Light propagation in multilayer metamaterials
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Introduction

Light as a physical phenomenon is all around us. It is used to observe our surroundings, it is applied in long-range telecommunications, it forms the largest renewable energy resource available to us. In all applications, the interaction between light and matter is crucial. A transparent dielectric material such as glass is a typical example of a material that is used to engineer the phase fronts and propagation direction of light. This control over light has enabled us to fabricate a wide variety of optical elements such as lenses, optical fibers and prisms.

These optical elements rely on the fact that light propagates through the dielectric medium with a velocity lower than the speed of light in vacuum, as dictated by the material refractive index. At the smallest length scales, materials are built up from atoms. A wave of light interacts with an atom by coupling between the oscillating electromagnetic field of the wave and the atomic charge distribution. The strength of this coupling is indicated by the polarizability of the material. However, due to the large characteristic length scale difference between the atomic structure of the material and the wavelength of light, we usually treat a material as an effectively homogeneous slab described by a refractive index. Glass typically has a refractive index of around $n = 1.5$. A material with one of the lowest refractive indices known is magnesium fluoride, at $n = 1.37$. Semiconductors are typically characterised by a higher refractive index, such as $n = 4.5$ for silicon, but this high refractive index is often accompanied with strong absorption of light. One of the main reasons why glass is used in so many different optical elements is its extremely low absorption, allowing for instance propagation through glass optical fibers with an attenuation of only 0.2 dB/km.

Our ability to manipulate light was originally limited to naturally occurring materials. However, as light can be described as a propagating wave, it was soon
realized that by structuring materials on a length scale larger than the atomic (Angstrom) length scale, but equal to or below the wavelength of light, novel interactions between light and materials can be achieved. One of the first examples was the design of a new type of radio wave antenna, in which the antenna was built up from closely spaced metal plates [1]. Such structures, then called "artificial dielectrics", were used to create an effective medium which showed a refractive index smaller than unity [2].

These demonstrations were designed to operate for radio waves, with a typical wavelength in the centimeter or meter range. Because of this long wavelength, fabricating sub-wavelength features in the material was still relatively straightforward. The recent advances in nanofabrication, primarily in electron beam lithography and focused ion beam milling, have enabled a spectacular scaling down of the typical dimensions at which materials can be structured. These advances have been primarily driven by the semiconductor industry, in its efforts to fabricate smaller transistors and memory elements. Today, the minimum achievable feature size has reached the nanometer length scale. This has enabled the fabrication of structures with features which are sub-wavelength for visible light. Apart from improved top-down fabrication techniques, a large effort has been devoted to developing self-assembly driven, bottom-up fabrication techniques. One specific example of bottom-up fabrication is called DNA origami [3], where DNA molecules are folded in a controlled manner, enabling the fabrication of a structure with a tailored optical response [4].

Such artificially structured materials are called metamaterials. Here meta, from the Greek word for "beyond", implies that structuring a material on a scale below the wavelength of light can lead to new effective properties beyond those found in natural materials. One example of such an effective property is a negative refractive index. In the 1960's [5], the Russian scientist Veselago considered how light would propagate through a negative-index material. This negative-index material was supposed to have a negative permittivity and a negative permeability. He found that a slab of such a material would act as a flat lens, creating an image of an object without using curved surfaces, but taking advantage of negative refraction. In 1999, the British theoretical physicist John Pendry showed two possible practical designs how to fabricate metamaterials with a negative magnetic permeability, which opened a practical route towards the realization of negative-index metamaterials. One geometry was the split ring resonator design shown in Figure 1.1a. This original work [6] inspired a large research effort by many groups all over the world.

The first experimental demonstration of a negative-index metamaterial was in the GHz range [7]. Here, the dimensions of the unit cell, formed by a metallic wire lattice combined with split ring resonators, were chosen such that the effective permittivity and permeability of this metamaterial were negative in the same frequency regime, leading to a negative effective index. This was experimentally verified by measuring the refraction angle from a prism constructed from this metamaterial [8]. Since then, advances in fabrication techniques and design of metamaterials have led to the demonstration of negative-index metamaterials also in the
infrared [9][10], and the near-infrared [11].

(a) (b)

Figure 1.1: a, Sketch of a split ring resonator metamaterial operating in the near-infrared and at longer wavelengths. b, 3D Metal-dielectric coupled waveguide metamaterial operating in the UV and visible spectral range.

Obviously, fabrication of a three-dimensional metamaterial composed of resonant nanostructures becomes much more challenging as the wavelength of operation is reduced [12]. Furthermore, optical losses due to Ohmic dissipation strongly increase for shorter wavelength. As a result, practical metamaterials based on the split-ring resonator design in the UV/visible spectral range have not been realized. One solution is to limit the sub-wavelength structures to a single interface, as is done in the field of metasurfaces, which have demonstrated interesting phenomena such as anomalous refraction [13], wavefront shaping [14] and three-dimensional holography [15].

A new method to realize metamaterials at high frequencies uses a fundamentally different design, based on coupled plasmonic waveguides. A metal-dielectric interface supports guided waves, formed by longitudinal charge density oscillations in the metal, called surface plasmon polaritons (SPPs) [16]. Such SPPs are characterized by a specific propagation constant and lateral field profile. Away from the interface the fields decay exponentially. If two metal-dielectric interfaces are brought in close vicinity, the SPPs travelling over the two surfaces can couple, to form symmetric or anti-symmetric SPP waves. A dielectric core cladded by metal then acts as a waveguide [17]. Because part of the field of the waveguide mode overlaps with the metal regions, which are characterized by a negative permittivity (in a certain spectral range), completely new propagation characteristics can be achieved, such as propagation with a negative modal index [18]. Such a negative-index waveguide mode has been experimentally observed in a 2D planar geometry [19], and the dispersion of different coupled plasmonic waveguide geometries has been systematically studied theoretically [20].

The waveguide geometries considered above consisted of two-dimensional de-
signs, where light is guided along two coupled interfaces. To achieve a 3D metamaterial at first sight these 2D waveguides seem unsuited as building blocks. It was realized however, that by closely stacking multiple metal-dielectric-metal waveguides onto each other and by engineering the adjacent coupling, also propagation of light in the third dimension can be achieved [21], rendering the coupled waveguide geometry a three-dimensional metamaterial. In fact, it was shown that by appropriate design of the unit cell geometry, the propagation constant can be made negative for TM-polarization and for all angles of propagation in the metamaterial. Such a multilayer structure then acts as a 3D metamaterial with a negative effective index.

The multilayer design has several advantages. First of all, the fabrication of the geometry is relatively straightforward. The deposition of thin films is a process that can be routinely carried out in a cleanroom facility, using thermal evaporation, sputter coating or atomic layer deposition. Furthermore, because light is no longer coupled to a localized resonance but rather a waveguide mode that resides mainly in the dielectric film, losses can be significantly reduced [21, 22]. Finally, due to the metal-dielectric-metal waveguide mode dispersion, the geometry can be engineered to exhibit a negative-index mode at very high frequencies [20], operating in the blue or ultraviolet spectral range. Figure 1.1b shows a sketch of the multilayer design.

While the coupled waveguide design enables 3D isotropic properties, the same geometry also enables optical properties which are strongly anisotropic when different layer thicknesses are considered. Taking advantage of the structural asymmetry in metal-dielectric waveguides, such geometries have been used in the realization of hyperbolic metamaterials [23, 24]. Hyperbolic metamaterials are artificial materials where the permittivity is described by a tensor; the permittivity along the longitudinal directions (parallel to the waveguides) has an opposite sign to the permittivity in the lateral direction (normal to the waveguides). This response is realized in 3D metal dielectric waveguide arrays, where light propagating as a waveguide mode along the interfaces probes a different effective permittivity than light propagating normal to the interfaces in the form of a Bloch wave. Hyperbolic metamaterials have unique features, such as a very high local density of optical states [25] and can serve to enhance the radiative decay of localized emitters [26]. They enable the conversion of evanescent waves in air to propagating waves, a feature that has been exploited to create hyperlenses, which are able to resolve sub-diffraction limit features in the far-field [27, 28].

1.1 Wave vector diagrams

In the description of wave propagation through a multilayered medium, a comprehensible way of presenting the directions of propagation is the wave vector diagram [29]. In such a diagram, the allowed combinations of wave vectors are shown at one frequency. Therefore these diagrams are referred to as isofrequency contours (IFCs). For the 3D planar designs considered here, only two wavevector directions
must be considered, by symmetry. Figure 1.2a shows a wave vector diagram for homogeneous media, for which the dispersion relation is: \( n^2 k_0^2 = k_x^2 + k_z^2 \) in the two-dimensional plane considered here, with \( k_0 = 2\pi/\lambda_0 \), where \( \lambda_0 \) is the free-space wavelength. The IFC is calculated for \( \lambda_0 = 364 \text{ nm} \). The inner green circle refers to wave propagation in air, \( n = 1 \). The outer green circle corresponds to propagation through a \( n = 1.5 \) material, such as glass. The red and blue circles are IFCs for slightly red and blue shifted frequencies. The IFC is a circle as light propagates with the same wave vector in all directions in a homogeneous medium. Increasing (decreasing) the frequency leads to a larger (shorter) wave vector.

**Figure 1.2:** a. Wave vector diagram showing the isofrequency contours for two homogeneous media. The inner green circle refers to light propagation with a wavelength of \( \lambda_0 = 364 \text{ nm} \) (\( k_0 = 17.3 \text{ \mu m}^{-1} \)) through air. The outer green circle refers to propagation through a medium with \( n = 1.5 \). The vertical gray line indicates the parallel wave vector momentum \( k_x \) of a plane wave with an angle of incidence in air of 30 degrees. The red and blue circles correspond to a small modulation to lower frequencies (red) and higher frequencies (blue). Refraction at an interface between air and an \( n = 1.5 \) medium can be constructed using this diagram. b. The isofrequency contour corresponding to a homogeneous negative-index material. The red and blue circles are inverted when compared to (a), and the Poynting vector is now oriented inward. As the direction of the \( z \) component of the Poynting vector is preserved, the \( x \) component changes sign, corresponding to negative refraction. c. IFC for a hyperbolic metamaterial. This metamaterial supports very high wavevectors, and the refracted Poynting vector \( S_r \) exhibits negative refraction.

The refraction of a plane wave at an air/glass interface can easily be constructed from such a wavevector diagram. The incident plane wave, propagating in the positive \( z \) direction, is described by an incident wavevector \( k_i \) and Poynting vector \( S_i \), which are colinear in a lossless, homogeneous medium. At the interface, the parallel wavevector momentum \( k_x \) is conserved, as indicated by the vertical gray line in Fig. 1.2a. The intersection between this line and the outer circle determines the refracted wavevector in glass. Note that there are two intersections between the outer circle and the line of conserved \( k_x \), which correspond to upward and downward propagating waves. The direction of energy propagation will determine which solution is valid. The incident plane wave has a positive vertical component.
of $S_r$, meaning light propagates in the positive $z$ direction. The group velocity is given by $v_g = \partial \omega / \partial k$, and in the absence of losses, it can be shown that the group velocity is equal to the energy velocity $v_e = v_g$ [30]. The energy velocity is colinear with the Poynting vector, which represents the energy flux direction. Therefore, the orientation of the Poynting vector will be normal to the tangent of the wavevector diagram, oriented towards IFCs corresponding to higher frequencies. Based on this, we see that for the case of a normal dielectric interface in Fig. 1.2a the Poynting vector is oriented outwards, and light propagation in the dielectric is described by the top intersection between the line of conserved $k_x$ and the outer IFC, as there the vertical component of $S_r$ is positive.

Next, we use the wavevector diagram to consider light propagation through a metamaterial with an isotropic negative index ($n = -1$), such as the coupled waveguide geometry proposed in [21]. Figure 1.2b shows the IFC for this metamaterial. The radius for the IFC is the same for the metamaterial and vacuum. However, the IFCs for the higher (blue) and lower (red) frequencies are now inverted. This means that the Poynting vector is now oriented inwards. Following the same construction as in Fig. 1.2a, with the incident light again propagating in the positive $z$ direction, the correct solution for $k_r$ is now the bottom intersection between the IFC and the line of conserved $k_x$ (gray), as in that case $S_r$ is oriented in the positive $z$ direction. This means $k_r$ and $S_r$ are anti-parallel, as sketched in the figure. As a consequence, this structure exhibits negative refraction, since the $x$-component of $S$ changes sign over the interface.

Fig. 1.2c shows the wavevector diagram of a hyperbolic metamaterial (HMM). This IFC is clearly very different from the homogeneous structures considered in Figs. 1.2a,b. Similar to the homogeneous metamaterial in Fig. 1.2b, the hyperbolic geometry shows negative refraction of energy as indicated by the fact that the Poynting vector in the metamaterial $S_r$ points in the negative $x$-direction. However, the wave vector $k_r$ is not anti-parallel with $S_r$, in contrast to the case in Fig. 1.2b. Hyperbolic metamaterials allow wavevectors with a very large magnitude, that carry very fine spatial details, enabling the fabrication of hyperlenses, which resolve features which are sub-diffraction limit [27, 28]. Another remarkable feature of these HMMs is that at high wavevectors, all waves propagate in the same direction, normal to the hyperbole, as indicated by $S_r$.

1.2 Outline of this thesis

In this thesis, we study the design and experimental realization of 3D metal-dielectric multilayer metamaterials. The goal of this research is to achieve fundamental insight in wave propagation in these metamaterials, to fabricate them, to experimentally determine their effective properties, and to apply these novel properties to demonstrate a flat lens. The following chapters present the results of this research.

Chapter 2 describes a multilayer metamaterial composed of a stack of thin Ag
and SiN multilayers. We determine the effective permittivity of this metamaterial from interferometric transmission measurements. In this experiment, light propagates along the waveguides, probing the negative permittivity of the metal and the positive permittivity of the dielectric. By changing the filling fraction of Ag in the unit cell, we tune the effective permittivity. We use this tunability to create a metamaterial of which the permittivity, or epsilon, crosses zero at a desired wavelength in the visible. Epsilon-near-zero (ENZ) metamaterials have the unique feature that light propagates with no phase advance through the material. We investigate the range of unit cell sizes for which this metamaterial can be fabricated, and study the angular response of the ENZ metamaterial.

In Chapter 3 we study how light propagates through a periodic metal dielectric multilayer structure, focussing on the Bloch-wave solutions to the wave equation in the $z$ direction. We study the effective optical properties of a metal dielectric multilayer structure with an isotropic negative index for the fundamental Bloch harmonic. We find that most of the energy is carried by a higher order, positive index harmonic. The theoretical findings are corroborated by a metamaterial prism refraction experiment, where the prism is sculpted from a Ag/TiO$_2$ multilayer stack using focussed ion beam milling.

In Chapter 4 we demonstrate a flat lens, fabricated from a Ag/TiO$_2$ multilayer. Due to negative refraction of energy, light from an object is focussed by a flat slab of such a material. Rather than using the double-periodic structure that has first been proposed to fabricate a flat lens, we show that by appropriately designing a single-periodic metal dielectric multilayer structure, an angle-independent effective negative index response can be achieved. Confocal transmission microscopy and near-field scanning optical microscopy are used to experimentally characterize the image formed by the flat lens.

While the Chapters 2-4 focussed on the propagation of light inside the metamaterial, Chapter 5 addresses the coupling efficiency of light into a metal-dielectric waveguide array metamaterial. Due to the large mismatch between the spatial field distribution of the incident plane wave and the complex field profiles in the metamaterial, light incoupling is normally quite inefficient. We design a dielectric coupling metamaterial that transforms the incident plane wave to a symmetric waveguide mode with strong field gradients. By controlling the relative position of the coupling metamaterial with respect to the waveguide array and by optimizing the coupling metamaterial dimensions, we achieve perfect coupling to an antisymmetric waveguide mode, which would not be excited by a plane wave without the coupling metamaterial. We show that even up to very large angles of incidence, a large fraction of incident light can be efficiently coupled into the waveguide mode of interest by appropriately designing the coupling metamaterial.

Chapter 6 discusses potential applications of the multilayer metamaterial design described in this thesis. We propose a flat lens for the near-infrared telecom wavelength band, and discuss an electrically tunable flat lens. We show that a hyperbolic metamaterial combined with a special cavity design creates a wave-attracting cavity resonance at optical frequencies. We describe how for circularly
polarized light the plasmonic spin-Hall effect could be exploited to achieve a deterministic wave attractor for arbitrary light emission around the cavity. Finally, we show how a dielectric multilayer structure can act as an angular filter, and how this can be used to control the emission characteristics of light-emitting diodes.

Overall, this thesis describes the effective optical properties of metamaterials formed by multilayer structures. Propagation of light through very thin metallic and dielectric layers exhibits unexpected behaviour, which we study using analytical calculations and numerical simulations. Based on this, we fabricate samples using cleanroom techniques, and characterize the effective optical response using phase-sensitive transmission measurements, angular refraction experiments and by confocal and near field microscopy. The results of these measurements help us further understand the interaction between light and new metamaterials, designed to operate in the UV and visible spectral range.