#### 1.1 INTRODUCTION :

Lode stone was first natural magnetic material known to man. Now a days ferrites are artificially prepared by mixing metal oxides and iron oxide. The ferrites are magnetic oxides containing iron oxide as their main component, regardless of their crystal structure.<sup>1</sup> Ferrites possess wide range of magnetization, shows high electrical resistivity which can be used to propagate high field electromagnetic waves with low attenuation.

Technological importance of ferrites can be gauged from the spectrum of applications in high frequency and pulse transformer, inductances, deflection coils, antenas, modulators and numerous other applications depending on the requirements of high permeability and low loss at high frequencies . Ferrites with square loop characteristics of B-H curve, find extensive use in computers, memory devices and flip-flops. Alternate selection and control of variables in preparation of ferrites has been used to give ferrites of lower permeability and larger magnetostriction. These are useful in accelerometers, mechanical filters and ultrasonic generators. Microwave applications depends on the nature of gyromagnetic resonance of ferrites which are exemplified in Faraday rotation, phase shifters, circulators and modulators.

#### 1.2 HISTORICAL :

The first known magnetic material was made available by nature itself, is magnetite. Hilpert<sup>2</sup> started the development of synthetic ferrites with an objective of reducing the eddy current losses in inductors and transformers. But ferrite prepared by him did not meet some of the above said properties and exhibited heavy losses, low permeability and could not be reproduced. In thirties Kato and Takai<sup>3</sup> Forestier<sup>4</sup> and Barth have studied ferrites from various point of view of its applicability and reliability.

Later on Snoek<sup>5,6</sup> carried out extensive and systematic studies of preparation of ferrites and their magnetic properties. This led to various applications of ferrites. Further Verway<sup>7</sup> showed that crystal structure of various ferrites with inverted spinel structure to be ferrimagnetic whereas those with normal structure to be nonmagnetic.

The basic theory of spin-spin interaction in ferrites was announced by Neel<sup>8</sup> and introduced the idea of magnetic sublattice. A more detailed examination of basic interaction was made by Anderson<sup>9</sup> and Van-Vleck<sup>10</sup> who developed the theory of superexchange. Yafet and Kittle<sup>11</sup> extended the theory of magnetic sub-lattices by postulating a 'triangular' arrangement of three sub-lattices, when antiferromagnetic exchange interactions between sub-lattices is comparable to that among spin moments within sub-lattices, formed by Fe<sup>3+</sup> ions on tetrahedral and octahedral sites. The direct proof of Neel's model

of ferromagnetism came up only with neutron diffraction studies on magnetite  $^{11}$  and zinc ferrite.  $^{12}$ 

Recently the work on ferrites has assumed large proportions during the last two decades. Now a days researchers working in different areas of science and technology are busy in manufacturing, synthesising and improving the ferrite properties in view of their tremendous demand in the field of microwave components, logic and memory circuits in modern digital computers. The substitutional types of ferrites are more useful in the field computer and high power applications.

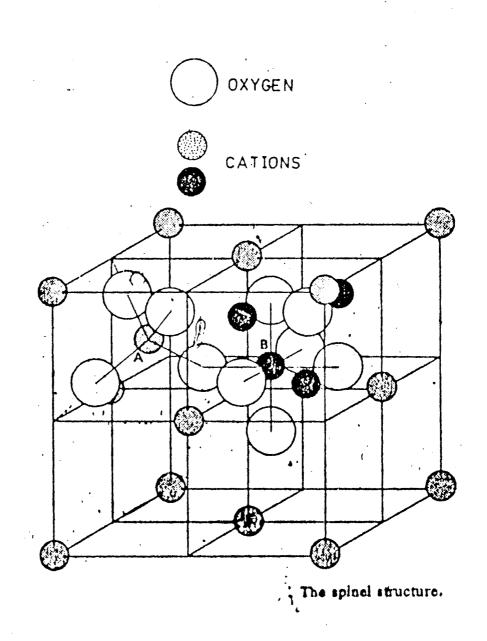
#### 1.3 CRYSTAL STRUCTURE :

Generally magnetic oxides show four types of crystal structure:

- (a) Spinel structure <sup>t</sup> (b) Hexagonal structure
- (c) Garnet structure (d) Perovskite structure.

## 1.3(a) <u>Spinel Structure</u> :

The ferrites exhibit the spinel structure, similar to the crystal structure of the mineral spinel  $MgAl_2O_4$ . Their chemical composition can be written in general form as  $M^{2+}Fe_2^{-3+}O_4^{-2-}$  where  $M^{2+}$  represents a divalent metal ion, such as Mg, Mn, Fe, Co, Cu, Zn or combination of these ions, such as  $(Li_{0.5}Fe_{0.5})$ . The  $Fe^{3+}$  ions are sometimes partially replaced by other trivalent ions such as  $Al^{3+}$  or  $Cr^{3+}$ . The spinel structure can be described in terms of cubic close packing of the larger oxygen ions with the smaller metal ions situated in the interstices. These interstices are of two types, the



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Fig. 1.1

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tetrahdral or A-site with four surrounding oxygen ions and the octahedral or B-site with six surrounding oxygen ions. In the unit cell of spinel which contains 32 oxygen ions, eight of tetrahedral sites and sixteen of the octahedral sites are occupied by metal ions. If divalent ions occupy 'A' sites the structure is called normal spinel structure and if they are found on B-sites the structure is called inverse spinel. Writing the ions on 'B' sites between the square bracket  $M^{2+}[Fe^{3+}]0^{2-}$  is a normal spinel and  $Fe^{3+}[M^{2+}Fe^{3+}]0^{2-}$  is an inverse spinel. It has been found that  $ZnFe_20_4$ ,  $CdFe_20_4$  are normal or nearly so and the other ferrites are inverse.

## 1.3(b) : Garnet Structure :

The garnet minerals existing in nature include quite a number of compounds characterized in general terms by the formula,

$$M_3^{2+}M_2^{3+}Si_3^{4+}O_{12}$$

where  $M^{2+}$  is Ca, Fe, Mg, or Mn and  $M^{3+}$  is Al, Cr, Fe or Mn. The natural garnet is grasslarite having formula  $Ca_3Al_2(SiO_4)_3$ . The most widely studied garnet is YIG or Yittrium-Iron-Garnet. Garnet structure was fully explained by Bertaut, Gellevand Giello.

## 1.3(c) <u>Hexagonal Structure</u> :

The hexagonal ferrites were discovered and investigated at the Phillips Research Laboratories.<sup>1</sup> The hexagonal structure has the chemical formula  $MFe_{12}O_{19}$  where 'M' can be Ba, Sr, Ca or Fe. The magnetoplumbite compounds also have the hexagonal structure and their 1.

chemical formula is x BaO :  $y \operatorname{Fe}_2 O_3' z M^{2+}O$  where 'M' is transition metal. They are generally known permanent magnetic materials.

#### 1.4(A) MAGNETIZATION IN FERRITES :

The ferrites are ferrimagnetic materials which show indirect exchange interaction. The intervening ions like oxygen give rise to the two sub-lattices, characterised by different oxygen environments for the metal atoms. Magnetic interactions within the sub-lattice give rise to a sub-lattice magnetizations. Of the two sub-lattice magnetization at a temperature are not equal, although they are opposite the net magnetization results as in the case of ferromagnetic materials. Therefore, the ferrites exhibit all the properties similar to those of ferromagnetic materials.

The properties of ferrites can be classified in two categories, intrinsic and structure sensitive. Saturation magnetization, anisotropy, magnetostriction and Curie temperature are basically the intrinsic properties, while permeability, hysteresis, resistivity and dielectric constant etc. are very much structure sensitive. The structural aspects like grain size, porosity, impurities and inclusions of non-magnetic ions, size distribution of inclusion affect the structure sensitive properties.<sup>13</sup>

The two sub-lattice model is used to explain the magnetization process in spinel ferrites.

The normal spinels are non-magnetic while inverse spinels are magnetic. However, for mixed ferrite system on a simplistic approach the observed magnetization could not be explained. For the ferrite with inverse spinel structure formula is,

$$(Fe^{3+})_{A} [M^{2+}Fe^{3+}]_{B}^{0}_{4}$$

The Fe<sup>3+</sup> ions on A-site are coupled with their spins antiparallel to those of Fe<sup>3+</sup> ions on B-site, so that net moment is only due to divalent  $M^{2+}$  metallic ions.

$$(Fe_A 5\mu_B) (Fe_B 5\mu_B M_B) \dots (1.1)$$

Suppose that 'M' is transition element with n-electrons in d-shell. The magnetic moment per unit formula is  $(n.\mu_B)$  or  $(10-n)\mu_B$  depending on d-shell which is filled less than half or more than half respectively. The degree of inversion is a fraction 'x' of the divalent metal ions that are on B-site. The arrangement of moments could be written as,

$$[(1-x)M_A \times Fe_A] [(1-x)Fe_B \times M_B] \dots (1.2)$$

The net moment  $_{\rm R}$  is weritten as,

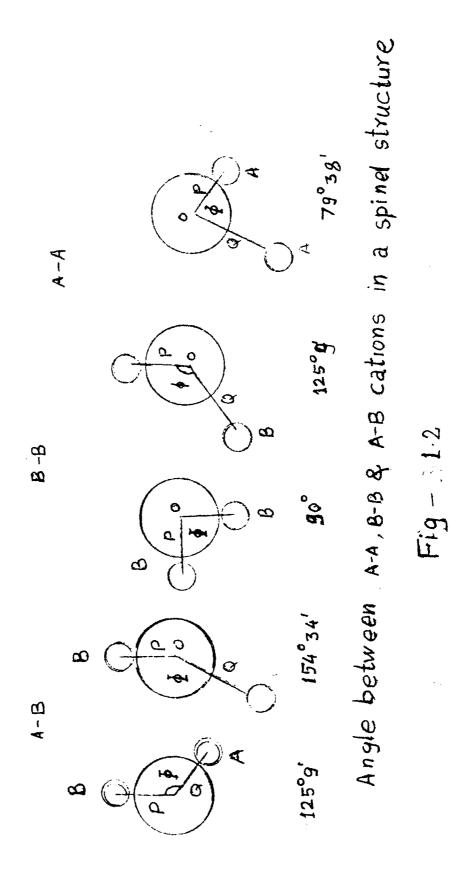
$$B = M[(1-x)-x] - S [1+(1-x) - x] \qquad \dots (1.3)$$

$$= M (1-2x) - 10 (1-x) \qquad \dots (1.4)$$

For normal spinels x = 0

inverse spinels x = 1

The indirect exchange coupling between metallic ions acting through the oxygen ions is maximum. For those ions A and B for which the angle AOB is close to 180°. For the perfect spinel lattice the major angles between the ions are AOB, 125°9' and 154°34'; BOB 90° and 125°9' and AOA 79°34' (Fig. 1.2). The nearest to perfect align ment are those  $Fe_A$  and  $Fe_B$  ions making 154°34' thus giving the strong antiferromagnetic coupling.<sup>14</sup>



1.4 (B) ANISOTROPY :

Anisotropy term is used to describe directionality of magnetization along certain crystallographic direction which is called as easy direction. It is also known as anisotropy energy which can be applied to change the direction of magnetization from easy direction to hard direction. It plays an important role in determining the properties like permeability, hysteresis and magnetostriction.<sup>15</sup>

For the bulk magnetization anisotropy energy is given by,

$$E_k = K_1 \sin^2 \theta$$

This expression applies to a crystal with uniaxial symmetry. For cubic materials the energy is described as,

$$E_{k} = K_{1}(\alpha_{1}^{2}\alpha_{2}^{2} + \alpha_{1}^{2}\alpha_{3}^{2} + \alpha_{2}^{2}\alpha_{3}^{2}) + K_{2}(\alpha_{1}^{2}\alpha_{2}^{2}\alpha_{3}^{2}) \dots (1.5)$$

where  $K_1$  and  $K_2$  are called anisotropy constants and which vary from material to material and are temperature dependent and  $\alpha_{1,\alpha_{2,\alpha_{3}}}$  $\alpha_{3}$  are direction cosines. When magnetization vector deviates from preferred direction the anisotropy energy increases. This increase in energy is due to the effective field (Ha) acting on spins. The anisotropy constant varies with crystal structure and kind of magnetic ions involved. The electronic structure of magnetic ions also contributes to anisotropy constant.<sup>16</sup> Magnetic anisotropy may also arise due to stress anisotropy and shape anisotropy. Stress anisotropy may be reduced by slow cooling of specimen. Shape anisotropy may be reduced by preparing ferrite in such a way as voids are spherical and few in number with the help of prolonged sintering.<sup>17</sup>

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### 1.5 (A) NEEL'S THEORY FOR FERRIMAGNETISM :

Neel defined the interactions within the material from the Weiss molecular field viewpoint. The magnetic field acting upon an atom or ion is written in the form,

$$H = H_0 + H_m$$
 ... (1.6)

where  $H_{o}$  is the externally applied field and  $H_{m}$  is the internal or molecular field which arises due to interactions with other atoms or ions within the material. Thus when molecular field concept is applied to ferrimagnetic material we have,

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

$$H_{B} = H_{BB} + H_{BA}$$
(1.7)

where  $H_A$  is molecular field acting on an ion on the A-site and is sum of molecular fields  $H_{AA}$  from neighbouring 'A' atoms and  $H_{AB}$  from 'B' atoms.

Molecular field components may be written,

$$H_{AA} = \gamma_{AA} M_A \qquad H_{AB} = \gamma_{AB} M_B \qquad \dots (1.8)$$
$$H_{BB} = \gamma_{BB} M_B \qquad H_{BA} = \gamma_{BA} M_A$$

where  $\gamma$ 's are the appropriate molecular field coefficients and  $M^{}_{A}$  and  $M^{}_{B}$  are the magnetic moments of the A and B sub-lattices.

It may be shown that,

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$$\begin{split} \gamma_{AB} &= \gamma_{BA} \quad \text{but} \quad \gamma_{AA} \neq \gamma_{BB} \quad \text{unless the two sublattices} \\ \text{are identical. Neel showed that} \quad \gamma_{AB} < 0 \quad \text{favouring antiparallel} \\ \text{arrangements of } \text{M}_{A} \text{ and } \text{M}_{B} \text{ gives rise to ferrimagnetism.} \end{split}$$

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In the presence of an applied field  $H_0$  total magnetic field acting on each sublattice may be written by using equation (1.8),

$$H_{A} = H_{o} + H_{AA}$$

$$H_{A} = H_{o} + AA M_{A} + AB M_{B}$$

$$\dots (1.9)$$

$$H_{B} = H_{o} + H_{BB}$$

$$= H_{o} + BB M_{B} + BA M_{A}$$

# a) Paramagnetic region :

and

In an assembly of N free paramagnetic ions per unit volume each with angular momentum quantum number J the magnetization is given by,

$$M = \mu_{o} Ng^{2} \mu_{B}^{2} J(J+1) H / 3K_{B}T$$

and the volume susceptibility, defined  $\chi = M/H$  is

$$X = M_0 Ng^2 \frac{2}{B} J(J+1) / 3 kT = C/T \qquad ... (1.10)$$
  
$$\mu_0 Ng^2 \frac{2}{B} J(J+1) / 3 k \text{ is Curie constant.}$$

At high temperature if the magnetic field in the Curie law is taken to be (  $\rm H$  +  $\rm H_{m}$  ) then we have,

 $\frac{M}{(H+H_m)} = \frac{M}{(H+m^M)} = \frac{C}{T}$ 

or

where C =

$$= \frac{C}{(T-T_c)} \qquad \dots (1.11)$$

where  $T_c = \gamma C$  which leads to Curie-Weiss law,

Neel supposed the existance in the material of one type of magnetic ion only, of which a fraction appeared on A-sites and a fraction on B-sites. Thus,

$$\lambda + \mu = 1 \qquad \dots (1.12)$$

Then we can write,

$$M_A = \left(\frac{\lambda_C}{T}\right) H_A$$
 and  $M_B = \left(\frac{\mu_C}{T}\right) H_B$  ... (1.13)

Taking  $M = M_A + M_B$  it follows after the necessary algebraic manipulation that,

$$\frac{1}{\chi} = \frac{T^2 - C\gamma_{AB} (\lambda \alpha + \mu \beta) T + C^2 \lambda \mu \gamma_{AB}^{2} (\alpha \beta - 1)}{C \{ T - \lambda \mu C \gamma_{AB} (2 + \alpha + \beta) \}} \dots (1.14)$$

where

$$\alpha = (\gamma_{AA} / \gamma_{AB}) \text{ and } \beta = (\gamma_{BB} / \gamma_{AB})$$

For clear explanation we rewrite,

$$\frac{1}{\chi} = \frac{T}{C} + \frac{1}{\chi} - \frac{r}{T-\theta}$$
 (1.15)

where,

$$\frac{1}{\chi_{0}} = \Upsilon_{AB} \left( 2\lambda\mu_{-} \lambda^{2}\alpha_{-} \mu^{2}\beta_{-} \right)$$
  

$$\theta = \Upsilon_{AB} \lambda\mu C \left( 2+\alpha+\beta_{-} \right) \qquad \dots (1.16)$$
  

$$\xi = \Upsilon_{AB}^{2} \lambda\mu C \left\{ \lambda (1+\alpha) -\mu_{-} (1+\beta_{-}) \right\}^{2}$$

The first two term on right hand side give Curie-Weiss form of temperature dependance of  $1/\chi$ . Equation (1.15) represents hyper-bolla. Now well above Curie temperature ferrimagnetic interactions plays little part and the hyperbolla is asymptotic to the line,

$$\frac{1}{\chi} = \frac{T}{C} + \frac{1}{x_0} \qquad \dots (1.16)$$

which cuts the temperature exists at  $T^{}_{\rm A}$  = - C/ $^{}_{\chi_{\rm O}}$  Neel called this the asymptotic Curie point.

Paramagnetic Curie point  $\mathbb{T}_p$  is found by equating 1/x to zero which gives

$$T_{p} = \frac{\gamma_{AB}}{2} \left[ \lambda \alpha + \mu \beta + \{ (\lambda \alpha - \mu \beta)^{2} + 4\lambda \mu \}^{\frac{1}{2}} \right] \dots (1.17)$$

# (b) <u>Spontaneous Magnetization</u>:

Equation (1.17) can be weritten as,

$$T_{p} = \frac{1}{2} \gamma_{AB} C \left[ \lambda \alpha + \mu \beta + (\lambda \alpha - \mu \beta)^{2} + 4\lambda \mu \right]^{\frac{1}{2}} \qquad \dots (1.18)$$

$$T_{p} = \frac{1\gamma}{2} AB^{C} [\lambda \alpha + \mu \beta - \{(\lambda \alpha - \mu \beta)^{2} + 4\lambda \mu\}^{\frac{1}{2}}] \dots (1.19)$$

assuming positive square root is taken each time. Full analysis shows that  $T_p$  is the Curie temperature when  $M_A$  and  $M_B$  antiparallel and  $T_p$  Curie temperature when  $M_A$  and  $M_B$  parallel.

If  $T_p > T_p$  the antiparallel arrangement of magnetization will occure and be stable at all temperatures below  $T_p$ . This is in agreement with negative interaction of A and B sublattices.

The individual magnetizations  $M_A$  and  $M_B$  cannot be observed independently but net magnetization 'M' can be written as,

$$M = M_A + M_B$$

but as  ${\rm M}_{A}$  and  ${\rm M}_{B}^{}$  are oppositely directed then,

$$\mathbf{M} = |\mathbf{M}_{\mathbf{A}}| - |\mathbf{M}_{\mathbf{B}}| \text{ or } |\mathbf{M}_{\mathbf{B}}| - |\mathbf{M}_{\mathbf{A}}|$$

Spontantous magnetization can be written assuming the similar behaviour below and above Curie point,

$$M_{Asp} = Ng J \mu_B B_j (\mu_0 g J \mu_B H_A / KT) ... (1.20)$$

$$M_{Bsp} = \mu Ng J \mu_B B_J (\mu_0 g J \mu_B H_B / KT) \qquad \dots (1.21)$$

$$\frac{M_{Asp}}{M_{Asat}} = B_J (\mu_0 g \mu_B J H_A / KT) \qquad \dots (1.22)$$

$$\frac{M_{Bsp}}{M_{Bsat}} = B_J (\mu_0 g J \mu_B H_B / KT) \qquad \dots (1.23)$$

In general  $J \simeq S$  the spin quantum number since the orbital contribution is small.

Total observed spontan@ous magnetization is given by,

$$M_{sp} = M_{Bsp} - M_{Asp} \qquad \dots (1.24)$$

The solutions of  $M_{sp}$  with variation of temperatures cannot be found analytically. For solving these graphical methods<sup>18</sup> are used and known as ( $\sigma$ ,  $\tau$ ) curves where,

$$\sigma = \frac{M_{sp}}{M_{sat}} \quad \text{and} \quad \tau = \frac{T}{T_p} \qquad \dots (1.25)$$

 $(\sigma, \tau)$  curves must depend on relative values of interactions and relative magnitudes of sub-lattice magnetizations, must depend on  $\alpha, \beta, \lambda$  and  $\mu$  in simple Neel's model, where only one type of magnetic ion is present.

## 1.5 (B) <u>YAFET-KITTLE THEORY</u> :

or

Neel's model was inadequate for the spinels containing chief constituents other than 'Fe' where the observed magnetization is much smaller than calculated by Neel's model for spin arrangement. Therefore Yafet and Kittle<sup>19</sup> have proposed a triangular type of spin arrangement.

The interaction energy E between two atoms having spins  ${\rm S}_{\rm i}$  and  ${\rm S}_{\rm i}$  given by Heisenburg as

$$E = 2 JeSiSj$$
 ... (1.26)

where 'Je' is exchange integral measure of extent to which the electronic charge of the two atoms concerned overlap one another.

They showed that for certain ratios of exchange interaction in spinels there can exist three sublattices with triangular spin configuration and has lower energy than two sub-lattice structure. In such a case there exists a possibility of negative interaction within the sub-lattice itself giving rise  $B_1$ ,  $B_2$  sub-lattices which are neither exactly antiparallel to each other nor to A sublattice individual. However the resultant magnetization of  $B_1$  and  $B_2$  remain antiparallel with that of A sublattice, holding in store all the possibilities of ferrimagnetism and antiferromagnetism.

The interaction energy E for this case has been given as,

$$E = 8N[6J_{ab}S_{a}S_{b} \cos \phi - J_{bb}S_{b}^{2}(2 \cos \phi - 1)] \qquad \dots (1.27)$$

 $\phi$  is angle between magnetization of two sublattices. E is minimum for J<sub>ab</sub> negative and J<sub>bb</sub> positive if  $\phi = 0$  is Neel state. If J<sub>bb</sub> is also negative and ratio of exchange energies is given by ' $\gamma_{ex}$ ' as,

$$\gamma_{ex} = J_{bb} S_b / J_{ab} S_a > \frac{3}{4} \qquad \dots (1.28)$$

Then Neels state will not become minimum and magnetization vector of sublattice will bent from colinear position by an angle where,

$$\cos \phi = 3 J_{ab} S_a / 4 J_{ab} S_b \dots (1.29)$$

They proved that Neels structure is stable for  $\gamma_{ex} < 3/4$  if the total number of sublattices are restricted to six (i.e. number of different magnetic ion).

## 1.6 <u>ELECTRICAL PROPERTIES OF FERRITES</u> :

Ferrites are semiconductors by nature their resistance can vary from  $10^{-3} \Omega$  cm to  $10^{11} \Omega$  cm at room temperature.<sup>20</sup> The physical and chemical properties of ferrites are dependent on the mechanism of charge transport. The mechanism of charge transport can be studied or derived from the measurement of electrical conductivity, thermoelectric power, magneto resistance and Hall co-efficient. The charge transport can vary with the composition of ferrites and method of preparation of ferrites and also greatly affected by the presence of impurities.

# 1.6 (a) <u>Conductivity</u> :

Ferrites are semiconductors and their resistivity decreases with increase in temperature according to relation,

$$\rho = \rho_{o} \exp (E_{g} / K T)$$
 ... (1.30)

where  $E_g$  is energy required to cause an electron jump. If a graph of log  $\rho$  vs 1/T is plotted, then it is straight line and break

occurs at a temperature which corresponds closely with observed Curie temperature.<sup>21</sup> The value of  $E_g$  results between 0.1 eV to 0.5 eV. The high ' $E_g$ ' is associated with high resistivity at room temperature. In general the substitution of a cation that tends to stay in lower valance state leads to p-type conduction and substitution of cation that tends to stay in higher valance state leads to n-type conduction

Electronic conduction of ferrites can be associated with the presence of ions of given element in more than one valance states distributed randomly over crystallographically equivalent lattice points. In ferrites conductivity is attributed to the occurrance of both  $Fe^{2+}$  and  $Fe^{3+}$  ions on identical lattice sites in spinel structure. If this is the situation the electron can move from trivalent iron ion to divalent iron ion within octahedral positions without causing the change in the energy state of a crystal as a result of transitions. The classic example is  $Fe^{2+}Fe_2^{3+}O_4$ , one of the non-metallic conductors.<sup>22</sup> The presence of  $Fe^{2+}$  ion will be considered to assure reduced resistivity in ferrite system. To achieve high resistivity it is common practice to avoid an excess of  $Fe_2O_3$  and oxidising atmosphere during sintering.

The resistivity will also be caused by following factors apart from the inherent properties of the material (1) porosity and porosity filled by air (2) The grain size of individual crystallite influence the conduction path due to number of grain to grain contact<sup>18</sup> (3) Chemical inhomogeneity caused by preparation or heat treatment.

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General methods for determination of resistivity is two probe contact method or four probe contact method. The four probe contact method is always free from contact resistance difficulties and is always used to measure low resistivity materials.

# 1.6 (b) <u>Dielectric Properties</u> :

Ferrite is also an insulator, but eventhough in a ferrite eddycurrents may flow due to dielectric displacement. In ferrite three principle mechanism of polarisation can be distinguished within a frequency range extending from zero frequency to U.V. frequency region. At optical frequencies the polarisation mechanism is electronic. Total electronic polarisation is sum of polarisation of individual electrons plus effect of interactions. At higher optical frequency and at U.V. frequency, absorption due to actual electron transitions become important. The absorption caused by conduction electrons migrating between iron ions on the octahedral sites by transition from 3d to 4s states and by transition involving the oxygen anions and various cations. At frequencies below infrared the mechanism is atomic or ionic polarisation, and arises from the movement of cations anions within the crystal lattice. The part of permitivity at microwave frequency is due to this mechanism. Below the microwave frequency region the polarisation arises from the macro structure of polycrystalline material and from the migration of free charge carriers. These charge carriers are trapped at the grain boundaries. Field distortion results due to build up of space charge and shows an increase in permitivity This phenomenon is known as

interfacial polarisation. The ferrite have permittivity between 10 to 20. Abnormally high dielectric constant have been reported by several authors as a resdult of low frequency measurement of ferrites.

 $Koops^{25}$  described good phenomenological theory of dielectric dispersion in ferrites at low frequencies. The dielectric dispersion theory is based on the assumption of parallel resistance and capacitance of material results from an equivalent circuit and which is equivalent to solid state model consisting of compact of relatively good conducting grains separated by poor conducting layers or grains. The theoretical and observed frequency variation dielectric constant and resistivity are in good agreement for some samples. Activation energy obtained by Kamiyoshi<sup>23</sup> and Fairweather<sup>24</sup> from temperature variation with relaxation time agree well with those obtained with temperature variation of resistivity. The high dielectric constant may arise at low frequency due to oxygen ion which have three B-site and one A-site neighbours, or in the case of A-site ion through a distortion of its surroundings of oxygen tetrahedra. This is justified in view of the fact that oxygen possesses the maximum polarisability.

### 1.7 ORIENTATION OF PROBLEM

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Since 1950 magnetic oxides specially ferrites arose considerable interest because of their high electrical and magnetic values They are widely used in electrical, electronic, microwave and computer applications. Being technological important they have been

extensively studied from point of view of determination of their magnetic and electrical properties to check the suitability for certain applications. Basically they are synthesized from iron-oxide and some divalent metal oxides. Their physical properties can be varied by changing the composition, heat treatment and some additives Recently the bulk properties have shown interesting modulation on addition of materials like Ti, V.... etc.<sup>26</sup> The influence of grain size, porosity, orientation of crystallites densification and grain growth which gives useful magnetic properties of practical importance

Especially eddy-current losses prevent the use of magnetic metals at high frequencies and are usually not importance in ferrites It is possible to reduce the corresponding eddy-current losses by giving grain boundaries a high resistance. The overall resistance of the material is thereby increased whereas the ferrous ions inside the grains are still useful for magnetic properties.<sup>27</sup> There are two methods of obtaining an insulated layer on grain boundaries viz. reorientation and the formation of non-magnetic second phase. The forma tion of non-magnetic second phase method uses the addition of small amount of CaO, SiO<sub>2</sub>, TiO<sub>2</sub>, GeO<sub>2</sub>, etc.<sup>28,29</sup> When these oxides are added in right proportion a markedly increased resistivity is observed and corresponding a decrease of eddy-current loss.

Mg-ferrite is sensitive to heat treatment. Secondly it has high resistance, whereas mixed ferrites of 'Mg' are used in microwave applications.<sup>30</sup> Recently, substitution of Ti, Zr with Ni-Zn and Mg-Zn mixed ferrites have been studied with their electrical and magnetic

properties by many workers.<sup>31,32</sup> Therefore in order to understand the physical properties like resistivity, magnetization in Mg-ferrite by substituting  $\text{TiO}_2$  and  $\text{ZrO}_2$  have undertaken. The following studies will be carried out.

- i) Preparation of  $Mg_{1+t}Ti_tFe_{2-2t}O_4$ (t = 0, 0.1, 0.2, 0.3, 0.4, 0.5) by ceramic method at an appropriate temperature.
- X-ray diffraction studies of these samples for finding Bravias lattices, lattice parameter and impurity phases.
- iii) D.C. electrical conductivity, thermo e.m.f. measurement studies for understanding the conduction mechanism and effect of substitution on resistivity.
- iv) A.C. conductivity measurement for the studies of dielectric losses and utility of high frequency applications.
- v) Magnetization studies and Curie temperature measurement on these ferrites for finding out the cation distribution to under stand the role of substitution.

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