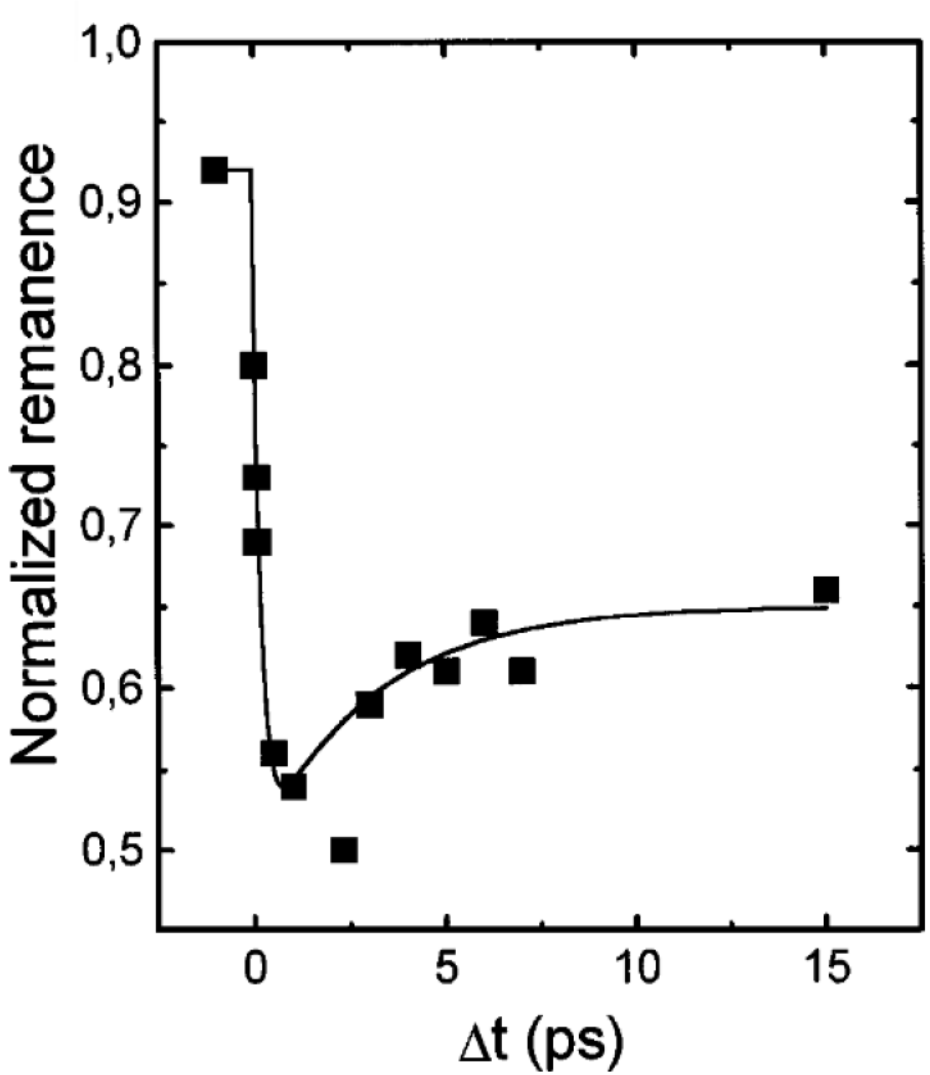
Ultrafast laser-driven spin dynamics in antiferromagnetic and ferromagnetic solids: Ab-initio studies with real-time TDDFT

E.K.U. Gross

Max-Planck Institute of Microstructure Physics Halle (Saale)

First experiment on ultrafast laser induced demagnetization



Beaurepaire et al, PRL 76, 4250 (1996)

<u>Possible mechanisms for demagnetisation</u>

- Direct interaction of spins with the magnetic component of the laser
  Zhang, Huebner, PRL **85**, 3025 (2000)

- Spin-flip electron-phonon scattering
  (ultimately leading to transfer of spin angular momentum to the lattice)
  Koopmans et al, PRL **95**, 267207 (2005)

- Super-diffusive spin transport
  Battiato, Carva, Oppeneer, PRL **105**, 027203 (2010)

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

Time-dependent density-functional theory

(E. Runge, E.K.U.G., PRL 52, 997 (1984))

Basic 1-1 correspondence:

$$v(\mathbf{r}t) \overset{1\text{-}1}{\longleftrightarrow} \rho(\mathbf{r}t)$$

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

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

of an auxiliary non-interacting (KS) system

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

with the local potential

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

Time-dependent density-functional theory

(E. Runge, E.K.U.G., PRL 52, 997 (1984))

Genuine ab-initio approach, in principle exact

Basic 1-1 correspondence:

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

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

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

Generalization: Non-collinear-Spin-TDDFT with SOC

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

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

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

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

Generalization: Non-collinear-Spin-TDDFT with SOC

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

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

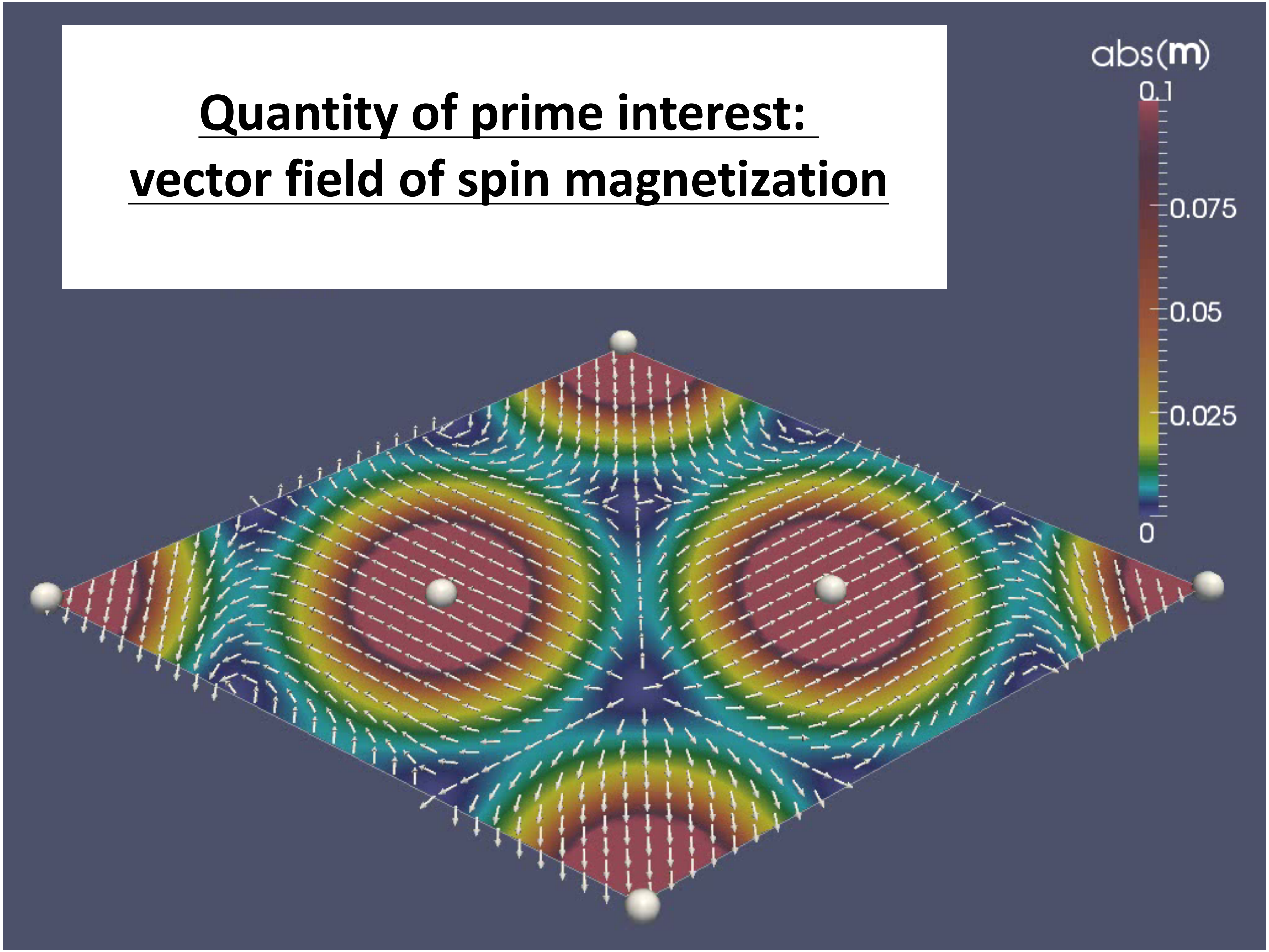
$$B_S[\rho,\boldsymbol{m}](r,t)=B_{external}(r,t)+B_{xc}[\rho,\boldsymbol{m}](r,t)$$

<u>Universal</u> functionals of ρ and m

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

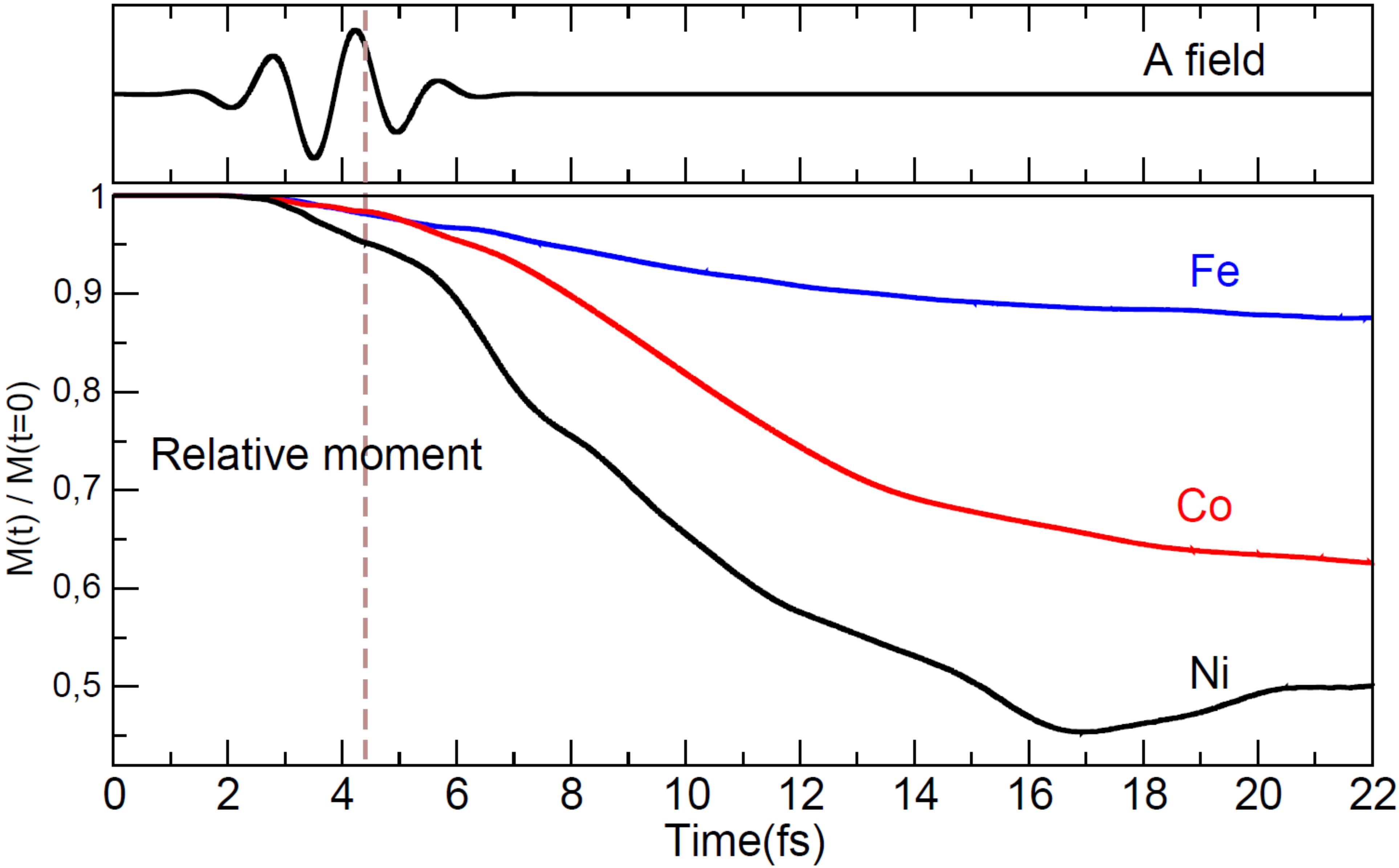
Quantity of prime interest: vector field of spin magnetization

Quantity of prime interest: vector field of spin magnetization



Cr monolayer in ground state

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/)**

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/)**

Kay Dewhurst

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

Sangeeta Sharma

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

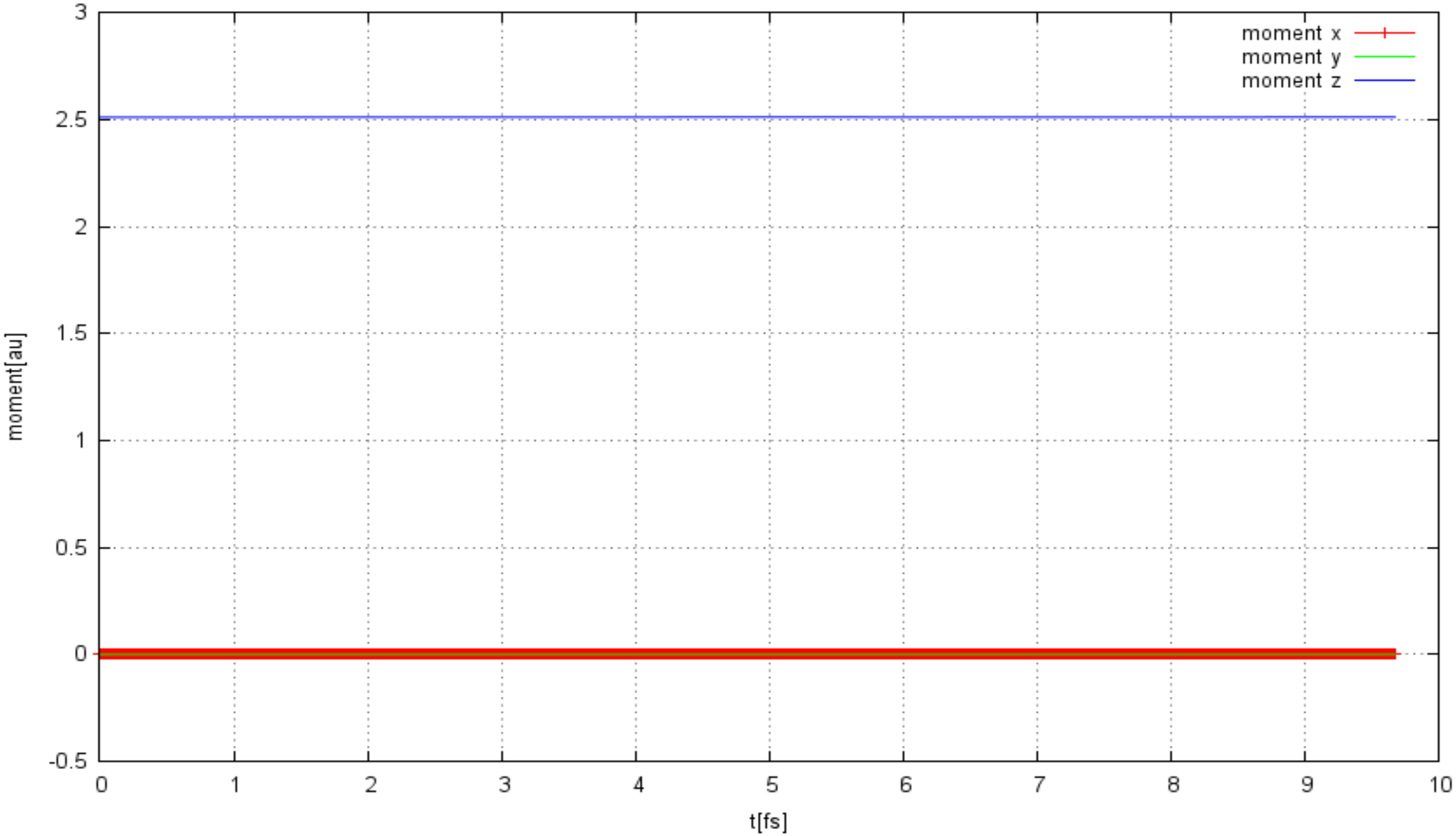
- **Periodicity of the TDKS Hamiltonian is preserved!**

- **Implementation in ELK code (FLAPW) (http://elk.sourceforge.net/)**

Analysis of the results

Calculation without spin-orbit coupling

components of spin moment



Exact equation of motion

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

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

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

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

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

Exact equation of motion

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

Global torque exerted by B_{KS}

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

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

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

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

Exact equation of motion

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

Global torque exerted by B_{KS}

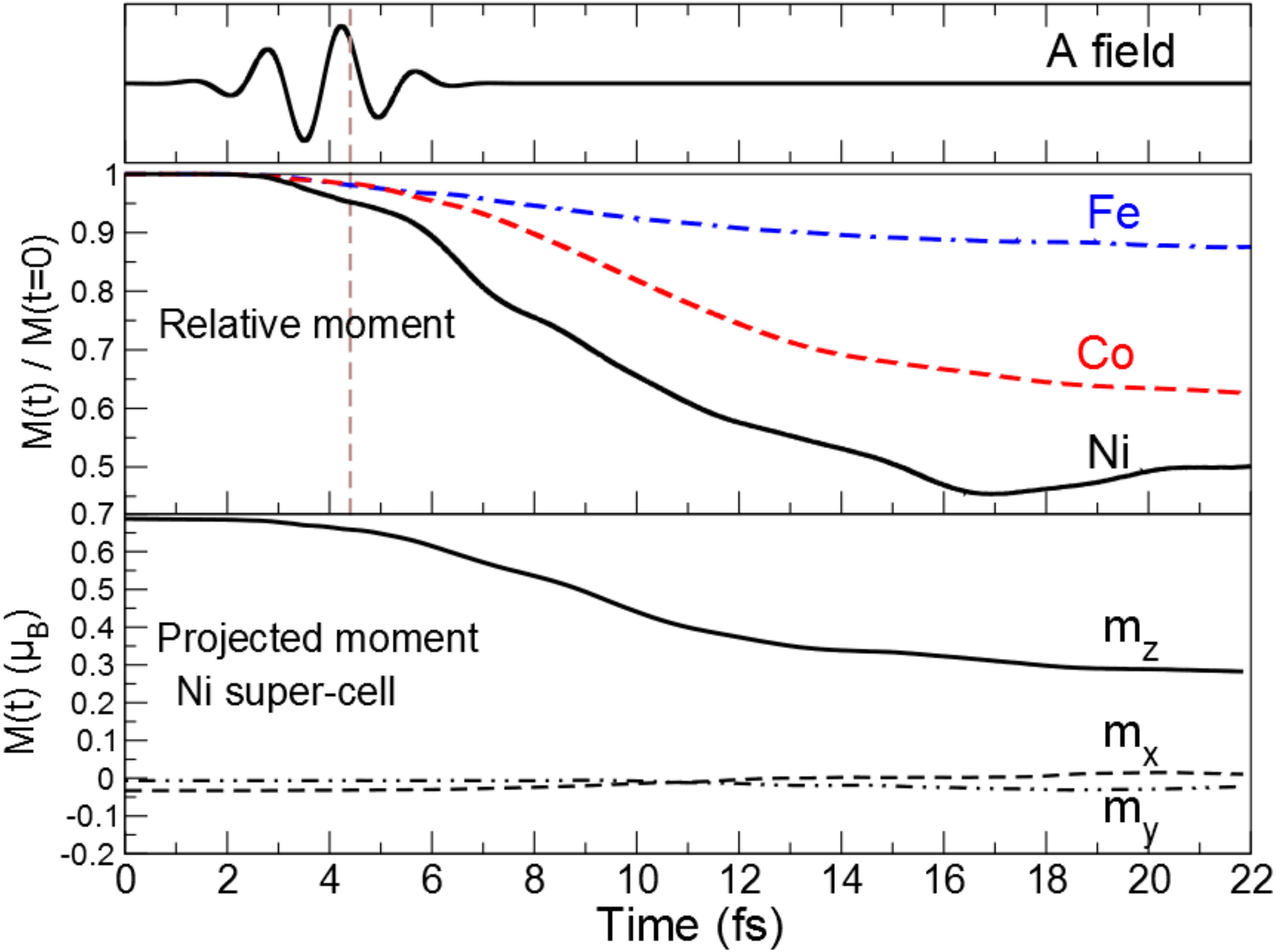
$$= \int d^3r \left\{M_x(r,t)B_{KS,y}(rt) - M_y(r,t)B_{KS,x}(rt)\right\}$$

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

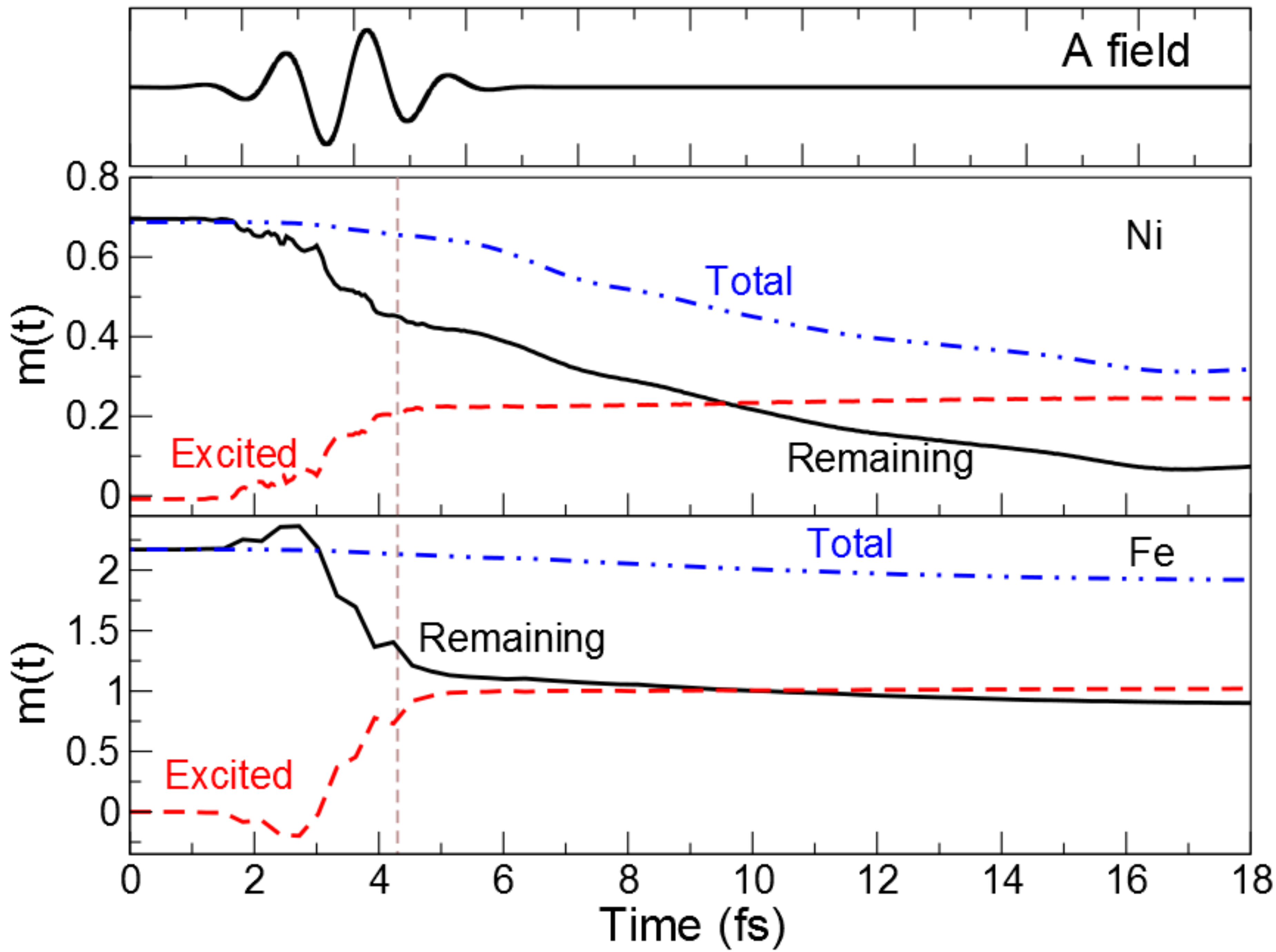
$\overset{\leftrightarrow}{j}(r,t) = \langle\hat{\sigma}\otimes\hat{p}\rangle$ spin current tensor

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

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



Note: Ground state of bulk Fe, Co, Ni is collinear

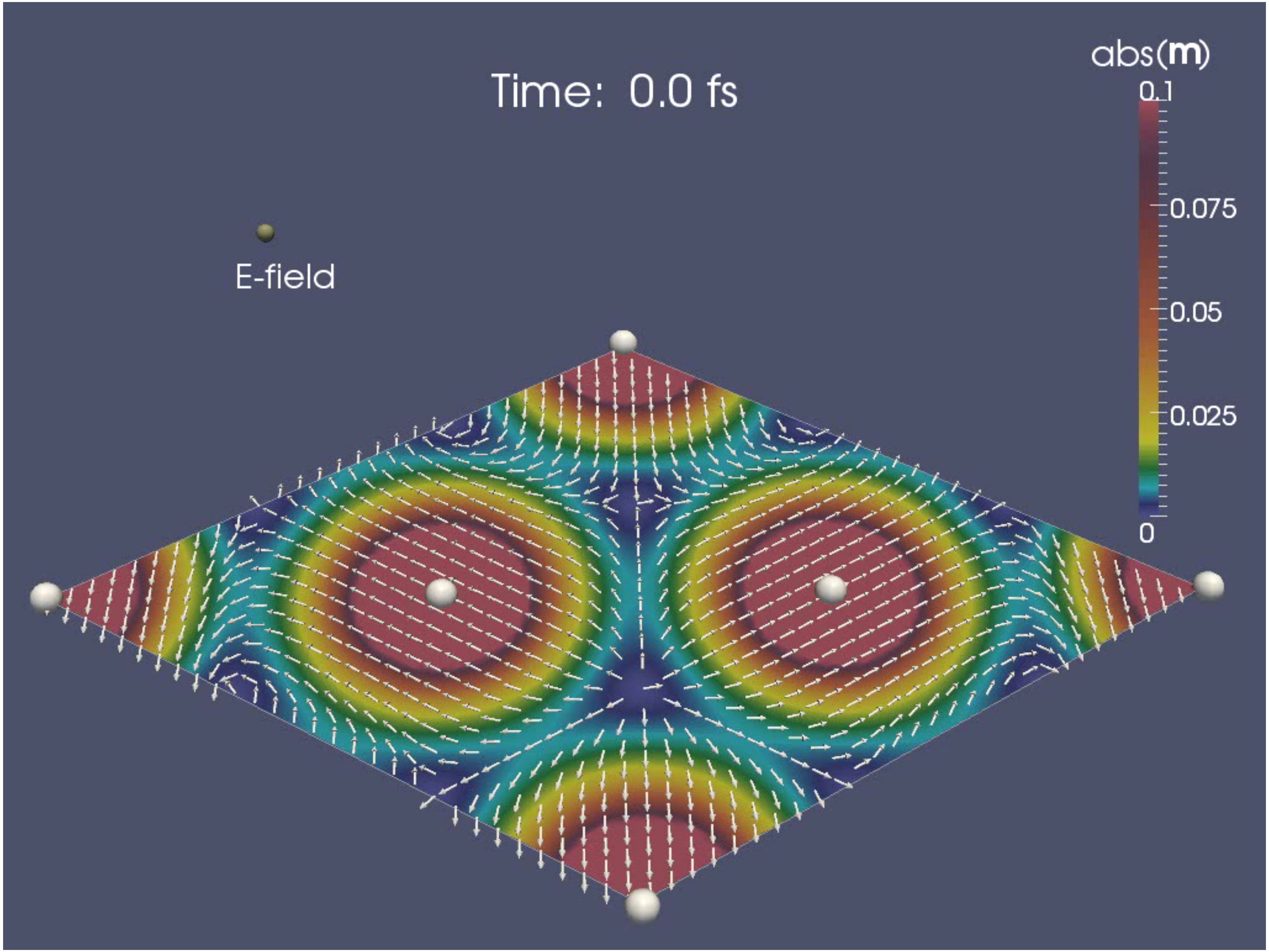

A field
0.8
0.6
0.4
0.2
0
m(t)
Ni
Total
Excited
Remaining
Fe
Total
Remaining
Excited
2
1.5
1
0.5
0
m(t)
0
2
4
6
8
10
12
14
16
18
Time (fs)

Demagnetization occurs in two steps:

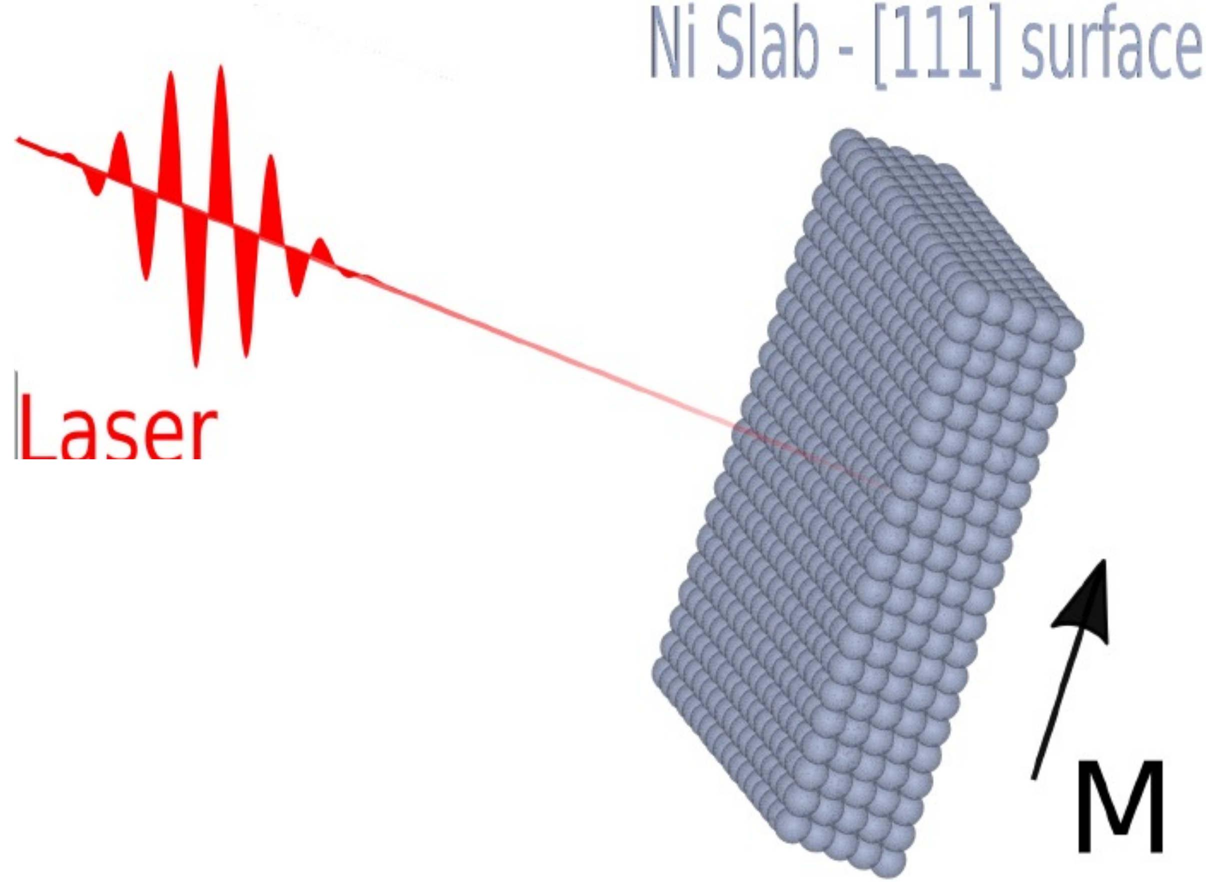
- Initial excitation by laser *moves* magnetization from atomic region into interstitial region. Total Moment is basically conserved during this phase.

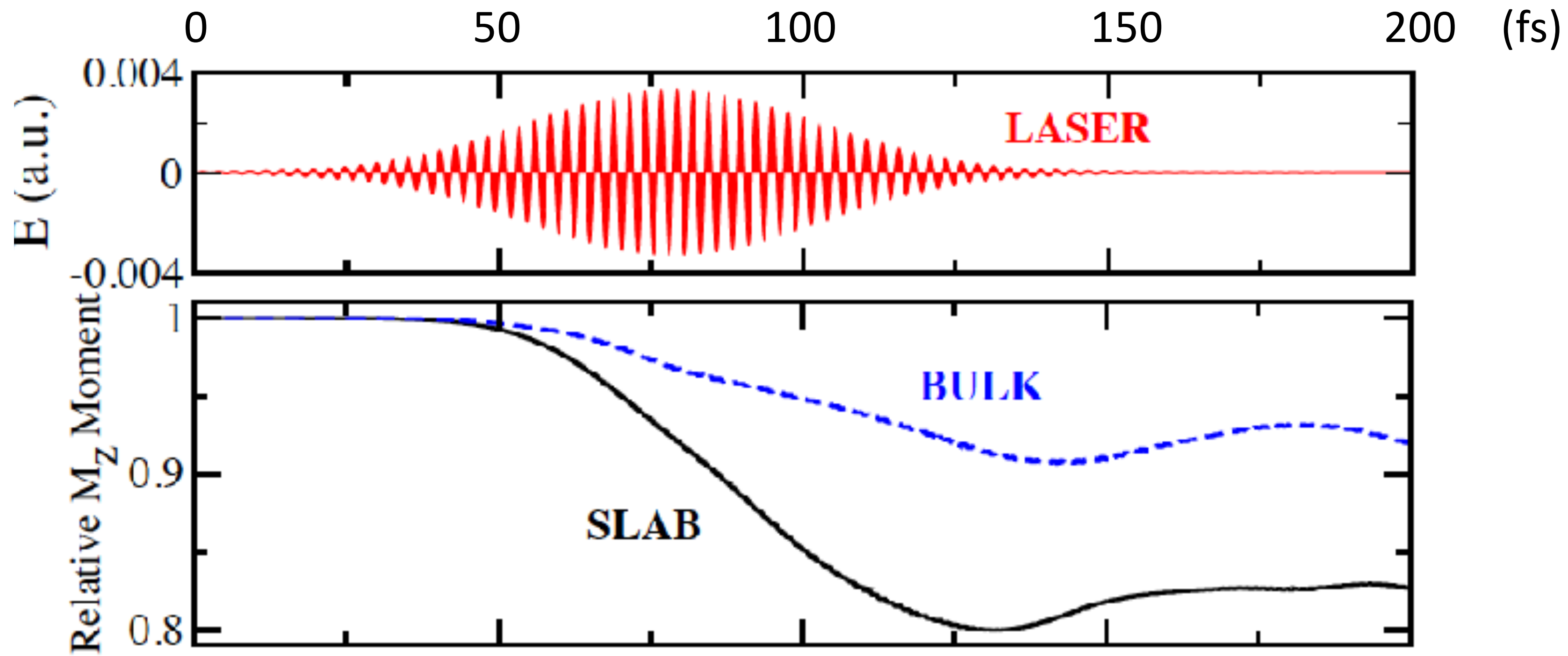
- Spin-Orbit term drives demagnetization of the more localized electrons until stabilization at lower moment is achieved

Beyond 3D bulk

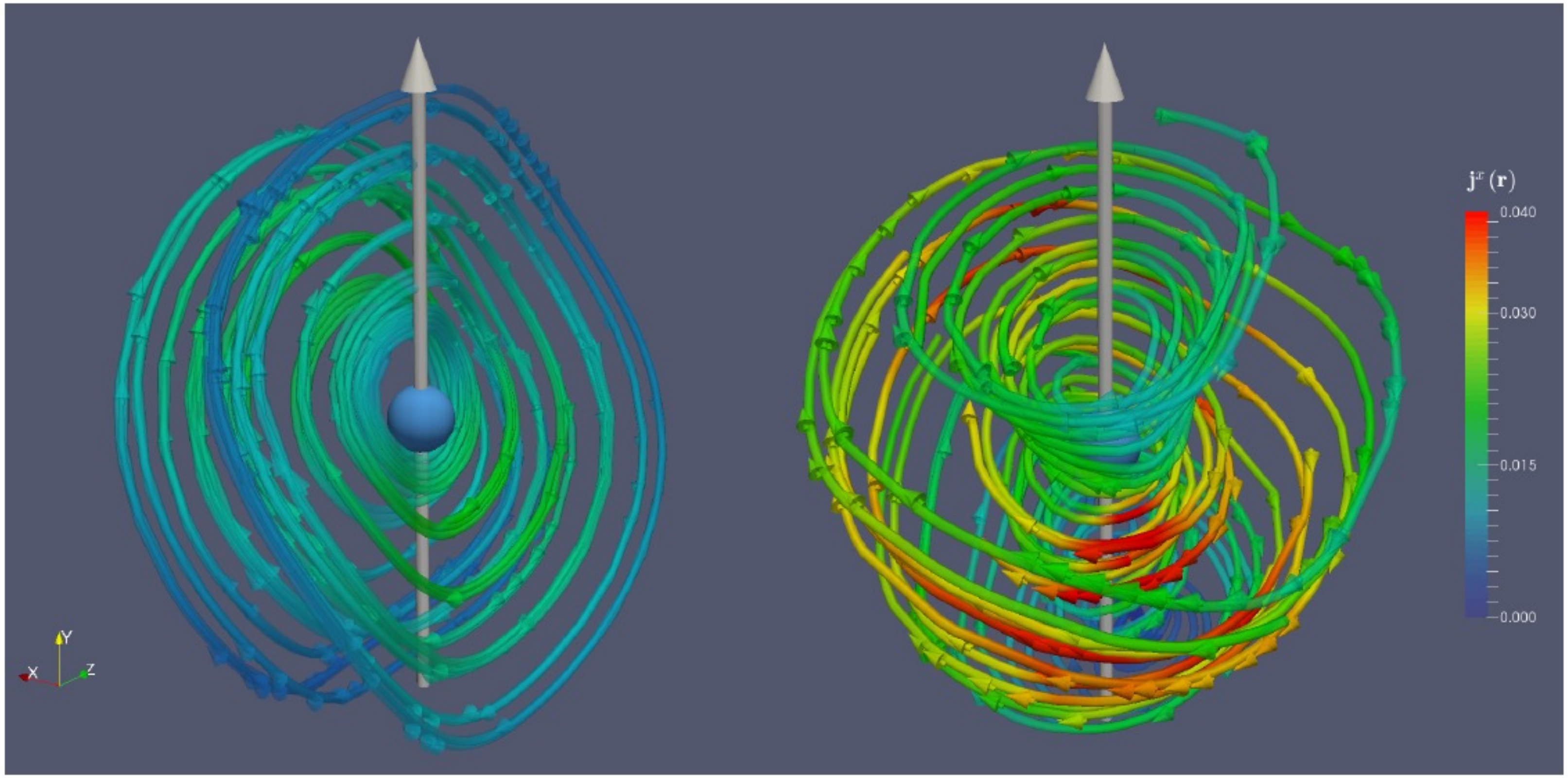


Cr monolayer

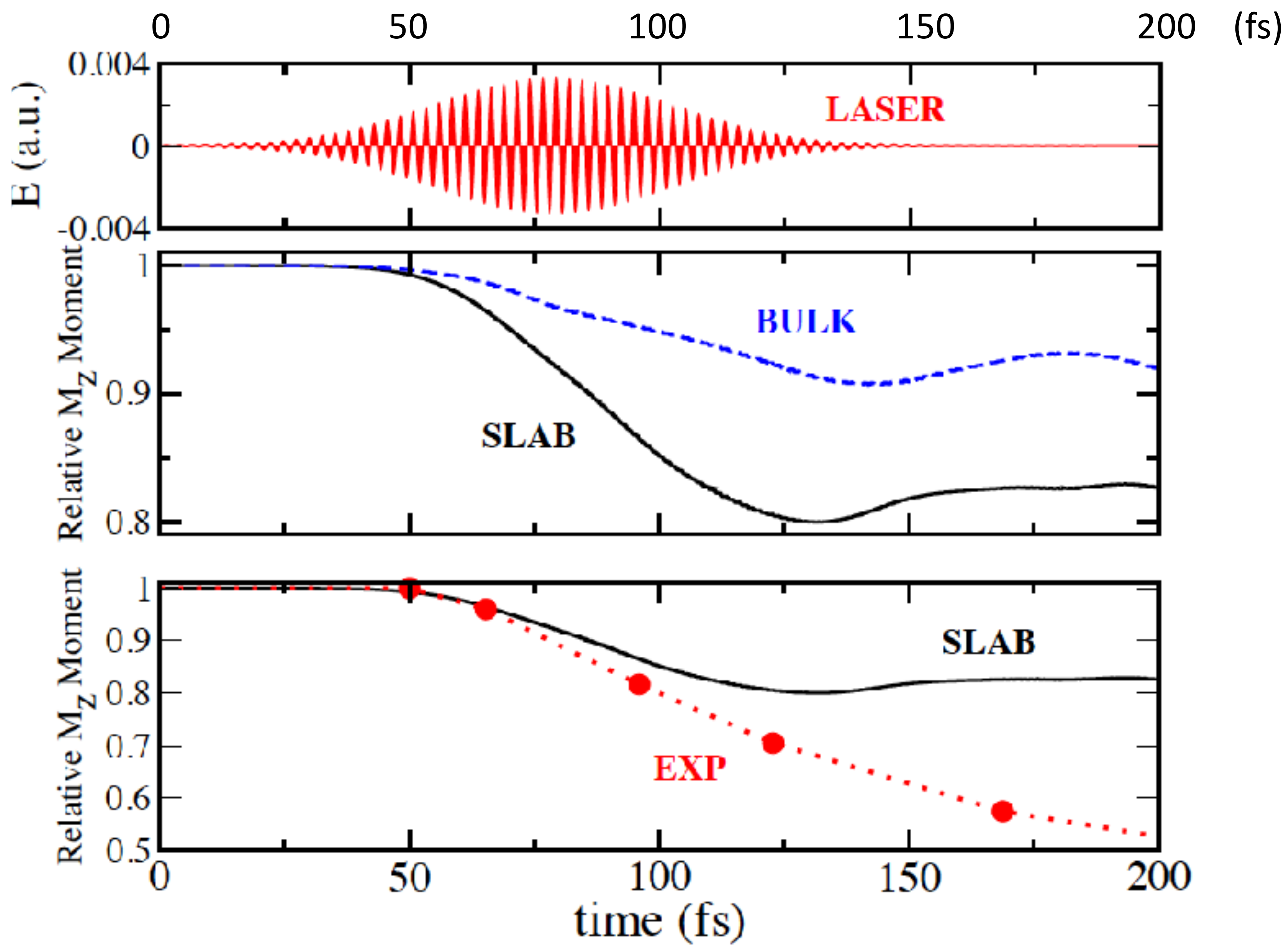

Ni Slab - [111] surface
Laser
M



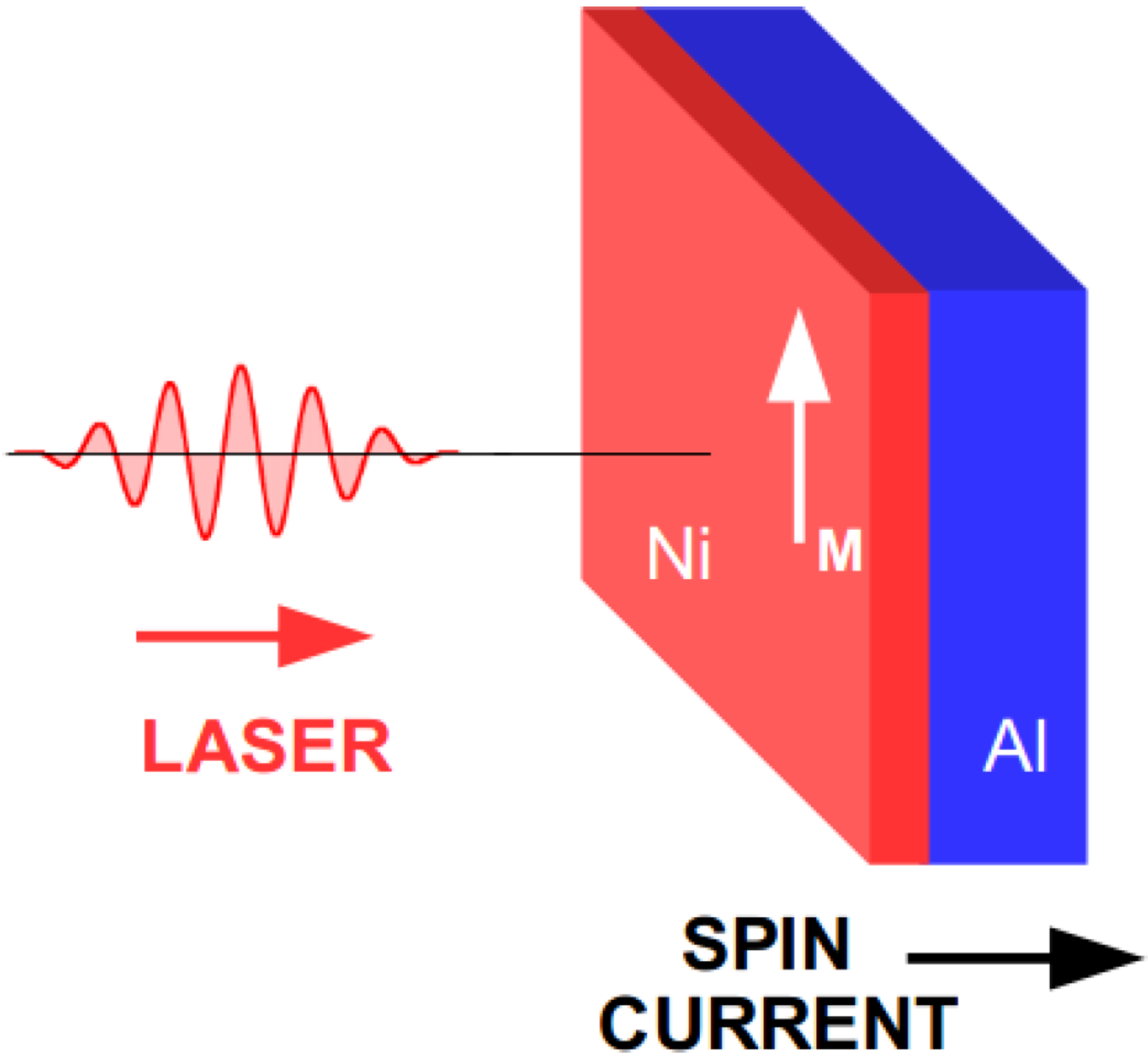
0
50
100
150
200
(fs)
0.004
0
-0.004
E (a.u.)
LASER
1
0.9
0.8
Relative M_Z Moment
BULK
SLAB



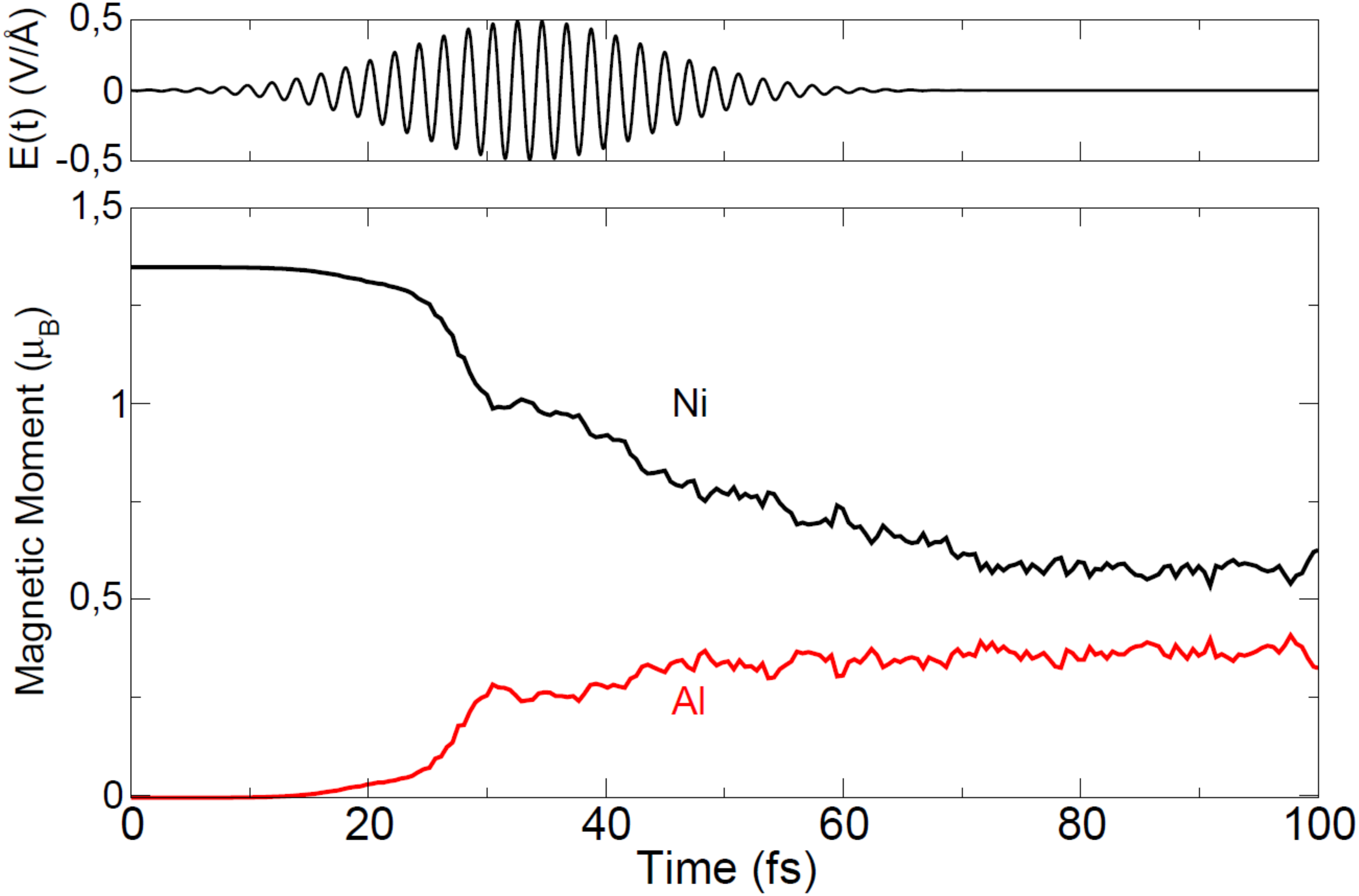
Streamlines for J_x, the spin-current vector field of the x component of spin, around a Ni atom in bulk (left) and for the outermost Ni atom in the slab (right).

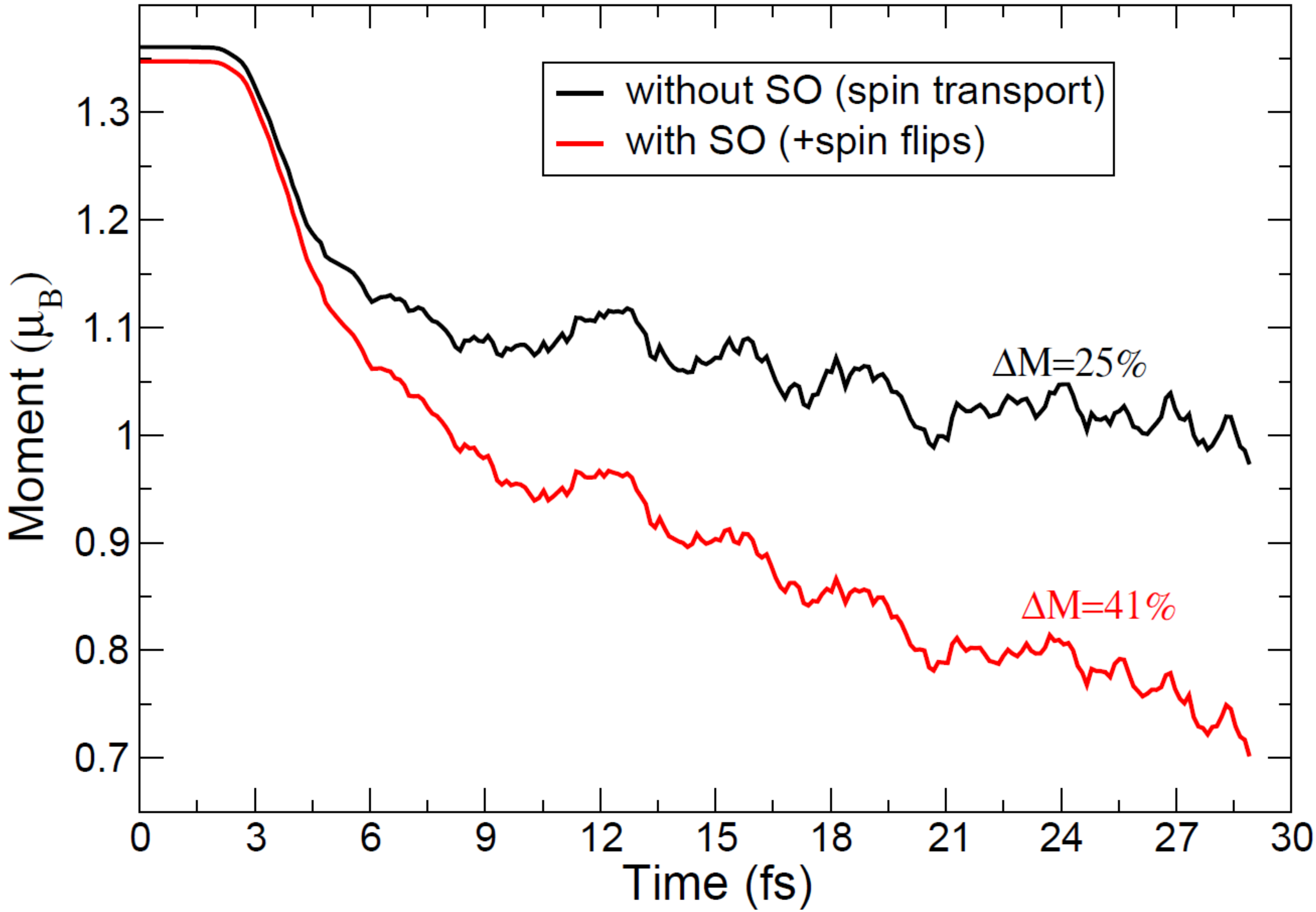


0
50
100
150
200
(fs)
0.004
0
-0.004
E (a.u.)
LASER
1
0.9
0.8
Relative M_Z Moment
BULK
SLAB
1
0.9
0.8
0.7
0.6
0.5
Relative M_Z Moment
SLAB
EXP
0
50
100
150
200
time (fs)


Ni
M
Al
LASER
SPIN
CURRENT

Effect of spin transport across interfaces: Ni@Al

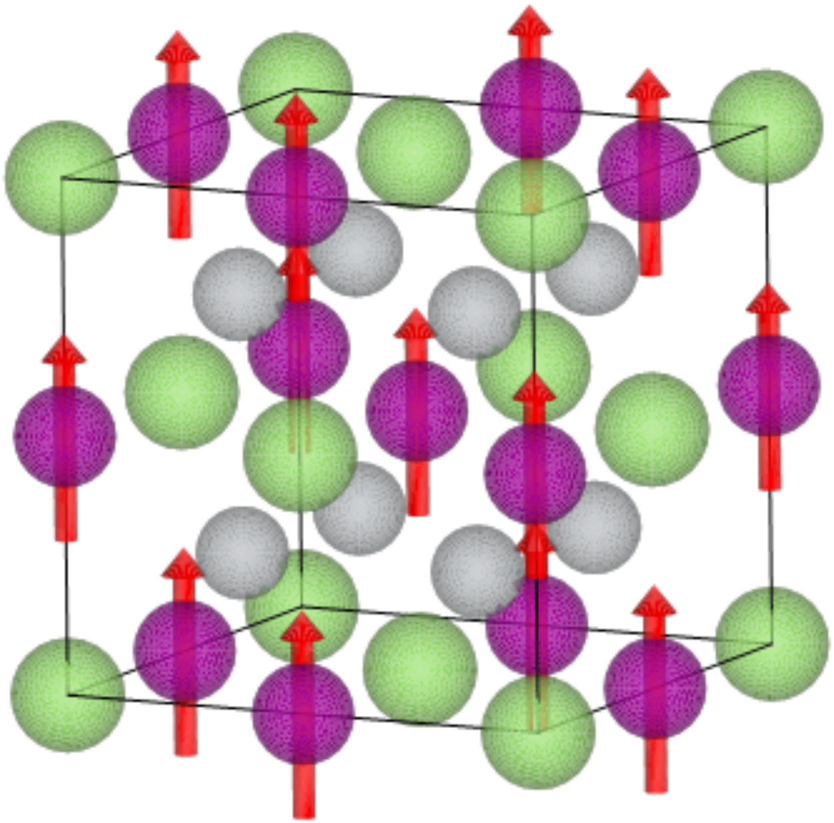




without SO (spin transport)
with SO (+spin flips)
ΔM=25%
ΔM=41%
Moment (μB)
Time (fs)
1.3
1.2
1.1
1
0.9
0.8
0.7
0
3
6
9
12
15
18
21
24
27
30

Heusler compounds

Ni_2MnGa

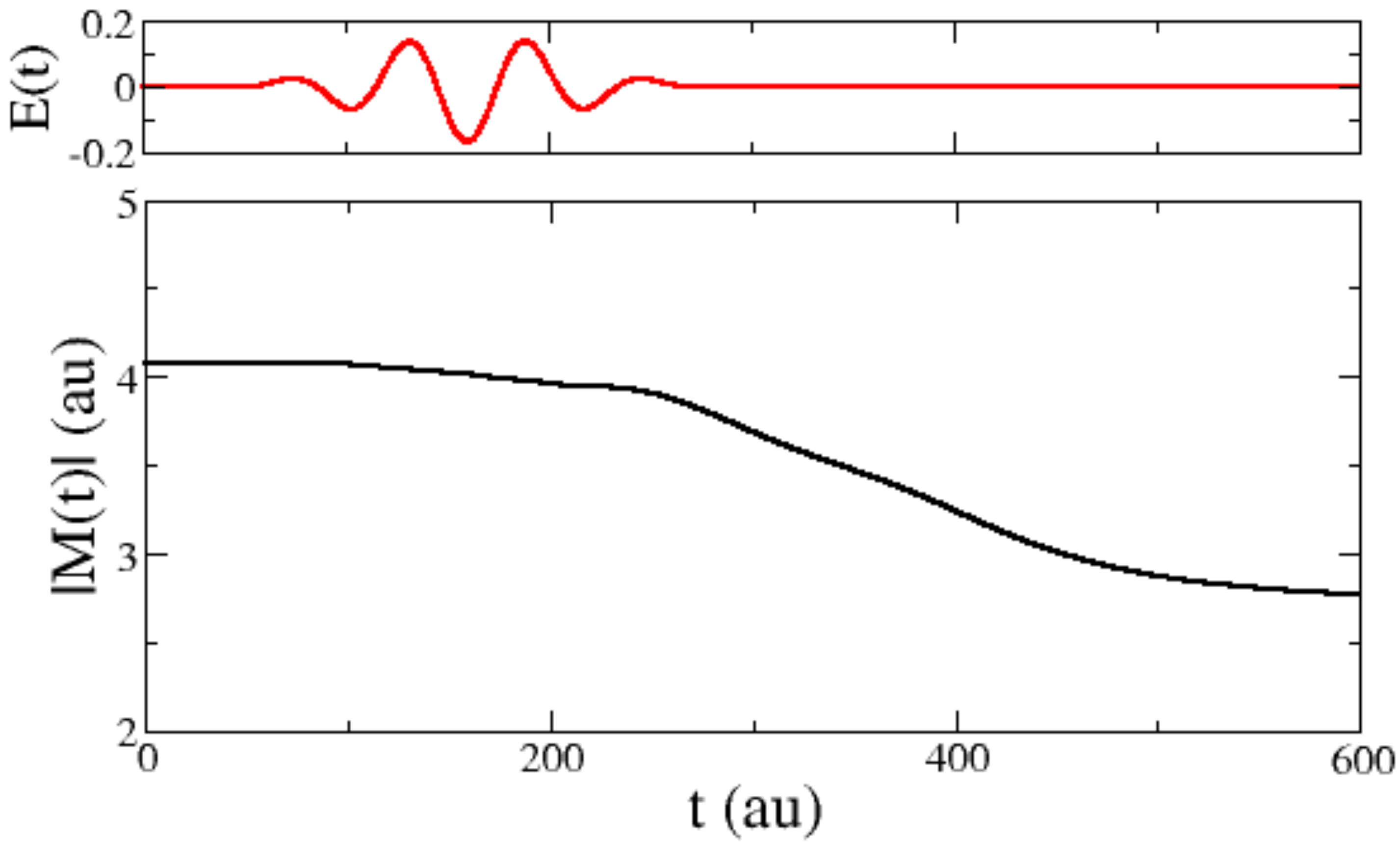


Ga	0.02 μB
Mn	-3.14 μB
Ni	-0.37 μB

Ni_2MnGa

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

See loss in global moment

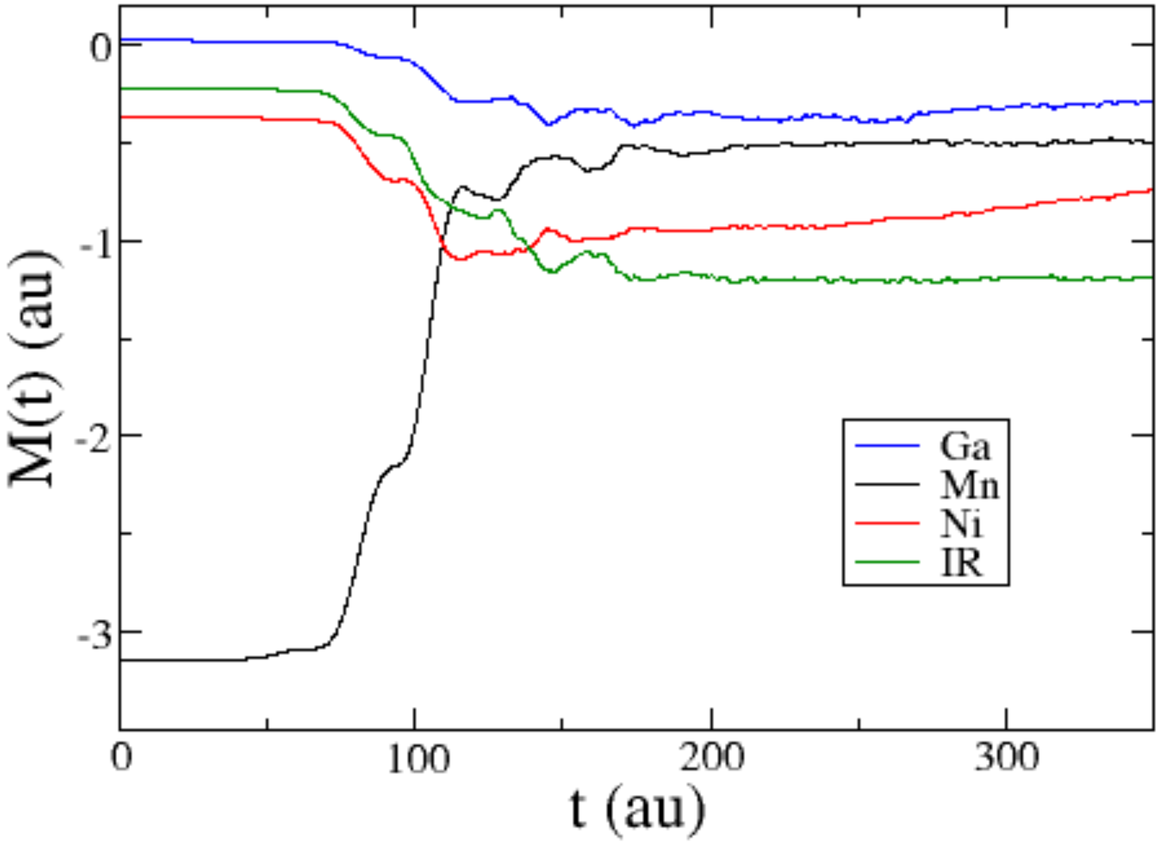


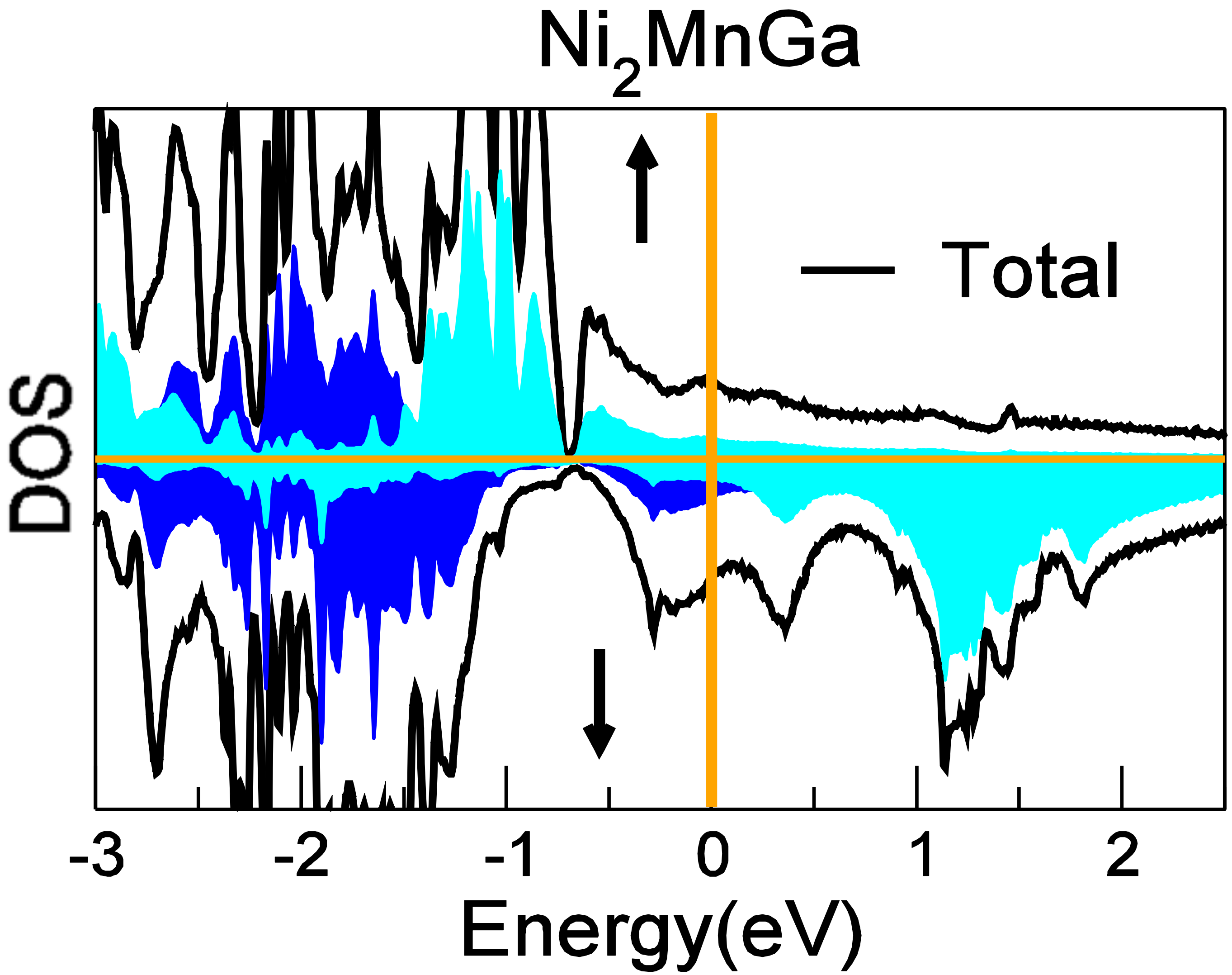
Ni_2MnGa

Also change in local moments

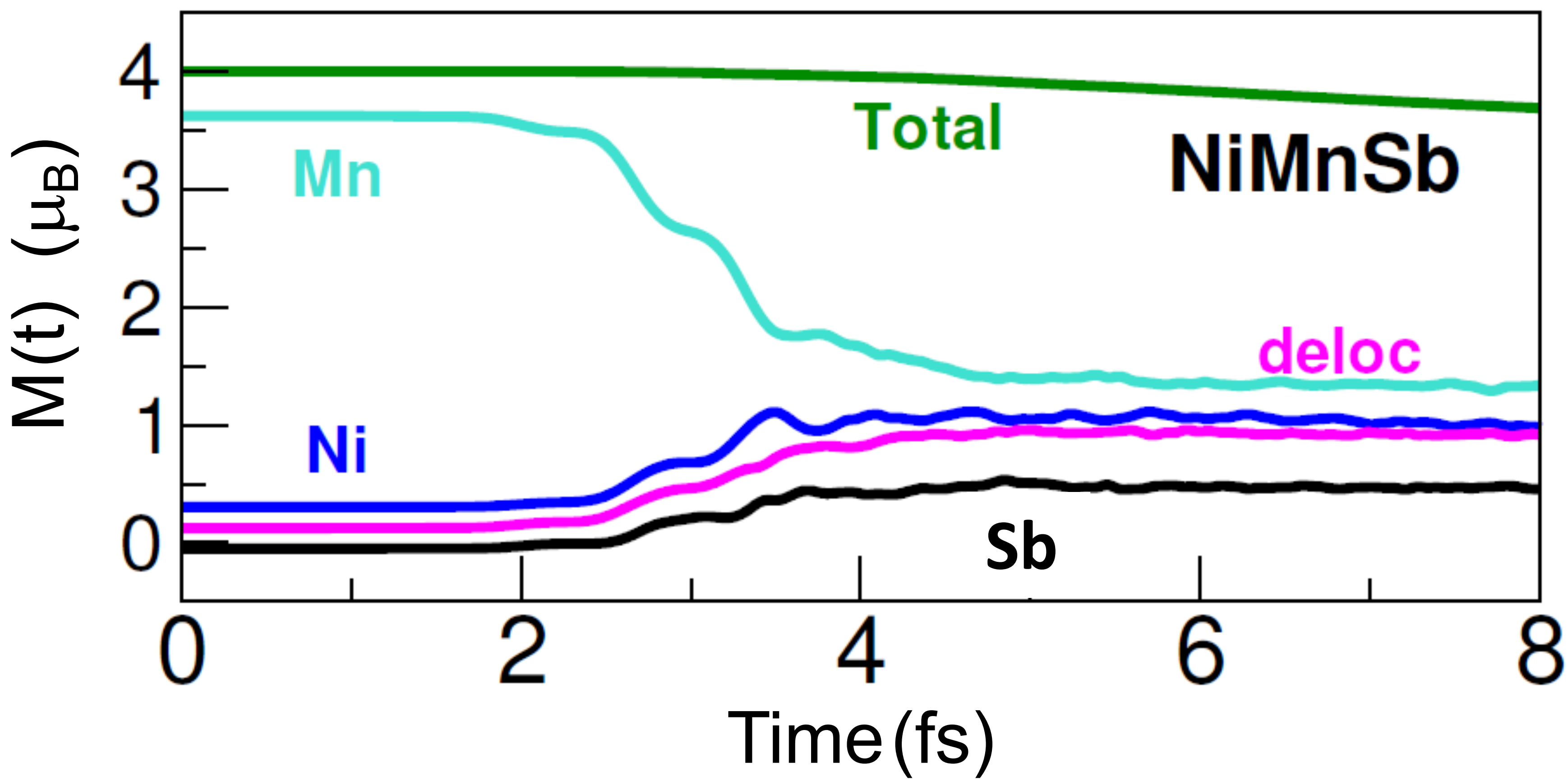
Transfer of moment from Mn to Ni (does not require SOC)

Followed by spin-orbit mediated demagnetization on Ni



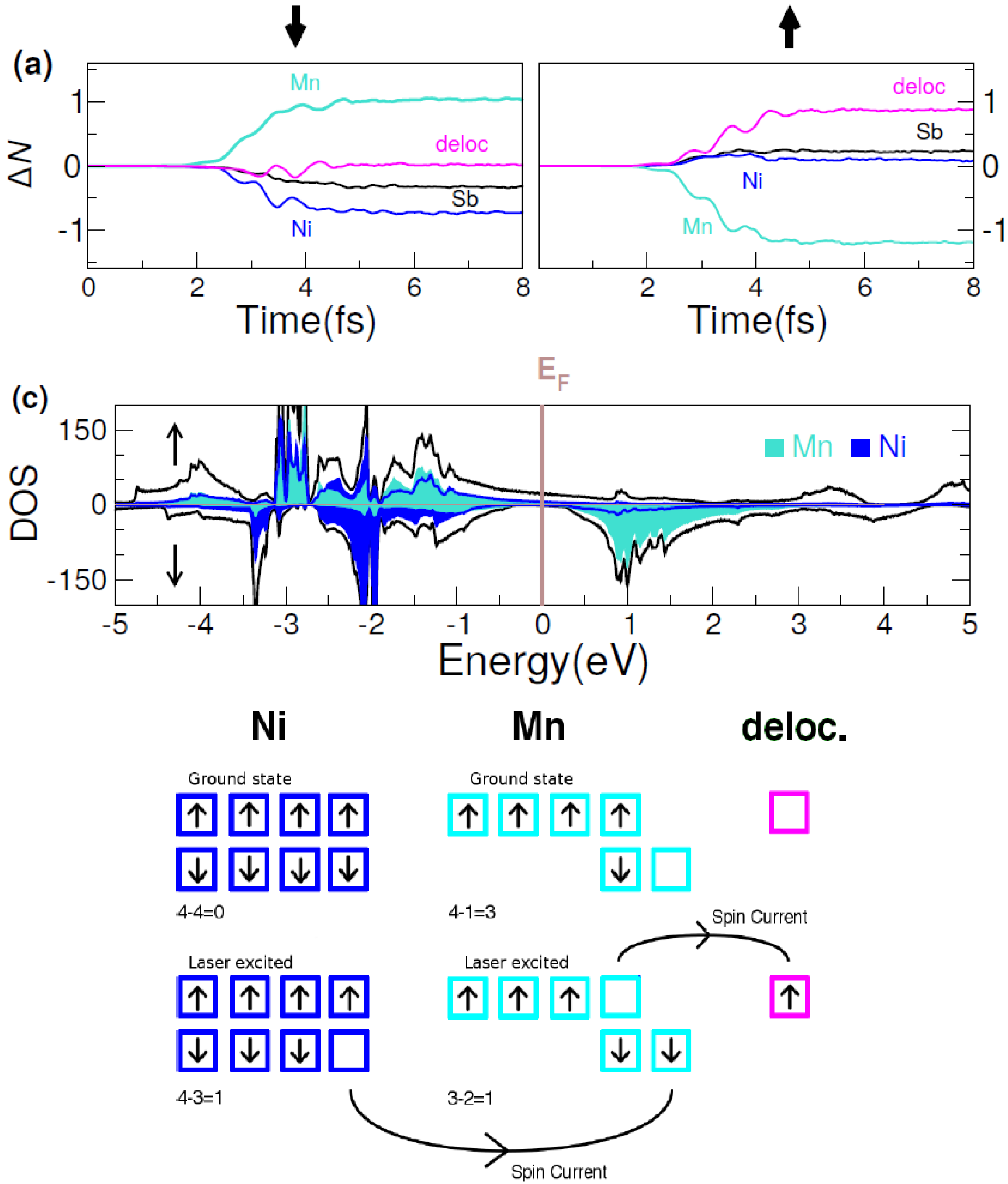


P. Elliott, T. Mueller, K. Dewhurst, S. Sharma, E.K.U.G., arXiv 1603.05603

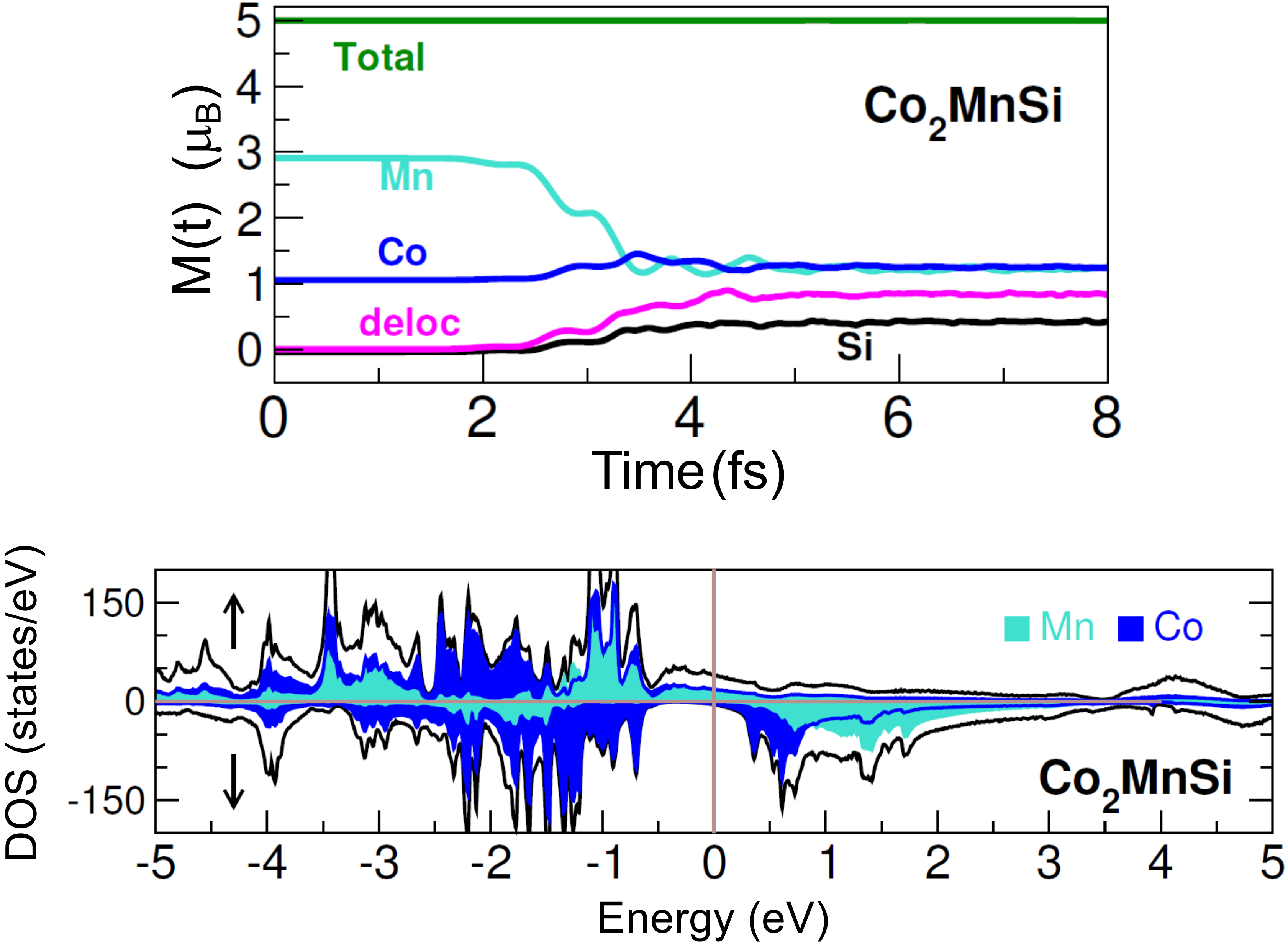


NiMnSb
Total
Mn
deloc
Ni
Sb
M(t) (μB)
Time (fs)
0
1
2
3
4
0
2
4
6
8

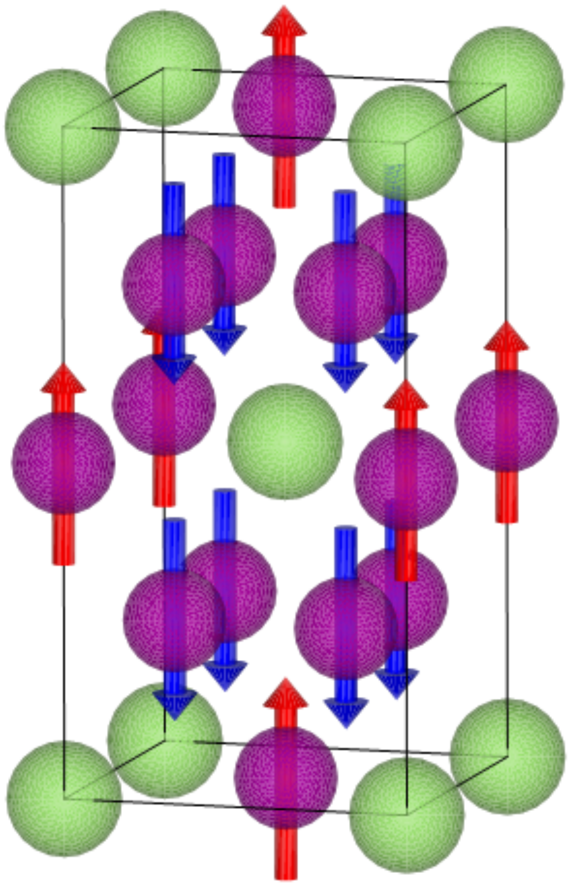
NiMnSb



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


Total
Co2MnSi
Mn
Co
deloc
Si
M(t) (μB)
Time (fs)
Mn
Co
Co2MnSi
DOS (states/eV)
Energy (eV)

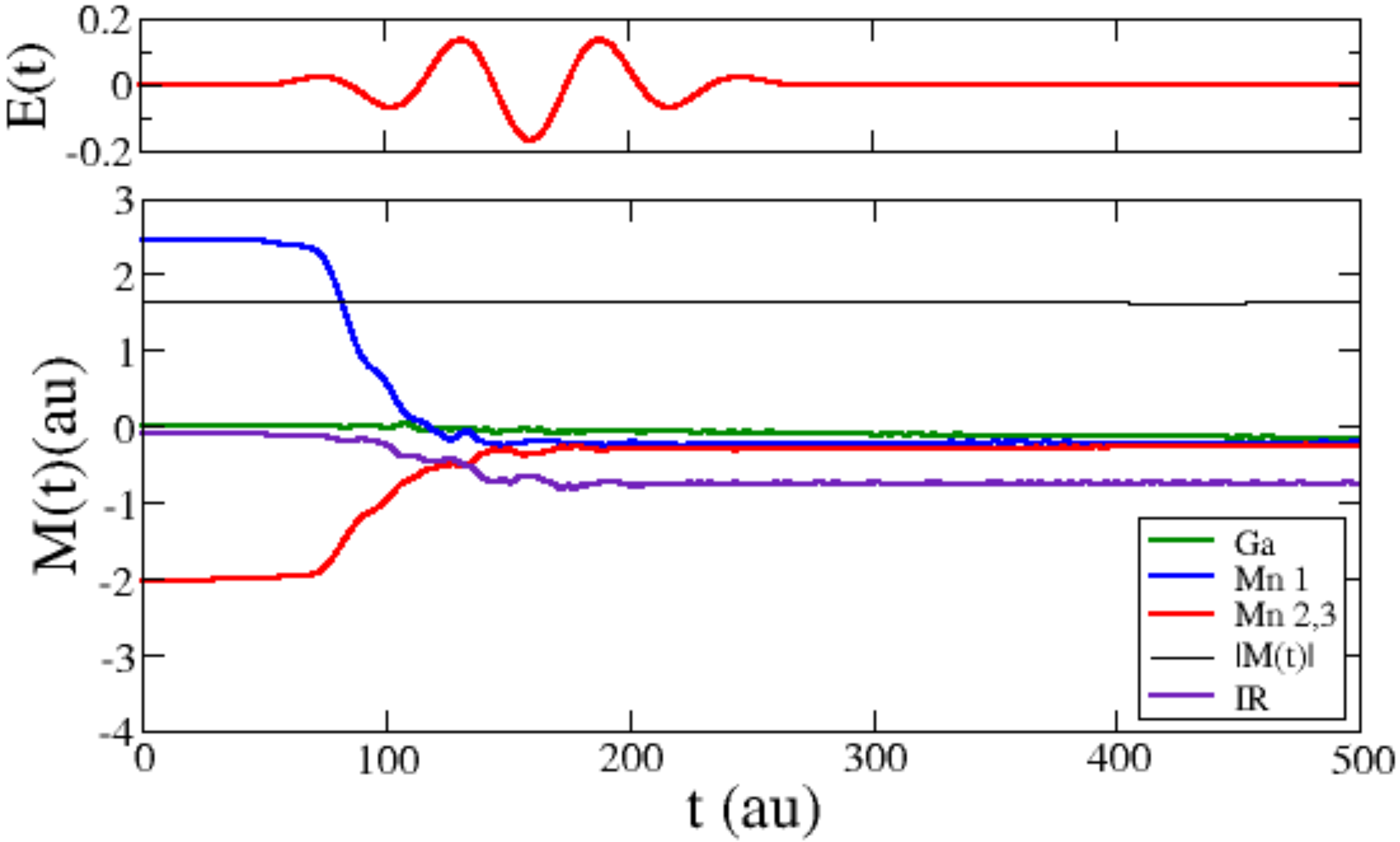
Mn_3Ga



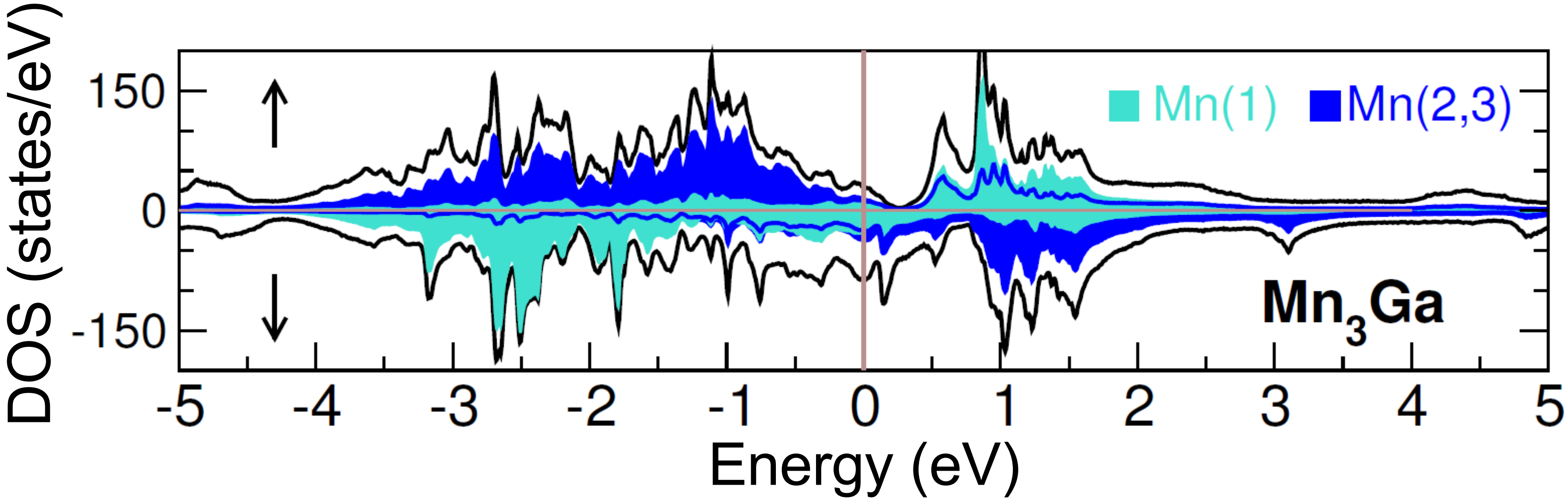
Mn_3Ga

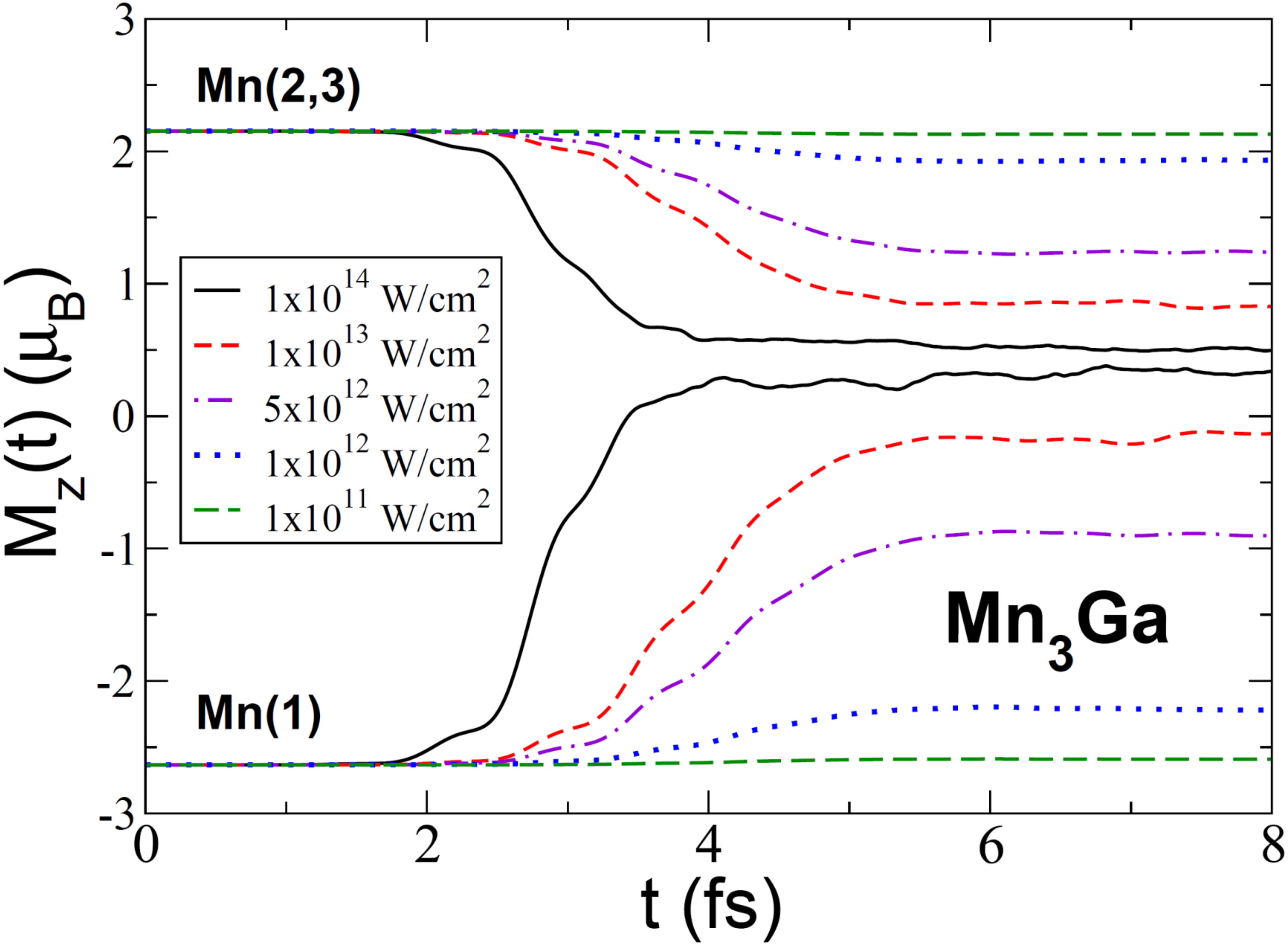
Laser parameters: ω=2.72eV Ipeak= 1x1015 W/cm2 J = 935 mJ/cm2 FWHM = 2.42 fs

Global moment |M(t)| preserved
Local moments around each atom change



P. Elliott, T. Mueller, K. Dewhurst, S. Sharma, E.K.U.G., arXiv 1603.05603


DOS (states/eV)
150
0
-150
↑
↓
Mn(1)
Mn(2,3)
Mn_3Ga
-5
-4
-3
-2
-1
0
1
2
3
4
5
Energy (eV)



Mn(2,3)
Mn(1)
Mn3Ga
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
Mz(t) (μB)
t (fs)
3
2
1
0
-1
-2
-3
0
2
4
6
8

Summary

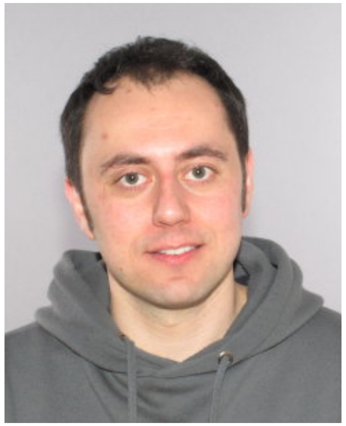
- Demagnetization in first 50 fs is a **universal** two-step process:
  1. Initial excitation of electrons into highly excited delocalised states (without much of a change in the total magnetization)
  2. Spin-orbit coupling drives demagnetization of the more localized electrons

- 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 o the basis of the ground-state DOS

Coworkers



Kevin Krieger

Sangeeta Sharma

Florian Eich

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 $v_{xc}\left[\rho\left(r't'\right)\right]\left(rt\right)$

- **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**