Quantum Phase Estimation-based Quantum Computational Chemistry

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R. Sakuma, S. Kanno, K. Sugisaki, T. Abe, N. Yamamoto,
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AIP Adv. 14, 095021 (2024)

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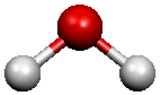
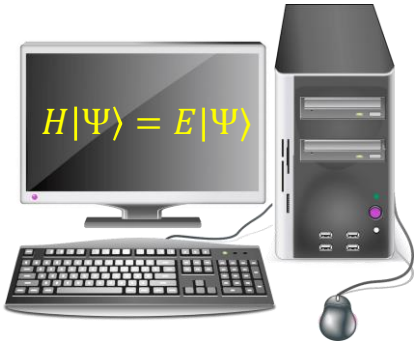
S. Kanno, K. Sugisaki, H. Nakamura, H. Yamauchi, R. Sakuma,
T. Kobayashi, Q. Gao, N. Yamamoto,
arXiv:2408.04946

Quantum chemical calculations

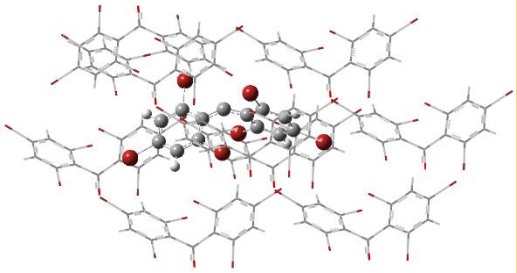
Solve a Schrödinger equation that is a **fundamental equation in quantum mechanics**, to theoretically reveal the **electronic structures of atoms and molecules**.



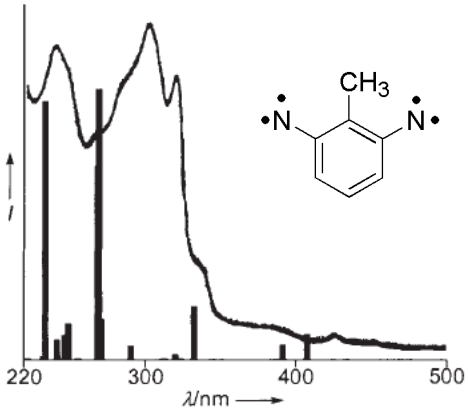
Atomic coordinates of molecules



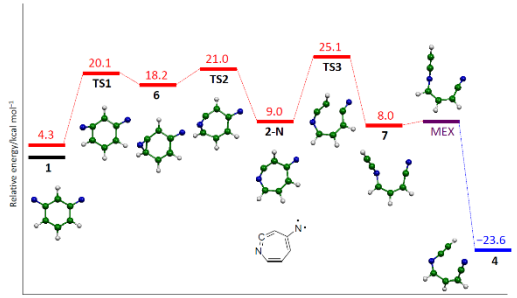
Total energy and wave function of the molecules at the input coordinates



Search for stable conformations



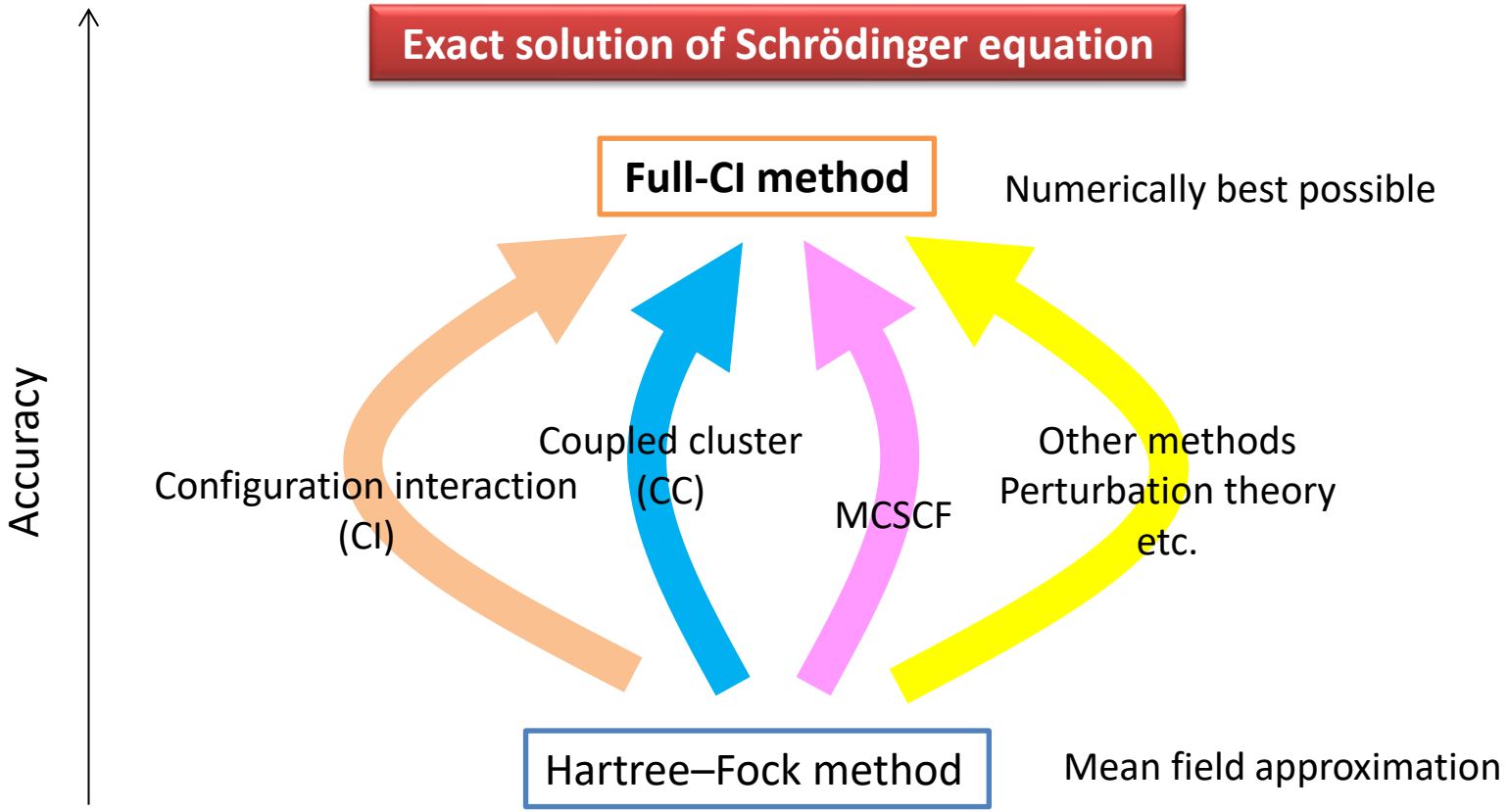
Molecular properties



Reaction pathways

Accurate quantum chemical calculations potentially open the door to "in silico" chemistry R&D

Quantum chemical calculations



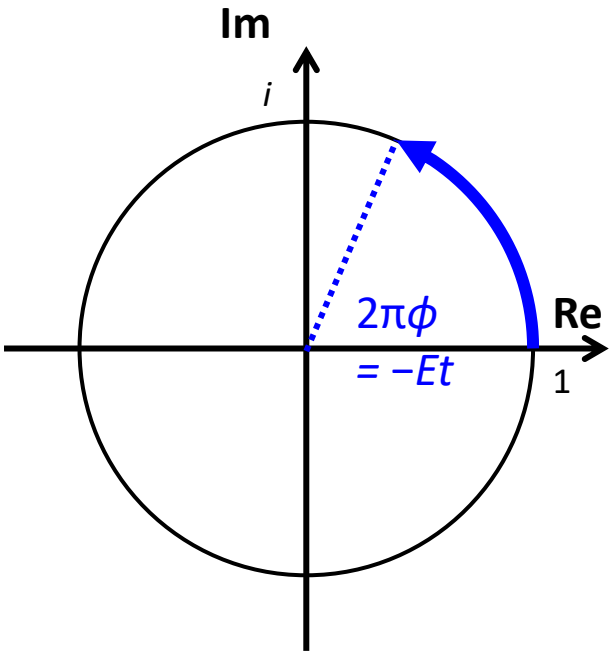
[Current status]

- **"Gold standard" approaches have been established**
  Such approaches become impractical for large molecules due to computational cost
- **Semi-quantitative or qualitative calculations are usually possible**
  Not "always" but "usually". QCC sometimes fail to predict experimental results even qualitatively

Quantum phase estimation (QPE)

$$\exp(-iHt)|\Psi\rangle = \exp(-iEt)|\Psi\rangle = \exp(i2\pi\phi)|\Psi\rangle$$

Time evolution of wave function → **Phase shift depending on the energy**

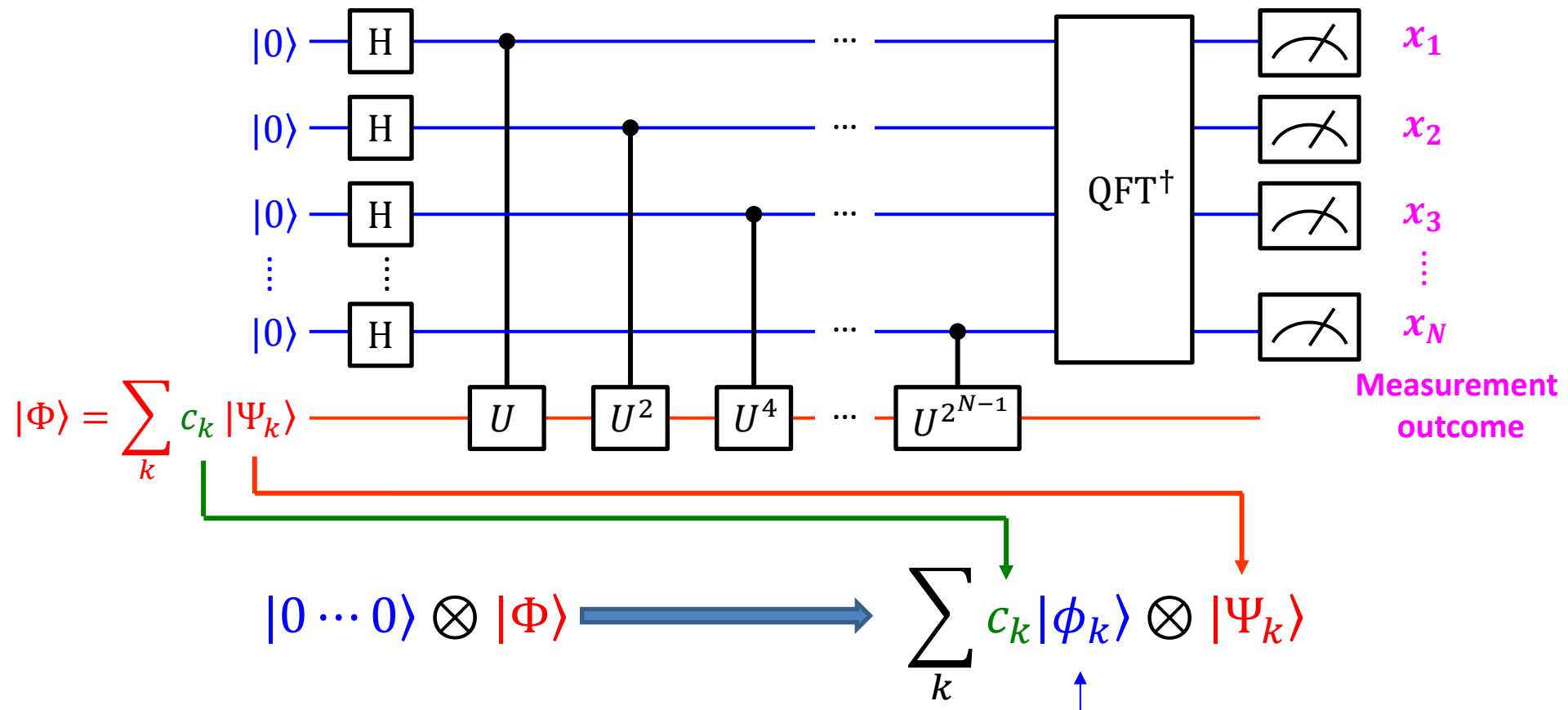


$$0 \leq \phi < 1$$

$$E = -\frac{2\pi\phi}{t}$$

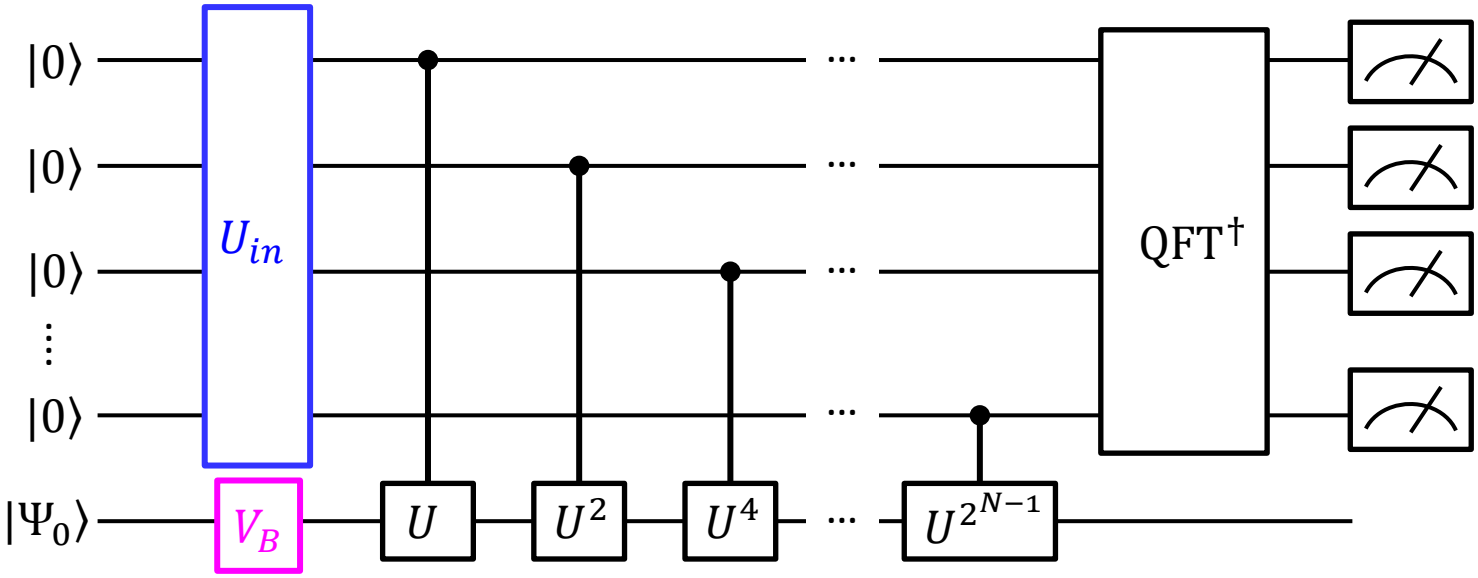
By determining the magnitude of phase shift caused by the time evolution, we can extract the energy eigenvalue!

Quantum phase estimation (QPE)



The probability of which electronic state is obtained in QPE is proportional to the square of overlap between the approximated and full-CI wave functions!

QPE-based dynamical response function calculations



$$U = \exp(-i(H - E_0)t)$$

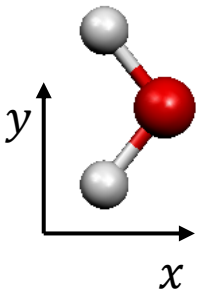
Theoretical calculations of spectroscopic properties is important.

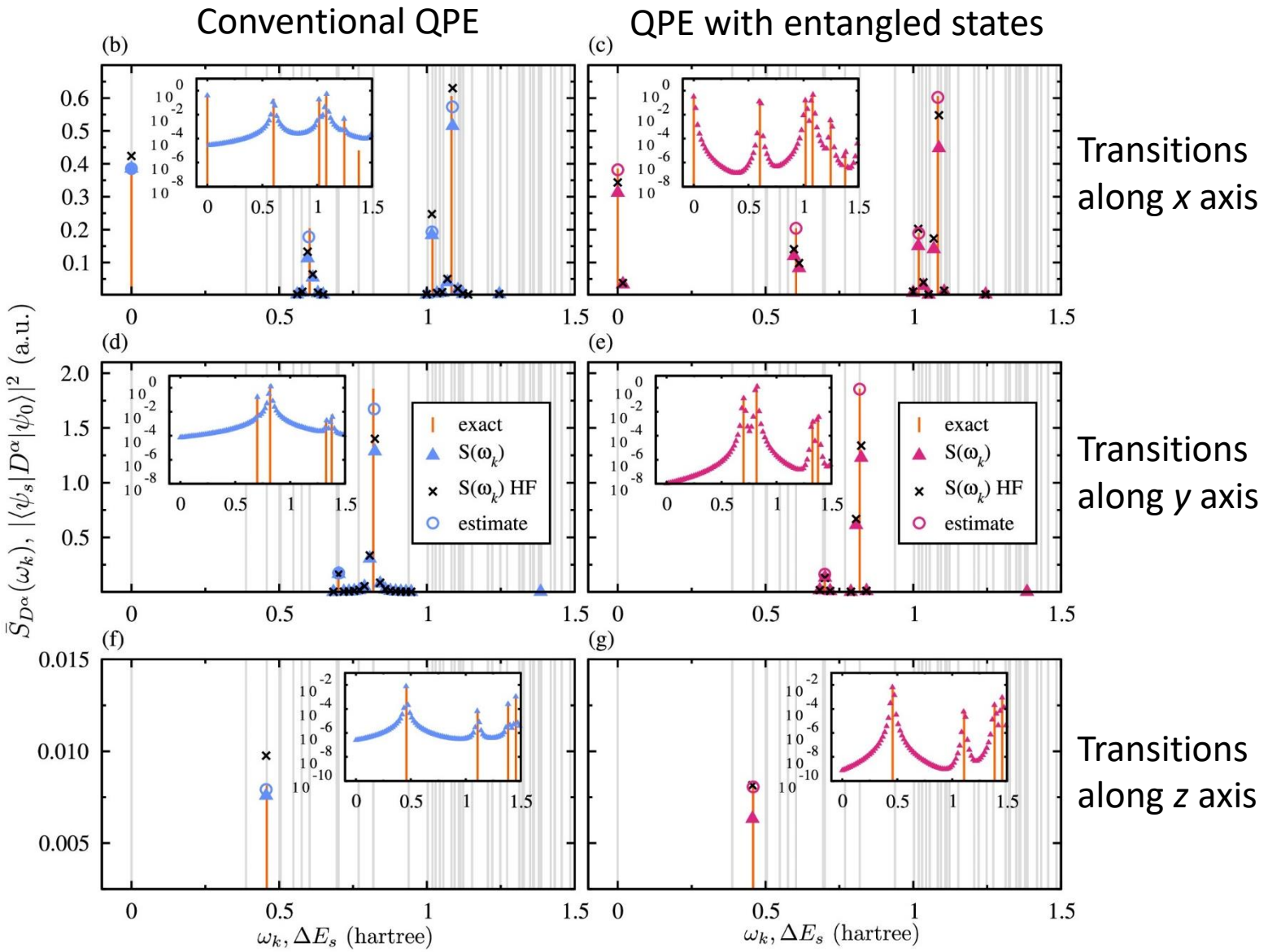
For example, photo-absorption spectra can be calculated by introducing the electron dipole operator as V_B in the QPE quantum circuit.

Conventional QPE suffers from the "spectral leakage" problem, which makes the spectral line broader.

By using **entangled states as the input for ancillary qubits**, the QPE spectral peaks are more localized, allowing us to estimate peak positions and heights very accurately!

QPE-based dynamical response function calculations



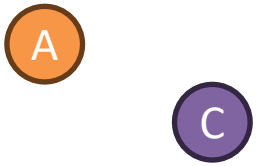


Transitions along *x* axis

Transitions along *y* axis

Transitions along *z* axis

Fragment MO method and size consistency



Sum of monomer energies

$$E^{\mathrm{FMO}} = E_A + E_B + E_C + (E_{A+B} - E_A - E_B) + (E_{B+C} - E_B - E_C) + (E_{C+A} - E_C - E_A)$$

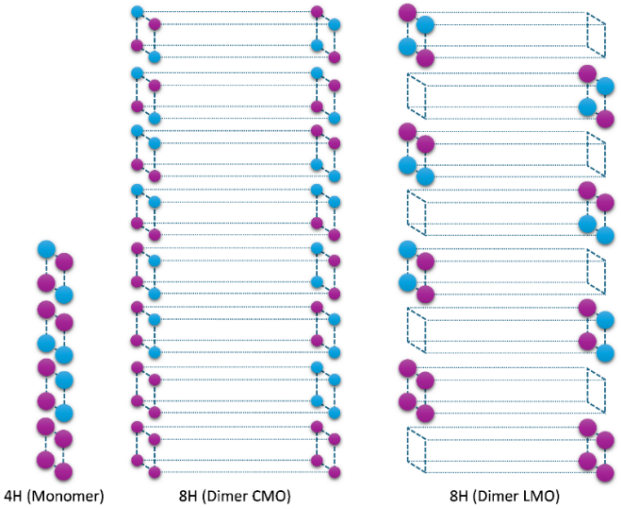
Interaction energy for fragments A–B

Interaction energy for fragments B–C

Interaction energy for fragments C–A

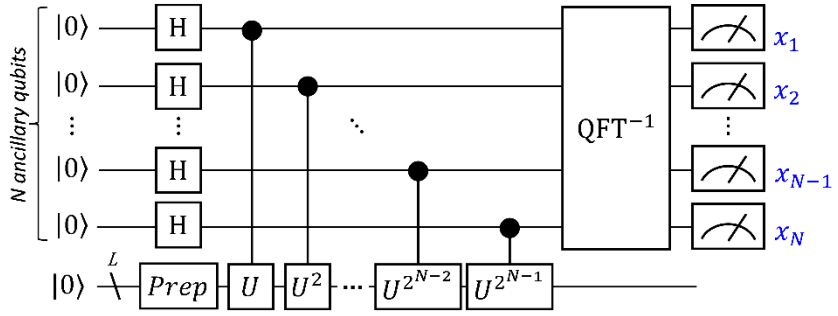
In the FMO-based quantum chemical calculations, $E(A \cdots A) = 2 \times E(A)$ for spatially well separated two molecules, known as **size consistency**, is essential.

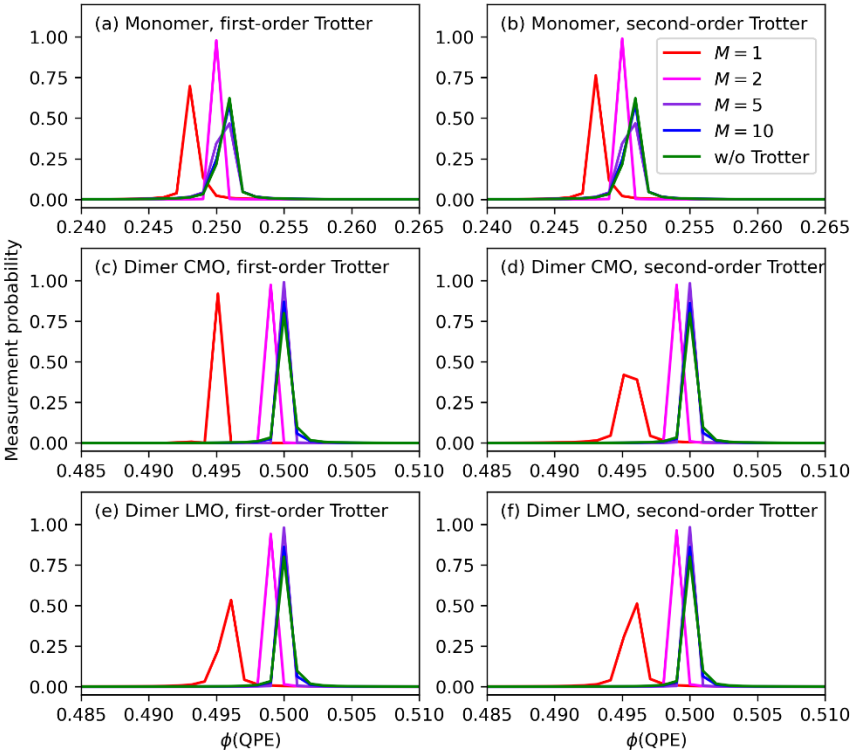
Recently we found that size consistency is violated by Trotterization in VQE-UCCSD, when the molecular orbitals delocalized on the dimer are used.



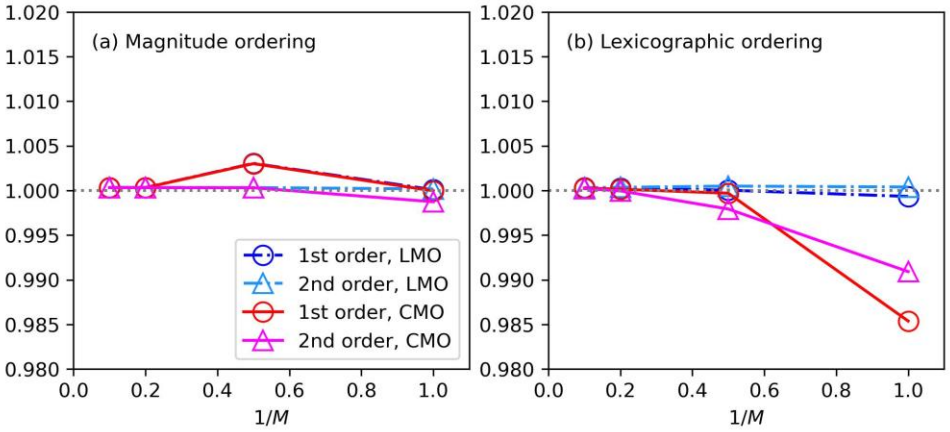
System	Energy error kcal/mol
Monomer	0.8102
Dimer (LMO)	1.6207
Dimer (CMO)	5.0319

Size consistency in QPE-based full-CI





Deviation from size consistency in triple bond dissociation of acetylene



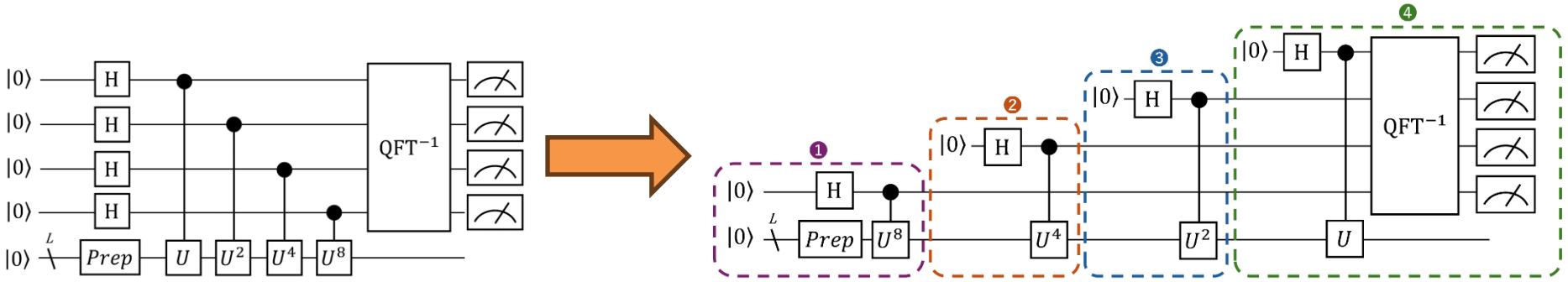
Localized MO is more suitable for satisfying size consistency.

Operator ordering in the Trotterized time evolution operator is also very important.

Acceleration of QPE quantum circuit simulations

Numerical simulations of QPE quantum circuits are very time-consuming!

By adopting the strategy of sequential addition of ancillary qubits and by performing quantum circuit simulations on GPUs, we can simulate larger-scale QPE efficiently!



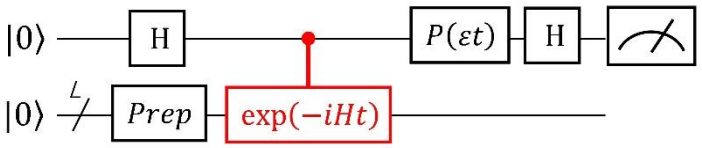
Simulation time of 26-qubit QPE (18 qubits for the system + 8 ancillary qubits)

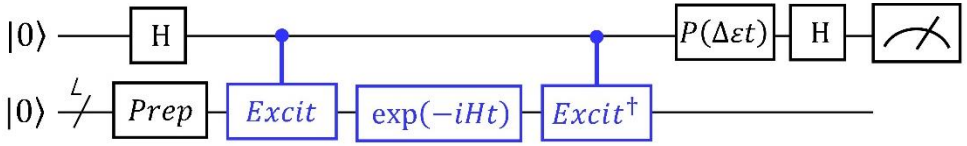
QPE implementation	GPU accelerations	Simulation time
Naïve implementation	No	13.2 days
Sequential addition of ancillary qubits	No	9.6 hours
Sequential addition of ancillary qubits	Yes	11.9 minutes

× 33

× 48.5

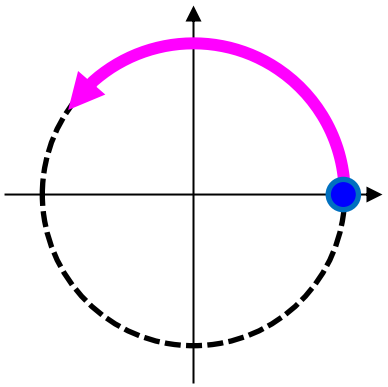
Quantum phase difference estimation (QPDE) algorithm





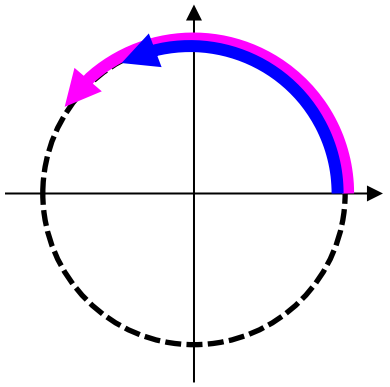
【QPE】

Determine the phase difference between $|0\rangle|\Psi\rangle$ and $\exp(-iEt)\,|1\rangle|\Psi\rangle$, to read out energy E



【QPDE】

Determine the phase difference between $\exp(-iE_0t)\,|0\rangle|\Psi_0\rangle$ and $\exp(-iE_1t)\,|1\rangle|\Psi_1\rangle$, to read out energy difference $\Delta E = E_1 - E_0$



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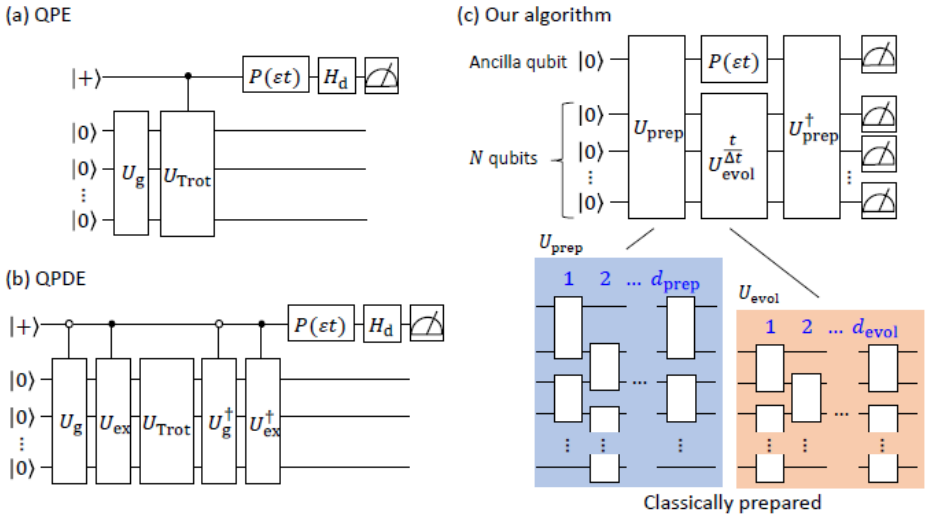
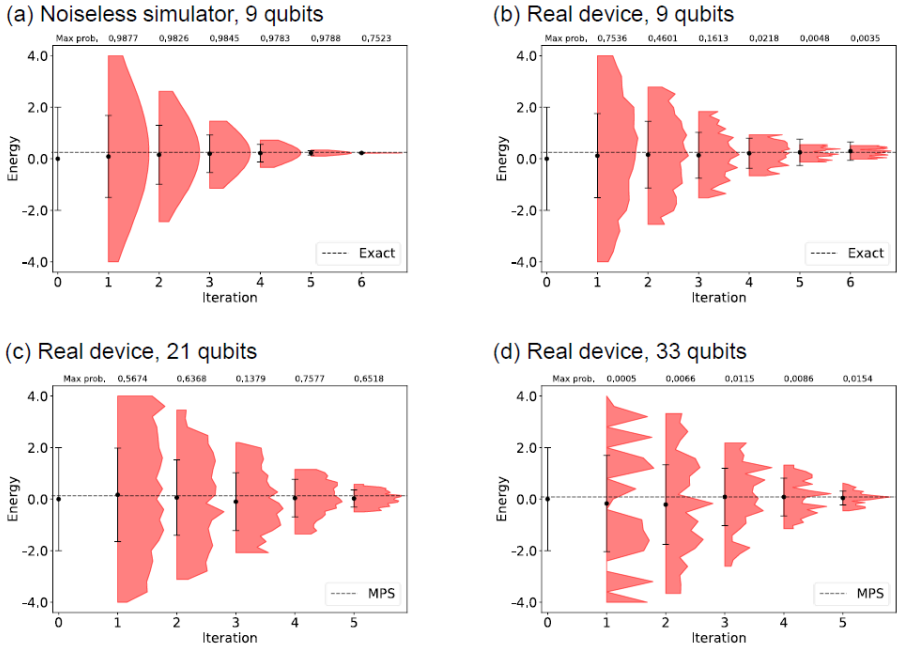
Tensor-based QPDE on a superconducting quantum hardware

It has been regarded that large-scale (more than 10 system qubits) QPE demonstrations on a real device are impossible in the current era, because the quantum circuit is too deep.

We have reported **33-qubit QPDE on a superconducting quantum hardware**.
Key techniques are

- MPS and MPO-based circuit compression on a classical computer
- Control-free time evolution in the QPDE framework
- State-of-the-art error suppression techniques in Q-CTRL





Summary

The extension of QPE-based methods is important for practical quantum computation in chemistry.

Although I skipped it due to time constraints, state preparation is one of the most important and still open problems for QPE-based quantum chemical calculations.

As we have shown in size consistency in QPE-based full-CI, there are problems that manifest themselves in the calculation of larger systems. In this context, acceleration of quantum circuit simulations is very important.

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