SAND2013-7190C

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Converting a real quantum bath to an effective classical noise

Windsor 2013

Wayne Witzel, Sandia National Laborators, NM

U.S. DEPARTMENT OF ENERGY

NNSA National Nuclear Security Administration

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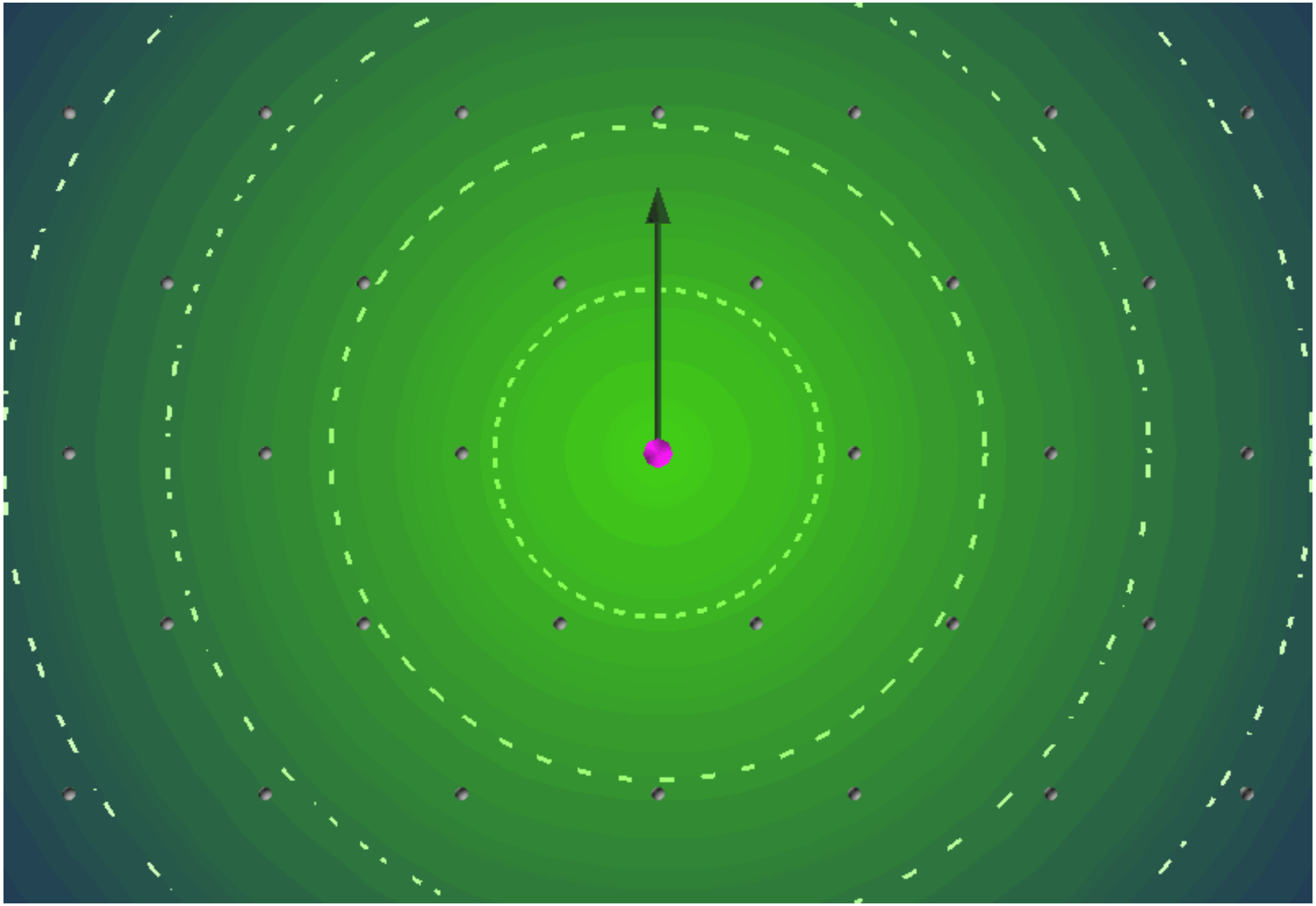
Outline

- Central Spin Decoherence Problem
  - Spectral Diffusion (SD)
  - Spin Echo
- Cluster Expansion Solutions
  - Natural Silicon SD
  - Other Nuclear-Induced SD Cases (and Successes)
  - Generic Dipolar-Only Problem
  - Enriched Silicon
- Correlation Functions (generic dipolar-only problem)
  - Computed Directly from a Cluster Expansion
  - Comparing Echo Decay Results
  - Using for Optimization
- Summary of New Findings

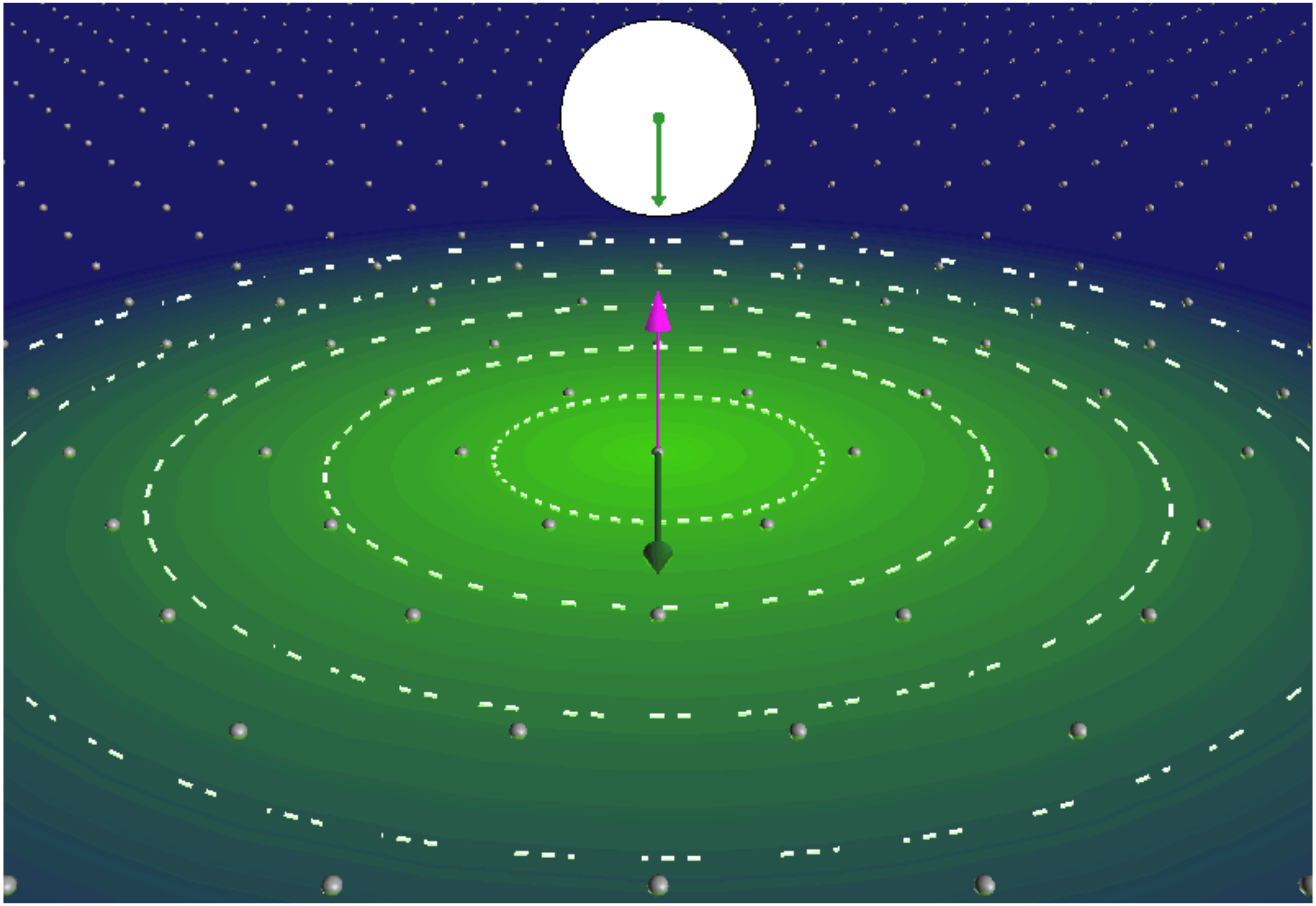
Central Spin Decoherence

localized solid state electron



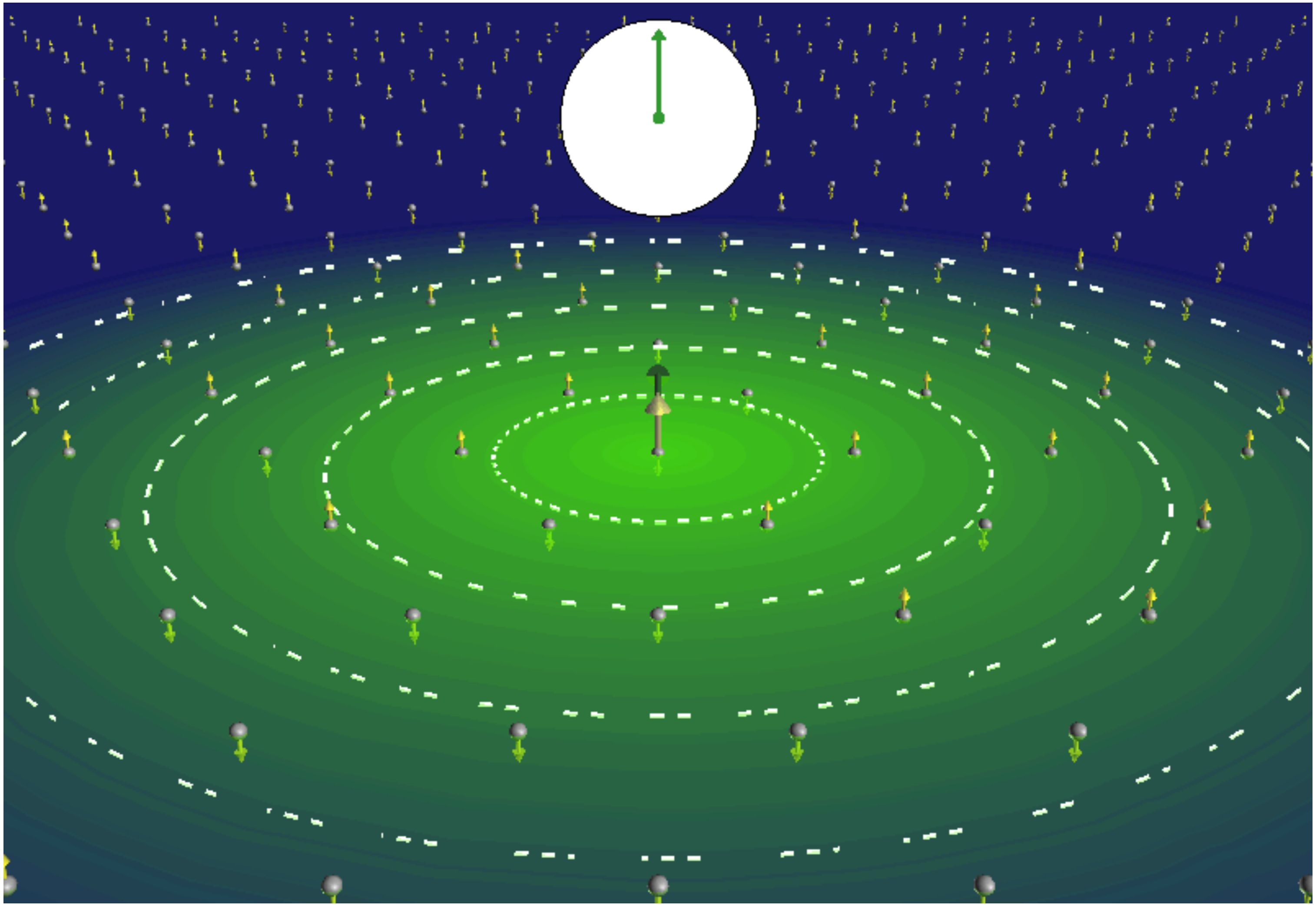
Central Spin Decoherence

localized solid state electron



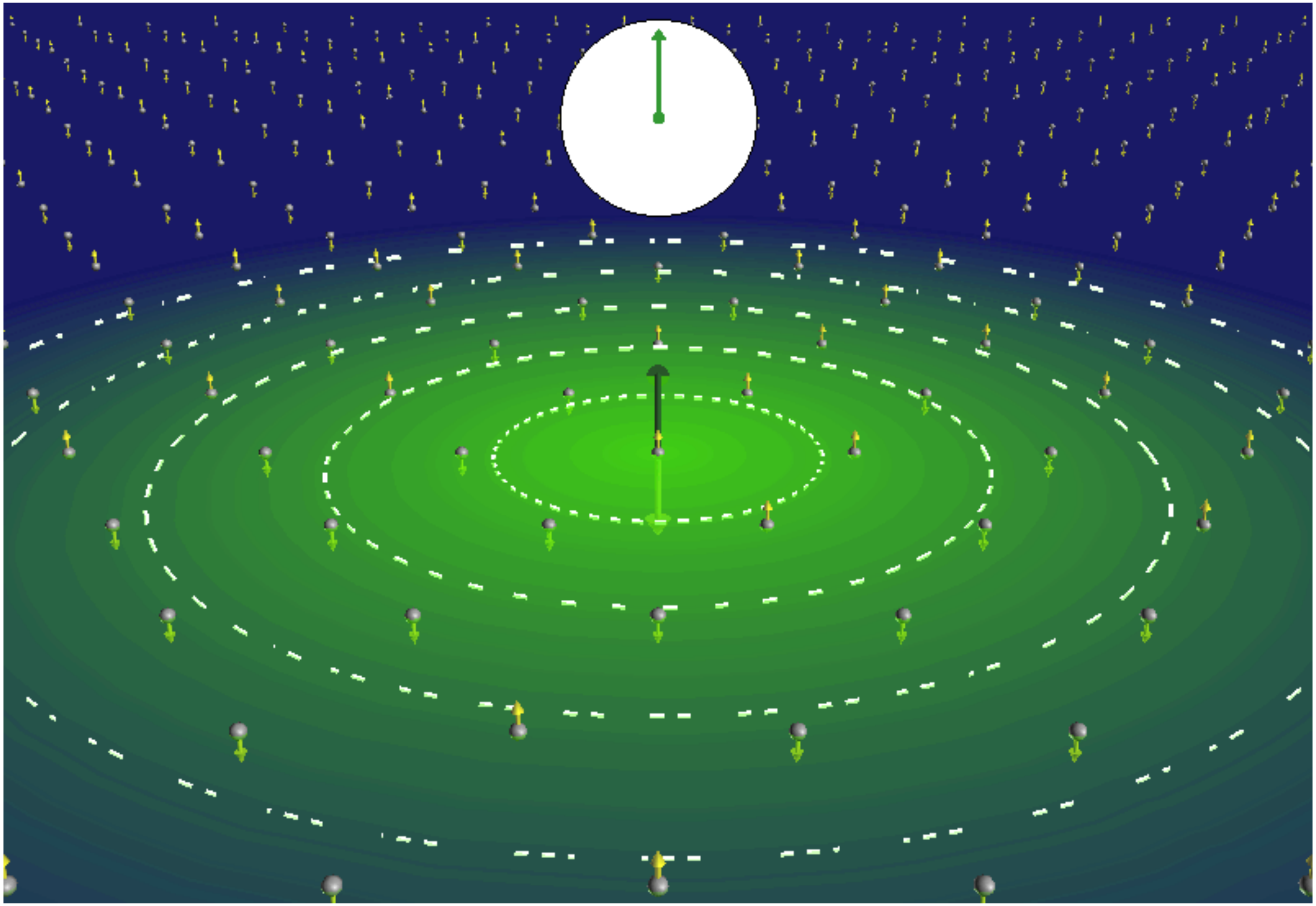
Central Spin Decoherence

random nuclear field



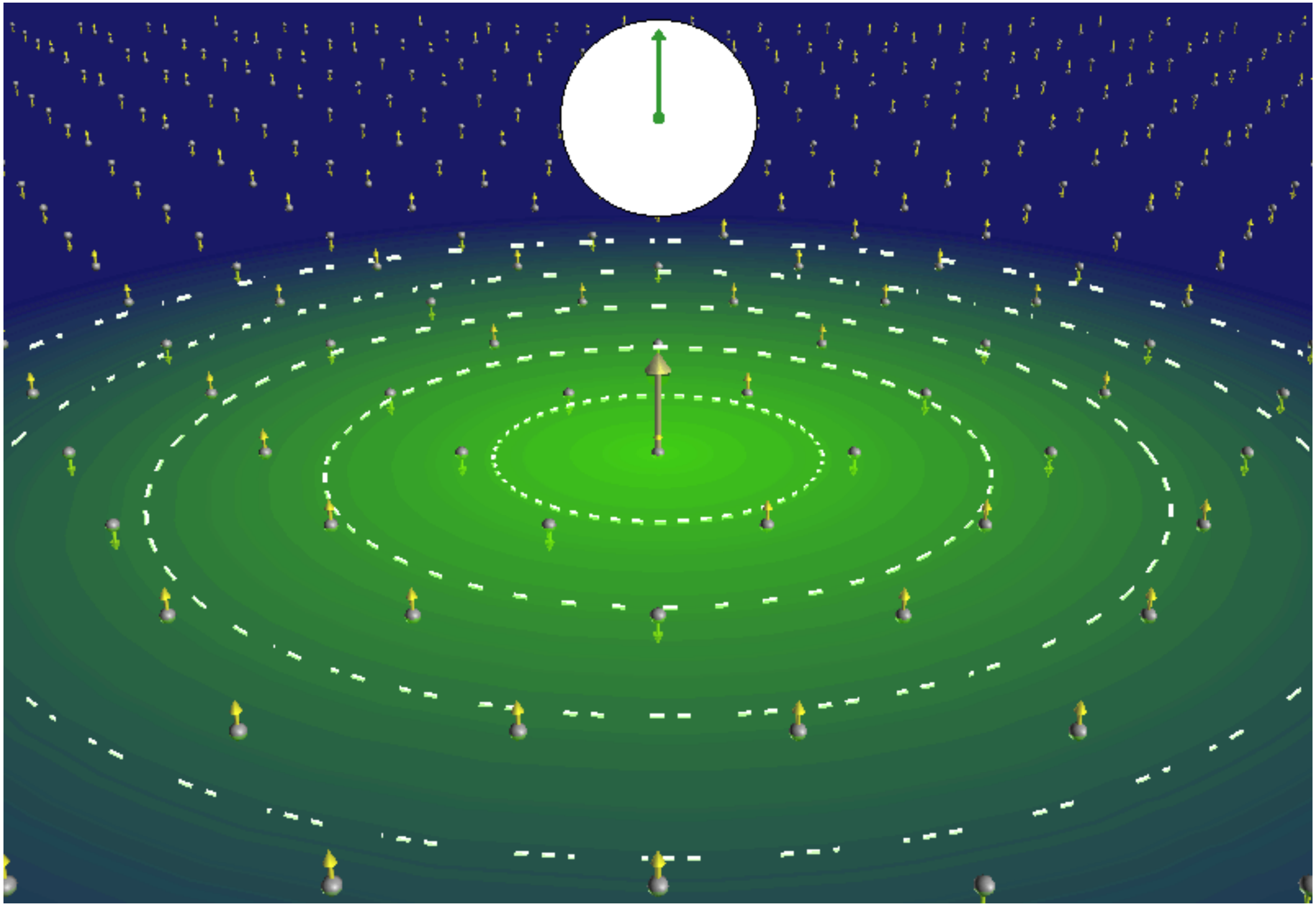
Central Spin Decoherence

random nuclear field



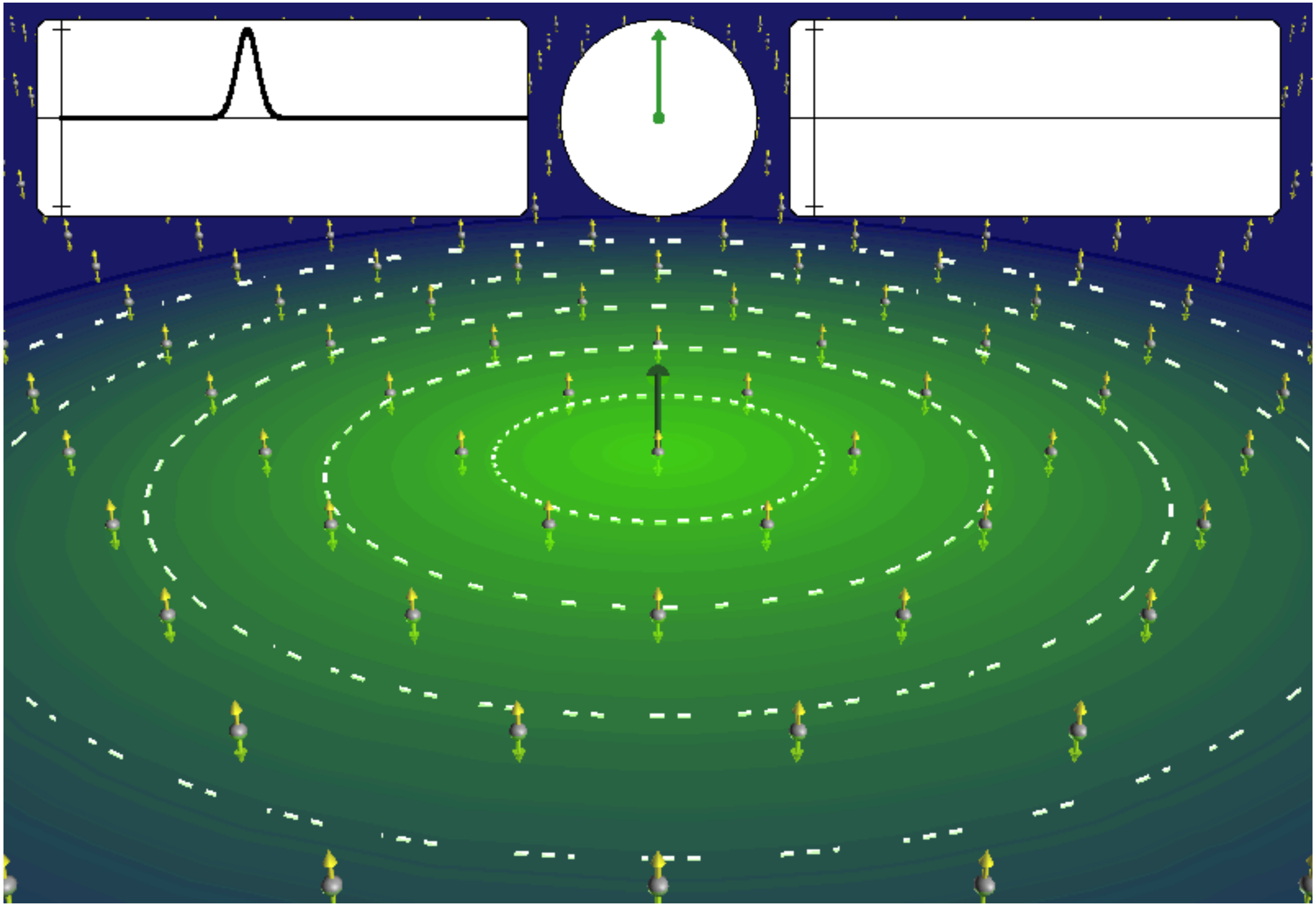
Central Spin Decoherence

random nuclear field



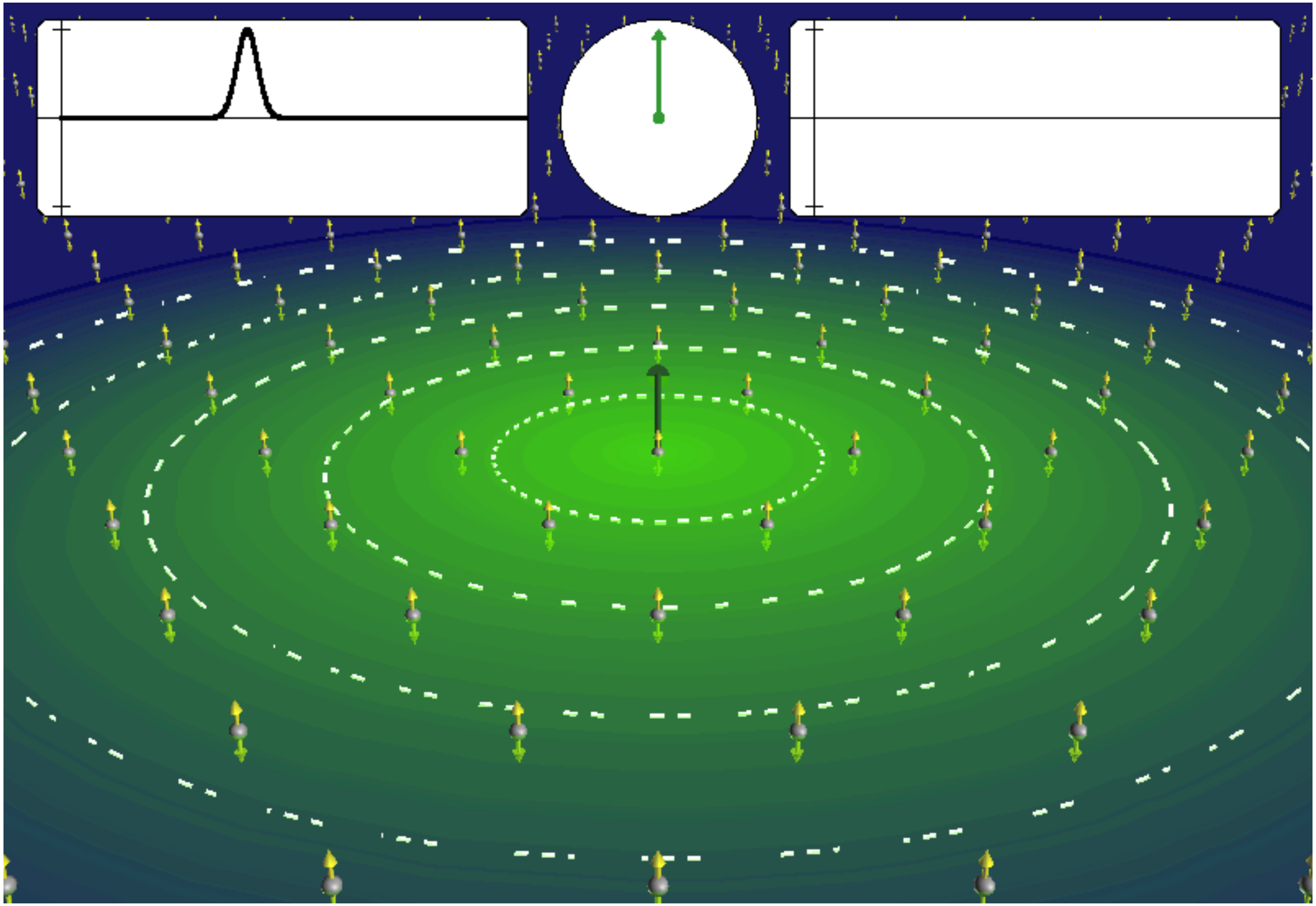
Central Spin Decoherence

ensemble: inhomogeneous broadening



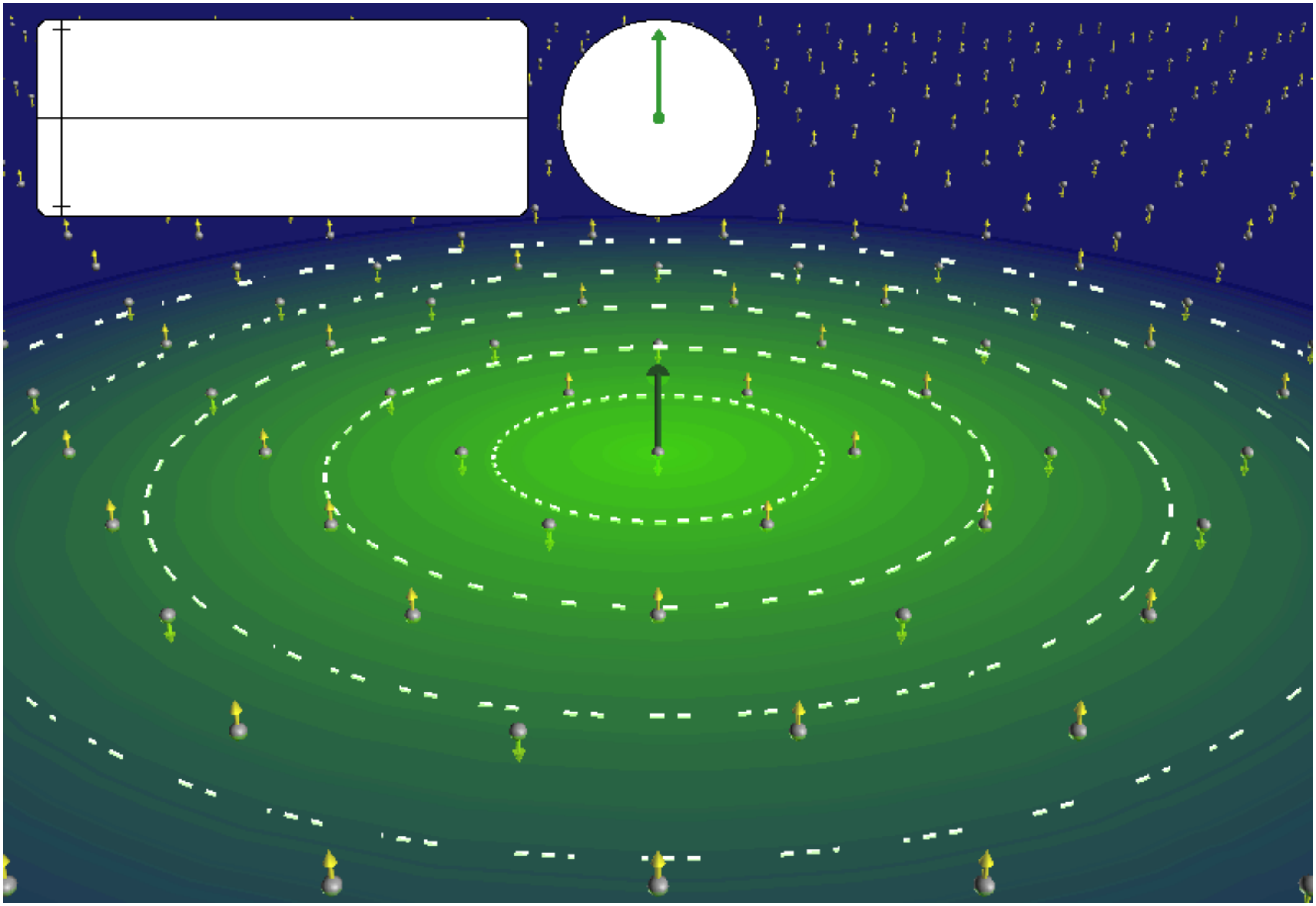
Central Spin Decoherence

Hahn spin echo



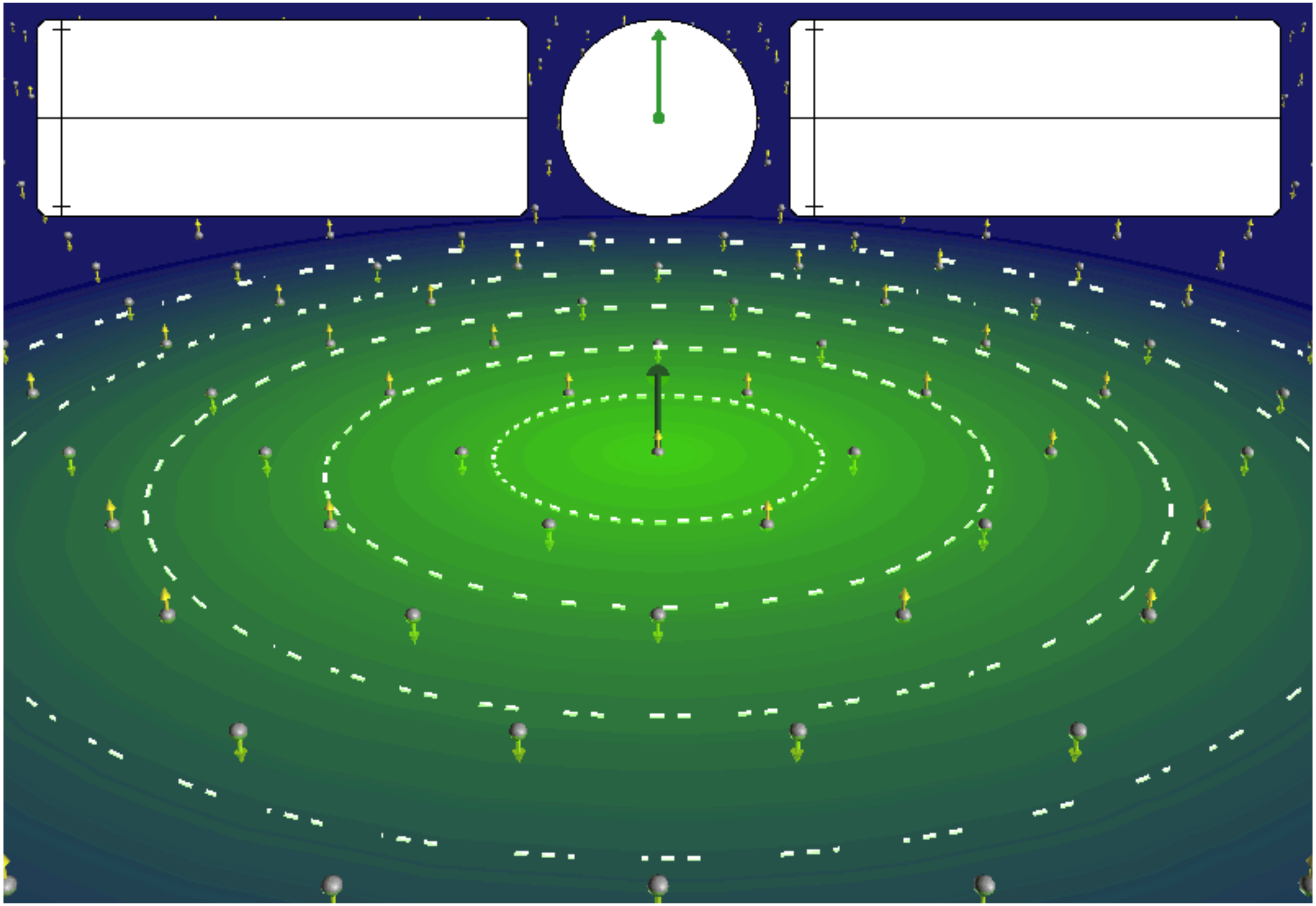
Central Spin Decoherence

random fluctuations

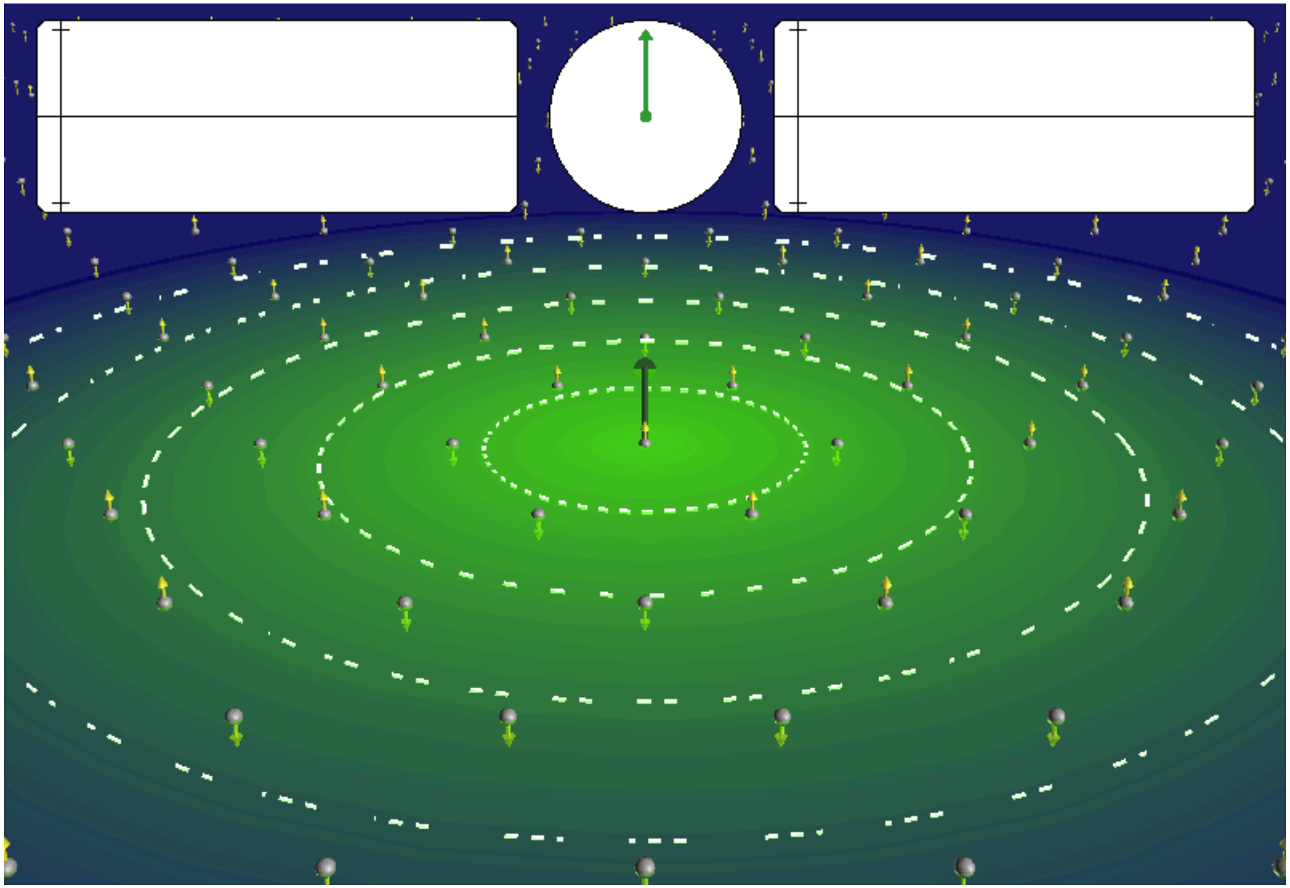


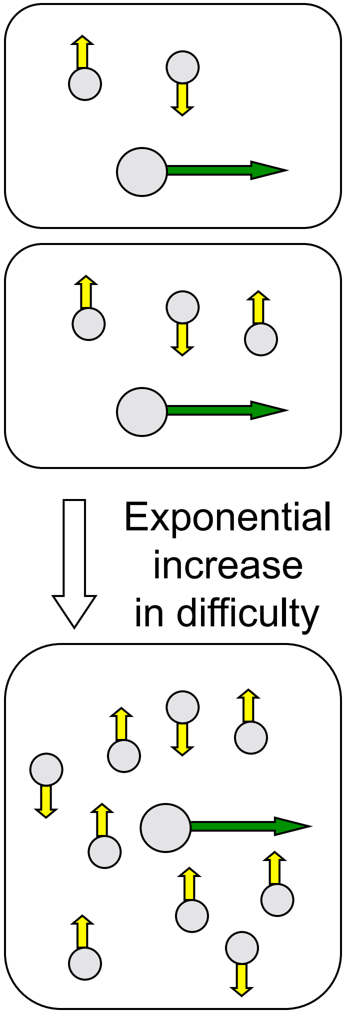
Central Spin Decoherence

ensemble: spectral diffusion



Challenge

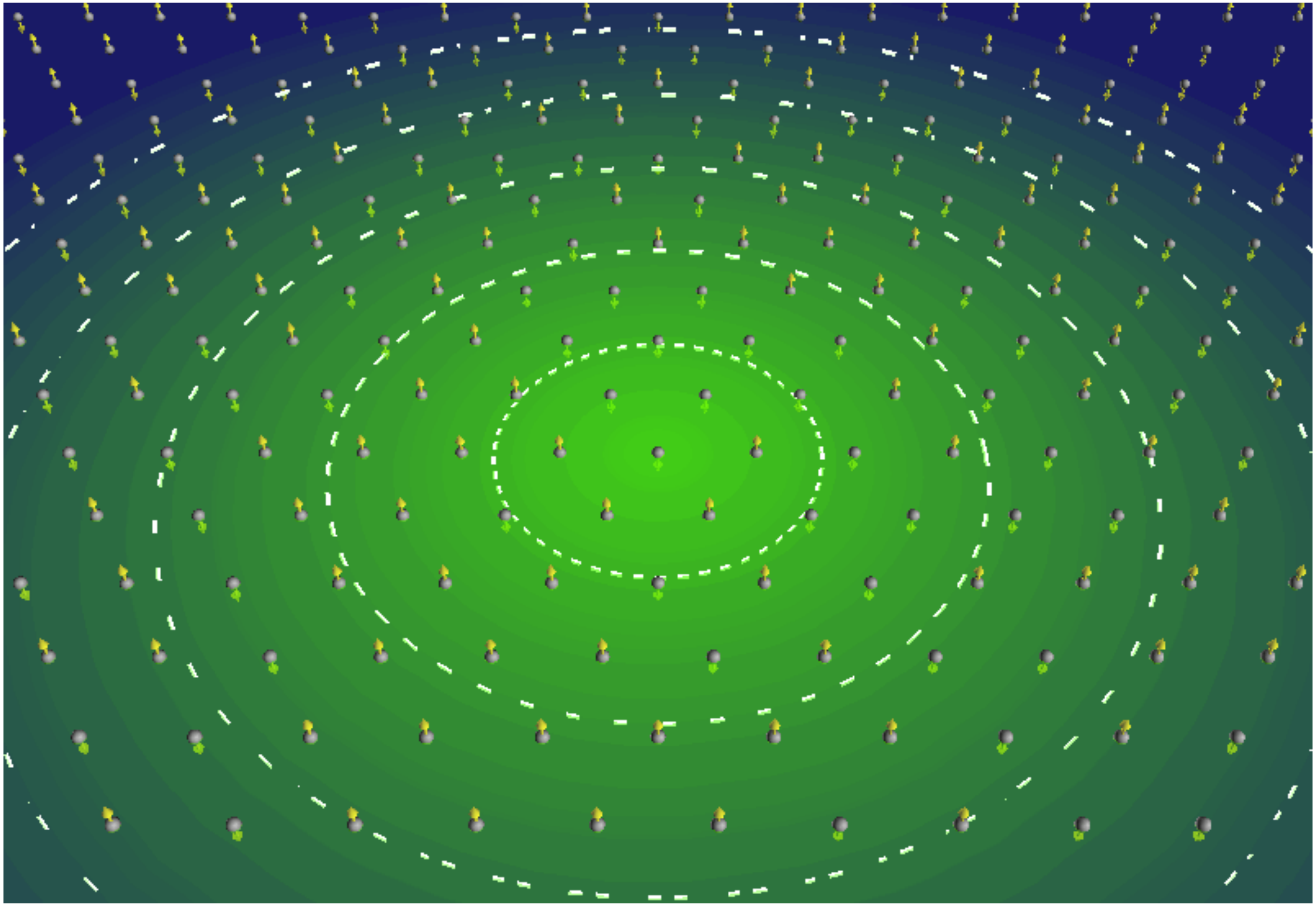




Spin Echo: $U(\tau) = \mathrm{e}^{-i\mathcal{H}\tau}\sigma_{x,e}\mathrm{e}^{-i\mathcal{H}\tau}.$

$$\rho(t) \;=\; \hat{U}\rho(0)\hat{U}^{\dagger}, \quad \rho_q(t) \;=\; \mathrm{Tr}_B\, \rho(t)$$

Visualizing Clusters



Nuclear-induced Spectral Diffusion: Problem Hamiltonian and Evolution

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Effective Hamiltonian (large B-field)

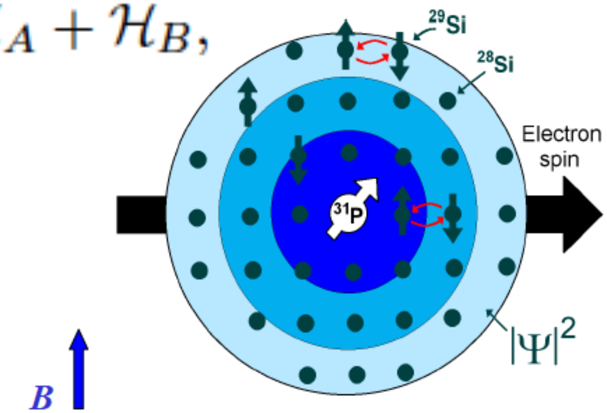
$$\mathcal{H} = \mathcal{H}_{Ze} + \mathcal{H}_{Zn} + \mathcal{H}_A + \mathcal{H}_B,$$

$$\mathcal{H}_{Ze} = \gamma_S B S_z,$$

$$\mathcal{H}_{Zn} = -\gamma_I B \sum_n I_{nz},$$

$$\mathcal{H}_A = \sum_n A_n I_{nz} S_z,$$

$$\mathcal{H}_B = \sum_{n \neq m} b_{nm}(I_{n+}I_{m-} - 2I_{nz}I_{mz}).$$



Hahn echo evolution

$$U(\tau) = e^{-i\mathcal{H}\tau}\sigma_{x,e}e^{-i\mathcal{H}\tau}$$

Initialize transverse spin, thermal bath:

$$\rho_0 = \frac{1}{2M}|y_e\rangle\langle y_e| \otimes e^{-\mathcal{H}_n/k_B T}$$

$$\rho(\tau) = U(\tau)\rho_0 U^\dagger(\tau)$$

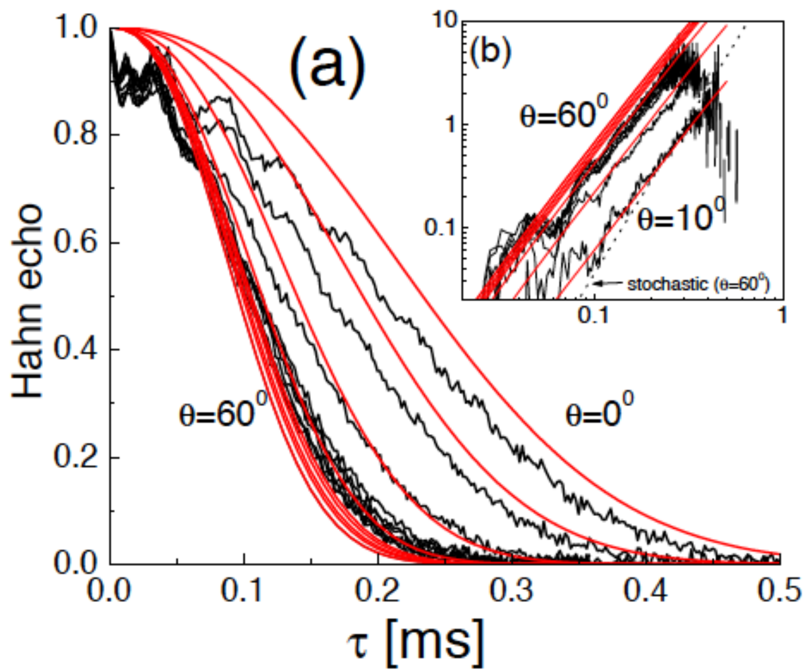
Echo decay: $v_E(\tau) = 2\left|\mathrm{Tr}\left\{(S_x + iS_y)\rho(\tau)\right\}\right|$

- Distant pairs or pairs with similar A_n: can flip-flop, but electron can't feel it.
- Pairs with very different A_n: cannot flip-flop in presence of the electron.
- Pairs with $(A_n - A_m) \sim b_{nm}$: flip-flopping causes electron decoherence.
- Larger $A_n - A_m$ causes faster but smaller decoherence (not full flip-flops).

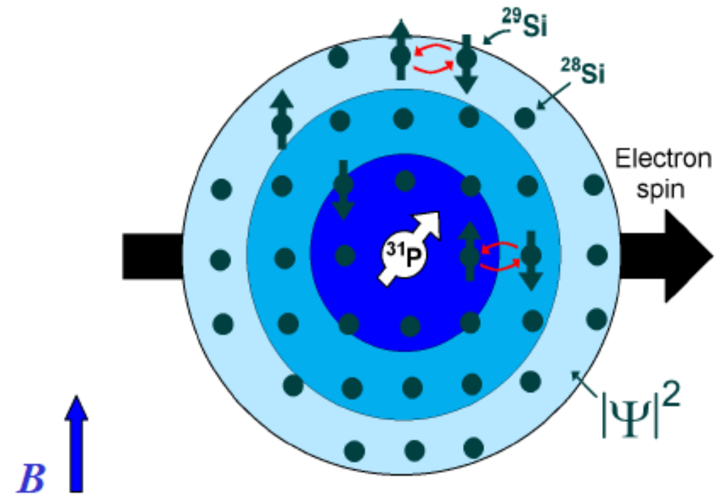
Witzel, de Sousa, Das Sarma, PRB 72, 161306(R) (2005)

Cluster Expansion Calculation of Spectral Diffusion (Spin Bath)

- Successive approximation in size of bath “cluster”.
- For nuclear-induced spectral diffusion, simply compose solutions from pairs of nuclear spins.

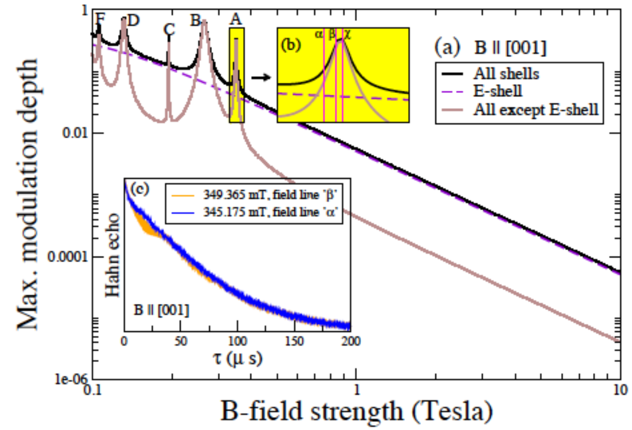


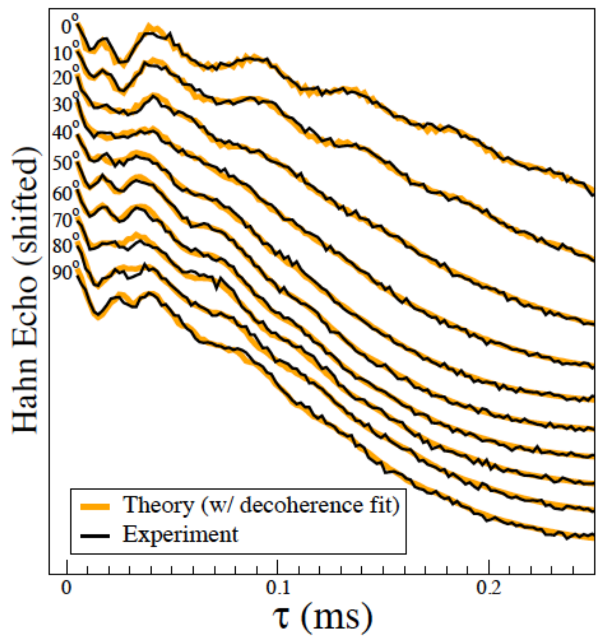
No fitting parameters!



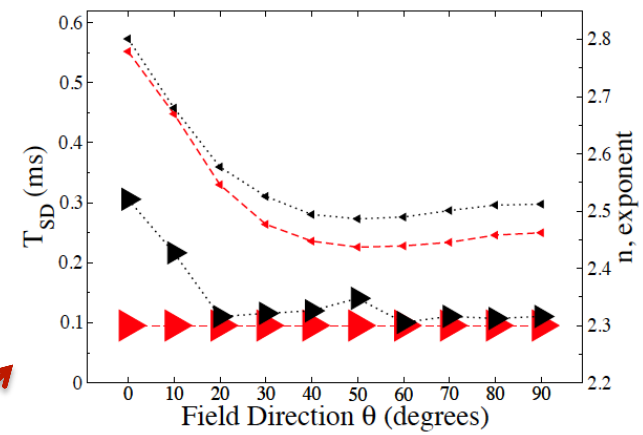
Electron spin echo envelope modulations

- Anisotropic interactions cause modulations of the echo envelope.
- By happenstance, Si:P echo experiments used B-field in which this effect is strong.



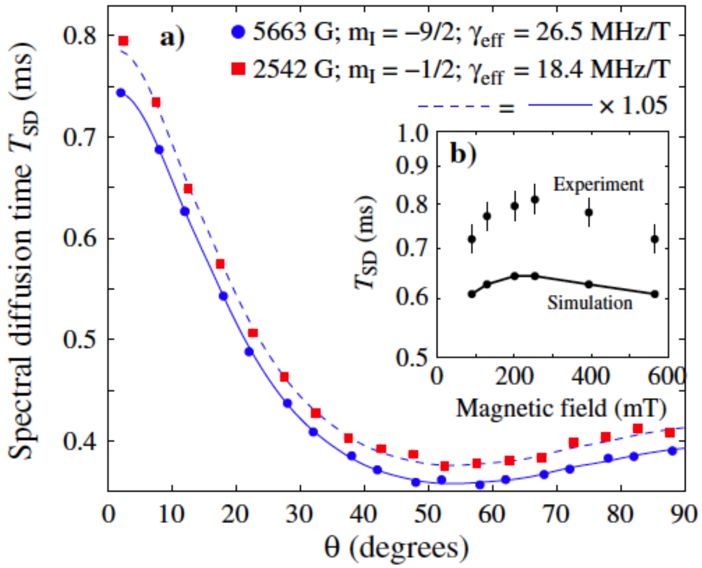


2 global fitting parameters plus 2 parameters per curve.

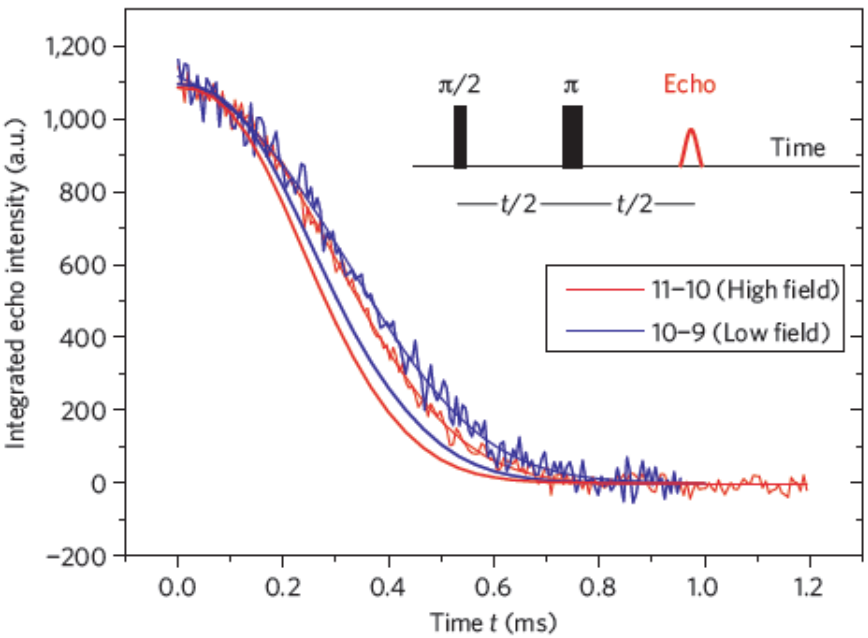


Comparison of 2 per-curve fitting parameters with theory.

Other Nuclear-Induced Decoherence Cases: Si:Bi in Natural Silicon

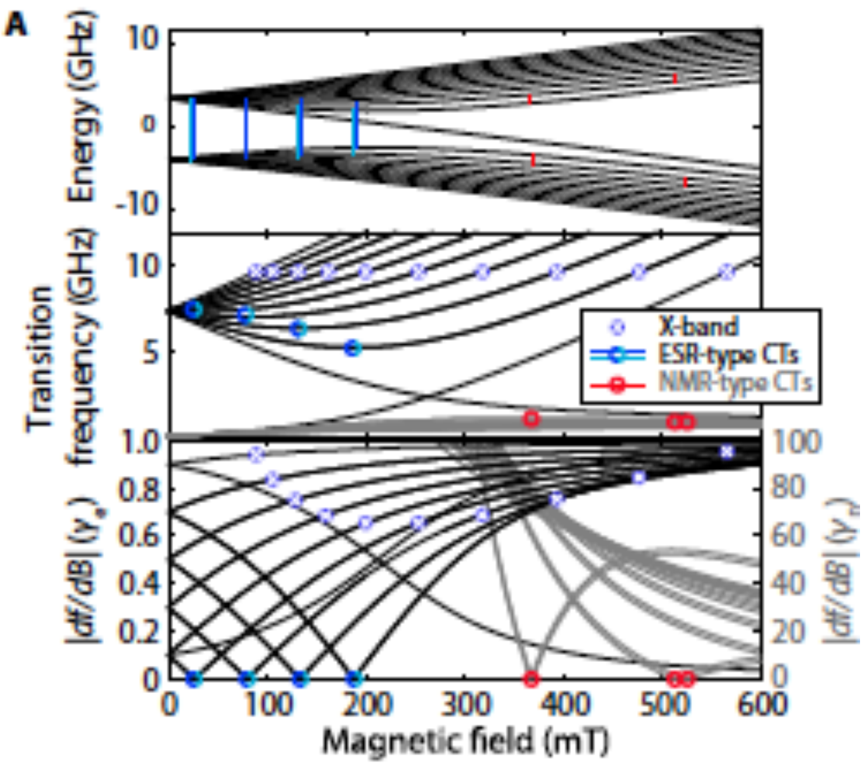


George et al, PRL **105**, 067601 (2010)

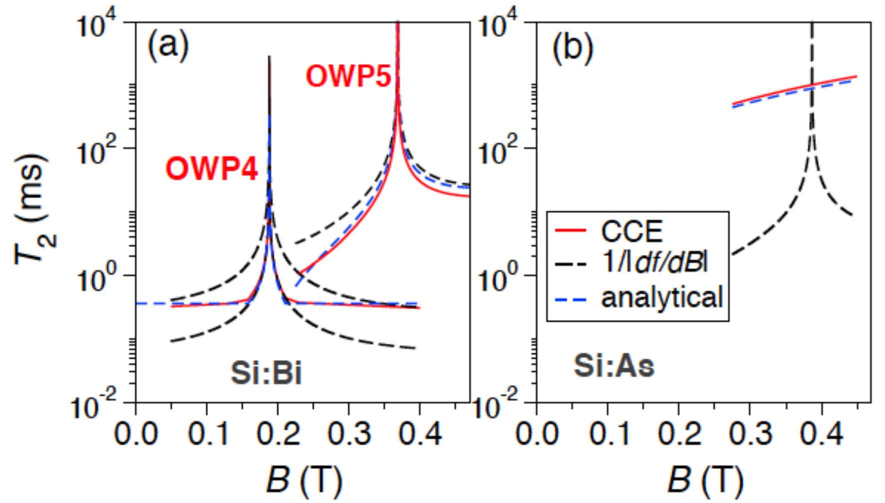


Morley et al, Nat. Mat. **12**, 103 (2013)

Si:Bi and Si:As at Optimal Working Points (Clock Transitions) in Natural Silicon

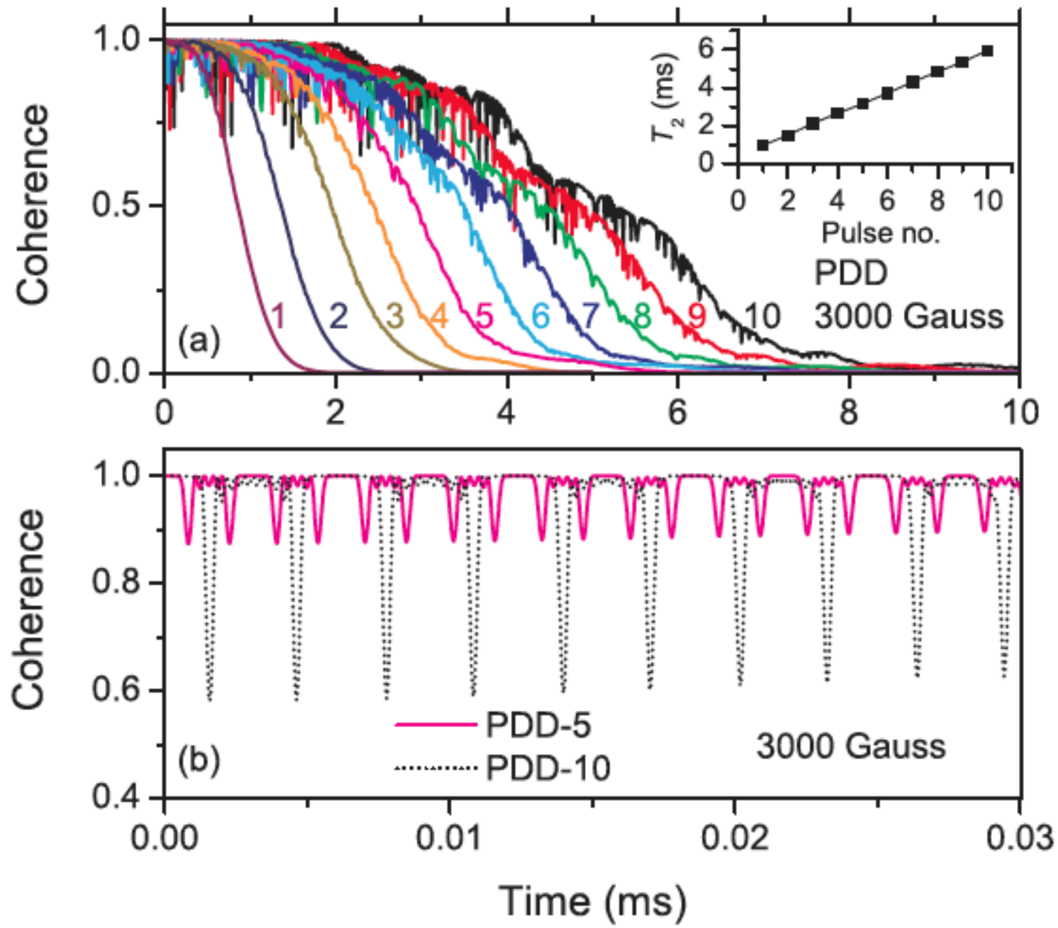


Wolfowicz et al., arXiv:1301.6567



Balian, Wolfowicz, Morton, Monteiro, arXiv:1302.1709

Other Nuclear-Induced Decoherence Cases: NV Centers in Diamond



Other Nuclear-Induced Decoherence Cases: GaAs Quantum Dots

$$\mathcal{L}_{+,-}(2\tau) \cong \prod_{k \in K_B} e^{-2\tau^4 E_k^2 B_k^2 \,\mathrm{sinc}^4(h_k^B \tau/2)}$$

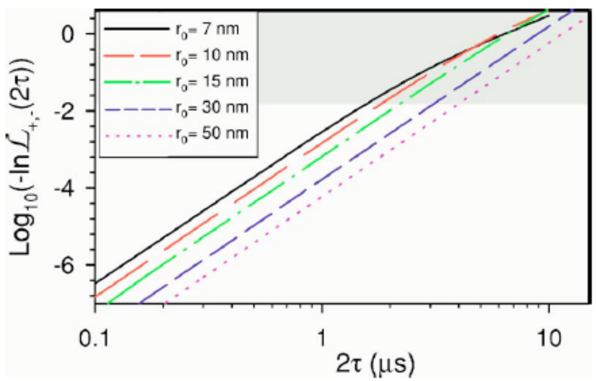
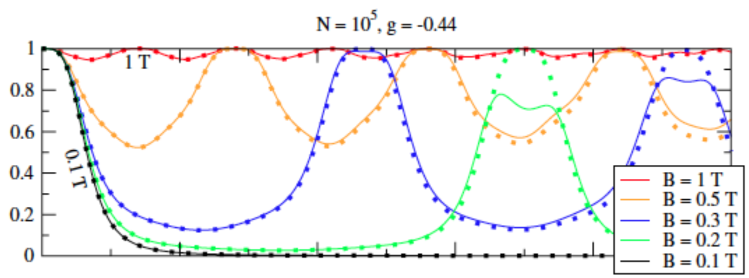
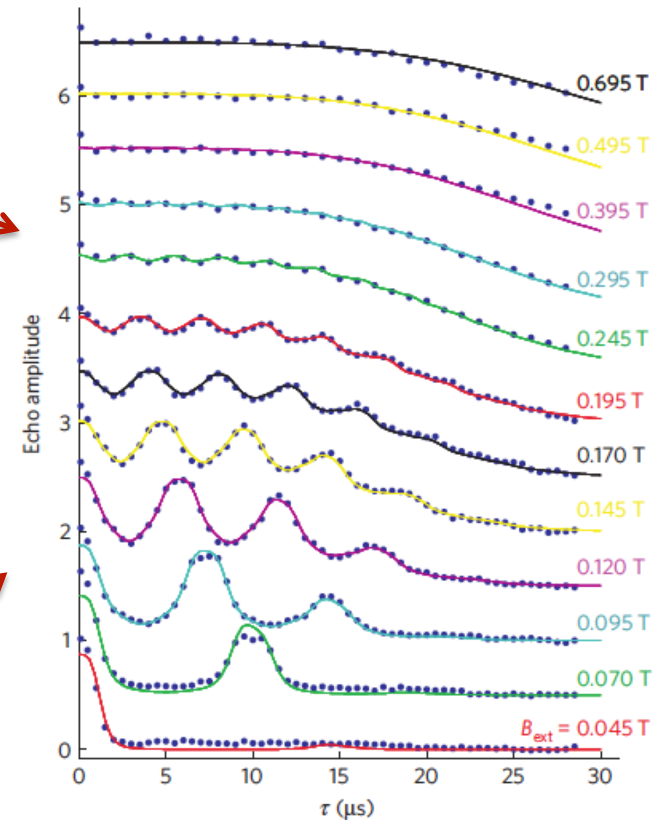


FIG. 3. (Color online) Spin-echo signal for dots of $L_{[001]} = 2.8$ nm and various r_0 at $B_{ext} = 10$ T.

Yao, Liu, Sham, PRB **74**, 195301 (2006)



Cywinski, Witzel, Das Sarma, PRL **102**, 057601 (2009)



Bluhm et al, Nature Physics 7, **109** (2011)

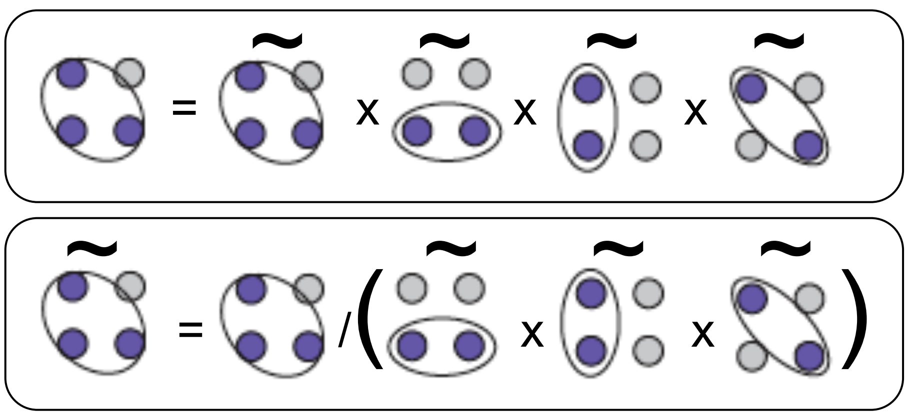
Cluster Correlation Expansion for Computing Echo Decay

Cluster correlation expansion:

$$L = \langle \rho_q^{+-}(t) / \rho_q^{+-}(0) \rangle$$

$L_{\mathcal{S}} = L$ with the exclusion of spins outside of $\mathcal{S}$: $\mathcal{S}$

$$L_{\mathcal{S}} = \prod_{\mathcal{C} \subseteq \mathcal{S}} \tilde{L}_{\mathcal{C}}, \quad \tilde{L}_{\mathcal{S}} = L_{\mathcal{S}} / \prod_{\mathcal{C} \subset \mathcal{S}} \tilde{L}_{\mathcal{C}}$$



$$L_{\mathrm{CCE}}^{(k)} = \prod_{\|\mathcal{S}\| \leq k} \tilde{L}_{\mathcal{S}}.$$

Works great for weakly coupled baths (perturbative orders equate with cluster size).

Strongly coupled baths require more care.

Wen Yang and Ren-bao Liu, PRB 78, 085315 (2008)

"Canonical" Dipolar-only Problem

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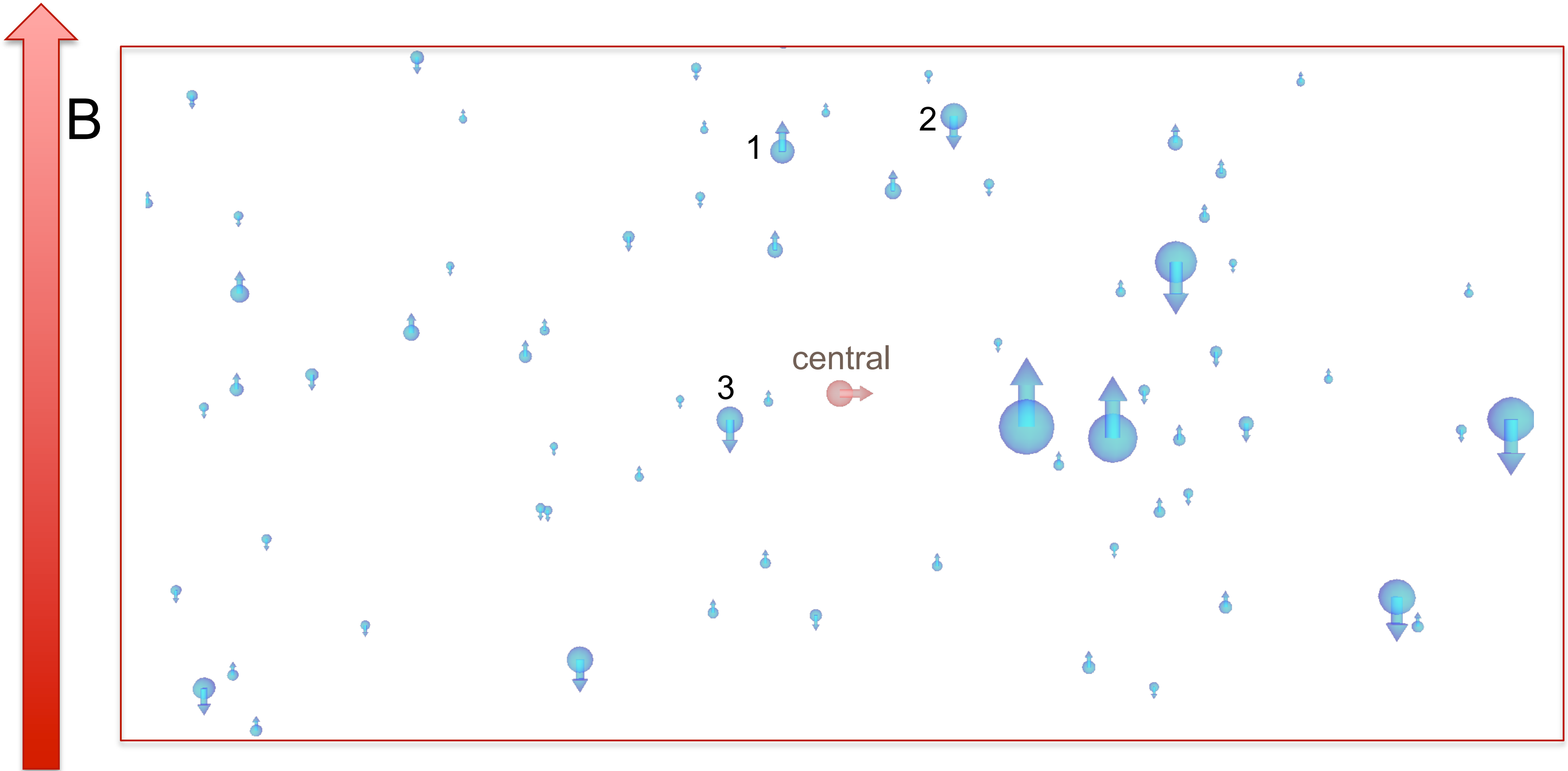
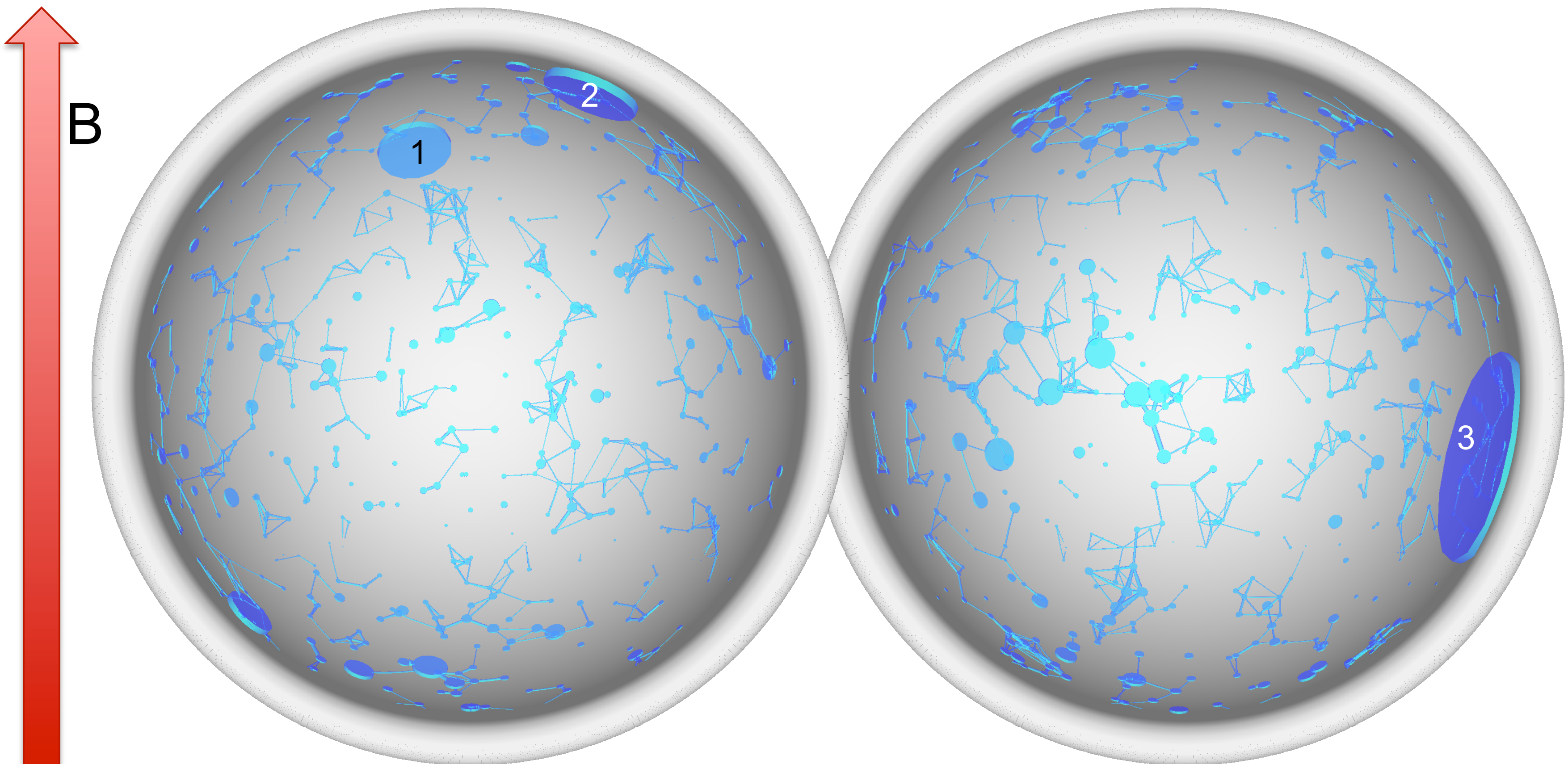


$$\mathcal{H}_{\text{eff}} = \sum_{n,m>0} b_{n,m} \hat{S}_n^+ \hat{S}_m^- - 2 \sum_{n,m} b_{n,m} \hat{S}_n^z \hat{S}_m^z \qquad b_{n,m} = -\frac{1}{4}(g\mu_B)^2 \hbar \frac{1 - 3\cos^2\theta_{nm}}{R_{nm}^3}$$

Central spin off resonance.

Our recent PRB studies several cases in depth.
Closely related to isotopically enriched Si:P.

“Canonical” Dipolar-only Problem



$$\mathcal{H}_{\text{eff}} = \sum_{n,m>0} b_{n,m} \hat{S}_n^+ \hat{S}_m^- - 2 \sum_{n,m} b_{n,m} \hat{S}_n^z \hat{S}_m^z \qquad b_{n,m} = -\frac{1}{4}(g\mu_B)^2 \hbar \frac{1 - 3\cos^2\theta_{nm}}{R_{nm}^3}$$

Note scale invariance: concentration $\longleftrightarrow$ time^{-1}.
The timescales of our figures are for g=2 electrons at a concentration of $10^{13}/cm^3$.

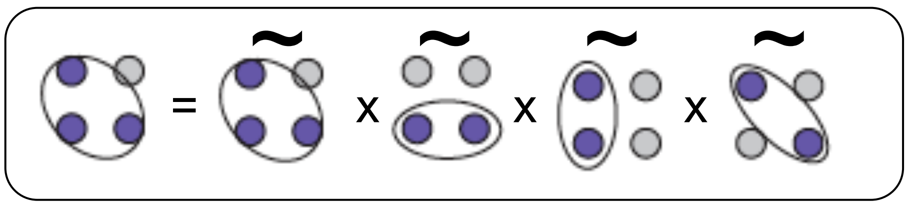
Adapting the Cluster Correlation Expansion

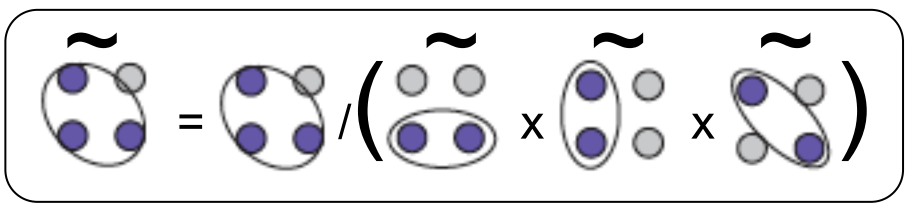
Cluster correlation expansion:

$$L = \rho_q^{+-}(t)/\rho_q^{+-}(0)$$

$L_{\mathcal{S}} = L$ with the exclusion of spins outside of $\mathcal{S}$: $\mathcal{S}$

$$L_{\mathcal{S}} = \prod_{\mathcal{C}\subseteq\mathcal{S}} \tilde{L}_{\mathcal{C}}, \quad \tilde{L}_{\mathcal{S}} = L_{\mathcal{S}} / \prod_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_{\mathcal{C}}$$





$$L_{\mathrm{CCE}}^{(k)} = \prod_{\|\mathcal{S}\|\leq k} \tilde{L}_{\mathcal{S}}.$$

Witzel et al., PRB 86, 035452 (2012)

Adapting the Cluster Correlation Expansion

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Cluster correlation expansion:

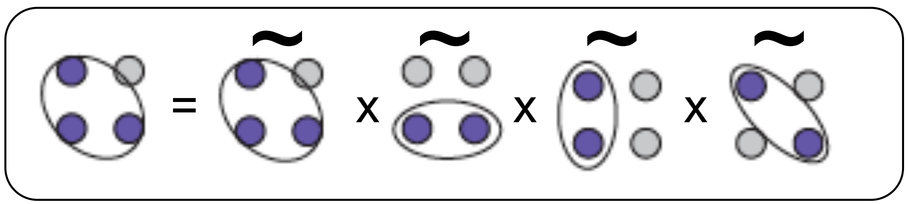
$$L = \langle \rho_q^{+-}(t) / \rho_q^{+-}(0) \rangle$$

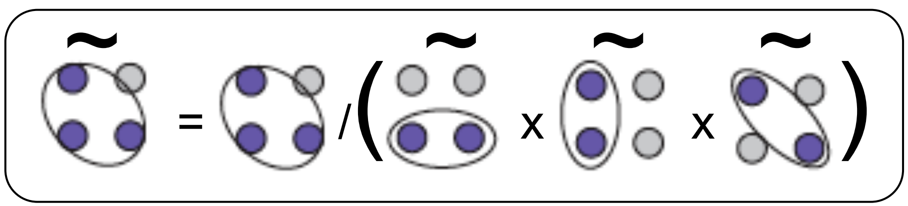
Internal Spin Averaging (numerical instability)

$L_{\mathcal{S}} = L$ only allowing flip-flops within set $\mathcal{S}$

External Awareness

$$L_{\mathcal{S}} = \prod_{\mathcal{C} \subseteq \mathcal{S}} \tilde{L}_{\mathcal{C}}, \quad \tilde{L}_{\mathcal{S}} = L_{\mathcal{S}} / \prod_{\mathcal{C} \subset \mathcal{S}} \tilde{L}_{\mathcal{C}}$$





Interlaced Spin Averaging: strategy we employ to successively approximate internal spin averaging.

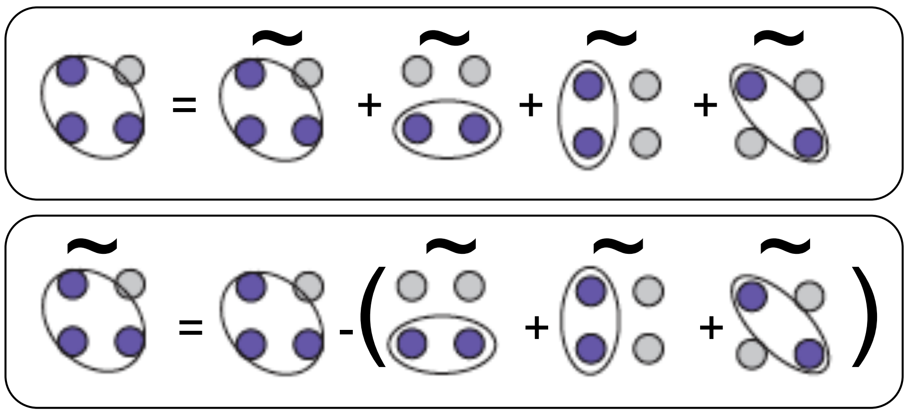
$$L_{\mathrm{CCE}}^{(k)} = \prod_{||\mathcal{S}|| \leq k} \tilde{L}_{\mathcal{S}}.$$

Easier Way to Obtain Numerical Stability: Additive Expansion

$$L_{\mathcal{S}} = \langle \rho_q^{+-}(t)/\rho_q^{+-}(0) \rangle - 1 \;\; ; \;\; L_{\mathcal{S}} = L$$

only allowing flip-flops within set $\mathcal{S}$

$$L = \sum_{\mathcal{S}} \tilde{L}_{\mathcal{S}}, \;\; L_{\mathcal{S}} = \sum_{\mathcal{C} \subseteq \mathcal{S}} \tilde{L}_{\mathcal{C}}, \;\; \tilde{L}_{\mathcal{S}} = L_{\mathcal{S}} - \sum_{\mathcal{C} \subset \mathcal{S}} \tilde{L}_{\mathcal{C}}$$

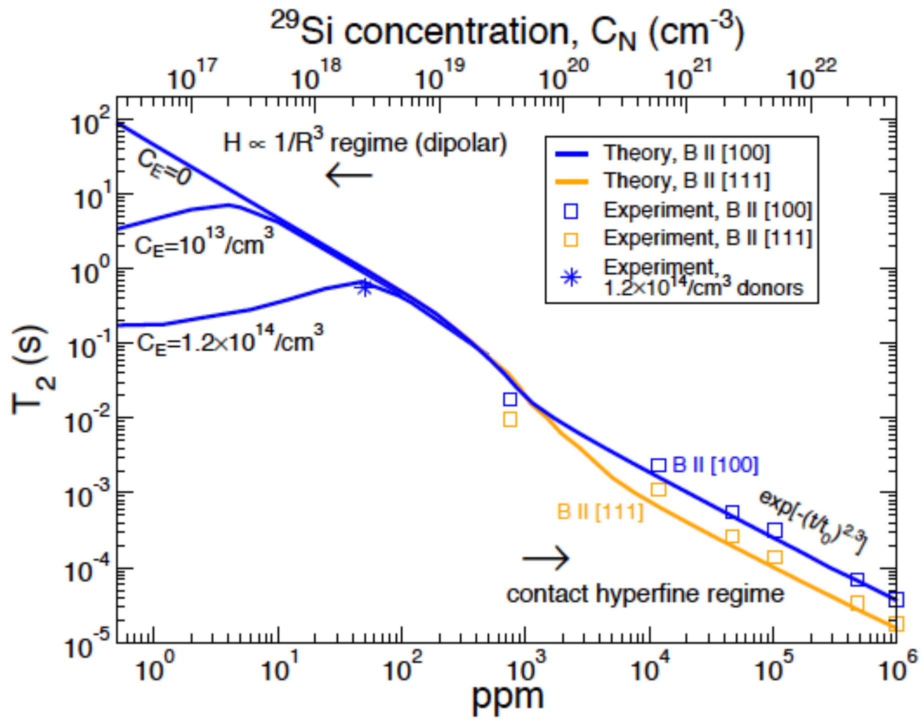


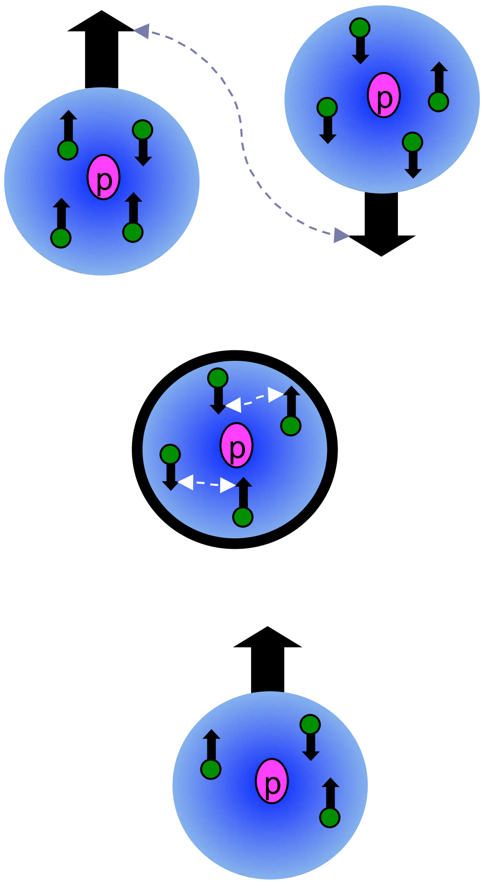
$$L_{\mathrm{CCE}}^{(k)} = \prod_{||\mathcal{S}|| \leq k} \tilde{L}_{\mathcal{S}}.$$

Witzel et al., arXiv:1307.2597 (2013)

Spectral Diffusion with Isotopic Enrichment

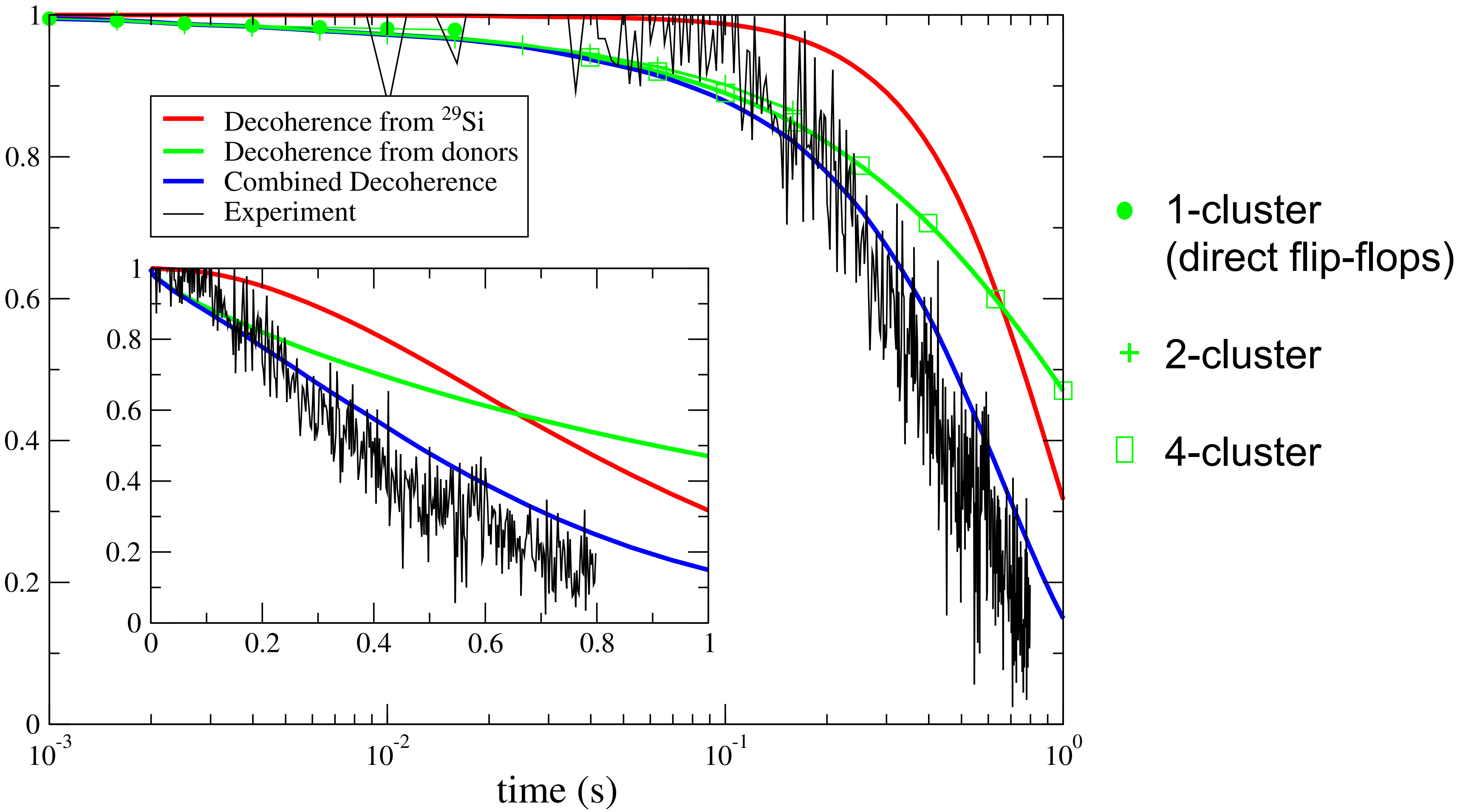
- Nuclear spins are eliminated with isotopic enrichment of Si.
- With few nuclear spins, other donor electron spins dominate decoherence.



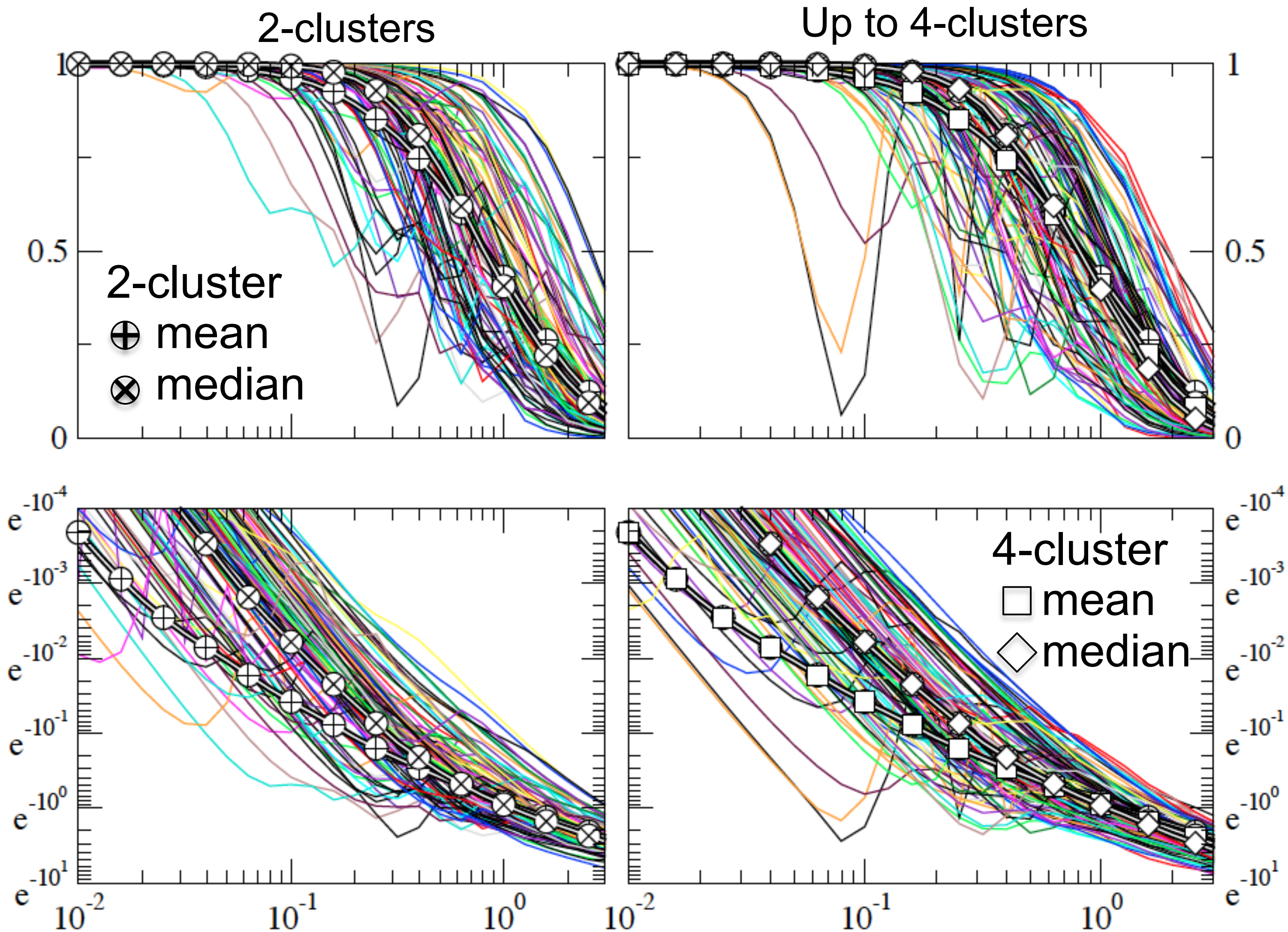


Witzel et al., PRL 105, 187602 (2010)

Enriched Si:P theory/experiment comparison

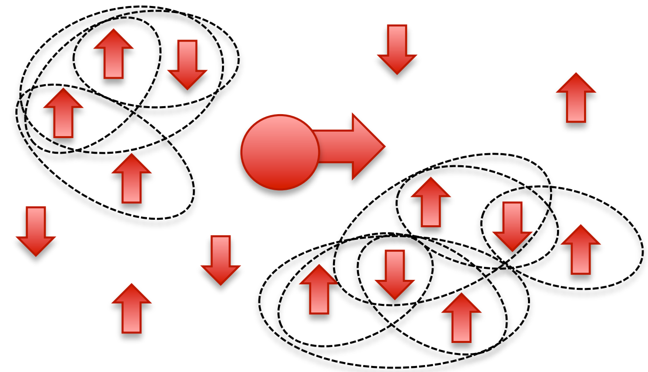


Hahn Echo for Many Spatial Realizations



Calculating Correlation Functions Using Spin-Cluster Approach

Use the additive cluster expansion with the correlation function as the quantity of interest (L).



$$C(t) = \langle \hat{B}_z(t)\hat{B}_z(0)\rangle, \text{ where } \mathcal{H} = \hat{B}_z\hat{S}_z + \mathcal{H}_B$$

$$L_\mathcal{S} = C(t) - C(0), \text{ freezing spins outside } \mathcal{S}$$

$$L_\mathcal{S} = \tilde{L}_\mathcal{S} + \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_\mathcal{C},$$

Contribution from cluster S

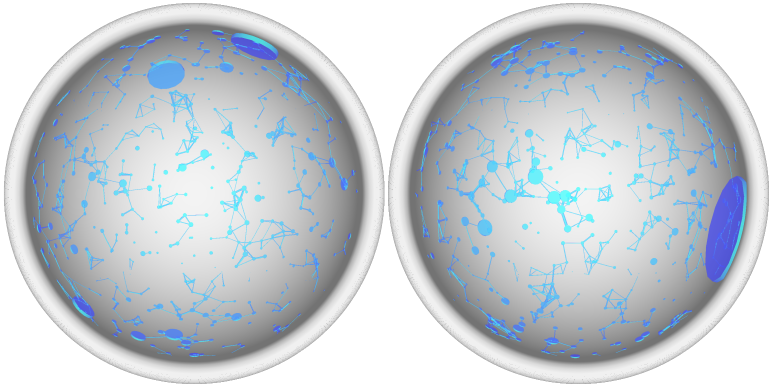
$$\tilde{L}_\mathcal{S} = L_\mathcal{S} - \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_\mathcal{C},$$

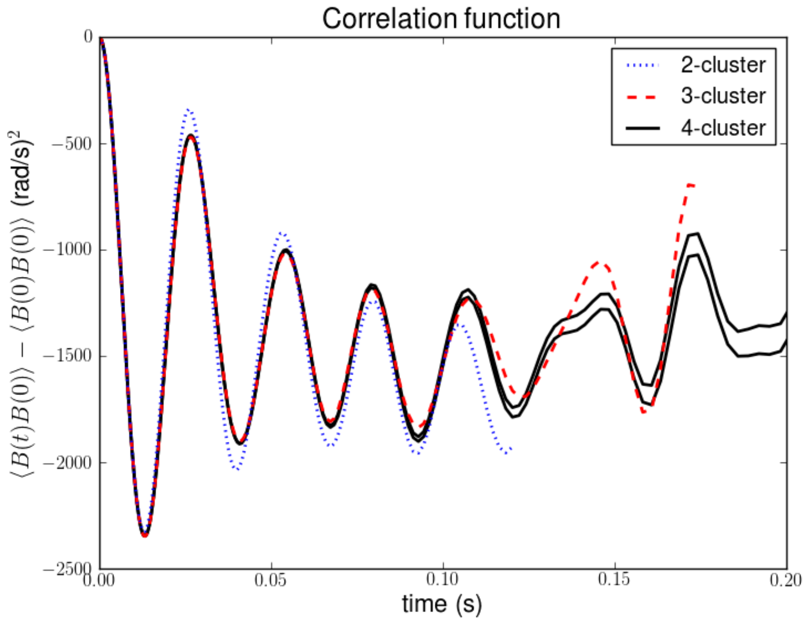
Correlation Function: Case A

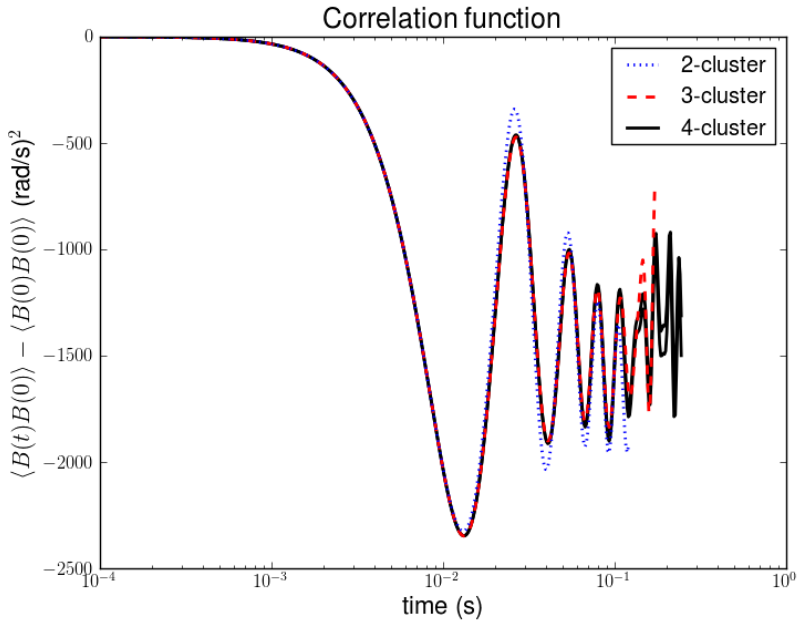
$$C(t) = \langle \hat{B}_z(t)\hat{B}_z(0)\rangle, \text{ where } \mathcal{H} = \hat{B}_z\hat{S}_z + \mathcal{H}_B$$

$$L_\mathcal{S} = C(t) - C(0), \text{ freezing spins outside } \mathcal{S}$$

$$L_\mathcal{S} = \tilde{L}_\mathcal{S} + \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_\mathcal{C}, \quad \tilde{L}_\mathcal{S} = L_\mathcal{S} - \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_\mathcal{C},$$







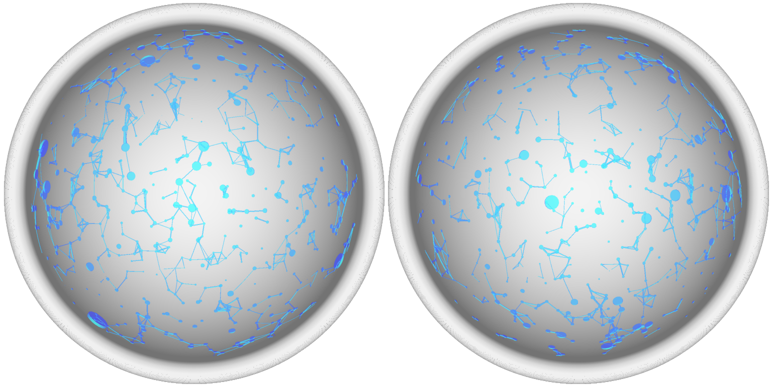
Witzel et al., arXiv:1307.2597 (2013)

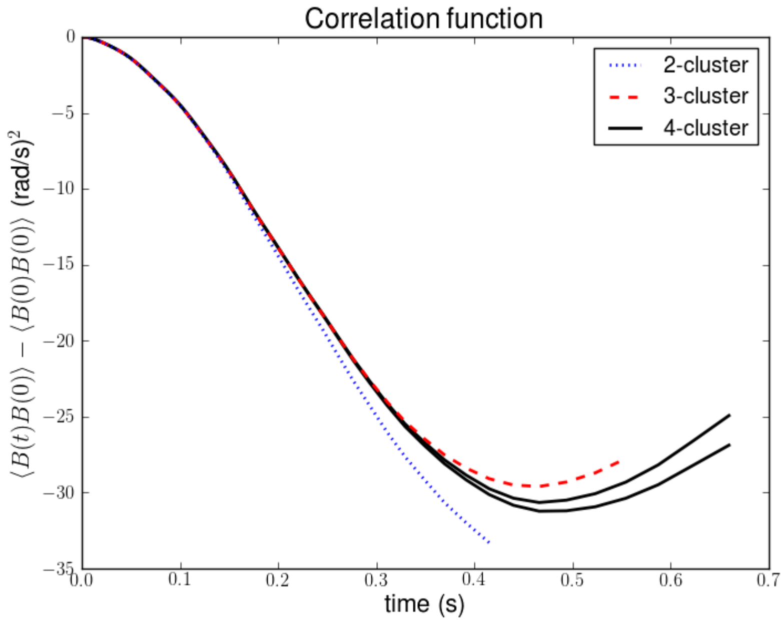
Correlation Function: Case B

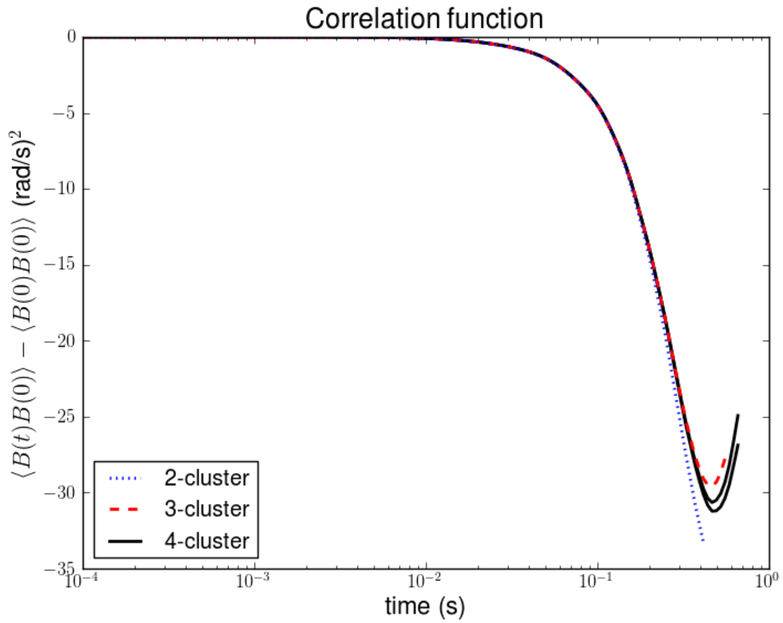
$$C(t) = \langle \hat{B}_z(t)\hat{B}_z(0)\rangle, \text{ where } \mathcal{H} = \hat{B}_z\hat{S}_z + \mathcal{H}_B$$

$$L_{\mathcal{S}} = C(t) - C(0), \text{ freezing spins outside } \mathcal{S}$$

$$L_{\mathcal{S}} = \tilde{L}_{\mathcal{S}} + \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_{\mathcal{C}}, \quad \tilde{L}_{\mathcal{S}} = L_{\mathcal{S}} - \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_{\mathcal{C}},$$







Correlation Function: Case C

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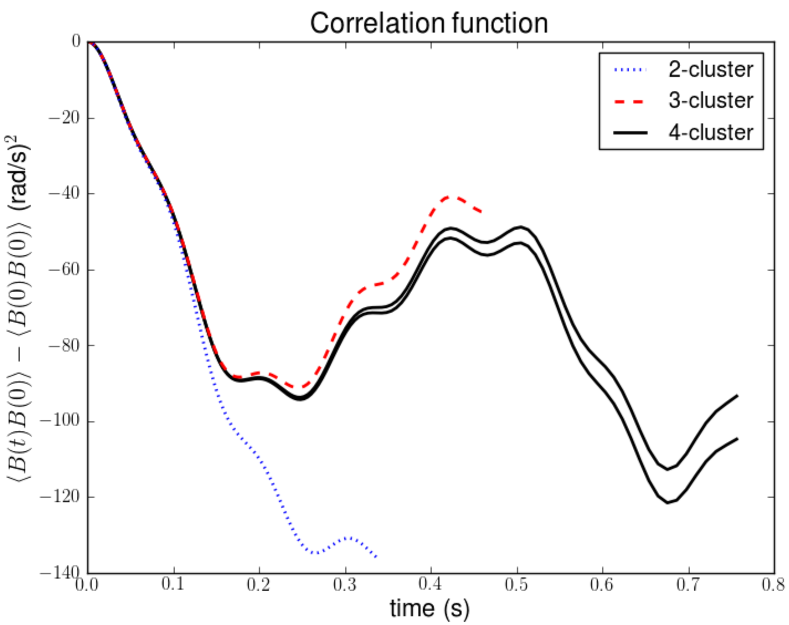
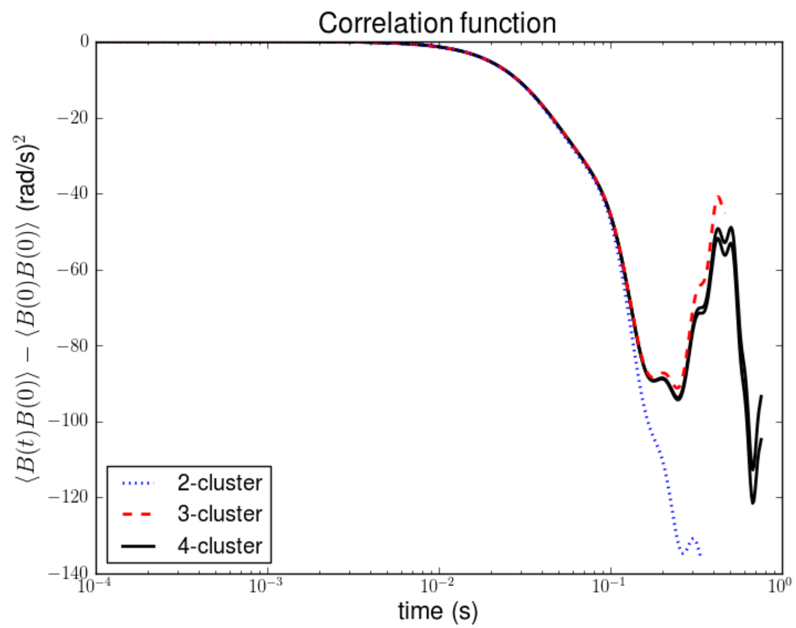
$$C(t) = \langle \hat{B}_z(t)\hat{B}_z(0)\rangle, \text{ where } \mathcal{H} = \hat{B}_z\hat{S}_z + \mathcal{H}_B$$

$$L_{\mathcal{S}} = C(t) - C(0), \text{ freezing spins outside } \mathcal{S}$$

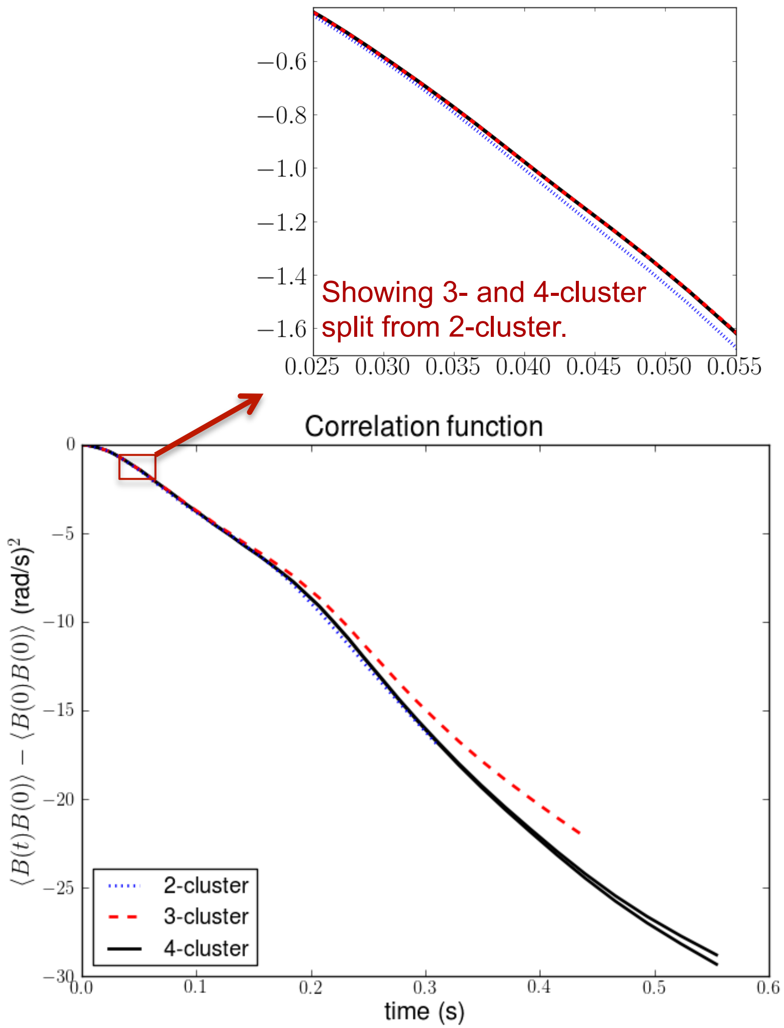
$$L_{\mathcal{S}} = \tilde{L}_{\mathcal{S}} + \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_{\mathcal{C}}, \quad \tilde{L}_{\mathcal{S}} = L_{\mathcal{S}} - \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_{\mathcal{C}},$$

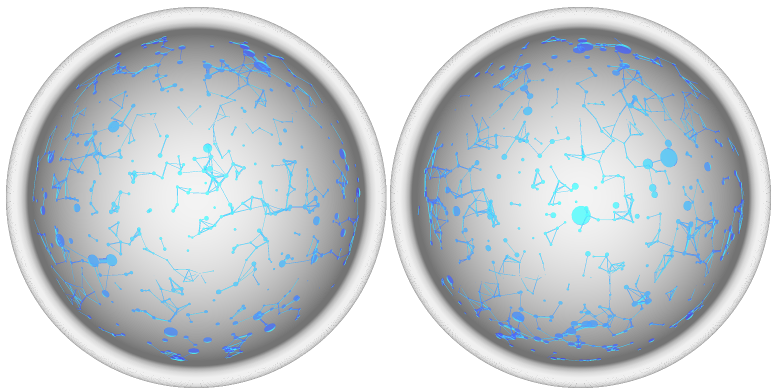


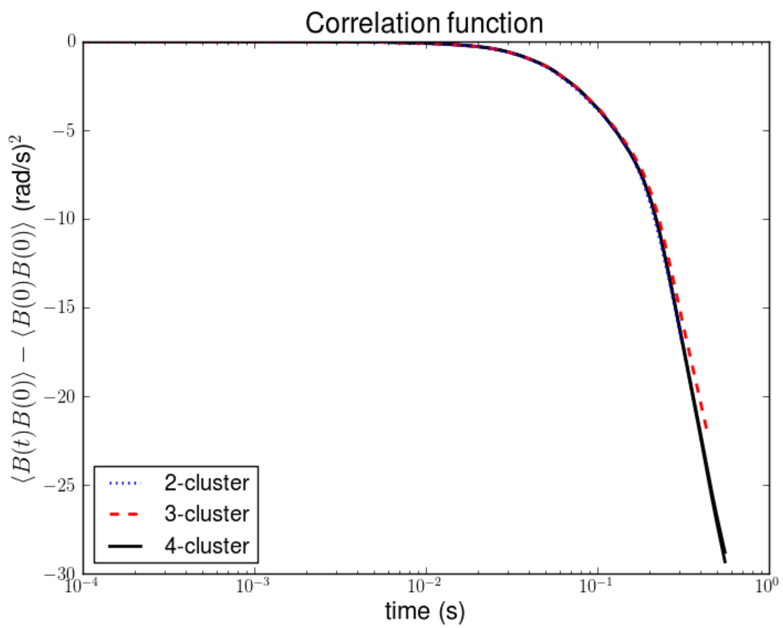


Witzel et al., arXiv:1307.2597 (2013)

Correlation Function: Case D







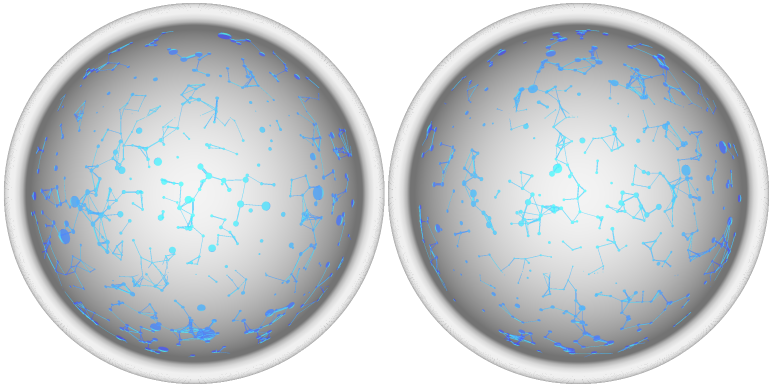
Witzel et al., arXiv:1307.2597 (2013)

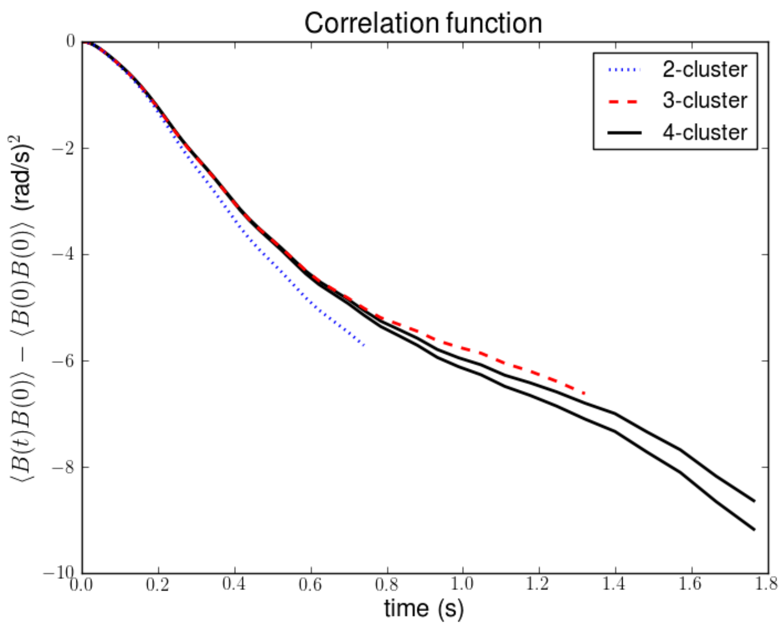
Correlation Function: Case E

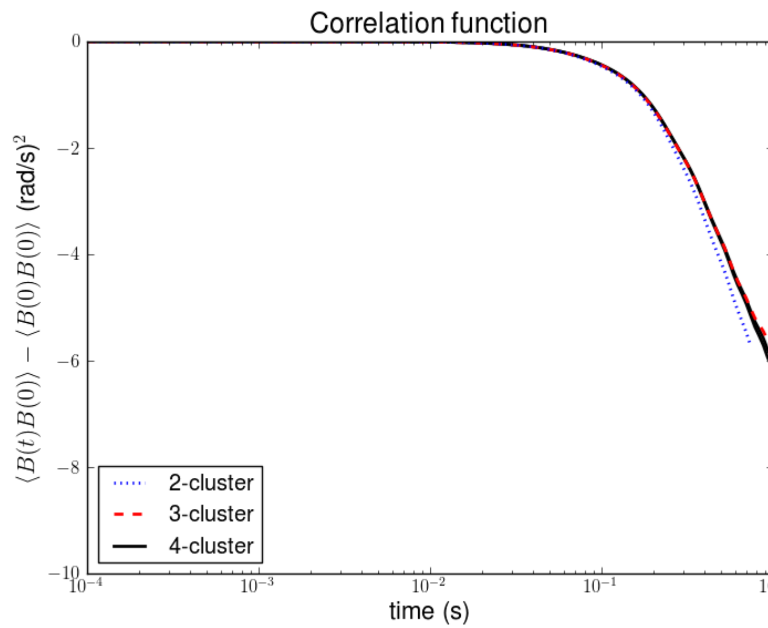
$$C(t) = \langle \hat{B}_z(t)\hat{B}_z(0)\rangle, \text{ where } \mathcal{H} = \hat{B}_z\hat{S}_z + \mathcal{H}_B$$

$$L_{\mathcal{S}} = C(t) - C(0), \text{ freezing spins outside } \mathcal{S}$$

$$L_{\mathcal{S}} = \tilde{L}_{\mathcal{S}} + \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_{\mathcal{C}}, \quad \tilde{L}_{\mathcal{S}} = L_{\mathcal{S}} - \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_{\mathcal{C}},$$







Correlation Function: Case F

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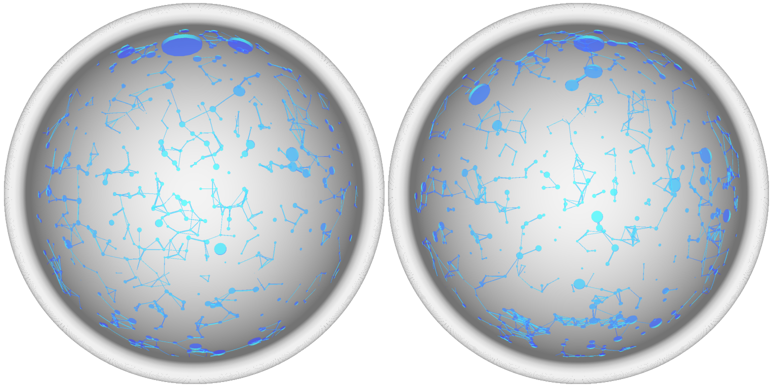
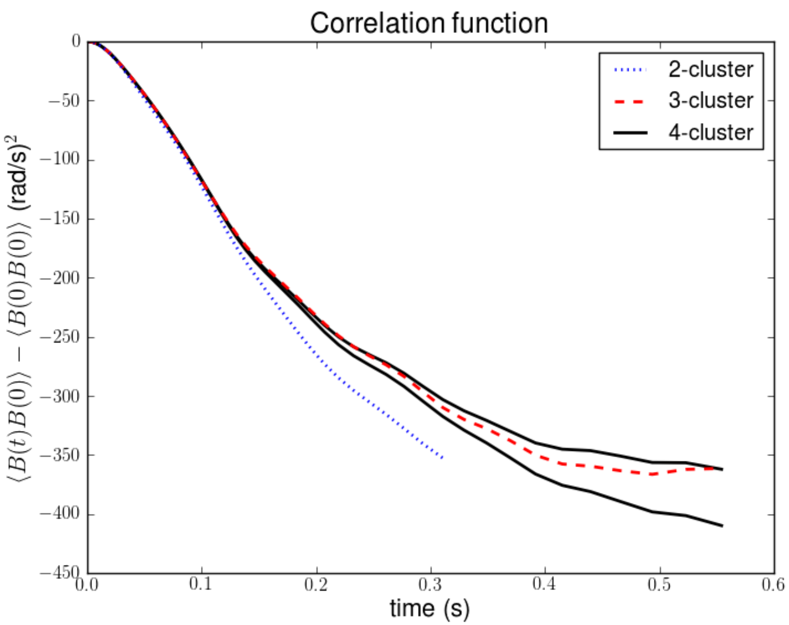
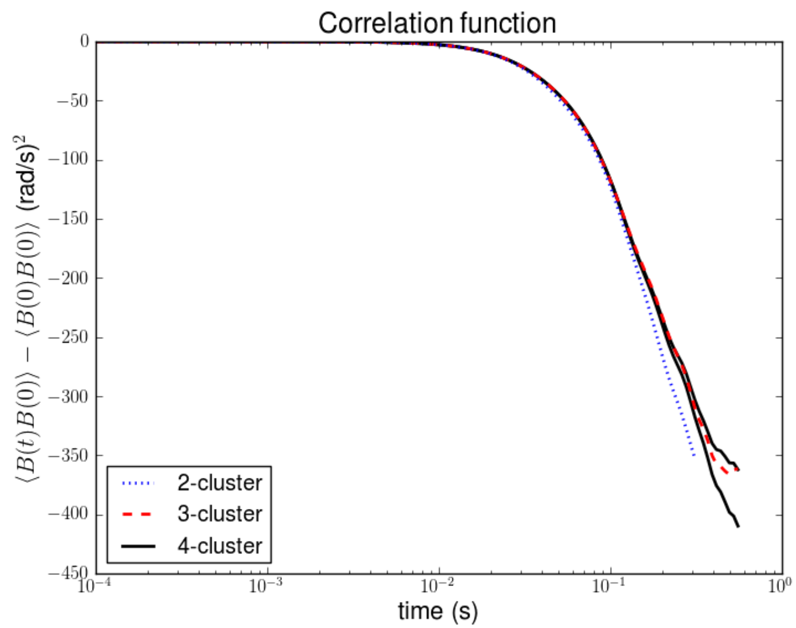
$$C(t) = \langle \hat{B}_z(t)\hat{B}_z(0)\rangle, \text{ where } \mathcal{H} = \hat{B}_z\hat{S}_z + \mathcal{H}_B$$

$$L_{\mathcal{S}} = C(t) - C(0), \text{ freezing spins outside } \mathcal{S}$$

$$L_{\mathcal{S}} = \tilde{L}_{\mathcal{S}} + \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_{\mathcal{C}}, \quad \tilde{L}_{\mathcal{S}} = L_{\mathcal{S}} - \sum_{\mathcal{C}\subset\mathcal{S}} \tilde{L}_{\mathcal{C}},$$







Witzel et al., arXiv:1307.2597 (2013)

Calculating Echo Decay via Correlation Function

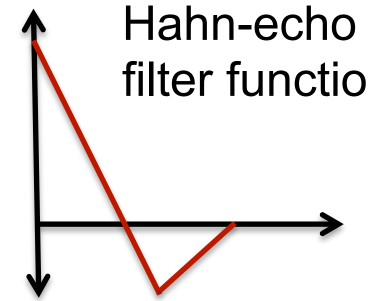
In rotating frame with "classical" field: $\tilde{H}(t) = B_z(t)y(t)\sigma_z$

Pi-pulse controls flip sign of B-field: $y(t) = \pm 1$

$$\begin{aligned}\langle \sigma_+(t) \rangle &= \frac{1}{2}\exp\left(-\int_0^t dt_1 \int_0^t dt_2 \, \langle B_z(t_1)B_z(t_2)\rangle \, y(t_1)y(t_2)\right) \\ &= \frac{1}{2}\exp\left(-\int_0^t du \, C(u)F_t(u)\right)\end{aligned}$$

Filter Function Defined By Pulse Sequence:

$$F_t(u) = \int_u^{2t-u} dv \; y\left(\frac{v+u}{2}\right) y\left(\frac{v-u}{2}\right)$$



Hahn-echo filter function

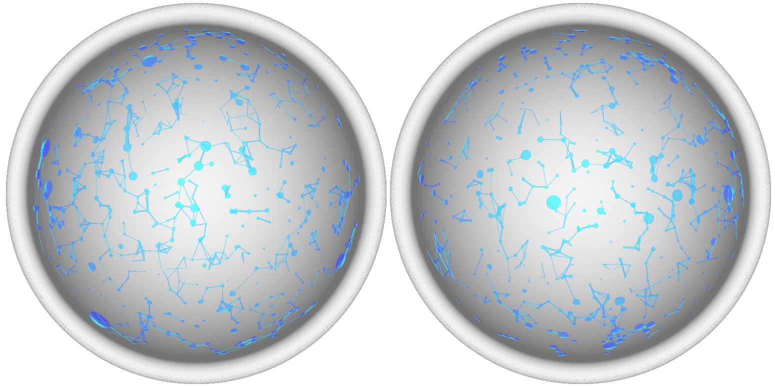
Coherence decay can be quickly and easily calculated from the filter function formalism, so can be easily incorporated into an optimal control loop.

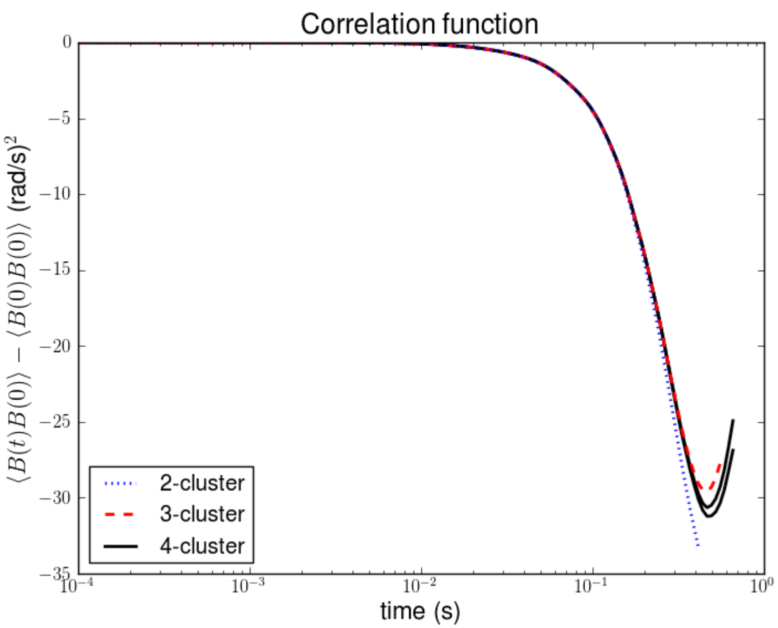
Echo Decay Comparison: Case B (we will look at Case A later)

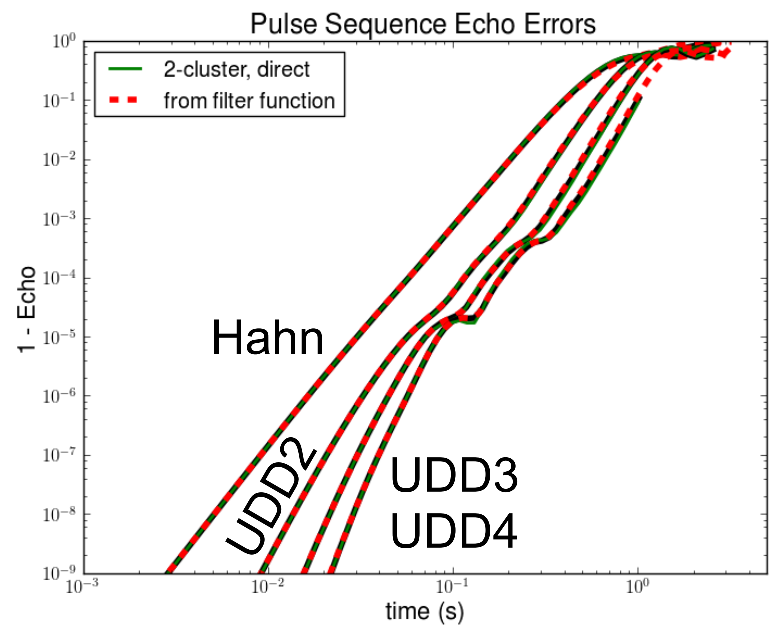
- Using correlation and filter functions:

$$\langle \sigma_+(t) \rangle = \frac{1}{2} \exp\left(-\int_0^t dt_1 \int_0^t dt_2 \, \langle B_z(t_1) B_z(t_2) \rangle \, y(t_1) y(t_2) \right)$$
$$= \frac{1}{2} \exp\left(-\int_0^t du \, C(u) F_t(u) \right)$$

- Compared with direct "interlaced spin averaging" approach.







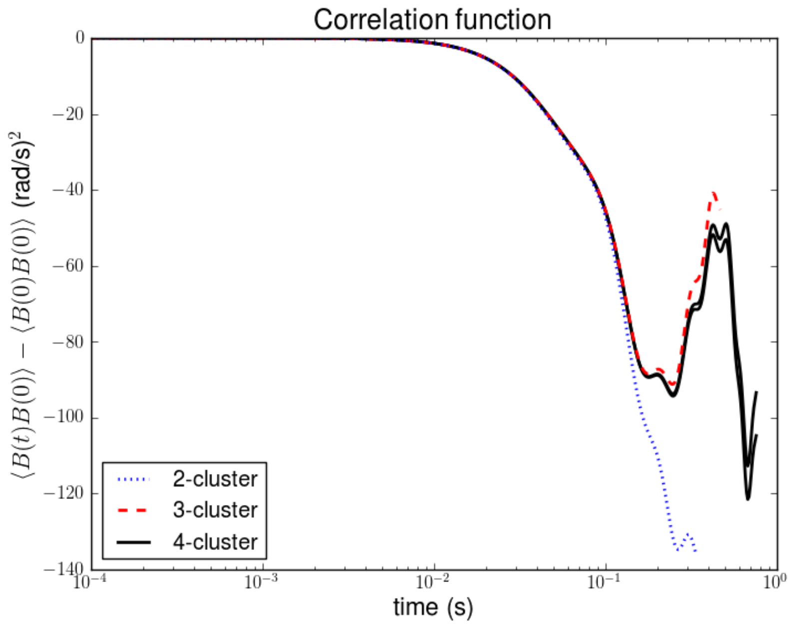
Witzel et al., arXiv:1307.2597 (2013)

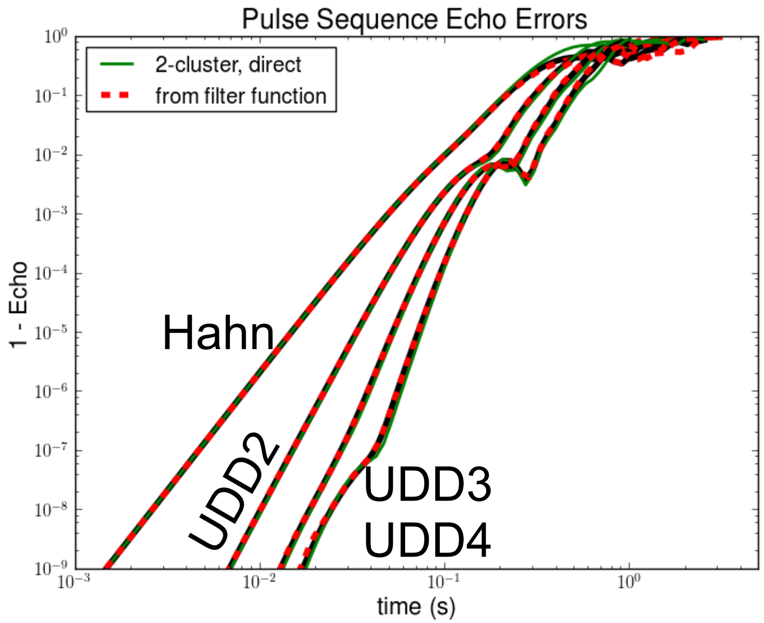
Echo Decay Comparison: Case C

- Using correlation and filter functions:

$$\langle \sigma_+(t) \rangle = \frac{1}{2} \exp\left(-\int_0^t dt_1 \int_0^t dt_2 \, \langle B_z(t_1) B_z(t_2) \rangle \, y(t_1) y(t_2) \right)$$
$$= \frac{1}{2} \exp\left(-\int_0^t du \, C(u) F_t(u) \right)$$

- Compared with direct "interlaced spin averaging" approach.





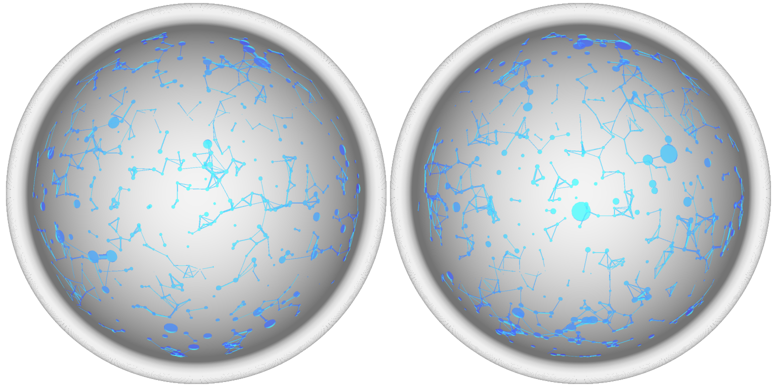
Witzel et al., arXiv:1307.2597 (2013)

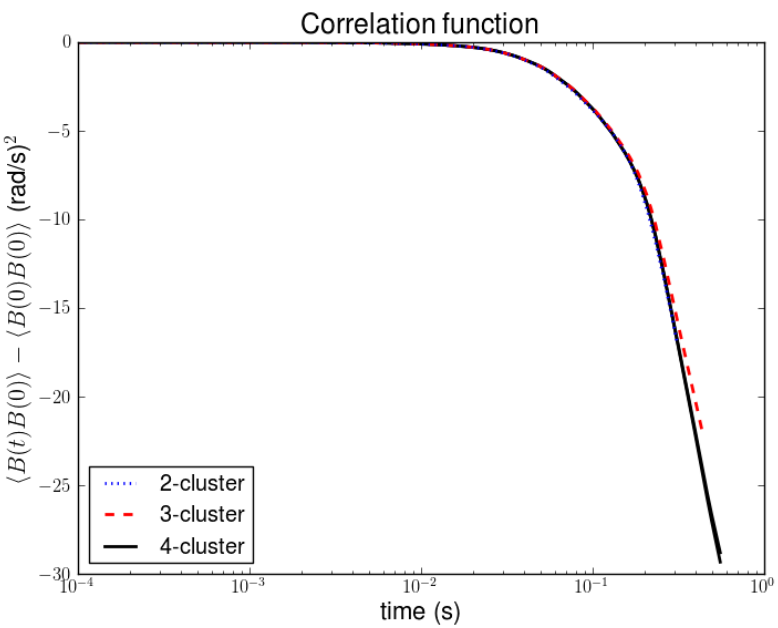
Echo Decay Comparison: Case D

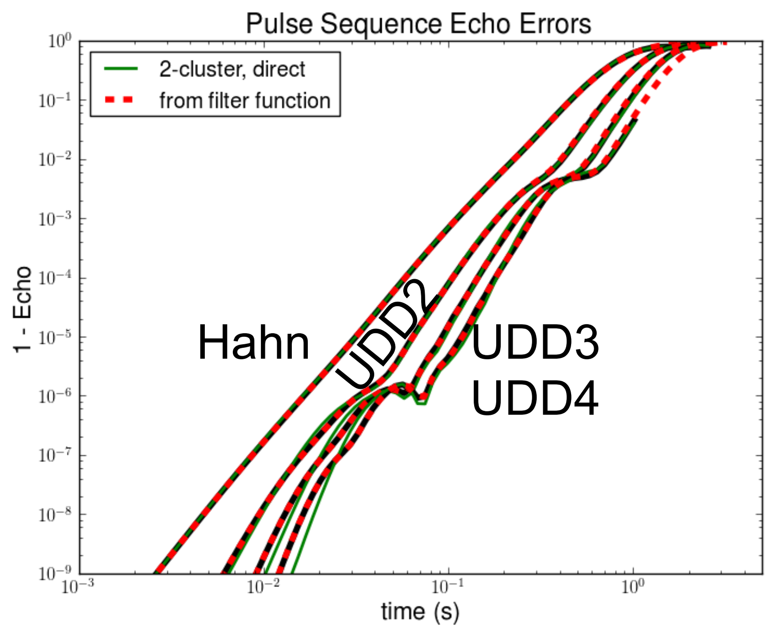
- Using correlation and filter functions:

$$\langle \sigma_+(t) \rangle = \frac{1}{2} \exp\left(- \int_0^t dt_1 \int_0^t dt_2 \, \langle B_z(t_1) B_z(t_2) \rangle \, y(t_1) y(t_2) \right)$$
$$= \frac{1}{2} \exp\left(- \int_0^t du \, C(u) F_t(u) \right)$$

- Compared with direct “interlaced spin averaging” approach.







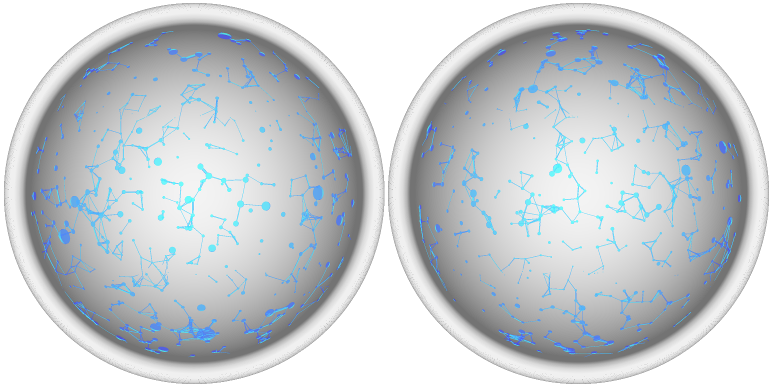
Witzel et al., arXiv:1307.2597 (2013)

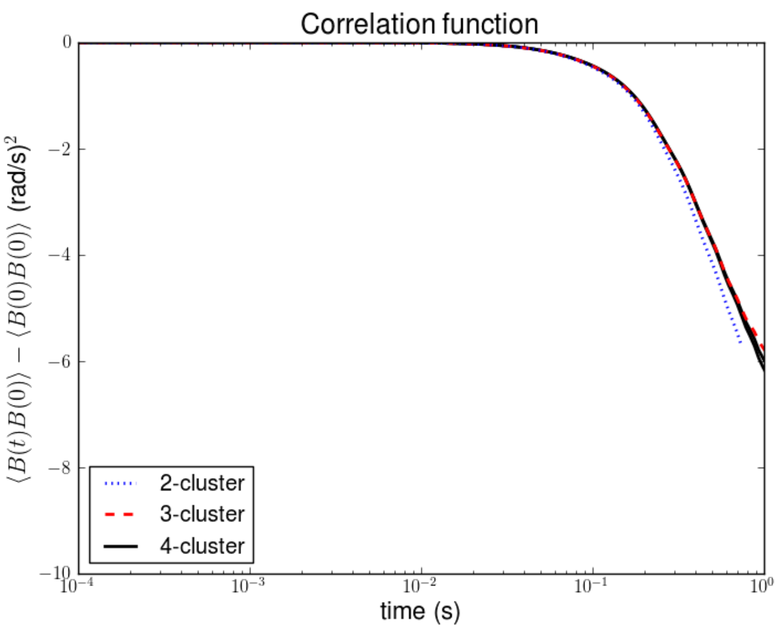
Echo Decay Comparison: Case E

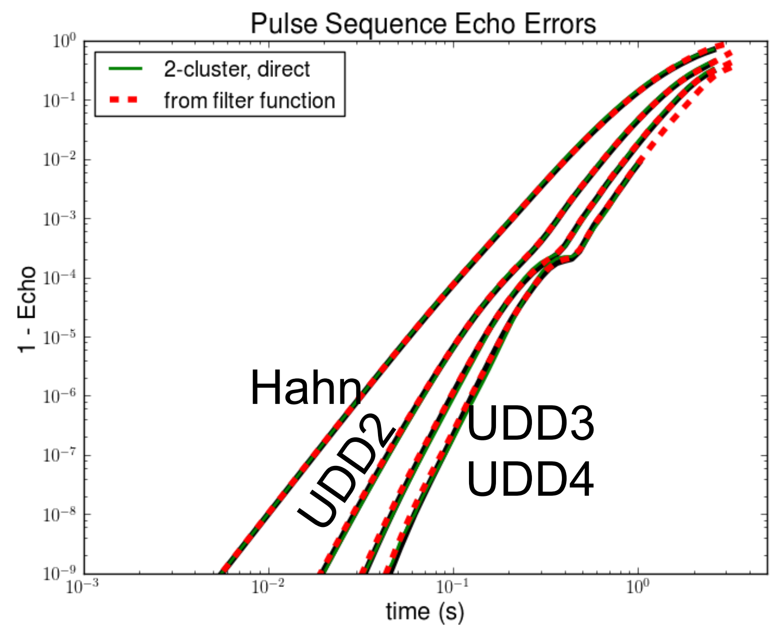
- Using correlation and filter functions:

$$\langle \sigma_+(t) \rangle = \frac{1}{2}\exp\left(-\int_0^t dt_1 \int_0^t dt_2 \, \langle B_z(t_1)B_z(t_2)\rangle \, y(t_1)y(t_2)\right)$$
$$= \frac{1}{2}\exp\left(-\int_0^t du \, C(u)F_t(u)\right)$$

- Compared with direct "interlaced spin averaging" approach.







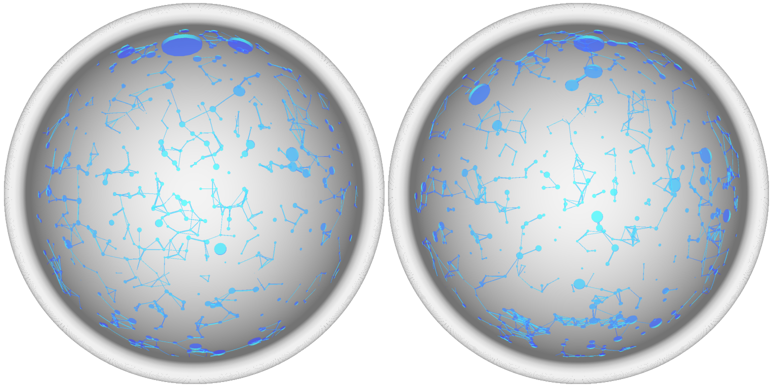
Witzel et al., arXiv:1307.2597 (2013)

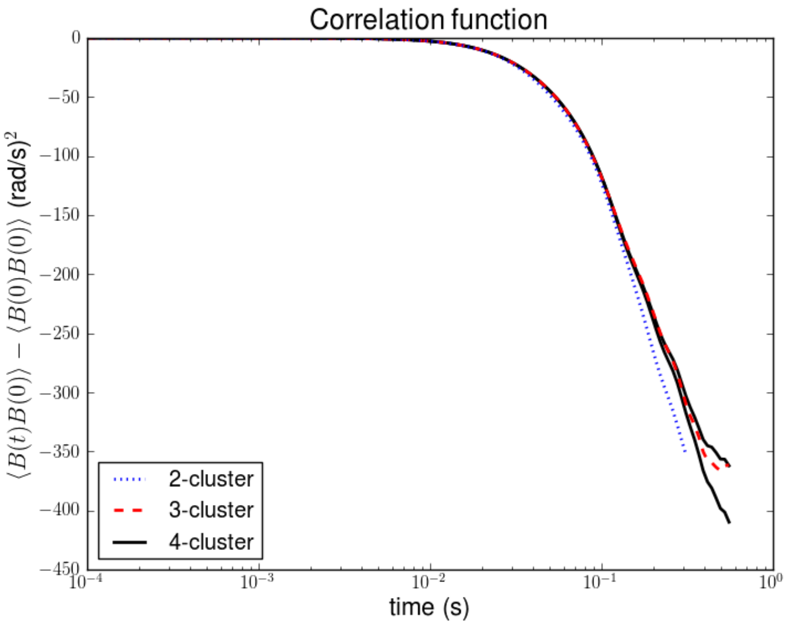
Echo Decay Comparison: Case F

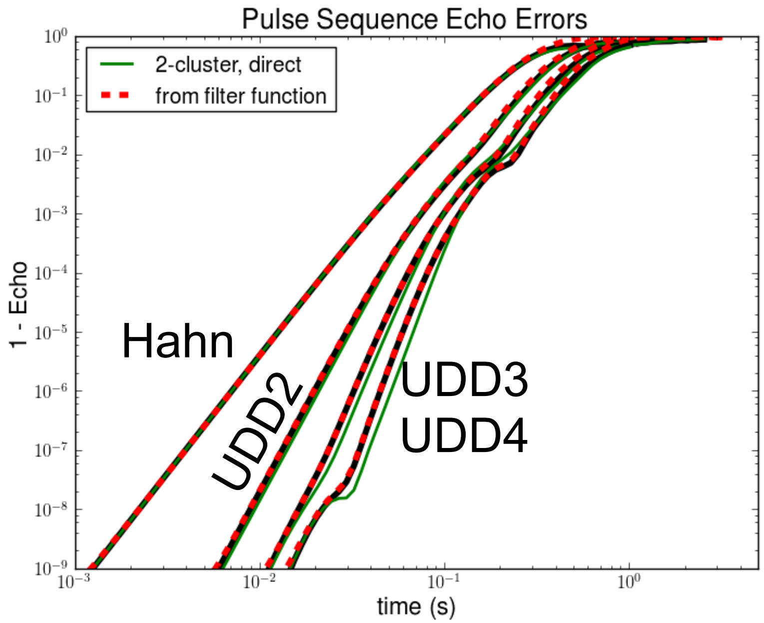
- Using correlation and filter functions:

$$\langle \sigma_+(t) \rangle = \frac{1}{2} \exp\left(- \int_0^t dt_1 \int_0^t dt_2 \, \langle B_z(t_1) B_z(t_2) \rangle \, y(t_1) y(t_2) \right)$$
$$= \frac{1}{2} \exp\left(- \int_0^t du \, C(u) F_t(u) \right)$$

- Compared with direct "interlaced spin averaging" approach.







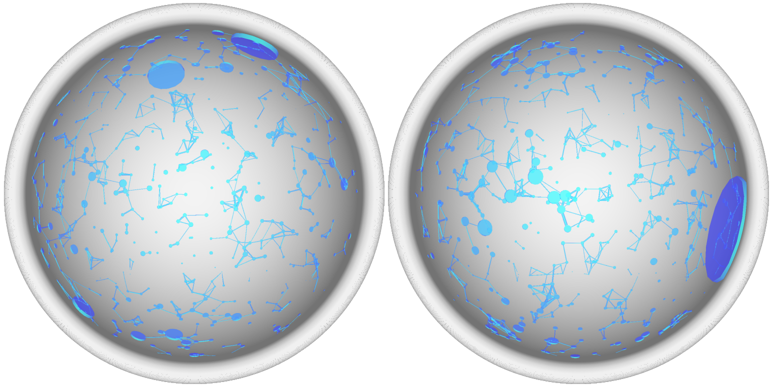
Witzel et al., arXiv:1307.2597 (2013)

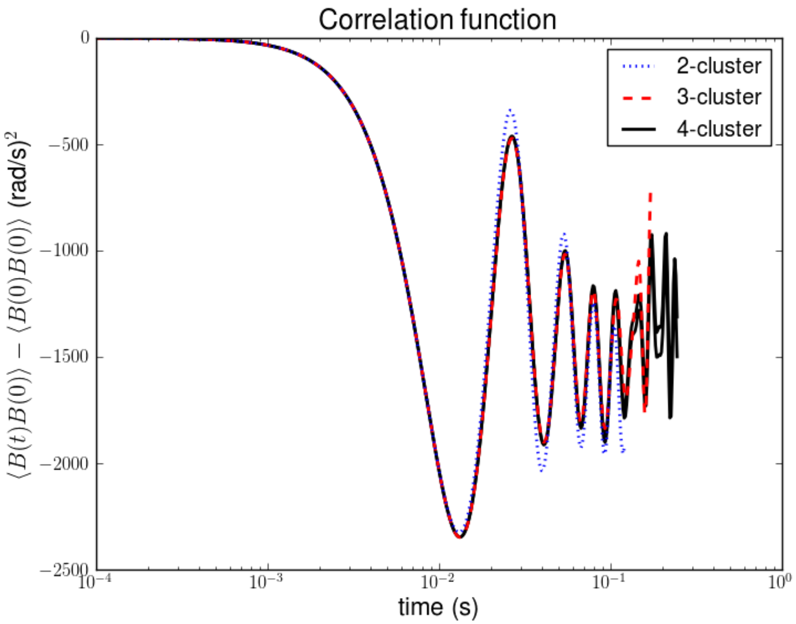
Echo Decay Comparison: Case A (an odd case)

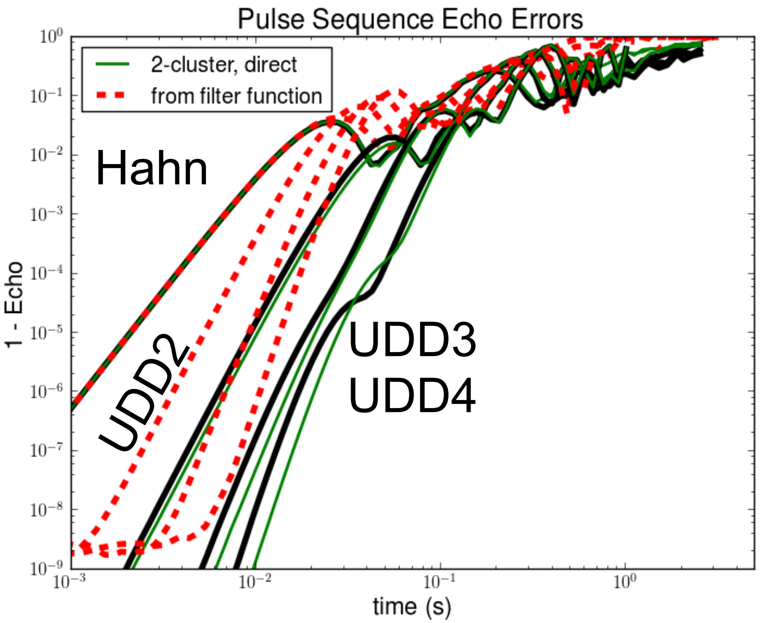
- Using correlation and filter functions:

$$\langle \sigma_+(t) \rangle = \frac{1}{2} \exp\left(- \int_0^t dt_1 \int_0^t dt_2 \, \langle B_z(t_1) B_z(t_2) \rangle \, y(t_1) y(t_2) \right)$$
$$= \frac{1}{2} \exp\left(- \int_0^t du \, C(u) F_t(u) \right)$$

- Compared with direct "interlaced spin averaging" approach.







Witzel et al., arXiv:1307.2597 (2013)

Optimizing Pulse Sequences

- Each spin state calculation of a 2-cluster spin-cluster calculation of 100 time points for a single pulse sequence takes about 1 minute (speed-ups are still possible).
- Computing 1000 different pulse sequences using correlation and filter functions takes about 1.5 minutes (in Python without optimization).
- We exploit the fact that filter functions for ideal pulses are piecewise linear.

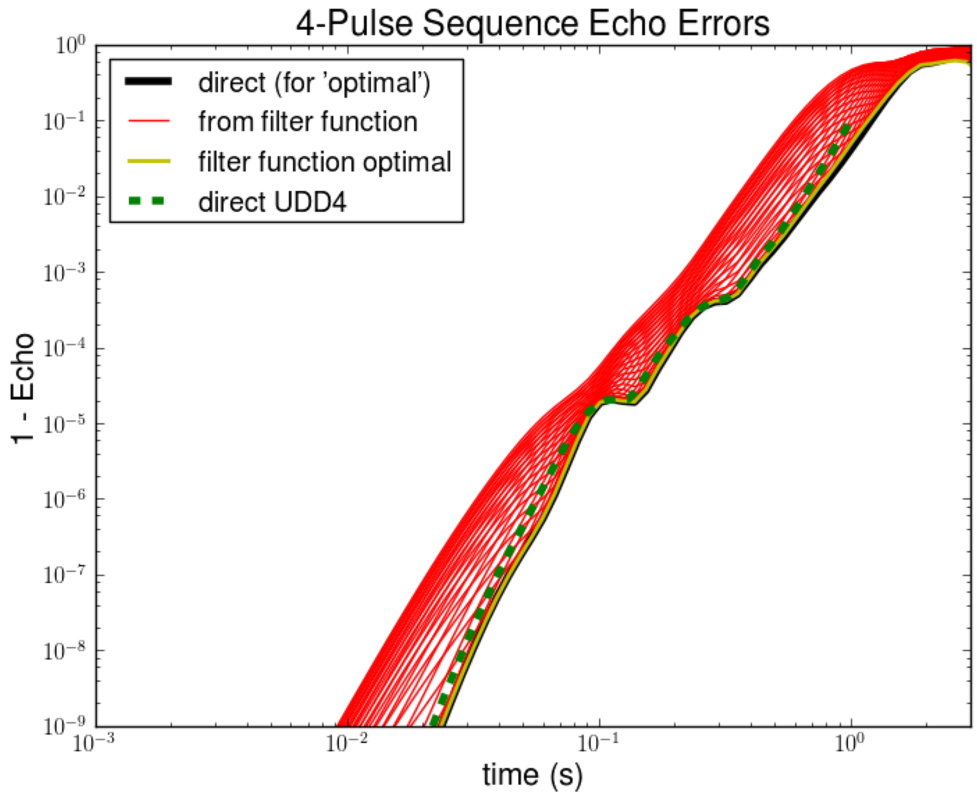
Precompute:

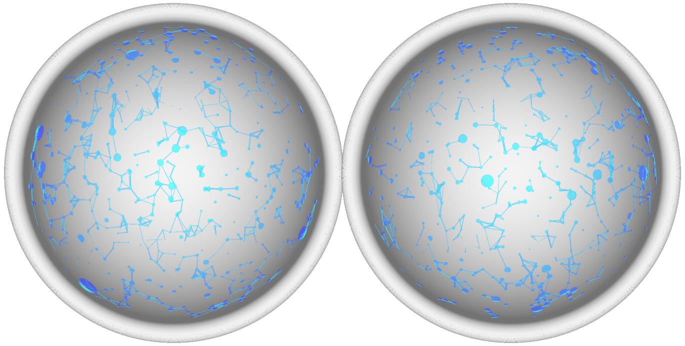
$$\begin{aligned} A(t) &= \int_0^t du\ C(u), \\ B(t) &= \int_0^t du\ uC(u) \end{aligned}$$

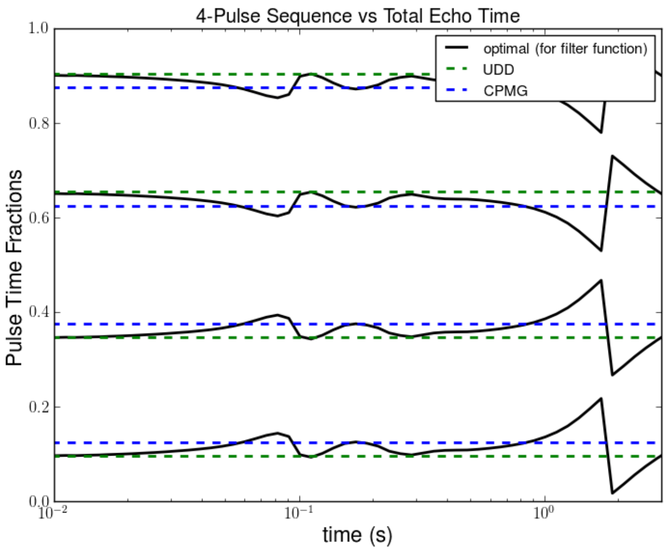
Now trivial:

$$\begin{aligned} F_t(u) &= \alpha(t) + \beta(t)u, \\ \langle \sigma_+(t) \rangle &= \frac{1}{2} \exp\left(- \int_0^t du\ C(u) F_t(u) \right) \\ &= \frac{1}{2} \exp\left(\alpha(t) A(t) + \beta(t) B(t) \right) \end{aligned}$$

Optimal 4-Pulse Sequence: Case B

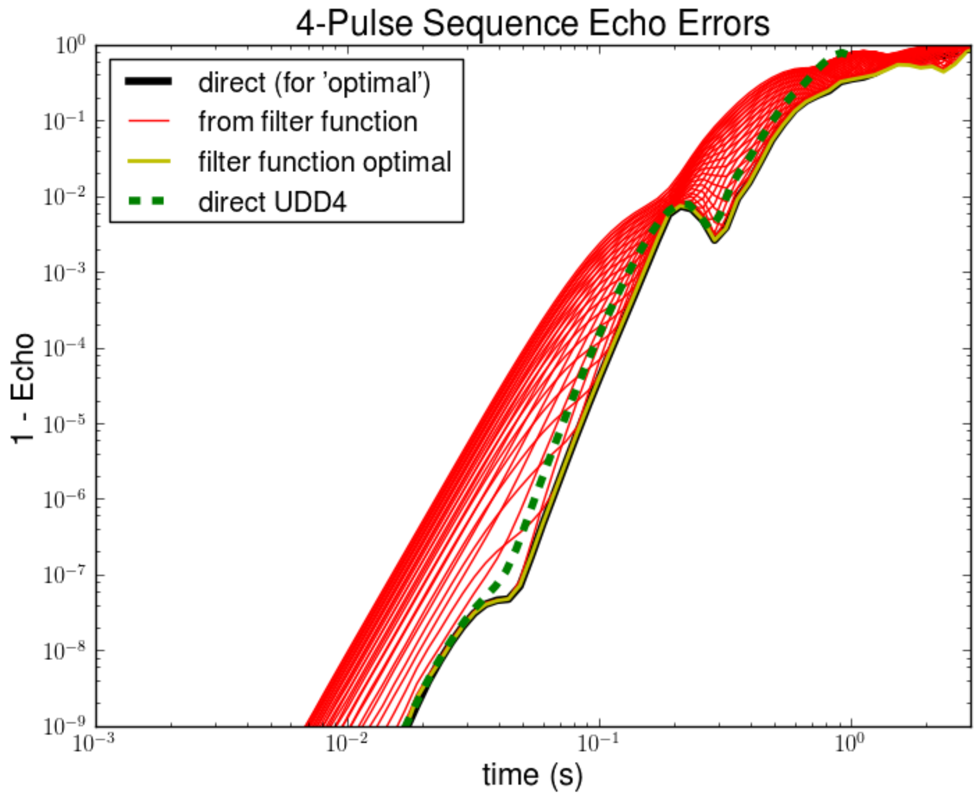


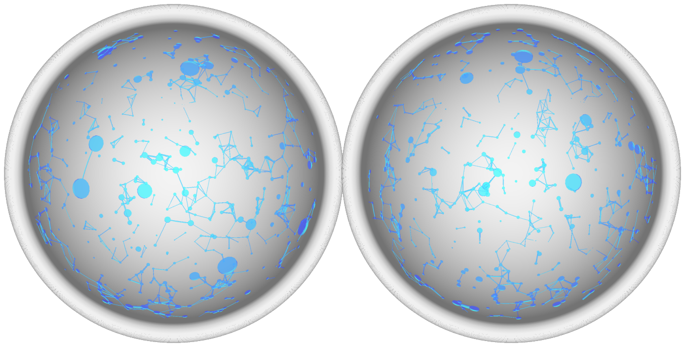


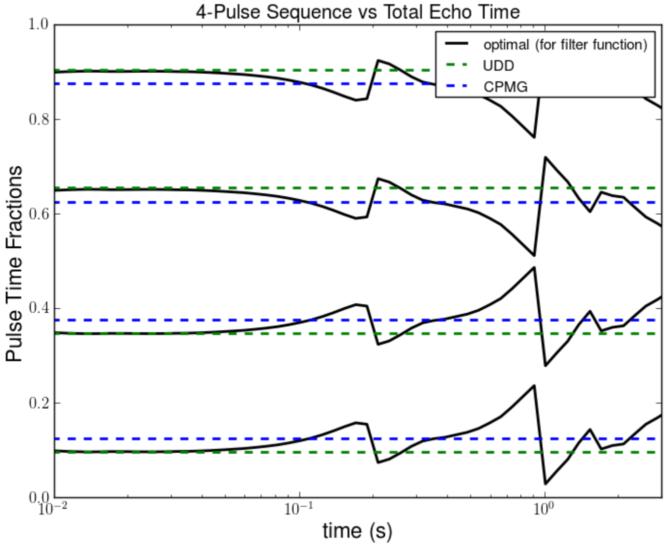


Witzel et al., arXiv:1307.2597 (2013)

Optimal 4-Pulse Sequence: Case C

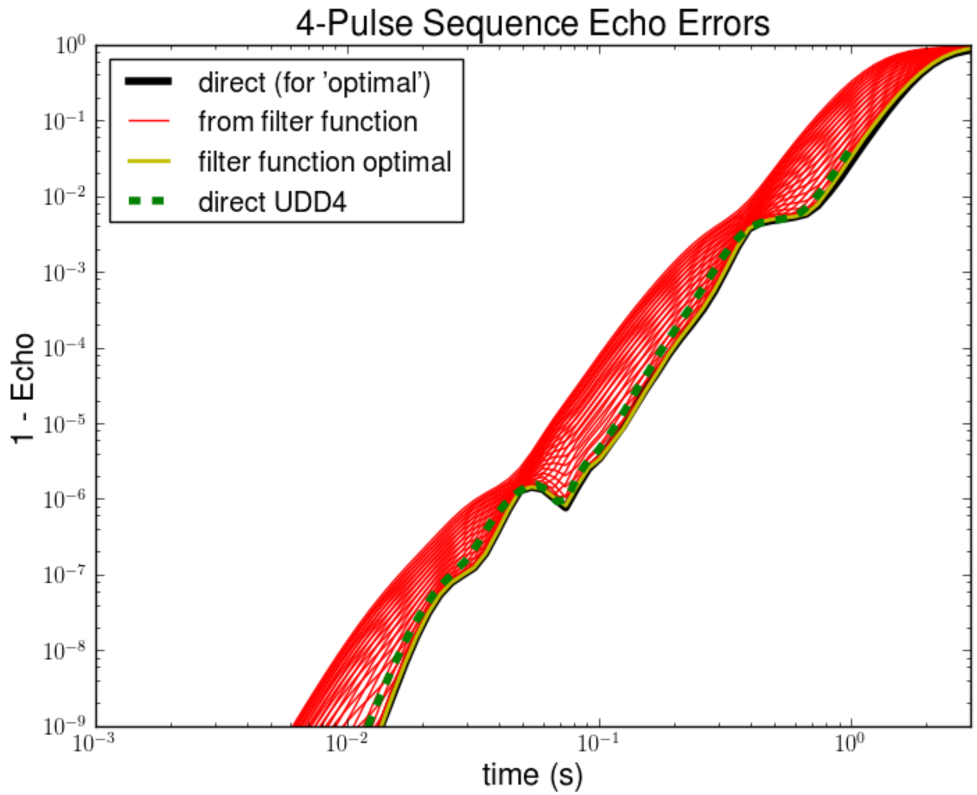


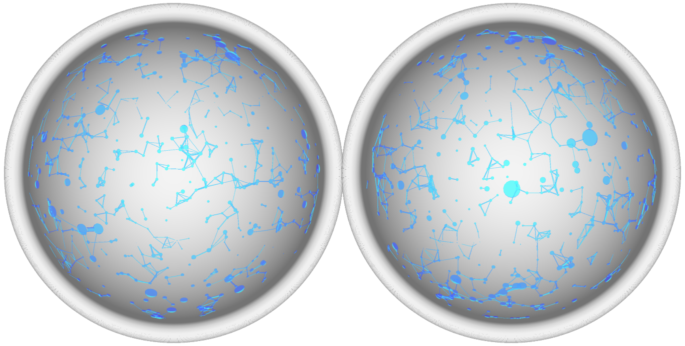


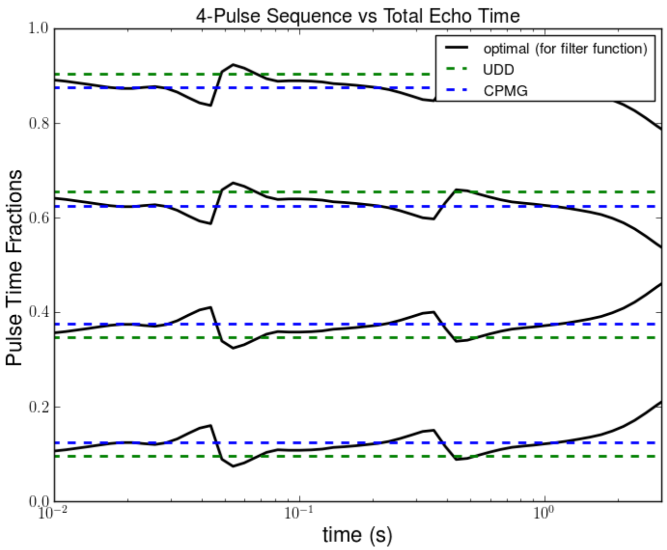


Witzel et al., arXiv:1307.2597 (2013)

Optimal 4-Pulse Sequence: Case D

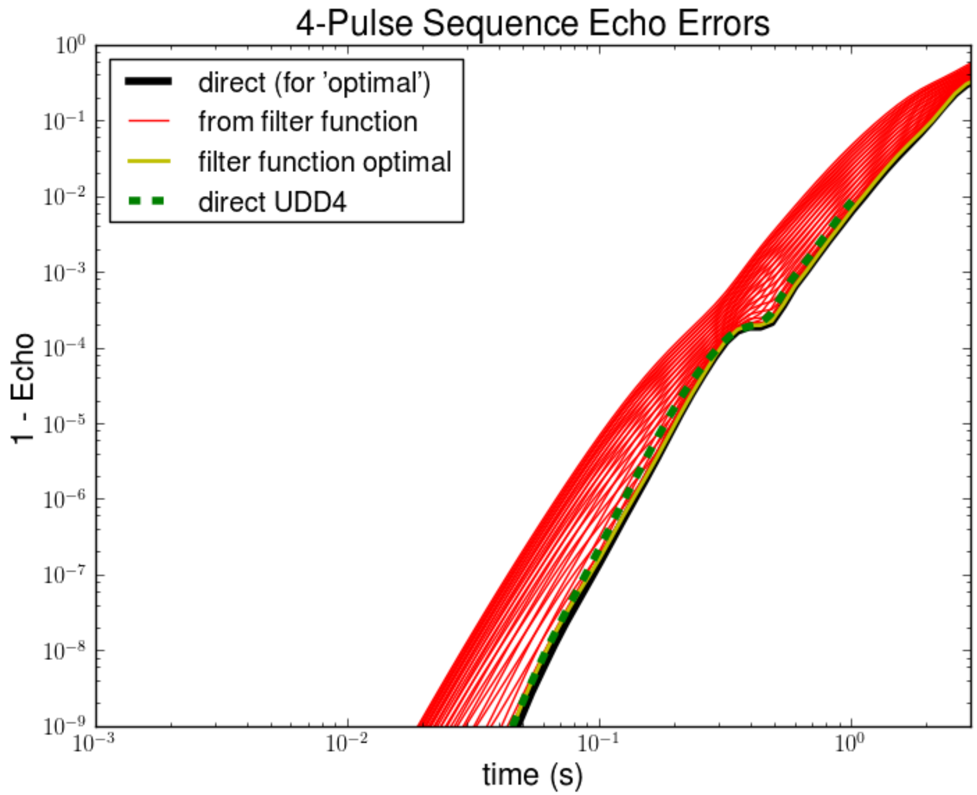


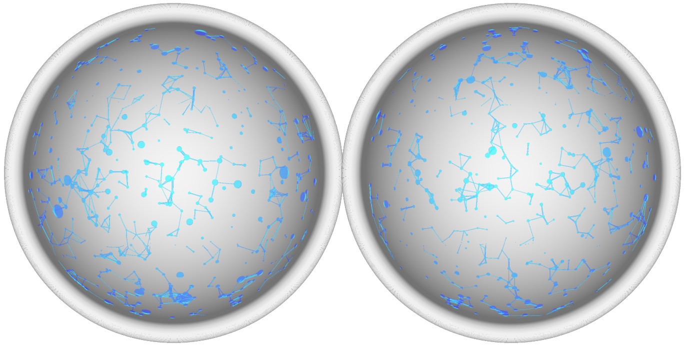


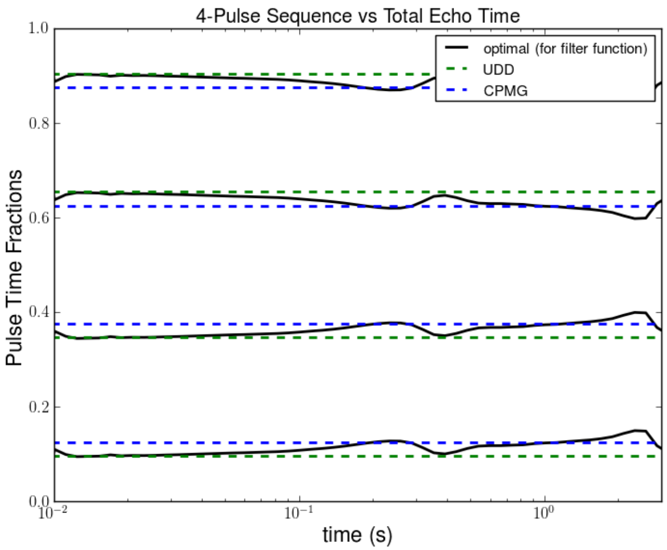


Witzel et al., arXiv:1307.2597 (2013)

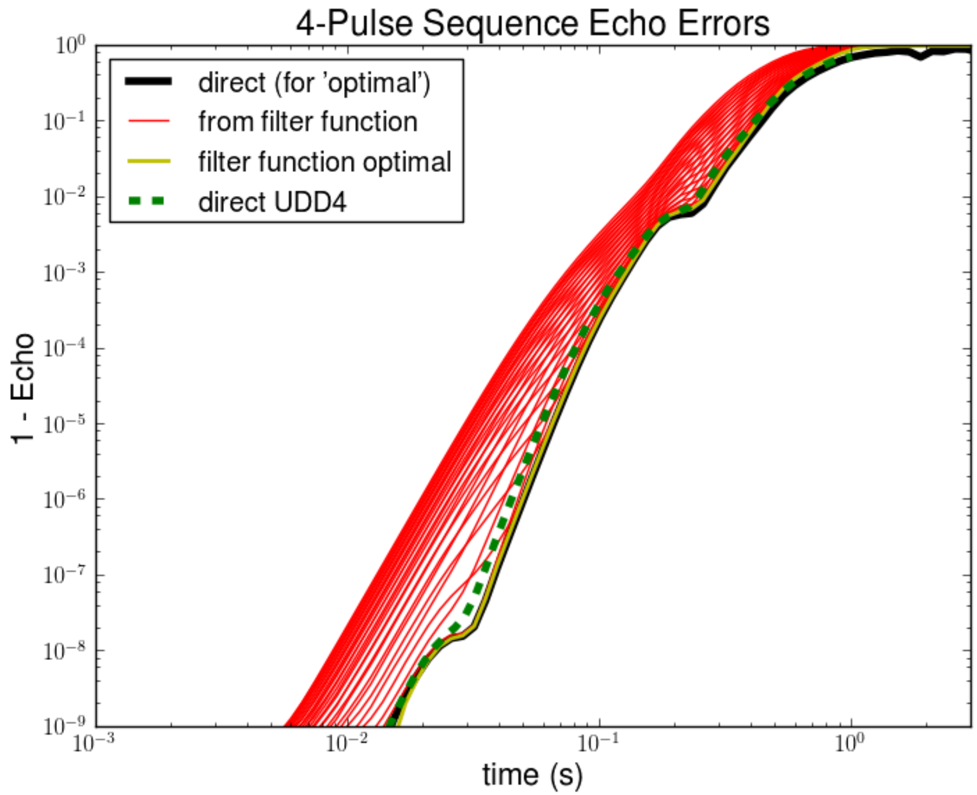
Optimal 4-Pulse Sequence: Case E

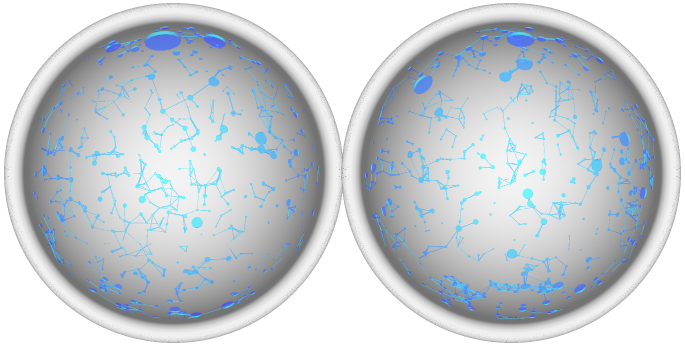


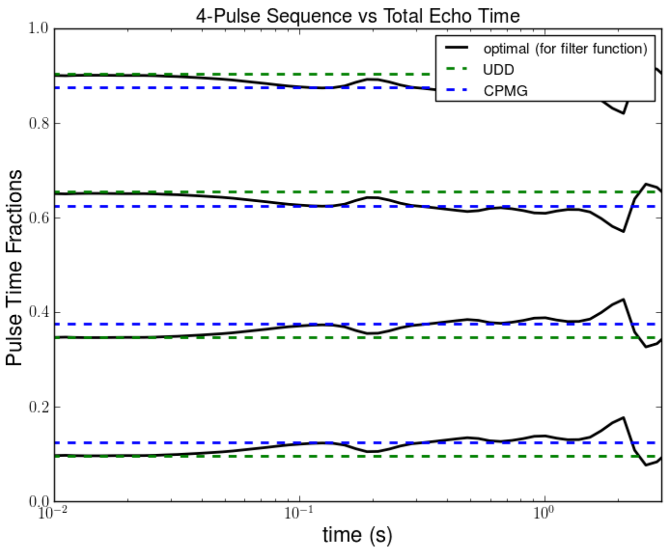




Optimal 4-Pulse Sequence: Case F

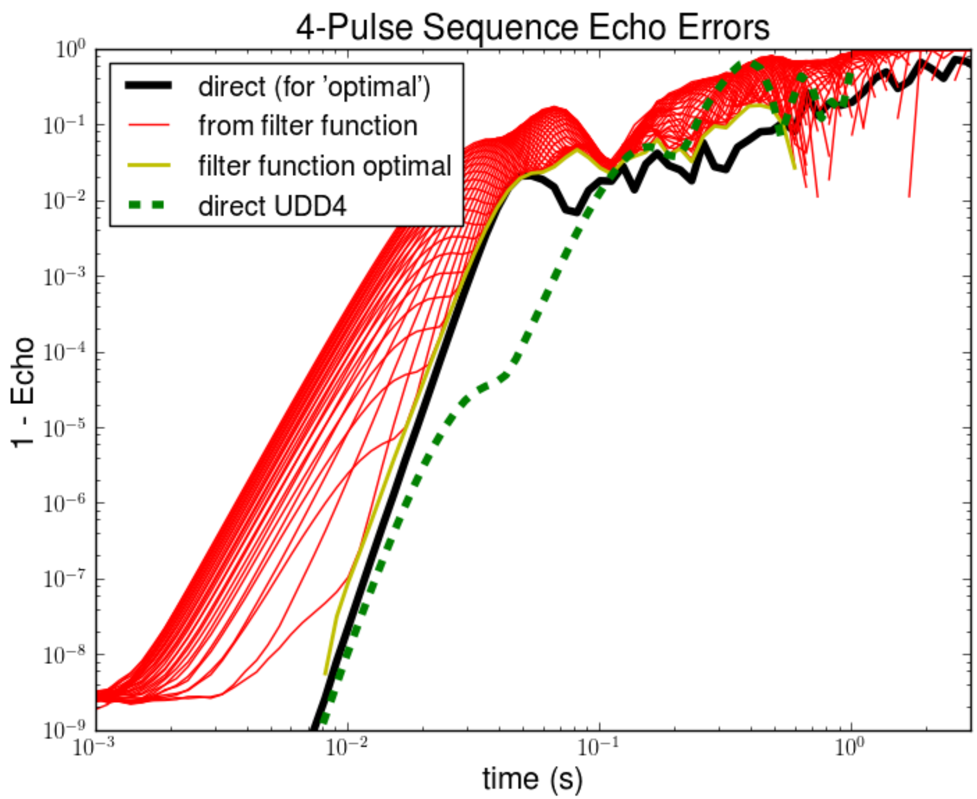


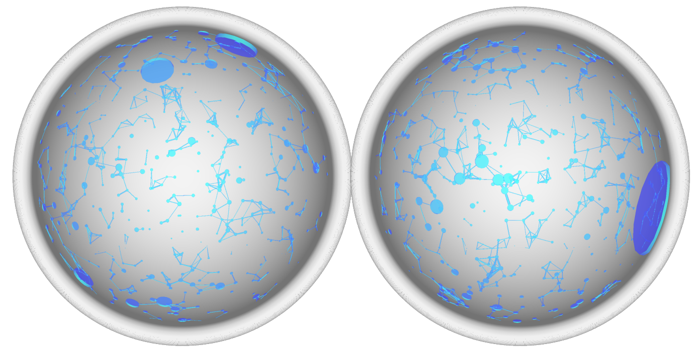


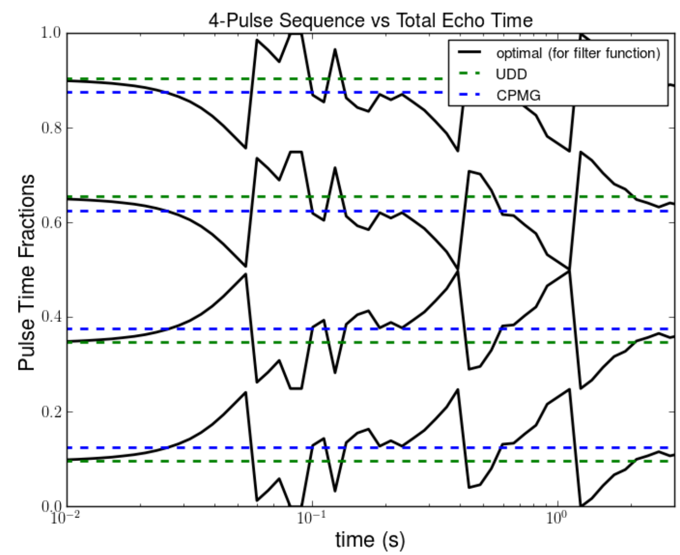


Witzel et al., arXiv:1307.2597 (2013)

Optimal 4-Pulse Sequence: Case A







Witzel et al., arXiv:1307.2597 (2013)

Summary of new findings

- Using a spin-cluster method, we've computed quantum (expectation value) correlation functions directly and shown its convergence behavior with cluster size.

- For a "canonical" all-dipolar problem, we find that the echo decay computed by assuming classical correlation functions matches well with direct calculations with some interesting exceptions.

- We've demonstrated the advantage of correlation functions for optimizing pulse sequences.