Correlation Rényi entropy

Mohamad Niknam

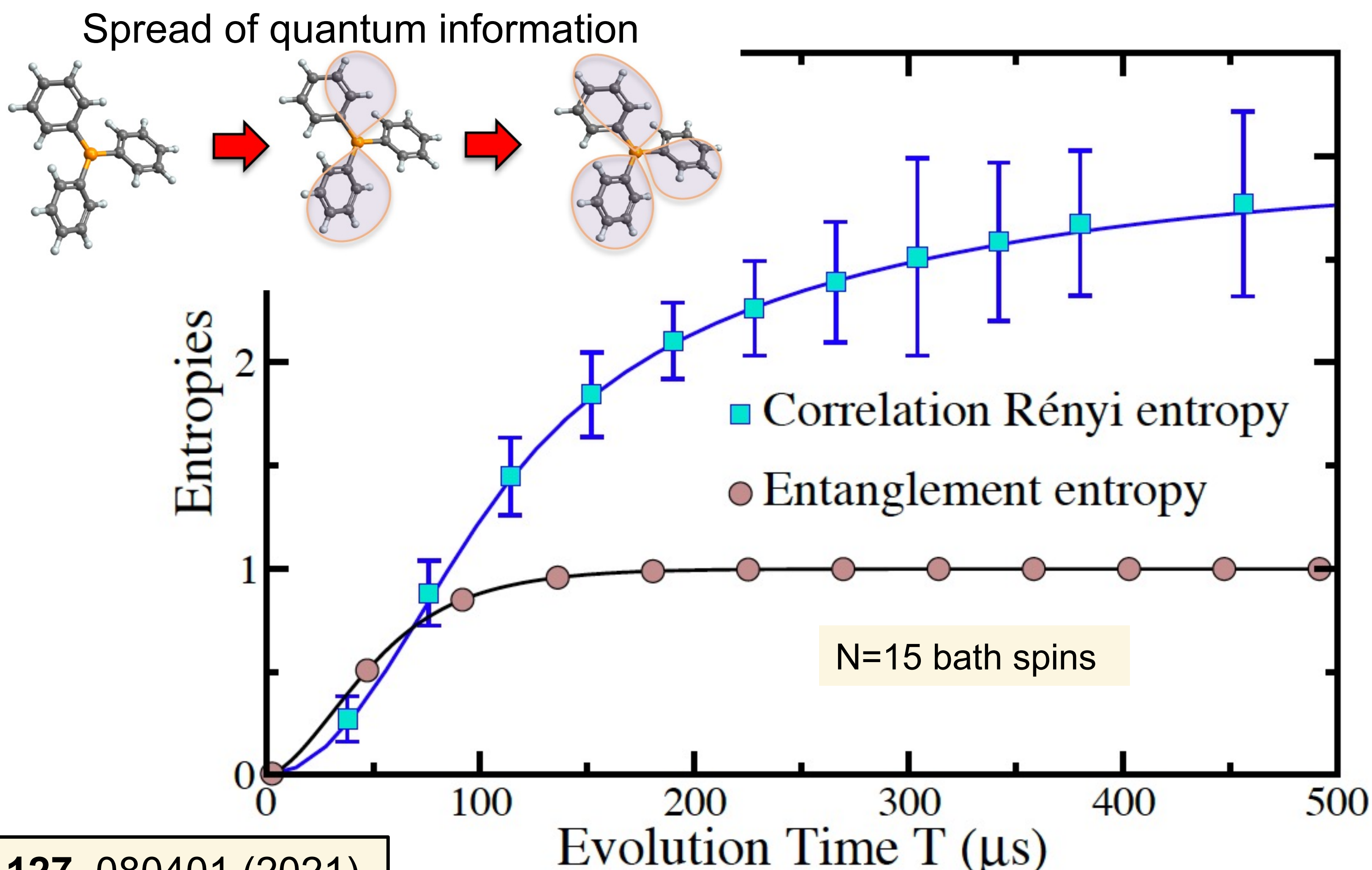
David Cory

NMR experiment

Experimental Detection of the Correlation Rényi Entropy in the Central Spin Model

PRL **127**, 080401 (2021)

Correlation Rényi entropy



PRL **127**, 080401 (2021)

Eigenstates: Degree of Delocalization

$$|\alpha\rangle = \sum_{n=1}^{D} C_n^{\alpha} |n\rangle$$

$$|\alpha\rangle = \begin{pmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \\ c_5 \\ \dots \end{pmatrix}$$

Generalized Inverse Participation Ratio

$$IPR_q^{\alpha} = \sum_n |C_n^{\alpha}|^{2q}$$

Inverse Participation Ratio

$$IPR_{q=2}^{\alpha} = \sum_n |C_n^{\alpha}|^4$$

IPR: large (**localization**)

IPR: small (**delocalization**)

Rényi entropy of order q

$$q > 1$$

$$S_q = \frac{1}{1-q} \log(IPR_q^{\alpha})$$

Entanglement Entropy

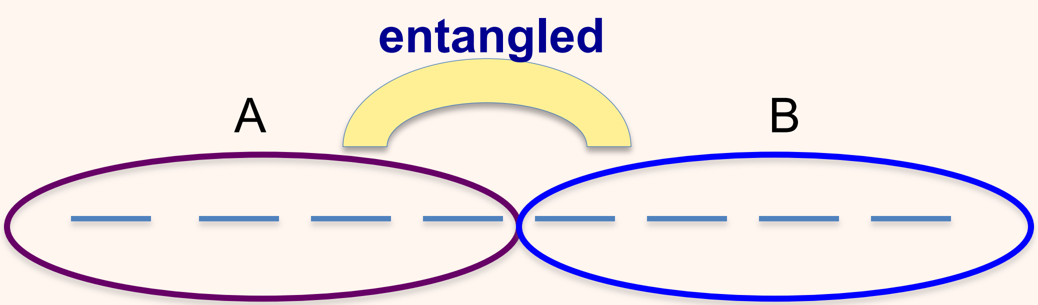
$$|\alpha\rangle = \sum_{n=1}^{D} C_n^{\alpha} |n\rangle$$

$$|\alpha\rangle = \begin{pmatrix} c_1 \\ c_2 \\ c_3 \\ c_4 \\ c_5 \\ \cdots \end{pmatrix}$$

$$\rho = |\alpha\rangle\langle\alpha|$$

$$\rho_A = \mathrm{Tr}_B(\rho)$$

$$S_{ent}^A = -\mathrm{Tr}(\rho_A \ln \rho_A)$$



Delocalization vs Entanglement

This state is **delocalized**, but not **entangled**

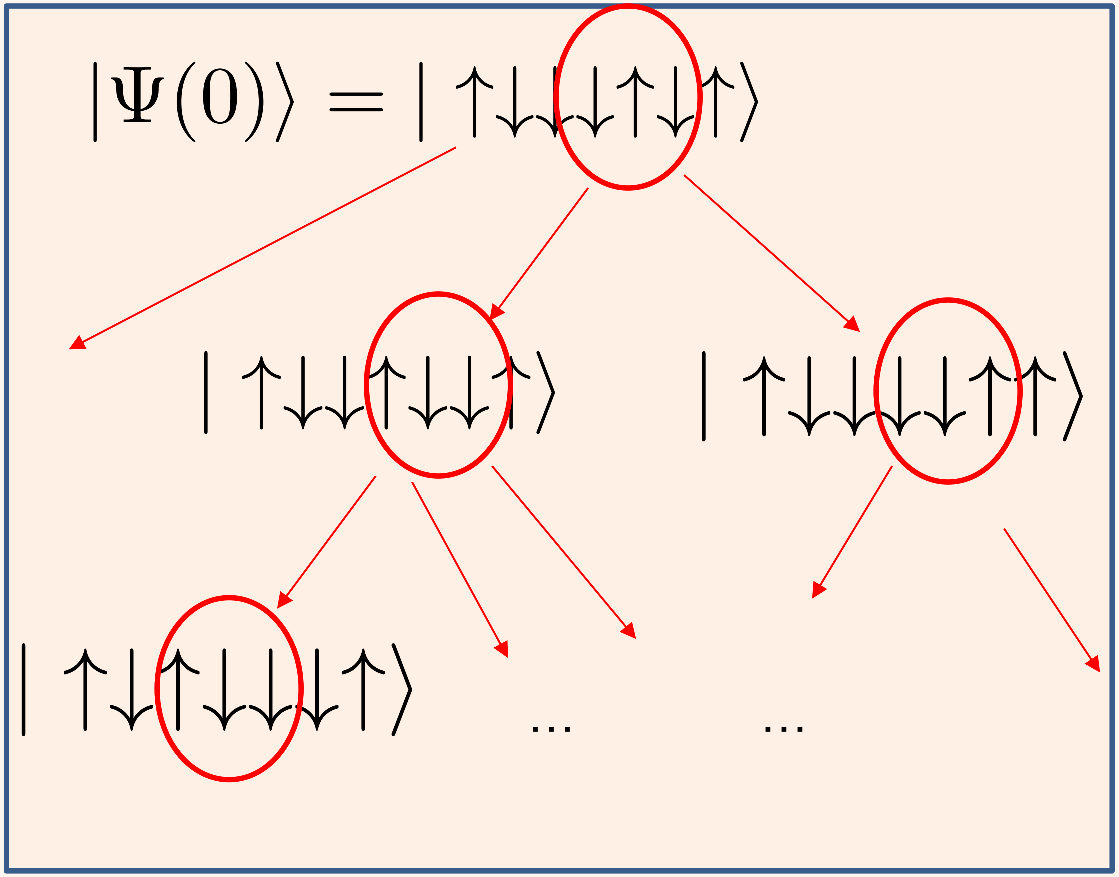
z: $$\frac{1}{2}|11\rangle + \frac{1}{2}|10\rangle + \frac{1}{2}|01\rangle + \frac{1}{2}|00\rangle$$

$$\frac{1}{2}|1\rangle(|1\rangle + |0\rangle) + \frac{1}{2}|0\rangle(|1\rangle + |0\rangle)$$

$$\frac{1}{2}(|1\rangle + |0\rangle)(|1\rangle + |0\rangle)$$

x: $$|+\rangle|+\rangle$$

Spread in the many-body Hilbert space



LFS, Borgonovi, Izrailev
PRL **108**, 094102 (2012)
PRE **85**, 036209 (2012)

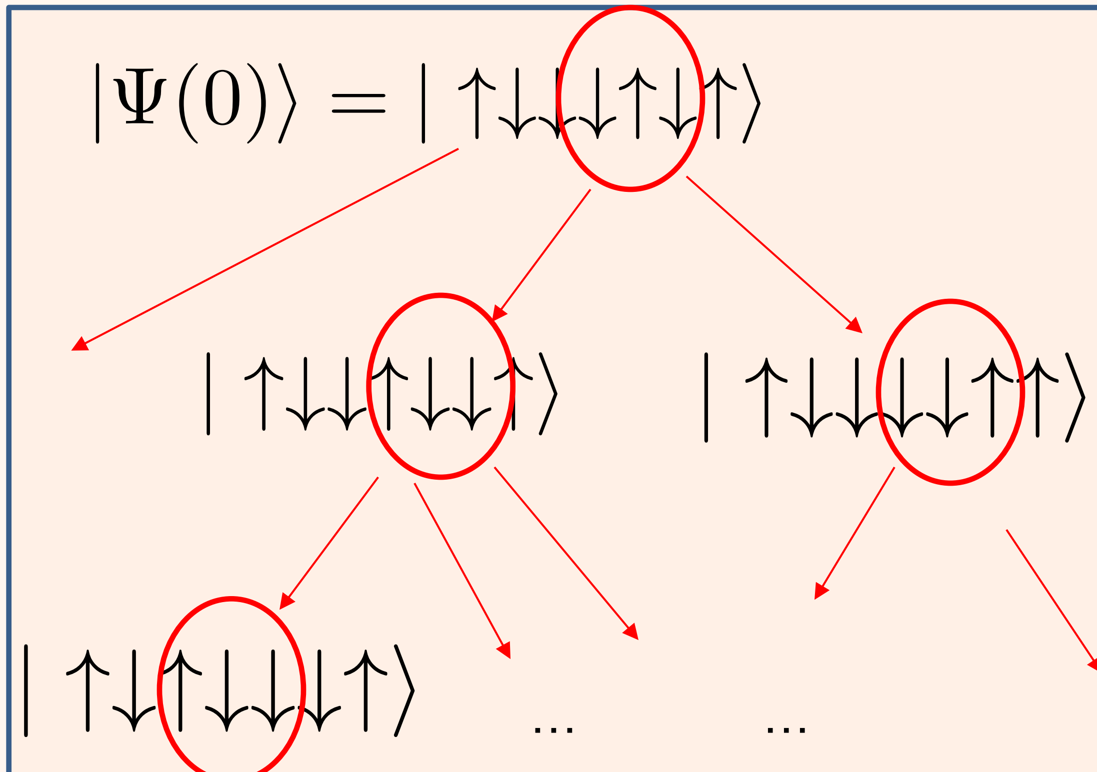
Torres & LFS,
Ann. Phys. **529**, 1600284 (2017)

Rényi entropy of components of ψ(t)

$$|C_k(t)|^2 = |\langle \phi_k | \Psi(t)\rangle|^2$$

$$|\langle \uparrow\uparrow \ldots \downarrow\downarrow | \Psi(t)\rangle|^2$$

$$|\langle \uparrow\downarrow \ldots \uparrow\downarrow | \Psi(t)\rangle|^2$$



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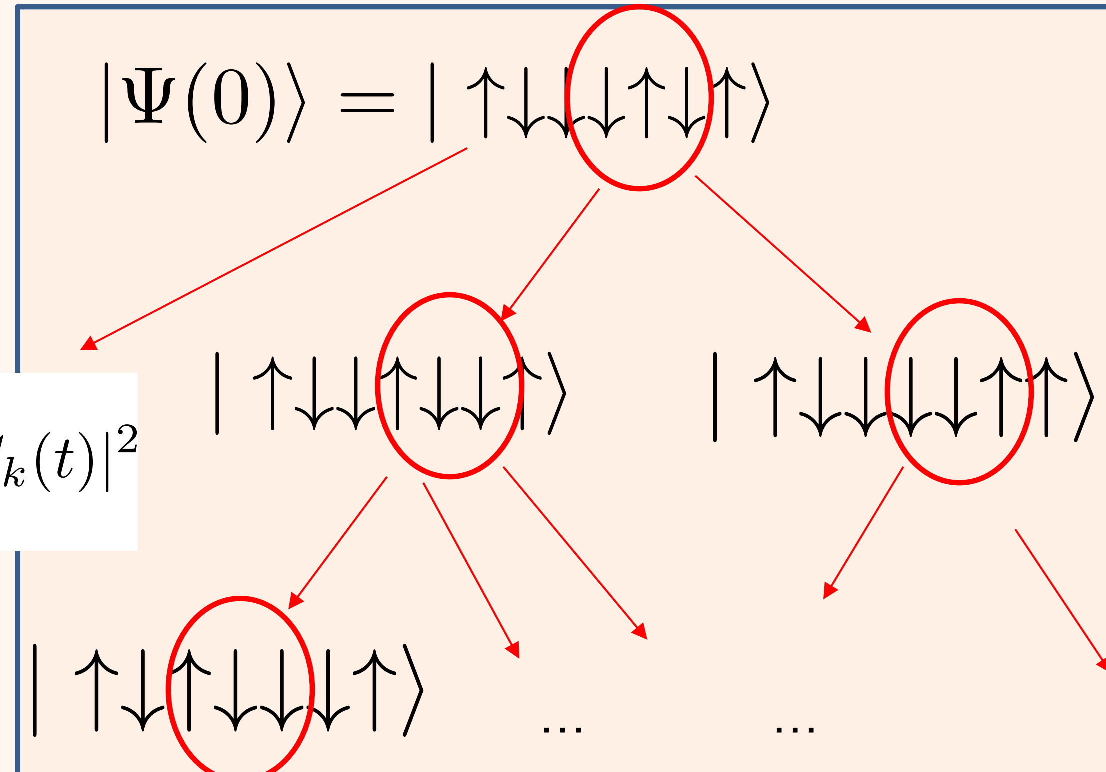

Shannon entropy

$$S_{Sh}(t) = -\sum_{k=1}^{Dim} |C_k(t)|^2 ln |C_k(t)|^2$$

2nd-order Rényi entropy

$$S_2(t) = -ln\left(\sum_{k=1}^{Dim} |C_k(t)|^4\right)$$

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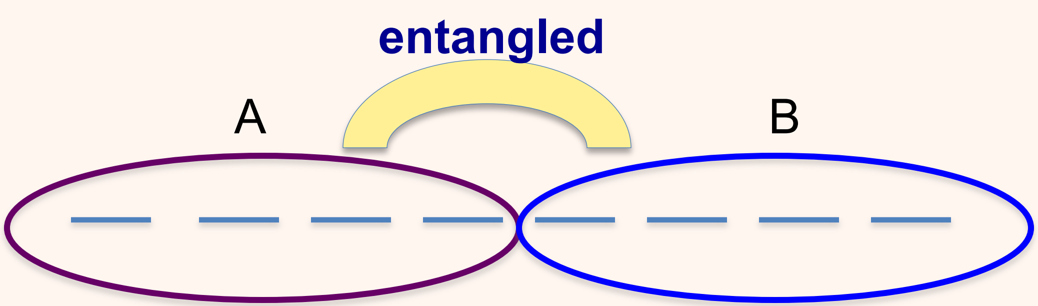
Entanglement entropy

Entanglement entropy: von Neumann entropy of the reduced density matrix

$$\rho_A(t) = \mathrm{Tr}_B\left[\rho(t)\right]$$

$$\boxed{S_{ent}^A = -\mathrm{Tr}(\rho_A \ln \rho_A)} = S_{ent}^B = -\mathrm{Tr}(\rho_B \ln \rho_B)$$



Entanglement entropy: Superconducting qubits

nature physics | LETTERS
PUBLISHED ONLINE: 11 JULY 2016 | DOI: 10.1038/NPHYS3830

Ergodic dynamics and thermalization in an isolated quantum system

C. Neill[1*†], P. Roushan[2†], M. Fang[1†], Y. Chen[2†], M. Kolodrubetz[3], Z. Chen[1], A. Megrant[2], R. Barends[2], B. Campbell[1], B. Chiaro[1], A. Dunsworth[1], E. Jeffrey[2], J. Kelly[2], J. Mutus[2], P. J. J. O'Malley[1], C. Quintana[1], D. Sank[2], A. Vainsencher[1], J. Wenner[1], T. C. White[2], A. Polkovnikov[3] and J. M. Martinis[1,2]

Driven system

$$U = e^{-i(\kappa/2j\hbar)J_z^2} e^{-i(\pi/2\hbar)J_y}$$

Use state tomography to reconstruct the density matrix

$$S = -\mathrm{Tr}\,\rho_{sq}\log_2(\rho_{sq})$$

ρ_{sq} is density matrix of a single qubit

Quantum system of **only three** superconducting qubits.

$$\mathcal{H}(t) = \frac{\pi}{2\tau}J_y + \frac{\kappa}{2j}J_z^2 \sum_{n=1}^{N} \delta(t - n\tau)$$

$$J_z = (\hbar/2)\sum_i \sigma_z^{(i)}$$

a three-qubit ring of planar transmons with tunable interqubit coupling

Each qubit is initially in a coherent state

$$|\theta_0, \varphi_0\rangle = \cos(\theta_0/2)|+\sigma_z\rangle + e^{-i\varphi_0}\sin(\theta_0/2)|-\sigma_z\rangle$$

Nat. Phys. **12**,1037 (2016)

Entanglement entropy: Superconducting qubits

nature physics

LETTERS

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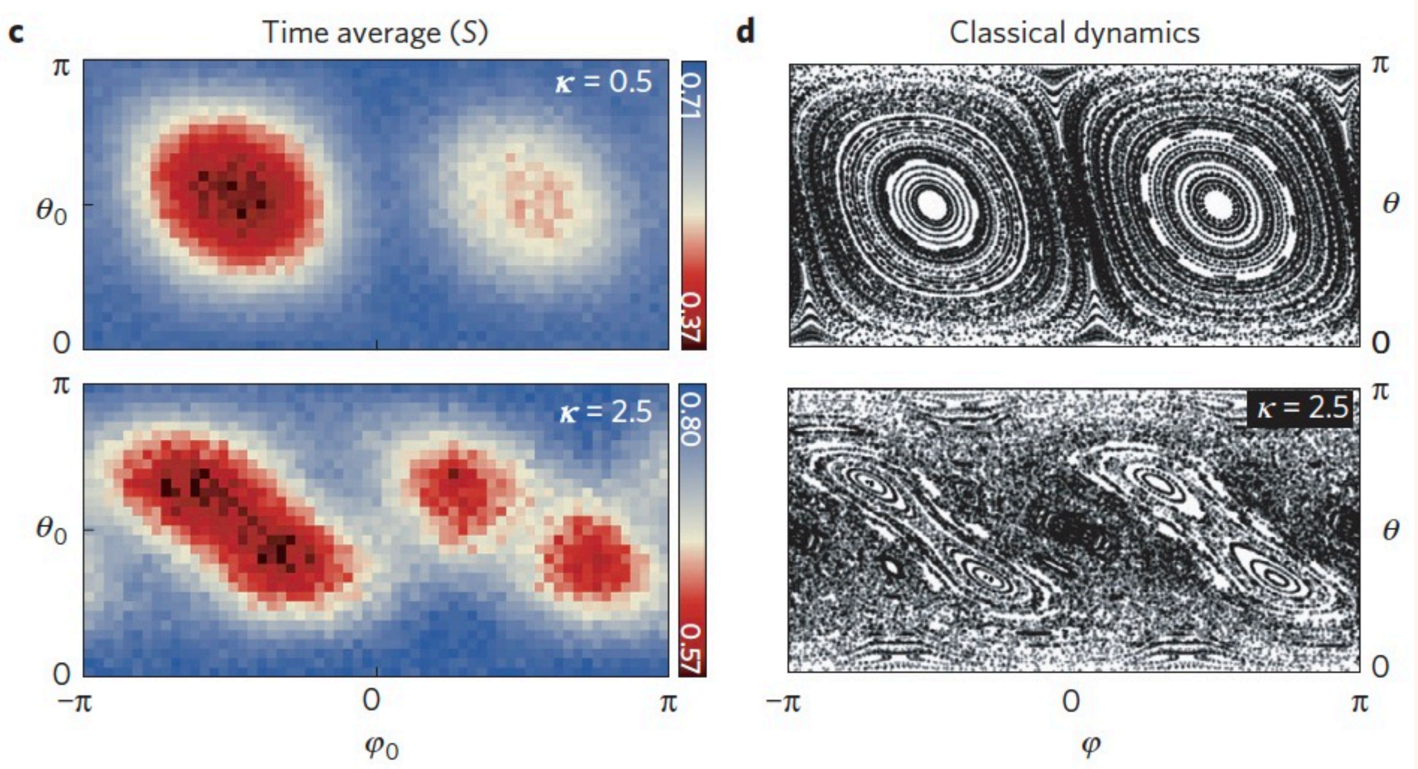


angles describing
the orientation of
the single-qubit states

Entanglement entropy: Cold atoms

Quantum thermalization through entanglement in an isolated many-body system

Adam M. Kaufman, M. Eric Tai, Alexander Lukin, Matthew Rispoli, Robert Schittko, Philipp M. Preiss, Markus Greiner*

Science **353**,794 (2016)

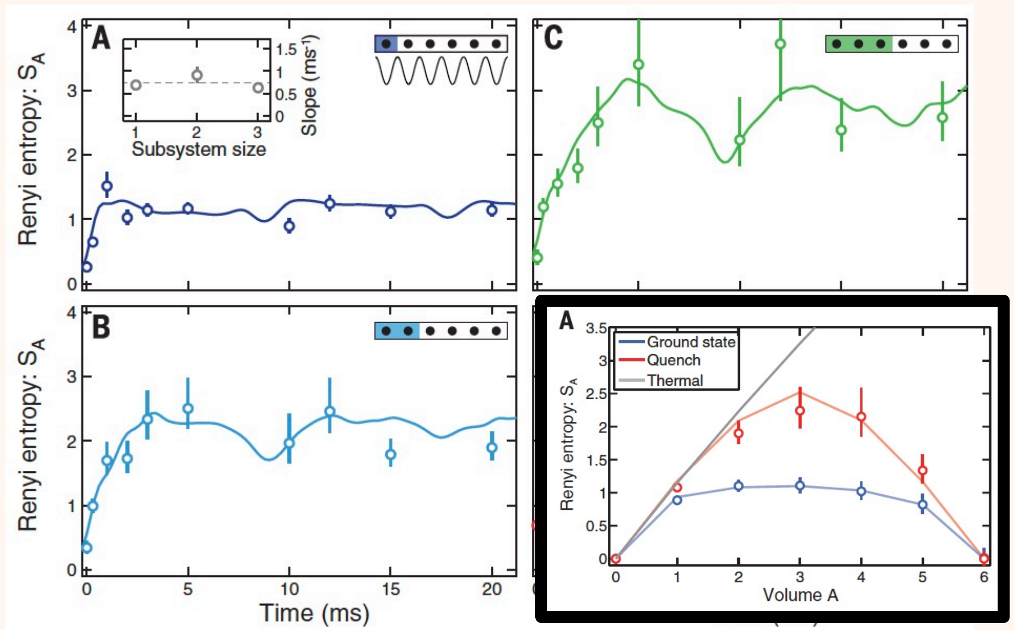
They can measure the square of the (reduced) density matrix

$$S_A(t) = -\ln\left[\mathrm{Tr}(\rho_A^2(t))\right]$$

Quantum system of <u>**only six**</u> sites.

$$H = -J\sum_i a_i^\dagger a_{i+1} + \frac{U}{2}\sum_i n_i(n_i - 1)$$

Each site has initially a single cold atom.
Dynamics starts by turning the hopping J on.



Entanglement entropy: Ion traps

Probing Rényi entanglement entropy via randomized measurements

Tiff Brydges[1,2*], Andreas Elben[1,2*], Petar Jurcevic[1,2], Benoît Vermersch[1,2], Christine Maier[1,2], Ben P. Lanyon[1,2], Peter Zoller[1,2], Rainer Blatt[1,2], Christian F. Roos[1,2†]

Science **364**, 260 (2019)

Their reduced density matrix can have from 1 to 10 qubits.

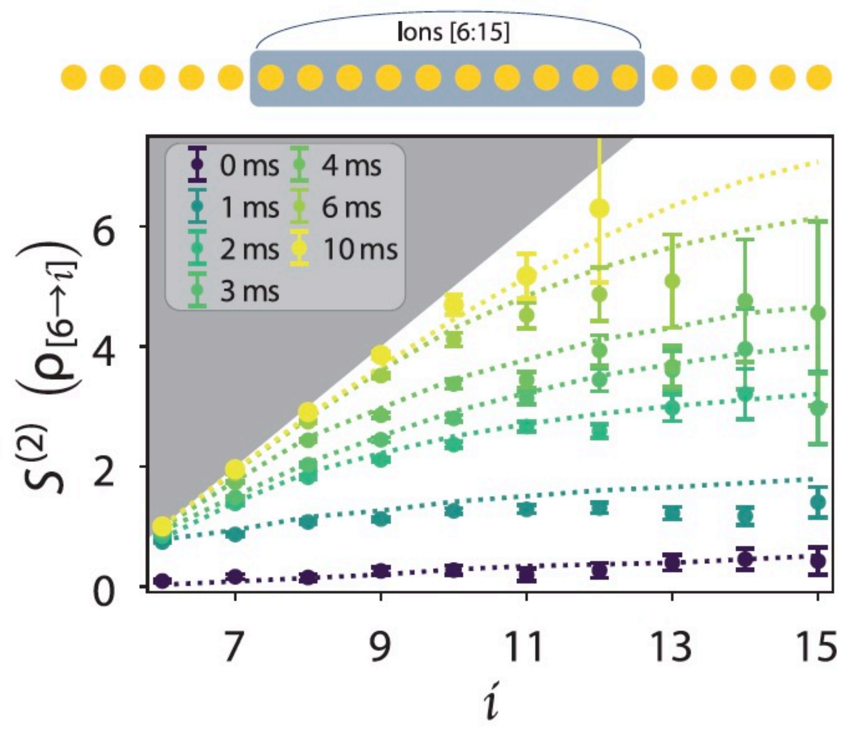
$$S_A(t) = -\ln\left[\mathrm{Tr}(\rho_A^2(t))\right]$$

Strings of up to **20** trapped $^{40}Ca^+$ ions.

$$H_{XY} = \hbar \sum_{i<j} J_{ij}(\sigma_i^+\sigma_j^- + \sigma_i^-\sigma_j^+) + \hbar B \sum_j \sigma_j^z$$

$$J_{ij} \approx J_0/|i-j|^\alpha \qquad 0 < \alpha < 3$$

Initial state: Néel



Spread of quantum information: OTOC

$$W(t) = e^{iHt} W e^{-iHt}$$

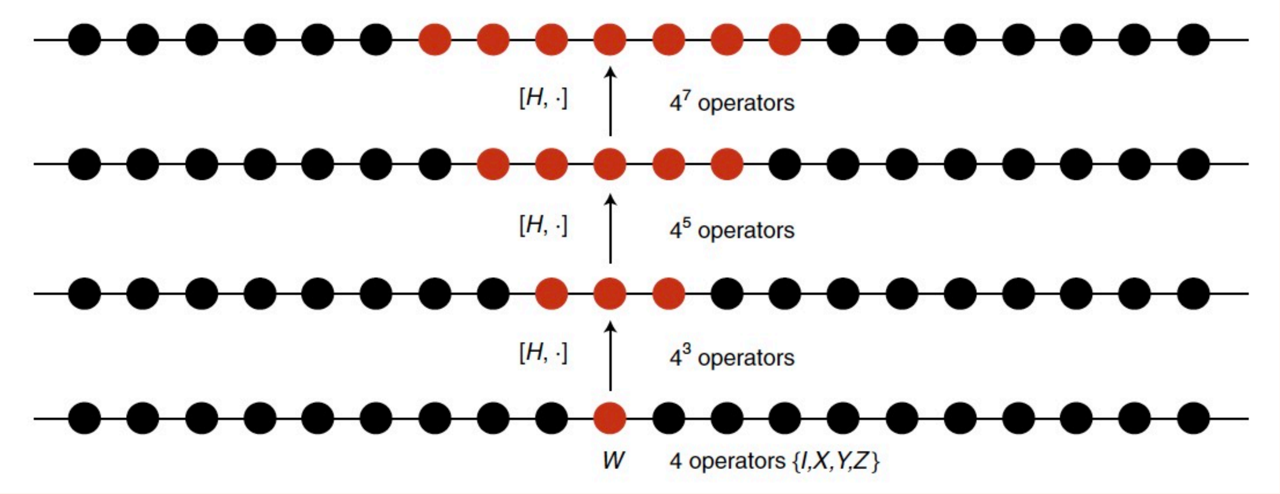
$$C(t) = \langle |[W(t), V(0)]|^2 \rangle$$

W is a local operator, e.g. spin operator on a site.

(nested commutators)

W(t) spreads over many sites

$$W(t) = \sum_{\ell=0}^{\infty} \frac{(it)^{\ell}}{\ell!} [H, \ldots [H, W], \ldots]$$



OTOC
measures this growth

$$C(t) = \langle [W(t), V]^{\dagger} [W(t), V] \rangle$$

B. Swingle
Nat. Phys. **14**, 988 (2018)

Spread of quantum information: Correlation Rényi entropy

$$\rho(0) = \sigma_x^{CS} \otimes \mathbb{I}^{\otimes N}$$

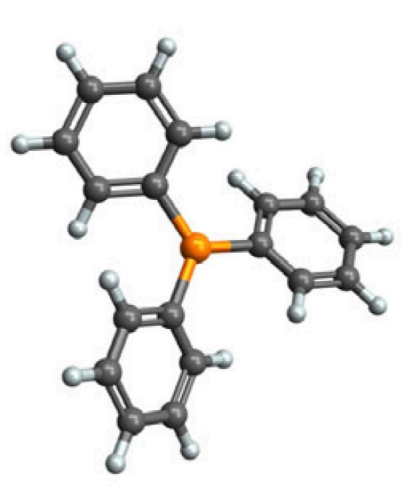
$$\rho(T) = e^{-iH_{\text{CSB}}T}\rho(0)e^{iH_{\text{CSB}}T}$$
$$= \rho(0) + iT[\rho(0), H_{\text{CSB}}]$$
$$- \frac{T^2}{2}\Big[[\rho(0), H_{\text{CSB}}], H_{\text{CSB}}\Big] + \cdots$$

$$\sigma_y^{CS} \otimes \sigma_z^j \otimes \mathbb{I}^{\otimes N-1}$$

$$\sigma_y^{CS} \otimes \sigma_z^j \otimes \sigma_z^k \otimes \mathbb{I}^{\otimes N-2}$$

The **correlation Rényi entropy** quantifies the buildup of **multispin correlations**.

Central Spin-1/2 Model



The Triphenylphosphine molecule has a ^{31}P nucleus at the central spin position and fifteen ^{1}H nuclei as the surrounding spins.

The total NMR signal is induced by an ensemble of molecules with more than 10^{17} spins.

Lea F. Santos

Central spin (CS) coupled with 15 bath spins

Heteronuclear dipolar interaction (~1.2 kHz)

$$H_{\mathrm{CSB}} = \sum_{j}^{N} \omega_j \sigma_z^{\mathrm{CS}} \otimes \sigma_z^j \otimes \mathbb{1}^{\otimes N-1}$$

$$\omega_j \propto (3\cos^2\theta_j - 1)/r_j^3$$

Coupling between bath (B) spins

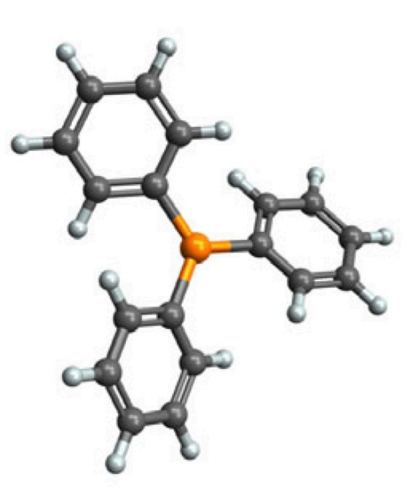
Homonuclear dipolar interaction (<20 kHz)

$$\mathcal{H}_{\mathrm{B}} = \mathbb{1}^{\mathrm{cs}} \otimes \sum_{j<k}^{N} \Omega_{jk} \left[\sigma_Z^j \sigma_Z^k - \frac{1}{4}(\sigma_+^j \sigma_-^k + \sigma_-^j \sigma_+^k) \right]$$

Experimental Detection of the Correlation Rényi Entropy in the Central Spin Model

PRL **127**, 080401 (2021)

Central Spin-1/2 Model



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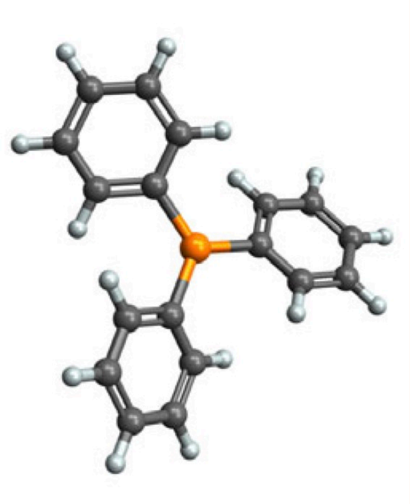

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$$\omega_j \propto (3\cos^2\theta_j - 1)/r_j^3$$

- Room temperature.
- Good isolation from an external bath.
- The central spin is initially polarized and coupled with the 15 unpolarized surrounding spins.

Experimental Detection of the Correlation Rényi Entropy in the Central Spin Model

PRL **127**, 080401 (2021)

Evolution under the ZZ interaction

$$\rho(0) = \rho^{\text{CS}}(0) \otimes \rho^{B}(0)$$

$$\rho^{\text{CS}}(0) = (\mathbb{1} + \epsilon\sigma_x)/2$$

Fully mixed:

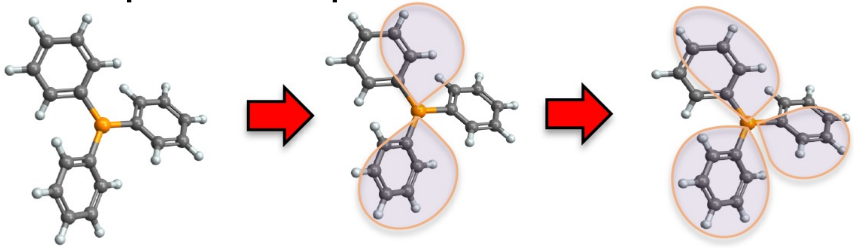
$$\rho^{B}(0) = (\mathbb{1}/2)^{\otimes N}$$

$$H_{\text{CSB}} = \sum_{j}^{N} \omega_j \sigma_z^{\text{CS}} \otimes \sigma_z^j \otimes \mathbb{1}^{\otimes N-1}$$

$$\rho(T) = U_{\text{CSB}}(T)\rho(0)U_{\text{CSB}}^{\dagger}(T)$$

As the CSB system evolves under the ZZ interaction, information from the central spin gets shared with the bath spins.

Spread of quantum information



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FID and entanglement entropy

The loss of information from the central spin can be quantified with the

- Free induction decay (FID):

$$\mathrm{FID}(T) = \mathrm{Tr}\{\sigma_x^{\mathrm{CS}} \rho^{\mathrm{CS}}(T)\} = \frac{\epsilon}{2^{N+1}} \sum_{k=1}^{2^{N+1}} \cos(2\langle \varphi_k | H_{\mathrm{CSB}} | \varphi_k \rangle T)$$

$$|\uparrow\downarrow\downarrow\ldots\uparrow\rangle$$

$$\rho^{CS}(T) = \mathrm{Tr}_B[\rho(T)]$$

FID and entanglement entropy

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$\rho^{CS}(T) = \mathrm{Tr}_B[\rho(T)]$

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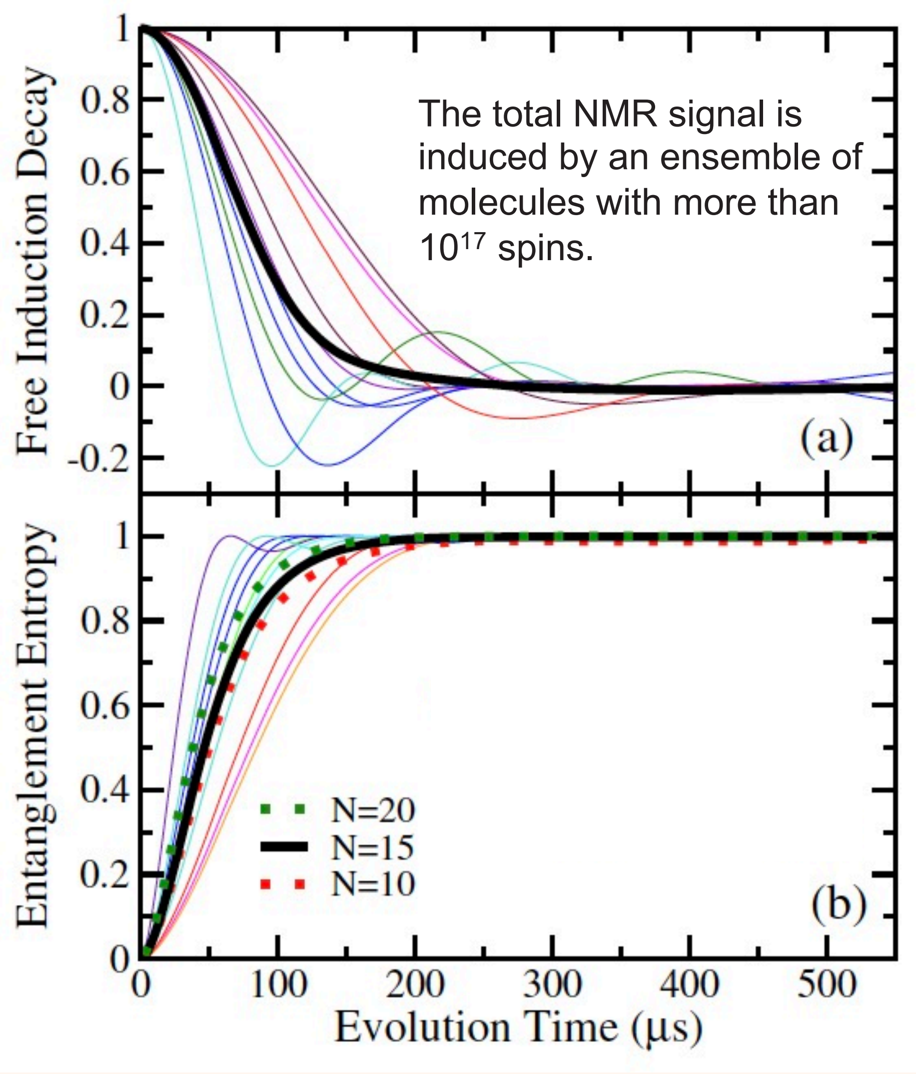
$|\uparrow\downarrow\downarrow\ldots\uparrow\rangle$

- Entanglement entropy

$$\begin{aligned} S_{\mathrm{ent}}(T) &= -\mathrm{Tr}\{\rho^{\mathrm{CS}}(T) \log_2[\rho^{\mathrm{CS}}(T)]\} \\ &= -[f^{+}(T) \log_2 f^{+}(T) + f^{-}(T) \log_2 f^{-}(T)] \end{aligned}$$

$$f^{\pm}(T) = (1/2) \pm \mathrm{FID}(T)/2$$

FID and entanglement entropy



Thin lines are for each realization of sets of ω_j,
and
thick line gives the average over 300 random orientations of the molecules.

$$H_{\mathrm{CSB}} = \sum_{j}^{N} \omega_j \sigma_z^{\mathrm{CS}} \otimes \sigma_z^j \otimes \mathbb{1}^{\otimes N-1}$$

Multispin correlations

As the CSB system evolves under the ZZ interaction, information from the central spin gets shared with the bath spins giving rise to clusters of multispin correlations.

$$\begin{aligned}\rho(T) &= e^{-iH_{\mathrm{CSB}}T}\rho(0)e^{iH_{\mathrm{CSB}}T} \\ &= \rho(0) + iT[\rho(0), H_{\mathrm{CSB}}] \\ &\quad - \frac{T^2}{2}\big[[\rho(0), H_{\mathrm{CSB}}], H_{\mathrm{CSB}}\big] + \cdots\end{aligned}$$

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$$= \rho(0) + iT[\rho(0), H_{\text{CSB}}]$$
$$- \frac{T^2}{2}\big[[\rho(0), H_{\text{CSB}}], H_{\text{CSB}}\big] + \cdots$$

$$\rho^B(0) = (\mathbb{1}/2)^{\otimes N}$$

$$\rho^{\text{CS}}(0) = (\mathbb{1} + \epsilon\sigma_x)/2$$

$$H_{\text{CSB}} = \sum_j^N \omega_j \sigma_z^{\text{CS}} \otimes \sigma_z^j \otimes \mathbb{1}^{\otimes N-1}$$

$$[\sigma_x^{CS}, \sigma_z^{CS} \otimes \sigma_z^j] = -2i\sigma_y^{CS} \otimes \sigma_z^j$$

The commutators above lead to terms with a different number m of nonidentity bath spins operators.

Correlation Rényi entropy

As the CSB system evolves under the ZZ interaction, information from the central spin gets shared with the bath spins giving rise to clusters of multispin correlations.

$$\rho(T) = \frac{1}{2^{N+1}} \Big\{ \mathbb{1}^{\otimes N+1} + \epsilon C_0^z(T) \sigma_x^{\mathrm{CS}} \otimes \mathbb{1}^{\otimes N} + \epsilon \sum_j^N C_1^{z(j)}(T) \sigma_y^{\mathrm{CS}} \otimes \sigma_z^j \otimes \mathbb{1}^{\otimes N-1} + \epsilon \sum_{j_1<j_2}^N C_2^{z(j_1 j_2)}(T) \sigma_x^{\mathrm{CS}} \otimes \sigma_z^{j_1} \otimes \sigma_z^{j_2} \otimes \mathbb{1}^{\otimes N-2} + \cdots \Big\}$$

$$S_1(t) = -\sum_{k=0}^{15} |C_k(t)|^2 log_2 |C_k(t)|^2$$

Our entropy quantifies the volume of correlations among qubits.

Correlation Rényi vs entanglement entropy

$$S_{\text{ent}}(T) = -\text{Tr}\{\rho^{\text{CS}}(T)\log_2[\rho^{\text{CS}}(T)]\}$$

$$\rho^{CS}(T) = \text{Tr}_B[\rho(T)]$$

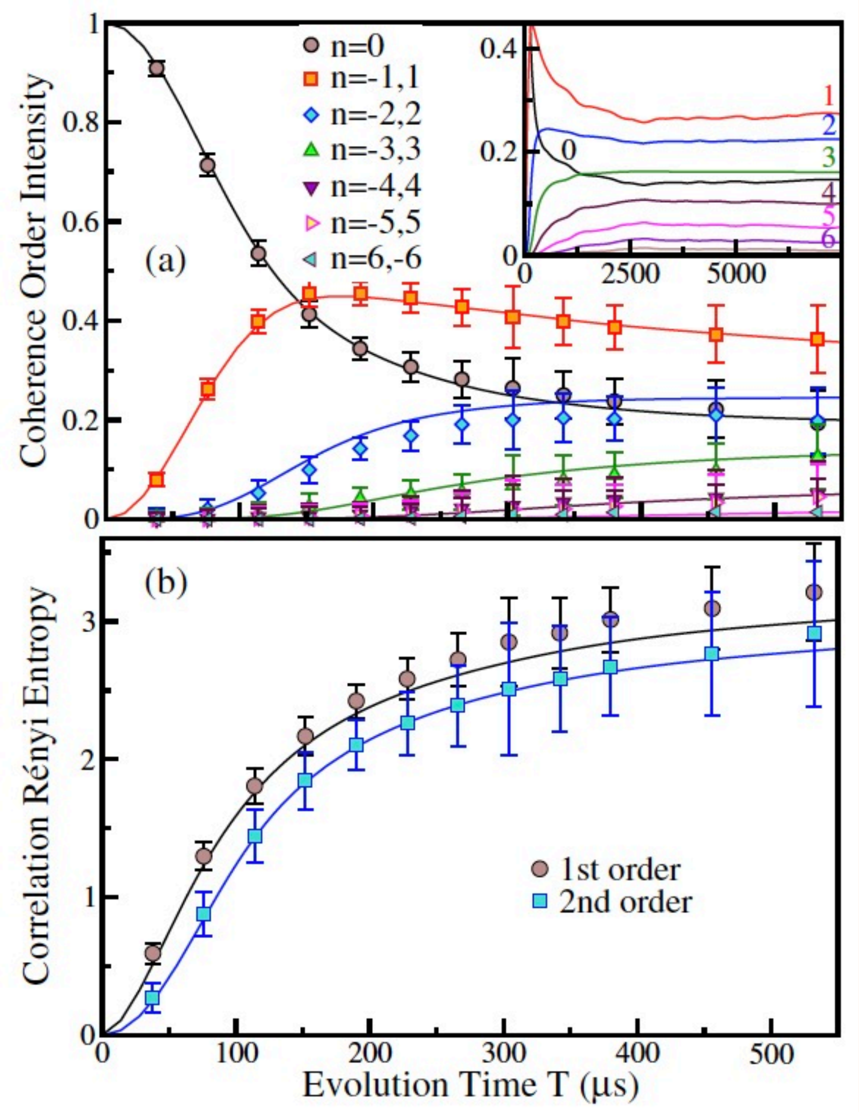
The uncorrelated term with amplitude **$C_0^z(T)$** is the only one that survives the partial trace used to obtain $\rho^{CS}(T)$, thus the only one that contributes to **FID**(T) and **entanglement entropy**.

The decay of $C_0^z(T)$ describes the loss of information from the central spin,
causes the decline of the observable NMR signal
the growth of the entanglement entropy.

Our entropy captures the **buildup of multispin correlations**
as determined by the higher order terms with $m > 0$.

$$S_1(t) = -\sum_{k=0}^{15} |C_k(t)|^2 log_2 |C_k(t)|^2$$

Correlation Rényi entropy



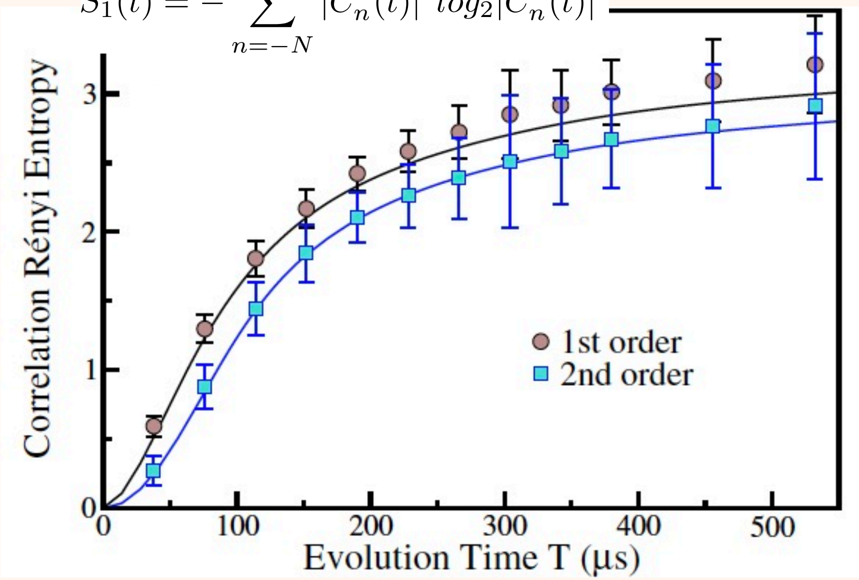
$$S_1 = -\sum_{n=-N}^{N} |C_n^x(T)|^2 \log_2 |C_n^x(T)|^2,$$

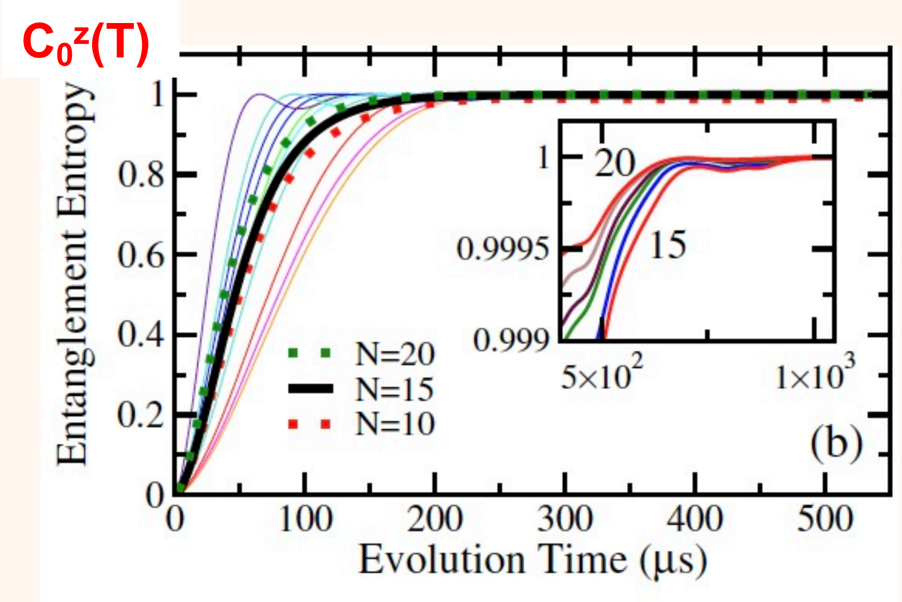
$$S_2 = -\log_2 \left(\sum_{n=-N}^{N} |C_n^x(T)|^4 \right).$$

Correlation Rényi entropy vs Entanglement entropy

Correlation Rényi entropy
keeps **growing** even after the entanglement entropy has already saturated.

$$S_1(t) = -\sum_{n=-N}^{N} |C_n^x(t)|^2 log_2 |C_n^x(t)|^2$$





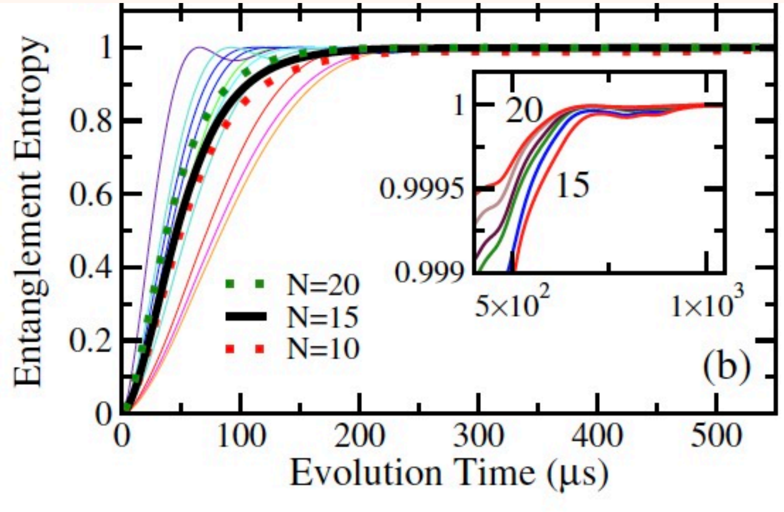
Entanglement entropy quantifies the loss of information from the central spin.

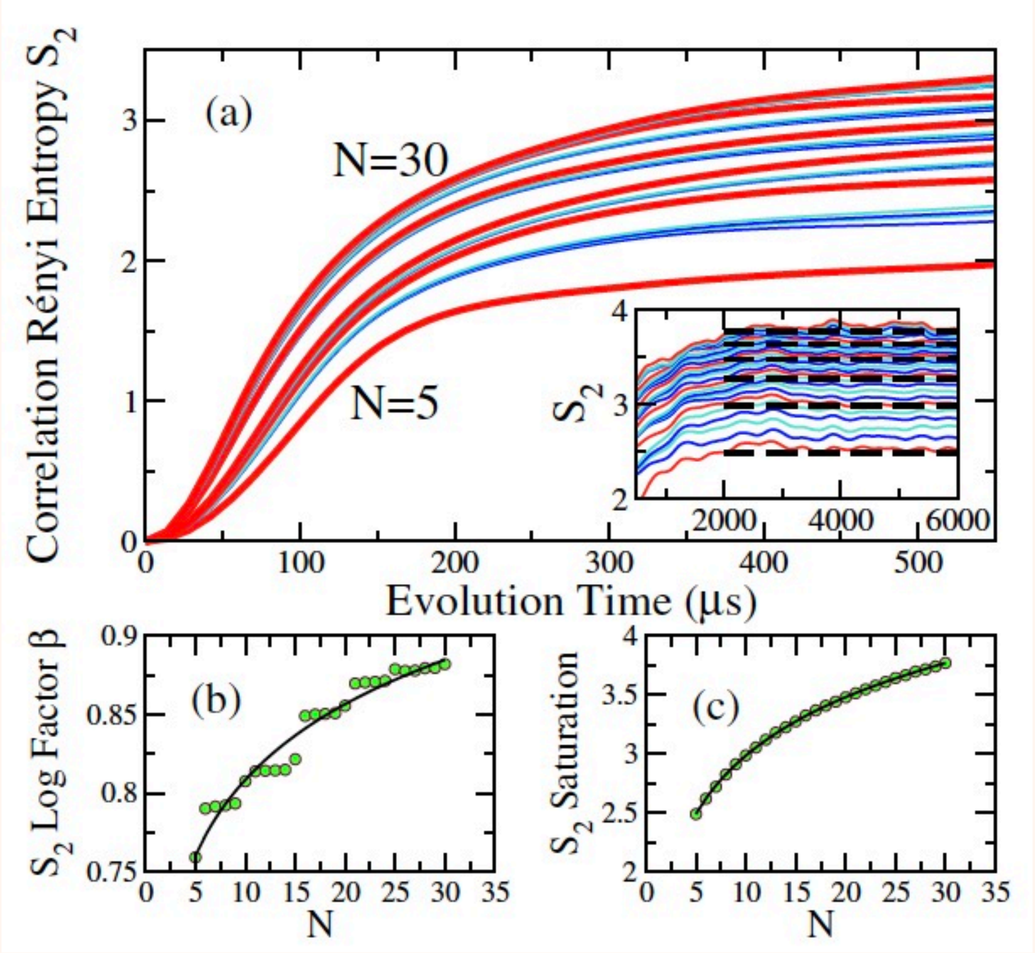
Correlation Rényi entropy measures the **volume of multispin correlations**,
captures how that **information gets shared** between
the central spin and the bath spins through the correlations;
provides a more detailed picture of the dynamics of the composite system.

Timescales

$$T_{eq} = (2052.1 \pm 163.7)\ \mu s$$

Equilibration time for the **correlation Rényi entropy** is nearly independent of the system size.





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Equilibration time?

Correlation Rényi entropy saturates at a time that is 1 order of magnitude larger than the saturation time for entanglement entropy.
Equilibration time is nearly independent of the bath size.

PRL **127**, 080401 (2021)

Equilibration time depends on the **quantity** and on dynamical features considered.

EXTRA SLIDES

The experiment: x-basis

The experiment cannot directly measure $C_z^{(m)}(T)$.
The experiment can directly measure the coherence order intensities $C_x^{(n)}(T)$.

Write the bath spin operators in terms of coherence raising and lowering operators in the x-quantization basis

$$\sigma_{\pm}^{x} := (\sigma_y \pm i\sigma_z)/\sqrt{2}$$

$$\rho(T) = \frac{1}{2^{N+1}}\Big\{ \mathbb{1}^{\otimes N+1} + \epsilon \sum_k C_0^{x,k}(T)\rho_0^{x,k} + \epsilon \sum_k [C_{+1}^{x,k}(T)\rho_{+1}^{x,k} + C_{-1}^{x,k}(T)\rho_{-1}^{x,k}] + \epsilon \sum_k [C_{+2}^{x,k}(T)\rho_{+2}^{x,k} + C_{-2}^{x,k}(T)\rho_{-2}^{x,k}] + \cdots \Big\}$$

The stages of the experiment

The coherence detection technique exploits the collective response of the spins.

$$\rho_\phi(2T) = U^\dagger_{CSB}(T) R_x(\phi) U_{CSB}(T) \rho(0) U^\dagger_{CSB}(T) R^\dagger_x(\phi) U_{CSB}(T)$$

$$R_x(\phi) = \exp\left(i(\phi/2) \sum_j \mathbb{1}^{\mathrm{CS}} \otimes \mathbb{1}^1 \otimes \cdots \otimes \sigma_x^j \otimes \cdots \otimes \mathbb{1}^N\right)$$

Intensities encoded in the phase

The coherence detection technique exploits the collective response of the spins.

$$\rho_\phi(2T) = U^\dagger_{CSB}(T) R_x(\phi) U_{CSB}(T) \rho(0) U^\dagger_{CSB}(T) R^\dagger_x(\phi) U_{CSB}(T)$$

$$R_x(\phi) = \exp\left(i(\phi/2) \sum_j \mathbb{1}^{\rm CS} \otimes \mathbb{1}^1 \otimes \cdots \otimes \sigma_x^j \otimes \cdots \otimes \mathbb{1}^N\right)$$

$$\rho_\phi(2T) = \frac{1}{2^{N+1}} \left\{ \mathbb{1}^{\otimes N+1} + \epsilon \sum_{n=-N}^{N} e^{in\phi} |C_n^x(T)|^2 \sigma_x^{\rm CS} \otimes \mathbb{1}^{\otimes N} \right\}$$

$$S(2T) = \mathrm{Tr}[\mathrm{Tr}_B[\rho_\phi(2T)] \sigma_x^{\rm CS}]$$

The signal is recorded for various increments of rotation angle $\phi \in [0, 2\pi]$.
By performing a Fourier transform of this array of observed signals,
one gets the intensities

$$|C_n^x(T)|^2$$

$$\sum_{n=-N}^{N} |C_n^x(T)|^2 = 1$$

Lea F. Santos

Dynamical decoupling

$$H = J\sigma_1^z \sigma_2^z$$

$$U(T) = e^{-iJ\sigma_1^z\sigma_2^z T}$$

$$U(2T) = P_x e^{-iJ\sigma_1^z\sigma_2^z T} P_x e^{-iJ\sigma_1^z\sigma_2^z T}$$

π-pulse:
instantaneous rotation of spin 1
of 180^o around the x-axis

$$U(2T) = -\mathbb{I}$$

$$P_x = e^{-i\pi S_1^x}$$

$$P_x = e^{-i\frac{\pi}{2}\sigma_1^x}$$

$$P_x = -i\sigma_1^x$$

$$\begin{aligned} e^{i\theta\hat{\mathbf{u}}\cdot\sigma} &= \sum_{k=0}^{+\infty} \frac{(i\theta)^k}{k!}(\hat{\mathbf{u}}\cdot\sigma)^k \\ &= \sum_{k=0}^{+\infty} \frac{i^{2k}\theta^{2k}}{(2k)!}(\hat{\mathbf{u}}\cdot\sigma)^{2k} + \sum_{k=0}^{+\infty} \frac{i^{2k+1}\theta^{2k+1}}{(2k+1)!}(\hat{\mathbf{u}}\cdot\sigma)^{2k+1} \\ &\overset{(5)}{=} \sum_{k=0}^{+\infty} \frac{(-1)^k\theta^{2k}}{(2k)!} I + \sum_{k=0}^{+\infty} \frac{i(-1)^k\theta^{2k+1}}{(2k+1)!}(\hat{\mathbf{u}}\cdot\sigma) \\ &= \cos\theta I + i(\hat{\mathbf{u}}\cdot\sigma)\sin\theta \end{aligned}$$

Dynamical decoupling

$$U(2T) = P_x e^{-iJ\sigma_1^z\sigma_2^z T} P_x e^{-iJ\sigma_1^z\sigma_2^z T}$$

$$U(2T) = \boxed{P_x P_x} \underbrace{P_x^\dagger e^{-iJ\sigma_1^z\sigma_2^z T} P_x}_{e^{-iP_x^\dagger H P_x T}} e^{-iJ\sigma_1^z\sigma_2^z T}$$

-1

$$\mathbb{I} - iP_x^\dagger H P_x T - \frac{T^2}{2!} P_x^\dagger H^2 P_x + i\frac{T^3}{3!} P_x^\dagger H^3 P_x + \ldots$$

Dynamical decoupling

$$U(2T) = P_x e^{-iJ\sigma_1^z\sigma_2^z T} P_x e^{-iJ\sigma_1^z\sigma_2^z T}$$

$$U(2T) = \boxed{P_x P_x} \underbrace{P_x^\dagger e^{-iJ\sigma_1^z\sigma_2^z T} P_x}_{e^{-iP_x^\dagger H P_x T}} e^{-iJ\sigma_1^z\sigma_2^z T}$$

-1

$$P_x = -i\sigma_1^x$$

$$(i\sigma_1^x)J\sigma_1^z\sigma_2^z(-i\sigma_1^x) = -J\sigma_1^z\sigma_2^z$$

$$U(2T) = -e^{+iJ\sigma_1^z\sigma_2^z T} e^{-iJ\sigma_1^z\sigma_2^z T}$$

Average Hamiltonian

$$
\begin{aligned}
U(T_c) &= P_m U(t_m, t_{m-1}) P_{m-1} U(t_{m-1}, t_{m-2}) \ldots P_2 U(t_2, t_1) P_1 U(t_1, 0) \\
&= \underbrace{(P_m P_{m-1} \ldots P_1)}_{U_c(T_c) = \mathbb{1}} \underbrace{(P_{m-1} \ldots P_1)^\dagger U(t_m, t_{m-1})(P_{m-1} \ldots P_1)}_{e^{-i(P_{m-1}\ldots P_1)^\dagger H_0 (P_{m-1}\ldots P_1)\tau_m}} \ldots \\
&\quad \underbrace{(P_2 P_1)^\dagger U(t_3, t_2)(P_2 P_1)}_{e^{-i(P_2 P_1)^\dagger H_0 (P_2 P_1)\tau_3}} \underbrace{P_1^\dagger U(t_2, t_1) P_1}_{e^{-i P_1^\dagger H_0 P_1 \tau_2}} \underbrace{U(t_1, 0)}_{e^{-i H_0 \tau_1}} \\
&= \exp\left[-i H_{m-1} \tau_m\right] \ldots \exp\left[-i H_2 \tau_3\right] \exp\left[-i H_1 \tau_2\right] \exp\left[-i H_0 \tau_1\right] \\
&= \exp\left[-i \bar{H} T_c\right].
\end{aligned}
$$

Magnus expansion

$$= \exp\left[-\mathrm{i}H_{m-1}\tau_m\right]\ldots\exp\left[-\mathrm{i}H_2\tau_3\right]\exp\left[-\mathrm{i}H_1\tau_2\right]\exp\left[-\mathrm{i}H_0\tau_1\right]$$
$$= \exp\left[-\mathrm{i}\bar{H}T_{\mathrm{c}}\right].$$

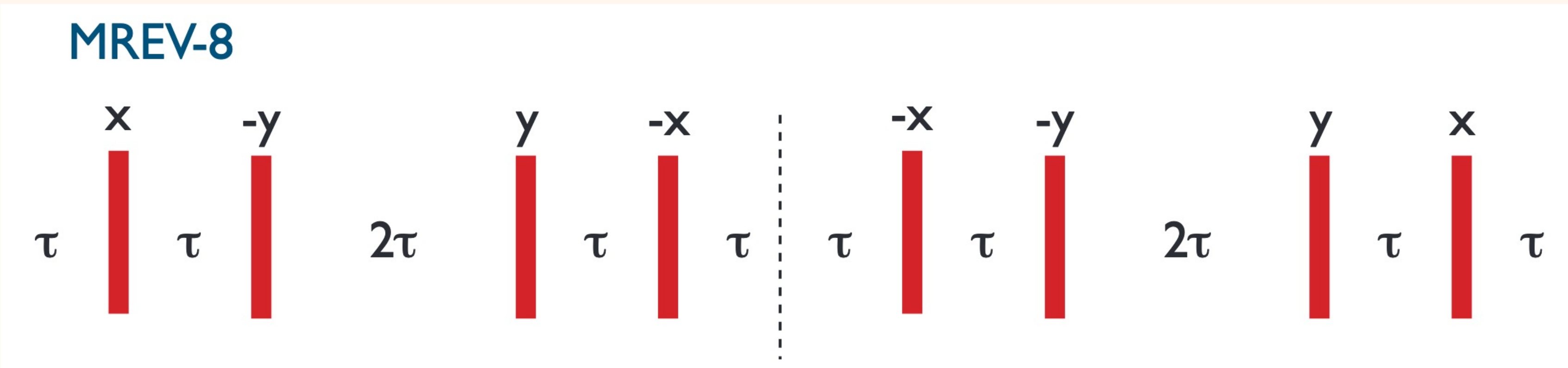
Baker–Campbell–Hausdorff expansion

$$\bar{H}^{(0)} = \frac{\Delta t}{T_{\mathrm{c}}}\sum_{k=1}^{|\mathcal{G}|} H_k,$$

$$\bar{H}^{(1)} = -\frac{\mathrm{i}(\Delta t)^2}{2T_{\mathrm{c}}}\sum_{l=2}^{|\mathcal{G}|}\sum_{k=1}^{l-1}[H_l, H_k],$$

$$\bar{H}^{(2)} = -\frac{(\Delta t)^3}{6T_{\mathrm{c}}}\left\{\sum_{m=3}^{|\mathcal{G}|}\sum_{l=2}^{m-1}\sum_{k=1}^{l-1}\left\{[H_m, [H_l, H_k] + [H_m, H_l], H_k]\right\}\right.$$
$$\left. + \frac{1}{2}\sum_{l=2}^{n}\sum_{k=1}^{l-1}\left\{[H_l, [H_l, H_k] + [H_l, H_k], H_k]\right\}\right\}.$$

MREV-8



Sequence with 8 $\pi/2$-pulses, so that the zeroth order of the average Hamiltonian becomes effectively

$$H_{\rm CSB} = \sum_{j}^{N} \omega_j \sigma_z^{\rm CS} \otimes \sigma_z^j \otimes \mathbb{1}^{\otimes N-1}$$

$$\mathcal{H}_E = \mathbb{1}^{\rm cs} \otimes \sum_{j<k}^{N} \Omega_{jk} \left[\sigma_Z^j \sigma_Z^k - \frac{1}{4} (\sigma_+^j \sigma_-^k + \sigma_-^j \sigma_+^k) \right]$$

Coherence orders

To explain how the experimental measurements of the coherence orders intensities are done, we take as an example the case of a central spin coupled with two bath spins, $N = 2$. After the cross polarization step, the central spin and the bath spins are uncorrelated and the density matrix for a single molecule is given by

$$\rho(0) = \frac{\mathbb{1}^{\otimes 3}}{2^3} + \frac{\epsilon}{2^3}\sigma_{\rm X}^{\rm CS}\otimes\mathbb{1}\otimes\mathbb{1}. \tag{S2}$$

The homonuclear dipolar interaction among the bath spins is turned off with the MREV-8 pulse sequence, so the system evolves under the heteronuclear dipolar interaction only, $U_{\text{CS-B}}(T) = \mathrm{e}^{-i\mathcal{H}_{\text{CS-B}}T}$, where

$$\mathcal{H}_{\text{CS-B}} = \frac{\omega_1}{2}\{\sigma_{\rm Z}^{\rm CS}\otimes\sigma_{\rm Z}^{1}\otimes\mathbb{1}\} + \frac{\omega_2}{2}\{\sigma_{\rm Z}^{\rm CS}\otimes\mathbb{1}\otimes\sigma_{\rm Z}^{2}\}. \tag{S3}$$

After the evolution time T, the density matrix becomes

$$\begin{aligned}\rho(T) = \frac{\epsilon}{8}\Big\{ & \cos(\omega_1 T)\cos(\omega_2 T) \quad \sigma_{\rm X}^{\rm CS}\otimes\mathbb{1}\otimes\mathbb{1} \\ & + \sin(\omega_1 T)\cos(\omega_2 T) \quad \sigma_{\rm Y}^{\rm CS}\otimes\sigma_{\rm Z}\otimes\mathbb{1} \; + \cos(\omega_1 T)\sin(\omega_2 T) \quad \sigma_{\rm Y}^{\rm CS}\otimes\mathbb{1}\otimes\sigma_{\rm Z} \\ & - \sin(\omega_1 T)\sin(\omega_2 T) \quad \sigma_{\rm X}^{\rm CS}\otimes\sigma_{\rm Z}\otimes\sigma_{\rm Z}\Big\} \\ & + \frac{\mathbb{1}^{\otimes 3}}{2^3}.\end{aligned} \tag{S4}$$

In the equation above, the number of non-identity bath spin operators in the first line is zero, $m = 0$ and the amplitude of this term is $\mathrm{C}_0^{\rm z} = \cos(\omega_1 T)\cos(\omega_2 T)$. Both terms in the second line have one non-identity bath spin operator, $m = 1$, one with amplitude $\mathrm{C}_1^{\mathrm{z}(1)} = \sin(\omega_1 T)\cos(\omega_2 T)$ and the other with amplitude $\mathrm{C}_1^{\mathrm{z}(2)} = \cos(\omega_1 T)\sin(\omega_2 T)$. The third line has $m = 2$ and $\mathrm{C}_2^{\mathrm{z}(1,2)} = -\sin(\omega_1 T)\sin(\omega_2 T)$.

Using raising and lowering operators in the x-quantization basis, $\sigma_+^{\rm X} := \frac{\sigma_{\rm Y} + i\sigma_{\rm Z}}{\sqrt{2}}$ and $\sigma_-^{\rm X} := \frac{\sigma_{\rm Y} - i\sigma_{\rm Z}}{\sqrt{2}}$, the density matrix above can be written as

$$\begin{aligned}\rho(T) = \frac{\epsilon}{8}\Big\{ & \cos(\omega_1 T)\cos(\omega_2 T) \quad \sigma_{\rm X}^{\rm CS}\otimes\mathbb{1}\otimes\mathbb{1} \\ & + \sin(\omega_1 T)\cos(\omega_2 T)\frac{1}{\sqrt{2}i}[\sigma_{\rm Y}^{\rm CS}\otimes\sigma_+^{\rm X}\otimes\mathbb{1} - \sigma_{\rm Y}^{\rm CS}\otimes\sigma_-^{\rm X}\otimes\mathbb{1}] + \cos(\omega_1 T)\sin(\omega_2 T)\frac{1}{\sqrt{2}i}[\sigma_{\rm Y}^{\rm CS}\otimes\mathbb{1}\otimes\sigma_+^{\rm X} - \sigma_{\rm Y}^{\rm CS}\otimes\mathbb{1}\otimes\sigma_-^{\rm X}] \\ & + \sin(\omega_1 T)\sin(\omega_2 T)\frac{1}{2}[\sigma_{\rm X}^{\rm CS}\otimes\sigma_+^{\rm X}\otimes\sigma_+^{\rm X} + \sigma_{\rm X}^{\rm CS}\otimes\sigma_-^{\rm X}\otimes\sigma_-^{\rm X} - \sigma_{\rm X}^{\rm CS}\otimes\sigma_+^{\rm X}\otimes\sigma_-^{\rm X} - \sigma_{\rm X}^{\rm CS}\otimes\sigma_-^{\rm X}\otimes\sigma_+^{\rm X}]\Big\} \\ & + \frac{\mathbb{1}^{\otimes 3}}{2^3}.\end{aligned} \tag{S5}$$

Coherence orders

The coherence order n of each term in the equation above is determined by the difference between the number n_+ of σ_+^{x} operators and the number n_- of σ_-^{x} operators. One sees that the second line in Eq. (S5), which had $m = 1$ in Eq. (S4), now has terms with coherence order +1 and with coherence order -1. The third line in Eq. (S5), which had $m = 2$ in Eq. (S4), now has four terms with coherence orders respectively given by $n = 2, -2, 0$ and 0. Using the notation of the Eq. (6) in the main text, we have that

$$\begin{array}{lll}
\rho_0^{\mathrm{x},1} = \sigma_{\mathrm{X}}^{\mathrm{cs}} \otimes \mathbb{1} \otimes \mathbb{1} & \rho_0^{\mathrm{x},2} = \sigma_{\mathrm{X}}^{\mathrm{cs}} \otimes \sigma_+^{\mathrm{x}} \otimes \sigma_-^{\mathrm{x}} & \rho_0^{\mathrm{x},3} = \sigma_{\mathrm{X}}^{\mathrm{cs}} \otimes \sigma_-^{\mathrm{x}} \otimes \sigma_+^{\mathrm{x}} \\
\rho_1^{\mathrm{x},1} = \sigma_{\mathrm{Y}}^{\mathrm{cs}} \otimes \sigma_+^{\mathrm{x}} \otimes \mathbb{1} & \rho_1^{\mathrm{x},2} = \sigma_{\mathrm{Y}}^{\mathrm{cs}} \otimes \mathbb{1} \otimes \sigma_+^{\mathrm{x}} & \\
\rho_{-1}^{\mathrm{x},1} = \sigma_{\mathrm{Y}}^{\mathrm{cs}} \otimes \sigma_-^{\mathrm{x}} \otimes \mathbb{1} & \rho_{-1}^{\mathrm{x},2} = \sigma_{\mathrm{Y}}^{\mathrm{cs}} \otimes \mathbb{1} \otimes \sigma_-^{\mathrm{x}} & \\
\rho_2^{\mathrm{x},1} = \sigma_{\mathrm{Y}}^{\mathrm{cs}} \otimes \sigma_+^{\mathrm{x}} \otimes \sigma_+^{\mathrm{x}} & & \\
\rho_{-2}^{\mathrm{x},1} = \sigma_{\mathrm{Y}}^{\mathrm{cs}} \otimes \sigma_-^{\mathrm{x}} \otimes \sigma_-^{\mathrm{x}}. & &
\end{array} \tag{S6}$$

Our experiment can measure the sum of the absolute square of the amplitudes of terms with the same coherence order. This sum is denoted by $|\mathrm{C}_n^{\mathrm{x}}(T)|^2$ and for Eq. (S5) they correspond to

$$|\mathrm{C}_0^{\mathrm{x}}(T)|^2 = \cos(\omega_1 T)^2 \cos(\omega_2 T)^2 + \frac{1}{2} \sin(\omega_1 T)^2 \sin(\omega_2 T)^2 \tag{S7}$$

$$|\mathrm{C}_1^{\mathrm{x}}(T)|^2 = \frac{1}{2}(\cos(\omega_1 T)^2 \sin(\omega_2 T)^2 + \sin(\omega_1 T)^2 \cos(\omega_2 T)^2)$$

$$|\mathrm{C}_{-1}^{\mathrm{x}}(T)|^2 = \frac{1}{2}(\cos(\omega_1 T)^2 \sin(\omega_2 T)^2 + \sin(\omega_1 T)^2 \cos(\omega_2 T)^2)$$

$$|\mathrm{C}_2^{\mathrm{x}}(T)|^2 = \frac{1}{4} \sin(\omega_1 T)^2 \sin(\omega_2 T)^2$$

$$|\mathrm{C}_{-2}^{\mathrm{x}}(T)|^2 = \frac{1}{4} \sin(\omega_1 T)^2 \sin(\omega_2 T)^2$$

The experiment: Coherence orders

The experiment cannot directly measure $C_z^{(m)}(T)$.
The experiment can directly measure the coherence order intensities $C_x^{(n)}(T)$.

Write the bath spin operators in terms of coherence raising and lowering operators in the x-quantization basis

$$\sigma_\pm^x := (\sigma_y \pm i\sigma_z)/\sqrt{2}$$

$$n = n_+ - n_-$$

$$\sigma_x^{CS} \sigma_z^{j_1} \sigma_z^{j_2}$$

$\sigma_x^{CS} \sigma_+^{xj_1} \sigma_+^{xj_2}$ Coherence order 2

$\sigma_x^{CS} \sigma_-^{xj_1} \sigma_-^{xj_2}$ Coherence order - 2

$\sigma_x^{CS} \sigma_-^{xj_1} \sigma_+^{xj_2}$ Coherence order 0

$\sigma_x^{CS} \sigma_+^{xj_1} \sigma_-^{xj_2}$ Coherence order 0