

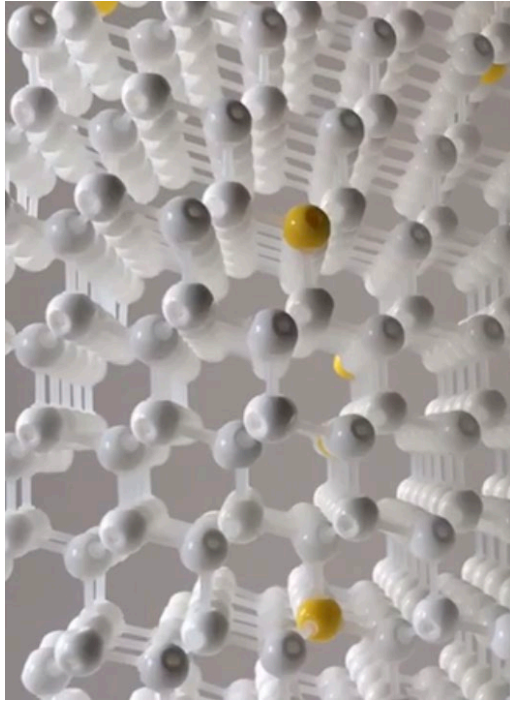
Time crystals with spins in diamond



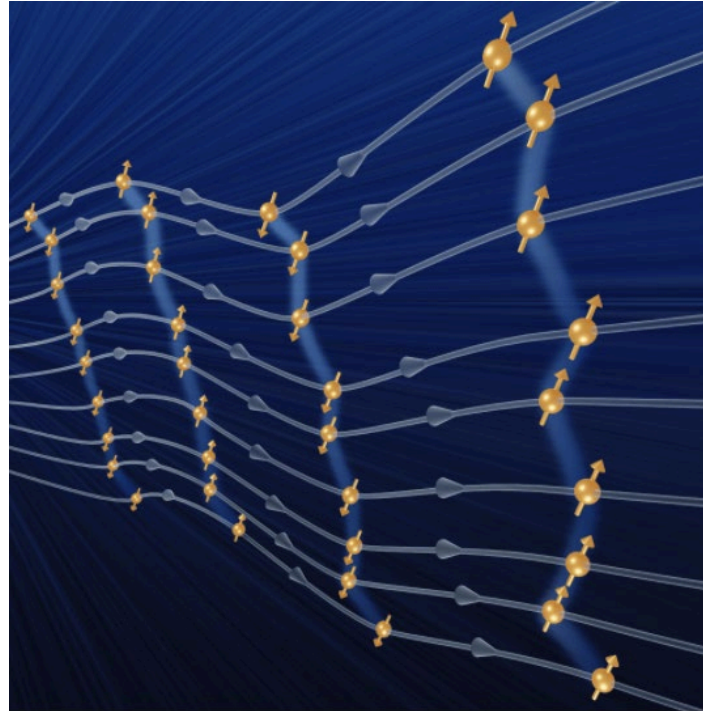
Tim Hugo Taminiau

Time crystals?

Concept proposed in 2012 by Frank Wilczek

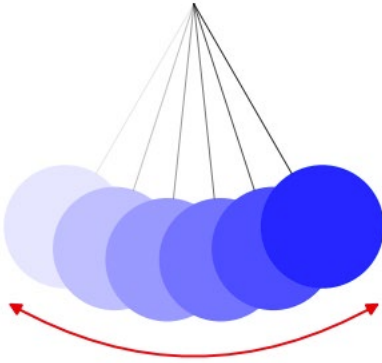


(Spatial) Crystal
– A diamond –

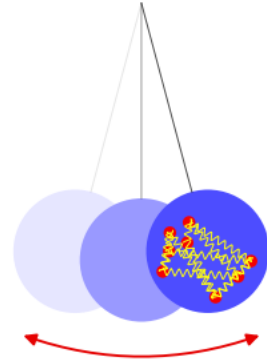


Discrete time Crystal
– Artist's impression –

Key element: a closed many-body system



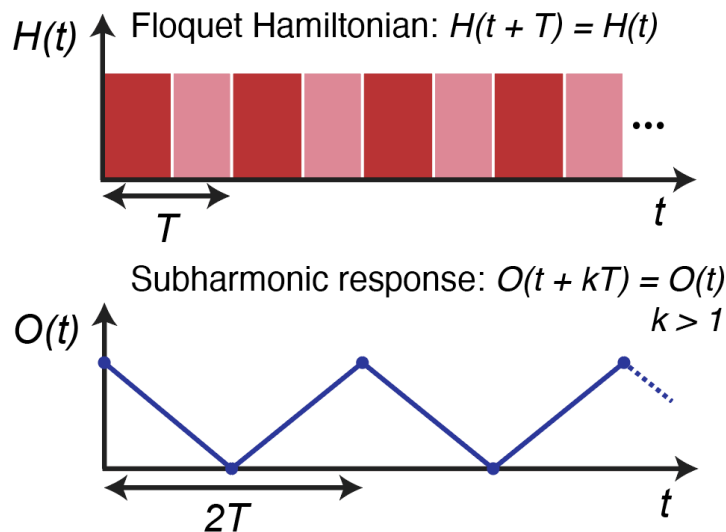
Perfect “single-mode” pendulum



Perfect many-body pendulum

The Discrete Time Crystal (DTC) phase

- **Periodically driven** Floquet Hamiltonian
- **Closed interacting many-body** quantum system
- **DTC** breaks *discrete* time translation symmetry: subharmonic response.
- **Out-of-equilibrium phase of matter**: robust to perturbations of the parameters.
- **Stable**: Prevent heating. Breaking ergodicity through many body localization (**MBL**).
- **Eigenstate order**: Spatio-temporal order from **all initial states**.



See:

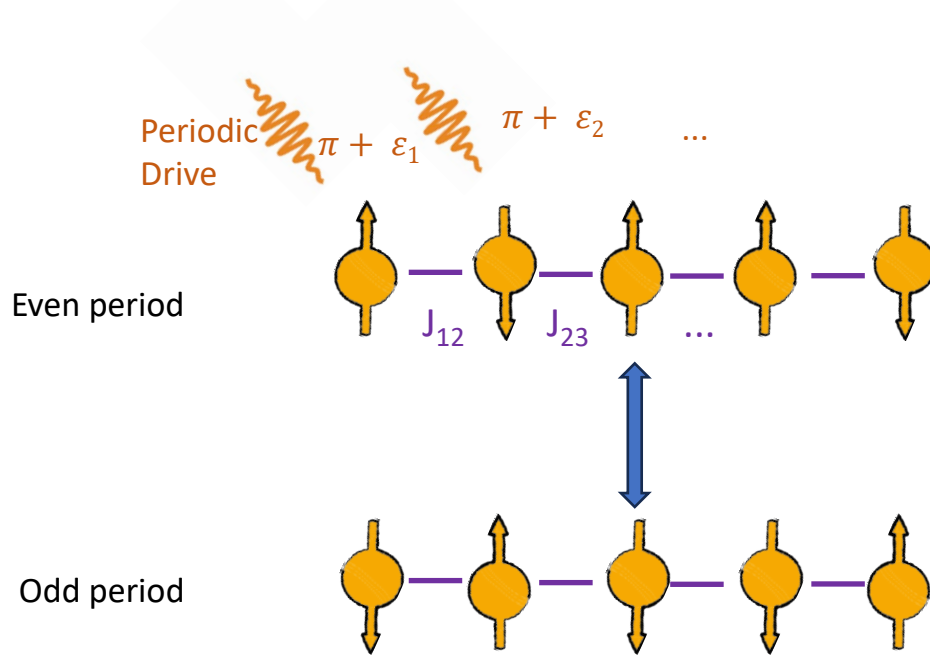
M. Ippoliti et al., PRX Quantum 2, 030346 (2021)

V. Khemani, arXiv:1910.10745 (2019)

D. V. Else, Annu. Rev. Condens. Matter Phys. 11, 467(2020)

Zaletel et al., Rev. Mod. Phys. 95, 031001 (2023)

A concrete example: a 1D chain of spins

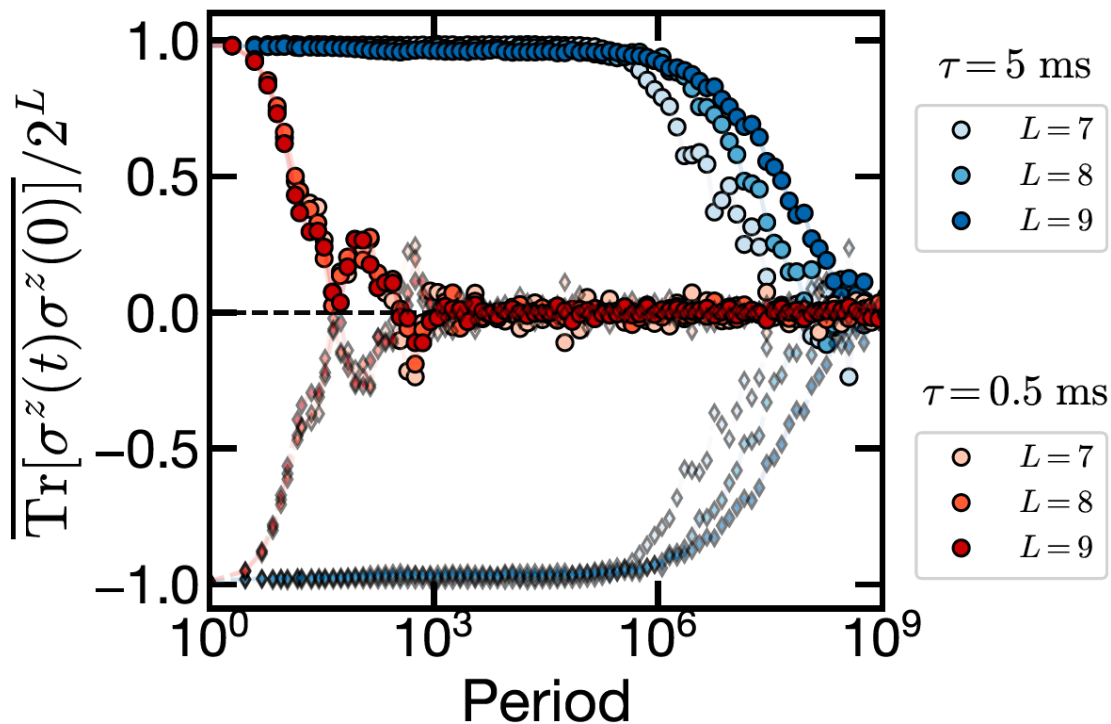


$$H_{\text{int}} = \sum_j^L (B + h_j) \sigma_j^z$$

- Imperfect drive, closed system
- Disordered couplings
- Low dimensional system and short range interactions required (debate on exact requirements).
- Exponential growth of stability by **only increasing the size** of the chain for **any initial state**

Note: these are open theoretical questions!

Numerical example of a length L spin chain



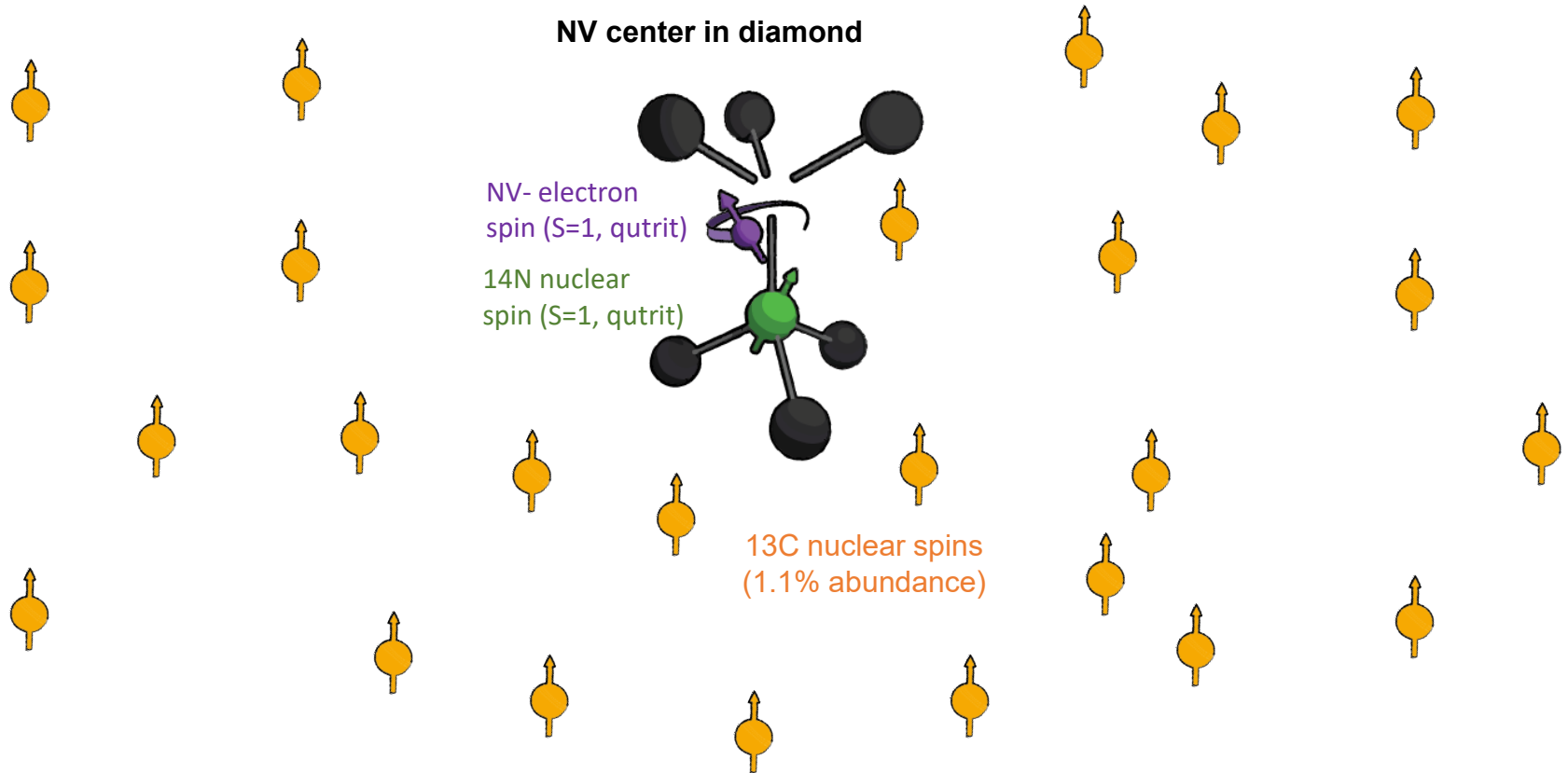
Theory shows long-lived response with exponential growth in system size

Consistent with the concept of DTC phase



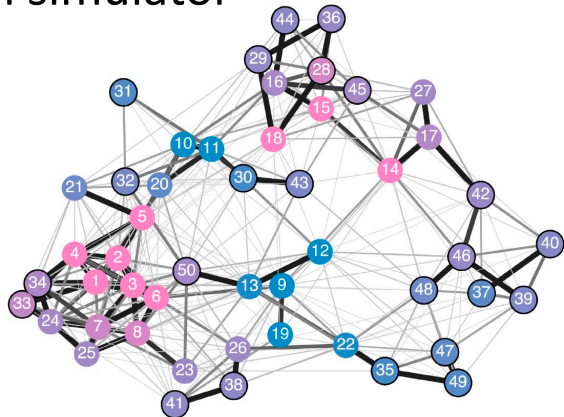
Simulation by Francisco Machado

System: ^{13}C nuclear spins + NV center in diamond

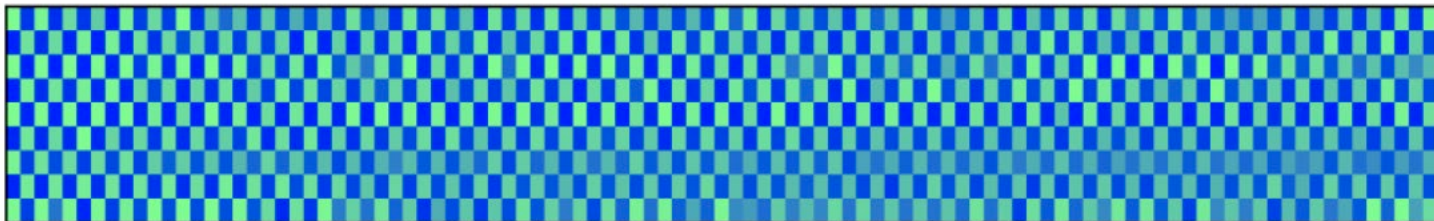


Outline

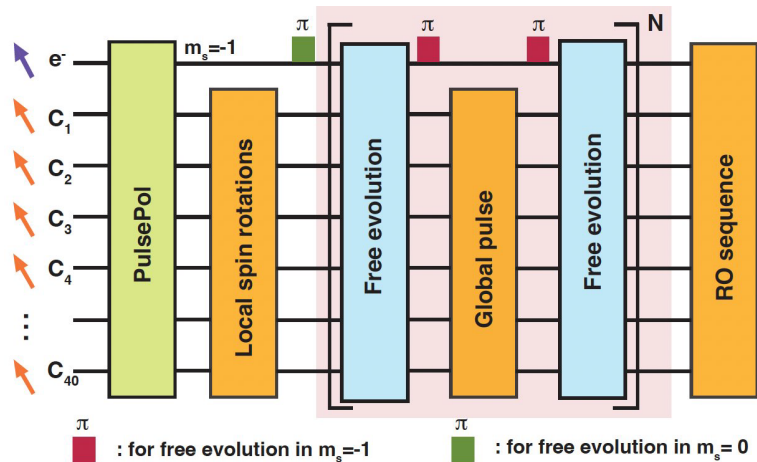
(1) System: a programmable diamond quantum simulator



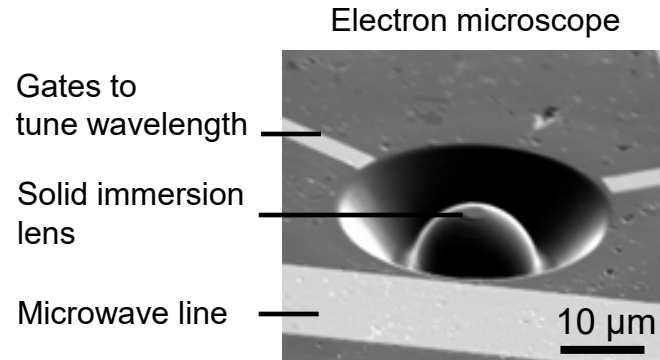
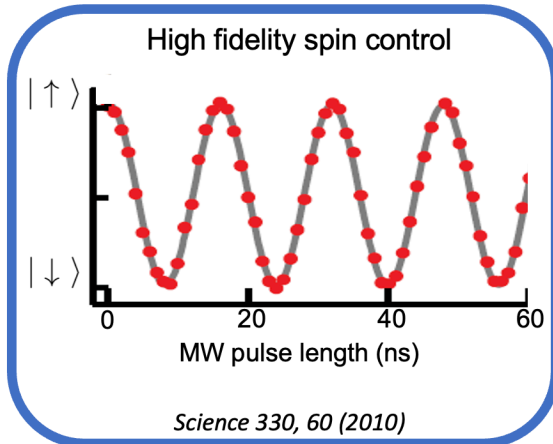
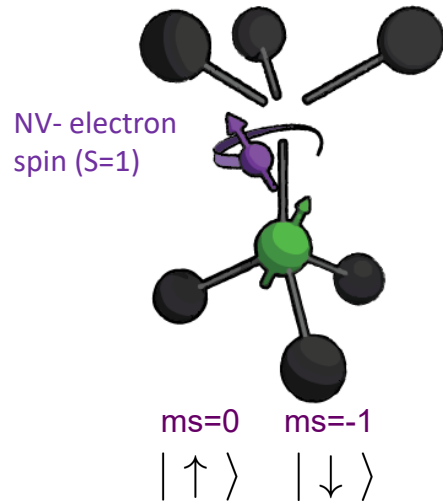
(2) 1D: Discrete time crystals



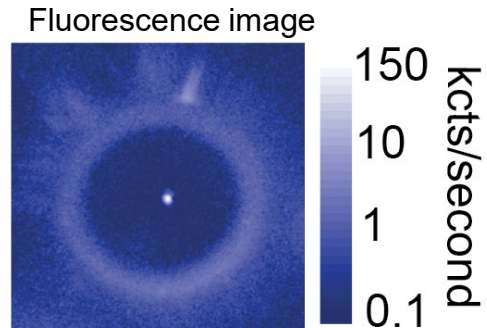
(3) Going 3D: ‘Critical’ Time Crystals



Our diamond devices

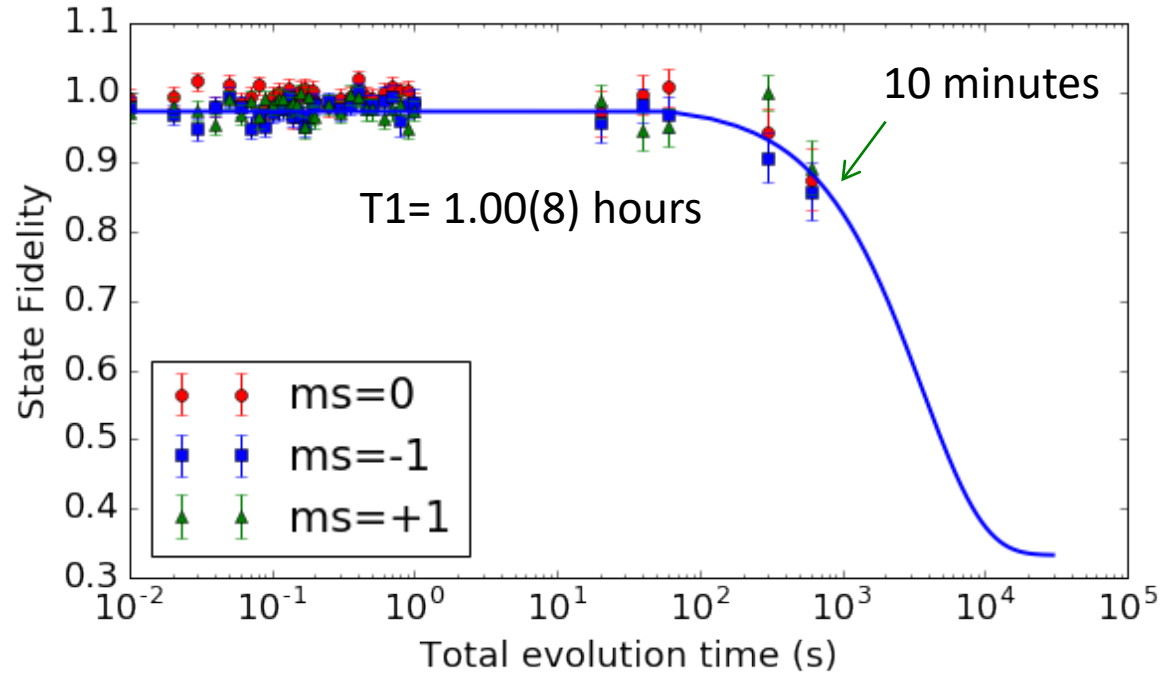


Temperature = 4 K



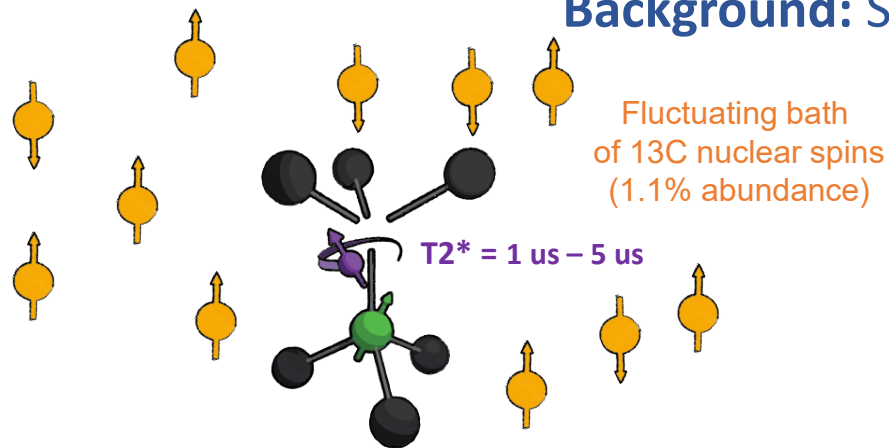
Pioneering work by Stuttgart,
Harvard, Chicago, Ulm, HP and many others

Electron spin relaxation (T1): a highly isolated system

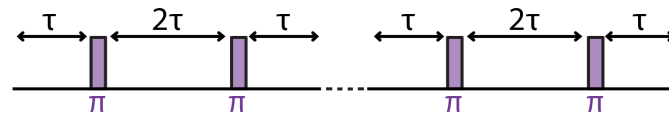


”Clean” system: ground state physics
is purely spin–spin interactions.

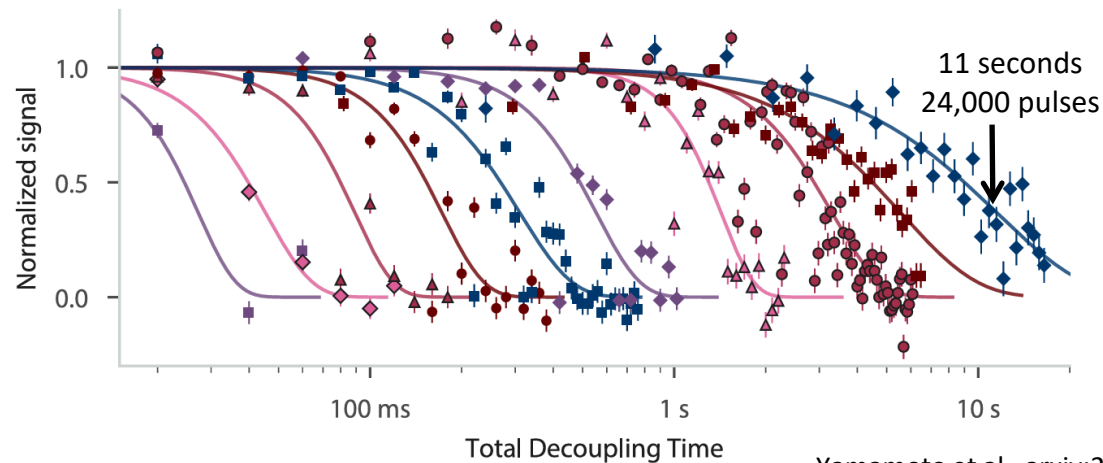
Background: Spin coherence



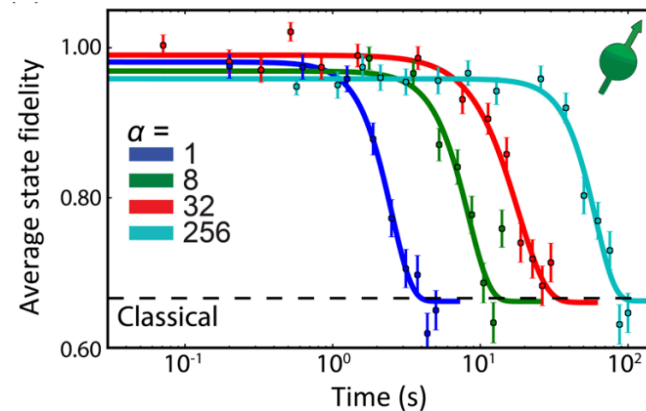
Decoupling pulse sequence (e.g. CPMG, XY8)



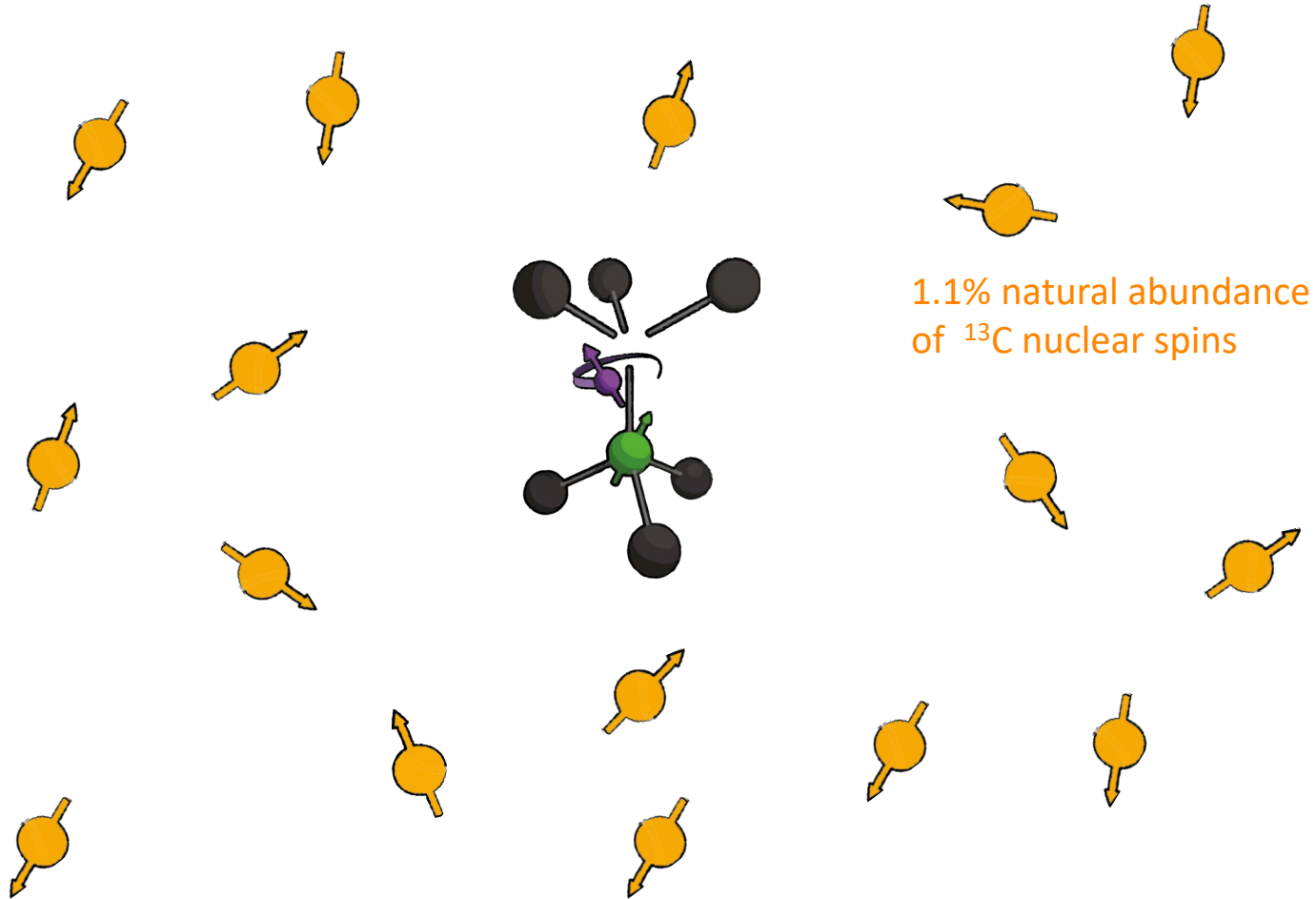
Electron spin dynamical decoupling (T_2)



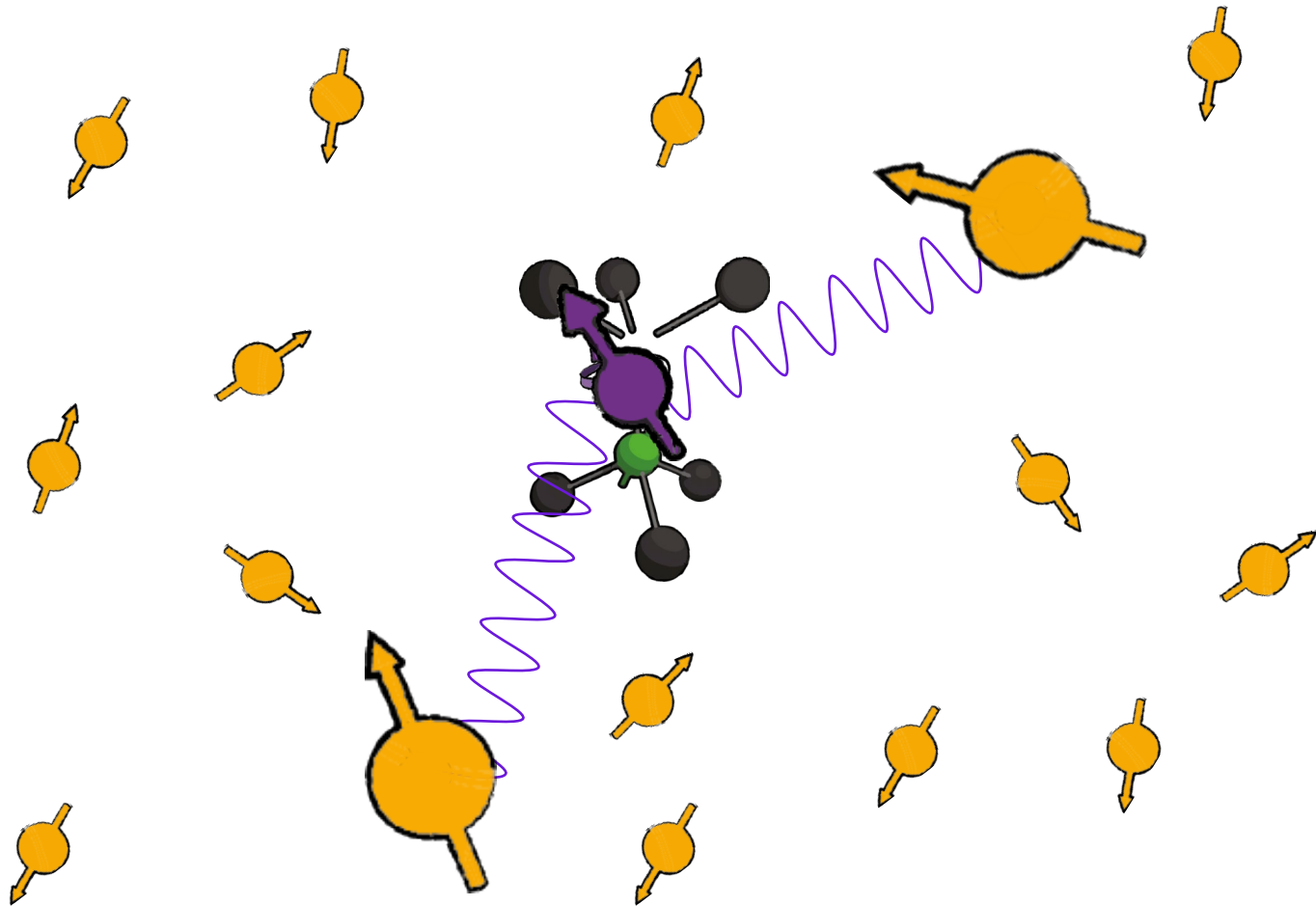
Minutes for nuclear spins



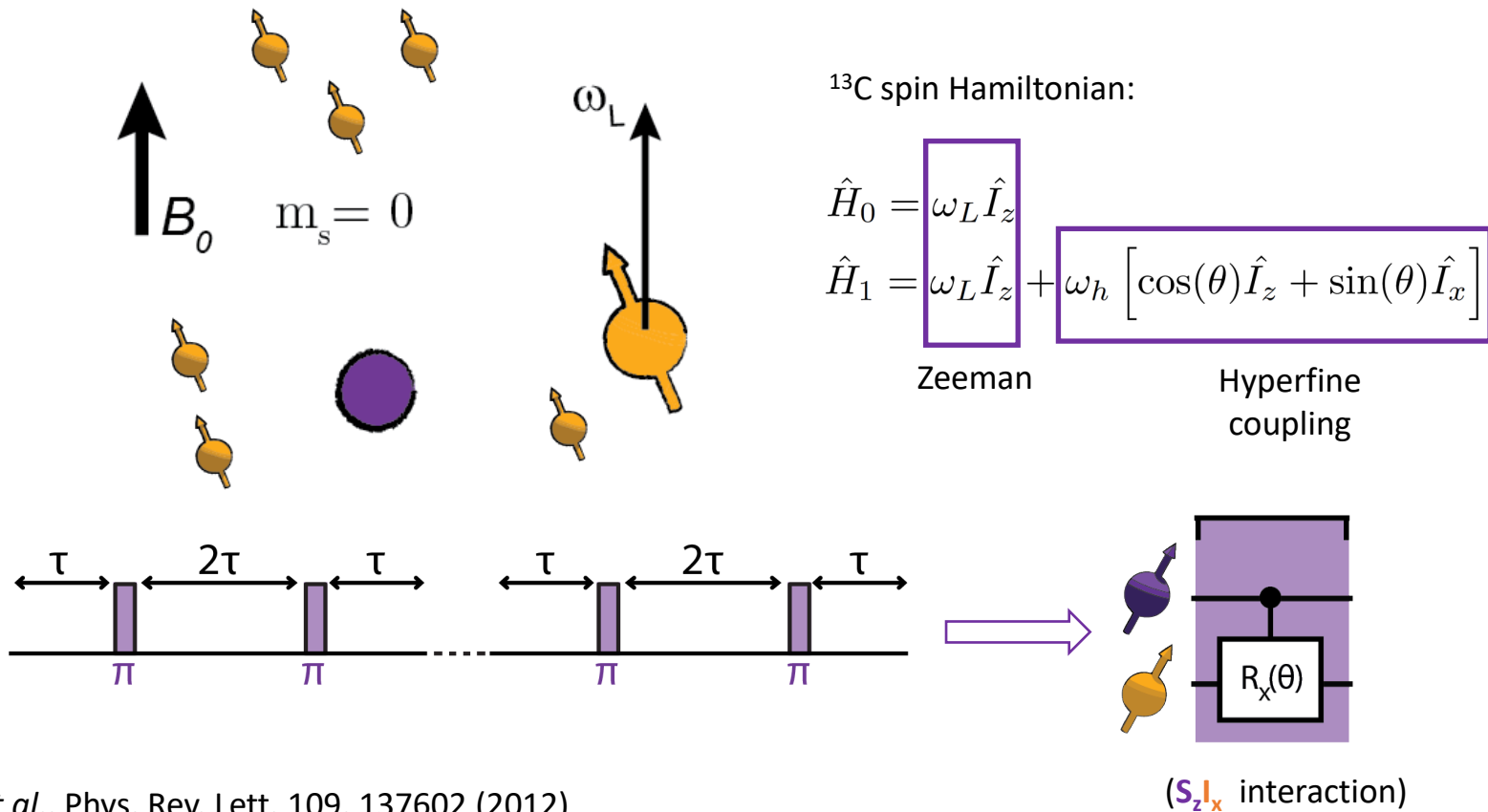
More spins: ^{13}C nuclear spins



More spins: ^{13}C nuclear spins



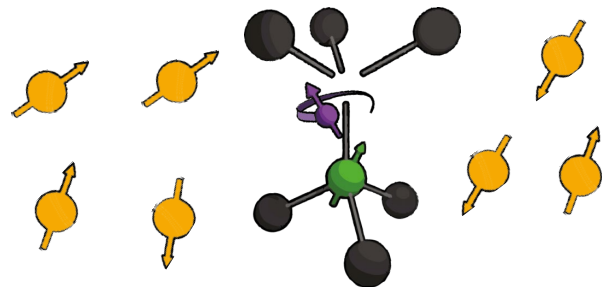
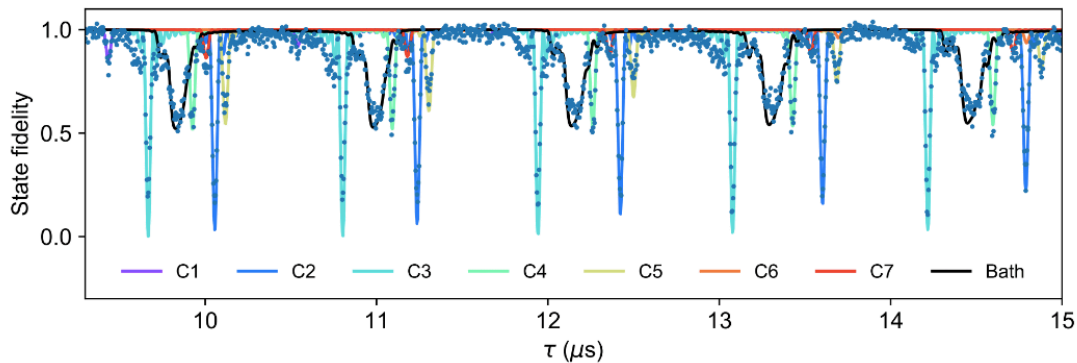
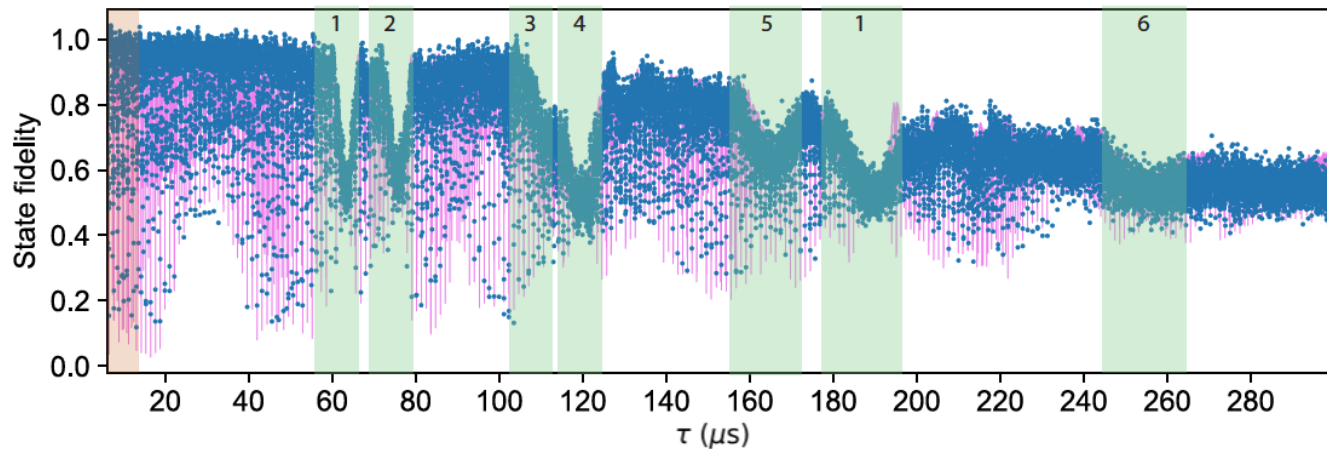
Controlling nuclear spin qubits (quantum picture)



Taminiau *et al.*, Phys. Rev. Lett. 109, 137602 (2012)

Taminiau *et al.*, Nature Nano. 9, 171-176 (2014)

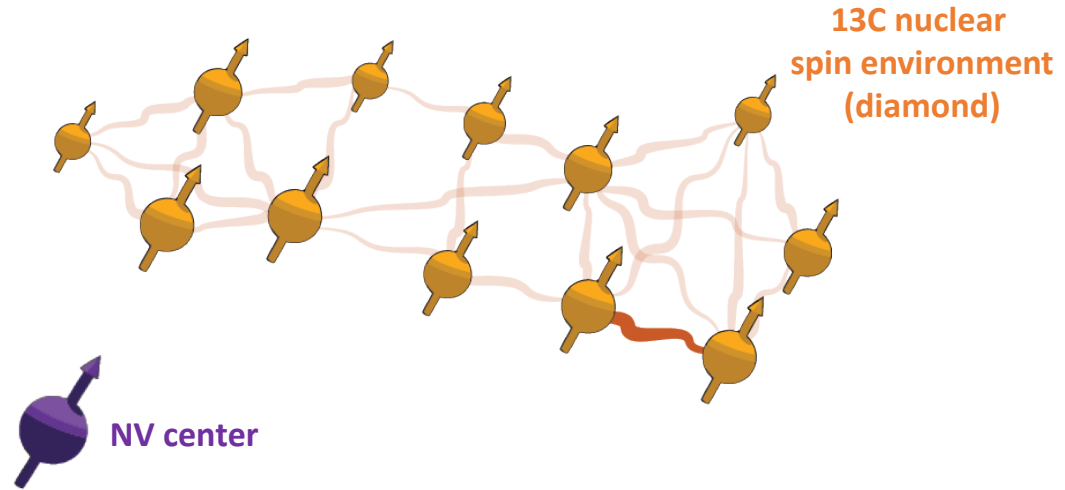
Dynamical decoupling: probing the spin environment



Abobeih et al., Nat. Commun. (2018)

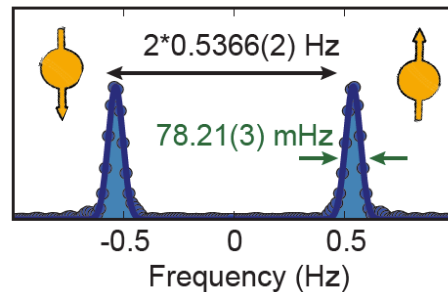
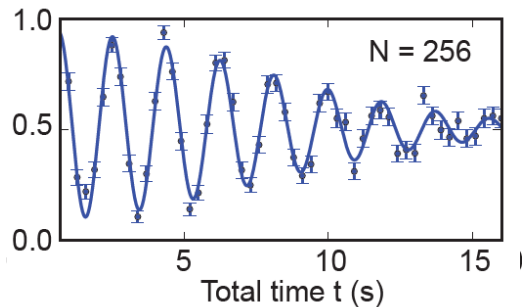
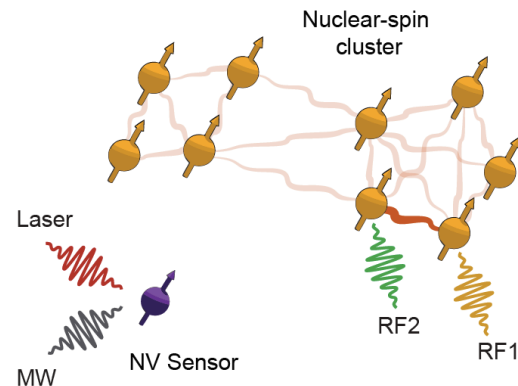
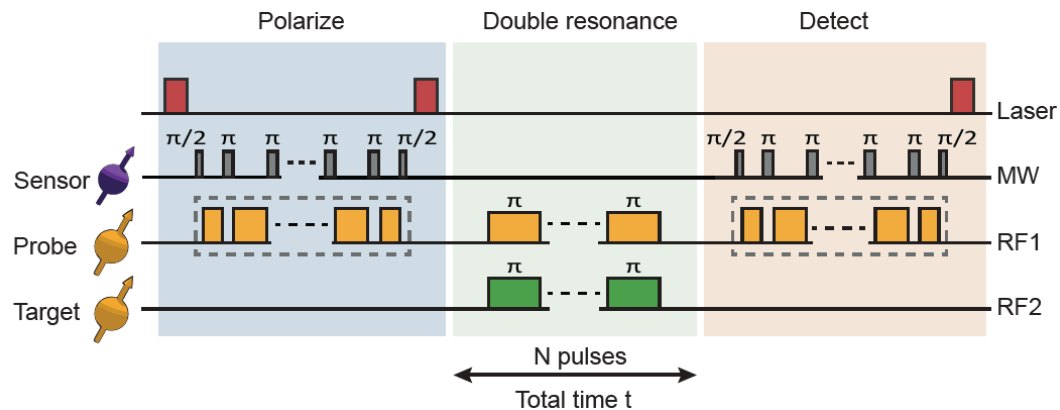
Atomic-scale imaging of your quantum simulator

1. Measure the **nuclear-nuclear interactions**.
2. These reveal the **interconnectivity** of the processor.
3. Transform the measured couplings into **3D structure**.
(= the Hamiltonian)

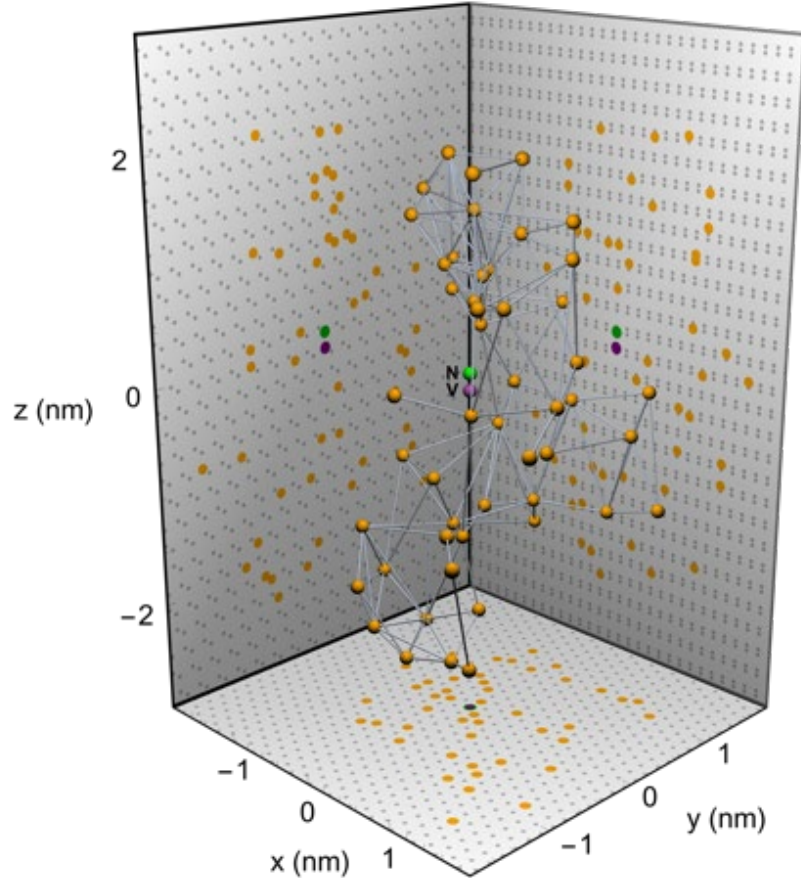


*Electron spin field gradient tags the nuclear spins
Nuclear-nuclear interactions reveal structure*

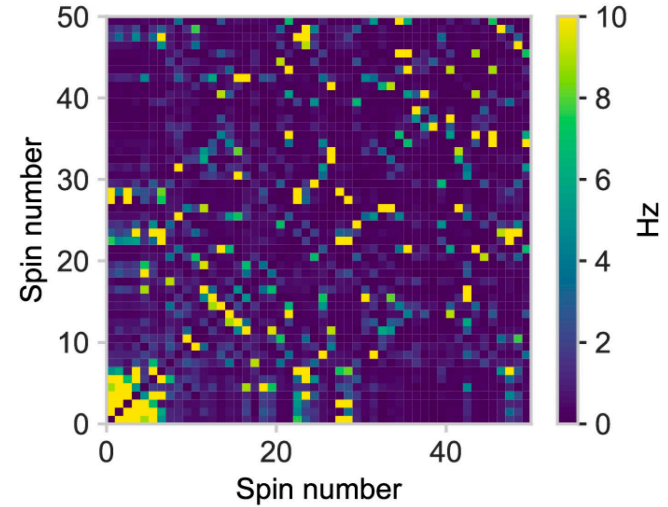
Measure nuclear-nuclear couplings (SEDOR)



A 52-spin-qubit quantum system



All 1225 nuclear-nuclear spin interactions



DTF calculations give full hyperfine tensors:

Takács Comm. Physics, 2024

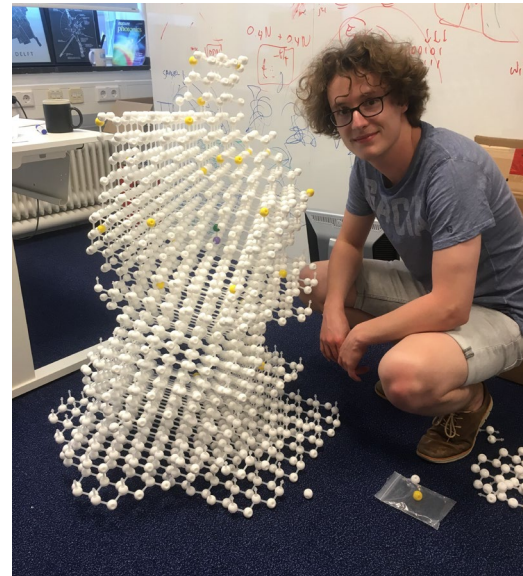
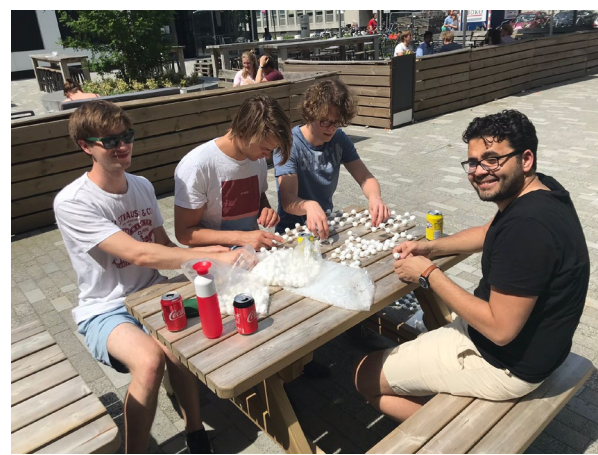


PhD Student
Guido van de Stolpe

A characterized quantum simulator

Abobeih et al., Nature 2019
Bradley et al., PRX 2019
Bartling et al., PRX 2022





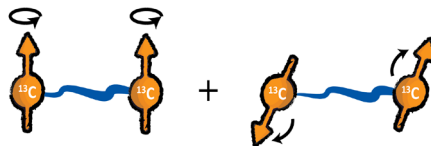
The basic Hamiltonians

~50 hyperfine
tensors known.

hyperfine frequency
shifts



dipolar interactions



RF rotations : tunable rotation
angle & axis

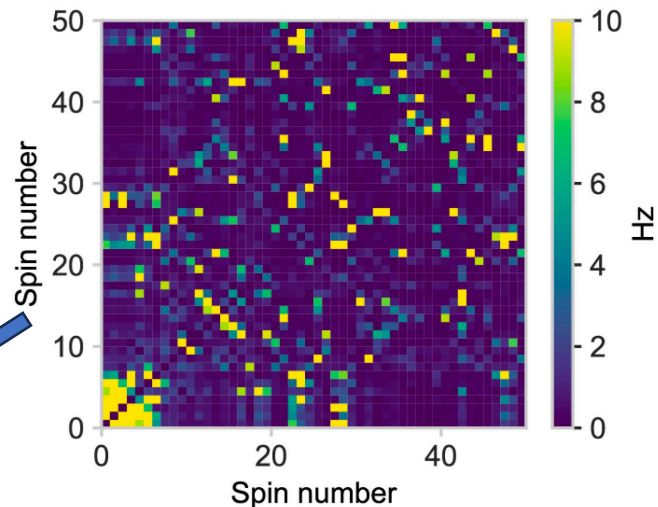


$$H(m_s = 1) = \underbrace{\sum_{i=1}^N h_i I_i^z}_{\text{disorder}} + \underbrace{\sum_{i=1}^N \sum_{j=1}^i J_{ij} \left[I_i^z I_j^z - \frac{1}{2} \left(\cancel{I_i^+ I_j^-} + \cancel{I_i^- I_j^+} \right) \right]}_{\text{ZZ interactions only}} + \underbrace{\sum_{i=1}^N \Omega_i(t) [\cos(\phi_i) I_i^x + \sin(\phi_i) I_i^y]}_{\text{selective: individual qubit control}}$$

$$H(m_s = 0) = \underbrace{\sum_{i=1}^N \cancel{h_i I_i^z}}_{\text{no disorder}} + \underbrace{\sum_{i=1}^N \sum_{j=1}^i J_{ij} \left[I_i^z I_j^z - \frac{1}{2} (I_i^+ I_j^- + I_i^- I_j^+) \right]}_{\text{ZZ + flip-flop interactions}} + \underbrace{\sum_{i=1}^N \Omega_i(t) [\cos(\phi_i) I_i^x + \sin(\phi_i) I_i^y]}_{\text{global qubit control}}$$

The basic Hamiltonians

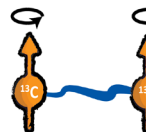
All 1225 nuclear-nuclear spin interactions



hyperfine frequency shifts



dipole



rotation



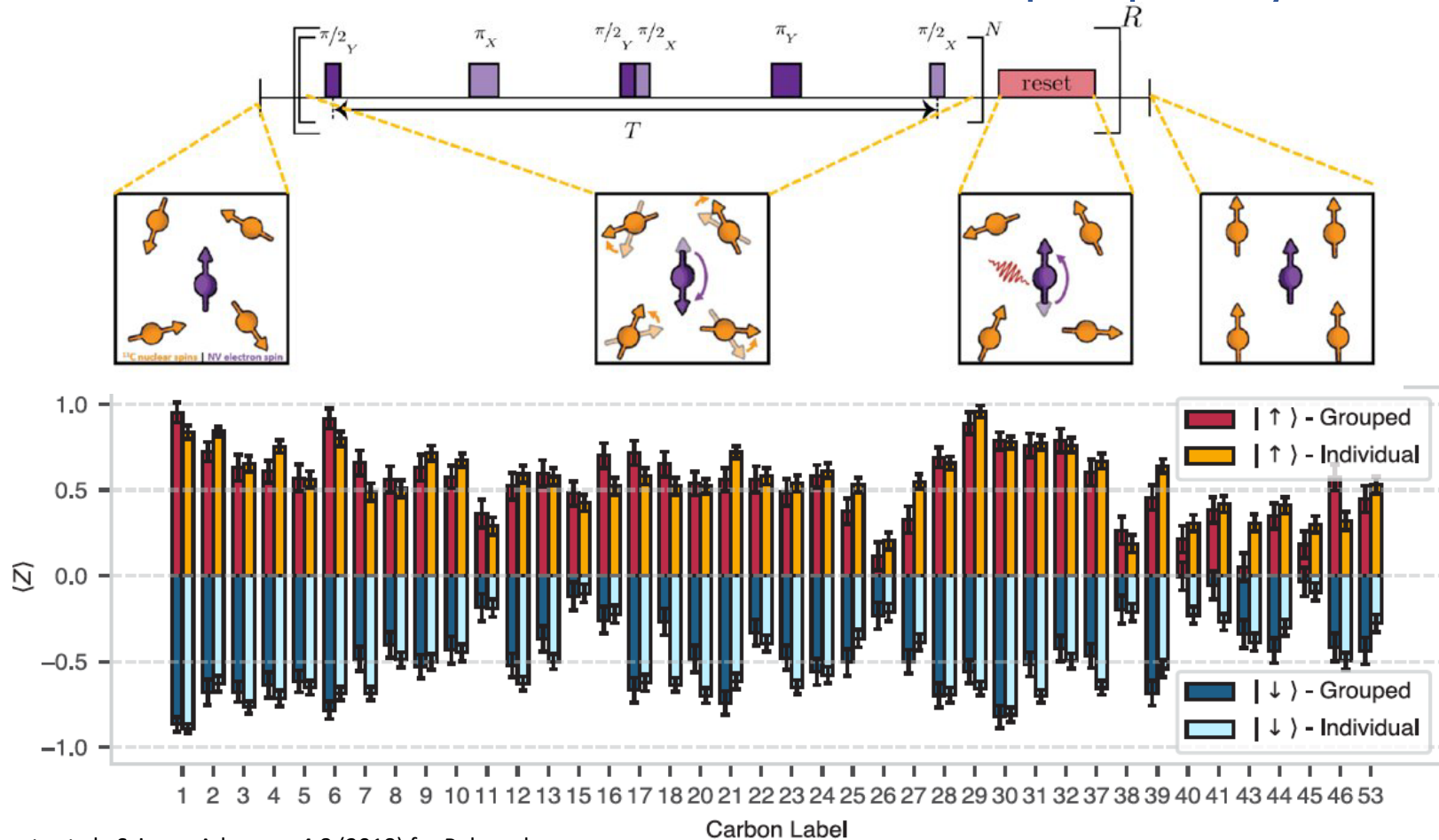
$$H(m_s = 1) = \underbrace{\sum_{i=1}^N h_i I_i^z}_{\text{disorder}} + \underbrace{\sum_{i=1}^N \sum_{j=1}^i J_{ij} \left[I_i^z I_j^z - \right]}_{\text{ZZ interactions only}}$$

$$I_i^x + \sin(\phi_i) I_i^y]$$

selective: individual qubit control

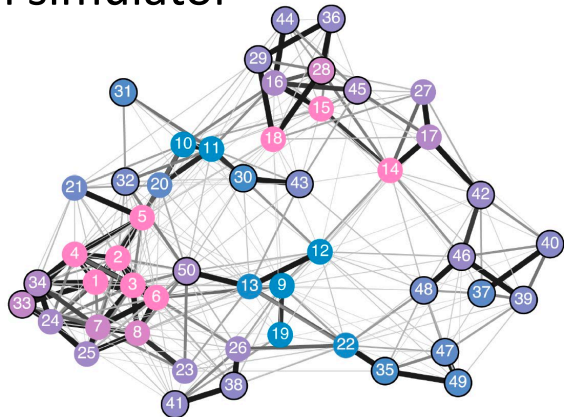
$$H(m_s = 0) = \underbrace{\sum_{i=1}^N \cancel{h_i I_i^z}}_{\text{no disorder}} + \underbrace{\sum_{i=1}^N \sum_{j=1}^i J_{ij} \left[I_i^z I_j^z - \frac{1}{2} (I_i^+ I_j^- + I_i^- I_j^+) \right]}_{\text{ZZ + flip-flop interactions}} + \underbrace{\sum_{i=1}^N \Omega_i(t) [\cos(\phi_i) I_i^x + \sin(\phi_i) I_i^y]}_{\text{global qubit control}}$$

Initialization and measurement of a 40-spin qubit system

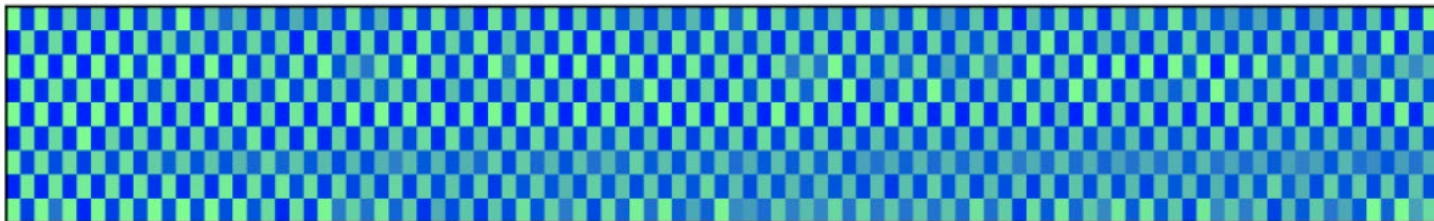


Outline

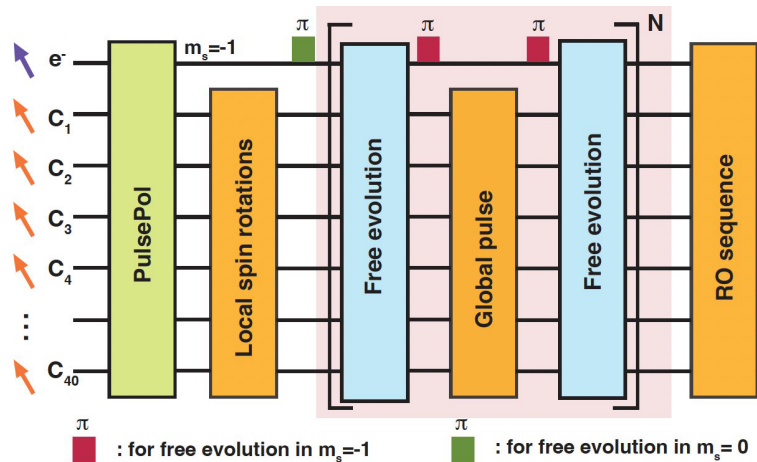
(1) System: a programmable diamond quantum simulator



(2) 1D: Discrete time crystals

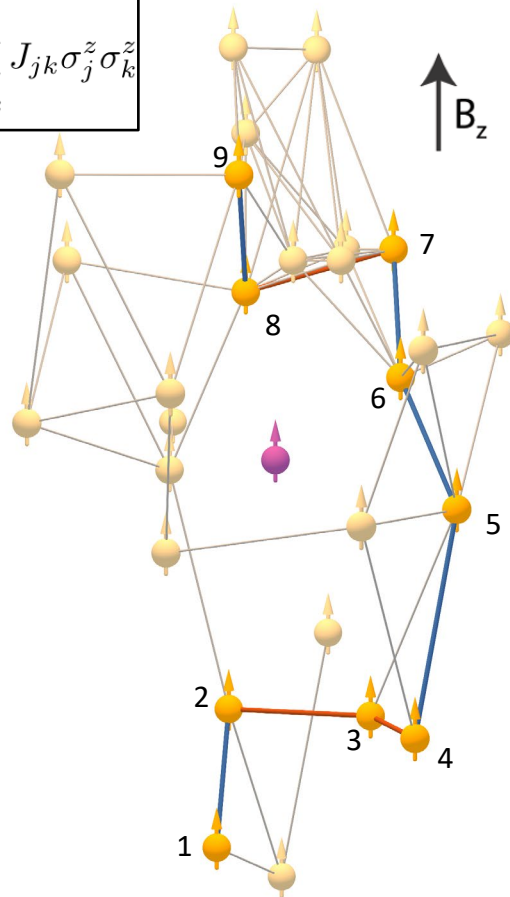


(3) Going 3D: ‘Critical’ Time Crystals



Programming a 1D spin chain

$$H_{\text{int}} = \sum_j^L (B + h_j) \sigma_j^z + \sum_{j < k}^L J_{jk} \sigma_j^z \sigma_k^z$$



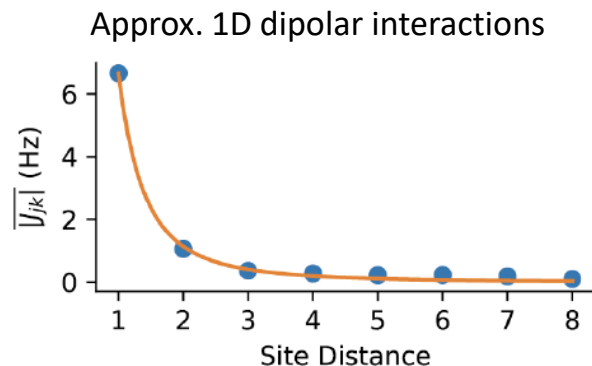
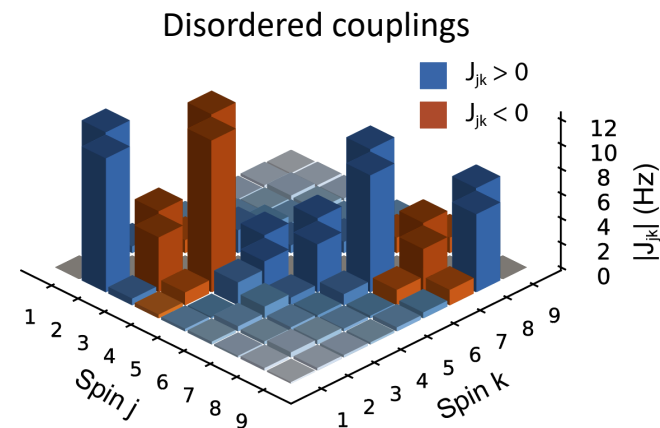
Collaboration with:



Norman
Yao

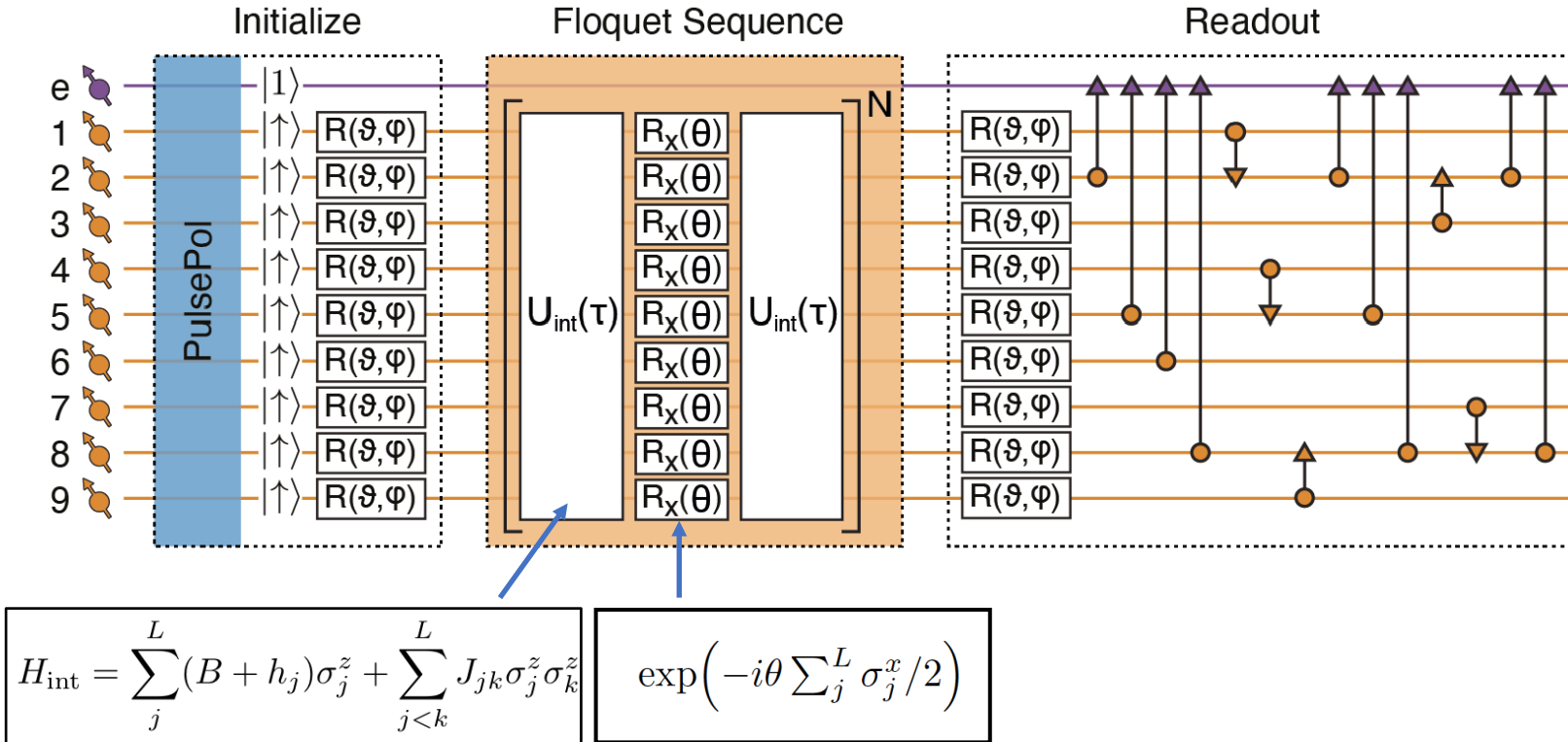


Francisco
Machado

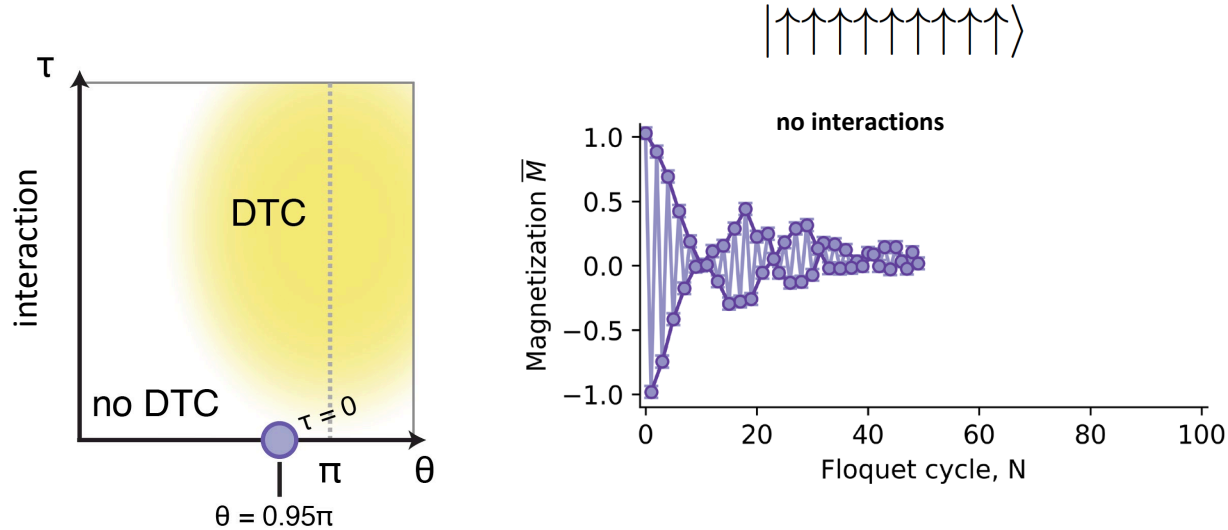


J. Randall et al., *Science* 374, 6574 (2021)

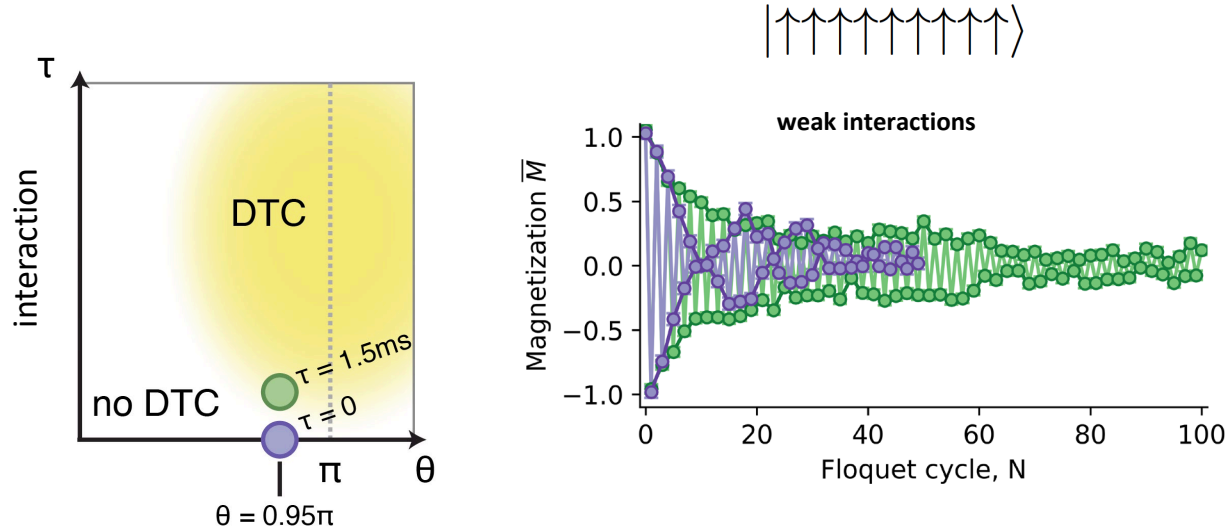
Programming the DTC Hamiltonian



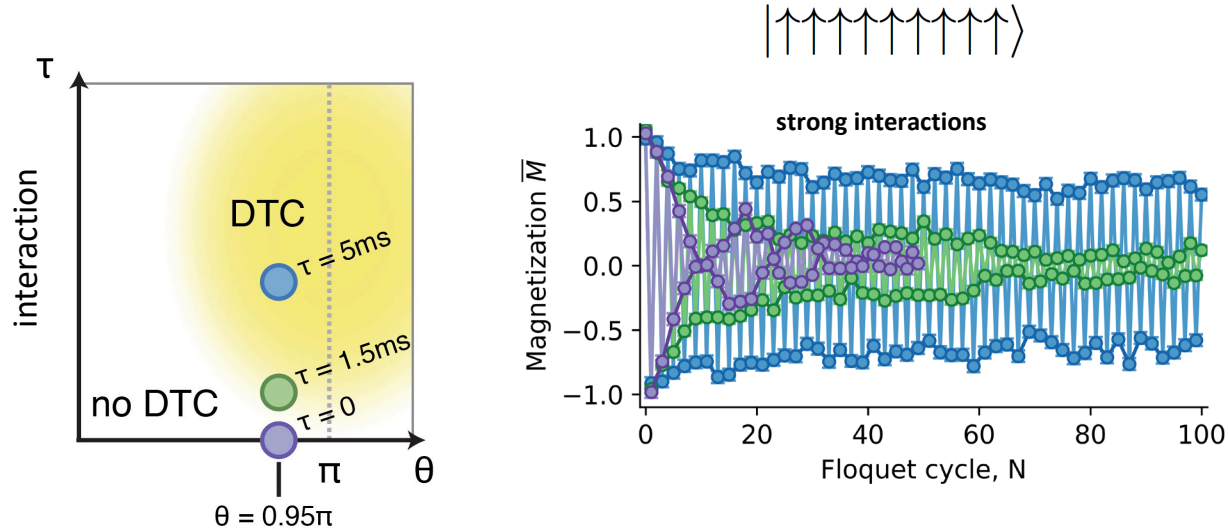
Experiment: Tuning through the DTC phase transition



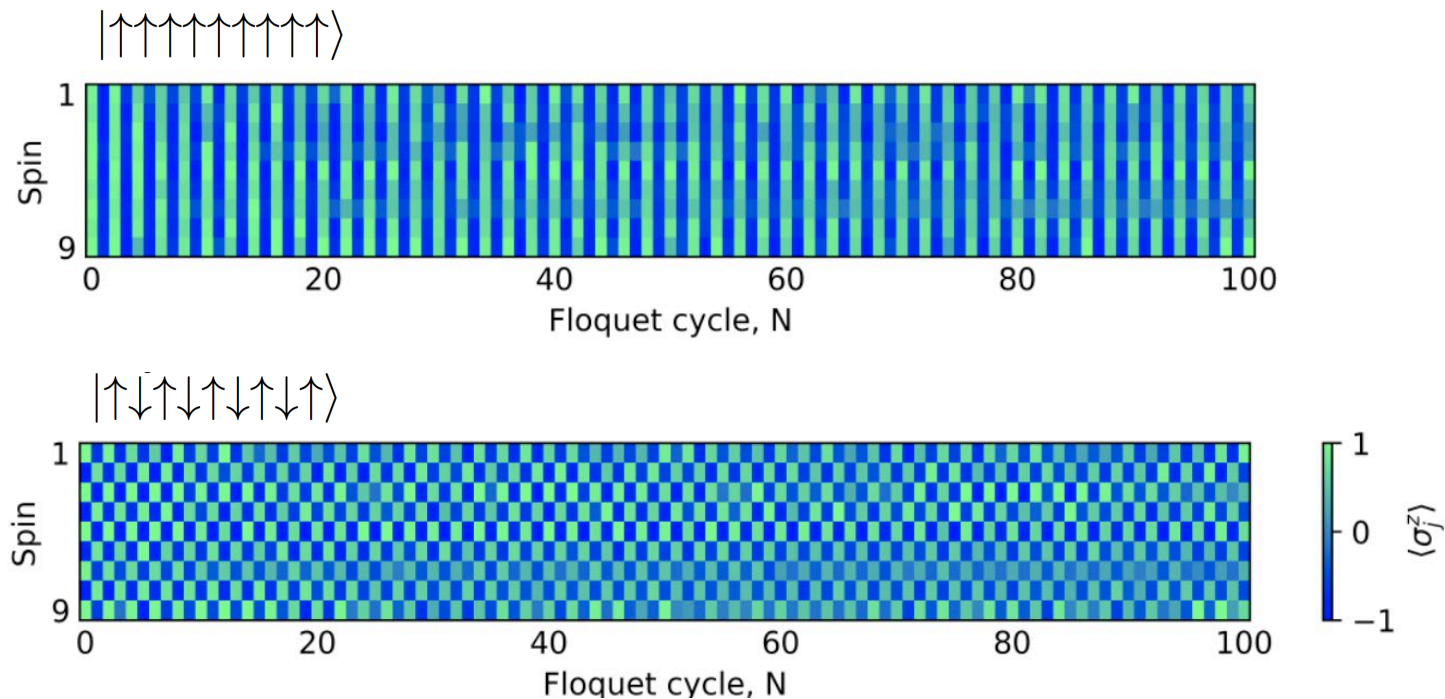
Experiment: Tuning through the DTC phase transition



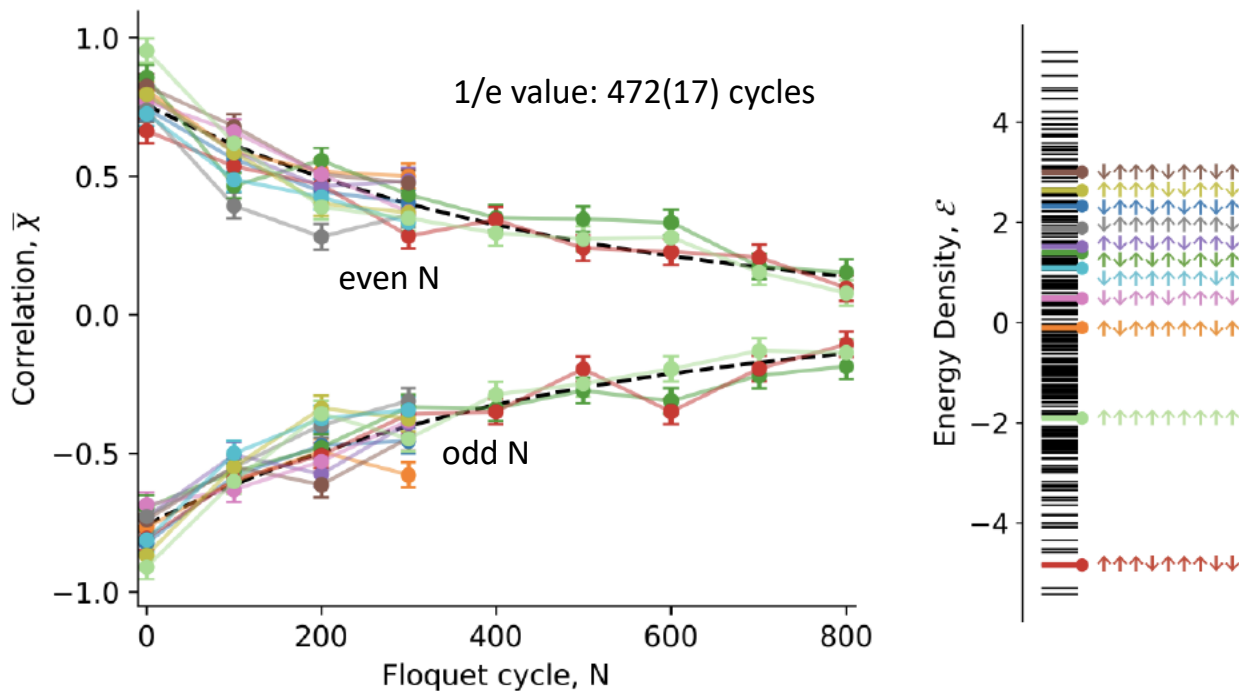
Experiment: Tuning through the DTC phase transition



Signatures of MBL DTC order



Observing the signatures of MBL DTC order



See also superconducting qubits:

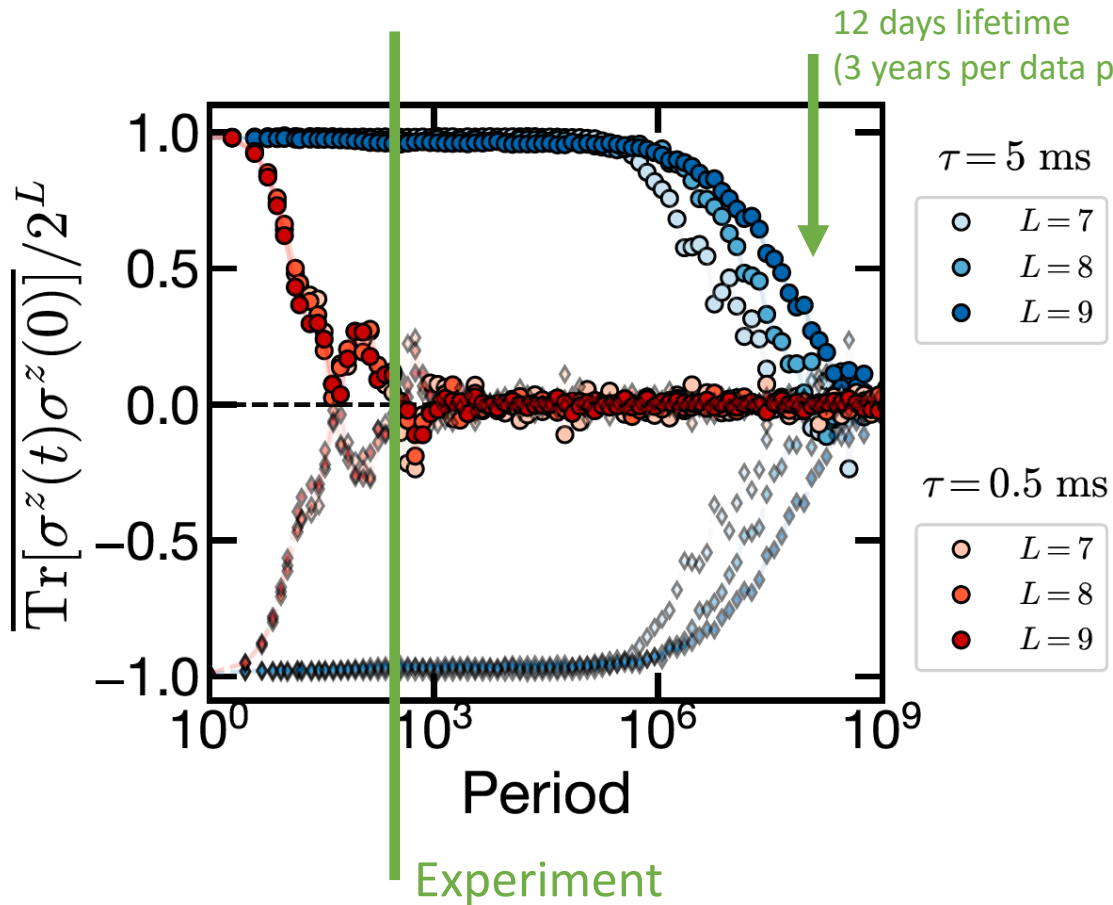
Mi et al., Nature 601, 531 (2022)

Frey and Rachel., Sci. Adv.8, eabm7652 (2022)

$$\bar{\chi} = \frac{1}{L} \sum_{j=1}^L \langle \sigma_j^z(N) \rangle \text{sgn}[\langle \sigma_j^z(0) \rangle]$$

J. Randall et al., Science 374, 1474 (2021)

Comparing to the theoretical DTC lifetimes



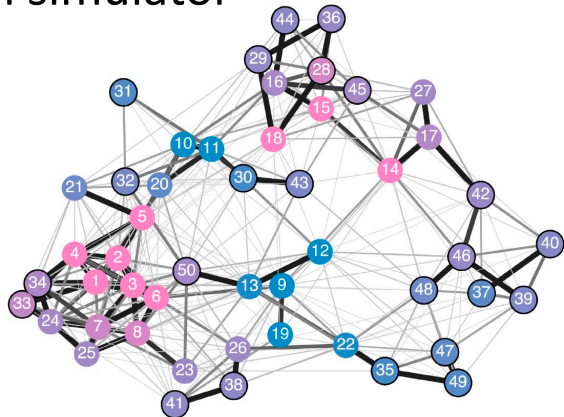
Experiment is not limited by finite-size effects

Experiment can be distinguished from the cases that are certainly not a MBL DTC:

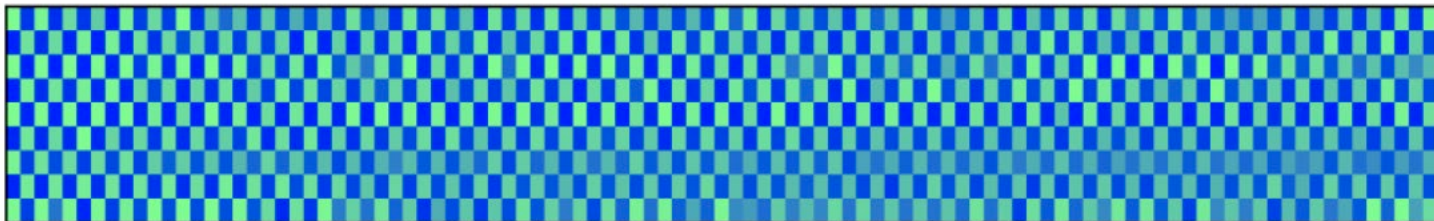
- Weak or no interactions
- Weak or no Ising-even disorder
- Higher dimensional system, such as 3D (?)

Outline

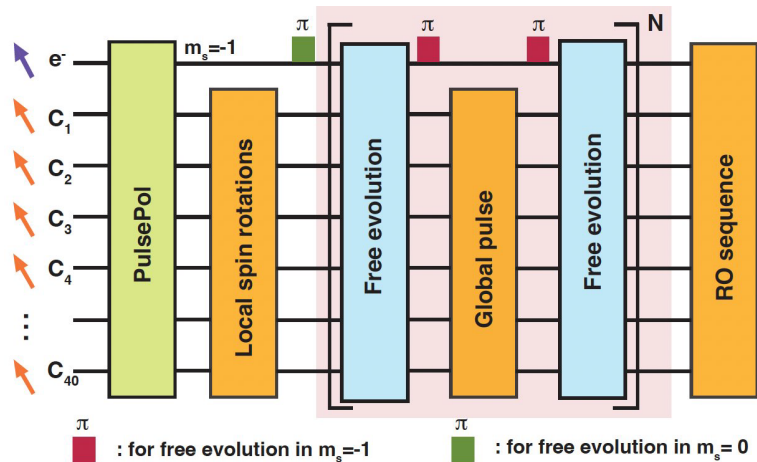
(1) System: a programmable diamond quantum simulator



(2) 1D: Discrete time crystals



(3) Going 3D: ‘Critical’ Time Crystals



What about 3D? The “critical time crystal”

Critical Time Crystals in Dipolar Systems

[Wen Wei Ho](#)¹, [Soonwon Choi](#)², [Mikhail D. Lukin](#)², and [Dmitry A. Abanin](#)¹

Phys. Rev. Lett. **119**, 010602 – Published 6 July, 2017

Observation of discrete time–crystalline order in a disordered dipolar many–body system

Soonwon Choi^{1*}, Joonhee Choi^{1,2*}, Renate Landig^{1*}, Georg Kucsko¹, Hengyun Zhou¹, Junichi Isoya³, Fedor Jelezko⁴, Shinobu Onoda⁵, Hitoshi Sumiya⁶, Vedika Khemani¹, Curt von Keyserlingk⁷, Norman Y. Yao⁸, Eugene Demler¹ & Mikhail D. Lukin¹

[Nature](#) **543**, 221–225 (2017)

- No MBL expected in 3D
- No true phase transition into a Discrete Time Crystal
- Slow thermalization in **3D disordered dipolar-coupled** systems predicted

Literature:

NV ensemble (3D, “Observation of discrete time-crystalline order in a disordered dipolar many-body system”, Choi et al. (2016))

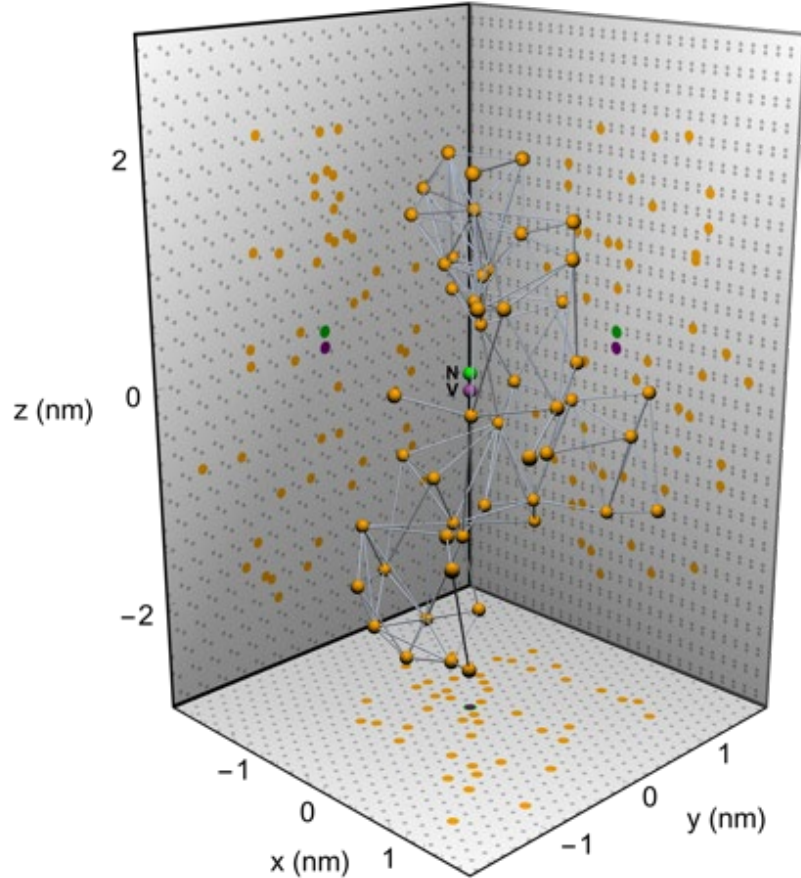
“Critical Time Crystals in Dipolar Systems”, Ho et al.

“Critical Thermalization of a Disordered Dipolar Spin System in Diamond”, Kucskoo et al.

“Critical prethermal discrete time crystal created by two-frequency driving”, Beatrez et al. (2023))

“Probing Quantum Thermalization of a Disordered Dipolar Spin Ensemble with Discrete Time-Crystalline Order”, Choi et al.)

3D dipolar-coupled nuclear-spin system



Global pulses → “all” spins participate

Microscopic control/observation → 40 individual spins

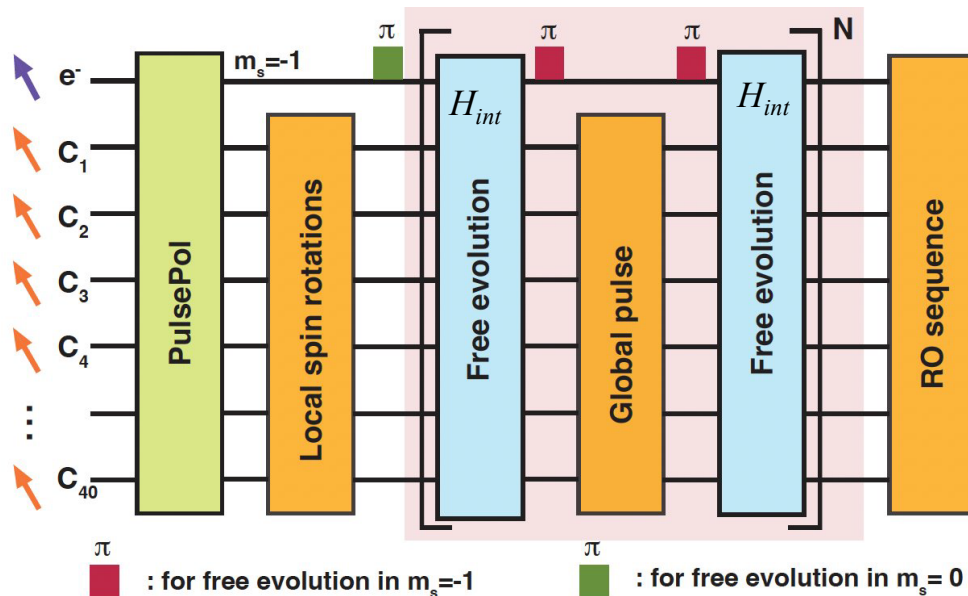
Microscopic knowledge → model individual spin dynamics



PhD Candidate

Christina Ioannou

Programming the 3D Critical Time Crystal Hamiltonian



$$H_{int}^{m_s=0} = \sum_{j \neq i}^N C_{ij} (I_i^z I_j^z - \frac{1}{2} (I_i^+ I_j^- + I_i^- I_j^+))$$

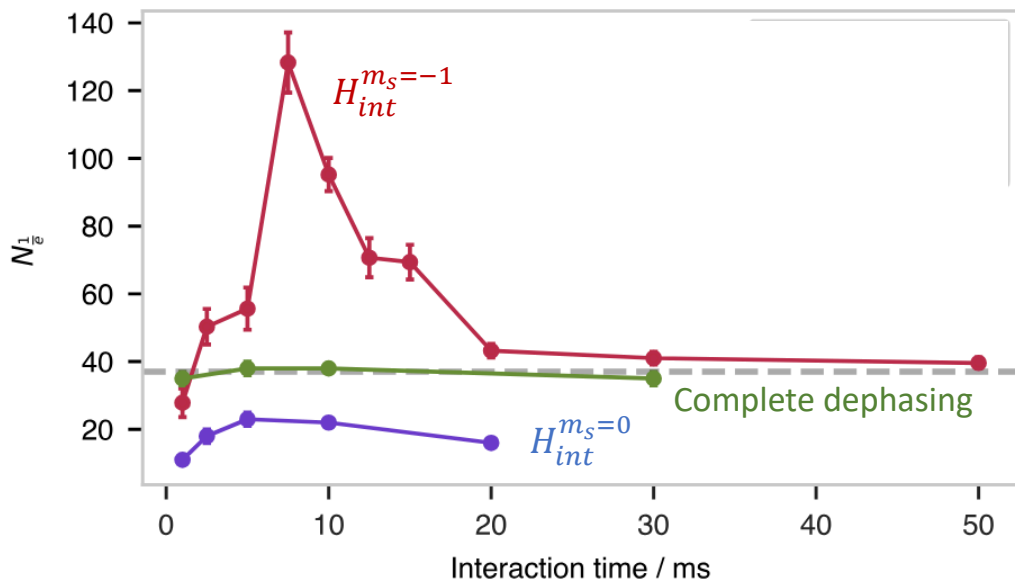
Ising Flip-flop term

Hypothesis:
No memory of initial state except
for special states

$$H_{int}^{m_s=-1} = \sum_{j \neq i}^N C_{ij} (I_i^z I_j^z - \cancel{\frac{1}{2} (I_i^+ I_j^- + I_i^- I_j^+)})$$

Hypothesis:
Time crystal-like response possible

Global response (averaged signal)



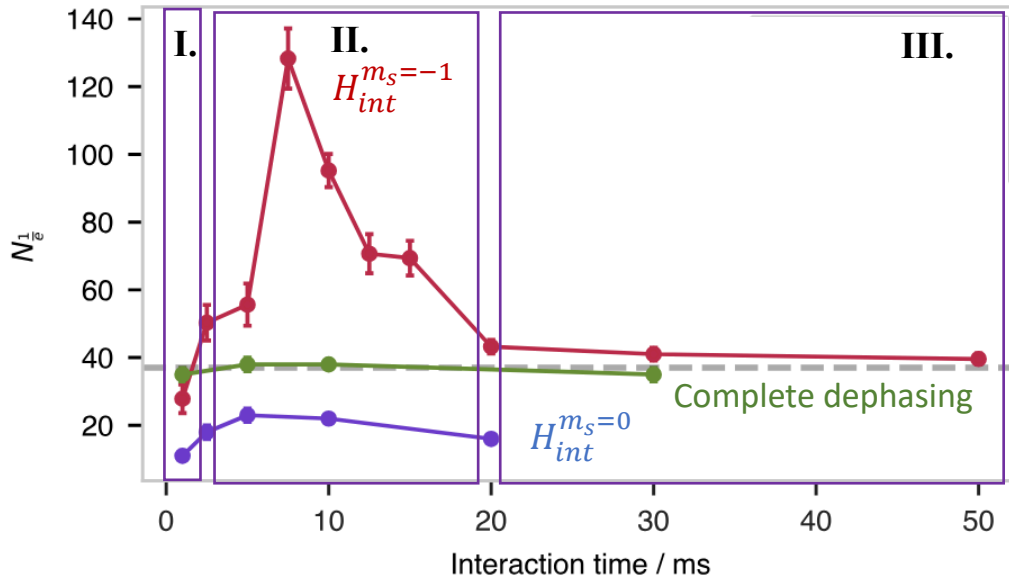
$$H_{int}^{m_s=-1} = \sum_{j \neq i}^N C_{ij} (I_i^z I_j^z - \frac{1}{2} (I_i^+ I_j^- + I_i^- I_j^+))$$

- Long lived response
- “Critical” time crystal

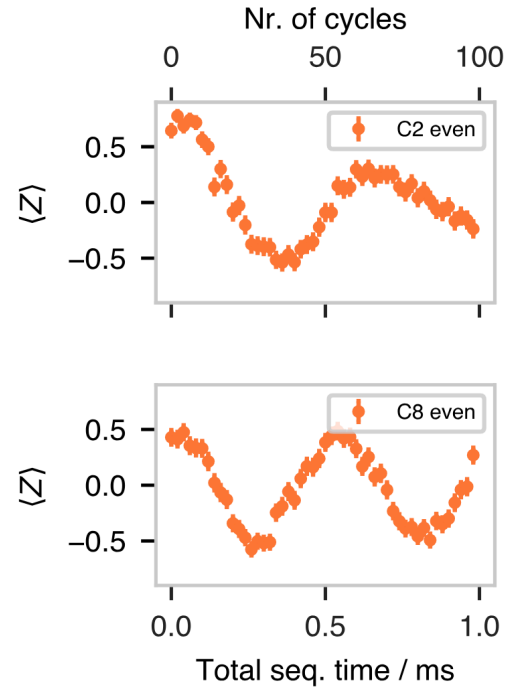
$$H_{int}^{m_s=0} = \sum_{j \neq i}^N C_{ij} (I_i^z I_j^z - \frac{1}{2} (I_i^+ I_j^- + I_i^- I_j^+))$$

- No long-lived response
- No time crystal (?)

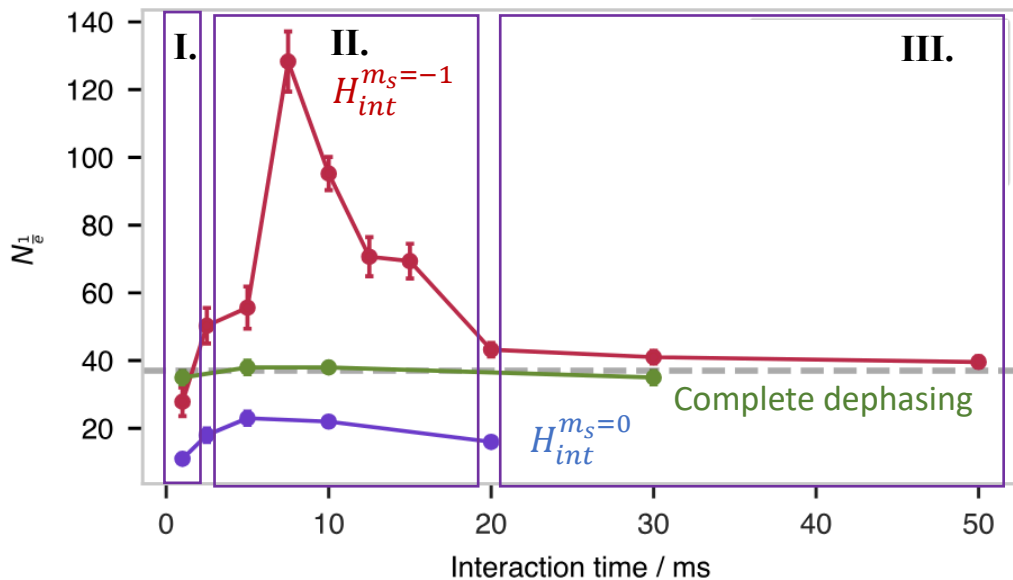
Three dynamical regimes



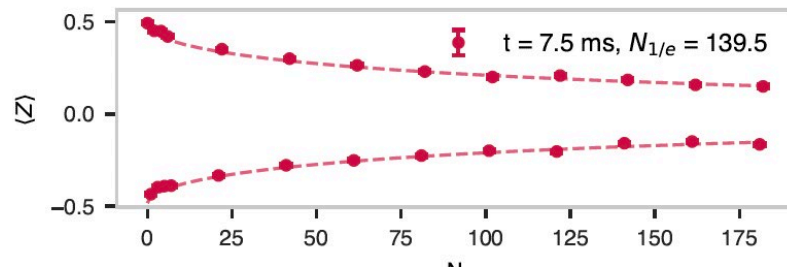
I. Coherent rotations (no interactions)



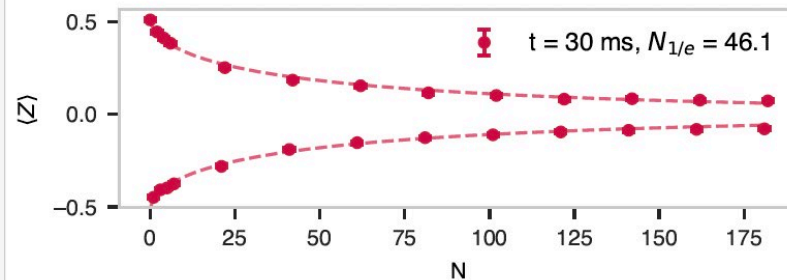
Three dynamical regimes



II. Critical time crystal

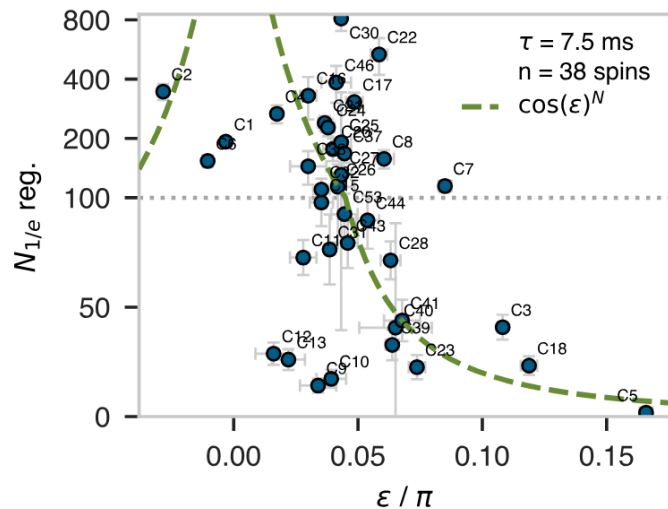


III. Dephasing (long tau)



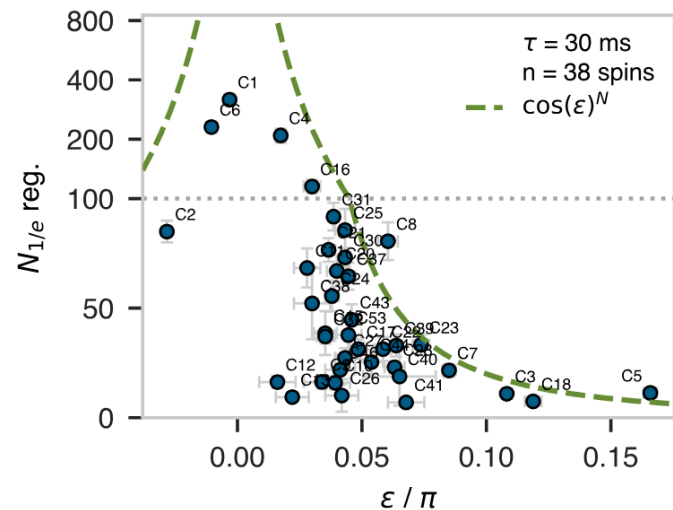
Microscopic: dynamical regimes

II. Critical Time Crystal



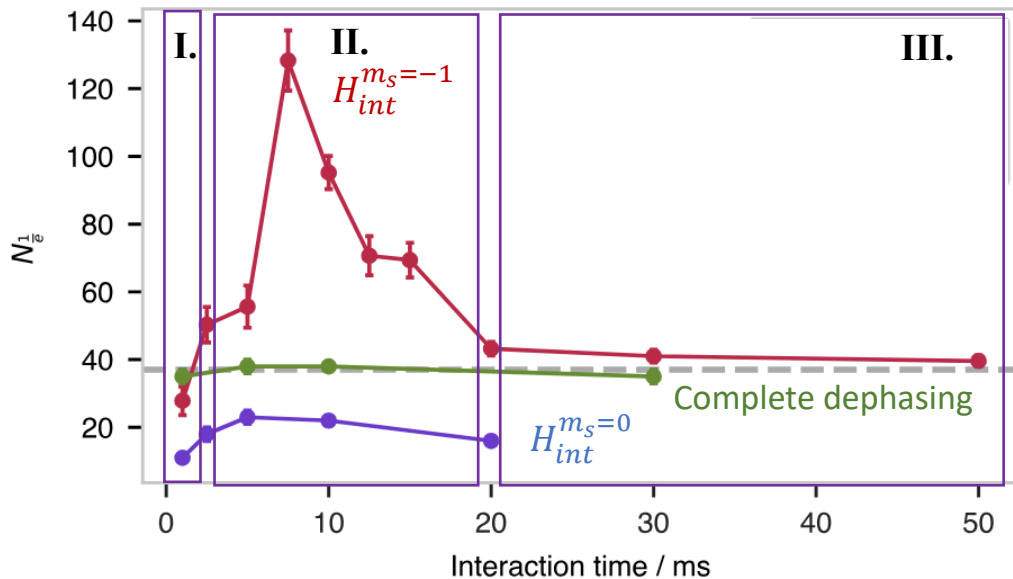
Large spread of individual lifetimes including long-lived spins

III. Large tau / dephasing

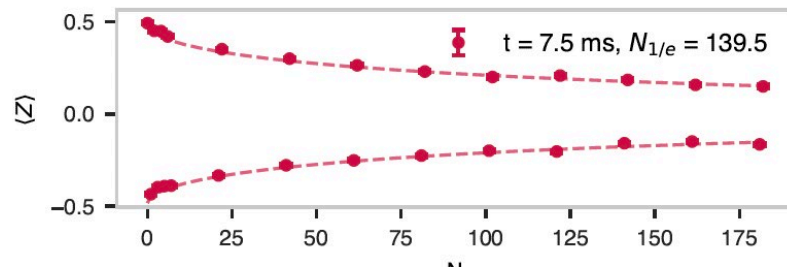


Individual spins are constrained by simple decoherence model

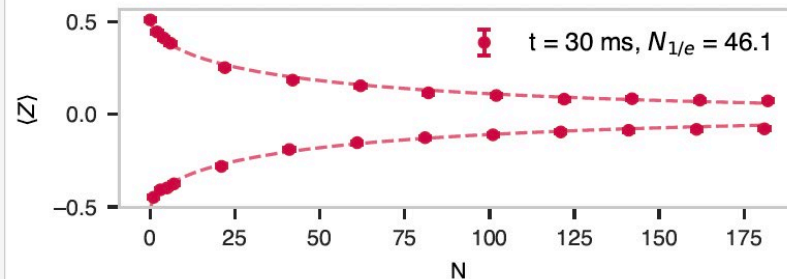
Microscopic: dynamical regime control experiment



II. Critical time crystal

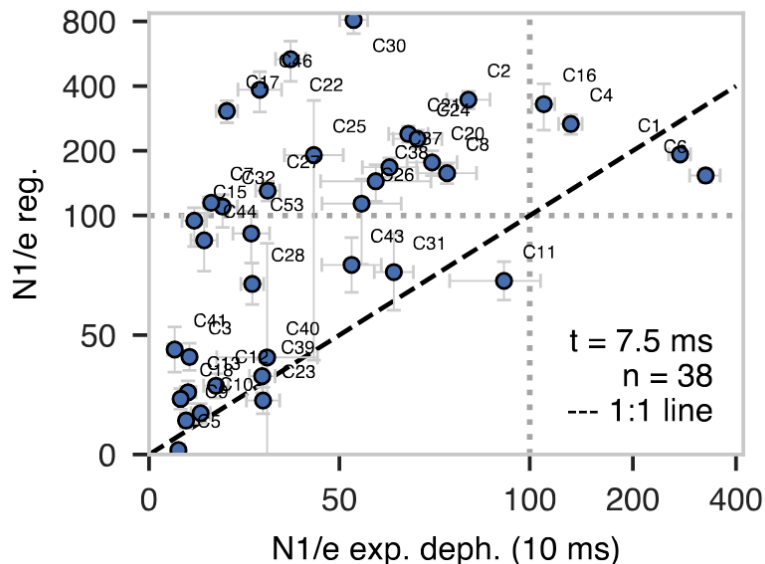


III. Dephasing (long tau)



Microscopic: dynamical regime control experiment

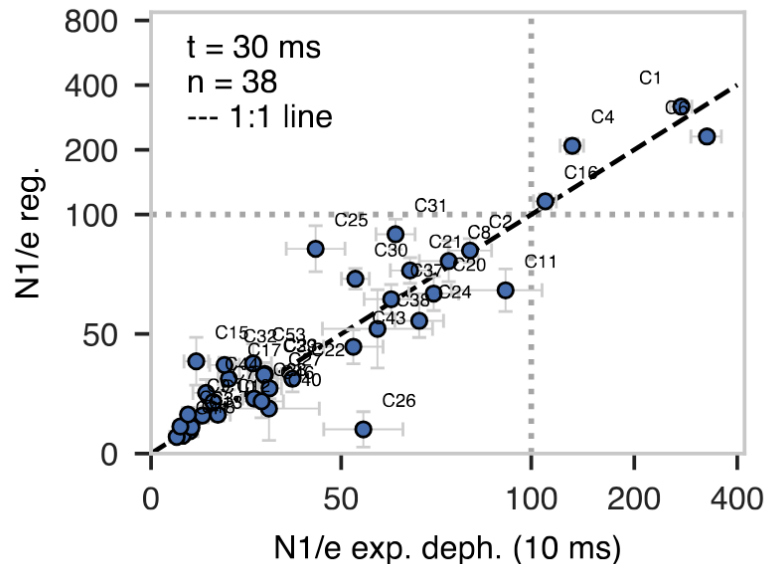
II. Critical Time Crystal



Large spread of individual lifetimes including long-lived spins

Not explainable by decoherence alone.

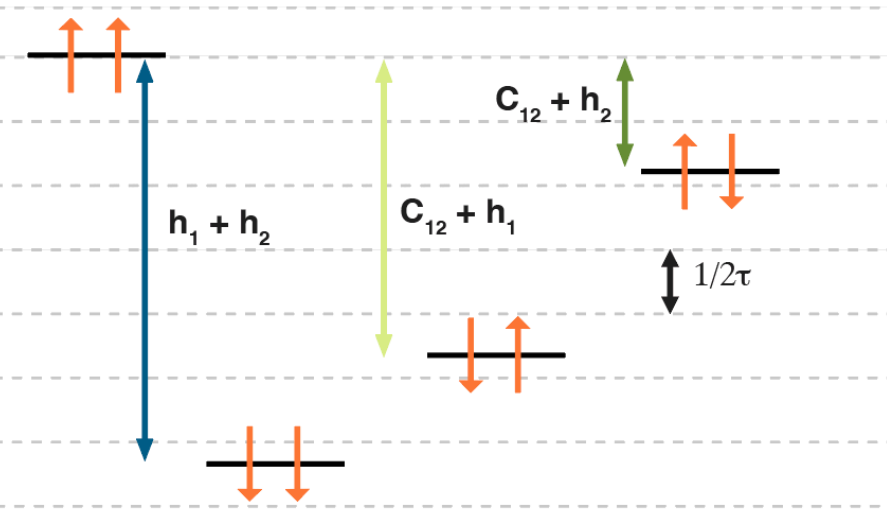
III. Large tau / dephasing



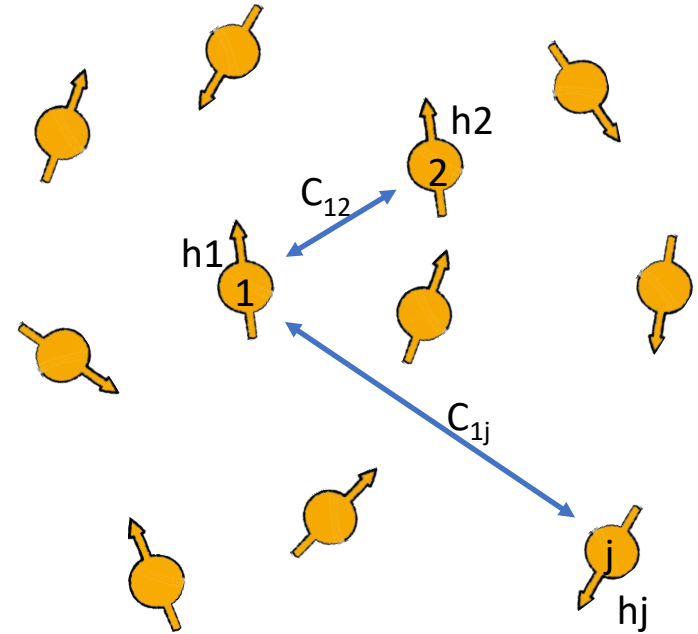
Each spin completely decoheres between pulses

Microscopic: Relaxation mechanism and resonances

Two-spin picture

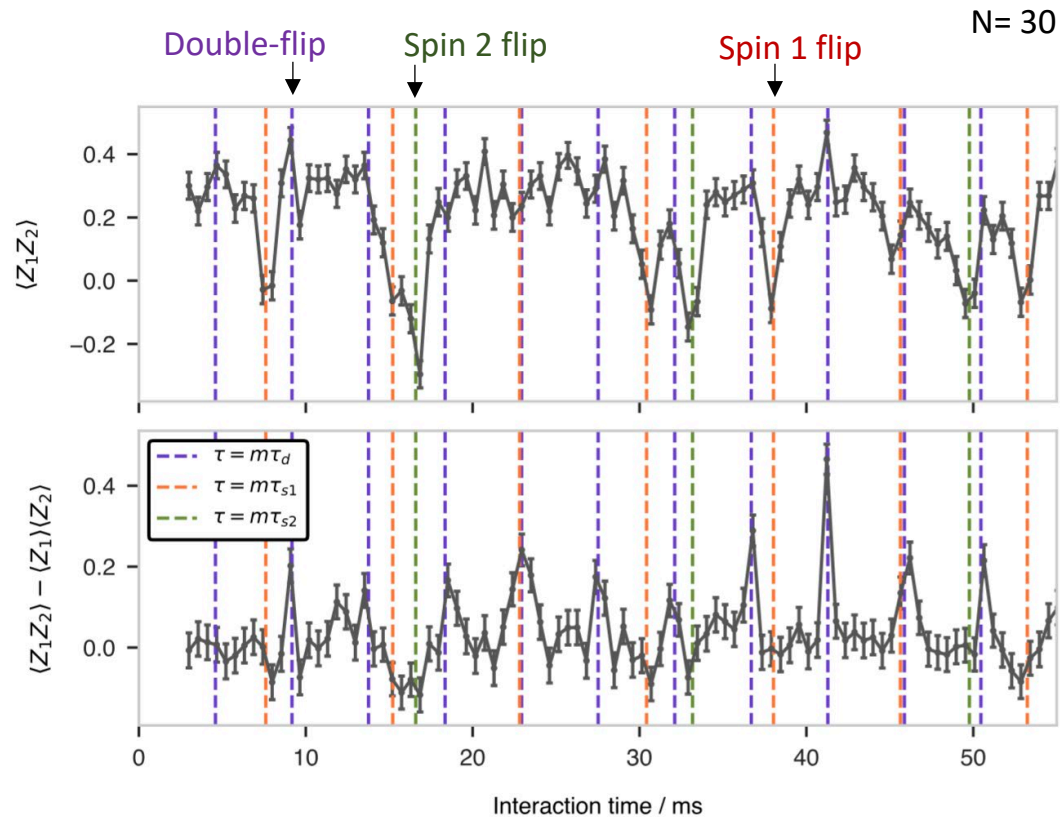
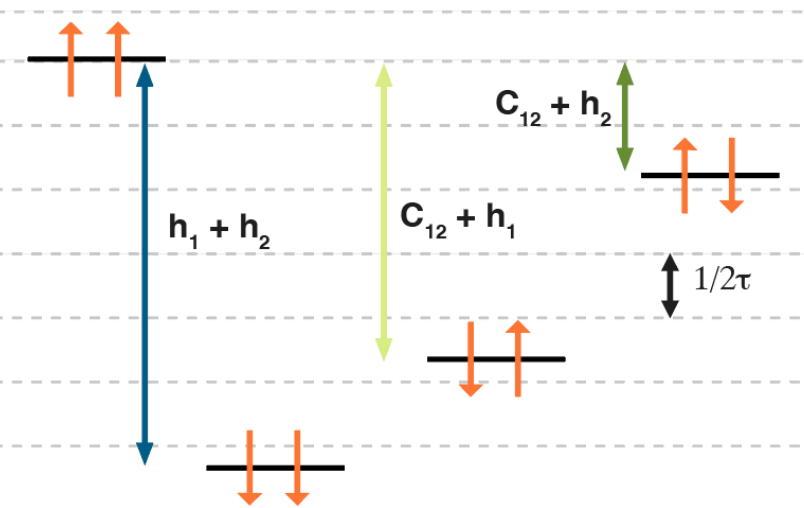


$$h_1 = \sum C_{1j} s_j$$



Microscopic: Relaxation mechanism and resonances

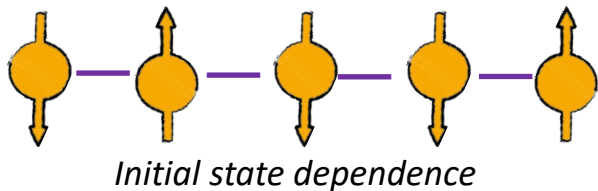
Two-spin picture



Outlook: understanding microscopic the dynamics

Critical time crystal (3D)

- Can we understand the **microscopic dynamics** of the experiment based on our **microscopic knowledge** of the system.



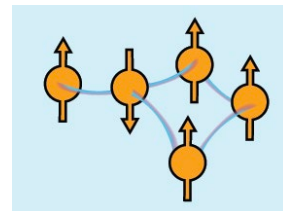
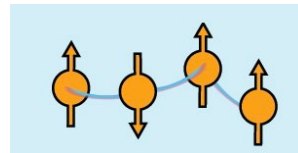
PhD Candidate
Christina Ioannou



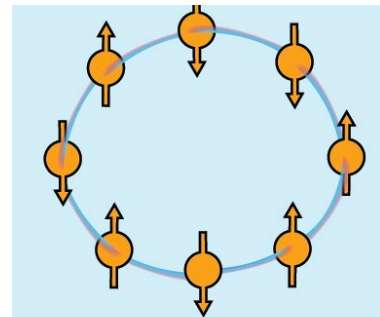
PhD Candidate
Małgorzata (Gosia) Kucharska

Discrete time crystal (MBL)

- What limits the lifetime?
- Transition from a long-lived phase to the crystal melting?

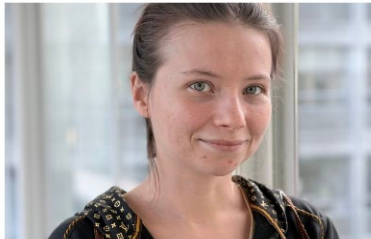
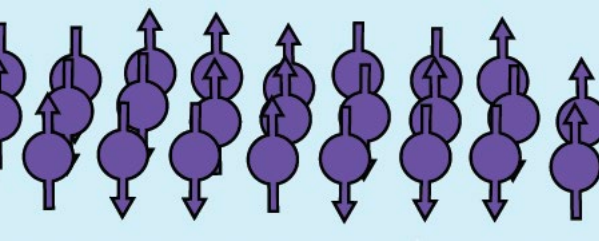


Connectivity



Outlook: engineering coupled electron-spin devices

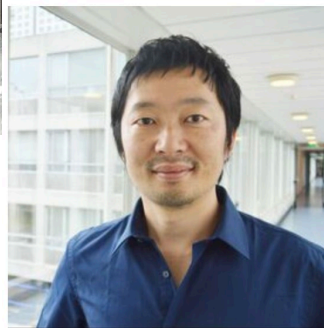
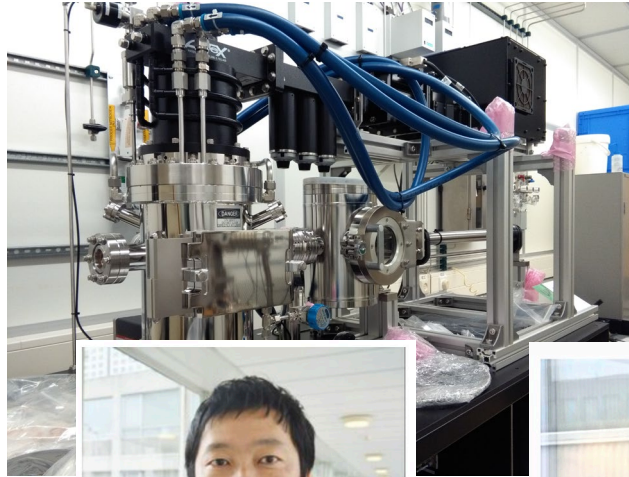
Coupled electron spins



PhD Candidate

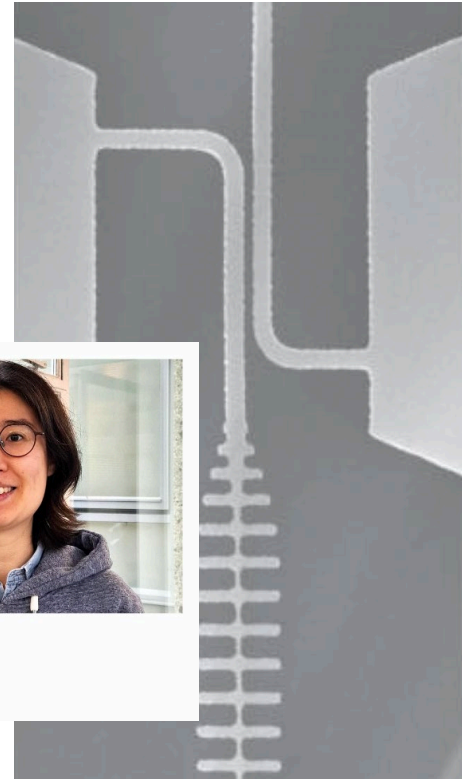
Małgorzata (Gosia) Kucharska

Diamond growth (delta doping of NV)



Takashi Yamamoto

Silicon-carbide devices

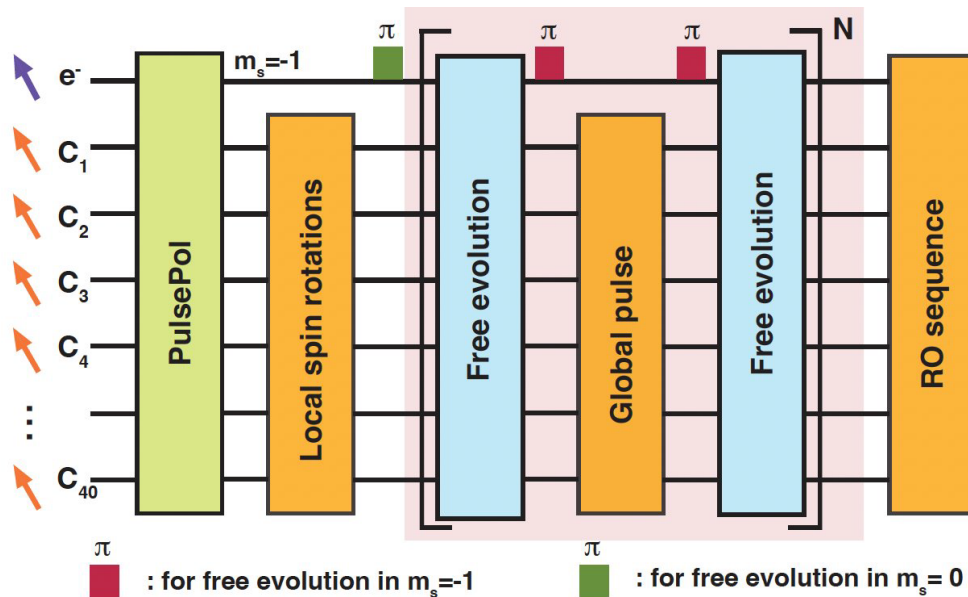


Postdoc

Lin Jin

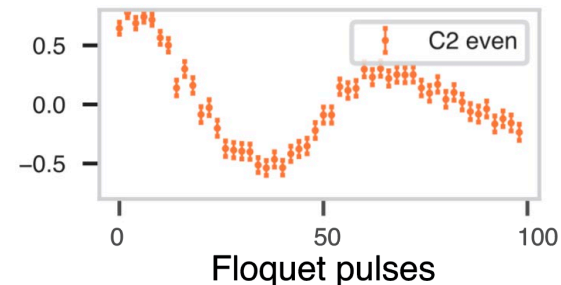


Programming the 3D Critical Time Crystal Hamiltonian

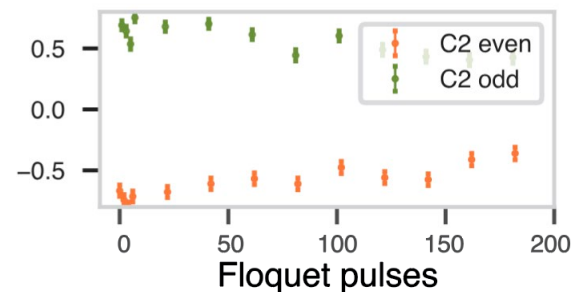


Example ($m_s = -1$)

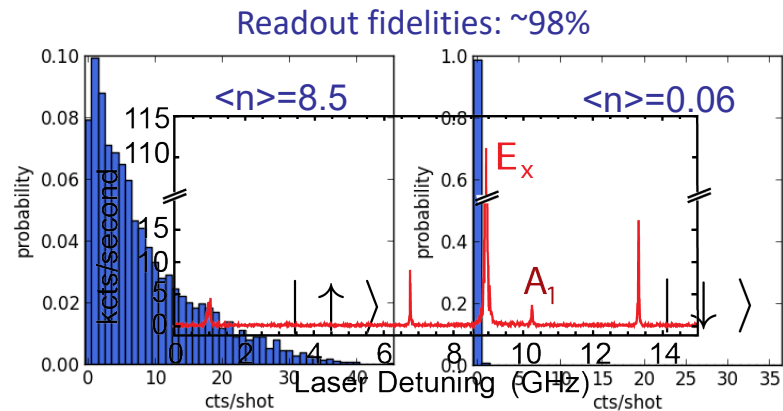
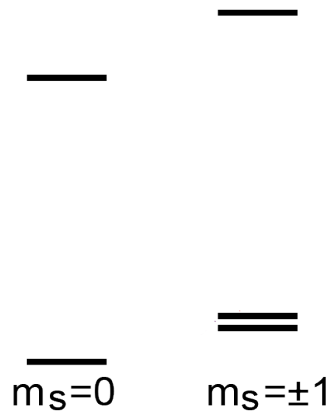
No interaction time



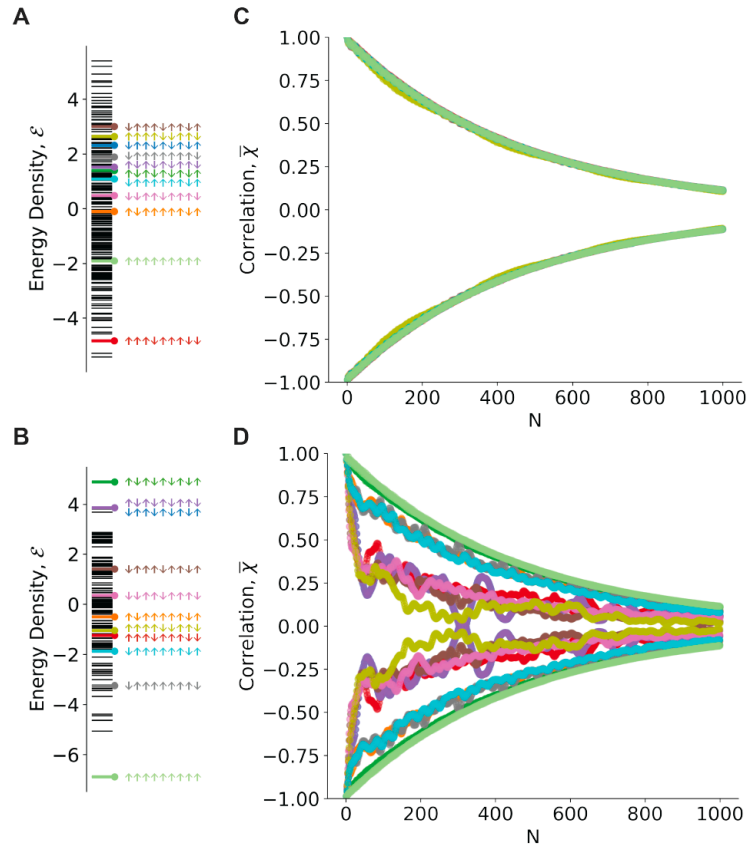
7.5 ms



Optical measurements of the electron spin at 4K



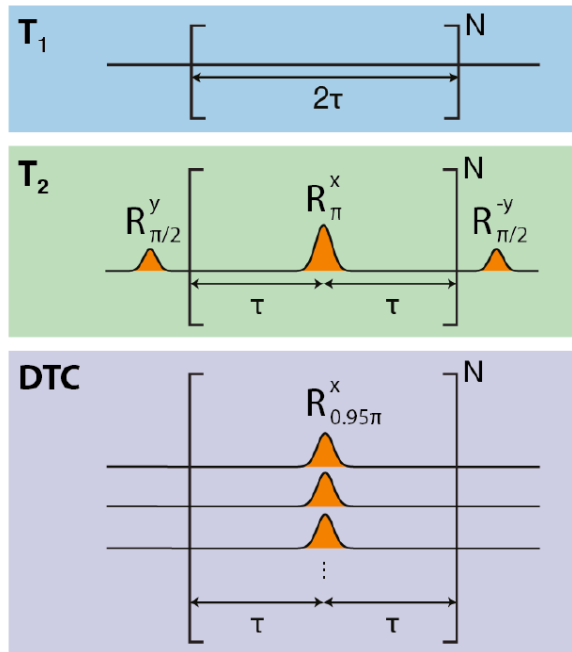
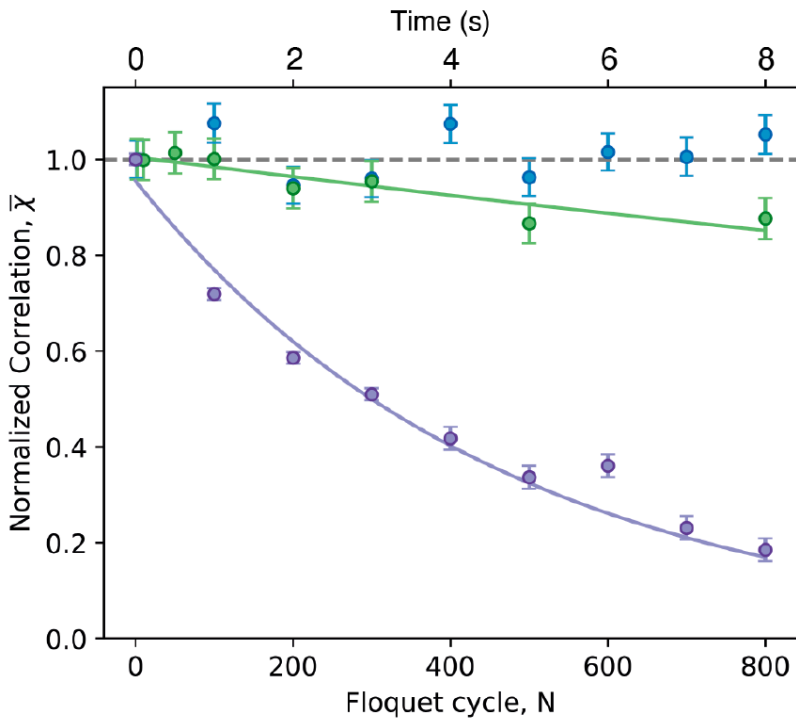
State (in)dependence? (theory)



Our spin chain

Disorder removed

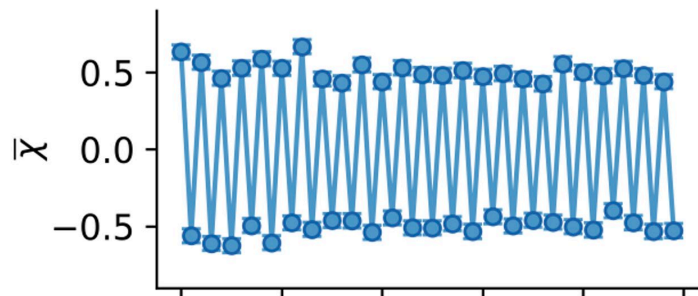
Coherence and where does the observed decay come from



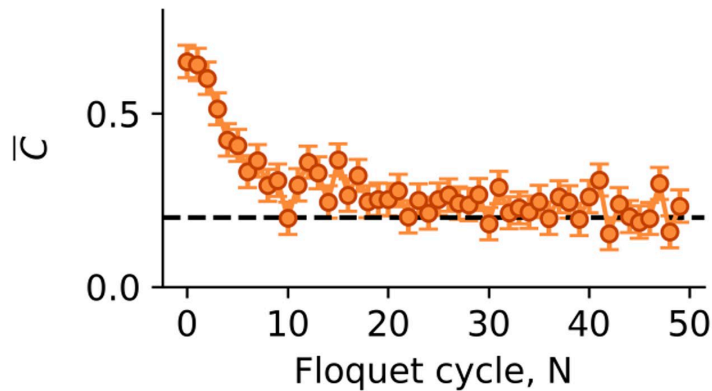
An interacting many-body system (?)

Initial state

$$[\cos(\pi/8) |\uparrow\rangle + \sin(\pi/8) |\downarrow\rangle]^{\otimes 9}$$

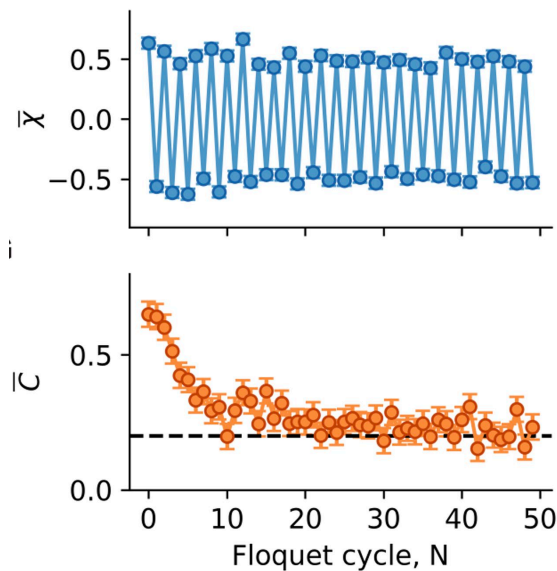
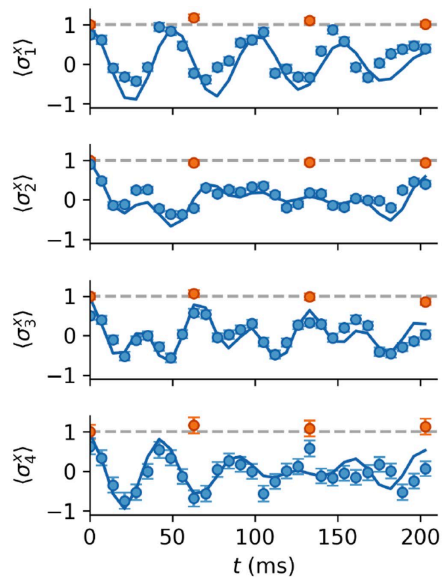
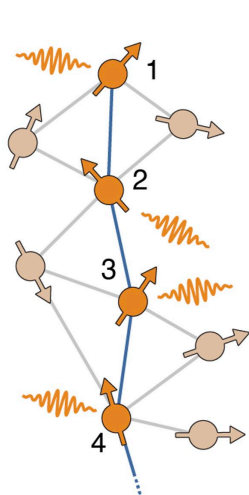


$$\bar{\chi} = \frac{1}{L} \sum_{j=1}^L \langle \sigma_j^z(N) \rangle \text{sgn}[\langle \sigma_j^z(0) \rangle]$$

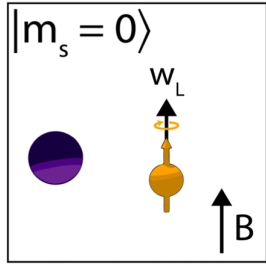


$$\bar{C} = \frac{1}{L} \sum_{j=1}^L \sqrt{\langle \sigma_j^x \rangle^2 + \langle \sigma_j^y \rangle^2}$$

Experiment: An isolated interacting system?



Global Rabi driving



Dipolar Hamiltonian:
$$H_{\text{dip}}^{(j)} = \sum_{\mu, \nu=x,y,z} S_{\mu} \alpha_{\mu\nu}^{(j)} I_{\nu}^{(j)}$$

$$\begin{pmatrix} \alpha_{xx} & \alpha_{xy} & \alpha_{xz} \\ \alpha_{yx} & \alpha_{yy} & \alpha_{yz} \\ \alpha_{zx} & \alpha_{zy} & \alpha_{zz} \end{pmatrix}$$

‘Enhanced’ g-factor:

$$\mathbf{g}^j(m_s) = \begin{pmatrix} 1 & & \\ & 1 & \\ & & 1 \end{pmatrix} - \frac{\mu_e}{\mu_n \Delta} (2 - 3|m_s|) \begin{pmatrix} \alpha_{xx}^{(j)} & \alpha_{xy}^{(j)} & \alpha_{xz}^{(j)} \\ \alpha_{xy}^{(j)} & \alpha_{yy}^{(j)} & \alpha_{yz}^{(j)} \\ 0 & 0 & 0 \end{pmatrix}$$

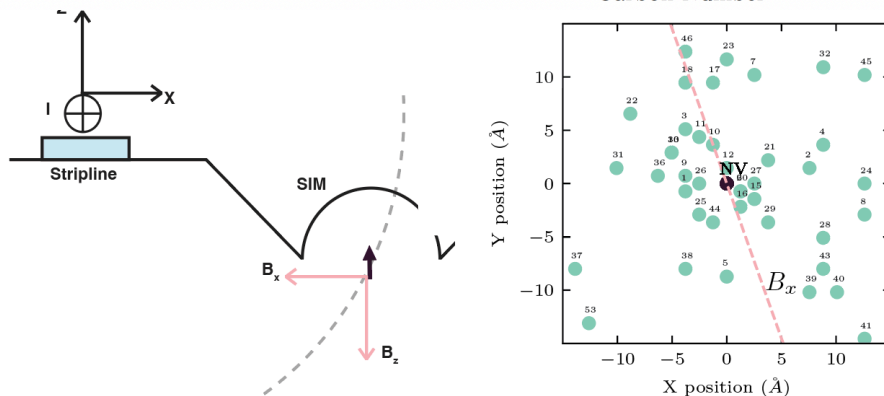
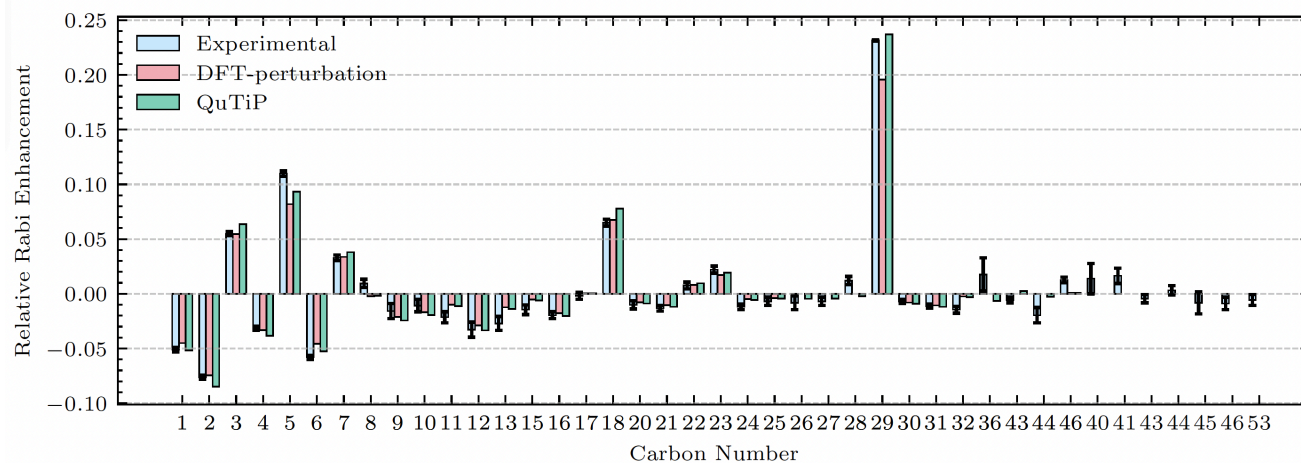
Relative Rabi frequency enhancement (perturbation theory):

$$\frac{\Omega_0}{\Omega} = \sqrt{[\eta \alpha_{xy} \sin \phi + (1 + \eta \alpha_{xx}) \cos \phi]^2 + [\eta \alpha_{xy} \cos \phi + (1 + \eta \alpha_{yy}) \sin \phi]^2}$$

ϕ : polarization RF field

$$\eta = \frac{-2\gamma_e}{\gamma_c \Delta_{ZFS}}$$

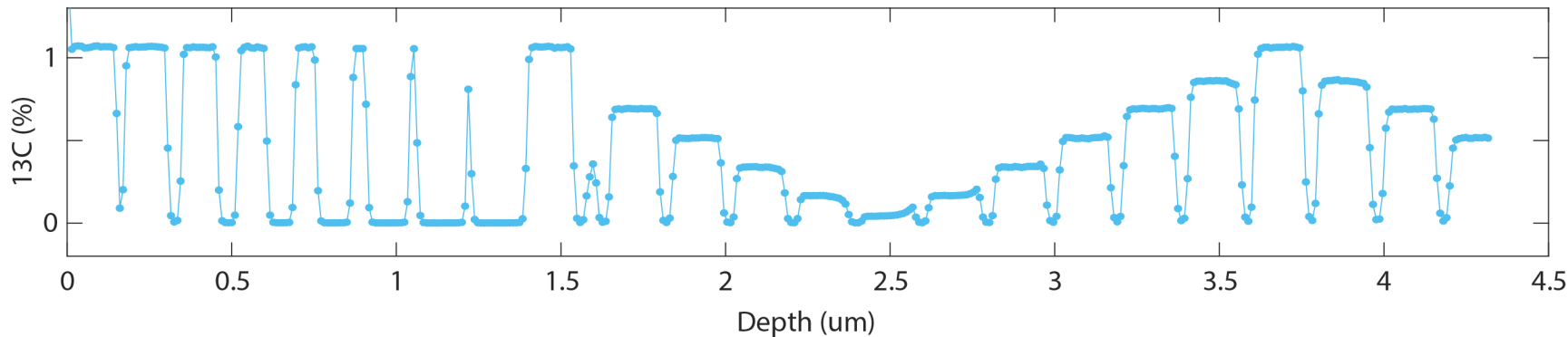
Global Rabi driving: enhanced g-factors



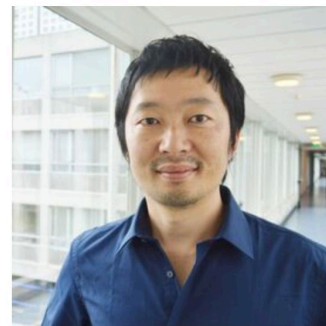
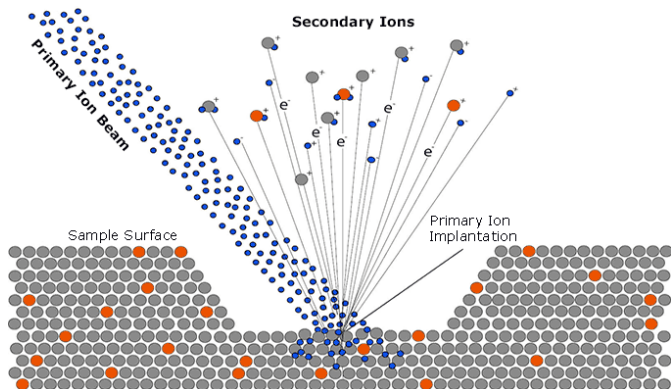
*Just one free parameter:
RF field polarisation!*

Growing diamonds by chemical vapor deposition (CVD)

Layers of different ^{13}C concentration



SIMS measurements



Takashi Yamamoto