Magnetic Structures and Their Determination

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Abstract. Neutron scattering is one of the most spread methods in solid state physics and soft matter studies. Electromagnetic interaction of neutron magnetic moment with periodically arranged moments of unpaired electrons leads to scattering phenomena similar to X-ray diffraction on spatially arranged atoms. Although are these phenomena similar they have their own specific features and problems. In this contribution we describe briefly experimental method of neutron diffraction on magnetic substances and the data analysis leading to determination of spatial distribution of magnetic moments.

Introduction

Over the last fifty years neutron scattering has been a valuable tool in solid state physics for probing the microscopic structure and processes in complex matter. Neutron scattering has decisively contributed to our knowledge of most magnetic phenomena from the experimental discovery of antiferromagnetism in the early 1950s to the first observation of spin fluctuations in high \( T_c \) superconductors most recently. In this contribution we describe briefly a small portion of experimental methods using neutrons, namely diffraction on magnetic substances and the main steps in the data reduction.

Basics of the neutron scattering and Magnetic Structures

Neutron has zero charge and non-zero magnetic moment. Therefore it can penetrate comparing to other charged particles or X-ray much deeper into solids and interacts on one side with nuclei (via strong interaction) on the other side with unpaired electrons (via electromagnetic interaction). Neutrons moderated at room temperatures have both the wavelengths and energies comparable with atomic distances and thermal excitations of the material. Neutrons scattered elastically (we neglect inelastic processes in which neutrons loose or gain energy) from a crystalline material exhibit constructive interferences due to a periodic distribution of atoms and magnetic moments (if they exist) [1]. The nuclear part is present at all temperatures but the magnetic one only below the magnetic phase transition temperature. The latter one further strongly depends on the scattering vector \( Q \) because of magnetic form factor, which decays with \( Q \) in a way typical for a given magnetic ion. On top of that a strong directional dependence with respect to the direction of \( Q \) exists. The
experiment is sensitive only to components of the magnetization perpendicular to $Q$. By subtraction of the reference signal obtained above the magnetic phase transition temperature one can get roughly the signal caused by magnetic moments.

Fig 1: Examples of magnetic structures (projected in two dimensions) in the real space (left) and in the reciprocal space (right). Nuclear (magnetic) reflections are denoted by closed (open) circles. (a) AF with a cell doubling along the $a$ axis with propagation vector $q = (1/2, 0, 0)$, (b) AF with a cell doubling along the $a$ and $b$ axes with $q = (1/2, 1/2, 0)$, (c) $F$ structure with the same size of magnetic and nuclear unit cell and $q = (0, 0, 0)$ and (d) ferrimagnetic structure consisting of two different magnetic moments the same size of magnetic and nuclear unit cell and $q = (0, 0, 0)$.

There are two basic types of magnetic structures: antiferromagnetic (AF, for each magnetic moment there exists exactly one that has the same magnitude but opposite direction) and ferromagnetic (F, having non-zero total magnetization). However, there are many different geometrical ways how to arrange individual magnetic moments leading to a large variety of different magnetic structures. Few very simple examples are shown in Fig. 1. In the case that some AF structure has larger periodicity then the parent crystal structure, are new Bragg reflections indexable by non-integer but rational numbers (in Fig. 1 cases (a) and (b)) defining the propagation vector(s) in the reciprocal space. If the size of magnetic and crystal structure is identical (for all the F structures - case c) only the intensity of some of the nuclear reflection increases below the magnetic transition temperature. In Fig. 1d we show an example of ferrimagnetic structure which involves two different magnetic ions.

**Magnetic Structure Determination**

Complete determination of some magnetic structure involves determination of 1, the magnetic phase transition, 2, the propagation vector 3, the magnetic moment directions and 4, the magnitudes of the moments. Point 1, is almost always known from bulk measurement. The determination of the propagation vector could be in fact the most difficult part of the magnetic structure determination. In the case of powder measurement it is easy to observe magnetic Bragg reflections that are necessary to be indexed. There exists a relatively
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A straightforward geometrical method [2], in which one compares radii of Ewald spheres with slices of the reciprocal space. The aim is to find such propagation vector(s) that would explain all the Ewald spheres observed in the powder experiment. In the case of a single-crystal experiment one has to search the reciprocal space directly.

From the knowledge, which symmetry operations leave the propagation vector invariant, it is possible to construct with the help of the group theory [3] all the irreducible representations (irr), to determine which magnetic moments are coupled by symmetry and how the basis vectors of the magnetic structures transforms under all the allowed symmetry elements for the given irr. Normally, the magnetic structure belongs only to a single irr (magnetic phase transition usually lowers the symmetry). Disadvantage of the method is that it could be numerically tedious. By fitting the experimental data to allowed models one arrives to the most probable solution which leads also the magnitudes of magnetic moments.

Limitations of the neutron scattering

Among the main limitations of neutron scattering belong besides the sensitivity (for powder samples 0.2-0.4 $\mu_B$, for single crystals probed by unpolarized neutrons 0.1 $\mu_B$ and probed by polarized neutrons 0.01 $\mu_B$ or even better) and necessity to apply various correction factors (absorption, extinction) also the inability to determine directions of magnetic moments in highly-symmetrical systems and ambiguity regarding the various types of magnetic domains. There are two basic types of domains; so-called K domains that exist in structures with several equivalent propagation vectors, and S domains that differ in the direction of magnetic moments within one K domain (Fig. 2). In zero field and ambient pressure are all domains normally equally populated. The equilibrium can be affected either by applied magnetic field or by uniaxial pressure that favours one of the K or S domain leading to changes in intensities. In this way one can resolve one of the principal problems of magnetic structure determinations: whether the structure consists from several single-K domains or whether is the structure described by multiple propagation vectors.

References