Out-of-Time-Order Correlators based Spectroscopy for Nuclear Spin Geometries

A Quantum Simulation Approach to Hamiltonian Learning via Many-Body Echo Spectroscopy

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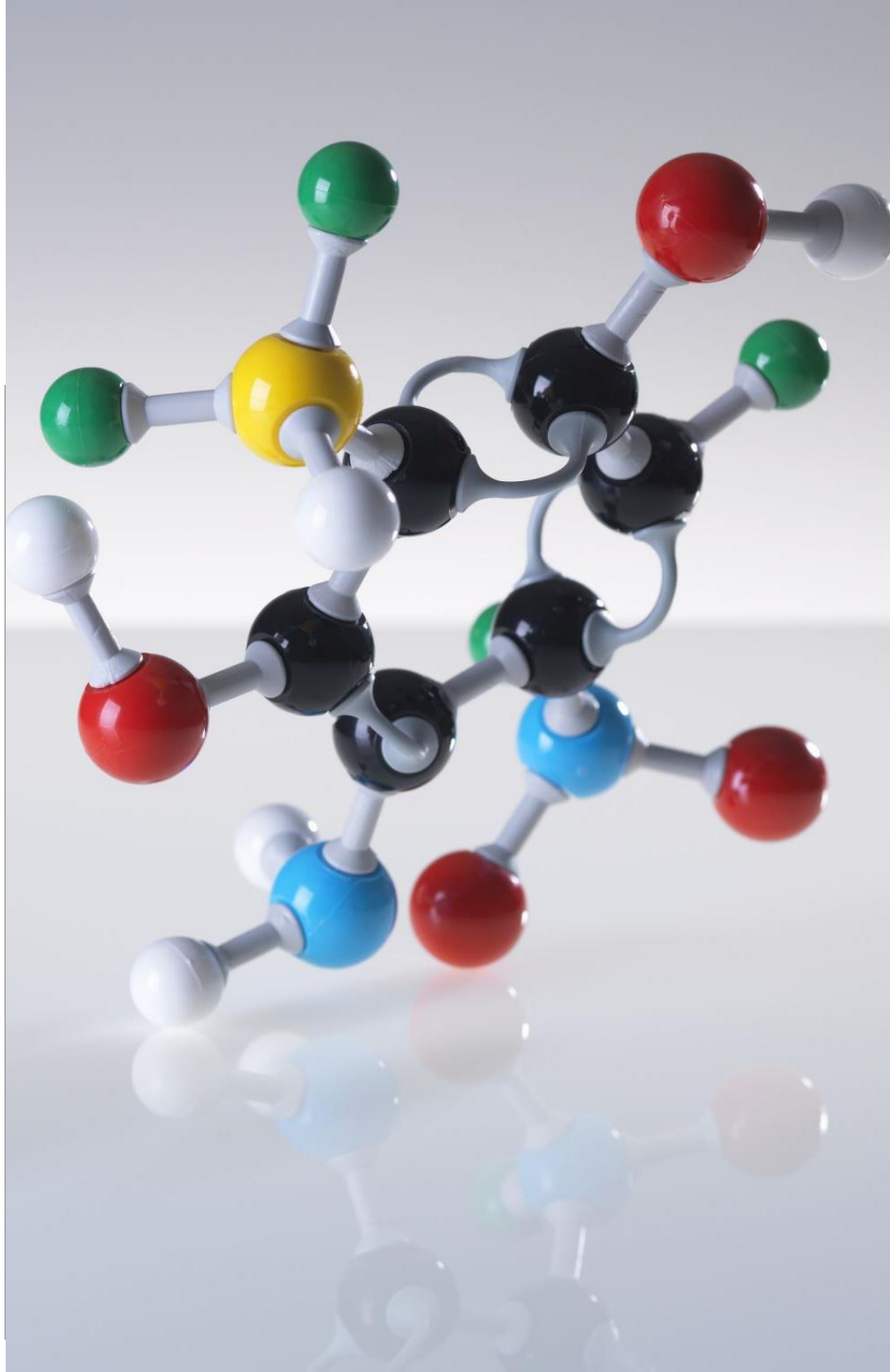
Motivation: The Challenge of Spin Networks in NMR

- **Problem in NMR/Molecular Physics:** Extracting precise long-range molecular distances (r_{ij}) and couplings (D_{ij}) in many-spin systems.
- **Decoherence:** Traditional signals (Time-Ordered Correlators, TOCs) decay exponentially due to **irreversible T_2 relaxation** and **information scrambling**.
- **Goal:** Utilize **time-reversal protocols** to selectively reverse reversible dynamics and isolate structural information encoded in the spin network Hamiltonian.

The Solution: Quantum Echoes

- **Loschmidt Echo (LE):** Measures the fidelity of time reversal.
- **Out-of-Time-Ordered Correlator (OTOC):** An enhanced echo that probes how a local perturbation spreads (scrambles) across the system.



Physical Model: The Secular Dipolar Hamiltonian

- **System:** N proton spins ($\mathbf{I}_i$) in a partially aligned medium.
- **Hamiltonian (Evolution Operator $U(t)$):**

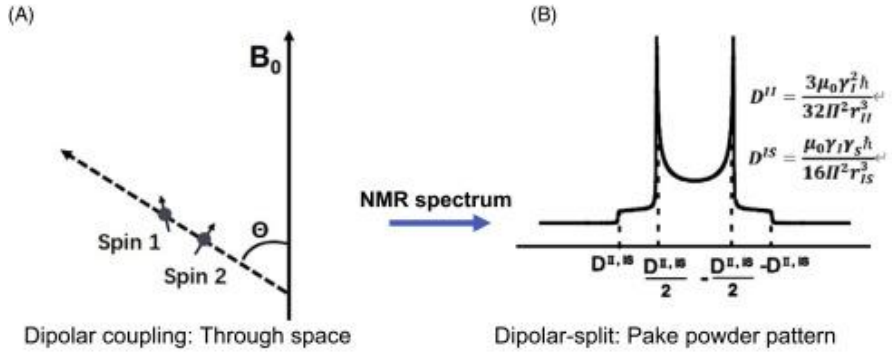
$$H_{\text{dip}} = \sum_{i<j} D_{ij} \left(2I_{z,i}I_{z,j} - I_{x,i}I_{x,j} - I_{y,i}I_{y,j}\right)$$
$$= \sum_{i<j} D_{ij}(2Z_iZ_j - \mathbf{I}_i \cdot \mathbf{I}_j) \quad \text{(up to constants)}$$

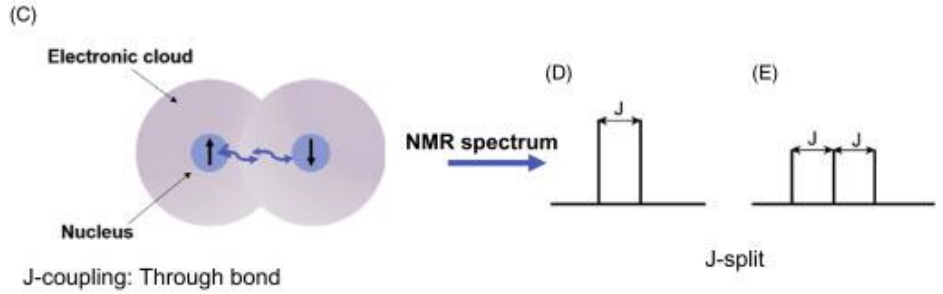
- **Residual Dipolar Coupling (RDC) D_{ij}:** The key parameter linking quantum dynamics to molecular geometry.

$$D_{ij} \propto \frac{1}{r_{ij}^3} \cdot S \cdot (3\cos^2\theta_{ij} - 1)$$

  - r_{ij}: Inter-spin distance.
  - S: Order parameter (degree of alignment).

Goal: Hamiltonian Learning by Extracting $\{D_{ij}\}$ from the OTOC signal.





The Quantum Probe: Out of Time Ordered Correlator (OTOC)

- **Definition:** The OTOC is a four-operator correlation function that probes the growth of the commutator $[W(t), V]^2$.
- **OTOC Fidelity ($F(t)$):** The experimentally measured quantity, where V and W are local spin operators.

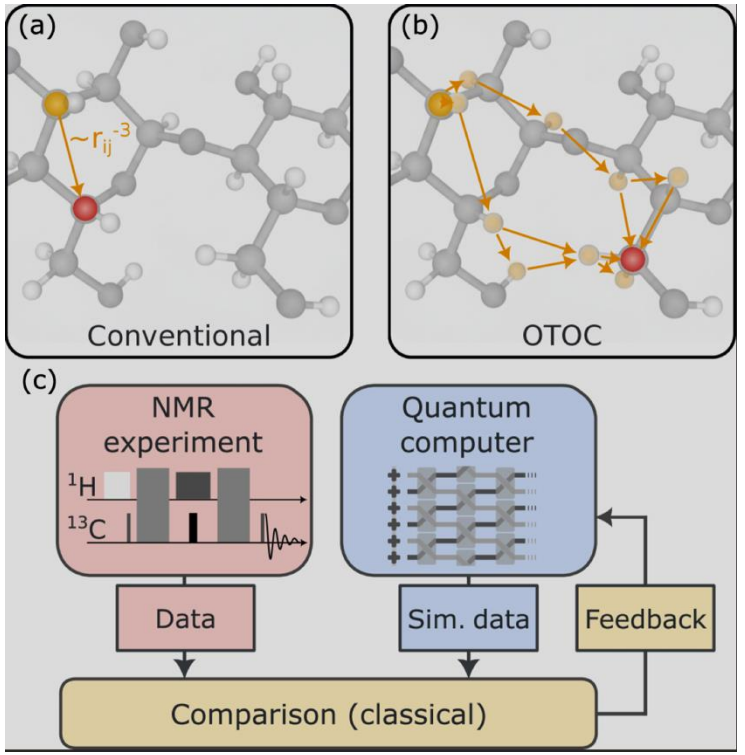
$$F(t) = \mathrm{Tr}\left[\rho_0 W^\dagger(t) V^\dagger W(t) V\right]$$

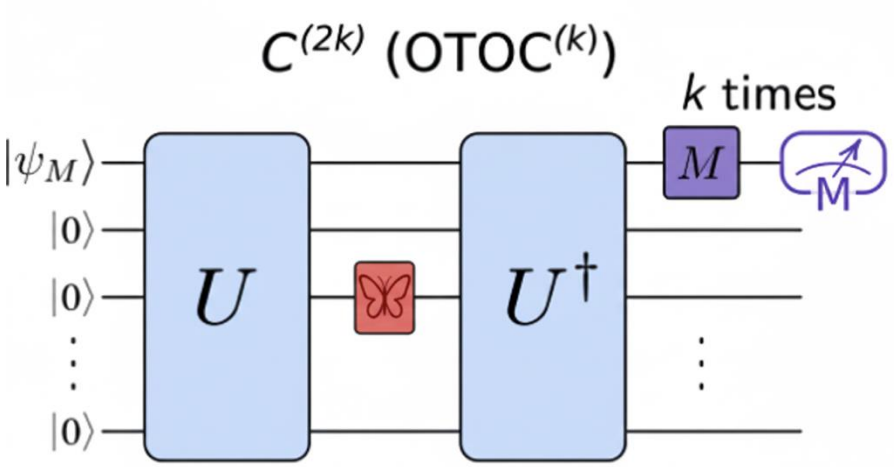
where $W(t) = U^\dagger(t) W U(t)$ and $U(t) = e^{-iHt}$.

- **Information Scrambling:** The decay of $F(t)$ from unity quantifies the spreading of operator V.

$$C(t) = 1 - \frac{1}{2}\,\mathrm{Tr}\left[\rho_0 [W(t), V]^2\right] \propto \mathrm{Re}(F(t))$$

- **OTOC Protocol (Quantum Echoes):**
  1. Prepare initial state ρ_0.
  2. Forward Evolution $U(t)$.
  3. Apply local perturbation V (Butterfly).
  4. Backward Evolution $U^\dagger(t)$.
  5. Measure local observable W.



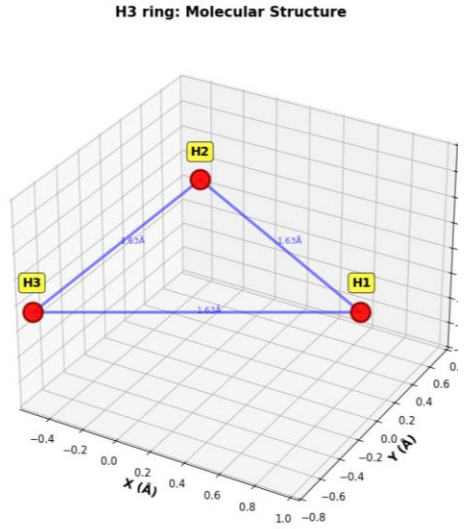


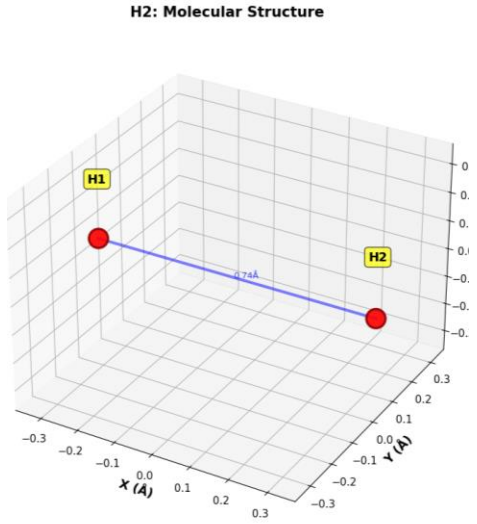
Simulation Samples (H2, H3, H6)

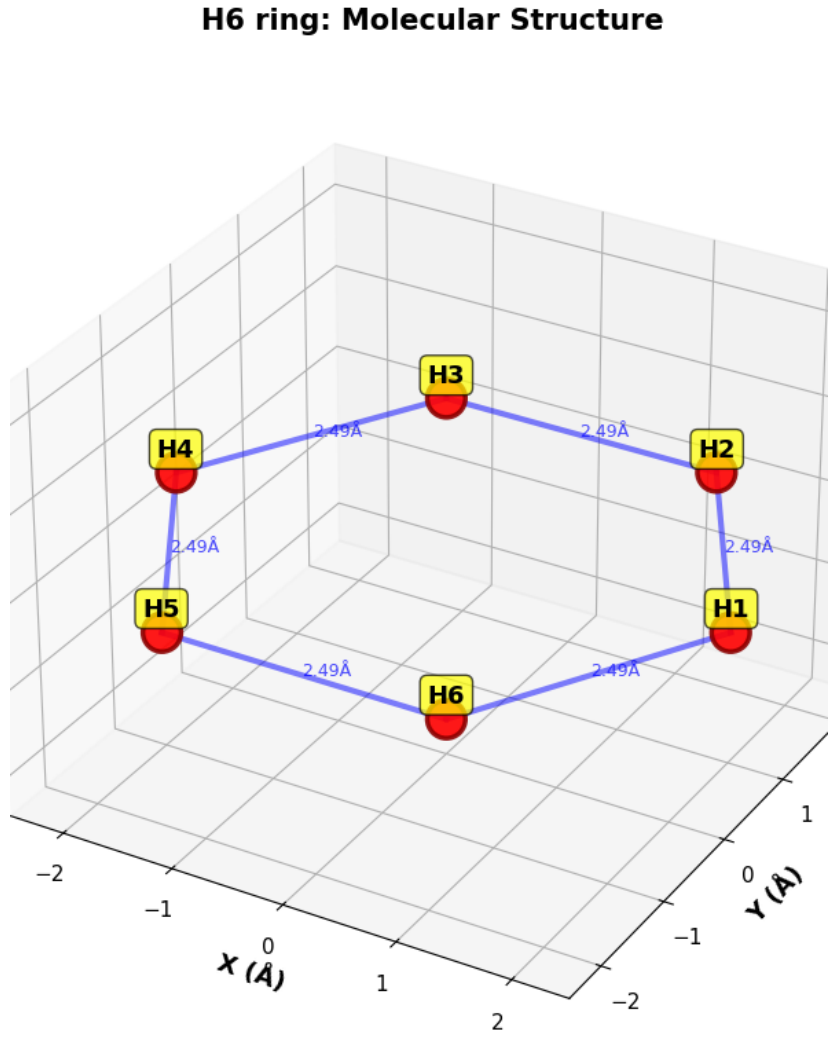
- Systems Simulated
- H2 Dimer (2 Qubits): Single coupling D01.
- H3 Ring (3 Qubits): Equilateral triangle topology.
- H6 Ring (6 Qubits): Benzene-like topology with complex, multiple
- Simulation Parameters
- H2 {n_qubits: 2, delta_nu: [0, 4000] Hz, J_{12}: 144 Hz, T_2: 0.50 s, dt: 20 µs, total_time: 0.50 s}
- H3 {n_qubits: 3,delta_nu: [0, 3000, 6000] Hz,J_{12}=144 Hz, J_{23}=7 Hz, J_{13}=0 Hz, T_2: 0.35 s, dt: 20 µs, total_time: 0.60 s}
- H6 {n_qubits: 6, delta_nu: [0,200,400,600,800,1000] Hz, J_ortho = 7.5 Hz, J_meta = 1.4 Hz, J_para = 0.6 Hz, T_2: 0.25 s, dt: 10 µs, total_time: 0.80 s}
- Result
- The OTOC signal exhibits a rapid, non-monotonic decay

whose features are directly determined by the inter-spin couplings,

confirming structural sensitivity







Quantum Implementation and Observable

- **Trotterization:** The many-body evolution $U(t)$ is approximated by a first-order product formula for quantum simulation.

$$U(t) = e^{-iHt} \approx \left(\prod_{i<j} e^{-iH_{ij}\Delta t} \right)^n$$

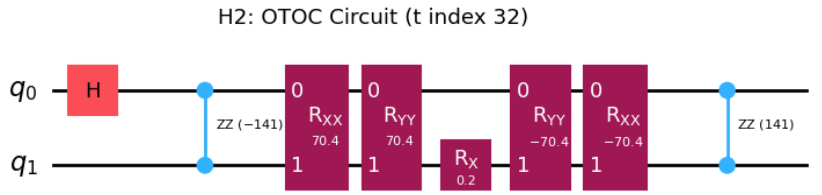
- **Experimental Signal $\langle X \rangle$:** The OTOC is measured as a modified expectation value on the measurement spin m:

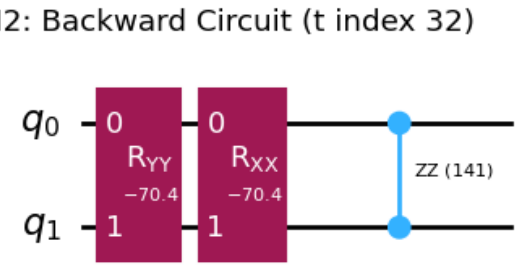
$$M_{\mathrm{OTOC}}(t) = \langle \psi_0 | U^\dagger(t) V^\dagger I_{x,m} U(t) V | \psi_0 \rangle$$

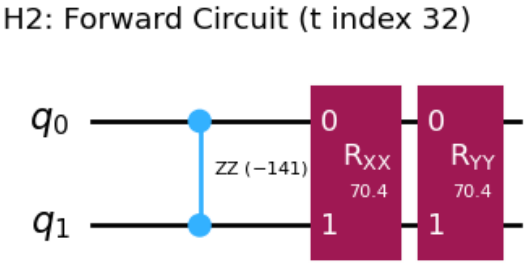
- **Loschmidt Echo (LE) Baseline:** Setting the butterfly operator $V = I$ provides a baseline decay.

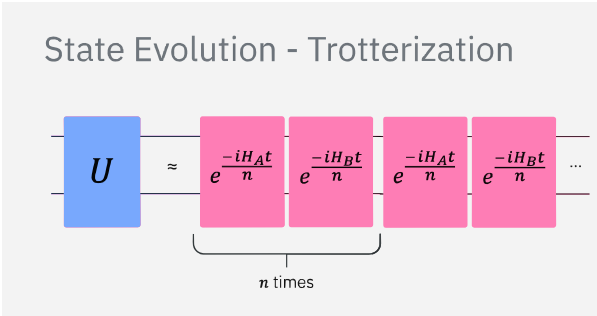
$$M_{\mathrm{LE}}(t) = \langle \psi_0 | U^\dagger(t) I_{x,m} U(t) | \psi_0 \rangle$$

- **Structural Sensitivity:** The **difference** between $M_{\mathrm{LE}}(t)$ and $M_{\mathrm{OTOC}}(t)$ isolates the structural information encoded by the coupling between the measured qubit (W) and the butterfly qubit (V).







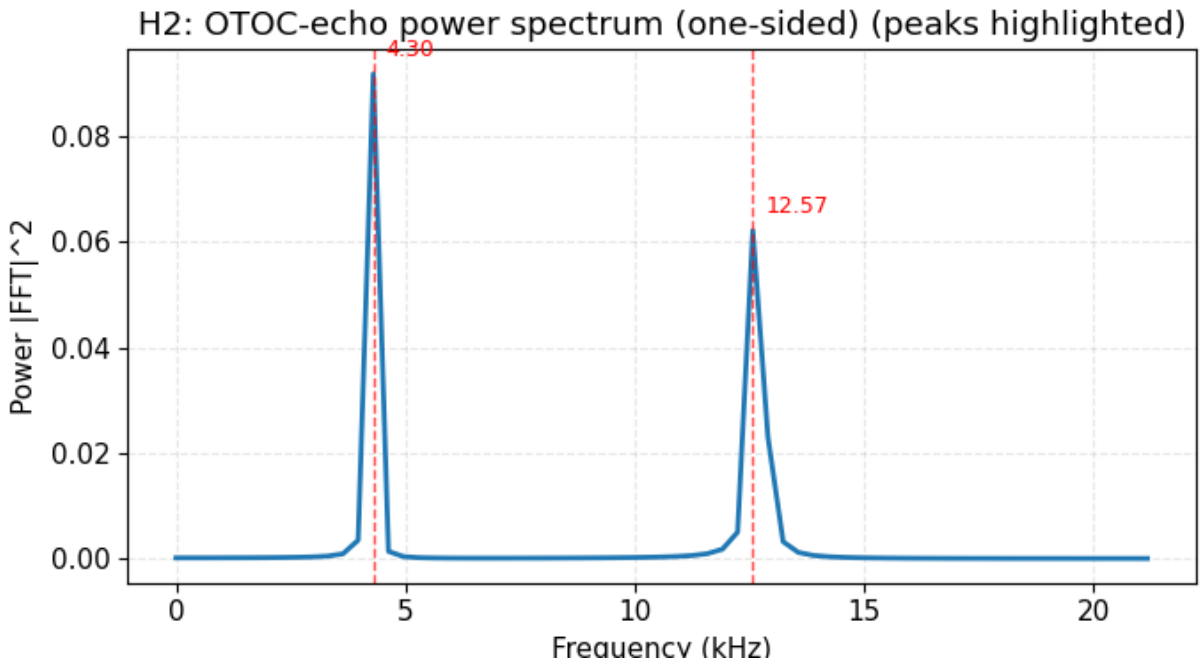


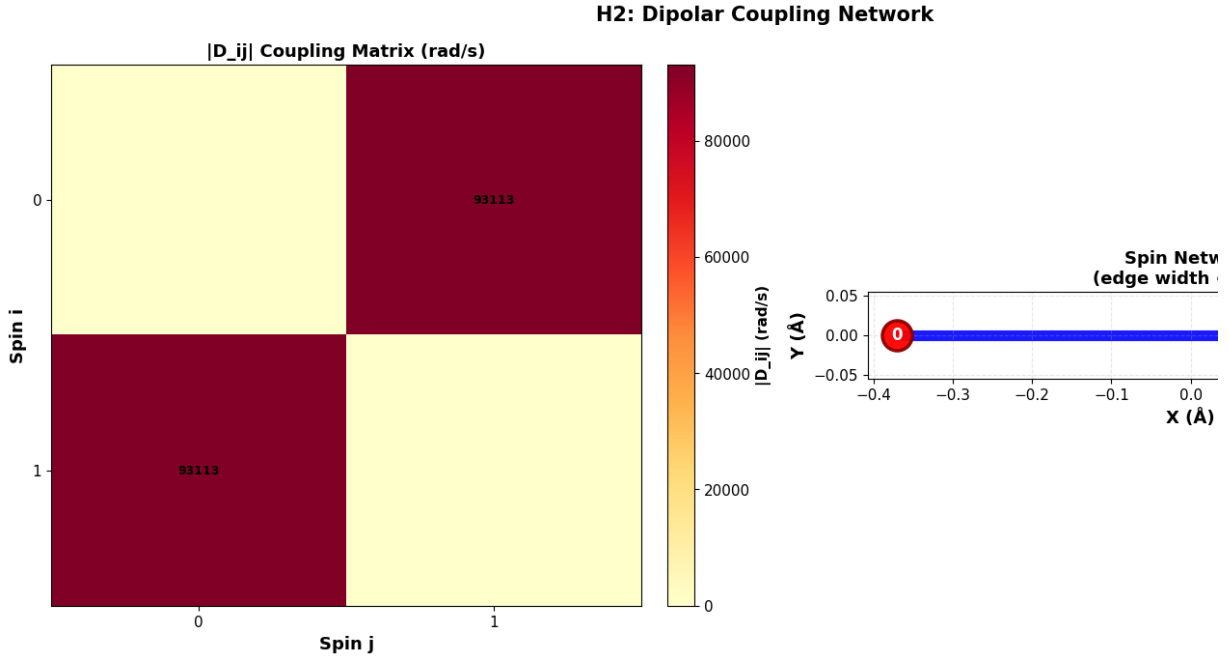
Data Analysis: Spectral Extraction of Couplings

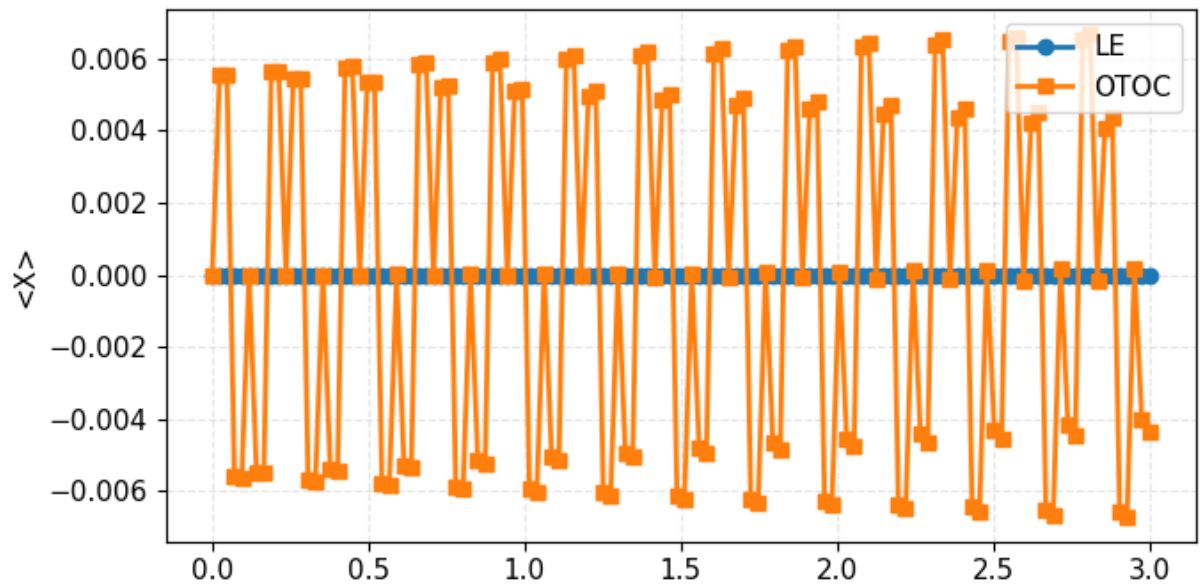
- **Principle:** The time evolution $U(t)$ is a superposition of frequencies corresponding to the eigenvalues of H, which are functions of the coupling strengths D_{ij}.
- **Method:** Apply a **Fourier Transform ($\mathcal{F}$)** to the OTOC time-trace $M_{\mathrm{OTOC}}(t)$.

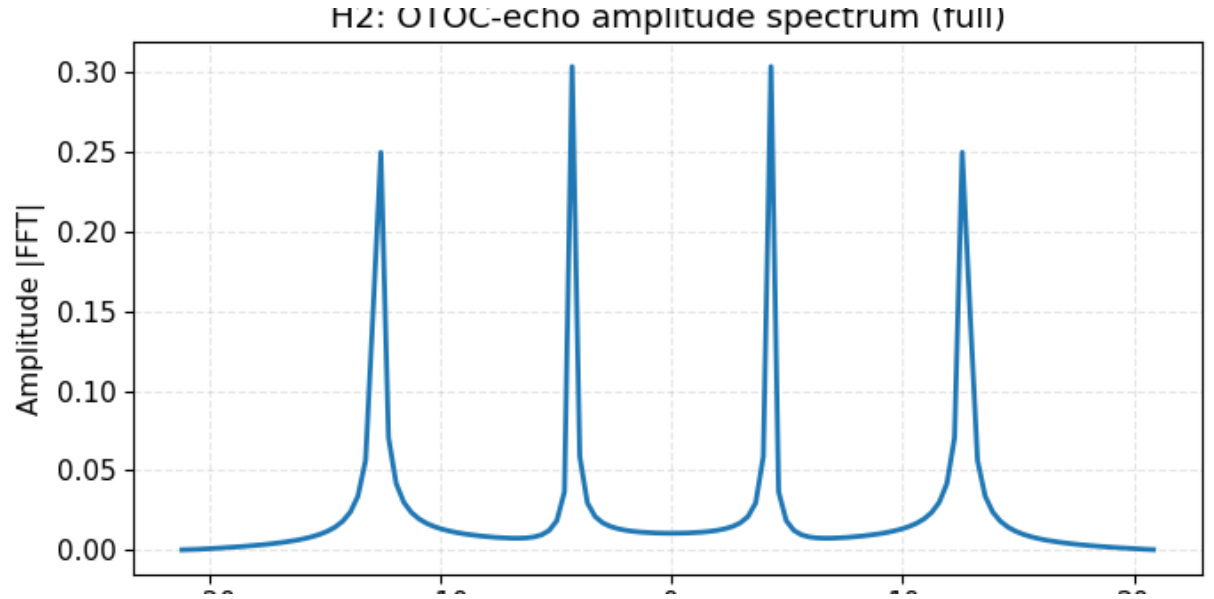
$$\text{Spectrum}(\omega) = |\mathcal{F}\{M_{\mathrm{OTOC}}(t)\}|^2$$

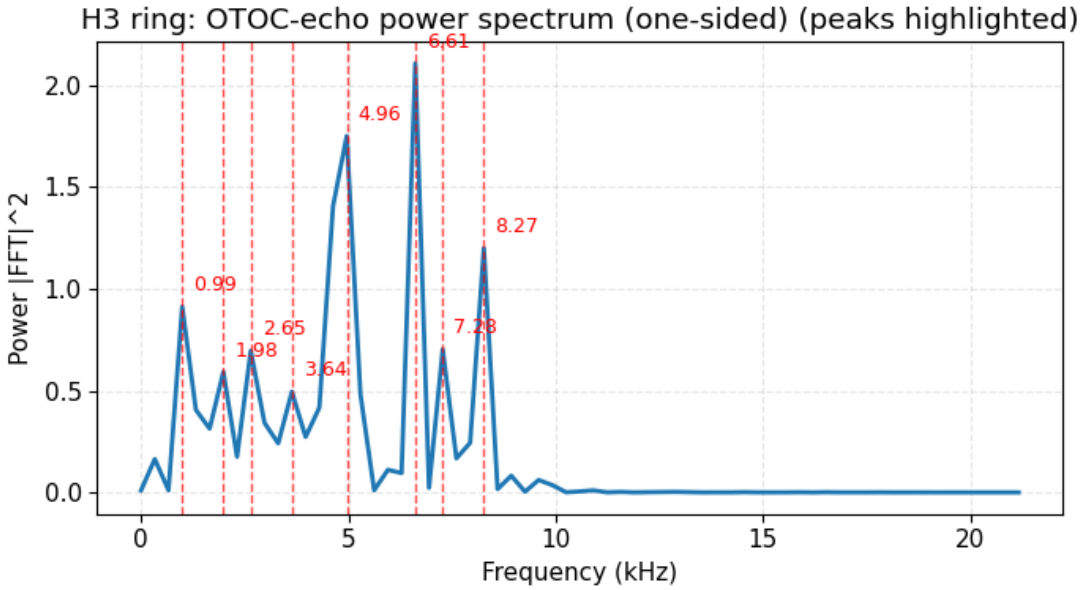
- **Result (Hamiltonian Learning):**
  1. The power spectrum yields distinct, sharp frequency peaks ω_k.
  2. These peaks ω_k correspond to specific linear combinations of the coupling strengths $\{D_{ij}\}$.
  3. By identifying these peaks, we **infer the D_{ij} values**, effectively performing quantum spectroscopy to determine the molecular geometry.

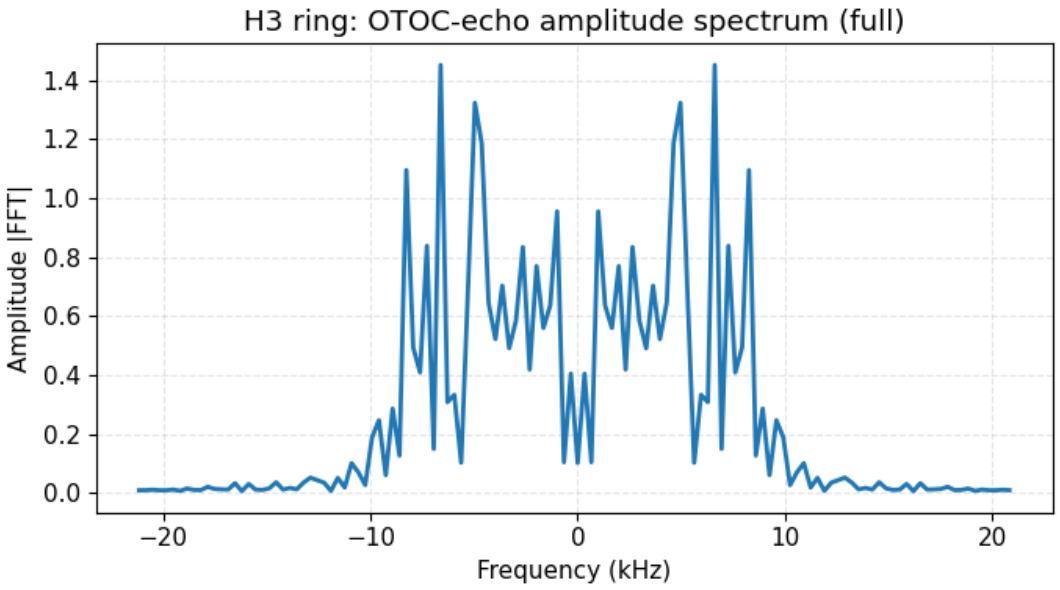

H2: OTOC-echo power spectrum (one-sided) (peaks highlighted)
4.30
12.57
Power |FFT|^2
Frequency (kHz)

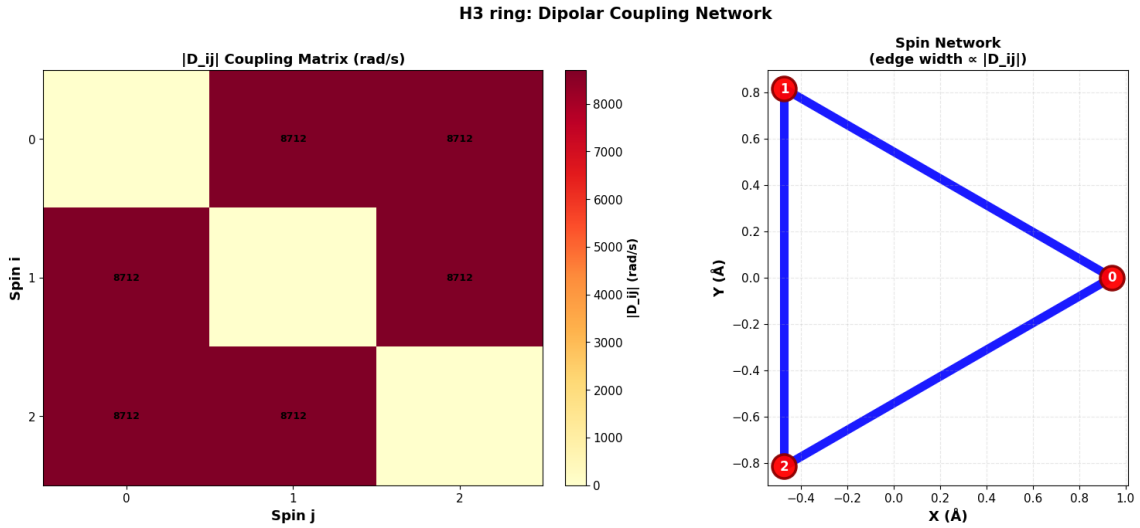

H2: Dipolar Coupling Network
|D_ij| Coupling Matrix (rad/s)
93113
93113
Spin i
Spin j
|D_ij| (rad/s)
Spin Netw
(edge width
Y (Å)
X (Å)


LE
OTOC
<X>


H2: OTOC-echo amplitude spectrum (full)
Amplitude |FFT|


H3 ring: OTOC-echo power spectrum (one-sided) (peaks highlighted)
Power |FFT|^2
Frequency (kHz)
0.99
1.98
2.65
3.64
4.96
6.61
7.28
8.27


H3 ring: OTOC-echo amplitude spectrum (full)
Amplitude |FFT|
Frequency (kHz)


H3 ring: Dipolar Coupling Network
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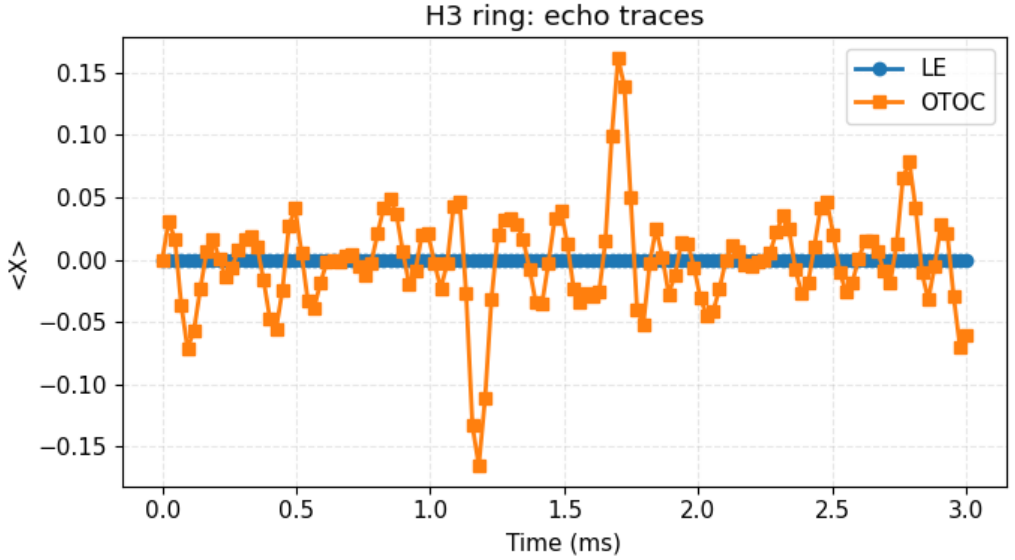
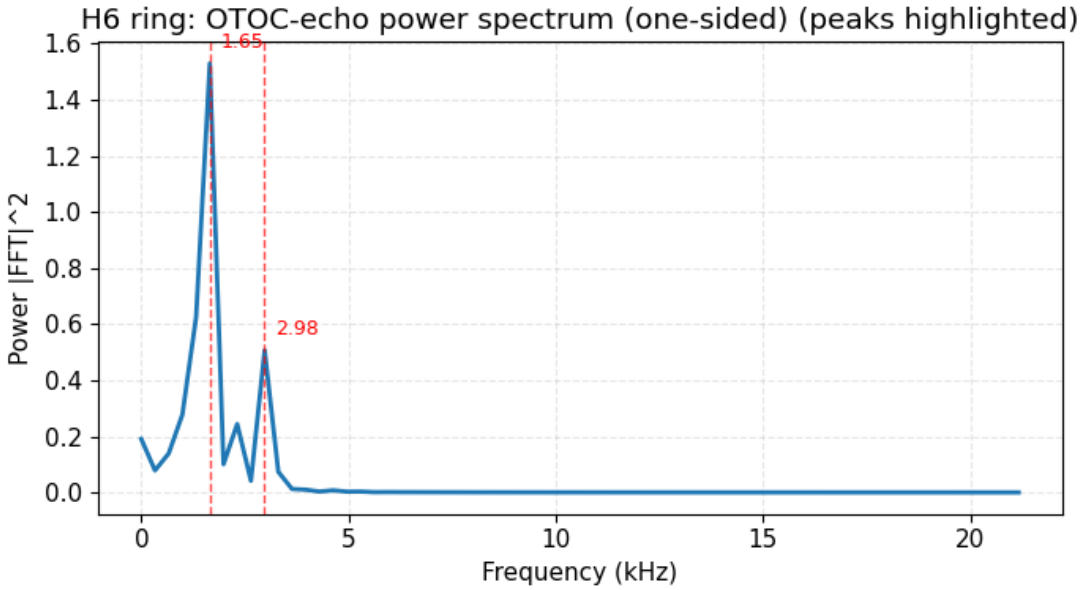
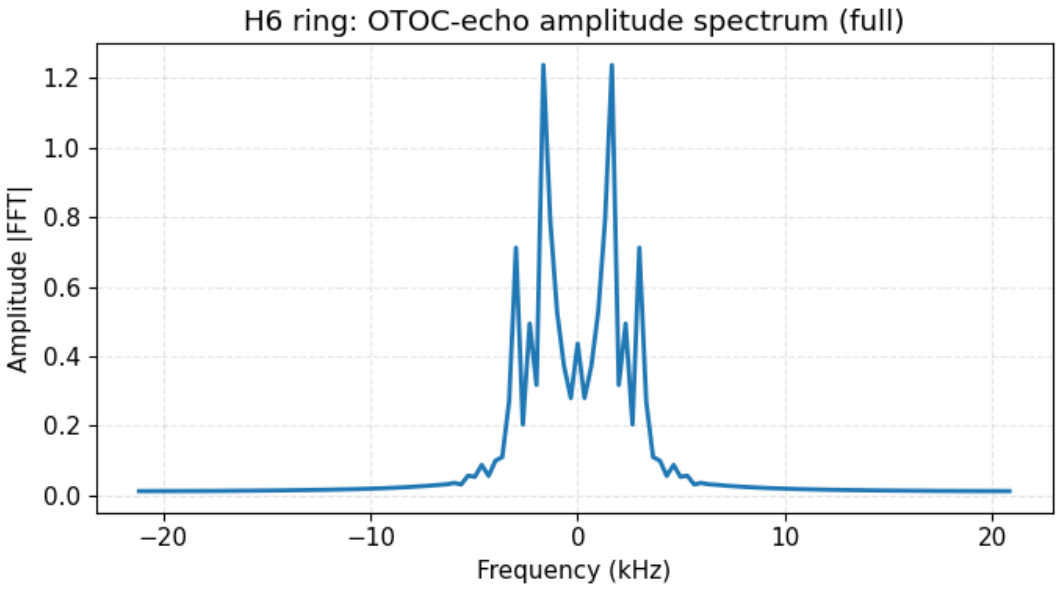
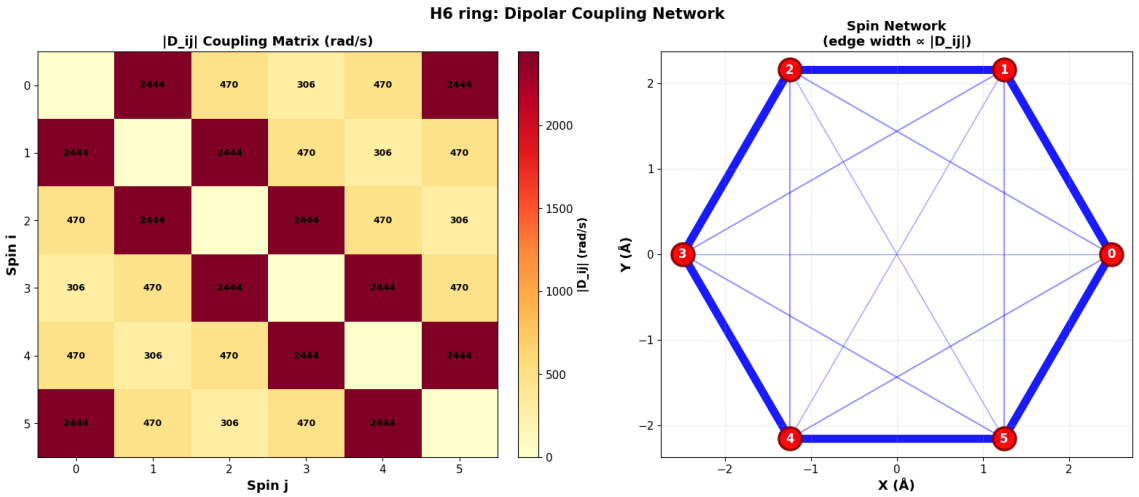
Spin i
Spin j
|D_ij| (rad/s)
Spin Network
(edge width ∝ |D_ij|)
Y (Å)
X (Å)


H3 ring: echo traces
LE
OTOC
<X>
Time (ms)


H6 ring: OTOC-echo power spectrum (one-sided) (peaks highlighted)
1.65
2.98
Power |FFT|^2
Frequency (kHz)


H6 ring: OTOC-echo amplitude spectrum (full)
Amplitude |FFT|
Frequency (kHz)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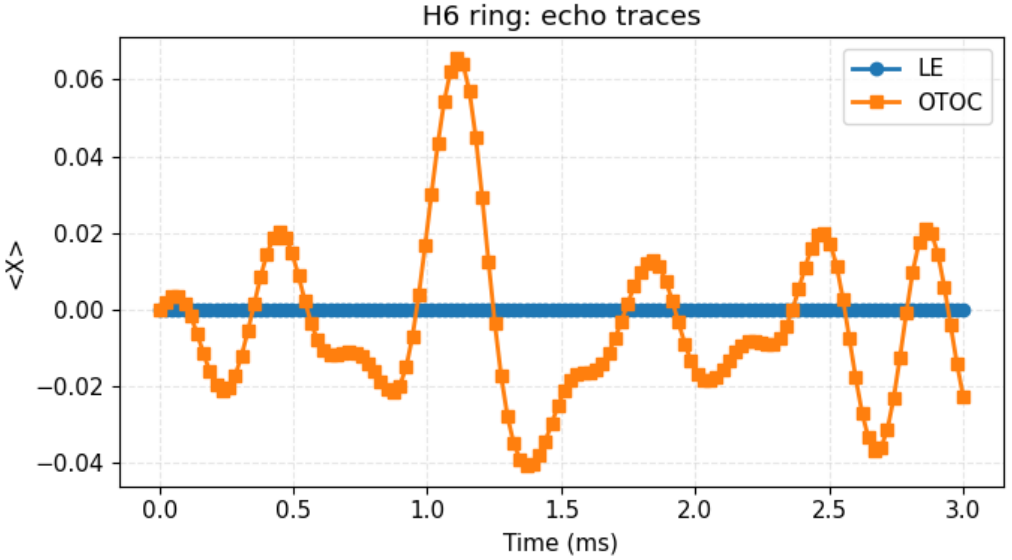
Spin j
|D_ij| (rad/s)
Spin Network
(edge width ∝ |D_ij|)
Y (Å)
X (Å)


H6 ring: echo traces
LE
OTOC
<X>
Time (ms)

The Quantum Advantage, A Molecular Ruler

- **Robustness:** The echo sequence inherently cancels systematic errors (e.g., imperfect pulses) and is highly **robust to static noise**.
- **Amplification:** Unlike traditional signals (TOCs) which decay rapidly, the OTOC's time-reversal allows the structural signal to be **amplified and read out clean**.
- **Speedup:** The "Quantum Echoes" algorithm has been demonstrated to perform Hamiltonian learning up to **13,000 times faster** than the best classical supercomputing methods for this specific task.
- **Sensing Scrambling:** The OTOC signal measures the **butterfly velocity** (v_B) of quantum information spread, providing a fundamental structural constant.

Conclusion and Future Direction

- **Conclusion:**
  1. The OTOC protocol successfully models a novel spectroscopic method for **Hamiltonian learning** in interacting spin systems.
  2. Time-domain simulation and frequency-domain analysis validate that OTOC can **spectrally resolve the molecular dipolar couplings** (D_{ij}).
  3. This approach leverages quantum mechanics to overcome the exponential complexity of many-body dynamics.
- **Future Work:**
  1. **Hardware Implementation:** Run the simulated Trotter circuits on current NISQ (Noisy Intermediate-Scale Quantum) devices.
  2. **Error Mitigation:** Integrate advanced error mitigation and post-processing techniques.
  3. **Inverse Modeling:** Develop computational schemes to solve the inverse problem: mapping the observed spectral peaks ω_k back to the full set of inter-atomic distances $\{r_{ij}\}$.

References

- GitHub Repository
- Observation of constructive interference at the edge of quantum ergodicity
- Quantum computation of molecular geometry via many-body nuclear spin echoes