Deep Wavelet Scattering for Quantum Energy Regression

2016 March APS Meeting
Predicting and Classifying Materials via High-Throughput Databases and Machine Learning I

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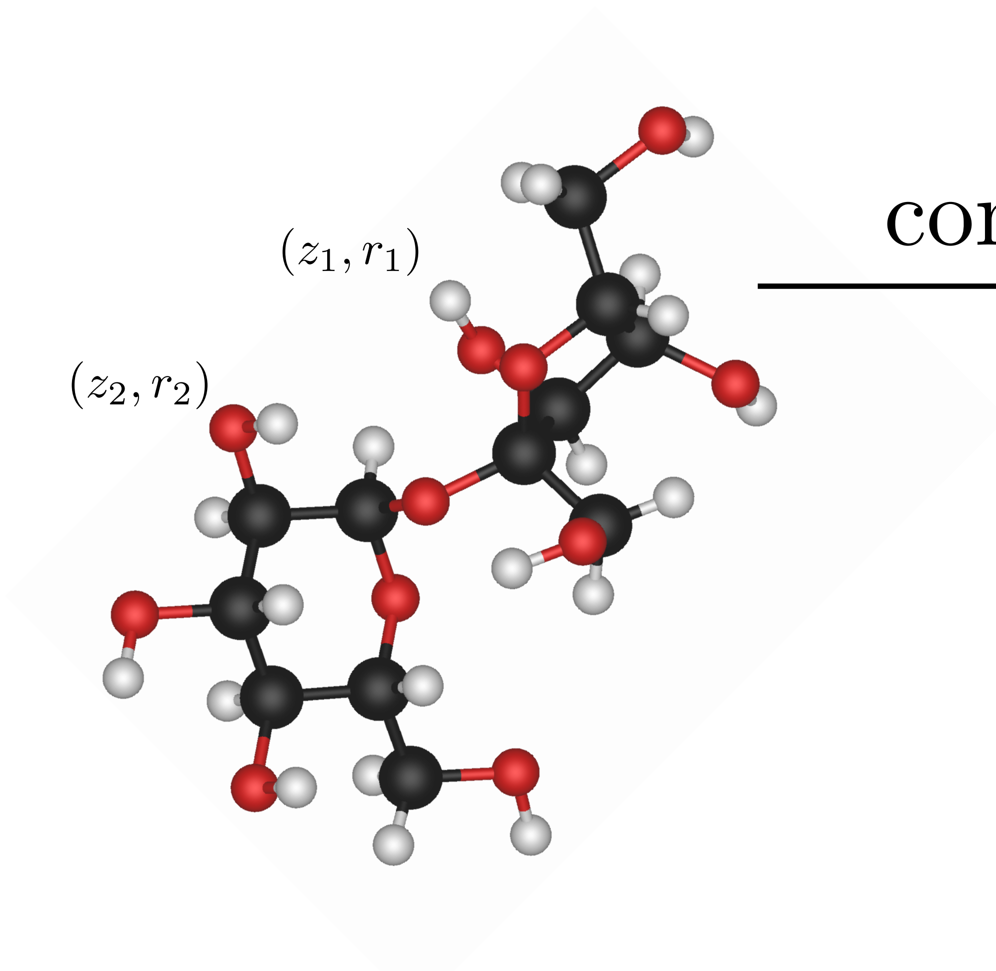
What do we want do?



Molecule

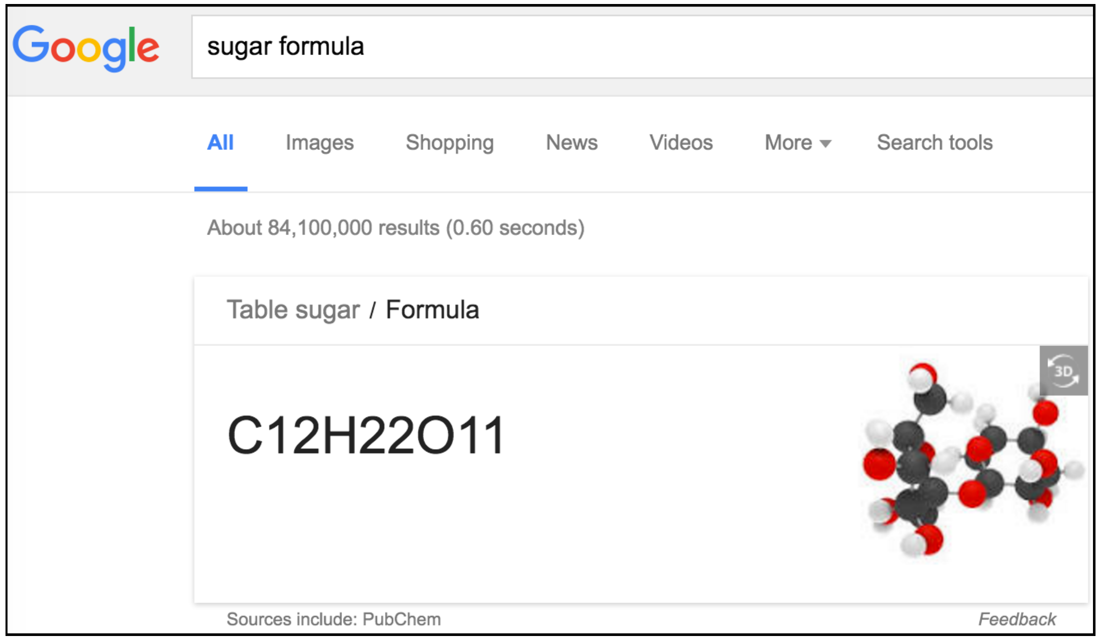
$$x = \{\text{charges, positions}\}$$
$$= \{(z_k, r_k)\}_k$$

$\xrightarrow{\text{compute}}$ Ground state energy

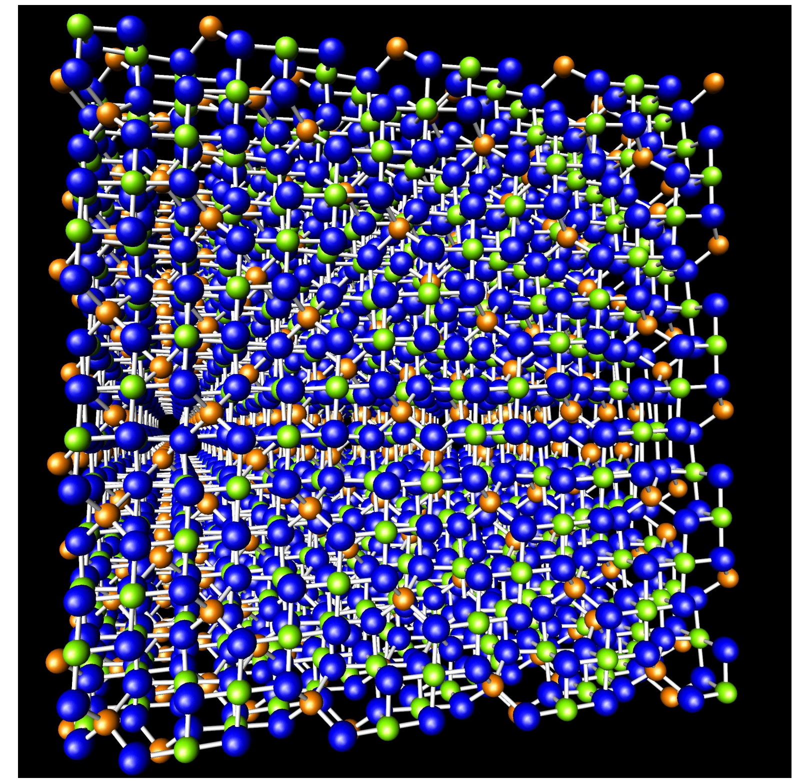
$$f(x) = E_0(x) + \sum_{k>l} \frac{z_k z_l}{|r_k - r_l|}$$

Energy from quantum electron interactions

Why do we want to do it?



Google of molecules



Materials design

Drug discovery

So what's the problem?

$$f(x) = E_0(x) + \sum_{k>l} \frac{z_k z_l}{|r_k - r_l|}$$

Schrödinger equation

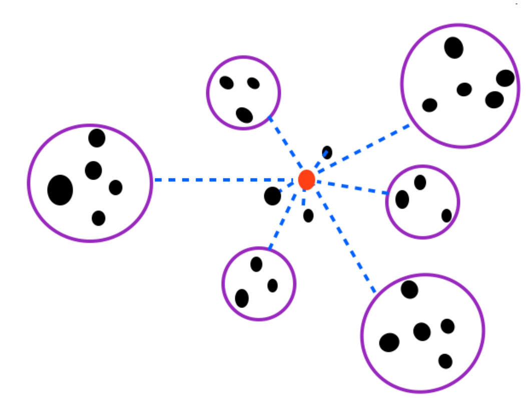
$$H[x]\Psi_0[x] = E_0(x)\Psi_0[x]$$

Not this

Fast multipole methods

(*Greengard, Rokhlin*)

Quantum mechanical (QM) approaches:



- Direct attacks (very small systems)
- Wave-function methods (small systems)
- Density functional theory (larger systems)

$O(N^\beta), \;\; \beta \geq 3$

computational cost

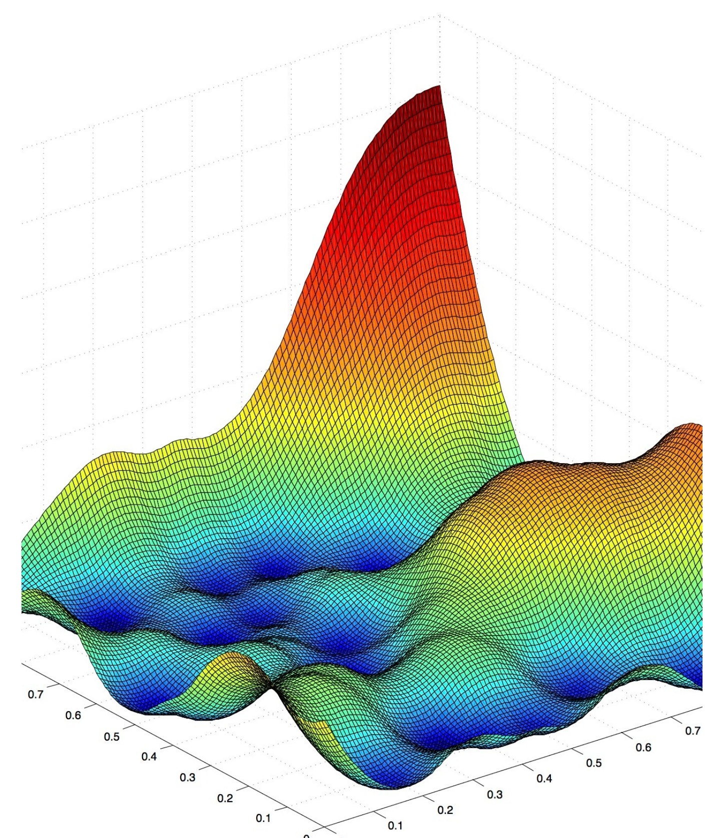
$N = \#$ electrons

Recent idea: Interpolation from known samples

- Use QM to compute training samples $\{(x_i, f(x_i))\}_{i \leq n}$
- Interpolate $f(x)$ from the training samples

<u>Issue:</u> Curse of dimensionality

- ϵ accuracy requires $O(\epsilon^{-d})$ samples
- $d = \text{dimension} = O(\#\,\text{atoms})$



Recent idea:
Interpolation from known samples

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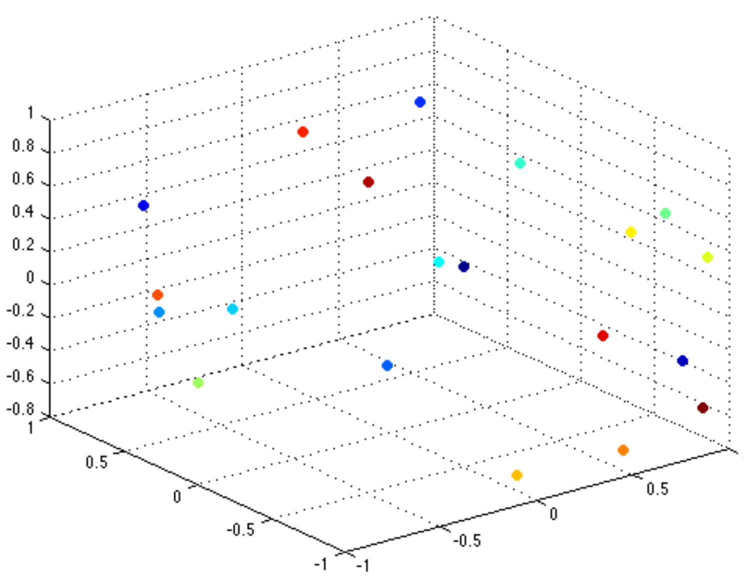


Need to take advantage of ***invariants*** *and* ***regularity*** *of f*

Energy
invariants and regularity

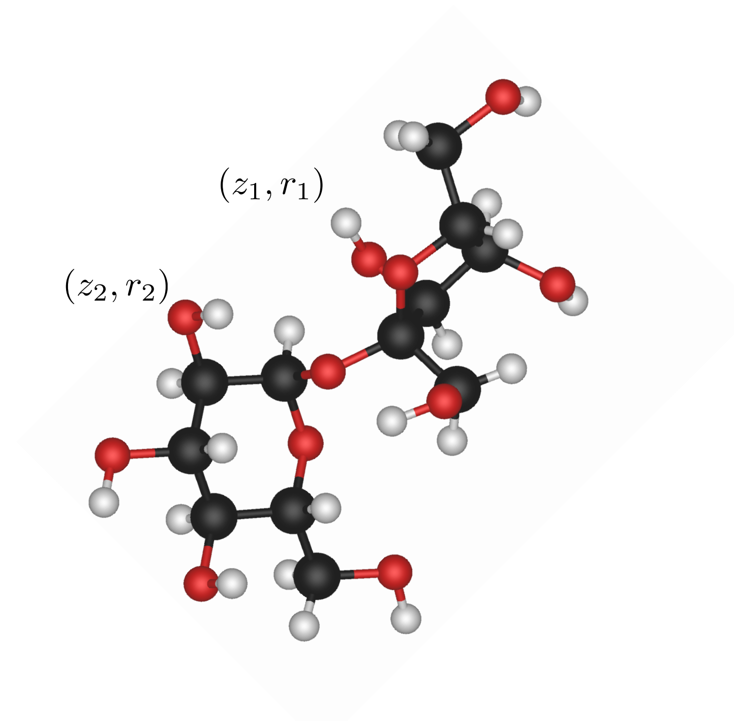
- <u>**Permutation invariance**</u>
  Invariant to permutations of the atom index k

- <u>**Isometry invariance**</u>
  Invariant to translations, rotations and reflections

- <u>**Deformation stability**</u>
  Lipschitz stable to diffeomorphisms of the molecule

(Bartók, Kondor, Csányi)

$$f(x) = E_0(x) + \sum_{k>l} \frac{z_k z_l}{|r_k - r_l|}$$



Molecule

$$x = \{\text{charges}, \ \text{positions}\}$$
$$= \{(z_k, r_k)\}_k$$

Regression over a dictionary

- New representation of the molecule x:
$$\Phi(x) = (\phi_k(x))_k$$

- Linear regression (interpolation) of f using Φ:
$$\widetilde{f}(x) = \langle w, \Phi(x) \rangle = \sum_k w_k \phi_k(x)$$

- Weights $(w_k)_k$ learned from the training data

(*Bartók, Kondor, Csányi: SOAP kernels*)
(*Rupp, Tkatchenko, Müller, von Lilienfeld: Coulomb matrices*)
(*And many others...*)

Dictionary properties

$$\widetilde{f}(x) = \langle w, \Phi(x) \rangle = \sum_k w_k \phi_k(x)$$

The dictionary Φ should:

- Have the same invariants and regularity as f
  - permutation invariance
  - isometry invariance
  - deformation stability

- Span a large enough space to approximate f to high accuracy, with as few terms as possible

$$\|w\|_0 \leq M$$

Dictionary structure

Decompose Φ as:

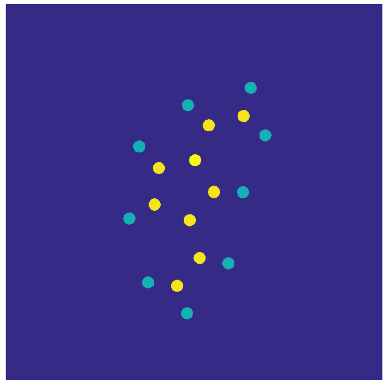
$$\Phi(x) = \Theta\rho[x], \quad x = \{(z_k, r_k)\}_k$$

- $\rho[x] : \mathbb{R}^3 \to \mathbb{R}$
- Permutation invariant
- Isometry covariant

- Isometry invariant
- Lipschitz stable to diffeomorphisms
- *This part is the main difficulty*

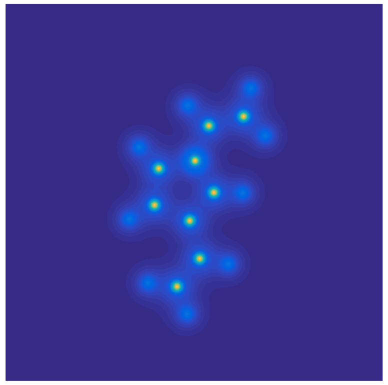
Non-interacting density

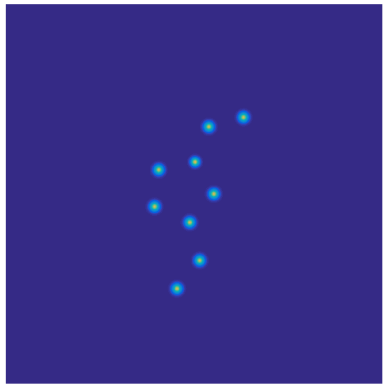
$$\rho[x](u) = \sum_k \rho[z_k](u - r_k), \quad x = \{(z_k, r_k)\}_k$$



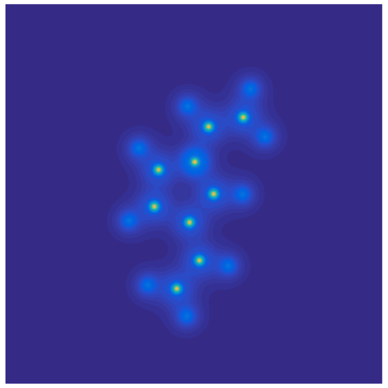
Candidates for $\rho[z_k]$:

- $\rho[z_k] = z_k \delta$
- $\rho[z_k] = \text{Isolated atomic density}$
  $= \rho_{\text{cor}}[z_k] + \rho_{\text{val}}[z_k]$





core



valence

- $\rho[z_k] = (\rho_{\text{cor}}[z_k], \rho_{\text{val}}[z_k])$

Fourier and autocorrelation

Dictionary $\Phi = \Theta\rho$

Now we focus on Θ; recall goals:

- Isometry invariant (translations, rotations, reflections)
- Stable to deformations

"Classic" translation invariant representations:

- Autocorrelation: $\Theta\rho(\tau) = \int \rho(u)\rho(u-\tau)\,du$
- Fourier modulus: $\widehat{\Theta}\rho(\omega) = |\widehat{\rho}(\omega)|^2$

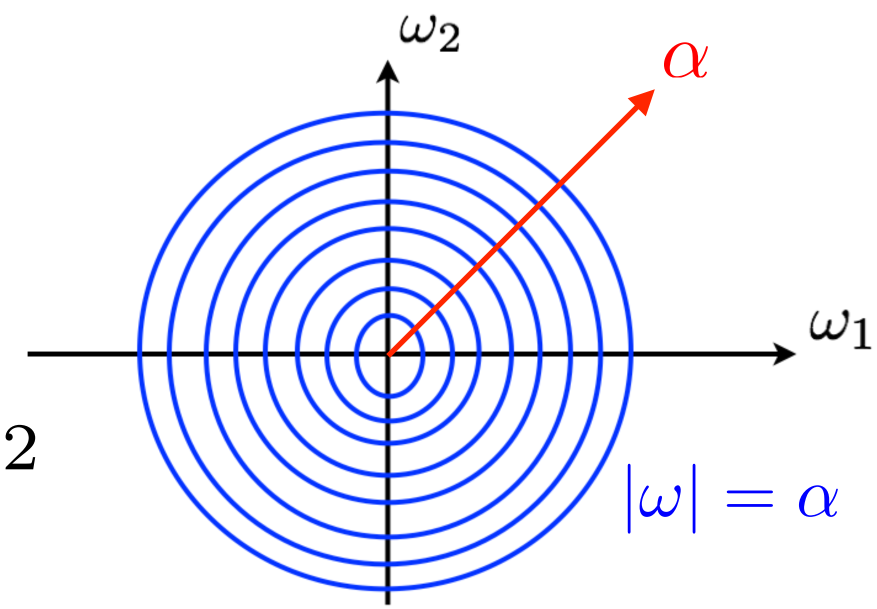
Integrate over rotations to obtain isometry invariance

Fourier and autocorrelation

Invariant Fourier operator:

$$\widehat{\Theta}\rho(\alpha) = \|\widehat{\rho}_\alpha\|_2^2 = \int_{S^2} |\widehat{\rho}(\alpha\eta)|^2 \, d\eta$$

$$\omega = \alpha\eta, \; (\alpha, \eta) \in \mathbb{R}^+ \times S^2$$



Pros:

- Isometry invariant
- Diagonalizes Coulomb:

$$U(\rho) = \iint \frac{\rho(u)\rho(v)}{|u-v|} \, du \, dv = \frac{1}{2\pi^2} \int_0^\infty \alpha^{-2} \|\widehat{\rho}_\alpha\|_2^2 \, d\alpha$$

Fourier and autocorrelation

<u>Invariant Fourier operator:</u>

$$\widehat{\Theta}\rho(\alpha) = \|\widehat{\rho}_\alpha\|_2^2 = \int_{S^2} |\widehat{\rho}(\alpha\eta)|^2 \, d\eta$$

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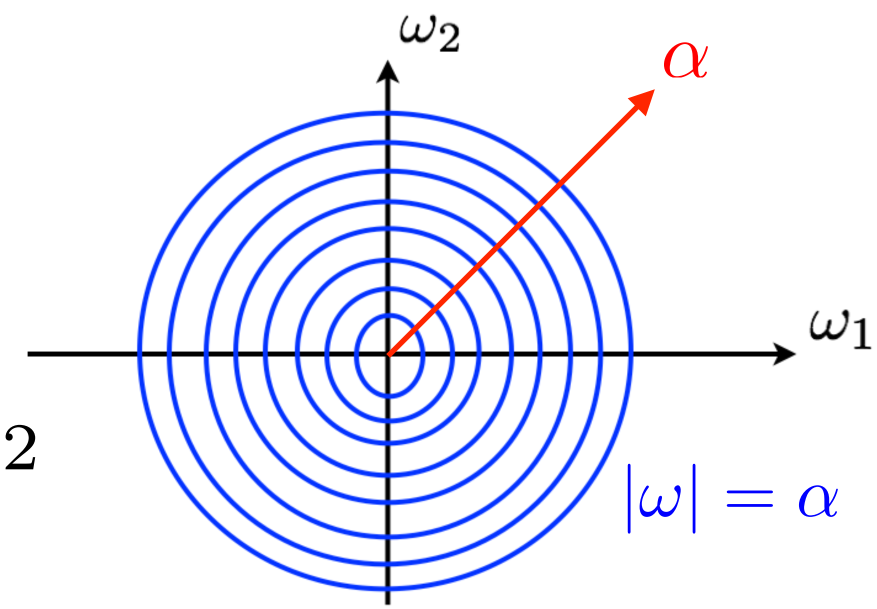
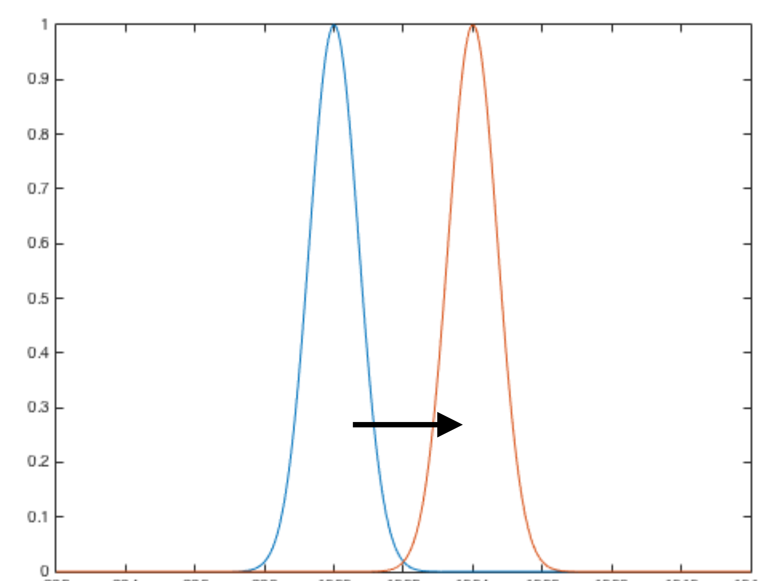


<u>Cons:</u>

- Not sparse: $U(\rho) = \sum_{k=1}^{\epsilon^{-2}} w_k \|\widehat{\rho}_{k\epsilon}\|_2^2 + O(\epsilon)$
- Not stable to deformations:

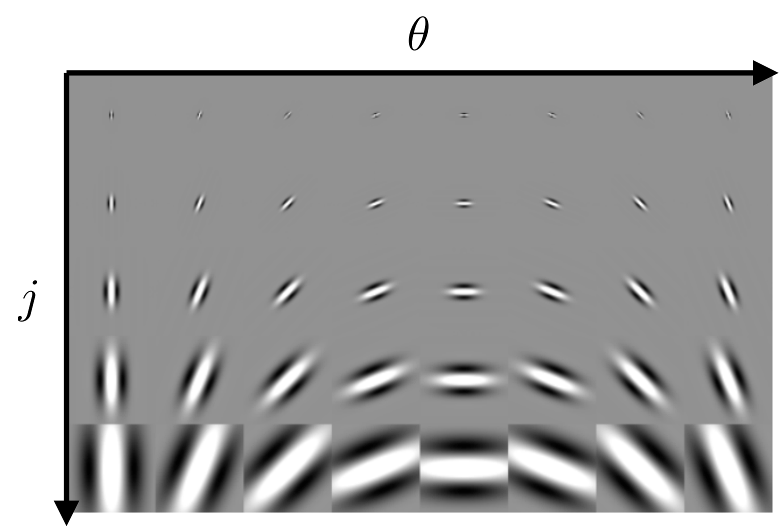
  Invariant autocorrelation:
  – Bumps located at $\{|r_k - r_l|\}_{k,l}$
  – Δ sized diffeomorphism
  moves them $\Delta|r_k - r_l|$ distance



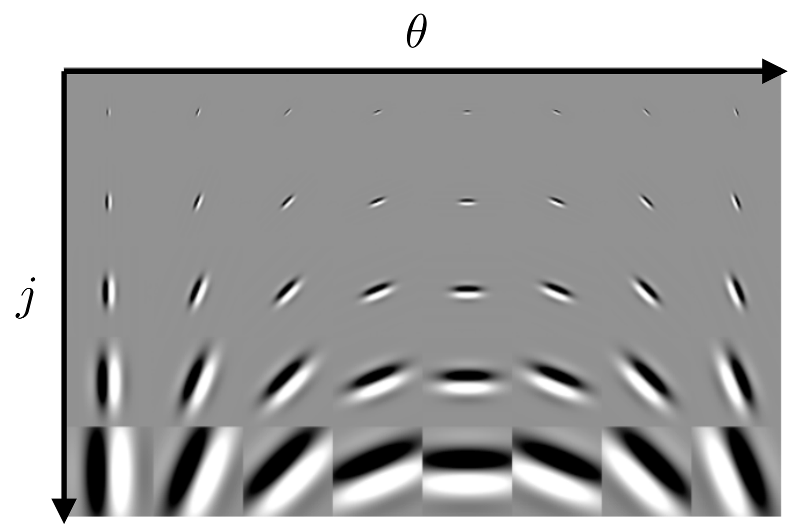
$|r_k - r_l| \gg 0$

Wavelets

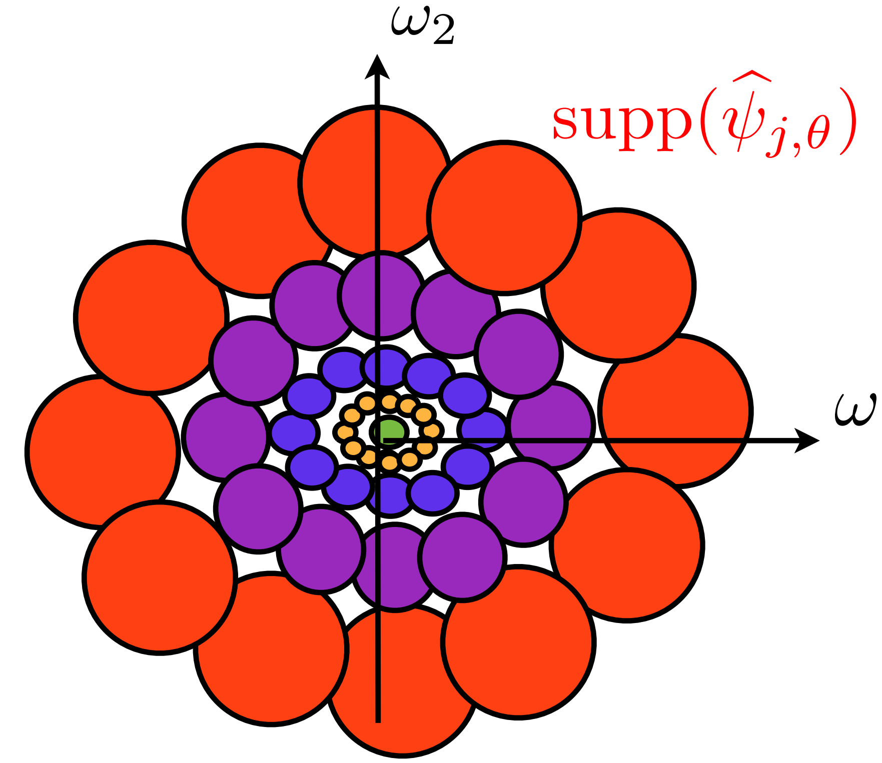
- Wavelet $\psi(u) = e^{-|u|^2}(e^{i\eta_0 \cdot u} - C), \qquad \int \psi = 0$
- Dilated and rotated:
  $\psi_{j,\theta} = 2^{-3j}\psi(2^{-j}R_\theta^{-1}u), \quad j \in \mathbb{Z}, \;\; R_\theta \in \mathrm{O}(3)$



real($\psi_{j,\theta}$)



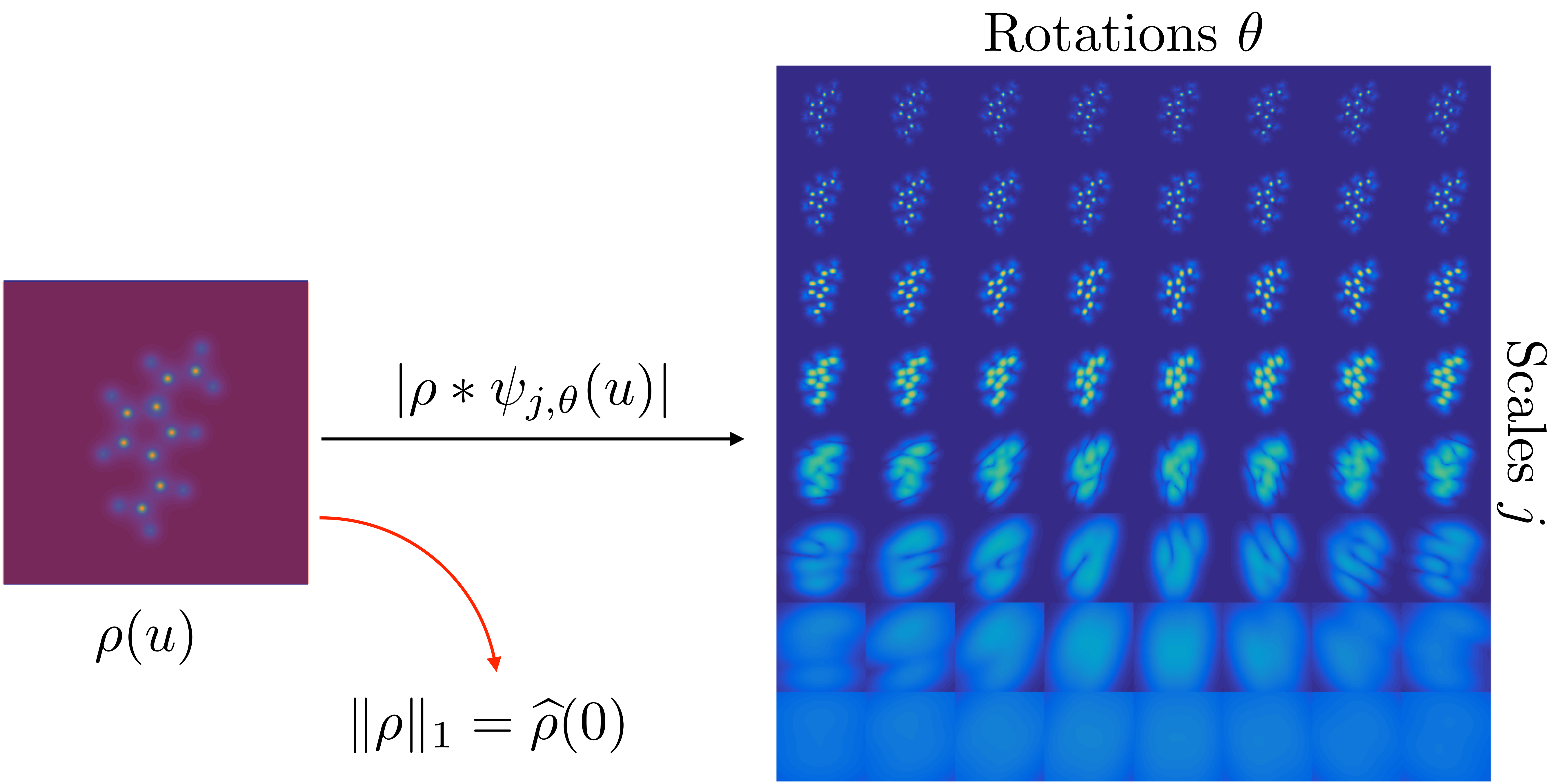
imag($\psi_{j,\theta}$)



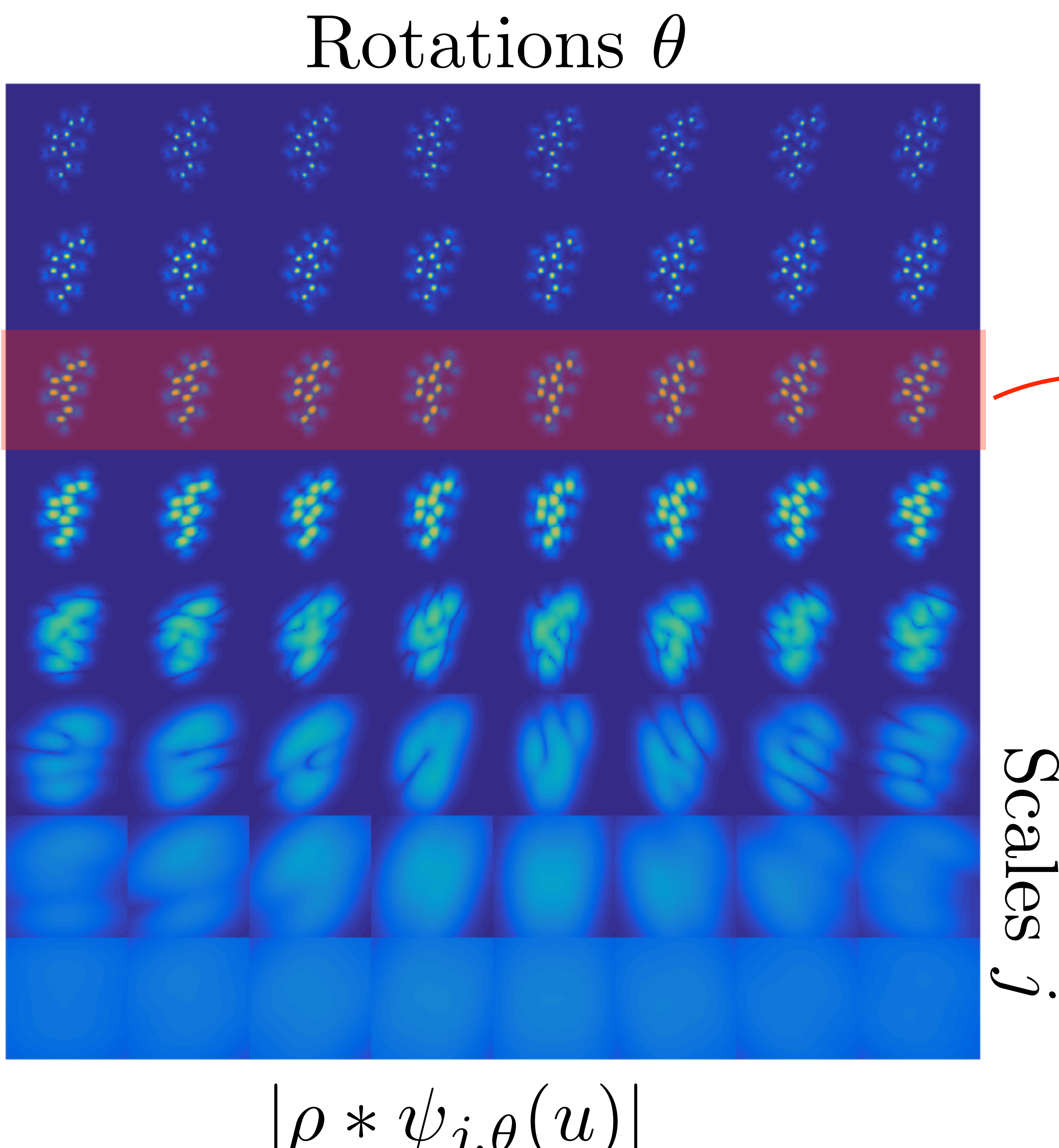
<u>Wavelet Transform:</u> $\rho \mapsto \{\int \rho, \;\; \rho * \psi_{j,\theta}(u)\}$

Interactions at scale 2^j in direction θ

Wavelet modulus

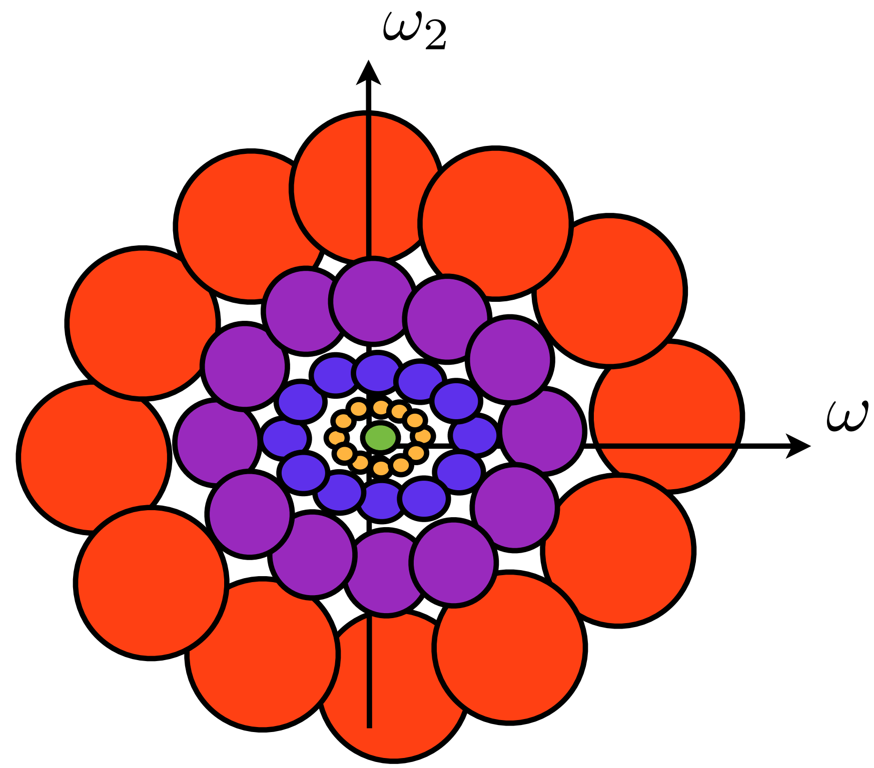


Wavelet invariants



$$\|\rho * \psi_{j,\cdot}\|_1 = \int_{\mathrm{O}(3)} \int_{\mathbb{R}^3} |\rho * \psi_{j,\theta}(u)| \, du \, d\theta$$

Invariant wavelet operator



Invariant wavelet operator:

$$\Theta\rho(j) = \|\rho * \psi_{j,\cdot}\|_1$$

Pros:

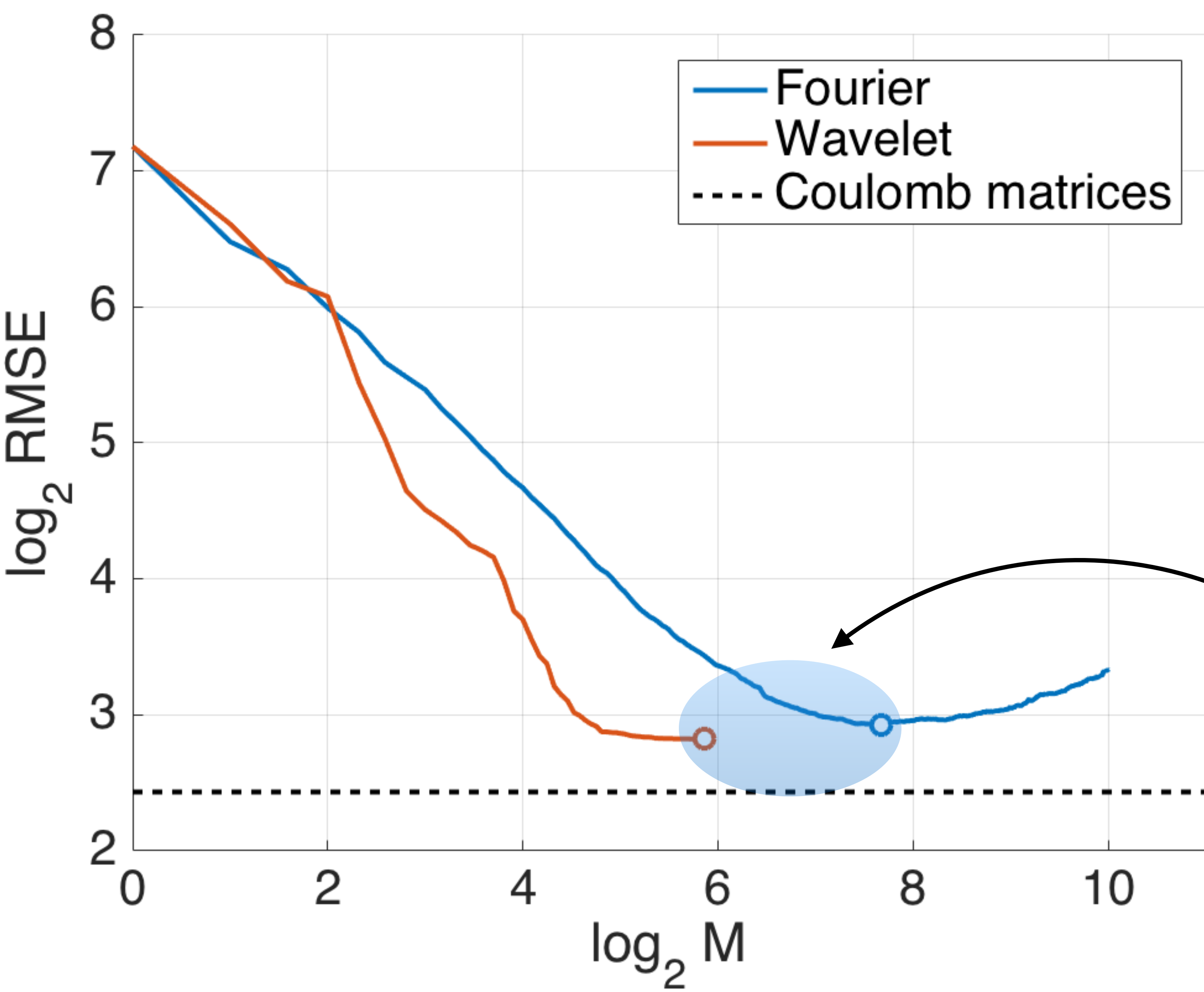
- Isometry invariant
- Stable to deformations (*Mallat*)
- Diagonalizes Coulomb and is sparse:

$$U(\rho) = \sum_{j=\log\epsilon}^{-\log\epsilon} w_j \|\rho * \psi_{j,\cdot}\|_2^2 + O(\epsilon)$$

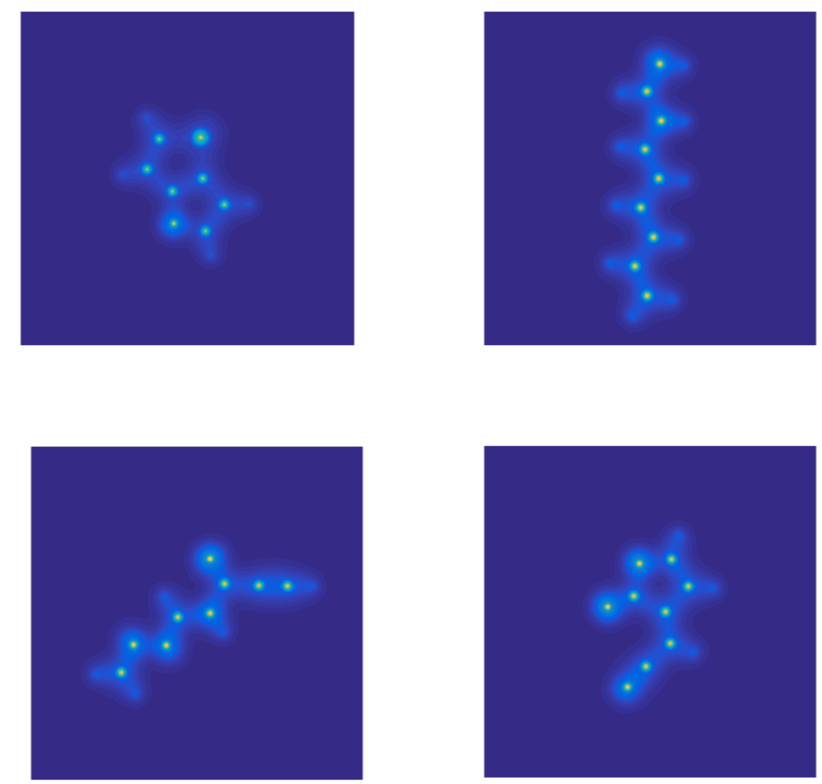
Cons: Encoding the invariants removes a lot of information

M-term regression error

$$\widetilde{f}_M(x) = \sum_{m=1}^{M} w_{k_m} \phi_{k_m}(x)$$



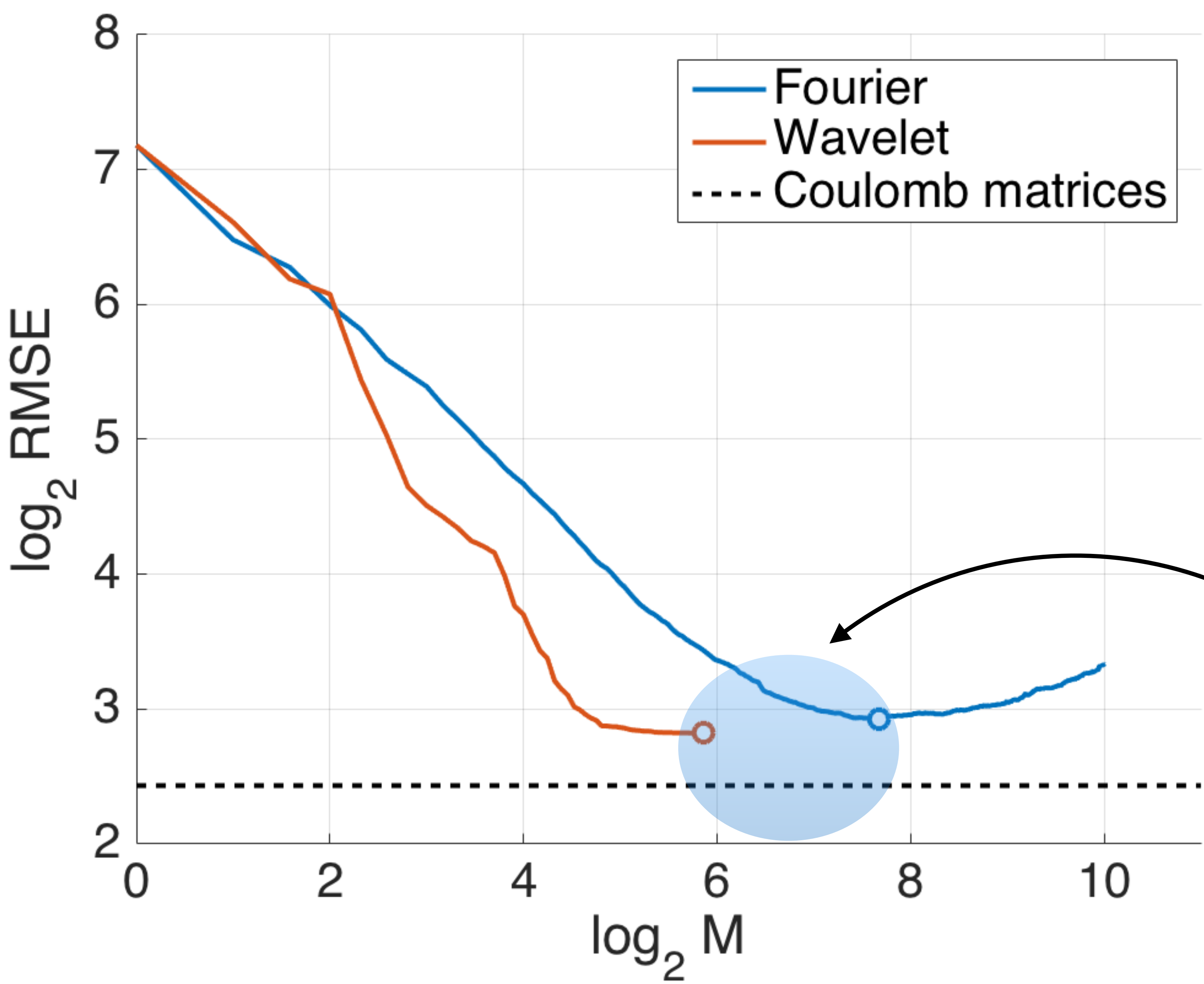
Data set:
Planar organic molecules



Multiscale wavelet dictionary
is sparser than Fourier dictionary

M-term regression error

$$\widetilde{f}_M(x) = \sum_{m=1}^{M} w_{k_m} \phi_{k_m}(x)$$



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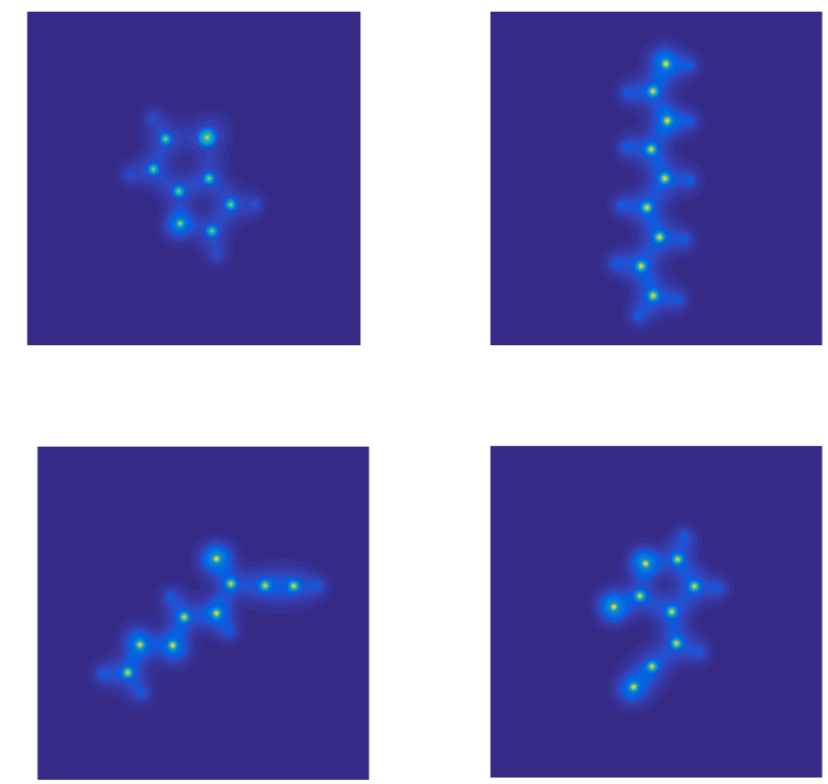
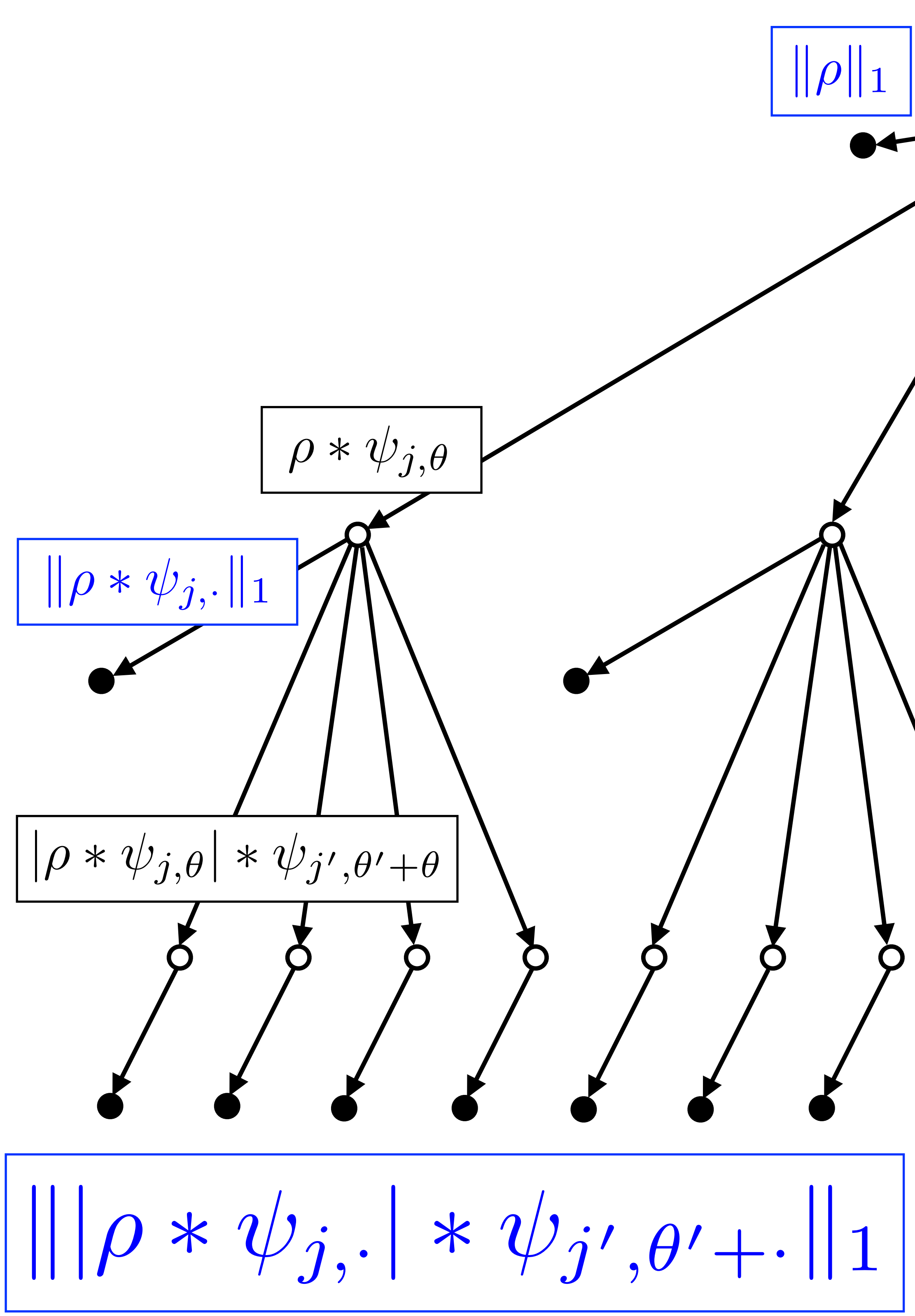


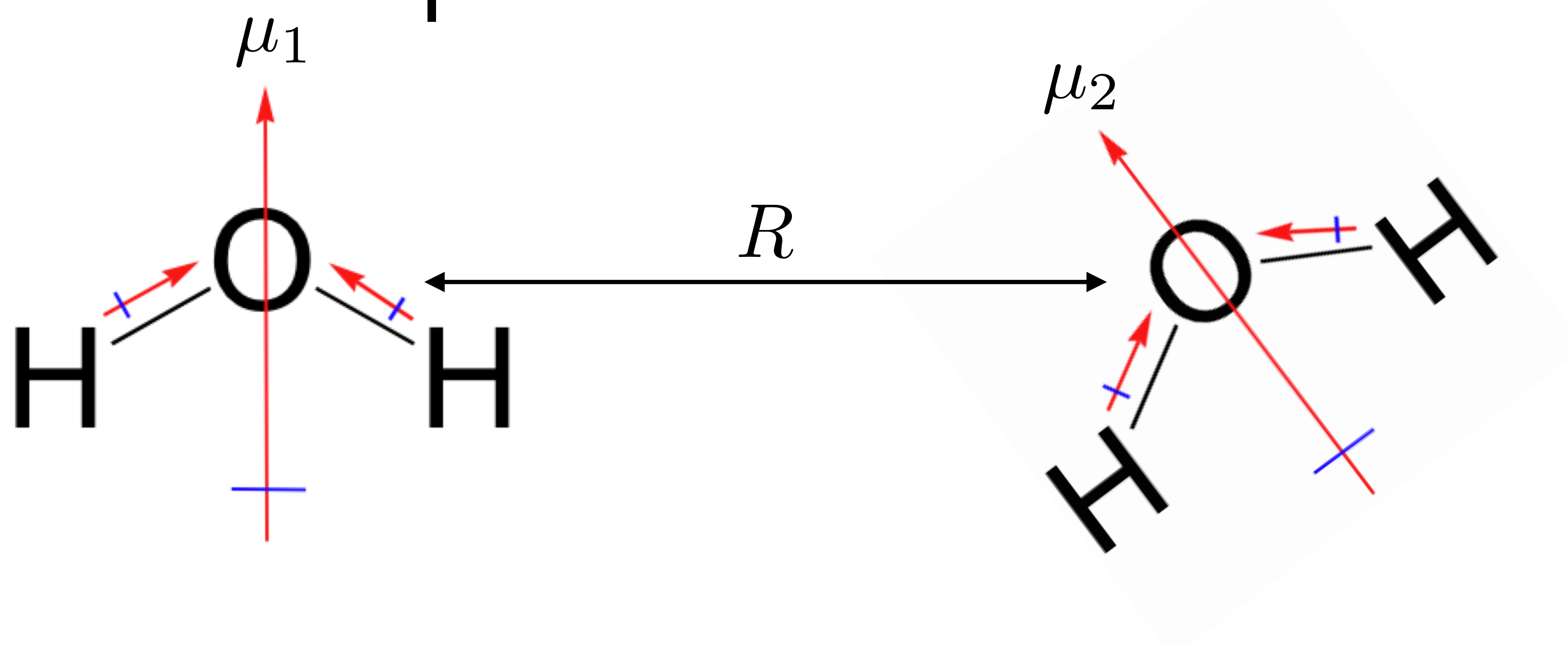
Coulomb matrices outperform both Fourier and wavelets

Invariant scattering operator



Couples wavelet features at two scales to encode complex interactions that move across scales

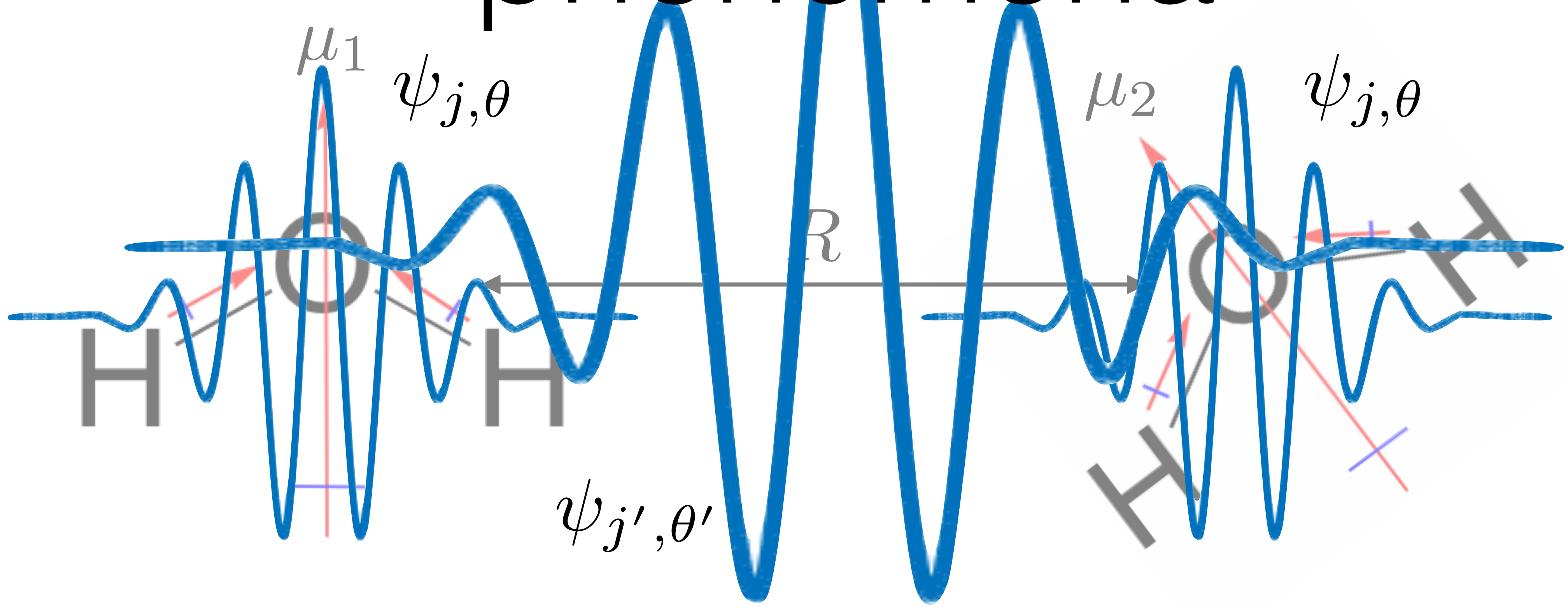
Coupled multiscale phenomena



Multipole expansion dipole-dipole moment:

$$\frac{\mu_1 \cdot \mu_2}{R^3}$$

Coupled multiscale phenomena



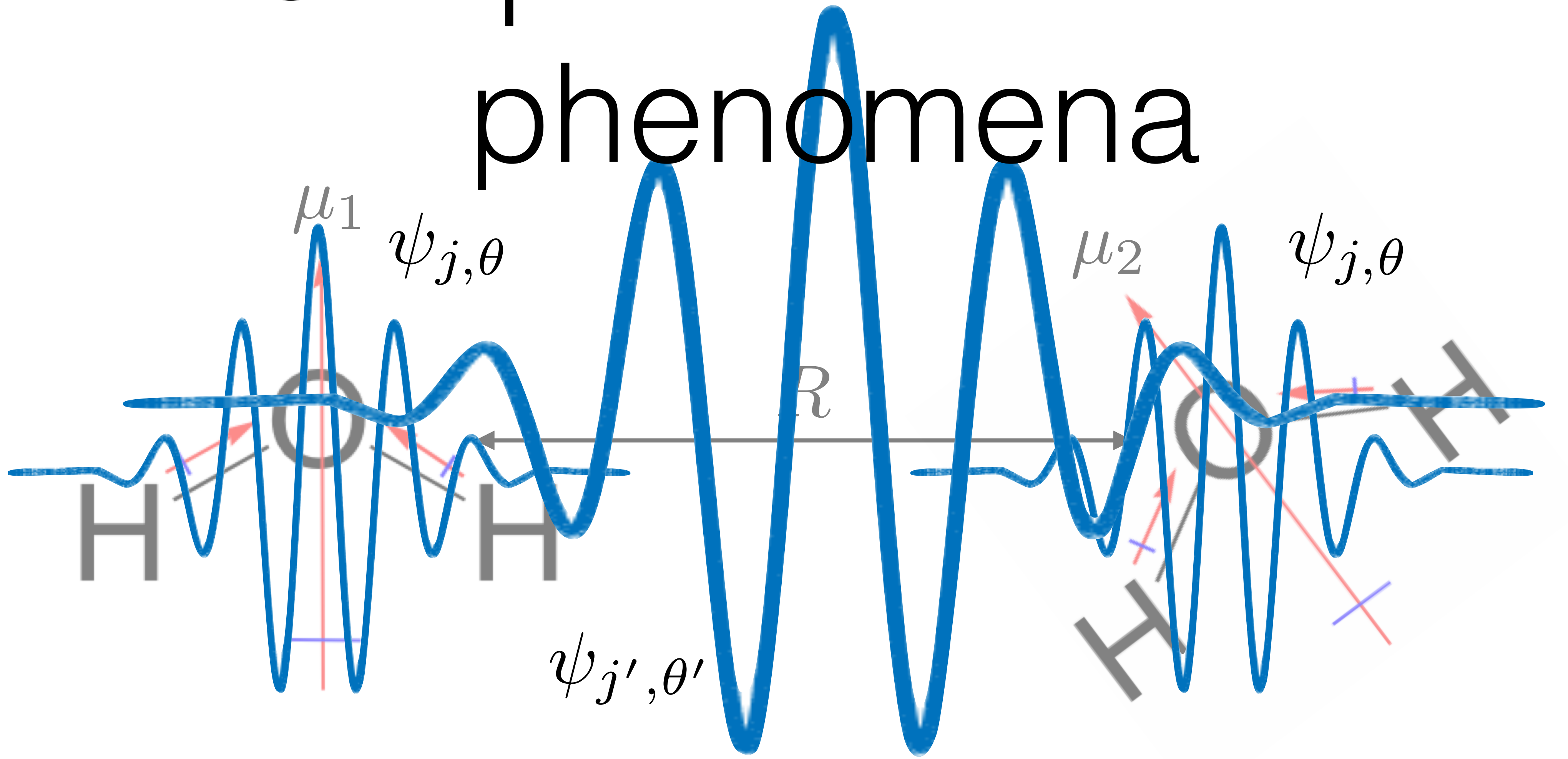
Small scale wavelets $\psi_{j,\theta}$ can learn dipole orientations

Large wavelets $\psi_{j',\theta'}$ can learn the distance between molecules

But there is no linear expansion of the dipole-dipole moment as:

$$\frac{\mu_1 \cdot \mu_2}{R^3} \neq \sum_{j \text{ small}} w_j \|\rho * \psi_{j,.}\|_1 + \sum_{j' \text{ large}} w_{j'} \|\rho * \psi_{j',.}\|_1$$

Coupled multiscale phenomena



2^{nd} order scattering features on the other hand:

First apply the small scale wavelet transform $\rho * \psi_{j,\theta} \ldots$

…then apply the large scale wavelet transform $|\rho * \psi_{j,\theta}| * \psi_{j',\theta'+\theta}$

Intuition and numerical evidence supports (theory in progress):

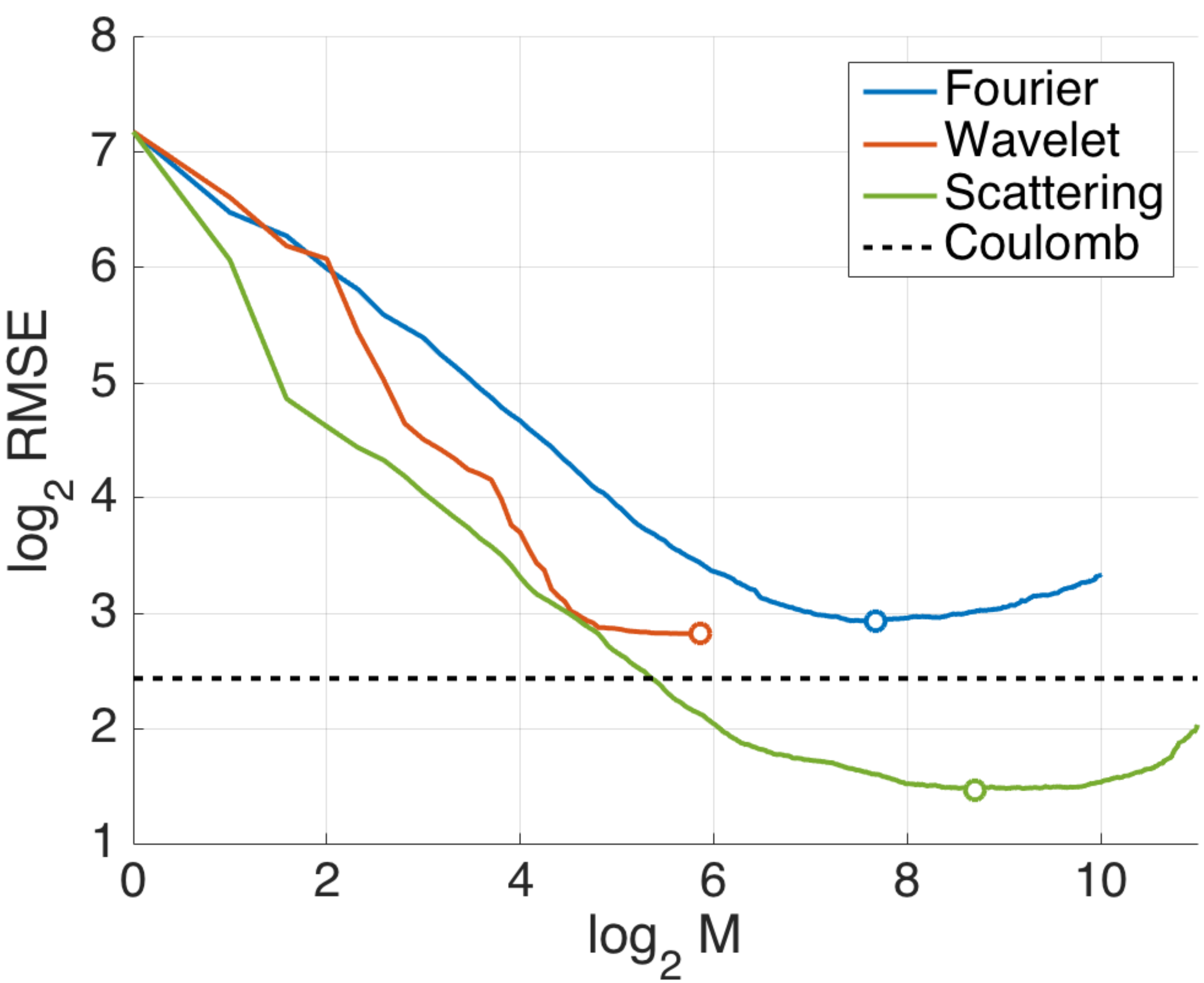
$$\frac{\mu_1 \cdot \mu_2}{R^3} \approx \sum_{j \text{ small}} \sum_{j' \text{ large}} \sum_{\theta'} w_{j,j',\theta'} \| |\rho * \psi_{j,\cdot}| * \psi_{j',\theta'+\cdot} \|_1$$

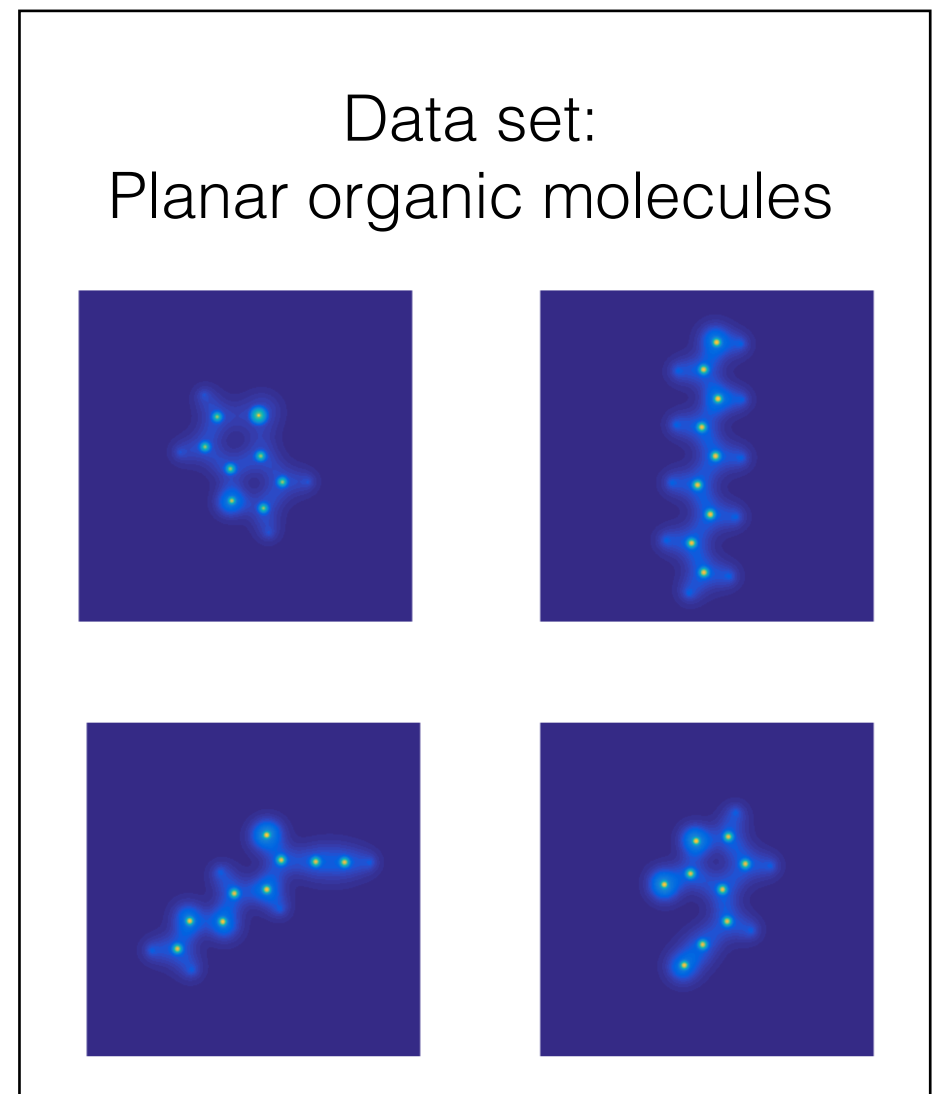
M-term regression error

$$\widetilde{f}_M(x) = \sum_{m=1}^{M} w_{k_m} \phi_{k_m}(x)$$

Errors (kcal/mol)

	Coulomb	Fourier	Wavelet	Scattering
MAE	2.4	5.3	5.4	1.7
RMSE	5.4	7.2	7.1	2.6

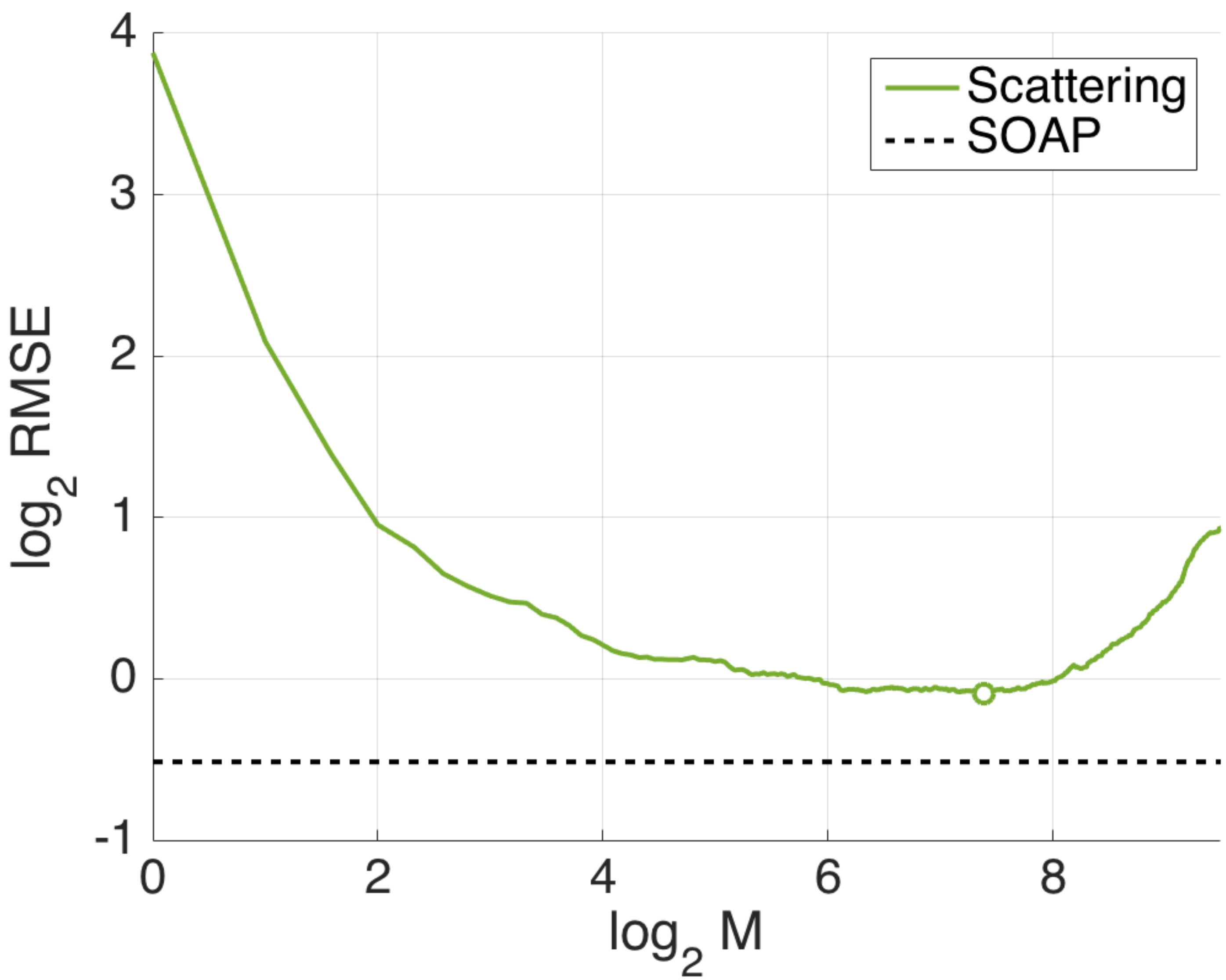




Water Molecules

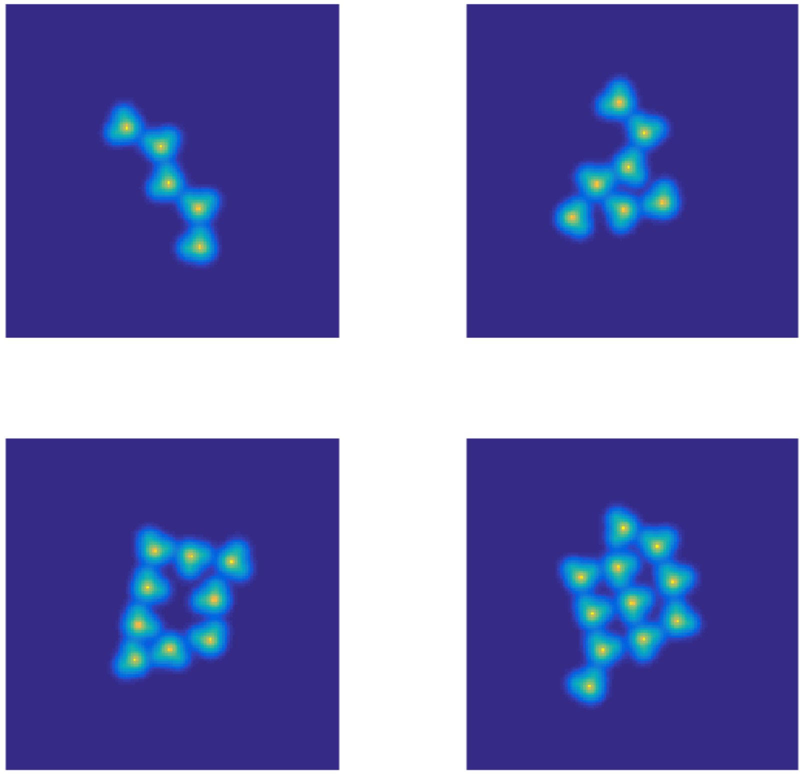
Root mean square error (meV/atom)

	SOAP	Scattering
700 Training	0.70	0.94



Long range interactions are $O(1/R^3)$

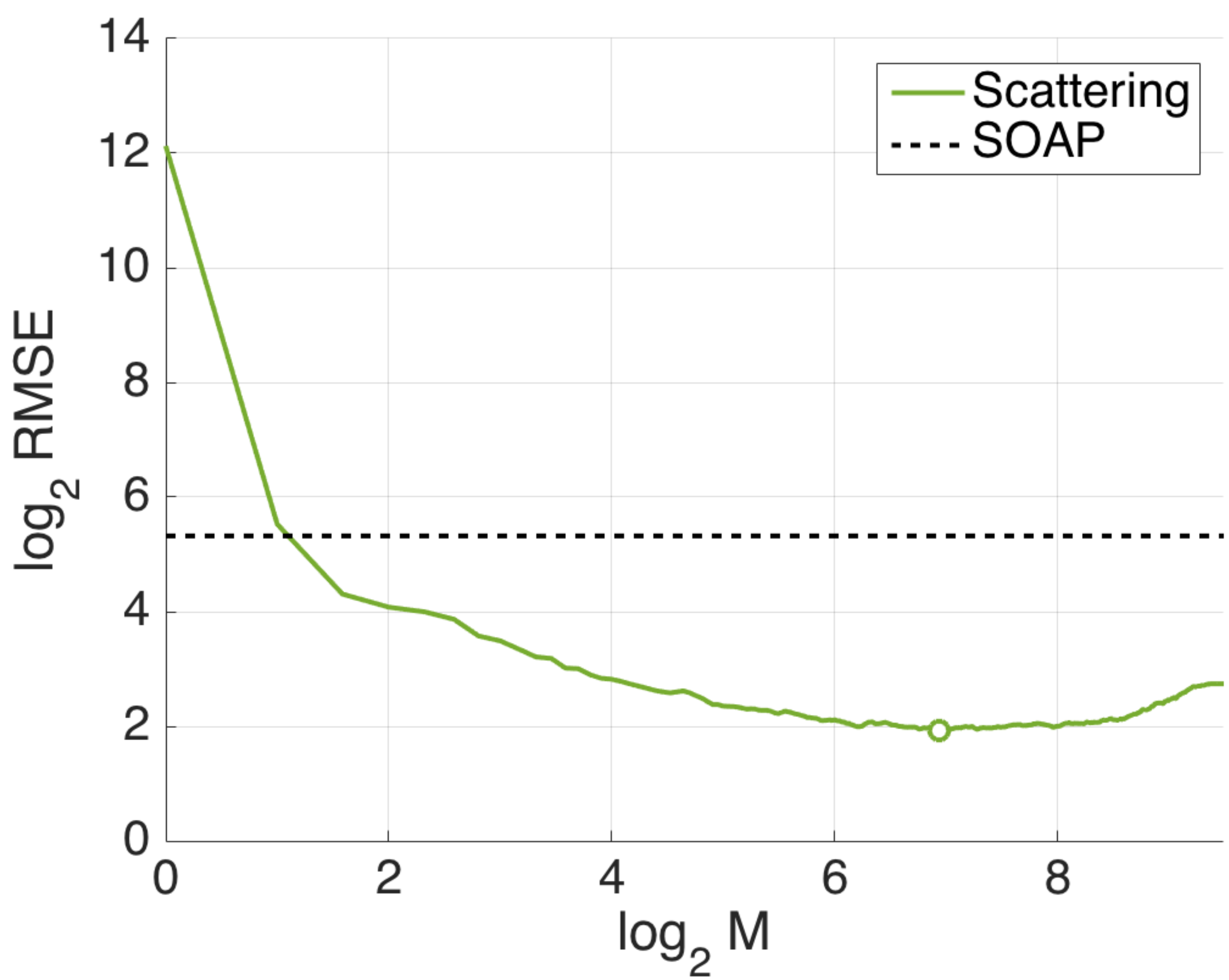
Data set:
Water molecules



Water Lithium Ion Systems

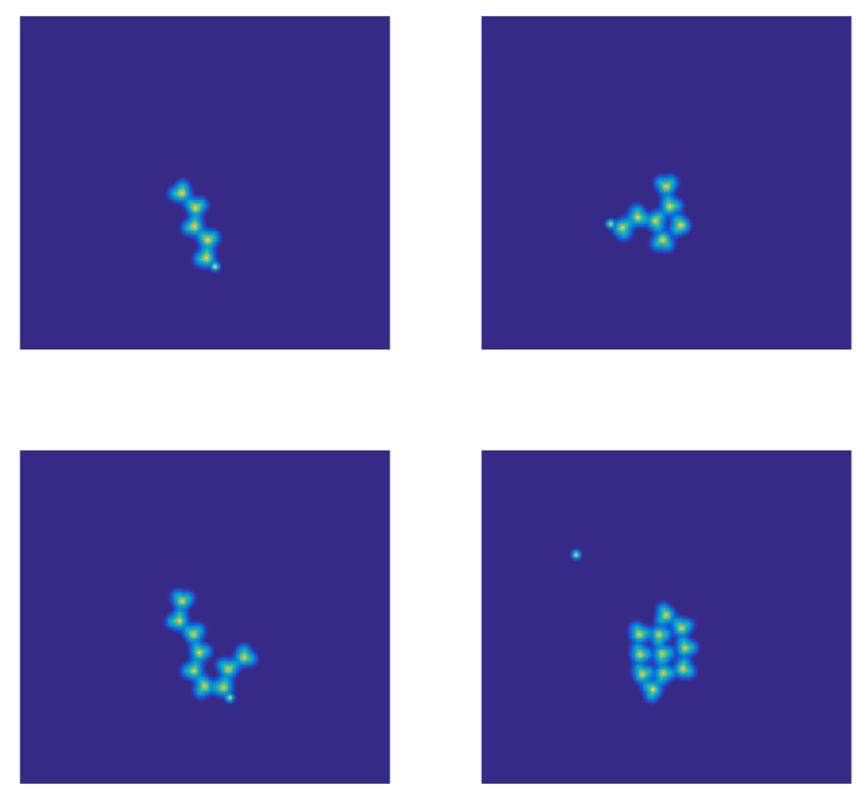
Root mean square error (meV/atom)

	SOAP	Scattering
700 Training	40	3.8



Long range interactions are $O(1/R^2)$

Data set:
Water & Lithium Ion



Future directions

- Efficient 3D software
- Rigorous link of scattering second layer with multipole methods
- Theoretical analysis of kinetic and exchange-correlation energies
- Utilization of forces
- Extensions to solid state physics, drug discovery, other many body problems in physics, other physical systems exhibiting complex multiscale behavior
- Connections with deep learning

Conclusions

To apply machine learning to many body problems, we must attack the curse of dimensionality. To do so we need to:

- Interpolate in low dimensional approximation spaces by…
- …nonlinear separation of original variables into learned non-interacting variables.

To achieve these goals, we utilize:

- Scale and angular separation over groups (translation, rotation)
- Symmetry and invariance properties
- Stability to deformations
- Cascade of operators (wavelet modulus) with the previous three properties to recover lost information