Dual splitting based fast spectral sum-of-Gaussian method for quasi-2D electrostatic systems

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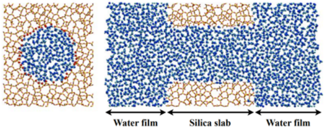
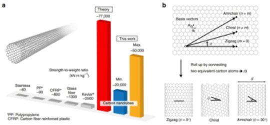
Content

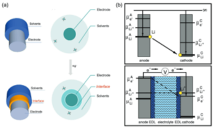
Molecular dynamics in different systems

- Molecular dynamics can be applied in different domains:
  - Triply periodic domain: protein folding/unfolding and DNA aggregation.
  - **Doubly periodic domain:** electrode–electrolyte interface and polyelectrolyte brushes.
  - Singly periodic domain: nanopores and nanotubes.
  - Non-periodic domain: acoustic scattering, non-parametric statistics, and machine learning.

Photo credit[1][2][3]







[1] Ian C Bourg and Carl I Steefel. "Molecular dynamics simulations of water structure and diffusion in silica nanopores". In: *The Journal of Physical Chemistry C* 116.21 (2012), pp. 11556–11564.

[2] Akira Takakura et al. "Strength of carbon nanotubes depends on their chemical structures". In: *Nature communications* 10.1 (2019), p. 3040.

[3] Chong Yan et al. "Toward critical electrode/electrolyte interfaces in rechargeable batteries". In: *Advanced Functional Materials* 30.23 (2020), p. 1909887.

Quasi-2D electrostatic model

- Consider a system of N sources with charge q_n at position $\boldsymbol{r}_n$ in $\Omega = [-\frac{Lx}{2}, \frac{Lx}{2}] \times [-\frac{Ly}{2}, \frac{Ly}{2}] \times [-\frac{Lz}{2}, \frac{Lz}{2}]$.
- Charge neutrality $\sum_{i=1}^{N} q_i = 0$.
- The system has periodicity in x, y direction, while it is free in z direction. Hence the Poisson's equation describes the electrostatic potential Φ with

$$-\Delta\Phi(\boldsymbol{r}) = 4\pi \sum_{\boldsymbol{n}\in\mathcal{N}_2} \sum_{j=1}^{N} q_j \delta\left(\boldsymbol{r} - \boldsymbol{r}_j + \boldsymbol{n} \circ \boldsymbol{L}\right)$$

  where $\mathcal{N}_2 = \{(m_x, m_y, 0), m_x, m_y \in \mathbb{Z}\}$ and $\boldsymbol{L} = (L_x, L_y, L_z)$.
- Free direction restriction need to be set by $\nabla\Phi$ vanishes at z-direction. Also, one needs that $\int_{\mathbb{R}} \int_{[-\frac{Lx}{2}, \frac{Lx}{2}] \times [-\frac{Ly}{2}, \frac{Ly}{2}]} \Phi(x) dx = 0$.

Source kernel splitting

- With these settings, one can use Green function to analytically get the solution (prime means the discarding of $i = j$ and $\boldsymbol{n} = \mathbf{0}$)

$$\Phi\left(\boldsymbol{r}_i\right) = \sum_{j=1}^{N} \sum_{\boldsymbol{n}\in\mathcal{N}_2}' \frac{q_j}{|\boldsymbol{r}_{ij} + \boldsymbol{n} \circ \boldsymbol{L}|}$$

which converges slowly and causes unacceptable $\mathcal{O}(N^2)$ complexity.

- So, the source $f(\boldsymbol{r})$ is commonly decomposed into the near-field and far-field components that

$$f = f^{\mathcal{N}} + f^{\mathcal{F}}, \quad f^{\mathcal{N}} = f - (f * \tau), \quad f^{\mathcal{F}} = f * \tau$$

where τ is an introduced screening function. Hence the potential can be splitted into

$$\Phi\left(\boldsymbol{r}_i\right) = \Phi^{\mathcal{N}}\left(\boldsymbol{r}_i\right) + \Phi^{\mathcal{F}}\left(\boldsymbol{r}_i\right) + \Phi_i^{\text{self}}$$

where Φ_i^{self} is added to exclude the self-interactions.

Notations on quasi-2D system

Definition

Denote $\dot{\boldsymbol{r}} = (x, y)$ and $\dot{\boldsymbol{k}} = (k_x, k_y)$ for the periodic part of position and Fourier mode, where $\dot{\boldsymbol{r}} \in \mathcal{R}^2$ and $\dot{\boldsymbol{k}} \in \mathcal{K}^2$ with

$$\mathcal{R}^2 := \left\{\dot{\boldsymbol{r}} \in \left[-\frac{L_x}{2}, \frac{L_x}{2}\right] \times \left[-\frac{L_y}{2}, \frac{L_y}{2}\right]\right\}, \quad \mathcal{K}^2 := \left\{\dot{\boldsymbol{k}} \in \mathbb{R}^2 : \dot{k}_d \in \frac{2\pi}{L_d}\mathbb{Z},\ d = x, y\right\}.$$

Definition

For any function on 2-periodic space $g : \mathcal{K}^2 \times \mathbb{R} \to \mathbb{C}$, the mixing summation functional $\mathcal{L}$ is defined by

$$\mathcal{L}[g(\boldsymbol{k})] = \mathcal{L}[g([\dot{\boldsymbol{k}}, k_z])] := \frac{1}{2\pi L_x L_y} \sum_{\dot{\boldsymbol{k}} \in \mathcal{K}^2} \int_{\mathbb{R}} g\left([\dot{\boldsymbol{k}}, k_z]\right) \mathrm{d}k_z.$$

Ewald2D decomposition: Formulation

- Hence, for any $f([\dot{\boldsymbol{r}}, z])$ in quasi-2D system, the Fourier transform is presented by

$$\hat{f}\left(\left[\dot{\boldsymbol{k}}, k_z\right]\right) := \int_{\mathcal{R}^2} \int_{\mathbb{R}} f([\dot{\boldsymbol{r}}, z]) e^{-\mathrm{i}\dot{\boldsymbol{k}}\cdot\dot{\boldsymbol{r}}} e^{-\mathrm{i}k_z z} \mathrm{d}z\, \mathrm{d}\dot{\boldsymbol{r}}$$

with the inverse

$$f([\dot{\boldsymbol{r}}, z]) = \mathcal{L}\left[\hat{f}\left(\left[\dot{\boldsymbol{k}}, k_z\right]\right) e^{\mathrm{i}\dot{\boldsymbol{k}}\cdot\dot{\boldsymbol{r}}} e^{\mathrm{i}k_z z}\right].$$

- For Ewald summation, the screening function for splitting is selected to be

$$\tau(\boldsymbol{r}) = \xi^3 \pi^{-3/2} e^{-\xi^2 r^2}, \qquad \widehat{\tau}(\boldsymbol{k}) = e^{-k^2/4\xi^2}$$

- The far-field potential is then

$$\Phi^{\mathcal{F}}(\boldsymbol{r}_i) = 4\pi \sum_{j=1}^{N} q_j \mathcal{L}\left[\frac{e^{-(\dot{k}^2+k_z^2)/4\xi^2}}{\dot{k}^2 + k_z^2} e^{\mathrm{i}\dot{\boldsymbol{k}}\cdot\dot{\boldsymbol{r}}_{ij}} e^{\mathrm{i}k_z z_{ij}}\right].$$

Ewald2D decomposition: Challenges

- In the lattice-based method, the integral along z-axis is discretized using the trapezoidal rule on a uniform grid in Fourier space so that the FFT is feasible.
- **Singularity.** The point $k_z = 0$ is singular when $\dot{\boldsymbol{k}} = \mathbf{0}$.
- **Rapidly change.** When $\dot{\boldsymbol{k}} \neq \mathbf{0}$ but quite small, the integrand will vary rapidly when k_z is close to 0.
- In recent studies, the truncated kernel method (TKM)[4] and adaptive upsampling are introduced to address these issue[5][6]. However, the modified Green's function introduces additional oscillations in the Fourier space and a possibly unacceptable factor in the system of $L_z \ll \min\{L_x, L_y\}$.
- Quasi-2D problem is still open and interesting!

[4] Felipe Vico, Leslie Greengard, and Miguel Ferrando. "Fast convolution with free-space Green's functions". In: *Journal of Computational Physics* 323 (2016), pp. 191–203.

[5] Davoud Saffar Shamshirgar, Joar Bagge, and A-K Tornberg. "Fast Ewald summation for electrostatic potentials with arbitrary periodicity". In: *The Journal of Chemical Physics* 154.16 (2021).

[6] Davoud Saffar Shamshirgar and Anna-Karin Tornberg. "The Spectral Ewald method for singly periodic domains". In: *Journal of Computational Physics* 347 (2017), pp. 341–366.

Content

- The BSA is derived from a Gaussian integral[7][8]

$$\frac{1}{r} = 2\int_0^\infty G_\sigma(rt)dt, \quad G_\sigma(r) = e^{-r^2/2\sigma^2}/\sqrt{2\pi\sigma^2}$$

employing geometrically spaced quadrature $t = b^j$ ($b > 1$ positive constant)

$$\mathcal{B}_b^\sigma(r) \triangleq 2\log(b)\sum_{j=-\infty}^{\infty} b^{-j} G_\sigma\left(b^{-j}r\right) = \frac{2\log(b)}{\sqrt{2\pi\sigma^2}}\sum_{j=-\infty}^{\infty}\frac{1}{b^j}\exp\left[-\frac{1}{2}\left(\frac{r}{b^j\sigma}\right)^2\right]$$

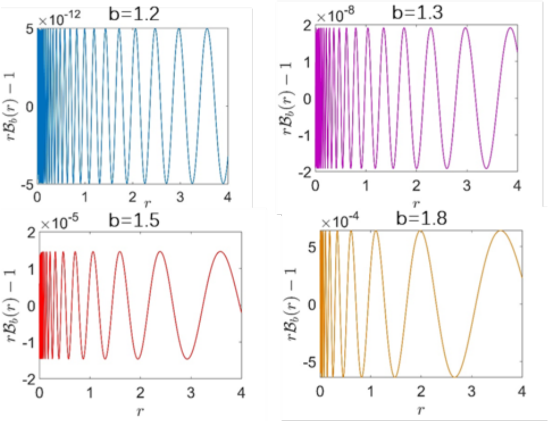
- The relative error of the BSA has the asymptotic bound as $b \to 1$

$$M_b = |r\mathcal{B}_b^\sigma(r) - 1| \lesssim 2^{3/2}\exp\left(-\frac{\pi^2}{2\log(b)}\right)$$

[7] Gregory Beylkin and Lucas Monzón. "On approximation of functions by exponential sums". In: *Appl. Comput. Harmon. Anal.* 19.1 (2005), pp. 17–48.

[8] Gregory Beylkin and Lucas Monzón. "Approximation by exponential sums revisited". In: *Appl. Comput. Harmon. Anal.* 28.2 (2010), pp. 131–149.

Photo credit[9]



[9] Cristian Predescu et al. "The u-series: A separable decomposition for electrostatics computation with improved accuracy". In: *J. Chem. Phys.* 152.8 (2020), p. 084113.

U-series decomposition of $1/r$

- The u-series[10] remains the far part of BSA

$$\mathcal{F}_b^\sigma(r) = \sum_{\ell=0}^{\infty} \omega_\ell e^{-r^2/s_\ell^2} \quad \begin{cases} w_\ell = (\pi/2)^{-1/2} b^{-\ell} \sigma^{-1} \ln b \\ s_\ell = \sqrt{2} b^\ell \sigma \end{cases}$$

and using the complement as the near part

$$\mathcal{N}_b^\sigma(r) = \begin{cases} 1/r - \mathcal{F}_b^\sigma(r), & \text{if } r < r_c \\ 0, & \text{if } r \geq r_c \end{cases}$$

- The cutoff r_c is selected as the smallest root of $r\mathcal{F}_b^\sigma(r) - 1 = 0$, such that the potential is exact up to the cutoff radius and it is continuous at the cutoff point.

[10] Cristian Predescu et al. "The u-series: A separable decomposition for electrostatics computation with improved accuracy". In: *J. Chem. Phys.* 152.8 (2020), p. 084113.

SOG2D decomposition: Remove singularity!

- The u-series decomposition requires the screening function be

$$\tau(\boldsymbol{r}) = \frac{1}{4\pi}\sum_{\ell=0}^{M}\omega_\ell\left(\frac{6}{s_\ell^2} - \frac{4r^2}{s_\ell^4}\right)e^{-r^2/s_\ell^2}, \quad \widehat{\tau}(\boldsymbol{k}) = \frac{\sqrt{\pi}}{4}\sum_{\ell=0}^{M}\omega_\ell s_\ell^3 k^2 e^{-s_\ell^2 k^2/4}$$

  which removes the $1/k^2$ from Laplacian operator

$$\Phi^{\mathcal{F}}(\boldsymbol{r}_i) = 4\pi\sum_{j=1}^{N}q_j\mathcal{L}\left[\frac{\widehat{\tau}(\boldsymbol{k},\omega,s,M)}{k^2}e^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_{ij}}\right]$$

- Explicitly, the far-field potential can be written by

$$\Phi^{\mathcal{F}}(\boldsymbol{r}_i) = \frac{\sqrt{\pi}}{2L_xL_y}\sum_{j=1}^{N}q_j\sum_{\ell=0}^{M}w_\ell s_\ell^3\sum_{\boldsymbol{k}\in\mathcal{K}^2}\int_{\mathbb{R}}e^{-s_\ell^2(k^2+k_z^2)/4}e^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_{ij}}e^{\mathrm{i}k_zz_{ij}}\,\mathrm{d}k_z$$

Truncation Errors of SOG2D

- The truncation error of SOG2D decomposition is only from the deviation beyond the cutoff r_c (H denotes the Heaviside step function)

$$\phi_{\mathrm{err}}(\boldsymbol{r}_i) := \sum_{\boldsymbol{n}\in\mathcal{N}_2}\sum_{j=1}^{N} q_j K(|\boldsymbol{r}_j - \boldsymbol{r}_i + \boldsymbol{n}\circ\boldsymbol{L}|), \quad K(r) = \left(\frac{1}{r} - \sum_{\ell=0}^{M} w_\ell e^{-r^2/s_\ell^2}\right) H(r - r_c)$$

- Follow the analysis in[11], one finds it easily to get

Theorem

The following estimates provide the convergence rate of the potential and force error,

$$|U_{err}| \simeq O\left((\log b)^{-3/2} e^{-\pi^2/2\log b} + b^{-M} + w_{-1} e^{-r_c^2/s_{-1}^2}\right)$$

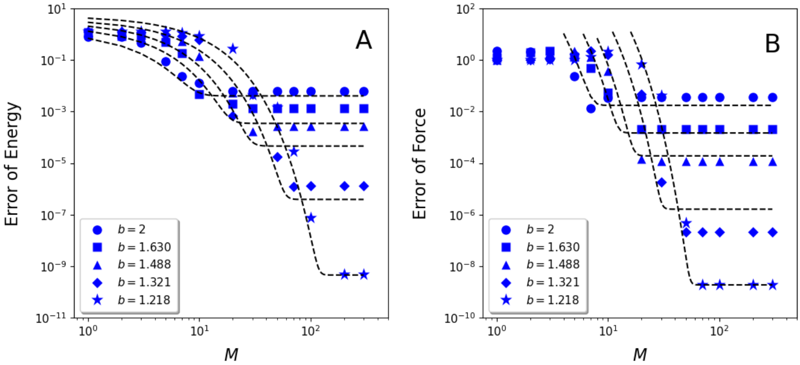
$$|\boldsymbol{F}_{err}(\boldsymbol{r}_i)| \simeq O\left((\log b)^{-3/2} e^{-\pi^2/2\log b} + b^{-3M} + w_{-1}(s_{-1})^{-2} e^{-r_c^2/s_{-1}^2}\right)$$

[11] Jiuyang Liang, Zhenli Xu, and Qi Zhou. *Error estimate of the u-series method for molecular dynamics simulations.* 2023. arXiv: 2305.05369 [math.NA].

Numerical experiment: Random particle

$$U = C_1(\log b)^{-3/2} e^{-\pi^2/2\log b} + C_2 b^{-M} + w_{-1} e^{-r_c^2/s_{-1}^2}$$

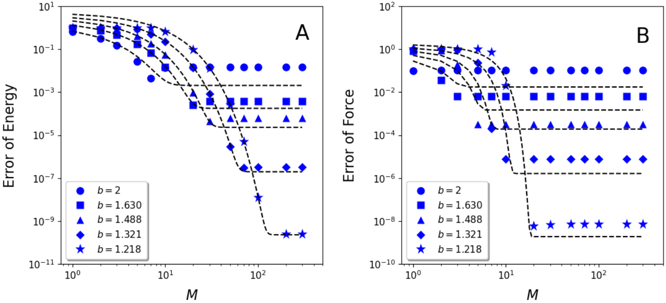
$$|\boldsymbol{F}(\boldsymbol{r}_i)| = C_1(\log b)^{-3/2} e^{-\pi^2/2\log b} + C_2 b^{-3M} + w_{-1}\,(s_{-1})^{-2}\, e^{-r_c^2/s_{-1}^2}$$



Numerical experiment: Quasi-2D NaCl

$$U = C_1 (\log b)^{-3/2} e^{-\pi^2/2\log b} + C_2 b^{-M} + w_{-1} e^{-r_c^2/s_{-1}^2}$$

$$|\boldsymbol{F}(\boldsymbol{r}_i)| = C_1 (\log b)^{-3/2} e^{-\pi^2/2\log b} + \mathbf{exp}(-C_2 b^M) + w_{-1} (s_{-1})^{-2} e^{-r_c^2/s_{-1}^2}$$



Next, one can obtain a reasonable estimate of $\boldsymbol{F}_G^{\text{up}}(\boldsymbol{r}_i)$ by finding the gradient of E_G^{up} (using Eq. (3.32)). Note that $\mathcal{T}(r_{ij}, r_c)$ has zero derivative when $r_{ij} \neq r_c$, and has no definition at $r_{ij} = r_c$. One can simply take the continuous approximation and obtain

$$\boldsymbol{F}_G^{\text{up}}(\boldsymbol{r}_i) \simeq \frac{2\log b}{\sqrt{\pi}} \sum_j \sum_{\ell=M+1}^{\infty} \frac{q_i q_j \boldsymbol{r}_{ij}}{s_\ell^3} e^{-r_{ij}^2/s_\ell^2} = O\left(\frac{1}{s_M^3}\right). \tag{B.7}$$

Calculate method: by closed form?

- The far-field potential is now smooth without singularity

$$\Phi^{\mathcal{F}}(\boldsymbol{r}_i) = \frac{\sqrt{\pi}}{2L_xL_y}\sum_{j=1}^{N} q_j \sum_{\ell=0}^{M} w_\ell s_\ell^3 \sum_{\boldsymbol{k}\in\mathcal{K}^2}\int_{\mathbb{R}} e^{-s_\ell^2\left(k^2+k_z^2\right)/4} e^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_{ij}} e^{\mathrm{i}k_z z_{ij}}\,\mathrm{d}k_z$$

  which also has a closed form

$$\Phi^{\mathcal{F}}(\boldsymbol{r}_i) = \frac{\pi}{L_xL_y}\sum_{j=1}^{N} q_j \sum_{\ell=0}^{M} w_\ell s_\ell^2 e^{-z_{ij}^2/s_\ell^2} \sum_{\boldsymbol{k}\in\mathcal{K}^2} e^{-s_\ell^2 k^2/4} e^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_{ij}}$$

- **By closed form:** Expensive $\mathcal{O}(N^2)$! Earlier spectral Ewald methods such as periodization[12], truncation[13], or regularization[14] have made some improvements but still unsatisfied.

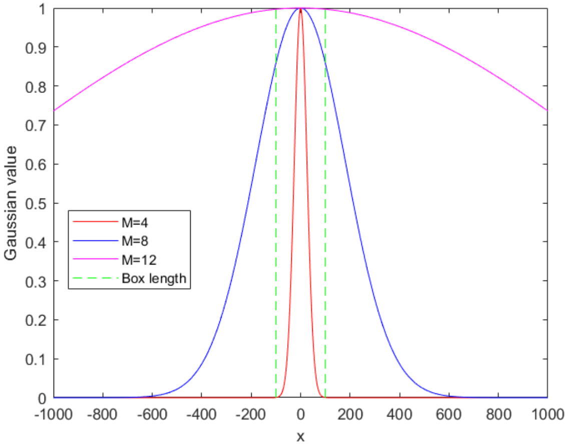
[12] Dag Lindbo and Anna-Karin Tornberg. "Fast and spectrally accurate Ewald summation for 2-periodic electrostatic systems". In: *The Journal of chemical physics* 136.16 (2012).

[13] Peter Minary et al. "A new reciprocal space based treatment of long range interactions on surfaces". In: *The Journal of chemical physics* 116.13 (2002), pp. 5351–5362.

[14] Franziska Nestler, Michael Pippig, and Daniel Potts. "Fast Ewald summation based on NFFT with mixed periodicity". In: *Journal of Computational Physics* 285 (2015), pp. 280–315.

Calculate method: direct FFT accleration?

- Indiscriminately applying the FFT is not efficient, since there are different kind of Gaussians...



- Since bandwith parameter s_ℓ varies from medium to extreme large, the corresponding Fourier transforms will become rapidly decaying, which requires a large-scale zero-padding in FFT acceleration to achieve better resolution in the frequency domain.
- Quantitively, for tolerance ϵ and bandwidth s_ℓ, the cutoff range in k-space need to be

$$s_\ell^2 K_{max}^2/4 \geq -\log(\epsilon)$$

which causes the zero-padding number be directly proportional to

$$\mu \sim s_\ell L_z^{-1}$$

- This problem cannot be directly addressed by either the TKM or its variants[15][16], as the support set of the source term expands with increasing ℓ, and truncating the Green's function proves inefficient.

[15] Felipe Vico, Leslie Greengard, and Miguel Ferrando. "Fast convolution with free-space Green's functions". In: *Journal of Computational Physics* 323 (2016), pp. 191–203.

[16] Leslie Greengard, Shidong Jiang, and Yong Zhang. "The anisotropic truncated kernel method for convolution with free-space Green's functions". In: *SIAM J. Sci. Comput.* 40.6 (2018), A3733–A3754.

Low-rank Chebyshev approximation

- For far-field Gaussians with $s_\ell \gg L_z$, real-space Chebyshev approximation can be effectively resolve these part.

Lemma

Let the Chebyshev polynomial of degree n on $[-1,1]$ be defined by

$$T_n(\cos(\theta)) = \cos(n\theta), \quad \theta \in [0,\pi].$$

Let $f(x)$ be a smooth function on the interval $[-1,1]$. Then

$$f(x) = {\sum_{n=0}^{\infty}}' a_n T_n(x),$$

where the prime indicates that there a factor of $1/2$ is multiplied in front of a_0, and the coefficients are strictly given by

$$a_n = \frac{2}{\pi}\int_0^{\pi} f(\cos(\theta))\cos(k\theta)d\theta$$

Lemma

The Chebyshev nodes of the first kind are the zeros of $T_n(x)$, given by

$$\left\{x_i = \cos\left[\frac{(i-1/2)\pi}{P}\right], i = 1, \cdots, P\right\}.$$

We define the $P \times P$ basis matrix $\boldsymbol{V}$ by $V(i,j) = T_j(x_i)$. Given the vector of function values $\boldsymbol{f} = (f(x_1), \cdots, f(x_P))$, $f(x)$ can be approximated by a P-term truncated Chebyshev series where the coefficients $\boldsymbol{a} = (a_1, \cdots, a_P)$ can be obtained as

$$\boldsymbol{a} = \boldsymbol{V}^{-1}\boldsymbol{f}$$

Given a set of N additional points $\{y_j \in [-1,1], j = 1, \cdots, N\}$, we define the $N \times P$ evaluation matrix $\boldsymbol{E}$ by $E(j,n) = T_n(y_j)$, so that $\boldsymbol{Ea}$ is the value of the interpolant at the additional points.

- From the two basic lemmas of Chebyshev approximation, one has the following error estimation for far-field Gaussians.

Theorem

Assume $s_\ell \geq \eta L_z$ with $\eta > 0$. By the Chebyshev interpolation, approximating the Gaussian function e^{-z^2/s_ℓ^2} at the interval $[-L_z/2, L_z/2]$ has an error bound

$$\left| e^{-z^2/s_\ell^2} - \sum_{n=0}^{P-1}{}' a_n T_n(z) \right| \leq \frac{1}{2P!(2\eta)^P}$$

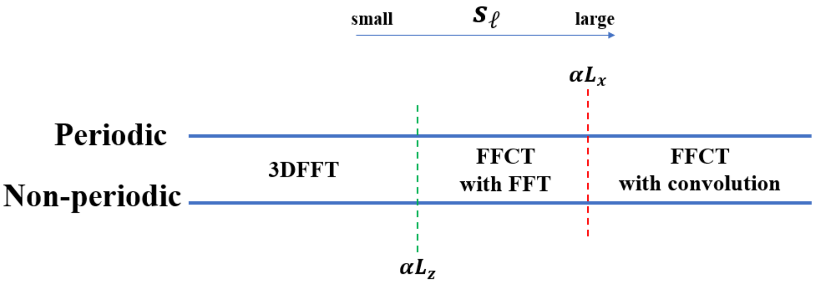
- Hence, for given $\alpha > 0$, we do the following dual splitting for far-field SOGs that
  - **Mid-ranged.** $s_\ell \leq \alpha L_z$.
  - **Long-ranged.** $s_\ell > \alpha L_z$.
- Denote $\mathcal{M}_1$ the critical term, then the far-field potential

$$\Phi^{\mathcal{F}} = \sum_{\ell=0}^{\mathcal{M}_1} \Phi_\ell^{\mathcal{F}} + \sum_{\ell=\mathcal{M}_1+1}^{M} \Phi_\ell^{\mathcal{F}} := \Phi^{\mathcal{M}_1} + \Phi^{\mathcal{M}_2}$$

Content

Overview of algorithm idea

- Based on dual splitting and different geometry of simulation box, FSSOG can be described by the following schema.



Mid-range part: Fourier spectral solver

- Since the mid-range Gaussians have relatively short effect range, the accuracy of Fourier spectral solver on z-direction can be promised.
- To directly caluclate the following expression is expensive

$$\Phi^{\mathcal{M}_1}(\boldsymbol{r}_i) = 4\pi \sum_{j=1}^{N} q_j \mathcal{L}\left[\frac{\widehat{\mathcal{T}}^{\mathcal{M}_1}(\boldsymbol{k}, \omega, s, M)}{k^2} e^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_{ij}}\right]$$

- Hence a window function W with compact support is introduced for calculating more effectively.
- A trivial fact is

$$1 \equiv \widehat{W}(\boldsymbol{k})[\widehat{W}(\boldsymbol{k})]^{-2}\widehat{W}(\boldsymbol{k})$$

so one can insert those components into the potential

$$\Phi^{\mathcal{M}_1}(\boldsymbol{r}_i) = 4\pi\mathcal{L}\left[\widehat{W}(\boldsymbol{k})e^{\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_i}\frac{\widehat{\mathcal{T}}^{\mathcal{M}_1}(\boldsymbol{k}, \omega, s, M)}{|\boldsymbol{k}|^2}[\widehat{W}(\boldsymbol{k})]^{-2}\sum_{j=1}^{N} q_j \widehat{W}(\boldsymbol{k})e^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_j}\right]$$

- **Step 1: Gridding.** Interpolate all sources into uniform real grids

$$H(\boldsymbol{r}) = \sum_{j=1}^{N} q_j W(\boldsymbol{r} - \boldsymbol{r}_j)_*$$

- **Step 2: R2F.** Using 3D-FFT to get

$$\widehat{H}(\boldsymbol{k}) := \sum_{j=1}^{N} q_j \widehat{W}(\boldsymbol{k}) e^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_j}$$

- **Step 3: Scaling.** Multiplicating scaling factor for each Fourier mode

$$\widehat{\widetilde{H}}(\boldsymbol{k}) := \frac{\widehat{\mathcal{T}}^{\mathcal{M}_1}(\boldsymbol{k}, \omega, s, M)}{|\boldsymbol{k}|^2} [\widehat{W}(\boldsymbol{k})]^{-2} \widehat{H}(\boldsymbol{k})$$

- **Step 4: F2R.** Using 3D-IFFT to get $\widetilde{H}(\boldsymbol{\dot{r}}, z)$.
- **Step 5: Gathering.** Using Plancherel's theorem to gather all contributions (with compact support) into sources by

$$\Phi^{\mathcal{M}_1}(\boldsymbol{r}_i) = 4\pi \int_{\mathbb{R}} \int_{\mathcal{R}^2} \widetilde{H}(\boldsymbol{\dot{r}}_j, z_j)\, W(\boldsymbol{\dot{r}}_i - \boldsymbol{\dot{r}}_j)_*\, W(z_i - z_j)\, d\boldsymbol{\dot{r}}_j dz_j$$

Window functions

- **Gaussian window**[17]**.** Fast, smooth, decay rapidly in Fourier space, need to be truncated.

$$\mathrm{W}_{\mathrm{G}}(x) = \begin{cases} e^{-\alpha(x/w)^2}, & |x| \le w \\ 0, & \text{otherwise} \end{cases} \qquad \widehat{\mathrm{W}}_{\mathrm{G},\ \text{untrunc}}(k) = \sqrt{\frac{\pi}{\alpha}} w e^{-k^2 w^2/4\alpha}$$

- **Kaiser-Bessel window**[18]**.** Require a significantly smaller half-width to achieve the same target accuracy. Need to be truncated.

$$\mathrm{W}_{\mathrm{KB}}(x) = \begin{cases} \frac{I_0\left(\beta\sqrt{1-(x/w)^2}\right)}{I_0(\beta)}, & |x| \le w \\ 0, & \text{otherwise} \end{cases} \qquad \widehat{\mathrm{W}}_{\mathrm{KB}}(k) = \frac{2w \sinh\left(\sqrt{\beta^2 - k^2 w^2}\right)}{I_0(\beta)\sqrt{\beta^2 - k^2 w^2}}$$

[17] Dag Lindbo and Anna-Karin Tornberg. "Spectral accuracy in fast Ewald-based methods for particle simulations". In: *Journal of Computational Physics* 230.24 (2011), pp. 8744–8761.

[18] J Kaiser and R Schafer. "On the use of the I 0-sinh window for spectrum analysis". In: *IEEE Transactions on Acoustics, Speech, and Signal Processing* 28.1 (1980), pp. 105–107.

- **Cardinal B-spline window[19].** Finite support, easy and fast to implement, polynomial degree of smoothness.

$$M_2(x) = \begin{cases} 1 - |x-1|, & 0 \le x \le 2 \\ 0, & \text{otherwise} \end{cases} \qquad M_p(x) = \frac{x}{p-1} M_{p-1}(x) + \frac{p-x}{p-1} M_{p-1}(x-1)$$

- **Exponential of semicircle window[20].** Achieve high precision w.r.t. KB window with the same width, cheaper to evaluate. Unknown of its Fourier transform, not differentiable at endpoints.

$$\mathrm{W}_{\mathrm{ES}}(x) = \begin{cases} \frac{e^{\beta\sqrt{1-(x/w)^2}}}{e^{\beta}}, & -w \le x \le w \\ 0, & \text{otherwise.} \end{cases}$$

[19] Ulrich Essmann et al. "A smooth particle mesh Ewald method". In: *J. Chem. Phys.* 103.19 (1995), pp. 8577–8593.

[20] Alexander H Barnett, Jeremy Magland, and Ludvig af Klinteberg. "A parallel nonuniform fast Fourier transform library based on an "exponential of semicircle" kernel". In: *SIAM Journal on Scientific Computing* 41.5 (2019), pp. C479–C504.

Long-range part: Fourier-Chebyshev spectral solver

- For the long-range contribution

$$-\Delta\Phi^{\mathcal{M}_2}(\boldsymbol{r}) = \left(f * \tau^{\mathcal{M}_2}\right)(\boldsymbol{r})$$

we will ultilize a Fourier solver for the first two periodic dimensions, and a well-conditioned Chebyshev solver for the free dimension.

$$-\frac{\partial^2 \widehat{\Phi}^{\mathcal{M}_2}(\dot{\boldsymbol{k}}, z)}{\partial z^2} + \dot{k}^2 \widehat{\Phi}^{\mathcal{M}_2}(\dot{\boldsymbol{k}}, z) = \widehat{f * \tau^{\mathcal{M}_2}}(\dot{\boldsymbol{k}}, z)$$

- Indeed, we apply a basis expansion in the function space by

$$f(x, y, z) = \sum_{l=-\infty}^{+\infty} \sum_{m=-\infty}^{+\infty} \sum_{n=0}^{+\infty} f_{l,m,n} e^{i\frac{2\pi}{L_x} lx} e^{i\frac{2\pi}{L_y} my} T_n^{L_z/2}(z)$$

which allows us to reduce all differential(integral) relation of functions into corresponding relation of basis. Denote $T_n(z) = T_n^{L_z/2}(z)$ for convenience.

Lemma

Let $f(x)$ be a smooth function on $[-1,1]$ given by a Chebyshev series $f(x) = {\sum_{n=0}^{\infty}}' a_n T_n(x)$. Then the integral of f has a Chebyshev expansion of the form

$$\int_{-1}^{x} f(t)dt = \sum_{n=1}^{\infty} b_n T_n(x) + b_0,$$

where b_0 is a constant term and other coefficients are given by

$$b_n = \frac{1}{2n}\left(a_{n-1} - a_{n+1}\right).$$

The double integral of f also has an expansion of the form

$$\int_{-1}^{x}\int_{-1}^{\tau} f(t)dtd\tau = \sum_{n=2}^{\infty} c_n T_n(x) + c_1 x + c_0,$$

where $c_1 = b_0, c_0$ is a constant term and

$$c_n = \frac{1}{2n}\left[\frac{a_{n-2} - a_n}{2(n-1)} - \frac{a_n - a_{n+2}}{2(n+1)}\right].$$

- Hence with the P-term Chebyshev expansion

$$\frac{\partial^2 \widehat{\Phi}^{\mathcal{M}_2}}{\partial z^2}(\dot{\boldsymbol{k}}, z) = \sum_{n=0}^{P-1} \hat{a}_n(\dot{\boldsymbol{k}}) T_n(z), \quad \widehat{\Phi}^{\mathcal{M}_2}(\dot{\boldsymbol{k}}, z) = \sum_{n=0}^{P-1} \hat{c}_n(\dot{\boldsymbol{k}}) T_n(z)$$

  and

$$\widehat{f * \tau^{\mathcal{M}_2}}(\dot{\boldsymbol{k}}, z) = \sum_{n=0}^{P-1} \hat{f}_n(\dot{\boldsymbol{k}}) T_n(z),$$

  one can derive the coefficients relation that

$$-\hat{a}_n(\dot{\boldsymbol{k}}) + \frac{\dot{k}^2 L_z^2}{4} \hat{c}_n(\dot{\boldsymbol{k}}) = \hat{f}_n(\dot{\boldsymbol{k}}), \quad n = 0, \cdots, P-1 \tag{1}$$

- From the lemma above, one can represent $\hat{c}_n$ as terms of $\hat{a}_n$. So here are P equations for $P+2$ free unkowns, $\hat{a}_0, \cdots, \hat{a}_{P-1}, \hat{c}_1, \hat{c}_0$. The two extra restriction will be offered by boundary conditions so that the integration coefficients $\hat{c}_1, \hat{c}_0$ can be determined.
- $\dot{\boldsymbol{k}} = \mathbf{0}$ will cause irreducibility of that system, for which we can apply Chebyshev-FGT[21] to obtain this contribution with only linear cost.

[21] Johannes Tausch and Alexander Weckiewicz. "Multidimensional fast Gauss transforms by Chebyshev expansions". In: *SIAM Journal on Scientific Computing* 31.5 (2009), pp. 3547–3565.

FFT acceleration for Fourier-Chebyshev solver

- Similar from Fourier spectral solver, we can define

$$\widehat{\Phi}_W^{\mathcal{M}_2}(\dot{\boldsymbol{k}}, z) = \frac{1}{\widehat{W}(\dot{\boldsymbol{k}})}\widehat{\Phi}^{\mathcal{M}_2}(\dot{\boldsymbol{k}}, z), \quad \widehat{H}_W(\dot{\boldsymbol{k}}, z) := \widehat{W * f * \tau^{\mathcal{M}_2}}(\dot{\boldsymbol{k}}, z)$$

- Hence the equation will turn to

$$-\frac{\partial^2 \widehat{\Phi}_W^{\mathcal{M}_2}(\dot{\boldsymbol{k}}, z)}{\partial z^2} + \dot{k}^2 \widehat{\Phi}_W^{\mathcal{M}_2}(\dot{\boldsymbol{k}}, z) = [\widehat{W}(\dot{\boldsymbol{k}})]^{-2} \widehat{H}_W(\dot{\boldsymbol{k}}, z) := \widetilde{\widehat{H}}_W(\dot{\boldsymbol{k}})$$

- And the final gathering step can also be implemented

$$\Phi^{\mathcal{M}_2}(\dot{\boldsymbol{r}}, z) = \int_{\mathcal{R}^2} \Phi_W^{\mathcal{M}_2}(\dot{\boldsymbol{r}}_j, z)\, W(\dot{\boldsymbol{r}} - \dot{\boldsymbol{r}}_j)_* \, d\dot{\boldsymbol{r}}_j$$

from the solver of

$$\Phi_W^{\mathcal{M}_2}(\dot{\boldsymbol{r}}_j, z) = \sum_{n=0}^{P-1} c_n(\dot{\boldsymbol{r}}_j)\, T_n(z)$$

How to implement Gridding step?

- Different from 3DFFT, the gridding step poses a significant chanllenge since the Chebyshev polynomials do not have corresponding convolution theorem.

$$H_W(\dot{\boldsymbol{r}}, z) = W * f * \tau^{\mathcal{M}_2}(\dot{\boldsymbol{r}}, z)$$

- Since $s_l > \alpha L_z$, the Q-term Taylor expansion reads

$$e^{-z^2/s_\ell^2} = \sum_{n=0}^{Q-1} \frac{1}{n!} \left(-\frac{z^2}{s_\ell^2}\right)^n + O\left(\frac{z^{2Q}}{s_\ell^{2Q}}\right)$$

- One can also represents $\tau^{\mathcal{M}_2}$ by the same series

$$\tau^{\mathcal{M}_2}(\dot{\boldsymbol{r}}, z) = \sum_{n=0}^{Q-1} A_n(\dot{\boldsymbol{r}}) \left(\frac{z^2}{L_z^2}\right)^n \tag{2}$$

where

$$A_n(\dot{\boldsymbol{r}}) = \begin{cases} \sum_{\ell=\mathcal{M}_1+1}^{M} \frac{w_\ell}{4\pi n!} e^{-\dot{r}^2/s_\ell^2} \left(\frac{6}{s_\ell^2} - \frac{4\dot{r}^2}{s_\ell^4} + 4n\right) \left(\frac{L_z^2}{s_\ell^2}\right)^n, & n = 0, 1, \cdots, Q-2, \\ \sum_{\ell=\mathcal{M}_1+1}^{M} \frac{w_\ell}{\pi(Q-2)!} e^{-\dot{r}^2/s_\ell^2} \left(\frac{L_z^2}{s_\ell^2}\right)^{Q-1}, & n = Q-1, \end{cases}$$

- **Step 1: Gridding.** It can be down hierarchically on $N_x \times N_y \times P$ Uniform-Chebyshev mixing grids

$$H_W^n(\dot{\boldsymbol{r}}, z) = W * f * \left(\frac{z^2}{L_z^2}\right)^n = \sum_{j=1}^{N} q_j W(\dot{\boldsymbol{r}} - \dot{\boldsymbol{r}}_j) \left[\frac{(z-z_j)^2}{L_z^2}\right]^n, \quad n = 0, \cdots, Q-1,$$

- **Step 2: RC2FC.** Hierarchically using 2DFFT to evaluate $\widehat{H}_W^n(\dot{\boldsymbol{k}}, z)$.
- **Step 3: Scaling and Integrating.** Using precomputed coefficients $\widehat{A}_n(\dot{\boldsymbol{k}}) = 2\pi \int_0^\infty J_0(\dot{k}r) A_n(r) r dr$ to calculate

$$\widehat{H}_W(\dot{\boldsymbol{k}}, z) = \sum_{n=0}^{Q-1} \widehat{H}_W^n(\dot{\boldsymbol{k}}, z) \widehat{A}_n(\dot{\boldsymbol{k}})$$

- **Step 4: FC2FCs.** Convert $\widehat{H}_W(\dot{\boldsymbol{k}}, z)$ on Fourier-Chebyshev grids into Fourier-Chebyshev series for solving equation.

Boundary condition

- For the last two conditions, we adopt Dirichlet-type BC at $z = \pm L_z/2$ of

$$\widehat{\Phi}_W^{\mathcal{M}_2}(\boldsymbol{k}, z) = \frac{1}{\widehat{W}(\boldsymbol{k})} \cdot \pi \sum_{\ell=\mathcal{M}_1+1}^{M} w_\ell s_\ell^2 e^{-s_\ell^2 k^2/4} \sum_{j=1}^{N} q_j e^{-\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}_j} e^{-\left(z-z_j\right)^2/s_\ell^2}$$

- One just needs to compute

$$\mathcal{B}_W^\ell(\boldsymbol{r}, \pm L_z/2) = \sum_{j=1}^{N} q_j W(\boldsymbol{r} - \boldsymbol{r}_j)_* \, e^{-\left(\pm L_z/2 - z_j\right)^2/s_\ell^2} \tag{3}$$

and apply 2DFFT to get $\widehat{\mathcal{B}}_W^\ell\left(\boldsymbol{k}, \pm L_z/2\right)$, so that the BC reads

$$\widehat{\Phi}_{\pm L_z/2}^{\mathcal{M}_2}(\boldsymbol{k}) = \pi \sum_{\ell=\mathcal{M}_1+1}^{M} w_\ell s_\ell^2 e^{-s_\ell^2 k^2/4} \widehat{W}^{-2}(\boldsymbol{k}) \widehat{\mathcal{B}}_W^\ell\left(\boldsymbol{k}, \pm L_z/2\right)$$

- **Step 5: SolEqn.** The full system then can be expressed by

$$\begin{pmatrix} \boldsymbol{A} & \boldsymbol{B} \\ \boldsymbol{C} & \boldsymbol{D} \end{pmatrix} \begin{pmatrix} \widehat{\boldsymbol{a}} \\ \widehat{c}_0 \\ \widehat{c}_1 \end{pmatrix} = \begin{pmatrix} \widehat{\boldsymbol{f}} \\ \widehat{\Phi}^{\mathcal{M}_2}_{L_z/2} \\ \widehat{\Phi}^{\mathcal{M}_2}_{-L_z/2} \end{pmatrix}$$

where $\boldsymbol{B}$ is $P \times 2$, $\boldsymbol{C}$ is $2 \times P$, $\boldsymbol{D}$ is 2×2, and $\boldsymbol{A}$ is a $P \times P$ pentadiagonal matrix with only 3 nonzero diagonals. Hence the Schur complement and FEBS method can be applied to efficiently solve

$$\left(\boldsymbol{C}\boldsymbol{A}^{-1}\boldsymbol{B} - \boldsymbol{D}\right) \begin{pmatrix} \widehat{c}_0 \\ \widehat{c}_1 \end{pmatrix} = \boldsymbol{C}\boldsymbol{A}^{-1}\widehat{\boldsymbol{f}} - \begin{pmatrix} \widehat{\Phi}^{\mathcal{M}_2}_{L_z/2} \\ \widehat{\Phi}^{\mathcal{M}_2}_{-L_z/2} \end{pmatrix}$$

and coefficients $\widehat{\boldsymbol{a}} = (\widehat{a}_0, \cdots, \widehat{a}_{P-1})$ can be obtained by back-substitution

$$\widehat{\boldsymbol{a}} = \boldsymbol{A}^{-1}\left(\widehat{\boldsymbol{f}} - \boldsymbol{B}\begin{pmatrix} \widehat{c}_0 \\ \widehat{c}_1 \end{pmatrix}\right)$$

- **Step 6: FCs2RCs.** Using 2DFFT to obtain $\Phi_W^{\mathcal{M}_2}(\dot{\boldsymbol{r}}_j, z)$ (in the form of Chebyshev series).
- **Step 7: Gathering.** Using Plancherel's theorem to gather all contributions (with compact support) into sources by

$$\Phi^{\mathcal{M}_2}(\dot{\boldsymbol{r}}, z) = \int_{\mathcal{R}^2} \Phi_W^{\mathcal{M}_2}(\dot{\boldsymbol{r}}_j, z)\, W(\dot{\boldsymbol{r}} - \dot{\boldsymbol{r}}_j)_*\, d\dot{\boldsymbol{r}}_j$$

Remark

The fast Fourier-Chebyshev solver applies FFT to accelerate the calculation of x, y-direction. Indeed, for isotropic system with $L_x = L_y = L_z$, there are only two possible cases for judging mid-range or long-range Gaussians. As for the long-range Gaussian, the Fourier method only requires a small amount of grids. Therefore, the direct convolution in Fourier-space can also be effective.

FSSOG v.s. Former algorithms

- **TKM+Upsampling**[22].
  - Need a nearly $\sqrt{d}+1$ optimized upsampling factor on FFT/IFFT process. Even worse in $Lz \ll \min\{Lx, Ly\}$ systems.
  - The non-zero mode and zero mode in periodic directions need to be calculated separately in FFT/IFFT part, costing approximatively.
- **Fourier-Chebyshev solver**[23].
  - It only fits for those sources with nearly compact support, since Chebyshev approximation does not have analogous convolution theorem, and hence all interpolation nodes effect on each sources.
- **FSSOG.**
  - No extra zero-padding. Modified by Chebyshev solver to reduce cost.
  - The non-zero mode and zero mode can be disposed uniformly.
  - Better smoothness w.r.t. Ewald splitting. Better energy conservation in NVE ensemble.

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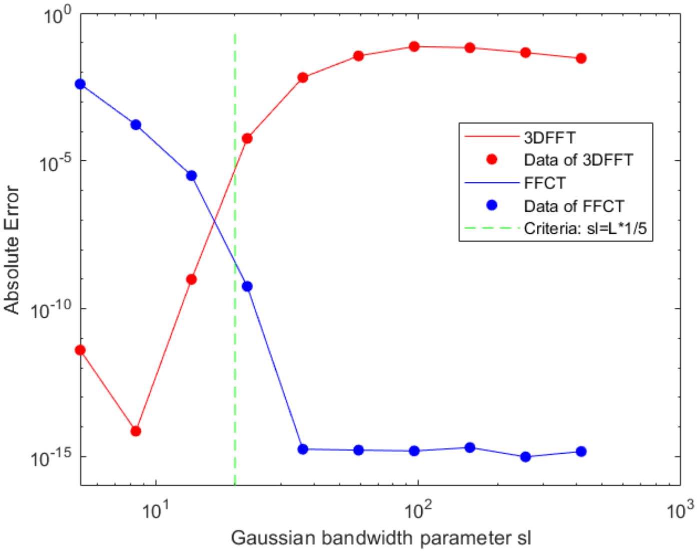
[22] Leslie Greengard, Shidong Jiang, and Yong Zhang. "The anisotropic truncated kernel method for convolution with free-space Green's functions". In: *SIAM J. Sci. Comput.* 40.6 (2018), A3733–A3754.

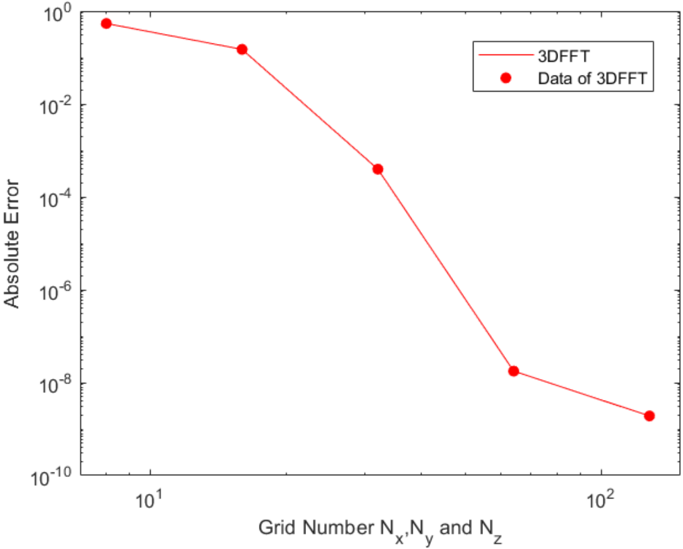
[23] Ondrej Maxian et al. "A fast spectral method for electrostatics in doubly periodic slit channels". In: *J. Chem. Phys.* 154.20 (2021), p. 204107.

Content

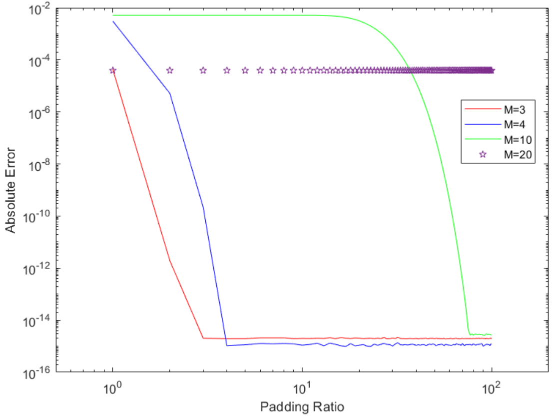
Importance of dual splitting



FSSOG Spectral convergence

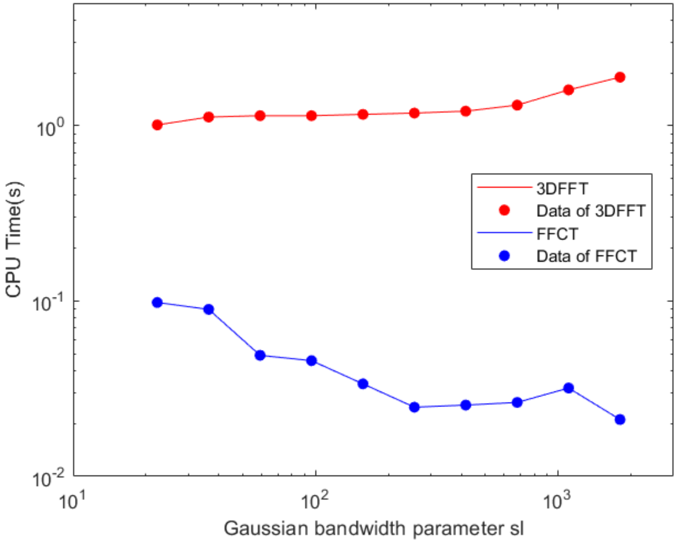


Inefficient of zero-padding 3DFFT



Gaussian Term	3	4	5	6	7	8	9	10	11	12
Zero-padding ratio	0.4	1.2	2.8	5.4	10	16	29	49	79	139

Time consumption: zero-padding V.S. FFCT



Content

Conclusion

- We develop an efficient and accurate FSSOG method for solving quasi-2D systems, which is based on u-series splitting and dual splitting of far-field potentials.
- The FSSOG has rigorous mathematical foundation for achieving the given accuracy, and it can provide a prior parameter selection without the information of particles.
- **Future works:** Continue to give more numerical examples to illustrate the high efficiency of FSSOG; Finish the article from FSSOG note; Design the random batch SOG2D method based on this fast spectral method.

Thank you for listening!