

# 1

## Introduction to Magnetic Materials

### 1.1 Theory and Fundamentals of Magnetization

Almost everyone knows about magnetic materials and the magnetic force, but very few are familiar with the underlying mechanisms that cause the magnetic force. The existence of an inextricable correlation between electricity and magnetism is the key to understanding this phenomenon. In the nineteenth century, research on magnetism and electricity resulted in the discovery of electromagnetism theory by Faraday and Maxwell. They proposed that the magnetic field can be induced in a material in the presence of electric current, which attributes to the understanding of the origin of magnetism. Indeed, for any material volume, matter consists of electrons with their motion around atoms and their interaction, which indicates how magnetism is induced from matter. This simply means that the magnetism originates from the contribution of electron motion around their own axis, which is considered as spin magnetic moment ( $\mu_s$ ), and the motion of the electron around the nucleus of the atom, which is considered as orbital magnetic moment ( $\mu_{\text{orb}}$ ). The combination of these moments results in magnetic moment generation, which affects the type of magnetism for each element or material. According to Halliday et al. [1], the spin magnetic moment is an intrinsic property of the electron and is attributed to the spin angular moment ( $S$ ) as follows:

$$\mu_s = \frac{e}{m} S \quad (1.1)$$

where  $e$  is the electron charge and  $m$  is the electron mass.  $S$  is quantized and can only be  $\pm 1/2$ . Since only the  $z$  component of  $S$  is measurable, the  $z$  component of  $\mu_s$  is given as [2]:

$$\mu_{s,z} = \pm \frac{e\hbar}{4\pi m} \quad (1.2)$$

where  $\hbar$  is Planck's constant. The positive value of this equation is equal to  $9.27 \times 10^{-24}$  J/T. This is considered as Bohr magneton ( $\mu_B$ ), which is the most basic unit of the magnetic moment in magnetism, and magnetic materials are elucidated based on this quantity.

The magnetic moment of solids normally arises from partially filled inner electron shells of transition metal atoms. In Hund's rules, the theory is closely related to the magnetism of free atoms or ions, which expect the orbital and spin moment as the role of the number of inner-shell electrons [3]. Indeed, some factors give rise to the origin of magnetism in ferrites, which are: (i) superexchange adjacent metal ions, (ii) unpaired electrons in 3d or 4f shells, and (iii) nonequivalence in the number of magnetic moments in tetrahedral and octahedral sites [4]. In oxide compounds such as ferrites, the orbital magnetic moment is typically quenched by the electronic fields induced by the surrounding oxygens around the metal ions [4]. The atomic magnetic moment then becomes equal to the electron spin moment as follows:

$$m = n\mu_B \quad (1.3)$$

where  $n$  is the number of unpaired electrons and  $\mu_B$  is a Bohr magneton unit. For more details, the readers are referred to Ref. [5].

## 1.2 Type of Magnetism

In general, the magnetic materials in the micro or macroscopic scale are classified into five distinct groups based on their response to an externally applied magnetic field. They are diamagnetic, paramagnetic, ferromagnetic, anti-ferromagnetic, and ferromagnetic, which are briefly described below according to Refs. [5–8]. Additionally, Table 1.1 introduces the quantities and units that are taken into account in magnetism [4].

**Table 1.1** Presented quantities and units considered in magnetism [5].

| Quantity                                  | Symbol      | Gaussian unit      | Conversion Factor           | SI unit             |
|---|-------------|--------------------|-----------------------------|---------------------|
| Mass magnetization                        | $\sigma, M$ | emu/g              | 1                           | Am <sup>2</sup> /kg |
| Magnetic flux                             | $\Phi$      | Maxwell (Mx)       | 10 <sup>-8</sup>            | Weber (Wb)          |
| Volume magnetization                      | $4\pi m$    | G                  | 10 <sup>3</sup> /4 $\pi$    | A/m                 |
| Magnetic dipole moment                    | $j$         | emu                | 4 $\pi$ × 10 <sup>-10</sup> | Wbm                 |
| Volume susceptibility                     | $\chi$      | Dimensionless      | 4 $\pi$                     | Dimensionless       |
| Magnetic field strength                   | $H$         | Oersted (Oe)       | 10 <sup>3</sup> /4 $\pi$    | Ampere/m (A/m)      |
| Mass susceptibility                       | $\chi_p$    | cm <sup>3</sup> /g | 4 $\pi$ × 10 <sup>-3</sup>  | m <sup>3</sup> /kg  |
| Magnetic flux density, magnetic induction | $B$         | Gauss (G)          | 10 <sup>-4</sup>            | Tesla (T)           |
| Magnetic moment                           | $m$         | Mu                 | 10 <sup>-3</sup>            | Am <sup>2</sup>     |

Source: Hajalilou et al. [5]/with permission of Springer Nature.

### 1.2.1 Diamagnetism

In this state, the atoms do not exhibit a magnetic moment in the absence or presence of an externally applied field. But, when the external field is applied, it causes an induced magnetic field in the opposite direction, which results in repulsive force. This simply means that the diamagnetic materials are repelled by a magnetic field. Indeed, the orbital velocity of the electrons, around their nuclei, would change in the presence of an applied magnetic field, which results in magnetic dipole moment variation in the opposite direction of the applied field [9]. In terms of electronic configuration, such characteristics occur in those materials with filled electronic sub-shells, in which, the magnetic moments are paired and overall cancel off each other [5].

### 1.2.2 Paramagnetism

In this state of magnetism, the materials represent a permanent magnetic dipole moment with an unpaired electron shell typically in the 3d or 4f shells [5, 9, 10]. Several theories have been suggested to explain such magnetism, which is only valid for a specific type of material. For instance, the Langevin model, which is considered for describing the non-interacting concentrated electrons' behavior, declares that the magnetic moment of each atom orientates in a random state because of thermal agitation [5]. The small alignment of these magnetic moments is caused by applying an external magnetic field, which accordingly gives rise to the creation of low magnetization in the applied field direction. Owing to the contribution of both the electron spin and the orbital angular moment to the magnetization, a positive susceptibility between  $10^{-2}$  and  $10^{-4}$  is obtained at room temperature. The magnetization ( $M$ ) is proportional to the applied field ( $H$ ) at relatively lower applied fields, but it deviates from proportionality at a higher applied field where saturation magnetization ( $M_s$ ) initiates to occur. A slight alignment is taken into account by the inverse correlation of susceptibility ( $\chi$ ) with temperature ( $T$ ), which is known as Curie law and is expressed as:

$$\chi = C/T \quad (1.4)$$

Based on this relation, the temperature would enhance at a relatively high applied magnetic field, which increases thermal agitation. Therefore, the alignment of the magnetic moment becomes very difficult. The Curie law also pronounces the positive susceptibility and is normally given through [11]:

$$\chi = \frac{C\mu_0 NM^2}{KT} \quad (1.5)$$

where  $N$  is the number of magnetic dipoles ( $M$ ) per unit volume,  $C$  is a constant,  $K$  is the Boltzmann constant,  $T$  is the absolute temperature, and  $\mu_0$  is the permeability of the vacuum. Except for very low temperatures, typically less than 5 K, some paramagnetic materials track this equation at most temperatures. However, most paramagnetic materials follow the Curie-Weiss Law as:

$$\chi = \frac{C}{T - \theta_c} \quad (1.6)$$

where  $\theta_c$  is a critical temperature and  $C$  is a Curie constant.

### 1.2.3 Ferromagnetism

In this state, the materials represent a powerful interaction with magnets like iron. Parallel alignment of the net moments of the same magnitude in these materials results in a remarkable net magnetization even in the absence of an externally applied field. Consequently, these materials have two key characteristics: (i) the presence of magnetic ordering temperature and (ii) spontaneous magnetization, which is the net magnetization existing inside of an even magnetized microscopic volume.

The susceptibility of a ferromagnetic material is typically large and positive in value. Moreover, it is a microstructure-dependent factor.

The atomic moments in these materials are produced by the electronic exchange force and give rise to the parallel alignment of atomic moments. A massive exchange force is equivalent to a field of about 1000 T.

Furthermore, the exchange force is a quantum mechanical phenomenon due to the orientation of the relative spins of two electrons. The electrons of neighboring atoms interact with one another in a process compared to the weaker diamagnetism and paramagnetism. This process is called exchange coupling [12]. If the material has a strong flux density and crystalline structure, a direct coupling between moments would occur (e.g. Fe, Ni, and Co) [9]. In the absence of an applied magnetic field, the aligned moment would yield spontaneous magnetization in the ferromagnetic material. Each material, which keeps permanent magnetization in the absence of an applied field, is considered a hard magnet.

### 1.2.4 Antiferromagnetism

This characteristic of magnetic materials can be observed once the two sub-lattices (namely tetrahedral and octahedral) are equally located in an opposite direction, which leads to zero net moments. Antiferromagnetic materials such as transition metal oxides present similar traits as ferromagnetic materials. A discrepancy is that the exchange interaction between the adjacent atoms gives rise to an anti-parallel alignment of the atomic magnetic moments. Hence, the magnetic moments cancel out each other, and the material would behave like low magnetization materials, i.e. paramagnetic. Susceptibility characteristics above the Néel temperature ( $T_N$ ) are the clue to elucidate the antiferromagnetism property. Indeed, above  $T_N$ , a sufficient thermal energy results in equally but oppositely aligned atomic magnetic moments to cancel out each other. Thus, their long-range order vanishes due to the random fluctuation alignment, which is the behavior of a paramagnetic material [10].

### 1.2.5 Ferrimagnetism

In this case of magnetism, the distribution of atoms represents an opposite magnetic moment, as in antiferromagnetism; however, in ferrimagnetic materials, the opposing moments are unequal and a spontaneous magnetization remains [13]. This happens when the distribution consists of different materials or ions such as

$\text{Fe}^{2+}$  and  $\text{Fe}^{3+}$ . Typically, within a magnetic domain, a net magnetic moment is induced from the anti-parallel arrangement of adjacent non-equivalent sublattices. Thus, the macroscopic property of ferrimagnetism is effectively a ferromagnetism behavior.

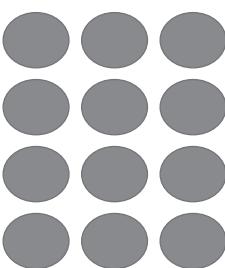
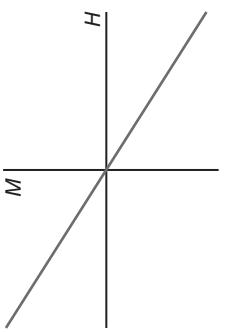
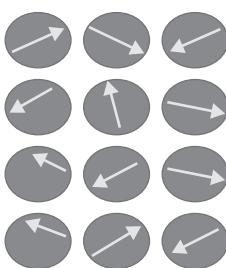
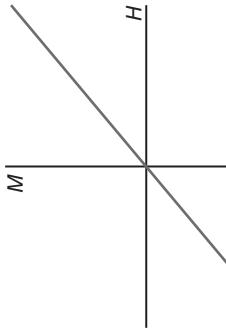
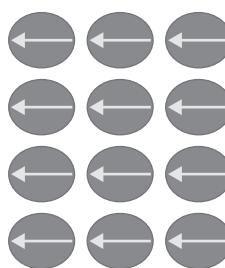
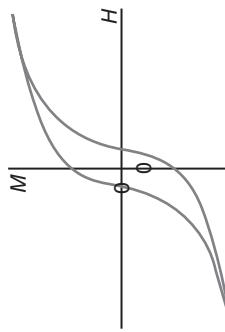
Ferrimagnetism is normally observed in ferrites such as  $\text{Fe}_3\text{O}_4$  and magnetic garnets, and ionic compounds with more complex crystal structures. The oldest known magnetic substance, magnetite ( $\text{Fe}_3\text{O}_4$ ), is a ferrimagnet; it was originally classified as a ferromagnet before Néel discovered ferrimagnetism and antiferromagnetism in 1948 [14]. Other known ferrimagnetic materials include yttrium iron garnet (YIG), cubic ferrites composed of iron oxides and other elements such as aluminum, cobalt, nickel, manganese, and zinc, and hexagonal ferrites such as  $\text{PbFe}_{12}\text{O}_{19}$  and  $\text{BaFe}_{12}\text{O}_{19}$ , and pyrrhotite,  $\text{Fe}_{1-x}\text{S}$  [15].

In a ferrimagnet, the magnetic moments of one type of ion on one type of lattice site are aligned antiparallel to those of an ion on another lattice site. Since the magnetic moments are not of the same magnitude, they only partially cancel each other, and the material has a net magnetic moment. Ferrimagnetism has several similarities to ferromagnetism in which the cooperative alignment between magnetic dipoles leads to a net magnetic moment even in the absence of an applied field. Ferrimagnetism is lost above curie temperature ( $T_C$ ), as it has the paramagnetic behavior above the  $T_C$ . Details and characteristics of the foregoing-mentioned magnetic states are given in Table 1.2.

### 1.3 Extrinsic and Intrinsic Characteristics of Magnetic Materials

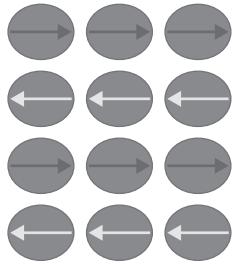
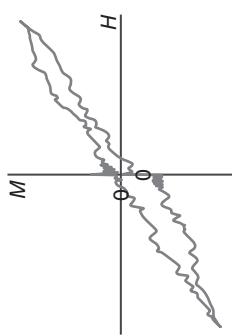
Understanding the concept of the intrinsic and extrinsic properties of magnetic materials is important to handle the microstructure and thus their application. It is believed that the ferrites' magnetic behavior relies not only on the intrinsic properties but also on the extrinsic behaviors. Therefore, understanding both of them is required to evaluate the magnetic behavior of a material. The intrinsic properties of a material are typically dependent on the chemistry and crystal structure such as crystal anisotropy, magnetostriction, and magnetostatic energy. While the extrinsic parameters such as pores, inclusions, and the nature of grain boundaries are highly microstructure sensitive. Atomic-scale quantum mechanics and relativistic effects have been modified to produce high-performance magnetic materials. Thus, due to the importance of ferrites in our daily lives, many attempts have been devoted to improving their versatility for various applications, e.g. from science to technology. In addition to the magnetic properties, the electrical properties are also important for describing their characteristics. Thus, it is necessary to correlate these properties with the relevant chemical, physical, and microstructural characteristics to improve their performance. Especially, for optimizing the properties of ferrimagnetic ceramics, it is necessary to understand both the intrinsic and extrinsic properties of a magnetic material. Intrinsic properties are those that are insensitive to variations in the microstructure feature of a sample [16]. Some characteristics of magnetic

Table 1.2 Characteristics of different types of magnetism.

| Type           | Atomic/magnetic behavior  | Spin orientation  | $M$ – $H$ curves   | Example  |
|----------------|---|---|--|--|
| Diamagnetism   | <ul style="list-style-type: none"> <li>Atoms have no magnetic movement</li> <li>Susceptibility is small and negative, <math>-10^{-6}</math> to <math>10^{-5}</math></li> </ul>                |   |   | <ul style="list-style-type: none"> <li>Inert gases, e.g. Ar, N<sub>2</sub></li> <li>Nonmetallic elements e.g. B, Si, P, S;</li> <li>Many ions, e.g. Na<sup>+</sup>, Cl<sup>-</sup> and their salts;</li> <li>Diatomic molecules, e.g. H<sub>2</sub>, N<sub>2</sub>, H<sub>2</sub>O;</li> <li>Most organic compounds</li> </ul> |
| Paramagnetism  | <ul style="list-style-type: none"> <li>Atoms have randomly oriented magnetic moments</li> <li>Susceptibility is small and positive, <math>+10^{-5}</math> to <math>+10^{-3}</math></li> </ul> |   |   | <ul style="list-style-type: none"> <li>Some metals, e.g. Al;</li> <li>Some diatomic gases, e.g. O<sub>2</sub> and NO;</li> <li>Ions of transition metals and rare earth metals, and their salts;</li> <li>Rare earth oxides</li> </ul>   |
| Ferromagnetism | <ul style="list-style-type: none"> <li>Atoms have parallel aligned magnetic moments</li> <li>Susceptibility is large (below <math>T_c</math>)</li> </ul>                                      |  |  | <ul style="list-style-type: none"> <li>Transition metals, like Co, Ni, and rare earth with <math>64 \leq Z \leq 69</math>;</li> <li>Alloys of ferromagnetic elements and some alloys of Mn, e.g. MnBi, Cu<sub>2</sub>MnAl</li> </ul>   |

### Antiferromagnetism

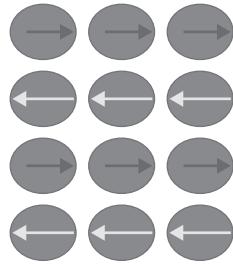
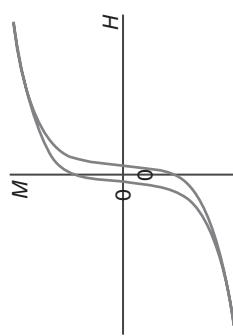
- Transition metals: Mn, Cr, Ni, Zn, and many of their compounds, e.g.  $\text{MnO}$ ,  $\text{CoO}$ ,  $\text{NiO}$ ,  $\text{Cr}_2\text{O}_3$ ,  $\text{MnS}$ ,  $\text{MnSe}$ , and  $\text{CuCl}_2$



- Atoms have anti-parallel aligned magnetic moments
- Susceptibility is small and positive,  $+10^{-5}$  to  $+10^{-3}$

### Ferrimagnetism

- $\text{Fe}_3\text{O}_4$  (magnetite);
- $\gamma\text{-Fe}_2\text{O}_3$  (maghemite);
- Mixed oxides of iron and other elements



- Atoms have mixed parallel and antiparallel aligned magnetic moments
- Susceptibility is large (below  $T_C$ )

materials such as spontaneous magnetization ( $M_o$ ), saturation magnetization ( $M_s$ ), crystal anisotropy or magnetocrystalline anisotropy ( $K$ ), ferromagnetic resonance (FMR), and Curie temperature ( $T_C$ ) are independent of the microstructure once the chemical composition of a material is fixed. As a result, it is useless to attempt to change, e.g. the curie temperature, by adjusting the microstructure. On the other hand, the extrinsic properties such as coercivity ( $H_c$ ), remanence magnetization ( $M_r$ ), permeability, hysteresis loop, and magnetic losses (hysteresis, eddy current, and residual) are highly microstructure sensitive. These parameters are influenced by the synthesis process, which affects the chemical homogeneity, crystallite or grain sizes, density, presence of nonmagnetic inclusions, and distribution of pores. From the physicochemical point of view, however, it is very difficult to fully separate the intrinsic and extrinsic properties of a material, and some of them are distinguishable based on the microstructural characteristics.

### 1.3.1 Intrinsic Properties

The intrinsic or intensive behaviors of magnetic materials, such as saturation, magnetization, anisotropy, Curie temperature, and magnetostriction, are those traits that belong to the material's characteristics and are not influenced by the microstructure, i.e. crystallite/grain, porosity size, and their distribution [9]. Intrinsic properties normally describe the atomic origin of magnetism and quantum phenomena such as exchange, crystal-field interaction, interatomic bonding, and spin-orbital coupling [17, 18]. The following sub-subsections deal with the details of intrinsic properties.

#### 1.3.1.1 Saturation Magnetization ( $M_s$ )

Saturation magnetization ( $M_s$ ) is given as the net magnetic moment per unit volume of a material. The saturation state only takes place in the situation wherein no further increase in magnetization happens by increasing the externally applied field. In this case, all domains would favor to line up in the same direction of the applied field. Increasing the externally applied field intensity enhances the domain alignment. The variation of magnetization with particle size as a result of temperature variation can be given as [19, 20]:

$$M_s = 8.1661 \exp(3T/1000) \quad (1.7)$$

where  $T$  is temperature.

The net  $M_s$  is equivalent to the vector of the magnetization of A and B sublattices in spinel ferrite, as:

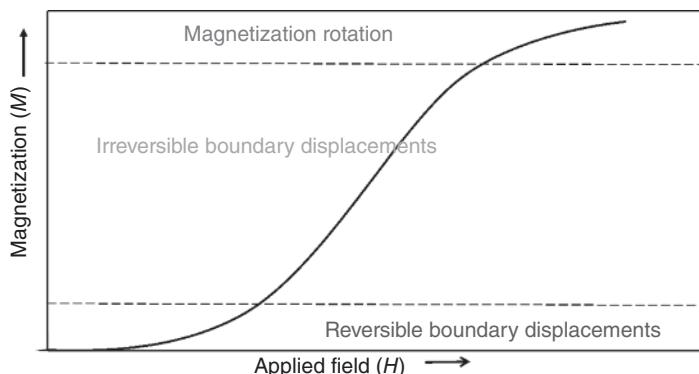
$$M_s = |M_B - M_A| \quad (1.8)$$

The exchange interaction among the electrons' ions would yield various magnetization values in A and B sites. Typically, the interaction between magnetic ions of A and B sublattices is the strongest in the state of the A-B configuration. B-B configuration is the weakest and A-A interaction is the intermediate state. The dominant A-B

configuration leads to either a partial (noncompensated) or fully ferrimagnetism in magnetic materials. Typically, the magnetic moment value in A-lattice is much smaller than that in B-lattice, which results in the above-mentioned equation [21]. As mentioned, saturation magnetization ( $M_s$ ) refers to the net magnetic moment per unit volume of a given material. This can be associated with the formula:

$$M_s = \frac{n_B \mu_B N}{M} d \quad (1.9)$$

where  $n_B$  is the number of Bohr magnetons per atom or ion (number of unpaired electron spins per atom or ion),  $\mu_B$  is the value of a Bohr magneton ( $1.1654^{-29}$  Vsm),  $N$  is the Avogadro number ( $6.025 \times 10^{23}$  atoms per mole or molecules per mole),  $M$  is the molecular weight, and  $d$  is the density. This formula is used for converting the moment in Bohr magnetons per atom or ion to units of bulk magnetization,  $M$  in emu/cm<sup>3</sup> or emu/g, where  $n \times \mu_B$  is the moment of the atom or ion in emu. As is known, Bohr magneton is the fundamental unit of the magnetic moment in Bohr theory, and it has been found that the magnetic moment is associated with the spin moment, which is almost identically equal to one Bohr magneton. The magnetic moment or magnetization is usually measured at 0 K to allow the correlation with the number of Bohr magnetons [22]. As shown in Figure 1.1, the magnetization of the sample follows the magnetization field. The slope of the curve in each region, from the origin to a point on the curve, or the ratio  $M/H$  is defined as magnetic susceptibility. The magnetization curve is divided into three major divisions. The lower section is for a reversible domain wall movement and rotation, which is considered as the initial susceptibility region. Being reversible means that after changing the magnetization slightly with an increase in the applied field, the original magnetization condition returns, once the field intensity is reduced to the original value. The second division of the magnetization curve is the one in which the irreversible domain wall motion occurs, and the slope increases greatly. The third section of the curve is the irreversible domain rotations. In this section, the slope is very flat, indicating that a large amount of energy is required to rotate the remaining domain magnetization in line with the magnetic field.



**Figure 1.1** Divisions on the  $M$ - $H$  curve based on domain magnetization.

### 1.3.1.2 Curie Temperature ( $T_C$ )

Curie temperature is a critical temperature where the magnetic behavior of a material or type of magnetization varies. Below this temperature, the ordered magnetic moment is dominant. At the Curie temperature, magnetic moments would change their directions. Above the Curie temperature, disorder magnetic moments occur, and ferromagnetic materials lose their spontaneous magnetization and hence behave similarly to paramagnetic materials. In fact, by increasing the temperature above the Curie point, thermal agitation causes a weaker alignment of the magnetic moments and leads to the net magnetization reduction. Hence, the increased thermal motion of the atoms tends to randomize the directions of any moments that may be aligned. In this circumstance, the atomic thermal motions counteract the coupling forces between the adjacent atomic dipole moment, causing some dipole misalignment without being affected by the presence of an external field. This results in saturation magnetization reduction. The saturation magnetization is maximum at 0 K, at which the thermal vibrations are in a minimum state. With the increase in temperature, the saturation magnetization diminishes gradually and then abruptly drops to zero, at what is called the Curie temperature ( $T_C$ ). At  $T_C$ , the mutual spin coupling forces are destroyed in such a way that the ferromagnetic materials behave as paramagnetic at temperatures above  $T_C$ .

### 1.3.1.3 Magnetic Anisotropy

The variation of magnetic properties concerning the magnetic moment orientation is well known as magnetic anisotropy. In other words, it refers to the dependence of the internal energy on the magnetization direction. That is, the spins of the magnetic ions are bound to a specific crystallographic direction. In crystalline materials, the magnetic moments tend to align more readily along certain crystallographic axes called easy magnetization directions. The source of this magnetic anisotropy is the interaction of magnetic ions with the electrostatic field formed typically by the first nearest neighbors of oxygen. Magnetic anisotropy is the energy of a magnetic solid related to the orientation of magnetization regarding the crystal axes. Magnetic anisotropy is typically used to discuss the dependence of the internal energy on the direction of spontaneous magnetization in ferromagnetic materials. Magnetic anisotropy affects the shape of the hysteresis loops when measured in different directions. Each permanent magnet requires a high magnetic anisotropy to retain magnetism on every magnet in the required direction. Soft magnets contained a very low anisotropy because of the easy change of their magnetization. Different types of anisotropy are as follows:

- Magnetocrystalline anisotropy,
- Shape anisotropy,
- Stress anisotropy,
- Exchange anisotropy, and
- Surface anisotropy.

These magnetic anisotropies play an important role in a variety of magnetic properties such as coercive force, hysteresis losses, magnetization processes, domain structure, shape of the hysteresis loop, and magnitudes of permeability [14].

**Magnetocrystalline Anisotropy** Magnetocrystalline anisotropy is an intrinsic property and is induced by the interaction between orbit and spin known as spin-orbit coupling. This is used to study the modification of magnetic properties along various crystal axes. Different types of crystal axes are easy, intermediate, and hard axes, which display the highest, intermediate, and lowest magnetic moment at the applied magnetic field, respectively. Indeed, there are a set of directions in all ferromagnetic substances in which the magnetization tends to be oriented in the desired direction, depending on the amount and direction of an applied field. Anisotropy of materials describes the competition between the electrostatic crystal-field interaction and spin-orbit coupling. When an applied field adjusts the spin of an electron, its' orbit shows a resistance, since the orientation of the electron's orbit is strongly fixed to the crystal lattice. The magnetocrystalline anisotropy energy is the required energy for breaking the spin-orbit coupling to magnetize a material by reorienting its spin direction.

The magnetization process is different when the field is applied along with different crystallographic directions, and the anisotropy reflects the crystal symmetry. Its origin is in the crystal-field interaction and spin-orbit coupling, or else the interatomic dipole-dipole interaction.

Magnetocrystalline anisotropy is the energy necessary to deflect the magnetic moment from the easy to an arbitrary direction. The magneto anisotropy energy ( $E_K$ ) for the uniaxial crystals is evaluated based on the direction in which the magnetization is rotated. Its value is obtained by considering some constants:

$$E_K = K_1 \sin^2 \theta + K_2 \sin^4 \theta + \dots \quad (1.10)$$

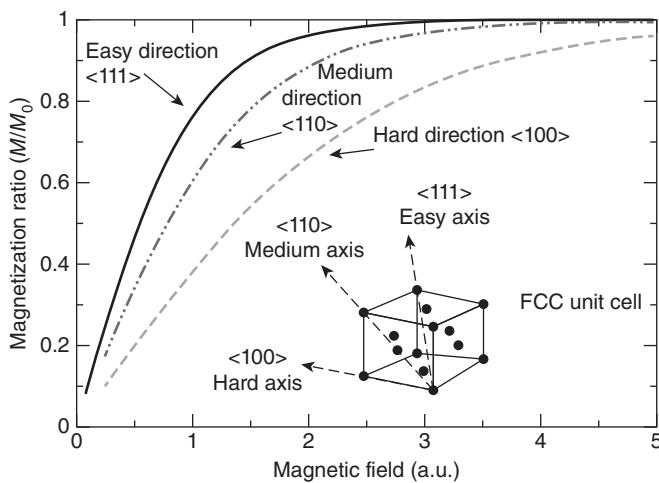
where  $\theta$  is the angle between  $M_s$  and the easy axis.

The magnetization inclines to locate along with fixed crystallographic directions, which are called easy directions or easy axes due to the influence of magnetocrystalline anisotropy. This all happens because of the spontaneous domain magnetization directions of a crystalline material in the demagnetized state. There is cubic symmetry of the easy axes in the  $\langle 100 \rangle$  direction for body-centered cubic (BCC) and  $\langle 111 \rangle$  for the face-centered cubic (FCC) lattices in the crystals. A variety of crystal directions for a cubic crystal are shown in Figure 1.2 [23]. The situation is more complete in the case of hexagonal crystals when the easy direction may lie either along the  $c$  axis of the crystal, or on the basal plane, or even in a cone about the  $c$  axis.

For spinel ferrite, magneto anisotropy energy is more formally stated as follows [9]:

$$E_K = K_1 (\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2) + K_2 (\alpha_1^2 \alpha_2^2 \alpha_3^2) + \dots \quad (1.11)$$

where  $K_1$  and  $K_2$  are the constants of anisotropy and  $\alpha_1$ ,  $\alpha_2$ , and  $\alpha_3$  are the angles between  $M_s$  and cubic axes of  $a$ ,  $b$ , and  $c$ , respectively [14]. Commonly the first



**Figure 1.2** Various possible axes of magnetization in a cubic FCC crystal. In this example, (111) is the easy axis, and (100) is the hard axis. Source: Fernando [23]/with permission of Elsevier.

constant of anisotropy ( $K_1$ ) has a key role and is dominant. Except for  $\text{CoFe}_2\text{O}_4$ , in other spinel ferrites,  $K_1$  has a negative value. Therefore, in most spinel ferrites, the easy direction is the cubic diagonal [111]. In soft magnetic ferrite,  $K_1$  should be as minimum as possible in absolute magnitude.

For polycrystalline materials of cubic symmetry, the average magnetostriction or deformation ( $\lambda_s$ ) is given by:

$$\lambda_s = \frac{2}{5}\lambda_{100} + \frac{3}{5}\lambda_{111} \quad (1.12)$$

where  $\lambda_{100}$  and  $\lambda_{111}$  are the maximum linear deformation with a field in the [100] and [111] crystallographic directions, respectively. In most ferrites, the body diagonal [111] is the easy direction except for cobalt ferrite, which exhibits an easy direction of magnetization along the cube edges at [100], and a single crystal of iron tends to magnetize in the directions of the cube edges at [100] [13]. The induced anisotropy energy is given as:

$$E = K_u \sin^2(\theta - \theta_u) \quad (1.13)$$

where  $(\theta - \theta_u)$  is the angle of the measuring field ( $\theta$ ) relative to the annealing field ( $\theta_u$ ) and  $K_u$  is the induced anisotropy constant.

**Shape Anisotropy** Shape anisotropy derives from the demagnetizing field which relies on the direction of magnetization in the sample, and it is not an intrinsic property of the material.

Shape anisotropy is the force that orders the magnetization throughout a certain particle axis. Shape anisotropy happens in extended particles. A polycrystalline sample has no macroscopic crystal anisotropy, and its grains have no specific orientation. If the sample is spherical, the direction of an applied field will not affect

its magnetization. If a magnetic sample is not spherical, it will be magnetized along the long axis than the short one. This is because the demagnetizing field along the short axis is stronger, and thus, a longer field is needed to create the same true field inside the sample.

**Stress Anisotropy** This kind of anisotropy occurs due to the change in physical dimensions when the sample is subjected to an externally applied field. The effect is associated with a fractional change in length and is called magnetostriction ( $\lambda_s$ ). Stress anisotropy is also affected by external or internal stresses caused by rapid cooling, application of external pressure, etc. It may also be induced by annealing a sample in a magnetic field, plastic deformation, or ion beam irradiation [24].

Magnetostriction ( $\lambda$ ) results from the fractional variation in length (or dimension) of a material, which is called a strain, in relevance with the constants of anisotropy (variation of magnetization). The reduction in length occurs in all ferrites during magnetization, except for ferrous ferrites. This variation can be related to the energy sensitivity to mechanical stress, which happens during the synthesis and operation process. The control of such stresses is somehow difficult. To achieve the desired quality of soft ferrites, magnetostriction should be as low as possible. In magnetostrictive materials, however, magnetostriction and magnetocrystalline are the intrinsic behavior, and through a selection of proper crystal structure and chemistry, they can affect the magnetic factors for certain utilization.

**Exchange Anisotropy** The exchange anisotropy is almost a new phenomenon, which is produced by an interaction between an antiferromagnetic and ferromagnetic material [25]. The source of the interaction that lines up the spins in the magnetic system is a result of the exchange interaction. The occurrence of such anisotropy has been considered to have a fundamental role in information storage technology.

**Surface Anisotropy** The contribution of surface anisotropy in magnetic particles occurs once their sizes reduce to a nanosized scale. It mostly seems to appear in grain boundaries and has a crystal field nature [26].

### 1.3.2 Extrinsic Properties

Extrinsic or extensive properties of magnetic materials depend on the quantity of matter (for example, weight). Some examples of the extrinsic properties of magnetic materials are hysteresis loop, magnetic loss, and permeability. These properties are influenced by microstructure characteristics [9]. However, chemical microstructure may possess deleterious influences; sometimes taking benefit of chemical aspects is carried out to develop microstructural as well as properties such as electrical and magnetic properties. Undoubtedly, the microstructure conundrums are the most crucial issues that must be considered to achieve a desirable property and reproduce a high quality of ferrites. To improve the property of ferrite to be used in high-frequency applications, the control of microstructure characteristics,

i.e. pores, sintered density, grain size, and distribution, is more crucial than the choice of chemistry [20, 27–35]. Indeed, the porosity as well as grain size and their distribution, as a microstructure feature, are the two main factors that affect magnetic properties such as initial permeability [20, 36, 37]. Recent studies on nickel ferrite and nickel-zinc ferrite, for example, revealed that grain size has a more key role in initial permeability than porosity. In soft ferrites, sometimes, the effects of these parameters on permeability become more complicated. Therefore, the importance of controlling the microstructure characteristics becomes more evident. However, the experimental effects of grain size are well known, and a systematical science about physical phenomena has not been completely developed. In fine-structured materials, normally there is no domain wall within the grain; thus, the permeability is accomplished by the means of the rotational process. However, in the single-domain state, one cannot easily determine the critical grain size. As the size of the grain enhances, the domain walls within the grains would increase. They may either be nailed at intergranular pores or grain boundaries. Porosities have an important role in the restriction of the domain wall movement as well as in the contribution of the demagnetizing field. Therefore, the domain wall motion is sensitive to porosity distribution and grain size as well. The existence of porosity at the grain boundaries and within the grains can affect the hysteresis, coercivity, and permeability.

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