Chapter 3. Microwave Theory and Background

In conventional thermal demagnetisation heat is applied to a sample which creates lattice vibrations (phonons). These phonons are in a higher energy state than the surrounding magnetic system so they exchange energy with the magnetic system, and spin waves (magnons) are created (Walton *et al.*, 1992; 1993). The generation of spin waves within the magnetic grains enables the individual domain magnetisations to reverse and thus demagnetise in zero field (Walton, 1986) or to realign with an ambient fixed field to produce a TRM. In microwave demagnetisation / remagnetisation, the first steps are bypassed; magnons are directly excited with the use of high frequency microwaves thus eliminating the need to heat the bulk sample. Some heating of the bulk sample does occur due to the generation of phonons in the relaxation process but to a much lesser extent than in conventional heating.

This chapter provides the basic theory and background to the process of microwave excitation and the different ways the microwave energy is lost to the system. This is described for non magnetic and magnetic material before concentrating on the process of ferromagnetic resonance, FMR with which the Liverpool microwave system operates. Microwaves are used in industry but for heating rather than demagnetisation, this is also discussed.

3.1. Introduction

Microwaves are electromagnetic waves that have a frequency range from around 0.3 GHz (there is no actual specified lower frequency limit) to 300 GHz with corresponding wavelengths ranging from 1m to 1mm. Microwaves are coherent and polarised in contrast to visible waves (apart from lasers). They obey the laws of optics and can be transmitted, absorbed or reflected depending on the type of material, as illustrated in Fig. 3.1.

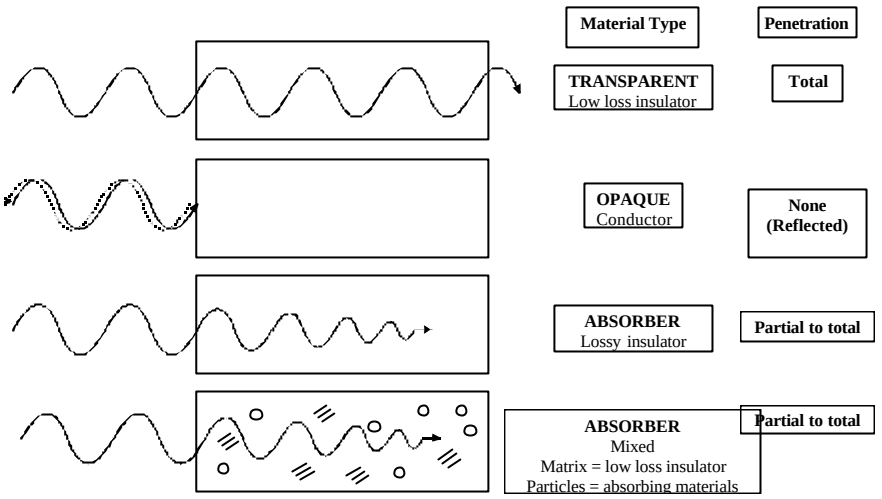

Figure 3.1 The interaction of microwaves with different materials (after Sutton, 1989).

Everyone is familiar with the domestic microwave oven; indeed the majority of households contain one. The microwave heating process is however, fundamentally different from the heating process used in conventional ovens. With microwaves, heat is generated internally within the material as opposed to originating from external heating sources. As a result, the thermal gradients and flow of heat is the reverse of those in materials heated by conventional means. A conventionally cooked Baked Alaska has the ice cream on the inside whereas a microwave cooked one has the ice cream on the outside! It is possible to heat both large and small shapes very rapidly and uniformly and as the absorption of microwave energy varies with composition and structure it is also possible to have selective heating.

Selective heating is desirable for palaeomagnetic purposes as the magnetic constituents can be specifically targeted. In fact it is possible to go one better by using ferromagnetic resonance (FMR) to demagnetise directly the magnetic particles with the microwave energy, before the energy is transferred to the lattice as heat. This leads to reduced heating of the bulk matrix of the sample and hence less alteration during experiments. As far as the magnetic particles are concerned, however, microwave heating is exactly the same as with conventional heat. In the demagnetisation process, the spin system of the magnetic grains is excited (Walton, 1986, Section 1.1.3) both with microwaves and conventional heat. It is solely the method of getting the energy to the spin system that differs between the

two processes. In the Liverpool microwave technique the mechanism of FMR is used.

There are different mechanisms by which microwaves (and lower frequency electromagnetic waves) can couple to a material and a whole host of ways that the microwave energy is subsequently lost to the system. The main loss mechanisms are electric, conduction (eddy current), hysteresis and resonance (domain wall and electron spin (FMR)). It is often difficult to ascertain which loss mechanism, or combination of mechanisms is occurring for a particular sample in given conditions. The different mechanisms do however have different dependencies on certain properties such as sample type and microstructure, frequency and temperature. A brief description of these different loss mechanisms will be given below for background purposes before a detailed description of ferromagnetic resonance (FMR) phenomena including high power effects is given. Finally the role of microwave heat in industry will be touched upon with particular reference to magnetite.

3.2. Loss Mechanisms

The two main loss mechanisms for non-magnetic materials are dielectric (dipolar) losses and conduction losses. Conduction losses dominate in metallic, high conductivity materials and dipolar losses dominate in dielectric insulators. Magnetic materials also exhibit conduction losses with additional magnetic losses such as hysteresis, domain wall resonance and electron spin resonance (FMR).

3.2.1. Losses due to the Oscillating Electric Field

In dielectric (electrically insulating) materials such as industrial ceramics and the bulk matrix of a lava, the absorption (degree of interaction) of microwaves is related to the material's complex permittivity e .

$$e = e_0(e' - ie'')$$

Where e_0 is the permittivity of free space (e_0 = 8.86 x 10^{-12} F/m), the real part e' is the relative dielectric constant and the imaginary part e'' is the effective relative dielectric loss factor.

When microwaves penetrate and propagate through a dielectric material the internal field generated within the effected volume induces translational

motions of free or bound charges such as electrons or ions, and rotates charge complexes such as dipoles. Inertial, elastic and frictional forces resist these induced motions and cause losses, a consequence of which is volumetric heating (Sutton, 1989). These loss mechanisms are combined together for convenience to give the one electric loss parameter e''. Fig. 3.2 illustrates the variation in e' and e'' for a dielectric showing 'Debye' relaxation.

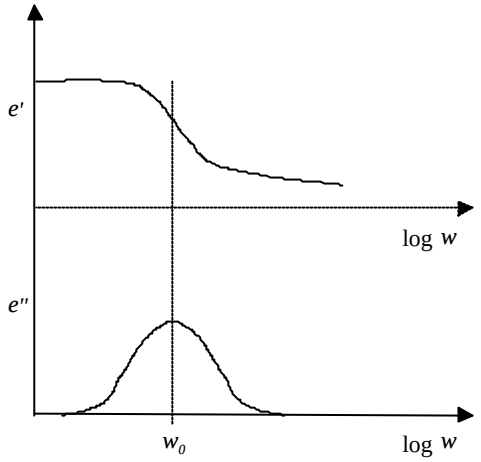

Figure 3.2 Variation in permittivity with frequency for a dielectric showing 'Debye' relaxation, w_0 being the resonance frequency (after Moulson & Herbert, 1990).

The loss tangent $\tan d$ is also commonly used to describe these losses, which is defined as

$$\tan d = \frac{e''}{e'} = \frac{s}{2pfe_0e'}$$

where s is the total effective conductivity (S/m) caused by ionic conduction and displacement currents and f is the frequency.

The power P that is absorbed per unit volume (W/m^3) of the sample at any instant of time can be described by (Sutton, 1989)

$$P = s|\mathbf{E}|^2 = 2pfe_0e'\tan d|\mathbf{E}|^2$$

where $|\mathbf{E}|$ (V/m) is the magnitude of the internal electric field. It has been assumed that the power is uniform throughout the volume and that thermal equilibrium has been achieved. This is not always the case and in addition $\mathbf{E}$, $\tan d$, e' and f are all in fact interdependent. However, it does provide a useful approximation for the power absorbed and describes the basic relationships

between the four variables. It can be seen that the power absorbed varies linearly with frequency, the relative dielectric constant, and the loss tangent, and varies with the square of the electric field.

3.2.2. Losses due to the Oscillating Magnetic Field

The permeability μ, of a material is defined as

$$\mathbf{B} = \mu\mathbf{H} = \mu_0(\mathbf{H} + \mathbf{M})$$

where $\mathbf{B}$ is the flux density (T), $\mathbf{H}$ is the magnetic field intensity (A/m), $\mathbf{M}$ is the magnetisation (A/m) and μ_0 is the permeability of free space (4π x 10^{-7} H/m). For small fields the magnetisation is proportional to the field intensity which means that the initial relative permeability, μ_r, is a constant.

$$\mu_r = \frac{\mu}{\mu_0} = 1 + \frac{\mathbf{M}}{\mathbf{H}} = 1 + \chi$$

In an analogous way to the electric losses, the losses that occur due to a time varying magnetic field can be described by a complex relative permeability

$$\mu_r = \mu' - i\mu''$$

where μ' is the permeability and μ'' describes all the magnetic losses. An analogous magnetic loss tangent, $\tan\delta_\mu$ can also be defined where

$$\tan\delta_\mu = \frac{\mu''}{\mu'}$$

In a similar way, the power P that is absorbed per unit volume (W/m^3) of the sample at a given instant in time can be described by (Krage, 1981)

$$P = 2\pi f\mu_0\mu''|\mathbf{H}|^2 = 2\pi f\mu_0\mu'\tan\delta_\mu|\mathbf{H}|^2$$

It can be seen that the power absorbed varies linearly with frequency, permeability, and the loss tangent and with the square of the magnetic field.

For a ferrite the loss tangent can be expressed in terms of three main contributors (Moulson & Herbert, 1990),

$$\tan\delta_\mu = \tan\delta_h + \tan\delta_e + \tan\delta_r$$

in which $\tan\delta_h$, $\tan\delta_e$ and $\tan\delta_r$ are the hysteresis, eddy current and 'residual' loss tangents respectively. The processes that contribute to the residual losses include the resonance losses and at high frequencies these will often dominate.

3.2.2.1. Hysteresis Losses

As a result of hysteresis, energy is dissipated as heat in a magnetic material as it travels around a **B** - **H** hysteresis loop. The hysteresis energy loss W_h per unit volume of material is

$$W_h = \oint \mathbf{B}\, d\mathbf{H}$$

This loss is controlled by factors that control low frequency permeability and coercivity such as porosity, grain size and impurities as well as the intrinsic properties.

3.2.2.2. Eddy Current Losses

The conductivity of the material is important as it determines the extent of losses due to eddy currents. For ferrites the conduction mechanism is believed to be electron hopping between ions of the same type on equivalent lattice sites, e.g. for magnetite $Fe^{3+} \leftrightarrow Fe^{2+}$ (Moulson & Herbert, 1990). This mechanism is also known as valence exchange (Sparks, 1964). Magnetite is one of the most conductive oxides with conductivity, s, approximately 10^4 S/m at room temperature (Moulson & Herbert, 1990).

The skin depth, d, in a conductive material is the depth of penetration of the magnetic field (as well as the current density) at which its value decreases by $1/e$ of its surface value. (This is analogous to the penetration depth for a dielectric in which case it is the electric field that penetrates.) The skin depth is given by (Metaxas, 1996)

$$d = \sqrt{\frac{2}{s\, 2pfm}}$$

When the skin depth is large compared to the sample size the influence of eddy currents on the magnetic field is entirely negligible. Hence, in calculations an infinite skin depth can be assumed and the energy dissipation for various shapes evaluated (Smit & Wijn, 1955; Sparks, 1964). The energy dissipation W can be evaluated using the equation

$$W = s \int \mathbf{E}^2 dV$$

The energy dissipation will differ for different sample shapes but in all cases it increases with the square of the frequency, magnetic field and sample dimension (radius for spheres and cylinders) and increases linearly with conductivity.

$$W \propto s\, f^2 \mathbf{B}^2 r^2$$

For high conductivity materials such as metals (or for very large samples), where the skin depth is less than the dimensions of the sample, the influence of eddy currents on the magnetic field ceases to be negligible. In this situation a broad range of spin waves are excited, as opposed to only the uniform precession (see Section 3.1.3) with the result that in metals the eddy current process often dominates.

The conductivity of a ferrite due to valence exchange increases with temperature T, in the form shown below (e.g. Tebble & Craik, 1969)

$$s = s_0 \exp -(E_p / kT)$$

where E_p is the activation energy and k is Boltzmanns constant (1.38 x 10^{-23} J/K). It can be seen that there is a log relation between eddy current losses and temperature and that eddy current losses are more likely to become important at high temperatures. This is illustrated in Fig. 3.3 for different sized samples where it can be seen that at high temperatures (where eddy current losses dominate) the larger the sample, the larger the loss.

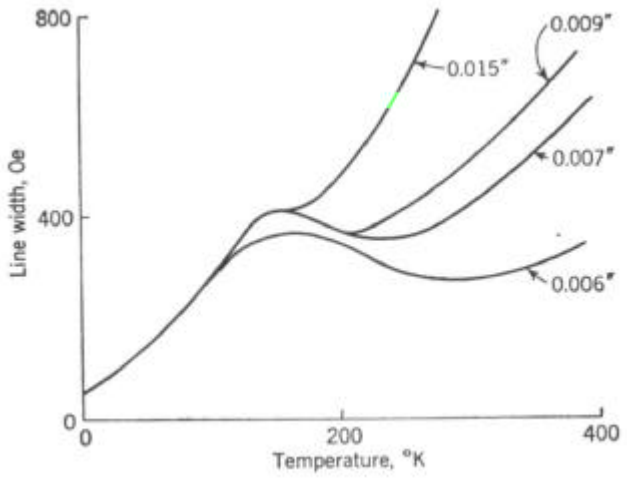

Figure 3.3 Eddy current contribution to line width (another way of representing absorption) for various sample sizes of a nickel ferrite (from Sparks, 1964 after Yager, Galt & Merritt, 1955).

3.2.2.3. Domain Wall Resonance Losses

The resonance phenomena can in general be divided into two distinct mechanisms; domain wall resonance and those due to electron spin (FMR). Domain wall resonance occurs at approximately one tenth of the frequency of FMR (Moulson & Herbert, 1990). The small displacements of a pinned domain wall with an applied field introduces restoring forces. Since the wall has inertia and its movement is accompanied by energy dissipation, an equation of motion can be written for a sinusoidal applied field (Moulson & Herbert, 1990)

$$m\ddot{x} + b\dot{x} + cx = 2\mathbf{MB}(t)$$

where x is the displacement normal to the wall, m is the inertia, b is the damping coefficient and c is the stiffness coefficient. This equation is in the form of damped forced harmonic motion so resonance will occur, if damping is small, at a characteristic angular frequency $w = (c/m)^{1/2}$. This resonance will become less significant the smaller the grain and will not occur in the case of single domain grains. This is illustrated in Fig. 3.4 for a single and multi domain nickel ferrite.

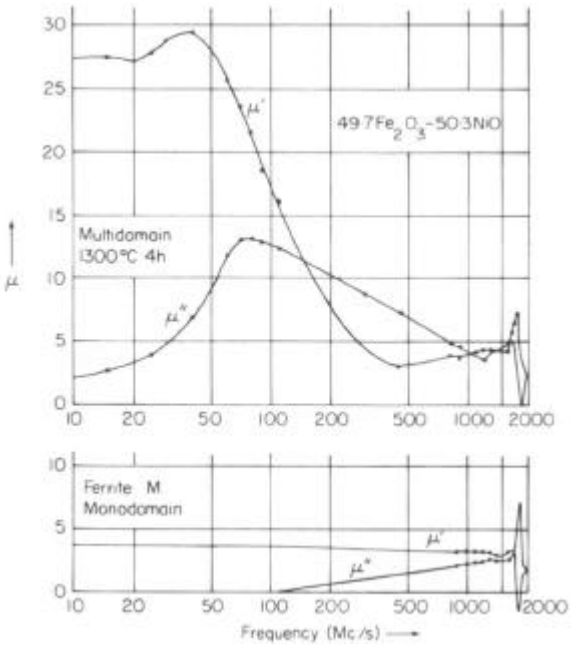

Figure 3.4 The effect of grain size on the permeability spectra of a nickel ferrite, both specimens have the same saturation magnetisation and spin resonance frequency (from Tebble & Craik, 1969 after Globus & Duplex, 1964).

3.3. Ferromagnetic Resonance (FMR)

The other resonance phenomenon (in addition to domain wall resonance) is ferromagnetic resonance. This is the mechanism by which palaeomagnetic samples are demagnetised with the Liverpool microwave equipment.

Griffiths reported the first observations of FMR in 1946 for electrolytically deposited films of iron, cobalt and nickel (Patton, 1975). Kittel (1948) advanced the understanding of the process considerably when the role of the demagnetising fields in determining the resonance condition was determined. Ferrites are treated in the same way as ferromagnets with the assumption that the two sub lattices remain coupled throughout and that it is the resultant magnetisation vector that precesses. This is a good approximation but there is an additional ferrimagnetic exchange resonance mode in which the sub lattices do not remain anti parallel but precess in opposite senses. This results in resonances in the optical and infra red regions, out of the microwave frequency range.

The physical basis for FMR is introduced firstly. The fundamentals of magnetism have been described previously in Sections 1.1.2 and 1.1.3 but in this section the energy terms are discussed in terms of frequency. The next section describes the high power process of parallel pumping which is used at Liverpool. Finally, the different relaxation channels of the microwave energy are discussed.

3.3.1. Physical Basis

In a simple model of a ferromagnet, the sample can be represented by a large number of electron spins, one for each ion (see Section 1.1.2). Each of these spins has a magnetic moment **m**, given by

$$\mathbf{m} = -g m_B \mathbf{S}$$

where g is the spectroscopic splitting factor (take as equal to 2), m_B is the Bohr magneton (9.274×10^{-24} Am^2) and $\mathbf{S}$ is the spin. The exchange force tends to align these spins parallel to each other (see Section 1.1.3). As the magnetic moment is associated with an angular momentum, the magnetic moment experiences a torque when in a static magnetic field. This field can be an applied field or else it is the anisotropy field of the grain. As a result, the moment precesses around this field with angular frequency w, where

$$w = gH$$

or

$$f = 35.2 \times 10^6 H \text{ GHz}$$

where g is the gyroscopic ratio ($g = gm_B/\hbar$ = 1.76 x 10^{11} $T^{-1}s^{-1}$), H is the magnetic field (A/m) and f is the frequency in GHz. This is analogous to a spinning top where the axis of rotation is at an angle to the gravitational field.

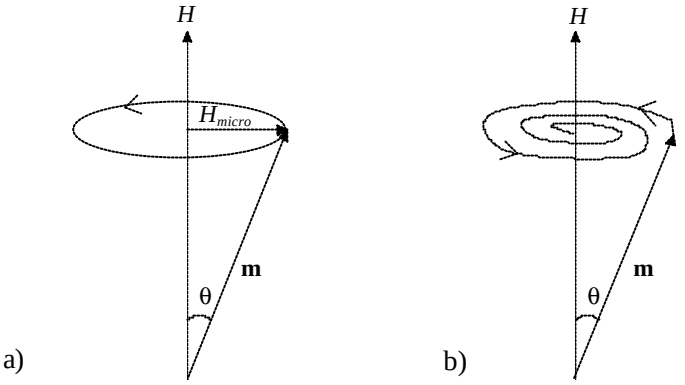

Figure 3.5 Precessional motion of magnetisation; a) precession maintained by a microwave field and b) the moment spiralling into line with *H* as the precessional energy is dissipated (after Moulson & Herbert, 1990).

If a microwave field is applied at right angles to the static field (Fig. 3.5a) at the same frequency as the precession (resonance condition) then the torque will always be in such a direction as to open up the angle of precession. Thus energy will be absorbed from the microwave field and the magnetisation will precess with a larger angle. If the microwave field is then removed the precessional energy is dissipated and the magnetic moment will spiral in until it is parallel with the static field since this is the minimum energy state (Fig. 3.5b). The static field induces the precession only it does not cause the relaxation back to the equilibrium position. The additional force required to drive the magnetic moment to the equilibrium value is supplied by the interaction of the excited magnetisation mode with other degrees of freedoms of the system, for example with lattice vibrations and other modes of the spin system.

The modes of importance that are excited by microwaves are the spin wave modes (see Section 1.1.3). A spin wave is a normal mode of the spin system and it is also known as a magnon (c.f. photons and phonons). As for photons and phonons in a solid, magnons are quantised. This means that only definite states

described by a characteristic dispersion relation $w_k = w(\mathbf{k})$ are allowed, where $\mathbf{k}$ is the spin wave vector and w_k is the spin wave angular frequency.

In general three energy terms contribute to the spin wave energy $\hbar w_k$, and hence the normal mode frequency w_k at a specified $\mathbf{k}$. These are the Zeeman energy, the exchange energy and the magnetostatic or dipolar energy (see Section 1.1.2 and 1.1.3).

The Zeeman and exchange energy terms can be described as below (e.g. Patton, 1975)

$$(\hbar w)_{Zeeman} = -g m_B \sum_i \mathbf{S}_i \cdot \mathbf{H}$$

$$(\hbar w)_{exch} = -2J \sum_{i,j} \mathbf{S}_i \cdot \mathbf{S}_j$$

where g is the spectroscopic splitting factor , m_B is the Bohr magneton, $\mathbf{H}$ is the static magnetic field (applied or anisotropy) and J is the exchange integral. The j summation is over the nearest neighbours of spin $\mathbf{S}_i$ and the i summations are over the N spins in the system. The ground state of the system has every spin in the sample aligned parallel to the static field. This corresponds to the minimum energy states for the Zeeman and the exchange energies. An initial guess at the first excited state could be to flip one spin anti-parallel to the magnetic field whilst keeping the others parallel. However, this state corresponds to a very high exchange energy and does not represent an eigenmode of the system. Instead, a twist on the system where each spin is tipped slightly away from the field direction gives a much smaller exchange energy (Fig. 3.6 and Fig. 1.1). Each spin is tipped through a small angle b, the precession angle, with respect to the static field and the angle a is the angle between successive spins. The exchange energy is small for small a and the Zeeman energy is small for small b. This disturbance on the spin system is called a spin wave and is a normal mode of the spin system.

The shortest distance between parallel spins (except in the plane where all spins are parallel) defines the propagation direction and the wavelength l. The wave vector $\mathbf{k}$ has magnitude $2p / l$ and is directed along the propagation direction.

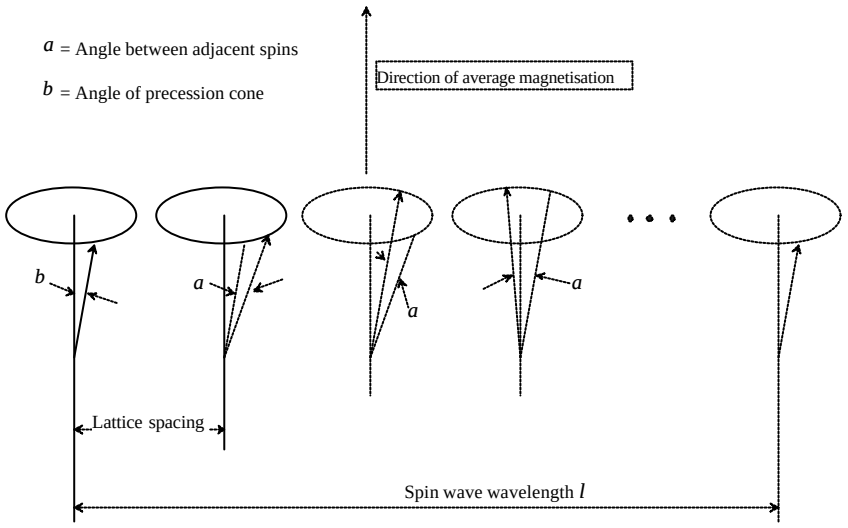

Figure 3.6 Spin configuration (spatial distribution) for spin wave mode (after Patton, 1975).

As well as direction the other information needed to describe a spin wave is the amplitude, which is quantised.

$$M_z = M_s(0)V - gm_B$$

where M_z is the total z component of the magnetic moment of the system, $M_s(0)$ is the saturation magnetisation at $T = 0$, and $M_s(0)V$ is the maximum possible value of M_z attained when all the spins are parallel. z is an arbitrary direction chosen as the direction of the static field.

When the spins are tipped through the small angle b the total change in magnetic moment is the equivalent to one spin being flipped over. Spin waves quantised in this way are known as magnons. When there is more than one magnon excited the total z component of the magnetic moment is reduced by gm_B for each magnon excited.

$$M_z = M_s(0)V - gm_B \sum_k n_k$$

where n_k is the number of magnons with wave vector k. This is strictly valid only when dipole-dipole forces are small. The equation shows that the excitation of spin wave modes causes a reduction in the average magnetisation. This is the process of thermal demagnetisation.

Using this qualitative spin wave picture, the general shape of the dispersion relation $w_k = w(\mathbf{k})$ can be obtained. The exchange energy between neighbouring spins is

$$(\hbar w)_{exch} \propto \mathbf{S}_i \cdot \mathbf{S}_{i+1} \propto -\cos a \approx -(1-\tfrac{1}{2}a^2)$$

for small a. As a and $\mathbf{k}$ are inversely proportional to the wavelength l ($a \approx 2pa/l$ where a is the spin separation distance), a is proportional to $\mathbf{k}$. Thus the exchange energy of the magnon is

$$(\hbar w)_{exch} = D\mathbf{k}^2$$

where the proportionality constant D is the exchange constant. This leads to a magnon energy comprised of the Zeeman and exchange energies given by

$$\hbar w = gm_B + D\mathbf{k}^2$$

where gm_B is the Zeeman energy. This is illustrated in Fig. 3.7.

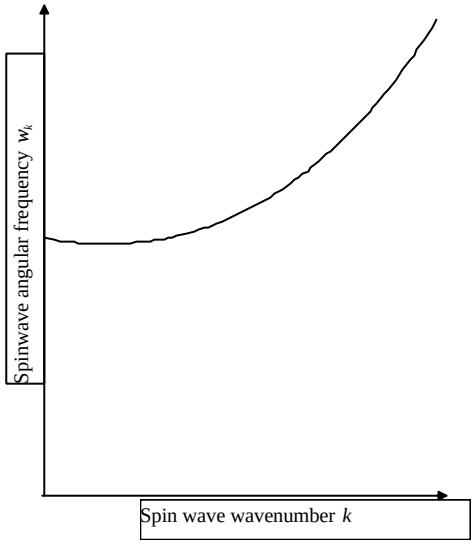

Figure 3.7 Dispersion relation with Zeeman and exchange energies included.

In general, the sample size is much less than the wavelength of the microwave field so that the microwave field is constant over the whole sample. Therefore, each spin experiences the same torque and the spins are parallel to each other as the field opens up the precession angle. Parallel spins imply an infinite wavelength, which corresponds to $\mathbf{k} = 0$. These $\mathbf{k} = 0$ magnons are important and are known as the uniform precession magnons.

The dispersion relation however, is still incomplete until the magnetostatic energy has been considered. The importance of this term is demonstrated in Fig. 3.8 where modes with $\mathbf{k}$ perpendicular and parallel to the static field and the average magnetisation are shown. The divergence of the magnetisation is zero when $\mathbf{k}$ and $\mathbf{H}$ are parallel making the magnetostatic energy term zero so that

the dispersion relation is essentially as given previously. For $\mathbf{k}$ perpendicular to $\mathbf{H}$ the magnetisation has a non zero divergence and dipolar fields are generated. These fields may be viewed as arising from magnetic charges. The dipolar fields that are generated when $\mathbf{k}$ and $\mathbf{H}$ are not parallel have two effects. The fields tend to raise the energy of the spin system and the fields alter the spin motions from circular precession to an elliptical precession cone.

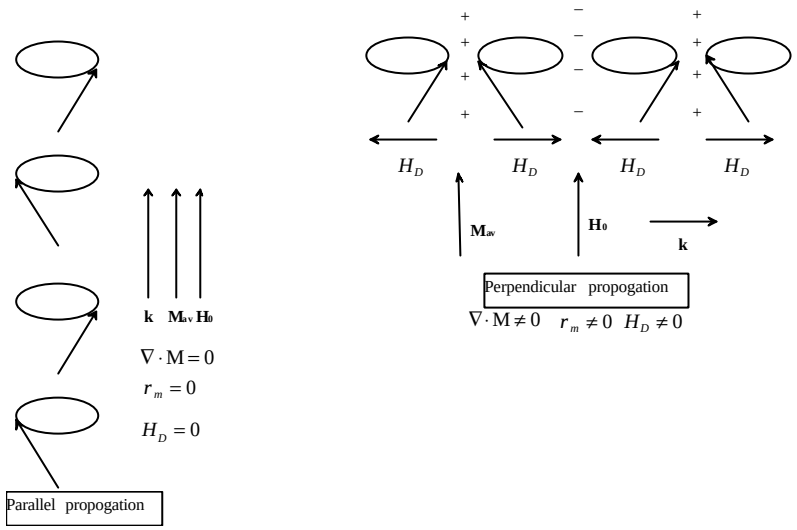

Figure 3.8 Schematic representation of magnetostatic charge distribution for spin wave modes (after Patton, 1975).

The spin wave demagnetisation fields pictured in Fig. 3.8 are volume demagnetisation fields. The surface demagnetisation fields can be neglected if the magnon wavelength is much smaller than the dimensions of the sample otherwise they must be taken into account. The resulting modes are called the magnetostatic modes. The uniform precession has $\mathbf{k} = 0$ and therefore surface demagnetisation fields are important and the resulting demagnetisation field is different for different sample shapes. Thus, when the demagnetisation energy is taken into account there are magnons that are degenerate with the uniform mode.

When the dipolar interaction is rigorously taken into account (see Sparks, 1964 or Patton, 1975 for detailed mathematical derivations) the dispersion relation is given by

$$\hbar w = \left[(D\mathbf{k}^2 + \hbar w_i)(D\mathbf{k}^2 + \hbar w_i + \hbar w_m \sin^2 q_k) \right]^{1/2}$$

where $$\hbar w_m = 4p\hbar g\mathbf{M}$$

and $\hbar w_i = \hbar g \mathbf{H}$

where g is the gyroscopic ratio and q_k is the angle between $\mathbf{k}$ and $\mathbf{H}$, where $\mathbf{H}$ is the internal field that comprises an applied field if there is one and the contributions of the demagnetisation factor (anisotropy field). The total dispersion relation including Zeeman, exchange and volume magnetostatic energies is shown in Fig. 3.9. The curves represent a quasi-continuum of magnon states which is called the spin wave manifold. The increase in w with $\mathbf{k}$ is due to exchange, the increase with q_k is due to dipolar interactions and the field dependant term $g\mathbf{H}$ is the Zeeman contribution.

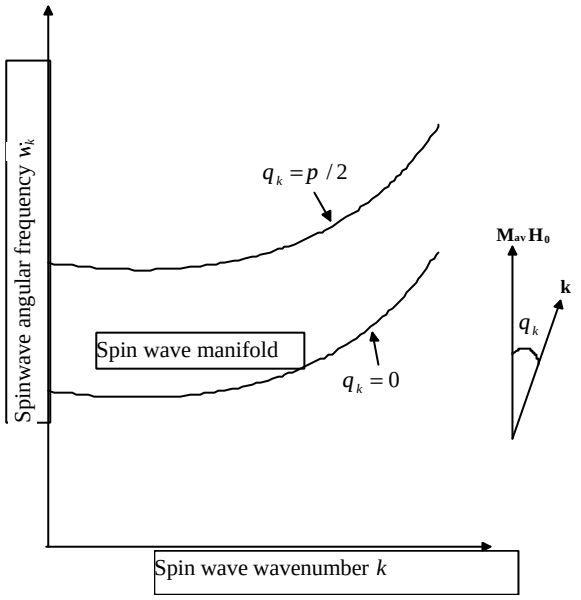

Figure 3.9 Magnon dispersion relation with exchange, Zeeman and dipole-dipole energies included (after Patton, 1975).

3.3.2. High Power Effects

At low microwave power levels the magnetisation response can be described adequately by a field independent susceptibility, the response being linear. However at high powers the coupling between the microwave field and spin wave modes can lead to a non-linear response. It was Suhl in the 50s (Patton, 1975) who first explained high power effects. At low power the uniform precession amplitude is small so that the spin wave amplitudes stay essentially at their thermal values. As the power level increases the spin wave amplitude

increases and more and more energy is pumped into the spin wave modes. At some critical power level the power input into the mode exceeds the rate at which it can be lost due to relaxation and some spin wave amplitudes increase beyond their thermal values. The analogy is that of a leaky bucket. At some critical point where the rate of water into the bucket exceeds the rate of leakage the bucket will fill with water. Fig. 3.10 illustrates the effect of critical power absorption.

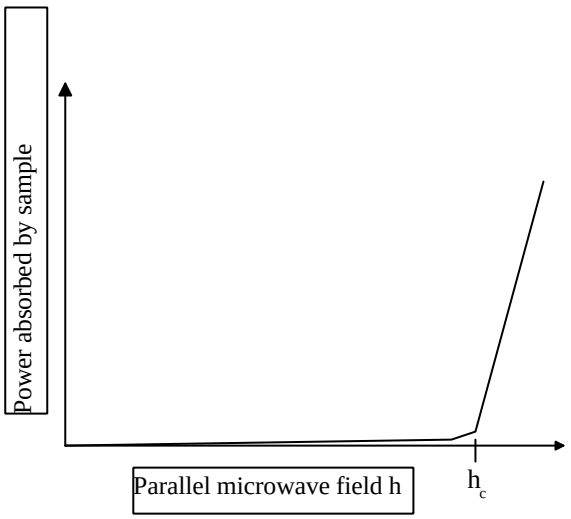

Figure 3.10 Power absorption for parallel pumping showing threshold behaviour (after Sparks, 1964).

Schlömann *et al.* (1960) and Morgenthaler (1960) both proposed the process of high power parallel pumping in which the microwave field is parallel to the static field (the early work had been on transverse pumping). Parallel pumping results in the parametric excitation of spin waves and associated instability effects.

It is spin wave ellipticity that allows the direct coupling between the parallel pump microwave field and certain spin wave modes as Fig.3.11 illustrates for a single precessing spin with an elliptical precession cone.

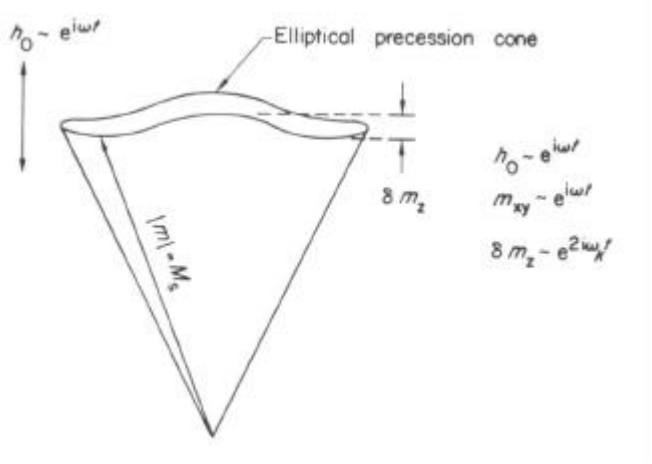

Figure 3. 11 Schematic representation of elliptical precession cone for spin waves which leads to the parallel pump instability (from Patton, 1975).

If the magnitude of the magnetisation is constant then the elliptical orbit results in a wobble dm_z in the z component. The wobble has an $\exp(2iw_k t)$ time dependence. Hence, a spin wave mode at w_k can couple to a microwave field with $w = 2w_k$. There is no ellipticity at $q_k = 0$ so the coupling and the amplitude of dm_z are zero. Maximum coupling and the maximum amplitude of dm_z occur for maximum ellipticity, which occurs at $q_k = p/2$.

The ellipticity is essentially a result of the dipole-dipole energy term in the dispersion relation so a reasonable assumption for the q_k dependence of the coupling strength is $|dm_z| \approx \sin^2 q_k$. More detailed theory gives the rate of energy input to spin waves at $w = 2w_k$ proportional to

$$P = (w_m / w)(gh_0 / 2)\sin^2 q_k$$

The threshold field h_c is the value of the static field h_0 at which P is equal to the relaxation rate h_k. It is given by

$$h_c = \frac{(w / w_m)\Delta H_k}{\sin^2 q_k}$$

where $\Delta H_k = 2h_k / g = 1/gT_k$ is the spin wave linewidth and T_k is the relaxation time of energy. When $q_k = 0$, h_c is infinite so that the threshold does indeed diverge at $q_k = 0$ due to the ellipticity being zero.

For a given w_k, q_k and $\mathbf{k}$ are related through the dispersion relation. The range of allowed $\mathbf{k}$ or q_k depends on the biasing conditions as illustrated in Fig. 3.12.

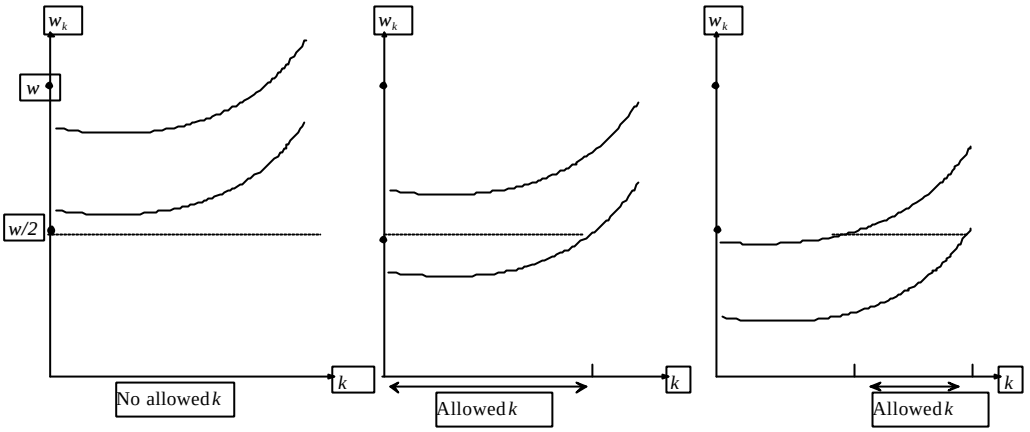

Figure 3.12 Position of spin wave manifold with respect to potentially unstable modes at $w_k = w/2$ for different static fields (after Patton, 1975).

3.3.3. Relaxation Processes

The basis for resonance is the precessional motion of the magnetisation that follows directly from the classical magnetic torque equation

$$(d\mathbf{M}/dt)_{precession} = -g(\mathbf{M} \times \mathbf{H})$$

However the torque equation as it stands contains no loss term which implies that once the microwave field excites the uniform precession mode, the motion will continue indefinitely if the exciting field is turned off. This is not the case for real materials where the coupling between the precessing spin system and the lattice, results in the damping of the precession motion. In the absence of a driving field the magnetisation will gradually relax to the equilibrium position along the static field direction for an isotropic sample (Fig. 3.5). There are different types of phenomenological damping terms that can account for the losses in the magnetic system, which are discussed in Patton (1975) and Sparks (1964).

The physical origin of relaxation is quite complicated. There are many possible physical relaxation channels by which the energy contained in the spin wave modes can be dissipated. A convenient way of describing the various

channels is by scattering interactions. The individual scattering mechanisms that may contribute are summarised in Fig. 3.13.

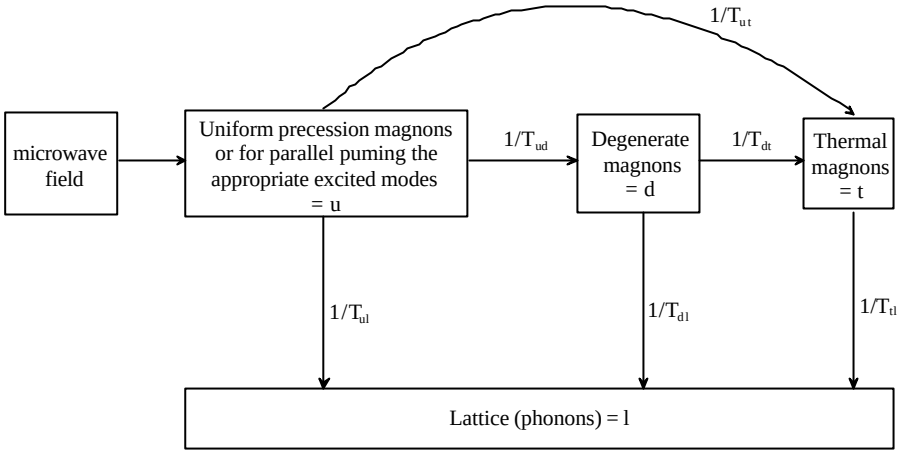

Figure 3.13 Classification of relaxation frequencies. The various channels are as follows:

$1/T_{ud}$ two magnon,
$1/T_{ul}$ magnon phonon, eddy current, rapidly relaxing impurity, slowly relaxing impurity, and valence exchange,
$1/T_{ut}$ three magnon such as Kasuya-LeCraw, four magnon,
$1/T_{dl}$ same as $1/T_{ul}$,
$1/T_{dt}$ three magnon,
$1/T_{tl}$ magnon phonon.

After Sparks (1964).

The microwave field excites a resonance and the magnons eventually relax into phonons (lattice heating) by various means. Different schemes have been suggested, and tried, for determining which processes are occurring. The dependencies of the different channels to different parameters such as temperature and frequency provide a way to distinguish between processes. The shape and width of the resonance and the relaxation times also provide information. The details of the different relaxation processes will not be discussed here; instead the reader is referred to Sparks (1964) and Patton (1975).

YIG (yttrium iron garnet) has been used extensively in FMR studies since it has simple, well known properties. It has a cubic structure, is a good insulator, has all magnetic ions identical and in the S state and has no detectable magnetic disorder. The simplest cases to study are magnetically saturated single crystals. More complex behaviour is seen for polycrystals, mixtures and partially magnetised materials, where in general line width broadening is seen. Natural ceramics will show complicated behaviour but these are magnetically much simpler than lava which contains a range of magnetic minerals and grain sizes.

For palaeomagnetic purposes it is of little practical interest which relaxation process is occurring as it is the demagnetisation of the sample that is of interest. It is, however, an interesting topic and could be a future area of study. Russian researchers (Raikher & Stepanov, 1992) have attempted to explain the observations from FMR experiments of ultra dispersed ferromagnets and ferrites on theoretical grounds. They investigated an ensemble of randomly oriented single domain, anisotropic, ferromagnetic particles and the effect of super paramagnetism to the resonance characteristics with some success at explaining the experimental observations.

3.4. Industrial Microwave Heating

In industry, microwaves are used for heating (the reduction of the magnetic moment of magnetic materials is of little importance to industry). High losses are desirable so that large amounts of heat can be imparted rapidly to the material in question. Microwave heating is used in industry for various processes such as sintering, food heating, gluing and for a variety of materials (e.g. Metaxas, 1996) and is also an area of on going research (e.g. Roy *et al.*, 1999).

In industry low frequency microwaves (often in MHz range) are generally used at high powers (hundreds of Watts). The principle processes of heating are from conduction losses (dominant in metallic, high conductivity materials) and dipolar losses (dominant process for dielectric insulators). Magnetite is a good absorber of microwave energy at low frequency and high powers as shown in Fig. 3.14.

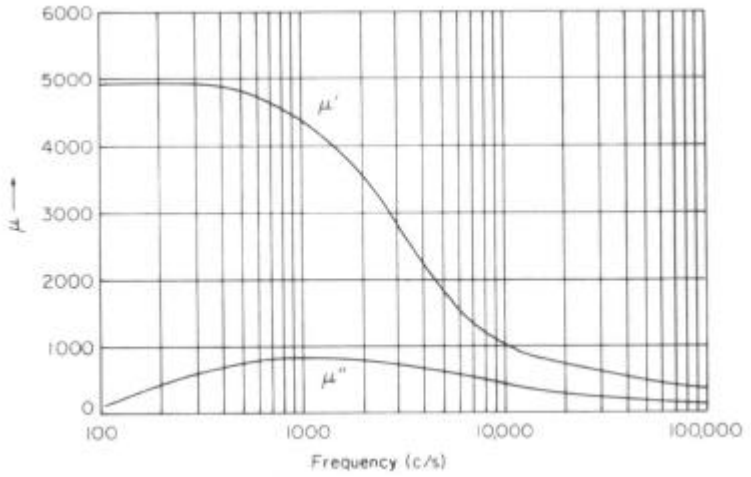

Figure 3.14 Permeability spectra for magnetite illustrating high relaxation losses at low frequencies (from Tebble & Craik, 1969 after Galt, 1952).

The condition of thermal runaway occurs in a microwave heated material at temperatures above a critical temperature when the loss tangent rises very rapidly so that the material begins to absorb the microwave energy more efficiently, which also raises the temperature. This in turn causes tanδ to rise even faster. The net result is an exponential increase (runaway) in temperature (Metaxas, 1996). The rate of the temperature rise and the critical triggering temperature vary widely for different materials (Sutton, 1989). Thermal runaway has been shown (McGill *et al.*, 1988) to occur at room temperature for magnetite (Fig. 3.15).

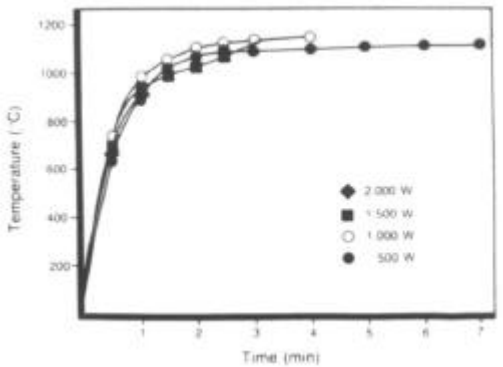

Figure 3.15 Microwave heating at 2.45 GHz of magnetite (from Sutton, 1989 after McGill *et al*. 1988).

Walkeiwicz *et al.* (1988) made a microwave heating study of various materials in a 1 kW, 2.45 GHz oven. A 25 g sample of magnetite powder reached

1258 °C in only 2.75 minutes making it one of the best absorbers of the 150 reagent grade elements, compounds and natural minerals they studied. As magnetite couples so well with microwaves at room temperature it can be used as an additive to microwave transparent material to increase the overall heating rate at lower temperatures. The added magnetite will be selectively heated in the microwave field. Fig. 3.16 illustrates the effect on microwave heating when 10 and 50 wt% magnetite is added to alumina powders. This is a similar scenario to palaeomagnetic samples where the magnetic minerals are dispersed in a non-magnetic matrix. Magnetite has also been used in microwave sintering of iron ore (Walkiewicz *et al.*, 1991) where the rapid thermal expansion of magnetite cracks the surrounding rock matrix thus reducing the energy required in the ore grinding process.

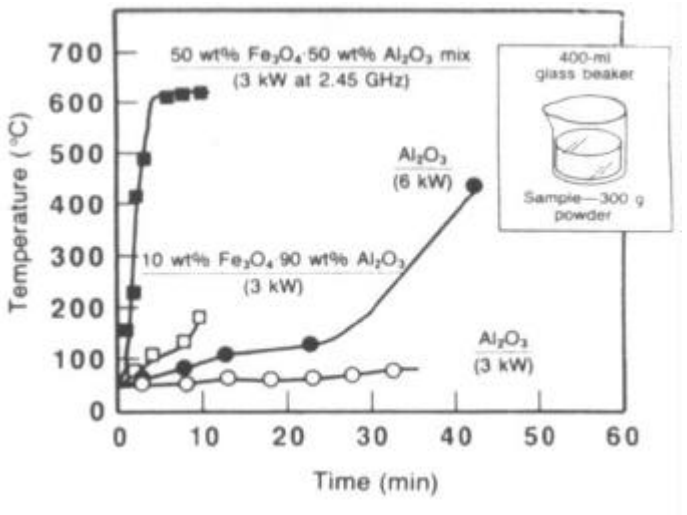

Figure 3.16 Effect of magnetite additions on the microwave heating of alumina powders (from Sutton, 1989 after Sutton, 1988).

Medicine is another area where microwaves and magnetite are combined. There has been recent concern (Kirschvink, 1996) that the small quantities of magnetite in human and animal tissue may absorb pollutant microwave radiation via FMR to a harmful level. In contrast, magnetite can be used in a beneficial way where the addition of fine grained magnetite to diseased cells can be used to selectively absorb microwaves and thus selectively destroy diseased tissue (Kletetschka, pers. comm.).

In a sample of ceramic or lava where the magnetic grains are surrounded by a non-magnetic matrix the magnetic grains will be preferentially heated. The heat will then be transferred to the bulk of the sample by conventional processes. The industrial method of using low frequency, high power microwaves has advantages over conventional heating in that the heat is applied much more rapidly reducing the possibilities of alteration (alteration is both time and temperature dependant (Tanguy, 1975; Walton, 1988)). In the time scales used (seconds) to demagnetise the sample it will probably reach thermal equilibrium so that the bulk sample will be heated considerably but for shorter times than in conventional experiments. It is preferable to use FMR, so that the spin systems of the magnetic minerals are directly excited and the sample demagnetised prior to the formation of heat in the relaxation process.

The only other published method of using microwaves for palaeomagnetic demagnetisation has been by Hale *et al.* (1978). They studied lunar material with microwaves of 2.64 GHz. The magnetic phases in the lunar rock are metallic iron or iron nickel alloys that are good electrical conductors. Hence induction heating takes place preferentially in the magnetic carriers rather than the dielectric matrix. They demonstrated with artificial mixtures of iron dispersed in an aluminium oxide matrix that heat gain and demagnetisation were a result of eddy current losses. The larger the grains, the greater the loss and the losses increased with length of exposure as expected. They attempted a microwave Thellier palaeointensity experiment but with little success. The matrix was still significantly heated and as a result chemical alteration took place.

3.5. Summary

A general description of the behaviour of material to microwave exposure has been given. For magnetic materials the principal loss mechanisms are hysteresis, eddy current and the resonance losses (domain wall and electron spin (FMR)). Eddy current heating is ideal for industrial processes where rapid heating of materials is desired. The two main mechanisms of importance for palaeomagnetic samples are eddy current heating and FMR. Demagnetisation of the magnetic vector is required with as little heat to the bulk sample as possible. This is achieved with FMR as the microwave energy first reduces the magnetic moment and then the energy is subsequently lost to the system as heat. The

appropriate frequency of microwaves has to be used so that FMR can occur in all magnetic particles present in a particular sample. If only a few magnetic particles are in resonance, then higher powers will be needed for demagnetisation so that heat from the relaxation process along with any eddy current heating occurring can demagnetise the rest of the magnetic minerals.

Magnetite is a good absorber of microwaves at low frequencies as shown in Fig. 3.15. In natural rock or ceramics the magnetic minerals only make up a small percentage of the total volume and are dispersed throughout a non-magnetic matrix. This is a similar situation to that shown in Fig. 3.16 where various proportions of magnetite have been added to non-magnetic alumina powder. When there is 10% magnetite dispersed in the alumina powder the temperature of the bulk sample only reaches 200 °C after 10 minutes of microwave exposure. This is encouraging for palaeomagnetic purposes.