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Digital quantum simulation of molecular dynamics and control

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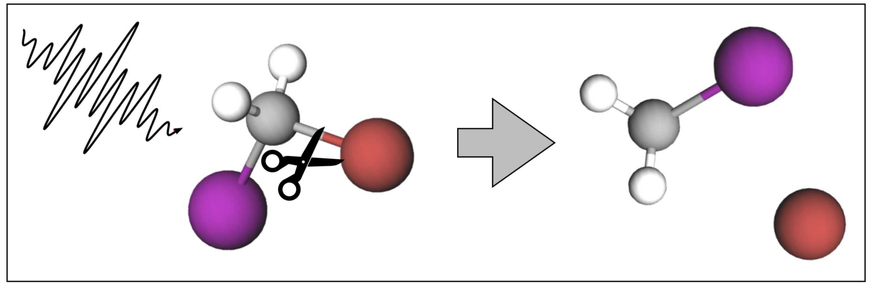
Quantum optimal control



Aim is to design a time-dependent field $f(t, \{\theta_i\})$, $t \in [0,T]$, to steer a quantum system towards a desired control target.
This is posed as the search for $\min_{\{\theta_i\}} J[T, \{\theta_i\}]$.

Simulations have two components:

1. Evaluation of control objective functional $J[T, \{\theta_i\}]$
2. Update of control parameters $\{\theta_i\}$

How well is desired control target reached?

Involves simulating the dynamics of a quantum system under the influence of an applied field

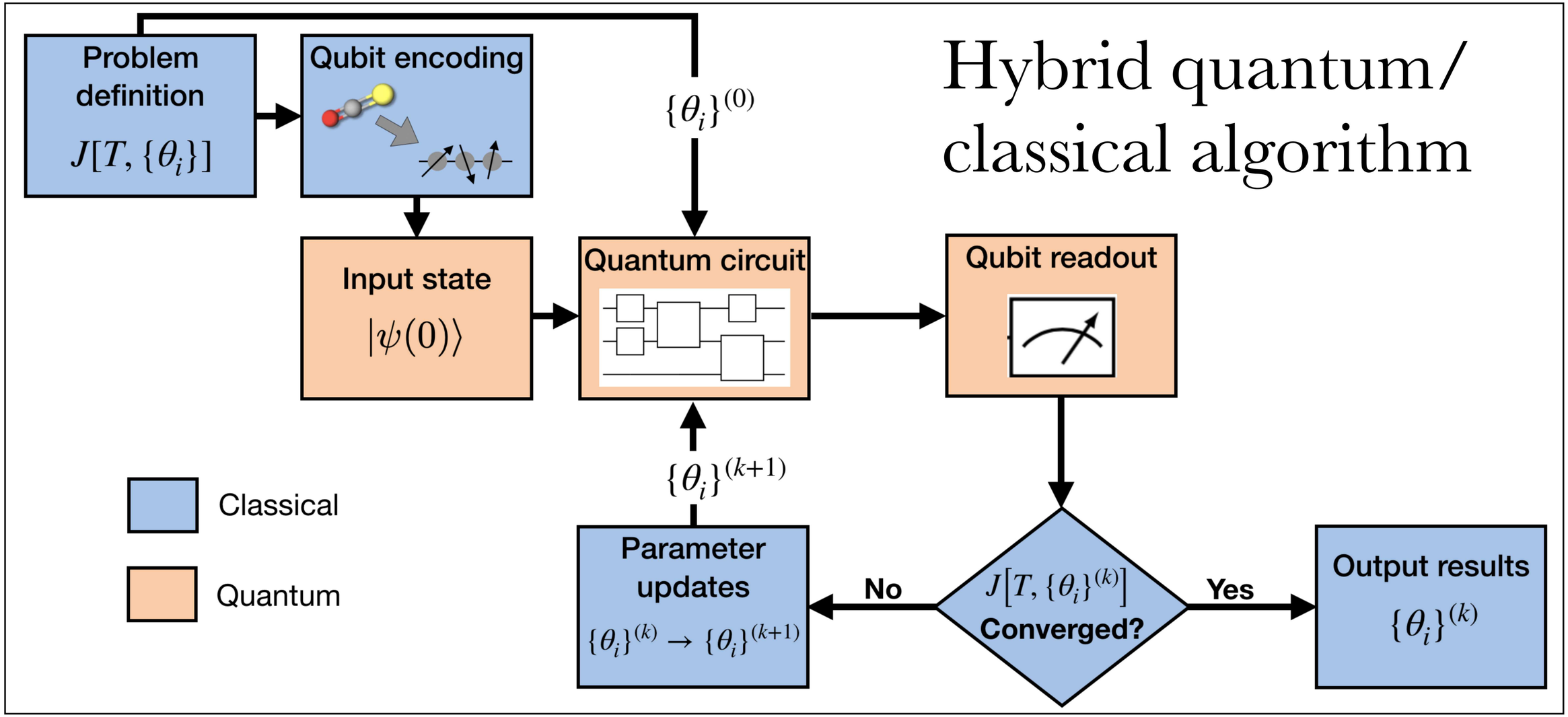
Iteratively update $\{\theta_i\}$ until $J[T, \{\theta_i\}]$ converges

- Computationally demanding
  - Computer memory/time costs scale exponentially in # quantum degrees of freedom

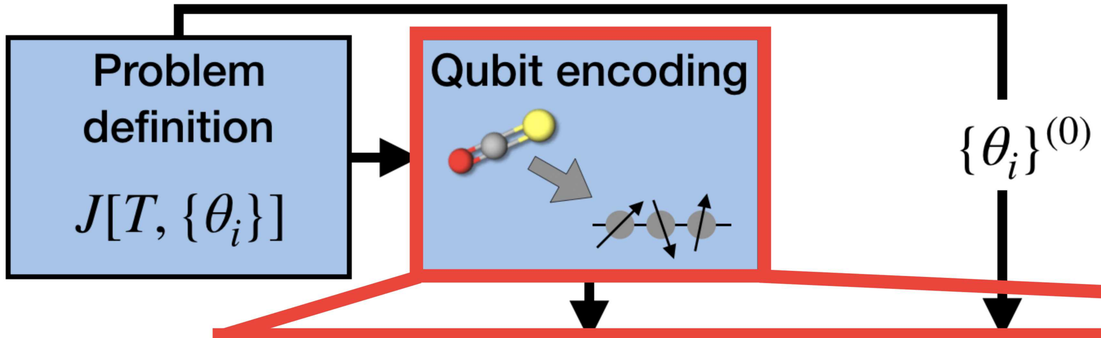
…not on a quantum computer

C. Brif, R. Chakrabarti, and H. Rabitz, New J. Phys. **12**, 075008 (2010).

Hybrid quantum/ classical algorithm



Hybrid quantum/ classical algorithm



Binary mapping to qubit states:

$$|v\rangle \mapsto |q\rangle$$
$$|0\rangle \mapsto |0\rangle|0\rangle\cdots|0\rangle|0\rangle|0\rangle$$
$$|1\rangle \mapsto |0\rangle|0\rangle\cdots|0\rangle|0\rangle|1\rangle$$
$$|2\rangle \mapsto |0\rangle|0\rangle\cdots|0\rangle|1\rangle|0\rangle$$
$$|3\rangle \mapsto |0\rangle|0\rangle\cdots|0\rangle|1\rangle|1\rangle$$

A single degree of freedom represented using d dimensions	$\mapsto \log_2(d)$ qubits
M degrees of freedom represented using d dimensions each	$\mapsto M \log_2(d)$ qubits

Mapping to qubit operators:

$$H = \sum_{v,v'} a_{v,v'} |v\rangle\langle v| \mapsto \sum_{q,q'} a_{q,q'} |q\rangle\langle q'|$$

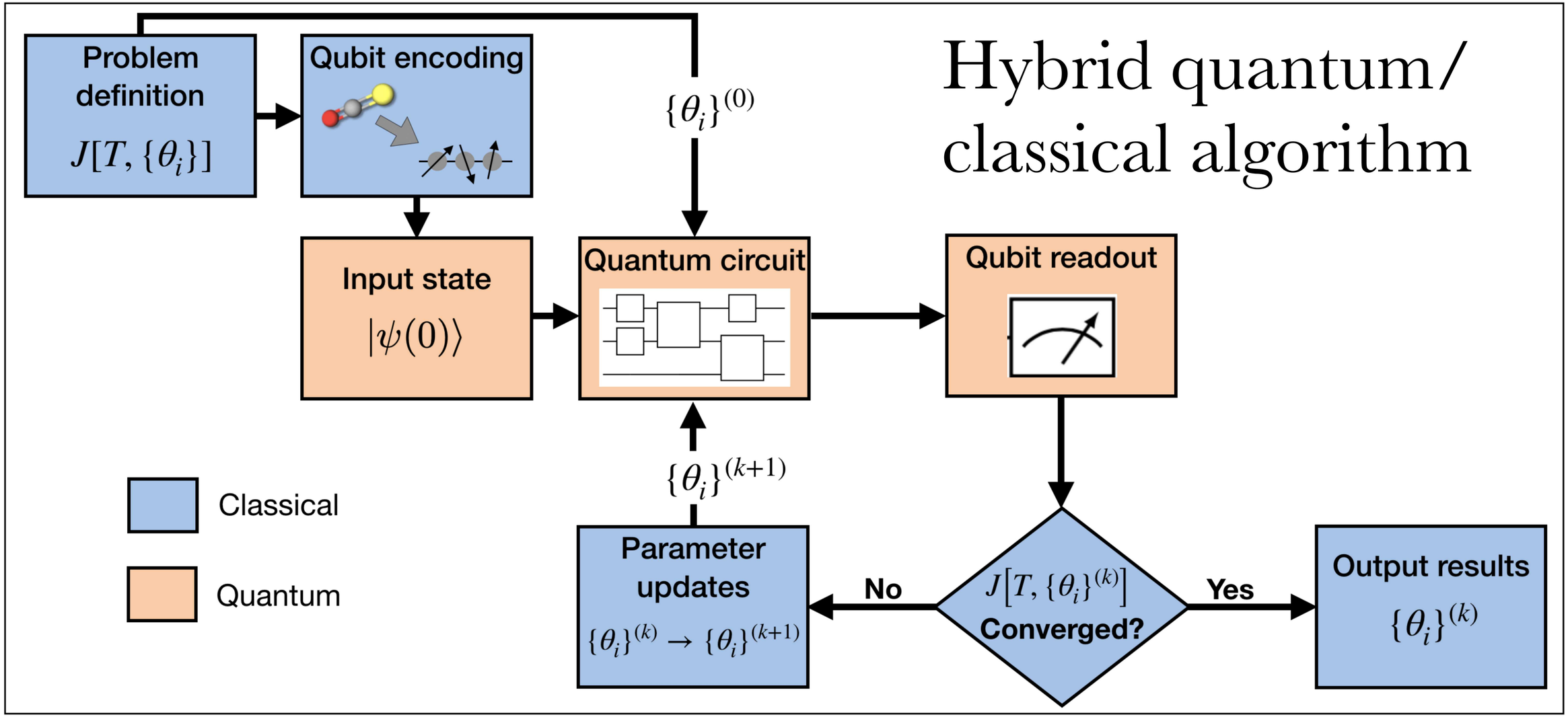
Projectors expressed as products of:

$$|1\rangle\langle 1| = \frac{I - \sigma_z}{2} \qquad |0\rangle\langle 0| = \frac{I + \sigma_z}{2}$$

$$|0\rangle\langle 1| = \frac{\sigma_x + i\sigma_y}{2} \qquad |1\rangle\langle 0| = \frac{\sigma_x - i\sigma_y}{2}$$

N. P. D. Sawaya, et al, (2019), arXiv:1909.12847 [quant-ph].

Hybrid quantum/ classical algorithm



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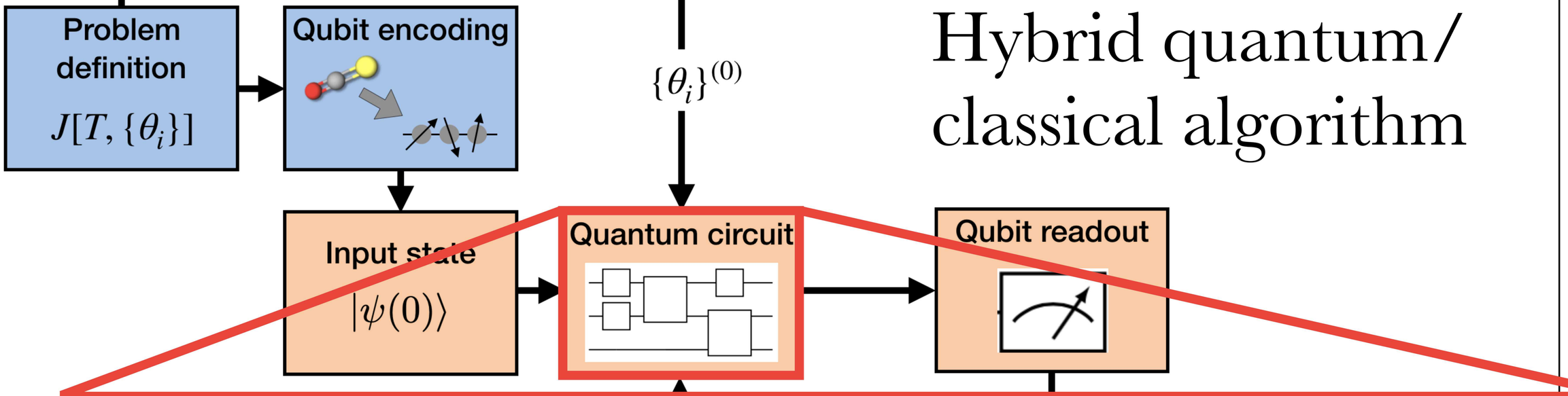
Hybrid quantum/ classical algorithm



Time evolution of initial state $|\psi(0)\rangle$ to terminal state $|\psi(T)\rangle$ simulated as

$$|\psi(T)\rangle = U(T,0)\,|\psi(0)\rangle$$

Approximate $U(T,0)$ as time-ordered product,

$$U(T,0) = U(T, T-\Delta t)\cdots U(2\Delta t, \Delta t)\, U(\Delta t,0)$$

Time evolution over each Δt then approximated using product formulas,

$$U(t+\Delta t, t) \approx U_{PF1}(t+\Delta t, t)$$
$$= \left(e^{-iH_1(t)\Delta t/n} e^{-iH_2(t)\Delta t/n} \cdots e^{-iH_L(t)\Delta t/n}\right)^n$$

Errors can be reduced with higher order product formulas,

$$U_{PF(2p)}(t+\Delta t, t) = \left(S_{2p}(t, \tfrac{-i\Delta t}{n})\right)^n$$

defined recursively as

$$S_{2p}(t,\lambda) = \left(S_{2p-2}(t,\gamma_p\lambda)\right)^2 S_{2p-2}\left(t,(1-4\gamma_p)\lambda\right)\left(S_{2p-2}(\gamma_p\lambda)\right)^2$$

$$S_2(t,\lambda) = \left(\prod_{l=1}^{L} e^{H_l(t)\lambda/2}\right)\left(\prod_{l=L}^{1} e^{H_l(t)\lambda/2}\right) \qquad \gamma_p = \frac{1}{4-4^{1/(2p-1)}}$$

M. Suzuki, J. Math. Phys **32**, 400 (1991).

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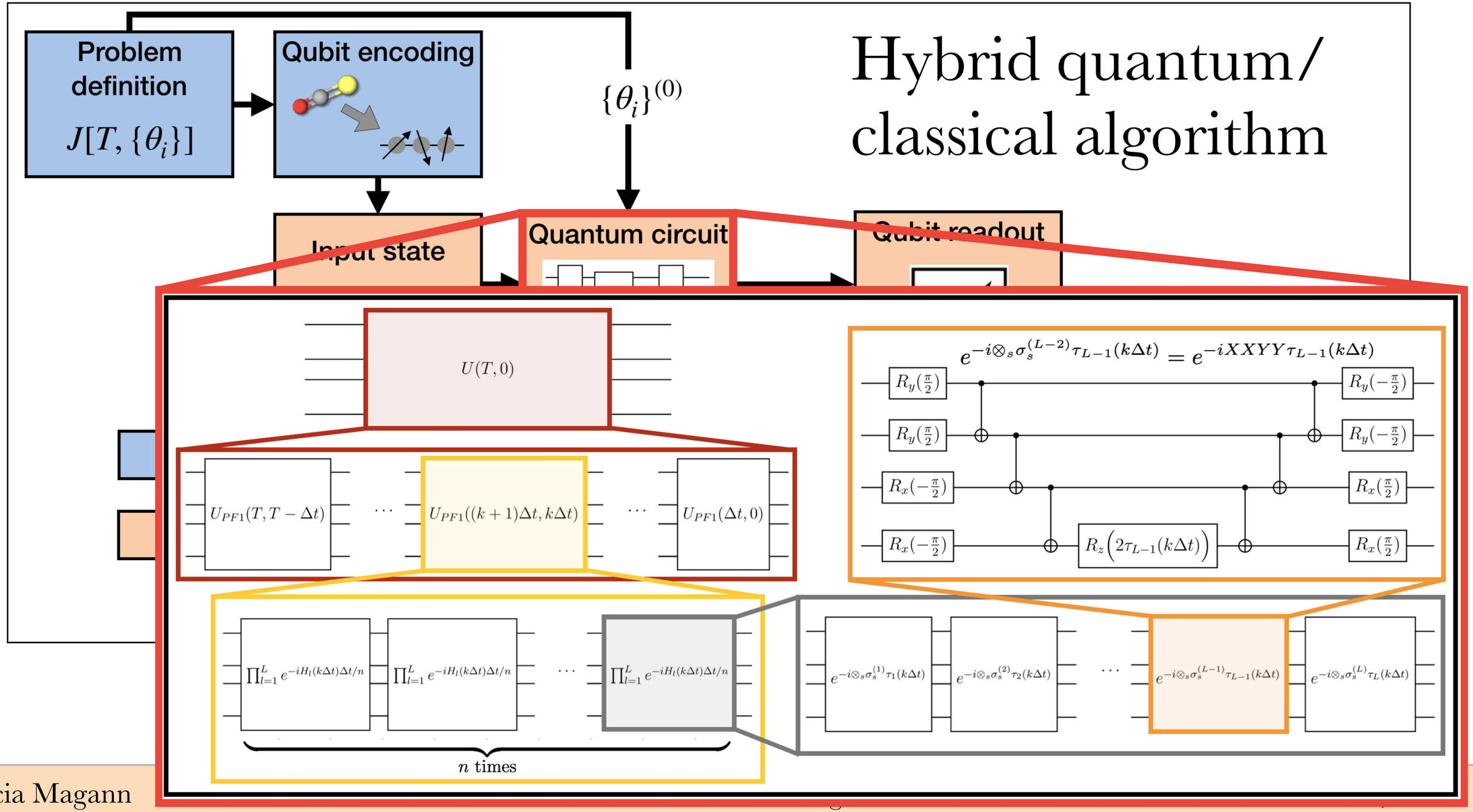
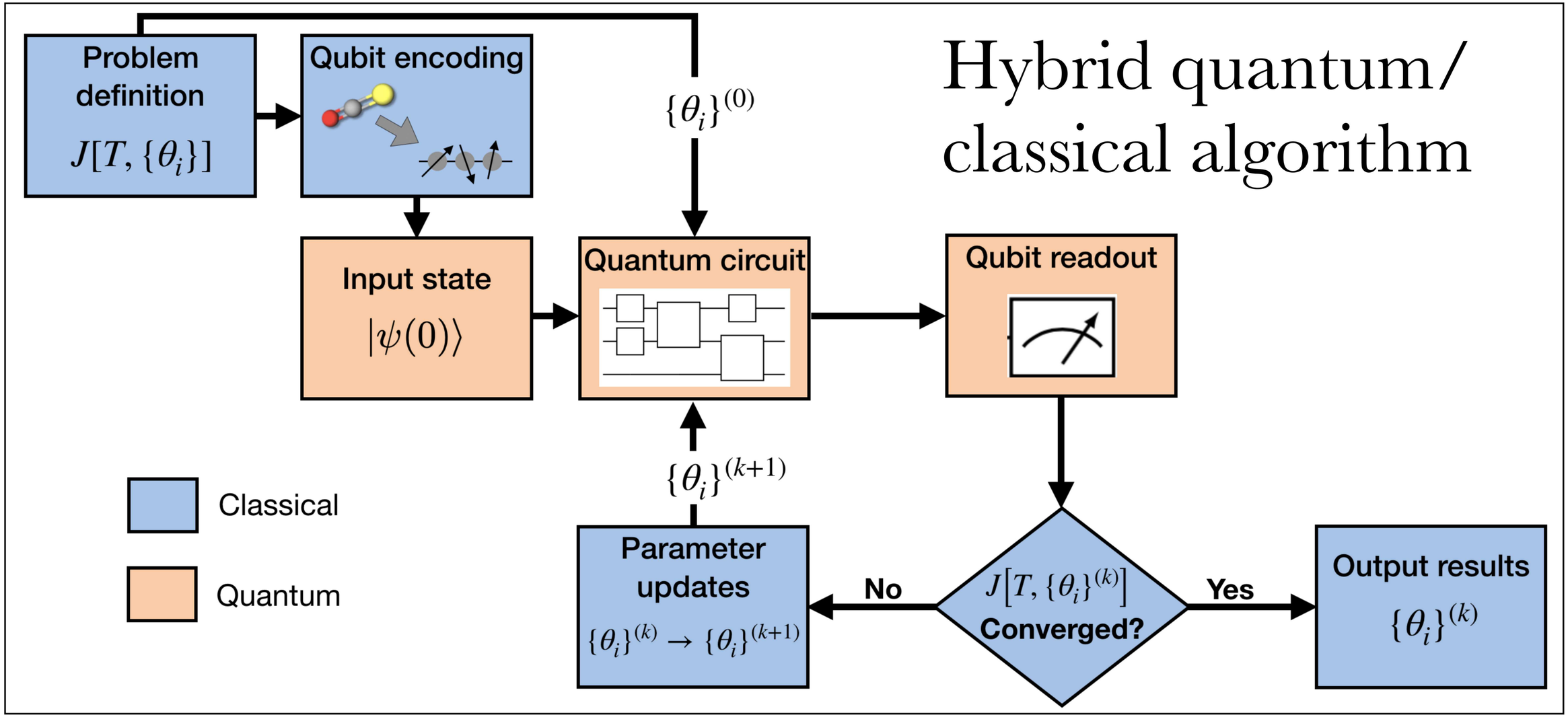
Hybrid quantum/ classical algorithm



Hybrid quantum/ classical algorithm



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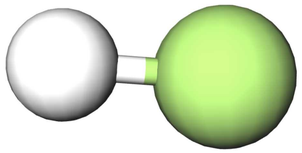
Numerical illustrations



Controlled bond stretching in HF

Goal: drive HF to target stretch $\gamma = 1.5r_0$ at terminal time T

$$J[T, \{\theta_i\}] = \left(\langle\psi(T)\,|\,r\,|\,\psi(T)\rangle - \gamma\right)^2$$

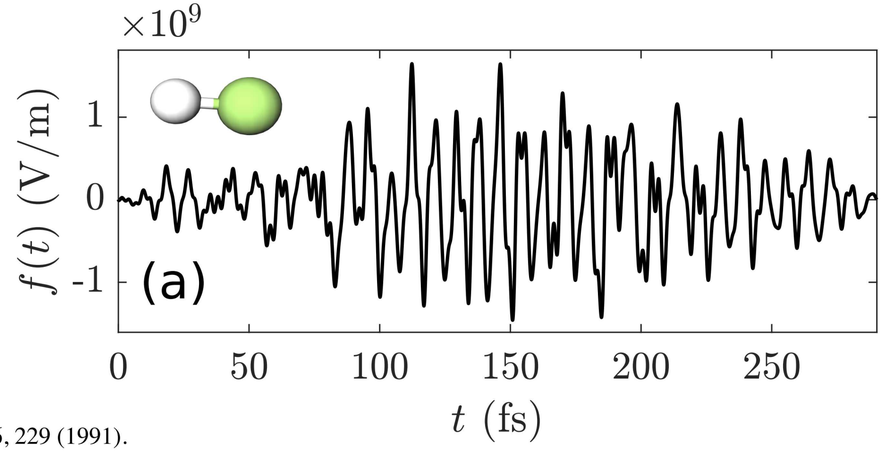
The field-induced vibrational dynamics of HF are simulated by modeling it as an anharmonic Morse oscillator

$$H(t) = \frac{p^2}{2m} + V(r) - \mu(r) f\left(t, \{\theta_i\}\right)$$

$$V(r) = D(1 - e^{-\alpha(r-r_0)})^2 - D$$

$$\mu(r) = \mu_e r e^{-\beta r^4}$$

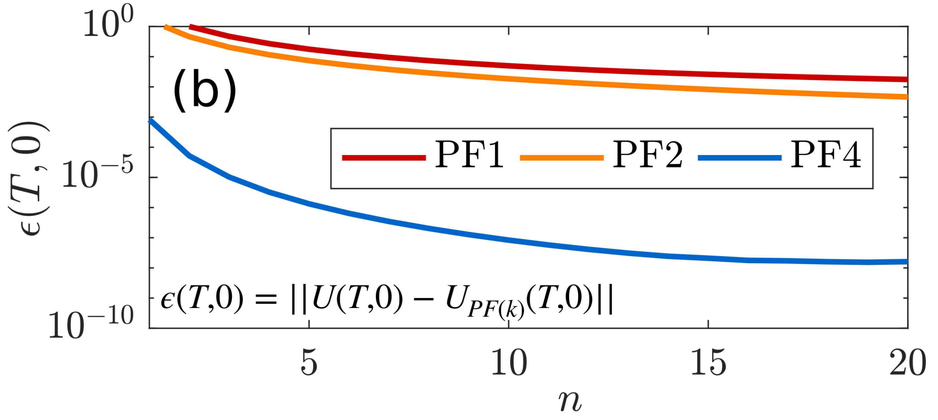
A. Guldberg and G. D. Billing, Chem. Phys. Lett **186**, 229 (1991).



Parameters $\{\theta_i\}$ taken to be amplitudes $\{a_i\}$, phases $\{\phi_i\}$, and detunings $\{\Delta_i\}$ of a mix of frequency components, and are updated using a genetic algorithm

$$f\left(t, p, a_1, \cdots, a_k, \Delta_1, \cdots, \Delta_k, \phi_1, \cdots, \phi_k\right) = \sin^{1/p}\left(\frac{\pi t}{T}\right)\left(\sum_{i=1}^{k} a_i \cos\left((\omega_i + \Delta_i) - \phi_i\right)\right)$$

The system is represented in the harmonic oscillator eigenbasis and encoded using $N = 4$ qubits with initialization $|\psi(0)\rangle = |0\rangle|0\rangle|0\rangle|0\rangle$



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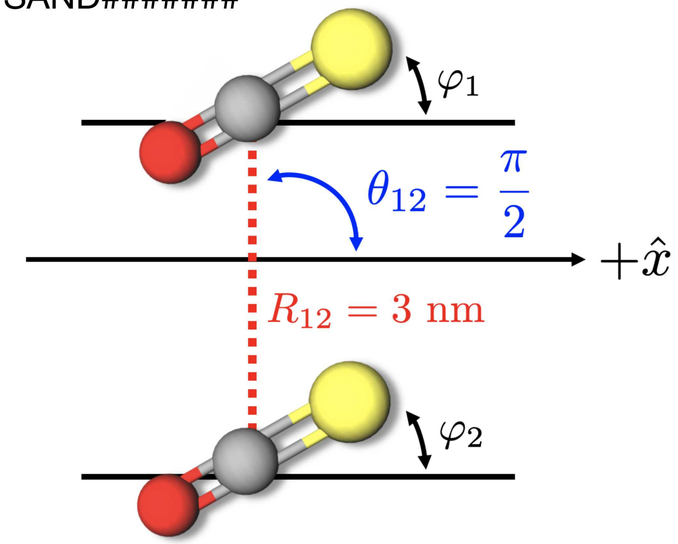
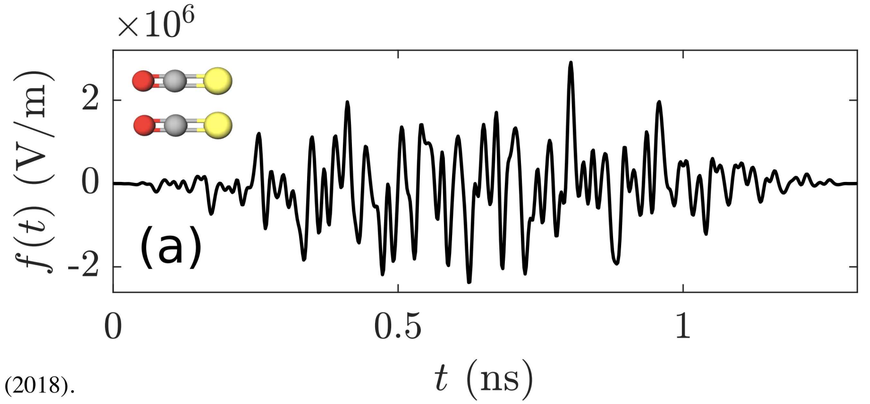


Controlled orientation of 2 dipole-dipole coupled OCS rotors



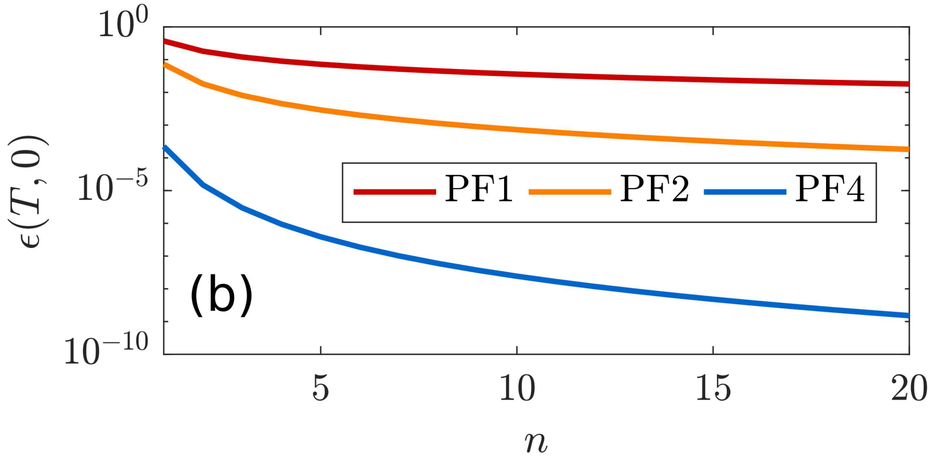
Goal: control the rotational dynamics of two planar dipole-dipole coupled OCS rotors

$$H(t) = \sum_{j=1}^{2} -\frac{\hbar^2}{2I}\frac{\partial^2}{\partial\varphi_j^2} + V_{12} - \mu(\cos\varphi_1 + \cos\varphi_2) f\big(t, \{\theta_i\}\big)$$

H. Yu, T.-S. Ho, and H. Rabitz, Phys. Chem. Chem. Phys. **20**, 13008 (2018).

such that they are oriented in the $+\hat{x}$ direction at time T

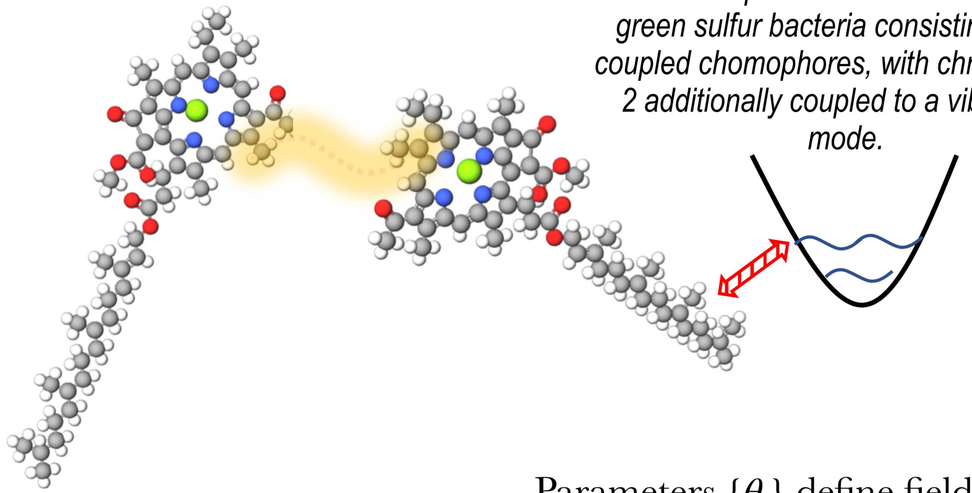
$$J[T, \{\theta_i\}] = -\langle\psi(T)|(\cos\varphi_1 + \cos\varphi_2)|\psi(T)\rangle$$



System modeled in free rotor eigenbasis and encoded using $N = 6$ qubits with initialization $|\psi(0)\rangle = |0\rangle|0\rangle\cdots|0\rangle$

Parameters $\{\theta_i\}$ taken to be amplitudes, phases, and detuning of a mix of frequency components; Updated using a genetic algorithm

Controlled excitonic state preparation in light harvesting complex



Model for portion of FMO complex of green sulfur bacteria consisting of two coupled chomophores, with chromophore 2 additionally coupled to a vibrational mode.

Parameters $\{\theta_i\}$ define field profile and updated with genetic algorithm

Goal: prepare an excitation on the second chromophore when the vibrational mode is initially in a thermal state $\sum_{v=0}^{v_{max}} c_v |v\rangle\langle v|$ at 300K, where $c_v = \frac{1}{tr\{e^{-\beta H_v}\}} e^{-\beta v}$.

$N = 5$ qubits are used for the encoding, and the control objective functional is given by

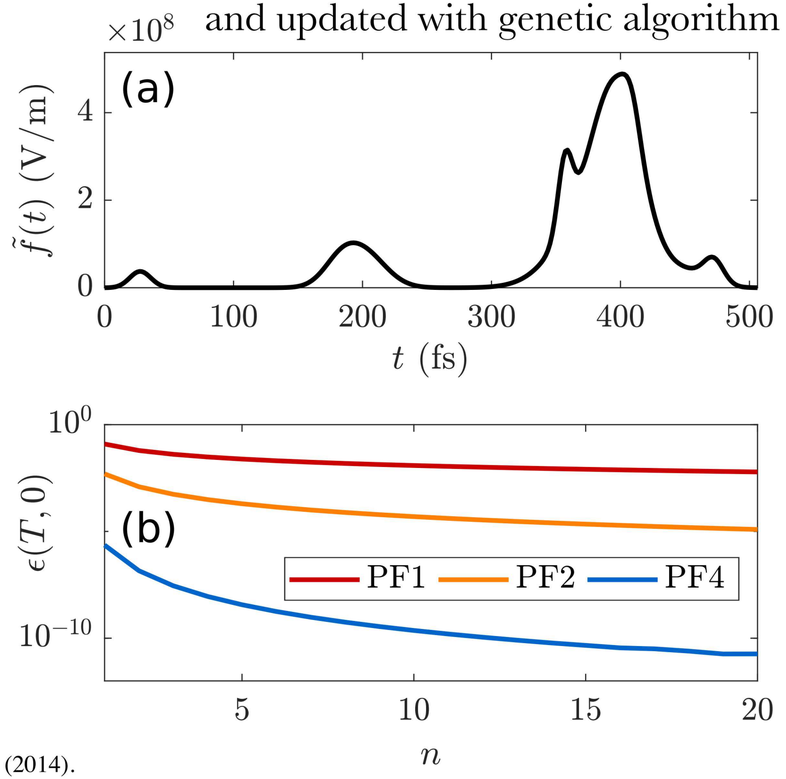
$$J[T, \{\theta_i\}] = \sum_{v=0}^{v_{max}} c_v J_v[T, \{\theta_i\}]$$

where $J_v[T, \{\theta_i\}] = 1 - \langle\psi(T)| \left(|g_1 e_2\rangle\langle g_1 e_2|\right) I_v |\psi(T)\rangle, \qquad |\psi(0)\rangle = |0\rangle|0\rangle|v\rangle$

and the Hamiltonian is given by,

$$H(t) = \sum_{j=1}^{2} E_j b_j^\dagger b_j + J(b_1^\dagger b_2 + b_2^\dagger b_1) + \nu a^\dagger a + g(b_2^\dagger b_2)(a + a^\dagger) + \sum_{j=1}^{2} (\mu_j (b_j + b_j^\dagger)) f(t, \{\theta_i\})$$

Model: S. Hoyer, et al, New J. Phys **16**, 045007 (2014).



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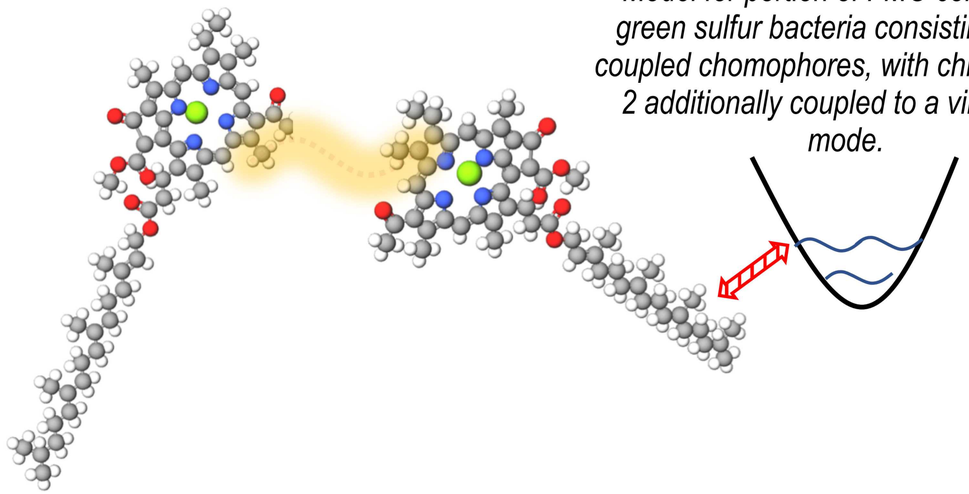
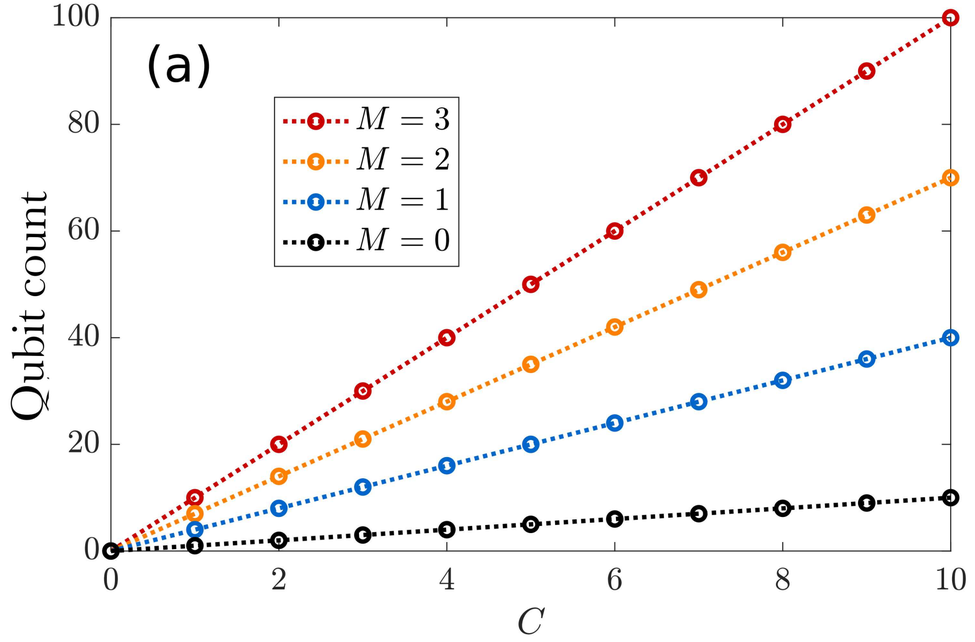
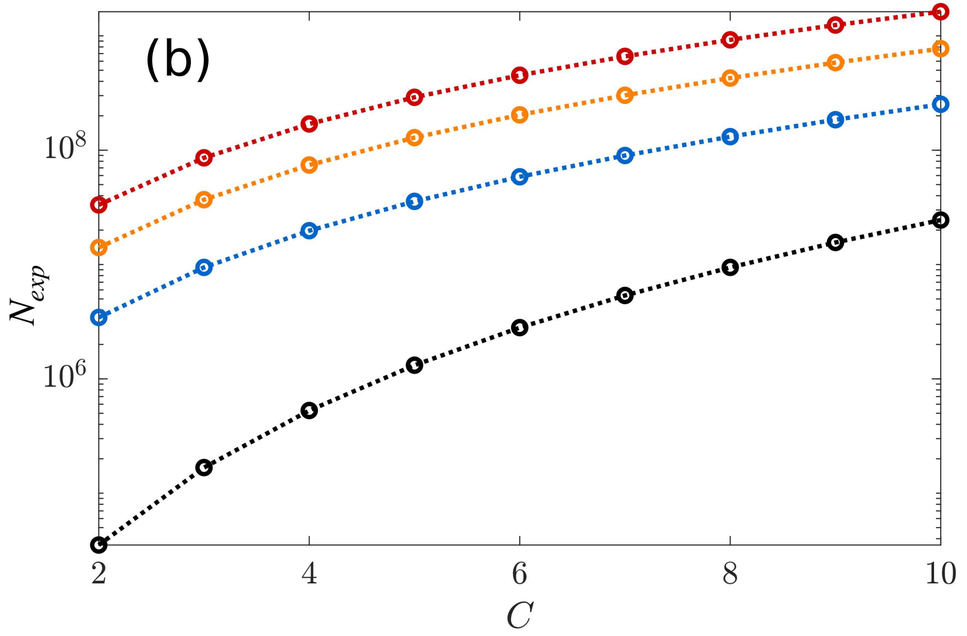


Model for portion of FMO complex of green sulfur bacteria consisting of two coupled chomophores, with chromophore 2 additionally coupled to a vibrational mode.

Resource analysis for simulating the dynamics of C coupled chromophores, each coupled to M vibrational modes, on a quantum computer





D. W. Berry, G. Ahokas, R. Cleve, and B. C. Sanders, Commun. Math. Phys **270**, 359 (2007).

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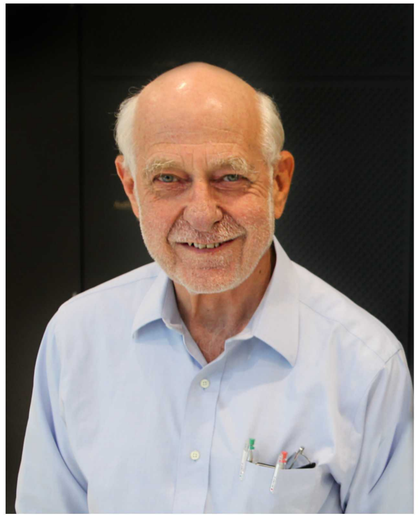
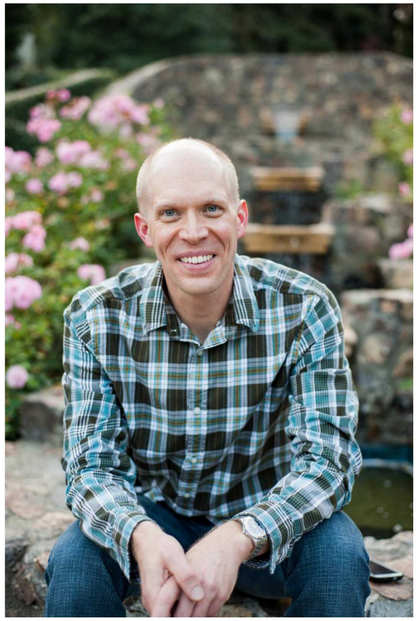
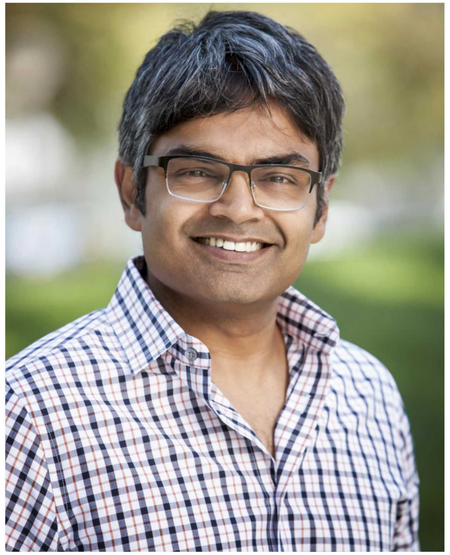
Thank you



Herschel Rabitz



Matthew Grace



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