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Time-dependent effects in atomistic spin dynamics

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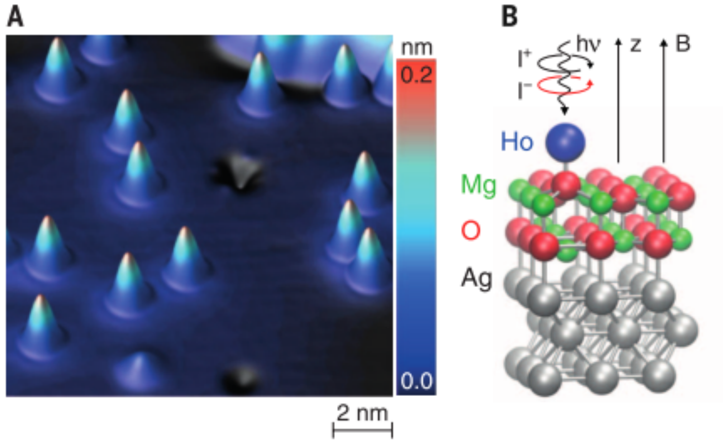
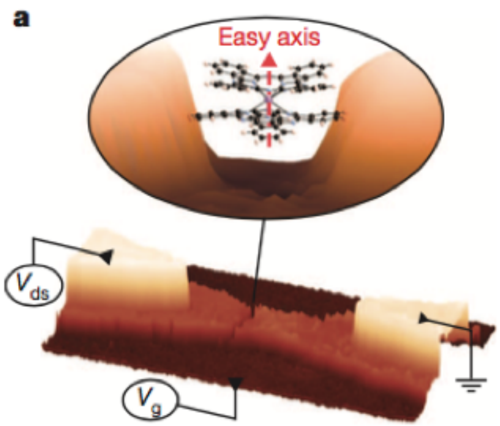
Single-molecule magnets

- Realization of stable single-atom magnets
- Control and read-out
- Spintronics devices
- Magnetic memories



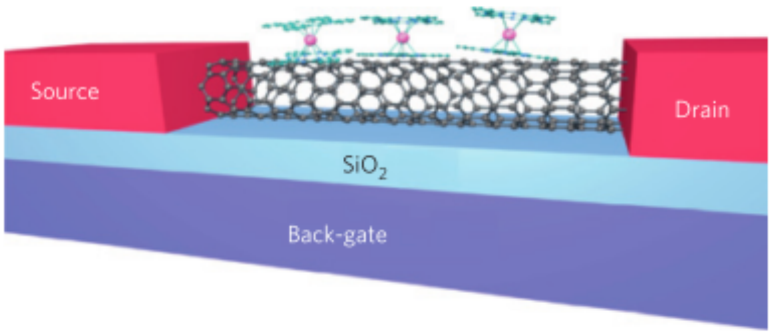
Science, 352(6283):318–321, 2016.



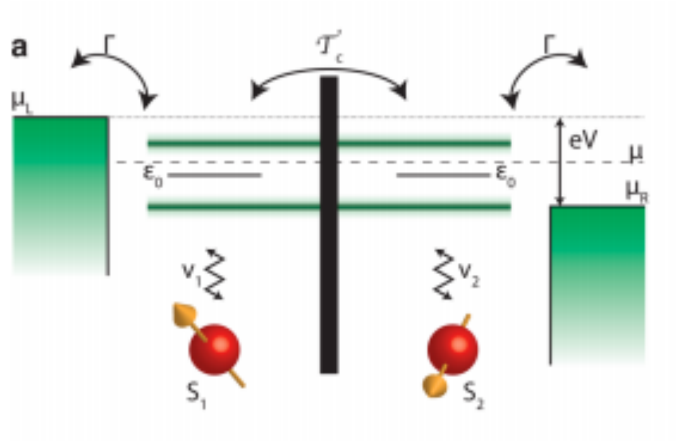
Nature, 488(7411):357–60, 2012.

Interesting physics ahead

- Dynamics of atomistic systems
- Systems out-of-equilibrium
- Need to extend theory of:
  - Exchange interaction
  - Electronic control
  - Spin dynamics

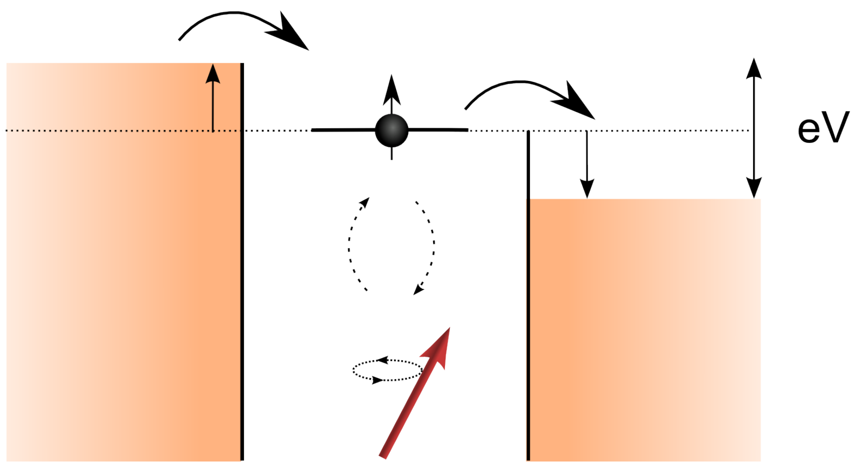


Nature materials, 10(7):502–506, 2011.



Nano Lett. 2016, 16, 2824−2829

Dynamics of a single-molecule system

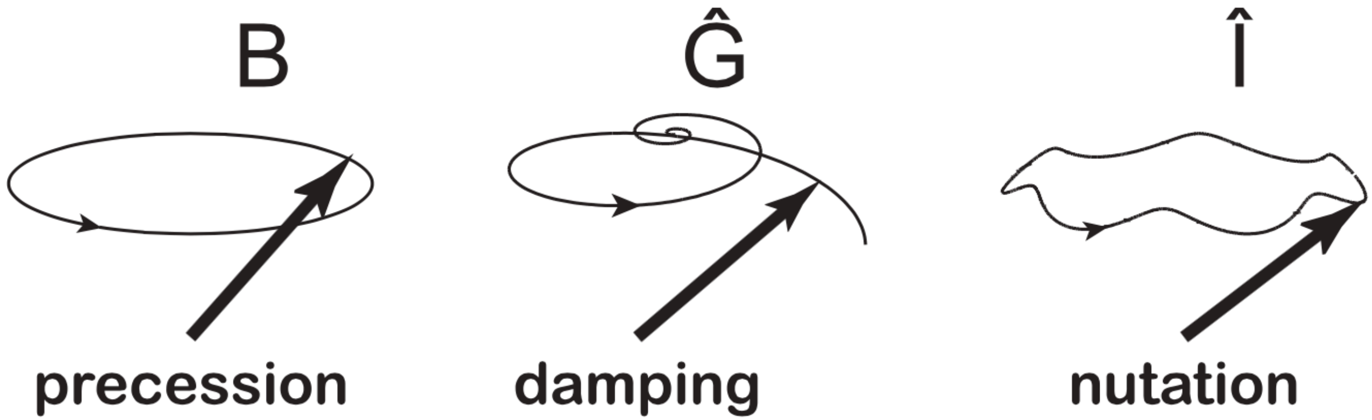


- Molecule in a tunnel junction
- Out-of-equilibrium
- Study transient behavior and fast dynamics in non-Markovian regime

Phenomological equation

- Spin dynamics for macroscopic systems:
  - Extended Landau-Lifshitz-Gilbert equation

$$\dot{\mathbf{S}} = \mathbf{S} \times \left(-\gamma \mathbf{B} + \hat{\mathbf{G}}\dot{\mathbf{S}} + \hat{\mathbf{I}}\ddot{\mathbf{S}}\right)$$



Phenomological equation

- Landau-Lifshitz-Gilbert equation

$$\dot{\mathbf{S}} = \mathbf{S} \times \left(-\gamma \mathbf{B} + \hat{\mathbf{G}}\dot{\mathbf{S}} + \hat{\mathbf{I}}\ddot{\mathbf{S}}\right)$$

- Going from macroscopic to atomistic systems

- How to treat quantum and nonequilibrium effects? How to take into account non-Markovian effects?

Spin action

- Derive the spin equation of motion

- Effective spin action for a spin in nonequilibrium

$$\mathcal{S} = \mathcal{S}_{WZWN} + \mathcal{S}_Z + \mathcal{S}_{int}$$

- Minimize the spin action and treat the spin classicaly

PRL 92, 107001 (2004)
PRL 108, 057204 (2012)
PRL 113, 257201 (2014)

Spin equation of motion

- Gives spin equation of motion

Exchange interaction

$$\dot{\mathbf{S}}(t) = \mathbf{S}(t) \times \left(-g\mu_B \mathbf{B}^{eff}(t) + \frac{1}{e} \int \mathbb{J}(t,t') \cdot \mathbf{S}(t') dt' \right)$$

Spin memory of all past spin states

- Effective magnetic field

$$\mathbf{B}^{eff}(t) = \mathbf{B} + \frac{1}{eg\mu_B} \int \boldsymbol{\epsilon}\mathbf{j}(t,t') dt'$$

Effective field due to charge background

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Spin equation of motion

$$\mathbf{J}(t,t') = \frac{ie}{2} v^2 \theta(t-t') sp \boldsymbol{\sigma} \mathbf{G}(t',t) \boldsymbol{\sigma} \mathbf{G}(t,t')$$

- Calculated from electronic structure given by some Hamiltonian $H = H_0 + H_M$

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Spin equation of motion

- Spin equation of motion

$$\dot{\mathbf{S}}(t) = \mathbf{S}(t) \times \left(-g\mu_B \mathbf{B}^{eff}(t) + \frac{1}{e} \int \mathbb{J}(t,t') \cdot \mathbf{S}(t') dt' \right)$$

- LLG equation

$$\dot{\mathbf{S}} = \mathbf{S} \times (-\gamma \mathbf{B} + \hat{\mathbf{G}}\dot{\mathbf{S}} + \hat{\mathbf{I}}\ddot{\mathbf{S}})$$

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Time-dependet parameters

- Derive LLG from our spin equation of motion
- First simplification: Time-dependent parameters, disregard spin history
- Taylor expand the exchange interaction

$$\frac{1}{e}\int \mathbb{J}(t,t')\cdot\mathbf{S}(t')dt' \approx \frac{1}{e}\Big(\int \mathbb{J}(t,t')dt'\mathbf{S}(t) - \int \mathbb{J}(t,t')(t-t')dt'\dot{\mathbf{S}}(t) + \int \mathbb{J}(t,t')(t-t')^2 dt'\ddot{\mathbf{S}}(t)/2\Big).$$

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Time-dependent parameters

- Identify

$$\mathbf{B}^{eff}(t) = \mathbf{B} + \frac{1}{eg\mu_B}\int \boldsymbol{\epsilon}\mathbf{j}(t,t')dt' + \int \mathbb{J}(t,t')dt'\mathbf{S}(t)$$

$$\hat{\mathbf{G}} = -\frac{1}{e}\int \mathbb{J}(t,t')(t-t')dt'$$

$$\hat{\mathbf{I}} = \frac{1}{2e}\int \mathbb{J}(t,t')(t-t')^2 dt'$$

PRL 108, 057204 (2012)

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Stationary limit

- Simplify it further: Stationary limit

$$\hat{\mathbf{G}} = -\frac{1}{e}\int \mathbb{J}(t,t')(t-t')dt' = -\frac{1}{e} lim_{\epsilon\to 0} i\partial_{\epsilon}\mathbb{J}(\epsilon)$$

$$\hat{\mathbf{I}} = \frac{1}{2e}\int \mathbb{J}(t,t')(t-t')^2 dt' = -\frac{1}{2e} lim_{\epsilon\to 0} i\partial_{\epsilon}^2\mathbb{J}(\epsilon)$$

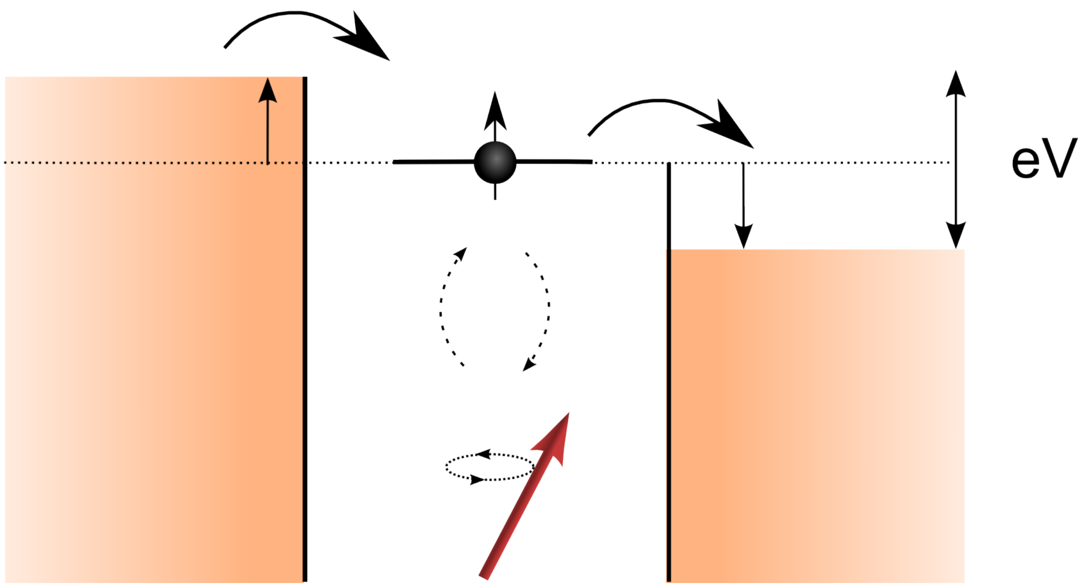
PRL 108, 057204 (2012)

Hamiltonian

Lead Tunneling QD

$$\mathscr{H} = \sum_{\mathbf{k}\chi\sigma}(\varepsilon_{\mathbf{k}\chi\sigma} - \mu_\chi)c^\dagger_{\mathbf{k}\chi\sigma}c_{\mathbf{k}\chi\sigma} + \sum_{\mathbf{k}\chi\sigma} T_\chi \left(c^\dagger_{\mathbf{k}\chi\sigma}d_\sigma + H.c\right) + \sum_\sigma \varepsilon_\sigma d^\dagger_\sigma d_\sigma - g\mu_B \mathbf{S}\cdot\mathbf{B} - J\mathbf{s}\cdot\mathbf{S}.$$

Field Interaction



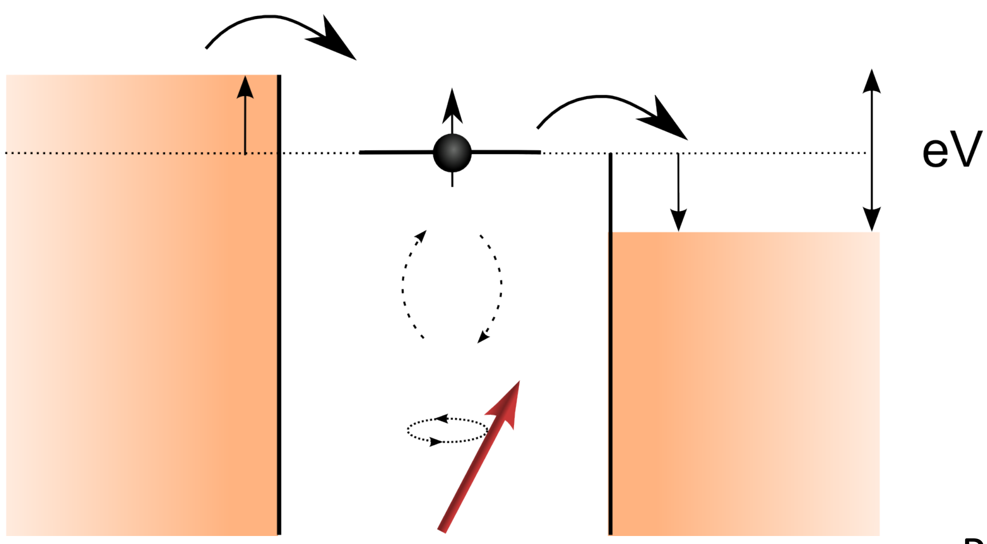
PRB 94 054311 (2016)

Simple system

- Green's function

H_0 H_M

$$\mathbf{G}(t,t') = g_0(t,t') - v \oint_C g_0(t,\tau) \langle \mathbf{S}(\tau) \rangle \cdot \boldsymbol{\sigma} g_0(\tau,t') d\tau.$$



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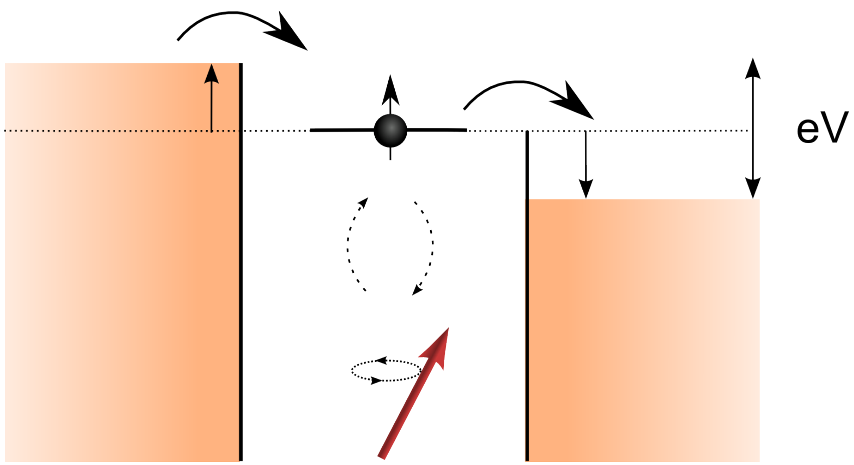
Time-dependent calculations

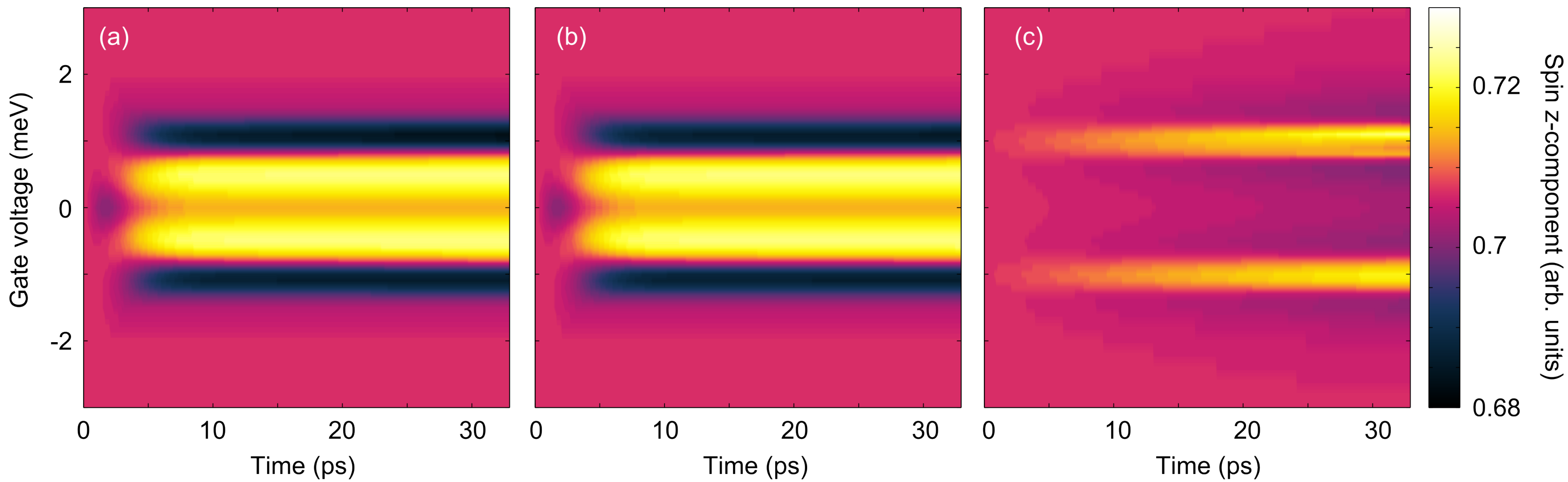
- Initial solution for spin, rotating in 45 degrees
- At time t=0: apply bias voltage and interactions



Spin evolution

$$\dot{\mathbf{S}}(t) = \mathbf{S}(t) \times \left(-g\mu_B \mathbf{B}^{eff}(t) + \frac{1}{e} \int \mathbb{J}(t,t') \cdot \mathbf{S}(t') dt' \right)$$

Full solution | Time-dependent parameters | Stationary parameters



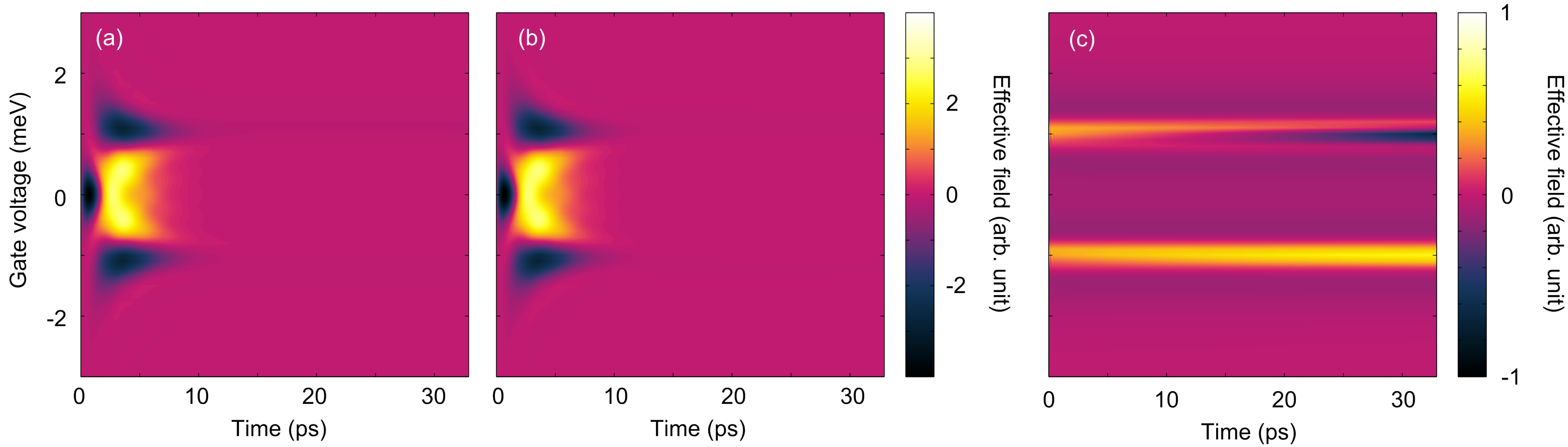
$$\int \mathbb{J}(t,t') \cdot \mathbf{S}(t') dt'$$

$$\int \mathbb{J}(t,t')(t-t') dt' \dot{\mathbf{S}}(t)$$

$$\hat{\mathbf{G}} \cdot \dot{\mathbf{S}}(t)$$

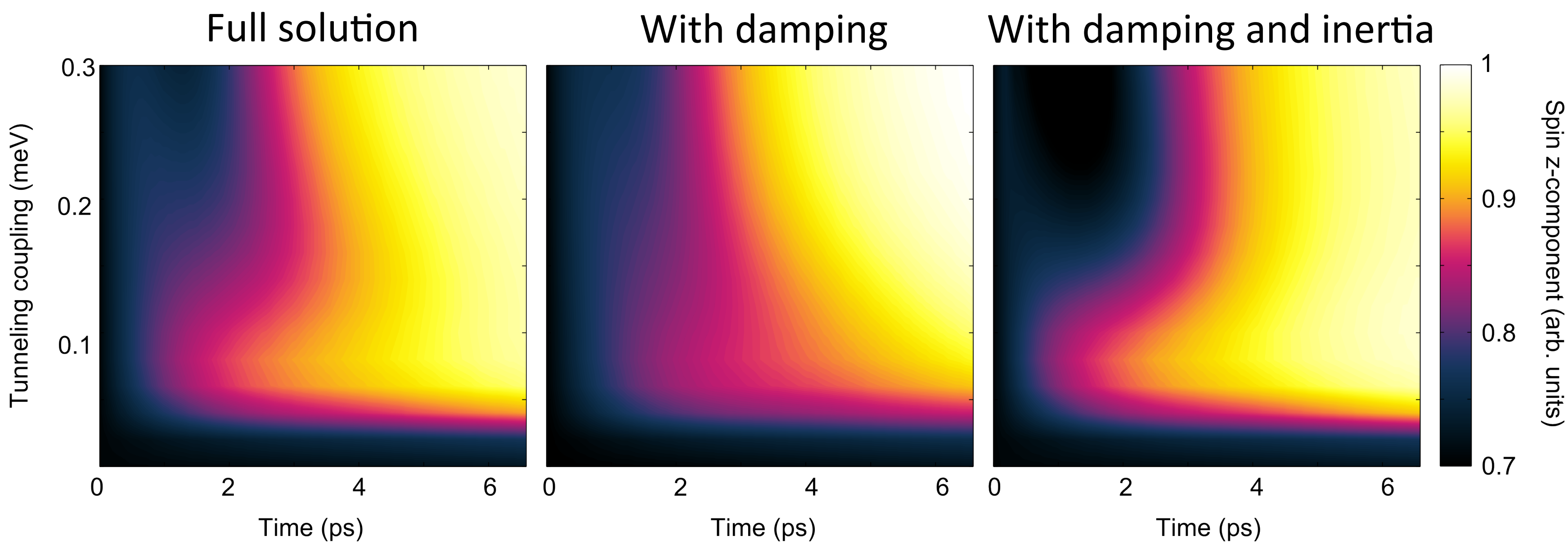
Effective field

Full solution Time-dependent parameters Stationary parameters



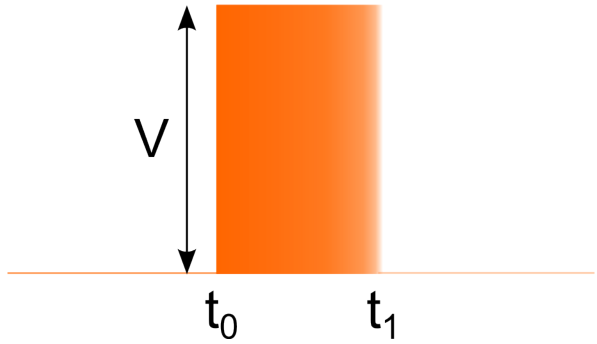
$$\dot{\mathbf{S}}(t) = \mathbf{S}(t) \times \left(-g\mu_B \mathbf{B}^{eff}(t) + \frac{1}{e} \int \mathbb{J}(t,t') \cdot \mathbf{S}(t') dt' \right)$$

Low coupling regime



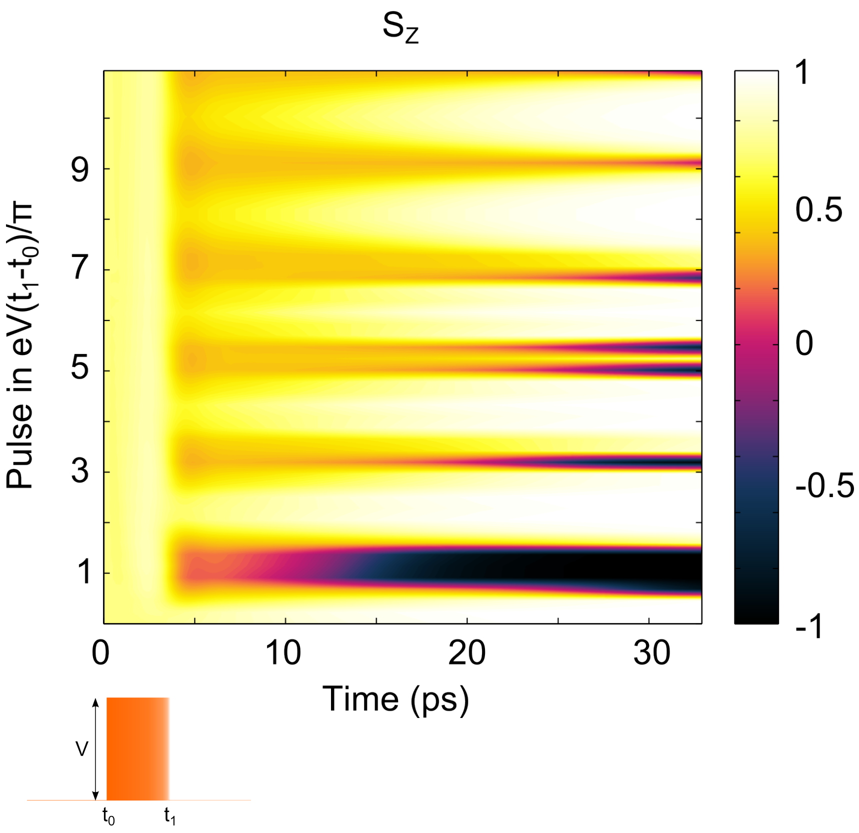
Pulse switching

- Calculate full solution
- At t_0, apply pulse
- At t_1, turn off



Phase-induced switching

- Memory effects / Non-Markovian
- Phase-induced switching
- $eV^*(t_1 - t_0)/\pi$ - periodicity



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Prospects

- Implications for atomistic spin dynamics
- Move towards quantum spin dynamics simulations
- Extension of the model

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Summary

- Compared field theoretical derivation of spin equation of motion with LLG equation

- Time-dependent LLG-parameters important in spin dynamics, time-independent doesn't capture transient regime

- Phase-induced switching dynamics for voltage pulses

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Exchange interaction

- Spin equation of motion

$$\dot{\mathbf{S}}(t) = \mathbf{S}(t) \times \left(-g\mu_B \mathbf{B}^{eff}(t) + \frac{1}{e} \int \mathbb{J}(t,t') \cdot \mathbf{S}(t') dt' \right)$$

- Decompose

$$\begin{aligned} \mathbf{S}(t) \times \mathbb{J}(t,t') \cdot \mathbf{S}(t') = & J_H(t,t') \mathbf{S}(t) \times \mathbf{S}(t') && \text{Heisenberg} \\ & + \mathbf{S}(t) \times \mathbb{I}(t,t') \cdot \mathbf{S}(t') && \text{Ising} \\ & - \mathbf{S}(t) \times \mathbf{D}(t,t') \times \mathbf{S}(t') && \text{Dzyaloshinski-Moriya} \end{aligned}$$

Effective Hamiltonian

$$\mathscr{H} = \sum_{mn} \mathbf{S}_m \cdot \left(J^H_{mn} \mathbf{S}_n + \mathbb{I}_{mn} \cdot \mathbf{S}_n + \mathbf{D}_{mn} \times \mathbf{S}_n \right)$$

m, n – indices for different spins and different times

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Exchange interaction

Isotropic Heisenberg

$$J_H(t,t') = iev^2\theta(t-t')\big(G_0^<(t',t)G_0^>(t,t') - G_0^>(t',t)G_0^<(t,t') - \mathbf{G}_1^<(t',t)\cdot\mathbf{G}_1^>(t,t') + \mathbf{G}_1^>(t',t)\cdot\mathbf{G}_1^<(t,t')\big),$$

Anisotropic Ising

$$\mathbb{I}(t,t') = iev^2\theta(t-t')\Big(\mathbf{G}_1^<(t',t)\mathbf{G}_1^>(t,t') - \mathbf{G}_1^>(t',t)\mathbf{G}_1^<(t,t') + \big[\mathbf{G}_1^<(t',t)\mathbf{G}_1^>(t,t') - \mathbf{G}_1^>(t',t)\mathbf{G}_1^<(t,t')\big]^t\Big),$$

Anisotropic Dzyaloshinski-Moriya

$$\mathbf{D}(t,t') = -ev^2\theta(t-t')\big(G_0^<(t',t)\mathbf{G}_1^>(t,t') - G_0^>(t',t)\mathbf{G}_1^<(t,t') - \mathbf{G}_1^<(t',t)G_0^>(t,t') + \mathbf{G}_1^>(t',t)G_0^<(t,t')\big).$$

$$\mathbf{G}^{</>}(t,t') = G_0^{</>}(t,t')\sigma^0 + \boldsymbol{\sigma}\cdot\mathbf{G}_1^{</>}(t,t')$$

Exchange interaction

- Stationary Dzyaloshinski-Moriya

$$\mathbf{D} = \frac{2v^2}{\pi\Gamma^2} \sum_{\chi\chi'} \Gamma^{\chi}\Gamma^{\chi'} \int (f_{\chi}(\omega) - f_{\chi'}(\omega))$$

$$\mathrm{Im} G_0^r(\omega) \mathrm{Im} \mathbf{G}_1^r(\omega) d\omega$$

$$\hat{\mathbf{G}}(\mathbf{D}) = \frac{-2v^2}{\pi\Gamma^2} \sum_{\chi\chi'} \Gamma^{\chi}\Gamma^{\chi'} \int (f'_{\chi}(\omega) + f'_{\chi'}(\omega))$$

$$\mathrm{Im} G_0^r(\omega) \mathrm{Re} \mathbf{G}_1^r(\omega) d\omega$$

Spin action

- Consider the spin action.

$$\mathcal{S} = \mathcal{S}_{WZWN} + \mathcal{S}_Z + \mathcal{S}_{int}$$

- Interaction term:

$$\mathcal{S}_{int} = \frac{1}{e} \int (\boldsymbol{\epsilon}\mathbf{j}(t,t') + \mathbf{S}(t) \cdot \mathbb{J}(t,t')) \cdot \mathbf{S}(t') dt dt'$$

PRL 92, 107001 (2004)
PRL 108, 057204 (2012)
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Spin action

$$\mathcal{S}_{WZWN} = \frac{1}{S^2}\int \mathbf{S}^q(t)\cdot[\mathbf{S}^c(t)\times\dot{\mathbf{S}}^{\mathbf{c}}(t)]dt$$

$$\mathcal{S}_Z = g\mu_B\int \mathbf{B}(t)\cdot\mathbf{S}^q(t)dt$$

$$\mathcal{S}_{int} = \frac{1}{e}\int (\epsilon\mathbf{j}(t,t') + \mathbf{S}^q(t)\cdot\mathbf{J}(t,t'))\cdot\mathbf{S}^c(t')dtdt'$$

PRL 92, 107001 (2004)
PRL 108, 057204 (2012)
PRL 113, 257201 (2014)

Exchange interaction

- Decompose damping and inertia terms

$$\dot{\mathbf{S}} = \mathbf{S} \times \left(-\gamma \mathbf{B} + \hat{\mathbf{G}}\dot{\mathbf{S}} + \hat{\mathbf{I}}\ddot{\mathbf{S}}\right)$$

$$\mathbf{S} \times \hat{\mathbf{G}} \cdot \dot{\mathbf{S}} = \hat{G}(J_H)\mathbf{S} \times \dot{\mathbf{S}} + \mathbf{S} \times \hat{G}(\mathbb{I}) \cdot \dot{\mathbf{S}} - \mathbf{S} \times \hat{G}(\mathbf{D}) \times \dot{\mathbf{S}}$$

$$\mathbf{S} \times \hat{\mathbf{I}} \cdot \ddot{\mathbf{S}} = \hat{I}(J_H)\mathbf{S} \times \ddot{\mathbf{S}} + \mathbf{S} \times \hat{I}(\mathbb{I}) \cdot \ddot{\mathbf{S}} - \mathbf{S} \times \hat{I}(\mathbf{D}) \times \ddot{\mathbf{S}}$$

Heisenberg Ising Dzyaloshinski-Moriya

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Theory

- Observables - currents

$$I_L^C(t) = -e\partial_t \left\langle \sum_{\mathbf{k}\sigma} n_{\mathbf{k}\sigma} \right\rangle = iesp\partial_t \sum_{\mathbf{k}} \mathbf{G}_{\mathbf{k}}^{<}(t,t')$$

$$I_L^S(t) = -e\partial_t \left\langle \sum_{\mathbf{k}\sigma\sigma'} c_{\mathbf{k}\sigma}^{\dagger} \boldsymbol{\sigma}_{\sigma\sigma'} c_{\mathbf{k}\sigma'} \right\rangle = iesp\boldsymbol{\sigma}\partial_t \sum_{\mathbf{k}} \mathbf{G}_{\mathbf{k}}^{<}(t,t')$$

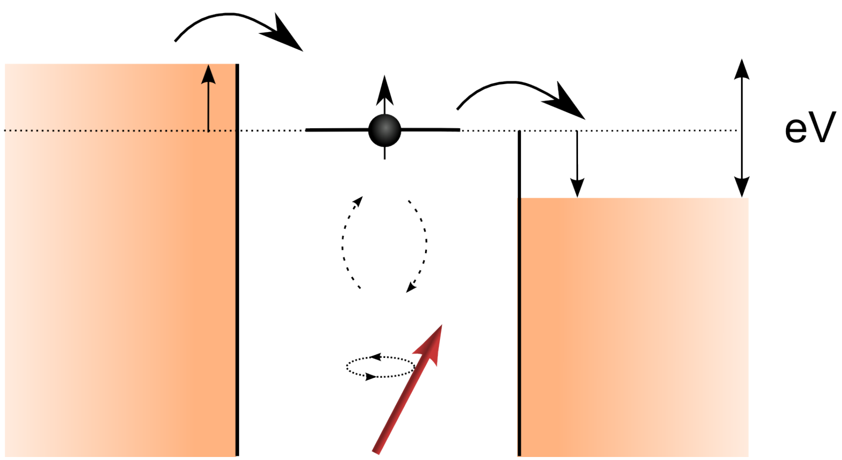
Theory

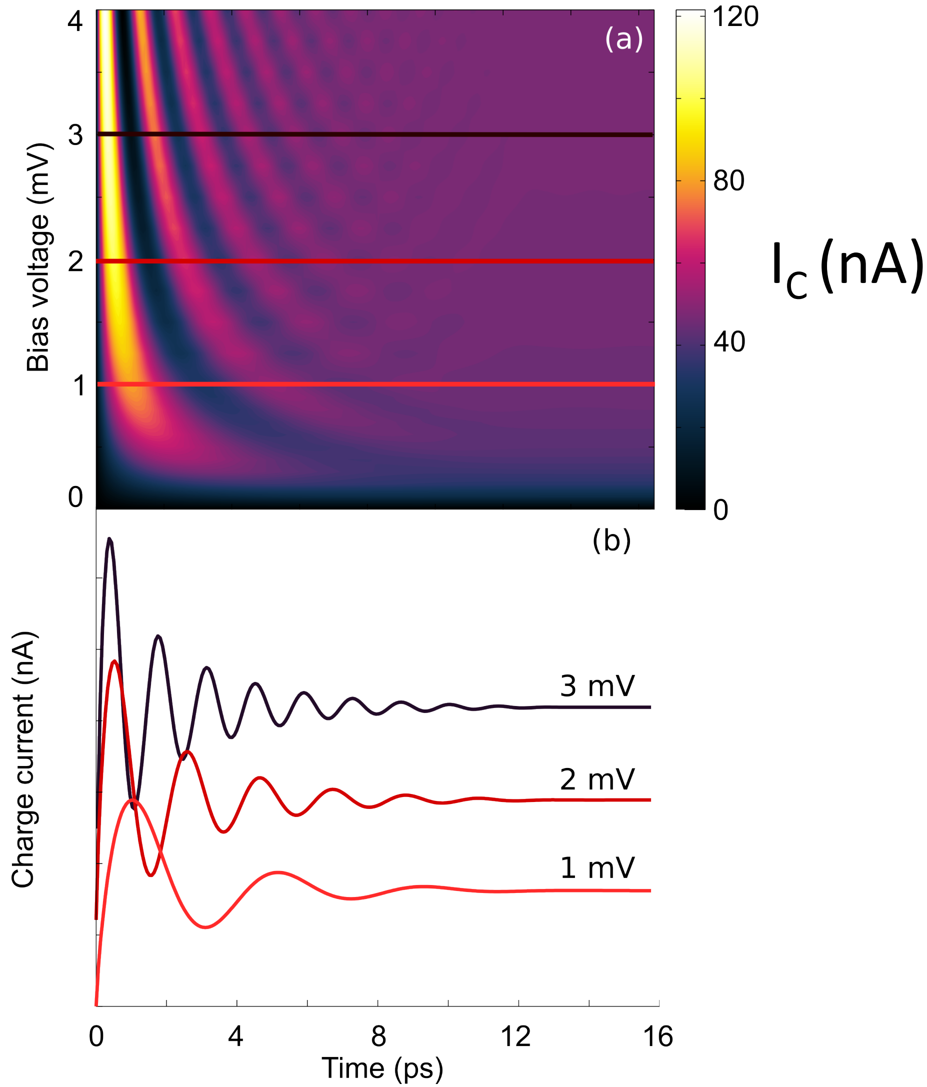
- QD Green's function

$$\mathbf{g}^{</>}(t,t') = \int \mathbf{g}^{r}(t,\tau)\mathbf{\Sigma}^{</>}(\tau,\tau')\mathbf{g}^{a}(\tau',t')d\tau d\tau'$$

$$g_{\sigma}^{r/a}(t,t') = (\pm i)\theta(\pm t \mp t')e^{-i(\varepsilon_{\sigma} \mp i\Gamma_{\sigma})(t-t')}$$

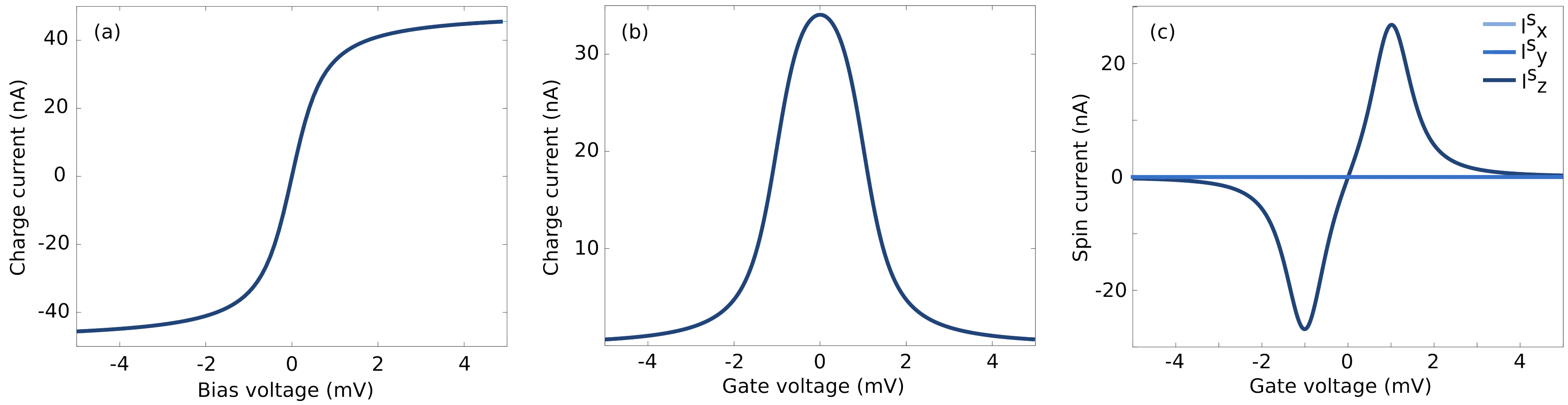
Time evolution of the charge current

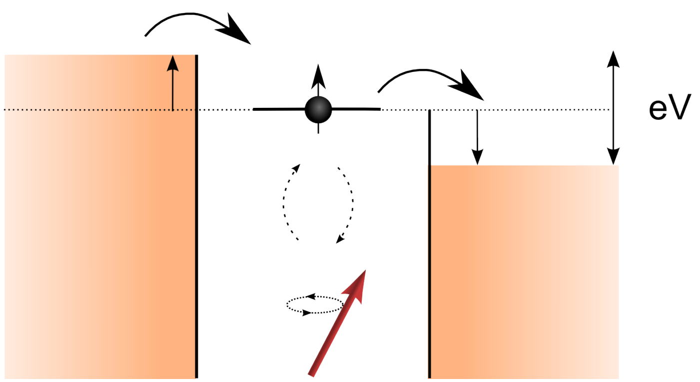




Simple system

$$\mathbf{G}_1 = -v g_0 g_0 S_z \hat{\mathbf{z}}$$





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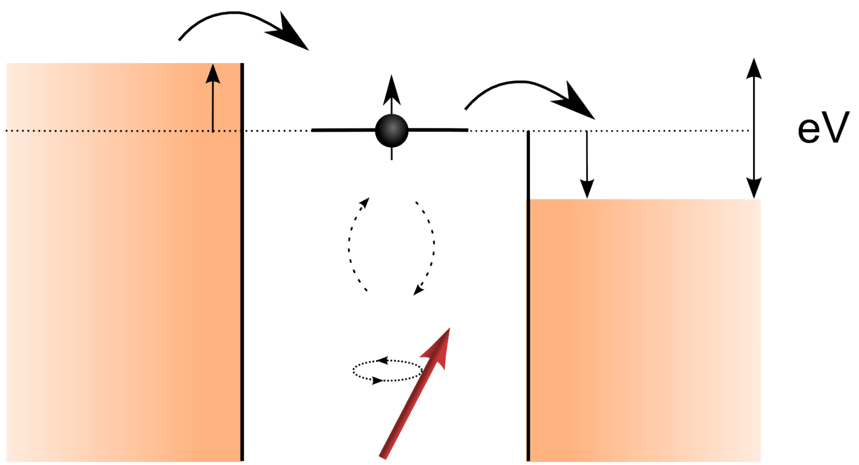
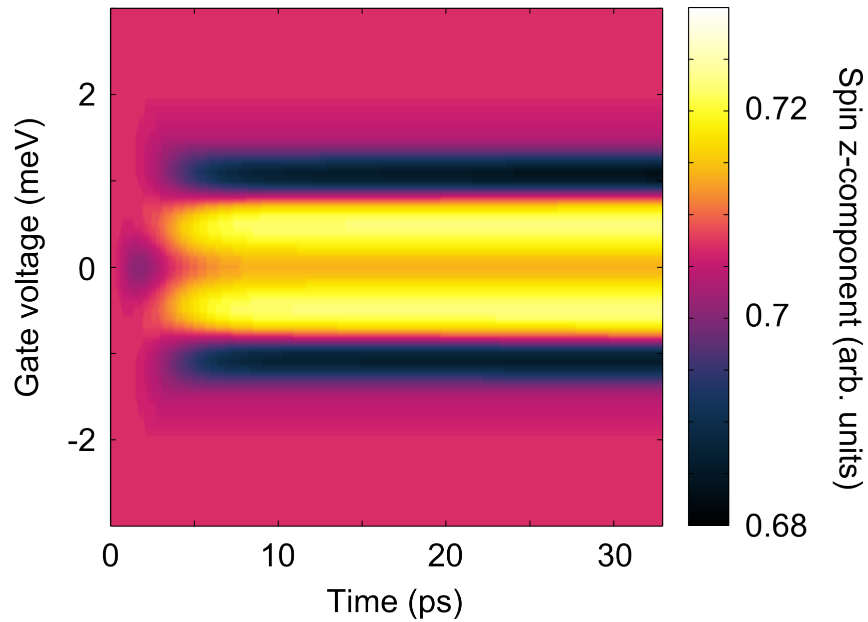
Spin evolution

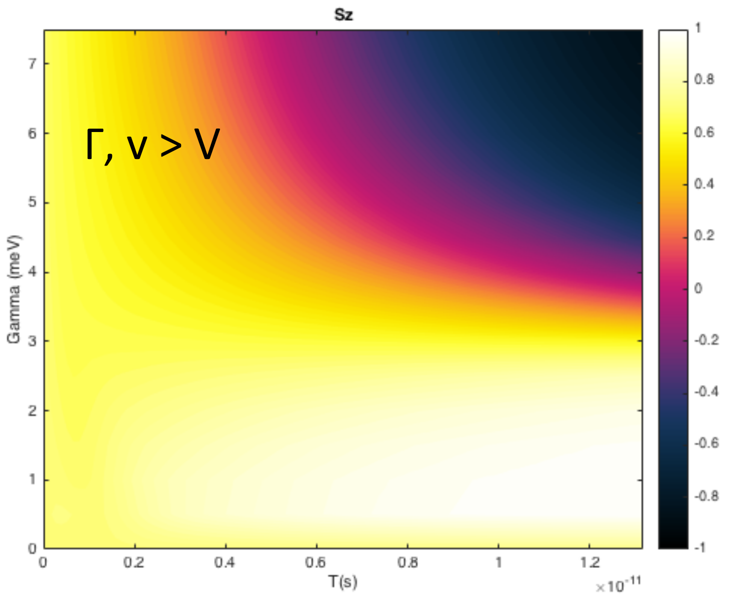




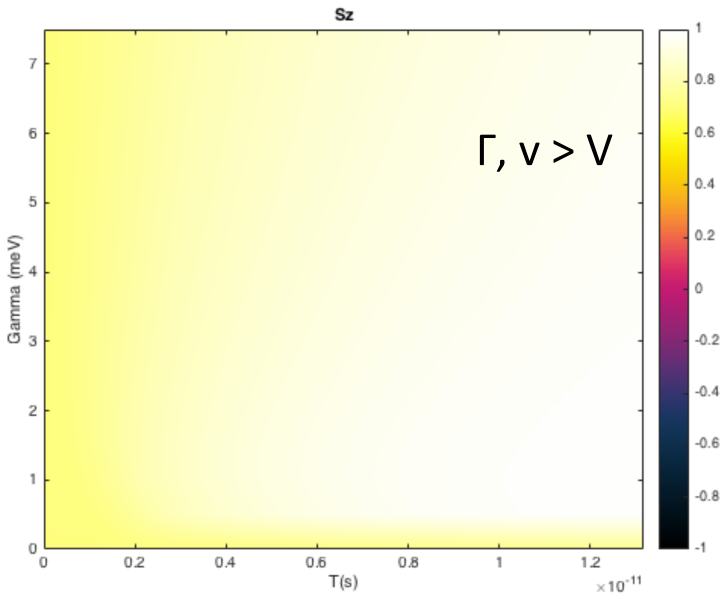
$$\dot{\mathbf{S}}(t) = \mathbf{S}(t) \times \left(-g\mu_B \mathbf{B}^{eff}(t) + \frac{1}{e} \int \mathbb{J}(t,t') \cdot \mathbf{S}(t') dt' \right)$$

Strong coupling regime

Spin evolution - Full solution



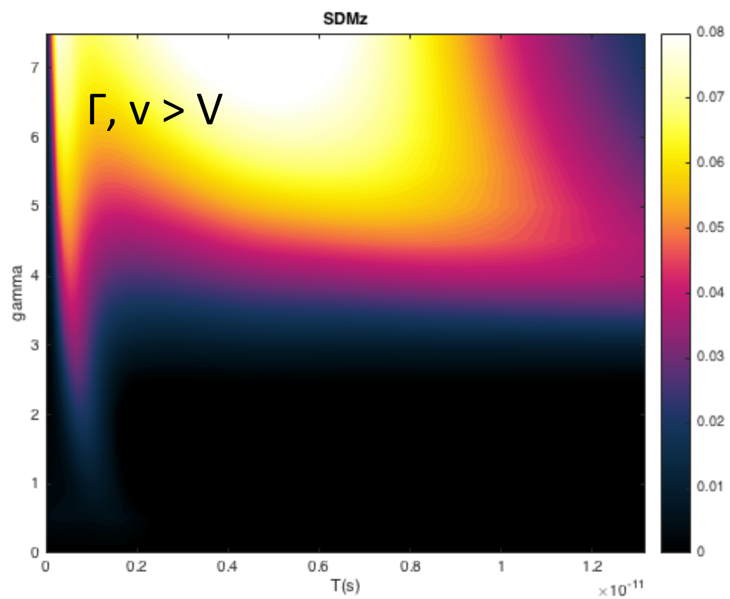
Spin evolution - Time-dependent damping



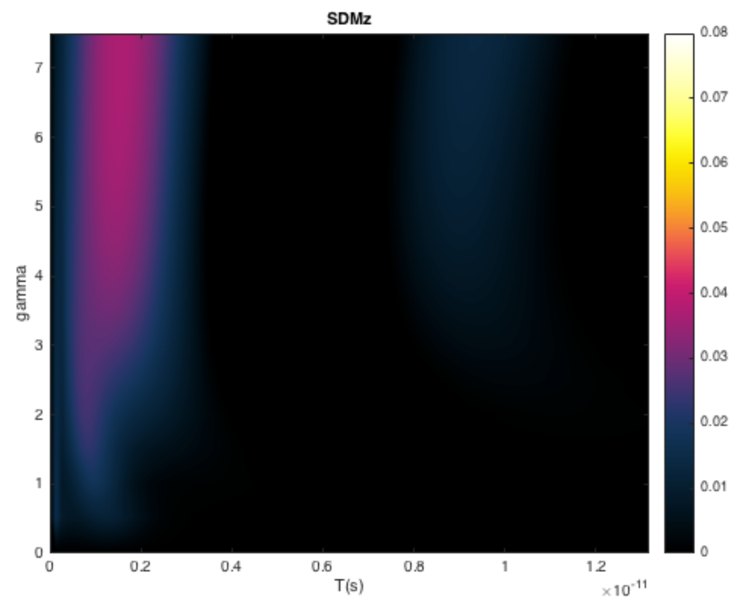
Time-dependent solution breaks down in high coupling regime

Strong coupling regime

DM interaction - Full solution



DM interaction - Time-dependent damping



$$
\begin{aligned}
\mathbf{S}(t) \times \mathbb{J}(t,t') \cdot \mathbf{S}(t') = & J_H(t,t')\mathbf{S}(t) \times \mathbf{S}(t') \\
& + \mathbf{S}(t) \times \mathbb{I}(t,t') \cdot \mathbf{S}(t') \\
& \boxed{- \mathbf{S}(t) \times \mathbf{D}(t,t') \times \mathbf{S}(t')}
\end{aligned}
$$