

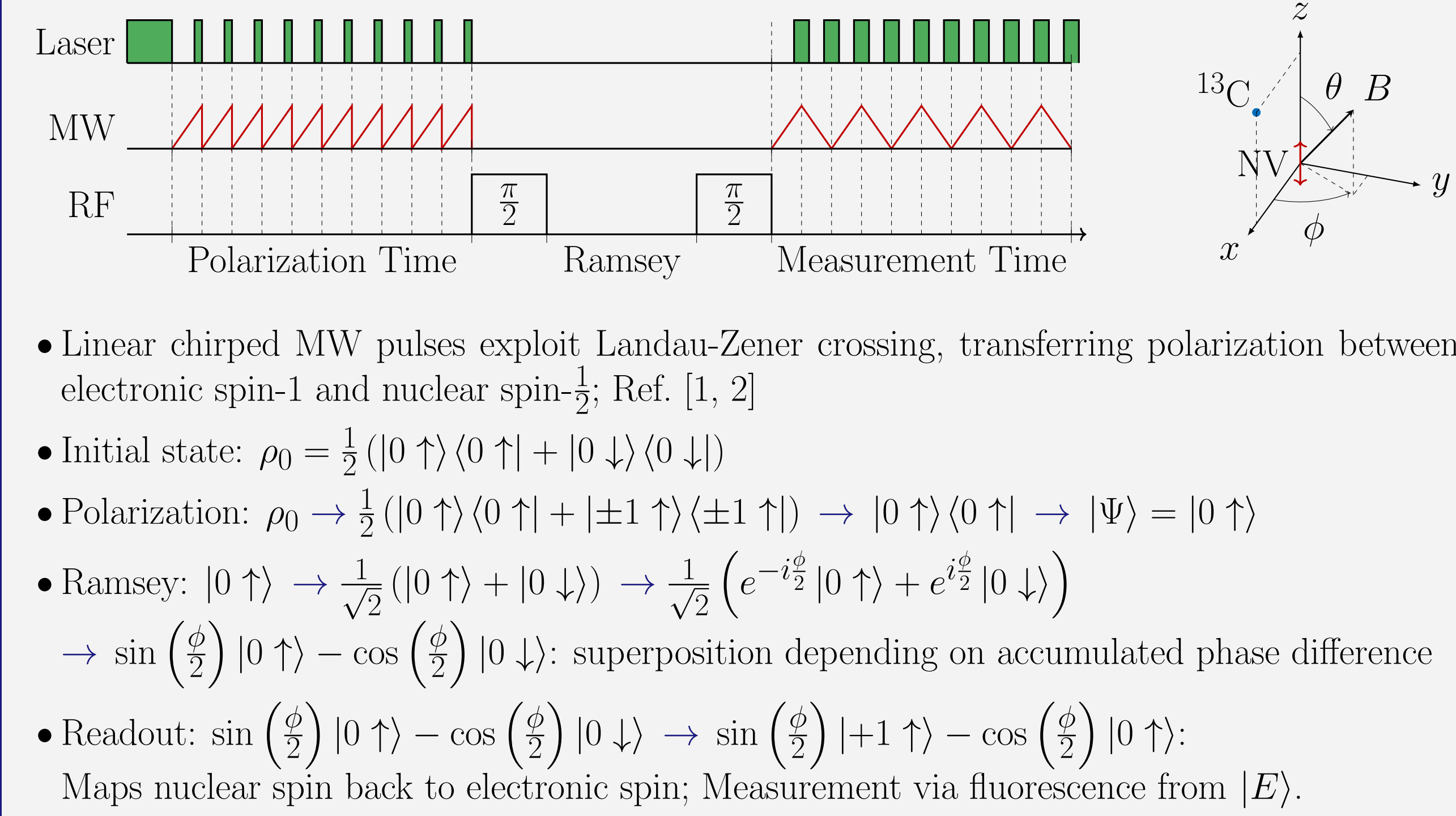
Microwave Pulse Trains for Quantum Metrology with ^{13}C in Diamond



Michael H. Goerz¹, J. Chathanathil¹, M. D. Aiello², J. Smits², S. C. Carrasco¹, A. Jarmola^{1,3}, D. Budker^{3,4,5}, V. Acosta², and V. S. Malinovsky¹

¹DEVCOM Army Research Laboratory, Adelphi, MD, USA ²University of New Mexico, Albuquerque, NM, USA ³University of California at Berkeley, CA, USA ⁴Johannes Gutenberg-Universität Mainz, Germany ⁵Helmholtz-Institut Mainz, Germany

Metrology Pulse Scheme for ^{13}C Nuclear Spin



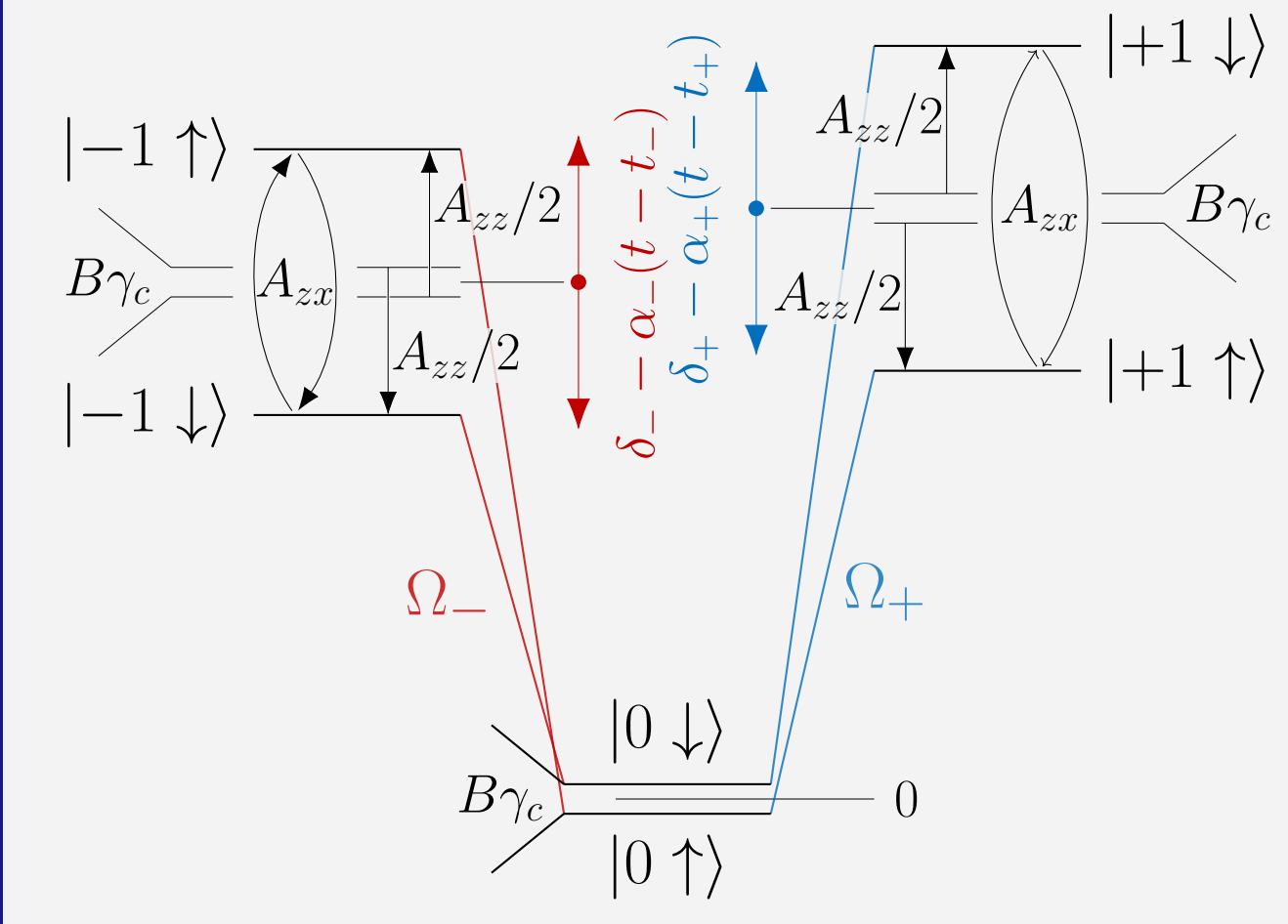
Interaction with MW Field

Linear Chirp Hamiltonian

In RWA w.r.t. drive frequencies $\omega_{\pm}$, detuned by $\delta_{\pm}$ from $|0\rangle \leftrightarrow |\pm 1\rangle$,

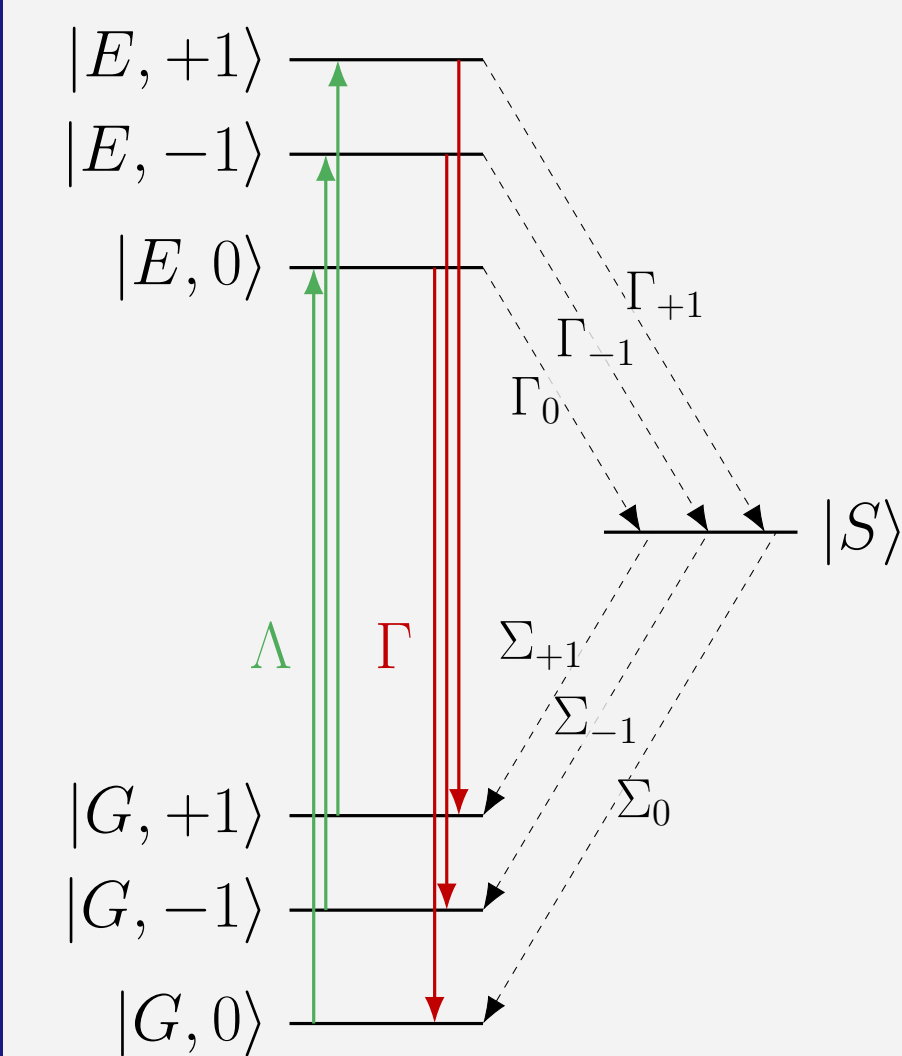
$$\Omega_{\pm}^{(\text{lab})}(t) = \Omega_{\pm} \cos(\omega_{\pm} t + \phi_{\pm}(t))$$

with $\phi_{\pm}(t) = \frac{\alpha_{\pm}}{2}(t - t_{\pm})^2$



$$\hat{H} = \begin{pmatrix} E_{-1\uparrow} & \frac{A_{zx}}{2} & \frac{\Omega_-}{2} & 0 & 0 & 0 \\ \frac{A_{zx}}{2} & E_{-1\downarrow} & 0 & \frac{\Omega_-}{2} & 0 & 0 \\ \frac{\Omega_-}{2} & 0 & E_{0\uparrow} & 0 & \frac{\Omega_+}{2} & 0 \\ 0 & \frac{\Omega_-}{2} & 0 & E_{0\downarrow} & 0 & \frac{\Omega_+}{2} \\ 0 & 0 & \frac{\Omega_+}{2} & 0 & E_{+1\uparrow} & -\frac{A_{zx}}{2} \\ 0 & 0 & 0 & \frac{\Omega_+}{2} & -\frac{A_{zx}}{2} & E_{+1\downarrow} \end{pmatrix}$$
$$E_{-1\uparrow\downarrow} = \pm \frac{A_{zz}}{2} \mp \frac{B\gamma_c}{2} + \underbrace{\delta_- - \alpha_-(t - t_-)}_{\equiv \delta_-(t)}$$
$$E_{0\uparrow\downarrow} = \mp \frac{B\gamma_c}{2}$$
$$E_{+1\uparrow\downarrow} = \mp \frac{A_{zz}}{2} \mp \frac{B\gamma_c}{2} + \underbrace{\delta_+ - \alpha_+(t - t_+)}_{\equiv \delta_+(t)}$$
$$\delta_{\pm} = (E_{\pm 1} - E_0) - \omega_{\pm} = (\Delta \pm B\gamma_c) - \omega_{\pm}$$

Interaction with Laser



Dissipative Model

with excited manifold E and metastable S

- incoherent (!) drive to $|E\rangle$ with rate Λ
- spontaneous decay to $|G\rangle$ with rate $\Gamma = 1/12\text{ns}$
- fluorescence proportional to $\Gamma \cdot \langle E \rangle$
- decay to metastable state with $\Gamma_0 \approx 0$, $\Gamma_{\pm 1} = 1/25\text{ns}$
- decay from metastable state with $\Sigma_{0,\pm 1} = 1/219\text{ns}$

Role of Laser Field

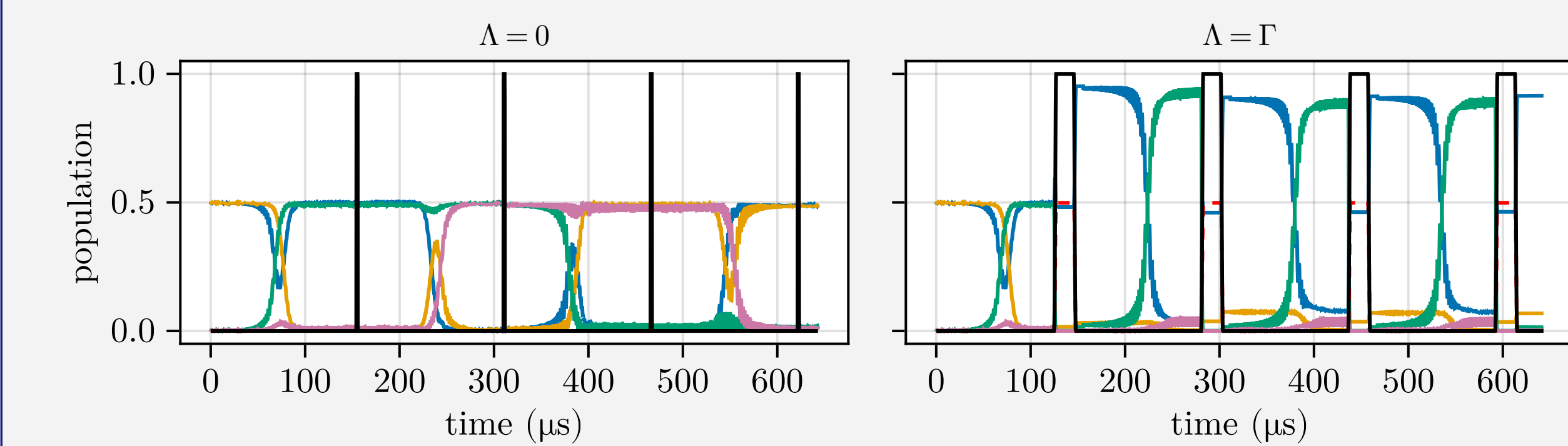
- Reset electronic spin after chirped pulse
- Read out electronic spin after Ramsey scheme (weak measurement)

Pulse Trains for Polarization

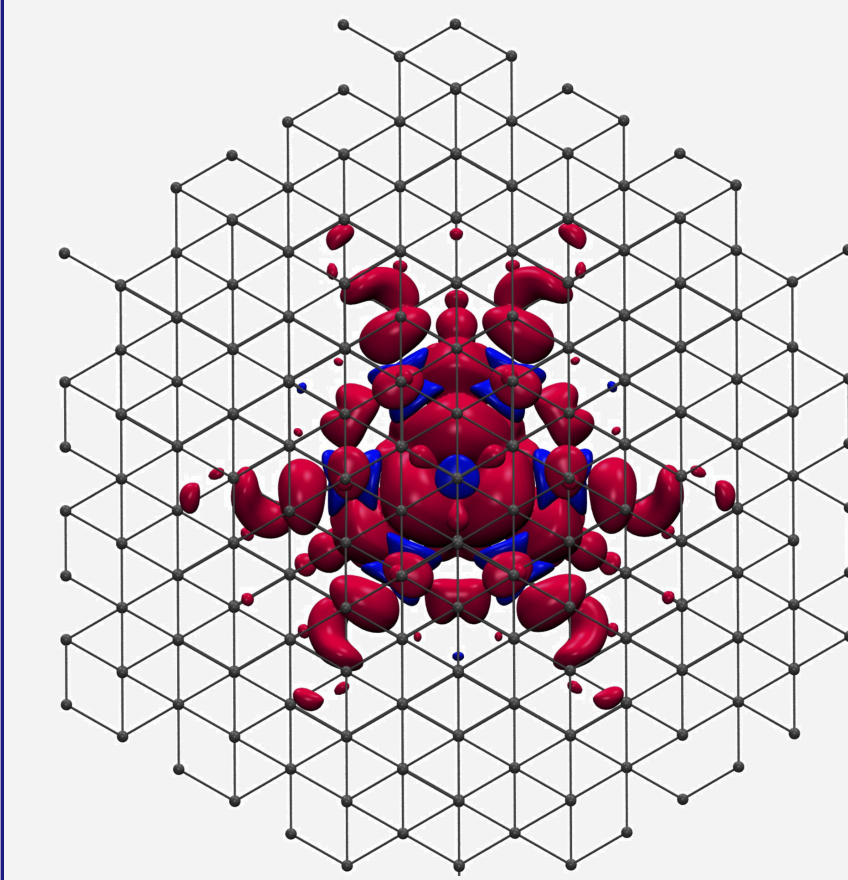
Chirped MW pulse polarizes nuclear spin but depolarizes electronic spin.
 $\Rightarrow$ need for optical re-pumping

Parameters

- $B = 120\text{ G}$
- $A_{zz} = 1\text{ MHz}$, $A_{zx} = 300\text{ kHz}$
- $\Omega_{\pm} = 347\text{ kHz}$, $t_{\pm} = 77.9\text{ }\mu\text{s}$



Hyperfine Interaction



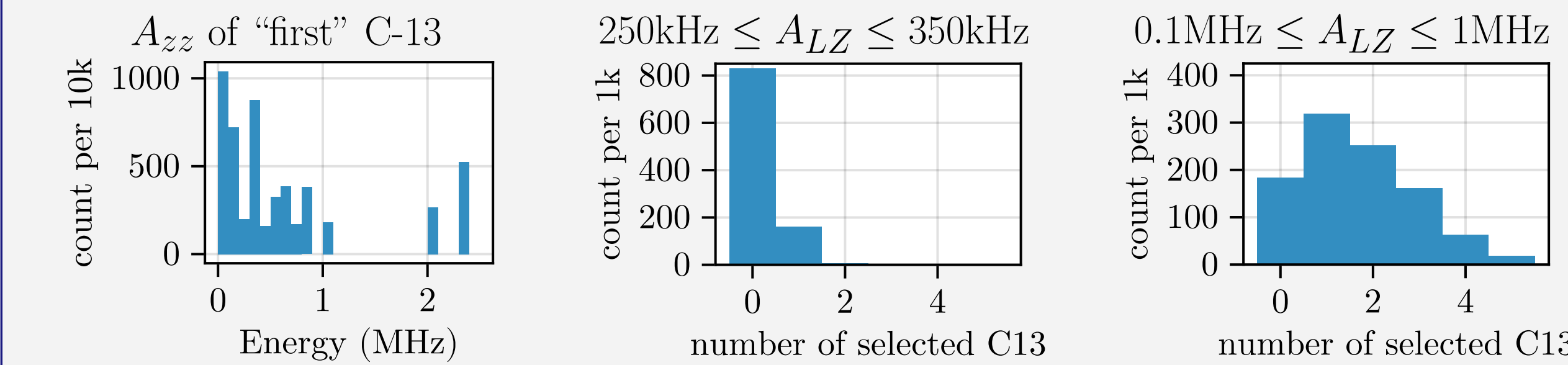
Adapted from [3].

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

- 3×3 hyperfine tensor $\mathbf{A}$ for coupling of 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)

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

- Short range: Fermi interaction (MHz)
- Natural abundance of C-13: 1.1%
- Generalized LZ interaction: $A_{LZ} = \sqrt{A_{zx}^2 + A_{zy}^2}$



Diffusion

Nuclear spins diffuse to neighboring ^{13}C atoms $\rightarrow$ pulse trains build up bulk polarization

- Include diffusion in dissipative model with effective Lindblad operator [4]; $\gamma_{\text{diff}} \sim \frac{D}{a^2}$
- Track P_{bulk} as classical variable

References

- [1] Ajoy *et al.*, Sci. Adv. 4, eaar5492 (2018)
- [2] Zangara *et al.* Proc. Natl. Acad. Sci. U.S.A 116, 2512 (2019)
- [3] Takács and Ivády, Commun. Phys. 7, 178 (2024)
- [4] Chessari *et al.*, Phys. Rev. B 107, 224429 (2023)

Pulse Trains for Readout

Alternating MW chirps with positive and negative chirp rate to map between nuclear and electronic spin $\Rightarrow$ weak fluorescence measurement

Application:

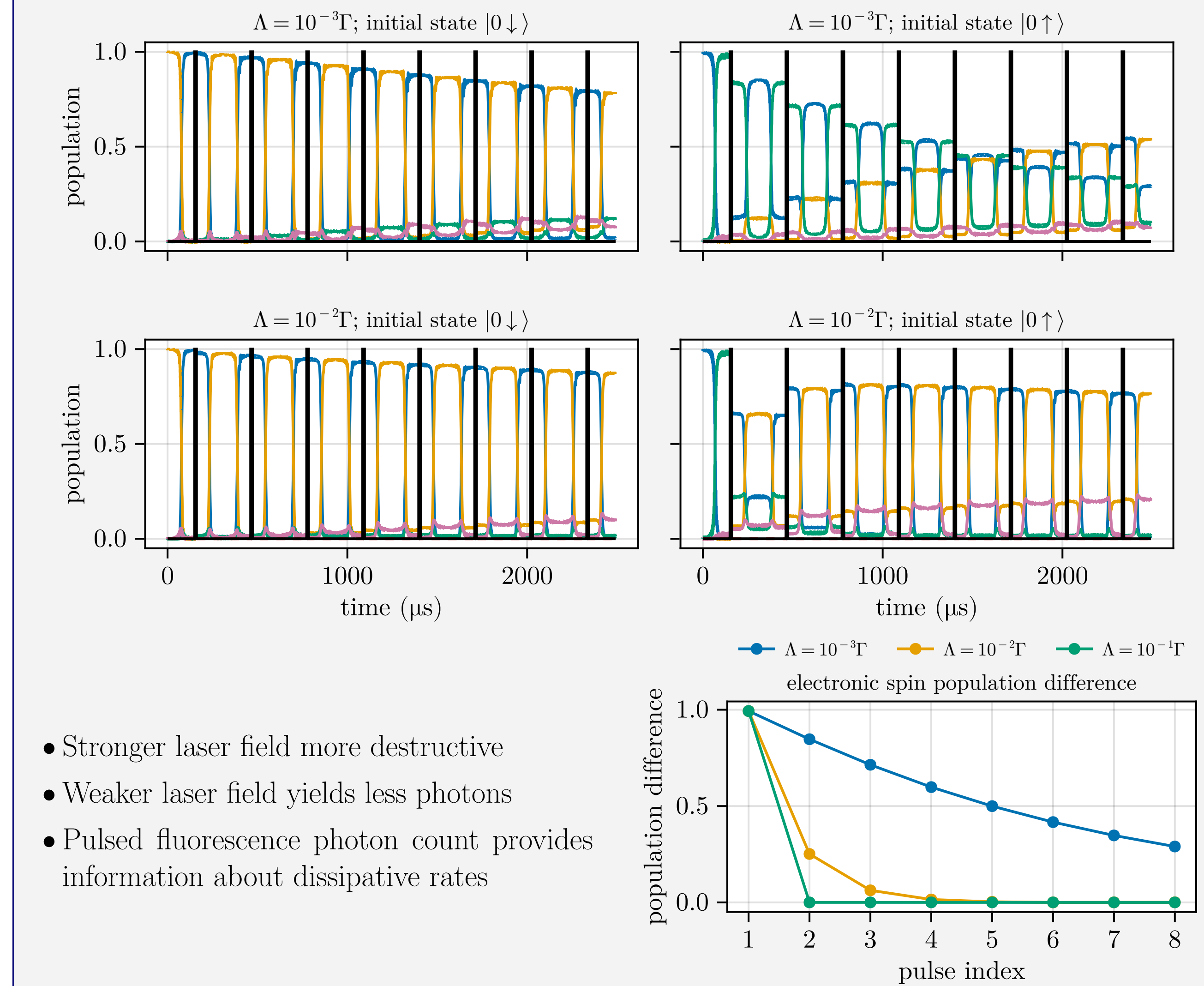
For positive chirp:

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

For negative chirp:

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

$$\begin{aligned} |\Psi\rangle &= \sin\left(\frac{\phi}{2}\right)|0 \uparrow\rangle - \cos\left(\frac{\phi}{2}\right)|0 \downarrow\rangle \\ (\pm) &\approx \sin\left(\frac{\phi}{2}\right)|+1 \uparrow\rangle - \cos\left(\frac{\phi}{2}\right)|0 \uparrow\rangle \\ (\mp) &\approx \sin\left(\frac{\phi}{2}\right)|0 \uparrow\rangle - \cos\left(\frac{\phi}{2}\right)|0 \downarrow\rangle \end{aligned}$$



- Stronger laser field more destructive
- Weaker laser field yields less photons
- Pulsed fluorescence photon count provides information about dissipative rates

Outlook: Optimal Control and Machine Learning

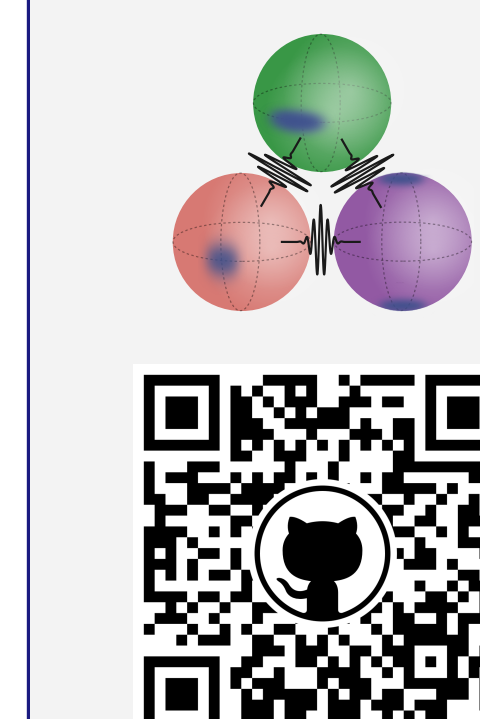
OCT

- Non-linear, optimized chirp ramps
- Polarize via $|+1\rangle$ and $|-1\rangle$ simultaneously
- Ensemble opt. for hyperfine couplings
- Robustness w.r.t. to field imperfections

Reinforcement Learning

- Select pre-optimized shapes for pulse trains
- Laser strength and switch-on/-off timing
- Learn system parameters from pulsed fluorescence response

QuantumControl.jl Framework



JuliaQuantumControl Organization
— <https://github.com/JuliaQuantumControl>

julia

- QuantumPropagators.jl**: Simulate time dynamics
- GRAPE.jl**: Gradient-ascent pulse engineering with Semi-AD
- Krotov.jl**: Krotov's method with Semi-AD
- QuantumControl.jl**: High-level framework for dynamics and control