Laser-induced spin dynamics at the femto-second time scale: Understanding switching mechanisms with real-time TDDFT

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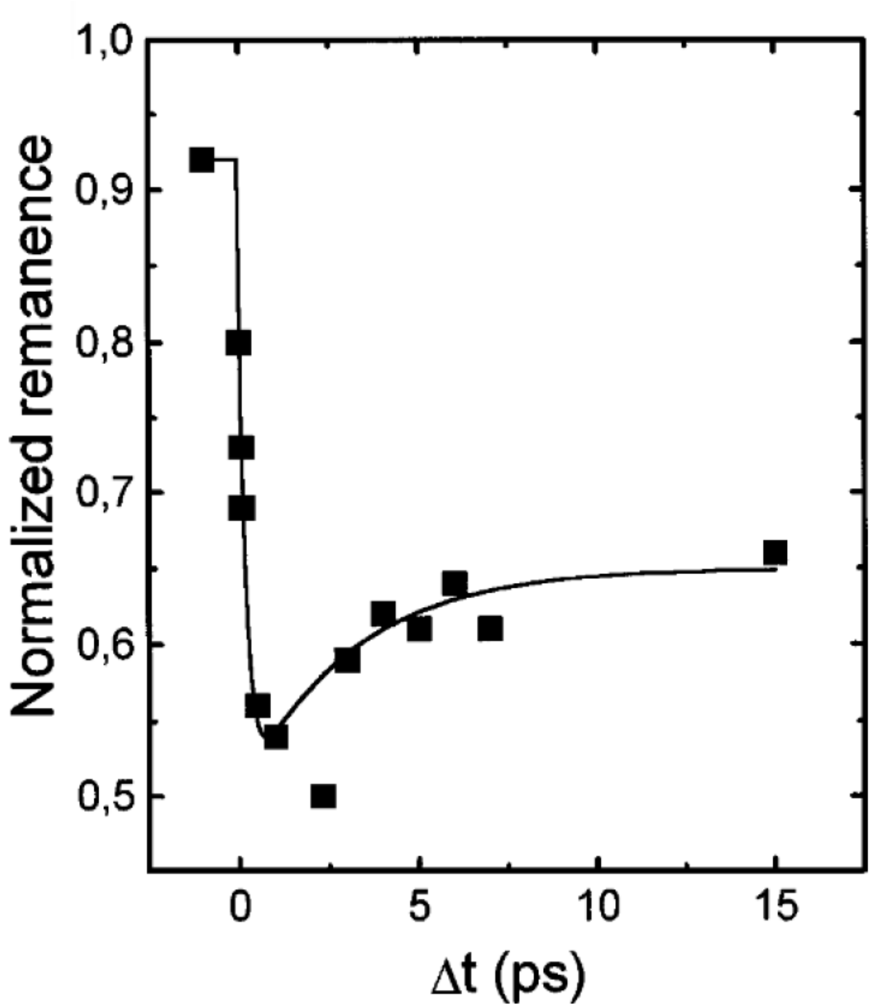
Halle (Saale)

First experiment on ultrafast laser induced demagnetization



Beaurepaire et al, PRL 76, 4250 (1996)

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- **Our proposal for the first 50 fs:**
  Laser-induced charge excitation followed by spin-orbit-driven demagnetization of the remaining d-electrons

Quantity of prime interest:
vector field of spin magnetization

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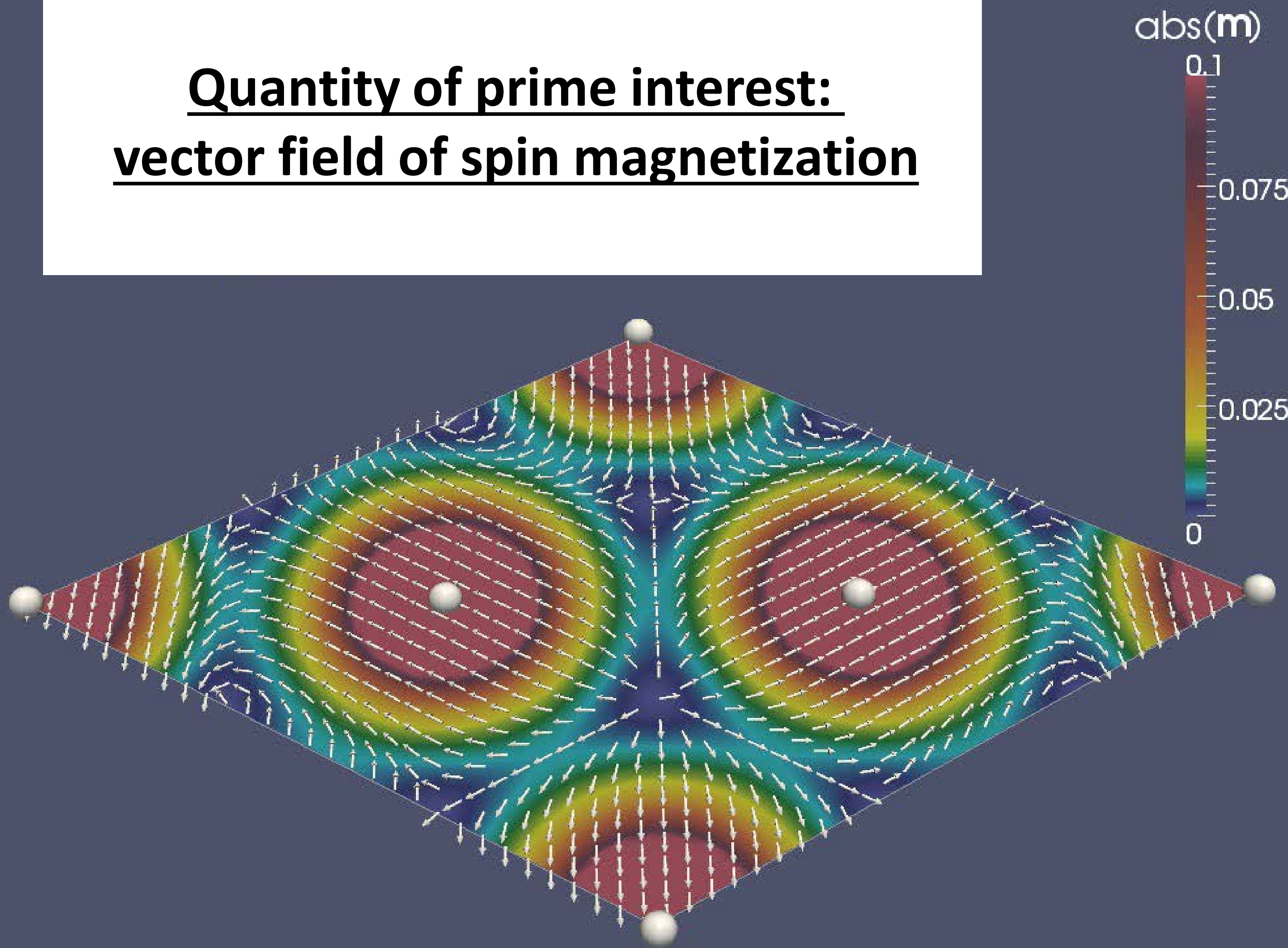

Cr monolayer in ground state

Theoretical approach:

Time-dependent density-functional theory
(E. Runge, E.K.U.G., PRL 52, 997 (1984))

Basic 1-1 correspondence:

$$v(rt) \overset{1\text{-}1}{\longleftrightarrow} \rho(rt)$$

The time-dependent density determines uniquely the time-dependent external potential and hence all physical observables for fixed initial state.

KS theorem:

The time-dependent density of the interacting system of interest can be calculated as density

$$\rho(rt) = \sum_{j=1}^{N} \left|\varphi_j(rt)\right|^2$$

of an auxiliary non-interacting (KS) system

$$i\hbar\frac{\partial}{\partial t}\varphi_j(rt) = \left(-\frac{\hbar^2\nabla^2}{2m} + v_S[\rho](rt)\right)\varphi_j(rt)$$

with the local potential

$$v_S\left[\rho(r't')\right](rt) = v(rt) + \int d^3r' \frac{\rho(r't)}{|r-r'|} + v_{xc}\left[\rho(r't')\right](rt)$$

Generalization: Non-collinear-Spin-TDDFT with SOC

$$i\frac{\partial}{\partial t}\varphi_k(r,t) = \left[\frac{1}{2}\left(-i\nabla - A_{laser}(t)\right)^2 + v_S[\rho,\boldsymbol{m}](r,t) - \mu_B\sigma\cdot B_S[\rho,\boldsymbol{m}](r,t) + \frac{\mu_B}{2c}\sigma\cdot\left(\nabla v_S[\rho,\boldsymbol{m}](r,t)\right)\times\left(-i\nabla\right)\right]\varphi_k(r,t)$$

$$v_S[\rho,\boldsymbol{m}](r,t) = v_{lattice}(r) + \int\frac{\rho(r',t)}{|r-r'|}d^3r' + v_{xc}[\rho,\boldsymbol{m}](r,t)$$

$$B_S[\rho,\boldsymbol{m}](r,t) = B_{external}(r,t) + B_{xc}[\rho,\boldsymbol{m}](r,t)$$

where $\varphi_k(r,t)$ are Pauli spinors

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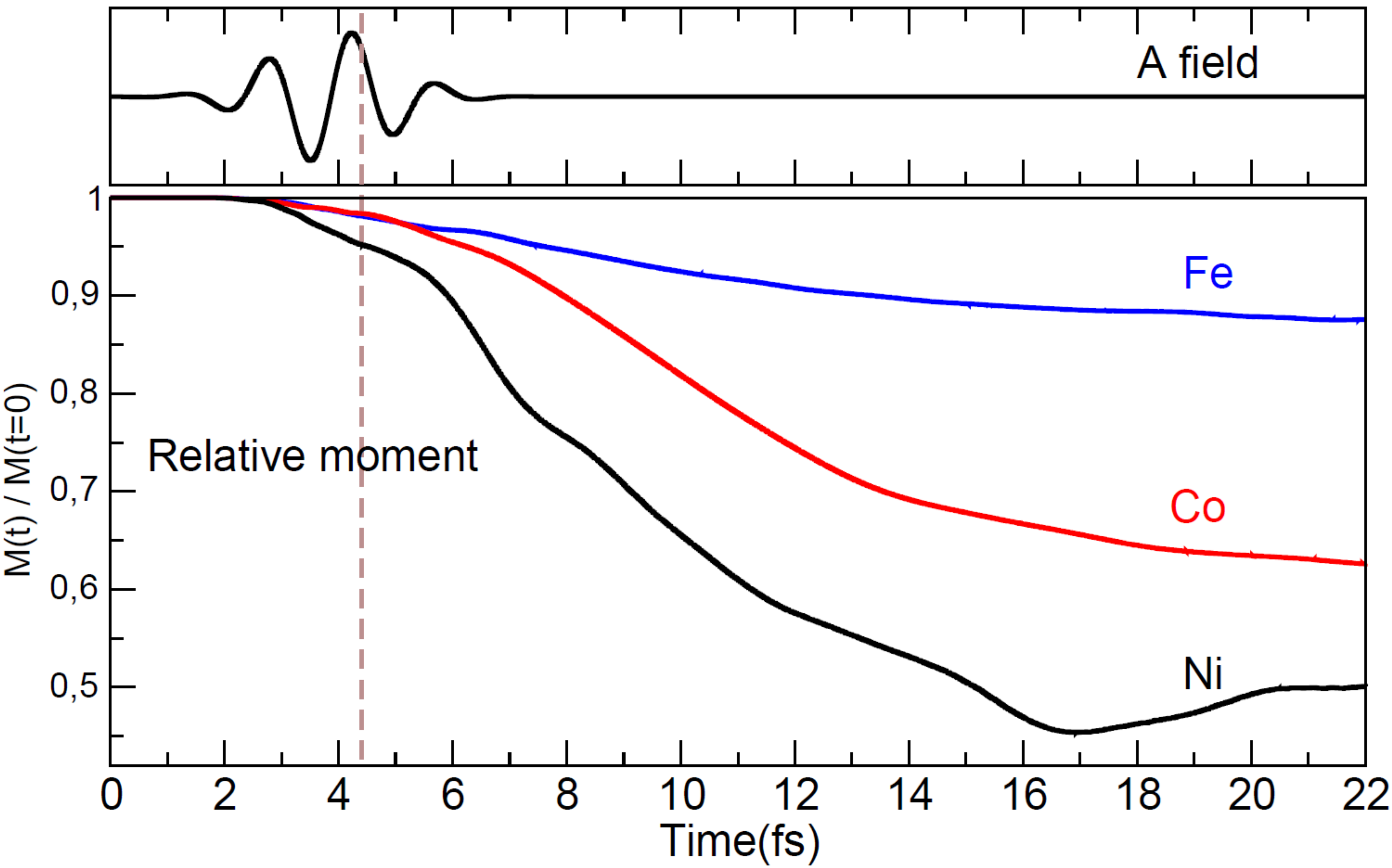
$$v_S[\rho,\boldsymbol{m}](r,t)=v_{lattice}(r)+\int\frac{\rho(r',t)}{|r-r'|}d^3r'+v_{xc}[\rho,\boldsymbol{m}](r,t)$$

$$B_S[\rho,\boldsymbol{m}](r,t)=B_{external}(r,t)+B_{xc}[\rho,\boldsymbol{m}](r,t)$$

Universal functionals of ρ and m

where $\varphi_k(r,t)$ are Pauli spinors

Demagnetisation in Fe, Co and Ni



K. Krieger, K. Dewhurst, P. Elliott, S. Sharma, E.K.U.G., JCTC 11, 4870 (2015)

Aspects of the implementation

- Wave length of laser in the visible regime (very large compared to unit cell)

  ⇒ Dipole approximation is made (i.e. electric field of laser is assumed to be spatially constant)

  ⇒ Laser can be described by a purely time-dependent vector potential

- **Periodicity of the TDKS Hamiltonian is preserved!**
- **Implementation in ELK code (FLAPW) (http://elk.sourceforge.net/)**

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Kay Dewhurst

ELK = Electrons in K-Space
or
Electrons in Kay's Space

Sangeeta Sharma

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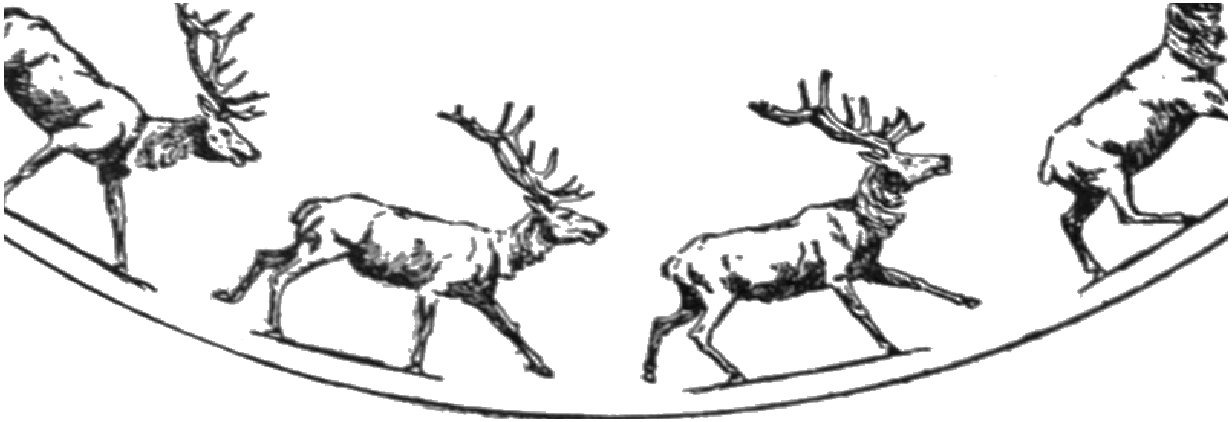


Algorithm for time propagation

1. Set $\psi_j(\mathbf{r},t) = \sum_i c_{ij}(t)\chi_i(\mathbf{r})$

2. Compute $\rho(\mathbf{r},t)$ and $\mathbf{m}(\mathbf{r},t)$

3. Compute $v_s(\mathbf{r},t)$, $\mathbf{B}_s(\mathbf{r},t)$, $\mathbf{A}_s(\mathbf{r},t)$ to give $\hat{H}_{KS}(t)$

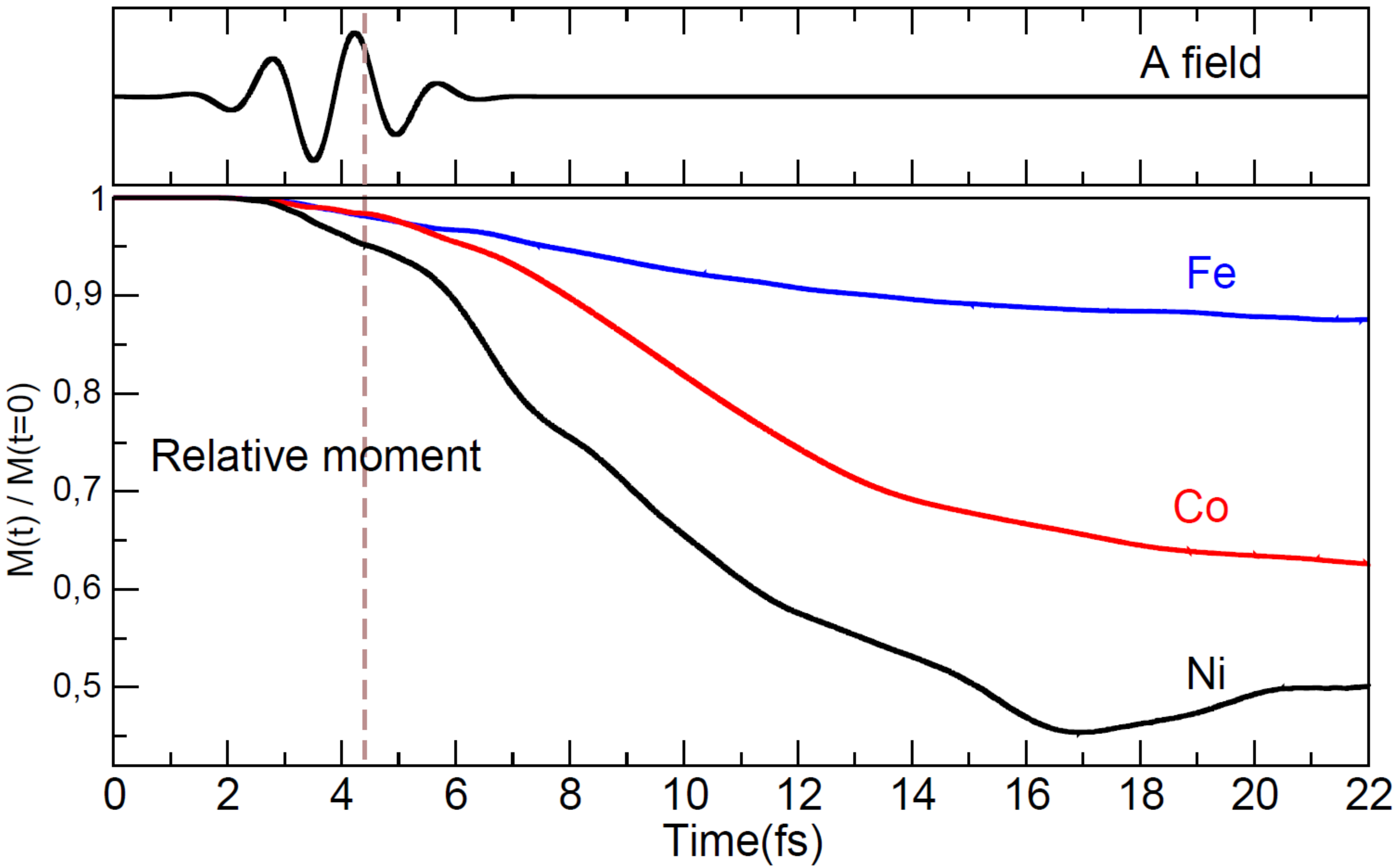
4. Compute $H_{ij} \equiv \left\langle \chi_i \middle| \hat{H}_{KS}(t) \middle| \chi_j \right\rangle$

5. Solve $H_{ik} d_{kj} = \in_j d_{ij}$ for d and $\in$

6. Compute $c_{ij}(t+\Delta t) = \sum_{kl} d^*_{jk} d_{lk} e^{-i\in_k \Delta t} c_{il}(t)$

7. Goto 1

Demagnetisation in Fe, Co and Ni

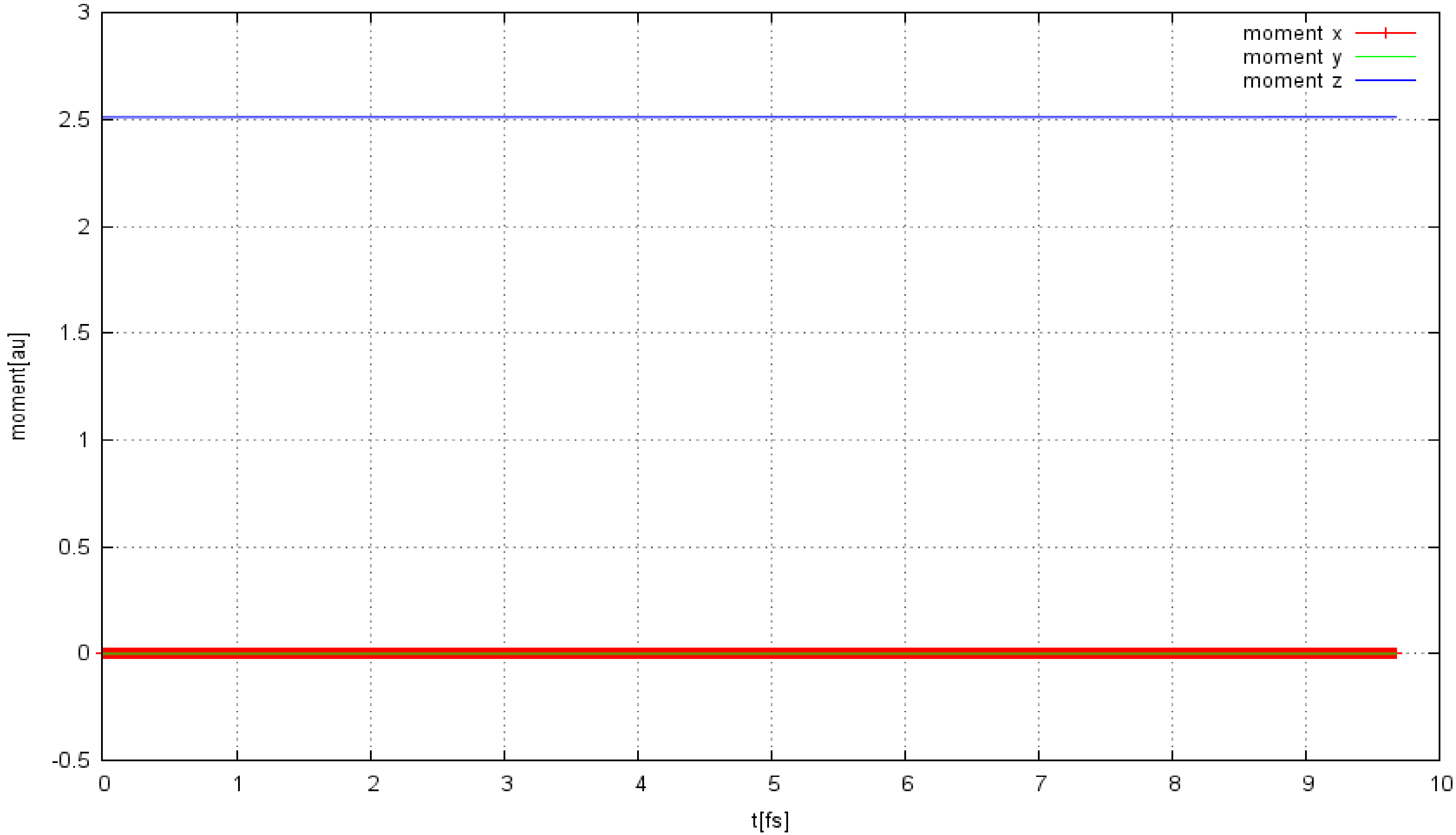


K. Krieger, K. Dewhurst, P. Elliott, S. Sharma, E.K.U.G., JCTC 11, 4870 (2015)

Analysis of the results

Calculation without spin-orbit coupling

components of spin moment



Exact equation of motion

$$\frac{\partial}{\partial t} M_z(t) = \frac{i}{\hbar}\left\langle \left[\hat{H}_{KS}, \hat{\sigma}_z \right] \right\rangle$$

$$= \int d^3r \left\{ m_x(r,t) B_{KS,y}(rt) - m_y(r,t) B_{KS,x}(rt) \right\}$$

$$+ \int d^3r \frac{1}{2c^2} \left\{ \hat{x} \cdot \left[\nabla v_s(r,t) \times j_y(r,t) \right] - \hat{y} \cdot \left[\nabla v_s(r,t) \times j_z(r,t) \right] \right\}$$

$\overleftrightarrow{j}(r,t) = \left\langle \hat{\sigma} \otimes \hat{p} \right\rangle$ spin current tensor

$$B_{KS}(rt) = B_{ext}(rt) + B_{XC}(rt)$$

Exact equation of motion

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Global torque exerted by B_{KS}

$$= \int d^3r \left\{ m_x(r,t) B_{KS,y}(rt) - m_y(r,t) B_{KS,x}(rt) \right\}$$

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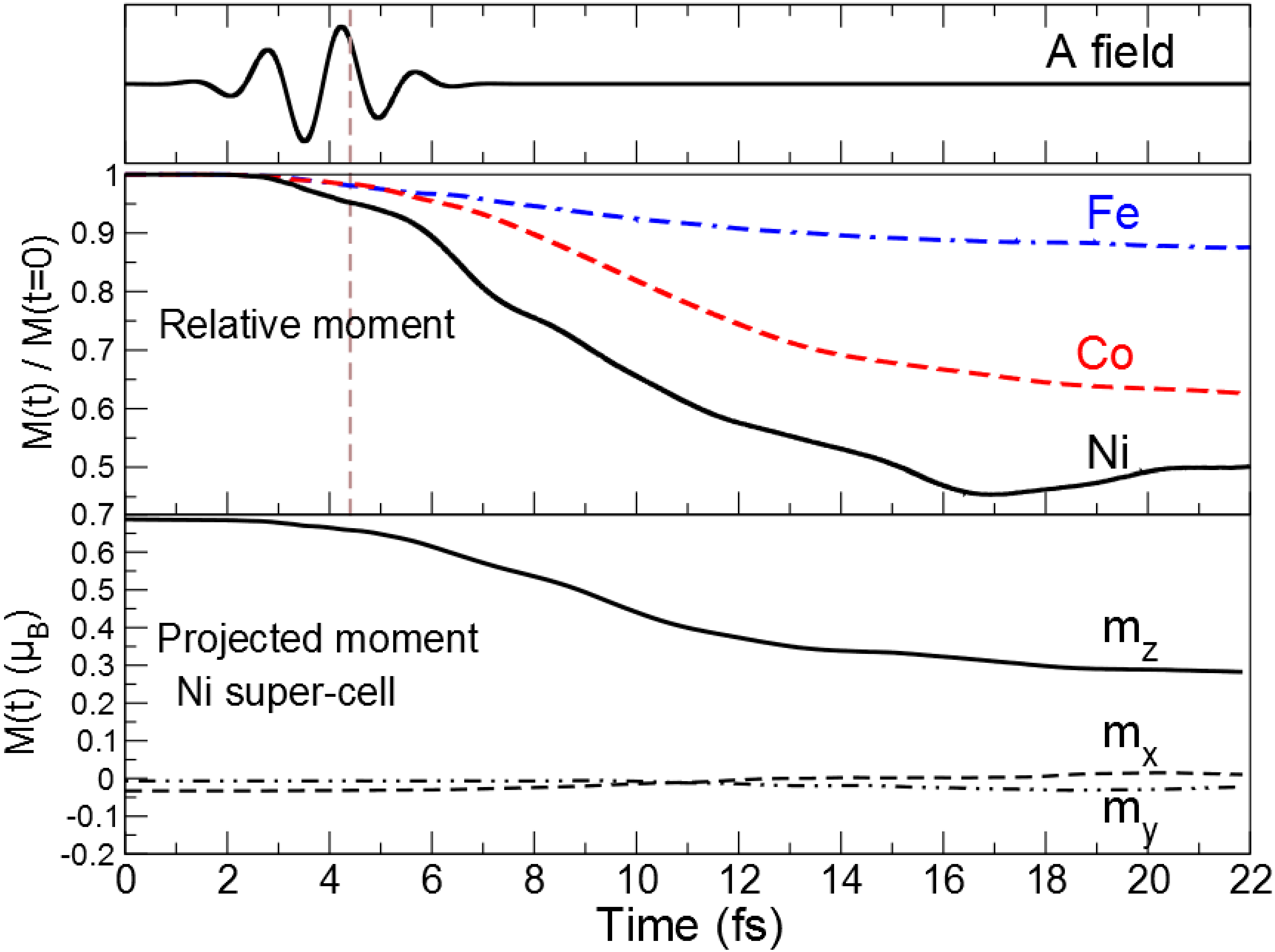
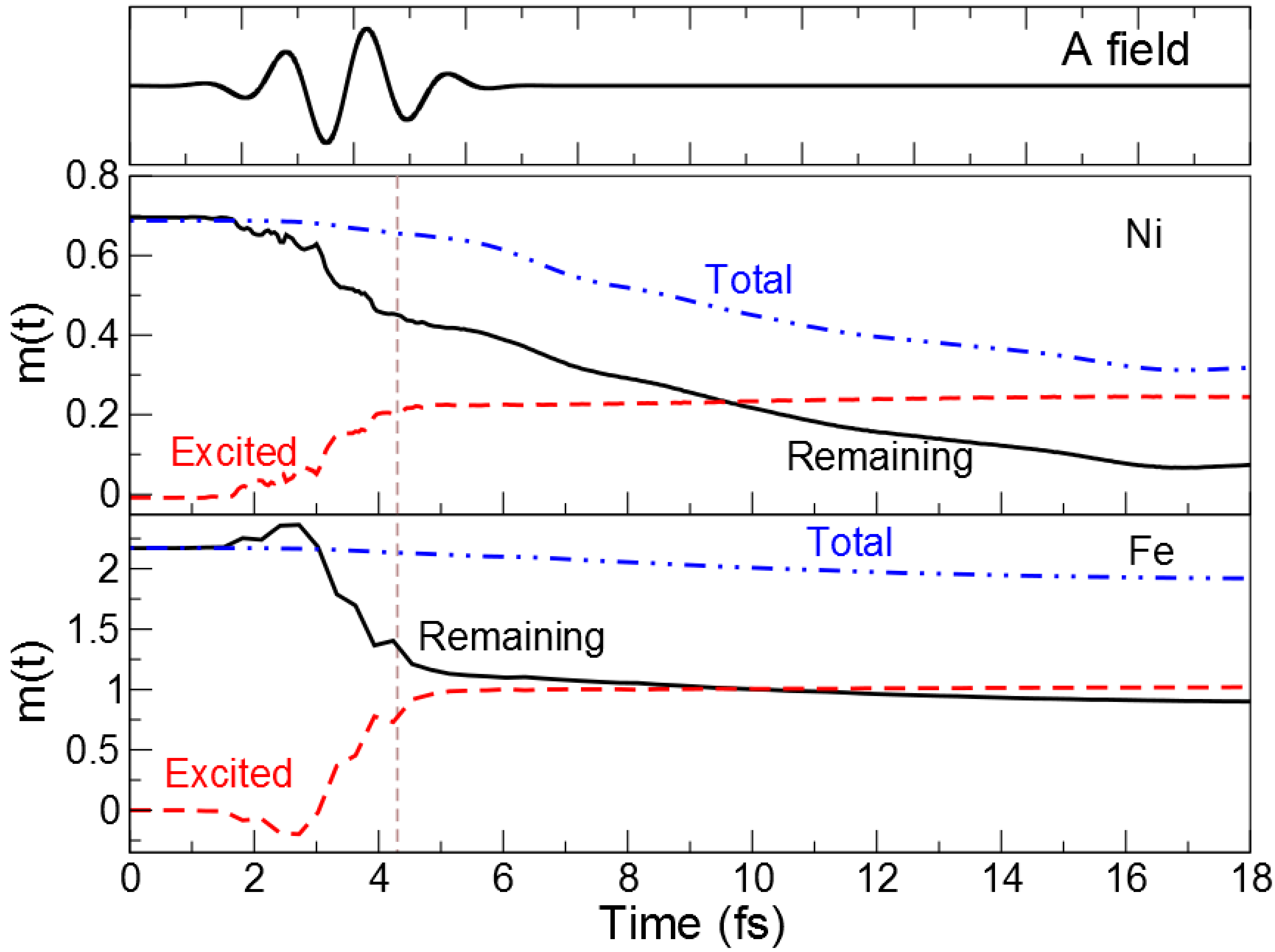
Global torque = 0, if $B_{ext} = 0$
(due to zero-torque theorem for B_{xc})



<u>Note: Ground state of bulk Fe, Co, Ni is collinear</u>



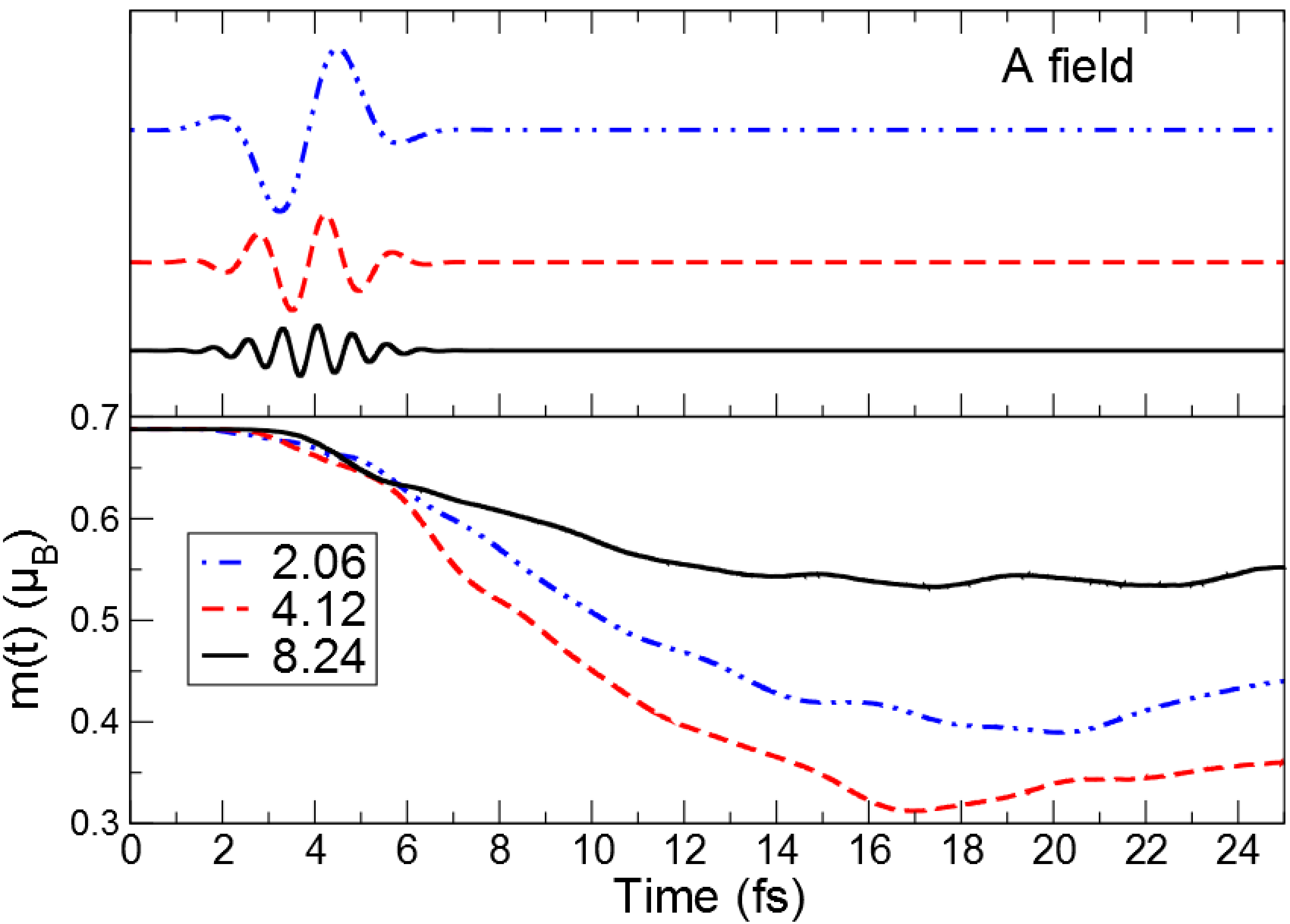
A field
0.8
0.6
0.4
0.2
0
m(t)
Ni
Total
Excited
Remaining
Total
Fe
2
1.5
1
0.5
0
m(t)
Remaining
Excited
0
2
4
6
8
10
12
14
16
18
Time (fs)

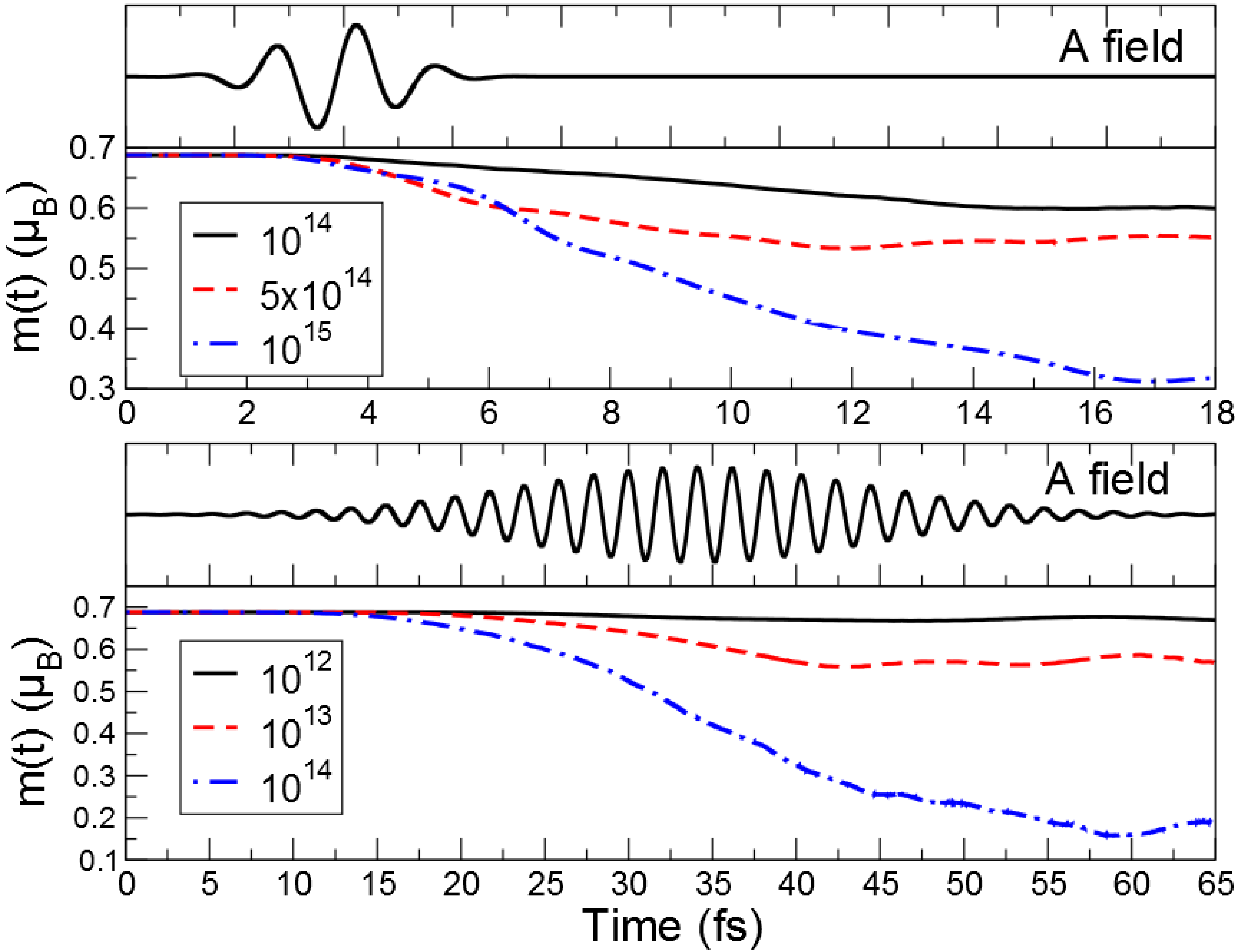
Demagnetization occurs in two steps:

- Initial excitation by laser *moves* magnetization from atomic region into interstitial region. Total Moment is basically conserved during this phase.

- Spin-Orbit term drives demagnetization of the more localized electrons until stabilization at lower moment is achieved

Playing with laser parameters


A field
2.06
4.12
8.24
m(t) (μ_B)
0.3
0.4
0.5
0.6
0.7
0
2
4
6
8
10
12
14
16
18
20
22
24
Time (fs)


A field
0.7
0.6
0.5
0.4
0.3
m(t) (μ_B)
10^{14}
$5x10^{14}$
10^{15}
0
2
4
6
8
10
12
14
16
18
A field
0.7
0.6
0.5
0.4
0.3
0.2
0.1
m(t) (μ_B)
10^{12}
10^{13}
10^{14}
0
5
10
15
20
25
30
35
40
45
50
55
60
65
Time (fs)

Influence of approximation for xc functional

The four steps of any functional theory

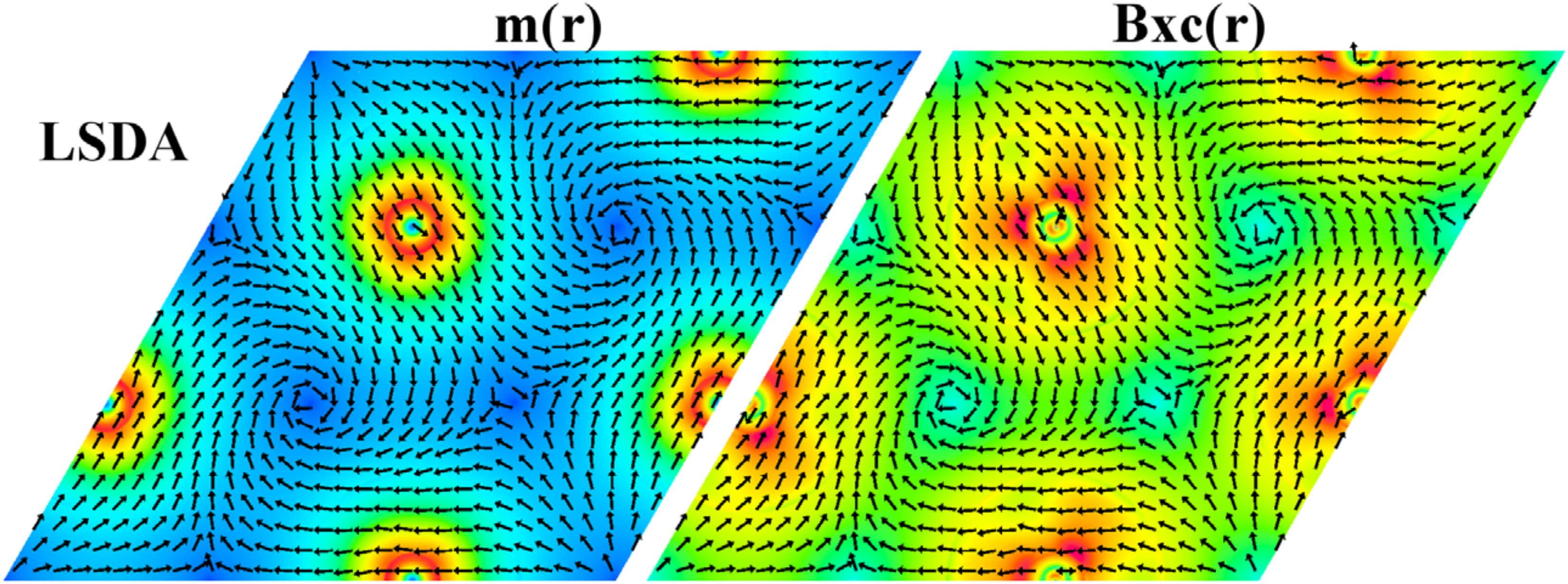
Step 1: Basic Theorems (Hohenberg-Kohn-Sham/ Runge-Gross)

Step 2: Find approximate functionals for $v_{xc}\left[\rho\left(r't'\right)\right]\left(rt\right)$

Step 3: Write code that solves the KS equations efficiently

Step 4: Run code for interesting systems/questions

Problem: In all standard approximations of E_{xc} (LSDA, GGAs) m(r) and B_{xc}(r) are locally parallel



S. Sharma, J.K. Dewhurst, C. Ambrosch-Draxl, S. Kurth, N. Helbig, S. Pittalis, S. Shallcross, L. Nordstroem E.K.U.G., Phys. Rev. Lett. 98, 196405 (2007)

Why is that important?

Ab-initio description of spin dynamics:

microscopic equation of motion (following from TDSDFT)

$$\dot{\vec{m}}(\vec{r},t) = \vec{m}(\vec{r},t)\times\vec{B}_{XC}(\vec{r},t) - \vec{\nabla}\cdot\overleftrightarrow{J}_S(\vec{r},t) + \text{SOC}$$

in absence of external magnetic field

Consequence of local collinearity: $m \times B_{xc} = 0$:
→ possibly wrong spin dynamics
→ how important is this term in real-time dynamics?

Construction of a novel GGA-type functional

Traditional LSDA: Start from uniform electron gas in collinear magnetic state. Determine $e_{XC}[n,m]$ from QMC or MBPT and parametrize $e_{XC}[n,m]$ to use in LSDA.

New non-collinear functional: Start from spin-spiral phase of e-gas. Determine $e_{XC}[n,\vec{m}]$ from MBPT and parametrize $e_{XC}[n,\vec{m}]$ to use as non-collinear GGA.

F.G. Eich and E.K.U. Gross, Phys. Rev. Lett. 111, 156401 (2013)

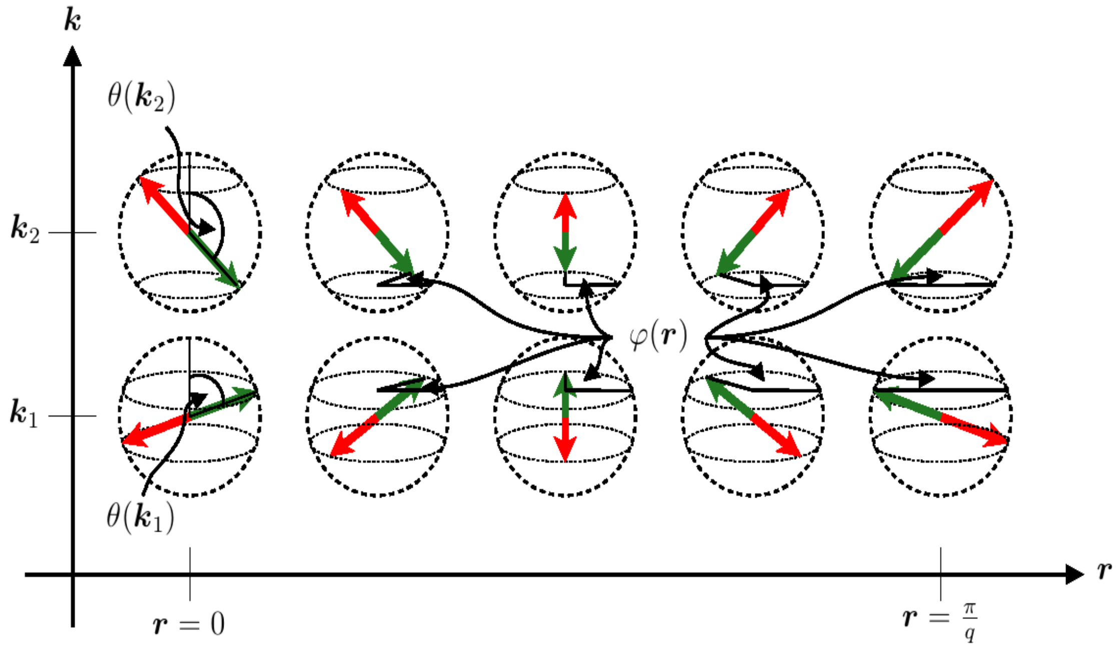


Illustration of spin spiral waves along one spatial coordinate for two different choices of wavevector $q=k_{1/2}$.

Magnetisation of a spin-spiral state in the uniform electron gas

$$\mathrm{m}(\mathbf{r}) = \mathrm{m}\begin{pmatrix} \mathrm{s}\cos(\mathbf{q}\cdot\mathbf{r}) \\ \mathrm{s}\sin(\mathbf{q}\cdot\mathbf{r}) \\ \sqrt{1-\mathrm{s}^2} \end{pmatrix}$$

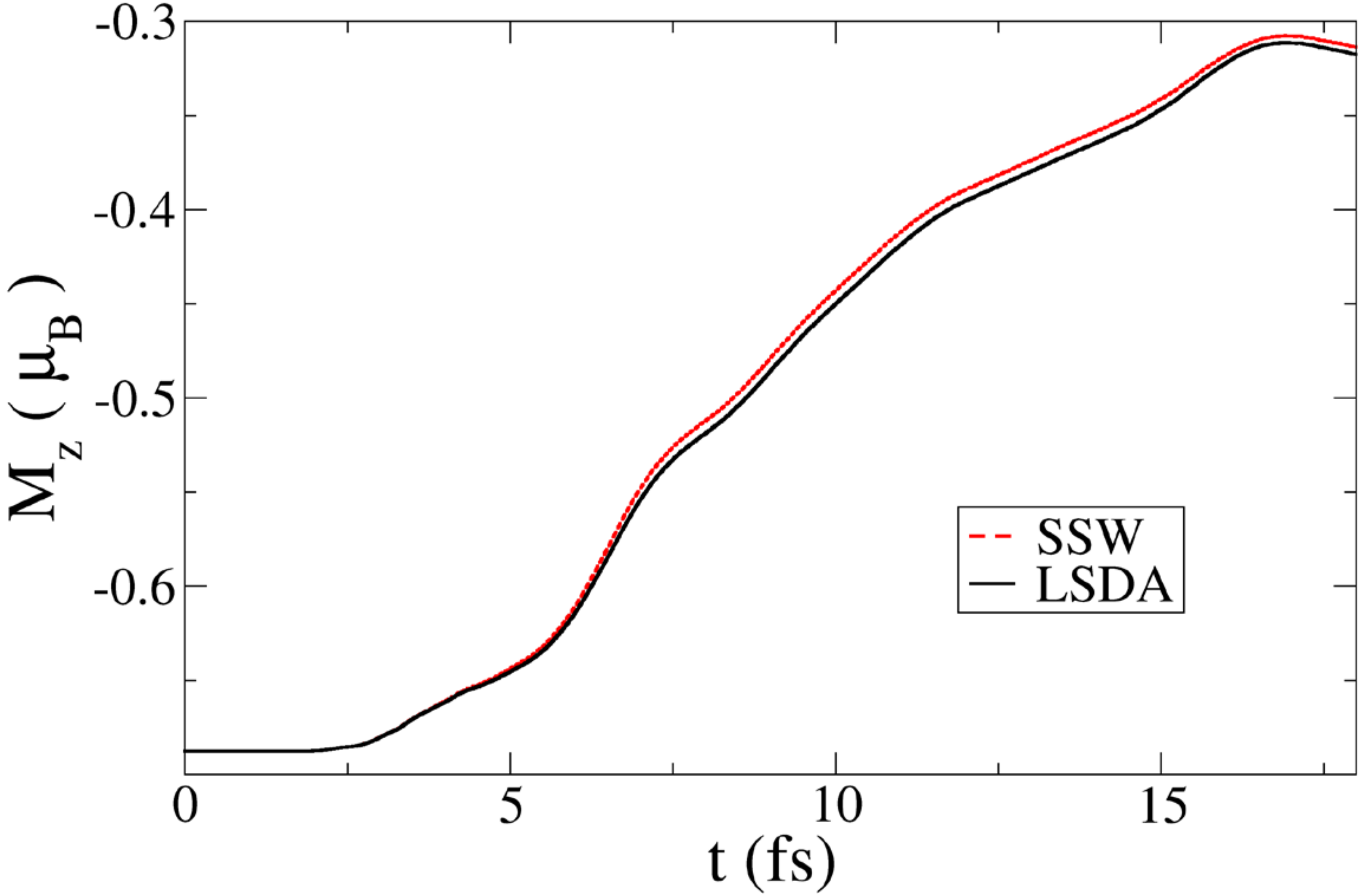
$$\varepsilon_{\mathrm{xc}}^{\mathrm{SSW}} = \varepsilon_{\mathrm{xc}}^{\mathrm{SSW}}(\mathrm{n},\mathrm{m},\mathrm{q},\mathrm{s})$$

$$E_{xc}^{GGA}[n,\vec{m}] = \int d^3r\, n(\mathbf{r})\, \varepsilon_{xc}^{SSW}\left(n(\mathbf{r}), m(\mathbf{r}), q(\mathbf{r}), s(\mathbf{r})\right)$$

$$s^2(\mathbf{r}) = \frac{D_T^2(\mathbf{r})}{D_T^2(\mathbf{r}) + m^4(\mathbf{r})\, d_T(\mathbf{r})} \qquad q^2(\mathbf{r}) = \frac{D_T^2(\mathbf{r}) + m^4(\mathbf{r})\, d_T(\mathbf{r})}{m^4(\mathbf{r})\, D_T(\mathbf{r})}$$

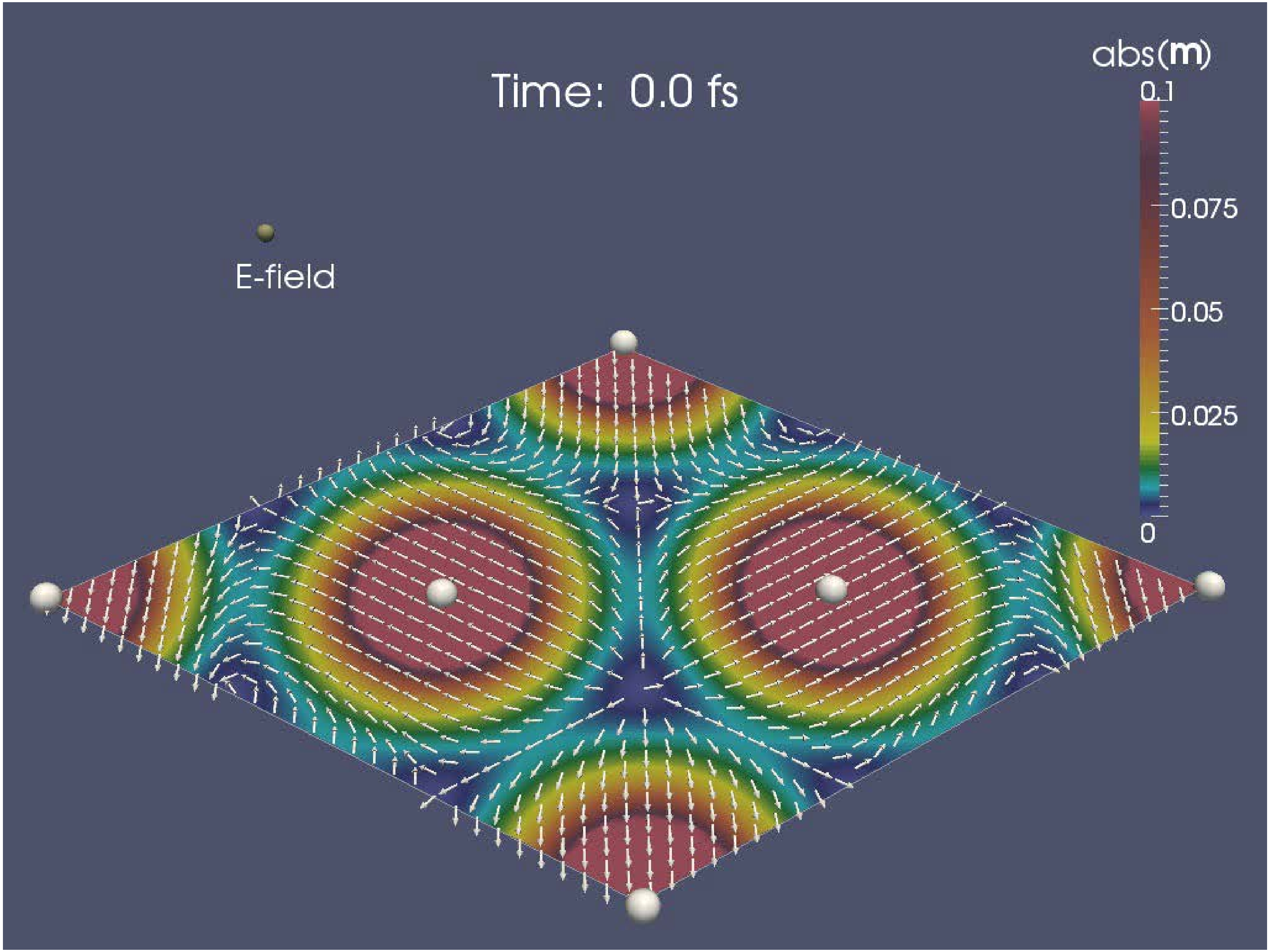
$$D_T(\mathbf{r}) = \left|\vec{m}(\mathbf{r}) \times \left(\nabla \otimes \vec{m}(\mathbf{r})\right)\right|^2 \qquad d_T(\mathbf{r}) = \left|\vec{m}(\mathbf{r}) \times \left(\nabla^2 \vec{m}(\mathbf{r})\right)\right|^2$$

F.G. Eich and E.K.U. Gross, Phys. Rev. Lett. 111, 156401 (2013)

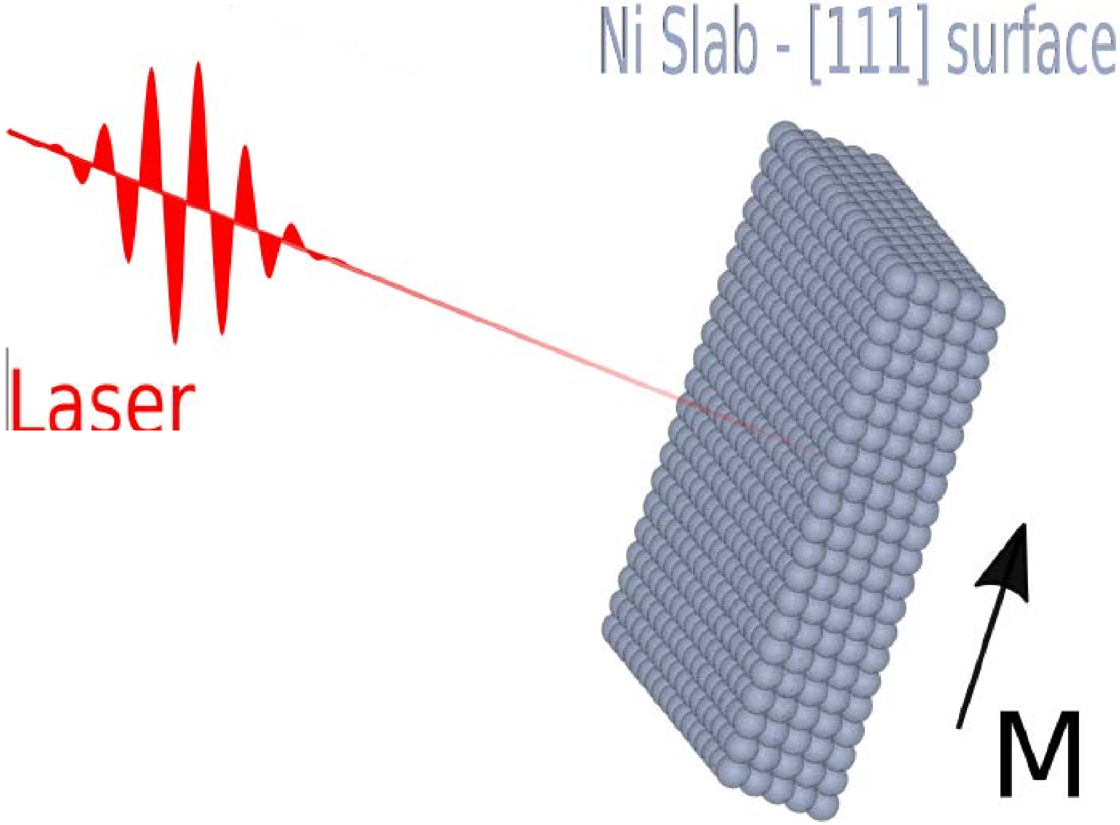


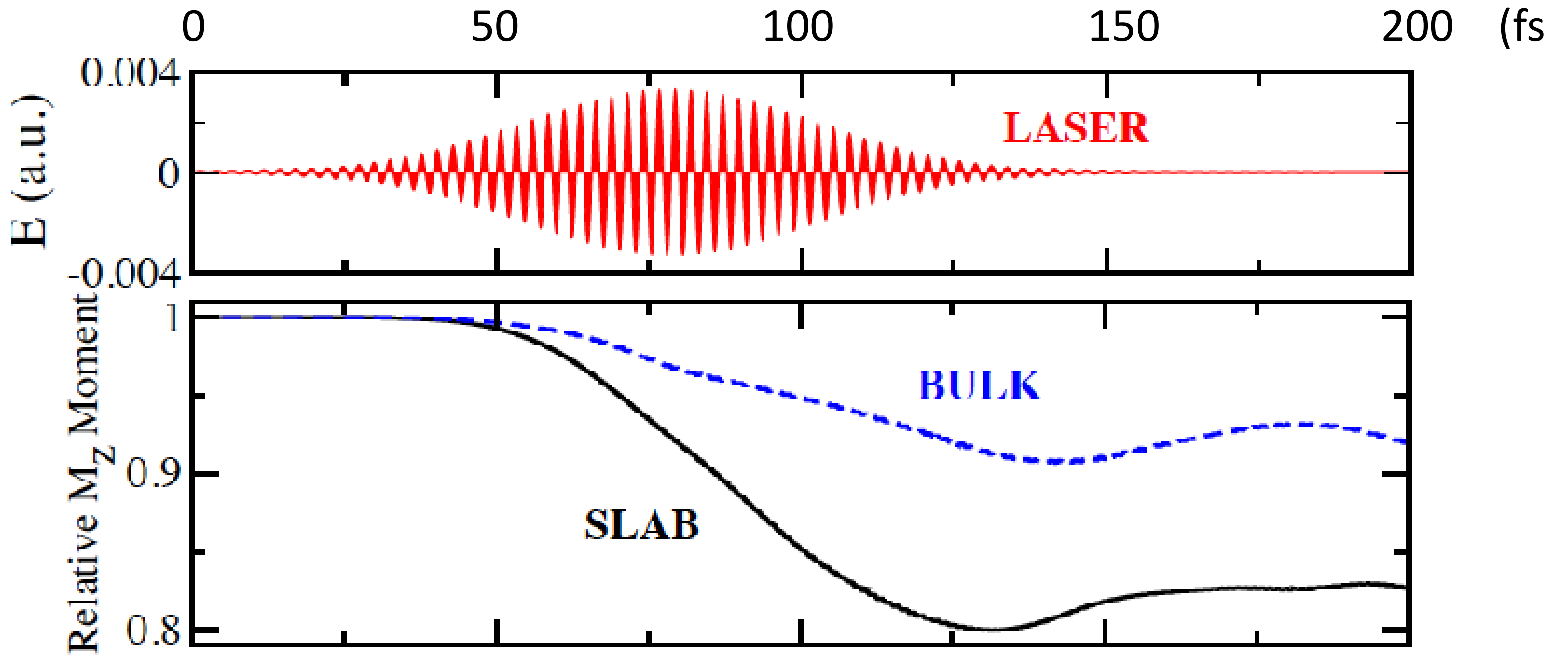
-0.3
-0.4
-0.5
-0.6
0
5
10
15
t (fs)
M_z (μ_B)
SSW
LSDA

Beyond 3D bulk

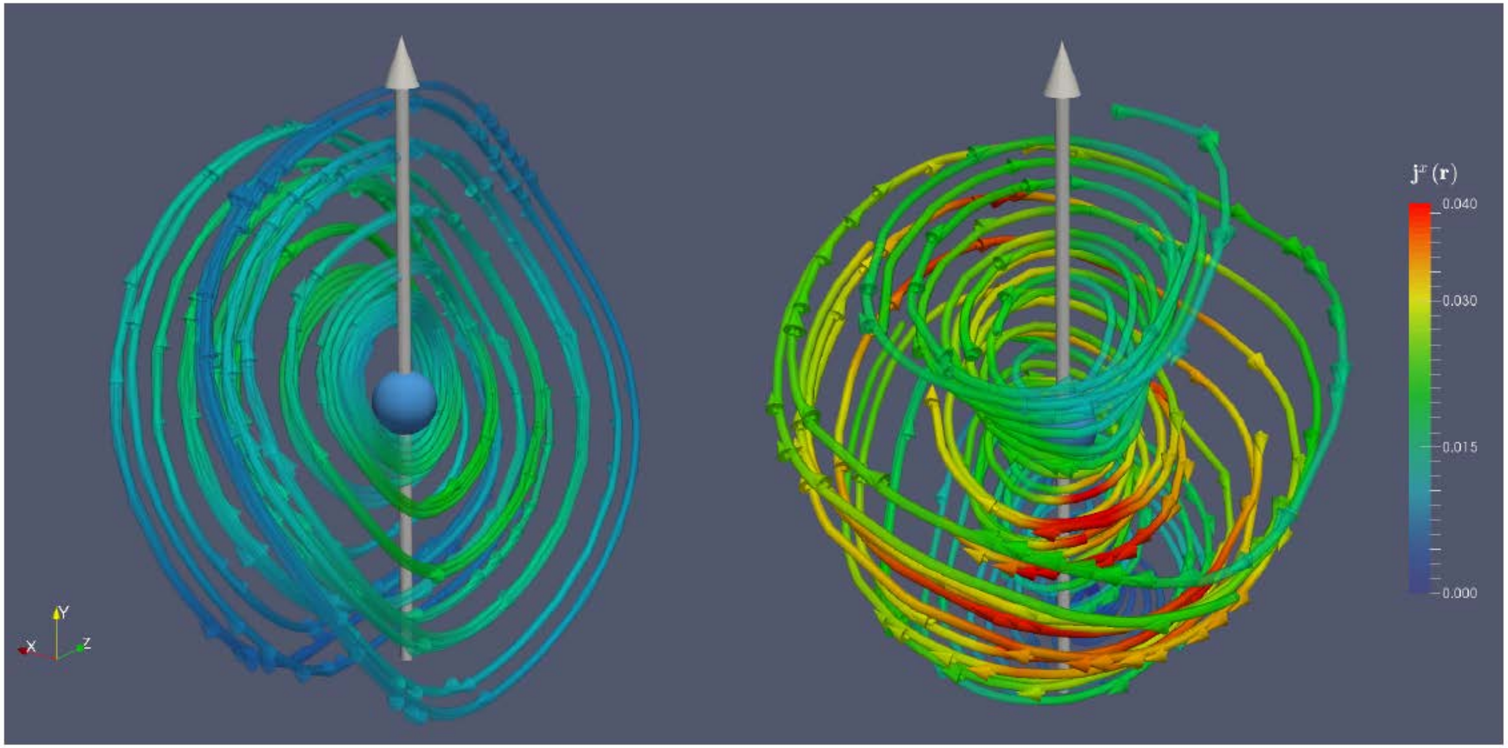


Cr monolayer

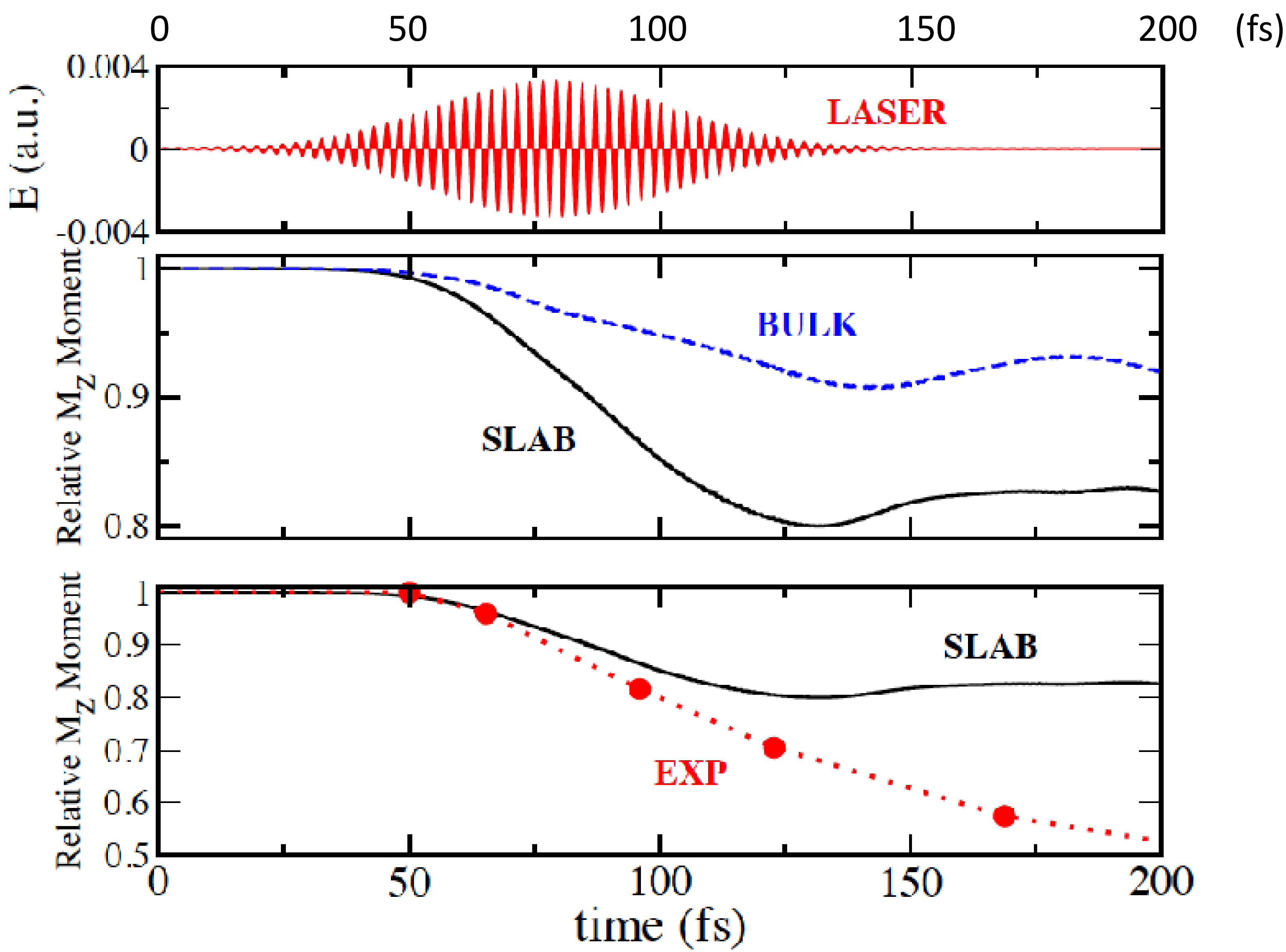

Ni Slab - [111] surface
Laser
M



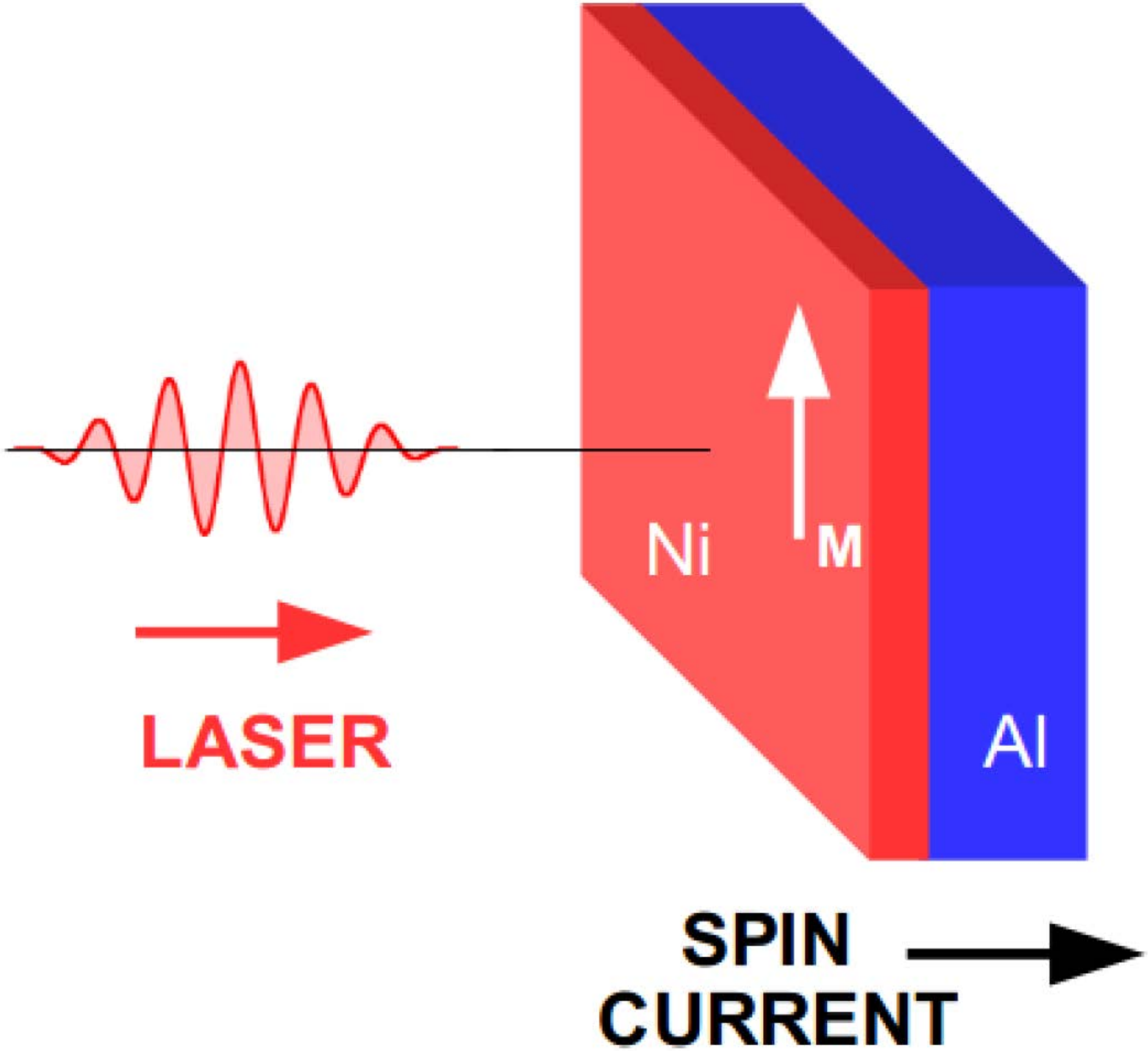
0
50
100
150
200
(fs)
0.004
0
-0.004
E (a.u.)
LASER
1
0.9
0.8
Relative M_Z Moment
BULK
SLAB



Streamlines for J_x, the spin-current vector field of the x component of spin, around a Ni atom in bulk (left) and for the outermost Ni atom in the slab (right).

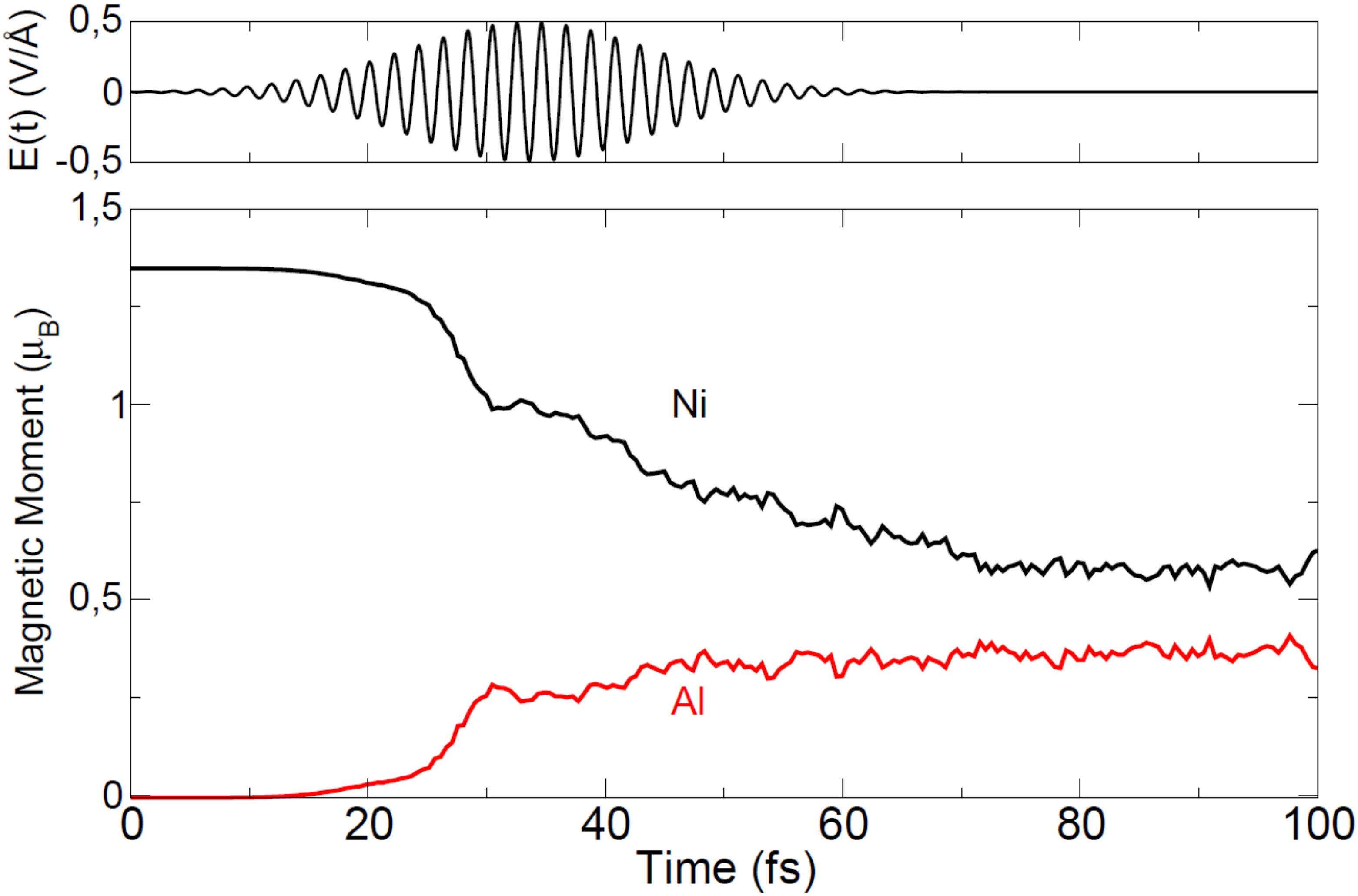


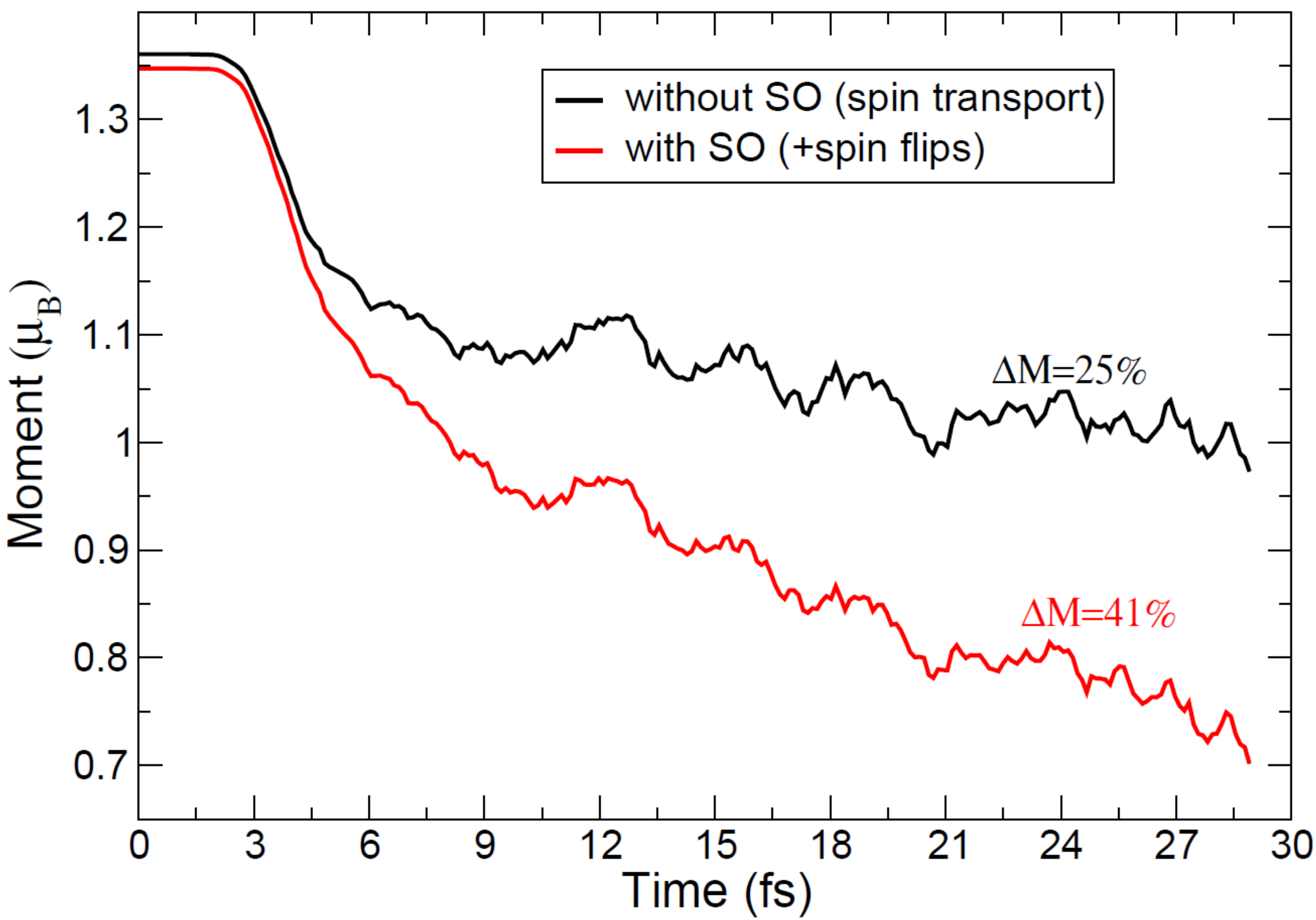
0
50
100
150
200
(fs)
0.004
0
-0.004
E (a.u.)
LASER
1
0.9
0.8
Relative M_Z Moment
BULK
SLAB
1
0.9
0.8
0.7
0.6
0.5
Relative M_Z Moment
SLAB
EXP
0
50
100
150
200
time (fs)



Ni
M
Al
LASER
SPIN
CURRENT

Effect of spin transport across interfaces: Ni@Al

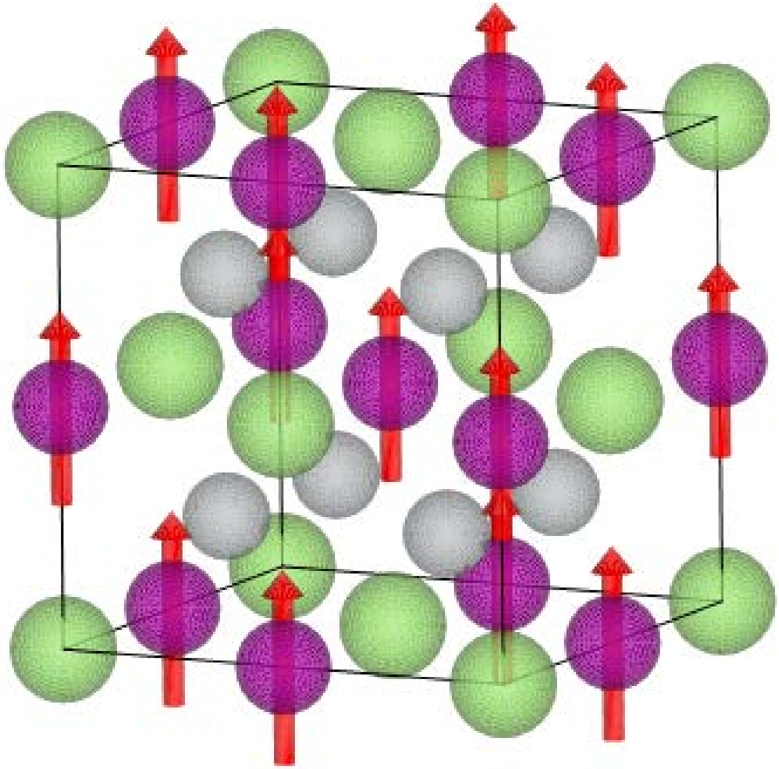




without SO (spin transport)
with SO (+spin flips)
ΔM=25%
ΔM=41%
Moment (μB)
1.3
1.2
1.1
1
0.9
0.8
0.7
0
3
6
9
12
15
18
21
24
27
30
Time (fs)

Heusler compounds

Ni_2MnGa

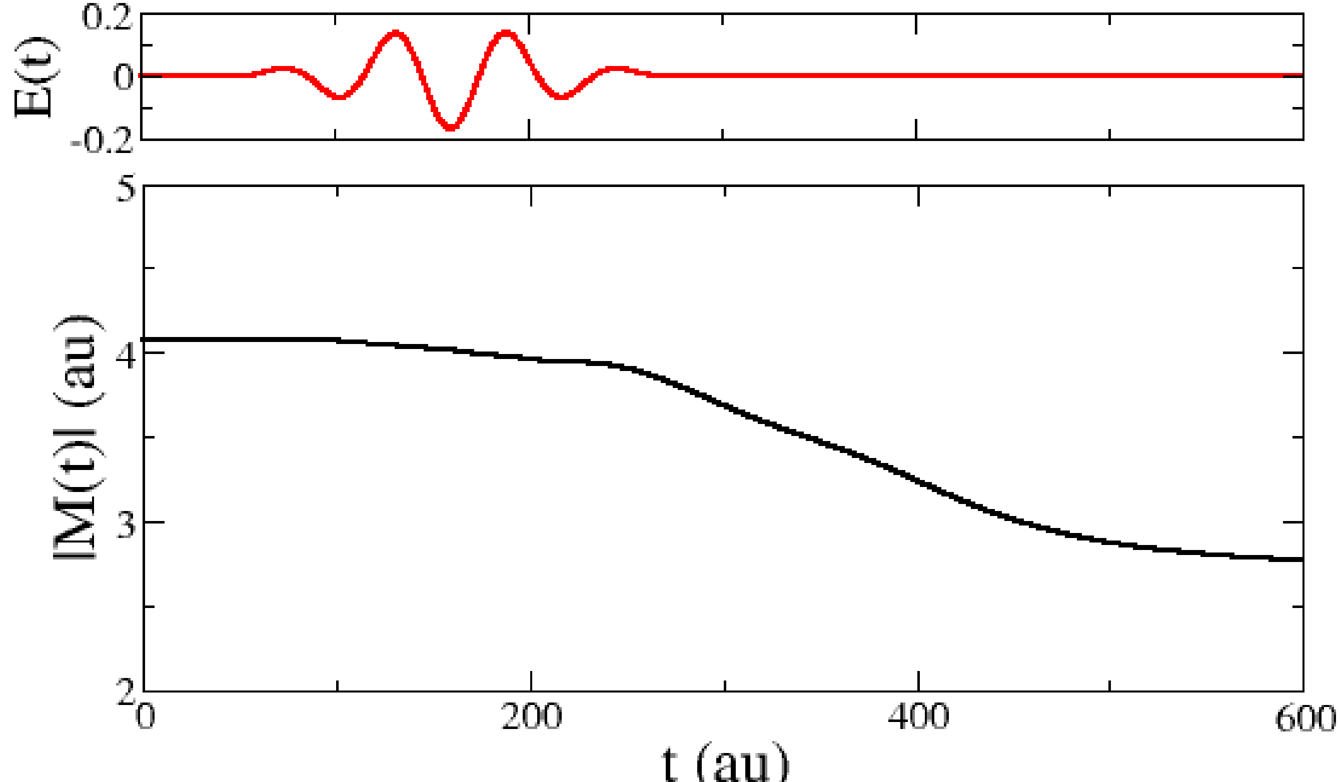


Ga	0.02 μB
Mn	-3.14 μB
Ni	-0.37 μB

Ni_2MnGa

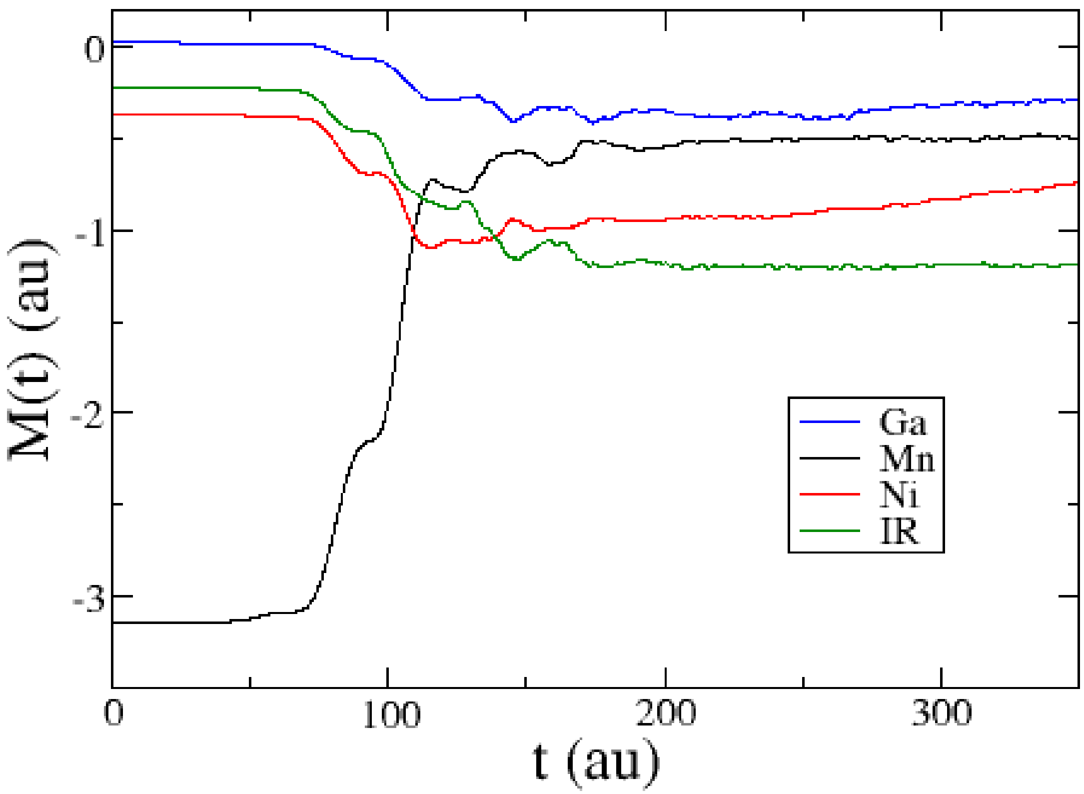
Laser parameters: ω=2.72eV Ipeak= 1x1015 W/cm2 J = 935 mJ/cm2 FWHM = 2.42 fs

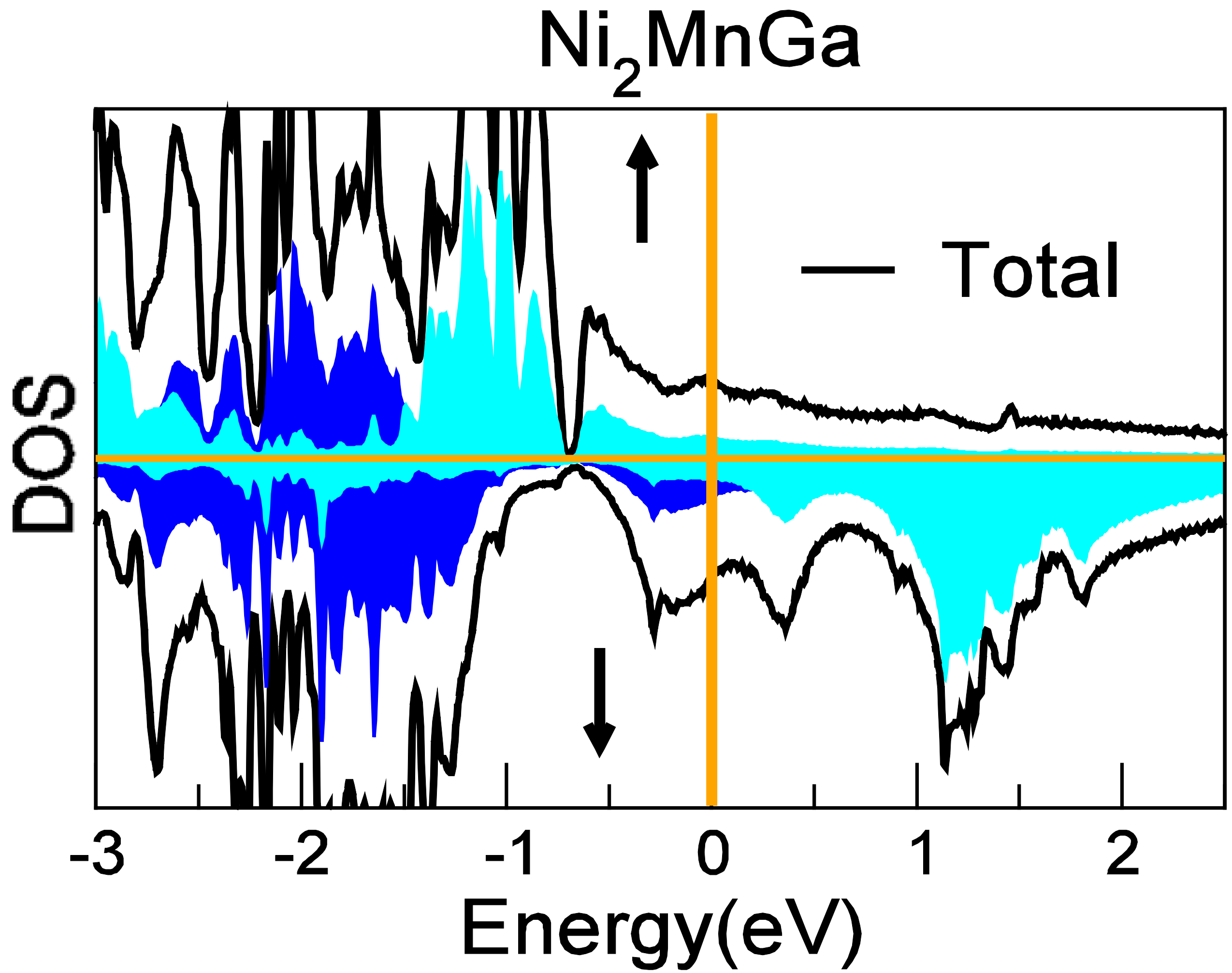
Loss in global moment, as usual



Ni_2MnGa

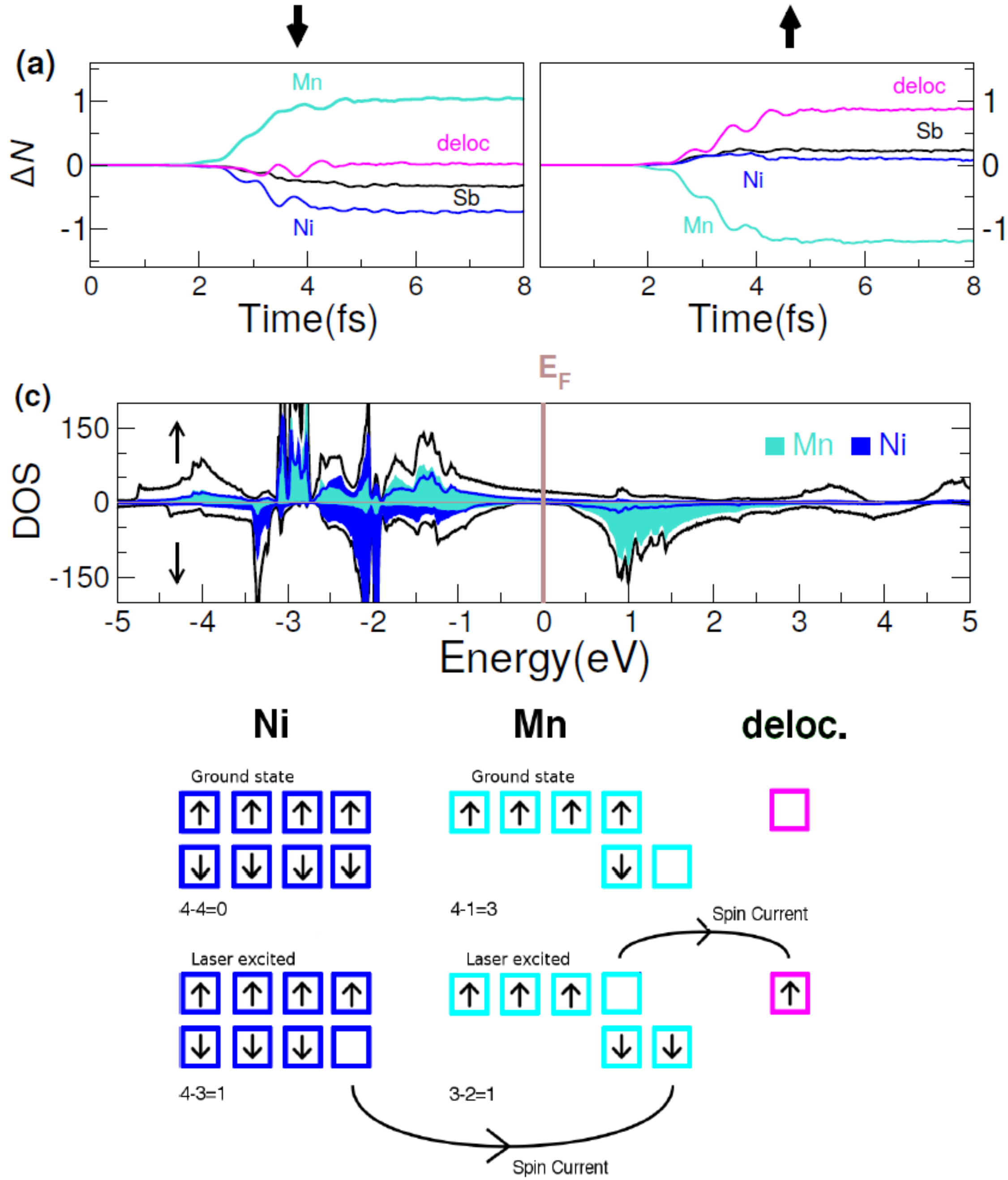
Change in local moments
Transfer of moment from Mn to Ni (does not require SOC)
Followed by spin-orbit mediated demagnetization, mainly on Ni



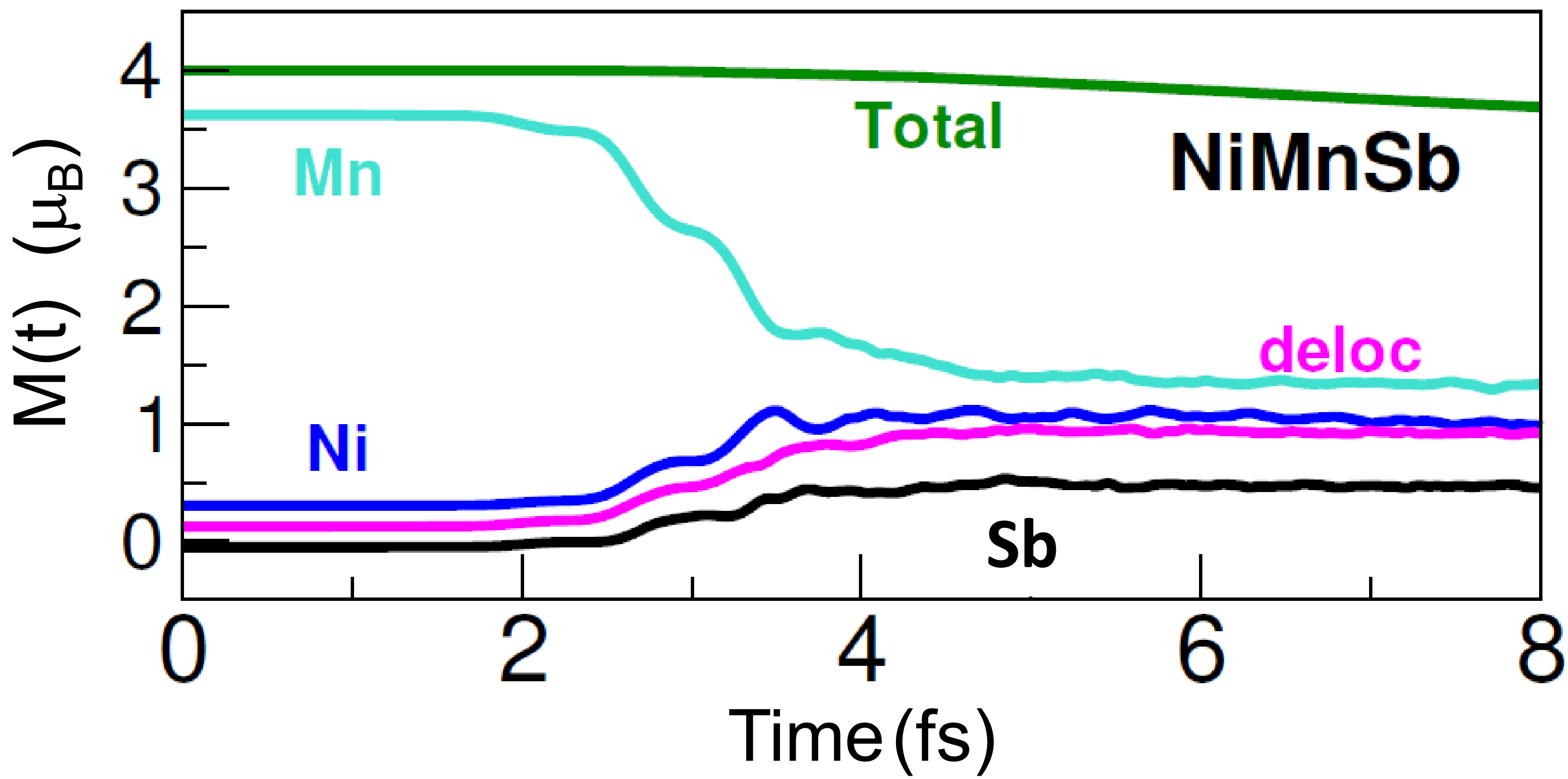


P. Elliott, T. Mueller, K. Dewhurst, S. Sharma, E.K.U.G., Scientific Reports 6, 38911 (2016)

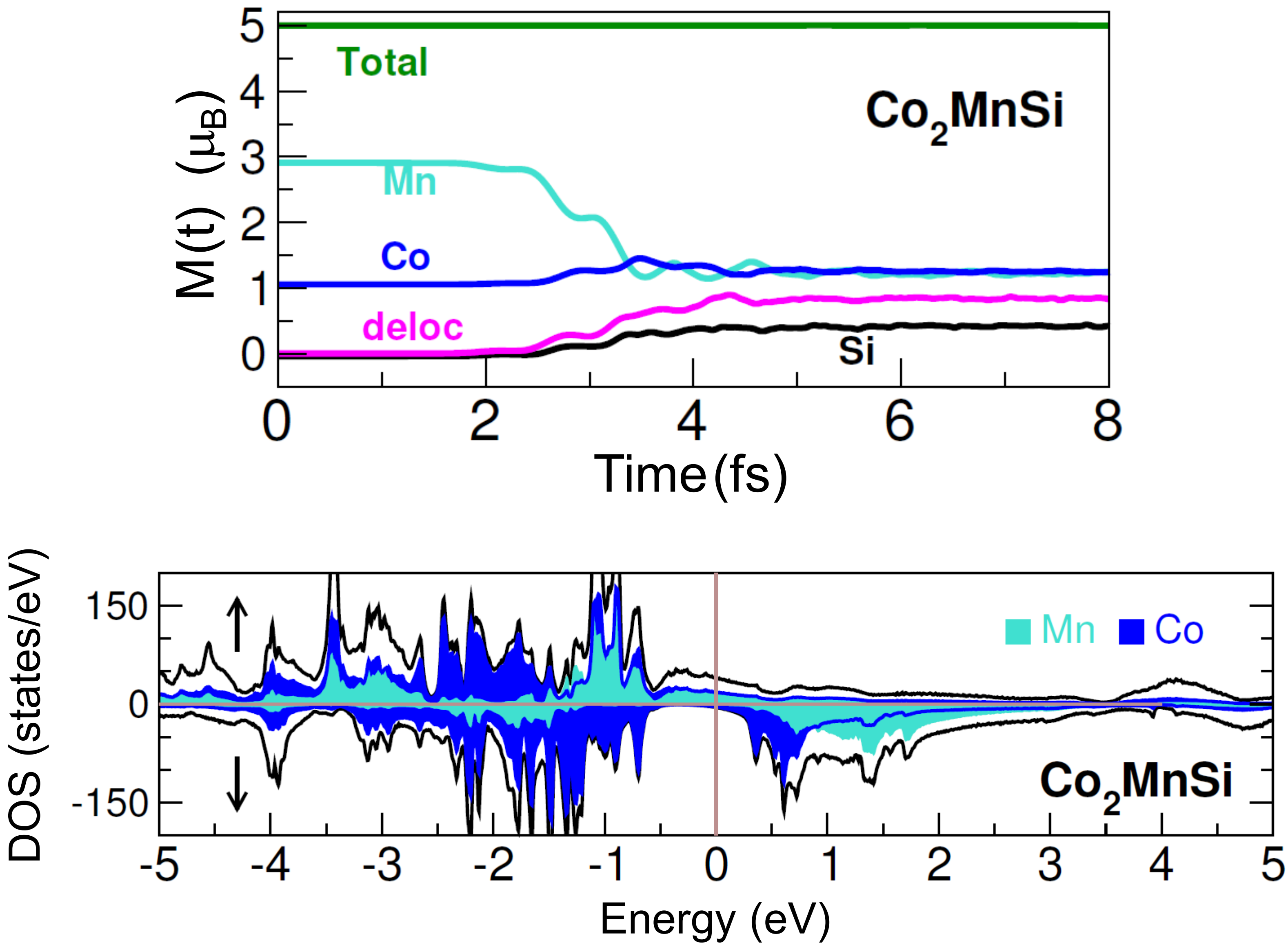
NiMnSb



(a)
Mn
deloc
Sb
Ni
ΔN
Time(fs)
(c)
E_F
DOS
Mn
Ni
Energy(eV)
Ni
Mn
deloc.
Ground state
4-4=0
4-1=3
Spin Current
Laser excited
4-3=1
3-2=1
Spin Current

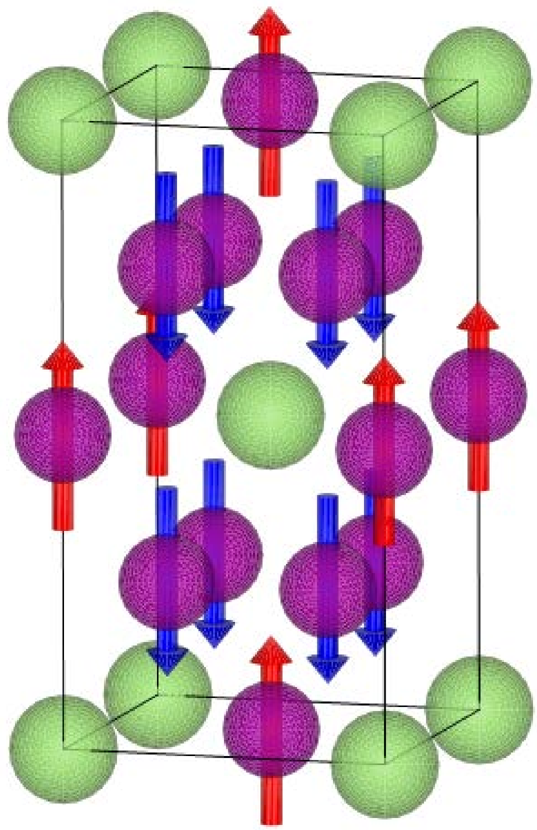


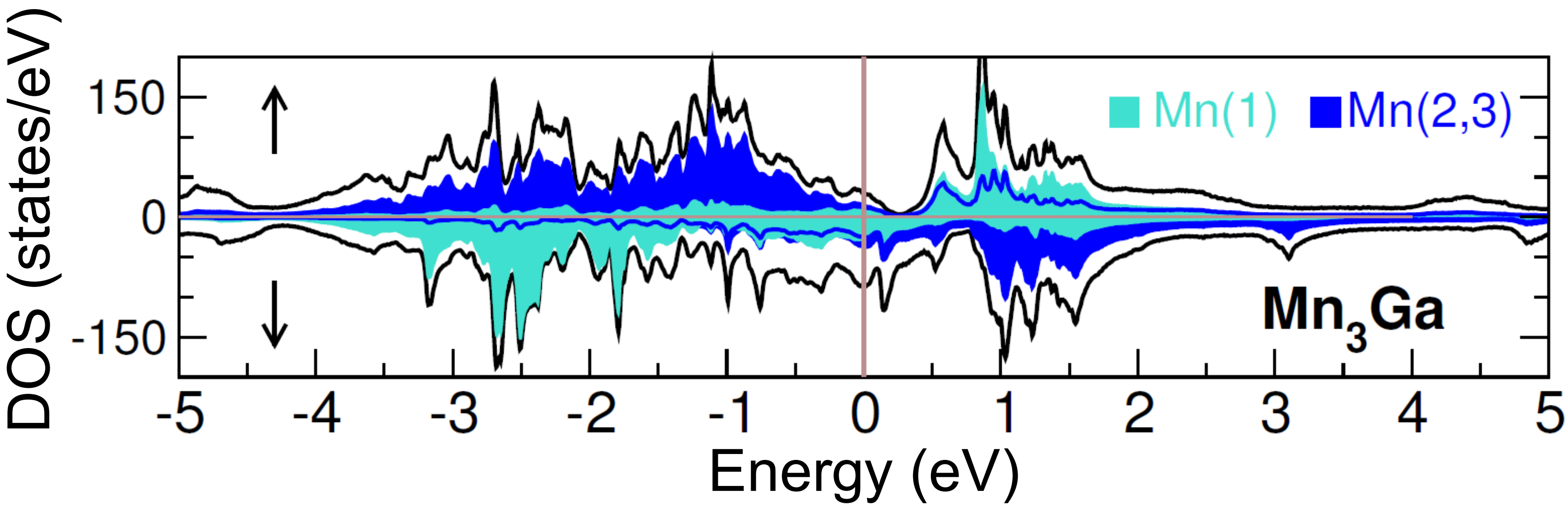
Total
NiMnSb
Mn
deloc
Ni
Sb
M(t) (μB)
Time (fs)
0
1
2
3
4
0
2
4
6
8



M(t) (μB)
Total
Co2MnSi
Mn
Co
deloc
Si
0
1
2
3
4
5
0
2
4
6
8
Time (fs)
DOS (states/eV)
150
0
-150
Mn
Co
Co2MnSi
-5
-4
-3
-2
-1
0
1
2
3
4
5
Energy (eV)

Mn_3Ga



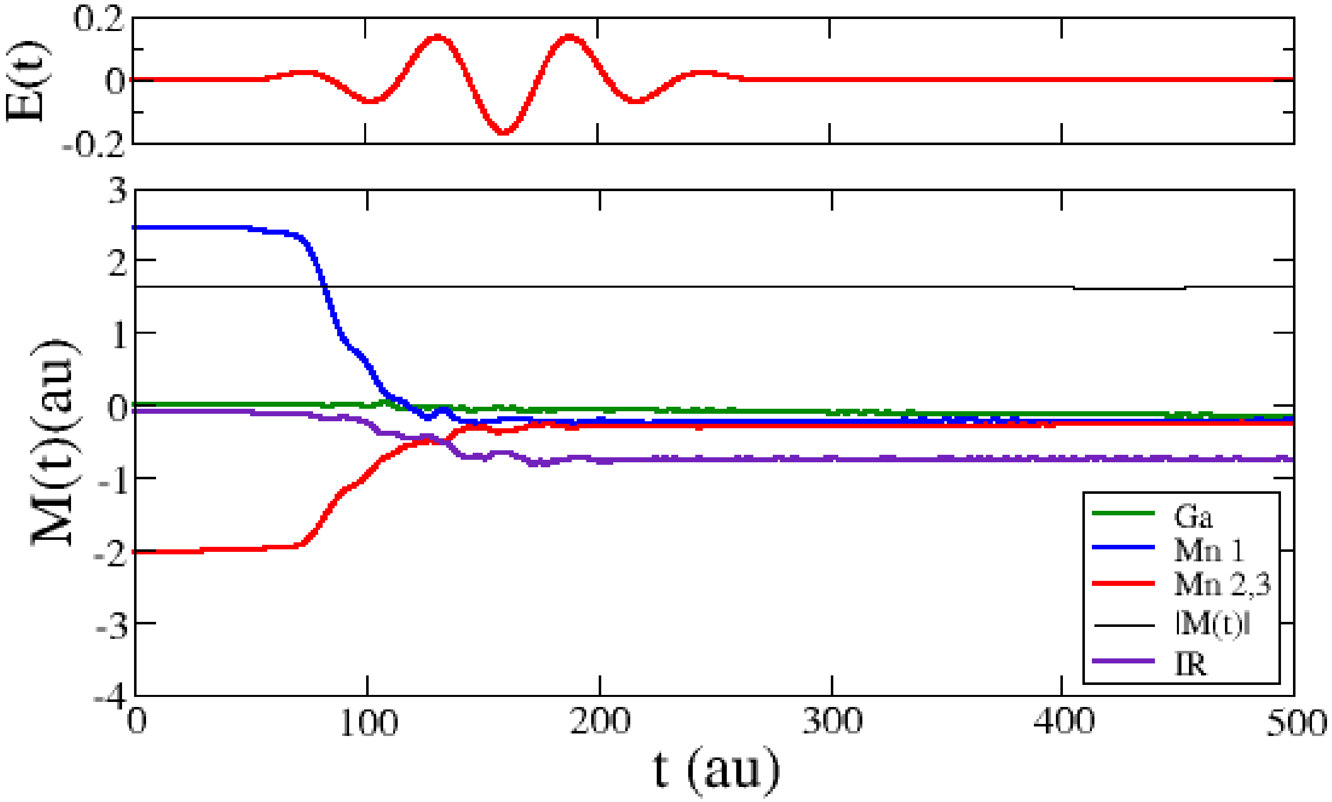


DOS (states/eV)
150
0
-150
↑
↓
Mn(1)
Mn(2,3)
Mn3Ga
-5
-4
-3
-2
-1
0
1
2
3
4
5
Energy (eV)

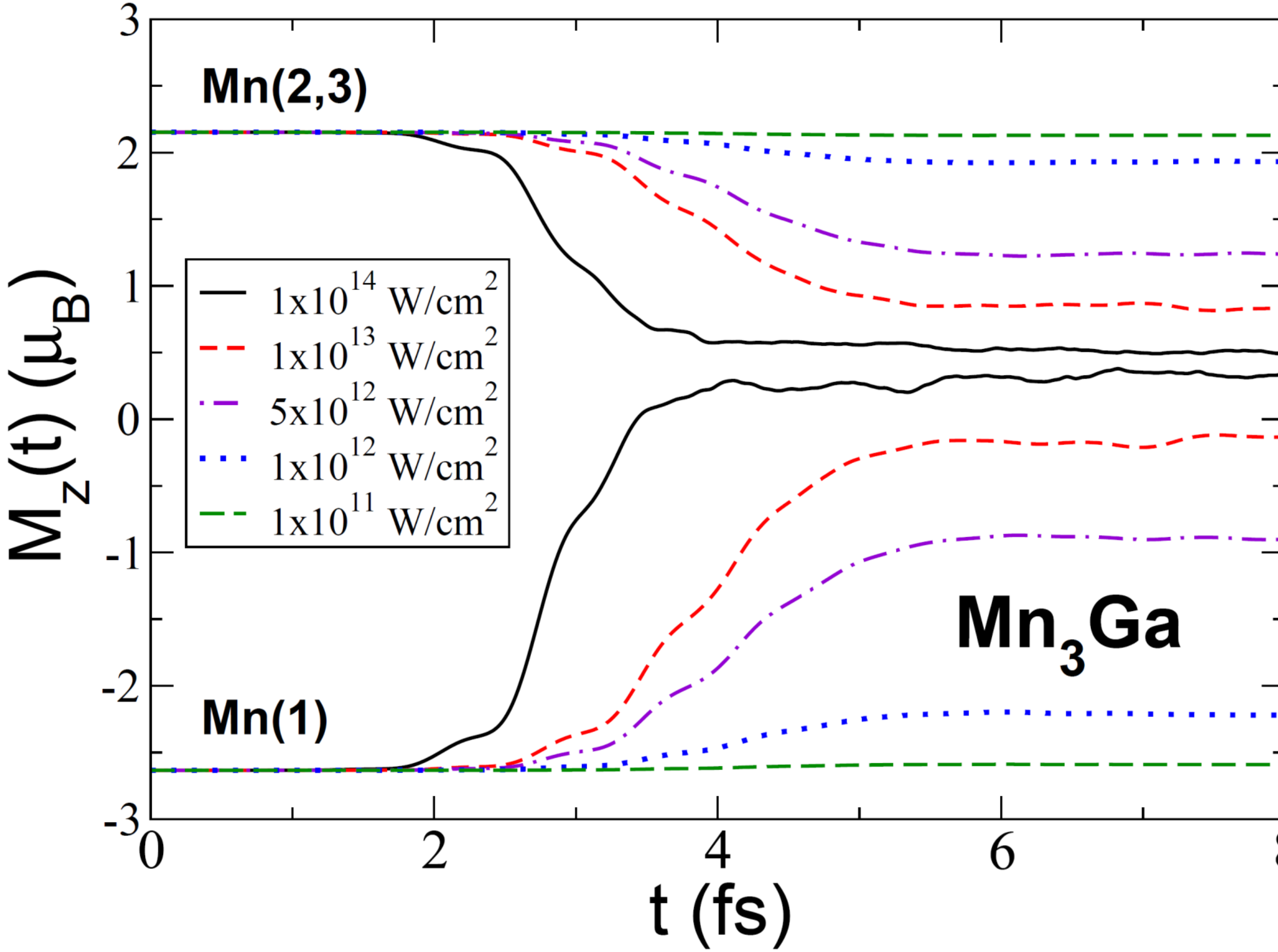
Mn_3Ga

Laser parameters: ω=2.72eV Ipeak= 1x1015 W/cm2 J = 935 mJ/cm2 FWHM = 2.42 fs

Global moment |M(t)| preserved
Local moment on each sublattice reduced



P. Elliott, T. Mueller, K. Dewhurst, S. Sharma, E.K.U.G., Scientific Reports 6, 38911 (2016)



Mn(2,3)
Mn(1)
Mn_3Ga
1x10^14 W/cm^2
1x10^13 W/cm^2
5x10^12 W/cm^2
1x10^12 W/cm^2
1x10^11 W/cm^2
$M_z(t)$ (μ_B)
t (fs)
3
2
1
0
-1
-2
-3
0
2
4
6
8

Summary

- Real-time TDDFT implemented in an all-electron solid state code (http://elk.sourceforge.net/)

- Demagnetization in first 50 fs is a **universal** two-step process:
    1. Initial excitation of electrons into highly excited delocalised states (without much of a change in the total magnetization)
    2. Spin-orbit coupling drives demagnetization of the more localized electrons

- No significant change in M_x and M_y in bulk Fe, Co, Ni

- Interfaces show spin currents as important as spin-orbit coupling

- Ultrafast (3-5 fs) transfer of spin moment between sublattices of Heusler compounds by **purely optical** excitation: Easily understood on the basis of the ground-state DOS

Coworkers

Kevin Krieger

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Kay Dewhurst

Peter Elliott

Future:

- **Include relaxation processes due to el-el scattering**
  - in principle contained in TDDFT,
  - but not with adiabatic xc functionals
  - need xc functional approximations with memory $v_{xc}\left[\rho(r't')\right](rt)$
- **Include relaxation processes due to el-phonon scattering**
- **Include relaxation due to radiative effects**
  simultaneous propagation of TDKS and Maxwell equations
- **Include dipole-dipole interaction to describe motion of domains**
  construct approximate xc functionals which refer to the dipole int
- **Optimal-control theory to find optimized laser pulses**
  to selectively demagnetize/remagnetize, i.e. to switch,
  the magnetic moment
- **Create Skyrmions with suitably shaped laser pulses**