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DOCTORAL THESIS

Demonstration of Magnetism in Thin Films at Optical Frequencies

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Declaration of Authorship

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"And your doubts can become a good quality if you school them, they must grow to be knowledgeable, they must learn to be critical, as soon as they begin to spoil something for you, ask them why a thing is ugly, demand hard evidence, test them, and you will perhaps find them at a loss and short of an answer or perhaps mutinous. But do not give in, request arguments, and act with this kind of attentiveness and consistency every single time, and the day will come when instead of being demolishers they will be among your best workers - - perhaps the canniest of all those at work on the building of your life."

Letters to A Young Poet - Rainer Maria Rilke

Abstract

Demonstration of Magnetism in Thin Films at Optical Frequencies

by Edward Hilding KROCK

Obtaining a non-unity value for magnetic permeability, $\mu \neq 1$, at optical frequencies is typically associated with subwavelength arrays of structured materials known as metamaterials, this is achieved by the shaping of current densities into circulating currents that can give rise to magnetic multipoles.

Such magnetic responses can enable the design of new devices at optical frequencies utilizing phenomena normally restricted to radio and micro-wave frequencies such as s-polarized Brewster angles, negative refraction lenses and transverse electric surface waves. However, given that these magnetic responses are confined to complex and difficult to fabricate metamaterials, there is limited demonstration of these phenomena, and fewer direct applications. Therefore, the demonstration and application of these magnetic responses in simpler systems, such as thin films, is of great interest, and is the focus of this work.

In this thesis, we derive an angle dependent multipole decomposition model, allowing us to decompose the reflection and transmission spectra of a sub-wavelength SiN thin film in terms of electric and magnetic multipoles. We

then devise an experiment to verify these predictions across visible and near-infrared frequencies. Showing that well known phenomena, such as Fabry Perot modes and p-polarized Brewster angles can only exist if one includes magnetic responses by accounting for magnetic dipoles, quadrupoles and octupoles.

We identify Fabry Perot modes as arising from the destructive interference, in the direction of reflection, between even and odd parity multipoles, which include both electric and magnetic responses, regardless of polarization or mode order.

The p-polarized Brewster angle is identified as resulting from the direction of reflection and electric dipole moment aligning, resulting in zero reflection. This is in agreement with literature, however, we demonstrate an additional, previously unobserved condition to observe the Brewster angle, the destructive interference between electric and magnetic multipoles.

We notice the possibility of additional Brewster angles associated with electric and magnetic multipoles other than the electric dipole, which arise from the alignment between the direction of reflection and the associated multipole moment. These new Brewster angles are hidden in the reflection spectra of our thin film, as the destructive interference between remaining multipoles is absent, but it is not possible to accurately model reflection without these hidden Brewster angles. We suggest several cases where metamaterials may be capable of supporting these new Brewster angles and demonstrate methodologies to unambiguously identify them.

Next we explore how to quantify the effective permittivity and permeability

of thin films arising from these multipoles, by evaluating how these multipoles polarize our thin films. We demonstrate how the presence of spatial dispersion, the wavevector dependence of the electric permittivity can arise from either all magnetic multipoles or from electric multipoles that are not the electric dipole. Spatial dispersion plays a vital role in magnetic responses at optical frequencies, given that it is typically understood as being inseparable from magnetic permeabilities at optical frequencies.

Next we move to study thin films that are significantly larger than the wavelength of the incident light, by exploring the reflection of a $50\text{ }\mu\text{m}$ thick film of SiO_2 . We demonstrate how spatial dispersion may be identified by the dispersion relation of the Fabry Perot mode in cases when multipole decomposition is difficult to practically use.

Having demonstrated how thin films may support magnetic responses through magnetic multipoles and higher order electric multipoles, we then move to demonstrate an application of the magnetic response. We modify the response of a low loss Fabry Perot resonator consisting of 1200 nm of PMMA by depositing it on a thin film of 1 nm Ti and 40 nm Au. Through eigenmode analysis we demonstrate the existence of several modes that arise from the coupling between the Fabry Perot mode in the PMMA cavity and a transverse electric surface plasmon in the metal layers.

A transverse surface wave is identified by two features in the structure. Firstly, the asymmetric Fano lineshapes, which are associated with coupled modes, which were observed in both measurement and model. Secondly, eigenmode analysis identifies a strong magnetic field enhancement along the direction of

propagation of the mode, which is a feature unique to transverse electric surface plasmon modes. Simulations introducing an air spacer layer in between the Au and PMMA layers shows this TE surface wave as an evanescent mode confined to the interface between the Au and air layers. However we propose further work to unambiguously identify this surface wave as a surface plasmon in measurements.

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First and foremost, my utmost thanks to Professor John Francis Donegan's mentorship as my PhD supervisor and his continued interest in the work of this thesis. The support, guidance and patience he has provided has been an invaluable asset though this journey. I could not ask for a more ideal supervisor!

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A final thanks to the keg of mead that I brewed four years ago at the beginning of this journey and plan to open to mark the end of the journey. I will soon find out if it has become vinegar!

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List of Abbreviations

TM	Transverse Magnetic
TE	Transverse Electric
GaP	Gallium Phosphide
SiO₂	Silicon O₂Dioxide
SiN	Silicon Nitride
YVO₄	Ytrrium Orthovanadate
Ti	Titanium
Au	Aurum/Gold
Cr	Chromium
PMMA	Poly(methyl methacrylate)
D	Dimension
FP	Fabry Perot
S-parameter	Scattering Parameter
ED	Electric Dipole
MD	Magnetic Dipole
EQ	Electric Quadrupole
MQ	Magnetic Qudarupole
EO	Electric Octupole
MO	Magnetic Octupole
VIS	Visible
NIR	Near Infra Red

PL	Propagation Length
FSR	Free Spectral Range

Physical Constants

Speed of Light $c_0 = 2.997\,924\,58 \times 10^8 \text{ m s}^{-1}$

Permittivity of Free Space $\epsilon_0 = 8.854\,187\,82 \times 10^{-12} \text{ m}^{-3} \text{ kg}^{-1} \text{ s}^4 \text{ A}^2$

Permeability of Free Space $\mu_0 = 1.256\,637\,06 \times 10^{-6} \text{ kg m s}^{-2} \text{ A}^{-2}$

List of Publications

1. **Edward H.Krock**, John F. Donegan. "Observation of Dipole, Quadrupole and Octupole Brewster Angles in Thin Films". (In Preparation).
2. **Edward H.Krock**, Mughaid Ali, John F. Donegan. "Exciting a TE Surface Plasmon with the Magnetic Response of a Thin Film" (In Preparation).
3. *Poster* **Edward H.Krock**, John F. Donegan. "Slow and Fast Light in Coupled Waveguide-ENZ Modes". Photonics Ireland 2023 Conference, 4-6 th of September.

List of Symbols

ω	Angular Frequency	Rad
λ	Wavelength	m
$\vec{k}$	Wavevector	m^{-1}
$\vec{E}$	Electric Field	V m^{-1}
$\vec{H}$	Magnetic Field Strength	A m^{-1}
$\vec{P}$	Electric Polarization Field	C m^{-1}
$\vec{D}$	Electric Displacement Field	C m^{-1}
$\vec{B}$	Magnetic Field	T
$\vec{J}$	Current Density Field	A m^{-2}
$j_n(a)$	Bessel Function of the First Kind	Unitless
$Y(\theta, \phi)$	Spherical Harmonic Function	Unitless
ϵ	Electric Permittivity	Unitless or F m^{-1}
μ	Magnetic Permeability	Unitless or H m^{-1}
γ	Decay Constant	Unitless
θ	Angle	Degrees
n	Refractive Index	Unitless
$k(\lambda)$	Absorption Coefficient	Unitless
r	Reflection Coefficient	Unitless
t	Transmission Coefficient	Unitless
R	Reflection	Unitless

ϕ	Phase	Unitless
$\hat{U}$	Unit Tensor	Unitless
$\vec{r}$	Positional Vector	m
$\vec{p}$	Electric Dipole Moment	C m ⁻¹
$\vec{m}$	Magnetic Dipole Moment	A m ⁻²
$\hat{E}Q$	Electric Quadrupole Moment	C m ⁻²
$\hat{M}Q$	Magnetic Quadrupole Moment	A m ⁻³
$\hat{E}O$	Electric Octupole Moment	C m ⁻³
$\hat{M}O$	Magnetic Octupole Moment	A m ⁻⁴
$\Delta\nu_{FSR}$	Free Spectral Range	m
$\Delta\nu_c$	Linewidth	m

Chapter 1

Introduction

1.1 The Difficulty of Obtaining $\mu \neq 1$ at Optical Frequencies

When studying the electric and magnetic properties of bulk materials for sufficiently high frequency fields, it is common to ignore the magnetic properties, i.e, we set the magnetic permeability, μ , to one, and only consider the electric properties, i.e, the electric permittivity, ϵ , is not one.

This convention has been verified over the last century by a variety of researchers. In particular in 1961, Landau and Lifshitz [1] were the first to theoretically explore the problem. Merlin in 2009 [2] and Agranovich in 2006 [3] expanded on this issue for more nuanced cases of large permittivity values and spatial dispersion respectively.

Experiments to demonstrate this fading magnetic permeability predate theoretical descriptions. In 1945, Allanson [4] reviewed a large collection of literature measuring the high frequency magnetic permeability of various wires, regardless of material or structural properties of the studied wires, the magnetic permeability would always trend to one as the frequency approached microwave frequencies. H.Němec et al [5] performed more recent measurement that measured this phenomena to the highest frequency to date, in the mid to far-infra red range.

However, despite these demonstrations, observations of $\mu \neq 1$ are still feasible at optical frequencies. In 1999, Pendry [6] proposed that structures smaller than the wavelength of light, known as metamaterials, may be capable of

supporting effective magnetic permeabilities at optical frequencies, a regime many of orders of magnitude higher than microwave frequencies. This work was soon followed by a practical demonstration of phenomena associated with $\mu \neq 1$, negative refraction, by Smith et al [7].

The use of metamaterials to achieve magnetic responses is now typically referred to as "Optical Magnetism", whereby one shapes a subwavelength structure to engineer the correct current densities to support a magnetic dipole [8].

We move to discuss how these two separate views, the bulk and metamaterial cases are not in conflict with each other, but rather complement each other, by exploring the nature of electric and magnetic fields through Maxwell's equations and the nature of electric and magnetic permeabilities.

1.2 Maxwell's Equations, Wave Equation and the Speed of Light

The speed of light is a fundamental physical constant, approximately equal to $3 \times 10^8 \text{ m s}^{-1}$. This speed is altered when light interacts with matter.

Free space light can be described as a transverse, traveling wave with electric, $\vec{E}$, and magnetic, $\vec{B}$ fields, the properties of these fields are described with Maxwell's equations, as shown below

$$\nabla \cdot \vec{E} = 0 \quad (1.1)$$

$$\nabla \cdot \vec{B} = 0 \quad (1.2)$$

$$\nabla \times \vec{E} = -\frac{\partial \vec{B}}{\partial t} \quad (1.3) \quad \nabla \times \vec{B} = -\mu_0 \epsilon_0 \frac{\partial \vec{E}}{\partial t} \quad (1.4)$$

where μ_0 and ϵ_0 are the permeability and permittivity of free space, which describes the electric and magnetic properties of vacuum.

Equation 1.1 describes Gauss' Law for electric fields. Equation 1.2 describes Gauss' Law for magnetism. These equations describe the outward flux of these fields. In the absence of free electric charge, the divergence of equation 1.1 is zero. The divergence of equation 1.2 is always zero, given that there is no equivalent free magnetic charge in nature.

Equation 1.3 describes the Maxwell-Faraday equation, describing how the spatial derivative of the electric field will be accompanied by a time varying magnetic field. Equation 1.4 describes the Maxwell-Ampere Law, showing that the reverse of equation 1.3 is also true, the spatial derivative of the magnetic field will be accompanied by a time varying electric field.

From Maxwell's equations, it is possible to derive the wave equation that governs the speed of light, c , to do so we apply a second curl to equations 1.3 and 1.4.

$$\nabla \times (\nabla \times \vec{E}) = -\nabla \times \frac{\partial \vec{B}}{\partial t} \quad (1.5)$$

$$\nabla \times (\nabla \times \vec{B}) = -\nabla \times \mu_0 \epsilon_0 \frac{\partial \vec{E}}{\partial t} \quad (1.6)$$

We note that $\nabla \times (\nabla \times \vec{A}) = \nabla(\nabla \cdot \vec{A}) - \nabla^2 \vec{A}$, which is known as the curl of the curl identity and using this we obtain the wave equation in terms of both electric and magnetic fields. In addition the curl of B is equal to the time derivative of E, and vice versa through equations 1.3 and 1.4.

$$\mu_0 \epsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} - \nabla^2 \vec{E} = 0 \quad (1.7)$$

$$\mu_0 \epsilon_0 \frac{\partial^2 \vec{B}}{\partial t^2} - \nabla^2 \vec{B} = 0 \quad (1.8)$$

To accurately define the electric and magnetic fields, which are both transverse fields that are perpendicular to each other, we must define those fields as a function of frequency of the wave, ω , wavevector, $\vec{k}$ and the maximum amplitude of the electric/magnetic wave, E_0/B_0 . The E, B fields are perpendicular to each other as a result of equation 1.3 and 1.4, cross products produce vectors that are perpendicular to original vectors, in this case the curl is a cross product between the spatial derivative of the field and the field itself. We note that the spatial derivative can be replaced with the wavevector of the light, which highlights how the direction of propagation, and the E and B fields are all perpendicular to each other, which is commonly described via the Poynting Vector. The equations to define these time varying fields are presented below, these two fields are perpendicular to one another for transverse waves. We only consider the 1D case with scalar fields for simplicity.

$$E = E_0 e^{i(kx - \omega t)} \quad (1.9)$$

$$B = B_0 e^{i(kx - \omega t)} \quad (1.10)$$

Substituting equations 1.9 and 1.10 into equation 1.7 and 1.8 respectively, and simplifying these equations results in the singular wave equation for both electric and magnetic waves.

$$\mu_0 \epsilon_0 \omega^2 - k^2 = 0 \quad (1.11)$$

We note that $\mu_0 \epsilon_0$ has the units of seconds per meter squared, $s^2 m^{-2}$, regardless of whether we are dealing with electric or magnetic fields. Isolating equation 1.11 for the inverse of $\mu_0 \epsilon_0$ will give us the speed of light, c , in terms of these fundamental constants, which is seen below in equation 1.12.

$$c = \frac{1}{\sqrt{(\mu_0 \epsilon_0)}} = \frac{\omega}{k} \approx 3 \times 10^8 \text{ m s}^{-1} \quad (1.12)$$

When traveling through a medium that is not vacuum, the values of ϵ and μ deviate from their free space values, in this case it is typical to describe the relative permittivity, ϵ_r , and permeability, μ_r of the medium.

$$\epsilon_r = \frac{\epsilon_{\text{medium}}}{\epsilon_0} \quad (1.13)$$

$$\mu_r = \frac{\mu_{\text{medium}}}{\mu_0} \quad (1.14)$$

Inserting equations 1.13 and 1.14 into equation 1.11, we can now describe the speed of light in a medium, be it vacuum or a dense medium.

$$v = \frac{c}{\sqrt{\epsilon_r \mu_r}} \approx \frac{3 \times 10^8 \text{ m s}^{-1}}{\sqrt{\epsilon_r \mu_r}} \quad (1.15)$$

It is common to combine the relative permeability and permeability as a single dimensionless constant, n , the refractive index of the medium.

$$n = \sqrt{\epsilon_r \mu_r} \quad (1.16)$$

For a vacuum, $n = \epsilon_r = \mu_r = 1$ and $v = c \approx 3 \times 10^8 \text{ m s}^{-1}$, however dielectric materials such as SiO_2 , the refractive index can vary with frequency, for example consider 150 THz (2000 nm), the refractive index is 1.44, but at 750 THz or 400 nm the refractive index increases to 1.47 [9]. We explore the nature of dispersion in more detail in the following chapters.

As seen in Figure 1.1, the frequencies of electromagnetic waves available to work with are incredibly broad, from radio-waves to visible light and our work is focused at the near-infra red to visible frequencies. In particular, we are interested in frequencies between 150-750 THz or 400-2000 nm, and at these frequencies we find that the magnetic permeability, μ_r is unity on the basis of theory [1, 2] and experiment [4, 5].

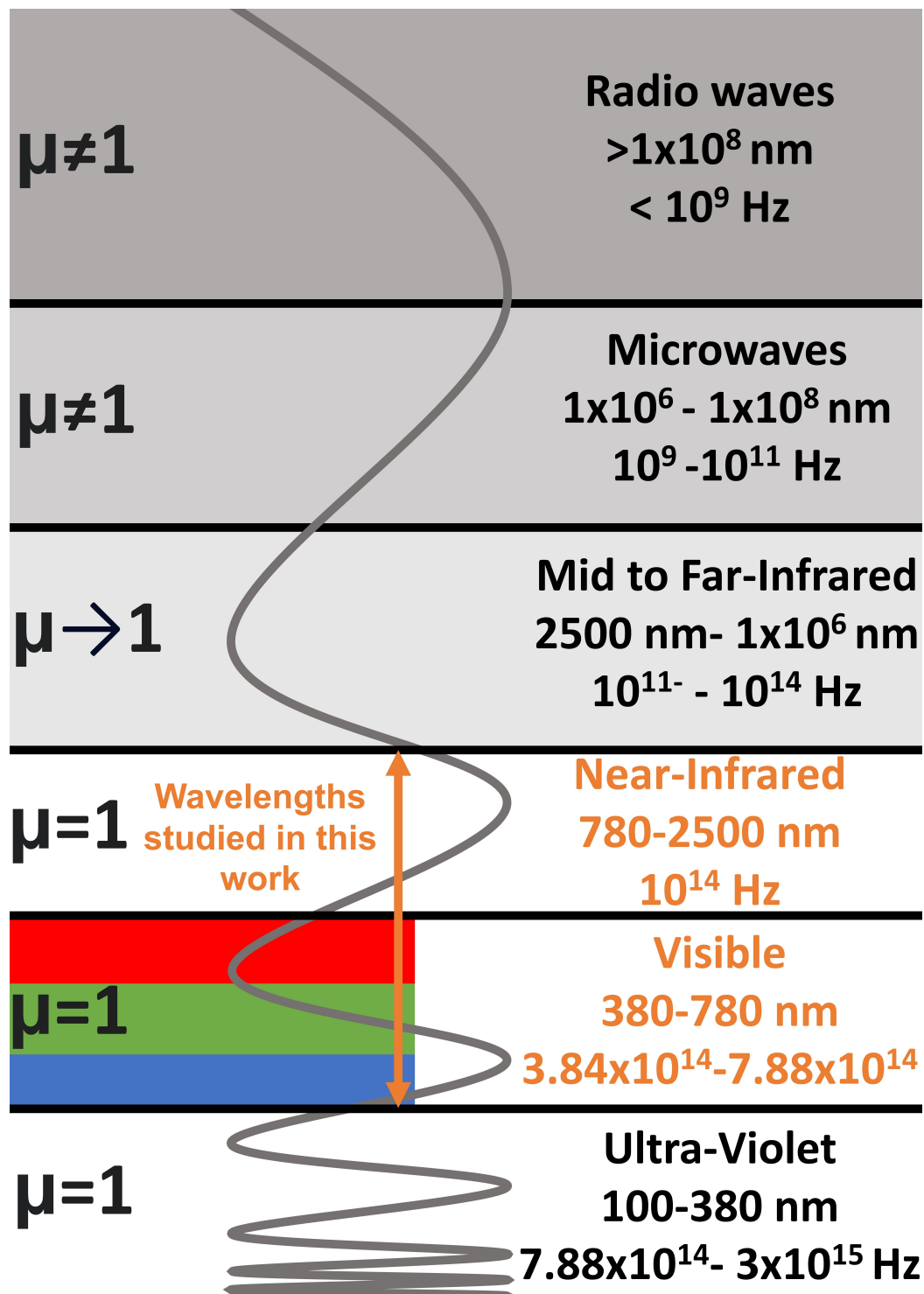


FIGURE 1.1: Schematic of frequencies and wavelengths from radio to ultraviolet waves. Orange marks the range of wavelength studied in this thesis.

1.3 Dispersion of Magnetic Permeability

In the electrostatic/magnetostatic regime, magnetism can be understood in terms of spin angular momentum or the angular orbital momentum of valence electrons [10]. In the case of spin angular momentum, this arises from the unpaired electrons in the valence orbitals (s, p, f etc) of the atoms, which are rotating around their center of mass. This magnetic dipole moment is described below.

$$m = \frac{e}{m_e} s \quad (1.17)$$

S is the intrinsic spin angular momentum, which can be $\pm\frac{1}{2}$. e is the charge of an electron and m_e is the mass of the electron.

For angular orbital momentum, a magnetic dipole moment can arise from the orbiting of electrons around the nucleus.

$$m = \frac{e}{2} \vec{r} \times \vec{v} \quad (1.18)$$

where r is the radius of the angular motion, and v is the velocity of the angular motion.

As noted by Coey [10], there is a rather interesting conflict between this quantum mechanical case and the classical charged particle case. “The remarkable difference between an electron in a quantum-mechanical stationary state and

a classical charged particle is that the former can circulate indefinitely in its orbit as some sort of perpetual motion or electronic supercurrent – whereas the classical particle, or an electron in an unquantized orbit, must radiate energy on account of its continuous centripetal acceleration. Classical orbital motion will soon cease as a result of radiation loss”.

At non-zero frequencies, one must consider the frequency dispersion of the magnetic permeability. Figure 1.1, highlights how the magnetic permeability of a medium is not unity at frequencies from radio-waves to microwaves, and trends to unity from the mid- to far-infrared ranges. This phenomena was first described by Landau-Lifshitz [1] in 1961, and expanded on by Merlin [2] in 2009.

Landau and Lifshitz began with the definition of the magnetic dipole moment of a medium, which is a common convention to describe the magnetic response of a medium, assuming the wavelength is significantly larger than the atomic dimensions of that medium.

$$\vec{m} = \int \vec{r} \times \vec{j} dV \quad (1.19)$$

where V is the volume over which the magnetic dipole moment is integrated over, $\vec{r}$, is the positional vector associated with $\vec{j}$. $\vec{j}$ is current density associated with both magnetic and electric fields as described by Maxwells equations, described as below.

$$\vec{J} = c \nabla \times \vec{M} + \frac{d\vec{P}}{dt} \quad (1.20)$$

where $\vec{M}$ is the magnetization field, $\vec{M} = \vec{B} - \vec{H}/\mu_0$ and $\vec{P}$ is the electric polarization field, $\vec{P} = \vec{D} - \epsilon_0 \vec{E}$. These fields are non-zero only if there is a non-unity value for the permeability or permittivity respectively, and assume purely magnetic/electric dipole response associated with wavelengths much greater than atomic dimensions of the medium.

Hence a magnetic dipole moment, arising from a circulating current density, has two sources

$$\vec{m} = \int \vec{M} dV + \int \vec{r} \times \frac{d\vec{P}}{dt} dV \quad (1.21)$$

As the frequency of light trends to higher values, the electric polarization plays an increasing role in the overall current density and it is difficult to separate electric and magnetic contributions, hence $\mu = 1$.

The ideal condition to obtaining $\mu \neq 1$ is where the object is as small as possible, which will enhance the contribution of the curl, a spatial derivative, over the time dependent derivative of the electric polarization. Landau identifies an inequality to identify this ideal regime.

$$l^2 \ll \frac{(\mu_r - 1)c^2}{\omega^2} \quad (1.22)$$

where l is the size of the object, for a magnetic permeability to be different from one, this inequality must hold, and Landau makes the following observation from inequality 1.20.

"Thus there is no meaning in using the magnetic susceptibility from optical frequencies onward, and in discussing such phenomena we must put $\mu=1$ ".

However Landau critiques this observation when considering a wavevector dependence on the permittivity, a feature common to single crystal media.

"The above treatment gives a somewhat different view of the assertion in (1.20) that the permeability μ has no meaning at optical frequencies. In that range, the effects due to the difference of μ from 1 are in general indistinguishable from those of the spatial dispersion of the permittivity".

Further discussion on spatial dispersion will be expanded in the next chapter, but we wish to highlight that the current density defined in equation 1.20 is not unique, given that $\vec{E}$ and $\vec{B}$ can not be considered independent of each other, which is highlighted by Maxwells equations, equations 1.3 and 1.4 in particular. This results in inequality 1.20 being insufficient to cover all possible regimes.

To our knowledge measurements of magnetic permeability have not been attempted at optical frequencies, the earliest attempt at measuring the dispersion of the magnetic permeability date back to the 1950's and are reviewed by Allanson [4] in excellent detail as shown in Figure 1.2 (a), who discussed the measured magnetic permeability of wires of various materials and diameters, from frequencies between $10^6 - 10^{10}$ Hz or c/s (cycles per second), which is

firmly in the microwave range in Figure 1.1.

The single constant observation throughout the datasets presented in Figure 1.2 (a) is that the magnetic permeability decreases as one goes further into the microwave range, as predicted by inequality 1.20. It appears that changing the dimensions of these wire compensates for this disappearance of magnetic permeability, which suggests that the size dependence of the inequality 1.20 is qualitatively verified.

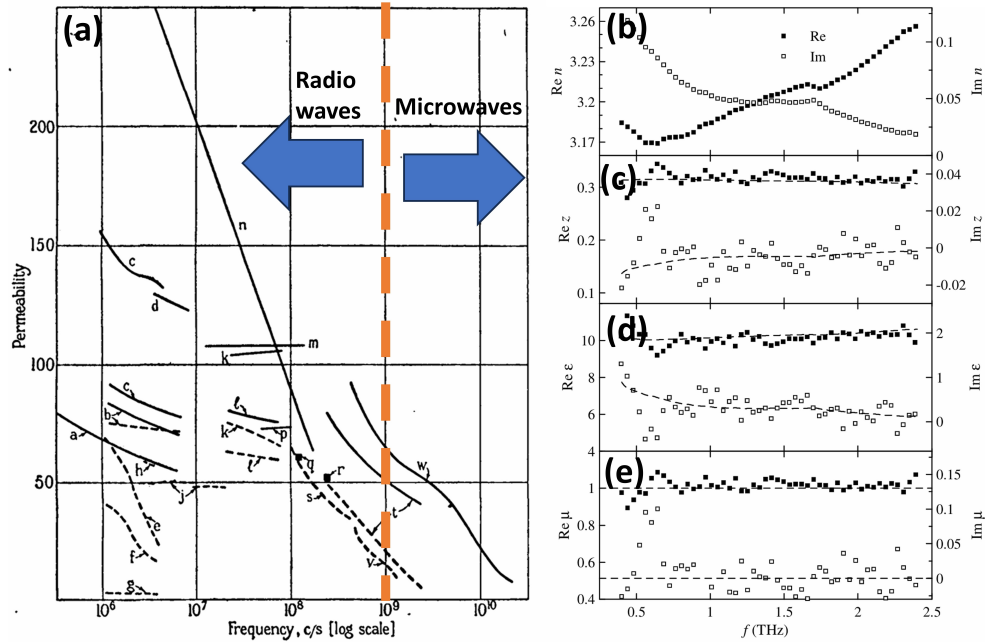


FIGURE 1.2: (a) Plot of measured dispersion of magnetic permeability. Orange line and blue arrows are our additions to this graph to make clear the radio and micro wave regions. c/s stands for cycles per second, presented along the x-axis is equivalent to Hz. Graphs sourced from [4]. (b)-(e) Measurements of ZnTe refractive index, permittivity and permeability. (b) Refractive index. (b) Impedance, which refers to the wave impedance of the medium, which is defined as the square root ration of the permeability and permittivity. (c) Permittivity. (d) Permeability. Dashed lines represent the assumption that $\mu=1$. Graph sourced from [5].

However the periodic and subwavelength nature of the samples suggests that

this paper may have been one of the first papers on metamaterials, and hence may not be a real indication of bulk material parameters that Landau discussed.

For a discussion on bulk material parameters, H.Němec et al [5] measured the electric permittivity and magnetic permeability for ZnTe up to a frequency of 2.5 THz or a wavelength of 120,000 nm, which is in the far-infra red range shown in Figure 1.1. This is the highest frequency at which such a measurement has been made as of writing this thesis. It is clear from Figure 1.2 (e) that the the measured magnetic permeability is unity, and the authors state that it was not possible to separate the magnetic permeability from permittivity, making it valid to set the permeability to 1, which corresponds to the dashed line in Figure 1.2. This agrees with Landau's argument which we outlined earlier.

1.4 Demonstration and Application of Magnetic Permeabilities in Literature

The first question to ask is whether is it possible to observe any magnetic response at optical frequencies? The answer is within inequality 1.20, a magnetic response may be observable if the structure is smaller than the wavelength of visible light, which would be a size limit between 400-780 nm. Ideally the smaller the structure relative to the incident wavelength, the easier it is to obtain a non-unity value for the permeability. Pendry [6], was the first to postulate a practical realization of this by exploring the effective permeability arising from metamaterials.

The following question is to ask is why would we desire non-unity value of magnetic permeability at high frequencies? An experiment by Shelby et al [11] demonstrated an exciting phenomenon associated with a medium with both negative permittivity and permeability, negative refraction, which is the bending of light in the "incorrect" direction after transmitting across a surface. This phenomena first postulated by Veselago [12], however no real life example existed to support negative refraction at the time.

The work done by Pendry [6] and Shelby et al [11] has been reviewed by Smith et al [7], and we show this in Figure 1.3. A specifically engineered metamaterial can be tailored for negative permeability and permittivity, resulting in a negative refraction flat lens. Such a lens has been proposed to be a "perfect lens" [13], but in reality it's resolution is limited by absorption rather than diffraction, which is still an improvement over a traditional diffraction limited lens.

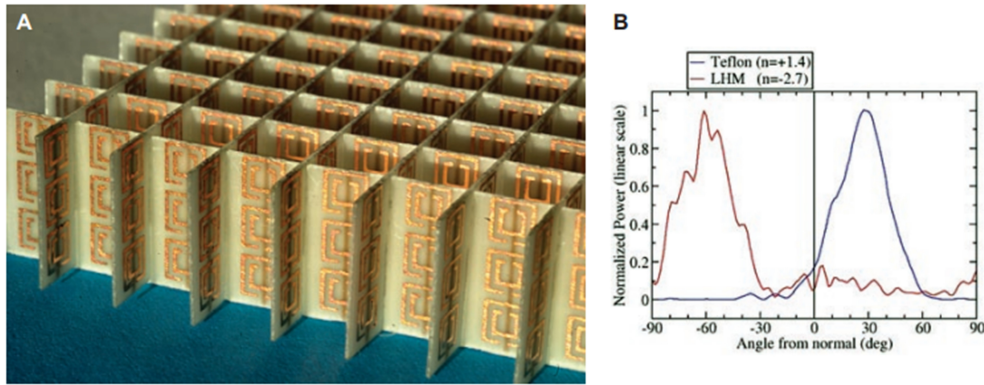


Fig. 3. (A) A negative index metamaterial formed by SRRs and wires deposited on opposite sides lithographically on standard circuit board. The height of the structure is 1 cm. (B) The power detected as a function of angle in a Snell's law experiment performed on a Teflon sample (blue curve) and a negative index sample (red curve).

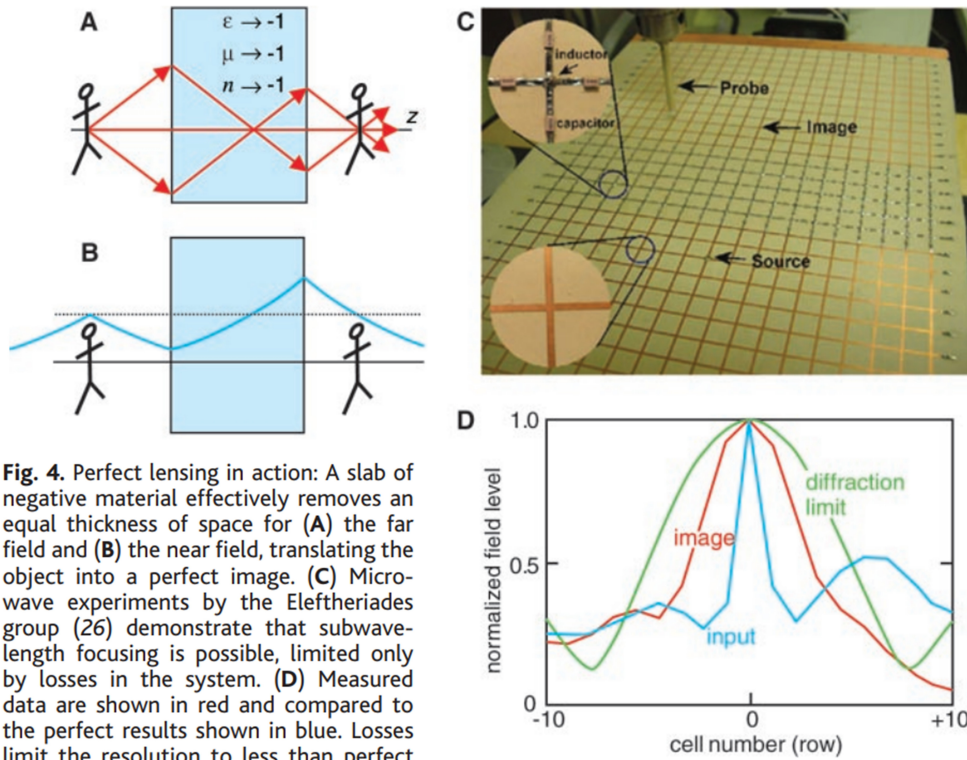
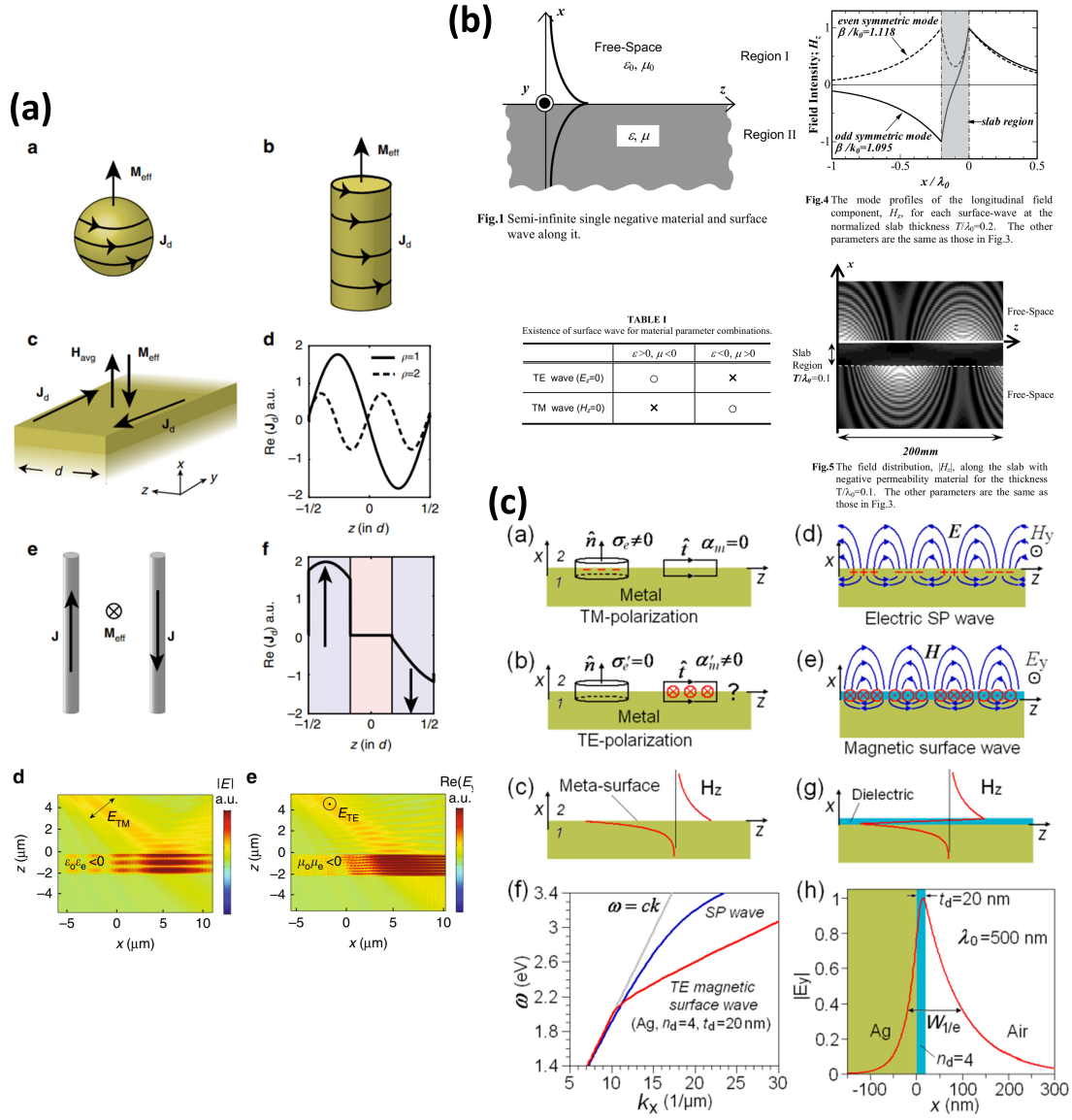


Fig. 4. Perfect lensing in action: A slab of negative material effectively removes an equal thickness of space for (A) the far field and (B) the near field, translating the object into a perfect image. (C) Microwave experiments by the Eleftheriades group (26) demonstrate that subwavelength focusing is possible, limited only by losses in the system. (D) Measured data are shown in red and compared to the perfect results shown in blue. Losses limit the resolution to less than perfect but better than the diffraction limit shown in green.

FIGURE 1.3: Practical demonstration of negative refraction. Top 2 figures show a metamaterial, which is an array of subwavelength structures that produce an "optical magnetic" response, which results in negative refraction. Bottom 3 figures further demonstrated this negative refraction, showing that resolution of a negative refraction lens is better than the diffraction limit. The frequencies studied were from $8 \times 10^9 - 1.2 \times 10^{10}$ Hz or roughly 3×10^7 nm. Graphs sourced from [7].

Another particularly interesting application of non-unity magnetic permeabilities is the excitation of unusual surface waves. At the interface between two media with differing electric permittivity, a transverse magnetic (TM) surface plasmon may exist, for differing magnetic permeabilities, a transverse electric (TE) surface plasmon may exist.

As shown in Figure 1.4, there have been several theoretical investigations into these TE plasmons. In sub-figure (b) Hotta et al [14] showed the existence surface waves in materials that had magnetic permeabilities. In sub-figure (c) Sun et al [15] highlighted that by placing a single thin film of high refractive index onto of a Ag substrate, a TE surface plasmon may exist at ultra-violet frequencies. A more practical demonstration of this TE surface wave was achieved by Papadakis et al [16] in Figure 1.4 (a), who experimentally demonstrated that planar metamaterials made of a series of subwavelength thin films were capable of supporting a TE surface plasmon. They argued that specifically shaped current densities, $\vec{J}$, were responsible for an "optical magnetic" response. A variety of structures, including thin films, can support anti-parallel current densities, leading to an effective magnetization, and hence an "optical magnetic" response.



To expand on the possibility of thin films providing an "optical magnetic" response, Li et al [17] in Figure 1.5, demonstrated that in order to correctly describe the reflection of a subwavelength thin film of GaP, one must include the magnetic dipole / quadrupole contribution to the reflection spectrum. These magnetic multipoles arise from circulating current densities, the same as those identified by Papadakis et al [16], Pendry [6, 18] and discussed in reviews on optical magnetism [8] as giving rise to an "optical magnetic" response, highlighting that such "optical magnetic" responses may not be unique to metamaterials, and may be intrinsic to all subwavelength structures, including simple thin films.

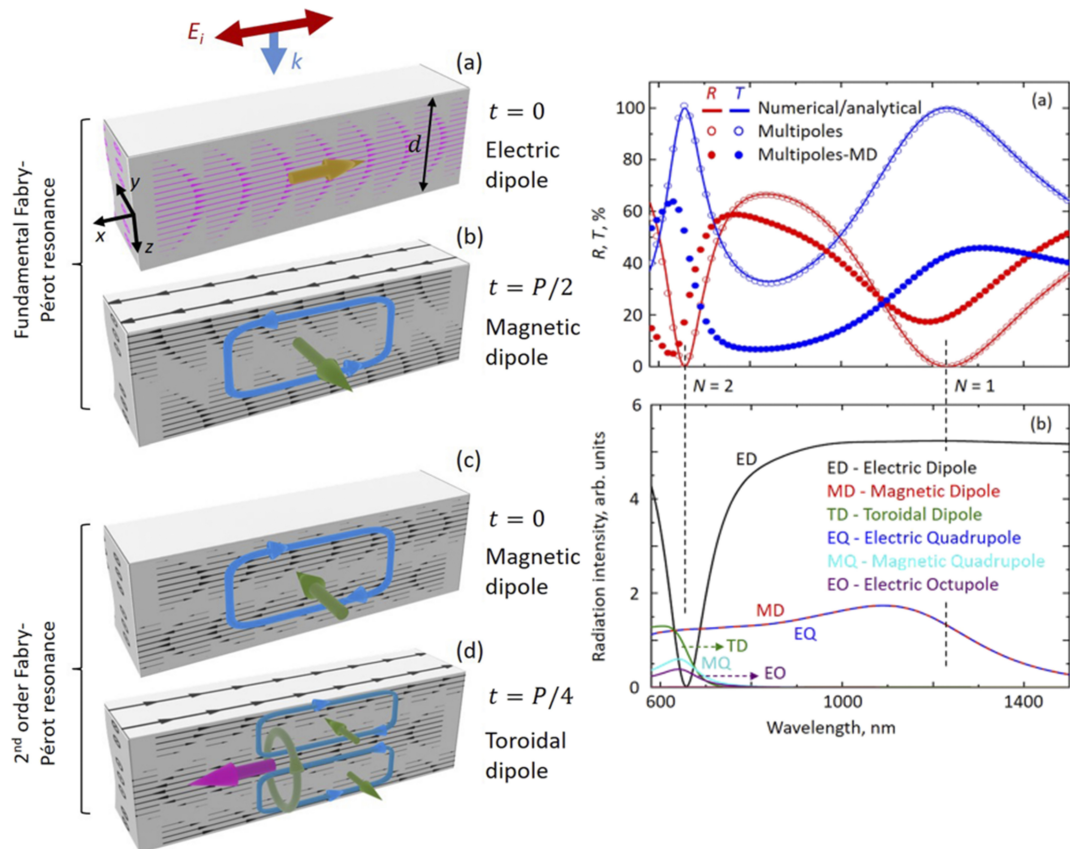


FIGURE 1.5: Demonstration of an "optical magnetic" response in subwavelength GaP thin films, graphs sourced from [17].

1.5 Thesis Structure and Aims

Achieving a magnetic response at visible frequencies is a highly sought phenomenon, permitting the design of new devices using unusual physics traditionally confined to radio and microwave frequencies. Devices such as flat lenses relying on negative refraction [11] or polarizers using s-polarized Brewster angles rather than p-polarized Brewster angles, [19, 20, 21] could be realized if one could practically realize magnetic responses at optical frequencies. Current approaches to achieving this magnetic response require complex sub-wavelength structures that are not scalable and are expensive to fabricate.

We aim to explore the role of magnetism in dielectric thin films, highlighting that a magnetic response can not be ignored in these simple structures and how such a magnetic response may be engineered to observe a TE surface plasmon in a simple metal-dielectric system. This thesis is structured as follows.

Chapter 1. Introduce high frequency magnetism and the difficulties of observing a magnetic response at optical frequencies to give context to the work of this thesis.

Chapter 2. Serves as a guide to teach readers on the nature of dispersion in bulk media and thin films, which is a topic vital to understanding the work and literature in Chapters 1, 4 and 5.

Chapter 3. Discusses methods in building and designing apparatus used for measurements in Chapter 4 and 5, including our novel design of the "Dual

Kretschmann Configuration" for prism coupling measurements using in Chapter 5, which offers accurate angle and frequency resolved measurements that are easier to perform than traditional approaches. Methods to model these experiments are outlined as well, including modeling of reflection via S-parameter, transfer matrix and multipole decomposition methods and eigenmode analysis.

Chapter 4. Use multipole decomposition to understand the angle and frequency dependent behavior of optical magnetism in free-standing films of Si_3N_4 thin films, highlighting how Fabry Perot modes and the Brewster angle are the result of combined electric and magnetic responses rather than purely electric responses. Then demonstrate the role of spatial dispersion in observing magnetic responses, and how electric poles may contribute to magnetic responses.

Chapter 5. Demonstrate how these magnetic responses in thin films may be applied to engineer the existence of a TE surface plasmon with simple dielectric-metal structures.

Chapter 2

Dispersion in Bulk Media and Thin Films

2.1 Dispersion of Bulk Media

Having given an overview of the problem of obtaining non-unity magnetic permeability at optical frequencies in Chapter 1, we now move to explain the concepts behind frequency and spatial dispersion, as well as the ambiguity between magnetic permeability and spatial dispersion. Examples of frequency dispersion will be vital, as these media will be those studied in future chapters.

We then discuss dispersion arising from thin films, including Fabry Perot modes and TM/TE surface plasmons that are also the focus of study of this thesis.

2.1.1 Frequency Dispersion, $\epsilon(\omega)$

In the classical microscopic picture, where the wavelength is much larger than the atomic dimensions, materials can be thought of as dipole oscillators, where the electrons are driven by the electric field of the incident light. In dielectrics, these are bound electrons in the valence bands. In this case these dipoles are atomic electric dipoles, which are polarized by the incident light, by polarizing these atoms, a restoring force is generated from the uneven distribution of charge. This can be treated as a single Lorentzian oscillator that follows the following equation of motion[\[22\]](#).

$$m_e \frac{d^2x}{dt^2} + m_e \gamma \frac{dx}{dt} + m_e \omega_0^2 x = eE \quad (2.1)$$

Where m_e is the mass of the electron, γ is the damping/restoring force, ω_0 is the frequency, e is the charge of the particle and E is the external driving force which is the electric field of the incident light.

The maximum displacement of these electrons can be derived by assuming the displacement oscillates in time as seen below.

$$x(t) = x_0 \text{Re}(e^{-i\omega t + \Phi'}) \quad (2.2)$$

The maximum displacement according to this Lorentzian dipole oscillator is as follows.

$$x_0 = \frac{-eE/m_e}{\omega_0^2 - \omega^2 - i\gamma\omega} \quad (2.3)$$

As described by Fox, the resonant polarization field in terms of the electric dipole moment can be described in terms of the number of particles N and the electric dipoles moment, which itself is the product of maximum displacement, N , and the charge of the electron, e .

$$\vec{P}_{resonant} = N\vec{p} = -Nex_0 = \frac{Ne^2}{m_0} \frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} \vec{E} \quad (2.4)$$

$\vec{P}_{resonant}$ contains the resonant behavior near the resonant frequency of our

atomic oscillator and the background behavior far away from the resonant frequency, which can be described via the electric susceptibility, χ .

$$\vec{D} = \epsilon_0 \vec{E} + \vec{P} = \epsilon_0 \vec{E} + \vec{P}_{background} + \vec{P}_{resonant} = \epsilon_0 \vec{E} + \epsilon_0 \vec{E} \chi + \vec{P}_{resonant} \quad (2.5)$$

Where $\vec{D}$ is the electric displacement field. these equations can be re-organized to isolate for the frequency dispersion of a dielectric as seen below.

$$\epsilon_r = 1 + \chi + \frac{Ne^2}{m_0} \frac{1}{\omega_0^2 - \omega^2 - i\gamma\omega} \vec{E} \quad (2.6)$$

In theory, it may be possible to ignore this frequency dispersion if one is sufficiently far away from the resonant frequency and only consider the effect of the background polarization.

We note that in this case, the material lacks spatial dispersion/anisotropy as the permittivity is a scalar value, a spatially dispersive material requires a diagonal 2nd order tensor.

As described in the previous chapter, the relation between the frequency and wavevector of a wave can be described as follows, in terms of the relative permittivity, assuming no magnetic response, $\mu_r = 1$.

$$\epsilon_r \omega^2 = k^2 \quad (2.7)$$

A medium is said to be dispersive if ϵ_r is dependent on either frequency or wavevector, and is typically a complex number if dispersive. Many materials exhibit frequency dispersion, where the permittivity varies with frequency, that is $\epsilon_r(\omega) = e_{r,real}(\omega) + ie_{r,imag}(\omega)$, which arises from the well-known Kramers Kroneig relation [23]. Several examples of measured frequency dispersion are presented below in Figure 2.1.

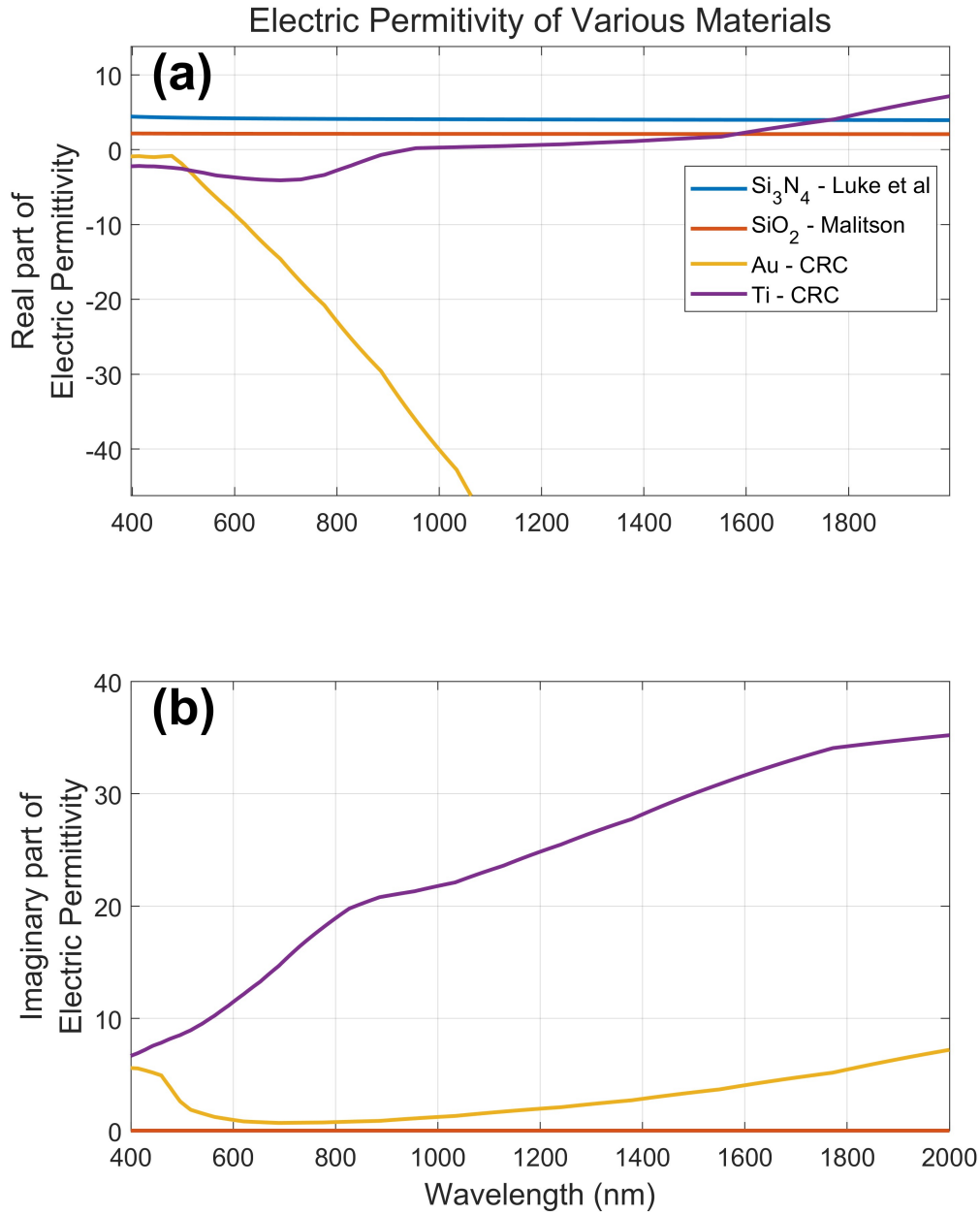


FIGURE 2.1: Examples of frequency dispersion from literature. In all these cases, $\mu_r = 1$. (a) Real part of the permittivity. (b) Imaginary part of the permittivity which arises from absorption of light. These particular materials are key materials that were studied in this thesis. Si_3N_4 data was sourced from [24], SiO_2 data was sourced from [9], Au and Ti data was sourced from [25] and [26] respectively.

Low loss materials such as SiO_2 and Si_3N_4 are weakly dispersive, as they are weakly absorbing across the visible and near infra-red wavelengths, highlighted by the Kramers-Kronig relation [23]. Lossy materials, those with large imaginary parts, $\epsilon_{r,imag}$, show large changes in the real part of the dispersion, making them highly dispersive, this includes metals such as Au and Ti in Figure 2.1.

Another method to identify frequency dispersion than in Figures 2.1 is by plotting the dispersion relation of the permittivity as defined by equation 1.11. Linear trends are dispersionless, while any degree of dispersion will appear as a non-linear trend. Figure 2.2 plots this dispersion relation for each of these materials, $\omega = \sqrt{\epsilon_r}k$, for the same media presented in Figure 2.1. None of these media show a linear response, and hence are all dispersive to some degree. SiO_2 and Si_3N_4 may appear to be linear, but that is a result of the small changes in refractive index with frequency, and measurements by Luke et al [24] and Maltison [9] have identified this dispersion.

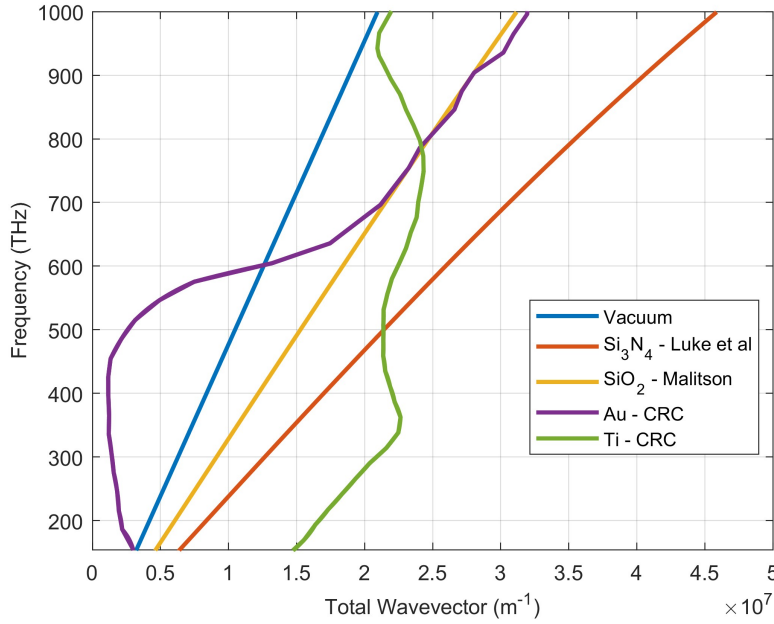


FIGURE 2.2: Frequency dispersion of various materials shown in Figure 2.1. In all these cases, $\mu_r = 1$.

2.1.2 Spatial Dispersion, $\epsilon(\vec{k})$

As well as the frequency dispersion mentioned above, it is possible for the permittivity to show spatial dispersion.

Re-visiting the microscopic image discussed in the previous section, in the case of non-local properties or spatial dispersion, the permittivity becomes a 2nd order tensor, with the permittivity depending on the direction of the electric field relative to the crystal axis of the anisotropic crystal, normally these effects are negligible if the wavelength of the incident light is significantly larger than crystal's lattice constant, however it is still weakly observable in YVO₄ [27].

A good example of spatial dispersion is the Doppler broadening of absorption

lines in a gas, if our atomic oscillators are moving in this gas, then the spectrum is broadened by the product of the wavevector of the light and the speed of the particle, and spatial dispersion is observable in the limit shown, $\vec{k} > \frac{|\omega - \omega_0|}{v_t}$, where v_t is the thermal velocity of the gas atoms.

Another example is in the case of metals, if the mean free path of electrons is larger than the incident wavelength, then spatial dispersion may become significant, however the mean free path of electrons in Au is approximately 50 nm and at optical frequencies[28], and spatial dispersion is not significant at optical frequencies.

Re-visiting the definition of relative permittivity described earlier, we note that wavevector dependency can also affect the permittivity. Given that the wavevector is defined as $k = \frac{\omega}{c}$, it can be difficult to separate the frequency and wavevector dispersion, in order to separate the two, one must measure the angle dependent behavior of the permittivity, which requires a 2nd order tensor to describe the permittivity, this is what we refer to as spatial dispersion [1, 29].

$$\hat{\epsilon}_{ij} = \begin{bmatrix} \epsilon_{xx} & 0 & 0 \\ 0 & \epsilon_{yy} & 0 \\ 0 & 0 & \epsilon_{yy} \end{bmatrix} \quad (2.8)$$

In the case of the simplest form bulk spatial dispersion, a uniaxial crystal, we only consider two different permittivities along the two axis of the crystal structure, the ordinary and extraordinary permittivity, $\epsilon_{ordinary}$ and $\epsilon_{extraordinary}$. An example of these parameters is shown in Figure 2.3 for YVO₄ crystal [27].

We note how frequency dispersion is also present, however depending on the direction of propagation, i.e, the wavevector of the light relative to the crystal axis, the permittivity experienced by the wave changes. We note that the degree of spatial dispersion, which can be defined as the difference in permittivities in Figure 2.3 (b), increases as frequency dispersion becomes non-negligible, further highlighting how difficult it is to separate these two phenomena.

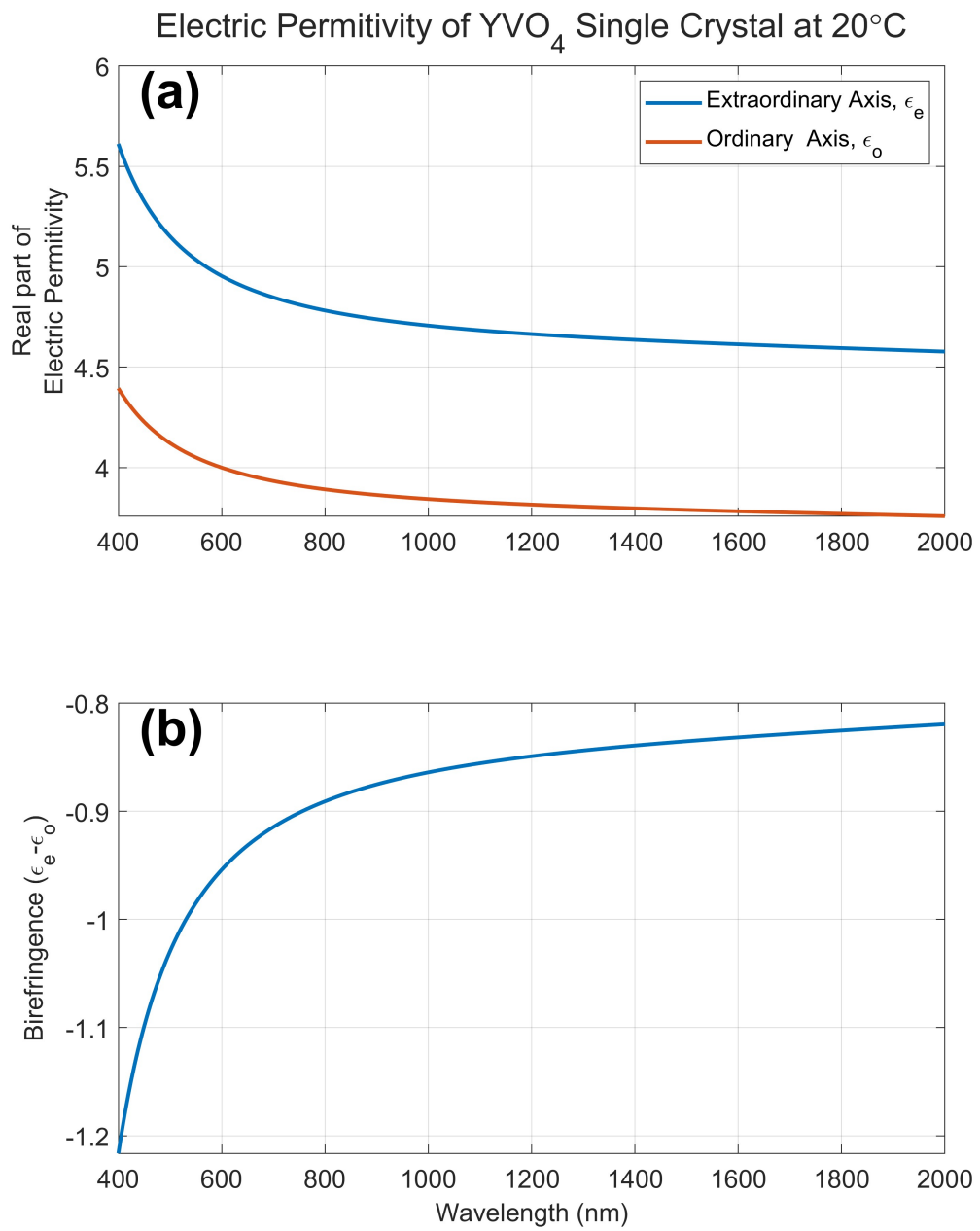


FIGURE 2.3: Frequency and spatial dispersion of YVO_4 crystal at 20°C , $\mu_r = 1$ [27].

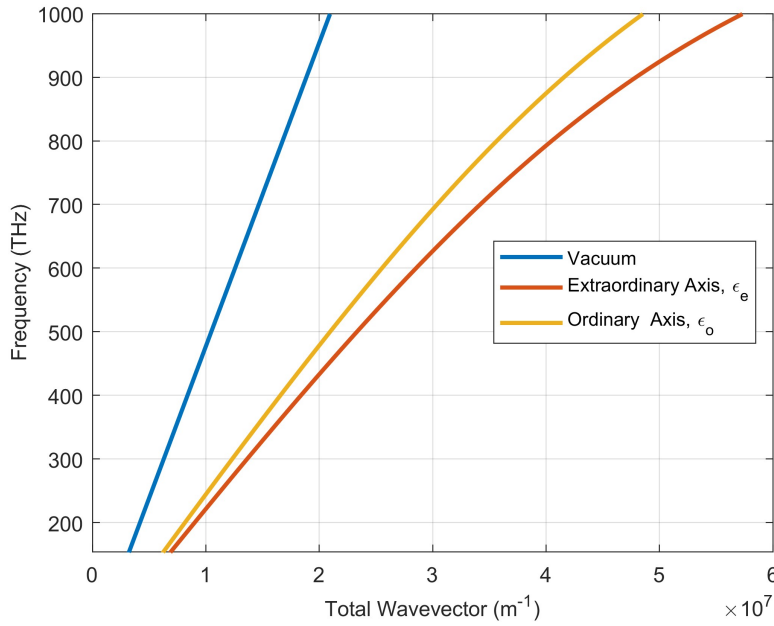


FIGURE 2.4: Frequency and spatial dispersion of YVO₄ crystal at 20° C, $\mu_r = 1$.

In Figure 2.4, we can plot the dispersion relation of the YVO₄ crystal, a major difference compared to Figure 2.2, is that there are now two dispersion relations corresponding to the ordinary and extraordinary axis. Further dispersion lines are possible depending on the orientation of the crystal axis relative to the wavevector of the light, for simplicity only two are presented.

2.1.3 Ambiguity Between $\epsilon(\vec{k})$ and $\mu(\omega)$

Having described both frequency and spatial dispersion, we revisit a comment made by Landau regarding spatial dispersion in the permittivity and frequency dispersion in magnetic permeability.

"...the effects due to the difference of μ from 1 are in general indistinguishable from those of the spatial dispersion of the permittivity".

First we must explain in detail why they are indistinguishable, Dirdal et al provides a more nuanced explanation of this [30]. We first must revisit the definition of the current density, $\vec{J}$ from equation 1.21 and seen below.

$$\vec{J} = c \nabla \times \vec{M} + \frac{d\vec{P}}{dt} \quad (2.9)$$

In the case of eigen-modal propagation, where we assume that the wavenumber is $k = \frac{\epsilon_{\perp}\mu\omega^2}{c^2}$, where $\epsilon_{\perp}$ is the permittivity in the transverse direction, as the frequency increases, the wavevector must approach the vacuum case, where $\epsilon_{\perp}\mu = 1$. This corresponds to a transformation of the field associated with $\epsilon_{\perp}$ and μ , so $\vec{M}$ and $\vec{P}$ becomes $\vec{M}'$ and $\vec{P}'$. It is noted that the current density is unchanged, despite changing the values of the vectors $\vec{M}$ and $\vec{P}$, this gives rise to the following relation below.

$$c \nabla \times \vec{M} + \frac{d\vec{P}}{dt} = c \nabla \times \vec{M}' + \frac{d\vec{P}'}{dt} \quad (2.10)$$

As seen from equation 2.4, and highlighted in the introduction, the current density is not uniquely defined, there are an many combinations of ϵ and μ that can give the same value for the current density, $\vec{J}$.

Dirdal [30] highlights that the Landau-Lifshitz argument, where $\mu = 1$, is a

particular example of this ambiguity where it is convenient to describe a spatially dispersive permittivity only, i.e, $\epsilon(\omega, \vec{k}) \neq 1$ and $\mu = 1$.

Agranovich [3, 31] expands on this issue, by refining Maxwell's equations, by forcing $\vec{B} = \vec{H}$ and only considering $\vec{E}$, $\vec{D}$ and $\vec{H}$ fields. This formulation of Maxwell's equations acts to remove the ambiguity between permittivity and permeability discussed by Dirdal [30] by including electric and magnetic responses into the electric fields.

$$\nabla \cdot \vec{D}' = 0 \quad (2.11) \quad \nabla \cdot \vec{B} = 0 \quad (2.12)$$

$$\nabla \times \vec{E} = -\frac{d\vec{B}}{dt} \quad (2.13) \quad \nabla \times \vec{B} = -\mu_0\epsilon_0\frac{d\vec{D}'}{dt} \quad (2.14)$$

where the new electric displacement field, $\vec{D}'$, is defined in terms of permittivity and permeability.

$$\vec{D}' = \epsilon(\omega)\vec{E} - \frac{c^2}{\omega^2} \left(1 - \frac{1}{\mu(\omega)}\right) \nabla \times \vec{B} \quad (2.15)$$

These equations are equivalent to the traditional Maxwell's equations seen in the introduction, but now equation 2.9 accounts for spatial dispersion, as it a particular case of the more general equation seen below.

$$\vec{D}' = \epsilon_{ij}(\omega, \vec{k})E \quad (2.16)$$

where ϵ_{ij} is an element of the permittivity tensor used to describe anisotropic crystals in the previous section.

In Equations 2.9 and 2.10, spatial dispersion is the result of including a magnetic response into the electric displacement field, but also highlight how specific electrical responses can behave as magnetic responses. For Agranovich's case, he highlighted that electric quadrupoles may behave as magnetic dipoles, and highlighted examples of metamaterials where electric quadrupoles contribute to a negative permeability [32], this will prove an important consideration in future chapters, when we discuss how the electric quadrupole may polarize a medium.

Setting $\vec{B} = \vec{H}$ has important implications for the current density, $\vec{J}$, and inequality 1.20 derived from it. We only deal with the electric polarization and do not separate the current density into electric and magnetic fields, making the derivation of inequality 1.20 impossible and hence it is not possible to define an ideal object size where a magnetic response is easy to observe. But this does not exclude the possibility of observing a magnetic response.

2.2 Dispersion of Thin Films

Having explored the nature of dispersion in bulk media, both frequency and spatial dispersion in permittivity and the ambiguity between spatial dispersion in permittivity and frequency dispersion in permeability, we move onto the dispersion of thin films.

When considering thin films, and resonances unique to those structures, such as Fabry-Perot and surface plasmons, these resonances require us to account for the dispersion of the bulk medium and the structure that the medium takes. We must consider the role of thickness on the dispersion of thin film modes, which has been highlighted as a form of spatial dispersion [33], but is not a common convention in literature.

2.2.1 Fabry Perot Modes

A well known form of resonance in thin films is the Fabry Perot mode, arising from the constructive interference between reflected and transmitted waves in a thin film, leading to a resonate enhancement of light in the cavity at a specific wavevector.

Fabry Perot modes are solutions to the wave equation 1.7 with the assumption that the boundary conditions, at co-ordinates 0 and d are defined as $E(0, t) = E(d, t) = 0$, which is made when working with a lossless cavity as it is assumed that the electric fields at the boundaries of the cavity are zero, where d is the thickness of the Fabry Perot cavity [34].

For a standing wave confined to this cavity, the electric field is defined in terms of it's spatial and time dependence as

$$E(x, t) = E_0 \sin(k_q x) \quad (2.17)$$

k_q is the wavenumber of the q th order Fabry Perot mode, where q is a positive

non-zero integer. Equation 2.11 is zero at the boundaries of the cavity, and may satisfy the wave equation above if the following is true

$$k_q d = \pi q \quad (2.18)$$

This confines k_q to discrete values, so the wavevector of each mode is the following.

$$k_q = \frac{\pi q}{d} \quad (2.19)$$

If the wavevector is confined to distinct values, then the frequency is as well. c is the speed of light.

$$\omega_q = \frac{q c}{2d} \quad (2.20)$$

or in terms of wavelength

$$\lambda_q = \frac{2n(\omega, \vec{k})d}{q} \quad (2.21)$$

The above equations defines the wavelengths at which a Fabry Perot mode of q th order may exist at, but suggests no angle dependence in these modes. The solution to introduce angle dependence into the equation is to change the thickness, d , to the optical path length in the thin film, d_{OP} .

$$\lambda_q = \frac{2n(\omega, \vec{k})d_{OP}}{q} \quad (2.22)$$

At normal incidence, this matches the thickness of the film but will vary with angle. The optical path length will depend on the angle of transmission in the thin film, which depends on Snell's law.

$$n_i \sin(\theta_{incident}) = n_t \sin(\theta_{transmitted}) \quad (2.23)$$

This can be re-arranged as follows.

$$\theta_{transmitted} = \sin \left(\frac{n_i \sin(\theta_{incident})}{n_t} \right)^{-1} \quad (2.24)$$

Now the optical path length can be defined in terms of the thickness and angle of transmission as follows.

$$d_{OP} = d \cos(\theta_{transmitted}) = d \cos \left(\sin \left(\frac{n_i \sin(\theta_{incident})}{n_t} \right)^{-1} \right) \quad (2.25)$$

Substituting this into equation 2.18 we obtain

$$\lambda_q = \frac{2nd \cos \left(\sin \left(\frac{n_i \sin(\theta_{incident})}{n_t} \right) \right)^{-1}}{q} \quad (2.26)$$

We test the validity of this equation by plotting these resonant wavelengths

and angles over the reflection spectra of a thin film, with a thickness of 500 nm and refractive index of 2, surrounded by air. As seen in Figure 2.5 below, the equation perfectly predicts the resonant wavelength and angle dependence of the first 3 Fabry Perot modes, as it overlaps with regions of zero reflection, which corresponds to perfect transmission, a feature that arises from the constructive interference of transmitted and reflected waves seen in Fabry Perot modes.

We briefly note the similarity between equation 2.20 and the condition required to observe the Bragg angle in crystal/spatially dispersive media [35]. The Bragg angle is controlled by the spacing of atomic layers and the angle of incidence for a given wavelength. The Fabry Perot mode is the spacing of the boundaries of the thin film and angle of incidence for a given wavelength. If our Fabry Perot resonator were made of two reflecting media separated by vacuum, these two equations would be even more similar, the only difference being a cosine dependence for the Fabry Perot modes and sine dependence for the Bragg angle. This highlights the potentially spatially dispersive nature of thin films.

In the case of lossy films, additional considerations are required, in particular sine functions in Snell's law must be rewritten with Euler identities. $\sin(x) = \frac{e^{ix} - e^{-ix}}{2i}$.

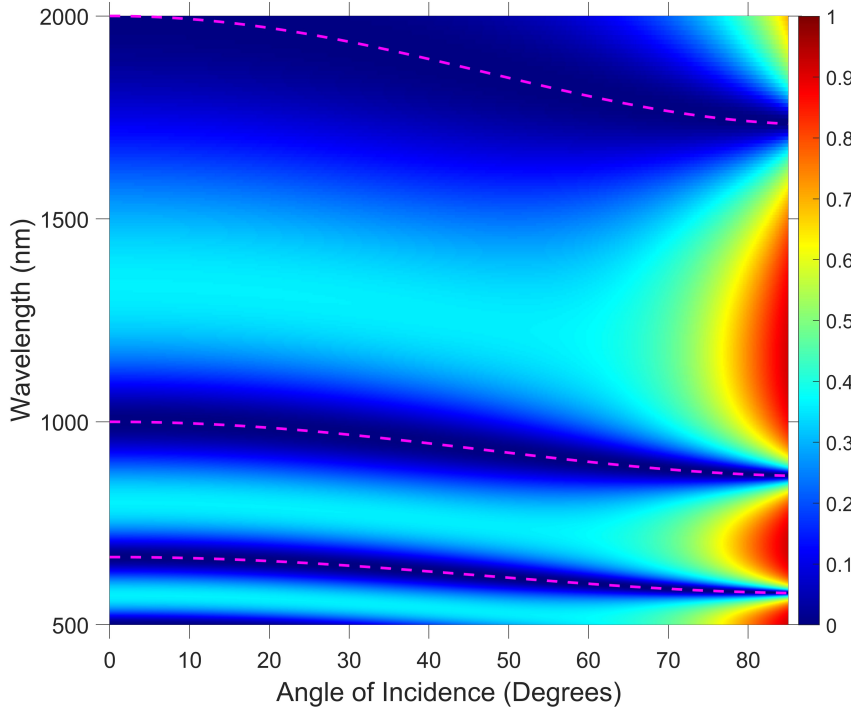


FIGURE 2.5: Plot of modeled unpolarized reflection (in the range 0 to 1) for 500 nm of medium with refractive index $n=2$ surrounded by a medium with $n=1$ showing the first 3 Fabry Perot modes, ($q=1,2$ and 3) in as dashed magenta lines overlapping with regions of zero reflection, which is expected from the reflection of a Fabry Perot mode. Unpolarized reflection is defined as the average of s-/p-polarization reflection, which is a form of linearly polarized light with fields that are parallel/perpendicular to the surface of the thin film respectively.

2.2.2 Propagating Surface Plasmons

Propagating surface plasmons are traveling surface waves confined to the interface between a dielectric material and metal, and can propagate along this interface. They are primarily transverse magnetic (TM) modes, that is they have two electric field components, and 1 magnetic field component, with electric fields along the direction of propagation (longitudinal), and perpendicular (transverse) to it, while the magnetic field is purely transverse.

Even more unusual is the transverse electric (TE) surface plasmon, a surface wave with two magnetic fields and 1 electric field, with magnetic fields along the direction of propagation and perpendicular to it, while the electric field is purely transverse.

We must discuss two important cases when dealing with propagating surface plasmons. First, the thick film case, and secondly the thin film case as there is a well known transition in the nature of these surface plasmons as one reduces the thickness of the metal film.

For thick metal films, we deal with a single interface between two infinitely large media. It is possible to derive a wavevector that the surface plasmon exists at, for a given permittivity and permeability of each medium.

To derive this conditions for a single interface TM surface plasmon we must derive the conditions where the electric field along the direction of propagation are continuous, otherwise the TM surface plasmon cannot exist. We begin with the well-known boundary conditions for the continuity of electric fields between interfaces [36].

$$\epsilon_{dielectric}\gamma_{metal} = -\epsilon_{metal}\gamma_{dielectric} \quad (2.27)$$

where γ_n is the decay constant, related to the decay of the electric field into medium n. We can then write the decay constants in terms of k , ϵ , μ of each medium using the following relation.

$$\frac{k^2 - \epsilon_{metal}\mu_{metal}}{\epsilon_{metal}^2} = \frac{k^2 - \epsilon_{dielectric}\mu_{dielectric}}{\epsilon_{dielectric}^2} \quad (2.28)$$

Solving for k gives the following equation for the TM surface plasmon dispersion relation.

$$k_{TM} = k \sqrt{\frac{\epsilon_{dielectric}\mu_{dielectric}\epsilon_{metal}^2 - \epsilon_{metal}\mu_{metal}\epsilon_{dielectric}^2}{\epsilon_{metal}^2 - \epsilon_{dielectric}^2}} \quad (2.29)$$

where k is the wavevector of the light in medium 1. If $\mu_r = 1$, this simplifies to the most well-known example of this equation. It is clear from these two equations that if the permittivities of each medium are equal, then a TM surface plasmon cannot exist.

$$k_{TM} = k \sqrt{\frac{\epsilon_{dielectric}\epsilon_{metal}}{\epsilon_{dielectric} - \epsilon_{metal}}} \quad (2.30)$$

For TE surface plasmons, we require a similar continuity in the magnetic field along the direction of propagation.

$$\mu_{dimagnet}\gamma_{metal} = -\mu_{metal}\gamma_{dimagnet} \quad (2.31)$$

where μ_m is the magnetic permeability in medium m . The derivation follows the same steps as the TM case, resulting in the following equation.

$$k_{TE} = k \sqrt{\frac{\epsilon_{dimagnet} \mu_{dimagnet} \epsilon_{metal}^2 - \epsilon_{metal} \mu_{metal} \epsilon_{dimagnet}^2}{\mu_{metal}^2 - \mu_{dimagnet}^2}} \quad (2.32)$$

It is obvious from equation 2.26 that a TE surface plasmon for thick films is not possible, as the denominator would be zero for bulk materials at optical frequencies.

Now we must consider what happens as one reduces the thickness of the thick metal film in equations 2.17 and 2.19. As the thickness of the metal film is reduced, the coupling between surface plasmons at the surfaces of the metal can not be ignored. These plasmon modes are referred to as "even" and "odd", "symmetric" and "asymmetric" or "long range propagating surface plasmon" and "short range propagating surface plasmon".

Such equations can not be analytically solved, and must be solved numerically, but those equations are shown below [36]. Another more practical solution is to use eigenmode analysis to identify the surface plasmons, as will be shown in Figures 2.6, 2.7 and 2.8.

For the even and odd TM modes

$$\sqrt{\epsilon_{metal} \mu_{metal} - \epsilon_{dielectric} \mu_{dielectric} - \kappa^2} = \frac{\epsilon_{dielectric}}{\epsilon_{metal}} \tan\left(\frac{k_0 \kappa}{d_{metal}}\right) \quad (2.33)$$

$$\sqrt{\epsilon_{metal} \mu_{metal} - \epsilon_{dielectric} \mu_{dielectric} - \kappa^2} = \frac{-\frac{\epsilon_{dielectric}}{\epsilon_{metal}}}{\tan\left(\frac{k_0 \kappa}{d_{metal}}\right)} \quad (2.34)$$

For the even and odd TE modes

$$\sqrt{\epsilon_{metal}\mu_{metal} - \epsilon_{dimagnet}\mu_{dimagnet} - \kappa^2} = \frac{\mu_{dimagnet}}{\mu_{metal}} \tan\left(\frac{k_0\kappa}{d_{metal}}\right) \quad (2.35)$$

$$\sqrt{\epsilon_{metal}\mu_{metal} - \epsilon_{dimagnet}\mu_{dimagnet} - \kappa^2} = \frac{-\frac{\mu_{dimagnet}}{\mu_{metal}}}{\tan\left(\frac{k_0\kappa}{d_{metal}}\right)} \quad (2.36)$$

We note that the physical dimensions of the film affect the dispersion of these even and odd modes, highlighting how the dimensions of a thin film affect the dispersion, i.e, spatial dispersion is present in these thin film surface plasmons.

The dispersion of these modes is plotted in Figure 2.6 for a 40 nm thick film of Au surrounded by a dielectric of 1 and 1.51, while in the case of the single interface, it is an Au-n=1.51 interface. We selected 40 nm of Au given it's relevance to our work in following chapters. The effect of reducing the film thickness is the splitting of the single surface plasmon mode into two separate modes, which deviate from either the n=1 or n=1.51 light line, which corresponds to angle of incidence of 41.2 and 90 degrees respectively, hence only the symmetric mode can be directly excited via incident light at these wavelength. This region where a surface plasmon may be directly excitable, between the n=1 and n=1.51 light lines, is typically referred to as the light cone.

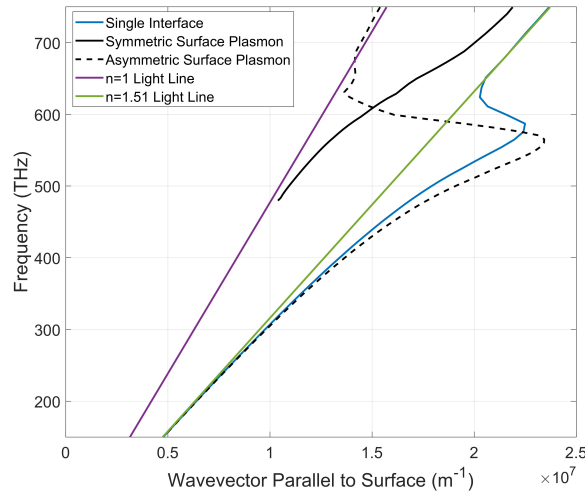


FIGURE 2.6: Dispersion relation for single interface plasmons, symmetric and asymmetric surface plasmons. For symmetric and asymmetric surface plasmons, the thickness of the Au layer is 40 nm.

The electric and magnetic fields of these modes can be plotted in Figure 2.7 and 2.8. In the case of modes that are outside the light cone, that is the single interface and asymmetric modes, their fields are purely decaying fields, with no radiative component. For the symmetric mode, as it lies in the light cone with an effective index similar to the $n=1.51$ substrate, it will radiatively decay into that substrate, which is seen with the sinusoidal electric field in Figure 2.8.

The results in Figure 2.7 and 2.8 match expectations for the distribution of electric and magnetic fields. Being TM surface plasmons, we see the expected two electric field components and one magnetic field component, with the electric and magnetic fields being transverse to each other, and another electric field component along the direction of propagation (y-direction). In the case of thin films in Figure 2.8, the symmetric and asymmetric nature of the modes is seen in the fields. For example, E_x for the symmetric surface plasmon and asymmetric plasmon. The asymmetric plasmon has two anti-parallel fields that are

on the $n=1.51$ and $n=1$ sides, the symmetric mode does not have this.

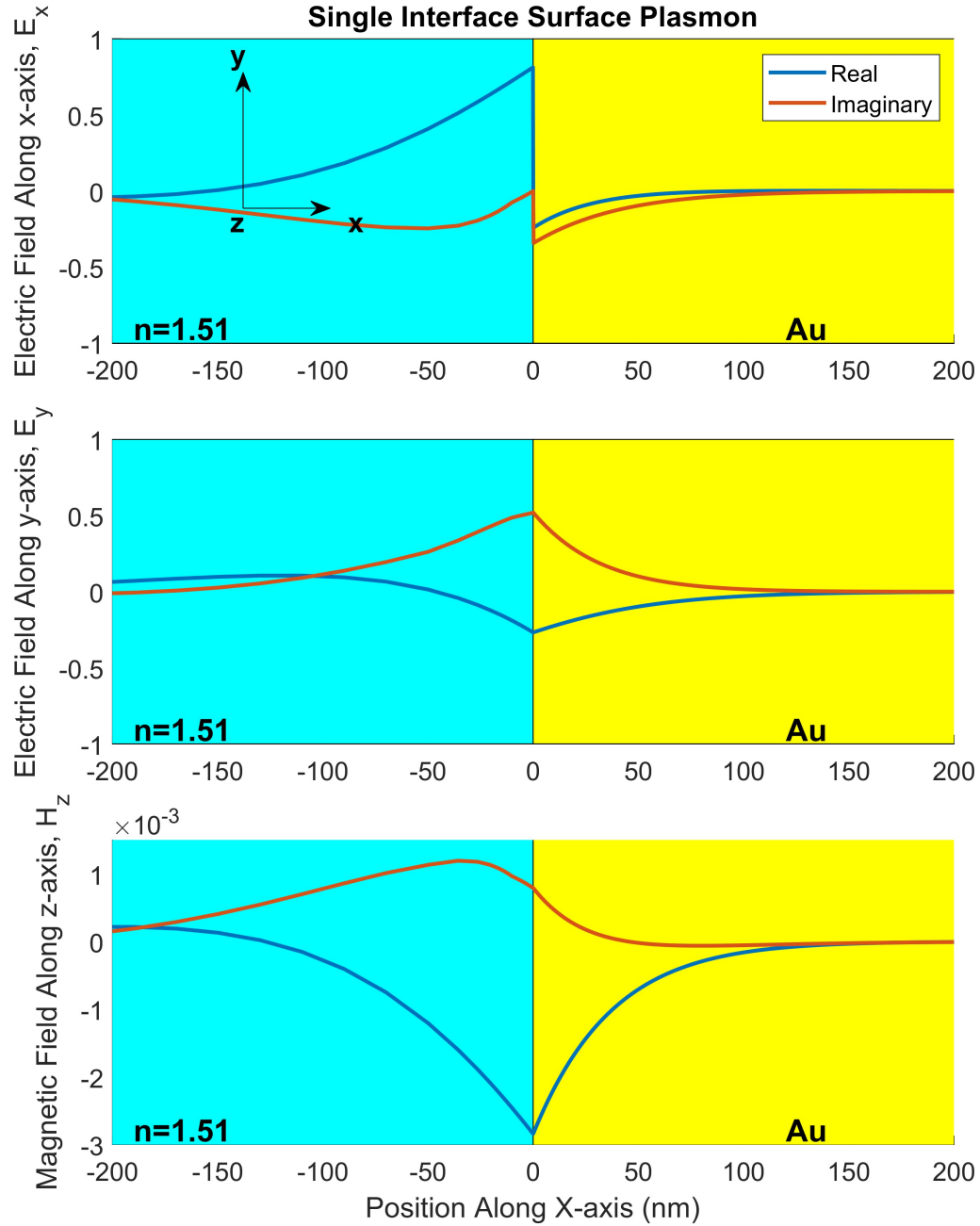


FIGURE 2.7: Electric and magnetic field profile of the single interface TM plasmon at 500 nm/600 THz. Units are in V/m.

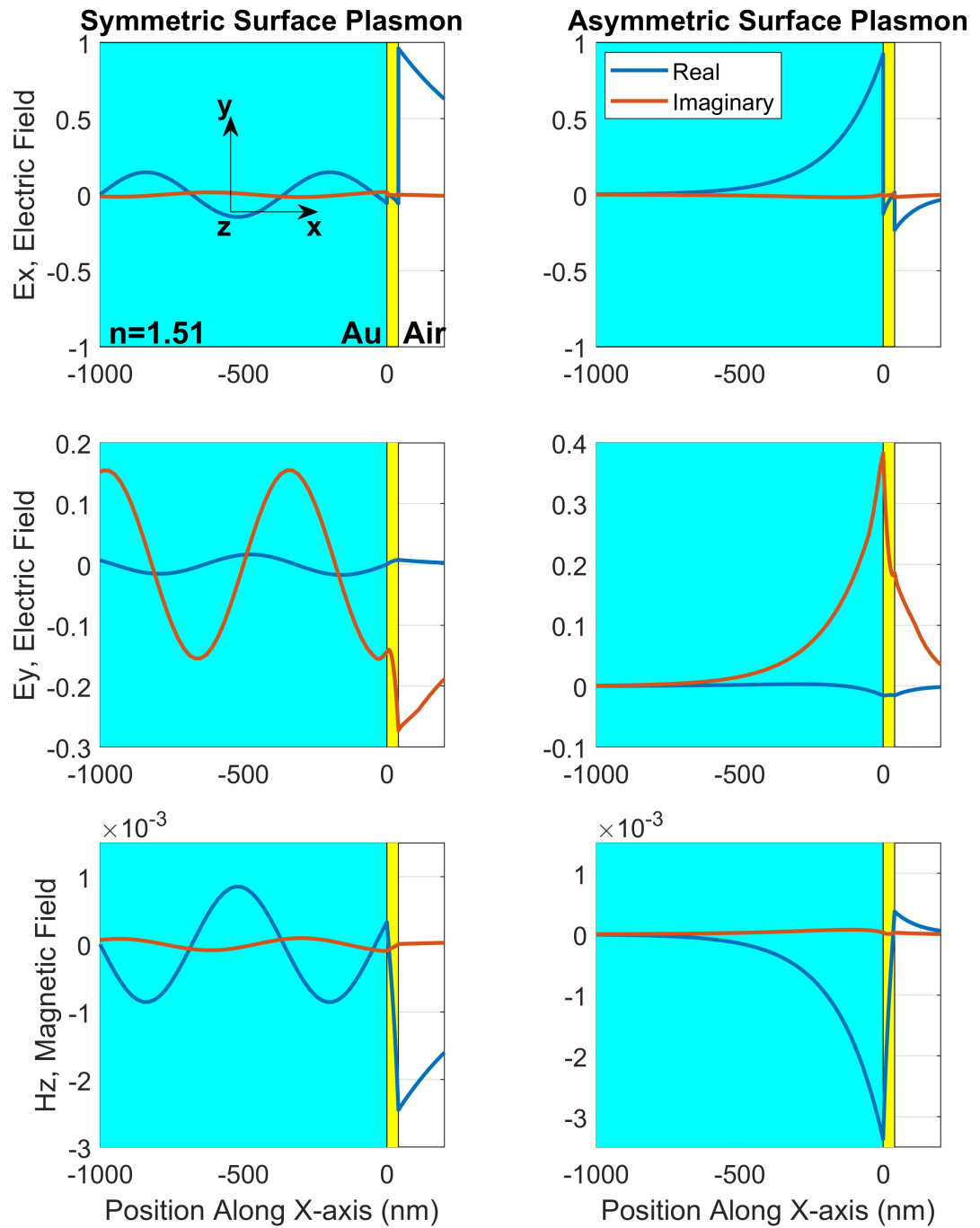


FIGURE 2.8: Electric and magnetic field profile of the symmetric and asymmetric plasmon at 700 nm/428 THz. Units are V/m.

2.3 Summary

In this chapter we discussed the concept of frequency dispersion, discussing several bulk material examples such as Si_3N_4 , SiO_2 , Ti and Au. These materials are key to this work of thesis, as we will see in the following chapters.

We introduced the concept of spatial dispersion, which is a wavevector dependence on the electric permittivity, a feature typically seen in bulk crystals, and associated with the structural anisotropy of the crystal lattice.

Next we revisited a comment discussed in the introduction, pertaining to the ambiguity between the dispersion of the magnetic permeability and spatial dispersion in permittivity, highlight how at optical frequencies the two are indistinguishable, providing an alternative route to achieving optical magnetism rather than the subwavelength case highlighted by inequality 1.20.

Having discussed dispersion in bulk materials, we highlighted how thin films may affect the dispersion already present in bulk materials, which is a form spatial dispersion arising from the finite thickness of those thin films. We discussed two examples relevant to our studies, Fabry Perot modes, which arise from the constructive and destructive interference of propagating waves, as well as the TM and TE propagating surface plasmons in thick and thin films, which are surface waves that propagate along interface between media. We noted that TE surface plasmon only exist for non-unity permeabilities while TM modes exist for non-unity permeabilities.

Chapter 3

Methodology

3.1 Approaches to Study Thin Films

In this chapter we outline our methodologies to study thin films. We selected thin films as candidates for study given the ease of obtaining and fabrication the required thin films. Furthermore it is easier to model the properties of thin films, than for example, structured metamaterials, which can be treated as 1 dimensional structures rather than modeling full 3D models.

To capture the effect of both frequency dispersion and spatial dispersion we need to be able to study the frequency and angle dependent reflection of these thin films, which is required to separately and identify frequency and spatial dispersion independently, as the angle of incidence and wavevector of the wave are related to each other.

Firstly, we outline the details of our samples, including measurements of refractive index and thickness of the samples. Two of these samples are simple thin films consisting of SiN and SiO₂, the third is a multilayer stack of Ti-Au-PMMA that is deposited on a SiO₂ substrate.

Secondly, we discuss the design of experiments to measure the frequency and angle dependent transmission of our SiN and SiO₂ thin films.

Thirdly, our novel "Dual Prism" approach to measure the frequency and angle dependent reflection from our Au-PMMA sample in a prism coupling configuration is presented. This is a modification of a well known prism coupling scheme known as the Kretschmann configuration. Compared to the traditional approach, ours is easier to construct and more reliable, while being limited to

a smaller range of angles.

Finally we discuss our modeling approaches to predicting the reflection and transmission of our samples with both COMSOL and Lumerical, including the theory behind models and practical usage of these software. Three separate approaches are used to model reflection, an analytical transfer matrix approach, a numerical S-parameter approach and another numerical approach, multipole decomposition. Practical use of both COMSOL and Lumerical to produce our results is then outlined.

3.2 Details of Thin Film Samples

Figure 3.1 (a) shows our Si_3N_4 thin film suspended in air, which is the green layer seen in the figure. It is supported by a silicon frame, as seen with the purple layer. The sample is brittle and hence, stored in a plastic pill to protect it. Our samples were purchased from Silson [37]. Figure 3.1 (b) shows our SiO_2 sample, which is not supported by any frame, these samples were purchased from Präzisions Glas & Optik GmbH [38]. Figure 3.1 (c) shows our Au-PMMA sample, we fabricated this sample in house, and the details of the fabrication are discussed later.

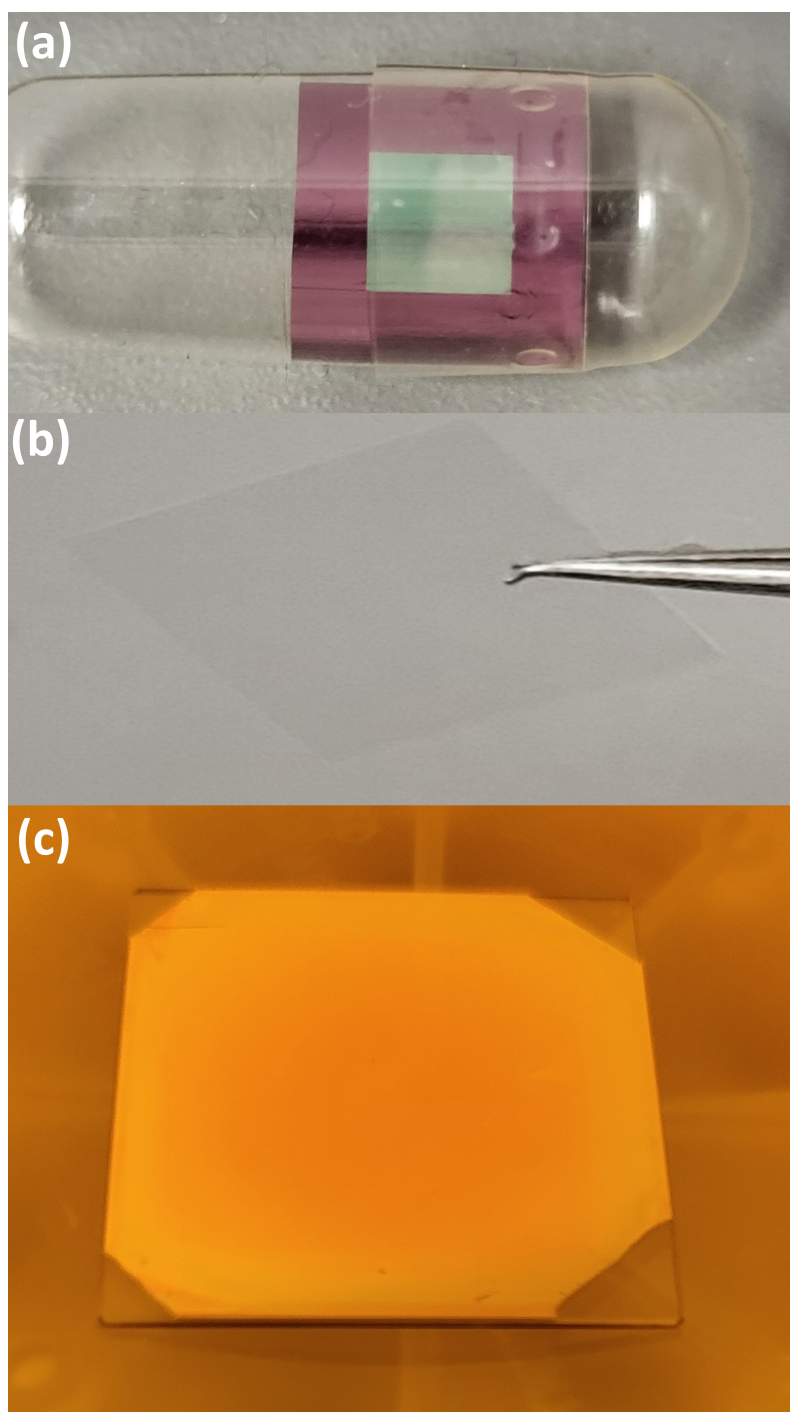


FIGURE 3.1: Images of samples studied throughout this thesis. (a) 500 nm of Si_3N_4 studied in Chapter 4. (b) 50 μm of SiO_2 studied in Chapter 4. (c) PMMA-Au Sample sample fabricated and studied in Chapter 5.

3.2.1 Refractive Index and Thickness Determination of Si_3N_4 Thin Film

We would like to thank Dr. Christopher Smith for ellipsometry measurements, who measured the refractive index of our Si_3N_4 sample, which is presented in Figure 3.2 (a) and (b) for wavelengths of 220-900 nm.

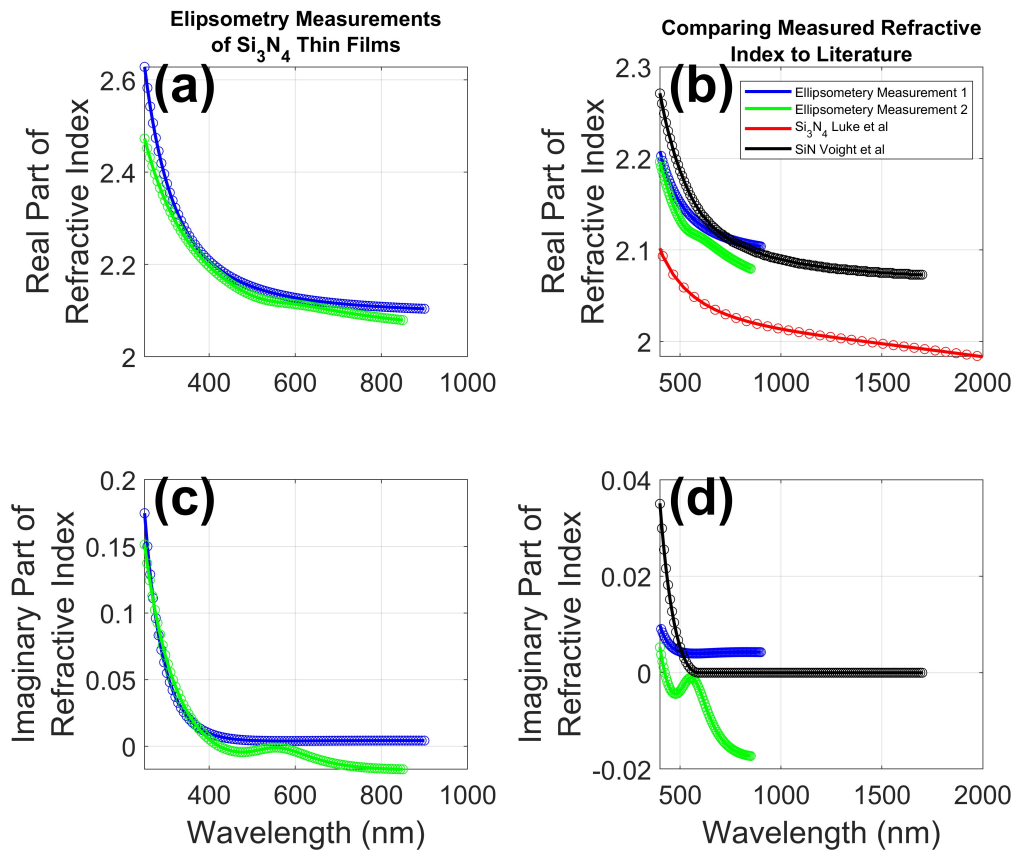


FIGURE 3.2: Ellipsometry measurements of SiN samples. (a) and (c) Measurements of the complex refractive index of our samples. (b) and (d) Comparison of measured complex refractive index to literature values.

The refractive index was extracted by fitting an absorbing Cauchy equation to the reflection of the thin film, which is shown below.

$$n(\lambda) = A + \frac{B}{\lambda^2} + \frac{C}{\lambda^3} \quad (3.1) \quad k(\lambda) = D + \frac{E}{\lambda^2} + \frac{F}{\lambda^3} \quad (3.2)$$

The fitted parameters, A, B, C, D, E, F are described in the table below with their corresponding uncertainties.

Parameter	Measurement 1	Measurement 2
A	$+2.065 \times 10^{+0} \pm 9.851 \times 10^{-3}$	$+2.032 \times 10^{+0} \pm 2.074 \times 10^{-3}$
B	$+9.848 \times 10^{-3} \pm 1.429 \times 10^{-3}$	$+2.671 \times 10^{-2} \pm 2.520 \times 10^{-3}$
C	$+1.518 \times 10^{-3} \pm 1.046 \times 10^{-4}$	$+5.626 \times 10^{-5} \pm 1.954 \times 10^{-5}$
D	$+2.996 \times 10^{-3} \pm 4.695 \times 10^{-3}$	$-2.382 \times 10^{-2} \pm 1.116 \times 10^{-3}$
E	$-2.002 \times 10^{-3} \pm 4.116 \times 10^{-4}$	$+4.053 \times 10^{-2} \pm 2.344 \times 10^{-2}$
F	$+2.775 \times 10^{-4} \pm 3.448 \times 10^{-5}$	$+1.283 \times 10^{-5} \pm 1.575 \times 10^{-5}$

TABLE 3.1: Table of fitted parameters for the Cauchy equation above.

The difference between measurements 1 and 2 in Figure 3.2 and Table 3.1 is the fitting method itself. In measurement 1 we assume a single layer, that is a smooth thin film with no rough surface. Measurement 2 involved fitting with 3 layers, the smooth thin film as with measurement 1, but with two rough surfaces on either side, these layers are a 50/50 mix of air and SiN whose thicknesses were estimated to be 13 and 5 nm. However both fittings produce the same thickness for the homogeneous SiN layer of $455 \text{ nm} \pm 5 \text{ nm}$. Measurement 2 does involve some fitting artifacts, specifically a negative imaginary part in Figure 3.2 (d), which suggests gain rather than absorption, which we

do not expect, however this can be treated as an artifact given the relatively low values of $k(\lambda)$, and in all cases we assume $k(\lambda) = 0$, but this assumption is only valid for wavelengths between 400-2000 nm.

In Figure 3.2 (c) and (d), we compare our measured refractive index to the refractive index from Luke et al [24] used for simulations in our work as well as another dataset for non-stoichiometric SiN from Vogt et al [39], identifying that our assumed refractive index is lower than the measured index of the sample by roughly 5-10%.

We find that the measured refractive index is a closer match to refractive index values for non-stoichiometric samples measured by Vogt [39]. This is a reasonable result given that free standing SiN samples can only be made by increasing the silicon content, relative to stoichiometric samples. The Vogt et al samples were fabricated via plasma deposition and used a Tauc-Lorentz oscillator model following the method outlined by [40] to extract the complex refractive indices, but further details on the specifics of deposition, surface roughness are not discussed in detail.

In addition the fitted thickness was lower than that stated by the supplier, as our ellipsometry measurements suggested that the thickness of the sample was $455 \text{ nm} \pm 5 \text{ nm}$ rather than 500 nm. Both the difference in modelled parameters will affect agreement between measurements and models, as we will show in Chapter 4.

In the case of anisotropic materials [41], the relative orientation of the crystal axis relative to the thin film is important consideration, as a permittivity tensor

is typically considered diagonal tensor, i.e, only the diagonal elements are non-zero, this corresponds to a crystal whose principal axis is aligned with the x , y , z co-ordinates of the fitting model. In the case where these axes are not aligned, we must consider the possibility that the non-diagonal elements. The best practice is to have detailed knowledge of the orientation of the crystal axis before measurements are taken.

Secondly anisotropic crystals can convert the linearly polarized incident ray into circularly polarized light, similarly to the effect of a quarter wave plate polarizer. This introduces additional complexity into measurement. For isotropic media only the amplitude ratio of s/p polarized light and phase difference between these two linear polarizations is measured, which is two independent parameters. For anisotropic media, the amplitude ratio/phase difference must be considered for cases where polarization is conserved and in cases where the polarization is converted, which is a total of six total parameters.

3.2.2 Fabrication of Au-PMMA samples

Our Au-PMMA samples consist of 1 nm Ti, 40 nm Au and 1200 nm of PMMA deposited on an SiO_2 . The SiO_2 substrates were cleaned in separate baths of acetone, IPA and deionized water in that order in an ultrasonic bath for 5 minutes each. Then the surface was cleaned in via O_2 plasma etching to remove remaining impurities.

The first deposition performed was the deposition of metal layers onto the cleaned substrate of SiO_2 . 1 nm of Ti was deposited first, to aid the deposition

and adhesion of a uniform Au film. Then 40 nm of Au was deposited on top of the Ti layer. We would like to thank Dr. Ali Mughaid for the deposition of the Ti and Au metal layers.

Then this Au surface was cleaned following the same procedure outlined earlier, with one additional step. To ensure that no dust particles remained on the surface, the sample was heated on a hotplate for 15 minutes at a temperature of 80 degrees Celsius.

Then deposition of the PMMA layer was performed via spin-coating. The spin coating of PMMA A11 495K from EMresist was performed at 1500 rpm for 3 minutes, after spin coating the sample was placed on a hotplate for 1 minute at 180 degrees Celsius. Visual inspection of the PMMA film was then performed. As seen in Figure 3.4 (a), ideal, uniform thin films are of a constant red hue, when under the light of the cleanroom. The presence of rings indicates a non-uniform thickness as seen in Figure 3.4 (c). After this was observed, the PMMA layer was removed with acetone, and the spin coating was repeated. These features are made clearer by increasing the contrast in Figure 3.4 (b) and (d).

We would also like to thank Dr. Christopher Smith for ellipsometry measurements, determining that the thickness of the spin coated PMMA was 1201 ± 1 nm. In our case, we assumed the refractive indices of the SiO_2 substrate, air superstrate and PMMA to be constant values of 1.51, 1, and 1.51 respectively [42]. The Au values were from Johnson and Christy [25] values while Ti was from CRC values [26]. However, such assumptions could be sources of error themselves given different deposition techniques between our samples and theirs.

An issue encountered during experiment was the contamination of the sample with the index matching fluid used to couple the light in the prism into the SiO_2 substrate. We found that it leeches into the PMMA and dissolves it, similar to paper chromatography. An example of such a contaminated film is seen in Figure 3.4 (e). This makes long term storage of the samples difficult, and typically the PMMA layer is destroyed after a single measurement is taken, however cleaning the sample, and redepositing the PMMA layer is possible.

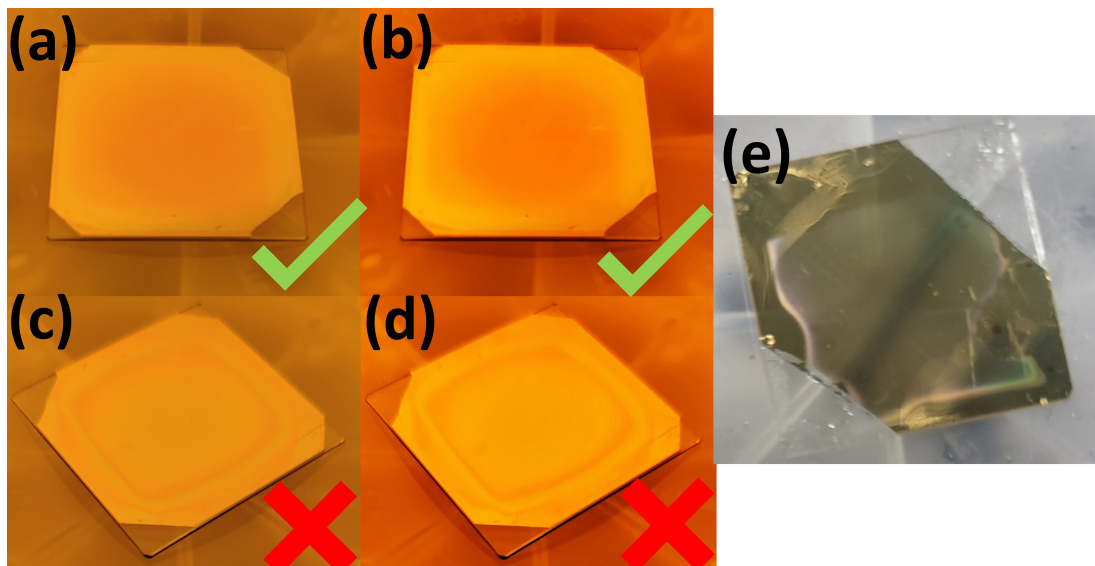


FIGURE 3.3: Example of ideal (a), (b) and non-ideal (c), (d) PMMA coatings. Sub-figures (b) and (d) are manipulated, with contrast increased by 300% relative to subplots (a) and (c) respectively, to make surface features more visible to reader. Subplots (a) and (b) appear orange due to the light of the clean-room. (e) Example of sample damaged after long term exposure to index matching fluid used in experiment. It appears that the index matching fluid dissolved, then reformed the PMMA surface after evaporating, making surface unusable after.

As for concerns regarding potential chemical changes during ellipsometry/exposure to natural light, which may change the refractive index of our PMMA sample. PMMA is not typically used as a photo-resist given that the chain scission chemical reaction required for patterning requires wavelengths that are

smaller than 250 nm and more pronounced the shorter the wavelength as the absorption of the material increases [43]. For lithography at longer wavelengths additional chemical alternation is required, which was not performed, nor is the standard for commercially available PMMA. The only case of well documented photolithography being used is in the X-ray region well beyond the wavelengths studied in this work. Measurements of the complex refractive index of PMMA are seen below and sourced from [44, 45, 46]. Exposure to short wavelengths should reduce the length of the PMMA polymer chains, however no such data on the impact of chain length of refractive index is documented in literature. No reference or concern was mentioned regarding this issue changing n - k during these ellipsometry measurements.

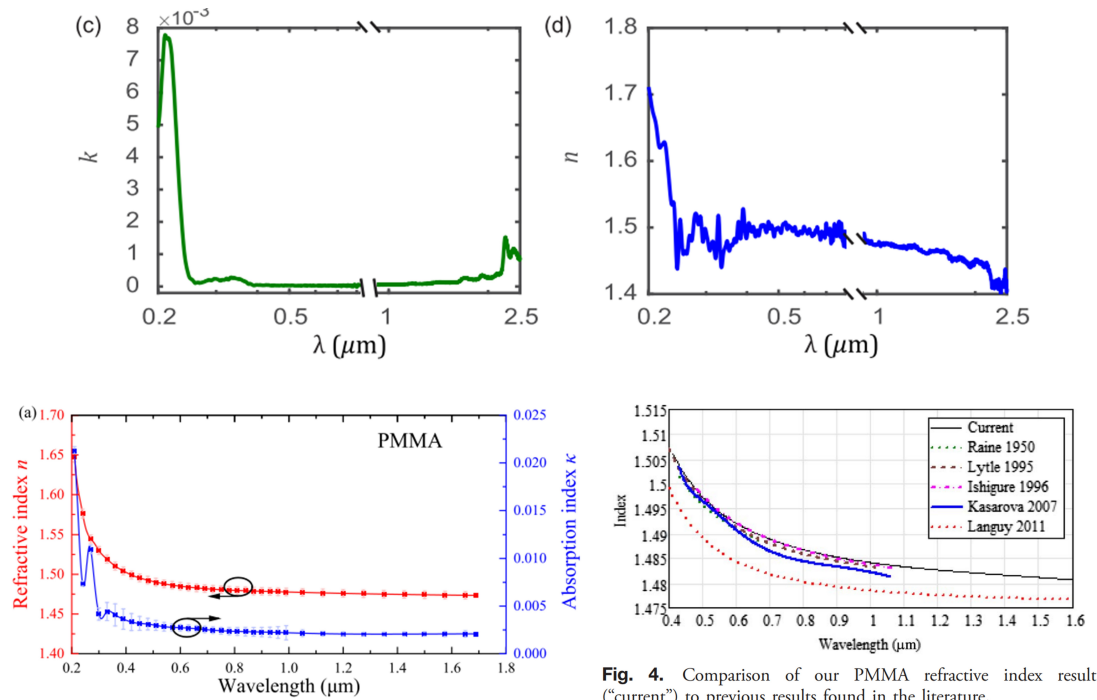


FIGURE 3.4: Examples of measured refractive index sourced from [44, 45, 46].

Fig. 4. Comparison of our PMMA refractive index result ("current") to previous results found in the literature.

3.3 Measuring Reflection and Transmission of Thin Films

3.3.1 Measuring Transmission Through Thin Films at Glancing Angles

In order to measure the transmission through SiN and SiO₂ thin films, the set up outlined in Figure 3.7 (a) was constructed. To achieve reliable transmission measurements approaching glancing angles, i.e. towards 90 degrees angle of incidence, a highly divergent beam is recommended for the following reasons.

Firstly, there will always be a ray passing through the center of the rotation of the motor, and hence the beam will not “run off” the surface as we change the angle of incidence.

Secondly, as the sample is rotated towards larger angles, the surface area of the sample exposed to the beam is reduced. If the sample’s surface area is significantly smaller than the beam diameter, one will measure a signal from both the sample surface, but also free space and the sample corners, reducing the accuracy of the measured transmission. This issue with large angles of incidence is demonstrated below in Figure 3.5. Further optimization at larger angles is feasible by increasing the distance between the iris and sample, or using a magnifying lens placed in-between iris and sample.

This is a desirable problem as it makes alignment of the beam with the set up significantly easier and is a critical consideration in the calibration procedures

required for an accurate measurement of transmission, as will be shown in the following discussion.

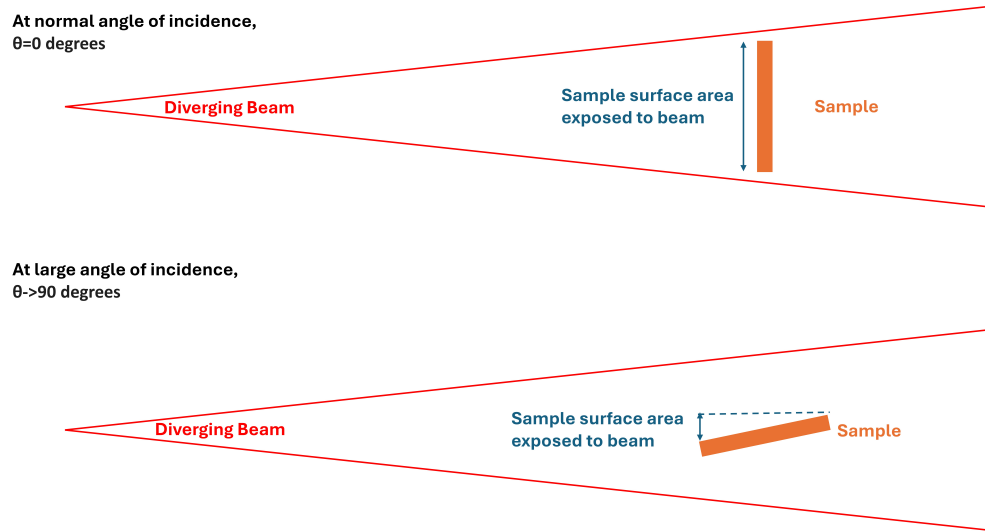


FIGURE 3.5: Issue with measurements at very large angles.

The samples are held in place by attaching them to a thicker more stable SiO_2 substrate, as seen in Figure 3.6. An index matching fluid is placed at the edge of the sample, this region is then pressed against the thicker holding substrate, a combination of atmospheric pressure, surface tension and molecular bonding keeps the sample connected to the glass substrate without applying stress or damaging the sample. This technique is known as "wringing" [47] and is applied to our dual prism approach as well. Figure 3.6 shows a sample being held in place through this "wringing" approach and can be seen in Figure 3.6, where our $50\ \mu\text{m}$ SiO_2 sample is in contact with a thicker SiO_2 sample.

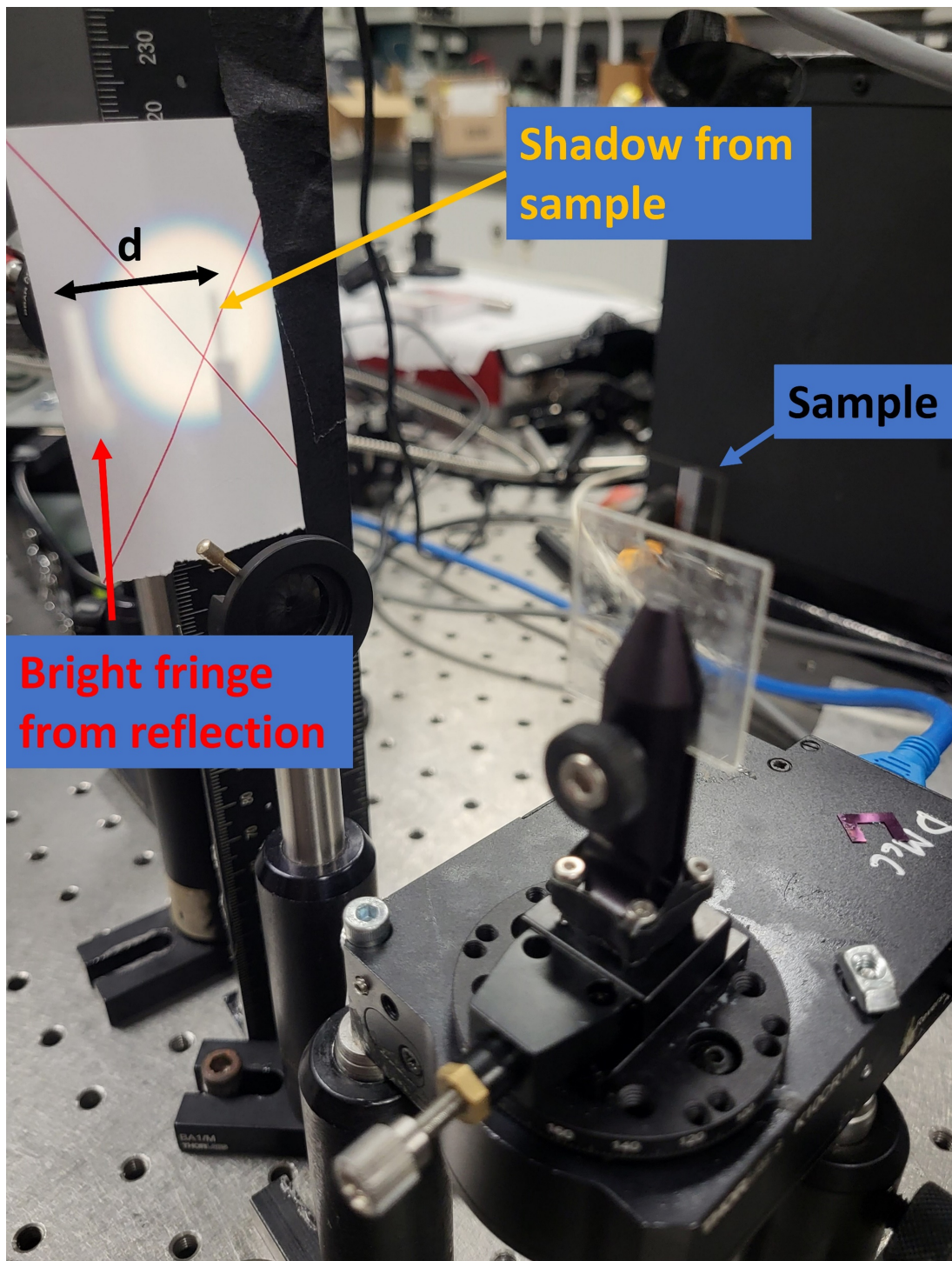


FIGURE 3.6: Image of set up in Figure 3.7 with various parts labeled.

To correctly calibrate the setup outlined in Figures 3.7 (a), 3 steps are outlined

in Figures 3.7 (b) and (c) and Figure 3.9. The first step outlined in Figure 3.7 (b), to know the true angle of incidence, we search for the ± 90 -degree angle of incidence. This can be found by matching the shadow of the sample with a bright fringe resulting from the near perfect reflection seen in all films towards 90 degrees. An angle of incidence of 90 degrees is obtained when this bright fringe disappears or overlaps with the shadow of the sample, this fringe and shadow are shown in Figure 3.6.

Step 2 is outlined in Figure 3.7 (c). The possibility remains that the sample is not aligned with the center of rotation of the motor. This will be observable when rotating the sample between ± 90 degrees, as the shadow of the sample will be laterally displaced. The center of rotation will lie in between the position of the two shadows and will be the region where the sample shadow must be moved to.

If steps 1 and 2 have been performed correctly, mirror symmetry through an angle of incidence of 0 when measuring reflection, as seen in Figure 3.9 is observed.

The reference measurement that accounts for optical losses in the setup, is presented in Figure 3.9 (b). It is the ratio of the measured intensity from the sample and the reference intensity without the sample that gives us measured reflection in Figure 3.9 (c).

It is important to note that the signal to noise is particularly weak below 400 nm, as the spectral power of the tungsten lamp is quite low in that region. Very large angles have major issues as the corners play a non-negligible role in the measured reflection, making observation of the Fabry Perot modes at large

angles difficult. This is seen at very large angles in Figure 3.9(c), with the large red regions, which suggest large values of reflection, but arise from this corner issue.

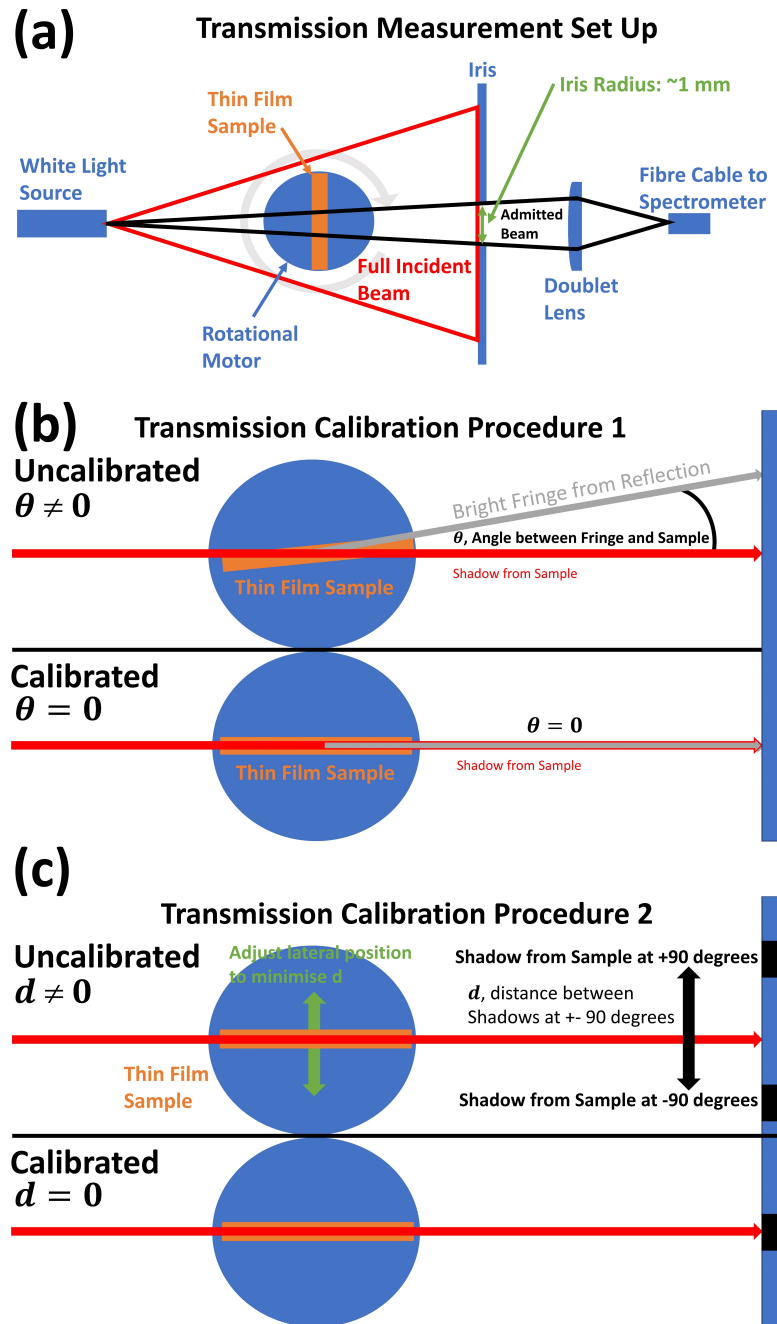


FIGURE 3.7: (a) Schematic diagram of set-up used to measure transmission for Si_3N_4 and SiO_2 samples in Chapter 4. (b) First procedure to calibrate the angle of incidence. The overlap between the bright fringe resulting from reflection (grey arrow) and the shadow (red arrow after sample) corresponds an angle of zero between the shadow and fringe, which corresponds to an angle of incidence of 90 degrees between the sample and incident beam. (c) Second calibration procedure to reduce the mismatch between the center of the sample and the center of rotation.

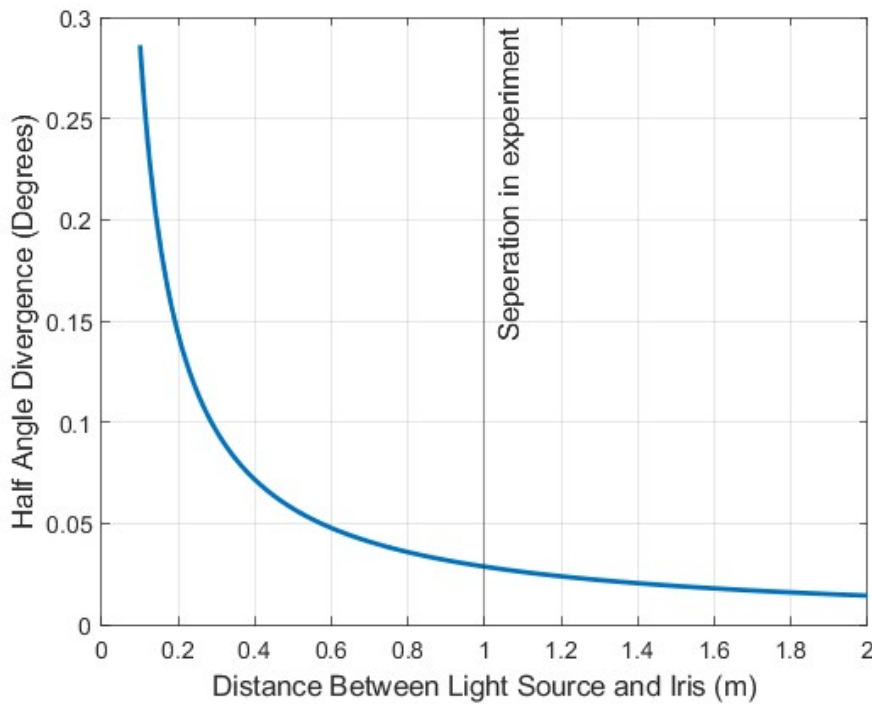


FIGURE 3.8: Calculated half angle divergence

Another consideration in designing this experiment is the half angle divergence, as this adds certainty to the angle of incidences measured. If the half angle divergence is sufficiently large relative to a given resonance, then the resonance will not be correctly resolved. To minimize the half angle divergence, an iris is placed between the beam and fiber cable port in 3.7 (a). The half angle divergence of this measured beam can be calculated with the tangent function, $\tan(\theta_{HalfAngleDivergence}) = \frac{Iris/2}{Seperation}$. This is plotted in Figure 3.8. The vertical line marks the separation in experiment, showing that the half angle divergence is roughly 0.025 degrees. This suggests that the uncertainty in angle of incidence is ± 0.025 degrees which is sufficiently small to avoid any issues related to very large divergences.

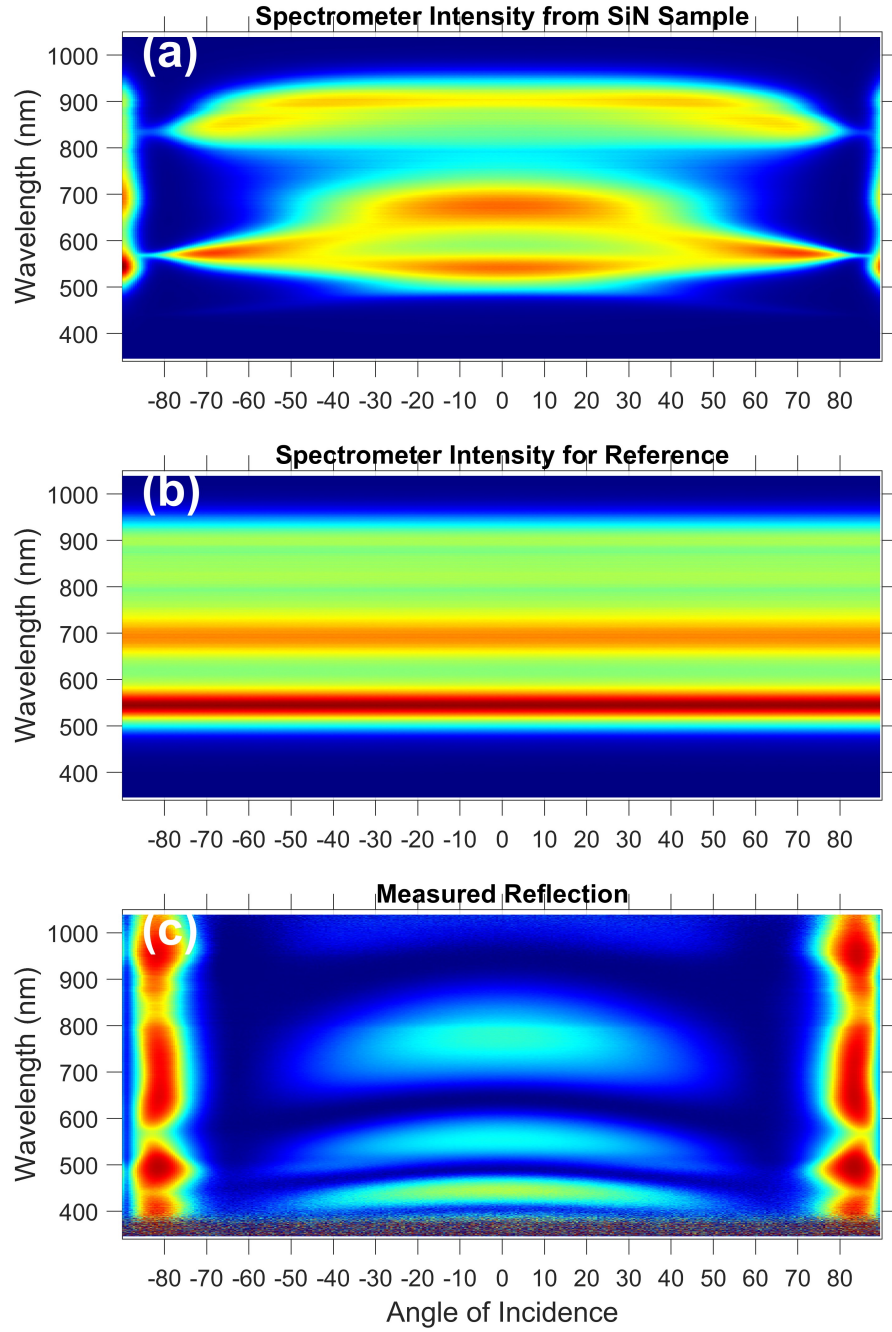


FIGURE 3.9: Example of verified measurement after steps outlined in Figure 3.7 for p-polarized light in the visible spectrum. (a) Measured intensity from SiN sample. (b) Measured intensity used for reference, i.e. intensity in the absence of SiN sample. (c) Measured reflection, calculated from measured transmission, which is the produced by dividing subplot (a) by subplot (b).

3.3.2 Measuring Reflection in Prism Coupling/Kretschmann Configuration Through the Dual Prism Approach

The following is a method to measure reflection in the well-known prism coupling method, however to our knowledge this exact approach has not been demonstrated in literature. Current methods involve two rotating arms as used in optical ellipsometry, involving delicate alignment between those arms to properly measure the reflected signal.

We developed a novel approach to measure the reflection that uses a dual prism arrangement as show in Figure 3.10. By connecting two prisms with index matching fluid and placing them at the center of rotation of the motor we remove the requirement for two motors, and the angular motion of the beam is replaced by a small lateral displacement of the reflected beam. This small lateral displacement can be accounted for by using a doublet lens, which is designed with off axis performance in mind [48].

Calibration of the angle of incidence requires the user to measure the reflection and observe the angle at which the reflection values are greater than 1.

The reflection from a sample is calculated by dividing the reflected signal from the sample, Figure 3.13 (b), with the reflected signal from a glass substrate, Figure 3.13 (a). This calculated reflection is only less than one above the critical angle of glass, as both samples are perfectly reflecting above that angle. Below the critical angle, the Au-PMMA sample reflects more light than the glass reference, resulting in a reflection value greater than 1. This limits measurements to angles above the critical angle.

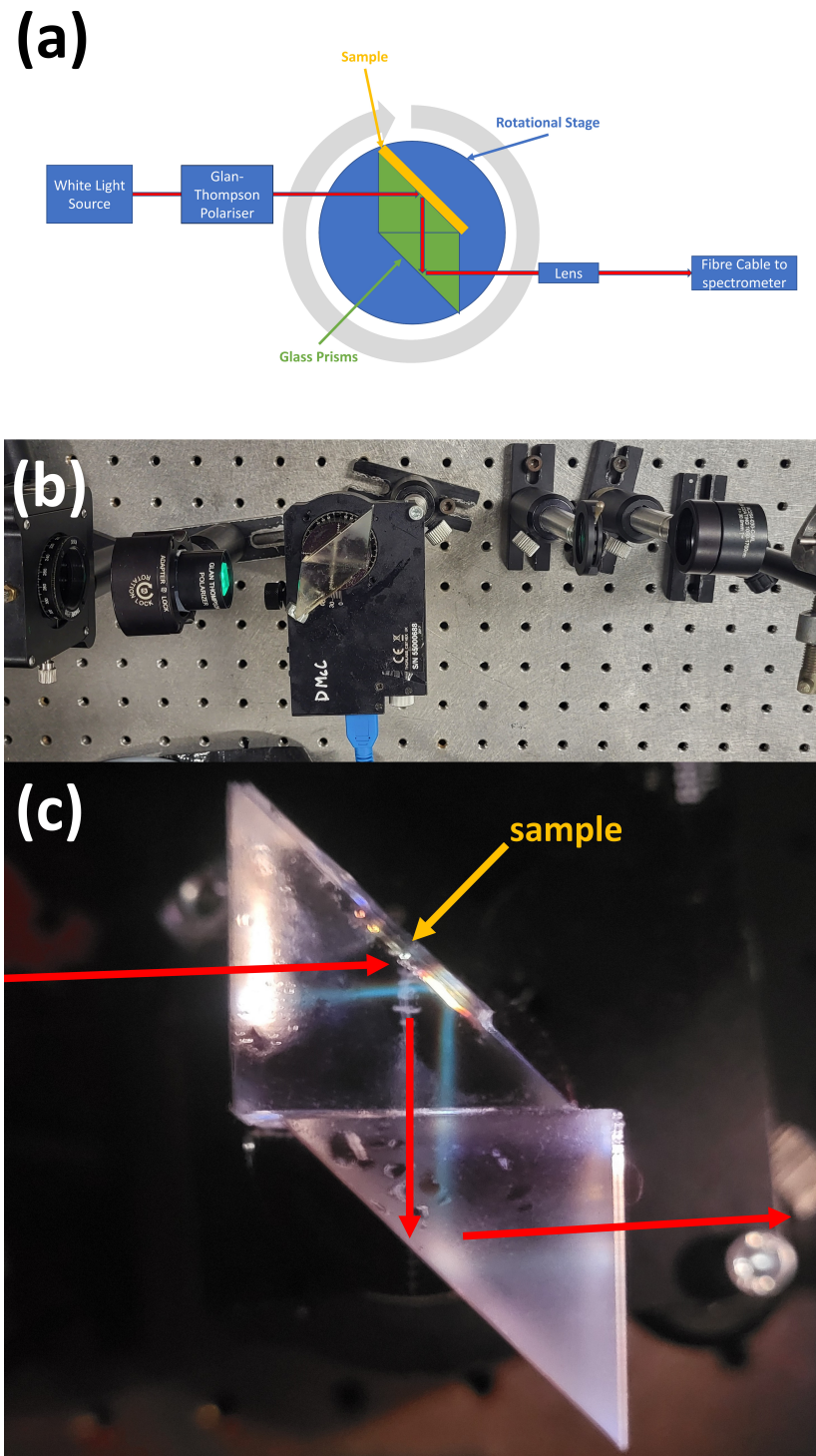


FIGURE 3.10: (a) Schematic of set up used to measured reflection via the Kretschamn configuration for the Au-PMMA sample studied in Chapter 5. (b) Image of set up described in (a). (c) Path of beam inside prism, shown by the bright blue path, which is fluorescence from the glass prism after being pumped by a super continuum laser.

This can be seen in Figure 3.11 below, when modeling the reflection at 1000 nm as a function of angle, the ideal case for the reference is to perfectly reflect the incident light so that any change in reflection can be assumed to come from the sample. This only occurs above the critical angle at 42.1 degrees, which corresponds to the critical angle of the SiO₂-air interface. Using a sufficiently thick metallic reference may extend the minimum measurable angle to angles below the critical angle, but would require require the glass reference to calibrate the angle of incidence, and the metal reference to measure the reference signal.

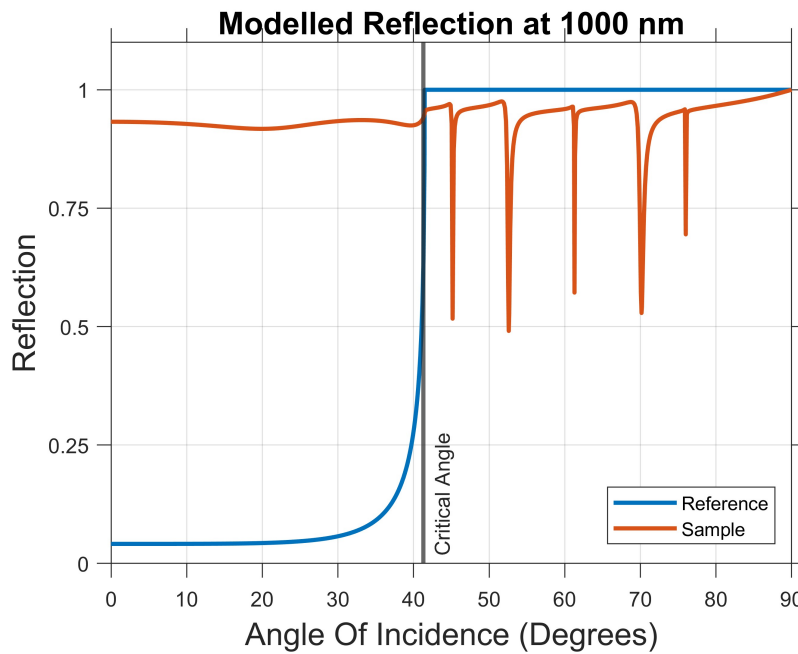


FIGURE 3.11: Modeled reflection using transfer matrix method for unpolarized light. Sample is the Au-PMMA sample studied in Chapter 5. Reference refers to a thick film of SiO₂. The reference is only valid as a reference when it perfectly reflects light, which occurs above the critical angle.

Once the critical angle has been identified, the follow steps are taken to convert the programmed angle of incidence which is associated with the motor, to the true angle of incidence striking the sample. The steps taken to calibrate the true angle of incidence from the programmed angle are as follows:

Step 1. Identify the programmed angle that corresponds to the critical angle. In Figure 3.6 (a), it is marked by the rapid increase in the reflected signal at -8.1 degrees, which should corresponds to a true angle of incidence of 42.1 degrees, the critical angle of the glass-air interface.

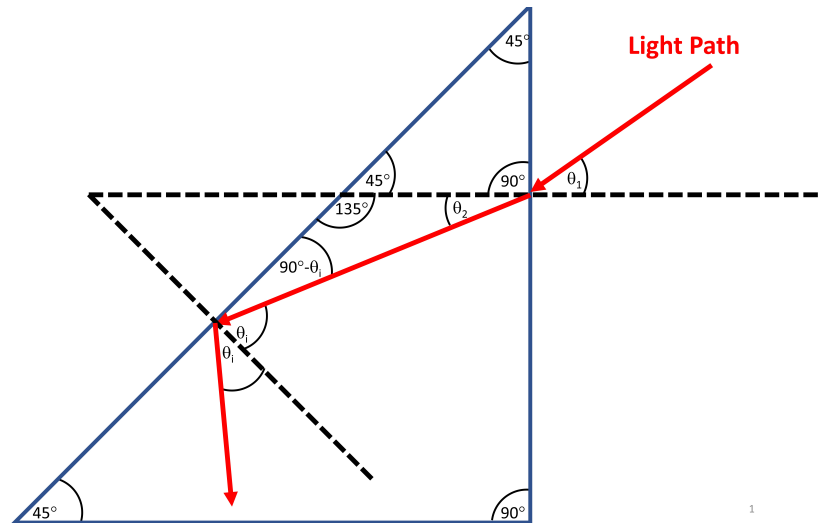


FIGURE 3.12: Schematic diagram of rays traveling through the first prism

Step 2. We now calculate θ_1 , the angle of incidence at the interface between the air and prism, as seen in Figure 3.12. To do so we need to derive a relation between θ_1 and θ_i , where θ_i is the true angle of incidence presented in the main text.

First, we define a triangle consisting of the following angles, $90^\circ - \theta_i$, 135° and θ_2 . This triangle is shown in Figure 3.12. The sum of these angles is equal to 180° .

$$180^\circ = 90^\circ - \theta_i + 135^\circ + \theta_2 \quad (3.3)$$

Simplifying this equation gives us the following.

$$\theta_i = 45^\circ + \theta_2 \quad (3.4)$$

Now we introduce Snell's Law.

$$n_1 \sin(\theta_1) = n_i \sin(\theta_i) \quad (3.5)$$

Combining these two equations we obtain the following relation between θ_1 and θ_i

$$\theta_i = \sin^{-1} \left(\frac{\sin(\theta_1)}{n_2} \right) + 45^\circ \quad (3.6)$$

or in reverse.

$$\theta_1 = \sin^{-1}(n_2 \sin(\theta_i - 45^\circ)) \quad (3.7)$$

Step 3. Now we convert $\theta_i = 42.1$ degrees to θ_1 , which is 5.2 degrees. The difference between 5.2 and -8.1 is then subtracted from the programmed angle. This converts the programmed angle of incidence, to the angle of incidence striking the air-prism interface (θ_1).

Step 4. Now using equation 3.6, we convert these new angles to the angle of incidence, θ_i , striking the sample.

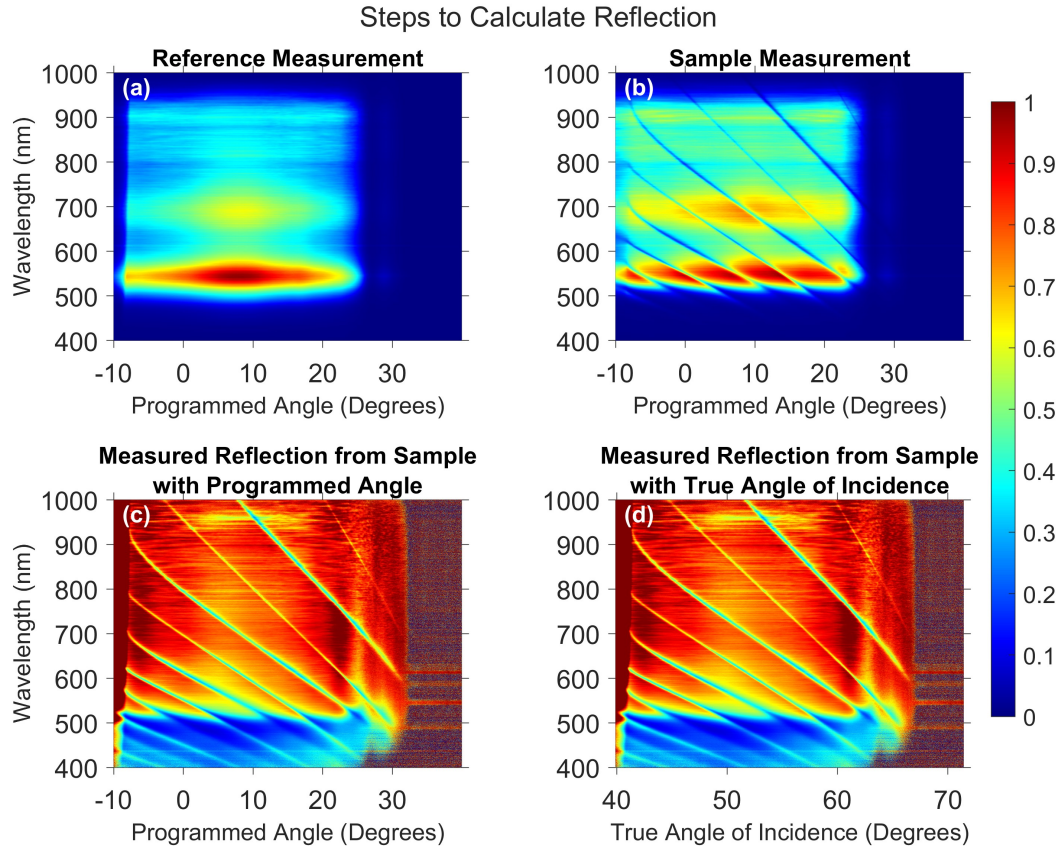


FIGURE 3.13: Demonstration of calibration procedure for reflection set up described in Figure 3.10. (a) Measured intensity for reference signal. (b) Measured intensity for sample. (c) Measured reflection with angles associated with the motor. (d) Measured reflection with angles associated with the true angle of incidence.

3.4 Modeling Reflection of Thin Films

There are a wide variety of approaches to modeling the reflection of a plane wave from a thin film to verify the measurements we have discussed so far. Here we outline three approaches used in this thesis.

The transfer matrix method, which provides an analytical solution when numerical simulations are too time intensive. This approach relies on modeling the series of reflected and transmitted rays which interfere with each other. By considering the magnitude of transmission and reflection from each interface, and the change in phase of transmitted and reflected beams as they interact with interface and as they propagate through layers of finite thicknesses.

The S-parameter or scattering parameter method, which is performed alongside the calculations required for multipole decomposition when numerical calculation are feasible. This approach relies on numerically solving Maxwell's equations and getting the ratio of the magnitude of the reflection divided by the magnitude of the incident wave in the reflected or transmitted direction.

Multipole decomposition is a relatively new approach, allowing for the interpretation of reflection through a more fundamental approach, by converting the light induced current densities into electric and magnetic multipoles. This third approach provides the most information of the three. Unlike the previous methods, we do not directly model the propagation of light through the thin film. Instead we numerically solve Maxwell's equations, and calculate the electric current density in the thin film, we then use these vectors to calculate the moment of electric and magnetic multipoles to predict reflection [49]. This

allows us to decompose the electric fields studied by the s-parameter technique into electric and magnetic multipoles.

3.4.1 Transfer Matrix Method

The transfer matrix method relies on a series of matrices to describe the reflection and transmission at interfaces and propagation matrices to capture the phase the light accumulates as it propagates through a medium.

We begin with the Fresnel equations for reflection and transmission at single interfaces for both s- and p- polarized light between two media, m and $m + 1$. n_m is the refractive index of the m th layer.

$$r_s = \frac{n_m \cos(\theta_i) - n_{m+1} \cos(\theta_t)}{n_m \cos(\theta_i) + n_{m+1} \cos(\theta_t)} \quad (3.8)$$

$$t_s = \frac{2n_m \cos(\theta_i)}{n_m \cos(\theta_i) + n_{m+1} \cos(\theta_t)} \quad (3.9)$$

$$r_p = \frac{n_{m+1} \cos(\theta_i) - n_m \cos(\theta_t)}{n_{m+1} \cos(\theta_i) + n_m \cos(\theta_t)} \quad (3.10)$$

$$t_p = \frac{2n_m \cos(\theta_i)}{n_{m+1} \cos(\theta_i) + n_m \cos(\theta_t)} \quad (3.11)$$

The subscripts s and p correspond to the two states that linearly polarized light can be polarized along, either s- or p-polarized. For s-polarization the electric field oscillates in plane with the surface it strikes, while for s-polarized light the electric field oscillates perpendicular or out of plane with the surface it strikes. At normal incidence, the electric fields of both polarizations are perpendicular to the surface, and hence the Fresnel equations produce the same result regardless of polarization, it is only at non-normal angles of incidence that the difference in polarization becomes apparent.

The matrix T below defines the reflection and transmission coefficients at the interface between two media, m and $m + 1$.

$$T_m = \frac{1}{t_{m,m+1}} \begin{pmatrix} 1 & r_{m,m+1} \\ r_{m,m+1} & 1 \end{pmatrix} \quad (3.12)$$

Describing the change in phase that the light experiences as it propagates through medium m , the following matrix is used.

$$P_n = \begin{pmatrix} e^{-i\delta_m} & 0 \\ 0 & e^{i\delta_m} \end{pmatrix} \quad (3.13)$$

The change in phase is calculated from the following equation, d is the thickness of the layer, λ is the vacuum wavelength of the incident light.

$$\delta = \frac{2\pi d n_m}{\lambda} \quad (3.14)$$

To describe the reflection and transmission of N layers, one performs matrix multiplication for every layer as seen below.

$$M = T_1 P_1 T_2 P_2 \dots T_N P_N \quad (3.15)$$

To extract the reflection and transmission coefficient, r and t , are described as follows.

$$r = \frac{M_{21}}{M_{11}} \quad (3.16)$$

$$t = \frac{1}{M_{11}} \quad (3.17)$$

The amplitude of equations 3.18 and 3.19 are related to the intensity of the reflected and transmitted light respectively. This is equivalent to the measurements outlined in Figures 3.13 and 3.10, the amplitude of $E_{incidence}$ are equivalent the measured intensity of the reference signal, while $E_{reflection}$ and $E_{transmitted}$ is equivalent to measured intensities of the reflected and transmitted signals.

3.4.2 S-parameter Method

S-parameter calculations are the numerical equivalent to the Transfer Matrix Method outlined in the previous section, while S-parameter calculations are involved with circuits and microwave theory, we focus on the application of S-parameters at optical frequencies. COMSOL provides built in features to calculate the S-parameters which are valuable for numerically extracting reflection and transmission coefficients, once Maxwell's equations have been solved in the simulation volume. The following discussion is taken from COMSOL's own guide to calculate s-parameters in their Wave Optics Module [50].

We begin with the definition of the electric field at a "port", the port refers to the points of the simulation where an electric field is either introduced into a simulation, or removed from that simulation. In our case, two ports are used, a port to introduce the incident light, which calculates reflection and a port to remove the transmitted light, which calculates transmission.

$$E_c = E_a + \sum_{i=1} S_{i1} E_i \quad (3.18)$$

E_c is the computed electric field at the port where the field is incidence, E_a is the electric field pattern at port a, which the light is not incident upon. $\sum_{i=1} S_{i1} E_i$ are the mode fields at the port. Next we define the scattering matrix, $\hat{S}$.

$$\hat{S} = \begin{bmatrix} S_{11} & S_{12} & S_{13} \\ S_{21} & S_{22} & S_{23} \\ S_{31} & S_{32} & S_{33} \end{bmatrix} \quad (3.19)$$

$$S_{port\ a,b} = \int_{port\ a} \frac{(E_c - E_b) \cdot E_b^*}{E_b \cdot E_b^*} dA \quad (3.20)$$

To extract the reflection and transmission coefficients, we select S_{11} and S_{21} respectively, these are equivalent to equations 3.18 and 3.19, but are associated with the complex electric fields after numerical solving Maxwell's equations in the simulation volume.

3.4.3 Multipole Decomposition

Multipole decomposition is a widely used technique that allows for probing the fundamental light matter interactions for the scattering of light by particles, regardless of shape or size. Several well cited works describe methods to derive the scattered field as a function of electric/magnetic poles [51, 52, 53, 49, 54, 55].

The following is an overview of the derivations published by Evlyukhin et al [49]. Alternative derivations available elsewhere by R. Alaee et al [56], but both produce the same equations. This model is valid for an object of any general shape or size as long as the following conditions are considered, larger structures will require higher order multipoles to capture their behavior, if the

structure's center of mass is outside the structure, then this model is invalid. The following model only considered the current density to arise from the electric polarization of the medium, and assumes that the magnetic permeability is unity, hence this model may not work for bulk magnetic permeabilities different from one.

Our model is based on the work of reference [49], is the exact spherical multipole decomposition in the cartesian basis plane assuming a plane wave. The nature of toroidal moments appears controversial in literature [57], and we assume the perspective that these moments are associated with their respective electric/magnetic moments, i.e the toroidal dipole and electric dipole are considered to contribute to the total electric dipole moment. We took this stance for the EQ moment in equation 3.25, where the second term is related to the toroidal quadrupole moment.

We define the scattering of a monochromatic electric field by a scatter of any size or shape, with frequency as $e^{ik_d r}$ in the far field in the direction $\vec{n}$

$$E_n(r) = i\omega\mu_0 \frac{e^{ik_d r}}{4\pi r} (\hat{U} - \vec{n}\vec{n}) \int_V e^{-ik_d r} \vec{J}(r) dr \quad (3.21)$$

The above equation describes the scattered field in the scattering direction, $\vec{n}$.

$$e^{-ik_d r} = 4\pi \sum_{l=1}^{\infty} \sum_{m=-l}^l (-i)^l j_l(k_d r') Y_{lm}^*(\theta, \phi) Y(\theta, \phi) \quad (3.22)$$

ω is the angular frequency, $\hat{U}$ is the unit tensor, $\vec{n}\vec{n}$ is the outer tensor product of

the unit vector for the direction of the scattered field. k is the wavenumber. $\vec{J}$ is the current density. The exponential term can be described in the Cartesian basis using plane wave representation in spherical harmonics. $j_l(k)$ is the spherical Bessel function, Y are the scalar spherical harmonics. It is important to note the following, we do not consider the small argument/long-wavelength approximations to the spherical Bessel functions, as these introduce errors as the structure studied approaches the size of the wavelength. Bessel functions are easily available in COMSOL, Lumerical and many programming languages, there is no reason to apply this approximation.

From equation 3.22 one can derive any electric/magnetic pole by setting l and m to various integers. For example, to derive the electric dipole, we set $l=0$, $m=0$ and obtain the following. So the electric dipole moment, $\vec{p}_a$, along cartesian co-ordinate a , is described as follows.

$$\vec{p}_a = \frac{i}{\omega} \int_V j_0(k_d r) J_a dV + \int_V \frac{ik_d^2}{\omega^2} (\vec{r} \cdot \vec{J}) \cdot \vec{r} - \vec{r}^2 \cdot \vec{J} dV \quad (3.23)$$

$\vec{J}(r)$, is the current density that we have previously discussed in Chapters 1 and 2. By setting $l=1$, $m=-1, \dots, +1=-1, 0, 1$ we get both the magnetic dipole and electric quadrupole. Electric multipoles are easily identified by a dot product between the positional vector and current density, while magnetic multipoles use a cross product.

The magnetic dipole moment, $\vec{m}_a$, along cartesian co-ordinate a , is described as follows.

$$\vec{m}_a = \frac{3ik_d}{2} \int_V \frac{j_1(k_d r)}{k_d r} (\vec{r} \times \vec{J})_a dV \quad (3.24)$$

The magnetic dipole moment, and all following multipoles have a origin dependence, and the moments of these multipoles depend on selection of reference point, which has claimed to be an unphysical result, however this is untrue, as selecting a reference point should consider the boundaries of the medium [58]. In our case we set our origin to the x, y, z co-ordinates 0, 0, 0 and ensure that the center of our thin film/unit cell is located at this origin.

The electric quadrupole moment is

$$E\hat{Q}_{ab} = \frac{-ik_d}{2} \int_V \frac{j_1(k_d r)}{k_d r} (3(\vec{r}_a \vec{J}_b - \vec{J}_a \vec{r}_b) - 2(\vec{r}_a \cdot \vec{J}_b) \hat{U}_{ab}) dr + \frac{6ik_d^2}{\omega} \int_V \frac{j_3(k_d r)}{k_d^3 r^3} [5(\vec{J} \cdot \vec{r}) \vec{r}_a \vec{r}_b - r^2(\vec{J}_a \vec{r}_b + \vec{r}_a \vec{J}_b) - (\vec{J} \cdot \vec{r}) r^2 \hat{U}] dr \quad (3.25)$$

By setting $l=2$, $m=-2, -1, 0, 1, 2$ we get both the magnetic quadrupole and electric octupole.

$$M\hat{Q}_{ab} = \frac{5k_d^2}{2} \int_V \frac{j_2(k_d r)}{(k_d r)^2} ([\vec{r} \times \vec{J}]_a \vec{r}_b + \vec{r}_a [\vec{r} \times \vec{J}]_b) dr \quad (3.26)$$

$$E\hat{O}_{abc} = \frac{15k_d^2}{\omega} \int_V \frac{j_2(k_d r)}{(k_d r)^2} (\vec{J}_a \vec{r}_b \vec{r}_c + \vec{r}_a \vec{J}_b \vec{r}_c + \vec{r}_a \vec{r}_b \vec{J}_c - \hat{A}) dr \quad (3.27)$$

By setting $l=3$, $m=-3, -2, -1, 0, 1, 2, 3$ we get both the magnetic octupole and electric 16 pole. However, we do not include the 16 poles in our model, nor do

we show it below.

$$M\hat{O}_{abc} = \frac{105}{4} \int_V \frac{j_3(k_d r)}{(k_d r)^3} ([\vec{r} \times \vec{J}]_a \vec{r}_b \vec{r}_c + \vec{r}_a [\vec{r} \times \vec{J}]_b \vec{r}_c + \vec{r}_a [\vec{r} \times \vec{J}]_b \vec{r}_c + \vec{r}_a \vec{r}_b [\vec{r} \times \vec{J}]_c - \hat{A}'_{abc}) dr \quad (3.28)$$

To the calculate the scattered field from each pole in direction, $\vec{n}$, we use the following equation.

$$E_n^{scattered}(r) = iw\mu_0 \frac{e^{ik_d r}}{4\pi} [\vec{n} \times (\vec{p} \times \vec{n}) + \frac{1}{v_d} [\vec{m} \times \vec{n}] + \frac{ik_d}{6} [\vec{n} \times (\vec{n} \times (\vec{n} \times \hat{Q}(\vec{n}\vec{n})))] + \frac{ik_d}{2v_d} [\vec{v} \times \vec{M}\vec{v}] + \frac{k_d^2}{6} [\vec{n} \times [\vec{n} \times \hat{O}\vec{n}\vec{n}]] + \frac{k_d^2}{6v_d} [\vec{n} \times [\vec{n} \times \hat{O}\vec{n}\vec{n}]]] \quad (3.29)$$

3.5 Deriving Angle Dependence in Multipole Decomposition

We begin with a novel expansion of multipole decomposition outlined above to all angles of incidence. To extend this to all angles of incidence, we make the scattering direction, $\vec{n}$, varying as a function of the angle of incidence, as seen below in equation 3.27 and perform a series of inner tensor products and cross products to derive the angle dependence contribution to the reflected field from each multipole. When investigating the reflected field, the scattering

angle, θ will be rotated by -180 degrees relative to the angle of reflection. The relation between the scattering angle in the reflection direction, and the angle of reflection is as follows, $\theta_{scattering} = \theta_{angle\ of\ reflection} - 180^\circ$. The direction of scattering/angle of reflection can be defined as a unit vector that would be associated with the wavevector of the light and is defined below.

$$\vec{n} = \begin{bmatrix} \sin(\theta) \\ 0 \\ \cos(\theta) \end{bmatrix} \quad (3.30)$$

To calculate the angle dependence for the dipoles, we perform a cross product between the dipole vector, $\vec{a}$, and the unit vector $\vec{n}$.

$$\vec{a} \times \vec{n} = \begin{bmatrix} a_y \cos(\theta) \\ a_z \sin(\theta) - a_x \cos(\theta) \\ -a_y \sin(\theta) \end{bmatrix} \quad (3.31)$$

Equation 3.28 is sufficient for the magnetic dipole. To obtain the equations for the electric dipole contribution, we must perform another cross product.

$$\vec{n} \times (\vec{a} \times \vec{n}) = \begin{bmatrix} \cos(\theta)(a_z \sin(\theta) - a_x \cos(\theta)) \\ a_y \\ \sin(\theta)(a_z \sin(\theta) - a_x \cos(\theta)) \end{bmatrix} \quad (3.32)$$

The vectors described in equations 3.28 and 3.29 describe the scattered field

from the magnetic and electric dipoles for unpolarized light. For s-polarized light we only need the middle element in each vector, for p-polarized light we can use either the 1st or 3rd elements, or both, the other elements being zero. For simplicity, we only use the 1st element, as it is the only one valid at normal incidence.

To derive the contribution for the quadrupoles, we perform a tensor inner product between the quadrupole tensor M and the unit vector $\vec{n}$, giving us a vector to perform cross products on.

$$\hat{M}\vec{n} = \begin{pmatrix} M_{11} & M_{12} & M_{13} \\ M_{21} & M_{22} & M_{23} \\ M_{31} & M_{32} & M_{33} \end{pmatrix} \begin{pmatrix} \sin(\theta) \\ 0 \\ \cos(\theta) \end{pmatrix} = \begin{pmatrix} M_{11} \\ M_{21} \\ M_{31} \end{pmatrix} \sin(\theta) + \begin{pmatrix} M_{13} \\ M_{23} \\ M_{33} \end{pmatrix} \cos(\theta) \quad (3.33)$$

$$\hat{M}\vec{n} = \begin{pmatrix} M_{11}\sin(\theta) + M_{13}\cos(\theta) \\ M_{21}\sin(\theta) + M_{23}\cos(\theta) \\ M_{31}\sin(\theta) + M_{33}\cos(\theta) \end{pmatrix} \quad (3.34)$$

To obtain the magnetic quadrupole terms, a cross product between the above and the unit vector is performed.

$$\vec{n} \times (\hat{M}\vec{n}) = \begin{pmatrix} \sin(\theta) \\ 0 \\ \cos(\theta) \end{pmatrix} \times \begin{pmatrix} M_{11}\sin(\theta) + M_{13}\cos(\theta) \\ M_{21}\sin(\theta) + M_{23}\cos(\theta) \\ M_{31}\sin(\theta) + M_{33}\cos(\theta) \end{pmatrix} \quad (3.35)$$

$$\vec{n} \times (\hat{M}\vec{n}) = \begin{pmatrix} M_{21}\sin(\theta) + M_{23}\cos(\theta) \\ -\cos(\theta)(M_{11}\sin(\theta)^2 - M_{33}\cos(\theta)^2) \\ M_{21}\cos(\theta) + M_{23}\sin(\theta) \end{pmatrix} \quad (3.36)$$

Another cross product obtains the electric quadrupole contribution.

$$\vec{n} \times (\vec{n} \times (\hat{M}\vec{n})) = \begin{pmatrix} -\cos(\theta)(M_{11}\sin(\theta)^2 - M_{33}\cos(\theta)^2) \\ (M_{21}\sin(\theta) + M_{23}\cos(\theta)) \\ -\sin(\theta)(M_{11}\cos(\theta)^2 - M_{33}\sin(\theta)^2) \end{pmatrix} \quad (3.37)$$

As with the dipoles, the 1st element is for p-polarized light, and the 2nd element is for s-polarized light.

The derivation follows the same steps as outlined for the octupole with an additional tensor product to reduce the order of the 3rd order octupole tensor, to a 1st order tensor.

$$\hat{O}(\vec{n}\vec{n}) = \begin{pmatrix} \sin(\theta)^2 O_{xxx} + \sin(\theta) O_{zxx} + \cos(\theta)^2 O_{zzx} \\ \sin(\theta)^2 O_{xyx} + \sin(\theta) O_{zyx} + \cos(\theta)^2 O_{zyz} \\ \sin(\theta)^2 O_{xzx} + \sin(\theta) O_{zzx} + \cos(\theta)^2 O_{zzz} \end{pmatrix} \quad (3.38)$$

The magnetic octupole becomes the following.

$$\vec{n} \times \hat{O}(\vec{n}\vec{n}) = \begin{pmatrix} -\cos(\theta)(\sin(\theta)^2 O_{xyx} + \sin(\theta) O_{zyx} + \cos(\theta)^2 O_{zyz}) \\ O_{xzx}(\sin(\theta)^3 + \sin(\theta)^2 \cos(\theta)) + O_{zzx}(\cos(\theta)^3 - \sin(\theta)^2) + O_{zz}\cos(\theta)^3 \sin(\theta)^2 + O_{xxx}\sin(\theta)^2 \cos(\theta) \\ \sin(\theta)(\sin(\theta)^2 O_{xyx} + \sin(\theta) O_{zyx} + \cos(\theta)^2 O_{zyz}) \end{pmatrix} \quad (3.39)$$

And to obtain the electric octupole we perform a final cross product.

$$\vec{n} \times (\vec{n} \times \hat{O}(\vec{n}\vec{n})) = \begin{pmatrix} \cos(\theta)(O_{xzx}(\sin(\theta)^3 + \sin(\theta)^2 \cos(\theta)) + O_{zzx}(\cos(\theta)^3 - \sin(\theta)^2) + O_{zz}\cos(\theta)^3 \sin(\theta)^2 + O_{xxx}\sin(\theta)^2 \cos(\theta)) \\ O_{xzx}(\sin(\theta)^3 + \sin(\theta)^2 \cos(\theta)) + O_{zzx}(\cos(\theta)^3 - \sin(\theta)^2) + O_{zz}\cos(\theta)^3 \sin(\theta)^2 + O_{xxx}\sin(\theta)^2 \cos(\theta) \\ \sin(\theta)(O_{xzx}(\sin(\theta)^3 + \sin(\theta)^2 \cos(\theta)) + O_{zzx}(\cos(\theta)^3 - \sin(\theta)^2) + O_{zz}\cos(\theta)^3 \sin(\theta)^2 + O_{xxx}\sin(\theta)^2 \cos(\theta)) \end{pmatrix} \quad (3.40)$$

To predict the reflection coefficients from these multipoles we use the equation below, using the cross product terms derived earlier to calculate the reflection coefficient in terms of these multipoles [59].

$$r_{s-pol} = \frac{ik_d}{E_0 2S_L \epsilon_0 \epsilon_d} (p_y + \frac{1}{v_d} m_y + \frac{ik_d}{6} Q_y + \frac{ik_d}{2v_d} M_y + \frac{k_d^2}{6} O_y^e + \frac{ik_d^2}{6v_d} O_y^m) \quad (3.41)$$

$$r_{p-pol} = \frac{ik_d}{E_0 2S_L \epsilon_0 \epsilon_d} \left(p_x + \frac{1}{v_d} m_x + \frac{ik_d}{6} Q_x + \frac{ik_d}{2v_d} O_x^e + \frac{k_d^2}{6} O_x^m \right) \quad (3.42)$$

E_0 is the incident electric field, S_L is the surface area exposed to the incident light, ϵ_d is the electric permittivity of the surrounding dielectric and v_d is the speed of light in the surrounding dielectric.

p_a and m_a are the electric and magnetic dipole moments along the a-axis, where $a = x, y, z$.

$$p_a = (\vec{n} \times (\vec{d} \times \vec{n}))_a \quad (3.43) \quad m_a = (\vec{d} \times \vec{n})_a \quad (3.44)$$

where Q_a and M_a are the electric and magnetic quadrupole moments.

$$Q_a = (\vec{n} \times (\vec{n} \times (\hat{M}\vec{n})))_a \quad (3.45) \quad M_a = (\vec{n} \times (\hat{M}\vec{n}))_a \quad (3.46)$$

where O_a^e and O_a^m are the electric and magnetic octupole moments.

$$O_a^e = (\vec{n} \times (\vec{n} \times \hat{O}(\vec{n}\vec{n})))_a \quad (3.47) \quad O_a^m = (\vec{n} \times \hat{O}(\vec{n}\vec{n}))_a \quad (3.48)$$

These derivations, though somewhat cumbersome, help to explain the difference in s-polarized and p-polarized reflection and transmission in all structures, be they thin films or metamaterials. The number of terms for each multipole depends on the polarization of the light and the order of the multipole. At normal incidence the s- and p-polarized terms reduce to single terms that match each other, which in turn matches the Fresnel equations, which predicts

that at normal incidence the two polarizations give the same result when considering reflection from a thin film.

Equations 3.41 and 3.42 have an interesting implications for the contribution of any multipole to the scattered field. It is possible for a multipole to be present, but not contribute to reflection, the reflected field from each pole is calculated via a cross product, which is zero if the direction of the reflected field (angle of reflection) and the vector associated with the multipole are parallel or anti-parallel with each other, then the resulting reflected field is zero.

In the case of the electric dipole, this permits p-polarized light to be perfectly transmitted, while only s-polarized light is reflected. This is the well-known Brewster angle [60], which is commonly associated with surface electric dipoles, whose dipole moment aligns with the direction of the reflected field, suppressing reflection for p-polarized light.

Fascinatingly, our derivations highlight that there is more than one Brewster Angle, each electric and magnetic multipole may theoretically support one or more Brewster angles regardless of polarization, given that all multipoles include at least 1 cross product in their derivations, as shown in our derivations earlier. We will present results on these Brewster angles in Chapter 4.

To characterize thin films that extend to infinity in the plane, we follow the same procedure applied to metamaterials, we define a single unit cell of the infinitely wide thin film, in this case that is a cube with dimensions equivalent to the thickness of the film, this matches previous models used to characterize thin films with multipole decomposition. We compute the scattered field associated with the multipole moments by integrating the current density over

the volume of this cube. Reflection is calculated by dividing this scattered field by the incident field at the center of an empty unit cell. This surface area term arises from the Green Tensor which describes the reflected field from an infinitely large periodic lattice consisting of our cubic unit cells [61].

3.5.1 COMSOL Simulation For Multipole Decomposition and S-parameter

In Figure 3.14 we outline the simulations details for our COMSOL model. In Figure 3.14 (a), the dimensions of the simulations are outlined. We note that it is vital to keep the shape of the thin film as a cube, with dimensions equal to that of the thickness of the thin film modelled, which in our case was 500 nm x 500 nm x 500 nm, this matches the approach by Li et al [17] for their GaP thin film.

Boundary conditions are shown in Figure 3.14 (b), in order to model an infinitely wide thin film, boundary conditions perpendicular to the film surface are set to periodic boundary conditions, Floquet boundaries must be used for non-zero angles of incidence.

Controlling whether the incident light is s- or p-polarization can be done through the port boundary conditions, Figure 3.14 (c) outlines the chosen parameters for s-polarization for Port 1 in Figure 3.14 (b). An s-polarized wave is defined by setting the electric field along the y direction to a value that is not 1, the magnitude does not matter, as long as it is kept constant across different simulations. To change the polarization to p-polarization, we change the input

quantity to magnetic field. The angle of incidence is controlled through the "Elevation angle of incidence" section.

A parameter not typically made clear in literature is the definition of the incident field in absence of the thin film. This is required to calculate reflection for multipole decomposition, which is E_0 in equations 3.41 and 3.42, as it is given as the ratio between the reflection from the multipoles and incident light. Figure 3.14 (d) highlights how it can be done with a point probe. We would like to thank Dr. Pavel D. Terekhov for their assistance in highlighting this issue through a private communication [62].

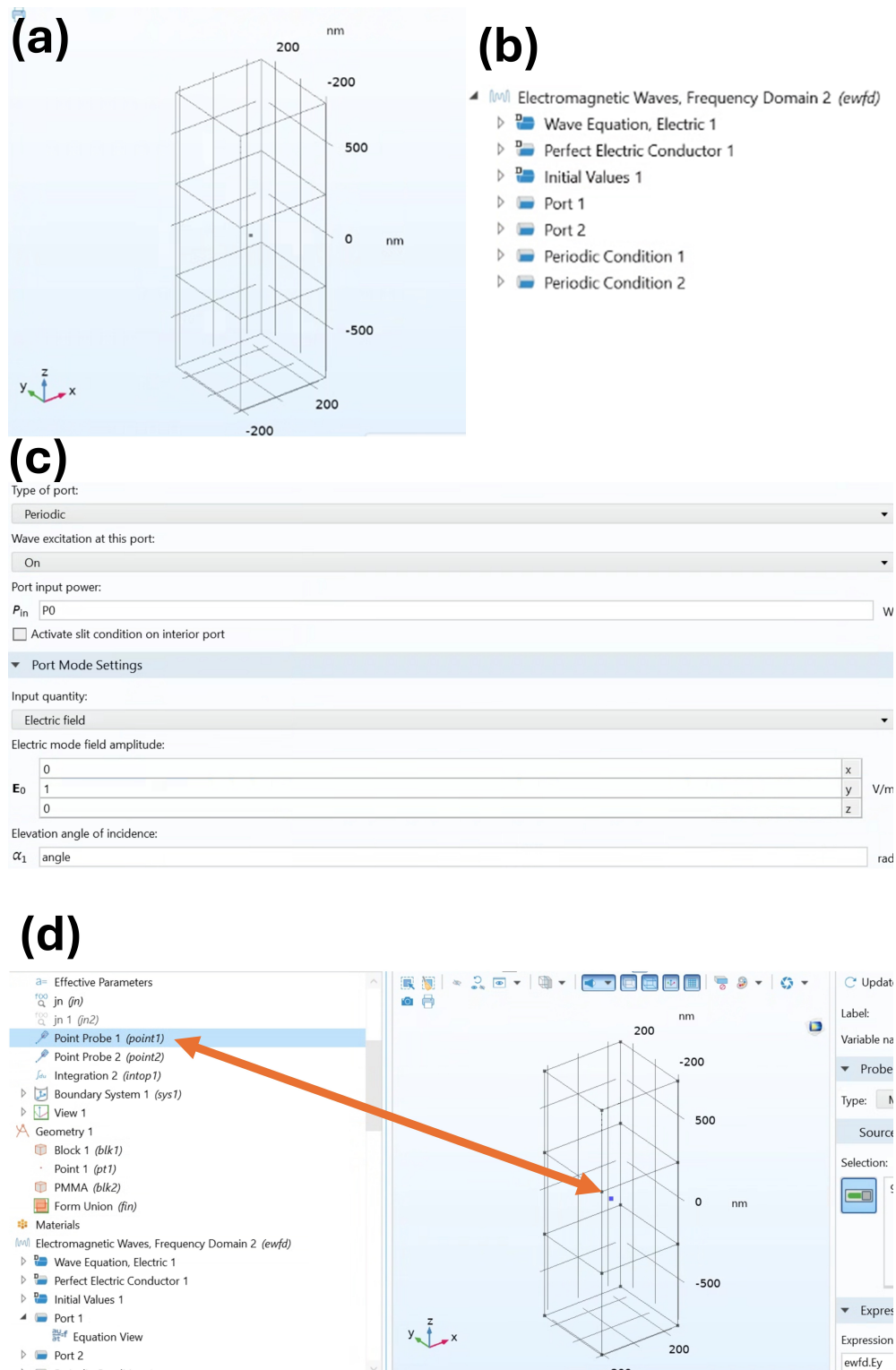


FIGURE 3.14: Images of the COMSOL model used. (a) 3D structure used. (b) Details of boundary conditions used. (c) Details of port 1 used to define the angle and polarization of the incident plane wave. (d) Details of the point probe used to calculate the incident electric field, E_0 .

3.5.2 Lumerical Simulation for Eigenmode Analysis

For modeling the possible eigenmodes in Chapter 5, we employed Lumerical's Finite Difference Eigenmode Solver (FDE), this is a common technique employed in fiber optic cables and waveguides to identify the modes that propagate in those structures. It is also useful for extracting other propagating modes, such as propagating surface plasmons.

Figure 3.15 outlines the steps taken to model this. While we define a 3D structure, the simulation region is a 1D dimensional region, treating the structure as an infinitely wide thin film, the only physical dimension that is defined in the simulation is perpendicular to the surface, i.e the thickness of the films.

The boundary conditions selected are as follows. For the SiO_2 substrate we use Perfectly Matched Boundaries (PML), these act to absorb the radiative mode, however we would recommend selecting a structure or wavelength to minimize this radiative problem, given that PML are difficult to use in eigenmode solvers as they introduce modes confined to the PML BCs that make it difficult to identify physical modes, requiring the user to increase the number of trial modes, and increasing computation time.

For the air layer, we use metallic boundary conditions that are sufficiently far away from the PMMA-Air interface, to avoid the evanescent wave from interacting with this BC, we suggest at least 2000 nm separation between interface and BC.

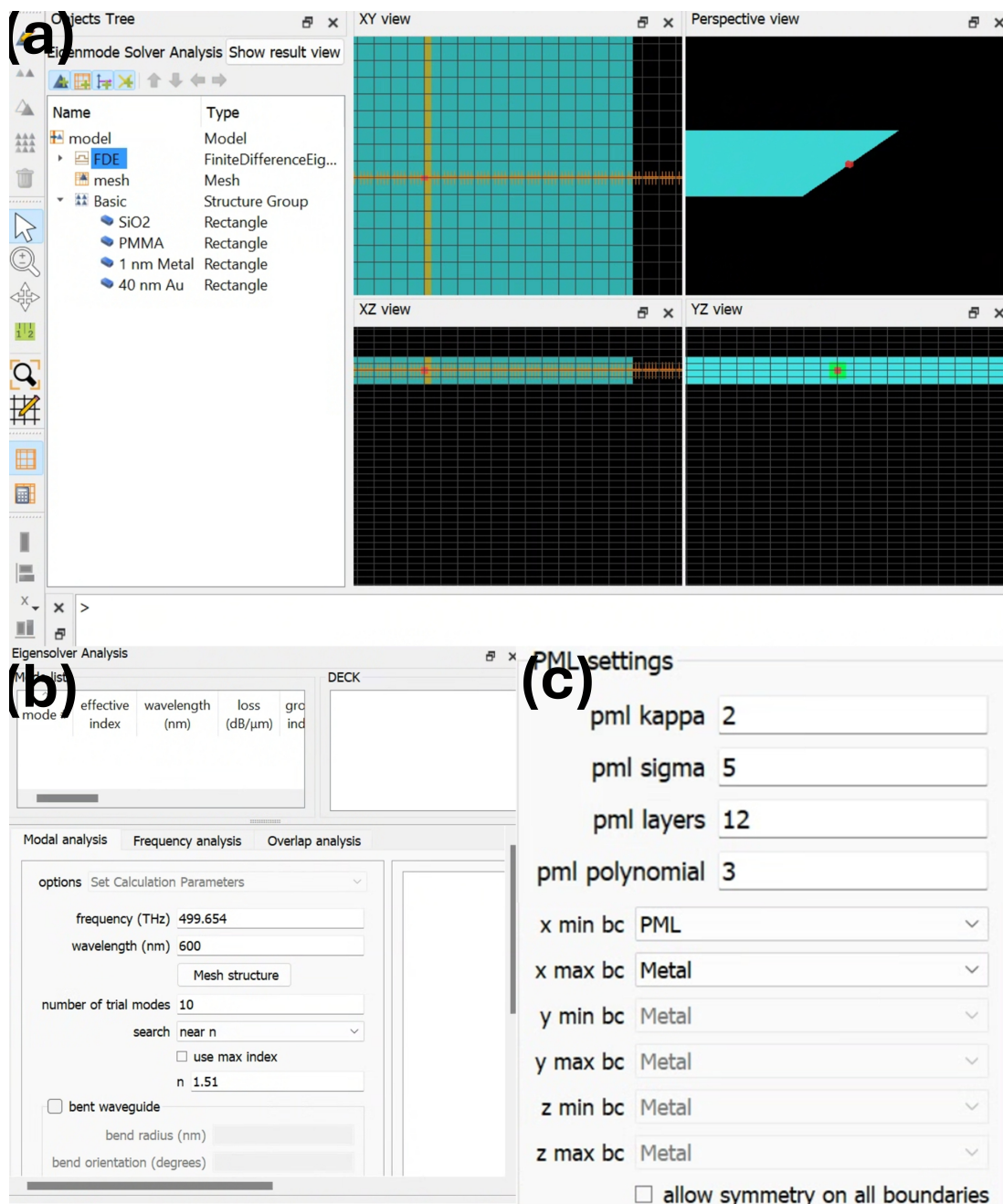


FIGURE 3.15: Images of Lumerical model used for Chapter 5. (a) Details of structure, meshing used for simulation. (b) Example of the modal analysis tab used to find eigenmodes at specific frequencies and effective indices. (c) Settings of the boundary conditions and the settings of the PML boundary conditions used.

3.6 Summary

To summarize this chapter, we outlined the samples we selected for our studies, that being a collection of thin films of SiN and SiO₂, and a multilayers structure of Ti-Au-PMMA.

Ellipsometry measurements highlighted a difference between our SiN sample, and the expected sample. In particular our sample's index is higher than expected for stoichiometric Si₃N₄, while the thickness of the film is roughly 50 nm thinner than expected.

Steps to fabricate the Ti-Au-PMMA sample are outlined, and detailed methods to identify optimal PMMA deposition are discussed, furthermore we identified that our PMMA thickness was roughly 1200 nm through ellipsometry measurements.

Design of experiments to study these thin films were then outlined, two separate approaches were discussed. For SiN and SiO₂ thin films, an experiment to measure the frequency and angle dependent transmission was devised. However we only present reflection data, as it is possible to calculate the reflection from transmission in the absence of absorption as the sum of reflection and transmission is one. It is safe to assume absorption is zero given that ellipsometry showed that absorption is negligible across the wavelengths we intend to study. For our multilayered structure of Ti-Au-PMMA, we devised a new prism coupling scheme to measure the frequency and angle dependent reflection. Both methods rely on the measurement of the reflected/transmitted intensity of the light, which is related to squared amplitude of the complex

electric fields, however we can not directly measure these electric fields.

Techniques to verify these experiments were outlined, we approached modeling of reflection with a threefold approach, a more standard transfer matrix or S-parameter method is applied to model transmission or reflection. Multipole decomposition is the third model, which allows us to study the electric and magnetic response of our thin films in terms of electric and magnetic multipoles.

The three models and measurement correspond to a different layer of physics, with each we reveal deeper layers and more phenomena. In our measurements we measure the intensity of the reflected/transmitted fields. The S-parameter model describes those intensities in terms of complex electric fields. Multipole decomposition describes those complex electric fields in terms of electric and magnetic multipoles.

Additionally, practical implementation of COMSOL for S-parameter and multipole decomposition models is outlined. Eigenmode analysis is outlined to identify resonant modes in our Ti-Au-PMMA sample in Chapter 5.

Chapter 4

Electric and Magnetic Multipoles in Thin Films at Optical Frequencies

4.1 Introduction

Having discussed our methodologies for both measurements and models in Chapter 3, we now move to discuss our study of our SiN and SiO₂ thin films whose experiment was outline in Section 3.2.1 and models outline in Section 3.3.

As highlighted in the introduction, the possibility of thin films supporting magnetic multipoles and hence, a magnetic response is the focus of this Chapter. In the previous chapter, our development of an angle-dependent multipole decomposition model to predict reflection in terms of electric and magnetic multipoles and having built a set up to measure this reflection, we move to verify the validity of our model, and understand the properties of thin films in terms of these electric and magnetic multipoles.

Thin films play a major role in applications, from interferometers [63], lasers [64], micro-combs [65], optical absorbers [66] and structural color [67]. Creating a fundamental model to understand the optical electric and magnetic properties of thin films provides us with new perspective on these everyday applications, with the possibility of developing a better understanding of thin films and proposing new applications from those everyday devices.

4.2 Measured and Modeled Reflection of Sub-wavelength Si_3N_4 Thin Films

In order to test the validity of our multipole model in predicting the optical properties of thin films, we chose to measure the transmission through a 500 nm thick Si_3N_4 thin film suspended in air, as shown in Figure 4.1.

We begin with Figure 4.1, comparing the measured reflection to our Multipole model and S-parameter calculations from COMSOL for 500 nm of Si_3N_4 suspended in air. The S-parameter technique relies on extracting the complex electric fields at the input and output ports of a COMSOL simulation to calculate the reflection from the Si_3N_4 , providing an excellent standard to test our multipole model against.

Comparing s-polarized reflection in Figure 4.1 (a), (c) and (e) we see that the two models agree perfectly while partially disagreeing with the measured reflection. The mismatch in between experiment and model is the result of different thicknesses and refractive indices, ellipsometry measurements showed that the true thickness of the film was roughly 455 nm, with a higher refractive index than the modelled refractive index sourced from Luke et al [24].

Furthermore, the plotting of the 1st order Fabry Perot mode in Figure 4.1 (a) using the measured thickness and refractive index overlaps well with the measured wavelengths and angles where zero reflection is observed, highlighting how our sample can behave as an ideal thin film supporting Fabry Perot modes.

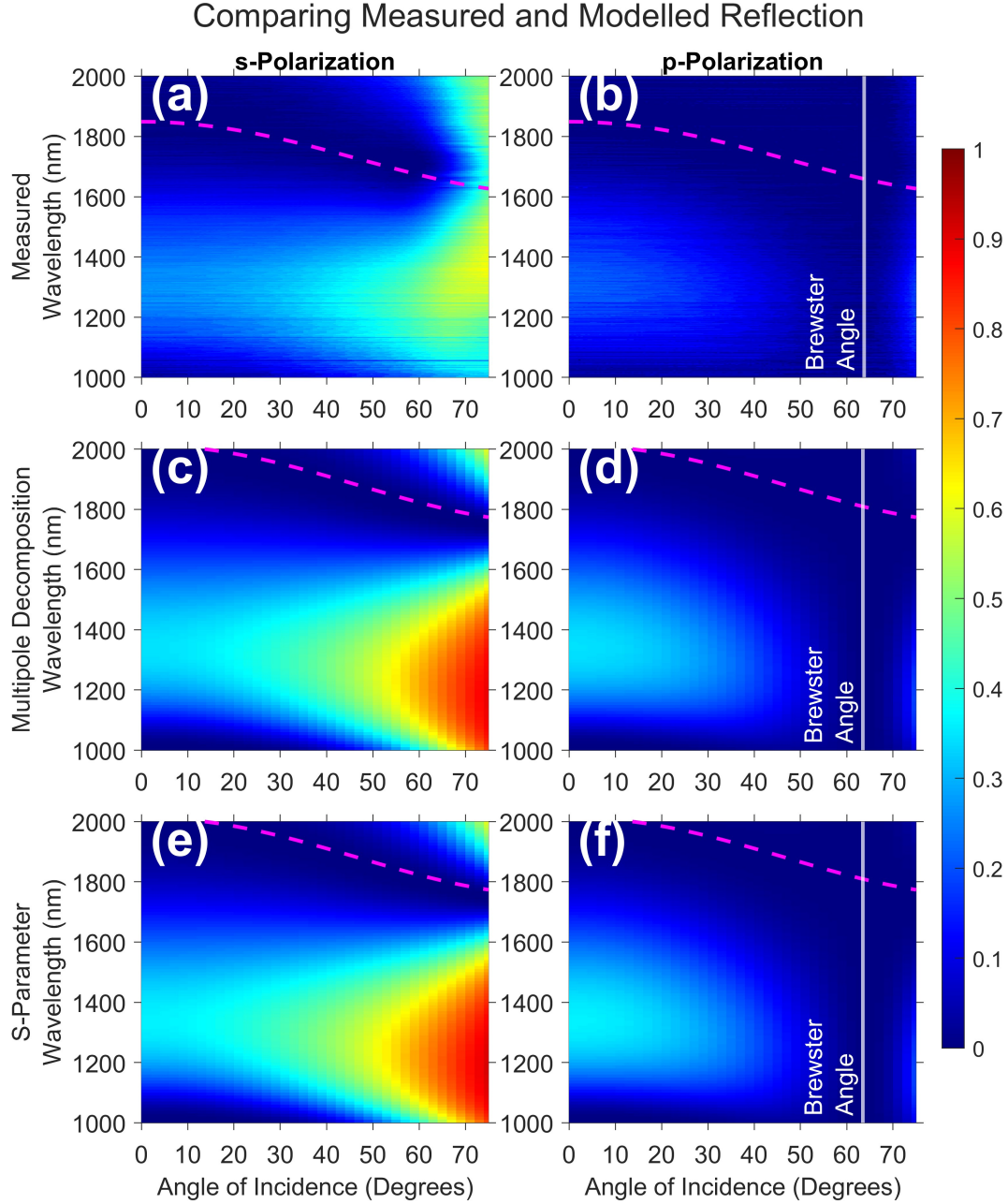


FIGURE 4.1: Comparison between measured and modeled reflection model with multipole decomposition for s and p-polarized light for wavelengths in the range of 1000-2000 nm. Dashed magenta line marks the 1st order Fabry Perot mode. (a) Measured reflection for s-polarized light. (b) Measured Reflection for p-polarized light. (c) Multipole reflection for s-polarized light. (d) Multipole reflection for p-polarized light. (e) S-parameter reflection for s-polarized light. (f) S-parameter reflection for p-polarized light.

The p-polarized measurements and models in Figure 4.1 (b), (d) and (f) show the same result, the two models agree excellently while the measurement shows some disagreement. However we do note that the measured position of the Brewster angle is very similar to predictions from the multipole model and the S-parameter model, showing that our multipole model is capable of capturing the physics associated with the reflectivity being zero at the Brewster Angle.

In Figure 4.2, we plot the same data as Figure 4.1, investigating the reflection versus wavelength for three different angles of incidence, 0, 30 and 60 degrees. We note that our theoretical models predict the positions of the Fabry Perot modes, plotted with dashed magenta lines, to be at longer wavelengths than observed in the measurements, which we attribute to differences in refractive indices and sample thicknesses between measurement and models.

Both experiment and theory show that the resonant wavelengths of the Fabry Perot modes shifts to shorter wavelengths with increasing angle of incidence. This is shown clearest in the s-polarized results in Figure 4.2 (a), (c), (e) where the Brewster angle is not present. The presence of the Brewster angle at 62.5 degrees makes observation of the Fabry Perot modes difficult in Figure 4.2 (f).

In Figure 4.3, we investigate the reflection vs the angle of incidence for three wavelengths, 1300 nm, 1500 nm and 1700 nm. The general trend of increasing reflection with angle of incidence is captured by both the multipole model and the S-parameter model, however the rate at which that increase occurs does not match very well, nor is it consistent. For example, Figure 4.3 (a), our models under-predict the reflection, while, in Figure 4.3 (c), they over-predict the reflection. However our models are capable of capturing the Brewster Angle at roughly 60 degrees, as seen clearly in Figure 4.3 (f).

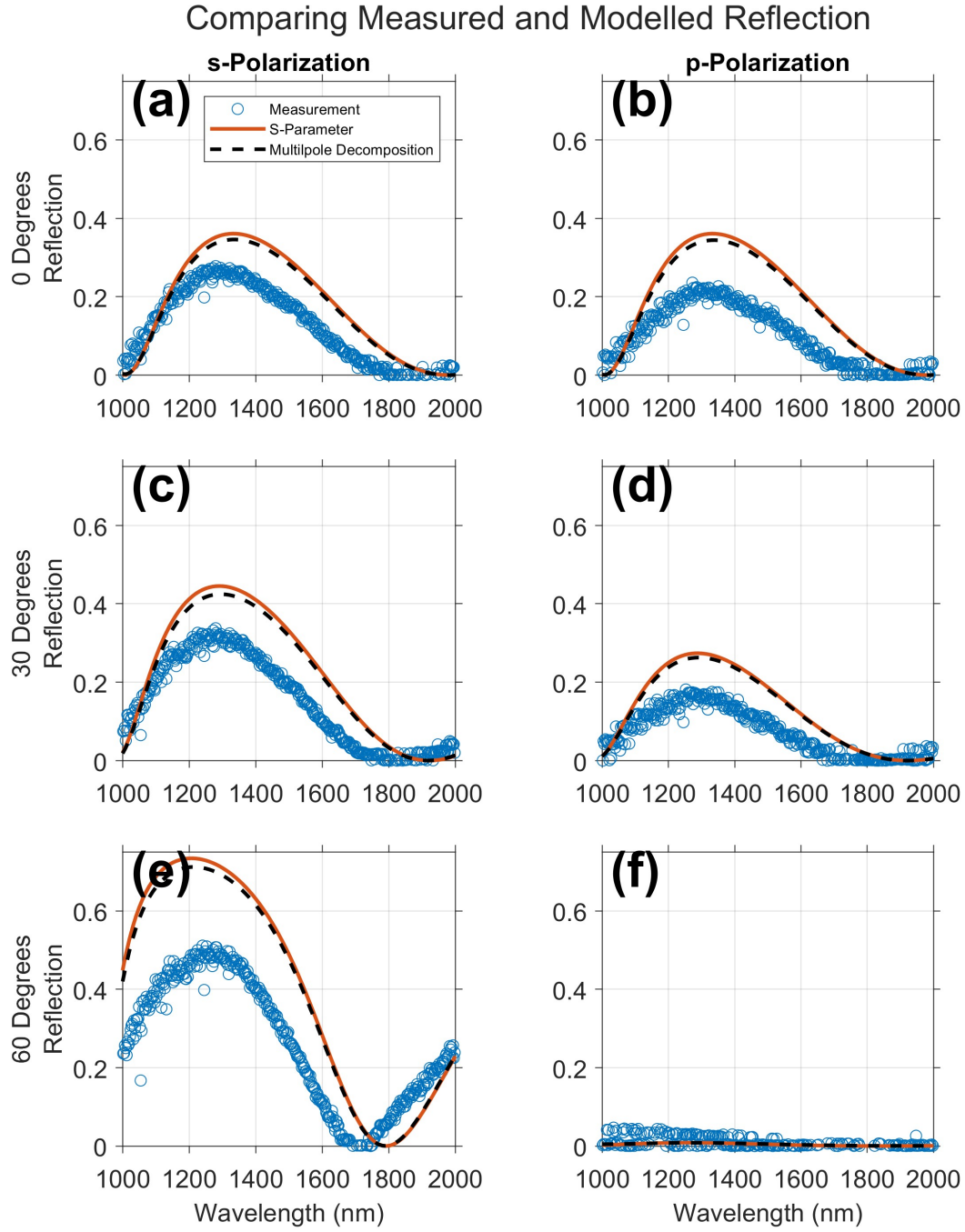


FIGURE 4.2: Plots of reflection vs wavelength at 0, 30 and 60 degrees. Data is taken from that presented in Figure 4.1

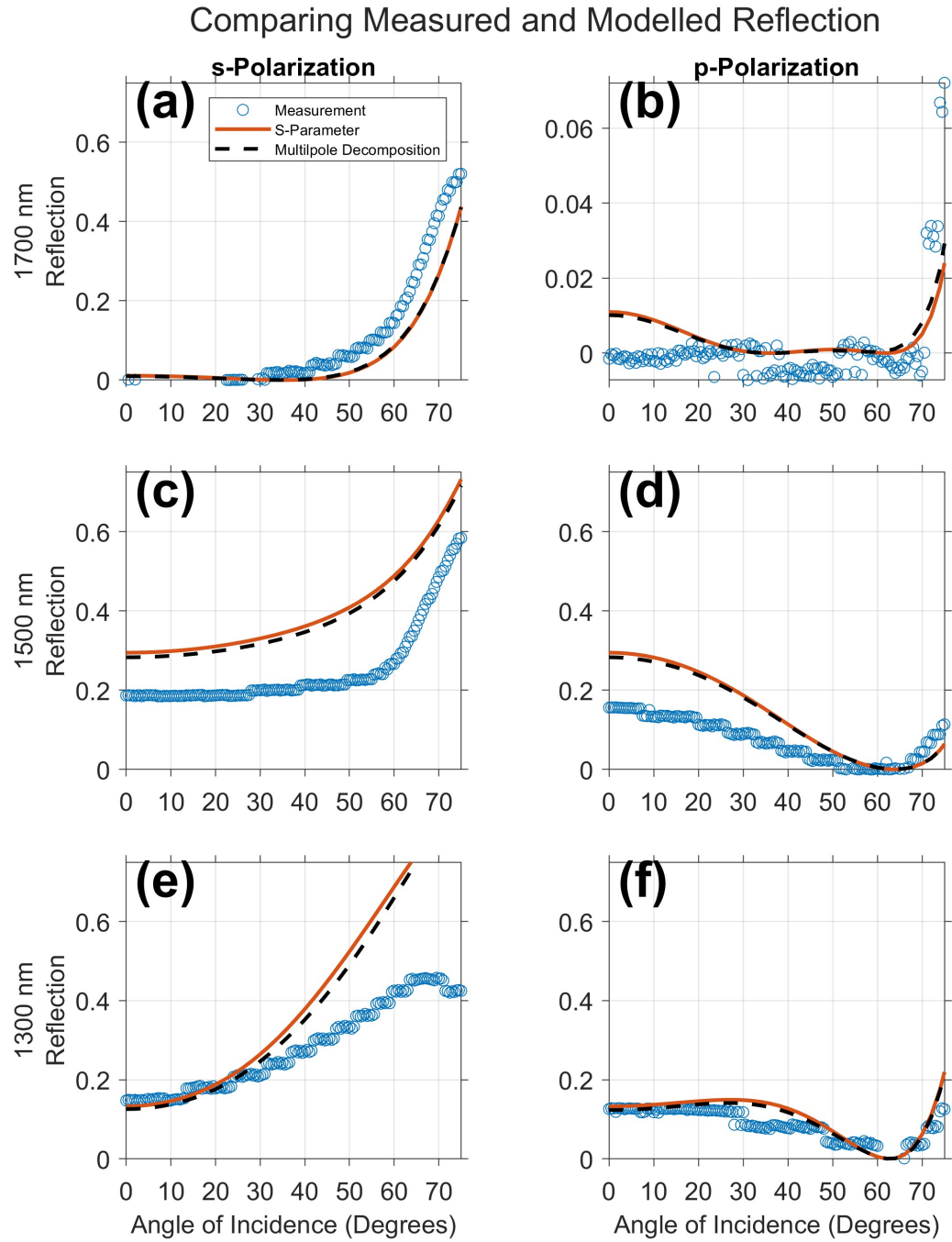


FIGURE 4.3: Plots of Reflection vs wavelength at 0,30 and 60 degrees. Data is take from that presented in Figure 4.1

4.2.1 Role of Multipoles in 1st and 2nd Order Fabry Perot Modes

We have verified the validity of our multipole model in Figures 4.1, 4.2 and 4.3 through comparison to measurements and the S-parameter technique. Mismatches are the result of the experimentally studied film being thinner than expected, and having a higher refractive index than in the model. Our multipole model is capable of reproducing the expected angle dependence of Fabry Perot modes in thin films. We can now further explore the physics behind these Fabry Perot modes in terms of our multipole model, by investigating how each multipole contributes to the overall reflection investigated in Figures 4.1, 4.2 and 4.3.

The following equation describes either s- or p-polarized reflection as linear combinations of electric/magnetic dipoles, quadrupoles and octopoles. In order to more easily understand the role of these multipoles in the reflection of thin films, one may use the phasor representation, given that the reflection coefficient is a complex number.

$$r_{multipole} = |r_{multipole}|e^{-i\phi} \quad (4.1)$$

$$\phi = \tan\left(\frac{re(r)}{im(r)}\right) \quad (4.2)$$

The magnitude of the multipole, $|r_{multipole}|$ indicates the degree to which the multipole contributes to the total reflection at a specific angle and wavelength.

ϕ is the phase of the reflection coefficient, which gives us useful information on how the phase response of the incident electric field is shifted by each multipole, which is important when understanding the constructive and destructive interference that occurs in thin films in terms of multipoles.

Another important consideration when dealing with the phase of multipoles is the parity of the multipoles. An excellent understanding of these parities has been explored in nanophotonics, when investigating the reflection and transmission of subwavelength structures [68, 69, 70], we summaries these observations and discuss adjustments to phase to account for this parity in table 4.1 below.

Even Parity	Odd Parity
Electric Dipole (ED)	Magnetic Dipole (MD)
Magnetic Quadrupole (MQ)	Electric Quadrupole (EQ)
Electric Octupole (EO)	Magnetic Octupole (MO)

TABLE 4.1: Parity of multipoles studied in this work as explained by [68, 69, 70].

A change of parity occurs when we change order (dipole, quadruple, etc) or when we change from electric to magnetic multipoles, if both change, parity is preserved.

The origin of these parity rules is seen in the positive/negative signs of the derived reflection coefficients/scattered field equations seen in equations 3.31-3.40 and originate from two possible sources, 1. The direction of the scattered field $\mathbf{n}$, which is a negative value in the reflection direction, 2. Cross products

may introduce negative/positive signs. We note that errors in the sign will introduce unphysical π phase shifts that will appear as deviations from these well-known parity rules.

Multipoles of same parity will constructively interfere with each other for both the reflected and transmitted direction. Multipoles of opposite parity will constructively interfere with each other in the transmitted direction and destructively interfere in the reflected direction.

For simplicity we focus on the phase of reflection coefficients. For reflection coefficients, constructive interference, which is a $0, 2\pi, 4\pi$, etc phase difference results in perfect reflection, while destructive interference, $1\pi, 3\pi, 5\pi$ etc results in zero reflection as light is perfectly transmitted.

In Figure 4.4 we plot the phase of the reflection coefficients for the electric dipole, magnetic dipole, and electric quadrupole versus wavelength as they are the only ones that significantly contribute to the reflected field in Figures 4.7 and 4.8.

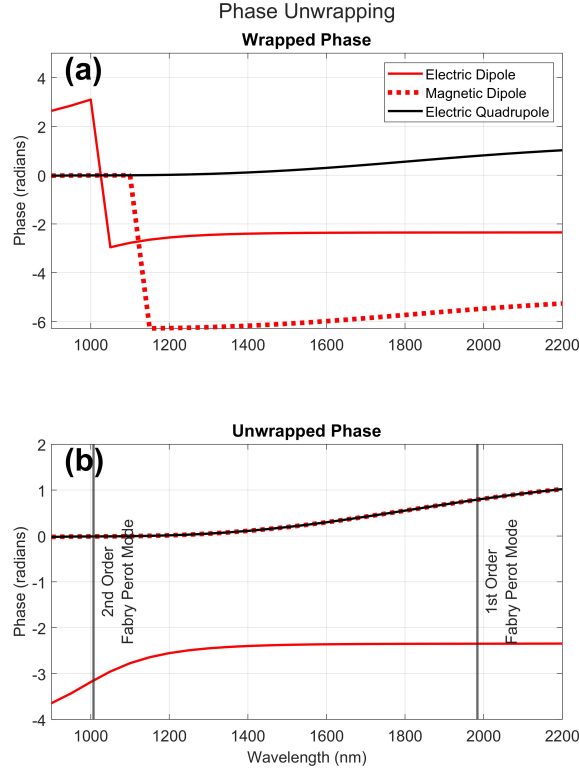


FIGURE 4.4: Phase of p-polarized electric dipole, magnetic dipole, and electric quadrupole at normal incidence across the NIR region. Phase unwrapping is performed by subtracting 2π from the phase.

Given that the trigonometric function, tangent, is only uniquely defined between 0 and 2π , one may observe 2π shifts in phase, we perform phase unwrapping in Figure 4.4 (b) by removing these 2π shifts at roughly 1000 nm, this procedure has been performed in all following datasets involving phase.

Figure 4.5 further explores the role of phase in the 1st order Fabry Perot Mode, we match the analytical prediction of the FP mode to the reflection minima observed in the S-parameter and multipole models in Figure 4.5 (a). Figure 4.5 (b) and (c) shows that the electric and magnetic dipoles, with the electric quadrupole are responsible for a non-zero reflection coefficient at the FP Perot

mode.

In Figure 4.5 (d) we see that there is complete destructive interference between even and odd parity multipoles, which produces zero reflection. The electric dipole is destructively interfering with the magnetic dipole and electric quadrupole, which are in phase. We also note that the sum of reflection coefficients of the magnetic dipole and electric quadrupole is exactly equal to the reflection coefficient of the electric dipole.

The identification of destructive interference at the Fabry Perot resonance does not match that made by Li et al [17], this is a matter of convention, Li et al dealt with the phase of the transmitted/reflected field rather than the phase of the reflection coefficients. As highlighted by Table 4.1, the parity of the multipoles will change the phase of the reflection coefficients, not the phase of the fields. If we were to examine the phase of the reflected fields, we would obtain the same result as they did.

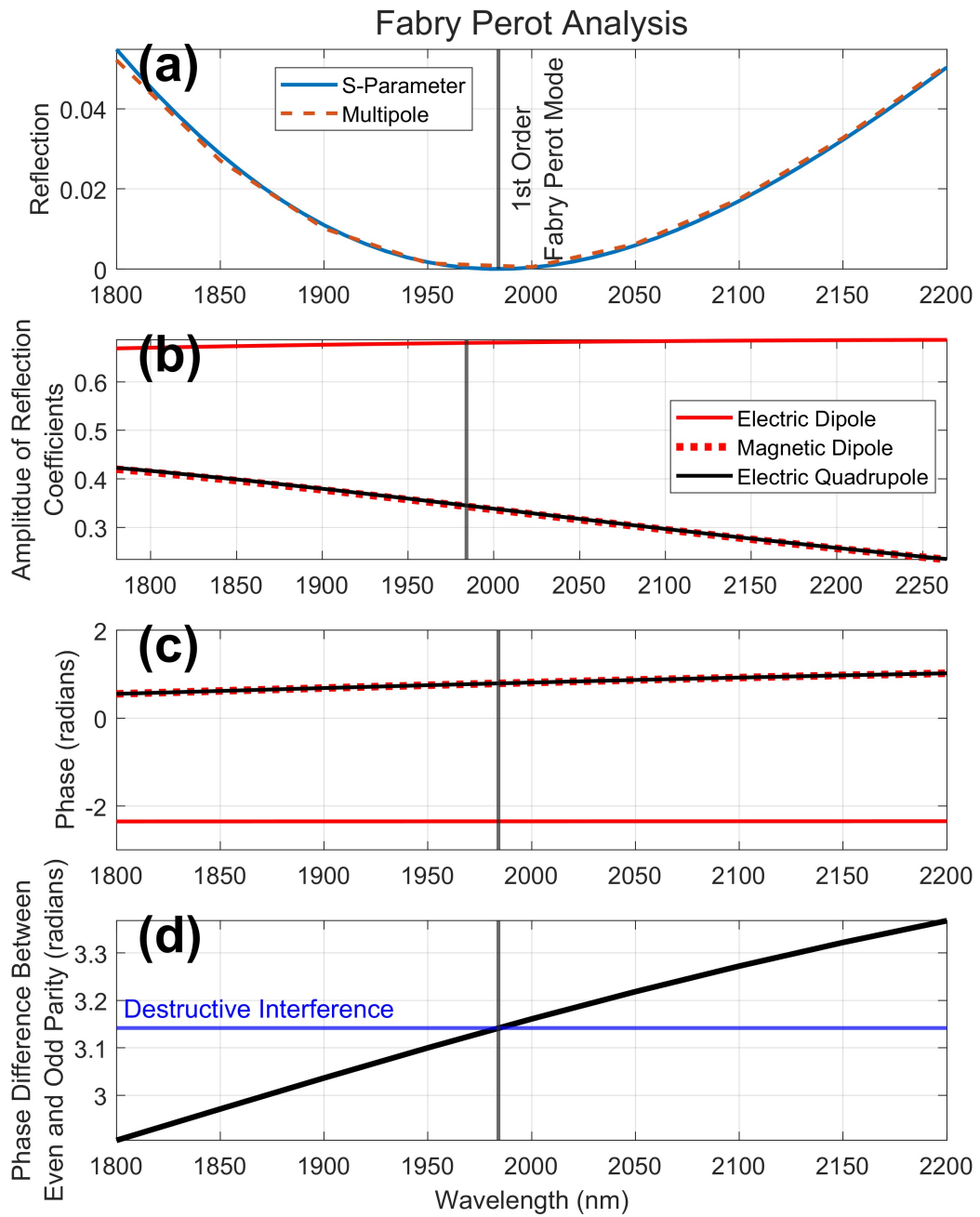


FIGURE 4.5: Analysis of 1st order Fabry Perot mode at normal incidence. (a) Reflection from S-parameter and multipole models. (b) Amplitude of reflection coefficients of individual multipoles. (c) Phase of individual multipoles. (d) Phase difference between even and odd parity multipoles (MD, EQ).

We repeat the analysis performed in Figure 4.5 at a non-zero angle of incidence of 50 degrees in Figure 4.6 for both s-polarized mode in Figure 4.6 (a), (c), (e), (g) and p-polarized modes in Figure 4.6 (b), (d), (f), (h).

We note that the position of the FP mode has shifted to a wavelength of 1800 nm, with the s-polarized mode being narrower than the p-polarized mode, which is a result of it's proximity to the Brewster angle.

The s-polarized FP mode behaves similarly to the FP mode at normal incidence, with the same contribution from ED, MD and EQ as well as interference effects.

However the p-polarized FP differs from it's s-polarized counterpart. In Figure 4.6 (d), the EQ contributes the most to the reflection coefficient, then the MQ and the ED contributes the least, however destructive interference between even and odd parity required to see the Fabry Perot mode still occurs in Figure 4.6 (h).

We suggest that the difference in multipoles between polarizations is the result of the proximity of the p-polarized mode to the Brewster angle near 60 degrees, and should be studied independently of the Fabry Perot mode to understand how these multipoles give rise to the Brewster angle, which we study in the following section.

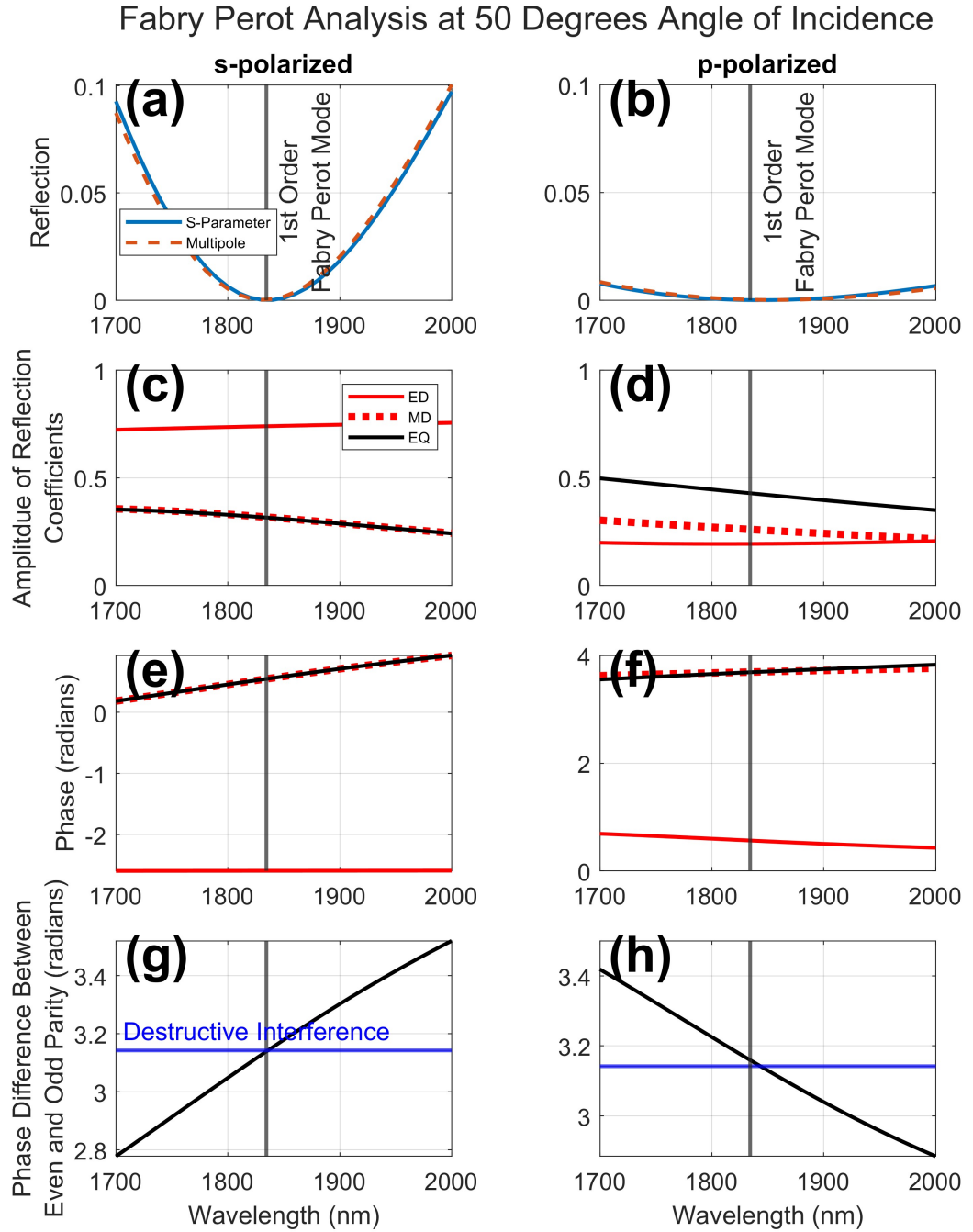


FIGURE 4.6: Analysis of 1st order Fabry Perot mode at 50 degrees angle of incidence, for s and p-polarizations.

In Figure 4.7 and 4.8 we plot the magnitude of the reflection coefficients for each multipole and polarization. The existence of a magnetic dipole in thin

films has been shown in other work at normal incidence for GaP thin films [17], however the angular dependence of this magnetic dipole has not been discussed. Magnetic dipoles are typically discussed in sub-wavelength structures, not sub wavelength films and it is commonly stated that only metamaterials with periodic arrays or sub-wavelength structures can support magnetic dipoles [8, 2, 71].

In Figure 4.8, the magnitude of the s-polarized reflection coefficients are shown. We see that the result diverge from Figure 4.7 at non-normal angles of incidence. This can be explained from the derivation of the angular terms shown in previous sections, which only agree at normal incidence. The electric dipole is the most dominant at longer wavelengths, followed the magnetic dipole and electric quadrupole which are equal in magnitude regardless of wavelength or angle.

We notice that magnetic dipole contribution is strongest in regions of low reflection in Figure 4.8 (b), and it's magnitude matches that of the electric quadrupole, which has been observed at normal incidences [17] .

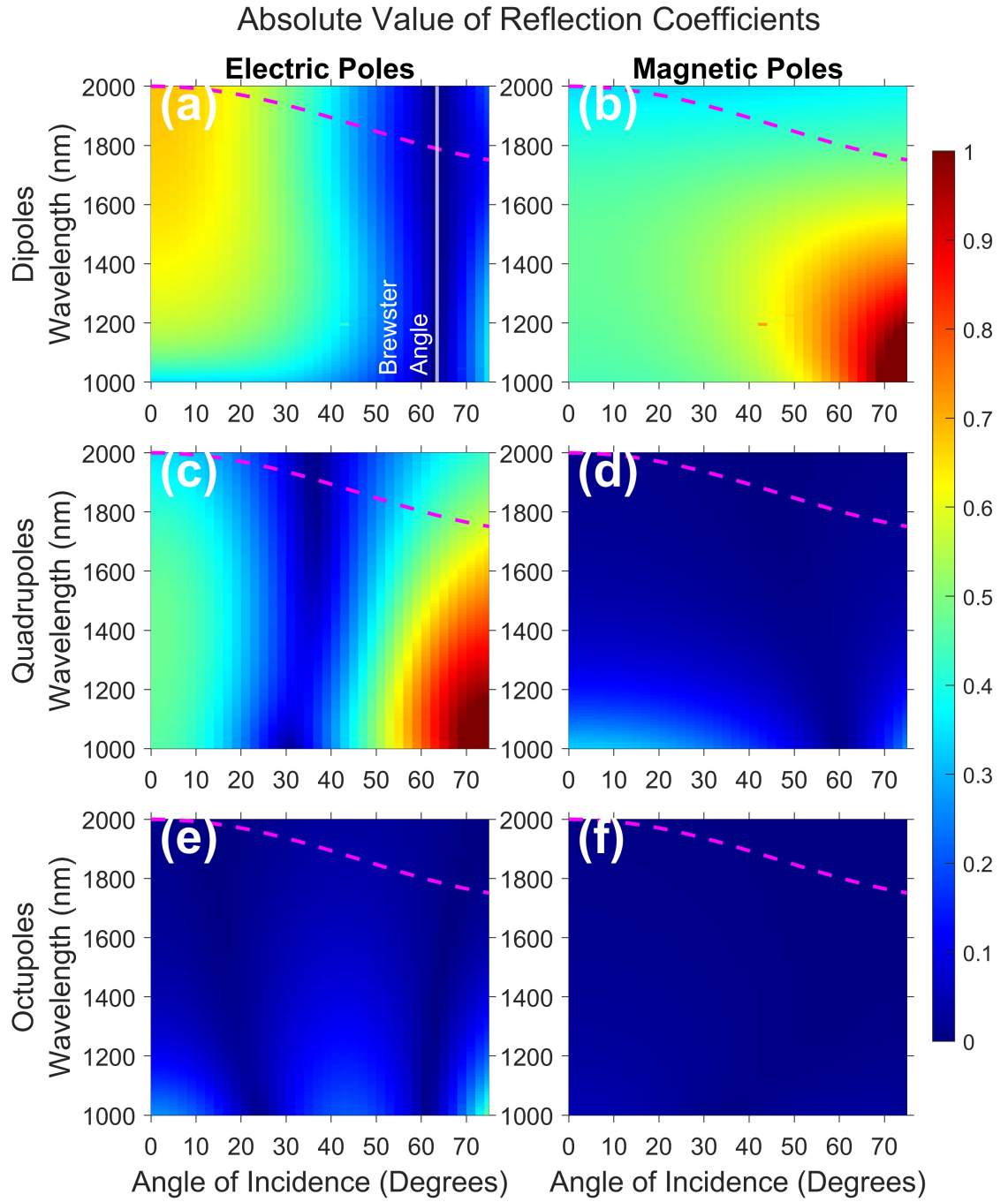


FIGURE 4.7: Amplitude of the p-polarized reflection coefficients for individual multipoles in the NIR region. (a), (c), (e) Reflection from electric dipole, quadrupole and octupole respectively. (b), (d), (f) Reflection from magnetic dipole, quadrupole and octupole respectively.

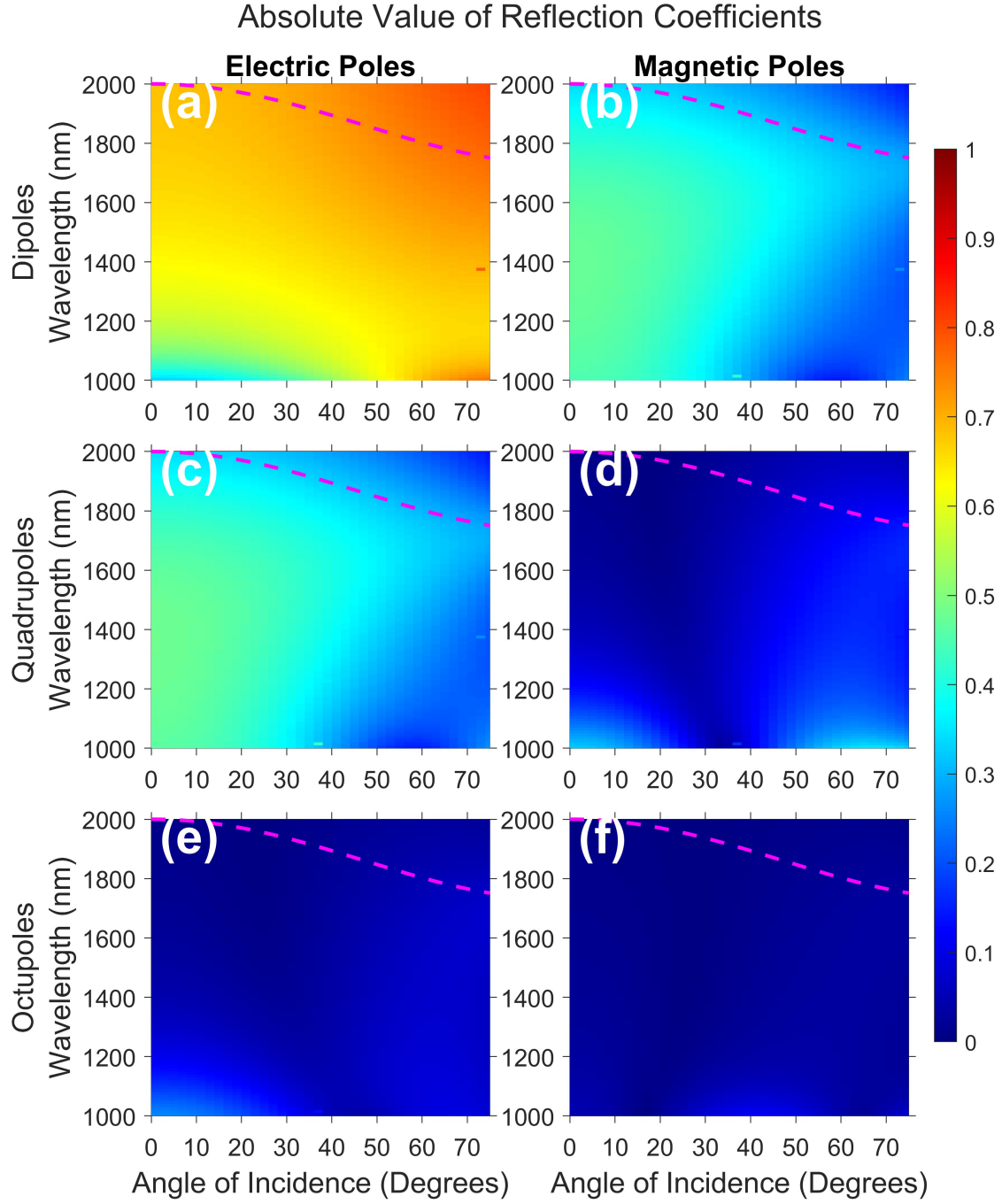


FIGURE 4.8: Amplitude of the s-polarized reflection coefficients for individual multipoles in the NIR region. (a), (c), (e) Reflection from electric dipole, quadrupole and octupole respectively. (b), (d), (f) Reflection from magnetic dipole, quadrupole and octupole respectively.

4.2.2 Role of Multipoles in the Brewster Angle

To explore the physics of the Brewster effect independently of the Fabry Perot modes, we study the multipoles at 1500 nm for varying angles of incidence in Figure 4.8, similar to the approach in Figure 4.5 and 4.6. In Figure 4.9 (a), the Brewster angle is observed at 63.1 degrees, Figure 4.9 (b) shows that the amplitude of the electric dipole reflection coefficient is zero, while the magnetic dipole and electric quadrupole reflection coefficients are equally contributing to the total reflection.

The electric dipole not contributing to reflection is expected, as it is well discussed that the Brewster angle is the result of the electric dipole moment and reflection direction aligning, resulting in the cross product between the two becoming zero and therefore not contribution to the reflection. Similar observations have been made in dielectric metamaterials [72], our work is the first observation of this with multipole decomposition in thin films. A further indication of this Brewster angle is seen in Figure 4.9 (c), where a π phase shift is seen in the vicinity of the Brewster angle.

However, the presence of the magnetic dipole and electric quadrupole should result in non-zero reflection at the Brewster angle. This issue is resolved in Figure 4.9 (c) and (d) as we see that the magnetic dipole and electric quadrupole are undergo complete destructive interference at the Brewster angle as the phase difference between them is π .

We note an apparent similarity in this result to the Kerker effect, where sub-wavelength nanoparticles perfectly transit light, or suppress back-scattering,

is only observed when the electric permittivity and magnetic permeability are equal to each other, or in other words the magnitude of scattering coefficients of the electric and magnetic multipoles are equal to each other [73]. Such a relation between the Kerker effect and the Brewster effect was proposed to exist in dielectric metamaterials [72].

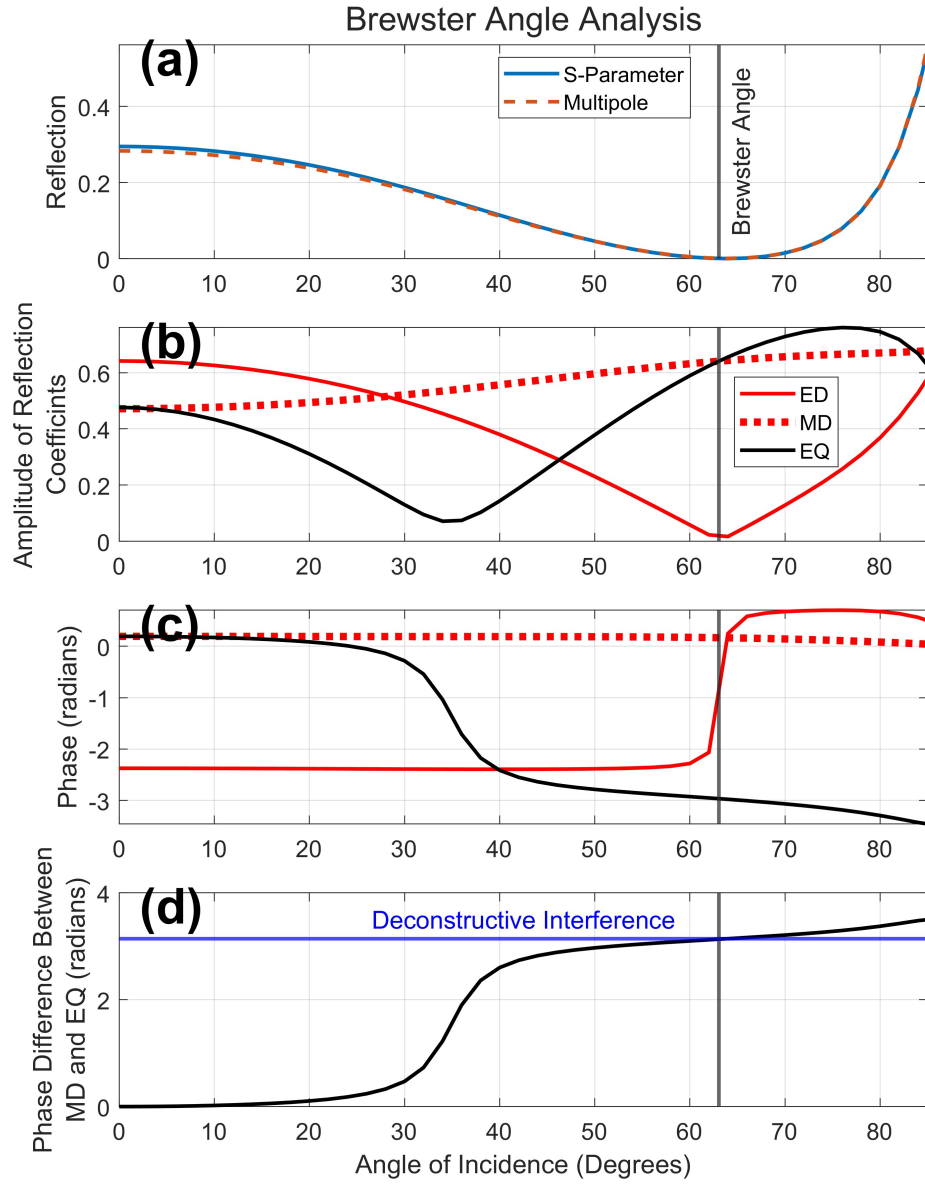


FIGURE 4.9: Phase of electric dipole, magnetic dipole, and electric quadrupole at 1500 nm for p-polarized light to explore the Brewster angle behavior

However, the electric and magnetic scattering coefficients are not equal to each other in our case, it is the cross products of the electric and magnetic scattering coefficients with the direction reflection that are equal to each other. The electric dipole still has a non-zero scattering coefficient at the Brewster angle, but the cross product between the angle of reflection and scattering coefficient is zero. The electric dipole would only have a non-zero scattering coefficient if there was no light in the film, which is at odds with the Brewster angle permitting the perfect transmission of p-polarized light into the film.

To our knowledge, the Brewster effect has only been understood in terms of the electric dipole aligning with the angle of reflection. However, our multipole model reveals a second, perviously unobserved requirement for the Brewster angle, the destructive interference between electric and magnetic multipoles in the reflection direction. Given that these multipoles cancel each other to enable zero reflection at the Brewster angle, only considering the electric dipole in the Brewster effect gives a correct result but is a physically incomplete picture.

This can be seen from Figure 4.10, modeling only the electric dipole produces the Brewster angle as seen with the blue dashed line, adding the electric quadrupole, the green dashed line, removes the Brewster Angle. It is only upon adding the magnetic dipole that the Brewster angle re-appears, with a dramatic improvement with overall agreement between the multipole model and the S-parameter model, as seen with the black dashed line and the red line respectively.

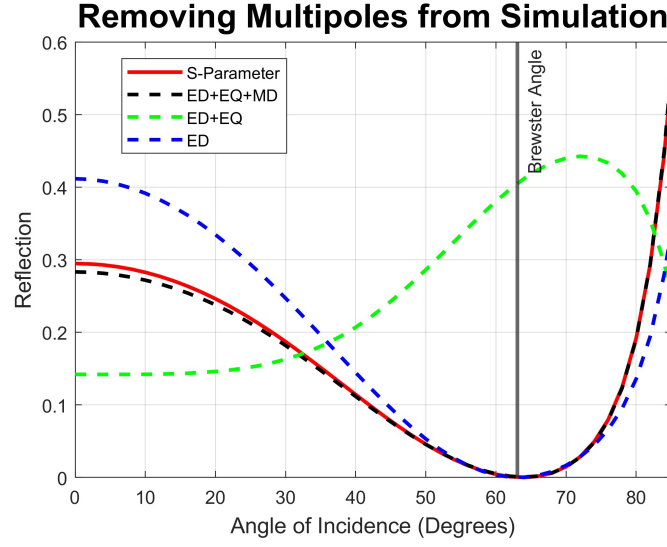


FIGURE 4.10: Demonstration of the importance of the magnetic dipole and electric quadrupole response to the existence of the Brewster Angle.

If we use the current definition of the Brewster Angle, that the cross product between scattering angle and multipole moment is zero, as the two vectors align, resulting in no reflection, then the Brewster Angle could occur for higher order multipoles. We observe that other multipoles will not reflect light, as seen in Figures 4.7 and 4.8. These regions of zero reflection result from alignment of scattering angle with the higher order multipole moments.

To demonstrate this more clearly, we explore the role of multipoles at 1000 nm with varying angle in Figure 4.11 for both s- and p-polarizations, the features of these Brewster angles arising from higher order multipoles can be seen with zero values in the amplitude of their reflection coefficients, as seen in Figure 4.11 (c) and (d), or with the π phase shift seen in Figure 4.11 (e) and (f), which was used to identify the Brewster angle in Figure 4.9. It is important to note that these Brewster angles exist for both polarizations, but the angle they occur

at varies with polarization.

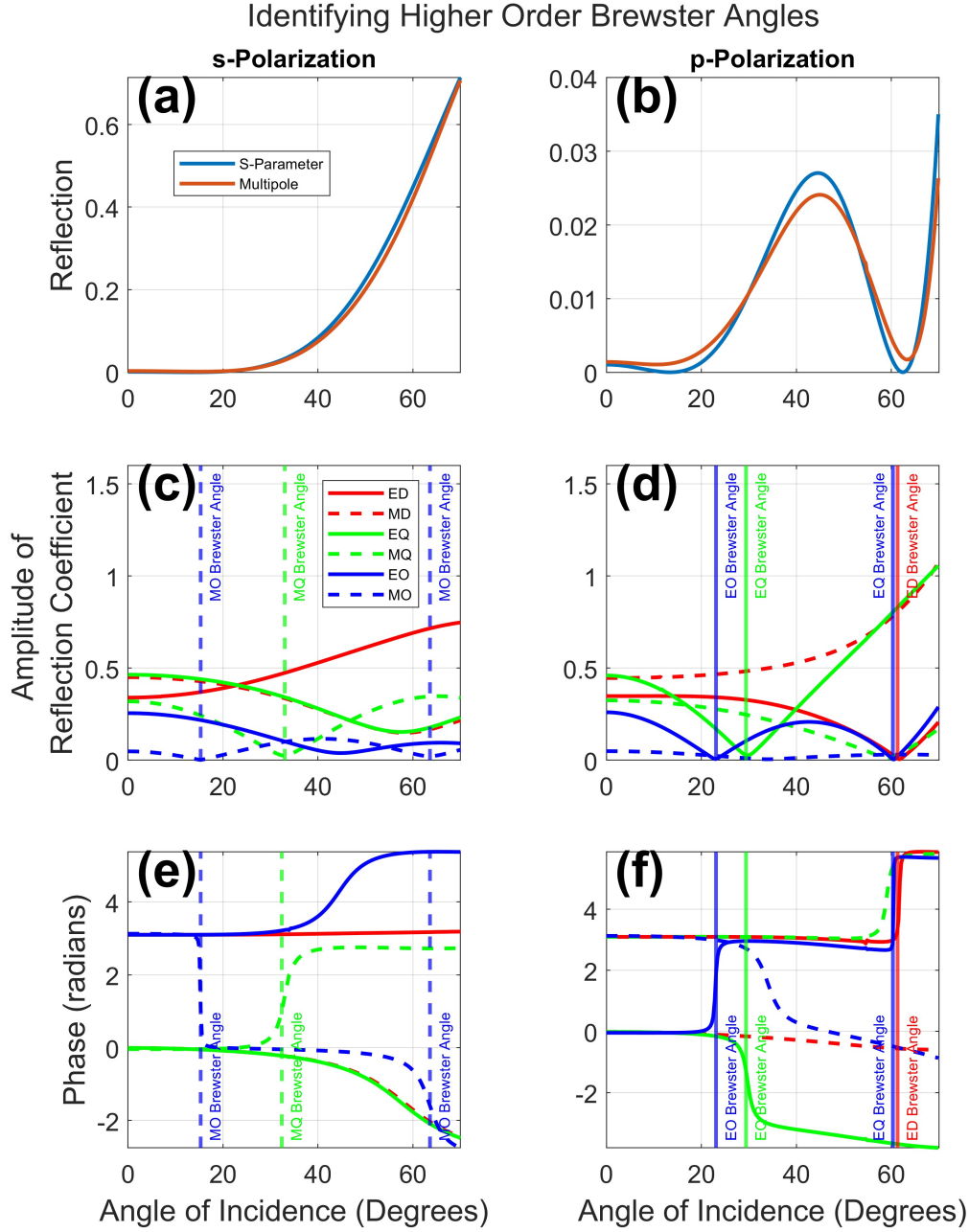


FIGURE 4.11: Demonstration of new Brewster angles arising from higher order multipoles at 1000 nm. (a)-(b) Reflection for s- and p- polarization respectively. (c)-(d) Amplitude of reflection coefficients for s- and p- polarization respectively. (e)-(f) Phase of reflection coefficients for s- and p- polarization respectively

In the case of s-polarized Brewster angles in Figure 4.11 (c), only the magnetic multipoles produce true Brewster angles, it does appear as if the electric quadrupole, the solid green line, also reaches 0 at 58 degrees, but it only comes close to zero. This near zero value can be more easily identified with the phase in Figure 4.11 (e), the π shift we previously associated with Brewster angles occurs over a larger range of angles than the true Brewster angle case.

For p-polarized Brewster angles in Figure 4.11 (d) point to a similar result, only electric multipoles produce a true Brewster angle, with magnetic multipoles coming close to zero, but not perfectly zero.

We note cases in literature of "pseudo-Brewster Angles" in absorbing materials, where reflection minima appear similarly to Brewster angles, but with non-zero reflection minima [74, 75], similar to those in the previous paragraph. While absorbing thin film are not of interest to our work, we highlight there may be potentially interesting observations to be made in such systems using our model. It may be that these "pseudo-Brewster Angles" may arise from these magnetic multipoles. However they could be easily explained by partial destructive interference instead of the complete destructive interference we observed in Figure 4.11.

While these Brewster angles are not visible in the reflection, as there is no destructive interference between remaining multipoles, as the only points of zero reflection result from either the 2nd order Fabry Perot mode or electric dipole Brewster angle in Figure 4.11 (b). However it would not be possible to correctly model the reflection, and obtain good agreement with measurements and the S-parameter model without these higher order Brewster angles, making them vital in understanding the properties of thin films.

It may be possible to observe these hidden Brewster angles for both s- and p-polarizations. However, one must ensure destructive interference between the remaining electric/ magnetic multipoles or by isolating that multipole itself. It does not seem likely that a simple thin film enough could be engineered to isolate a given multipole, given our results. However, there is much work at present in structuring materials that alter the contributions of the different multipoles.

For example, a metamaterial supporting a resonance from a single electric/magnetic multipole may support a Brewster angle for either s or p-polarization. Claims of s-polarized Brewster angles in thin films have been made previously [21], there has not been sufficient evidence to show that they are truly Brewster angles and not Fabry Perot modes. Likewise, there are several other works where there appears to be a Brewster angle, but it is not clear whether those are true Brewster angles [19, 20, 21, 76]. Multilayered films have been shown to support s-polarized Brewster angles, suggesting that sufficiently complex film arrangements may support s-polarized Brewster angles [20], assuming they truly observed a Brewster angle.

The required evidence to unambiguously demonstrate a Brewster Angle would be to expand multipole decomposition to include substrate and superstrate effects and more complex structural effects and perform a similar analysis as shown in Figure 4.9 and 4.11.

It is important to note that zero reflection at a specific angle, even if that is polarization dependent is not unique to the Brewster angle. The Borraman [77, 78] effect, the enhanced transmission of x-rays through periodic crystals at the

Bragg angle for specific polarizations of light is an example of this. This phenomena is the result of x-rays forming standing waves in that periodic crystal in-between the atoms, preventing absorption from occurring.

It is a rather unique interplay between the cancellation of a single multipole, resulting from the cross product of two aligned vectors and the interference between remaining multipoles that should be defined as the Brewster angle.

Such a definition is distinct from the multipole description of Fabry Perot resonances, which arise from the destructive interference of even and odd parities only. However we note that this definition of the Fabry Perot is very similar to the Kerker effect [73].

4.3 Measured and Modeled Reflection of Wavelength Scale Si₃N₄ Thin Films

Continuing our analysis of our Si₃N₄ thin film, we now investigate the optical electric and magnetic response for wavelengths in the visible range, from 400-1000 nm, which are of particular interest to us, given that to our knowledge, multipole models at wavelength scale and greater have not been studied in literature yet. We do this simply by moving to shorter wavelengths of light while staying with the same 500 nm thick Si₃N₄ suspended film.

Figure 4.12, like Figure 4.1 verifies our multipole model by comparison to the measured, S-parameter and multipole models.

Overall difference between experiment and model has increased, we note that at very short wavelength and large angles, the multipole model begins to predict the disappearance of the Brewster angle in Figure 4.12 (d), while the S-parameter calculation does not. We attribute this error to the lack of higher order multipoles required to describe reflection at shorter wavelengths. It can be seen in Figure 4.12 (f), that S-parameter result shows that the Brewster angle is moving to greater angles, indicating that the refractive index is increasing with wavelength. Regardless of the source the error, this indicates that our multipole model is no longer sufficient to capture the two conditions required to observe the Brewster angle as detailed in the last section.

The location of the $n=2$, $n=3$ and $n=4$ Fabry Perot modes, shown in the dashed lines is diverging from predictions made by the multipole model, but the general shape of the modes is correct.

In Figure 4.13, we plot the reflection vs angle of incidence for wavelengths of 500, 700 and 1000 nm, using the same data in Figure 4.12. The error in the multipole model with increasing angle is quite clear, with only 1000 nm giving reasonable agreement.

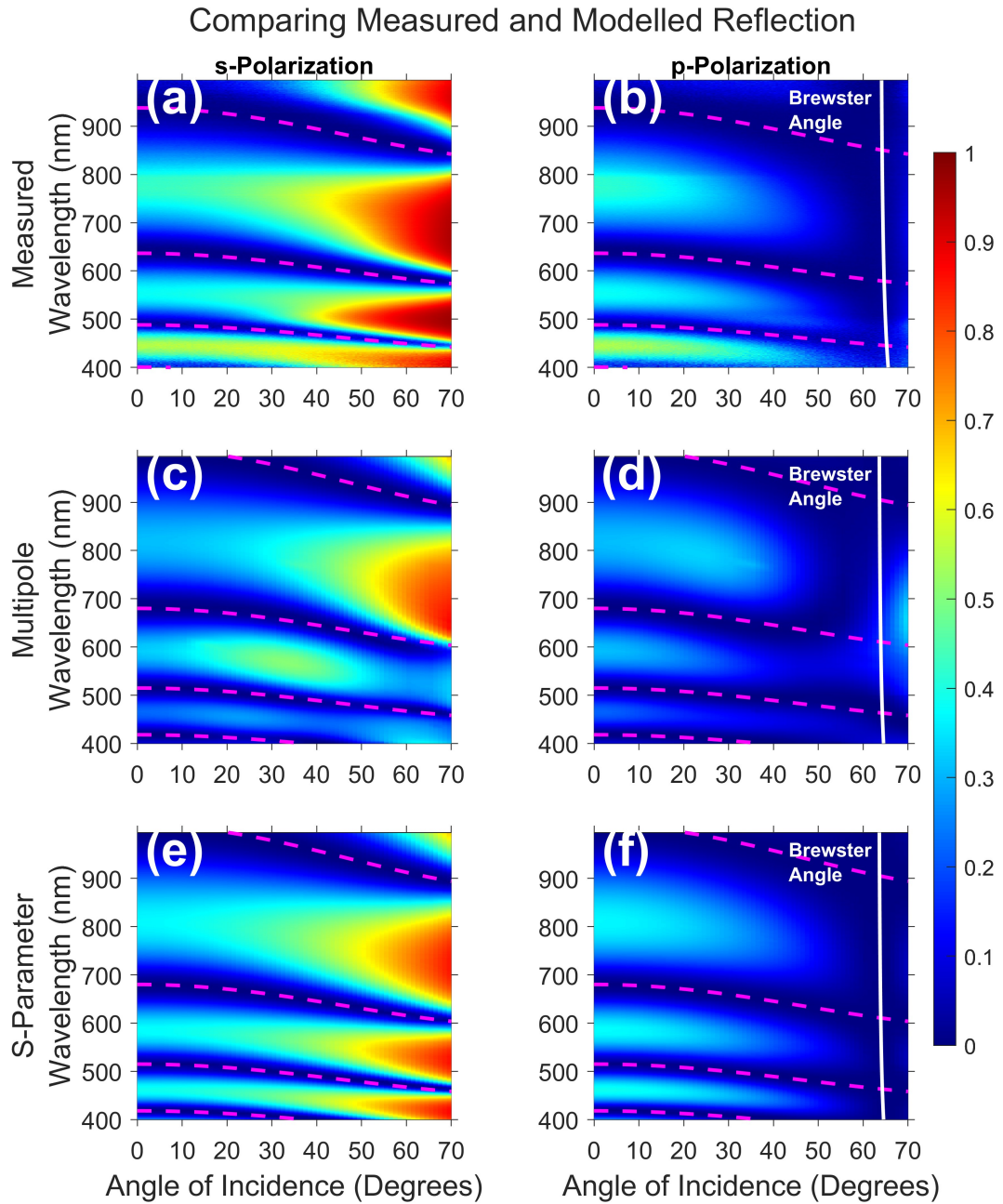


FIGURE 4.12: Comparison between measured reflection and reflection model with multipole decomposition for s and p-polarized light for wavelengths in the range 400-1000 nm. (a) Measured reflection for s-polarized light. (b) Measured reflection for p-polarized. (c) Multipole reflection for s-polarized light. (d) Multipole reflection for p-polarized. (e) S-Parameter reflection for s-polarized light. (f) S-Parameter reflection for p-polarized.

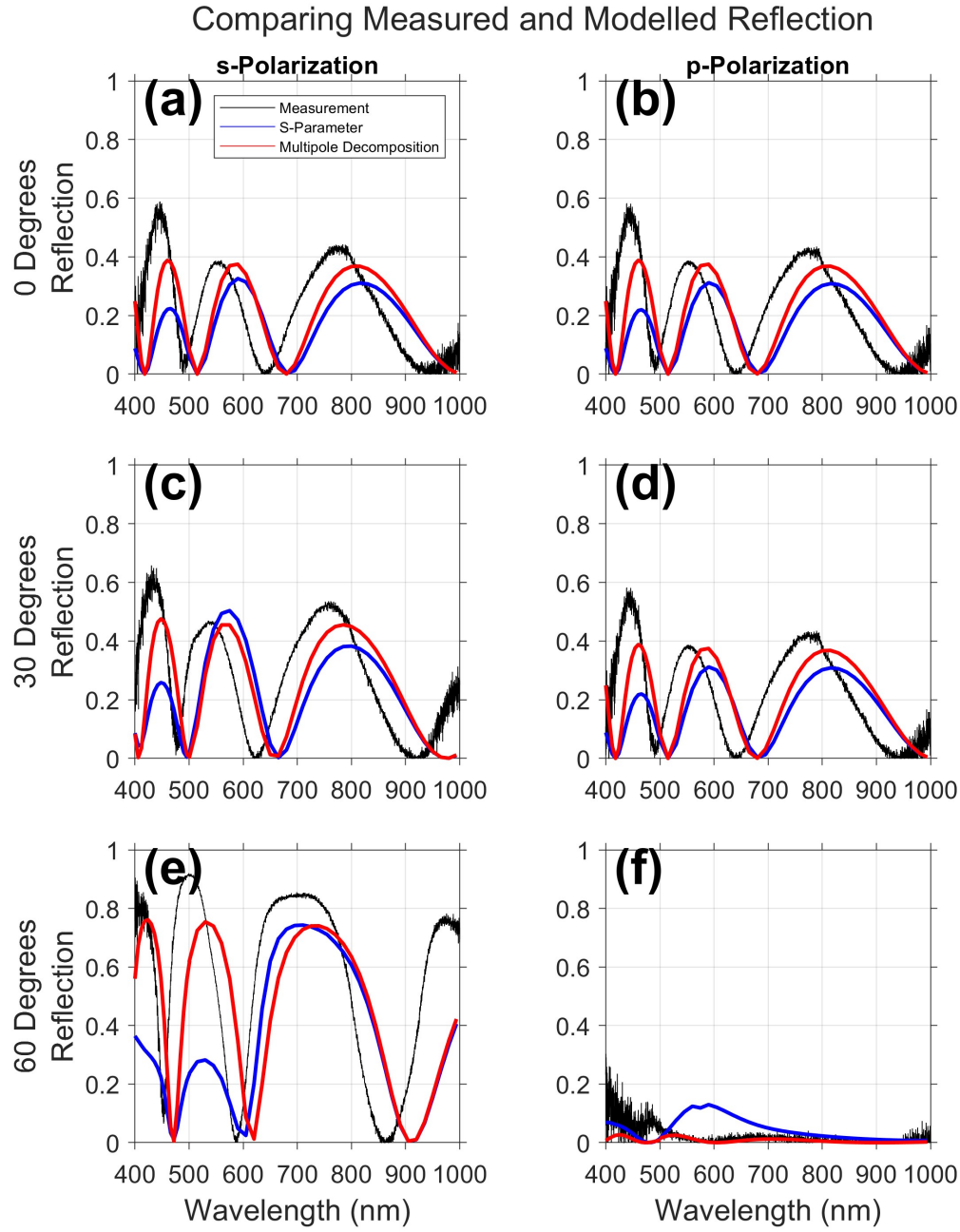


FIGURE 4.13: Angle Scans for the data presented in Figure 4.12 at 0, 30 and 60 degrees angle of incidence.

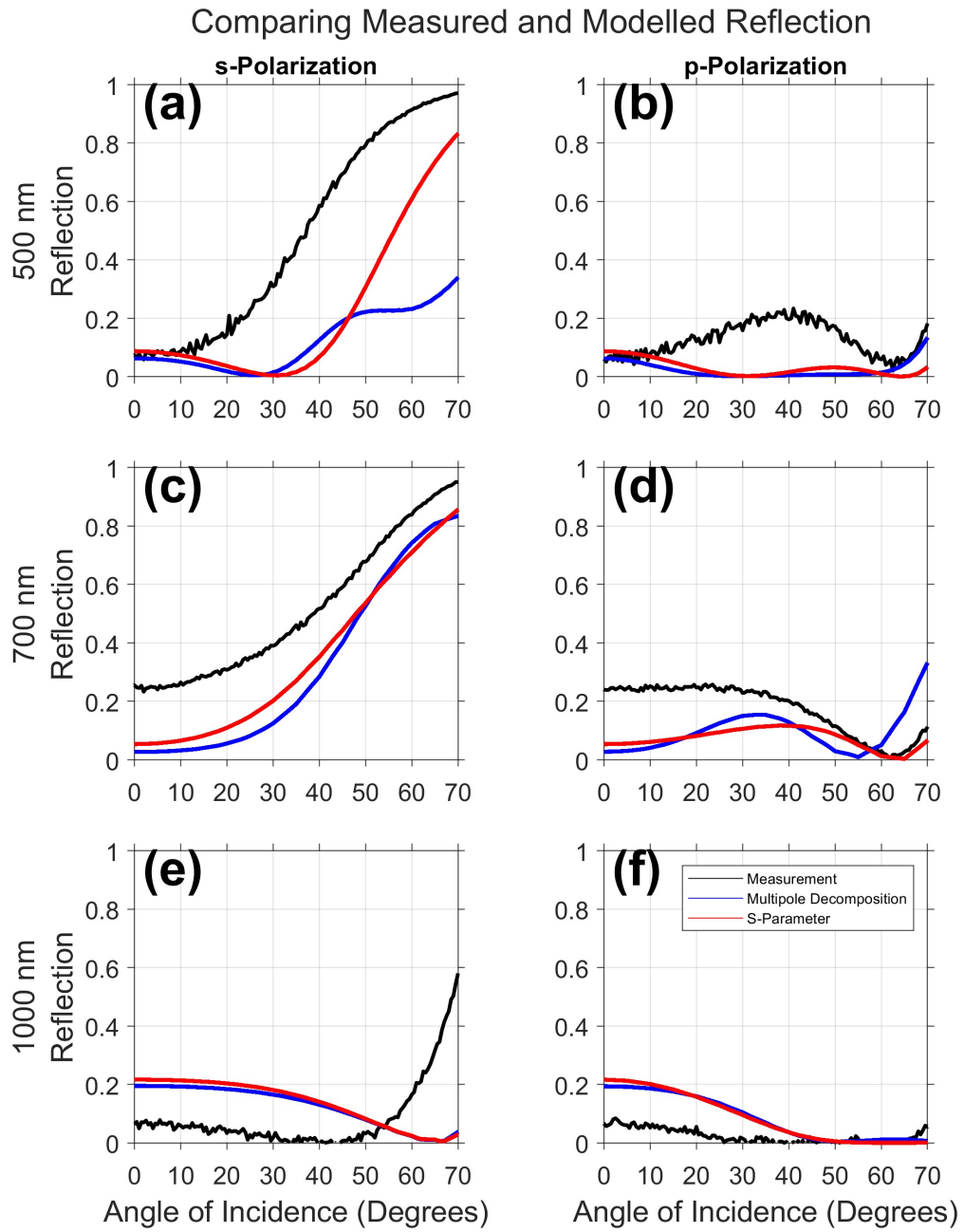


FIGURE 4.14: Angle Scans for the data presented in Figure 4.1.
 (a)-(b) 0, (c)-(d) 30 and (e)-(f) 60 degrees angle of incidence for s
 and p-polarization

4.3.1 Role of Multipoles in 3rd, 4th and 5th Order Fabry Perot Modes

We perform the same phasor investigation as we did in the last section. In Figure 4.15 and 4.16, the magnitude of the reflection coefficients is plotted. We observe that now the octupoles can no longer be ignored and the dipoles are becoming less significant.

This highlights a rather interesting property as we approach higher frequencies/shorter wavelengths, relative to the thickness of the Si_3N_4 film. The contribution to reflection from lower order multipoles reduces, and is replaced by contribution from high order multipoles. This problem is two fold, higher order multipoles exist at shorter wavelengths, but the lower multipoles have higher order terms that may become more significant at shorter frequencies [79], this is seen in Figure 4.15 (a) where the zero reflection in the electric dipole reflection coefficient disappears, suggesting the electric dipole may be lacking terms to describe the Brewster angle.

This restricts the validity of our multipole model, in order to study the structure at even shorter wavelengths, one requires the derivation of more and more terms with larger and larger tensors, such as the 16 and 32 multipoles, which are 4th and 5th order tensor respectively. At normal incidence they should only require 1 term, however they will require a large number of terms to capture the angular dependence.

We note that the magnitude of the magnetic dipole and electric quadrupole for the s-polarized coefficients in Figure 4.15 (b) and (c) are the same, as seen

previously in the NIR region. In addition there are several regions where we see a large magnetic response from the magnetic multipoles, in Figure 4.15 (c) the magnetic quadrupole has a large reflection coefficient at 900 nm and 70 degrees, while in Figure 4.15 (f) the magnetic quadrupole has a large reflection coefficient. For p-polarized reflection coefficients in Figure 4.16 (b), there are similarly large regions for the magnetic dipole at 900 nm and 70 degrees for the magnetic dipole.

Building on our discussion of Brewster angles arising from higher order multipoles, we see evidence of them in Figures 4.15 and 4.16, regions of where the reflection coefficients are zero are widespread across these datasets. We note the complexity of these new Brewster angles, given they can occur at several angles for a specific wavelength, and their location can change as a function of angle and wavelength, unlike the electric dipole Brewster angle which is unique to a specific angle for a given refractive index.

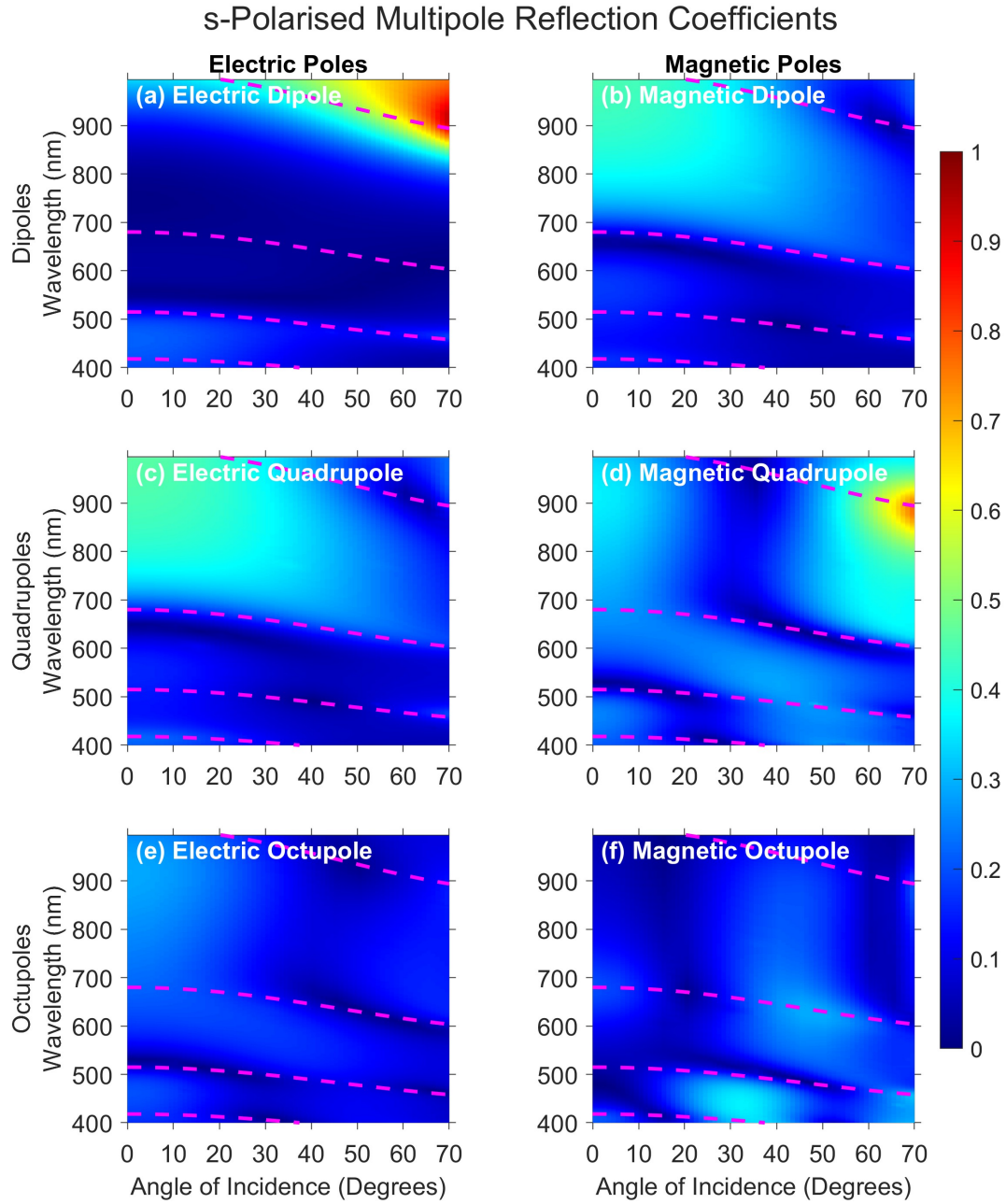


FIGURE 4.15: Amplitude of the s-polarized reflection coefficients for individual multipoles in the visible region. (a), (c), (c) Reflection from electric dipole, quadrupole and octupole respectively. (b), (d), (f) Reflection from magnetic dipole, quadrupole and octupoles respectively.

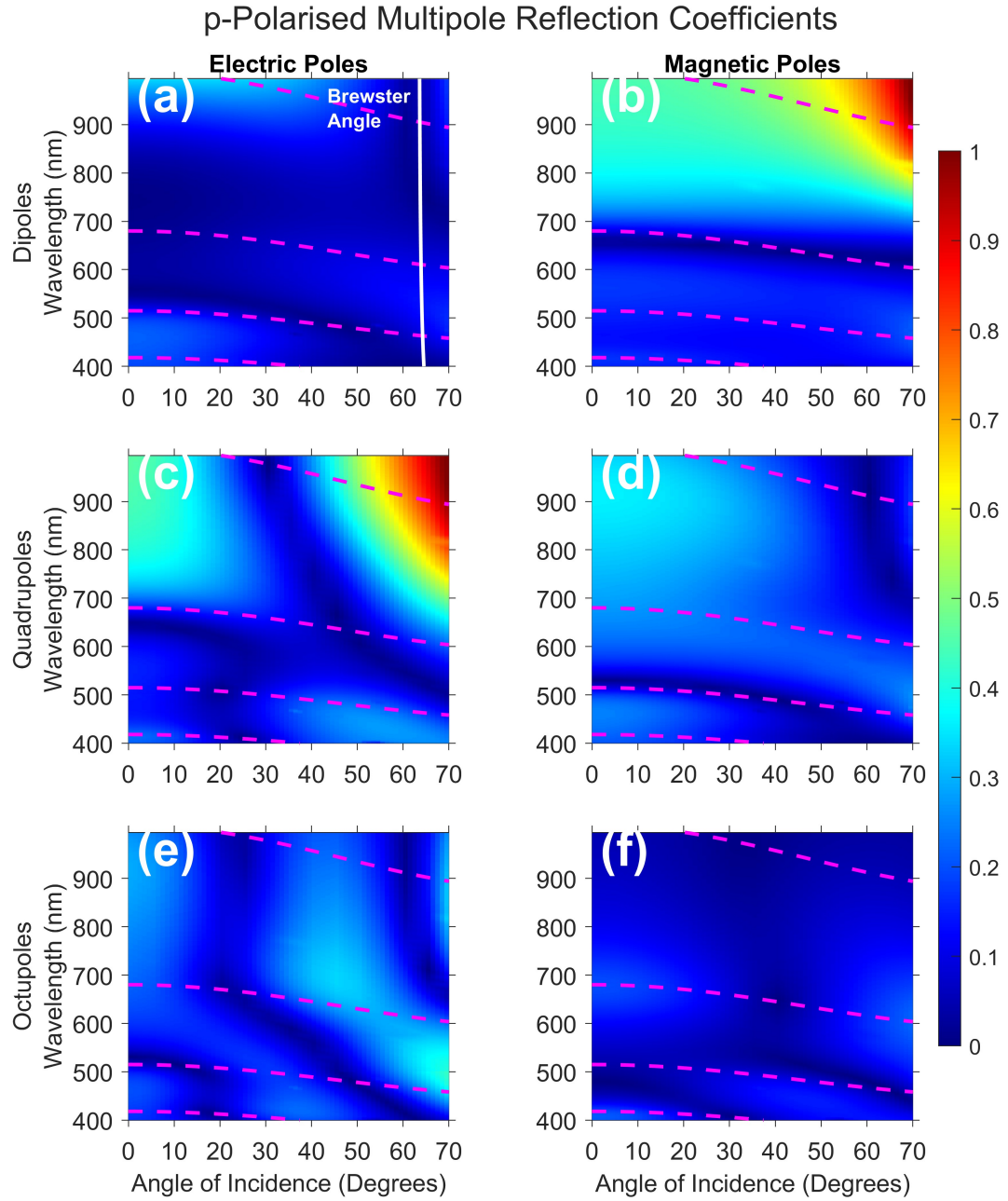


FIGURE 4.16: Amplitude of the p-polarized reflection coefficients for individual multipoles in the VIS region. (a), (c), (c) Reflection from electric dipole, quadrupole and octupole respectively. (b), (d), (f) Reflection from magnetic dipole, quadrupole and octupole respectively.

4.4 Estimating a Spatially Dispersive Extended permittivity From Electric Multipoles

To further expand on our understanding of electric multipoles in thin films, we estimate an extended permittivity resulting from the same electric multipoles we have studied so far. Such models have been used before [80, 81], as they are much more fundamental than traditional effective medium models that rely on reflection and transmission coefficients to calculate an effective permittivity, and in our case do not require an additional, potentially nonphysical homogenization procedure to estimate an extended permittivity.

The following model may be thought of an effective medium model, given its similarity to the work of field averaging effective medium theories that considered higher order multipoles [6, 18, 82].

However, one critical feature is missing, a homogenization procedure, as there is little physical justification in homogenizing a thin film, making a direct comparison with traditional effective medium theories difficult.

Instead, our permittivity model is a multipole expansion of the permittivity itself to account for quadrupoles and the presence of spatial dispersion and hence we refer to it as a "expanded permittivity". In the electric dipole case, its values match the permittivity of bulk SiN, which is expected given that the bulk permittivity bulk SiN is treated as an electric dipole response and demonstrated the validity of our approach at longer wavelengths. However, given that our multipoles model can only capture the reflection of our thin film

by including quadrupoles, we must also consider the permittivity from these multipoles as well, hence the use of a multipole expansion on the permittivity.

We begin with the well known electric displacement field, $\vec{D} = \vec{E}\epsilon_{relative}$, which is defined as the material's response to an incident electric field, and is the sum of incident electric field, $\vec{E}\epsilon_0$, and electric polarization field, $\vec{P}$.

$$\vec{D} = \vec{P} + \vec{E}\epsilon_0 = \vec{E}\epsilon_{relative} \quad (4.3)$$

For bulk materials such as Si_3N_4 , we approximate the electric polarization as an electric dipole. But in reality a multipole expansion of this vector can be performed [83].

$$\vec{P} = \vec{p} - \frac{1}{3}\nabla \cdot \hat{Q} \quad (4.4)$$

In the equation above we present the electric polarization vector in terms of the electric dipole and quadrupole multipole moments in their traceless, symmetric form.

Combining equations 4.3 and 4.4 gives us an estimation for the extended permittivity resulting from these electric multipoles as follows.

$$\epsilon_{effective,a} = 1 + \int \frac{\vec{P}_a}{\vec{E}_a} dV \quad (4.5)$$

The above equation defines an extended permittivity resulting from the spatial averaging of the ratio between the multipole moment, and electric field in cavity, where a defines one of the Cartesian axes.

For s-polarized light, we calculate the extended permittivity along the y-axis of the simulation, i.e the electric field is in-plane with the surface of the thin film.

$$\epsilon_{effective,y} = 1 + \int \frac{p_y - (\frac{dQ_{xy}}{dx} + \frac{dQ_{yz}}{dz})}{\vec{E}} dV \quad (4.6)$$

For p-polarized light, we calculate the extended permittivity along the x-axis of the simulation, i.e the electric field is in-plane to the surface of the thin film, but perpendicular to the y-axis.

$$\epsilon_{effective,x} = 1 + \int \frac{p_x - (\frac{dQ_{xx}}{dx} + \frac{dQ_{xz}}{dz})}{\vec{E}} dV \quad (4.7)$$

The permittivities described in equations 4.6 and 4.7 are for directions parallel to the surface of the thin film, as only these directions were required to predict reflection from the thin film in our study so far. We note it is possible to calculate the permittivity perpendicular to the surface, i.e the z-axis, however that is only non-unity for p-polarization, as only p-polarized light has a non-zero value for E_z for non-zero angles of incidence.

These multipole moments are the same as those presented earlier in our derivation of the angle dependent multipole decomposition mode, which were verified by comparison to experiment in Figures 4.1 and 4.12. However there are

several difference between the prediction of reflection and extended permittivity calculations that must be highlighted.

Firstly, the calculation of reflection requires one to consider the direction of reflection and perform a series of cross products with the multipole moments to calculate the reflection coefficient associated with a given multipole moment. For our effective permittivity calculation, such a consideration is not taken, given that a multipole may have a non-zero moment, but may not contribute to reflection. We highlighted such a possibility during our study of Brewster angles, where a vector that is either the multipole moment, or associated with that multipole aligns with the direction of reflection, resulting in zero reflection. If we consider the electric dipole Brewster angle, the electric dipole still polarizes the medium, but does not contribute to reflection. We note this will lead to significant differences in presented data between effective medium parameters and reflection coefficients.

Secondly, the field inside the cavity, E , is not extracted in the same manner of the incident field, E_0 , for the multipole model presented so far, as it is taken from within the thin film, and is related to the field enhancement inside the cavity rather than the incident field that approaches the cavity.

It is important to note that the vectors and tensors shown above are all complex valued, however in our case, the complex extended permittivity has a sufficiently small positive imaginary part, $< 10^{-5}$, that it may be treated as purely real, which is in agreement with the bulk permittivity of Si_3N_4 [24] over most of the wavelengths we studied. In Figure 4.17, we present the extended permittivity resulting from equation 4.6 and 4.7 across the visible and near-infrared regions studied.

In the case of electric dipoles in Figures 4.17 (a) and (b), there is no angle dependence in the permittivity, and no difference between s and p-polarization. Isotropic media with electric dipole responses, i.e, all non-crystalline bulk media such as SiN, SiO₂, are generally not known to be spatially dispersive, as that would suggest that all isotropic materials would be spatially dispersive, which is not true. However anisotropic crystalline media with electric dipole responses are spatially dispersive, as the electric dipole response depends on the crystal axis.

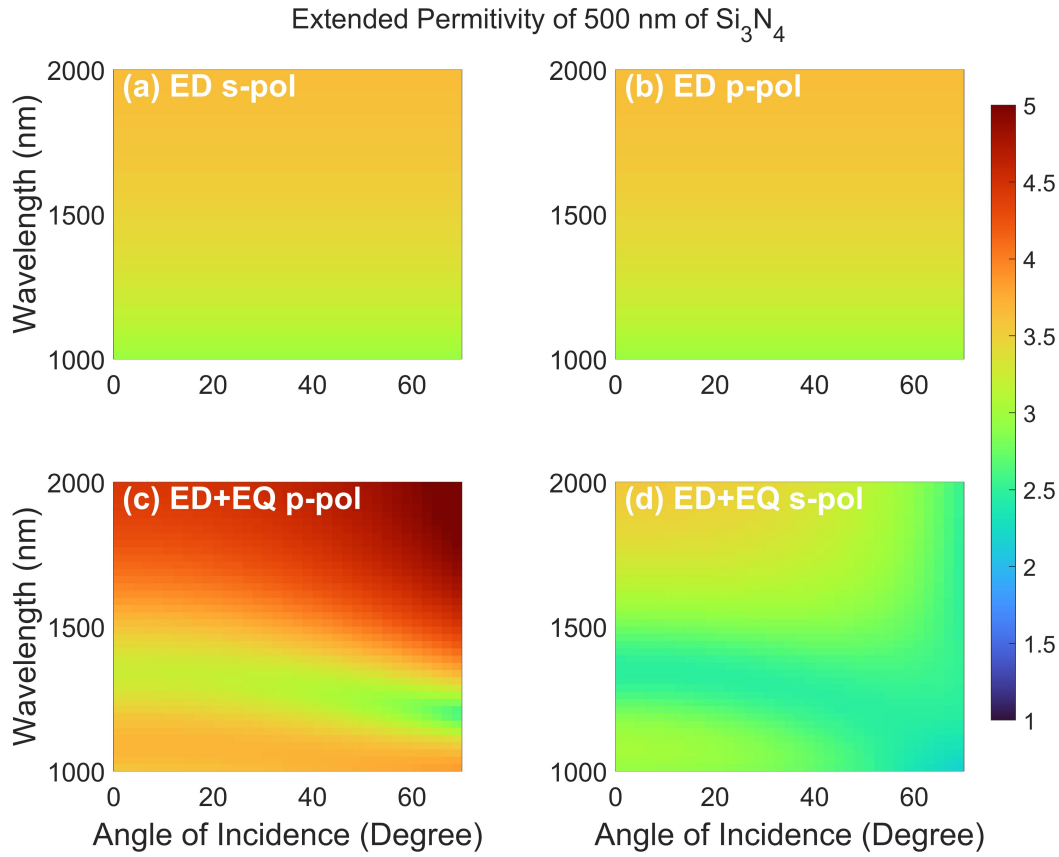


FIGURE 4.17: extended permittivity Calculation for 500 nm of Si₃N₄ for both polarizations. (a)-(b) Dipole contribution. (c)-(d) Dipole and Quadrupole contribution.

The addition of electric quadrupole in Figures 4.17 (c) and (d), shows the presence of an angular dependence in the permittivity, which is also referred to

as spatial dispersion in the permittivity. The two polarizations produce different effective permittivities given that the angle dependence for the quadrupole varies with polarization, which can be seen in previous datasets throughout this work.

The critical result in Figure 4.17 is the demonstration of spatial dispersion, that is a wavevector dependence on the permittivity, which can be observed as an angular dependence on the permittivity.

Spatial dispersion in the permittivity has many potential applications, given that it is indistinguishable from a non-unity value of magnetic permeability [1, 31], which is difficult to obtain at visible or NIR wavelengths without optical metamaterials [1, 84].

4.4.1 Estimating the Contribution of Magnetic Multipoles to Spatial Dispersion

In the presence of magnetic multipoles and subwavelength structures, it is common to calculate a magnetic permeability [16]. However in the case of spatial dispersion in the permittivity, which we previously identified in the EQ contribution in Figure 4.17, it is not possible to define the electric and magnetic permeability uniquely [1, 3] and was previously discussed as in Chapter 2.

The solution to this issue is described by Agranovich [3], he redefines the electric displacement field, $\vec{D}$, in terms of electric and magnetic fields, any magnetic response is included in the spatial dispersion of the electric permittivity.

$$\vec{D}' = \epsilon \vec{E} - \frac{c}{\omega} \left(1 - \frac{1}{\mu(\omega)} \right) \nabla \times \vec{B} \quad (4.8)$$

Another derivation was performed independently of Agranovich to resolve the same issue, but in terms of electric and magnetic multipoles [83].

$$\vec{D}' = \vec{p} - \frac{1}{3} \nabla \cdot \hat{Q} + \frac{i}{\omega} \nabla \times \left(\vec{m} - \frac{1}{3} \nabla \cdot \hat{M} Q \right) \quad (4.9)$$

These equations permit a calculation whereby we account for the role of a magnetic response on the electric permittivity.

In the case of s-polarization, we obtain the following equation

$$\begin{aligned} \epsilon_{effective,y} = 1 + \int \frac{p_y - \left(\frac{dQ_{xy}}{dx} + \frac{dQ_{yz}}{dz} \right)}{E_y} dV \\ + \int \frac{\frac{dm_x}{dz} - \frac{d}{dz} \left(\frac{dM_{xx}}{dx} + \frac{dM_{xz}}{dz} \right) - \frac{d}{dx} \left(\frac{dM_{xz}}{dx} + \frac{dM_{zz}}{dz} \right)}{H_x} dV \end{aligned} \quad (4.10)$$

and in the case of p-polarization

$$\epsilon_{effective,x} = 1 + \int \frac{p_x - \left(\frac{dQ_{xx}}{dx} + \frac{dQ_{xz}}{dz} \right)}{E_x} dV + \int \frac{\frac{dm_y}{dz} - \frac{d}{dx} \left(\frac{dM_{xy}}{dx} + \frac{dM_{zy}}{dz} \right)}{H_y} dV \quad (4.11)$$

The first term in the above equations, matches that described earlier when accounting for electric multipoles, the second term accounts for magnetic multipoles.

In Figure 4.18, we investigate the role of the magnetic dipole and magnetic quadrupole on the extended permittivity. The magnetic dipole in Figure 4.18 (a) and (b) plays a major role in the extended permittivity, while the magnetic quadrupole does not make noticeable contribution across the NIR, which was seen previously for the reflection coefficients for the magnetic quadrupole.

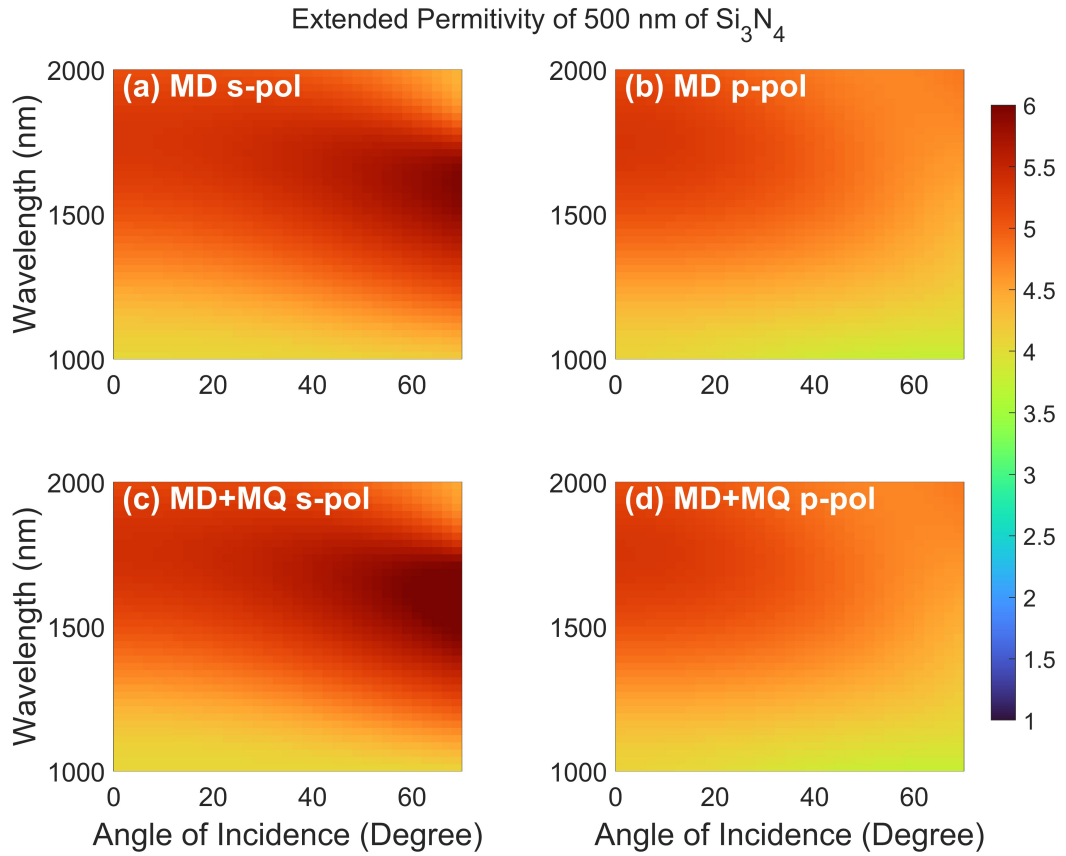


FIGURE 4.18: extended permittivity calculation for 500 nm of Si_3N_4 for both polarizations. (a)-(b) Dipole contribution. (c)-(d) Dipole and quadrupole contribution. Contributions from the electric multipoles are not considered.

An important difference between Figures 4.17 and 4.18 is that the electric dipole is not spatially dispersive, while the magnetic dipole is spatially dispersive and this dispersion is polarization dependent, similarly to the electric quadrupole case.

This highlights that both electric and magnetic responses, in this case, the electric quadrupole and magnetic dipole can both contribute to spatial dispersion, which itself is used to describe the magnetic response of a given media as highlighted in Chapter 2.

Agranovich [31] highlights how such an electric quadrupole may behave as a magnetic dipole as they would both contribute to spatial dispersion. He similarly highlights research performed by Cho et al [32], who demonstrated how an electric quadrupole resonance may give rise to a negative permeability.

Landau makes a similar comment [1] when discussing spatial dispersion, "the effects due to the difference of μ from 1 are in general indistinguishable from those of the spatial dispersion of the permittivity".

A similar comment made by Johnson et al [85] when theoretically exploring the electrodynamics of a vacuum-dielectric interface when accounting for spatial dispersion, they made the following comment, "We see that any material exhibiting spatial dispersion is "magnetic" ".

An important observation to make is that while the true magnetic response arising from magnetic multipoles is only valid in the subwavelength regime, according to the Landau and Lifshitz [1], an electric response does not follow such a restriction, and if that electric response is spatially dispersive, then it

may behave as a magnetic response.

However our model is not valid towards shorter wavelengths, as we lack the higher order multipoles required to correctly describe the behavior at such frequencies, regardless we note that spatial dispersion is an intrinsic property of higher order electric multipoles, and in isotropic systems such as ours, it is only the electric dipole that is not spatially dispersive.

4.4.2 Spatial Dispersion in Dispersion Relations

In the case where our effective medium calculations can not be performed, that is when we change the properties of the substrate, add more structural complexity, or as highlighted earlier, when the film is sufficiently thick to require higher order multipoles than those we modelled, the relation between a wave's frequency and wavevector, the dispersion relation of the wave, may prove to be a useful tool to identity this spatial dispersion, while lacking a more fundamental analysis that we have provided above.

While the possibility of thin films being spatially dispersive has been previously highlighted by Kinsler [33], his argument was related to the dispersion relation, that is the plot of frequency vs wavevector as shown in Figure 4.19 below. However Kinsler did not deal in detail with Fabry Perot modes, whereas we explore angle dependence of these modes.

For a dispersionless wave, the following equation holds true where ω is the frequency of the wave, k is the wavevector and c is the speed of light.

$$\omega^2 = c^2 k^2 \quad (4.12)$$

We note that in this case, equation 4.12 behaves as a linear equation in the form

$$y = mx \quad (4.13)$$

where the slope of the straight line, m , is determined by the speed of light. Kinsler argues that any deviation from this straight line described by equation 4.12 is evidence of spatial dispersion, his criteria for a non-spatially dispersive case is the following, the dispersion relation must be linear, the dispersion relation must pass through 0, and the relation must be continuous. If any of these three criteria are broken, then the wave is experiencing dispersion, however separating frequency and spatial dispersion may be difficult [33].

Figure 4.19 plots the dispersion relation for several cases. First are the bulk material cases, the solid blue line is a material with no frequency or spatial dispersion, that is a constant refractive index of 2. The dashed orange line is the bulk dispersion of Si_3N_4 , which contains frequency dispersion only, which was previously in Figure 2.2 and taken from Luke et al [24]. The remaining lines are the dispersion plots for the first 5 Fabry Perot modes for 500 nm of Si_3N_4 .

It is clear from the three equations below that the three are different. The equation for the dispersionless case of $n=2$ is.

$$\omega^2 = \frac{c^2 k^2}{4} \quad (4.14)$$

The equation for the frequency dispersion case is

$$\omega^2 = \frac{c^2 k^2}{n(\omega)_{Si_3N_4}^2} \quad (4.15)$$

This frequency dispersion case is not linear given the refractive index varies with frequency. Using the equation for locating the resonant wavelength and angle of a Fabry Perot mode in Chapter 2, equation 2.19, we can derive a similar equation for the dispersion relation of the Fabry Perot mode dispersion below.

$$\omega^2 = c^2 k_{FP}^2 = \frac{c^2}{\lambda_{FP}^2} = \frac{c^2}{n(\omega)_{Si_3N_4}^2} \left(\frac{q}{2d \cos(\sin(\frac{\sin(\theta_{incident})}{n(\omega)_{Si_3N_4}}) - 1)} \right)^2 \quad (4.16)$$

where d is the thickness of the thin film, q is the mode order and $\theta_{incident}$ is the angle of incident.

The Fabry Perot case has additional limitations, the mode order, q , will prevent the relation from being continuous, confining each mode to a limited range of wavevectors and frequencies.

In Figure 4.19 (a), we note that the dispersion of the Fabry Perot modes lies along the continuous line that is the frequency dispersion of Si_3N_4 , however each mode is confined to a limited range of frequencies and wavevectors for a given mode number, which is a result of the derivation of the Fabry Perot modes from Chapter 2 equations 2.12, where the solution to the wave equation is valid for specific waves of a given wavevector, not frequency. Kinsler argues this is spatial dispersion intrinsic to all Fabry Perot modes.

An interesting observation in Figure 4.19 (a) is that the separation between dispersion relations of adjacent Fabry Perot modes reduces with higher order modes, for a sufficiently high order mode, there may be little or no separation between adjacent modes, making it appear as if the dispersion relation is continuous, but it is still not truly continuous.

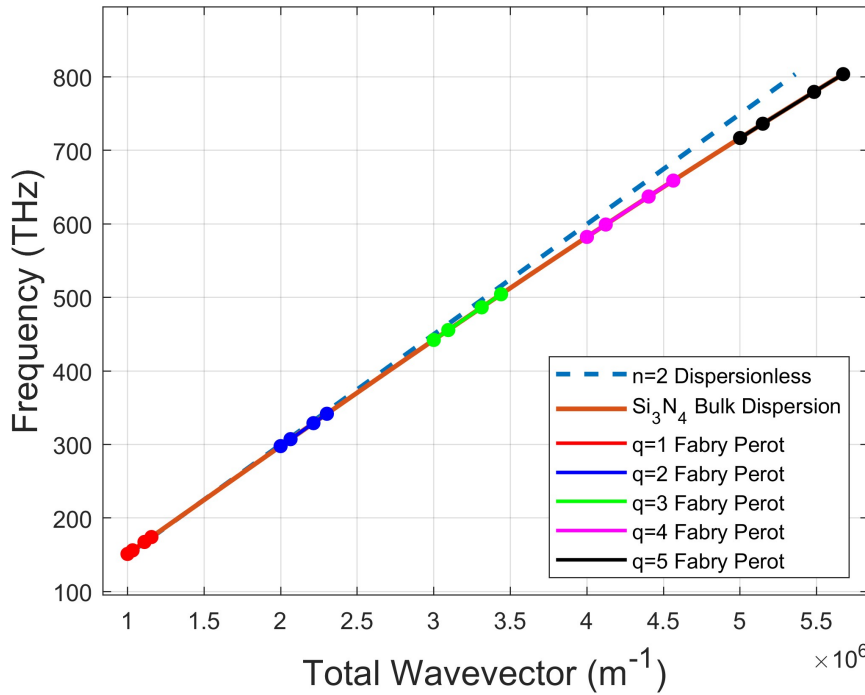


FIGURE 4.19: Dispersion relation of bulk materials and Fabry Perot Modes. The circles mark points that correspond to 0, 30, 60 and 90 angles of incidence.

In simple terms, the source of this spatial dispersion in Figures 4.17, 4.18 and 4.19 is that Si_3N_4 thin film has a finite thickness, if the Si_3N_4 had no boundaries, then spatial dispersion would not be observed. In Figure 4.17 and 4.18, this film of finite thickness supports magnetic dipoles and electric quadrupole that are spatially dispersive, where as in Figure 4.19 spatial dispersion is the result of the mode order, q , in equation 4.16 restricting the possible values of frequency and wavevector that are possible, turning a continuous function into a partially discrete one.

This origin of spatial dispersion is very different to bulk materials that have been observed to support spatial dispersion, we outline several forms of spatial dispersion, and compare to spatial dispersion in our work.

One source of spatial dispersion arises from the structural anisotropy of a crystal lattice, which is traditionally referred to as the ordinary and extraordinary refractive indices of materials such as YVO_4 [86]. These media have a pure electric dipole response that depends on the direction of propagation, unlike our case where our electric dipole response was independent of the direction of propagation.

A second source of spatial dispersion can also be observed in isotropic crystals, in the vicinity of absorption features, which has been predicted theoretically [87], and observed in Si [88], CdS [89, 90], GaAs [91], BaF_2 and CaF_2 [92] single crystals.

Our work highlights a third source of spatial dispersion, magnetic multipoles and higher order electric multipoles. Previously the role of electric multipoles in spatial dispersion has only been studied in metamaterials [58] but it is not

typical for spatial dispersion to be considered in effective medium models. Mathematically speaking, equation 4.9 shows this origin of spatial dispersion. Spatial dispersion in the quadrupole results from the spatial derivative, either from divergences ($\nabla \cdot \vec{A}$) or curls ($\nabla \times \vec{A}$) of the multipole moment, which the non-spatially dispersive electric dipole lacks.

Unlike metamaterials, thin films can support these electric multipoles at thickness comparable, or greater than the wavelength. However it seems it may be difficult to isolate and control individual multipoles in a single thin film, making metamaterials more ideal when such control is desirable.

4.5 Reflection Beyond the Wavelength Scale in Thick Films of SiO₂

Continuing our extension of our study of spatial dispersion to films thicker than the wavelength of light, we investigate the measured and modeled reflection of a 50 μm film of SiO₂. The comparison between measured and modeled reflection through the analytical transfer matrix method is shown in Figure 4.20. The analytical transfer matrix is provided by Lumerical STACK and outlined in Chapter 3, Section 3.3.1.

Overall, all we see good agreement between the measured and calculated reflectivity, we only plot 600-750 nm as that is the range of wavelengths that a single Fabry Perot mode exists over the range of angles reliably measured, as seen by the magenta line in Figures 4.20 (b), (d) and (e), where we have plotted

the resonant wavelengths and angles for the 200 th Fabry Perot mode.

The Brewster angle is weakly seen in the measurements in Figure 4.21 at roughly 56 degrees, which is expected for SiO₂ samples. Given the thickness of the sample, and the large number of modes, it is easy to observe changes in the overall shape of these modes, unlike our Si₃N₄ sample, while they are all Lorentzian lineshapes, the width of these lineshapes is an important parameter to explore, and is typically referred to as the full width half maximum or spectral linewidth.

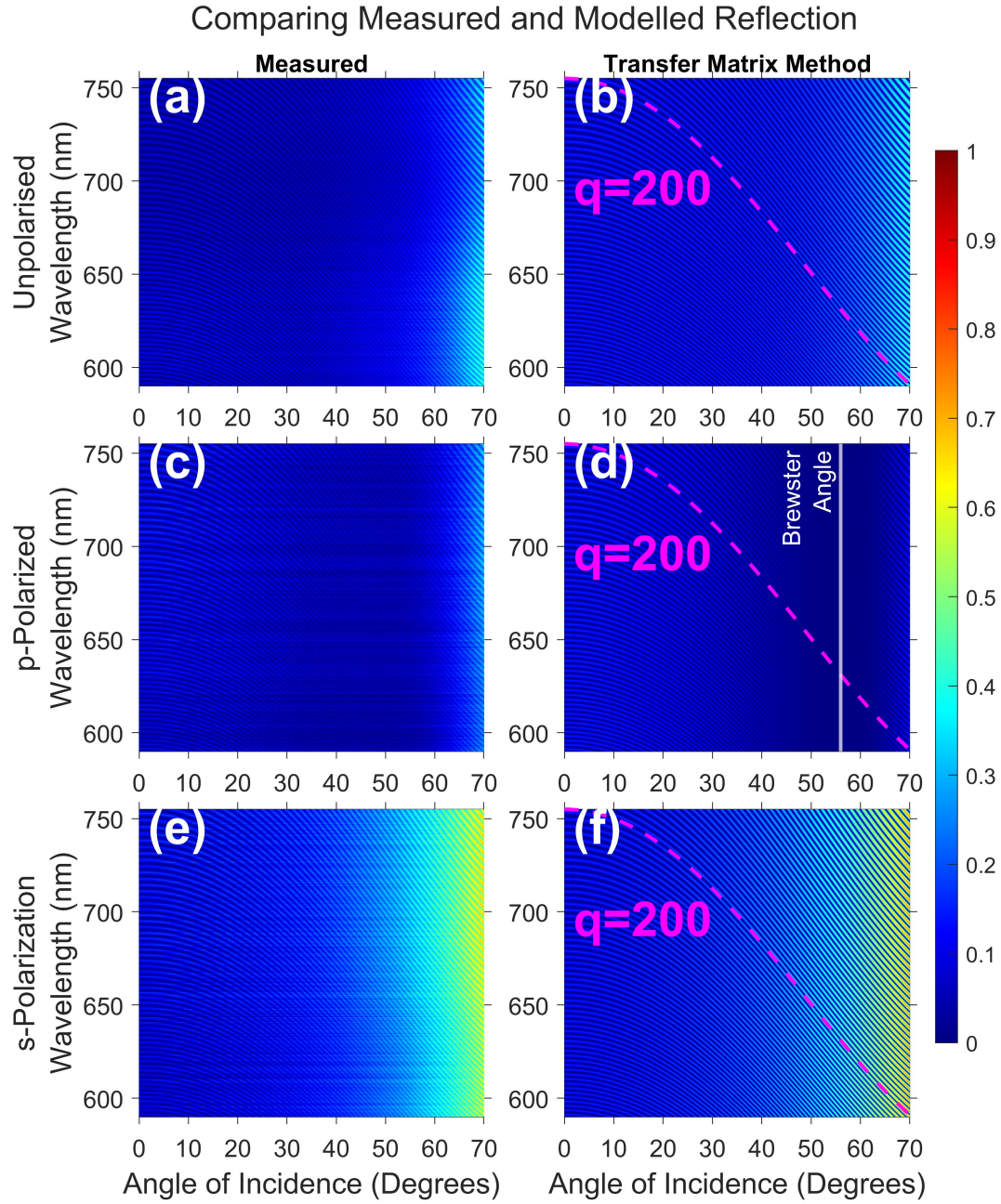


FIGURE 4.20: Comparison of measured and modeled reflection for 3 polarizations of $50\ \mu\text{m}$ of SiO_2 . Dashed magenta line marks the location of the 200 th order Fabry Perot mode. (a)-(b) Measured and modeled reflection for unpolarized light. (c)-(d) Measured and modeled reflection for p-polarized light. (e)-(f) Measured and modeled reflection for s-polarized light.

We observe that increasing the angle of incident decreases the spectral linewidth, with the s-polarized linewidths being narrower than their p-polarized counterparts. It is possible to analytically predict the linewidth, by calculating the finesse of Fabry Perot modes, as long as they follow a Lorentzian shape [93].

$$Finesse = \frac{\Delta\nu_{FSR}}{\Delta\nu_c} = \frac{2\pi}{-\ln(R_1 R_2)} \quad (4.17)$$

where $\Delta\nu_{FSR}$ is the free spectral range, defining the spacing between adjacent modes. $\Delta\nu_c$ is the linewidth of the Lorentzian resonance. R_1 and R_2 defines the reflection from each surface, in this case, the air-glass and glass-air surface of our sample, which can be easily calculated with the Fresnel equation. It should be immediately clear that the Finesse between polarizations deviates at non-zero angles of incidence, given the Fresnel equations do so.

Figure 4.21 (c) shows the plot of equation 4.17, and we do observe that s-polarized Fabry Perot modes are indeed narrower than their p-polarized counterpart, which was observed in both Figure 4.21 (a) and (b).

However it is not fully clear as to whether both FSR and linewidth are changing in equations 4.18, we explore this with Figure 4.22. While it is difficult to fully discern the locations of the measured peaks at such a large angle, and hence extract the true linewidth of those modes, it is still possible to compare the FSR between measurement and model in Figure 4.22 (c). The agreement of the measured and modeled FSR is quite good, demonstrating that the FSR between polarizations does not vary, and only the linewidth between polarizations is deviating.

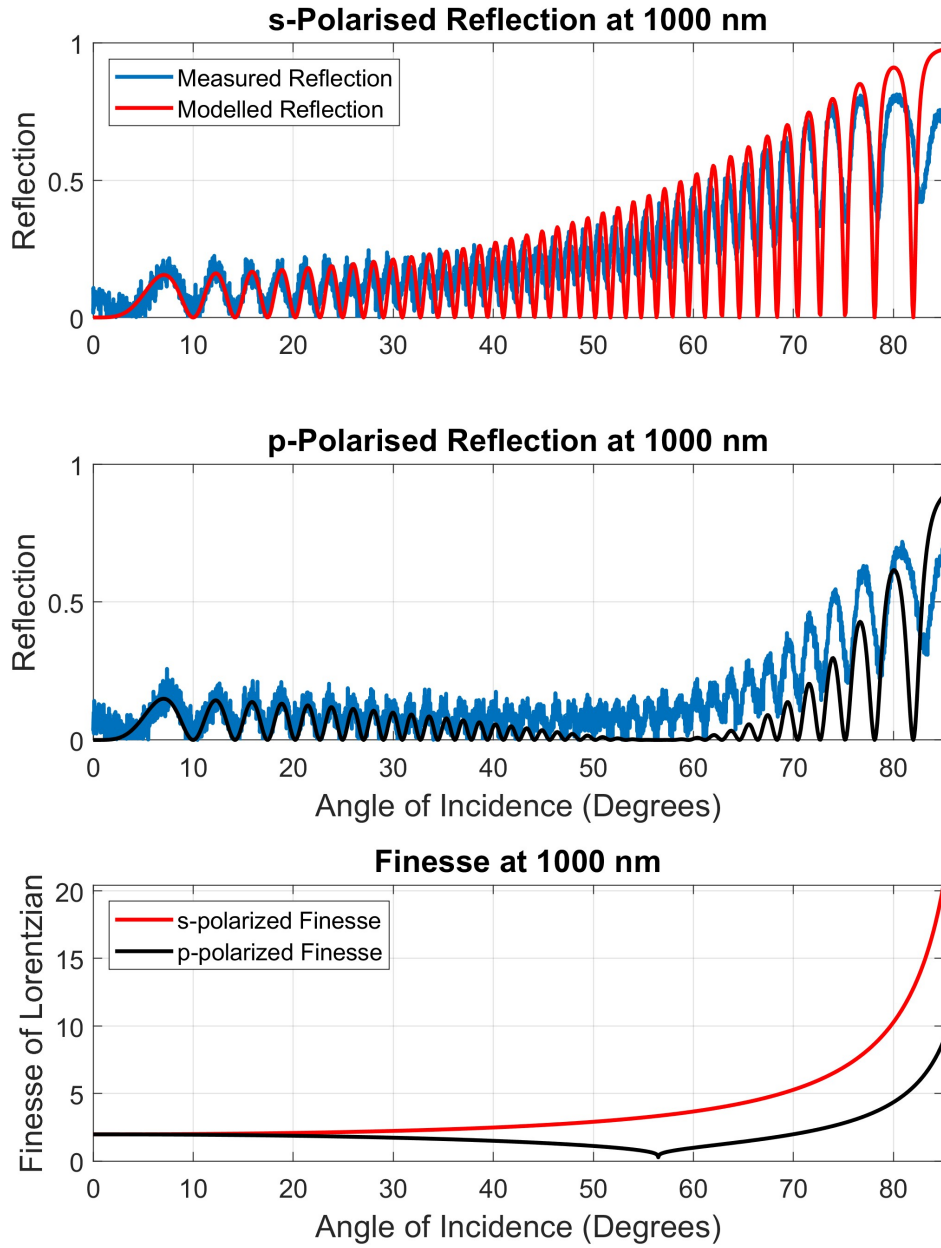


FIGURE 4.21: Comparison between measured and modeled reflection at 1000 nm. (a) s-polarized reflection. (b) p-polarized reflection. (c) Plot of analytically predicted finesse, calculated by equation 4.17.

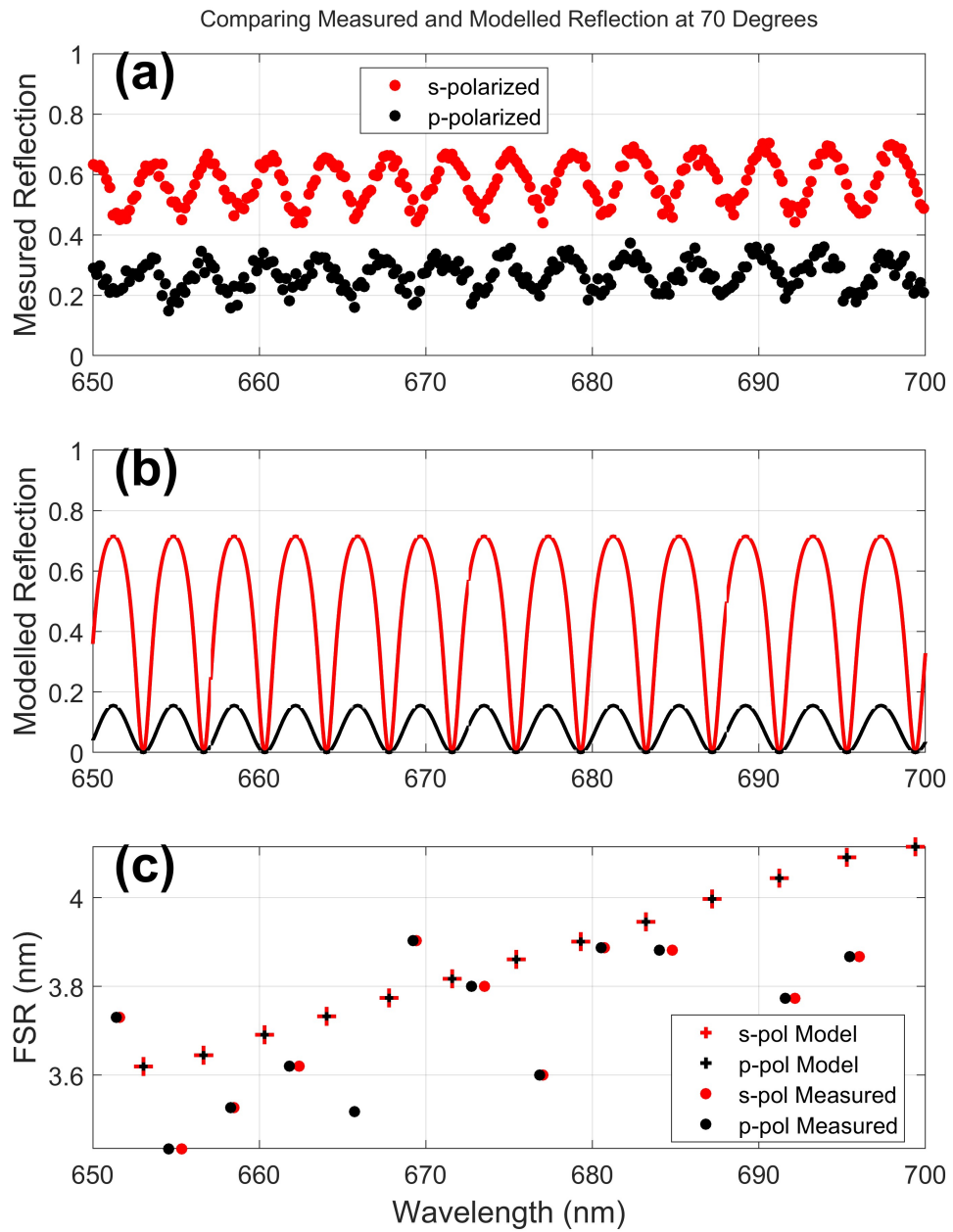


FIGURE 4.22: Comparison between measured and modeled reflection at an angle of incidence of 70 degrees. (a) Measured reflection for both polarizations. (b) Modelled reflection for both polarizations. (c) Comparison between measured and modeled FSR, calculated from subplots (a) and (b).

4.5.1 Presence of Spatial Dispersion in SiO₂ Thin Films

To explore the role of spatial dispersion in our SiO₂ we perform the same dispersion plot previously shown in Figure 4.19, given our multipole model does not have the higher order multipoles required to estimate an extended permittivity for such a thick sample.

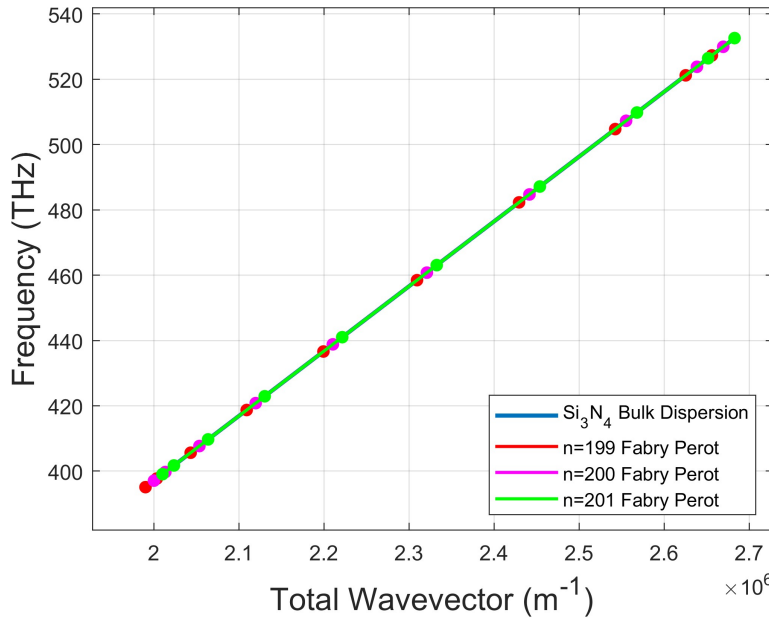


FIGURE 4.23: Dispersion relations of the 199th, 200th and 201st Fabry Perot modes. Circles mark wavevectors and frequencies that correspond to angles of incidence of 0, 30, 60 and 90 degrees.

In Figure 4.23, we plot the dispersion of the 200th mode, which we showed in Figure 4.20 as well as the adjacent Fabry Perot modes. As predicted previously, we see that the three modes overlap, unlike for 500 nm of Si₃N₄, where there was a clear separation between Fabry Perot modes. The overlap of modes suggests that these Fabry Perot modes can exist at the same frequency, with different wavevectors/angles of incidences, a result observed in Figure 4.21, where several modes are observed at 1000 nm.

A more nuanced discussion of this spatial dispersion has already been made in Section 4.3. We simply wish to highlight that this spatial dispersion is present in thin films that are significantly larger than the incident wavelength, permitting potential applications of this spatial dispersion to structures that are far larger than the wavelength of light.

4.6 Conclusion

Describing the optical response of a thin films in terms of electric and magnetic multipoles permits for a fundamental perspective on the behavior of well known phenomena such as Fabry Perot modes and Brewster angles.

We highlight how all Fabry Perot modes are the result of destructive interference in the reflection direction, resulting in perfect reflection. At non-zero angles of incidence the behavior of multipoles deviates with polarization, highlighting how selecting angle, wavelength and polarization may be tailored for a specific electric or magnetic response.

Our multipole model verifies that the long held understanding that the Brewster angle is the result of the electric dipole moment and direction of reflection aligning, resulting in zero reflection. However it also identifies a previously unobserved condition to observe the Brewster angle, the destructive interference between electric and magnetic multipoles, without this effect the Brewster angle would not be observable.

This new observation highlights that higher order multipoles may also align

with the direction of reflection and support their own Brewster angles, but are hidden by the lack of destructive interference. Regardless of whether they are hidden or not, we cannot accurately model reflection without these new Brewster angles.

Finally we highlight how spatial dispersion may arise from electric and magnetic multipoles. As highlighted in the introduction, spatial dispersion is equivalent to a non-unity value of magnetic permeability, we show how this equivalency is captured through the spatial derivatives of the magnetic dipole, quadrupole and electric quadrupole, suggesting that certain electric responses can give rise to magnetic responses.

Chapter 5

Exciting TE and TM Surface Waves in PMMA-Au Multilayers

5.1 Demonstrating a Magnetic Response

Building upon the work so far, we now move on to explore a potential application of these magnetic responses that arise from both higher order electric multipoles and magnetic poles.

As highlighted in the introduction, the observation of optical magnetism through modeling is an important step. A practical application of that optical magnetism is of great interest, and is an important step in verifying the presence of optical magnetism.

Towards this goal, we modify the angle dependent response of the low loss Fabry Perot modes studied so far, by depositing a thin transparent film of PMMA, which acts as a lossless Fabry Perot resonator onto of a thin metal film of Au that is deposited on an SiO₂ substrate. The PMMA thickness was selected to be in the range of 1 μm given that multiple models would be visible across a wide range of angles/wavelengths making it easier to observe them in measurement. The 40 nm Au thickness was chosen for a similar reason, as simulations suggested per perfect absorption at that thickness.

Of particular interest to us is the propagating surface plasmon, which exists at the interface between two bulk media, and is one of most well-known and understood cases of surface plasmons. TM surface plasmons require differing electric permittivities across the interface, while TE plasmons require differing magnetic permeabilities, which we have demonstrated in the last chapter, is an intrinsic feature of thin films.

We begin this work by verifying that the measured reflection of fabricated structure outlined in Chapter 3 matches the theoretical predicted reflection, highlighting features of these modified Fabry Perot resonators, before directly identifying these TE plasmons through eigenmode analysis.

5.2 Comparing Measured and Modeled Reflection for our Au-PMMA Samples

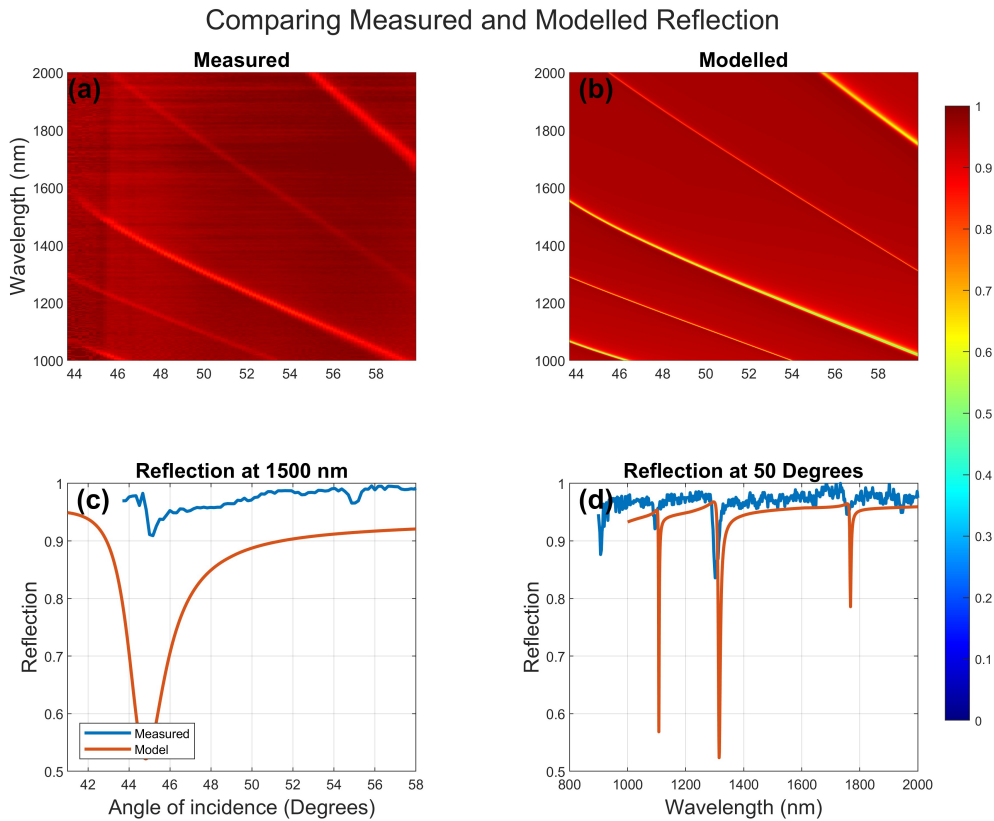


FIGURE 5.1: Measured reflection for unpolarized light for a sample with an estimated thickness of 1500 nm. (b) Modeled thickness was set to 1500 nm. (c) Measured and modeled reflection spectra at 1500 nm as a function of angle of incidence. (d) Measured and modeled reflection spectra at 50 degrees. Measured modes appear broader and less clear than their modelled counterparts due to the low resolution of the spectrometer relative to the linewidth of the reflection dips.

To make a comparison between modeled and measured reflection, we used Lumerical STACK to perform modeling of the reflection using the Transfer Matrix Method. We set the thickness of the layers to match those deposited, the

refractive index of SiO_2 and PMMA were set to 1.51 [42], which is dispersionless. For the refractive index of Au, we used the Johnson and Christy values [25] and for Cr, we used the CRC dataset [26].

In Figure 5.1, we show the measured unpolarized reflection for a sample in the NIR, we see good agreement between the position of reflection dips in both model and experiment. However, the measured reflection dips are weak and broader than their modeled counterparts, which is the result of the spectrometer lacking the resolution to resolve the spectral linewidth of these dips and the divergence of the incident beam being larger than the angular line width of the resonances. Given the resolution limitation of our NIR spectrometer, we move our study to the visible spectrum, as our visible spectrometer has higher spectral resolution.

In Figure 5.2, the reflection spectra from wavelengths in the range of 400-1000 nm are defined by the appearance of several narrow reflection dips moving to larger angles with shorter wavelengths. By changing the polarization of the incident light in Figure 5.2 (c), (d), (e), (f), we can select the s-polarized or p-polarized reflection dips. The s-polarized modes are noticeably narrower than their p-polarized counterparts, which is in agreement with measured linewidths for Si_3N_3 and SiO_2 samples that we studied in the previous chapter.

Comparing measured reflection to modelled, we see a generally good agreement at all studied angles. The lack of data below the critical angle is the result from the set up built for these measurements and is detailed in Chapter 3, section 3.2.2. It is important to note that during the measurement, the beam moves across the surface, the area of the sample measured is 1x1 cm, meaning that we can infer a very high surface uniformity and roughly constant thickness across

the 1 cm region measured.

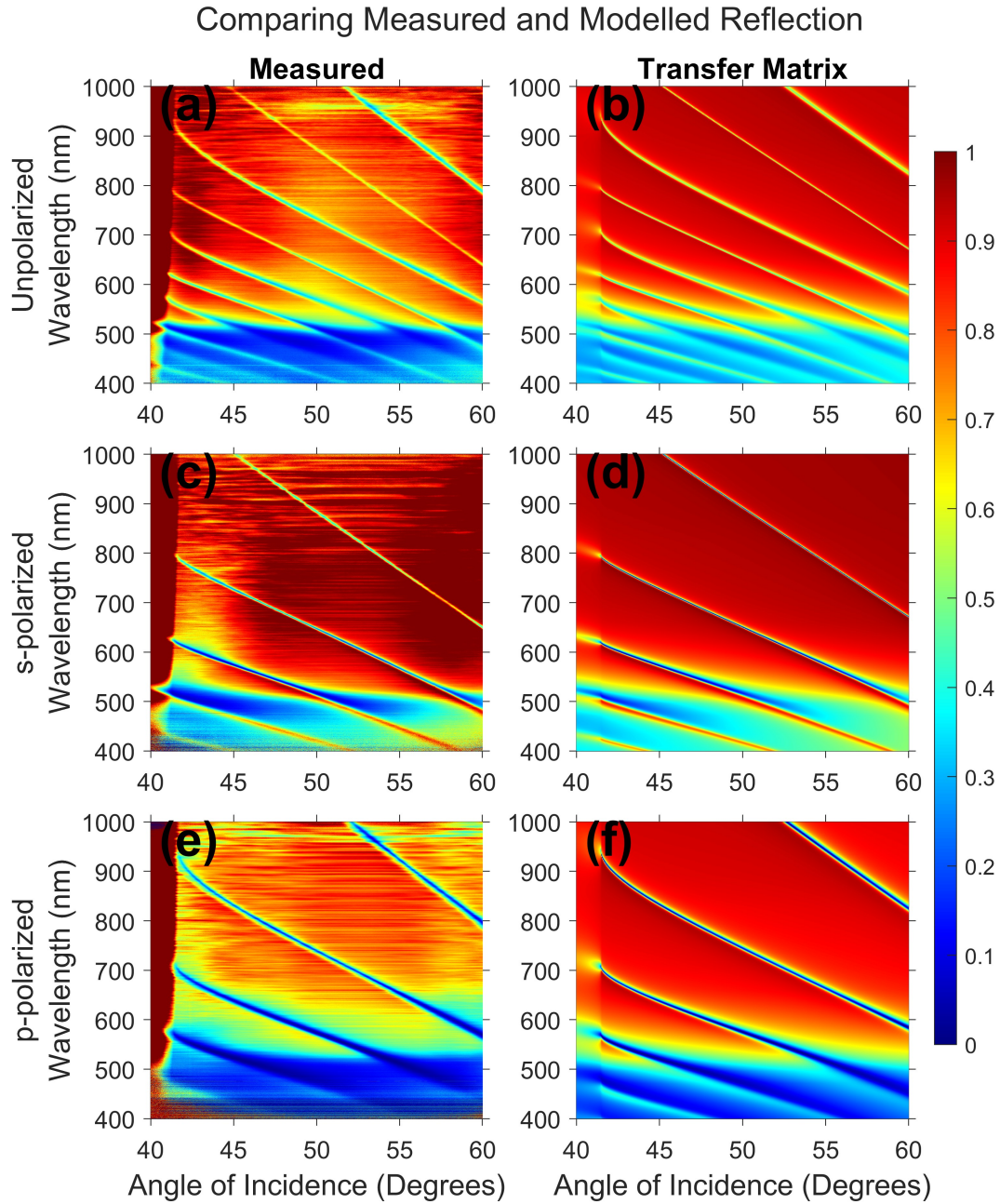


FIGURE 5.2: Comparison between measured and modelled reflection for the Au-PMMA structure for wavelengths in the range of 400-1000 nm and angles in the range of 40-60 degrees. (a)-(b) Measured and modelled reflection for unpolarized light. (c)-(d) Measured and modelled reflection for s-polarized light. (e)-(f) Measured and modelled reflection for p-polarized light.

To further investigate the agreement between model and measurement, we plot the reflection as a function of angle of incidence at various wavelengths in Figure 5.3. We note that p-polarized modes are much broader than their s-polarized counterparts, and that predicted and measured angular positions are typically in agreement. The only case this was not true was for Figure 5.3 (e), where the measurement was too noisy to identify any reflection features, which we attribute to the weak signal to noise of the tungsten halogen lamp at 450 nm. This issue is shown in reference measurements in Chapter 3, Figure 3.13.

We continue the comparison in Figure 5.4, where we study the reflection as a function of wavelength for various angles. The overall agreement between measured and modelled resonance wavelength is good regardless of the dataset.

The most striking result is the transition in lineshapes as the wavelength is reduced. In Figure 5.3, we saw a reduction in reflection off resonance regardless of polarization, which is seen in Figure 5.4 as well, this can be explained as the reflectivity of metal layers is lower at shorter frequencies, as expected of the Au layer. This allows for the non-resonant transmission into the PMMA. However, the asymmetric nature of these lines shapes is much clearer, such as Figure 5.4 (c), where a line shape with strong reflecting peak and low reflecting minimum as close together at 500 nm and 50 degrees. Similar features can be seen for p-polarized reflection, for example, Figure 5.4 (d), but they are not as dramatic as the s-polarized case. In addition, the resonances have transitioned from strongly absorbing/low reflection minima at larger wavelengths to weakly absorbing/highly reflection maxima at shorter wavelengths. The shorter the wavelength, the broader the resonance.

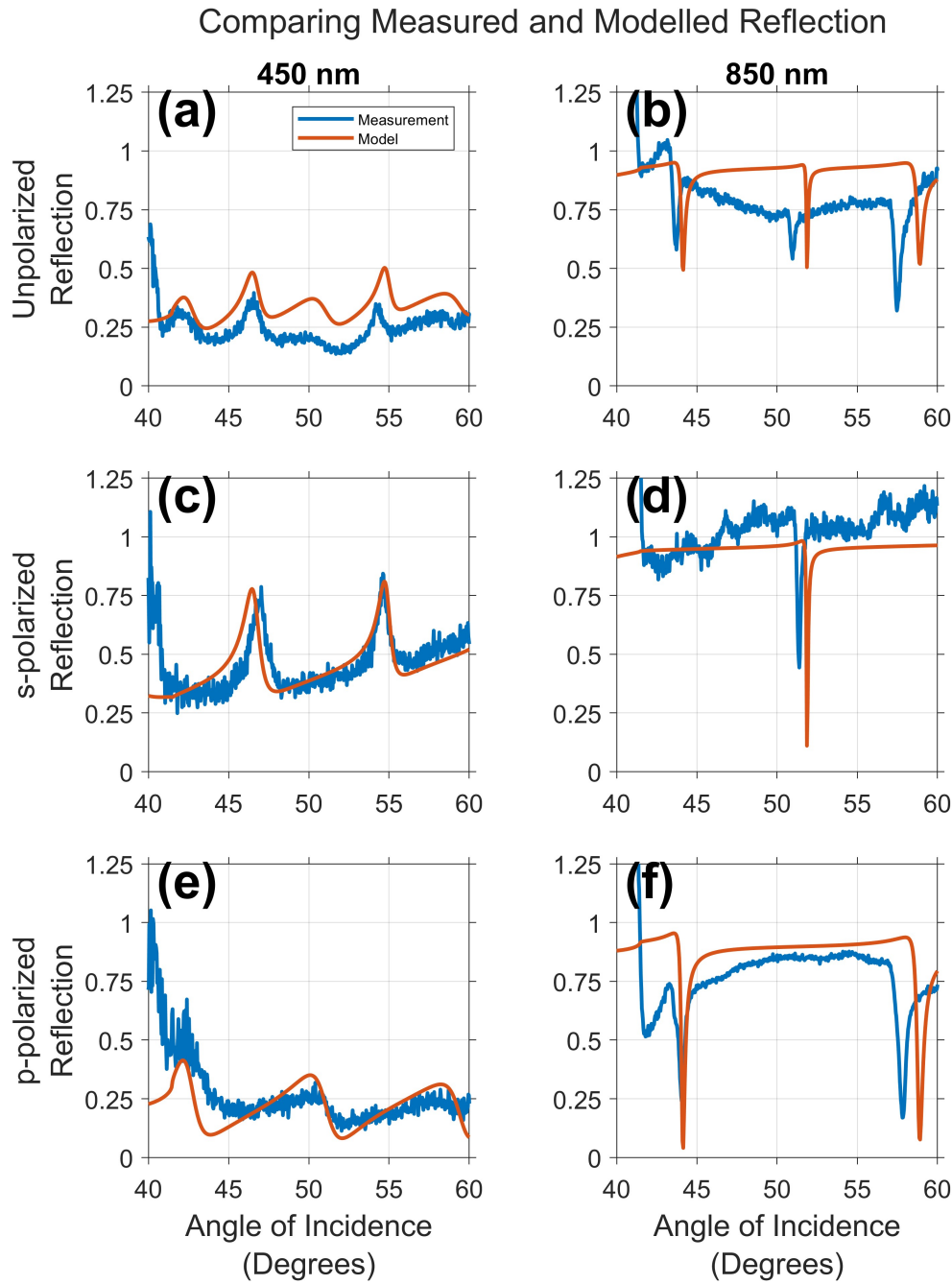


FIGURE 5.3: Reflection as a function of angle of incidence for various wavelengths and polarization. Orange line is predicted reflection from transfer matrix method. Dataset is the same data presented in Figure 5.2.

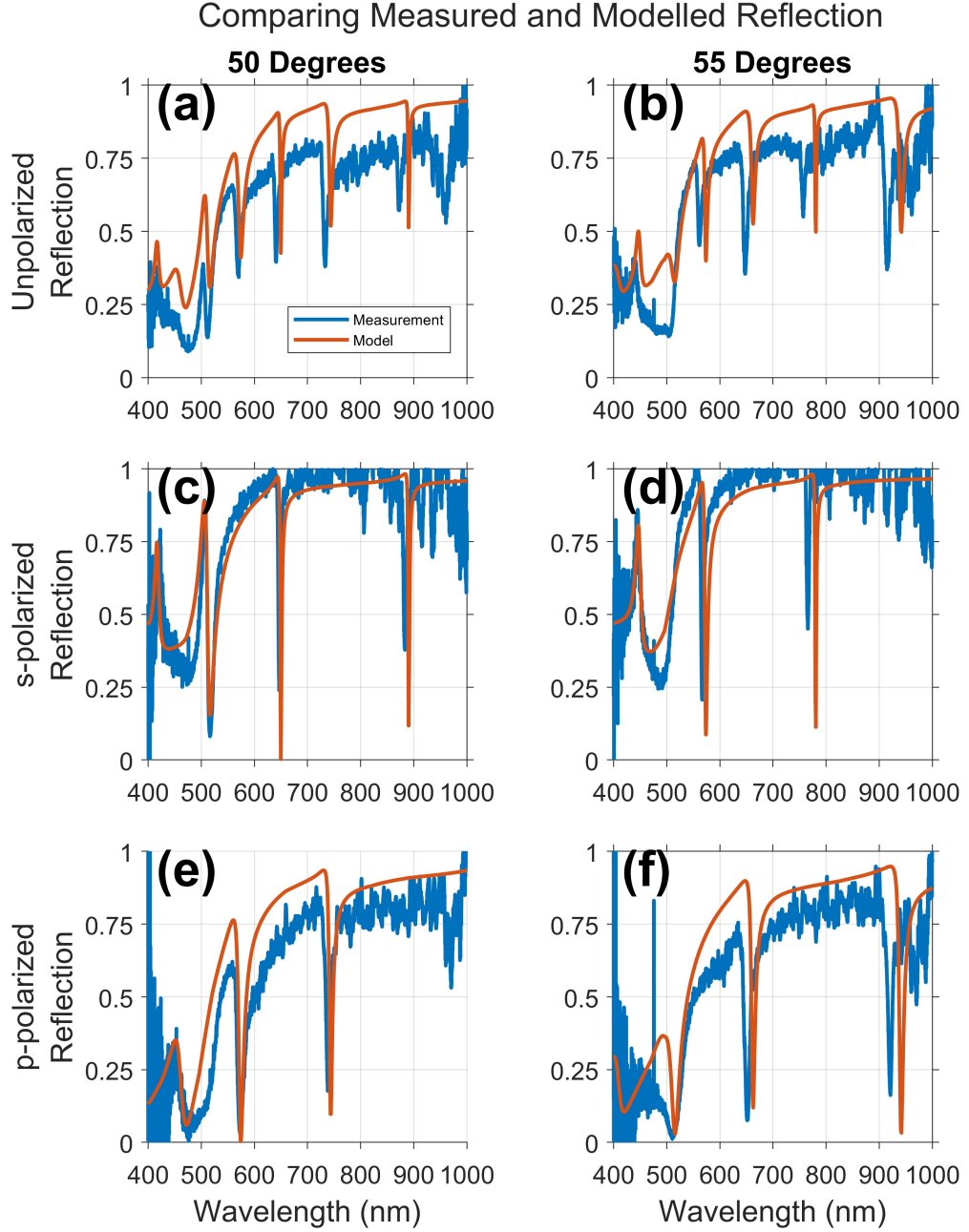


FIGURE 5.4: Reflection as a function of wavelength for various angles of incidence for all polarizations. Orange line is predicted reflection from transfer matrix method. Dataset is the same data presented in Figure 5.1.

We can further explore this structure through the transfer matrix method, by

changing the PMMA thickness to half and twice that of the PMMA layer studied, that is 600 nm and 2400 nm respectively. In Figure 5.5, we plot the unpolarized reflection for 600, 1200 and 2400 nm respectively. It is seen that the reflection peaks are blue shifted with increasing thickness as more modes begin to appear. This suggests that these resonances are Fabry Perot modes whose resonant position is moving with the changing thickness of the PMMA cavity.

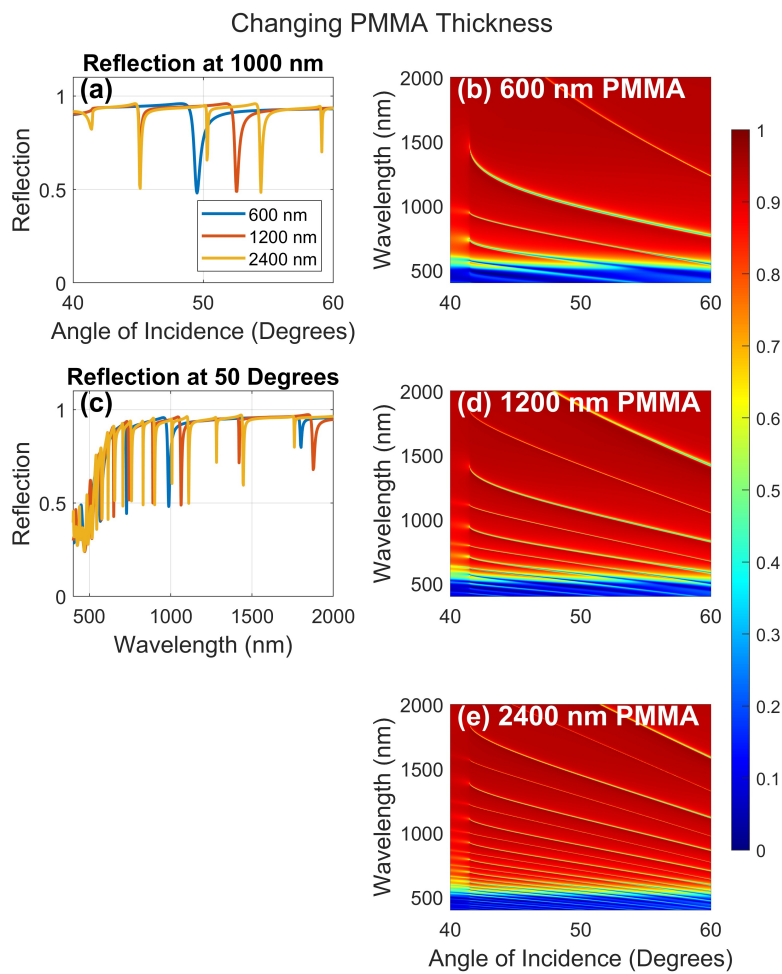


FIGURE 5.5: Modeled unpolarized reflection for 600, 1200 and 2400 nm of PMMA, thicknesses of Au and Ti are kept constant.

Questions regarding the reflection below the critical angle remain. As highlighted in Chapter 3 Figure 3.11, our set up is not capable of measuring reflection below the critical angle and relies on this issue for angle calibrations. However our transfer matrix allows for the modeling of this reflection, which is shown in Figure 5.6 for 0-, 15-, 30- and 45-degrees angles of incidence.

We find that for normal incidence, s- and p- polarizations are the same, which was demonstrated in the previous chapter for a Si_3N_4 thin film, increasing angles shows a deviation in s- and p-polarization, in particular, the resonances of the s-polarized cases are narrower, and more well defined then their p-polarized counterpart. These resonances are expected to be typically Fabry Perot modes studied in the previous chapter.

The most dramatic change is seen at 45 degrees, which corresponds to the narrow line shapes we have been studying. This dramatic change may be best understood as a change in reflection at the PMMA-Air interface, which undergoes total internal reflection at 41 degrees and with reflectivity equal to 1, as highlighted in Chapter 4, Figure 4.21 and equation 4.18.

As highlighted in our investigation into SiO_2 thin films, we demonstrated how s-polarized lineshapes are narrower then their p-polarized counterparts as the angle of incidence increased, and how that could be interpreted as resulting from an increase in reflection, which varies dramatically in the vicinity of the critical angle. However Chapter 4, Figure 4.21 and equation 4.18 does not explain the change in FSR or resonant wavelength with increasing angle of the Fabry Perot modes, suggesting additional factors must be considered.

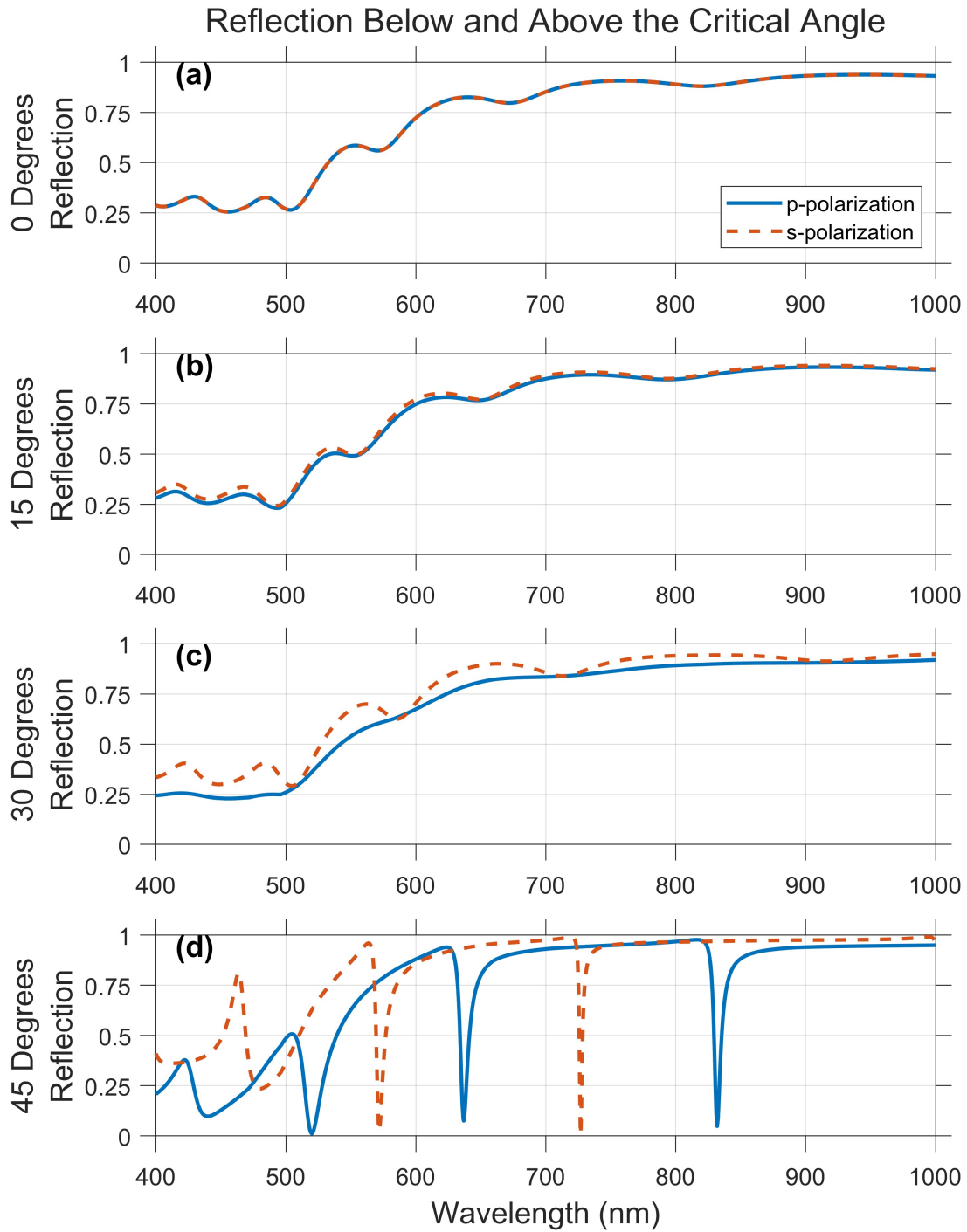


FIGURE 5.6: Modelled Reflection for various angles of incidence across the visible spectrum. PMMA thickness is 1200 nm, Ti thickness is 1 nm and Au thickness is 40 nm for all these datasets.

5.2.1 Determining Asymmetry of Lineshape

The asymmetric line shapes we have observed so far are commonly referred to Fano line shapes [94] and are distinctly different from the symmetrical Lorentzian line shapes as we found for the Fabry Perot resonances in Chapter 4. The observation of Fano line shapes is typically associated with coupling between a discrete (low loss) state and continuous state (lossy) and seen in metamaterials [84] where different modes couple and exchange energy with each other. The Fano line shape is described by the following equations.

$$\sigma(x) = \frac{D^2(q + \Omega)}{1 + \Omega^2} + a \quad (5.1)$$

$$\Omega = \frac{2(x - x_0)}{\tau} \quad (5.2)$$

where q is the Fano parameter describe the degree asymmetry of the line shape. D is associated with the phase shift of the continuum, τ is the resonance line width, x_0 is the resonant frequency/angle and a is the background fitting, which adjusts the height of the line shape to match the off resonance absorption. In Figures 5.7 and 5.8, we fit the above equation to the calculated unpolarized absorption, which was calculated by subtracting 1 from the measured reflection, given that the sum of reflection and absorption should be equal to one, as transmission is zero, and we assume energy conservation.

In Figure 5.7, comparing the angular line widths for model and experiment

shows a good agreement at larger angles, but noise in measurements makes it difficult to fit the Fano equation to resonances at smaller angles.

In Figure 5.8, comparing the spectral line widths shows similar difference at shorter wavelengths, where the background may be introducing further error in the fitted parameters.

Our primary interest is in the Fano parameter, which is an indication in the degree of coupling between the discrete and continuous states. No coupling occurs if the Fano parameter is either 0, or at an exceptionally large value. In these cases, the asymmetric Fano line shape becomes a symmetric Lorentzian line shape.

The asymmetric line shapes are quite clear in the modeled absorption, and are occasionally obscured in the measurement, which can be attributed to noise in the measurement. In Figure 5.7, our fitted Fano parameters show good agreement between experiment and model, except for the resonance at 59 degrees, in this case the fit to the measurement appears symmetric, rather than the expected asymmetric line shape. This can also be seen in the fitted Fano parameter, which appears to be quite large, indicating a line shape that is more symmetric than other line shapes.

In Figure 5.8, the agreement in fitted Fano parameters is much better. Overall, the Fano parameter is small enough to produce asymmetric line shapes. This small, non-zero value for the fitted Fano parameter demonstrates the potential for coupling between the Fabry Perot mode in the PMMA layer, whose presence is evident in the resonance shifts with changing PMMA thickness in Figure 5.7 and 5.8, and a lossy plasmonic mode, being excited by s-polarized

light, which suggests a TE surface plasmon.

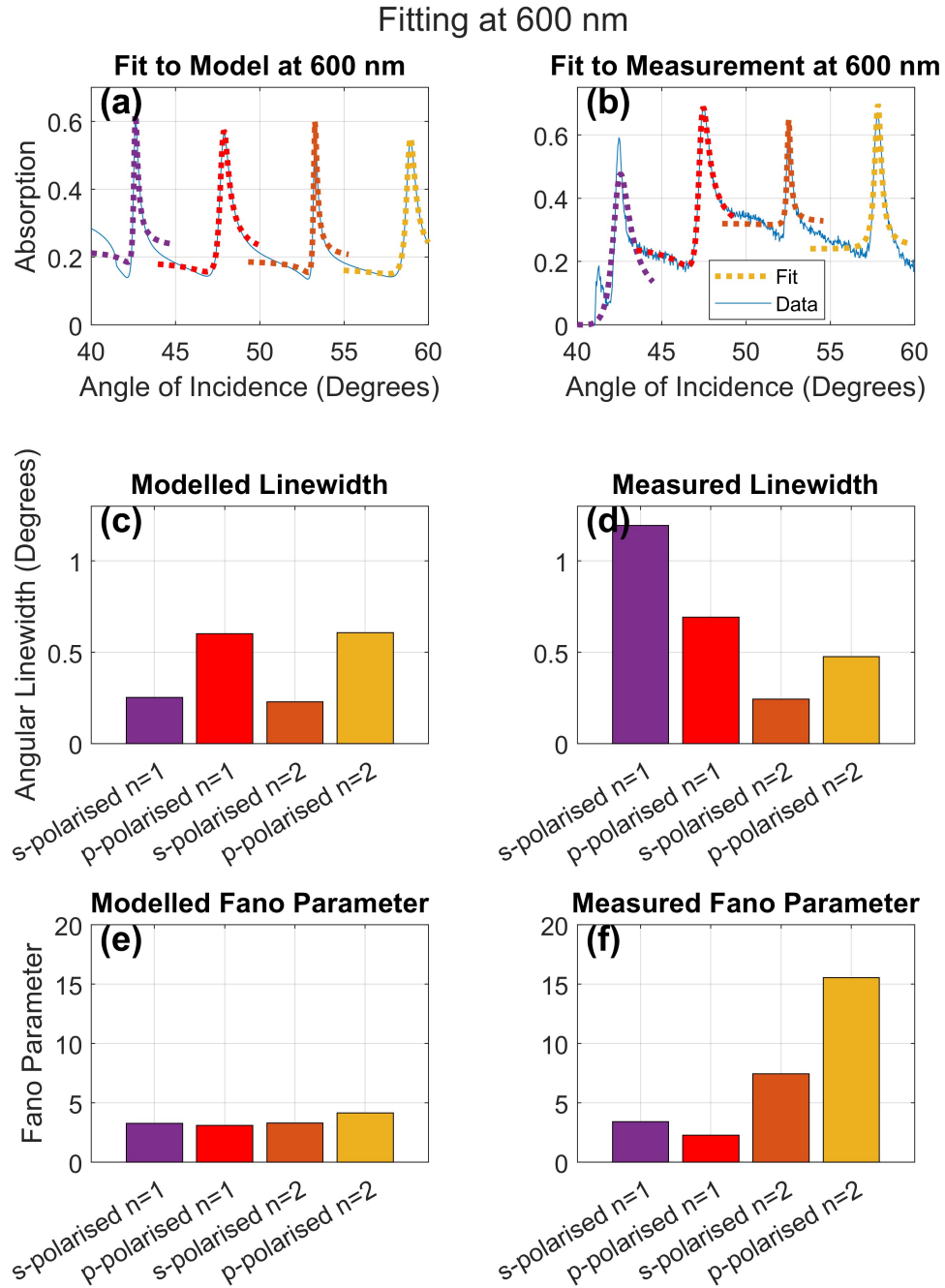


FIGURE 5.7: Fitting to absorption as a function of angle of incidence at 600 nm. (a) Plot of modeled unpolarized absorption and fitting to absorption peaks. (b) Plot of measured unpolarized absorption and fitting to absorption peaks. (c) Bar chart of fitted line width extracted from (a). (d) Bar chart of fitted line width extracted from (b). (e) Bar chart of fitted line width extracted from (a). (f) Bar chart of fitted line width extracted from (b).

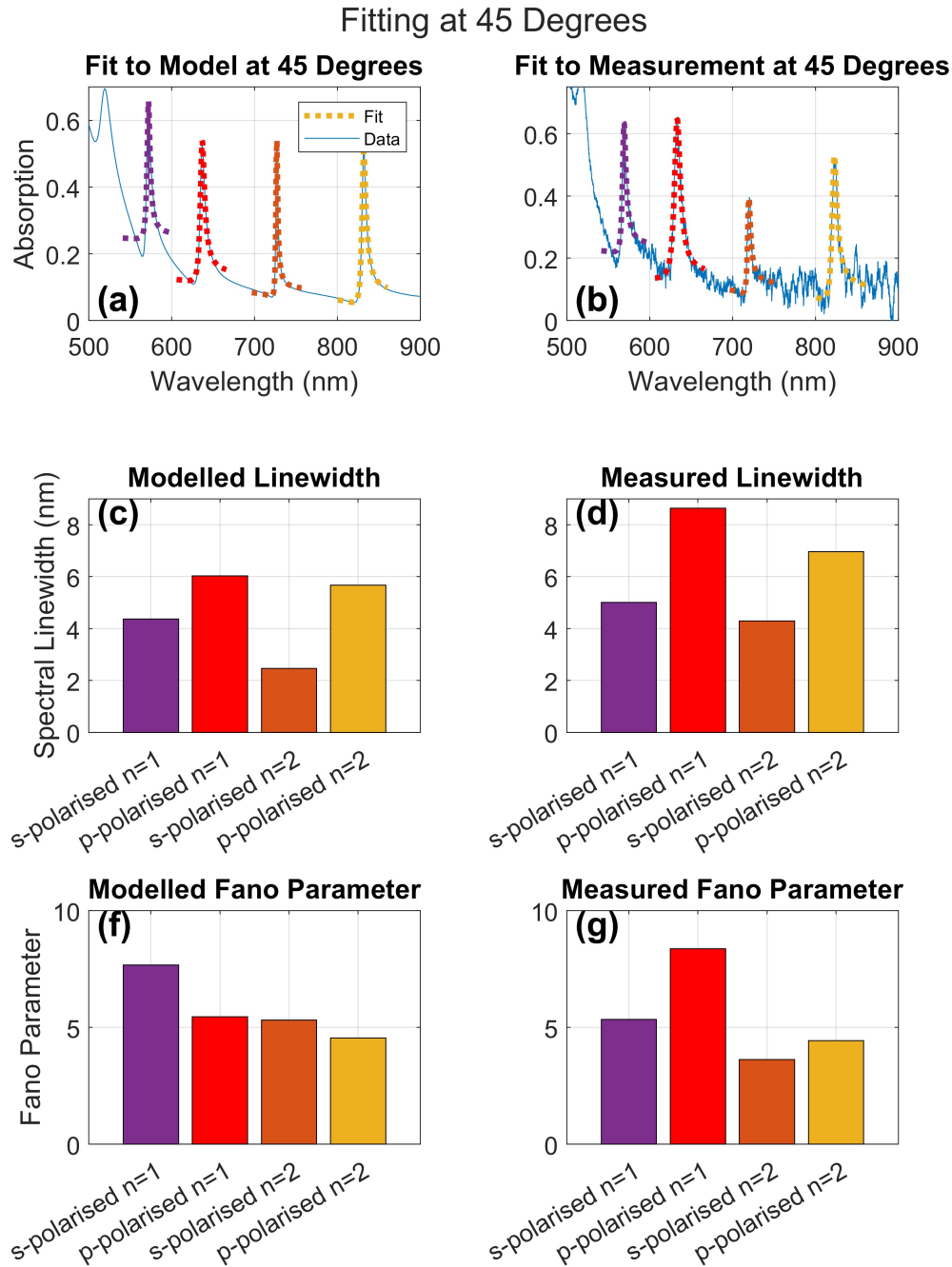


FIGURE 5.8: Fitting to absorption as a function of angle of incidence of 45 Degrees. (a) Plot of modeled unpolarized absorption and fitting to absorption peaks. (b) Plot of measured unpolarized absorption and fitting to absorption peaks. (c) Bar chart of fitted line width extracted from (a). (d) Bar chart of fitted line width extracted from (b). (e) Bar chart of fitted line width extracted from (a). (f) Bar chart of fitted line width extracted from (b).

5.3 Modal Analysis of TE and TM Modes in PMMA Layer

To explore the nature of the s- and p-polarized modes, eigenmode analysis was performed through Lumerical FDE to find resonant modes at the wavelengths and angles we studied in Figure 5.2. The thicknesses and refractive indices of the layers remain the same as in the transfer matrix method previously.

Eigenmode analysis is typically used in the study of optical and plasmonic waveguides, being able to identify resonant modes within a structure. In our case, we searched for resonances from 400 – 2000 nm and effective indices from 1.0 - 1.51, which correspond to angles of incidences between 41-90 degrees.

The effective indices of these resonant modes were converted to angles of incidence through the equation, $\theta_{incidence} = \sin(\frac{n_{eff}}{n_{SiO_2}})^{-1}$, and plotted over the modeled reflection spectrum in Figure 5.10 (a) and (b). It was found that single modes were found at the wavelength and angles where the reflection dips are were observed in the modeled data. We note a slight error in resonance positions, which is roughly 5% at most, which we attribute to numerical error in introduced PML boundary conditions into the simulation.

The wavelength and angular positions of the TE and TM modes are plotted as lines over the reflection peaks, indicating that the s-polarized reflection dips are the result of the TE modes and p-polarized reflection dips are the result of TM modes . We also note the appearance of the symmetric surface plasmon in Figure 5.10 (b) seen by the very broad resonance at larger angles and longer

wavelengths. This mode has been moved to longer wavelengths relative to the dispersion we plotted in Chapter 2, Figure 2.6, which is the effect of the PMMA film, changing it's thickness would move location of the symmetric surface plasmon.

TE and TM modes are definitions applied to optical and plasmonic waveguides, defining the polarization of these modes, similarly to s- and p-polarized definitions for plane waves. TE stands for Transverse Electric, which defines a mode with one electric field component, and 2 magnetic field components. The electric field is transverse to the direction of propagation, while the magnetic fields are both transverse and longitudinal. TM stands for Transverse Magnetic, which defines a mode with two electric and one magnetic field component. The magnetic field component is transverse to the direction of propagation, while the electric fields are transverse and longitudinal to the direction of propagation.

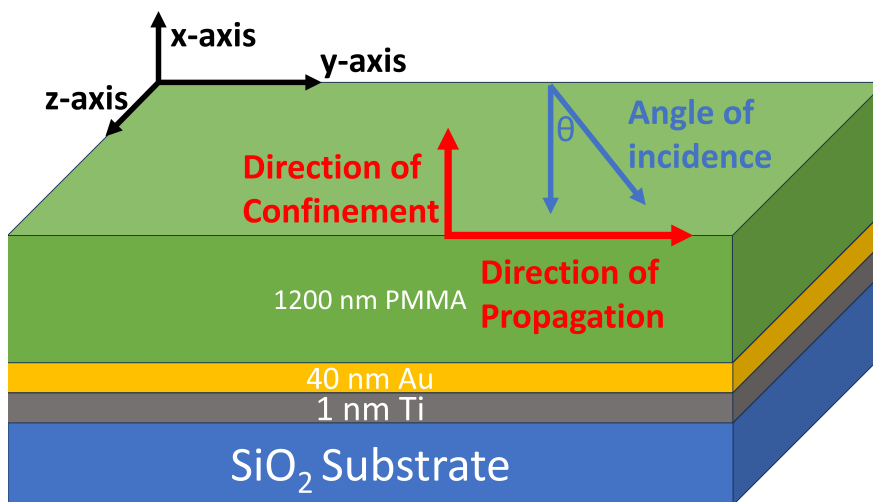


FIGURE 5.9: 3D Illustration of simulation for modal analysis. All modes propagate along the y-direction, with the thickness of each layer defined along the x-direction. Actual simulation is a 1D simulation assuming the x-direction is finite, and all other directions are infinite. The angle of incidence is shown by the blue arrows and is along a direction in the x-y plane.

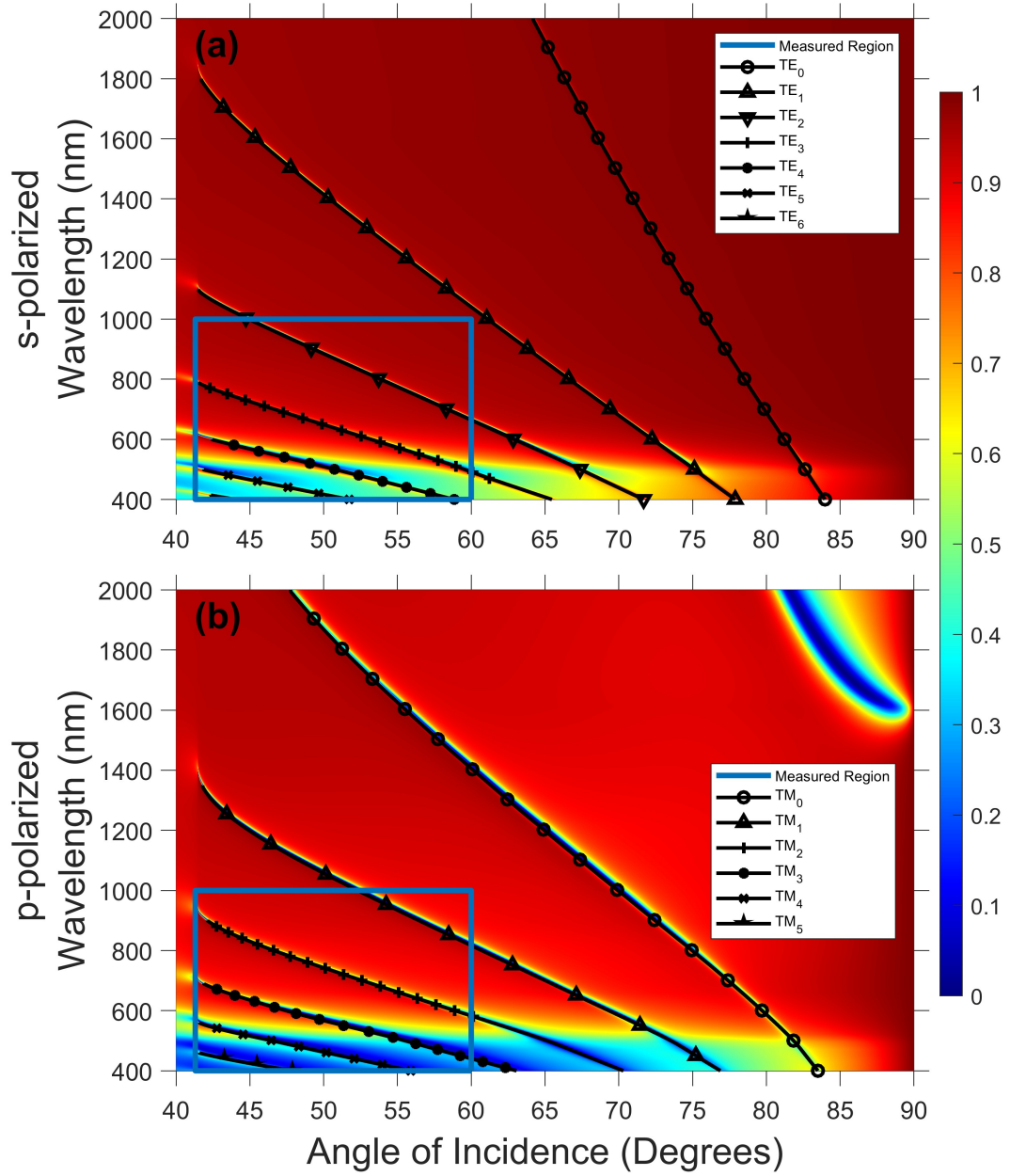


FIGURE 5.10: Modeled reflection for s and p-polarized light. The blue box marks the region measured in Figure 5.2. We note the presence of the symmetric surface plasmon at long wavelengths and large angles in subplot (b).

Eigenmode analysis also provides the near fields of the modes in Figure 5.10, we plot the near field profiles of the TE_1 and TE_2 as well as the TM_1 and TM_2 at 900 nm in Figures 5.11 and 5.12. The number indicated the number of additional half wave patterns seen in the PMMA layer, for example the TE_0 has a single half wave pattern and TE_1 has two half wave patterns. These are standard definitions used to define waveguides. These modes are the result of FP like resonances confined to the PMMA layer, with a radiative losses seen in the SiO_2 substrate, which is absorbed by the PML boundary conditions.

In the case of TM modes in Figure 5.11, we observe two electric field components along the x-direction, which is the direction of confinement that the film thickness are defined along, as well as along the y-direction, which is direction of propagation. A single magnetic field component is seen along the z-direction, in plane with the films, but is perpendicular to the direction of propagation.

Discontinuities in electric fields in Figure 5.11 (a) and (b) are the result of different permittivities between mediums, as per the electromagnetic boundary conditions between media [95, 36], which states that electric fields perpendicular to the surface, i.e, along the x-axis, are discontinuous if the permittivities are unequal.

In Figure 5.12, we see a similar result in the s-polarized/TE modes, most of the field is confined to the PMMA layer like a Fabry Perot mode, however there are a single electric field component and two magnetic field components. No discontinuities are seen in the electric field, given that there is no electric field perpendicular to the interfaces. No discontinuities are seen in the H fields perpendicular to the surface, or along the x-axis, which is expected, given that

the material permeability is set to 1.

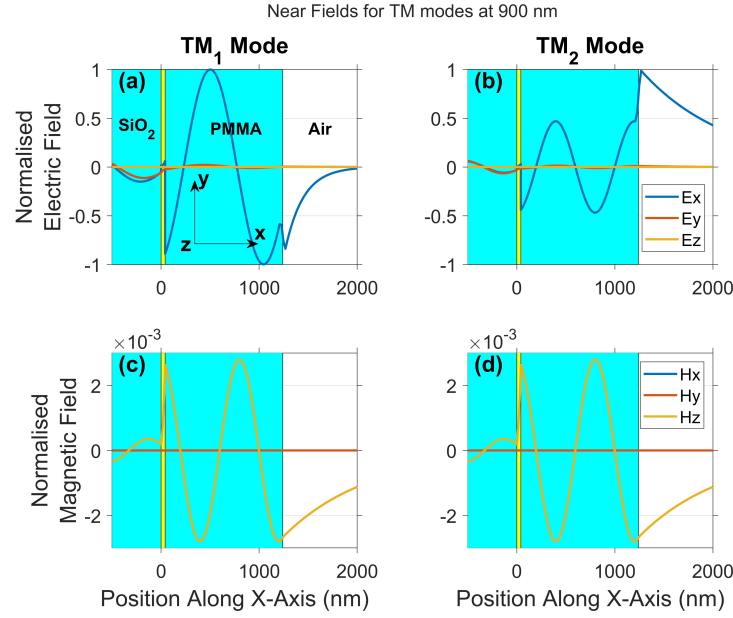


FIGURE 5.11: Near field profiles of the TM_1 and TM_2 modes plotted in Figure 5.10 (b) at 900 nm the TM_2 mode was directly measured, while the TM_1 exists at an angle outside of our range of measured angles.

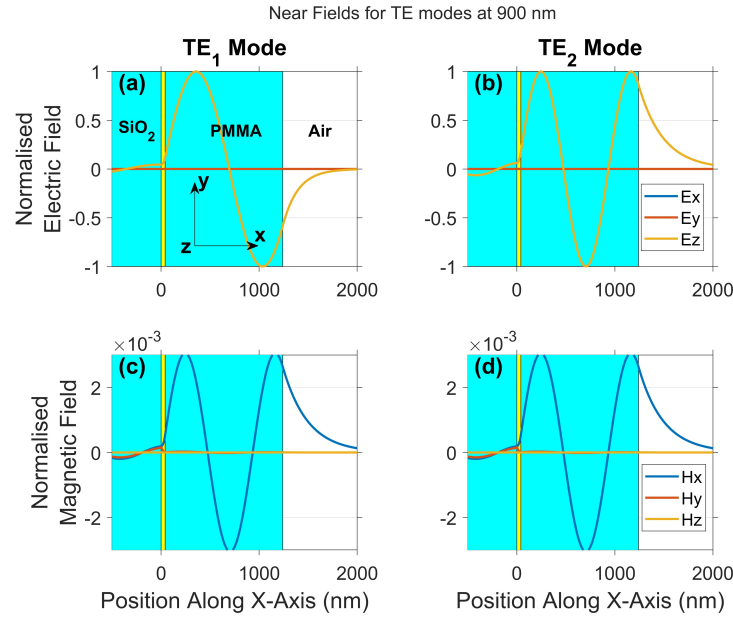


FIGURE 5.12: Near field profiles of the TE_1 and TE_2 modes plotted in Figure 5.10 (a) at 900 nm, the TE_2 mode was directly measured, while the TE_1 exists at an angle outside of our measurement range. Note in all cases, the evanescent tail into the air.

The values of the electric field are normalized with respect to largest electric field, and hence 1 corresponds to the point where the electric field is the strongest. The magnetic field is normalized relative to this electric field as well, given that there is a relation between the magnitude of the electric and magnetic fields, $E = \mu_0 \epsilon_0 H = Z_0 H$, where Z_0 is the impedance of free space and equal to 377 ohms. Therefore if $E = 1$, the largest magnetic field would be $H = \frac{1}{377} \approx 2.65 \times 10^{-3} A m^{-1}$, this matches the largest magnetic field value in Figures 5.11 and 5.12.

5.3.1 Identifying the TE Surface Plasmon

To identify a surface wave, we must look at the magnetic field along the direction of propagation, that is along the y-axis, for TE surface waves, as this a common feature of TE surface plasmons [14, 15]. Surface plasmons are surface waves with both transverse and longitudinal components whereas Fabry Perot modes are purely transverse, observing a magnetic field in the longitudinal direction or along the direction of propagation in the metal layers would be a clear indication of a TE surface plasmon.

In Figure 5.13, we plot the magnetic field along the direction of propagation across the wavelengths that the TE_1 and TE_2 modes are present across the visible and NIR regions. We note that regardless of mode order, there is some non-zero magnetic field at the SiO_2 -Ti interface, the non-zero electric field in the SiO_2 layer is the product of radiative losses.

We note that below 500 nm, there is a transition in the location of the magnetic

field from the interface to the center of the Au layer, which matches the shift in lineshapes over the same wavelength range which was identified in Figure 5.3 for s-polarized modes. These localized magnetic fields along the direction of propagation are a clear indicator of a TE plasmon, however we can explore the wave in more detail by modifying the structure as seen in Figure 5.14.

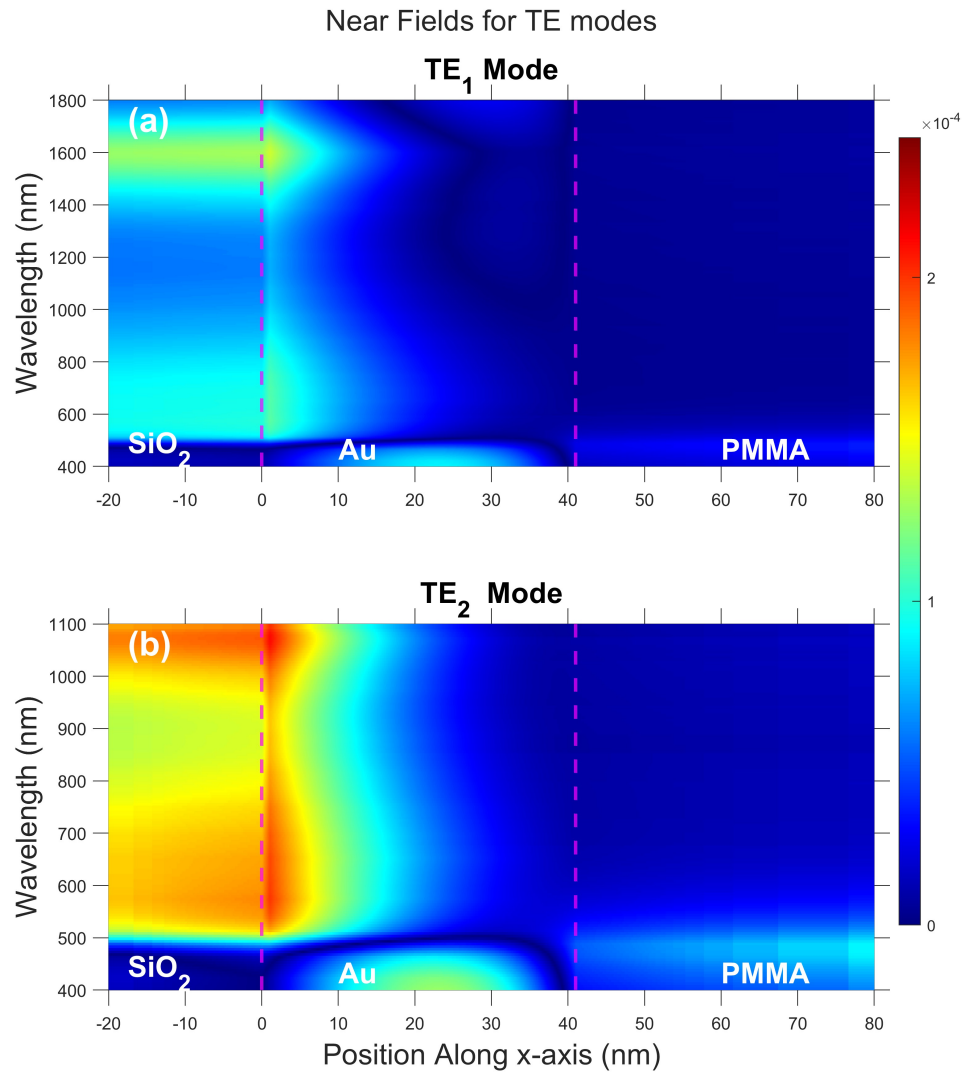


FIGURE 5.13: Demonstration of TE surface wave in TE₁ and TE₂ modes. (a) Magnetic field along direction of propagation for TE₁ mode. (b) Magnetic field along direction of propagation for TE₂ mode. Colour map is magnetic field in A/m.

The surface waves can be more clearly seen by separating the PMMA from the Au layer, as seen in Figure 5.14 (c) and (d). Lower index spacer regions are typically used to optimize the coupling between waveguide modes and TM surface plasmons [96]. By introducing a $1\text{ }\mu\text{m}$ spacer layer of $n=1$ in between the Au and PMMA layers, we may optimize coupling between plasmon and cavity mode.

For the TE case in Figure 5.14 (c), the evanescent tail into the air is made clear, while the same field in the metal is maintained. For the TM case in Figure 5.14 (d), the TM surface plasmon appears, with a similar evanescent tail into the air.

Finally, to simplify the structure further, we replace the SiO_2 substrate with an thick Au film in Figure 5.14 (e) and (f). This clearly shows both TE and TM surface waves being excited. A unique feature of the TE surface plasmon is the large negative value for magnetic field in the metal, which is present in Figure 5.13 and Figure 5.14 (a), (c) and (e). The TM surface plasmon has a much smaller, negative value for the electric field in a similar location in Figure 5.14 (f).

In addition, the magnetic field profile of our TE surface plasmon in Figure 5.14 (e) matches an observation of an “artificial TE surface wave” observed in Ag substrates covered by sub wavelength dielectric films of $n=4$ [15] at similar wavelengths and discussed in Figure 1.4. A similar observation in field profile was made in hyperbolic metamaterials [16], where the “bump” in the metal region, seen throughout Figures 5.13 and 5.14 in the TE mode, was observed for a TE surface plasmon in metallic multilayers, also discussed in Figure 1.4.

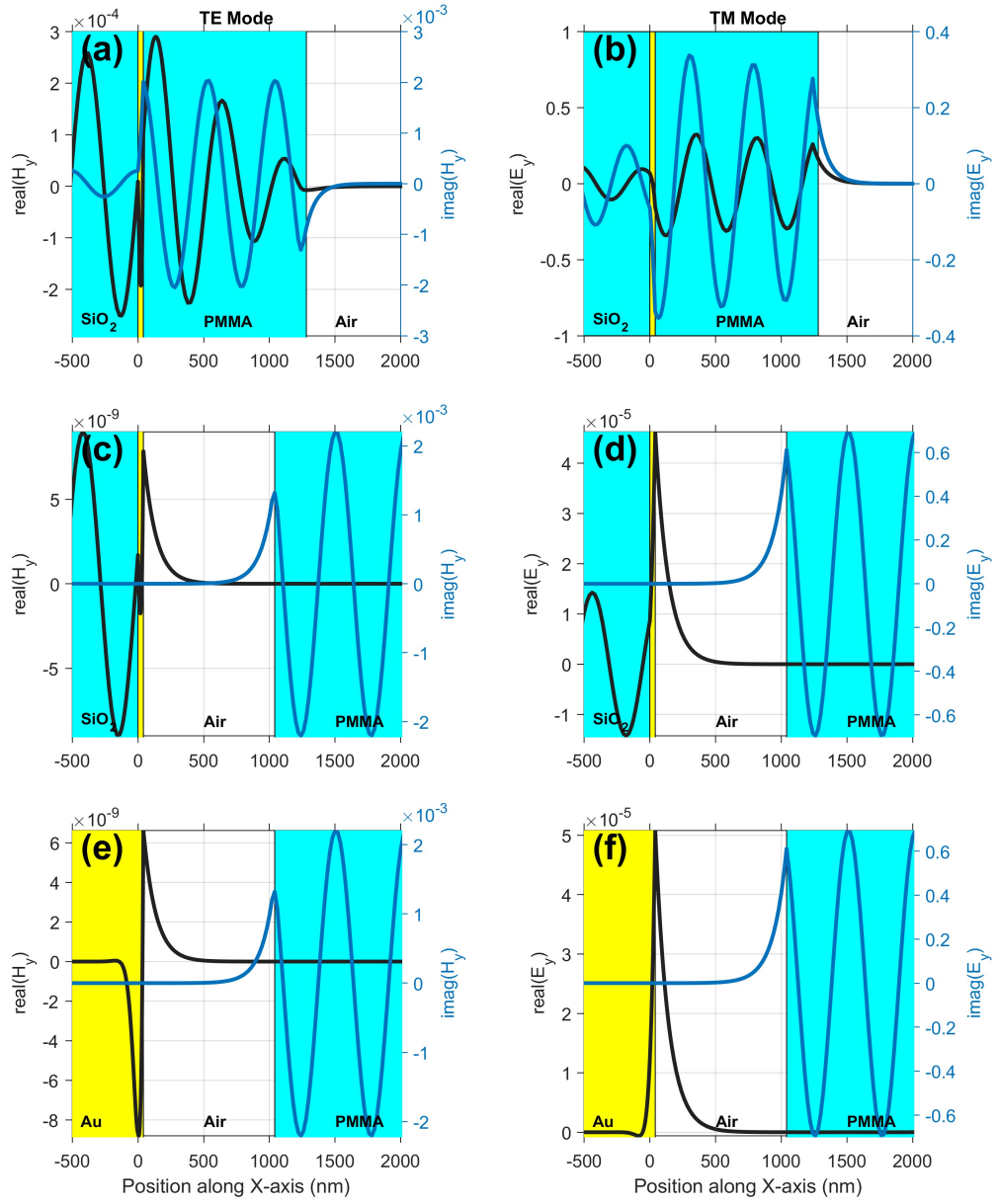


FIGURE 5.14: Change in TE and TM surface waves with introduction of spacer layer and Au substrate for TE₂ and TM₂ modes at 900 nm. (a)-(b) Fields with no variation. (c)-(d) Fields with 1 μm $n=1$ space between Au and PMMA. (e)-(f) SiO₂ substrate converted to Au.

We can further exploring the coupling to this TE surface plasmon, by changing the thickness of the $n=1$ spacer layer in figure Figure 5.14 (e). This analysis is shown in Figure 5.15, where we set the thickness to 100, 300 and 500 nm. As the separation increases, the magnetic field strength, and hence to coupling is reduced, which is expected as the distance between the Fabry Perot mode and metal film affects the degree of coupling between modes.

At sufficiently small gaps, the TE plasmon radiates into the PMMA layer, as it's evanescent field overlaps with the PMMA layer, this would suggest that the effective index of the TE plasmon is near $n=1.51$, which is the refractive index of the PMMA, similar results were seen for TM plasmons in Chapter 2 Figure 2.7 and 2.8.

The overall confinement of this TE surface wave is weak, given the mode is radiative, it is weakly confined to the metal layer, however confinement can be improved through modifications to structure, i.e spacer etc, which may prove more optimal for practical confinement.

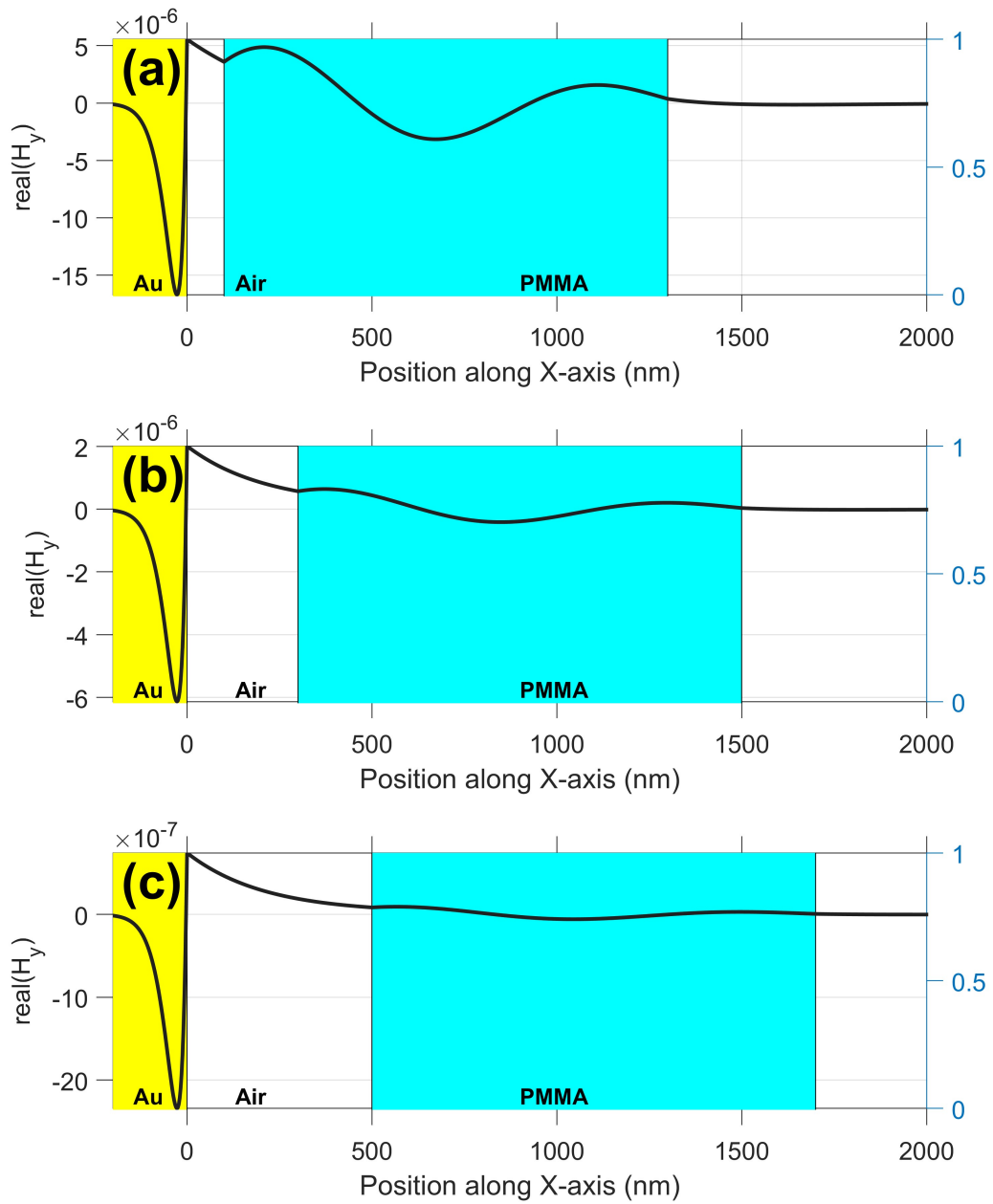


FIGURE 5.15: Effect of changing distance between thick Au layer and PMMA from Figure 5.14 (e). (a) 100 nm spacer distance. (b) 300 nm spacer distance. (c) 500 nm spacer distance

5.4 Propagation Length of TE Modes

A final parameter than can be extracted from eigenmode analysis is the propagation length of the mode, the effective index plotted in Figure 5.10 contains an imaginary component arising from loss from the mode, either from absorption or radiative losses. The propagation length defines the distance at which the mode retains 33% of it's initial energy, as serves as a vital parameter for practical applications for plasmonics. The propagation length is defined as follows [97, 98].

$$PL = \frac{1}{2k_{neff,imaginary}} \quad (5.3)$$

where $k_{neff,imaginary}$ is the imaginary part of the mode's wavevector as a function of wavelength.

$$k_{neff,imaginary} = \frac{2\pi n_{eff,imaginary}}{\lambda} \quad (5.4)$$

In Figure 5.16 we plot the propagation length of the TE₀, TE₁ and TE₂ and their TM counterparts. We find that lower order modes typically have higher propagation lengths then their higher order counterparts. It is also observed that TE modes have lower loss then their TM counterparts. This result agrees with the spectral measurements showing the TE modes are narrower than their TM counterparts.

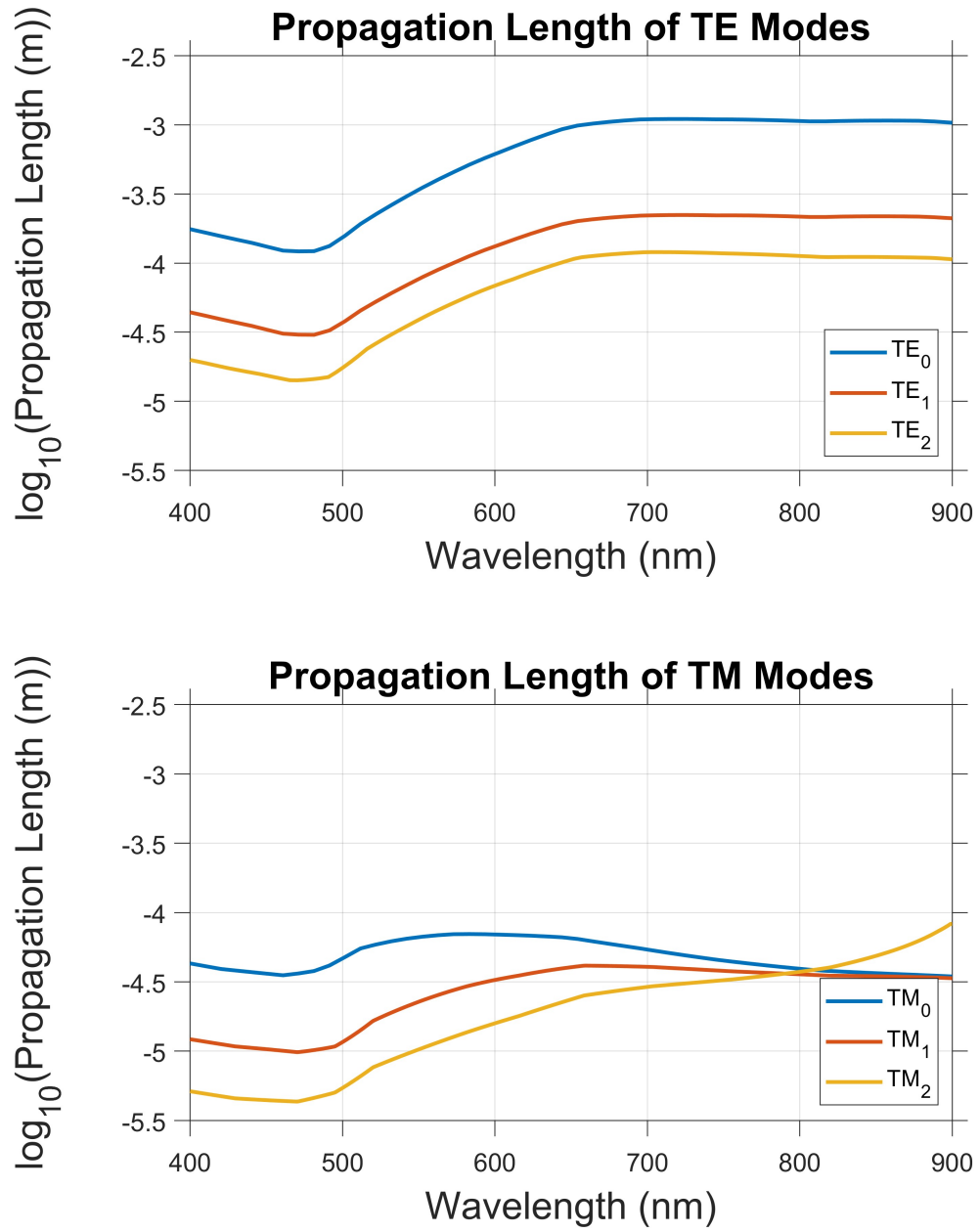


FIGURE 5.16: Propagation lengths of modes plotted in Figure 1 across the visible spectrum. Propagation lengths were calculated from the imaginary part of the effective modal index.

We note that TE modes have a minimum propagation length at below 500 nm, which matches the enhancement in magnetic field seen in Figure 5.12, suggesting that optimal coupling between FP modes and TE plasmon are achieved at that wavelength.

The propagation length of lower order TE modes is exceptionally large, ranging between 0.1-1 mm distances, this is roughly a 100 fold improvement over traditional TM plasmons, which are well known to have low propagation lengths that make applications more difficult, given that the propagation length of a TM plasmon is typically in the range of micrometers [99]. This parameter also plays a role in sensors, where high values may be desirable.

5.4.1 Role of Metal on Propagation Length

It is common to use different metals for TM surface plasmons, such as Ag, which has lower losses than Au and most other metals. To explore the effect of different metals on the TE modes, we investigate replacing Au with Ag and Cr for the TE₄ mode in Figure 5.17. It is found that the propagation length of Ag and Au cases is the same at longer wavelengths, while diverging at shorter wavelengths. However, we find that the Cr case is overall lossier, with much shorter propagation lengths than the Ag or Au case.

Furthermore, there seems to be little correlation between the change in propagation length with wavelength and the imaginary part of the refractive index for each metal. This lack of correlation is better understood by looking at the magnetic field in the metal layer as seen in Figure 5.17 (b), (d) and (e). We

see that Cr shows the largest magnetic field relative to Au or Ag at longer wavelengths, where we saw shorter propagation lengths. Ag shows no clear excitation of the TE surface wave at any wavelength, while Au only supports the surface wave at 500 nm, the region where propagation lengths were seen to deviate in Figure 5.17 (a).

This highlights the importance of selecting the correct metal for exciting the TE surface wave, we find that Cr is more optimal for increasing coupling between the Fabry Perot mode and the TE surface plasmon.

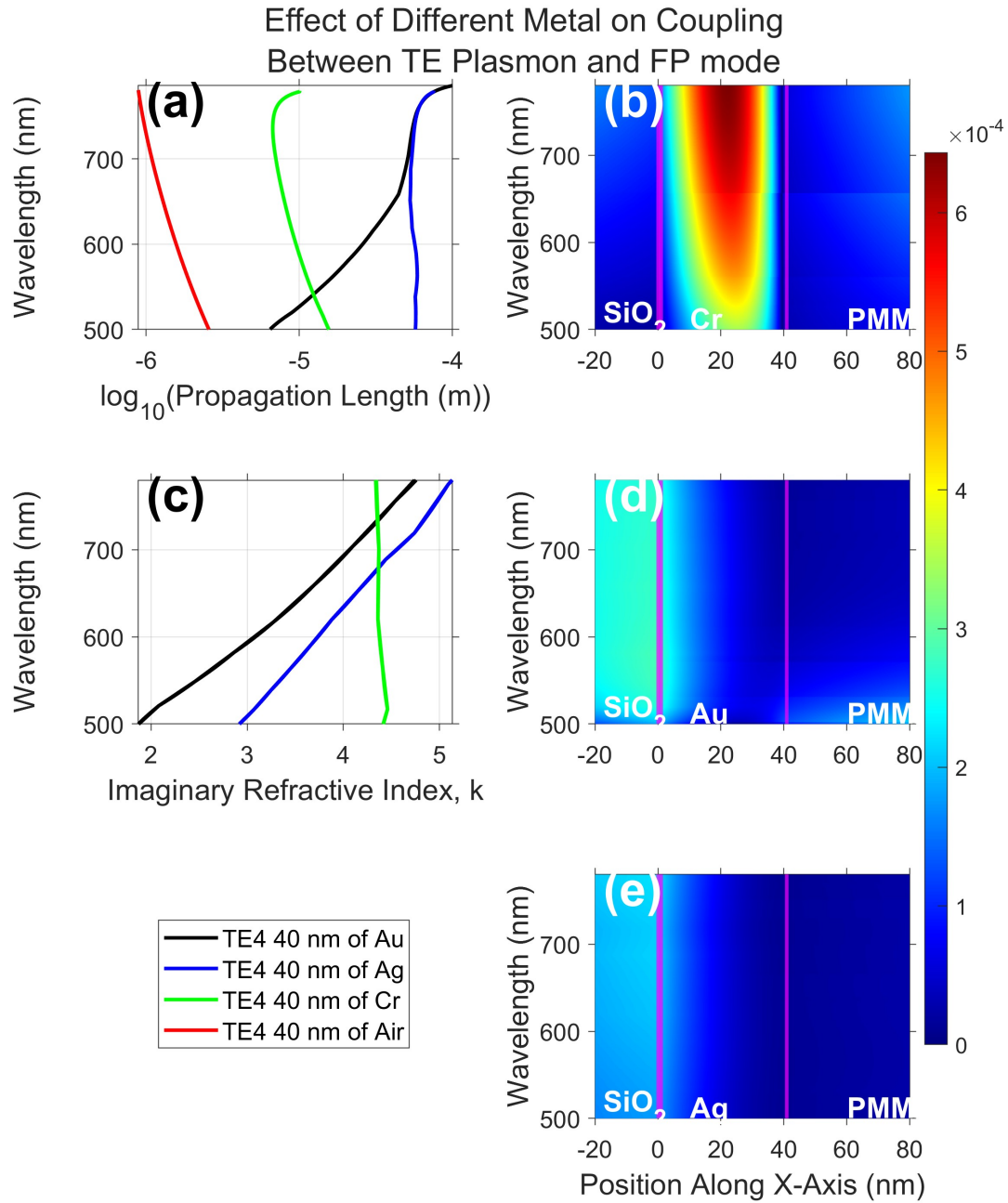


FIGURE 5.17: Study on the effect of changing the metal. (a) Propagation for Au, Cr, Ag and Air layers. (b), (d) and (e) Magnetic fields along the direction of propagation for different metals. (c) Absorption coefficients of metals for reference.

5.4.2 Conclusion

In this chapter we demonstrated how one may excite a TE surface plasmon by using a thin film of PMMA on top of a thin film of Au that is sufficiently thick to support Fabry Perot modes, which as highlighted in Chapter 4 are the result of interference between electric and magnetic multipoles, which can both contribute to spatial dispersion or non-unity values of magnetic permeability.

First we demonstrated that our fabricated sample matched our models, we then demonstrated how the location of the reflection dips may be engineered by changing both angle of incidence and thickness, with thicker films producing narrower modes. Unlike Chapter 4 we demonstrated that our modes are Fano lineshapes as opposed to the Lorentzian lineshapes seen in Chapter 3, which were the result of coupling to a lossy plasmonic mode, in the case of s-polarized reflection dips, this must be a TE plasmon rather than a TM plasmon.

Using eigenmode analysis, the reflection dips were demonstrated to result from a coupled Fabry Perot-Plasmonic mode, a combination of propagation lengths and magnetic fields along the direction of propagation were excellent indicators of the existence of this surface plasmon, with lower propagation lengths and larger magnetic fields correlating to increasing coupling between Fabry Perot and plasmonic modes. For our Au sample, optimal coupling was seen at wavelengths below 500 nm, where dramatic changes in lineshapes and magnetic field enhancement were observed.

This TE plasmon was made clearer in simulations by adding an air spacer between cavity and metal layer, while changing the thin metal film to a thick

film, which perfectly matches previous observations of TE plasmons in literature [15]. Coupling can be further controlled by selection of metal such as unconventional metals such as Cr, while popular plasmonic metals such as Ag seem to weakly support the surface plasmon.

The presence of a TE surface wave, matching the field profile of the TM plasmon is very clear and present in theoretical simulations of simpler structures, as seen in Figure 5.14 and 5.15, the introduction of PMMA and SiO₂ enable radiative losses that made the mode properly bound, but the main feature of the surface wave, the magnetic field along the direction of propagation is still present regardless of changes to the overall structure.

However, we more direct evidence is desirable. The least ambiguous method of identifying this TE wave as a surface plasmon is to identify near field focusing, a feature that has been experimentally observed by previous work in the group. Designing a near field transducer specifically for TE operation and observing the scattered field from this near field focusing would show that this TE surface wave behaves similarly to its TM counterpart. Further evidence may come from the comparison with the TE behavior in this work, and that of the actual TM plasmon mode, whose presence was identified in simulation.

The possibility of this TE plasmon being another form of surface wave, such as Tamn mode or Dyakonov should be considered as well. Typically, Tamn modes are associated with Bragg mirrors, Bragg mirrors are complex multilayered structures that are strongly reflective, and these modes appear in regions where the reflection is nearly perfect, as we see in our structure [100]. Plasmonic Tamn modes are proposed to exist at the interface between metals and

Bragg mirrors [101] with the arrangement of the multilayers is critical in observing them [102] and if these waves are treated as purely transverse waves, which makes them distinct from surface plasmons.

There is no analysis of the field inside the metal, while these modes are centered in the metal, as are ours, they appear to have radiative components, like ours. Furthermore, we note that dispersion relations of Tamn modes match the dispersion of FP modes and waveguide modes perfectly [33]. Direct comparison between Tamn modes and our modes is complicated, and possibly not suitable given this result, especially when comparing the complexity of a Bragg mirror to a simple thin film. In addition, there is little to no acknowledgment of magnetic behaviors in Bragg mirrors, despite them being made of thin films, which have been shown to support magnetic responses in our work and the works of others[17].

Dyankonv surface waves [103] exist between the interface of isotropic and anisotropic media/dispersion, which itself is indistinguishable from magnetic permeabilities. While it is possible, one could argue that Dyakonov waves are surface plasmons themselves. Making comparisons to our structure difficult.

Chapter 6

Conclusions and Future Work

6.1 Conclusions

In conclusion, this thesis explored the optical magnetic responses of thin film films in SiN and SiO₂ sample, highlighting how Fabry Perot modes and Brewster angle are the result of electric and magnetic responses. A practical demonstration of these magnetic responses was shown through the excitation of a TE surface plasmon, which requires a non-unity magnetic permeability to exist.

To expand multipole decomposition to thicker films and non-zero angles of incidence, we derived a novel expansion on the current multipole model, demonstrating excellent agreement between our multipole model, a more traditional S-parameter model and measurement for 500 nm of Si₃N₄ across the visible and near infrared.

The Brewster angle is demonstrated to arise from the alignment between the

electric dipole and direction of reflection, however our novel model highlights a previously unobserved condition required to observe the Brewster angle in thin films, the destructive interference between the other electric and magnetic multipoles that have not aligned with the angle of reflection, such a phenomena has previously only been observed in dielectric metamaterials.

We also demonstrated that the alignment between multipole moment and wavevector can occur for every multipole, not just the electric dipole. These new Brewster Angles have not been reported in literature as of writing this thesis, remaining "invisible" in the reflection spectrum of thin films, as no destructive interference between remaining electric and magnetic multipole occurs. However they are required to accurately model reflection, and can not be ignored.

Building upon this model, we calculate the effective permittivity induced by the electric multipoles, using an established effective medium model. This calculation shows that all electric multipoles, except for the electric dipole, are spatially dispersive.

Spatial dispersion in the electric permittivity has many potential applications, given that it is equivalent to a non-unity magnetic permeability at optical frequencies, a property only achievable in subwavelength metamaterials, without being limited to subwavelength structures. Our observation suggests it may be possible to replace these difficult to fabricate, and delicate structure, with more traditional thin films, which are easier to fabricate and more robust. However an important limitation is that our thin films can not isolate and control the response of individual multipoles, making metamaterials more desirable when such control is needed. We suggest that further work is needed to identify

regimes where the lack of control over magnetic responses is suitable for applications.

Spatial dispersion is then explored in a thicker film of 50 μm SiO_2 thin film, showing that spatial dispersion is an intrinsic feature of Fabry Perot modes.

To realize a practical application of this spatial dispersion in thin films, we deposited 1200 nm of PMMA on a 40 nm of Au on a SiO_2 substrate and designed a novel set up to reliably measure the reflection spectrum as a function of angle in the Kretschmann configuration, which we refer to as the "Dual Prism Kretschmann configuration".

We identify the presence of several s- and p-polarized absorption features. Using eigenmode analysis, we demonstrated the presence of a magnetic field enhancement along the direction of propagation, a tell tale feature of the transverse electric surface plasmon, which is associated with non-unity magnetic permeability, or in our case, spatial dispersion in the permittivity.

Optimal coupling between this TE surface wave and the Fabry Perot is explored by calculating the propagation length, a parameter extracted from eigenmode analysis. It is shown that optimal coupling occurs below 500 nm. Different metals demonstrate drastically different coupling strengths, as a traditionally popular plasmonic metal, Ag, does not seem to support the TE surface wave at any wavelength in the visible spectrum, while Cr supports a very strong coupling across the entire spectrum.

The identification of spatial dispersion and its practical application demonstrates a new approach to observing phenomena associated with magnetic permeability, in dramatically simpler structures than what is currently available. Non-unity magnetic permeability are associated with negative refraction, s-polarized GH shifts and TE surface waves, phenomena that are difficult to achieve without metamaterials at optical frequencies. We demonstrate that thin films are capable of demonstrating such phenomena with spatial dispersion, a feature that appears intrinsic to all thin films.

6.2 Future Work

6.2.1 Introducing Gain into PMMA through Quantum Dots and Laser Dyes

A common problem in plasmonics is the unusually large loss experienced by a propagating surface plasmon [104], as a result of the large field enhancement and large absorption coefficient of the metal supporting it.

One possible solution is the introducing of gain to compensate for this loss, typically in the form of a Surface Plasmon Amplification by Stimulated Emission of Radiation or SPASER [105]. We aim to introduce PbS Quantum Dots into the PMMA layer of the structure studied in Chapter 5 to introduce gain in the Fabry Perot mode that would couple to the TE surface plasmon.

In initial experiments involved spin coating the quantum dots onto a glass

substrate to form what is known as a Colloidal Quantum Dot (CQD) Thin Film, and fluorescence measurements of these films are presented in Figure 6.1. While several artifacts are present in the measurements, there is a clear fluorescence peak observed from the CQD films.

Upon introducing the QDs into the PMMA, and spin coating onto of 40 nm of Au, we prototyped an in house measurement to detect fluorescence from our TE modes, as seen in Figure 6.2. The set up is the same as that used in Chapter 5, with an additional component, a fiber cable placed in the vicinity of the air-PMMA surface. Given that the incident light strikes the surface at angles above the critical angle, this approach will only detect the scattered light from the sample as the Fresnel equations will forbid transmission [106].

In Figure 6.2 (b)-(c), we demonstrate that this approach is capable of detecting the scattered light when a TE mode is excited, however no fluorescence was seen at the wavelengths seen in Figure 6.1 (e). This failure has not been determined as of writing this thesis, but is hypothesized to be a failure in fabrication.

Fluorescence Measurements of CQD Films

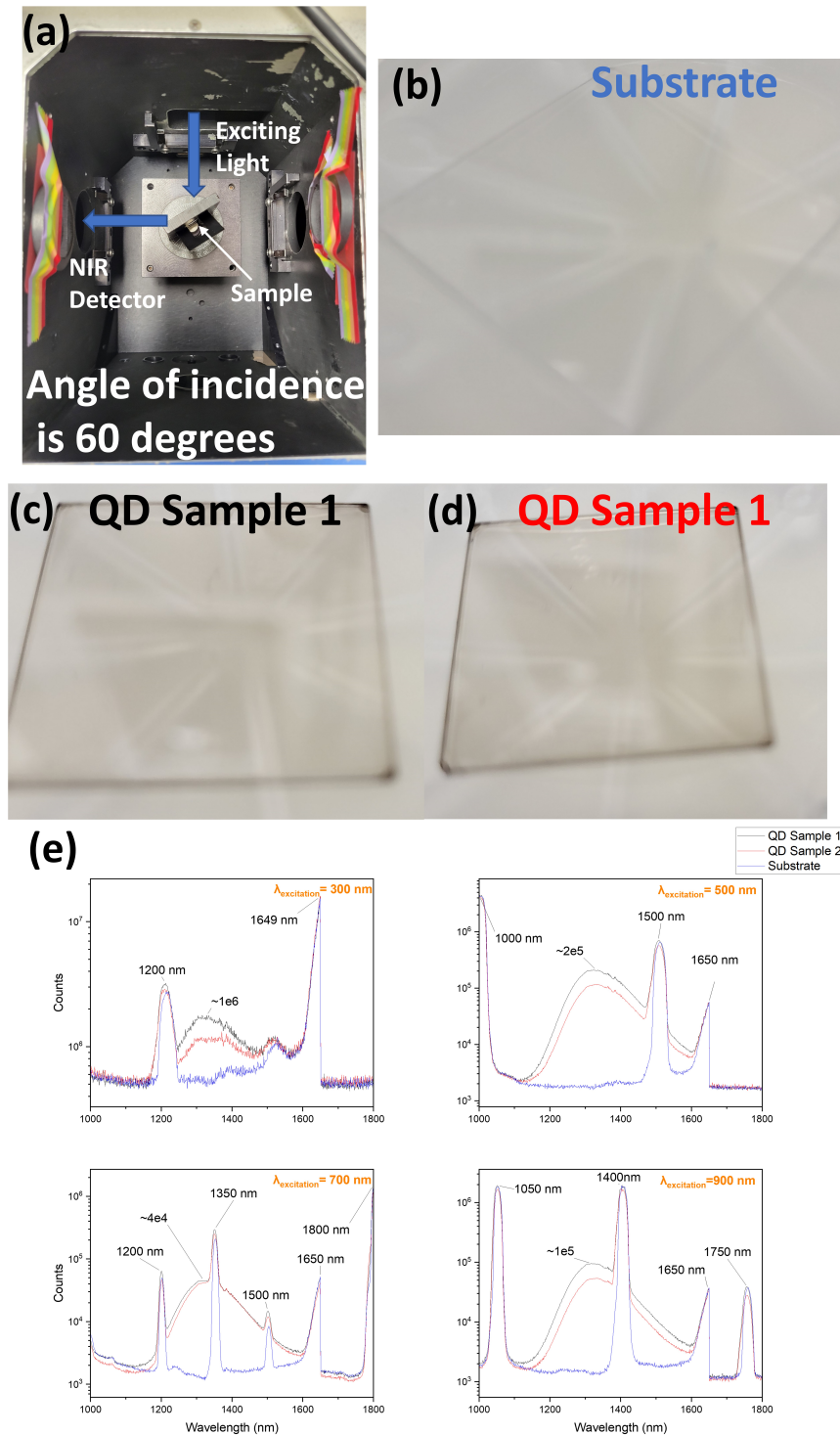


FIGURE 6.1: Outline of fluorescence measurements of colloidal quantum dot (CQD) thin films of PbS quantum dots. (a) Fluorolog set up Used. (b) Reference sample used for comparison. (c)-(d) Samples of CQD films deposited on glass substrates. (e) Measurements of fluorescence at pump wavelengths of 300, 500, 700 and 900 nm.

Fluorescence Measurement of Doped PMMA

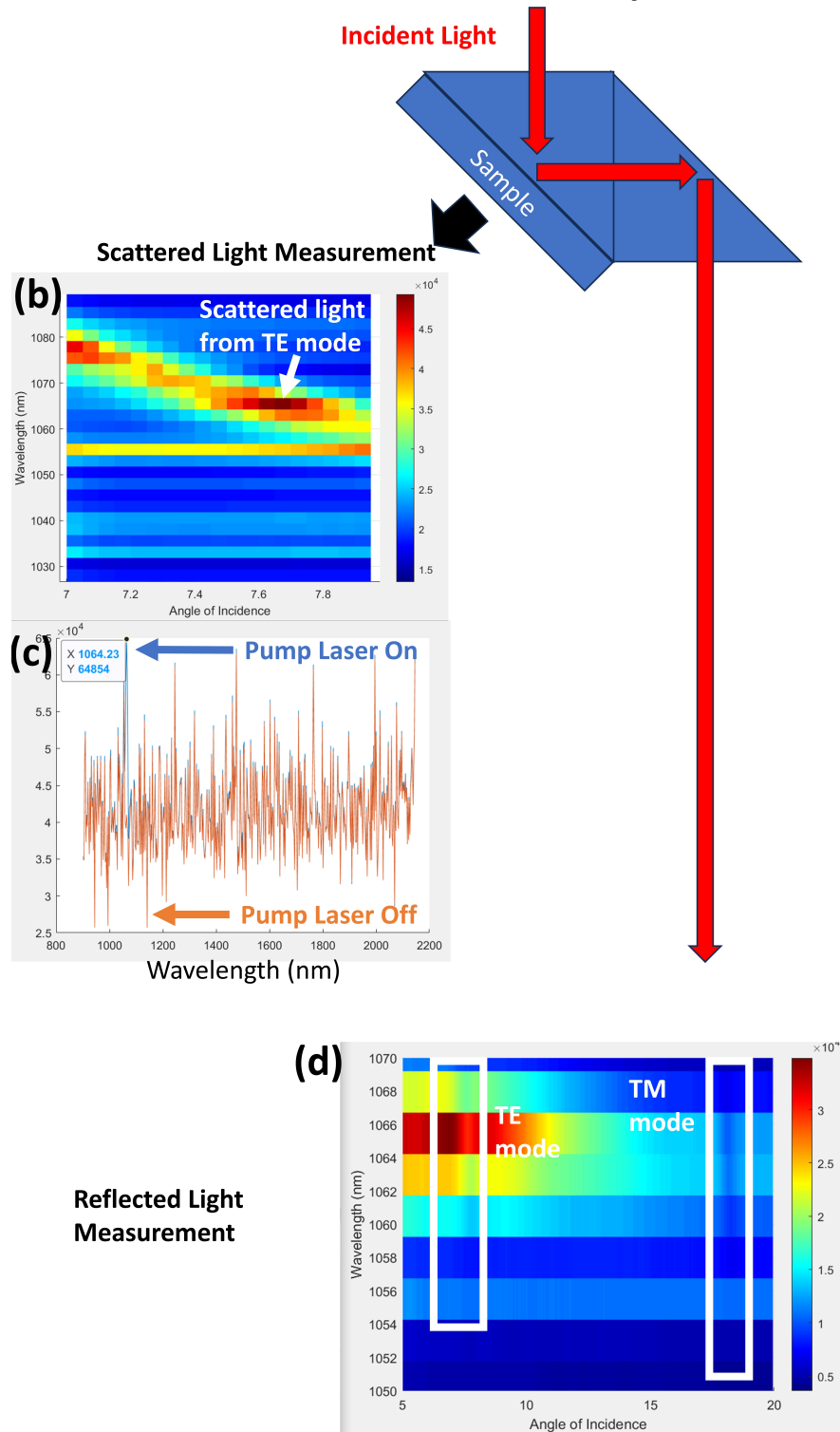


FIGURE 6.2: Outline of attempted fluorescence measurements of PMMA doped with PbS quantum dots, deposited on 40 nm of Au. (b) Measurement of scattered field from pump laser and scattered field from TE mode. (c) Comparison between measured scattered field with pump laser on and off, no fluorescence is seen. (d) Measurement of reflected light, similar to Figure 5.2.

6.2.2 Utilizing the Coupled Fabry Perot Plasmonic Mode as a Spectroscopic Source

Regardless of the failure of the work outlined in Figure 6.2, the design may serve practical application as a spectroscopic source, given that in Figure 6.2 (b) we show it is possible to convert a broad spectrum from a supercontinuum laser into a very narrow linewidth whose resonant position can be tuned with angle, and thickness of dielectric layer as shown in Figure 5.5. Most devices designed for spectroscopy rely on diffraction grating to convert broad sources to narrow ones, and may require several of these adding cost and bulk to the final design. Our design may outperform traditional approaches, with reduced cost and size.

Furthermore it may be possible to deposit molecules or nanoparticles on the surface of the PMMA, or embed them into the PMMA as we attempted to with the PbS quantum dots. It may be possible to then tune the incident wavelength by changing the angle of incidence, and measure the scattered field from those molecules/quantum dots. The larger electric field enhancements in the PMMA cavity, as well as the evanescent fields stretching into the air layer in Figure 5.11 and 5.12 may permit detection of these molecules/quantum dots. These molecules/quantum dots could then be characterized through the measurement of the scattered field, as in Figure 6.2 (b) and/or through changes in the measured linewidth in the reflection, and these measurements could be performed simultaneously.

For fluorescent molecules/quantum dots, the fluorescence may be used to excited another mode at longer wavelengths than the excitation wavelength if

embedded in the PMMA cavity, permitting for a laser design or further spectroscopic applications.

6.2.3 Optimizing Coupling Between Fabry Perot Mode and TE surface Plasmon

Building on results obtained in Figure 5.17, we observed that coupling between the TE surface plasmon and Fabry Perot mode is difficult. Simulations showed that optimal coupling in our Au sample occurs at wavelengths shorter than 500 nm, with a minimum in propagation length being observed. Similar simulations with Ag, a popular, low loss metal in plasmonics does not support this coupling at all, while Cr, which is not usually used for surface plasmon, is a highly optimal metal with good coupling across the visible range.

We suggest that the right selection of refractive indices and thicknesses of the metal and cavity may dramatically improve coupling, which could be optimized by looking for structures that minimize propagation length.

6.2.4 Observing Higher Order Brewster Angles

An interesting observation made in Chapter 2, was the potential identification of multiple Brewster angles associated with higher order electric and magnetic multipoles.

While the current definition of the Brewster angle may need amending, given that we have identified a previously unidentified property of the Brewster angle, the constructive interference of electric and magnetic multipoles. The current definition permits these higher order Brewster angles.

We hypothesis it may be possible to design a metamaterial to support these higher angle Brewster angles. The two possible approaches are.

- Isolating a single multipole that will align with the scattering vector to produce zero reflection
- 3 multipoles together may support a Brewster angle, one multipole will align with the scattering vector to produce zero reflection, while the remaining two multipole must be an electric and magnetic multipole that constructively interfere with each other.

The first approach is simpler and easier to achieve, while the second may only be feasible in thin films. To aid in the identification of these Brewster angles, we derive a series of equations to identify the angles that these Brewster angle may exist at.

For the electric dipole, we revisit our derivation for the angle dependent reflected fields below.

$$\vec{n} \times (\vec{p} \times \vec{n}) = \begin{bmatrix} \cos(\theta)(a_z \sin(\theta) - a_x \cos(\theta)) \\ a_y \\ \sin(\theta)(a_z \sin(\theta) - a_x \cos(\theta)) \end{bmatrix} \quad (6.1)$$

The Brewster angle will occur when either reflected field is zero for either polarization, i.e when, the first (p-polarization) or second element (s-polarization) are zero.

$$E_{p-pol} = p_z \sin(\theta) - p_x \cos(\theta) = 0 \quad (6.2)$$

$$E_{s-pol} = p_y = 0 \quad (6.3)$$

It should be immediately clear that a s-polarized Brewster angle arising from the electric dipole is impossible, given that equation 6.3 has no angle dependence, and would only be zero if there were no electric field in the cavity.

Equation 6.2 however may be zero and we proceed to derive the angle at which this would occur.

$$p_z \sin(\theta) - p_x \cos(\theta) = 0 \quad (6.4)$$

Re-arranging this we find the following

$$\frac{\sin(\theta)}{\cos(\theta)} = \tan(\theta) = \frac{p_x}{p_z} \quad (6.5)$$

We note that p_x and p_z can be described in terms of the angle of transmission into a thin film, these equations are listed below

$$p_x = \int_V E_i \sin(\theta_{\text{transmission}}) dV \quad (6.6)$$

$$p_z = \int_V E_i \cos(\theta_{\text{transmission}}) dV \quad (6.7)$$

Substituting these equation 6.6 and 6.7 into equation 6.5 we obtain a new equation in terms of the angles of reflection and transmission.

$$\frac{\sin(\theta_{\text{reflection}})}{\cos(\theta_{\text{reflection}})} = \frac{\cos(\theta_{\text{transmission}})}{\sin(\theta_{\text{transmission}})} \quad (6.8)$$

Simplifying equation 6.8 with the trigonometric identities, $\sin(a) \sin(b) = \frac{1}{2}(\cos(a - b) - \cos(a + b))$ and $\cos(a) \cos(b) = \frac{1}{2}(\cos(a - b) + \cos(a + b))$, gives us the following relation.

$$\cos(\theta_{\text{reflection}} + \theta_{\text{transmission}}) = 0 \quad (6.9)$$

or in simpler terms.

$$\theta_{\text{reflection}} + \theta_{\text{transmission}} = 90^\circ \quad (6.10)$$

If one notices that the angle of reflection is equivalent to the angle of incidence, and the angle of transmission is equivalent to the angle of refraction, then it is immediately clear that equation 6.10 is the well known condition required to derive the Brewster angle from Snell's Law, which we can now derive from equation 6.10.

$$n_1 \sin(\theta_{incidence}) = n_2 \sin(\theta_{transmission}) \quad (6.11)$$

Substituting in equation 6.10 into 6.11 gives us the well known Brewster angle equation as seen below.

$$\tan(\theta_{Brewster}) = \frac{n_2}{n_1} \quad (6.12)$$

And hence, we have derived the typical Brewster angle from our multipole decomposition model and have tied it's origin to the electric dipole. This method can be extended to any multipole discussed in this thesis by taking the equivalent vector presented in 6.1 and deriving the condition where the scattered field is zero. We show this in detail for the magnetic dipole next.

$$\vec{m} \times \vec{n} = \begin{bmatrix} m_y \cos(\theta) \\ m_z \sin(\theta) - m_x \cos(\theta) \\ -m_y \sin(\theta) \end{bmatrix} \quad (6.13)$$

$$E_{p-pol} = m_y \cos(\theta) = 0 \quad (6.14)$$

Now for p-polarized case, a magnetic dipole will never support a Brewster angle, as the reflected electric field will be zero when there is no field in the cavity or the angle of incidence is 90 degree, both of these conditions are unrealistic.

$$E_{s-pol} = m_z \sin(\theta) - m_x \cos(\theta) = 0 \quad (6.15)$$

However for a s-polarized Brewster angle is possible and we now derive this condition.

We find that the condition is similar to the electric dipole case, equation 6.5.

$$\frac{\sin(\theta)}{\cos(\theta)} = \tan(\theta) = \frac{m_x}{m_z} \quad (6.16)$$

However a problem arises, as of writing this thesis, an analytical description for the magnetic dipole moment, m , has not been derived, as the origin dependence of this terms can not be simplified. However there is another solution, we can describe m_x and m_z in terms of the total magnetic dipole moment, and the angle of that dipole.

$$m_x = |m_t| \sin(\theta_{MD}) \quad (6.17) \quad m_z = |m_t| \cos(\theta_{MD}) \quad (6.18)$$

Substituting these into equation 6.16 gives the following equation, which matches equation 6.8

$$\frac{\sin(\theta_{reflection})}{\cos(\theta_{reflection})} = \frac{\cos(\theta_{MD})}{\sin(\theta_{MD})} \quad (6.19)$$

This can be simplified the same way equation 6.8 was, giving the following condition for the magnetic dipole Brewster angle.

$$\theta_{reflection} + \theta_{MD} = 90^\circ \quad (6.20)$$

It appears that the condition to observe the magnetic dipole Brewster angle is not related to the angle of transmission, as the electric dipole Brewster angle is, but instead the angle of the magnetic dipole moment, which can be numerically calculated from the following equation.

$$\theta_{MD} = \tan\left(\frac{m_x}{m_z}\right)^{-1} \quad (6.21)$$

We note that in Chapter 4, we did not clearly observe a magnetic dipole Brewster angle in Figures 4.8 and 4.15, this Brewster angle may occur outside our range of studied angles or wavelengths. Next we briefly overview the electric quadrupole.

$$\vec{n} \times (\vec{n} \times (\hat{M}\vec{n})) = \begin{pmatrix} -\cos(\theta)(M_{11}\sin(\theta)^2 - M_{33}\cos(\theta)^2) \\ (M_{21}\sin(\theta) + M_{23}\cos(\theta)) \\ -\sin(\theta)(M_{11}\cos(\theta)^2 - M_{33}\sin(\theta)^2) \end{pmatrix} \quad (6.22)$$

$$E_{p-pol} = M_{11}\sin(\theta)^2 - M_{33}\cos(\theta)^2 = 0 \quad (6.23)$$

$$E_{s-pol} = M_{21}\sin(\theta) + M_{23}\cos(\theta) = 0 \quad (6.24)$$

Unlike the dipoles, it appears the electric quadrupole supports both s- and p-polarized Brewster angle, but the conditions may be polarization dependant. Applying the same methodology for the dipoles, we find the following conditions for these Brewster angles.

$$\tan(\theta_{EQ \text{ } p-pol \text{ Brewster}})^2 = \frac{M_{33}}{M_{11}} \quad (6.25)$$

$$\tan(\theta_{EQ \text{ } s-pol \text{ Brewster}}) = \frac{M_{23}}{M_{21}} \quad (6.26)$$

It is difficult to attribute an angle to a 2nd order tensor, and hence we were unable to derive simpler equations, but these equations can still be numerically calculated.

Following the same method, the magnetic quadrupole Brewster angle are as follows.

$$\tan(\theta_{MQ \text{ } p-pol \text{ Brewster}}) = \frac{M_{23}}{M_{21}} \quad (6.27)$$

$$\tan(\theta_{MQ \text{ } s-pol \text{ Brewster}})^2 = \frac{M_{33}}{M_{11}} \quad (6.28)$$

The electric and magnetic quadrupole Brewster angles were observed in our earlier studies, in Chapter 4 Figure 4.11, and may be confirmed through the applications of these equations.

Future work will involve verifying these equations, as it is possible to numerically calculate these Brewster angles and verify their existence through multipole decomposition. Then the octupole Brewster angles may be derived as well.

Appendix A

Reflection using Matching

Thicknesses and Refractive Indices

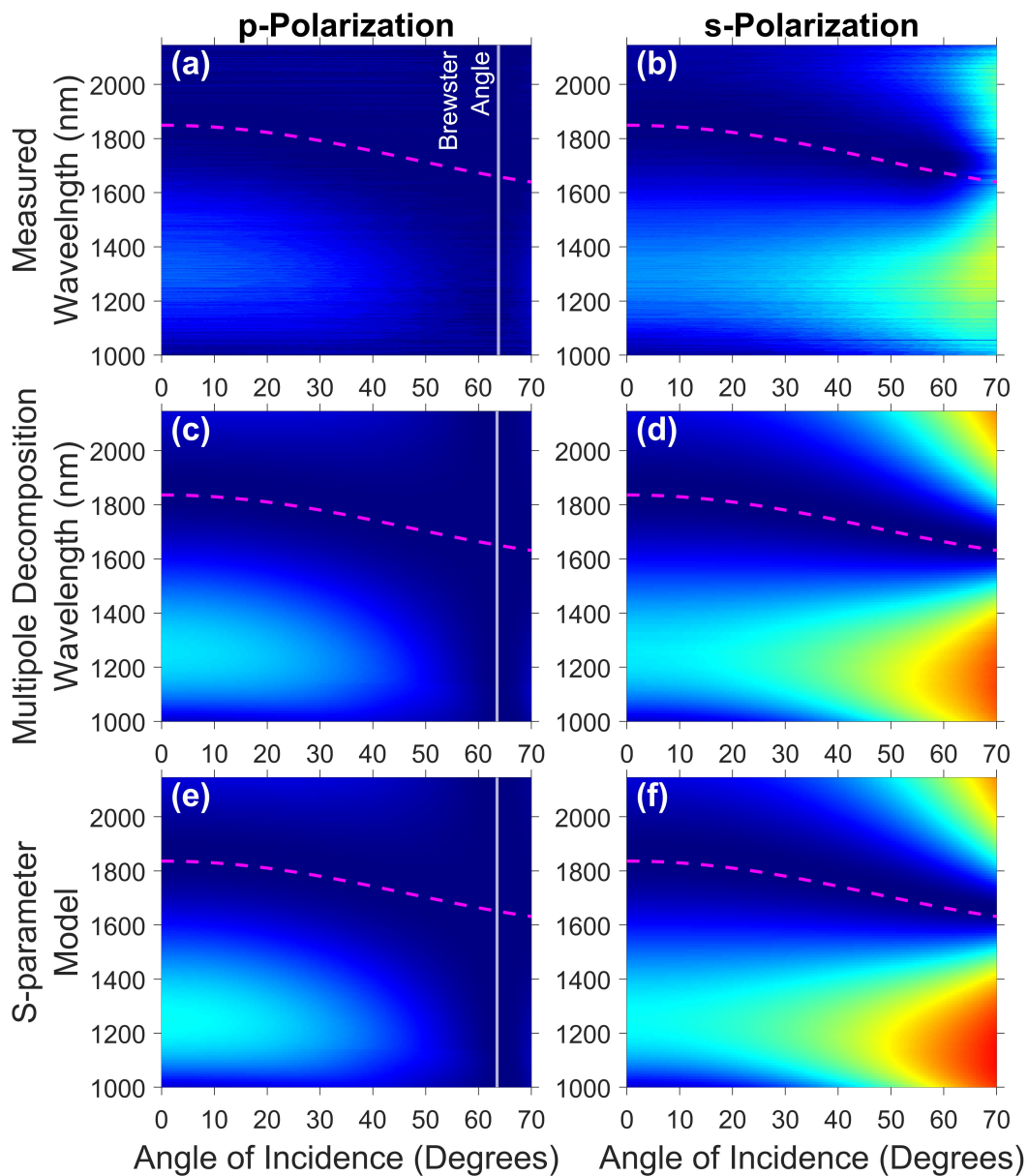


FIGURE A.1: NIR Reflection for measurement, multipole decomposition and S-parameter for s- and p-polarizations. Thickness is 455 nm, refractive indices used are from "Mesaurement 1" in Chapter 3.

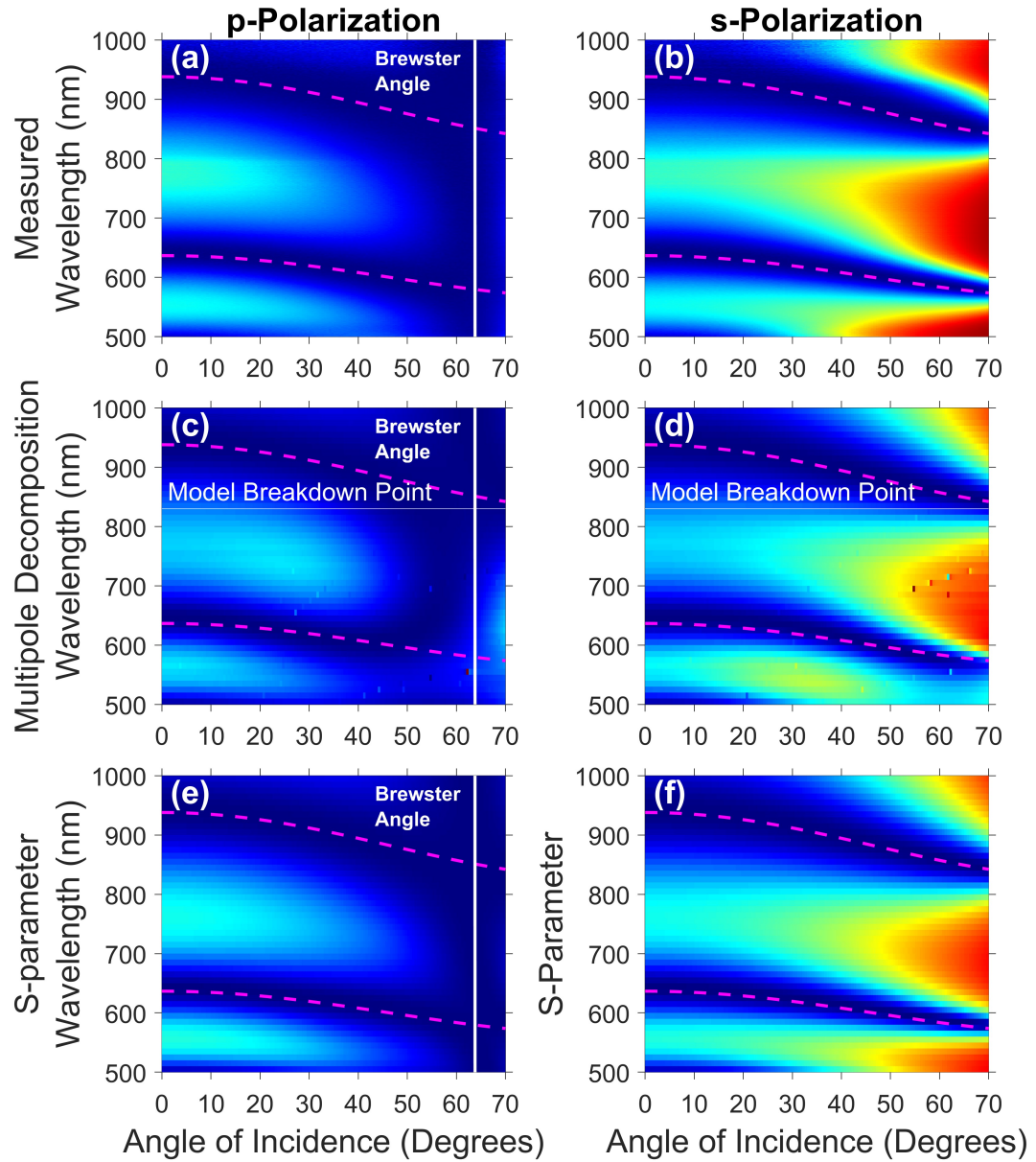


FIGURE A.2: VIS Reflection for measurement, multipole decomposition and S-parameter for s- and p-polarizations. Thickness is 455 nm, refractive indices used are from "Mesaurement 1" in Chapter 3.

FIGURE A.3

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