ULTRAFAST SPIN, CHARGE AND NUCLEAR DYNAMICS

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Ultrafast spin, charge and nuclear dynamics

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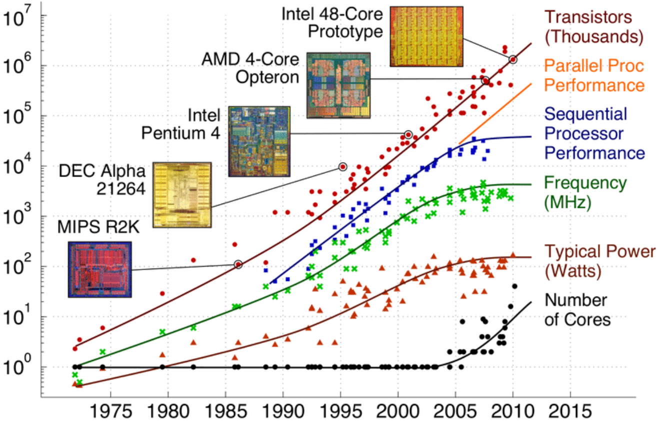
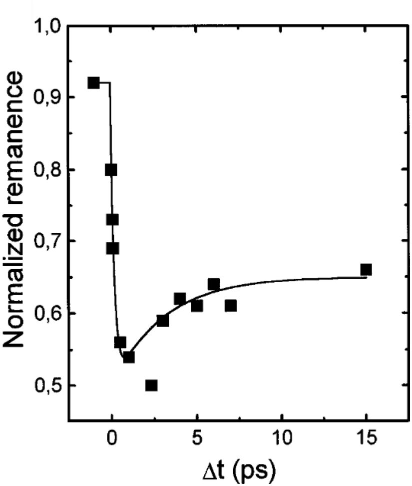
Funding: DFG– SFB (TRR227), SPP(1840), Normalverfahren, Eigenestelle

Evolution of multi-core processors



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

Leading order terms in expansion of QED H

$$
\begin{aligned}
H = {} & \sum_i \frac{1}{2m}\mathbf{p}_i^2 - eV_{\mathrm{ext}}(\mathbf{r}_i) + \sum_{i<j} \frac{e^2}{r_{ij}} + \frac{1}{c}\sum_i \frac{e}{m}\mathbf{p}_i \cdot \mathbf{A}_{\mathrm{ext}}(\mathbf{r}_i) \\
& + \frac{1}{c}\sum_i \frac{e}{m}\mathbf{s}_i \cdot \mathbf{B}_{\mathrm{ext}}(\mathbf{r}_i) + \frac{1}{c^2}\sum_i \frac{e}{2m^2}\mathbf{s}_i \cdot [\mathbf{E}_{\mathrm{ext}}(\mathbf{r}_i) \times \mathbf{p}_i] \\
& + \frac{1}{c^2}\sum_i \frac{e^2}{2m}\mathbf{A}_{\mathrm{ext}}^2(\mathbf{r}_i) - \frac{1}{c^2}\sum_i \frac{1}{8m^3}\mathbf{p}_i^4 + \frac{ie}{4m^2c^2}\mathbf{p}_i \cdot \mathbf{E}_{\mathrm{ext}}(\mathbf{r}_i) \\
& - \frac{1}{c^2}\frac{e^2}{2m^2}\sum_{i<j}\mathbf{p}_i \cdot \left[\frac{(\mathbf{r}_i - \mathbf{r}_j)(\mathbf{r}_i - \mathbf{r}_j)}{r_{ij}^3} + \frac{1}{r_{ij}}\right] \cdot \mathbf{p}_j \\
& - \frac{1}{c^2}\frac{e^2}{m^2}\sum_{i<j}\mathbf{s}_i \cdot \left[\frac{3(\mathbf{r}_i - \mathbf{r}_j)(\mathbf{r}_i - \mathbf{r}_j)}{r_{ij}^5} - \frac{1}{r_{ij}^3}\right] \cdot \mathbf{s}_j \\
& - \frac{1}{c^2}\frac{e^2}{m^2}\sum_{i \neq j}\frac{1}{r_{ij}^3}\left(\mathbf{s}_i \cdot [(\mathbf{r}_j - \mathbf{r}_i) \times \mathbf{p}_j] + \frac{1}{2}\mathbf{s}_i \cdot [(\mathbf{r}_i - \mathbf{r}_j) \times \mathbf{p}_i]\right) \\
& + \ldots
\end{aligned}
$$

Still only an approximation to QED...

Theory: fully ab-initio approach

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

Theory: fully ab-initio approach

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

Effective potentials (we use ALDA):

$$\mathbf{B}_s[\rho,\mathbf{m}] = \mathbf{B}_{\mathrm{ext}} + \mathbf{B}_{\mathrm{xc}}[\rho,\mathbf{m}], \qquad v_s[\rho,\mathbf{m}] = v_{\mathrm{cl}} + v_{\mathrm{xc}}[\rho,\mathbf{m}] \quad (2)$$

Theory: fully ab-initio approach

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

Effective potentials (we use ALDA):

$$
\mathbf{B}_s[\rho,\mathbf{m}] = \mathbf{B}_{\rm ext} + \mathbf{B}_{\rm xc}[\rho,\mathbf{m}], \qquad v_s[\rho,\mathbf{m}] = v_{\rm cl} + v_{\rm xc}[\rho,\mathbf{m}] \tag{2}
$$

Density:

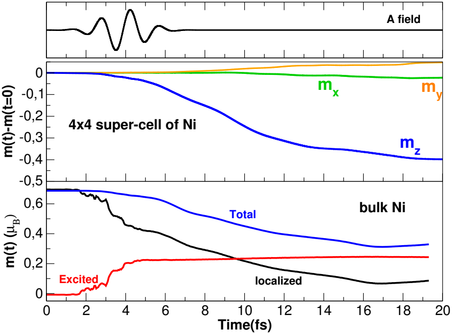
$$
\rho(\mathbf{r},t) = \sum_j \psi_j^*(\mathbf{r},t)\psi_j(\mathbf{r},t) \tag{3}
$$

Magnetization density(unconstrained vector field):

$$
\mathbf{m}(\mathbf{r},t) = \sum_j \psi_j^*(\mathbf{r},t)\overrightarrow{\sigma}\psi_j^*(\mathbf{r},t) \tag{4}
$$

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

Demagnetisation in elemental magnet : Ni



- Inter-site non-collinearity and magnons do not contribute in early times
- $\frac{1}{4c^2}\overrightarrow{\sigma} \cdot (\nabla v_s(\mathbf{r},t) \times i\nabla)$
- The physics is dominated by spin-orbit induced spin-flips

Krieger et al. JCTC **11**, 4870 (2015),
Elliott et al. New J. Phys. **18**, 013014 (2016),

Dewhurst et al. Computer Phys. Comm. **209**, 92 (2016)

Nuclear degrees of freedom

Treat nuclei as classical point particles: Ehrenfest dynamics

Quantum nuclei required for superconductivity!

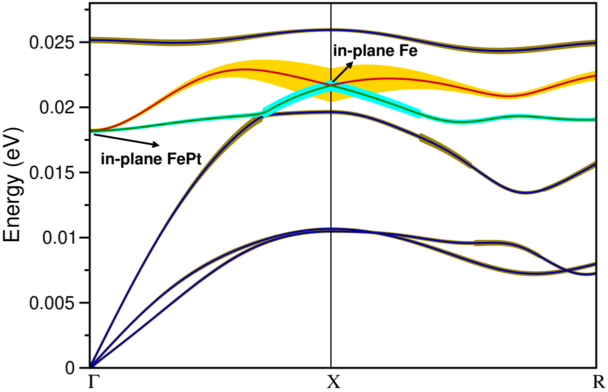
phono- magnetism

$$\begin{aligned}\hat{H}_s &= \frac{1}{2}\left(i\nabla + \frac{1}{c}\mathbf{A}_{\mathrm{ext}}(t)\right)^2 + v_s(\mathbf{r},t) + \frac{1}{2c}\overrightarrow{\sigma}\cdot\mathbf{B}_{\mathrm{xc}}(\mathbf{r},t)\\ &+ \frac{1}{4c^2}\overrightarrow{\sigma}(\nabla v_s(\mathbf{r},t)\times i\nabla),\end{aligned}$$

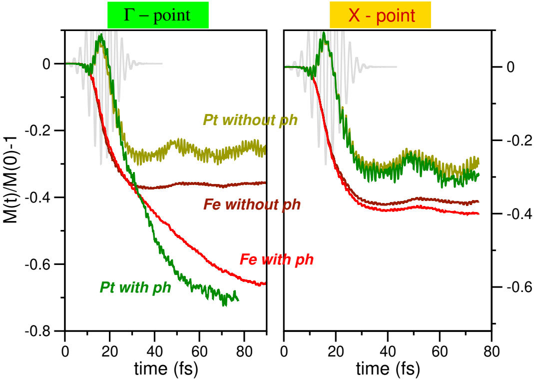
$$v_s(\mathbf{r},t) \longrightarrow v_s(\mathbf{r},t) - \sum_{p\alpha}\frac{\partial v_{\mathrm{cl}}(\mathbf{r})}{\partial u_\alpha^p}\delta u_\alpha^p(t), \tag{5}$$

$$F_{p\alpha}(t) = -\omega\delta u_\alpha^p(t). \tag{6}$$

Phonons and electron-phonon coupling: FePt



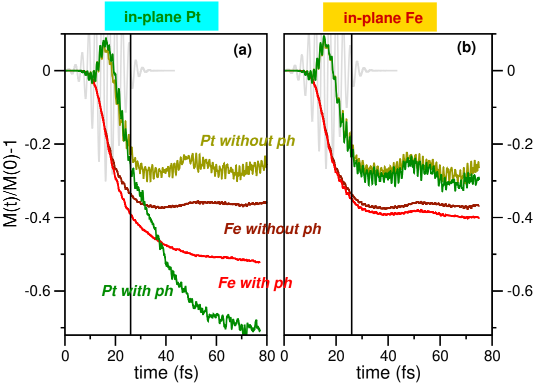
Femto- phono- magnetism: FePt



- EPC is not necessarily a good guide.
- Magnetization dynamics due to non-adiabatic coupling.

Sharma et al. arxiv/2203.14234 (2022)

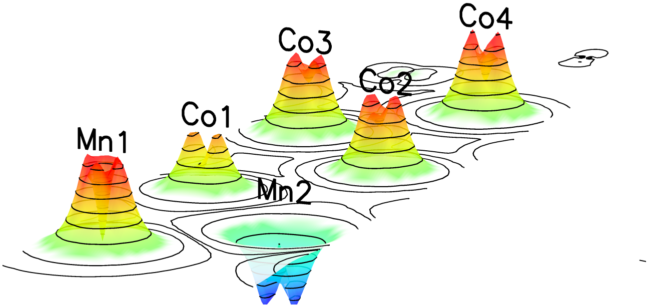
Femto- phono- magnetism: FePt



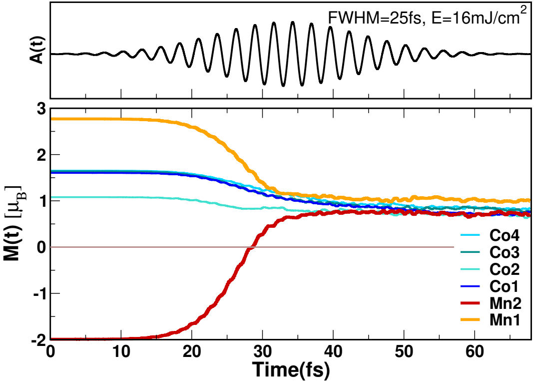
- Early time physics is the same.
- Phenomena of OISTR domiates in early times.

Sharma et al. arxiv/2203.14234 (2022)

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



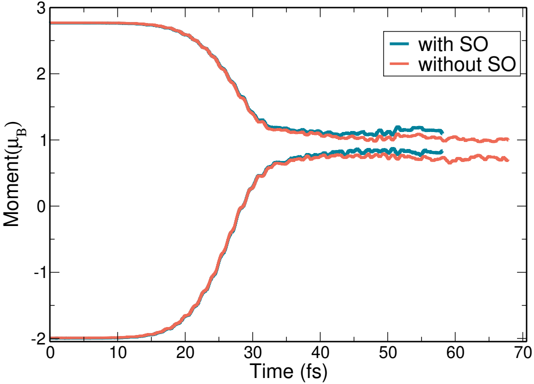
Magnetization dynamics in Mn2/Co4/Cu(001)



- 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
- Saturation is due to missing nuclear dynamics .

Dewhurst et al. Nano Lett. **18**, 1842, (2018)

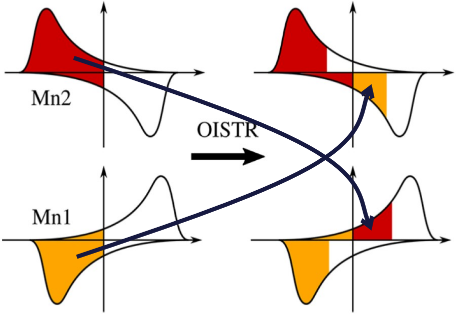
Magnetization dynamics without SOC

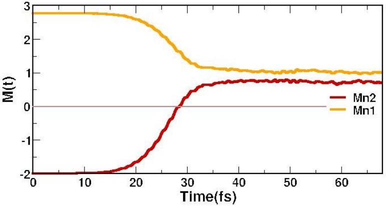


- Spin-flips and spin-orbit play no role in switching of magnetic order.

Dewhurst et al. Nano Lett. **18**, 1842, (2018)

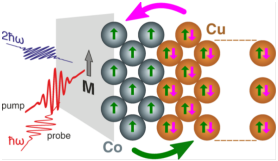
Optical inter-site spin transfer (OISTR)



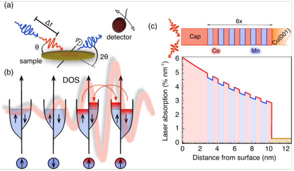


$$m = n_{\mathrm{up}} - n_{\mathrm{dn}}$$

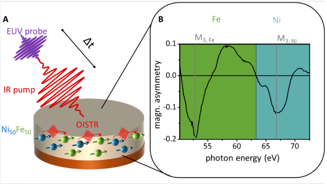
Experimental confirmation of OISTR



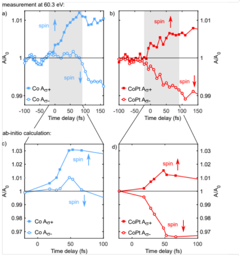
Chen *et al.*, Phys. Rev. Lett.
122, 067202 (2019)

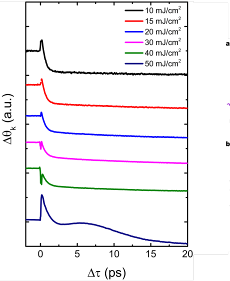


Golias et al., Phys. Rev. Lett.
126, 107202 (2021)



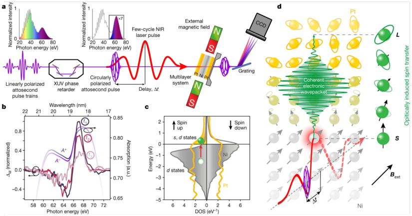
Hofherr *et al.* Science Advs.
6, eaay8717 (2019)





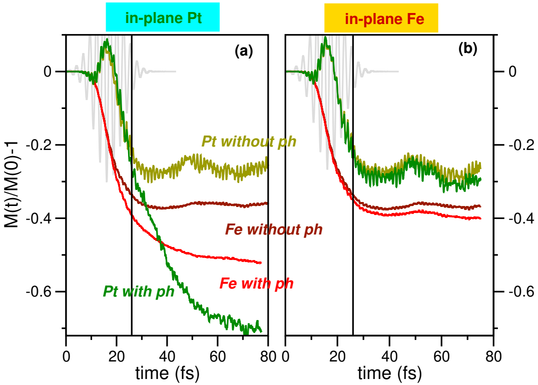
F. Willems *et al.*, Nature Comm.
11, 1 (2020)

Steil *et al.*, Phys. Rev. Res.
2, 023199 (2020)



Siegrist *et al.* Nature
571, 240 (2019)

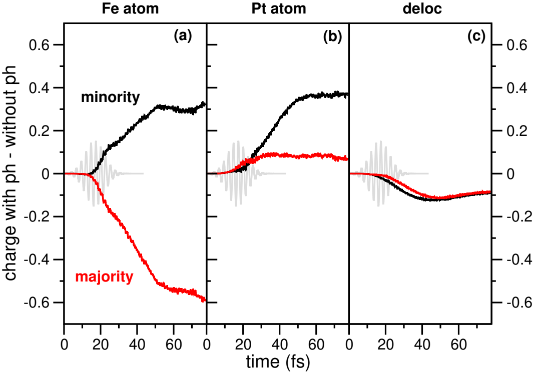
Femto- phono- magnetism: FePt



- Early time physics is the same.
- Phenomena of OISTR domiates in early times.

Sharma et al. arxiv/2203.14234 (2022)

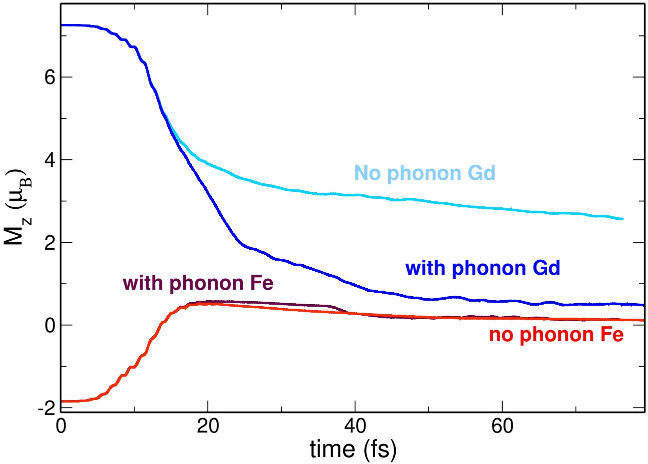
Spin currents due to phonons



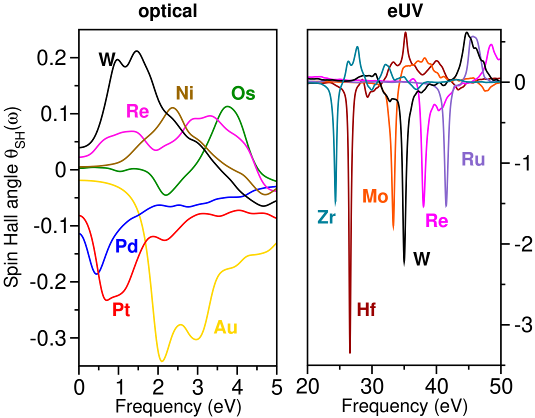
- Minority spin current flow from Fe to Pt (this demagnetizes Pt).
- Increase in minority states on Fe atoms leads to enhanced spin-flips (loss in Fe moment).

Sharma et al. arxiv/2203.14234 (2022)

Faster switching with phono- magnetism: GdFe



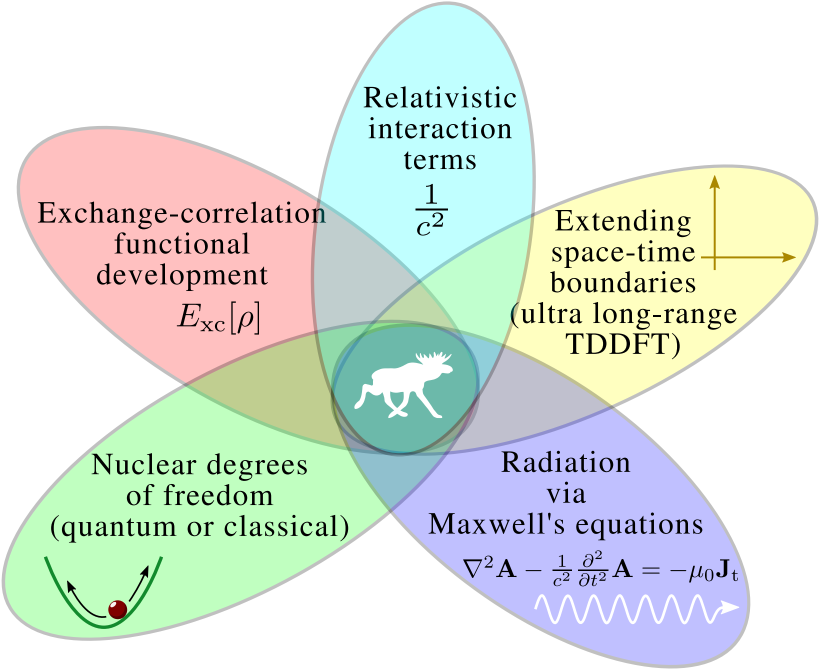
Giant optical spin-Hall angles



- $\theta^{\mathrm{SH}}(\omega) = \frac{\mathbf{j}^S_{xy}(\omega)}{\mathbf{j}_z(\omega)}$: finite frequency extension of SHA leads to a giant effect.
- In eUV range the SHA is larger than 1.

Elliott et al. arxiv/2204.01538 (2022)

What is missing from the theory?


Relativistic interaction terms
$\frac{1}{c^2}$
Exchange-correlation functional development
$E_{\mathrm{xc}}[\rho]$
Extending space-time boundaries (ultra long-range TDDFT)
Nuclear degrees of freedom (quantum or classical)
Radiation via Maxwell's equations
$\nabla^2\mathbf{A} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{A} = -\mu_0\mathbf{J}_{\mathrm{t}}$

Various combinations yield new predictiveness

$$\text{long-range} + \text{Maxwell's equations} + \text{TDDFT} \implies \begin{cases} \text{light propagation} \\ \text{density wave propagation} \end{cases}$$

long-range + spin-orbit + spin-TDDFT $\implies$ skyrmion dynamics

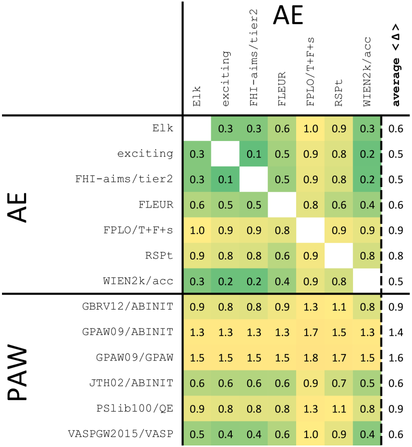
long-range + dipole-dipole interaction + spin-DFT $\implies$ magnetic domains

quantum nuclei + TD-long-range + superconductivity $\implies$ TD-supercond

New physics can be found at competing energy scales!

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

- Early time demagnetization in elemental magnets is dominated to SOC.
- In multi-sub-lattice systems OISTR is the first mechanism that causes spin-dynamics.
- OISTR can be coherently controlled by lasers.
- Large SHA can be obtained for finite frequencies.
- Femto- phono- magnetism: phonons for femtosecond magnetization dynamics