Magnetic Ordering: Some Structural Aspects

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Abstract

An overview of some structural aspects of magnetic ordering is presented. Magnetic symmetry operations, point groups, and Bravais lattices will be utilized to describe the magnetic symmetry of various magnetic materials. Throughout the talk, the utilization of the theory of magnetic symmetry will be emphasized. The magnetic space groups of Fe, Co, Ni, α'-FeCo, CoPt, $CrPt_3$, and Cu_2MnAl will be examined.

Introduction

One of the most interesting and perhaps most neglected phase transformations in the solid state is the *paramagnetic* to *ferromagnetic* transformation. One of the reasons for this is that for many years in the metallurgical community, the transformation was not thought to be a transformation at all! Indeed the paramagnetic β phase in Fe was removed from the iron phase diagrams and replaced with the symbol used for ferromagnetic α-Fe. This confusion amongst metallurgists arose from a faulty understanding of symmetry and its relationship to "structure" and the definition of "phase".

A common definition of phase is "…a portion of the system whose properties and composition are homogeneous and which is physically distinct from other parts of the system"[1]. If we accept this definition it alone, demonstrates that that *paramagnetic* to *ferromagnetic* "change" is indeed a phase change, since a paramagnetic phase has different magnetic properties than a ferromagnetic phase. In older definitions, a phase was said to have a distinct "structure" and it is here where the problem arose. As far as could be determined by x-ray diffraction, the structure of ferromagnetic Fe was the same as that of paramagnetic Fe.[2] Thus no change of phase was thought to have occurred when Fe lost its magnetism at the Curie temperature. A good definition of a phase includes the fact that each phase has a set of order parameters, (η_1, η_2, η_3, …) which specify its physical properties.[3] Such order parameters include composition, structure, atomic order parameter, magnetization, etc. By including "order parameters" in the definition of a phase it becomes clear that a discontinuous change in order parameter occurs at a phase change. If the change is from a zero value to an infinitesimal one, the transformation is of higher order thermodynamically. Such is the case for most *paramagnetic* to *ferromagnetic* transformations.

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Even if the word "structure" is included explicitly in the definition, the *paramagnetic* to *ferromagnetic* transition should be classified as a phase transformation. This is because there is a change in symmetry during the *paramagnetic* to *ferromagnetic* transition. For the case of Fe, this transformation causes a change from paramagnetic cubic β-Fe to ferromagnetic tetragonal α-Fe. The changes in symmetry during magnetic transformations are the topic of this paper.

Symmetry Operations

Classical Symmetry Operations

It is well known that the following point symmetry operations are the only possible in a three dimensional crystal:

$$\begin{array}{ll} N & 1,\ 2,\ 3,\ 4,\ 6 \\ \bar{\mathbf{N}} & \bar{1}, \mathrm{m}, \bar{3}, \bar{4}, \bar{6} \end{array}$$

Of these, five are called *proper symmetry* operations (or symmetry operations of the first kind), since when operating on an object they do not change its handedness or chiralty. These are *1*, *2*, *3*, *4*, and *6*. The remainder of the operations, known as *improper symmetry* operations (or symmetry operations of the second kind), change the handedness of an object on which they act. A general point in classical crystallography is represented by its three-dimensional co-ordinates *x, y, z*.[4]

Magnetic Symmetry Operations

When a magnetic moment is present in a crystal an additional type of symmetry operation may be present, often denoted as $\Re$. This operation can be thought of as changing the sign of the moment when it operates on an object that has a magnetic moment or spin. Thus, the operation $\Re$ changes the moment on the left to the one on the right of Figure 1.

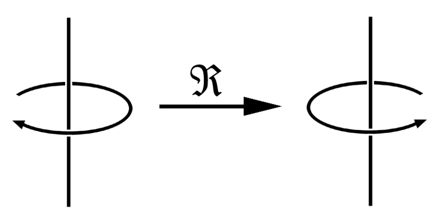


Figure 1: The magnetic moment on the left is converted to the one on the right by the operation of the symmetry element $\Re$.

The antisymmetry operation, $\Re$, can also be thought of as time inversion, as the 'current' denoted by the arrow in Figure 1, on the left would change to that on the right if time were reversed. It can be seen that the symmetry of a magnetic moment is $\infty/mm'm'$ (in Curie limiting group notation).

By using the $\mathfrak{R}$ operation on each of the classical symmetry elements, a new set of magnetic symmetry elements is formed. These are represented as:

$$\mathfrak{R} \otimes n = n' \quad \text{and} \quad \mathfrak{R} \otimes \bar{n} = \bar{n}'$$

Here, the symbol ⊗ represents the operation of $\mathfrak{R}$ on the given symmetry operation. For example, Figure 2 shows the operation of $\mathfrak{R}$ applied to a mirror plane, giving rise to the symmetry operation m’. Other examples of magnetic symmetry operations are shown in Figure 3, where the operations *2′* and *4′* are represented in stereograms.

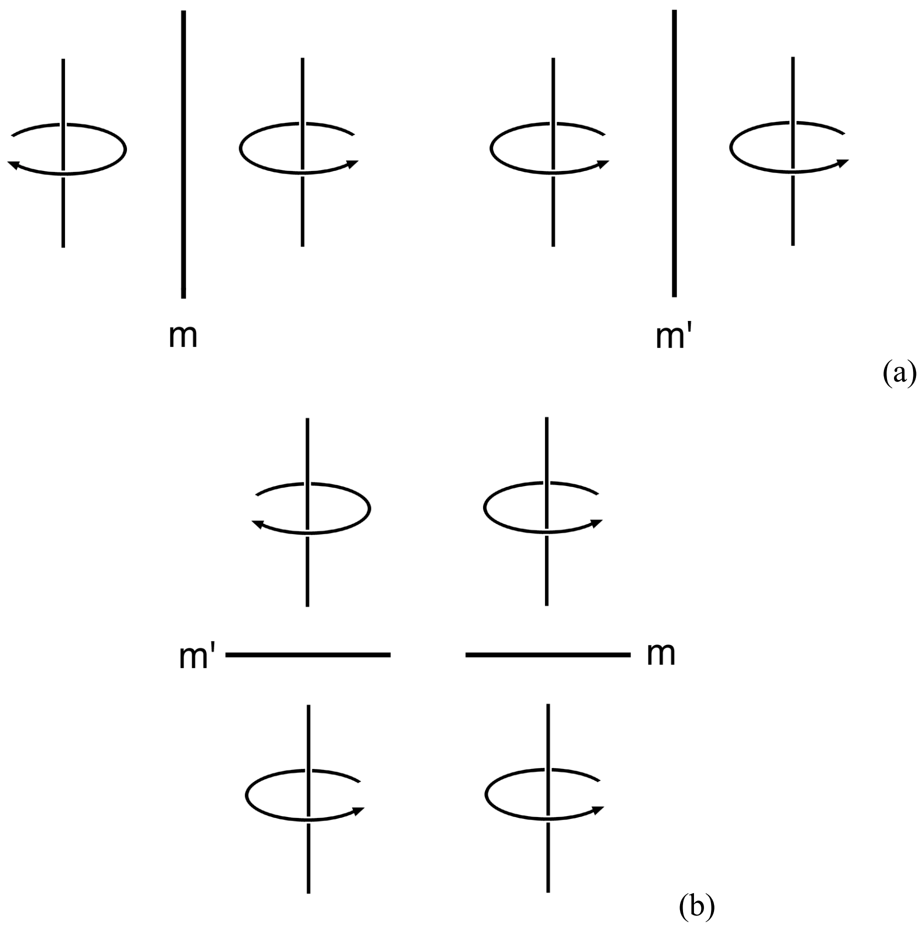


Figure 2: Axial vector representations of the parallel and perpendicular m’ magnetic symmetry operation

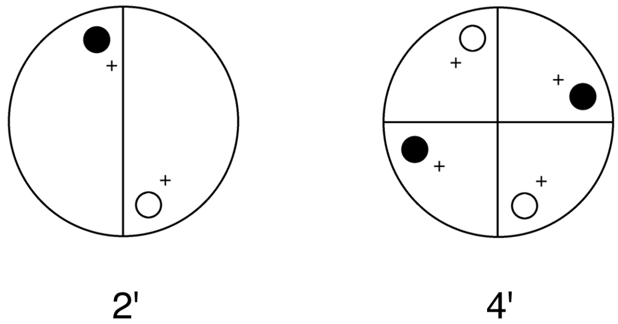


Figure 3: Stereograms of the *2'* and *4'* point groups of black/white symmetry

These new operations need not be magnetic ones. They could be thought of as ferroelectric ones (changing a plus polar vector into a minus polar vector) or changing the color of an object from white to black etc. Magnetic symmetry operations are a subgroup to the black/white symmetry operations.[5-9] In this type of crystallography a general point is represented in four dimensions, namely, x, y, z, s, where s takes a value of +1 or -1. The last digit could represent white & black, or two spins or plus and minus charge. The symmetry operation $\mathfrak{R}$ is usually called the anti-identity operation since it changes one type of site (with $s = 1$) into the other type (with $s = -1$).

Figure 4 illustrates the relationships of black/white symmetry. The various "commas" are related to each other by the symmetry operations *1*, *m*, *1'* and *m'* as depicted in the figure. Here *1* is the identity, *m* is a mirror plane, *1'* is the anti-identity element we called $\mathfrak{R}$ above and *m'* is an anti-mirror plane.

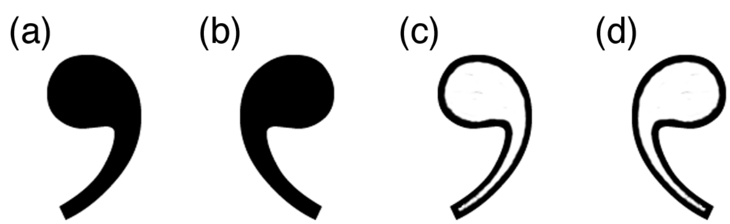


Figure 4: Examples of the antisymmetry operator and mirror planes

This can be seen form the following relationships: (a) operated on by *1* yields (a); (a) operated on by *m* yields (b); (a) operated on by *1'* yields (c); (a) operated on by *m'* yields (d).

Point Groups

Classical Crystallographic Point Groups

The above mentioned *classical symmetry operations* may be combined in a number of ways about a point in space. These combinations give rise to the well known 32 crystallographic point groups, which we term here *the classical crystallographic point* *groups.* These point groups do not have $\Re$ as a symmetry operation and hence can not be used to fully describe the symmetry of crystals that have magnetic moments associated with the atoms. Their representations on stereograms are shown in most books on crystallography.

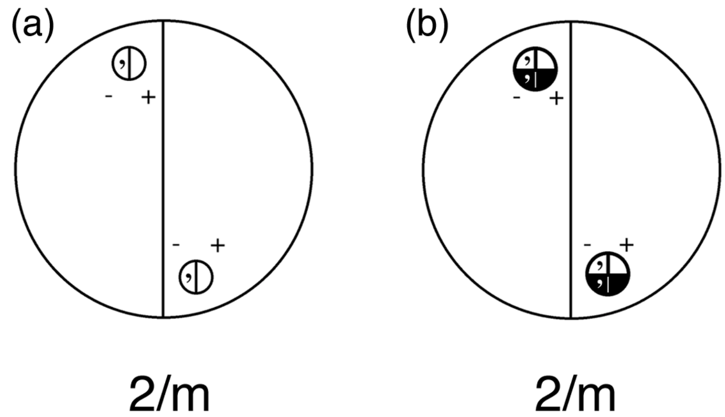
The Gray Point Groups

The magnetic point groups are obtained in a similar fashion as the classical crystallographic point groups, namely by the grouping of all the symmetry elements into possible combinations. Before we look at the magnetic groups we must discuss the so-called "gray point groups." These are point groups whose set of general co-ordinates is $x, y, z, \pm s$. The term "gray" comes about from the black and white symmetry analog: if every equivalent site has <u>both</u> a white colored object and a black colored object the overall color will appear to be gray. In the magnetic case these groups represent the paramagnetic point symmetry, where each atom has a moment which is either *up* or *down* and therefore has two possible states. The moments on the atoms are independent of one another in sign and direction. The sum of the moments in a paramagnetic crystal is zero. It is clear that each of the classical point groups can have a gray point group associated with it. There are therefore 32 gray point groups. The order of each of the gray point groups is twice that of its corresponding classical point group.

Black/White Point Groups

We will start with a classical point group, namely the one designated *2/m*. A representation of this is shown in Figure 5(a). It can be seen that it is of order four, as it has four symmetry operations in its group. The co-ordinates of each of the generated points are: $xyz \quad xy\bar{z} \quad \bar{x}\bar{y}z \quad \bar{x}\bar{y}\bar{z}$. To obtain the four symmetry operations in this group we ask what operation can take the point at $x\ y\ z$ to the four equivalent sites. This is shown in Figure 5.

Figure 5(b) shows the gray point group *2/m*. This point group is of order 8 since each site has a value of s equal to ±1 represented as white and black, respectively. In addition to the four symmetry operations listed for the *2/m* classical point group, the gray *2/m* group has $1'$, $2'$, m' and $\bar{1}'$. In Figure 6, three possible subgroups of the gray group *2/m* are shown.



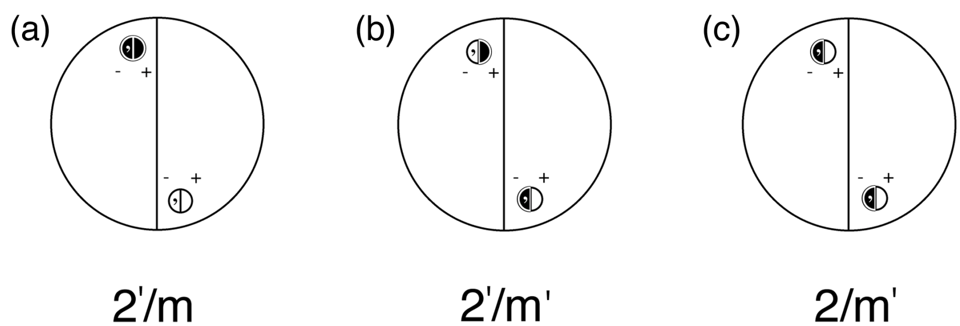
(a)

$x, y, z, +1$ E
$\bar{x}, \bar{y}, z, +1$ 2
$x, y, \bar{z}, +1$ m
$\bar{x}, \bar{y}, \bar{z}, +1$ $\bar{1}$

(b)

$x, y, z, +1$ E
$x, y, z, -1$ E'
$\bar{x}, \bar{y}, z, +1$ 2
$\bar{x}, \bar{y}, z, -1$ 2'
$x, y, \bar{z}, +1$ m
$x, y, \bar{z}, -1$ m'
$\bar{x}, \bar{y}, \bar{z}, +1$ $\bar{1}$
$\bar{x}, \bar{y}, \bar{z}, -1$ $\bar{1}$'

Figure 5: White (Classical) point group and Gray point group representations for *2/m*



(a)

$x, y, z, +1$ E
$\bar{x}, \bar{y}, z, -1$ 2'
$x, y, \bar{z}, +1$ m
$\bar{x}, \bar{y}, \bar{z}, -1$ $\bar{1}$'

(b)

$x, y, z, +1$ E
$\bar{x}, \bar{y}, z, -1$ 2'
$x, y, \bar{z}, -1$ m'
$\bar{x}, \bar{y}, \bar{z}, +1$ $\bar{1}$

(c)

$x, y, z, +1$ E
$\bar{x}, \bar{y}, z, +1$ 2
$x, y, \bar{z}, -1$ m'
$\bar{x}, \bar{y}, \bar{z}, -1$ $\bar{1}$'

Figure 6: Subgroups of the gray point group *2/m*. Only *2'/m'* can be ferromagnetic, and this only if the axis of magnetization is perpendicular to the 2-fold axis. The other two could be antiferromagnetic.

When all possible combinations are made there turns out to be 58 white/black point groups. These added to the 32 gray point groups and the 32 classical point groups make a total of 122 point groups.[4] Of these, only those point groups that are subgroups of the Curie limiting group $\infty/mm'm'$ can support ferromagnetism. There are 31 such point groups out of the 122.[4,7]

Bravais Lattices

Classical Bravais Lattices and Black/White Bravais Lattices

In classical crystallography there are 14 distinct Bravais lattices. Let us consider the primitive cubic Bravais lattice shown in Figure 7(a). If an opposite "colored" site is placed in the center of the cell we obtain one of the black and white symmetry Bravais lattices.

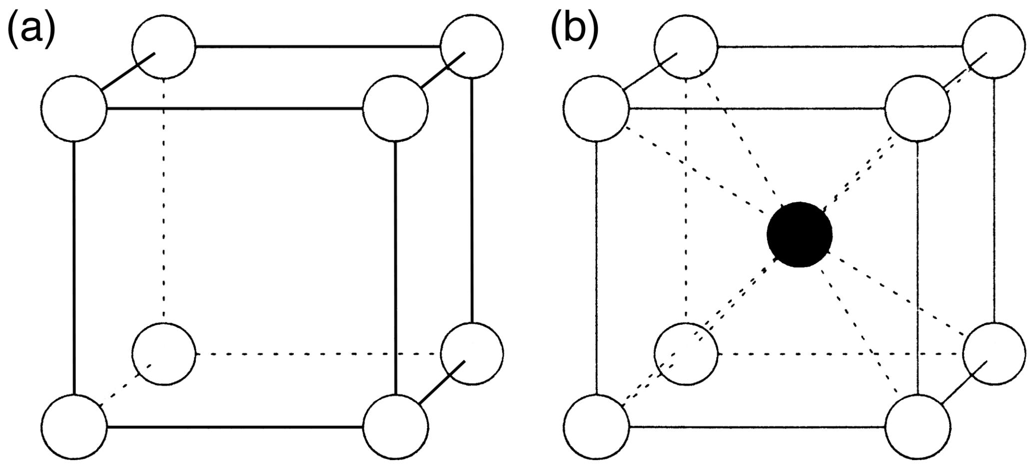


Figure 7: Primitive cubic "classical" Bravais Lattice (cP) and primitive cubic black/white Bravais Lattice. (cP')

This lattice is seen to be primitive since the two points are not equivalent, but opposite in colors (or ±1). In order to be a black/white Bravais lattice the number of white sites (+1) must equal the number of black sites (-1). Hence, these black and white symmetry Bravais lattices are used in antiferromagnetic materials. The Bravais lattice shown in Figure 7(b) is designated as cP' to distinguish it from the primitive cubic Bravais lattice cP. There are 22 additional black/white Bravais lattices.

Space Groups

Classical Space Groups

If the above mentioned 32 classical point groups are combined with those Bravais lattices that are consistent with their symmetry a total of 73 symmorphic space groups are obtained.[4,7] Each of these space groups can be derived from the translational symmetry of one of the Bravais lattices and the symmetry operations delineated above. Other space groups can be found when new symmetry elements, sometimes called microsymmetry elements are also included. These include screw axes (denoted as n_i, where n is the order of the rotation axis and i is related to the pitch of the screw) and glide planes, which are combinations of mirror symmetry and translations. In both of these types of microsymmetry operations, the translations are fractions of the unit cell translations. The addition of these symmetry operations increases the number of possible space groups to 230. The symmetries of these 230 groups are detailed in the *International Tables for Crystallography*.

Black/White Space Groups

The addition of the anti-identity element to the point operations and the addition of the 22 black/white Bravais lattices leads to an increase in the number of space groups to 1651.[4,7] As with the point groups, all of these can not support ferromagnetism. Only those space groups whose point symmetry is a subgroup of $\infty/mm'm'$ can support ferromagnetism.

Types of Magnetism

Different forms of magnetic behavior involve different symmetry considerations. All materials exhibit some magnetic behavior. Classification of the different types of magnetism is accomplished by considering the response of the material to a magnetic field, and the magnitude and orientation of magnetic moments (if applicable) in the unit cell. By consideration of the symmetry of these systems, the anisotropic magnetic properties of the single crystals can be described.

A simple approach to finding the symmetry for the ferromagnetic, antiferromagnetic, and ferrimagnetic structures is possible by considering the intersection of the atomic crystal structure with the symmetry of the magnetic moments taking into account their orientations. In some magnetic structures, such as helimagnetism, the stacking of certain magnetic planes is also an important parameter.

Magnetic behavior of most materials can be classified by the following groups: diamagnetism, paramagnetism, ferromagnetism, antiferromagnetism, or ferrimagnetism. Additional types of less common magnetic behavior are characterized by consideration of the relations of the new behavior to these five groups.[10]

Differences in magnetic behavior arise from differences in magnetocrystalline anisotropy and exchange interactions for different materials. Magnetocrystalline anisotropy is linked to the symmetry of the crystal and involves the electron spin-orbital interactions and crystal fields.[10] Magnetocrystalline anisotropy generates the directions for the magnetic moments that have minimum energy. The exchange interactions are quantum mechanical phenomena required by the Pauli exclusion principle. For strong exchange, the magnetic moments have a strong coupling to one another. This is called cooperative phenomena, while small exchange interactions yield non-cooperative phenomena. These ideas are more thoroughly described in references 10 and 11.

Diamagnetism

Diamagnetic materials have a slightly negative susceptibility (or permeability slightly less than one). The atoms that make up the diamagnetic materials have no permanent magnetic moment and therefore the materials maintain the same symmetry as the atomic crystal structure unless a field is applied. When a field is applied, an induced magnetic moment in the direction that opposes the magnetic field is produced. This yields a magnetic symmetry that is the intersection of the crystal structure with those of the applied field and the induced moment. This generally reduces the symmetry. Without an applied field for these materials, the symmetry of the structures may always be considered in the classical space group notation.

Paramagnetism

Paramagnetic materials have a slightly positive susceptibility (or permeability of nearly unity). Local magnetic moments are present on the atoms, but are not aligned due to thermal fluctuations. An applied magnetic field will align the moments in the direction of the magnetic field (for a large enough field). The symmetry of the saturated paramagnet becomes the intersection of the field and moments along the direction of the applied field. However, saturation of paramagnets requires enormous fields and therefore, the magnetic moments of a paramagnet are only statistically oriented with the field. Qualitatively, the paramagnetic material will have symmetry less than that of the atomic crystal structure due to the statistical alignment of the moments with the field. The general case for a paramagnet without an applied field is that of gray symmetry, since the magnetic moments at each site are considered both parallel and antiparallel (can be either of two spins).

Ferromagnetism

In this article, we consider the ferromagnetic materials to involve only localized moments. Ferromagnetism occurs due to the exchange interactions between magnetic moments in the materials in direct or indirect manners. The direct exchange involves the overlap of atomic orbitals and the interactions of the electron spins with one another to form a positive interaction. Superexchange, a form of indirect exchange, involves interactions mediated by intermediate atoms, such as anions in ferrimagnetic spinels. Spontaneous magnetic moments in these alloys yield lower or equal point group symmetry from the point of view of classical point group symmetry. If we consider the paramagnetic phase from the point of view of gray point group symmetry, on ferromagnetic ordering it loses at least one half of its symmetry elements since all the moments of the ferromagnet have the same sense. Cutting in half the order of the group allows these transitions to be of second order (higher order thermodynamic transitions) by the well-known Landau rules.[12,13]

Antiferromagnetism

Antiferromagnetic structures can be divided into two sublattices. Both lattices have equal numbers of formula units. One sublattice has moments which are opposed to those in the other sublattice so that the net magnetization is zero. These sublattices reduce the symmetry of the structure from that of the classical structure in much the same way as in ferromagnets. They also may be characterized by the black/white Bravais lattices.

Ferrimagnetism

Ferrimagnetic materials have sublattices much the same as in the antiferromagnetic state. However, ferrimagnets have a net magnetization due to the uncompensated atomic moments in the unit cell. Examples of these magnetic materials include garnets and ferrites.

Application

An understanding of the symmetry difference between the *paramagnetic* and *ferromagnetic* phases can be helpful in determining the possible domain orientations for a given material. In the application of magnetic symmetry to structures, the assumption of localized moments is made. The following figures indicate the direction of the magnetic moments in the upper right. The black/white/gray color of the atoms in some of the structures is not an indication of the symmetry color of the site, but has been used to indicate different atoms. Values for the lattice parameters, magnetic moments, Curie temperatures, and easy axes are well known in some cases, but three references are given for the less well known data.[14-17]

Elements

α-Iron

The intersection of the magnetic moments along the [001] with the atoms on the bcc lattice reduces the point symmetry from $m\bar{3}m$ to *4/mm'm'*. This is a simple case where the symmetry is often thought to be cubic, but is really tetragonal due to the axial symmetry magnetic moment. Cubic symmetry does not support ferromagnetism! This is because the moments always produce a special direction.

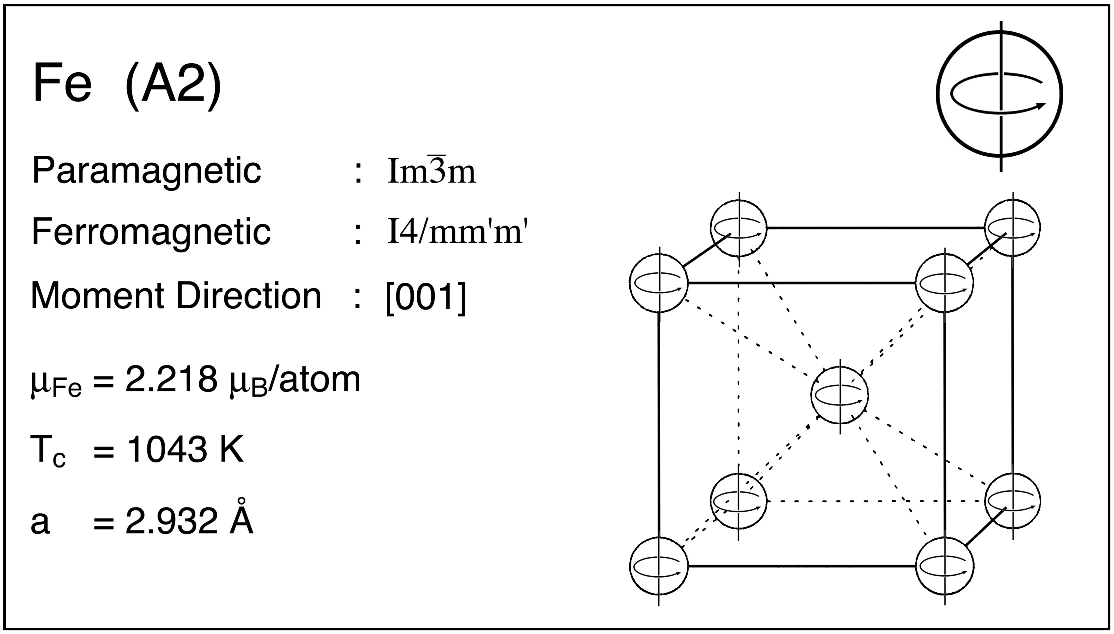


Figure 8: α-Fe crystal structure and magnetic information.

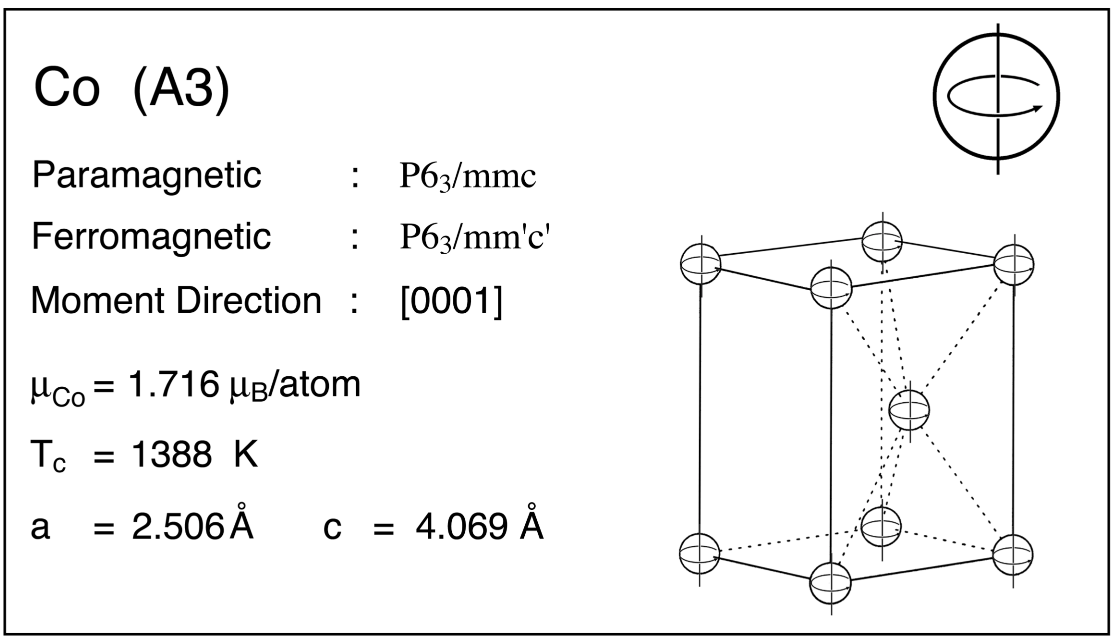


Figure 9: ε-Co structure and magnetic information

ε-Cobalt

In hcp cobalt, the magnetic moments are parallel to the [0001] direction at high temperatures. The phase transformation from paramagnetic to ferromagnetic states reduces the symmetry by a factor of two, represented by the intersection $6_3/mmc \cap \infty/mm' = 6_3/mm'c'$. The paramagnetic point group is a gray point group so its order is

48. The order of the point group of the ferromagnetic phase is 24. The introduction of the antisymmetry mirror plane yields the change from *c* glide to *c'* glide among other changes.

Nickel

A reduction in the symmetry of the Ni crystal structure from fcc occurs due to the alignment of the magnetic moment along the [111]. This produces a special direction from the intersection $m\bar{3}m \cap \infty/m$ along the [111]. The symmetry is reduced to $\bar{3}m'$.

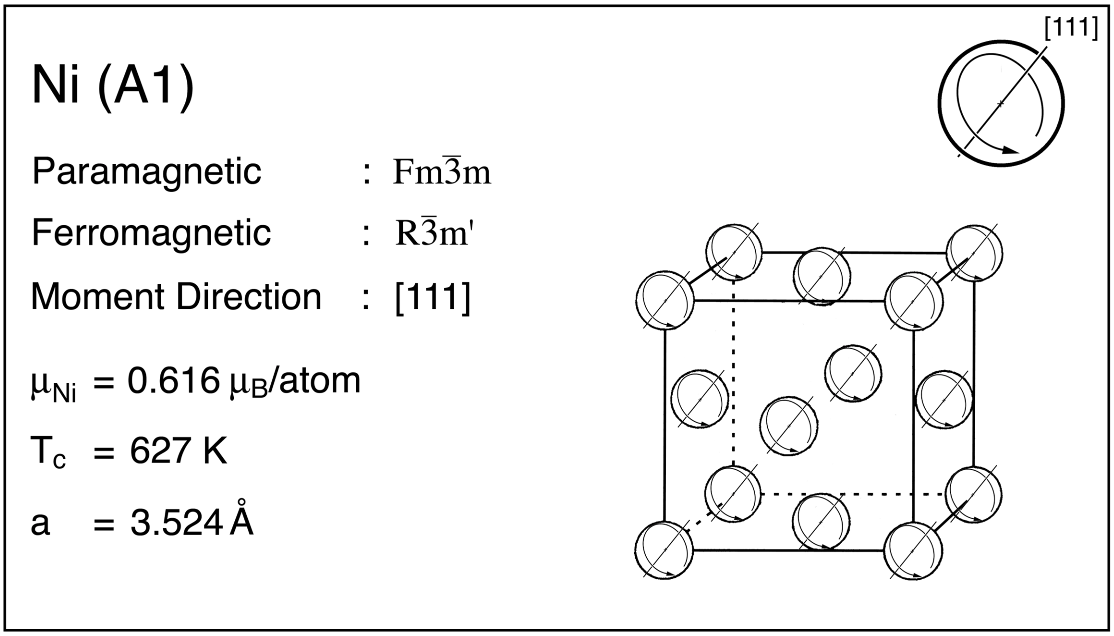


Figure 10: Ni crystal structure and magnetic information

Ordered Intermediate Phases

α'-FeCo

The α'-FeCo phase has the CsCl structure (B2) if the magnetic symmetry is ignored. Reduction of the symmetry is found when the magnetic moments are considered to be along the [001]. The magnetic space group of α'-FeCo is *P4/mm'm'*.

The change of symmetry during the transformation from *paramagnetic* to *ferromagnetic* phases has interesting implications for the domain structure. The case of α'-FeCo is not typical for this consideration since the first order transformation of ferromagnetic α-(Fe,Co) to paramagnetic fcc (Fe,Co) phase transformation occurs before a Curie temperature is reached. However, the hypothetical case of a paramagnetic B2 alloy undergoing the ferromagnetic ordering transformation yields 6 magnetic domains. This occurs due to the reduction of the gray symmetry point group ($m\bar{3}m$) with order 96 to the black/white symmetry point group (*4/mm'm'*) of order 16. The moments in these domains will lie along the <100>, which remain equivalent energetically, as the magnetocrystalline anisotropy retains the cubic symmetry.

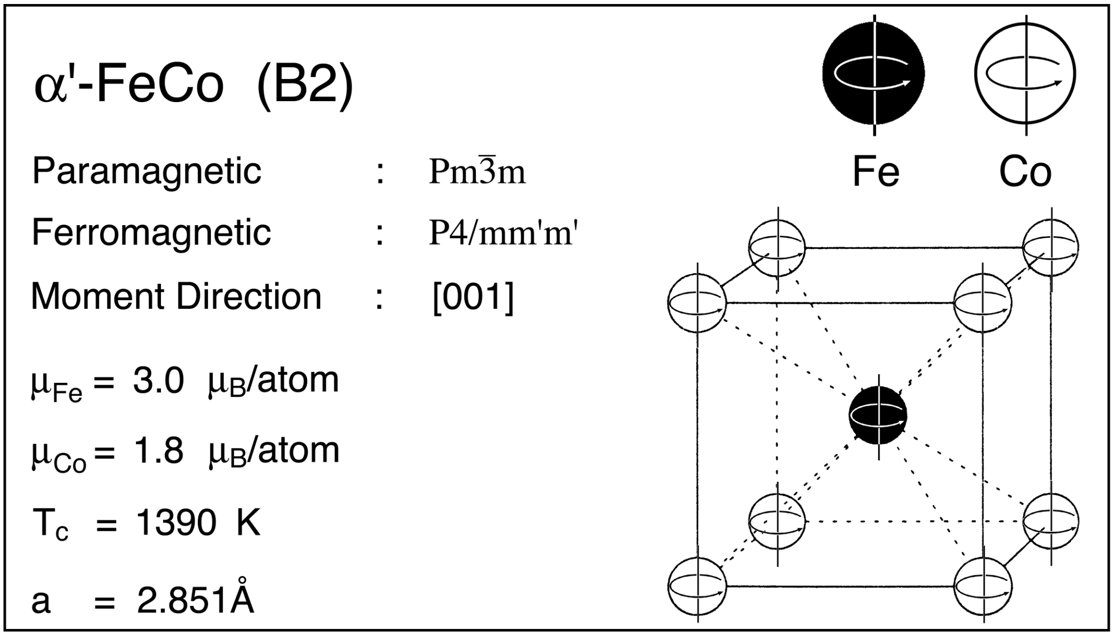


Figure 11: α'-FeCo crystal structure (B2) and magnetic information

CoPt

CoPt has the AuCu ($L1_0$) chemical structure. This structure has a paramagnetic (gray) symmetry of *P4/mmm*. The addition of the magnetic moments yields the lower symmetry of *P4/mm'm'*. Notice that the ferromagnetic space group is the same for CoPt as for α'-FeCo. However, the number of domains developed during the phase transformation from the paramagnetic phase to the ferromagnetic phase is only two in this case. That is because the order of the paramagnetic CoPt phase is only 32 whereas it is 96 in the α'-FeCo case. This has interesting implications for the domain structures for these materials.

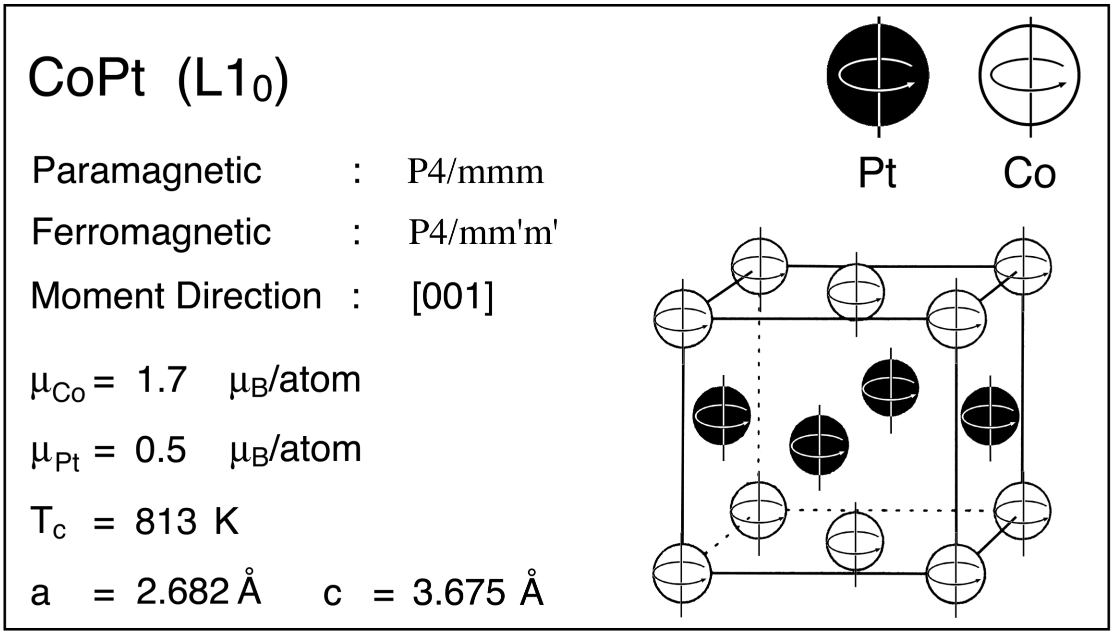


Figure 12: CoPt crystal structure ($L1_0$) and magnetic information

$CrPt_3$

$CrPt_3$ has the Cu_3Au ($L1_2$) chemical structure. The Pt atoms have polarized moments, but these moments are antiferromagnetically coupled to the Cr atoms, as opposed to the previous case of CoPt where the polarized moments are ferromagnetically coupled. The antiferromagnetic coupling creates a ferrimagnetic arrangement with one Co moment opposing the three Pt moments. Since the magnetic moments oppose each other, and the moment has a mirror plane perpendicular to its axis, the symmetry of the $CrPt_3$ structure is the same as CoPt. The number of variants for domains in this case is six (as in the α'-FeCo case).

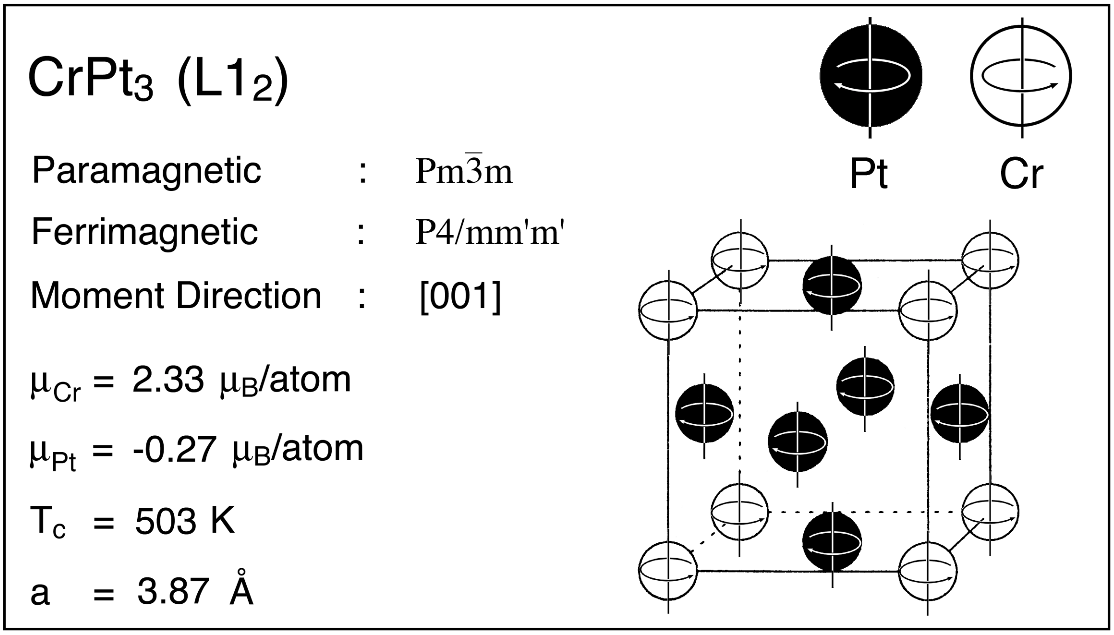


Figure 13: $CrPt_3$ crystal structure ($L1_2$) and magnetic information

Cu_2MnAl (Heusler Alloy)

The Cu_2MnAl or Heusler alloy has the $L2_1$ chemical structure. The magnetic moments in this case are along the [111] direction, making it unique with respect to the other 3-fold axes of the paramagnetic structure. This case is similar to that of Ni.

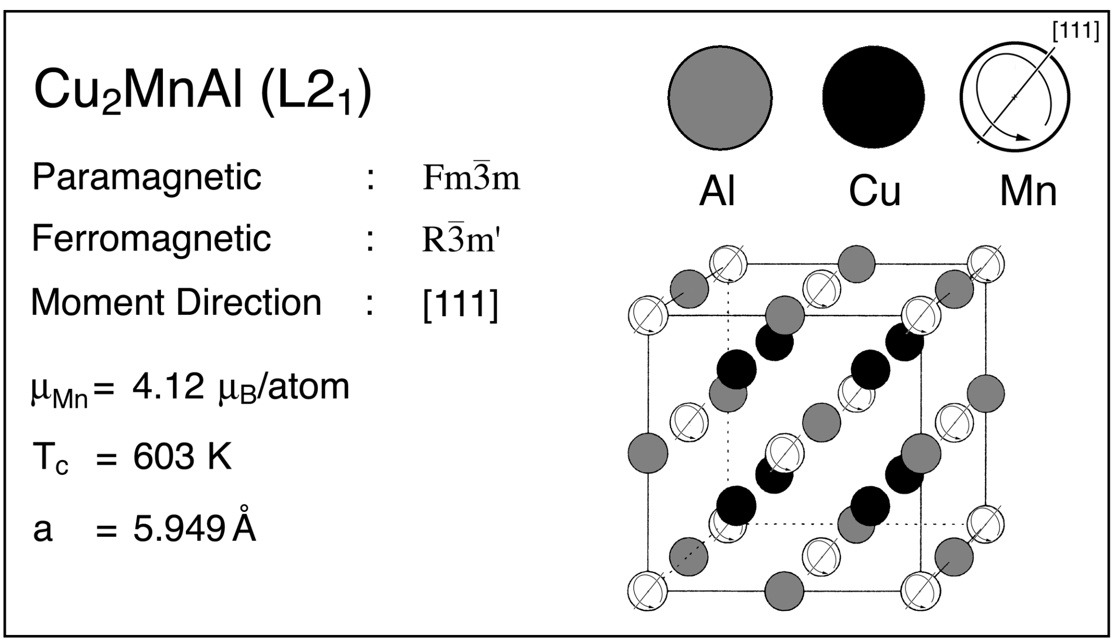


Figure 14: Cu_2MnAl (Heusler) crystal structure ($L2_1$) and magnetic information

Concluding Remarks

We have presented an overview of some of symmetry and structural aspects of magnetic ordering. By including the symmetry of the magnetic moments (∞/mm'm') the magnetic space groups of several magnetic phases have been presented. In some cases, magnetic space groups are detectable by diffraction experiments. Shen and Laughlin have shown by convergent beam diffraction that the projected point group symmetry along the [0001] of $PrCo_5$ is reduced from *6mm* to *6* on magnetic ordering.[18] This implies that the space group changed from *P6/mmm* in the paramagnetic state to *P6/mm'm'* in the ferromagnetic state.

Other implications of magnetic ordering are also evident. The thermodynamic order of the paramagnetic to ferromagnetic transition for uniaxial crystals should be second order (higher order), since from the point of view of the gray point groups the symmetry is reduced by a factor of two on magnetic ordering. Also, the number of magnetic domains present after ordering can be predicted in a straightforward manner by the use of group theory. For example in the cubic to tetragonal transition of alpha iron, the order of the point group changes from 96 (paramagnetic, cubic) to 16 (ferromagnetic, tetragonal). The ratio of the orders of the groups is 6, showing that six domains will exist for this transition. Other phenomena in magnetic materials, such as domain interactions, magnetocrystalline anisotropy, magnetostriction, etc. can be analyzed better by using the full symmetry aspects of the magnetic state.

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Acknowledgments

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