### Simulation of Stochastic Non-Equilibrium Thermal Effects of Particle Inclusions within Fluid Interfaces and Membranes

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We formulate theoretical modeling approaches and develop practical computational simulation methods for investigating the non-equilibrium statistical mechanics of fluid interfaces with passive and active immersed particles. Our approaches capture phenomena taking into account thermal and dissipative energy exchanges, hydrodynamic coupling, and correlated spontaneous fluctuations. Our methods allow for modeling non-uniform time-varying temperature fields, fluid momentum fields, and their impacts on particle drift-diffusion dynamics. We show how practical stochastic numerical methods can be developed for these systems by performing analysis to factor operators analytically to obtain efficient algorithms for generating the fluctuating fields. We demonstrate our methods in a few simulation studies showing how they can be used to capture particle heating of the interface and the roles of thermal gradients on hydrodynamic fluctuations. The methods developed provide modeling and simulation approaches that can be used for further investigations of non-equilibrium phenomena in active soft materials, complex fluids, and biophysical systems.

### Introduction

Many soft materials and biophysical systems exhibit phenomena involving passive and active energy exchanges in regimes out of thermodynamic equilibrium. Examples include the viscoelastic responses of soft materials and complex fluids [1-5], activity in biological cell transport and mechanics [6-14], and experimental assays and processing techniques using thermal effects to manipulate colloids and other small particles [15-19]. Investigating phenomena in these systems requires taking into account energy exchanges arising from dissipative mechanisms in the mechanics, gradients in temperature, and roles played by fluctuations. This includes exchanges from dissipation during coupling in hydrodynamic flows and temperature gradients arising from particle chemical activity or external heating [13, 14, 16, 17, 20].

Capturing these effects in a tractable and consistent manner poses significant challenges for modeling and simulation. This requires stochastic spatial-temporal models accounting for both the mechanical and thermodynamic exchanges along with appropriate fluctuations. Most coarse-grained modeling and simulation approaches treat fluctuations in a limiting regime where temperatures have equilibrated to a global constant value. For soft materials and complex fluids, this includes simulation approaches such as Brownian-Stokesian Dynamics [21, 22], Coarse-grained approaches with Langevin thermostats [3, 23, 24], and continuum mechanics formulations such as Stochastic Immersed Boundary Methods (SIBMs) [14, 25, 26] and Stochastic Eulerian Lagrangian Methods (SELMs) [27, 28]. These computational simulation methods are based on statistical mechanics primarily in regimes in thermodynamic equilibrium.

For investigations of soft materials with active microstructural elements, biophysical systems. or experimental assays having external sources of energy, such as laser heating of particles, requires further theoretical modeling and simulation approaches to capture the relevant roles of thermal effects and other energy exchanges. For this purpose, we develop here theoretical formulations and practical computational methods for non-equilibrium thermodynamic regimes. We build on our past work on equilibrium and non-equilibrium statistical mechanics and on Stochastic Eulerian Lagrangian Methods (SELMs) [14, 26–30]. As a specific case to demonstrate these approaches. we focus on the context of fluid interfaces / membranes having particle inclusions. We formulate our models to account for non-uniform temperature fields, hydrodynamic coupling, heat exchanges between the particles and fluid, and the roles of fluctuations in the hydrodynamics and particle drift-diffusion dynamics. We develop SELM approaches for non-equilibrium regimes for investigating fluid interfaces with active and passive particles. Part of the motivations for this work include the roles of activity of proteins in cellular processes [12, 20, 31–35], active inclusions within lipid bilayer membranes [14, 36–38], recent experimental systems that use lasers to thermally excite and manipulate particles [15–19], and other modeling and simulation challenges for active processes in soft materials and biophysical systems [6, 39-41].

### Materials and methods

#### Stochastic Non-Equilibrium Model for Thermal Effects and Fluctuations

We first formulate our theoretical modeling approach for capturing the mechanics and thermal effects within fluid-particle systems. We then discuss how practical stochastic numerical methods can be developed for performing simulations. We then perform a few simulations of specific models to demonstrate the approaches.

We model the system using the following stochastic fluctuating hydrodynamics description coupled to a collection of thermodynamic energy exchange equations of the form

$$\frac{d\mathbf{X}}{dt} = \mathbf{V}, \quad m\frac{d\mathbf{V}}{dt} = -\gamma \left(\mathbf{V} - \Gamma \mathbf{u}\right) - \Xi \mathbf{V} - \nabla_{\mathbf{X}} \Phi(\mathbf{X}) + \mathbf{G}_{thm,pp} \tag{1}$$

$$\rho \frac{\partial \mathbf{u}}{\partial t} = \nabla \cdot \boldsymbol{\sigma} + \boldsymbol{\tau} - \nabla p + \Lambda \left[\gamma \left(\mathbf{V} - \Gamma \mathbf{u}\right)\right] + \mathbf{g}_{thm,p_F}, \quad \nabla \cdot \mathbf{u} = 0$$

$$\frac{\partial \theta_P}{\partial t} = -\frac{\kappa_{PI} \left(\theta_P - \theta_I\right)}{c_P} + \frac{\dot{q}_P^T D_P(Y) \dot{q}_P}{c_P} + \mathbf{G}_{thm,\theta_P} \tag{2}$$

$$\frac{\partial \theta_F(x,t)}{\partial t} = \frac{\nabla \cdot \left(\kappa_{FF} \nabla \theta_F(x)\right)}{c_F} - \frac{\kappa_{FI}(x;X) \left(\theta_F - \theta_I\right)}{c_F} + \frac{\dot{q}_F^T D_F(Y) \dot{q}_F}{c_F} + \mathbf{g}_{thm,\theta_F}(x,t)$$

$$\frac{\partial \theta_I}{\partial t} = \frac{\kappa_{PI} \left(\theta_P - \theta_I\right)}{c_I} + \int \frac{\kappa_{FI}(x;X) \left(\theta_F - \theta_I\right)}{c_I} dx + \frac{\dot{q}_{P,F}^T D_I(Y) \dot{q}_{P,F}}{c_I} + \mathbf{G}_{thm,\theta_I}.$$

The first part of our model employs momentum equations for the particle drift-diffusion X(t), V(t) coupled to fluctuating hydrodynamic [14, 25, 38] equations for u(x,t). For tractable computations, we treat the fluid-structure interactions using a coarse-grained interaction model having similarities to Immersed Boundary Methods (IBMs) [25, 42] and Stochastic Eulerian Lagrangian Methods

(SELMs) [27, 28]. More details are discussed below. The second part of the equations accounts for the irreversible dissipative energy exchanges within the system which generate heat and direct heat exchanges that impact the temperature fields. We model our system with three types of heat bodies (i) particles with temperature  $\theta_P$ , (ii) fluid interface with temperature field  $\theta_F(x)$ , and (iii) interfacial region involved in the fluid-structure interactions with temperature  $\theta_I$ . The  $\mathbf{G}_{thm,*}, \mathbf{g}_{thm,*}$ are stochastic terms accounting for fluctuations. We denote the vector-valued terms as  $\mathbf{G}_{thm}(t)$  and the stochastic spatial fields as  $\mathbf{g}_{thm}(x, t)$ . A schematic of the model can be found in Figure 1.



Figure 1: Stochastic Non-Equilibrium Model for the Particle-Fluid Interface. The model accounts for energy exchanges and fluctuations in the particle-fluid system from reversible and irreversible processes. This includes exchanges between the particle and fluid for non-uniform temperature fields, hydrodynamic flows, and other dissipative mechanisms. The fluid-particle coupling is handled through tractable approximations to the fluid-structure interactions based on the averaging operators  $\Lambda$ ,  $\Gamma$  and for thermal exchanges through a spatially varying thermal conductivity  $\kappa_{F,I}(x; X)$ . The approaches allow for capturing phenomena in non-equilibrium regimes arising from temperature gradients, heat exchanges, and fluctuations, see equation 1

We now give some more details. For this purpose, it is convenient to collect together all of the degrees of freedom of the system by letting

$$Y = \left[\mathbf{X}, \boldsymbol{\phi}, \mathbf{R}, m\mathbf{V}, \rho \mathbf{u}, \tilde{m} \dot{\mathbf{R}}, \theta_P, \theta_F, \theta_I\right]^T = [\mathbf{q}, \mathbf{p}, \boldsymbol{\theta}]^T,$$
(3)

where  $\mathbf{q} = [q_P, q_F, q_I] = [\mathbf{X}, \boldsymbol{\phi}, \mathbf{R}]$  and  $\mathbf{p} = [p_P, p_F, p_I] = [m\mathbf{V}, \rho \mathbf{u}, \tilde{m}\dot{\mathbf{R}}]$ , and  $\boldsymbol{\theta} = [\theta_P, \theta_F, \theta_I]$ . For completeness and to allow for simplified common expressions within our derivations, we include a few additional degrees of freedom. These are  $\boldsymbol{\phi}$  for the fluid displacement map,  $\mathbf{R}$  the interface state vector,  $\tilde{m}\dot{\mathbf{R}}$  the interface pseudo-momentum. In practice, these do not play a significant role in our current model but are helpful to include so terms can be treated similarly in our derivations.

The fluid-structure interactions are treated by a drag force in the particle equations referencing the local flow environment and an equal-and-opposite body-force term in the fluid equations. For this purpose, the  $\Gamma[u] = \int \delta_a(x - \mathbf{X})u(x, t)dx$  operator is used to probe the local flow environment to determine a reference velocity at the particle location  $\mathbf{X}$ . The  $\Lambda$  operator is used to distribute forces spatially to a region of the fluid nearby to  $\mathbf{X}$ . For this purpose we use the adjoint operator  $\Lambda = \Gamma^T$  with  $\Lambda[F] = F\delta_a(x - X)$ . Many other choices can be made for the forms of  $\Gamma, \Lambda$ , we use throughout the above with  $\delta_a$  the Peskin  $\delta$ -function with  $a = \Delta x$  [14, 25, 42]. The  $\gamma$  provides the strength of the drag coupling. The  $\Xi$  is a symmetric positive semi-definite operator accounting for any friction effects treated as directly between the particles. The  $\Phi(\mathbf{X})$  provide a potential energy resulting in forces acting on the particles.

The in-plane hydrodynamic stress is denoted by  $\boldsymbol{\sigma}$  and the traction stress with the surrounding medium is denoted by  $\boldsymbol{\tau}$ . We use a Newtonian in-plane stress  $\boldsymbol{\sigma} = \mu \left( \nabla \mathbf{u} + \nabla \mathbf{u}^T \right)$  with  $\mu$  the fluid viscosity and p the fluid pressure. In this initial work, we use for simplicity the traction stress  $\boldsymbol{\tau} = -\lambda u$  with the dissipative viscosity  $\lambda$ . In our model and numerical methods, alternative in-plane and traction stresses also can be treated readily in the methods to use other hydrodynamic models and theories arising for thin films and membranes [14, 38, 43, 44].

The temperatures  $\theta_P$ ,  $\theta_F(x)$ ,  $\theta_I$  account for energy exchanges from dissipation in the mechanics and direct heat exchanges. Associated with these temperatures are three types of heat bodies. These are related to different dissipative mechanisms reflecting the rates at which the system does work in the form of friction. For the fluid-particle system these are

$$\dot{q}_P^T D_P(Y) \dot{q}_P = V^T \Xi V \tag{4}$$

$$\dot{q}_F^T D_F(Y) \dot{q}_F = \mu \nabla u : \left( \nabla u + \nabla u^T \right)$$
(5)

$$\dot{q}_{P,F}^T D_I(Y) \dot{q}_{P,F} = V^T \gamma \left( V - \Gamma u \right) - u^T \gamma \Lambda \left[ V - \Gamma u \right].$$
(6)

The  $\dot{q}$  gives the rates of change of the mechanical configuration. We distinguish the different parts of the system using the notation  $\dot{q}_F = u$ ,  $\dot{q}_P = V$ , and  $\dot{q}_{P,F} = [V, u]^T$ . We remark for simplicity and brevity in this initial work, we handle the  $-\lambda u$  by treating the coupling and  $\lambda$  as small having a negligible effect on the surrounding medium and the system.

In practice, depending on the system and regime this may require further consideration and more detailed treatment of the surrounding medium.

We briefly discuss the thermodynamics of our system associated with these energy exchanges and fluctuations. As a basic model we use the following internal energies and entropies. For the particles and interfacial region we formulate entropies as  $S^{(1)} = s_1(\theta_P) = c_P \ln(\theta_P)$ ,  $S^{(2)} = \int s_2(\theta_F) dx = \int c_F \ln(\theta_F(x)) dx$ ,  $S^{(3)} = s_3(\theta_I) = c_I \ln(\theta_I)$ . The  $c_P, c_F, c_I$  denote the specific heats. The fluid body is spatially extended and we emphasize that this is formulated using the local entropy densities  $s_2(x, \theta_F(x)) = c_F \ln(\theta_F(x))$ . This gives the system total entropy  $S = S^{(1)} + S^{(2)} + S^{(3)}$ . Similarly, we have for the particles and interfacial region internal energies  $\mathcal{U}^{(1)} = u_1(\theta_P) = c_P \theta_P$ ,  $\mathcal{U}^{(3)} = u_3(\theta_I) = c_I \theta_I$ . Since the fluid body is spatially extended, we formulate using the local internal energies  $u_2(x, \theta_F(x)) = c_F \theta_F(x)$  giving the global energy  $\mathcal{U}^{(2)} = \int u_2(x, \theta_F(x)) dx = \int c_F \theta_F(x) dx$ . This gives for the system the total energy

$$\mathcal{E} = \frac{1}{2m}p_P^2 + \Phi(q_P) + \int \frac{1}{2\rho}p_F(x)dx + \frac{1}{2\tilde{m}}p_I^2 + \mathcal{U}^{(1)} + \mathcal{U}^{(2)} + \mathcal{U}^{(3)}.$$
(7)

The terms involving the momentum  $p_P, p_F, p_I$  give the kinetic energy of the system. The  $\Phi(q_P)$  is the energy associated with the particle potential energy in equation 1.

In the context of soft materials and biophysical systems, we are often interested in phenomena in regimes having spatial-temporal scales where diffusive effects play an important role. This requires a non-trivial extension of the above formulation to account for the entropy production from dissipation and spontaneous energy exchanges from thermal fluctuations. As part of this modeling, we will use as a guideline the abstract framework of non-equilibrium statistical mechanics referred to as *GENERIC* [45, 46]. This framework generalizes Ginzsburg-Landau theory [45, 47–51] and requires that the reversible and irreversible processes be formulated in terms of an anti-symmetric operator L and a positive semi-definite symmetric operator K [45–47, 50, 52]. These act respectively on the energy gradient  $\nabla E$  and entropy gradient  $\nabla S$  to obtain dynamics that have the abstract form  $dY_t = L\nabla E dt + K\nabla S dt + k_B (\nabla \cdot K) dt + B dW_t$ . The fluctuations satisfy  $BB^T = 2k_B K$ , where  $k_B$ is the Boltzmann constant. We will also build on our prior work for equilibrium and non-equilibrium fluctuations for SELMs and other systems with uniform and non-uniform stochastic fields [25, 27, 28, 30, 53].

Given the three types of heat-bodies involved in the energy exchanges, our prior derivations shows it is convenient to treat the system using a collection of operators  $K^{(j)}$  each corresponding to one of the heat bodies. This gives dynamics of the general abstract form

$$dY_t = L\nabla E dt + \sum_j K^{(j)} \nabla S^{(j)} dt + k_B (\nabla \cdot K^{(j)}) dt + B^{(j)} dW_t^{(j)}.$$
(8)

In this formulation, the fluctuations satisfy  $B^{(j)}B^{(j),T} = 2k_B K^{(j)}$ . The  $S^{(j)}$  denotes the entropy of the  $j^{th}$  heat body. For the particle-fluid interface system this yields the stochastic dynamics incorporating both the temperature fluctuations and momentum fluctuations of the particle-fluid system.

To obtain more specific forms for the stochastic fluctuations  $\mathbf{G}_{thm}, \mathbf{g}_{thm}$ , we perform derivations below in the context of development of our stochastic numerical methods. We remark that to obtain practical simulation methods for equation 1 requires we develop numerical approaches for approximating both the differential operators and stochastic terms  $\mathbf{G}_{thm}, \mathbf{g}_{thm}$ . This can be done most conveniently by reformulating our model in equation 1 in terms of the above more abstract framework in equation 8. This requires we identify the operators  $L, K^{(j)}$  corresponding to equation 1 and perform some additional analysis. In numerical methods for the stochastic terms, we need to be able compute the stochastic terms having statistics equivalent to  $\zeta = \sqrt{2k_B}B^{(j)}\xi$  where  $\xi$  are Gaussians with  $\langle \xi\xi^T \rangle = I$ , and  $\langle \zeta\zeta^T \rangle = 2k_B B^{(j)}B^{(j),T} = 2k_B K^{(j)}$ . While in principle, Choleksy factorizations could be used for this purpose [54, 55], given that  $B^{(j)}(Y)$  depends on the state of the system this computation would be required each time-step incurring a cost of  $O(n^3)$ . The *n* is the number of rows of  $K^{(j)}$ , which can be large especially for the discretized fluid fields. For this purpose, we will derive analytically factorizations  $B^{(j)}$  for  $K^{(j)}$  allowing for more efficient generation of the stochastic driving fields for performing simulations.

### **Stochastic Numerical Methods**

We now discuss briefly the stochastic numerical methods and fluctuations in our model in equation 1. To provide a unified approach to deriving the fluctuations and our numerical discretizations, we reformulate our system in terms of stochastic dynamics of the general form

$$dY_t = L\nabla E dt + \sum_j K^{(j)} \nabla S^{(j)} dt + k_B (\nabla \cdot K^{(j)}) dt + B^{(j)} dW_t^{(j)},$$
(9)

where  $B^{(j)}B^{(j),T} = 2k_B K^{(j)}$ . This stochastic differential equation is to be given an Ito interpretation [56]. The Y denotes the state of our fluid-particle system with components

$$Y = \left[\mathbf{X}, \boldsymbol{\phi}, \mathbf{R}, m\mathbf{V}, \rho \mathbf{u}, \tilde{m} \dot{\mathbf{R}}, \theta_P, \theta_F, \theta_I\right]^T = [\mathbf{q}, \mathbf{p}, \boldsymbol{\theta}]^T.$$
(10)

This can be expressed as  $\mathbf{q} = [q_P, q_F, q_I] = [\mathbf{X}, \boldsymbol{\phi}, \mathbf{R}]$  and  $\mathbf{p} = [p_P, p_F, p_I] = [\mathbf{m}\mathbf{V}, \rho\mathbf{u}, \tilde{m}\dot{\mathbf{R}}]$ , and  $\boldsymbol{\theta} = [\theta_P, \theta_F, \theta_I]$ . For completeness and for allowing for simplified common expressions within our derivations, we include a few additional degrees of freedom. These are  $\boldsymbol{\phi}$  for the fluid displacement map,  $\mathbf{R}$  the interface state vector,  $\tilde{m}\dot{\mathbf{R}}$  the interface pseudo-momentum. In practice, these do not play a significant role in the current stochastic dynamics allowing us to avoid tracking them in our implementations, but they do provide convenience for making a common treatment of expressions in our derivations.

We start in this initial work with the basic stochastic time-step integrator

$$Y^{n+1} = Y^n + L(Y^n)\nabla E(Y^n)\Delta t + \sum_j K^{(j)}(Y^n)\nabla S^{(j)}(Y^n)\Delta t$$

$$+ \sum_j \left(\nabla \cdot K^{(j)}(Y^n)\right)\Delta t + \mathbf{g}^n.$$
(11)

This is based on the Euler-Maruyama approach [57]. The  $Y^n = Y(t_n)$ ,  $\Delta t$  is the time-step, and  $\mathbf{g}^n$  denotes the stochastic forcing with  $\langle \mathbf{g}^{n_1} \mathbf{g}^{n_2} \rangle = \delta_{n_1,n_2} 2k_B \sum_j K^{(j)} \Delta t$ . Other time-step integrators preserving additional structures also can be developed readily based on stochastic Verlet, Runge-Kutta, or Multistep schemes [57]. This initial work focuses primarily on the spatial discretizations, generation of fluctuations, and related numerical considerations.

For brevity, and to help make clear the structures that need to be preserved in our discretizations, we present at the same time our derivations for the stochastic driving fields for the fluctuations and for our numerical methods. An important underlying structure in these derivations arises from the properties of the gradient operator  $\nabla$  and divergence operator  $\nabla \cdot = \operatorname{div}$ , and their negative adjoint relationship  $\nabla \cdot = \operatorname{div} = -\nabla^T$ . As such, we retain the notation  $\nabla$  and  $\nabla \cdot$  for our operators in our derivations and in our numerical methods replace these with numerical discrete operators with  $\mathcal{G} \sim \nabla$  and  $\mathcal{D} \sim \operatorname{div} = \nabla \cdot$ , while imposing that  $\mathcal{D} = -\mathcal{G}^T$ . We give below a finite volume discretizations for  $\mathcal{D}$  and  $\mathcal{G}$  satisfying these conditions after we present the general derivations. We then use these derivations to generate our numerical methods by replacing  $\nabla$  and  $\nabla \cdot = \operatorname{div}$  with  $\mathcal{G}$ and  $\mathcal{D}$  throughout in our analytic expressions. This approach provides a systematic way to obtain discretizations and stochastic numerical methods preserving important structures of the dynamics helping to ensure they yield reliable physical results.

#### **Operators of the Reversible and Irreversible Processes**

This formulation of our dynamics uses operators that act on the energy gradient  $\nabla E$  and entropy gradient  $\nabla S$ . Relative to the continuum formulation there is a subtle but important distinction in

our numerical discretizations. We discretize our system using a finite volume perspective, where we replace spatial integrals  $\int (\cdot) dx$  by the corresponding finite sum  $\sum_m (\cdot)_{x_m} \delta V$ . We treat continuum bodies as divided into a discrete finite collection of boxes each having volume  $\delta V$ . The  $x_m$  denotes the location of the  $m^{th}$  finite volume box. We use for our total entropy gradient and energy gradients

$$\nabla \mathcal{S} = [\partial_{\mathbf{q}} \mathcal{S}, \partial_{\mathbf{p}} \mathcal{S}, \partial_{\theta} \mathcal{S}]^{T} = [0, \dots, 0, c_{P}/\theta_{P}, c_{F} \delta V/\theta_{F}, c_{I}/\theta_{I}]^{T}$$
(12)

$$\nabla \mathcal{E} = \left[\partial_{\mathbf{q}} \mathcal{E}, \partial_{\mathbf{p}} \mathcal{E}, \partial_{\theta} \mathcal{E}\right]^{T} = \left[\nabla_{\mathbf{X}} \Phi(\mathbf{X}), 0, 0, \mathbf{V}, \mathbf{u} \delta V, \dot{\mathbf{R}}, c_{P}, c_{F} \delta V, c_{I}\right]^{T}.$$
(13)

We further distinguish entropy gradient components depending on which irreversible process is being considered. For this purpose, we use the notation

$$\nabla S^{(1)} = \nabla_Y S^{(1)}(\theta_P) = [0, \dots, 0, c_P/\theta_P, 0, 0]^T$$
(14)

$$\nabla S^{(2)} = \nabla_Y S^{(2)}(\theta_F) = [0, \dots, 0, 0, c_F \delta V / \theta_F, 0]^T$$
(15)

$$\nabla S^{(3)} = \nabla_Y S^{(3)}(\theta_P, \theta_F, \theta_I) = [0, \dots, 0, c_P/\theta_P, c_F \delta V/\theta_F, c_I/\theta_I]^T.$$
(16)

The extra factor of  $\delta V$  arises in these gradients from the energy and entropy density contributions. In the energy setting the kinetic energy density now contributes in the finite volume discretization for each box as  $\frac{1}{2} \frac{p_F^2}{\rho} \delta V = \frac{1}{2} \frac{(\rho u)^2}{\rho} \delta V$ . Since our degrees of freedom are  $p_F(x_m) = \rho u(x_m)$  for the  $m^{th}$ box, we now also have an additional factor of  $\delta V$ . This similarly holds for the entropy gradients. This will also contribute to our discretizations for the operators.

Using these gradients, we next reformulate our model in terms of the operators L and  $K^{(j)}$ . The reversible parts of the dynamics can be reformulated similar to Hamiltonian mechanics to use the anti-symmetric operator

$$L = \begin{bmatrix} 0 & 0 & I & 0 \\ 0 & 0 & 0 & I/\delta V \\ -I & 0 & 0 & 0 \\ 0 & -I/\delta V & 0 & 0 \end{bmatrix}_{p_F}^{q_F}$$
(17)

For brevity in our notation for the operators, we show just the non-zero subset of the rows and columns of the operators. For L this corresponds to the degrees of freedom  $q_P, q_F, p_P, p_F$  for the fluid and particles, as defined above. The matrix representation for the operator acting for the full system  $\nabla_Y E$  can be obtained straight-forwardly by padding the other rows and columns with zeros.

We can reformulate the irreversible parts of our dynamics in terms of the operators  $K^{(j)}$ . For the dissipation in the mechanics of the particles we have

$$K^{(1)} = \begin{bmatrix} \theta_P D_P(Y) & -\frac{\theta_P D_P(Y) \dot{q}_P}{c_P} \\ -\frac{\dot{q}_P^T \theta_P D_P(Y)}{c_P} & \frac{\dot{q}_P^T \theta_P D_P(Y) \dot{q}_P}{c_P c_P} \end{bmatrix}_{\theta_P}^{p_P}.$$
(18)

where  $D_P$  is given in equation 4. This can be verified by considering the action of this operator on the gradient of the entropy  $K^{(1)}\nabla S^{(1)}$  giving the contributions to the dynamics in equation 1, and by considering the net energy exchanges in the system which from our accounting of both mechanical and internal energy requires  $K^{(1)}\nabla_{Y^{(1)}}E = 0$ . Similarly, we can reformulate our dynamics for the dissipation in the hydrodynamics to obtain the operator

$$K^{(2)} = \begin{bmatrix} -\frac{\wp \operatorname{div}(\mu\theta_F(\nabla + \nabla^T)\wp^T)}{\delta V} & \frac{\wp \operatorname{div}(\Box \mu\theta_F(\nabla u + \nabla u^T))}{c_F \delta V} \\ \frac{-\nabla u:(\mu\theta_F(\nabla + \nabla^T)\wp^T)}{c_F \delta V} & \frac{\nabla u:(\Box \mu\theta_F(\nabla u + \nabla u^T))}{c_F c_F \delta V} + \frac{-\nabla \cdot (\tilde{\kappa}_0 \nabla)}{c_F c_F \delta V} \end{bmatrix}_{\theta_F}^{p_F}.$$
(19)

We take here  $\tilde{\kappa}(\theta_F) = \kappa_0 \theta_F^2$ . The  $\Box$  in this notation indicates the location in the expression where to insert the terms on which this operator acts. The  $\wp$  denotes the projection operator associated with the incompressibility constraint on the fluid [25, 28]. We further use its adjoint  $\wp^T$  and that the operator is self-adjoint  $\wp^T = \wp$ . We also use that from the dynamics that  $\wp u = u$  throughout. We remark that in the discrete case, we would replace the integrals  $\int (\cdot) dx$  by the finite volume sums of the form  $\sum_m (\cdot)_{x_m} \delta V$ . For clarity and to avoid clutter in expressions, we retain the integral notation throughout our derivations.

For the particle-fluid interface dissipation and direct heat exchanges we have

$$K^{(3)} = \begin{bmatrix} \theta_I D_I(Y) & 0 & 0 & \frac{-\theta_I D_I(Y) \dot{q}_{P,F}}{c_I} \\ 0 & \frac{\kappa_{PI} \theta_I \theta_P}{c_{P,P}} & 0 & -\frac{\kappa_{PI} \theta_P \theta_I}{c_{P,I}} \\ 0 & 0 & \frac{\operatorname{diag}(\kappa_{FI} \delta V \theta_F \theta_I)}{c_{F,F} \delta V \delta V} & -\frac{\kappa_{FI} \delta V \theta_I \theta_F}{c_{F,I} \delta V} \\ \frac{-\dot{q}^T \theta_I D_I(Y)}{c_I} & -\frac{\kappa_{PI} \theta_I \theta_P}{c_{I,P}} & -\frac{(\kappa_{FI} \delta V \theta_I \theta_F)^T}{c_{I,F} \delta V} & \frac{\dot{q}^T \theta_I D_I(Y) \dot{q}}{c_{I,I}} + \frac{\kappa_{PI} \theta_P \theta_I + \theta_I \int \kappa_{FI} \theta_F d_I d_F}{c_{I,I}} \end{bmatrix} \theta_I$$

$$(20)$$

We use notation  $c_{ij} = c_i \cdot c_j$  for brevity. We now give the divergence  $\nabla \cdot K^{(j)}$  for each of these terms. For the particle dissipation we have

$$\nabla_{Y^{(1)}} \cdot K^{(1)} = \begin{bmatrix} -\frac{D_P(Y)\dot{q}_P}{c_P} \\ \frac{-m^{-1}\theta_P \operatorname{tr}(D_P(Y))}{c_P} + \frac{\dot{q}_P^T D_P(Y)\dot{q}_P}{c_P c_P} \end{bmatrix}_{\theta_P}^{p_P}$$
(21)

where  $Y^{(1)} = [p_P, \theta_P]^T$ . In the notation to help keep expressions compact, we only show the non-zero rows of the divergence vector. Similarly, we highlight this through  $Y^{(1)}$  showing the only non-zero terms we need to consider when taking the derivatives. This vector can then be expanded to the full set of degrees of freedom by padding with zeros in the stochastic dynamics in equation 9. For the fluid dissipation, we have the divergence

$$\nabla_{Y^{(2)}} \cdot K^{(2)} = \begin{bmatrix} \frac{\wp \operatorname{div}\left(\frac{\partial \mu \theta_F\left(\nabla u + \nabla u^T\right)}{\partial \theta_F}\right)}{c_F} \\ \frac{-\rho^{-1} \nabla \left(\left(\mu \theta_F\left(\nabla + \nabla^T\right) \wp^T\right)}{c_F}\right) + \frac{\nabla u \left(\frac{\partial \mu \theta_F\left(\nabla u + \nabla u^T\right) \wp^T}{\partial \theta_F}\right)}{c_F c_F} + \frac{\partial - \nabla \left(\left(\tilde{\kappa}(\theta_F) \nabla\right)}{\partial \theta_F}\right)}{c_F c_F} \end{bmatrix}_{\theta_F}^{p_F}.$$
 (22)

The  $Y^{(2)} = [p_F(x), \theta_F(x)]^T$ . For the divergence for the dissipation from the interfacial fluid-particle coupling and the direct heat exchanges between particles, fluid, and interface we have

$$\nabla_{Y^{(3)}} \cdot K^{(3)} = \begin{bmatrix} \frac{-D_I(Y)\dot{q}_{P,F}}{c_I} & & \\ \frac{\kappa_{PI}\theta_I}{c_P,P} - \frac{\kappa_{PI}\theta_P}{c_{P,I}} & & \\ \frac{\kappa_{FI}\delta V \theta_I}{c_F,F\delta V\delta V} - \frac{\kappa_{FI}\delta V \theta_F}{c_{F,I}\delta V} & \\ \frac{-\theta_I \operatorname{tr}(D_I(Y)M_{P,F}^{-1})}{c_I} + \frac{\dot{q}^T D_I(Y)\dot{q}}{c_Ic_I} - \frac{\kappa_{PI}\theta_I}{c_{I,P}} - \frac{\int \kappa_{FI}dx \theta_I}{c_{I,F}\delta V} + \frac{\kappa_{PI}\theta_P + \int \kappa_{FI}\theta_F dx}{c_{I,I}} \end{bmatrix}_{\theta_I}^{p_{P,F}}$$
(23)

The  $Y^{(3)} = [p_{P,F}, \theta_P, \theta_F, \theta_I]^T$  with  $p_{P,F} = [p_P, p_F]^T$  and  $M_{P,F}^{-1} = \frac{d\dot{q}_{P,F}}{dp_{P,F}}$ .

To obtain practical stochastic numerical methods requires we are able to generate the thermal fluctuations for each time-step. We now present some methods avoiding Cholesky factorizations by performing analysis to obtain directly factorizations  $B^{(j)}(Y)$  for each of the  $K^{(j)}(Y)$ .

### Methods for Generating the Thermal Fluctuations

We now briefly discuss our approach for generating the needed stochastic fields for the thermal fluctuations with correlations  $B^{(j)}$ . We will perform analysis that yields direct factorizations of  $K^{(j)}$ . We establish a few identities useful in obtaining factors. As one identity, we will use that for operators that can be decomposed into a sum of positive semi-definite operators  $C = C_1 + C_2$  can be factored using  $C_1 = R_1 R_1^T$  and  $C_2 = R_2 R_2^T$  to generate the needed noise  $\zeta$ . To obtain the covariance  $\langle \zeta \zeta^T \rangle = C$  we use for our generation procedure  $\zeta = \zeta_1 + \zeta_2$  with  $\zeta_1 = R_1 \xi_1$  and  $\zeta_2 = R_2 \xi_2$  generated independently with  $\xi_1 \sim \eta(0, I)$  and  $\xi_2 \sim \eta(0, I)$ . The  $\eta(0, I)$  denotes the distribution of vectors having standard Gaussian components yielding mean zero and covariance the identity matrix I. This gives the needed stochastic variates, since  $\langle \zeta \zeta^T \rangle = \langle \zeta_1 \zeta_1^T \rangle + \langle \zeta_2 \zeta_2^T \rangle = R_1 R_1^T + R_2 R_2^T = C_1 + C_2 = C$ , using  $\langle \zeta_1 \zeta_2^T \rangle = 0$ . We show how this and other factorization techniques can be employed to obtain practical algorithms for generating the needed fluctuations for the spatial hydrodynamic fields, particles, and thermal fields.

We generate fluctuations for the particles X(t), V(t) and their temperature  $\theta_P$  by decomposing the operator as  $K^{(1)} = RR^T$ . For this purpose, we use the factor

$$R = \begin{bmatrix} \sqrt{\theta_P} R_D(Y) \\ -\frac{\sqrt{\theta_P} \dot{\mathbf{q}}_P^T R_D(Y)}{c_P} \end{bmatrix}_{\theta_P}^{p_P}.$$
(24)

This requires we use  $R_D$  so that  $R_D R_D^T = D_P(Y)$ , where  $D_P$  is from equation 4. For the particles, this depends on the form of the operator  $D_P(Y) = \Xi$ . In the present work, we take  $\Xi = 0$ , making this trivial. More generally in the worst case a Cholesky factorization of just  $\Xi$  can be utilized, if a factorization is not readily available. The expression above gives a general procedure for leveraging knowledge of a factorization of  $\Xi$  for use in our modeling and simulation framework. We further remark that fluctuations in this case only require random variates of the same dimension as  $p_P$ and do not require independent terms for the temperature. Intuitively, this can be explained since the temperature fluctuations are from the same spontaneous mechanism of energy exchange that impacts the particle momentum fluctuations. This requires the specified correlations to ensure the energy balances.

We generate fluctuations for the hydrodynamics u(x,t) and their temperature fields  $\theta_F(x)$  by using the following factor

$$R = \begin{bmatrix} -\operatorname{div}(R_{visco}) & 0\\ \underline{-\nabla u:R_{visco}}\\ c_F & \underline{R_{heat}}\\ p_F & \theta_F \end{bmatrix} \stackrel{p_F}{\theta_F}$$
(25)

The  $R_{visco}R_{visco}^T = K_{visco}$  and  $R_{heat}R_{heat}^T = K_{heat}$ . We define the operator  $K_{visco}A = \frac{\mu\theta_F(x)}{\delta V} (A + A^T)$ , which acts on a tensor to take the symmetric part and multiply by the local temperature  $\theta_F(x)$  and viscosity  $\mu$ . We define  $K_{heat} = -\operatorname{div}(\tilde{\kappa}\nabla)$ .

For factorization of  $R_{visco}$ , it is convenient to express the operator as  $K_{visco} = \frac{\mu\theta}{\delta V} \mathcal{T}$  with  $\mathcal{T}A = A + A^T$ . To obtain  $R_{visco}$  we use the following properties of  $\mathcal{T}$ . The operator is self-adjoint, since  $\mathcal{T}A = A + A^T$  gives  $\langle \mathcal{T}A, B \rangle = \langle A + A^T, B \rangle = A : B + A^T : B$  and  $\langle A, \mathcal{T}B \rangle = \langle A, B + B^T \rangle = A : B + A : B^T = A : B + A^T : B = \langle \mathcal{T}A, B \rangle$ . Hence the operator  $\mathcal{T} = \mathcal{T}^T$  is self-adjoint and has a symmetric matrix representation. We further have that  $\mathcal{T}^2A = \mathcal{T}(A + A^T) = (A + A^T) + (A^T + A) = 2(A + A^T) = 2\mathcal{T}A$ . If we let  $R = \frac{1}{\sqrt{2}}\mathcal{T}$  then  $RR^T = \frac{1}{2}\mathcal{T}\mathcal{T}^T = \frac{2}{2}\mathcal{T} = \mathcal{T}$ . This gives the needed factorization. The  $R_{visco}A = \sqrt{\frac{1}{2}\frac{\mu\theta}{\delta V}}(A + A^T) = \sqrt{\frac{1}{2}\frac{\mu\theta}{\delta V}}\mathcal{T}A$ . This gives  $R_{visco} = \left(\sqrt{\frac{1}{2}\frac{\mu\theta}{\delta V}}\right)\mathcal{T}$ . We can readily verify that  $R_{visco}R_{visco}^T = \frac{1}{2}\frac{\mu\theta}{\delta V}\mathcal{T}^2 = \frac{\mu\theta}{\delta V}\mathcal{T} = K_{visco}$ .

For the  $R_{heat}$ , we have  $K_{heat} = -\operatorname{div}(\tilde{\kappa}\nabla)$ . This yields factorization  $R_{heat} = -\operatorname{div}(\sqrt{\tilde{\kappa}}) = (-\nabla \cdot)\sqrt{\tilde{\kappa}}I$ , where we use  $(-\nabla \cdot)^T = \nabla$  and  $R_{heat}R_{heat}^T = K_{heat}$ . We see with these terms that  $RR^T = K^{(2)}$  and we can generate the fluctuating hydrodynamic contributions to the stochastic driving fields by using  $\zeta = \sqrt{2k_B}R\xi$ .

For the fluid-particle interface, we split the matrix  $K^{(3)}$  into two decoupled parts as

$$K^{(3)} = K_1^{(3)} + K_2^{(3)}.$$

The  $K_1^{(3)}$  correspond to the dissipation from the mechanics and the  $K_2^{(3)}$  to the direct heat exchanges. We let

$$K_{1}^{(3)} = \begin{bmatrix} \theta_{I} D_{I}(Y) & 0 & 0 & \frac{-\theta_{I} D_{I}(Y)\dot{q}}{c_{I}} \\ 0 & 0 & 0 & 0 \\ \frac{-\dot{q}_{P,F}^{T}\Theta(Y)D(Y)}{c_{I}} & 0 & 0 & \frac{\dot{q}_{P,F}^{T}\Theta(Y)D(Y)\dot{q}_{P,F}}{c_{I,I}} \end{bmatrix}_{\theta_{I}}^{p_{P,F}} \theta_{I}$$

$$(26)$$

For consistency between the cases, we also show here some of the zero rows and columns. Similar to the particle case, we can decompose this into a factor of the form

$$R_{1} = \begin{bmatrix} \sqrt{\theta_{I}} R_{D}(Y) \\ 0 \\ 0 \\ -\frac{\sqrt{\theta_{I}} \dot{\mathbf{q}}_{P,F}^{T} R_{D}(Y)}{c_{I}} \end{bmatrix}_{\theta_{I}}^{p_{P,F}} \begin{pmatrix} \theta_{P} \\ \theta_{F} \\ \theta_{F} \end{pmatrix}$$
(27)

We need to derive  $R_D$  so that  $R_D R_D^T = D_I(Y)$ . This would yield  $R_1 R_1^T = K_1^{(3)}$ . In the interface case, we have the dissipative tensor  $D_I$  given by equation 4. For this purpose, we factor using

$$R_D = \sqrt{\gamma} \begin{bmatrix} I \\ -\Lambda \end{bmatrix}_{p_F}^{p_P}.$$
(28)

We remark that this only requires generating a random value that is the size of the particle degrees of freedom  $p_F$ . This arises from the strong correlations required to ensure the energy balance between the fluctuations that inter-convert between the fluid temperature fields and momentum fields.

For the second factor corresponding to the heat exchanges, we have

$$K_{2}^{(3)} = \begin{bmatrix} 0 & 0 & 0 & 0 \\ 0 & \frac{\kappa_{PI}\theta_{I}\theta_{P}}{c_{P,P}} & 0 & -\frac{\kappa_{PI}\theta_{P}\theta_{I}}{c_{P,I}} \\ 0 & 0 & \frac{\operatorname{diag}(\kappa_{FI}\delta V \theta_{F}\theta_{I})}{c_{F,F}\delta V\delta V} & -\frac{\kappa_{FI}\delta V \theta_{I}\theta_{F}}{c_{F,I}\delta V} \\ 0 & -\frac{\kappa_{PI}\theta_{I}\theta_{P}}{c_{I,P}} & -\frac{(\kappa_{FI}\delta V \theta_{I}\theta_{F})^{T}}{c_{I,F}\delta V} & \frac{\kappa_{PI}\theta_{P}\theta_{I} + \theta_{I}\int \kappa_{FI}\theta_{F}dx}{c_{I,I}} \end{bmatrix}_{\theta_{I}}^{p_{P,F}}$$

We again retained some of the zero rows and columns for comparison with the other parts of the decomposition. For the continuum fluid fields, we split this further into a decomposition of the form  $K_2^{(3)} = K_{21}^{(3)} + \int K_{22}^{(3)}(x) dx$ . In the discrete case, we would replace the integrals by the finite volume sums of the form  $\sum_m K_{22}^{(3)}(x_m) \delta V$ . For clarity and to avoid clutter in expressions, we retain the integral notation. We can factor these terms as

$$K_{21}^{(3)} = \begin{bmatrix} \frac{\kappa_{PI}\theta_{I}\theta_{P}}{c_{P,P}} & -\frac{\kappa_{PI}\theta_{P}\theta_{I}}{c_{P,I}} \\ -\frac{\kappa_{PI}\theta_{I}\theta_{P}}{c_{I,P}} & \frac{\kappa_{PI}\theta_{P}\theta_{I}dx}{c_{I,I}} \end{bmatrix}_{\theta_{I}}^{\theta_{P}} \\ = \kappa_{PI}\theta_{P}\theta_{I} \begin{bmatrix} \frac{1}{c_{P,P}}\mathbf{e}_{P}\mathbf{e}_{P}^{T} - \frac{1}{c_{P,I}}\mathbf{e}_{P}\mathbf{e}_{I}^{T} - \frac{1}{c_{I,P}}\mathbf{e}_{I}\mathbf{e}_{P}^{T} + \frac{1}{c_{I,I}}\mathbf{e}_{I}\mathbf{e}_{I}^{T} \end{bmatrix} \\ = R_{21}R_{21}^{T}.$$

$$(30)$$

The  $\mathbf{e}_I$  and  $\mathbf{e}_P$  give the standard basis vectors with all zero entries except for the components corresponding to the  $\theta_I$  and  $\theta_P$  degrees of freedom of the system. This gives the factor

$$R_{21} = \sqrt{\kappa_{PI}\theta_{I}\theta_{P}} \begin{bmatrix} \frac{1}{c_{P}}\mathbf{e}_{P} \\ -\frac{1}{c_{I}}\mathbf{e}_{I} \end{bmatrix} = \sqrt{\kappa_{PI}\theta_{I}\theta_{P}} \begin{bmatrix} \frac{1}{c_{P}} \\ -\frac{1}{c_{I}} \end{bmatrix}_{\theta_{I}}^{\theta_{P}}.$$
(31)

We also expressed this factor using our notation for non-zero entries. As we will see in our derivations below, it is useful to have flexibility between these ways of describing the system. The second factor is similar for each spatial location since  $\kappa_{FI} = \kappa_{FI}(x)$  with

$$K_{22}^{(3)} = \int \begin{bmatrix} \frac{\kappa_{FI}\theta_{I}\theta_{F(x)}}{c_{F,F}\delta V\delta V} & -\frac{\kappa_{FI}\theta_{F(x)}\theta_{I}}{c_{F,I}\delta V} \\ -\frac{\kappa_{FI}\theta_{I}\theta_{F(x)}}{c_{I,F}\delta V} & \frac{\kappa_{FI}\theta_{F(x)}\theta_{I}}{c_{I,I}} \end{bmatrix}_{\theta_{I}}^{\theta_{F}(x)} dx$$

$$= \int \kappa_{FI}\theta_{F(x)}\theta_{I} \begin{bmatrix} \frac{1}{c_{F}\delta V}\mathbf{e}_{\theta_{F}(x)} \\ -\frac{1}{c_{I}}\mathbf{e}_{\theta_{I}} \end{bmatrix} \begin{bmatrix} \frac{1}{c_{F}\delta V}\mathbf{e}_{\theta_{F}(x)} \\ -\frac{1}{c_{I}}\mathbf{e}_{\theta_{I}} \end{bmatrix}^{T} dx$$

$$= \int R_{22}(x)R_{22}^{T}(x)dx,$$

$$(32)$$

where

$$R_{22}(x) = \sqrt{\kappa_{FI}(x)\theta_I\theta_F(x)\delta V} \begin{bmatrix} \frac{1}{c_F\delta V} \mathbf{e}_{\theta_F(x)} \\ -\frac{1}{c_I} \mathbf{e}_{\theta_I} \end{bmatrix}$$
(33)

$$= \sqrt{\kappa_{FI}(x)\theta_I\theta_F(x)\delta V} \begin{bmatrix} \frac{1}{c_F\delta V} \\ -\frac{1}{c_I} \end{bmatrix}_{\theta_I}^{\theta_F(x)}$$
(34)

The fluctuations are generated using

(35)  
$$\mathbf{g}_{2} = \mathbf{g}_{21} + \int \mathbf{g}_{22}(x)dx, \quad \mathbf{g}_{21} = \sqrt{k_{B}\Delta t}R_{21}\boldsymbol{\xi}_{21}, \quad \mathbf{g}_{22}(x) = \sqrt{k_{B}\Delta t}R_{22}(x)\boldsymbol{\xi}_{22}(x).$$

As mentioned above, for brevity we express things in terms of integrals, but in practice, we replace these expressions by the finite volume sums  $\int (\cdot) dx \sim \sum_m (\cdot)_{x_m} \delta V$ . This yields the needed correlations in the fluctuations for  $K^{(3)}$ . Since the  $\xi_{ij}(x_1), \xi_{ij}(x_2)$  have zero correlation when  $x_1 \neq x_2$ . We further have

$$\begin{aligned} \langle \mathbf{g}_2 \mathbf{g}_2^T \rangle &= \langle \mathbf{g}_{21} \mathbf{g}_{21}^T \rangle + \int \langle \mathbf{g}_{22} \mathbf{g}_{22}^T \rangle dx \\ &= 2k_B R_{21} R_{21}^T \Delta t + \int 2k_B R_{22}(x) R_{22}(x)^T dx \Delta t \\ &= 2k_B K_{21}^{(j)} \Delta t + 2k_B \int K_{22}^{(j)}(x) dx \Delta t = 2k_B K_2^{(3)} \Delta t. \end{aligned}$$

This provides stochastic driving fields with the required statistics. We can combine these factors into one large *B* matrix as follows. If  $\mathbf{g} = \sqrt{2k_B\Delta t}R_1\boldsymbol{\xi}_1 + \sqrt{2k_B\Delta t}R_2\boldsymbol{\xi}_2$  then  $\mathbf{g} = \sqrt{2k_B\Delta t}[R_1|R_2][\boldsymbol{\xi}_1|\boldsymbol{\xi}_2]^T$ . We similarly use this with  $\mathbf{g}_1 = \sqrt{2k_B\Delta t}R_1\boldsymbol{\xi}_1$  to obtain the thermal force  $\mathbf{g} = \mathbf{g}_1 + \mathbf{g}_2$ .

We have now derived analytically factors  $B^{(j)}$  for each  $K^{(j)}$ . This provides our algorithms for generating the stochastic driving fields  $\mathbf{G}_{thm}(t)$  and  $\mathbf{g}_{thm}(x,t)$  in equation 1. This allows for simulating thermal effects and fluctuations arising from the temperature gradients, hydrodynamics, and particle drift-diffusion taking into account the correlations from the fluid-structure coupling and thermal exchanges of heat.

(36)

### Spatial Discretizations for the Operators

We briefly discuss how we obtain numerical approximations that preserve needed structure in the operators. For the gradient  $\nabla$  and divergence  $\nabla$ , we use discretizations based on finite volume methods with  $\mathcal{G}_{\mathbf{m}}^{(d)}\mathbf{u} = (\mathbf{u}_{\mathbf{m}+\mathbf{e}_d} - \mathbf{u}_{\mathbf{m}-\mathbf{e}_d})/2\Delta x$ . The  $\mathbf{m} = (m_1, m_2)$  gives the lattice index,  $\Delta x$  the mesh spacing, and  $\mathbf{e}_d$  is the standard basis vector in direction d. For the divergence we use  $\mathcal{D} \cdot = -\mathcal{G}^T$ . For the thermal exchanges in the fluid we discretize using directly Fourier's law for the finite volumes. In this initial work, we used for simplicity the most basic methods based on a uniform lattice and central differences. Given some of the limitations of these types of discretizations, alternatives also can be developed using finite volume methods on staggered meshes, or other spatial discretizations based on spectral or finite element methods [53, 55, 58–61]. A key property we use in our discretizations for our stochastic numerical methods, we replace in our derivations the terms  $\nabla$  with  $\mathcal{G}$  and  $\nabla \cdot = \operatorname{div}$ with  $\mathcal{D}$ . We also replace the spatial integrals  $\int (\cdot) dx$  by the finite volume sums  $\sum_m (\cdot)_{xm} \delta V$ . This provides spatial discretizations that preserve key properties of the operators. This approach provides viable stochastic numerical methods for performing simulations of the particle-fluid system incorporating fluctuations.

## Results

### **Simulation Studies**

As a demonstration of the approaches, we perform a few basic simulations showing some of the thermal effects and related phenomena that can be captured by the methods. We consider first a particle that has been externally heated, such as excitation from exposure to a laser, which then cools by transferring heat to the interface. We then consider an interface that has a spatially varying temperature gradient and simulate its impact on hydrodynamic fluctuations.

### Particle Heating of the Interface

In recent experiments, particles within fluid interfaces are manipulated using lasers and other external driving fields inducing thermal effects [15–19]. As a basic model, we consider the energy exchanges of an initially heated particle that then cools by transferring heat to the surrounding regions of the interface. This heat is further dispersed from Fourier's law laterally within the interface. We show simulation results for the temperature fields and fluctuations in Figure 2.

When the particle is hottest there is initially a rapid transfer of energy from the particle to the interface region. Heat accumulates in the interfacial region in the vicinity of the particle making the temperatures comparable. This then decreases the rate of energy transfer from the particle with the heat transferred laterally within the interface. Over time the particle and interface equilibrate toward a common spatially uniform temperature. As the fluid heats up this also results in increased fluctuations in the temperature fields. We show quantitative results for the fluid-particle system for the temperature and energy exchanges over time in Figure 3.



Figure 2: Particle Heating of the Interface. We show the simulation results for a particle that is externally driven to be at an initially higher temperature and then cools to heat the surrounding regions. The heat transfer operators are based on  $\Lambda$  operator which give the thermal exchanges near the particle location X through a spatially varying thermal conductivity  $\kappa_{F,I}(x; X)$ . The heat disperses according to Fourier's law with stochastic fluxes associated with spontaneous fluctuations.

| Parameter  | Value               | Parameter      | Value     | Parameter      | Value     |
|------------|---------------------|----------------|-----------|----------------|-----------|
| ρ          | 0.9                 | $\mu$          | 0.08      | m              | 1.1       |
| $\gamma$   | 5                   | $k_B$          | $10^{-5}$ | $\theta_P(0)$  | 1.5       |
| $\kappa_0$ | $4.2 \times 10^{6}$ | $\kappa_{P,I}$ | 130       | $\kappa_{F,I}$ | 102       |
| $c_P$      | 1.2                 | $c_F$          | 130       | $c_I$          | 1.4       |
| $n_x$      | 20                  | $n_y$          | 20        | $\Delta x$     | $10^{-1}$ |
| $\Delta t$ | $10^{-3}$           | $n_t$          | 8000      | $\delta V$     | $10^{-2}$ |

**Table 1:** Parameters. For simulations of the case when the particle heats the interface. The  $n_x, n_y$  give the number of mesh sites in each direction,  $\Delta x$  the spatial discretization,  $\Delta t$  the time-step,  $n_t$  the total number of time-steps. The other parameters are discussed in the context of the model equations 1.



**Figure 3: Particle Heating of the Interface.** We show the results of the fluid-particle system where a hot particle heats the fluid interface. The temperature of the particle and the average temperature of the fluid is shown on the (left). We also show the individual entropies of the particle, fluid, and the total entropy in the inset. The energy exchanges between the particle and the fluid are shown on the (right).

#### **Temperature Gradients and Hydrodynamic Fluctuations**

As another basic demonstration indicating some of the phenomena that can be captured with the methods, we perform simulations when the interface initially has a significant temperature gradient. We show these results in Figure 4.



**Figure 4: Hydrodynamic Fluctuations in a Temperature Gradient.** The simulation results show the coupling where regions having larger temperature exhibit larger spontaneous hydrodynamic fluctuations. These spatial variations in fluctuations drive inhomogeneous diffusivity of the particles and other statistical effects. As the temperatures equilibrate and the gradient diminishes, the spontaneous hydrodynamic fluctuations exhibit more spatial uniformity.

| Parameter  | Value               | Parameter      | Value     | Parameter      | Value     |
|------------|---------------------|----------------|-----------|----------------|-----------|
| ρ          | 0.9                 | $\mu$          | 0.08      | m              | 1.1       |
| $\gamma$   | 5                   | $k_B$          | $10^{-5}$ | $\theta_P(0)$  | 3.0       |
| $\kappa_0$ | $4.2 \times 10^{6}$ | $\kappa_{P,I}$ | 130       | $\kappa_{F,I}$ | 102       |
| $c_P$      | 1.2                 | $c_F$          | 130       | $c_I$          | 1.4       |
| $n_x$      | 20                  | $n_y$          | 20        | $\Delta x$     | $10^{-1}$ |
| $\Delta t$ | $10^{-3}$           | $n_t$          | 16,000    | $\delta V$     | $10^{-2}$ |

**Table 2:** Parameters. For simulations of the case of the temperature gradient and hydrodynamic fluctuations. The  $n_x, n_y$  give the number of mesh sites in each direction,  $\Delta x$  the spatial discretization,  $\Delta t$  the time-step,  $n_t$  the total number of time-steps. The other parameters are discussed in the context of the model equations 1.

We see that the regions having the larger temperature exhibit hydrodynamic fluctuations having larger variance as one may expect. This results in the temperature gradient creating a hydrodynamic environment for the particles that results in larger fluctuations and diffusivities in the larger temperature regions. These asymmetries can cause statistical effects in the drift-diffusion of particles, which are related to the local hydrodynamic fluctuations [25, 28]. We show quantitative results for the temperature fields and the variance  $\sigma^2(\mathbf{x})$  of the hydrodynamic fluctuations in Figure 5.

These simulation results demonstrate how the methods can be used simultaneously for thermal effects taking into account non-uniform temperature fields, fluctuations in the hydrodynamics, and



Figure 5: Hydrodynamic Fluctuations in a Temperature Gradient. We show the temperature distribution as a function of  $x_1$  and how it changes over time on the (left). From the lateral transfer of heat the temperature distribution becomes more uniform over time. We show estimates of the average variance  $\sigma^2(x_1)$  of the hydrodynamic fluctuations over the first 8,000 simulation steps as a function of  $x_1$  on the (right). The variance of the fluctuations is seen to be smaller in the regions of lower temperature.

the particle drift-diffusion dynamics. The methods can be used for further investigations in active soft materials, complex fluids, and biophysical systems.

# Conclusions

We developed theory and modeling approaches for investigating the non-equilibrium statistical mechanics of particle inclusions within fluid interfaces. Our approaches allow for taking into account the energy exchanges, hydrodynamic coupling, and correlated spontaneous fluctuations of the non-uniform temperature fields, fluid momentum fields, and the particle drift-diffusion dynamics. We developed practical numerical methods for spatially discretizing the system and for efficiently generating the stochastic driving fields yielding the correlated fluctuations. We performed analysis to obtain stochastic algorithms based on analytic factorizations of the operators. In this initial work, we presented some basic demonstrations for how the methods can be used to capture thermal effects for particles heating the interface and for how hydrodynamic fluctuations are impacted by temperature gradients. The approaches provide methods for modeling and simulation of non-equilibrium statistical mechanics of thermal effects and fluctuations in active and passive fluid-particle systems. The simulation methods can be used for further investigations in active soft materials, complex fluids, and biophysical systems.

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