Novel Instrumentation for High Frequency Electrical Conductivity and Magnetisation Dynamics Characterisation

Microwave Reflectometry and SQUID-based FMR

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Declaration

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Summary

The drive towards smaller devices and faster operational speeds places a challenge on techniques capable of probing high-frequency processes both on-chip and in bulk devices. The objective of this work is to expand the range of high-frequency broadband measurements for electrical conductivity and in turn magnetisation dynamics.

This work bridges the gap between conventional methods for characterisation of synthetic thin films and scattering-based methods used on random and pseudo-random self-organised structures. It further develops methods of non-destructively characterising nanostructures. Samples consist of poly(vinylpyrrolidone) (PVP) surface coated Ag and oxide-passivated Ni nanowires (NW) with average diameters of 86 nm and 80 nm respectively. NWs are spray-cast into thin films of varying optical transmittance ($\approx 65 - 90 \%$ $T$) on dielectric and semi-conducting substrates. Contactless (inductive) broadband (CW (Continuous Wave) VNA (Vector Network Analysis) $< 3.3$ GHz and pulsed TDR (Time-Domain Reflectometry) $< 50$ GHz) measurements of conductance and distributed capacitance are performed. Direct (galvanic) pulsing of the aforementioned networks via integrated slotline structures is used in an attempt to reach the athermal transport-limited avalanche regime for the wire-to-wire contacts at room temperature. In-situ impedance measurements demonstrate the enhanced conductivity and hence activation or part-activation of the network following microwave pulsing. Initial conductances of $3.9 - 0.8$ ($\mu\Omega$m)$^{-1}$ are shown to increase to $4 - 1.4$ ($\mu\Omega$m)$^{-1}$ following successive pulsing. Laser specular scattering techniques are exploited to enhance understanding of film surface topography. The characteristic NW lengths are determined to be approximately 50-70 $\mu$m and 30 $\mu$m for Ni and Ag NWs respectively.

Sets of finite bandwidth cavity waveguides are designed, simulated, constructed and used to evaluate inkjet printed silver films as potential coatings for EMI shielding applications. Comprehensive microwave reflectometry analysis is performed in three distinct frequency bands (X-Ku: 7-14 GHz, K: 15-20 GHz and Ka: 21-42 GHz) for films of varying thickness, in order to establish the threshold at which the films transition from a microwave transparent state to a microwave reflective state. It is shown that films with a thickness beyond 120 nm are consistently effective EMI shields but the rate at which they transition to shielding behaviour depends on the frequency of the stimulus. This analysis is further extended...
to higher frequencies and shorter wavelengths through high-visible-UV densitometry, which indicates that the frequency dispersion holds true at optical frequencies. Supplementary to this, the effect of geometric structuring of Ag films on the microwave propagation is discussed for applications where minimising the mass of the shield without compromising the shielding behaviour is desired. The structures consist of grids where the metallic linewidth is kept a constant 1 mm and the separation between lines is varied. It is shown that for grids with metallic fill factor greater than 55% desirable EMI shielding can be achieved. This is however, strongly dependent on the intended operational frequency band.

Also two new techniques for the detection of broadband (100 MHz - 20 GHz) ferro/ferromagnetic resonance in single and poly-crystalline materials, which rely on SQUID-based gradiometry detection of small changes in the magnetisation are developed. In the first method small changes in the along-the-applied-field projection of the coupled magnetic moment ($\Delta m_z$) are detected as the material is driven into resonance. Absolute measurement of the longitudinal component of the magnetisation and the resonance induced lowering of this moment makes estimation of the precession cone angle accessible, which is typically difficult to extract using conventional cavity or stripline based detection methods. The second method involves the change in $\Delta m_z$ with the resonance-induced thermal heating ($\frac{dm_z}{dT}$). Magnetisation dynamics in bulk Y$_3$Fe$_5$O$_{12}$ is observed over a broad range of experimental temperatures (4 K - 400 K) and fields (10 - 500 mT). The inhomogeneous microwave excitation allows for the observation of higher magnetostatic modes and the convenient tracking of very broad resonances. The two SQUID-detection techniques when combined with conventional broadband VNA-FMR, low-frequency magnetic susceptibility and DC magnetometry, all easily realised, essentially concurrently, using the same module, greatly expand the amount of static and dynamic information accessible.
To Martin and Margaret O'Reilly
“If you want to join the revolution, innovate that’s my solution”

Sasha Velour
Keywords and Abbreviations

Keywords
contactless, transmission line, microwave reflectometry, metallic nanowire networks, inkjet printing, electromagnetic interaction shielding, magnetisation dynamics, ferromagnetic resonance

Abbreviations

AHE  Anomalous Hall Effect
CPW  CoPlanar-Waveguide
CW   Continuous Wave
DUT  Device Under Test
EDX  Energy Dispersive X-ray
EMI  Electromagnetic Interaction
EPR  Electron Paramagnetic Resonance
FFT  Fast Fourier Transform
FIT  Finite Integration Technique
FMRFM Ferromagnetic Resonance Force Microscopy
IF   Intermediate Frequency
iFFT Inverse Fast Fourier Transform
IMD  InterModulation Distortion
IR   Infrared
LED  Light Emitting Diode
LSS  Laser Specular Scattering
MOKE Magneto-Optical Kerr Effect
MOMR Magneto-Optical Magnetic Resonance
MPMS Magnetic Property Measurement System
MWS MicroWave Studio
NMR  Nuclear Magnetic Resonance
NW   NanoWire
NWN  NanoWire Network
PCB  Printed Circuit Board
PID  Proportional Integral Derivative
RBW  Receiver BandWidth
RF   Radio Frequency
SAS  Small Angle Scattering
SE   Shielding Effectiveness
SQUID Superconducting Quantum Interference Device
SIS  Superconductor-Insulator-Superconductor
TDR  Time-Domain Reflectometry
TDT  Time-Domain Transmission
TE   Transverse Electric
TEM  Transverse Electromagnetic
TM   Transverse Magnetic
UV   UltraViolet
VNA  Vector Network Analysis
VSM  Vibrating Sample Magnetometry
XMCD X-ray Magnetic Circular Dichroism
XRD  X-Ray Diffraction
YIG  Yttrium Iron Garnet

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Nomenclature

$m$ Magnetic moment (A m^2)
$R$ Resistance (Ω)
$L$ Inductance (H, T m^2 A^{-1})
$C$ Capacitance (F)
$G$ Conductance (S)
$Z$ Impedance (Ω)
$E$ Energy (J)
$t$ Time (s)
$k$ Wave-vector (m^{-1})
$f$ Frequency (Hz)
$μ$ Magnetic permeability (T m A^{-1})
$ε$ Dielectric permittivity (F m^{-1})
$v_{ph}$ Phase velocity
$v_g$ Group velocity
$k_c$ Cut-off wavenumber
$β$ Wave propagation constant
$f_{mn}$ Cut-off frequency for the $[m n]$ mode (Hz)
$K$ Elliptical integral of the first kind
$γ$ Gyromagnetic ratio (GHz T^{-1})
$g$ Lande g-factor
$B$ Magnetic Field (T)
$L/S$ Orbital/Spin quantum number
$μ_B$ Bohr magneton ($9.274 \times 10^{-24}$ A m^2)
$Γ$ Torque (N m, J rad^{-1})
$J$ Total angular momentum quantum number
$q$ Elementary charge $≈ 1.602... \times 10^{-19}$ C
$m_e$ Electron mass ($9.11 \times 10^{-31}$ kg)
$I$ Current (A)
\( \rho_r \)  Reflection coefficient
\( \rho_{sc} \)  Cooper pair density
\( \rho \)  Resistivity (\( \Omega \) m)
\( I_c \)  Critical Current (A)
\( N \)  Demagnetising tensor
\( K_{an} \)  First order magnetocrystalline anisotropy (J m\(^{-3}\))
\( M_{tot} \)  Total magnetisation (A m\(^{-1}\))
\( \omega_0 \)  Resonant frequency (Hz)
\( h \)  Alternating field component (RF) (A m\(^{-1}\))
\( \mu_0 H \)  Applied DC field (T)
\( H_{eff} \)  Effective Field (A m\(^{-1}\))
\( m \)  RF alternating component of magnetisation (A m\(^{-1}\))
\( \chi \)  Susceptibility
\( \Delta H \)  Resonance linewidth (field) (A m\(^{-1}\))
\( M_s \)  Saturation magnetisation (A m\(^{-1}\))
\( G \)  Gilbert damping coefficient
\( \alpha \)  Gilbert damping parameter
\( S_{xy} \)  Scattering parameter (dB)
\( n \)  Integer number
\( P \)  Power (W)
\( e_n \)  Electric voltage for cell edges (V)
\( b_n \)  Magnetic flux for cell faces (Wb, T m\(^2\))
\( e \)  Napier’s constant (\( \approx 2.71828 \))
\( \Phi \)  Magnetic flux (Wb, T m\(^2\))
\( n \)  Number of impurities (mol, ppm)
\( N_A \)  Avogadro’s number (6.022 \times 10^{23} \text{ mol}^{-1})
\( k_B \)  Boltzmann constant (1.3807 \times 10^{-23} \text{ J K}^{-1})
\( D \)  Size distribution (m)
\( A \)  Absorption
$R$  Reflection
$T$  Transmission
$D$  Pitch (spacing) (m)
$\delta$  Skin-depth (m)
$\sigma$  Conductivity (S m$^{-1}$)
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Chapter 1

Introduction and Literature Review

1.1 Introduction

Soft ferro/ferrimagnetic materials form the basis of most applications that require magnetic flux to be concentrated or directed, for example in transformers, generators, or on a smaller scale in circulators and isolators. They are used in a variety of applications, ranging from static to dynamic (from less than 1 Hz to hundreds of GHz) frequencies. At static or very low-frequencies they are primarily used for flux guidance as cores in electromagnets. Metallic cores can be used for flux-direction in transformers, generators, motors and inductors in the low frequency and audio frequency ranges but insulating ferrites are required for high frequency applications in the MHz to GHz ranges, in order to minimize Eddy-current losses that are associated with metallic materials. The operational frequency ranges for soft-magnets and examples of applications and materials are depicted in figure 1.1.

| Materials          | Applications                  
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<tbody>
<tr>
<td>Fe, Fe-Co</td>
<td>Electromagnets, Relays</td>
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<tr>
<td>Ni-Fe</td>
<td></td>
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<tr>
<td>Permalloy Foil,</td>
<td></td>
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<td>YIG Ferrite,</td>
<td></td>
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<tr>
<td>Mn-Ze Ferrite,</td>
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<td>Si Steel,</td>
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<td>Sendust (Fe-Si-Al)</td>
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<td>Li Ferrite</td>
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<td>Ni-Zn Ferrite</td>
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<td>Generators</td>
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<td>Phase shifters,</td>
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Figure 1.1: Frequency applications of soft magnetic materials. The region of interest is the microwave regime (> 1 GHz).

1.1.1 Frequency and Length Scales of Magnetic Characterisation Techniques

At low frequencies (< MHz), i.e. of the time scale of domain wall depinning, SQUID (superconducting quantum interference device) magnetometry or VSM (vibrating sample magnetometry) are all viable detection methods. At high-MHz frequencies, domain wall dynamics (propagation) is observed, typically using stroboscopic techniques. For
high frequencies in excess of one GHz, magnetisation processes involve precession of either electrons (EPR-electron paramagnetic resonance) or the magnetisation (FMR-ferro/ferrimagnetic resonance). The discussed effects and their corresponding frequency ranges are shown in figure 1.2a. The length scales of magnetic interactions is broad, ranging from Å to millimetre scales and generally each magnetic effect corresponding to the length scale will be treated using a different methodology. Figure 1.2b shows the sensitivity limits of some of the more commonly used magnetometry techniques and their typical probing volumes. Of the given examples, scanning SQUID magnetometry is the most sensitive, capable of detecting magnetisations as low as $10^{-12}$ Am$^2$.

![Figure 1.2: Frequency and length scales of magnetic interactions.](image)

With the recent drive towards device miniaturisation, increased operational speeds and in turn capability to access higher frequencies both on-chip and in integrated devices there has been an interest in designing devices with broadband microwave functionality. This develops a need to create techniques capable of probing the reaction of a device to a microwave stimulus, be it microwave conductivity or magnetisation dynamics in the RF/microwave regime. The primary criteria for such tests would be that they be non-destructive and have a suitable bandwidth for the effect being
measured. Ferromagnetic resonance (FMR) effects are typically probed with GHz frequency stimuli.

1.2 Thesis Layout

The work detailed in this thesis attempts to create and develop novel instrumentation to expand the frequency bandwidth of dynamic measurements and extend the techniques and methodologies to random, quasi-random and template nanostructures. The original concept of this research doctorate was to bridge the gap between the methods for thin film characterisation of synthetic thin film structures (e.g. microwave reflectometry, magnetometry) and typically scattering-based method used on random and pseudo-random self-organised structures (e.g. laser speckle scattering). Through this work the opportunity arose to participate in the development of a novel form of characterisation for examining magnetisation dynamics using SQUID (Superconducting QUantum Interference Device) magnetometry. This then became the core focus of this work due to the interest and novelty of the new characterisation technique. As such, the work described in this thesis focuses on two main areas, the first being broadband microwave reflectometry and specifically its application to random or templated nanostructures, which previously have been examined using DC conductivity measurements. This includes pulsed time domain reflectometry (TDR), continuous wave vector network analysis (VNA) in addition to the design, simulation and construction of high-frequency wave carriers in both planar and cavity geometries. The work done on high frequency waveguide design was pertinent in the creation of two novel methodologies for broadband detection of FMR, which rely on SQUID-magnetometry detection.

This chapter will discuss the underlying concepts that govern the treatment of high frequency wave propagation, namely transmission line theory and will describe some commonly used waveguide structures. Leading on from wave propagation, the theory of, and detection methods for, measuring ferromagnetic resonant absorption of electromagnetic energy will be discussed.

Chapter 2 will deal with the high-frequency instrumentation and simulation tools such as pulsed and continuous wave reflectometry techniques, magnetometry and optics
to name but a few, that are key aspects of this work. The more novel magnetisation
dynamics techniques will be elaborated on in later chapters.

In chapter 3 the main motivation behind and results of broadband microwave reflectometry techniques in analysing the electrical conductivity of randomly assembled nanostructures are discussed. Here TDR and VNA provide for a non-destructive analysis of networks consisting of either surface coated or native oxide passivated nanowires, where the conductivity depends on both the conductivity of the material and the density of the network. In a rather more potent form, the change of the conductivity under high power microwave stressing using bursts of CW is also evaluated. This is done in order to gain an understanding of the evolution of the transport-limited avalanche regime for wire-to-wire contacts and examine the transition from a resistive to an ‘activated’ state.

Cavity-based narrowband reflectometry is discussed in chapter 4, which includes the simulation and fabrication of high frequency cavity waveguides with operational bandwidths ranging from 7 GHz to 42 GHz. The primary application of such waveguides is illustrated in analysing the effectiveness of inkjet printed Ag films for electromagnetic interference (EMI) shielding applications. Measurements are established in three distinct frequency bands; X-Ku (7-14 GHz), K (15-20 GHz) and Ka (21-42 GHz).

As an extension of chapters 3 and 4 to higher frequencies and shorter wavelengths, the optical properties of both the nanowire networks and the inkjet-printed films are established in chapter 5. For the case of the printed films this involves high-visible-UV spectroscopy (densitometry), measuring both transmittance and reflectance. The geometric features, length scales and diameters of the nanowire networks are examined using the less well known contactless technique, laser specular scattering (LSS).

The design of high-frequency, large bandwidth, planar waveguides played a key role in the excitation of ferromagnetic resonance, which is discussed in chapter 6. In this chapter, two novel techniques for the detection of broadband (100 MHz - 20 GHz) ferro/ferrimagnetic resonance are introduced, which rely on SQUID-based gradiometry detection of small changes in the magnetisation. In the first method small changes in
the along-the-applied-field projection of the coupled magnetic moment ($\Delta m_z$) are detected as the material is driven into resonance. Absolute measurement of the moment along the applied field direction and the resonance induced lowering of this moment makes estimation of the precession cone angle easily accessible. The second method invokes the change in $\Delta m_z$ with the resonance-induced thermal heating ($\frac{dm_z}{dT}$). Measurement of the thermal derivative of the magnetisation, while not absolute, provides for high sensitivity FMR detection, which is especially sensitive at low temperatures. The inhomogeneous microwave excitation generated by the microstrip carrier allows for the observation of higher magnetostatic modes in addition to the primary spatially uniform mode and the tracking of very broad resonances at low static fields. The capability of both strategies are demonstrated in analysing the magnetisation dynamics over a broad range of experimental temperatures (4 K - 400 K) and fields (10 - 500 mT) in both polycrystalline and single-crystalline $Y_3Fe_5O_{12}$. The two SQUID-detection techniques when combined with broadband VNA-FMR, low-frequency magnetic susceptibility and DC magnetometry, all easily realised, essentially concurrently, greatly expand the amount of static and dynamic information accessible using a single module.

1.3 Theory and Literature Review

1.3.1 Microwave Propagation and Transmission Line Theory

As described in D. Pozar’s *Microwave Engineering* [1] the term transmission line is typically used to describe media that transmit primarily transverse electromagnetic (TEM) waves. While traditional circuit analysis deals with a situation where component sizes are far less than the wavelength of the electric field, in transmission line theory the component dimensions may be comparable in size or multiples of the wavelength. As a result, a lumped circuit analogy is typically employed in describing the electrical behaviour of a transmission line. As can be seen in figure 1.3, for a length $\Delta z$, the transmission line is schematically represented as two conducting lines and electrically represented in terms of a series of $R$, $L$, $C$ and $G$. Where $R$, $L$, $C$ and $G$ are the series resistance, inductance, shunt capacitance and conductance per unit length respectively. The general expression for the characteristic impedance $Z_0$ of a
transmission line is given in equation 1.1a where \( j \) is an imaginary unit and \( \omega \) is the angular frequency. According to this theory the ideal lossless transmission line can be represented by a lumped element equivalent circuit consisting of only inductors in series and parallel capacitors, assuming a negligible line resistance and no dielectric losses (i.e. \( R, G = 0 \)). Equation 1.1a then transforms to equation 1.1b.

\[
Z_0 = \sqrt{\frac{R + j\omega L}{G + j\omega C}} \quad (1.1a)
\]

\[
Z_0 = \sqrt{\frac{L}{C}}. \quad (1.1b)
\]

Figure 1.3: (a) Voltage and current for two line conductor schematic of transmission line. (b) Equivalent circuit for a lossy transmission line of finite length \( \Delta z \).

In the case where an incident wave encounters a discontinuity in the transmission line, part of the wave not originating at the load is reflected back toward the source and the characteristic and load impedance \( Z_L \) are no longer equal. Hence, the quality of a transmission line is characterised by the reflection coefficient \( \rho_r \) which is the ratio of reflected \( E_r \) to incident energy \( E_i \) when an electromagnetic wave passes through the transmission line and is given by

\[
\rho_r = \frac{E_r}{E_i} = \frac{Z_L - Z_0}{Z_L + Z_0}. \quad (1.2)
\]

For a transmission line with zero discontinuities, perfectly matched to the source this would be zero.
1.3.1.1 Transmission Line Geometries

Transmission line geometries come in many forms, for example planar, cavity or coaxial, each having its strengths in terms of bandwidth, frequency or field uniformity. One of the earliest forms of transmission lines are rectangular, hollow waveguides consisting of a single conductor filled with a homogeneous dielectric, most commonly air. Providing access to frequency bands ranging from 1 to 220 GHz, cavity waveguides are still indispensable in RF and microwave transmission, particularly in accessing frequency bands 20 GHz and beyond. As they consist of only a single conductor, they are capable of carrying either TE or TM mode waves but TEM mode propagation is not possible. Figure 1.4 shows a schematic of a typical metallic cavity waveguide where $a > b$, filled with a uniform material of dielectric permittivity $\varepsilon$ and magnetic permeability $\mu$.

\[ \begin{align*}
\text{Figure 1.4: Rectangular, hollow waveguide filled with a uniform material with dielectric permittivity } \varepsilon \text{ and magnetic permeability } \mu.
\end{align*} \]

For the case of simplicity, propagation of the transverse electric (TE\textsubscript{mn}) modes (all combinations of $m$ and $n$) will be discussed here. Transverse magnetic modes and derivations are included in the appendix A. The transverse electric modes are characterised by $E_z = 0$ and $H_z \neq 0$, where $H_z$ must satisfy the reduced wave equation

\[ \left( \frac{\partial^2}{\partial y^2} + \frac{k_z^2}{\varepsilon} \right) h_z(x, y) = 0 \] (1.3)
Where \( k_c = \sqrt{k^2 - \beta^2} \) is the cut-off wavenumber and \( H_z(x, y, z) = h_z(x, y)e^{-i\beta z} \). Using the separation of variables \( h_z(x, y) = X(x)Y(y) \), equation 1.3 reduces to

\[
\frac{1}{X} \frac{d^2X}{dx^2} + \frac{1}{Y} \frac{d^2Y}{dy^2} + k_c^2 = 0.
\] (1.4)

Each term within the equation must be equal to a constant as the first and second terms depend on only \( x \) and \( y \) respectively. The constants \( k_x \) and \( k_y \) are now introduced, where \( k_x^2 + k_y^2 = k_c^2 \). Therefore

\[
\frac{d^2X}{dx^2} + k_x^2 X = 0
\] (1.5a)

and

\[
\frac{d^2Y}{dy^2} + k_y^2 Y = 0
\] (1.5b)

A general solution for \( h_z \) is given by

\[
h_z(x, y) = (A \cos k_x x + B \sin k_x x)(C \cos k_y y + D \sin k_y y)
\] (1.6)

For a rectangular cavity, as shown in figure 1.4 the boundary conditions that the electric field components tangential to the cavity walls falls to zero. i.e.

\[
e_x(x, y) = 0 \text{ at } x = 0, b
\] (1.7a)

\[
e_y(x, y) = 0 \text{ at } y = 0, a
\] (1.7b)

hold. This can be used to determine the the constants \( A, B, C, D \) in the generalised equation 1.6. The transverse components \( e_x \) and \( e_y \) can be formulated as

\[
e_x = -\frac{j\omega \mu}{k_c^2} \frac{\partial H_z}{\partial y} = -\frac{j\omega \mu}{k_c^2} (A \cos k_x x + B \sin k_x x)(-C \cos k_y y + D \sin k_y y) = 0 \] (1.8a)

\[
e_y = -\frac{j\omega \mu}{k_c^2} \frac{\partial H_z}{\partial x} = -\frac{j\omega \mu}{k_c^2} (-A \cos k_x x + B \sin k_x x)(C \cos k_y y + D \sin k_y y) = 0. \] (1.8b)
Therefore \( D, B = 0, k_y = \frac{m\pi}{b} \) and \( k_x = \frac{n\pi}{a} \), such that the solution to \( H_z(x, y, z) \) is given by

\[
H_z(x, y, z) = F \cos \frac{m\pi x}{a} \cos \frac{n\pi y}{b} e^{-j\beta z}
\]  

(1.9)

where \( F \) is a constant consisting of the two constants \( A \) and \( C \). The transverse field components \( H_x, H_y, E_x \) and \( E_y \) are described by

\[
H_x = \frac{-j\beta}{k^2c} \frac{\partial H_z}{\partial x} = \frac{j\beta m\pi}{k^2c a} F \sin \frac{m\pi x}{a} \cos \frac{n\pi y}{b} e^{-j\beta z}
\]  

(1.10a)

\[
H_y = \frac{-j\beta}{k^2c} \frac{\partial H_z}{\partial y} = \frac{j\beta n\pi}{k^2c b} F \cos \frac{m\pi x}{a} \sin \frac{n\pi y}{b} e^{-j\beta z}
\]  

(1.10b)

\[
E_x = \frac{-j\omega\mu}{k^2c} \frac{\partial H_z}{\partial y} = \frac{j\omega\mu n\pi}{k^2c b} F \cos \frac{m\pi x}{a} \sin \frac{n\pi y}{b} e^{-j\beta z}
\]  

(1.10c)

\[
E_y = \frac{j\omega\mu}{k^2c} \frac{\partial H_z}{\partial x} = \frac{-j\omega\mu m\pi}{k^2c a} F \sin \frac{m\pi x}{a} \cos \frac{n\pi y}{b} e^{-j\beta z}
\]  

(1.10d)

where \( \beta \) is the propagation constant and can be represented as

\[
\beta = \sqrt{k^2 - k_c^2} = \sqrt{k^2 - \left(\frac{m\pi}{a}\right)^2 - \left(\frac{n\pi}{b}\right)^2}.
\]  

(1.11)

Each TE mode will have a cut-off frequency below which the mode will not propagate. Cavity waveguides operate in finite frequency bands bound by upper and lower cut-off frequencies. The cut-off frequency is the frequency beyond which the transmitted signal intensities are strongly attenuated and relates to the cut-off wavefunction \( k_c \), which for a rectangular cavity can be written in terms of the cavity dimensions \( a \) and \( b \) such that

\[
k_c = \sqrt{\left(\frac{m\pi}{a}\right)^2 + \left(\frac{n\pi}{b}\right)^2}
\]  

(1.12)

The cut-off frequency can be then represented by

\[
f_{mn} = \frac{k_c}{2\pi\sqrt{\mu\varepsilon}} = \frac{1}{2\pi\sqrt{\mu\varepsilon}} \sqrt{\left(\frac{m\pi}{a}\right)^2 + \left(\frac{n\pi}{b}\right)^2}
\]  

(1.13)

The dominant mode is that which gives the lowest cut-off frequency, which for the case of the rectangular cavity waveguide is the TE_{10} (i.e. \( m = 1 \) \( n = 0 \)). The cut-off
frequency for the dominant TE\textsubscript{10} mode is therefore given by

\[ f_{10} = \frac{1}{2a\sqrt{\mu\varepsilon}} \]  

(1.14)

and relates to the width, \( a \) of the rectangular waveguide. TE and TM modes with non-zero cut-off frequencies exist, which limit the operational bandwidth of the cavity waveguide. The secondary lowest modes are TE\textsubscript{01} and TE\textsubscript{20} where \( f_{01} = \frac{1}{2b\sqrt{\mu\varepsilon}} \) and \( f_{20} = \frac{1}{a\sqrt{\mu\varepsilon}} \). Assuming that \( b \leq a \) it follows that \( f_{10} \leq f_{01} \). For the case of \( b \leq \frac{a}{2} \), then \( \frac{1}{a} \leq \frac{1}{2b} \), such that \( f_{20} \leq f_{01} \). On the other hand if \( \frac{a}{2} < b \leq a \) then \( f_{01} < f_{20} \).

The two conditions described are represented graphically in figure 1.5 which shows that in order to maximise the operational bandwidth, \( b \leq \frac{a}{2} \) and the widest bandwidth is achieved when \( b = \frac{a}{2} \) (Image adapted from [2]). The bandwidth interval is then defined by \([f_{10}, 2f_{10}]\). Most commercially available waveguides are designed such that \( b = \frac{a}{2} \) in order to maximise the available bandwidth.

![Figure 1.5: Two conditions for cavity geometries and their corresponding operational bandwidth.](Image)

With the constant drive toward device miniaturization and integration of RF and microwave circuitry, planar transmission line geometries have overtaken their hollow rectangular predecessors. What they lack in terms of field homogeneity, they make up for in terms of minimal size, ease of fabrication and broadband transmission capabilities. While microwave conductivity analysis using cavities typically involves sets of discrete frequency band waveguides, a single planar geometry guide can have bandwidths in the 10s of GHz and beyond, provided the dielectric materials used are non-dispersive. This work primarily uses broadband planar transmission lines, in either microstrip (figure 1.6a) or coplanar-waveguide (CPW) (figure 1.6b) configuration.
In the microstrip geometry depicted in Fig. 1.6a the majority of the field lines are contained within the dielectric region between the signal conductor and the ground plane but some are in the air above the dielectric, as depicted on the right hand side of figure 1.6a. As the field is not contained within a single homogeneous material and the phase velocities in air ($v_{ph} \approx c$) and the dielectric of relative permittivity $\varepsilon_r$ ($v_{ph} \approx \frac{c}{\sqrt{\varepsilon_r}}$) are different, wave propagation will possess a longitudinal component and therefore cannot be considered to be a purely TEM mode. This is referred to as quasi-TEM mode propagation.

Similarly, coplanar-waveguides also possesses quasi-TEM mode propagation. The primary advantage of the CPW over the microstrip geometry is that active and passive (ground) lines are contained within the same plane and shunting active components does not require drilling vias, which can be a difficult task with brittle substrates such as alumina. Also, the electric field distribution as depicted in the right hand side of Fig. 1.6b is such that a larger proportion of the field lines are in the air above the dielectric, making it an ideal candidate for contactless electrical conductivity measurements, where the sample is placed at the surface of the CPW.

In 1965 Wheeler [3] introduced a method of computing the characteristic impedance
of the microstrip based on conformal mapping of the microstrip geometry, resulting in a
simplified model of the microstrip as a planar waveguide where the top and bottom are
electrical conductors separated by a dielectric and the sides are magnetic conductors.
The waveguide will have an effective width ($W$) and height ($H$) defined in terms of the
dimensions of the microstrip

$$W = w + \frac{t}{\pi} \left[ \ln \left( \frac{2h}{t} \right) + 1 \right]$$  \hspace{1cm} (1.15a)

$$H = h - 2t.$$  \hspace{1cm} (1.15b)

For ($\frac{W}{H}$) < 1 approximations for the effective dielectric constant $\varepsilon_{eff}$ and characteristic
impedance $Z_0$ are given by

$$\varepsilon_{eff} = \frac{\varepsilon_r + 1}{2} + \frac{\varepsilon_r - 1}{2} \left[ \frac{1}{\sqrt{1 + 12 \left( \frac{H}{W} \right) \left( \frac{W}{H} \right)^2}} + 0.04 \left( 1 - \left( \frac{W}{H} \right)^2 \right) \right]$$  \hspace{1cm} (1.16a)

$$Z_0 = \frac{60}{\sqrt{\varepsilon_{eff}}} \ln \left( 8 \left( \frac{H}{W} \right) + 0.25 \left( \frac{W}{H} \right) \right).$$  \hspace{1cm} (1.16b)

For the case of ($\frac{W}{H}$) > 1

$$\varepsilon_{eff} = \frac{\varepsilon_r + 1}{2} + \frac{\varepsilon_r - 1}{2} \left[ \frac{1}{\sqrt{1 + 12 \left( \frac{H}{W} \right)}} \right]$$  \hspace{1cm} (1.17a)

$$Z_0 = \frac{120}{\sqrt{\varepsilon_{eff}}} \left[ \frac{W}{H} + 1.393 + 0.667 \ln \left( \frac{W}{H} + 1.444 \right) \right].$$  \hspace{1cm} (1.17b)

In reality this is a rather rough approximation and a more comprehensive analysis of
the line’s impedance can be done by measuring the incident and reflected energy $E_i$
and $E_r$ at a point using a microwave reflectometer, using the reference characteristic
impedance $Z_0 = 50 \Omega$ and rearranging equation 1.2

$$Z_L = \frac{E_i Z_0 + E_r Z_0}{E_i - E_r}.$$  \hspace{1cm} (1.18)

By knowing the group velocity $v_g$ and the time delay $\Delta t$ i.e. the transit time from the
Introduction and Literature Review

1.3 Pulse launch to the end of the media and back again, the length of the transmission media may also be determined:

\[ L = \frac{v_g \Delta t}{2}. \]  

(1.19)

The factor of \( \frac{1}{2} \) accounts for the fact that the pulse traverses the media twice.

The characteristic impedance and effective dielectric constant for a CPW depends largely on the width of the central conductor \( S \), the pitch between the signal line and each ground plane \( W \) and the dielectric thickness \( H \). The dielectric thickness \( H \) does not govern the characteristic impedance, provided \( H \) is greater than the pitch between the signal line and each ground plane \( W \). This can be advantageous for double sided board geometries and high frequency applications. The effective dielectric constant is defined as [4]

\[ \varepsilon_{eff} = 1 + \frac{\varepsilon_r - 1}{2} \frac{K(k')K(k_1)}{K(k)K(k'_1)} \]  

(1.20)

where \( k \) and \( k_1 \) are related to \( S, W \) and \( H \) and are given by

\[ k = \frac{S}{S + 2W} \]  

(1.21a)

\[ k_1 = \frac{\sinh \left( \frac{\pi S}{4H} \right)}{\sinh \left( \frac{(S+2W)\pi}{4\pi} \right)} \]  

(1.21b)

\( K \) is the elliptical integral of the first kind and \( k' = \sqrt{1 - k^2} \). Approximate values for the ratio of the complete elliptical integrals are given by Hilberg [5]:

\[ \frac{K(k)}{K(k')} = \begin{cases} \frac{1}{2\pi} \ln \left[ \frac{2\sqrt{1+k}+\sqrt{4k}}{\sqrt{1+k}-\sqrt{4k}} \right] & \text{for } 1 \leq \frac{k}{k'} \leq \infty, \frac{1}{\sqrt{2}} \leq k \leq 1 \\ \frac{2\pi}{\ln \left[ \frac{2\sqrt{1+k}+\sqrt{4k}}{\sqrt{1+k}-\sqrt{4k}} \right]} & \text{for } 0 \leq \frac{k}{k'} \leq 1, 0 \leq k \leq \frac{1}{\sqrt{2}} \end{cases} \]  

(1.22)

The characteristic impedance is calculated as

\[ Z_0 = \frac{30\pi}{\sqrt{\varepsilon_{eff}}} \frac{K(k')}{K(k)} \]  

(1.23)
A brief discussion of some other widely used transmission line geometries are given in appendix B.

1.3.2 Ferromagnetic Resonance (FMR)

One of the main uses of both planar and cavity transmission lines in magnetics research is in the excitation and detection of resonance effects in magnetic media, namely nuclear magnetic resonance (NMR), electron paramagnetic resonance (EPR) and ferro/ferrimagnetic resonance (FMR).

1.3.2.1 On the Theory of Ferromagnetic Resonance

Magnetic resonance occurs when the energy levels of a quantised system of moments are Zeeman split by a uniform static magnetic field \( B_0 \) and the system absorbs energy from a transverse AC magnetic field at sharply defined frequencies which correspond to transitions between the energy levels [6]. The moment may take the form of an isolated electron, as is the case for EPR (otherwise known as electron spin resonance ESR), a nuclear spin as in NMR or the coupled electrons resulting in a net magnetisation as for FMR. This occurs at the Larmor frequency

\[
\omega_0 = \gamma B_0
\]  

(1.24)

where \( \omega_0 = 2\pi f_L \) and \( \gamma \) is known as the gyromagnetic ratio, the ratio of magnetic moment \( \mathbf{M} \) to angular moment \( \mathbf{J} \). For the case of EPR this is given by

\[
\gamma = \frac{gq}{2m_e} = \frac{g\mu_B}{\hbar}.
\]  

(1.25)

Here \( g \) represents the Lande’ g-factor, a dimensionless value resulting from the first order energy perturbation of an atom under the influence of a weak magnetic field and \( q, m_e, \mu_B \) and \( \hbar \) are electron charge, mass, the Bohr magneton (\( \mu_B = 9.274 \times 10^{-24} \text{ Am}^2 \)) and the reduced Planck constant respectively. For paramagnetic ions, the value of \( g \)
may be determined using:

\[ g = \frac{3}{2} + \frac{S(S + 1) - L(L + 1)}{2J(J + 1)} \]  

(1.26)

where \( L \) and \( S \) are the orbital and spin quantum numbers respectively.

Figure 1.7: Conventional axis system used. Where \( B_0 \) is the magnetic field \( B_0 = \mu_0 H \) and \( B_1 \) is the applied magnetic field oscillating at microwave frequencies.

Figure 1.7 depicts the conventional axis system employed, where a sample placed in a DC field \( B_0 \) is subjected to a perpendicular AC field \( B_1 \). Ferromagnetic resonance was first reported in 1946\(^1\) by Griffiths who observed absorption at specific microwave frequencies for thin-films of Fe, Ni and Co [8]. Taking FMR the resonant body is taken as the entire coupled magnetic moment. If a ferromagnetic material with moment \( M \) is placed in an externally applied magnetic field \( B = \mu_0 H \), \( \mu_0 \) being vacuum permeability, the moments will tend to align parallel to the field, such that \( M \times H = 0 \) as this is the lowest energy configuration. In order to do so the magnetic moment experiences a torque \( \Gamma = M \times \mu_0 H \). The following equation

\[ M = \frac{gq}{2m_e} J \]  

(1.27)

relates the moment \( M \) to the total angular momentum \( J \) for a charged particle. Substituting equation 1.25, and knowing that \( \Gamma = \frac{dJ}{dt} \), it follows that the equation of motion

\[ \frac{dM}{dt} = \gamma \mu_0 M \times H \]  

(1.28)

\(^1\)It may have been demonstrated in years prior but was not disclosed due to Griffith’s efforts in WWII. [7].
describes the precession about the magnetic field direction in the absence of damping. Magnetisation precession excluding the effects of damping is depicted in figure 1.8a, where the magnetisation vector precesses about the direction of \( \mathbf{H} \) at an angle \( \theta \), commonly known as the precession cone angle.

![Figure 1.8](image)

**Figure 1.8:** (a) Precession of the magnetisation vector in a magnetic field \( \mathbf{H} \). (b) Precession of magnetisation in a magnetic field including the effects of damping.

The effects of relaxation need to be accounted for. The Landau-Lifschitz-Gilbert [9, 10] equation

\[
\frac{d\mathbf{M}}{dt} = \gamma \mu_0 \mathbf{M} \times \mathbf{H}_{\text{eff}} - \left( \frac{\alpha}{M_s} \right) \mathbf{M} \times \frac{d\mathbf{M}}{dt}
\]

(1.29)
describes the damped precession of the moment about the effective magnetic field \( \mathbf{H}_{\text{eff}} \), described by

\[
\mathbf{H}_{\text{eff}} = \mathbf{H} + H_{\text{an}} + H_{\text{demag}}
\]

(1.30)

for a ferromagnetic material, accounting for the external applied field \( \mu_0 \mathbf{H} \), magnetocrystalline anisotropy \( \mu_0 H_{\text{an}} \) and the demagnetising field \( \mu_0 H_{\text{demag}} \). The magnetocrystalline anisotropy field is given as \( H_{\text{an}} = \frac{2K_{\text{an}}}{\mu_0 M_s} \) for uniaxial magnetocrystalline anisotropy, where \( K_{\text{an}} \) is the first order magnetocrystalline anisotropy term. Here \( \mathbf{M} \) has a magnitude \( M_s \) and precesses around the direction of \( \mathbf{H} \). \( \alpha \) is the Gilbert damping parameter, a dimensionless sample dependent phenomenological value, typically of the order \( \alpha = 0.01 \) for thin ferromagnetic films. The damping term on the right hand side causes the magnetisation to spiral toward the direction of \( \mathbf{H} \), as depicted in figure 1.8b.
If an alternating magnetic field \( h = h_0 \cos \omega t \) is applied perpendicular to the direction of the saturating field \( H \), the torque modifies to \( \Gamma = M \times \mu_0(H_{\text{eff}} + h) \) hence equation 1.28 transforms to \( \frac{dM}{dt} = \gamma \mu_0 M \times (H_{\text{eff}} + h) \). The magnetisation will also possess a steady and alternating part, such that \( M_{\text{tot}} = M + m \). If one considers the case where the easy axis of the material lies along the applied field direction \( x \), then the magnetisation and field vectors take the form

\[
H = \begin{pmatrix} H \\ 0 \\ 0 \end{pmatrix} \quad \quad h = \begin{pmatrix} 0 \\ h_y \\ 0 \end{pmatrix} \quad \quad (1.31a)
\]

\[
M = \begin{pmatrix} M \\ 0 \\ 0 \end{pmatrix} \quad \quad m = \begin{pmatrix} m_x \\ m_y \\ m_z \end{pmatrix} \quad \quad (1.31b)
\]

Therefore in the absence of the RF field \( H \) and \( M \) are in the same direction and \( M = M_s \). If the demagnetising tensors \( N_x, N_y \) and \( N_z \) are taken as diagonal then the demagnetising field may be described by

\[
\mu_0 H_{\text{demag}} = -\mu_0 N_x M_s \hat{x} - \mu_0 N_y m_y \hat{y} - \mu_0 N_z m_z \hat{z} \quad \quad (1.32)
\]

where \( \hat{x}, \hat{y} \) and \( \hat{z} \) are unit vectors. Assuming that the RF field is perpendicular to the applied DC field (i.e. precession is in the \( y - z \)-plane) then the effective field can be written in terms of the DC and RF components of field and magnetisation and the demagnetising tensors as

\[
H_{\text{eff}} = H + h + H_{\text{an}} - \mu_0 N_x M_s \hat{x} - \mu_0 N_y m_y \hat{y} - \mu_0 N_z m_z \hat{z} \\
= (H + H_{\text{an}} - N_x M_s) \hat{x} + (h - N_y m_y) \hat{y} - N_z m_z \hat{z}. \quad \quad (1.33)
\]

\( H_{\text{an}} \) contains all anisotropy besides the demagnetisation anisotropy field. The leading term of \( H_{\text{an}} \) is the magnetocrystalline anisotropy field. The cumulative magnetisation is described by

\[
M_{\text{tot}} = M_s \hat{x} + m_y \hat{y} + m_z \hat{z}. \quad \quad (1.34)
\]


Substituting equations 1.33 and 1.34 into equation 1.29 the equations of motion may be written as

\[
\frac{dm_x}{dt} = 0 = -\gamma \mu_0 (-m_y N_z m_z - m_z (h - N_y m_y)) + \frac{\alpha}{M_s} (m_y \frac{dm_z}{dt} - m_z \frac{dm_y}{dt}) \tag{1.35a}
\]

\[
\frac{dm_y}{dt} = -\gamma \mu_0 (m_x (H + H_{an}) - M_s N_x m_z) + \alpha \frac{dm_z}{dt} \tag{1.35b}
\]

\[
\frac{dm_z}{dt} = -\gamma \mu_0 (M_s (h - N_y m_y) - m_y (H + H_{an} - N_x M_s)) + \alpha \frac{dm_y}{dt} \tag{1.35c}
\]

The generalised linear form of equation 1.29 is described by

\[
\frac{dM}{dt} = \gamma \mu_0 (m \times H_{eff}^* + M_{tot} \times h) \tag{1.36}
\]

Where \(H_{eff}^* = H_{eff} - h\). To simplify equations 1.35, two new terms are defined as

\[
\omega_M = \gamma \mu_0 M_s \tag{1.37a}
\]

and

\[
\omega_H = \gamma \mu_0 (H + H_{an}). \tag{1.37b}
\]

Substituting 1.37 and neglecting higher order terms of \(h\) and \(m\) and considering the time variation of \(m \propto \exp^{-i\omega t}\) equations 1.35 are reduced to

\[
0 = -i\omega m_y + (\omega_H + (N_z - N_x)\omega_M - i\alpha \omega) m_z \tag{1.38a}
\]

\[
\omega_M h = i\omega m_z + (\omega_H + (N_y - N_x)\omega_M - i\alpha \omega) m_y. \tag{1.38b}
\]

This may be written in matrix form relating \(h\) to \(m\) as

\[
\begin{pmatrix} 0 \\ h \end{pmatrix} \omega_M = \begin{pmatrix} -i\omega & \omega_H + (N_z - N_x)\omega_M - i\alpha \omega \\ \omega_H + (N_y - N_x)\omega_M - i\alpha \omega & -i\omega \end{pmatrix} \begin{pmatrix} m_y \\ m_z \end{pmatrix} \tag{1.39}
\]

The dynamic susceptibility tensor \(\hat{\chi}\) is defined as the response of the RF magnetisation to the alternating RF field given as
\( \mathbf{m} = \hat{\chi} \mathbf{h} \). \hfill (1.40)

To obtain solutions for \( \chi \) the matrix given in equation 1.39 must be inverted. Doing so yields

\[
\hat{\chi} = \begin{pmatrix}
\chi_{yy} & i\chi_{yz} \\
-i\chi_{zy} & \chi_{zz}
\end{pmatrix}
\hfill (1.41)
\]

where the elements of the matrix are described by

\[
\chi_{yy/zz} = \frac{\omega_0 M}{\omega_0^2 - \omega^2(1 + \alpha^2) - i\alpha\omega}[2\omega H + (N_y + N_z - 2N_x)\omega M]
\hfill (1.42a)
\]

and

\[
\chi_{yz/zy} = \frac{\omega_0 M}{\omega_0^2 - \omega^2(1 + \alpha^2) - i\alpha\omega}[2\omega H + (N_y + N_z - 2N_x)\omega M]
\hfill (1.42b)
\]

where \( \omega_0 \) is defined as the resonant frequency. The general expression for the frequency at which a ferromagnet’s moment resonates is given by Kittel’s Law [11]. Considering the cumulative demagnetising factors \( N_x, N_y \) and \( N_z \), \( H_{eff} = H + H_{an} + NM \) Kittel’s Law takes the form

\[
\omega_0 = \mu_0 \gamma[(H + (N_z - N_x)M + H_{an})(H + (N_y - N_x)M + H_{an})]^\frac{1}{2}
\hfill (1.43)
\]

for an applied DC field along the \( x \) direction. Table 1.1 represents the demagnetisation tensors and corresponding resonance conditions for different shaped ferromagnetic specimen.

<table>
<thead>
<tr>
<th>Shape</th>
<th>( N_x )</th>
<th>( N_y )</th>
<th>( N_z )</th>
<th>( \omega_0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Sphere</td>
<td>( \frac{1}{3} )</td>
<td>( \frac{1}{3} )</td>
<td>( \frac{1}{3} )</td>
<td>( \gamma \mu_0 \mathbf{H} )</td>
</tr>
<tr>
<td>Needle z long axis (parallel)</td>
<td>( \frac{1}{2} )</td>
<td>( \frac{1}{2} )</td>
<td>0</td>
<td>( \gamma \mu_0 (\mathbf{H} + \frac{M}{2}) )</td>
</tr>
<tr>
<td>Needle z long axis (perpendicular)</td>
<td>( \frac{1}{2} )</td>
<td>0</td>
<td>( \frac{1}{2} )</td>
<td>( \gamma \mu_0 [(\mathbf{H}(\mathbf{H} - \frac{M}{2})]^\frac{1}{2} )</td>
</tr>
<tr>
<td>Thin-film (x-z plane) (parallel)</td>
<td>0</td>
<td>1</td>
<td>0</td>
<td>( \gamma \mu_0 [\mathbf{H}(\mathbf{H} + \mathbf{M})]^\frac{1}{2} )</td>
</tr>
<tr>
<td>Thin-film (x-z plane) (perpendicular)</td>
<td>0</td>
<td>0</td>
<td>1</td>
<td>( \gamma \mu_0 [\mathbf{H}(\mathbf{H} + \mathbf{M})] )</td>
</tr>
</tbody>
</table>
\( \hat{\chi} \) is complex can be represented as the summation of the real and imaginary contributions to the susceptibility (i.e. \( \hat{\chi} = \chi' + i\chi'' \)). The real and imaginary terms are considered dispersive and dissipative in nature, respectively. It was previously assumed that the RF field was applied along the y-axis, therefore \( \chi' = \chi'_{yy} \) and \( \chi'' = \chi''_{yy} \) and are given by

\[
\chi'_{yy} = \frac{\omega_M (\omega_H + (N_z - N_x)\omega_M)(\omega_0^2 - \omega^2)}{[\omega_0^2 - \omega^2]^2 + \alpha^2 \omega^2 [2\omega_H + (N_y + N_z - 2N_x)\omega_M]^2}
\] (1.44a)

and

\[
\chi''_{yy} = \frac{\alpha \omega M [(\omega_0^2 - \omega^2) + (\omega_H + (N_z - N_x)\omega_M)(2\omega_H + (N_y + N_z - 2N_x)\omega_M)]}{[\omega_0^2 - \omega^2]^2 + \alpha^2 \omega^2 [2\omega_H + (N_y + N_z - 2N_x)\omega_M]^2}
\] (1.44b)

where it is assumed that the Gilbert damping parameter \( \alpha \) is small and only linear terms are considered. Taking \( \alpha = 0.01, \gamma = 28 \text{ GHz/T} \) and \( \omega_0 = 3 \text{ GHz} \) for an applied field of 50 mT equations 1.44 can be used to graphically represent the real \( \chi'_{yy} \) and imaginary \( \chi''_{yy} \) parts of the susceptibility as in figure 1.9.

![Figure 1.9](image)

**Figure 1.9:** The real \( \chi'_{yy} \) and imaginary \( \chi''_{yy} \) parts of the susceptibility. The imaginary contribution shows the characteristic Lorentzian profile whereas the real part is antisymmetric about the resonant frequency.

As previously stated the real part is dispersive and possesses an asymmetric profile,
whereas the imaginary contribution is dissipative and depicts the amplitude of the precession in the form of a Lorentzian lineshape. It is typically the imaginary contribution $\chi'' = \chi''_{yy}$ that is probed in an FMR experiment, via monitoring microwave losses as a function of a swept field.

1.3.2.2 Origin of Ferromagnetic Resonance Linewidth

In analysing novel materials for spintronic devices it is important to estimate and gain an understanding of the relaxation mechanisms at play that result in linewidth broadening. The magnitude of the linewidth (the full-width-half-maximum of the resonant absorption peak) and its change with temperature variance can tell a lot about crystallinity and quality of magnetic materials. The field linewidth $\Delta H$ is typically described as the sum of two contributions, a homogeneous broadening $\Delta H_{\text{hom}}$ and an inhomogeneous $\Delta H_{\text{inhom}}$, such that $\Delta H = \Delta H_{\text{hom}} + \Delta H_{\text{inhom}}$. $\Delta H_{\text{hom}}$ arises due to intrinsic magnetisation damping mechanisms, whereas the $\Delta H_{\text{inhom}}$ contribution is due to sample inhomogeneities and imperfections such as defects, porosity, polycrystallinity or surface roughness. For the case of angular independent broadening, such as that seen when static magnetisation and the external applied magnetic field are in the plane of a film $\Delta H_{\text{hom}}$ may be expressed as

$$\Delta H_{\text{hom}} = \frac{4\pi \alpha f}{\mu_0 \gamma}$$

therefore

$$\Delta H = \Delta H_{\text{inhom}} + \frac{4\pi \alpha f}{\mu_0 \gamma}$$

(1.46)

where $\alpha$ is the Gilbert damping parameter. In this case, line width broadening is said to be dominated by magnetic inhomogeneities and is therefore an extrinsic effect. For the case of polycrystalline specimen, which exhibit much broader linewidths than their single-crystal counterparts, the inhomogeneous contribution to linewidth broadening can be said to originate from two main sources, anisotropy of the grains and porosity [12, 13] such that $\Delta H_{\text{inhom}} = \Delta H_{\text{anisotropy}} + \Delta H_{\text{porosity}}$. $\Delta H_{\text{anisotropy}}$ arises due the fact that each microscopic crystalline grain will have a different resonant field (or frequency)
due to the random orientation of the anisotropy axes for each grain [15]. This term is given by [16]

$$\Delta H_{\text{anisotropy}} = 1.08 \frac{H_{\text{an}}^2}{4\pi M} J$$

(1.47)

where $4\pi M$ is the saturation magnetisation and can be written as $M_s$ and $J$ is a shape factor, which for a sphere is given by

$$J = \sqrt{\frac{3}{2}} H_0 - \frac{H_0 M_s}{3} + \frac{5}{12} \left( \frac{M_s}{3} \right)^2$$

(1.48)

and relates to the resonant field $H_0$, the applied field $H$ and the saturation magnetisation [17].

Schlömann provides a detailed discussion for anisotropy induced broadening for two cases [18]. The first takes an independent grain approximation approach where each grain is taken as resonating independently from surrounding grains. The second approach considers strong coupling between grains in the material. For the case of weak coupling, randomly orientated grains are considered to resonate independently and resonant absorption is essentially proportional to the number of grains, whose resonant field lies within an interval $H$ to $H + \Delta H$. In the strong coupling case, where $M_s > H_{\text{an}}$, it is assumed that non-uniform spinwave modes exist. If the wavelength of the spinwaves is comparable to or greater than the grain size, energy will be transferred from the normal homogeneous resonance mode into spinwave modes with the same frequency as the normal mode, which results in linewidth broadening. The degree of broadening, thus depends largely on the number of spinwaves which are degenerate with the homogeneous resonant mode. This strong coupling term is generally referred to as ‘two-magnon scattering.’

The contribution of porosity to the inhomogeneous broadening factor in polycrystalline materials is widely discussed [12, 13, 15, 17]. Inhomogeneous line broadening is said to be attributed to demagnetisation fields created by pores at grain boundaries, where the width of the broadening is directly related to the fluctuation of the static magnetic field resulting from the demagnetising field of a spherical void. One approach to computing the field distribution is to subdivide the sample into equal volumes $V$, ...
each containing a single pore of volume $v$ [17], which results in a linewidth broadening given by

$$\Delta H_{\text{porosity}} = 0.5024\pi M_p J$$

(1.49)

where $p$ is the porosity given by $p = v/V$. Pores may also appear in the form of secondary phases within the material. Surface roughness is another factor which contributes to linewidth broadening, in a similar fashion to the porosity. For the case of spherical samples the quality of the surface polishing strongly effects the resonance linewidth.

Temperature is another important factor that governs the ferromagnetic linewidth and once again the broadening will have both intrinsic and extrinsic components. The intrinsic temperature dependent linewidth in yttrium iron garnet ($\text{Y}_3\text{Fe}_5\text{O}_{12}$ YIG) was originally described by Kasuya and LeCraw [19] by a two-magnon process, whereby a relaxing magnon couples to a phonon and provided the thermal energy $k_B T$ is greater than the energy of the phonon and magnons involved in the process ($T > 100$ K), the linewidth is directly proportional to the temperature. This theory, which predicts linewidth broadening with increasing temperature is only valid at high temperature ($T > 150$ K). At low temperatures extrinsic, impurity related broadening effects dominate [20, 21]. At temperatures below 150 K, the linewidth will increase with decreasing temperature with a peak between 40-150 K, below which the linewidth will again decrease. This is shown to be the case for non-S-state rare-earth ion impurities, except europium. Rare-earth ions with large orbital momentum will exchange-couple with iron ions leading to an extra relaxation mechanism for the normal precessional mode. A secondary source of impurity resultant broadening is the presence of Fe$^{2+}$ or Fe$^{4+}$ ions formed by the addition of tetravalent or divalent impurities to the crystal lattice. Such ions can lead to charge transfer conduction of either p-type or n-type respectively. The damping coefficient in YIG is strongly dependent on the impurity level, as the charge transfer conduction between iron ions provides for an extra relaxation mechanism known as valence exchange relaxation [22, 23]. This manifests in YIG as increased damping at low temperatures (around liquid nitrogen temperatures) and
therefore increased linewidth broadening.

### 1.3.2.3 Magnetostatic Modes in a Spherical Sample

For a ferromagnetic sample in an inhomogeneous RF excitation field, such as those created by a planar waveguide, higher magnetostatic modes in addition to the the spatially uniform primary mode exist. Non-uniform modes may also be excited in a uniform static field due to inhomogeneity in the internal fields such as those created by magnetostriction, crystal inhomogeneities, lattice distortions or surface imperfections [24]. When coupled to the primary mode, higher modes contribute to lineshape distortion or asymmetry or multiplicity of resonant absorption peaks supplementary to the primary peak described by Kittel dynamics (equation 1.43). Field-inhomogeneity-induced modes are examined experimentally by White and Solt by analysing the effect of the sample position within a resonant cavity on the resonant absorption spectra [25]. Mercereau and Feynman provide calculations for some of the simpler higher order modes in a sphere [26]. A generalised theory for all magnetostatic modes in a spherical specimen is provided by Walker [27] and expanded by Fletcher and Bell for a sphere [28]. Fletcher and Bell explicitly tabulate the primary features of 33 observable modes (63 are observed but 33 are formally identified), such as the frequency dependence of the resonant field, the magnetic potential and the RF moment distribution for each mode, as well as the saturation magnetisation.

The calculations described here are based on those first detailed by Walker. The distribution of the magnetisation in a spherical sample can be formulated by neglecting propagation and assuming Maxwell’s magnetostatic conditions hold and are given by

$$\nabla \times \mathbf{H} = 0 \text{ and } \nabla \cdot \mathbf{B} = 0.$$  \hspace{1cm} (1.50)

This implies that $\mathbf{H}$ can be described by a gradient of a magnetic scalar potential $\psi$

$$\mathbf{H} = \nabla \psi$$  \hspace{1cm} (1.51)
Assuming that the sample is a spheroid with the axis of symmetry lying along the applied field direction $z$, the ratio of the longitudinal axis $b$ to the transverse axis $a$ is given by $\alpha$ ($\alpha = b/a$). The spherical coordinates $\xi$, $\mu$, and $\phi$ are now introduced and can be represented as

\begin{align*}
    x(\xi, \mu, \phi) &= \sqrt{a^2 - b^2} \sqrt{1 + \xi^2} \sqrt{1 - \mu^2} \cos(\phi) \\
y(\xi, \mu, \phi) &= \sqrt{a^2 - b^2} \sqrt{1 + \xi^2} \sqrt{1 - \mu^2} \sin(\phi) \\
z(\xi, \mu, \phi) &= \sqrt{a^2 - b^2} \xi \mu.
\end{align*}

The surface of a prolate spheroid in Cartesian coordinates is given by

\[ \frac{x^2}{a^2} + \frac{y^2}{a^2} + \frac{z^2}{b^2} = 1 \] (1.53)

and

\[ \xi = \xi_0 \text{ where } \xi_0 = \sqrt{\frac{\alpha^2}{1 - \alpha^2}} \] (1.54)

Solutions to the Legendre functions for the potential $\psi$ take the form of

\[ \psi_e = Q^m_n(i\xi) P^m_n(\mu) e^{(im\phi)} \] (1.55)

where $Q^m_n$ and $P^m_n$ are the Legendre polynomials of the second and first kind respectively, $n$ and $m$ denote the mode indexes and $e^{(im\phi)}$ is the angular variation. On this surface of the spheroid, where $\xi = \xi_0$, equation 1.55 is reduced to

\[ \psi_e(n, m, \xi, \mu, \phi) = P^m_n(\mu) e^{(im\phi)}. \] (1.56)

The gradients are then given by

\[ \nabla \psi_\mu(n, m, \xi, \mu, \phi) = \frac{1}{\xi_0} dP^m_n(\mu) e^{(im\phi)} \] (1.57)

\[ \nabla \psi_\phi(n, m, \xi, \mu, \phi) = \frac{im}{\xi_0 \mu} P^m_n(\mu) e^{(im\phi)}. \] (1.58)
For magnetostatic modes other than the primary mode, larger amplitudes close to the surface of the sphere are generally observed, this leads to a larger surface anisotropy contribution to the eigenvalues of the Hamiltonian corresponding to those modes. In turn, there is a larger contribution from surface anisotropy to the internal field and as a result, to the resonance condition (equation 1.43) such that higher order modes are observed at higher frequencies than the uniform mode. By surface anisotropy here we understand an energy term that is originating in the fact that surface ions have some unsatisfied exchange integrals when compared to the bulk symmetric crystallographic positions.

1.3.2.4 FMR-based Devices; Overview and Challenges

One of the most commonly used anisotropic ferro/ferrimagnetic materials for microwave applications are garnets (A$_3$Fe$_5$O$_{12}$). Yttrium iron garnet (Y$_3$Fe$_5$O$_{12}$) is the prototypical garnet used in FMR-based devices as it has an exceptionally sharp linewidth and high thermal stability ($T_c \approx 560$ K). As previously discussed, in the presence of an external bias field, the magnetic moments will tend to align with the field and undergo a damped precession. A microwave signal, when polarised in the same direction as the FMR precession in the material, will couple strongly with the moment. The interaction is much weaker when the FMR signal is polarised in the opposite direction. This ferro/ferrimagnetic resonance effect governs the operation of the majority of directional devices, such as circulators, isolators and filters, all fundamental features of high bandwidth mobile communications.

One of the most widely employed devices is the circulator, which is effectively a three-port non-reciprocal device. Here a signal entering port 1 will be received at port 2, an input signal at port 2 will transmit to port 3 and a signal at port 3 will transmit to port 1, but will not transmit in the reverse direction. The circulator may be used as an isolator by terminating one of the ports with a matched load impedance.

At low input power, the directional behaviour of the ferrite can be described by the linear equation of motion (equation 1.36) as described in section 1.3.2. At higher powers, non-linear effects become prominent. It is generally observed that at sufficiently
high powers, the amplitude of the resonant absorption peak will decrease with increasing power and gain a supplementary absorption peak [29]. Suhl considered spin-wave instability, when the amplitude of the spatially uniform resonance mode exceeds a critical threshold as the governing effect for non-linearity [30]. In this theory, spin-waves arising due to non-uniform magnetisation are coupled in second and higher orders to the primary mode by the demagnetising field, such that they extract power from the primary uniform mode. At low power this is only a small loss in absorption but at higher power levels the power losses to the spin-waves are significant and result in the amplitude of the spin-waves growing in time until they approach a very turbulent, unstable regime. In addition to this, dissipative non-linearity can introduce mode-mixing, beyond the realm of that which is accounted for by the fluctuation-dissipation theorem [31].

For the case of directional devices permanent magnets are typically used for the applied bias field, so spin-wave instability is not the dominating effect. Instead a second form of non-linearity exists for the condition of precession of the uniform magnetisation. At higher powers, higher order terms of the RF magnetisation and RF external field (\(m\) and \(h\) see equation 1.36) must be considered [32, 33] and may lead to second order or higher harmonics.

With the drive toward higher power devices (in excess of 100 W/cm\(^2\)) and increased density of high-performance devices in communication masts, addressing non-linear, high-power effects such as intermodulation distortion (IMD) or harmonic generation, becomes a critical limit to the density of high bandwidth communications for the foreseeable future (5G and beyond).

1.3.2.5 FMR Detection Methods; Conventional and State of the Art

FMR techniques are a key tool for evaluating materials for spin electronic applications and microwave devices, which allow for the probing of not only the gyroscopic properties of the materials but also the magnetic ground state and relaxation rates. To understand the drive to develop novel, non-inductive detection methods, conventional cavity or co-planar waveguide-based inductive techniques must first be discussed.
When discussing inductive detection techniques we refer to the monitoring of microwave losses in a transmission media (cavity, slotline, microstrip, etc.) inductively coupled to the sample being examined. Non-inductive methods rely on examining other phenomena for example the Kerr effect, thermal wave generation, or magnetic deflection as in magnetic force microscopy. Taking a look at cavity-based FMR, the earliest form of FMR detection, the fundamental components are a microwave generator (Gunn diode or klystron), an electromagnet, a ferrite circulator, a fixed-frequency cavity, a diode detector and a lock-in amplifier as shown in figure 1.10. Typically the resonant cavity is kept at a fixed frequency (usually X-band 9.6 GHz) and the electromagnet is swept within a finite range.

![Figure 1.10: Standard cavity-based FMR experimental set-up.](image)

Resonance detection relies on impedance matching as previously discussed in section 1.3.1, whereby the fixed frequency cavity behaves as a load impedance \( Z_L \), the impedance of which is matched to the source by adjusting the cavity coupling via an ‘iris,’ until system reflections are minimized. This is done when the sample is inserted into the cavity but prior to external field excitation. When a resonant excitation occurs, the load impedance will change, thus the matching criteria for minimal reflections no longer holds. Therefore the system reflections will increase and are detected by a diode. The signal is then lock-in amplified and digitized. Lock-in amplification requires that the signal is modulated at a known frequency (typically 10s of Hz). The modulation
is provided a set of Helmholtz coils powered by an AC source between the poles of the electromagnet which imposes a small field ($\approx 1$ Oe) modulation on the larger static DC field. Only at resonance will the output signal be at the same frequency as the frequency of the field modulation. The output signal is fed to the lock-in amplifier and the output is then a DC signal proportional to $\frac{dP}{dH}$: where $P$ is the absorbed microwave power and $H$ is the applied DC field. Measuring $\frac{dP}{dH}$ improves the achievable signal to noise ratio.

One advance in inductive FMR detection would be vector network analysis (VNA)-FMR, which greatly expands the bandwidth of resonance detection at the expense of sensitivity. The VNA injects RF into the sample either by microstrip or co-planar waveguide and simultaneously measures the amplitude and phase of reflected and transmitted signals through the waveguide. If the system is used in a single-port configuration, and if reflections are solely monitored, enhanced reflections are observed for a resonant interaction provided the cavity is designed to be impedance matched to the source. If the VNA is a two-port system, both reflections and transmitted signals can be measured to estimate resonant absorption. While both methods of FMR detection are easily employed, neither cavity nor microstrip/CPW FMR give absolute FMR detection, as the resonant absorption is inferred from monitoring microwave losses.

In the quest to improve absolute sensitivity, measurement bandwidth, particularly beyond 10 GHz and expand the overall scope of static and dynamic magnetisation characterisation, novel non-inductive methods have emerged. A number of state-of-the-art non-inductive detection techniques are discussed here.

In terms of improved sensitivity for smaller dimension samples, ferromagnetic resonance force microscopy (FMRFM) has led the way by making FMR detection on sub-micrometer (as small as atomic scale) length scales achievable [34, 35]. It does so by sensing the force between a probe micromagnet mounted on a tip attached to a cantilever and the magnetic moment of the test material in the presence of an RF excitation. The interaction force is detected by monitoring the deflection of the cantilever. This has proven capability in examining magnetisation dynamics in thin films
[36] and micron dots and is potentially sufficiently sensitive to selectively probe layered structures [37].

In 1984 Netzelmann et. al. introduced a method of FMR imaging by monitoring photothermal deflection [38]. Optical or microwave resonant absorption in a ferro/ferri-magnet results in oscillatory heating. In this detection method, the sample is immersed in a gas or liquid and a laser is used at grazing incidence. A change in the temperature of the gas or liquid, resulting from the resonant heating of the sample will change the gas/liquid’s reflective index which invokes a change in the deflection angle of the laser.

More recently it has been shown that x-ray magnetic circular dichroism (XMCD) is a useful tool for probing magnetic resonance effects [39]. This is done by measuring the longitudinal change in the magnetisation $m_z$ resulting from moment precession on the Fe sites. Magneto-optical magnetic resonance (MOMR) takes a similar approach, where the magneto-optical Kerr effect (MOKE) monitors this longitudinal drop in the projection of the magnetisation via a Kerr rotation [40].

These non-inductive methods laid the foundations for the novel SQUID FMR-detection discussed in later chapters, whereby athermal absolute ($\Delta m_z$) and thermal-detection, similar to the work by Netzelmann et. al. ([38]) but using the longitudinal magnetisation as a probe, can both be easily addressed by SQUID magnetometry. These can also be combined in real-time with VNA-FMR, AC susceptibility measurements and DC magnetometry, opening up a broad-range of analysis possible with a single module.
Chapter 2

Experimental Methods

This chapter describes the instrumentation that played an integral part in this work. This is subdivided into five separate forms of characterisation; microwave reflectometry, electromagnetic simulations, magnetometry, optical methods and compositional analysis techniques.

2.1 Microwave Reflectometry

As explained in Section 1.3.1, microwave reflectometry deals with the monitoring of reflected and transmitted electromagnetic waves as they travel through a device under test (DUT) and the figure of merit for the device is given by the reflection coefficient in equation 1.2. This method of monitoring incident and reflected microwave energy has been used extensively in the characterisation of the conductivity of thin films [41, 42] and its applicability in the analysis of randomly dispersed nano-structured media, such as metallic nanowire arrays or printed nano-platelets is now investigated. This ‘echo’ technique is performed in two forms; pulsed time-domain reflectometry (TDR) and continuous wave (CW) vector network analysis (VNA). The inductive broadband microwave reflectometry techniques described in this section form the basis of the work on analysing the conductivity of randomly distributed Ag nanowire networks presented in Chapter 3. Additionally frequency domain VNA is utilised to evaluate inkjet printed Ag films and grids for electromagnetic shielding application in Chapter 4.

2.1.1 Pulsed Time Domain Reflectometry

Performed up to 50 GHz, TDR involves launching a fast edge voltage pulse into the DUT [43]. Microwave reflections are then detected by a broadband receiver and monitored with an oscilloscope. If the load impedance is matched to the standard 50 Ω line the incident energy will be fully transmitted and the oscilloscope will display just the incident stepped pulse. If a discontinuity is encountered, part of the wave is reflected back toward this source. This appears on the oscilloscope added algebraically.
to the source pulse but separated in time represented by either a reflected voltage (mV) or reflection coefficient (Ω). Hence signals arising from discontinuities can be easily identified and characteristic impedances can be extracted. The shape of the trace reveals not only the magnitude of the reflection but also its nature. Figure 2.1 shows the shape profile for five different situations which may easily be identified using TDR: (a) an open circuit, where the load impedance is considered to be $Z_L \rightarrow \infty$, (b) a short circuit, where $Z_L = 0$, (c) a line terminated by a load impedance equal to twice the characteristic impedance of the input signal $Z_0$ i.e. $Z_L = 2Z_0$, (d) inductive termination and (e) capacitive termination.

![TDR Spectra Diagram]

**Figure 2.1:** The nature of the discontinuity can be identified from the shape of the TDR spectra. Here five profiles are displayed; (a) open circuit where the load impedance is considered to be $Z_L \rightarrow \infty$ (b) short circuit $Z_L = 0$ and (c) a circuit terminated with $Z_L = 2Z_0$ (d) inductive termination. Here the inductor will not accept a sudden change in the current and initially behaves as an open circuit. At some stage the current in the inductor builds exponentially and the impedance drops to zero. It then behaves as a short circuit. (e) capacitive termination. Similarly the capacitor will not accept a sudden change in voltage, behaving initially as a short circuit. After some change time the voltage builds on the capacitor and the impedance rises. It then behaves as an open circuit.

The system can simultaneously be used in a time domain transmission (TDT) configuration, where the microwave excitation exiting the DUT is the input for another
channel on the oscilloscope. Signals from both reflection and transmission will then appear separated in time, corresponding to the length from the excitation port to the input port (i.e. the length of the cables, microstrip and DUT). Examining both the reflection and transmission spectra resulting from interaction with a device, the reflection and transmission coefficients can easily be extracted as well as gain/loss and propagation length for the device. The advantages and disadvantages of pulsed TDR characterisation are summarized in table 2.1.

<table>
<thead>
<tr>
<th>Advantages</th>
<th>Limitations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Large frequency range from DC to maximum of TDR source (for properly de-phased dual source &gt; 50 GHz)</td>
<td>Post processing (FFT) is required to convert to the frequency domain</td>
</tr>
<tr>
<td>Broadband receiver allows for rapid measurements</td>
<td>Lower signal to noise ratio in comparison to narrowband measurements</td>
</tr>
<tr>
<td>Low cost step pulse generators compared to continuous wave sources</td>
<td>Limited excitation power</td>
</tr>
</tbody>
</table>

Figure 2.2a shows the rough schematic of a typical TDR set-up. Here the sample under test is subjected to a pulsed signal via a microstrip transmission line. It is then either shorted with a load impedance $Z_L$ or returned to a secondary port on the spectrum analyser.
2.1.2 Continuous Wave Vector Network Analysis

VNA on the other hand involves the signal generator transmitting a sinusoidal wave of swept frequency \((f < 20 \text{ GHz})\) through the DUT. Transmitted and reflected signals are monitored by an analyser and displayed as the addition of signals from all discontinuities within the DUT. Figure 2.2b shows the experimental set-up for the VNA. Reflected and transmitted signals are represented mathematically by the scattering parameters \(S_{xy}\), where \(x\) represents the measuring port and \(y\) is the number of the source port. For the case of a two-port system, the transmission characteristics are described by the \(S_{11}\) forward reflection, the \(S_{22}\) reverse reflection, \(S_{21}\) the forward transmission and the \(S_{12}\) reverse transmission as seen in figure 2.3.
The $S$-parameters can be described in terms of the normalized incident and reflected voltages $a \left( = \frac{V^+ (x)}{\sqrt{Z_0}} \right)$ and $b \left( = \frac{V^- (x)}{\sqrt{Z_0}} \right)$, such that

\begin{align*}
S_{11} & = \frac{b_1}{a_1} \tag{2.1a} \\
S_{12} & = \frac{b_1}{a_2} \tag{2.1b} \\
S_{21} & = \frac{b_2}{a_1} \tag{2.1c} \\
S_{22} & = \frac{b_2}{a_2} \tag{2.1d}
\end{align*}

It then follows that the scattering parameters can be represented by the matrix

\[
\begin{pmatrix}
    b_1 \\
    b_2 \\
\end{pmatrix}
= 
\begin{pmatrix}
    S_{11} & S_{12} \\
    S_{21} & S_{22} \\
\end{pmatrix}
\times 
\begin{pmatrix}
    a_1 \\
    a_2 \\
\end{pmatrix}
\tag{2.2}
\]

for a two-port system. By this reasoning, a three port system is described by

\[
\begin{pmatrix}
    b_1 \\
    b_2 \\
    b_3 \\
\end{pmatrix}
= 
\begin{pmatrix}
    S_{11} & S_{12} & S_{13} \\
    S_{21} & S_{22} & S_{23} \\
    S_{31} & S_{32} & S_{33} \\
\end{pmatrix}
\times 
\begin{pmatrix}
    a_1 \\
    a_2 \\
    a_3 \\
\end{pmatrix}
\tag{2.3}
\]

and measurements with any number of ports can be described in a similar fashion. The $S$-parameters can be given as either a linear ratio as in equations 2.1 or a logarithmic
amplitude in dB, as in equation 2.4, which is given in terms of the power ratio.

\[
S_{xy}[^{\mathrm{dB}}] = 20 \log \left( \frac{P_y}{P_x} \right)
\]  

(2.4)

With a VNA, measurement of the phase of reflected or transmitted signals in addition to the magnitude is possible, which allows for a much more detailed analysis of the device. At each VNA port, either a directional coupler or bridge separates incident and reflected signals and directs them to a dedicated heterodyne receiver where both the magnitude and phase of the signal are extracted. The receiver essentially takes the input RF and downconverts it to an intermediate frequency (IF) by mixing the input signal with a signal originating from a local oscillator. The downconverted signal is converted from an analog to a digital signal and processed. Downconversion of the RF signal serves a number of purposes, namely that it allows for simpler processing at lower frequencies and also better noise-rejection. It also improves frequency selectivity, making it easier to distinguish features that lie very close in frequency range. This downconversion process is illustrated in figure 2.4. Each port will have at least two receivers, exclusively for detecting the incident input signal and the reflected signal.

![Diagram](image)

**Figure 2.4:** The VNA-receiver measurement strategy employs downconversion of the RF input, analog to digital conversion and digital signal processing.

As results are displayed in the frequency domain, an inverse fast Fourier transform (iFFT) is required to convert to the time domain for determining the location of discontinuities [44]. This requires measurement of both the magnitude and phase of measured signals.
### Table 2.2: Advantages and Limitations of Continuous Wave Vector Network Analysis

<table>
<thead>
<tr>
<th>Advantages</th>
<th>Limitations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Measurements are taken directly in the frequency domain (no post-processing required to examine a frequency response)</td>
<td>Interpreting spectra is more complex than time domain analysis</td>
</tr>
<tr>
<td>Narrowbanding the receiver is possible to improve the signal to noise ratio</td>
<td>Measurements can only have a finite frequency range, which for high end analysers is typically &lt; 20 GHz</td>
</tr>
<tr>
<td>Possess a large dynamic range (&gt; 100 dB)</td>
<td>Higher cost than time-domain systems</td>
</tr>
<tr>
<td>The signal to noise ratio is overall far greater for VNA than TDR</td>
<td>The location of discontinuities is only know after converting to the time domain via a iFFT</td>
</tr>
</tbody>
</table>

Despite the greater difficulty in interpretation of results, it has the advantage of greater sensitivity than TDR analysis as the noise level is proportion to the square root of the bandwidth. TDR analysis can typically probe much lower frequencies than VNA, therefore reliability engineers typically use both techniques for device integrity evaluation. The main advantages and limitations of vector network analysis are given in table 2.2. The sensitivity limits of both TDR and VNA microwave reflectometry techniques are heavily dependent on the conductivity of the test material. For insulating dielectrics 10-100s µm of material would be required to produce a reflected signal, whereas reflections may be seen from a few monolayers of highly conductive materials, such as Au.

#### 2.1.2.1 Transfer Matrices

There are a number of ways of expressing two-port parameters; $Z$ (impedance), $Y$ (admittance), $h$ (hybrid), $ABCD$ (chain), $S$ (scattering, previously discussed) and $T$ (transfer) and it may be convenient to convert between parameters. For this work $T \rightarrow S$ parametric conversion is formulated. The transmission matrix $T$ may be defined
Experimental Methods 2.2

as

\[ T = \begin{pmatrix} T_{11} & T_{12} \\ T_{21} & T_{22} \end{pmatrix} \quad (2.5) \]

and the conversion between \( S \) and \( T \) parameters are described by the equations

\[ T = \frac{1}{S_{21}} \begin{pmatrix} S_{12}S_{21} - S_{11}S_{22} & S_{11} \\ -S_{22} & 1 \end{pmatrix} \quad (2.6a) \]

\[ S = \frac{1}{T_{22}} \begin{pmatrix} T_{12} & T_{11}T_{22} - T_{12}T_{21} \\ 1 & -T_{21} \end{pmatrix} \quad (2.6b) \]

for a two-port system. Conversion between parameters will be a useful tool for electromagnetic simulations of high complexity novel instrumentation described in later chapters.

### 2.2 Electromagnetic Simulations for Cavity and Waveguide Design

CST is a powerful simulation tool for solving high-complexity electromagnetic problems over a broad range of frequencies, from static to the optical regime. CST microwave studio, one of many packages in the CST environment is used in this study to monitor electric and magnetic fields, evaluate scattering parameters and quantify power losses in proposed designs for transmission lines, microwave cavities and adapters prior to fabrication. This section will have relevance in Chapter 4, where CST is used to simulate electromagnetic wave propagation in rectangular waveguides prior to their fabrication and use in analysing the electromagnetic interaction (EMI) shielding effectiveness of inkjet printed films and grids.

#### 2.2.1 Finite Integration Technique

CST Microwave Studio (MWS) utilizes the finite integration technique (FIT) to solve Maxwell’s equations for a defined structure [45]. This numerical approach discretises the integral form of Maxwell equations to allow for solutions to systems of complex
geometry. CST first restricts the electromagnetic problem to a volume of finite boundaries. It then splits the entire calculation domain (structure) into a number of discrete volume elements or mesh cells which may be hexahedral or tetrahedral depending on the solver used. Fig. 2.5 shows a graphical representation of the unit mesh cell defined by CST, where $e_n$ is the electric voltage at the cell edge and $b_n$ is the magnetic flux through the face of the cell.

![Graphical Representation of different ‘mesh cells’ defined by CST MWS using the finite integration technique. The unknowns are represented as the electric voltages at the edges of the cell, $e_n$ and the magnetic fluxes at the faces of each cell, $b_n$.](image)

Take for example, Faraday’s Law in its integral form, given by:

$$
\int \mathbf{E} \cdot d\mathbf{S} = \frac{d}{dt} \int \mathbf{B} \cdot d\mathbf{A} \quad (2.7)
$$

$\int \mathbf{E} \cdot d\mathbf{S}$ for a single cell may be described as the sum of the $e_n$ electric voltages at the cell edges such that

$$
\int \mathbf{E} \cdot d\mathbf{S} = e_k + e_j - e_i - e_l. \quad (2.8)
$$

Similarly $\int \mathbf{B} \cdot d\mathbf{A}$ can be described by the magnetic flux through the cell face $b_n$. It then follows that

$$
e_k + e_j - e_i - e_l = \frac{d}{dt} b_n \quad (2.9)
$$

Summing over all meshes within the structure gives the discretised form of Faraday’s Law

$$
\begin{pmatrix}
e_k \\
e_j \\
e_i \\
e_l
\end{pmatrix}
= \frac{d}{dt}
\begin{pmatrix}
b_n \\
\vdots
\end{pmatrix} \quad (2.10)
$$

The other Maxwell equations may be discretised in a similar fashion.
2.2.2 CST Solvers

CST MWS offers a number of different solvers depending on the application. The most commonly used being the transient time-domain and the frequency domain solvers [46]. When deciding on an appropriate solver the size of the device in relation to the wavelength of operation, the device’s complexity and it’s resonant nature must be considered. The transient solver is inherently suited to time-domain reflectometry analysis and also the extraction of broadband scattering (S) parameters and is the primary solver utilized in chapters 3 and 4. The frequency domain solver, while more computationally expensive is similar to the transient solver in that it provides high accuracy S-parameter extraction. It is typically better suited to the analysis of electrically small devices or those with a high Q-factor, such as ferrite-based circulators or isolators.

![Figure 2.6: CST solvers (a) Transient time domain mesh (b) tetrahedral frequency domain mesh](image)

For the case of the transient time domain solver the calculation geometry is divided into hexahedral cells as in figure 2.6a and a time domain signal is propagated in the defined device. This solver has issues with curved surfaces and meshes must be very sufficiently fine to compute them accurately, which can greatly increase the computational time. The accuracy of the calculation is strongly dependent on the number of mesh cells, which for MW Studio Student Edition is limited to 30,000 cells per calculation. Computing electromagnetic propagation through resonant devices also has long simulation times using the time domain solver and is much better suited to frequency domain solutions. The frequency domain solver uses the finite element method, which first defines the structure in terms of a surface triangular mesh. If the device has volume, this is extended to a tetrahedral mesh as shown in figure 2.6b. The main
limitations with this solver are the available random access memory (RAM) for the calculation and the hard 10,000 mesh cell limit.

2.2.3 CST Model Construction

The first step in simulation is creating a 3D construction of desired geometry for the application. MW Studio features a number of standard shapes; cube, cylinder, sphere, cone and torus and boolean addition and subtraction of shapes can be used to create custom shapes such as cavities or protrusions. CST has an in-built material library and each component can be defined as consisting of a different material with distinct values of relative permeability $\mu_r$ and permittivity $\varepsilon_r$.

Excitation is provided by defining either a discrete port or waveguide port. For $S$-parameter calculation the discrete port is modelled as a lumped circuit element source consisting of a current source with an internal impedance and is defined by picking two points as in figure 2.7a. This excitation is mainly suitable for low frequency simulations of planar devices such as slotline antennae. For high frequency waveguide simulations, the waveguide port is most suitable. Here the excitation field is assumed to be composed of modes from an quasi-infinitely long, lossless waveguide of equivalent geometry to the defined waveguide plane shown in figure 2.7b.

The swept frequency range may be defined and altered easily and field monitors are specified at frequencies of interest. $S$-parameters are determined for the full operational bandwidth but electric and magnetic fields are simulated at the defined frequencies. Manual mesh refinement is performed to maximize the accuracy while ensuring the
maximum number of cells is not exceeded. This is done by carefully adjusting the number of cells per wavelength and cells at box edges within the structure. For high complexity structures the model may be simplified by assuming lossy conductors as perfect electrical conductors and lossy dielectrics as lossless.

2.3 Magnetometry

2.3.1 SQUID Magnetometry

A Superconducting QUantum Interference Device (SQUID) is a high-sensitivity magnetometer capable of sensing $10^{-10} \text{Am}^2$ or better in DC mode or $10^{-15} \text{Am}^2$ in AC mode, relying on both the principles that the flux threading a superconducting loop is quantised and the Josephson effect. This section will give an overview of both phenomena, introduce the flux-locked loop readout circuitry and give a brief introduction to the newly developed SQUID-FMR technique. This technique is discussed in greater detail in Chapter 6.

2.3.1.1 Flux Quantisation in a Superconducting Loop

According to the BCS theory of superconductivity, electrons of opposite spin and momentum are weakly bound in correlated pairs [47]. The coherence length of such pairs is sufficiently long that they overlap and the quantum-mechanical wavefunction for each pair is considered to be coherent and in phase with that of the other pairs. This leads us to the situation where the superconducting state around the superconducting loop may be described by a single valued macroscopic wavefunction of the form

$$\Psi = |\Psi(r)| e^{i\kappa \cdot r}$$

(2.11)

where $\kappa \cdot r$ is position dependent phase and given as $\kappa \cdot r = \theta(r)$. $|\Psi(r)|^2$ is the local Cooper pair density and is given by

$$\rho_{sc} = |\Psi(r)|^2.$$  

(2.12)
\( \rho_{sc} \) may be considered spatially invariant, as is the case for low fields. We now consider a charged particle moving in a field of vector potential \( \mathbf{A} \) with an effective mass and charge of \( m^* \) and \( q^* \) respectively. Taking the Ginzburg-Landau phenomenological formulation for superconductivity, the Gibbs free energy must be invariant and given by

\[
\nabla \times (\nabla \times \mathbf{A}) + \frac{i q^*}{2m^*} (\Psi^* \nabla \Psi - \Psi \nabla \Psi^*) + \frac{q^*^2}{m^*} A |\Psi|^2 = 0. \tag{2.13}
\]

Using the definition for the potential vector \( \mathbf{B} = \nabla \times \mathbf{A} \), the Maxwell equation \( \nabla \times \mathbf{B} = \mu_0 \mathbf{I} \) and the London-Landau gauge \( \nabla \mathbf{A} = 0 \) an expression for the current density \( \mathbf{I} \) can be obtained as

\[
\mu_0 \mathbf{I} = -\frac{i h q^*}{2m^*} (\Psi^* \nabla \Psi - \Psi \nabla \Psi^*) + \frac{q^*^2}{m^*} A |\Psi|^2. \tag{2.14}
\]

The gradient of the quantum-mechanical wavefunction is given as

\[
\nabla \Psi = i \Psi \nabla \theta + e^{i\theta} \nabla |\Psi|. \tag{2.15}
\]

Subbing in equations 2.11 and 2.15 into equation 2.14 the expression for the current density takes the form

\[
\mu_0 \mathbf{I} = \frac{h q^*}{m^*} |\Psi|^2 \nabla \theta - \frac{q^*^2}{m^*} A |\Psi|^2. \tag{2.16}
\]

As previously stated, the macroscopic wavefunction given in equation 2.11 must be single-valued. In order for this to be the case the phase must be coherent and a multiple of \( 2\pi \) along the contour of the superconducting loop i.e.

\[
\oint \nabla \theta dl = 2\pi n \tag{2.17}
\]

where \( n \) is an integer number. Integrating expression 2.16 along the contour and substituting equation 2.17 we obtain the following expression

\[
\frac{\mu_0 m^*}{q^*^2} \oint \frac{I}{|\Psi|^2} \cdot dl = \frac{h}{q^*} \oint \nabla \theta \cdot dl - \oint \mathbf{A} \cdot dl = \frac{h 2\pi n}{q^*} - \oint \mathbf{A} \cdot dl \tag{2.18}
\]
Applying Stokes theorem, which states \( \oint A \, dl = \oint B \, dS = \Phi \), where \( \Phi \) is the magnetic flux, equation 2.18 becomes

\[
\frac{\mu_0 m^*}{q^*} \oint \frac{I}{|\Psi|^2} \cdot dl = \frac{\hbar 2 \pi n}{q^*} - \Phi = n\Phi_0 - \Phi
\]  

(2.19)

Here \( \Phi_0 \) is a flux quantum, where \( \Phi_0 = \frac{\hbar}{q^*} \). As \( q^* \) is the effective charge of the Cooper pair, then \( \Phi_0 = \frac{\hbar}{2q} = 2.067833758 \times 10^{-15} \text{ Wb} \). In the region of the superconducting loop where the density of the Cooper pairs is constant (i.e. away from the surface of the loop) there is zero current density and equation 2.19 may be further reduced to

\[
\Phi = n\Phi_0
\]  

(2.20)

Thus the flux threading a superconducting loop is quantized i.e. confined to an integral number of \( \Phi_0 \).

### 2.3.1.2 The Josephson Effect

The SQUID geometry consists of a superconducting loop with one (RF SQUID) or two (DC SQUID) weak junctions separated by thin insulating layers. These superconductor-insulator-superconductor (SIS) structures are known as Josephson junctions, which exhibit non-zero current in an unbiased junction due to the quantum-mechanical tunnelling of Cooper pairs through the insulating barrier. The effect was first theorised by Josephson [49, 50] in 1962 and then confirmed experimentally in 1963 by Philip Anderson and John Rowell of Bell Labs in Princeton [51]. Josephson calculated that the current \( I \) flowing through the SIS junction is given by

\[
I = I_c \sin \delta
\]  

(2.21)

where \( I_c \) is the critical current density and \( \delta \) is the phase difference between the condensates in the two superconductors (i.e. \( \delta = \phi_1 - \phi_2 \)). The equivalent circuit configurations for DC and RF SQUIDs are represented in figures 2.8a and 2.8b respectively. The DC SQUID essentially consists of a superconducting loop with two Josephson junctions, while an RF SQUID contains a singular weak junction.
A SQUID operates on the principle that the flux threading a superconducting loop is quantized i.e. confined to an integral number of $\Phi_0 = \frac{h}{2e} = 2.067833758 \times 10^{-15} \text{ Wb}$ as shown in section 2.3.1.1. Therefore when an external flux is applied, the loop will increase or screen the flux in order to reach $n\Phi_0$, creating a circulating current.

**2.3.1.3 Principle of Operation: DC SQUID**

The DC SQUID consists a superconducting loop of inductance $L$ interrupted by two Josephson junctions. The superconducting loop is biased with a DC current $I > 2I_c$, developing a DC voltage across the junctions. When the magnetic flux threading the superconducting loop changes, this induces a phase change $\delta$ that increases the current in one junction, while decreasing the current in the other junction. As this external flux increase/decreases so too does the DC voltage change across the loop. Figure 2.9 shows the I-V characteristics for a DC SQUID. If the SQUID is biased with a constant
current $I_B$, the voltage across the SQUID will oscillate with a period $\Phi_0$ as the external flux $\Phi$ steadily increases/decreases. As seen in figure 2.10, $(n + \frac{1}{2})\Phi_0$ and $\Phi_0$ correspond to the maxima and minima of oscillation respectively. For the case of the DC SQUID the $V - \Phi$ response is sinusoidal in nature. The bias current is chosen in order to maximise the the gain $\frac{dV}{d\Phi}$ and the voltage change across the junctions resulting from an external flux change is measured using a low-noise amplifier.

Due the variable nature of the gain it may be difficult to determine if the external flux is increasing or decreasing. It is therefore important to linearise the SQUID response. In order to linearise the $I$-$\Phi$ response a flux-locked loop is used, which maintains $\Phi = \frac{n}{2}\Phi_0$ i.e. the maxima. A modulating flux $\Phi_m < \frac{\Phi_0}{2}$ of frequency $\omega$ is applied to the SQUID by an oscillator. This modulation is supplied via inductive coupling to a modulation coil as shown in figure 2.8a. The SQUID signal is sent to a lock-in amplifier where it is referenced to the oscillator signal. The SQUID signal $V_S$ is periodic on $\frac{n}{2}\Phi_0$ and the lock-in amplifier signal is a periodic function of $2\omega$. If the SQUID signal is not a multiple of $\frac{n}{2}\Phi_0$, it contains a component at a frequency of $\omega$ and the lock-in amplifier sends out a voltage proportional to the signal at $\omega$. This lumped-circuitry is referred to as a flux-locked-SQUID system. It essentially ensures that the sum of the external flux and the feedback flux is constant and maintained at the optimal working point. If the modulation flux is applied at the points a, b and c as depicted in figure 2.10, the flux-locked loop feedback $V_L$ will vary as seen in figure 2.11 in order to maintain the SQUID flux at $\frac{n}{2}\Phi_0$. Figures below are adapted from Clarke et al. [52].
2.3.1.4 Principle of Operation: RF SQUID

The RF SQUID consists of a single Josephson junction with a critical current $I_c$ interrupting a superconducting loop of inductance $L$. Flux quantisation in the superconducting loop imposes the constraint

$$\delta = \frac{2\pi \Phi_{\text{total}}}{\Phi_0} = 2\pi n.$$  \hspace{1cm} (2.22)

on the total flux $\Phi_{\text{total}}$ in the superconducting loop. Using equation 2.21 and substituting the flux quantisation criteria the supercurrent across the junction can be given.
by
\[ I = I_c \sin \delta = I_c \sin \left( \frac{2\pi \Phi_{\text{total}}}{\Phi_0} \right). \] (2.23)

It then follows that the total flux in the superconducting loop is given by
\[ \Phi_{\text{total}} = \Phi_{\text{ext}} - LI = \Phi_{\text{ext}} - LI_c \sin \left( \frac{2\pi \Phi_{\text{total}}}{\Phi_0} \right) \] (2.24)

where \( \Phi_{\text{ext}} \) is the external applied flux. Equation 2.24 results in two distinct regimes of operation. For \( \beta_{\text{RF}} = 2LI_c/\Phi_0 < 1 \)
\[ \frac{\Phi_{\text{total}}}{\Phi_0} = \frac{\Phi_{\text{ext}}}{\Phi_0} - \beta_{\text{RF}} \sin \left( \frac{2\pi \Phi_{\text{total}}}{\Phi_0} \right) \] (2.25)

and as shown in figure 2.12 the curve is non-hysteretic and the slope \( d\Phi_{\text{total}}/d\Phi_{\text{ext}} = 1/[1 + \beta_{\text{RF}} \cos(2\pi)\Phi_{\text{total}}/\Phi_0] \) is positive everywhere. On the other hand if \( \beta_{\text{RF}} = 2LI_c/\Phi_0 > 1 \) the slope \( d\Phi_{\text{total}}/d\Phi_{\text{ext}} \) may be positive, negative or divergent. As only regions where the slope is positive are stable the device will make jumps between flux states on increasing (decreasing) the externally applied flux. This behaviour is described as hysteretic in nature.

![Figure 2.12: The total flux as a function of the external flux for three conditions. (a) The blue dashed line \( \Phi_{\text{total}} = \Phi_{\text{ext}} \) (b) The black curve \( \beta_{\text{RF}} = 0.5 \) and (c) the red curve \( \beta_{\text{RF}} = 2 \).](image)

The superconducting loop is coupled to a resonant tank LC circuit with an inductance \( L_{\text{tank}} \) via a mutual inductance \( M = k(LL_{\text{tank}}^{1/2}) \). The tank circuit is driven by an
RF current $I_{RF} \sin \omega_{RF} t$, which induces flux in the SQUID loop. The voltage across the tank circuit coil serves as a readout of the external applied flux, as it is dependent on both the RF current and is periodic as a function of the applied flux $\Phi_{ext}$ with a period of $\Phi_0$ as seen in figure 2.10. This triangular response is a linear detector of magnetic flux changes up to $1\Phi_0$. In order to increase the detection range a negative feedback flux-locked loop is used which locks onto either a peak or a trough of the triangular voltage response. Here an oscillator provides a flux modulation of amplitude $\pm \Phi_0/2$ via the coil of the LC tank circuit, while also providing a reference to a lock-in amplifier. When the total flux $\Phi_{total}$ is at a maximum or minimum of the triangular response then the amplitude of the modulation is zero, but it increases linearly as the flux departs from the extrema. A change in flux in the superconducting loop due to external flux results in an output signal from the lock-in amplifier that it fed back through a resistor to the coil of the tank circuit. The RF current in the coil induces a flux in the SQUID that counters the flux change due to external flux and maintains the SQUID locked at either a maxima or minima of the triangular response.

2.3.1.5 Gradiometer Coil Assemblies

One of the primary issues with using the SQUID directly as a magnetometer is the limiting small area of the device and it’s associated inductance. In order to increase the measurable area, most SQUID-magnetometer systems use a superconducting flux transformer to couple externally detected flux to the SQUID washer. This additionally allows for the SQUID-device to be maintained within a magnetically isolated environment. In this way the SQUID does not detect the magnetic properties of a sample directly. Instead, the sample passes through detection coils located at the centre of the superconducting magnet and the sample’s magnetic moment induces current in the sensing coils which in turn varies the total flux in the SQUID. Considering the RF SQUID, $\Phi_{ext}$ is measured by detection and amplification of the RF voltage in the resonant LC tank circuit. Hence measuring this voltage and knowing the correct calibration factor, one can accurately measure the magnetic moment of a sample.
The detection coil geometries for coupling to the RF or DC SQUID can come in many forms, namely the magnetometer (single-turn superconducting pick-up loop) or spatial gradiometer (multi-turn superconducting circuit) configurations. SQUID magnetometers or gradiometers are vector instruments, measuring only the magnetisation component perpendicular to the plane of the pick-up loop ($m_z$). Of the many optimised pick-up loop geometries the second-order gradiometer is considered one of the best in terms of rejection of noise and external environmental magnetic fields. The general arrangement of the SQUID second-order gradiometer sensing coils may be seen in figure 2.13, which consists of four single turns of superconducting wire typically wound on a material with a small thermal expansion coefficient (quartz, fibreglass, macor, etc.). The coils are configured in such a way that a uniform axial field $B_z$ induces zero net flux in the gradiometer, while a second-order gradient $\frac{dB_z^2}{dz^2}$ induces a proportional flux that is then coupled to the SQUID-device. In this way, the gradiometer acts as a filter for ambient magnetic noise. The moment of a sample is measured by translating the sample through the central position of the coil assembly and processing the flux coupled to the SQUID via a superconducting transformer as a function of the distance from the central position.
2.3.1.6 A Brief Introduction to the SQUID-FMR Technique

Figure 2.14: Change in the projection of the magnetic moment along z ($\Delta m_z$) during resonance.

Here a novel resonance experiment is proposed, whereby a sample placed in a static magnetic field produced by a superconducting magnet (along the $z$-axis, perpendicular to the plane of the gradiometer coils) is subjected to a broadband swept frequency via microstrip transmission lines (RF excitation in the $xy$-plane) and the change in the projection of the magnetic moment along the $z$-axis $\Delta m_z$ is monitored using a RF SQUID magnetometer. The total moment $m_z$ is measured in the conventional way by translating the sample through the gradiometer and processing the coupled flux as a function of distance by monitoring the RF voltage across the resonant tank circuit. In order to measure the comparatively small drop in the moment on resonance ($\Delta m_z$) the superconducting flux transformer to the SQUID washer must be momentarily quenched. At this point no supercurrent is flowing in the pick-up coil assembly and only relative changes in flux due to temperature, microwave absorption at resonance, etc. are measured. As the change in RF voltage to change in flux to change in moment is well calibrated the $\Delta m_z$ can be detected by directly monitoring the RF voltage across the tank circuit. As seen in figure 2.14, by measuring $\Delta m_z$ and $m_z$ the precession cone angle may be determined as follows

$$\cos \theta = \frac{m_z - \Delta m_z}{m_z} = 1 - \frac{\Delta m_z}{m_z}$$ \hspace{1cm} (2.26a)

$$\theta = \cos^{-1} \left( 1 - \frac{\Delta m_z}{m_z} \right).$$ \hspace{1cm} (2.26b)
Figure 2.14 shows the resonance mode for a ferrimagnet, which at microwave frequencies behaves like a single sub-lattice with a net moment $m$ which precesses about the applied field, where $m$ is the difference between the moments of the two antiparallel sub-lattices. At higher frequencies (IR) the sub-lattices no longer precess antiparallel and each sub-lattice will have a different resonance frequency proportional to the exchange interaction field $H_{ex}$ between the sub-lattices [53]. The resonance condition given by Kittel’s equation 1.43 is modified to

$$\omega_0 = \mu_0 \gamma \left( H + [H_{an}(H_{an} + 2H_{ex})]^{1/2} \right)$$

(2.27)

for antiferromagnetic/ferrimagnetic resonance at 0 K [54, 55]. This measurement technique will be expanded on in Chapter 6.

### 2.4 Optical Methods

This section will detail a number of methods which use the interaction of light with matter to probe the structure, density and frequency of random or quasi-random materials. The first of these methods, laser specular scattering (LSS) is used to characterise the length scales of metallic nanowires, which have been spray deposited onto dielectric substrates. The conductive properties of such networks are dealt with in Chapter 3 and their topography is examined using LSS in Chapter 5. This section also details Ultraviolet-visible (UV) spectroscopy in the context of examining the density of metallic inkjet printed films, which are examined as potential EMI shielding materials in 4. The results of the UV-visible densitometry for printed Ag films are presented in Chapter 5.

#### 2.4.1 Laser Specular Scattering (LSS)

Small angle optical scattering (SAS) or laser specular scattering has long been used for characterising the dispersion and average sizes of macromolecules such as polymers or proteins in colloidal suspensions [56, 57]. The technique is particularly useful in the structural analysis of disordered or partially disordered systems. Here a similar technique is employed for determining the radial distribution and average length
of nanowires in nanowire networks (NWNs) of various optical transmittance. This is performed under two modes; transmission mode and reflection mode as shown schematically in figure 2.15a and 2.15b respectively. Both configurations consist of a helium neon laser (HeNe) ($\lambda = 632.8$ nm) positioned $z=1$ m from a white screen containing a pinhole in the centre. The pinhole ensures the exclusion of the $I_0$ beam i.e. laser light directly from the source, not resulting from scattering from the NWN and helps with calibration when flattening and dewarping images. In transmission mode the sample is affixed to the front of the laser, allowing the beam to pass directly through the sample, producing a specular pattern on the screen. Hence this mode is suitable for transparent dielectric substrates. With reflection mode, the laser beam passes through a pinhole in the centre of the screen, impinges on a non-transparent (in this case semi-conducting) substrate and the light is scattered back to the screen. The specular image is then photographed from the screen using a digital camera at an angle. An example of a raw specular image is shown in figure 2.16.

![Figure 2.15: Experimental set-up for (a) transmission and (b) reflection mode small angle optical scattering](image)

![Figure 2.16: As captured specular scattering](image)
2.4.2 Ultraviolet–visible Densitometry

An Ocean Optics S1000, fibre-optic spectrometer is utilized to characterise the optical density of metallic films on transparent dielectric substrates. In this spectroscopic approach, light from a broadband UV-visible lamp (Xe-lamp) traverses a fibre-optic cable and feeds into a small aperture in a miniature adjustable optical bench. The beam illuminates the sample under investigation, which is clamped within the optical cavity. The transmitted light is then picked up by a secondary fibre-optic cable and detected by a spectrometer grating. The light is then dispersed onto a diode array, where each pixel in the array represents a different wavelength, such that the intensities of transmitted light are recorded. The S1000 detector is most sensitive for wavelengths 400 nm to 900 nm. Fig. 2.17 shows a schematic representation of the optical transmittance measurement. A representative transmitted intensity spectrum for the Xe-lamp is shown in figure 2.18.

![Schematic diagram of optical densitometry sensor configuration for measuring transmitted UV-visible optical intensities.](image-url)
In order to measure absorbance the optical reflections must be accounted for as:

\[ A + T + R = 1 \]  \hspace{1cm} (2.28)

where A, T and R are the optical absorption, transmission and reflection as light passes through the test media. Reflections are measured by reconfiguring the geometry in Fig. 2.17 to that shown in Fig. 2.19. In this configuration the optical waveguides are positioned at right angles to each other, interfacing an optical black box. The sample is placed in a custom made planar clamp at 45° to the incident light aperture. As only 45° reflections are measured, it is expected that the reflection coefficient will be underestimated because specular and diffuse reflections are not accounted for. The specular component of reflection may be large, especially for non-uniform or textured materials. Estimation of the specular reflection would require an integrating sphere with multiple detectors at multiple angles.
2.5 Material Characterisation

As the premise of this study is the development of novel microwave-based techniques and their application to a broad variety of materials, fabrication and structural characterisation played only a minor role. This was primarily in the evaluation of the crystallinity and stoichiometry of Yttrium Iron Garnet, used as a standard test material in chapter 6. Hence this section will provide a brief description of both x-ray diffraction (XRD) and energy dispersive x-ray spectroscopy (EDX).

2.5.1 X-Ray Diffraction (XRD)

XRD is a highly useful, non-destructive technique for evaluating the crystallinity of materials. The principle of XRD relies on the constructive interference of x-rays with an ordered crystalline structure. In a typical system, x-rays are produced by electron bombardment with a copper or molybdenum anode. The electrons are produced via thermionic emission from a tungsten filament and accelerated by a high voltage potential down a column to the anode. The wavelength of the x-rays are characteristic of
the anode target used. For example, the most commonly used target Cu produces $K_\alpha$ lines with wavelengths 1.54056 Å and 1.54439 Å.

![Figure 2.20: Graphical representation of Bragg’s formulation which describes a crystal in the form of planes of scattering centres and the distance between successive planes is $d$.](image)

Interference patterns produced by the interaction of x-rays with a crystalline material may be described in real-space by Bragg’s formulation. Here the structure is defined by periodic planes of atoms, which behave as coherent scattering centres separated by an inter-plane distance $d$ as represented in figure 2.20. The crystal lattice essentially behaves like a diffraction grating for the incident x-rays and x-rays exiting the material from successive planes will constructively interfere for specific conditions for a given specimen. The position, intensity and breadth of diffraction peaks can be used as ‘fingerprint’ identification for a particular material. Diffraction will be observed provided that conditions satisfy Bragg’s Law which states

$$n\lambda = 2d \sin \theta$$

(2.29)

where $n$ is an integer, $\lambda$ is the x-ray wavelength, the $d$-spacing is the separation between neighbouring planes of atoms and $\theta$ is the angle of incidence.

In the context of this study, a PANalytical X’pert Pro diffractometer was used to perform powder x-ray diffraction in order establish the polycrystalline nature of the $Y_3Fe_5O_{12}$ specimen used to demonstrate the operation of the SQUID FMR set-up in chapter 6. The sample was provided by colleagues in Bell labs, in the form of a die pressed cylinder typically used in a microwave filter assembly. Samples were carefully
cut from the cylinder using a diamond saw and for the case of XRD ground to a fine powder using a pestle and mortar.

### 2.5.2 Energy Dispersive X-Ray Spectroscopy (EDX)

EDX is a compositional analysis tool typically done in combination with scanning electron microscopy, which allows element identification and measurement of their relative proportions. Here, the sample is bombarded with a focused beam of high energy electrons. These electrons will interact with a core level electron, resulting in it’s removal and the generation of a vacancy. Higher level electrons will occupy the core level as this is the lowest energy configuration. In transitioning from a higher level to a lower level an x-ray is emitted, the energy of which is equal to the energy difference between the associated levels. X-ray lines and intensities produced will be characteristic for a specific element. This technique can be used selectively and surface elemental maps can be produced by rastering the electron beam across the sample’s surface. The spatial resolution is driven by the penetration depth of electrons within the material, which is ultimately governed by the material’s density and also the energy of the incident electrons. This technique is best suited to ultra-thin samples, where the electron beam is not cable of dispersing within the film.

To avoid charging in insulating materials the sample is coated with a thin layer ($\approx 10$ nm) of a highly conductive material such as Au or graphite which provides a conductive pathway to ground. Therefore Au or C x-ray lines are commonly observed in spectra.
Chapter 3

Microstrip Based Contactless Microwave Reflectometry and Galvanic Pulsing

3.1 Motivation

Transparent conductors have long played a vital role in optoelectronic devices such as electronic displays, LEDs and solar cells. In particular the doped metal oxide, indium tin oxide (ITO) has been the workhorse of these devices. However, due to the scarcity of indium and the brittle nature of ITO [58] there has been a drive to develop new materials with enhanced electrical properties and flexibility to meet the demand for such materials in sensors and displays. Silver nanowire networks (Ag NWNs) have been one interesting material emerging in the field, showing high flexibility, conductivity and transparency [59, 60]. While much effort has been made to exploit the potential of individual nanowires (NW), this has been met with limited fruition due to property variations at such small scales and deposition challenges associated with placement and contacting via electrodes [61]. An alternate focus has been placed on the development and characterisation of random nanowire networks (NWNs) [62]. Here deposition challenges are minimized as NWs can be solution processed and spray-cast into thin films on the desired substrate.

Compared to thin film structures, the known variety of methods for addressing and characterising NWNs is less diverse. Typically DC conductivity measurements are used to characterise electrical transport in such structures [63]. I-V characteristics of such networks are heavily dependent on the connections between individual NWs that compose the network, which in turn is dependent on the NW’s surface passivation layer. However, this is a destructive technique and involves the deposition of Ti/Au and Pd contact electrodes directly onto the networks [64]. This creates a need for a method of measuring electrical transport without direct contact. The primary objective of this chapter is to develop novel contactless microwave-based methods for characterizing...
the electrical properties for a broad range of devices, from bulk to thin-film and also random nanostructures such as NWNs. This includes the use of inductive broadband microwave reflectometry techniques for conductivity analysis, which has the advantage of being non-destructive and contactless. The methodologies of both pulsed time-domain reflectometry (TDR) and continuous wave vector network analysis (VNA) are detailed in section 2.1. These techniques are now demonstrated by showing the analysis of randomly ordered metallic Ag NWNs on dielectric substrates but may be applied to a variety of nanostructures, random or templated and thin-films.

For broadband measurements, planar microstrip or coplanar waveguide are typically used in place of rectangular cavities as they are not limited by cut-off frequencies and provided they are impedance matched to the source, provide a much larger bandwidth. The carrier microstrip used for microwave reflectometry was designed using TX Line (Transmission Line Calculator) software, which uses Wheeler’s equations (see equations 1.16 and 1.17) to provide estimates of the characteristic impedance based on given design parameters. The layout was designed using Diptrace schematic and PCB design software and the transmission lines were then produced commercially by BETA Layout, based on the given designs.

3.2 Pulsed Time Domain Reflectometry of Randomly Dispersed Ag Nanowire Arrays

Samples consist of poly(vinylpyrrolidone) (PVP) surface coated Ag and oxide passivated Ni NWs with diameters of 86 nm and 80 nm respectively. This work is done in collaboration with Prof. J. Boland’s\textsuperscript{1} group who produce networks by spray-casting into thin films of varying optical transmittance ($\approx 65 - 90 \% T$) on dielectric (borosilicate glass) and semi-conducting (Si) substrates with dimensions $2 \times 2$ cm$^2$. They have also performed the calculations of transmittance in the visible spectra ($\% T$) for the samples discussed in this chapter. For those deposited on non-transparent substrates, calibration samples deposited on transparent substrates were measured.

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For these broadband microwave measurements, the dielectric sample is placed in the centre of the transmission line and by monitoring the reflected and transmitted signals that result, one can determine characteristic impedances and distributed capacitances for the aforementioned NWNs. Figure 3.1 displays the total reflected and transmitted voltages for Ag NWNs subjected to the pulsed signal \( E_i = -0.25 \text{ V} \) from the TDR. Small reflections are observed from front-end SMA connectors and solder joints from the SMA end-launchers to the microstrip transmission line. 100 % optical transmittance corresponds to a blank borosilicate glass dielectric substrate. Reflections arising from the metallic nanowire arrays appear separated in time as capacitive discontinuities within the cumulative circuit reflections and therefore can be easily identified.

![Figure 3.1: Direct pulsed TDR measurements of reflection and transmission for Ag NWNs.](image)

Treating the reflected and transmitted signals separately and subtracting the background signal obtained in the absence of a sample figures 3.2 and 3.3 are obtained. As is evident from the figures, the reflected signal originating from the NWNs increases with increased network density, due to backscattering of the pulse when it encounters discontinuities in the form of NW junctions. The reflected voltage \( E_r \) is averaged over the length of the sample and the load impedance of the network is then calculated according to equation 1.18. Plotting \( Z_L \) of the network against the optical transmittance...
figure 3.4 is obtained, which displays a linear correlation between the two.

**Figure 3.2:** Reflected voltage ($V$) as a function of time ($t$).

**Figure 3.3:** Transmitted voltage ($V$) as a function of time ($t$). The plots are fitted with an exponential decay curve, assuming a capacitive discharge through the characteristic 50 Ω impedance ($R$) of the line in order to extract the voltage across the capacitor $V_c$, the maximum voltage $A$, the time constant $\tau = RC$ and from this the capacitance $C$ as shown in the table inset.
Figure 3.3 shows the capacitative discharge of the NWN through the standard 50 Ω impedance (R) of the line. The capacitive decay is described by

\[ V_c = V_0 e^{-\frac{t}{RC}} \]  

(3.1)

where \( V_c \) is the voltage across the capacitor and \( V_0 \) is the maximum voltage. By fitting this to the decay as shown by the solid fit lines in figure 3.3 estimates of the distributed capacitances of the networks are obtained, which as shown in figure 3.4 follows a polynomial decay \( (C = 0.010 pF(100 - T)^2) \) with decreasing density.

![Figure 3.4: Load impedance \( Z_L \) and distributed capacitance \( C_L \) as a function of optical transmittance for Ag NWNs.](image)

### 3.3 Broadband Continuous Wave Vector Network Analysis of Ag Nanowire Arrays

As mentioned previously, continuous wave broadband VNA has proven higher sensitivity and signal to noise than TDR but both can be used in conjunction to give an extensive picture of location and amplitude of reflections resulting from the test material’s composition, as well as wave propagation in devices. The same Ag NW arrays discussed in section 3.2 are characterised using a two port CW (300 kHz-3 GHz) vector network analysis system. The noise-floor of a typical VNA system ranges from -150
dB to -90 dB, which largely depends on the receiver bandwidth (RBW) used. The receiver bandwidth is the intermediate-frequency (IF) bandwidth and the noise-floor can be reduced by narrowing this bandwidth. There is however a trade-off between noise-reduction and scanning rate, as a very narrow IF bandwidth will increase the measurement time significantly. To achieve the best signal-to-noise for the frequency sweep, the optimum input power is established. Figure 3.5 shows both (a) the reflected and (b) the transmitted signal amplitude as a function of frequency for a blank substrate where the input power is varied from -34 dBm to 5 dBm. The largest dynamic range is observed for an input of +5 dBm, the maximum output of the signal generator, at such frequencies. The optimum +5 dBm is used for successive testing.

Figure 3.5: (a) Reflected and (b) transmitted amplitude (dB) as a function of frequency (GHz) for a blank substrate, varying the power of the incident signal.

As can be seen in figure 3.6, the signals appear as a summation of all reflections along
the transmission media, represented as a function of a swept frequency. BG indicates reflection and transmission from the transmission line in the absence of a sample and 100 % transmittance is for a blank dielectric substrate, in this case borosilicate glass. In agreement with the TDR data, the CW reflection analysis shows enhancement of the reflected amplitude with decreased % $T$. The low-frequency (< 1 GHz) dispersion is likely due to the fact that the wavelengths are far greater than the sample dimensions. Detection of frequency responses below 300 kHz are inaccessible with this system.

![Graph](image_url)

**Figure 3.6:** Reflected amplitude (dB) as a function of frequency (GHz) for Ag NWNs of varying % $T$. 
Figure 3.7 shows the transmitted signal intensity as a function of the swept frequency, for the same Ag NWNs. The $S_{21}$ transmission data follows an opposing trend to the reflection spectra, as the transmission increases with optical transmittance. Again, a strong frequency dispersion below 1 GHz is observed.

Figure 3.8: Phase of the reflected signal as a function of frequency.
As previously discussed, the phase of the reflected and transmitted signals are easily extracted using CW VNA. Figures 3.8 and 3.9 show the phase of the reflected and transmitted waves respectively. Here the phase is not an absolute measure but a ratio which compares the phase of the incident signal to the phase of the response signal (reflection). Knowledge of both the magnitude and phase of the signals allows for comprehensive signal dephasing and analysis of the real and imaginary signal components and subsequently extraction of the background (microstrip) subtracted linear (%) reflections and transmissions. Equivalent linear responses for figures 3.6 and 3.7 are shown in figure 3.10 where (a) represents the enhanced reflections resulting from the NWNs and (b) is the reduction in the transmitted signal. Small reflections of the order of 2 % are determined to originate from the dielectric substrate (T = 100 %). For the densest of the arrays examined, T = 70 %, a reduction of up to 50 % of the incident signal that reaches the S_{21} receiver is observed.
3.4 Thermal Activation of NW Networks using Direct Galvanic Pulsing

Within the network the connection between the individual nanowires consists of resistive junctions, in the form of a surface polymer or native oxide coatings. It has been previously shown that when an external electrical field is applied across such metallic NWNs, the junctions begin to breakdown and the conductivity of the network evolves under electrical stressing [64]. When a low resistance pathway from source to drain has been created, the network is said to be activated. Here the aim is to reach this transport limited avalanche regime for wire to wire contacts by using CW reflectometry.

Figure 3.10: Linearised (a) reflection and (b) transmission responses to an incident microwave stimulus for Ag NWNs of varying density.
in a more potent form where the networks are subjected to relatively short (0.5s) bursts of high power CW pulses.

Figure 3.11: Thermally evaporated slotline waveguides for direct galvanic pulsing of metallic nanowire networks.

Slotline structures are deposited via thermal evaporation through a shadow mask directly onto the NWNs on Si substrates as seen in figure 3.11a. Two different electrode spacings 30 µm and 120 µm were investigated. Each slotline was connected to the single port VNA. Bursts of CW power were applied starting at -34 dBm and increasing by 5 dBm up to 21 dBm. The sheet resistance was measured after each burst of CW. After a certain number of CW pulsings, a small but perceptible reduction in the sheet resistance of the networks was noted. After continuous pulsing, a point was reached where the conduction saturated and no further reductions in the sheet resistances $R$ were noted. Figure 3.12 shows a representative plot of the evolution of the sheet resistance for a 65.9 % $T$ Ag NWN under microwave stressing, which indicates an irreversible transition from a high resistance to low resistance state.
If the network contained between the two co-planar electrodes is modelled as a metallic film of length $D = 120 \ \mu\text{m}$ (the electrode spacing), having an area of contact with each electrode of $A = 2dL$ ($d$ being the NW diameter ($d = 86 \ \text{nm}$) and $L$ the length of the entire network ($L = 2 \ \text{cm}$)), assuming that on average two nanowires are overlapping at any point within the network, approximations for the resistivity $\rho$ and conductivities $\sigma$ can be obtained

$$\rho = \frac{RA}{D} = \frac{2RdL}{D} \quad (3.2a)$$

$$\sigma = \frac{1}{\rho}. \quad (3.2b)$$

As shown in Table 3.1 the initial conductances of $3.9 - 0.8 \ (\mu\Omega\text{m})^{-1}$ increase to $4 - 1.4 \ (\mu\Omega\text{m})^{-1}$ following successive pulsings (-20 dB to 20 dB, each CW pulsing lasting $< 1 \ \text{s}$) indicating partial activation of the network. This technique is limited by the highly reflective nature of the metallic NWs. Also the initial sheet resistance is too low to allow impedance matching to the swept microwave source.
Table 3.1: Monitoring the changes in conductances for Ag NWNs following microwave pulsing.

<table>
<thead>
<tr>
<th>Optical Transmittance (% T)</th>
<th>$\sigma_1$ ($\mu\Omega$m$^{-1}$)</th>
<th>$\sigma_2$ ($\mu\Omega$m$^{-1}$)</th>
<th>$\Delta\sigma$ ($\mu\Omega$m$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>65.9</td>
<td>3.97±0.05</td>
<td>4.009±0.05</td>
<td>0.039±0.07</td>
</tr>
<tr>
<td>75.4</td>
<td>4.543±0.04</td>
<td>4.598±0.06</td>
<td>0.055±0.06</td>
</tr>
<tr>
<td>90.6</td>
<td>0.8±0.02</td>
<td>1.405±0.02</td>
<td>0.605±0.03</td>
</tr>
</tbody>
</table>

Figure 3.13: Monitoring the change in the conductance of Ag NWNs as a function of optical transmittance % $T$. The increase in the conductance is shown to scale with the optical transmittance of the network.

Figure 3.13 shows that the increase in the conductance scales with the optical transmittance of the network. This is due to the fact that the sparser networks (larger % $T$) are more resistive and better matched to $Z_0 = 50 \text{ }\Omega$ allowing greater power absorption. No changes in conductivity were observed for the 30 $\mu$m electrode spacings as the initial sheet resistance was too low to allow matching to the frequency source.

3.5 Summary

The main aim of this chapter was to evaluate microwave reflectometry as a potential means of contactlessly characterising the electrical properties of randomly structured
media, in this case disordered of arrays of metallic NWs spray cast on dielectric substrates. The techniques shown are mainly limited by the conductivity of the test material. For a highly conductive material, for example Au, reflections would be apparent for thicknesses in the tens of nanometres, whereas a dielectric material would need to have millimetre thicknesses to observe a similar microwave response. The conduction properties for a range of Ag nanowire arrays have been non-destructively characterised using both continuous wave VNA (< 3 GHz) and pulsed TDR analysis (< 50 GHz). In the pulsed regime, the impedance of Ag NWNs has been shown to scale linearly with the optical transmittance of the network. It is shown for Ag NWNs that denser (smaller \( \% T \)) networks display enhanced reflected signals due to the increased number of capacitive junctions. This is in good agreement with the results obtained using the swept frequency reflectometer (VNA), which also show a systematic increase in the reflected signal with increasing network density (decreasing optical transmittance). Hence it can concluded that such contactless techniques, conventionally used for analysing thin-film structures have application in the characterisation of conductive disordered systems.

In a more destructive technique an attempt was made to achieve the ‘activated,’ low-resistance state for the NWNs using direct galvanic pulsing of CW power via a slotline geometry, thermally deposited directly onto the arrays. Marginal increases in conductivities of such networks are shown after the CW pulsing but as mentioned in Section 3.4 the technique is limited by the reflective properties of the NWNs, which behave like metallic thin films.
Chapter 4

High Frequency, Cavity-based,
Narrowband Microwave Reflectometry

4.1 Motivation

For high frequency, high sensitivity applications cavity waveguides have an advantage over their planar counterparts. They are composed of only a single conductor and as a result can transmit greater power than microstrip or CPW transmission lines, as the power handling capability is directly related to the separation between conductors in planar devices. As electric and magnetic fields are contained within a uniform dielectric, dielectric and radiation losses are also minimized. As mentioned in chapter 1 rectangular waveguides are efficient in operating in bands from 0.32 GHz to 325 GHz. To access this full frequency range multiple waveguides with varying dimensions must be used, as the cut-off frequency is largely dependent on the cavity profile. For this study, frequency bands of operation are classified according to the IEEE standard letter designations for radar-frequency bands [65] as shown in table 4.1.

Table 4.1: IEEE standard letter designations for radar-frequency bands

<table>
<thead>
<tr>
<th>IEEE</th>
<th>Frequency (GHz)</th>
<th>EIA</th>
<th>WG (w x h) mm</th>
<th>Free Space $\lambda$ (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>X</td>
<td>8-12</td>
<td>WR90</td>
<td>22.86 x 10.16</td>
<td>3.75-2.5</td>
</tr>
<tr>
<td>Ku</td>
<td>12-18</td>
<td>WR62</td>
<td>15.8 x 7.9</td>
<td>2.5-1.67</td>
</tr>
<tr>
<td>K</td>
<td>18-26.5</td>
<td>WR42</td>
<td>10.67 x 4.32</td>
<td>1.67-1.11</td>
</tr>
<tr>
<td>Ka</td>
<td>26.5-40</td>
<td>WR28</td>
<td>7.11 x 3.56</td>
<td>1.11-0.75</td>
</tr>
</tbody>
</table>

One application which relies on the ability to access high frequency bands is in the evaluation of materials for multi-band or broadband electromagnetic interaction (EMI) shielding applications. Unlike other methods of characterising EMI shielding, such as the coaxial holder or dual transverse electromagnetic cell methods which tend to suffer from a limited frequency range, cavity based EMI shielding characterisation
has proven excellent conformity over an unparalleled frequency range. In this chapter, finite band cavity waveguides are designed according to CST Maxwell solutions and used in conjunction with vector network analysis (VNA) (7 - 20 GHz) or pulsed TDR (21 - 42 GHz) to characterise the high frequency conductivity of Ag films and grids for electromagnetic interaction (EMI) shielding applications. This work is done in collaboration with Prof. J. Coleman’s group who prepare Ag films and grids by inkjet printing dispersions of Ag flakes in solution onto flexible, alumina-coated PET substrates. Here, film thickness is controlled by the concentration of Ag in the solution. Figure 4.1 shows a histogram of the distribution of lengths of the Ag flakes measured by Scanning Electron Microscopy (SEM) performed by Adam Kelly of Prof. J. Coleman’s group. It indicates that the majority of flakes are between 200 nm and 800 nm in length with the mean flack size being \( \approx 500 \) nm. Fabrication is done in a single pass thermal inkjet printing approach. The thickness of the Ag films are estimated to be 10 nm per pass through the inkjet printer and thicker films are produced by multiple printer passes.

![Figure 4.1: Distribution of flake length in the inkjet printed Ag films.](image)

The K and Ku-band cavities designed in this chapter are configured according to EIA standard dimensions for rectangular waveguides [66]. The microwave scattering parameters through the films and grids are quantified from 7 to 42 GHz in order to evaluate

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the dispersive nature of the inkjet materials at high frequencies.

4.2 Applications and Mechanisms of Electromagnetic Interference Shielding

In the design of electrical and magnetic devices, unwanted interaction with external electromagnetic fields is of great concern. Such external fields arise from many daily encountered sources such as radio transmitters, mobile phones, WiFi transmitters and radar and can have detrimental effects on for example TV, radio or mobile phone reception, or could effect the functionality of military or medical devices. Likewise, when designing radiative electrical devices it is important to establish the electromagnetic compatibility with external devices and governmental bodies have put regulations on the external fields a device can produce [67]. In recent years developing novel functional materials that reflect or absorb electromagnetic energy has been of great interest [68–70]. The main EMI shielding mechanisms, reflection, multi-reflection and absorption are shown in figure 4.2, showing a free standing shielding material. Metals are the most commonly used EMI shielding material, which rely primarily on the reflection of incident electromagnetic radiation. This mechanism for shielding is driven by mobile charge carriers in the metal which interact with the external electromagnetic radiation creating an impedance mismatch between the wave in free space and within the shielding material, such that a large portion of the radiation is reflected. For metallic systems, multiple reflections from surfaces or interfaces within the shield also serve to enhance the overall shielding capability of the material.

![Figure 4.2: The mechanisms of EMI shielding, reflection, re-reflection and absorption are shown.](image)
EMI shielding does not actually require a percolation pathway through the material but it’s effectiveness is enhanced by increasing conductivity. This is mainly due to the secondary shielding mechanism, absorption. Within the metallic shield, part of incident field will be sustained beyond the shield’s surface and will decay as it travels though the media. The decay rate is governed by the material’s conductivity and permeability. The depth in bulk metallic materials at which the field drops to 1/e of the incident value is termed the skin depth $\delta$ and is described by

$$\delta = \frac{1}{\sqrt{\pi f \mu \sigma}} \quad (4.1)$$

where $\omega$ is the frequency of the interacting wave and $\sigma$ is the shield’s conductivity. For the case of metallic thin films where the thickness is less than the skin depth, most of the EM radiation will be reflected or absorbed for films beyond a critical thickness, described by

$$d_0 = \frac{c}{2\pi \sigma} \quad (4.2)$$

As $\sigma$ has a frequency dependence, the critical thickness will shrink with increasing frequency and theoretically absorb more efficiently.

While shielding using bulk metallic sheets can be very effective, it is also bulky and has issues with rigidity and hence is not suitable for protecting light-weight, portable devices. As such, metal coatings produced by electroplating or vacuum deposition are more commonly used. These techniques can however be costly. A more economical approach would be inkjet printing of such materials on flexible dielectric substrates using commercially available printers. For the case of non-continuous Ag films produced by inkjet printing, shielding relies on the formation of 3-dimensional conducting pathways within the film matrix. The minimum thickness which results in conductive pathway formation is know as the percolation threshold. The figure of merit for EMI shields is often represented in terms of the shielding effectiveness (SE). In terms of the scattering parameters, which are simply the mathematical representations of the reflection and transmission through the system (as described in chapter 2), the reflective and
absorptive SE contributions are given by

\[
SE_{\text{Reflection}}[dB] = 10 \log \left( 1 - 10^{\frac{S_{11}}{10}} \right) \tag{4.3a}
\]

\[
SE_{\text{Absorption}}[dB] = 10 \log \left( \frac{10^{\frac{S_{21}}{10}}}{1 - 10^{\frac{S_{11}}{10}}} \right) \tag{4.3b}
\]

respectively. For a two-port measurement, where excitation is provided from both
port 1 and 2 \( S_{11} = S_{22} \) and \( S_{12} = S_{21} \). Inkjet printing Ag allows for easy tuning
of the EMI shielding properties via film composition (flake size, thickness) and opens
the possibility of printing other materials for electrically active shielding components.
The EMI shielding properties of the test materials are analysed using a rectangular
waveguide assembly (two straight sections and two SMA to waveguide adaptors) and a
VNA (see section 2.1.2). The VNA monitors the phase and magnitude of the scattering
parameters for the waveguide assembly when a sample is clamped between the two
straight waveguide sections.

\[\text{Figure 4.3: Schematic of experimental set-up for measuring the EMI shielding of inkjet printed films and}
\text{grids, showing the VNA, the SMA to waveguide adaptors and the straight waveguide sections with the sample}
\text{clamped between.}\]

4.3 X-Ku-Band Measurements

X (8-12 GHz) and Ku (12-18 GHz 'Kurtz-under' in reference to short wavelength
radar) bands are classified as a subset of the super high frequency region of the electro-
magnetic spectrum, dealing with frequencies from 3 to 30 GHz. This band corresponds
to centimetre wavelengths ranging from 3.75 cm to 1.67 cm for the highest frequency.
The primary uses of the X and Ku-bands include radar, wireless networks and satellite
communications, with a portion of the X-band being designated exclusively to deep space communications. This incurs power restrictions on commercially available signal generators capable of accessing these bands. X-band is widely used in military devices and communications, necessitating extensive study of potential shielding materials.

4.3.1 X-Ku-Band (7-14 GHz) EMI Shielding effectiveness of Inkjet Printed Ag Films

Figure 4.4 shows the evolution of the $S_{11}$ reflection scattering parameter (dB) as a function of frequency for different thicknesses of Ag Films. The spectra are shown to be flat band over the bandwidth with some features consistent in all spectra, which can be attributed to reflections in the measurement set-up, such as those created at the SMA connector to cavity adapters. These features may theoretically be extracted by careful decoupling of the geometric reflections from the overall signal but as they are only a minor contribution to the measured signal, this treatment is beyond the scope of this study. A small increase in the $S_{11}$ is observed after 9 passes with the inkjet printer, which is equivalent to a thickness of 90 nm. The reflection increases systematically and then begins to plateau beyond 12 passes. After 15 passes the reflection scattering parameter has essentially saturated.
Examining the $S_{12}$ for the same frequency band, as seen in figure 4.5 a similar transition is observed. After 9 passes the transmission drops dramatically, equivalent to just over 60 % reduction in the transmitted signal, on a linear scale. For the case of the thicker samples (> 200 nm) the transmitted signal approaches the detection
limit of the VNA ($\approx -100$ dB and with comprehensive decoupling of line reflections
$\approx -120$ dB [71]), hence there is a noticeable decrease in the signal to noise ratio.

Figure 4.6 and Figure 4.7 show the $S_{11}$ reflection and $S_{12}$ transmission respectively, averaged over the bandwidth of the Ku-band cavity as a function of the number of passes through the inkjet printer. The bare PET substrate is characterised as having an average $S_{11}$ of -17 dB and an $S_{12}$ parameter of -3dB. When the cavity is completely blanked off with a brass plate with a thickness far greater than the penetration depth of the microwave excitation (i.e. 0 % transmission) the $S_{11}$ and $S_{12}$ are measured as -3 dB and -90 dB respectively. Both plots indicate a percolation threshold of 90 nm in thickness beyond which the films transition from an insulating to a conductive state. After 15 passes (150 nm) the conductivity plateaus and the films are shown to be fully reflective.

Figure 4.6: Average X-Ku-band $S_{11}$ reflection as a function of the number of passes
For calculating the absorption ($A$) of the silver films

$$A = 1 - (T + R)$$

(4.4)

where $T$ and $R$ are the normalized linear transmission and total reflection, accounting for multiple reflections, respectively. Figure 4.8 shows the normalised linear reflection, transmission and absorption as a function of the number of passes. At the percolation threshold of 9 printing passes ($\approx 90$ nm), the system switches from a microwave transparent regime to reflective state. The transmission decays at -46 % per pass between from 9 to 11 passes, and begins to plateau thereafter. The films are shown to have an absorption of up to 50 % just above the percolation threshold for the Ag flake matrix. Following the transition from a transmissive to a reflective state the average baseline absorption is larger than prior to the transition. This may be attributed to a multi-reflection mechanism whereby the incident signal is reflected at the farthest interface and then reflected again at the nearest interface.
4.3.2 X-Ku-Band EMI Shielding effectiveness of Inkjet Printed Ag Grids

For EMI shielding in space and satellite applications being able to restrict the mass of the shield, while maximising the shielding behaviour is of great importance. One way to do so is to reduce the thickness of metallic films, however in doing so the EMI shielding effectiveness is likely compromised. Alternatively, a great deal of study has been put into the development of fine mesh metallic grids for EMI shielding, which reduce the mass while retaining the shielding capabilities [72, 73]. For the case of a metallic film with regular ordered square apertures of length D the shielding effectiveness (SE) may be described by

\[
SE(dB) = 20 \log \left( \frac{\lambda}{2D} \right) - 20 \log(n) \text{ when } D \leq \frac{\lambda}{2} \quad (4.5a)
\]

or

\[
SE(dB) = 0 \text{ when } D \geq \frac{\lambda}{2} \quad (4.5b)
\]

such that a maximum in radiated energy through the shield occurs when D is equal to half the wavelength \( \lambda \) of the incident radiation. Here, \( n \) is the total number of openings within a half wavelength.
This section probes the effect of systematic modification of the line spacing on the shielding characteristics for 200 nm thick Ag grids, deposited by inkjet printing. Figure 4.9 shows two representative grids, the left being the finest mesh investigated. The metal line-width remains constant (t=1 mm) for each grid and the separation between lines is varied.

Table 4.2 gives a brief description of each grid studied in terms of separation between lines, metallic fill factor and microwave transmission. For the case of the sparser meshes, placement within the cavity will have an effect on the microwave reflectivity and therefore special care was taken to maximize the metallic fill within the cavity area.

<table>
<thead>
<tr>
<th>D (mm)</th>
<th>Fill Factor (%)</th>
<th>$S_{12}$ Transmission (dB)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Blank PET</td>
<td>0</td>
<td>-4.02</td>
</tr>
<tr>
<td>6</td>
<td>26.5</td>
<td>-9.32</td>
</tr>
<tr>
<td>5</td>
<td>30.6</td>
<td>-10.57</td>
</tr>
<tr>
<td>4</td>
<td>36</td>
<td>-12.79</td>
</tr>
<tr>
<td>3</td>
<td>44</td>
<td>-16.32</td>
</tr>
<tr>
<td>2</td>
<td>56</td>
<td>-23.41</td>
</tr>
<tr>
<td>1</td>
<td>75</td>
<td>-34.64</td>
</tr>
<tr>
<td>Continuous Film</td>
<td>100</td>
<td>-48.85</td>
</tr>
</tbody>
</table>
The as-detected X-Ku-band reflectance and transmittance spectra are displayed in figures 4.10 and 4.11 and follow opposing trends, with the reflection component decreasing and transmission increasing for increasing sparsity within the mesh. Examining figure 4.11 it is evident that the meshes possess a non-dispersive frequency response over the bandwidth.

**Figure 4.10:** Evolution of the X-Ku-band $S_{11}$ with variation of the pitch between conductive lines.

**Figure 4.11:** Evolution of the X-Ku-band $S_{12}$ with systematic alteration of the pitch between conductive lines.
The X-Ku-bandwidth averaged $S_{11}$ reflection and $S_{12}$ transmission are displayed in figure 4.12. For a fill factor of 100% i.e. a continuous 200 nm thick Ag film, $S_{11}$ and $S_{12}$ are determined to be -2.8 dB and -48.9 dB, respectively. By reducing the fill factor of the continuous film by 25% a 16 dB increase in $S_{11}$ is observed. Figure 4.13 displays the normalised linear scattering parameters represented in the previous figure. The
transmission is observed to decay with increasing metallic fill factor. The 30.6% fill factor grid appears to be an anomaly within the representative data set and may be accredited to issues during the fabrication process resulting from the printing approach used. A near line quality (NLQ, Multiple passes with a slight offset and higher ink drop density) strategy could potentially improve the line conformity. For a fill factor of 75%, an attenuation of the transmitted signal and enhancement of the reflected signal of 99.9% and 90% respectively in comparison to the blank PET are observed. Therefore by reducing the fill factor by 25% the transparency and material weight can be reduced without significantly sacrificing the EMI shielding properties.

4.4 K-Band (18-24 GHz)

Operating between 18 and 24 GHz, the K-band experiences atmospheric attenuation due to resonant absorption at 22.3 GHz by water vapour, hence this band is more commonly used for short range communications and radar. One application which utilizes rain fade effects is in weather radar systems, where precipitation can be measured by monitoring the attenuation of radiative signals between two antennae. Shielding materials for operation in this band are frequently designed for the protection of short-range military devices.

4.4.1 Computer Aided Design and Electromagnetic Simulation of K-band SMA-Cavity Adaptors

In the design of high frequency waveguides, simulations based on Maxwell’s equations for electromagnetism can be an essential tool in visualizing TE or TM propagation, for a given set of design parameters. CST Microwave Studio allows for simple CAD generation and alteration to achieve the desired operating frequency band.
Figure 4.14a shows the schematic of a right-angle SMA coaxial to cavity transition adapter optimized for K-band operation. The SMA itself consists of a central conductor acting as an antenna surrounded by a PTFE dielectric which extends to the inner side of the cavity wall and the probe dimensions are optimized to have a 50 Ω impedance. This is encased in a circumferential conductor, connected to the outer wall of the waveguide. These SMA connectors show optimal performance for DC to 18 GHz, as PTFE becomes dispersive at higher frequencies. The rectangular cavity is interfaced with the RF coaxial via a simple probe antenna that protrudes to the mid-point of the waveguide’s height and is positioned at a distance D from a backplane short. This length essentially forms a secondary shorted transmission line, behind the probe, where a small portion of the incident energy at the antenna propagates towards and is reflected at the backplane. D is generally less than a quarter of the wavelength of the frequency at the centre of the operation band, such that EM energy reflected from the shorted backplane to the probe interacts constructively with the incident wave and propagates down the length of the cavity.

To simplify simulations the waveguide is treated as a seamless copper cavity, uniformly filled with air and only the primary TE\textsubscript{10} mode is considered. In reality the apparatus is designed in sections, two transition adapters and two straight waveguide lengths, between which the test sample is placed, as in figure 4.14b. Figure 4.15 shows the electric field distribution in a K-band cavity with two coaxial stimuli. This cavity
is excited at 15 GHz via port 1. The length of the cavity and adapter pieces are designed to be in quarter wavelength multiples such that a sample, when positioned in the centre of the assembly and subjected to the two simultaneous stimuli, experiences a constructive node in the stimuli as indicated by peak in the E-field intensity in figure 4.15.

Figure 4.15: Simulating E-field distribution in rectangular waveguides optimized for K-band (18-27 GHz) measurements. Only the primary TE_{10} mode is considered.

Figure 4.16: Simulating cut-off frequencies for TE_{10} mode propagation in rectangular waveguides optimized for K-band (18-27 GHz) measurements.
The simulated $S_{11}$ Reflection and $S_{12}$ transmission are shown in figure 4.16. The cut-off frequency is characterised as the frequency below which a sharp decrease in the $S_{12}$ scattering parameter is observed. The spectra indicate a cut-off frequency of 15 GHz and an operational bandwidth of 15 to 24 GHz. Comparing this with the $S_{11}$ and $S_{12}$, seen in figure 4.17 for cavities fabricated using the same geometries stipulated in the simulations, measured using a vector network analyser in a two port configuration, a cut-off frequency of 15 GHz is also observed.

![Figure 4.17: Measuring the cut-off frequencies for cavity waveguides fabricated to operate in the K-band.](image)

The waveguides described here are fabricated using commercially available WR42 (see table 4.1) rectangular tubes cut to the desired length using a diamond saw. WR42 connection flanges are custom built by the School of Physics mechanical workshop to tightly fit the WR42 and allow for connection of the waveguides. These are tin soldered in place and the surface of each flange is finely polished using a radial sander to allow a close connection between the waveguides. For the adapter pieces one end of the tube is terminated with a copper plate to cover the opening and soldered in place. The end plate is then sanded down to fit the dimensions of the WR42. Additionally an engraving tool was used to puncture a hole in the WR42 a distance D from the
backplane. An SMA Teflon covered probe is cut to the correct length \((h/2)\) using a diamond saw and the end of the probe is polished flat. The probe is inserted in the opening in the WR42 and after an initial test using the VNA to ensure the probe is in the correct position, is soldered in place. A very sharp increase in the \(S_{12}\) from -100 dB to -3 dB indicates low-loss cavities, with a dynamic range (i.e. difference between maximum and minimum signals measured) greater than 90 dB, which is comparable with commercially produced X-Ku-band waveguides used in the previous section. The operational bandwidth is shown to be 15 to 20 GHz. The VNA has a bandwidth of 100 MHz to 20 GHz, hence the upper limit of the frequency band is not observed.

4.4.2 K-Band EMI Shielding effectiveness of Inkjet Printed Ag Films

Figure 4.18 and 4.19 show the bandwidth averaged \(S_{11}\) and \(S_{12}\) scattering parameters respectively. As is also shown in section 4.3, a transition from transmissive to shielding behaviour is observed for samples with thicknesses greater than 9 passes or approximately 90 nm.

![Graph of average K-band \(S_{11}\) reflection as a function of the number of passes](image)

*Figure 4.18:* Average K-band \(S_{11}\) reflection as a function of the number of passes
Analysing the normalised linear scattering parameters, as in figure 4.20 the decay in the transmission at the shielding transition between 7 and 9 passes is slower at 44 \% per pass than that shown for the X-Ku-band (see figure 4.8), indicating that higher frequency signals penetrate deeper into the shielding material. Enhanced absorption for all films in comparison to X-Ku-band absorption (see figure 4.8) is observed, with most films exhibiting above 10 \% absorption, peaking at 60 \% just above the percolation threshold.
Figure 4.20: Percentage of K-band incident signal transmitted, reflected and absorbed as a function of the number of passes.

4.4.3 K-Band EMI Shielding effectiveness of Inkjet Printed Ag Grids

Once again the reflection and transmission coefficients are measured to evaluate the multi/broadband operation of non-uniform materials for EMI shielding applications where mass restrictions are an issue. As seen in figure 4.21, for a uniform film $S_{11}$ and $S_{12}$ are measured as -4 dB and -70.6 dB. On decreasing the metallic fill factor to 75 % the $S_{12}$ is determined to be -34.6 dB. However, if we examine the normalised scattering parameters as in figure 4.22 it may be seen that this corresponds to only a 0.09 % increase in the transmission, relative to the continuous Ag film. So exceptional shielding performance is indeed attainable for non-uniform films provided the metallic fill factor is kept above a critical threshold, which for the Ag grids is approximately 55 % i.e. a 2x2 mm aperture (at 55 % less than 2 % of the incident pulse is transmitted.). Above this threshold, varying the pitch between metallic lines allows us to easily adapt the metallic fill and hence the mass of the shield without dramatically effecting the microwave transparency.
Figure 4.21: Average K-band $S_{11}$ and $S_{12}$ in dB as a function of the percentage metallic fill factor.

Figure 4.22: Normalized linear scattering parameters as a function of varying metallic fill factor in the K-band.
4.5 Ka-Band Measurements

In recent years the Ka (Kurtz-above) band has drawn a great deal of interest for long range satellite communications and radar and high resolution short range radar for military aircraft systems. This is mainly due to the higher data transfer speeds than the Ku-band. Additionally, scaling to the higher frequencies also reduces the size of antennae and devices, making it a favourable option in applications where weight and size are important limitations. This band operates between 26.5 and 40 GHz and hence does not suffer from the same level of atmospheric attenuation experienced by the K-band.

4.5.1 Computer Aided Design and Electromagnetic Simulation of Ka-band SMA-Cavity Adaptors

Figure 4.23a shows the cad of an SMA to cavity transition adapter, which has been optimized for operation in the Ka-band, which follows the same design criteria as the adapters described in section 4.4.1. As discussed in chapter 2 the waveguide dimensions dictate the frequency of operation, such that Ka waveguides are smaller than the K-band previously described. Figure 4.23b shows the resulting fabricated Ka-band waveguide.

![CAD schematic of rectangular waveguide to SMA adapter optimized for Ka-band (26.5-40 GHz) measurements.](image1)

![Picture of Ka-band cavities](image2)

**Figure 4.23:** Ka-band Cavity
Again, the waveguide section and adapter lengths are designed in $\lambda/4$ multiples, where $\lambda$ is the wavelength at centre frequency for the desired band such that simultaneous excitation from both ports produces constructive interference at the waveguides centre, where the sample is positioned. Figure 4.24 shows the E-field distribution in a 9 cm seamless cavity excited via two SMA to coaxial adapters at each end.

Figure 4.24: Simulating E-field distribution in rectangular waveguides optimized for K-band (26.5-40 GHz) measurements. The waveguides are subjected to a 27 GHz stimulus at port 1. Only the primary TE$_{10}$ mode is considered.

Figure 4.25: Simulating cut-off frequencies for TE$_{10}$ mode propagation in rectangular waveguides optimized for Ka-band (26.5-40 GHz) measurements.
Figure 4.25 represents the simulated $S_{11}$ and $S_{12}$ scattering coefficients over a 20 to 50 GHz bandwidth. Examining only the $S_{12}$ transmission the designed cavities appear to have a lower frequency cut-off of 21 GHz and an upper cut-off around 43 GHz. The spectra appears to show multiple bands of operation, separated by sharply defined EM rejection frequencies where $S_{12}$ drops substantially.

4.5.2 Ka-Band EMI Shielding effectiveness of Inkjet Printed Ag Films

The Ka-band cavities are estimated to operate between 21 to 43 GHz, beyond the range of most commercially available continuous wave VNAs, with the higher end analysers operating up to 20 GHz. As such, a different approach was taken with this band. A pulsed time domain reflectometer, emits two dephased 24 GHz signals. The dephasing and combination of the excitation signals essentially doubles the capable bandwidth of the reflectometer. However this method suffers from the lower signal to noise ratio associated with TDR analysis in comparison to the CW VNA and measuring transmission through films beyond 180 nm is not feasible.

Figure 4.26: Raw TDR spectra obtained for a blank PET substrate.

Figure 4.26 shows the TDR spectra obtained for a bare PET substrate. As measurements are done in the time domain, a complex fast Fourier transform (FFT) is
then needed to convert to the frequency domain. Before computing the FFT, the zero offset must be accounted for by subtracting the average measured transmission, which for the representative spectra is -0.023 mV.

Figure 4.27 shows the Fourier transform of the time domain spectra taken for the bare PET substrate. Seven distinct bands are observed; 21-24 GHz, 24-25.5 GHz, 25.5-28 GHz, 28-30.5 GHz, 30.5-35 GHz, 35-37.5 GHz and 39-42 GHz, with the higher frequency bands being weaker in intensity. This is in agreement with figure 4.25 which indicates multiple bands of operation ranging from 21 to 43 GHz.

As seen in figure 4.28 after 10 passes (≈ 100 nm), all seven bands are shown to be greatly diminished in intensity, with the 21-24 GHz most notably reduced. After 18 passes (≈ 180 nm) as seen in figure 4.29 we approach the noise floor of the measurement and the samples are said to be entirely reflective for this bandwidth.
In order to quantify the average transmission \( T_{\text{avg}} \) for each sample the spectral data in each finite operational bandwidth is integrated over the frequency interval (i.e. resolution bandwidth)

\[
T_{\text{avg}} = \frac{\int_{f_1}^{f_2} \text{Transmission} \, df}{\int_{f_1}^{f_2} 1 \, df}
\]

where \( f_1 \) and \( f_2 \) are the lower and upper frequency bounds for the bandwidth. Figure 4.30 (a) displays the normalized average transmission as a function of the number of passes and in a similar fashion to figures 4.5 and 4.19 a shielding threshold is observed after 9 passes. It is of interest to note that the lower frequency bands tend to decay faster than higher frequency bands (inset figure 4.30 (b)), indicating that either the PET or silver films become dispersive at high frequency (i.e. higher frequencies are more penetrative in the shielding material). It is possible that this dispersive nature may become more pronounced as the optical or UV range of the EM spectrum is approached and hence Chapter 5 will discuss this issue.
4.5.3 Ka-Band EMI Shielding effectiveness of Inkjet Printed Ag Grids

Evaluation of the EMI shielding by Ag grids in the Ka-band is performed in a solely linear transmission detection method as previously described in section 4.5.2 for the Ag films.
As is seen in figure 4.31 the measured transmitted signal to noise ratio decreases for the higher frequency bands. As the frequency is increased we begin to approach the limit where the pitch becomes comparable to half the wavelength for the sparser meshes.

Figure 4.32: Normalized linear transmission as a function of metallic fill factor in the excitation bands ranging from 7 to 30.5 GHz
In figure 4.32 the normalized linear transmission as a function of the metallic fill factor is shown. If we take for example the 24-25.5 GHz band, half the mid bandwidth wavelength is equal to 6.06 mm, and according to equation 4.5b a maximum transmission should be observed for the 26.5 % fill factor grid, where D is equal to 6 mm. For this band 51.9 % of the incident signal is transmitted through the 26.5 % fill factor grid. Comparing to the 15-21 GHz band, where D < \(\lambda/2\) a transmission of only 29.5 % is observed. For bands exceeding 21 GHz, a transmission less than 100 % is recorded for the blank PET substrate, indicating that the PET becomes dispersive at high frequencies.

4.6 Summary

A comprehensive evaluation of the effectiveness of inkjet printed Ag films and grids as potential EMI protection has been presented. Consistent EMI shielding behaviour has been observed for all Ag films beyond a critical thickness (percolation threshold) which is determined to be approximately 120 nm. From a film only 120 nm thick 98-99 % of the incident signal is rejected, mainly in the form of reflection. For thicknesses beyond 120 nm a constant maximum in the reflection coefficient and minimum in the transmission coefficient is seen. This transition from a microwave transparent to a reflection regime is consistent in the three excitation bands used (X-Ku, K and Ka) indicating broadband/multi-band functionality of the Ag films. For the purpose of this study, K and Ka-band custom waveguides were fabricated in accordance with CST Microwave Studio geometric simulations. Both sets of cavities were observed to be low loss with a large dynamic range within their operational bandwidth. It is noted that for lower frequency bands, the microwave transmission tends to decay faster with increasing film thickness than the higher frequency bands, indicating that lower frequencies have a shorter penetration depth in the Ag/PET system.

The effect of pitch on the shielding effectiveness of 200 nm thick Ag grids was also investigated for applications where mass restrictions are an important factor (for example, in space or satellite applications). It was established that for grids with metallic fill factor greater than 55 % desirable EMI shielding can be achieved but this
is heavily dependent on the intended operational band. For example a 55 % dense grid will shield 98 % of incident energy in the K-band but will only shield 77 % in the 28-30.5 GHz band. Hence, meshes are not ideal for universal broadband application and the pitch must be tailored for shielding within a specific band. For a metallic fill factor greater than 75 % (D = 1 mm) uniformly high shielding characteristics are observed as D is far less than \( \lambda/2 \) within the measured bands.
Chapter 5

Optical Characterisation of Randomly Dispersed Media

5.1 Motivation

Chapter 3 introduced the need to develop novel transparent conductors and gave an insight into novel contactless methods for evaluating their electrical conductivity at microwave frequencies. The conductivity is just one of many important factors in establishing if a material is a suitable replacement for ITO. An equally important figure of merit is the material’s optical properties and it is important to find the right balance of enhanced conductivity while also providing maximum optical transmittance. This is also the case for the Ag films and grid structures discussed in chapter 4 where the main objective was to provide maximum RF shielding, which is directly related to the conductivity while minimizing the film thickness or metallic fill for the case of the grids.

Contactless optic-based methods are a pivotal tool in non-destructively characterising a material’s morphology. To this end ultraviolet-visible densitometry (optical spectroscopy) is used examine the density of the inkjet printed Ag films. As observed in chapter 4, the films become dispersive at high frequencies and higher frequency excitations tend to be more penetrative than lower frequencies and it is therefore important to establish if the trend extends to ultra-short optical wavelengths. If this is the case, it may be seen that a sample which is close to 100 % shielding in the microwave regime may have non-negligible transmission at optical frequencies.

In addition to optical densitometry laser specular scattering (LSS) techniques are exploited to enhance understanding of surface topography in disordered systems such as nanowire networks, which demonstrate significant specular scattering or reflection when laser illuminated. The specular pattern can provide not only information on the network’s order but also the length scale of network features.
5.2 Ultraviolet-Visible Densitometry

A Xe arc-lamp produces illumination which is fed into a fibre-optical cable. The Ocean Optics spectrometer detects optical intensity with wavelengths between 420 nm and 900 nm. Optical transmission is measured in a straight-trough configuration as in figure 2.17 while reflections are measured at a $45^\circ$ angle as seen in figure 2.19 for Ag films of varying thickness. Diffuse reflections are unaccounted for in this geometry so it’s expected that reported reflection coefficients will be underestimated. Absorption is then computed using $A = 1 - (T + R)$.

5.2.1 UV-VIS Transmission

The transmitted optical intensity at normal incidence as a function of the wavelength is shown in figure 5.1 for Ag films of varying thickness. As mentioned in chapter 4 the effective thickness is estimated to be 10 nm per pass of through the inkjet printer. $I_0$ represents the transmitted optical intensity in the absence of a test sample. In this case $I_0$ is attenuated using cellulose filter paper to avoid saturating the spectrometer detector. $I_0$ is measured twice, once at the start of the measurement and then again

Figure 5.1: High visible/UV transmitted intensity spectra for Ag films of varying thickness.
after the measurement series, in order to estimate the photodegradation of the filter paper [74]. Similarly the bare PET substrate is also measured before and after the measurement series. Both $I_0$ and the PET experience a small photodegradation induced reduction in the transmitted optical intensity over the time scale of the measurements, therefore it is important to have a short measurement time to minimize UV effects on the substrate. $I_0$ must also be measured at regular intervals.

The transmittance is defined as the ratio of the transmitted intensity to the incident intensity, such that

$$T = \frac{I}{I_0}$$

(5.1)

Using equation 5.1 the transmittance as a function of wavelength is shown in figure 5.2 for the sample series. Non-zero optical transmittance is observed for all films, even those with thicknesses beyond the microwave shielding threshold of $\approx 90 \text{ nm}$. A large source of dispersion appears to be the PET substrate as transmittance varies significantly over the spectral range (0.358 at 420 nm to 0.785 at 900 nm).

![Transmittance spectra for inkjet printed Ag films of varying thickness.](image)

**Figure 5.2:** Optical transmittance spectra for inkjet printed Ag films of varying thickness.
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5.2

The dependence of the optical transmittance on the film thickness is shown for two wavelengths; 420 nm and 900 nm in figure 5.3. The Beer-Lambert Law relates the optical transmittance through a solution to the materials absorbance $A$ and is typically defined as

$$A = \log \frac{I_0}{T} = \varepsilon cl$$

(5.2)

$c$ and $l$ are the concentration (mol dm$^{-3}$) and optical path length of the solution respectively. This relation is used to estimate the optical absorbance as a function of optical path length as in figure 5.4, where the absorbance can take a value between zero (100 % Transmittance) and 2 (0 % Transmittance). The absorbance of the PET (i.e. the 0 nm thickness) is non-negligible, particularly at shorter wavelengths (higher frequencies) where it becomes quite dispersive. Subtracting the absorbance of the PET substrate, the absorbance of a 9 pass (90 nm thick) Ag film is 0.78 a.u. (or 53 % absorption). If we compare this with microwave absorption in the K-band (see figure 4.20), previously discussed in Chapter 4 the absorption at the percolation threshold (9 passes) is approximately 60 %. The absorbance for the 90 nm thick film at 420 nm is

Figure 5.3: Optical transmittance as a function of film thickness is shown at two wavelengths and follows the Beer-Lambert Law.
only a few percent below that observed for microwave frequencies, indicating that the dispersion is deeper in the UV than the observation range. Indeed this should be the case, as the plasma cut-off wavelength for Ag is approximately \( \lambda \approx 350 \text{ nm} \) [75].

From equation 5.2 \( \varepsilon \) can be defined as the molar absorption coefficient \( \varepsilon = \frac{A}{c} \).

For the case of an optically transparent film the transmittance can be defined as

\[
T = \exp(-\alpha l)
\]

(5.3)

where \( \alpha \) is a linear absorption coefficient, an expression that represents how far the incident light at a specific wavelength can travel into the material before it is absorbed. An exponential decay curve is fitted to the experimental data to determine \( \alpha \). Figure 5.5 shows the absorption coefficients for all wavelengths between 420 nm and 900 nm. Only marginally longer absorption coefficients are observed for shorter wavelengths (i.e. higher frequencies) and the absorption is flat-band and consistent over the observed spectral range. It is therefore deduced that the frequency dispersion extends to frequencies in excess of what we are able to observe using UV-visible spectroscopy.
(\(\lambda < 350\) nm). \(\alpha\) is given as 51.99 nm at a 420 nm wavelength and 44.47 nm at 900 nm. The absorption coefficients for continuous bulk Ag films are reported in literature at 420 nm and 900 nm as 14.235 nm and 11.241 nm respectively, much shorter than those measured for the inkjet printed Ag films [76]. It is therefore assumed that the films are not fully dense continuous films but still rather porous.

\[\begin{array}{|c|c|}
\hline
\lambda (\text{nm}) & \alpha (\text{nm}) \\
\hline
420 & 51.99 \\
480 & 50.00 \\
540 & 48.00 \\
600 & 45.99 \\
660 & 44.99 \\
720 & 43.99 \\
780 & 42.99 \\
840 & 41.99 \\
900 & 40.99 \\
\hline
\end{array}\]

**Figure 5.5:** Absorption coefficient for inkjet printed films for all wavelengths between 420 nm and 900 nm

### 5.2.2 UV-VIS Reflection

The optical absorbance estimates based on optical transmittance data discussed in the previous section fail to account for a reflection contribution to the diminished transmitted intensity. For metallic systems, this contribution may be significant and consist of direct or diffuse reflections or a combination of both. Here direct optical reflections are estimated, by illuminating a sample at a 45° and detecting the reflections at right angle to the source, through a secondary fibre-optical cable. This method fails to account for diffuse reflections or specular reflections, which are typically measured using an integrating sphere which allows for multi-port illumination and spatially diverse optical detection. Figure 5.6 shows the as-measured reflected intensity spectra for a number of samples with varying thickness, which appear to show enhanced reflection.
with increasing thickness. In order to detect reflected signals the maximum gain of the detector, gain 8 was used.

Figure 5.6: Optical reflectance spectra at 45° incidence for inkjet printed Ag films of varying thickness.

Figure 5.7: Reflectance measurements taken at a 45° angle for two wavelengths; 420 nm and 900 nm as a function of film thickness.
The dependence of the $45^\circ$ optical reflection on the film thickness is shown in figure 5.7 for 420 nm and 900 nm at extreme ends of the optical detection bandwidth. Here it is assumed that measured reflections are underestimated, as for the case of the porous film, diffuse and specular reflections would be non-negligible. The $45^\circ$ reflections are consistently less than 1% of the incident intensity. The signal to noise ratio is much lower than that for the transmittance measurements due to the smaller detected signals.

5.2.3 UV-VIS Absorption

The high-vis UV absorption is calculated according to $A = 1 - (T + R)$ and displaced in figure 5.8. It is clear that samples with thicknesses $< 160$ nm fail to reach the saturation regime for metallic absorption, where the energy is fully attenuated.

![Figure 5.8: Estimates of the optical absorbency based on transmittance and reflectance measurements at two wavelengths; 420 nm and 900 nm as a function of film thickness.](image)

5.3 Laser Specular Scattering

Laser specular scattering (LSS) techniques are exploited to non-destructively examine the surface topography of randomly orientated NWs in dense arrays, where the NWs have been spray-cast onto dielectric (borosilicate glass) or semiconducting (Si)
substrates and are largely disordered or only partially ordered, due to the uncontrolled nature of the spray-casting method. This takes two forms; transmission analysis for Ni NWs on transparent substrates and reflection characterisation for Ag NWs on Si (i.e. optically non-transparent) substrates. Both geometries are further discussed in chapter 2 section 2.4.1.

5.3.1 Transmission Mode Evaluation of Ni Nanowire Arrays on Borosilicate Glass Transparent Substrates

For transmission mode analysis, samples consist of oxide passivated Ni nanowires with diameters of $\approx 80$ nm. Figure 5.9a shows a representative, as-captured specular scattering image taken for an Ni NWN ($% T \approx 70 \%$). Points within the captured image, specifically the corners of the screen and the pinhole are mapped to co-ordinates.
in the x and y directions, producing a dewarped image [77]. This process is illustrated in figure 5.9b. The background signal was subtracted and histogram equalisation was implemented for all images. This involves a linear transformation of the image whereby the new output pixel $P_{out}$ is confined between the 0 and 255 (256-bit) gray-scale. If the original pixel $P_{in}$ has a gray-scale x to (100-x), $P_{out}$ will be defined as:

$$P_{out} = \frac{255(P_{in} - c)}{d - c}$$  \hspace{1cm} (5.4)

where $c$ and $d$ are the $x^{th}$ and $(100 - x)^{th}$ percentiles [78]. The specular nature of the fast-Fourier-transform filtered image shown in figure 5.9c indicates scattering from a random array of nanowires with no preferential arrangement. The pronounced central ring indicates structural isotropy with no long range order within the network.  

The scattering wave vector may be calculated using the relation

$$q = \left(\frac{4\pi}{\lambda}\right) \sin\left(\frac{\theta}{2}\right)$$  \hspace{1cm} (5.5)

where $\theta$ is the scattering angle and may be calculated according to $\tan^{-1}(\frac{u}{z})$, $u$ being the radial distance from the central point on the screen and $z$ begin the distance between the illumination source to the screen ($z = 1$ m). The extracted scattering intensity is given as a function of the scattering wave vector for transmission mode analysis of Ni NWNs in figure 5.9. The peak shown for small $q$ values indicates that the $I_0$ beam was only partially excluded. After the initial peak both the transmission and reflection mode scattering intensity plateau with increasing $q$, further indicating no higher probability scattering angles and hence no preferential ordering.
Assuming classical Rayleigh scattering the size of the scattering particle $D$ and the scattering wave vector $q$ are related by

$$D = \frac{2\pi}{q}. \quad (5.6)$$

Plotting the normalised scattering intensity as a function of the size $D$, the size (NW length) distribution curve 5.11 is obtained for an Ni NWN with 70 % optical transparency. A broad peak in the distribution indicates a broad dispersion in NW lengths over the network, with the mean NW length being approximately 50 – 70 $\mu$m.
5.3.2 Reflection Mode Evaluation of Ag Nanowire Arrays on Si Substrates

As previously stated, reflection mode LSS is a viable method for contactlessly assessing structural features of NWNs on non-transparent substrates. For reflection mode analysis, samples consist of PVP coated Ag nanowires with diameters of \( \approx 86 \) nm. Figure 5.12a shows a raw specular image for an Ag NWN with 65.9 % optical transmittance. For reflection mode analysis, the \( I_0 \) beam features strongly in the specular image due to reflection from the Si substrate and is more difficult to exclude than for transmission mode measurements. The gray-scale converted and dewarped image is displayed in figure 5.12b, which shows the highly specular nature of the sample prior to background subtraction. Figure 5.12c shows the image obtained from conversion from the spatial domain via fast Fourier transform. The diffuse ring around the central position is again indicative of an isotropic material with no long-range structural order.
The scattering intensity as a function wave vector is shown in figure 5.13. For wave-vectors less than 0.2 \( \mu m^{-1} \) \( I_0 \) is not effectively attenuated, hence the sharp peak at low \( q \) values is observed. After the initial peak the scattering intensity plateaus with increasing \( q \), indicating no preferential ordering or higher probability scattering angles.

Again, equation 5.6 is used to convert from wave vector \( q \) to the size of scatterer, in this case length of the NWs, \( D \). A broad size distribution peak is observed, with the NWs length most likely in the range 20-40 \( \mu m \). This large distribution is due to the random nature of the network, where NWs are overlapping. The highest probability NW length is \( \approx 30 \, \mu m \) long.
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5.4

Figure 5.13: Reflection mode obtained scattering intensity as a function of the scattering wave vector for Ag NWNs.

Figure 5.14: Normalised scattering intensity as a function of size distribution $D(\mu m)$ for reflection mode detection (Ag NWNs)

A representative set for both transmission and reflection detection methods have been shown here and supplemental data of sparser networks is given in appendix C to
show the reproducibility of length scales measured.

5.4 Summary

This chapter examined non-destructive optical methods for evaluating structural features of randomly ordered metallic systems. This included high-visible to UV wavelength densitometry and small angle laser specular scattering for quantifying the length-scales of scattering features within a sample.

The ultraviolet-visible (420-900 nm) transmission, 45° reflection and absorption properties of Ag films, produced by inkjet printing, were measured. It was previously observed in chapter 4 that the films become dispersive at high frequencies and it was important to verify that this extended to higher frequencies/shorter wavelengths, beyond the RF/microwave frequency ranges discussed previously. The transmitted optical intensities for films of varying thickness were closely examined at two incident wavelengths, 420 nm and 900 nm. Absorption coefficients for 420 nm and 900 nm were measured as 51.99 nm and 44.47 nm respectively. While the absorption coefficients measured are slightly longer for shorter wavelengths, the absorption is consistent and flat band within the observable spectral range (microwave (Chapter 4) to UV). It is understood that dispersion will manifest at shorter wavelengths (< 350 nm), as it approaches the Debye length for Ag [75]. The measured absorption coefficients were much longer than the literature values for continuous Ag films indicating that the inkjet printing does not create dense films. UV-visible reflection spectroscopy was similarly performed but this was mainly limited by the fact that only 45° direct reflections were measured. This excluded diffuse reflections which may be non-negligible for disordered metallic sample.

In order to show that the structures of metallic nanowire networks previously examined in chapter 3 were truly disordered, the scattering of laser light from the NWN systems was examined. From the specular images obtained it was concluded that neither the Ni NWs on borosilicate glass substrates or Ag networks on Si substrates showed preferential ordering within the network. This was indicated by the specular nature of the original images and the central ring in the FFT filtered image. The central image
is indicative of surface structural isotropy with no long-range order over the area of the sample. The mean NW lengths were determined to be approximately 50-70 µm for Ni NWs on the transparent substrates and 30 µm for Ag on Si substrates. The mean NW length is also examined by scanning electron microscopy performed by Prof. Boland’s research group [79, 80]. They estimate the mean NW length of Ni and Ag NWs to be 10.6 µm and 6.6 µm respectively. LSS samples over a larger area due the longer coherence length of the HeNe laser and can be considered more representative of the entire network.
Chapter 6

SQUID-Based Ferro/Ferrimagnetic Resonance

6.1 Motivation

Resonance detection methods have been used extensively to examine the magnetisation dynamics in ferromagnetic and ferrimagnetic systems since the effect was reported in the late 1940’s [8, 81]. The main advantage of the FMR techniques is the low excitation energy (microwave) by which the magnetic ground state properties are probed. The magnetic relaxation rate can be probed in a time window of typically a few hundred picoseconds and the FMR signal is measured by monitoring the microwave losses as a function of the applied DC field. The most commonly used methods for characterising ferromagnetic resonance (FMR) rely on either resonant cavity or microstrip inductive detection of the reflected signals that originate from subjecting the material to an external microwave field and inferring the resonant absorption by comparing the incident and reflected signals. However, with the rapid growth in RF technologies in terms of device miniaturization and the push to higher frequencies and greater bandwidths inductive methods are falling short in terms of sensitivity and as resonance is inferred measurements are not absolute. As such, non-inductive techniques have emerged in the field more recently, most notably FMR force microscopy (FMRFM) [82, 83], X-ray magnetic circular dichroism (XMCD) detection [84], and optically-based methods, such as magneto-optic Kerr effect (MOKE) [85, 86]. While such methods have dramatically improved the sensitivity limit of FMR detection, inductive methods still have the upper hand in terms of simplicity of implementation and generally inductive methods are used in conjunction with non-inductive techniques for thorough magnetisation dynamics analysis.

In this chapter, a method of detecting FMR by directly measuring the change in the
longitudinal component of the magnetisation $\Delta m_z$ when a material is in resonance, using SQUID magnetometry is demonstrated. Such measurements have only been made previously using MOKE-based techniques [87]. Absolute measurement of the longitudinal component of the magnetisation and resonance induced lowering of this moment makes estimation of the precession cone angle feasible, which is typically difficult to extract using conventional cavity or stripline based detection methods. Such measurements have only been made previously using MOKE-based techniques[87] for FMR detection but such measurements are not absolute and extraction of the precession cone angle is inaccessible. While longitudinal SQUID-based detection has been applied before to the NMR case [88], there are specific differences associated primarily with the much higher resonance frequencies, much shorter decay times and much stronger coupling to the measurement system in the case of FMR. The SQUID-FMR module described in this chapter is also capable of operating as a conventional inductive VNA-FMR set-up and the detection methods are complementary.

6.2 SQUID-FMR Approach and Instrumentation Design

![Figure 6.1: Change in projection of the magnetic moment along z ($\Delta m_z$) during resonance.](image)

Here the ferro/ferrimagnetic material is considered as a macro-spin system, as in figure 6.1, where the length of the magnetisation vector, which is nominally saturated along the $z$ direction $m_z$, remains constant. As the system is excited at its resonance frequency it will absorb microwave excitation power and the precession cone will open.
Opening of the precession cone will lead to lowering of the $m_z$ projection of the magnetic moment, which is approximately quadratic as a function of the cone opening as inferred from the cosine expansion at small angles. Using a widely available commercial SQUID magnetometer (Quantum Design magnetic property measurement system MPMS® XL [89]) we can simultaneously obtain absolute measures of $m_z$ and $\Delta m_z$ and can subsequently gain a quantitative insight into the precession cone angle for a material. The MPMS system uses an RF SQUID driven at a 150 MHz bias frequency and modulated at 100 kHz in a flux-locked loop, such that the GHz excitations used in this study will not interfere.

Supplementary to this, we demonstrate magnetic resonance detection by monitoring $\Delta m_z$ as a function of changing temperature caused by resonant heating, using SQUID magnetometry. Here, the strong interaction with an unmodulated high-frequency signal which leads to precession cone opening has the added effect of periodic heating, which is especially pronounced at resonance. This oscillatory heating induces changes in $m_z$, which are directly detected via the SQUID output signal. This thermal detection, while not inherently absolute exhibits excellent sensitivity and resolution, even at low temperatures.
6.2.1 Resonator Design

Figure 6.2: Schematic of the SQUID-based FMR detector, showing the FR4-based microstrip contained within copper shielding, at the centre of the SQUID magnetometers sample space.

Figure 6.2 shows the design of the resonator consisting of a stainless steel coaxial cable for connecting to the swept frequency source and the FR4 (woven fibreglass and epoxy resin)-based microstrip transmission line shorted with a 50 Ω load resistance (0.5 mm in size), encased in a thin walled copper shielding. The adjusted 50 Ω termination minimizes the amplitude of the back-reflected wave at essentially all frequencies used, leaving only a small amplitude standing wave, the nodes of which vary in spatial position as a function of frequency. The sample can be positioned in the central (40 millimetre-long) region of the microstrip. Since the sample position is typically fixed during measurements, the small amplitude standing waves would project maximal $B$-field intensity only at a certain comb of frequencies. The z-axis profile of the $B$-field distribution of the input signal above the microstrip is essentially cosine in nature, with its zeroth node located at the 50 Ω terminator. The majority of microwave loss is incurred within the cryogenic coaxial line, with about 3 dB of loss at 1 GHz and 30 dB of loss at 18 GHz. A further 0.5 dB of loss is incurred in the microstrip at
1 GHz, further increasing to 5 dB at 18 GHz. The overall loss, at 18 GHz, to the termination point is 35 dB. This can be substantially improved upon by substituting the cryogenic coaxial line, with a low-loss alternative, however this can only be done on expense of a substantial increment in the z-axis heat loss, particularly important for low-temperature measurements.

Two venting holes are positioned at the end of the shield, perpendicular to one another (the vertical one being blind). These serve the sole purpose of preventing implosion of the shielding during pump-down and have no particular microwave functionality as are located a couple of millimetres below the 50 Ω termination of the microstrip. The 90° angle creates a maximum in reflection, ensuring that stray microwave field from the microstrip will not interfere with the flux-locked SQUID loop. The rectangular prism of bulk, polycrystalline YIG (dimensions: 2.5 x 1.6 x 0.7 mm) or polished single crystal YIG sphere (Diameter = 1 mm) is mounted (using varnish) adjacent to the signal plane of the transmission line, where the external RF magnetic field is at a maximum. The sample assembly is placed in the centre of the sensing coils (four-turn z-axis gradiometer, see Fig. 6.2), which are inductively coupled to the RF SQUID within the cryogenic, vacuum environment of the superconducting magnet system.

The total z-axis magnetic moment projection is measured, as usual, by translating the sample vertically around the central position and processing the dependence of the coupled flux on distance. This component can be large and is processed using the lowest gain ranges of the SQUID amplifier. The moment typically is on the order of several mAm². With the superconducting magnet in persistence mode and after a delay determined by a number of different physical factors (eddy current dissipation, on the scale of tens to hundreds of milliseconds, superconducting flux creep on the scale of seconds to minutes, to mention but a few) the superconducting transformer to the SQUID washer is quenched. The very large coupled flux is thereafter no longer measured. For the normal $M_z$ measurements, the SQUID is typically operated within much less than 1 $\Phi_0$ of coupled flux (the response being flux-periodic, modulo $\Phi_0$). It is essentially impossible to measure changes in moment corresponding to $10^{-6}$ of $\Phi_0$, while using the
flux-locked loop. Once the coupled flux is known within $1 \Phi_0$, a momentary quenching of the coupling transformer ensures that no super-current is flowing through the pick-up assembly and that only relative changes in flux (as a function of temperature, microwave absorption, etc.) are measured. Since the change in flux to change of sample moment ratio is well known (calibrated) for all gain ranges, the change in moment, relative to the total $M_z$ can be directly inferred. At this stage (while staying within $1 \Phi_0$), coupled fluxes of the order of $10^{-4}\Phi_0$ and below can be readily detected. It is at this point when the SQUID-FMR measurements begin. The above settling algorithm slows down the measurements of field dependence substantially, allowing for about one spectrum to be acquired every three to five minutes. Within this obligatory delay other measurements can be readily performed, including DC magnetometry, low-frequency AC susceptibility and recoil relaxometry. Measurements requiring the evolution of temperature at fixed field are much less restrictive and can often be performed ‘on the fly.’ A typical noise floor for such a detection system is below $3nAm^2$ with a time constant of the order of $1 s$. In measurement strategies where amplitude chopping or frequency modulation is utilised, the modulation frequency can be varied in the $1 \text{ mHz}$ to $20 \text{ kHz}$ range. This allows for at least two different measurement strategies, one athermal and one thermal as detailed later.

6.2.2 Magnetic and Electrical Evaluation of Transmission Lines

Prior to assembly of the resonance module the magnetic and electrical properties of the PCB material and transmission lines used for RF injection were evaluated.

6.2.2.1 Characterisation of Magnetic Impurities in Microstrip and Co-planar Waveguide Transmission Lines

In order to characterise magnetic properties of the printed circuit board FR4 (woven fibreglass and epoxy resin blend) based transmission lines, scans of the magnetic moment $m$ versus the applied magnetic field $\mu_0H$ were performed at $300 \text{ K}$ for a $5 \times 5 \text{ mm}^2$ piece of copper clad board. Figure 6.3a shows a representative plot of the signal following the subtraction of the large diamagnetic signal. The shape indicates trace amounts
of paramagnetic impurities, sufficiently small to allow detection of $m_z$, the change in the $z$ component of the moment arising when a sample placed on the transmission line.

As seen in figure 6.3b the moment increases significantly with decreasing temperature. This is due to the paramagnetic impurities, the magnetic susceptibility ($\chi = M/H$) of which diverges hyperbolically with decreasing temperature according to the Curie-Weiss Law for paramagnetism. The inset in figure 6.3b shows the susceptibility as a function of $1/T$. Using the equation $\chi = (n\mu_0\mu_B^2N_Ap_{eff}^2)/3k_BT$ [90], where $N_A$ is Avogadro’s number, $p_{eff}^2 = g^2(S(S+1)) = 35$ for $S = 5/2$ and $k_B$ is the Boltzmann constant, the number of Fe$^{3+}$/Mn$^{2+}$ impurities $n$ is determined to be $\approx 1.8$ $\mu$mol. As such, in order to examine low-temperature magnetisation dynamics, alternative dielectric materials must be considered for the transmission lines. Some candidates to consider would be low dielectric-constant materials, such as fused silica, quartz or ceramics which suit as magnetically clean alternates to the FR4. The later are readily available in the form of copper-clad boards, compatible with conventional PCB fabrication techniques. Ceramic boards are however less robust than PCB and cutting and shaping boards is more challenging due to the brittle nature of the material.
The module is fitted with a sliding clamp which is compatible with the transport option of the SQUID MPMS to allow translational motion through the detection coil assembly. Scans of the SQUID output voltage were performed over the entire length of the transmission line in order to check their magnetic homogeneity.

Figure 6.4: SQUID response voltage as a function of position along the line. The microstrip displays good homogeneity with vertical jumps corresponding to different integer numbers of flux quanta $\Phi_0$.

Figure 6.4 shows the 5T scan for a 3 mm signal line width microstrip. The initial peak and the fact that the moment increases as the end of the line is approached is an artefact of the system software modelling the line as two effective monopole moments separated by lines of continuous flux. With regular symmetrical samples it is assumed that the moment approximates an effective magnetic dipole moment. The line shows good homogeneity where vertical jumps correspond to integral jumps in the number of flux quanta.
6.2.2.2 Impedance Characterisation of Transmission Lines

The characteristic impedance and reflections of the transmission line are evaluated using pulsed time domain reflectometry. As seen in figure 6.5 reflections are minimal over the entirety of the line and the characteristic impedance is approximately 53Ω. Primary reflections within the set-up are seen at time scales corresponding to the front end SMA to microstrip soldering connection. The microstrip is terminated with a 50Ω load impedance.

![Figure 6.5: TDR measured microwave reflected voltage (V) as a function of pulse time for a 3 mm signal line width microstrip.](image)

6.3 Test Materials

6.3.1 Bulk Polycrystalline and Single-Crystalline Yttrium Iron Garnet $Y_3Fe_5O_{12}$

This chapter discusses the application of novel SQUID-based detection methods in analysing the magnetisation dynamics in the ferrimagnetic insulator, yttrium-iron garnet ($Y_3Fe_5O_{12}$ YIG), as an illustration of the capability of the newly developed SFMR in analysing precessional dynamics. YIG, due to it’s unique magnetic and thermal...
properties: very narrow resonance-line width, low spin-damping at microwave frequencies and a relatively high \( T_c \approx 560 \text{K} \), has been the basis of most insulator based radio frequency and microwave devices, and the standard test material for novel resonance phenomena in spintronics (i.e. spin pumping)[91–95]. It’s typically a fundamental component of non-reciprocal devices, such as circulators and isolators, etc. In order to gain an understanding of the magnetisation dynamics in YIG one must first look at its composition. YIG has a complex crystal structure with 160 atoms per unit cell, consisting of eight formula units of \( \text{Y}_3\text{Fe}_2^{3+}\text{Fe}_3^{3+}\text{O}_{12}^{2}\) per unit cell. The Fe atoms occupy two crystallographic positions, 16 a-type octahedrally coordinated by oxygen and 24 d-type tetrahedrally coordinated by oxygen as shown in figure 6.28 [96], with an unbalanced, trivalent Fe atom giving rise to a net spontaneous magnetisation [97]. In the microwave regime YIG is typically described using a two sublattice approximation and the resonant energy absorption by the macroscopic magnetisation is typically interpreted using Kittel’s theory for ferromagnetic resonance [11]. Despite its profuse industrial applications, much is still to be understood about the details of magnetisation dynamics at low temperatures as YIG’s magnonic spectra are shown to have 20 distinct magnon branches, which are not all necessarily in phase with each other [98–101]. This study will characterise the gyroscopic properties of both bulk poly- and single-crystalline yttrium iron garnet.
As mentioned in the experimental section, the polycrystalline sample was provided by colleagues in Bell labs, in the form of a die pressed cylinder and samples were carefully cut from the cylinder using a diamond saw. Volumetric densitometry measurements report a density of 4180 kg/m$^3$, an almost 7 % reduction in the literature theoretical bulk density of 5170 kg/m$^3$. Densification is largely dependent on the pressing and sintering procedure used and forming a cylindrical piece with close to theoretical density is difficult [102]. Figure 6.7 illustrates the EDX spectra obtained for a piece of the YIG ring. The carbon peak is a result of the graphite used to avoid surface charging and is typically removed from the atomic percentage calculations. Yttrium, iron and oxygen peaks indicate 14.9 %, 23.1 % and 60.6 % atomic percentages respectively. 1.4 % indium is also identified which indicates that the YIG is in fact indium substituted $Y_3In_xFe_{5-x}O_{12}$ where $x = 0.3$. Previous studies of the microwave characteristics of In substituted YIG found that for $x = 3$ a small decrease in the $\Delta H$ linewidth is observed in comparison to the non-substituted YIG. For $x > 3$ the linewidth broadens [103].
The powder X-ray diffraction pattern for the $Y_3In_{0.3}Fe_{4.7}O_{12}$ is shown in figure 6.8. The most intense diffraction peaks are indexed to (420), (422), (640) and (642) of a YIG cubic cell. The presence of several other peaks could suggests that many randomly orientated crystallographic phases are indeed present. Indium is not directly detected, this is due to the concentration being below the resolution limit of the XRD system.
Temperature dependent magnetisation curves for the bulk YIG are shown in figure 6.9. Consistently low remanence and coercive fields are observed over the range of experimental temperatures and the large room temperature saturation magnetisation increases systematically with decreasing temperature as shown in the inset in figure 6.9.

6.4 Single Port Resonator

This section describes the operation of the single port SQUID-FMR module, where the RF injection microstrip is 50 $\Omega$ terminated.

6.4.1 Bulk Polycrystalline $Y_3Fe_5O_{12}$

6.4.1.1 Conventional Single Port VNA-FMR Inside the MPMS

The experimental set-up may be operated as a conventional broadband (100 MHz - 20 GHz) single-port vector network analyser (VNA)-FMR spectrum analyser, where FMR spectra can be obtained over a broad range of static fields and experimental temperatures. The unprocessed output signal appears as a complex series of peaks in
the cumulative reflected amplitude, as shown in figure 6.10 where the signal to noise ratio largely depends on the filtering and sampler used.

**Figure 6.10:** Unprocessed VNA-FMR spectra in a 50 mT static field. Insets display (a) the $S_{11}$ scattering parameters and (b) the phase as a function of frequency, simulated using transmission line theory.

In order to process the FMR signal one must account for reflections due to discontinuities within the experimental apparatus. Modelling of the reflected signals amplitude and phase are possible using transmission line theory, as seen in figure 6.10 (a) and (b), which accounts only for reflections due to impedance mismatches within the flexible coaxial cable, semi-rigid coaxial and PCB board. Modelling of all reflections within the experimental set-up using transmission line theory is not a trivial task due to the dispersive nature of the materials, FR4 and polytetrafluoroethylene (PTFE) at high frequencies. If the set-up were to be sufficiently compartmentalized, the reflections originating from each module could be calibrated (and de-embedded) and theoretically one could provide a more accurate model for all reflections but this is beyond the scope of this work. Additionally, compartmentalizing the system could intrude additional reflections as each non-continuous join will behave as a scattering point.
Figure 6.11: Evolution of the resonant signal at low fields (10-30 mT) for the polycrystalline cuboid. Inset shows the normalised amplitude for the same applied field values, in order to render the very broad, low-field resonance visible. The resonant signal for the 10 mT applied field (black trace) is only rendered by normalisation. The sharp peaks at 1.8 and 2.4 GHz are due to GSM and WiFi interference, respectively.

At low frequencies, where good signal to noise ratios can be readily achieved, resonances can be simply tracked by subtracting a spectrum measured either at remanence, in a multi-domain state, or in complete saturation at fields large enough to place the resonance frequency of the lowest magnetostatic mode at frequencies way beyond the Nyquist threshold and the front end filter cut-off of the VNA. This process is illustrated in figure 6.11. The exacerbated broadening at very low fields is attributed to the large distribution of internal demagnetising fields in the specimen below magnetisation saturation. The lineshape at fields below 25 mT is not purely Lorentzian but rather a Gaussian convoluted Lorentzian, well fitted by a pseudo-voight function.
**Figure 6.12:** VNA-FMR spectra for a rectangular prism of bulk YIG. Frequency dependence of the background subtracted reflected amplitude under a static field $\mu_0 H$.

**Figure 6.13:** VNA-FMR spectra obtained for 100 MHz-20 GHz excitation frequency and 0-400 mT external applied field. Two resonant modes are identified which are strongly coupled to the transmission line electromagnetic modes. Demagnetising fields are determined; $H_{d1} = 100$ mT, $H_{d2} = 110$ mT, $\gamma = 27.8(3)$ GHz/T

The broadband (100 MHz - 10 GHz) FMR signals shown in figure 6.12 are extracted from the unprocessed data by subtracting an off-resonance background spectra and the field-independent peaks in the reflected amplitude. The resonant frequency ($f_{res}$) is
indicated by a sharp increase in the reflected amplitude which shifts to higher frequencies with increasing applied field, in agreement with Kittel’s equation for ferromagnetic resonance[11]. VNA-FMR spectra in figure 6.13 suggest the existence of two resonant modes which are strongly coupled to geometric reflections within the experimental apparatus. The higher and lower frequency modes are shown to have demagnetising fields of 110 mT and 100 mT respectively and correspond to the two different demagnetising directions $x$ and $y$. The gyromagnetic ratio $\gamma$ is measured as 27.8(3) GHz/T.

6.4.1.2 Athermal Detection

The athermal SQUID-FMR circuitry is shown in figure 6.14. A Rohde and Schwarz 100 MHz to 20 GHz VNA provides the RF excitation to the FMR module which is maintained within the four-turn $z$-axis gradiometer detection coils (see Fig. 6.2), inductively coupled to the SQUID washer. The TTi arbitrary waveform generator provides $2 V_{pp}$ SQUID modulation. The SQUID output voltage is detected by lock-in amplification and visualised on an oscilloscope to ensure that the SQUID remains flux-lock. If the flux drifts out of range a momentary reset is used to temporarily reset the flux-locked loop (FLL), such that the output is zero to within 1 flux quantum. The lock-in amplifier provides triggering to the oscilloscope. To reduce the $1/f$ noise and narrowband the measured output signal, the GHz excitation signal provided by the 100 MHz - 20 GHz signal generator is chopped, the frequency of which dictates the operating regime of the measurement. At a chopping frequency of 1 kHz, resonant absorption is in the athermal regime, where the change in the lock-in demodulated and processed SQUID output voltage is directly proportional to the change in the $z$ projection of the macro-spin.
Figure 6.14: Circuit design of athermal SQUID FMR detection.

Figure 6.15: The dependence of the SQUID output on the input power is quadratic (on linear scale) as expected from the cosine expansion at small angles. The secondary y-axis shows the corresponding change of macrospin moment projection.

Figure 6.15 shows the dependence of the SQUID output voltage on the input power when excited at 5 GHz in a 120 mT in-plane field, which on a linear scale is shown to be quadratic, as expected from the cosine expansion at small angles. Therefore a small increase in the input power can produce a substantial change in terms of sensitivity. As previously stated, the change in the SQUID-voltage to change in flux and hence change in moment along the applied field direction is well calibrated for each gain-range. The secondary y-axis in figure 6.15 illustrates the change in the moment corresponding to the measured SQUID output voltage.
In a static longitudinal magnetic field, the frequency is swept and resonance is indicated by a sharp increase in the SQUID output voltage. Figure 6.16 compares a sample SQUID resonance spectra in a nominal 50 mT applied magnetic field with that obtained using VNA-FMR for the same conditions. The spectra indicate a primary peak in the resonance absorption at 3.02 GHz followed by a secondary weaker peak at 3.25 GHz. In addition the VNA-FMR-detected resonant peaks appear broader than those obtained by SFMR. This may be attributed to the intermediate frequency filtering within the VNA itself. The linewidth, in the case of SQUID-detection is limited by the chopping or modulation frequency, or if that is sufficiently low, by the fundamental broadening for the sample.

Figure 6.16: Dependence of both the VNA-FMR measured reflected amplitude and on a secondary y-axis, the SQUID output voltage on the applied excitation frequency in a nominal $\mu_0 H = 50$ mT field.
Figure 6.17 compares the SQUID detected resonance frequency of the primary resonant mode with the VNA obtained $f_{\text{res}}$ as a function of the applied field. Excellent agreement is shown for the two FMR detection methods, which also show agreement with the high-bias field range of Kittel dynamics[11]. The gyromagnetic ratio $\gamma$ and demagnetising field are shown to be $27.4(2)$ GHz/T and 61.9 mT respectively.

6.4.1.3 Thermal Regime Detection

At low frequency chopping ($< 50$ Hz) FM resonant absorption generates oscillatory thermal waves, which induce lowering of the macro-spin’s projection along the $z$-axis. Here, we take a macroscopic view of heat-wave propagation across the structure, with the typical frequencies of excited waves ranging from mHz to several tens of kHz. The corresponding wavelengths would be on the order of $\mu m$ to several centimetres, depending on the thermal conductance and heat capacitance of the the various material involved (sample, support structure - microstrip, varnish etc.) These are not the short-wavelength phonons of YIG, and are not explicitly quantized, but rather posses a very smooth continuous spectrum and wavelength dispersion. The sample would essentially
never be uniformly heated by the precession losses. The dissipation (heat source distribution) would vary spatially, depending on the macroscopic magnetostatic modes excited (i.e. would be substantially smaller at the modes’ nodes), while the heat sinking is typically also rather non-uniform, with the majority of it provided by Newtonian heat conduction to the microstrip structure and ultimately to the thermal bath. A substantially lower thermal conductivity is effected by gaseous convection (for the relatively low pressures or residual helium in the sample space - pressures are typically $10^{-2}$ mBar or lower), and by IR emission, which only becomes important at high temperatures (as it varies as $\approx T^4$). The dynamic thermal gradients across the sample can be substantial and at high microwave powers can reach several K/mm. In this regime, direct detection of $\frac{dm_z}{dT}$ is demonstrated.

Figure 6.18 shows a direct comparison between resonance spectra obtained in the athermal and thermal regime at different experimental temperatures (4 K - 400 K). In the thermal regime the SQUID output voltage is shown to be substantially stronger, especially at lower temperatures.
Figure 6.18: Resonance spectra obtained at different temperatures (90 - 400 K) by (a) monitoring the the SQUID output voltage which is proportional to the change in the projection of the in-plane magnetisation. (b) In this regime the SQUID output voltage is proportional to the change in $m_z$ resulting from a temperature change $dT$ caused by resonant heating.

Figure 6.19 (a) shows the thermal conductivity $\sigma$ of bulk YIG with varying temperature, which displays a clear maximum at 20 K [104, 105]. In this regime the heat capacity $c$ shown in figure 6.19 (b) has a prominent role in the resonant excitation and governs the SQUID output voltage.
Values of both $\sigma$ and $c$ are obtained from a number of sources [104–106], and are interpolated ($\beta$-splined) together to form a complete picture of the thermal properties as a function of changing temperature.

The thermal time constant, defined as $\tau = c\rho_{YIG}/\sigma A$, where $\rho_{YIG} = 5.11 \times 10^3$ kg/m$^3$ and $A$ is the surface area of the rectangular prism of YIG, is dominated by the heat
capacity and constrains to zero at low temperatures. If the thermal coupling of the sample to the SQUID heat bath is considered as an equivalent RLC circuit, as in figure 6.21 (b) the thermal impedance of the system can be effectively simulated. Accounting for this impedance and the fit to the Magnetisation curve seen inset in figure 6.22, the material dependent $\frac{dm_z}{dT}$ can be modelled, as seen in figure 6.21 (a).

![Graph](image)

**Figure 6.21:** (a) Simulated $\frac{dm_z}{dT}$. (b) Inset shows a rough schematic of the equivalent circuit model, illustrating the thermal coupling of the sample to the SQUID heat bath.

Figure 6.22 shows the variation of resonance frequency with temperature in a static 50 mT magnetic field. Both regimes show the resonance frequency plateauing below a critical temperature (T=160 K). The phase shift at low temperatures, shown in figure 6.23 suggests a transition from thermal to athermal regime. The behaviour of the thermal system is essentially one of a conventional low-pass filter, the theoretical limit for the phase shift is $-90 \text{deg}$. In the limit of long thermal time constants, on the order of seconds to several seconds, which is the case for weakly coupled samples of millimetric dimensions, close to room temperature, the thermal time constant can be measured by the conventional pulsed method, i.e. by abruptly switching on and off the microwave excitation and tracking the heat up and cool down exponents (see figure 6.35). The same approach becomes impractical for thermal time constants shorter than 100 ms, at which point an AC de-phasing approach becomes more practical. It is, in principle, possible to mount millimetre and sub-millimetre samples on a thin wire (25 $\mu$m diameter, gold-plated tungsten) symmetric line (twinax$^\text{®}$) assembly, in order to perform absolute calorimetry. Examples of wire-suspension type calorimeters are
abundant, including some widely used commercial systems i.e. the Quantum Design PPMS Calorimeter Option [107]. The analysis in this case is done in three consecutive steps. First, the temperature/time traces of the bare assembly are recorded - these are responses of the type \( \Delta T \exp(-t-t_0/\tau) \) and \( 1-\Delta T \exp(-t-t_0/\tau) \), on each positive/negative front of the heat pulses. Second, the smallest practical amount of an adhesive (often a hard vacuum grease, i.e. H-type) is then added to the nominal mounting position of the sample, and the thermal behaviour re-measured. Third, the sample is added, without removing any of the adhesive, thus preserving the quality of mass balance. Again, the thermal exponents are measured, as required, as a function of field, temperature and any other intensive thermodynamic parameters. Last the combined datasets are used to model the thermal conductivity and capacity of the calorimeter, the losses to thermal bath and ultimately extract the heat capacity and conductivity of the sample. The strategy, in the case of short time constants, is to work in Fourier space, following exactly the same mathematical approach. Heat capacitance is treated as a generalized susceptibility and the real and imaginary parts of the same are measured as a function of thermal excitation frequency and all required intensive parameters. Such additional functionality goes beyond the scope of this work.

![Figure 6.22](image-url)  
**Figure 6.22:** Temperature dependence of the resonant frequency detected by athermal \((\Delta m_z)\) and thermal \((\Delta m_z/dT)\) regime measurements of the SQUID output voltage, for bulk YIG in a 50 mT static field.
6.4.2 Continuous-Wave Absorption and Detection

With minor modification to the set-up circuitry, continuous wave excitation and detection is possible. The experimental set-up is illustrated in figure 6.24. Again the FMR module is within the detection coils, inductively coupled to the SQUID. Here an external waveform generator provides a triggering pulse to both the oscilloscope and the RF generator to initiate the frequency sweep. The oscilloscope directly displays both the digitized SQUID voltage and the sweep information, once the triggering conditions are met.

The level of thermal coupling between the sample and the cryostat’s isothermal shield ultimately determines the rate at which the excitation frequency can be swept. At very low sweep rates, comparable with the characteristic time-constant of the proportional integral derivative (PID) temperature controller, the back action of the same leads to line-shape distortion and significant attenuation of the amplitude of the detected signal. Moreover, the conventional strategy of using different optimised PID values at different temperatures makes it much more tractable to analyse the behaviour of small samples of low heat capacity, or when \( \tau_{\text{sample}} \ll \tau_{\text{cryostat}} \). As this version
of the technique does not require amplitude (chopping) or frequency modulation, the ultimate frequency resolution can be much higher and even allow for the measurement of paramagnetic resonance.

As displayed in figure 6.25, continuous wave (CW) excitation with thermal detection is achievable and absorption spectra are obtained by the direct digitization of the output voltage as the sample is subjected to a continuous swept frequency excitation.
Figure 6.26 shows a colour map of the resonant modes at varying temperatures. In agreement with figure 6.13, two primary absorption modes, converging at very low and high temperatures are displayed. Once again we see strong coupling of the FM resonant modes and electrostatic modes of the microstrip transmission line at specific frequencies.

![Color Map of Resonant Modes](image)

**Figure 6.26:** Continuous wave microwave absorption indicates two resonant modes which converge at high and low temperatures. This finding is congruent with the VNA-FMR map displayed in figure 6.13.

The FR4 dielectric actually provides some advantages in thermal detection regime. At low temperatures, the additional change in moment and susceptibility of the paramagnetic impurities in the board of the microstrip (which, itself can be thermally well anchored to the sample) actually improve sensitivity. This is due to the hyperbolic divergence of the susceptibility of these impurities, at low temperature (see Figure 6.3b). A small change in temperature of the region of the microstrip (in vicinity of the sample) due to the sample’s resonant absorption, produces a large change in the overall magnetic moment and the corresponding flux coupled to the SQUID gradiometer. This strongly suggests that an alternative approach to sensitivity improvement would be to immerse the sample in a ‘cocoon’ of an ‘ideal’ paramagnetic salt such as Mohr’s salt or ammonium iron (III) sulphate dodecahydrate (ferric ammonium alum - FAA). Of
course, such an approach is only really beneficial at temperatures below few K, for an applied field of $\mu_0 H \sim T$.

Figure 6.27 shows SQUID output voltage as a function of the chopping frequency applied to the GHz excitation. At low frequency ($< 50$ Hz) resonant absorption generates oscillatory thermal waves which alter the projection of $m_z$. Between 50 Hz and 2 kHz measurement is stable and allows for direct detection of $\Delta m_z$. Above 100 kHz the slew-rate of the SQUID-system is exceeded and the SQUID no longer remains flux-locked. This dependency is largely dependent on the test materials composition and shape and hence there will be a different optimum chopping frequency for absolute and thermal regime detection for each sample.

![Figure 6.27: SQUID output voltage as a function of the chopping frequency of the GHz excitation. Three regimes are displayed; Thermal (1 - 20 Hz) where resonant heating changes the projection of $m_z$, athermal $\Delta m_z$ (50 Hz - 2 kHz) where the change in the projection during resonant absorption is directly measured and the flux-locked loop cut-off.](image)

### 6.4.3 Single Crystal Ferrites (YIG)

The polished single-crystal YIG sphere has long been the standard material for demonstrating novel resonance detection methods mainly due to it’s exceptionally low damping at microwave frequencies and narrow line width. In 1958 LeCrew et al. observed "an extremely narrow line width of 520 millioersteds, (0.052 mT),” for a 350 $\mu$m YIG sphere using fixed frequency (9 GHz) cavity-based FMR [108]. Given that the
gyromagnetic ratio $\gamma$ of YIG is taken as 28 GHz/T and $\Delta H = \frac{\Delta f}{\gamma}$, this corresponds to a swept frequency resonant full width half maximum of just 1.46 MHz. It is therefore expected that much narrower peaks will be observed in comparison to section 6.4.1 which dealt with bulk, polycrystalline YIG.

### 6.4.3.1 Conventional Single Port VNA-FMR Inside the MPMS

Figure 6.28 shows tracing of the VNA-FMR resonance spectra with an applied magnetic field ranging from 70 mT to 450 mT for low temperature (a) 5 K and room temperature (b) 300 K. Once again, enhancement, broadening or splitting of the FMR spectra is observed at frequencies where there is strong coupling of the sphere’s FMR mode with the electrostatic modes of the microstrip.
Figure 6.28: VNA-FMR spectra for a single-crystal YIG sphere at (a) 5 K and (b) 300 K. The observed linewidths are much narrower than those shown for the polycrystalline specimen (fig.6.12).

The FMR spectra can be detected up to 14 GHz, beyond which signals are greatly diminished. The characteristic Lorentzian lineshape is observed for the range of applied fields and is much narrower than the spectra obtained for the polycrystalline specimen, as in figure 6.12. In comparison to the polycrystalline sample, post-processing is greatly reduced as the resonant interaction is much stronger and more sharply defined hence the signal to noise ratio is greater.
Figure 6.29: VNA FMR spectra obtained in a nominal 150 mT applied magnetic field. Strong coupling of the electrostatic modes of the microstrip with the resonant mode leads to line width broadening, as observed in the 5 K spectrum.

Figure 6.29 examines two resonant peaks obtained in a 150 mT excitation field at 5 K and 300 K. In agreement with Kittel’s equation, a shift in the resonance peak to a higher frequency is observed with decreasing temperature. Lateral broadening of the resonance peak is also observed. The full-width half-maximum, which is the standard measure of the resonance linewidth is extracted by fitting the curves to a Lorentzian profile. At 300 K the linewidth is measured as 8 MHz, which increases over fivefold to 44 MHz at low temperatures. This temperature dependent linewidth broadening for polished spheres of single crystal YIG was first reported by Dillon in 1956 [109, 110]. This he attributed to the increase in the first order magnetocrystalline anisotropy constant $\frac{K_1}{M_s}$, with decreasing temperature. The relaxation mechanisms that result in this temperature dependent broadening are described in section 1.3.2.2.
6.4.3.2 Athermal Absolute Detection

The SQUID-based absolute FMR detection method is again employed to characterise the magnetisation dynamics in the polished sphere of yttrium iron garnet. As mentioned previously, each material and shape of sample will have a different optimum
level of the chopping frequency of the input for the desired mode of operation. The
dependence of the SQUID output voltage on this chopping frequency under 2.67 GHz
and 100 mT field resonant excitation is shown for the YIG sphere in figure 6.31. The
three distinct regimes are again observed. From 0.1 to 2 Hz the excitation is weakly
modulated leading to the resonant generation of oscillatory thermal waves which alter
the projection of $m_z$. For 5 Hz - 2 kHz chopping absolute $\Delta m_z$ is detected and beyond
this we approach the cut-off of the flux-locked SQUID loop.

![Graph showing SQUID output voltage as a function of chopping frequency.](image)

**Figure 6.31:** SQUID output voltage as a function of the chopping frequency of the GHz excitation. Three
regimes are displayed: Thermal (0.1 - 2 Hz) where resonant heating changes the projection of $m_z$, athermal
$\Delta m_z$ (5 Hz - 2 kHz) where the change in the projection during resonant absorption is directly measured and
the flux-locked loop cut-off.

To evaluate resonance conditions in the athermal $\Delta m_z$ regime, an optimal chopping
frequency of 1 kHz was used. The dependence of the output signal on the input power,
as shown in figure 6.32 is once again quadratic on a linear scale, hence the maximum
power output of the signal generator, 21 dBm was used.
Figure 6.32: The dependence of the SQUID output on the input power is quadratic (on linear scale) as expected from the cosine expansion at small angles. Again the secondary y-axis indicates the corresponding change in the moment (nAm²) and hence opening of the precession-cone at resonance.

Figure 6.33: Comparison of a VNA and SQUID-detected resonant peak for the single crystal YIG sphere in a static 150 mT field.

To compare the VNA-FMR with the SQUID-detected method developed in this work, two resonant peaks in a nominal 150 mT field at 300 K are closely examined in
The characteristic sharp Lorentzian profile is observed, with the FWHM of the VNA-detected spectra measured as 11 MHz. The SQUID detected method measures an much sharper linewidth of just 4 MHz. This is attributed to the intermediate frequency filtering within the VNA itself. The linewidth, in the case of SQUID-detection is limited by the chopping or modulation frequency, or if that is sufficiently low, by the fundamental broadening for the sample. In the case of single-crystalline YIG spheres, this has been shown to be related to the quality of the surface polishing [108].

Figure 6.34 shows the correlation between the SQUID-detected resonant frequency and the applied magnetic field which displays good agreement with the 300 K VNA-FMR scan in figure 6.30. The gyromagnetic ratio is determined to be 28.2(4) GHz/T, which is even closer to than the VNA measured value to the standard $\gamma = 28$ GHz/T.

6.4.3.3 Thermal CW Detection

The same issue of diminished $\Delta m_z$ signal with decreasing temperature outlined for the polycrystalline sample in figure 6.18 is also evident for the YIG single-crystalline
sphere, hence detection of the thermal derivative of the magnetisation \((dm_z/dT)\) is used for low temperature FMR identification. Here the temperature dependence of the resonant frequency is examined in a continuous wave excitation and perpendicular 175 mT applied field. Figure 6.35 shows a characteristic thermal SQUID-FMR absorption curve at 25 K, again obtained by direct oscilloscope digitization of the SQUID voltage.

![Graph](image)

**Figure 6.35:** CW Microwave excitation and absorption detection via direct digitization (inset) of the SQUID output voltage \(U_{\text{SQUID}}\) at 20 K. For the case of single crystal YIG, a single sharp resonant peak is observed.

Figure 6.36 shows the absorption curve at 120 K, which appears to indicate extensive broadening with increasing temperature. While the FMR signal tends to narrow with increasing temperature via the mechanisms described in section 1.3.2.2, the thermal time constant, which increases with increasing temperature has a dominating effect on linewidth in thermal regime detection. Hence a superposition of two exponents is observed; two-magnon scattering relaxation which is rapid at higher temperatures and the dominant thermal relaxation which increases with increasing temperature. One approach to counteract this thermal induced broadening would be reduce the frequency sweep rate and allow thermal relaxation over a longer time. However, as discussed in section 6.4.2 the primary issue with this being that the SQUID PID temperature controller will have time to register the changing temperature resulting from the resonant
excitation and react to counteract it, thus significantly reducing the measured \( \frac{dm_z}{dT} \).

**Figure 6.36:** Thermally detected resonance spectra taken at 125 K. The CW thermal regime detection shows extensive broadening at temperatures above 100 K due to the increase in the thermal time constant (i.e. longer thermal relaxation times). The noise floor is set by the digital noise due to the 8-bit output of the scope.

**Figure 6.37:** The shift in the resonant frequency with changing temperature for the single-crystal sphere is less pronounced than that observed for the bulk polycrystalline piece as shown in Fig. 6.26. In addition to the primary [1 1] mode, two magnetostatic modes are identified [2 2] and [3 3]. It could be noted that a much weaker mode is seen at higher frequencies, which may be identified as the [2 1] mode.
The temperature dependence of the thermally detected resonant frequency for the YIG sphere is shown in Fig. 6.37. The signal is again strongest at low temperature, where the thermal time constant tends to zero and resonant signals are measurable at temperatures down to 5 K, limited by the power dissipation at the 50 Ω termination resistor and the amount of available cooling power. Above 100 K, as \( \tau \) becomes significant (see Fig. 6.20) linewidth broadening becomes especially pronounced. The inhomogeneity of the RF excitation excites higher-order non-uniform resonant modes, the strongest of which are indexed as [1 1], [2 2] and [3 3] \([27, 28, 93]\). The shift in the resonant frequency of the [1 1] mode with increasing temperature is shown to be much less dispersive than that observed for the polycrystal (Fig. 6.26) and also less dispersive than the higher modes. This is likely due to the higher contribution of surface anisotropy to the resonance conditions for the higher modes. As anisotropy tends to gap the dispersion relations of the magnetostatic modes, their population intensities tend to drop correspondingly, at low temperatures.

### 6.5 Two port Resonator

In order to gain the additional ability to characterise thin films or random/templated nano-structures the power of the excitation would need to be increased but we are currently limited by the fact that the signal generator is operated in a single-port configuration and the power is dissipated at the 50 Ω termination at the end of the microstrip. As such, increasing the power would lead to an increase in the residual heating within the sample space, which may be detrimental during low temperature measurements. In order to circumvent this issue, a new reflection-transmission (return-track geometry) design based on the same principle is constructed. Power can then be dissipated in a 50 Ω resistor externally at ambient temperature or for the case of VNA-FMR be connected to a secondary port, such that the two-port S parameters can be determined. Knowledge of both the reflection and transmission coefficients allows quantitative extraction of the resonant absorption coefficients. In addition to this, external heat dissipation would enable the measurement of ferromagnetic resonance at very high excitation power (in excess of tens or even hundreds of watts) and access
to very non-linear dynamics, at large precession angles. The same is important, for example, for the definition of the fundamental limits of intermodulation distortion in critical high-performance microwave devices, such as filters and stripline circulators.

The new set-up contains many of the same features as the previous, such as the 8 mm diameter Cu shielding, except two semi-rigid coaxial lines feed into either side of a double sided co-planar waveguide (i.e. two CPWs with a shared dielectric) carrier PCB. The thickness of the dielectric (t=1.6 mm) is far greater than the pitch between signal and ground lines (w=0.3 mm) and hence is sufficient to avoid cross-talk between lines while still fitting within the 8 mm shield. One side acts as the input port and the sample is positioned, just to the side of the CPW’s signal conductor. The input CPW is then connected at the end to the opposing side by a via (vertical interconnect access), which feeds externally through to the secondary coaxial line. The two coaxial lines are held together using evenly spaced 4.6 mm diameter, 2 cm length stainless steel shrouds and Teflon washers to allow the rod to glide smoothly through the MPMS system. The top of the rod has a longer tube section, to which the transport sliding clamp is anchored. The Swagelok-based clamp as shown in figure 6.38 was made to fit the outer diameter of the stainless steel shroud.

![Figure 6.38: The Swagelok-based clamp is designed to fit the diameter of the stainless steel shroud which is used to encapsulate the two coaxial lines. When hooked into the MPMS transport module, it allows motorized transport of the CPWs though the SQUID detection coils.](image)

The longer tube section is fitted with a custom made vacuum assembly, commonly known as a ’slide-seal,’ fitting based on the Quantum-Designs model [111, 112]. This assembly, when installed at the top of the sample transport option provides a vacuum seal while allowing the stainless steel rods to move and the CPW to translate through the gradiometer coils. Figure 6.39 shows a schematic of the vacuum seal part which
consists of a chamber maintained using spring compression which divides two sealing sections. In figure 6.39, parts 2 form an inner seal on the machined brass piece, while 3 are smaller rubber o-rings which form an outer seal on the stainless steal shroud used to house the two co-axial lines. All components are held within the brass piece using snap-ring compression.

The Quantum-Design model uses a series of Teflon spacers and rubber lip-seals to maintain the chamber, in place of the spring compression design discussed here. However, rubber seals are more prone to degradation, both thermal and friction if the rod it not kept well lubricated with vacuum grease. When in operation, the chamber is continuously flushed with helium gas from the dewar via the two-ports on the brass piece, thus minimizing the amount of air pulled through the chamber as the rod moves through the seal. Having both the ‘slide-seal’ assembly and the clamp to lock on to the transport motor means that it is possible to perform AC magnetometry. The complete ‘dual-port’ assembly is shown in figure 6.40.
Figure 6.40: Complete return-track SFMR rod consisting of (A) the Swagelok-based clamp for attaching to the transport motor, (B) the ‘slide-seal’ assembly and (C) the copper shield which houses the double-sided PCB. Metallic spacers are placed at regular intervals to strengthen the rod and give the two-line system rigidity. Teflon spacers allow the rod to smoothly glide through the sample-space of the QD MPMS system.

Figure 6.41 shows the TDR-detected, reflected and transmitted signals through the entire rod, where a fast edge pulse is launched into the SMA attached to one coaxial line and the transmitted signals are fed into the second TDR port through the SMA on the other line. Small losses are observed in coaxial 1 and coaxial 2, most likely due to their length. The two coplanar waveguides are labelled as two well defined capacitive distortions to the incident signal. This may be due to a number of factors, the most
likely of which are an impedance mismatch in the CPWs, or discontinuities due to solder connections or vias.

![TDR measurement](image)

**Figure 6.41:** TDR measured reflected and transmitted signals arising from the return-track set-up consisting of two CPWs and two co-axial feed lines.

To assess the capability of the return-track module and compare with the 50 Ω terminated design, magnetisation dynamics is again examined in the polished sphere of single-crystalline YIG.

Automatic centering within the gradiometer detection coils, while maintaining vacuum is made possible by the combination of the ‘slide-seal’ piece and the transport clamp. Figure 6.42 shows the moment scan for the length of the excitation region of the SFMR module, where the strong peak at 4.2 cm indicates the position of the YIG sphere. The shape is a result of the QD software modelling the sample as a point dipole moment moving through the gradiometer coils. Centering is performed at room-temperature (300 K) and then again at 5 K, when performing low-temperature measurements.
Figure 6.42: Moment scan over the length of the return-track FMR module in order to centre the sample within the detection coils. The central peak is indicative of the strong moment from the YIG sample. *The curvature at the maximum position (12 cm) is due to the moment of the brass end-piece of the SFMR module.

6.5.1 Semi-Automated Athermal Detection

The primary advantage of a two-port system is that it allows for higher input power and hence higher power absorption by the sample, which can greatly improve the sensitivity of the technique. This improvement is demonstrated in the automated sweeping of the athermally-detected SFMR spectra. Here the first module port provides 1 kHz chopped excitation signal to the sample via a 1-22 GHz signal generator and the second port feeds the exiting signal into a 30 GHz bandwidth scalar spectrum analyser, such that the power dissipation can be monitored. The resonant response is again monitored by the lock-in demodulated and processed SQUID voltage. A pulsed signal from a TTi arbitrary waveform generator provides synchronised triggering to both the signal generator (to initialise the sweep) and an oscilloscope connected to the lock-in amplifier (to begin the trace). The digitized SQUID-response can then be recorded through GPIB interface using Labview software. The circuitry is depicted in figure 6.43.
Figure 6.43: Circuit design for semi-automated scanning of athermal SQUID FMR.

Figure 6.44: The transmitted signal through the rod assembly shows strong attenuation beyond 10 GHz due to the dispersive nature of the FR4. In addition, losses occur at sharply defined frequencies corresponding to line resonances within the assembly.

Some of the issues with the previous design are still unavoidable, for example, at specific frequencies resonant modes of the sample may couple strongly to electrostatic modes of the transmission lines and lead to distortion of the lineshape and if the
interaction is constructive will lead to thermal propagation. At higher frequencies, in excess of 10 GHz power delivery becomes an issue due to the dispersion of the FR4 PCB board, which becomes highly lossy at high frequencies. Figure 6.44 shows the amplitude of the transmitted signal over the 22 GHz bandwidth, detected using the scalar spectrum analyser, which shows strong attenuation for frequencies in excess of 12 GHz and also high losses at specific frequencies corresponding to line resonances within the SFMR assembly. The coupling of FMR modes with line resonances, resulting in thermal propagation is depicted in figure 6.45a, where below 125 mT the amplitude of the primary mode increases significantly and the linewidth broadens and gains an asymmetric shape, characteristic of thermal relaxation.
For applied fields in excess of 135 mT, multiple higher magnetostatic modes in addition to the primary mode are easily distinguished. Three of these higher modes are traced in figure 6.45b but more may be seen at certain fields. For frequencies above 5.5 GHz, the amplitude of the resonant response is diminished, hence the signal to
noise ratio decreases.

Figure 6.46: Taking a close look at the field dependence of the automatically scanned athermal SFMR spectra, the higher order magnetostatic modes appear to be more dispersive than the primary mode.

Figure 6.46 shows a close examination of the evolution of the resonant peaks with increasing field strength. It appears that higher order modes are more dispersive than lower order ones (i.e., the separation between successive modes increases with the applied field).

6.6 Summary

This study culminated in the design and operation of two contactless SQUID-based FMR detection systems, capable of operating in a broad range of experimental temperatures and fields. This is believed to be the first reported detection of ferromagnetic resonance effects using SQUID-magnetometry. SQUID-FMR detection allows for absolute detection of the absorbed power on resonance, unlike conventional cavity or stripline-based FMR, which gauge resonant absorption by monitoring microwave losses. Additionally it has the added advantage of allowing for a qualitative insight into the precession dynamics. In addition to SQUID-detection, conventional VNA-FMR detection is possible and the two methods are complementary. The modules have been
constructed to fit the sample space of a Quantum Design magnetic property measurement system MPMS® XL[89] but are compatible with most commercial SQUID magnetometer systems. By chopping the external microwave excitation at different frequencies, the system may be operated in two regimes, thermal and athermal which give absolute values of $\Delta m_z$ and $dm_z/dT$ respectively, as the material is driven at its resonance frequency.

The operation of the single port, 50 $\Omega$ terminated SQUID-FMR module has been successfully used to observe the magnetisation dynamics of two resonant modes in a rectangular piece of bulk $Y_3Fe_5O_{12}$ and the much more sharply defined resonant modes of a sphere of single crystal YIG with varying magnetic field and temperature. Due to the inhomogeneous contribution to the linewidth, linewidths for the polycrystalline specimen are shown to be much broader than those of the single-crystalline sphere. For the case of the single crystal, linewidths as narrow as 4 MHz are reported using the athermal SQUID-detection strategy and the linewidth increases with decreasing temperature due to the increase in the magnetocrystalline anisotropy. In addition to the spatially uniform primary mode, higher order magnetostatic modes are observed in the single-crystalline sphere, which are driven by the inhomogeneity of the RF excitation field.

A secondary return-track geometry SFMR module is constructed which allows for power dissipation external to the cryogenic environment of the sample space. This allows for greater input excitation power and hence greater absorption by the sample, which can significantly improve the absolute sensitivity of the technique. This is demonstrated by performing automated frequency sweeping of the athermal SFMR for the single crystal YIG sphere. Using this method, multiple higher order modes (three are tracked) in addition to the uniform mode are directly observable.

Potentially, all three detection strategies demonstrated (absolute $\Delta m_z$, thermal $dm_z/dT$ and VNA-FMR) could be combined (in real-time) with DC and AC magnetisation measurements, dramatically expanding the amount of sample characterisation
data, and allowing for the quantitative discrimination between dissipative and dispersive resonant and non-resonant effects. The strategies discussed can be generalised to vacuum cavity waveguides in order to access higher frequencies (> 20 GHz) that are otherwise inaccessible. For frequencies in the range of 20 GHz to 50 GHz this can be done in sets of shorted discrete frequency cavities and beyond 50 GHz it would even be possible to use a return track geometry. Provided the power can be delivered to and absorbed by the sample, FMR, EPR and high-field-NMR can all be detected using SQUID-based techniques.
Chapter 7
Conclusions and Outlook

7.1 Conclusions

The premise of this thesis was to expand the range of high-frequency broadband measurements for electrical conductivity and in turn magnetisation dynamics. To start, a complete understanding of wave propagation in high-frequency waveguides was established. This was done for both planar and rectangular cavity geometries for the applications discussed. This formed a strong basis for, the main outcome of this work, which was the creation of novel instrumentation for broadband FMR detection. The main findings are detailed in this section.

This work shows the capability of microwave techniques, conventionally used for thin-film characterisation in analysing wave propagation in randomly orientated nanostructures. The conduction properties for a range of nanowire arrays have been non-destructively characterised using both CW VNA ($< 3$ GHz) and pulsed TDR analysis ($< 50$ GHz). In the pulsed regime, the impedance of Ag NWNs has been shown to scale linearly with the optical transmittance of the network (see figure 3.4 Chapter 3). It is shown for Ag NWNs that denser (smaller % T) networks display enhanced reflected signals due to the increased number of capacitive junctions. This is in good agreement with the results obtained using the swept frequency reflectometer (VNA). Hence it can concluded that such contactless techniques, conventionally used for analysing thin-film structures have application in the characterisation of conductive disordered systems. In a more destructive technique an attempt was made to achieve the low-resistance state for the NWNs using direct galvanic pulsing of CW power via CPW. Marginal increases in conductivities of such networks are shown after the CW pulsing but as mentioned in Section 3.4 the technique is limited by the reflective properties of the NWNs, which behave like metallic thin films. Laser specular scattering techniques have been exploited to enhance understanding of film surface topography. The characteristic NW lengths were determined to be approximately 50-70 $\mu$m and 25 $\mu$m for Ni and Ag NWs.
respectively.

Using the same principles established in the results previously discussed, sets of finite bandwidth frequency cavities were simulated, constructed and used to evaluate inkjet printed silver films as potential coatings for EMI shielding applications. Comprehensive microwave reflectometry analysis was performed in three distinct frequency bands (X-Ku: 7-14 GHz, K: 15-20 GHz and Ka: 21-42 GHz) for films of varying thickness, in order to establish the threshold at which the films transition from a microwave transparent system to a reflective regime. It was shown that films with a thickness beyond 120 nm are consistently effective EMI shields but the rate at which they transition to shielding behaviour does depend on the frequency of the stimulus. That is, the higher frequency stimuli show greater penetration depth within the Ag/PET films. This analysis was further extended to higher frequencies and shorter wavelengths through high-visible-UV densitometry, which indicated that the frequency dispersion hold true at optical frequencies i.e. a film that is close to 100 % reflection at microwave frequencies would have significant UV/visible transmission.

Supplementary to this, the effect of geometric structuring of Ag films on the microwave propagation was also discussed for applications where retaining the maximum visual throughput without compromising the shielding behaviour is desired. The structures consisted of grids where the metallic linewidth was kept a constant 1 mm and the pitch between lines was varied. It was shown that for grids with metallic fill factor greater than 55 % desirable EMI shielding can be achieved. This is however strongly dependent on the intended operational band. For example a 55 % dense grid will shield 98 % of incident energy in the K-band but will only shield 77 % in the 28-30.5 GHz band. Hence, meshes are not ideal for universal broadband application and the pitch must be tailored for shielding within a specific frequency band. For a metallic fill greater than 75 % ($D = 1$ mm) uniformly high shielding characteristics are observed.

Novel instrumentation and strategies for FMR detection were introduced. These techniques rely on SQUID-detection of small changes in the magnetisation projection along the applied field direction, resulting from FMR absorption. To demonstrate the
functionality of the new techniques, magnetisation dynamics was analysed in bulk polycrystalline and single crystalline YIG. The inhomogeneity of the microstrip/CPW RF field allows for the excitation and detection of higher-order magnetostatic modes, in addition to the primary spatially uniform one described by Kittel dynamics. In addition, tracking of the evolution of the very broad resonances at low fields is accessible, which is rather difficult to extract in classical inductive FMR detection strategies. Two different module geometries have been constructed. The first is a single track, 50 Ω terminated system and the second is a return-track design, which potentially allows for high power excitation and therefore analysis of non-linear effects resulting from very large angle precession cone opening. The two new techniques can be combined, essentially in real time, with conventional VNA-FMR, DC magnetometry and AC susceptibility to greatly expand the amount of static and dynamic information available using a single module.

7.2 Outlook

The quest to access high frequencies in materials and devices is an ongoing process but SQUID-FMR detection has shown potential as a method of contactlessly assessing resonant excitations. Provided power can be efficiently delivered to and absorbed by the sample, SQUID-based detection is accessible. This opens the possibility of being able to analyse not only FMR but also EPR or even NMR at sufficiently high fields to have the resonant frequency in the GHz range. There is an existing initiative within the European high Magnetic Field Lab (EMFL), in particular the GHMFL, for constructing NMR spectrometers operating at fields in excess of 42 T and performing scalar NMR experiments at frequencies of up to 2 GHz. The process suggested above should be applicable to high field SQUID magnetometers (systems already in existence in operation of fields up to 24 T). A back-of-the-envelope style calculation shows that apart from the order of magnitude of the total moment of a plausible sample of say water, being certainly sufficiently large (about $2 \times 10^{-4}$ Am$^2$ for a 0.25 mm$^3$ sample, when compared to a 1 mm diameter YIG sphere of moment $7 \times 10^{-6}$ Am$^2$), as the Q-factor of the resonance is hundreds to thousands of times higher, the detectivity of
the same should be in orders of magnitude better. One would, of course, use few turns of copper coils, rather than microstrips or coplanar waveguides at the frequencies of interest for FMR at fields below 5 T - 100 MHz, or so.

To perform higher frequency excitation, alternatives to the PCB dielectric would need to be assessed, as the dispersive nature of the material is the primary limit in delivering power to the sample. Low loss high frequency ceramics may be considered to address this issue. The methods developed are not limited to microstrip or CPW excitation and can be generalised to other waveguide forms. In-fact, to access frequencies beyond 20 GHz, rectangular cavities can be used. Here the main limit is the diameter of the sample-space within the SQUID-magnetometer system but this would not be an issue for the higher frequency waveguides, as the cavity dimensions are inversely proportional to the operating frequency. It may be even possible to create return-track modules for waveguides with operating frequencies in excess of 40 GHz.

As briefly mentioned in section 6.4.1.2 it would, in principle, be possible to utilise the thermal regime detection to perform absolute calorimetry. This could potentially be done by mounting millimetre and sub-millimetre samples on a thin wire (25 µm diameter, gold-plated tungsten) symmetric line (twinax®) assembly.

High-field FMR can, in theory be performed using the same modules designed here, in the Quantum Design physical property measurement system (PPMS) which would allow external fields of up to 14 T. Here it would be operated in a purely conventional VNA-FMR fashion but would still allow for easy assessment of both NMR and broadband FMR.

To enhance the capability of the return-track SFMR assembly, a larger power dynamic alternating field source or alternatively a suitable broadband, high power amplifier must be employed to increase the power capable of being absorbed by the sample. This would allow for access to the non-linear dynamic regime at very large precession angles. This effectively allows for the critical evaluation of the limits of intermodulation distortion in high-power, high-performance, FMR-based non-reciprocal devices such as circulators, isolators and filters. In effect, this will set the limits of the density
of the high bandwidth mobile communications network of the future (5G and beyond).
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Presentations and Publications

- **Poster** “Contactless Characterisation of Random Nanowire Arrays; Microwave Conductivity and Permittivity and Laser Specular Scattering” European School of Nanoscience and Nanotechnology, Grenoble, France, August 2014

- **Poster** “Contact and Contact-less Nanowire Characterisation - Single Wires and Networks” AMBER Review, Dublin, February 2015

- **Poster** “Addressing Random Nanowire Arrays Using Contactless Characterisation Techniques” IEEE Magnetics Society Summer School, Minneapolis, Mn, USA, June 2015

- **Poster** “SQUID-Detected Broadband Ferrimagnetic Resonance in Bulk Y₃Fe₅O₁₂” Joint European Magnetic Symposia, Glasgow, UK, August 2016

- **Talk** “SQUID-Detected Broadband Ferrimagnetic Resonance in Bulk Polycrystalline Y₃Fe₅O₁₂” Intermag2017, Dublin, Ireland, April 2017

Appendix

A Transverse Magnetic Propagation in Cavity Waveguides

This section shows solutions to Maxwell’s equations for TM wave propagation in a hollow rectangular waveguide and is formulated similarly to that described in section 1.3.1.1 for TE propagation.

**Assumption**: $H_z = 0$ and $E_z \neq 0$ and satisfies

$$\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} + k_c^2\right)e_z(x, y) = 0. \quad (1)$$

$E_z(x, y, z) = e_z(x, y)e^{-j\beta z}$

**General Solution**: Again the separation of variables described in section 1.3.1.1 yields a general solution for $E_z$ of the form

$$e_z(x, y) = (A \cos k_x x + B \sin k_x x)(C \cos k_y y + D \sin k_y y) \quad (2)$$

**Boundary Conditions**:

$$e_x(x, y) = 0 \text{ at } x = 0, b \quad (3a)$$

$$e_y(x, y) = 0 \text{ at } y = 0, a \quad (3b)$$

which can be applied directly to the general solution giving $A, C = 0$, $k_y = \frac{n\pi}{b}$ and $k_x = \frac{m\pi}{a}$, which gives the solution

$$E_x(x, y, z) = G \sin \frac{m\pi x}{a} \sin \frac{n\pi y}{b} e^{-j\beta z} \quad (4)$$

where $G$ is a constant consisting of the two constants $B$ and $D$.

**Transverse Field Components**:

$$H_x = \frac{j\omega \varepsilon}{k_c^2} \frac{\partial E_z}{\partial y} = \frac{j\omega \varepsilon n\pi}{k_c^2 b} G \sin \frac{m\pi x}{a} \cos \frac{n\pi y}{b} e^{-j\beta z} \quad (5a)$$

$$H_y = \frac{-j\omega \varepsilon}{k_c^2} \frac{\partial E_z}{\partial x} = \frac{j\omega \varepsilon m\pi}{k_c^2 a} G \cos \frac{m\pi x}{a} \sin \frac{n\pi y}{b} e^{-j\beta z} \quad (5b)$$
B  Additional Transmission Line Geometries

The table below illustrates the cut-off frequency ($f_{\text{cut-off}}$) and characteristic impedance ($Z_0$) for a number of commonly used transmission media. The corresponding geometries are shown in figure 1b.

Table 1: Different Waveguide Geometries

<table>
<thead>
<tr>
<th>Name</th>
<th>$f_{\text{cut-off}}$</th>
<th>$Z_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Circular waveguide (a)</td>
<td>$\frac{k_c}{2\pi\sqrt{\mu\varepsilon}} = \frac{p'<em>{nm}}{2\pi\sqrt{\mu\varepsilon}}$ where $p'</em>{nm}$ is the root of the derivative of the first kind (see [1])</td>
<td></td>
</tr>
<tr>
<td>Coaxial line (b)</td>
<td>$\frac{k_c}{2\pi\sqrt{\varepsilon_r}}$</td>
<td>$\frac{\nu k}{\beta}$</td>
</tr>
<tr>
<td>Stripline (embedded microstrip) (c)</td>
<td>na</td>
<td>$Z_0 = \frac{\beta}{\sqrt{\varepsilon_r}} W_{\text{eff}} + 0.441H$</td>
</tr>
</tbody>
</table>

$w_{\text{eff}} = \left\{ \begin{array}{ll} 0 & W > 0.35 \\ (0.35 - \frac{W}{H})^2 & \frac{W}{H} < 0.35 \end{array} \right.$

$k, k_c, \beta$ and $w_{\text{eff}}$ are the wavenumber, cut-off wavenumber, propagation constant and effective width respectively.

$k_c, k, \beta$ and $w_{\text{eff}}$ are the wavenumber, cut-off wavenumber, propagation constant and effective width respectively.

---

(a) Circular waveguide of radius $r$.  
(b) Coaxial transmission line.  
(c) Stripline transmission line.

Figure 1: Different transmission line geometries.
C Supplementary Laser Specular Scattering data

This appendix includes the supporting data for sections 5.3.1 and 5.3.2 to illustrate the reproducibility of the NW lengths measured in the main body of the text. Each section displays the raw laser specular image, the dewarped specular image, the image after background subtraction and the corresponding plot of the scattering intensity as a function of the scattering wave number. The captions indicate the measured NW lengths in each case.

Transmission Ni NWNs 80 % T

(a) As captured specular scattering on a 10x10 cm screen.
(b) Dewarped specular image which includes the background specular reflections from the substrate.
(c) Dewarped specular image excluding the background signal

Figure 2: Transmission mode small angle optical scattering for 80 % T Ni NWNs
Figure 3: Transmission mode obtained scattering intensity as a function of the scattering wave vector for 80 % T Ni NWNs. The average NW length is estimated to be 36-66 µm.

Transmission Ni NWNs 90 % T

(a) As captured specular scattering
(b) Dewarped specular image which includes the background specular reflections from the substrate.

(c) Dewarped specular image excluding the background signal

Figure 4: Transmission mode small angle optical scattering for 90 % T Ni NWNs
Figure 5: Transmission mode obtained scattering intensity as a function of the scattering wave vector for 90 % T Ni NWNs. The average NW length is estimated to be 46-63 µm.

Reflection Ag NWNs 75.4 % T

Figure 6: Reflection mode small angle optical scattering for 75.4 % T Ag NWNs
Figure 7: Reflection mode obtained scattering intensity as a function of the scattering wave vector for 75.4 % T Ag NWNs. The average NW length is estimated to be 25-40 µm.

Reflection Ag NWNs 90.6 % T

Figure 8: Reflection mode small angle optical scattering for 90.6 % T Ag NWNs
Figure 9: Reflection mode obtained scattering intensity as a function of the scattering wave vector for 90.6 % T Ag NWs. The average NW length is estimated to be 23-37 μm.