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)
Possible mechanisms for demagnetisation

• 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)

• Our proposal for the first 50 fs:
  Laser-induced charge excitation followed by spin-orbit-driven
demagnetization of the remaining d-electrons
Basic 1-1 correspondence:
\[ v(\text{rt}) \leftrightarrow^{1-1} \rho(\text{rt}) \]
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
\[ \rho(\text{rt}) = \sum_{j=1}^{N} \left| \phi_j(\text{rt}) \right|^2 \]
of an auxiliary non-interacting (KS) system
\[ \text{i} \hbar \frac{\partial}{\partial t} \phi_j(\text{rt}) = \left( -\frac{\hbar^2 \nabla^2}{2m} + v_s[\rho](\text{rt}) \right) \phi_j(\text{rt}) \]
with the local potential
\[ v_s[\rho(r't')](\text{rt}) = v(\text{rt}) + \int d^3r' \frac{\rho(r't')}{|r-r'|} + v_{xc}[\rho(r't')](\text{rt}) \]
Basic 1-1 correspondence:

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Generalization: Non-collinear-Spin-TDDFT with SOC

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

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

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

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

where \( \varphi_k(r,t) \) are Pauli spinors
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Universal functionals of \( \rho \) and \( m \)
Quantity of prime interest:
vector field of spin magnetization
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Cr monolayer in ground state
Demagnetisation in Fe, Co and Ni

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

Kay Dewhurst

Sangeeta Sharma
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  (very large compared to unit cell)

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Analysis of the results
Calculation without spin-orbit coupling

components of spin moment

momemt x
moment y
moment z

momemt[au] vs. t[fs]
Exact equation of motion

\[
\frac{\partial}{\partial t} M_z(t) = \frac{i}{\hbar} \left\langle \left[ \hat{H}_{KS}, \hat{\sigma}_z \right] \rightangle \\
= \int d^3r \left\{ M_x(r,t)B_{KS,y}(rt) - M_y(r,t)B_{KS,x}(rt) \right\} \\
+ \int d^3r \frac{1}{2c^2} \left\{ \hat{x} \cdot [\nabla v_s(r,t) \times j_y(r,t)] - \hat{y} \cdot [\nabla v_s(r,t) \times j_z(r,t)] \right\}
\]

\[
\cdot j(r,t) = \langle \hat{\sigma} \otimes \hat{p} \rangle \quad \text{spin current tensor}
\]

\[
B_{KS}(rt) = B_{ext}(rt) + B_{XC}(rt)
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Exact equation of motion

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B_{KS}(rt) = B_{\text{ext}}(rt) + B_{\text{XC}}(rt)
\]

**Global torque** = 0, if \( B_{\text{ext}} = 0 \)

(due to zero-torque theorem for \( B_{\text{xc}} \))
Note: Ground state of bulk Fe, Co, Ni is collinear
**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

Time: 0.0 fs

E-field
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).
Effect of spin transport across interfaces: Ni@Al
Heusler compounds
$\text{Ga} \quad 0.02 \ \mu\text{B}$

$\text{Mn} \quad -3.14 \ \mu\text{B}$

$\text{Ni} \quad -0.37 \ \mu\text{B}$
Ni$_2$MnGa

Laser parameters: $\omega=2.72\text{eV}$ $I_{\text{peak}}=1\times10^{15}\text{ W/cm}^2$ $J=935\text{ mJ/cm}^2$ $\text{FWHM}=2.42\text{ fs}$

See loss in global moment
Ni$_2$MnGa

Also change in local moments

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

Followed by spin-orbit mediated demagnetization on Ni
The graph shows the evolution of $M(t)$ (in $\mu_B$) with time (in fs) for different elements in NiMnSb. The total magnetic moment decreases over time, with contributions from Mn and Ni becoming more evident as time progresses. The graph also highlights the delocalization effect (deloc) as a factor in the observed behavior.

- **Mn** shows a sharp decrease in magnetic moment shortly after the initial time point, indicating a quick response to the changing conditions.
- **Ni** displays a gradual increase in magnetic moment over time, suggesting a delayed or more gradual reaction.
- The **Total** magnetic moment decreases consistently, with contributions from both Mn and Ni, and the delocalization effect (deloc) is noted as a possible influence.

The graph is a crucial visualization for understanding the magnetic dynamics and interplay between different elements in NiMnSb systems.
NiMnSb

(a) 

\[ \Delta N \] vs. Time (fs)

(b) 

\[ \Delta N \] vs. Time (fs)

(c) 

DOS vs. Energy (eV)

Ni
Mn
deloc.

Ground state
- 4-4=0
- 4-1=3

Laser excited
- 4-3=1
- 3-2=1

Spin Current

Spin Current
\( \text{Mn}_3\text{Ga} \)
Mn$_3$Ga

Laser parameters: $\omega=2.72$ eV $I_{\text{peak}}=1\times10^{15}$ W/cm$^2$ $J=935$ mJ/cm$^2$ $\text{FWHM}=2.42$ fs

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

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 or the basis of the ground-state DOS
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 $v_{xc}[\rho(r,t')]_{(rt)}$

• **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**