

Engineering Materials

Gagan Kumar Bhargava

Sumit Bhardwaj

Mahavir Singh

Khalid Mujasam Batoo *Editors*

Ferrites and Multiferroics

Fundamentals to Applications

 Springer

Engineering Materials

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Editors

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Editors

Gagan Kumar Bhargava
Physics
Chandigarh University
Mohali, India

Sumit Bhardwaj
Physics
Chandigarh University
Mohali, India

Mahavir Singh
Physics
Himachal Pradesh University
Shimla, India

Khalid Mujasam Batoo
King Saud University
Riyadh, Saudi Arabia

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Preface

Ferrites or Magnetic materials are an important class of materials as they possess high resistivity, high saturation magnetization and initial permeability and negligible eddy current losses. The high resistivity of ferrites makes them suitable for microwave applications and with the tremendous increase in the development of microwave and millimeter wave-based communication technologies, the production of dielectric resonators has emerged as one of the most rapid growth areas in electro-ceramic manufacturing. High saturation magnetization of ferrites is a prerequisite condition for making them useful in storage sectors. In ferrites, the electrical and magnetic properties mainly depend on the synthesizing technique, type of metal ions and their distribution between the tetrahedral (A) and octahedral (B) sites. So due to the ability of ferrite materials to distribute the metal ions over A- and B-sites, material scientists became very much curious far a long time and are still working on the synthesis and characterization of various possible ferrites substituted with different metal ions to cope with the advanced technologies and applications. At the other end, multiferroic materials, i.e., coexistence of ferroelectric and ferromagnetic character have attracted much interest in their potential applications in information storage, the emerging field of spintronics, and sensors. The magnetization can be rotated or even reversed by the reversal of external electric field. The polarization can also be reversed by the reversal of external magnetic field. One of the main aim in preparing this book was to highlight the complex magnetic behavior along with unique coupled magneto electric behavior. This book highlights the fundamentals of ferrites and multiferroic materials with special attention to their structure, types, and properties. The book will provide a platform for critical evaluation of many aspects of dielectric, ferromagnetic coupled electromagnetic issues at the forefront of material science today. It highlights a comprehensive survey about the ferrite and multiferroic materials. There are 12 chapters organized in a systematic way. The first chapter takes the broad view of ferrites and their structures. Second chapter describes the effect of synthesizing techniques on the properties of ferrites. The third and fourth chapter deals with the substitution effects on the electric and magnetic properties of the ferrites. The fifth, sixth and seventh chapters explore the role of ferrites in bio-medical, high frequency antenna and water purification applications

respectively. Chapter eight takes the broad view of multiferroic materials. The chapter nine describes the effect of synthesis on the properties of BFO multiferroic while the chapter ten deals with the multiferroic phenomenon in different forms of materials. The chapter eleven describes the synthesis and properties of $\text{BiFeO}_3\text{-BaTiO}_3$ multiferroic while the twelfth chapter deals with the various applications of multiferroic materials. We hope that the book will emerge as the primary text dealing with general aspects of ferrite and multiferroics and will prove useful for all the various people interested in ferrites and multiferroics: from graduate level to advanced specialists in both academic and industrial settings. At the end we are thankful to Sh. Jagdish Chand and Mrs. Ritika Bhargava for constant encouragement and suggestions during the entire process.

Mohali, India
December 2021

Dr. Gagan Kumar Bhargava

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and Ritesh Verma

About the Authors

Dr. Gagan Kumar Bhargava is currently working as a Professor at the Department of Physics, Chandigarh University, Gharuan, Mohali, Punjab, India. He has obtained his Ph.D. degree in Physics from Himachal Pradesh University, Shimla, India. His current research interests are synthesis of nanomaterials for antenna, EMI shielding, electromagnet and bio-medical applications. He has more than 14 years of teaching and research experience. Before joining the Chandigarh University, he was the Head of the Applied Science Department at Green Hills Group of Institutes, Solan, India and also served as an Associate Professor at IEC University, Solan, India. He has published more than 50 research papers in different peer-reviewed international journals and 6 book chapters. He has guided 2 Ph.D's, 1 M.Phil and 12 M.Sc. dissertations. He was the former Editorial Board Member of American Journal of Nanosciences and acted as Editor for American Institute of Physics Conference Proceedings in 2018 for AMDFA-2018.

Dr. Sumit Bhardwaj is currently working as an Assistant Professor at the Department of Physics, Chandigarh University, Gharuan, Mohali, Punjab, India. He has obtained his M.Tech. degree in Metallurgical and Materials Engineering from Thapar University, India and Ph.D. degree in Materials Science and Engineering from National Institute of Technology, Hamirpur, India. His current research interests are in the field of ferrite, multiferroic and composite materials. He has more than 12 years of research and teaching experience. Before joining to Chandigarh University, he has worked at Punjab Engineering College (Deemed to be University), Chandigarh, India. He has published more than 23 research papers in different peer-reviewed international journals and 3 book chapters. He has supervised two M.Tech. dissertations, and currently he is supervising three Ph.D. students.

Dr. Mahavir Singh is a Professor at the Department of Physics and Dean Physical Science Himachal Pradesh University, Shimla, India. He has more than 30 years of teaching experience. He remains head of the department during 2007-2009, 2015 and Dean Engineering Himachal Pradesh University 2017-2018. He was Common Wealth Fellow, UK (2006-2007), Visiting Fellow, Faculty of Engineering, University

of Dundee, UK (2006-2007), Visiting Professor at, Faculty of Engineering University De Bretagne Occidentale, Brest France (2007-2008) and Vice Chancellor at IEC University, Solan, India (2015-17). He was awarded with 100 Most Influential Vice Chancellor Award 2016 by World Education Congress, Global Achiever Award 2014 by Indian Heritage Institute and Calcutta Bharat Excellence Award 2015 from Friendship Forum New Delhi. He has authored book chapter Complex magnetic nanostructure (Springer), The Basis of Nanomagnetism: An Overview of Exchange Bias and Spring Magnets CRC Press, more than 5 textbooks on different areas of physics. He has published more than 190 research papers in different peer-reviewed international journals and about 100 Invited lectures delivered in International conferences. He has guided 23 Ph.D. students and evaluated 60 Ph.D. thesis. He was among Top 2% Scientist Globally as per Stanford University USA 2020 lists.

Dr. Khalid Mujasam Batoo is an Associate Professor at King Abdullah Institute for Nanotechnology, King Saud University, Riyadh, Saudi Arabia. He has authored more than 145 research papers published in peer-reviewed Journals of International commendation and conference papers. He is currently serving as the editorial board member of more than 13 international Journals. Among his various research interests include the study of magnetic nanomaterials, graphene materials, solar cells, batteries, e-textiles, magnetic tunnel Junctions and spintronic materials with an emphasis to understand micro and nano-structural properties of these materials. He has authored several book chapters published by Bentham Science Publishers and Materials Research foundation.

Basics of Ferrites: Types and Structures



Pooja Dhiman, Garima Rana, Dipanshi Goyal, and Ankush Goyal

Abstract This chapter summarizes the detailed introduction, types of magnetism, classifications of ferrites and their crystal structure. In addition, this chapter has the main focus on the magnetic properties possessed by ferrites on the basis of the soft and hard nature of ferrite materials. This chapter summarizes the recent work on the magnetic and dielectric properties of ferrites. Ferrites are primarily known for their high resistivity and highly magnetic nature which makes them suitable material over a wide range of applications like magnetic storage devices, antennas, transformers and other high-frequency devices. A great benefit of ferrites nanoparticles is the porosity, mandatory for sensor applications too. In short, ferrites belong to the materials which are a potential candidate for widely spread applications.

Keywords Ferrites · Nanomaterials · Soft and hard ferrite

1 Introduction

A ferrite ‘a ceramic substance’ composed primarily of iron oxide (Fe_2O_4), with a small number of metals like barium, manganese, nickel, and zinc etc. [1–3]. The term “ferrite” comes from the Latin word “Ferrum,” this is meant for “iron.” Both the iron oxide and the metal are ferrimagnetic. Ferrite, a ceramic substance created by combining iron oxide with a metal. It is thought that ferrites were first found in ancient Greece around 800 BC. Ferrite belongs to iron oxide-based magnetic oxide. Forestier used a heat treatment procedure to manufacture ferrites in 1928 [4]. Snoek [5] developed many ferrites as commercially important materials in 1947. Ferrimagnetic materials have uneven opposing magnetic moments, which allows them to keep their spontaneous magnetization [6, 7]. Chemical composition, particle size, and particle

P. Dhiman (✉) · G. Rana · D. Goyal · A. Goyal
School of Physics & Materials Science, Shoolini University of Biotechnology and Management Sciences, Bajhol, Solan 173229, H.P., India

P. Dhiman
International Research Centre of Nanotechnology for Himalayan Sustainability (IRCNHS),
Shoolini University, Solan, India

interaction with the neighbouring matrix all influence the properties of ferrite NPs. Ferrite NPs have a wide range of applications in various fields ranging from biomedical to industrial applications [8, 9]. Ferrites NPs are commonly used magnetic materials and have substantial scope in the field of biomedical sciences, such as tumour treatment, drug delivery [10, 11], magnetic resonance imaging (MRI) [12–14], biomagnetic separation [15], controlled drug release, cellular therapy, tissue repair, cell separation, purifying of cells, magnetoception, severe inflammation, disability [16–18]. In industrial applications, ferrite nanoparticles are often used as adsorbents and catalysts [19–25], manufacturing of electronic materials [26–28], and wastewater treatment [29–32]. These materials attracted a lot of attention as magnetic nanoparticles due to their unique structural features, surface reactivity, electrical, and magnetic properties, all of which are impacted by the nano-structured phase [2, 33], and these features of nanomaterials differ from bulk materials. The particle sizes of nanoparticles are the range of 1–100 nm. At the nano-scale, nanoparticles have distinctive physical, chemical, and biological properties as compared to their counter parts at larger dimensions. This is due to a greater surface area to volume ratio, increased chemical reactivity or stability, increased mechanical strength, and so on. The macro-structured parameters are the same as the bulk material's. As the abundance of surface particles increases, the particle size decreases. Surface atoms have a minor coordination number than inside atoms, allowing them to move freely. Quantum size effects are observed when the NPs' size is closer to the de-Broglie wavelength limit and their width is less than the quasiparticle interaction [34]. Controlling and manipulating the properties of nanomaterials is attainable by controlling their size during production using various approaches. A metal's electronic areas are characterised by its electronic band structure. The confirmation of band structure is determined by particle size. The delocalized bands can be seen in the molecular states. The energy gap between consecutive lines is determined by the particle size. The distance between the energy levels rises when the size decreases. There are various classifications of magnetic materials, i.e. dia, para, ferro, antiferro, and ferromagnetic materials [35]. In 1831, Faraday published a novel disciplinary law of magnetic induction [36]. Following that, in the nineteenth and twentieth centuries, the Bohr model of the atom and the Dirac hypothesis explained the circulation of currents in atoms. Weiss introduced the next domains theory to explain ferromagnetic behaviour [37]. Neel proposed the next radical approach. Neel suggested a theory to explain the hysteresis loop and magnetism. In almost every field, magnetic materials are extremely popular. Magnetic materials are used in soft magnets, permanent magnets, recording, magnetic storage, and alluring levitation, among other things [38]. All of the properties of nanomaterials are primarily resolute by their shape and size. Controlling and manipulating the properties of nanomaterials has become achievable by monitoring the particle size distribution by varying the synthesis methods [39, 40].

1.1 Advantages of Ferrite NPs

In terms of electrical resistivity, ferrites nanoparticles outperform other magnetic materials, resulting in reduced eddy current losses over a high frequency. The application of ferrites in quality filter circuits, transformers which works at high frequencies, wide-band transformers, and adjustable inductors has increased due to additional properties such as high permeability and temperature constancy. Ferrites nanoparticles are regularly utilized to fabricate the electrical circuits used for low frequency applications and devices operating at the high-frequency efficiency of other circuit components improve. Ferrites nanoparticles are the finest interior material option for frequencies between 10 kHz and 50 MHz because they provide the best arrangement of high Q, low cost, high stability, and low volume. In terms of magnetic and mechanical parameters, ferrites are unrivalled flexibility.

- High resistivity
- High Q/small package
- High permeability low loss achieved
- Low cost
- Large selection material
- Shape versatility
- Flexibility in the choice of core shapes
- Economical assembly
- Wide frequency range
- Temperature and time stability.

2 Magnetic Material and Types of Magnetism

The electronic structure of an atom within a compound is used to classify its magnetic behaviour. The main significant property of a magnetic material, magnetic susceptibility which is expressed by:

$$\chi = M/H \tag{1}$$

where M is used for the magnetization value and H corresponds to the applied magnetic field, with units of A/m . Magnetic susceptibility is dissimilar for a piece of material and depends upon the temperature and is given by:

$$\chi = C/(T - \theta) \tag{2}$$

Here, C and θ are constants that are dissimilar for each material [41]. Figure 1 shows the classification of magnetism. There are different types of magnetism.

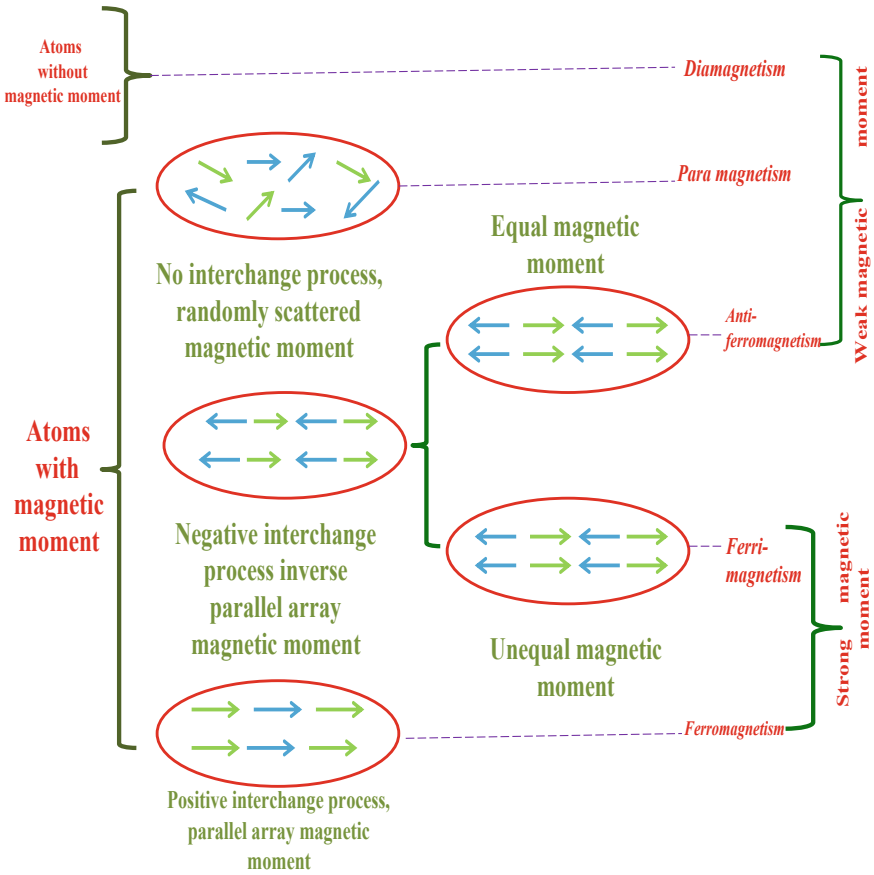


Fig. 1 Schematic presentation of classification of magnetism

2.1 Diamagnetism

The spin and orbital moments of electrons in an enclosed shell in an atom are normally aligned in such a way that the atom as a whole has zero net moment. Diamagnetic materials exhibit magnetic permeability values of ≤ 1 , as susceptibility is expressed as $\chi \cdot \nu = \mu \cdot \nu - 1$, and the values lie between -10^{-6} and -10^{-5} . If the material contains a net magnetization and long-ranged arrangement of magnetic moments, the impacts of these atomic current loops are mitigated.

2.2 Paramagnetism

The number of electrons which are not paired and reside in atomic shells, particularly the '3d' and '4f' shells of every other atom, is responsible for all other sorts of magnetic activity found in materials. When the applied magnetic field is eliminated, however, fluctuations which occur thermally cause the magnetization of paramagnetic atoms to shift arbitrarily. These materials have a comparative magnetic permeability of ≥ 1 i.e. $\mu > \mu_0$ indicating that their susceptibility is +ve and tiny, on the scale of 10^{-3} to 10^{-5} , and so are influenced to magnetic fields. Curie's law describes this phenomenon in the availability of a particularly poor magnetic field.

$$\chi = \frac{M}{H} = \frac{C}{T} \quad (3)$$

Here C is known as the Curie constant.

2.3 Ferromagnetism

Ferromagnetic materials contain 'atomic magnetic moments which are oriented ones' of comparable size, and their crystalline structure facilitates direct coupling contacts between the moments, which can boost flux density significantly. Moreover, the oriented moments in magnetic particles can impart magnetic behaviour in the absence of an exterior magnetic field. Hard magnets are those materials that maintain permanent magnetization even when no magnetic field is applied. Ferromagnetic materials have very high magnetic susceptibilities and the atom has a parallel aligned magnetic moment, which produces strong magnetization. The value of susceptibility (χ) is large and lies below the Curie temperature.

$$\chi = \frac{C}{T - \theta} \quad (4)$$

where C is known as the Curie temperature, which is the temperature at which exchange forces are present, i.e., randomization happens because of thermal energy, as it happens in a paramagnetic system.

2.4 Ferrimagnetism

In zero applied fields beyond a certain temperature is called Neel temperature. Ferrimagnetism is a property of the material whose atoms or ions interact to congregate an order, but not a parallel arrangement. The antiparallel alignment of ions leads in

a significant net magnetization within a magnetic region in most cases. In comparison to ferromagnetic materials, these have a higher saturation magnetization. The value of susceptibility (χ) is large and exists below the Curie temperature. The value of susceptibility (χ) is large and exists below the Curie temperature. Ferrites (for example, Fe_3O_4) are ferrimagnetic minerals in which metal cations and oxygen anions are arranged in the crystal lattice.

2.5 Antiferromagnetism

Antiferromagnetism belongs to a type of magnetism in which neighbouring ions act as minuscule magnets and impulsively align them into opposing or antiparallel configurations throughout the material at relatively low temperatures, resulting in a material with essentially zero external magnetism. Very feeble magnetic susceptibility (χ) of the order of paramagnetic materials is the characteristics of these materials. Above the Neel temperature (TN), thermal energy is sufficient to randomize the atomic moments aligned in the opposite direction, causing their long-range order to vanish. The material shows paramagnetic behaviour in this state (Table 1).

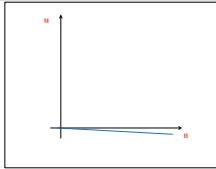
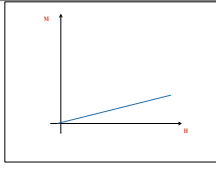
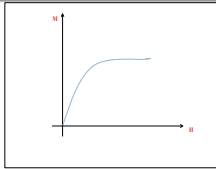
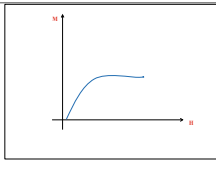
3 Classifications of Ferrite NPs According to Magnetic Behavior

Soft and hard ferrites are classified according to their potential to be magnetized and/or demagnetized instead of their capacity to withstand penetration or erosion [42, 43]. Coercive force (H_c) is the prime essential qualities of magnetic substances, and it distinguishes both types of ferrites from each other. Figure 2 shows the hysteresis loop of soft ferrite and hard ferrite (Table 2).

3.1 Soft Ferrites

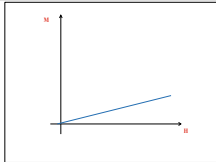
Soft ferrites are easily magnetized and demagnetized because they have low resistivity. A small hysteresis loop occurs in soft ferrite. These materials have soft magnetic properties like high permeability, low coercive value and high saturation magnetization value. Soft ferrites have diverse applications in the electronic industry, including the development of transformer cores, high frequency inductors, and microwave components [6]. Soft ferrites are those that contain Ni, Zn, or Mn and are utilized in transformers or electromagnetic cores. Because at high frequency, the soft ferrites possess very low losses therefore can be commonly used in RF transformers and also in inductors.

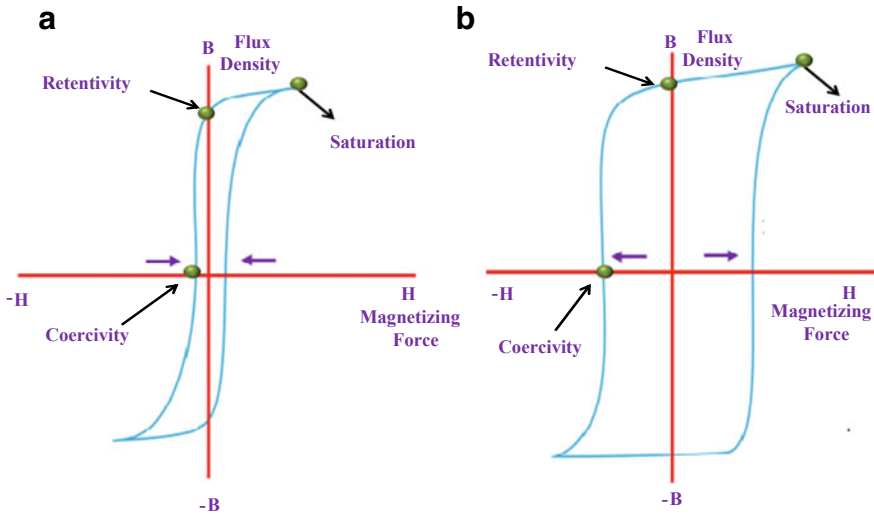
Table 1 Comparison of different types of magnetism

Type	Temperature dependence	M-H loops	Value of χ	Example
Diamagnetic	Temperature independent		-10^{-6} to -10^{-5}	As in covalent solids such as Au, Cu, and Ag, the atoms' shells are enclosed
Paramagnetic	Follows Curie's law $\chi = \frac{M}{H} = \frac{C}{T}$		$+10^{-5}$ to $+10^{-3}$	As in transition metal ions, atoms possess arbitrarily oriented magnetic moments
Ferromagnetic	Below Curie's temperature		Positive and large Fe $\sim 100,000$	In some transition metals and rare earths, such as cobalt and nickel, atoms with identically induced magnetic moments process substantial persistent magnetization without an external magnetic field
Ferrimagnetic	Below Curie's temperature		Positive and large Ba ~ 3	Atoms having antiparallel aligned magnetic moments, such as magnetite and ferrite, have high magnetism in the absence of a magnetic field

(continued)

Table 1 (continued)

Type	Temperature dependence	M-H loops	Value of χ	Example
Anti-ferromagnetic	Below Neel temperature		$+10^{-5}$ to $+10^{-3}$	Magnetic moments are present in both parallel and antiparallel oriented atoms. The most frequent transition metal oxides and salts are MnO and NiO

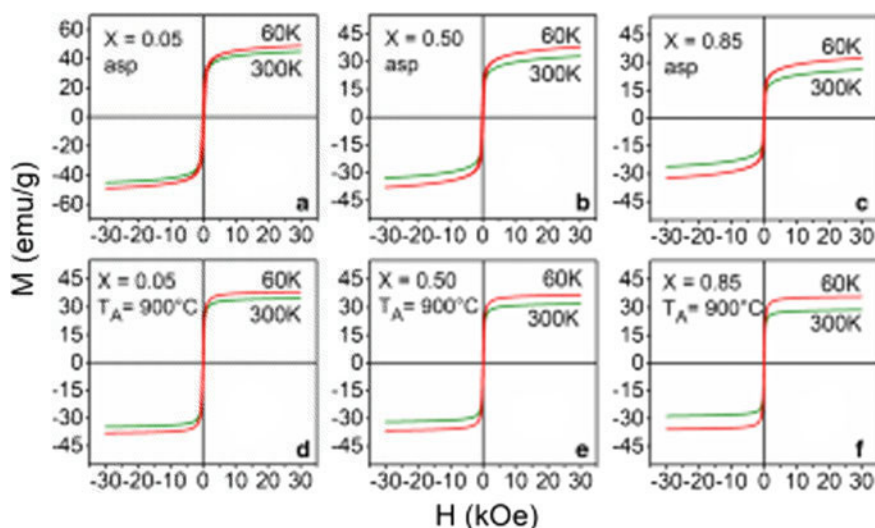
**Fig. 2** Depicts the hysteresis loop of **a** soft ferrite, **b** hard ferrite

3.1.1 Magnetic Property of Soft Magnet: Nickel Ferrite (NiFe_2O_4)

Magnetic materials, depending on the spin orientation, exhibit various kinds of magnetic ordering. The magnetization of ferrites is due to the interaction of the spins with the exchange of neighbouring atoms. Anumol et al. [44] used the sol-gel process to make magnesium-substituted Ni-ferrite ($\text{Mg}_x\text{Ni}_{1-x}\text{Fe}_2\text{O}_4$; $0 \leq x \leq 1$) nanoparticles, which were then annealed at 550 and 900 °C. Figure 3 shows the (as prepared and annealed at $T = 900$ °C) $\text{Mg}_x\text{Ni}_{1-x}\text{Fe}_2\text{O}_4$ nanoparticles samples with

Table 2 Essential properties of both type of magnetic ferrites (soft and hard)

Soft ferrite	Hard ferrite
Low coercivity	High coercivity
High electrical resistivity	Not important
High Curie temperature	High Curie temperature
Low anisotropy	High anisotropy
Low magnetostriction	Not important
High permeability	Low permeability
High saturation magnetization (1–2 T)	High saturation magnetization (0.3–6 T)
High Curie temperature	High Curie temperature

**Fig. 3** Hysteresis loop of the **a–c** as-prepared and **d–f** annealed ($T_A = 900\text{ }^\circ\text{C}$) Mg–NiFe₂O₄ nanoparticles sample [44]. Reprinted with permission from Springer Nature

$x = 0.05, 0.50,$ and 0.85 magnetic hysteresis (M–H) loops. The hysteresis loops did not drencheven at a maximum magnetic field strength of 30 kOe, as shown in Fig. 3. Furthermore, the annealed samples' loops seem to be more saturated than the as prepared samples. The corresponding magnetization values were calculated by extending the M–H loop from high field to zero applied fields. At 300 and 60 K with an increase in the concentration of Ca²⁺, the values of magnetic saturation increase. At room temperature, Chavan et al. [45] found that the highest M_S of 64.5 emu/g for $x = 0$, i.e. Nickel ferrite NPs. At room temperature, Moradmard et al. [46] registered a maximum M_S value of 28.8 emu/g for $x = 0$. For $x = 0.05$, the maximum M_S values were 42 and 46 emu/g at 300 and 60 K, respectively. In addition, the M_S value of the annealing temperature at 550 °C is smaller than that of the asp sample. The M_S

value for annealed at 900 °C samples declined gradually from the value of 37 emu/g ($x = 0$) to 29 emu/g ($x = 0.25$) at 300 K.

The M_S value increased marginally to 31 emu/g as the x value increased to 0.50, but then decreased as the Mg^{2+} concentration increased. However, for the samples which were annealed at 900 °C, a similar pattern was observed at 60 K. The bare Mg-ferrite sample, annealed at 550 °C, had the lowest M_S values of 21 emu/g and 26 emu/g at 300 and 60 K respectively. At both 300 and 60 K, H_C decreased as Mg^{2+} doping (%) increased. It's important to note that at 300 K, the H_C values for the 900 °C-annealed samples were consistently higher than those for the as prepared and 550 °C-annealed samples. Though the H_C values were higher at 60 K, they were the lowest of the 900 °C-annealed samples. The as-prepared and 550 °C-annealed samples with $x = 1$ had the lowest H_C of 10 Oe at 300 K, while the asp sample with $x = 0$ had the maximum H_C of 340 Oe at 60 K. The non-saturated magnetization of samples increased with increasing Mg^{2+} content for the as-prepared samples and the samples annealed at 550 °C with substitution values between 0.05 and 1, i.e. for $0.05 \leq x \leq 1$, where as it did not change for the samples annealed at 900 °C [44]. The observed magnetic behaviour in these nanoparticles samples is explained by crystallite size distribution. Another factor is decreased ionic magnetic moment, and the most important cation distribution in the spinel type of ferrites, and decreased anisotropy with increasing Mg^{2+} concentration [47].

According to Maaz et al., nickel ferrite coercivity increases with particle size (for microscopic particles) when the particle changes its behaviour from a super-paramagnetic type, i.e. from blocked to unblocked. This happens for very narrow sized particles because thermal energy overcomes the anisotropy, which is volume dependent. While the coercivity reduces with size for larger particles because of two unique mechanisms; first one is that bulky particles can maintain domain wall motion, and the second one is that surface role varies and bulk anisotropies are observed as the size is reduced [48]. Atif et al. prepared Zn doped nickel ferrite by the sol-gel method and the highest magnetic saturation values were found, i.e. 76 emu/g. The saturation magnetization increases with an increase in the zinc content [49]. The value of magnetic saturation and coercivity increase with an increase in the concentration of dopant [50].

3.1.2 Electrical Property of Soft Ferrite

The ferrite system acts as a heterogeneous system with various conducting properties of grains and grain boundaries. Koop believed that both the grains and the limits were parallel to R.C. Circuit loops. When an A.C. is exposed to a dielectric material, the series arrangement of these parallel circuits replicates the conduct of the inhomogeneous dielectrics as a whole, due to the comparable current flow through grains and grain boundaries. There is a relative movement of positive and negative ions in the electric field, which results in an electric dipole moment. Atif et al. prepared Zn doped $NiFe_2O_4$ nanoparticles by sol-gel technique and the value of dielectric permittivity ranged from 2.77 to 12.2 kHz [49]. Moradmard et al. magnesium doped

nickel ferrite nanoparticles are fabricated by co-precipitation technique and study the various parameters like dielectric loss, dielectric constant, and ac conductivity with frequency range 1–12GHz [46]. The related dielectric pictures are shown in Fig. 4 (Fig. 5).

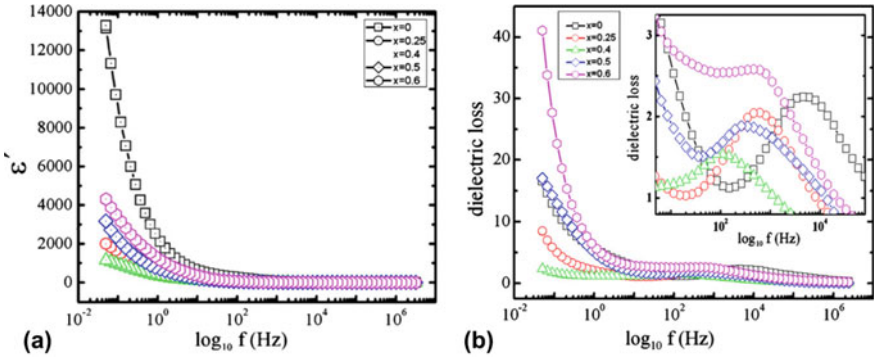


Fig. 4 a Variation of dielectric for the $Ni_{1-x}Zn_xFe_2O_4$. b Variation of dielectric loss for the $Ni_{1-x}Zn_xFe_2O_4$ [49]. Reprinted with permission from Elsevier

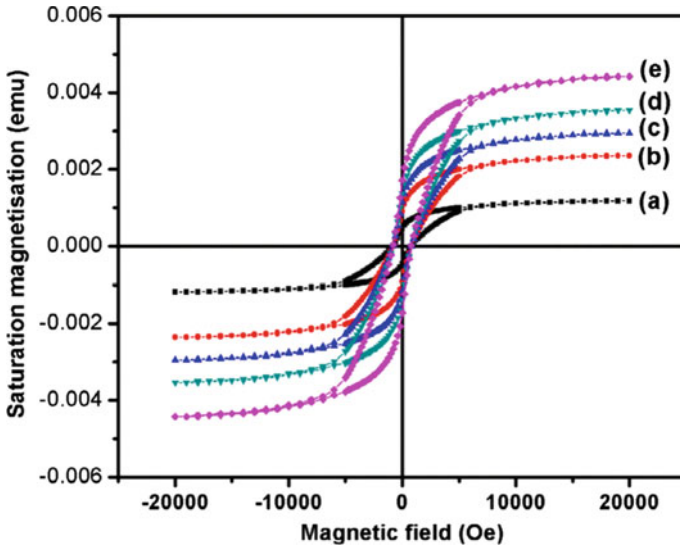


Fig. 5 M–H curve of a Pure barium ferrite and b Cu-2 mol%, c Cu-4 mol%, d Cu-6 mol%, and e Cu-8 mol% doped barium ferrite [51]. Reprinted with permission from Elsevier