Modelling quantum states using normalizing flows

Álvaro Fernández, Yahya Saleh, Andrey Yachmenev, Jochen Küpper

Controlled Molecule Imaging Group

Center for Free-Electron Laser Science, Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany
Department of Physics, Universität Hamburg
Department of Chemistry, Universität Hamburg
Center for Ultrafast Imaging, Universität Hamburg

CUI THE HAMBURG CENTER FOR ULTRAFAST IMAGING

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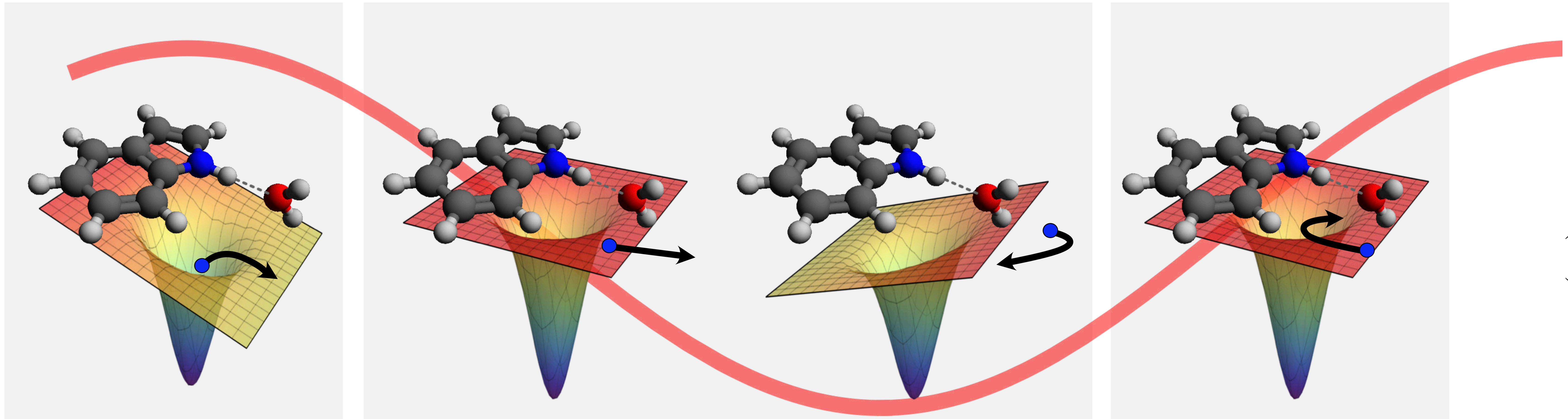
CFEL SCIENCE
Controlled Molecule Imaging

DESY.

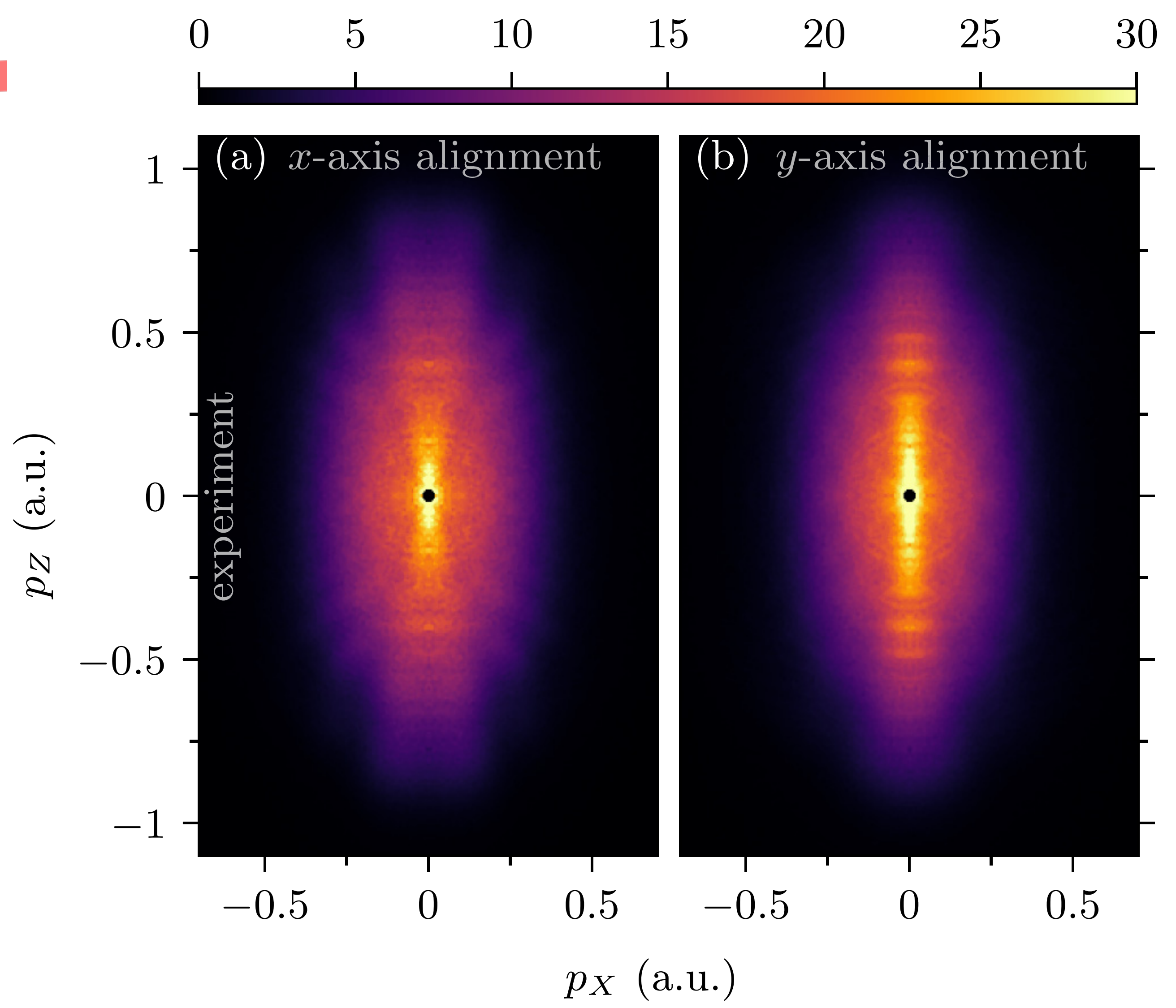
Objectives

- Create simulations for the LIED experiment.

Ultrafast imaging of dissociation dynamics with LIED



Ionisation | *Propagation* | *Rescattering*



Quantum | Classical | Classical → **solution of classical ODEs with neural networks, PAD features ↔ trajectory type**

Quantum | Quantum | Quantum → **solution of quantum ODEs with neural networks (LSTM, quantum flows, …)**

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Time Independent Schrödinger Equation in Molecules

- Partial Derivative Eigenvalue equation:

$$H(x_1, x_2, \dots)\Psi_i(x_1, x_2, \dots) = [-\frac{1}{2}\Delta_i^2 + V(x_1, x_2, \dots)]\Psi_i(x_1, x_2, \dots) = E_i\Psi_i(x_1, x_2, \dots)$$

- Generally not solvable due to high degree of freedom.
- Need of numerical approximations.
  - Basis expansion:

$$\Psi_i(x_1, x_2, \dots) = \sum_{i_1, i_2, \dots = 0}^{N} c_{i_1, i_2, \dots}\phi_{i_1}(x_1)\phi_{i_2}(x_2)\dots$$

  - How can we improve this approximation?

Normalizing flows

- Augment the basis by a flow of the intrinsic coordinates to a support space. Let this flow be a Neural Network:

$$(x_1, x_2, \ldots, x_n) = g_\theta(q_1, q_2, \ldots, q_n) \longrightarrow H(x_1, x_2, \ldots, x_n) = H_\theta(q_1, q_2, \ldots, q_n)$$

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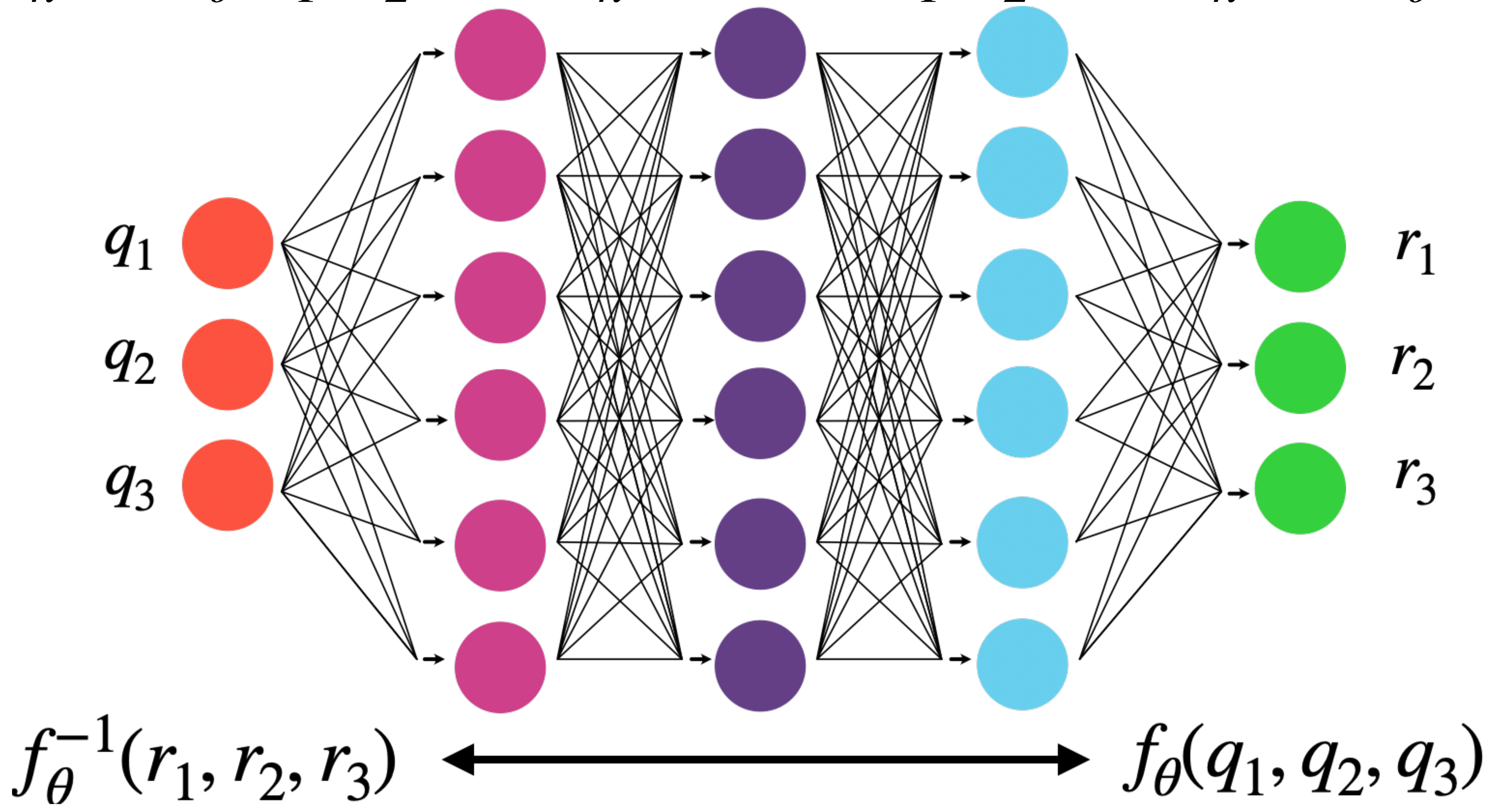

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[1] Invertible Residual Networks. Jens Behrmann, Will Grathwohl, Ricky T. Q. Chen, David Duvenaud, Joern-Henrik Jacobsen. arXiv:1811.00995

Normalizing flows

- We can use support space for performing integrals:

$$< \phi_i^A | \phi_j^A > = \int \phi_i^*(q)\phi_j(q)dq = \delta_{i,j}$$

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- Using many states variational principle, the eigenvalues of the Hamiltonian matrix ($\lambda_i(\theta)$) can only be bigger or equal to the real energies:

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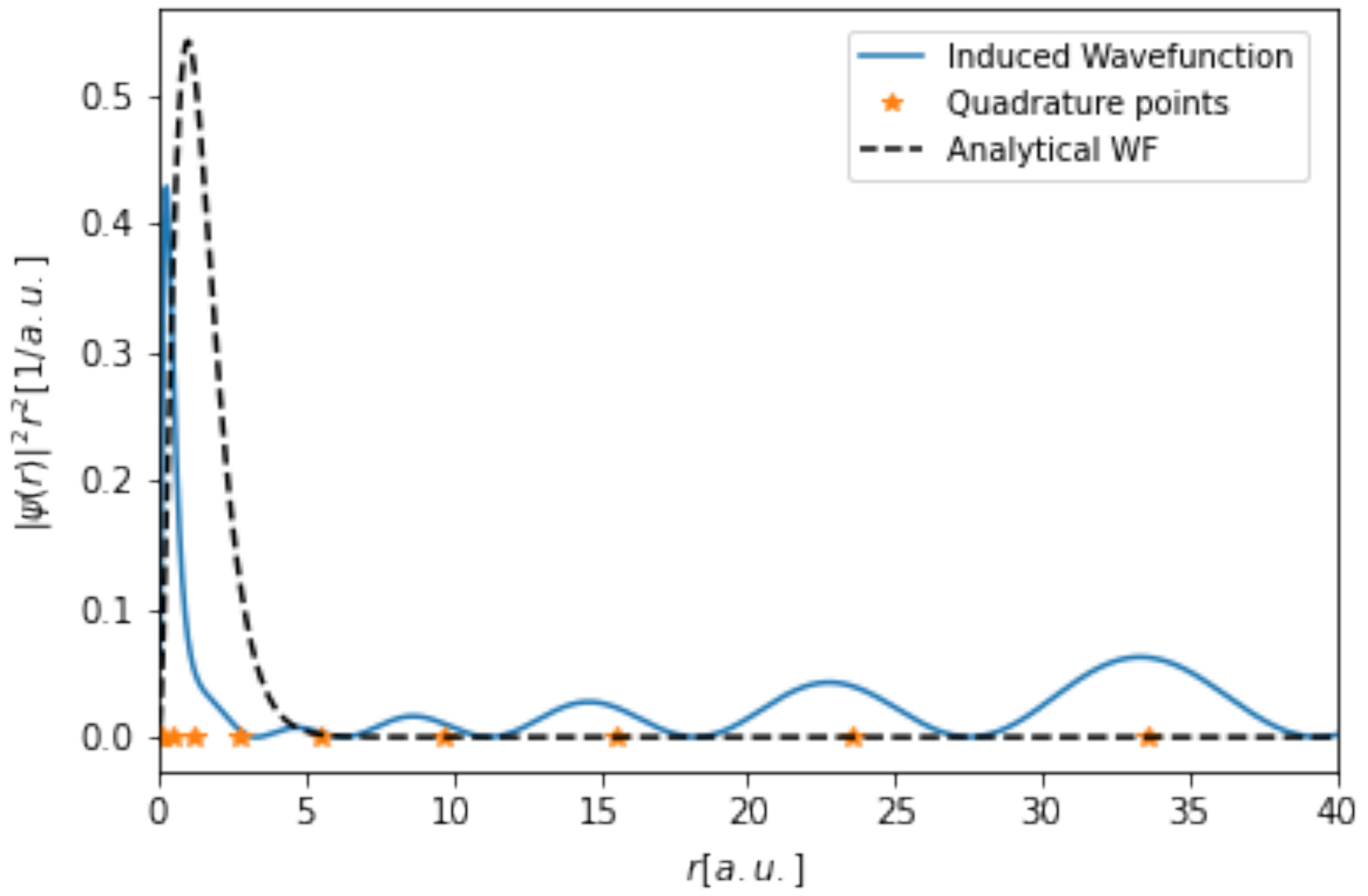
- We can apply numerical approximations for integrals in support space. How?

Integrals using quadratures

- Very precise integrals. Memory usage scales approximately as N^{dim}, so very useful for low dimensional problems.
- Drawback: during training, wave function is only studied at quadrature points. Resulting eigenfunction is biased.



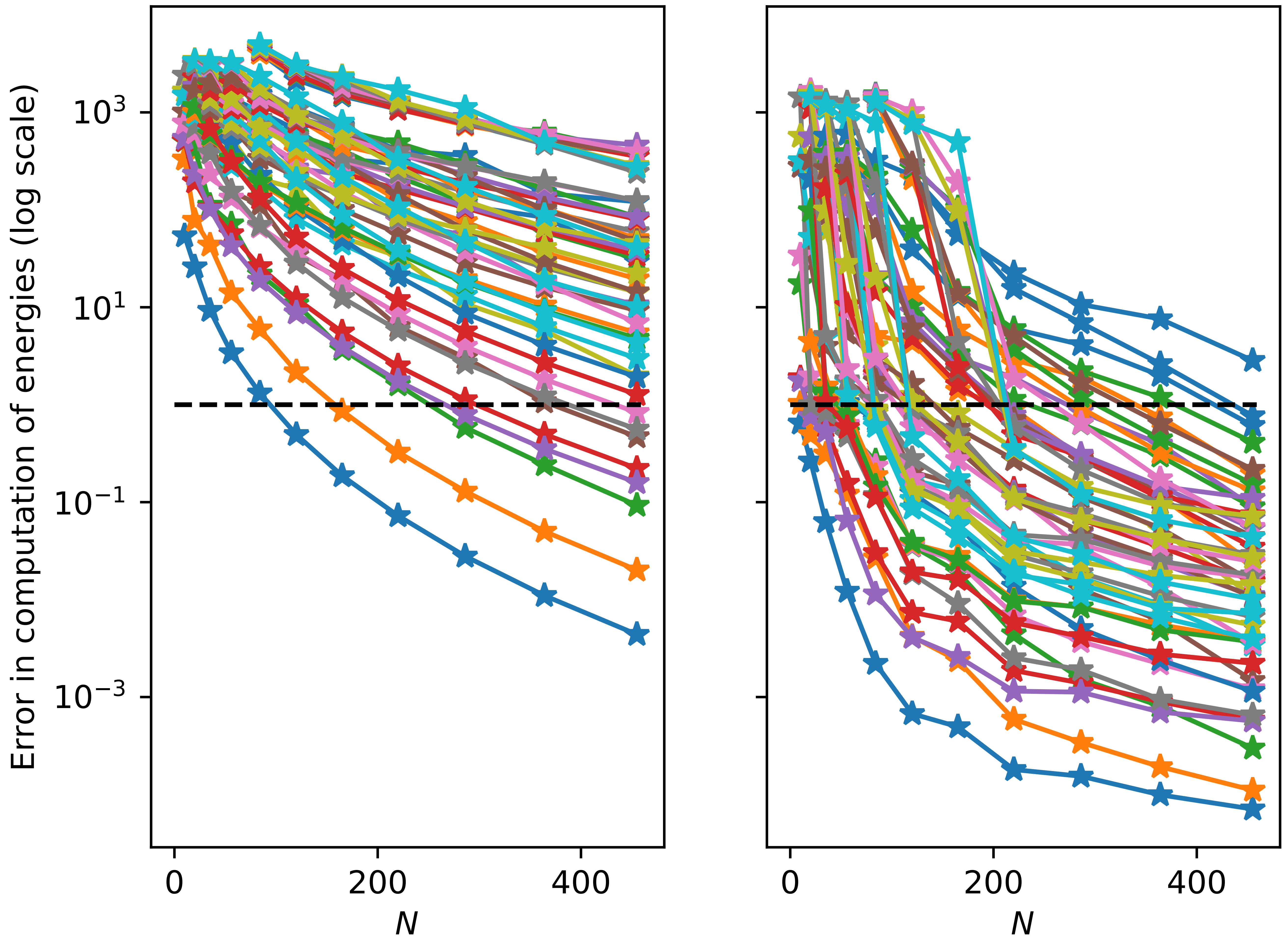
Integrals using Langevin MonteCarlo

- Starting from a point in space, create a Markov Chain that accepts points based on a density. Using $\rho_i(q) = |\phi_i^A(q)|^2$ as our density, we obtain a distribution of points $q_\alpha \sim |\phi_i^A(q)|^2$. Then any integral can be approximated by:

$$< \phi_i^A | f(q) | \phi_j^A > = \int \rho_i(q) \frac{f(q)\phi_j^A(q)}{\phi_i^A(q)} \simeq \frac{1}{M} \sum_{q_\alpha \sim |\phi_i^A(q)|^2}^{M} \frac{f(q_\alpha)\phi_j^A(q_\alpha)}{\phi_i^A(q_\alpha)}$$

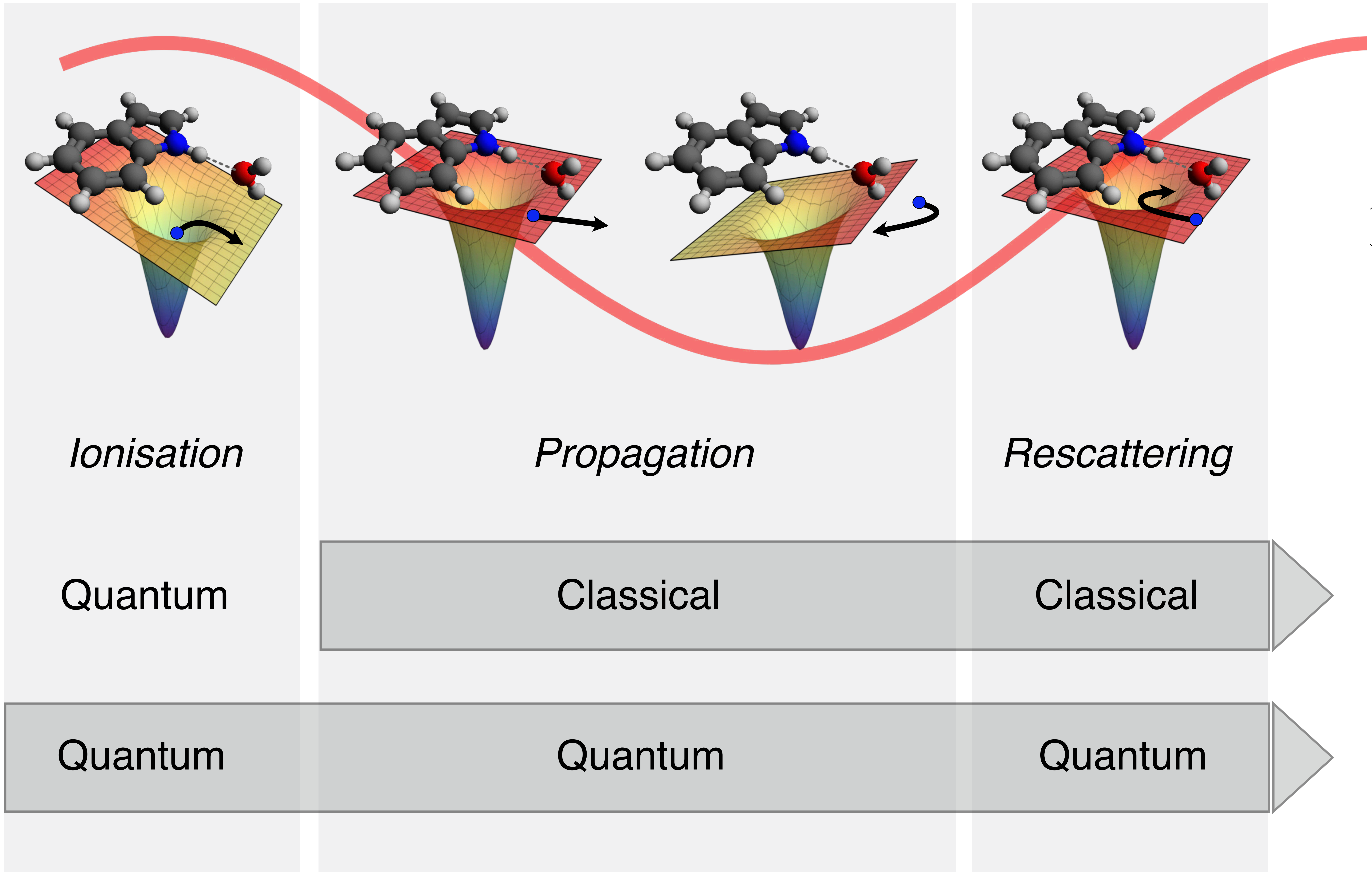
- The integral can cover all the space during training. To do so, choose M to be a big number and divide the training process in minibatches of m points.
- The memory usage scales now as $M \cdot dim$, of which only $m \cdot dim$ enter the training at the same time. Thus, it is more efficient for problems with many intrinsic degree of freedom.

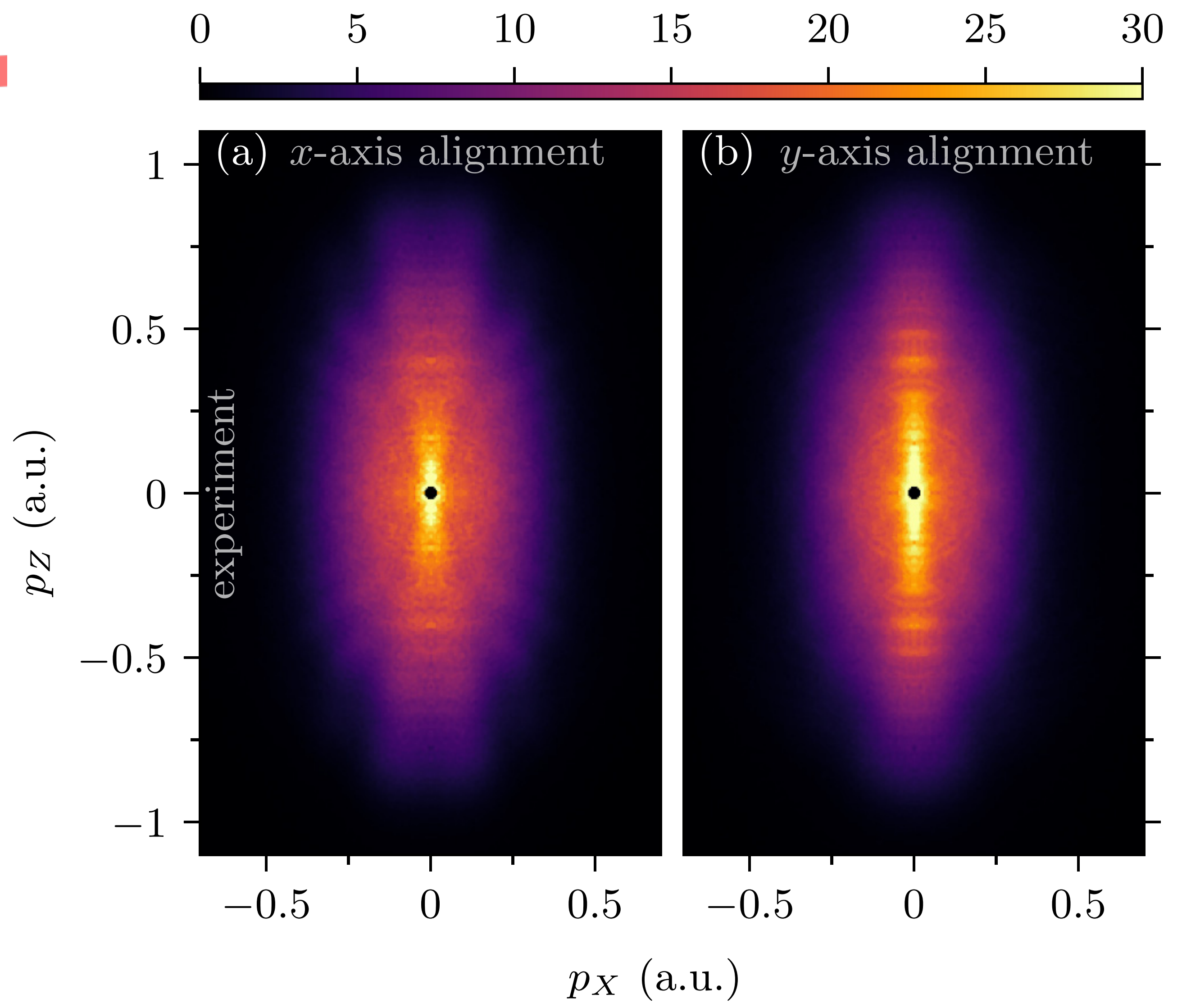
Results



- Outperform the convergence of classical basis expansion w.r.t. number of basis functions.
- Convergence for many excited states.
- Tested for vibrational and electronic states.

Ultrafast imaging of dissociation dynamics with LIED





solution of classical ODEs with neural networks,
PAD features ↔ trajectory type

solution of quantum ODEs with neural networks (LSTM, quantum flows, …)

Time Dependent Schrödinger Equation

- Partial Derivatives equation:

$$H(x_1, x_2, \ldots; t)\Psi(x_1, x_2, \ldots; t) = = i\partial_t \Psi(x_1, x_2, \ldots; t)$$

With boundary condition: $\Psi(x_1, x_2, \ldots; t = t_0) = \Psi_0(x_1, x_2, \ldots)$

- In the basis expansion approximation, initial condition is given by:

$$\Psi_0(x) = \sum_i^N c_i(t_0)\phi_i(x)$$

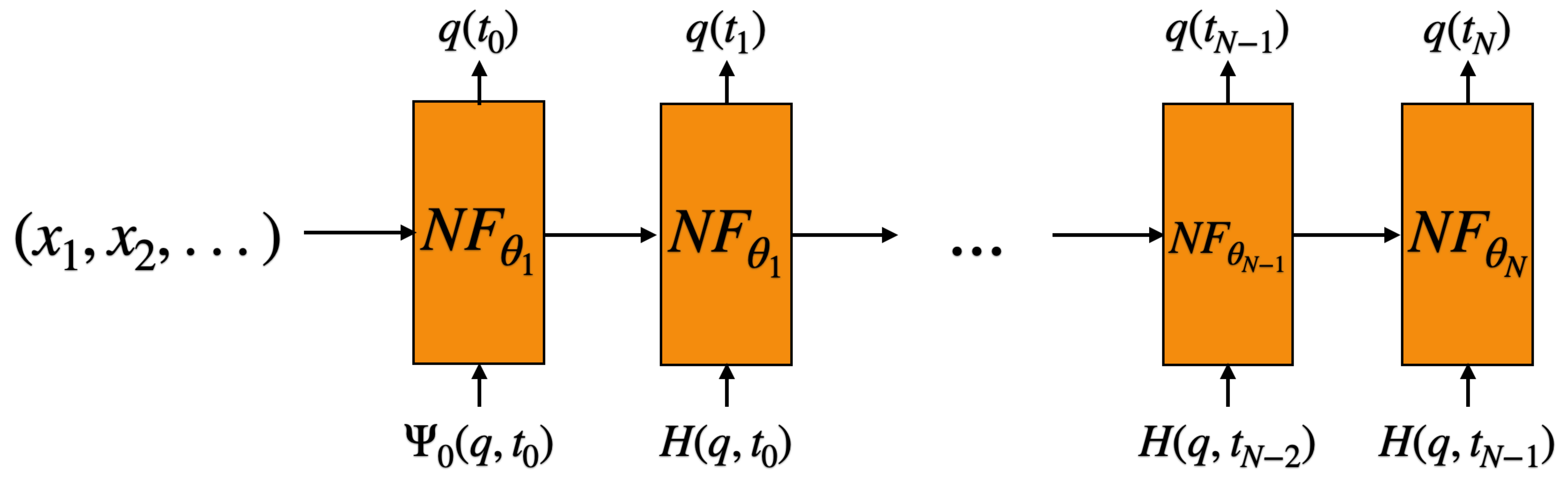
- Then, our approximation to the wave function is given by propagating the coefficients in small time steps at each time:

$$c_i(t) = \sum_j^N e^{<\phi_i(x)|H(x;t-\Delta t)|\phi_j(x)>} c_j(t - \Delta t)$$

- The approximation only holds for small time steps and a big number of basis functions.

Time Dependent Schrödinger Equation

- Can we use a similar strategy to solve a time dependent equation?
  - Hopefully! Using recurrent Neural Networks each for one time step:



  - We again create an augmented basis at each time:

$$\phi_i^A(q,t) = \phi_i(q)\sqrt{|\det \frac{\partial q}{\partial g_{\theta,t}^{-1}(q)}|}$$

  - And our approximation to the wave function:

$$\Psi^A(q,t) = \sum_i^N c_i(t)\phi_i^A(q,t)$$

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CFEL Controlled Molecule Imaging Group

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DFG

EXC Center for Ultrafast Imaging

EXC Advanced Imaging of Matter

SPP 1840 Quantum Dynamics in Tailored Light Fields

MC-ITN MEDEA

MC-ITN ASPIRE

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