Magnetic Order

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The vocabulary of magnetism has no strict rules, but for most people working in the field use the term *magnetic materials* to mean materials where spontaneous magnetic ordering takes place. The stability of the ordered state is due to the exchange interaction; at high temperature thermal agitation prevents ordering. Accordingly, the two important aspects in the study of magnetic order concern the nature of the order, i.e. the ground-state arrangement of the magnetic moments, and the process of ordering itself, that is, the phase transition and critical behaviour near the transition temperature. In fact, models of magnetic systems and experiments on materials well described by such models have been central in the study of critical phenomena. This lecture will be about the types of order and the models for the interactions that bring about the ordered states.

The simple classification of ordered states into ferromgnets, antiferromagnets and ferrimagnets cannot do justice to the rich variety of possible ordered patterns of three-dimensional vectors at the sites of all crystal lattices possible in three dimensions. To begin with, we associate these magnets with collinear structures, $\uparrow\uparrow\uparrow\uparrow\uparrow$, $\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow$ and $\uparrow\uparrow\downarrow\uparrow\uparrow\downarrow$, whereas the angle between neighbouring magnetic moments is not limited to zero and π and the orientation of moments with respect to the crystal axes is also an important variable, which can indeed vary with temperature. Apart from the exchange interaction, magneto crystalline anisotropy is also an important factor in determining the nature of the ordered state.

In the rare-earth – transition-metal ferromagnets, the anisotropy energy of the rare earth component is much larger than that of the transition metal, because the spin-orbit coupling is an order of magnitude stronger. As the orientations preferred by these anisotropy energies can be different and so are their temperature dependences, the overall effect is also temperature dependent, so that the orientation of the net ferromagnetic moment changes with temperature.

Similar effects in antiferromagnets are not conspicuous, because there is no net magnetisation. However, some antiferromagnets, most notably $\alpha\text{-}Fe_2O_3$, $MnCO_3$, $CoCO_3$ and CrF_3, show a small magnetic moment ("weak ferromagnets"), which signals a slight noncollinearity of the oppositely oriented moments, due to an anisotropic superexchange interaction. The orientation of magnetic moments in antiferromagnets and ferrimagnets can only be measured by means of neutron diffraction.

The concept of sublattice magnetisation is an important tool in the description of antiferromagnetic and ferrimagnetic ordering. Two sublattices will suffice for the simplest antiferromagnets and the ferrimagnets where the two sublattices are occupied by different ions, carrying moments of different magnitude. Most of the common ferrimagnets are more complicated and need a model with many superlattices. For

instance, magnetite, Fe_3O_4, can be looked upon as being built up of two sublattices occupied by Fe^{3+} ions whose magnetic moments cancel each other and a sublattice of Fe^{2+} ions, which provide the measured magnetisation of approximately 4 Bohr magnetons per formula unit. It should be noted that superlattices are not suitable for all antiferromagnetic structures (ordered structures with zero net magnetisation). In spiral structures, which are quite common with rare-earth metals, the magnetic moments in a crystallographic plane are rotated by a given angle with respect to those in a neighbouring plane. Clearly, if this angle is not $\pi/2$, the structure is not collinear and if it is not a rational fraction of 2π, each plane would have to be defined as a distinct sublattice, which is not very helpful.

Magnetic ordering always breaks at least one symmetry of the crystal, the invariance under time inversion. Invariance under rotations around axes not coinciding with the ordered moments also disappears. This symmetry braking is not related to asymmetries in the spin Hamiltonian. Spin Hamiltonians are constructed to describe the behaviour of the degrees of freedom of atoms or ions occupying lattice sites. Therefore, they have the full symmetry of the lattice. For instance, in a cubic lattice the bilinear term describing the interaction between magnetic moments must be of the Heisenberg form,

$$H_{Heisenberg} = -\frac{J}{2}\sum_{i,k}\left(S_x^{(i)}S_x^{(j)} + S_y^{(i)}S_y^{(j)} + S_z^{(i)}S_z^{(j)}\right).$$

Here only nearest-neighbour interaction is included. Nearest neighbours are equivalent in a cubic lattice, hence the unique exchange parameter J. In a tetragonal lattice, z is not equivalent with x and y, the interaction Hamiltonian will include two exchange parameters and have the form

$$H = -\frac{1}{2}\sum_{i,k}\left[J\left(S_x^{(i)}S_x^{(j)} + S_y^{(i)}S_y^{(j)}\right) + J'S_z^{(i)}S_z^{(j)}\right].$$

Evidently, anisotropic exchange is allowed. Disregarding single-ion magneto crystalline anisotropy, if $J'/J > 1$, the latter Hamiltonian describes a magnet with easy-axis anisotropy, if $J'/J < 1$, with easy-plane anisotropy. In the case of extreme anisotropy we find the Hamiltonians which define two import models:

$$H_{Ising} = -\frac{J}{2}\sum_{i,k}S_z^{(i)}S_z^{(j)} \; ;$$

$$H_{XY} = -\frac{J}{2}\sum_{i,k}\left(S_x^{(i)}S_x^{(j)} + S_y^{(i)}S_y^{(j)}\right).$$

As the Ising and XY models exclude some components of the spin vectors, the entities they describe can be looked upon as one- and two-dimensional moments, respectively. However, this does not automatically make them low-dimensional models. The dimensionality is determined by the meaning of the indices i and j. Those indicate sites in a lattice of well defined dimension. Thus one can study three-dimensional Ising models or one-dimensional XY models; there is no contradiction in terms here.