PSINC FUNCTIONS & FFT BOXES

Arash A. Mostofi

The Thomas Young Centre for Theory and Simulation of Materials

Departments of Physics and Materials, Imperial College London

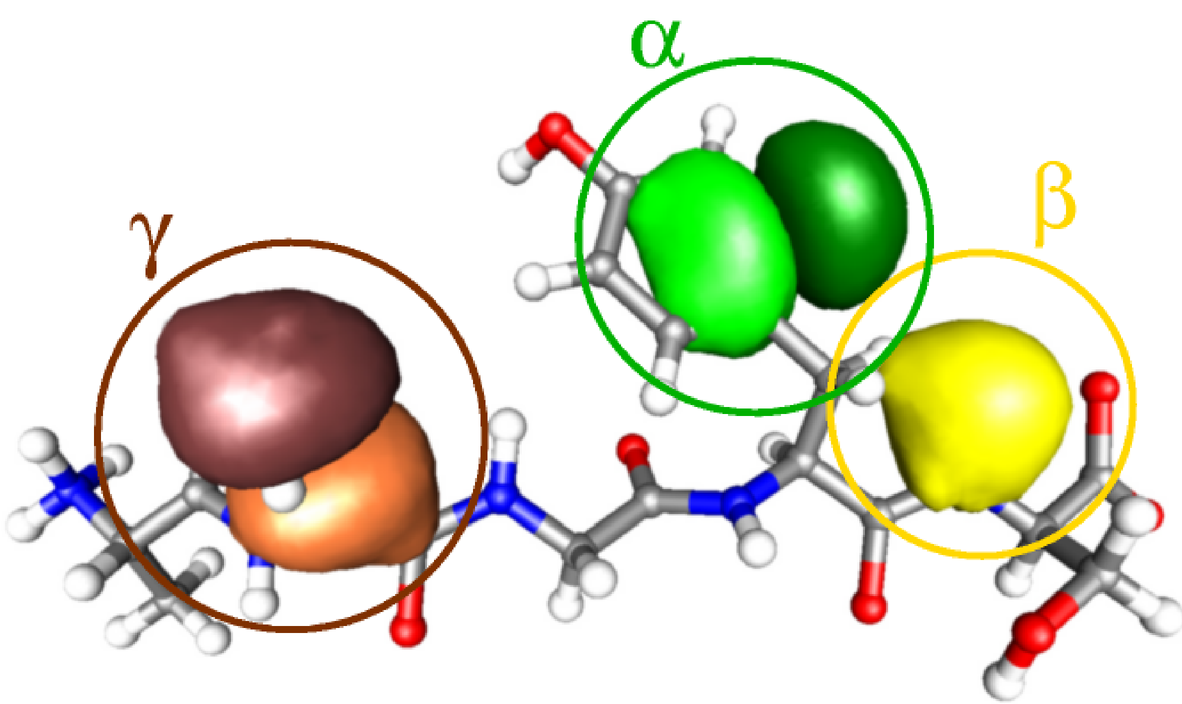
Overview

- Basis Sets for Linear-Scaling Methods
- Grids and Fast Fourier Transforms (FFT)
- The ONETEP Basis – Psinc functions
  - Definition & Properties
- Total Energy: O($N^2 \log N$)
- The FFT Box
- Total Energy: O(N)

Localisation

$$\rho(\mathbf{r},\mathbf{r}') = \sum_{\alpha\beta} \phi_\alpha(\mathbf{r}) K^{\alpha\beta} \phi^*_\beta(\mathbf{r}')$$

Impose spatial cut-offs on orbitals (NGWFs)



Degrees of Freedom

$$\rho(\mathbf{r},\mathbf{r}') = \sum_{\alpha\beta} \phi_\alpha(\mathbf{r}) K^{\alpha\beta} \phi^*_\beta(\mathbf{r}')$$

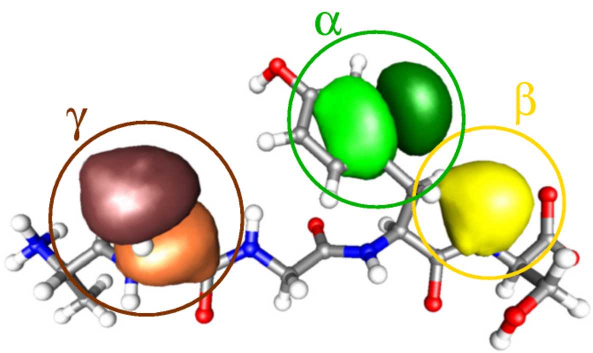
$$E = E[\rho] = E\left[\mathbf{K}, \{\phi\}\right]$$

- Minimise E only wrt $\mathbf{K}$, keeping $\{\phi\}$ fixed
  - Need many NGWFs per atom for good accuracy
  - Large matrices

- Minimise E wrt $\mathbf{K}$ and $\{\phi\}$
  - Minimal number of NGWFs per atom
  - Better accuracy, but more work

Localised Basis

$$\phi_\alpha(\mathbf{r}) = \sum_i D_i(\mathbf{r}) C_{i\alpha}$$



- $\{C_{i\alpha}\}$ are expansion coefficients of $\phi_\alpha(\mathbf{r})$ in basis $D_i(\mathbf{r})$
- $\phi_\alpha(\mathbf{r})$ localised ⇨ convenient if $D_i(\mathbf{r})$ also localised
- Plenty of choices
  - Gaussians, wavelets, spherical waves, splines, finite elements and grids

Comment About Plane-Waves

- Pros
  - $e^{i\mathbf{G}\cdot\mathbf{r}} = \cos(\mathbf{G}\cdot\mathbf{r}) + i\sin(\mathbf{G}\cdot\mathbf{r})$
  - Systematically controllable accuracy: resolution determined by $\mathbf{G}_{max}$
  - Fourier basis: efficient FFTs to switch between real and reciprocal space
  - No pulay forces

- Cons
  - Delocalised
  - Uniform resolution in all space: one pays for vacuum

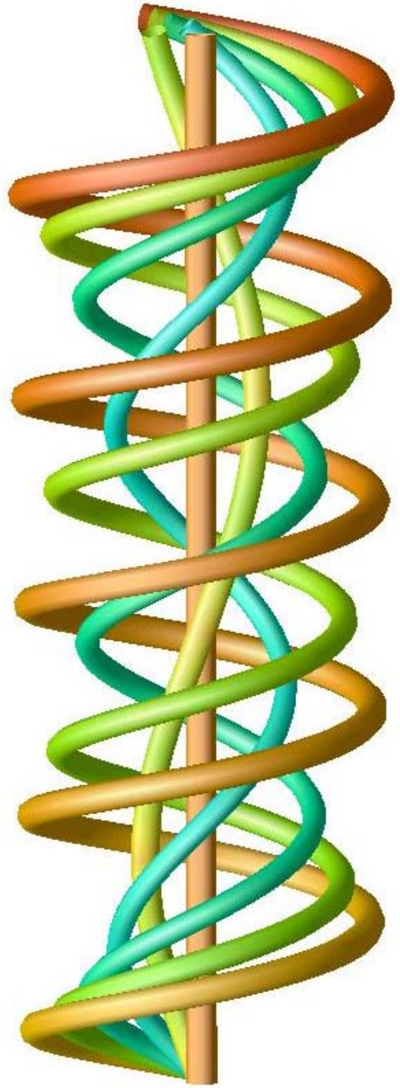


Image courtesy of Dr. Ismaila Dabo
(École Nationale des Ponts et Chaussées, Paris)

Position vs momentum

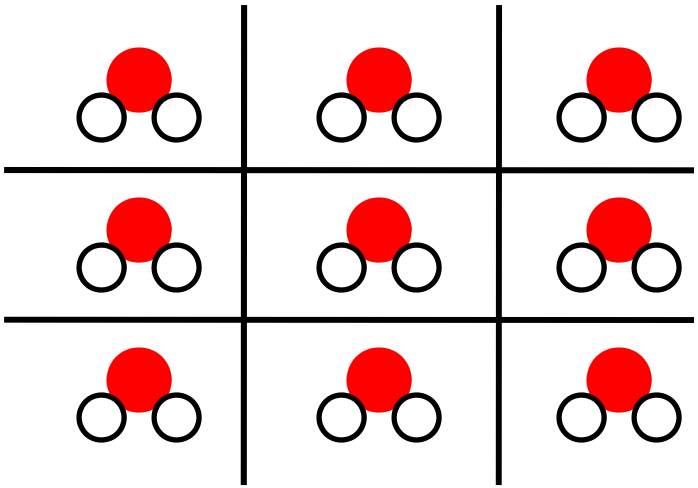
- Hamiltonian:

$$\hat{H}_{\mathrm{KS}} = -\tfrac{1}{2}\nabla^2 + V_{\mathrm{eff}}[n](\mathbf{r})$$

$$n(\mathbf{r}) = \sum_n f_n \left|\psi_n(\mathbf{r})\right|^2$$

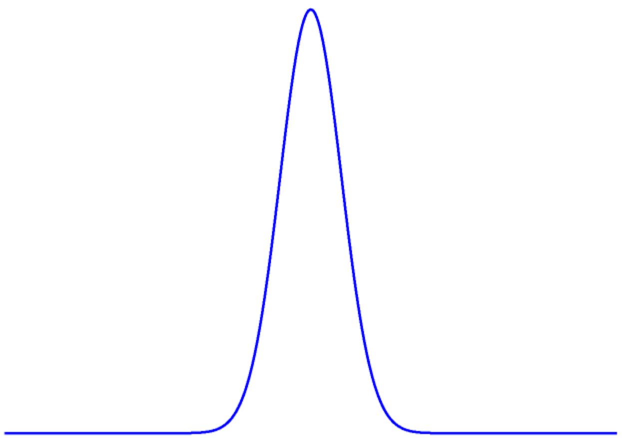
- Bloch + periodic boundary conditions:

$$\begin{aligned} \psi(\mathbf{r}+\mathbf{R}) &= \mathrm{e}^{\mathrm{i}\mathbf{k}\cdot\mathbf{R}}\,\psi(\mathbf{r}) \\ \Rightarrow \psi(\mathbf{r}) &= \sum_{\mathbf{G}} c_{\mathbf{G}}\,\mathrm{e}^{\mathrm{i}(\mathbf{k}+\mathbf{G})\cdot\mathbf{r}} \end{aligned}$$

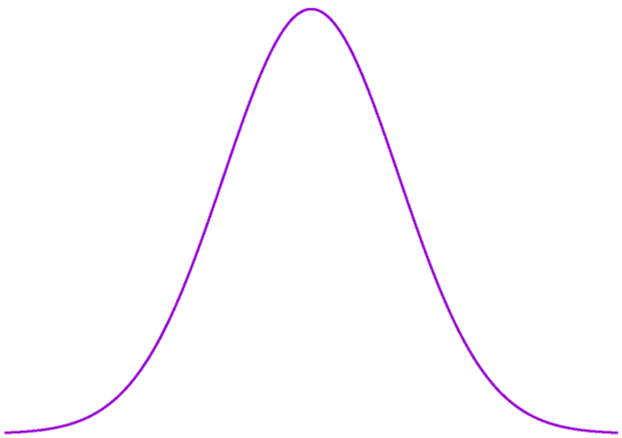


- Fast Fourier transforms

Fourier transforms



$\rightleftarrows$



$$f(x) = \frac{1}{2\pi}\int_{-\infty}^{\infty} dk\, \tilde{f}(k)\, \mathrm{e}^{\mathrm{i}kx}$$

$$\tilde{f}(k) = \int_{-\infty}^{\infty} dk\, f(x)\, \mathrm{e}^{-\mathrm{i}kx}$$

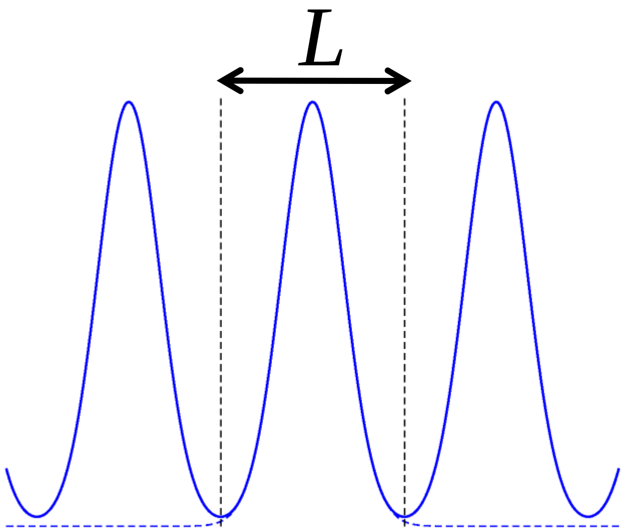
Infinite domain

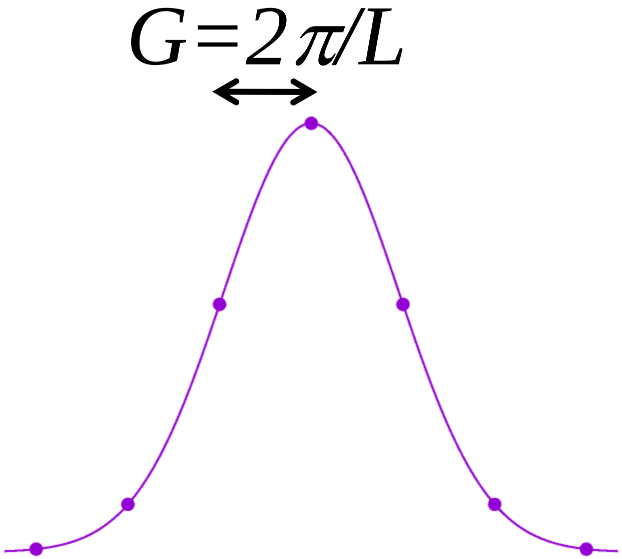
Continuous

Infinite domain

Continuous

Fourier transforms



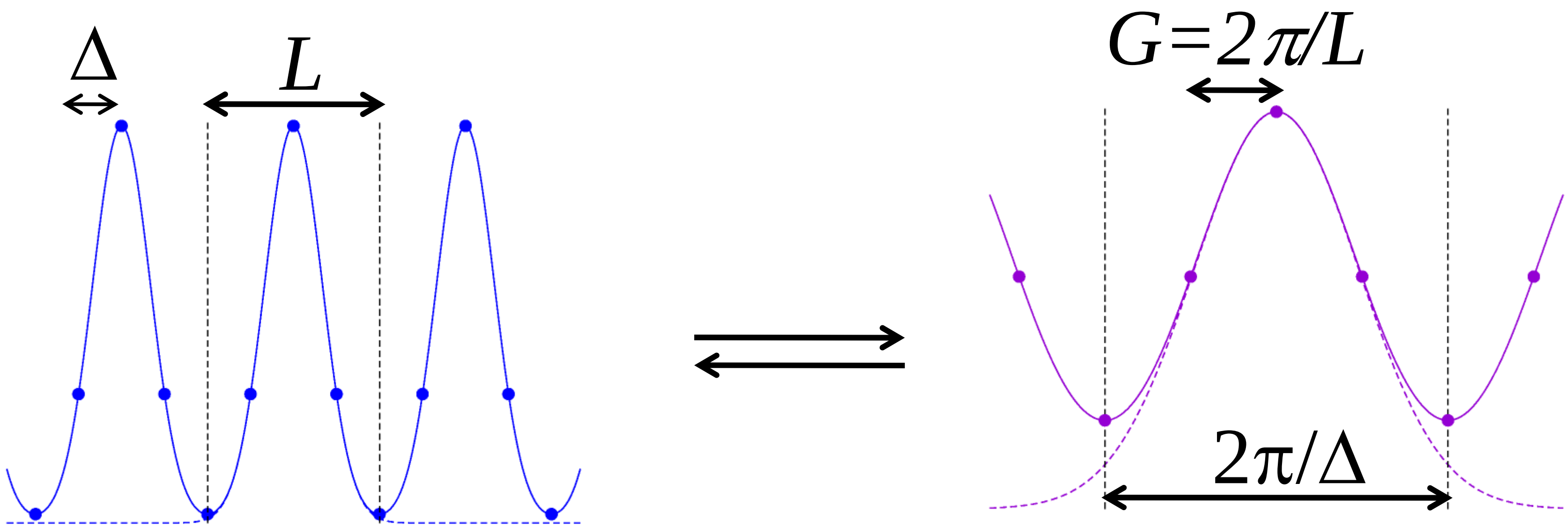


$$f(x) = \frac{1}{L} \sum_{n=-\infty}^{\infty} \tilde{f}_n \, \mathrm{e}^{\mathrm{i}nGx}$$

$$\tilde{f}_n = \int_0^L f(x) \, \mathrm{e}^{-\mathrm{i}nGx}$$

Periodic	Infinite domain
Continuous	Discrete

Fourier transforms

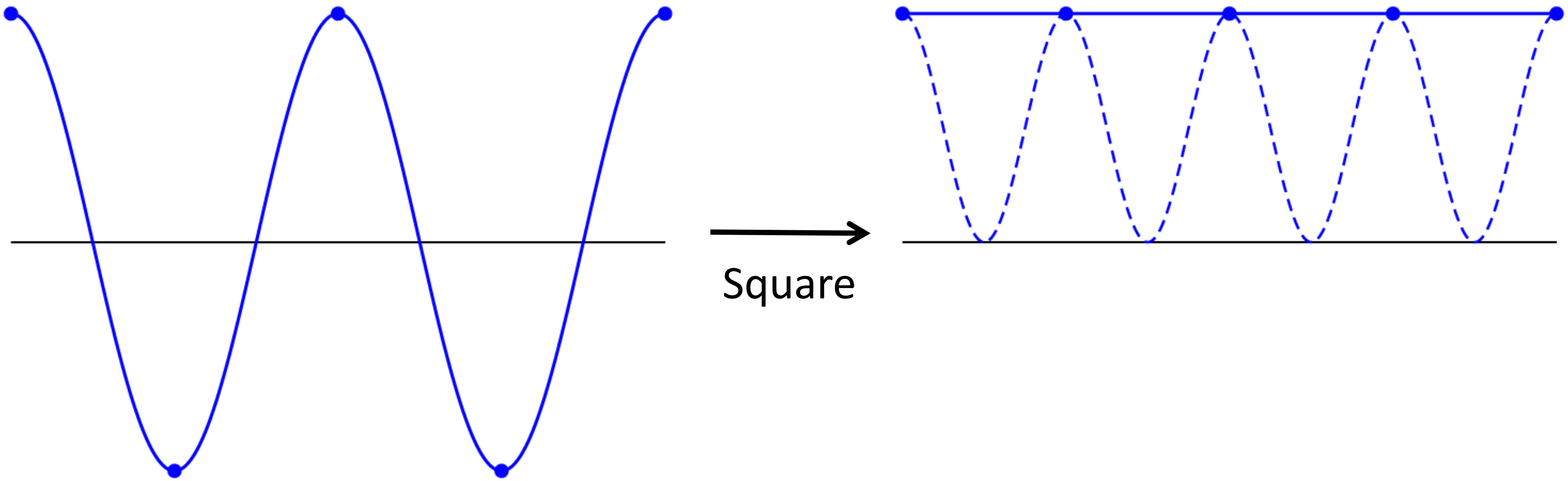


$$f_m = \frac{1}{N}\sum_{n=0}^{N-1} \tilde{f}_n \, \mathrm{e}^{\mathrm{i}nmG\Delta}$$

$$\tilde{f}_n = \sum_{m=0}^{N-1} f_m \, \mathrm{e}^{-\mathrm{i}nmG\Delta}$$

Periodic	Periodic
Discrete	Discrete

Aliasing

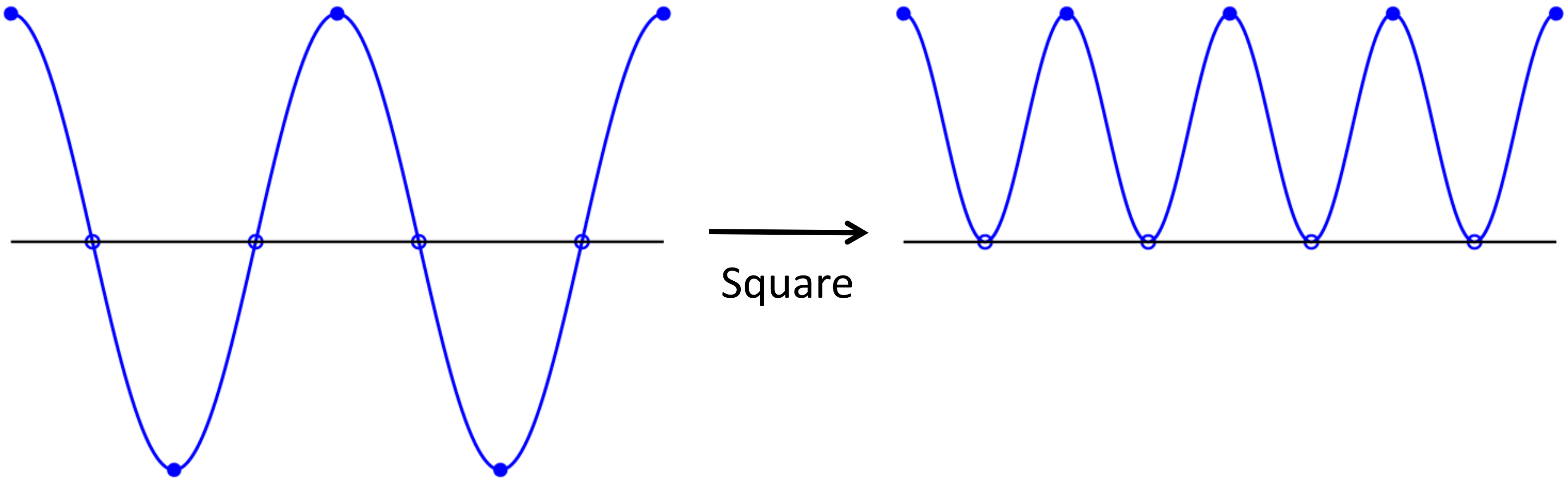


$$\cos(\pi x) \longrightarrow \cos^2(\pi x) = \frac{1}{2} + \frac{1}{2}\cos(2\pi x)$$

sampled at Nyquist frequency

aliased to unity

Aliasing



$$\cos(\pi x) \longrightarrow \cos^2(\pi x) = \frac{1}{2} + \frac{1}{2}\cos(2\pi x)$$

interpolated to twice Nyquist frequency

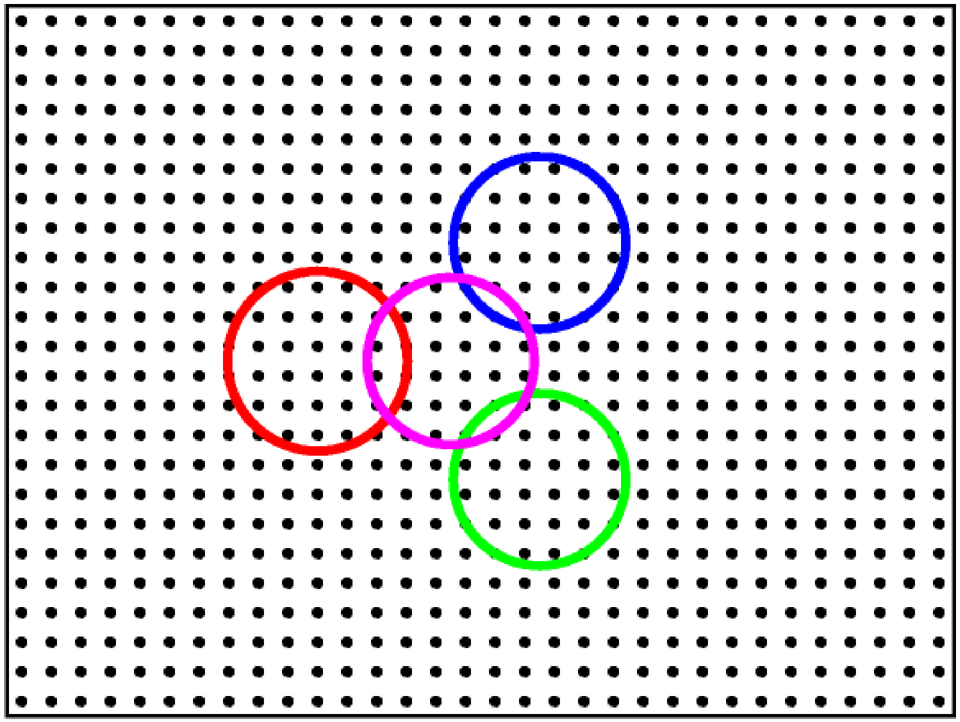
no aliasing

Real-Space Grid

$$\phi_\alpha(\mathbf{r}) = \sum_i D_i(\mathbf{r}) C_{i\alpha}$$

- Represent ϕ as values on a regular real-space grid
- Can choose our basis D_i to be the grid-points themselves, ie, a set of Dirac delta-functions

$$\delta(\mathbf{r}) = \int \frac{\mathrm{d}\mathbf{G}}{(2\pi)^3} \mathrm{e}^{\mathrm{i}\mathbf{G}\cdot\mathbf{r}}$$



Implicit Basis

$$\delta(\mathbf{r}) = \int \frac{\mathrm{d}\mathbf{G}}{(2\pi)^3} \mathrm{e}^{\mathrm{i}\mathbf{G}\cdot\mathbf{r}}$$

- Grid points are not δ-functions
- Periodicity of unit cell ⇨ discrete $\mathbf{G}$-vectors
- Grid ⇨ $\mathbf{G}_{\text{max}}$ (maximum representable wavevector)

⇩

$$D(\mathbf{r}) = \frac{1}{N} \sum_{\mathbf{G}}^{\mathbf{G}_{\text{max}}} \mathrm{e}^{\mathrm{i}\mathbf{G}\cdot\mathbf{r}}$$

psinc function

Psinc Functions: Definition (i)

$$D_m(\mathbf{r}) \equiv D(\mathbf{r} - \mathbf{r}_m)$$
$$= \frac{1}{N_1 N_2 N_3} \sum_{p_1=-J_1}^{J_1} \sum_{p_2=-J_2}^{J_2} \sum_{p_3=-J_3}^{J_3} \mathrm{e}^{\mathrm{i}(p_1\mathbf{B}_1 + p_2\mathbf{B}_2 + p_3\mathbf{B}_3)\cdot(\mathbf{r}-\mathbf{r}_m)},$$

where the reciprocal lattice vectors $\{\mathbf{B}_i\}$ are

$$\mathbf{B}_1 = \frac{2\pi}{V}(\mathbf{A}_2 \times \mathbf{A}_3), \quad \text{etc.},$$

satisfying orthogonality relations

$$\mathbf{B}_i \cdot \mathbf{A}_j = 2\pi\delta_{ij},$$

and $\{\mathbf{r}_m\}$ are grid points of the simulation cell,

$$\mathbf{r}_m = \frac{m_1}{N_1}\mathbf{A}_1 + \frac{m_2}{N_2}\mathbf{A}_2 + \frac{m_3}{N_3}\mathbf{A}_3 = \sum_{i=1}^{3} \frac{m_i}{N_i}\mathbf{A}_i, \qquad \mathbf{r} = \sum_{i=1}^{3} \frac{\xi_i}{N_i}\mathbf{A}_i, \quad \xi_i \in \mathbb{R}.$$

Psinc Functions: Definition (ii)

By orthogonality of {**A**} and {**B**}:

$$D_m(\mathbf{r}) = \mathcal{D}^{(1)}_{m_1}(\xi_1)\mathcal{D}^{(2)}_{m_2}(\xi_2)\mathcal{D}^{(3)}_{m_3}(\xi_3)$$

$$\mathcal{D}^{(i)}(\xi) = \frac{1}{N_i}\sum_{p=-J_i}^{J_i} \mathrm{e}^{2\mathrm{i}\pi p\xi/N_i}$$

This is a geometric sum, first term *a* and common ratio *r*:

$$a = \frac{1}{N_i}\mathrm{e}^{-\mathrm{i}\pi\xi(1-1/N_i)}, \quad r = \mathrm{e}^{2\mathrm{i}\pi\xi/N_i}$$

$$\mathcal{D}^{(i)}(\xi) = \frac{a(1-r^{N_i})}{1-r} = \frac{1}{N_i}\frac{\sin(\pi\xi)}{\sin(\pi\xi/N_i)}$$

Psinc vs Sinc (i)

$$\mathcal{D}^{(i)}(\xi) = \frac{1}{N_i} \frac{\sin(\pi\xi)}{\sin(\pi\xi/N_i)}$$

Consider the limit of a psinc function as $N_i \to \infty$

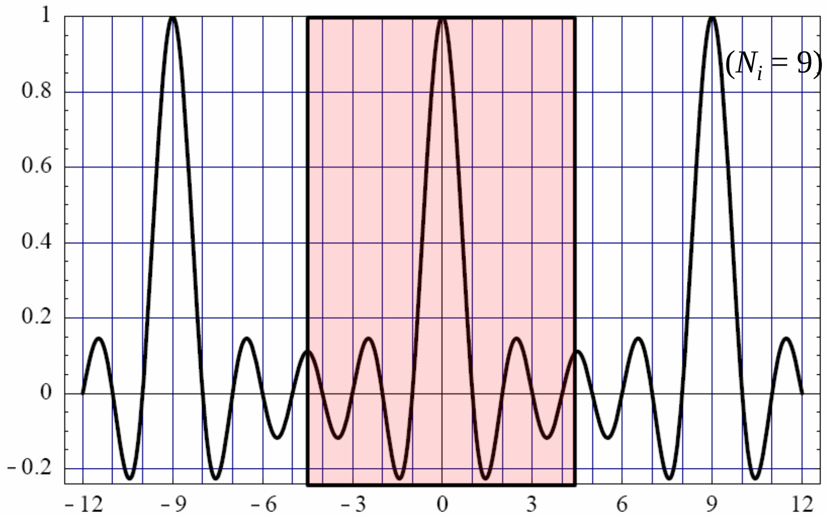
$$\begin{aligned} \mathcal{S}(\xi) &\equiv \lim_{N_i \to \infty} \mathcal{D}^{(i)}(\xi) \\ &= \lim_{N_i \to \infty} \frac{1}{N_i} \frac{\sin(\pi\xi)}{\sin(\pi\xi/N_i)} \\ &= \frac{\sin(\pi\xi)}{(\pi\xi)} = \operatorname{sinc}(\pi\xi), \end{aligned}$$

Cardinal sine (sinc) function

Psinc vs Sinc (ii)

$$\mathcal{D}^{(i)}(\xi) = \frac{1}{N_i} \sum_{p=-J_i}^{J_i} e^{2i\pi p\xi/N_i}$$

Periodic cardinal sine (sinc)



$$\mathcal{S}(\xi) = \frac{1}{2\pi} \int_{-\pi}^{\pi} dk\, e^{ik\xi}$$

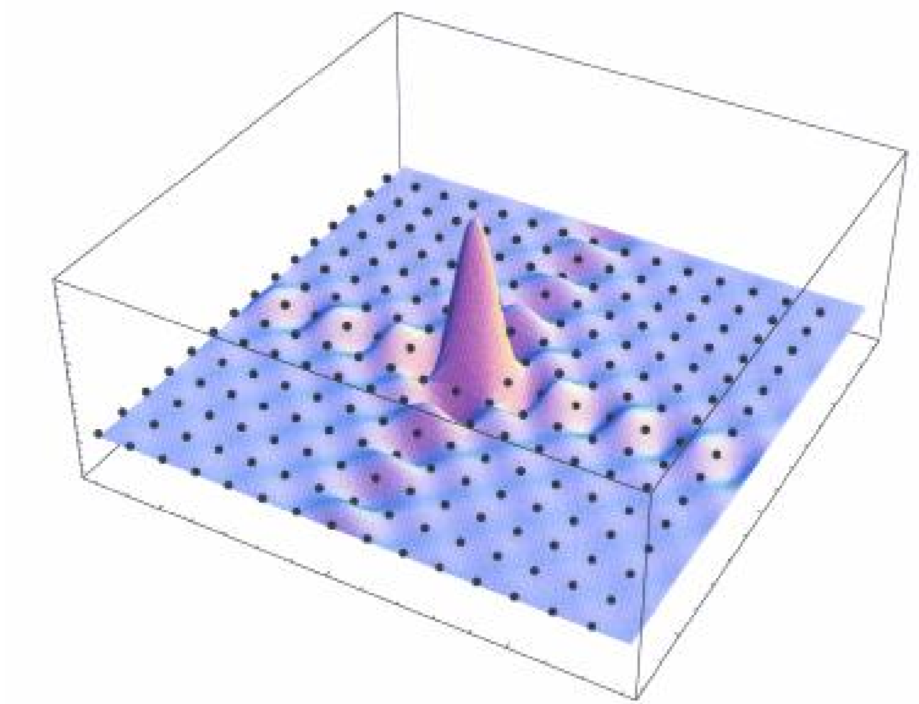
Cardinal sine (sinc)

Psinc Basis Set

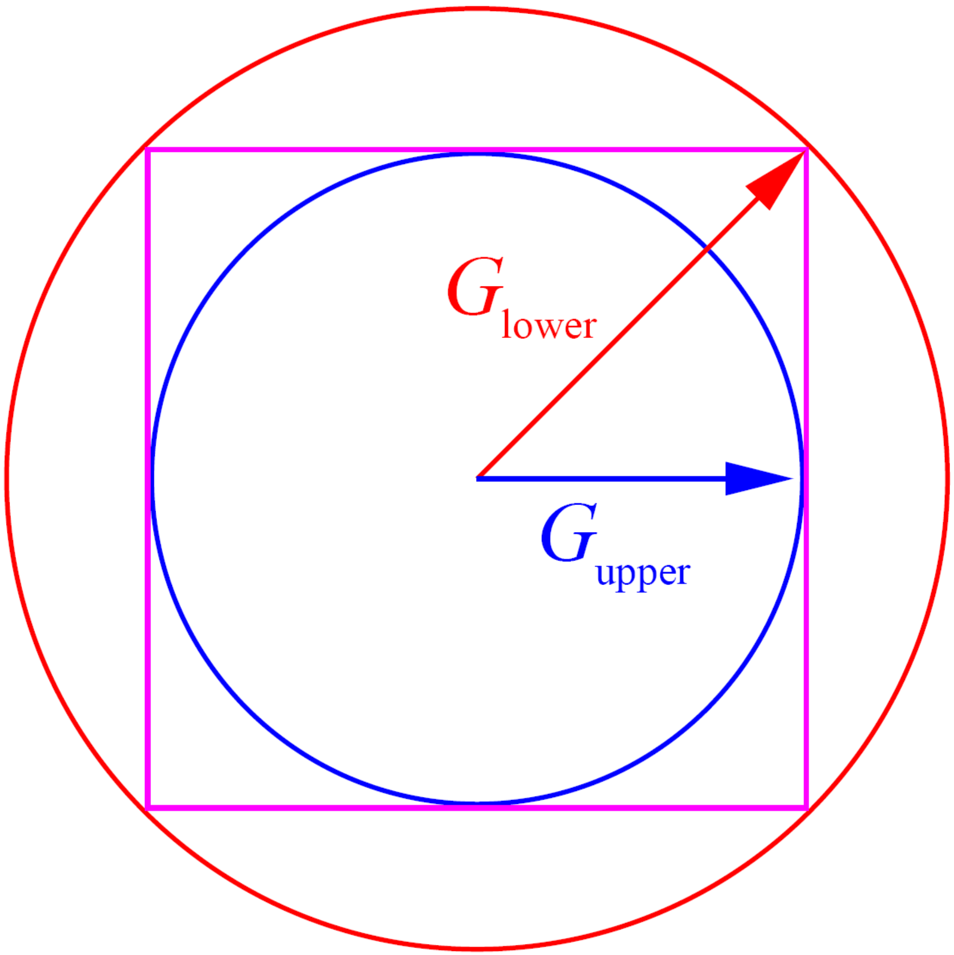
- Localised: $D_i(\mathbf{r}_j) = \delta_{ij}$
- Orthogonal: $\langle D_i | D_j \rangle \propto \delta_{ij}$
- Real
- Fixed in space
- Equivalent to plane-waves

$$\phi_\alpha(\mathbf{r}) = \sum_i D_i(\mathbf{r}) C_{i\alpha}$$

One PSINC on each gridpoint ***i*** of a regular real-space grid



$$E_{\mathrm{PW}}(\mathcal{E}_{\mathrm{cut}} = \frac{1}{2}G^2_{\mathrm{lower}}) < E_{\mathrm{ONETEP}} < E_{\mathrm{PW}}(\mathcal{E}_{\mathrm{cut}} = \frac{1}{2}G^2_{\mathrm{upper}})$$



Localisation

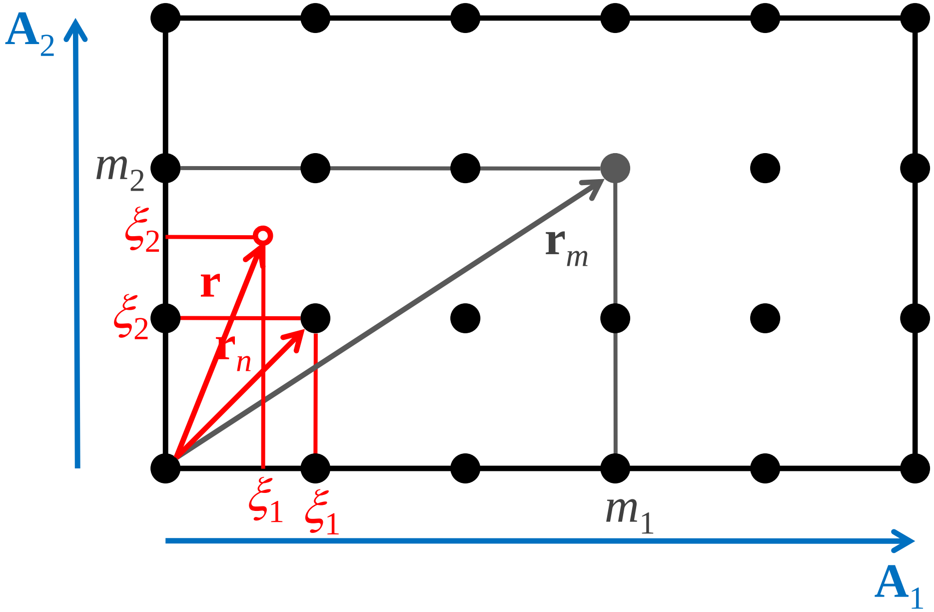
$$D_m(\mathbf{r}) = \mathcal{D}^{(1)}_{m_1}(\xi_1)\mathcal{D}^{(2)}_{m_2}(\xi_2)\mathcal{D}^{(3)}_{m_3}(\xi_3)$$

$$\mathcal{D}^{(i)}(\xi) = \frac{1}{N_i}\sum_{p=-J_i}^{J_i} \mathrm{e}^{2\mathrm{i}\pi p\xi/N_i}$$

$$D_m(\mathbf{r}_n) = D(\mathbf{r}_n - \mathbf{r}_m)$$

$$\mathbf{r}_n - \mathbf{r}_m = \sum_{i=1}^{3} \frac{l^{nm}}{N_i}\mathbf{A}_i, \qquad l_i^{nm} \in \mathbb{Z}$$

$$\mathcal{D}^{(i)}(l) = \frac{1}{N_i}\sum_{p=-J_i}^{J_i} \mathrm{e}^{2\mathrm{i}\pi pl/N_i} = \begin{cases} 1 & \text{if } l = 0, \pm N_i, \pm 2N_i, \ldots \\ 0 & \text{otherwise} \end{cases}$$



$$D_m(\mathbf{r}_n) = \delta_{nm}$$

Orthogonality

$$D(\mathbf{r}) = \frac{1}{N} \sum_{\mathbf{G}}^{\mathbf{G}_{\max}} \mathrm{e}^{\mathrm{i}\mathbf{G}\cdot\mathbf{r}}$$

$$\begin{aligned}
s_{ij} &= \int_V \mathrm{d}\mathbf{r}\, D_i^*(\mathbf{r}) D_j(\mathbf{r}) = \langle D_i | D_j \rangle \\
&= \frac{1}{N^2} \sum_{\mathbf{G}_p}^{\max} \sum_{\mathbf{G}_q}^{\max} \mathrm{e}^{\mathrm{i}\mathbf{G}_p\cdot\mathbf{r}_i - \mathrm{i}\mathbf{G}_q\cdot\mathbf{r}_j} \boxed{\int_V \mathrm{d}\mathbf{r}\, \mathrm{e}^{\mathrm{i}(\mathbf{G}_q - \mathbf{G}_p)\cdot\mathbf{r}}} \rightarrow V\,\delta_{pq} \\
&= \frac{V}{N^2} \sum_{\mathbf{G}_p}^{\max} \sum_{\mathbf{G}_q}^{\max} \mathrm{e}^{\mathrm{i}(\mathbf{G}_p\cdot\mathbf{r}_i - \mathbf{G}_q\cdot\mathbf{r}_j)} \delta_{pq} \\
&= \frac{V}{N^2} \boxed{\sum_{\mathbf{G}_p}^{\max} \mathrm{e}^{\mathrm{i}\mathbf{G}_p\cdot(\mathbf{r}_i - \mathbf{r}_j)}} \rightarrow N D_j(\mathbf{r}_i) = N\delta_{ij} \\
&= w\delta_{ij}, \qquad w = V/N
\end{aligned}$$

Integrals

- Consider two cell periodic functions:

$$f(\mathbf{r}) = \frac{1}{V} \sum_{\mathbf{G}_p}^{\infty} \tilde{f}(\mathbf{G}_p) \mathrm{e}^{\mathrm{i}\mathbf{G}_p \cdot \mathbf{r}} \qquad f_D(\mathbf{r}) = \frac{1}{V} \sum_{\mathbf{G}_p}^{\max} \tilde{f}(\mathbf{G}_p) \mathrm{e}^{\mathrm{i}\mathbf{G}_p \cdot \mathrm{r}}$$

- Using orthogonality, can show that projection onto a psinc function is

$$\int_V \mathrm{d}\mathbf{r}\; f^*(\mathbf{r}) D_i(\mathbf{r}) = w f_D^*(\mathbf{r}_i) = \int_V \mathrm{d}\mathbf{r}\; f_D^*(\mathbf{r}) D_i(\mathbf{r})$$

- Very useful: overlap between $f(\mathbf{r})$ and a function represented in the psinc basis can be evaluated *exactly* as a sum over the grid

Calculating the Total Energy

$$\hat{H} = -\frac{1}{2}\nabla^2 + \int d^3r' \frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} + \sum_I V_I^{\mathrm{ps,loc}}(\mathbf{r}) + \sum_I V_I^{\mathrm{ps,nl}}(\mathbf{r}) + V^{\mathrm{xc}}(\mathbf{r})$$

$$H_{\alpha\beta} = \langle \phi_\alpha | \hat{H} | \phi_\beta \rangle$$

- $E = \mathrm{Tr}[\mathbf{KH}] - E_{\mathrm{DC}}$

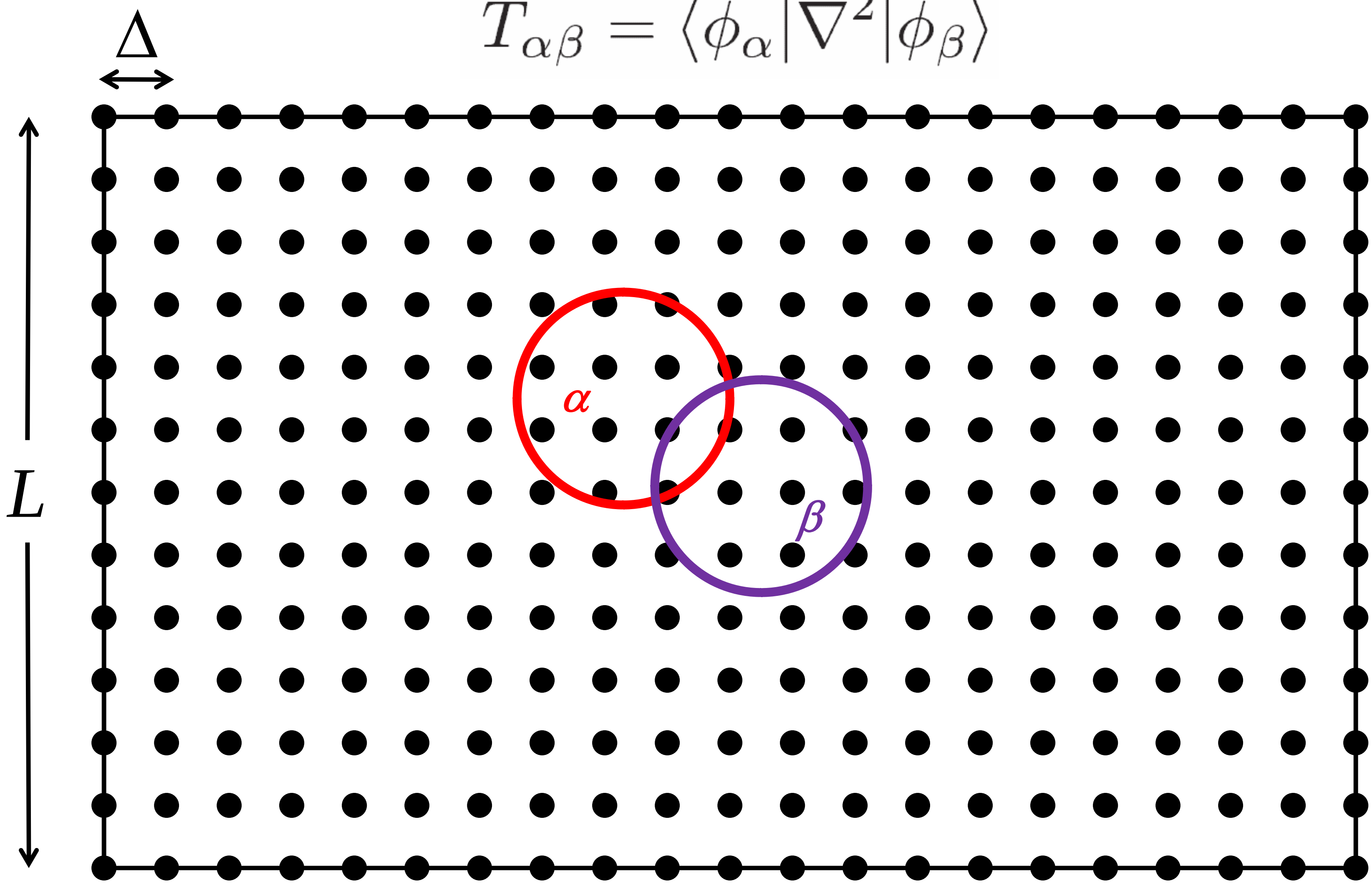
Kinetic

$$T_{\alpha\beta} = \langle \phi_\alpha | \nabla^2 | \phi_\beta \rangle$$

- Fourier transform $\phi_\beta(\mathbf{r}) \rightarrow \tilde{\phi}_\beta(\mathbf{G})$
- Apply Laplacian in reciprocal space: $-\mathbf{G}^2 \tilde{\phi}_\beta(\mathbf{G})$
- Fourier Transform back: $\nabla^2 \phi_\beta(\mathbf{r})$
- Calculate dot product with $\phi_\alpha(\mathbf{r})$ on the grid

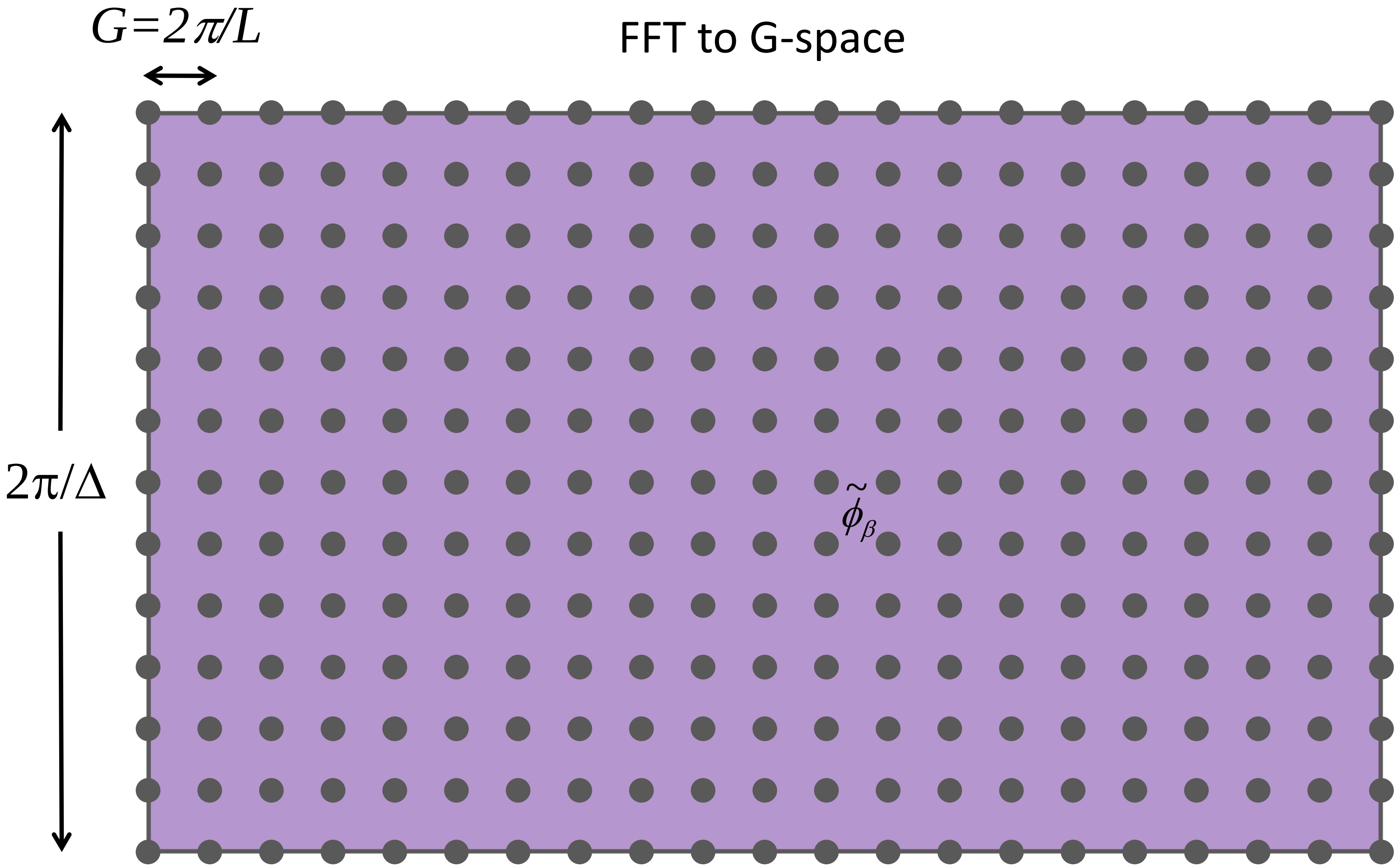
Kinetic

$$T_{\alpha\beta} = \langle \phi_\alpha | \nabla^2 | \phi_\beta \rangle$$



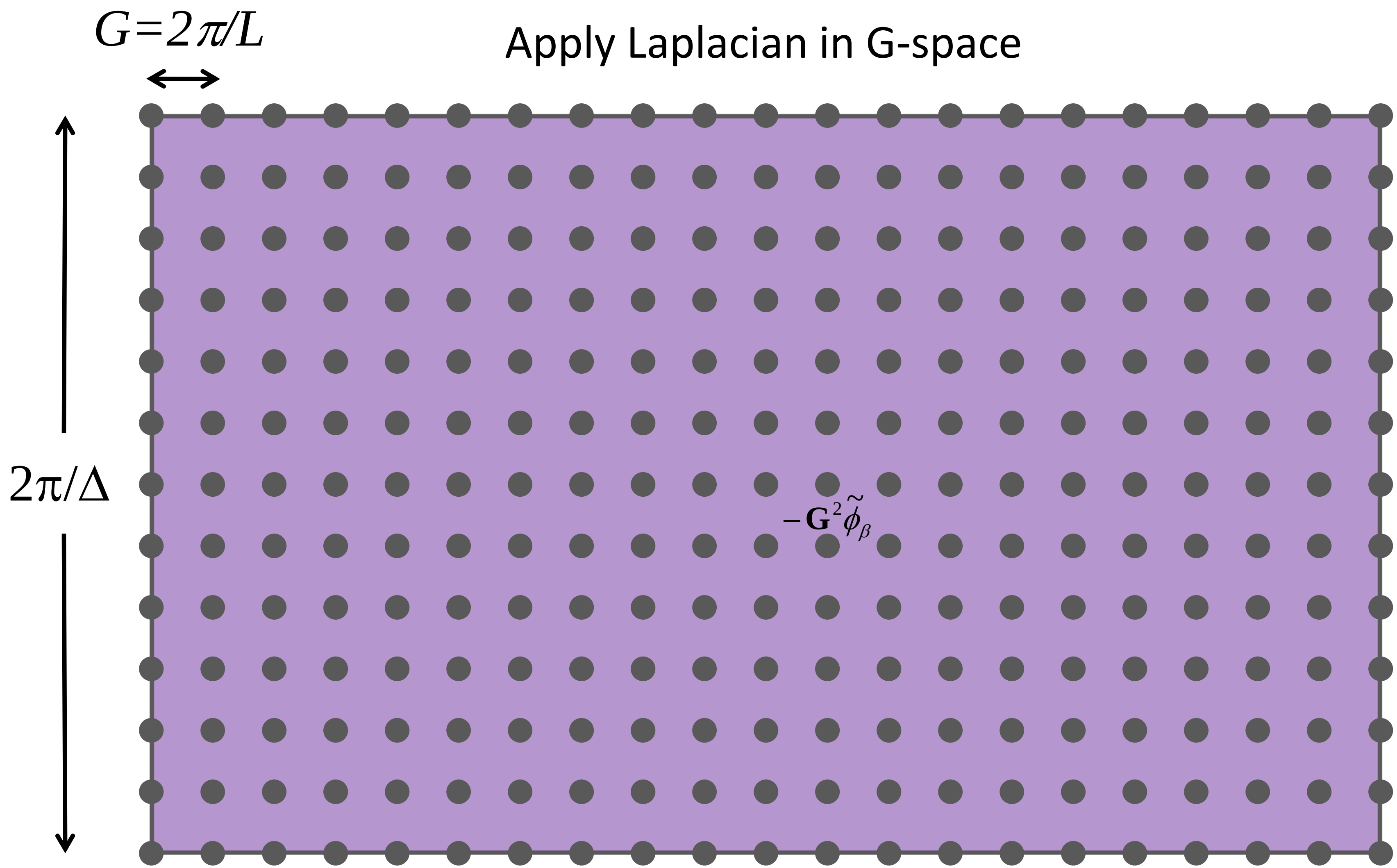
Kinetic

FFT to G-space



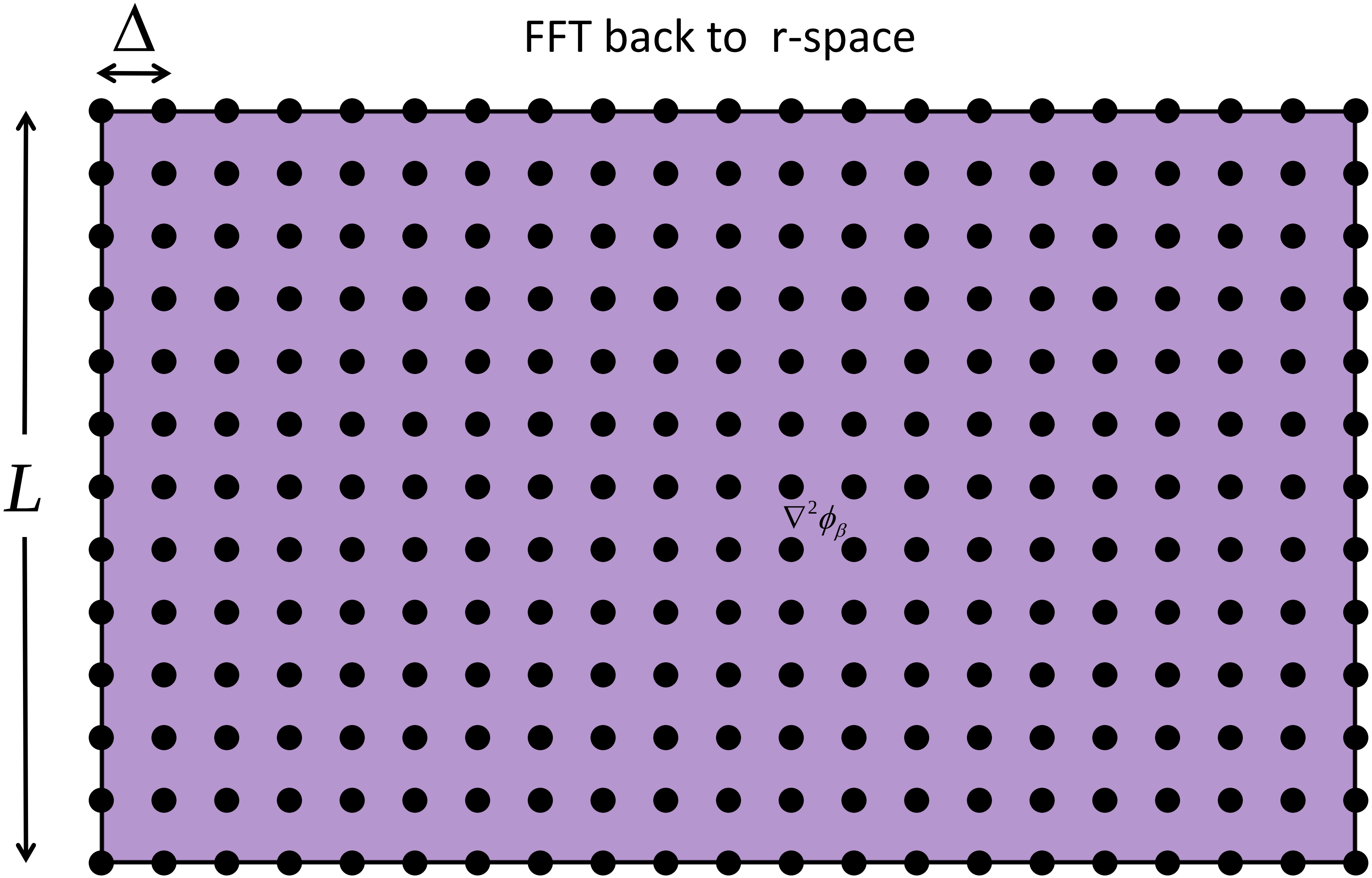
Kinetic

Apply Laplacian in G-space



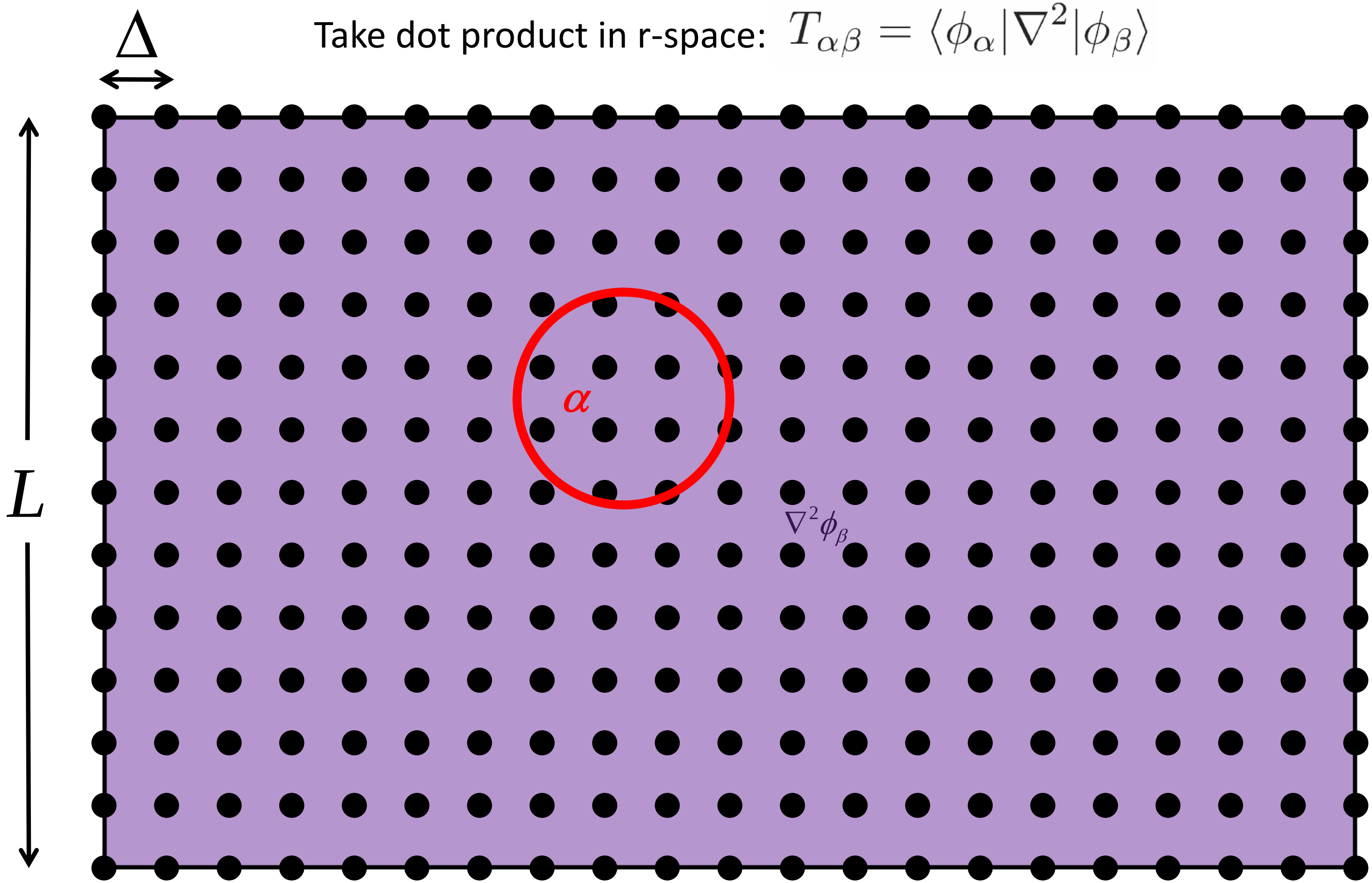
Kinetic

FFT back to r-space



Kinetic

Take dot product in r-space: $T_{\alpha\beta} = \langle\phi_\alpha|\nabla^2|\phi_\beta\rangle$



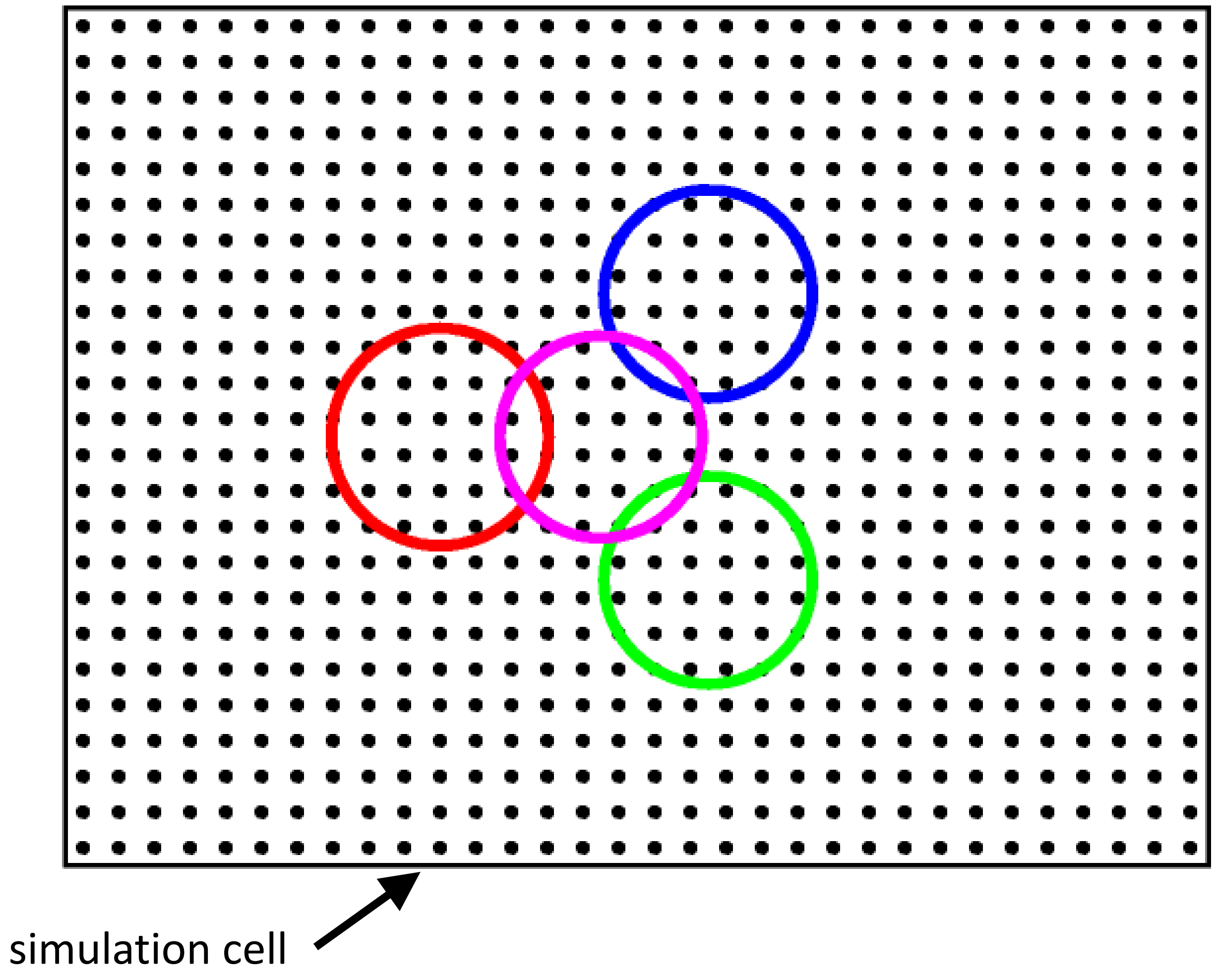
Computational Cost

- For each $T_{\alpha\beta}$ require 2 FFTs on whole grid
- $O(N \log N)$ per element
- There are $O(N)$ elements
- Overall cost $O(N^2 \log N)$
- Not linear-scaling!

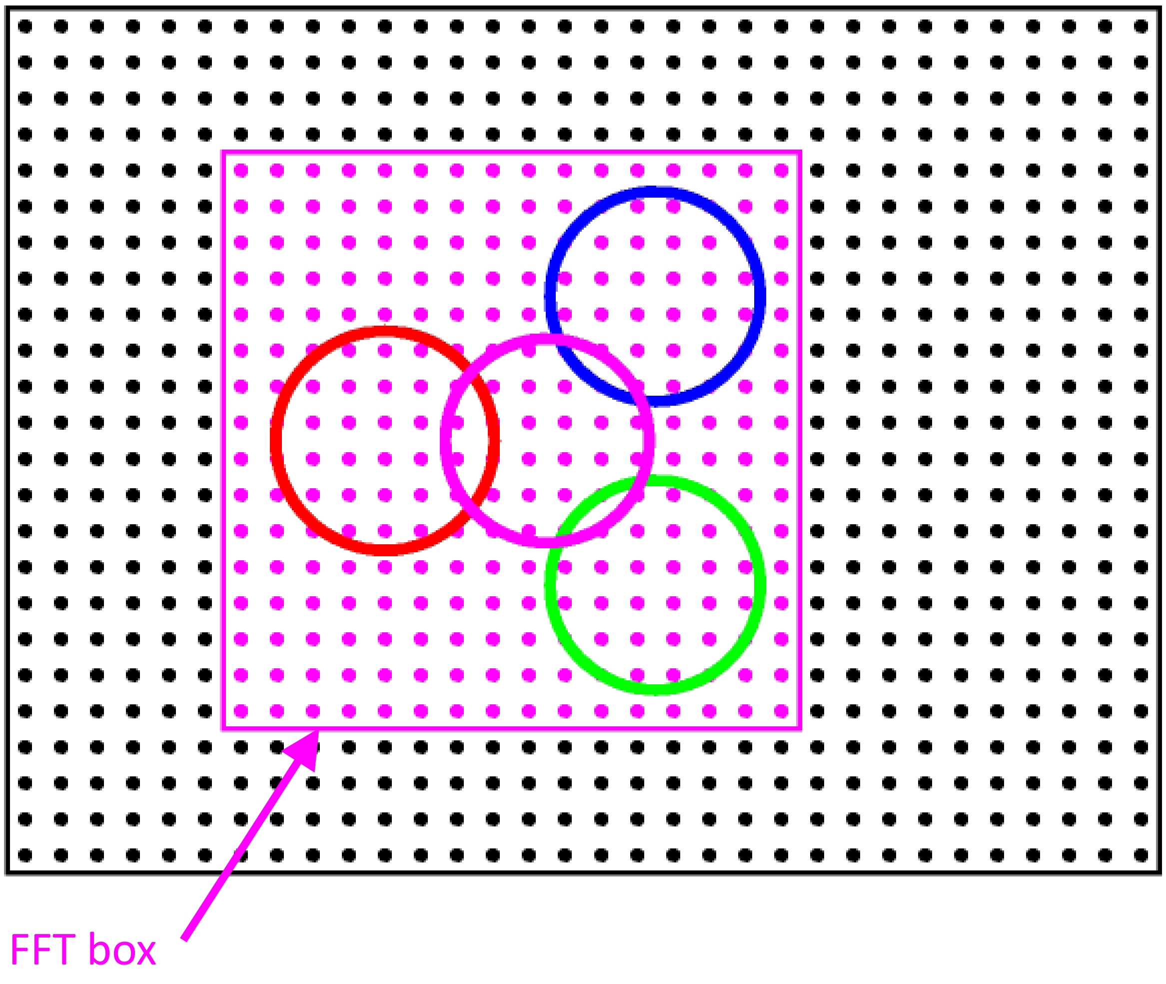
Exploit Localisation

- Each NGWF is only non-zero within a well-defined region
- Most of the effort goes into FFTing strings of zeros!
- FFT boxes...

FFT Box



FFT Box



FFT Box: Definition

- Miniature version of simulation cell
- Same grid spacing and shape
- Origin always coincides with a grid-point of the simulation cell
- Universal shape and size

Hermiticity and Consistency

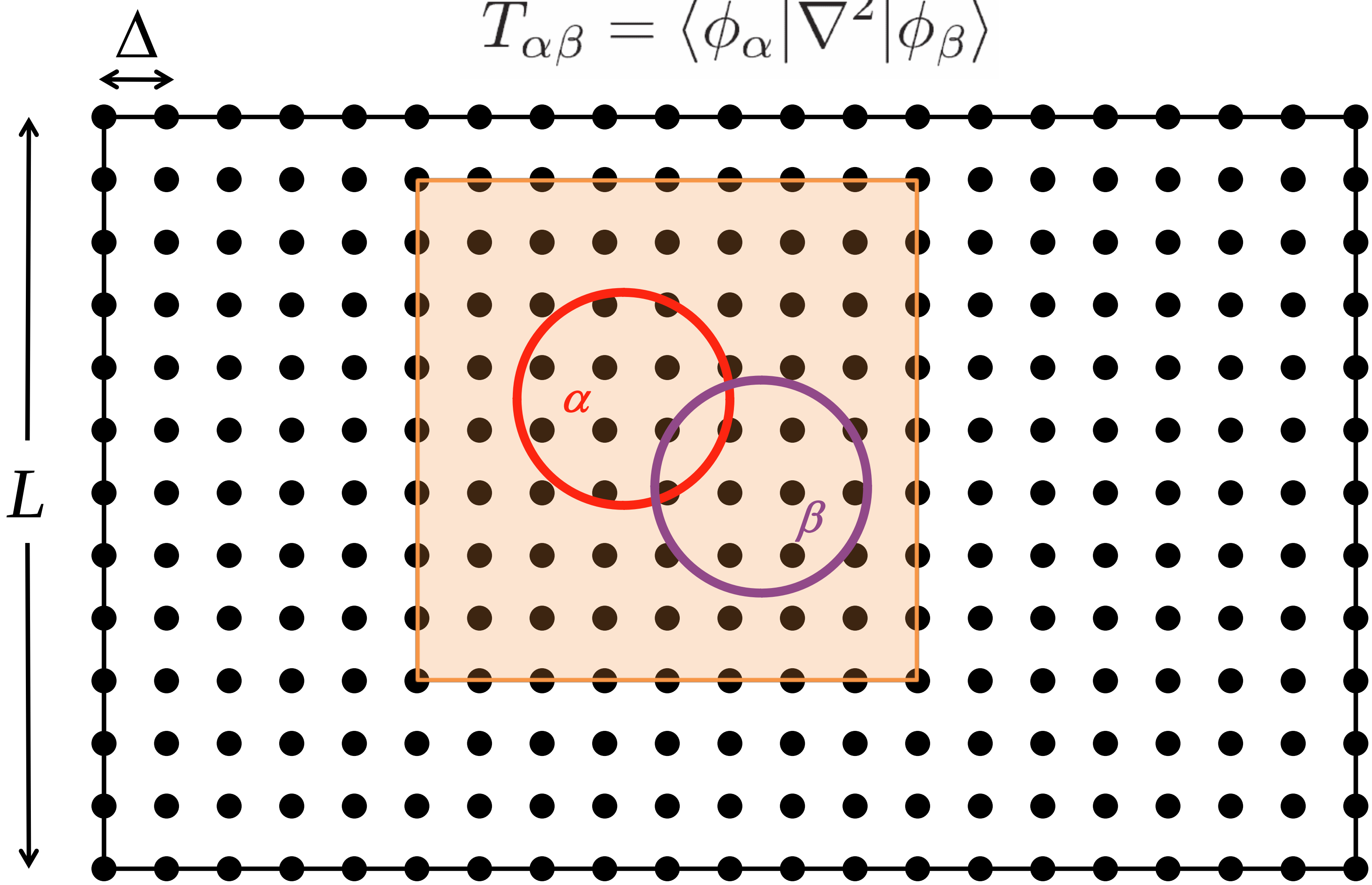
- Should ensure Hermiticity of operators is maintained when using FFT box:

$$O_{\alpha\beta} = \langle \phi_\alpha | \hat{O} | \phi_\beta \rangle = O^*_{\beta\alpha}$$

- Should ensure consistency of representation: when calculating $O_{\alpha\gamma}$ and $O_{\beta\gamma}$ the quantity $\hat{O}\left|\phi_\gamma\right\rangle$ is required and in both cases should be identical

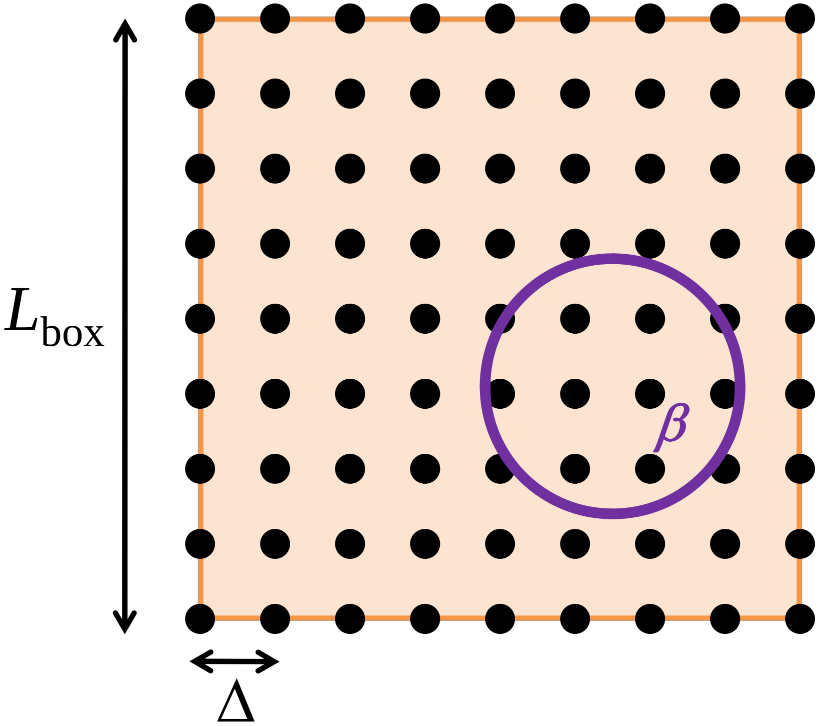
Kinetic with FFT Box

$$T_{\alpha\beta} = \langle \phi_\alpha | \nabla^2 | \phi_\beta \rangle$$



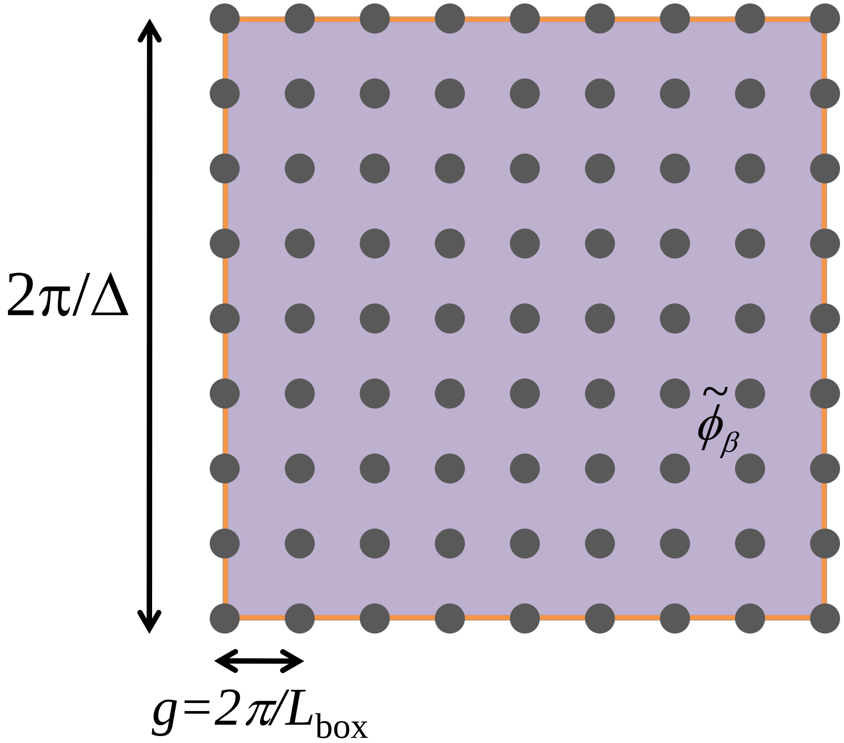
Kinetic with FFT Box

Put function in FFT box



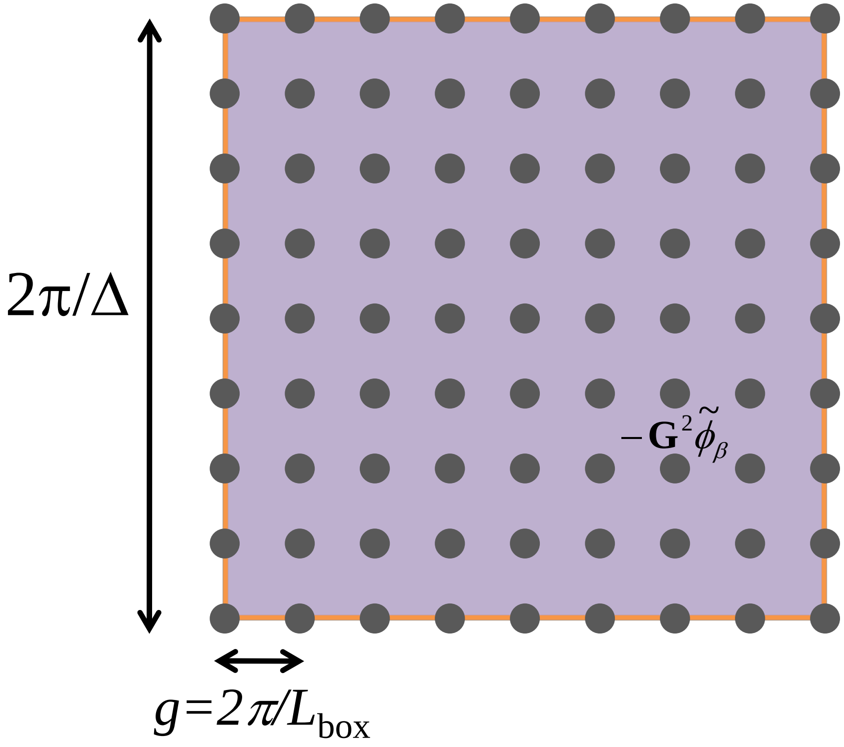
Kinetic with FFT Box

FFT to G-space



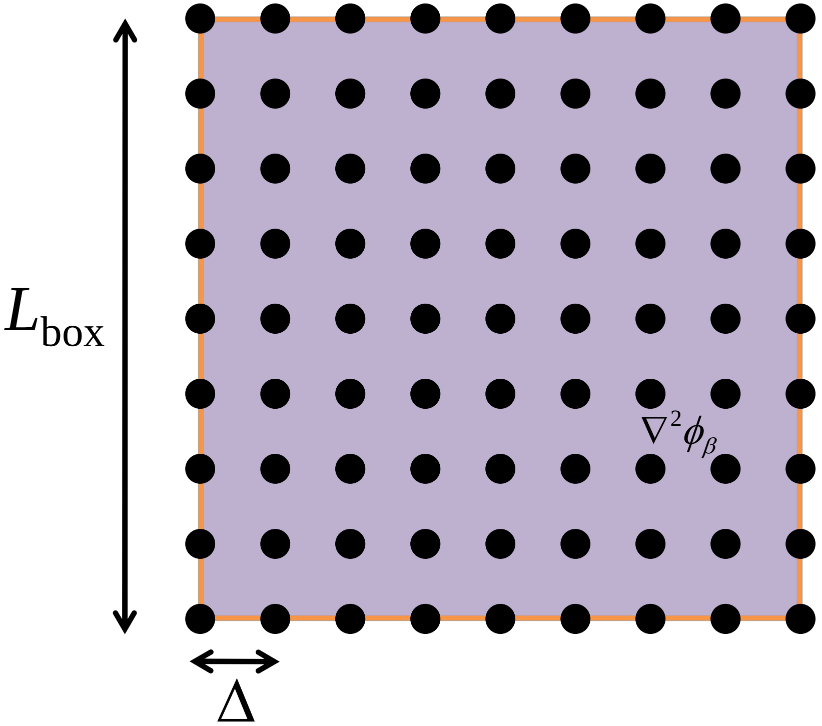
Kinetic with FFT Box

Apply Laplacian



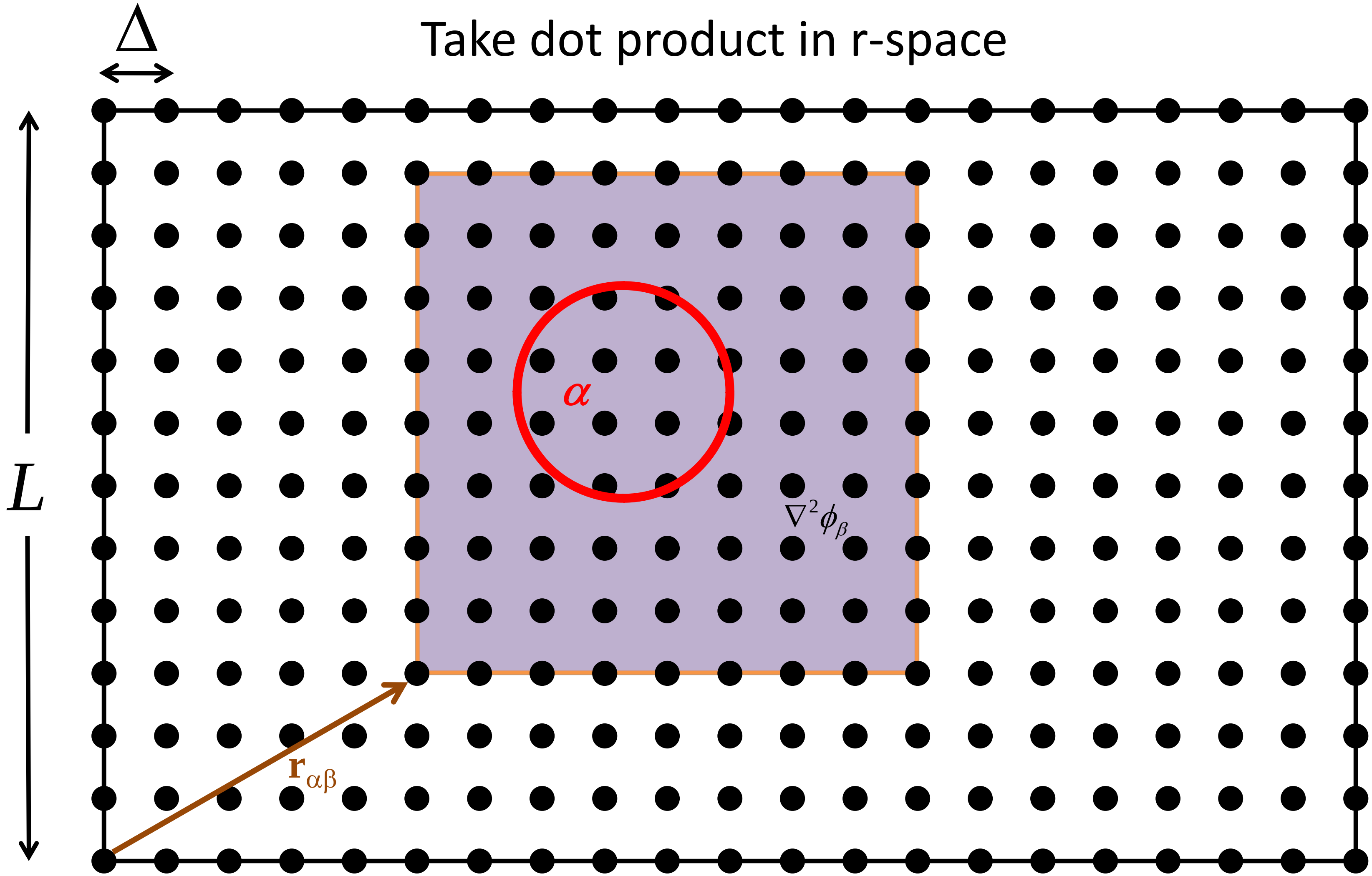
Kinetic with FFT Box

FFT back to r-space



Kinetic with FFT Box

Take dot product in r-space



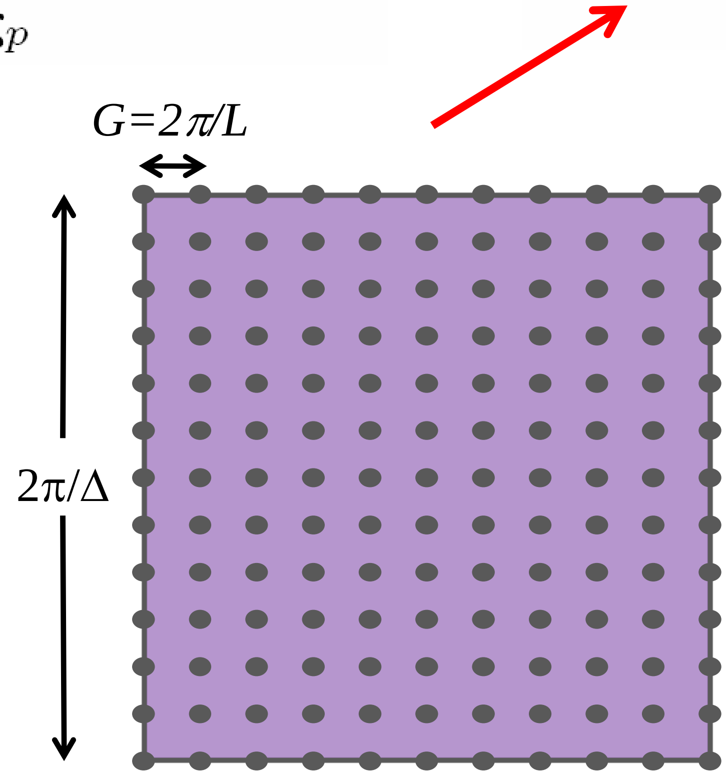
Coarse Sampling in G-Space

FFT Box

$$d_m(\mathbf{r}) = \frac{1}{n}\sum_{\mathbf{g}_p}^{\max} \mathrm{e}^{\mathrm{i}\mathbf{g}_p\cdot(\mathbf{r}-\mathbf{r}_m)}$$

Simulation Cell

$$D_m(\mathbf{r}) = \frac{1}{N}\sum_{\mathbf{G}_p}^{\max} \mathrm{e}^{\mathrm{i}\mathbf{G}_p\cdot(\mathbf{r}-\mathbf{r}_m)}$$



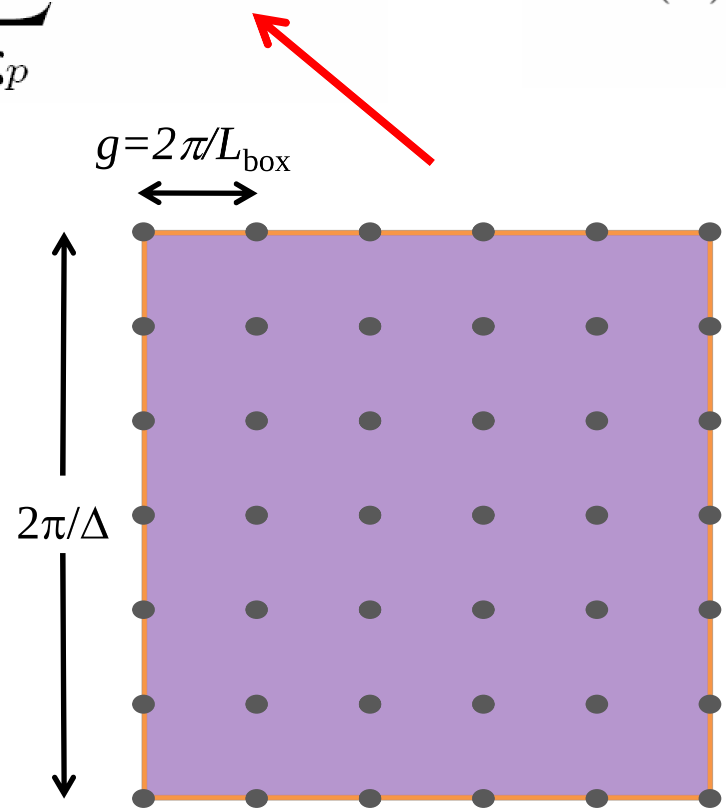
Coarse Sampling in G-Space

FFT Box

$$d_m(\mathbf{r}) = \frac{1}{n}\sum_{\mathbf{g}_p}^{\max} e^{i\mathbf{g}_p\cdot(\mathbf{r}-\mathbf{r}_m)}$$

Simulation Cell

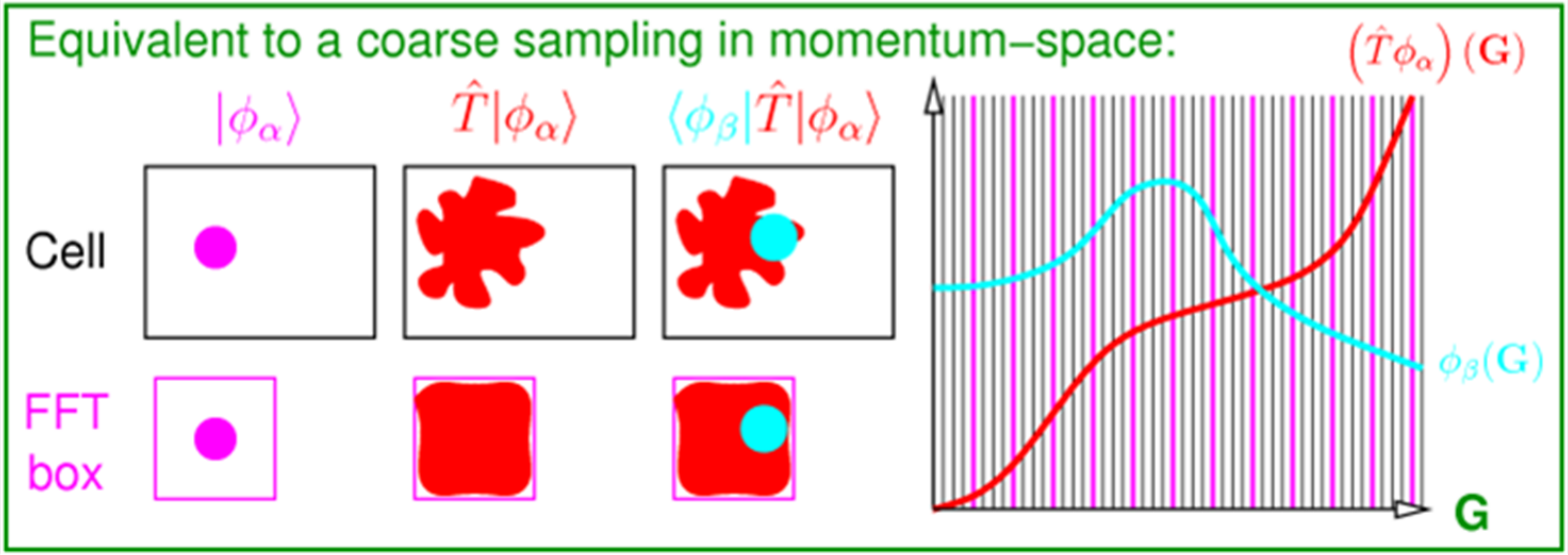
$$D_m(\mathbf{r}) = \frac{1}{N}\sum_{\mathbf{G}_p}^{\max} e^{i\mathbf{G}_p\cdot(\mathbf{r}-\mathbf{r}_m)}$$

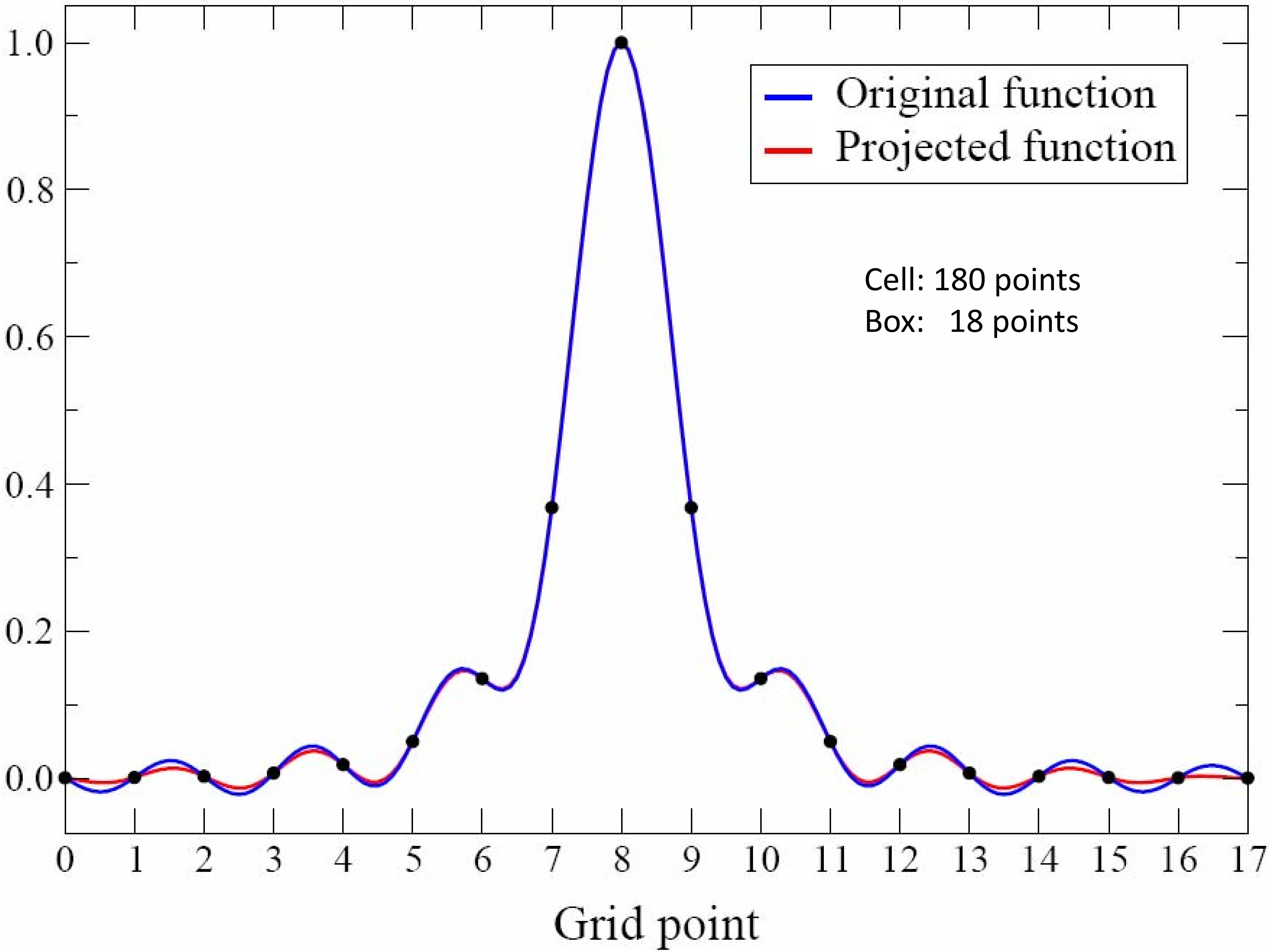


FFT Box as a Projection

$$\hat{\mathcal{P}}_{(\alpha\beta)} = \frac{1}{w}\sum_{m}^{\text{box}} |d_m\rangle\langle D_{m+(\alpha\beta)}| \;\rightarrow\; \mathcal{P}_{(\alpha\beta)}(\mathbf{r},\mathbf{r}') = \frac{1}{w}\sum_{m}^{\text{box}} d(\mathbf{r}-\mathbf{r}_m)D^*(\mathbf{r}'-\mathbf{r}_m-\mathbf{r}_{(\alpha\beta)})$$

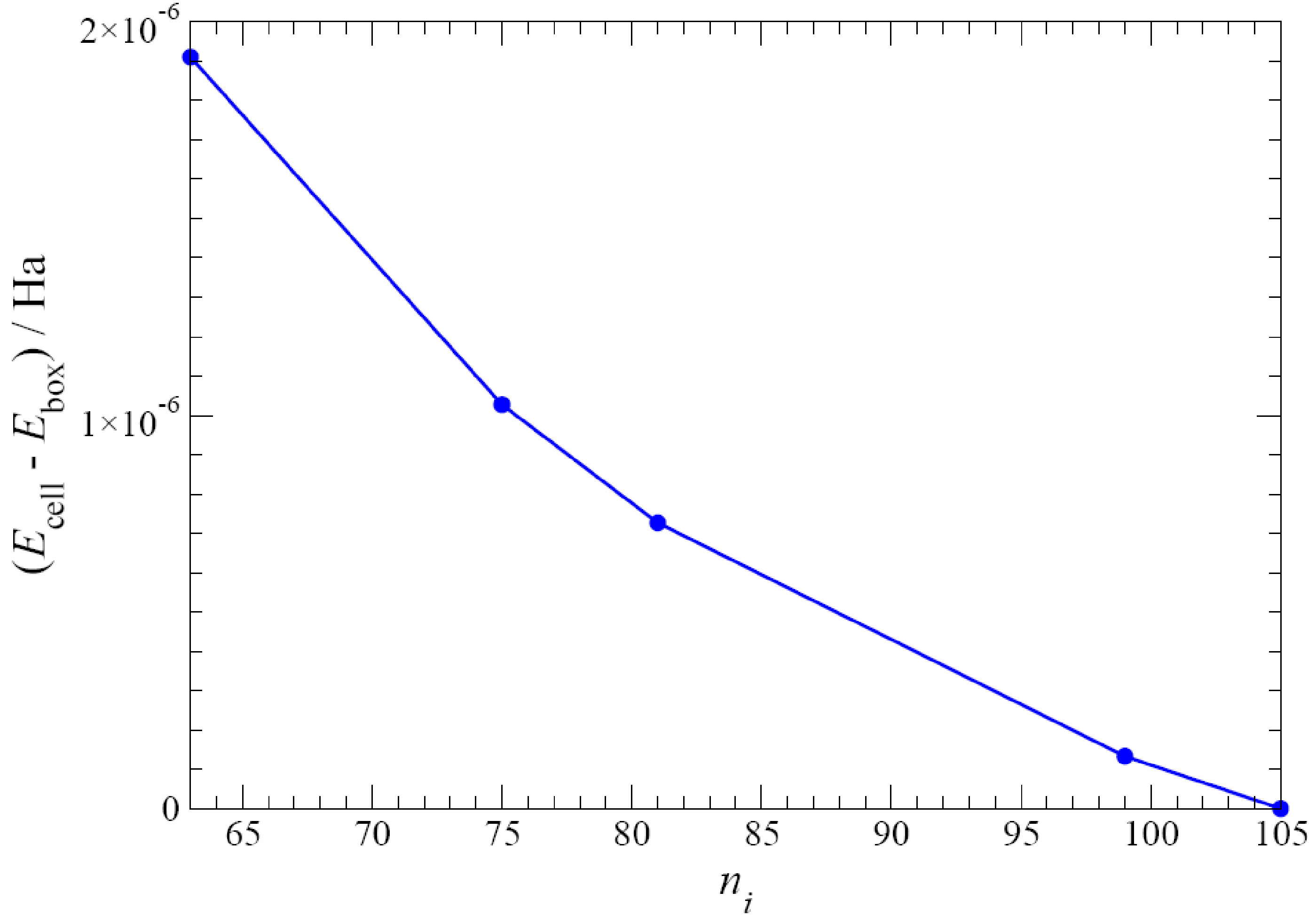
$$|\phi_\alpha^{(\alpha\beta)}\rangle \equiv \hat{\mathcal{P}}_{(\alpha\beta)}|\phi_\alpha\rangle \qquad \langle\phi_\alpha|\hat{H}|\phi_\beta\rangle \;\rightarrow\; \langle\phi_\alpha^{(\alpha\beta)}|\hat{H}|\phi_\beta^{(\alpha\beta)}\rangle = \langle\phi_\alpha|P^\top\hat{H}P|\phi_\beta\rangle$$





Original function
Projected function
Cell: 180 points
Box: 18 points
1.0
0.8
0.6
0.4
0.2
0.0
0
1
2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
Grid point

Accuracy of FFT Box



The End

Extra Slides

Hartree Energy

$$V^{\mathrm{H}}_{\alpha\beta} = \langle \phi_\alpha | V^{\mathrm{H}}(\mathbf{r}) | \phi_\beta \rangle$$

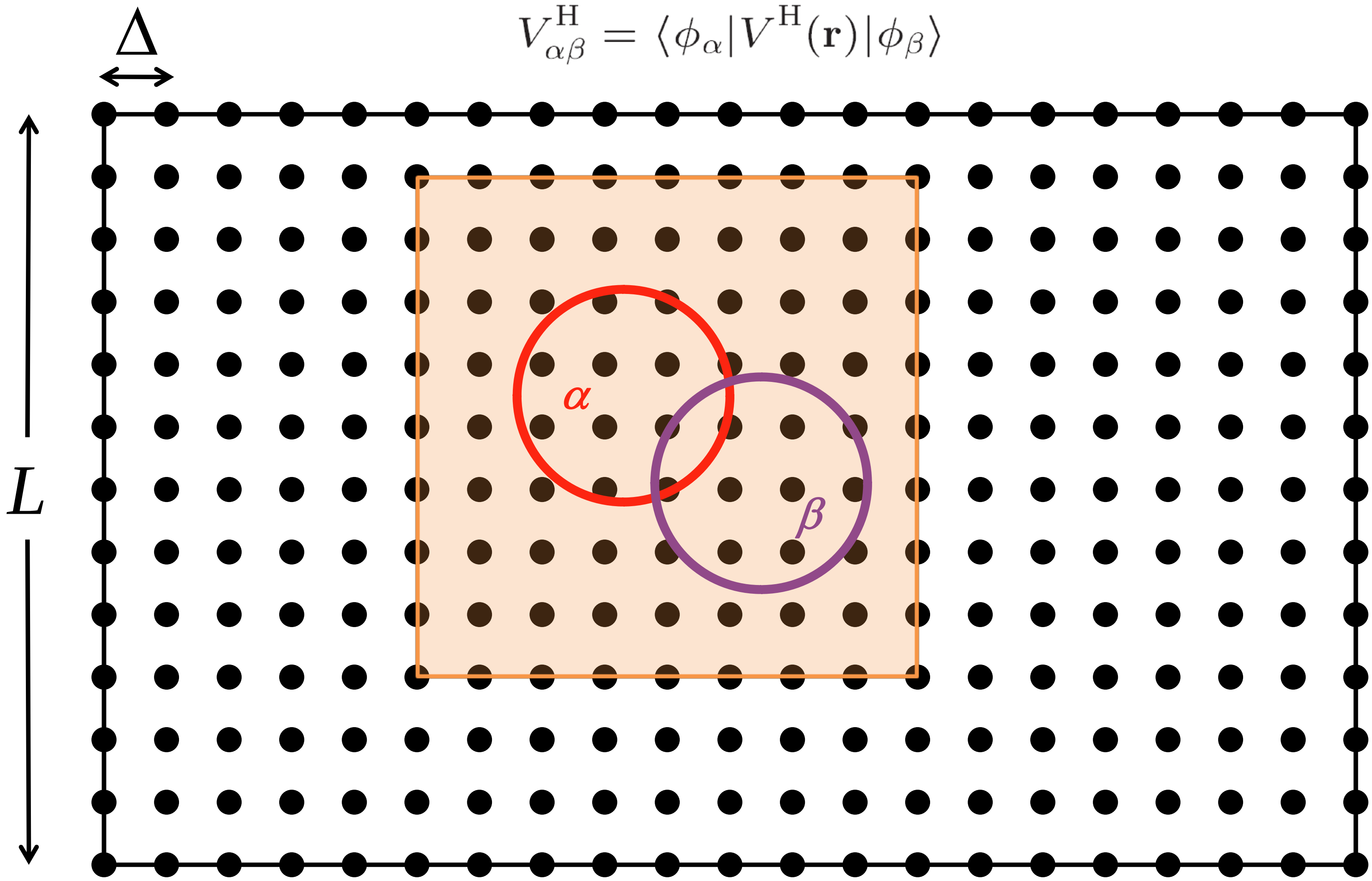
$$V^{\mathrm{H}}(\mathbf{r}) = \int d^3r' \frac{n(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|}$$

Require density on the r-space fine grid

$$n(\mathbf{r}) = K^{\alpha\beta} \phi_\alpha(\mathbf{r}) \phi_\beta(\mathbf{r})$$

Hartree Energy

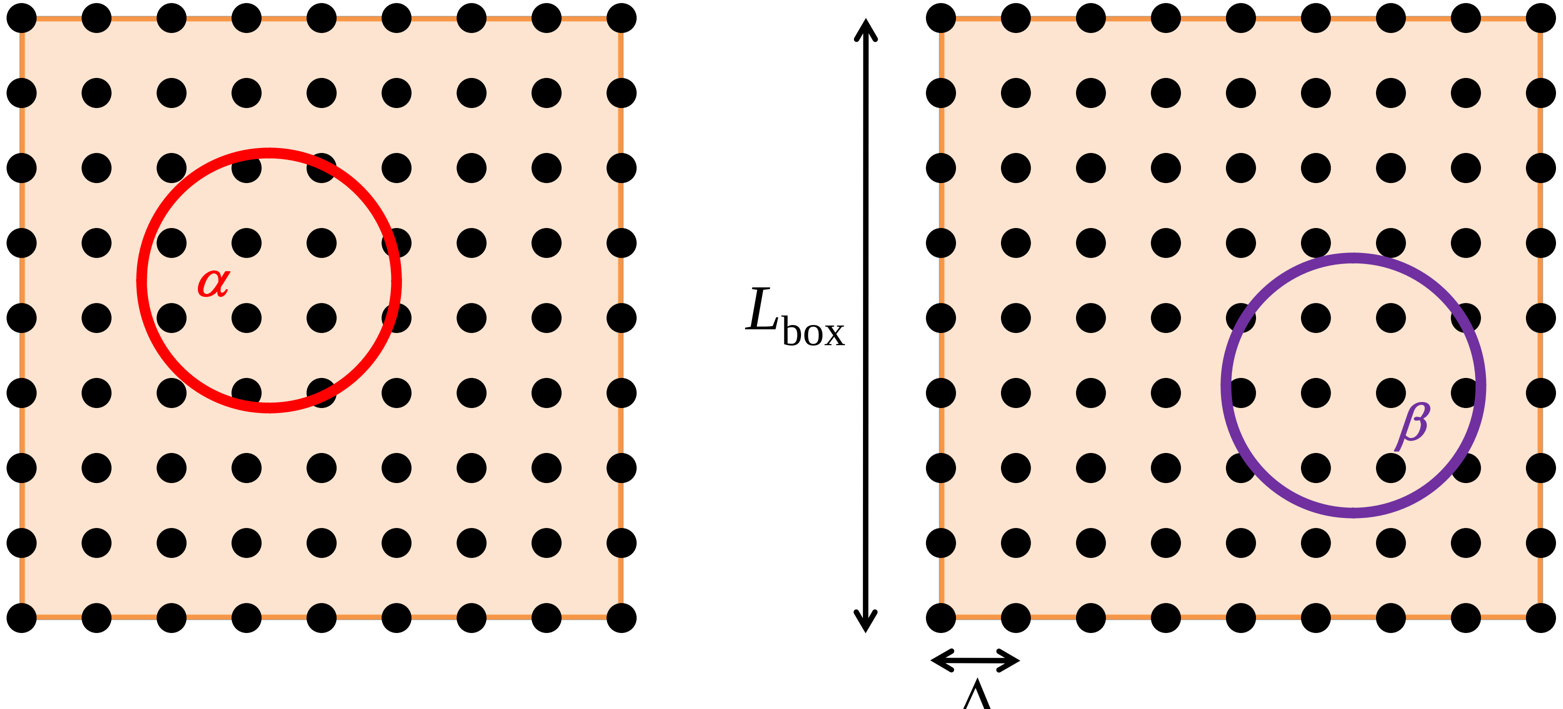
$$V^{\mathrm{H}}_{\alpha\beta} = \langle \phi_\alpha | V^{\mathrm{H}}(\mathbf{r}) | \phi_\beta \rangle$$



Density

$$n(\mathbf{r}) = K^{\alpha\beta}\phi_\alpha(\mathbf{r})\phi_\beta(\mathbf{r})$$

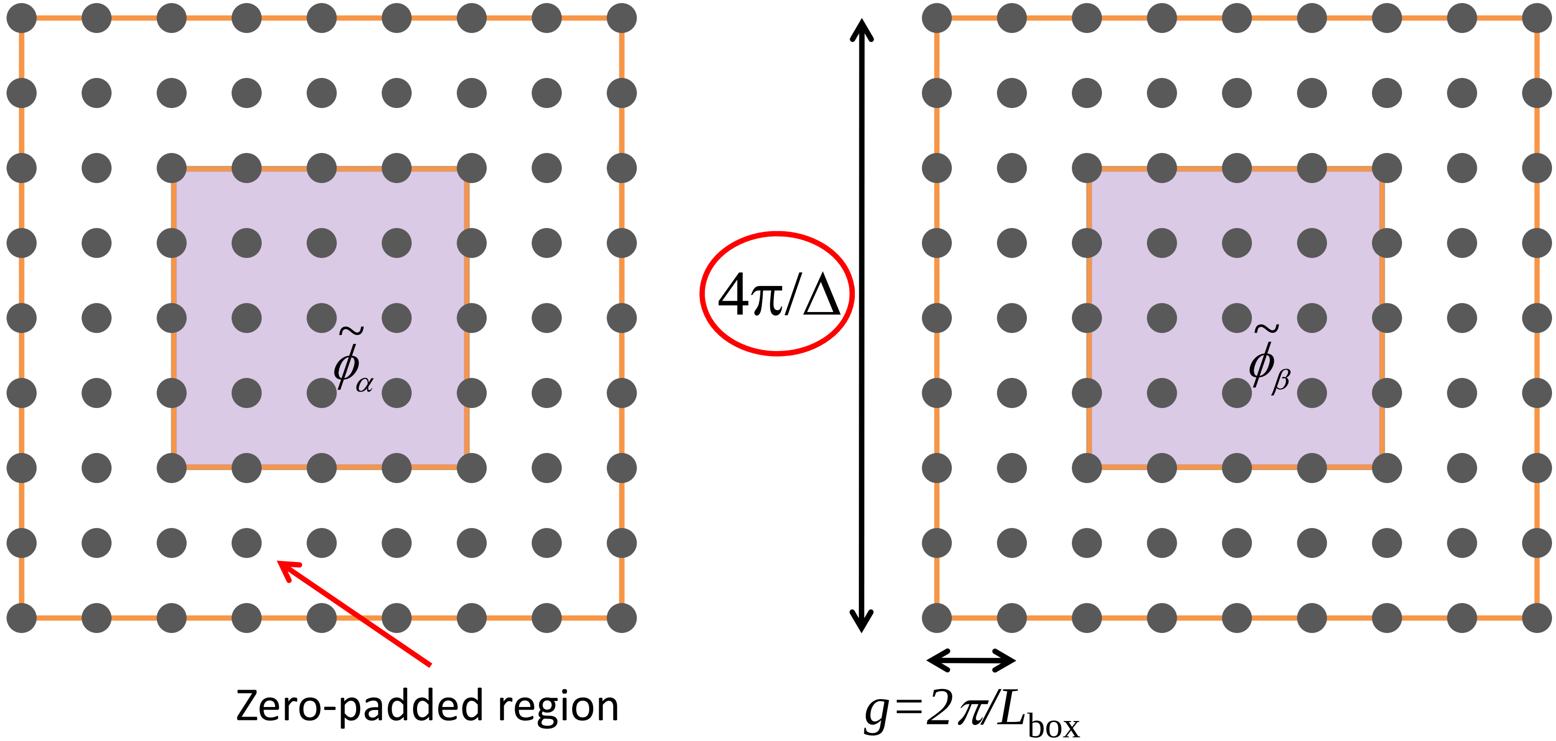
Put both functions in FFT box



Density

$$n(\mathbf{r}) = K^{\alpha\beta}\phi_\alpha(\mathbf{r})\phi_\beta(\mathbf{r})$$

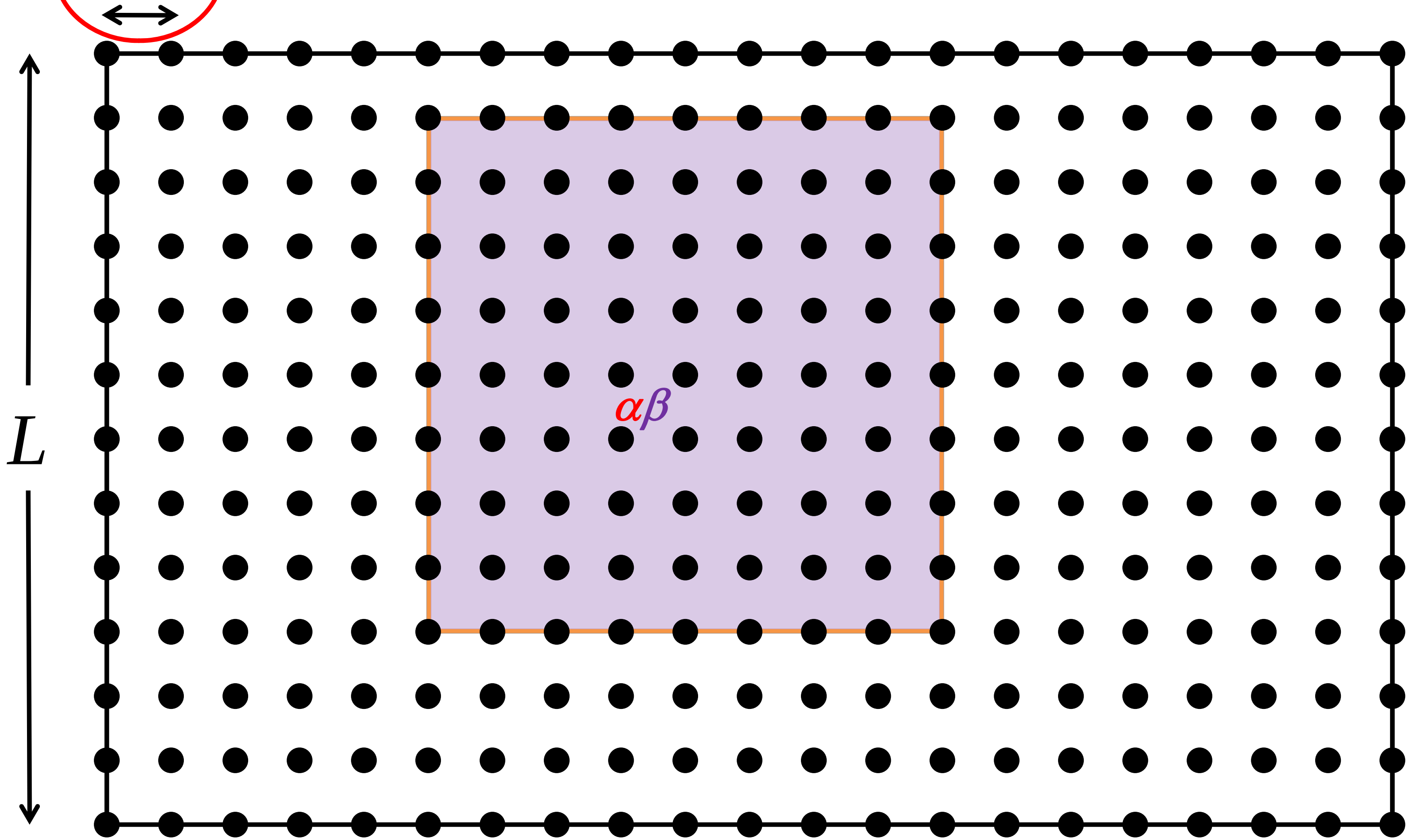
FFT to G-space and zero-pad to $2\mathbf{G}_{\max}$



$$n(\mathbf{r}) = K^{\alpha\beta}\phi_\alpha(\mathbf{r})\phi_\beta(\mathbf{r})$$

Density

$\Delta / 2$ FFT back to r-space *fine grid* FFT box and take product



Density & Hartree Potential

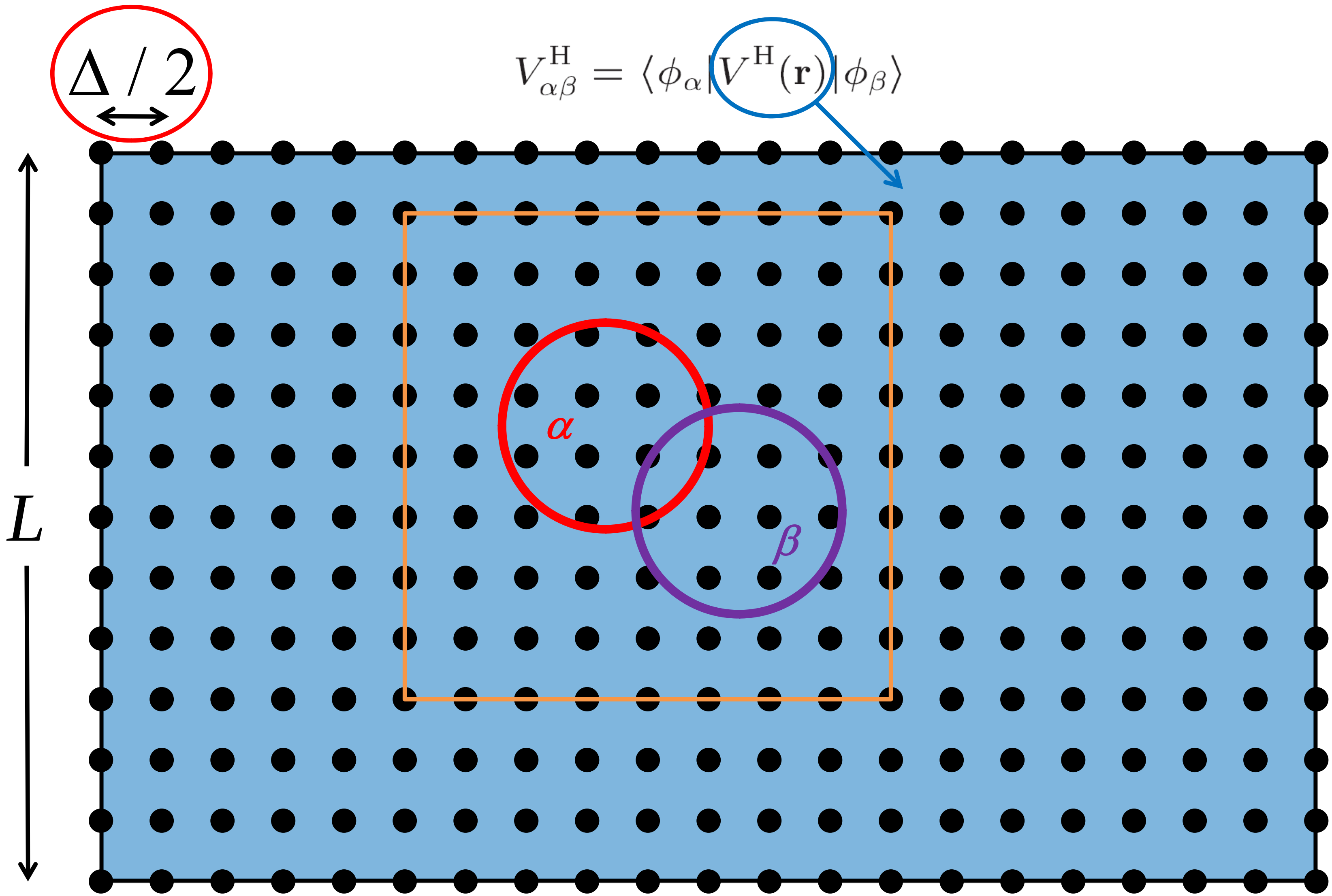
$$n(\mathbf{r}) = K^{\alpha\beta}\phi_\alpha(\mathbf{r})\phi_\beta(\mathbf{r})$$

$$V^{\mathrm{H}}(\mathbf{r}) = \int d^3r' \frac{n(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}$$

- Repeat for all overlapping pairs of NGWFs
- Accumulate result to obtain $n(\mathbf{r})$ on r-space fine grid
- To obtain V^{H}: FFT $n(\mathbf{r})$ to G-space, divide by G^2 at each point and FFT back to r-space: $O(N\log N)$

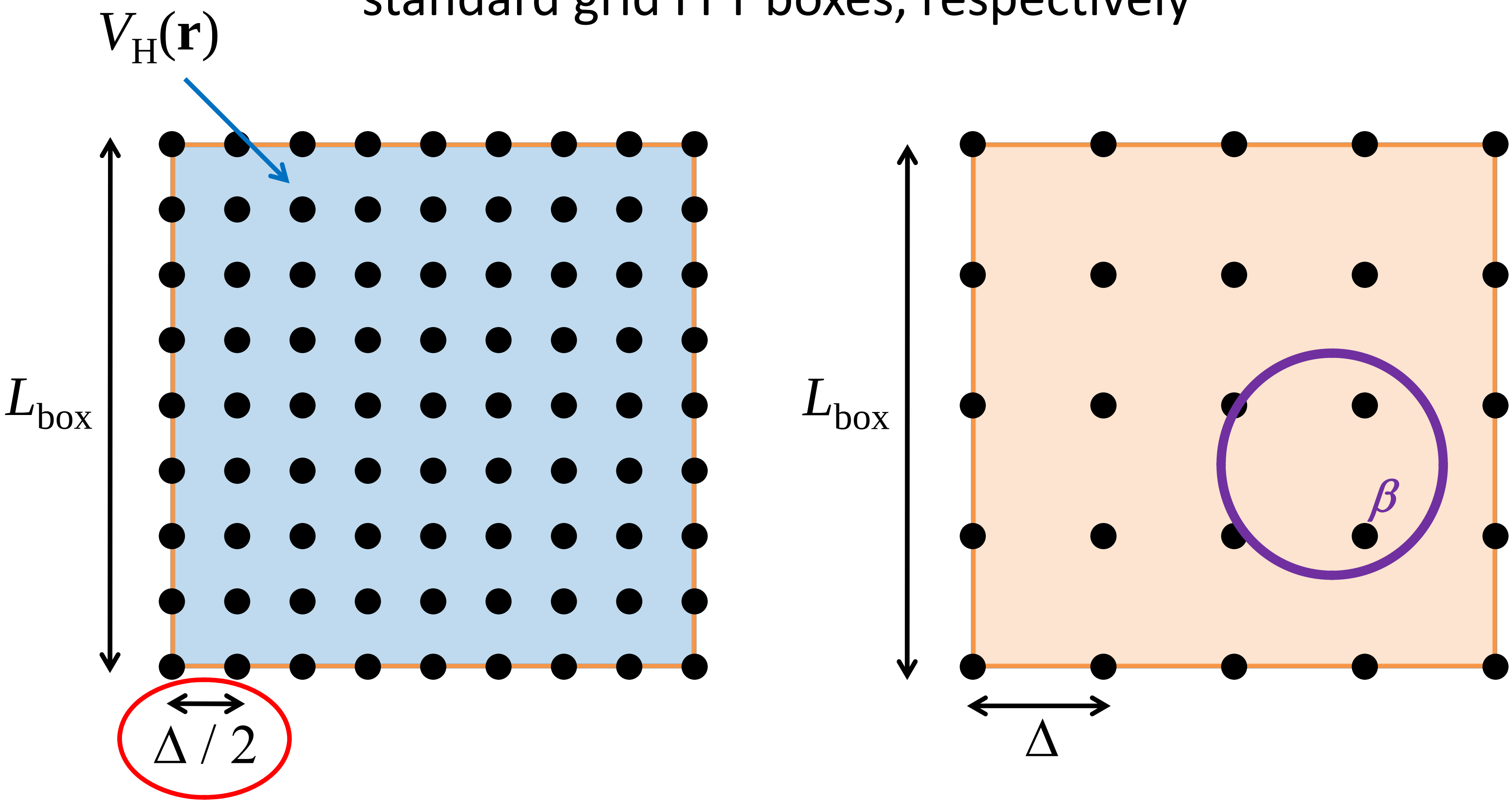
Hartree Matrix

$$V^{\mathrm{H}}_{\alpha\beta} = \langle \phi_\alpha | V^{\mathrm{H}}(\mathbf{r}) | \phi_\beta \rangle$$

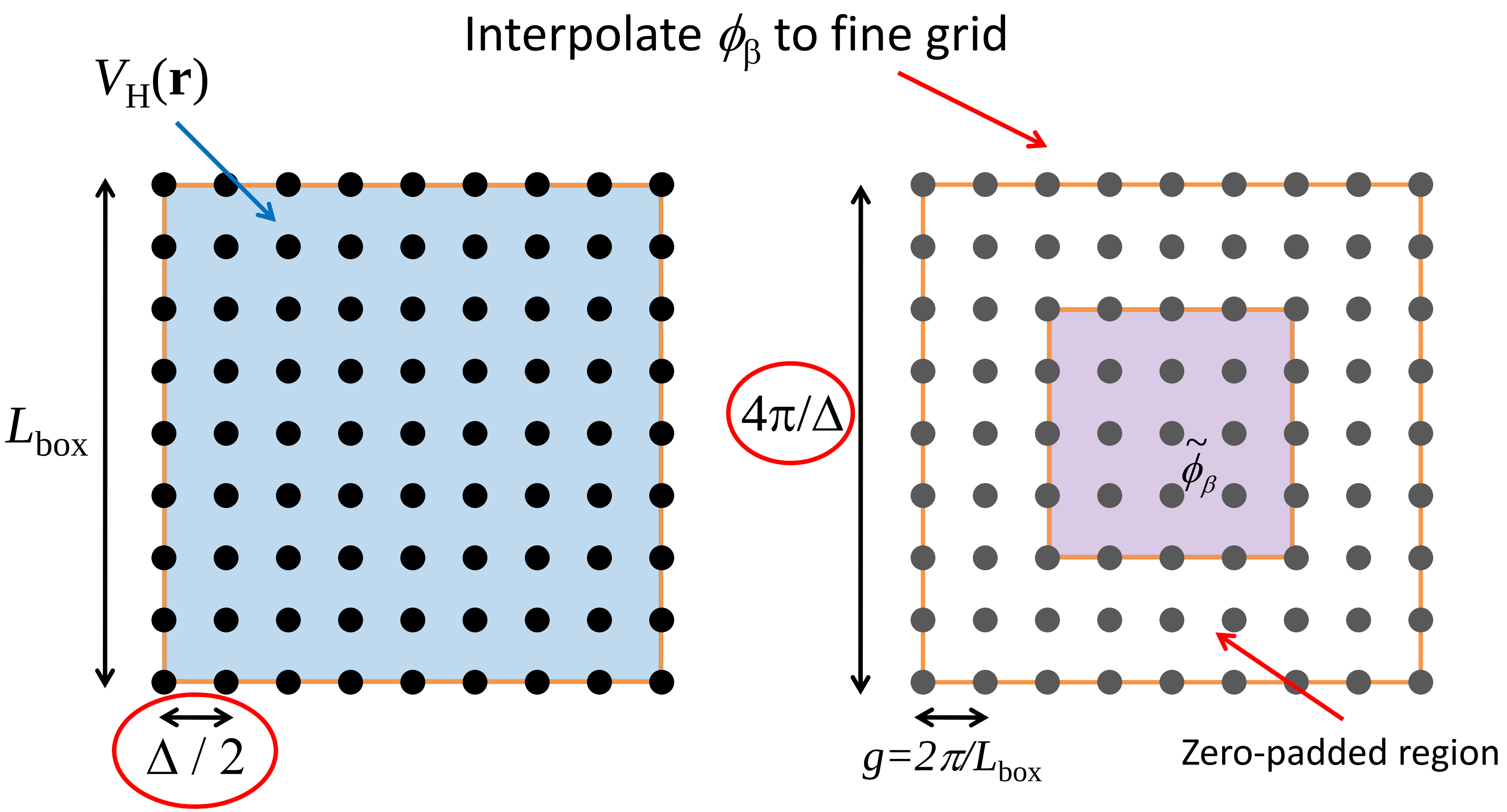


Hartree Matrix

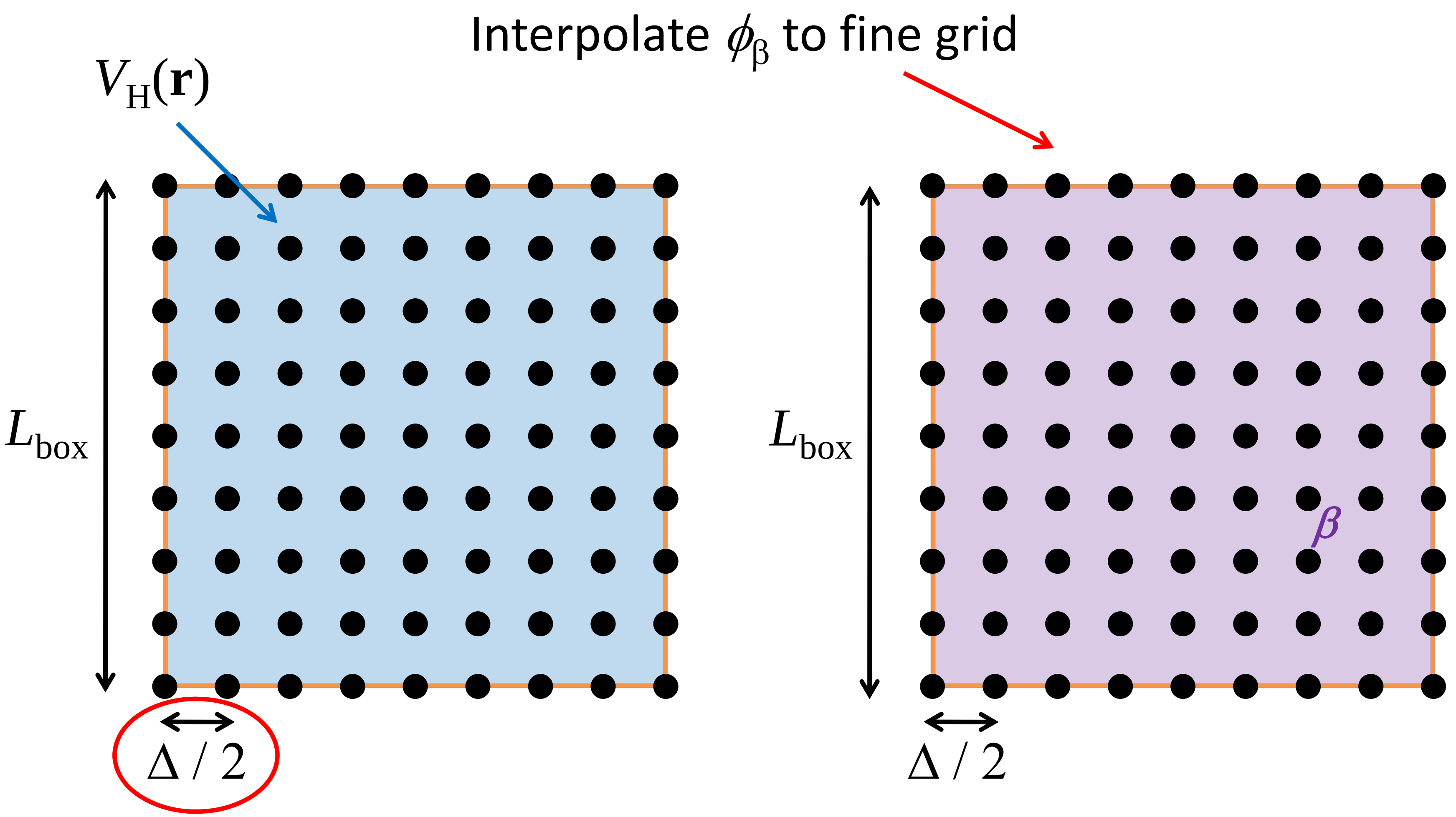
Extract $V_{\mathrm{H}}(\mathbf{r})$ and ϕ_{β} to fine grid and standard grid FFT boxes, respectively



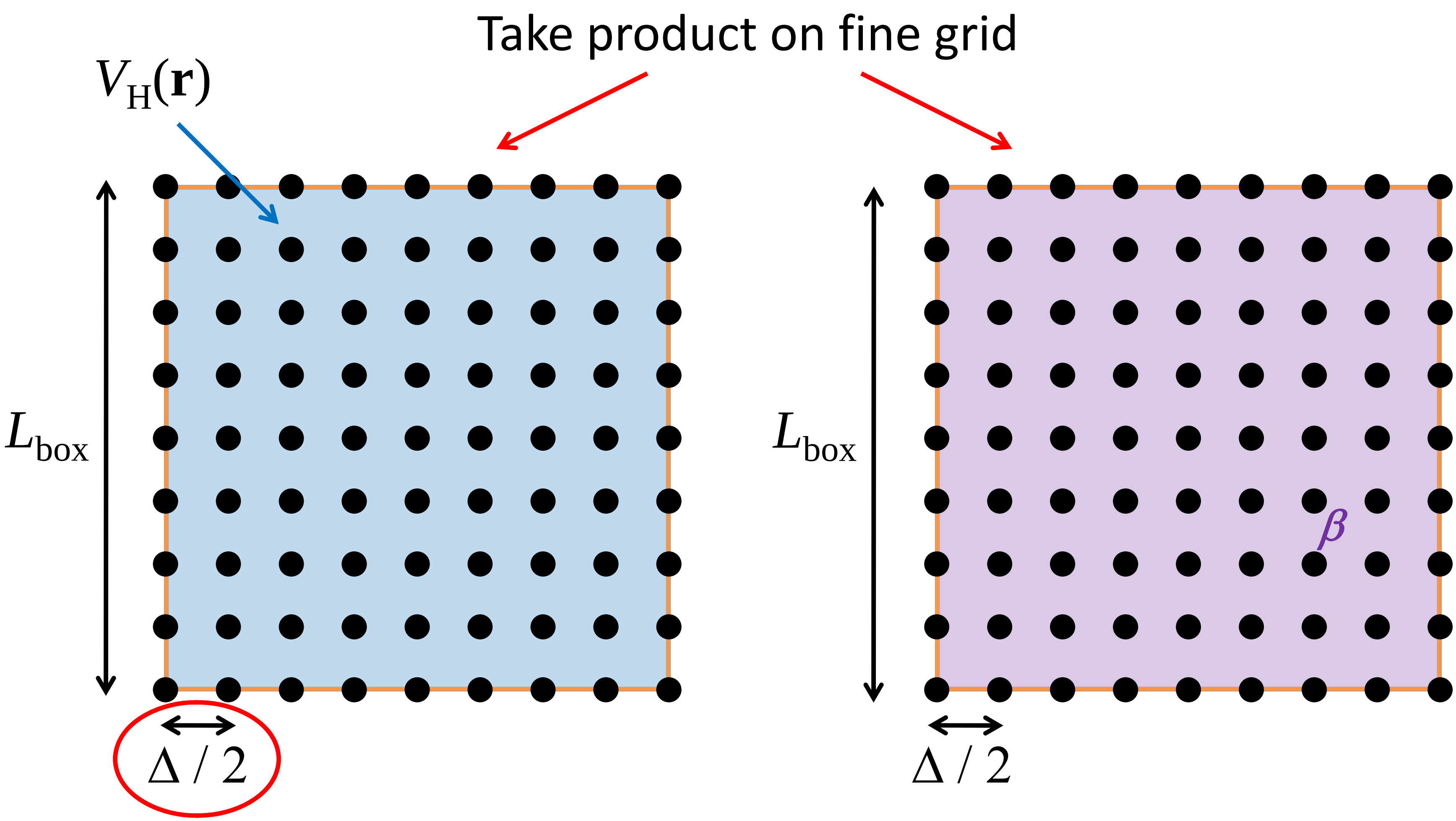
Hartree Matrix



Hartree Matrix

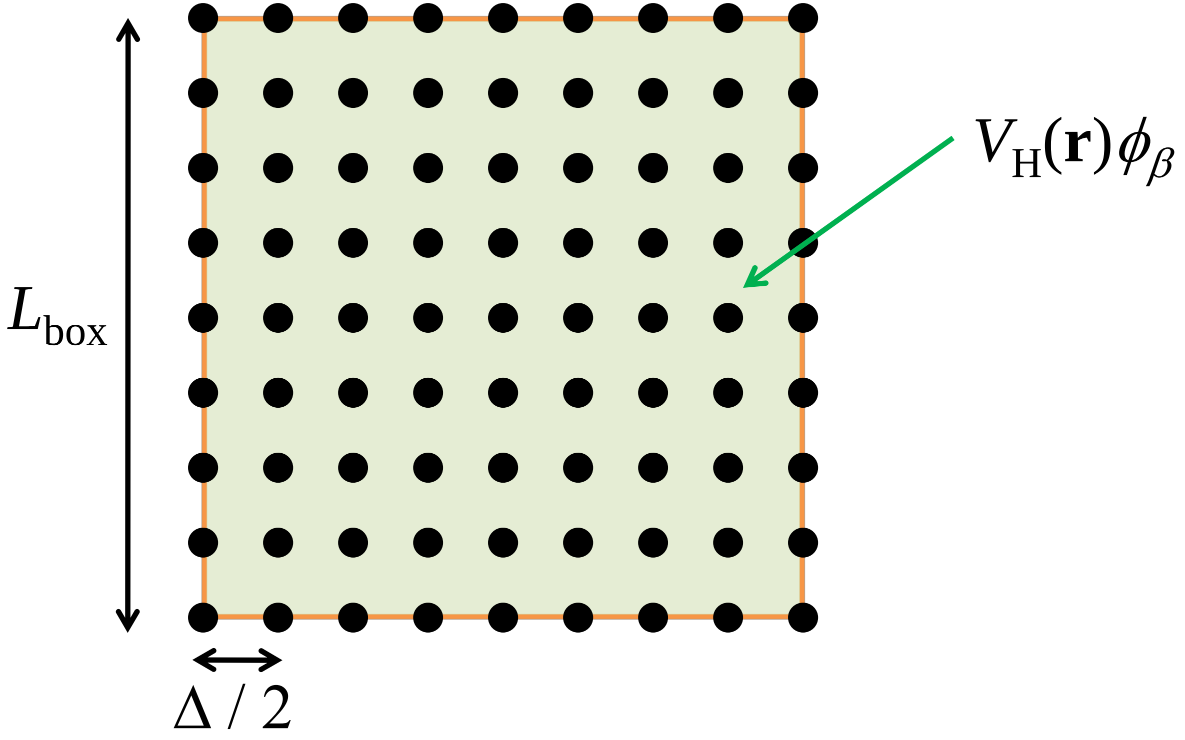


Hartree Matrix



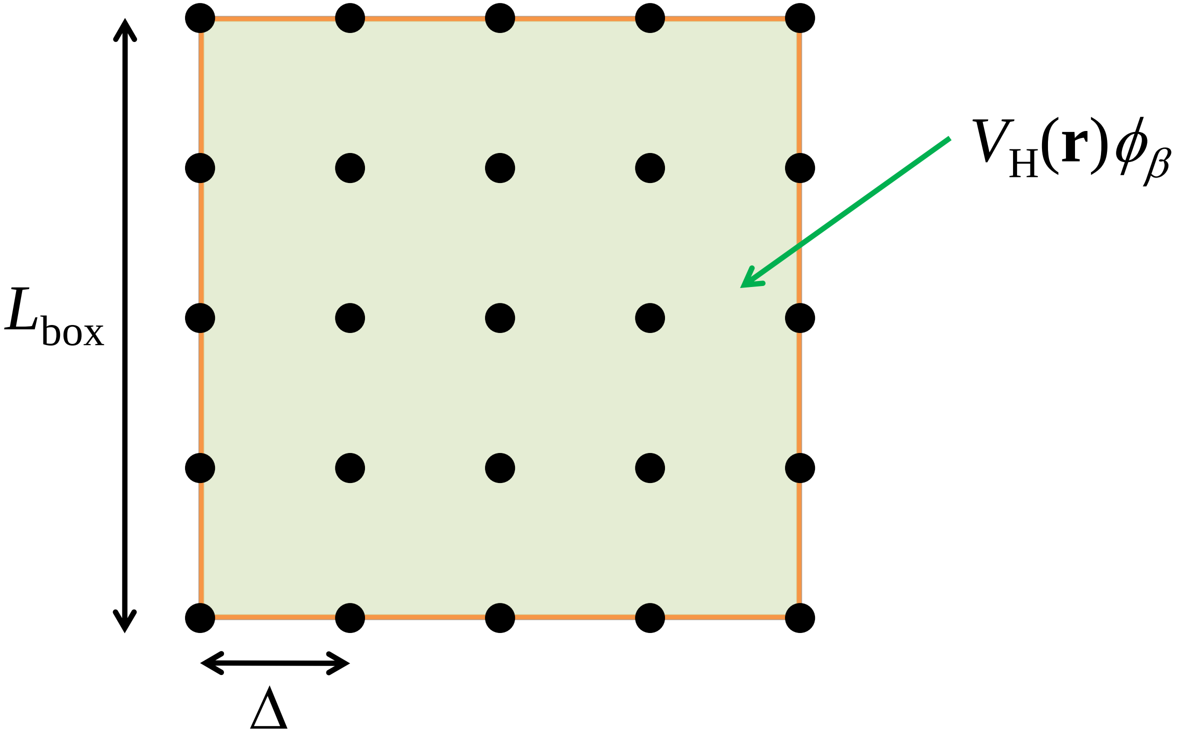
Hartree Matrix

Take product on the fine grid



Hartree Matrix

“Compress” to standard grid



Hartree Matrix

$$V_{\alpha\beta}^{\mathrm{H}} = \langle \phi_\alpha | V^{\mathrm{H}}(\mathbf{r}) | \phi_\beta \rangle$$

Take product of $V_{\mathrm{H}}(\mathbf{r})\phi_\beta$
with ϕ_α on standard grid