
chapter I

Introduction to Ferrites

CHAPTER - I

INTRODUCTION TO FERRITES

1.1 Introduction

Ferrites are industrially important ferrimagnetic materials. In which ferric-oxide is the basic constituent and the remaining part is some divalent oxide. The structural, magnetic and the electrical properties of these material have been the subject of tremendous interest, to physicist, chemist and materials scientists. They are semiconductor in nature possess very high resistivity in the range 10^2 to $10^{11}\Omega$ -cm. The magnetic properties like high permeability small coercive forces and the electrical properties like resistivity, low power loss due to eddy currents along with low electrical conductivity of ferrites depend upon the chemical constituents and the method of preparation cation distribution etc.

For a particular application in an electrical or electronic industry these can be tailor made by using correct method of preparation and the constituents.

The Fig. (1.1) shows the wide spectrum of application of ferrites as "ferrite tree".

1.2 Historical

The 'ferrites' are an artificial version of load stone [Fe_3O_4] also called ferrous ferrite, the first magnetic material known to man.

During 600 BC [1] Greeks discover magnetism in the mineral lodestone (Magnetite), the first ferrite. About 1000 AD, first application come out. Mariners use lodestone in compasses. The modern period began with the work of Snoek (1947) and the development of ferrite material started as outlined significantly in Table [1.1].

Table 1.1

Scientist	Area of work	Significance	Ref.
Pierre, Weiss (1907)	Magnetic Properties & Curie temp. of Fe_3O_4	Development of Ferrites	2
Du Bios (1890)	Measurement of saturation magnetisation of Fe_3O_4	Structure of ferrite	3
Hilpert (1909)	Synthesis of ferrites	Success to reduce losses in inductor and transformer	4
Hedwall (1912)	Preparation of ferrites by solid state reaction	Established the theory of solid state reaction	5
Kato, Takai, Barth, Forrester (1930, 1939, 1932)	Magnetization and other views	The structure of ferrites was established to be spinel type	6, 7, 8
Snoek (1936, 1947)	Preparation & Magnetic prop.	Developed commercialy usefull Ferrites	9, 10
Verwey (1947)	Electrical properties	Established the reason of electronic conductivity in ferrites	11

	Crystal structure	Shows inverse structure ferrites are ferrimagnetic & normal spinel structure are non-magnetic	
Anderson & Van Vleck (1950)	Basic interactions	Developed theory of super- exchange interactions	12
Neel (1948, 1952)	Spin - spin interaction	Provide a theoretical key & introduce the concept of mag.sub-lattice	13 14
Yafet & Kittel (1952)	Theory of mag. sublattices	Formulate theory of a "triangular" arrangement three sublattices	15
Gorter & Guiland (1954, (1951))	Magnetization	Gave direct expt. proof of Neel's theory	16 17
Smorter & Gorter (1954)	Magnetisation & microwave resonance	Corelated independently the cation distribution	18 19
Gilleo (1957)	Magnetisation	Corelate Curie temperature with cation distribution & magnetisation	20
Shull & Strauser (1951)	Neutron diff- raction	Confirm Neel's theory for Magnetite & Zn-ferrite	21
Koops (1951)	Conductivity	Gave phenomenological theory to explain conduction behaviour	22
Bertaut's (1951)	X - ray diffraction study	Cation distribution in ferrites	23
Gorte (1954)	Microwave reso- nance and magnetisation	Cation distribution in ferrites	24

1.3 Crystal Structure of Ferrite Material

Generally ferrites shows different types of crystal structures.

- 1) Spinel structure
- 2) Hexagonal structure
- 3) Garnet structure
- 4) Perovskite structure

Out of these four crystal structure, the spinel structure is discussed in detail. Its details are given below.

1.3.1 Chemical structure of spinel ferrites

Ferrites are magnetic substances exhibiting the spinel structure [25] Fig. 1.2 having the general chemical formula $Me.Fe_2O_4$ or $MeO.Fe_2O_3$ or MFe_2O_4 where M is a divalent metal ion [M^{2+}] like Co, Ni, Mn, Mg, Cu, Cd and iron is a trivalent metal ion [Fe^{3+}].

Ferrites formed by single divalent atoms for ex. $CdFe_2O_4$, $MgFe_2O_4$, $ZnFe_2O_4$ etc. are termed as simple or single ferrites whereas those in which divalent atoms are partially by other metal ion and partially replaced by one metal ion are called binary or mixed ferrites.

For ex. $ZnCo_{1-x}Fe_2O_4$, $Cd_xMg_{1-x}Fe_2O_4$ etc.

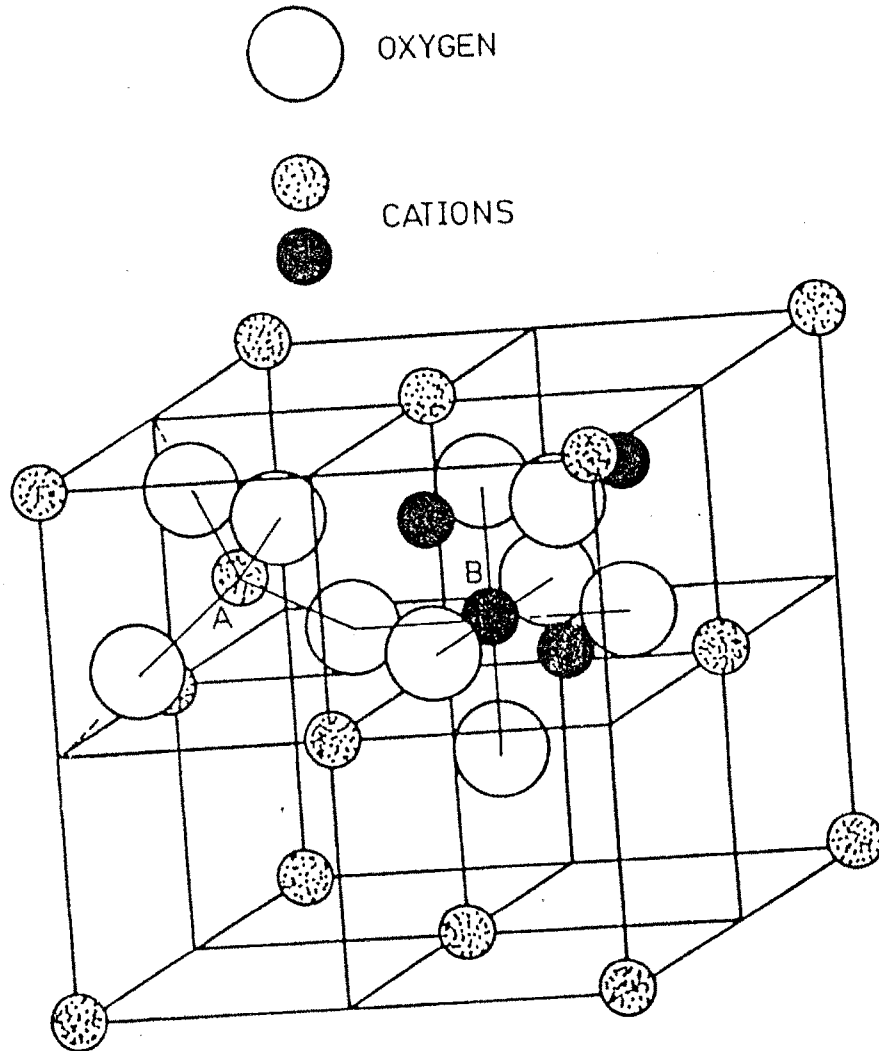
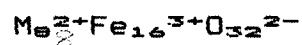
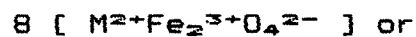


Fig.12 THE SPINEL STRUCTURE .

1.3.2 Lattice structure of spinel ferrites

Crystallographically the structure of ferrites may be derived from naturally occurring compound $MgAl_2O_4$. The spinel crystallize in the cubic system and contain eight formula units $MgAl_2O_4$ in the unit cell. Thus the ferrites derived from $MgAl_2O_4$ by substituting Fe^{3+} for Al have the spinel structure. The spinel ferrite is an ionic crystal and essentially have cubic face centered crystal structure. The smallest cubic unit cell consists of eight molecules of MFe_2O_4 having the lattice parameter 'a' from 8.3 A to 8.6 A. Hence the unit cell formula becomes



The 32 oxygen ions are arranged on a more or less close packed face centered cubic lattice. There are 96 interstices available between these 32 oxygen ions. Out of which the metal ions occupy suitably 24 interstices. The oxygen close packed fcc lattice structure is shown in Fig. (1.2). The 96 interstitial sites are divided into two types.

1) Tetrahedral or A-site in which metal ion is surrounded by four oxygen ions located at the corners of a tetrahedron as shown in Fig. (1.3a) and

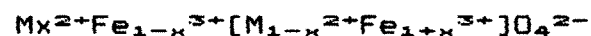
2) Octahedral or B-site in which the metal ion is surrounded by six oxygen ions placed at the vertices of an octahedron as shown in Fig. (1.3b).

Out of these 96 interstitial positions 64 are tetrahedral and 32 are octahedral. Again of these only 8 tetrahedral and 16 octahedral sites are occupied by cations per unit cell.

According to Goodenough and Loeb [26] the site preference energies play an important role in determining the cation distribution.

1.3.3 Classification of ferrites

On the basis of cation distribution Barth and Posnjak [8] have classified ferrites in three groups. The general formula for cation distribution of a spinel formula for cation distribution of a spinel ferrite may be written as



Where M stands for divalent metal ion

1.3.3a Normal Spinel ferrites

In normal spinel ferrites, all the 8A sites are occupied by divalent metal ions (8Me²⁺) and all the 16B sites are occupied by trivalent ions (16Fe³⁺). The cation distribution can be written as

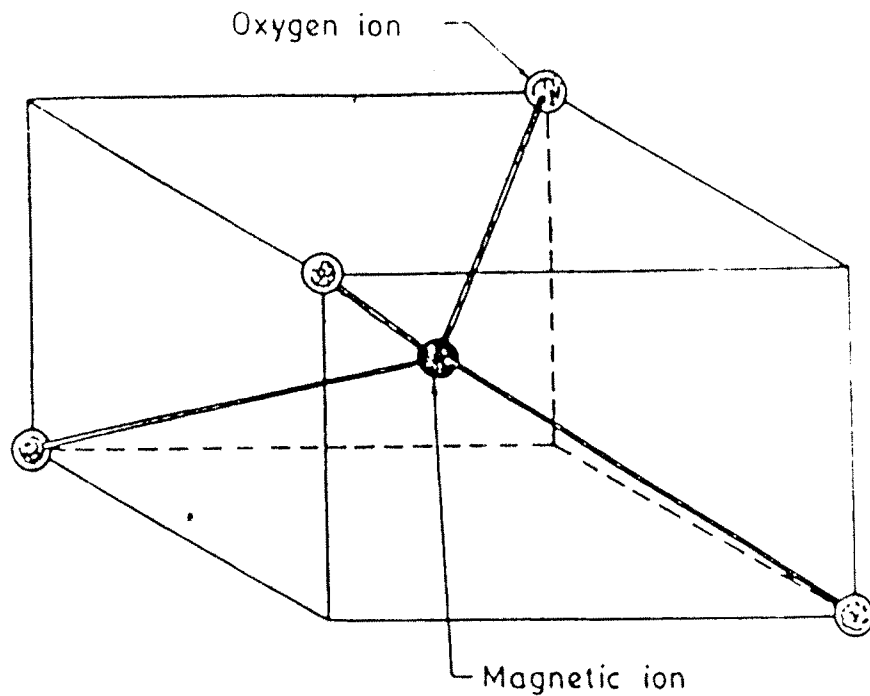


Fig.1.3. a) THE MAGNETIC ION OCCUPIES A "TETRAHEDRAL LATTICE SITE" WHERE IT IS SURROUNDED BY FOUR NEAR-NEIGHBOUR OXYGEN IONS.

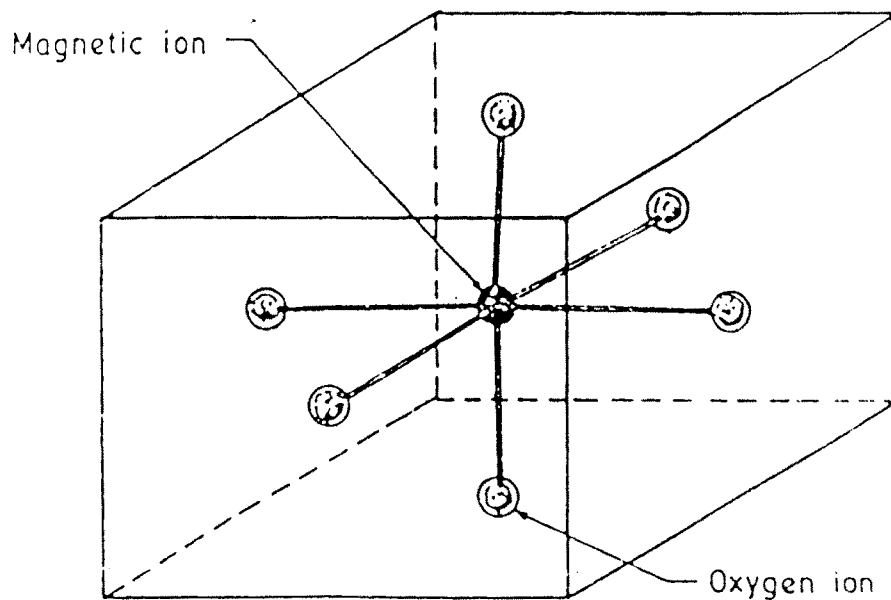
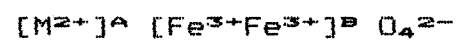


Fig.1.3 b): THE MAGNETIC ION IS SAID TO OCCUPY AN "OCTAHEDRAL LATTICE SITE" WHERE IT IS SURROUNDED BY SIX NEAR-NEIGHBOUR OXYGEN IONS.

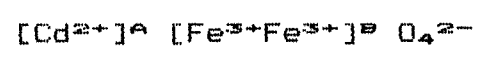


This type of ferrite are non magnetic in nature.

E.g. CdFe₂O₄, ZnFe₂O₄

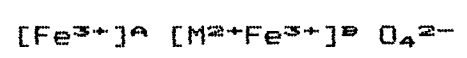
The normal ferrites do not show ferrimagnetic behaviour.

The cation distribution of CdFe₂O₄ is thus given by



1.3.3b Inverse spinel ferrite

In inverse spinel structure 8 metal ions (M²⁺) occupy B-sites and 8 (Fe³⁺) ions occupy A-sites and remaining 8 Fe³⁺ ions occupy B-site. The structural arrangement can be written as



E.g. CoFe₂O₄, Fe₃O₄, NiFe₂O₄, CuFe₂O₄

This types) of ferrites are magnetic in nature.

1.3.3c Random spinel structure

Completely normal and inverse ferrite represents an extreme case. X-ray and neutron diffraction studies have shown some intermediate structure known as random spinel ferrite. When the divalent [Me²⁺] and trivalent [Fe³⁺] metal ions are randomly distributed over A and B sites depending on their physicochemical conditions of

preparation and compositional variation. Such a ferrite is partially inverse.

The general cation distribution may be represented as



Where x = coefficient of normalcy

$1-x$ = coefficient of inversion

$x = 0$ for normal spinel ferrite

$x = 1$ for inverse spinel ferrite

E.g. MnFe_2O_4 , CuFe_2O_4 , MgFe_2O_4

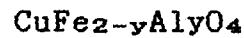
1.3.4 Substitution ferrites

In the substitutional ferrites divalent metal [M^{2+}] ions as well as trivalent metal [Fe^{3+}] ions can be replaced by other magnetic or non-magnetic ions. The substitution may take place either by replacement of M^{2+} ions by other divalent ions or by replacement of Fe^{3+} ions with other trivalent ions. The net magnetization in this ferrites also depends on the cation distribution.

E.g. In copper ferrite copper ion may be replaced by zinc partially or wholly



This is zinc substituted mixed ferrite. Trivalent iron ion may be replaced by another trivalent non-magnetic element generally. Such as aluminium or chromium



1.3.5 Magnetic interactions

In ferrites every oxygen ion is surrounded by three B cations and one A cation. The angles between A-O-A and B-O-B, A-O-B are [27] given below

$$\text{A-O-A} = 79^\circ 58'$$

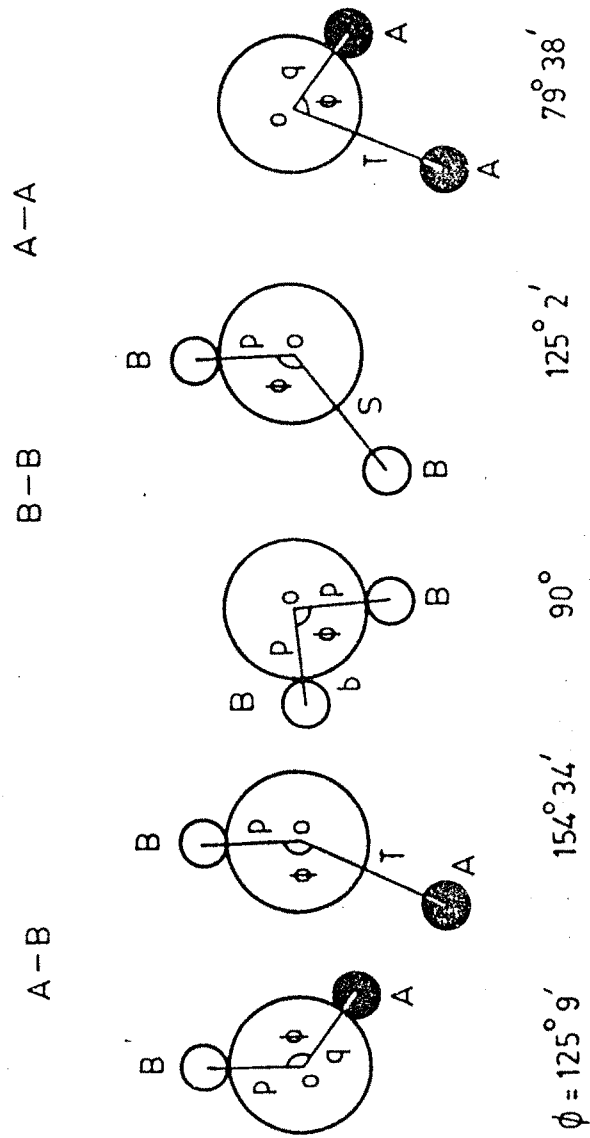
$$\text{B-O-B} = 90^\circ \text{ and}$$

$$\text{A-O-B} = 125^\circ 9'$$

The angle between A-A, A-B and B-B is shown in Fig. [1.4]. When the distance between M-O-M is small and the angle MOM is 180° [28] the interaction is expected to be strong. Thus we can conclude that B-B interaction is weak, the A-A interaction is weaker while the A-B interaction is strong.

1.4 Ferrimagnetism

The magnetic properties of the materials are due to free or unpaired spin in the atomic state. The material is paramagnetic or diamagnetic according as it possesses unpaired spins or paired spins. The ferromagnetism is due to permanently ordered spins in the same direction. In order to explain low magnetism Neel introduced two sublattice model in the spinel ferrites^{of} Ferrimagnetism. The ferrimagnetic material possesses weak permanent



ANGLE BETWEEN A-A B-B & A-B CATIONS IN A SPINEL STRUCTURE .

Fig. 1.4

magnetization due to their unequal antiparallel magnetic momenta. The mathematical theory of ferrimagnetism was developed by Neel [14] by considering two sub-lattice model. Yafet-Kittel [15] extended Neel's theory proposing triangular arrangement of spins.

1.5 Neel's Theory of Ferrimagnetism

The Knowledge of crystal structure and the nature of magnetic interaction in it helps us to account for the saturation magnetic moment.

Neel [14] [29] explained his theory of ferrimagnetism on the basis of two sublattice model. He assumed that the crystal lattice would be divided into two sub-lattices such as tetrahedral A-site and octahedral B-site in a spinel structure in which one type of magnetic ion of which fraction λ appeared on A-site and fraction μ appeared on B-site so that

$$\lambda + \mu = 1$$

Neel used the Weiss [2] molecular field theory for the actual magnetic field acting on an atom or ion which is written in the form

$$H = H_o + H_m$$

Where H_o is the external applied field and H_m is internal or molecular field. When the concept of molecular field

is applied to ferrimagnetic material on each lattice site we have

$$H_A = H_{AA} + H_{AB}$$

$$H_B = H_{BB} + H_{BA}$$

Here the molecular field H_A , acting on an ion on A-site is represented as the sum of the molecular field H_{AA} due to neighbouring A ions and H_{AB} due to its neighbouring on B-sites. A similar definition holds for the molecular field H_B , acting on a B-site ion.

The molecular field components may then be written as

$$H_{AA} = \lambda_{AA} M_A, \quad H_{AB} = \lambda_{AB} M_B$$

$$H_{BB} = \lambda_{BB} M_B, \quad H_{BA} = \lambda_{BA} M_A$$

Where the λ 's are the molecular field coefficients and M_A and M_B are the magnetic moments of the A and B sublattice. It can be shown that $\lambda_{AB} = \lambda_{BA}$ but $\lambda_{AA} \neq \lambda_{BB}$ unless the two sublattices are identical. Neel showed that $\lambda_{AB} < 0$ favours antiparallel arrangement of M_A and M_B resulting in the ferrimagnetism.

The total magnetic fields on each lattice site in the presence of an applied field H_0 can be written as

$$\begin{aligned} H_a &= H_0 + H_A \\ &= H_0 + \lambda_{AA} M_A + \lambda_{AB} M_B \end{aligned}$$

$$\begin{aligned} \text{and } H_b &= H_0 + H_B \\ &= H_0 + \lambda_{AB} M_B + \lambda_{BA} M_A \end{aligned}$$

I) Paramagnetic region

Ferrimagnetic substances at higher temperatures behave like paramagnetic substances.

Consider N free paramagnetic ions per unit volume each with total angular momentum quantum number J. Then the magnetization $M = N g J \mu_B B_J(x)$

Where N is number of free ions, g is Lande's splitting factor, J is total angular momentum quantum number, $B_J(x)$ is Brillouin function, μ_B the Bohr magneton.

The Brillouin function $B_J(x)$ is given by

$$B_J(x) = \frac{2J+1}{2J} \coth \left[\frac{(2J+1)x}{2J} \right] - \frac{1}{2J} \coth \left[\frac{x}{2J} \right]$$

Where $x = \mu_0 g J M_B H / kT$

k = Boltzman constant

T = absolute temperature.

Writing $\chi = M/H$ and $M = M_A + M_B$ and after necessary simplification, Neel expressed the equation of susceptibility as

$$\frac{1}{\chi} = \frac{T}{C} + \frac{1}{\chi_0} - \frac{\xi}{T-\theta} \dots 1$$

Where $\frac{1}{\chi_0} = \frac{1}{\chi_{AB}} [2 \lambda \mu - \lambda^2 \alpha - \mu^2 \beta]$

$$\theta = \frac{1}{\chi_{AB}} \lambda \mu c [2 + \alpha + \beta]$$

and $\xi = \frac{1}{\chi_{AB}^2} \lambda \mu c [\lambda(1+\alpha) - \mu(1+\beta)]^2$

The first two terms in equation (1) give Curie-Weiss form of temperature dependence of $1/\chi$ as observed in

ferromagnetics about Curie temperature but the term $\xi/T-\theta$ has no counterpart in ferromagnetism. This term is special to ferrimagnetic materials.

The plot of equation (1) i.e. $1/\chi$ versus T in $^{\circ}K$ represents the hyperbola as shown in Fig.(1.5).

II) Spontaneous magnetization

The spontaneous magnetization for ferrimagnetic substance is given by

$$M_{sp} = N g J \mu_B B_J(x)$$

$$\text{Where } x = \frac{\mu_0 g J \mu_B H}{kT}$$

For A sublattice, the spontaneous magnetization

$$M_{ASP} = \lambda N g J \mu_B B_J \left[\frac{\mu_0 g J \mu_B H_A}{kT} \right]$$

For B sublattice, the spontaneous magnetization

$$M_{BSP} = \mu N g J \mu_B B_J \left[\frac{\mu_0 g J \mu_B H_B}{kT} \right]$$

Where B_J 's represents Brillouin function and N represents number of atoms per unit volume of respective lattice.

The net observable spontaneous magnetization

$$M_{sp} = M_{BSP} - M_{ASP}$$

If the M_s values are plotted against T , different types of curves are obtained as shown in Fig. (1.6).

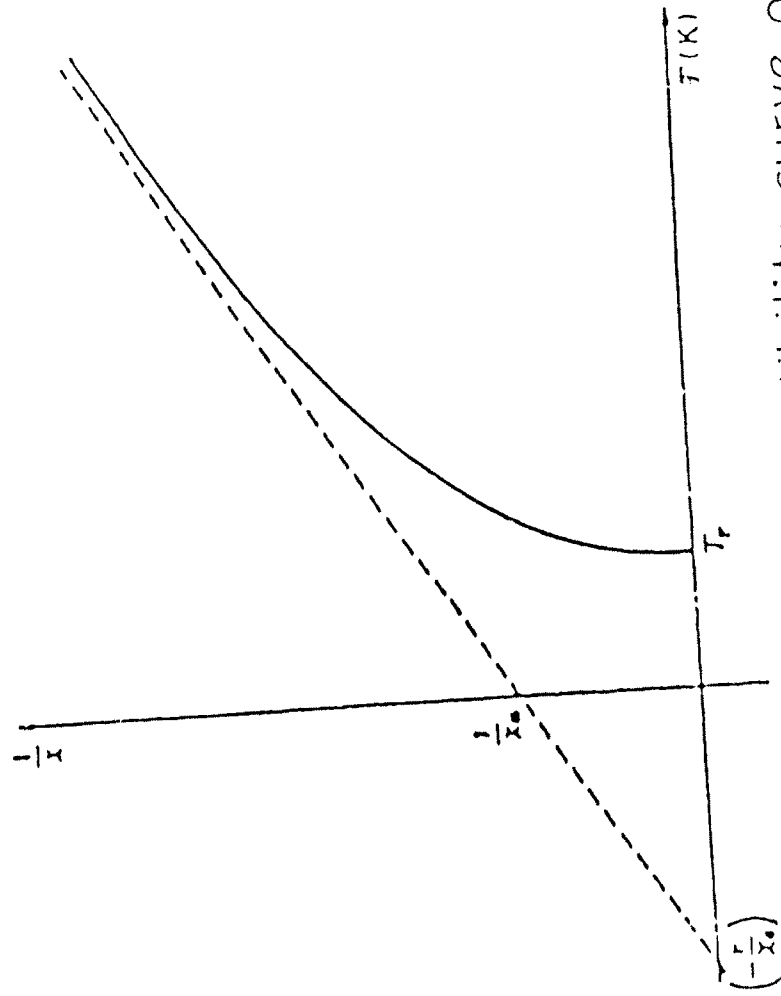


Fig. 1.5 -- Predicted susceptibility curve of a ferrimagnetic material above the Curie point.

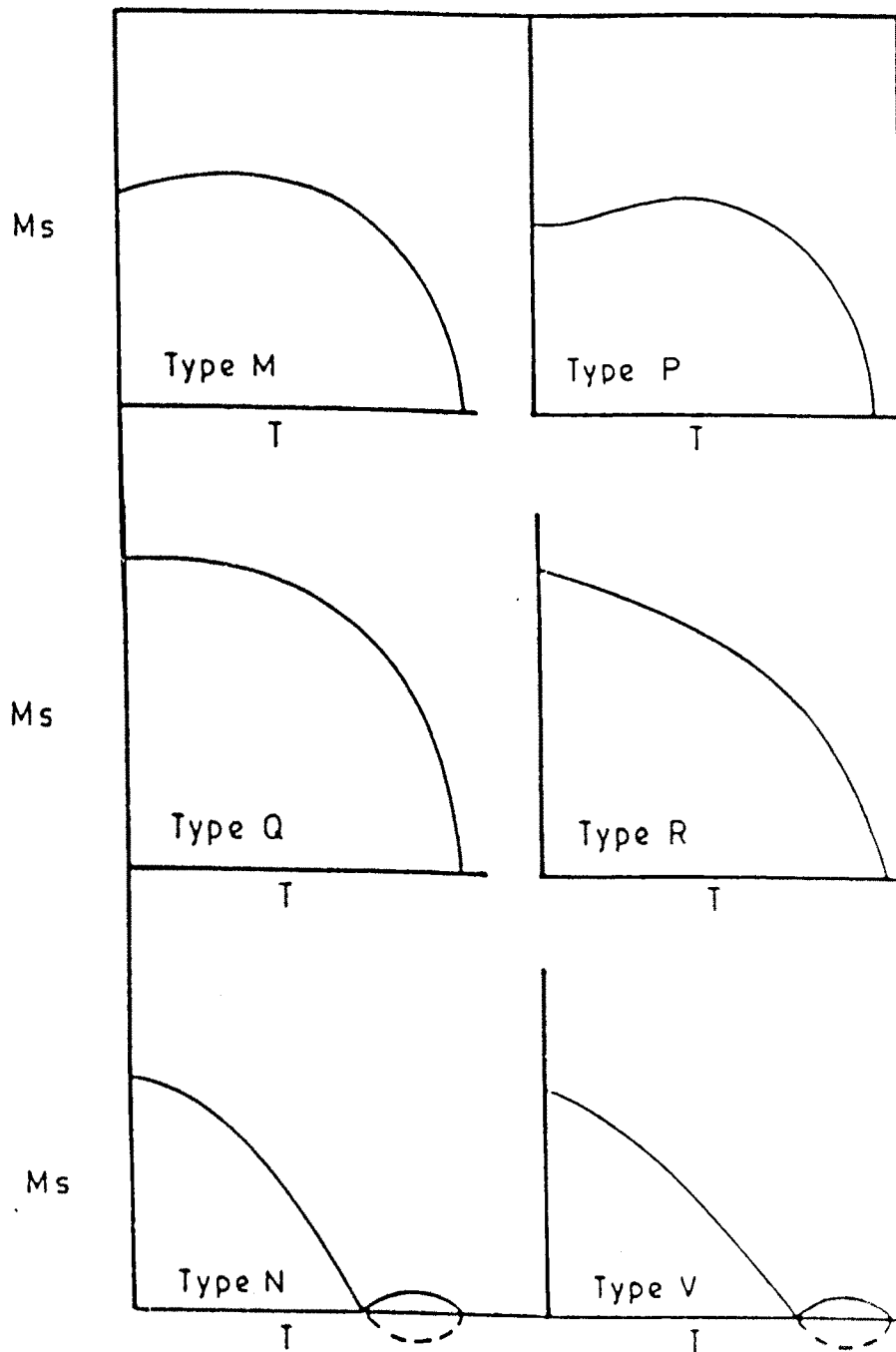


Fig. 1-6 - M_s vs T Curves for ferrimagnetic substances predicted by Neel's Theory.

These curves provide useful information about the ferrites from the application point of view.

1.6 Yafet-Kittel Theory

In 1952 Yafet-Kittel [15] suggested modifications in Neel's theory as it was observed that for the spinels containing metal ions other than Fe, the magnetization value is smaller than Neel's Model. They proposed triangular arrangement of moments by considering the possibility of subdivision of each sublattice of B as shown in the Fig (1.7). They showed that spins on sublattice B form canting angles with each other. On the basis of canting angles on B-site the magnetic properties of Cu-Zn, Ni-Zn, Fe-Zn, Cu-Co and many other systems have been explained successfully. Effect of presence of Yafet-Kittel angles on the magnetic properties of ferrites have been reported by Josyulu et al. Kulkarni and Paniker have calculated Yafet-Kittel angles in Cu-Cd ferrites. The important contribution of Kulkarni and Panikar on the studies of Cu-Cd ferrites is the formula suggested by them to calculate theoretical values of a canting angle given by

$$\cos \alpha_{YK} = \frac{5(1-x)^2\alpha + 25(1-x)^2\beta}{(1-x)^2\tau + 25(1+x)^2\delta + 10(1-x)^2\epsilon}$$

where α , β , τ , δ and ϵ are molecular field coefficients.

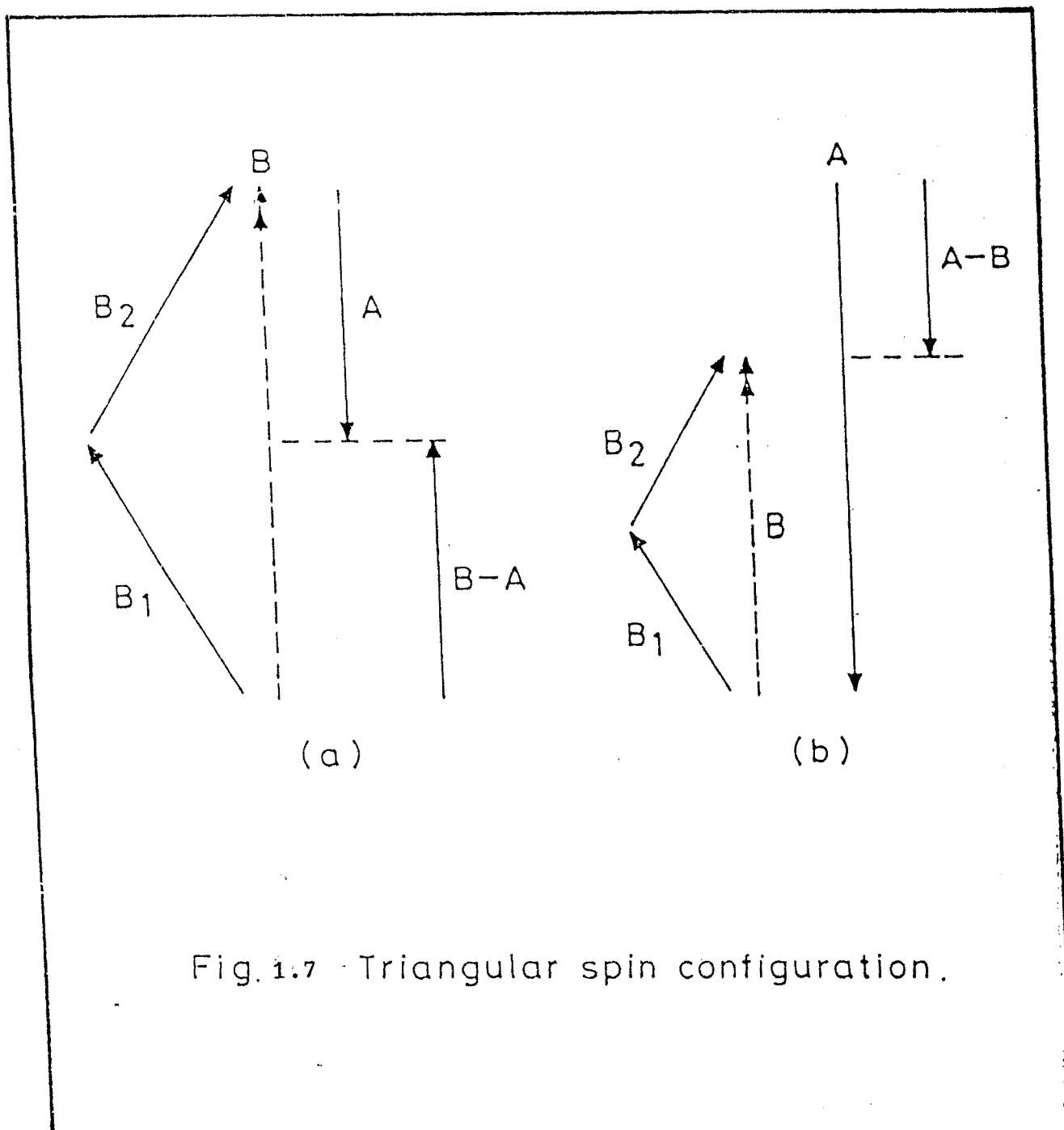


Fig. 1.7 - Triangular spin configuration.

When strong negative interactions exist within the sublattice B, it is divided into two equivalent substructures B₁ and B₂ with their magnetization neither exactly antiparallel to each other nor to A sublattice. Instead they align at some angle other than 180°. There is a net magnetization of the B lattice and which is resultant of B₁ and B₂ and, is antiparallel to the magnetization of A sublattice.

The interaction energy for this case is given by

$$E = 8N = 6 J_{ab} S_a S_b \cos \theta - J_{bb} S_b^2 (2 \cos \theta - 1)$$

where θ is an angle between a and subdivisions of B and J's are exchange integrals

Interaction energy E is minimum for J_{ab} negative and J_{bb} positive. $\theta = 0$ is the Neel state. If J_{bb} is also negative, the ratio of exchange energy is given by

$$\tau_{ex} = J_{bb} S_b / J_{ab} S_a > 3/4$$

under such conditions, Neel's state will not be minimum and magnetization of sublattice B₁ and B₂ will be inclined to sublattice A at an angle given by

$$\cos \theta = (3/4) (J_{ab} S_a / J_{bb} S_b)$$

Yafet and Kittel proved that for $\tau_{ex} < 3/4$ the Neel structure is stable if the total number of sublattices is restricted to six.

1.7 Properties of ferrites

The properties of ferrites are classified into two categories-

- i. Intrinsic properties
- ii. Extrinsic or structure sensitive properties

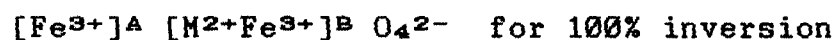
magnetization, saturation magnetization, anisotropy, Curie temperature and permeability are the intrinsic properties while hysteresis, dielectric constant, resistivity etc. are the structure sensitive properties.

The parameters such as porosity, grain size, impurities affect the structure sensitive properties.

1.7.1 Magnetic properties

1.7.1(a) Saturation magnetization

This is one of the intrinsic properties of ferrites and is governed by the chemical composition, cation distribution. The normal spinels are paramagnetic where as inverse spinels are magnetic at room temperature. The valence state of cations along with their distribution on two sublattice is written as



The Fe^{3+} ions on A-site are coupled with their spins antiparallel to those of Fe^{3+} ions on B-site. Therefore, the net magnetic moment is only due to divalent M^{2+} metallic.

1.7.1(b) Magnetostriction

Ferrites change their dimensions when they are magnetized. The small change in length is called longitudinal magnetostriction. This plays an important role in domain theory and in practical use of transformer materials. The incremental change may be positive or negative for different materials at saturation using substitutional ferrites, the magnetostrictive effect can be minimized [29].

1.7.1(c) Permeability

The permeability of the magnetic materials is due to the reversible displacement of domain walls within the material.

In general it is defined as the ratio of the magnetic flux density B in a substance to the external magnetic field strength H i.e.

$$\mu = B/H$$

It is generally found that the permeability increases with increase in temperature. Also pores, density, grain size of material are affecting the permeability.

1.7.1(d) Hysteresis

The lagging of magnetic induction to the magnetizing field is referred as magnetic hysteresis. The hysteresis

study on ferrites gives valuable data on saturation magnetization, remanance and coercive forces. Ferrites with high coercive force are called hard ferrites and those with low coercive force are called soft ferrites. Hard ferrites are used in permanent magnets. Whereas soft ferrites are applicable in a switching circuits. Hysteresis properties depend mainly on the chemical composition, crystal structure, cation distribution.

1.7.2 Electrical properties

1.7.2(a) D.C. Conductivity

Ferrites are semiconductors having wide range of resistivity. The ferrites show resistivities of the order of 10^{-3} to 10^{11} ohm-cm at room temperature.

The temperature dependence of resistivity obeys the law,

$$\rho = \rho_0 \text{Exp} (E_g/kT)$$

Where E_g = activation energy

k = Boltzmann constant

T = absolute temperature

The straight line graph of $\log \rho$ versus $1/T$ enable to determine the activation energy and observe ferrimagnetic Curie temperature. The activation energy ranges from 0.1 eV to 0.5 eV.

The chemical inhomogeneity, the grain size of individual crystallite, the porosity are the factors responsible for the resistivity.

1.7.2(b) Dielectric behaviour

A dielectric material when subjected to an alternating electric field, the positive and negative charges within the material get displaced with respect to one another and the system acquires an electric dipole moment. The dipole moment per unit volume is called as polarization.

Ferrites exhibit abnormally high dielectric constants and dispersion of dielectric constant in the frequency range from few Hertz to few MHz [30, 31]. The dielectric parameters of ferrites depend on the preparation technique grain size and porosity [32].

Koop's [22] has suggested that the dielectric constant is inversely proportional to the square root of conductivity.

1.7.2(c) Thermoelectric power

The thermo-electric properties are widely used in the interpretation of the conduction mechanism in semiconductors. However, in the case of low mobility materials, such as ferrites, thermoelectric power

measurement is the only alternative. The sign of thermo-emf gives vital information about the type of conduction in ferrites. Whether it is p-type or n-type. Another important significance of thermo-emf is that it enables one to calculate the values of fermi-energy and carrier concentration [33].

1.6 Applications

Ferrites have both high electrical resistivity and reasonably good magnetic permeability combined together in one. Therefore they find numerous applications in many fields. In addition to this they have switching property. Ferrites find wide applications in electronic products, microwave communication data processing devices, radar, television etc. Ferrites also find applications in MW antenna in radio receivers, mechanical filter, ultrasonic generators tape recorders etc. [34]. Some of the applications are given below.

1. Due to high resistivity, they find applications in audio devices such as recording and erasing heads in the tape recorders.
2. Ferrites having high permeability are used for band pass filters in telephone circuits as inductors in frequency range of 50 Hz to 150 KHz.

3. The iron core transformers produce eddy currents and hence ferrites are used in place of iron core, because ferrites do not produce eddy currents. These transformers are used in radio receivers due to their high resistivity and televisions.
4. Ferrites with square loop characteristics are more suitable for flip-flop changes in computers and memory devices.
5. Hexagonal ferrites being permanent magnetic materials are used in loud speakers.
6. Ferrites with large magnetostriction or lower permeability are used in accelerators, filter and ultrasonic generators.
7. Ferrites cores are widely used in the field of memory and switching circuits in computers.
8. The high uniaxial anisotropy as hard ferrites allow them to be used in loudspeakers, motors and generators, sticking devices and magneto therapy.

1.9 Orientation of the present work

The ferrites ferrimagnetic oxides have aroused great interest in last three decades from the point of view of their electrical and magnetic properties. These materials have a large potential from the point of application and research development.

These materials have high resistivity and low eddy current losses and also high permeability, hence these are widely used in industrial electronic, microwave and computer industries. They are useful for transformers, chokes, transformers cores, induction tuners, antenna rods, magnetic pole pieces of high frequency induction heaters. They are also used in T.V. revivers, telephone circuits, loudspeakers and in the field of memory and switching devices.

In spinel ferrites electric and magnetic properties are structure sensitive and can be influenced by substituting the cations and heat treatment. The mixed Cu-Co ferrite system has been studied by many workers from the point of view of crystal structure, d.c. electrical and magnetic properties, etc. However data on aluminium substituted Cu-Co ferrite is scarce in literature. Hence it will be interesting to study and compare the d.c. electrical and magnetic properties of the Cu-Co mixed ferrites with those which are substituted with aluminium. Therefore by substituting aluminium in Cu-Co ferrite system, the following studies have been carried out,

1. Preparation of aluminium substituted Cu-Co ferrite system viz.



Where $x = 0, 0.2, 0.4, 0.6, 0.8$ and 1

2. XRD studies to confirm the solid state reaction, determination of crystal structure and calculation of lattice parameters.
3. Spectroscopic investigation by I.R.
4. The d.c. electrical resistivities of the ferrite materials.
5. Magnetic properties like hysteresis and Curie temperature and a.c. susceptibility.

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