Neutron scattering and its applications to study magnetic excitations.

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Abstract
Neutron scattering techniques are an essential tool for the study of magnetic properties of materials. Magnetic as well as nuclear interacting mechanisms are described together with differential cross sections for both elastic and inelastic neutron scattering. The seminar focuses on magnetic inelastic scattering which is used for measurements of magnetic excitations. These excitations are modelled with a spin-only Hamiltonian of a quantum many-body system. Antiferromagnetic Heisenberg chain is analysed using the linear spin-wave theory and the dispersion relation for its magnetic excitations is derived. The derived solution describes the low energy excitations of the ground state, named *magnons* or spin-waves. Neutron scattering measurements on CuO are presented, as they can be precisely modelled using the presented linear spin-wave theory, allowing the extraction of the main exchange interactions.
1 Introduction

Scattering is a physical process where some form of radiation, e.g. light, sound, or moving particles, is forced to deviate from a straight trajectory. Typically this occurs due to localized non-uniformities in the medium through which they pass. From the analysis of the scattered part the information regarding the medium can be obtained. One of the most known techniques is the X-ray scattering which reveals information about the crystal and electronic structure and corresponding excitations. Another valuable tool for investigating many important features of matter are neutrons. A major advantage of neutrons compared to X-rays is that they possess a magnetic dipole moment and can therefore interact with the unpaired electrons in the crystal, leading to the so-called magnetic neutron scattering. Two types of magnetic neutron scattering are described in this paper. Elastic scattering that gives the information on the arrangement of electron spins and the inelastic scattering that gives the energies of the magnetic excitations. Neutron scattering is therefore essential for the study of magnetic properties of materials and their scattering is a versatile technique that is well suited to study atomic and magnetic structures as well as excitation properties of strongly correlated electron systems. Several neutron properties, important for understanding neutron scattering are listed in Tab. 1.

<table>
<thead>
<tr>
<th>Basic properties of neutron:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rest mass</td>
</tr>
<tr>
<td>$m_n = 1.001374 m_p$</td>
</tr>
<tr>
<td>Charge</td>
</tr>
<tr>
<td>0</td>
</tr>
<tr>
<td>Spin</td>
</tr>
<tr>
<td>$\frac{1}{2}$</td>
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<tr>
<td>Magnetic dipole moment</td>
</tr>
<tr>
<td>$\mu_n = -1.913 \mu_N$</td>
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</tbody>
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<table>
<thead>
<tr>
<th>Values of physical constants:</th>
</tr>
</thead>
<tbody>
<tr>
<td>Elementary charge</td>
</tr>
<tr>
<td>$e = 1.602 \times 10^{-19} \text{C}$</td>
</tr>
<tr>
<td>Proton rest mass</td>
</tr>
<tr>
<td>$m_p = 1.673 \times 10^{-27} \text{kg}$</td>
</tr>
<tr>
<td>Nuclear magneton</td>
</tr>
<tr>
<td>$\mu_N = 5.051 \times 10^{-27} \text{J T}^{-1}$</td>
</tr>
</tbody>
</table>

Table 1: Basic properties of the neutron and values of physical constants.
The de Broglie wavelength of "thermal" neutrons (energy: 1-100 meV) is comparable to interatomic spacings in solids and liquids ($\approx \AA$). Thus, interference effects occur, which yield information on the structure of the scattering system. Relation between energy $E$ and wavelength $\lambda$ is $E = \frac{h \nu}{\lambda}$, where $h$ is the Planck constant and $c$ the speed of light. Because the neutron has no electrical charge and it can penetrate deep into the matter, and more importantly, it comes close to the atoms’ nuclei, because there is no Coulomb barrier to overcome. Neutrons are thus scattered from the nuclei due to nuclear forces and the scattering depends on the number of protons and neutrons in the nucleus. An important example is hydrogen which is virtually transparent to X-rays but has high probability for neutron scattering.

From the production viewpoint, the energetic neutrons produced in the nuclear reactors and spallation sources can be slowed down, or moderated, by collisions with atoms of similar mass, such as hydrogen or deuterium. The neutrons energy spectrum is therefore well determined and with the use of the so-called *Time of Flight* method, neutrons with exact wanted energy are obtained. With the evolution of nuclear reactors, that allow high neutron fluxes and development of subsequent processing that offers neutrons with well determined energy spectrum, neutron scattering has evolved in a powerful tool to study condensed matter.

This seminar focuses on the use of neutrons for magnetic-excitations studies. In the first part of the seminar scattering theory is presented together with theoretical analaysis of magnetism with the introduction of quasi-particles *magnons* on a simple one-dimensional antiferromagnetic Heisenber chain. The second part focuses on experiments, starting with neutron sources and followed by the description of the three-axis spectrometer, which made neutron scattering widely accepted. At the end, the comparison of neutron scattering measurements and the theory is presented.

2 Theory

2.1 Scattering Theory

The scattering of a photon or a neutron on a sample is characterized by the resultant change in its momentum $\mathbf{P}$, and energy $E$. This is shown schematically in Fig. 1 where a particle incident with a wave-vector $\mathbf{k}_i$ and angular frequency $\omega_i$ emerges with a final wave-vector $\mathbf{k}_f$ and frequency $\omega_f$.

![Figure 1: A schematic representation of a particle being scatter by a sample.](image)

The momentum transfer can be expressed as

$$ \mathbf{P} = h\mathbf{k}_i - h\mathbf{k}_f = h\mathbf{Q}, $$

(1)
where the $\hbar = \frac{h}{2\pi}$ is the Planck constant and

$$Q = k_i - k_f,$$  \hspace{1cm} (2)

and similarly, the energy transfer can be expressed as

$$E = \hbar \omega \quad \text{where} \quad \omega = \omega_i - \omega_f. \hspace{1cm} (3)$$

The momentum and energy gained by the scattered particle is equal to that lost by the sample and vice versa. For now let’s limit ourselves only to neutron scattering where the number of neutrons that are scattered per second into a certain direction with a certain range of energy values is expressed as the partial differential cross section. To obtain the expression we consider the probability of a transition of the neutron-target system (sample) from an initial state $\lambda_i$ to a final state $\lambda_f$. The interacting potential between the neutron and the target can be treated as a perturbation and Fermi’s Golden Rule can be applied to calculate the transition probability. The partial differential scattering cross section for the whole process is a sum over all initial and final states of the system and over all possible initial and final spin states of neutron \([1], [2]\):

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i} \left( \frac{m_n}{2\pi\hbar^2} \right)^2 \sum_{\sigma, \sigma_f, \lambda_i, \lambda_f} p_{\lambda_i} |\langle \sigma_f \lambda_f | V(Q) | \sigma_i \lambda_i \rangle|^2 \delta(E_{\lambda_f} - E_{\lambda_i} - \hbar \omega), \hspace{1cm} (4)$$

where $V(Q)$ is the Fourier transform of the neutron-matter interaction potential $V(r)$, and $p_{\lambda_i}$ is the probability distribution for initial state $\lambda_i$ and $p_{\sigma_i}$ is the probability distribution for the initial spin state of the neutron $\sigma_i$. The scattering cross section is therefore dependent on the type of interaction between the neutron and the matter it scatters from, described by the interaction potential $V(r)$.

The main advantage for the use of neutrons to investigate magnetic properties is that they do not carry an electric charge, but they have an intrinsic magnetic moment. Therefore they interact only with magnetic moments (via magnetic dipole-dipole interaction) and atoms’ nuclei (via nuclear interaction). They can easily penetrate the sample and form a scattering pattern that is a combination of both, nuclear and magnetic contributions. Hence information about the crystal and magnetic structure is obtained simultaneously. Furthermore, the scattering can be divided into elastic ($E_f = E_i$), so-called Bragg scattering and inelastic ($E_f \neq E_i$), as presented on Fig. \([2]\). From elastic information about atomic and magnetic structure of the crystal is obtained and with inelastic the excitations of the system are measured e.g., atomic motion - phonons or magnetic excitations - spin waves or magnons.

![Figure 2: Schematic diagram of Bragg scattering \((a)\) and inelastic scattering, where the neutron gains energy \((b)\)](image)

\((a)\) Elastic scattering $|k_i| = |k_f|$

\((b)\) Inelastic scattering $|k_i| \neq |k_f|$

2.1.1 The Nuclear Interaction

In any scattering experiment the dominant contribution to the total scattering will come from nuclear elastic scattering which arises from the neutron interacting with nuclei in the sample.
through the strong nuclear force. Neutrons interact with nuclei \( j \) at positions \( r_j \) through an interaction potential which can be approximated by \[1\]

\[
V_N(r) = \frac{2\pi\hbar^2}{m_n} \sum_j b_j \delta(r - r_j),
\]

where \( b_j \) are the scattering lengths of each atomic nucleus. The partial differential cross section can be expressed as a sum of coherent and incoherent terms. The coherent terms represents the interference effects between the nuclei and the neutron as elastic Bragg scattering (correlations of atomic positions) and inelastic phonon scattering (atomic motion). Incoherent terms grant no useful information and are for most nuclei negligible. Yet, for some nuclei, e.g., proton and vanadium, they are strong and may overshadow the coherent scattering. Coherent nuclear elastic cross section is given by \[1\]

\[
\left( \frac{d^2\sigma}{d\Omega dE_f} \right)_{\text{nuclear elastic}} = N \frac{(2\pi)^3}{V_0} |F_N(Q)|^2,
\]

where \( N \) is the number of unit cells in the crystal, \( V_0 \) is the volume of the unit cell and \( F_N \) is the nuclear structure factor given as

\[
F_N(Q) = \sum_j b_j e^{iQr_j} e^{-W_j(Q,T)}.
\]

Here the sum runs over all atoms \( j \). The Debye-Waller factor \( W_j \) takes into account that atoms are not frozen to their lattice sites, but they rather experience a certain amount of thermal motion around an equilibrium position.

### 2.1.2 The Magnetic Interaction

Magnetic scattering of neutrons occurs due to an interaction between the magnetic dipole moment of the incident neutron and the magnetic field due to the intrinsic spin and orbital momentum of unpaired electrons. The neutron magnetic moment is

\[
\mu_n = -\gamma \mu_N \sigma,
\]

where \( \mu_N \) is the nuclear magneton, \( \gamma \approx 1.913 \) is the gyromagnetic ratio and \( \sigma \) is the Pauli spin operator with eigenvalues of \( \pm 1 \). The interaction potential for magnetic scattering takes the form

\[
V_M(r) = -\mu_n \cdot B(r),
\]

where \( B \) represents the local magnetic flux density from the unpaired electrons, due to both their intrinsic spin and angular momentum. The scattering cross-section for magnetic elastic scattering is given by \[1\]

\[
\left( \frac{d^2\sigma}{d\Omega dE_f} \right)_{\text{mag. elastic}} = \frac{N m (2\pi)^3}{V_{0m}} \frac{\gamma r_0}{2} \sum_{\alpha,\beta} \left( \left( \delta_{\alpha,\beta} - \hat{Q}_\alpha \hat{Q}_\beta \right) F^\alpha(Q) F^{\beta\ast}(Q) \right) \delta(Q - \tau_M) \delta(\hbar\omega),
\]

where \( \tau \) is a reciprocal magnetic lattice vector, \( V_{0m} \) is the volume of the magnetic unit cell, \( r_0 \) is the classical electron radius and the sum over \( \alpha \) and \( \beta \) is a sum over all combinations of two Cartesian axes. \( \hat{Q}_\alpha \hat{Q}_\beta \) are the unit vector components in direction of \( Q \). \( F^\alpha(Q) \) is the \( \alpha \) component of the magnetic structure factor, given by

\[
F^\alpha(Q) = \sum_j \mu_j^\alpha f_j(Q)e^{iQr_j}e^{-W_j(Q,T)},
\]
where $\mu_j^\alpha$ is the $\alpha$ component of the magnetic moment of the $j$th ion, $r_j$ is its position within the magnetic unit cell and $f_j$ is the magnetic form factor which arises from dipole approximation of the magnetic interaction.

Large part of this seminar will concentrate on explanation of magnetic excitations, such as magnons. Magnetic excitations are probed by magnetic inelastic scattering measurements and the corresponding cross section is given by

$$\frac{d^2\sigma}{d\Omega dE_f} = \left(\frac{\gamma r_0}{2}\right)^2 f^2(Q)e^{-2W(Q,T)} \frac{k_f}{k_i} \sum_{\alpha,\beta} \left\langle \left(\delta_{\alpha,\beta} - \hat{Q}_\alpha \hat{Q}_\beta\right) S^{\alpha\beta}(Q,\omega) \right\rangle,$$

where $S^{\alpha\beta}$ are the space and time Fourier transforms of the time dependent spin-spin correlation functions

$$S^{\alpha\beta}(Q,\omega) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \sum_{j,j'} e^{iQ|(r_j - r_{j'})} e^{-i\omega t} \left\langle S^{\alpha}_j(0) S^{\beta}_{j'}(t) \right\rangle dt,$$

where $S^{\beta}_j(t)$ is the $\beta$ component of the spin at site $j$ and at time $t$ and $\langle...\rangle$ denotes a statistical average over the initial states of the system. A simplified expression can be written for a system at zero temperature, where the excitations are out of the ground state only, which has a wave function $|0\rangle$ and energy $E_0$, and the spin-spin correlation function is given by

$$S^{\alpha\alpha}(Q,\omega) = \sum_{\lambda} \langle\lambda|S^{\alpha}(Q)|0\rangle^2 \delta(h\omega - E_0 - E_\lambda),$$

where the sum is over all eigenstates $|\lambda\rangle$ of the final state of the system with energy $E_\lambda$. $S^{\alpha}(Q)$ is the Fourier transform of the $\alpha$ component of the spin $S_j^\alpha$.

### 2.1.3 The Response Function $S(Q,\omega)$

It is often convenient to express the cross section in general in terms of the response function $S(Q,\omega)$, which depends only on $Q$ and $\omega$:

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{k_f}{k_i} S(Q,\omega),$$

where $S(Q,\omega)$ is defined so as to absorb all the factors in Eq. except for $(k_f/k_i)$. The advantage of this view is that it factorizes the cross section into a part that depends on the setup of the experiment $(k_f/k_i)$, and a function that depends only on the properties of the system $S(Q,\omega)$. The measured response function with neutron scattering can therefore easily be compared with theory [1] [2].

### 2.2 Magnetic Excitations in the Heisenberg Antiferromagnet

In this section the focus is the case where magnetic moments of the magnetic ions interact solely with their nearest neighbours. In particular, the excitations model with a spin-only Hamiltonian of a quantum many-body system, where only nearest neighbours contribute to the exchange interaction. This is formally written as Heisenberg Hamiltonian [3]:

$$\hat{H} = \sum_{\langle ij \rangle} J_{ij} \hat{S}_i \cdot \hat{S}_j = \sum_{\langle ij \rangle} J_{ij} (\hat{S}^x_i \hat{S}^x_j + \hat{S}^y_i \hat{S}^y_j + \hat{S}^z_i \hat{S}^z_j).$$

Here $J_{ij}$ is the exchange constant and $\langle ij \rangle$ denotes the sum over all nearest neighbours. In the following the focus is on the antiferromagnetic exchange interactions, i.e. $J > 0$, which favor
antiparallel alignment of the magnetic moments [3]. Depending on the magnetic lattice, i.e.,
the arrangement of the magnetic ions, such Hamiltonian can lead to different types of magnetic
ground state, ranging from disordered spin liquids to static long-range orders. For simplicity,
a Neel state on a simple chain is considered, which consists of two ferromagnetic sublattices,
i.e. that spins on sublattice A point in one direction and the spins on sublattice B point in
the opposite direction. This is shown schematically in Fig. 3. Despite the fact that quantum
fluctuations suppress long-range order on a simple spin chain, this example still serves as a
reference for spin-wave calculation.

\[
\hat{S}_B^x = \hat{S}_A^x \quad \hat{S}_B^y = -\hat{S}_A^y \quad \hat{S}_B^z = -\hat{S}_A^z
\]

Figure 3: A schematic representation of the Neél state on an antiferromagnetic spin chain.

The Holstein-Primakoff transformations are used to write the spin operator components \( \hat{S}_x, \hat{S}_\pm = \hat{S}_x \pm i\hat{S}_y \) in terms of Bose operators in the lowest order

\[
\hat{S}^- = (2S)^{1/2}a^\dagger, \quad \hat{S}^+ = (\hat{S}^-)^\dagger, \quad \hat{S}^z = S - a^\dagger a,
\]

for which the commutator is \([a, a^\dagger] = 1\) and \(n = a^\dagger a\). From this the Heisenberg Hamiltonian
in the spin-wave approximation can be written, where the exchange constant \(J\) is equal for all
spins on chain

\[
\hat{H} = -J \sum_i \left( \hat{S}_i^z \hat{S}_{i+1}^z - \frac{1}{2} (\hat{S}_i^+ \hat{S}_{i+1}^- + \hat{S}_i^- \hat{S}_{i+1}^+) \right)
\]

\[
= -NJ S^2 + JS \sum_i \left( 2a_i^\dagger a_i + a_i a_{i+1} + a_{i+1}^\dagger a_i^\dagger \right) + O(S^0).
\]

The Bose operators can be written in terms of their discrete Fourier transform

\[
a_k = \frac{1}{\sqrt{N}} \sum_{n=1}^{N} e^{i k n} a_n, \quad a_n = \frac{1}{\sqrt{N}} \sum_k e^{-i k n} a_k, \quad [a_k, a_{k'}^\dagger] = \delta_{kk'} \]

and used to write the Hamiltonian in terms of \(a_k\):

\[
\hat{H} = -NJ S(S + 1) + JS \sum_k \left( a_k^\dagger a_k - \frac{1}{2} \right) \left( \frac{1}{\cos k} \begin{pmatrix} 1 & \cos k \\ \cos k & 1 \end{pmatrix} \right) a_k^\dagger a_k + O(S^0).
\]

The Bogoliubov transformation (preserves commutation relations) of \(a_k\) and \(a_{-k}^\dagger\) is used

\[
\left( \begin{array}{c} \alpha_k \\ \alpha_{-k}^\dagger \end{array} \right) = \left( \begin{array}{cc} \cosh \theta_k & \sinh \theta_k \\ \sinh \theta_k & \cosh \theta_k \end{array} \right) \left( \begin{array}{c} a_k \\ a_{-k}^\dagger \end{array} \right)
\]

and removing the off-diagonal terms by setting \(\cosh(2\theta_k) = \sqrt{1 - \cos^2 k} = |\sin k|\) to achieve
the final solution of Hamiltonian

\[
\hat{H} = -NJ S^2 + 2JS \sum_k |\sin k| |\alpha_k^\dagger \alpha_k + \frac{1}{2}| + O(S^0)
\]

with the dispersion relation

\[
\omega_k = 2JS |\sin k|.
\]
3 Inelastic neutron scattering experiment

This section will focus on experimental equipment used in order to determine magnetic excitation with neutron scattering. First part will focus on neutron sources, second on the Three Axis Spectrometer, where all the main parts of the instrument will be described. In the last part measurements on CuO (cupric oxide) will be presented where magnon excitations were measured with inelastic neutron scattering.

3.1 Neutron Sources

The neutrons used in a scattering experiment can be obtained from a nuclear reactor, with the nuclear fission with fissile material, i.e., $^{235}\text{U}$, as described in Eq. (26), or from a spallation source, as described in Eq. (27), where the neutrons are produced by bombarding a heavy target with high-energy protons

\[
n + ^{235}\text{U} \rightarrow 2.5n + \text{nuclei} + 200 \text{MeV} \quad \text{(26)}
\]

\[
p + \text{heavy nucleus} \rightarrow 20n + \text{energy.} \quad \text{(27)}
\]

Neutrons in spallation sources come as a pulse in which 20-30 neutrons are expelled after each impact of proton, while in nuclear reactors neutrons are produced continuously in time where the neutron flux of current generation of high-flux reactors is $\approx 1 \times 10^{15}$ neutrons/$\text{cm}^2$/s, which is higher than spallation sources. Neutrons produced from sources are transported through beam tubes to the instruments, where measurements are performed. The shielding around the sources and the beam tubes contains a combination of materials that can scatter the fast neutrons and absorb them. In addition, a significant amount of lead is needed to absorb the dangerous $\gamma$-rays. For experiments different energies of neutrons are used. Neutrons produced from the sources are fast (hot neutrons) with energies $> 2$ MeV. They can be slowed down (cold neutrons) with collisions on light nuclei (hydrogen). These materials are called moderators which in combination with neutron-energy dependent absorbing filters enable us to get the desired spectrum of neutron energies.

3.2 Three Axis Spectrometer

The three-axis spectrometer is the most versatile instrument for the inelastic neutron scattering measurements, because it allows one to probe a broad region of the energy-momentum phase space. It was developed by Brockhouse in 1961. The derivation of the name Three Axis Spectrometer becomes clear from an inspection of Fig. 4.
There are three vertical axes about which parts of the instrument rotate. The first, labelled monochromator, allows a narrow band of neutron wavelengths to be chosen from the broad spectrum coming from the neutron source. The spectrum of this band is described with Bragg’s law for elastic neutron scattering. The angle of scattered neutrons is defined by the collimator located before the sample, which determines the width (in wavelength) of the spectrum of neutrons coming to the sample. The second axis of rotation passes through the sample. The rotation around this axis allows for the investigation of the neutron scattering properties of the sample as a function of the scattering angle. The third axis passes through the analyser crystal, which determines the center and the width of the band of neutron wavelengths which will finally fall on the detector and thus contribute to the measured signal. The principle by which this is accomplished is the same as that used in the case of the monochromator. The band width will be determined here by the angular divergence of the last two collimators and the characteristics of the analyser. The great power of this spectrometer is that it allows almost arbitrary choice of, $Q$ and $\omega$, which determine the momentum and energy transferred to the sample, respectively. This allows measuring previously defined response function $S(Q, \omega)$.

### 3.3 Magnetic Excitations in CuO

Despite its simple chemical formula, cupric oxide (CuO) (crystal and magnetic structure presented in Fig.5) has a surprisingly complex magnetic behaviour which remains only partly
understood. The crystal structure of CuO, shown in Fig. 5a, is monoclinic. Each Cu atom is surrounded by four coplanar O atoms making square plaquette. Two, more distant, O atoms above and below the plaquette complete a highly distorted octahedron. The structure can be regarded as having two types of buckled Cu-O chains. The magnetic structure of low temperature ordered moment is illustrated in Fig. 5b. Here we present the latest study \(^8\), where the neutron magnetic inelastic scattering was used to analyze the spin dynamics in the low temperature antiferromagnetic (AFM) phase of CuO. The low-energy excitation spectrum shows a well defined spin-wave dispersion, the linear spin-wave model for a Heisenberg AFM provides a very good description of the measured data, presented on Fig. 6.

![Crystal structure of CuO](image1)

![The antiferromagnetic structure on two chains](image2)

Figure 5: Crystal and magnetic structures of CuO. (a) four unit cells of the crystal structure of CuO with two CuO\(_4\) plaquettes highlighted. The red and blue arrows indicate the two chains shown in (b).

The measured response function, presented on Fig 6a, and the results of the linear spin-wave theory, presented on Fig. 6b, are in good agreement. We can conclude that the linear spin-wave theory, allowing for calculation of the magnon dispersion relation, is accurate for the description of low-energy magnetic excitations of CuO.

![Measured magnon spectrum](image3)

![Calculated magnon spectrum](image4)

Figure 6: The magnon excitation spectrum measured in CuO by neutron scattering, presented on Fig. 6a, showing the \(S(Q, \omega)\) as a function of energy transfer and wave vector along the chain. Right Fig. 6b, show the plotted linear spin-wave theory results for magnon excitations.
4 Conclusion

Neutron scattering is a powerful tool for investigating material properties. One of the most important applications are studies of crystal magnetic structures and their excitations, because they possess a magnetic dipole moment and can interact with unpaired electrons. Two different types of scattering were described, divided by energy transfer (elastic and inelastic), which are a consequence of nuclear or magnetic interaction. Linear spin-wave theory on Heisenberg antiferromagnet was used to introduce quasi-particles magnons in order to explain the measurements performed with Triple Axis Spectrometer on CuO. In the presented study, the excitation spectrum has been modelled using linear spin-wave theory with a high accuracy, which enable to extract the strength of the main exchange interactions.

References


