Microscopic theory of spin dynamics in transition metal nanostructures and the role of spin-orbit coupling

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Plan

- Motivation
- Formalism
- Spin waves in ultrathin films
- FMR, dynamic coupling and spin pumping
- Spin excitations and SOC

Main motivation

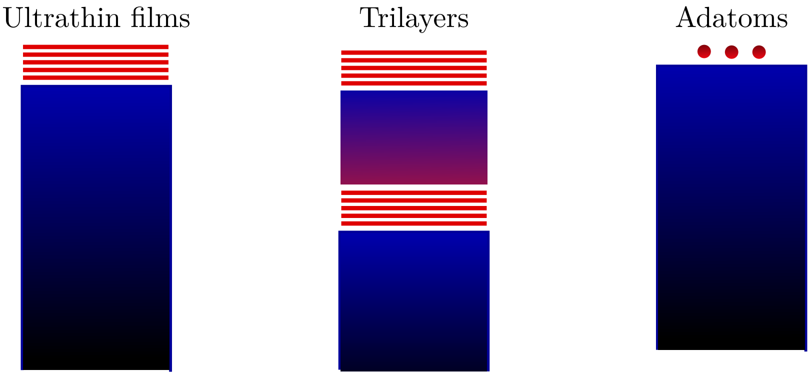
Understanding spin dynamics from the microscopic point of view

- Simple but realistic models
- Everything must come from the electronic structure
- Quantitative comparison to experimental results

Phenomena

- Intrinsic damping mechanism.
- Anisotropy gap in the spectra of spin excitations
- Dzyaloshinskii-Moriya coupling
- Anisotropic g-factors
- spin signal - charge signal interconversion*

Typical Systems



All substrates are metallic and non-magnetic.

Formalism

- Semi-empirical description of the electronic structure

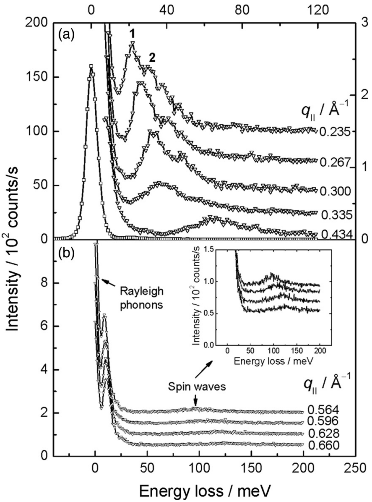
$$H = \sum_{l,l';\mu,\nu;\sigma} T_{ll'}^{\mu\nu} a_{l\mu\sigma}^{\dagger} a_{l'\nu\sigma} +$$

$$\sum_{l} \sum_{\mu,\nu,\mu'\nu} \sum_{\sigma,\sigma'} U_{l}^{\mu\nu\mu'\nu'} a_{l\mu\sigma}^{\dagger} a_{l\nu\sigma'}^{\dagger} a_{l\nu'\sigma'} a_{l\mu'\sigma}$$

- Linear response theory
- Random Phase Approximation

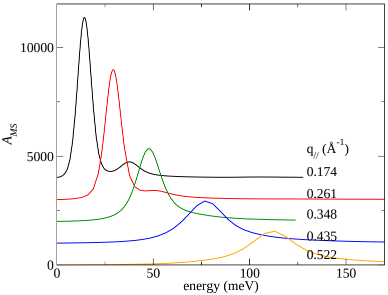
$$\chi_{ll'}(\Omega) = \chi_{ll'}^{0}(\Omega) + \sum_{m} \chi_{lm}^{0}(\Omega) U_m \chi_{ml'}(\Omega)$$

Spin wave spectra of ultrathin films - 8Co/Cu(001)



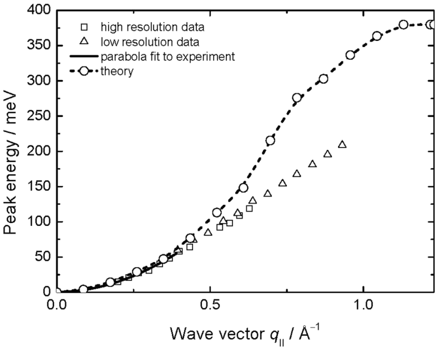
First experimental observation of optical spin wave modes on ultrathin metallic films.

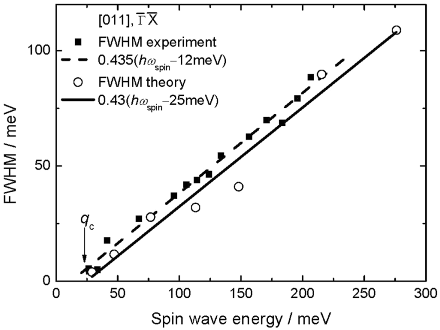
Theory



PRB **86**, 165436 (2012)

Spin waves on 8Co/Cu(001) - dispersion and linewidths





PRB **86**, 165436 (2012)

Including SOC

Within our method, including SOC corresponds to adding to the hamiltonian,

$$H_{SO} = \sum_l \sum_{\mu\nu} \frac{\lambda_l}{2} \left[L^z_{\mu\nu} (c^\dagger_{l\mu\uparrow} c_{l\nu\uparrow} - c^\dagger_{l\mu\downarrow} c_{l\nu\downarrow}) + L^+_{\mu\nu} c^\dagger_{l\mu\downarrow} c_{l\nu\uparrow} + L^-_{\mu\nu} c^\dagger_{l\mu\uparrow} c_{l\nu\downarrow} \right]$$

which is nothing but

$$\sum_l \lambda_l \vec{L}_l \cdot \vec{S}_l$$

in second-quantized form in terms of localized atomic orbitals $\{l, \mu\}$.

Including SOC

SOC couples transverse spin excitations, given by

$$\chi_{ll'}^{+-}(t) = -i\theta(t)\left\langle\left[S_l^{+}(t), S_{l'}^{-}(0)\right]\right\rangle$$

to longitudinal spin excitations and charge excitations,

$$\chi_{ll'}^{\uparrow-}(t) = -i\theta(t)\left\langle\left[n_l^{\uparrow}(t), S_{l'}^{-}(0)\right]\right\rangle$$
$$\chi_{ll'}^{\downarrow-}(t) = -i\theta(t)\left\langle\left[n_l^{\downarrow}(t), S_{l'}^{-}(0)\right]\right\rangle$$
$$\chi_{ll'}^{--}(t) = -i\theta(t)\left\langle\left[S_l^{-}(t), S_{l'}^{-}(0)\right]\right\rangle$$

Equations of motion for these **four matrices in four orbital indices** must be solved simultaneously.

PRB **82**, 014428 (2010)

Including SOC

The solution has a form that closely resembles the traditional RPA expression,

$$\vec{\chi} = \vec{\chi}^0 + (\Omega - B)^{-1}\bar{B}\vec{\chi},$$

which is solved by

$$\vec{\chi}(\Omega) = [I - (\Omega - B)^{-1}\bar{B}]^{-1}\vec{\chi}^0(\Omega),$$

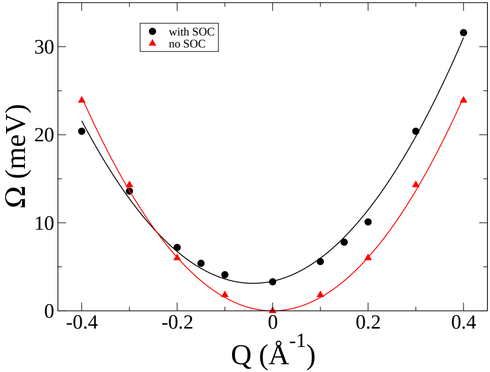
where the superscript ^{0} denotes mean-field quantities and the vector $\vec{\chi}$ is a compact notation for the set of four susceptibilities

$$\vec{\chi} = \left(\chi^{+-}, \chi^{\uparrow -}, \chi^{\downarrow -}, \chi^{--}\right)^T.$$

PRB **82**, 014428 (2010)

Results - Fe/W(110)

Anisotropy - gap in the SW spectrum



With SOC,

$$\Omega(Q_\parallel) = 3.4 + 11.8\,Q_\parallel + 143.4\,Q_\parallel^2.$$

Without SOC,

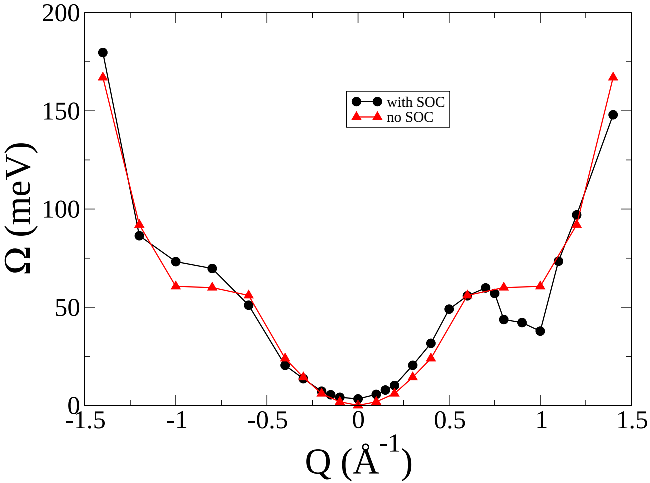
$$\Omega(Q_\parallel) = 143.4\,Q_\parallel^2.$$

(all energies in meV)

PRB **82**, 014428 (2010)

Results - Fe/W(110)

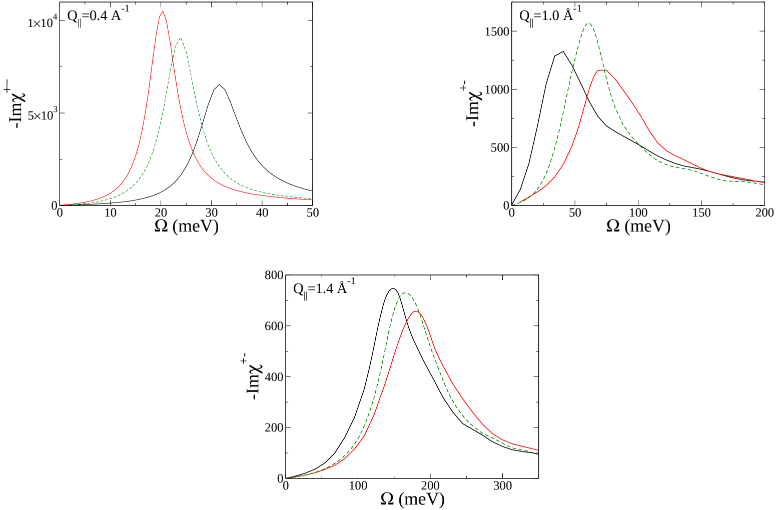
$\pm\vec{Q}$ asymmetry in the SW dispersion relation:



PRB **82**, 014428 (2010)

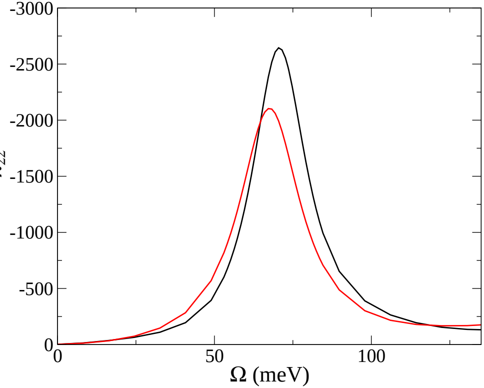
Results - Fe/W(110)

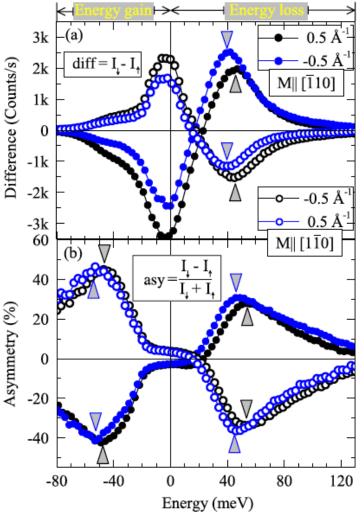
$\pm\vec{Q}$ asymmetry in the SW spectra



PRB **82**, 014428 (2010)

2Fe/W(110) - Comparison to experiment

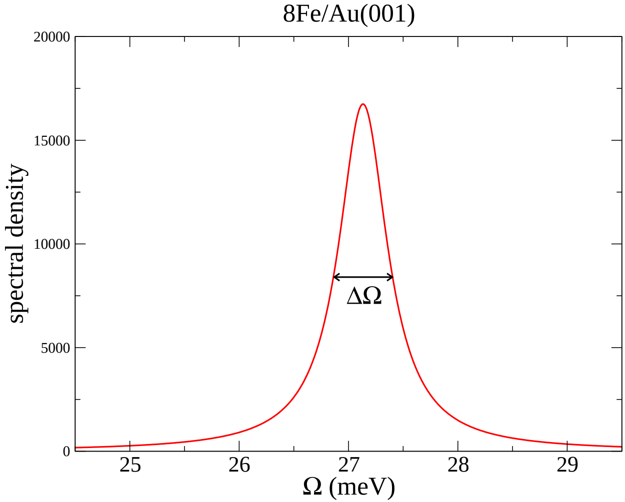




Experiments: Tang, Zhang, Tudosa, Prokop, Etzkorn and Kirschner, PRL **99**, 087202 (2007)

Spin Pumping

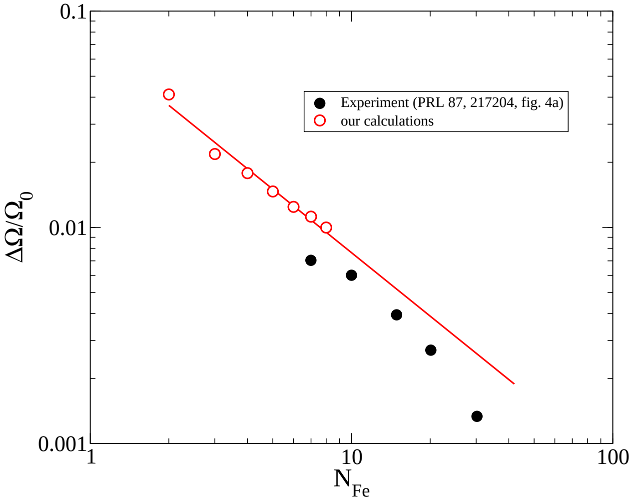
We obtain FMR spectra from our formalism by taking $|\vec{Q}| \to 0$.



PRB **73**, 054426 (2006)

Spin pumping without SOC

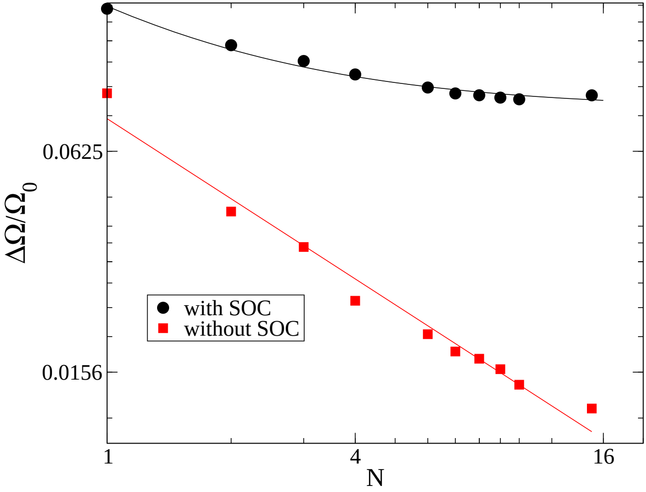
"Damping" comes only from "leakage" to the semi-infinite substrate.



PRB **73**, 054426 (2006)

Spin pumping with SOC

By including SOC in the calculation of FMR spectra we can evaluate the relative importance of intrinsic and spin-pumping contributions to the damping.



Single atoms on metallic surfaces

Ancient history: D. Mills and P. Lederer, 1967

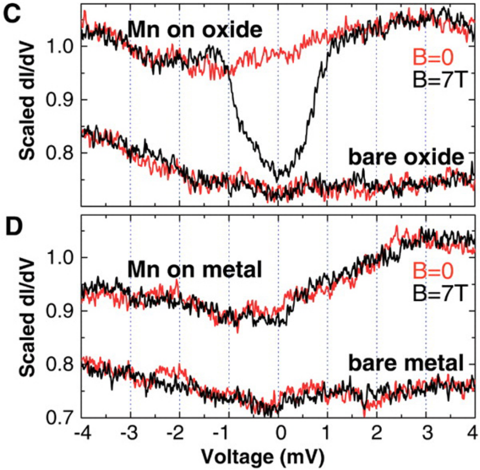
Single magnetic impurity in a transition metal host show a resonance of **zero linewidth if monitored by a long wavelength probe**. If **only the local response is measured**, there is a **considerable** g **shift and large linewidth**.

They employed a simple one-band model with intra-atomic repulsion in the impurity site only.

D. L. Mills and P. Lederer, Phys. Rev. **160**, 590 (1967).

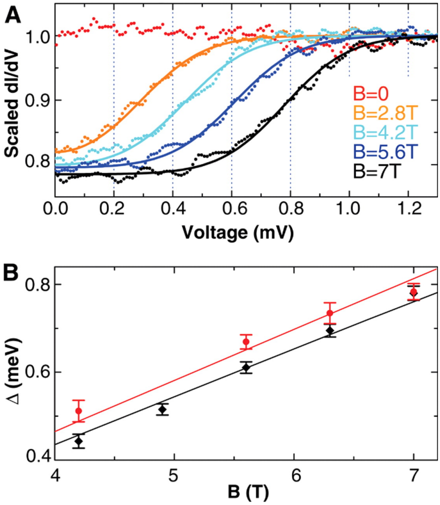
The experimental technique: ISTS

Inelastic **S**canning **T**unneling **S**pectroscopy: sample-tip bias is varied. Steps in the $\frac{dI}{dV}$ signal indicate excitation.



A. J. Heinrich *et al.*, Science **306**, 466 (2004).

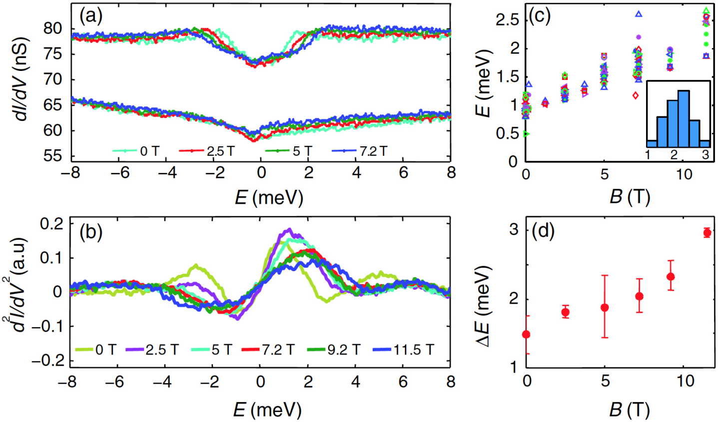
Is this a magnetic excitation?



A. J. Heinrich *et al.*, Science **306**, 466 (2004).

Red dots on panel B were obtained for a Mn atom at the edge of an oxide path.

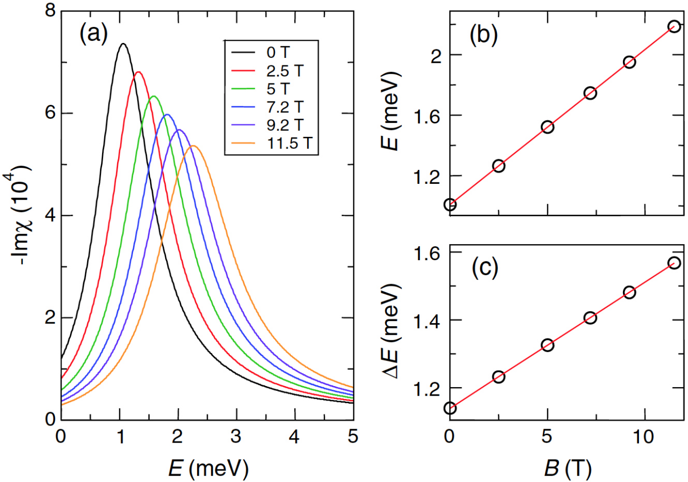
Metallic surfaces – Fe adatom on Cu(111) surface



$\bar{g} \approx 2$

PRL **106**, 037205 (2011).

Fe adatom on Cu(111) surface - Calculations



$g = 1.8$; magnetic anisotropy ~ 1 meV.

PRL **106**, 037205 (2011).

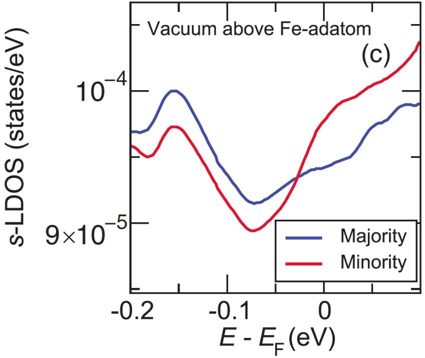
The role of surface states

Cu(111) × Ag(111)

Both have free-electron-like states localized at the surface layer.

- In Cu(1111), the surface state is well below the Fermi level.
- In Ag(111) it is very close to E_F ($\sim E_F - 50$ meV)

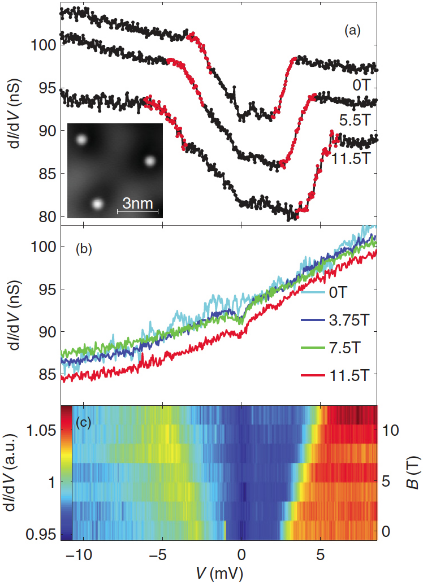
Coupling of the Fe adatom with the Ag(111) surface state generates a spin-split bound state



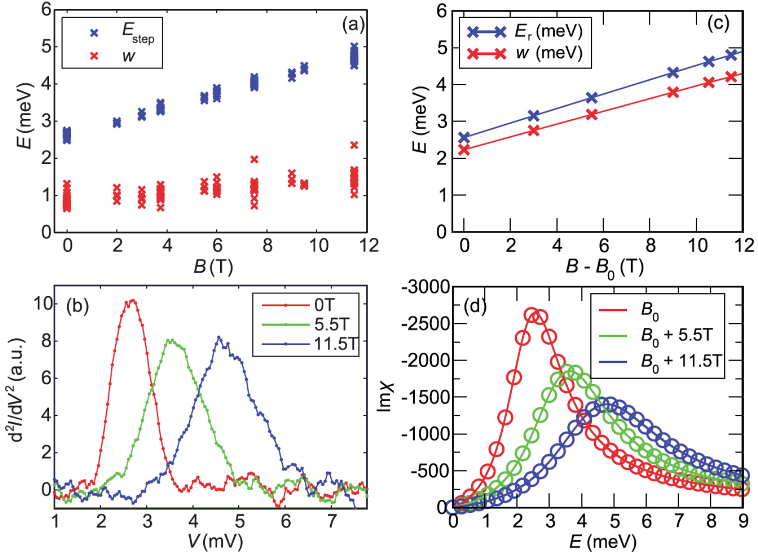
Fe adatom on Ag(111) - ISTS

ISTS at

(a) Fe adatom
(b) Ag substrate
(c) Fe adatom



Fe adatom on Ag(111) - theory



Experiment: $g \approx 3.1$. Theory: $g \approx 3.3$

Origin of the large g shift

It is difficult to present a simple explanation for the origin of the anomalous g (although we know it is connected to the surface state).
A hint: for a single magnetic site,

$$\chi = \frac{\chi_0}{1 + U\chi_0} = \frac{\chi_0^{(R)} + i\chi_0^{(I)}}{1 + U\chi_0^{(R)} + iU\chi_0^{(I)}}$$

But, for small Ω,

$$\chi_0^{(R)} \approx \chi_0(0) + \alpha\Omega, \quad \chi_0^{(I)} \approx -\beta\Omega.$$

Thus,

$$\chi^{(I)} \approx \frac{\beta\Omega}{\{1 + U[\chi_0(0) + \alpha\Omega]\}^2 + (U\beta\Omega)^2}.$$

Resonance condition

From the last expression we extract the resonance condition

$$\Omega_r = \frac{|\frac{1}{U} + \chi_0(0)|}{\sqrt{\alpha^2 + \beta^2}}.$$

In the absence of a Zeeman field B, $\chi_0(0) = -\frac{1}{U} \Rightarrow \Omega_r = 0$.

Fe is actually a rare case: Co, Cr and Mn all have $g \sim 2$.

Concluding remarks

- It is possible to understand several aspects of the spin dynamics from a microscopic point of view, with an *ab-initio*-like theory (*i.e.* relying only on information about the electronic structure of the system).
- SOC has been incorporated to our method of calculating spin excitations; the phenomenology of spin excitations in the presence of SOC is reproduced by our microscopic method.
- **Further developments:** IST spectrum directly from our method; dynamical coupling through "interacting" substrates (Pd).