**Fe paper structure**

# Introduction

Accurate *ab-initio* theory of magnetism of complex highly correlated electron systems plays critical role in description and prediction of properties of modern functional materials. Iron is a classic example of strongly correlated electron system, which magnetic susceptibility exhibits properties described both by localized and itinerant electronic states1,2. As such it becomes archetypal system2 for experimental and theoretical investigations, providing benchmark material, which magnetic properties should be thoroughly understood by any realistic theory and reproduced by any modelling code.

Magnetic properties of iron have been widely investigated experimentally using triple axis spectrometers and early versions of inelastic direct spectrometers3,4,5,6 in later 80th -90th. These experiments delivered results in low energy spin waves excitations range, so the whole picture of magnetic excitations in iron remains incomplete. First quantitative theoretical description of magnetic susceptibility of iron was initially performed using Band model and Random phase approximation (see, e.g7,8,9 and references therein) claiming good correspondence between theory and experiment, though the experiment and theory were mainly covering the low energy spin-waves excitations. Number of later publications have presented first-principles calculations of magnetic susceptibility of iron using various flavours of density functional theory (TDDFT)10,11,12,13,14,15,16,17, many-body perturbation theory18,19,20 or dynamical mean field theory21. All these theories give reasonable correspondence between each other and the experimental results for low energy high wavelength magnetic excitations but suggest different dispersion and structure (stoner excitations, spin-waves excitations spectra, etc.) in high energy range.

In this work we provide comprehensive experimental investigations of spin-wave excitations in iron over whole Brillouin zone. In addition to that, we use two modern *ab initio* TDFT codes15,22 which use substantially different approaches to solution of electronic structures and spin waves dispersion and compare their predictions with the results of the experiment.

# Experimental investigations

# Ab initio calculations

Measurements

* TOF technique
* What we did
* Horace
* Resolution convolution and Tobyfit

Results

* Overview
* Low energies
  + Stiffness
  + Intensities
  + Damping
  + Comparison with Mook et al
* High energies
  + Overview of features
  + Comparison with Paul and Mook

TD-DFT

* Theory
  + Buczek12
  + Questaal 23
  + Cao15

Discussion

* Energy scale at long wavelength limit
  + We agree with old data
* Intermediate energies (up to 150 meV say)
  + Marked discrepancy with old data
  + Comparison with calculation
* High energy
  + Energy scale
  + Different behavior of [100] direction, P point
  + Additional scattering
* Meaning

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References

1. Kübler, J. *Theory of Itinerant Electron Magnetism, 2nd Edition*. (Oxford University Press, 2021). doi:10.1093/oso/9780192895639.001.0001.

2. Wohlfarth, E. P. Handbook of Ferromagnetic Materials: Chapter 1 Iron, cobalt and nickel. in *Handbook of Ferromagnetic Materials* vol. 1 1–70 (Elsevier, 1980).

3. Collins, M. F., Minkiewicz, V. J., Nathans, R., Passell, L. & Shirane, G. Critical and Spin-Wave Scattering of Neutrons from Iron. *Phys. Rev.* **179**, 417–430 (1969).

4. Paul, D. M., Mitchell, P. W., Mook, H. A. & Steigenberger, U. Observation of itinerant-electron effects on the magnetic excitations of iron. *Phys. Rev. B* **38**, 580–582 (1988).

5. Perring, T. G. *et al.* High‐energy spin waves in bcc iron. *J. Appl. Phys.* **69**, 6219–6221 (1991).

6. Loong, C. ‐K. Neutron scattering investigation of magnetic excitations at high energy transfers (invited). *J. Appl. Phys.* **57**, 3772–3777 (1985).

7. Blackman, J. A., Morgan, T. & Cooke, J. F. Prediction of High-Energy Spin-Wave Excitation in Iron. *Phys Rev Lett* **55**, 2814–2817 (1985).

8. Dimmock, J. O. The Calculation of Electronic Energy Bands by the Augmented Plane Wave Method. in *Solid State Physics* (eds. Ehrenreich, H., Seitz, F. & Turnbull, D.) vol. 26 103–274 (Academic Press, 1971).

9. Cooke, J. F., LYNN, J. & DAVIS, H. Calculations of the dynamic susceptibility of nickel and iron. *Phys. Rev. B* **21**, 4118–4131 (1980).

10. Antropov, V. P., Harmon, B. N. & Smirnov, A. N. Aspects of spin dynamics and magnetic interactions. *J. Magn. Magn. Mater.* **200**, 148–166 (1999).

11. Pajda, M., Kudrnovský, J., Turek, I., Drchal, V. & Bruno, P. Ab initio calculations of exchange interactions, spin-wave stiffness constants, and Curie temperatures of Fe, Co, and Ni. *Phys. Rev. B* **64**, 174402 (2001).

12. Buczek, P., Ernst, A. & Sandratskii, L. M. Different dimensionality trends in the Landau damping of magnons in iron, cobalt, and nickel: Time-dependent density functional study. *Phys. Rev. B* **84**, (2011).

13. Singh, N., Elliott, P., Nautiyal, T., Dewhurst, J. K. & Sharma, S. Adiabatic generalized gradient approximation kernel in time-dependent density functional theory. *Phys. Rev. B* **99**, 035151 (2019).

14. Rousseau, B., Eiguren, A. & Bergara, A. Efficient computation of magnon dispersions within time-dependent density functional theory using maximally localized Wannier functions. *Phys. Rev. B* **85**, 054305 (2012).

15. Cao, K., Lambert, H., Radaelli, P. G. & Giustino, F. Ab initio calculation of spin fluctuation spectra using time-dependent density functional perturbation theory, plane waves, and pseudopotentials. *Phys. Rev. B* **97**, 024420 (2018).

16. Skovhus, T. & Olsen, T. Dynamic transverse magnetic susceptibility in the projector augmented-wave method: Application to Fe, Ni, and Co. *Phys. Rev. B* **103**, 245110 (2021).

17. Durhuus, F. L., Skovhus, T. & Olsen, T. Plane wave implementation of the magnetic force theorem for magnetic exchange constants: application to bulk Fe, Co and Ni. *J. Phys. Condens. Matter* **35**, 105802 (2023).

18. Şaşıoğlu, E., Schindlmayr, A., Friedrich, C., Freimuth, F. & Blügel, S. Wannier-function approach to spin excitations in solids. *Phys. Rev. B* **81**, 054434 (2010).

19. Müller, M. C. T. D., Friedrich, C. & Blügel, S. Acoustic magnons in the long-wavelength limit: Investigating the Goldstone violation in many-body perturbation theory. *Phys. Rev. B* **94**, 064433 (2016).

20. Okumura, H., Sato, K. & Kotani, T. Spin-wave dispersion of $3d$ ferromagnets based on quasiparticle self-consistent $GW$ calculations. *Phys. Rev. B* **100**, 054419 (2019).

21. Lichtenstein, A. I., Katsnelson, M. I. & Kotliar, G. Finite-Temperature Magnetism of Transition Metals: An ab initio Dynamical Mean-Field Theory. *Phys. Rev. Lett.* **87**, 067205 (2001).

22. Pashov, D. *et al.* Questaal: A package of electronic structure methods based on the linear muffin-tin orbital technique. *Comput. Phys. Commun.* **249**, 107065 (2020).

23. Questaal.