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First-principles approach to ultrafast logic functionalization of magnetic molecules

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Outline

I. History: theory of spin dynamics in extended systems

II. Introduction: theoretical and background aspects

III. Three magnetic centers: magnetic logic

IV. Four magnetic centers: spin SHIFT register & which-path interference

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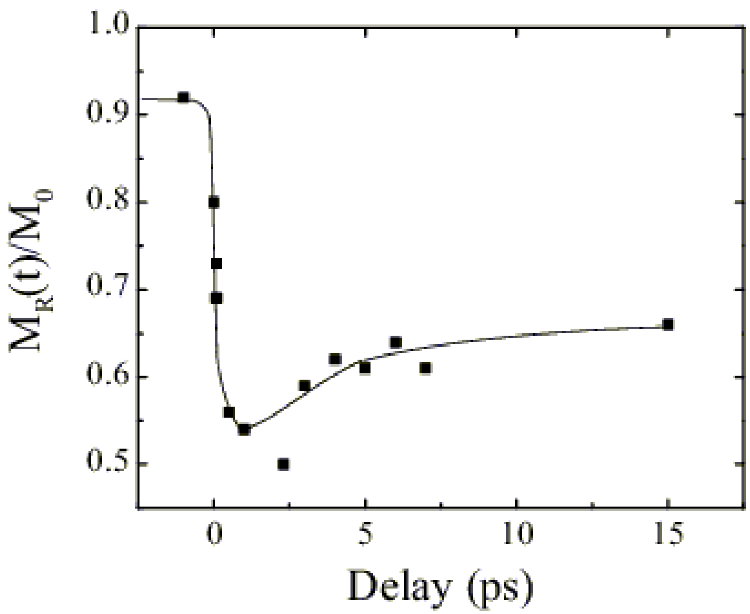
I. History: theory of spin dynamics in extended systems

II. Introduction: theoretical and background aspects

III. Three magnetic centers: magnetic logic

IV. Four magnetic centers: spin SHIFT register & which-path interference

I. History: pump-probe (magneto-)optics



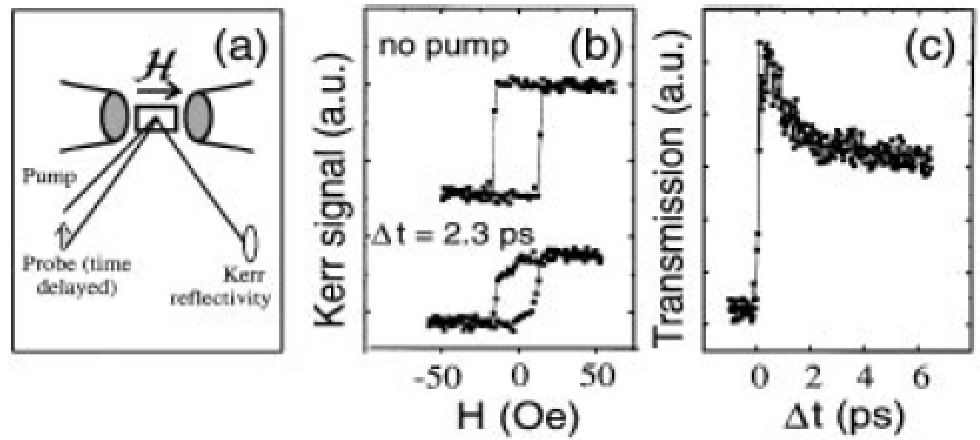
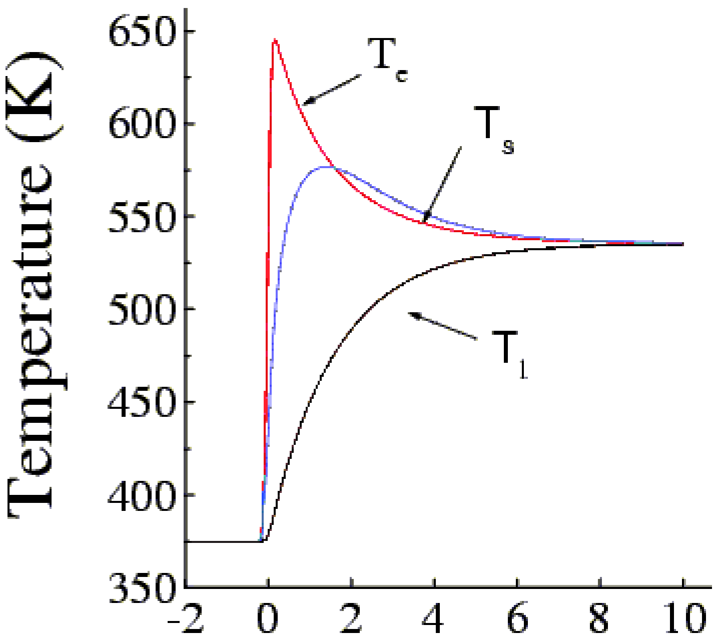


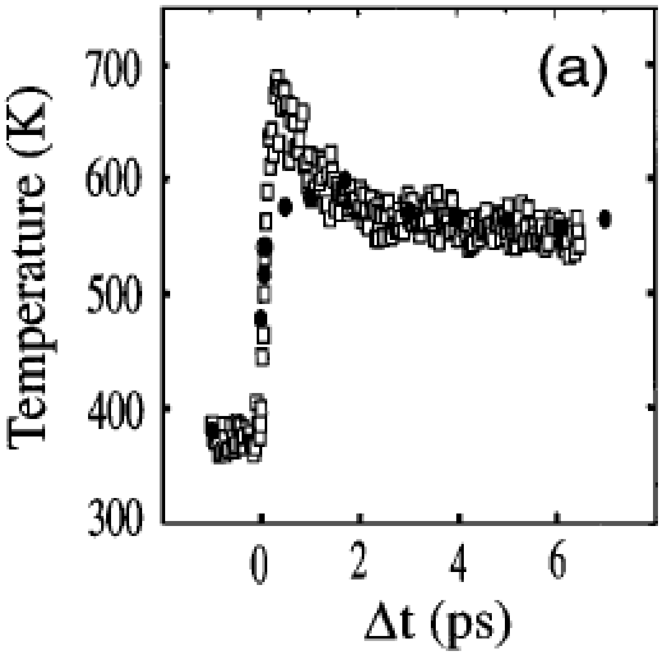
FIG. 1. (a) Experimental pump-probe setup allowing dynamic longitudinal Kerr effect and transient transmissivity or reflectivity measurements. (b) Typical Kerr loops obtained on a 22 nm thick Ni sample in the absence of pump beam and for a delay $\Delta t = 2.3$ ps between the pump and probe pulses. The pump fluence is 7 mJ cm^{-2}. (c) Transient transmissivity [same experimental condition as (b)].

- Reflectivity
- MOKE
- $\tau_{spin} < 1$ ps

E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Phys. Rev. Lett. **76**, 4250 (1996)

I. History: 3-Temperature model





$$C_e(T_e)dT_e/dt = -G_{el}(T_e - T_l) - G_{es}(T_e - T_s) + P(t),$$

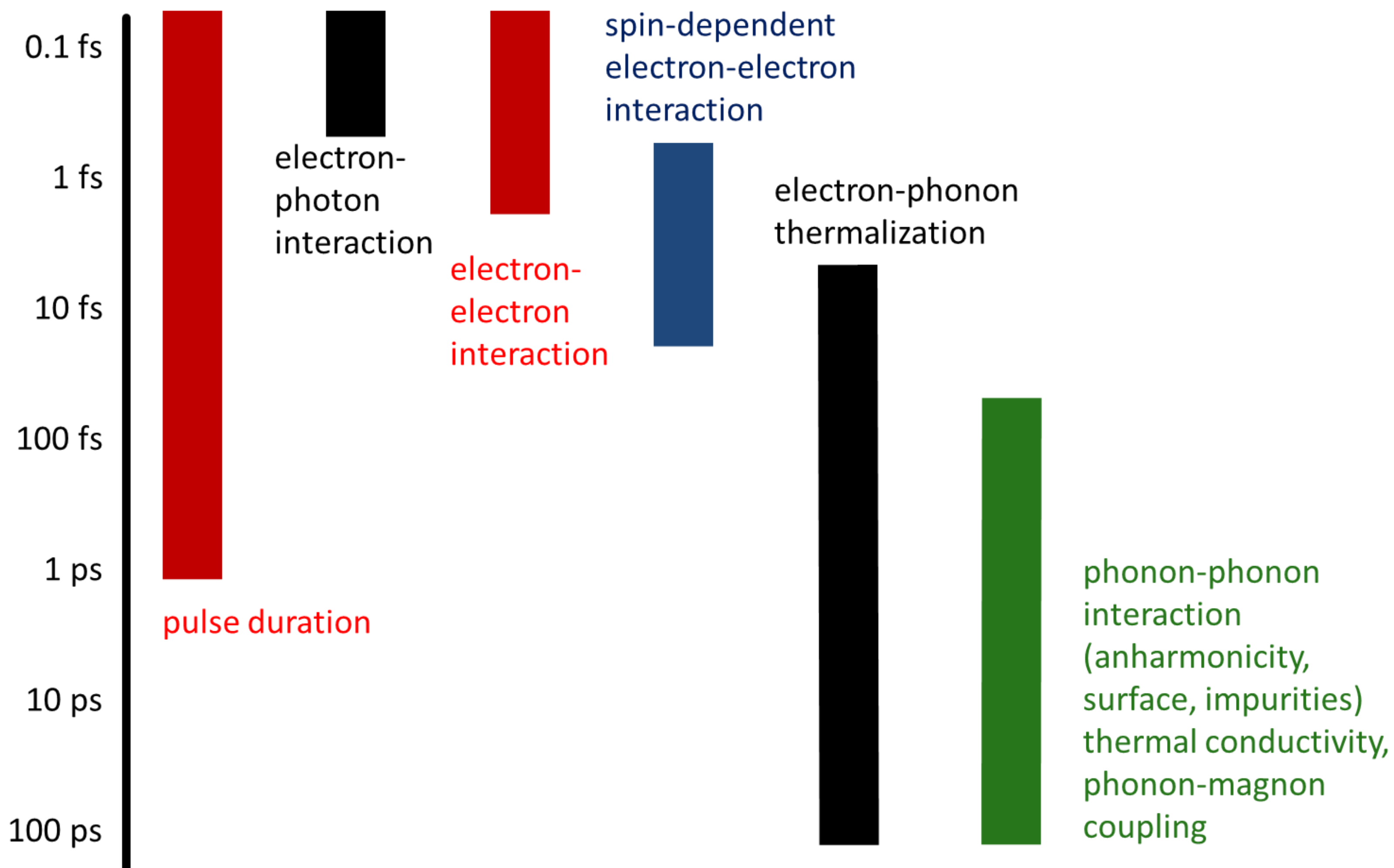
$$C_s(T_s)dT_s/dt = -G_{es}(T_s - T_e) - G_{sl}(T_s - T_l)$$

$$C_l(T_l)dT_l/dt = -G_{el}(T_l - T_e) - G_{sl}(T_l - T_s)$$

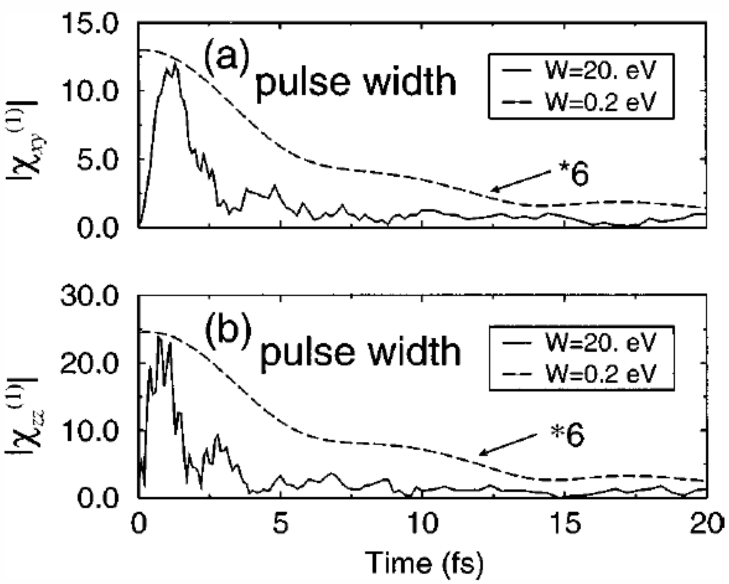
- Good agreement with experiment
- Uniform temperature profile

E. Beaurepaire, J.-C. Merle, A. Daunois, and J.-Y. Bigot, Phys. Rev. Lett. **76**, 4250 (1996)

I. History: Relevant time scales for the laser control of magnetism



I. History: theoretical achievements
Spectral width → bleaching in Ni



Wide pulse (in frequency domain) populates target states
→ transition paths blocked
→ bleaching

Affects both charge and spin dynamics

G. P. Zhang and W. Hübner, Phys. Rev. B **58**, R5920 (1998)

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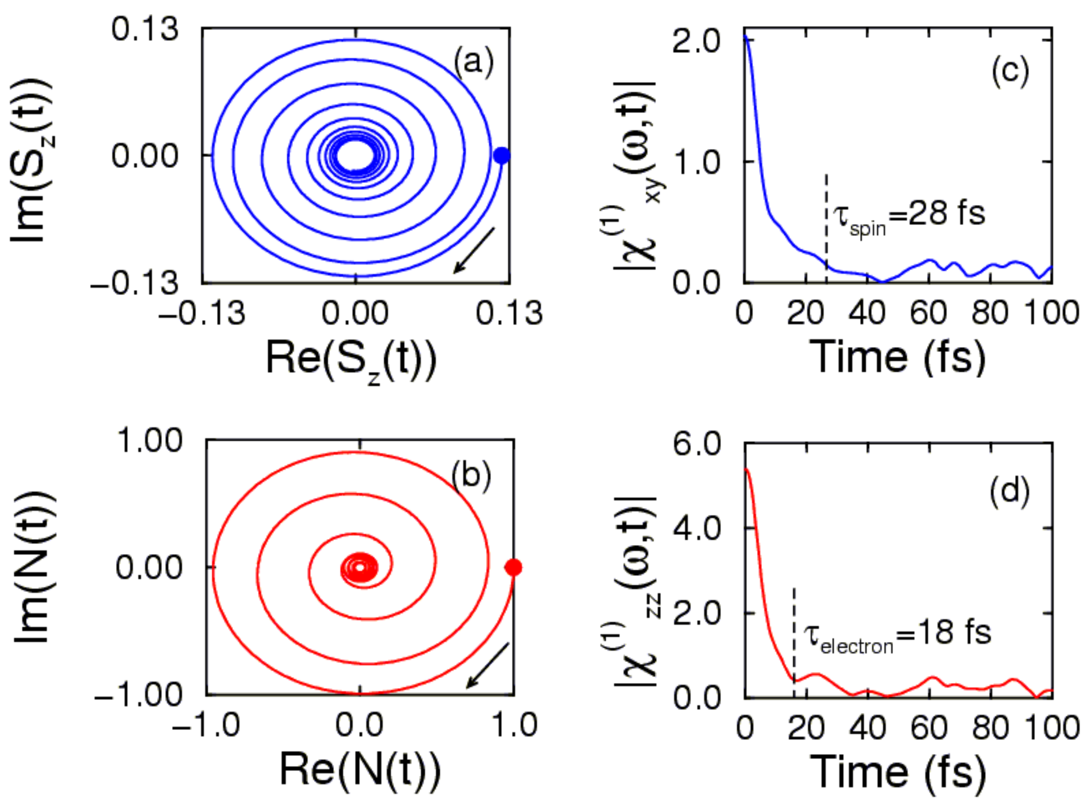
I. History: Theoretical achievements
Coherent dephasing intrinsic vs extrinsic quantities

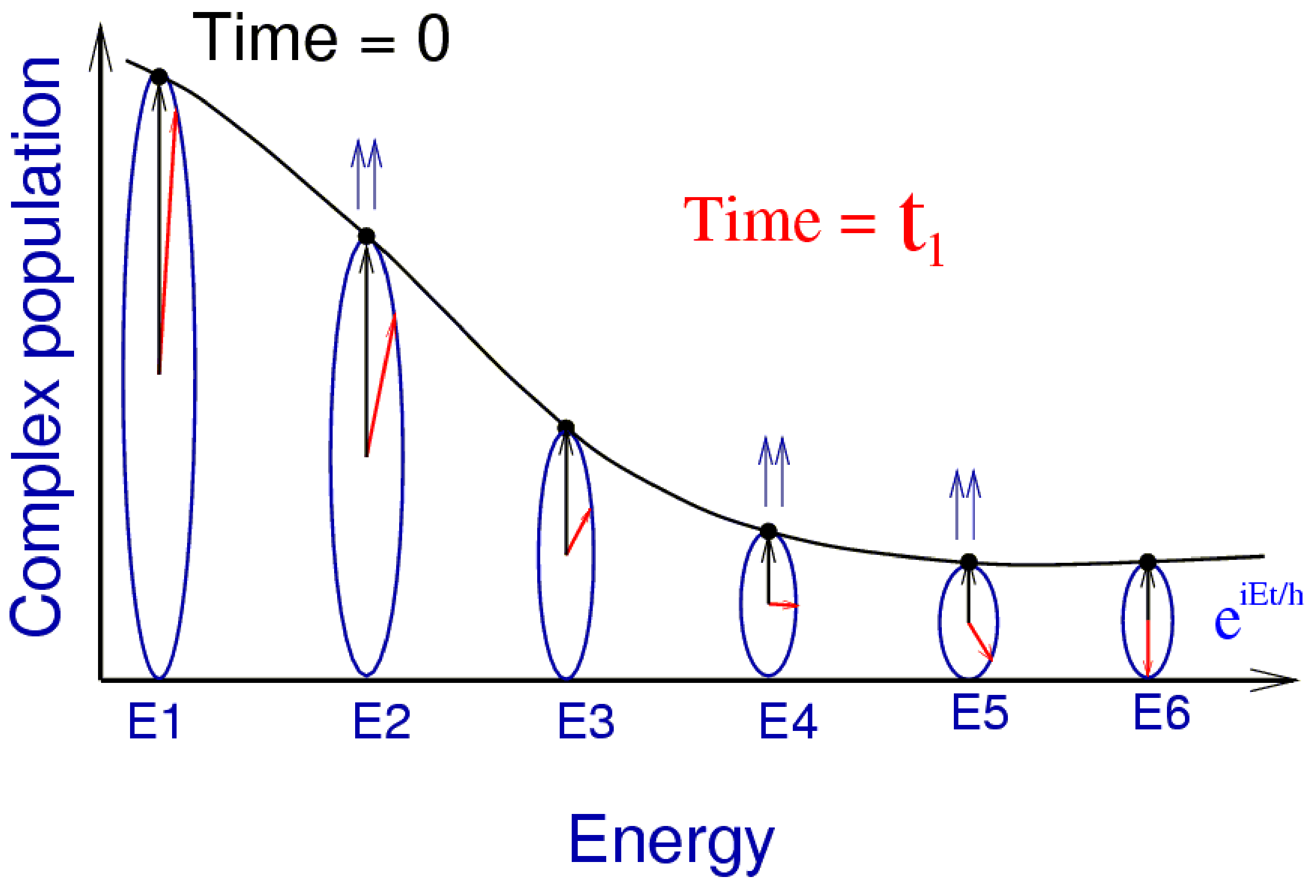


Ni

- Charge dynamics preceeds spin dynamics -> spin memory time
- Spin dynamics due to dephasing of states
- Fast decay results from loss of coherence
- Increased exchange interaction speeds up spin (rather than charge) dynamics
- ~10 fsec

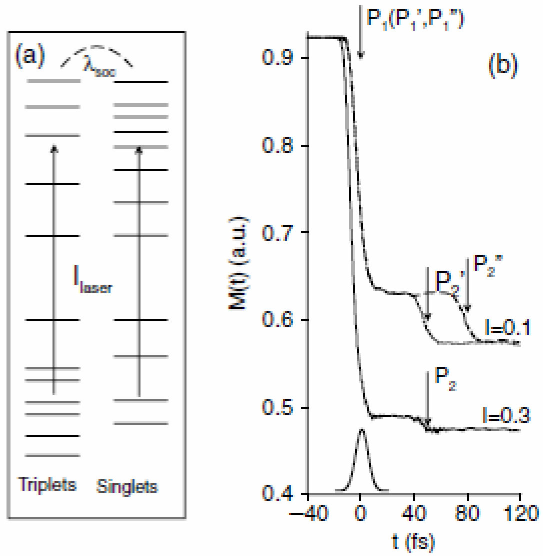
W. Hübner and G.P. Zhang, J. Magn. Magn. Mater. **189**, 101 (1998)
G. P. Zhang and W. Hübner, Appl. Phys. B **68**, 495 (1999)

I. History: Dephasing of the excited state



G. Zhang, W. Hübner, E. Beaurepaire, and J.-Y. Bigot (2001*). Laser-induced ultrafast demagnetization: femtomagnetism, a new frontier?* In K. Ounadjela and B. Hillebrands (Eds.), Spin Dynamics in Confined Magnetic Structures, Volume 83 of Topics Appl. Phys., pp. 245–289.

I. History: Theoretical achievements
Population dynamics → magnetization dynamics in FM



Time-dependent problem

$$i\hbar \frac{\partial}{\partial t}|\Psi(t)\rangle = H|\Psi(t)\rangle$$

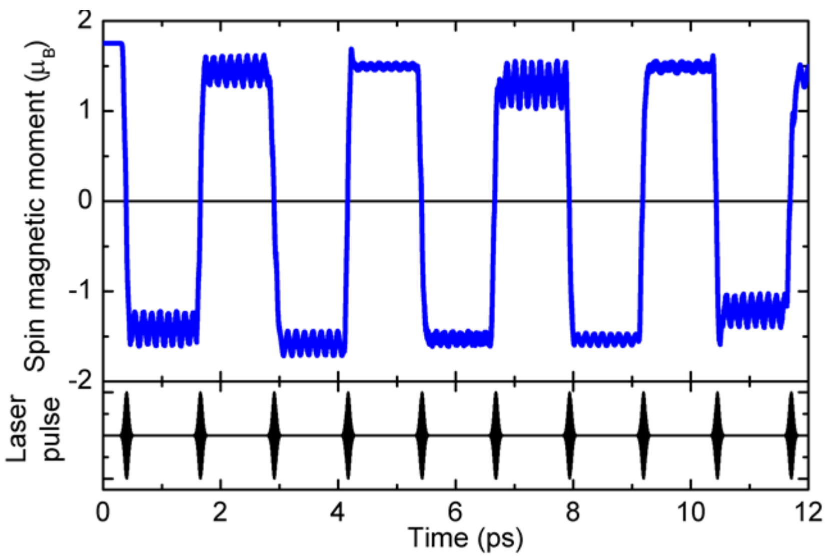
Ni

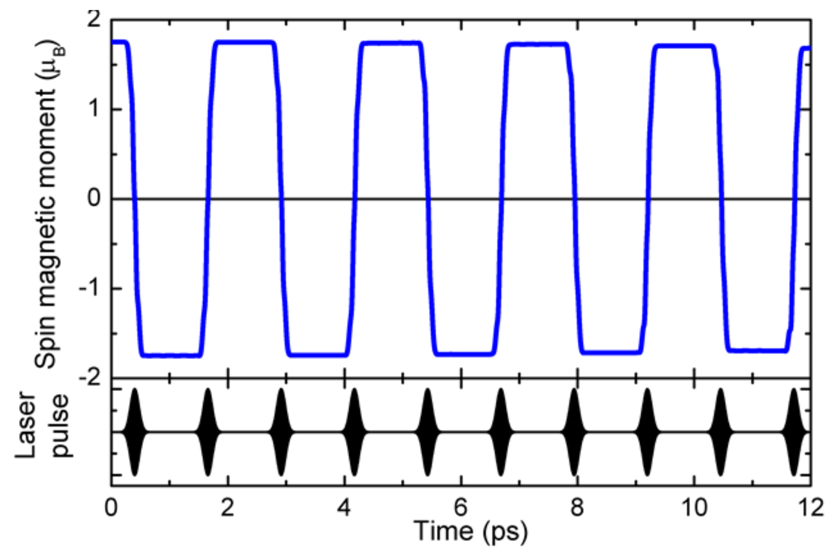
- Cooperative effect of laser pulse and SOC
- Controllable process!
- τ_1 ~ 40 fsec

G. P. Zhang and W. Hübner, Phys. Rev. Lett. **85**, 3025 (2000)

I. History: switching NiO (001)

- First results for NiO, showing the possibility of all optical spin switching in the subpicosecond regime
- Tuning photon energy, intensity and width of the laser pulse



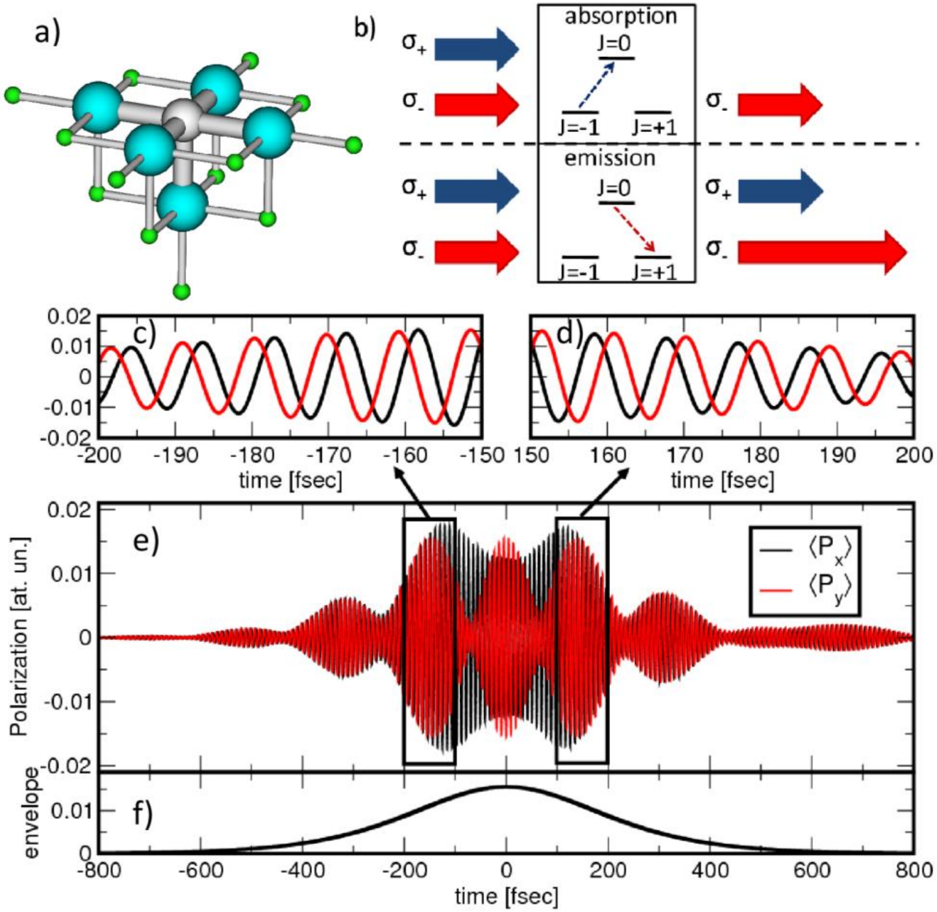


$\hbar\omega_0 = 0.422$ eV, l = 2933 nm
FWHM = 59 fs, $l_{\max} \approx 10^{14}$W/cm²

$\hbar\omega_0 = 1.645$ eV, l = 752 nm
FWHM = 117 fs, $l_{\max} \approx 1.2 \times 10^{14}$ W/cm²

R. Gómez-Abal, O. Ney, K. Satitkovitchai, and W. Hübner, Phys. Rev. Lett. **92**, 227402 (2004)

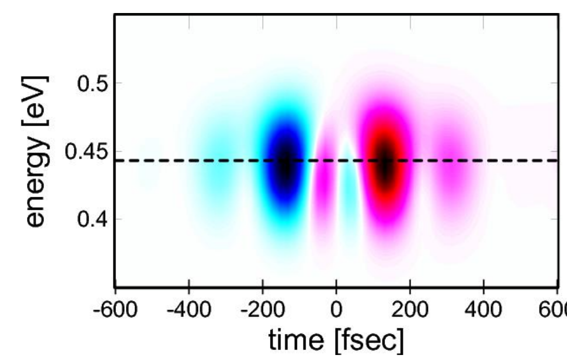
I. History: extended magnetic systems
What about angular momentum conservation?



conservation of angular momentum in spin-flip in antiferromagnetic **NiO**

induced material polarization $\langle \mathbf{P}(t) \rangle$

phase difference between $\langle P_x(t) \rangle$ and $\langle P_y(t) \rangle$
→ circularly polarized light



Stokes vector $\mathbf{S} = (I, Q, U, V)$

G. Lefkidis, G. P. Zhang, and W. Hübner, Phys. Rev. Lett. **103**, 217401 (2009)
G. Lefkidis and W. Hübner, Phys. Rev. B **87**, 014404 (2013)

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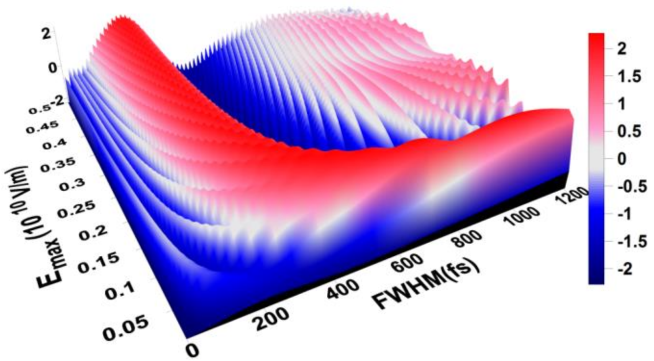
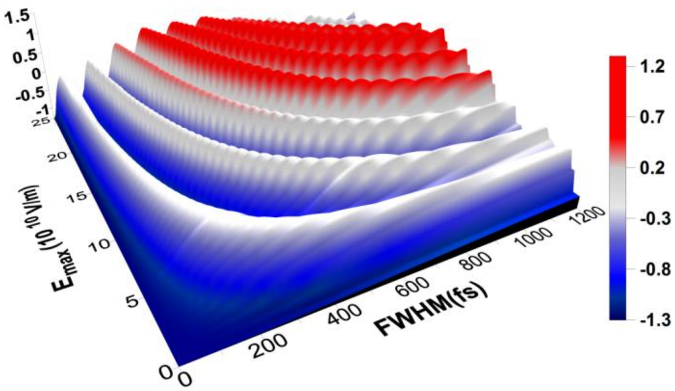
OPTIMAS light-spin-matter | SFB/TRR 88 3MET.de | SPIN+X SFB/TRR 173 Kaiserslautern • Mainz | TECHNISCHE UNIVERSITÄT KAISERSLAUTERN

I. History: Electron-phonon coupling in NiO

force matrix → normal modes → quantization → electron-phonon interaction



no phonons



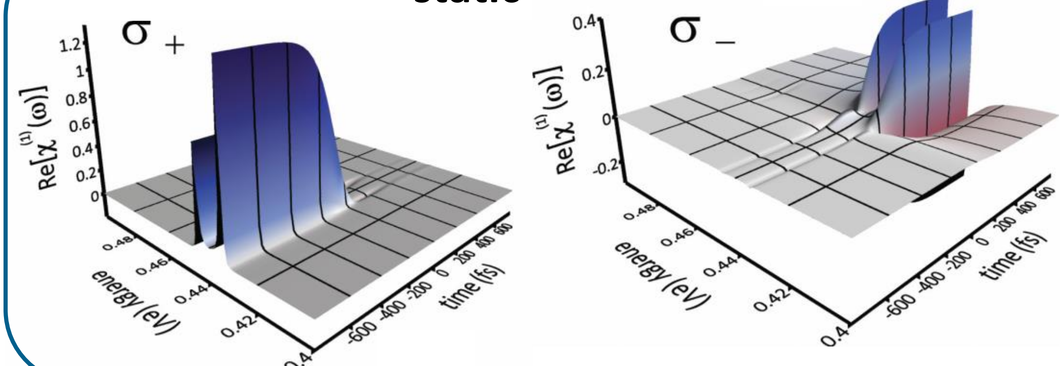
phonons

- phonons affect symmetry
  ⇒ different selection rules
- lattice temperature dependence

G. Lefkidis and W. Hübner, J. Mag. Mag. Mater. **321**, 979 (2009)

I. History: extended magnetic systems
What about the probe pulse?

static



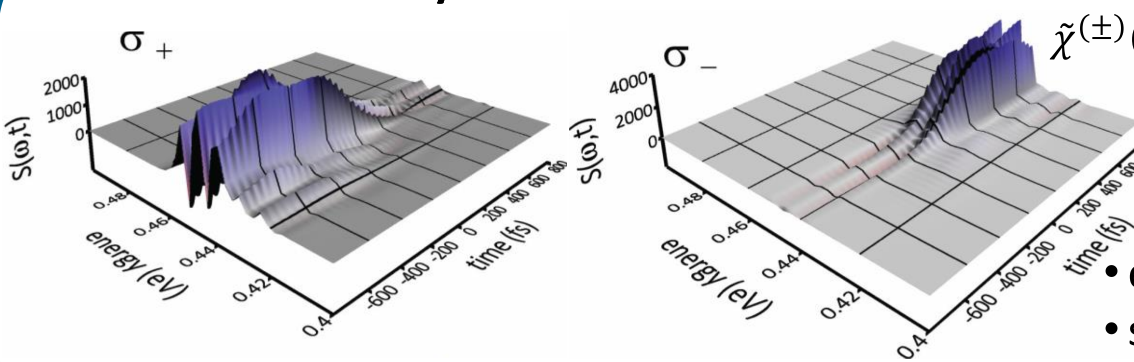
1-photon

occupations

$$\chi^{(1)}(\omega) \propto \sum_{a,b} D^*_{ab} D_{ba} \frac{n_a - n_b}{E_a - E_b + \hbar\omega + i\hbar\Gamma}$$

artificial broadening

dynamic



$$\tilde{\chi}^{(\pm)}(\omega, t) \propto \left. \frac{\int \langle \mathbf{P}(t') \rangle g(\sigma, t-t') e^{i\omega t'} dt'}{\int \mathbf{E}^{(\pm)}_{\mathrm{probe}}(t') g(\sigma, t-t') e^{i\omega t'} dt'} \right|_{\omega_1}$$

- exact ($\chi^{(\infty)}$ much better than $\chi^{(3)}$ only)
- small effects (new peaks, dynamic Stark effect, broadening)

G. Lefkidis and W. Hübner, Phys. Rev. B **87**, 014432 (2013)

I. History: theoretical achievements
Spin-lattice relaxation time $\tau_{SL}\approx$48 psec for Gd

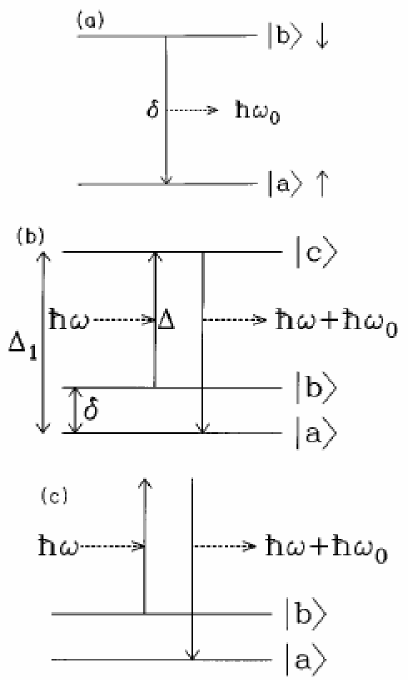


FIG. 1. (a) Direct process, (b) Orbach process, and (c) Raman process.

- Good agreement with experiment
- Time given by spin-orbit induced magnetocrystalline anisotropy energy
- Three phonon involving processes
  - Direct process (one-phonon scattering, very low T)
  - Orbach process (crystal-field splitting, low T)
  - Raman process (two-phonon scattering, moderate T)
- Phonon-magnon coupling
- 2-phonon processes → high-temperature theory (for low-temperature plateau)

rate equation

$$\dot{N}_b=\frac{9\sum_{mn}|\langle a|v_n^m|b\rangle|^2}{8\rho^2\pi^3\hbar^7 v_s^{10}}\int [N_a\bar{p}_0(\delta_2)[\bar{p}_0(\delta_1)+1]-N_b\bar{p}_0(\delta_1)[\bar{p}_0(\delta_2)+1]]\delta_1^6 d\delta_1$$

W. Hübner and K. H. Bennemann, Phys. Rev. B **53**, 3422 (1996)

I. History of theoretical achievements

a. Bleaching (<10 fsec)

b. Dephasing (10 fsec)

c. Population dynamics (40-80 fsec) with spin-orbit coupling

d. Electron-phonon coupling (<1 psec)

e. Spin-lattice relaxation (48 psec)

Outline

I. History: theory of spin dynamics in extended systems

II. Introduction: theoretical and background aspects

III. Three magnetic centers: magnetic logic

IV. Four magnetic centers: spin SHIFT register & which-path interference

II. Introduction: Which materials?

Extended systems (top-down approach, patterning, DFT)

- **Ferromagnets** → fast dynamics but no selective control possible
  (many broad bands i.e. no addressability of excited states)

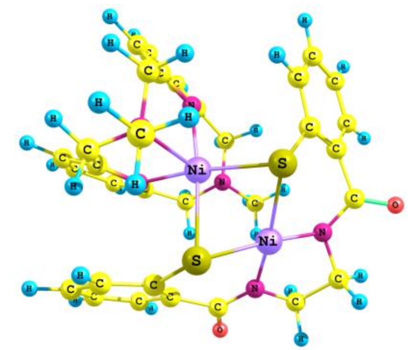
- **Antiferromagnets**
  - narrow bands → good addressability → switching
  - two magnetic centers
  - faster dynamics $\omega_{AFM} \propto \sqrt{H_{\text{eff}}\,(2H + H_{\text{eff}})}$

Localized systems (bottom-up approach, self-organization, quantum chemistry)

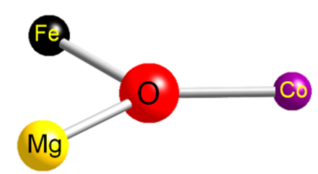
- **Magnetic molecules**
  - several distinguishable active centers
  - few discrete levels
  - even better addressability
  - spin localization → magnetic logic
  - however: difficulty of environment

II. Introduction: Which materials?

<u>Molecular magnets: four different experimental environments</u>



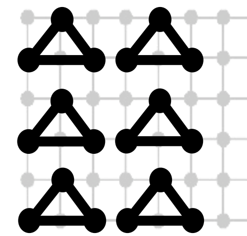
1. ligand-stabilized complexes (fluid phase/pellets)
   (+) conventional wet chemistry
   (+) exist already
   (-) far from device applications



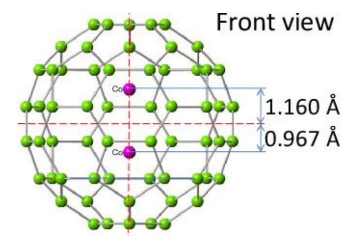
2. Gas phase of bare clusters (nozzle expansion)
   (+) few atomes
   (+) larger active-center/total-atoms ratio
   (+) charged particles → control through mass-selection
   (-) far from device applications

II. Introduction: Which materials?

Molecular magnets: four different experimental environments



3. Clusters on surfaces
   (+) close to application devices
   (-) exploit of additional features needed for selectivity
   (resonance selection/magnetic field gradient)
   (-) bottom-up preparation: good but not excellent structures, e.g. on (111) surfaces, magnetically fair



4. Endohedral fullerenes
   (+) synthesizable
   (+) inner cluster magnetically protected (e.g. from a surface)
   (+) formation of ordered superstructures on surfaces possible
   (-) orientation of cluster not always clear

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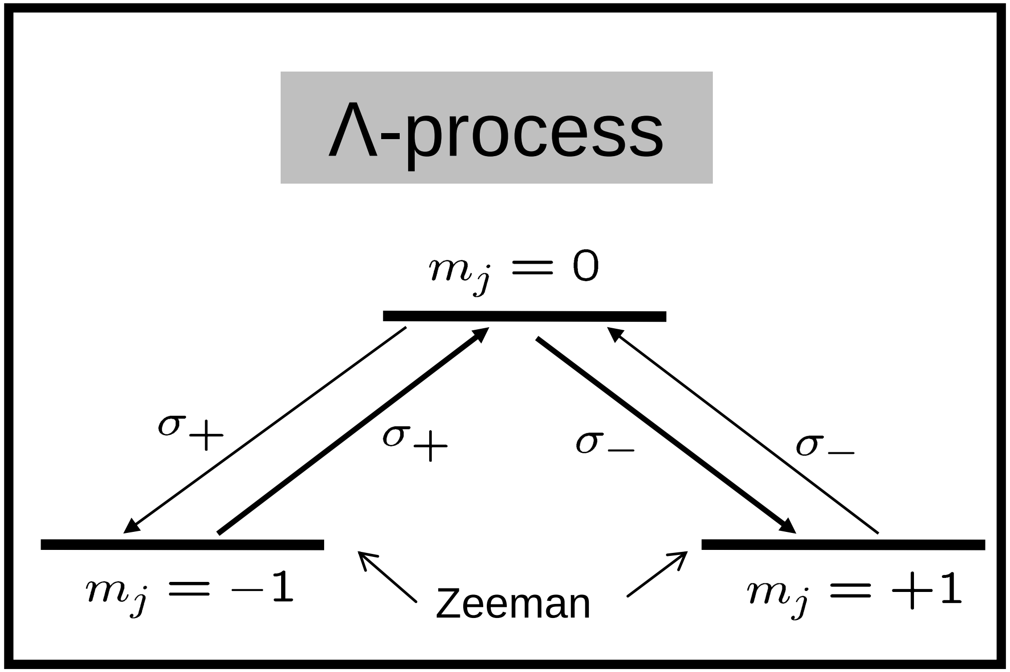
II. Introduction: Λ process



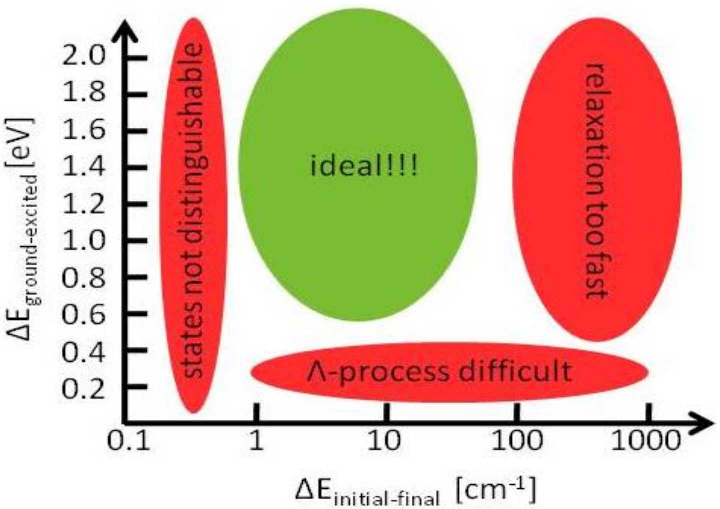
in silico: time minimization (< 1 psec)

propagation process $|A\rangle \xrightarrow{\hbar\omega} |B\rangle$

the spins of the states can have **different**

- magnitude → demagnetization
- orientation → spin flip
- localization → spin transfer

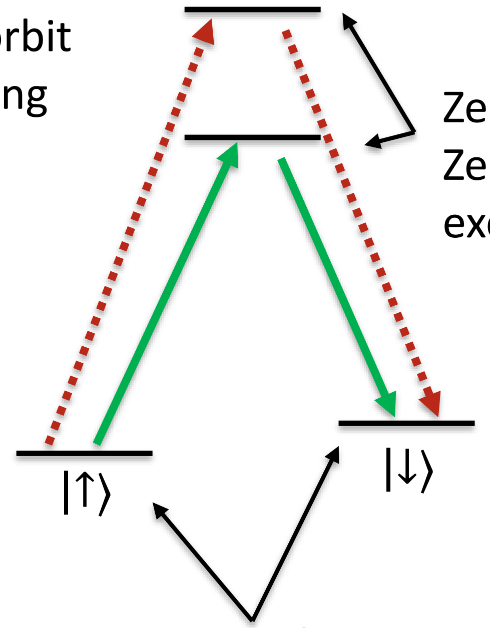
spin-orbit coupling AND laser →
spin dynamics AND functionalization !



Energy differences

II. Introduction: Anisotropy vs. thermalized population

spin-orbit coupling



Zero-field-splitting and Zeeman splitting of excited states

Zeeman splitting of ground states

The process depends on the population distribution of all states (not only the ground states)
→ selection rules can overcome thermal distribution problems!

- start from broken-spinsymmetry ground state after proper laser pulse
- start from metastable excited state
- external magnetic field + SOC
- rely on laser polarization (dichroism)

Magnetic preferential axis can be induced with

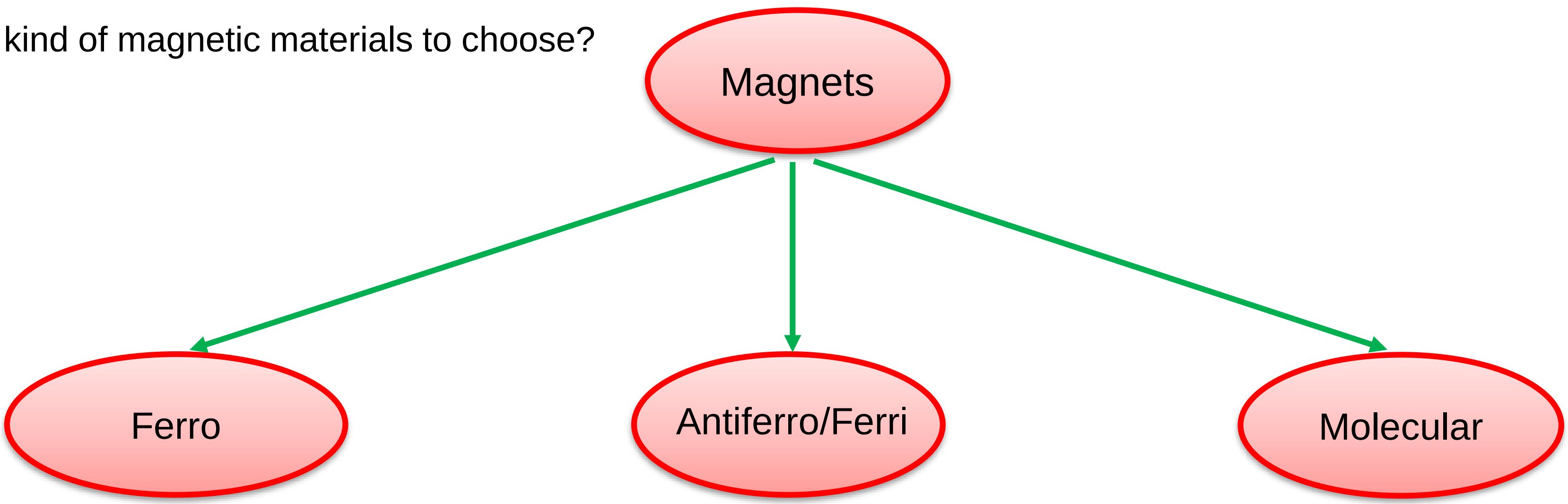
- magnetic anisotropy + magnetic field
- magnetic anisotropy + light helicity
- magnetic field + light helicity + spin-orbit-coupling

II. Introduction: Motivation

Towards nano-spintronics

- Larger storage density
- Faster information processing
- Low power consumption

What kind of magnetic materials to choose?



Ferro
- Broad bands
- Lack in selective control

Antiferro/Ferri
- Good addressability
- Narrow bands
- More than one magnetic center

Molecular
- Discrete energy levels
- Best addressability
- Functionalization of logic elements
- Spatially localized bits

Extended systems (Ferro, Antiferro/Ferri)

II. Introduction: How many centers for nanologic?

- **extended systems (1 point in k-space)**
- **vertical excitations**

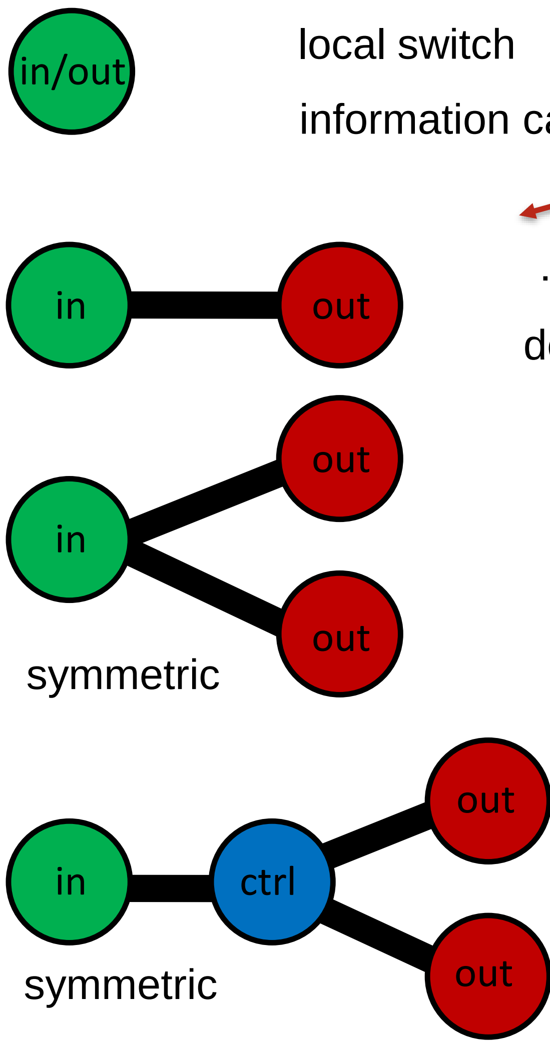
1 center — local switch, information carrier

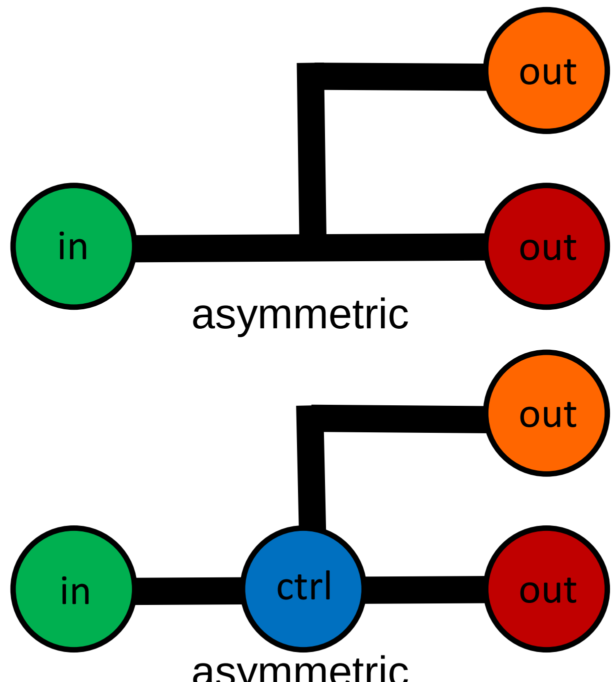
2 centers — …+ transfer

decoupling input/output and propagation of information

3 centers

4 centers





…+ branching

direction of information propagation, interference

…+ control

logic

II. Introduction: Hamiltonian

$$\widehat{H}^{(0)} = -\frac{1}{2}\sum_{i=1}^{N_{\mathrm{el}}} \nabla^2 - \sum_{i=1}^{N_{\mathrm{el}}}\sum_{a=1}^{N_{\mathrm{at}}} \frac{Z_a}{|\mathbf{R}_a - \mathbf{r}_i|} + \sum_{i=1}^{N_{\mathrm{el}}}\sum_{j=1}^{N_{\mathrm{el}}} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} + \sum_{a=1}^{N_{\mathrm{at}}}\sum_{b=1}^{N_{\mathrm{at}}} \frac{Z_a Z_b}{|\mathbf{R}_a - \mathbf{R}_b|}$$

Electronic correlations

$$\widehat{H}^{(1)} = \sum_{i=1}^{N_{\mathrm{el}}} \frac{Z_a^{\mathrm{eff}}}{2c^2 R_i^3} \hat{\mathbf{L}} \cdot \hat{\mathbf{S}} + \sum_{i=1}^{N_{\mathrm{el}}} \mu_L \hat{\mathbf{L}} \cdot \mathbf{B} + \sum_{i=1}^{N_{\mathrm{el}}} \mu_S \hat{\mathbf{S}} \cdot \mathbf{B} + \sum_{i=1}^{N_{\mathrm{el}}} \sum_{\mathbf{q}} \lambda_i^{\mathbf{q}} \langle \mathbf{q} \rangle$$

SOC | Static B-field | phonons

$$\widehat{H}^{(2)}(t) = \langle \widehat{\mathbf{D}} \rangle^{(0+1)} \cdot \mathbf{E}_{\mathrm{laser}}(t) + \langle \widehat{\mathbf{M}} \rangle^{(0+1)} \cdot \mathbf{B}_{\mathrm{laser}}(t)$$

laser

II. Introduction: quantum-chemistry calculations

Hartree-Fock

$$\hat{H}^{(0)} = -\frac{1}{2}\sum_{i=1}^{N_{\mathrm{el}}} \nabla^2 - \sum_{i=1}^{N_{\mathrm{el}}}\sum_{a=1}^{N_{\mathrm{at}}} \frac{Z_a}{|\mathbf{R}_a - \mathbf{r}_i|} + \frac{1}{2}\sum_{i=1}^{N_{\mathrm{el}}}\sum_{j=1}^{N_{\mathrm{el}}} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|} + \frac{1}{2}\sum_{a=1}^{N_{\mathrm{at}}}\sum_{b=1}^{N_{\mathrm{at}}} \frac{Z_a Z_b}{|\mathbf{R}_a - \mathbf{R}_b|}$$

no correlations

CCSD state (correlated reference state)

$$|\Phi^g\rangle = e^{(\hat{S}_1 + \hat{S}_2 + \cdots)}|\Psi_{\mathrm{HF}}\rangle = \left(1 + \hat{S}_1 + \hat{S}_2 + \frac{1}{2!}\hat{S}_1^2 + \frac{1}{2!}\hat{S}_2^2 + \frac{1}{2!}\hat{S}_1\hat{S}_2 + \cdots\right)|\Psi_{\mathrm{HF}}\rangle$$

cumulative expansion
(up to quadruple excitations)

EOM-CCSD states (correlated excited states)

$$|\Phi^e\rangle = \hat{R}|\Phi^g\rangle = (\hat{R}_1 + \hat{R}_2 + \cdots)|\Phi^g\rangle$$

$$\bar{H}\hat{R}|\Phi^g\rangle = E\hat{R}|\Phi^g\rangle$$

$$\bar{H} = e^{-(\hat{S}_1 + \hat{S}_2\,)}\,\hat{H}e^{\hat{S}_1 + \hat{S}_2}$$

correlations on top of correlations

Additional static properties: spin-orbit coupling, Zeeman splitting

$$\hat{H}^{(1)} = \sum_{i=1}^{N_{\mathrm{el}}} \frac{Z_a^{\mathrm{eff}}}{2c^2 R_i^3}\hat{\mathbf{L}}\cdot\hat{\mathbf{S}} + \sum_{i=1}^{N_{\mathrm{el}}} \mu_L \hat{\mathbf{L}}\cdot\mathbf{B} + \sum_{i=1}^{N_{\mathrm{el}}} \mu_S \hat{\mathbf{S}}\cdot\mathbf{B}$$

perturbative inclusion

D. J. Rowe, Rev. Mod. Phys. **40**, 153 (1968)
A. I. Krylov, Ann. Rev. Phys. Chem. **59**, 433 (2008)

II. Introduction: theoretical and background aspects

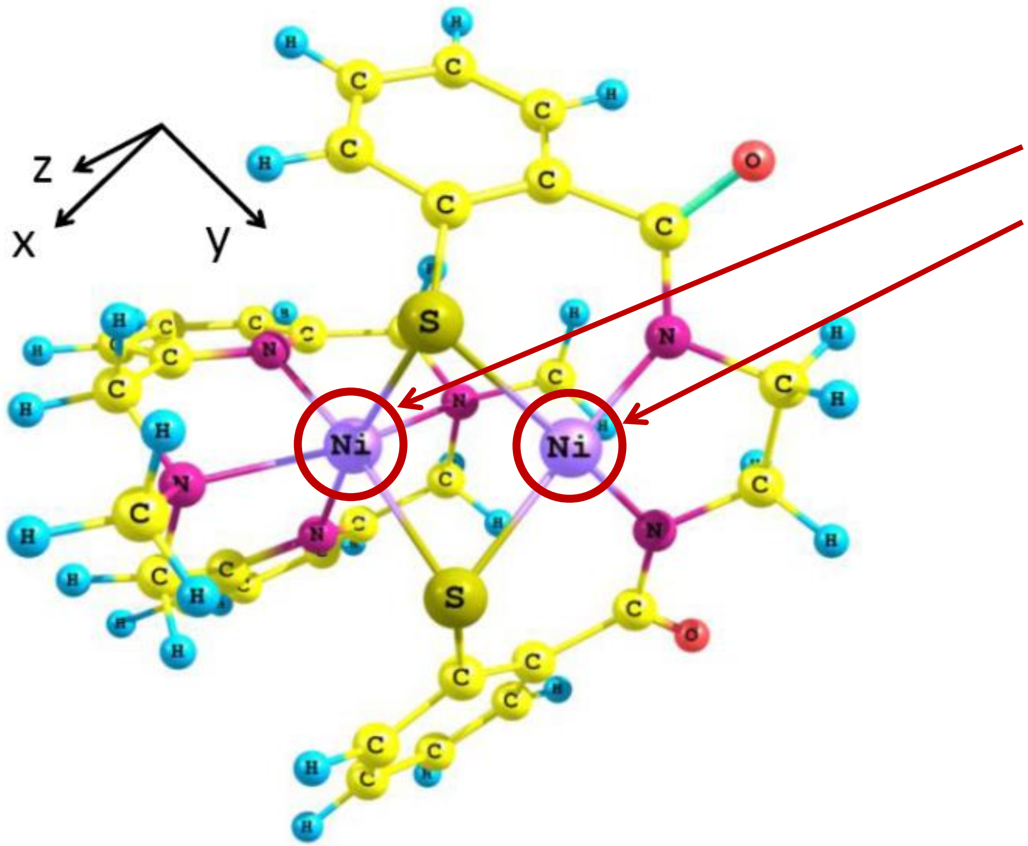
a. Molecular magnets

b. Strong correlations

c. Spin-orbit coupling

d. Laser-induced Λ processes

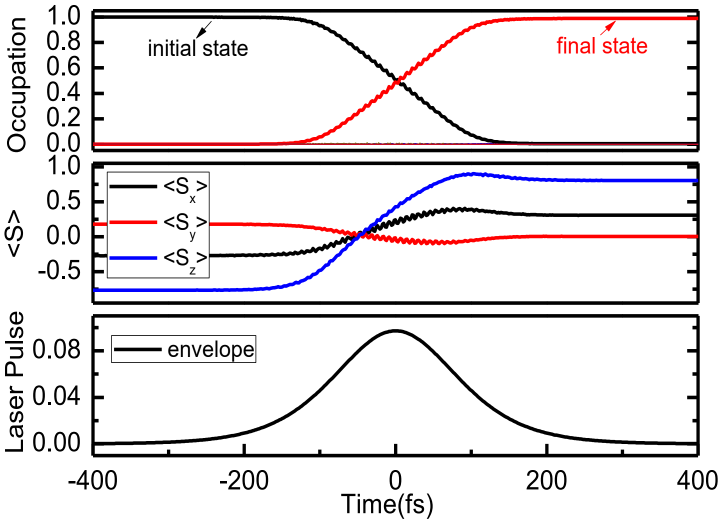
II. Introduction: $[Ni^{II}_2(L\text{-}N_4Me_2)(emb)]^+$ complex



- high-spin (triplet) $Ni_{oct.}$
- low-spin (singlet) $Ni_{sqpl.}$
- synthesized and characterized (Krüger group)
- spectra also from excited states (Diller group)
- **spin dynamics** on triplet, **charge dynamics** on singlet

G. Lefkidis, M. Blug, H. Kelm, C. Li, G. Pal, H.-J. Krüger, and W. Hübner, J. Phys. Chem. A **115**, 1774 (2011)
W. Jin, F. Rupp, K. Chevalier, M. M. N. Wolf, M. Colindres Rojas, G. Lefkidis , H.-J. Krüger, R. Diller, and W. Hübner, Phys. Rev. Lett. **109**, 267209 (2012)

II. Introduction: $[Ni^{II}_2(L\text{-}N_4Me_2)(emb)]^+$ complex



- local spin switch within 300 fsec (Λ process)
- CT states involved belong mainly to $Ni_{sqpl.}$
- **logic** (spin-flip) on $Ni_{oct.}$

Main states	Energy (ev)	$<S_x>$	$<S_y>$	<Sz>	<S>	Spin localization $Ni_{oct.}$	Spin localization $Ni_{sqpl.}$	Charge densities $Ni_{oct.}$	Charge densities $Ni_{sqpl.}$
40	5.905	0.269	0.000	0.670	0.722	1.148	0.308	-0.109	**0.514**
38	5.904	-0.270	0.000	-0.669	0.722	1.148	0.308	-0.109	**0.514**
9	0.849	0.027	0.002	0.057	0.063	1.863	0.004	-0.426	-0.556
6	0.739	0.015	0.031	0.008	0.036	1.861	0.006	-0.424	-0.558
3	0.003	0.376	-0.090	0.730	0.826	**1.917**	0.001	-0.48	**-0.552**
2	0.002	-0.273	0.180	-0.766	0.833	**1.917**	0.001	-0.48	**-0.552**
1	0.000	-0.104	-0.090	0.036	0.142	1.917	0.001	-0.48	-0.552

CT

G. Lefkidis, M. Blug, H. Kelm, C. Li, G. Pal, H.-J. Krüger, and W. Hübner, J. Phys. Chem. A **115**, 1774 (2011)
W. Jin, F. Rupp, K. Chevalier, M. M. N. Wolf, M. Colindres Rojas, G. Lefkidis , H.-J. Krüger, R. Diller, and W. Hübner, Phys. Rev. Lett. **109**, 267209 (2012)

Outline

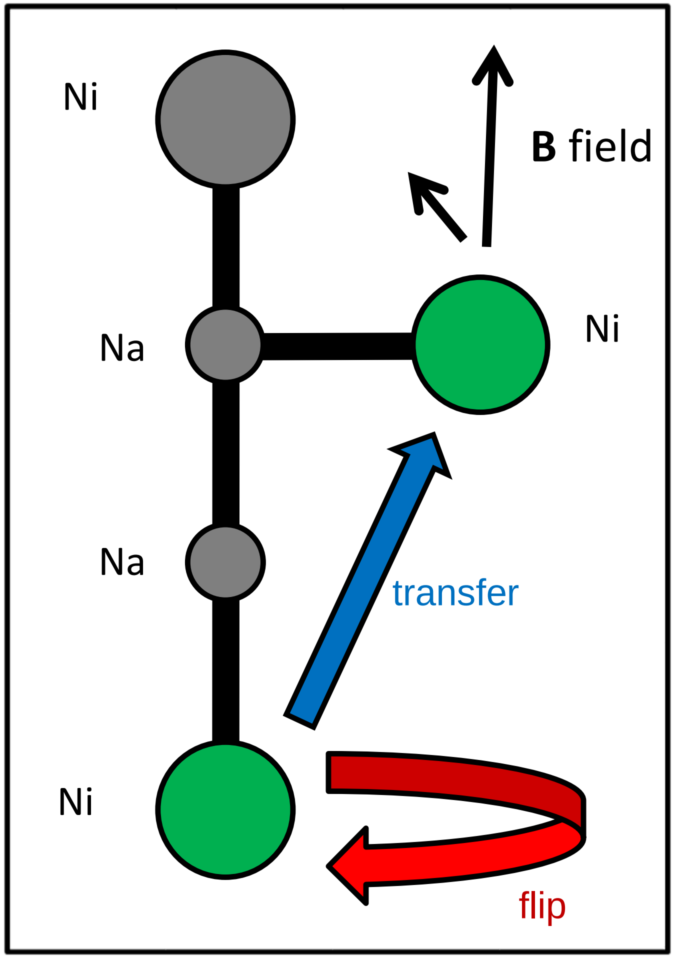
I. History: theory of spin dynamics in extended systems

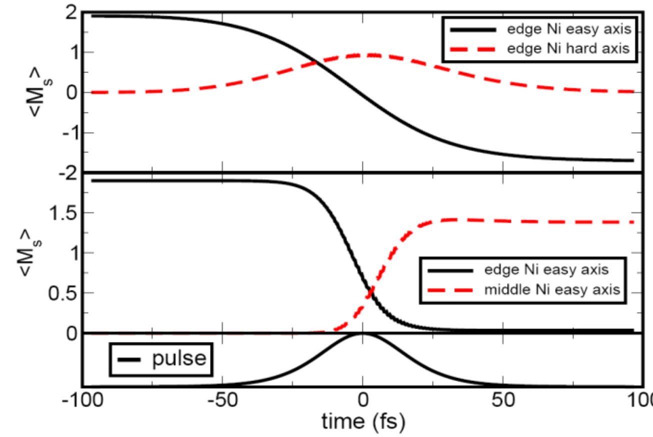
II. Introduction: theoretical and background aspects

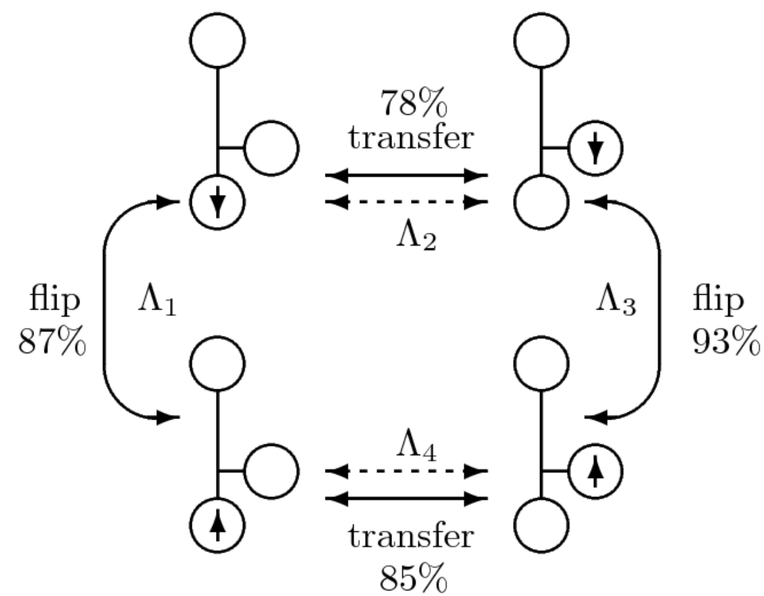
III. Three magnetic centers: magnetic logic

IV. Four magnetic centers: spin SHIFT register & which-path interference

III. Logic: spin logic (AND, OR, XOR gates)

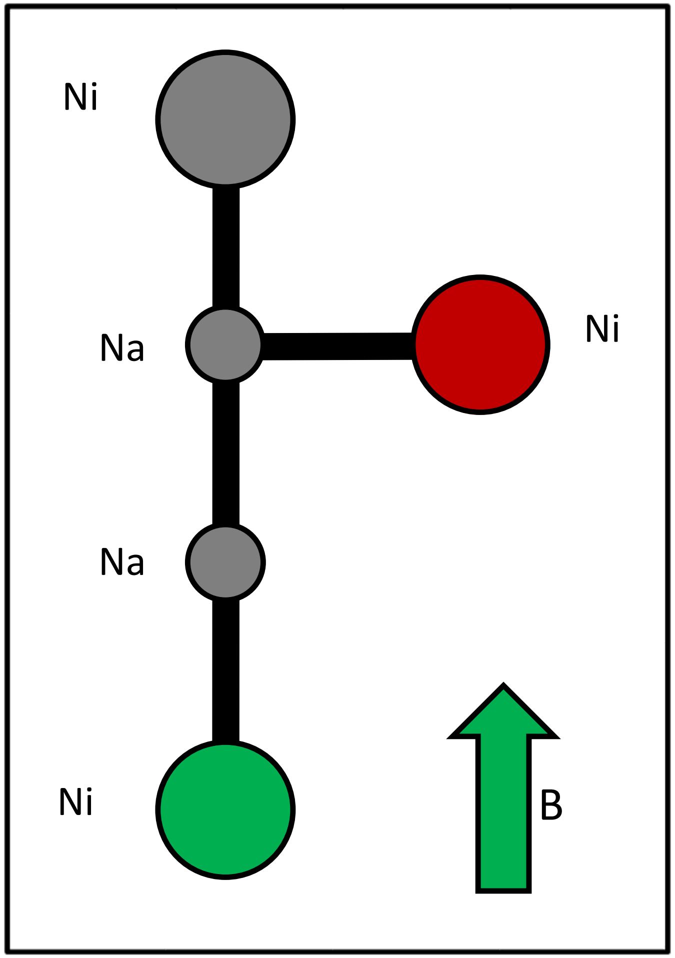






W. Hübner, S. Kersten, and G. Lefkidis, Phys. Rev. B **79**, 184431 (2009)

III. Logic: spin logic (AND, OR, XOR gates)



AND gate

input 1 spin	input 2 **B**-field	output spin+position
1 (edge↑)	1 ($\theta = 0°$)	1 (middle↑)
0 (edge↓)	1 ($\theta = 0°$)	0 (middle↓)
1 (edge↑)	0 ($\theta = 78°$ and $\phi = 96°$)	0 (edge↑)
0 (edge↓)	0 ($\theta = 78°$ and $\phi = 96°$)	0 (edge↓)

OR gate

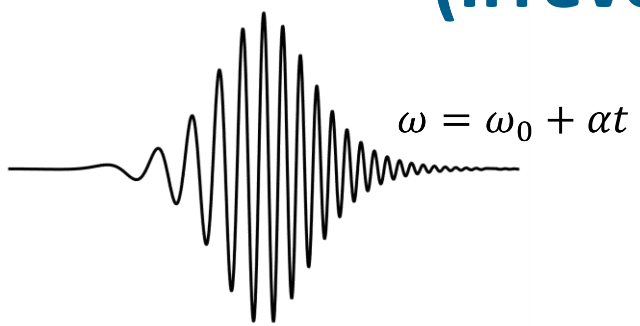
input 1 spin	input 2 **B**-field	output spin+position
0 (edge↑)	0 ($\theta = 0°$)	0 (middle↑)
1 (edge↓)	0 ($\theta = 0°$)	1 (middle↓)
0 (edge↑)	1 ($\theta = 78°$ and $\phi = 96°$)	1 (edge↑)
1 (edge↓)	1 ($\theta = 78°$ and $\phi = 96°$)	1 (edge↓)

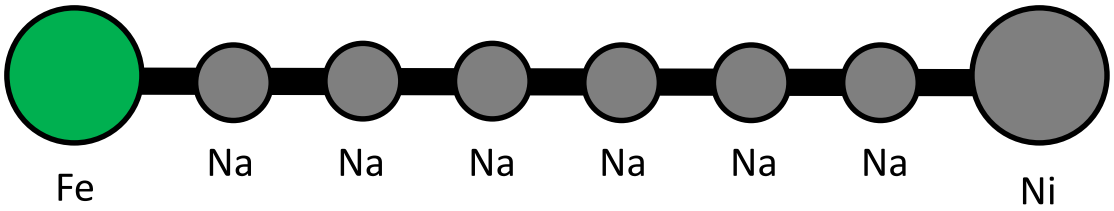
XOR gate (needed for CNOT)

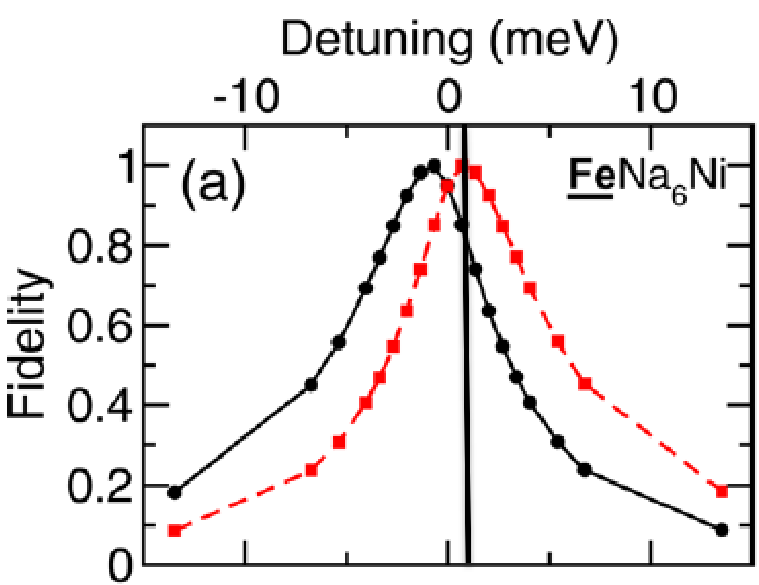
input 1 spin	input 2 **B**-field	output spin
1 (edge↑)	1 ($\theta = 78°$ and $\phi = 96°$)	0 (middle↓)
0 (edge↓)	1 ($\theta = 78°$ and $\phi = 96°$)	1 (middle↑)
1 (edge↑)	0 ($\theta = 0°$)	1 (middle↑)
0 (edge↓)	0 ($\theta = 0°$)	0 (middle↓)

W. Hübner, S. Kersten, and G. Lefkidis, Phys. Rev. B **79**, 184431 (2009)

III. Logic: spin ERASE functionality (Irreversibility through chirp)







- Maximum for two directions of spin flip for opposite chirp
- Fidelitydrop from 99 % to 80 %
- works on pair of states (single Λ process)
- also analytically proven
- at that time limited to very few Na atoms

G. P. Zhang, G. Lefkidis , W. Hübner, and Y. Bai, J. Appl. Phys. **111**, 07C508 (2012)
G. Lefkidis and W. Hübner, Springer Proc. Phys. **159**, 128 (2015)

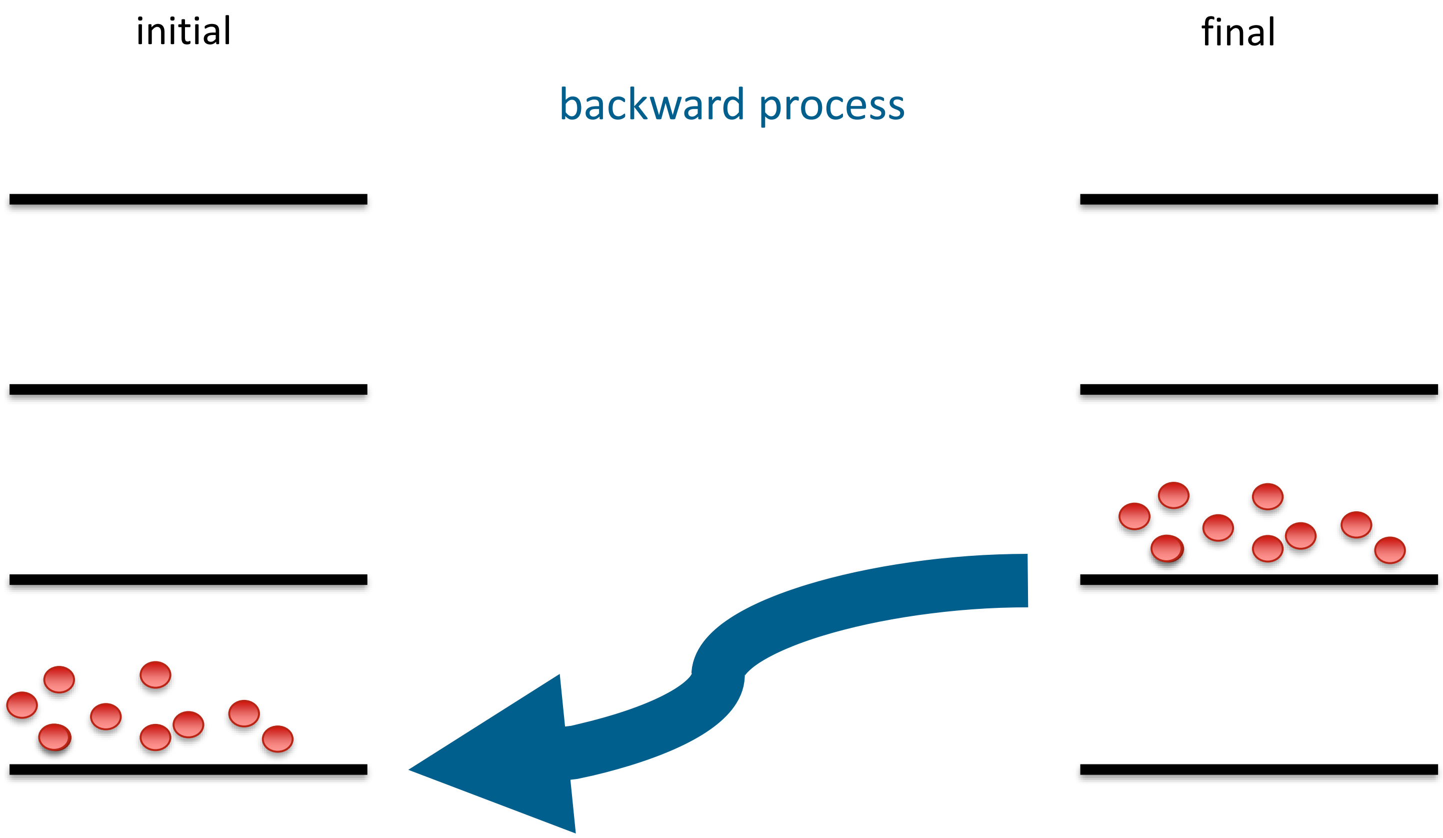
III. Logic: spin ERASE functionality (Irreversibility through quantum interference)

initial

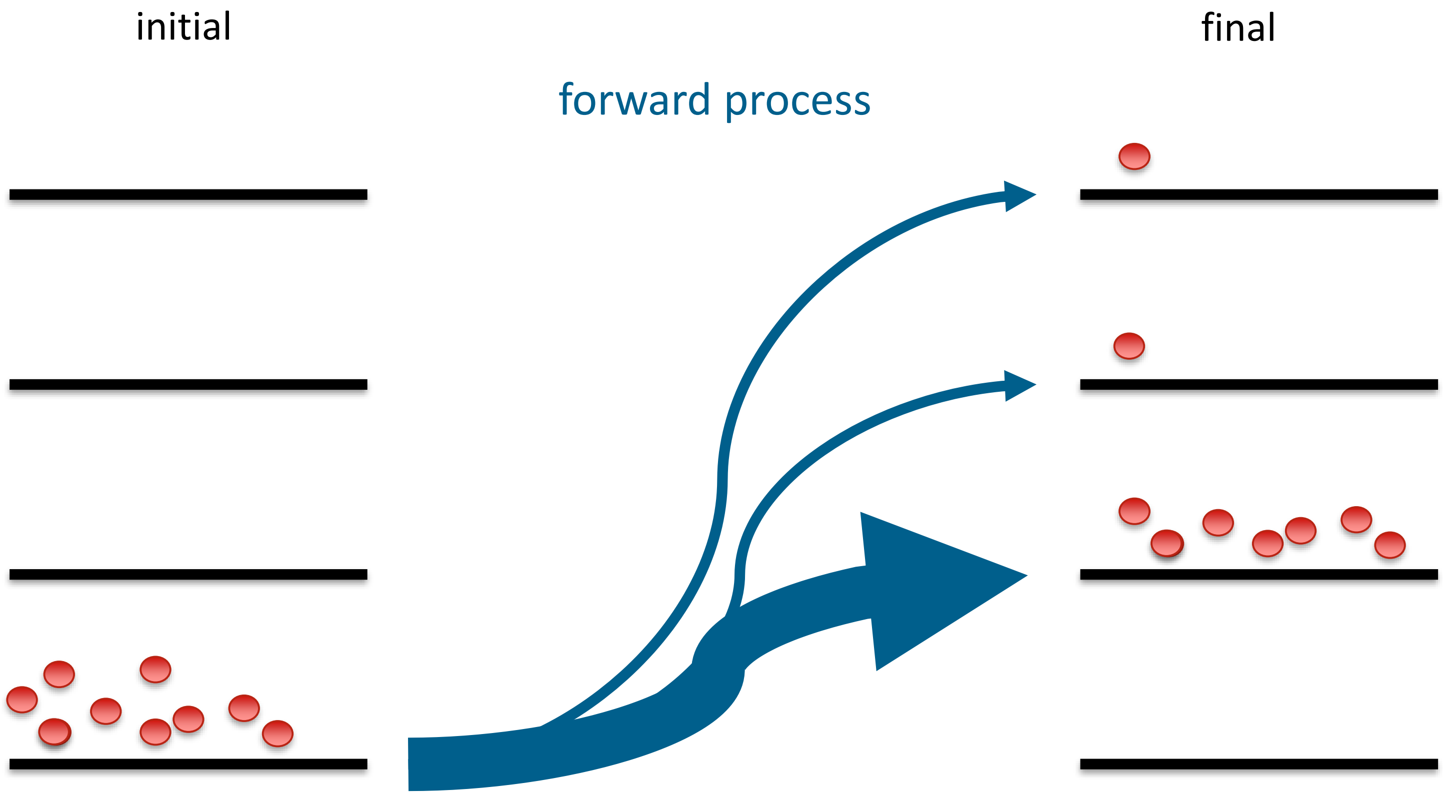
final

forward process

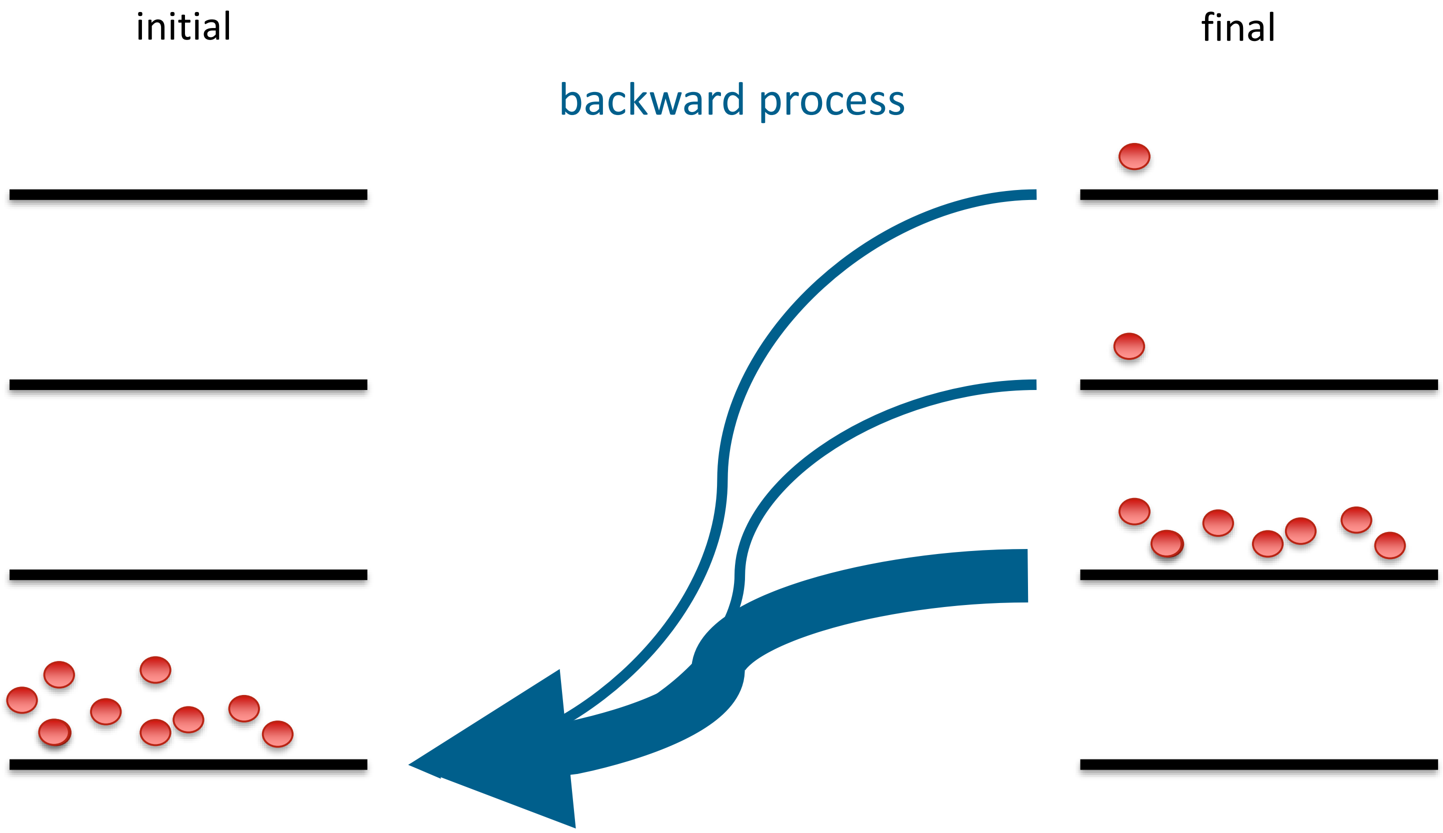
III. Logic: spin ERASE functionality (Irreversibility through quantum interference)



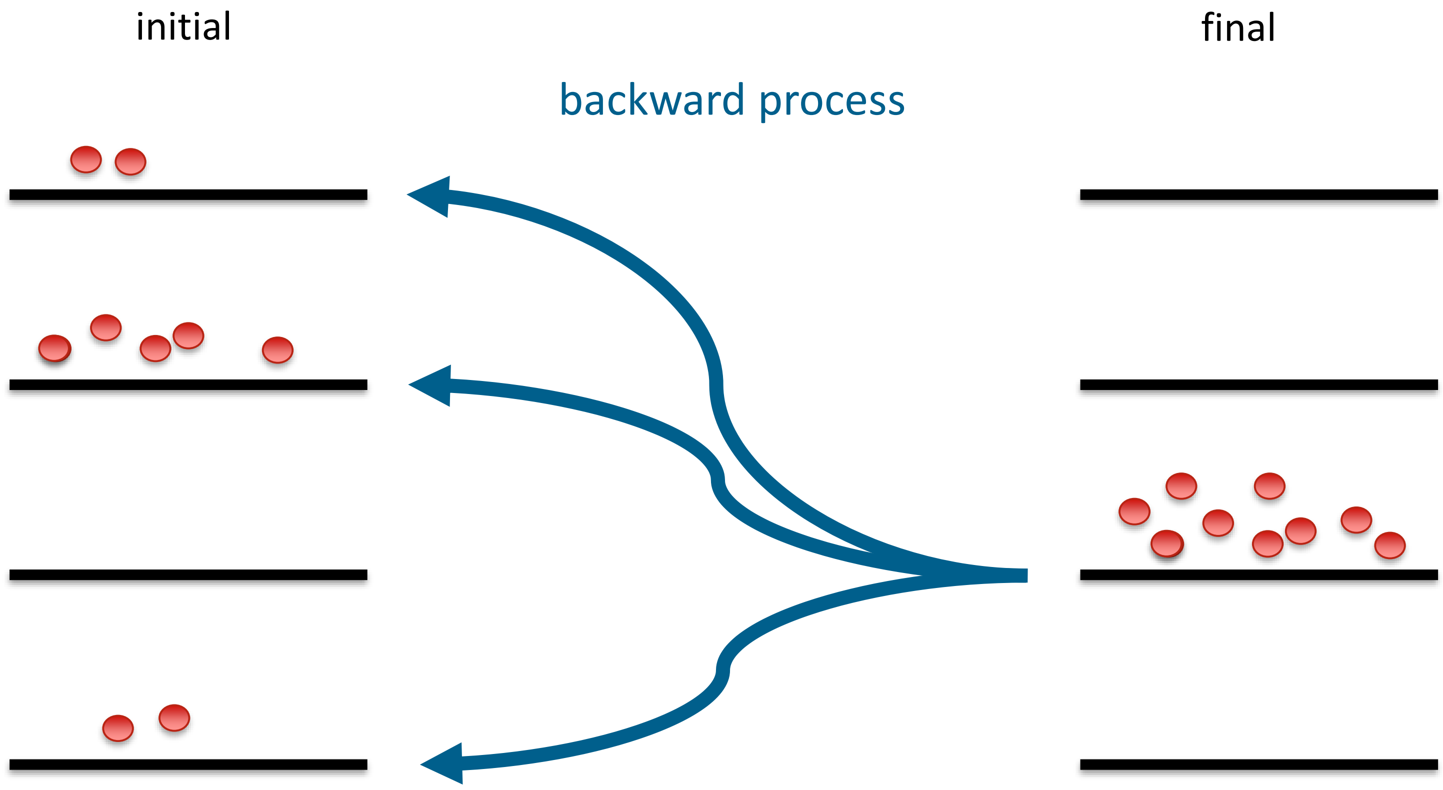
III. Logic: spin ERASE functionality (Irreversibility through quantum interference)



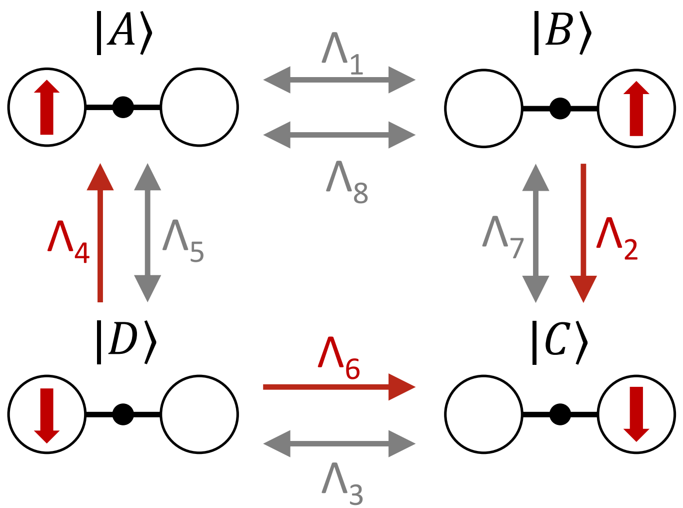
III. Logic: spin ERASE functionality (Irreversibility through quantum interference)

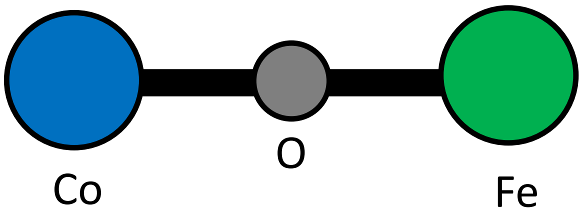


III. Logic: spin ERASE functionality (Irreversibility through quantum interference)



III. Logic: spin ERASE functionality (Irreversibility through quantum interference)





		Λ_2		Λ_6		Λ_5		Λ_6	
state	$\|A\rangle$	→	$\|A\rangle$	→	$\|A\rangle$	→	$\|D\rangle$	→	$\|C\rangle$
state	$\|B\rangle$	→	$\|C\rangle$	→	$\|C\rangle$	→	$\|C\rangle$	→	$\|C\rangle$
state	$\|C\rangle$	→	$\|C\rangle$	→	$\|C\rangle$	→	$\|C\rangle$	→	$\|C\rangle$
state	$\|D\rangle$	→	$\|D\rangle$	→	$\|C\rangle$	→	$\|C\rangle$	→	$\|C\rangle$
entropy	$S = 2$		$S \approx 1.56$		$S \approx 0.99$		$S \approx 0.98$		$S \approx 0.31$

Shannon entropy

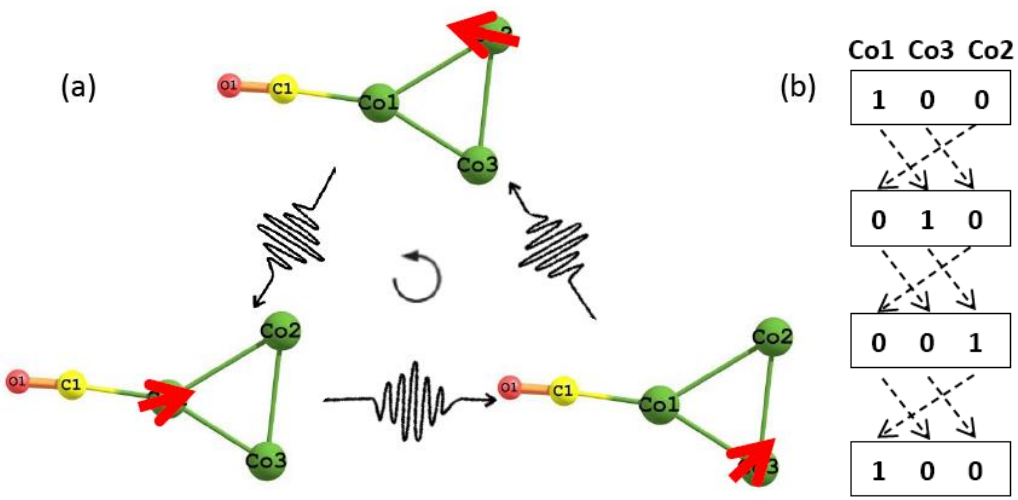
$$S = -\sum p_i \log_2 p_i$$

information gained

entropy decreases

C. Li, S. Zhang, W. Jin, G. Lefkidis, and W. Hübner, Phys. Rev. B **89**, 184404 (2014)

III. Logic: spin cyclic SHIFT register



- not indispensible but:
  - needed for information transfer
  - half-adder ⇒ full-adder
- complete cycle
- lowering of symmetry neccessary

W. Jin, PhD Thesis (Hübner group, 2013)

Outline

I. History: theory of spin dynamics in extended systems

II. Introduction: theoretical and background aspects

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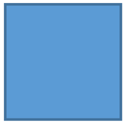
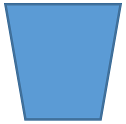
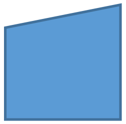
IV. Four magnetic centers: spin SHIFT register & which-path interference

IV. Four centers: Point groups and irreducible representations of Ni_4 (spin-orbit-coupling)



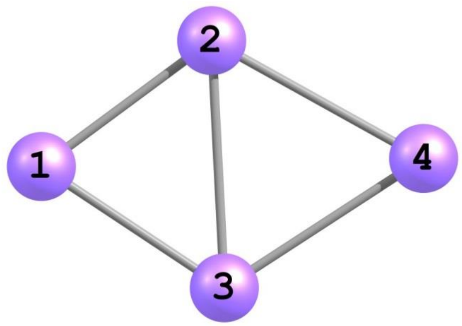



D_{4h}	SOC	D_{2h}	SOC	C_{2v}	SOC	C_s	SOC
A_{1g}	$A_{2u}+E_u$	A_g	$B_{1u}+B_{2u}+B_{3u}$	A_1	$B_1 + 2B_2$	A'	$2A'+A''$
A_{2g}	$A_{1u}+E_u$	B_{1g}	$A_u+B_{2u}+B_{3u}$	A_2	$2B_1 + B_2$	A''	$A'+2A''$
B_{1g}	$B_{2u}+E_u$	A_g	$B_{1u}+B_{2u}+B_{3u}$	A_1	$B_1 + 2B_2$	A'	$2A'+A''$
B_{2g}	$B_{1u}+E_u$	B_{1g}	$A_u+B_{2u}+B_{3u}$	B_1	$A_1 + 2A_2$	A''	$A'+2A''$
E_g	$A_{1u}+A_{2u}+B_{1u}+B_{2u}+E_u$	$B_{2g} + B_{3g}$	$2A_u+2B_{1u}+B_{2u}+B_{3u}$	$B_1 + B_2$	$3A_1 + 3A_2$	$A' + A''$	$3A' + 3A''$
A_{1u}	$A_{2g}+E_g$	A_u	$B_{1g}+B_{2g}+B_{3g}$	A_2	$2B_1 + B_2$	A''	$A'+2A''$
A_{2u}	$A_{1g}+E_g$	B_{1u}	$A_g+B_{2g}+B_{3g}$	A_1	$B_1 + 2B_2$	A'	$2A'+A''$
B_{1u}	$B_{2g}+E_g$	A_u	$B_{1g}+B_{2g}+B_{3g}$	A_2	$2B_1 + B_2$	A''	$A'+2A''$
B_{2u}	$B_{1g}+E_g$	B_{1u}	$A_g+B_{2g}+B_{3g}$	B_2	$2A_1 + A_2$	A'	$2A'+A''$
E_u	$A_{1g}+A_{2g}+B_{1g}+B_{2g}+E_g$	$B_{2u} + B_{3u}$	$2A_g+2B_{1g}+B_{2g}+B_{3g}$	$B_1 + B_2$	$3A_1 + 3A_2$	$A' + A''$	$3A' + 3A''$

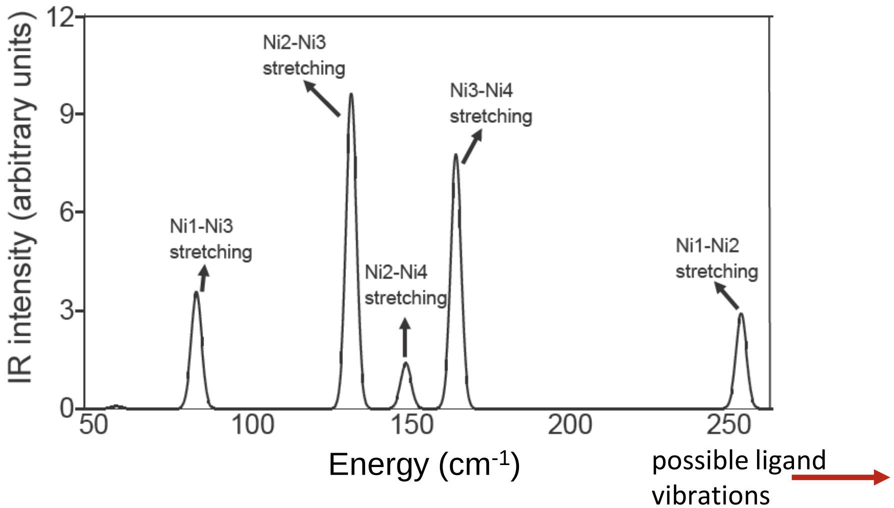
10 irreps 2×2D	8 irreps no 2D	4 irreps no 2D	2 irreps no 2D

IV. Four centers: Optimized geometry of Ni_4

- Geometry



- IR spectra



- Hartree-Fock optimized bond lengths (Å)

species	Ni1−Ni2	Ni2−Ni4	Ni4−Ni3	Ni1−Ni3
Ni_4	2.2709	2.5996	2.54763	2.3508

- All possible functionalities

- Spin flips
- Spin transfers
- Spin bifurcation
- Spin-SHIFT-register
- OR gate

PHYSIK

OPTIMAS light-spin-matter

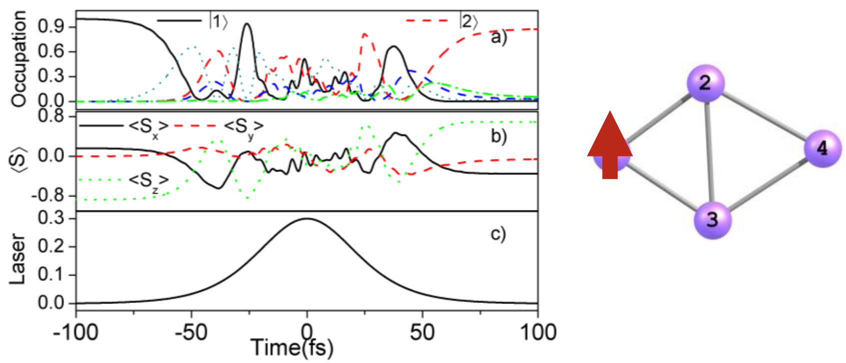
SFB/TRR 88 3MET.de

SPIN+X SFB/TRR 173 Kaiserslautern • Mainz

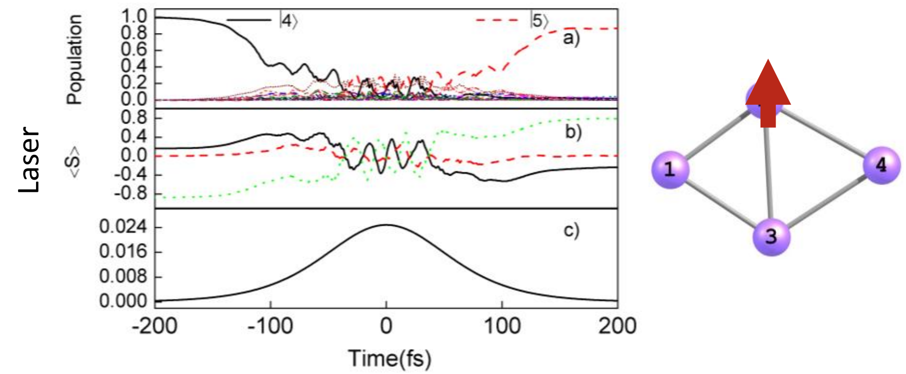
TECHNISCHE UNIVERSITÄT KAISERSLAUTERN

IV. Four centers: local spin-switch scenarios on Ni_4

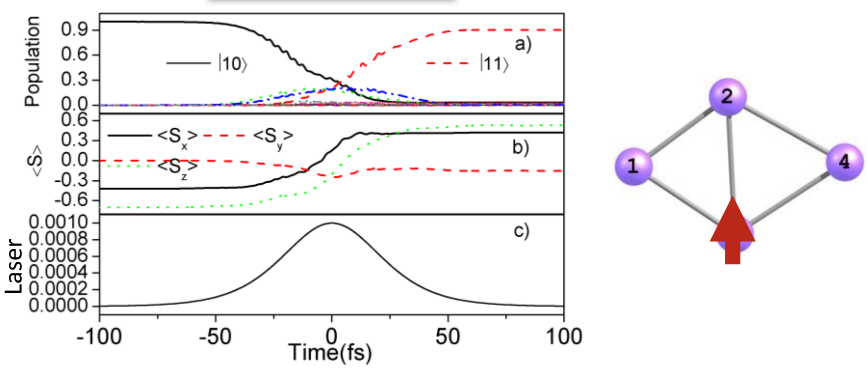
Ni1 < 150 fs



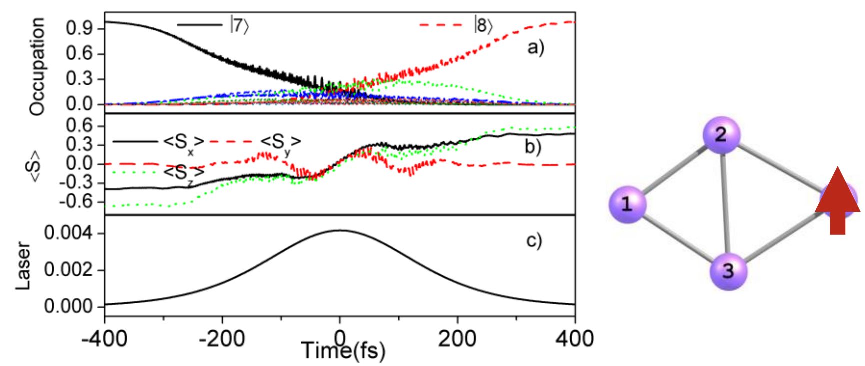
Ni2 < 200 fs



Ni3 < 90 fs



Ni4 < 500 fs

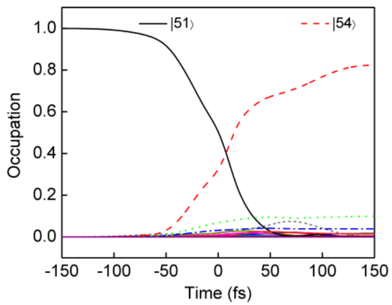


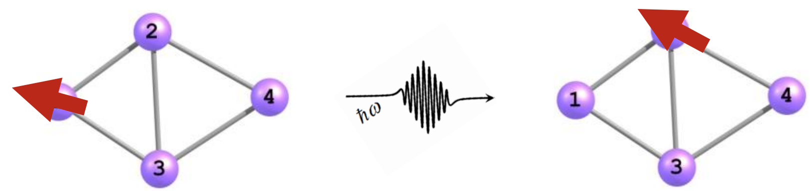
Fastest spin switch ever so far!!

D. Chaudhuri, G. Lefkidis, and W. Hübner, Phys. Rev. B **96**, 184413 (2017)

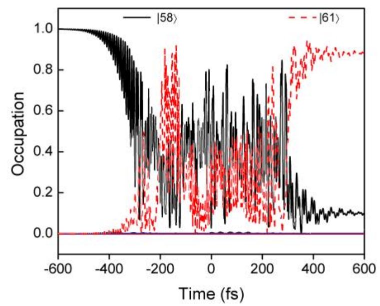
IV. Four centers: Three spin-transfer scenarios on Ni_4

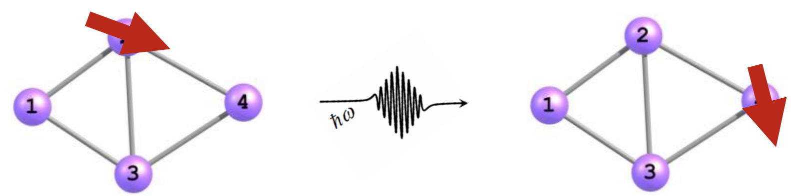
Ni1 -> Ni2



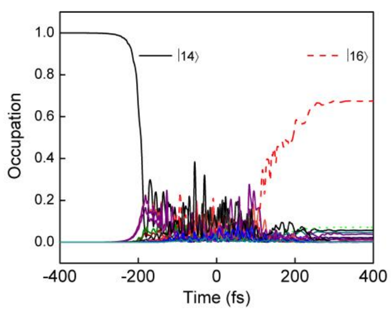


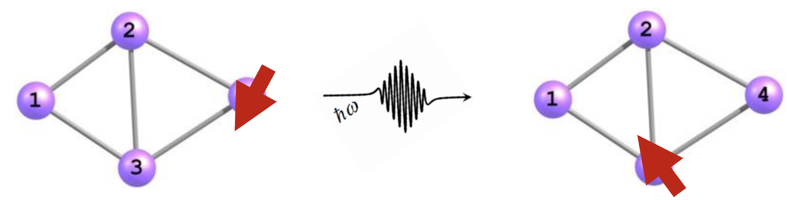
Ni2 -> Ni4





Ni4 -> Ni3

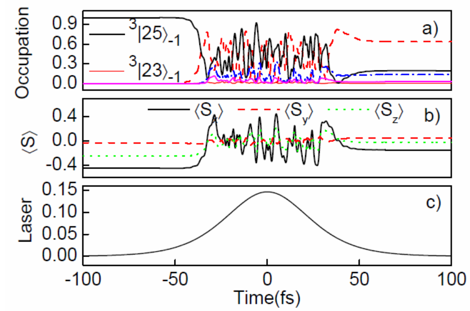


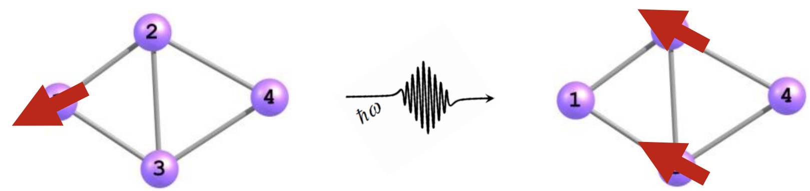


D. Chaudhuri, G. Lefkidis, and W. Hübner, Phys. Rev. B **96**, 184413 (2017)

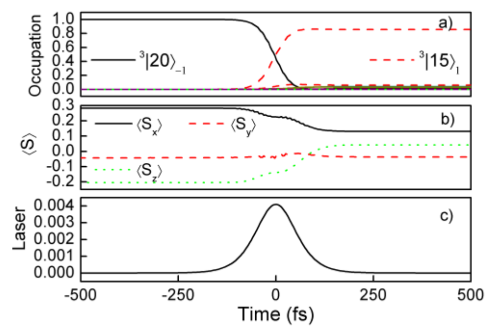
IV. Four centers: Spin bifurcation and spin merging on Ni_4

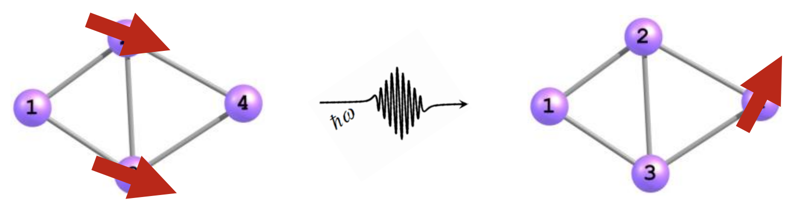
Ni1 -> Ni2+Ni3





Ni2+Ni3 -> Ni4

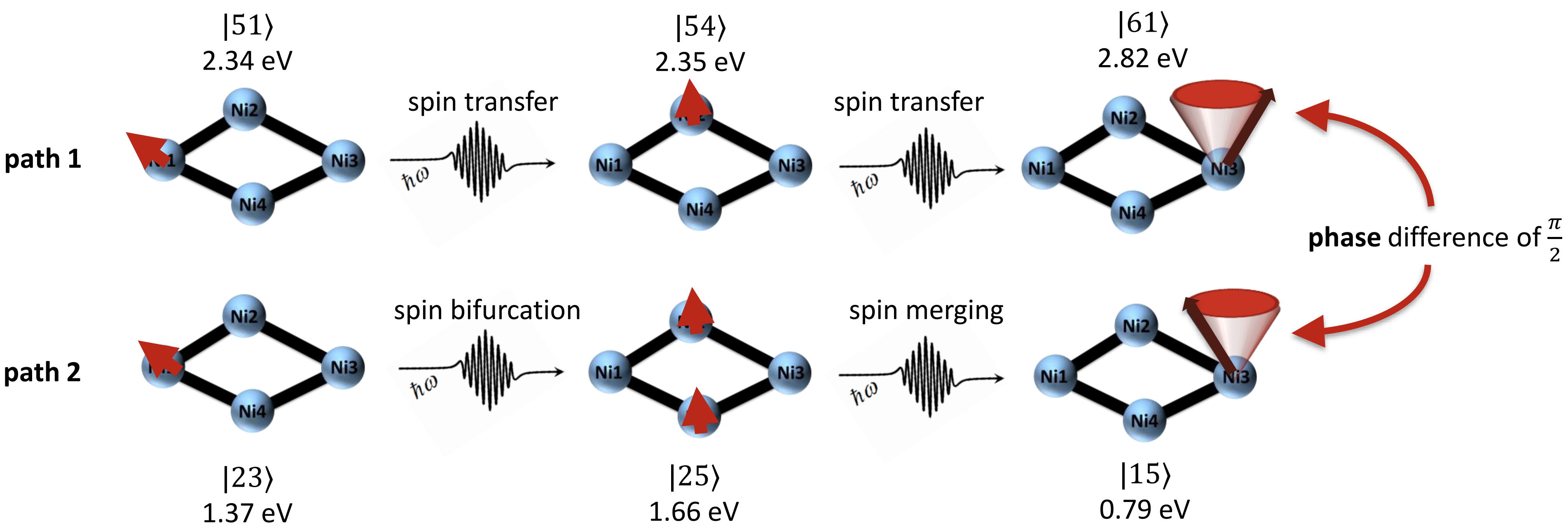




- After the bifurcation the spin is equidistributed on Ni2 and Ni3
- Both bifurcation and merging in less than 100 fs

D. Chaudhuri, G. Lefkidis, and W. Hübner, Phys. Rev. B **96**, 184413 (2017)

IV. Four centers: Quantum computing on Ni_4: *which-path* interference

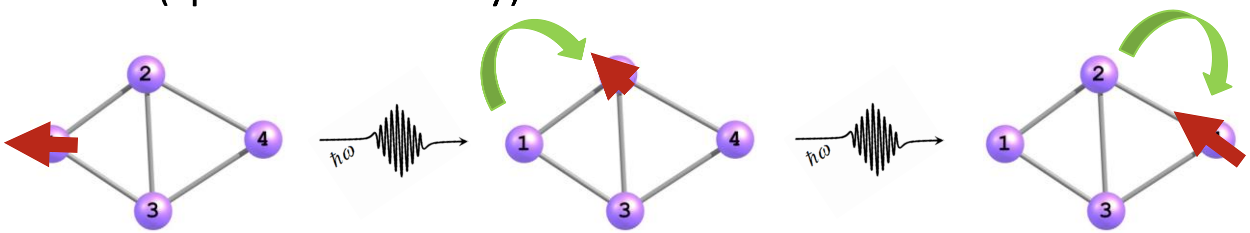


Phase of wavefunction after the whole process reveals the **path** traveled by the spin.

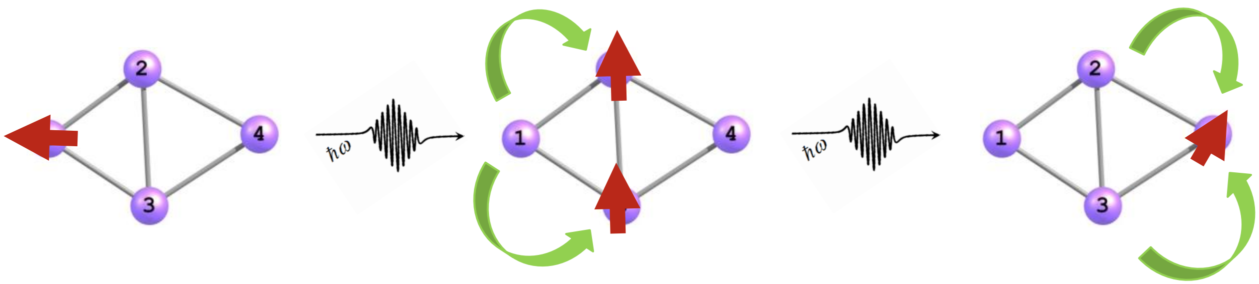
D. Chaudhuri, G. Lefkidis, and W. Hübner, Phys. Rev. B **96**, 184413 (2017)

IV. Four centers: Which-path interference on Ni_4

Path 1 (spin transfer only)



Path 2 (spin bifurcation and spin merging)



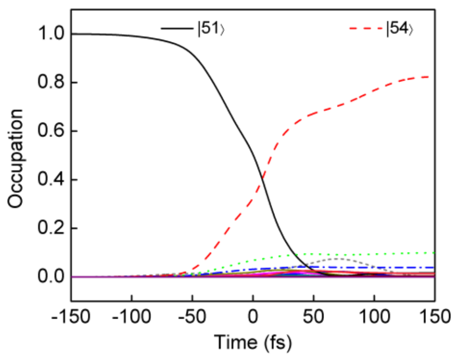
(the red arrows indicate the phase of the spin)

- $\frac{\pi}{2}$ phase difference in the final state
- detectable through interference with laser pulse

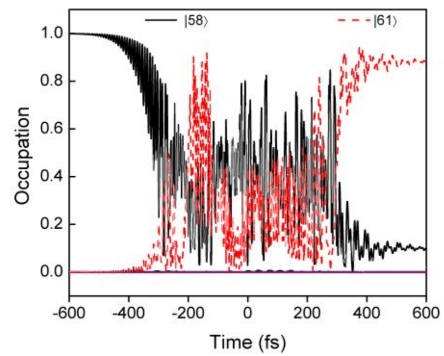
D. Chaudhuri, G. Lefkidis, and W. Hübner, Phys. Rev. B **96**, 184413 (2017)

IV. Four centers: Spin-SHIFT-register on Ni_4

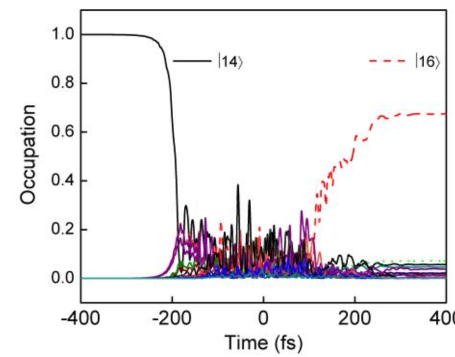
- Ni1 → Ni2



- Ni2 → Ni3

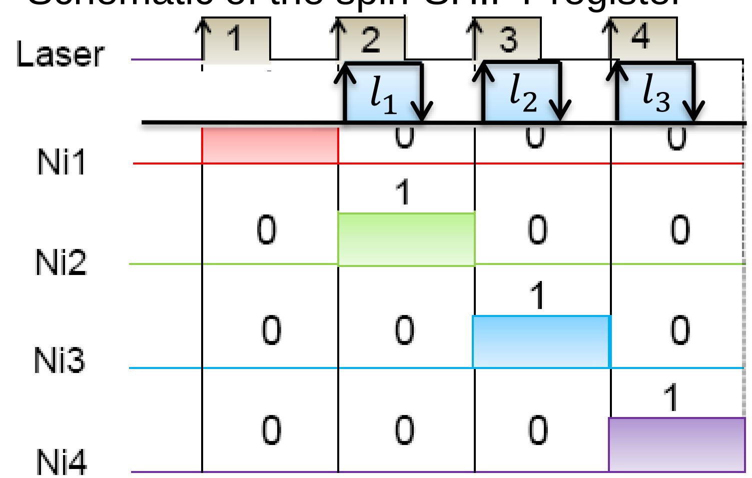


- Ni3 → Ni4



- Series of spin-transfer scenarios
- Ability to store and transfer binary bits
- Movement of data in a register to the right
- Prototypic spin-SHIFT- register

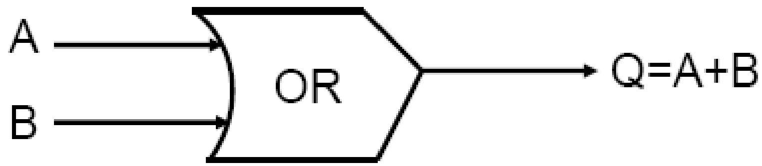
- Schematic of the spin-SHIFT-register



D. Chaudhuri, G. Lefkidis, and W. Hübner, Phys. Rev. B **96**, 184413 (2017)

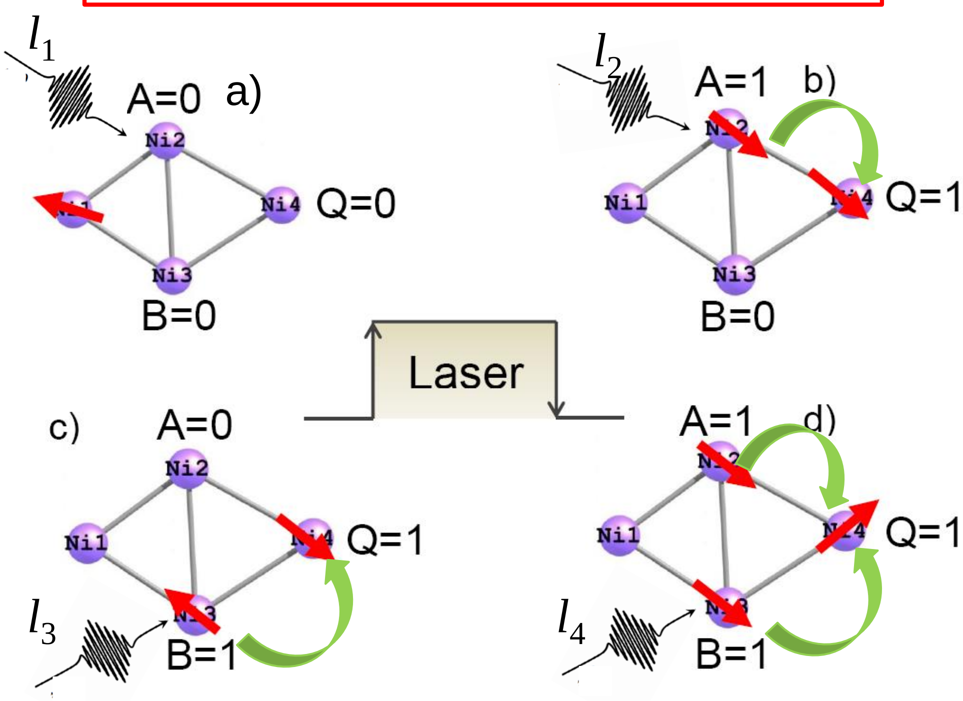
IV. Four centers: pure-spin OR gate on Ni_4

- OR gate



- Laser used as a trigger to transfer spins
- Atom with spin localized is considered 1, otherwise 0.
- A and B are inputs, Q is output

- Schematic of spin transfer scenarios

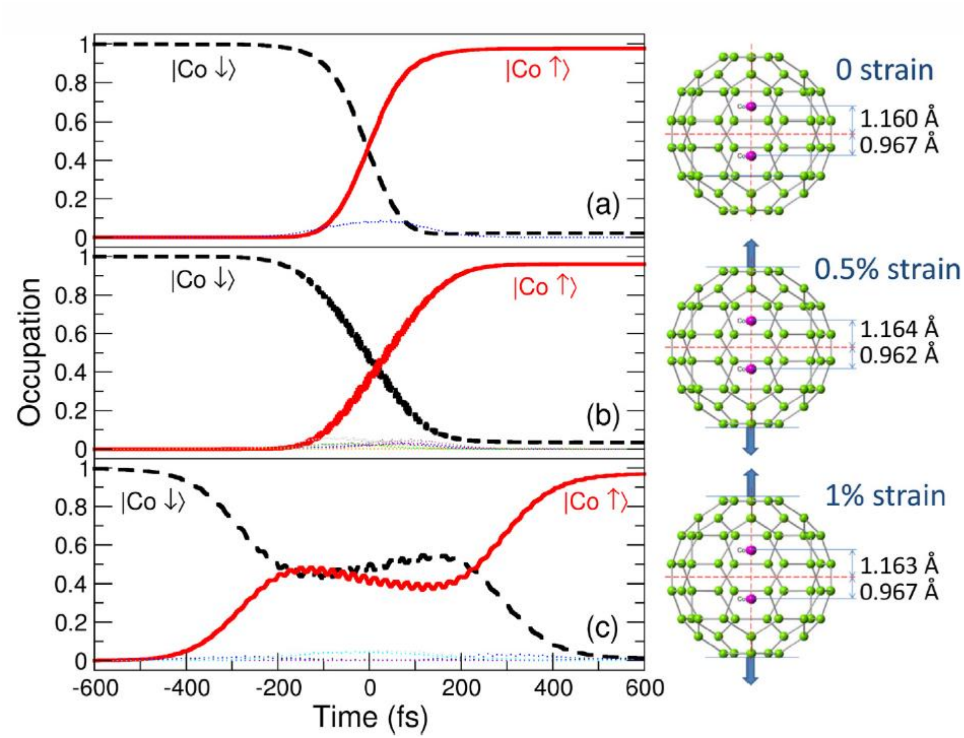


Laser pulse	Ni2 A	Ni3 B	Ni4 Q=A+B	fidelity (f)
l_1	0	0	0	–
l_2	1	0	1	97%
l_3	0	1	1	60%
l_4	1	1	1	84%

D. Chaudhuri, G. Lefkidis, and W. Hübner, Phys. Rev. B **96**, 184413 (2017)

Outlook: C_{60}-supported nanologic

Strain	Atom	State 1	State 2	State 3	State 4	State 5
0	Co1	0.072	1.820	0.082	1.841	0.066
0	Co2	1.870	0.060	1.773	0.067	1.810
0.5%	Co1	0.060	1.823	0.975	0.067	1.837
0.5%	Co2	1.870	0.064	0.940	1.782	0.058
1.0%	Co1	0.058	1.833	1.854	0.065	1.845
1.0%	Co2	1.877	0.060	0.038	1.786	0.064

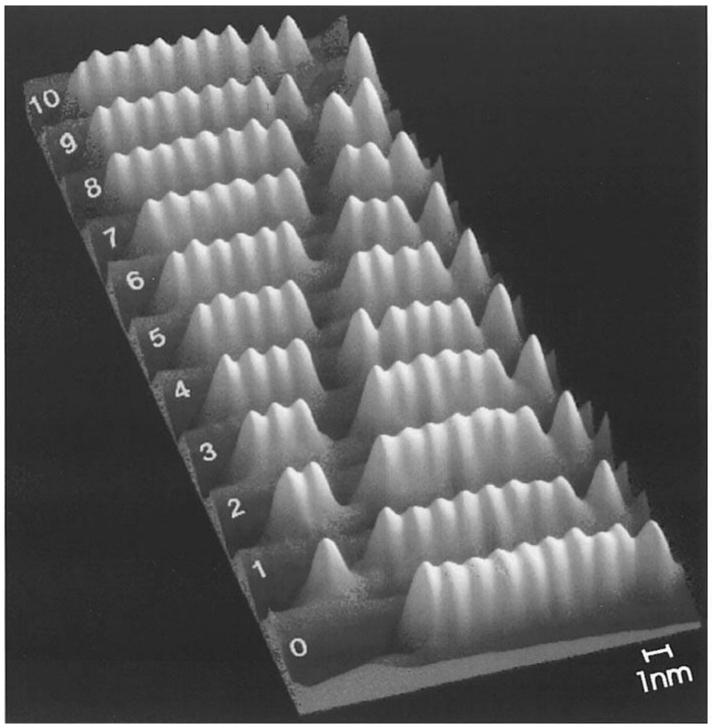


- C_{60} protects the Co_2 dimer
- mechanical control over spin and spin dynamics

C. Li, J. Liu, S. Zhang, G. Lefkidis, and W. Hübner, Carbon **87**, 153 (2015)

Outlook: C_{60}-supported nanologic

Strain	Atom	State 1	State 2	State 3	State 4	State 5



Abacus using C_{60} molecules
⇒ stability!

M. T. Cuberes, R. R. Schlittler, & J. K. Gimzewski, Appl. Phys. Lett. **69**, 3016 (1996)

C. Li, J. Liu, S. Zhang, G. Lefkidis, and W. Hübner, Carbon **87**, 153 (2015)

IV. Summary: magnetic molecules for logic applications

- Two and three magnetic centers (AND, OR, XOR, ERASE, and SHIFT functionalities)
- Four magnetic centers (pure-spin logic, quantum interference, quantum logic)
- Carbon chains possible route to optically triggered long-range spin transfer