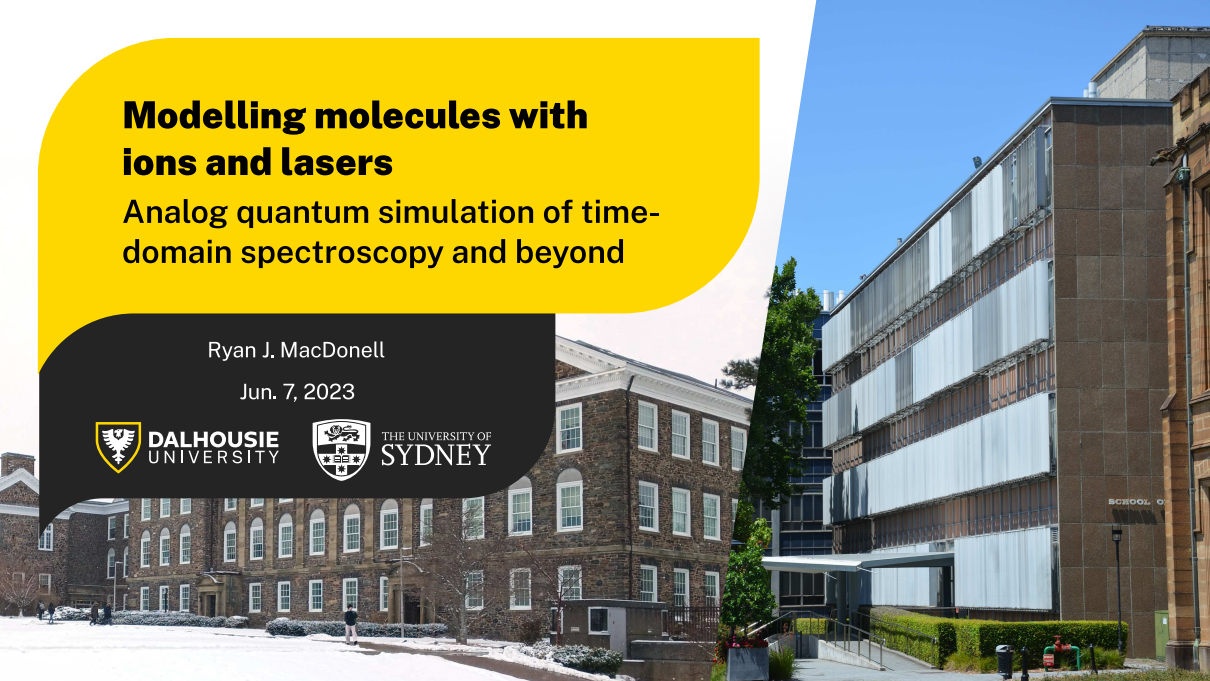
Modelling molecules with ions and lasers

Analog quantum simulation of time-domain spectroscopy and beyond

Ryan J. MacDonell

Jun. 7, 2023



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Acknowledgements

- Kassal group
  - Claire Dickerson (UCLA)
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  - Clare Birch (Blackbird VC)
  - Liam Flew
  - Alok Kumar (UPenn)
  - Ben Stewart
  - Vanessa Olaya Agudelo
  - Michael Currington (UNSW)
- Quantum control laboratory
  - Claire Edmunds (Innsbruck)
  - Cornelius Hempel (PSI)
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  - Ting Rei Tan

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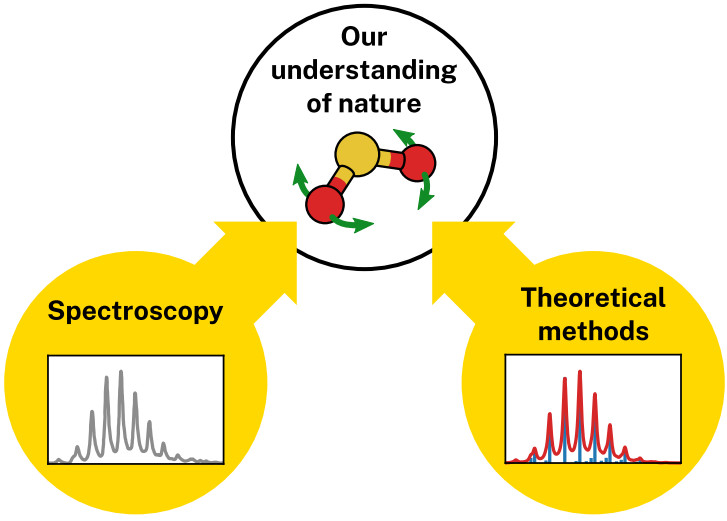
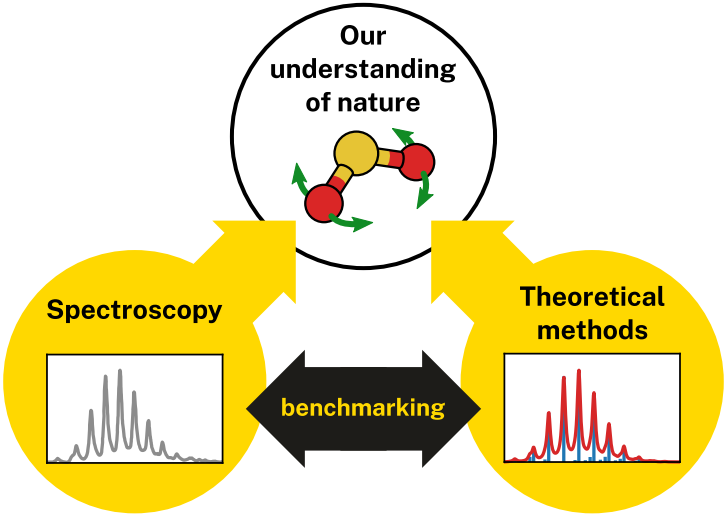
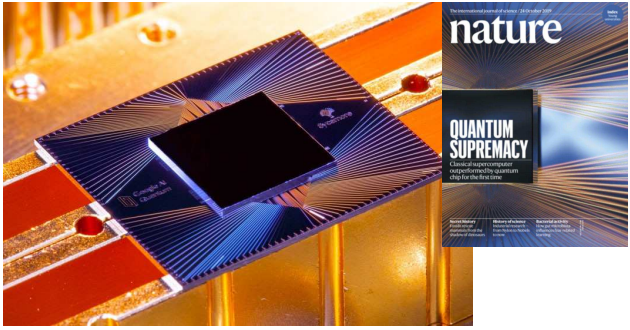
Spectroscopy and theory



Spectroscopy and theory

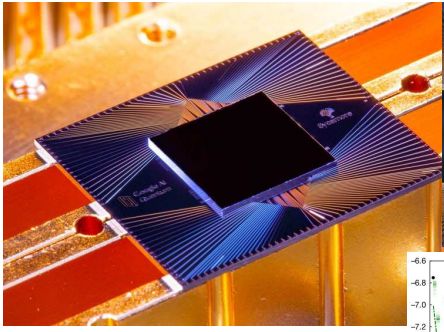


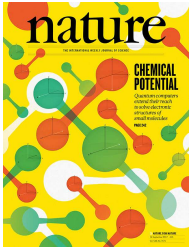
Simulation with quantum computers



Arute, F. et al. *Nature* **2019**, *574*, 505–510.

Simulation with quantum computers





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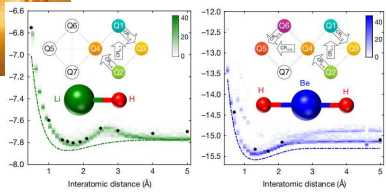
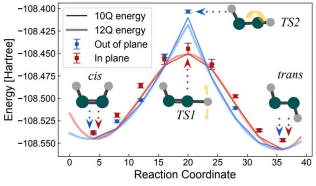
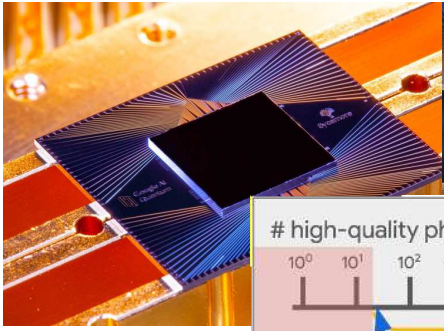




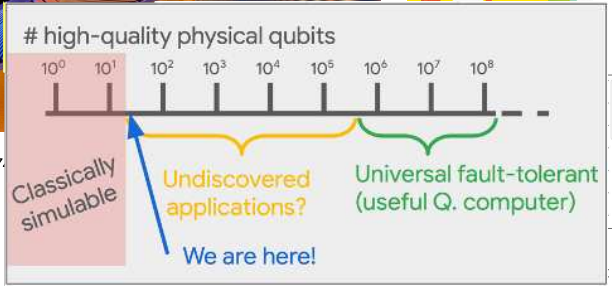
Kandala, A. et al. *Nature* **2017**, *549*, 242–246.

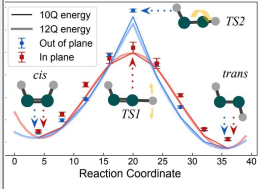
Rubin, N.C. et al. *Science* **2020**, *369*, 1084–1089.

Simulation with quantum computers



Arute, F. et al. *Nature* **2019**, *57*4





al. *Science* **2020**, *369*, 1084–1089.

Analog simulation

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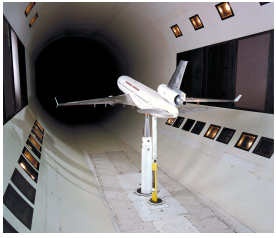
Classical



Analog simulation

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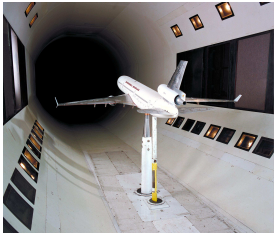
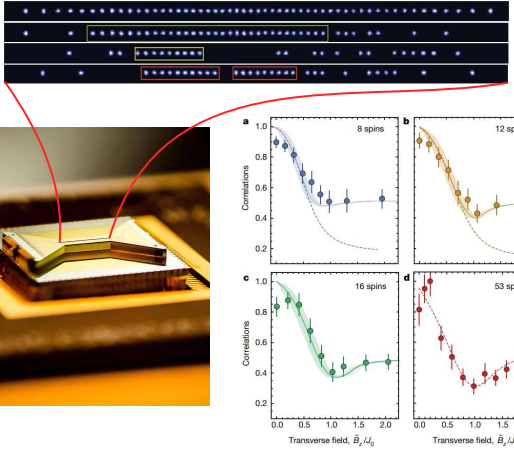


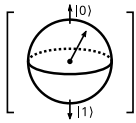
Quantum

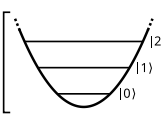


Zhang, J. et al. *Nature* **2017**, *551*, 601–604.

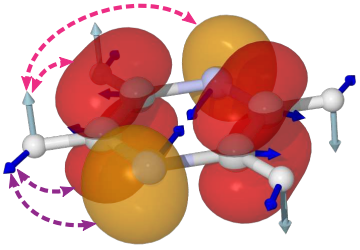
Analog MQB simulators

$$|\Psi\rangle = \left[\;\right]^{\otimes N} \otimes \left[\;\right]^{\otimes M}$$

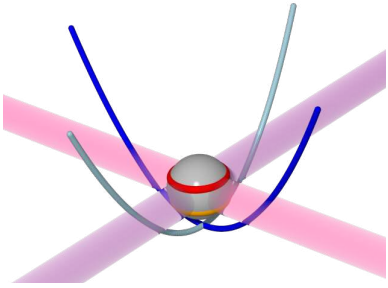




mapping:
electronic → qudit
vibrational → bosonic



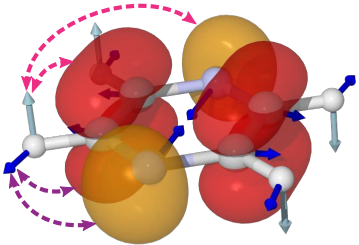
$$\hat{H}_{\text{mol}} = \hat{H}_{\text{sim}}$$



MacDonell, R.J. et al. *Chem. Sci.* **2021**, *12*, 9794–9805.

Analog MQB simulators

$$|\Psi\rangle = \left[\; |0\rangle \; |1\rangle \; \right]^{\otimes N} \otimes \left[\; |2\rangle \; |1\rangle \; |0\rangle \; \right]^{\otimes M}$$



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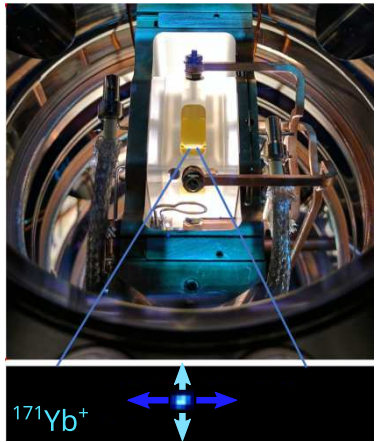

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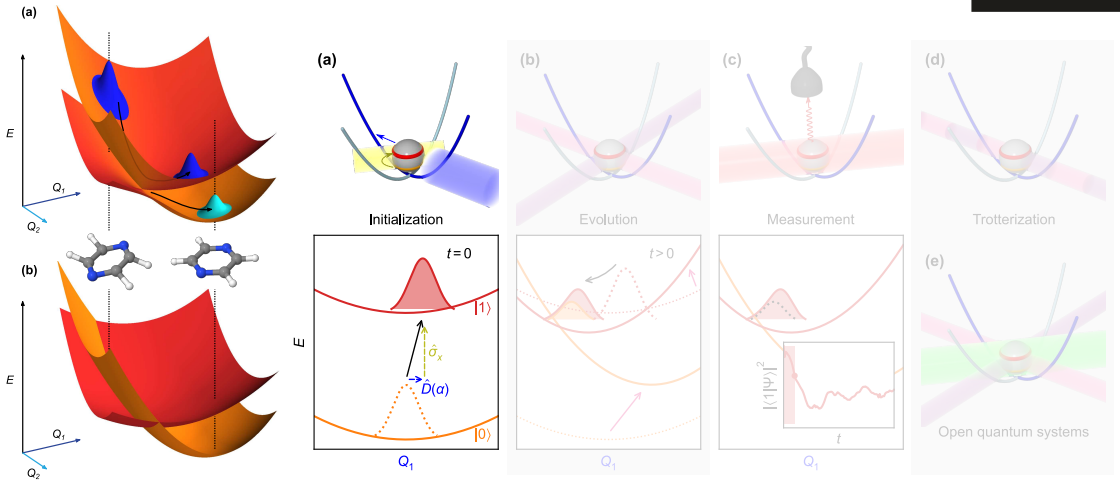
MQB simulation procedure



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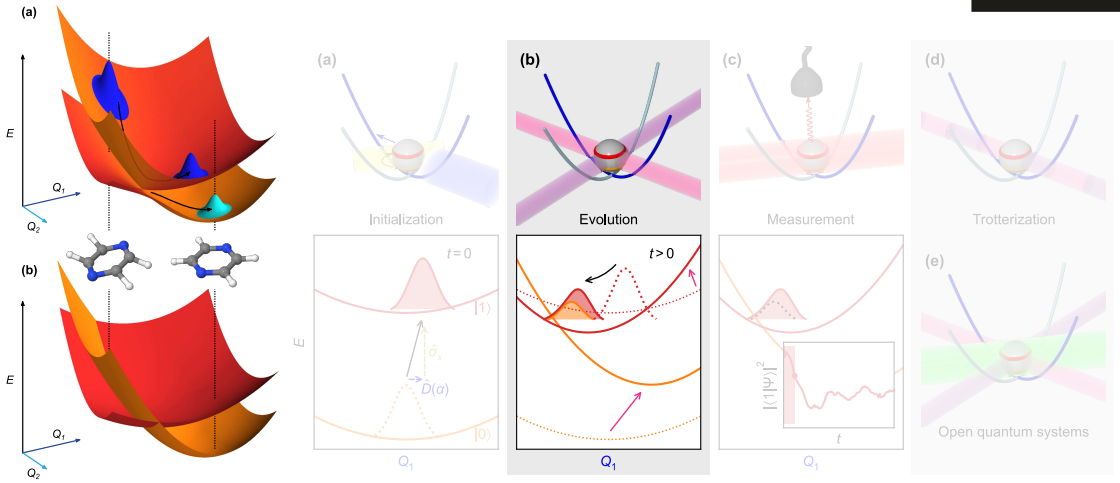

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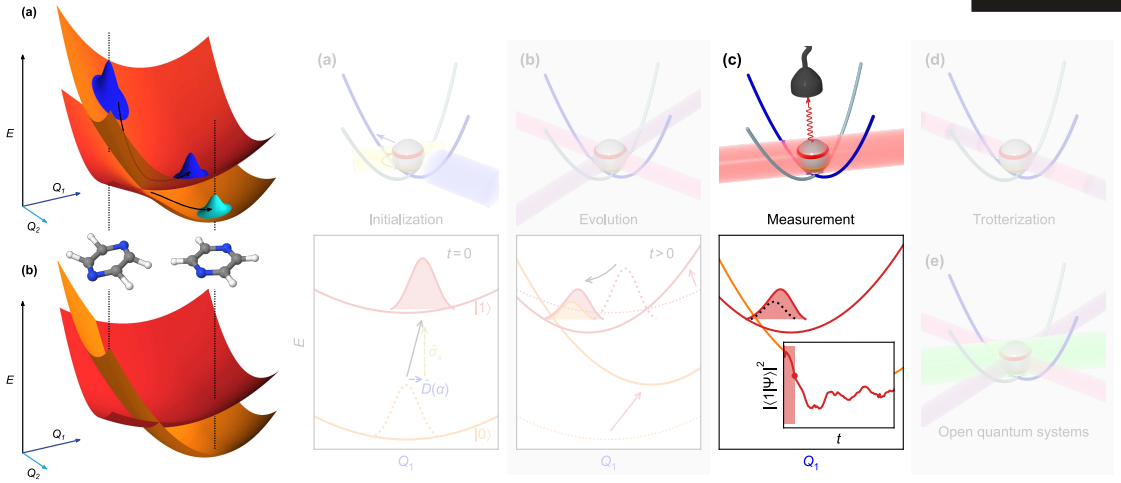

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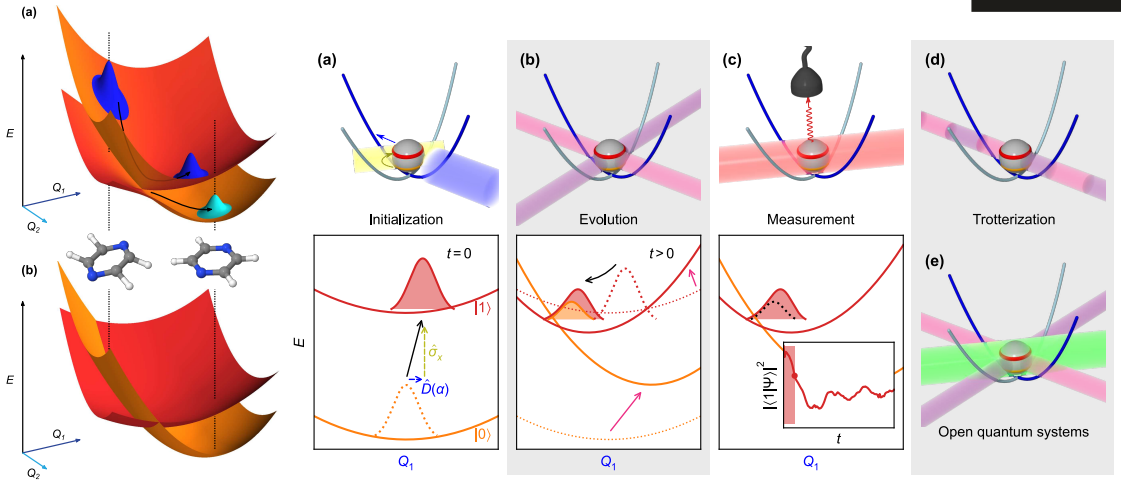

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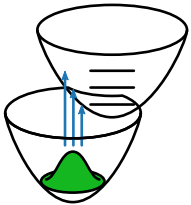
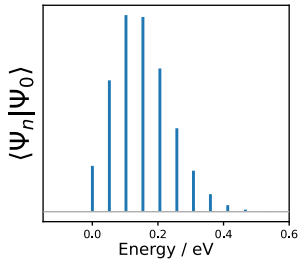
Spectroscopy in the time domain

Frequency domain:





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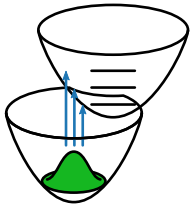
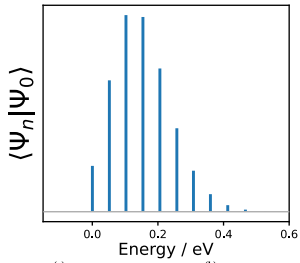
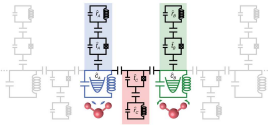
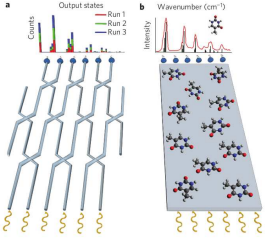
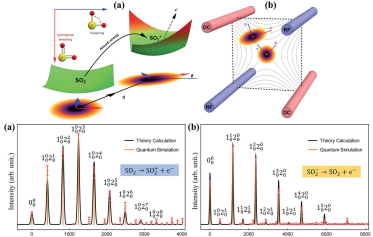




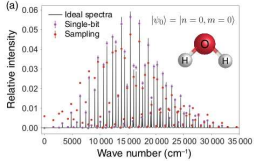




Huh, J. et al. *Nat. Photonics* **2015**, *9*, 615–620.



Shen, Y. et al. *Chem. Sci.* **2018**, *9*, 836–840.



Wang, C. et al. *Phys. Rev. X* **2020**, *10*, 021060.

Spectroscopy in the time domain

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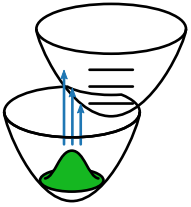

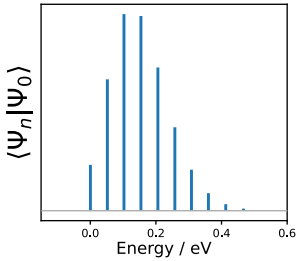

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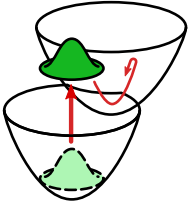

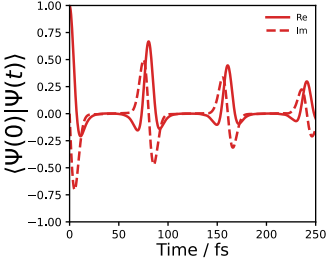

Spectroscopy in the time domain

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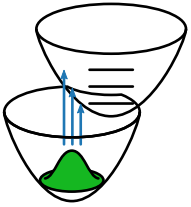

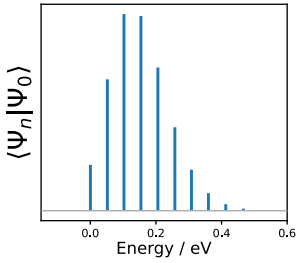

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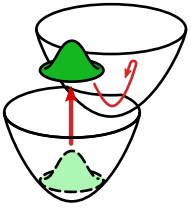

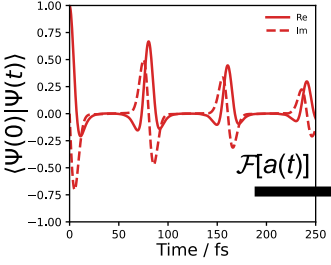

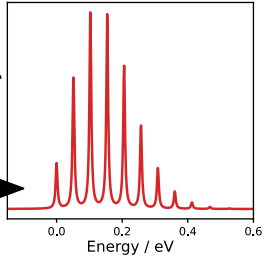

Spectroscopy in the time domain

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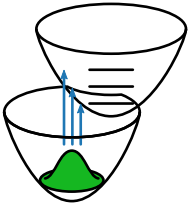
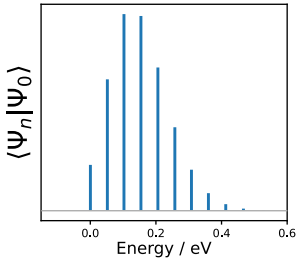
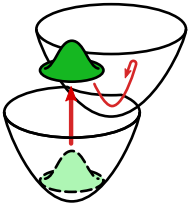


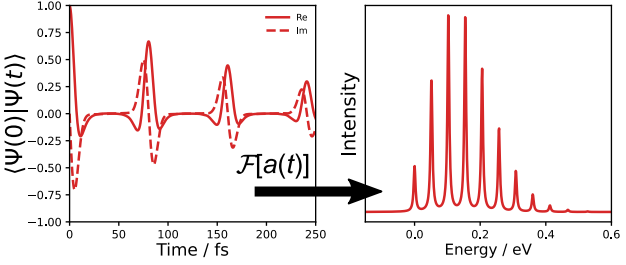


***a(t)* contains information for:**

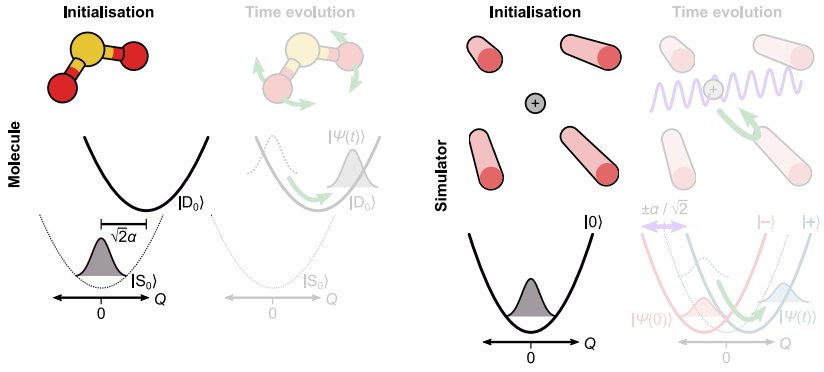
- intensities
- frequencies
- lineshapes

Time domain:





Measuring the autocorrelation function



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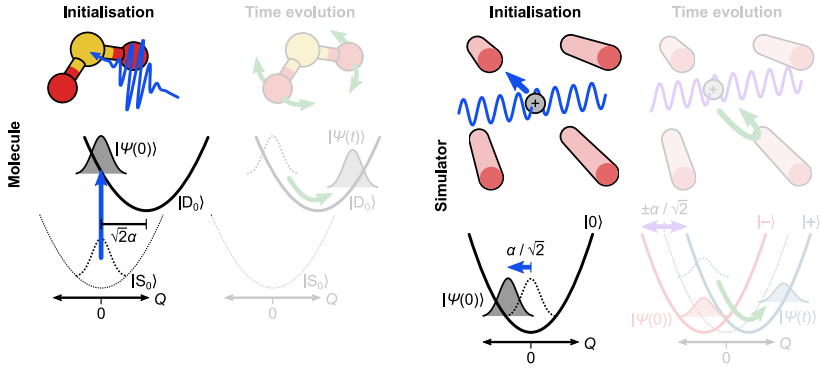

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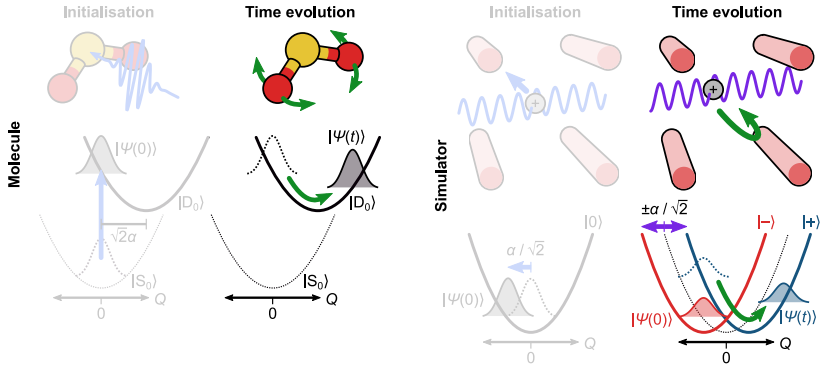

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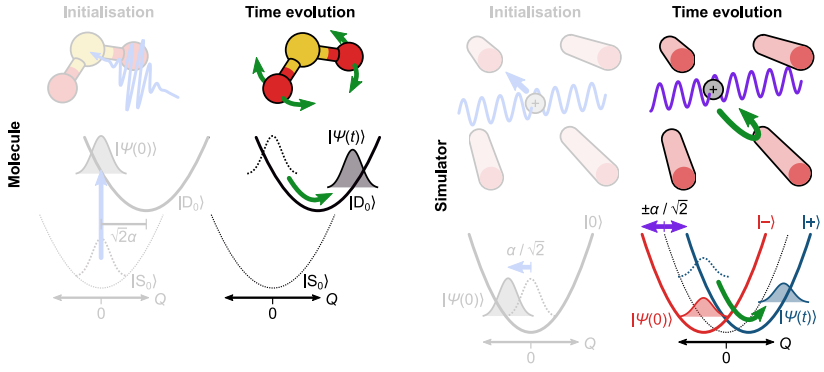

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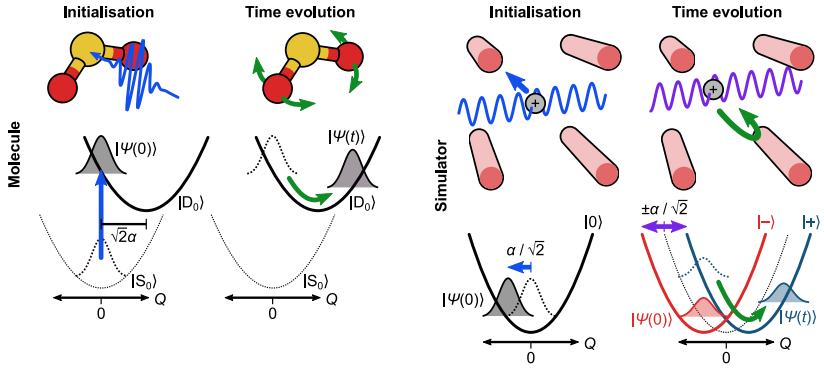
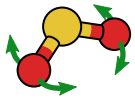
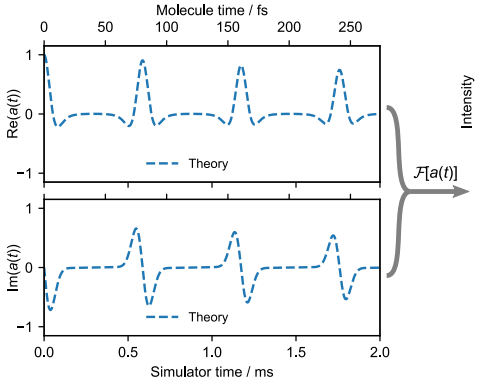


Experimental results



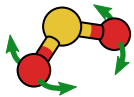
$S_0 \rightarrow D_0$



Model parameters: Lee et al. *J. Mol. Spec.* **2009**, *256*, 279–286.

Expt. spectrum: Holland et al. *Chem. Phys.* **1994**, *188*, 317–337.

Experimental results



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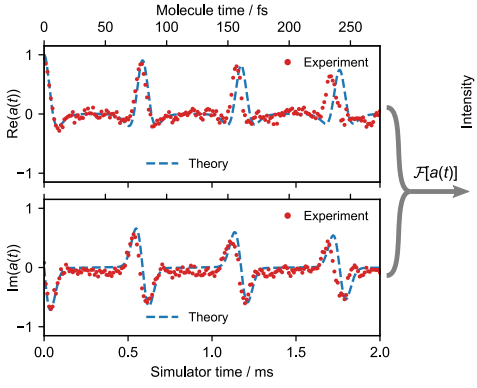

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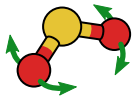

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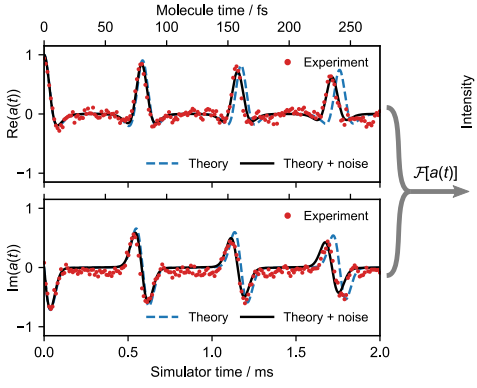

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Simulating spectra beyond SO_2

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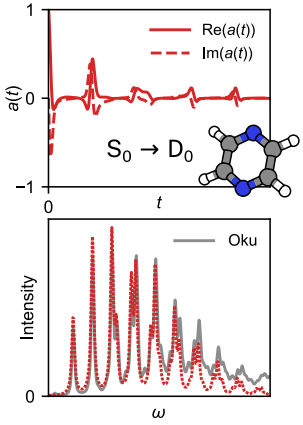
Multiple modes



Oku, M. et al. *J. Phys. Chem A* **2008**, *112*, 2293–2310.

Simulating spectra beyond SO_2

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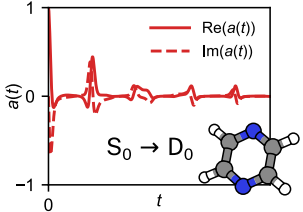

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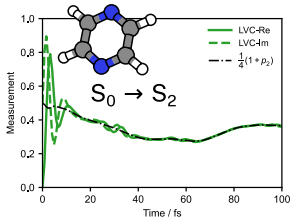
Vibronic coupling



Suzuki, Y.-I. et al. *J. Chem. Phys.* **2010**, *132*, 174302.

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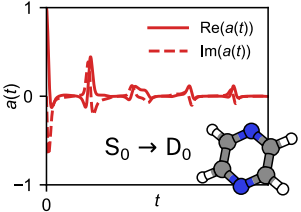

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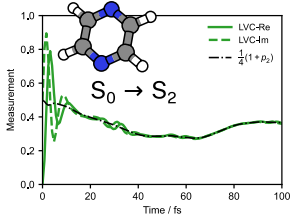

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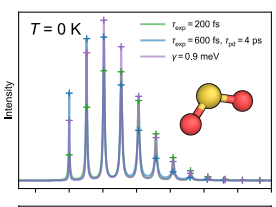
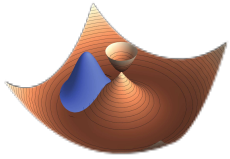
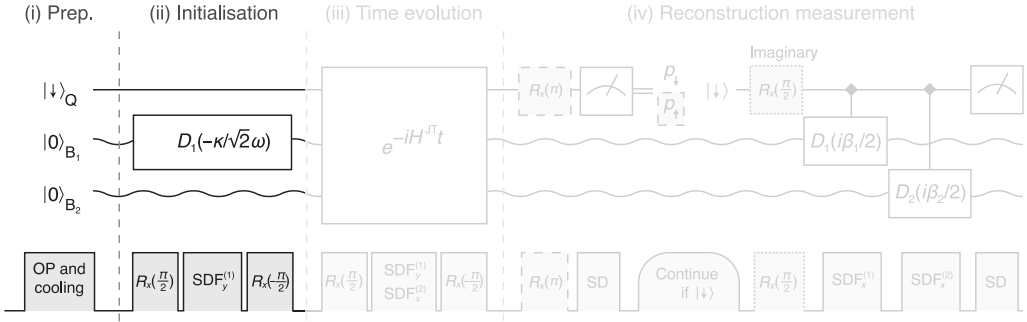
Open quantum systems



Holland et al. *Chem. Phys.* **1994**, *188*, 317–337.

Not spectroscopy: geometric phase





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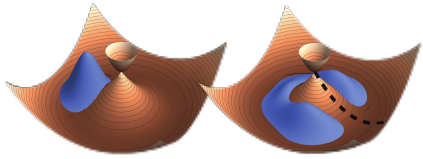

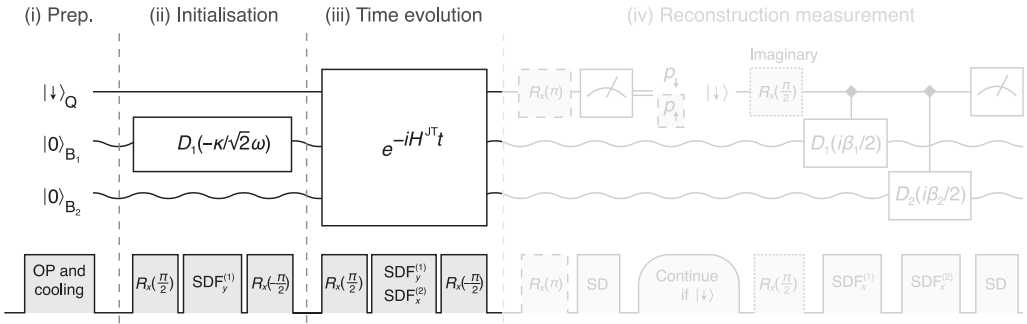

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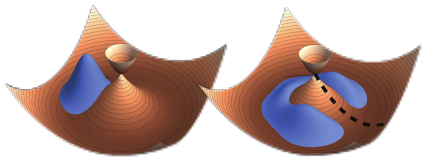
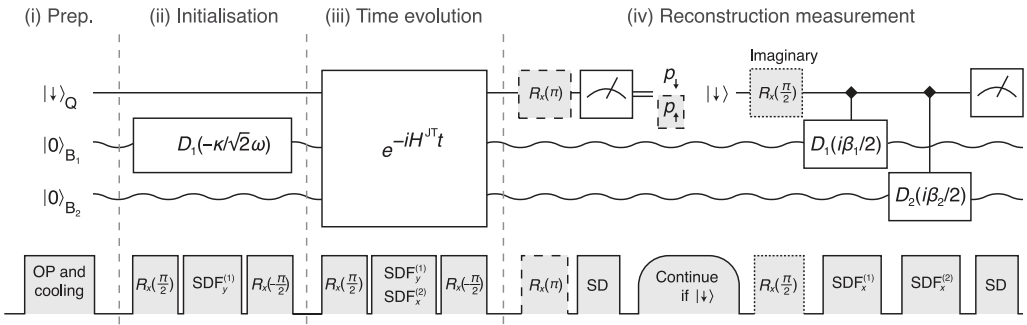
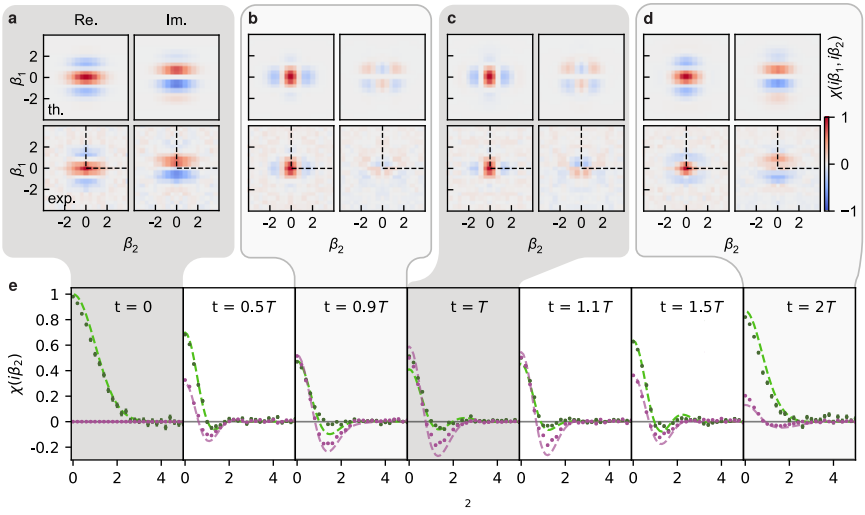


2D density reconstruction

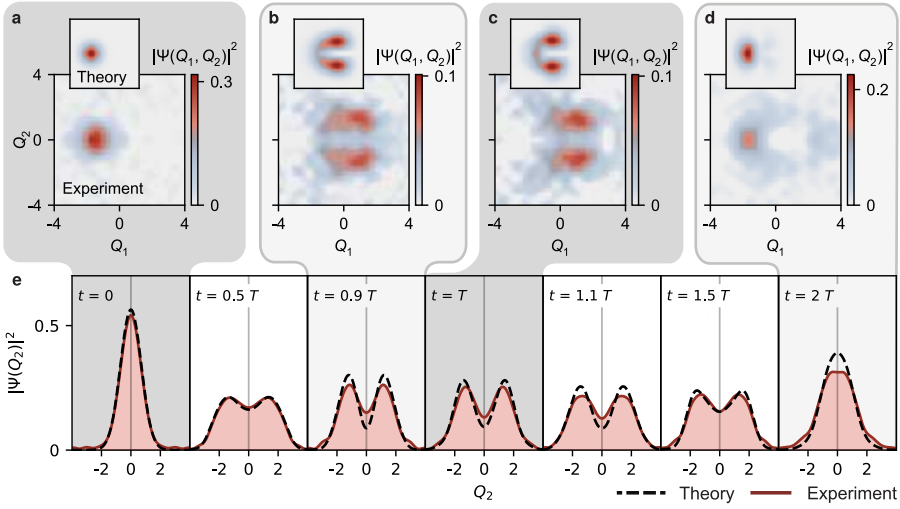
$$|\Psi(Q_1,Q_2)|^2 = \iint \frac{\mathrm{d}\beta_1 \mathrm{d}\beta_1}{2\pi^2} e^{-i\sqrt{2}(Q_1\beta_1+Q_2\beta_2)} \langle\Psi|D_1(i\beta_1)D_2(i\beta_2)|\Psi\rangle$$



Experimental measurements

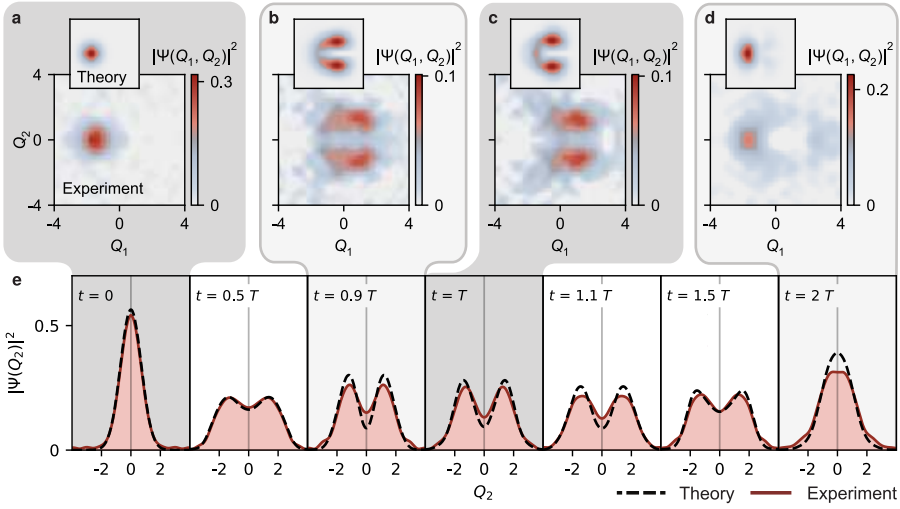


Reconstructed densities



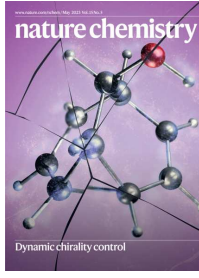
arXiv:2211.07320

Reconstructed densities



arXiv:2211.07320

Coming soon:



Conclusions

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- We can simulate vibronic dynamics with analog quantum devices
- We showed how absorption spectra can be obtained by measuring the auto-correlation function
- Our technique readily simulates vibronic coupling and open quantum systems
- The future of quantum simulation is **now**

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Read the preprint: arXiv:2209.06558

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MacDonell Group

quantum photochemistry

Grad student positions available!

email: rymac@dal.ca