Time-dependent density functional theory

OUTLINE

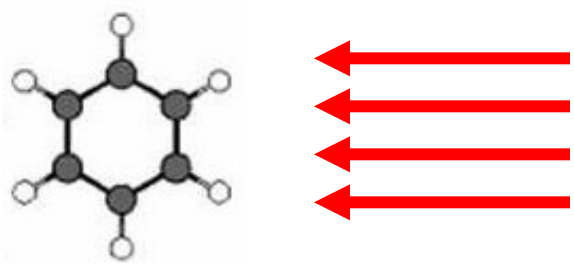
- **Basic theorems of TDDFT**

- **Linear response regime:**
  -- Calculation of photo-absorption spectra

- **Beyond the linear regime:**
  -- Laser-driven spin dynamics

What do we want to describe?

System in laser field:
Generic situation



$$\hat{H}(t) = \hat{T}_e + \hat{T}_n + \hat{W}_{ee} + \hat{W}_{nn} + \sum_{j,\alpha} - \frac{Z_\alpha e^2}{|r_j - R_\alpha|} + \vec{r}_j \cdot \vec{E}(t) \cdot \sin \omega t$$

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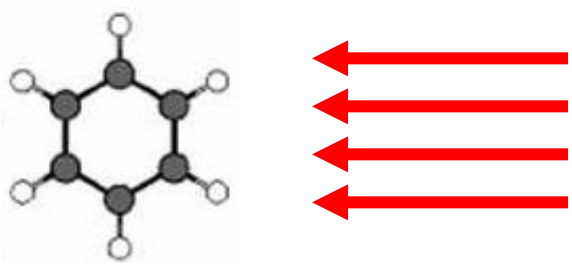

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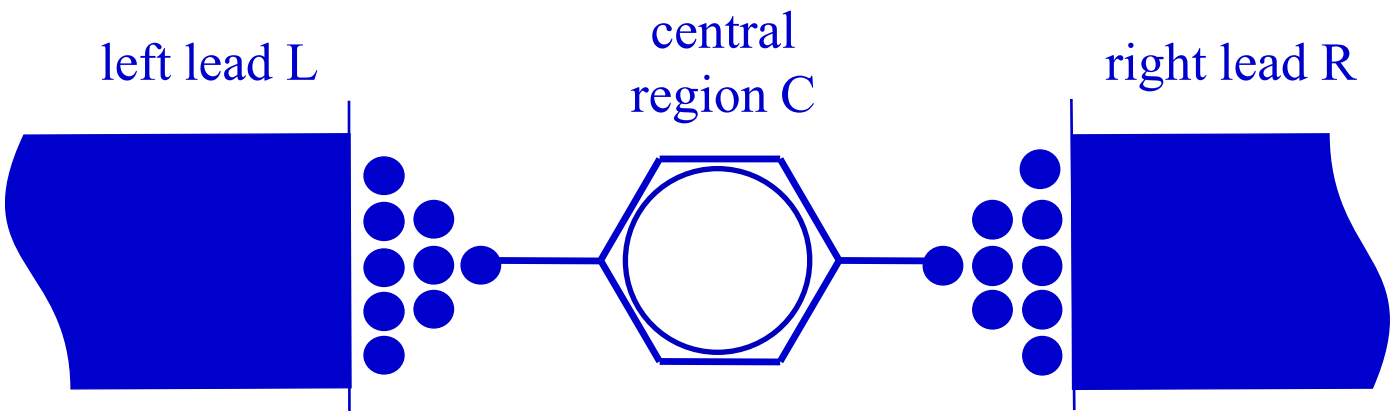
Electronic transport: Generic situation



Bias between L and R is turned on: U(t) ⟶ V

$$\hat{H}(t) = \hat{T}_e + \hat{W}_{ee} + \sum_{j,\alpha} - \frac{Z_\alpha e^2}{|r_j - R_\alpha|} + \vec{r}_j \cdot \vec{E}(t) \cdot \sin \omega t$$

Strong laser ($v_{laser}(t) \geq v_{en}$) :

Non-perturbative solution of full TDSE required

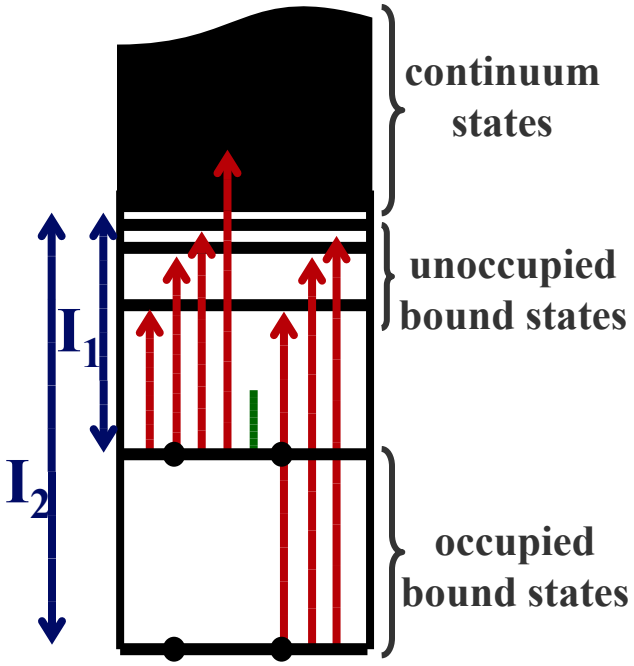
Weak laser ($v_{laser}(t) << v_{en}$) :

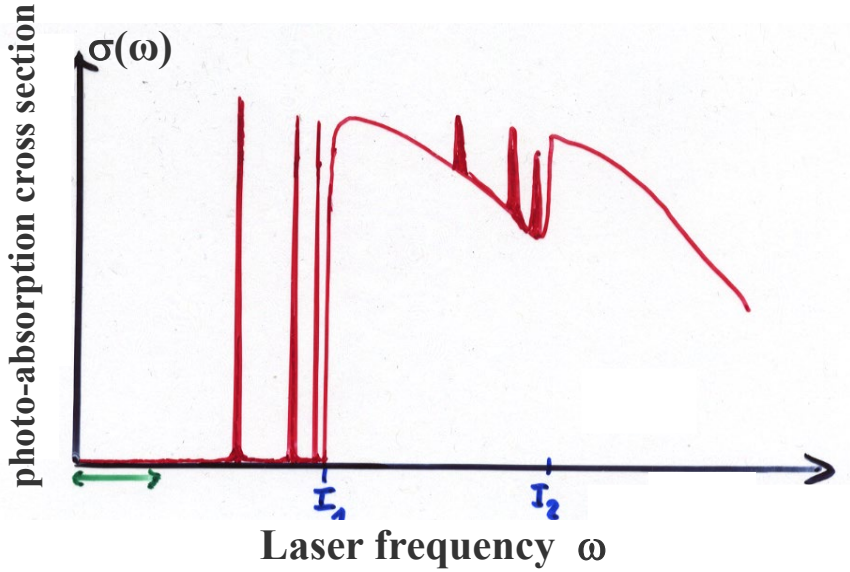
Calculate 1. Linear density response $\rho_1(\vec{r}\, t)$

2. Dynamical polarizability $\alpha(\omega) = -\frac{e}{E}\int z\, \rho_1(\vec{r}, \omega) d^3 r$

3. Photo-absorption cross section $\sigma(\omega) = -\frac{4\pi\omega}{c} \mathrm{Im}\, \alpha$

Photo-absorption in weak lasers





Basic theorems of TDDFT

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

1-1 correspondence (TD analogue of Hohenberg-Kohn theorem):

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

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

TDKS 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

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

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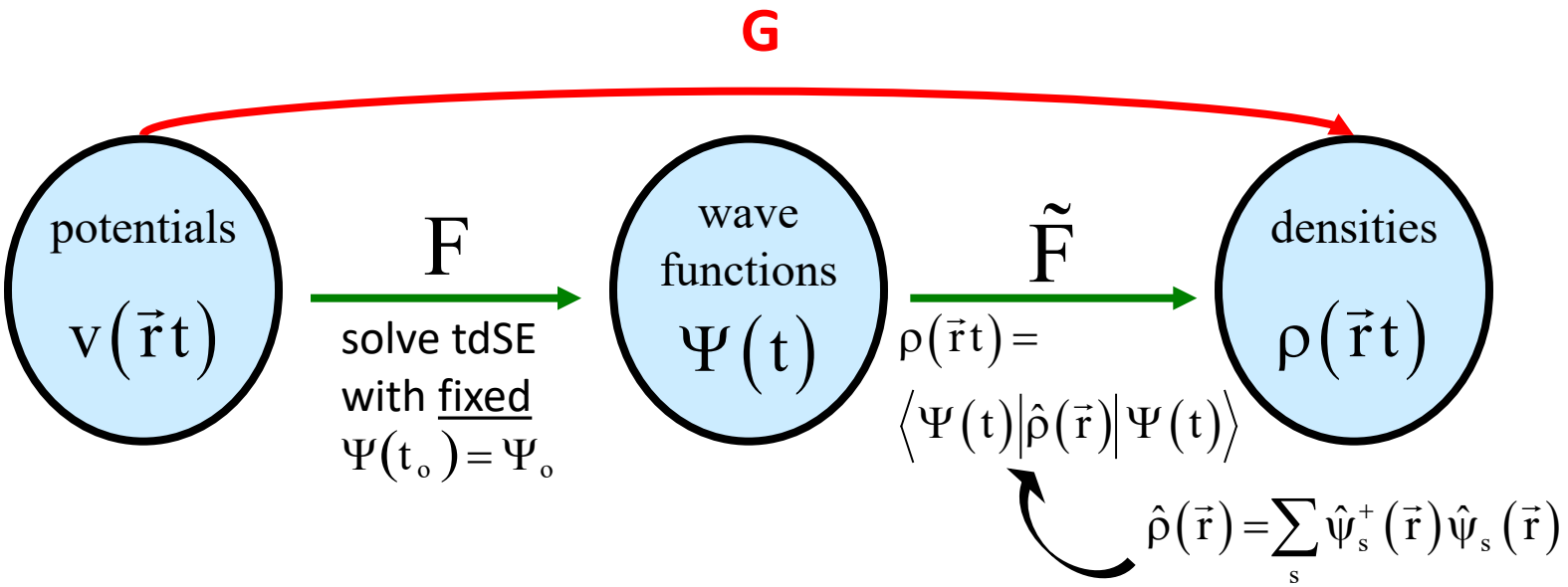
Proof of the 1-1 correspondence between $v(\vec{r}\,t)$ and $\rho(\vec{r}\,t)$

define maps $F: \; v(\vec{r}\,t) \mapsto \Psi(t)$ $\tilde{F}: \; \Psi(t) \mapsto \rho(\vec{r}\,t)$



$G: \; v(\vec{r}\,t) \mapsto \rho(\vec{r}\,t)$

to be shown that G is invertible for fixed Ψo

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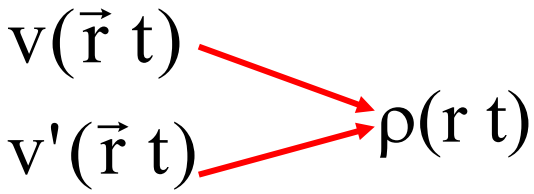


is impossible

i.e. $v(\vec{r}\ t) \neq v'(\vec{r}\ t) + c(t) \Rightarrow \rho(\vec{r}\ t) \neq \rho'(\vec{r}\ t)$

proof (basic idea):

$$v(\vec{r}\ t) \longrightarrow \vec{j}(\vec{r}\ t) \longrightarrow \rho(\vec{r}\ t)$$

$$v'(\vec{r}\ t) \longrightarrow \vec{j}'(\vec{r}\ t) \longrightarrow \rho'(\vec{r}\ t)$$

use

$$i\frac{\partial \vec{j}(\vec{r},t)}{\partial t} = \left\langle \Psi(t) \left| \left[\hat{j}(\vec{r}) \, , \, \hat{H}(t) \right] \right| \Psi(t) \right\rangle$$

equation of motion for $\vec{j}$

and

$$\frac{\partial \rho(\vec{r},t)}{\partial t} = - \operatorname{div} j(\vec{r},t)$$

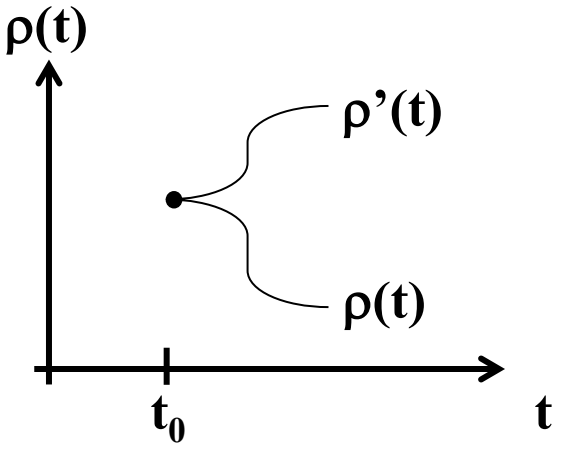
continuity equation

to show that there exists an integer k>0 such that

$$\left.\frac{\partial^k \rho}{\partial t^k}\right|_{t_0} \neq \left.\frac{\partial^k \rho'}{\partial t^k}\right|_{t_0}$$

$\Rightarrow$ ρ and ρ' will become different from each other infinitesimally later than t_0

This demonstrates uniqueness of the potential yielding a given TD density



The existence of a potential yielding a given TD density (TD v-representability) can also be shown (R. van Leeuwen, PRL 82, 3863 (1999) for arbitrary interaction. For the case of no interaction this guarantees the existence of the TD Kohn-Sham potential (non-interacting v-representability).

Simplest possible approximation for $v_{xc}[\rho](\vec{r}t)$

Adiabatic Local Density Approximation (ALDA)

$$v_{xc}^{ALDA}(\vec{r}\,t) := v_{xc,stat}^{hom}(n)\Big|_{n=\rho(\vec{r}\,t)}$$

$v_{xc,stat}^{hom}$ **= xc potential of static homogeneous e-gas**

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Any approximate ground-state-DFT xc functional can be used to generate an adiabatic approximation for TDDFT

$$v_{xc}^{adiab}(\vec{r}\,t) := v_{xc,gs}^{approx}[n]\Big|_{n=\rho(\vec{r}\,t)}$$

LINEAR RESPONSE THEORY

$t = t_0$: Interacting system in ground state of potential $v_0(r)$ with density $\rho_0(r)$

$t > t_0$: Switch on perturbation $v_1(r\,t)$ (with $v_1(r\,t_0)=0$).

Density: $\rho(r\,t) = \rho_0(r) + \delta\rho(r\,t)$

Consider functional $\rho[v](r\,t)$ defined by solution of interacting TDSE

Functional Taylor expansion of $\rho[v]$ around v_o:

$$\rho[v](r\,t) = \rho[v_0 + v_1\,](r\,t)$$

$$= \rho[v_0\,](r\,t) \longrightarrow \rho_o(r)$$

$$+ \int \left.\frac{\delta\rho[v](r\,t)}{\delta v(r'\,t')}\right|_{v_0} v_1(r'\,t')\,d^3r'\,dt' \longrightarrow \rho_1(r\,t)$$

$$+ \frac{1}{2}\int\int \left.\frac{\delta^2\rho[v](r\,t)}{\delta v(r'\,t')\delta v(r''\,t'')}\right|_{v_0} v_1(r',t')v_1(r'',t'')\,d^3r'\,d^3r''\,dt'\,dt'' \longrightarrow \rho_2(r\,t)$$

$$\vdots$$

$\rho_1(r,t)$ = linear density response of interacting system

$$\chi(r\,t, r'\,t') := \left.\frac{\delta\rho[v](r\,t)}{\delta v(r'\,t')}\right|_{v_0}$$ = density-density response function of interacting system

Lehmann representation of the full response function

$$\chi(r,r';\omega) = \lim_{\eta\to 0^+}\sum_m\left(\frac{\langle 0|\hat\rho(r)|m\rangle\langle m|\hat\rho(r')|0\rangle}{\omega-(E_m-E_0)+i\eta} - \frac{\langle 0|\hat\rho(r')|m\rangle\langle m|\hat\rho(r)|0\rangle}{\omega+(E_m-E_0)+i\eta}\right)$$

with the exact many-body eigenfunctions and energies of the initial unperturbed interacting system Hamiltnian $H(t_0)|m\rangle = E_m|m\rangle$

$\Rightarrow$ The exact linear density response

$$\rho_1(\omega) = \chi(\omega)\, v_1$$

has poles at the exact excitation energies $\Omega = E_m - E_0$

Analogous function $\rho_s[v_s](r\,t)$ for <u>non</u>-interacting system

$$\rho_S[v_S](r\,t) = \rho_S[v_{S,0} + v_{S,1}](r\,t) = \rho_S[v_{S,0}](r\,t) + \int \left.\frac{\delta\rho_S[v_S](r\,t)}{\delta v_S(r'\,t')}\right|_{v_{S,0}} v_{S,1}(r'\,t')\, d^3r'\, dt' + \cdots$$

$$\chi_S(r\,t, r'\,t') := \left.\frac{\delta\rho_S[v_S](r\,t)}{\delta v_S(r'\,t')}\right|_{v_{S,0}} = \textbf{density-density response function of } \underline{\textbf{non}}\textbf{-interacting system}$$

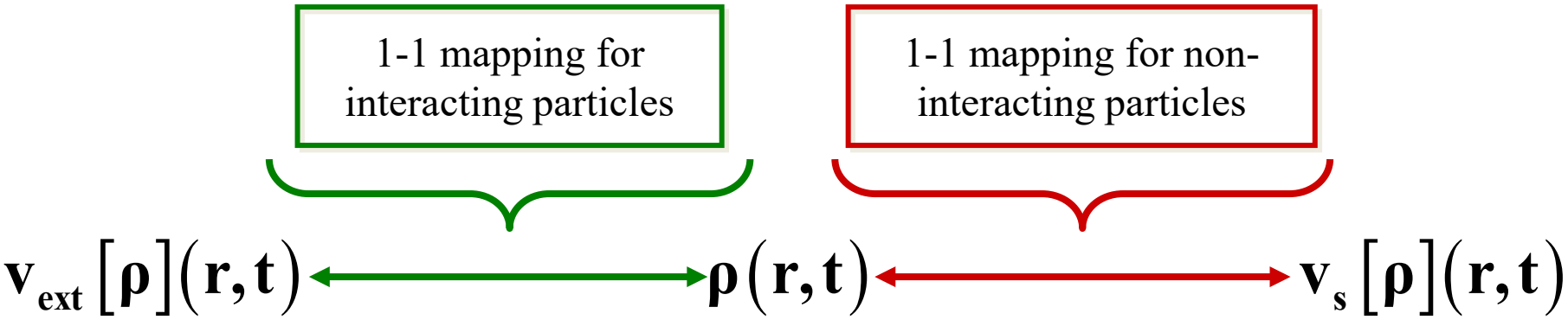
χ_S (r,r',ω) has also poles as function of ω, but at the non-interacting single-particle (KS) excitation energies.

GOAL: Find a way to calculate $\rho_1(r\,t)$ without explicitly evaluating $\chi(r\,t,r't')$ of the interacting system

starting point: Definition of xc potential

$$v_{xc}[\rho](r\,t) := v_S[\rho](r\,t) - v_{ext}[\rho](r\,t) - v_H[\rho](r\,t)$$

v_{xc} is well-defined through the non-interacting and the interacting 1-1 mapping.



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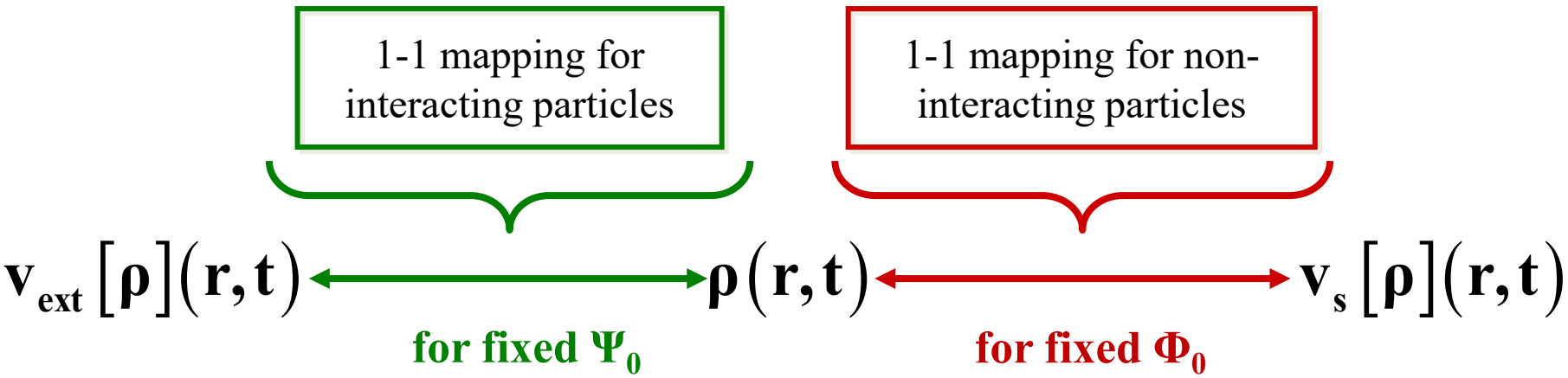

$$v_{xc}(r\,t) := v_{xc}[\rho, \Psi_0, \Phi_0](r\,t)$$

$$\left.\frac{\delta v_{xc}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} = \left.\frac{\delta v_{S}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \left.\frac{\delta v_{ext}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \frac{\delta(t-t')}{|r-r'|}$$

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$$\uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow$$

$$f_{xc}(r\,t, r'\,t') \qquad \chi_S^{-1}(r\,t, r'\,t') \qquad \chi^{-1}(r\,t, r'\,t') \qquad W_C(r\,t, r'\,t')$$

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$$\uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow \qquad\qquad \uparrow$$

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$$f_{xc} + W_C = \chi_S^{-1} - \chi^{-1}$$

$$\left.\frac{\delta v_{xc}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} = \left.\frac{\delta v_S[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \left.\frac{\delta v_{ext}[\rho](r\,t)}{\delta\rho(r'\,t')}\right|_{\rho_0} - \frac{\delta(t-t')}{|r-r'|}$$

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$$\chi_S\left(f_{xc} + W_C\right)\chi = \chi - \chi_S$$

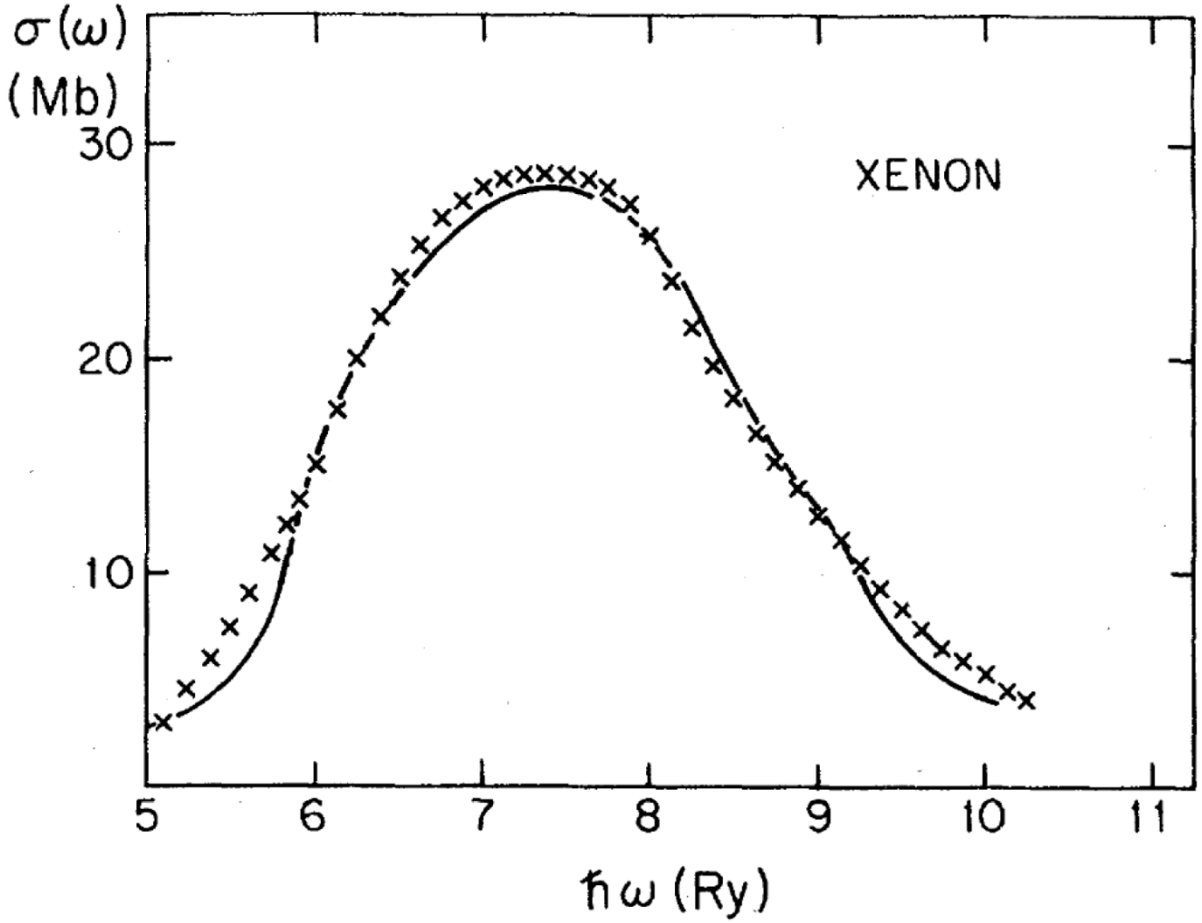
$$\chi = \chi_S + \chi_S \left(W_{ee} + f_{xc} \right) \chi$$

Act with this operator equation on arbitrary $v_1(r\,t)$ and use $\chi\, v_1 = \rho_1$:

$$\rho_1(r\,t) = \int d^3r'dt' \chi_S(r\,t, r't') \left[v_1(r\,t) + \int d^3r''dt'' \left\{ W_{ee}(r't', r''t'') + f_{xc}(r't', r''t'') \right\} \rho_1(r''t'') \right]$$

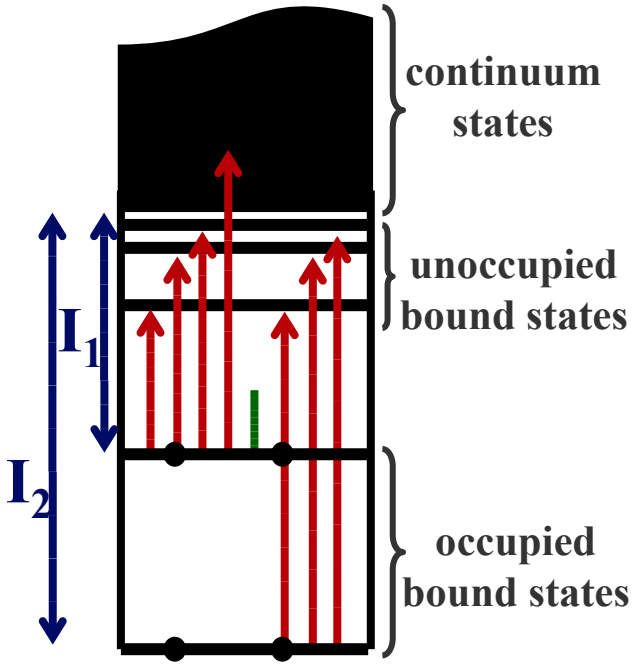
- Exact integral equation for $\rho_1(r\,t)$, to be solved iteratively
- Need approximation for $f_{xc}(r't', r''t'') = \left. \frac{\delta v_{xc}[\rho](r't')}{\delta \rho(r''t'')} \right|_{\rho_0}$
  (either for f_{xc} directly or for v_{xc})

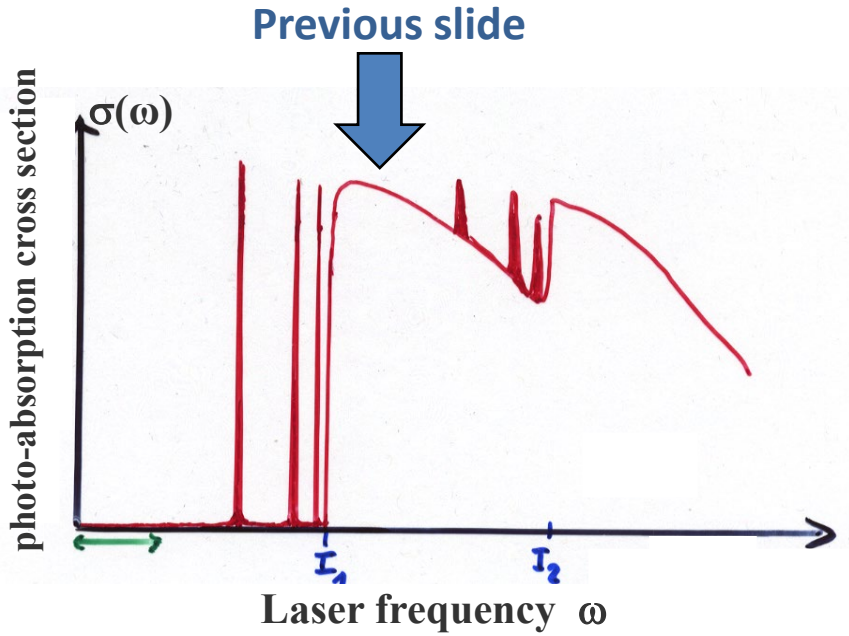
Total photoabsorption cross section of the Xe atom versus photon energy in the vicinity of the 4d threshold.



Solid line: self-consistent time-dependent KS calculation [A. Zangwill and P. Soven, Phys. Rev. A 21, 1561 (1980)]; crosses: experimental data [R. Haensel, G. Keitel, P. Schreiber, and C. Kunz, Phys. Rev. 188, 1375 (1969)].

Photo-absorption in weak lasers





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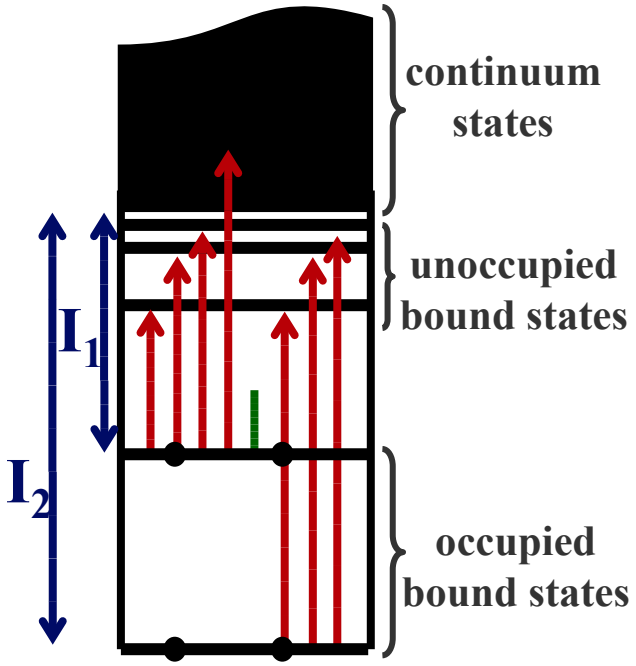

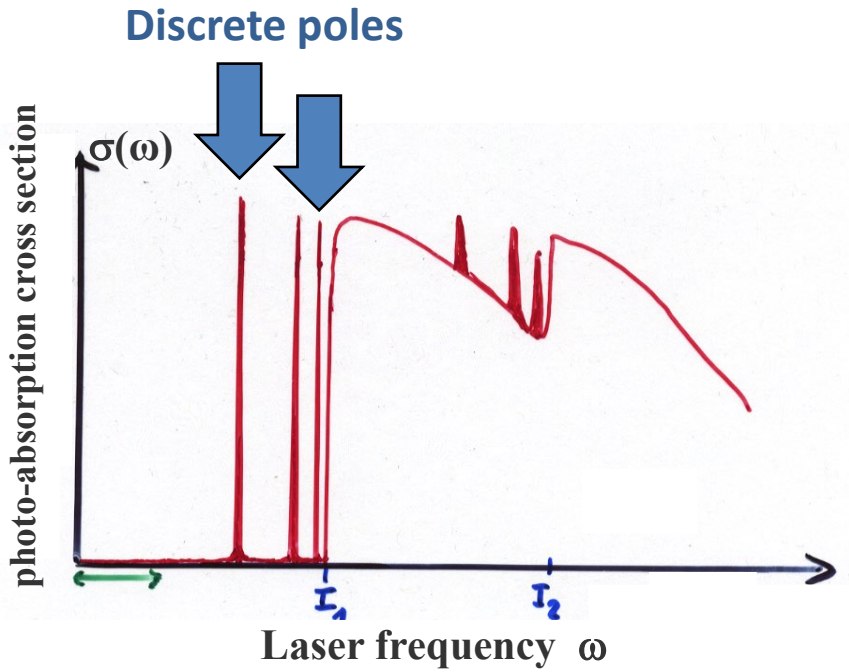

Looking at those frequencies, Ω, for which $\rho_1(\omega)$ has poles, leads to a (non-linear) eigenvalue equation

M. Petersilka, U. J. Gossmann, E.K.U.G., PRL 76, 1212 (1996)
T. Grabo, M. Petersilka, EKUG, J. Mol. Struc. (Theochem) 501, 353 (2000)
M.E. Casida, Recent Advances in Density Functional Methods I, 155 (1996)

$$\sum_{q'}\left(A_{qq'}(\Omega)+\omega_q\delta_{qq'}\right)\beta_{q'}=\Omega\beta_q$$

where

$$A_{qq'}=\alpha_{q'}\int d^3r\int d^3r'\,\Phi_q(r)\left(\frac{1}{|r-r'|}+f_{xc}(r,r',\Omega)\right)\Phi_{q'}(r')$$

$q=(j,a)$ double index $\qquad \alpha_q=f_a-f_j$

$\Phi_q(r)=\varphi_a^*(r)\varphi_j(r) \qquad \omega_q=\varepsilon_a-\varepsilon_j$

Atom	Experimental Excitation Energies $^1S \rightarrow {}^1P$ (in Ry)	KS energy differences $\Delta\epsilon_{KS}$ (Ry)	TDDFT
Be	0.388	0.259	0.391
Mg	0.319	0.234	0.327
Ca	0.216	0.157	0.234
Zn	0.426	0.315	0.423
Sr	0.198	0.141	0.210
Cd	0.398	0.269	0.391

from: M. Petersilka, U. J. Gossmann, E.K.U.G., PRL 76, 1212 (1996)

Excitation energies of CO molecule [mH]

State	$\Omega_{\text{expt KS}}$	KS-transition	$\Delta\epsilon_{KS}$	TDDFT
A $^1\Pi$	312.7	$5\sigma \rightarrow 2\pi$	252.3	310.2
a $^3\Pi$	232.3			221.4
I $^1\Sigma^-$	363.1	$1\pi \rightarrow 2\pi$	362.6	362.6
e $^3\Sigma^-$	363.1			362.6
a' $^3\Sigma^+$	312.7			314.9
D $^1\Delta$	375.9			380.7
d $^3\Delta$	344.0			339.6

Molecular excitation energies from time-dependent density-functional theory
T Grabo, M Petersilka, EKU Gross, J Mol Struc-Theochem 501, 353 (2000).

Failures of ALDA in the linear response regime

- **H_2 dissociation is incorrect:**

$$E\left(^1\Sigma_u^+\right) - E\left(^1\Sigma_g^+\right) \xrightarrow[R\to\infty]{} 0 \quad \textbf{(in ALDA)}$$

(see: Gritsenko, van Gisbergen, Görling, Baerends, J. Chem. Phys. 113, 8478 (2000))

- **response of long chains strongly overestimated**

(see: Champagne et al., J. Chem. Phys. 109, 10489 (1998) and 110, 11664 (1999))

- **in periodic solids,** $f_{xc}^{ALDA}(q,\omega,\rho) = c(\rho)$ **whereas,**

for insulators, $f_{xc}^{exact} \xrightarrow[q\to 0]{} 1/q^2$ **divergent.**

- **charge-transfer excitations not properly described**

(see: Dreuw et al., J. Chem. Phys. 119, 2943 (2003))

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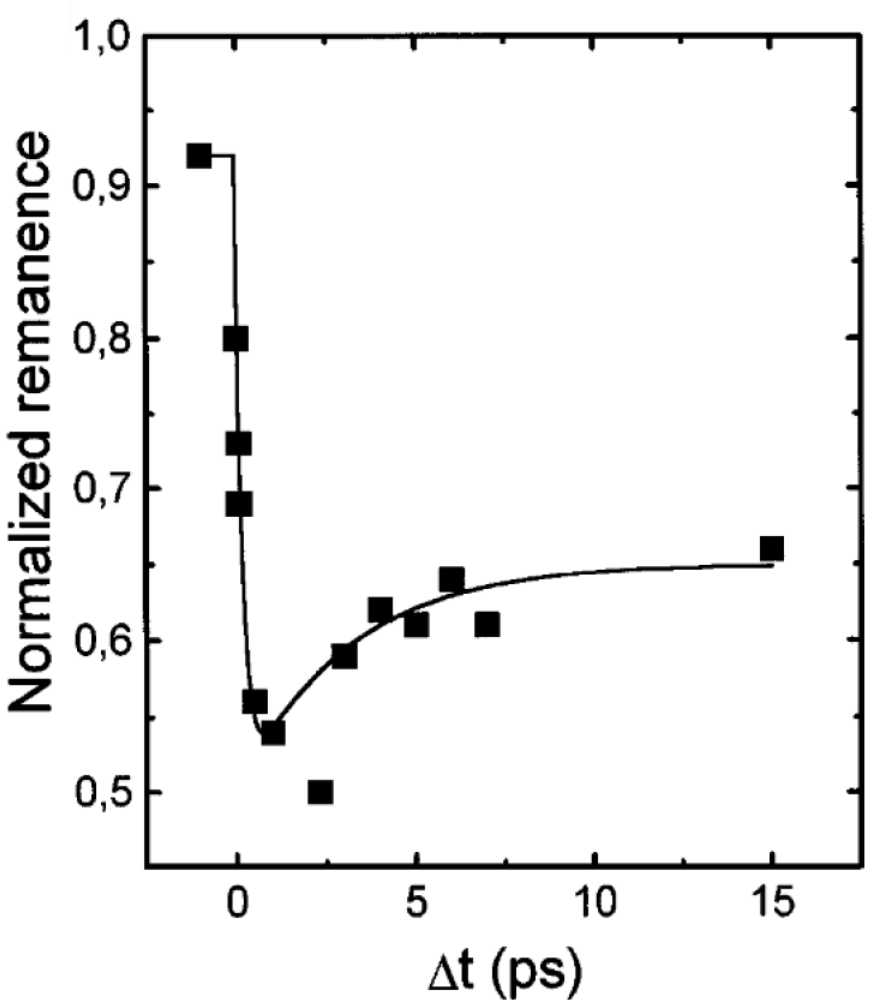
These difficulties have largely been solved by xc functionals more advanced than ALDA

Beyond the linear regime: Real-time TDDFT prediction of electron dynamics far from equilibrium

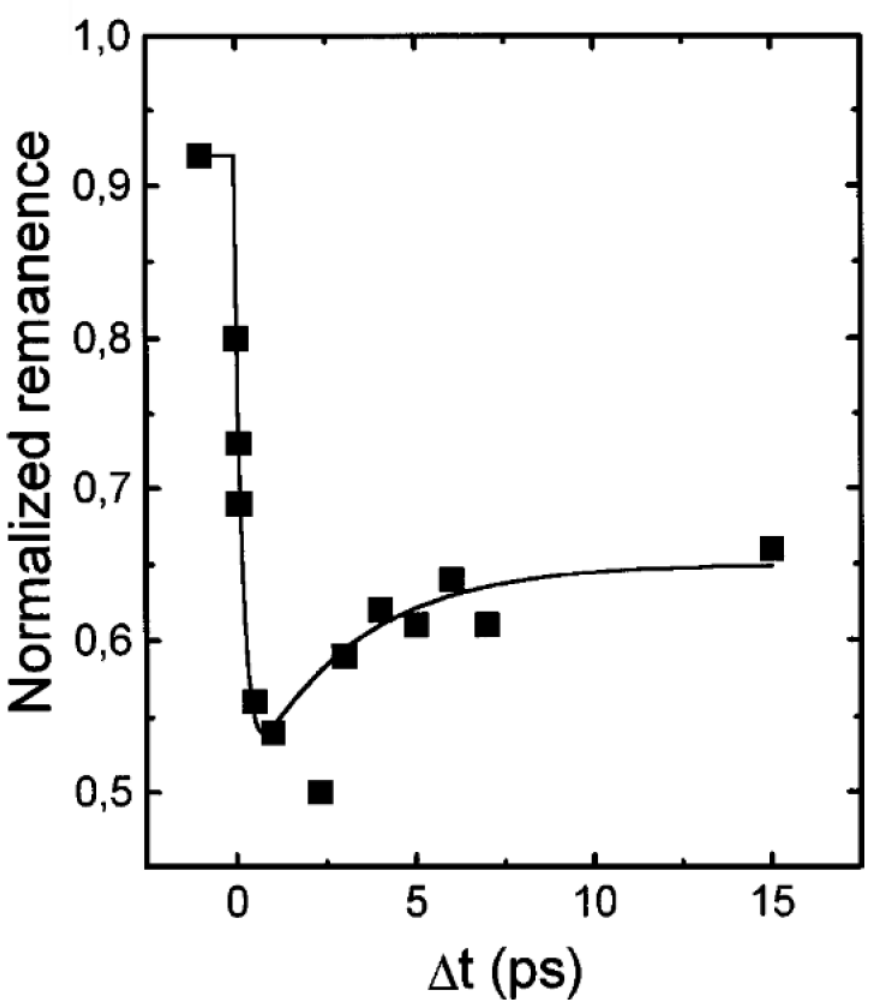
Ultrafast laser-driven spin dynamics in solids

First experiment on ultrafast laser induced demagnetization



Beaurepaire et al, PRL 76, 4250 (1996)

First experiment on ultrafast laser induced demagnetization



Beaurepaire et al, PRL 76, 4250 (1996)

Demagnetization in less than 100 fs has been demonstrated experimentally

Non-collinear-Spin TDDFT with Spin-Orbit-Coupling (weakly relativistic limit of relativistic TDDFT)

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

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

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

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

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

Universal functionals of ρ and m

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

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

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

Aspects of the numerical implementation

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

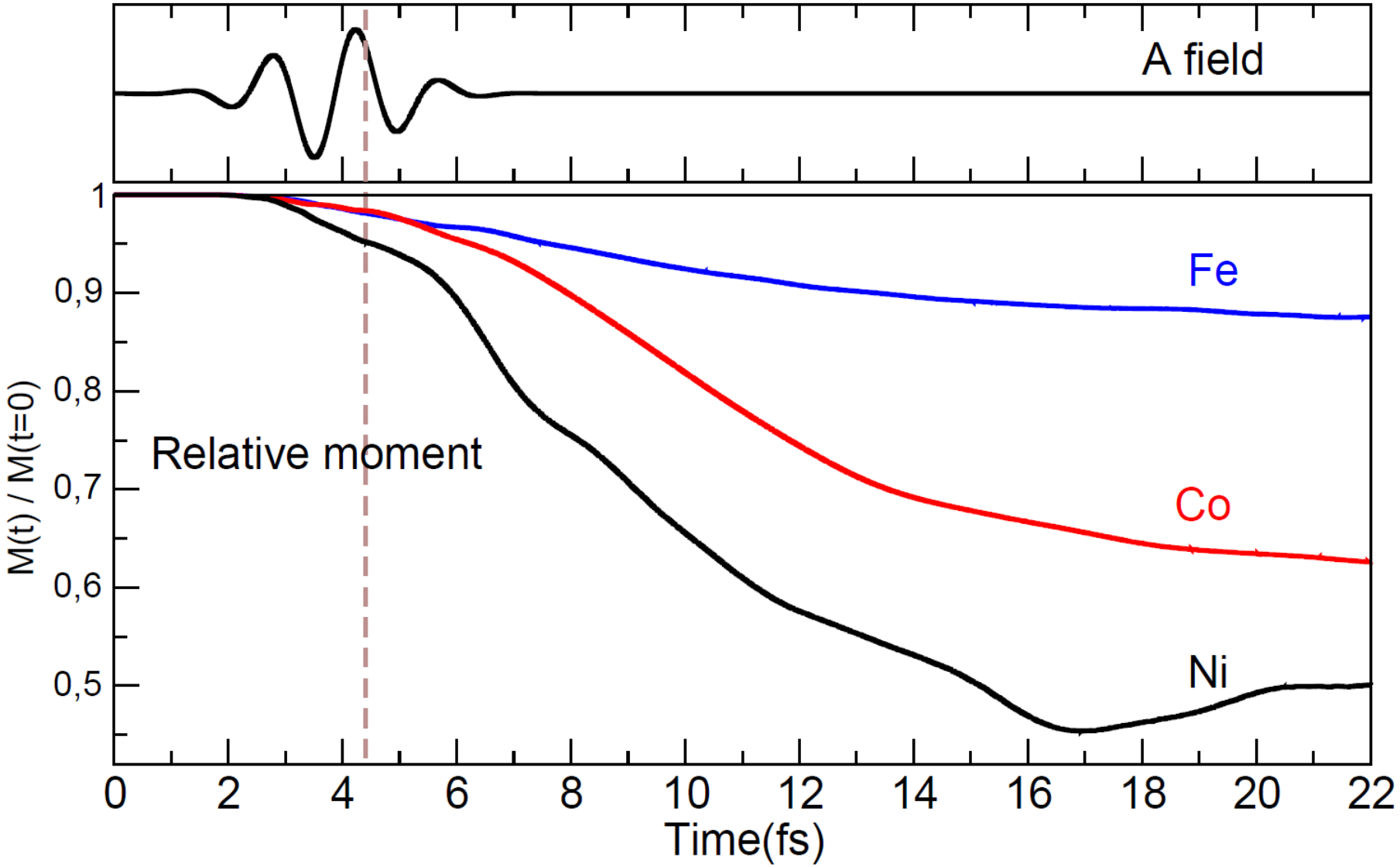
  ⇨ Dipole approximation is made
  (i.e. electric field of laser is assumed to be spatially constant)

  ⇨ 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/)**

Demagnetization in Fe, Co, and Ni

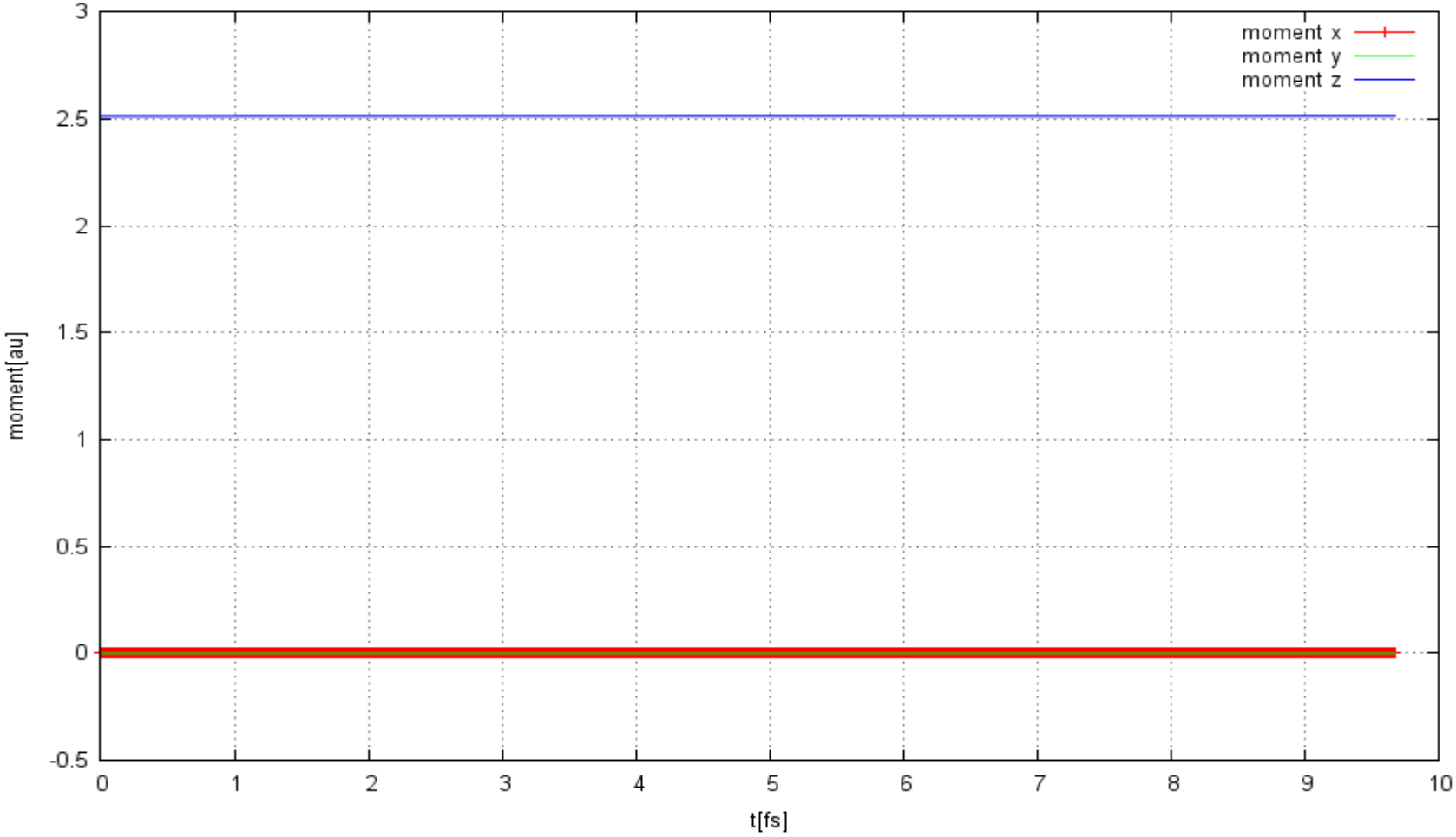


K. Krieger, K. Dewhurst, P. Elliott, S. Sharma, E.K.U.G., JCTC 11, 4870 (2015)

Analysis of the results

Calculation without spin-orbit coupling

components of spin moment



Exact equation of motion

$$\frac{\partial}{\partial t} m_z(r,t) = \frac{i}{\hbar}\left\langle \Phi_{KS}^{det}(t) \left| \left[\hat{H}_{KS}, \hat{m}_z(r) \right] \right| \Phi_{KS}^{det}(t) \right\rangle$$

$$= \left\{ m_x(r,t) B_{xc,y}(rt) - m_y(r,t) B_{xc,x}(rt) \right\}$$

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

$$- \nabla \cdot j_z(r,t)$$

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

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local spin torque $(m \times B_{xc})_z$

$$= \left\{ m_x(r,t) B_{xc,y}(rt) - m_y(r,t) B_{xc,x}(rt) \right\}$$

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SOC

$$-\nabla \cdot j_z(r,t)$$

"continuity"

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spin current tensor

Dynamics of **total** moment: Integrate over the unit cell

Exact equation of motion for total moment

$$\frac{\partial}{\partial t} M_z(t) = \frac{i}{\hbar} \int d^3 r \left\langle \Phi_{KS}^{det}(t) \left| \left[\hat{H}_{KS}, \hat{m}_z(r) \right] \right| \Phi_{KS}^{det}(t) \right\rangle$$

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

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

$$- \int d^3 r \left\{ \nabla \cdot j_z(r,t) \right\}$$ **=0**

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

Exact equation of motion for total moment

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Global torque exerted by $B_{xc} = 0$ (zero torque theorem)

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

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

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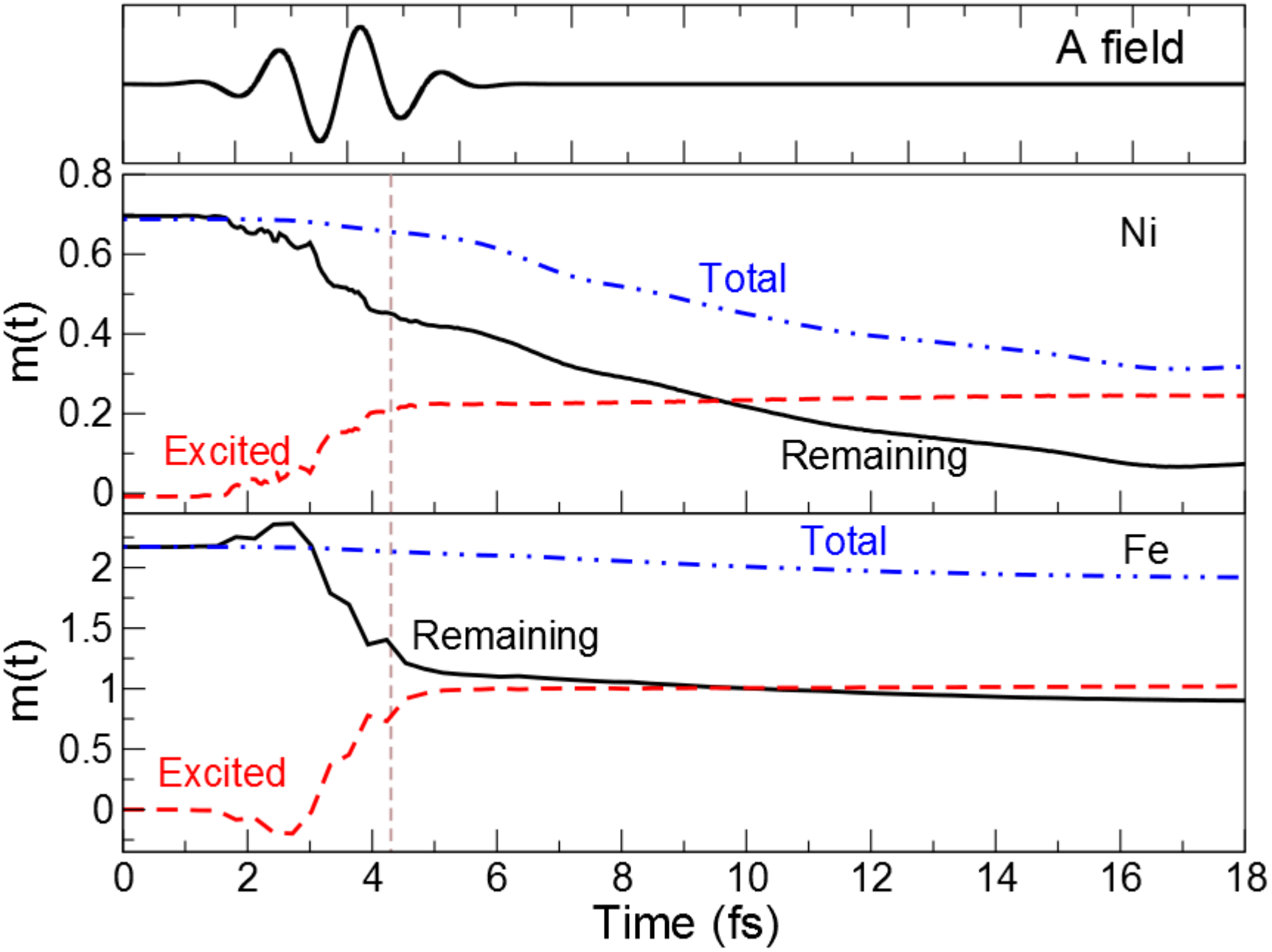
$$\overleftrightarrow{j}(r,t) = \left\langle \hat{\sigma} \otimes \hat{j}(r) \right\rangle$$ spin current tensor

SOC is the only term which can change the total moment!


A field
Ni
Total
Excited
Remaining
m(t)
Fe
Total
Remaining
Excited
m(t)
Time (fs)

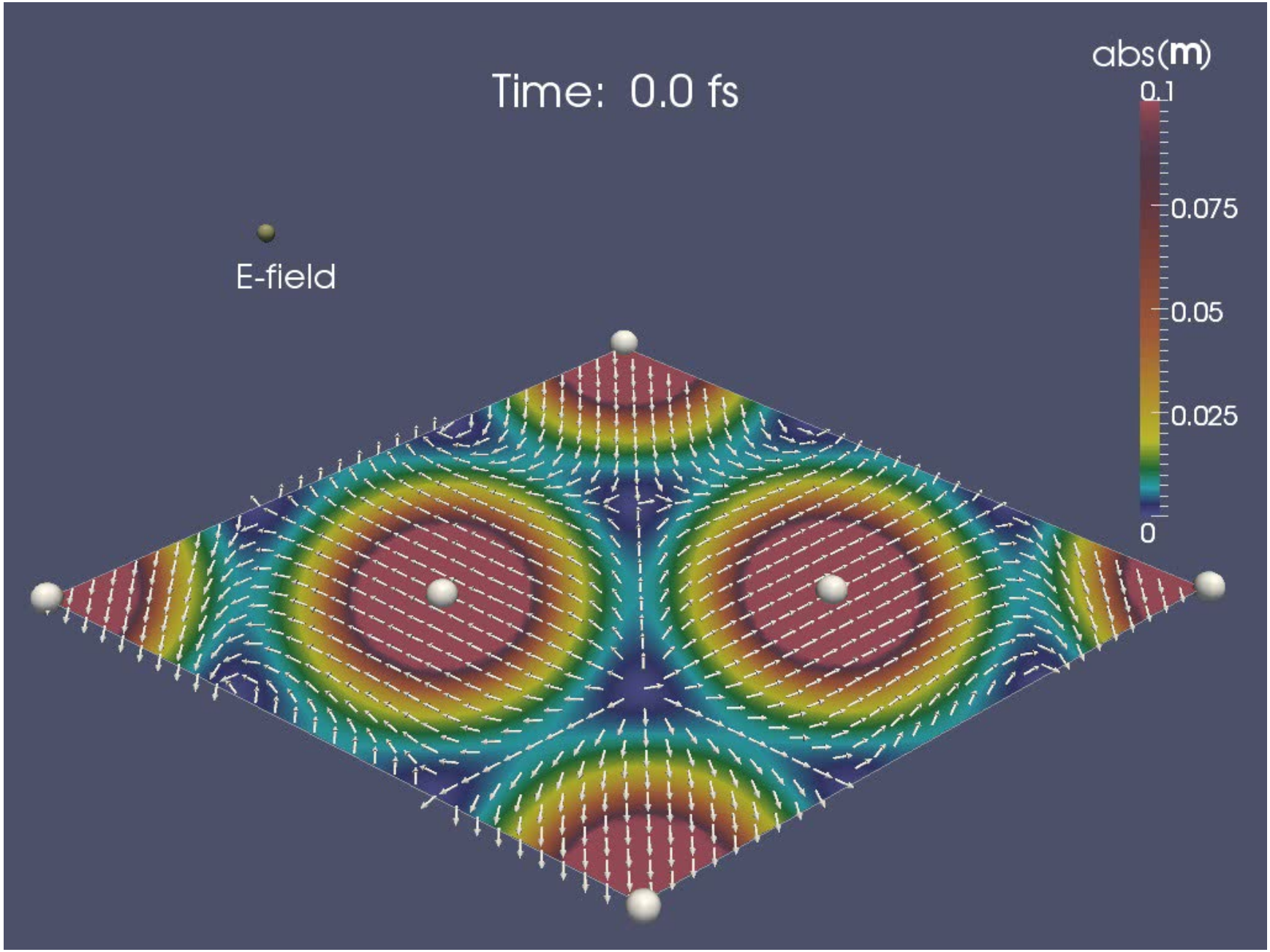
Demagnetization occurs in two steps:

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

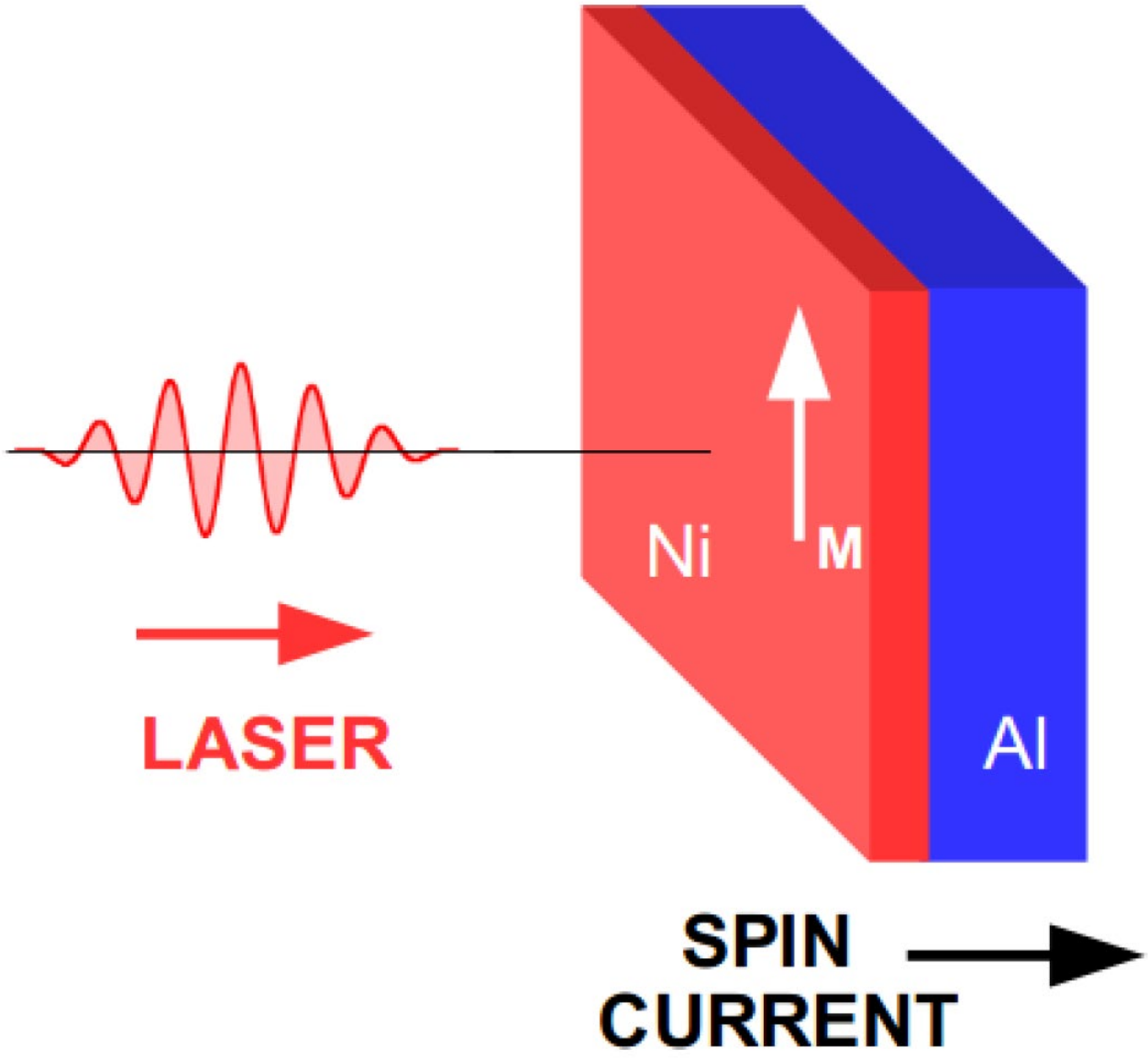
- Spin-Orbit term drives demagnetization of the more localized electrons until stabilization at lower moment is achieved

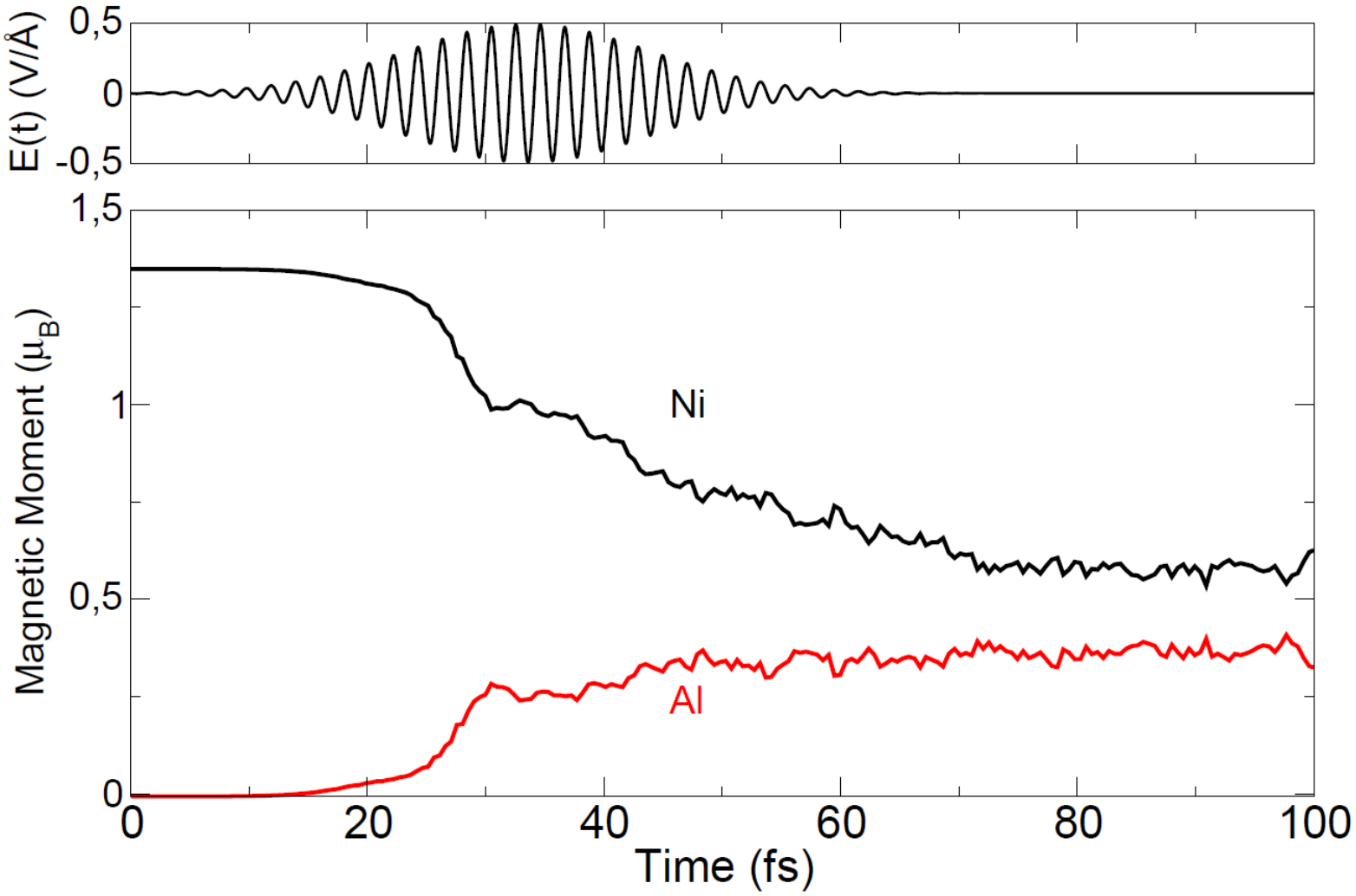
- This is a local mechanism, hence occurs in this form in essentially all systems, e.g. magnetic clusters (Sanvito et al) or magnetic mono-layer / few-layer systems

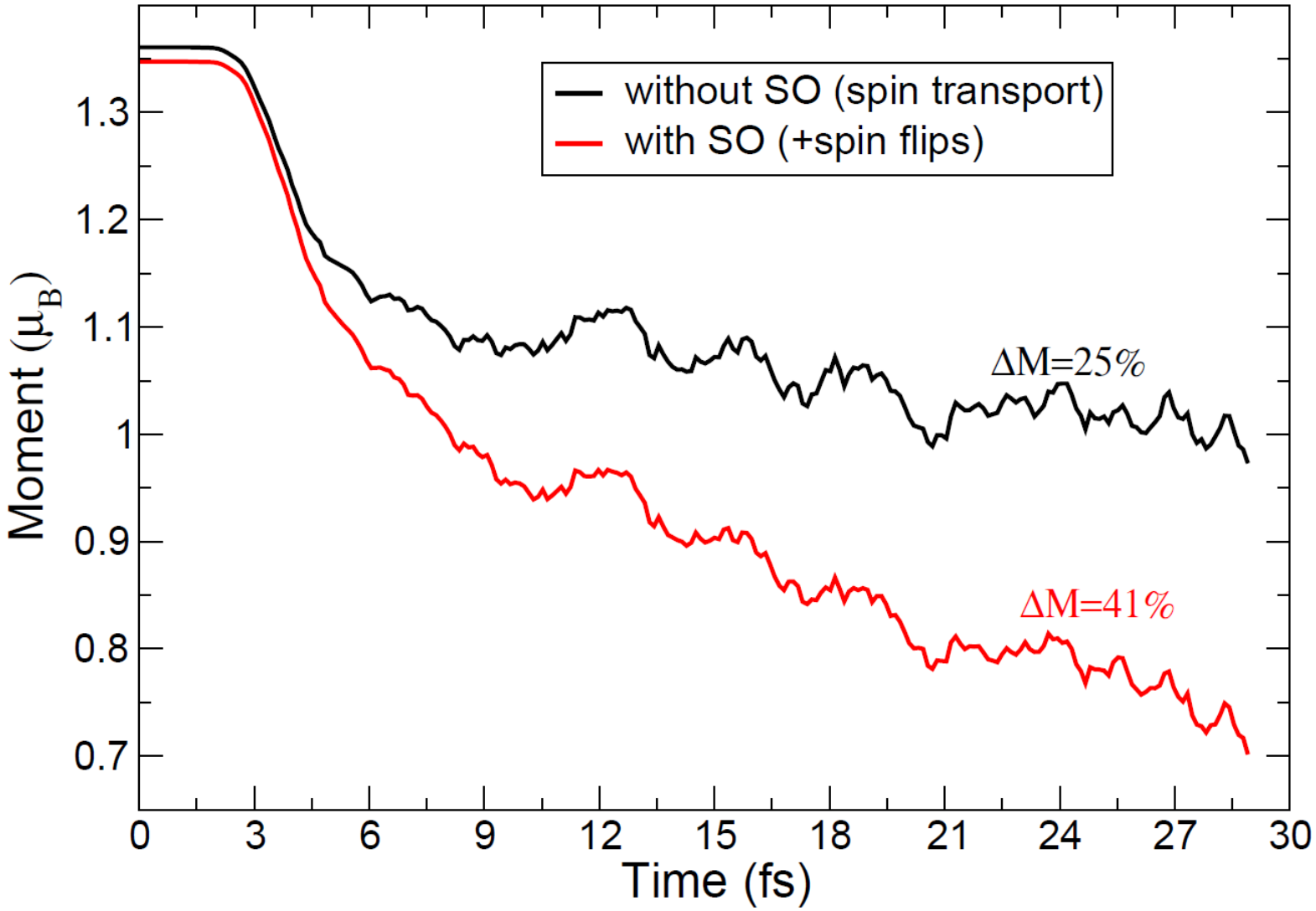
Beyond 3D bulk



Cr monolayer


Ni
M
Al
LASER
SPIN
CURRENT


E(t) (V/Å)
0,5
0
-0,5
1,5
1
0,5
0
Magnetic Moment (μ_B)
Ni
Al
0
20
40
60
80
100
Time (fs)


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

Experimental confirmation:

***Spin Flips versus Spin Transport in Nonthermal Electrons Excited by Ultrashort Optical Pulses in Transition Metals*,**
V. Shokeen, M. Sanchez Piaia, J.-Y. Bigot, T. Mueller, P. Elliott, J.K. Dewhurst, S. Sharma, E. K. U. Gross, Phys. Rev. Lett. 119, 107203 (2017).

Review article:

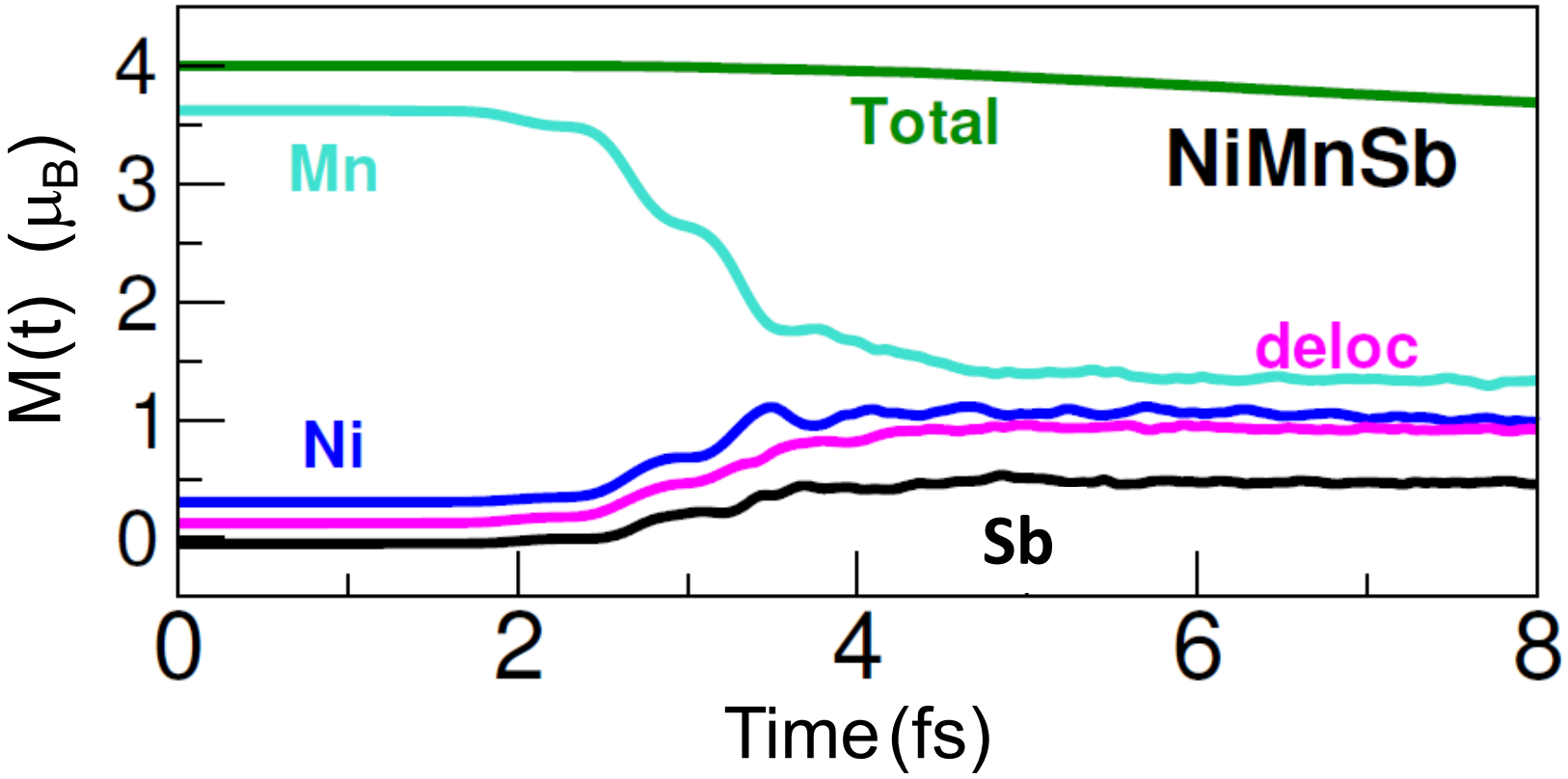
***Time-Dependent Density Functional Theory for Spin Dynamics*,**
P. Elliott, M. Stamenova, J. Simoni, S. Sharma, S. Sanvito, and E.K.U. Gross, in: Handbook of Materials Modeling, W. Andreoni, S. Yip eds, Springer (2020), p. 841

Optical intersublattice spin transfer (OISTR)

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

K. Dewhurst, P. Elliott, S. Shallcross, E.K.U. Gross, S. Sharma,
Nano Lett. 18, 1842 (2018)

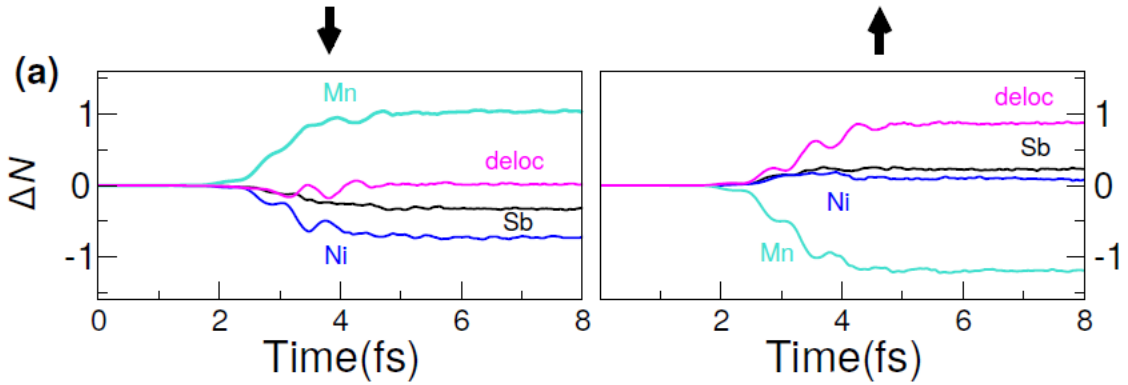
OISTR was first predicted with TDDFT and later found experimentally (Aeschlimann group, Kaiserslautern, 2018)

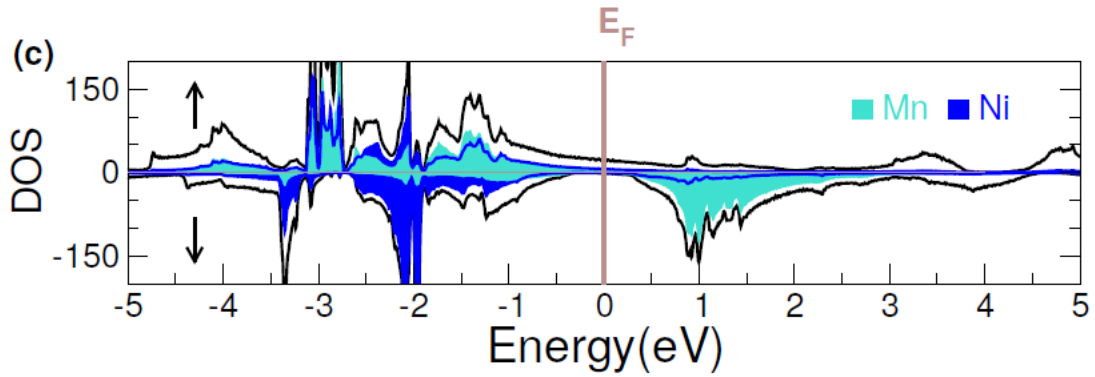


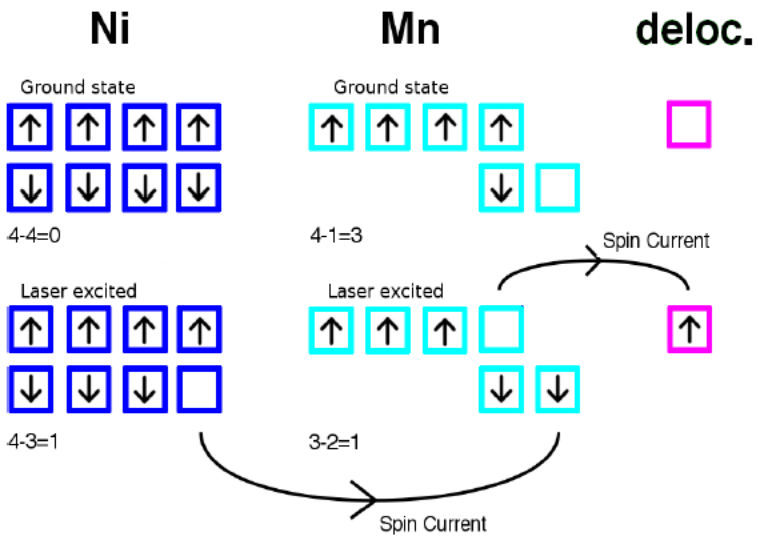
Global moment |M(t)| nearly preserved. Local moments change.

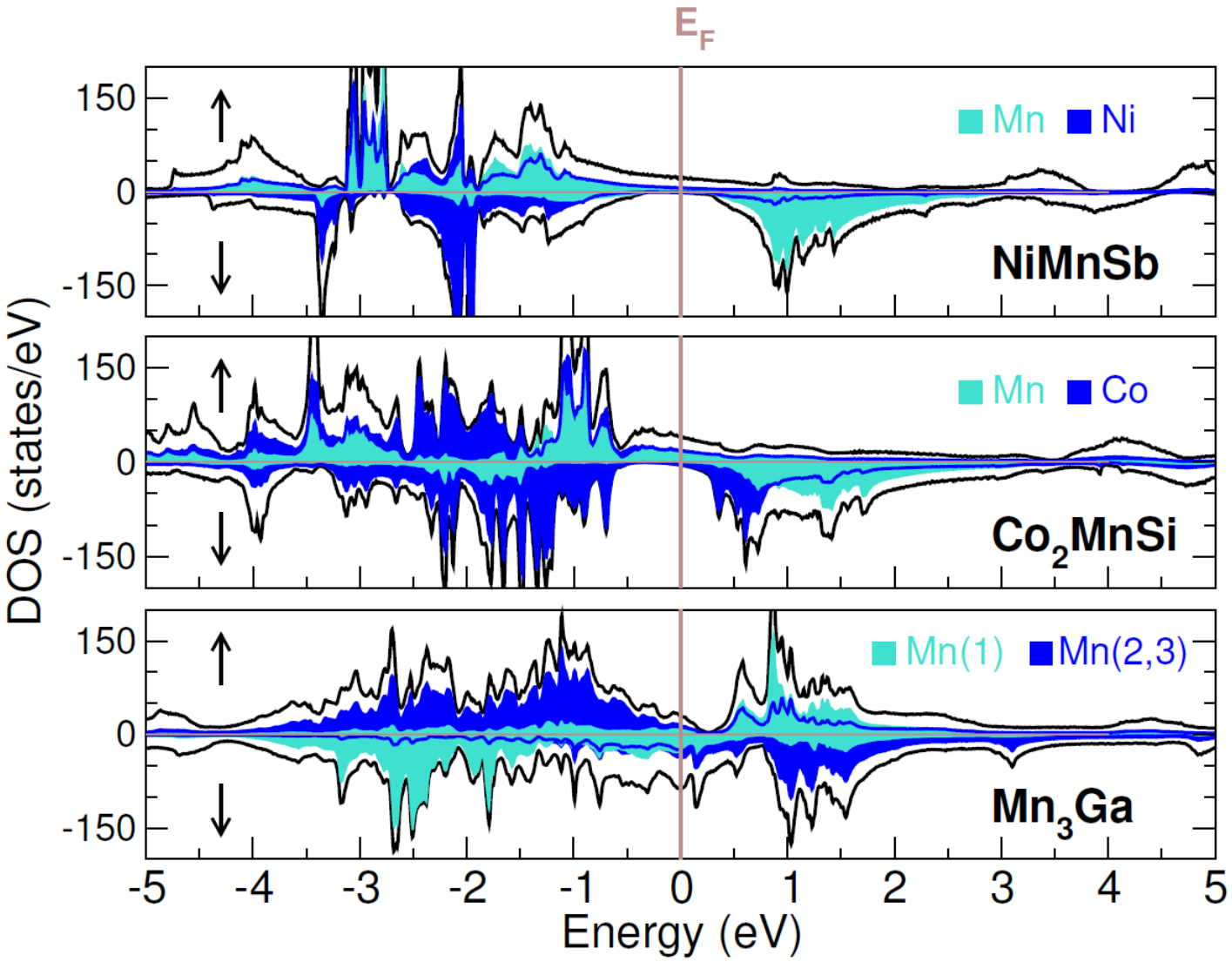
Laser parameters: ω= 2.72 eV, a FWHM of 2.42 fs, and fluence of 93.5 mJ/cm2, giving a peak intensity of 1 × 1014 W/cm2.

NiMnSb

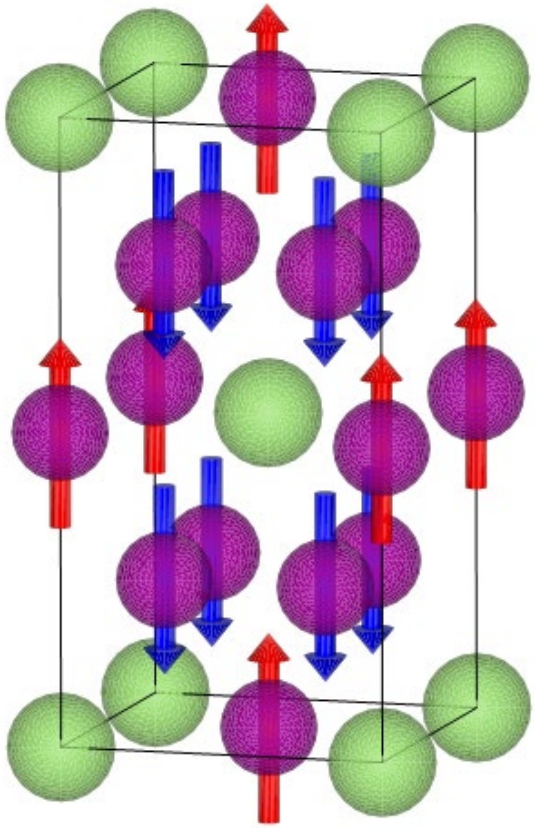

(a)
ΔN
Mn
deloc
Sb
Ni
Time(fs)
deloc
Sb
Ni
Mn
Time(fs)


(c)
E_F
DOS
Mn
Ni
Energy(eV)


Ni
Mn
deloc.
Ground state
4-4=0
Ground state
4-1=3
Spin Current
Laser excited
4-3=1
Laser excited
3-2=1
Spin Current


E_F
150
0
-150
Mn
Ni
NiMnSb
Co
Co2MnSi
Mn(1)
Mn(2,3)
Mn3Ga
DOS (states/eV)
-5
-4
-3
-2
-1
0
1
2
3
4
5
Energy (eV)

Mn_3Ga (ferri-magnet)



Ga

Mn

TDDFT prediction for Mn_3Ga: ferri → ferro transition within 4 fs

PHYSICS

Ultrafast optically induced spin transfer in ferromagnetic alloys

M. Hofherr[1,2], S. Häuser[1], J. K. Dewhurst[3], P. Tengdin[4], S. Sakshath[1], H. T. Nembach[4,5], S. T. Weber[1], J. M. Shaw[5], T. J. Silva[5], H. C. Kapteyn[4], M. Cinchetti[6], B. Rethfeld[1], M. M. Murnane[4], D. Steil[7], B. Stadtmüller[1,2], S. Sharma[8], M. Aeschlimann[1], S. Mathias[7]*

The vision of using light to manipulate electronic and spin excitations in materials on their fundamental time and length scales requires new approaches in experiment and theory to observe and understand these excitations. The ultimate speed limit for all-optical manipulation requires control schemes for which the electronic or magnetic subsystems of the materials are coherently manipulated on the time scale of the laser excitation pulse. In our work, we provide experimental evidence of such a direct, ultrafast, and coherent spin transfer between two magnetic subsystems of an alloy of Fe and Ni. Our experimental findings are fully supported by time-dependent density functional theory simulations and, hence, suggest the possibility of coherently controlling spin dynamics on subfemtosecond time scales, i.e., the birth of the research area of attomagnetism.

INTRODUCTION

Next-generation quantum materials will make it possible to surpass the speed and efficiency limits of current devices to generate faster, smaller, and more energy-efficient technological implementations (*1*–*8*). A promising approach to enhance data processing speed is to use ever shorter external stimuli for the manipulation and control of the state of matter. In this context, light represents the fastest means to alter the state of a material since laser pulses can now be generated with extremely short temporal duration down to a few tens of attoseconds. Visible lasers can deliver pulses with few-femtosecond durations that can be used to excite matter, while attosecond pulses can be generated in the extreme ultraviolet (EUV) and soft x-ray regions to probe the resulting dynamics (*9*–*12*). When combined with advanced spectroscopies, these new light sources are enabling comprehensive views into material results in a nonequilibrium hot charge distribution, which subsequently triggers a series of cascaded incoherent secondary processes including transport, (spin-flip) scattering, and quasiparticle generation, ultimately leading to macroscopic demagnetization of the magnetic material within <500 fs (*18*–*22*).

The fastest manipulation of the magnetic state should occur, however, through the direct (possibly coherent) interaction between the spin system of the material and the light field itself (*23*–*25*). While the first experiments have provided indications that such a direct manipulation scheme might be possible (*26*–*29*), to our knowledge, only one experimental study on magnetic metallic systems to date has focused on this challenging aspect of coherent ultrafast magnetism induced by femtosecond laser pulses (*24*). One particularly interesting and previously unknown scheme for the ultrafast

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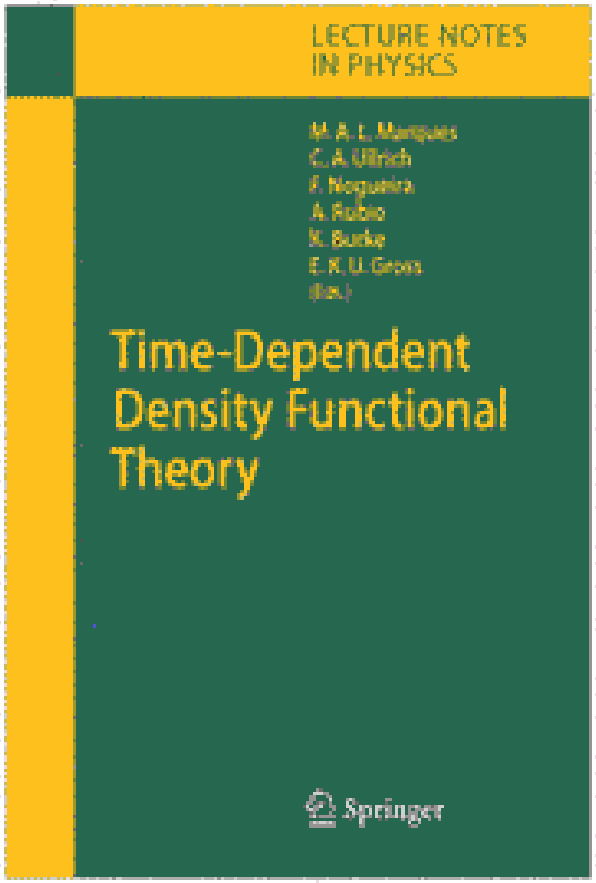
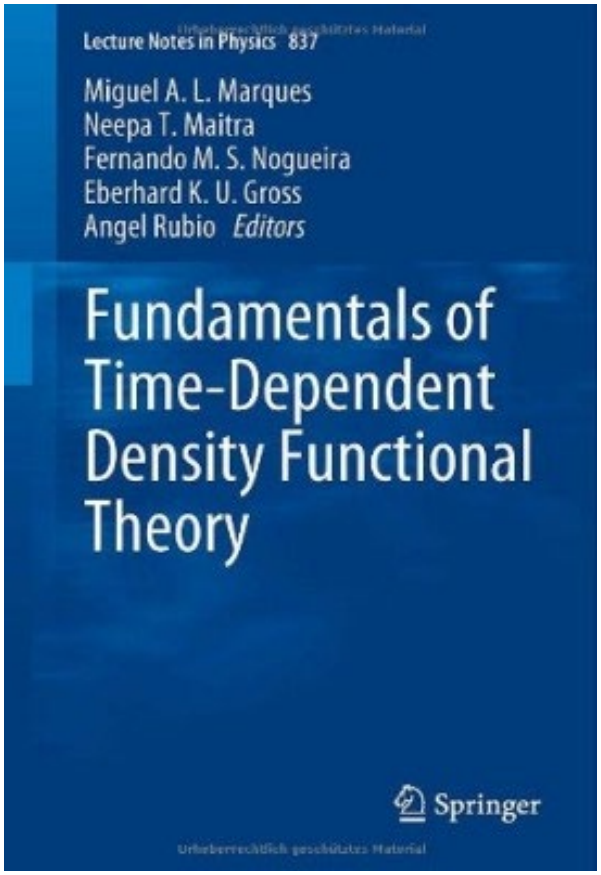


Lecture Notes in Physics 706 (Springer, 2006)



Lecture Notes in Physics 837 (Springer, 2012)