Laser induced inter-site spin transfer

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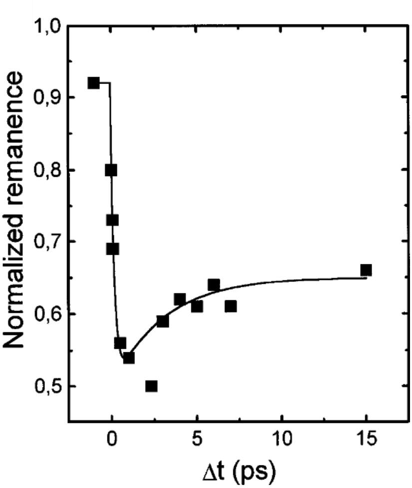
Femto-magnetism



Laser-induced **demagnetization**

Expt: Beaurepair et al. PRL 76 4250 (1996)

Ultrafast manipulation of spins with light

- Efficient devices based in spin currents
- Fast (femtoseconds) operational times

Exchange interaction time scales in femto-seconds

Metal	Exchange
Fe	52
Co	80
Ni	380

Table: Exchange interaction time is Heisenberg exchange parameter converted to time.

Exchange and spin-orbit times in femto-seconds

Metal	Exchange	Spin-orbit
Fe	52	50
Co	80	52
Ni	380	48

Table: Exchange time is Heisenberg exchange parameter converted to time. SO time is the time at which SO coupling causes spin flips in Fe/Mn, Co/Mn and Ni/Mn interfaces.

Demonstrate

1. Spin manipulation faster than spin-orbit.
2. Spin manipulation at sub-exchange time scales.
3. Sub-exchange, sub-spin-orbit switching the magnetic order.
4. Spin manipulation times entirely controlled by laser pulse.
5. Experimental demonstration that spins can be manipulated at sub-exchange time scales.

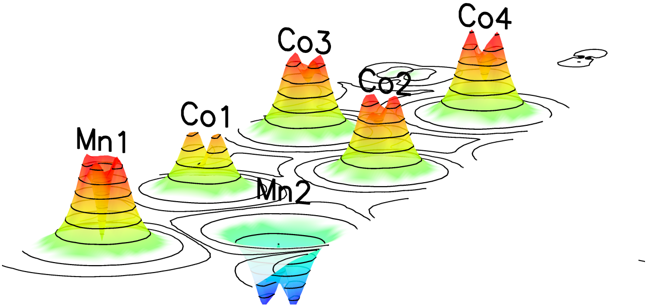
Theory

$$
\begin{aligned}
i\frac{\partial\psi_j(\mathbf{r},t)}{\partial t} &= \left[\frac{1}{2}\left(i\nabla+\frac{1}{c}\mathbf{A}_{\text{ext}}(t)\right)^2+v_s(\mathbf{r},t)\right. \qquad (1)\\
&+ \left.\frac{1}{2c}\overrightarrow{\sigma}\cdot\mathbf{B}_s(\mathbf{r},t)+\frac{1}{4c^2}\overrightarrow{\sigma}\cdot\left(\nabla v_s(\mathbf{r},t)\times i\nabla\right)\right]\psi_j(\mathbf{r},t)
\end{aligned}
$$

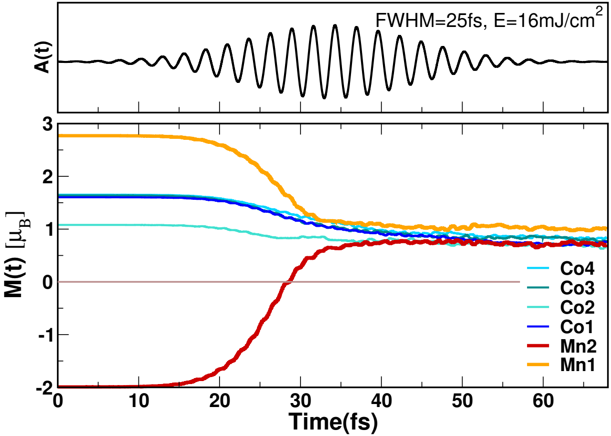
E. Runge and E. K. U. Gross, Phys. Rev. Lett. **52**, 997 (1984)

ab-initio: "from the beginning". A calculation is said to be *ab-initio* (or "from first principles") if it relies on basic and established laws of nature without additional assumptions or special models.

Ferri-magnetic layers: Mn2/Co4/Cu(001)



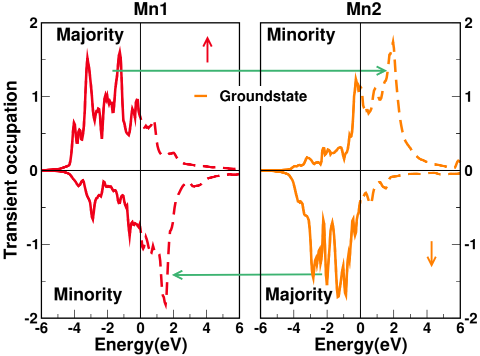
Magnetization dynamics in Mn2/Co4/Cu(001)



- Ground-state is Ferri-magnetic
- ∼ 29fs one of the Mn layers switches the direction of spin
- Stays in transient FM state at least till 150fs

Dewhurst, Elliott, Shallcross, Gross and Sharma, Nano Lett. **18**, 1842, (2018)

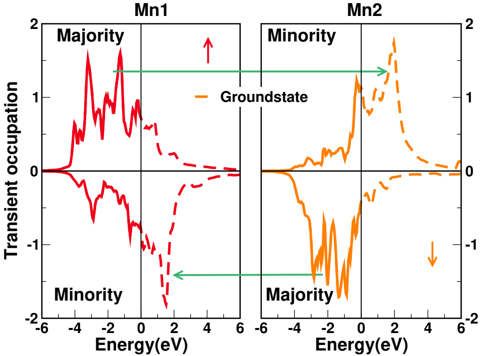
Transient density of states

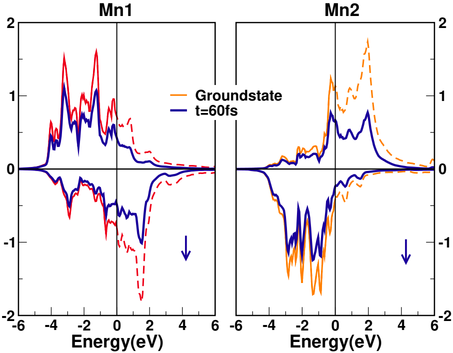


$$M = n_{\mathrm{maj}} - n_{\mathrm{min}}$$

- Optical inter-site spin transfer (OISTR) is responsible for switching
- Availability of states enforces OISTR

Transient density of states





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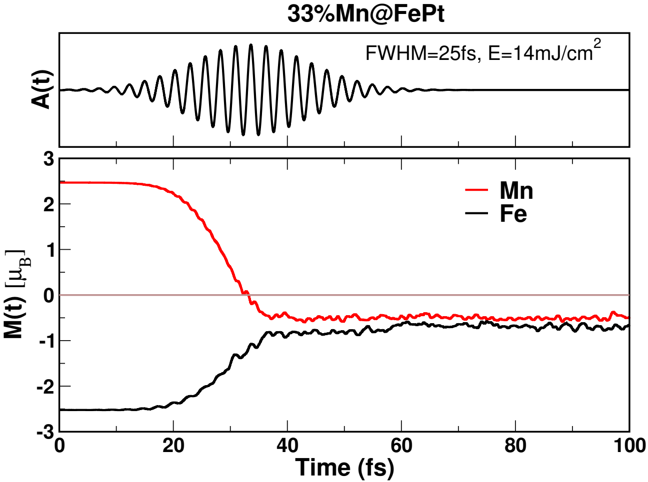
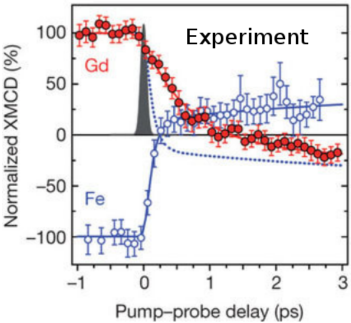
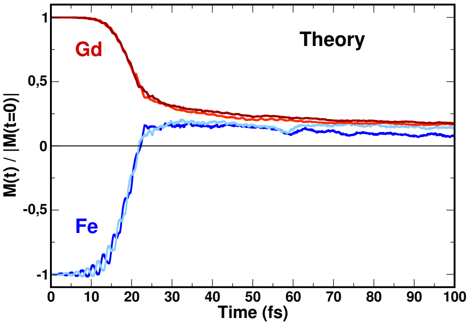
Ferri-magnetic bulk: FeMnPt



- OISTR leads to switching of order in bulk materials

Optical switching in GdFeCo– Theory





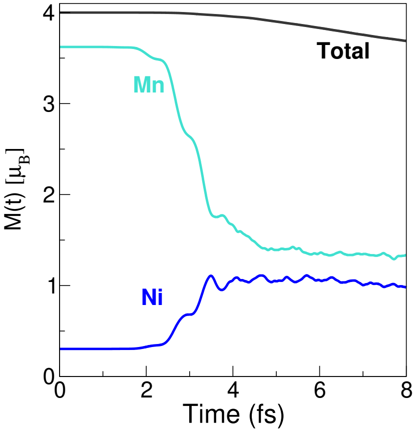
- Coupling goes from AFM to FM.
- Fe switches direction of magnetization faster.
- FM coupling between Fe and Gd persists.

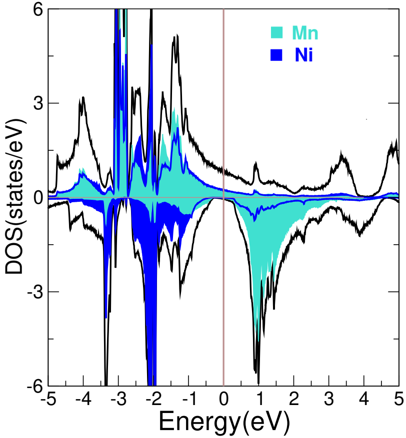
Ferro-magnets

Atom	Moment (μ_B)
Mn	3.62
Ni	0.30
Sb	0.04

Table: Local ground-state moments in half-heusler NiMnSb

Ferro-magnetic bulk: NiMnSb

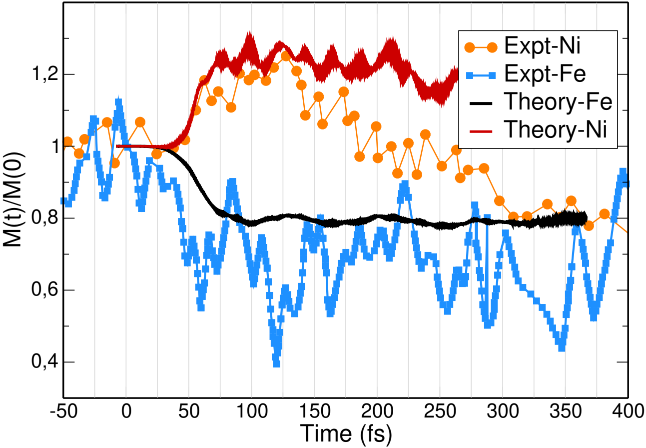




Elliott et al. Sci. Rep. 6 38911 (2016)

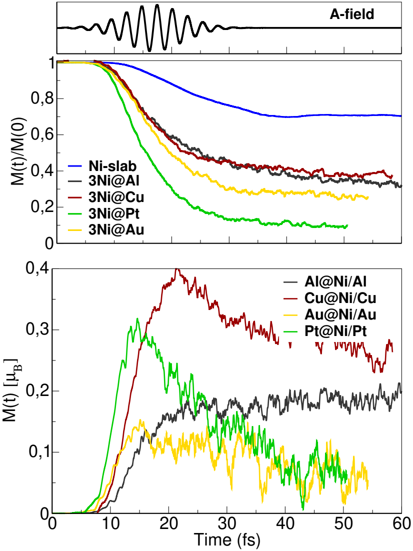
Kreiger et al. JPCM 29, 224001 (2017)

OISTR demonstrated in bulk: $Fe_{50}Ni_{50}$



Moritz Hofherr and Martin Aeschilmann (Kaiserslautern)

Ferro-magnetic layers: Ni@ Pt, Au, Al or Cu (001)

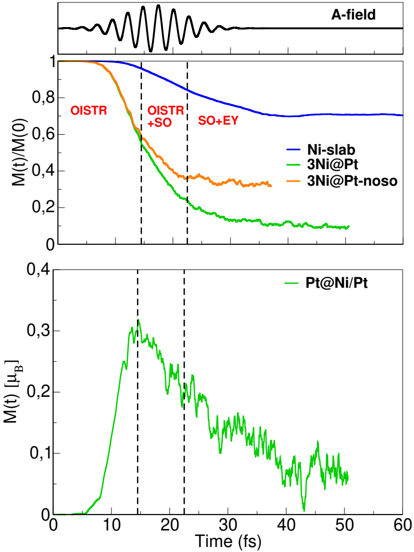


Dewhurst et al. Phys. Rev. Appl. (2018)

- OISTR leads to early-time demagnetization is multi-layers.
- In single sub-lattice bulk this is missing.
- There are 3 distinct time scales in the demagnetization process:

  1. OISTR
  2. OISTR+SO
  3. SO+Elliott-Yafet

Ferro-magnetic layers: Ni@ Pt, Au, Al or Cu (001)



Dewhurst et al. Phys. Rev. Appl. (2018)

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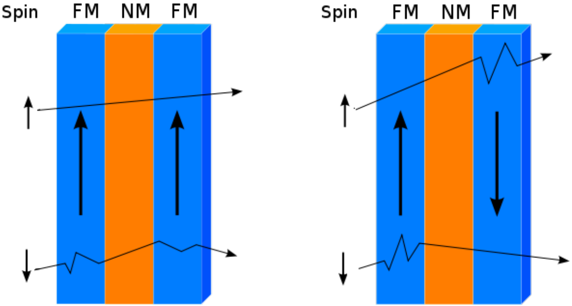
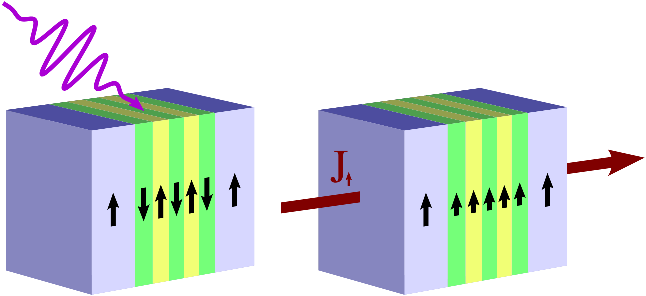
Outlook for OISTR: Ultrafast GMR

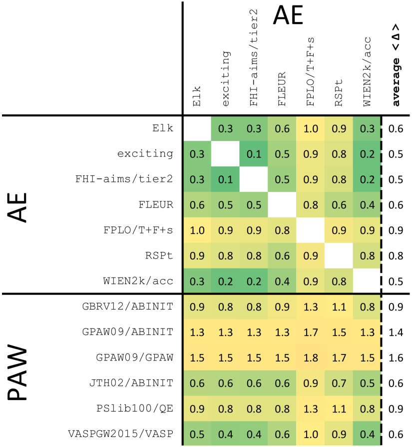


Laser induced transient GMR device



Elk code: full potential LAPW method

Science 351, 6280 (2016)



		AE							
		Elk	exciting	FHI-aims/tier2	FLEUR	FPLO/T+F+s	RSPt	WIEN2k/acc	**average <Δ>**
AE	Elk		0.3	0.3	0.6	1.0	0.9	0.3	0.6
	exciting	0.3		0.1	0.5	0.9	0.8	0.2	0.5
	FHI-aims/tier2	0.3	0.1		0.5	0.9	0.8	0.2	0.5
	FLEUR	0.6	0.5	0.5		0.8	0.6	0.4	0.6
	FPLO/T+F+s	1.0	0.9	0.9	0.8		0.9	0.9	0.9
	RSPt	0.9	0.8	0.8	0.6	0.9		0.8	0.8
	WIEN2k/acc	0.3	0.2	0.2	0.4	0.9	0.8		0.5
PAW	GBRV12/ABINIT	0.9	0.8	0.8	0.9	1.3	1.1	0.8	0.9
	GPAW09/ABINIT	1.3	1.3	1.3	1.3	1.7	1.5	1.3	1.4
	GPAW09/GPAW	1.5	1.5	1.5	1.5	1.8	1.7	1.5	1.6
	JTH02/ABINIT	0.6	0.6	0.6	0.6	0.9	0.7	0.5	0.6
	PSlib100/QE	0.9	0.8	0.8	0.8	1.3	1.1	0.8	0.9
	VASPGW2015/VASP	0.5	0.4	0.4	0.6	1.0	0.9	0.4	0.6

Gold standard for electronic structure of solids. Features include:

- Ground state
- Most single particle observables
- Structural optimization
- Many-body methods: GW and beyond, RDMFT, BSE ...
- Response functions: magnons, phonons, plasmons, excitons ...
- Wannier90 interface
- Tensor moments
- Non-equilibrium spin dynamics
- Superconductivity: calculation of Tc, Eliashberg

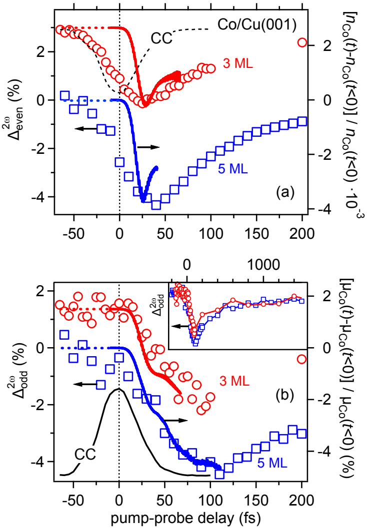
J. K. Dewhurst, S. Sharma, L. Nordström and E. K. U. Gross ....

http://elk.sourceforge.net/

Summary

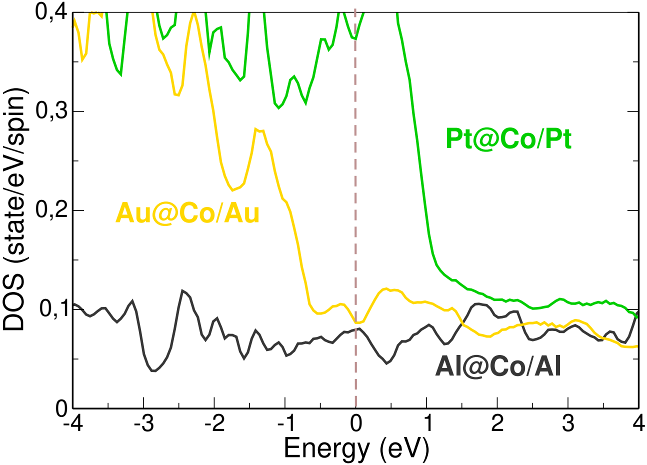
- In multi-sub-lattice systems OISTR is the first mechanism that causes spin-dynamics
- In ferro-magnets moment on one sub-lattice increases as the other decreases.
- In ferri-magnets long range magnetic order switches and a transient ferro-magmetic state appears.
- The time of this physics is decided by FWHM of the pump-pulse.
- Three distinct time scales in early time spin-dynamics identified.

Co@Cu

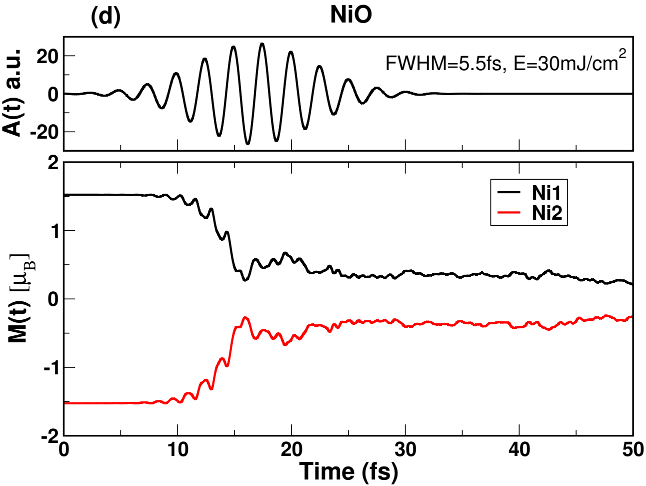


SHG expt: Andrea Eschenlohr and Uwe Bovensiepen (Duisburg)

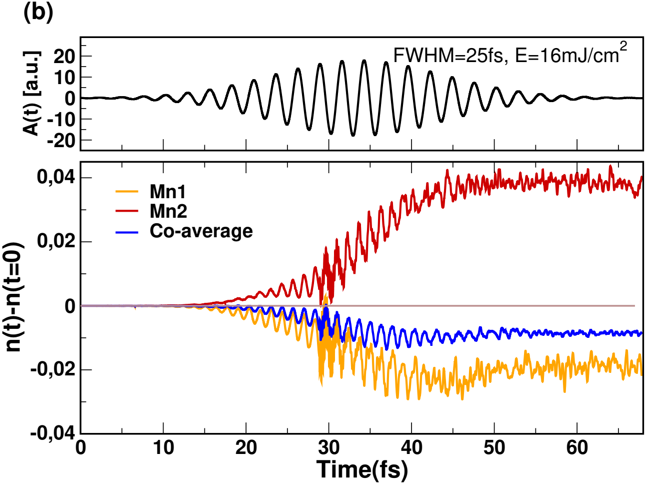
DOS for Ni@ Pt, Au and Al



What about compensated AFM?



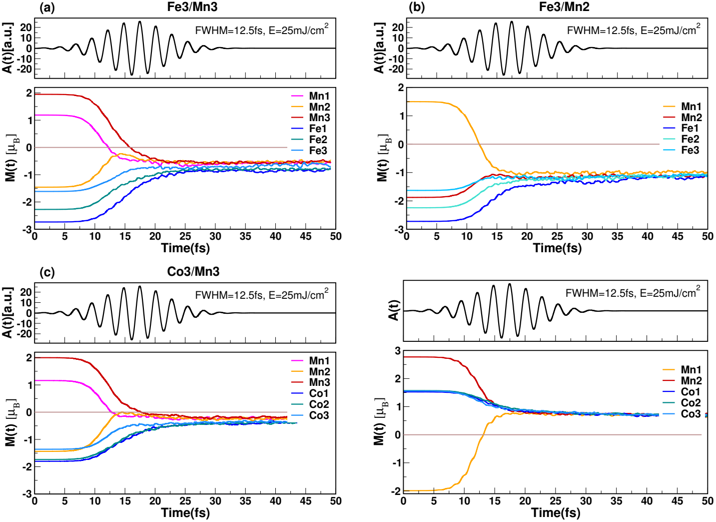
Does a dipole moment develop?



$$N = n_{\text{majority}} + n_{\text{minority}}$$

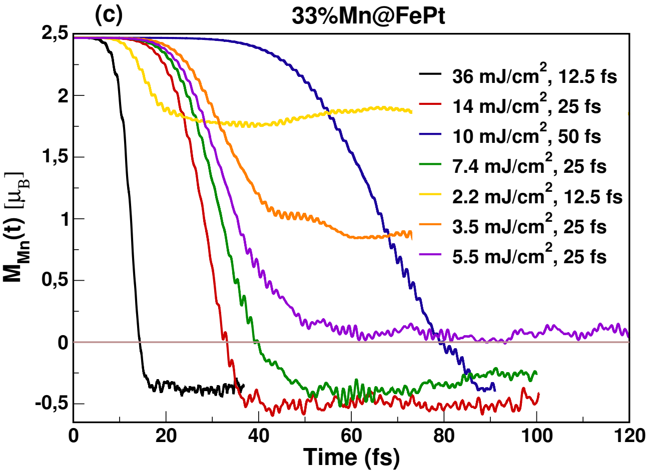
- Change in total charge very small despite large spin-selective charge transfer

Is it generic?



- Effect faster for shorter pulses.
- Effect is universally applicable to ferri-magnets.

Tunable via laser pulses

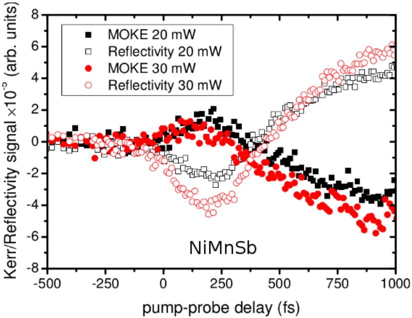


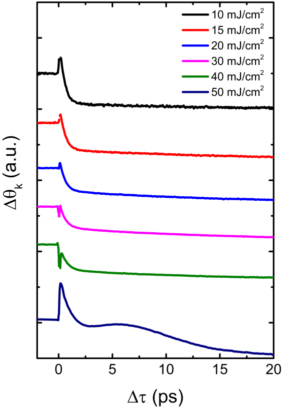
- Switching controllable by pulse tuning.

Dewhurst, Elliott, Shallcross, Gross and Sharma, Nano Lett. (2018)

http://dx.doi.org/10.1021/acs.nanolett.7b05118

Experimental confirmation for ferro-magnets

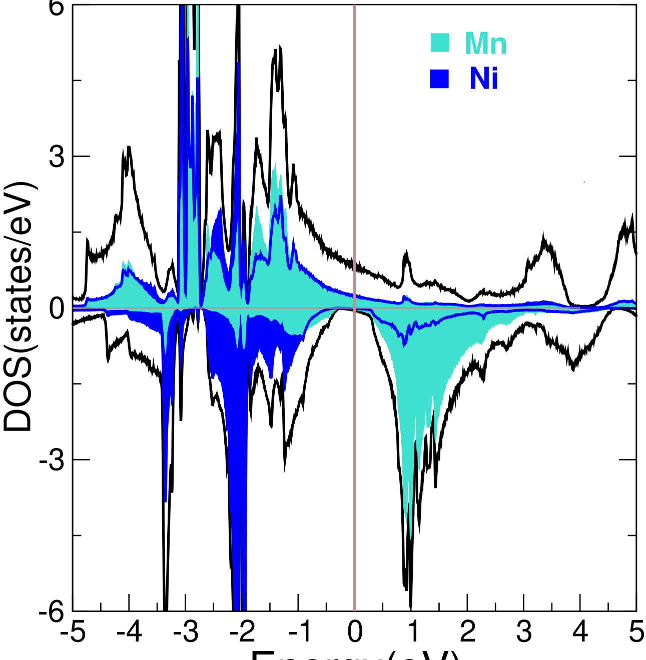




NiMnSb: D. Steil

CoMnSb: Mann and Münzenberg

Density of states: NiMnSb



Simplest (and first) exchange-correlation functional is the local density approximation (LDA)

$$E_{\rm xc}^{\rm LDA}[\rho] = \int d^3r \, \varepsilon_{\rm xc}\left(\rho(\mathbf{r})\right) \rho(\mathbf{r})$$

where $\varepsilon_{\rm xc}$ is the exchange-correlation energy density of the <u>homogeneous electron gas</u>

Easy to take a functional derivative

$$\begin{aligned} v_{\rm xc}^{\rm LDA}(\mathbf{r}) &= \frac{\delta E_{\rm xc}^{\rm LDA}[\rho]}{\delta \rho(\mathbf{r})} \\ &= \varepsilon_{\rm xc}'\left(\rho(\mathbf{r})\right) \rho(\mathbf{r}) + \varepsilon_{\rm xc}\left(\rho(\mathbf{r})\right) \end{aligned}$$

The exchange energy density of the homogeneous electron gas (HEG) is (relatively) easy to calculate

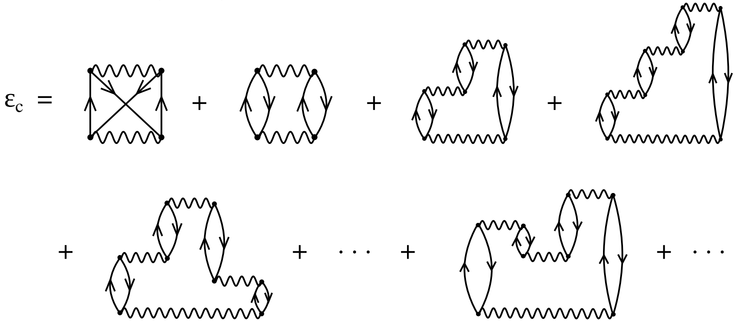
$$\varepsilon_{\mathrm{x}}(r_s) = -\frac{3e^2}{4\pi}\left(\frac{9\pi}{4}\right)^{1/3}\frac{1}{r_s}$$

where

$$r_s = \left(\frac{3}{4\pi\rho}\right)^{1/3}$$

Calculating the correlation energy density of the HEG is much more difficult

High density limit: M. Gell-Mann and K. A. Brueckner *Phys. Rev.* **106**, 364 (1957)



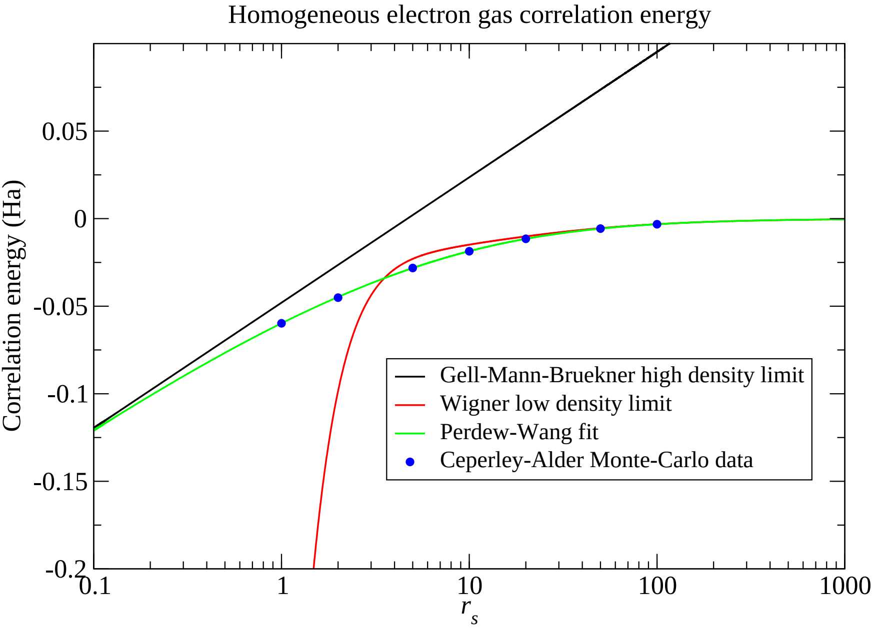
$$\varepsilon_{\mathrm{c}}(r_s) = A + B \ln r_s + \cdots$$

Low density limit: bcc Wigner lattice

$$\varepsilon_{\mathrm{c}}(r_s) = \frac{a_1}{r_s} + \frac{a_2}{r_s^{3/2}} + \frac{a_3}{r_s^2} + \cdots$$

The Perdew-Wang fit interpolates between these extremes
[*Phys. Rev. B* **45**, 13244 (1992)]

$$\varepsilon_{\mathrm{c}}(r_s) = -2A(1+\alpha_1 r_s) \ln \left[1 + \frac{1}{2A(\beta_1 r_s^{1/2} + \beta_2 r_s + \beta_3 r_s^{3/2} + \beta_4 r_s^{P+1})} \right]$$


Homogeneous electron gas correlation energy
Correlation energy (Ha)
0.05
0
-0.05
-0.1
-0.15
-0.2
0.1
1
10
100
1000
r_s
Gell-Mann-Bruekner high density limit
Wigner low density limit
Perdew-Wang fit
Ceperley-Alder Monte-Carlo data