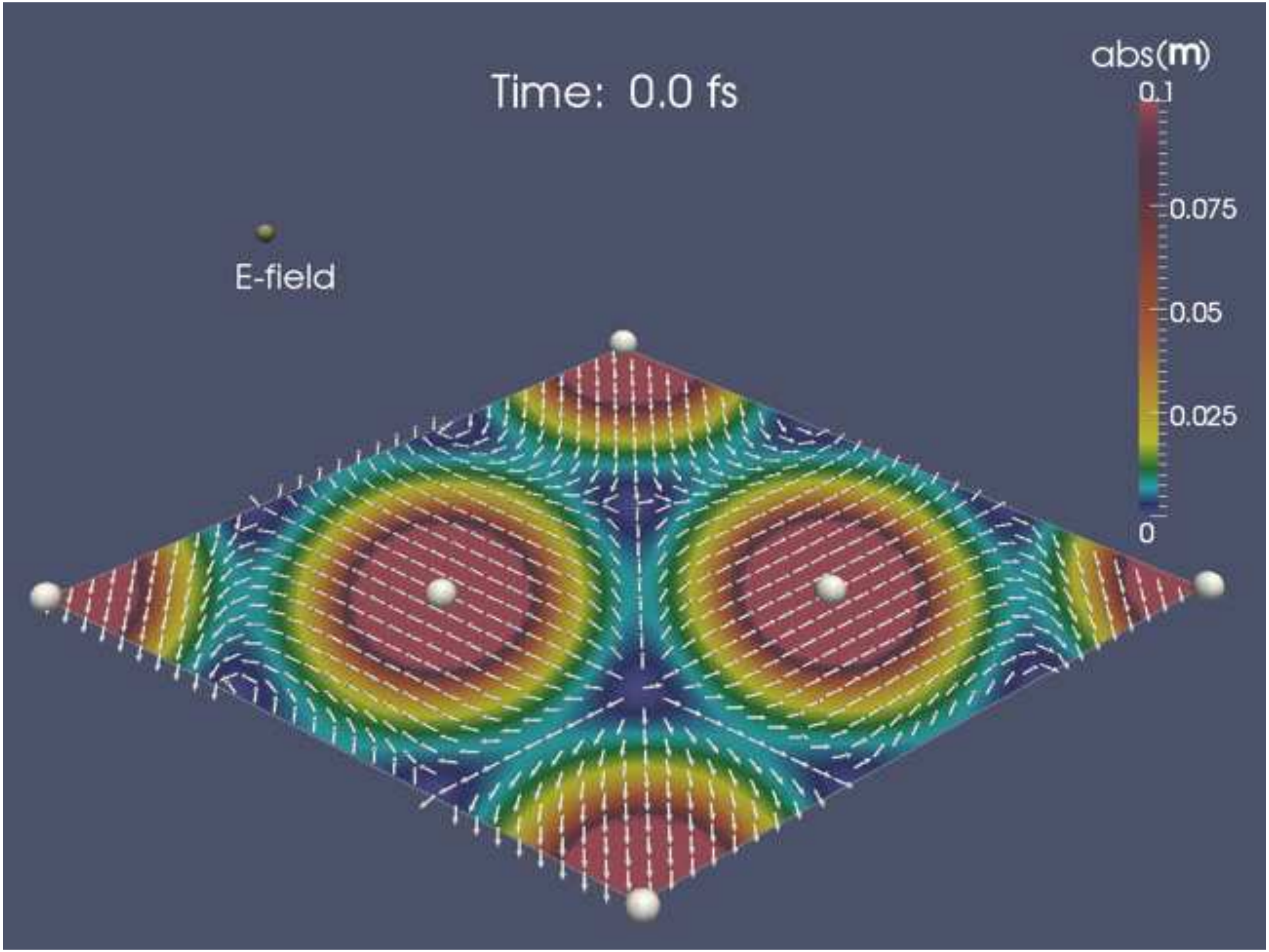
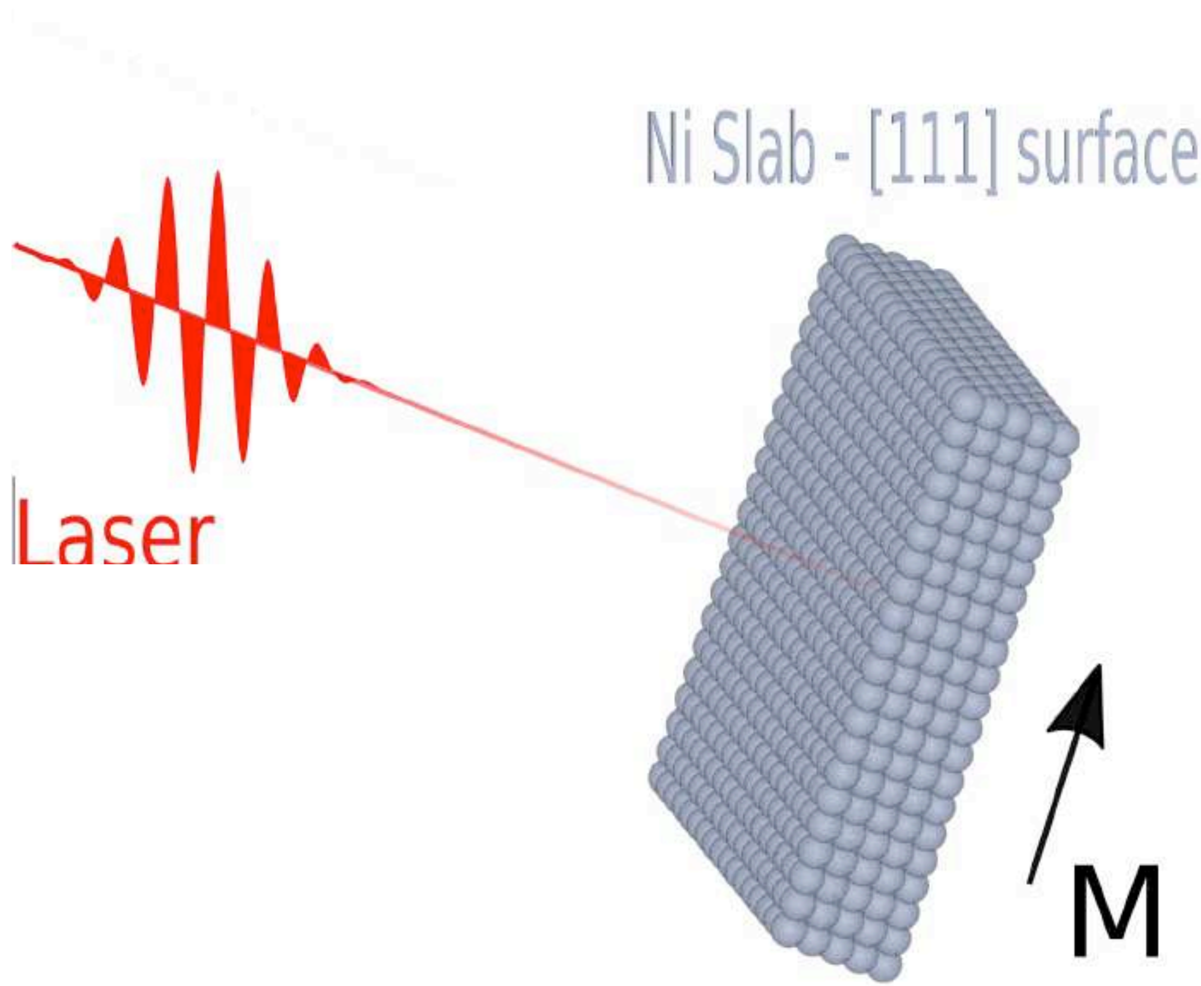
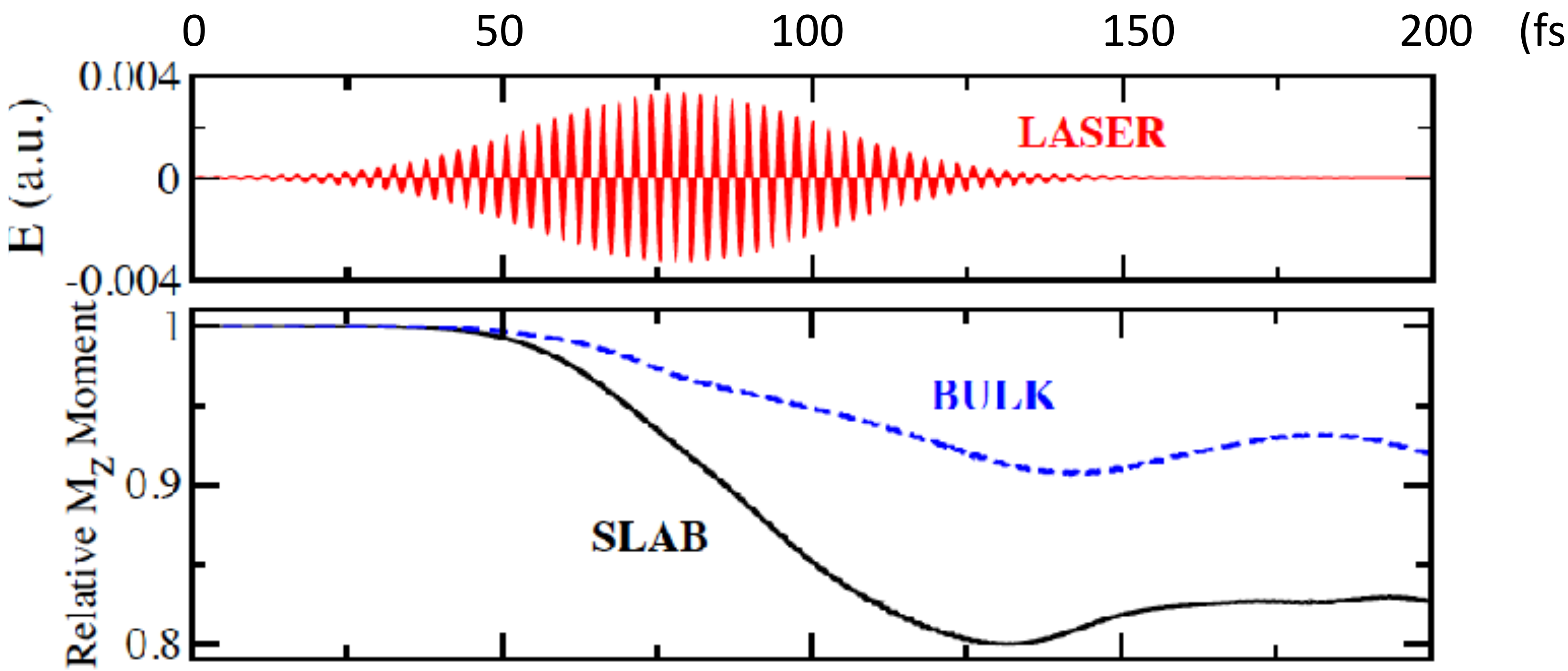
Beyond 3D bulk



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

Influence of the approximation for the xc functional

Ordinary LSDA yields GLOBAL collinearity

$$\vec{B}_{xc}(r) = \begin{pmatrix} 0 \\ 0 \\ B_{xc}(r) \end{pmatrix} \qquad \vec{m}(r) = \begin{pmatrix} 0 \\ 0 \\ m(r) \end{pmatrix}$$

$\vec{B}_{xc}, \vec{m}$ parallel to $\begin{pmatrix} 0 \\ 0 \\ 1 \end{pmatrix}$ everywhere in space

Construction of non-collinear LSDA

Kübler, Sandratskii (1980s)

$$\int \rho(r)v(r)d^3r - \int \vec{m}(r)\cdot\vec{B}(r)d^3r$$

$$\equiv \sum_{\alpha,\beta=\uparrow\downarrow} \rho_{\alpha,\beta}(r)v_{\alpha,\beta}(r)$$

$\{\rho(r),\vec{m}(r)\}$: 4 independent functions

$\rho_{\alpha\beta}$ is Hermitian $\Rightarrow$ 4 independent functions

Non-collinear LSDA:

$\vec{r}$ given point in space:

① Find unitary matrix U(r) such that

$$U^{+}(r)(\rho_{\alpha\beta})U(r) = \begin{pmatrix} n_{\uparrow}(r) & 0 \\ 0 & n_{\downarrow}(r) \end{pmatrix}$$

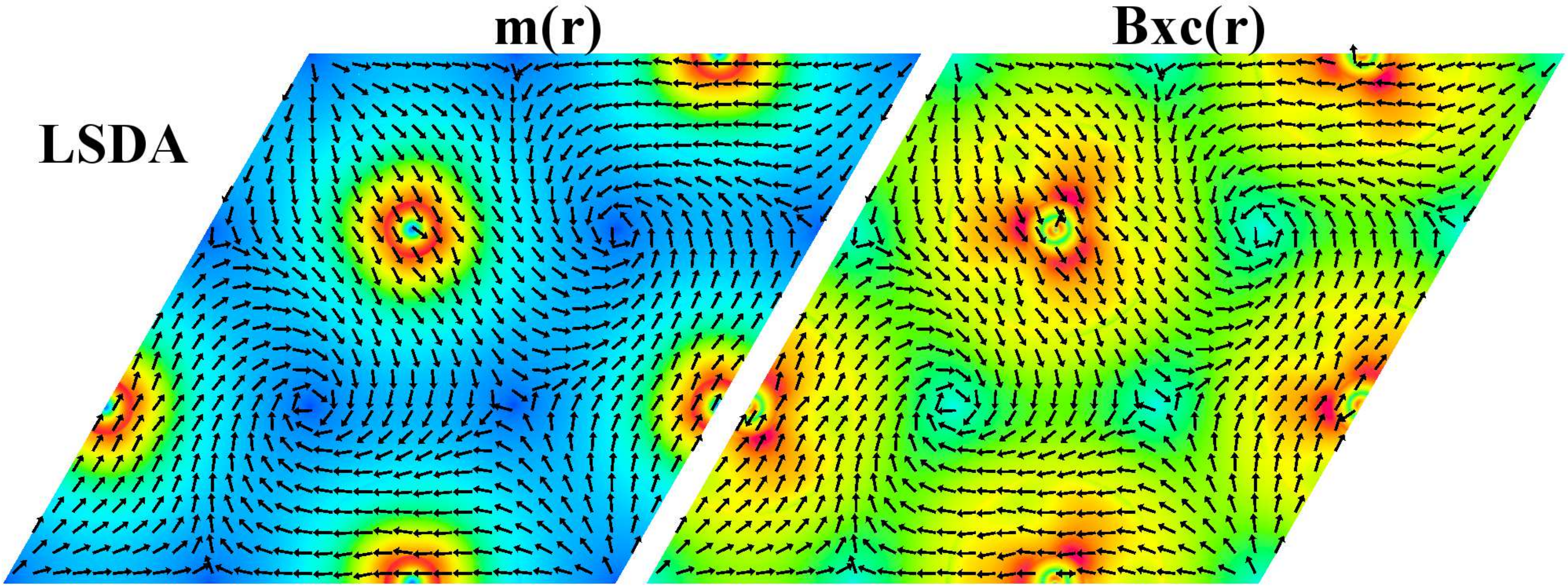
② Calculate $v_{xc}^{\uparrow}(r)$ and $v_{xc}^{\downarrow}(r)$ from $\{n_{\uparrow}, n_{\downarrow}\}$

using the normal LSDA expressions

③ $$(v_{xc}^{\alpha\beta}) = U(r)\begin{pmatrix} v_{xc}^{\uparrow}(r) & 0 \\ 0 & v_{xc}^{\downarrow}(r) \end{pmatrix}U^{+}(r)$$

in this approximation $\vec{B}_{xc}(r)$ and $\vec{m}(r)$ may change their direction in space, but locally they are always parallel

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)

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

in absence of external magnetic field

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

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 xc functional for which m(r) and B_{xc}(r) are not locally parallel

Enforce property of the exact xc functional:

$$\boldsymbol{B}_{xc}^{exact}\left(\boldsymbol{r}\right)=\nabla\times A_{xc}^{exact}\left(\boldsymbol{r}\right)$$

K. Capelle, E.K.U. Gross, PRL 78, 1872 (1997)

By virtue of Helmholtz' theorem, any vector field can be decomposed as:

$$\boldsymbol{B}_{xc}^{GGA}\left(\boldsymbol{r}\right)=\nabla\times A_{xc}\left(\boldsymbol{r}\right)+\nabla\phi(r)$$

Enforce exact property by subtracting source term!

Explicit construction:

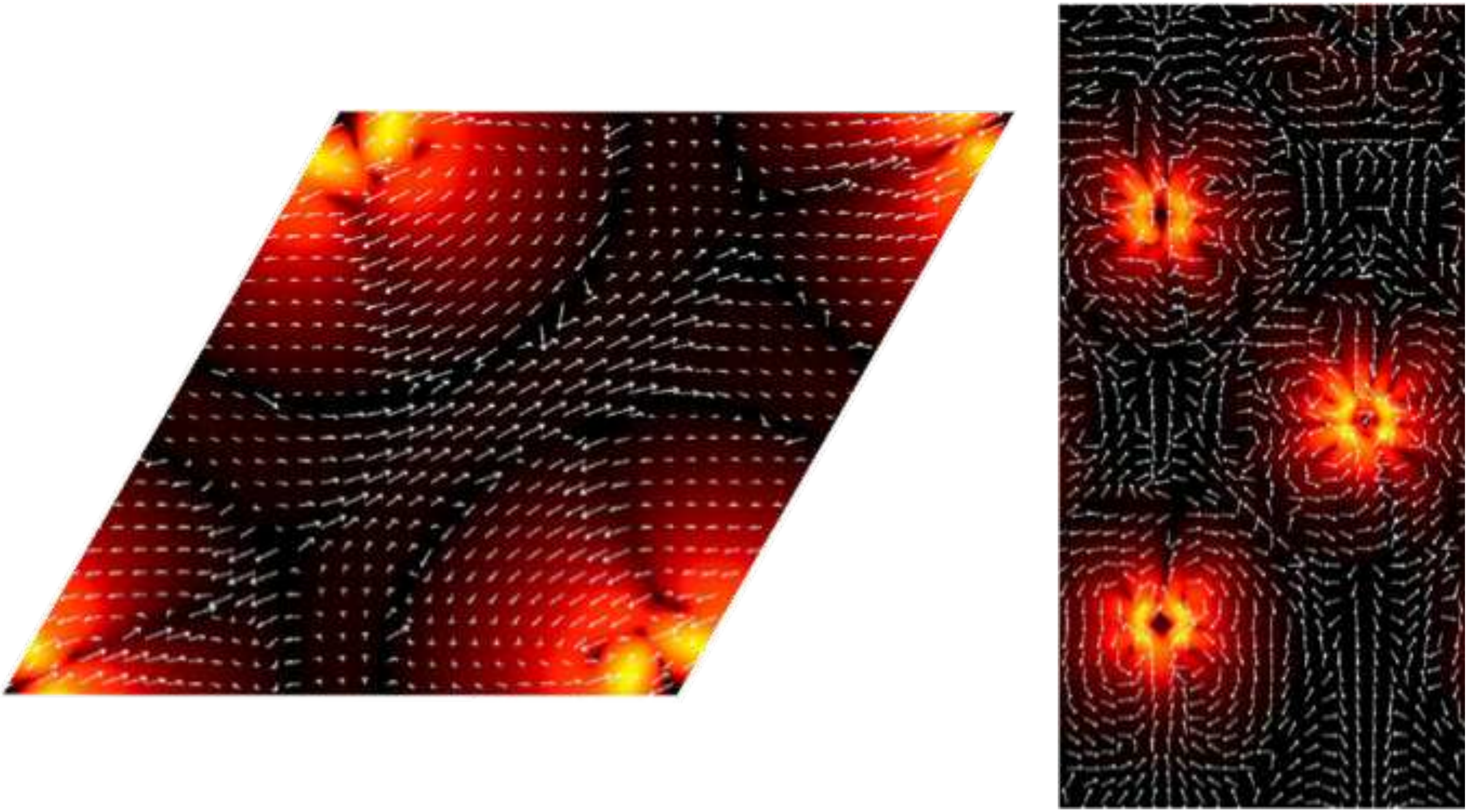
S. Sharma, E.K.U. Gross, A. Sanna, K. Dewhurst, JCTC14, 1247 (2018)

$$\nabla^2 \phi(\boldsymbol{r}) = 4\pi\nabla \cdot B_{xc}^{GGA}(\boldsymbol{r})$$

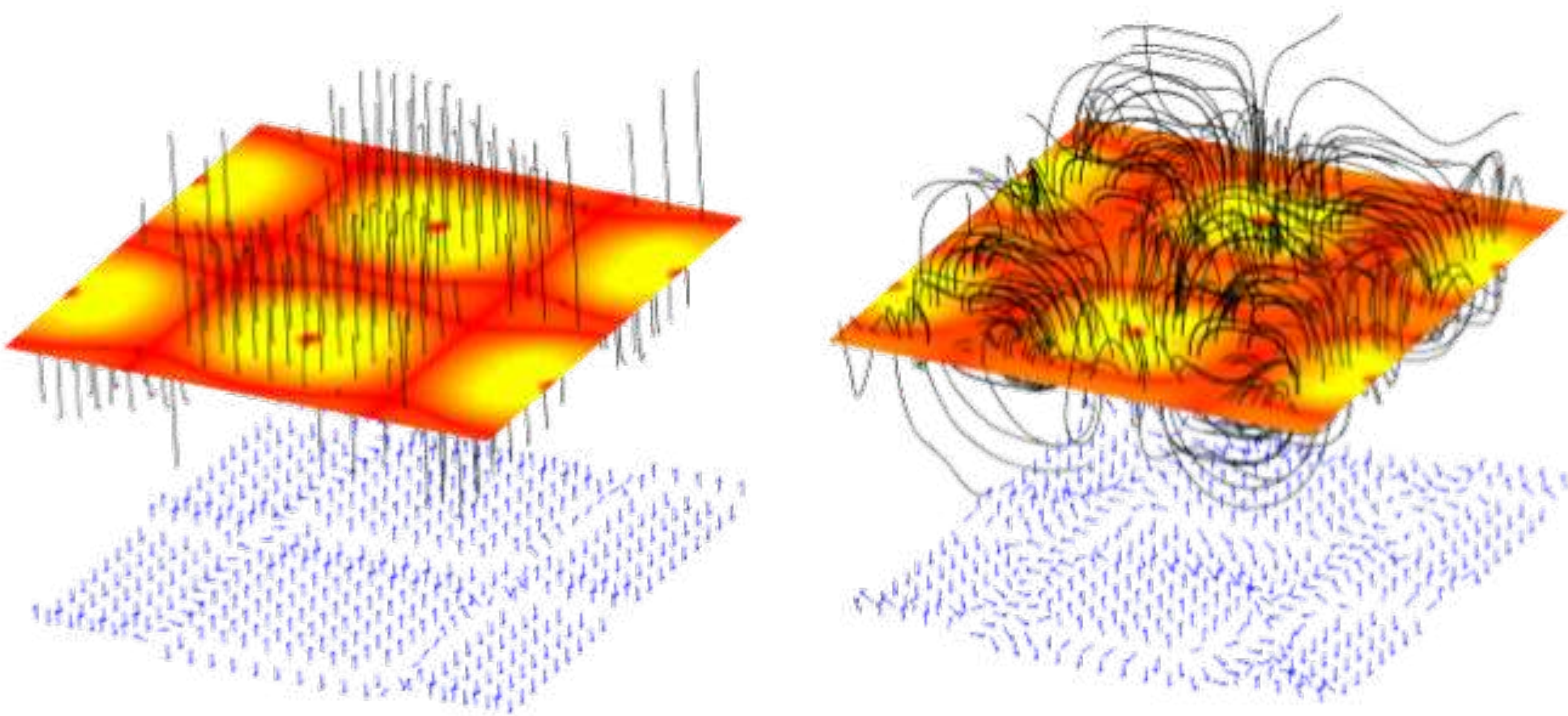
$$\tilde{B}_{xc}(\boldsymbol{r}) \cong B_{xc}^{GGA}(\boldsymbol{r}) - \frac{1}{4\pi}\nabla\phi(\boldsymbol{r})$$

$$B_{xc}^{SF}(\boldsymbol{r}) = s\,\tilde{B}_{xc}(\boldsymbol{r})$$

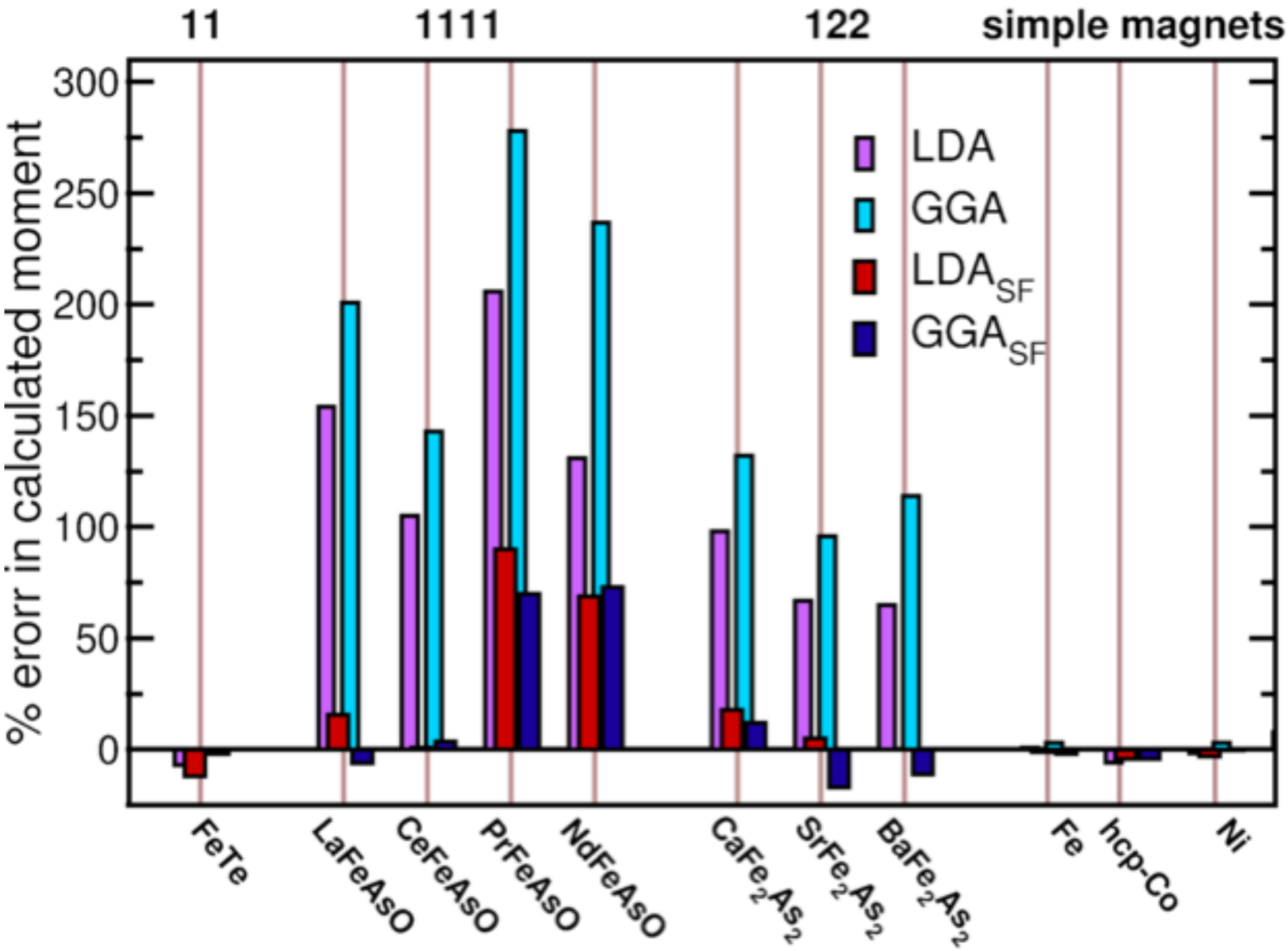
Scaling factor, s, only depends on underlying functional (GGA/LSDA), nothing else

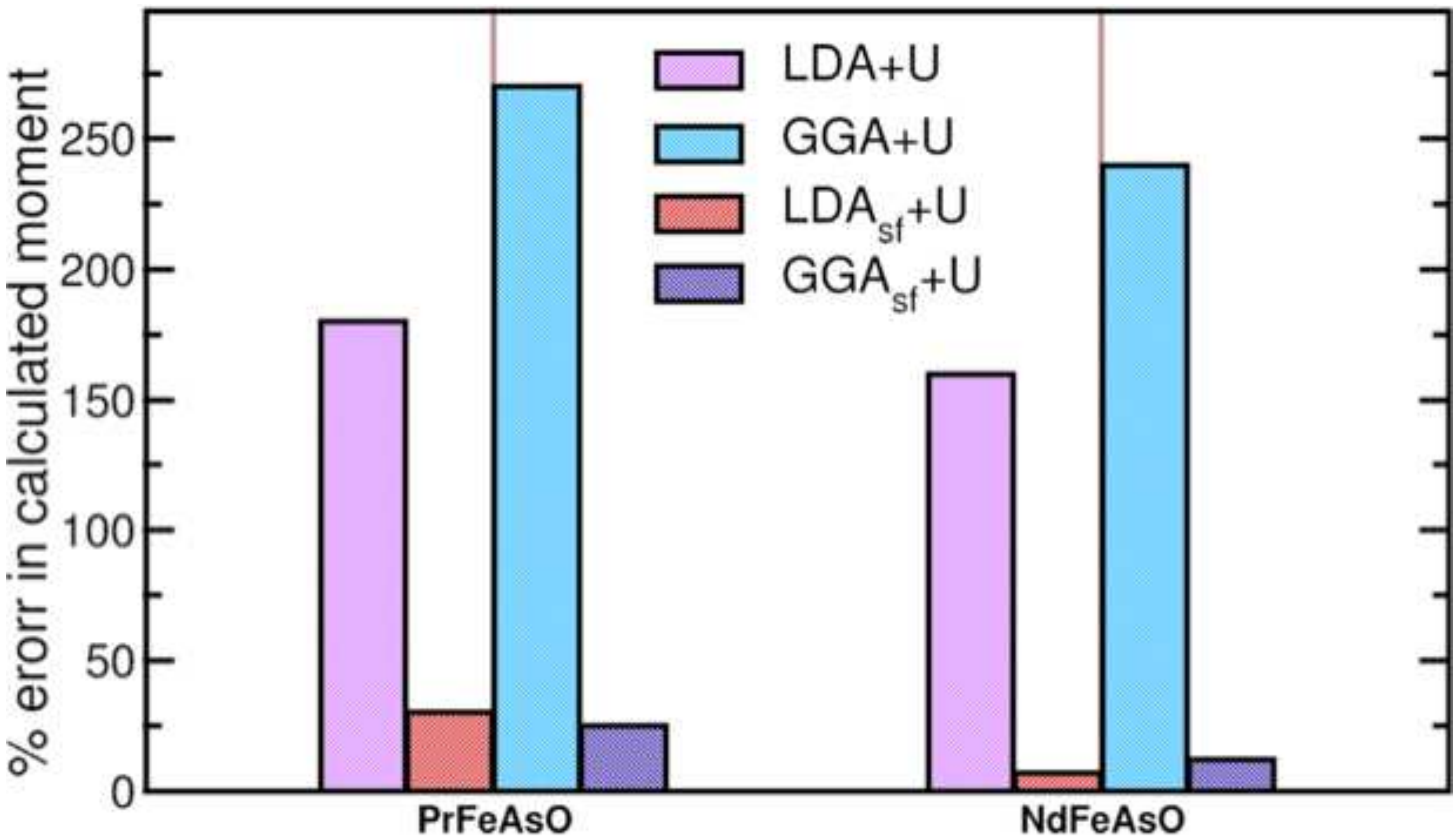


Left panel: Local xc torque for bulk Ni in (111) plane. Right panel: Local xc torque for 3ML Ni@5ML Pt in the (110) plane. The arrows indicate the direction and colors the magnitude.



The vector field B_{xc} for $BaFe_2As_2$ projected in a plane containing Fe atoms. Plot (a) is LSDA and plot (b) is source-free LSDA. The colored plane shows the magnitude of B_{xc} and the arrows indicate the direction. The black field lines originate from a regular grid in the plane and follow the vector field. LSDA field lines show a plane of magnetic monopoles while making LSDA source-free leads to more complicated but physical field lines. The arrows indicate that the removal of the source term leads to enhancement of non-collinearity.


11
1111
122
simple magnets
% erorr in calculated moment
300
250
200
150
100
50
0
LDA
GGA
LDA$_{SF}$
GGA$_{SF}$
FeTe
LaFeAsO
CeFeAsO
PrFeAsO
NdFeAsO
CaFe$_2$As$_2$
SrFe$_2$As$_2$
BaFe$_2$As$_2$
Fe
hcp-Co
Ni

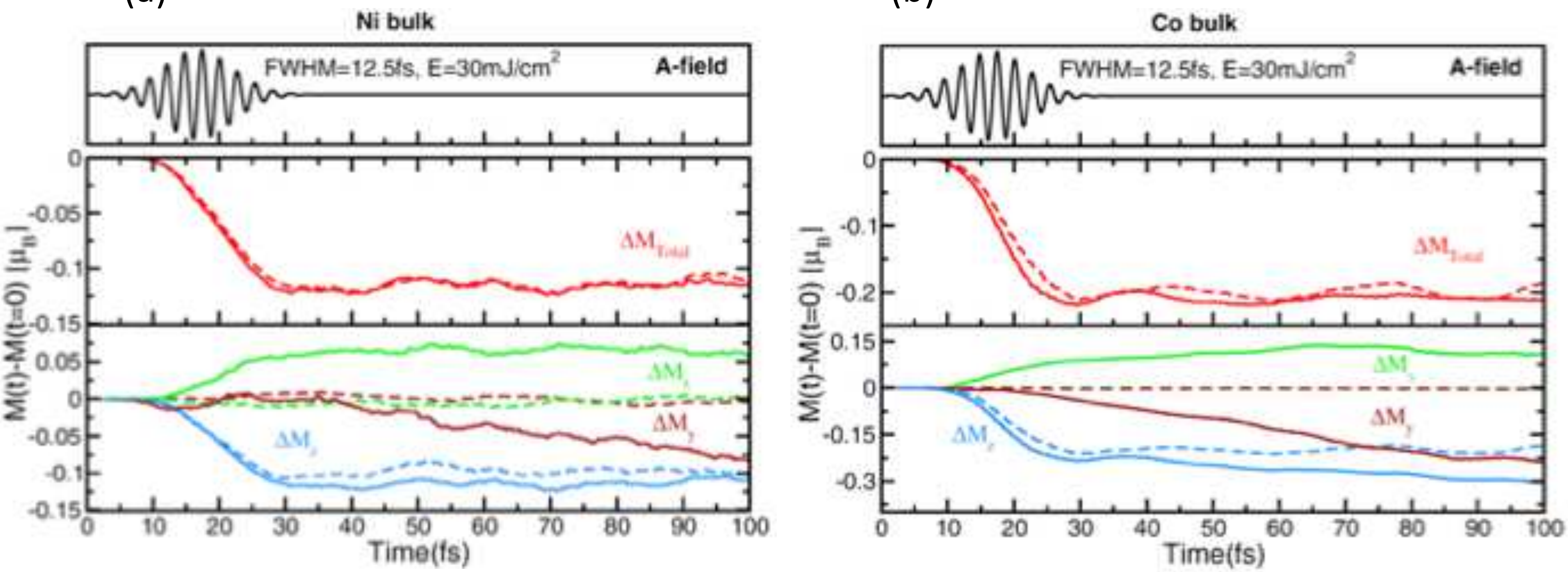

% erorr in calculated moment
0
50
100
150
200
250
LDA+U
GGA+U
LDA$_{sf}$+U
GGA$_{sf}$+U
PrFeAsO
NdFeAsO

Magnetic moment per atom. Calculations are performed using LSDA+U, PBE-GGA+U, $LSDA_{SF}$ + U and PBE-GGA_{SF} + U.

Material	Expt	LSDA	PBE-GGA	$LSDA_{SF}$	PBE-GGA_{SF}
PrFeAsO	Fe: 0.5	1.40	1.9	0.65	0.63
	Pr: 0.87	0.30	0.30	0.81	0.83
NdFeAsO	Fe: 0.54	1.42	1.84	0.50	0.61
	Nd: 0.9	2.44	1.25	0.80	0.89

(a)

(b)

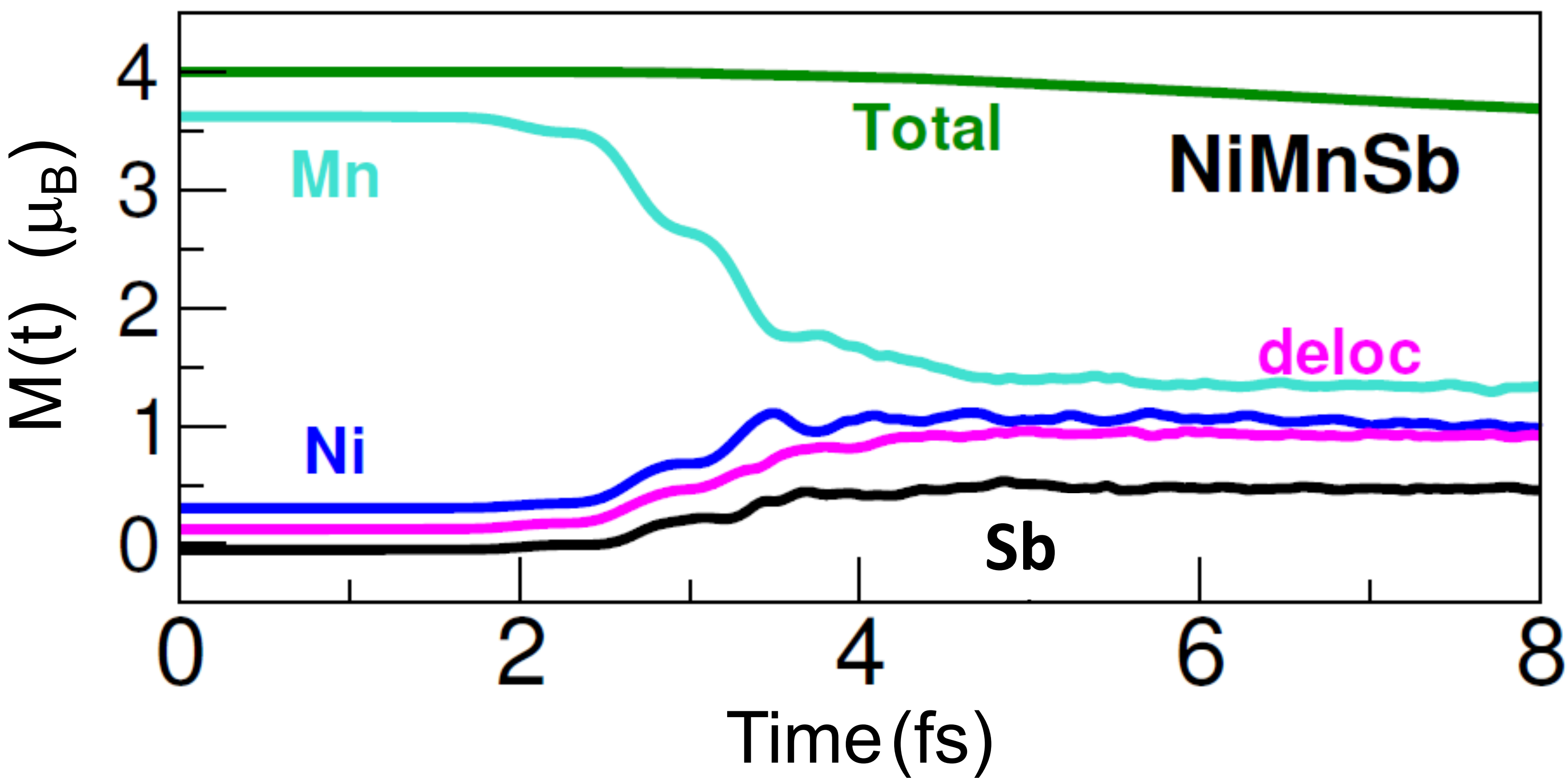


(a) Middle panel shows the total moment (red) and the bottom panel x (green), y (brown) and z (blue) projected moments for bulk Ni as a function of time. Dashed lines are the results obtained using the ALSDA and full lines the results obtained using the source-free functional. (b) The same as (a) but for bulk Co.

Optically induced spin transfer (OISTR)

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

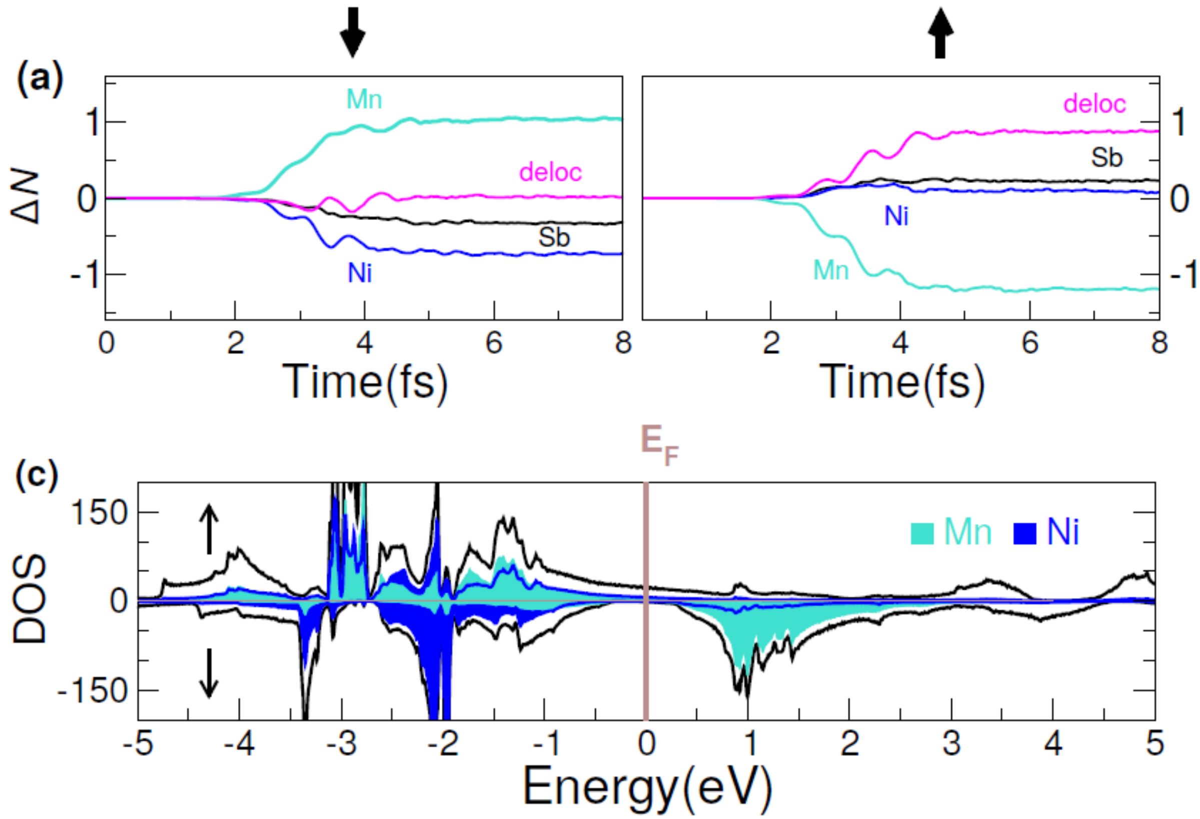
K. Dewhurst, P. Elliott, S. Shallcross, E.K.U. Gross, S. Sharma,
Nano Lett. 18, 1842 (2018)

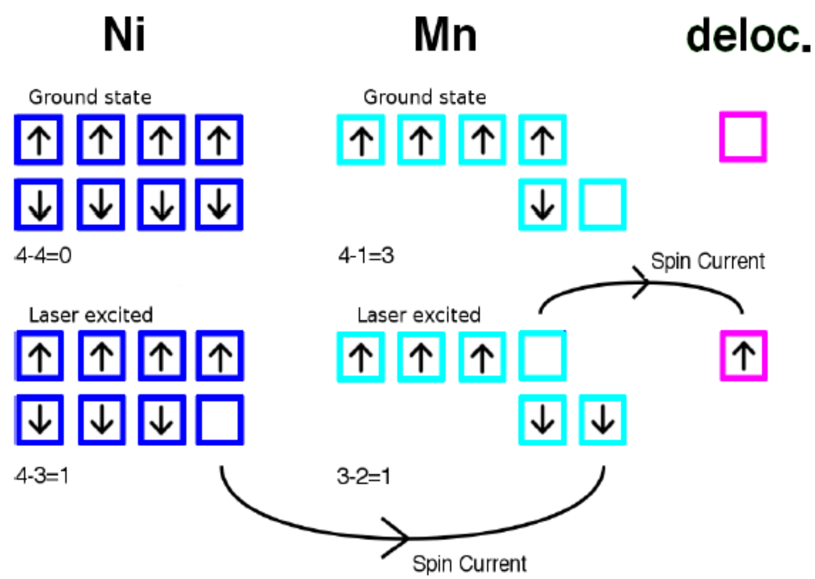


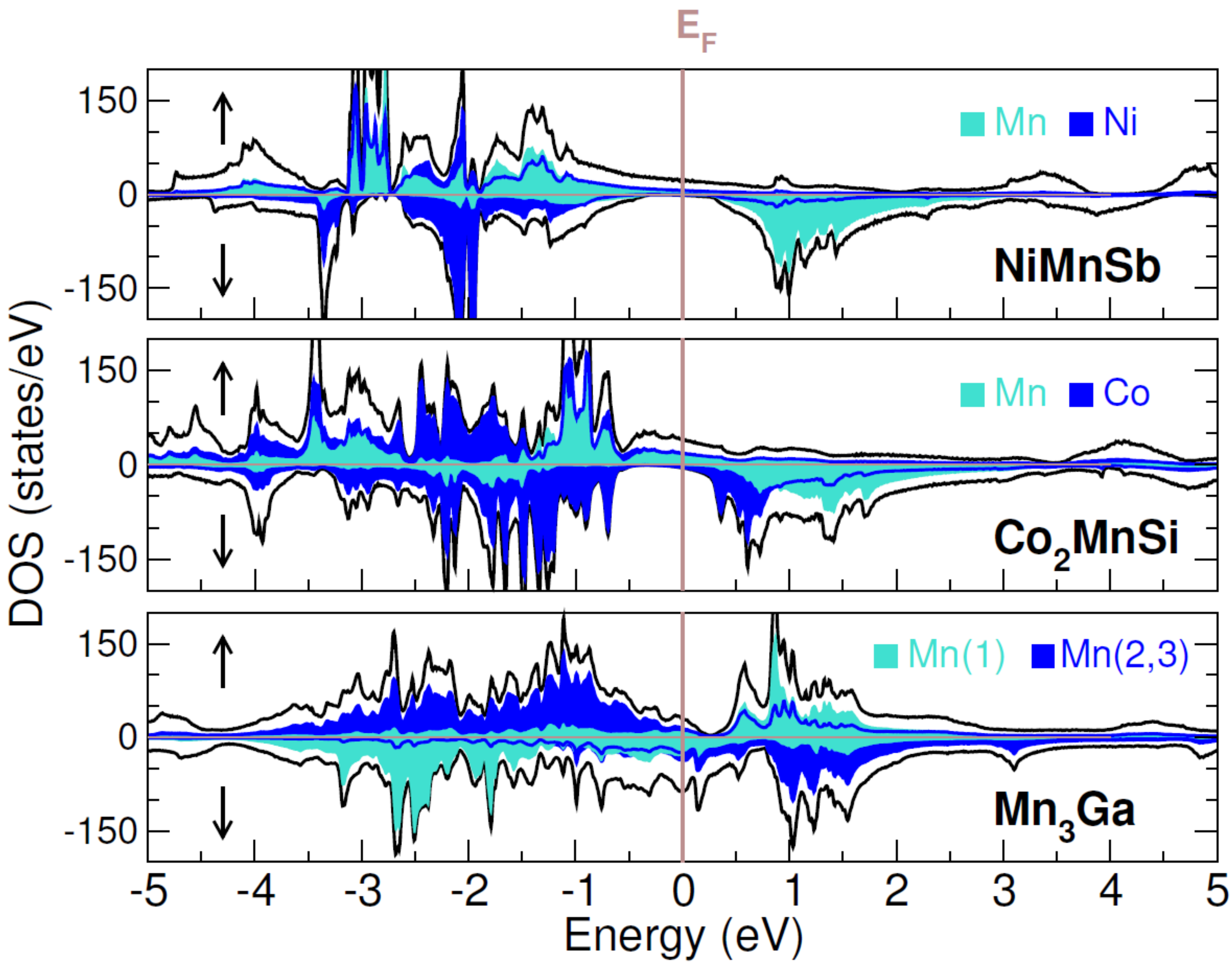
Global moment |M(t)| nearly preserved

Local moments around each atom change

NiMnSb

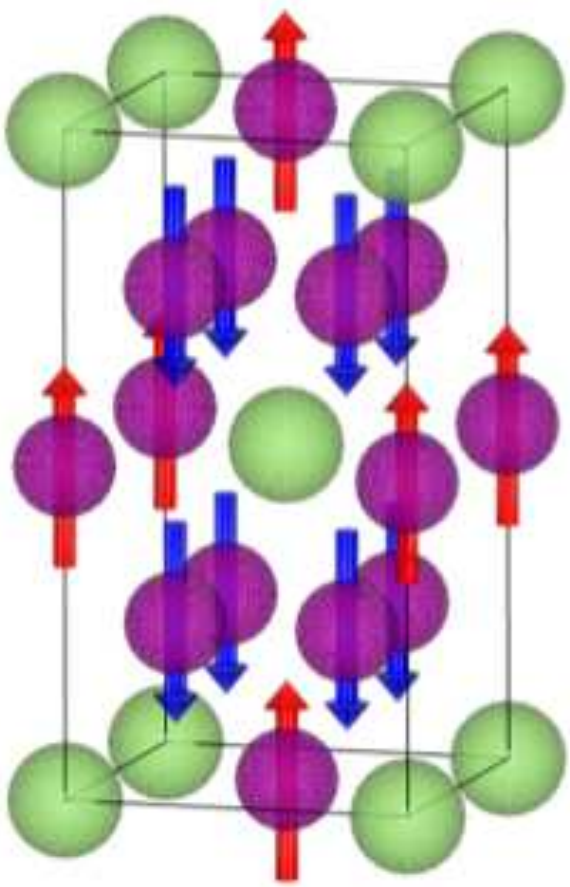

(a)
Mn
deloc
Sb
Ni
ΔN
Time(fs)
deloc
Sb
Ni
Mn
Time(fs)
(c)
E_F
DOS
Mn
Ni
Energy(eV)


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



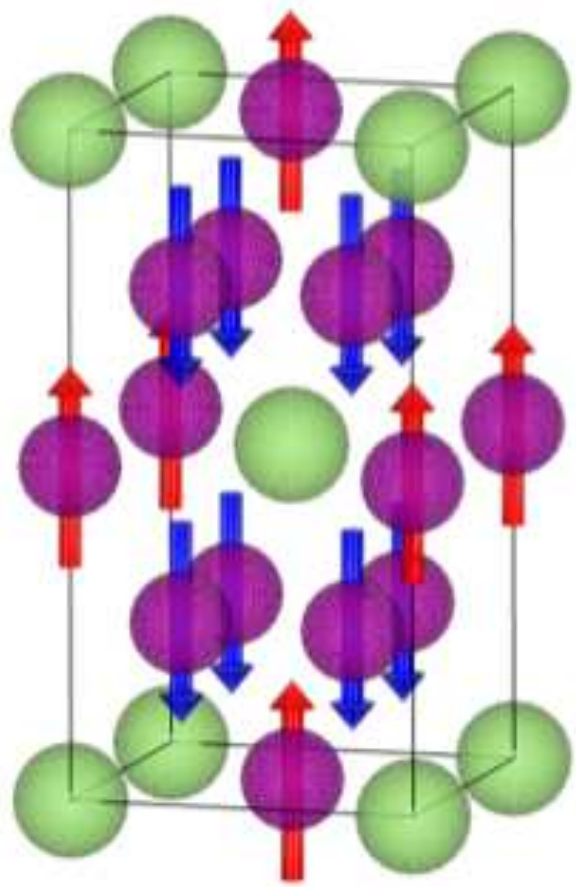
E_F
150
0
-150
Mn
Ni
NiMnSb
Mn
Co
Co2MnSi
Mn(1)
Mn(2,3)
Mn3Ga
DOS (states/eV)
-5
-4
-3
-2
-1
0
1
2
3
4
5
Energy (eV)

Mn$_3$Ga (ferri-magnet)



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

Mn_3Ga (ferri-magnet)



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

OISTR experimentally confirmed! (Aeschlimann group, 2018)

Future aspects in the field of laser-driven spin dynamics:

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

- **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**

Optimal control using short laser pulses

<u>**Review Article on Quantum Optimal Control Theory**</u>**:**
J. Werschnik, E.K.U. Gross, J. Phys. B 40, R175-R211 (2007)

Optimal Control Theory (OCT)

Normal question:

What happens if a system is exposed to a given laser pulse?

Inverse question (solved by OCT):

Which is the laser pulse that achieves a prescribed goal (target)?

possible targets:

a) system should end up in a given final state ϕ_f at the end of the pulse

b) wave function should follow a given trajectory in Hilbert space

c) density should follow a given classical trajectory r(t)

Optimal control of static targets (standard formulation)

For <u>given</u> target state Φ_f , maximize the functional:

$$J_1 = \left| \left\langle \Psi(T) \middle| \Phi_f \right\rangle \right|^2 = \left\langle \Psi(T) \middle| \Phi_f \right\rangle \left\langle \Phi_f \middle| \Psi(T) \right\rangle = \left\langle \Psi(T) \middle| \hat{O} \middle| \Psi(T) \right\rangle$$

Optimal control of static targets (standard formulation)

For <u>given</u> target state Φ_f , maximize the functional:

$$J_1 = \left| \left\langle \Psi(T) \middle| \Phi_f \right\rangle \right|^2 = \left\langle \Psi(T) \middle| \underbrace{\Phi_f \rangle \langle \Phi_f}_{\hat{O}} \middle| \Psi(T) \right\rangle = \left\langle \Psi(T) \middle| \hat{O} \middle| \Psi(T) \right\rangle$$

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$\hat{O}$

with the constraints:

$$J_2 = -\alpha \left[\int_0^T dt\, \varepsilon^2(t) - E_0 \right]$$

E_0 = <u>given</u> fluence

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with the constraints:

$$J_2 = -\alpha \left[\int_0^T dt\, \varepsilon^2(t) - E_0 \right]$$

E_0 = given fluence

$$J_3[\varepsilon, \Psi, \chi] = -2\,\mathrm{Im} \int_0^T dt \langle \chi(t) | -i\partial_t - \left[\hat{T} + \hat{V} - \mu\varepsilon(t) \right] | \Psi(t) \rangle$$

Optimal control of static targets (standard formulation)

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TDSE

Optimal control of static targets (standard formulation)

For <u>given</u> target state Φ_f , maximize the functional:

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TDSE

GOAL: Maximize $J = J_1 + J_2 + J_3$

Set the total variation of $J = J_1 + J_2 + J_3$ equal to zero:

Control equations

1. Schrödinger equation with initial condition:

$$\delta_\chi J = 0 \rightarrow \boxed{i\partial_t \psi(t) = \hat{H}(t)\psi(t), \quad \psi(0) = \phi}$$

2. Schrödinger equation with final condition:

$$\delta_\psi J = 0 \rightarrow \boxed{i\partial_t \chi(t) = \hat{H}(t)\chi(t), \quad \chi(T) = \hat{O}\psi(T)}$$

3. Field equation:

$$\delta_\varepsilon J = 0 \rightarrow \boxed{\varepsilon(t) = \frac{1}{\alpha}\,\mathrm{Im}\left\langle \chi(t) \middle| \hat{\mu} \middle| \psi(t) \right\rangle}$$

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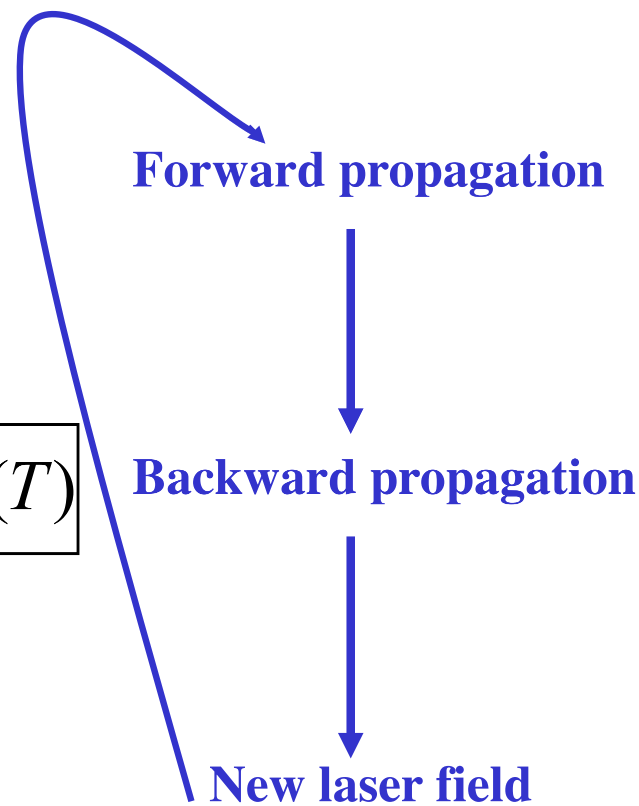
Algorithm



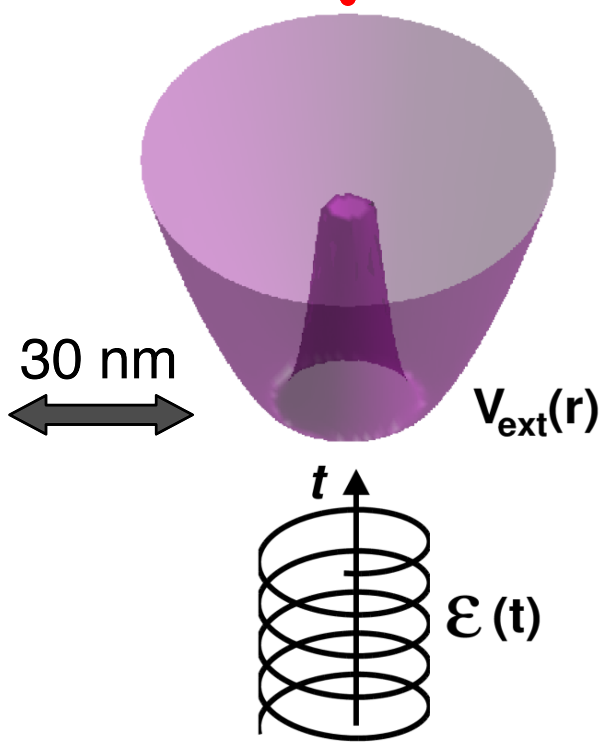
Algorithm monotonically convergent: W. Zhu, J. Botina, H. Rabitz, JCP 108, 1953 (1998))

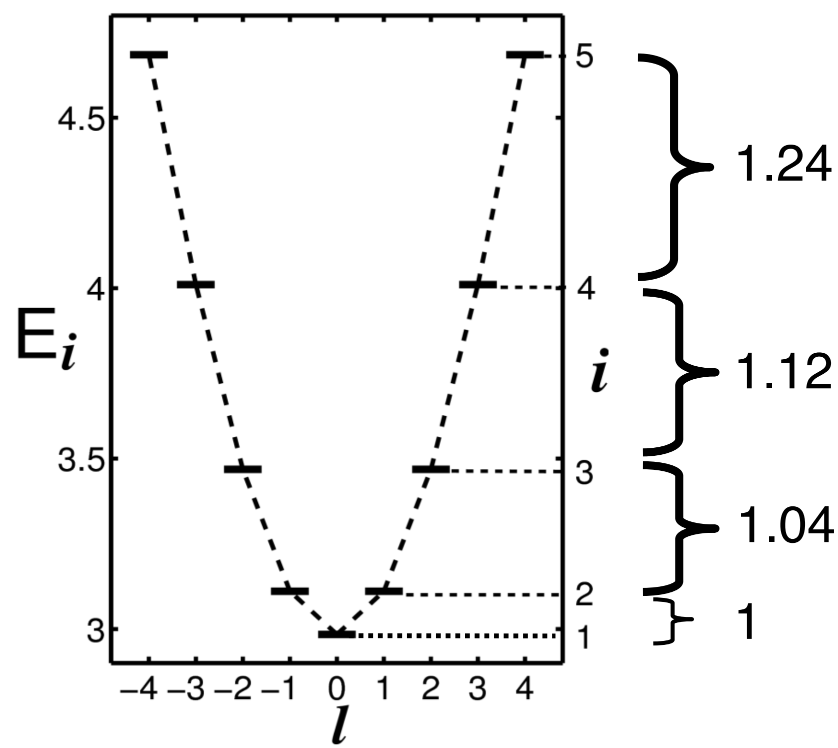
Quantum ring: Control of circular current

TDSE: $$i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r},t) = \left[\hat{H}_0 + e\,\mathbf{r}\,\epsilon(t)\right]\Psi(\mathbf{r},t)$$

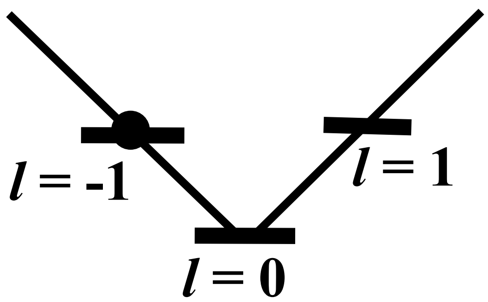
$$\hat{H}_0 = -\frac{\hbar^2}{2m^*}\nabla^2 + \frac{1}{2}m^*\omega_0^2 r^2 + V_0 e^{-r^2/d^2}$$

$$\epsilon(t) = (\epsilon_x(t), \epsilon_y(t))$$

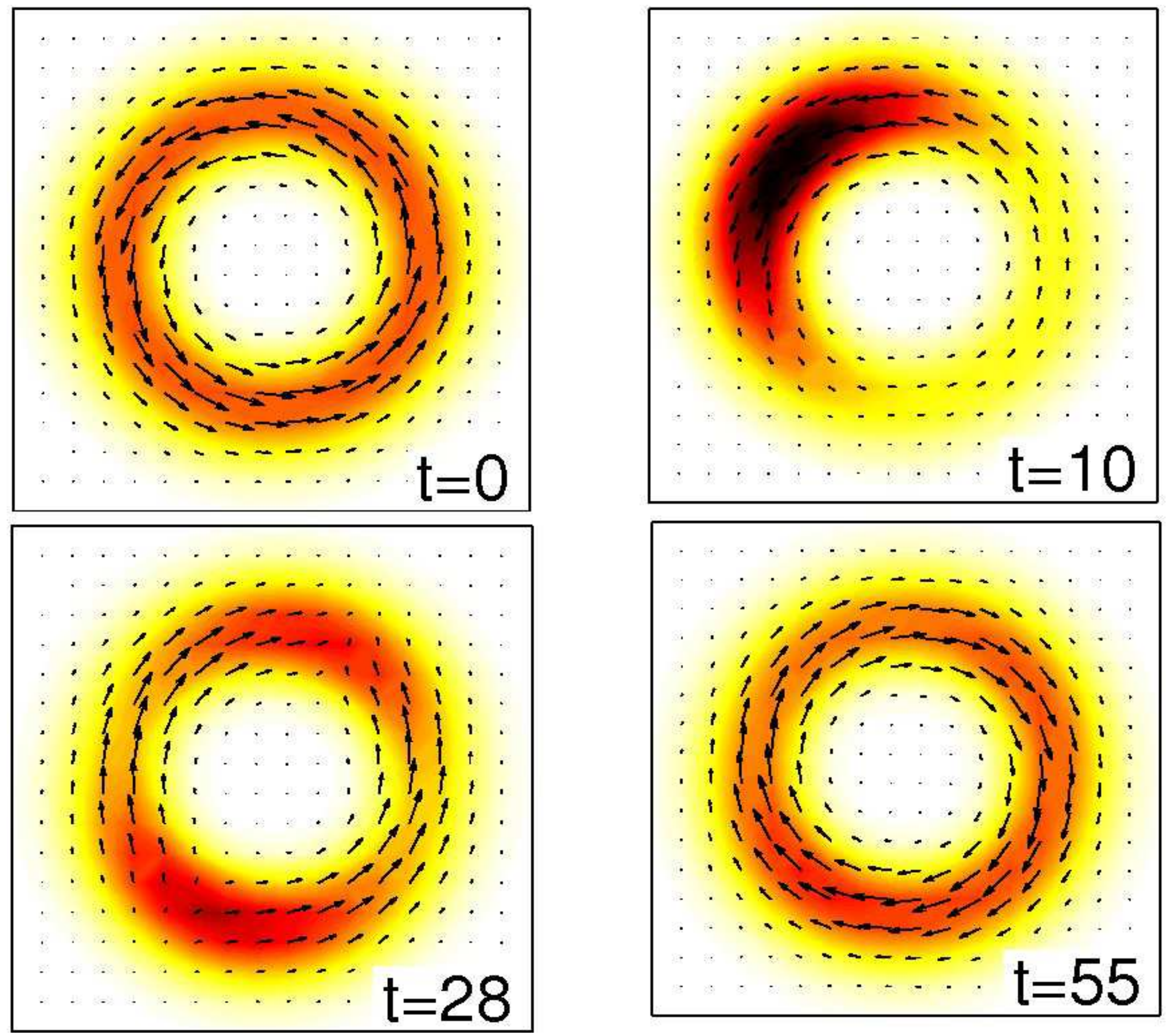




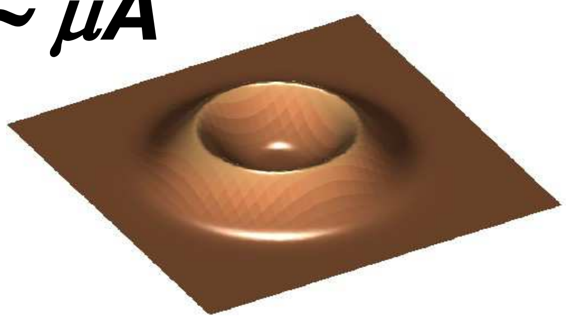
Control of currents



$|\psi(t)|^2$ and $\mathbf{j}(t)$



$I \sim \mu A$



E. Räsänen, A. Castro, J. Werschnik, A. Rubio, E.K.U.G., PRL 98, 157404 (2007)

OPTIMAL CONTROL OF TIME-DEPENDENT TARGETS

Maximize $$J = J_1 + J_2 + J_3$$

$$J_1[\Psi] = \frac{1}{T}\int_0^T dt \left\langle \Psi(t) \middle| \hat{O}(t) \middle| \Psi(t) \right\rangle$$

$$J_2 = -\alpha\left[\int_0^T dt\, \varepsilon^2(t) - E_0\right]$$

$$J_3[\varepsilon, \Psi, \chi] = -2\,\mathrm{Im}\int_0^T dt \left\langle \chi(t) \middle| -i\partial_t - \left[\hat{T} + \hat{V} - \mu\varepsilon(t)\right] \middle| \Psi(t) \right\rangle$$

Set the total variation of $J = J_1 + J_2 + J_3$ equal to zero:

Control equations

1. Schrödinger equation with initial condition:

$$\delta_\chi J = 0 \rightarrow \boxed{i\partial_t \psi(t) = \hat{H}(t)\psi(t), \quad \psi(0) = \phi}$$

2. Schrödinger equation with final condition:

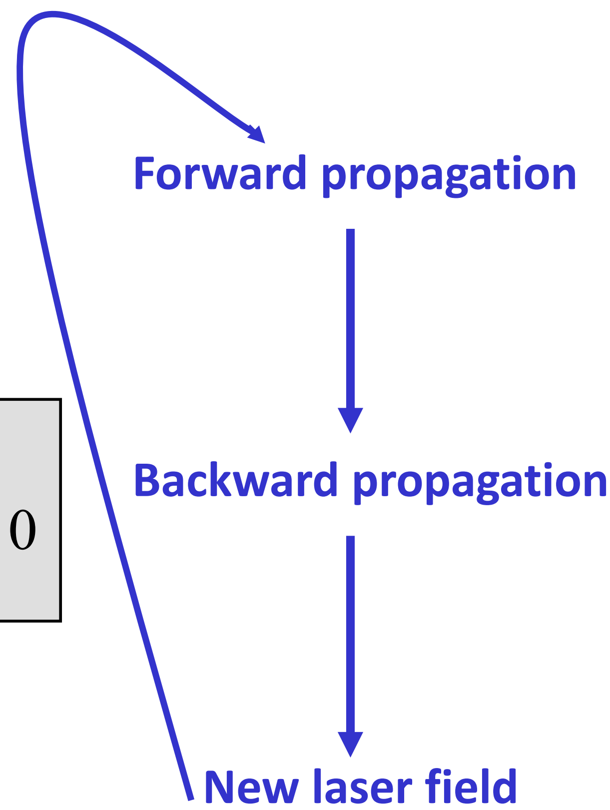
Inhomogenous TDSE :

$$\delta_\psi J = 0 \rightarrow \left[i\partial_t - \hat{H}(t) \right] \chi(t) = -\frac{i}{T} \hat{O}(t)\psi(t), \quad \chi(T) = 0$$

3. Field equation:

$$\delta_\varepsilon J = 0 \rightarrow \boxed{\varepsilon(t) = \frac{1}{\alpha} \mathrm{Im} \left\langle \chi(t) \middle| \hat{\mu} \middle| \psi(t) \right\rangle}$$

Algorithm



Algorithm monotonically convergent:
I. Serban, J. Werschnik, E.K.U.Gross., Phys. Rev. A 71, 053810 (2005)

Control path in real space

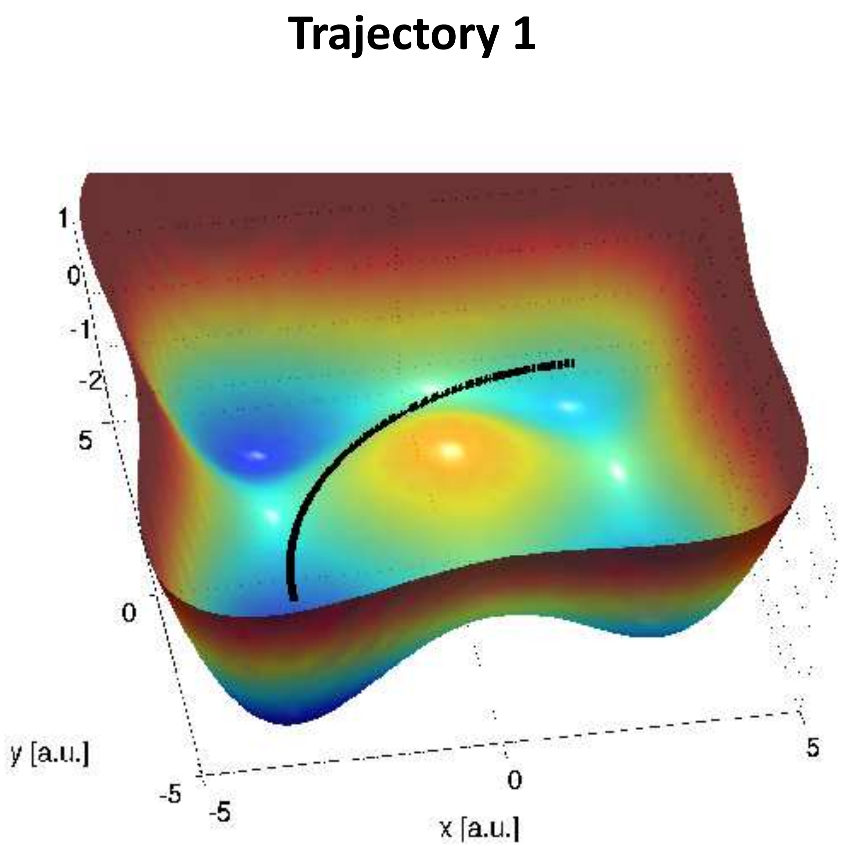
$$\hat{O}(t) = \delta(r - r_0(t)) \approx \frac{1}{\sqrt{2\pi\sigma^2}} e^{-(r-r_0(t))^2/2\sigma^2}$$

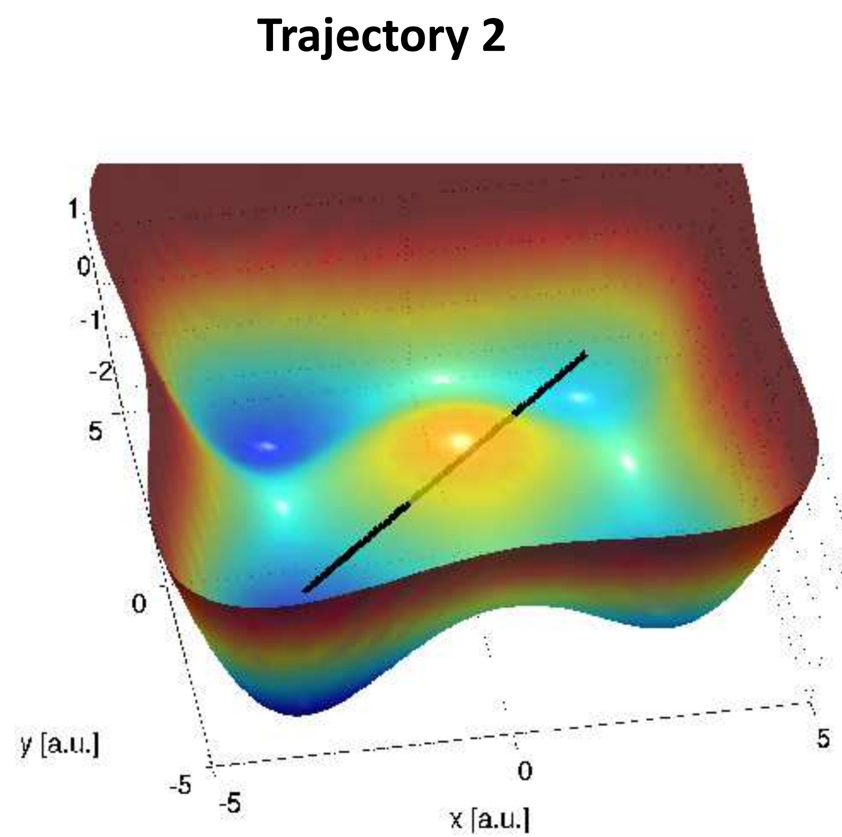
with <u>given</u> trajectory $r_0(t)$.

Algorithm maximizes the density along the path $r_0(t)$:

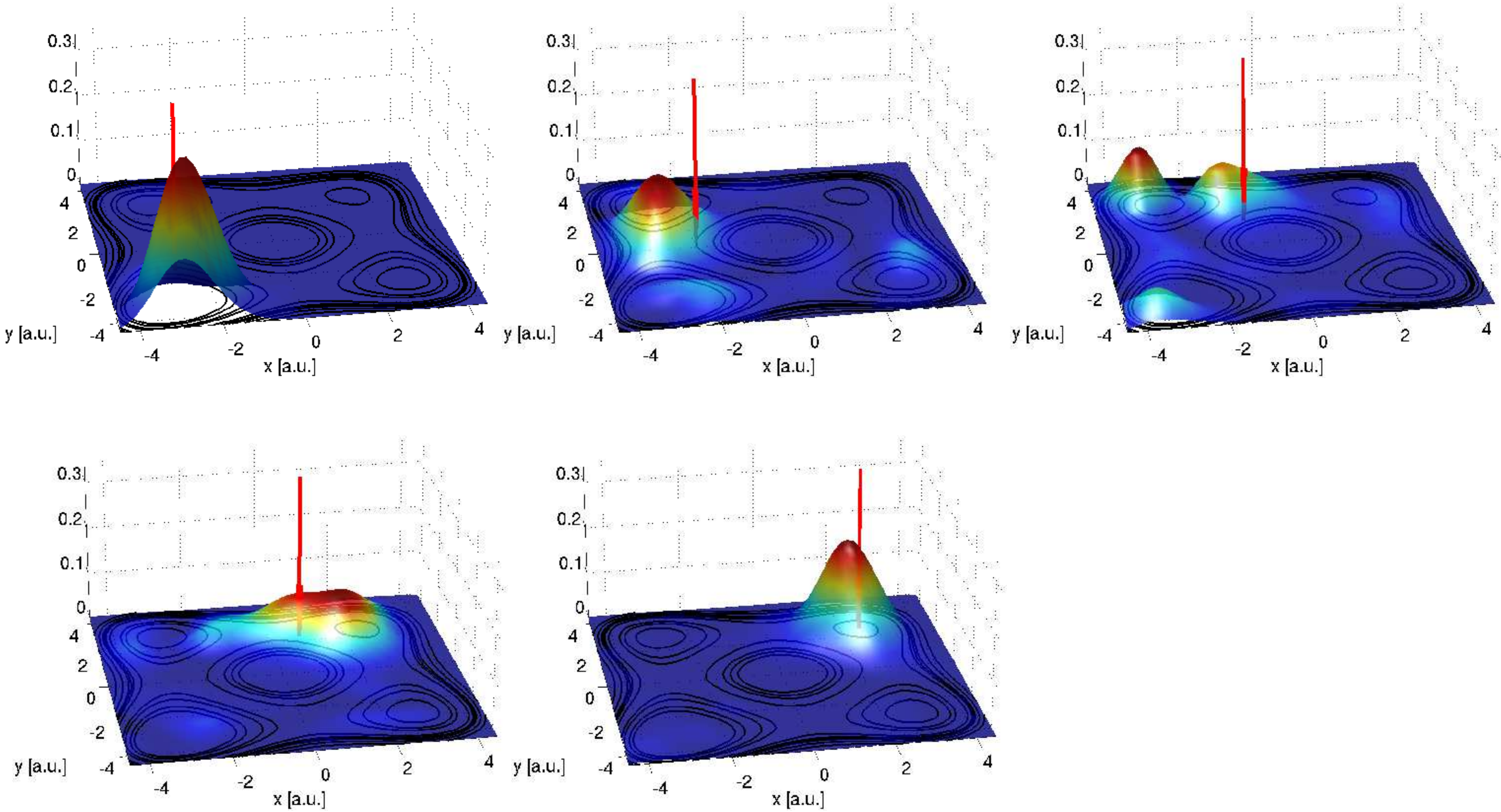
I. Serban, J. Werschnik, E.K.U.G. Phys. Rev. A 71, 053810 (2005)

Control of charge transfer along selected pathways

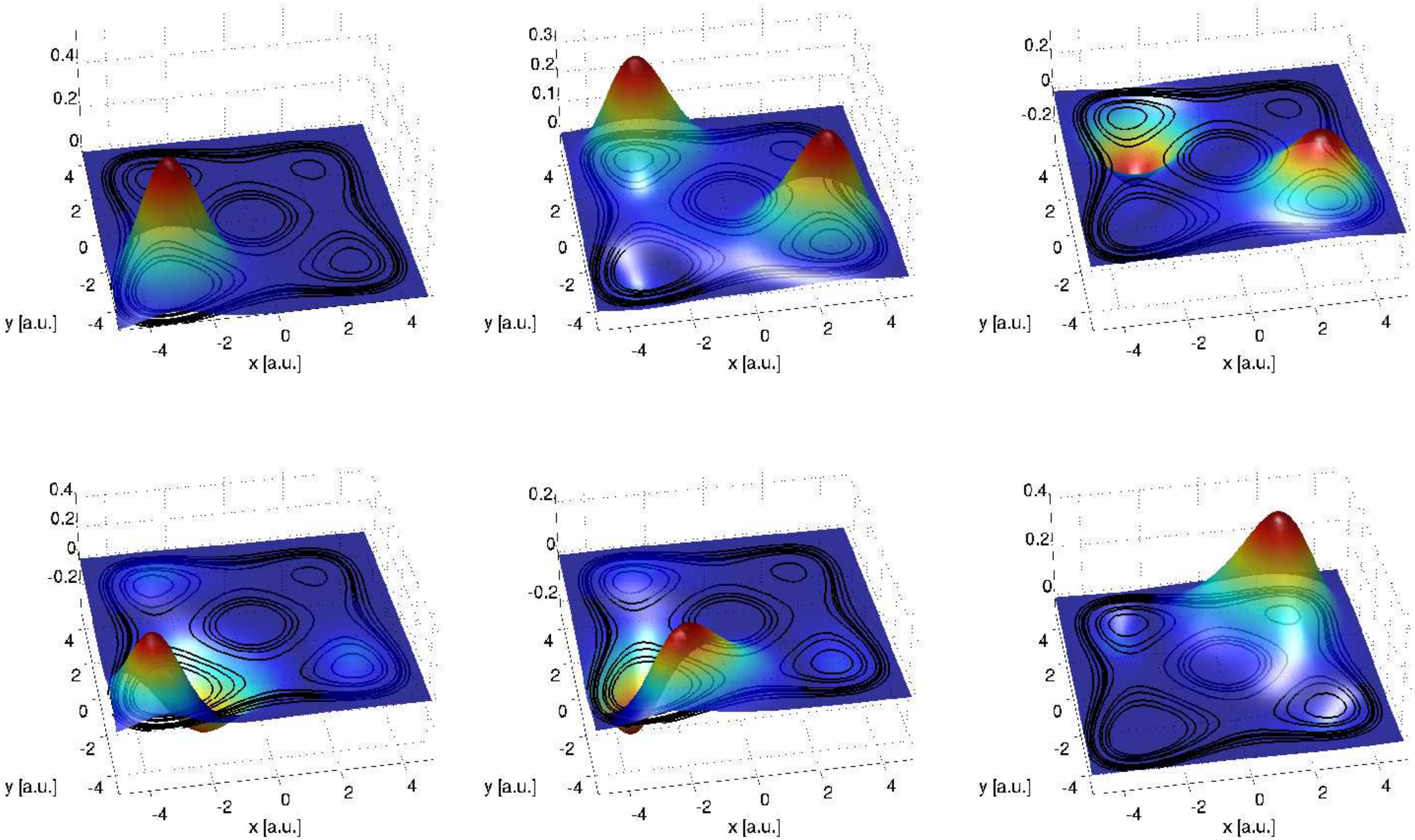




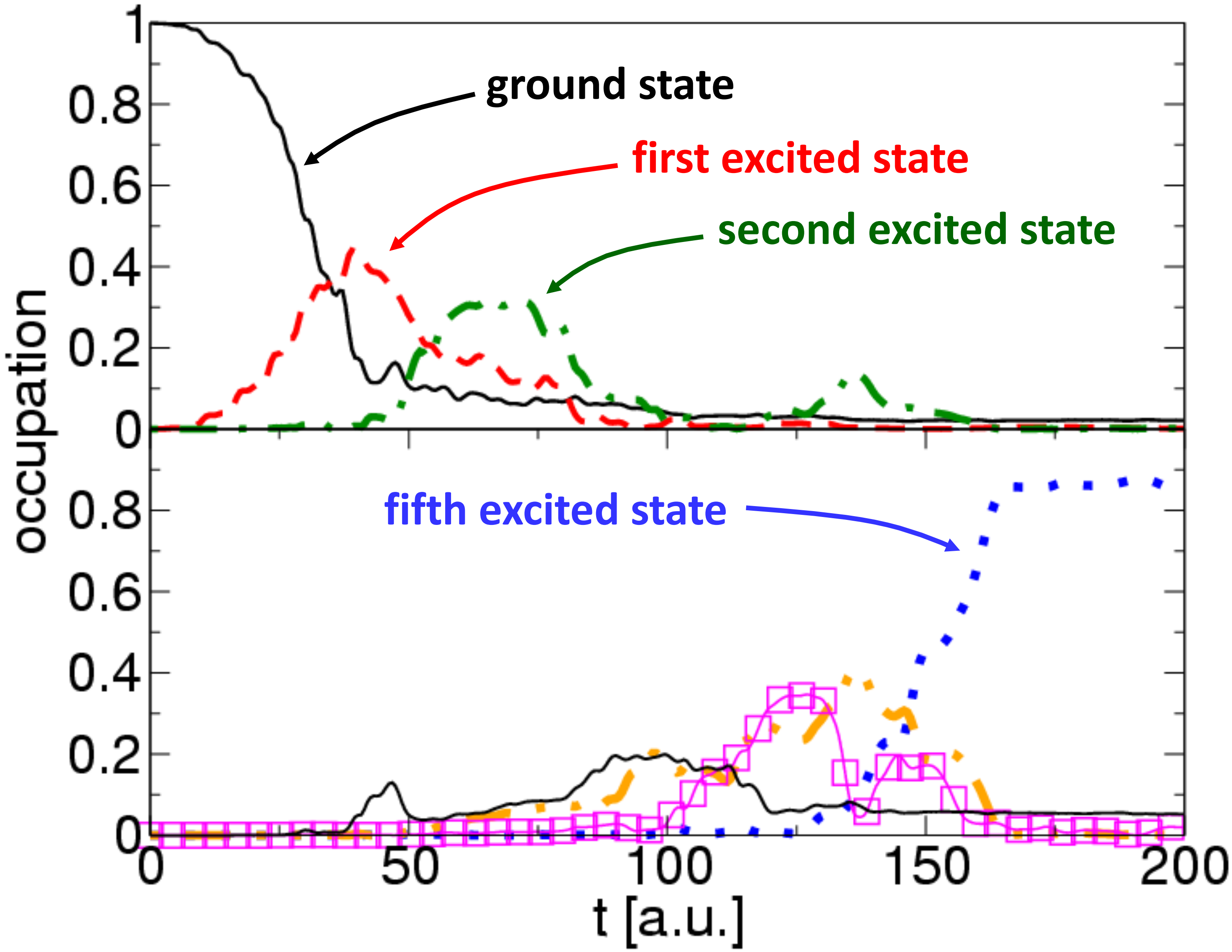
Time-evolution of wavepacket with the optimal laser pulse for trajectory 1



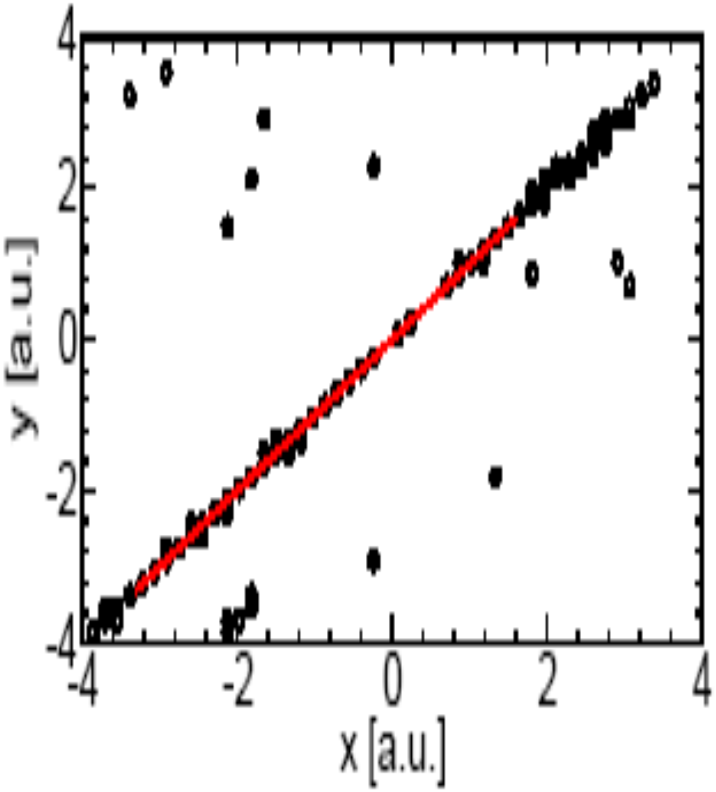
Lowest six eigenstates

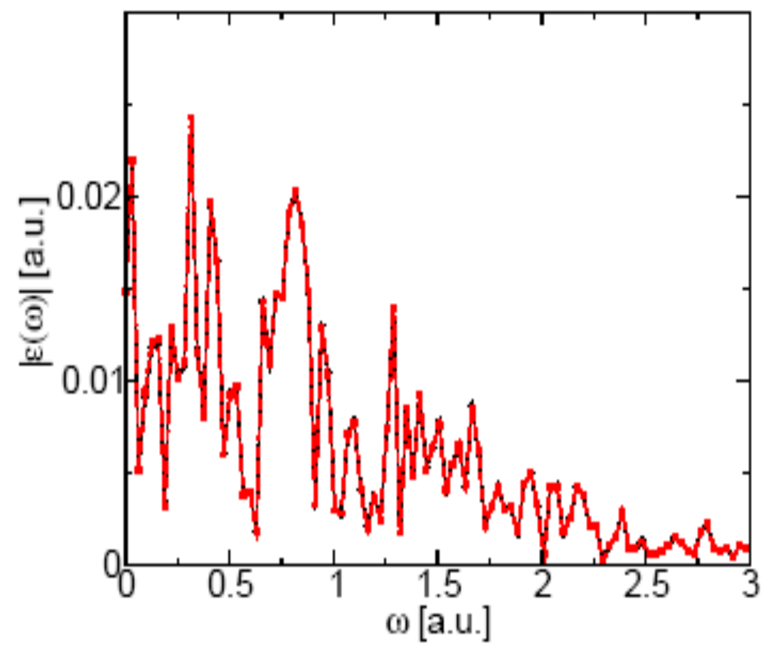


Populations of eigenstates



Trajectory 2





Control of many-body systems

- Formally the same OCT equations

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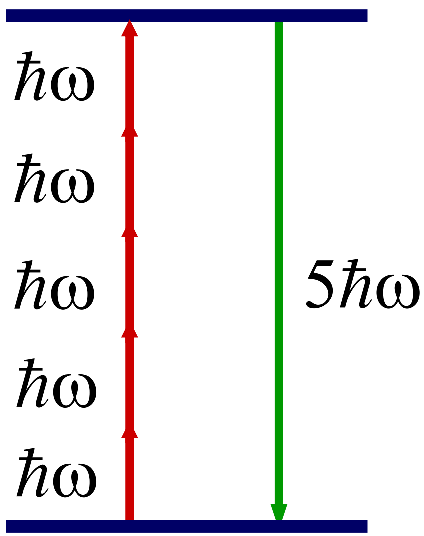
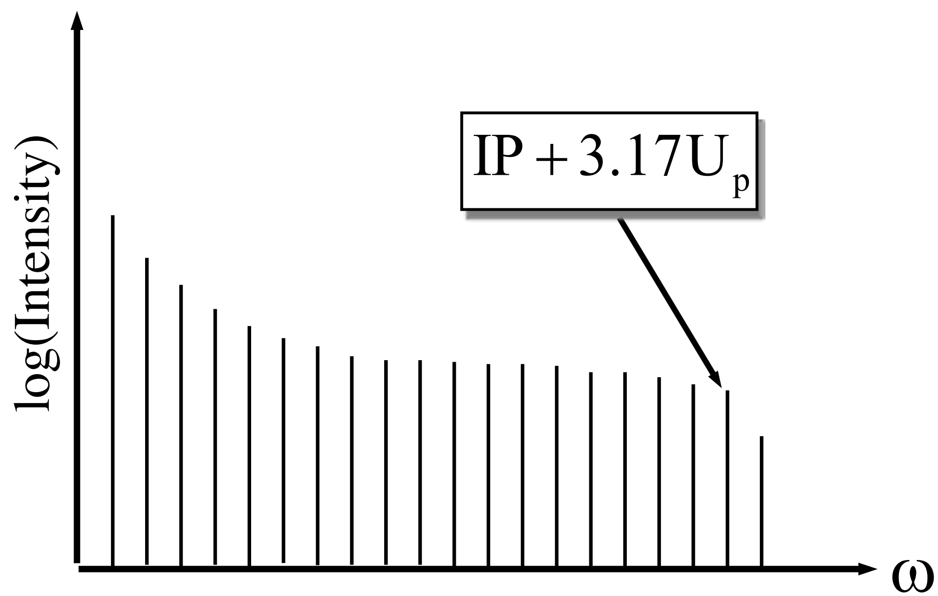
Important: Control target must be formulated in terms of the density!

Optimal Control of Harmonic Generation (example: Helium Atom)





Enhancement of a single harmonic peak

Harmonic Spectrum:

$$H(\omega) = \left| \int dt e^{i\omega t} \frac{d^2}{dt^2} \left\{ \int d^3 r z \rho(\vec{r}, t) \right\} \right|^2$$

Maximize:

$$F = \sum_k \alpha_k \max_{\omega \in [k\omega_0 - \beta, k\omega_0 + \beta]} H(\omega)$$

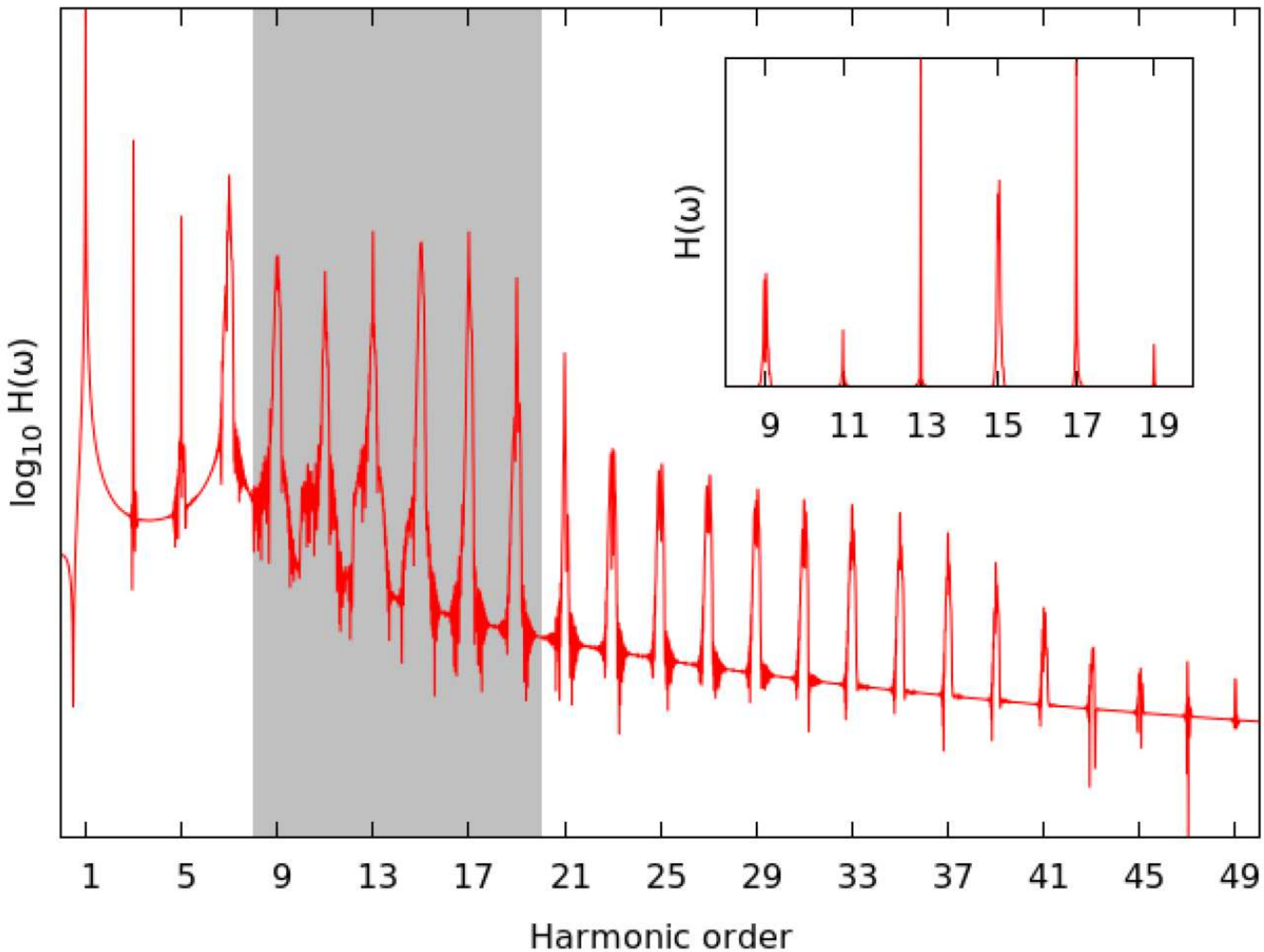
To maximize, e.g., the 7th harmonic of ω_0 , choose coefficients as
$\alpha_7 = 4, \quad \alpha_3 = \alpha_5 = \alpha_9 = \alpha_{11} = -1$

<u>Measure of enhancement</u>: Compare with reference pulse:

$$\varepsilon_{ref}(t) = \varepsilon_0 \cos\left(\frac{\pi}{2}\frac{2t - T}{T}\right)\cos(\omega t)$$

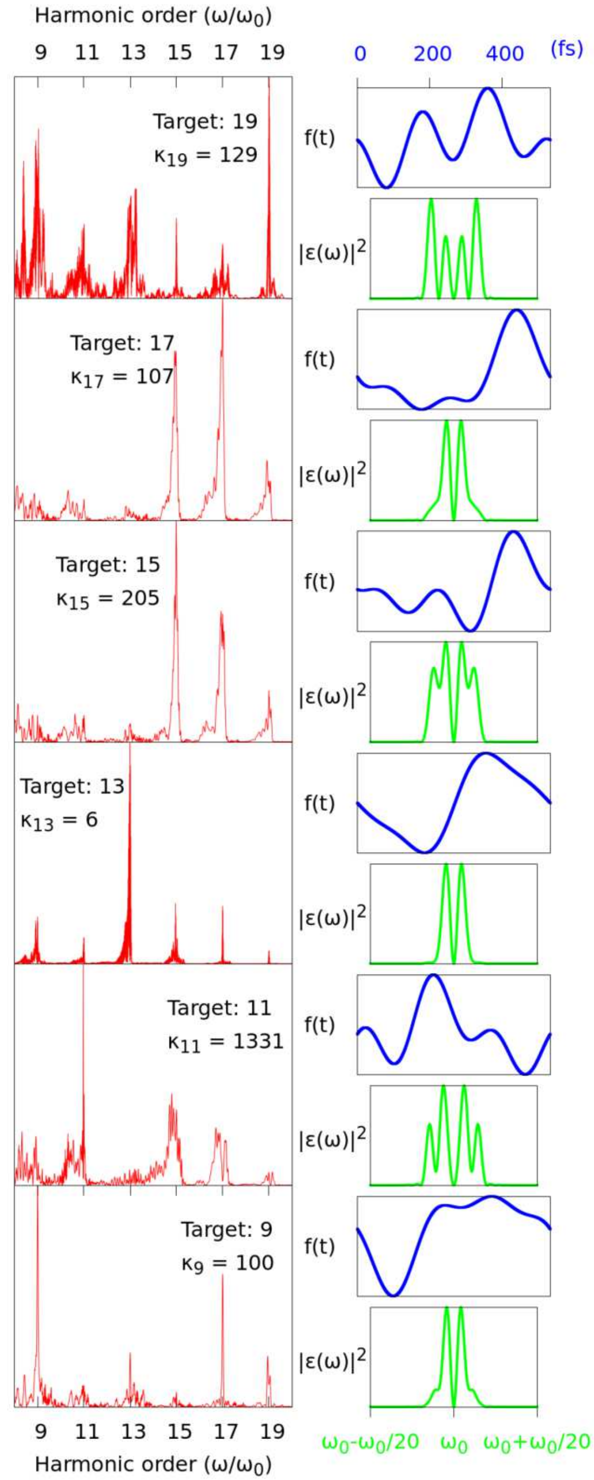
$$\kappa_j = \frac{\max_{\omega \in [j\omega_0 - \beta, j\omega_0 + \beta]} H(\omega)}{H_{ref}(j\omega_0)}$$

Harmonic spectrum of reference pulse for hydrogen atom



Results for Hydrogen atom

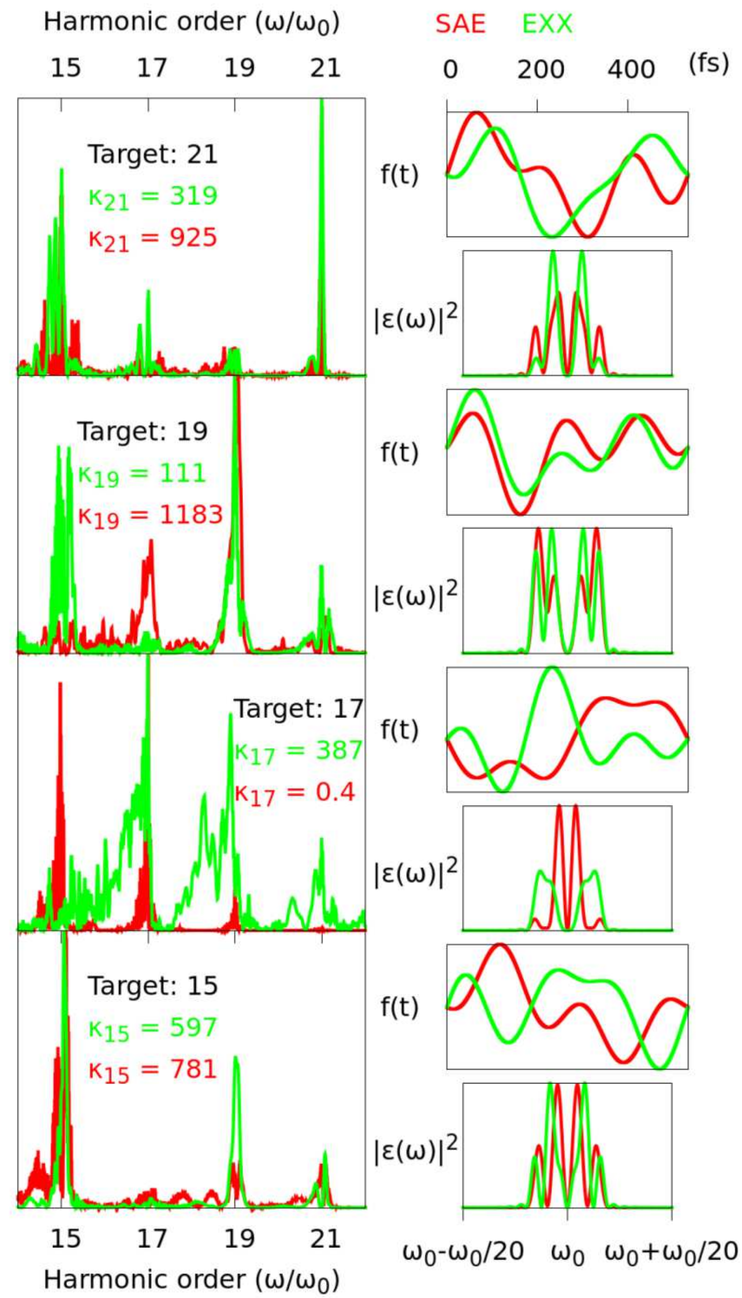
A. Castro, A. Rubio, E.K.U.Gross, Eur. Phys. J. B 88, 191 (2015).

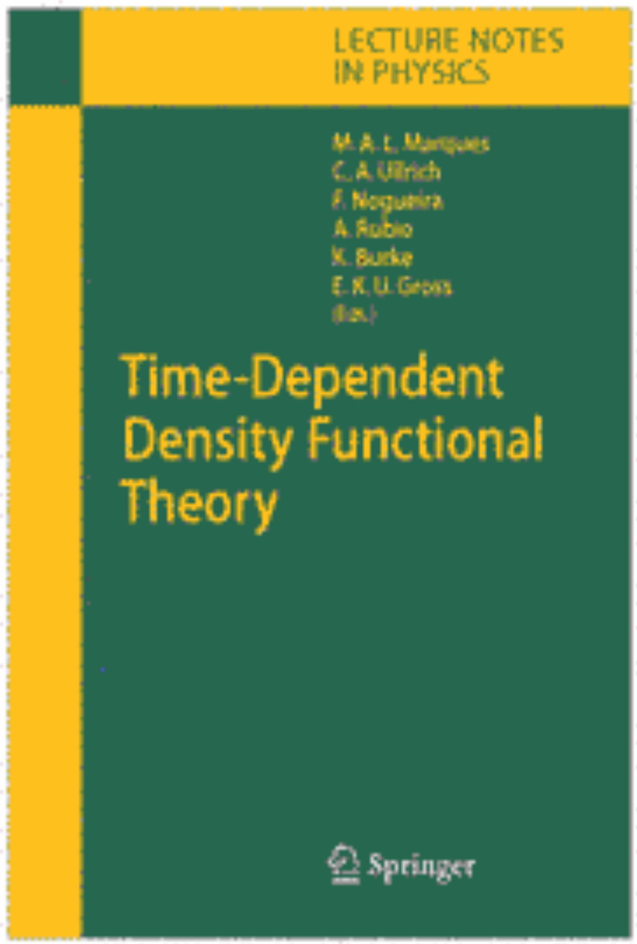


Results for Helium atom

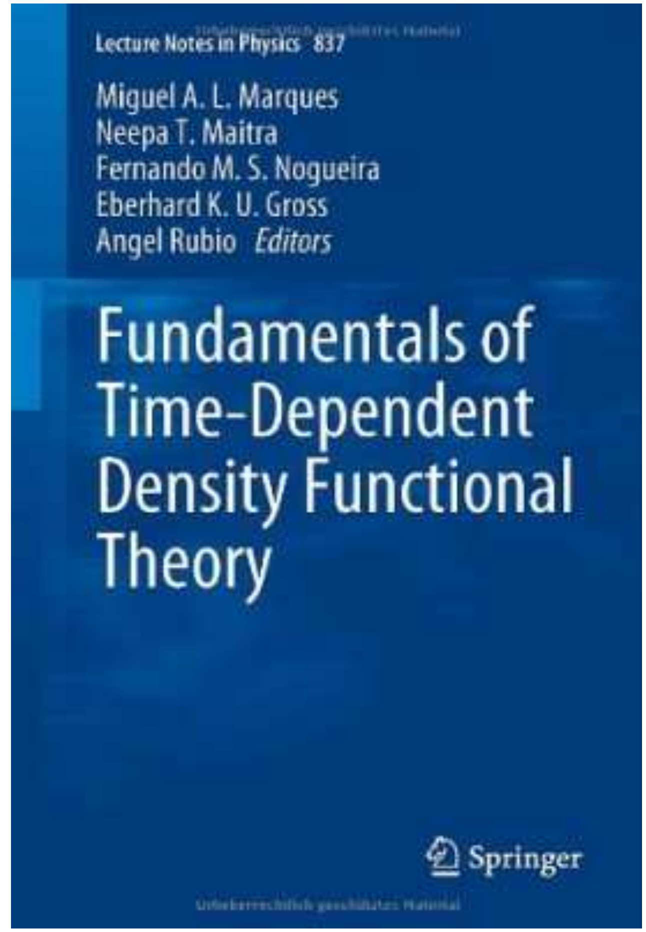
(Using TDDFT with EXX functional)

A. Castro, A. Rubio, E.K.U.Gross, Eur. Phys. J. B 88, 191 (2015).





Lecture Notes in Physics <u>706</u>
(Springer, 2006)



Lecture Notes in Physics <u>837</u>
(Springer, 2012)