



Optimal Microwave Pulse Trains for Ramsey Metrology of ^{13}C in Diamond

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Quantum Physics

*[Submitted on 30 Apr 2026]***Optically detected nuclear magnetic resonance of carbon-13 in bulk diamond**

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Precision measurements based on optically detected nuclear magnetic resonance offer exquisite sensitivity to absolute shifts in spin transition frequencies, with potential applications in fundamental physics experiments and inertial sensing. We investigate ^{13}C nuclear spins in diamond as a candidate system for solid-state implementations, which hold the promise for high-fidelity readout of large numbers of coherent nuclear spins in millitesla or lower magnetic fields. We demonstrate a technique that allows for both optical polarization and readout of large ensembles of $\sim 10^4$ polarized nuclear spins. Our method takes advantage of state-selective Landau-Zener transitions under microwave frequency sweeping, which bidirectionally transfer spin polarization between Nitrogen-Vacancy (NV) electron spins and remote ^{13}C nuclear spins. Using natural isotopic abundance diamonds with nitrogen densities of ~ 0.5 – 10 ppm, we perform optically-detected ^{13}C Ramsey spectroscopy and realize a nuclear-spin-dependent fluorescence contrast exceeding 0.5% peak-to-peak. We observe nuclear spin dephasing times $T_2^* \sim 2$ ms that only modestly improve with homonuclear dipolar decoupling, indicating that they are limited by the longitudinal spin relaxation of nearby NV electron spins. We study the magnetic field dependence of the optical readout and find comparable contrast and dephasing times for magnetic fields in the range 8–20 mT. Our method can be interpreted as a type of repetitive readout, where each NV center optically reads out the spin state of ~ 100 nuclei before nuclear spins depolarize.

Comments: 25 pages, 17 figures, 7 tables, 70 refs

Subjects: **Quantum Physics (quant-ph)**; Mesoscale and Nanoscale Physics (cond-mat.mes-hall); Atomic Physics (physics.atom-ph); Instrumentation and Detectors (physics.ins-det); Optics (physics.optics)

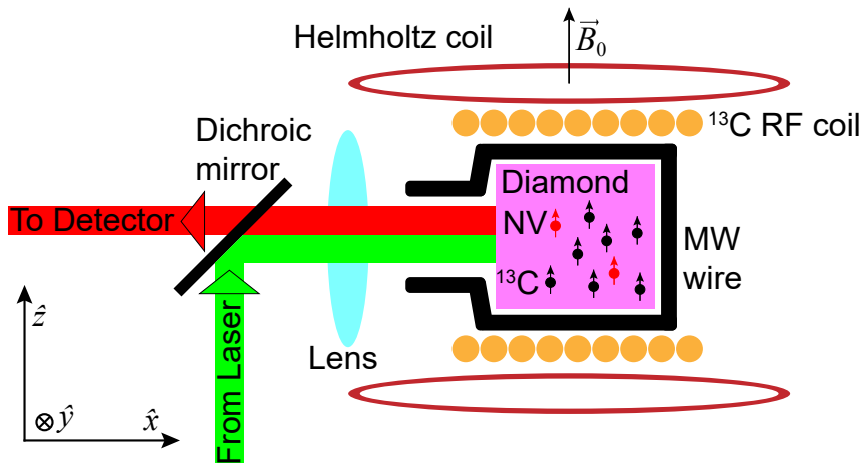
Cite as: arXiv:2605.00152 [quant-ph]
(or arXiv:2605.00152v1 [quant-ph] for this version)

<https://doi.org/10.48550/arXiv.2605.00152> 



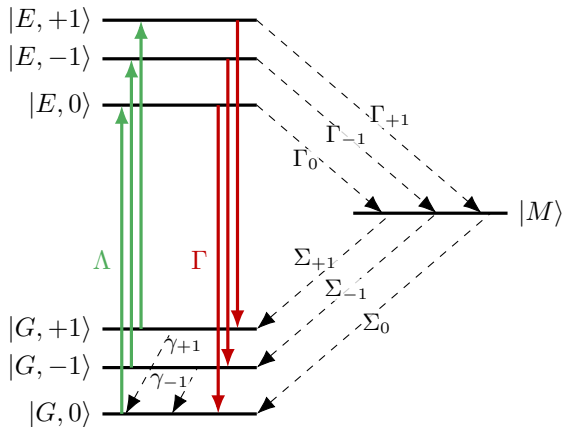
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Experimental Setup: NV Center in Diamond



— Aiello *et al.*, arXiv:2605.00152 (2026)

NV Center Levels



- Optical Manifold:

$|G\rangle, |E\rangle, |M\rangle$

- Electronic Spin of NV center:

$|+1\rangle, |0\rangle, |-1\rangle$

In addition:

- ^{13}C nuclear spin

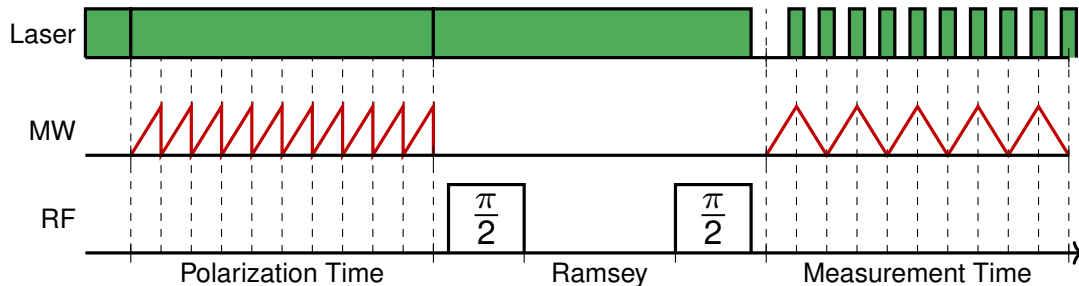
- single carbon:

$|\uparrow\rangle, |\downarrow\rangle$

- N carbons:

$|\uparrow\uparrow\rangle, |\uparrow\downarrow\rangle, |\downarrow\uparrow\rangle, |\downarrow\downarrow\rangle$, etc.

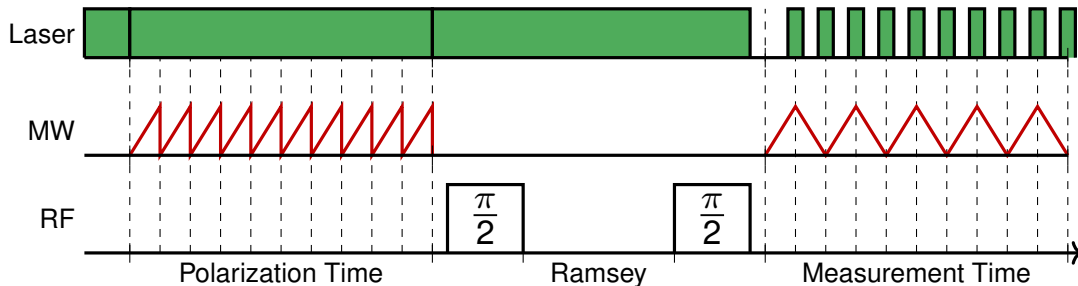
Pulse Scheme



Initial state:

$$\rho_0 = \frac{1}{2} (|0 \uparrow\rangle\langle 0 \uparrow| + |0 \downarrow\rangle\langle 0 \downarrow|)$$

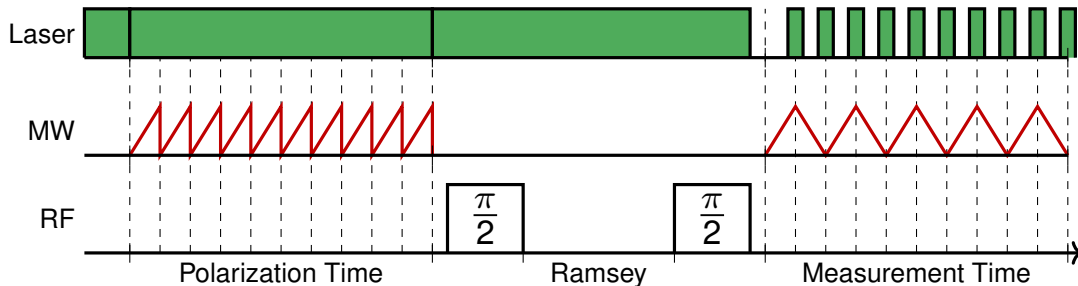
Pulse Scheme



Polarization:

$$\frac{1}{2} (|0 \uparrow\rangle\langle 0 \uparrow| + |0 \downarrow\rangle\langle 0 \downarrow|) \rightarrow \frac{1}{2} (|0 \uparrow\rangle\langle 0 \uparrow| + |\pm 1 \uparrow\rangle\langle \pm 1 \uparrow|)$$

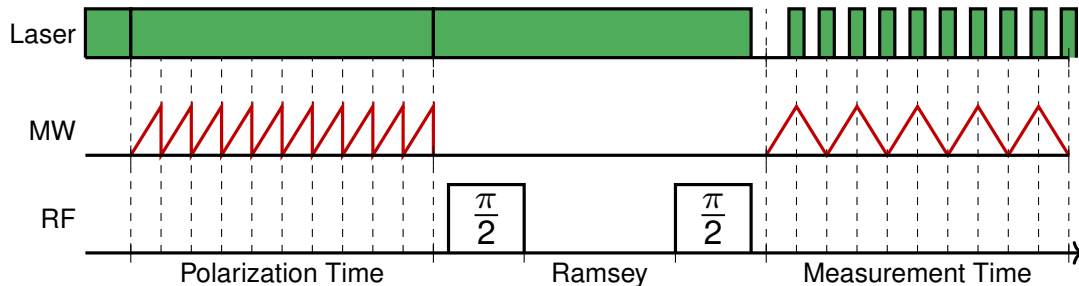
Pulse Scheme



Polarization:

$$\text{re-pumping} \rightarrow |0 \uparrow\rangle\langle 0 \uparrow| \rightarrow |\psi\rangle = |0 \uparrow\rangle$$

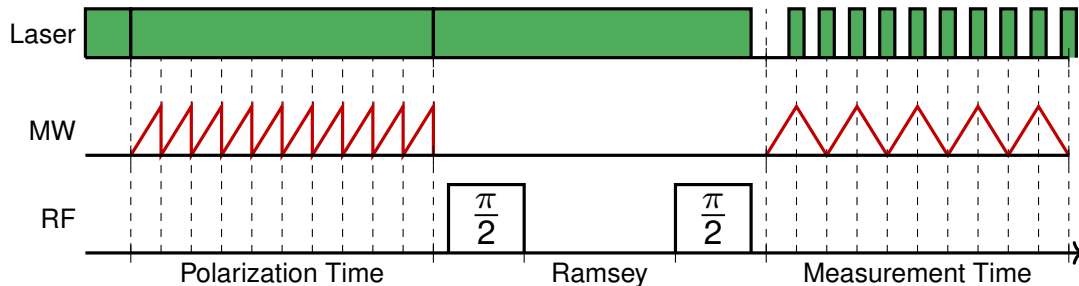
Pulse Scheme



Ramsey:

$$|0 \uparrow\rangle \rightarrow \frac{1}{\sqrt{2}} (|0 \uparrow\rangle + |0 \downarrow\rangle) \rightarrow \frac{1}{\sqrt{2}} \left(e^{-i\frac{\phi}{2}} |0 \uparrow\rangle + e^{i\frac{\phi}{2}} |0 \downarrow\rangle \right)$$

Pulse Scheme

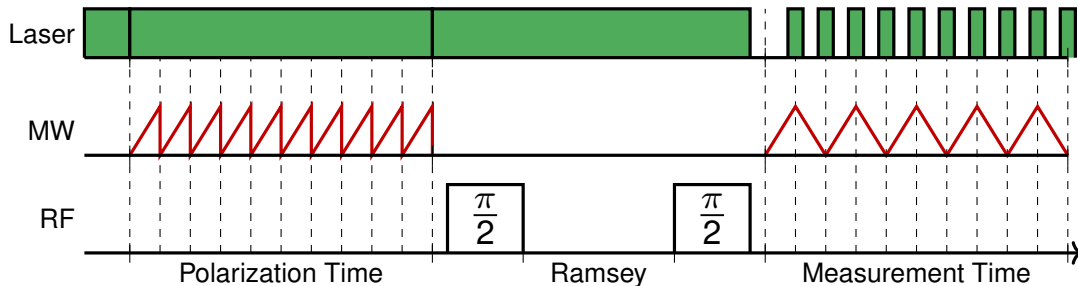


Ramsey:

$$\text{recombination} \rightarrow \sin\left(\frac{\phi}{2}\right) |0 \uparrow\rangle - \cos\left(\frac{\phi}{2}\right) |0 \downarrow\rangle$$

superposition depending on accumulated phase difference

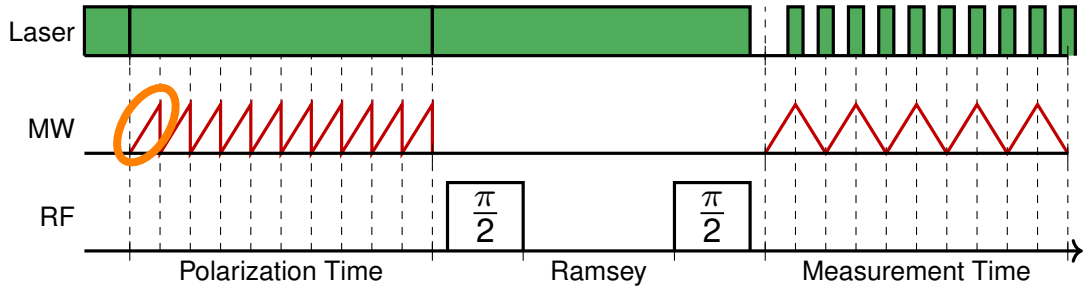
Pulse Scheme



Readout:

$$\sin\left(\frac{\phi}{2}\right) |0 \uparrow\rangle - \cos\left(\frac{\phi}{2}\right) |0 \downarrow\rangle \rightarrow \sin\left(\frac{\phi}{2}\right) |+1 \uparrow\rangle - \cos\left(\frac{\phi}{2}\right) |0 \uparrow\rangle$$

Polarization



Polarization:

$$\frac{1}{2} (|0 \uparrow\rangle\langle 0 \uparrow| + |0 \downarrow\rangle\langle 0 \downarrow|) \rightarrow \frac{1}{2} (|0 \uparrow\rangle\langle 0 \uparrow| + |\pm 1 \uparrow\rangle\langle \pm 1 \uparrow|)$$

Hamiltonian

$$\hat{H}_{\text{lab}} = \overbrace{D \hat{S}_z^2}^{\text{zero-field splitting}} + \underbrace{\gamma_e \mathbf{B} \cdot \hat{\mathbf{S}}}_{\text{electronic Zeeman}} + \sum_{n=1}^N \left(\underbrace{\hat{\mathbf{S}} \cdot A^{(n)} \hat{\mathbf{I}}^{(n)}}_{\text{hyperfine}} - \underbrace{\gamma_c \mathbf{B} \cdot \hat{\mathbf{I}}^{(n)}}_{\text{nuclear Zeeman}} \right) + \underbrace{\mu \Omega(t) \hat{S}_x}_{\text{MW drive}}$$

Hamiltonian

$$\hat{H}_{\text{lab}} = D \hat{S}_z^2 + \gamma_e \mathbf{B} \cdot \hat{\mathbf{S}} + \sum_{n=1}^N \left(\hat{\mathbf{S}} \cdot A^{(n)} \hat{\mathbf{I}}^{(n)} - \gamma_c \mathbf{B} \cdot \hat{\mathbf{I}}^{(n)} \right) + \mu \Omega(t) \hat{S}_x$$

In rotating frame, with $\Omega(t) = \sqrt{2} \Omega_{\pm}(t) \cos(\omega_{\pm} t + \phi_{\pm}(t))$:

$$\hat{H}_{\text{RWA}} = \underbrace{(\hat{\delta} - \hat{\omega}(t))}_{\text{detuning \& chirp}} + \sum_{n=1}^N \left(\underbrace{\sum_{j=x,y,z} A_{z,j}^{(n)} \hat{S}_z \otimes \hat{I}_j^{(n)}}_{\text{hyperfine}} - \underbrace{\gamma_c B \mathbf{1}_S \otimes \hat{I}_z^{(n)}}_{\text{nuclear Zeeman}} \right) + \underbrace{\hat{\Omega}(t)}_{\text{MW envelope}}$$

$$\hat{\delta} = \text{diag}[\delta_+, 0, \delta_-], \quad \hat{\omega}(t) = \text{diag}[\dot{\phi}_+(t), 0, \dot{\phi}_-(t)], \quad \hat{\Omega}(t) = \frac{\mu}{2} \begin{pmatrix} 0 & \Omega_+ & 0 \\ \Omega_+ & 0 & \Omega_- \\ 0 & \Omega_- & 0 \end{pmatrix}$$

Avoided Crossings

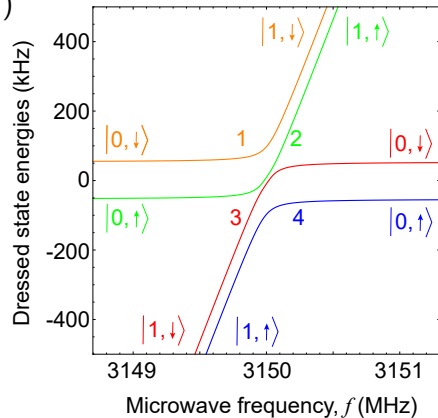
$$\Omega_+^{(\text{lab})}(t) = \Omega_+ \cos[\omega_+ t + \phi_+(t)], \quad \dot{\phi}_+(t) = \alpha(t - t_c)$$

RWA Hamiltonian in $|0\rangle - |+1\rangle$ subspace:

$$\hat{H}_+ = \begin{pmatrix} E_{1\uparrow} & \frac{1}{2}A_{zx} & \frac{1}{2}\Omega_+ & 0 \\ \frac{1}{2}A_{zx} & E_{1\downarrow} & 0 & \frac{1}{2}\Omega_+ \\ \frac{1}{2}\Omega_+ & 0 & E_{0\uparrow} & 0 \\ 0 & \frac{1}{2}\Omega_+ & 0 & E_{0\downarrow} \end{pmatrix}$$

$$E_{1\uparrow\downarrow} = \pm \frac{A_{zz}}{2} \mp \frac{B\gamma_c}{2} - \underbrace{\alpha(t - t_c)}_{\text{linear chirp}}, \quad E_{0\uparrow\downarrow} = \mp \frac{B\gamma_c}{2}$$

$$P_{\text{hop}} \sim \exp\left(-\frac{\pi}{2} \frac{\Delta^2}{\alpha}\right)$$



— Aiello *et al.*, arXiv:2605.00152 (2026)

Avoided Crossings

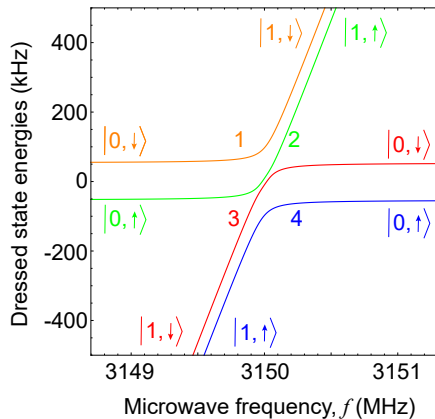
For positive chirp:

$$\begin{array}{ll}
 |0 \downarrow\rangle & \rightarrow |0 \uparrow\rangle \\
 |0 \uparrow\rangle & \rightarrow |+1 \uparrow\rangle \\
 |+1 \downarrow\rangle & \rightarrow |0 \downarrow\rangle \\
 |+1 \uparrow\rangle & \rightarrow |+1 \downarrow\rangle
 \end{array}$$

For negative chirp:

$$\begin{array}{ll}
 |0 \downarrow\rangle & \rightarrow |+1 \downarrow\rangle \\
 |0 \uparrow\rangle & \rightarrow |0 \downarrow\rangle \\
 |+1 \downarrow\rangle & \rightarrow |+1 \uparrow\rangle \\
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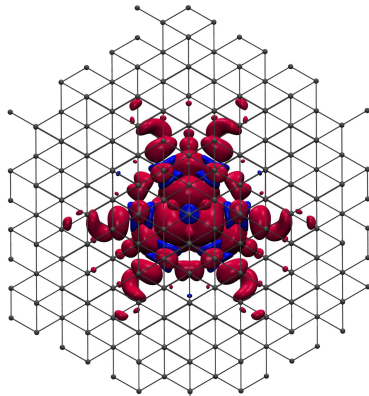
Interaction with Multiple Carbons

$$\hat{H} = D \hat{S}_z^2 + \gamma_e \mathbf{B} \cdot \hat{\mathbf{S}} + \hat{\mathbf{S}} \cdot \mathbf{A} \hat{\mathbf{I}} - \gamma_c \mathbf{B} \cdot \hat{\mathbf{I}}$$

- 3×3 hyperfine tensor $\mathbf{A}$ couples electronic spins $\hat{S}_x, \hat{S}_y, \hat{S}_z$ and nuclear spins $\hat{I}_x, \hat{I}_y, \hat{I}_z$
- Long range: dipole-dipole interaction (kHz)

$$A_{ij} = \frac{\mu_0}{4\pi} \frac{\gamma_e \gamma_n \hbar^2}{r^3} (\delta_{ij} - 3 \hat{r}_i \hat{r}_j)$$

- Short range: Fermi contact interaction (MHz)
- Natural abundance of ^{13}C : 1.1%



— Takács & Ivády, Commun. Phys. **7**, 178 (2024)

Polarization of Nuclear Spin – Statistics

Propagate

$$\hat{\rho}_0 = \frac{1}{2} (|0 \uparrow\rangle\langle 0 \uparrow| + |0 \downarrow\rangle\langle 0 \downarrow|)$$

over single chirp ramp:

$$\Delta f = 9 \text{ MHz}$$

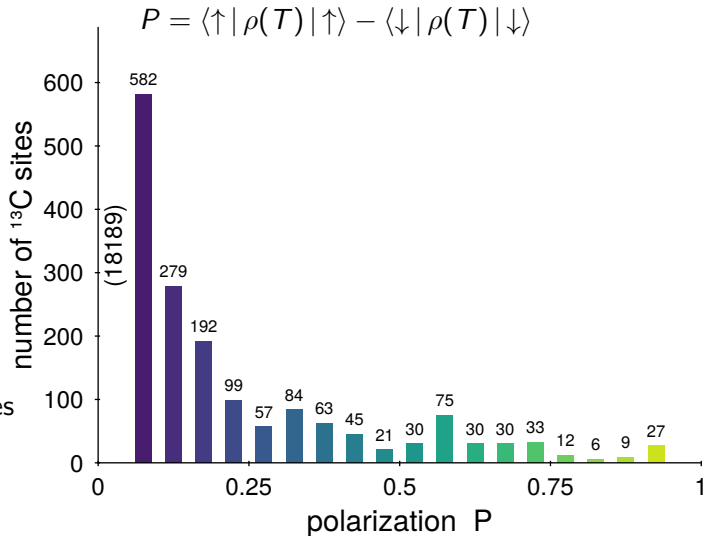
$$T = 833 \text{ } \mu\text{s}$$

$$\alpha = 10.8 \text{ MHz/ms}$$

$$B = 120 \text{ Gauss}$$

$$\Omega_0 = 100 \text{ kHz}$$

For all 20000 possible carbon sites



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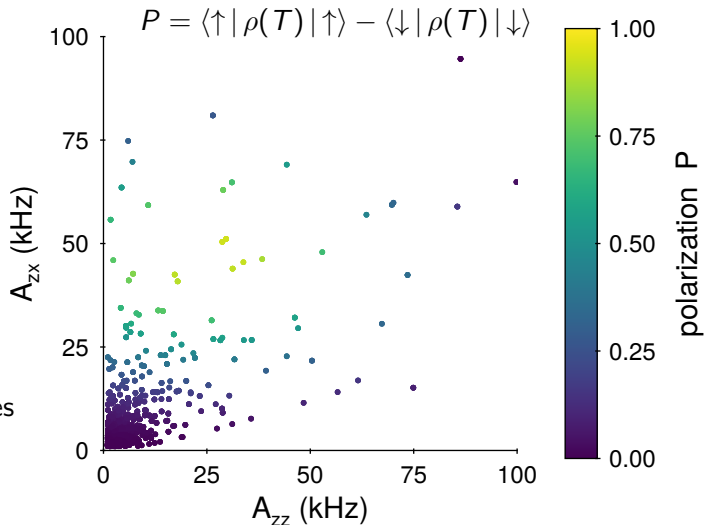
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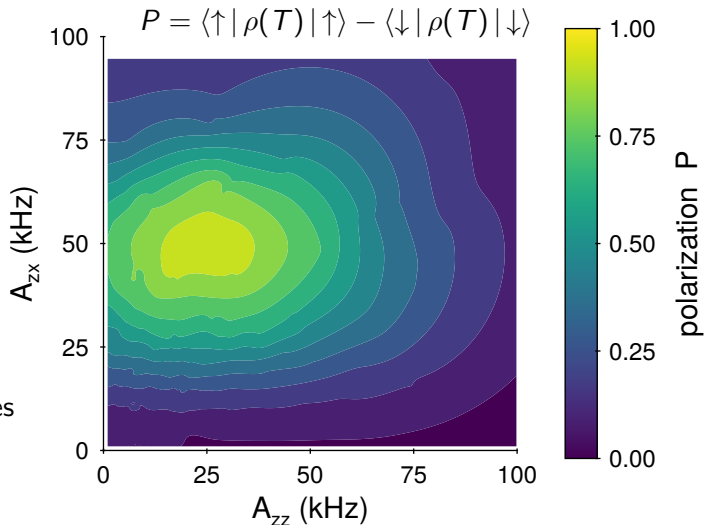
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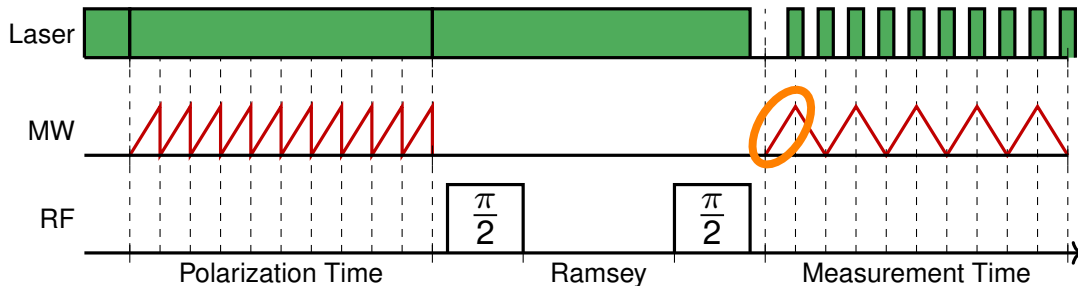
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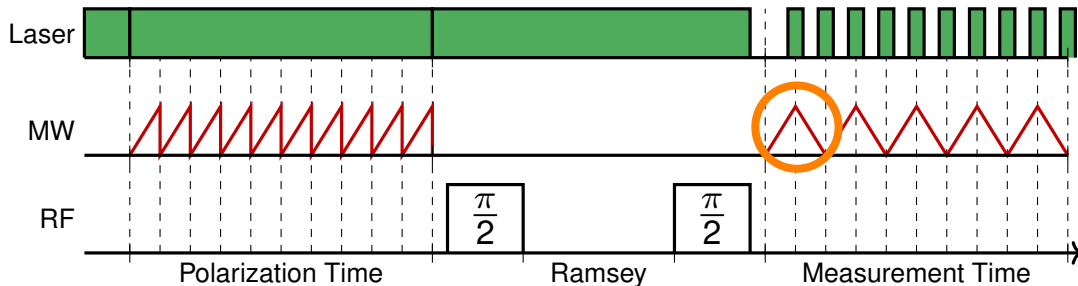
Readout



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Readout



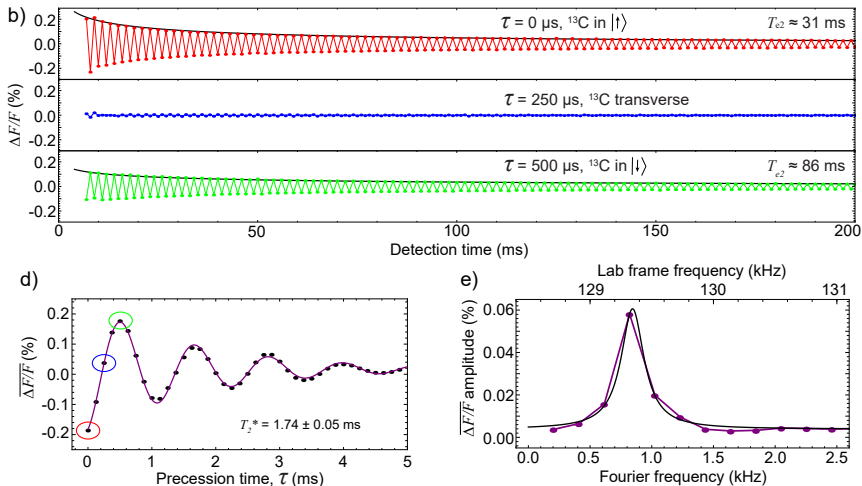
For positive chirp:

$$\begin{aligned}
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 |+1 \downarrow\rangle &\rightarrow |0 \downarrow\rangle \\
 |+1 \uparrow\rangle &\rightarrow |+1 \downarrow\rangle
 \end{aligned}$$

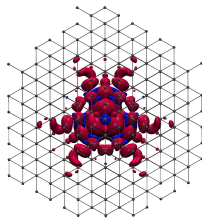
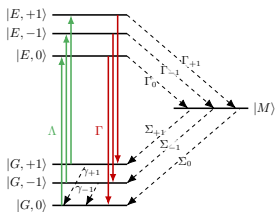
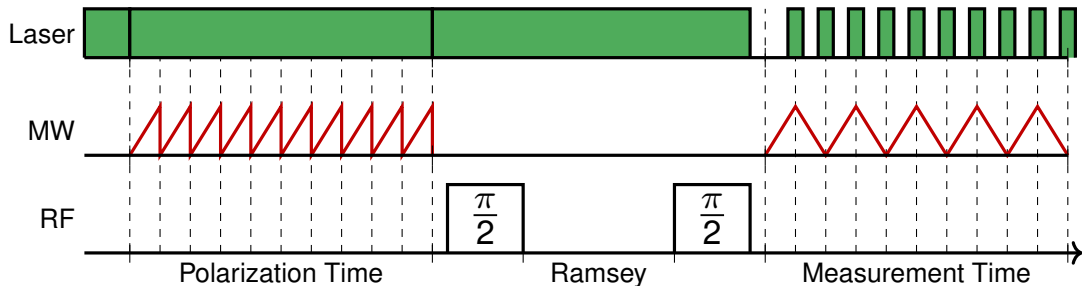
For negative chirp:

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 |0 \uparrow\rangle &\rightarrow |0 \downarrow\rangle \\
 |+1 \downarrow\rangle &\rightarrow |+1 \uparrow\rangle \\
 |+1 \uparrow\rangle &\rightarrow |0 \uparrow\rangle
 \end{aligned}$$

Readout

— Aiello *et al.*, arXiv:2605.00152 (2026)

Control Perspectives



- Effective multi-carbon open model
- Diffusion model
- Non-linear chirp
- Optical pulse protocol

Numerical Framework



JuliaQuantumControl

Julia Framework for Quantum Optimal Control

42 followers <https://juliaquantumcontrol.github.io...>

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A Julia Framework for Quantum Optimal Control.

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The [JuliaQuantumControl](#) organization collects packages implementing a comprehensive collection of methods of open-loop quantum optimal control.

[Quantum optimal control theory](#) attempts to steer a quantum system in some desired way by finding optimal control parameters or control fields inside the system Hamiltonian or Liouvillian. Typical control tasks are the preparation of a specific quantum state or the realization of a logical gate in a quantum computer. Thus, quantum control theory is a critical part of realizing quantum technologies, at the lowest level. Numerical methods of *open-loop* quantum control (methods that do not involve measurement feedback from a physical quantum device) such as [Krotov's method](#) and [GRAPE](#) address the control problem by [simulating the dynamics of the system](#) and then iteratively improving the value of a functional that encodes the desired outcome.

The packages in [JuliaQuantumControl](#) that implement specific individual methods should be used via the high-level package

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.github/workflows	Add workflow to render notes	5 months ago
docs	Clarify frame transformations	3 months ago
examples	Clarify frame transformations	3 months ago
notes	Clarify frame transformations	3 months ago
src	Add utility functions	3 months ago
test	Clarify frame transformations	3 months ago
.JuliaFormatter.toml	Customize package setup	last year

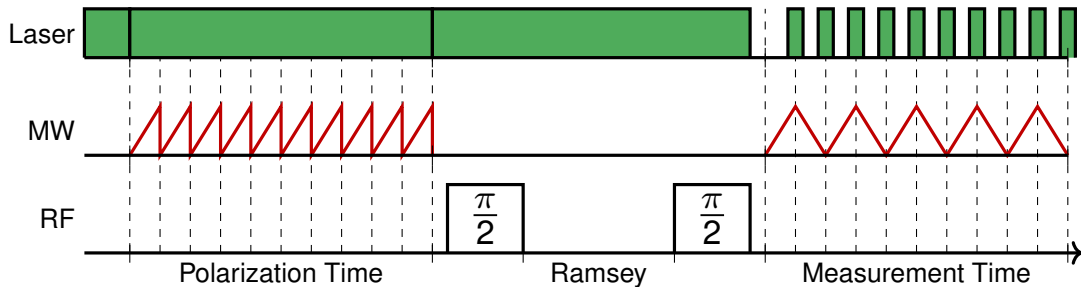
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Julia model for C13 interacting with and NV center in diamond

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