Journal of Magnetism and Magnetic Materials 350 (2014) 209–216

Contents lists available at ScienceDirect

Journal of Magnetism and Magnetic Materials

journal homepage: www.elsevier.com/locate/jmmm

Current Perspectives

Magnetic frustration probed by inelastic neutron scattering: Recent examples

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ARTICLE INFO

Available online 14 November 2013

Keywords:
Magnetic frustration
Neutron

ABSTRACT

Frustration, either due to the lattice geometry of competing interactions is now days the main source of exotic magnetic ground states, potentially tunable. Inelastic neutron scattering is a unique tool to study them at a microscopic level, by measuring the spin excitations, further modeled by interactions schemes. Some recent examples of this powerful technique are reviewed, involving three types of frustrated compounds: the pyrochlore magnets, the multiferroics and the chiral magnets.

1. Introduction

Magnetic frustration, namely the inability of a system to satisfy all pair of interactions simultaneously, has been for a long time one of the main research areas in condensed matter physics. It is the source of many exotic grounds states and excitations, whose description remains challenging both for theoreticians and experimentalists. Inelastic neutron scattering (INS) provides a microscopic description of these states in many aspects. It probes relaxation phenomena at short times scales, as well as spin excitations from a given ground state yielding the exchange interactions, and it also gives access to spin correlations.

The interest in frustrated magnets started in the 1980s, the main goal of research involving compounds with frozen chemical disorder, the so-called spin glasses or reentrant spin glasses [1]. Here frustration arises from a random distribution of competing ferromagnetic (F) and antiferromagnetic (AF) interactions. Mean field approaches predict a new kind of phase transition, which recalls the glass transition since the transition temperature is frequency dependent. The spin glass freezing transition is associated with the onset of a static local order parameter, but the ground state remains highly degenerated, since many configurations are energetically equivalent. INS greatly contributed to the knowledge of the spin dynamics associated with the spin glass freezing by probing the existence of a wide spectrum of relaxation times [2]. In model metallic spin glasses such as CuMn alloys, INS combined with other probes showed that the spin dynamics extends over more than ten time decades, greatly extending the information drawn from macroscopic susceptibility, muon or NMR data. In reentrant spin glasses with a dominant F interaction, spin waves measurements in the low temperature region where magnetization collapses, unambiguously probed the existence of a mixed phase which retains long range order [3], as predicted by mean field theory.

More recently, the interest of the community has switched to chemically ordered compounds. In geometrically frustrated magnets (GFMs), frustration arises from the lattice, built from weakly connected entities such as triangles or tetrahedra. The simplest example of a geometrically frustrated unit is a regular triangle of Ising spins coupled by AF interactions. It points out the three necessary ingredients of such frustration, namely: (i) the nature of first neighbor interaction (F or AF); (ii) the spin anisotropy (Ising, XY, Heisenberg); and (iii) the lattice geometry (How are the locally frustrated units connected?). From this simple starting point, a wide variety of exotic magnetic ground states and spin excitations can be stabilized. The most famous types, found in rare earth titanate pyrochlores $R_2Ti_2O_7$, are short range ordered and recall thermodynamical states of matter, leading to the concepts of spin liquids and spin ices [4], which emerge from a correlated paramagnetic state without well defined transition. Here too, the degeneracy of the magnetic ground state suggests the possibility of low energy excitations, or soft modes [5,6], between equivalent configurations, which can be studied by INS. More classically, INS also probes crystal field and cooperative excitations, yielding a direct access to the interactions and the local anisotropy.

Keeping a chemically ordered lattice, the competition of several magnetic interactions, or the coupling of several degrees of freedom (spin, orbit) or order parameters (elastic, magnetic, electric) is the source of complex magnetic orders [7]. Here, magnetic frustration prevents a simple Néel or F state to settle in, opening the route for more complex non-collinear or cycloidal orders, potentially tunable. In multiferroics, INS may probe the existence of the spin lattice coupling through hybrid phonon–magnon modes, and yields precious information about the competing interactions at play.

Chiral magnetic phases may result from the competition of magnetic interactions, or from the competition of a ferromagnetic

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0304-8853/$ - see front matter
http://dx.doi.org/10.1016/j.jmmm.2013.11.001

exchange and Dzyaloshinskii–Moryia (DM) anisotropy [8,9]. In centrosymmetric crystals, chiral properties usually cancel due to the presence of helical domains of equivalent populations. But interesting effects arise when the lattice lacks an inversion symmetry. Topological defects may be created [10], analogous to liquid crystal phases. Enantiopure magnetic structures with single helicity can be stabilized. Good examples of such cases are the itinerant magnet MnSi with cubic structure and the insulating langasites with triangular symmetry. Here, polarized inelastic neutron scattering probes the chirality directly, either in the ordered or in the paramagnetic phase.

Whatever the physics concerned, the information deduced from INS data must be associated to that obtained from Bragg diffraction or from diffuse scattering, namely by integrating the neutron response over all the energies of the out-coming neutron beam. Bragg diffraction measures the static Long Range Order (LRO) if it exists, whereas diffuse scattering yields the Fourier transform of the instantaneous magnetic correlations in the case of Short Range Order (SRO). Performed at very low temperature, these techniques give access to the magnetic ground state, whose knowledge is necessary to analyze and model the magnetic excitations. In multiferroics and chiral magnets where LRO dominates, Bragg diffraction provides a direct determination of the helical or cycloidal magnetic structures. In rare earth pyrochlores like spin ices where SRO exist alone, the diffuse scattering cross-section show pinch points, fingerprints of algebraic correlations, Coulomb phase and its peculiar excitations known as magnetic monopoles.

In all cases, the high complexity of the short or long range ordered magnetic phases induced by frustration, as well as their excitations can be unraveled using state of art neutron instrumentation. Spectrometers combining a high intensity with a high energy-resolution are now available, offering the possibility to map the reciprocal and energy space in great details from single crystal data. Energy analysis combined with polarization analysis of the outcoming neutron beam yields a clear determination of the spin components involved in the scattering process. Such experiments require sophisticated data treatment and software to analyze and simulate the neutron cross-sections. In turn, these results stimulate more and more elaborate theories. This paper shortly reviews a few recent examples of such cases.

2. Pyrochlore magnets

The pyrochlore lattice of Fd-3m symmetry, made from corner sharing tetrahedra (Fig. 1a) is the model system to study geometrical frustration in three dimensions [4]. For Heisenberg spins coupled via AF interactions, early mean field calculations predict this lattice to be fully frustrated [12]. The resulting short range ordered ground state with low energy flat modes has been called spin liquid or cooperative paramagnet. It has a continuous degeneracy since an infinite number of configurations minimize the energy in a given tetrahedron, keeping the local magnetization to zero (Fig. 1b). Recent calculations of the spin dynamics predict a relaxational behavior with a gradual temperature freezing and strongly anisotropic susceptibility [6].

Experimentally, $Tb_2Ti_2O_7$ shows spin liquid ground state of strongly correlated moments, which keep fluctuating down to the base temperature of 50 mK [13]. Since its scheme of interactions is quite far from simple Heisenberg AF, the reason for this behavior has remained a theoretical and experimental challenge for many years [14], and is still highly debated [15,16]. INS plays a major role in this debate, since a key feature of the spin liquid state resides on the crystal field (CF). The most important and controversial observation is that of a low energy excitation [17,18]. Taking into account the non-Kramers nature of the Tb^{3+} ion, it cannot result from the influence of a molecular field and it was attributed to the splitting of the GS crystal field doublet by a tetragonal distortion [15]. The resultant symmetry breaking induces a two-singlet ground state with entangled or tunnel-like wave functions, and large matrix elements between them, yielding a strong inelastic line at an energy close to the splitting of the GS doublet. Recent high flux and high resolution INS measurements [19] yield a precise determination of this excitation in wavevector and energy space. The very typical energy map in Fig. 2 is well accounted for by a simulation taking into account the symmetry breaking of the

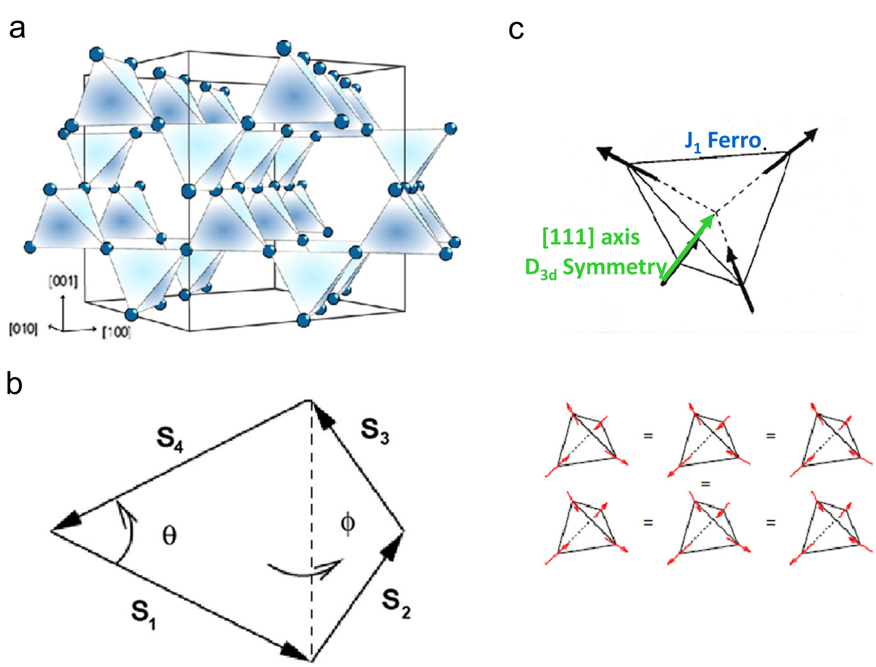

Fig. 1. (a) The pyrochlore lattice made of corner sharing tetrahedron. (b) Frustration of AF interactions. In a tetrahedron of Heisenberg spins coupled by AF interactions, the ground state configuration has continuous degeneracy (spin liquid case). (c) Frustration of ferromagnetic interactions. In a tetrahedron of Ising spins, F coupled and constrained by anisotropy along the local [111] axes, the "two-in-two out" state which minimizes the energy has six equivalent configurations (spin ice case).