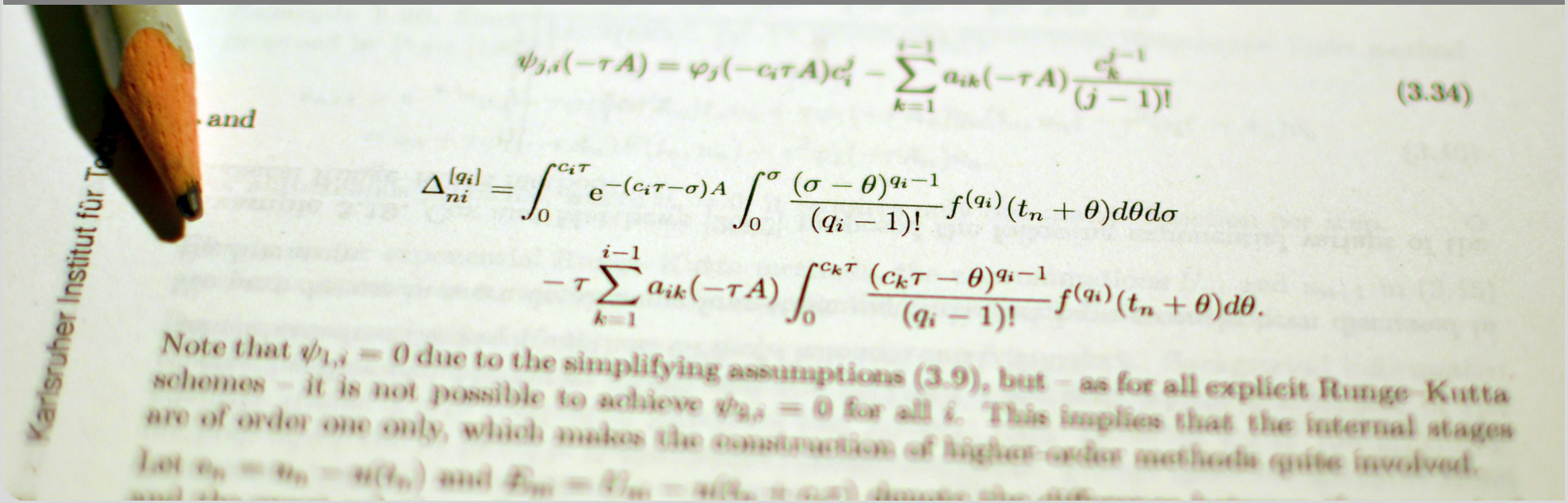
Karlsruhe Institute of Technology

A Fast Mollified Impulse Method for Flexible Water Simulations

Lukas Fath | Oct 6, 2015

INSTITUTE FOR APPLIED AND NUMERICAL MATHEMATICS 1 | MARLIS HOCHBRUCK



A Problem from Nanotribology

Ice-Ice Friction with MD

- phase transition, premelting, heat transport
- transition static to sliding friction
- multi scale phenomena

in cooperation with CME (University of Toronto) and INT (KIT)

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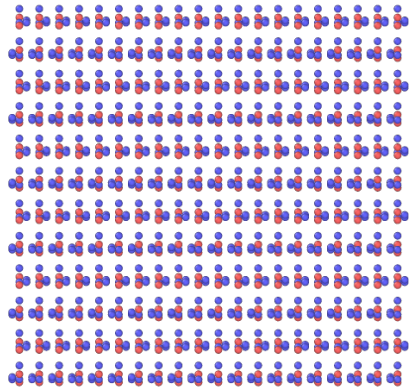

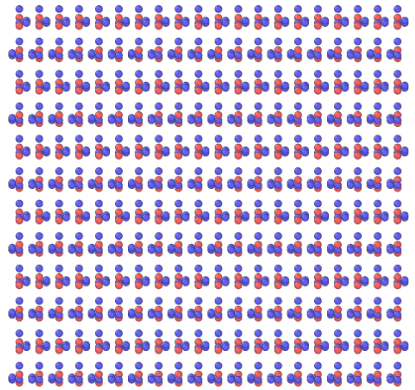

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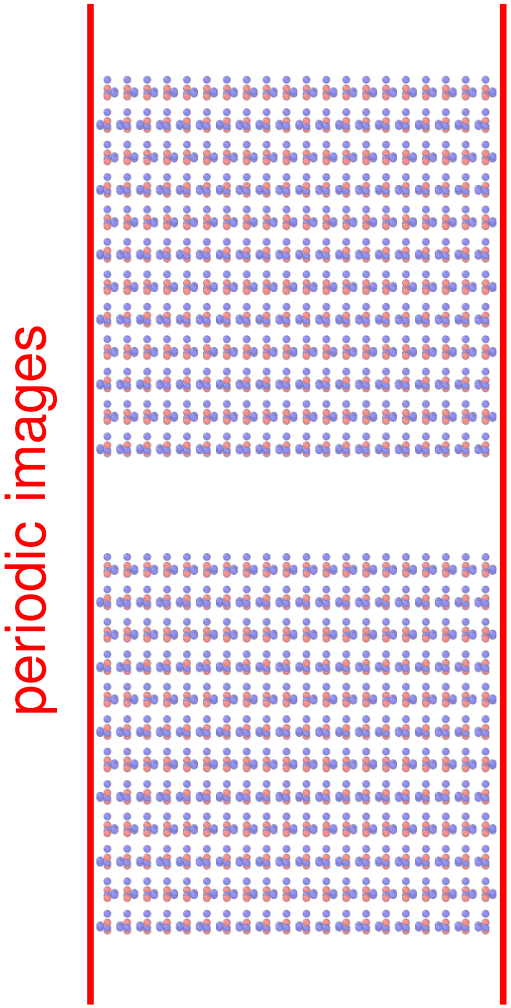

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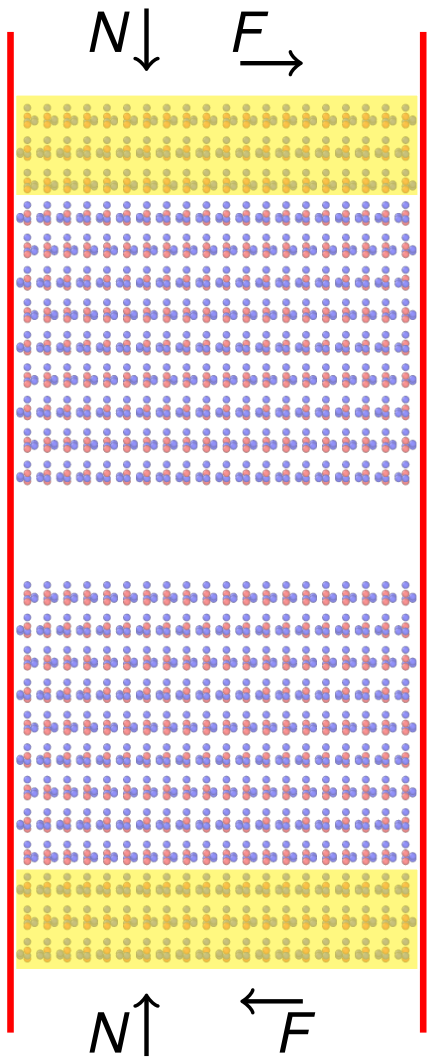

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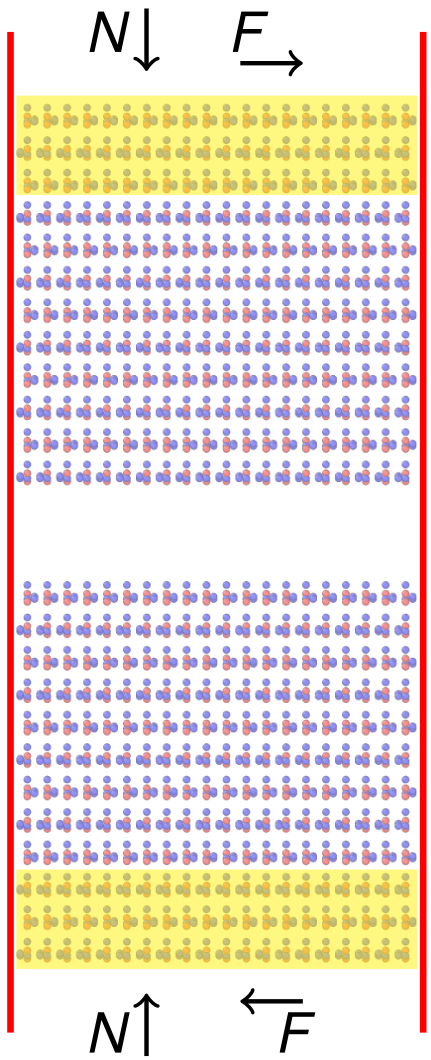
Hamiltonian System

$$H(p,q) = \frac{1}{2} p^T M^{-1} p + U(q)$$



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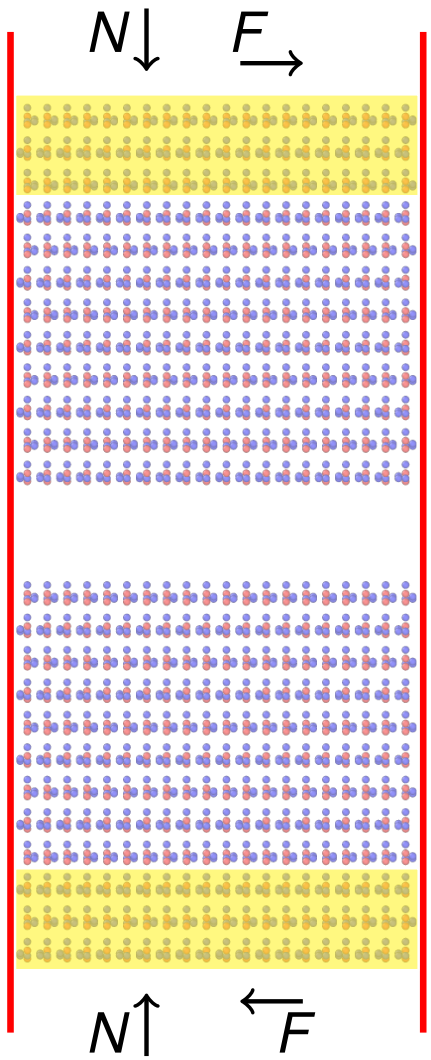
$$H(p,q) = \frac{1}{2} p^T M^{-1} p + U(q)$$

ODE: $$\begin{pmatrix} \dot{q} \\ \dot{p} \end{pmatrix} = \begin{pmatrix} M^{-1} p \\ -\nabla U(q) \end{pmatrix}$$



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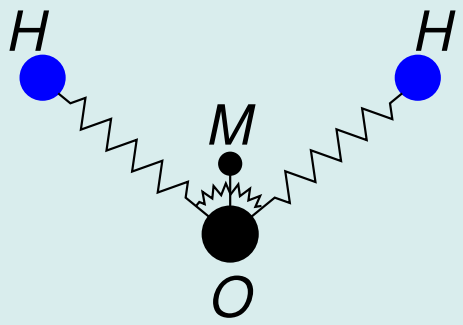
A Flexible Water Model: TIP4P/2005f[1]

$$U(q) = U_{bond}(q) + U_{short}(q) + U_{long}(q)$$

- 2 flexible bonds, 1 flexible angle per molecule
- short range attraction/repulsion between oxygen atoms
- electrostatic interaction between massless charge site *M*, hydrogen

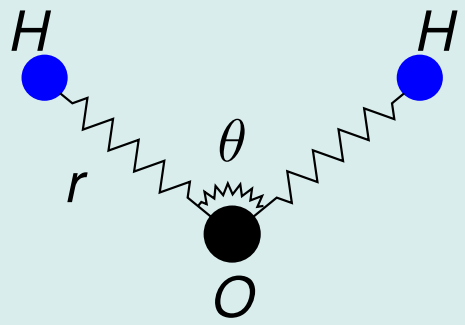


[1] [2011] González, Abascal

A Flexible Water Model: TIP4P/2005f[1]

$$U(q) = U_{bond}(q) + U_{short}(q) + U_{long}(q)$$

- $U_{morse}(r) = D[1 - e^{-\alpha(r-r_0)}]^2$
- $U_{angle}(\theta) = \frac{K_a}{2}(\theta - \theta_0)^2$



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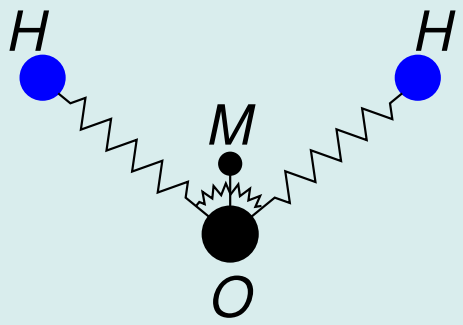
A Flexible Water Model: TIP4P/2005f[1]

$$U(q) = U_{bond}(q) + U_{short}(q) + U_{long}(q)$$

for interactions with $r < r_{cut}$:

- $U_{lj}(r) = 4\epsilon \left[\left(\frac{\sigma}{r} \right)^{12} - \left(\frac{\sigma}{r} \right)^{6} \right]$
- $U_{coul}(r) = \frac{e_i e_j}{4\pi\epsilon_0} \frac{1 - \mathrm{erf}(Gr)}{r}$



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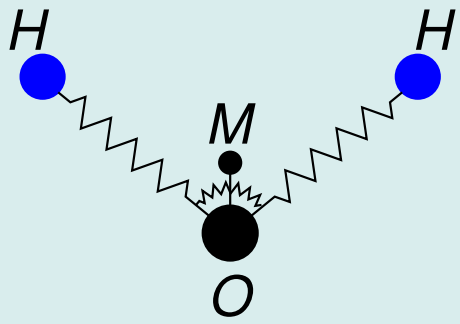
A Flexible Water Model: TIP4P/2005f[1]

$$U(q) = U_{bond}(q) + U_{short}(q) + U_{long}(q)$$

- long-range electrostatic interactions: PME, PPPM, FMM



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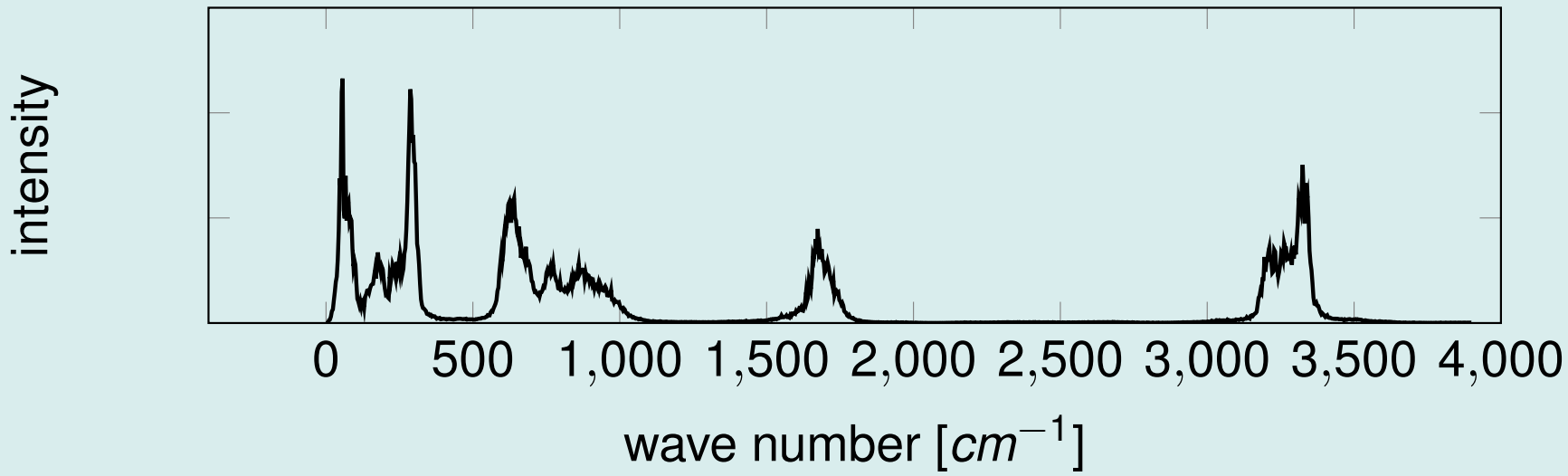


[1] [2011] González, Abascal

Time Integration

Hamiltonian $H(p, q) = T(p) + U(q)$

Verlet Method:

$$\Phi^U_{h/2} \circ \Phi^T_h \circ \Phi^U_{h/2}$$

$\rightarrow$ EXPENSIVE, time step restricted by fastest oscillations

Time Integration

Hamiltonian $H(p,q) = T(p) + U(q) = T(p) + U_{fast}(q) + U_{slow}(q)$

Verlet Method:

$$\Phi^{U}_{h/2} \circ \Phi^{T}_{h} \circ \Phi^{U}_{h/2}$$

$\rightarrow$ EXPENSIVE, time step restricted by fastest oscillations

Impulse Method:

$$\Phi^{U_{slow}}_{h/2} \circ \left(\Phi^{U_{fast}}_{h/2N} \circ \Phi^{T}_{h/N} \circ \Phi^{U_{fast}}_{h/2N} \right)^{N} \circ \Phi^{U_{slow}}_{h/2}$$

$\rightarrow$ CHEAPER, slow force is evaluated less often than fast force

Time Integration

Hamiltonian $H(p,q) = T(p) + U(q) = T(p) + U_{fast}(q) + U_{slow}(q)$

Verlet Method:

$$\Phi^{U}_{h/2} \circ \Phi^{T}_{h} \circ \Phi^{U}_{h/2}$$

$\rightarrow$ EXPENSIVE, time step restricted by fastest oscillations

Impulse Method:

$$\Phi^{U_{slow}}_{h/2} \circ \left(\Phi^{U_{fast}}_{h/2N} \circ \Phi^{T}_{h/N} \circ \Phi^{U_{fast}}_{h/2N} \right)^{N} \circ \Phi^{U_{slow}}_{h/2}$$

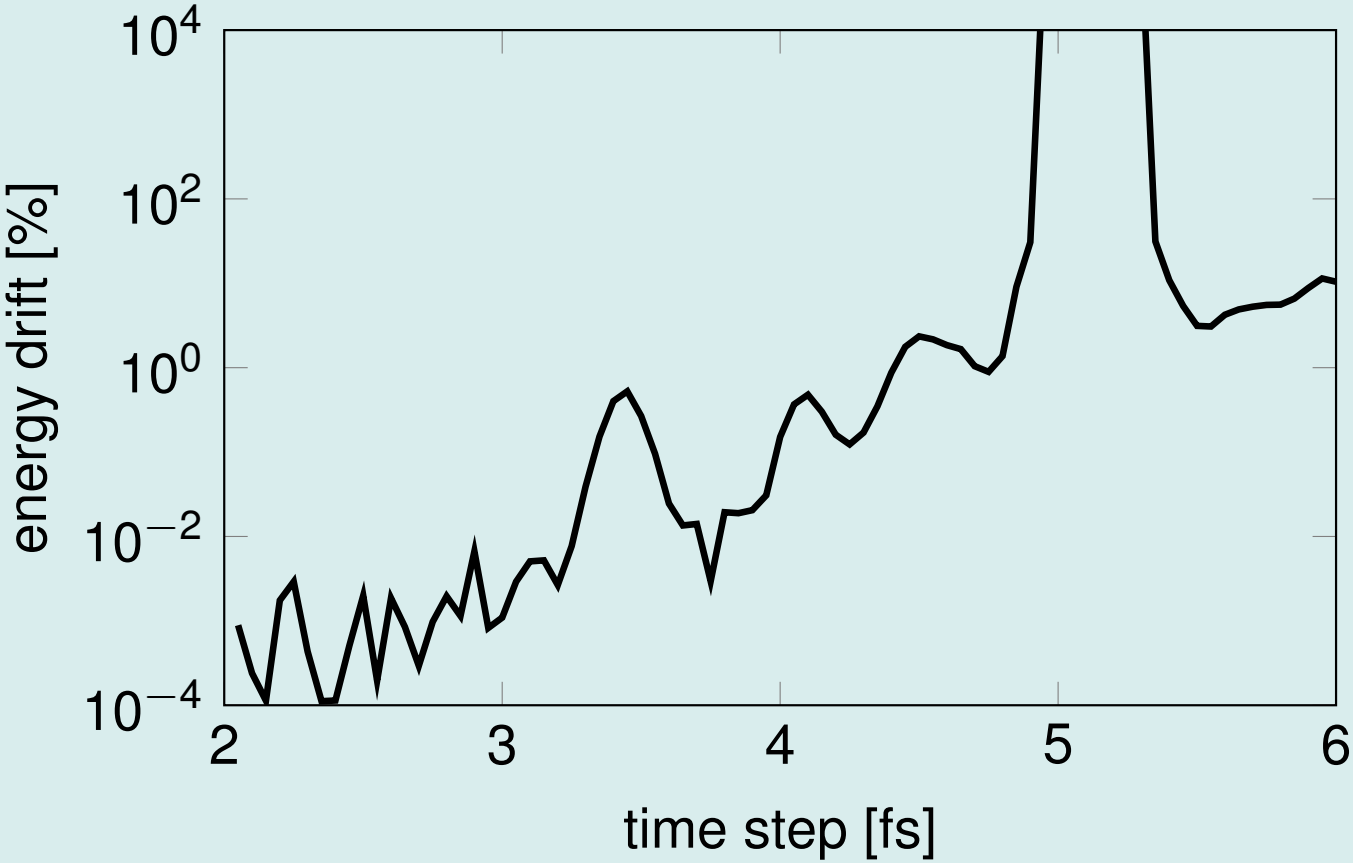
$\rightarrow$ CHEAPER, slow force is evaluated less often than fast force

BUT: resonance issues[2], '5fs barrier'[3]

[2] [2006] Hairer, Lubich, Wanner [3] [1998] Skeel, Izaguirre

Resonance Artifacts

Impulse Method, TIP4P/2005f (50 ps, NVE)



Avoiding Resonances

Techniques:

- freeze fastest dof
- exponential methods
- stochastic damping
- averaging

Avoiding Resonances

Mollified Impulse Method[4]:

- modify slow potential - insert filter Ψ:

$$\tilde{U}(q) = U_{fast}(q) + U_{slow}(\Psi(q))$$

[4] [1998] García-Archilla, Sanz-Serna, Skeel

Avoiding Resonances

Mollified Impulse Method[4]:

- modify slow potential - insert filter Ψ:

$$\tilde{U}(q) = U_{fast}(q) + U_{slow}(\Psi(q))$$

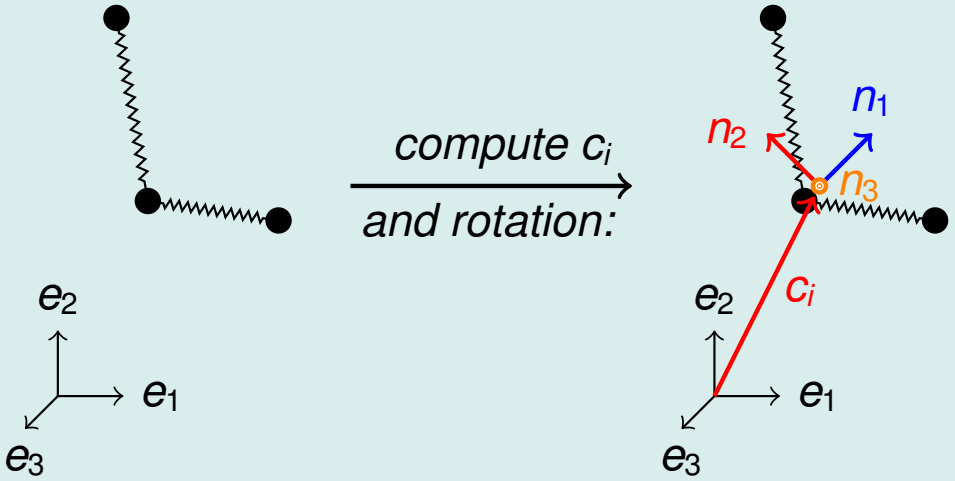
Choice of filter:
$\rightarrow$ close to identity map, derivative easy to compute

- averaging
- equilibrium

[4] [1998] García-Archilla, Sanz-Serna, Skeel

A Corotational Filter

Step 1: identify each molecule by center of mass c_i and vectors n_i:

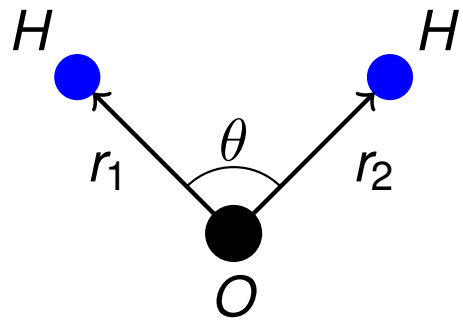


with

$$n_1 = \left(\frac{r_1}{||r_1||} + \frac{r_2}{||r_2||} \right) / \left|\left| \frac{r_1}{||r_1||} + \frac{r_2}{||r_2||} \right|\right|$$

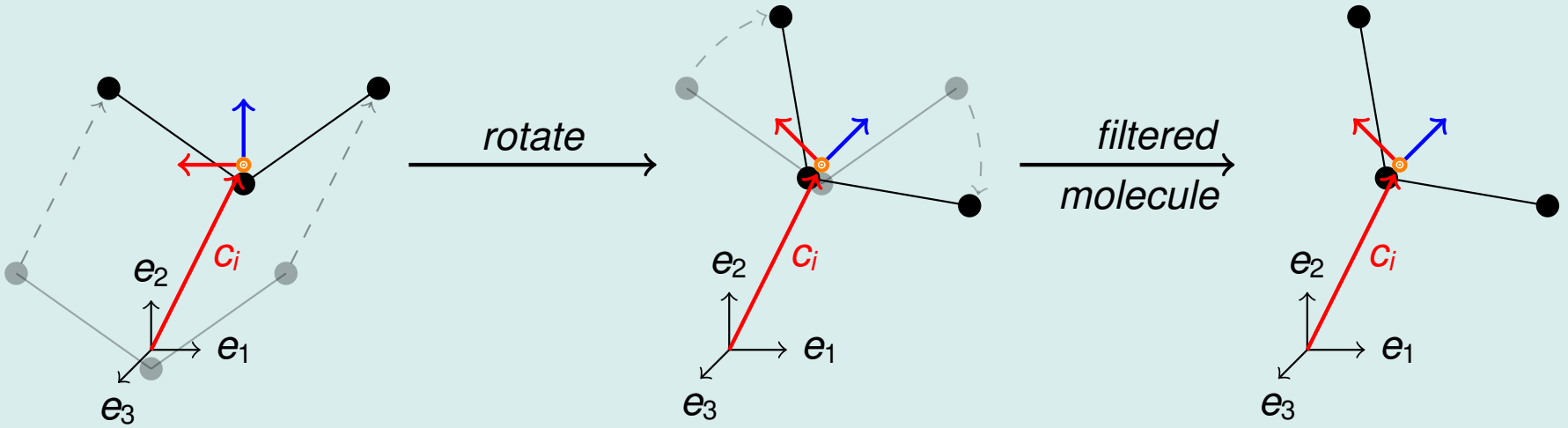
$$n_2 = (n_1 \times n_3) / ||n_1 \times n_3||$$

$$n_3 = (r_1 \times r_2) / ||r_1 \times r_2||$$



A Corotational Filter

Step 2: replace with unit molecule



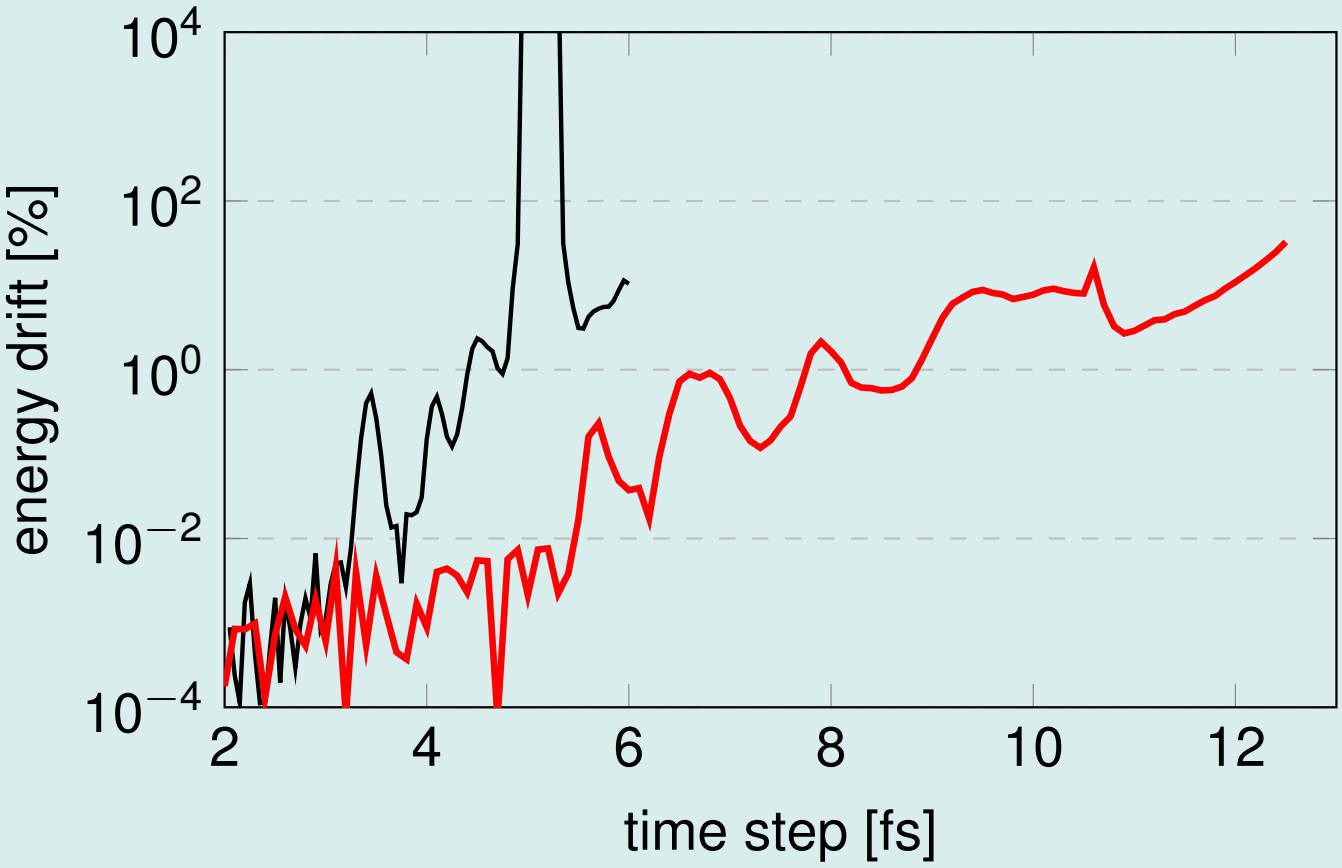
- $\Psi(q^i) = [1,1,1]^T \otimes c_i + (I_3 \otimes N_i N_0^T) q^0$

  (q^0 unit molecule, $N_i = (n_1, n_2, n_3)$ of molecule i)

- $\Psi(q), \Psi_q(q)$ can be computed explicitly!

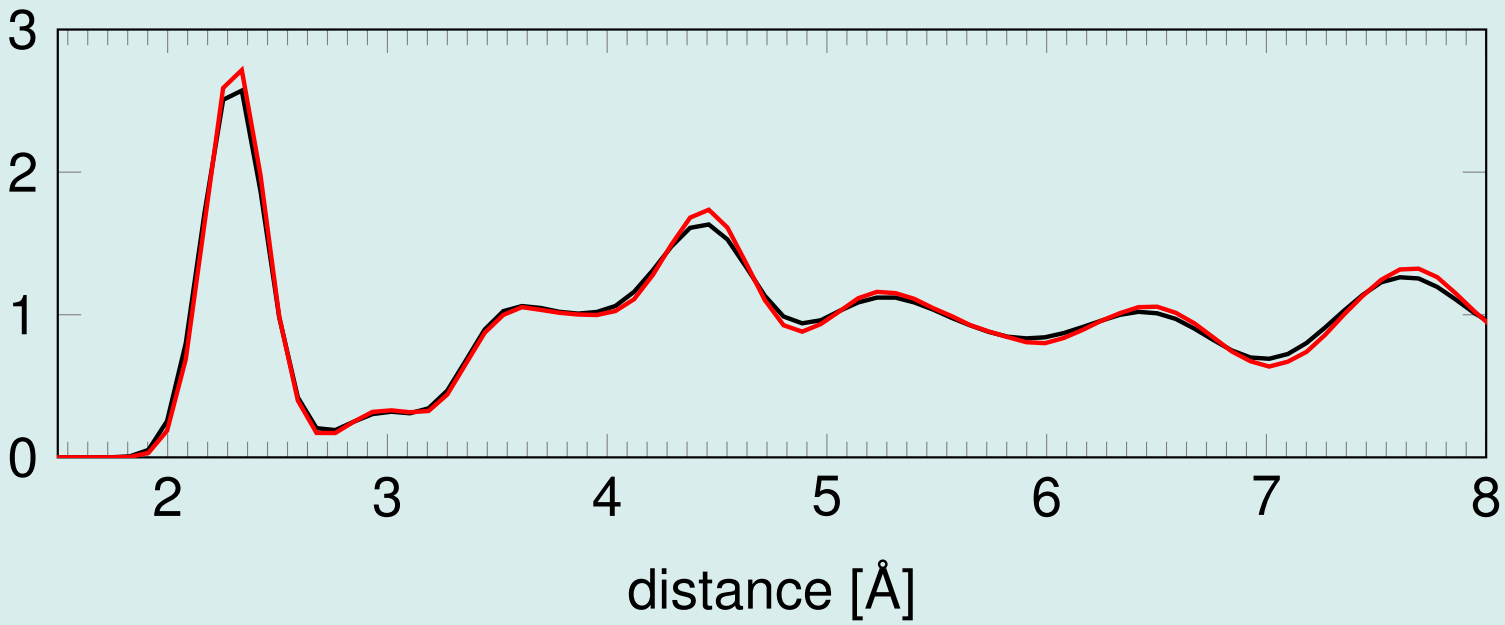
Energy Drift

Filtered Impulse Method, TIP4P/2005f (50 ps, NVE)



Properties of Filter

Radial Distribution Function



Bond Length / Angle

IR Spectra

Kinetic Energy

Potential Energy

Properties of Filter

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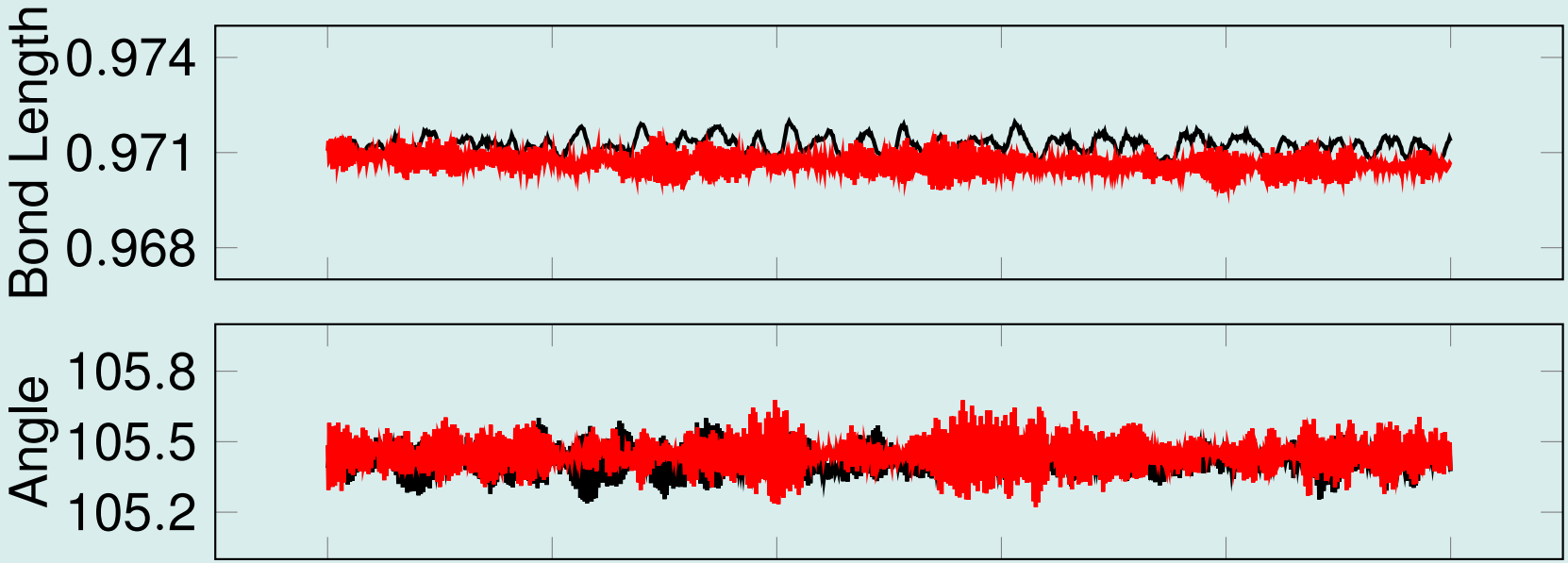
Radial Distribution Function

Bond Length / Angle



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Radial Distribution Function

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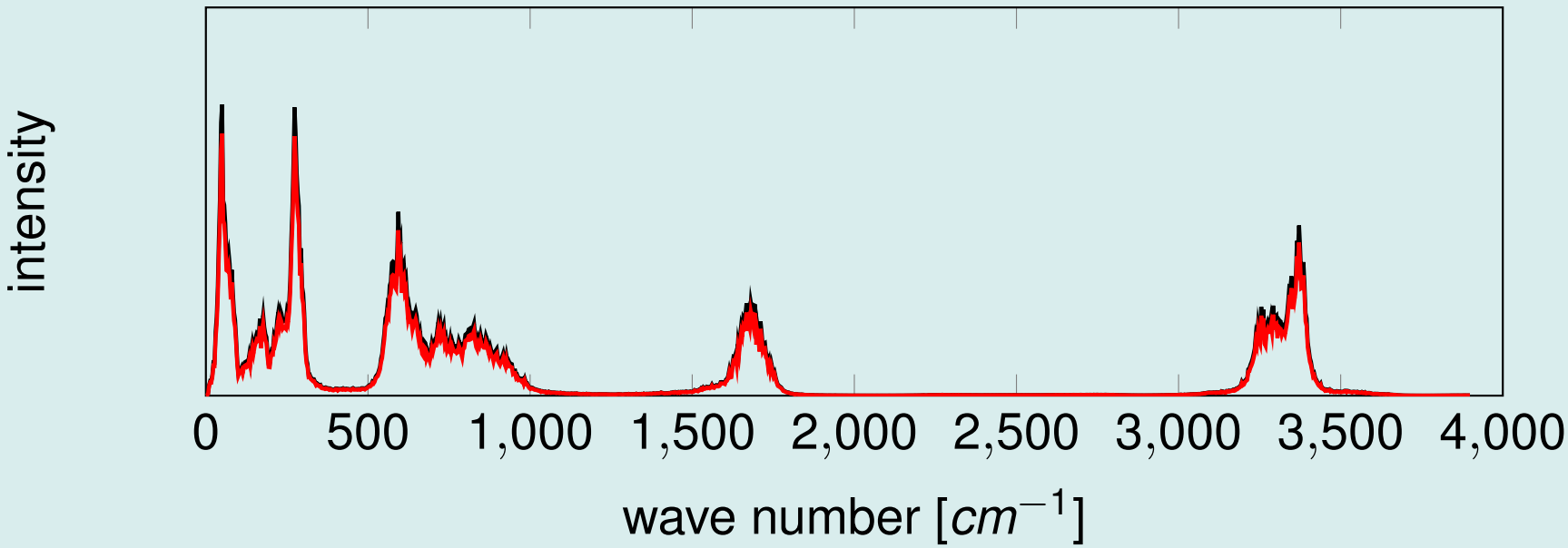
IR Spectra



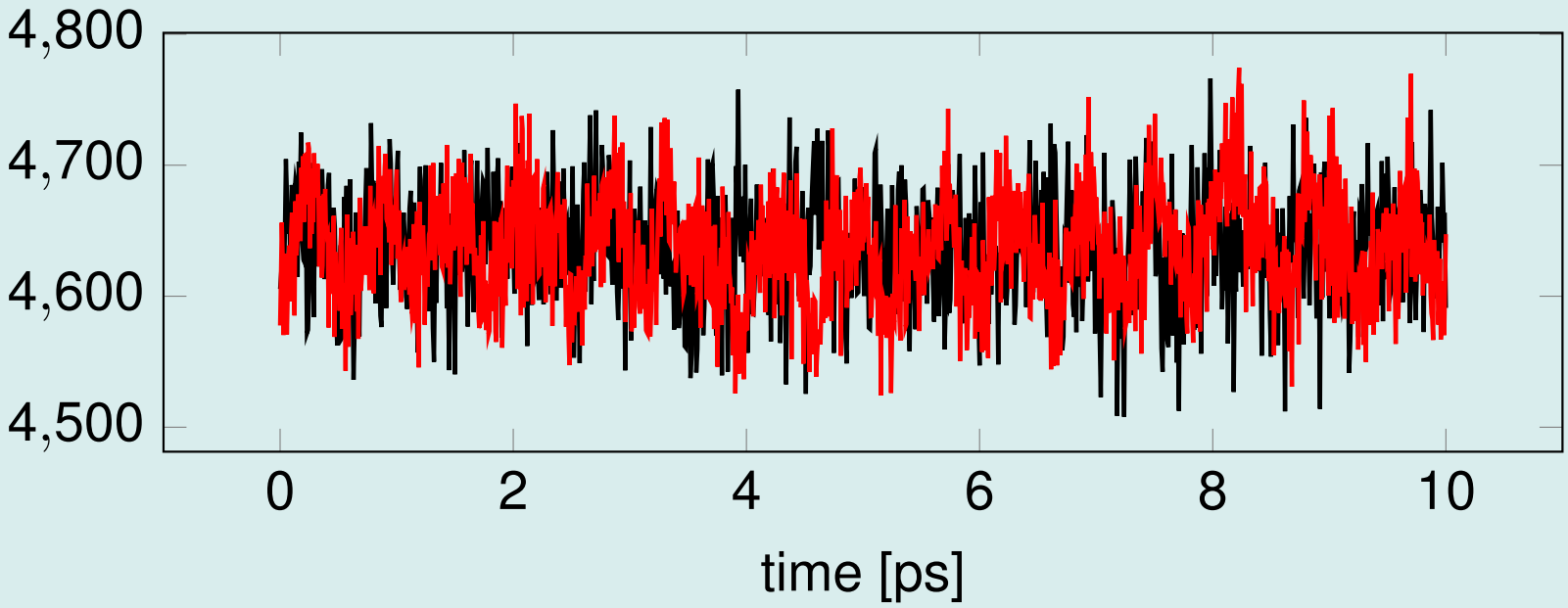
Kinetic Energy

Potential Energy

Properties of Filter

Karlsruhe Institute of Technology

- Radial Distribution Function
- Bond Length / Angle
- IR Spectra
- Kinetic Energy



- Potential Energy

Properties of Filter

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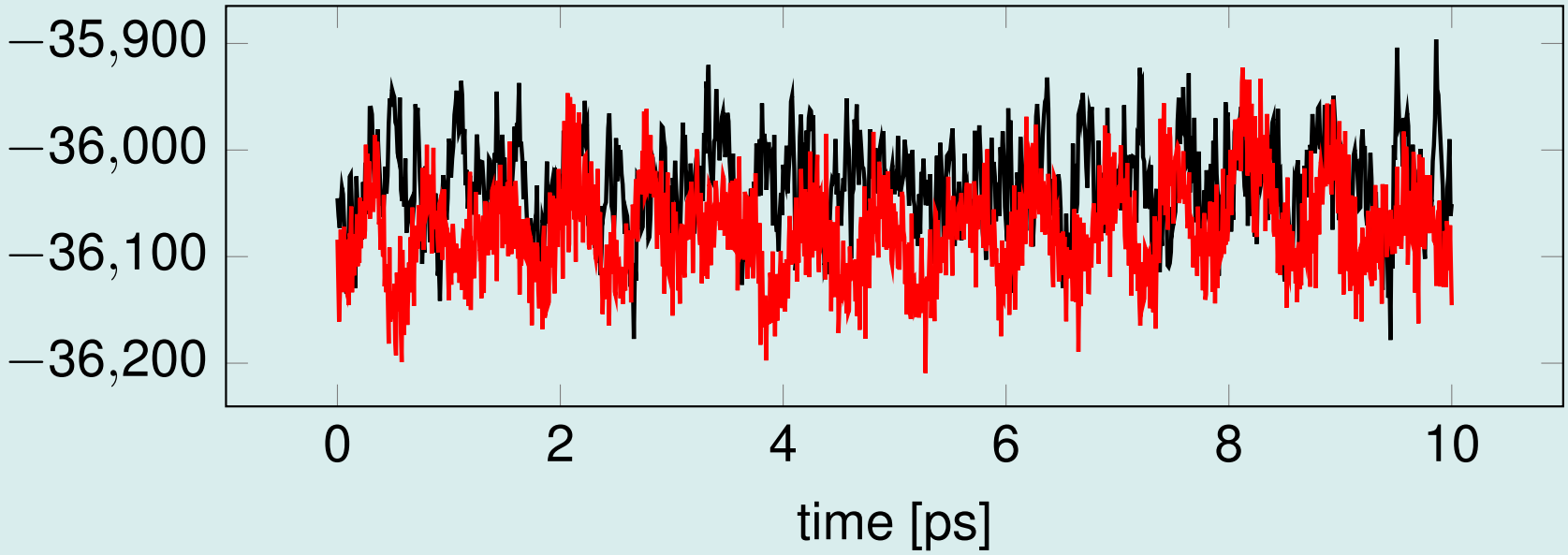
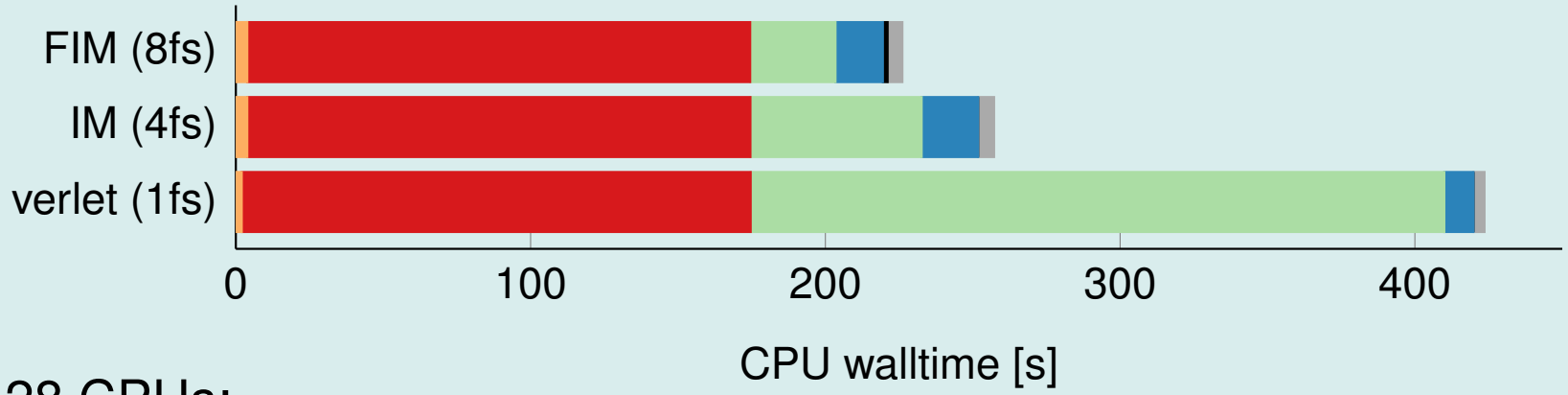
- Kinetic Energy
- Potential Energy



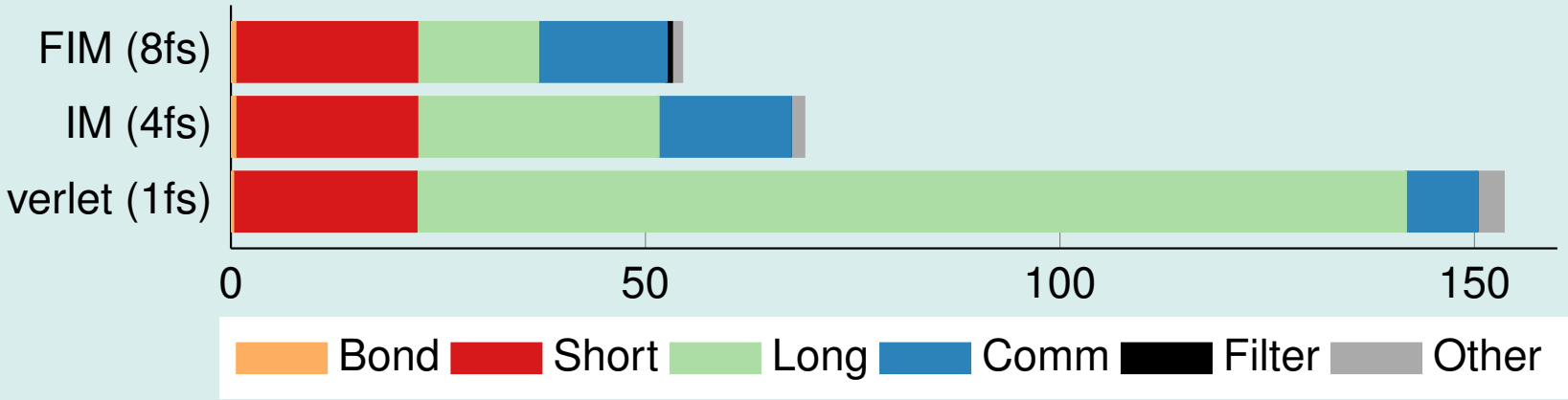
Computational Cost

TIP4P/2005f (8640 atoms, 50ps)

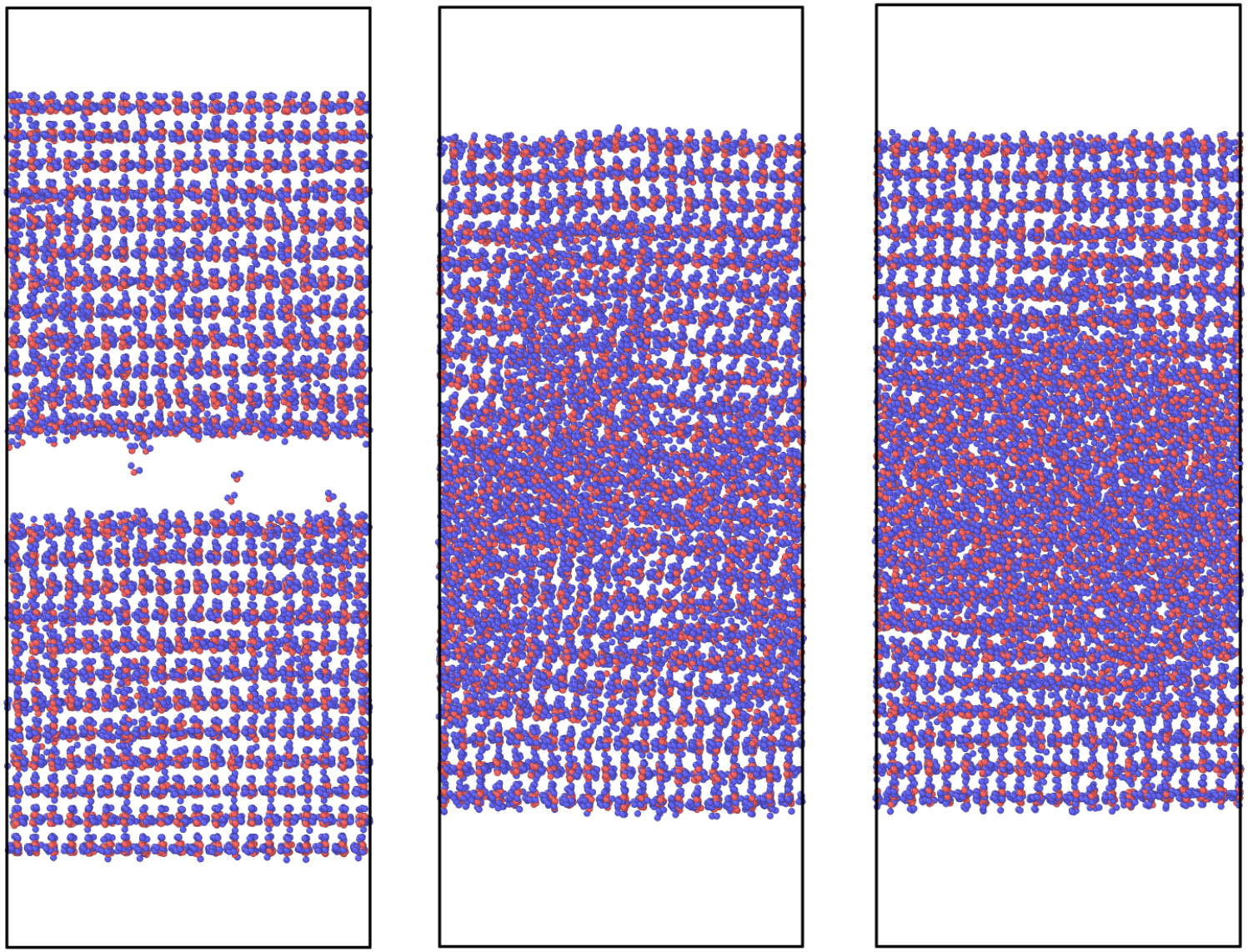
- 16 CPUs (2.6GHz):



- 128 CPUs:

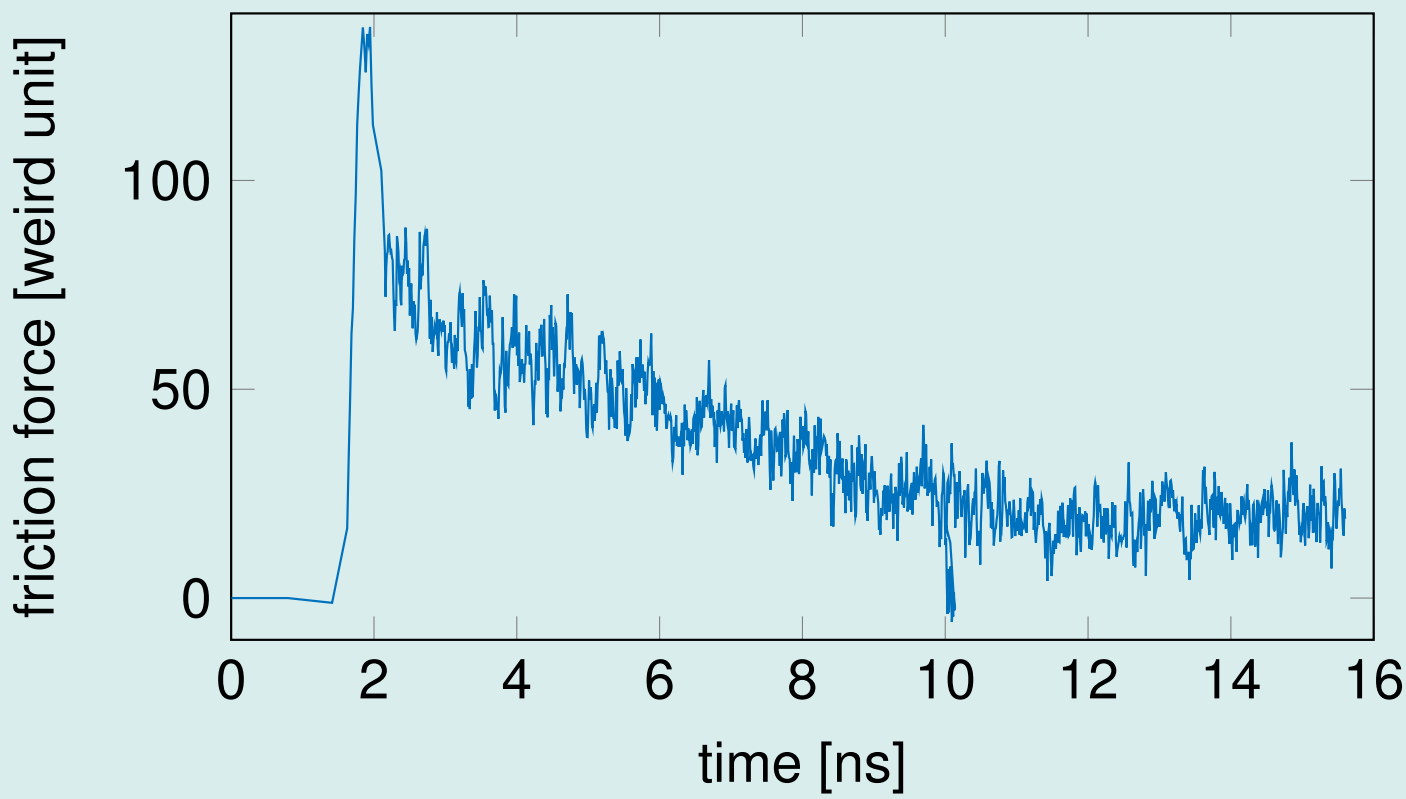


Results

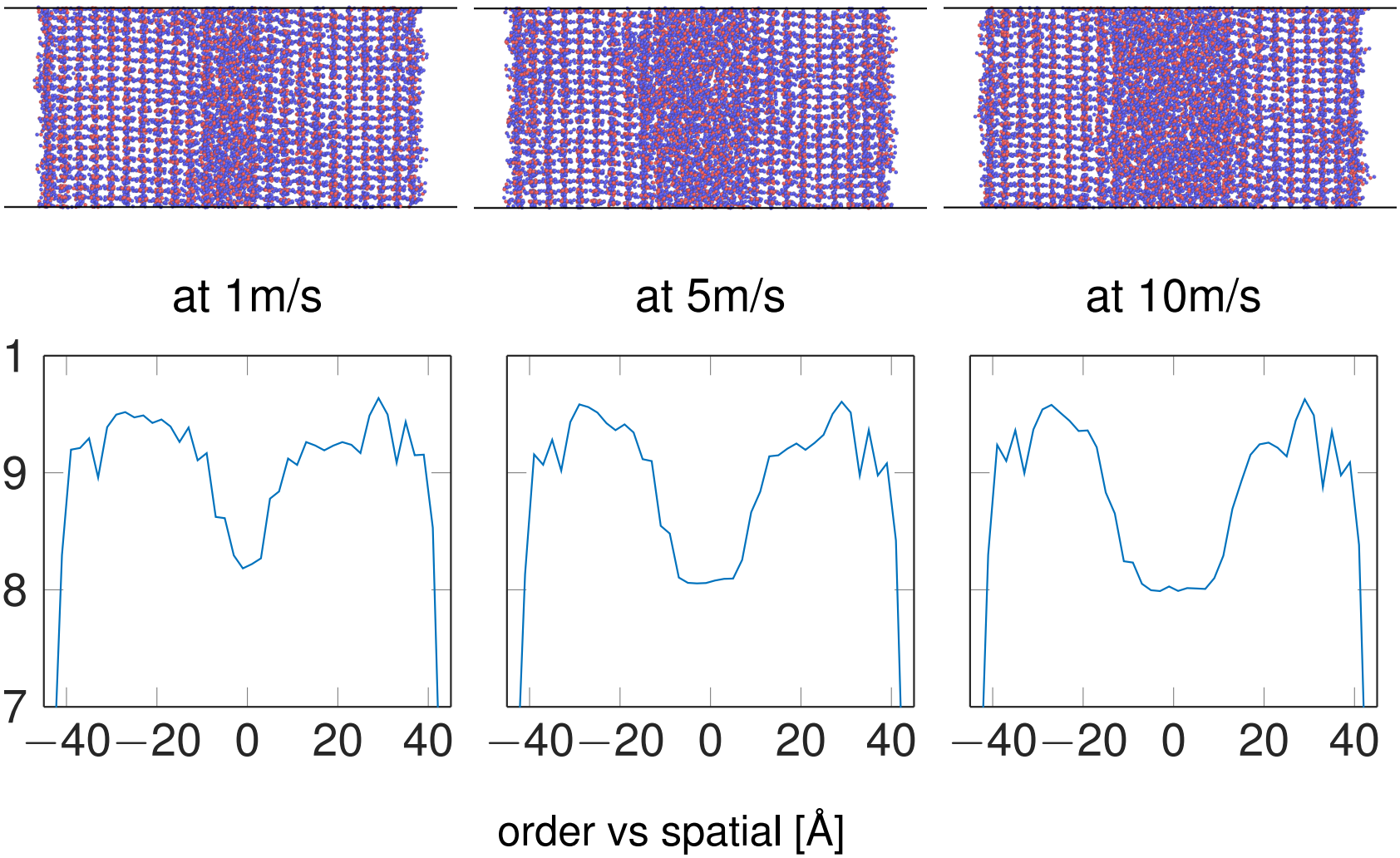


Friction Force

Behaviour of Friction Force



Thickness of Melted Layer



Conclusion

- filter effectively avoids fastest resonances
- outer step size roughly twice as large can be used with this filter
- compared to the impulse method (almost) half the computational time of long-range contributions can be saved