Ultrafast laser-induced demagnetization of solids: Understanding the mechanism with real-time TDDFT simulations

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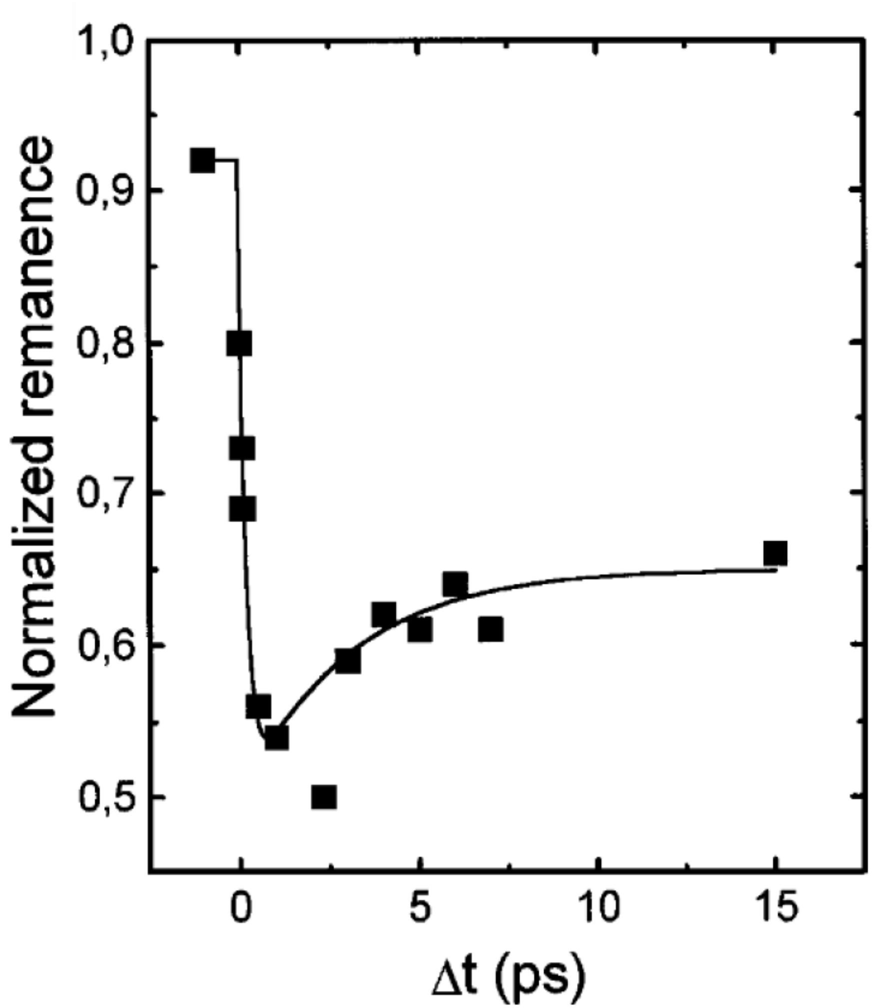
Halle (Saale)

First experiment on ultrafast laser induced demagnetization



Beaurepaire et al, PRL <u>76</u>, 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

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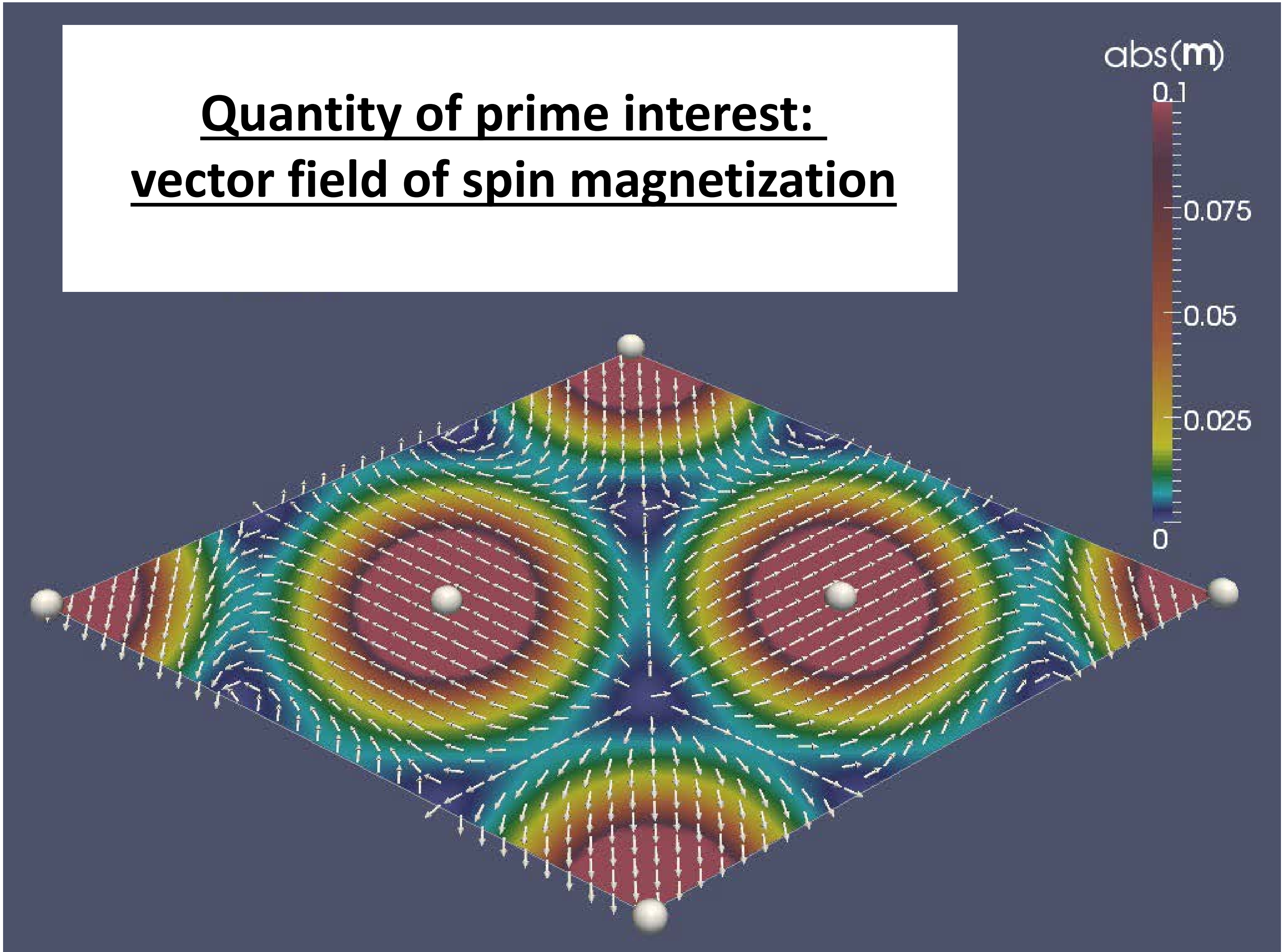


Cr monolayer in ground state

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

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

Generalization: Real-time TDDFT with SOC

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

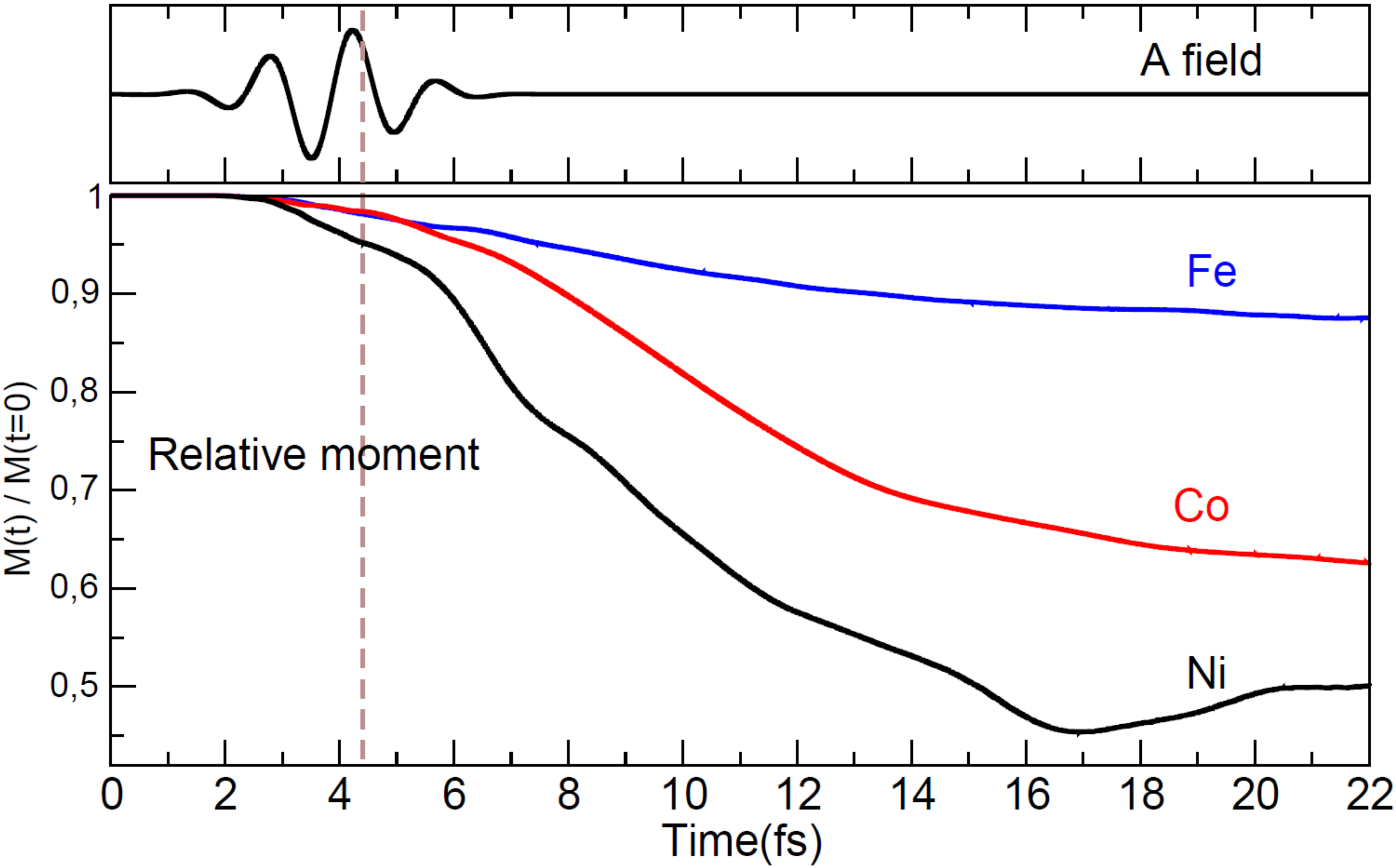
$$+\frac{\mu_B}{2c}\sigma\cdot\left(\nabla v_S[\rho,\boldsymbol{m}](r,t)\right)\times\left(-i\nabla\right)\Bigg]\varphi_k(r,t)$$

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

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

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

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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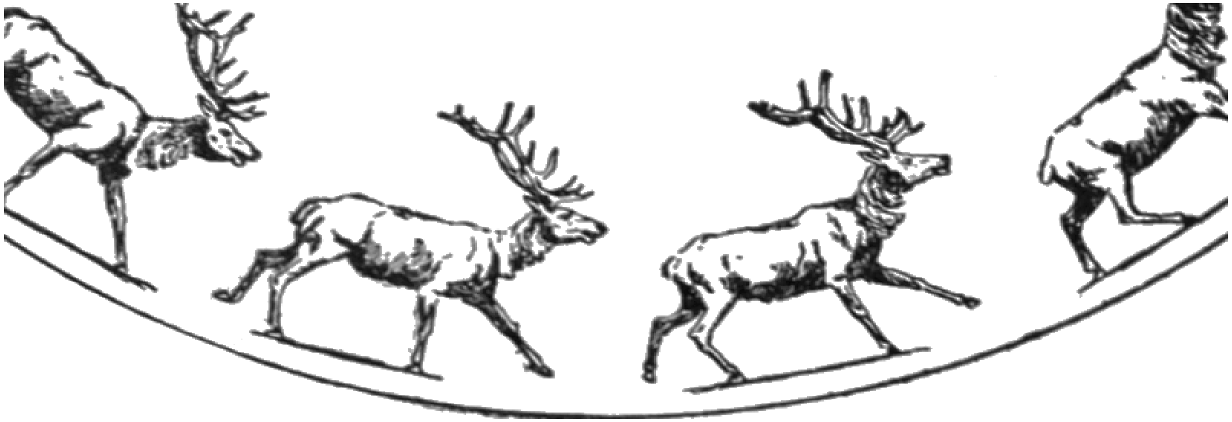


Algorithm for time propagation

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 $\hat{H}_{KS}(t)$

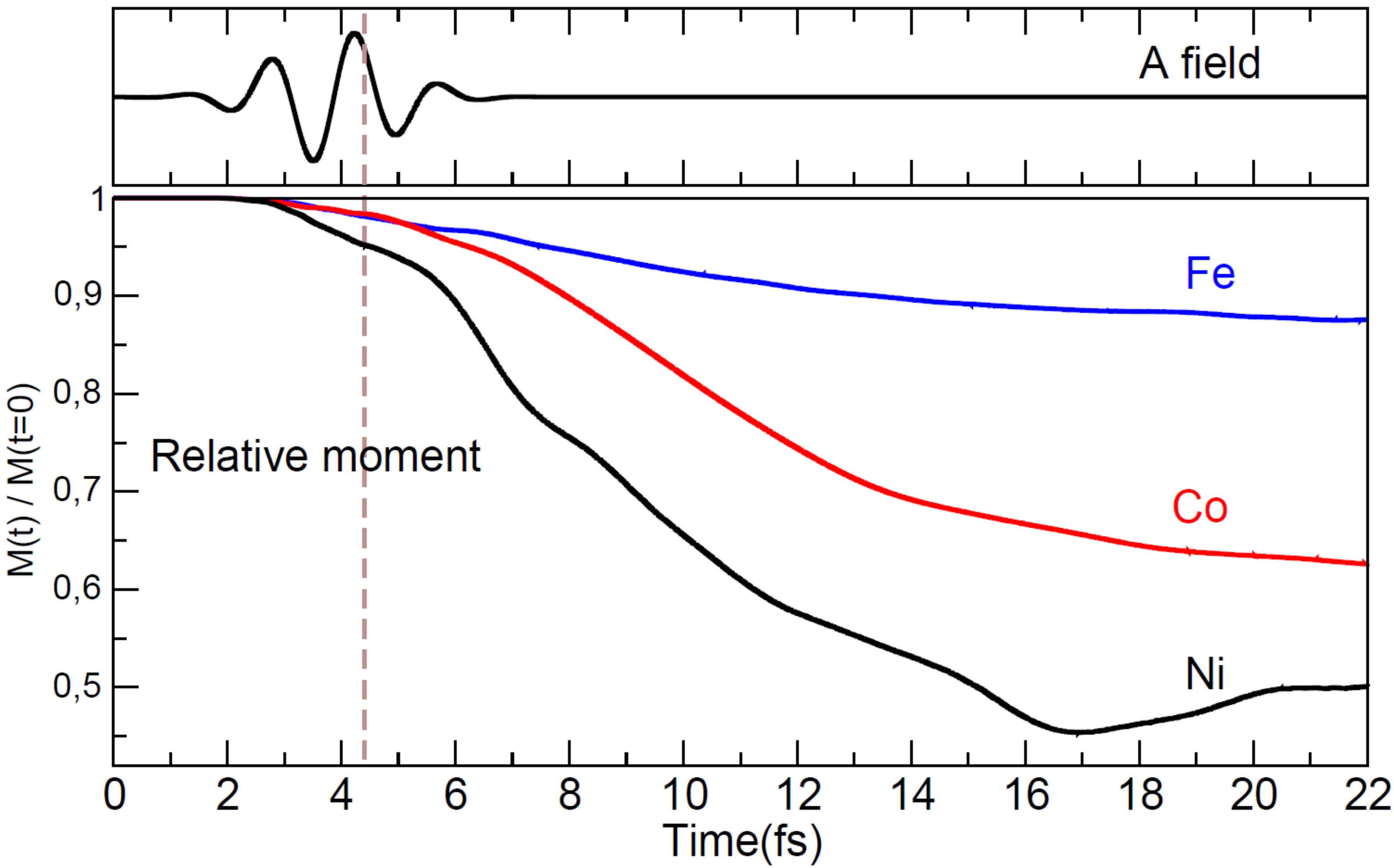
4. Compute $H_{ij} \equiv \left\langle \chi_i \middle| \hat{H}_{KS}(t) \middle| \chi_j \right\rangle$

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

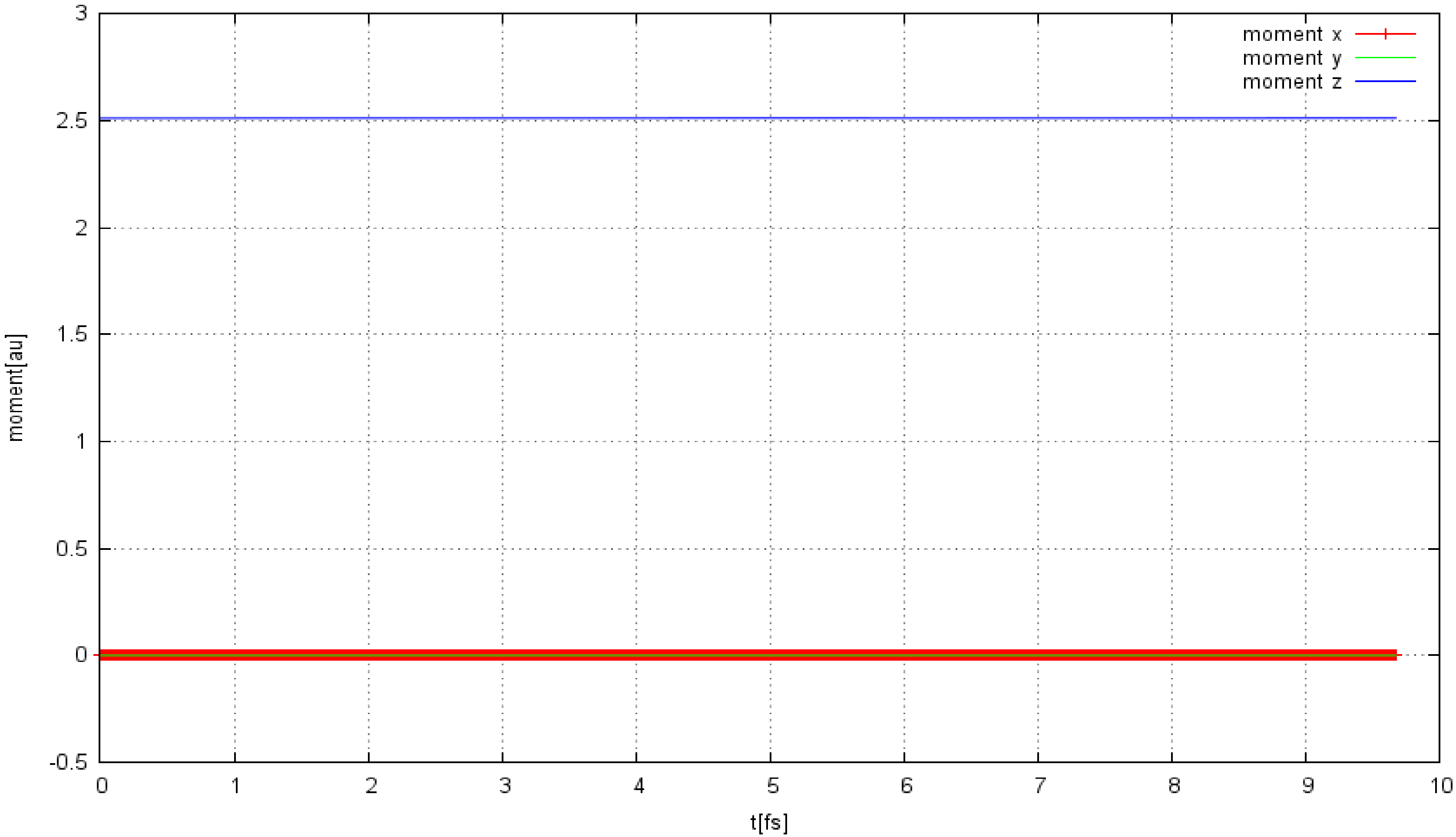


K. Krieger, K. Dewhurst, P. Elliott, S. Sharma, E.K.U.G., JCTC 11, 4870 (2015)

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

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

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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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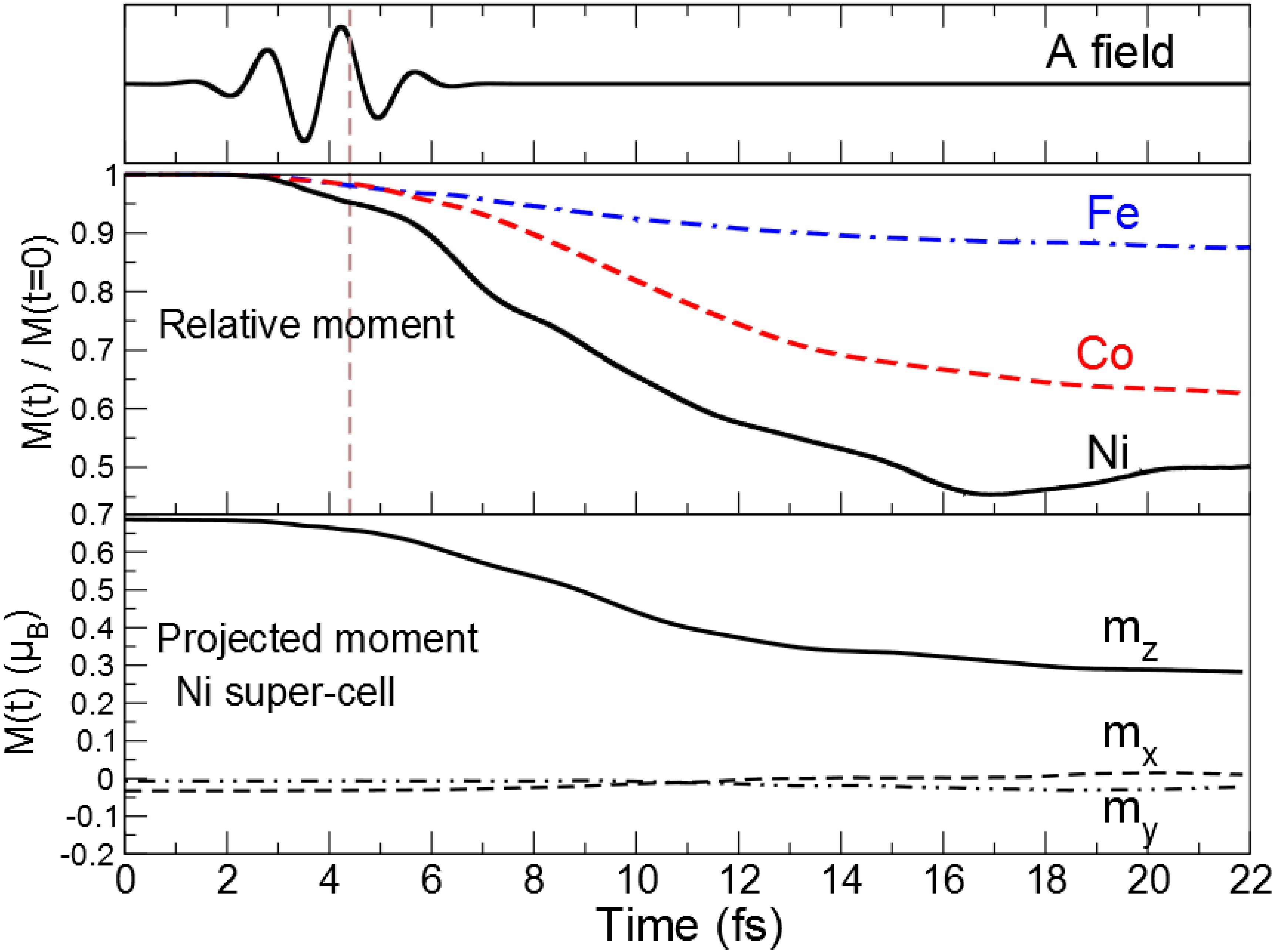
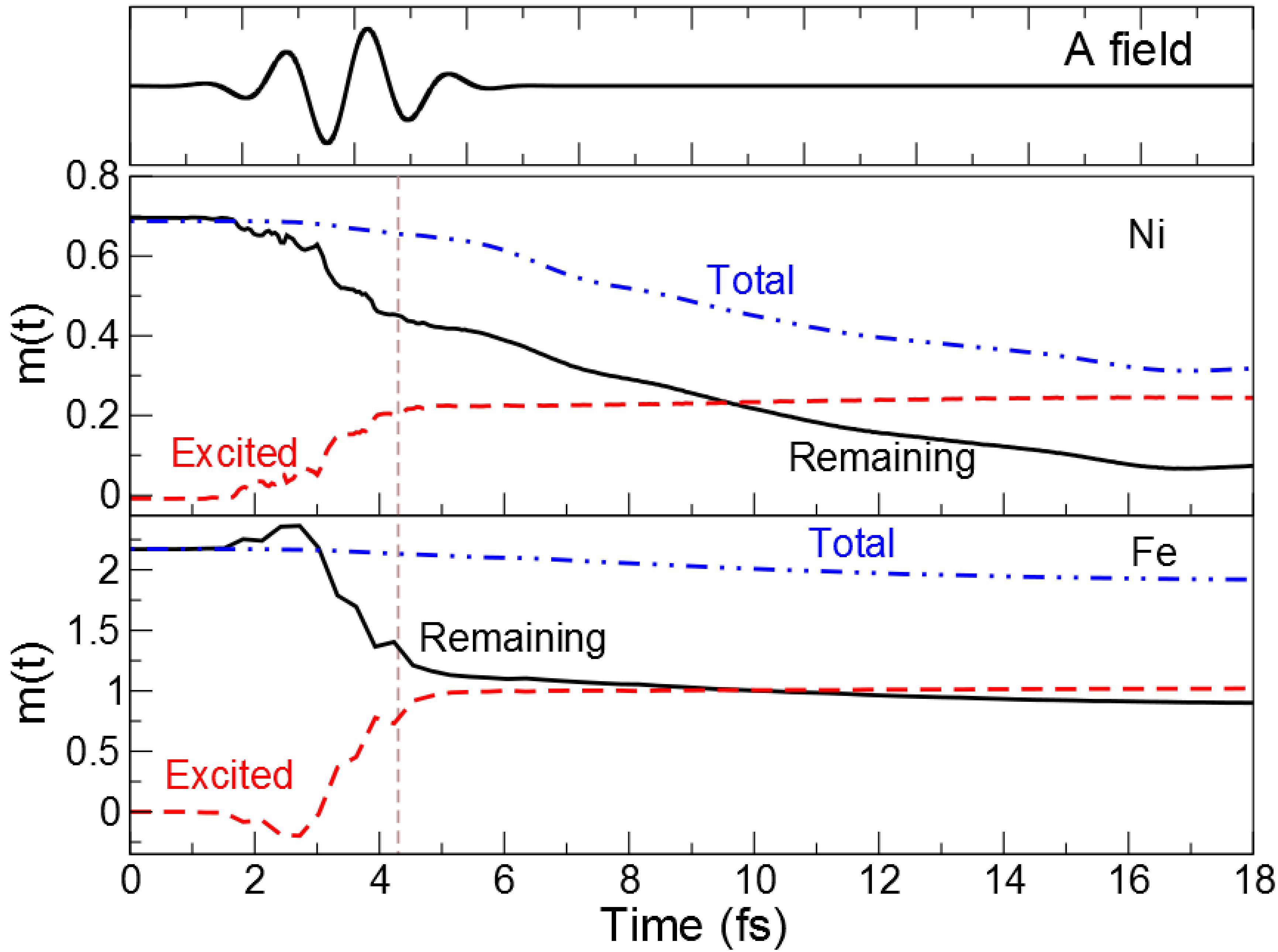
Global torque = 0, if $B_{ext} = 0$

(due to zero-torque theorem for B_{xc})



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



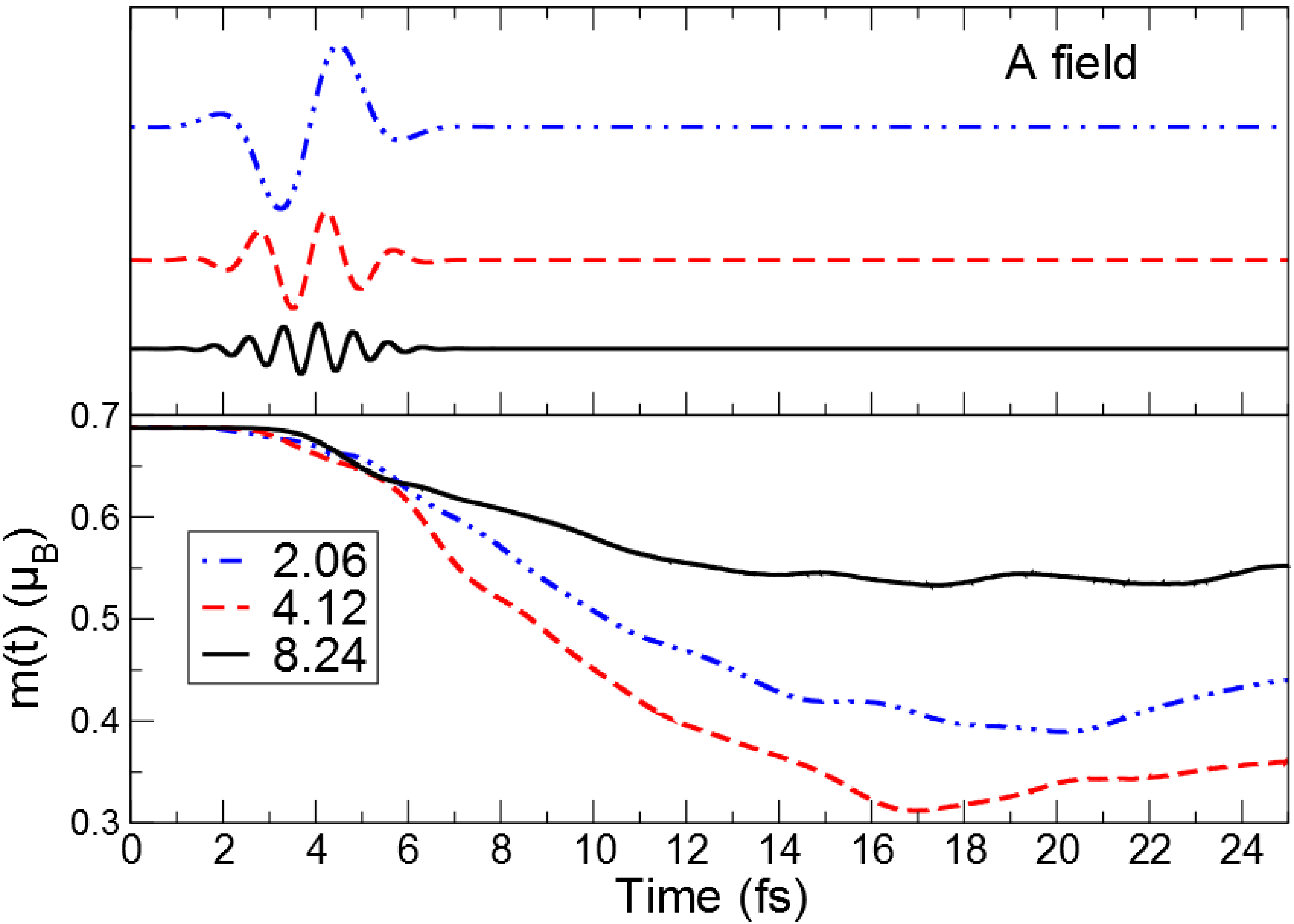
A field
Ni
Total
Excited
Remaining
Fe
Total
Remaining
Excited
m(t)
m(t)
0.8
0.6
0.4
0.2
0
2
1.5
1
0.5
0
0
2
4
6
8
10
12
14
16
18
Time (fs)

<u>Demagnetization occurs in two steps:</u>

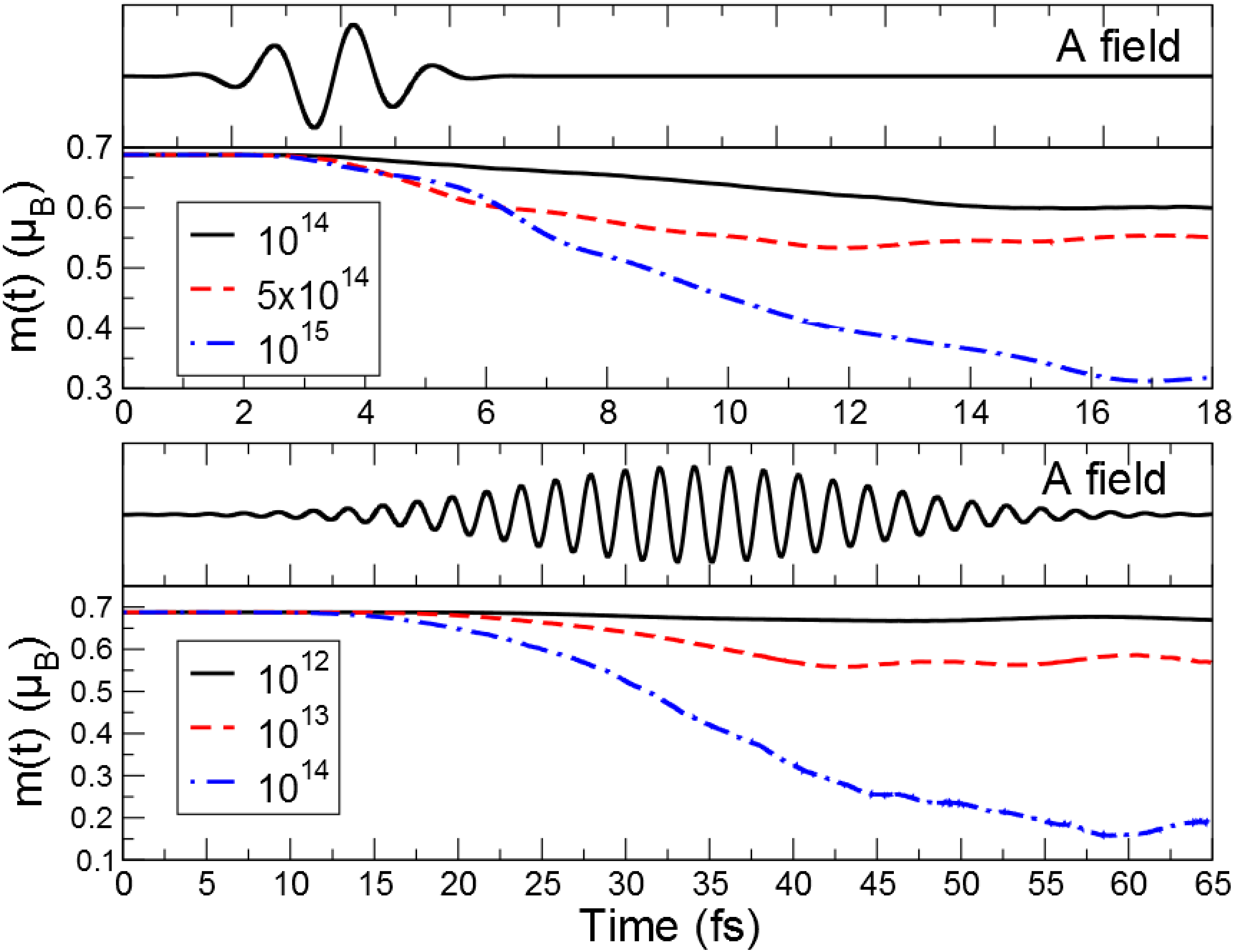
- Initial excitation by laser *moves* magnetization from atomic region into interstitial region. Total Moment is basically conserved during this phase.

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

Playing with laser parameters



A field
0.7
0.6
0.5
0.4
0.3
m(t) (μ_B)
2.06
4.12
8.24
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) (μB)
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) (μB)
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

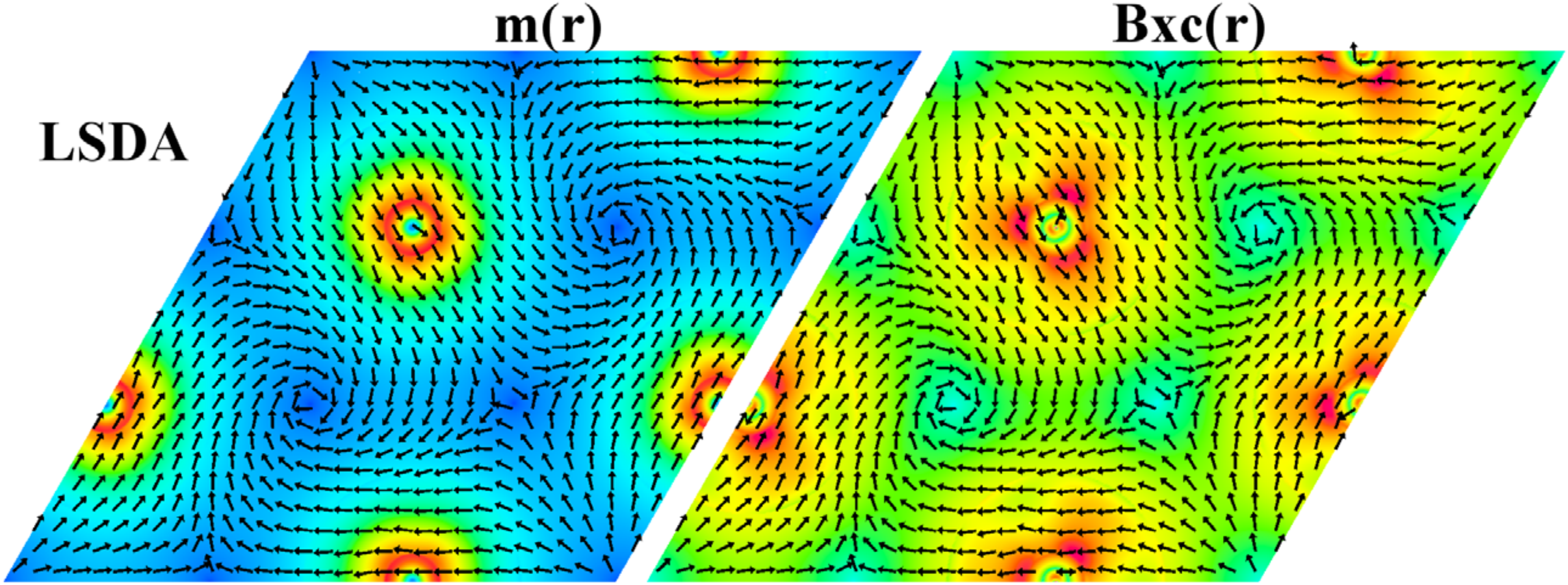
Step 1: Basic Theorems (Hohenberg-Kohn-Sham/ Runge-Gross)

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)

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

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)

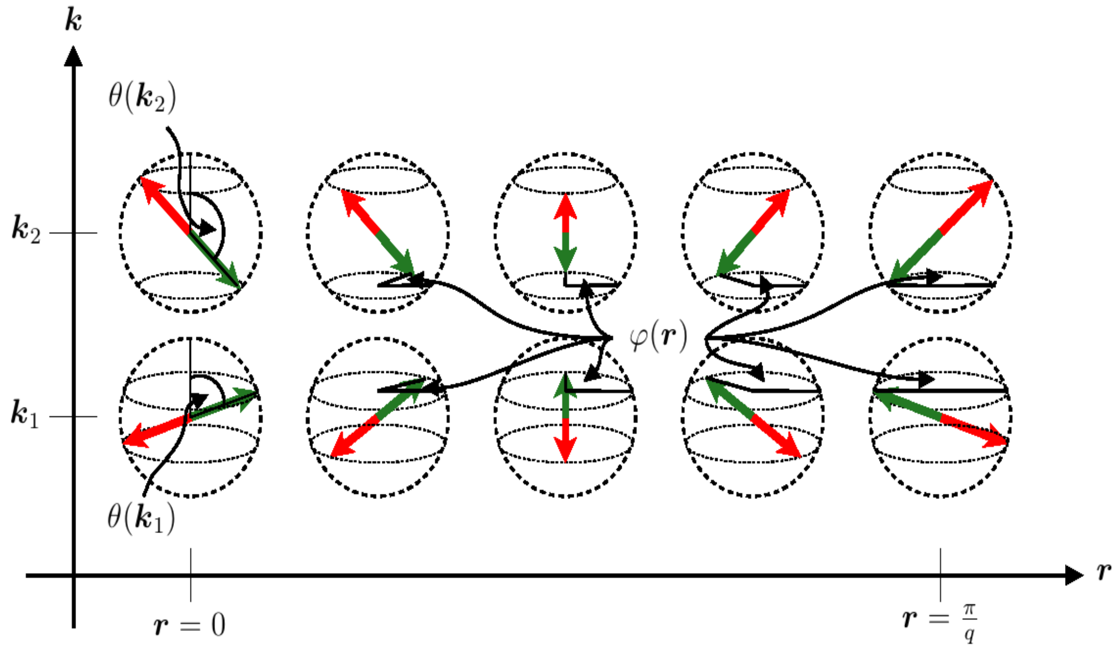


Illustration of spin spiral waves along one spatial coordinate for two different choices of wavevector $q=k_{1/2}$.

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

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

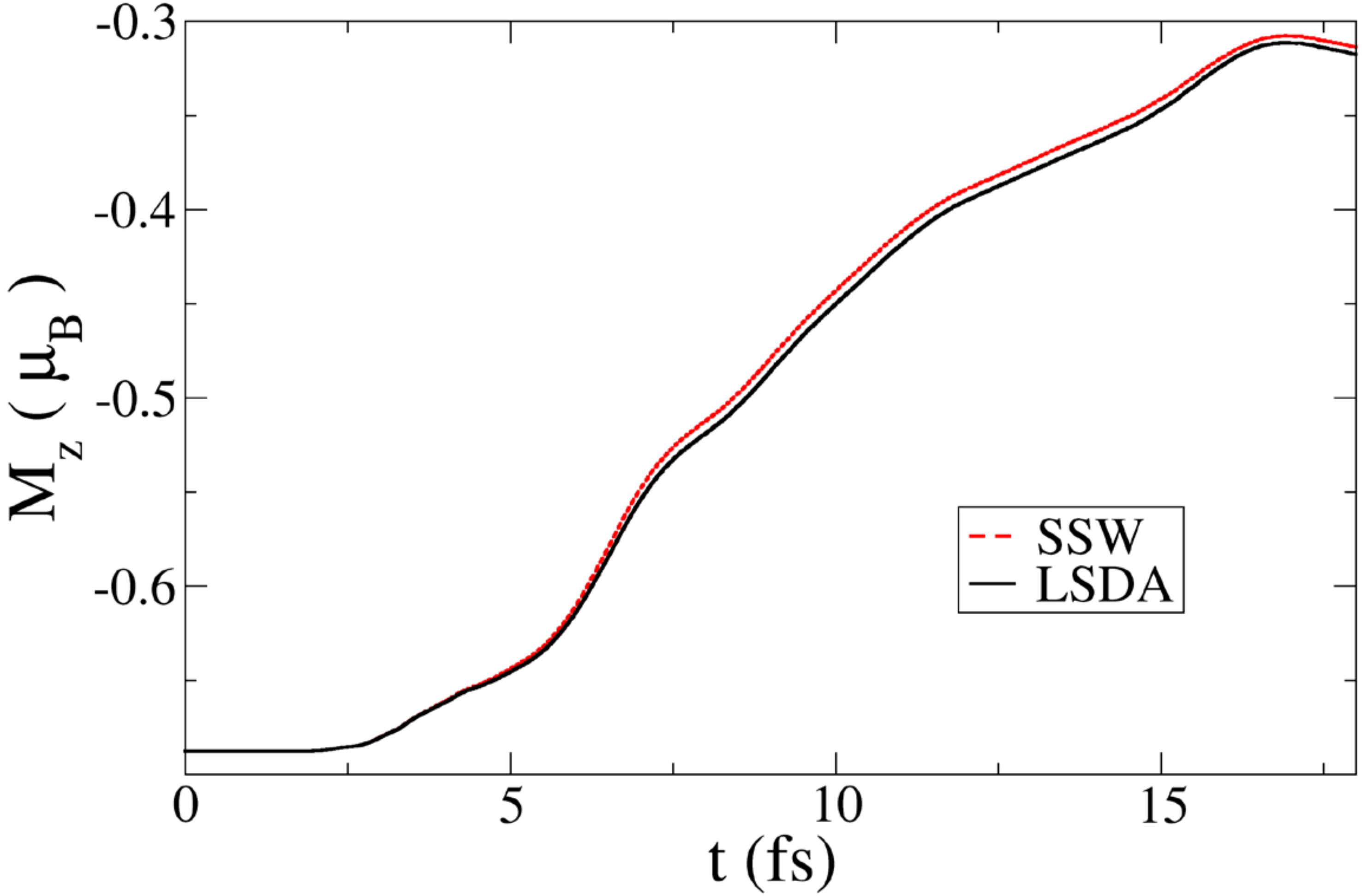
$$\varepsilon_{\mathrm{xc}}^{\mathrm{SSW}} = \varepsilon_{\mathrm{xc}}^{\mathrm{SSW}}(\mathrm{n},\mathrm{m},\mathrm{q},\mathrm{s})$$

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

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

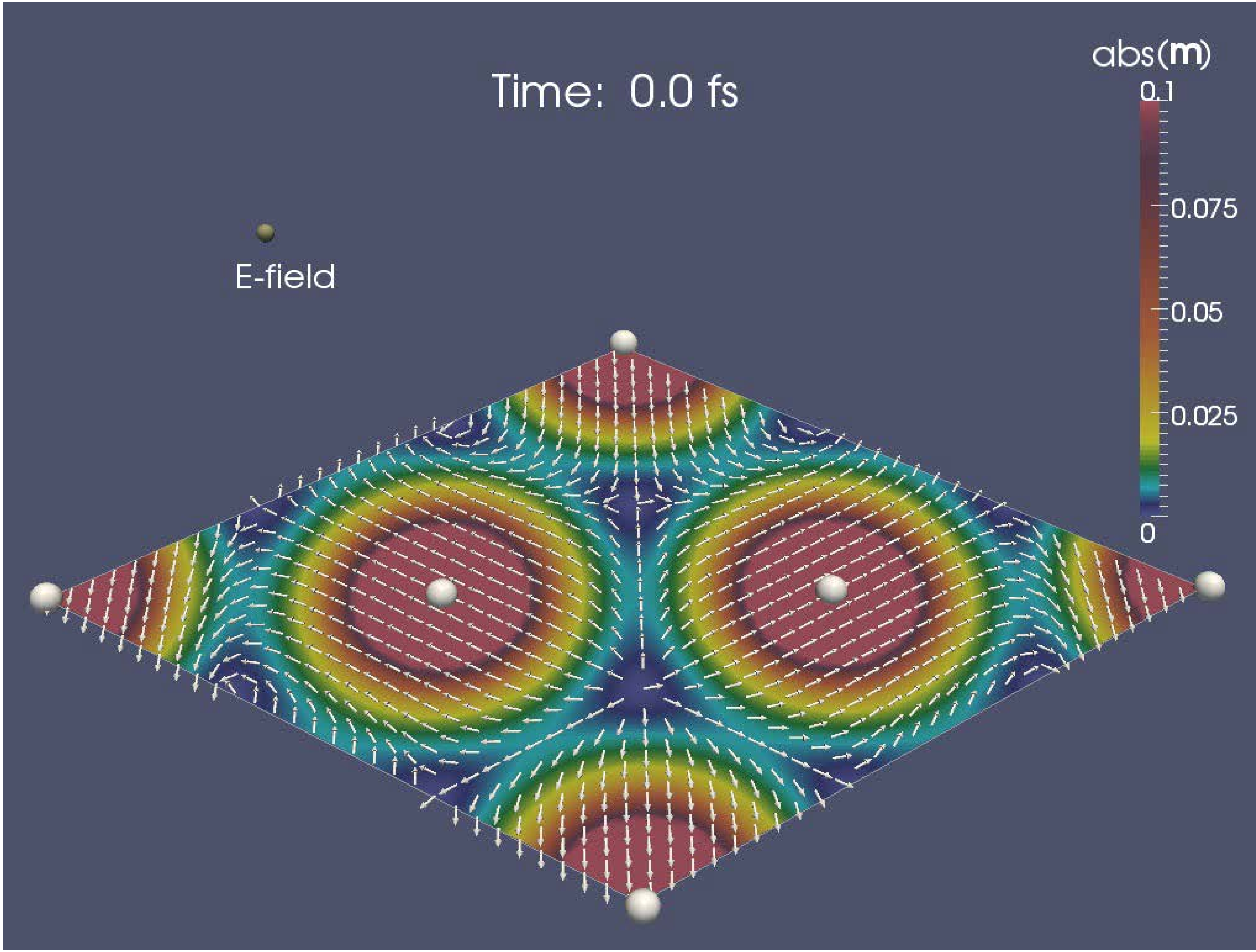
$$D_T\left(\mathbf{r}\right)=\left|\vec{m}\left(\mathbf{r}\right)\times\left(\nabla\otimes\vec{m}\left(\mathbf{r}\right)\right)\right|^2 \qquad d_T\left(\mathbf{r}\right)=\left|\vec{m}\left(\mathbf{r}\right)\times\left(\nabla^2\vec{m}\left(\mathbf{r}\right)\right)\right|^2$$

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

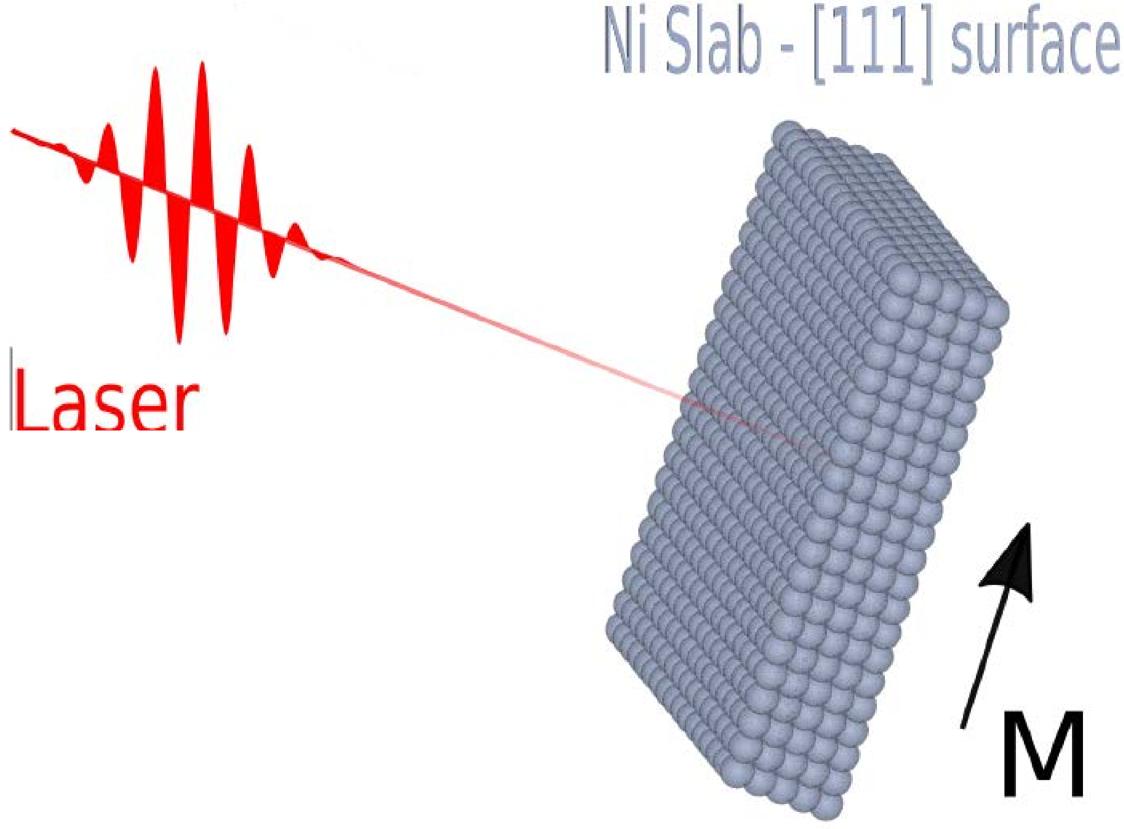


-0.3
-0.4
-0.5
-0.6
M_z (μ_B)
SSW
LSDA
0
5
10
15
t (fs)

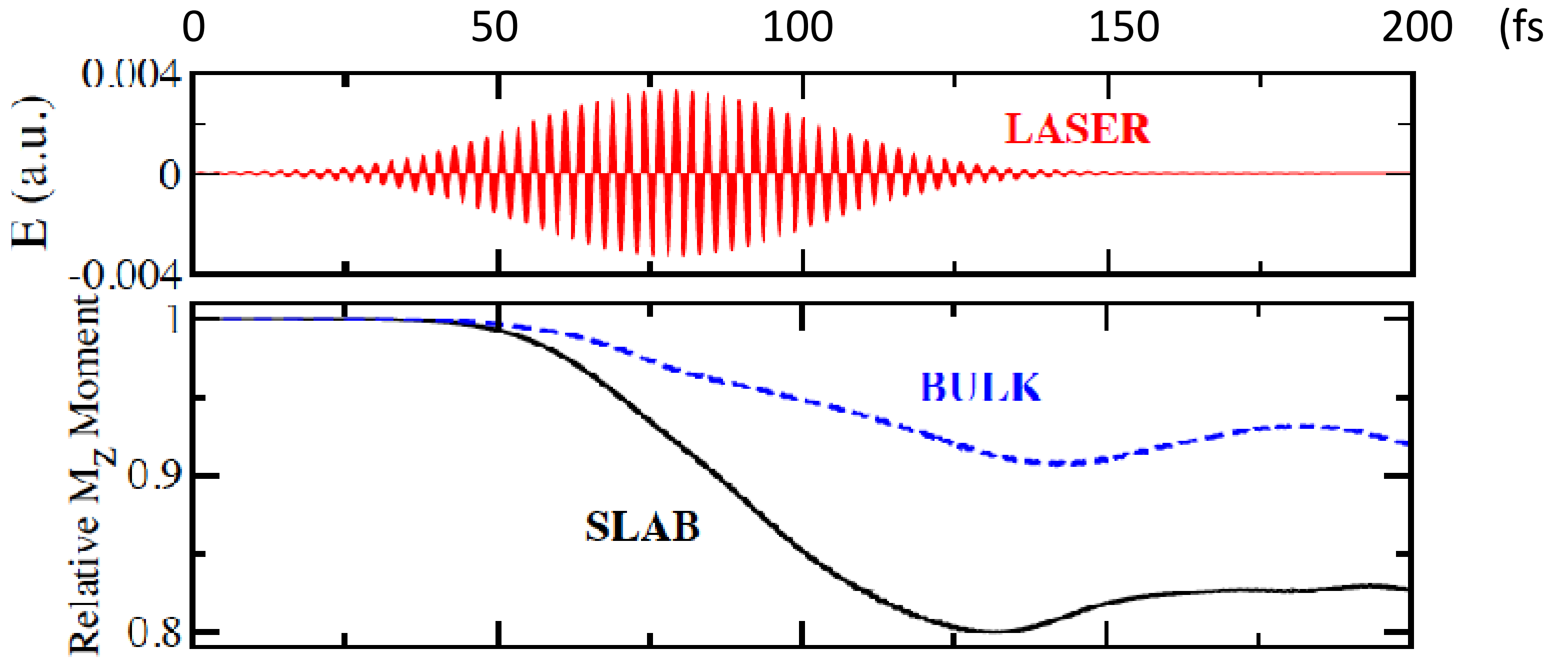
Beyond 3D bulk



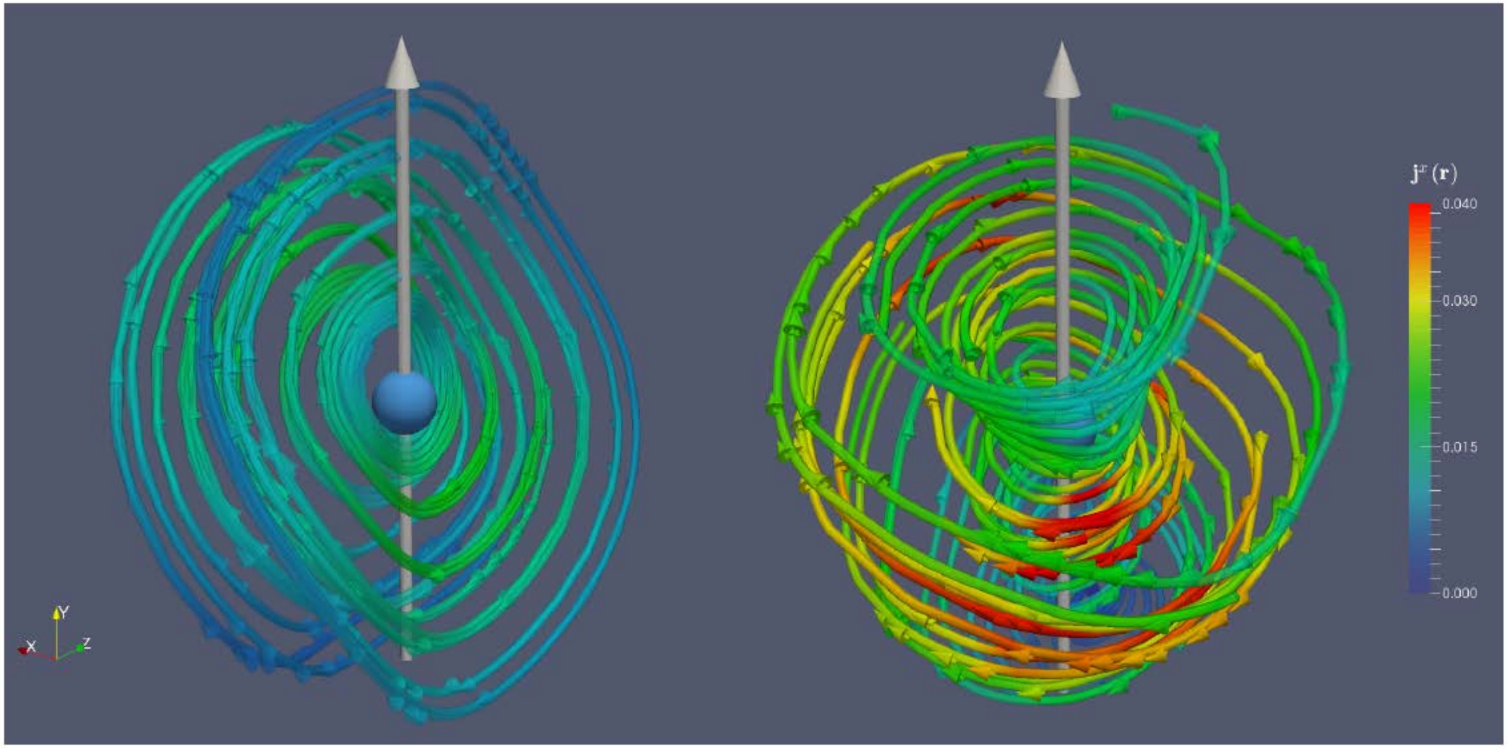
Cr monolayer



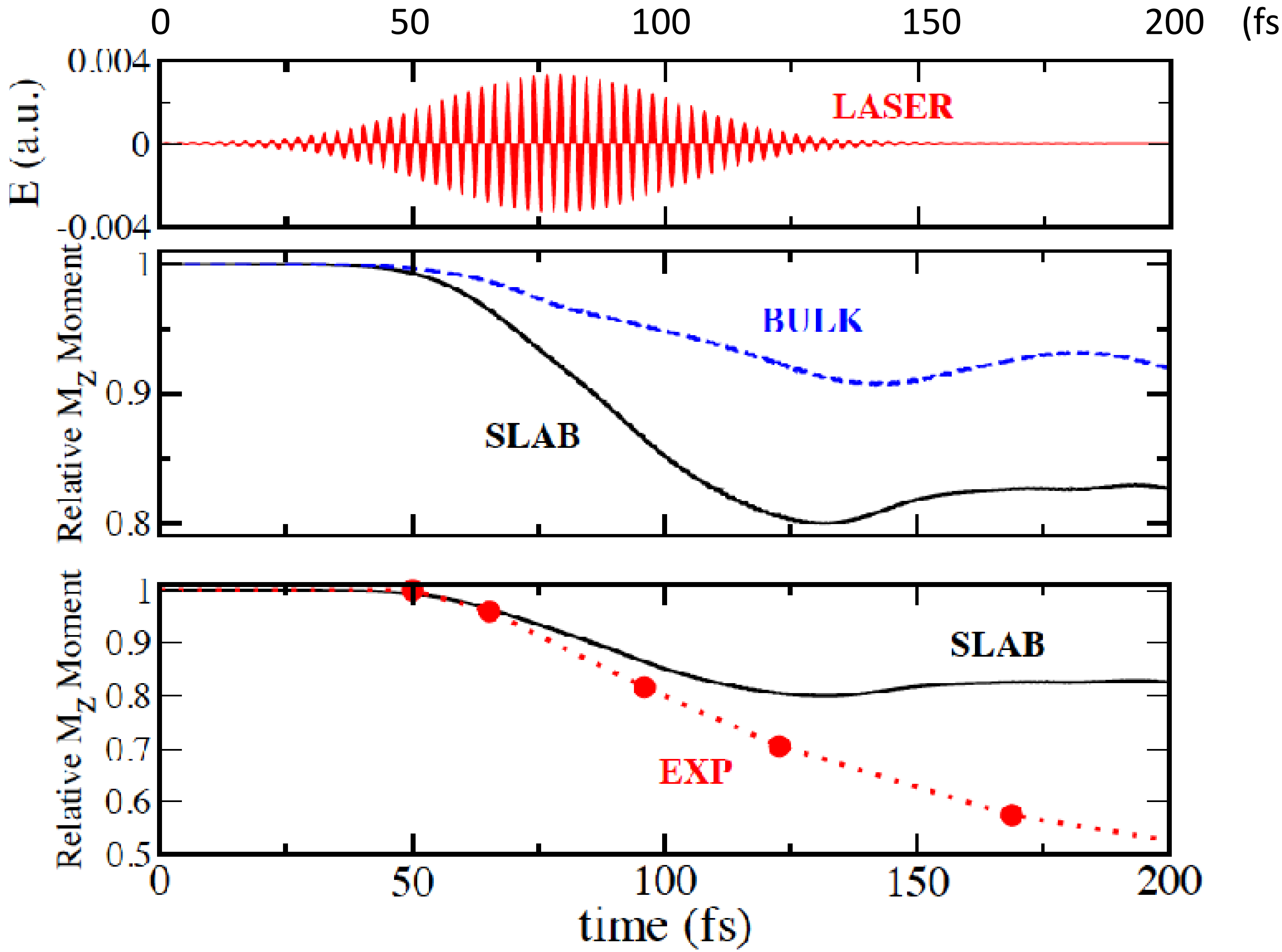
Ni Slab - [111] surface
Laser
M



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

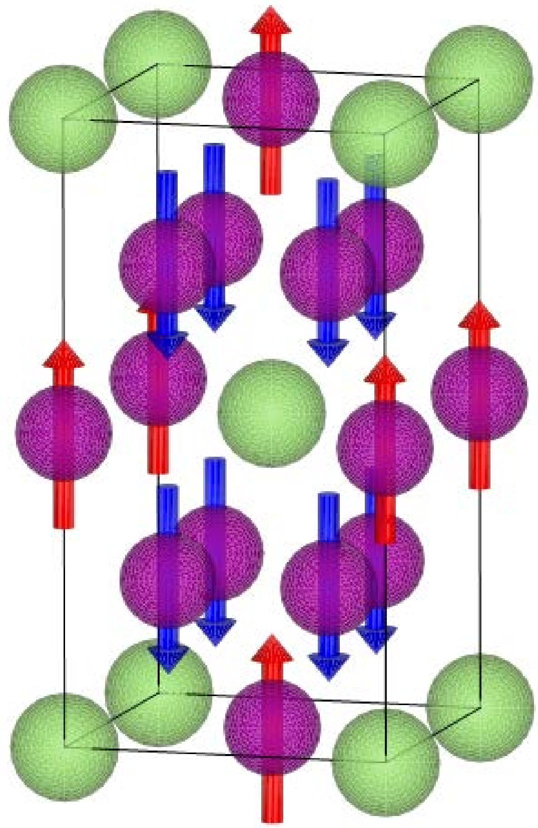


Streamlines for J_x, the spin-current vector field of the x component of spin, around a Ni atom in bulk (left) and for the outermost Ni atom in the slab (right).


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)

Heusler compounds

Mn_3Ga



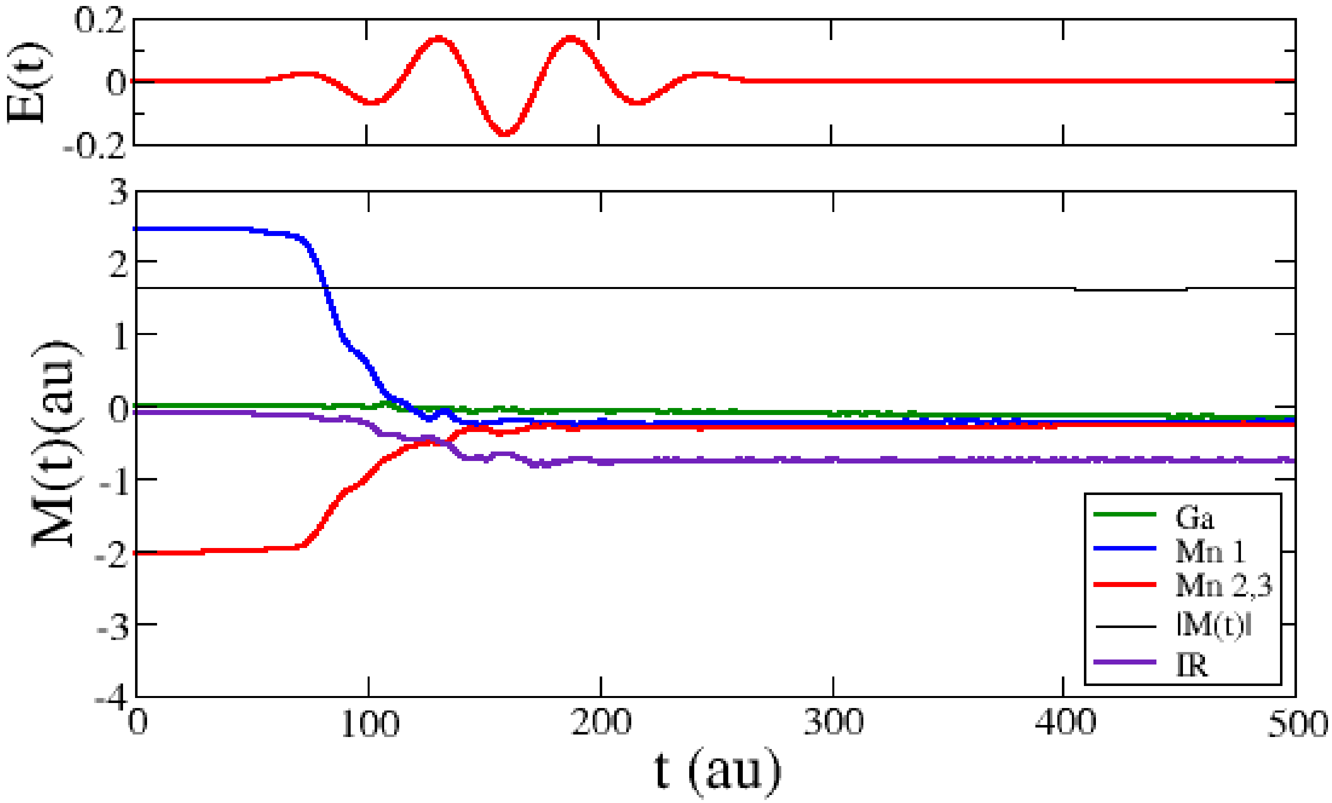
Ga

Mn

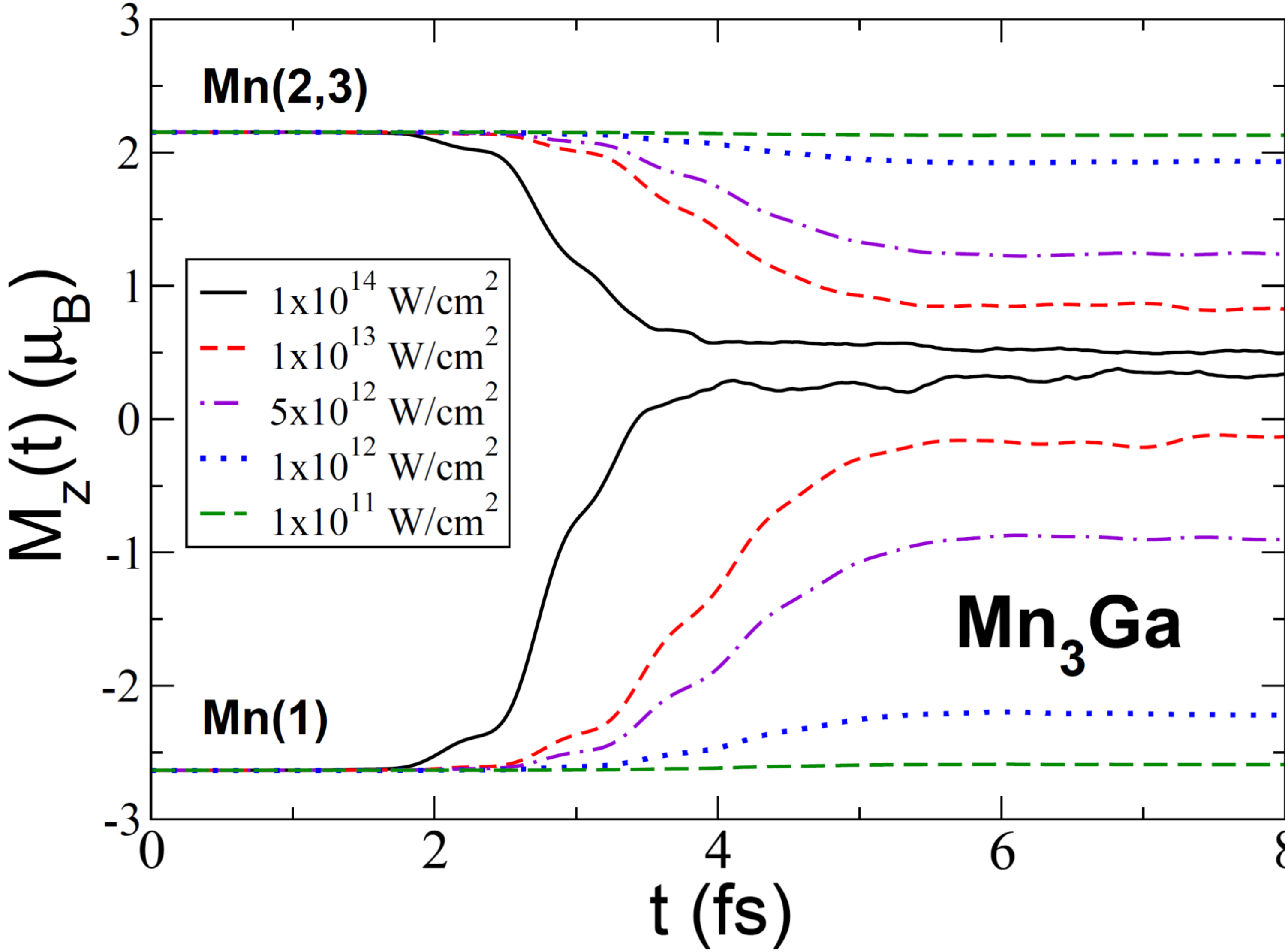
Mn_3Ga

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

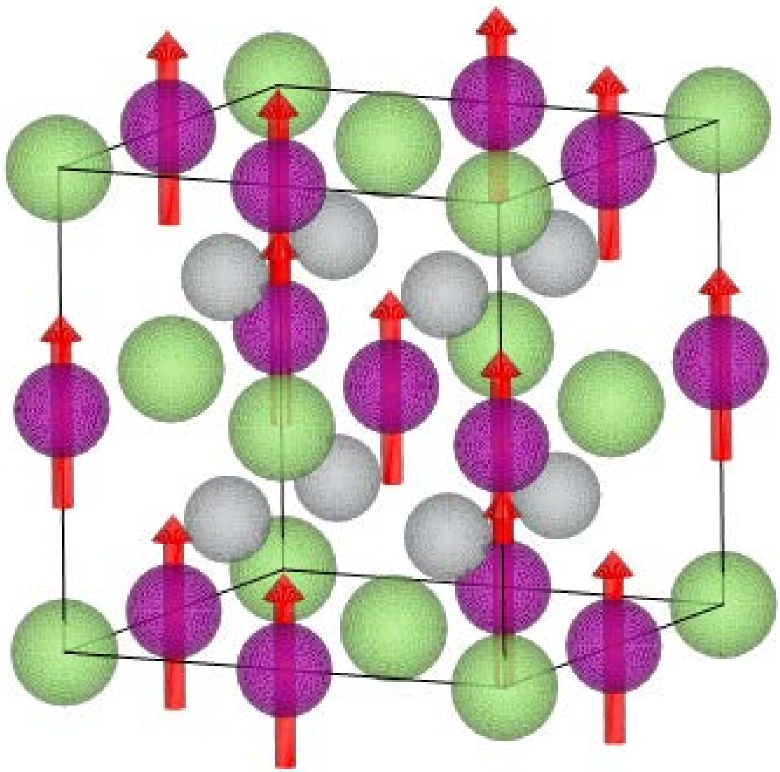


P. Elliott, T. Mueller, K. Dewhurst, S. Sharma, E.K.U.G., arXiv 1603.05603



Mn(2,3)
Mn(1)
Mn$_3$Ga
$1x10^{14}$ 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)

Ni_2MnGa

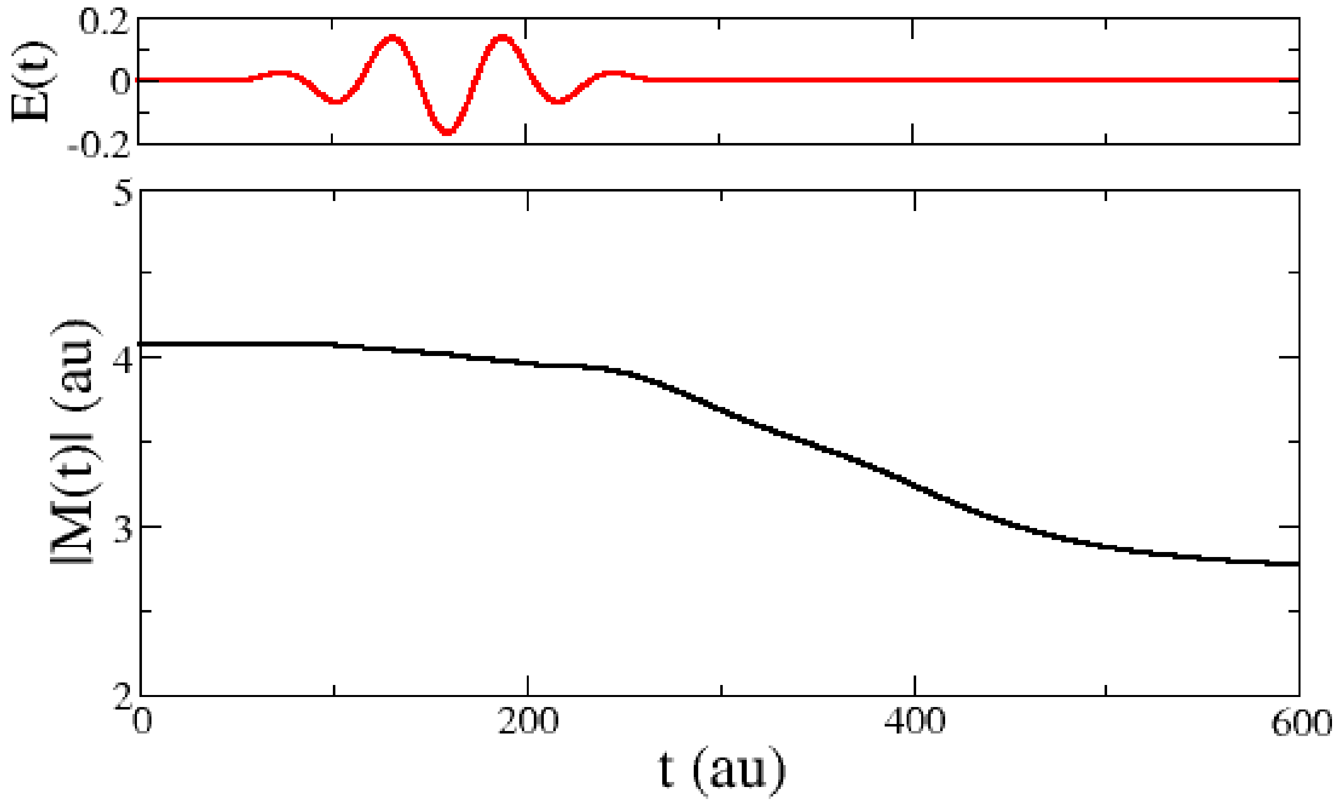


Ga	0.02 μB
Mn	-3.14 μB
Ni	-0.37 μB

Ni_2MnGa

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

See loss in global moment

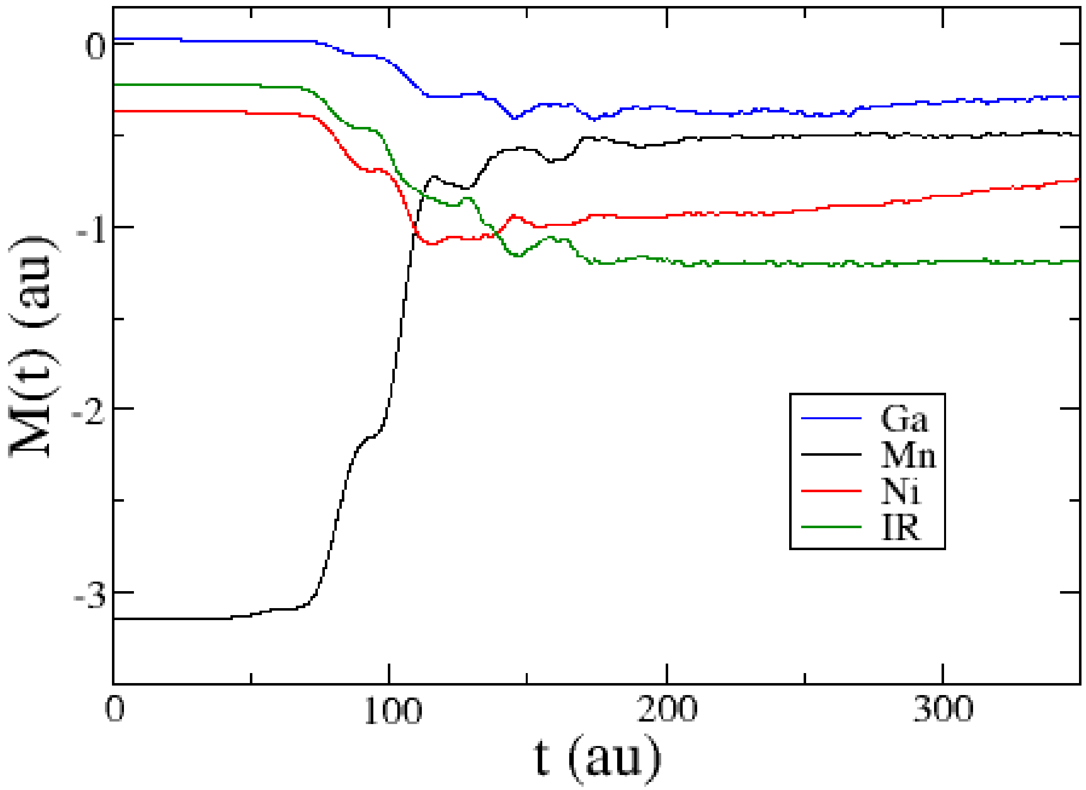


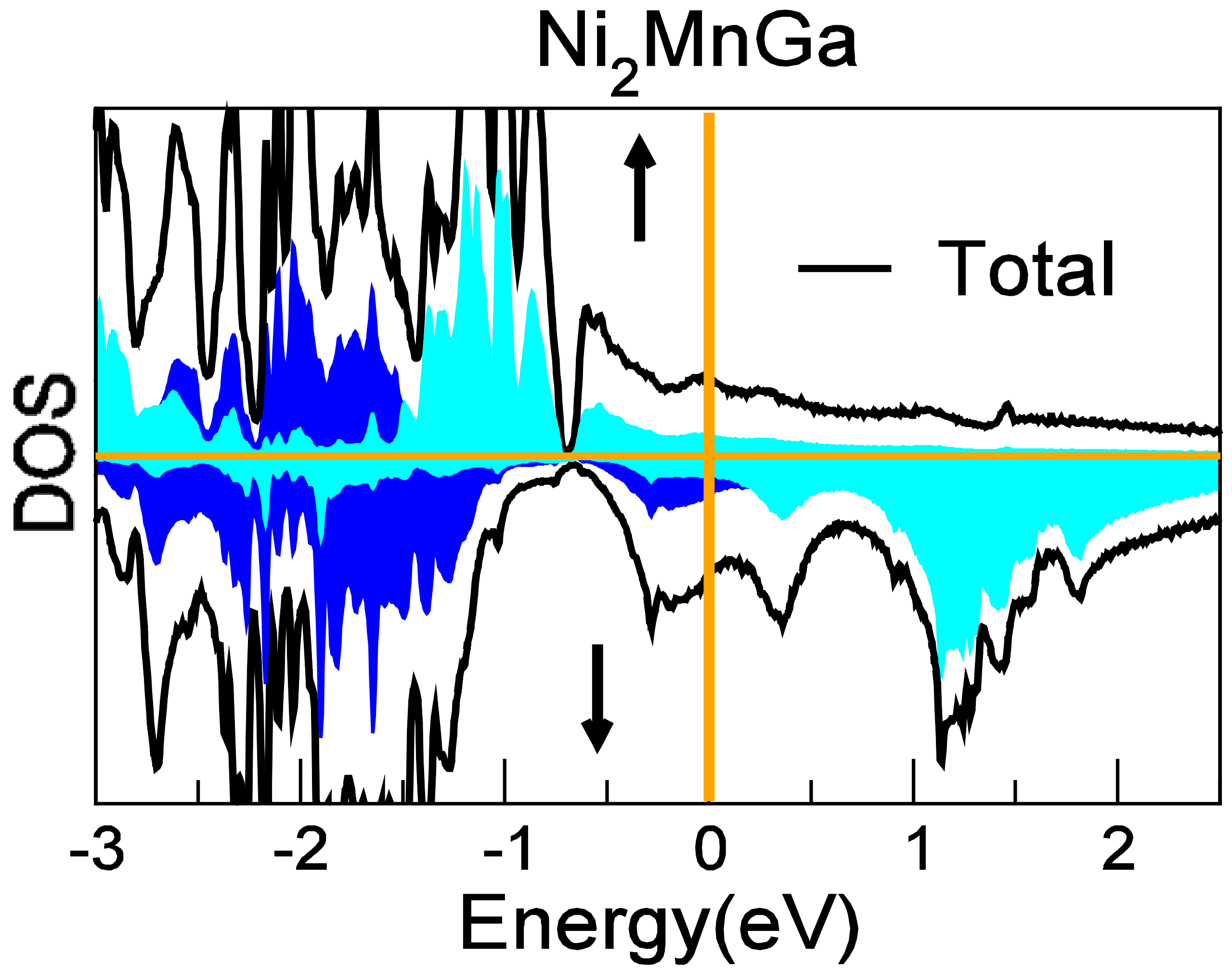
Ni_2MnGa

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

Summary

- No demagnetization without Spin-Orbit coupling

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

- No significant change in M_x and M_y

- New xc functional derived from spin-spiral phase of uniform e-gas yields results very similar to non-collinear LSDA

- Ultrafast transfer of spin moment between sublattices of Heusler compounds: Easily understood on the basis of the ground-state DOS

Coworkers

Kevin Krieger

Sangeeta Sharma

Florian Eich

Kay Dewhurst

Peter Elliott