**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

Experimental measurements of spin-wave excitations were performed on a 166g 54Fe10At%Si sample in ISIS (UK) facility using inelastic copper spectrometers MAPS23 running four experiments with incident neutron energies Ei=200,400,800 and 1400meV and using rotating crystal to cover the whole momentum transfer range available to the instrument. Multiple energy experiments allow us from one side, cover whole possible range of spin-wave excitations present in iron and from other side, obtain MAPS instrument energy and momentum resolution compatible with the width-s of actual spin wave excitations in the correspondent energy range. The instrumental resolution has been selected to be in range of E/E~4%.

Silicon has been added to iron sample to guarantee thermal stability of the lattice. Theoretical estimates suggest that used amount of silicon should not noticeably affect magnetic properties of bulk iron. The sample has been characterized using ISIS SXD neutron spectrometer24,25, revealing BCC lattice with crystal lattice parameter a=2.844±0.001Å, which corresponds to the values available in the literature (see e.g.26,27) for *FeSi* alloys correspondent concentration at temperature 8K. The change of lattice parameters from the standard iron value a= 2.86 Å also would not visibly affect the magnetic properties of the sample.

Initial inelastic data processing and conversion of raw experimental data into partial(per experiment) magnetic scattering cross-section expressed in mb/meV/sr we use standard Mantid28,29 inelastic data reduction scripts and partial magnetic cross sections results are combined into total differential magnetic cross-section and analysed using Horace&Tobyfit30,31 data analysis package.

# 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 32
  + 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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