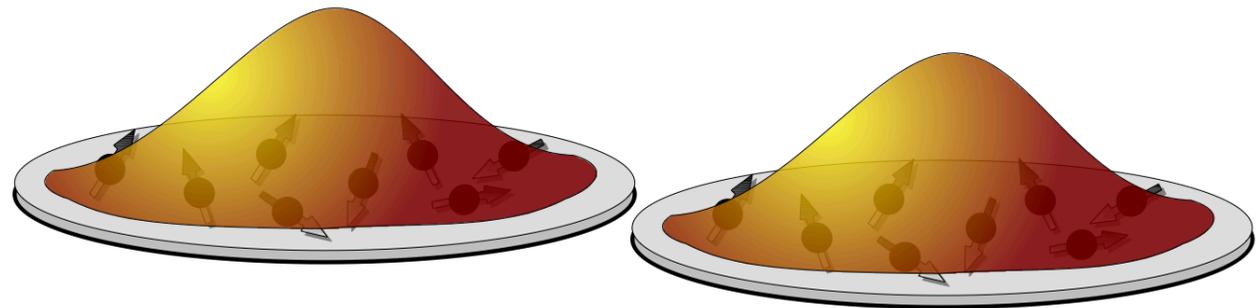


Dynamical nuclear polarization oscillations in quantum dots

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Niels Bohr Institute

3 October 2013



In collaboration with:

Izhar Neder (Tel Aviv)

Leonid Levitov (MIT)

Bert Halperin (Harvard)

For more information, see:

MR, I. Neder, L. S. Levitov, and B. I. Halperin, Phys. Rev. B **82**, 041311(R) (2010).

MR and L. S. Levitov, Phys. Rev. Lett. **110**, 086601 (2013).

I. Neder, MR, and B. I. Halperin, arXiv:1309.3027 (2013).

The Plan

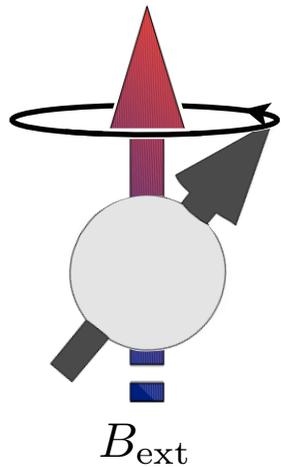
I. Motivation and background

II. Coherent interplay of hyperfine and spin-orbit coupling

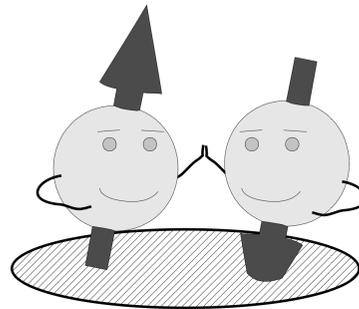
III. Nonlinear dynamics and self-oscillations of DNP

QDs offer a controlled platform for studying spin dynamics on a wide range of timescales

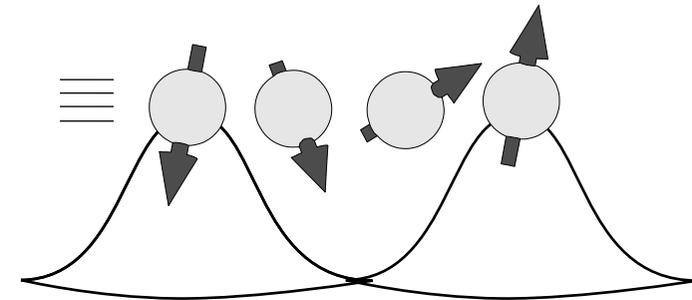
Larmor precession



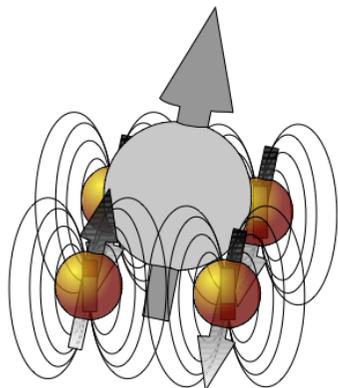
Exchange interaction



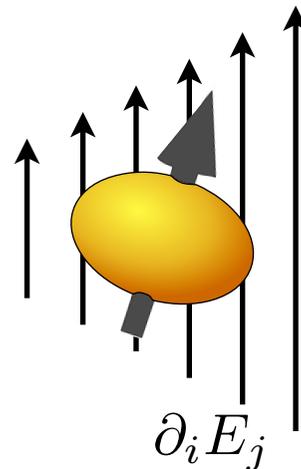
Spin-orbit coupling



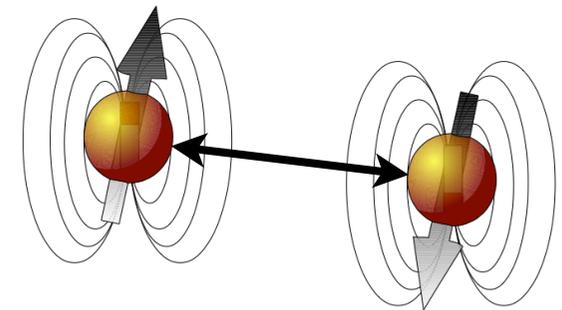
Hyperfine interaction



Quadrupolar coupling

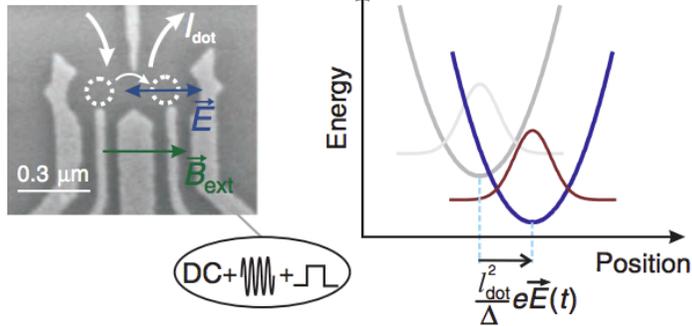


Dipole-dipole coupling;
nuclear spin diffusion



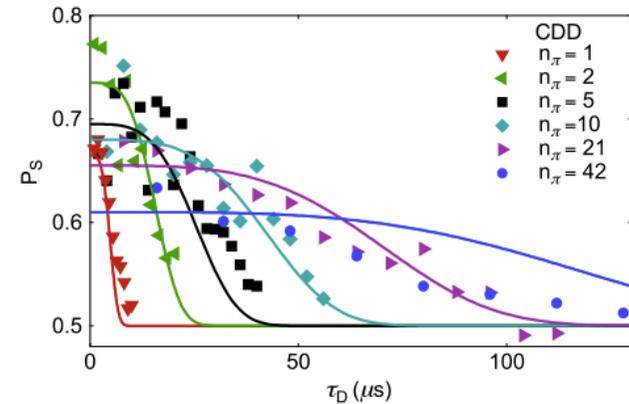
Phenomena relevant for spin-based information processing

Electric dipole spin resonance (EDSR)



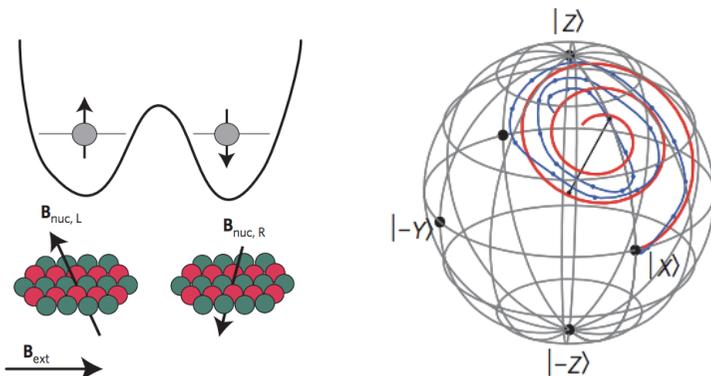
K. C. Nowack *et al.*, Science **318**, 1430 (2007)
 M. Pioro-Ladriere *et al.*, Nature Physics **4**, 776 (2008).

Electron spin decoherence



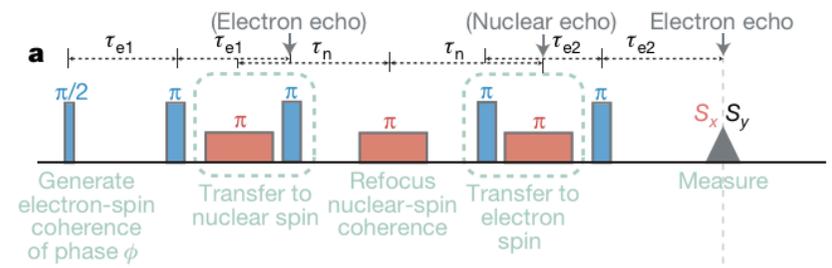
J. Medford *et al.*, PRL **108**, 086802 (2012)

Hyperfine fields for universal qubit control



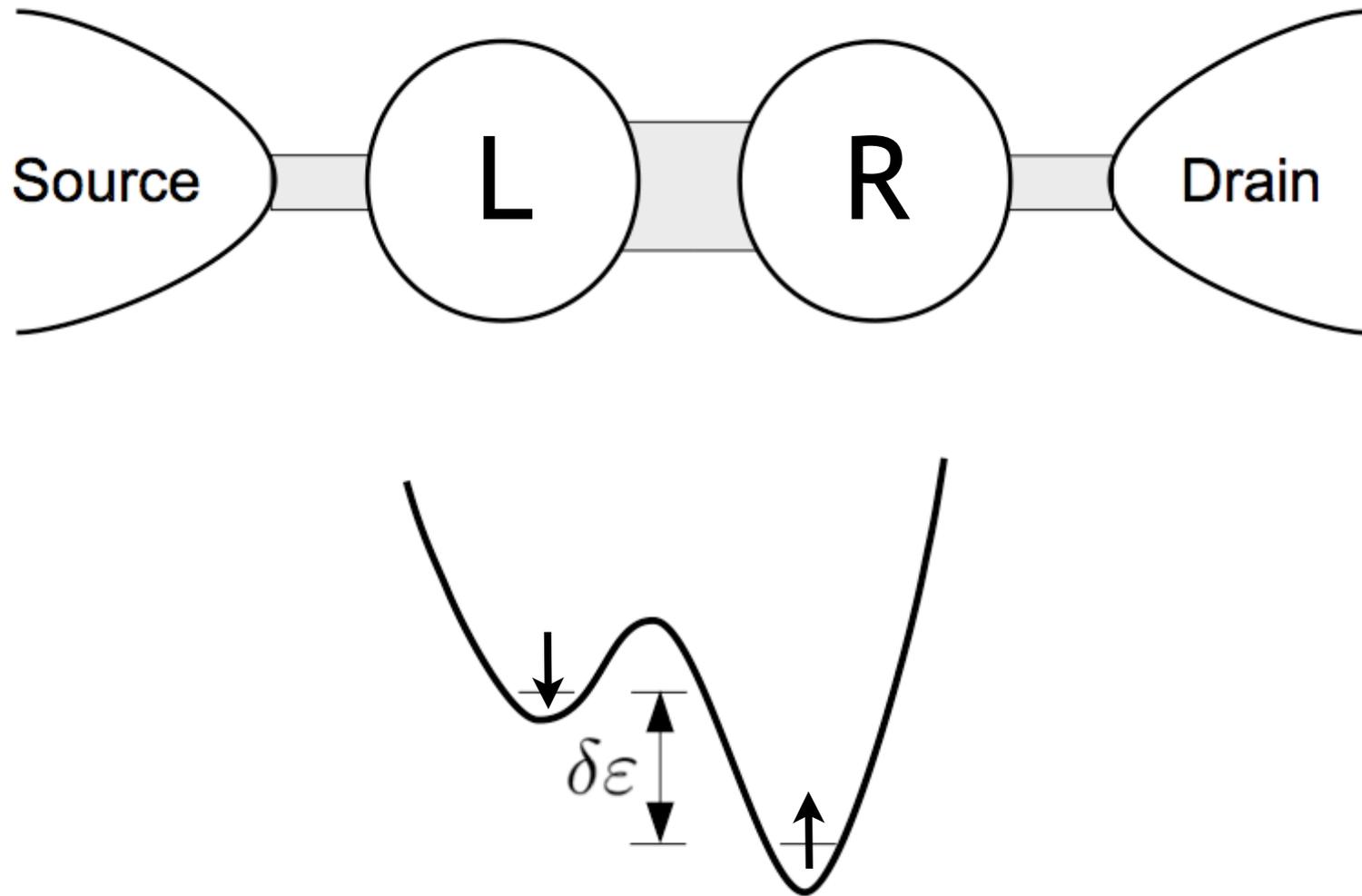
S. Foletti *et al.*, Nature Physics **5**, 903 (2009)

Long-lived quantum memory

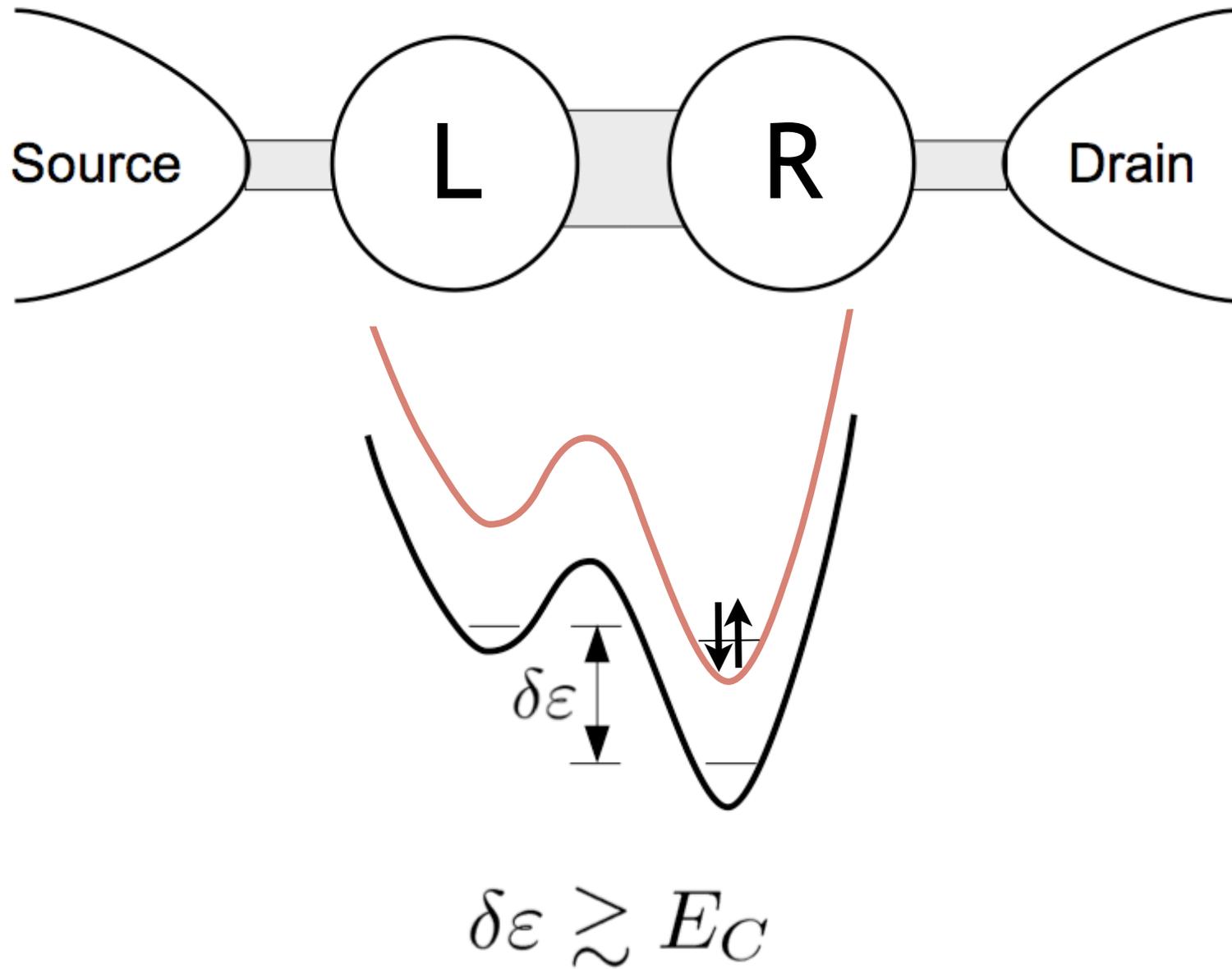


J. J. L. Morton *et al.*, Nature **455**, 1085 (2008)

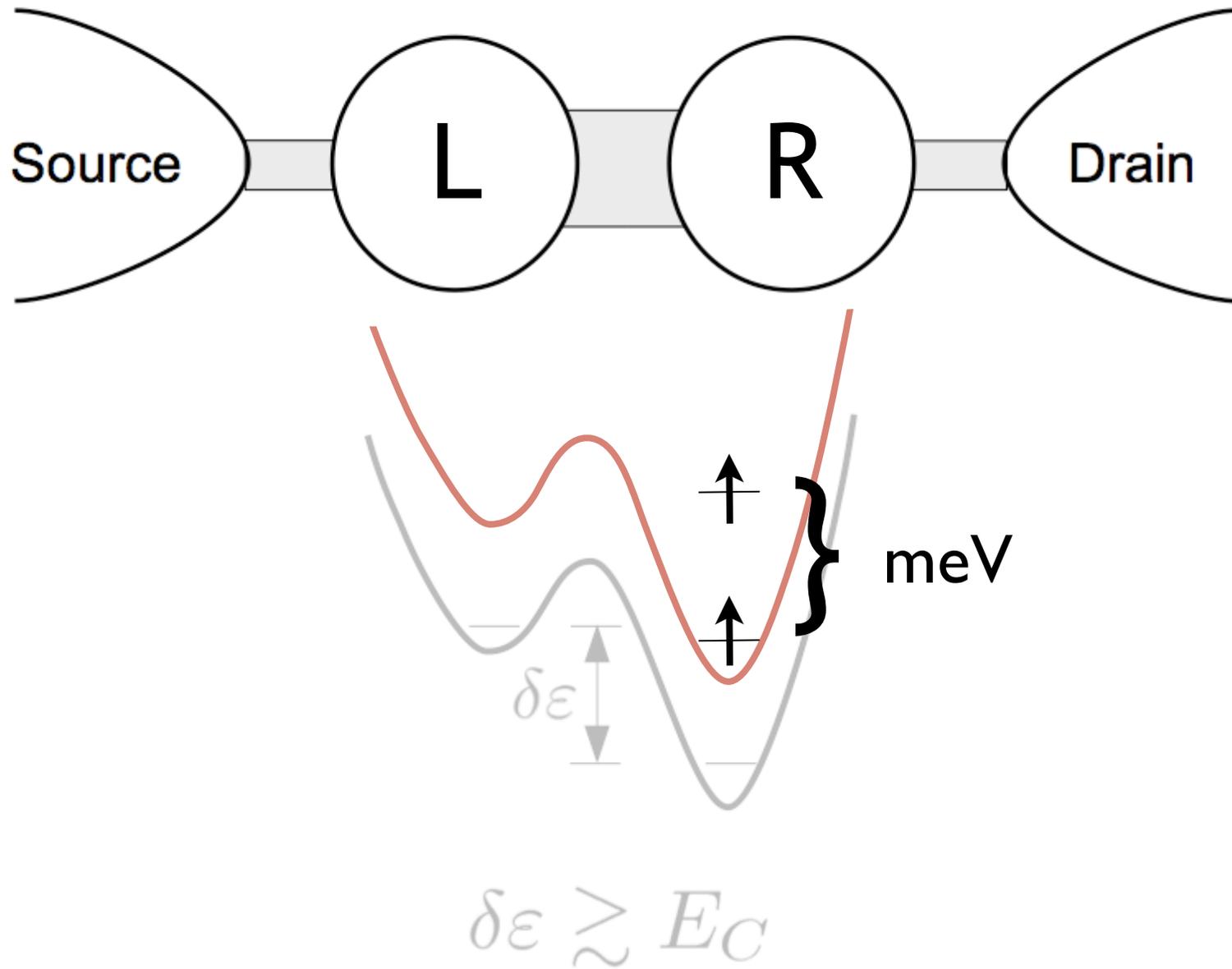
Pauli blockade exposes spin dynamics via charge motion



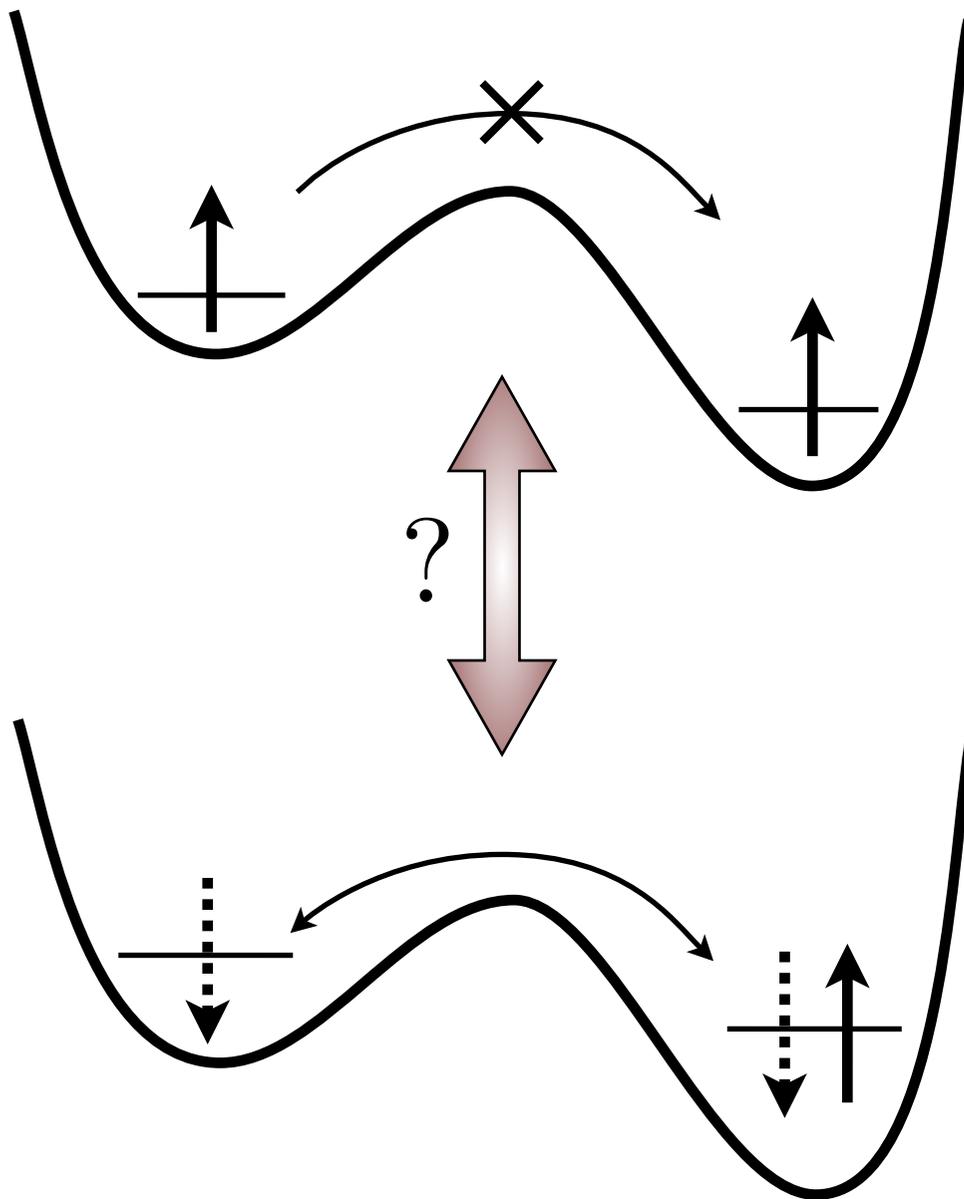
Pauli blockade exposes spin dynamics via charge motion



Pauli blockade exposes spin dynamics via charge motion

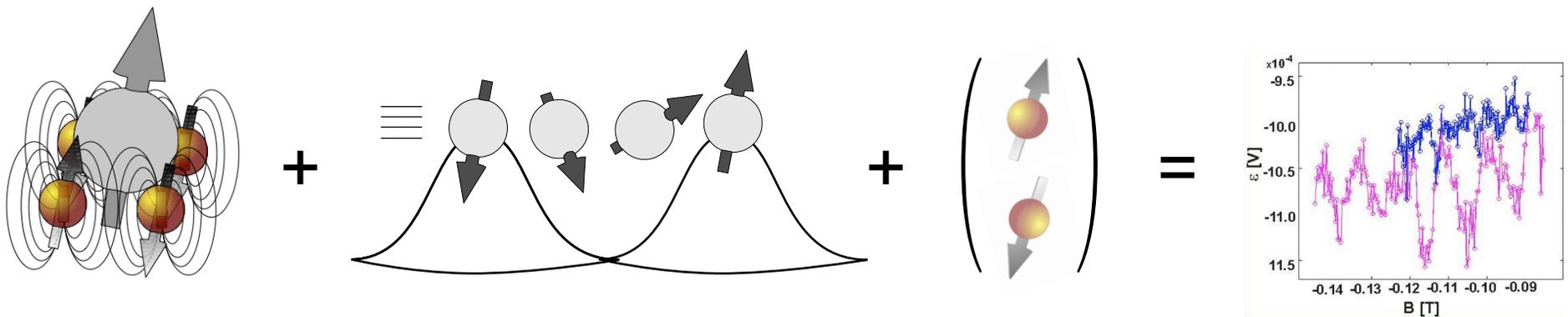


Dynamics controlled by coupling of singlet and triplet states



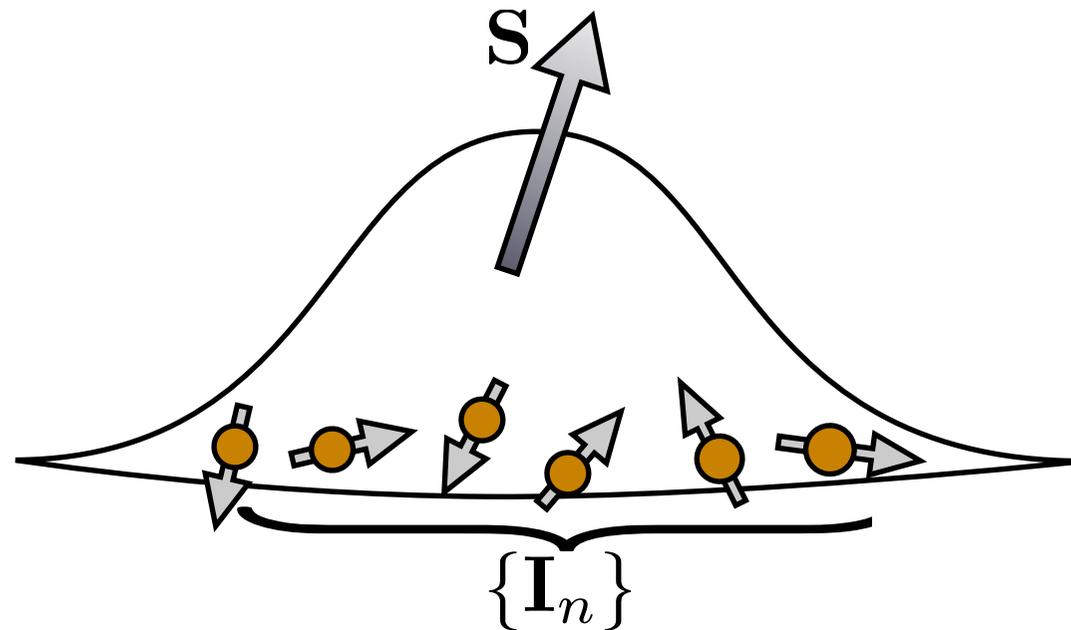
II. Coherent interplay of hyperfine and spin-orbit coupling

- * Polarization selection rules drastically altered
- * Long-lived nuclear spin coherence mediates interference
- * Effects revealed in pumping “commensuration resonances”



Hyperfine interaction couples electron, nuclear spins

$$H_{\text{HF}} = \sum_{n=1}^N A |\Psi(\mathbf{R}_n)|^2 \mathbf{I}_n \cdot \mathbf{S} \quad N \sim 10^6$$

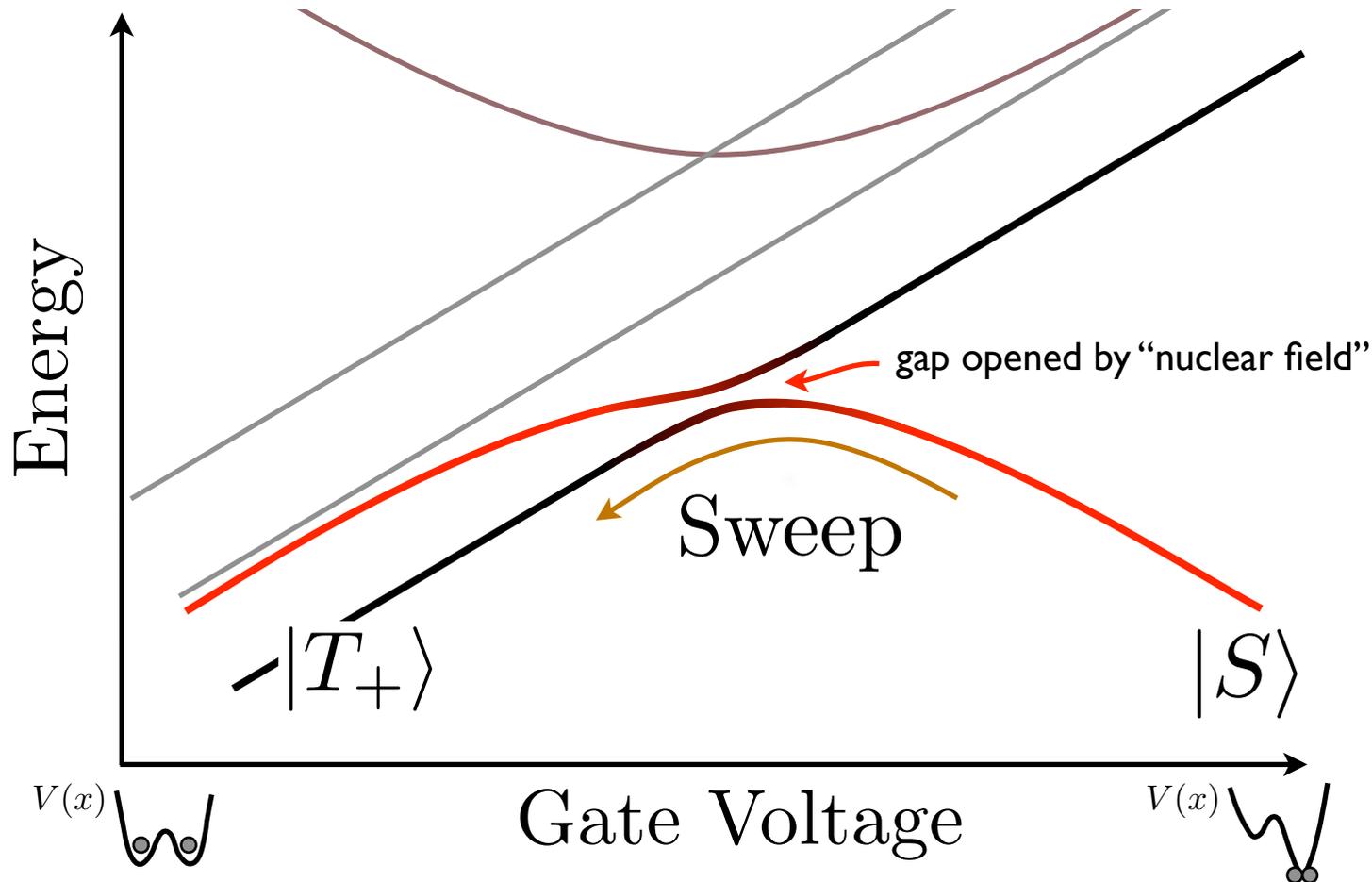


$$\mathbf{I} \cdot \mathbf{S} = \underline{I^z S^z} + \frac{1}{2} (\underline{I^+ S^-} + I^- S^+)$$

“Overhauser” term shifts
electron Zeeman energy

“flip-flop” terms allow exchange of
angular momentum

Sweeps through level crossing deposit angular momentum into nuclear spin bath

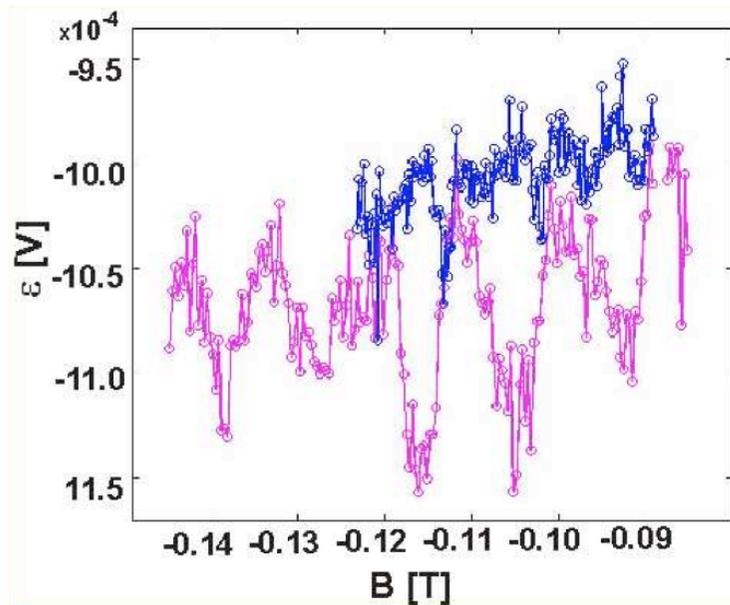


One nuclear "flop" per electron "flip"

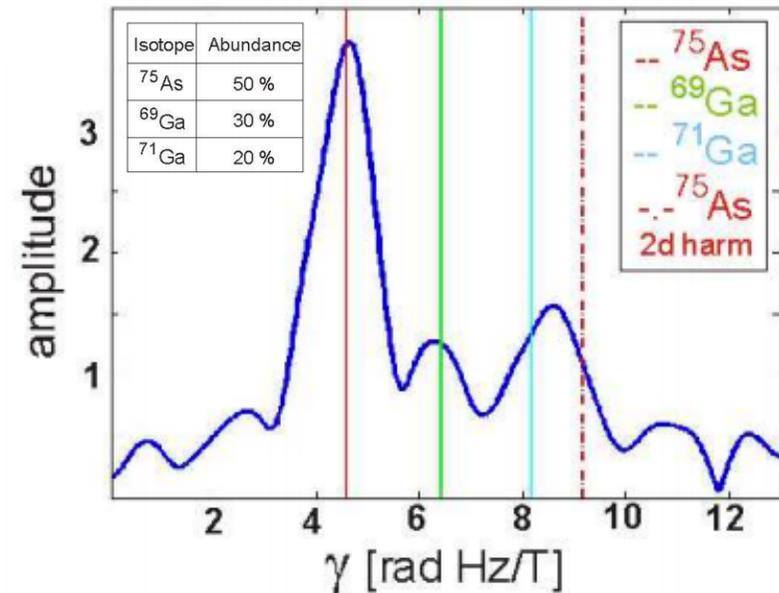
Sharp dips observed for cycle times commensurate with individual nuclear Larmor periods

Pumping with fixed total cycle time

Shift of $S-T_+$ resonance



Fourier transform of resonance shift



$$\gamma t_{\text{cycle}} \sim 2\pi / \Delta B$$

Why is pumping sensitive to precession in the *lab frame*?
How is the *sign* of polarization opposite to expectation?

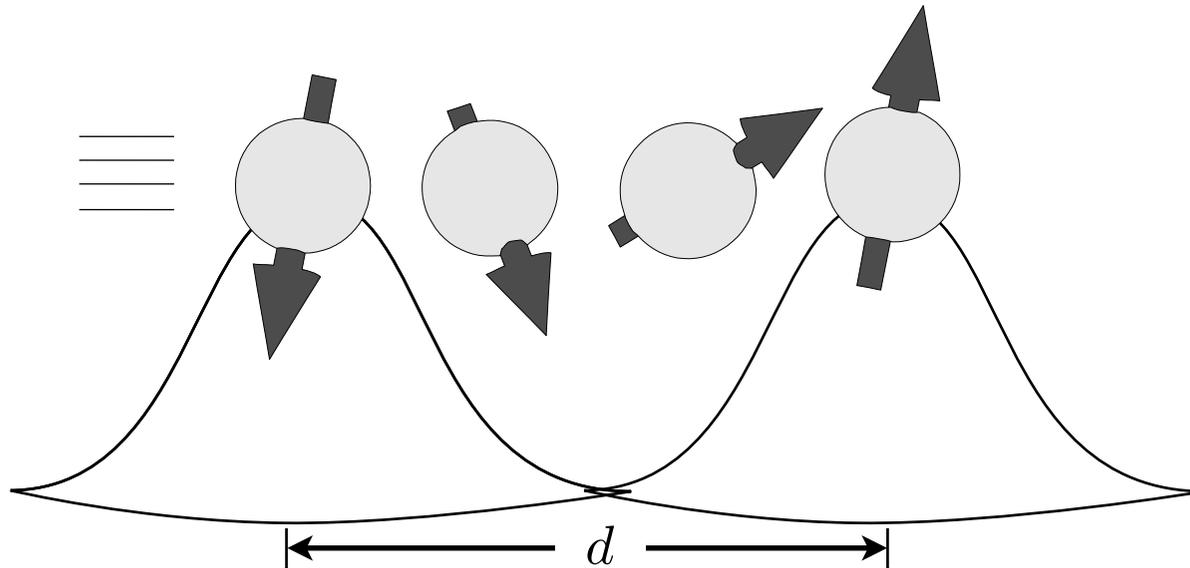
S. Foletti *et al.*, arXiv:0801.3613 (2008)

I. Neder, MR, and B. I. Halperin, arXiv:1309.3027 (2013).

Spin-orbit coupling induces spin rotation during tunneling

$$H_{\text{SO}} \sim \alpha(p_x \sigma_y - p_y \sigma_x)$$

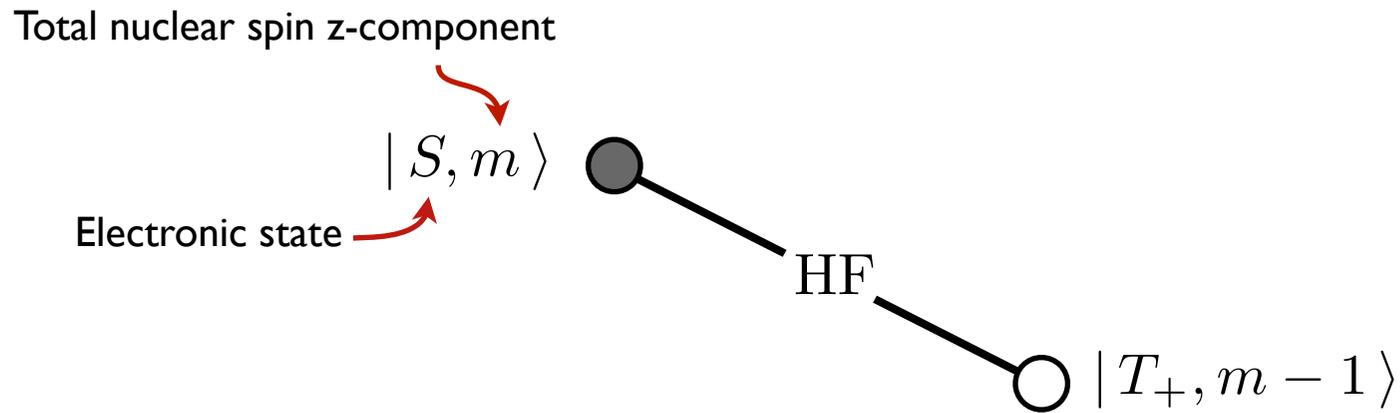
“spin-orbit field” \propto velocity



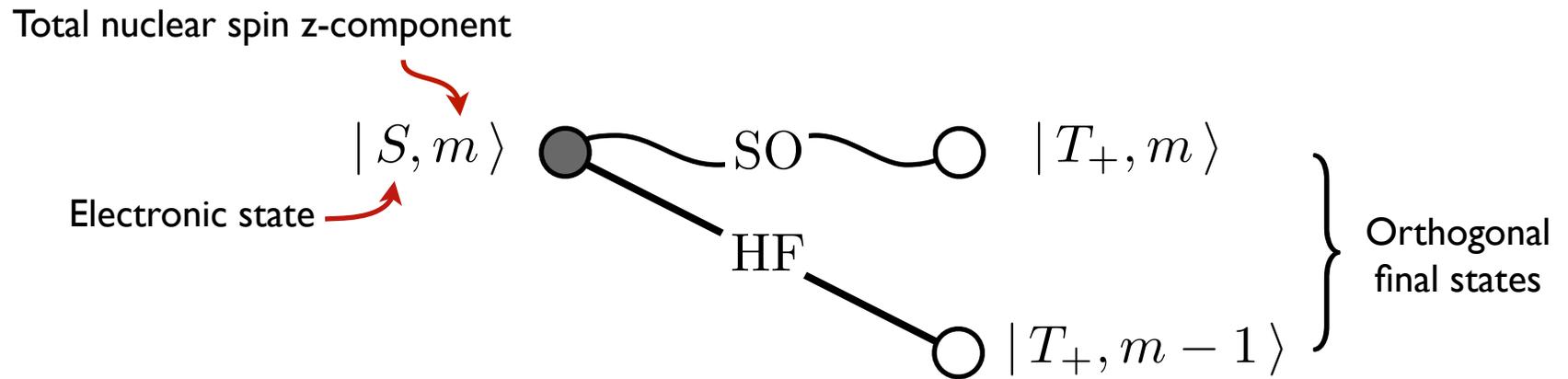
Displacement defines rotation angle

$$\theta_R \sim \frac{d}{\ell_{\text{SO}}}, \quad \ell_{\text{SO}} = \frac{\hbar}{m_* \alpha}$$

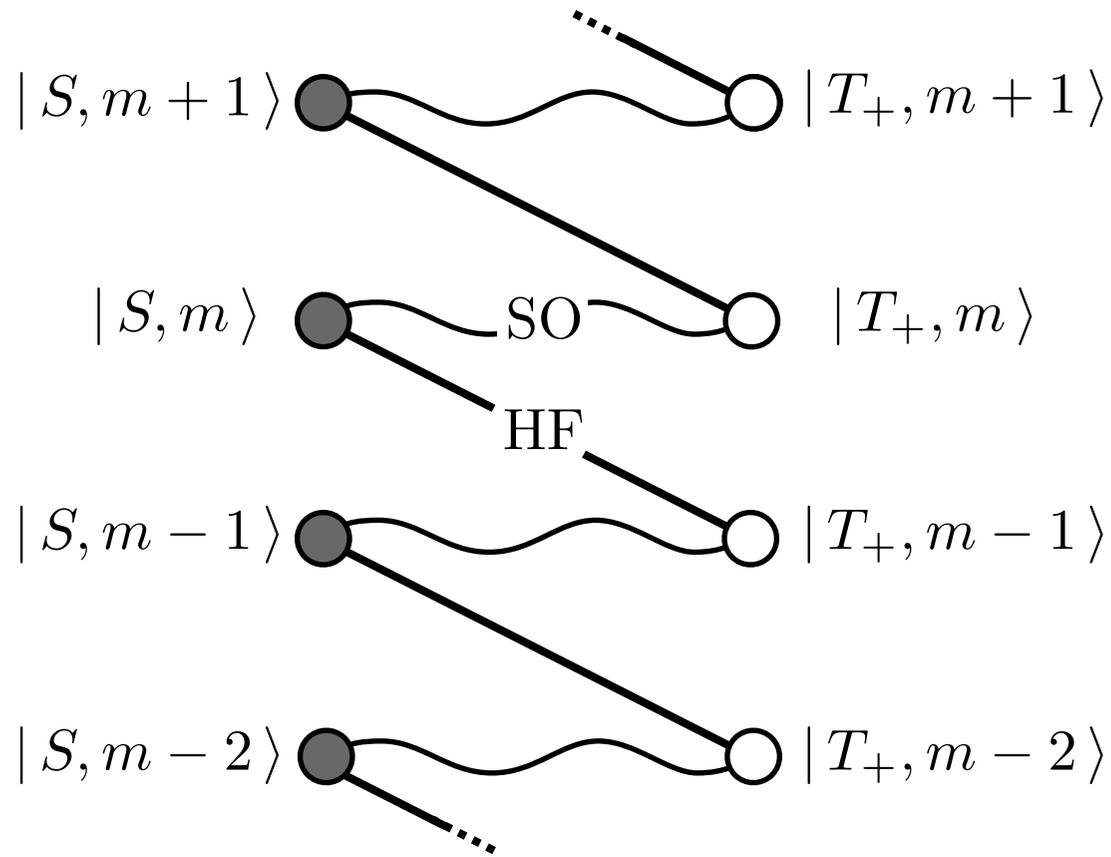
When spin flip mechanisms compete, no simple counting rule



When spin flip mechanisms compete, no simple counting rule



When spin flip mechanisms compete, no simple counting rule



Greatly expanded Hilbert space now accessible

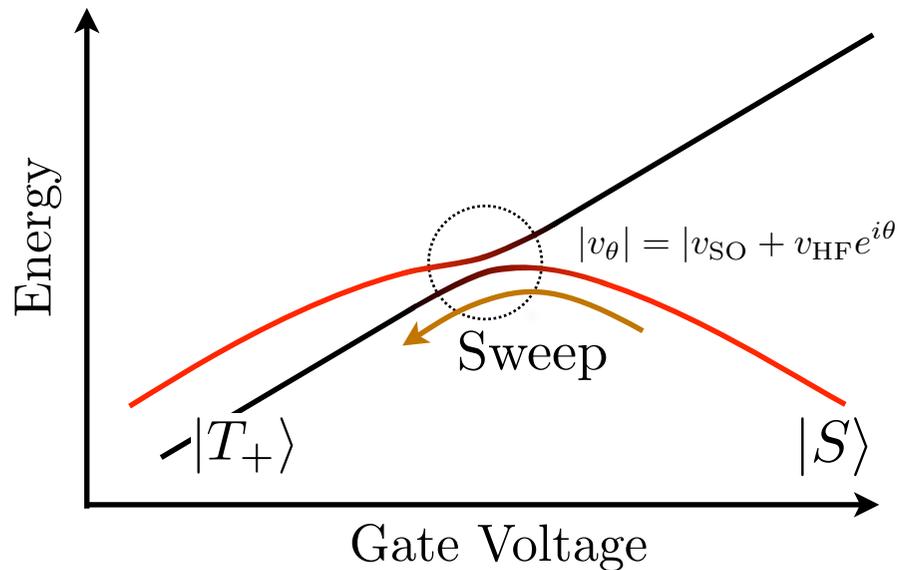
Assuming translation invariance in m , decouple problem into 2x2 blocks

Fourier space (Bloch) Hamiltonian

$$|\psi_\theta\rangle = \sum_m e^{im\theta} |\psi_m\rangle, \quad H_{ST_+}(\theta) = \begin{pmatrix} \varepsilon_{T_+}(t) & v_{SO} + v_{HF}e^{-i\theta} \\ v_{SO} + v_{HF}e^{i\theta} & \varepsilon_S(t) \end{pmatrix}$$

spin-orbit
matrix element

typical hyperfine
matrix element



see also, e.g.:

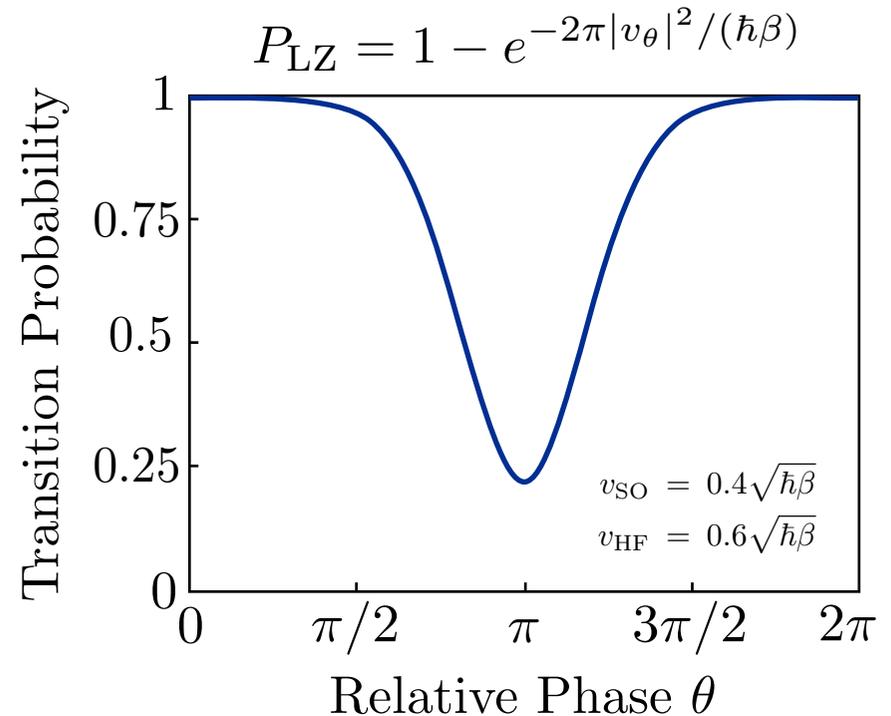
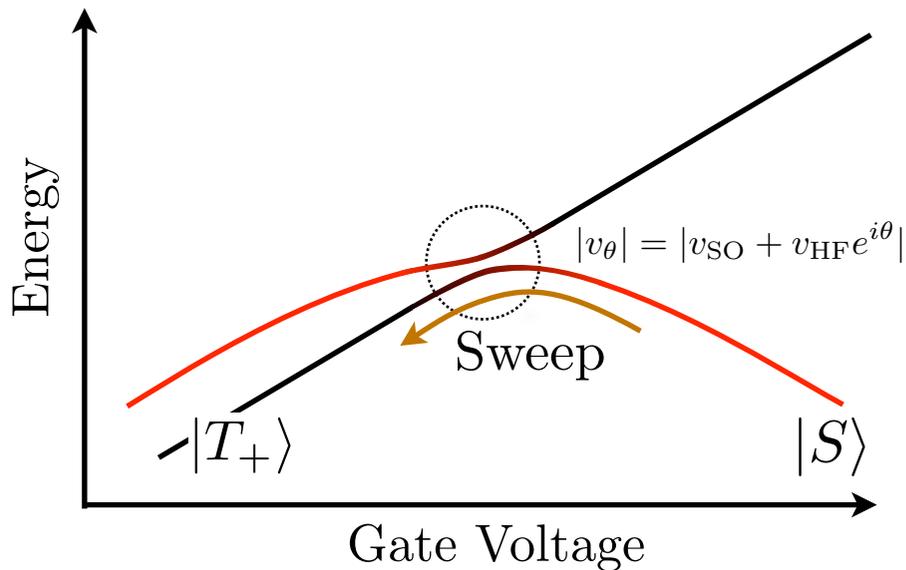
A. Brataas and E. I. Rashba, Phys. Rev. B **84**, 045301 (2011)

D. Stepanenko, MR, B. I. Halperin, and D. Loss, Phys. Rev. B **85**, 075416 (2012)

M. Gullans *et al.*, Phys. Rev. B **88**, 035309 (2013)

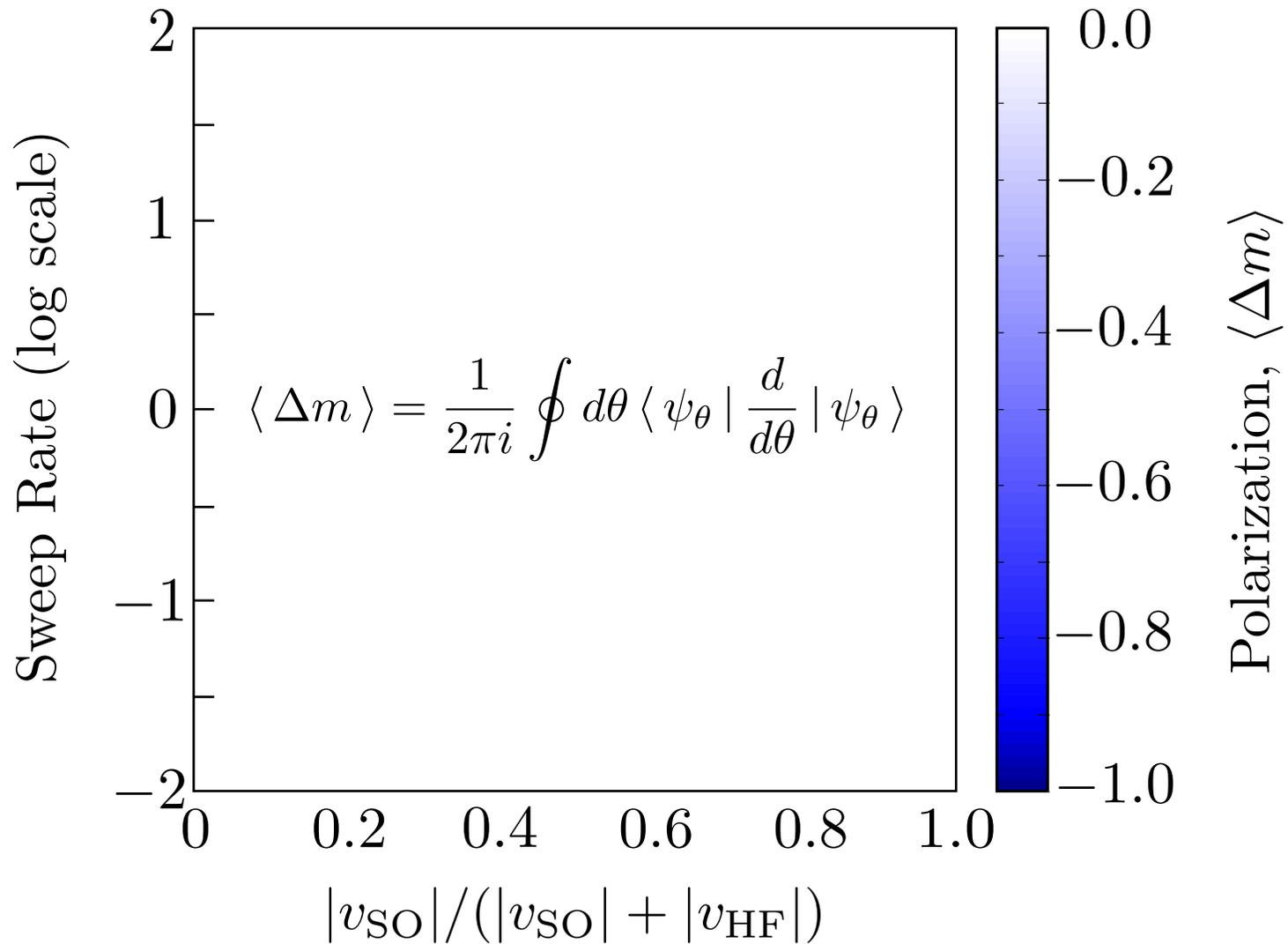
Phase θ controls gap for effective Landau-Zener problem

$$\varepsilon_S(t) - \varepsilon_{T_+}(t) = \beta t, \quad -T \leq t \leq T$$

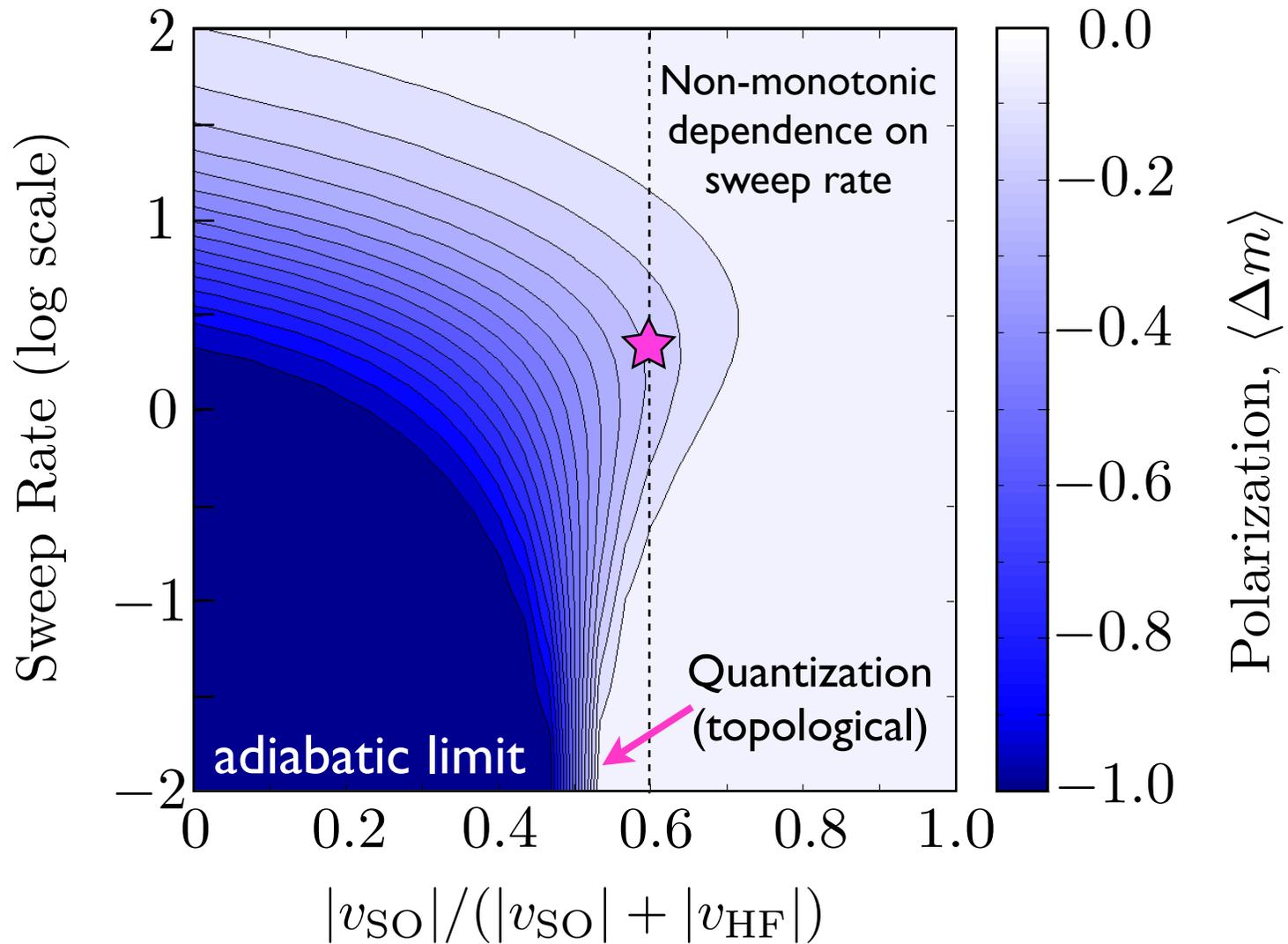


Hyperfine and spin-orbit processes interfere!

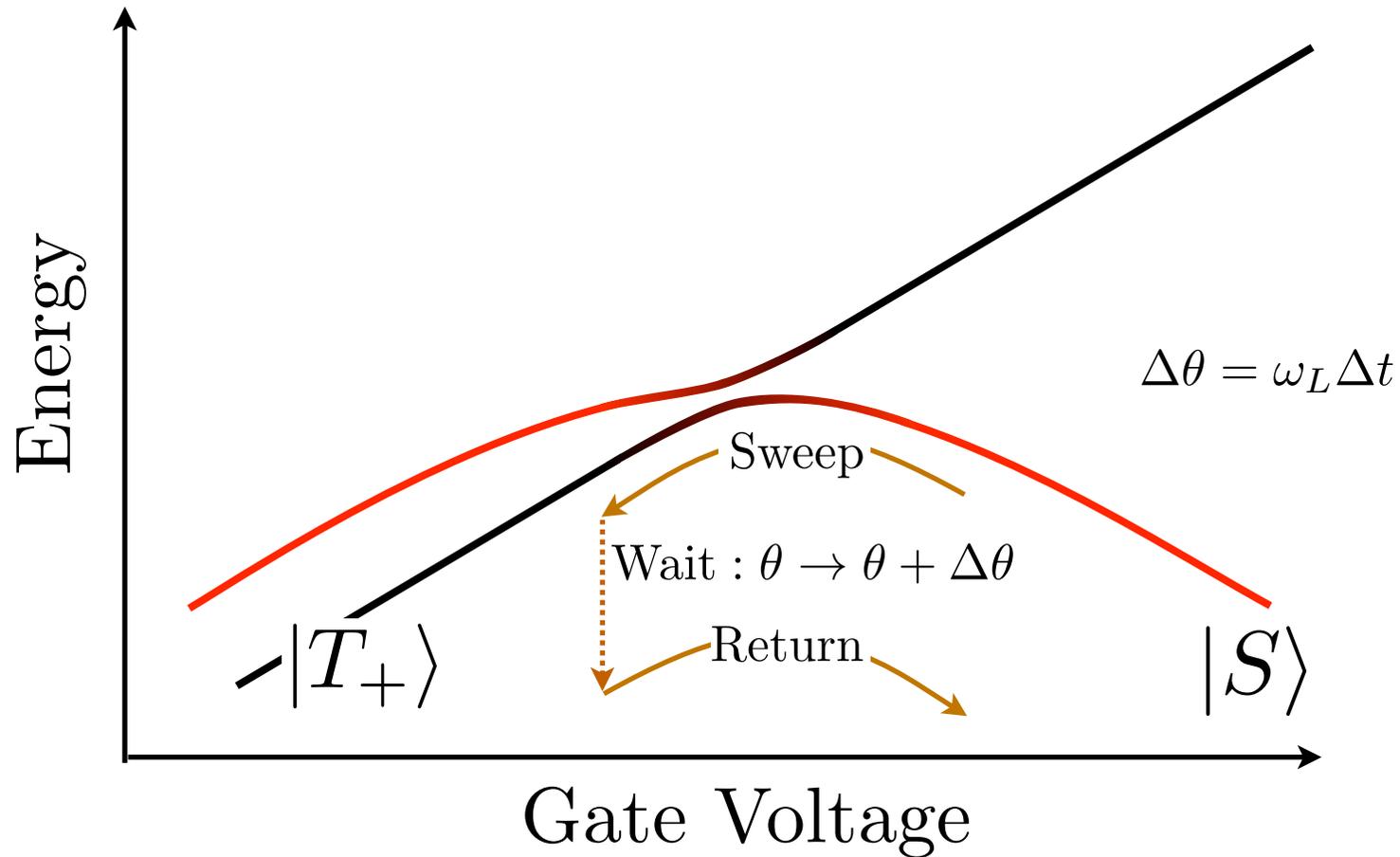
Pumped spin calculated from average displacement



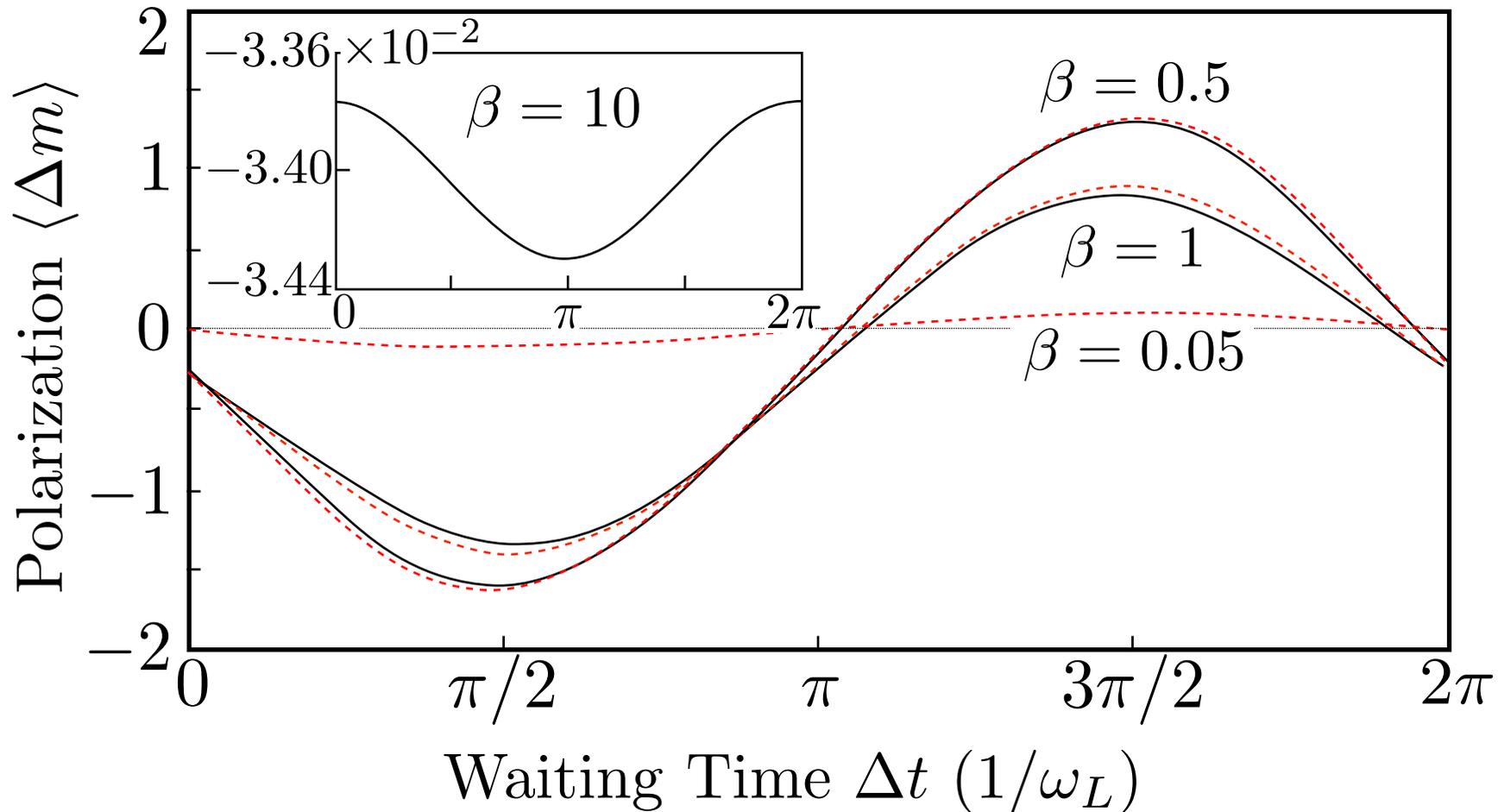
Adiabatic limit reveals complete suppression of nuclear spin pumping



Multiple sweeps: nuclear Zeeman energy causes Larmor precession during waiting period



Periodic dependence of pumped spin on Larmor precession angle!

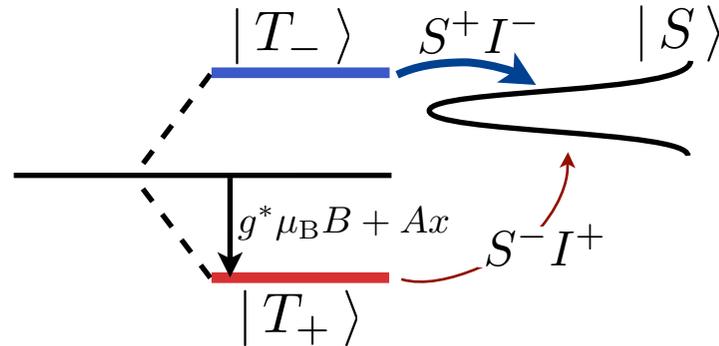


$$v_{\text{SO}}/v_{\text{HF}} = 2/3$$

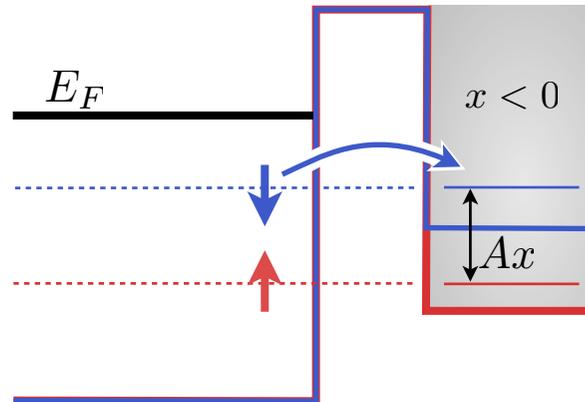
Spin-orbit field provides reference direction in lab frame

III. Nonlinear dynamics and self-oscillations of DNP

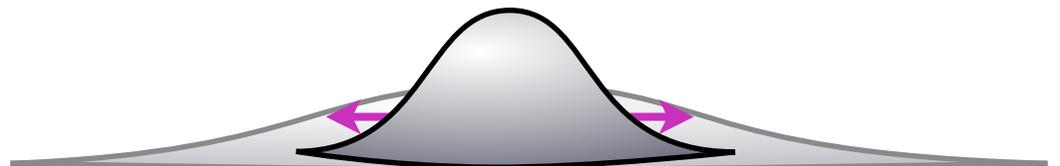
- * Energy-dependent hyperfine transition rates:



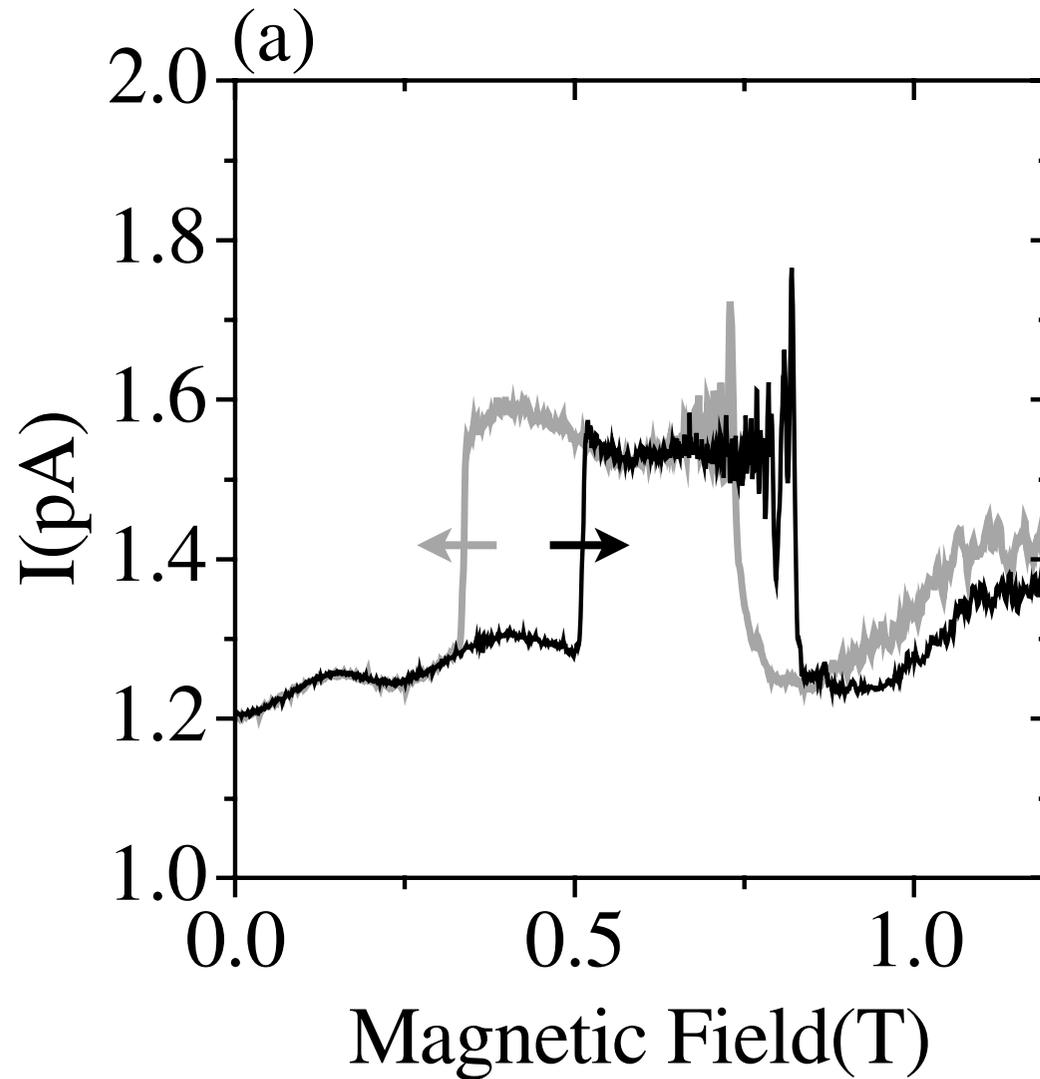
- * Spin-dependent tunneling due to inhomogeneous field:



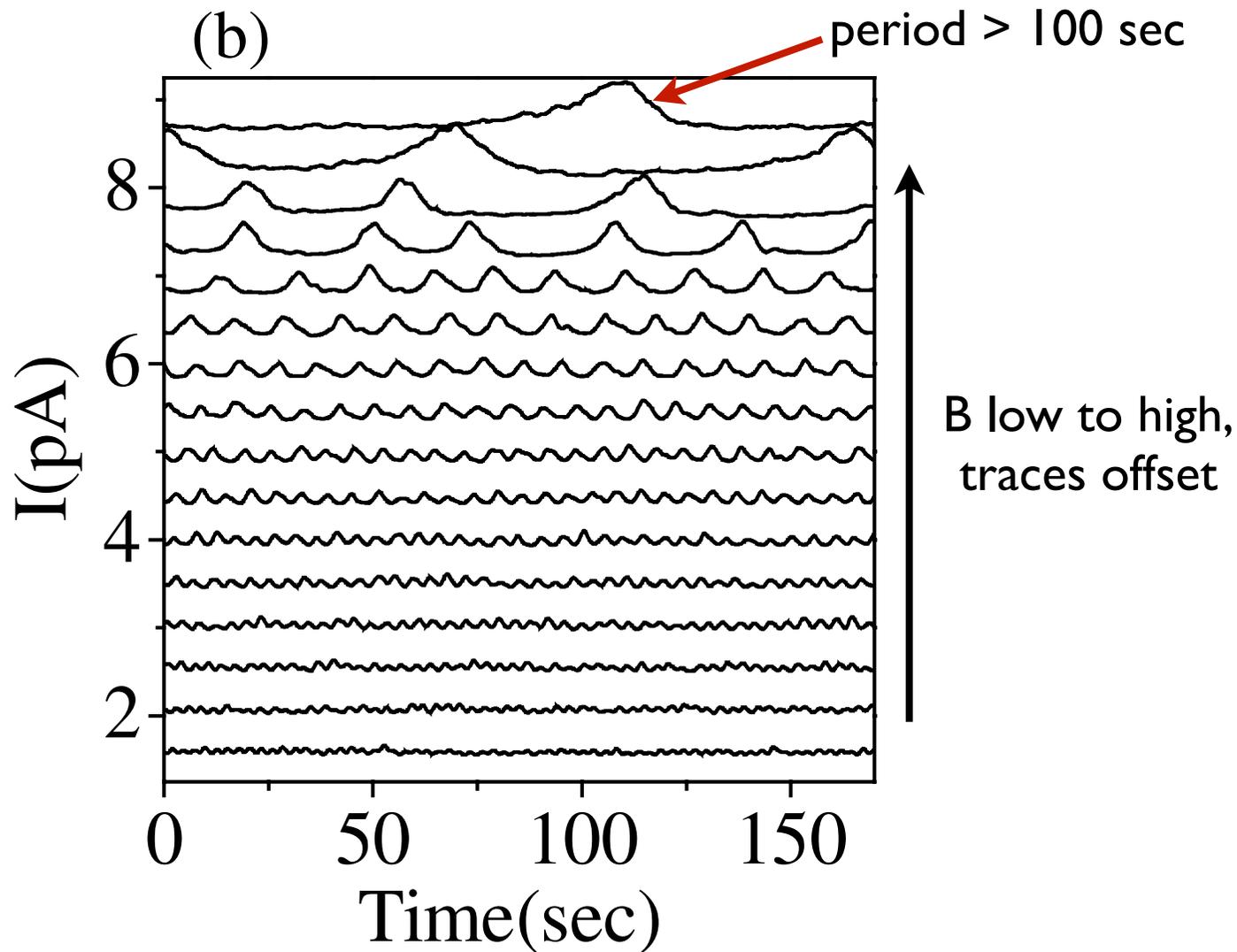
- * Nuclear spin diffusion:



Magnetic field dependence of current shows instabilities, hysteresis



Current oscillates for fixed B, with DC source-drain bias

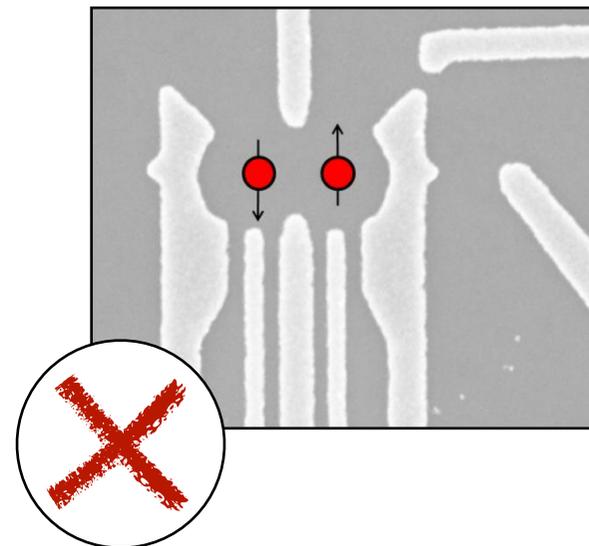
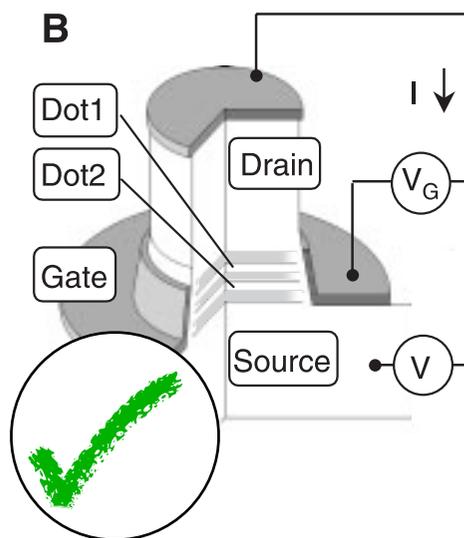


Theory must explain key experimental observations:

1. Extremely long oscillation timescale:

Oscillation period ~ 100 s
Single electron transit time (1 pA) ~ 100 ns
9 orders of magnitude separation!

2. Oscillations only observed in vertical DQDs



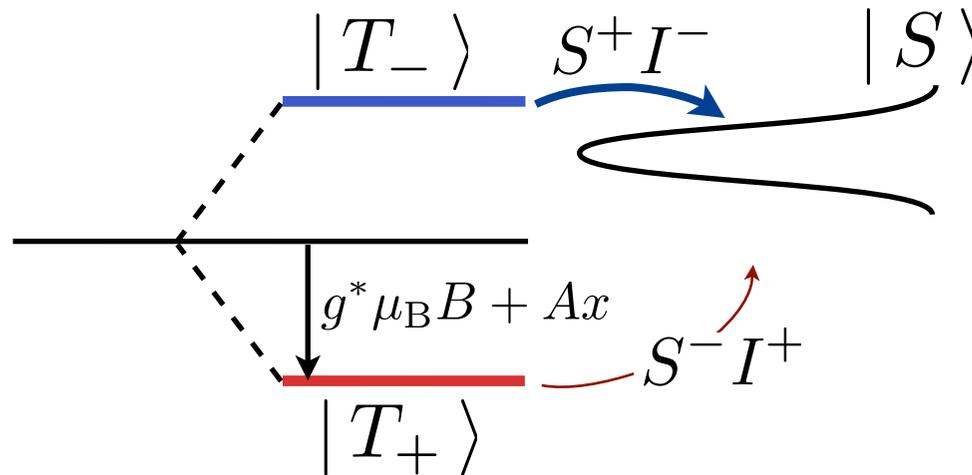
Nuclear polarization rate depends on *probabilities of up/down spin flip processes*

$$\Gamma_{\pm} \sim (\text{Attempt Freq.}) \cdot (\text{Prob. to load } T_{\pm}) \cdot (\text{Prob. of HF decay})$$

I/e

" f_{\pm} "

$W_{\pm}^{\text{HF}} / W_{\text{Tot}}$

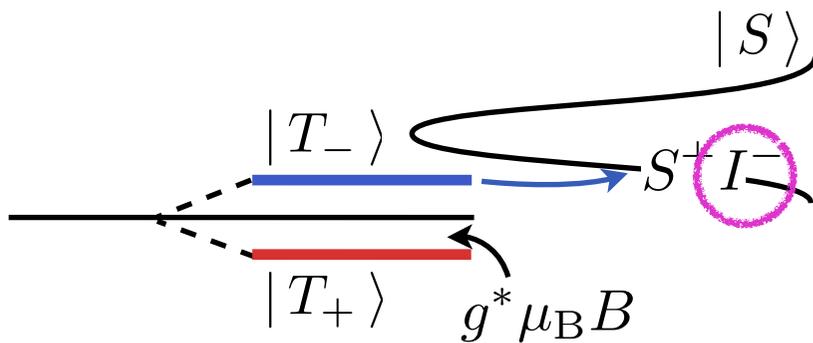


Rate equation for nuclear polarization, x :

$$\dot{x} = [f_{+}W_{+}^{\text{HF}} - f_{-}W_{-}^{\text{HF}}] / N \quad (W_{\text{Tot}} \gg W_{\pm}^{\text{HF}})$$

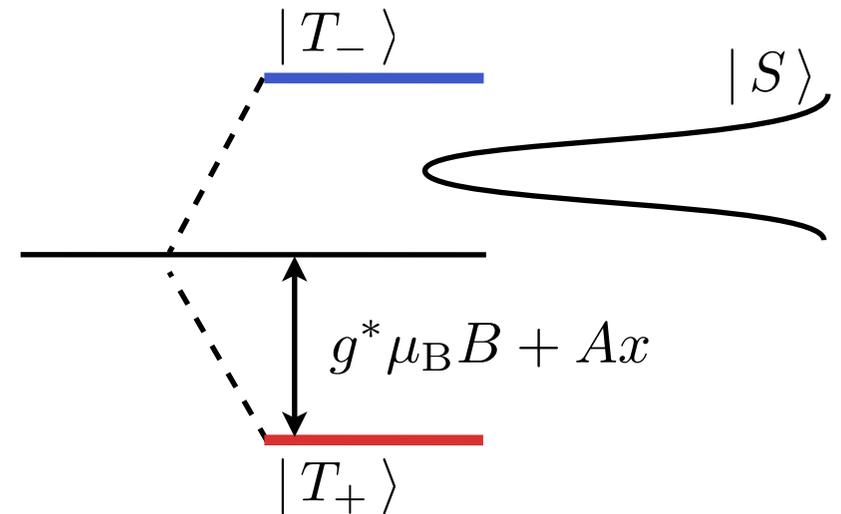
Dependence of polarization rate on Overhauser shift provides feedback, leads to instability

$$x \equiv \langle I_z \rangle = 0$$



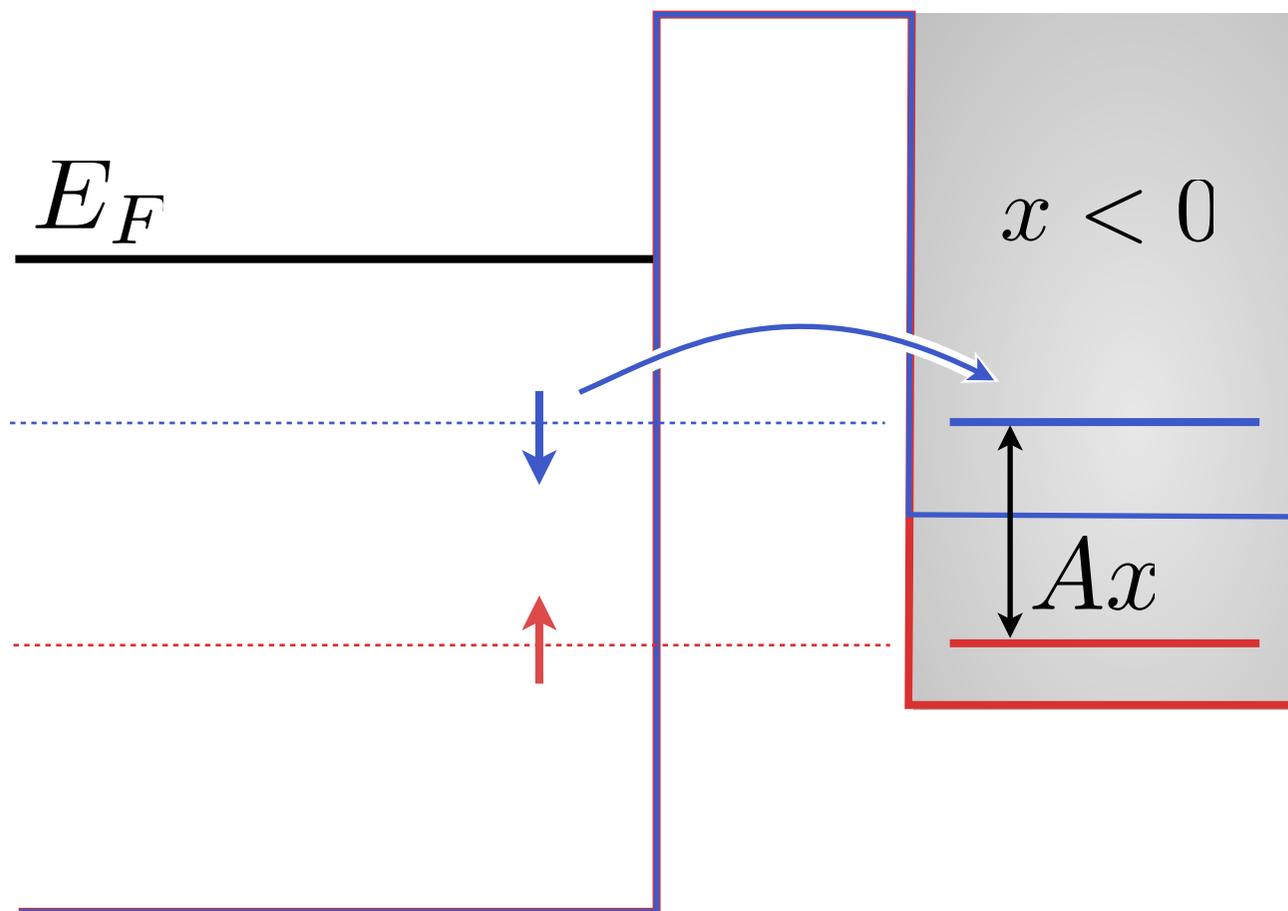
Hyperfine rates imbalanced

$$x < 0$$

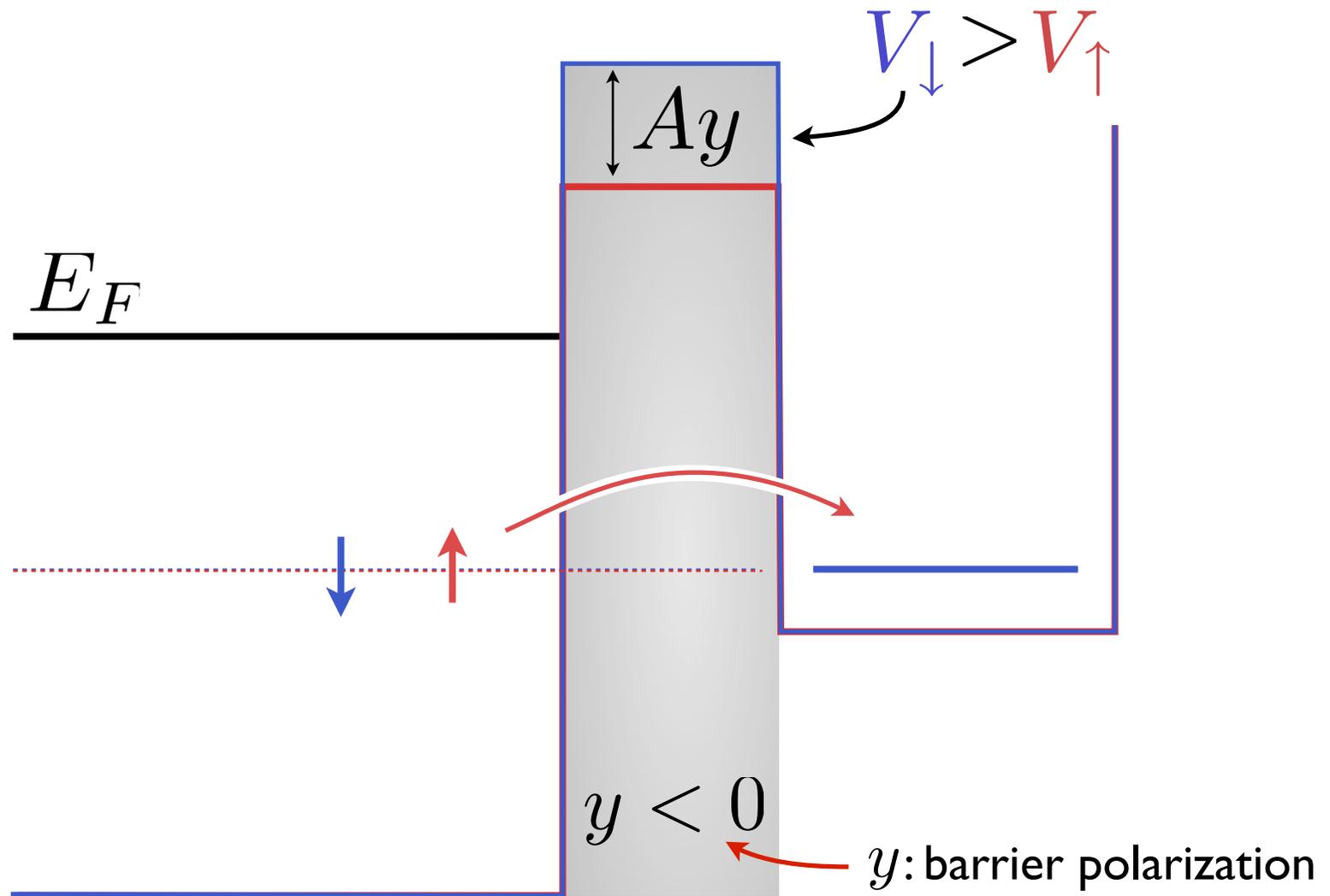


Polarization build-up inside dot,
saturates to steady state

Overhauser field only inside dot: down spins tunnel faster

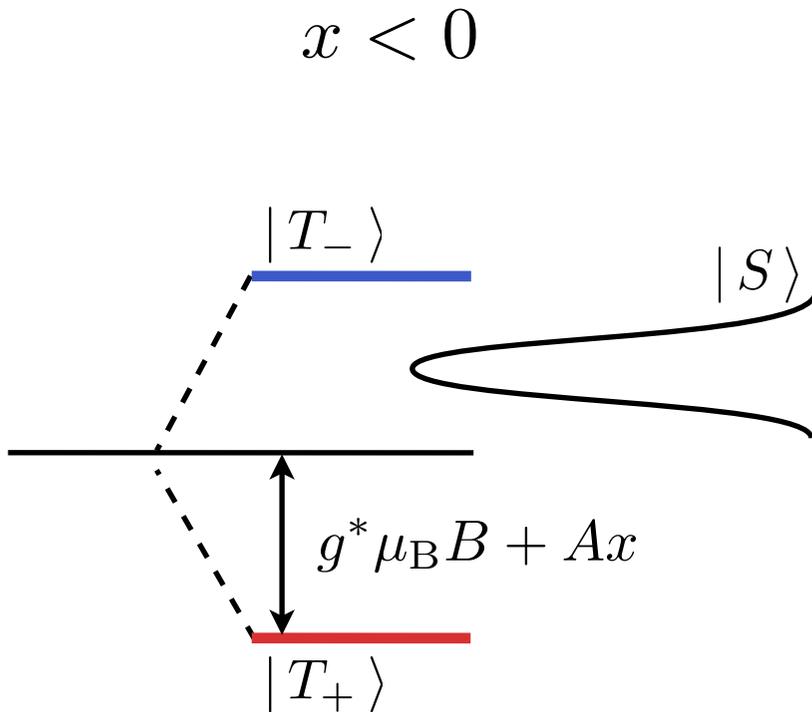


Overhauser field only inside barrier: up spins tunnel faster

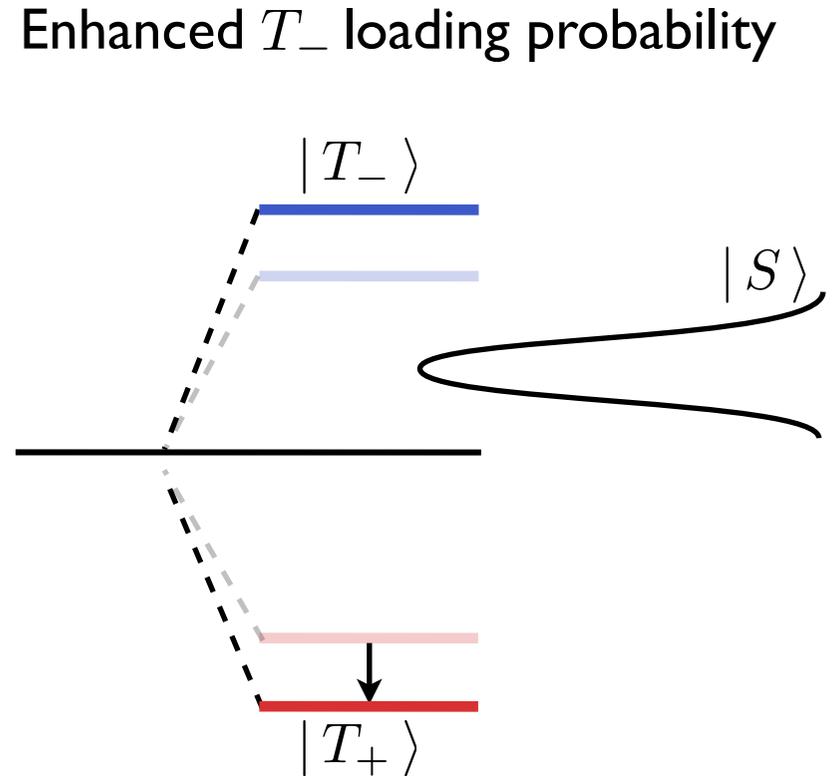


* Loading probabilities sensitive to DNP inhomogeneity

Polarization builds up inside dot; additional feedback due to spin-dependent tunneling



Polarization only inside dot



Polarization “overshoots”

Dot and barrier DNP coupled by slow nuclear spin diffusion

$$\begin{aligned}\dot{x} &= [f_+ W_+^{\text{HF}} - f_- W_-^{\text{HF}}] / N - 2\Gamma_D(x - y) \\ \dot{y} &= \Gamma_D x - 2\Gamma_D y\end{aligned}$$

Diffusion time constant: $\Gamma_D^{-1} \approx 10$ s for a few 10s of nm

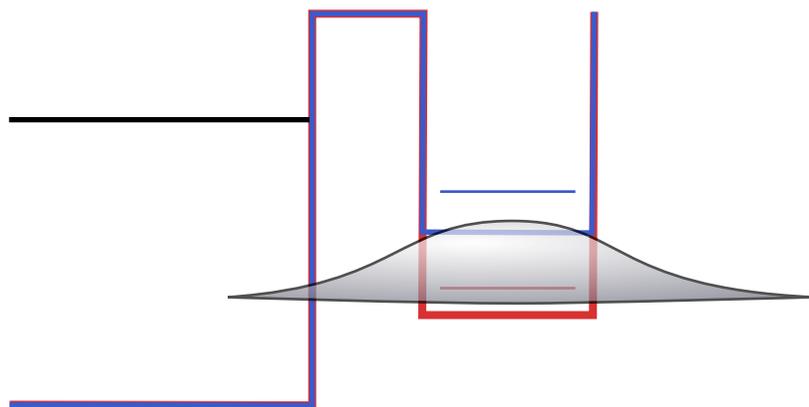
Loading probabilities $f_{\pm} \sim \frac{1}{4} [1 \pm \eta(x - y)]$ depend on DNP gradient

Diffusion references:

- D. Paget, Phys. Rev. B **25**, 4444 (1982)
- D. J. Reilly *et al.*, PRL **101**, 236802 (2008)

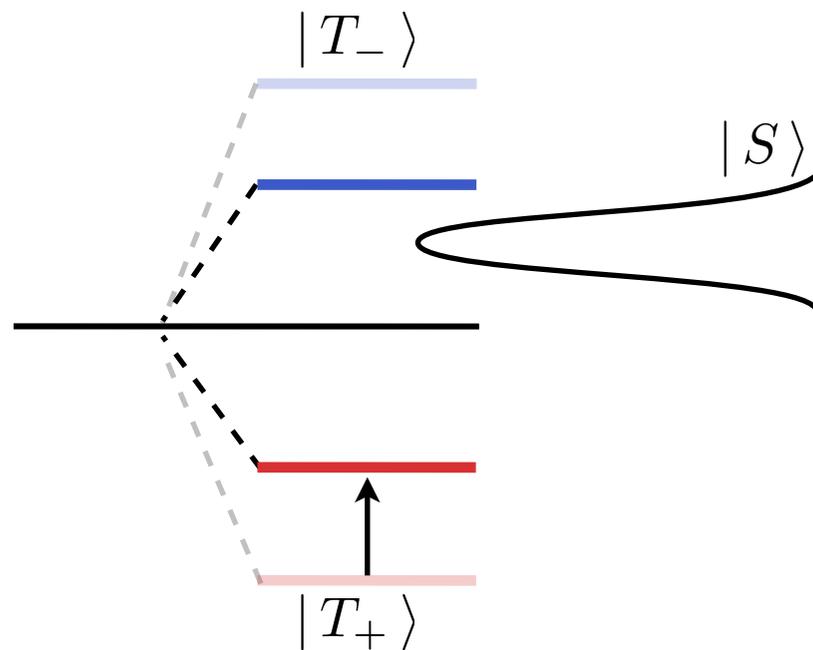
DNP diffuses into barrier, loading probabilities react with long time delay

Polarization diffuses to barrier



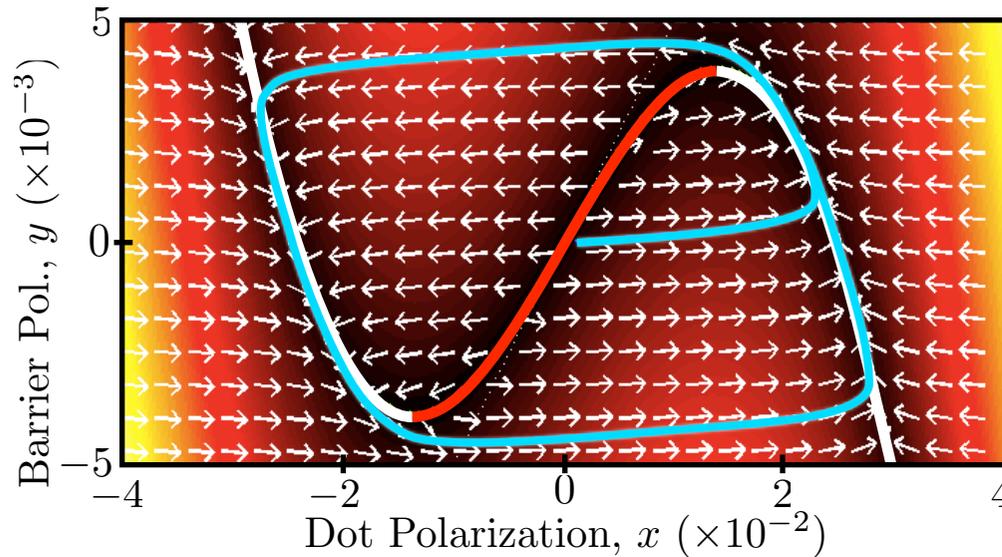
Loading probabilities equalized

Reduced T_- loading probability



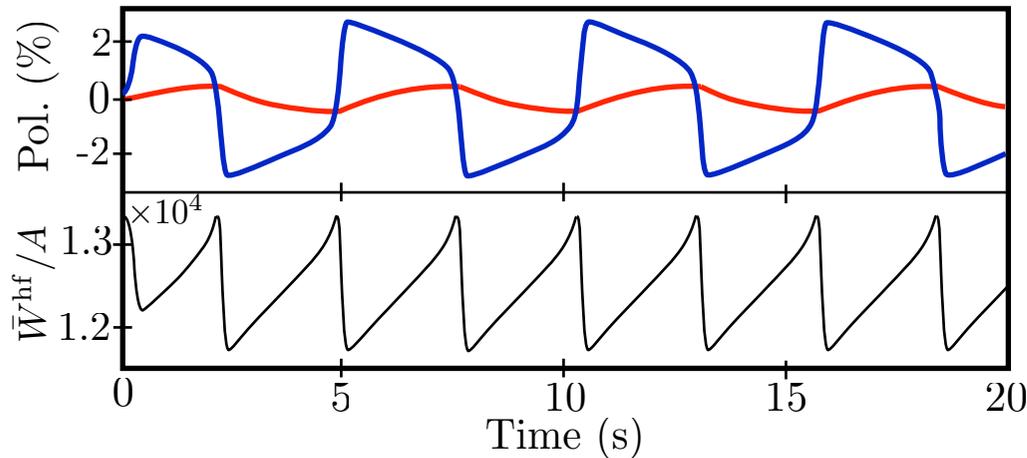
Polarization in dot driven back toward zero

Delayed feedback gives rise to stable limit cycle!



Arrows: flow direction
Color scale: flow velocity

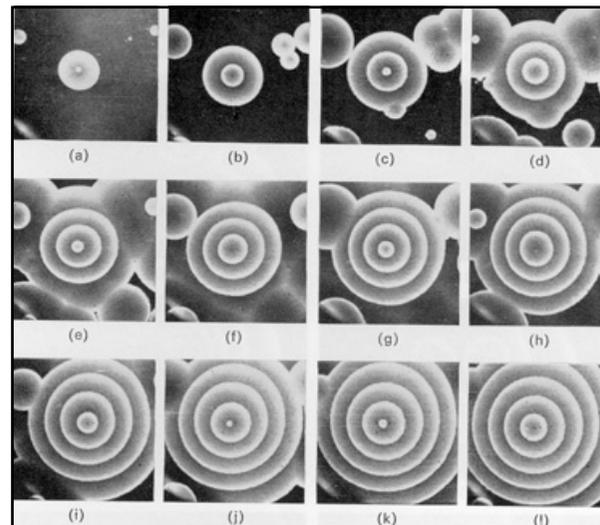
$\varepsilon/A = -0.0025$
 $\gamma/A = 0.075$
 $\Gamma_D/A = 10^{-12}$
 $B = 0$
 $\eta = 0.2$
 $N = 10^5$



- * Period set by nuclear spin diffusion, easily reaching tens of seconds
- * Directed spin diffusion into barrier expected only for vertical DQDs

Oscillatory phenomena provide new insight into the mechanisms of spin dynamics in QDs

- * Spin-orbit coupling drastically alters angular momentum counting
- * Long-lived nuclear spin coherence mediates interference between electronic spin flip pathways
- * Coupling of electronic degrees of freedom and spatial modes of nuclear polarization leads to intriguing new phenomena



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Assuming translation invariance in m , decouple problem into 2x2 blocks

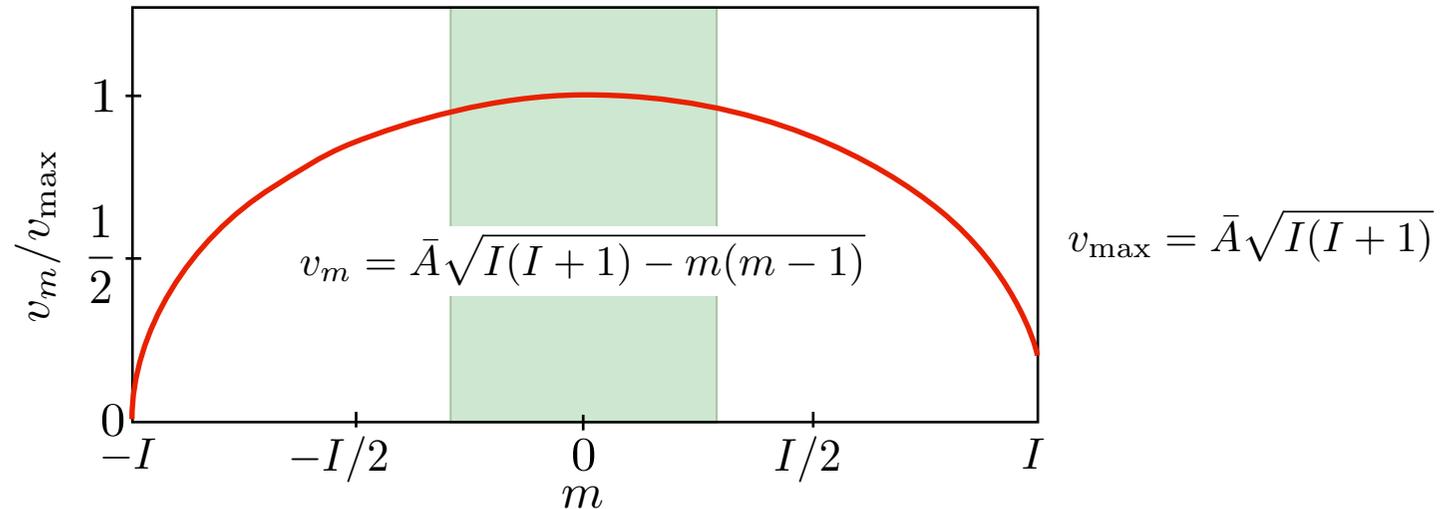
Fourier space (Bloch) Hamiltonian

$$|\psi_\theta\rangle = \sum_m e^{im\theta} |\psi_m\rangle, \quad H_{ST_+}(\theta) = \begin{pmatrix} \varepsilon_{T_+}(t) & v_{\text{SO}} + v_{\text{HF}} e^{-i\theta} \\ v_{\text{SO}} + v_{\text{HF}} e^{i\theta} & \varepsilon_S(t) \end{pmatrix}$$

spin-orbit
matrix element

typical hyperfine
matrix element

Hyperfine matrix element: $\langle T_+, m-1 | H_{\text{HF}} | S, m \rangle$



see also, e.g.:

A. Brataas and E. I. Rashba, Phys. Rev. B **84**, 045301 (2011)

D. Stepanenko, MR, B. I. Halperin, and D. Loss, Phys. Rev. B **85**, 075416 (2012)

M. Gullans *et al.*, Phys. Rev. B **88**, 035309 (2013)

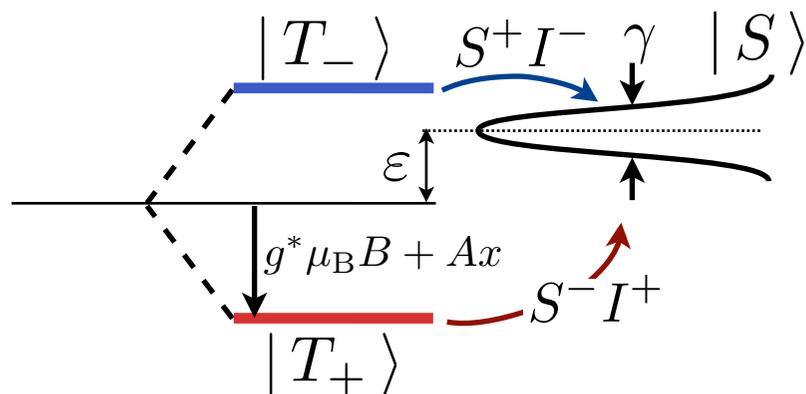
DNP production rate controlled by HF transition rates

$$\Gamma_{\pm} = (\text{Attempt freq.}) \cdot (\text{Prob. to load } T_{\pm}) \cdot (\text{Prob. to decay by spin flip})$$

average current

" f_{\pm} "

$$\frac{W_{\pm}^{\text{HF}}}{W_{\pm}^{\text{HF}} + W^{\text{in}}}$$



W^{in} : nuclear-spin-independent escape rate

W_{\pm}^{HF} : elastic T_{\pm} - S hyperfine transition rate

$$W_{\pm}^{\text{HF}} = \frac{A^2 (1 \mp x) \gamma}{N \epsilon_{\pm}^2 + \gamma^2}$$

A : hyperfine coupling

N_{\pm} : nuclear spin populations

$$\epsilon_{\pm} = \epsilon \pm g^* \mu_B B \pm Ax$$

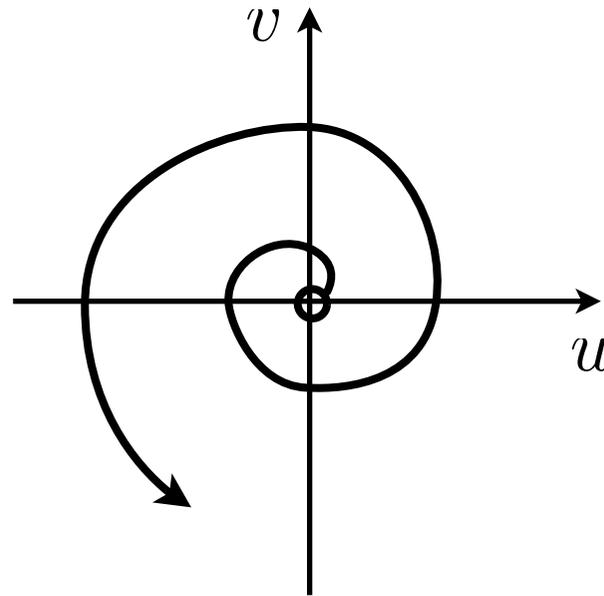
$$x = \frac{N_{+} - N_{-}}{N_{+} + N_{-}}$$

$$N = N_{+} + N_{-}$$

Rate equation for nuclear polarization:

$$\dot{x} = [\Gamma_{+}(x; B, \epsilon, \dots) - \Gamma_{-}(x; B, \epsilon, \dots)] / N$$

Finding oscillations: look for unstable spiral in linearized eqns



$$\dot{u} = \alpha u + v, \quad \dot{v} = -\mu u + \beta v$$

$$\lambda_{\pm} = \frac{1}{2}(\alpha + \beta) \pm \frac{1}{2}\sqrt{(\alpha - \beta)^2 - 4\mu^2}$$

Instability (positive real part of eigenvalues): $(\alpha + \beta) > 0$

Negative discriminant (complex eigenvalues): $(\alpha - \beta)^2 - 4\mu < 0$