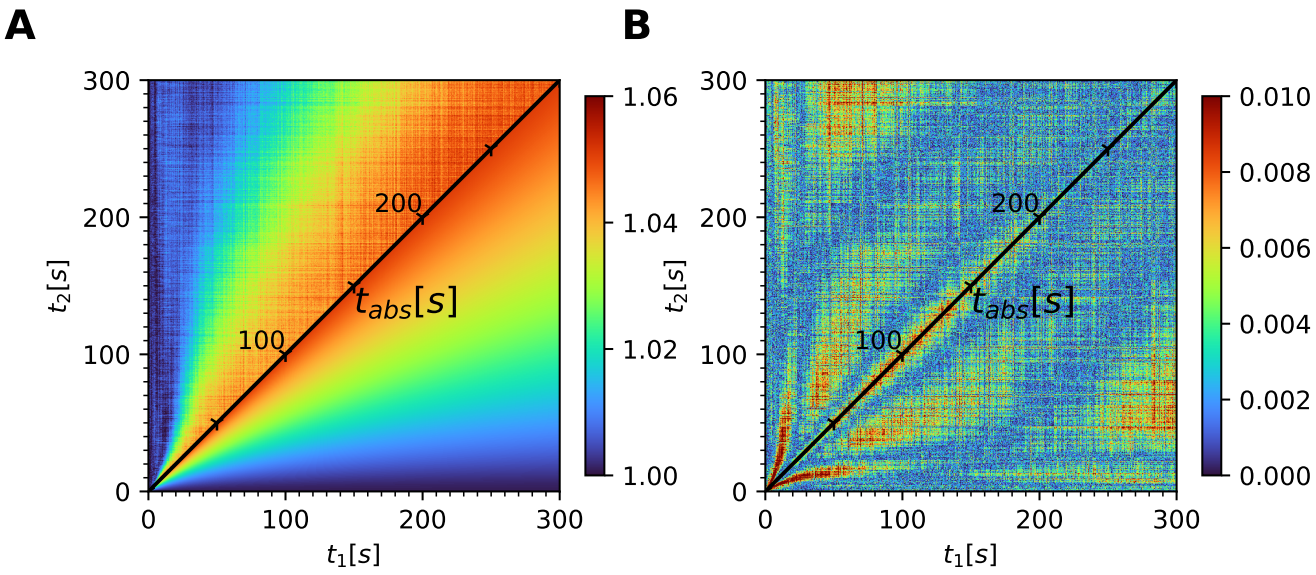

Fig. S8. Illustrating microscopically heterogeneous dynamics during recovery: By subtracting the diffusive dynamics attributed to c_2 based on the non-equilibrium model (Panel. A, lower right) from experimental c_2 measurements (Panel. A, upper left), the c_2 serves as evidence of microscopically heterogeneous dynamics in Panel B, indicating interference between at least two compositions with distinct dynamics in the system. The 'tailing' shape of the c_2 is largely attributed to the abrupt halt of a thin flow layer near the rotor during the recovery phase, mirroring the microscopically heterogeneous changes detailed in reference (7).

The temporal patterns observed in the residuals of c_2 shown in Fig. S8 suggest the presence of microscopically heterogeneous dynamics, as previously noted by (7). The pronounced 'tailing' pattern evident in S8B is indicative of shear banding, potentially resulting from the sudden stop of a thin flow layer near the rotor's surface during the recovery phase. This aligns with the heterogeneous dynamics discussed in (7). We are currently enhancing our methods to more effectively formalize these relationships, accurately describe the patterns seen in c_2, and quantify the associated heterogeneous dynamics.

7. Fitting Results and Residual for Figures in main text

A. Fitting Residual for Fig 2.

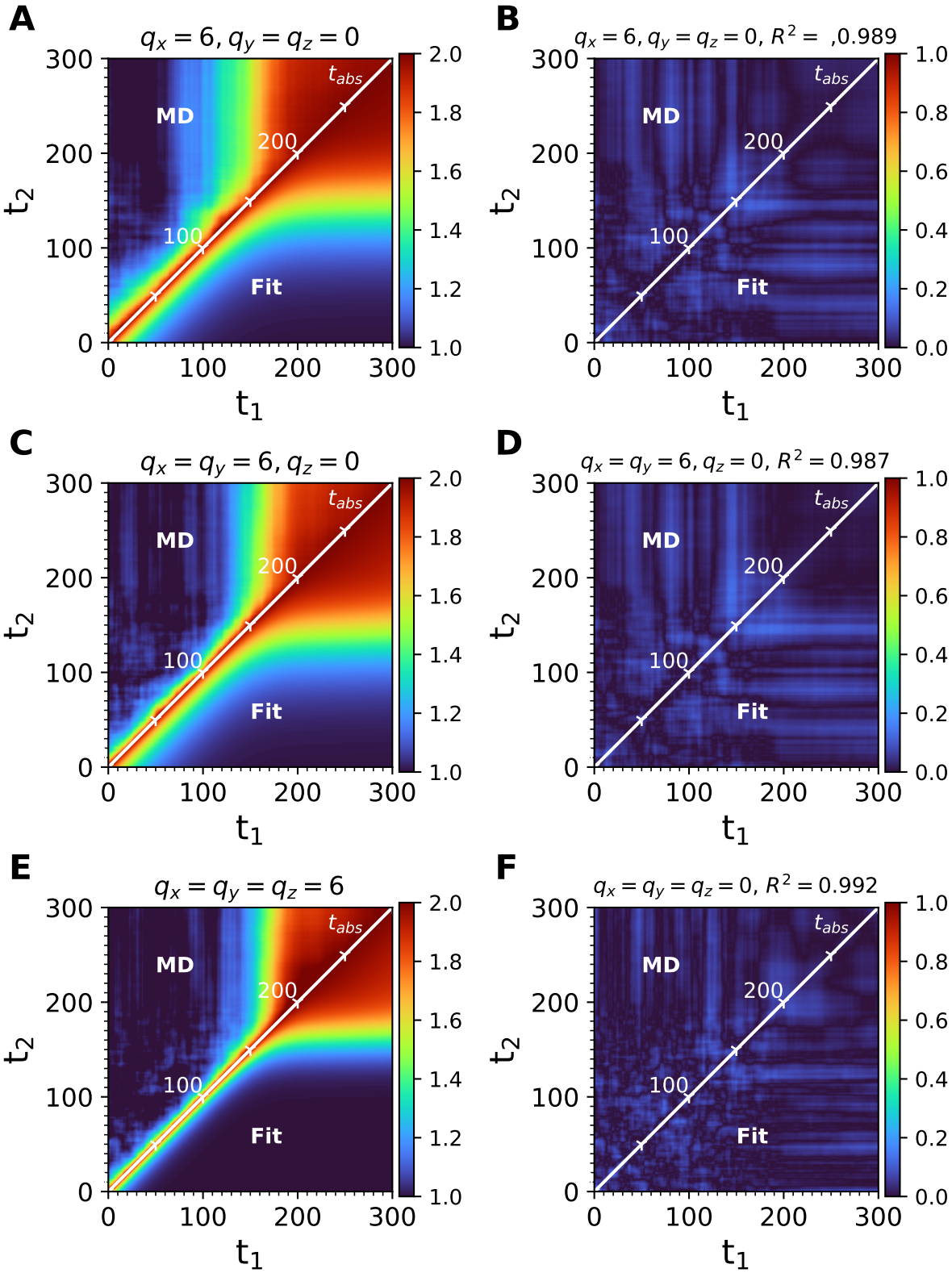

Fig. S9. illustration of the fitting results (Panels A, C, E) and residuals (Panels, B, D, F) obtained after the data fitting process shown in Fig. 2 of the main manuscript. The R^2 of the fitting is shown in the title of the residual plot.

B. Fitting Residual for Fig 3.

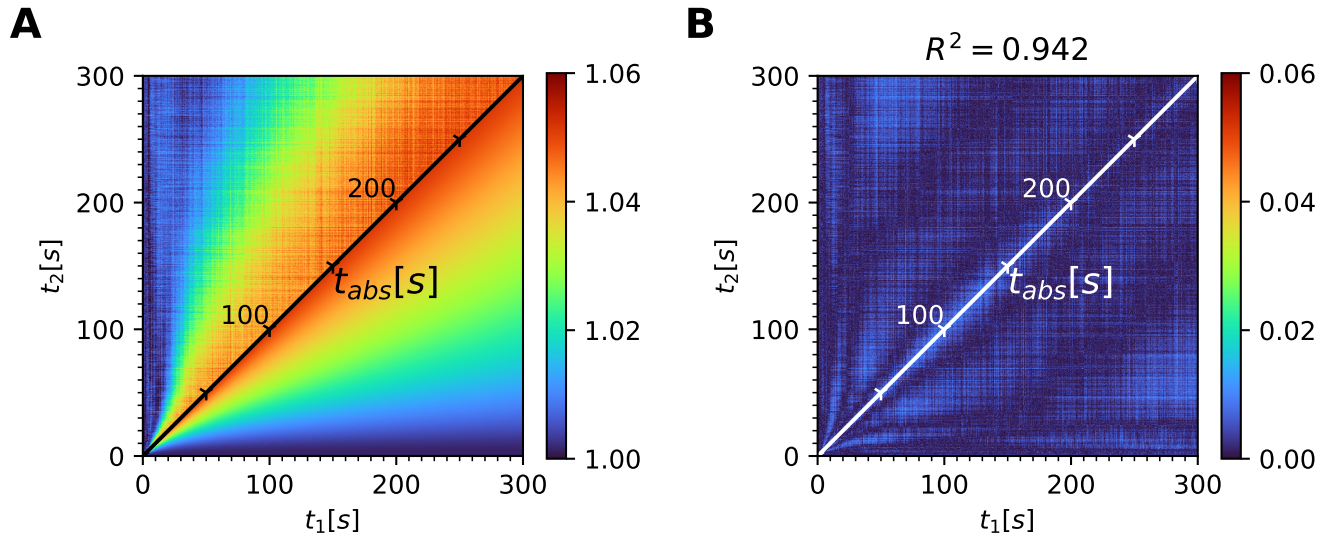

Fig. S10. Illustration of the fitting results (Panel. A) and residuals (Panel. B) obtained after the data from the (7) fitting process shown in Fig. 3A of the main manuscript. The R^2 of the fitting is shown in the title of the residual plot.

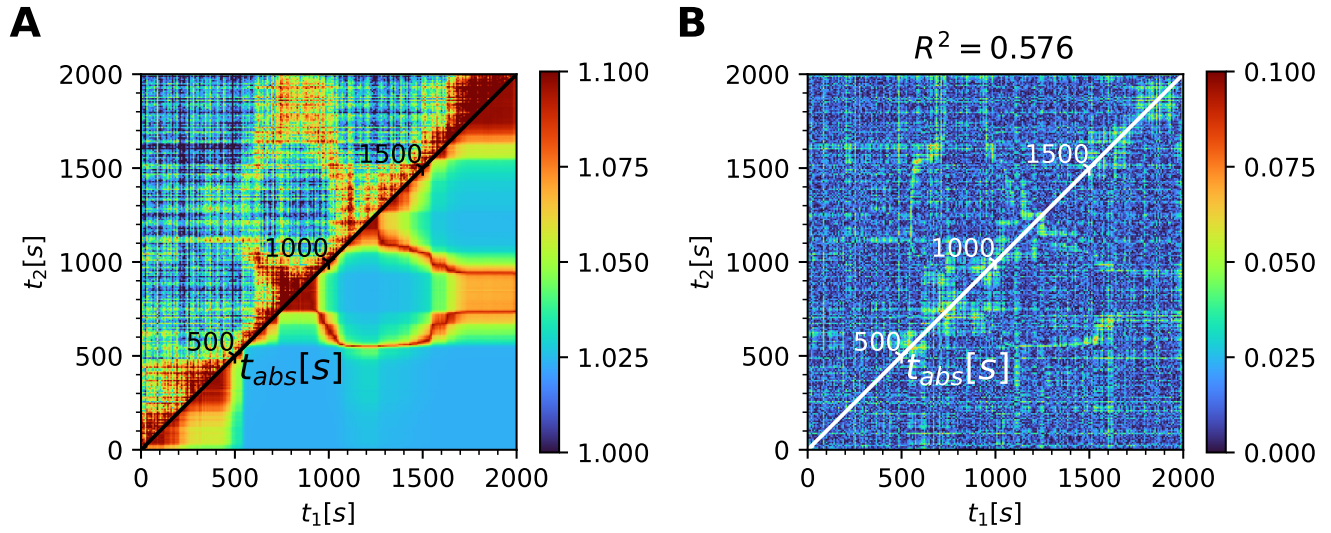

Fig. S11. Illustration of fitting results (Panel. A) and residuals (Panel. B) obtained after the data from (10) fitting process shown in Fig. 3C of the main manuscript. The R^2 of the fitting is shown in the title of the residual plot.

C. Fitting Result and Residual for Fig 4 in all ϕ directions.

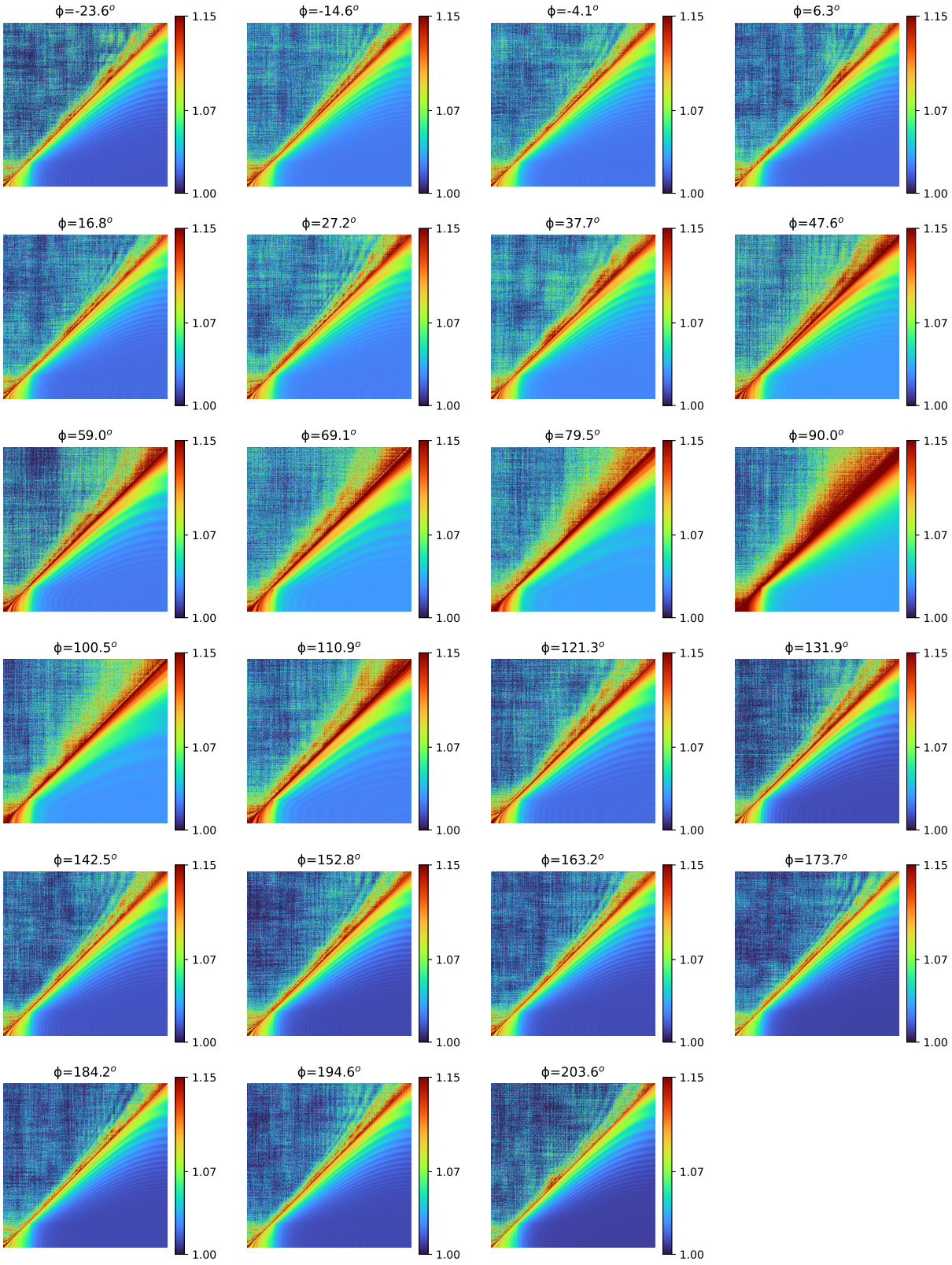

Fig. S12. Comprehensive c_2 data across all ϕ directions from Fig. 4 of the manuscript. The angle ϕ, corresponding to each bin on the field detector, is marked in the title of each panel for reference. One shown in Fig 4 of the main text is at $\phi = 6.3^o$.

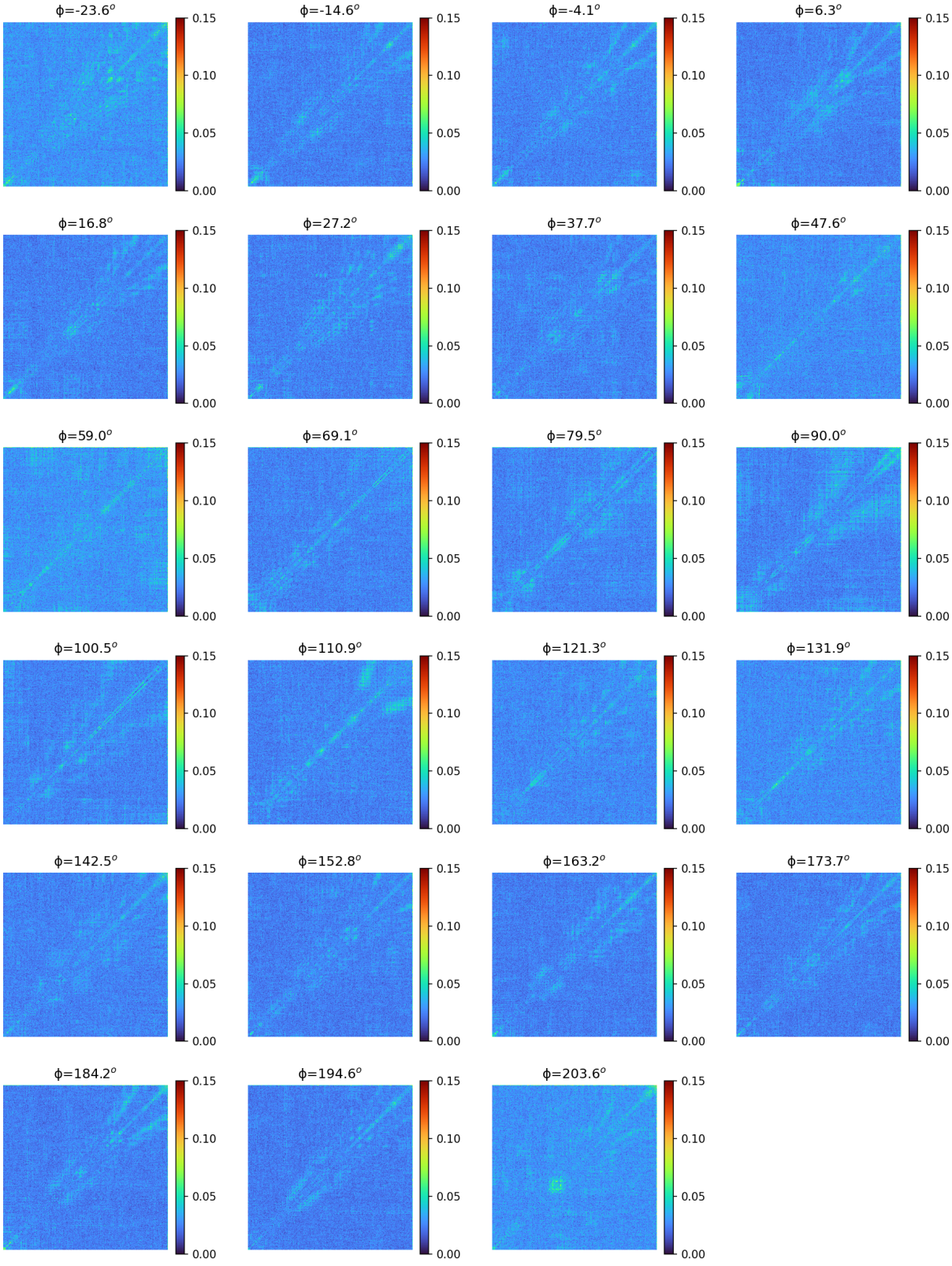

Fig. S13. Presentation of the residuals resulting from the analysis conducted in Fig. 4 of the manuscript, encompassing all ϕ directions.The angle ϕ, corresponding to each bin on the field detector, is marked in the title of each panel for reference.

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