Laser-induced spin dynamics in solids: Some predictions from real-time TDDFT

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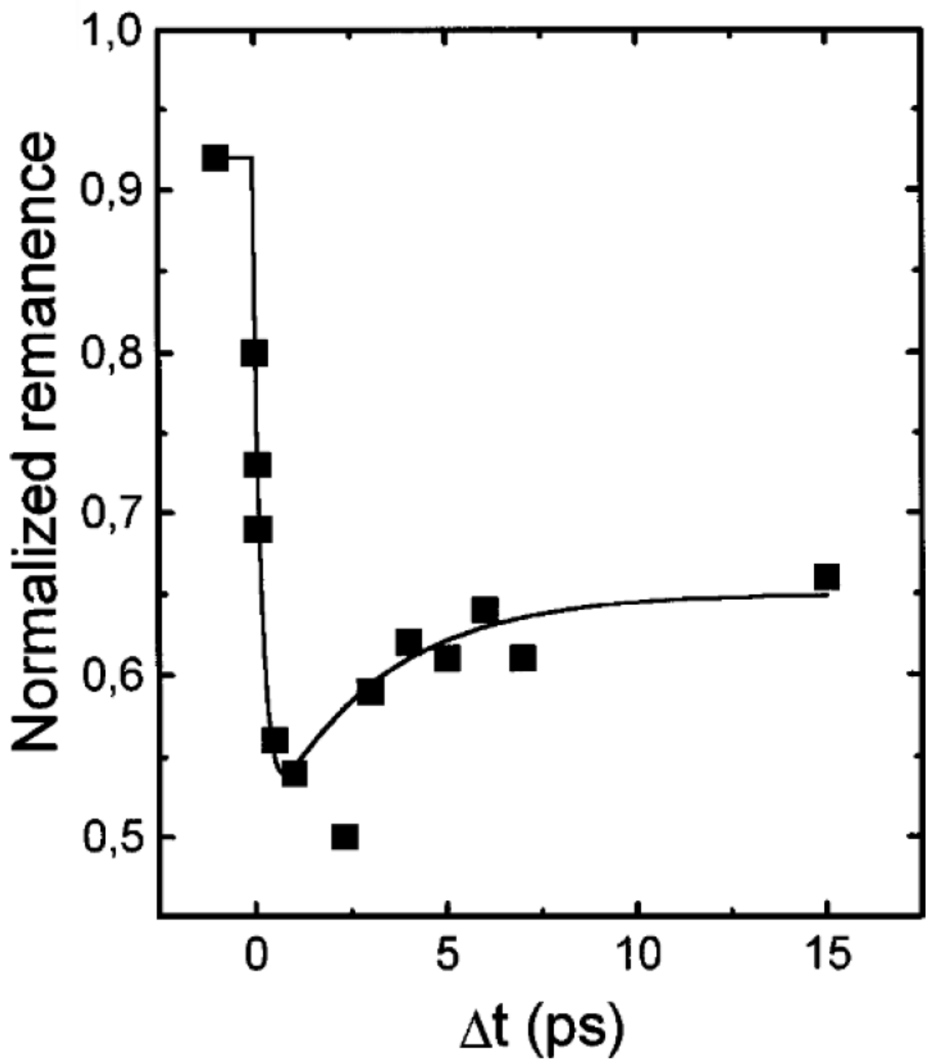
- Laser-induced demagnetisation (~ 50 fs)
  found experimentally in 1996,
  explained by TDDFT in 2015

- XC functionals for non-collinear magnetism

- Optically Induced Spin TRansfer OISTR (~ 5 fs)
  predicted by TDDFT in 2016
  found experimentally in 2018

First experiment on ultrafast laser induced demagnetization



Beaurepaire et al, PRL 76, 4250 (1996)

Possible mechanisms for demagnetisation

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

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- **Our proposal for the first 50 fs:**
  Laser-induced charge excitation followed by spin-orbit-driven demagnetization of the initially not excited electrons

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vector field of spin magnetization

$$\vec{m}(\vec{r}, t)$$

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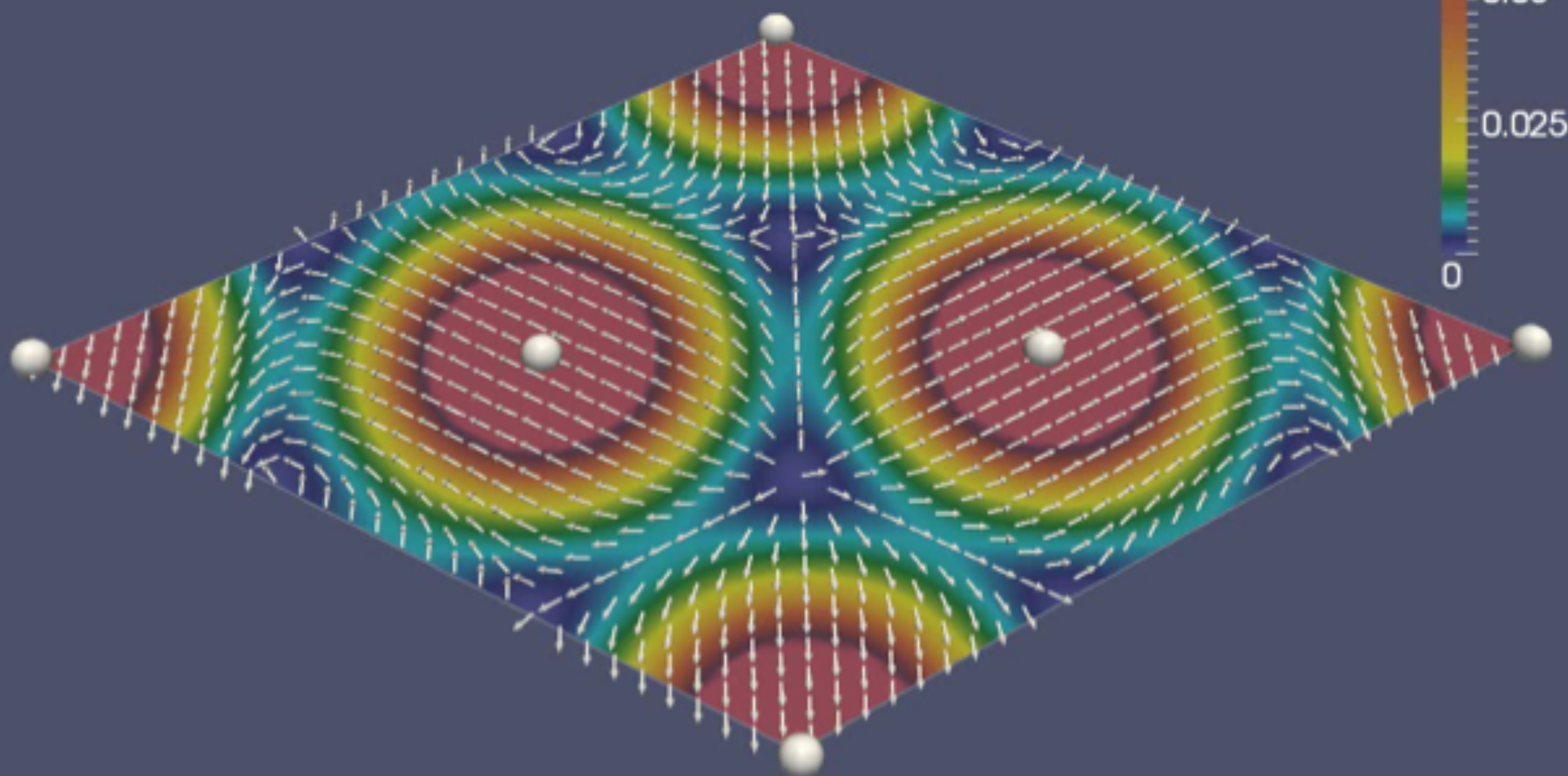

Cr monolayer in ground state

Theoretical Approach: Real-time non-collinear-spin TDDFT with TD spin-orbit coupling

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

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

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

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

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Universal functionals of ρ and m

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

$$n(\boldsymbol{r},t) = \sum_{j=1}^{N} \psi_j^{\dagger}(\boldsymbol{r},t)\,\psi_j(\boldsymbol{r},t)$$

$$\vec{\mathbf{m}}(\boldsymbol{r},t) = \sum_{j=1}^{N} \psi_j^{\dagger}(\boldsymbol{r},t)\,\vec{\boldsymbol{\sigma}}\,\psi_j(\boldsymbol{r},t)$$

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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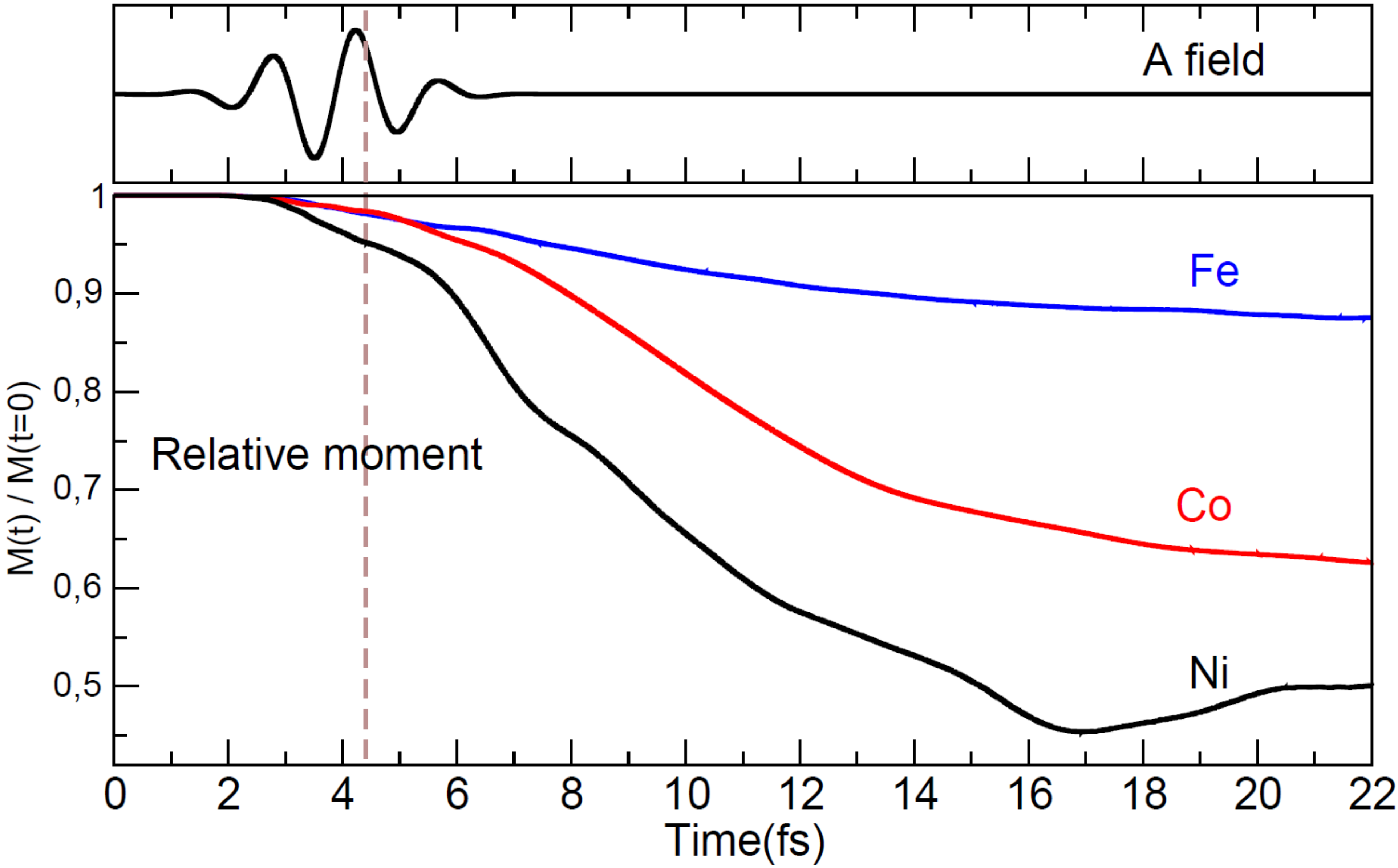
Demagnetisation in Fe, Co and Ni

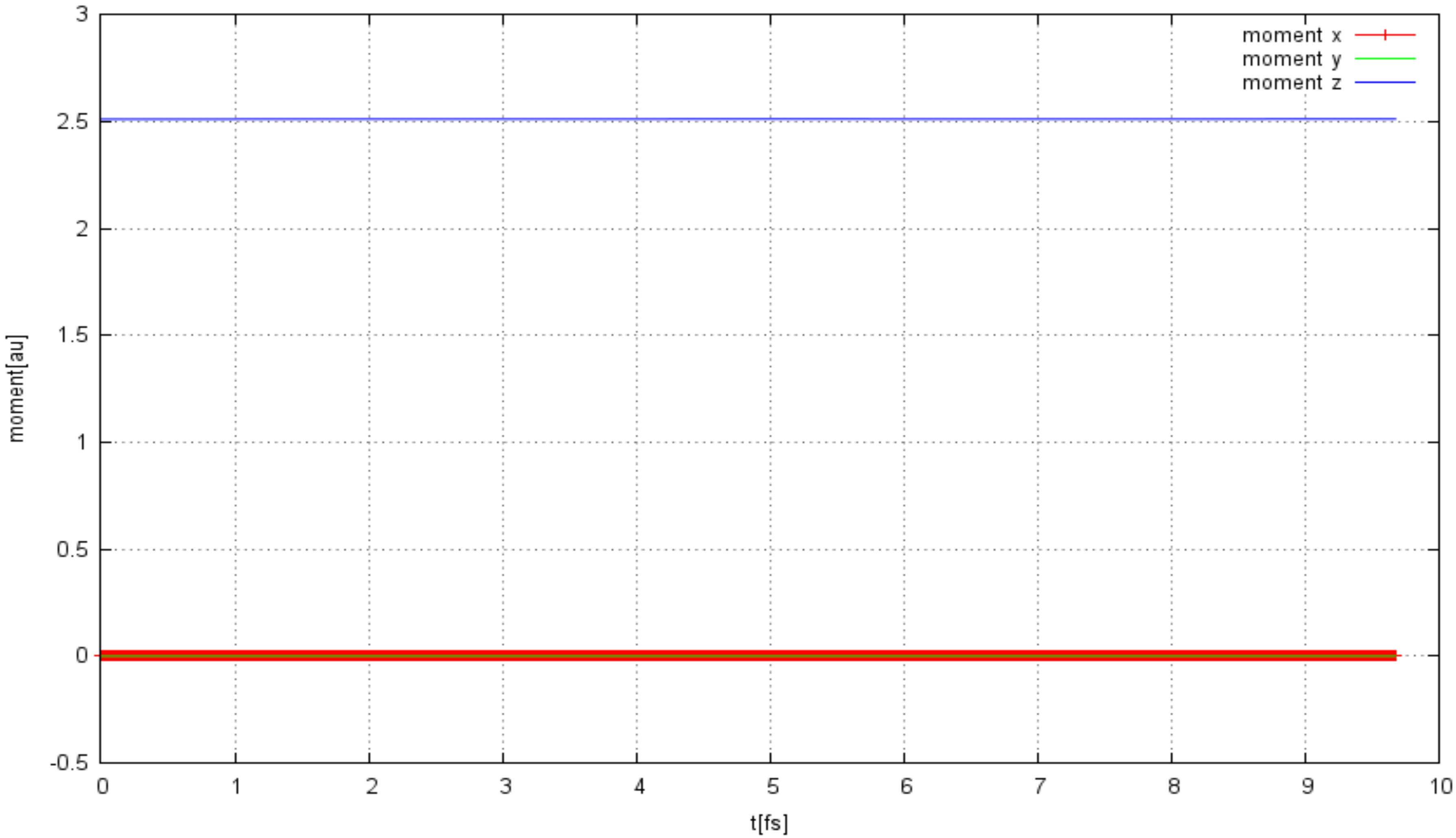


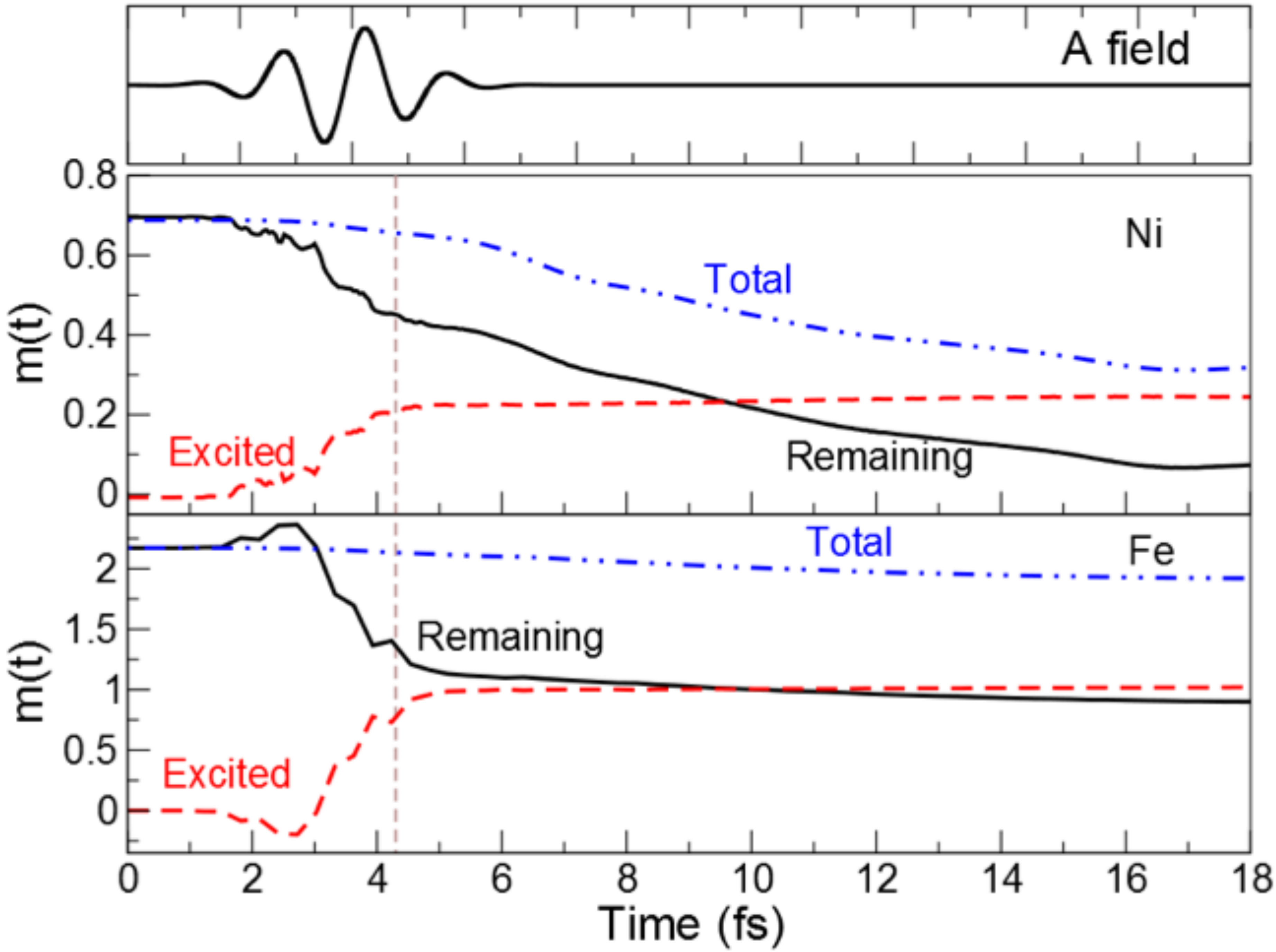
K. Krieger, K. Dewhurst, P. Elliott, S. Sharma, E.K.U.G., JCTC 11, 4870 (2015)

Analysis of the results

Calculation without spin-orbit coupling

components of spin moment



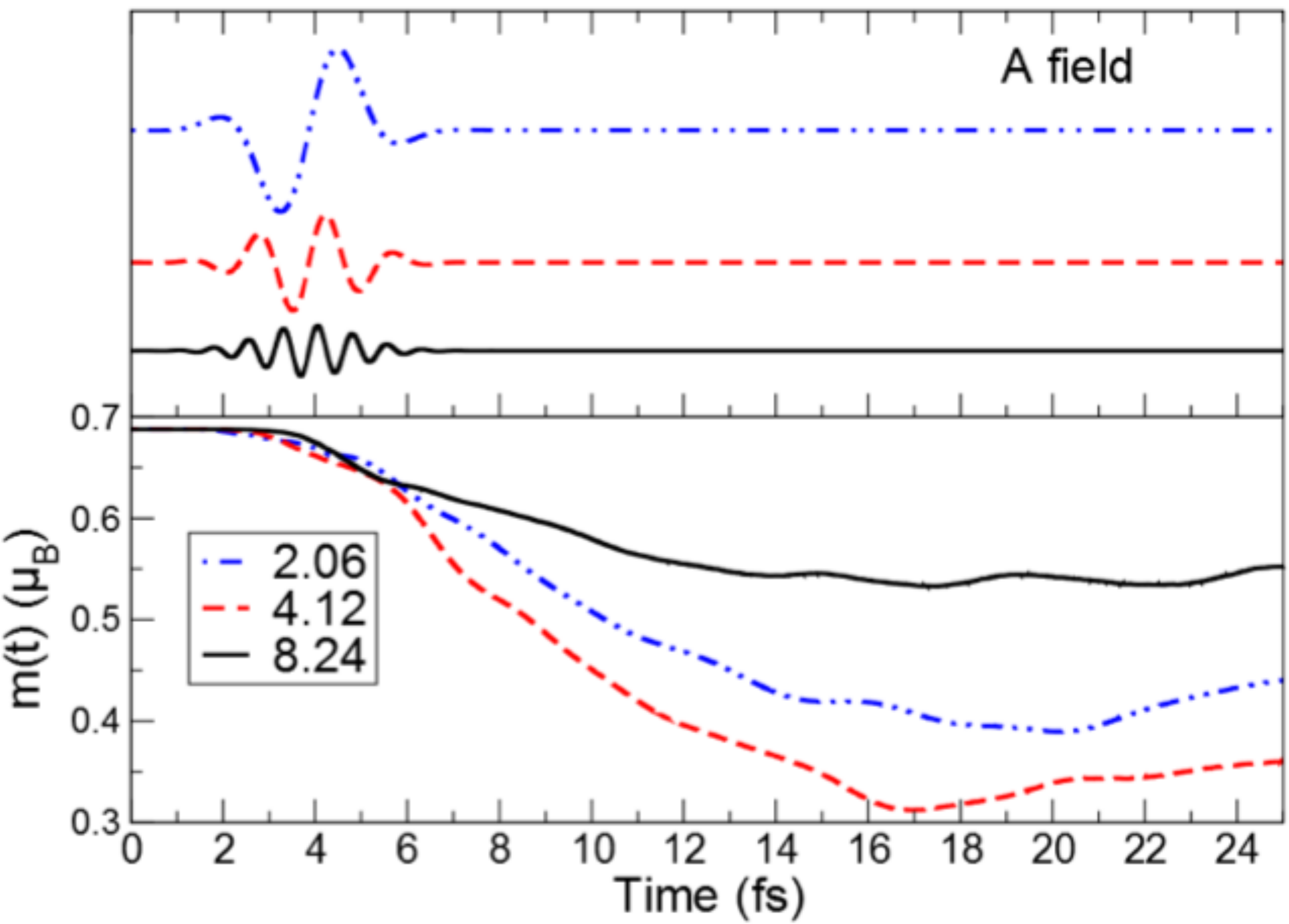

A field
0.8
0.6
0.4
0.2
0
m(t)
Ni
Total
Excited
Remaining
2
1.5
1
0.5
0
m(t)
Fe
Total
Remaining
Excited
0
2
4
6
8
10
12
14
16
18
Time (fs)

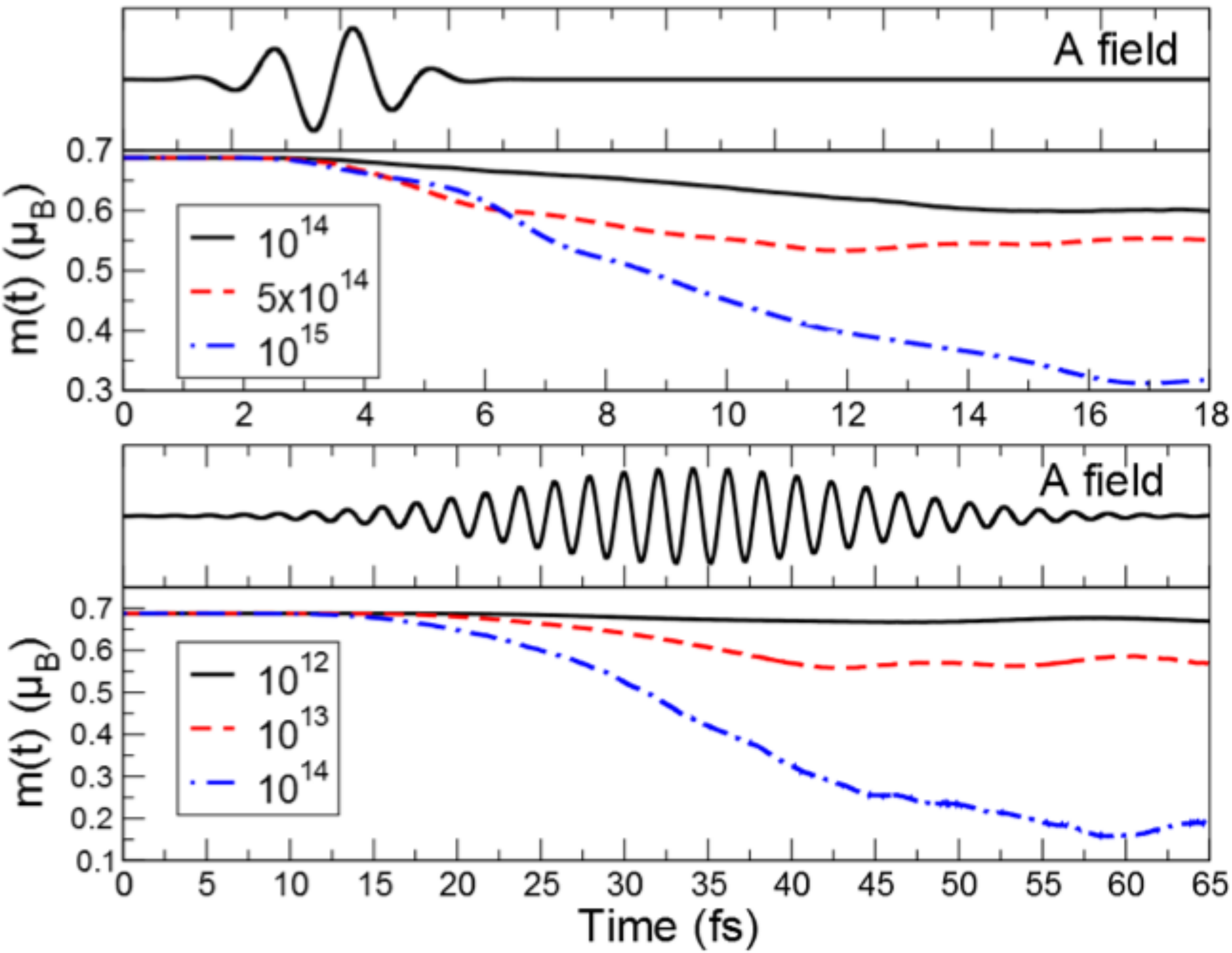
Demagnetization occurs in two steps:

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

- Spin-Orbit coupling 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) (μB)
2.06
4.12
8.24
0
2
4
6
8
10
12
14
16
18
20
22
24
Time (fs)


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