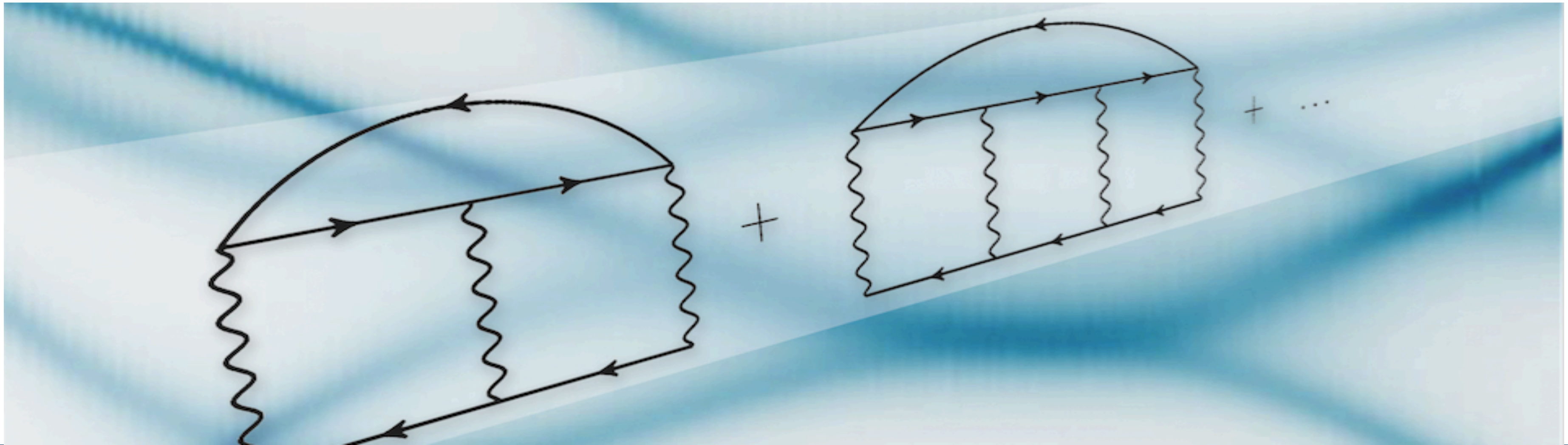


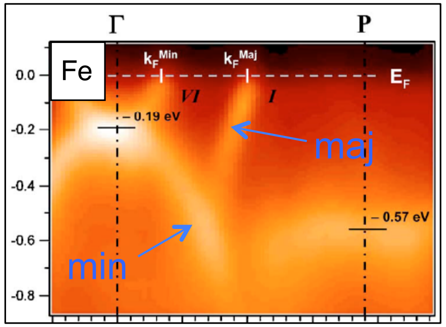
ELECTRON-MAGNON SCATTERING IN ELEMENTARY FERROMAGNETS FROM FIRST PRINCIPLES: LIFETIME BROADENING AND KINKS

CHRISTOPH FRIEDRICH

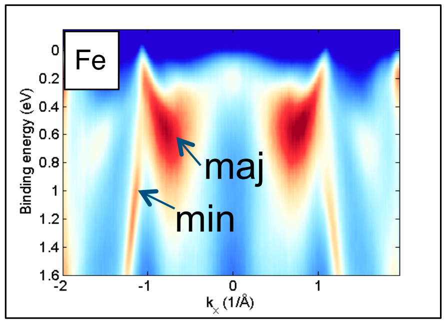
PETER GRÜNBERG INSTITUTE AND INSTITUTE FOR ADVANCED SIMULATION
FORSCHUNGSZENTRUM JÜLICH AND JARA, 52425 JÜLICH, GERMANY

ARPES MEASUREMENTS

Spin asymmetry in spectra

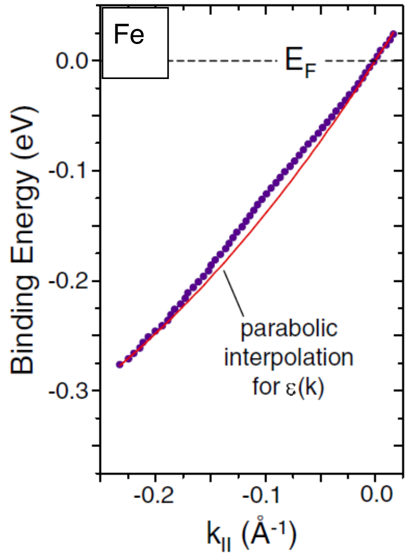


Schäfer *et al.*, PRL **72**, 155115 (2005)

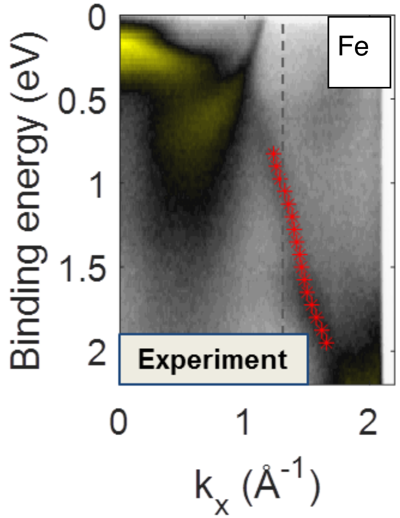


E. Mlynczak, L. Plucinski, unpublished

Anomalies in band dispersion of iron

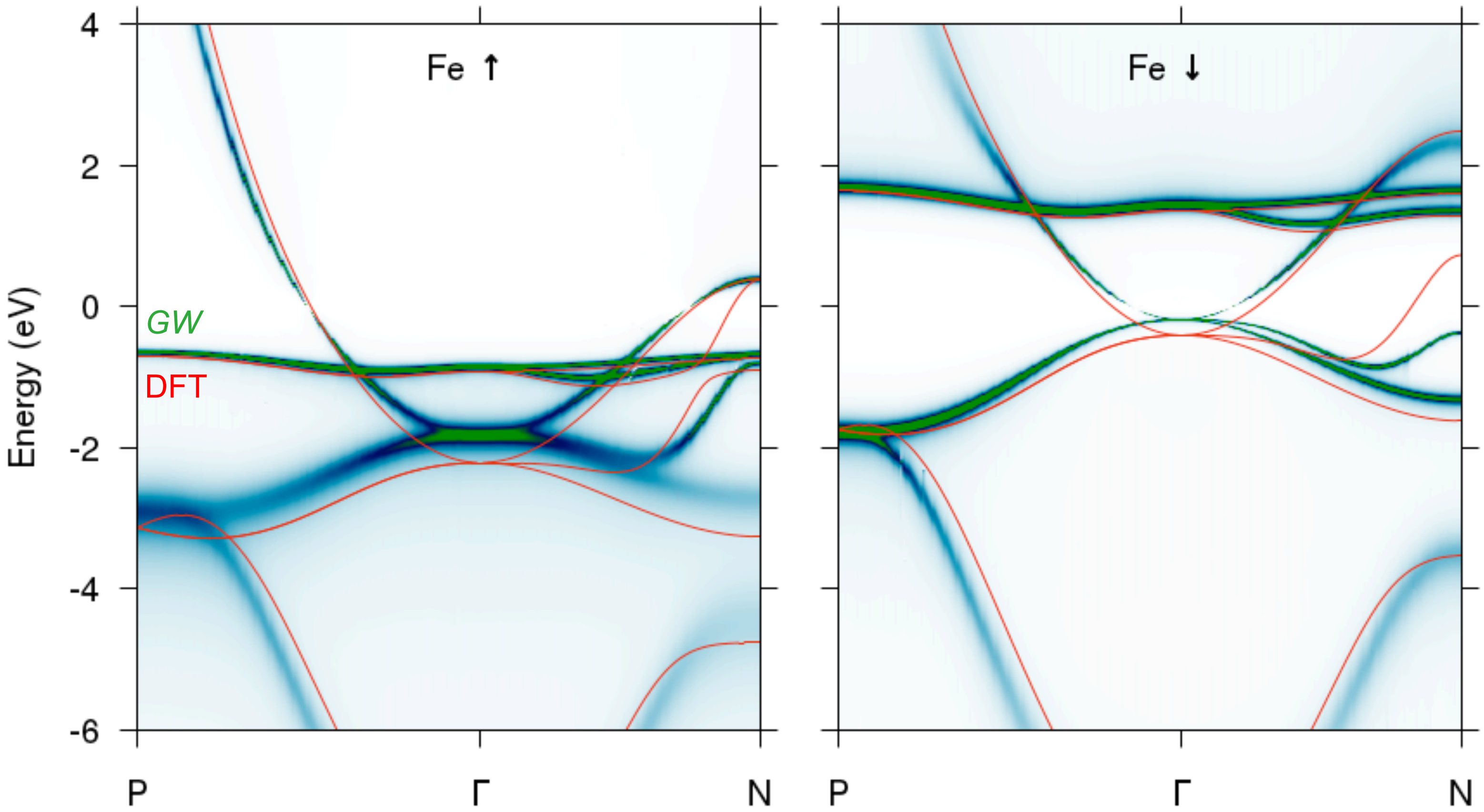


Schäfer *et al.*, PRL **92**, 097205 (2004)



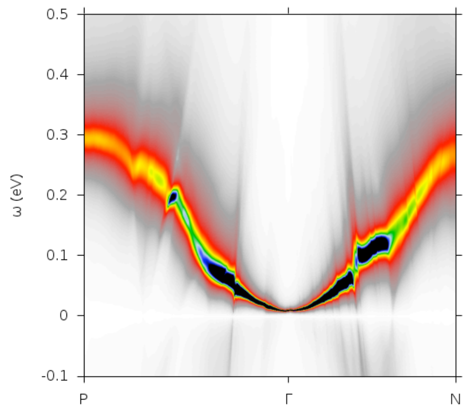
E. Mlynczak *et al.*,
Nature Communications 10,
505 (2019)

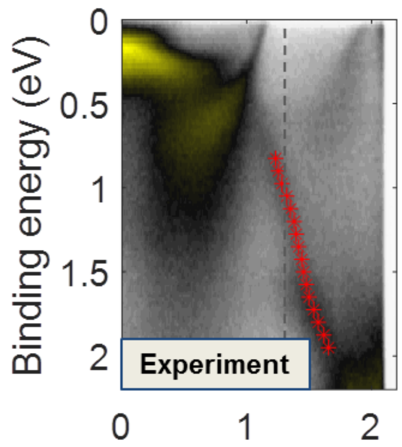
IRON *GW* CALCULATION



OVERVIEW

- Many-body spin excitations
  - Transverse magnetic response function
  - Bethe-Salpeter equation
  - Implementation (Wannier functions)
  - Transition-metal ferromagnets
  - Goldstone violation (resolved with COHSEX)
- Electron-magnon scattering
  - Iteration of Hedin equations (*GT* self-energy)
  - Aspects of implementation
  - Results for iron and nickel
    (lifetime broadening, kinks, band anomalies)
- Conclusions





MAGNETIC RESPONSE FUNCTION

JÜLICH Forschungszentrum

Response of the magnetization (electronic) density with respect to changes of the external magnetic (electric) field:

Spin-orbit coupling neglected

$$R(\mathbf{r}t,\mathbf{r}'t') = \begin{pmatrix} \frac{\delta\sigma_x(\mathbf{r},t)}{\delta B_x(\mathbf{r}',t')} & \frac{\delta\sigma_x(\mathbf{r},t)}{\delta B_y(\mathbf{r}',t')} & 0 & 0 \\ \frac{\delta\sigma_y(\mathbf{r},t)}{\delta B_x(\mathbf{r}',t')} & \frac{\delta\sigma_y(\mathbf{r},t)}{\delta B_y(\mathbf{r}',t')} & 0 & 0 \\ 0 & 0 & \frac{\delta\sigma_z(\mathbf{r},t)}{\delta B_z(\mathbf{r}',t')} & \frac{\delta\sigma_z(\mathbf{r},t)}{\delta V(\mathbf{r}',t')} \\ 0 & 0 & \frac{\delta\rho(\mathbf{r},t)}{\delta B_z(\mathbf{r}',t')} & \frac{\delta\rho(\mathbf{r},t)}{\delta V(\mathbf{r}',t')} \end{pmatrix}$$

$$B_x, B_y \rightarrow B^+ = B_x + iB_y$$
$$B^- = B_x - iB_y$$

Circularly polarized *B* field

MAGNETIC RESPONSE FUNCTION

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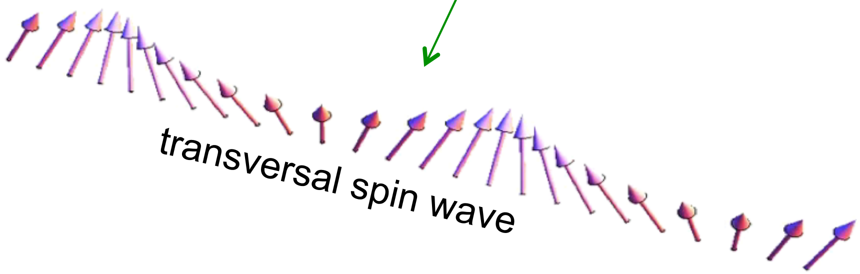
Response of the magnetization (electronic) density with respect to changes of the external magnetic (electric) field:

Spin-orbit coupling neglected

$$R(\mathbf{r}t,\mathbf{r}'t') = \begin{pmatrix} \frac{\delta\sigma^+(\mathbf{r},t)}{\delta B^+(\mathbf{r}',t')} & 0 & 0 & 0 \\ 0 & \frac{\delta\sigma^-(\mathbf{r},t)}{\delta B^-(\mathbf{r}',t')} & 0 & 0 \\ 0 & 0 & \frac{\delta\sigma_z(\mathbf{r},t)}{\delta B_z(\mathbf{r}',t')} & \frac{\delta\sigma_z(\mathbf{r},t)}{\delta V(\mathbf{r}',t')} \\ 0 & 0 & \frac{\delta\rho(\mathbf{r},t)}{\delta B_z(\mathbf{r}',t')} & \frac{\delta\rho(\mathbf{r},t)}{\delta V(\mathbf{r}',t')} \end{pmatrix}$$



$$R^{+-}(\mathbf{r}t,\mathbf{r}'t') = \frac{\delta\sigma^+(\mathbf{r},t)}{\delta B^+(\mathbf{r}',t')}$$

$$= \langle\Psi_0|\mathcal{T}[\sigma^+(\mathbf{r},t)\sigma^-(\mathbf{r}',t')]|\Psi_0\rangle$$

MAGNETIC RESPONSE FUNCTION

$$R^{+-}(1,2) = \frac{\delta\sigma^+(1)}{\delta B^+(2)} \qquad \sigma^+(1) = -i\sum_{\alpha,\beta}\sigma^+_{\beta\alpha}G_{\alpha\beta}(1,1^+)$$

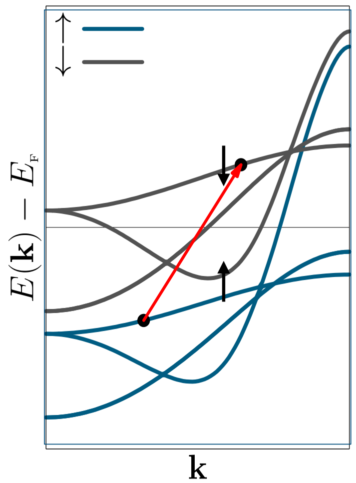
$$R = -i\frac{\delta G}{\delta B} = -i\frac{\delta}{\delta B}\left[G_0^{-1}-\Sigma\right]^{-1} = iGG\frac{\delta}{\delta B}\left[G_0^{-1}-\Sigma\right] = -iGG + GG\frac{\delta\Sigma}{\delta G}R$$

Dyson equation

Chain rule

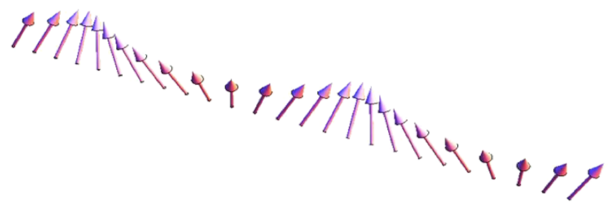
Stoner excitation

- Single particle
- Spin-flip
- Large energy scale (~eV)



Spin-wave excitation

- Collective excitation
- Small energy scale (~meV)



F. Aryasetiawan, K. Karlsson, PRB **60**, 7419 (1999)

BETHE-SALPETER EQUATION

JÜLICH
Forschungszentrum

Self-energy

$$\Sigma(12) = iG(12)W(1^{+}2)$$

$$\frac{\delta\Sigma}{\delta G} = iW + \cancel{iG\frac{\delta W}{\delta G}}$$

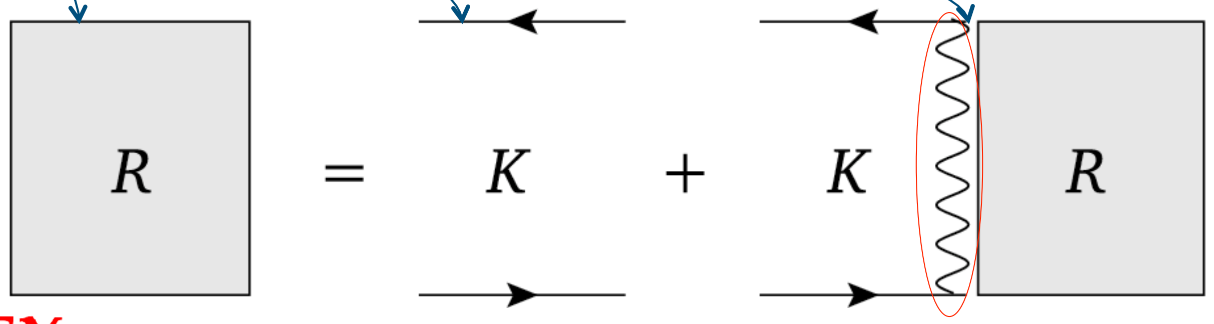
no SOC

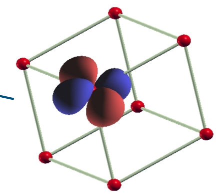
Bethe-Salpeter equation for spin excitations

$$R = K + K\,W\,R$$

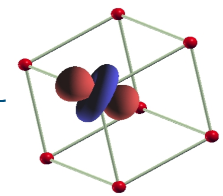
Static approximation:

$$W(\omega) \rightarrow W(\omega = 0)$$





Wannier functions

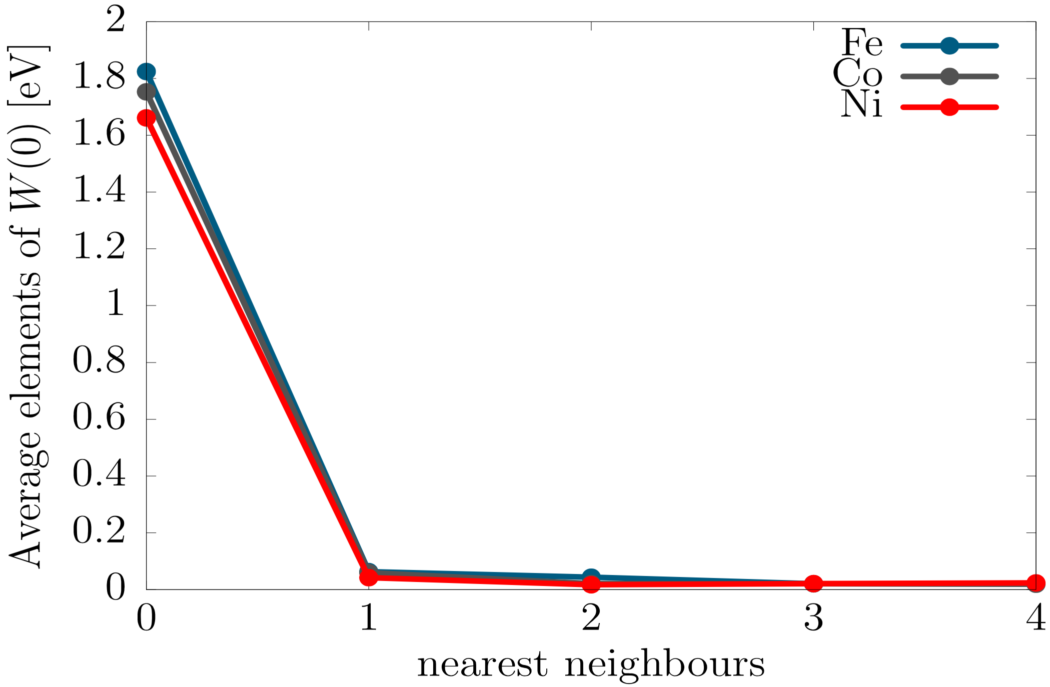


onsite approximation

SPEX

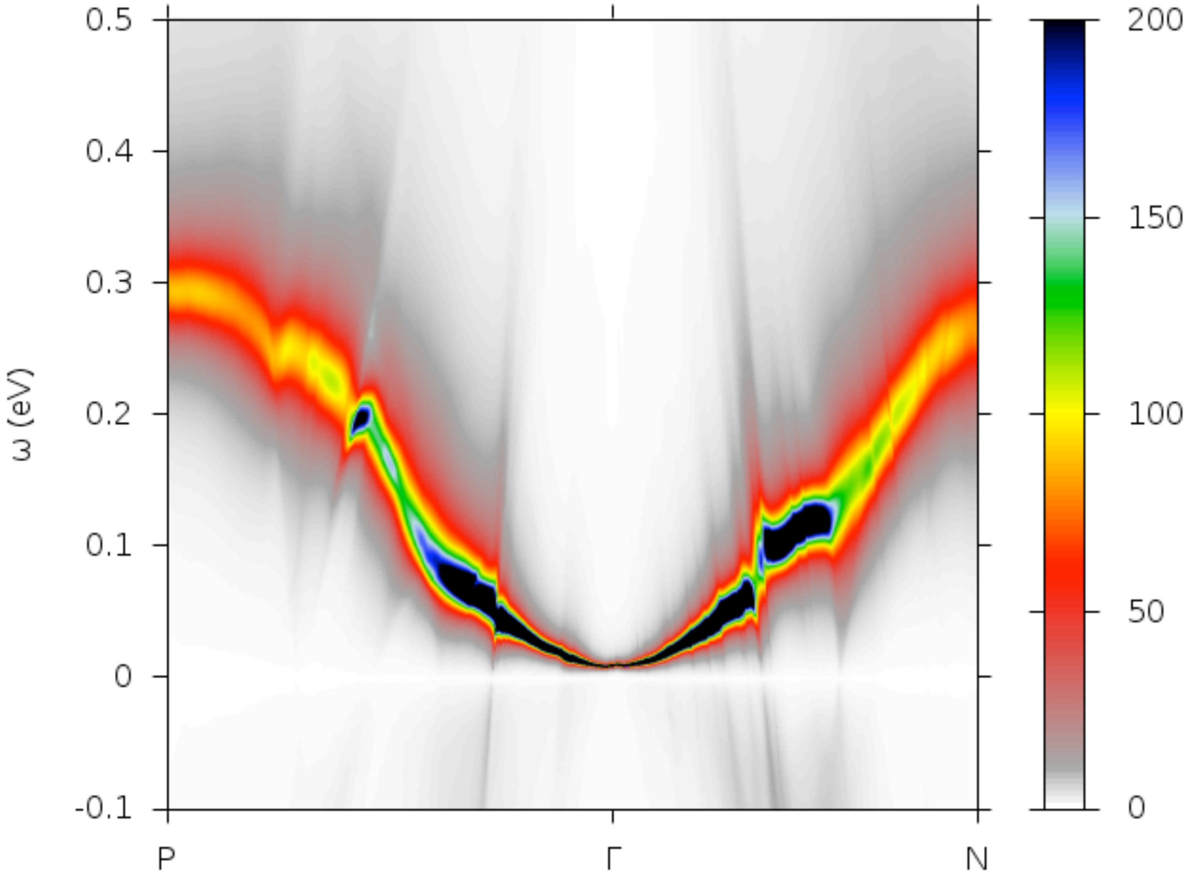
E. Sasioglu *et al.*, PRB **81**, 054434 (2010)

SPATIAL DEPENDENCE *W*

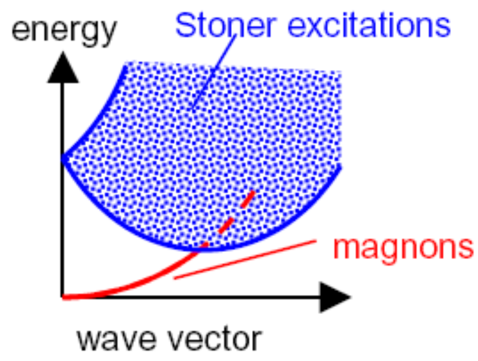


Largest contribution from the onsite interaction (~98%)

EXAMPLE: BCC IRON

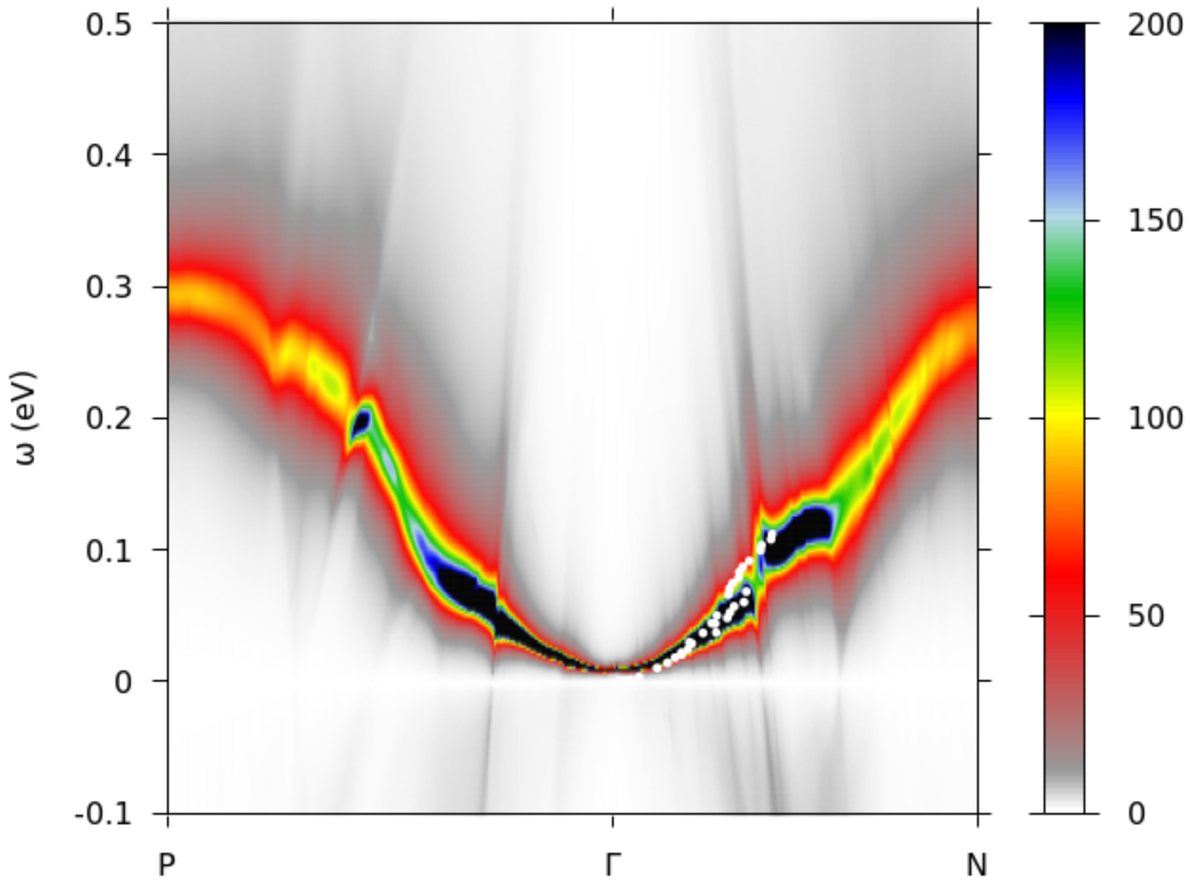


$$\mathrm{Im}\, R^{+-}(\mathbf{q},\omega)$$

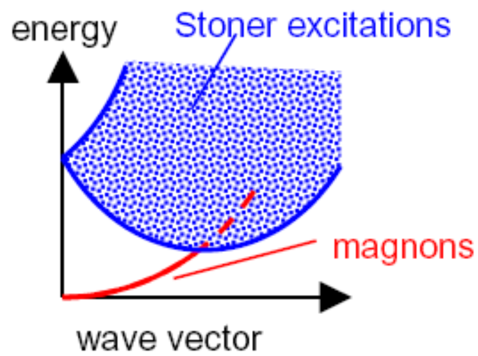


C. Friedrich, M.C.T.D. Müller, S. Blügel, "Handbook of Materials Modelling", Andreoni, Yip (eds), Springer (2019)

EXAMPLE: BCC IRON



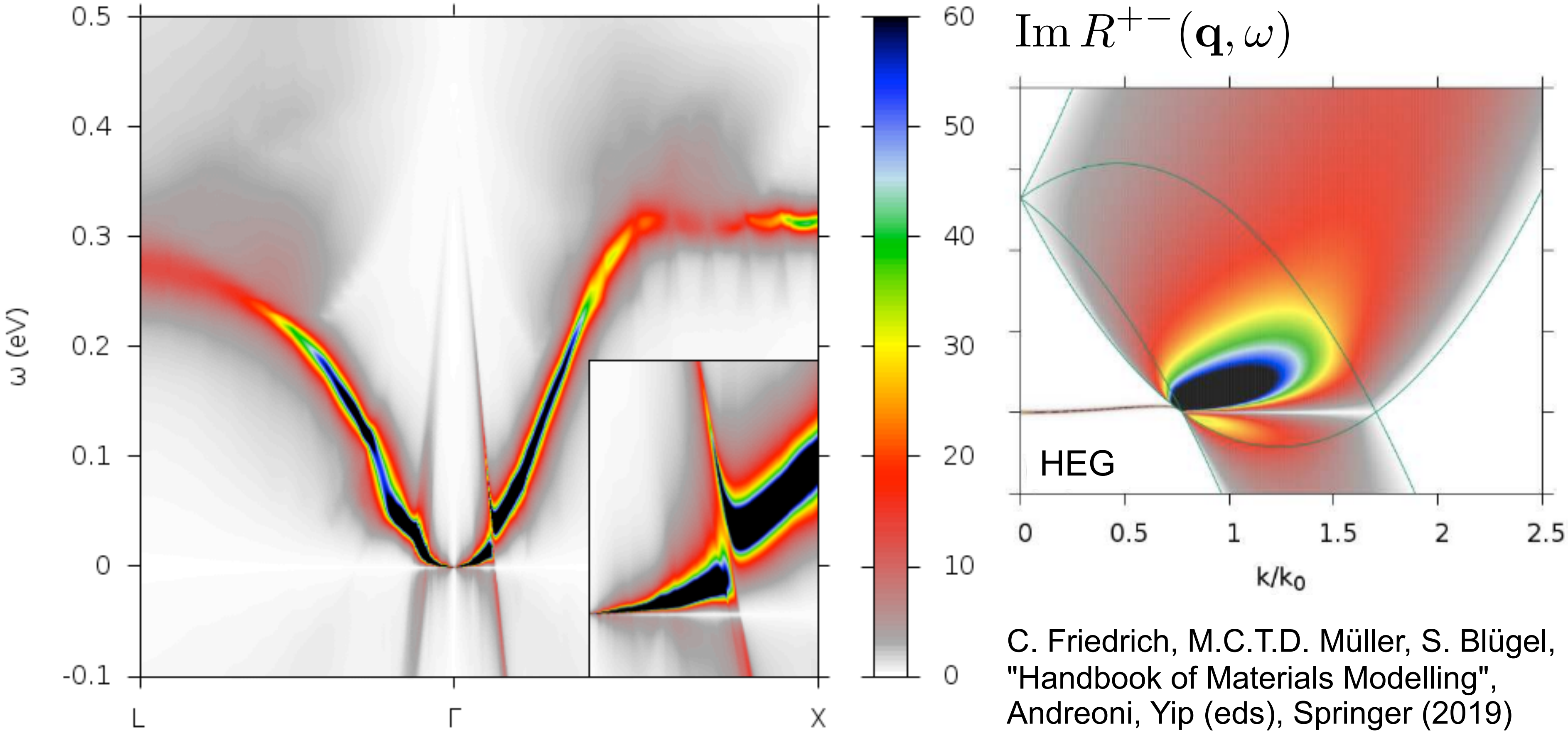
$$\mathrm{Im}\, R^{+-}(\mathbf{q}, \omega)$$



C. Friedrich, M.C.T.D. Müller, S. Blügel, "Handbook of Materials Modelling", Andreoni, Yip (eds), Springer (2019)

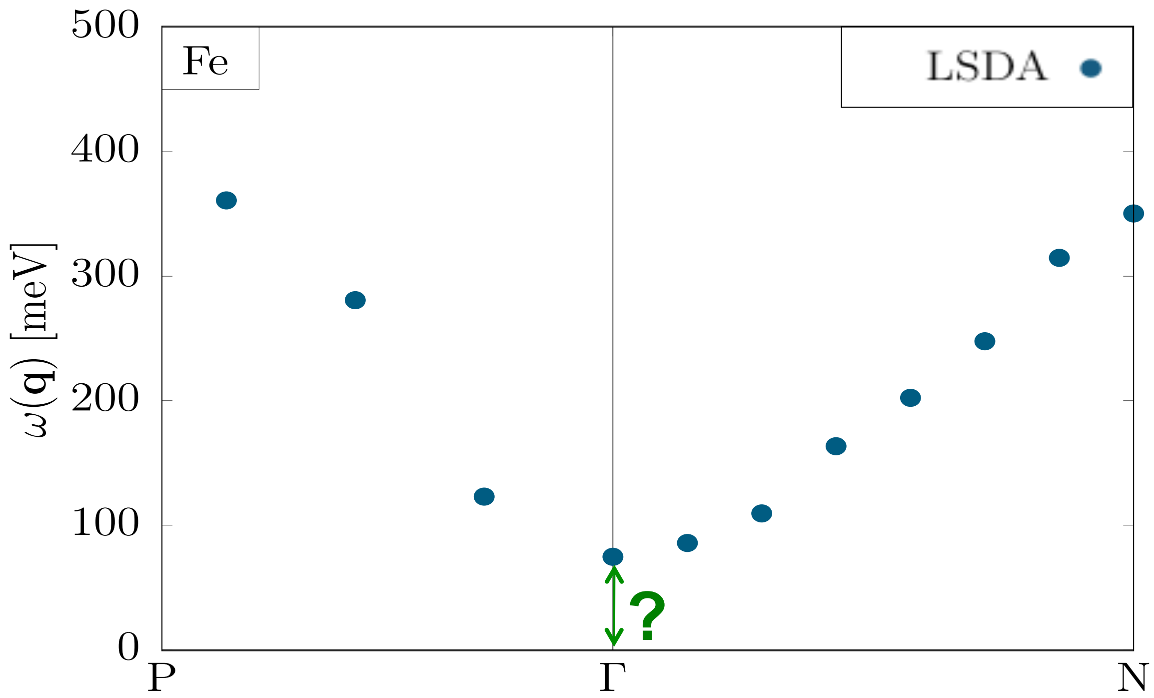
Experiments (white circles) Collins etal. PR 179, 417; Mook etal. PRB 7, 336

EXAMPLE: FCC NICKEL



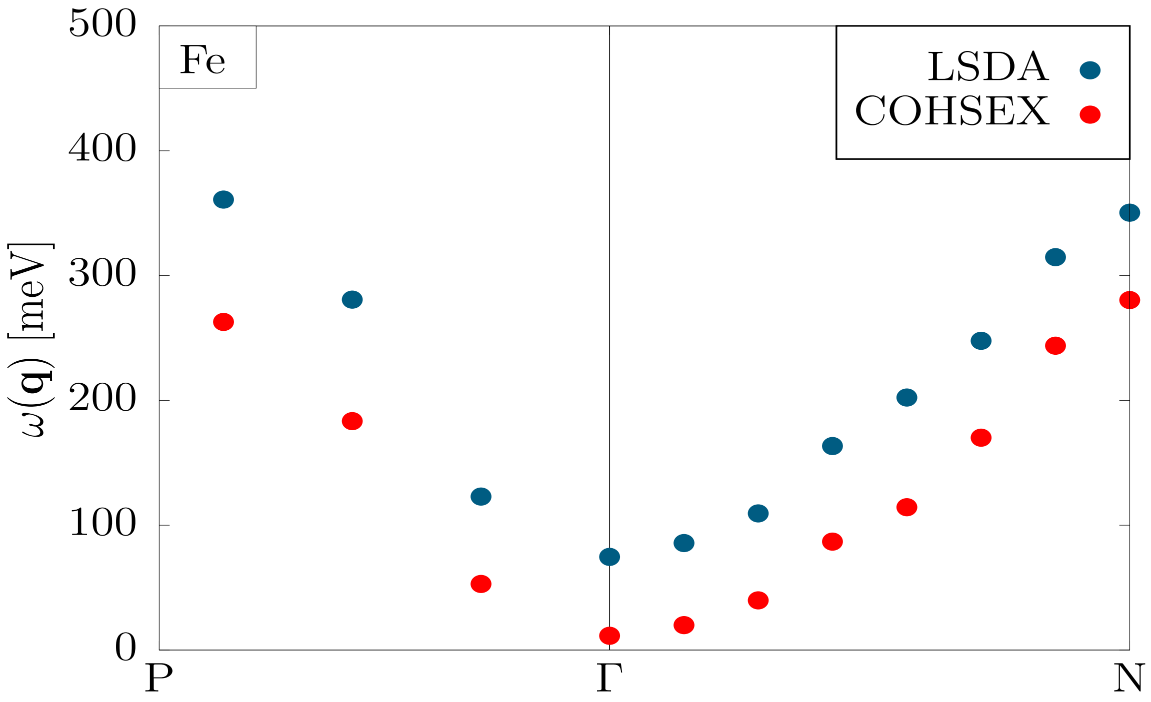
C. Friedrich, M.C.T.D. Müller, S. Blügel, "Handbook of Materials Modelling", Andreoni, Yip (eds), Springer (2019)

VIOLATION OF GOLDSTONE CONDITION



In the absence of spin-orbit coupling, the magnetization of a ferromagnet can be rotated without a cost of energy. ➔ $\lim_{q \to 0} \omega(\mathbf{q}) = 0$

VIOLATION OF GOLDSTONE CONDITION



M. Müller, C. Friedrich, S. Blügel, Phys. Rev. B **94**, 064433 (2016)

In the absence of spin-orbit coupling, the magnetization of a ferromagnet can be rotated without a cost of energy. ➔ $\lim_{q \to 0} \omega(\mathbf{q}) = 0$

INSIGHT FROM HUBBARD MODEL

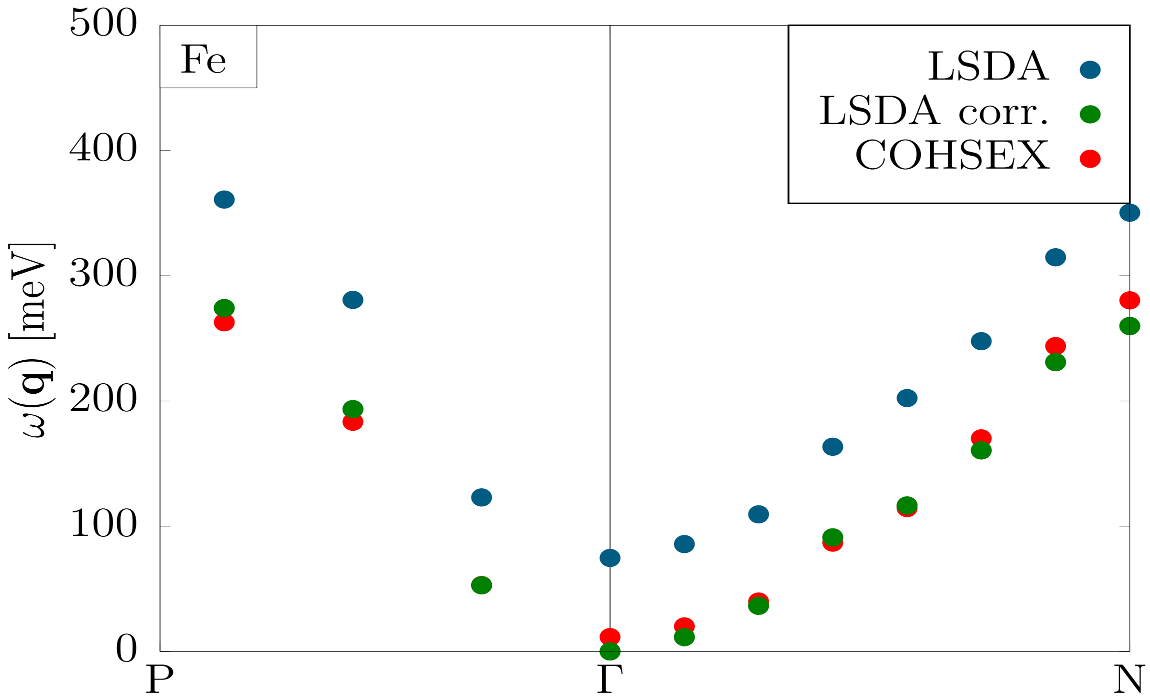
$$H = E_0 \sum_{i\sigma} a^\dagger_{i\sigma} a_{i\sigma} + \sum_{ij,\sigma} t_{ij} a^\dagger_{i\sigma} a_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

Goldstone mode (limit **q**→0, ω→0)

$$1 = \frac{mU}{\Delta_x}$$

Parameter for correcting G_{LSDA}

VIOLATION OF GOLDSTONE CONDITION



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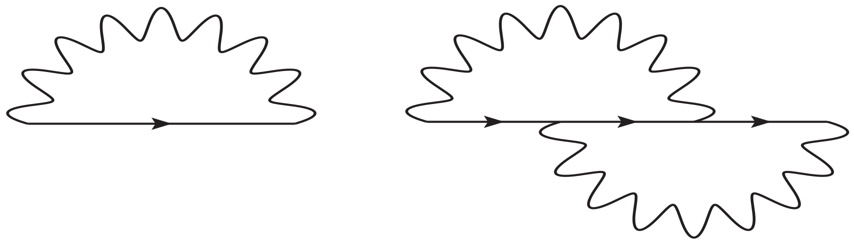
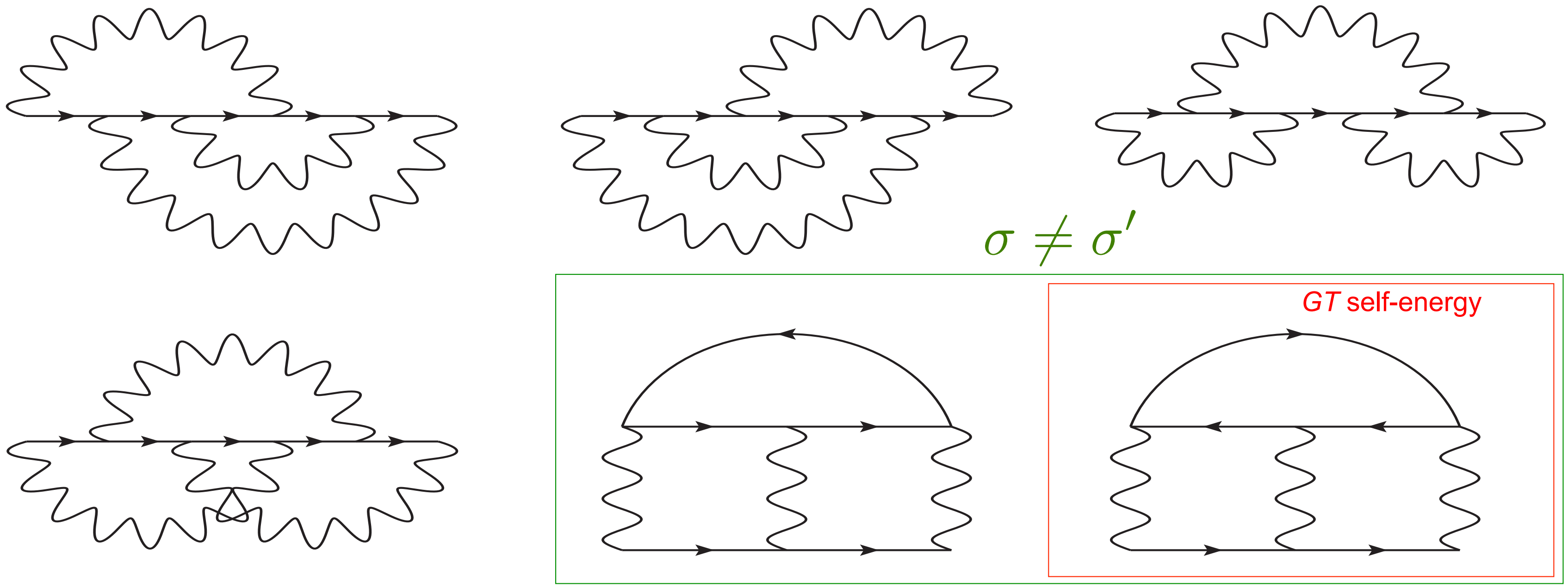
In the absence of spin-orbit coupling, the magnetization of a ferromagnet can be rotated without a cost of energy. ➔ $\lim_{q \to 0} \omega(\mathbf{q}) = 0$

SELF-ENERGY (HEDIN EQUATIONS)

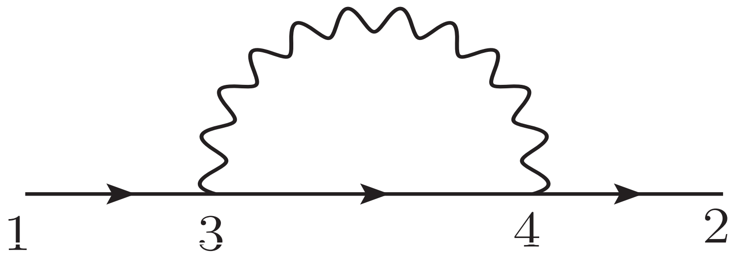
Diagrams up to third order in *W*:



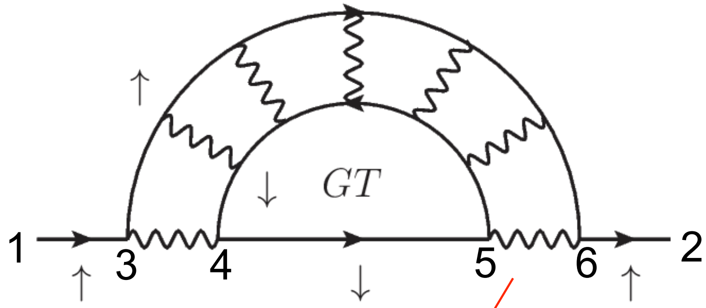


SELF-ENERGY

GW self-energy:

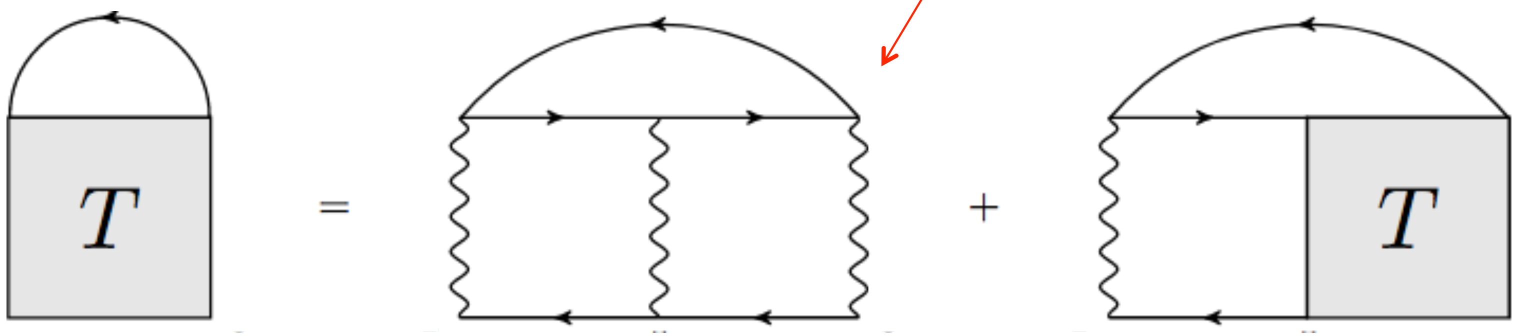


GT self-energy:



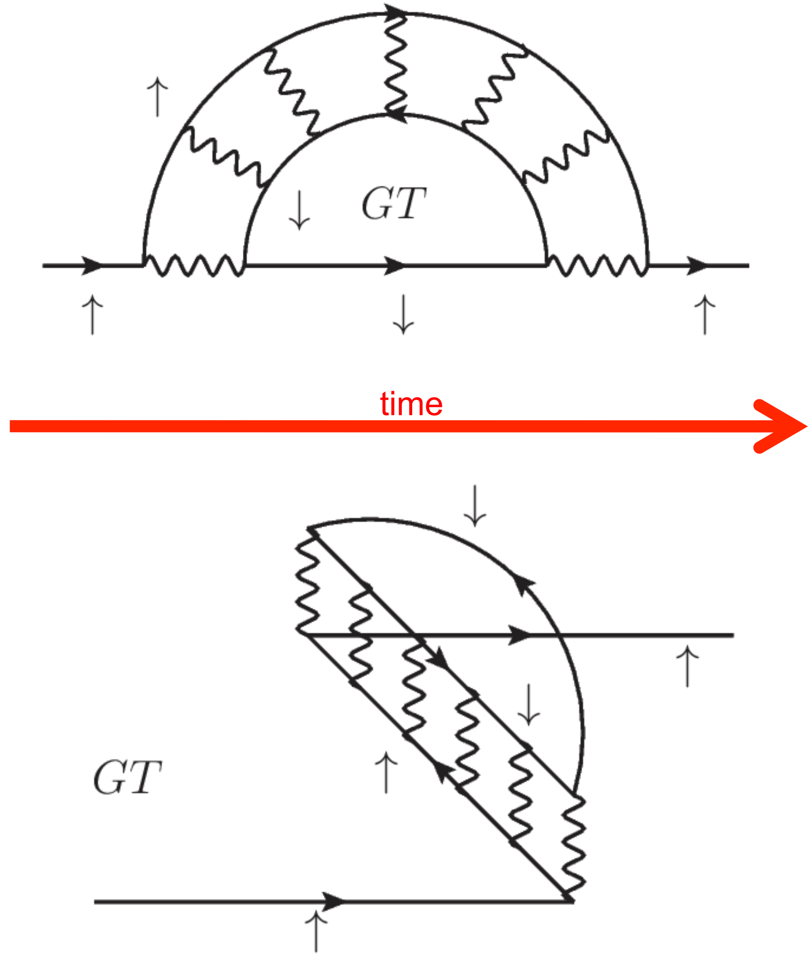
Bethe-Salpeter equation:

starting at 3rd order



SELF-ENERGY

magnon is emitted
before absorbed



magnon is absorbed
before emitted?
electron travels back in time?

IMPLEMENTATION

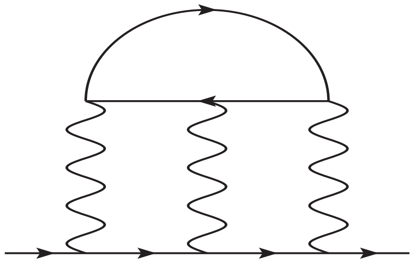
GW approximation:

$$G = G_0 + G_0[\Sigma - v_{\mathrm{xc}}]G$$

GT approximation:

$$G = G_0 + G_0 \Sigma G$$

COHSEX
or
LSDA corr.

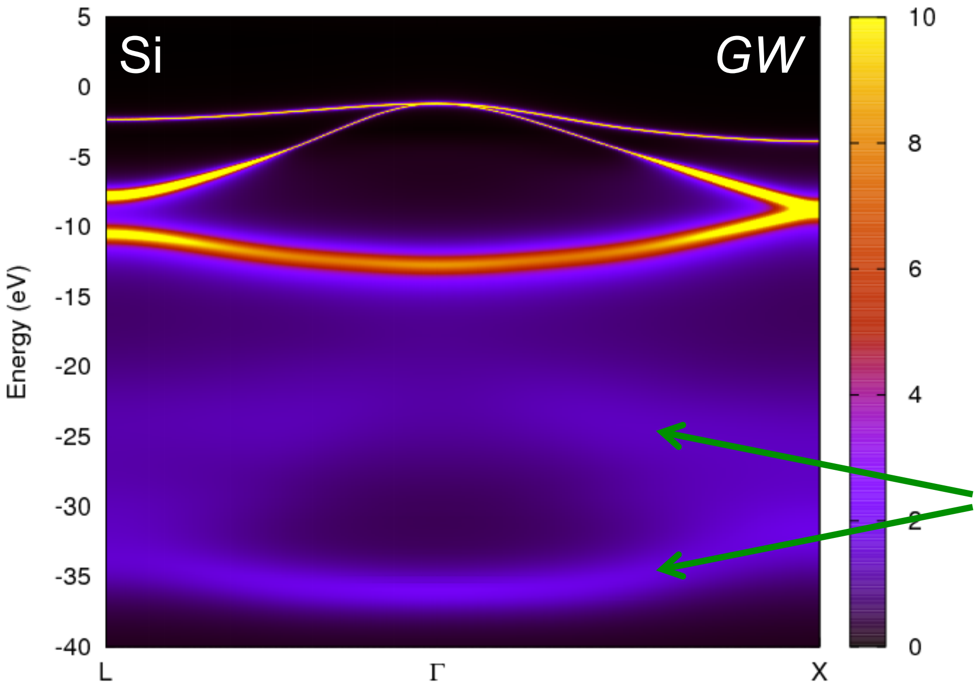


$+ \quad \dots$

$$S^{\sigma}(\omega, \mathbf{k})$$
$$= \frac{1}{\pi} \sum_{n} \mathrm{Im} \frac{1}{\omega - \epsilon^{\sigma}_{\mathbf{k}n} - \Sigma^{\sigma}_{\mathbf{k}n}(\omega) + \Delta_v}$$

alignment of chemical potential

SATELLITES

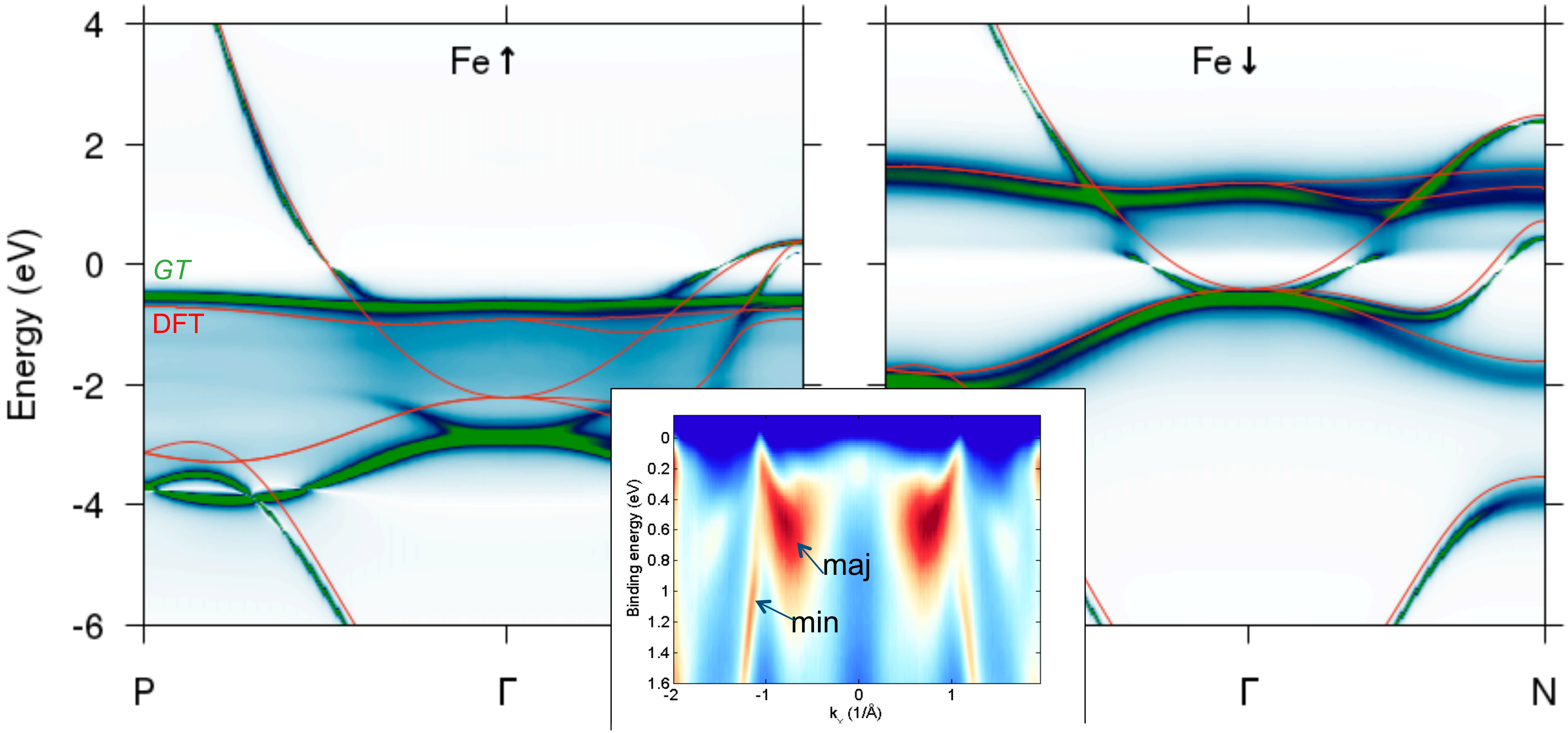


Spectral function

Plasmon satellites
$\omega_0 \approx 23$ eV

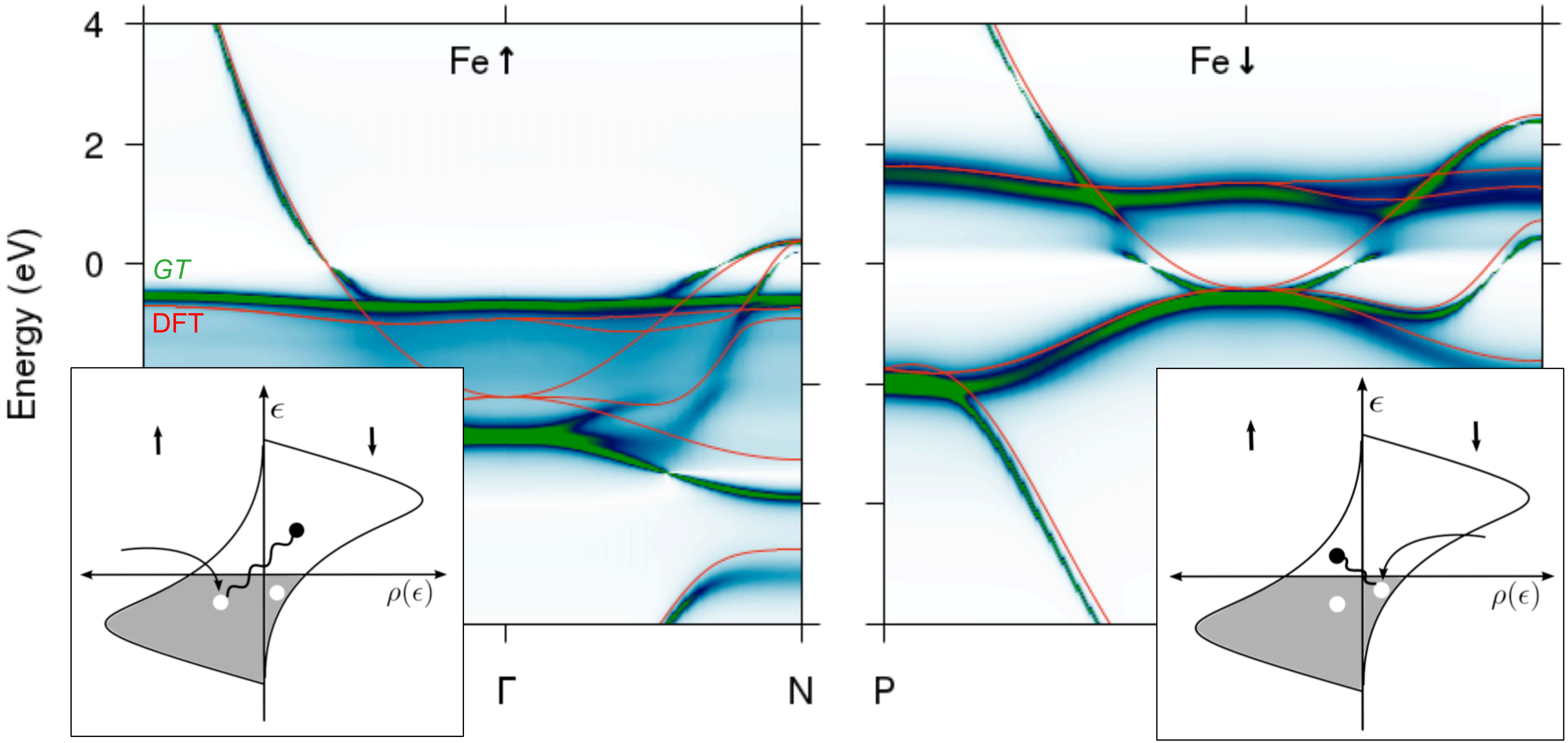
Magnon satellites?
$\omega_0 \sim 0\text{-}0.3$ eV

IRON BAND STRUCTURE

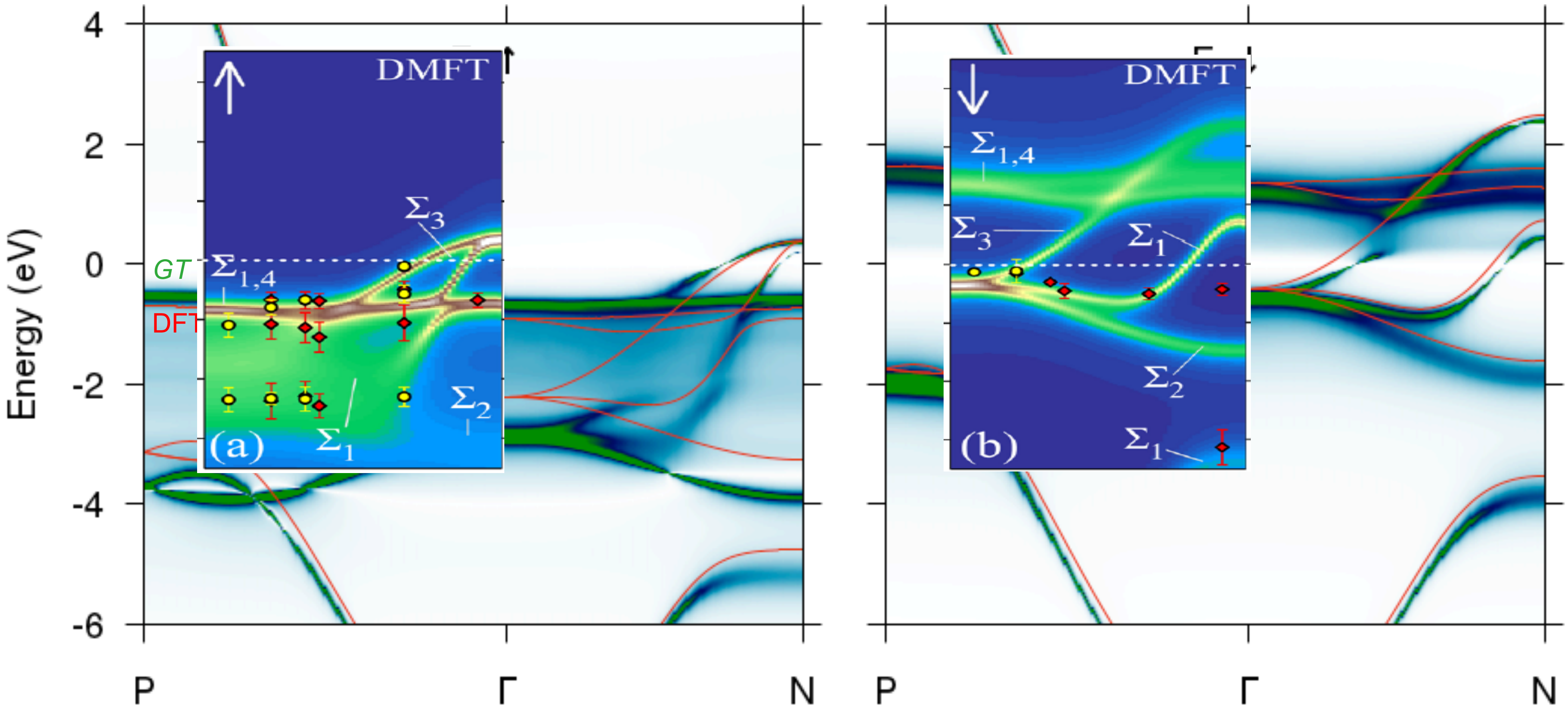


E. Mlynczak, L. Plucinski, unpublished

IRON BAND STRUCTURE

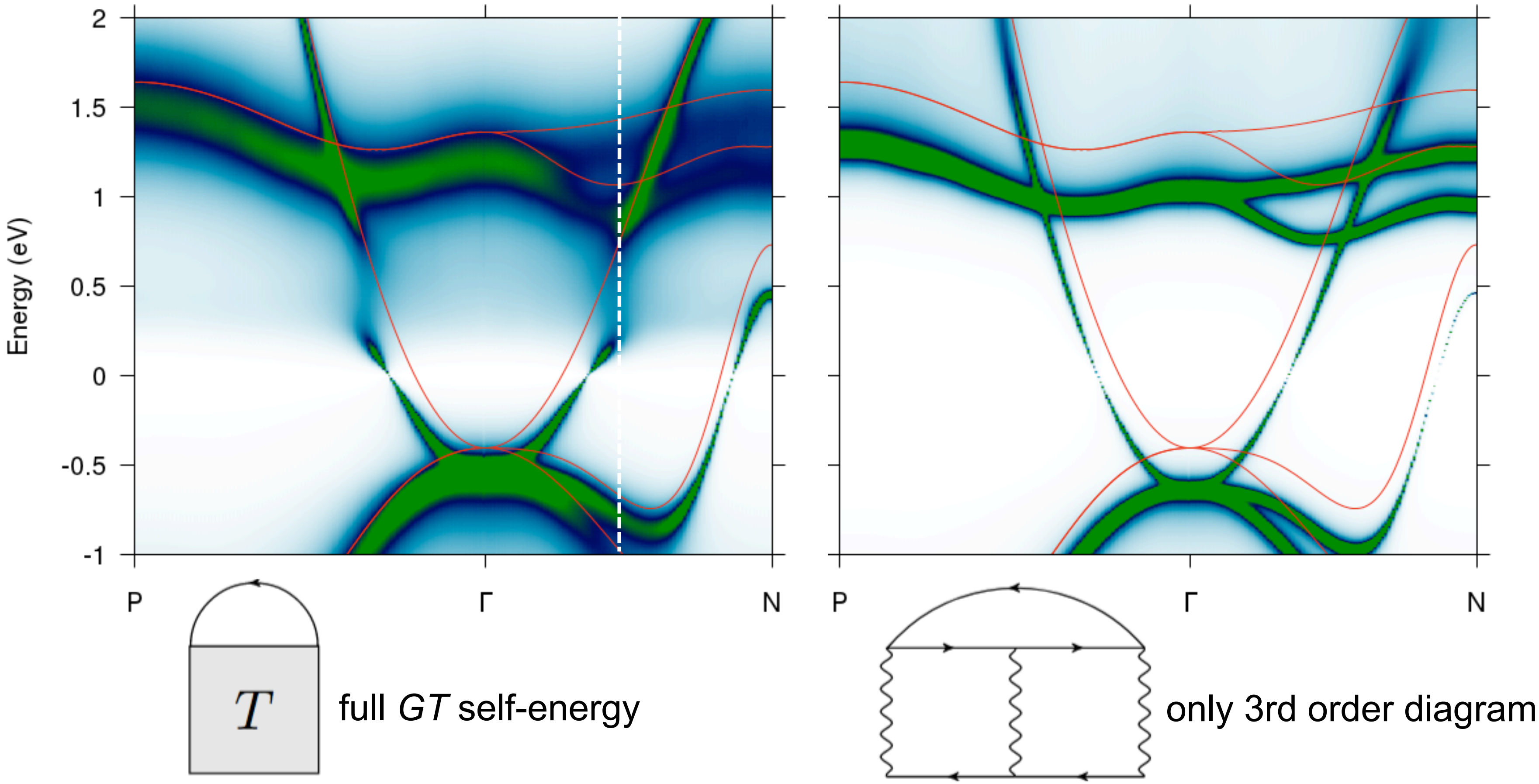


IRON BAND STRUCTURE

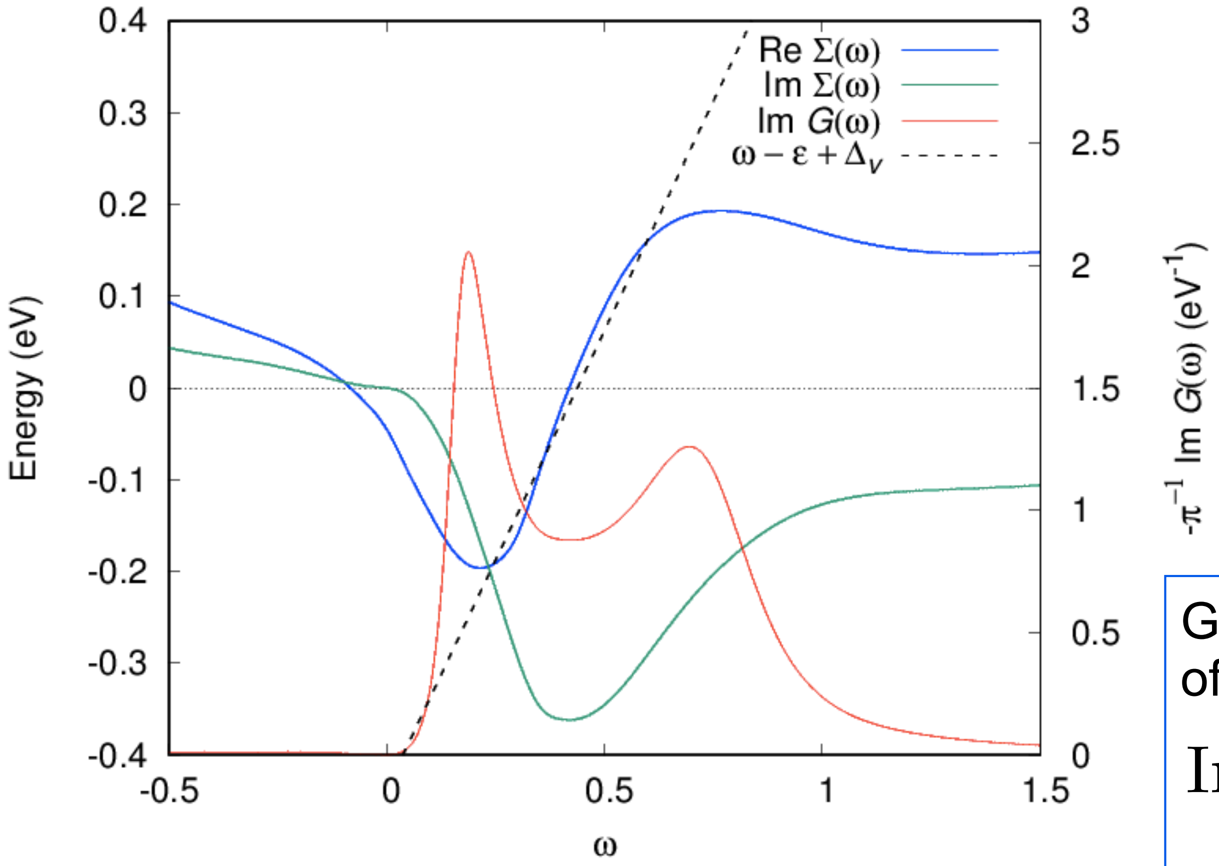


DMFT: J. Sánchez-Barriga et al., PRL **103**, 267203 (2009)

ANOMALY SPIN DOWN



IRON BAND ANOMALY (SPIN DOWN)

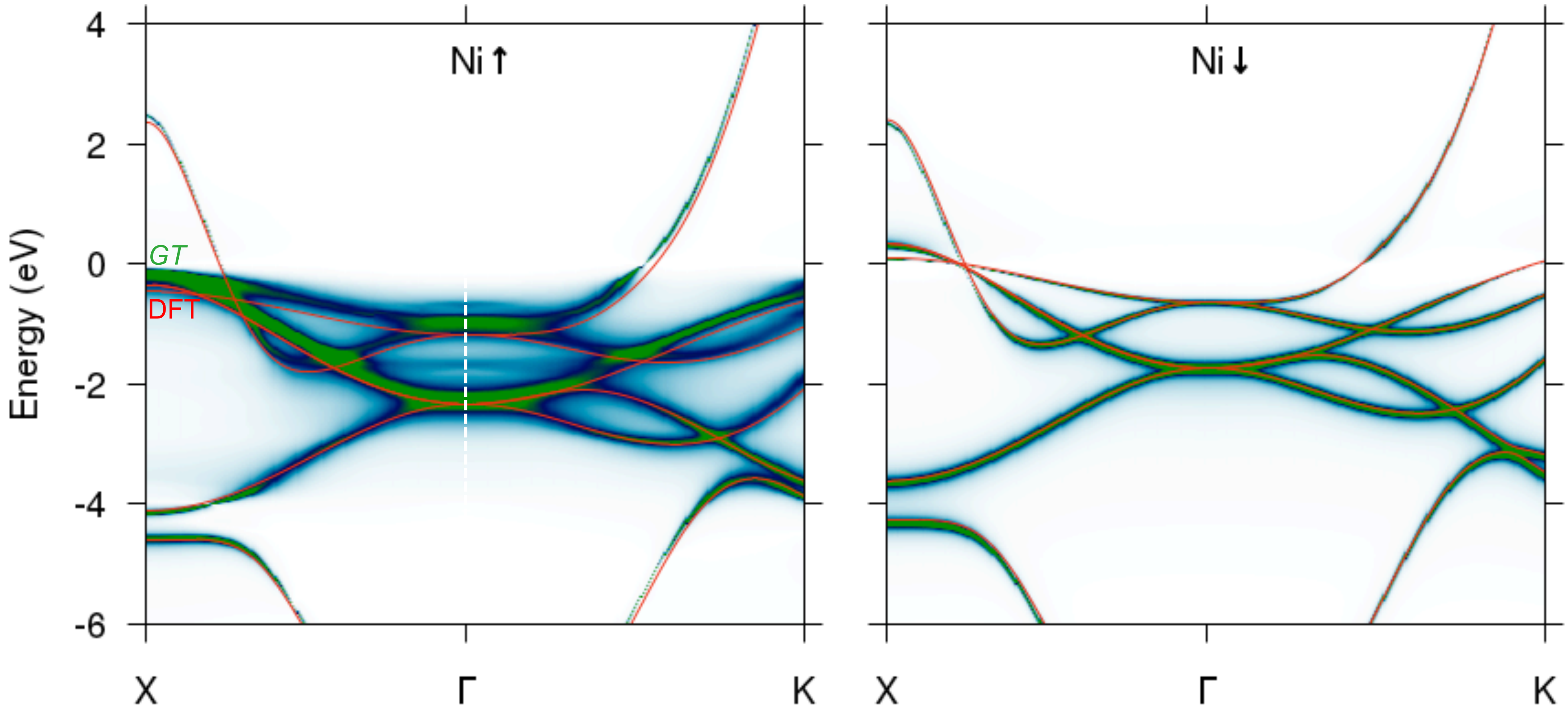


Graphical solution
of the Dyson equation:

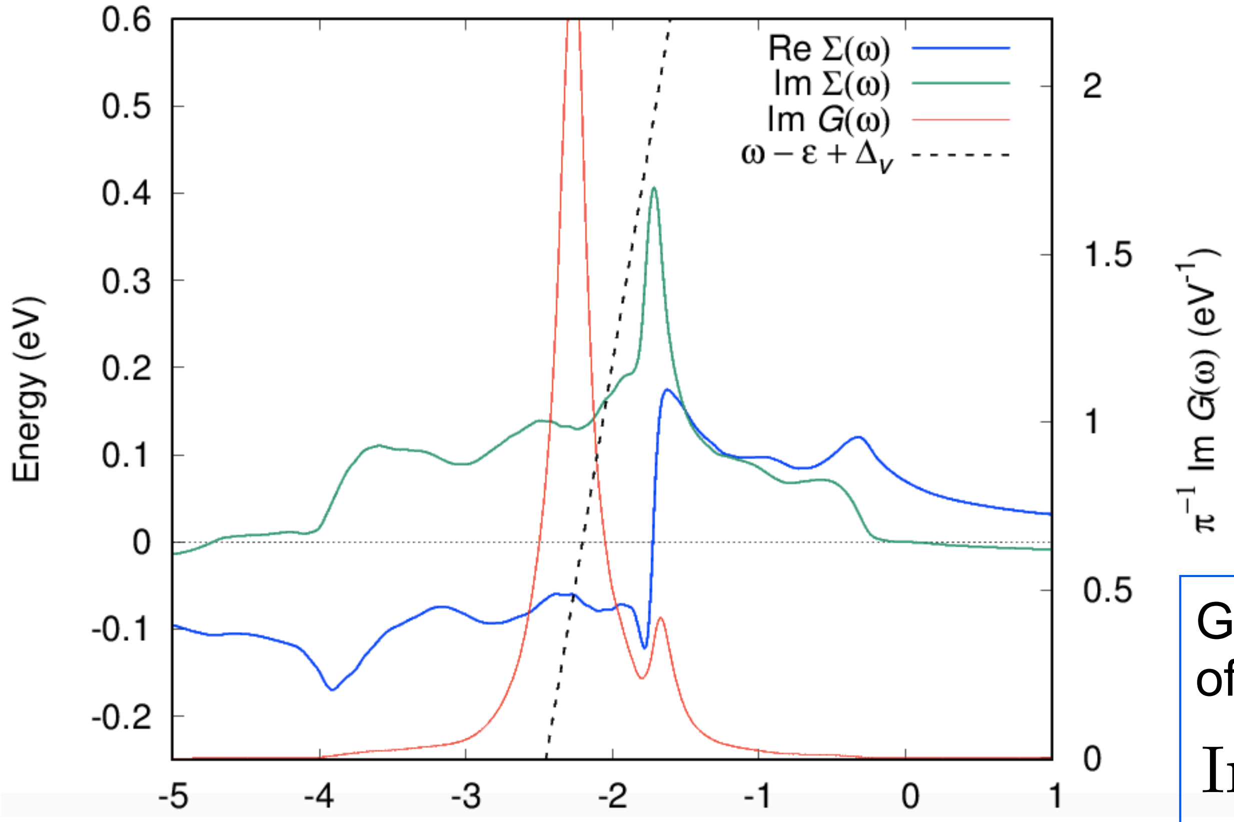
$$\mathrm{Im} G^{\sigma}_{\mathbf{k}n}(\omega)$$

$$= \mathrm{Im} \frac{1}{\omega - \epsilon^{\sigma}_{\mathbf{k}n} - \Sigma^{\sigma}_{\mathbf{k}n}(\omega) + \Delta_v}$$

NICKEL BAND STRUCTURE



NICKEL BAND ANOMALY (SPIN UP)

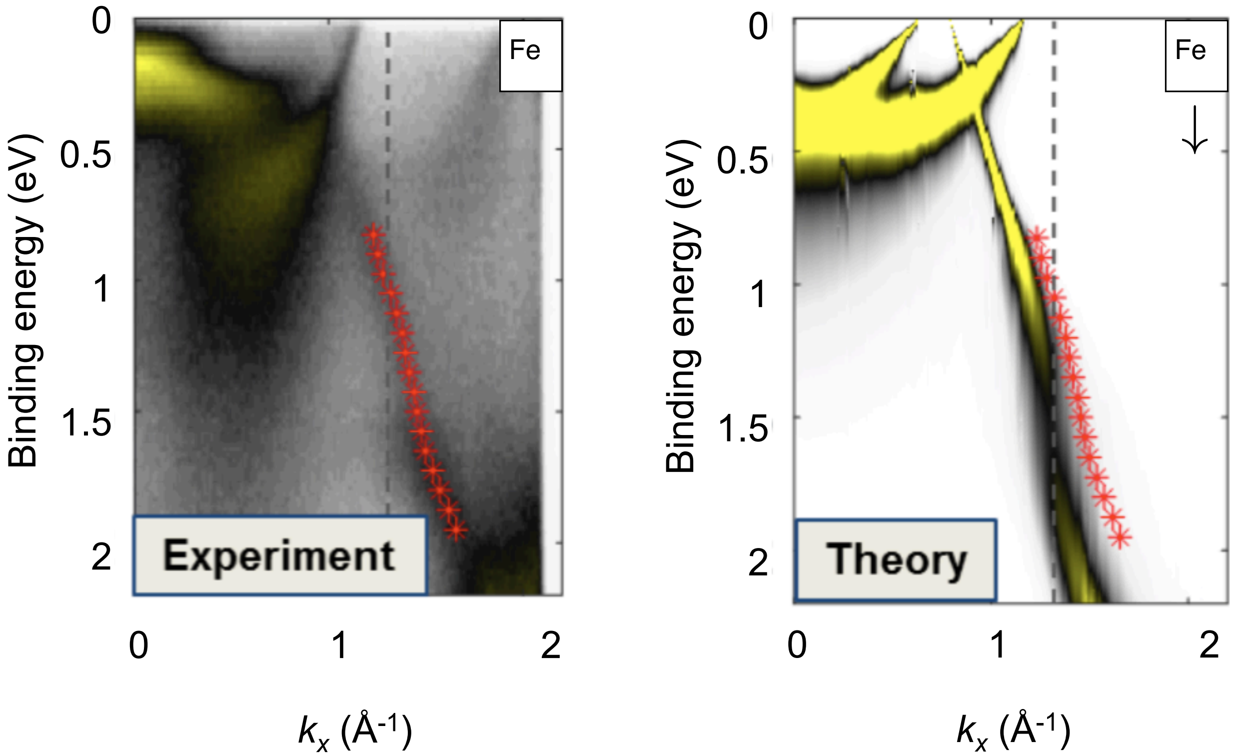


Graphical solution
of the Dyson equation:

$$\mathrm{Im}G^{\sigma}_{\mathbf{k}n}(\omega)$$
$$= \mathrm{Im}\frac{1}{\omega - \epsilon^{\sigma}_{\mathbf{k}n} - \Sigma^{\sigma}_{\mathbf{k}n}(\omega) + \Delta_v}$$

HIGH-ENERGY BAND ANOMALY (IRON)

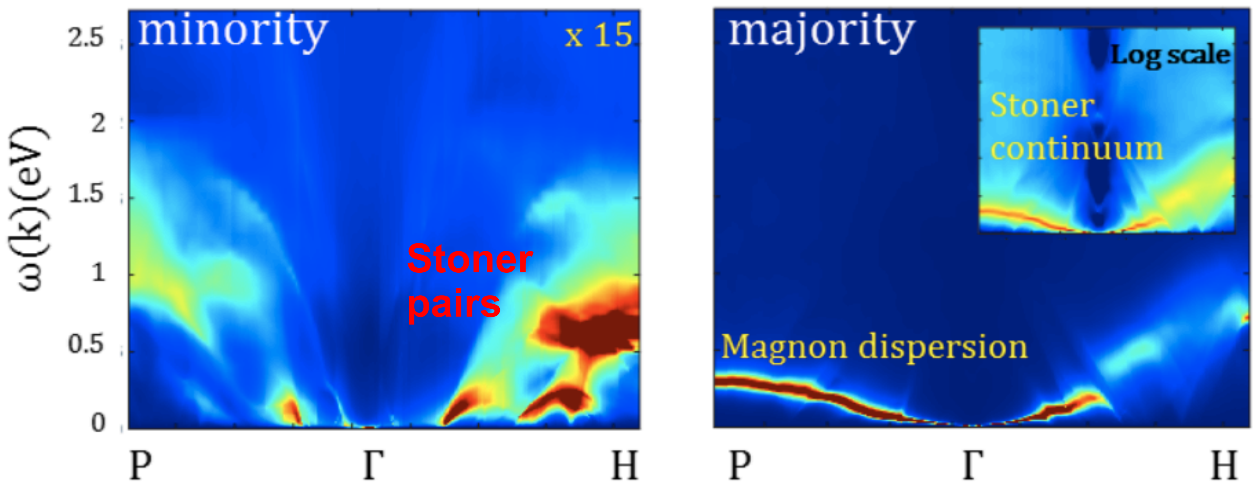
ARPES (Mlynczak *et al.*, Jülich)



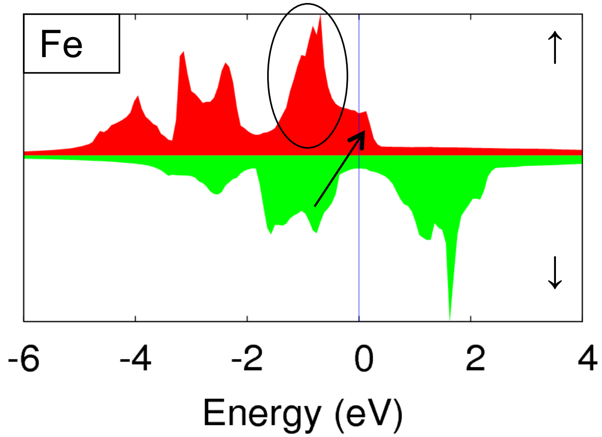
Mlynczak *et al.*, Nature Communications 10, 505 (2019)

HIGH-ENERGY BAND ANOMALY (IRON)

JÜLICH Forschungszentrum



Mlynczak *et al.*, Nature Communications 10, 505 (2019)



≈0.8eV ≈0.7eV

$$\frac{1}{2\pi}\int_{-\infty}^{\infty}\frac{1}{\omega-\omega'-\epsilon-\mathrm{i}\eta}\,\frac{1}{\omega'-\epsilon_{\mathrm{M}}-\mathrm{i}\eta}\,d\omega'$$

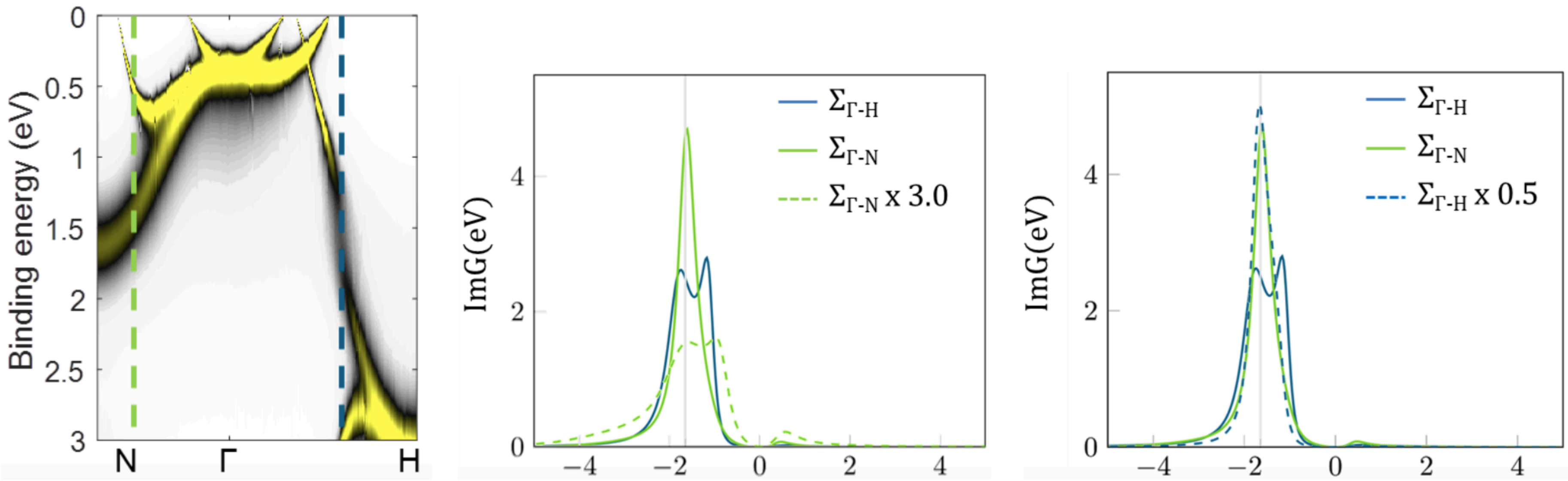
$$=\frac{\mathrm{i}}{\omega-(\epsilon+\epsilon_{\mathrm{M}})-\mathrm{i}\eta}$$

≈1.5eV

HIGH-ENERGY BAND ANOMALY (IRON)

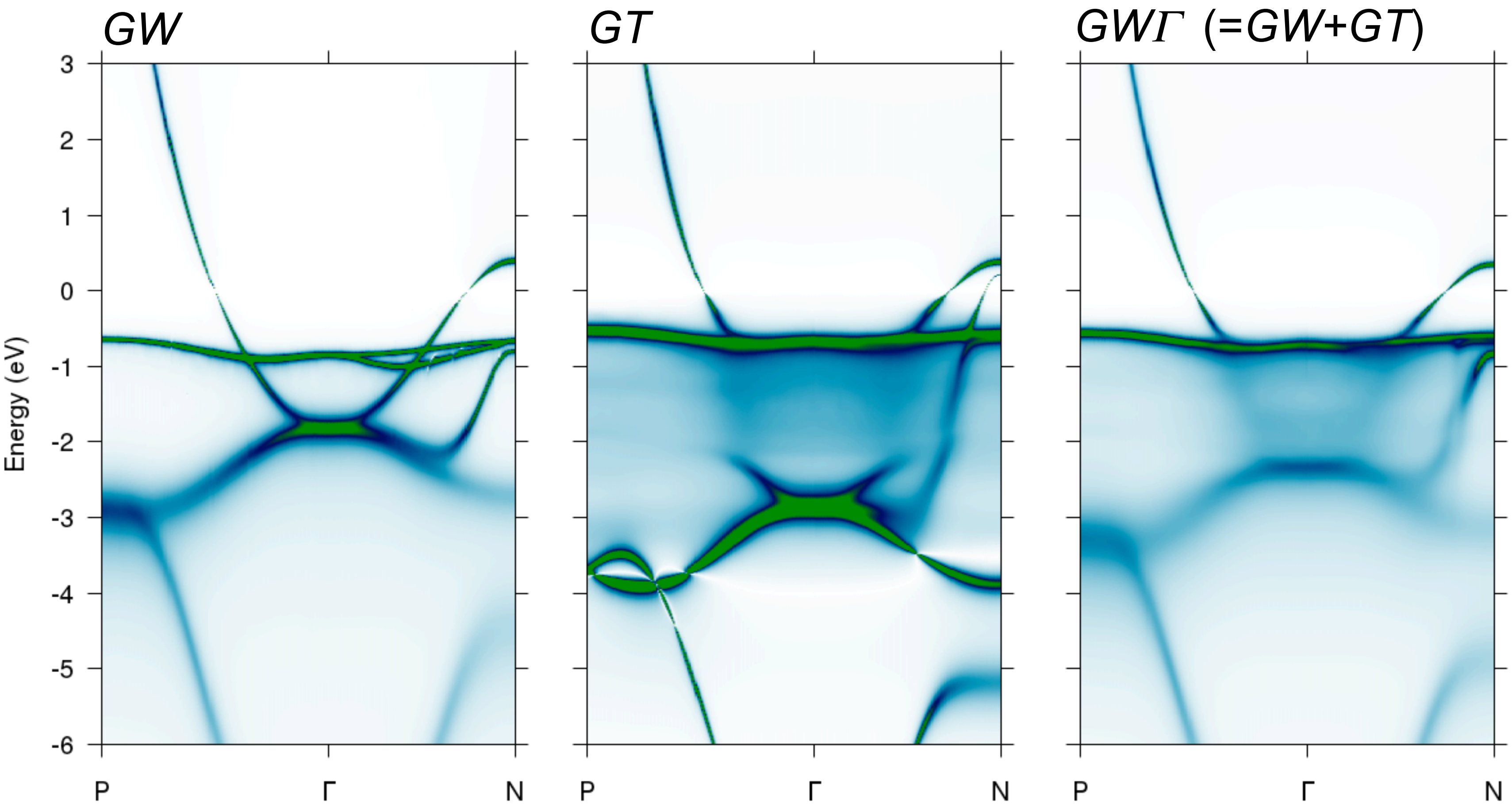
Mlynczak *et al.*, Nature Communications 10, 505 (2019)

Importance of **k** dependence



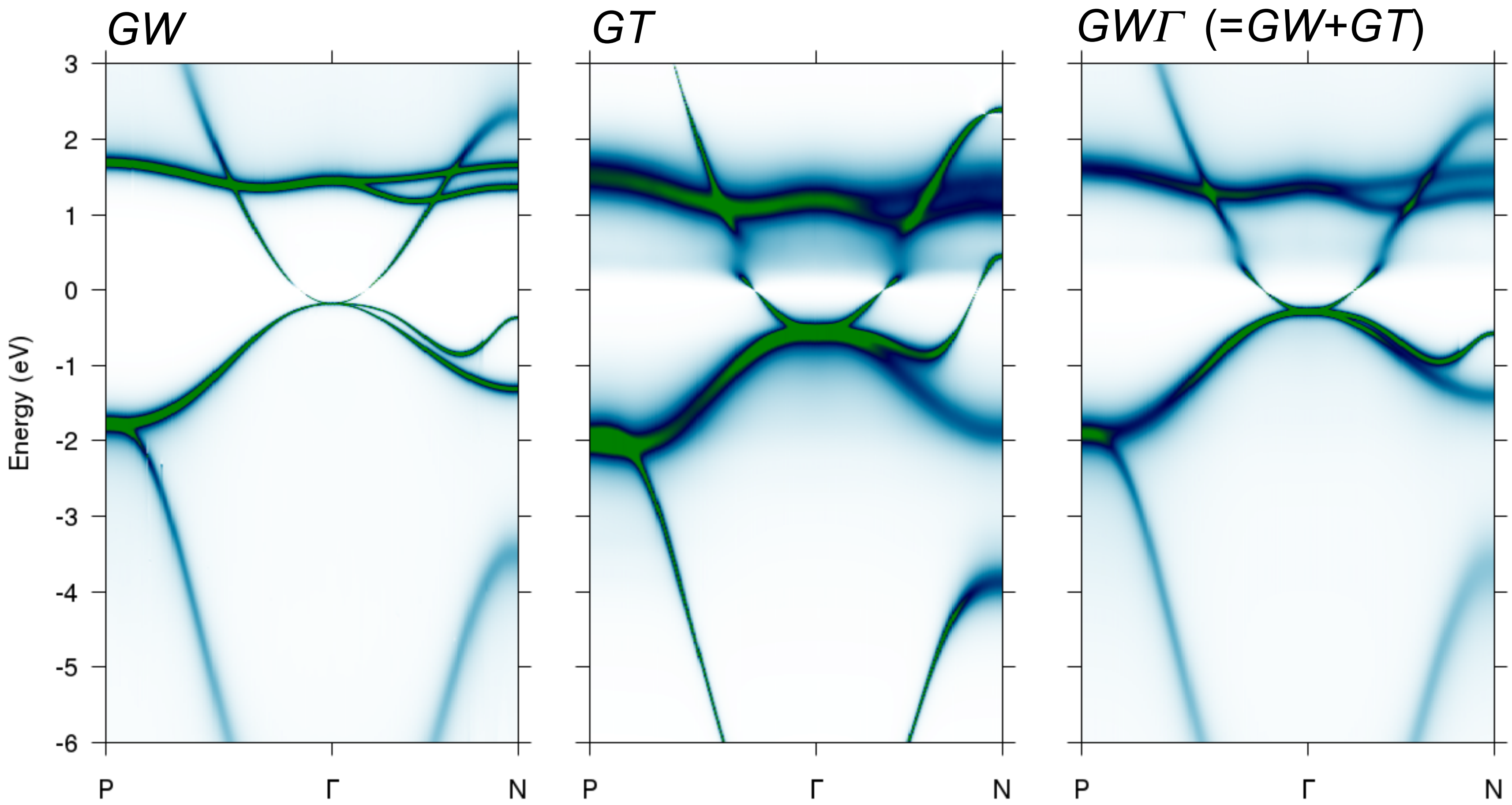
IRON BAND STRUCTURE (SPIN UP)

JÜLICH
Forschungszentrum



IRON BAND STRUCTURE (SPIN DOWN)

JÜLICH Forschungszentrum



SUMMARY

JÜLICH
Forschungszentrum

- Calculation of spin excitations (spin waves and Stoner excitations) implemented within FLAPW through the solution of the Bethe-Salpeter equation.
- Goldstone condition violated in the limit $\mathbf{q}\to 0$ and $\omega\to 0$ due to inconsistency of Green functions (LSDA vs. self-consistent)
- Static COHSEX approximation for the self-energy recovers correct dispersion of Goldstone mode. Alternative: corrected LSDA approach.
- Electron-magnon scattering described by *GT* self-energy.
- Strong spin asymmetry of lifetime broading in agreement with experiment. Majority *d* bands strongly renormalized (quasiparticle character lost) due to coupling to many-body spin excitations.
- Band anomalies due to many-body renormalization through coupling to spin-wave and Stoner excitations (the latter seen in recent ARPES experiment).

http://spex.readthedocs.org SPEX

ACKNOWLEDGMENTS

Mathias C.T.D. Müller

Stefan Blügel

Experiments:

Ewa Mlynczak

Lukas Plucinski