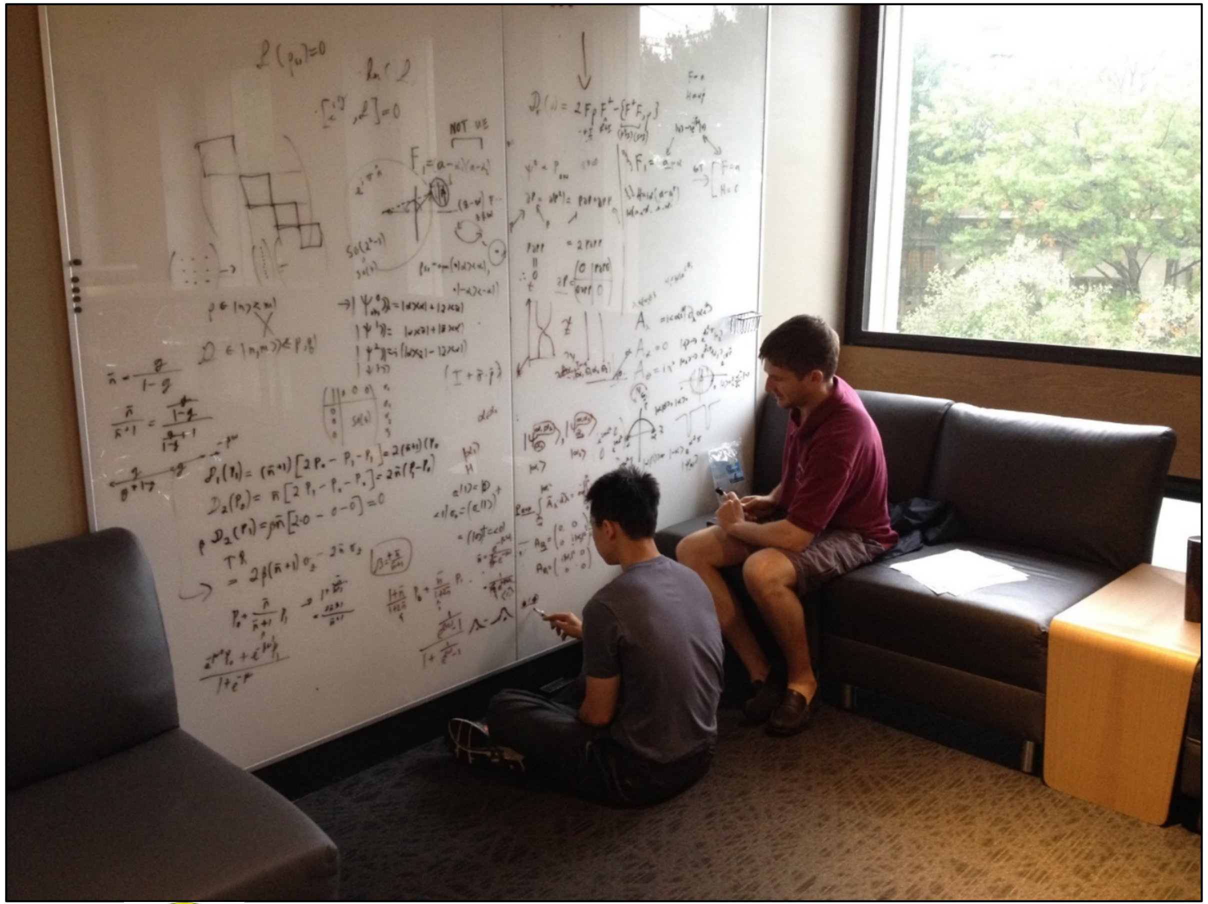
Experimental Simulation of Atomic Vibrational Spectra Using Efficient Boson Sampling

Steven Girvin
Yale Quantum Institute

<u>Experiment</u>

Michel Devoret
Luigi Frunzio
Rob Schoelkopf

Chris Wang
Jacob Curtis
Luyan Sun
Yvonne Gao
Brian Lester
Andrei Petrenko
Nissim Ofek
Reinier Heeres
Philip Reinhold
Yehan Liu
Zaki Leghtas
Brian Vlastakis
+…..



<u>Theory</u>

SMG
Liang Jiang
Leonid Glazman
M. Mirrahimi
Shruti Puri

Baptiste Royer
Shraddha Singh
Kevin Smith
Micheline Soley
Yaxing Zhang
+…..

Disclosure: SMG is a consultant and equity holder in Quantum Circuits, Inc. and an equity holder in IBM, Inc.

Research supported by ARO

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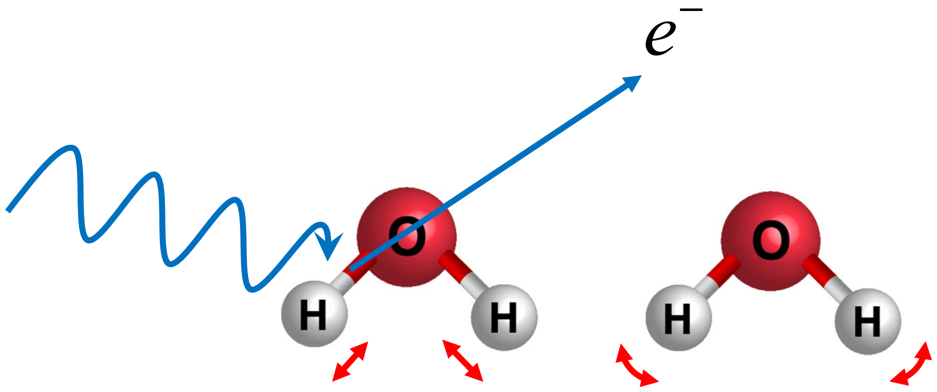
Goal: Study quantum non-equilibrium vibrational dynamics of small molecules following a sudden photoemission event



Experimental demonstration of a highly hardware efficient quantum 'boson sampling' simulator using microwave quanta to represent vibrational quanta

First-generation experiments: Chris Wang (Schoelkopf lab)

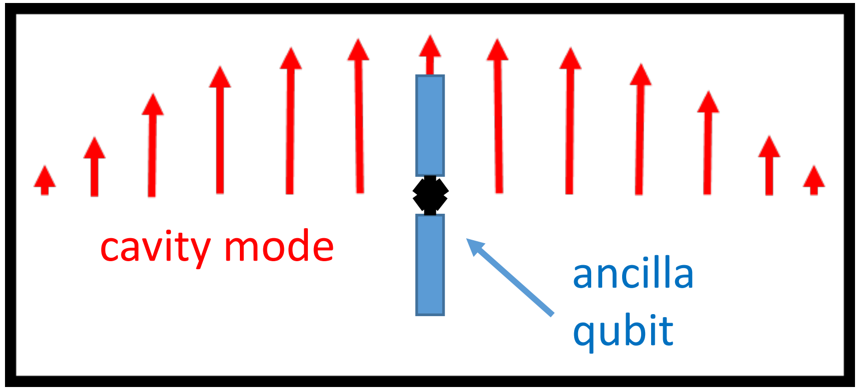
Phys. Rev. X 10, 021060 (2020)
Phys. Rev. X 13, 011008 (2023)

Programmable quantum simulator desiderata:

1. Universal control
    - Create initial non-classical states
    - Synthesize arbitrary Hamiltonian dynamics

2. Efficient and non-trivial measurements
    - Probe the simulation results
    - State tomography beyond capabilities typically available in the system being simulated

Universal control and measurement of hybrid qubit-oscillator systems in circuit QED.



Qubit-Cavity Strong Dispersive Coupling

$$\omega_c \neq \omega_q$$

$$H = \omega_c a^\dagger a + \frac{\omega_q}{2}\sigma^z + \chi\,\sigma^z\, a^\dagger a$$

Permits several possible instruction sets

Example 1: SNAP gate set:

(Recall this from Lecture 2)

Cavity-controlled qubit rotations + Cavity displacements:

$$U_{\mathrm{SNAP}}(\vec{\theta}) \equiv e^{i\sigma^z \sum_{n=0}^{n_{\max}} \theta_n \hat{P}_n}$$

$$\hat{P}_n = |n\rangle\langle n|$$

$$\vec{\theta} = (\theta_0, \theta_1, \ldots, \theta_{n_{\max}})$$

$$\mathcal{D}(\alpha) = e^{\alpha a^\dagger - \alpha^* a}$$

ISA Example 2: Phase-Space Displacements
Qubit-Controlled Cavity Displacement + Qubit Rotations

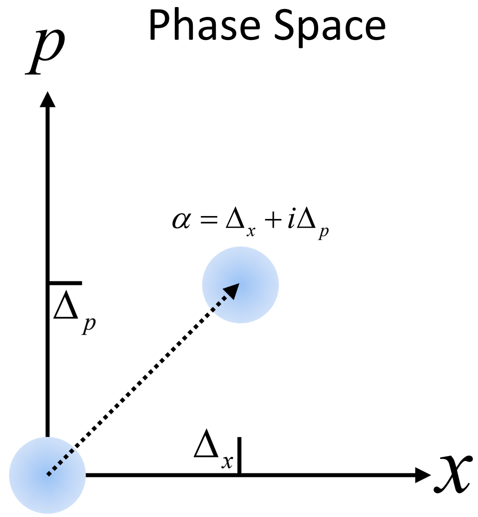
Phase Space Displacement by $\alpha = \Delta_x + i\Delta_p$

$$\mathcal{D}(\alpha) = e^{\alpha a^\dagger - \alpha^* a} = e^{2i[-\Delta_x \hat{p} + \Delta_p \hat{x}]}$$

$-2\hat{p}$ generates displacements
$+2\hat{x}$ generates boosts

'Wigner units': $\hat{x} = \frac{a + a^\dagger}{2}, \hat{p} = \frac{a - a^\dagger}{2i}$; $[\hat{x}, \hat{p}] = \frac{i}{2}$

$$a = \hat{x} + i\hat{p}$$



Starting from vacuum, this is a non-universal gate that can only produce coherent states:

$$|\alpha\rangle \equiv \mathcal{D}(\alpha)|0\rangle = \sum_{n=0}^{\infty} \frac{\alpha^n}{\sqrt{n!}}|n\rangle; \quad a|\alpha\rangle = \alpha|\alpha\rangle \quad \text{['Hilbert Hotel']}$$

ISA Example 2: Qubit-Controlled Cavity Displacement + Qubit Rotations

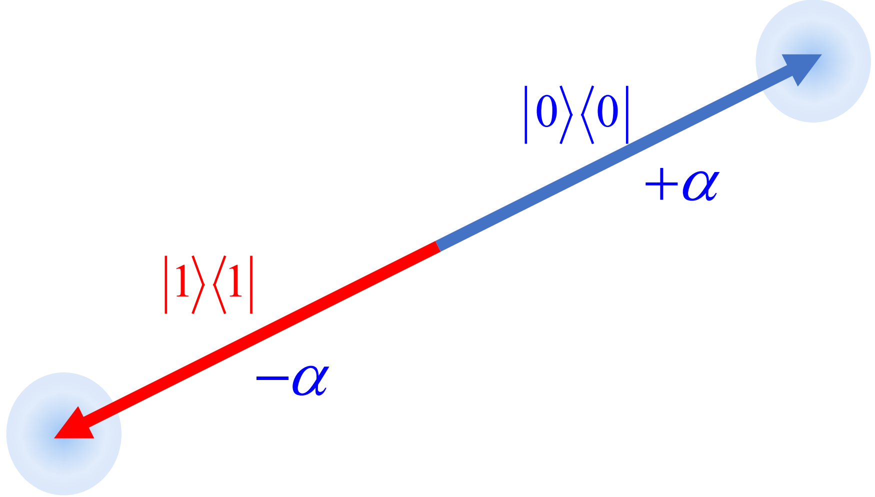
Fast Universal Control of an Oscillator with Weak Dispersive Coupling to a Qubit, A. Eickbusch et al. (Devoret Lab)
Nature Physics **18**, 1464 (2022)

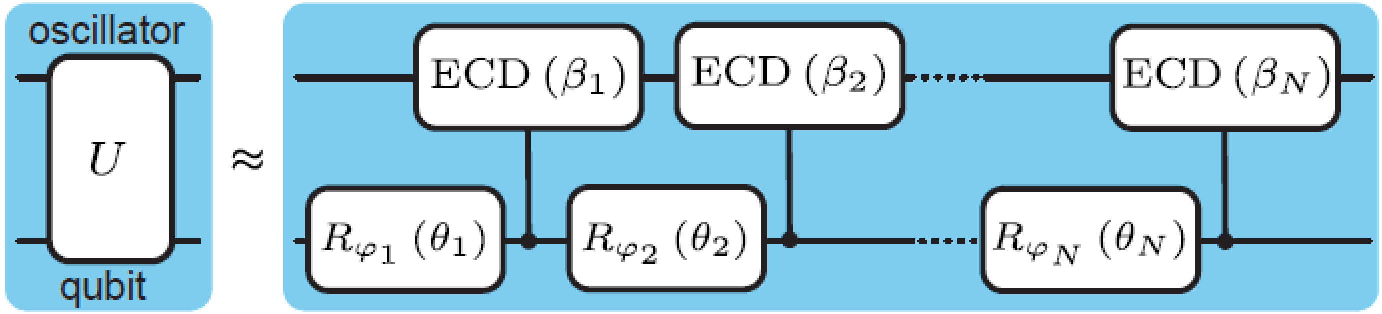
Conditional Displacement Gate

$$\mathcal{D}_c(\alpha) = |0\rangle\langle 0|\mathcal{D}(\alpha) + |1\rangle\langle 1|\mathcal{D}(-\alpha) = e^{[\alpha a^\dagger - \alpha^* a]\sigma^z}$$

Qubit Rotation Gate

$$R_\varphi(\theta) = e^{-i\frac{\theta}{2}\left[\cos\varphi\,\sigma^x + \sin\varphi\,\sigma^y\right]}$$





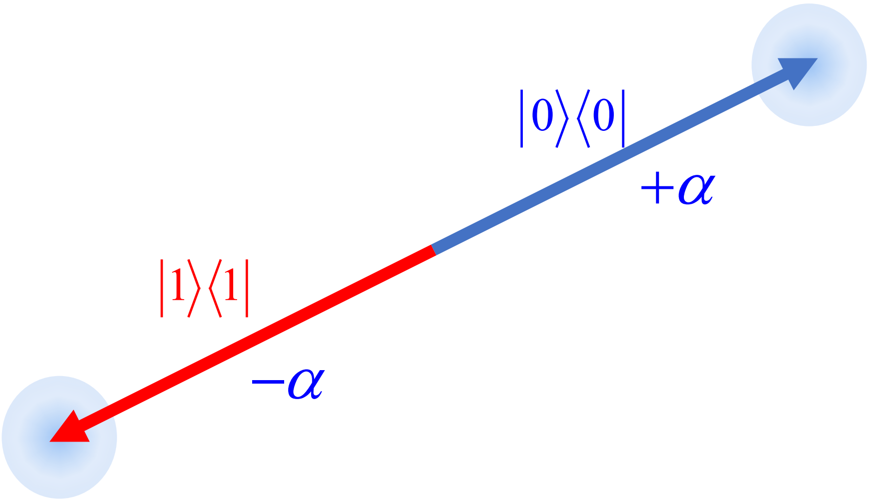
Circuit depth →

ISA Example 2: Qubit-Controlled Cavity Displacement + Qubit Rotations

Non-Commuting Geometry of Oscillator Phase Space $\otimes$ Bloch Sphere: Conditional Displacements

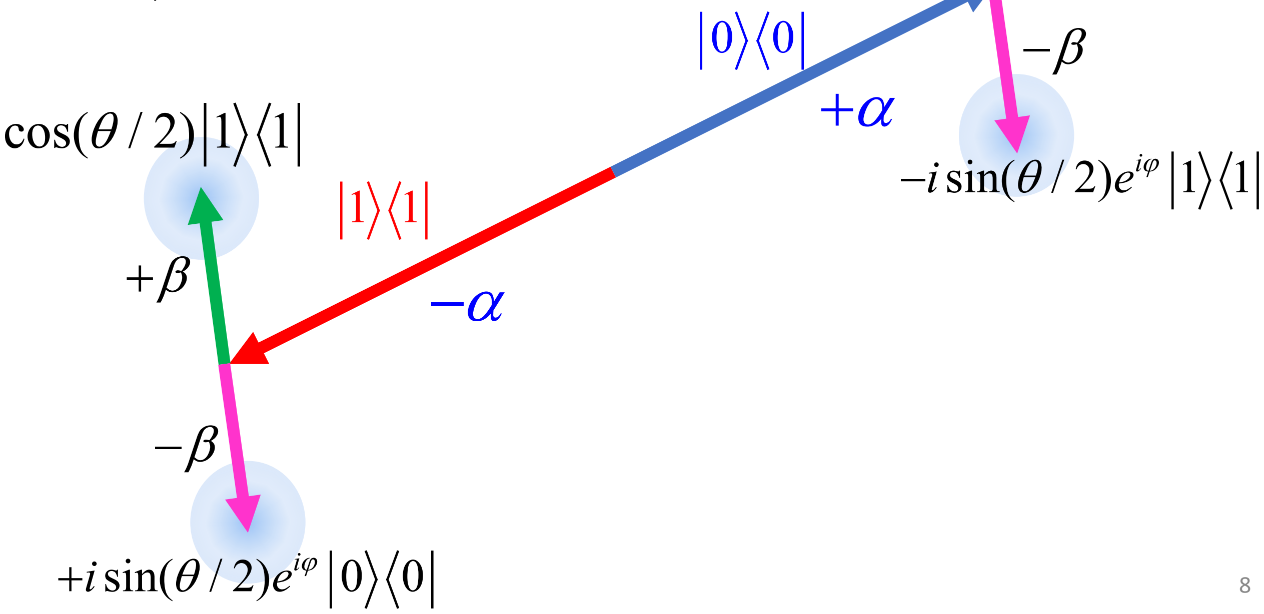
Universal Gate Set (Lie algebra does not close):
Composing conditional displacements and qubit rotations

$$\mathcal{D}_c(\alpha) = |0\rangle\langle 0|\mathcal{D}(\alpha) + |1\rangle\langle 1|\mathcal{D}(-\alpha)$$

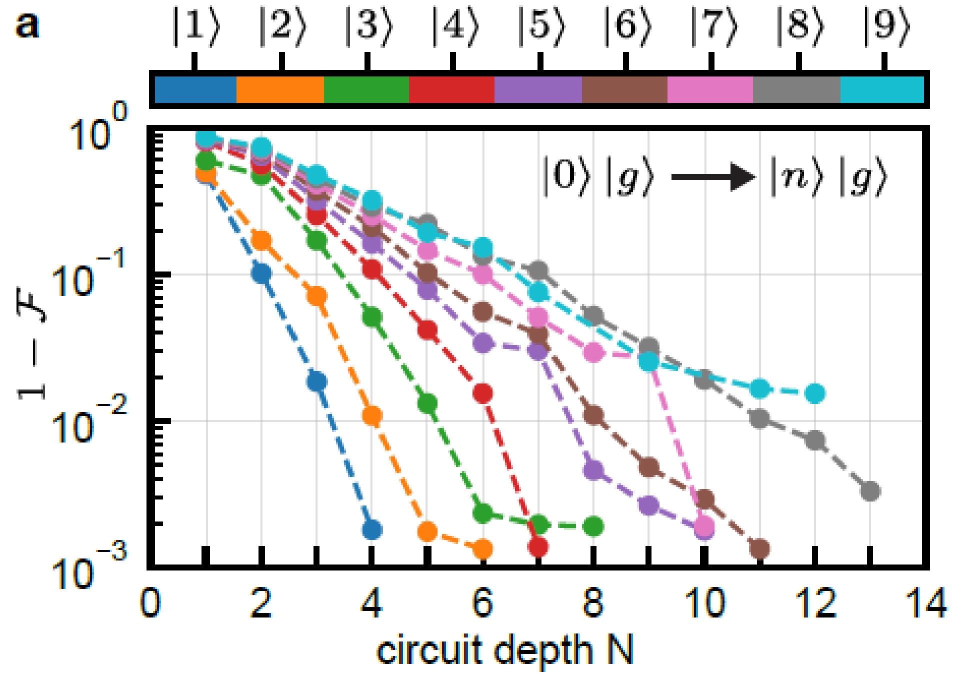


$$\mathcal{D}_c(\beta)R_\varphi(\theta)\mathcal{D}_c(\alpha)$$

$$R_\varphi(\theta) = e^{-i\frac{\theta}{2}\left[\cos\varphi\,\sigma^x + \sin\varphi\,\sigma^y\right]}$$



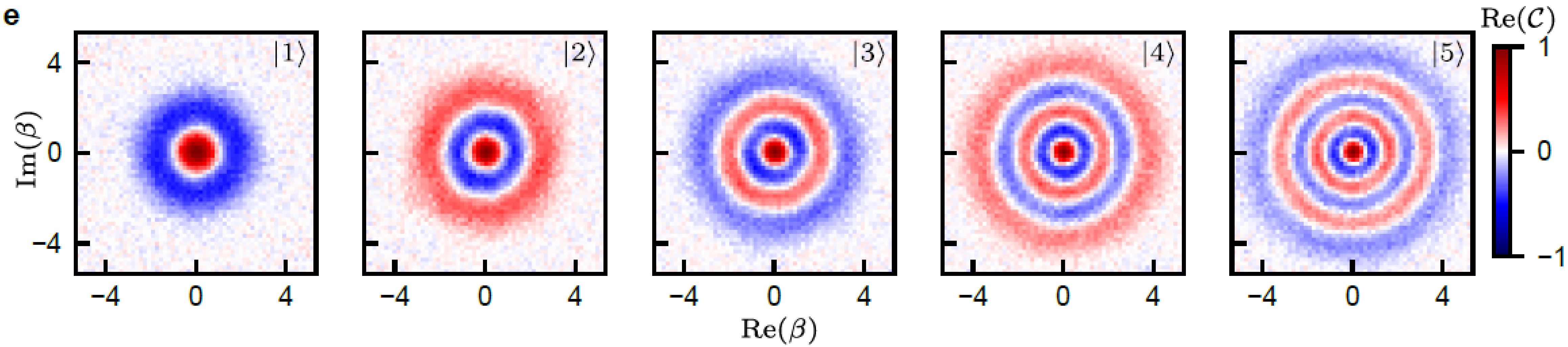
(Echoed) Controlled-Displacement ISA:



Fast Universal Control of an Oscillator with Weak Dispersive Coupling to a Qubit, A. Eickbusch et al. (Devoret Lab)
Nature Physics **18**, 1464 (2022)

[Wigner function and characteristic function phase space tomography plots will be explained later.]

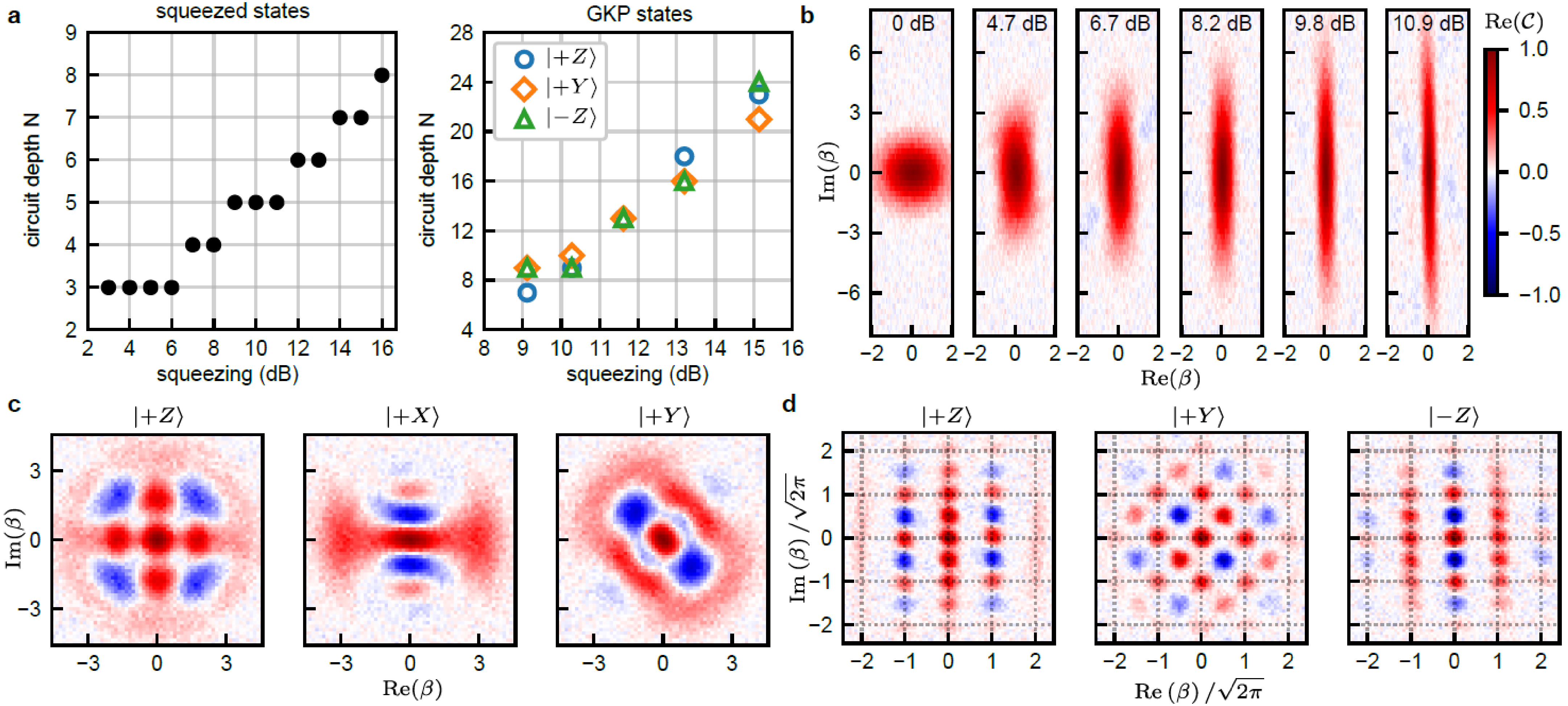
Photon Fock State Generation



(Echoed) Controlled-Displacement ISA:

Fast Universal Control of an Oscillator with Weak Dispersive Coupling to a Qubit, A. Eickbusch et al. (Devoret Lab)
Nature Physics **18**, 1464 (2022)

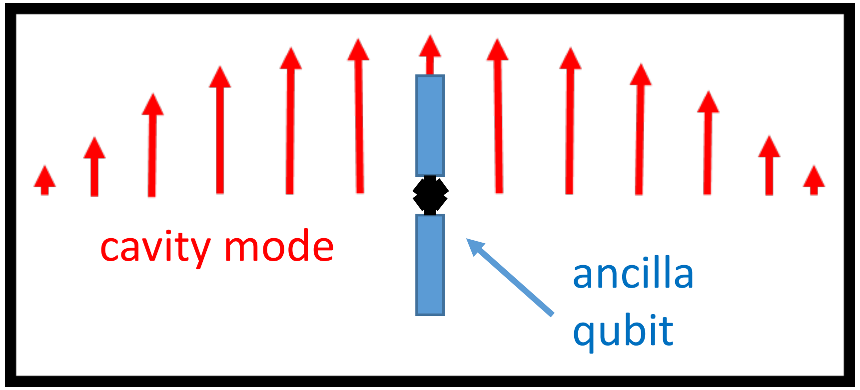
Squeezed states



Binomial QEC code word states

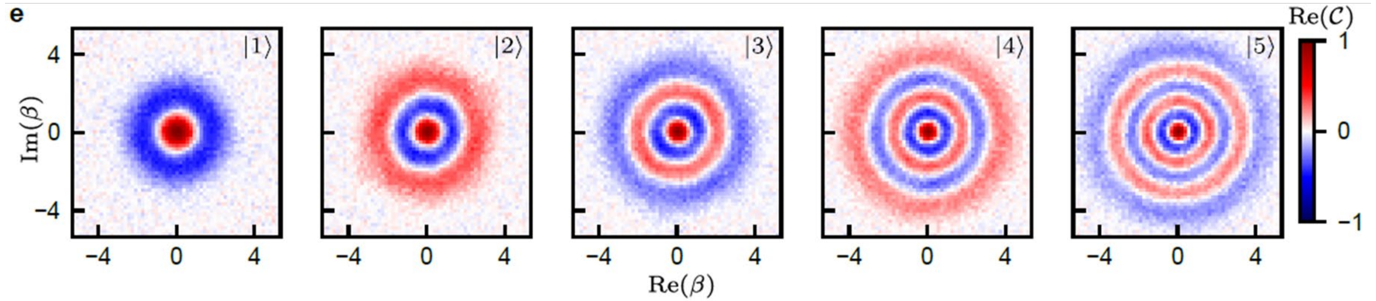
GKP QEC code word states

Universal control and measurement of hybrid qubit-oscillator systems in circuit QED.



Bosonic State Tomography with the Characteristic Function (related to density matrix: $\rho(x',x)=\langle x'|\Psi\rangle\langle\Psi|x\rangle$)
--Using the controlled displacement gate

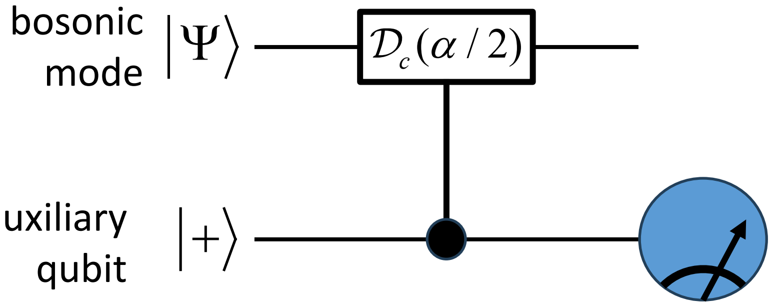
Photon Fock State Generation



Characteristic Function

$$C(\alpha)=\langle\Psi|\mathcal{D}(\alpha)|\Psi\rangle$$

How do we measure the overlap of the state with a displaced version of itself?



Measure phase kickback of controlled displacement

$$\langle\sigma^x\rangle=\mathrm{Re}\,C(\alpha)$$
$$\langle\sigma^y\rangle=-\mathrm{Im}\,C(\alpha)$$

$$\mathcal{D}_c(\alpha)=e^{[\alpha a^\dagger-\alpha^* a]\sigma^z}=e^{-2i[\Delta_x\hat{p}+i\Delta_p\hat{x}]\sigma^z}$$

Interpretation:
Think of this, not as a controlled displacement, but as a rotation of the qubit around the z axis by an angle dependent on the position and momentum of the oscillator.

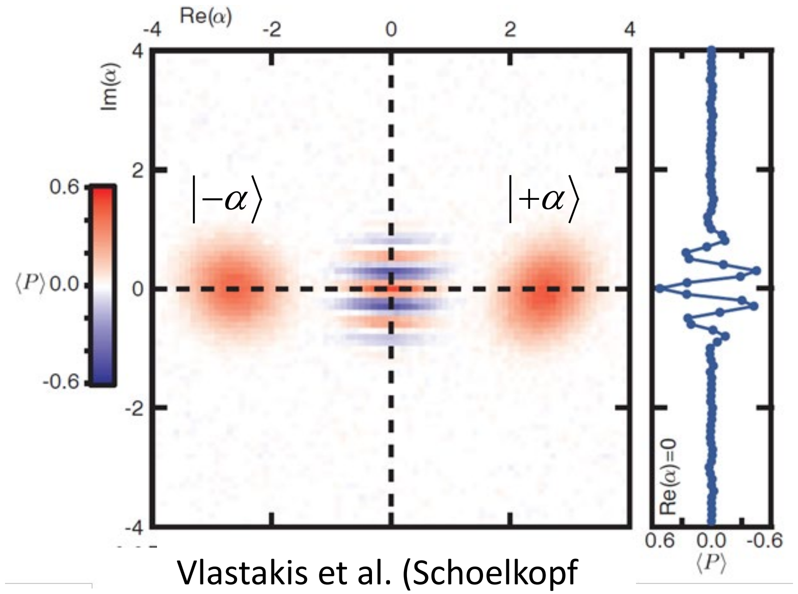
Bosonic State Tomography with the Wigner Function (also related to density matrix and characteristic function)

Theory | Experiment

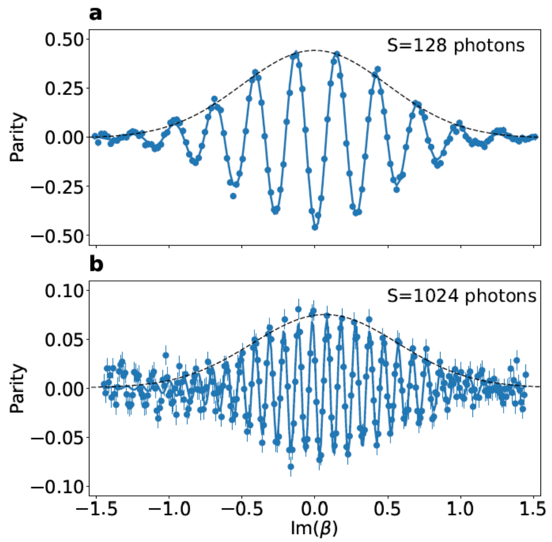
$$W(\beta) = \frac{1}{\pi^2}\int d^2\alpha\, C(\alpha) e^{\beta\alpha^* - \alpha\beta^*} = \langle \Psi | D^\dagger(\alpha)(-1)^{\hat{n}} D(\alpha) | \Psi \rangle$$

Fourier transform of Characteristic Function

Expectation value of displaced photon number parity operator



Vlastakis et al. (Schoelkopf group), Science (2013)



World's largest Schrodinger Cat:

Interference fringes in Wigner function prove coherent superposition (of coherent states)

$$\frac{1}{\sqrt{2}}\left[|+\alpha\rangle \pm |-\alpha\rangle\right]$$

Milul et al., (S. Rosenblum group) arXiv:2302.06442; See also J. Home ion-trap cats.

State Tomography in the Fock Basis:

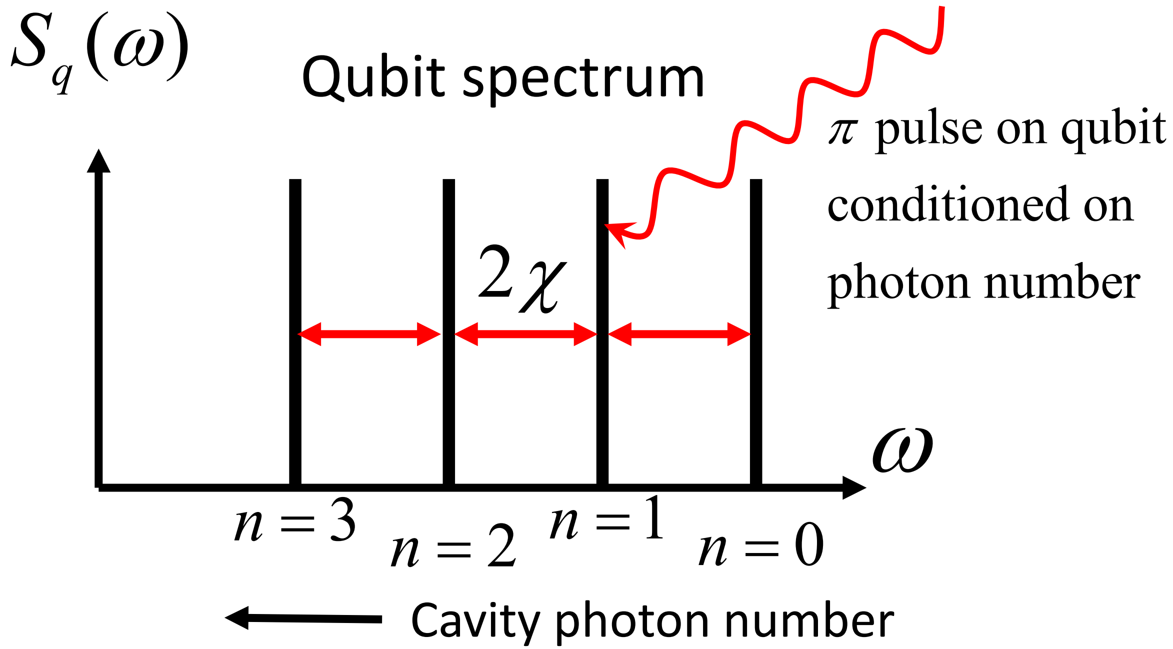
Efficient Boson Sampling from the Photon Number Distribution

-via binary search for the photon number

Is the photon number equal to
1? Yes or no?
13? Yes or no?

[If there are, say, 256 possible photon numbers, the answer is likely to be 'no' most of the time.]

Inefficient sampling implies
large query complexity.]

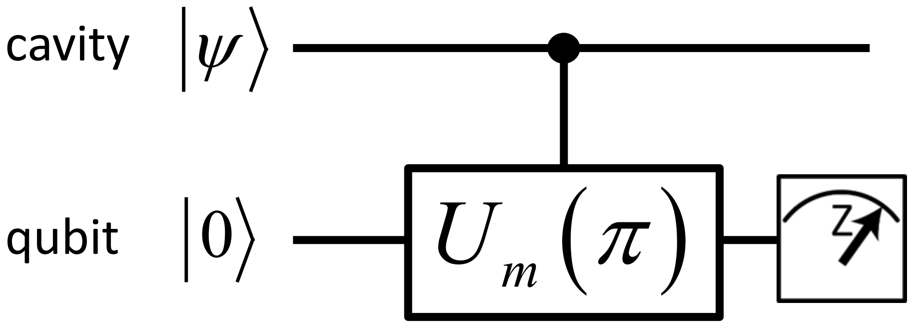


Qubit-Cavity Strong Dispersive Coupling

$$\omega_c \neq \omega_q$$

$$H = \omega_c a^\dagger a + \frac{\omega_q}{2}\sigma^z + \chi\,\sigma^z\, a^\dagger a$$

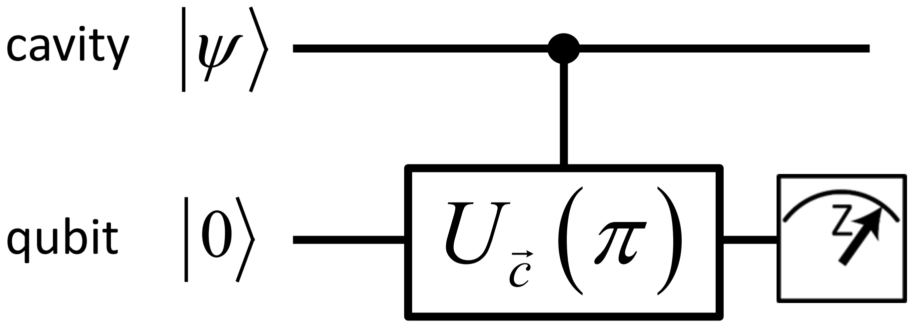
Measure cavity photon number by its effect qubit transition frequency. [QND]

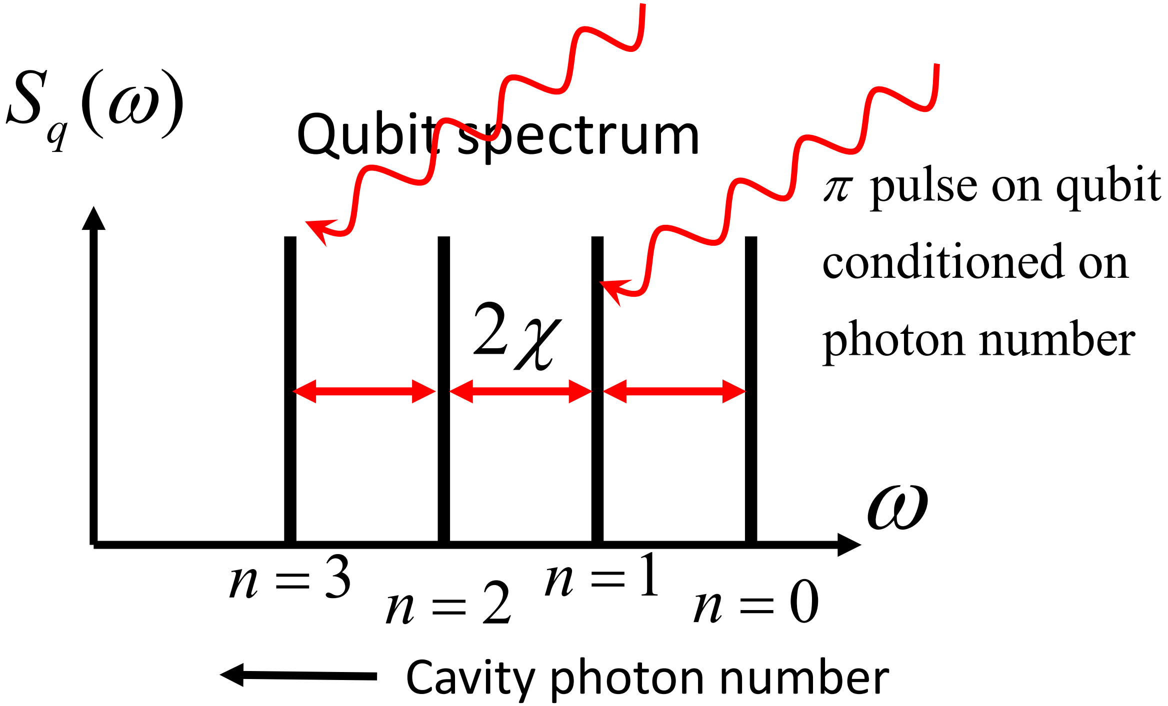


$$U_m(\pi) = e^{-i\frac{\pi}{2}\sigma^x \hat{P}_m} \qquad \hat{P}_m \equiv |m\rangle\langle m|$$

Measure <u>any arbitrary binary function</u> of the photon number. [QND]

Is the photon number equal to either 1 or 3?
Yes or no?



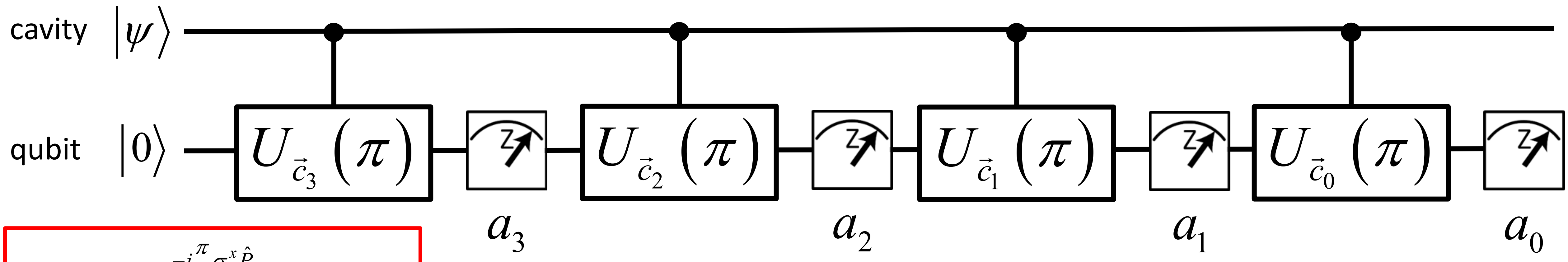


$$U_{\vec{c}}(\pi) = e^{-i\frac{\pi}{2}\sigma^x \hat{P}_{\vec{c}}}$$

$$\hat{P}_{\vec{c}} \equiv \sum_{m=0}^{n_{\max}} c_m \hat{P}_m, \quad c_j \in \{0,1\}$$

Example: binary search for photon number

More convenient than phase estimation—
no feedforward required + obtain most significant bits first



$$U_{\vec{c}}(\pi) = e^{-i\frac{\pi}{2}\sigma^x \hat{P}_{\vec{c}}}$$

$$\hat{P}_{\vec{c}} \equiv \sum_{m=0}^{n_{\max}} c_m \hat{P}_m \,, \quad c_j \in \{0,1\}$$

$$\vec{c}_0 = [1010101010101010]$$ Parity

$$\vec{c}_1 = [1100110011001100]$$

$$\vec{c}_2 = [1111000011110000]$$

$$\vec{c}_3 = [1111111100000000]$$

Walsh-Hadamard transform

circuit cost: $\log_2 n_{\max}$
efficient boson sampling
(exponential gain)

Binary digits in measured photon number

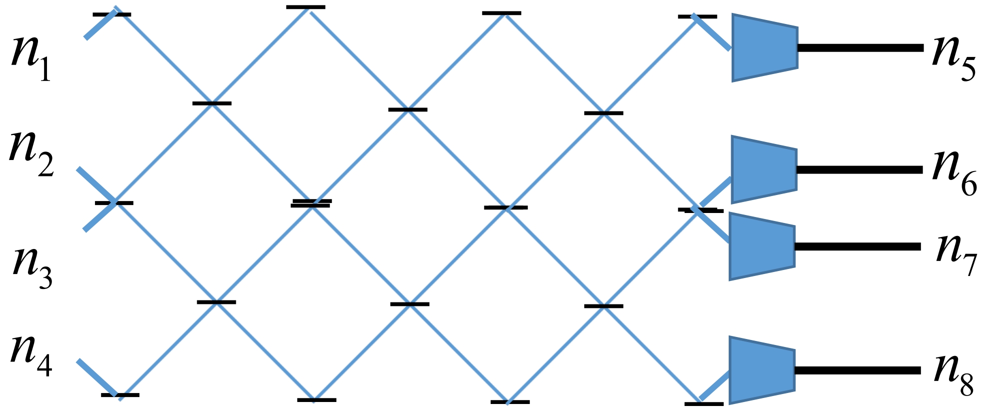
$$[b_3 b_2 b_1 b_0]$$
$$b_3 = a_3$$
$$b_2 = a_3 \oplus a_2$$
$$b_1 = a_3 \oplus a_2 \oplus a_1$$
$$b_0 = a_3 \oplus a_2 \oplus a_1 \oplus a_0$$

Using this control and measurement toolbox for

hardware-efficient simulation of physical models containing bosons.

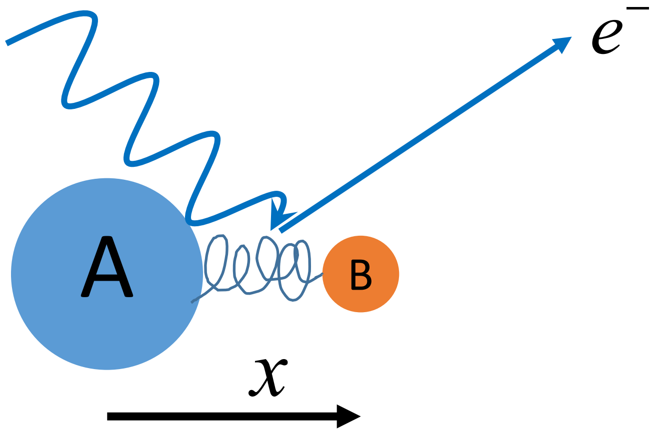
Experimental simulation of the optical spectra of vibrating molecules

Franck-Condon factors as a boson sampling problem



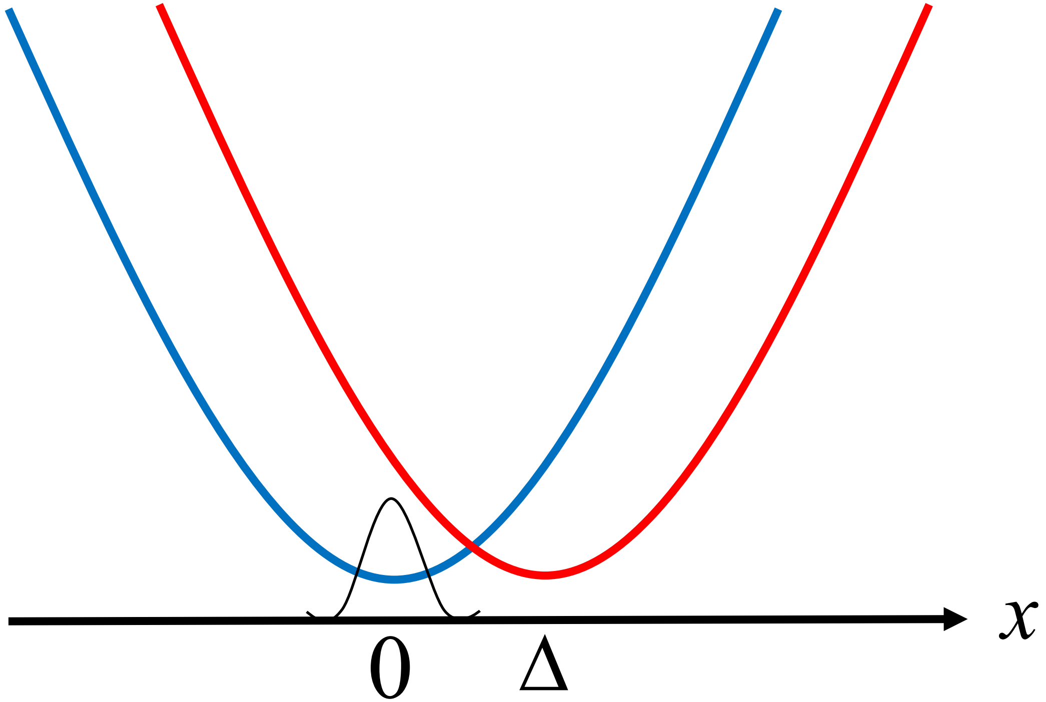
Huh et al. *Nature Photonics* **9** 615 (2015)

Warm up example: the suddenly displaced harmonic oscillator

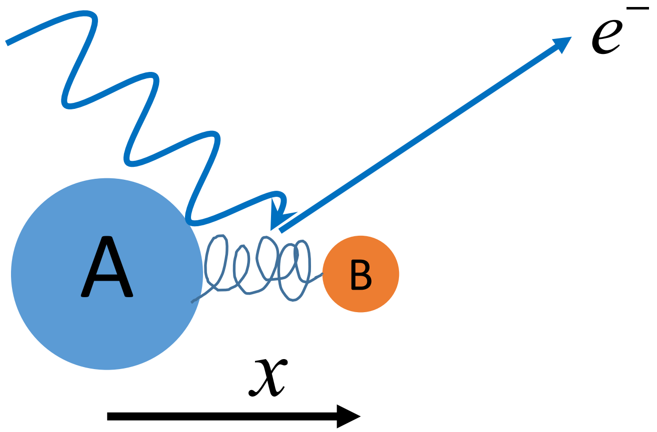


Photon emission ejects a bonding electron suddenly changing the equilibrium spacing of the two nuclei

$$H(t) = \frac{p^2}{2m} + \frac{1}{2} k \left[x - \Delta \theta(t) \right]^2$$



Warm up example: the suddenly displaced harmonic oscillator

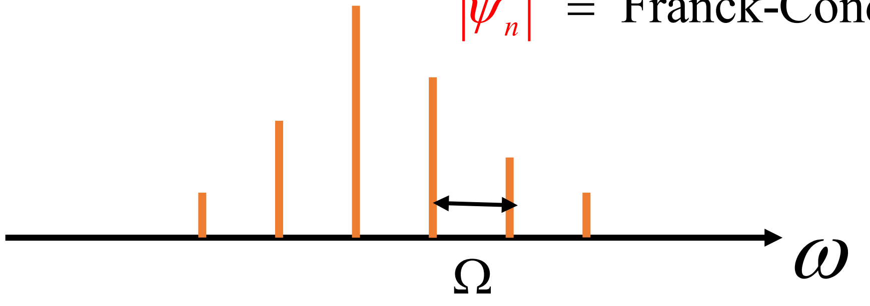


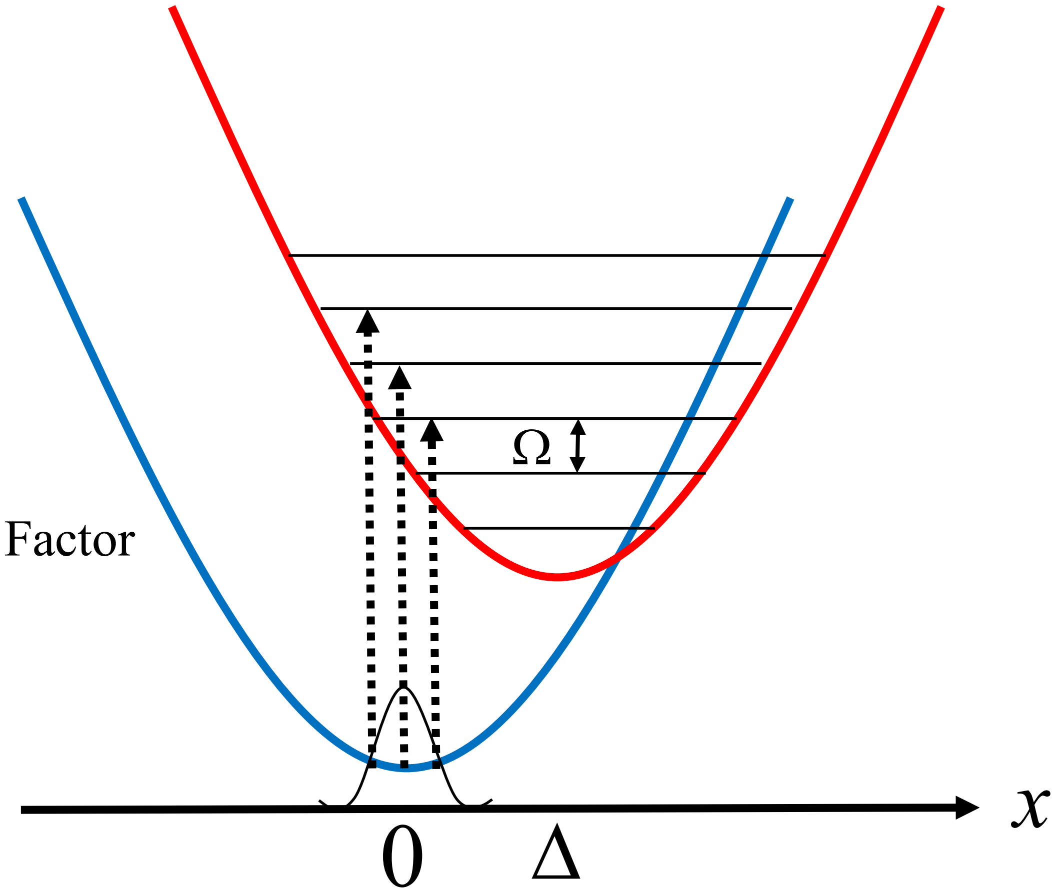
Nuclear wave function has no time to change. Sudden projection onto eigenstates of the new Hamiltonian.

$$|\Psi\rangle = \sum_n \psi_n |n\rangle$$

$$S(\omega) = \sum_n |\psi_n|^2 \delta(\omega - n\Omega)$$

$|\psi_n|^2$ = Franck-Condon Factor





Warm up example: the suddenly displaced harmonic oscillator

$$|\Psi\rangle_{\text{old basis}} = |0\rangle$$

$$|\Psi\rangle_{\text{new basis}} = e^{-\Delta[a^\dagger - a]}|0\rangle = |-\Delta\rangle$$

$$|\Psi\rangle_{\text{new basis}} = e^{-\frac{|\alpha|^2}{2}} \sum_n \frac{\alpha^n}{\sqrt{n!}}|n\rangle$$

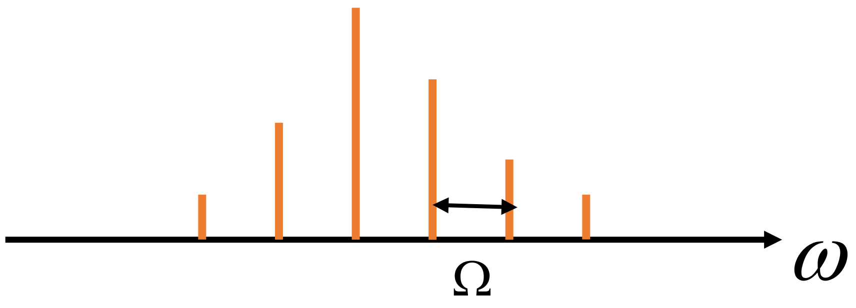
Nuclear wave function has no time to change. Sudden projection onto eigenstates of the new Hamiltonian.

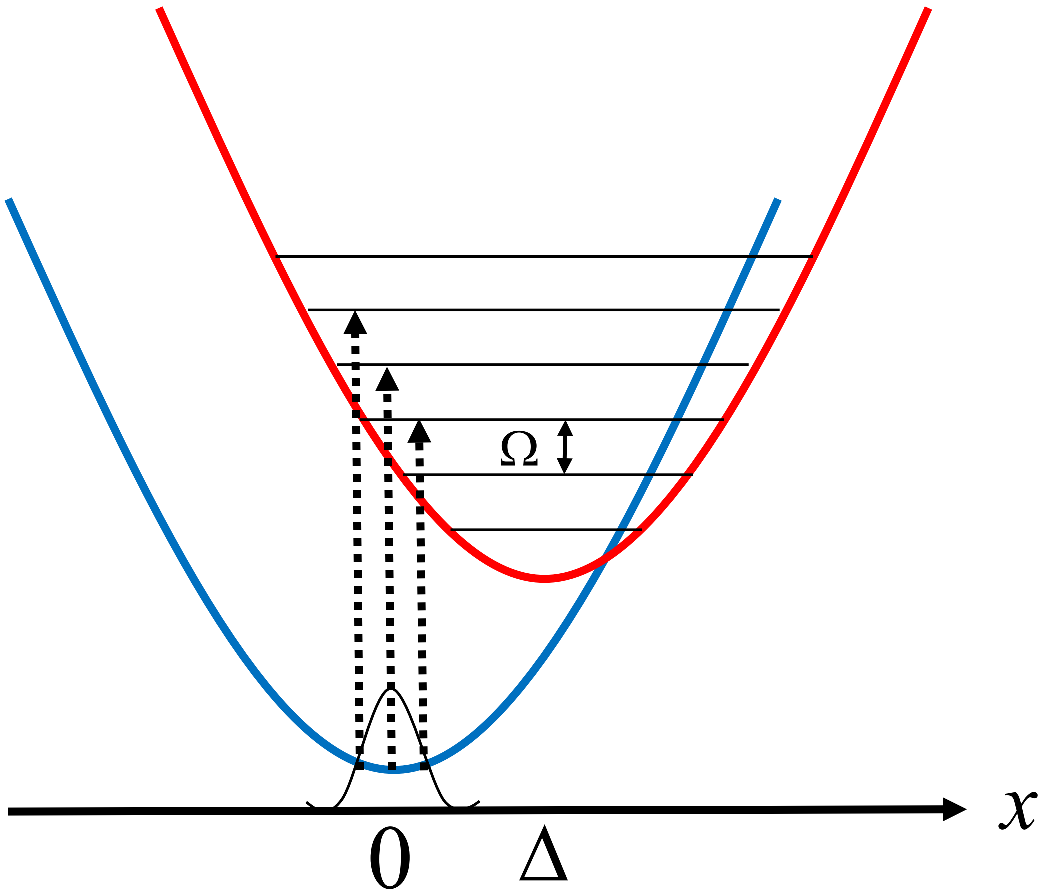
Spectral function: Franck-Condon Factor

$$S(\omega) = e^{-\bar{n}} \sum_n \frac{\bar{n}^n}{n!} \delta(\omega - n\Omega)$$

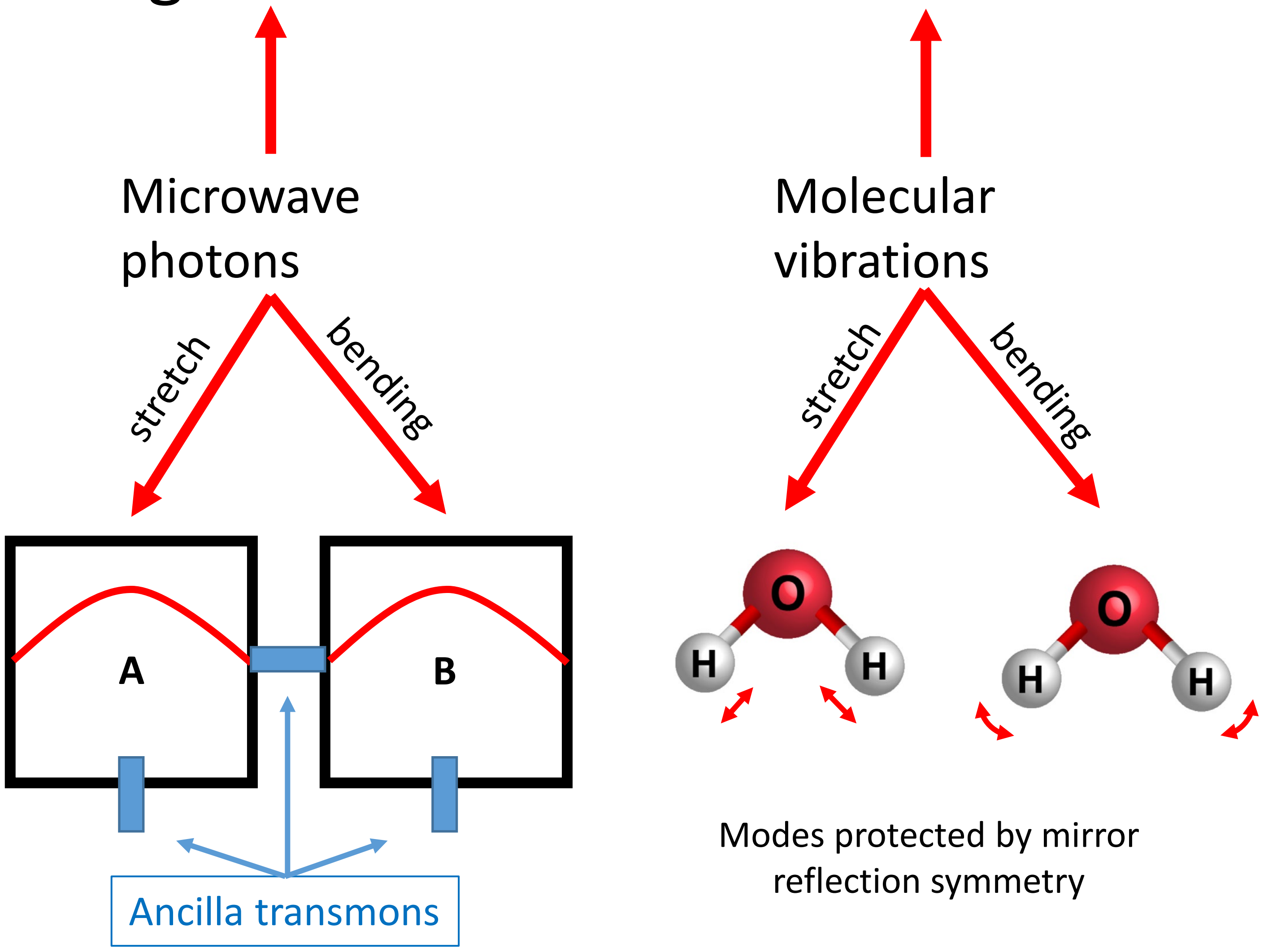
where $\bar{n} = |\alpha|^2 =$ mean excitation number.

Poisson distribution.



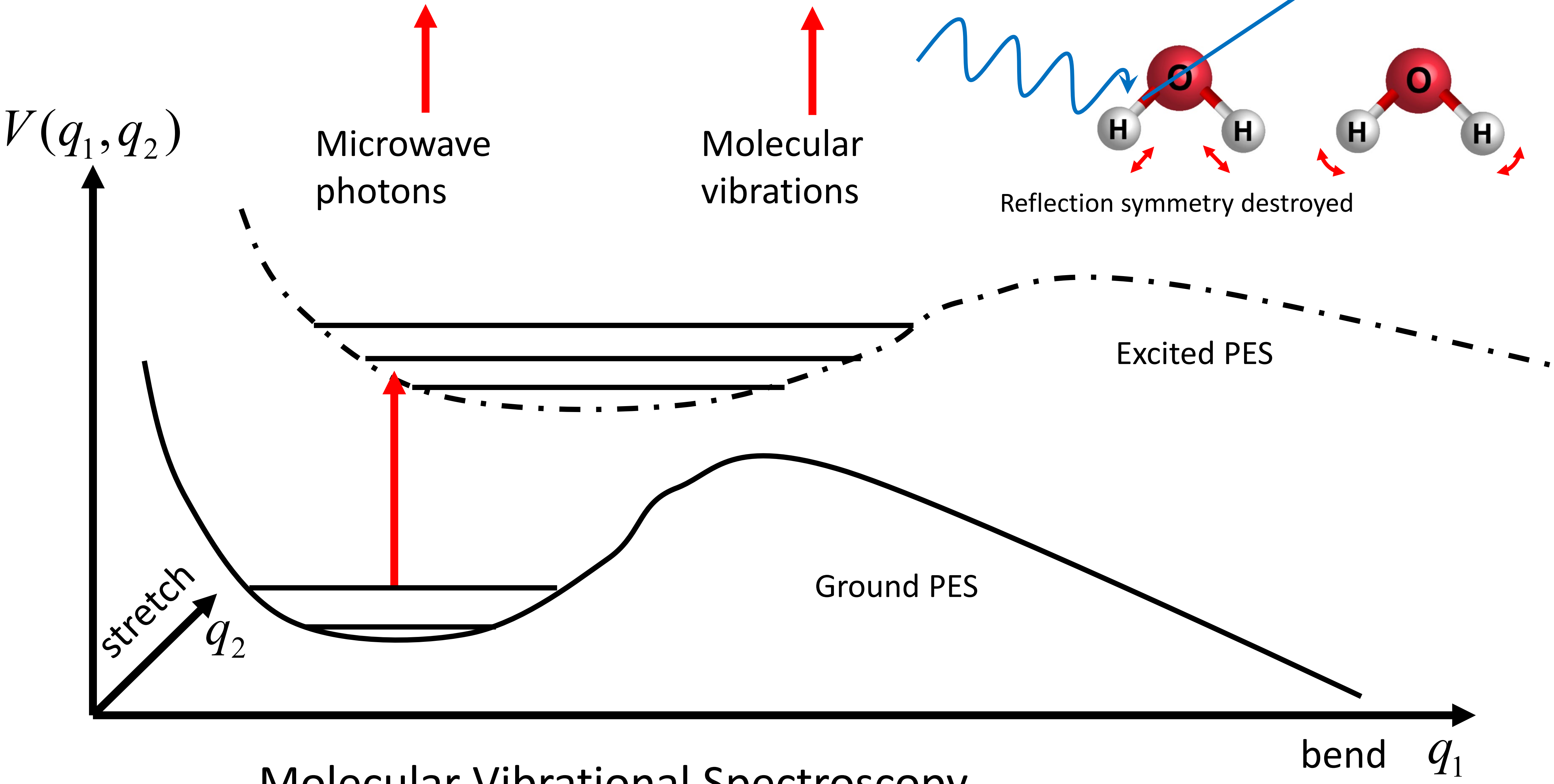


Using Bosons to Simulate Bosons

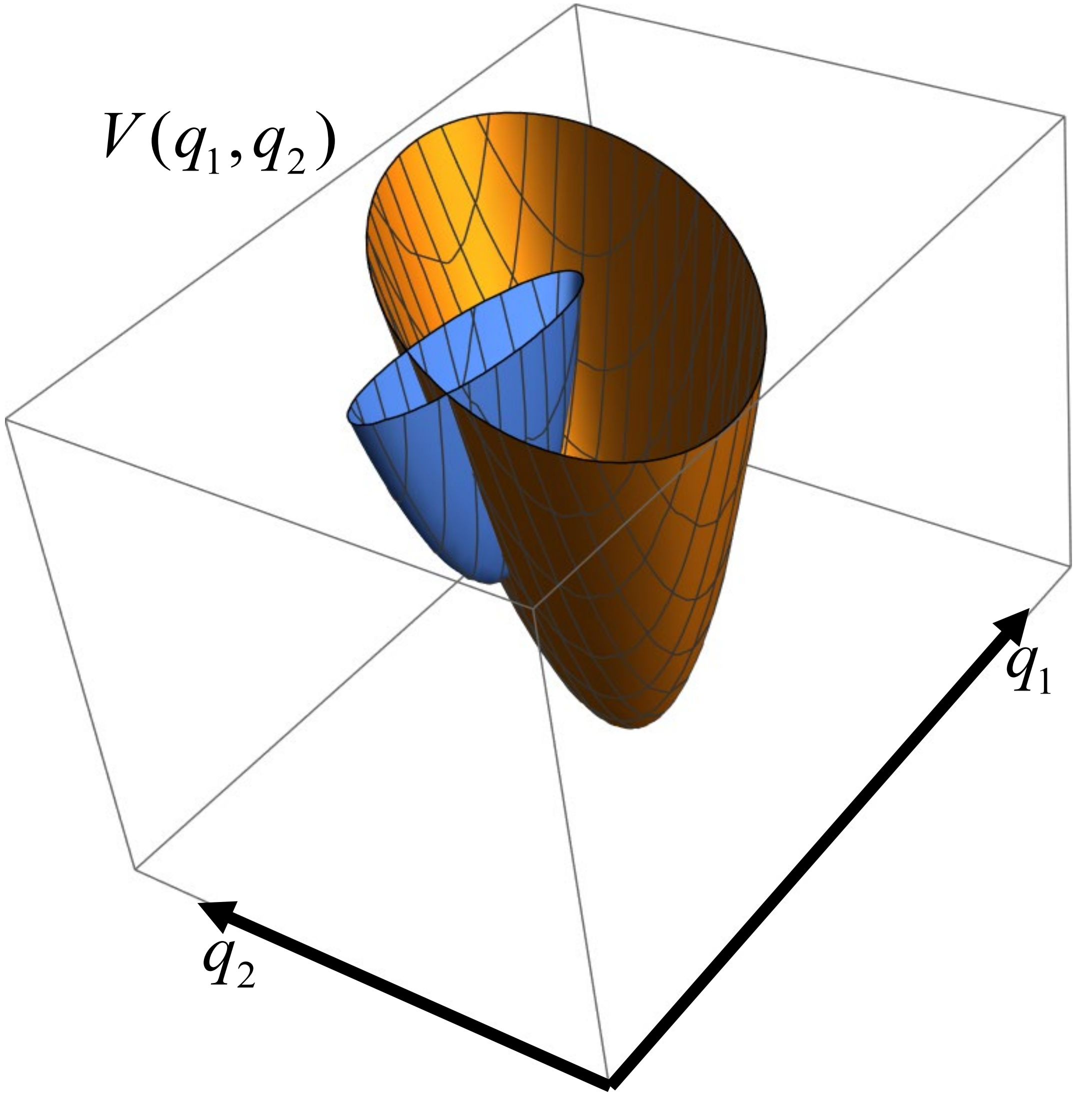


Molecular Vibrational Spectroscopy

Using Bosons to Simulate Bosons



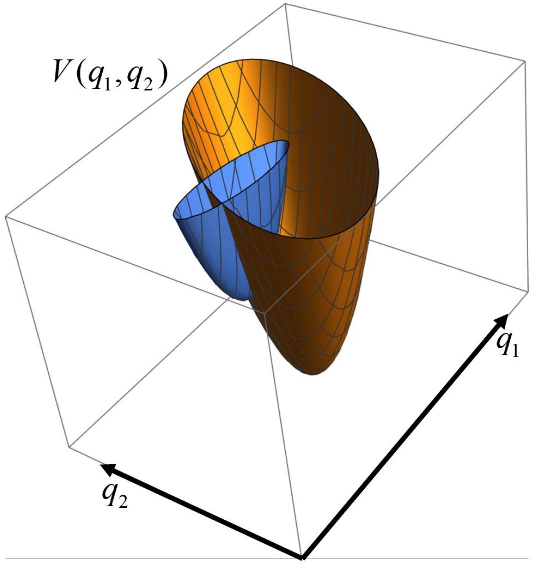
Molecular Vibrational Spectroscopy



First-generation experiment:
Chris Wang (Schoelkopf lab)
Phys. Rev. X 10, 021060 (2020)

1. Obtain nuclear PES from solving fermionic problem on classical computer.
2. Approximate nuclear PES as quadratic
3. But allow for different frequencies, displacement, squeezing, and orientation of symmetry axes (reflection symmetry destroyed) of PES between electronic ground and excited states.
4. Sudden approximation: Perform unitary transformation between eigenstates of ground and excited state Hamiltonians.

Doktorov et al. *J. Mol. Spec.* **64** 302-326 (1977)



Requirements:

(rotations between modes)

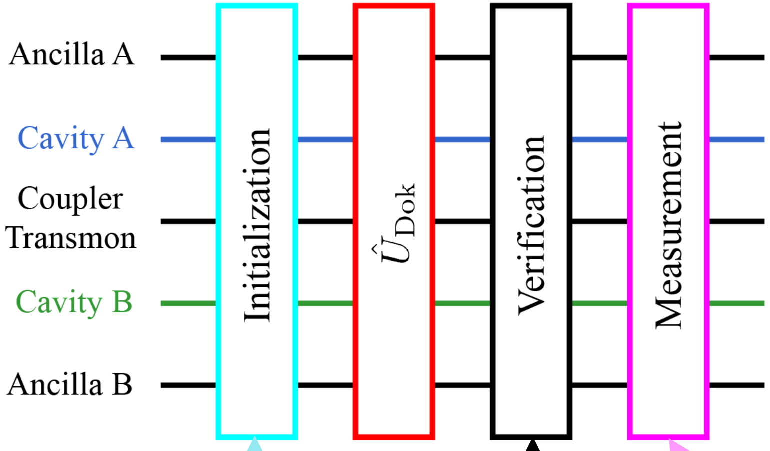
- bosonic modes
- Gaussian operations: beamsplitters, squeezing, displacements Gao et al. *PRX* **8** 2 (2018)
- non-Gaussian state preparation Heeres et al. *Nat. Comm.* **8** 1 (2017)
- number-resolved detection Wang et al. Phys. Rev. X 10, 021060 (2020)

Challenging in conventional quantum optics

Huh et al. *Nature Photonics* **9** 615 (2015)

Circuit implementation of the Franck-Condon simulation

Phys. Rev. X 10, 021060 (2020)



Non-Gaussian state preparation

Check error flags (ancillae)

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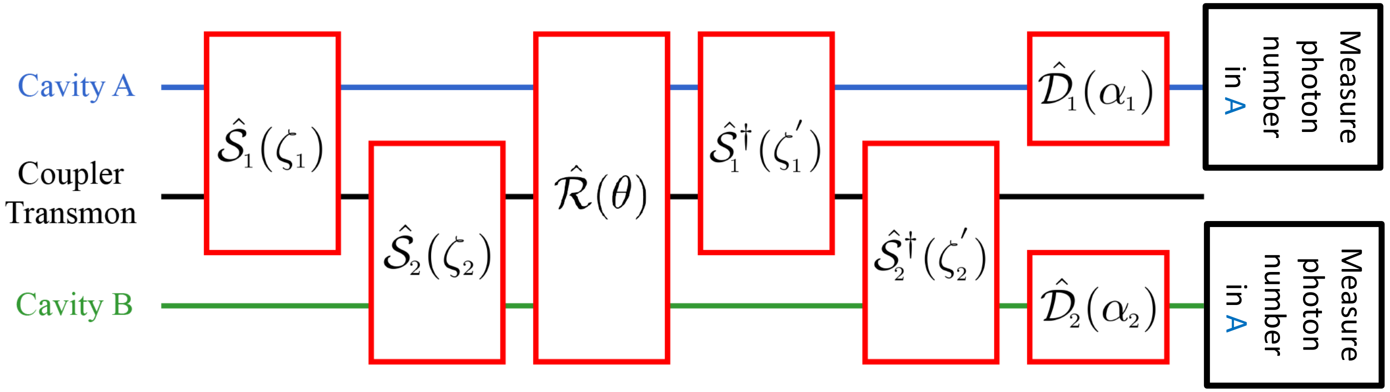
Number-resolving measurements in each cavity

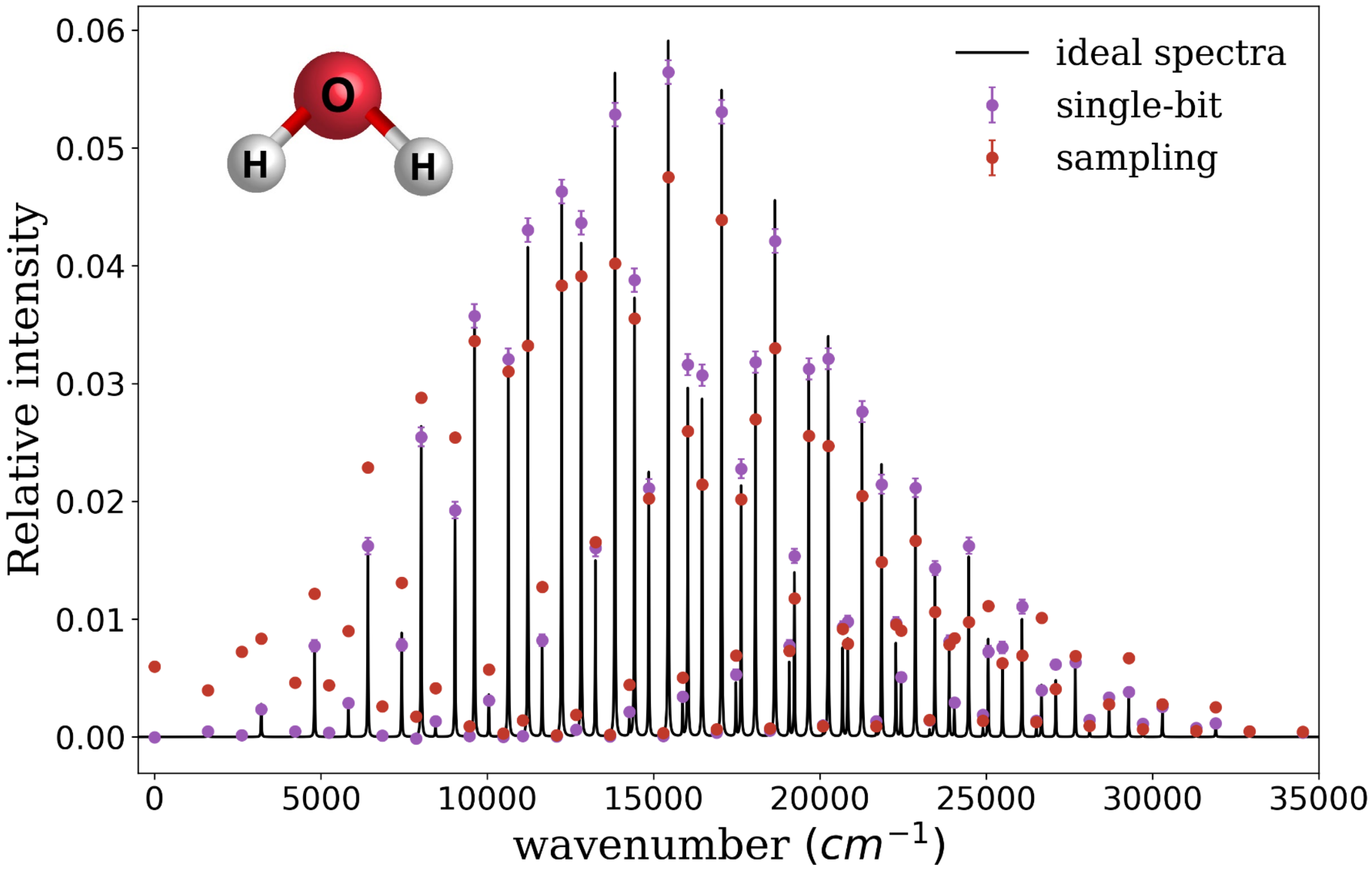
$\hat{U}_{\text{dok}}$ Unitary basis change between ground and excited PES



Histogram the measured photon numbers n_A, n_B from many shots to obtain the probability distribution $P(n_A, n_B)$ and the spectral density

$$S(\omega) = \sum_{n_A, n_B} P(n_A, n_B)\, \delta(\omega - n_A \Omega_A - n_B \Omega_B)$$

Experimentally simulated photoelectron processes via efficient boson sampling (photons represent phonons)



$$H_2O \rightarrow H_2O^+(\tilde{B}\,^2B_2) + e^-$$

$$|\psi_0\rangle = |0,0\rangle$$

L_1 distance between exact and experimental distributions:

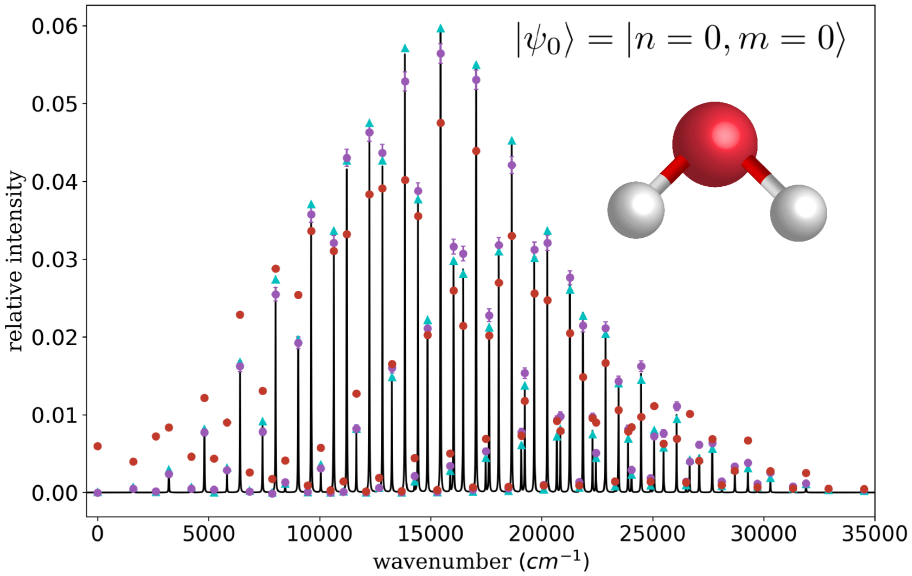
$$D = \frac{1}{2}\sum_{i,j}\left|p_{ij} - q_{ij}\right|$$

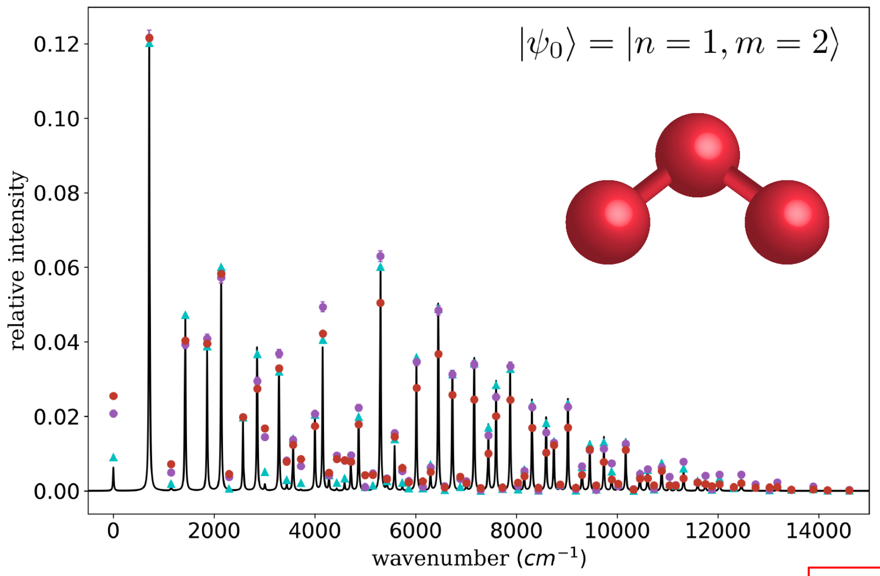
$$D_{\text{single-bit}} = 0.049$$

$$D_{\text{sampling}} = 0.152$$

Phys. Rev. X 10, 021060 (2020)

[Chris Wang]





Phys. Rev. X 10, 021060 (2020)

Typical photodetectors are not number resolving and are destructive.

Here we have efficient QND single-shot boson number sampling.

We measure which of D=256 photon states the two cavities are in by QND measurement of the 'digits' in the binary representation of the photon number:

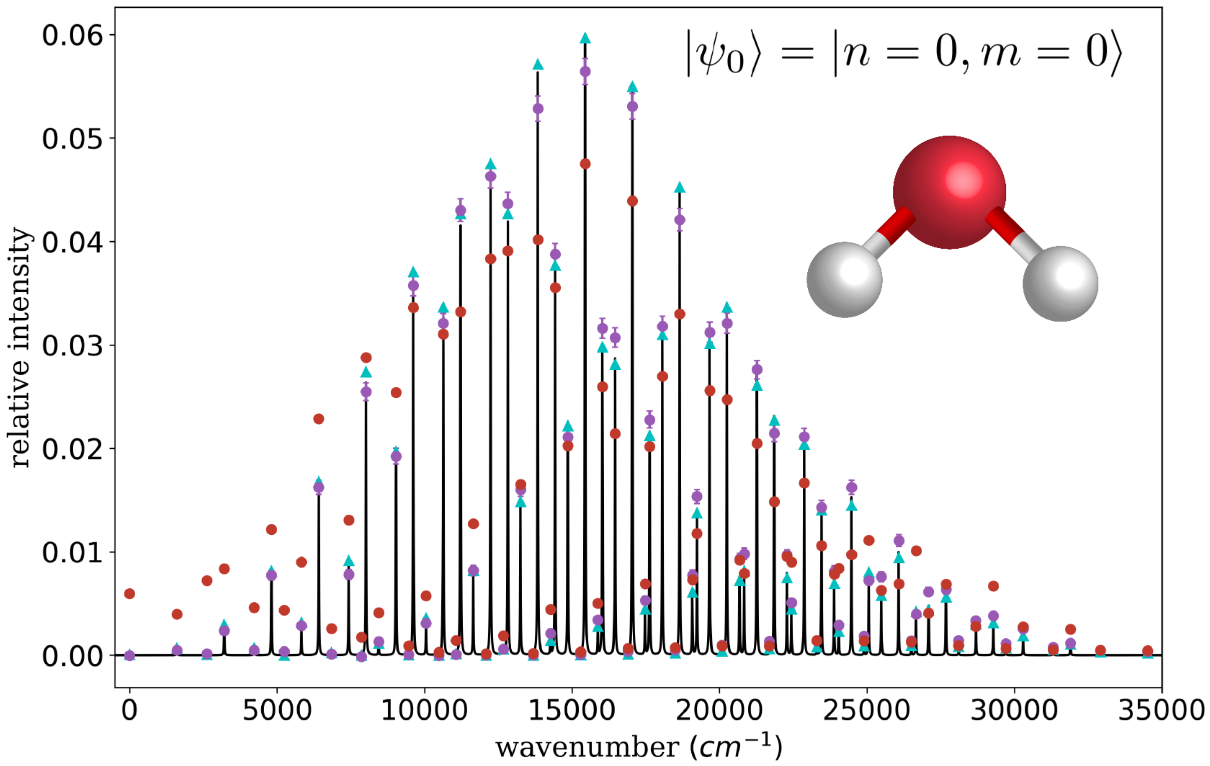
$$[n,m]=[(b_3,b_2,b_1,b_0),(c_3,c_2,c_1,c_0)]$$

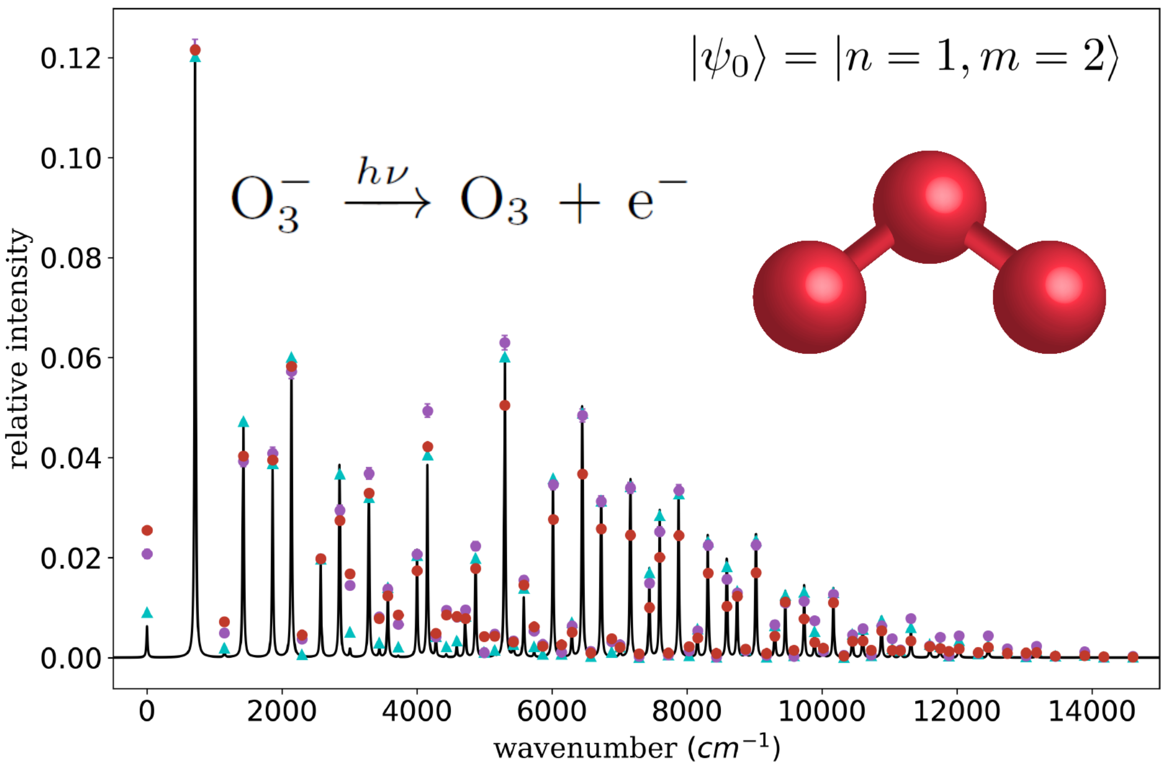
Circuit complexity cost is only log D, not D. (Exponential gain, true boson sampling)

$$H_2O \xrightarrow{h\nu} H_2O^+\left(\tilde{B}\,^2B_2\right) + e^-$$

Phys. Rev. X 10, 021060 (2020)

‘exact’ (cyan), single bit extraction (purple) and sampling (red) measurement

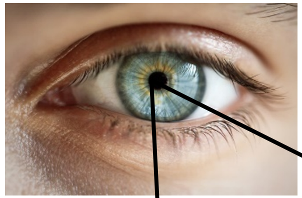




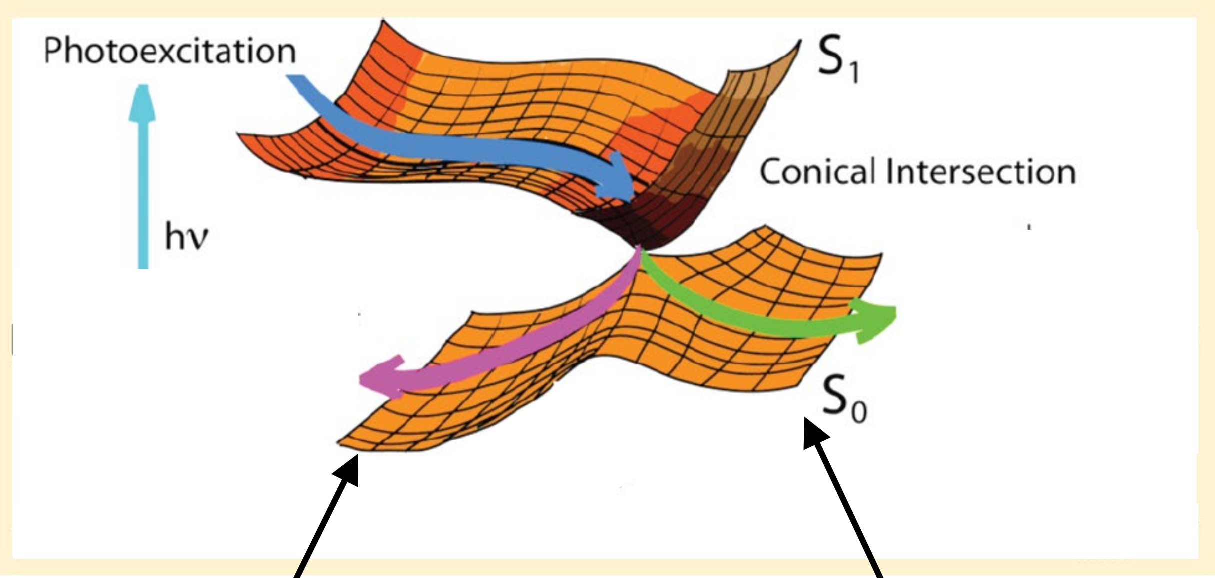
Microwave bosons to simulate vibrational bosons is highly advantageous.
Would have required >8 qubits and ~10^3 gates in an ‘ordinary’ quantum computer.

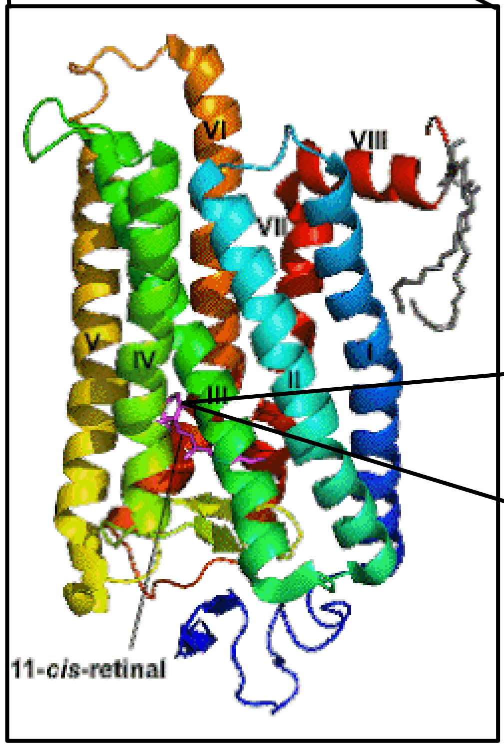
Conical Intersections and vision

Polli et al. *Nature* (2010).



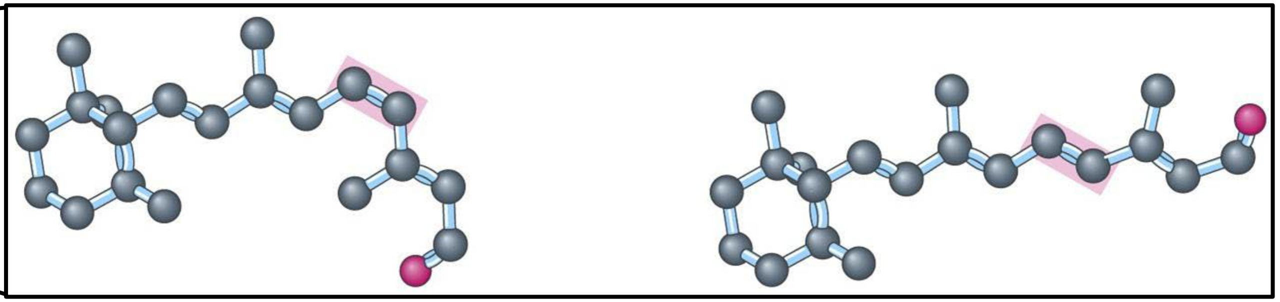
Conical intersections of molecular nuclear potential energy surfaces





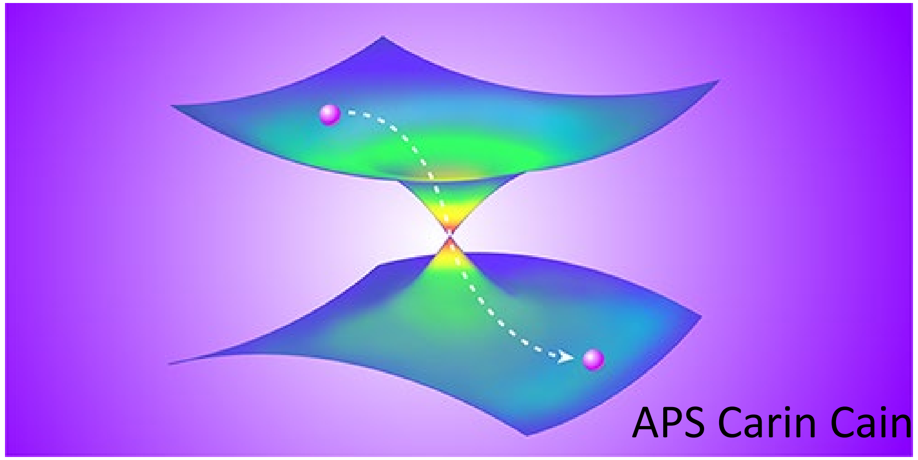
rhodopsin

retinal (vitamin A)



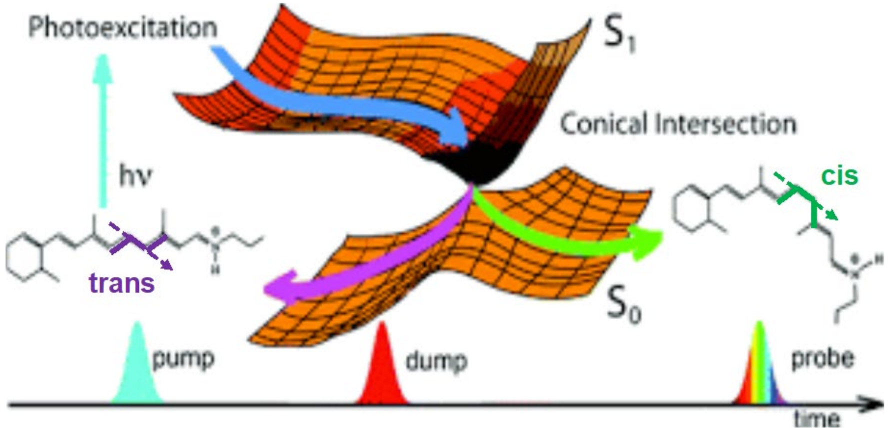
cis trans

Quantum Circuit Tackles "Diabolical" Photochemical Process
January 26, 2023• *Physics* 16, s14
https://physics.aps.org/articles/v16/s14



Conical Intersections activate human vision

trans/cis photoisomerization in retinal chromophore

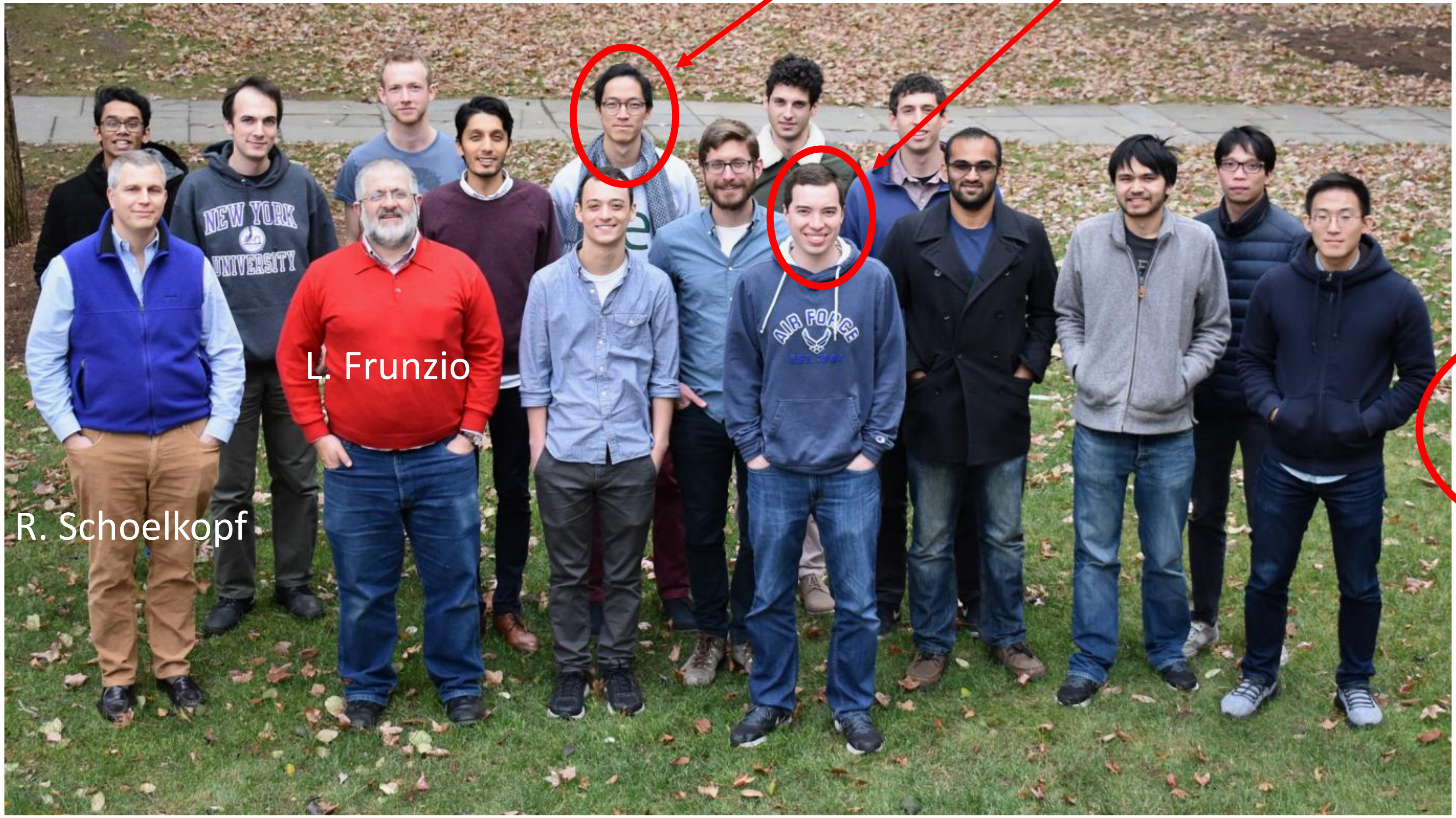


Challenges
- Nonadiabatic effects
- Anharmonicities
- Dissipation

Microwave Boson Simulation Experiment: Chris Wang (Schoelkopf lab)
Phys. Rev. X 13, 011008 (2023)

Breakdown of Born-Oppenheimer adiabatic approximation
Environmental dissipation

Schoelkopf Lab

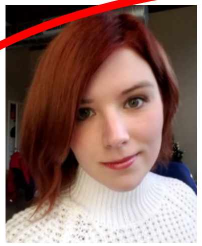


Actual Chemists


B. Lester

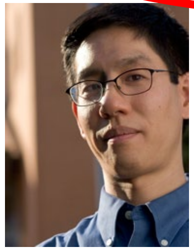

Y. Y. Gao


Y. Zhang


J. Freeze


V. Batista


P. Vaccaro


I. Chuang


L. Jiang


S. Girvin

U.S. DEPARTMENT OF ENERGY

QuantumInstitute.yale.edu

& many others!

Devoret Lab