

Magnetism from first-principles

Arthur Ernst

Max-Planck-Institut für Mikrostrukturphysik, Halle (Saale)

September 12th 2014

Magnetism from first-principles

I. First-principles material design

- Green function method
- Adiabatic spin wave approach
- Dynamical susceptibility approach

II. Spin waves in bulk

- Fe bcc as example
- Magnon lifetime in Heuslers

II. Spin waves in thin films

- Fe, Co, Ni films on Cu(001) and W(110)
- Landau Damping of Magnons in thin magnets
- Softening of spin waves in thin Fe films

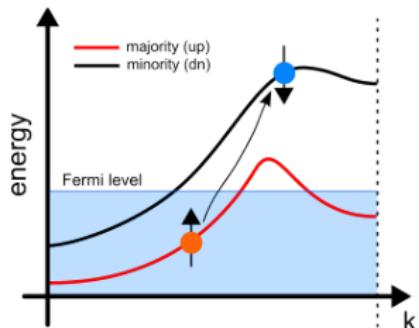
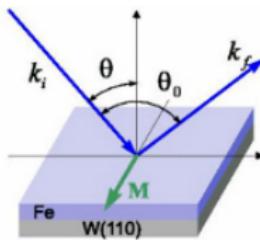
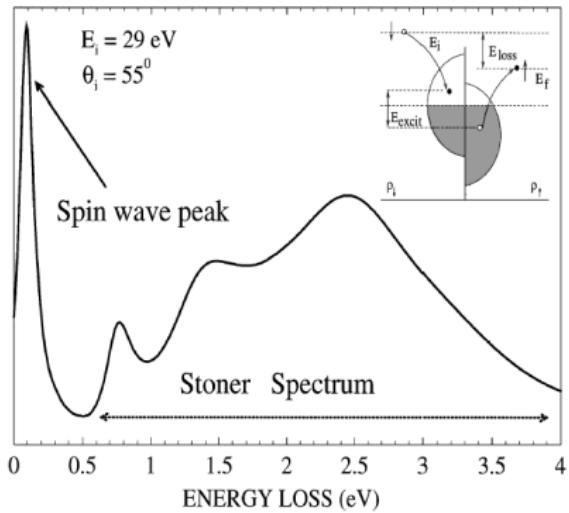
III. Summary & outlook

I. First-principles material design

I. First-principles material design

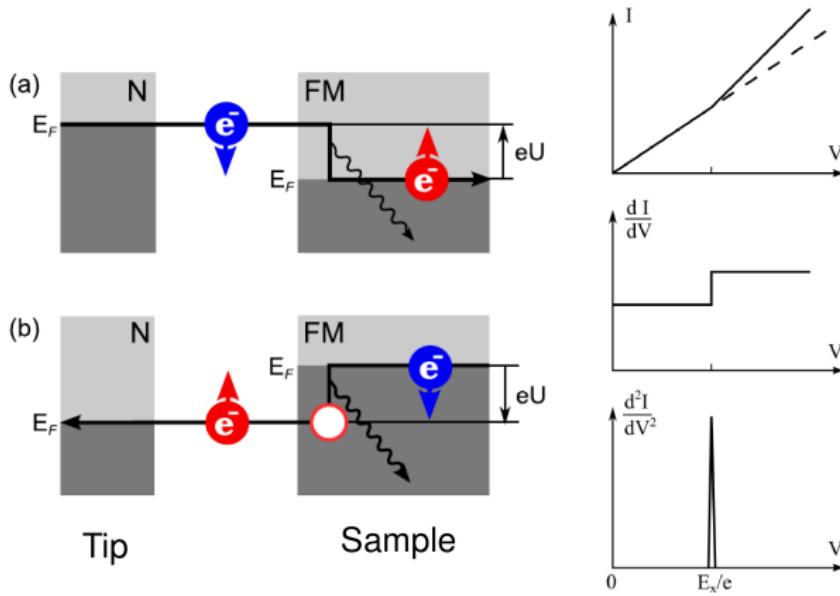
Spin waves and Stoner excitations in solids

SPIN FLIP LOSS SPECTRUM



I. First-principles material design

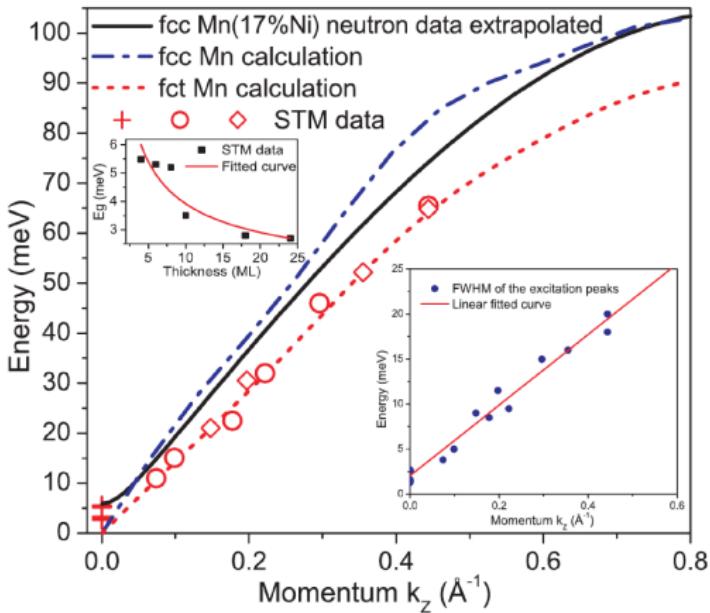
Magnon creation mechanism in inelastic STM



Experiment: Wulf Wulfhekel et al

I. First-principles material design

Spin wave dispersion

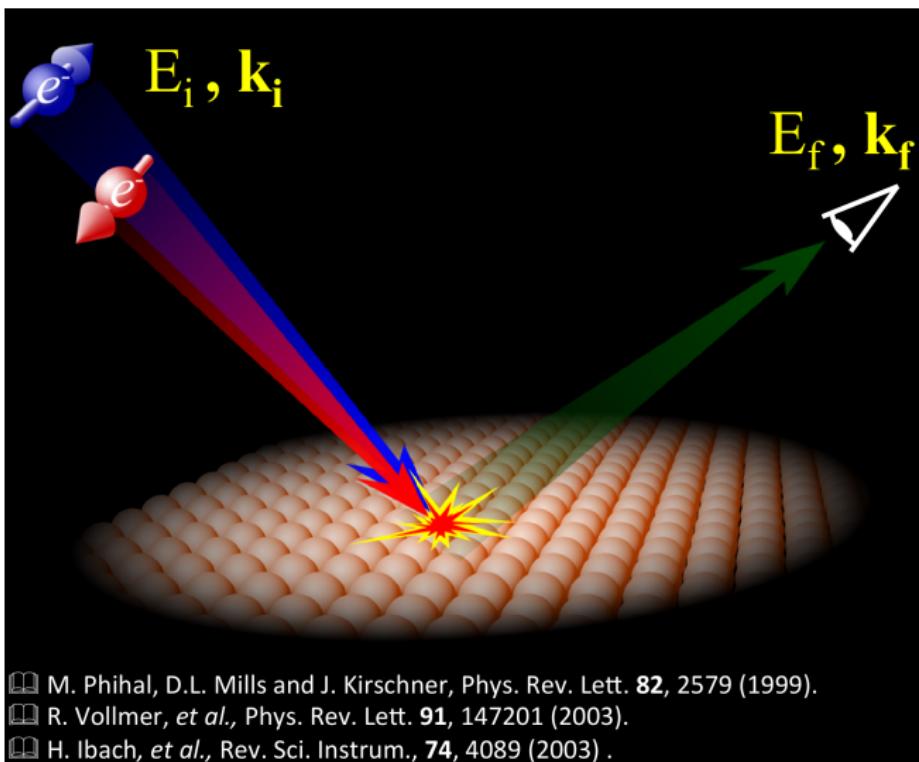


Adiabatic spin wave calculations with HUTSEPOT

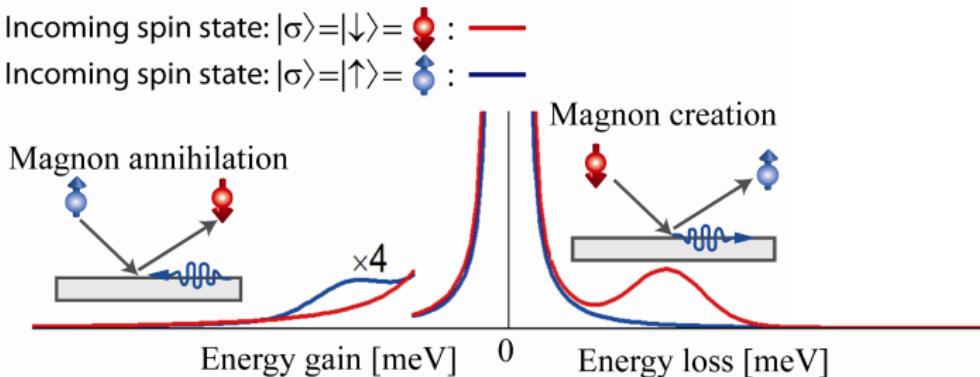
Phys. Rev. Lett. 101, 167201 (2008)

I. First-principles material design

SPEELS: Spin Polarized Electron Energy Loss Spectroscopy



I. First-principles material design



A magnon carries a total angular momentum of $1 \hbar$

- The magnon annihilation process is allowed for incident electrons of majority character
- The magnon creation process is allowed for incident electrons of minority character

Green function method

Parameter free simulations of realistic materials

Ab-initio Kohn-Sham approach

- Wave function equation

$$\left[\varepsilon + \frac{\hbar^2}{2m} \nabla^2 - V_{\text{eff}}(\mathbf{r}) \right] \Psi(\mathbf{r}; \varepsilon) = 0$$

- Green function equation

$$\left[\varepsilon + \frac{\hbar^2}{2m} \nabla^2 - V_{\text{eff}}(\mathbf{r}) \right] G(\mathbf{r}, \mathbf{r}'; \varepsilon) = \delta(\mathbf{r} - \mathbf{r}')$$

Korringa (1947), Kohn & Rostoker (1954)

- Dyson equation

$$G = G_0 + G_0 \Delta V_{\text{eff}} G$$

$$\Delta V_{\text{eff}} = V_{\text{eff}} - V_{\text{eff}}^0$$

Green function method

Explicit Green function for various systems

- Bulk

$$G_{bulk} = G_{free} + G_{free} V_{eff} G_{bulk}$$

- Surfaces & interfaces

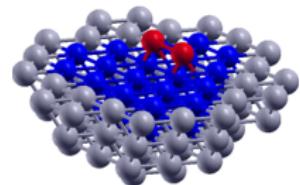
$$G_{surf} = G_{bulk} + G_{bulk} \Delta V_{eff} G_{surf}$$



Wildberger et al. (1997), Uiberacker et al. (1998)

- Defects in bulk & surfaces

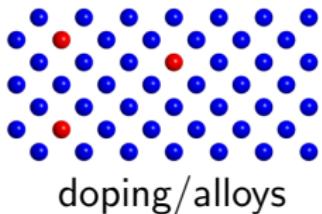
$$G_{cluster} = G_{host} + G_{host} \Delta V_{eff} G_{cluster}$$



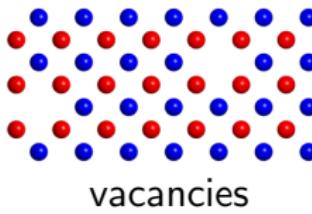
Zeller & Dederichs (1979)

Method: Coherent potential approximation

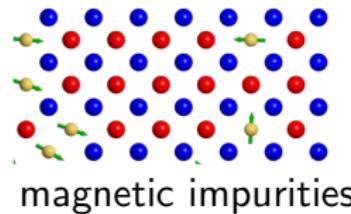
Alloys and pseudo-alloys



doping/alloys



vacancies

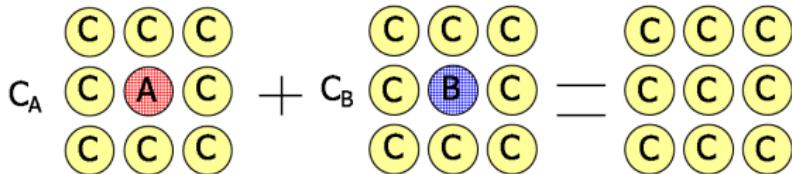


magnetic impurities

Coherent potential approximation

Soven (1967), Györffy (1972)

CPA equation for a binary alloy: $c_A G_A + c_B G_B = G_C$



Nonlocal CPA: Charge and Spin-Fluctuations

D. A. Rowlands, A. Ernst, J. B. Staunton, B. L. Györffy, PRB 73, 165122 (2006)

Green-Funktion-Methode

- Explicit Green Function
- Dimensions: 1D, 2D, 3D & Cluster
- $\mathcal{O}(N)$ method
- CPA for disordered alloys
- *multi-code approach*: crystalline structure from VASP or experiments

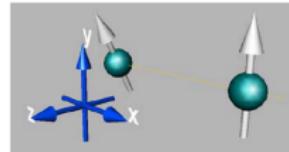
I. First-principles material design

Exchange interaction and spin waves in adiabatic approximation

Heisenberg Model: $H = -\frac{1}{2} \sum_{i \neq j} J_{ij} \vec{e}_i \cdot \vec{e}_j$

Exchange integrals: *Liechtenstein et al, 1984*

$$J_{ij} = \frac{1}{4\pi} \int_{-\infty}^{\varepsilon_F} d\varepsilon \Delta_i(\varepsilon) \tau_{\uparrow}^{ij}(\varepsilon) \Delta_j(\varepsilon) \tau_{\downarrow}^{ji}(\varepsilon)$$



Mean-field critical temperature: $k_B T_C^{MFA} = \frac{2}{3} \sum_{j \neq 0} J_{0j}$

Magnetization and critical temperature in the RPA

$$N_{\alpha\beta}(\vec{q}) = \delta_{\alpha\beta} \sum_{\gamma} J_{\alpha\gamma}(0) \left\langle \vec{e}_{\gamma}^z \right\rangle - \left\langle \vec{e}_{\alpha}^z \right\rangle J_{\alpha\beta}(\vec{q})$$
$$\left\langle \vec{e}_{\alpha}^z \right\rangle = L \left(2\beta \left\{ \frac{1}{\Omega} \int d\vec{q} \left[N^{-1}(\vec{q}) \right]_{\alpha\alpha} \right\}^{-1} \right)$$

I. First-principles material design

Linear response theory in the DFT

Change of the magnetization via an applied magnetic field $\mathbf{B}(\mathbf{q}; \omega)$:

$$\delta\mathbf{m}(\mathbf{q}; \omega) = \chi(\mathbf{q}; \omega)\mathbf{B}(\mathbf{q}; \omega)$$

Transverse susceptibility:

$$\chi^\pm(\mathbf{q}; \omega) = -\sum_j \frac{\langle 0|\hat{\sigma}^+(\mathbf{q})|j\rangle\langle j|\hat{\sigma}^-(\mathbf{-q})|0\rangle}{\omega - (E_j - E_0) + i0^+} + \text{H.c.}$$

Singularities of the susceptibility define magnons and their lifetime

I. First-principles material design

Linear response theory in the DFT

Calculation of the transverse susceptibility:

- Pauli susceptibility (non-interactive):

$$\chi_{ij}^0(\mathbf{x}, \mathbf{x}'; \omega) = \sum_{km} \sigma_{\alpha\beta}^i \sigma_{\gamma\delta}^j (f_k - f_m) \frac{\phi_k^*(\mathbf{x}\alpha)\phi_m(\mathbf{x}\beta)\phi_m(\mathbf{x}'\gamma)\phi_k(\mathbf{x}'\delta)}{\omega + (\varepsilon_k - \varepsilon_m) + i0^+}$$

- Dyson equation for the transverse susceptibility:

$$\begin{aligned} \chi(\mathbf{x}, \mathbf{x}'; \omega) &= \chi_0(\mathbf{x}, \mathbf{x}'; \omega) \\ &+ \int \int d\mathbf{x}_1 d\mathbf{x}_2 \chi_0(\mathbf{x}, \mathbf{x}_1; \omega) f_{xc}(\mathbf{x}_1, \mathbf{x}_2; \omega) \chi(\mathbf{x}_2, \mathbf{x}'; \omega) \end{aligned}$$

with the Kernel $f_{xc}(\mathbf{x}, \mathbf{x}'; t - t') = \frac{\mathbf{B}_{xc}((\mathbf{x}; t))}{\partial \mathbf{m}(\mathbf{x}'; t')}$

E. K. U. Gross & W. Kohn, (1985)

I. First-principles material design

Paramagnetic susceptibility

Static spin susceptibility:

$$\chi_{ij} = \frac{\beta}{3} \mu_i^2 \delta_{ij} + \frac{\beta}{3} \sum_k S_{ik}^{(2)} \chi_{kj}$$

Direct correlation function for the local moments:

$$S_{ik}^{(2)} = \frac{\partial^2 \langle \Omega \rangle}{\partial M_i \partial M_k}$$

generalized grand potential
magnetization

Fourier transformed paramagnetic susceptibility

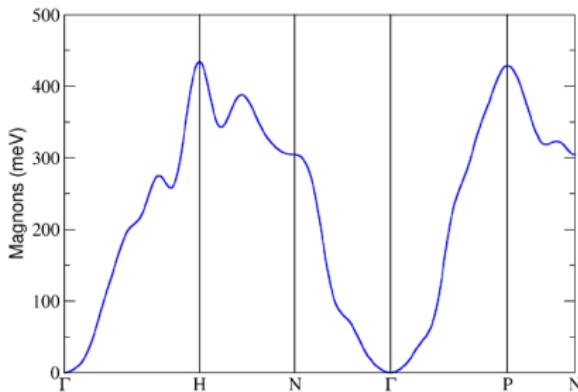
$$\chi(\vec{q}) = \frac{1/3 \beta \mu^2}{1 - 1/3 \beta S^{(2)}(\vec{q})}$$

CPA-KKR Problem

$$S^{(2)}(\vec{q}) = \int d\vec{k} \tau^C(\vec{k}) \tau^C(\vec{k} + \vec{q})$$

I. First-principles material design

Adiabatic calculations of spin waves



Fe, bcc

Mean-field: 1175 K

RPA: 770 K

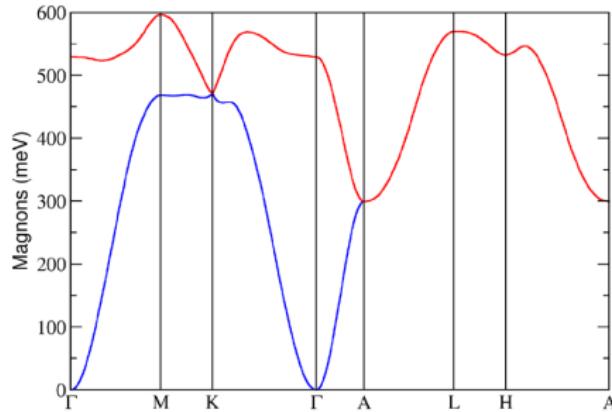
Experiment: 1040 K

Co, hcp

Mean-field: 1456 K

RPA: 1184 K

Experiment: 1311 K

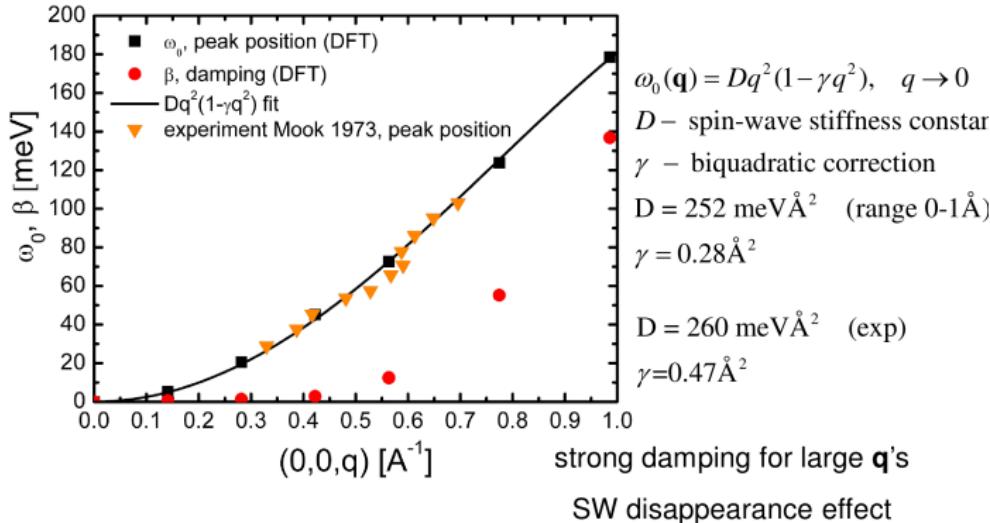


I. First-principles material design

Dynamical calculations of spin waves

Bulk bcc Fe

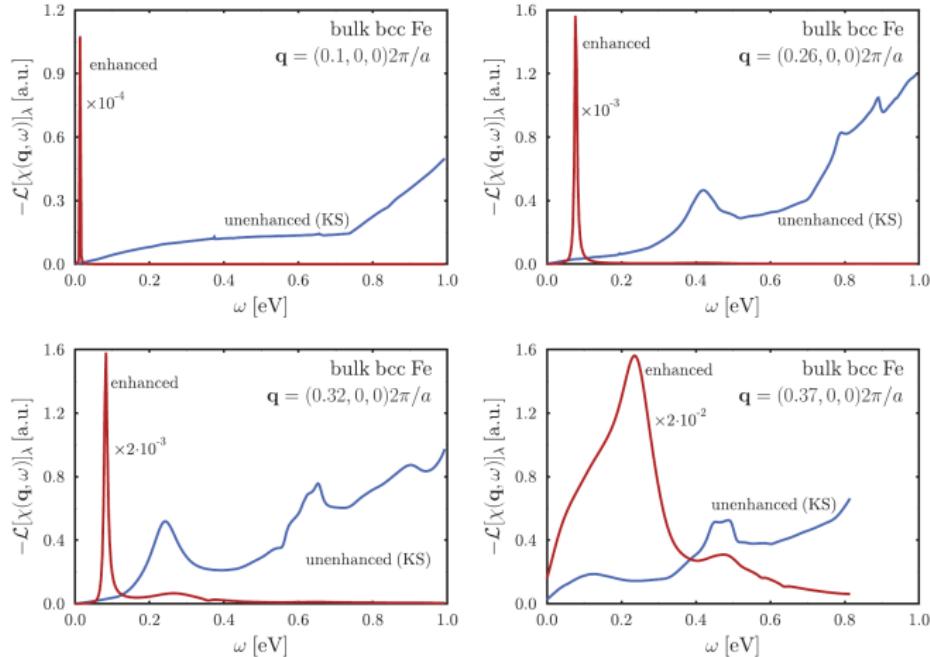
$$\chi(\omega, \mathbf{q}) = \frac{A(\mathbf{q})}{\omega - \omega_0(\mathbf{q}) + i\beta(\mathbf{q})}$$



I. First-principles material design

Dynamical calculations of spin waves

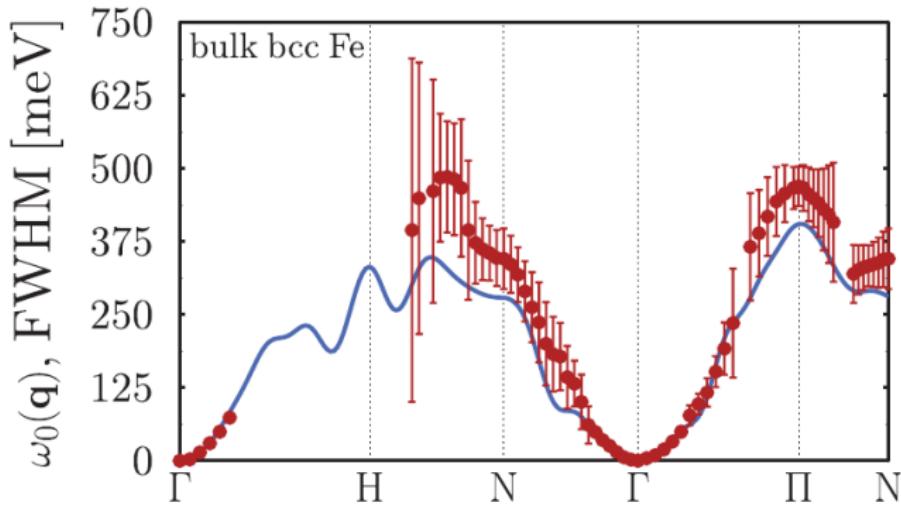
Examples of spin-flip spectra in Fe for different momenta along (100) direction



I. First-principles material design

Dynamical calculations of spin waves

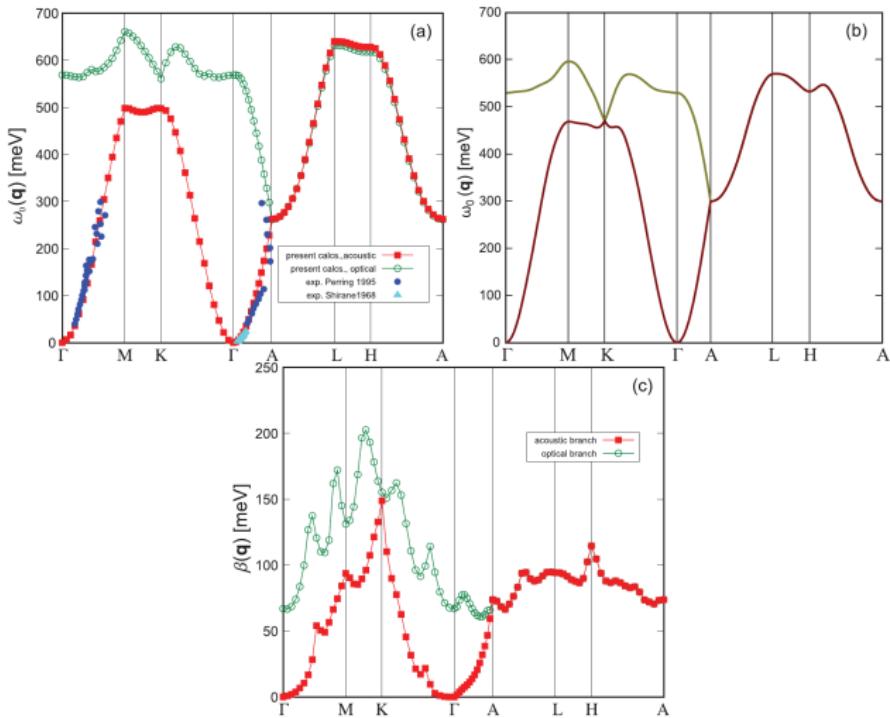
Spin waves in Fe: Adiabatic & Dynamical approaches



I. First-principles material design

Dynamical calculations of spin waves

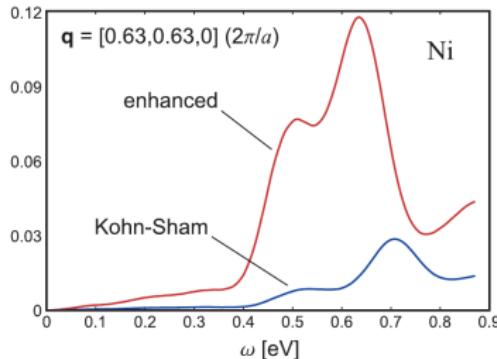
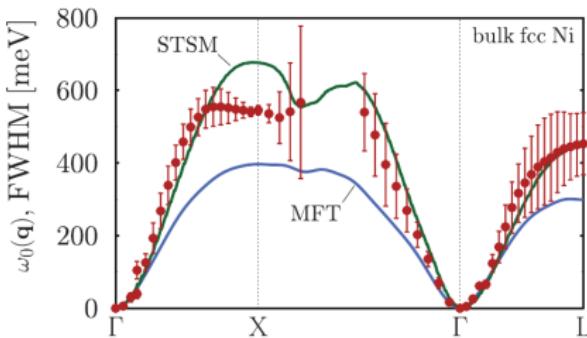
Spin waves in hcp Co: Adiabatic & Dynamical approaches



I. First-principles material design

Dynamical calculations of spin waves

Spin waves in fcc Ni: Adiabatic & Dynamical approaches



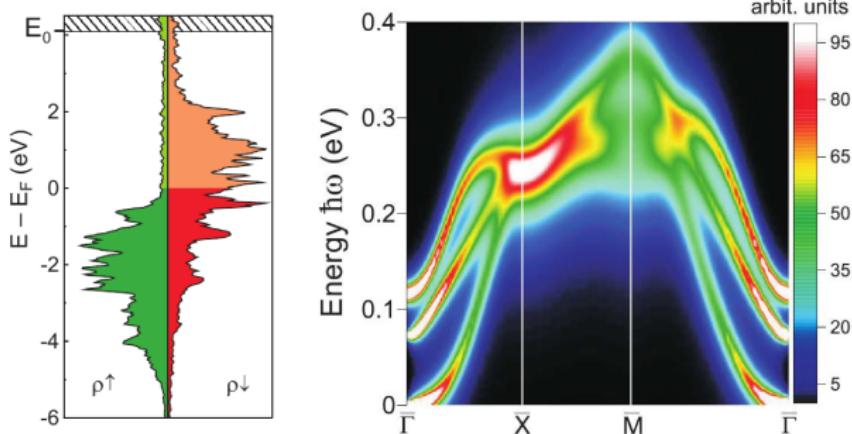
II. Spin waves in thin films

II. Spin waves in thin films

Goals

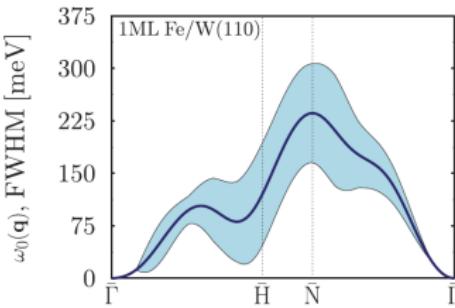
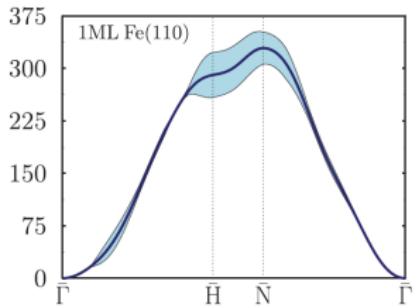
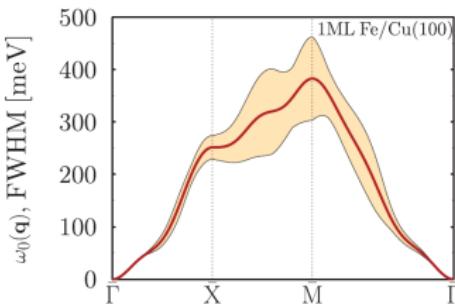
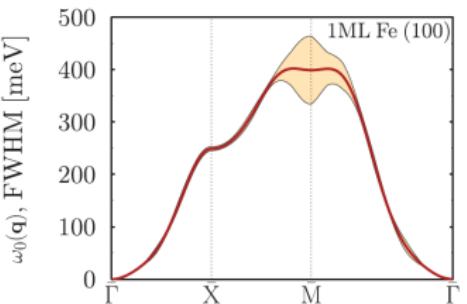
- Magnons in systems with a reduced symmetry
- Lifetime of magnons
- Role of the substrate
- Support experiments

Life time broadening of spin waves in 3 ML Fe/ Cu(001)



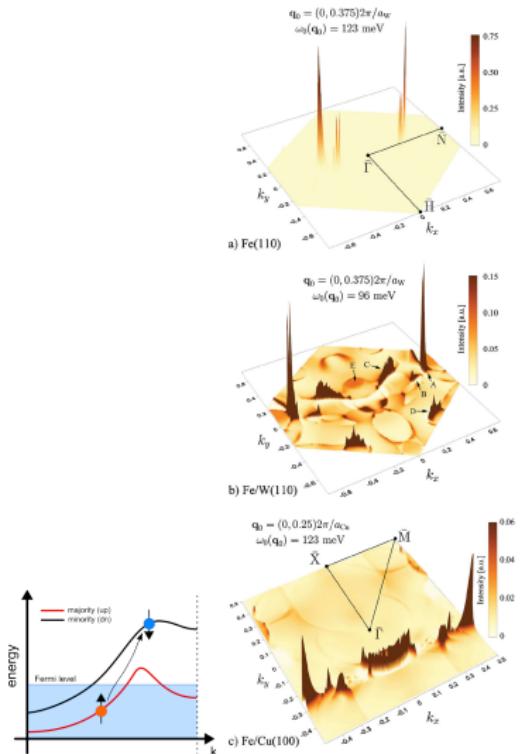
II. Spin waves in thin films

Fe films on Cu(001) & W(110)



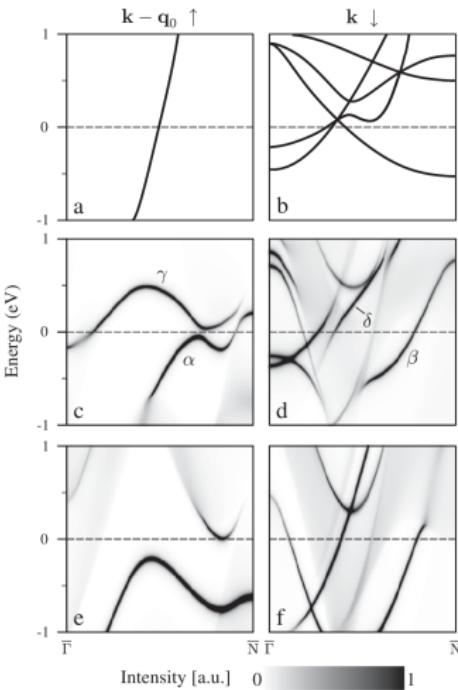
II. Spin waves in thin films

Landau damping of magnons in Fe films



Stoner transitions

A. Ernst, MPI Halle

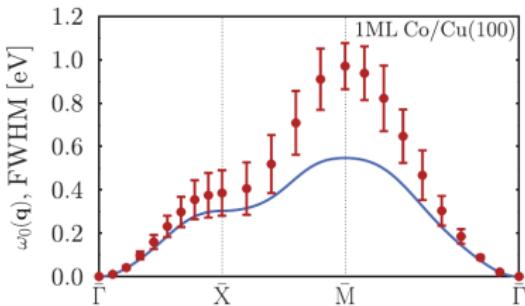
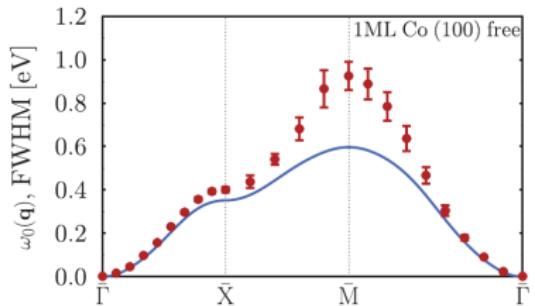


2D band structure

Magnetism from first-principles

II. Spin waves in thin films

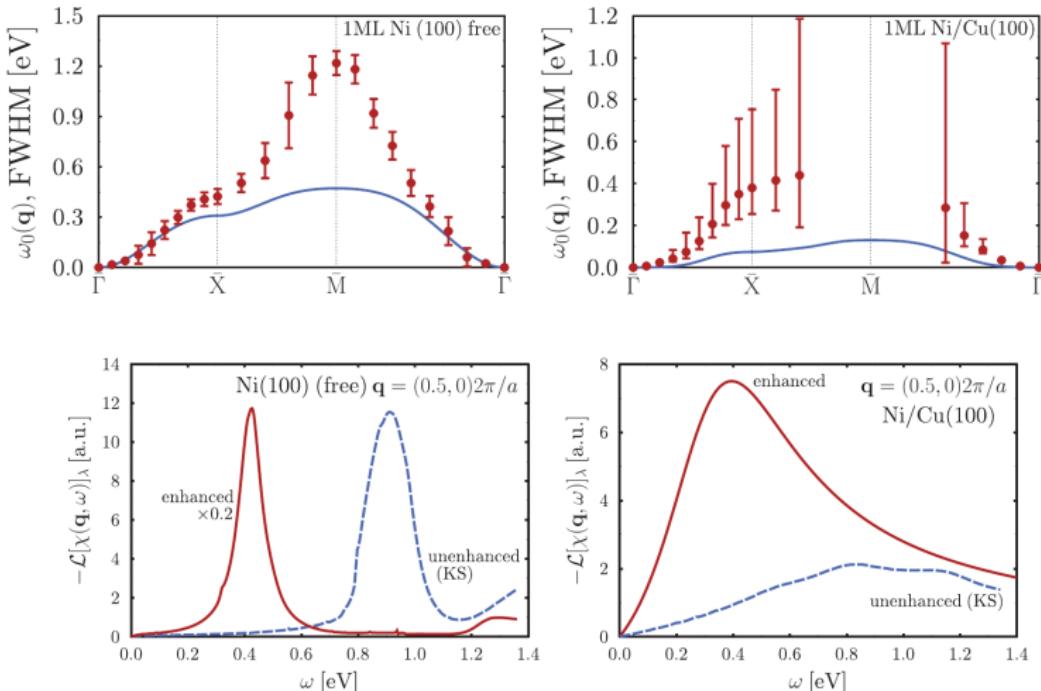
Co films on Cu(001)



Cu(001) substrate does not change much spin waves in Co films

II. Spin waves in thin films

Ni films on Cu(001)

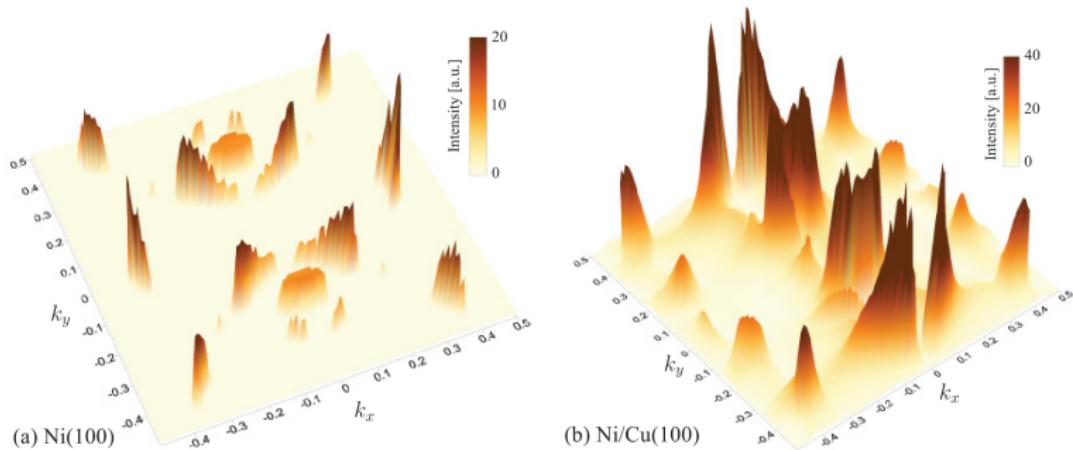


Significant broadening of the unenhanced susceptibility on Cu(001)

II. Spin waves in thin films

Ni films on Cu(001)

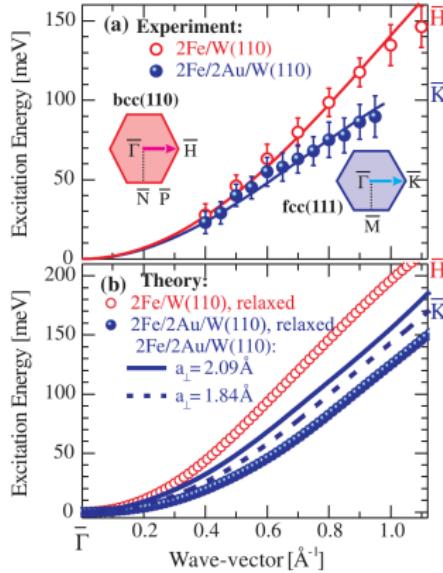
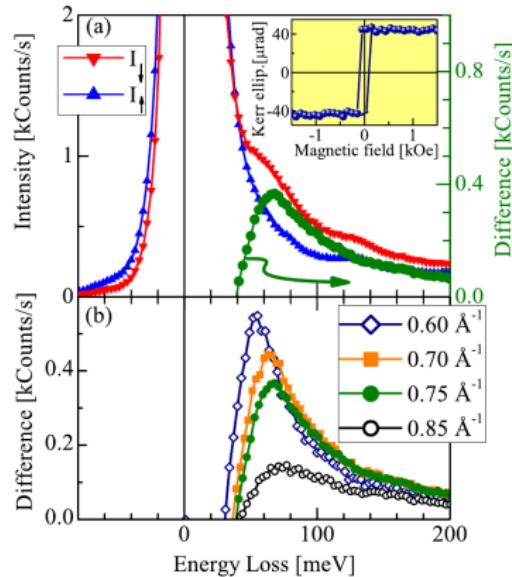
Landau complexes



Strong damping of magnons in Ni films due to Cu(001) substrate

II. Spin waves in thin films

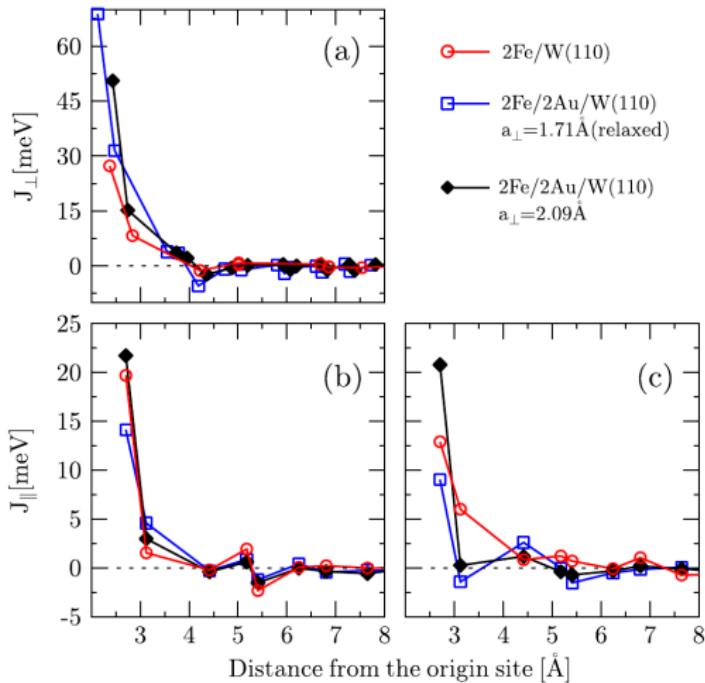
Fe films on W(110) and Au/W(110)



Motivation: Spin wave design

II. Spin waves in thin films

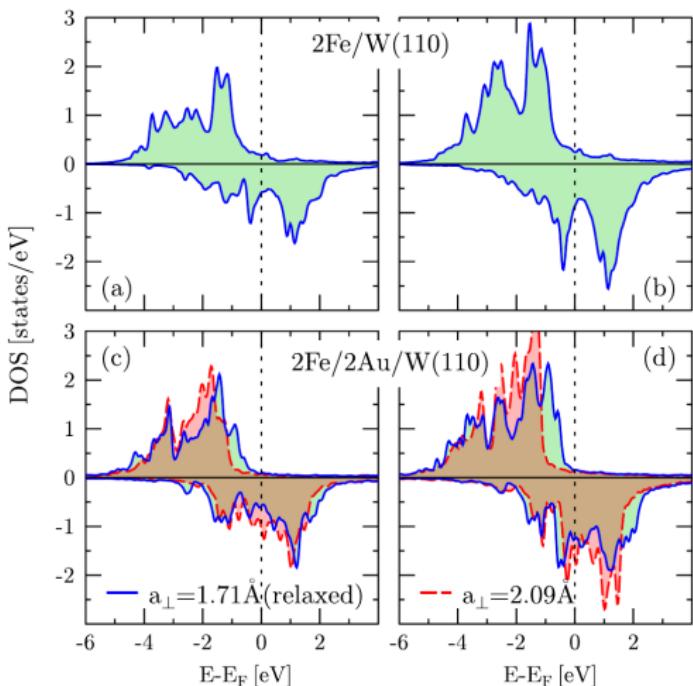
Fe films on W(110) and Au/W(110)



Exchange interaction in Fe film on W(110) and Au/W(110)

II. Spin waves in thin films

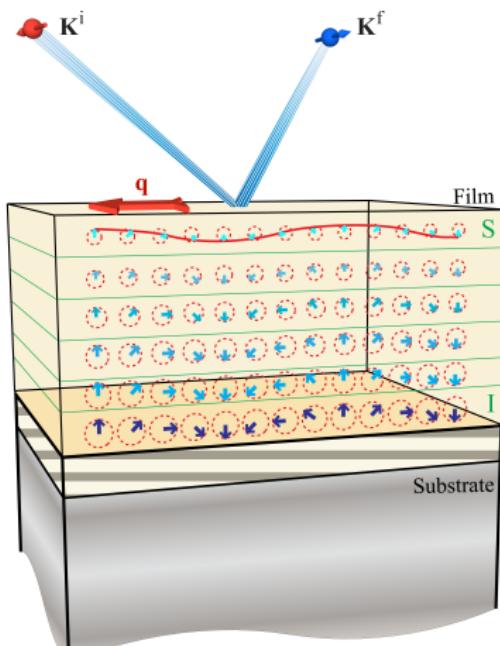
Fe films on W(110) and Au/W(110)



Density of states of Fe film on W(110) and Au/W(110)

II. Spin waves in thin films

Fe films on Ir(001)



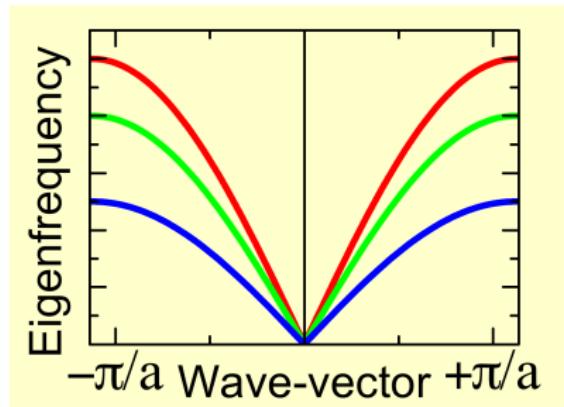
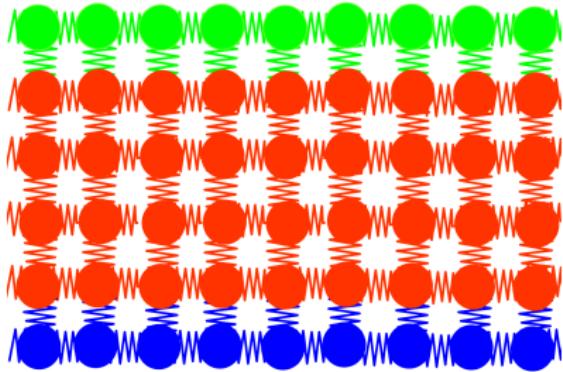
Goals

- Magnon softening in Fe films on Ir(001)
- Structure or origin of magnons
- Interpretation of experiment

II. Spin waves in thin films

Fe films on Ir(001)

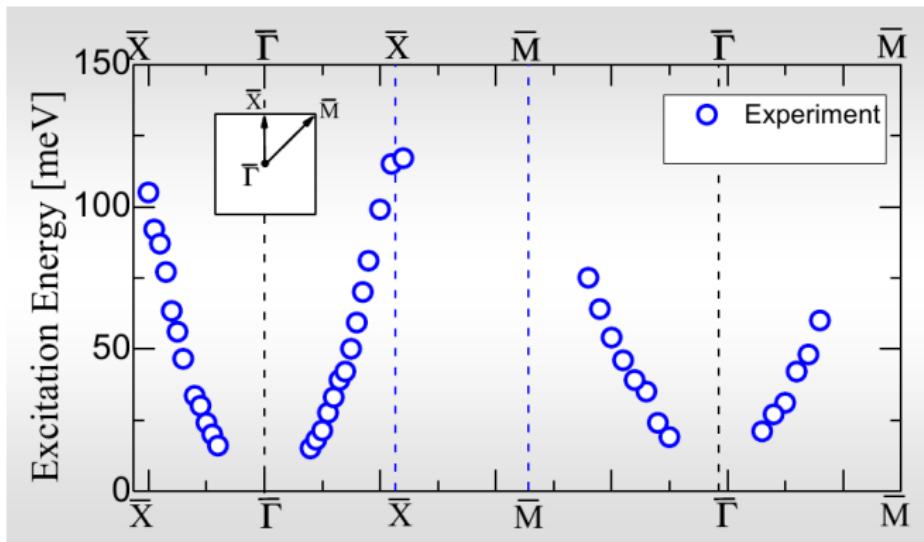
Simple model: An infinite array of coupled oscillators
Classical analog of magnons: Excitations in coupled oscillators



II. Spin waves in thin films

Fe films on Ir(001)

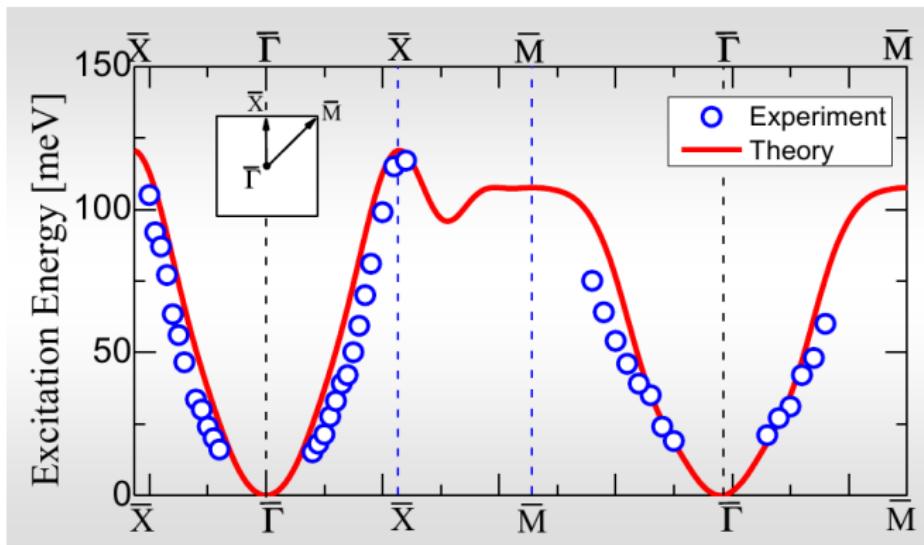
Magnons: Experiment



II. Spin waves in thin films

Fe films on Ir(001)

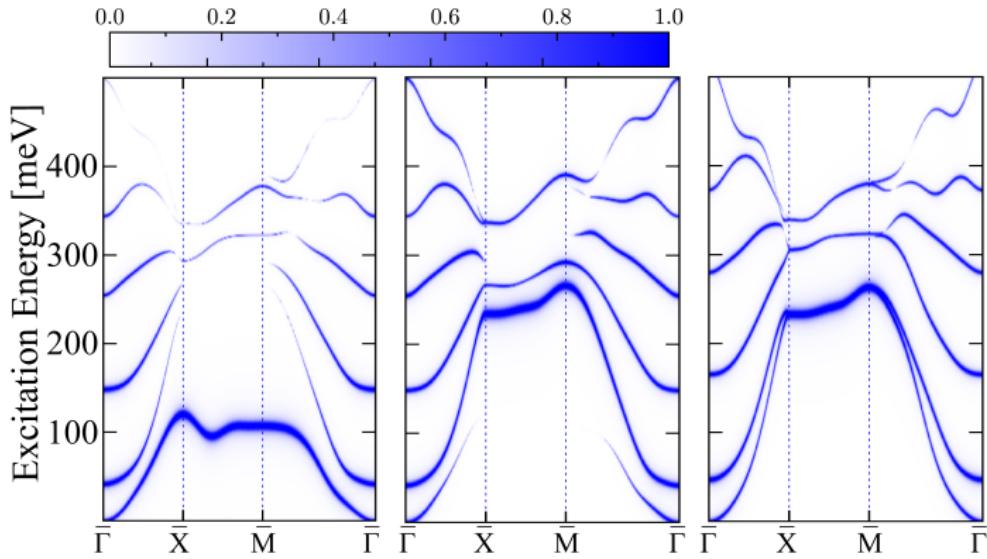
Magnons: Experiment & Theory



II. Spin waves in thin films

Fe films on Ir(001)

Layer-resolved transverse susceptibility

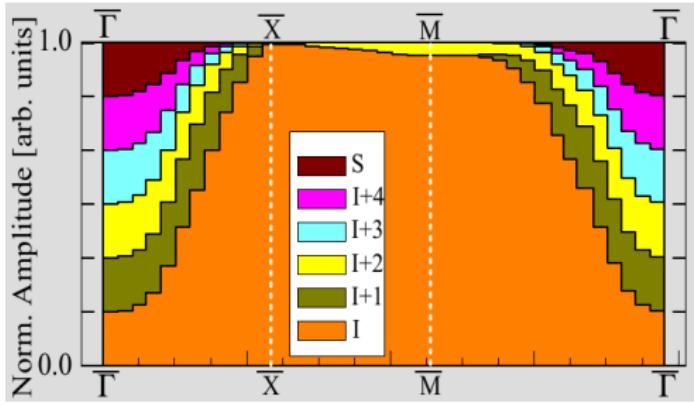
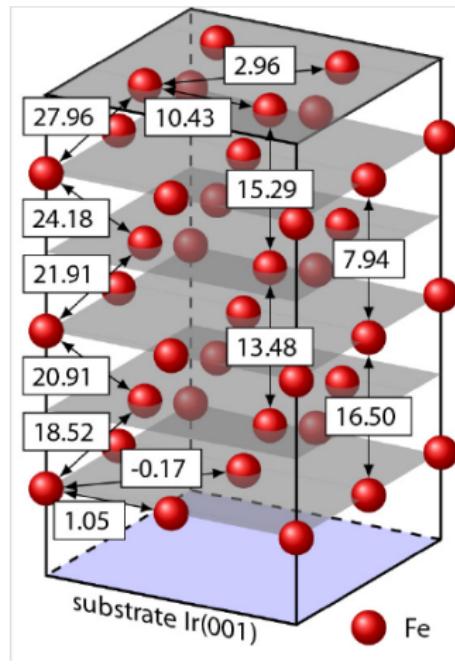


Interface mode Surface mode Free standing 6ML Fe

II. Spin waves in thin films

Fe films on Ir(001)

Exchange interaction in 6ML Fe/Ir(001)



Summary & Outlook

Results

- First-principles design of spin waves is possible
- Spin wave lifetime is related to the interaction of spin waves with the Stoner continuum
- Spin waves in thin films can be affected by the substrate

Future

- Relativistic extension of the code
- Coherent potential approximation for the susceptibility
- Electron-magnon interaction

Acknowledgment

Theory (MPI)

- Pawel Buczek
- Leonid Sandratskii

Experiment (MPI)

- Tzu-Hung Chuang
- Khalil Zakeri

Experiment (KIT)

- Timofey Balashov
- Wulf Wulfhekel