UNIVERSITY OF SOUTHAMPTON

FACULTY OF ENGINEERING, SCIENCE AND MATHEMATICS

Optoelectronics Research Centre

Novel Phenomena in Planar and Layered, Photonic and Microwave Metamaterials

by

Alexander Sven Schwanecke

Thesis for the degree of Doctor of Philosophy

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To my family.

UNIVERSITY OF SOUTHAMPTON

ABSTRACT

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Doctor of Philosophy

NOVEL PHENOMENA IN PLANAR AND LAYERED, PHOTONIC AND MICROWAVE METAMATERIALS

by Alexander Sven Schwanecke

This work is related to the investigation and development of novel concepts to utilise planar nanostructures in optics, aided by complementary research in the microwave domain—in particular:

Strong polarisation conversion capabilities are shown for planar chiral nanoholes in the near-infrared part of the spectrum. The efficiency is found to depend strongly on the mutual orientation of the chiral hole and the polarisation of the incident light.

Polarisation-dependent nano-focusing is seen for these nanoholes. Light is concentrated to a spot size of 42 % of the wavelength in the proximity of a nanohole.

Broken enantiomeric symmetry is discovered for planar chiral structures excited with circularly polarised light, it is explained numerically and analytically for optics.

Non-paraxial contributions are analytically proven to be crucial to the description of the nonlocal response of planar chiral structures.

Asymmetric optical transmission in anisotropically nanostructured planar chiral metamaterials is demonstrated, for the first time. This phenomenon is mediated by a new type of excitation: an enantiomerically sensitive plasmon. The normal incidence transmission of circularly polarised light displays a difference exceeding 25 % for opposite propagation directions.

Giant gyrotropy in bilayered chiral structures is shown for both optical and microwave spectral regions, for the first time. The maximum relative polarisation rotatory power in the microwave domain is found to be 5 orders of magnitude stronger than in a gyrotropic crystal of quartz; for optical frequencies it exceeds 2500 °/mm.

The first demonstration of an optical magnetic mirror is reported. The phase change for the electric field of a wave reflected by a nanostructured metal surface is found to be smaller than 90 ° (the signature of magnetic mirror behaviour) for a broad spectral range from 550 to 750 nm, reaching values below 25 °.

A unit of planar chirality is proposed, for the first time. Normalisation procedures for the class of triple-integrated and -summated measures of planar chirality are introduced. A new type of multiplicative measure is defined and its properties and applicability are demonstrated.

A process for nanoimprint lithography at room temperature is developed. The production of planar chiral metamaterials for optical applications is validated to be reliable and reproducible and to provide a strong polarisation effect.

Contents

List of Figures

List of Tables

Declaration of Authorship

I, Alexander Sven Schwanecke, declare that the thesis *Novel Phenomena in Planar and Layered, Photonic and Microwave Metamaterials* and the work presented in it are my own. I confirm that:

- this work was done wholly or mainly while in candidature for a research degree at this University;
- where any part of this thesis has previously been submitted for a degree or any other qualification at this University or any other institution, this has been clearly stated;
- where I have consulted the published work of others, this is always clearly attributed;
- where I have quoted from the work of others, the source is always given. With the exception of such quotations, this thesis is entirely my own work;
- I have acknowledged all main sources of help;
- where the thesis is based on work done by myself jointly with others, I have made clear exactly what was done by others and what I have contributed myself;
- part of this work has been published as articles, letters or communications:

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Some content has, furthermore, been made public at the following conferences and in their respective proceedings (if applicable):

1. Y. Chen, F. M. Huang, A. S. Schwanecke , N. I. Zheludev, and E. Huq. Nanolithography for metallic quasi crystals for nanobio applications. *Micro and Nano Engineering 2008 (MNE2008)* (Athens, Greece; 15 - 18 September 2008).
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Signature ______________________   Date ____________________

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Southampton, UK Alexander Sven Schwanecke
May, 2009

Supplementary Statements

The articles embedded in this thesis are followed by brief statements clarifying the extent of my respective contributions. They have been altered from their respective original by conversion of all internal and external references, and footnotes for consistency throughout this thesis. Short figure captions have been introduced for the overall list of figures to facilitate an effortless overview.

Several parts of the discussed results involve the usage of template structures of low symmetry and complexity. Historically, such elementary shapes or their variations may carry religious, political or other meaning in certain social contexts. Their individual use in this work was motivated purely by scientific merit and carries no further meaning whatsoever.

The bibliography contains the document object identifier (DOI) for many articles. In order to retrieve these articles online append the identifier to the URL `http://dx.doi.org/`. Moreover, most references throughout this thesis are hyperlinked to their corresponding journal web pages via this method, clickable in the PDF version of the document.

Abbreviations

CCD charge-coupled device

CMOS complementary metal oxide semiconductor

DP diffraction pattern

FDTD finite-difference time-domain (method)

FEM finite-element method

FM flow or field map (electromagnetic)

HSQ hydrogen silsequioxane

IR infra-red

RCP left–circularly polarised / polarisation

RHS right-hand side

PCS planar chiral structure

PSTD pseudospectral time domain (method)

PMMA poly(methyl methacrylate)

RAM random access memory

RIE reactive ion etching

RTNIL room-temperature nanoimprint lithography

RCP right–circularly polarised / polarisation

SEM scanning electron microscope

TE transverse electric (mode / field)

TM transverse magnetic (mode / field)

Introduction

1. Introduction

Rapid advances in micro- and nano-fabrication over the last decade have opened previously inaccessible regions of photonics to experimental physicists. At the same time, ever increasing computational capabilities have made complex numerical analysis more readily available. This was the fertile ground that fostered the mounting interest in what have come to be known as **metamaterials**—artificially designed materials, carefully structured on the subwavelength scale to create novel kinds of light-matter-interaction, beyond (meta) the bare physical properties of their constituent elements and substances.

Control of phase-, as well as frequency-related properties is the key to most types of metamaterials. While considerable effort was initially focussed on double-negative-index materials [Pendry, 2003; Smith et al., 2000; Veselago, 1968] and many early predictions and results emerged from the long-established microwave research community, the field has since evolved to encompass a significantly broader perspective.

This thesis is motivated by earlier work on planar chiral structures in Southampton, initially starting with basic theoretical considerations [Svirko et al., 2001], which sparked systematic experimental investigations for diffracting arrays of planar chiral nanostructures [Papakostas et al., 2003] and revisited the idea of putting numbers not only behind the physical properties of such structures, but also their mathematical degree of symmetry, namely their chirality [Potts et al., 2004; Schwanecke, 2004]. At the same time, suggested links between the polarisation and resonant properties of planar chiral structures and a possible "chiral route to negative refraction" [Pendry, 2004] further strengthened the outlook of the project. These aside, the prospect of polarisation-oriented functionality on the subwavelength scale and with planar dimensions would already possess considerable merits in its own right.

The word **chiral** stems from the Greek word for hand which is perhaps the simplest example of a chiral object, as they exist in left and right versions. The definition of chirality used nowadays goes back to Lord Kelvin and the beginning of the last century [Kelvin, 1904]:

> I call any geometrical figure (. . .) chiral, and say it has chirality, if its

image in a plane mirror (. . .) cannot be brought to coincide with itself.[1]

Accordingly, two-dimensional objects are deemed planar chiral if they cannot be found congruent with their own mirror image, obtained against a line in its plane of existence, by any translation and/or rotation within this plane. Two objects that are mirror images of one another are called **enantiomers**.

The terminology around chirality has largely been coined by the chemistry community, where various applications of this concept exist primarily on molecular level. It stems from the discovery that objects consisting of the same elements but in a mirror image configuration can have significantly different chemical properties. An instructive introduction into the relations of chemistry and chirality has been provided by the Nobel laureate Vladimir Prelog [1976].

In biology on the other hand, the concept of chirality allows, for example, one to distinguish between bacteria that share a mirror-symmetric buildup but differ considerably in their properties: Lactic acid exists in two enantiomeric forms, denoted D(−) and L(+), where the latter can be broken down much better by common biological systems like the human body [Wikipedia, 2009].

However, all these terms have their roots in the concept of optical activity. In 1846 Faraday discovered the rotation of the polarisation plane of linearly polarised light when applying a magnetic field. Two years later Pasteur noted equal but opposite amounts of polarisation azimuth rotation by different crystals of sodium ammonium tartrate. Ultimately, Lord Kelvin understood the difference between magnetic rotation and natural optical activity and introduced the notion of chirality to describe the latter phenomenon [Barron, 2000].

For this thesis, understanding the optics of individual subwavelength-sized planar chiral template structures, their symmetries, and interactions became the first step in bringing chiral metamaterial functionality to optics, not least because polarisation microscopy had led to puzzling results [Schwanecke et al., 2003]. Chapter 2 provides a detailed numerical analysis of planar chiral nanoholes, which is further expanded in Chapter 3. In order to judge and predict the symmetries of such light-matter interactions and to achieve further clarity regarding the nonlocal nature of the optical responses, Chapter 4 provides analytical and numerical detail, and more broadly considers arrays of chiral elements and time-resolved simulations.

Understanding of the resonant and polarisation properties observed and predicted for planar chiral structures requires the differentiation of planar achiral effects (part III of this thesis) from purely planar chiral effects (part I), and from volume or conventional three-dimensional chirality and resulting gyrotropy (part II). This thesis inves-

[1] Quotation taken from Le Guennec [2000].

tigates phenomena resulting from each of these three categories:

Fedotov et al. [2006] found evidence for microwave radiation that the normal incidence transmission of circularly polarized waves through an anisotropic planar chiral metamaterial is asymmetric for opposing propagation directions. Chapter 5 numerically predicts, while Chapter 6 experimentally confirms that this effect is present in the visible to near-IR part of the spectrum for adequate topologies of nanostructured surfaces. It explains how this interaction is mediated by the excitation of enantiomerically sensitive plasmons.

Conventionally, optical activity and gyrotropy are associated with bulk media, with multi-wavelength dimensions in all three spacial directions [Svirko and Zheludev, 1999]. A single layer with a regular topology of planar chiral templates allows the creation of resonator cavities solely in its plane, the symmetry of which can only be disturbed by a substrate supporting the layer. Yet two such layers, located well within their respective near-fields of interacting electromagnetic waves, would couple these two resonators. Introducing a respective twist between the individual template structures, with otherwise identical layers, allows the system to display full 3D-gyrotropy in an arrangement of well below a wavelength's thickness. Chapters 7 and 8 show experimentally and numerically how a two-layer arrangement of planar chiral structures can lead to gigantic specific gyrotropy, relative to the structural thickness and compared to previously known gyrotropic materials and configurations.

Effectively, all described structures consist of resonators, which produce a phase change in transmitted light waves. For planar chiral structures this phase change is less trivial and more sensitive to the relative orientation of the light polarisation and the chiral object. This phase change can also be utilised in achiral configurations: Chapter 9 demonstrates how a nanostructed metallic surface effectively interacts with light as a magnetic mirror. This compares to conventional (or electric) mirrors, where the electric field at the interface assumes a minimum, producing a reflected wave with reversed electrical component. A magnetic mirror, however, reflects a wave with an inverted magnetic component resulting in strong electric field values close to the interface—beneficial, for example, for the detection of small substances and nanoparticles.

The mathematical property of (planar) chirality in context with its physical manifestation in optics—through its effect on the polarisation of light transmitted or reflected from such media—has required researchers to develop measures and indices of symmetry. Many are motivated by simple planar chiral molecules and include optimisation routines to arrive at a value [Buda and Mislow, 1992; Harris et al., 1999; Le Guennec, 2000; Petitjean, 2003, and references therein]. Complex planar geometries would thus be difficult to handle and analyse coherently. However, Osipov et al. [1998] introduced a new type of measure, which was later improved upon and further revised by Potts et al. [2004]. Here, a straightforward triple-integration (or summation, respectively) leads to a unique and reproducible number for any given system. Chapter 10 not only develops and analyses a new sub-type of such triple-integrated chirality measures, but introduces a general concept to normalise them. Moreover and for the first time, it explains how a unit of planar chirality can be introduced for any such measure, enabling consistent and well defined exchange of values and corresponding templates between researchers.

All previously suggested continuous media consisted of templates arranged in planar crystal-symmetry patterns, mostly square lattices. The transmission of light waves through apertures smaller than the wavelength requires consideration of the strongly diminished transmission, predicted by Bethe [Bethe, 1944], versus a collective strengthening for apertures in arrangements of crystal symmetry [Ebbesen et al., 1998]. Chapter 11 demonstrates for the microwave domain how even a quasicrystalline structure can not only transmit much more energy than predicted by Bethe for its individual holes, but also that it hardly affects the phase and intensity of a transmitted electromagnetic wave—effectively rendering a screen with subwavelength apertures "invisible" for a broad spectral domain.

Given the previously described results on planar nanostructures, any further development of the field will have to take into account not only state-of-the-art fabrication techniques for scientific purposes, but also the mass-production prospects of such intricate surface topologies. Chapter 12 explains a method to manufacture planar chiral metamaterials through nanoimprint lithography at room-temperature and thus considerably reduced cost. Moreover, it demonstrates the presence of basic polarisation properties in test samples, alluding to the opportunity for industrial exploitation.

Exploring the prospects of nanostructured planar chiral metamaterials, this thesis describes various aspects and novel phenomena of light-matter interaction for structured interfaces and surfaces. It arrives at its conclusions through numerical modelling, analytical and symmetry considerations, experimental analogies in the microwave domain, and extensive optical experiments for a variety of arrangements and, ultimately, it provides a unifying perspective and promising outlook for the employment of the developed concepts.

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Appendix

Future Work

The multitude of phenomena for different spectral domains described in this thesis provides ample room for concepts and investigations deriving from it. Four very promising candidates should be highlighted.

All employed materials described in this work have been aimed at linear optical interactions. In particular, the fish-scale designs exemplify how strong resonances can be combined with chiral or gyrotropic functionality. Preparing the patterned or substrate material with active and non-linear components would be a natural next step—not only would it provide for hybrid switchable devices, but may also ultimately strengthen the observed effects, aggregating to viable devices for the optoelectronics industry.

The investigated magnetic mirrors display a strong concentration of electric field intensity in the proximity of their interface. This should lead to enhanced interaction with particles in their proximity. Given the considerable remaining cost of such nanostructured surfaces, which could merely be alleviated by employing nanoimprint techniques, this might be a promising technique for enhanced trace gas detectors.

On the route to miniaturised optically functional elements, the use of circular polarisation may be found advantageous. The described effects of polarisation-dependent nano-focusing and broken enantiomeric symmetry would provide for selective guidance of light on the smallest scale. These theoretically predicted phenomena can readily be verified with polarising scanning near-field microscopy, before taking them further.

This work explains how intricately structured interfaces can assume effective electromagnetic parameters decidedly different from those of their constituent materials. Reaching for more complex chiral template designs and cleverly arranging and stacking them will ultimately lead to double negative index media, proving a different route to intriguing applications like the proposed cloaking devices.

Eventually, developing the investigated metamaterial phenomena into practical devices and evaluating them for different as well as more specific and narrow spectral ranges, like the fibre telecommunication bands, will provide a fertile ground for the advancement of this field of science.

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