

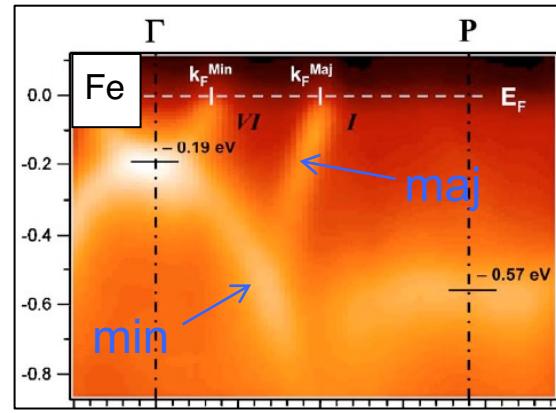
ELECTRON-PLASMON AND ELECTRON-MAGNON SCATTERING IN ELEMENTARY FERROMAGNETS FROM FIRST PRINCIPLES: GWT SELF-ENERGY

CHRISTOPH FRIEDRICH

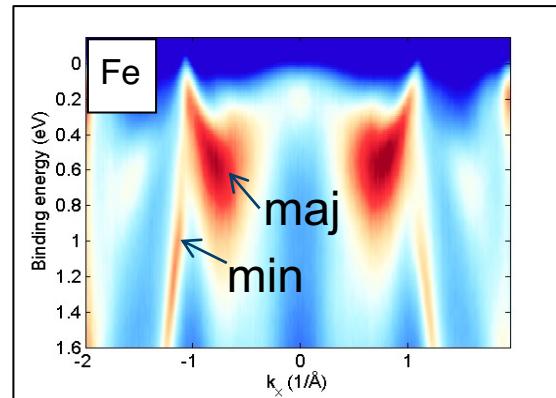
PETER GRÜNBERG INSTITUTE AND INSTITUTE FOR ADVANCED SIMULATION
FORSCHUNGSZENTRUM JÜLICH AND JARA, 52425 JÜLICH, GERMANY

ARPES MEASUREMENTS

Spin asymmetry in spectra

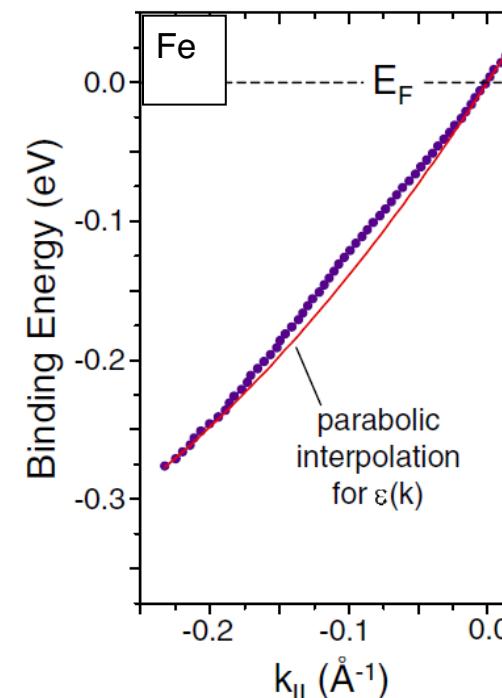


Schäfer *et al.*, PRL **72**, 155115 (2005)

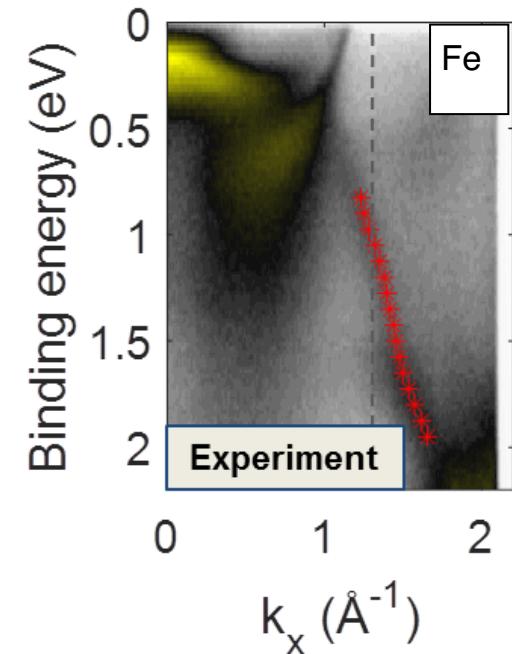


E. Mlynaczak, L. Plucinski, unpublished

Anomalies in band dispersion of iron

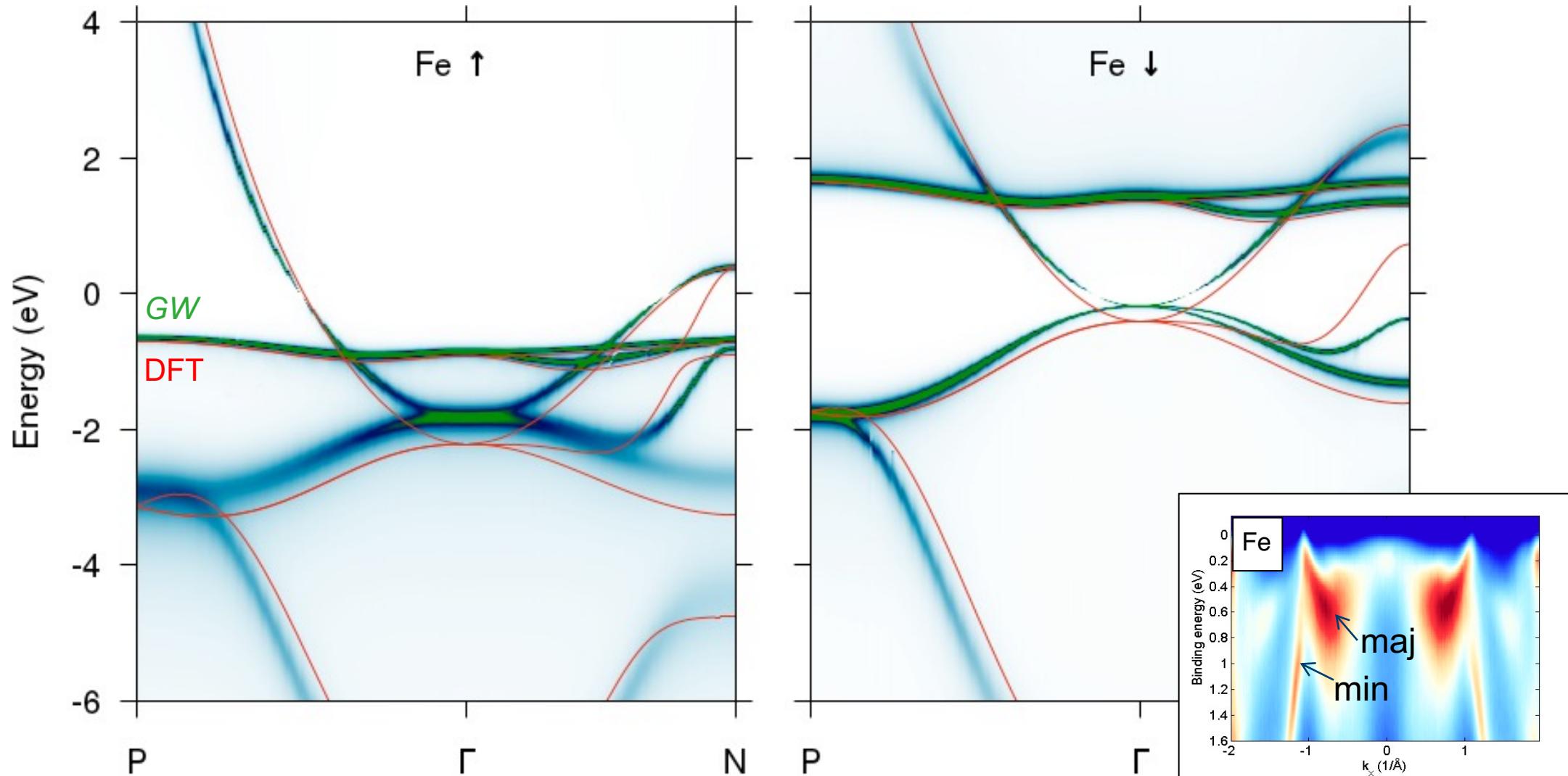


Schäfer *et al.*, PRL **92**, 097205 (2004)



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

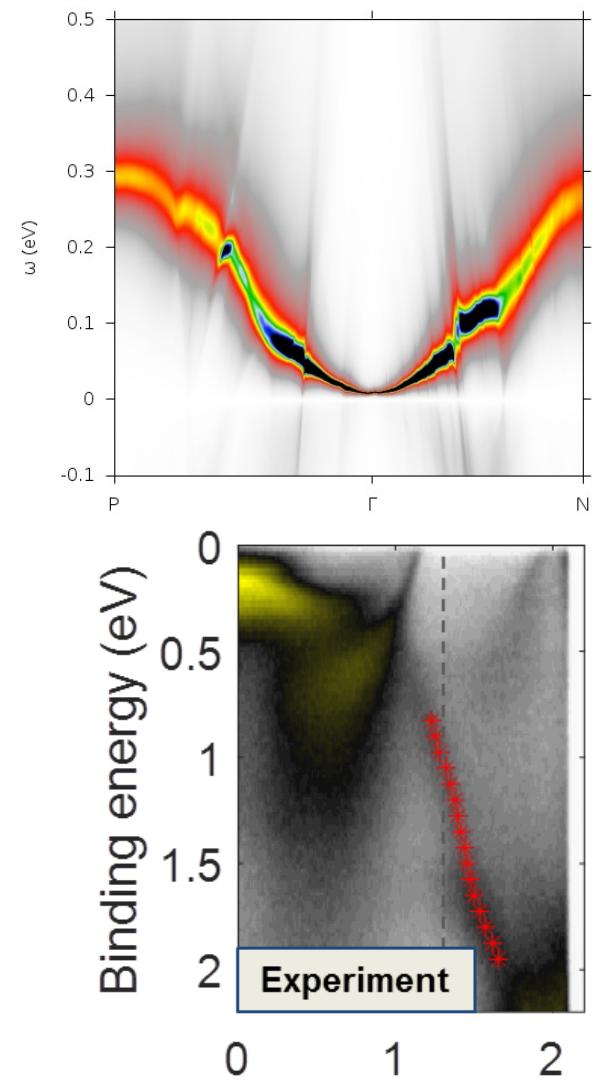
IRON GW CALCULATION



E. Mlynczak, L. Plucinski, unpublished

OVERVIEW

- Many-body spin excitations
 - Magnetic response function / T matrix
 - Bethe-Salpeter equation
 - Implementation (Wannier functions)
 - Examples
 - Goldstone violation
- Electron-magnon scattering
 - Iteration of Hedin equations (GWT self-energy)
 - Results for iron
 - comparison to DMFT and experiment
 - lifetime broadening, renormalizations, band anomalies
 - violation of causality (GT)
 - magnetic moments, d -band width, exchange splittings
- Conclusions



MAGNETIC RESPONSE FUNCTION

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

Spin-orbit coupling neglected

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

$$\begin{aligned} B_x, B_y &\rightarrow B^+ = B_x + iB_y \\ B^- &= B_x - iB_y \end{aligned}$$

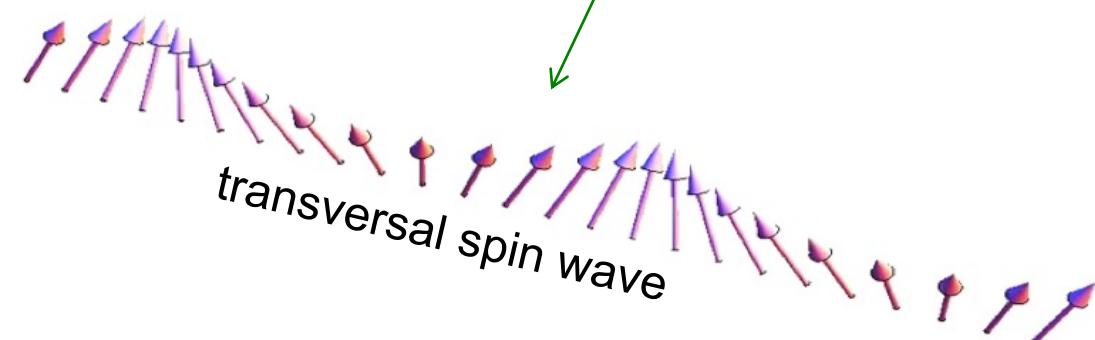
Circularly polarized B field

MAGNETIC RESPONSE FUNCTION

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

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

Spin-orbit coupling neglected



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

MAGNETIC RESPONSE FUNCTION

$$R^{+-}(1, 2) = \frac{\delta\sigma^+(1)}{\delta B^+(2)}$$

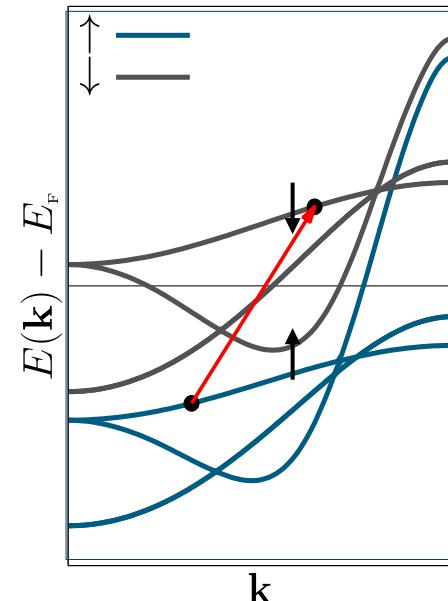
$$\sigma^+(1) = -i \sum_{\alpha, \beta} \sigma_{\beta\alpha}^+ G_{\alpha\beta}(1, 1^+)$$

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

Dyson equation Chain rule

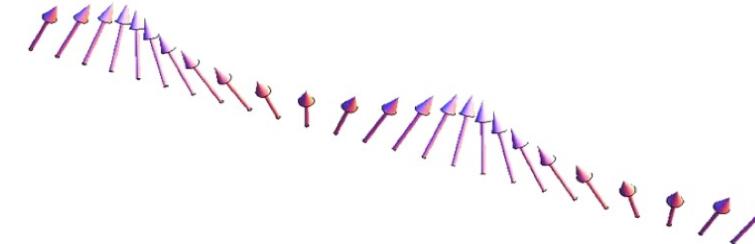
Stoner excitation

- Single particle
- Spin-flip
- Large energy scale (\sim eV)



Spin-wave excitation

- Collective excitation
- Small energy scale (\sim meV)



BETHE-SALPETER EQUATION

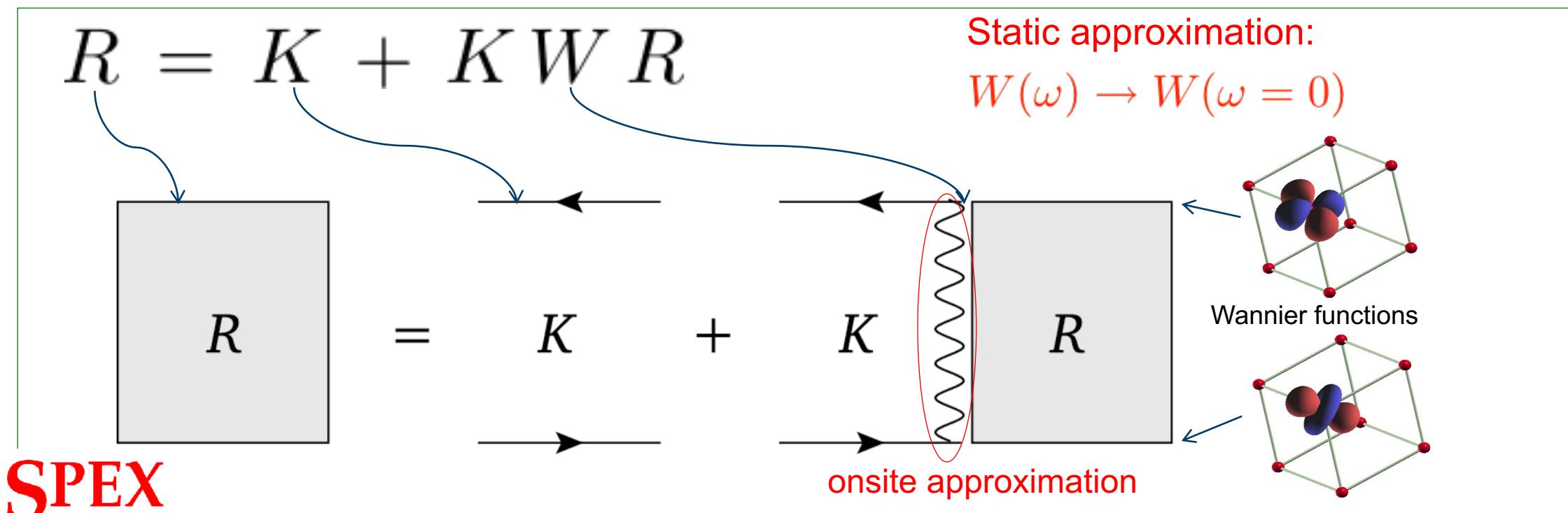
Self-energy

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

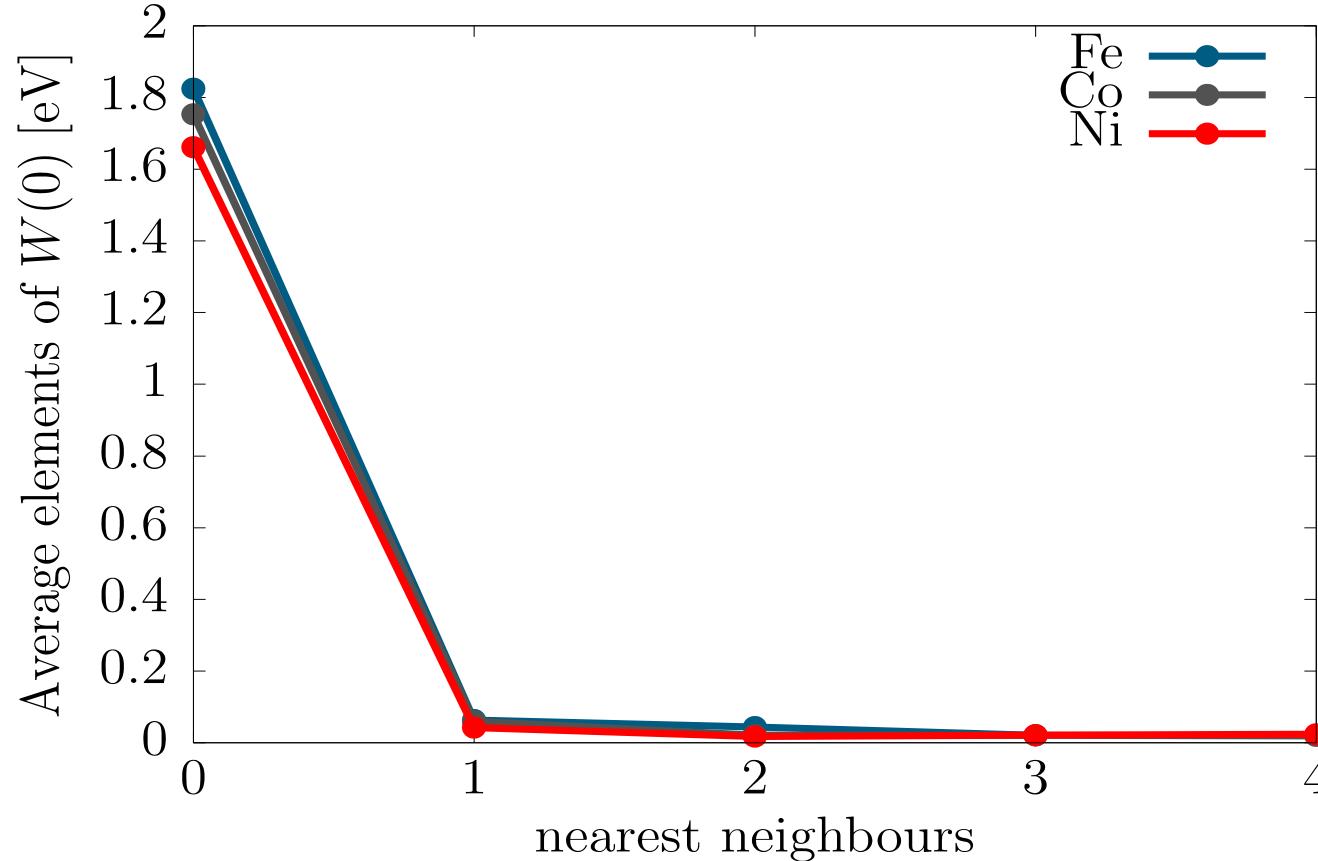
$$\frac{\delta \Sigma}{\delta G} = iW + iG \frac{\delta W}{\delta G}$$

no SOC

Bethe-Salpeter equation for spin excitations

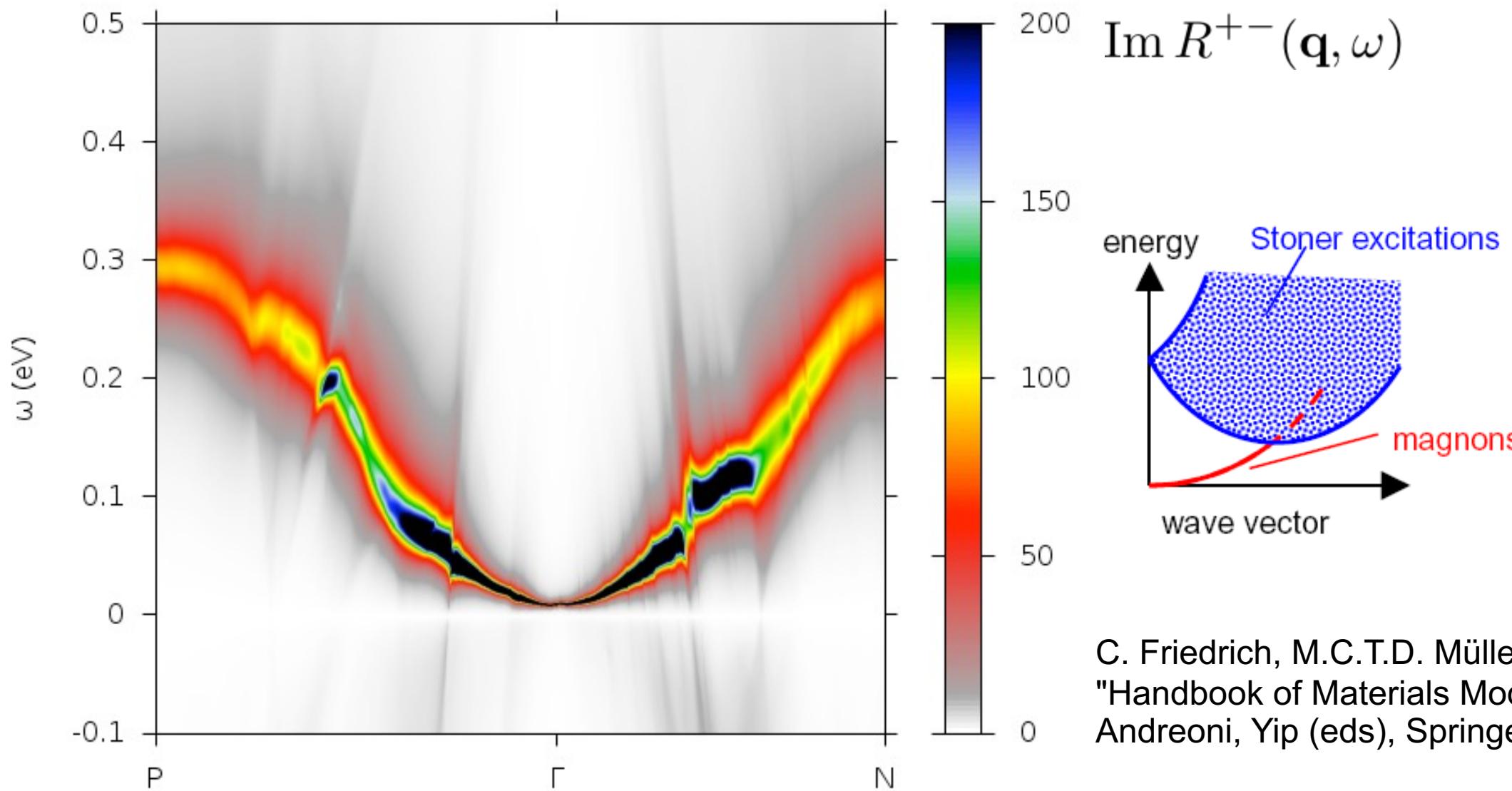


SPATIAL DEPENDENCE W



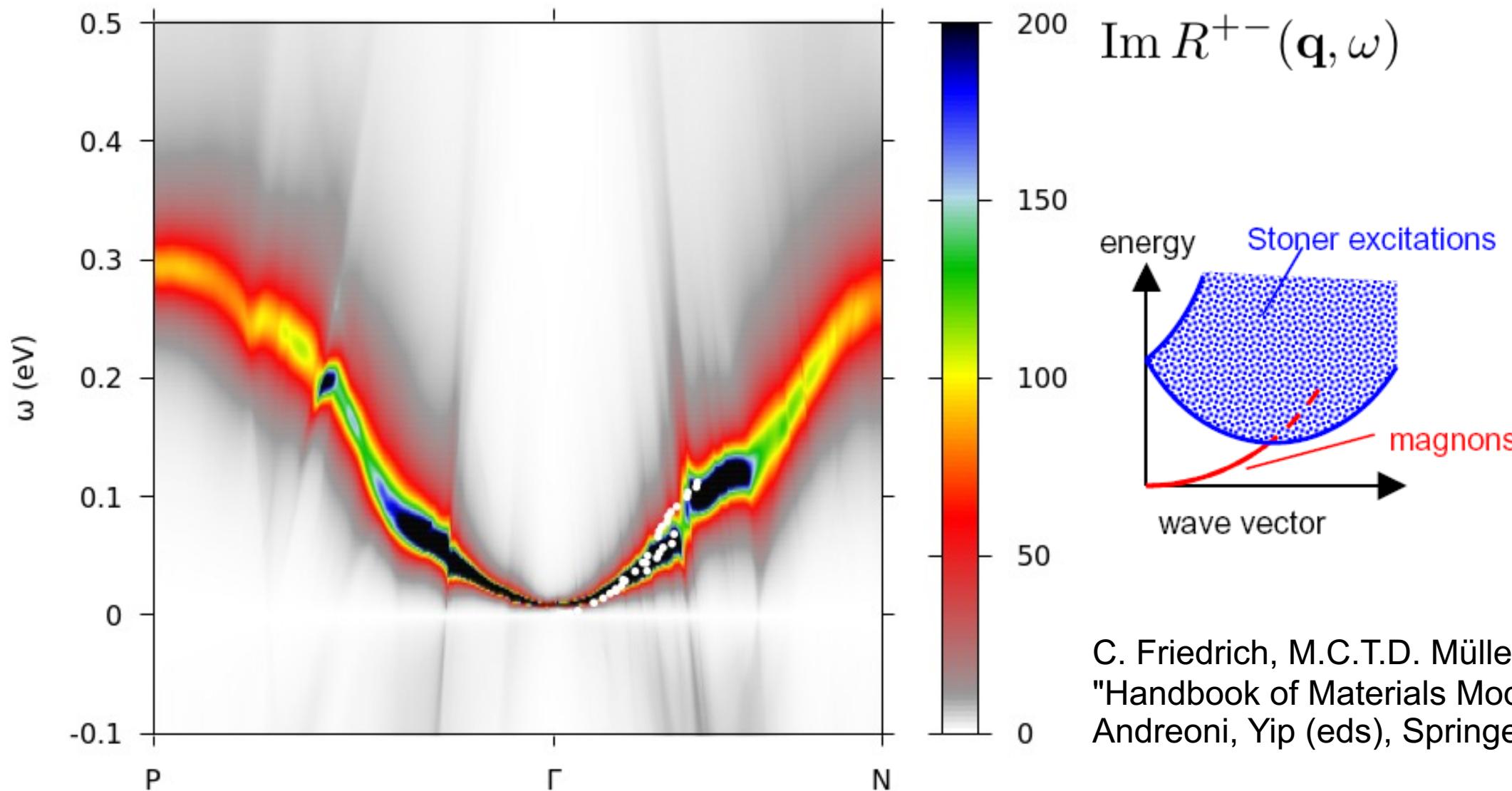
Largest contribution from the onsite interaction (~98%)

EXAMPLE: BCC IRON



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

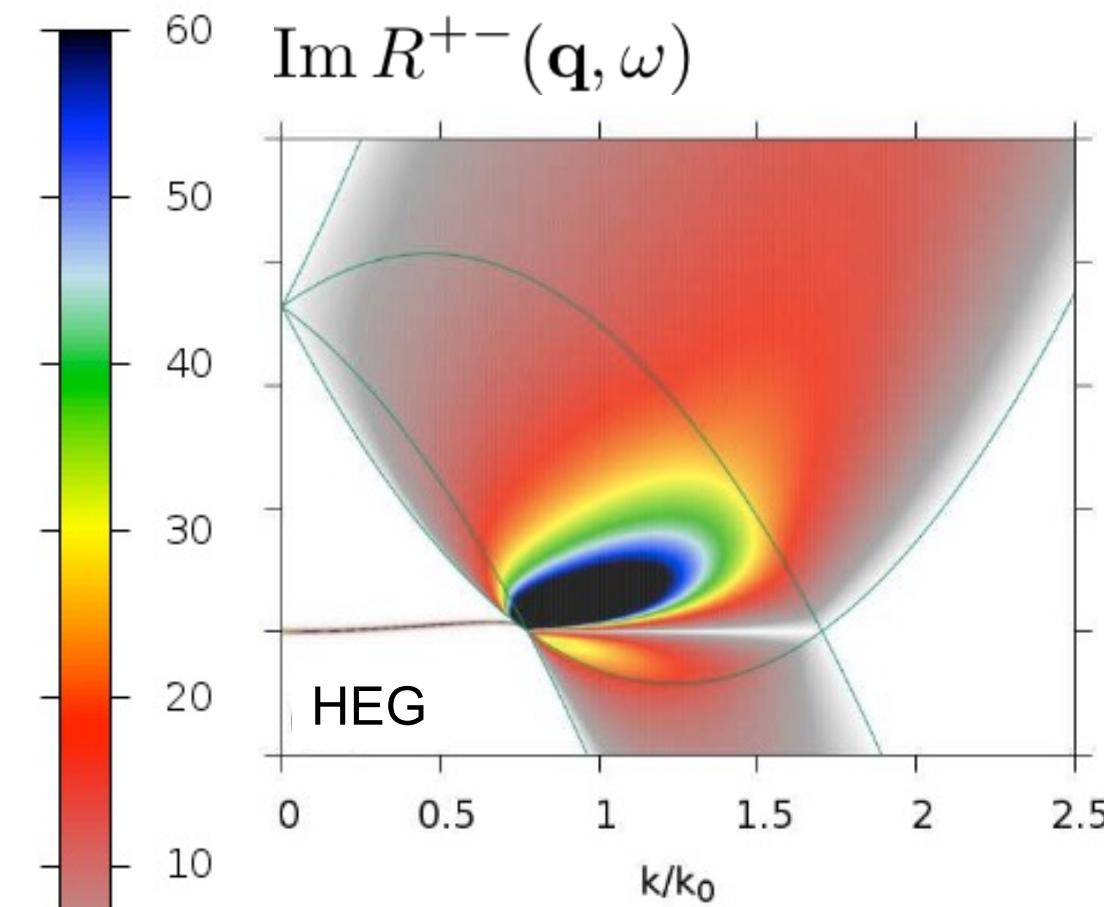
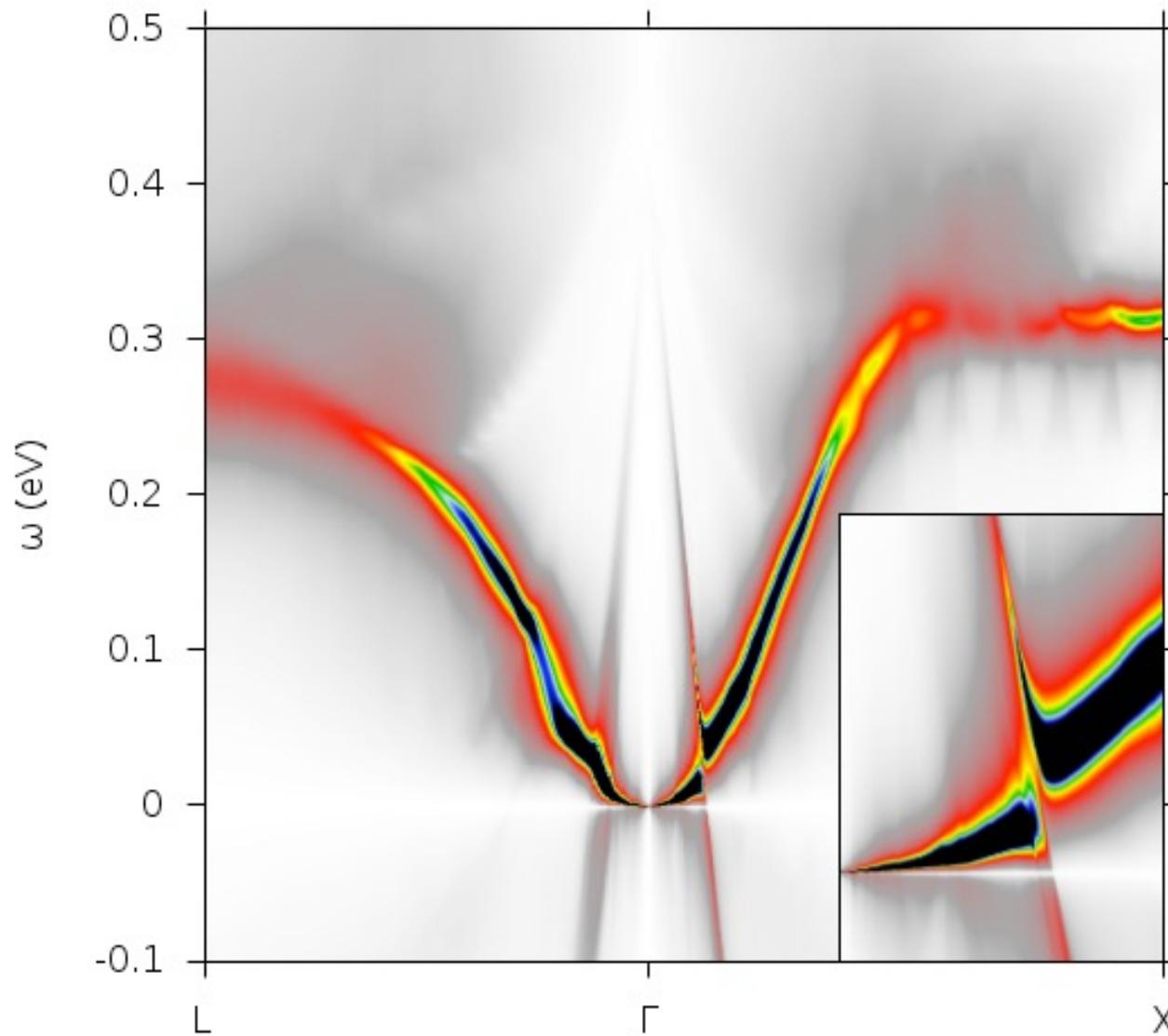
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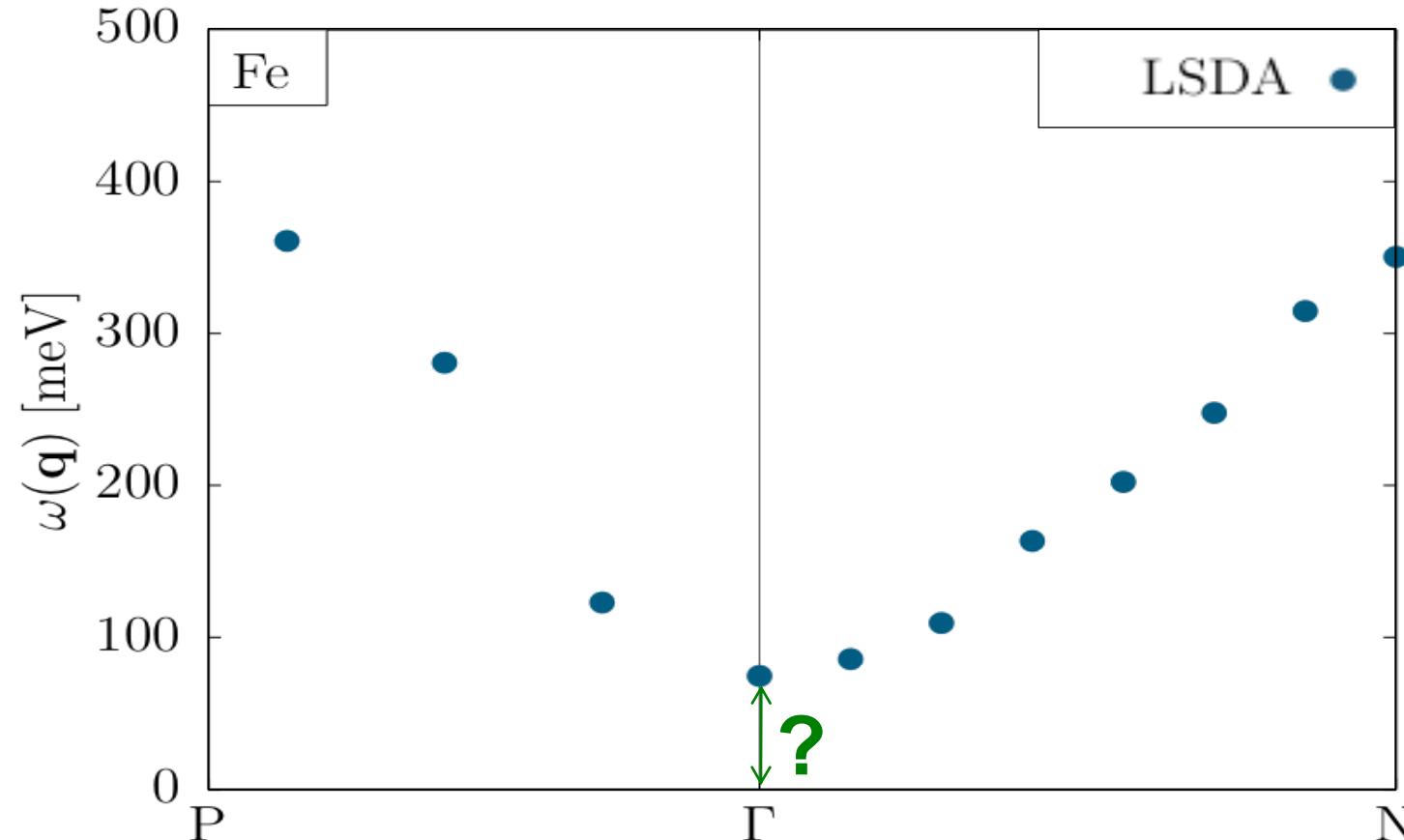
Experiments (white circles) Collins et al. PR 179, 417; Mook et al. PRB 7, 336

EXAMPLE: FCC NICKEL



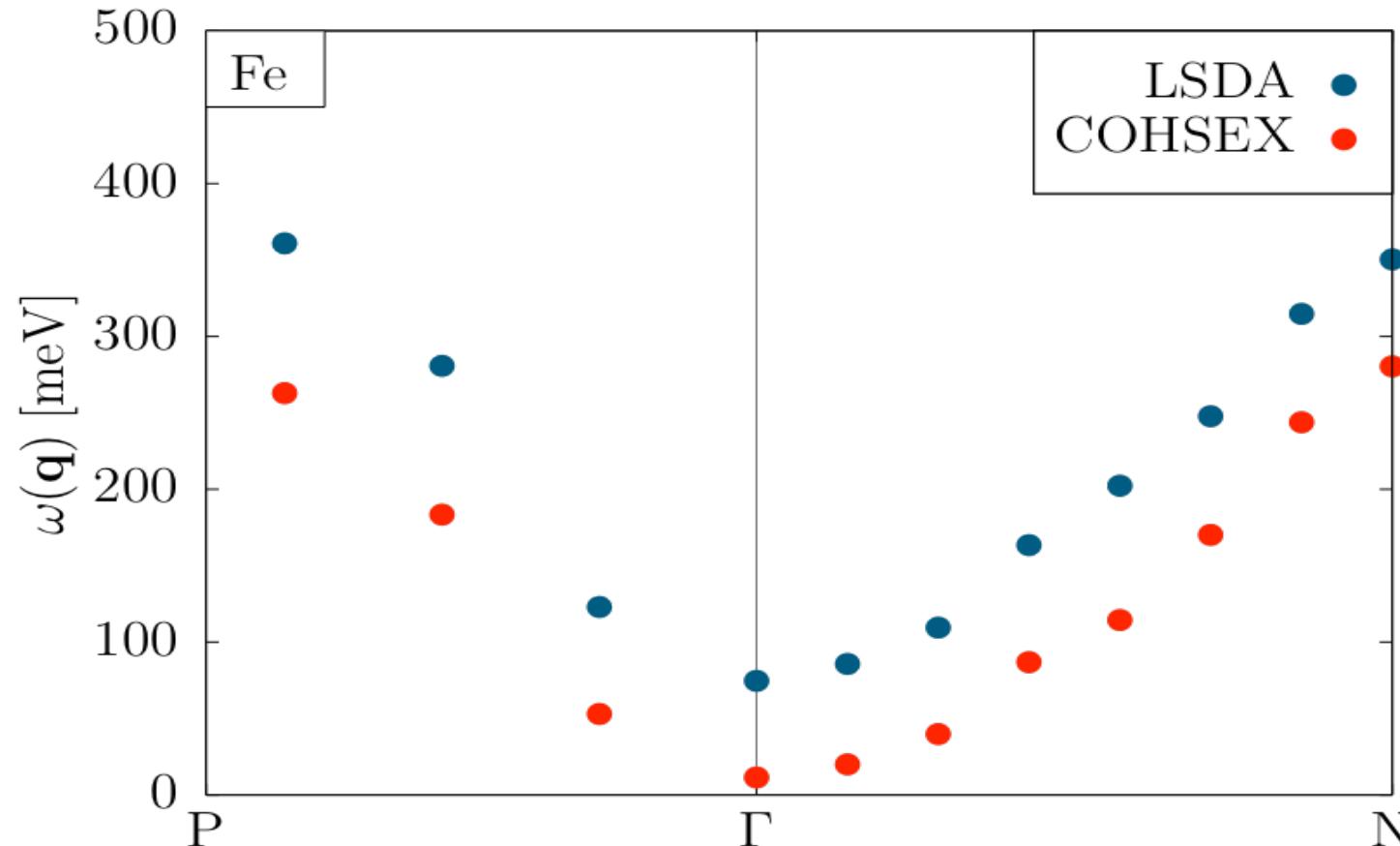
C. Friedrich, M.C.T.D. Müller, S. Blügel,
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VIOLATION OF GOLDSSTONE CONDITION



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

VIOLATION OF GOLDSTONE CONDITION



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

C. Friedrich, M.C.T.D. Müller, S. Blügel,
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In the absence of spin-orbit coupling, the magnetization of a ferromagnet can be rotated without a cost of energy. → $\lim_{q \rightarrow 0} \omega(\mathbf{q}) = 0$

INSIGHT FROM HUBBARD MODEL

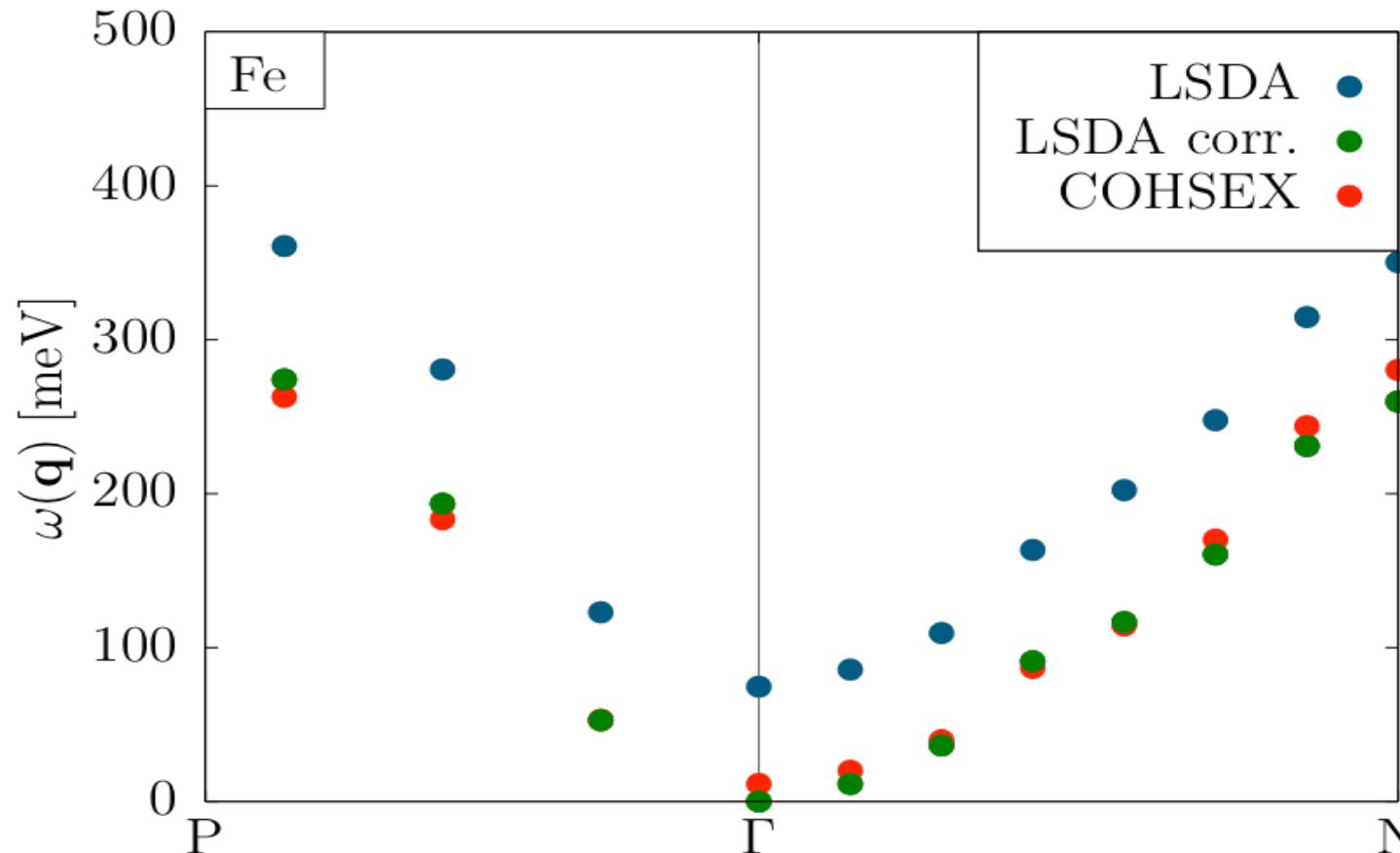
$$H = E_0 \sum_{i\sigma} a_{i\sigma}^\dagger a_{i\sigma} + \sum_{ij,\sigma} t_{ij} a_{i\sigma}^\dagger a_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

Goldstone mode (limit $\mathbf{q} \rightarrow 0, \omega \rightarrow 0$)

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

Parameter for correcting G_{LSDA}

VIOLATION OF GOLDSTONE CONDITION



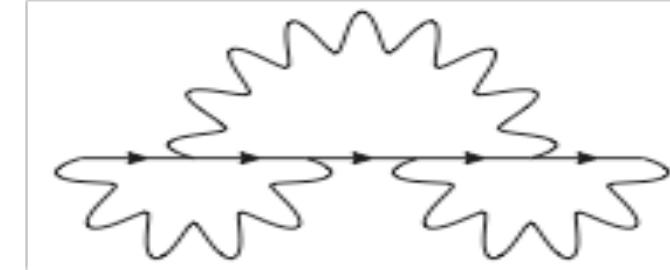
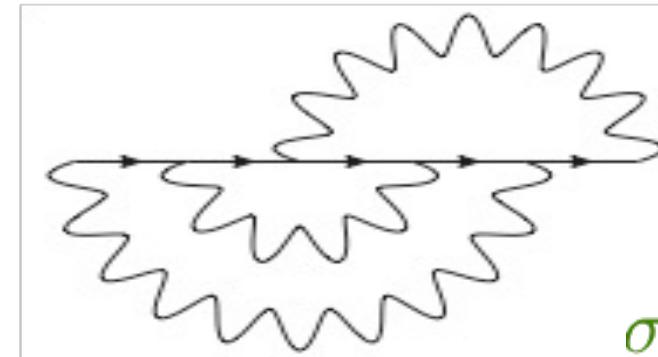
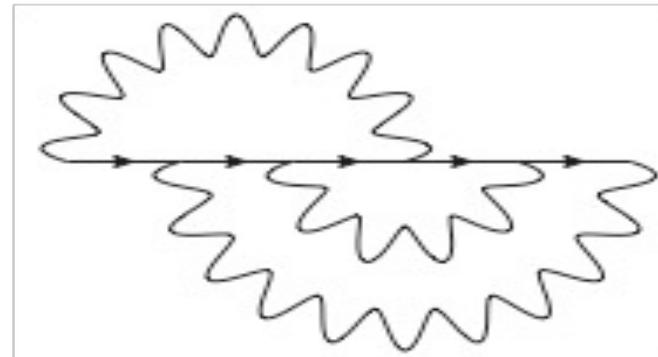
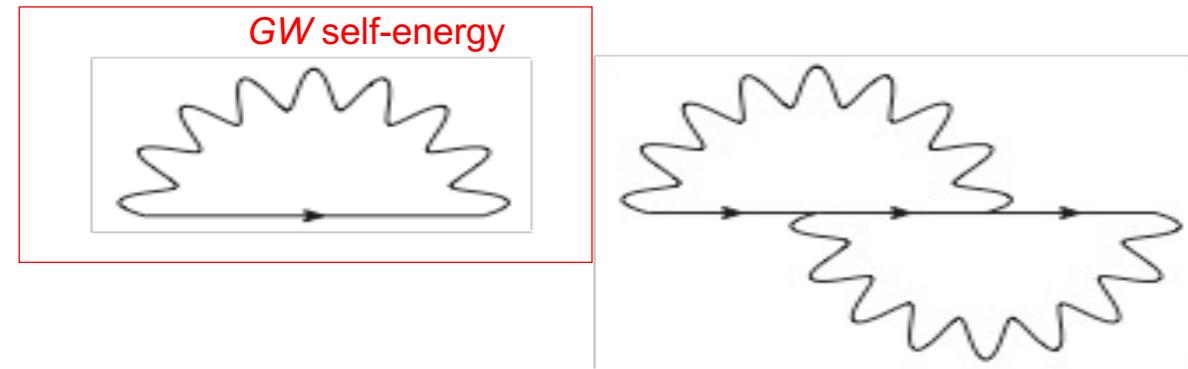
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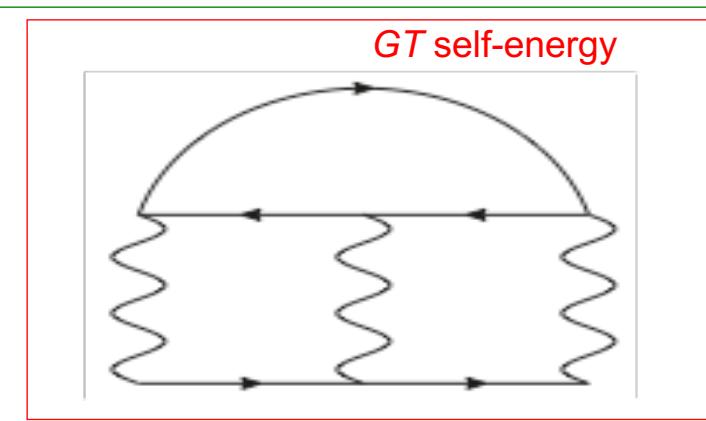
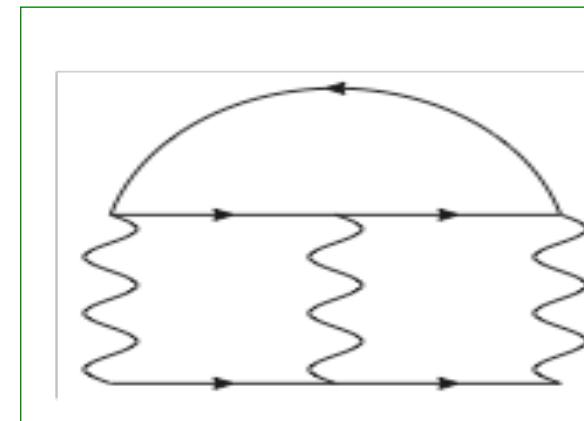
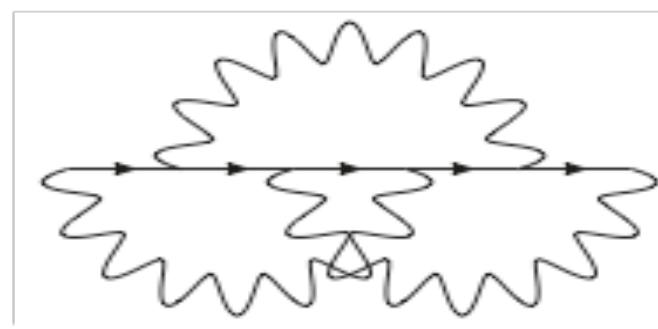
In the absence of spin-orbit coupling, the magnetization of a ferromagnet can be rotated without a cost of energy. → $\lim_{\mathbf{q} \rightarrow 0} \omega(\mathbf{q}) = 0$

SELF-ENERGY (HEDIN EQUATIONS)

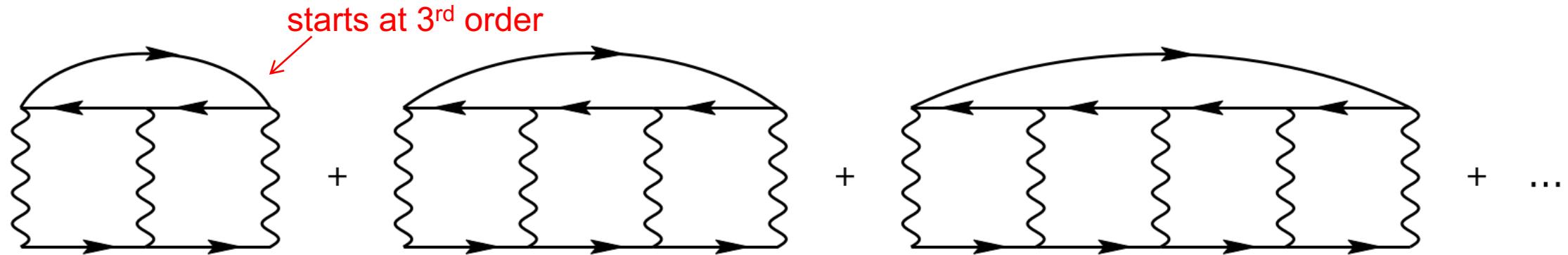
Diagrams up to third order in W :



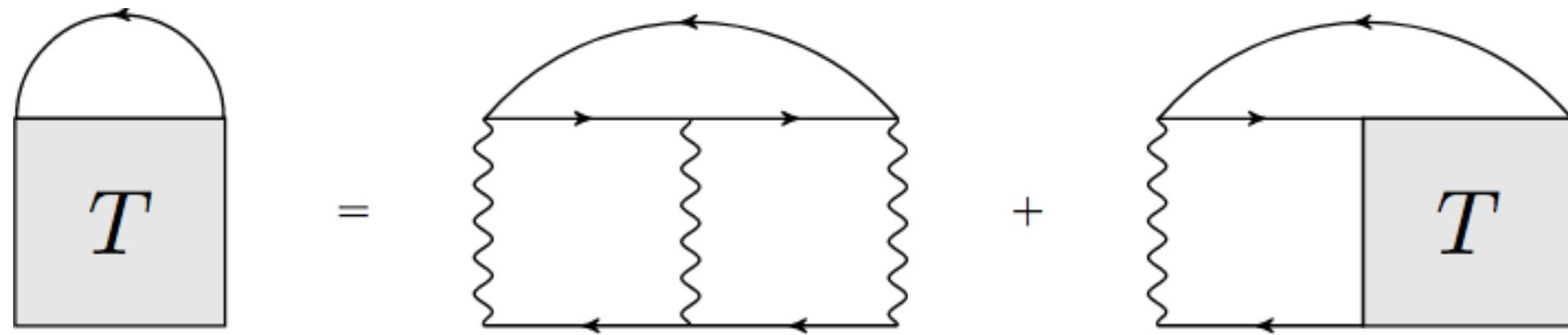
$\sigma \neq \sigma'$



GT SELF-ENERGY



Bethe-Salpeter equation:



Springer, Aryasetiawan, Karlsson, Phys. Rev. Lett. **80**, 2389–2392 (1998):

Zhukov, Chulkov, Echenique, Phys. Rev. Lett. **93**, 096401 (2004):

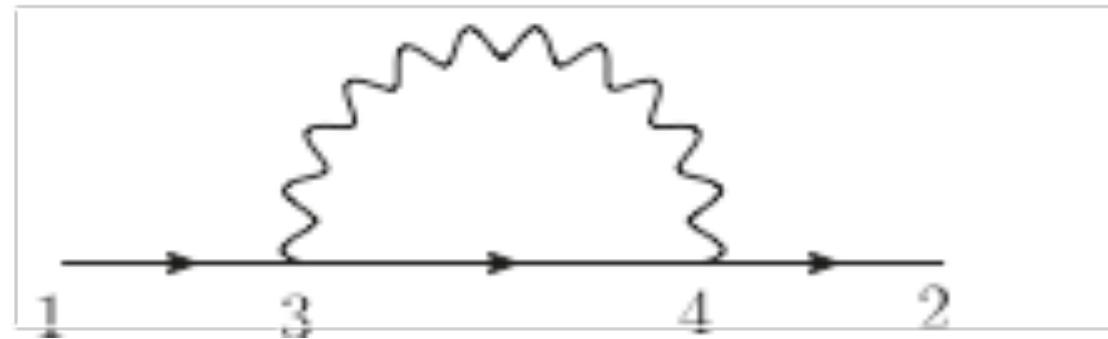
Romaniello, Bechstedt, Reining, Phys. Rev. B **85**, 155131 (2012):

Loos, Romaniello, J. Chem. Phys. **156**, 164101 (2022):

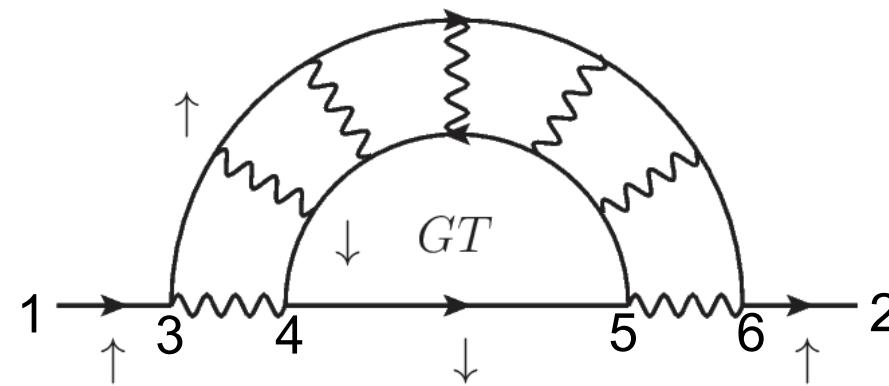
6 eV satellite in Ni
Lifetime effects in Ni and Fe
Diagrammatic expansion
pp channel (molecules)

INTERPRETATION

GW self-energy:

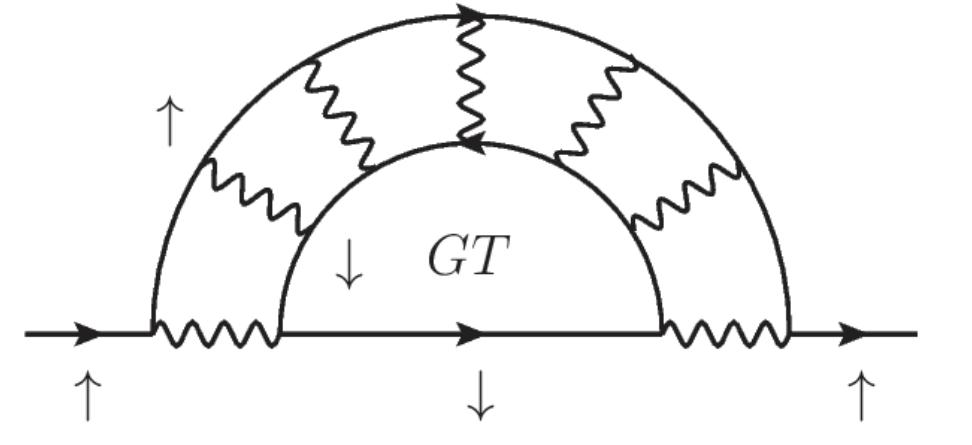


GT self-energy:



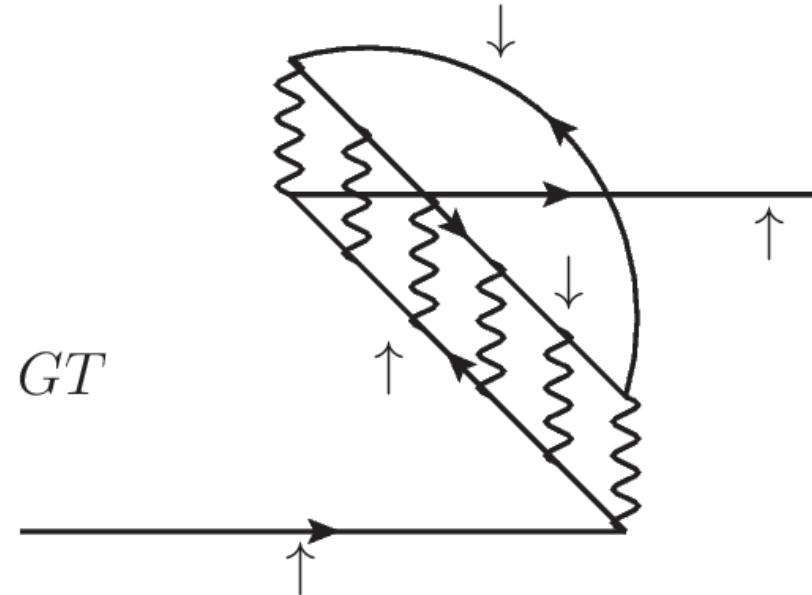
INTERPRETATION

magnon is emitted
before absorbed

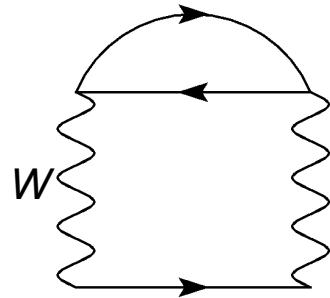
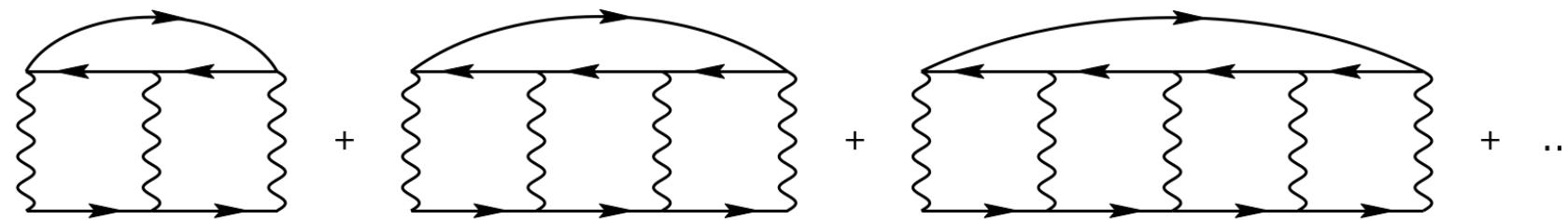


time 

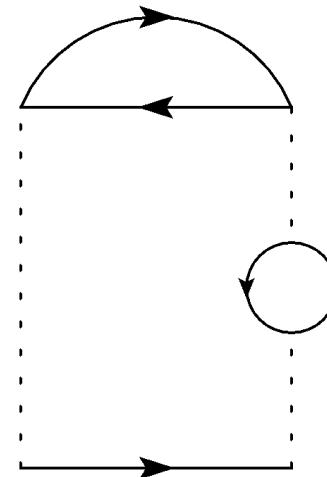
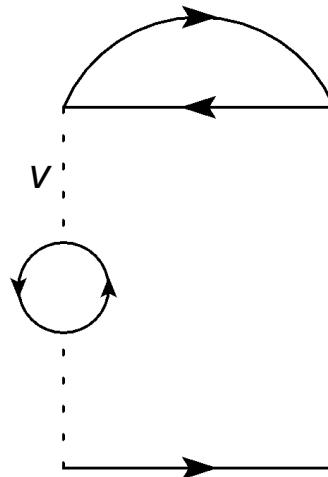
magnon is absorbed
before emitted?
electron travels back in time?



SELF-ENERGY



why not this
diagram?



1. already contained in GW (sort of)
2. unphysical double counting in diagram

IMPLEMENTATION

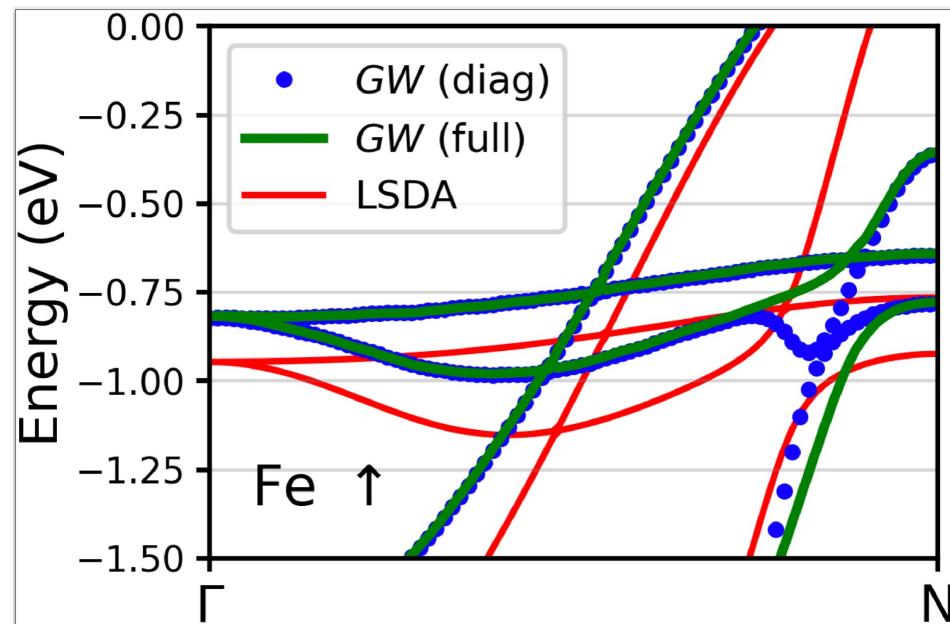
Dyson equation

$$G^\sigma(\mathbf{k}, \omega) = G_0^\sigma(\mathbf{k}, \omega) + G_0^\sigma(\mathbf{k}, \omega)[\Sigma^\sigma(\mathbf{k}, \omega) - v_{xc}^\sigma(\mathbf{k})]G^\sigma(\mathbf{k}, \omega)$$

Spectral function

$$A^\sigma(\mathbf{k}, \omega) = \frac{1}{\pi} \text{sgn}(\epsilon_F - \omega) \text{Im} \{ \text{tr}G(\mathbf{k}, \omega) \}$$

$$= \frac{1}{\pi} \text{sgn}(\epsilon_F - \omega) \text{Im} \{ \text{tr}[\omega I - H^\sigma(\mathbf{k}) - \Sigma^\sigma(\mathbf{k}, \omega) + v_{xc}^\sigma(\mathbf{k})]^{-1} \}$$



IMPLEMENTATION

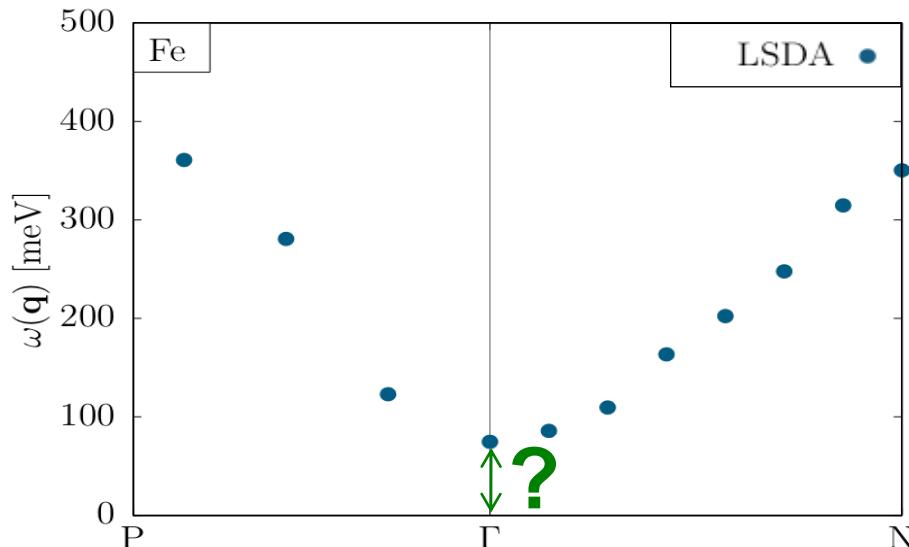
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M. Müller, C. Friedrich, S. Blügel, Phys. Rev. B **94**, 064433 (2016)

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IMPLEMENTATION

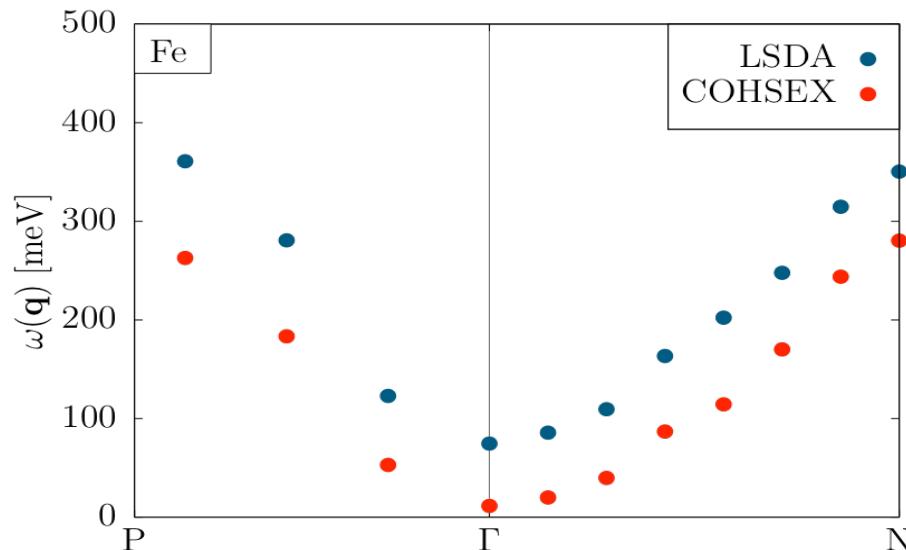
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IMPLEMENTATION

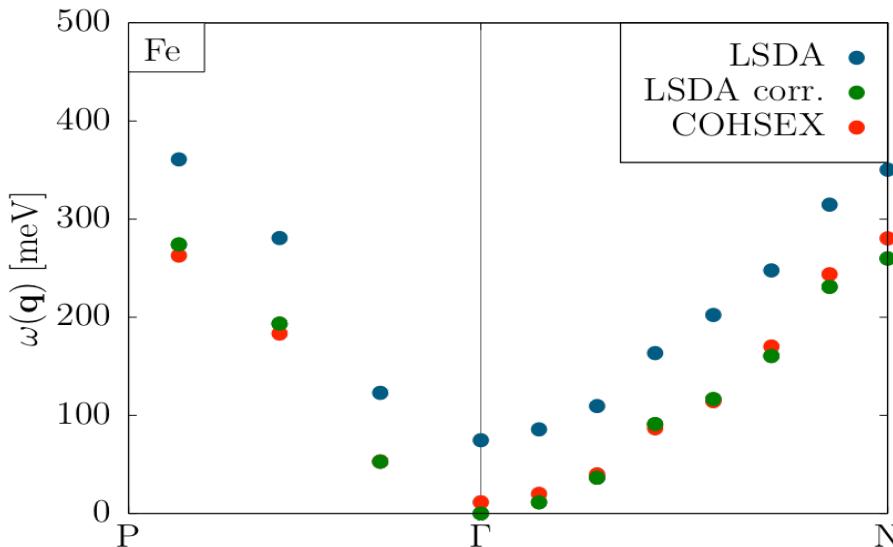
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IMPLEMENTATION

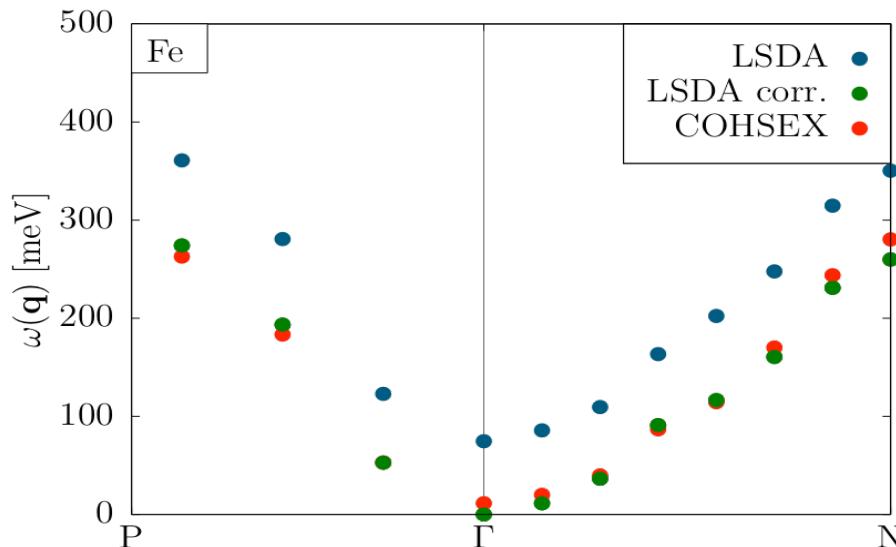
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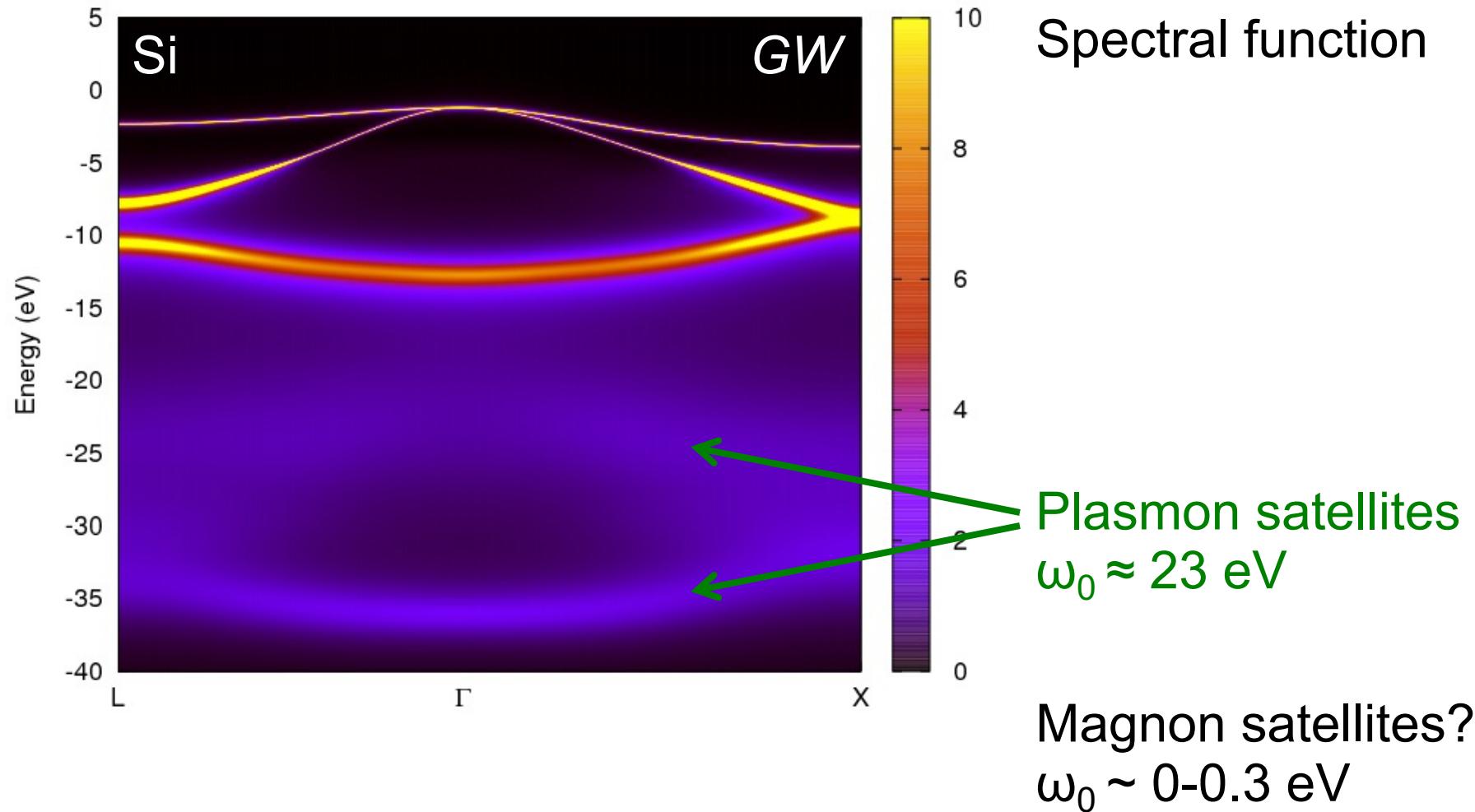


$$\boxed{\omega \pm \frac{\Delta_x}{2}}$$

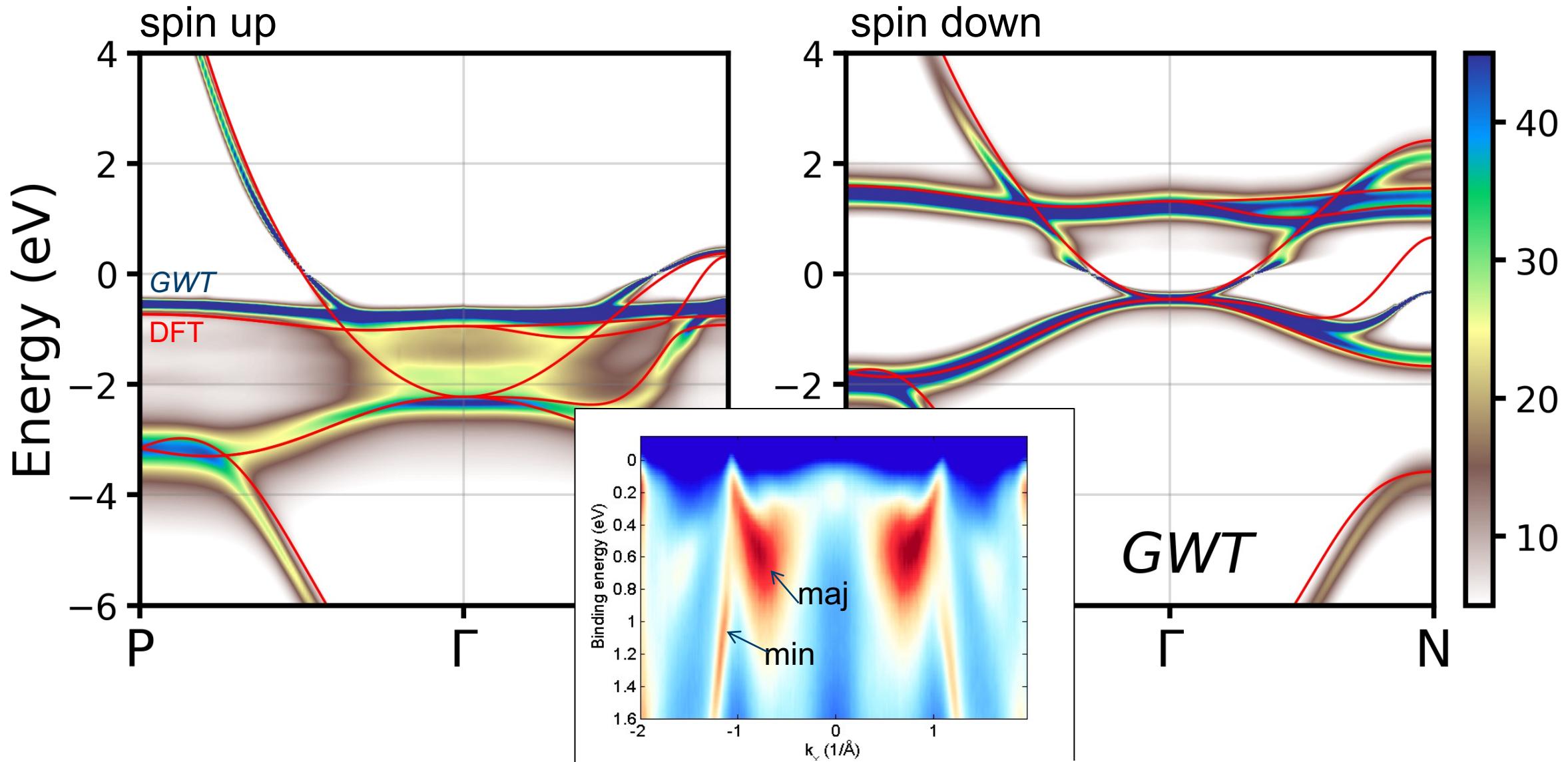
- to enforce Goldstone condition
- self-consistency condition
- $\Delta_x = 0.23 \text{ eV}$

$$\boxed{\omega - \Delta_v}$$

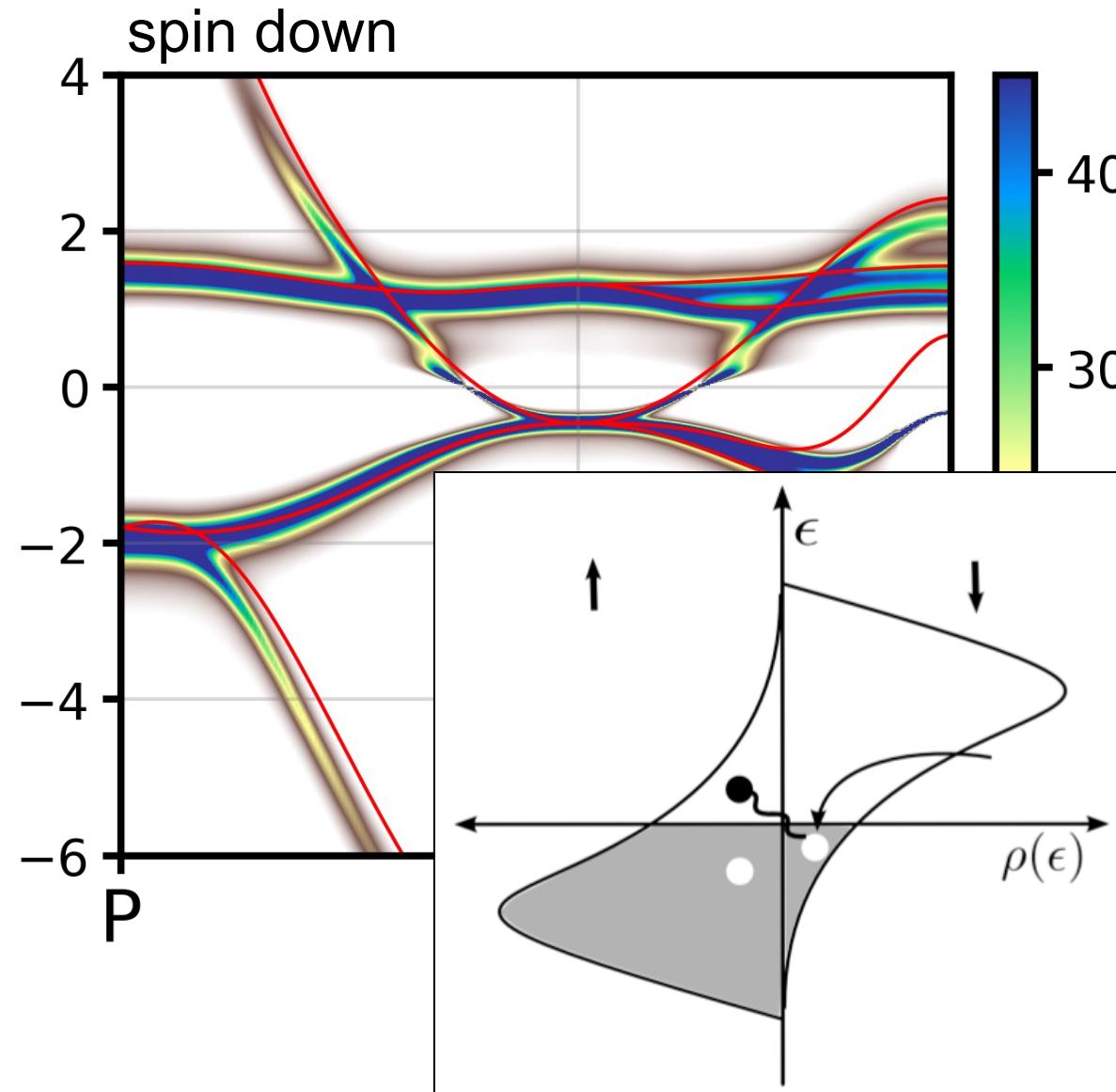
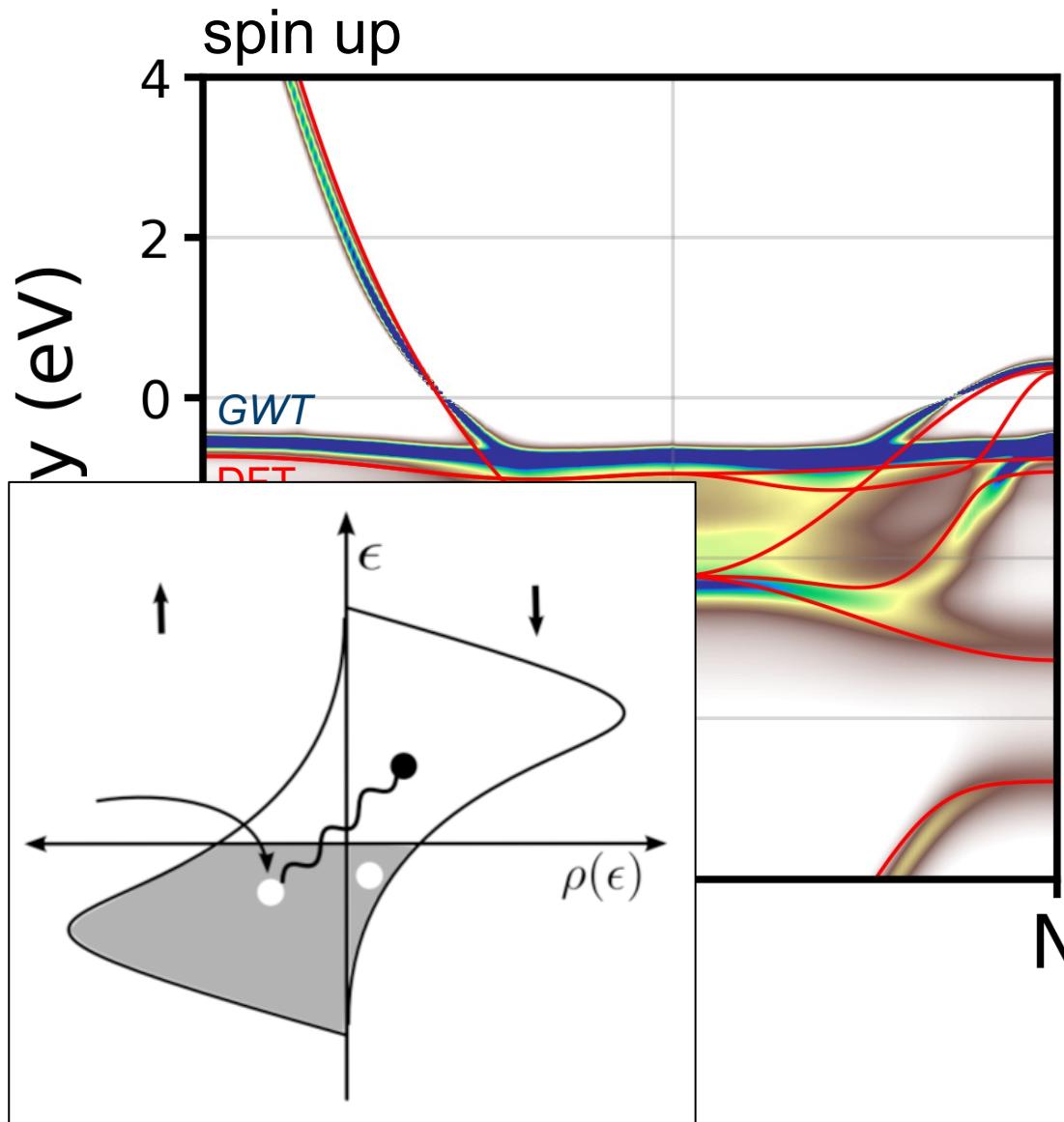
- self-consistency condition
- $\Delta_v = 0.96 \text{ eV}$



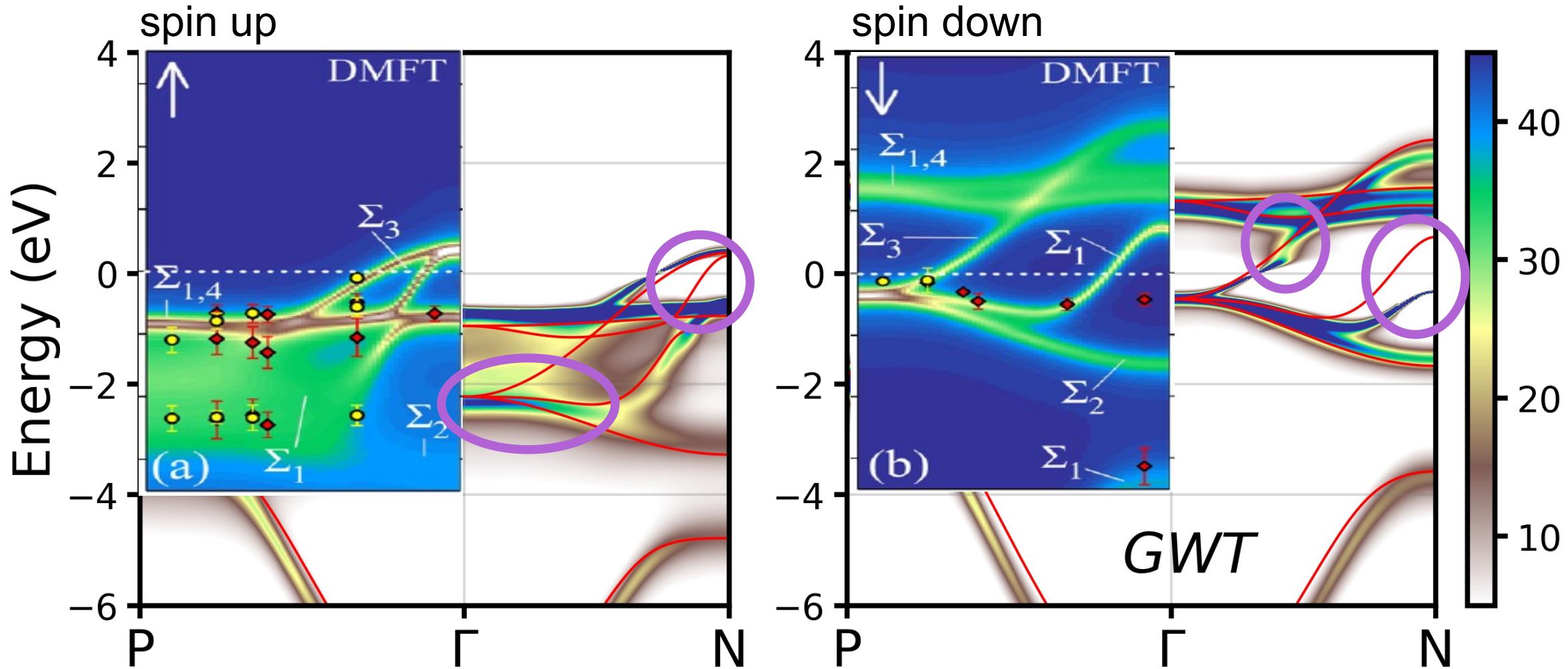
IRON BAND STRUCTURE (GWT)



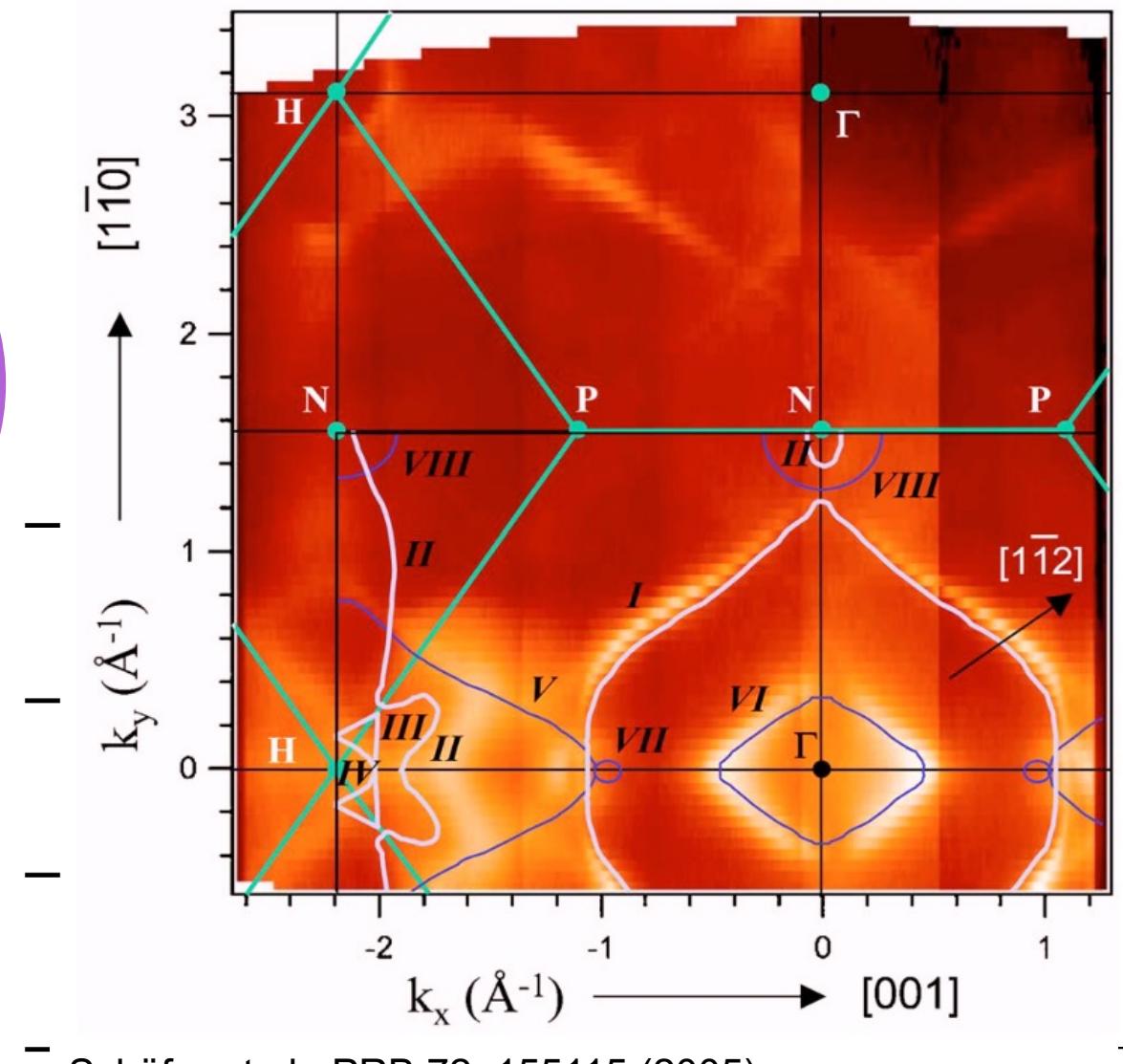
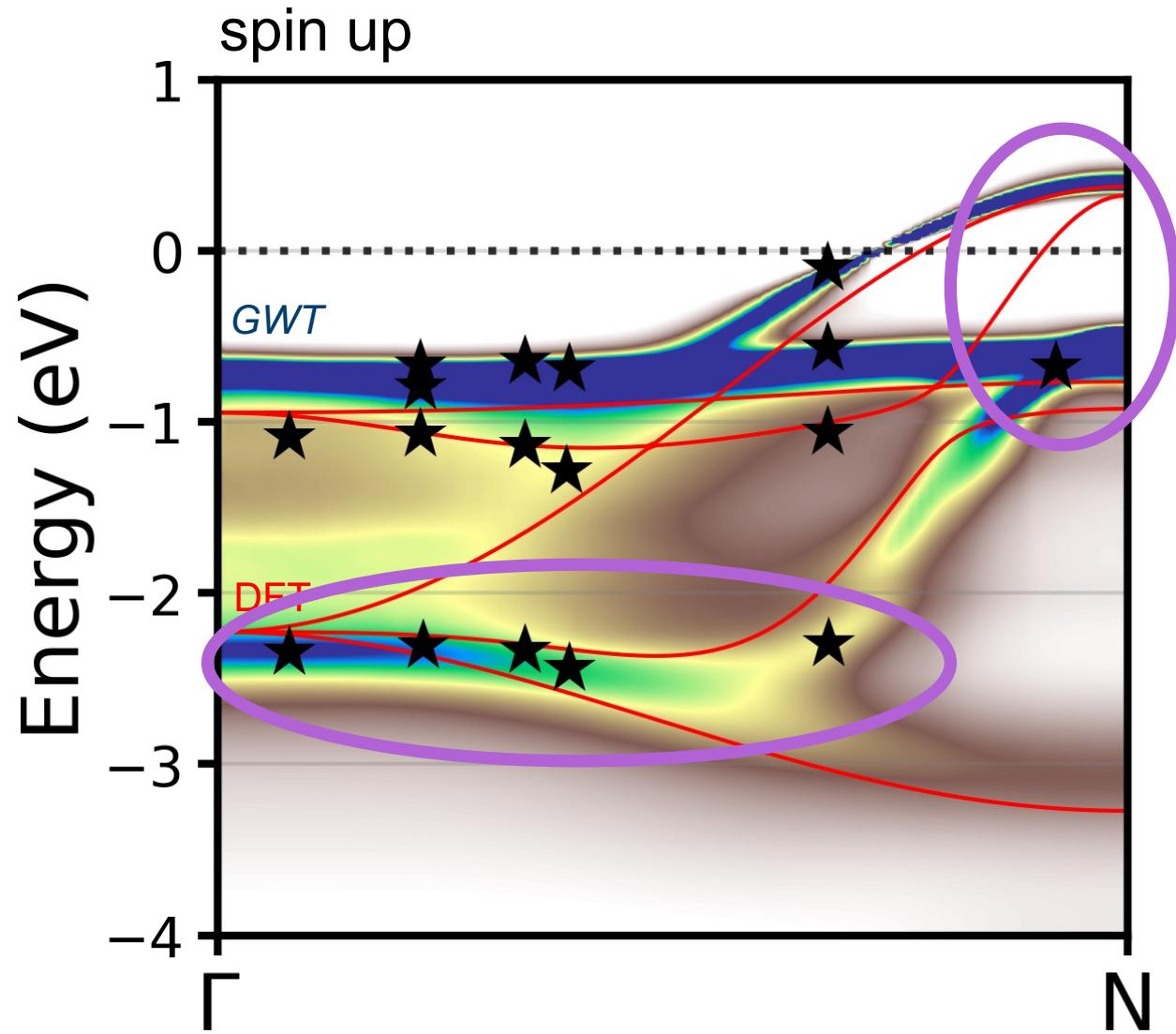
IRON BAND STRUCTURE (GWT)



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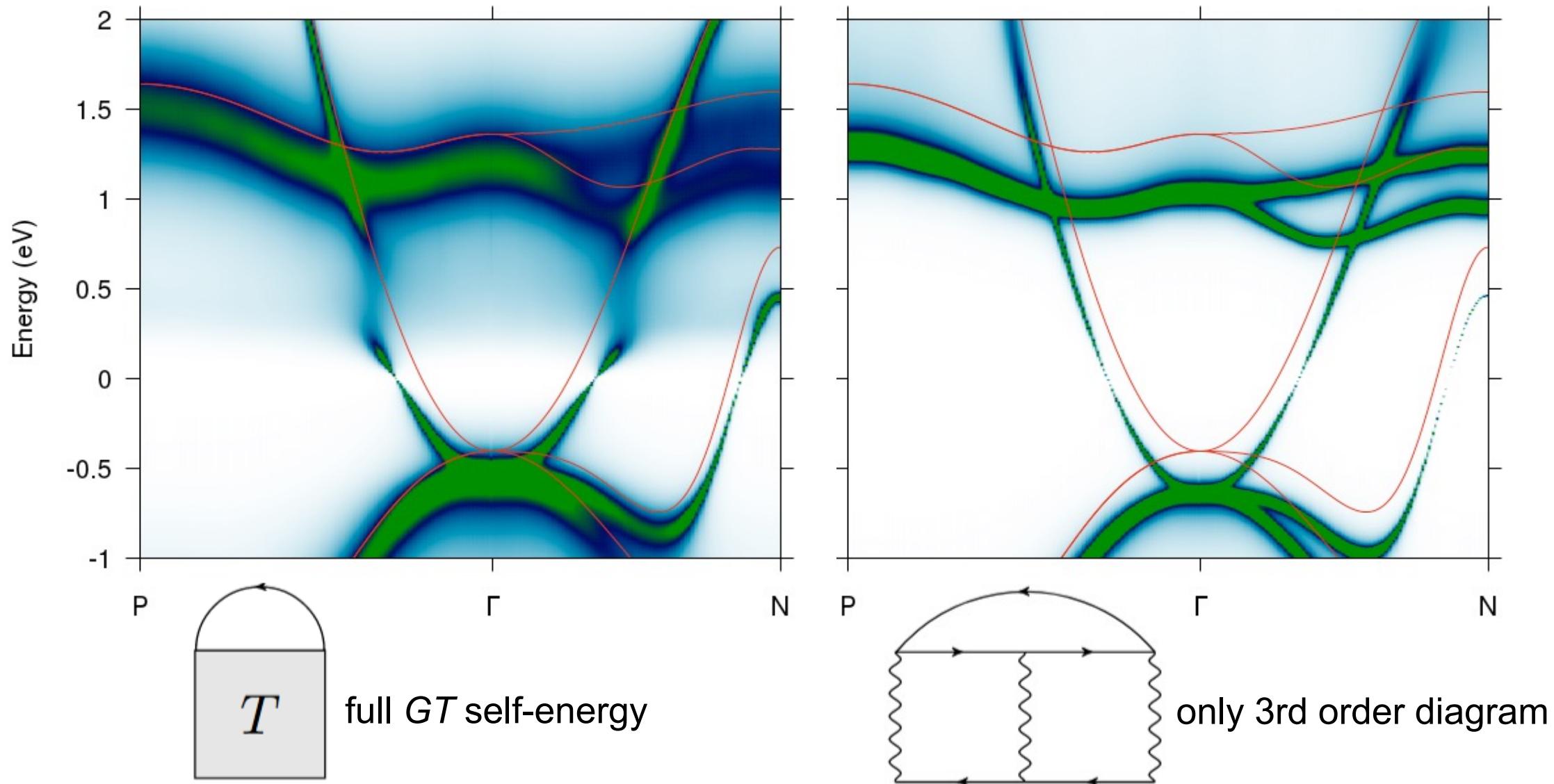
IRON BAND STRUCTURE (GWT)



Schäfer et al., PRB 72, 155115 (2005)
Mlynczak et al., PRB 105, 115135 (2022)

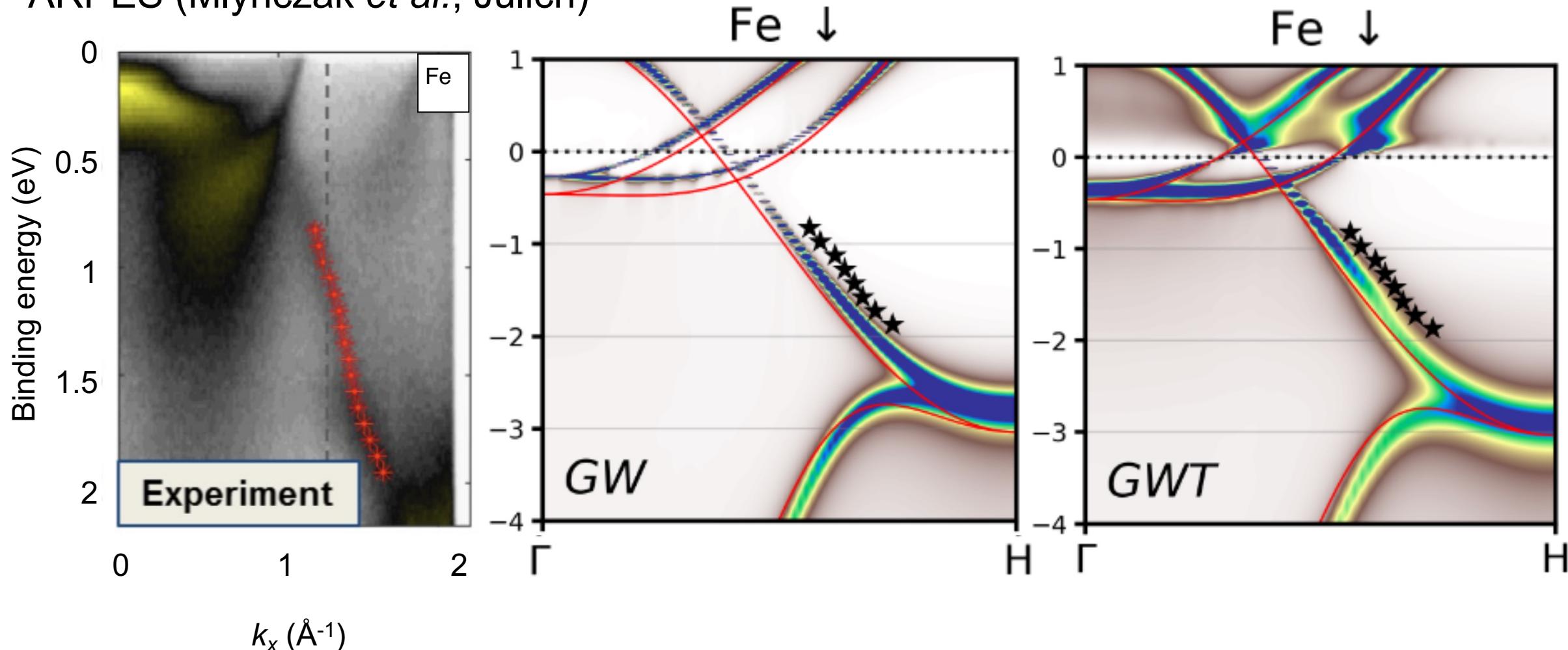
exp: J. Sánchez-Barriga et al., PRL 103, 267203 (2009)

ANOMALY SPIN DOWN (IRON)



HIGH-ENERGY BAND ANOMALY (IRON)

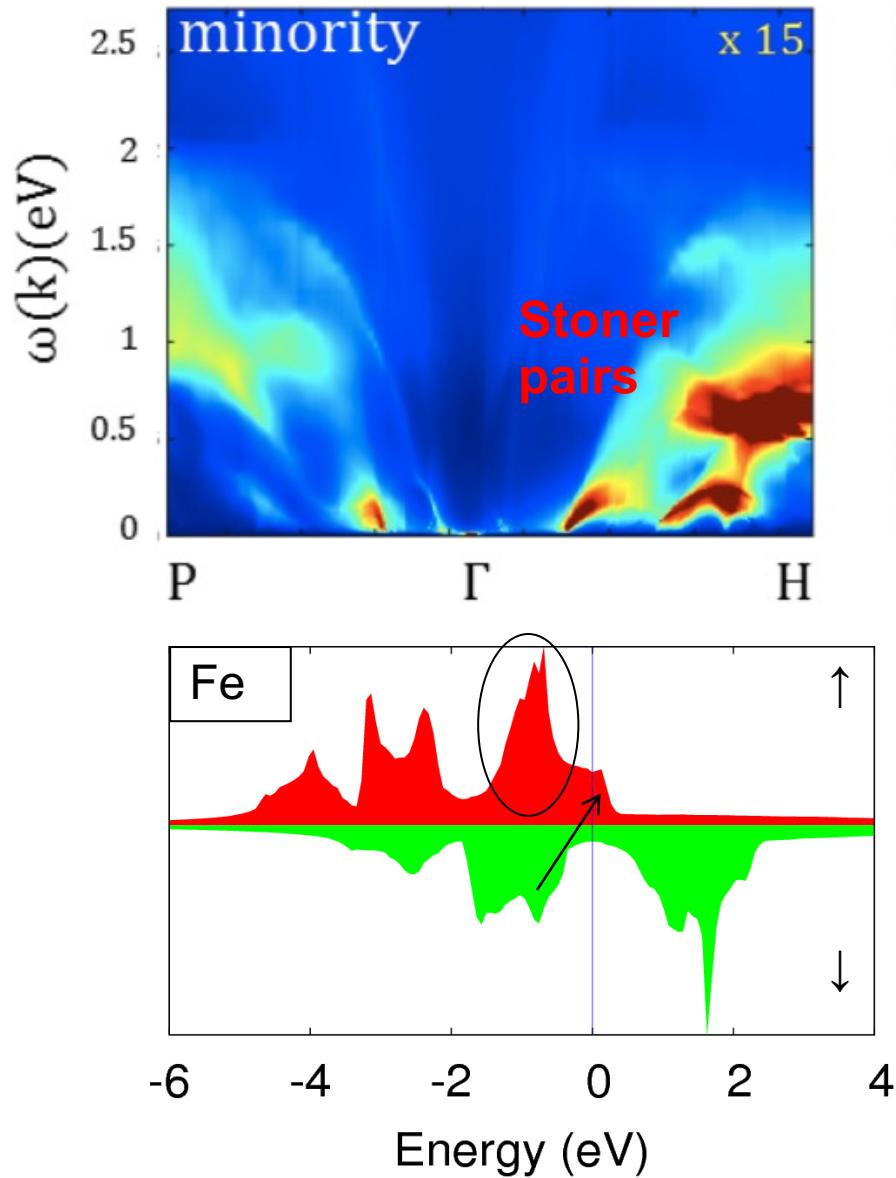
ARPES (Mlynczak *et al.*, Jülich)



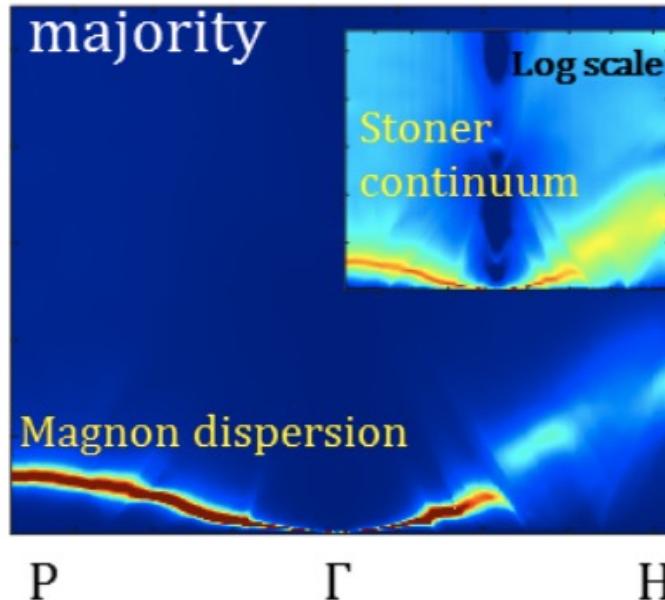
exp: Mlynczak *et al.*, Nature Comm. **10**, 1 (2019)

theo: Nabok *et al.*, npj Comput. Mater. **7**, 178 (2021)

HIGH-ENERGY BAND ANOMALY (IRON)



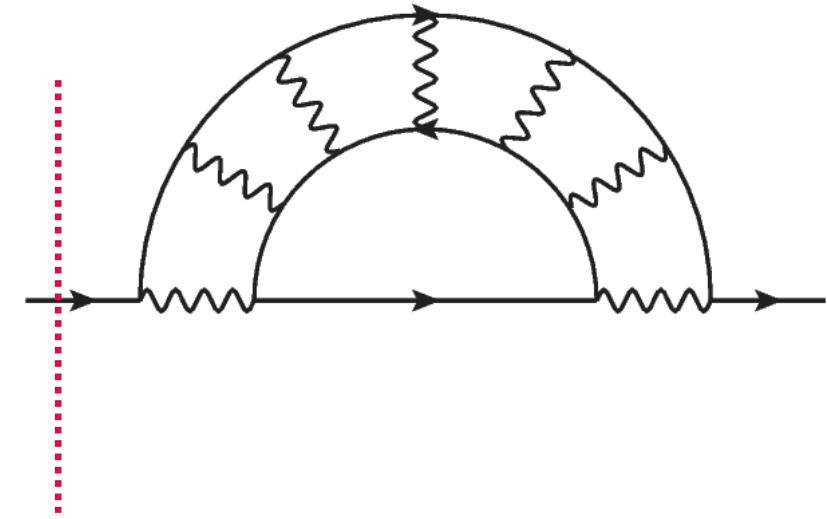
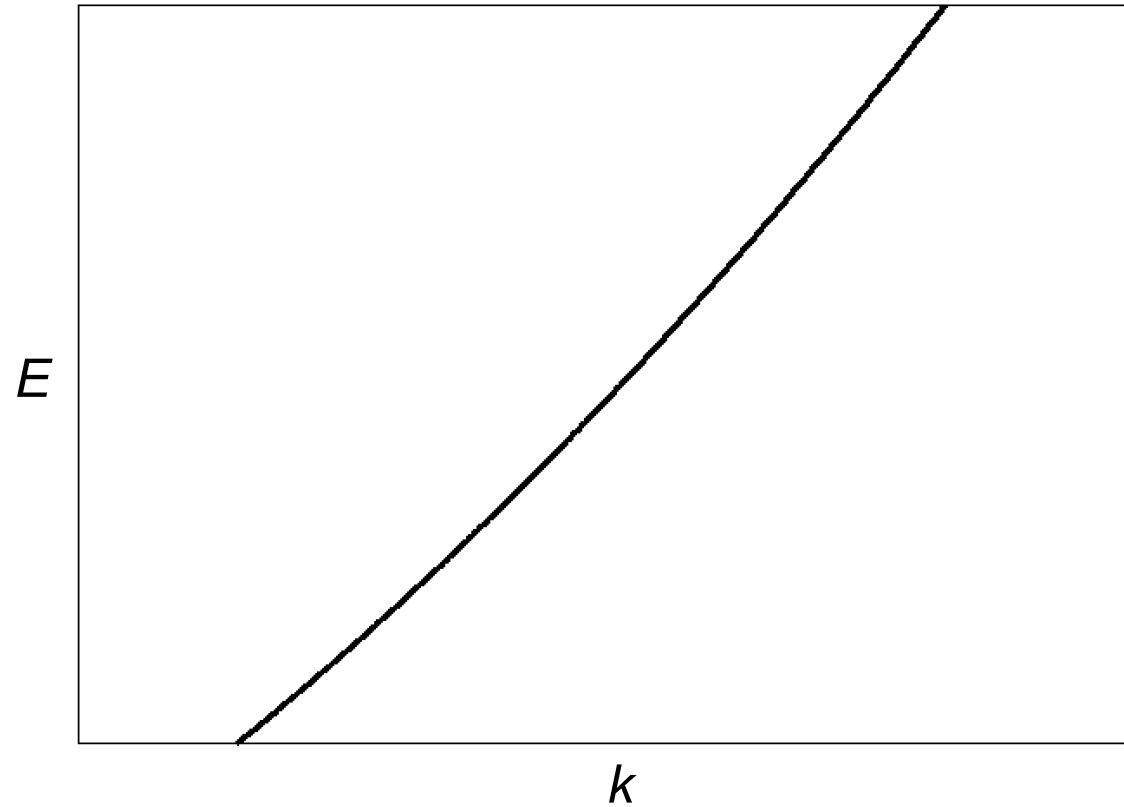
Mlynczak et al., Nature Communications 10, 505 (2019)



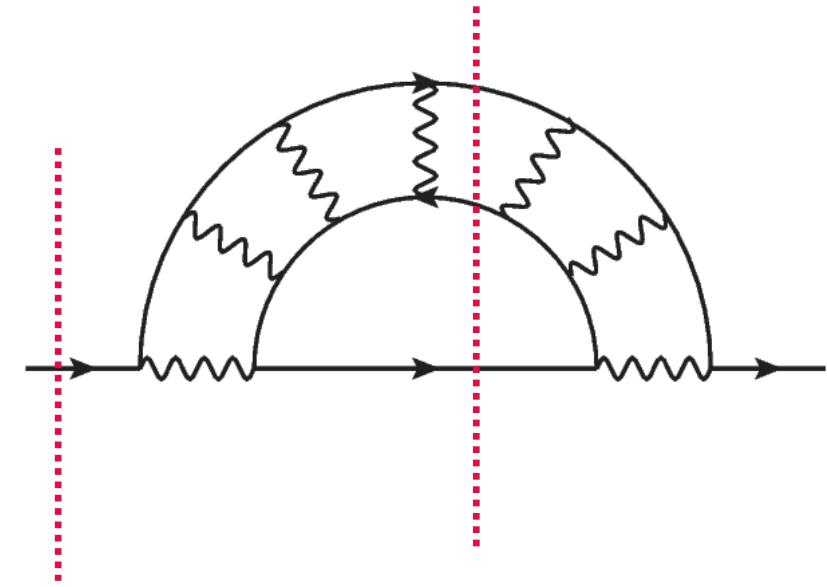
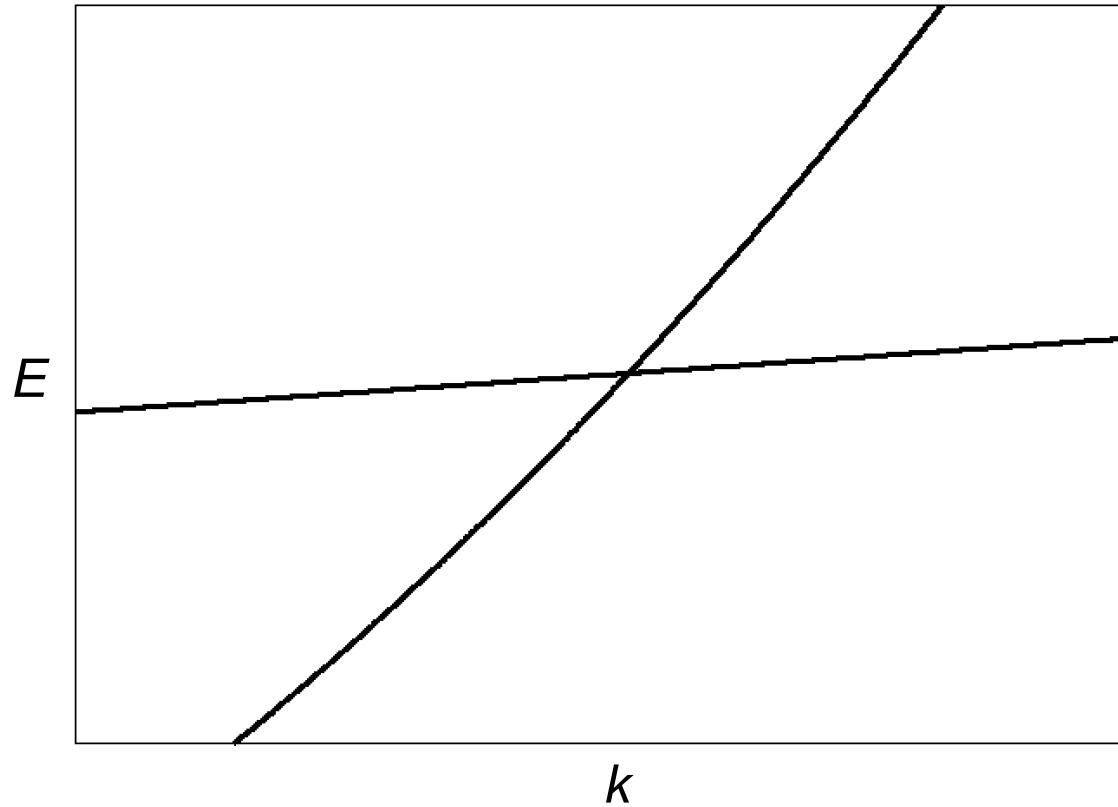
P Γ H

$$\begin{aligned}\Sigma_{GT} &= \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{1}{\omega - \omega' - \epsilon - i\eta} \frac{1}{\omega' - \epsilon_M - i\eta} d\omega' \\ &= \frac{i}{\omega - (\epsilon + \epsilon_M) - i\eta} \\ &\approx 0.8 \text{ eV} \quad \approx 0.7 \text{ eV} \quad \approx 1.5 \text{ eV}\end{aligned}$$

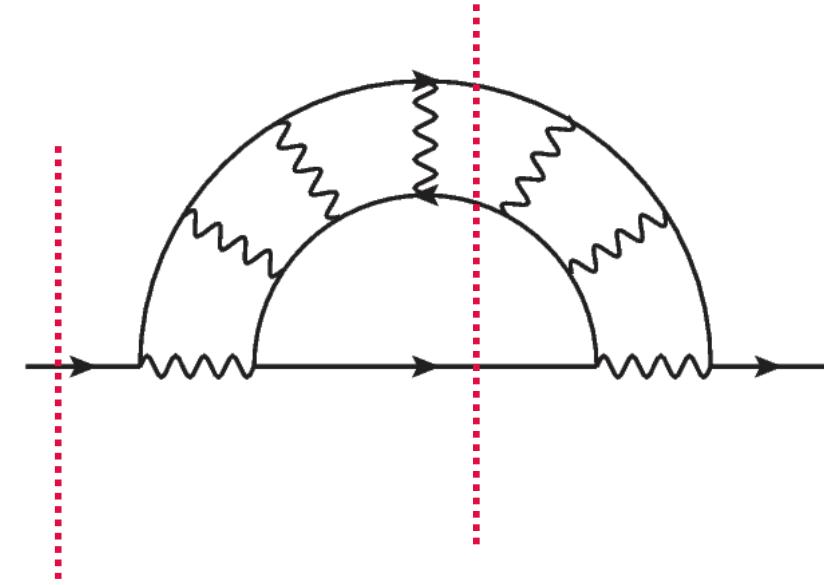
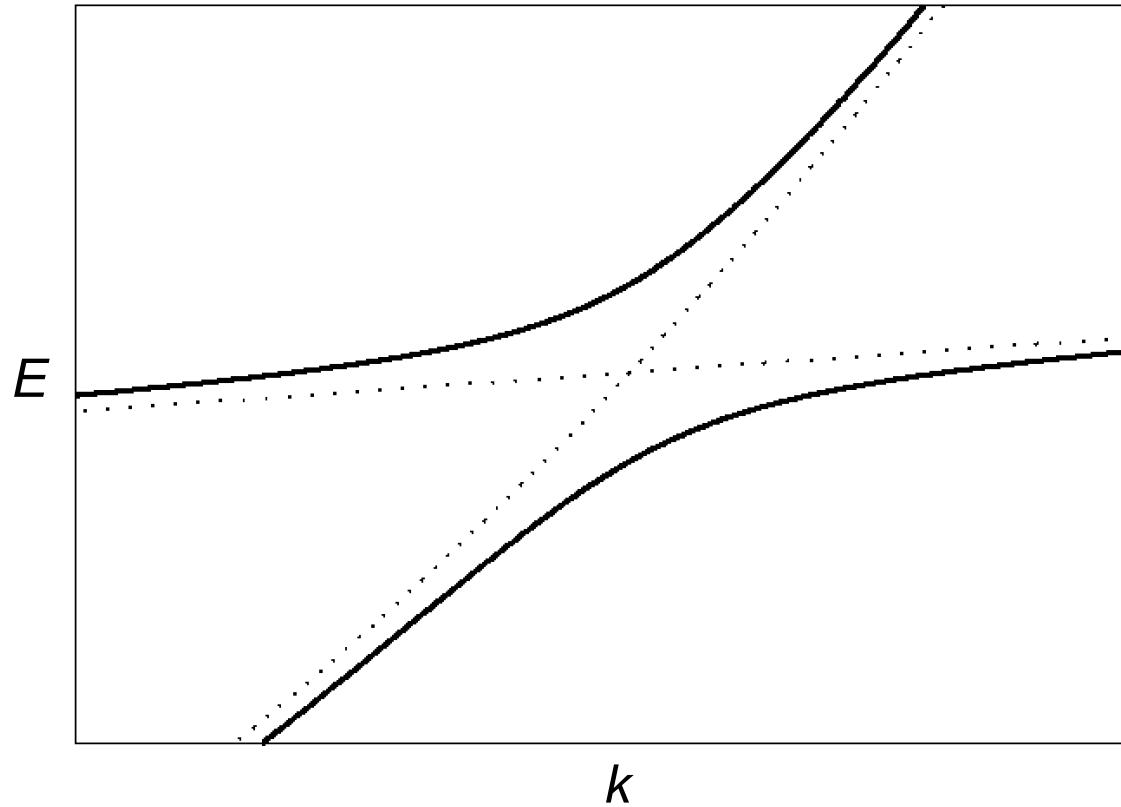
BAND ANOMALY (WATERFALL)



BAND ANOMALY (WATERFALL)

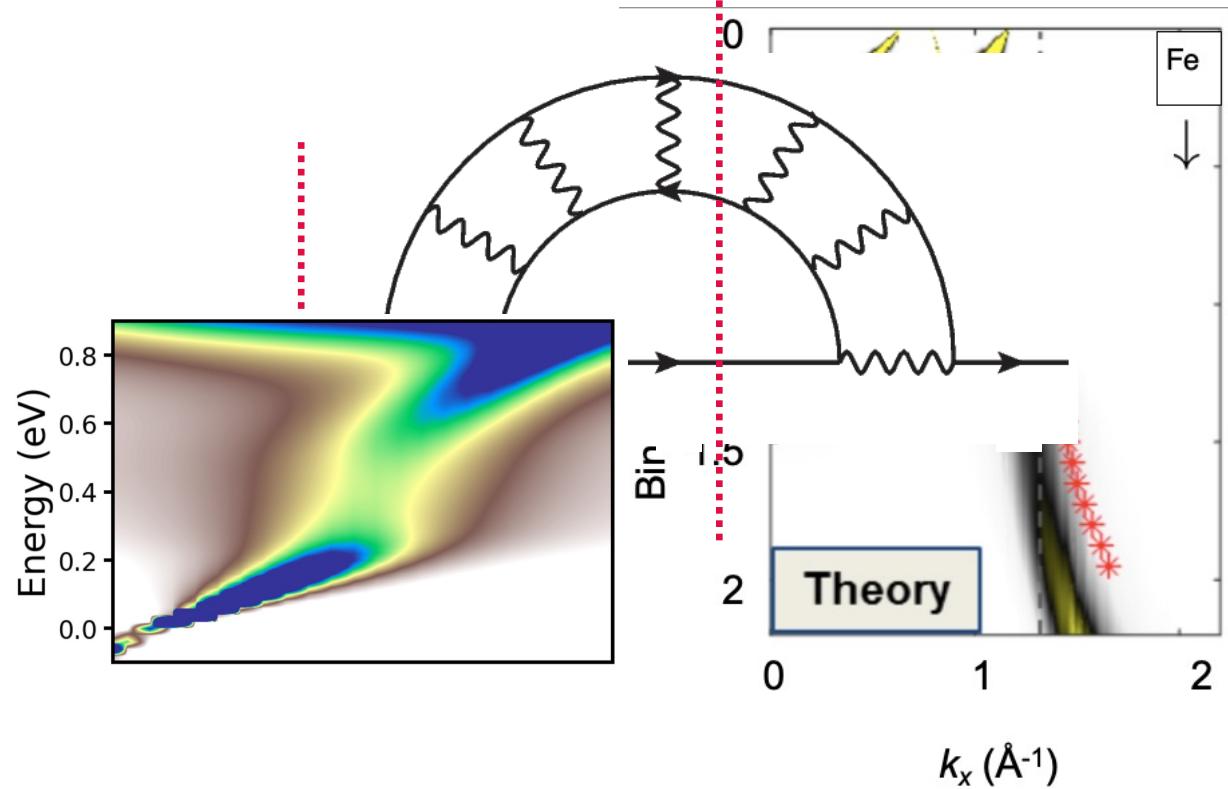
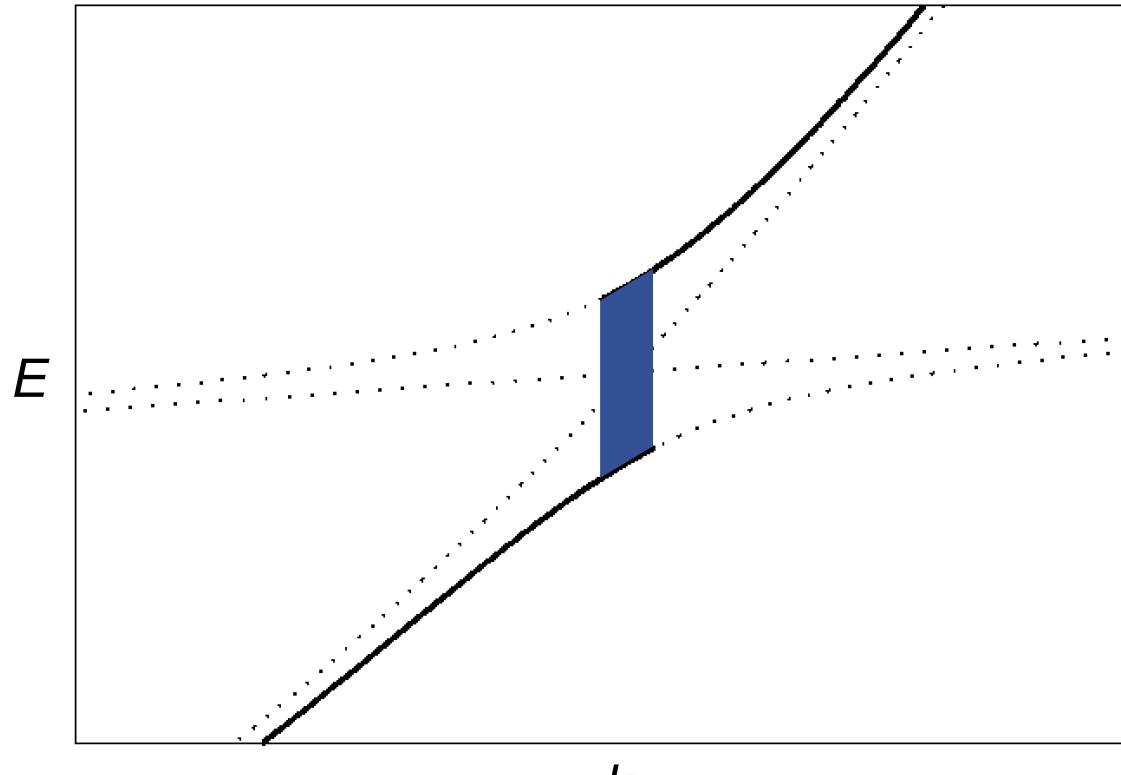


BAND ANOMALY (WATERFALL)



$$G(\mathbf{r}, \mathbf{r}'; \omega) = \sum_n \frac{\psi_n^{N-1}(\mathbf{r}) \psi_n^{N-1 *}(\mathbf{r}')}{\omega - \epsilon_n^{N-1} - i\eta} + ... N + 1 ...$$

BAND ANOMALY (WATERFALL)



$$G(\mathbf{r}, \mathbf{r}'; \omega) = \sum_n \frac{\psi_n^{N-1}(\mathbf{r}) \psi_n^{N-1 *}(\mathbf{r}')}{\omega - \epsilon_n^{N-1} - i\eta} + \dots N+1\dots$$

TABLES (IRON)

Table 1. Electron spin magnetic moments (in μ_B) of Fe, Co, and Ni computed with different techniques.

	LSDA	GW	GT	GWT	exp ¹
Fe	2.15	2.12 (2.17)	2.00	2.09	2.08
Co	1.58	1.57 (1.65)	1.28	1.54	1.52
Ni	0.58	0.59 (0.65)	0.49	0.59	0.52

GW values in brackets are calculated without the Δ_x correction (see text).

¹Table 12 of ref. ⁵².

Table 2. d bandwidths (in eV) for bcc Fe, fcc Co, and fcc Ni obtained from LSDA, GW, GT, and GWT and compared with experimental data.

W_d	LSDA	GW	GT	GWT	exp
Fe $N_{1\uparrow}$	4.74	4.29 (4.30)	5.10	4.67	4.50 ± 0.23^1
Fe $N_{1\downarrow}$	3.54	3.46 (3.35)	3.96	3.57	3.60 ± 0.20^1
Co $\langle L_1 \rangle$	4.64	4.18 (4.33)	4.67	4.40	3.8 ± 0.5^2 $> 4.0^3$
Ni $\langle L_1 \rangle$	4.58	4.12 (4.18)	4.56	4.12	3.9 ± 0.2^4

The bandwidths are estimated, as in the experiment, based on the specified states. The GW values in brackets are computed without the Δ_x correction (see text).

¹Ref. ⁵³.

²Ref. ³⁷.

³Ref. ³⁶.

⁴Ref. ¹⁶ (based on experimental data from ref. ²).

Table 3. The exchange splitting ΔE_x at special \mathbf{k} points computed with different techniques and compared with available experimental values.

ΔE_x	LSDA	GW	GT	GWT	exp
Fe					
Γ_{25}	1.77	1.41 (1.55)	2.09	1.90	2.08 ± 0.10^1
H_{12}	1.56	1.06 (1.19)	1.64	1.51	1.30 ± 0.30^1
P_4	1.36	0.96 (1.08)	1.50	1.19	1.35 ± 0.10^1
N_2	1.60	1.19 (1.32)	1.78	1.49	1.60 ± 0.15^1
Co					
Γ_{25}	1.44	0.98 (1.24)	1.39	1.18	1.20 ± 0.30^2
Γ_{12}	1.66	1.20 (1.48)	1.49	1.04	0.85 ± 0.20^2
L_3	1.48	1.00 (1.26)	1.33	1.18	1.15 ± 0.40^2
Ni					
L_3	0.56	0.37 (0.51)	0.51	0.37	0.31 ± 0.03^3
X_2	0.52	0.30 (0.45)	0.44	0.31	$\sim 0.2^4$

GW values in brackets are calculated without the Δ_x correction in the reference system (see text).

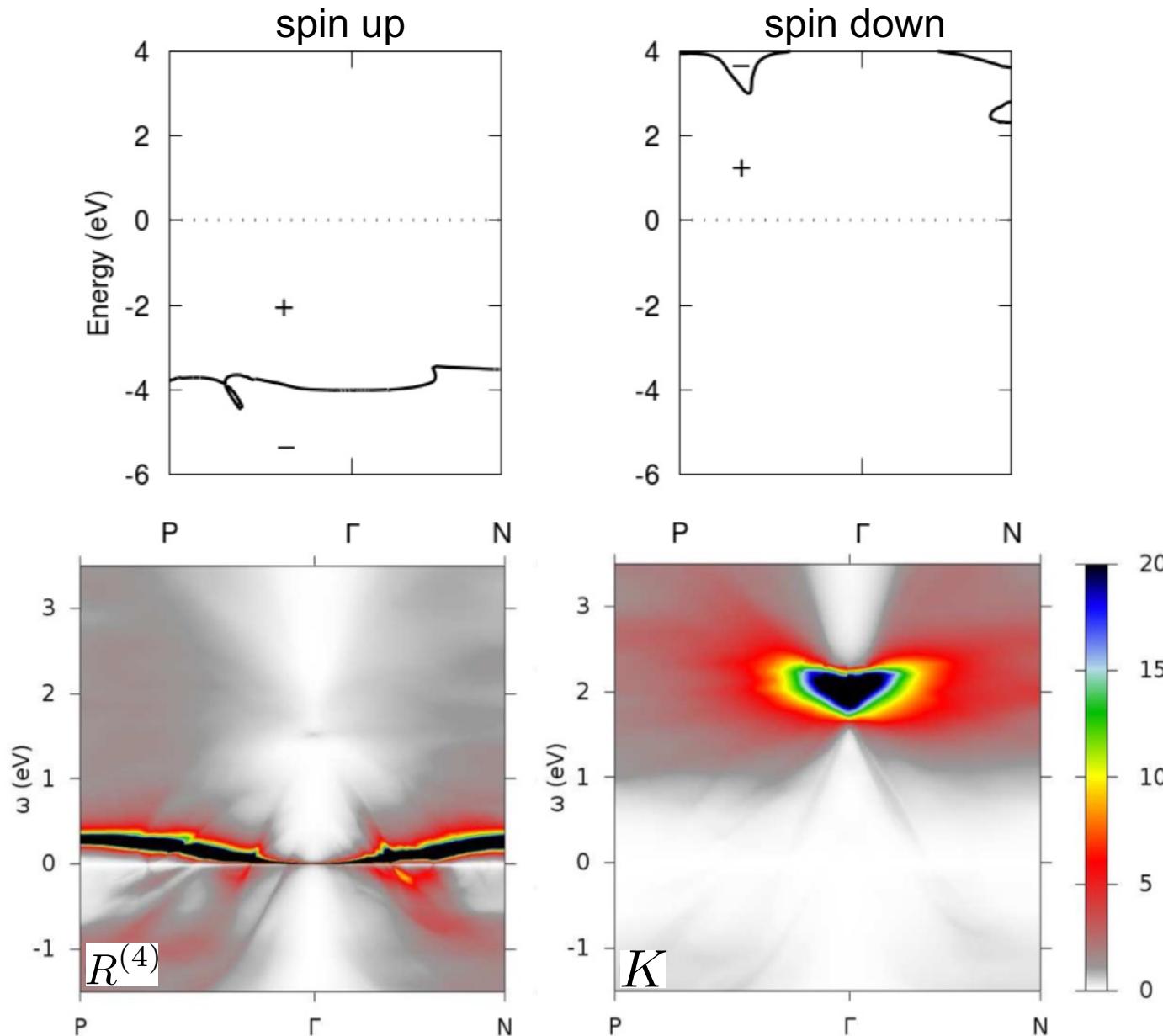
¹Ref. ⁵³.

²Ref. ³⁷.

³Ref. ⁵⁴.

⁴Ref. ⁵⁵.

VIOLATION OF CAUSALITY (GT)



There are regions where the GT spectral function turns **negative!**

Can be traced back to the imag. part of the **T -matrix** having negative sign.

$$\begin{aligned} T &= T_{\geq 2} - T_2 \\ &= W[R^{(4)} - K]W \end{aligned}$$

→ difference is **negative**

GW restores correct sign.

SUMMARY

- 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 $\mathbf{q} \rightarrow 0$ and $\omega \rightarrow 0$ due to inconsistency of Green functions (LSDA vs. self-consistent); self-consistent **COHSEX** recovers Goldstone mode.
- Electron-magnon scattering described by **GT self-energy**.
- Double-counting-free combination with GW self-energy gives the **GWT self-energy**.
- **Strong spin asymmetry** of lifetime broadening 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).
- **GWT band structure** of Fe is similar to DMFT spectral function but with improvements in the details:
 - quasiparticle band at 2.34 eV (Γ) and 3.2 eV (P),
 - 1.5 eV band anomaly,
 - no false prediction of hole pocket at N

ACKNOWLEDGMENTS



Dmitrii Nabok



Mathias C. T. D.
Müller



Stefan Blügel

Experiments:



Ewa Mlynczak



Lukas Plucinski