Coupling a Fluctuating Fluid with Suspended Structures Part I. Particle-Continuum Hybrid

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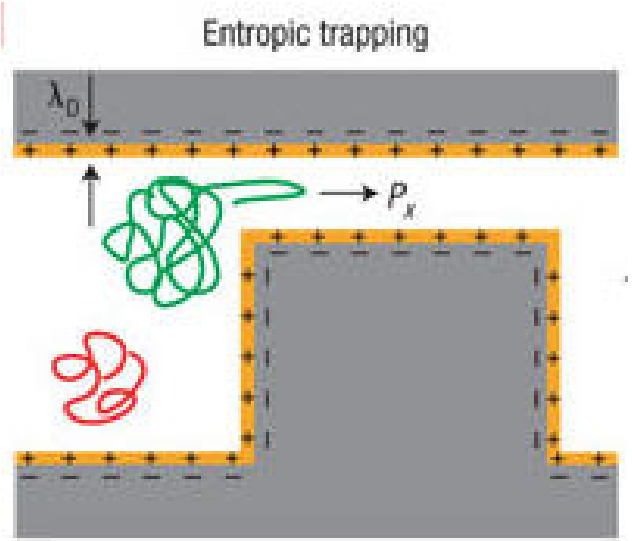
Outline

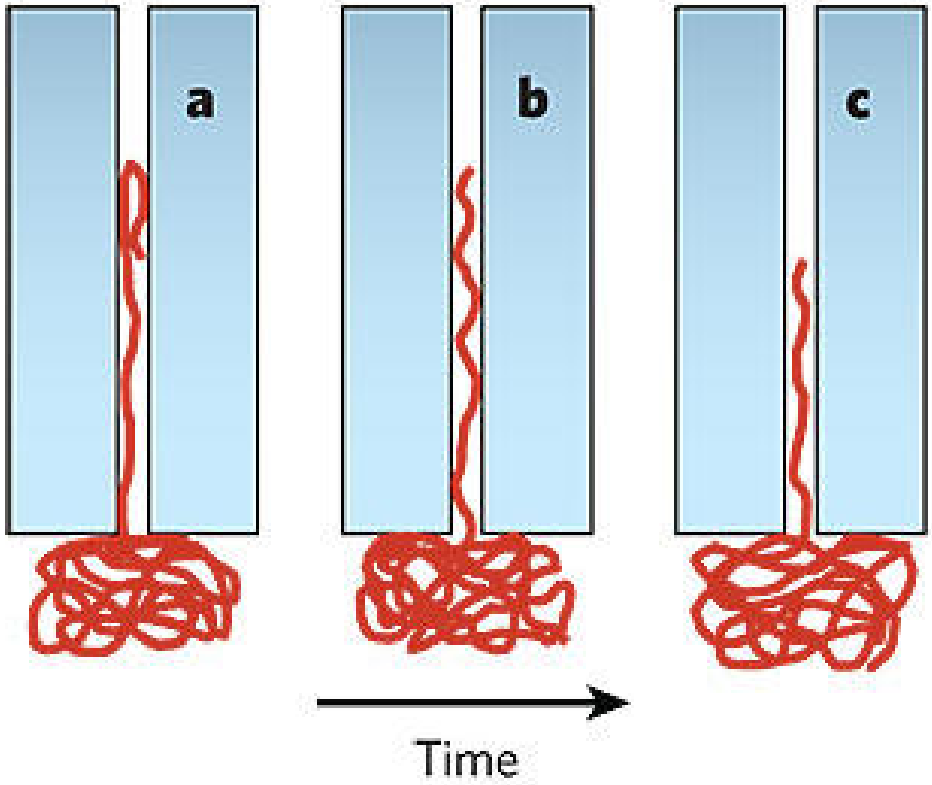
Micro- and nano-hydrodynamics

- Flows of fluids (gases and liquids) through micro- (μm) and nano-scale (nm) structures has become technologically important, e.g., **micro-fluidics, microelectromechanical systems (MEMS).**
- **Biologically-relevant** flows also occur at micro- and nano- scales.
- The flows of interest often include **suspended particles**: colloids, polymers (e.g., DNA), blood cells, bacteria: **complex fluids**.
- Essential distinguishing feature from "ordinary" CFD: **thermal fluctuations**!

Example: DNA Filtering

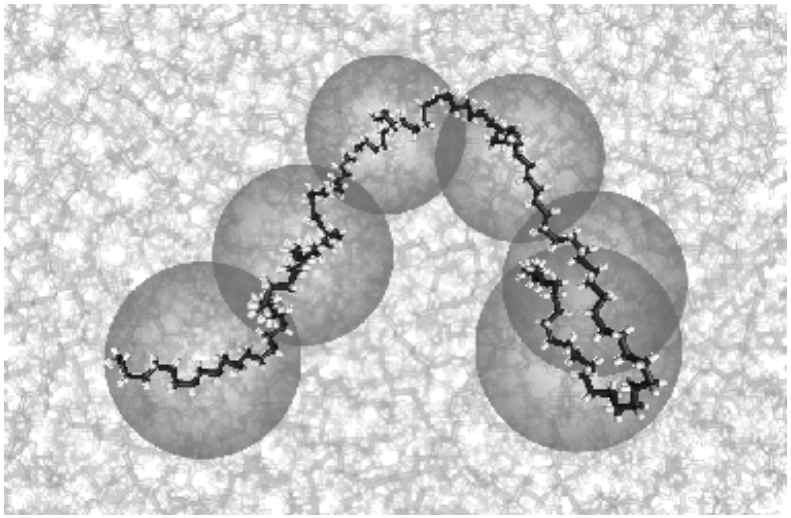


Fu et al., Nature Nanotechnology 2 (2007)



H. Craighead, Nature 442 (2006)

Polymer chains



Johan Padding, Cambridge

- Consider modeling of a polymer chain in a flowing solution, for example, DNA in a micro-array.
- The detailed structure of the polymer chain is usually **coarse-grained** to a model of spherical **beads**.
- E.g., Kuhn segments of the chain are represented as **spherical beads** connected by non-linear elastic springs (FENE, worm-like, etc.)

The issue: **How to coarse grain the fluid (solvent) and couple it to the suspended structures?**

Our approach: Particle/Continuum Hybrid

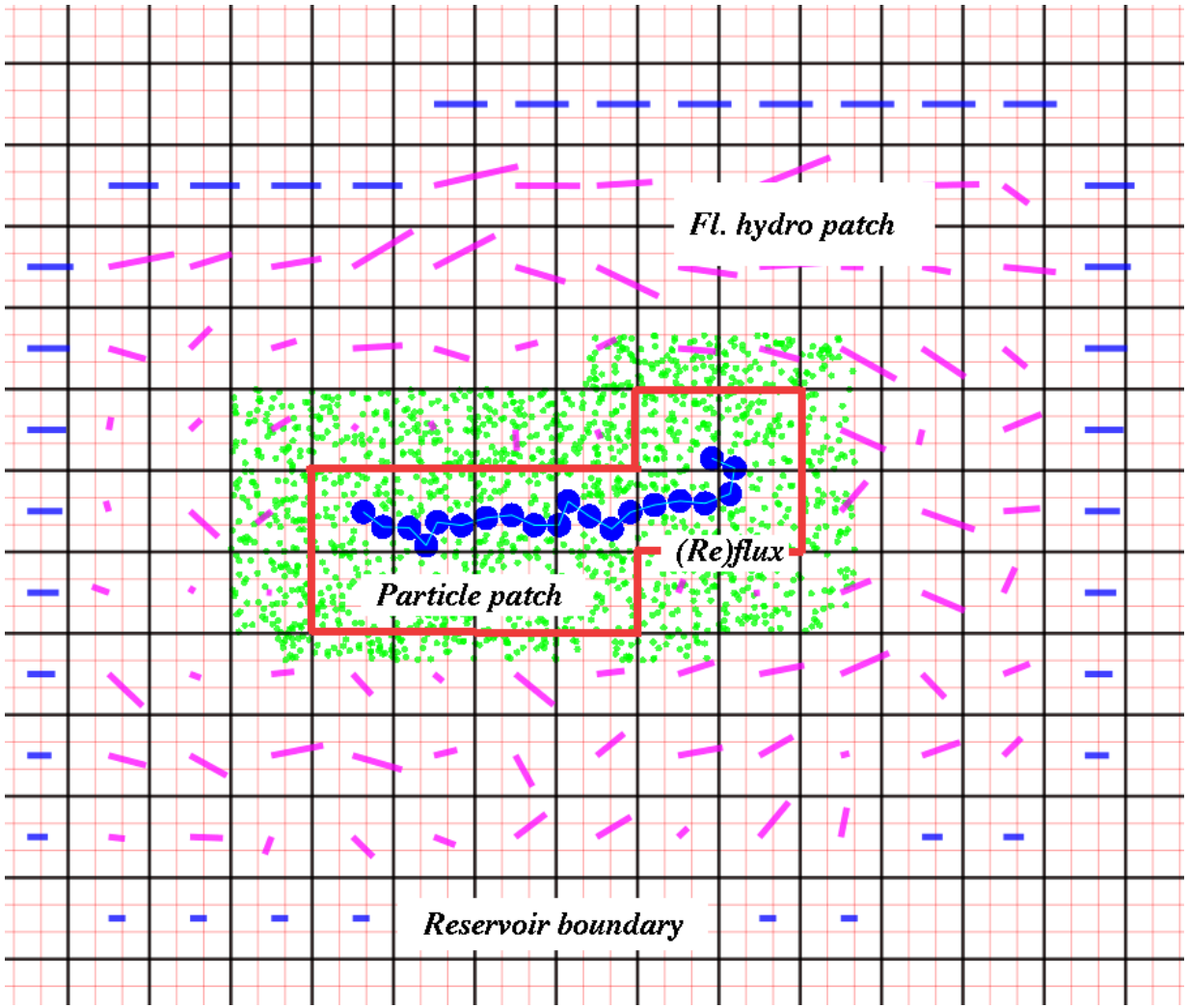


Figure: Hybrid method for a polymer chain.

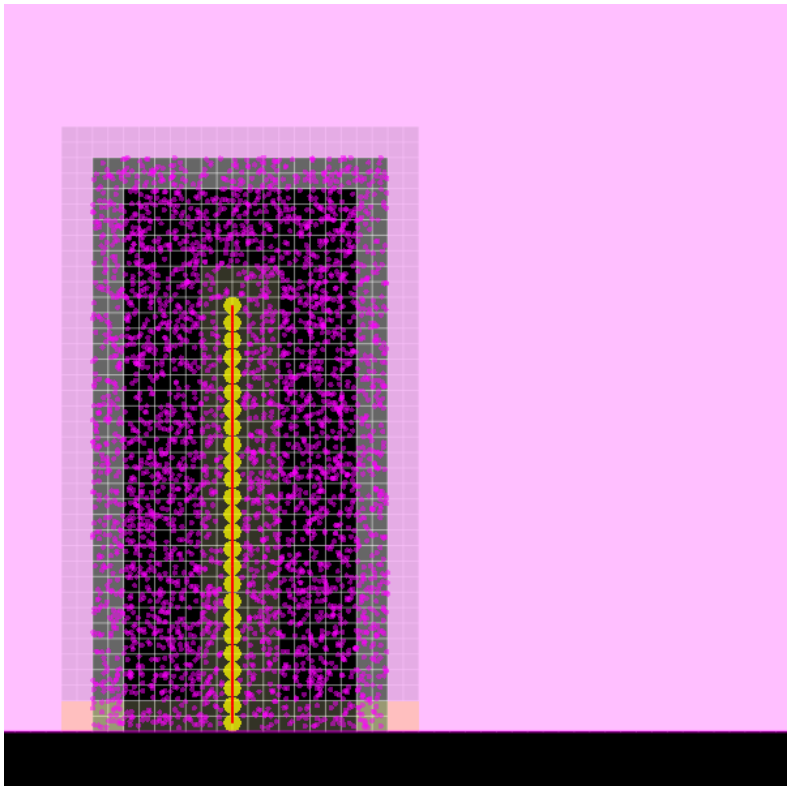
Particle Methods for Complex Fluids

- The most direct and accurate way to simulate the interaction between the **solvent** (fluid) and **solute** (beads, chain) is to use a particle scheme for both: **Molecular Dynamics (MD)**

$$m\ddot{\mathbf{r}}_i = \sum_j \mathbf{f}_{ij}(\mathbf{r}_{ij})$$

- The stiff repulsion among beads demands small time steps, and chain-chain crossings are a problem.
- Most of the computation is "wasted" on the *unimportant solvent particles*!
- Over longer times it is **hydrodynamics** (*local momentum* and energy **conservation**) and **fluctuations** (Brownian motion) that matter.
- We need to coarse grain the fluid model further: *Replace deterministic interactions with stochastic ones.*

Direct Simulation Monte Carlo (DSMC)



(MNG)

Tethered polymer chain in shear flow [1].

- **Stochastic conservative collisions** of randomly chosen nearby solvent particles, as in DSMC (also related to MPCD/SRD).
- Solute particles still interact with **both** solvent and other solute particles as hard or soft spheres [2].
- **No fluid structure**: Viscous ideal gas.
- One can introduce biased collision models to give the fluids consisten structure and a **non-ideal equation of state**. [3, 4].

The Need for Coarse-Graining

- In order to examine the time-scales involved, we focus on a fundamental problem:
  A single bead of size a and density ρ' suspended in a stationary fluid with density ρ and viscosity η (**Brownian walker**).
- By increasing the size of the bead obviously the **number of solvent particles** increases as $N \sim a^3$. But this is not the biggest problem (we have large supercomputers).
- The real issue is that a wide **separation of timescales occurs**: The *gap between the timescales of microscopic and macroscopic* processes widens as the bead becomes much bigger than the solvent particles (water molecules).
- Typical bead sizes are *nm* (nano-colloids, short polymers) or μm (colloids, DNA), while typical atomistic sizes are $1\mathring{\mathrm{A}} = 0.1nm$.

Brownian Bead

- Classical picture for the following dissipation process: *Push a sphere suspended in a liquid with initial velocity* $V_{th} \approx \sqrt{kT/M}$, $M \approx \rho' a^3$, *and watch how the velocity decays:*
  - **Sound waves** are generated from the sudden compression of the fluid and they take away a fraction of the kinetic energy during a **sonic time** $t_{sonic} \approx a/c$, where c is the (adiabatic) sound speed.
  - **Viscous dissipation** then takes over and slows the particle *non-exponentially* over a **viscous time** $t_{visc} \approx \rho a^2/\eta$, where η is the shear viscosity. Note that the classical **Langevin time** scale $t_{Lang} \approx m/\eta a$ applies only to unrealistically dense beads!
  - **Thermal fluctuations** get similarly dissipated, but their constant presence pushes the particle diffusively over a **diffusion time** $t_{diff} \approx a^2/D$, where $D \sim kT/(a\eta)$.

Timescale Estimates

- The mean collision time is $t_{coll} \approx \lambda / v_{th} \sim \eta / (\rho c^2)$, where the thermal velocity is $v_{th} \approx \sqrt{\frac{kT}{m}}$, for water

$$t_{coll} \sim 10^{-15} s = 1 fs$$

- The **sound time**

$$t_{sonic} \sim \begin{cases} 1ns \text{ for } a \sim \mu m \\ 1ps \text{ for } a \sim nm \end{cases}, \text{ with gap } \frac{t_{sonic}}{t_{coll}} \sim \frac{a}{\lambda} \sim 10^2 - 10^5$$

Estimates contd...

- **Viscous time** estimates

$$t_{visc} \sim \begin{cases} 1\mu s \text{ for } a \sim \mu m \\ 1ps \text{ for } a \sim nm \end{cases}, \text{ with gap } \frac{t_{visc}}{t_{sonic}} \sim \sqrt{C}\frac{a}{\lambda} \sim 1 - 10^3$$

- Finally, the **diffusion time** can be estimated to be

$$t_{diff} \sim \begin{cases} 1s \text{ for } a \sim \mu m \\ 1ns \text{ for } a \sim nm \end{cases}, \text{ with gap } \frac{t_{diff}}{t_{visc}} \sim \frac{a}{\phi R} \sim 10^3 - 10^6$$

which can now reach **macroscopic timescales**!

Levels of Coarse-Graining

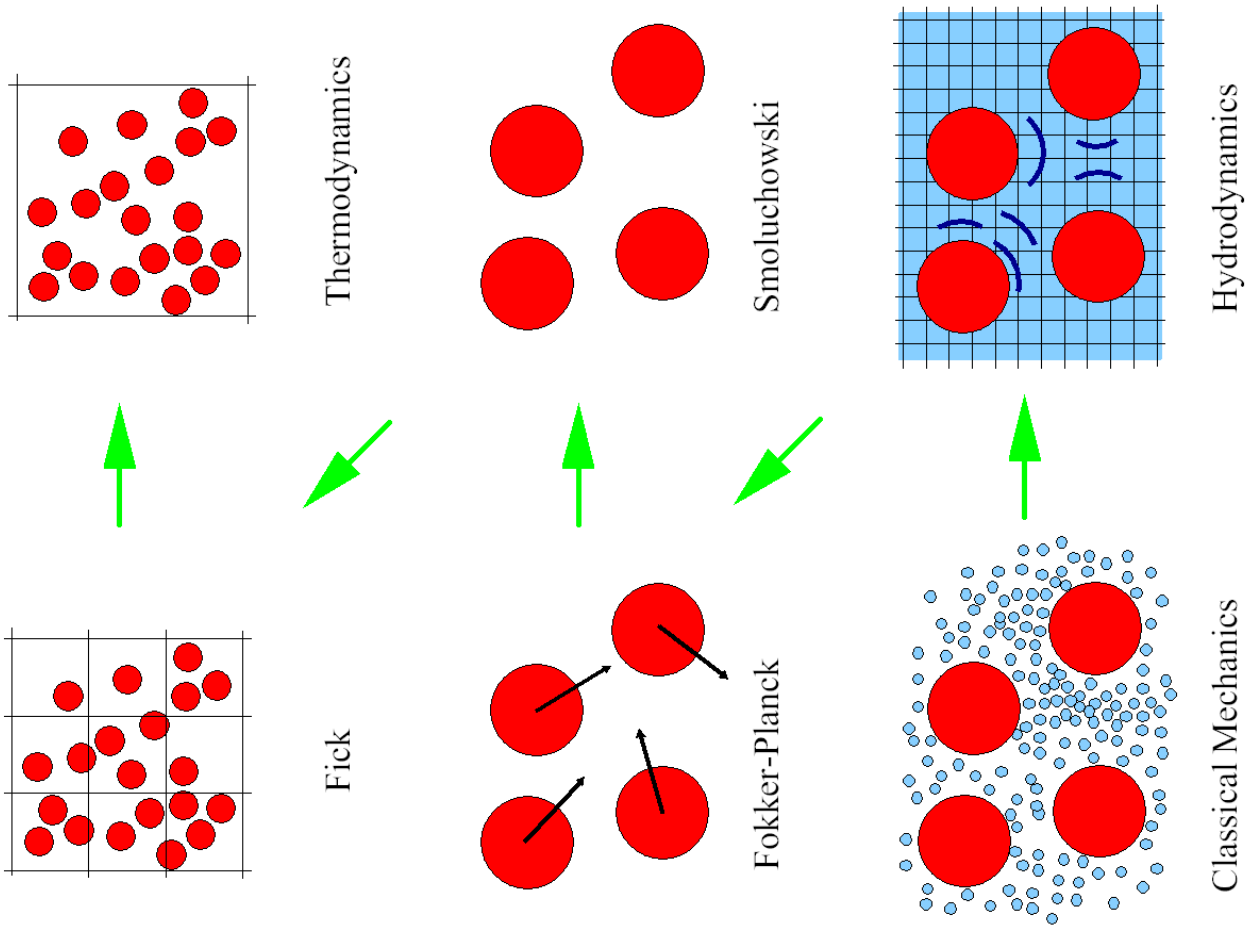


Figure: From Pep Español, "Statistical Mechanics of Coarse-Graining"

Continuum Models of Fluid Dynamics

- Formally, we consider the continuum field of **conserved quantities**

$$\mathbf{U}(\mathbf{r},t) = \begin{bmatrix} \rho \\ \mathbf{j} \\ e \end{bmatrix} \cong \widetilde{\mathbf{U}}(\mathbf{r},t) = \sum_i \begin{bmatrix} m_i \\ m_i \boldsymbol{\upsilon}_i \\ m_i \upsilon_i^2/2 \end{bmatrix} \delta\left[\mathbf{r} - \mathbf{r}_i(t)\right],$$

  where the symbol $\cong$ means that $\mathbf{U}(\mathbf{r},t)$ approximates the true atomistic configuration $\widetilde{\mathbf{U}}(\mathbf{r},t)$ over **long length and time scales**.
- Formal coarse-graining of the microscopic dynamics has been performed to derive an **approximate closure** for the macroscopic dynamics [5].
- This leads to **SPDEs of Langevin type** formed by postulating a random flux term in the usual Navier-Stokes-Fourier equations with magnitude determined from the **fluctuation-dissipation balance** condition, following Landau and Lifshitz.

The SPDEs of Fluctuating Hydrodynamics

- Due to the **microscopic conservation** of mass, momentum and energy,

$$\partial_t \mathbf{U} = -\boldsymbol{\nabla} \cdot [\mathbf{F}(\mathbf{U}) - \boldsymbol{\mathcal{Z}}] = -\boldsymbol{\nabla} \cdot [\mathbf{F}_H(\mathbf{U}) - \mathbf{F}_D(\boldsymbol{\nabla}\mathbf{U}) - \mathbf{B}\boldsymbol{\mathcal{W}}],$$

  where the flux is broken into a **hyperbolic**, **diffusive**, and a **stochastic flux**.
- Here $\boldsymbol{\mathcal{W}}$ is spatio-temporal **white noise**, i.e., a Gaussian random field with covariance

$$\langle \mathcal{W}_i(\mathbf{r}, t)\mathcal{W}_j^\star(\mathbf{r}, t')\rangle = (\delta_{ij})\,\delta(t - t')\delta(\mathbf{r} - \mathbf{r}').$$

- Adding stochastic fluxes to the **non-linear** NS equations produces **ill-behaved stochastic PDEs** (solution is too irregular), but we will ignore that for now...

Compressible Fluctuating Hydrodynamics

$$
\begin{aligned}
D_t\rho &= -\rho\boldsymbol{\nabla}\cdot\mathbf{v} \\
\rho\left(D_t\mathbf{v}\right) &= -\boldsymbol{\nabla}P + \boldsymbol{\nabla}\cdot\left(\eta\overline{\boldsymbol{\nabla}}\mathbf{v} + \boldsymbol{\Sigma}\right) \\
\rho c_p\left(D_t T\right) &= D_t P + \boldsymbol{\nabla}\cdot\left(\mu\boldsymbol{\nabla}T + \boldsymbol{\Xi}\right) + \left(\eta\overline{\boldsymbol{\nabla}}\mathbf{v} + \boldsymbol{\Sigma}\right) : \boldsymbol{\nabla}\mathbf{v},
\end{aligned}
$$

where the variables are the **density** ρ, **velocity** $\mathbf{v}$, and **temperature** T fields,

$$
\begin{aligned}
D_t\square &= \partial_t\square + \mathbf{v}\cdot\boldsymbol{\nabla}\left(\square\right) \\
\overline{\boldsymbol{\nabla}}\mathbf{v} &= \left(\boldsymbol{\nabla}\mathbf{v} + \boldsymbol{\nabla}\mathbf{v}^T\right) - 2\left(\boldsymbol{\nabla}\cdot\mathbf{v}\right)\mathbf{I}/3
\end{aligned}
$$

and capital Greek letters denote stochastic fluxes:

$$
\begin{aligned}
\boldsymbol{\Sigma} &= \sqrt{2\eta k_B T}\,\boldsymbol{\mathcal{W}}. \\
\left\langle \mathcal{W}_{ij}(\mathbf{r},t)\mathcal{W}^{\star}_{kl}(\mathbf{r}',t')\right\rangle &= \left(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk} - 2\delta_{ij}\delta_{kl}/3\right)\delta(t-t')\delta(\mathbf{r}-\mathbf{r}')
\end{aligned}
$$

Incompressible Fluctuating Navier-Stokes

- Ignoring density and temperature fluctuations, we obtain the **incompressible approximation**:

$$\rho D_t \mathbf{v} = \eta \boldsymbol{\nabla}^2 \mathbf{v} - \boldsymbol{\nabla}\pi + \sqrt{2\eta k_B T}\,(\boldsymbol{\nabla}\cdot\boldsymbol{\mathcal{W}}),$$
$$\boldsymbol{\nabla}\cdot\mathbf{v} = 0$$

  where the stochastic stress tensor $\boldsymbol{\mathcal{W}}$ is a white-noise random Gaussian tensor field with covariance

$$\langle \mathcal{W}_{ij}(\mathbf{r},t)\mathcal{W}^{\star}_{kl}(\mathbf{r}',t')\rangle = (\delta_{ik}\delta_{jl})\,\delta(t-t')\delta(\mathbf{r}-\mathbf{r}').$$

- We have algorithms and codes to solve the compressible equations, and we are now working on the incompressible ones.
- Solving them numerically requires paying attention to **discrete fluctuation-dissipation balance**, in addition to the usual deterministic difficulties [6].

Solute-Solvent Coupling using Particles

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- Split the domain into a **particle** and a **continuum (hydro) subdomains**, with timesteps $\Delta t_H = K \Delta t_P$.
- Hydro solver is a simple explicit **(fluctuating) compressible LLNS** code and is *not aware* of particle patch.
- The method is based on Adaptive Mesh and Algorithm Refinement (AMAR) methodology for conservation laws and ensures **strict conservation** of mass, momentum, *and* energy.

Continuum-Particle Coupling

- Each macro (hydro) cell is either **particle or continuum**. There is also a **reservoir region** surrounding the particle subdomain.
- The coupling is roughly of the **state-flux** form:
  - The continuum solver provides *state boundary conditions* for the particle subdomain via reservoir particles.
  - The particle subdomain provides *flux boundary conditions* for the continuum subdomain.
- The fluctuating hydro solver is **oblivious** to the particle region: Any conservative explicit finite-volume scheme can trivially be substituted.
- The coupling is greatly simplified because the particle fluid is ideal (no internal structure): **No overlap region**.

"*A hybrid particle-continuum method for hydrodynamics of complex fluids*", A. Donev and J. B. Bell and A. L. Garcia and B. J. Alder, **SIAM J. Multiscale Modeling and Simulation 8(3):871-911, 2010**

Hybrid Algorithm

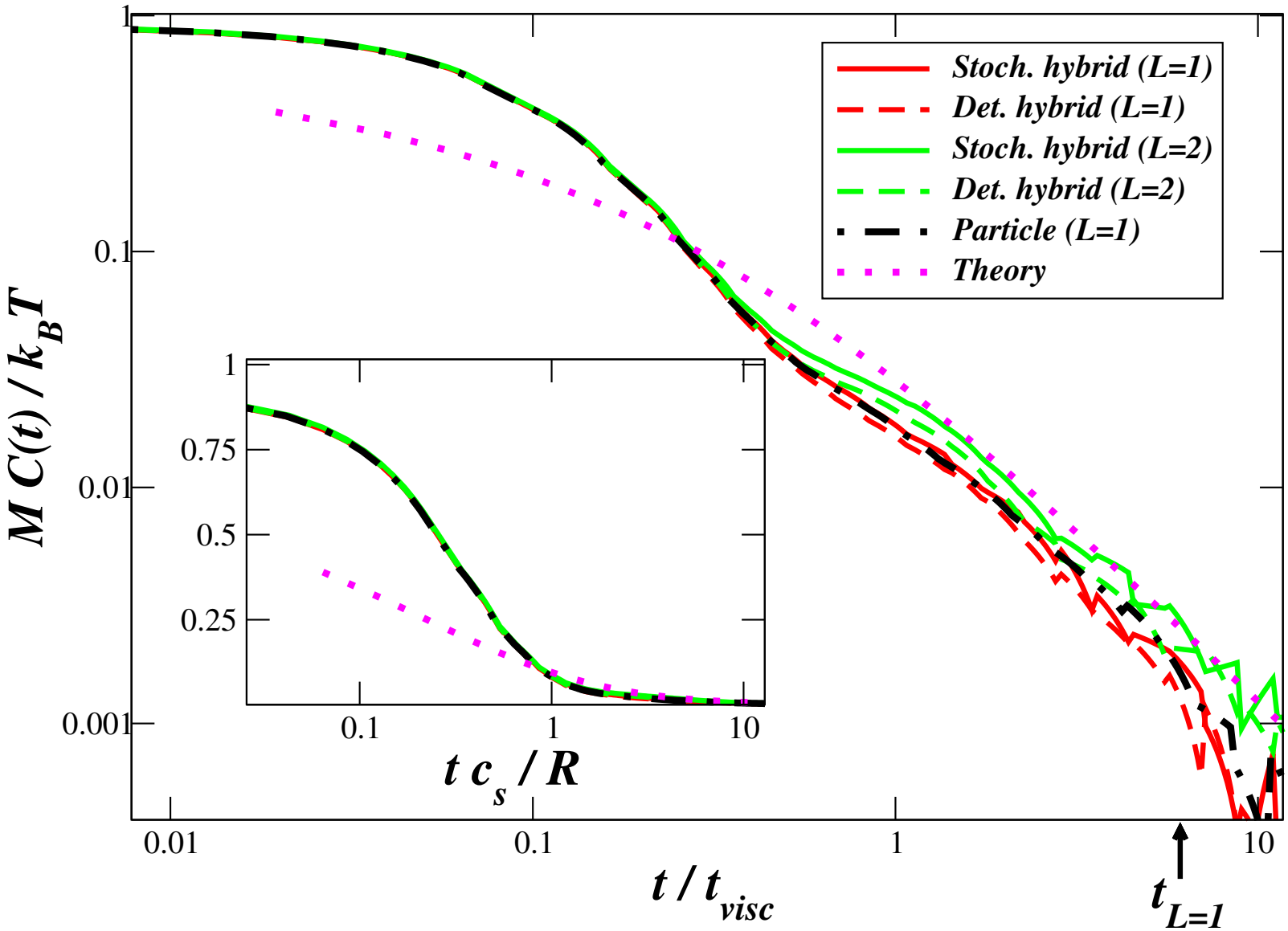
Steps of the coupling algorithm [7]:

1. The hydro solution is computed everywhere, including the **particle patch**, giving an estimated total flux $\mathbf{\Phi}_H$.
2. **Reservoir particles** are *inserted* at the boundary of the particle patch based on *Chapman-Enskog distribution* from kinetic theory, accounting for *both* collisional and kinetic viscosities.
3. Reservoir particles are *propagated* by Δt and *collisions* are processed (including virtual particles!), giving the total particle flux $\mathbf{\Phi}_p$.
4. The hydro solution is overwritten in the particle patch based on the particle state $\mathbf{u}_p$.
5. The hydro solution is corrected based on the more accurate flux, $\mathbf{u}_H \leftarrow \mathbf{u}_H - \mathbf{\Phi}_H + \mathbf{\Phi}_p$.

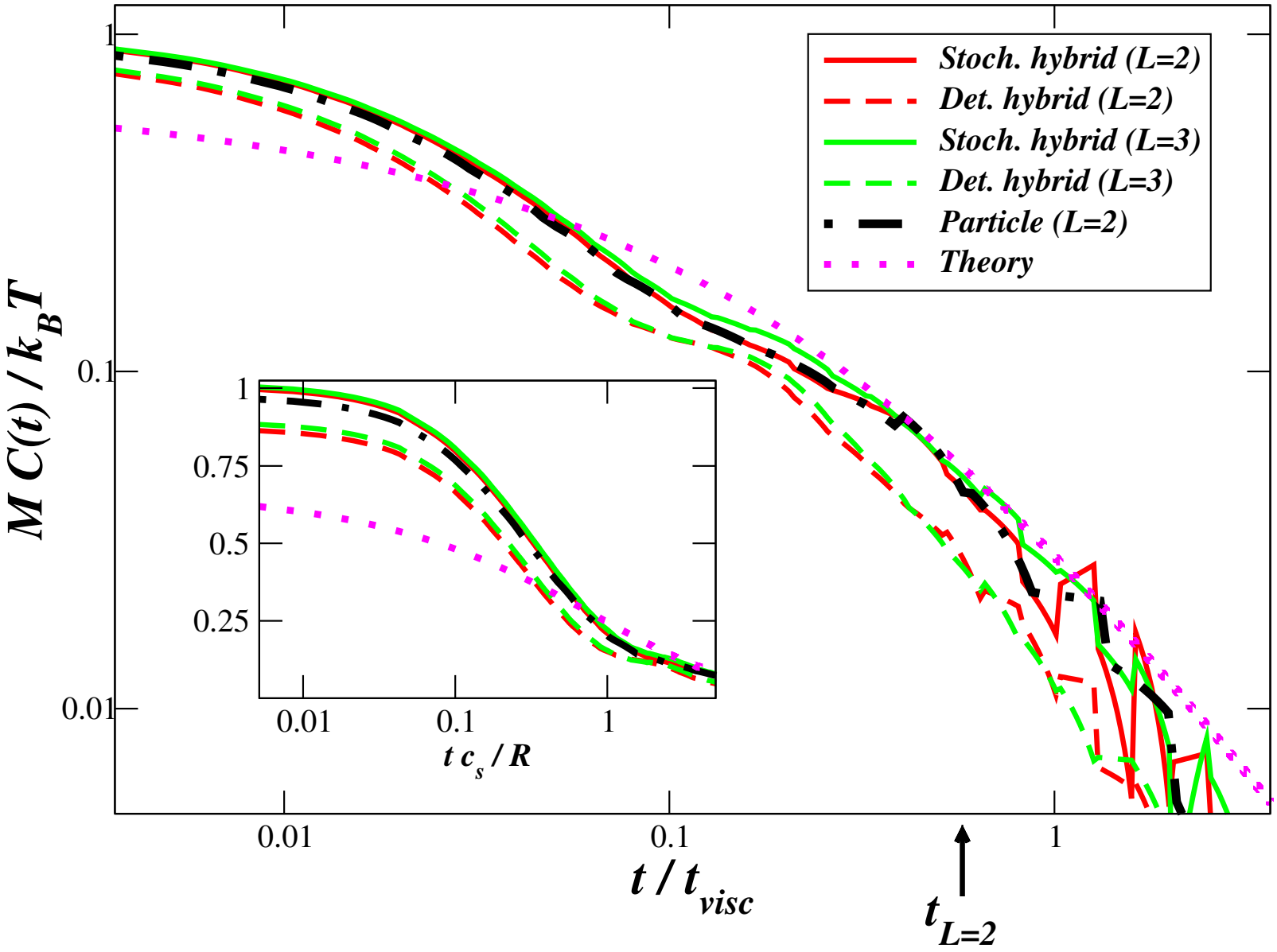
Velocity Autocorrelation Function

- We investigate the **velocity autocorrelation function** (VACF) for a Brownian bead
$$C(t) = \langle \mathbf{v}(t_0) \cdot \mathbf{v}(t_0 + t) \rangle$$
- From equipartition theorem $C(0) = kT/M$.
- For a **neutrally-boyant** particle, $\rho' = \rho$, incompressible hydrodynamic theory gives $C(0) = 2kT/3M$ because the momentum correlations decay instantly due to sound waves.
- Hydrodynamic persistence (conservation) gives a **long-time power-law tail** $C(t) \sim (kT/M)(t/t_{visc})^{-3/2}$ *not* reproduced in Brownian dynamics.

Small Bead (˜10 particles)



Large Bead (~1000 particles)



The adiabatic piston problem

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Relaxation Toward Equilibrium

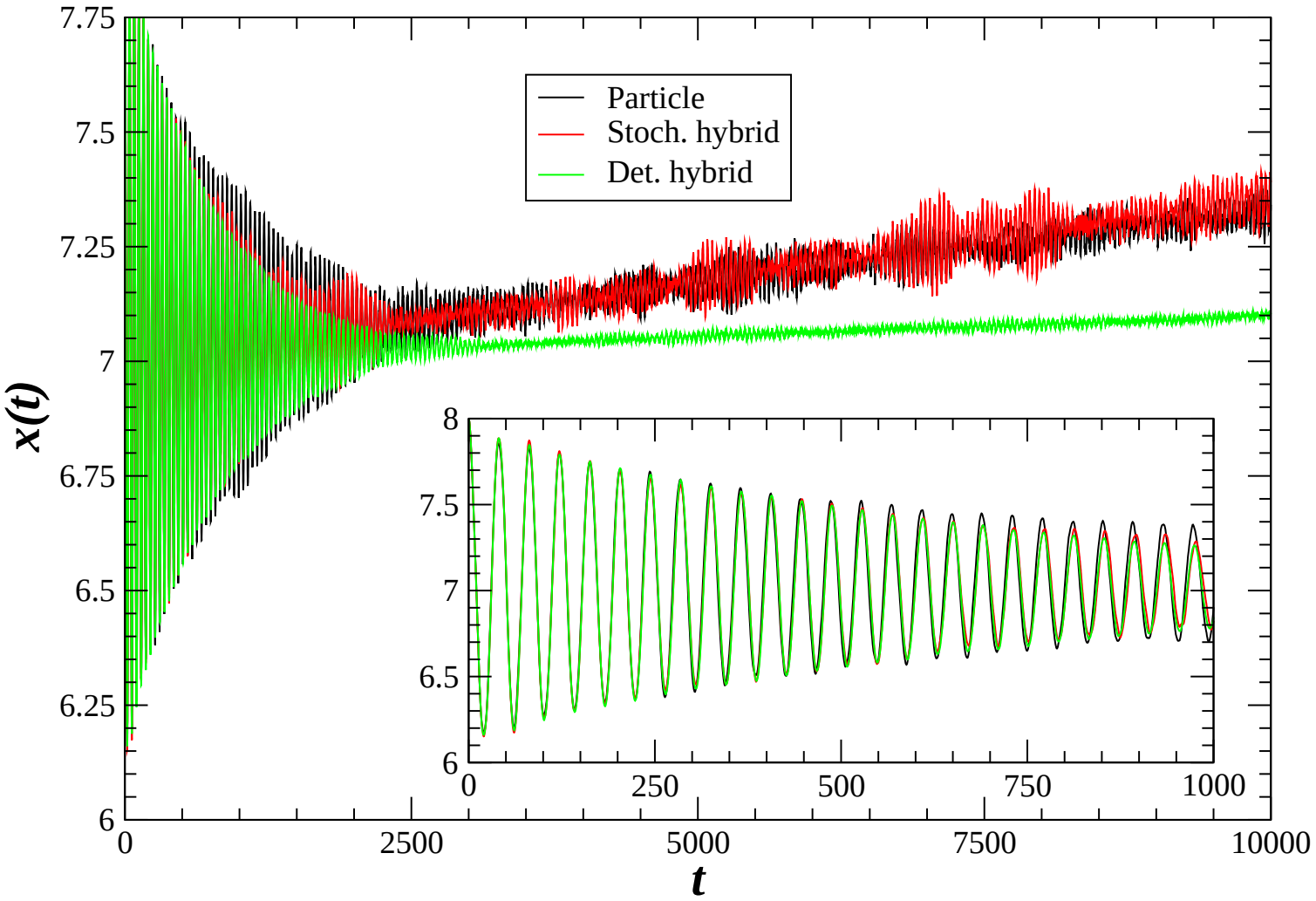

Figure: Massive rigid piston ($M/m = 4000$) not in mechanical equilibrium: **The deterministic hybrid gives the wrong answer!**

VACF for Piston

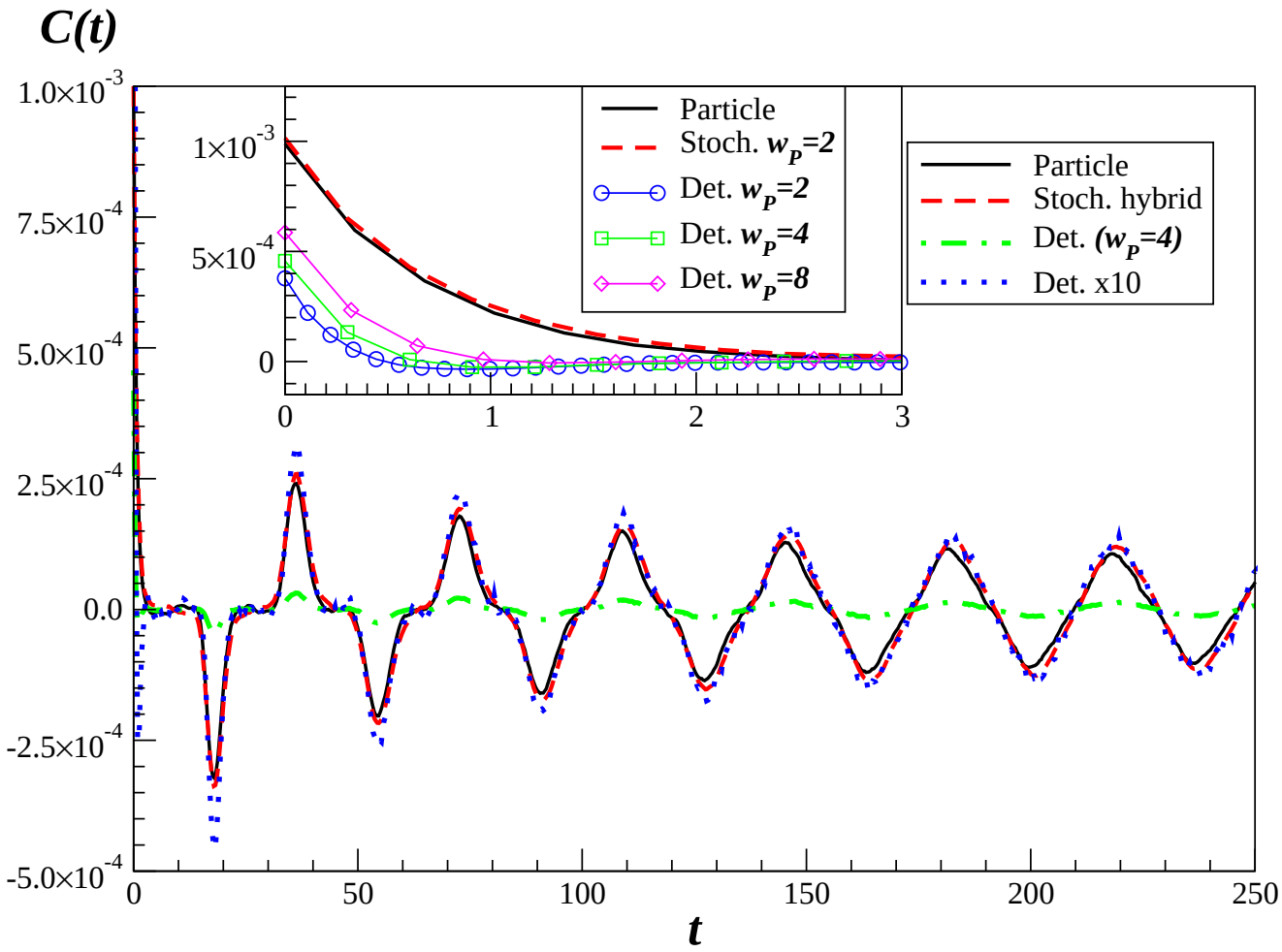

Figure: The VACF for a rigid piston of mas $M/m = 1000$ at thermal equilibrium: **Increasing the width of the particle region does not help: One must include the thermal fluctuations in the continuum solver**!

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