Magnetoelectric resonant metamaterial scatterers

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Metamaterials are a class of optical media that have earned much attention from the scientific community at the end of the 20th and the beginning of the 21st century. The name is derived from the Greek word *meta*, meaning ‘beyond’, and refers to the ability of the material to exhibit properties that are not found in nature. Metamaterials aim to mimic naturally occurring homogeneous media composed of atoms or molecules, yet made of artificially structured building blocks in order to go beyond parameters accessible with just molecular interaction with light. Beating the diffraction limit and arbitrary bending of light with the help of transformation optics are the visions that have resulted in a plethora of man-made meta-molecules for nanophotonics.

1.1 Negative index materials

Research in the field of metamaterials is driven by the possibility to control the properties of light on the nanoscale by using coupled resonant nanoscat-terers to create optical materials with very unusual effective medium parameters. Engineering arbitrary values for the effective permittivity $\varepsilon$ and permeability $\mu$ would allow new forms of light control based on achieving negative index materials [1–5], or transformation optics media [6, 7] that arbitrarily reroute light through space. The possibility of achieving negative index of refraction has coined the name ‘negative index materials’ (NIMs) [8]. Depending on the choice of parameters, a positive or a negative sign can be taken in the definition of the refractive index of a material

$$n = \pm \sqrt{\varepsilon\mu},$$

where $\varepsilon$ and $\mu$ are the relative electric permittivity and magnetic permeability, respectively. The conventional choice is to choose the positive sign of the square
root, appropriate for positive $\epsilon$ and $\mu$ [2, 9]. All naturally occurring materials have $\mu=1$ at optical frequencies since they exhibit no, or very weak, response to the magnetic field of light. Furthermore, insulators usually have $\epsilon > 1$, while metals have $Re(\epsilon) < 1$ due to the free electron gas. Light incident at an oblique angle $\theta_1$ from a medium with $n_1$ upon a material with $n_2$ will be refracted under an angle $\theta_2$ according to Snell’s law

$$n_1 \sin \theta_1 = n_2 \sin \theta_2. \tag{1.2}$$

Veselago [1] first suggested that when a material possesses both negative $\epsilon$ and $\mu$, a negative square root must be taken as a definition of refractive index to avoid violating causality. It follows that $n < 0$ results in two fundamental anomalies: (1) negative refraction of light at oblique incidences, and (2) phase velocity opposite to energy flow. Choosing $n_2 = -1$ in Eq. (1.2) implies a negative $\theta_2$, i.e., light will be refracted on the same side of the normal as the incident wave. This is unlike conventional materials where light is refracted on the other side of the normal and it always bends away or towards it, but never through it, as shown in Fig. 1.1 (a). Since boundary conditions require $k||$ vector conservation, negative refraction means that the Poynting vector $S$ has a negative sign, i.e., the energy flows away from the interface while the phase fronts travel towards the interface [1, 10]. This is easy to see from the fact that

$$S = E \times H = \frac{1}{\mu} E \times B \propto -k. \tag{1.3}$$

A consequence of negative refractive index is strong focusing of light through a thin negative index slab that led to the idea of perfect lensing. In order to understand how perfect lenses work, let us first consider a positive thin lens. The distance $v$ of an object to the lens and the distance $b$ of its image to the lens are related to the focal length $f$ of the lens via

$$\frac{1}{v} + \frac{1}{b} = \frac{1}{f}. \tag{1.4}$$

The magnification $M$ of a thin lens is given by the ratio of the distances of the image to the lens and the object to the lens. Turning our attention back to the perfect lens, Fig. 1.1 (b) shows a ray tracing diagram from a source at a distance $d/2$ to the image (black arrows) that is made by a slab of material with $n = -1$ and thickness $d$. Due to the negative refractive index, the rays are focused inside the material creating a virtual image (white arrow). A real upright image with unit magnification is created at a distance $d/2$ from the material slab. The distances of the image and the object to the lens have to conserve the relation

$$b + v + d = 2d. \tag{1.5}$$
1.1. Negative index materials

Figure 1.1: (a) A schematic of light propagation upon refraction from a conventional material and a NIM. The wavevector points in the opposite direction to the energy flow in a NIM. (b) An object placed at a distance $d/2$ in front of a slab with $n = -1$ and thickness $d$ will be imaged with magnification $M = 1$ on the other side of the slab. Note that a virtual image is formed within the slab. (c) If an object is placed at a distance $v < d$, it will also be imaged with the same magnification.

As the object is brought closer to the lens in Fig. 1.1 (c), the magnification remains 1 and the total object to image distance remains constant. A real image is formed when $v < d$, while for $v > d$ no image is formed. Surprisingly, when compared to a standard positive lens, the magnification for a slab of $n = -1$ is always 1.

The unique property that initially steered attention to the field of metamaterials is not solely that a slab of $n = -1$ acts as a lens, but that it would in fact outperform any conventional lens. A usual lens is limited to a resolution no better than $\lambda/2$ due to the following argument. Suppose one has an object rich in spatial information that radiates towards the lens. Its field can be represented as

$$E(r, z, t) = \int E(k_{||}, z_{obj}) e^{ik_{||}r + ik_{\perp}(z-z_{obj}) - i\omega t} dk_{||}, \quad (1.6)$$

where $z$ is the distance away from the object at a distance $z_{obj}$, and towards the lens. This representation Fourier transforms the field to parallel wave vector space $k_{||}$. The sharpest features carry the highest $k_{||}$. However, due to the dispersion relation for vacuum $k_{||}^2 + k_{\perp}^2 = \omega^2/c^2$, any feature with $k_{||} > \omega/c$ is exponentially damped as it propagates towards the lens, since $k_{\perp}$ is imaginary. These are evanescent waves characterized by $\text{Re}(k_{\perp}) = 0$ and $\text{Im}(k_{\perp}) \neq 0$. Since fine details do not reach any far field lens, far field optics is limited to the diffraction limit of Abbe [2]. Resolving finer detail is hence the hallmark of near-field optics, where detection is attempted directly inside the evanescent field of the object. The unique insight of Pendry [2] that triggered interest in metamaterials, is that when an object is placed at a small, typically subwavelength distance of a thin negative index
Introduction

slab, the negative index slab has a transmission coefficient far exceeding one for the evanescent wave. In fact, the perfect lens effectively amplifies the evanescent field components to such a degree that on the other side of the slab, the transmitted field exactly reconstitutes the object in the image plane [2, 11, 12]. To understand how such amplification can occur, let us analyze refraction of a propagating wave again. For $n = -1$, causality implies that the phase fronts must travel opposite to energy. In other words, the proper understanding of the ray diagram in Fig. 1.1, is that energy refracts negatively, and by causality, points from left to right throughout the diagram. Because $k$ opposes energy flow, this means that the wave acquires negative phase as it propagates through the lens. All the waves recombine at the real image with net zero phase, because the negative phase acquired in the lens exactly cancels that acquired in the air. This particular argument carries over to evanescent incident waves, where the full complex phase is negated by the negative index medium, i.e., both the phase and the evanescent decay. It is important to realize that the only way for this reasoning not to imply violation of causality, is that the lens will only operate in a narrow frequency band and is intrinsically limited by unavoidable dispersion and absorption in the lens material [13–18]. The ability of a material to act as a perfect lens has important implications for applications where high resolution is needed, such as medical imaging, data storage, material fabrication where high resolution is needed, and as optical components [12, 19–23].

We have just seen how one might manipulate light if we choose $\epsilon = \mu = -1$. In case of arbitrary $\epsilon$ and $\mu$, however, light can be manipulated in even more complex manner described by transformation optics. Leonhardt [6] and Pendry et al. [7] suggested that light can be bent in space continuously by an inhomogeneous distribution of $\epsilon$ and $\mu$. Therefore, metamaterials offer the possibility to smoothly guide light around any object by realizing appropriate spatial variations in material parameters, i.e., both $\epsilon$ and $\mu$. This has led to the idea of cloaking devices. An object placed within a sphere with carefully engineered values of $\epsilon$ and $\mu$ will seem invisible to any outside observer however the object is illuminated, since the cloak guides any near and far field around the object [24]. Bending of light is also of immediate importance for waveguiding, optoelectronics and photovoltaics.

1.2 Origin of material parameters

Realizing arbitrary $\epsilon$ and $\mu$ is far from trivial and requires building blocks with a response to both $\mathbf{E}$ and $\mathbf{H}$. Metamaterial building blocks are excellent candidates for realizing negative $\epsilon$ and $\mu$ provided they each have a large electric and a magnetic response to light. In order to quantify the response of such scatterers, we first need to examine how material response arises in known materials. Conventional materials consist of very dense assemblies of atoms and molecules, as shown in
1.2. Origin of material parameters

![Figure 1.2: (a) A schematic of a simple cubic crystal consisting of atoms (grey circles). (b) A schematic of the same cubic crystal with split rings as atoms.](image)

Fig. 1.2 (a). Typical spacings between, e.g., Si atoms in a diamond crystal structure are 3 to 5 Å [25]. Therefore, the wavelength usually exceeds the atomic spacing by a factor 1000 or more. Although generally most materials are charge neutral, they are made of polarizable particles that contribute to material polarization $P$ and magnetization $M$. Rather than having to treat the scattering of each atom separately, the response of the medium can be captured in constitutive parameters $\epsilon$ and $\mu$ that enter Maxwell’s relations via

$$\nabla \times E = -\mu \frac{\partial B}{\partial t}, \quad \nabla \times H = \epsilon \frac{\partial D}{\partial t}, \quad (1.7)$$

where $E$ is the electric and $H$ is the magnetic field of light. An incident electric field $E$ induces an electric displacement in the material

$$D = \epsilon_0 \epsilon E = \epsilon_0 E + P, \quad (1.8)$$

where $\epsilon_0$ is the electric permittivity in vacuum. Likewise the magnetic response $\mu$ is due to material magnetization via

$$B = \mu_0 \mu H = \mu_0 H + M, \quad (1.9)$$

where $\mu_0$ is the magnetic permeability in vacuum.

Since most materials respond strongly only to the electric field of light $E$, in order to understand how material parameters arise, let us consider the motion of an electron bound to an atom in such a material driven by an incident electric field $E$. According to the classical Lorentz model [26], the excitation of an electron bound to an atom is analogous to driving a harmonic oscillator with mass $m$ on
a spring with a damping constant $\gamma$. The response of such an oscillator to the monochromatic driving $E_0 e^{-i\omega t}$ is described by the differential equation

$$\frac{d^2 x}{dt^2} + \gamma \frac{dx}{dt} + \omega_0^2 x = \frac{q}{m} E_0 e^{-i\omega t}. \quad (1.10)$$

The resulting electron displacement is $x(t) = \frac{q}{m} E_0 \frac{e^{-i\omega t}}{\omega_0^2 - \omega^2 - i\omega \gamma}$. Therefore, an atom acquires a dipole moment proportional to the charge separation $x$ in Eq. (1.10),

$$p(t) = \frac{q^2/m}{\omega_0^2 - \omega^2 - i\omega \gamma} V E e^{-i\omega t} = \alpha(\omega) E e^{-i\omega t}, \quad (1.11)$$

where the Lorentzian prefactor $\alpha(\omega)$ is the polarizability of the atom that describes the frequency dependent strength of the resonant response, $\omega$ is the driving frequency, $\omega_0$ is the frequency at which the system is resonant, and $\gamma$ is the damping of the material. For a material consisting of many atoms, the material polarization density $P$ is equal to the volume average of the dipole moments $p$.

Material polarization arises whenever the applied electric field distorts the electron distribution inducing a dipole moment $p$, whether in a medium consisting of, e.g., molecules, plasmonic nanoparticles or metamaterial building blocks, schematically drawn in Fig. 1.3. In all cases the important figure of merit is the polarizability $\alpha$ defined above through $p = \alpha E$. A well known example where polarizability is applied not to an atom or molecule, but to a bigger scatterer, is that of a small sphere of dielectric constant $\epsilon$, which in the long-wavelength limit has polarizability

$$\alpha = 3V \left( \frac{\epsilon - 1}{\epsilon + 2} \right). \quad (1.12)$$

The fact that the denominator yields a resonance in $\alpha$ for $\epsilon = -2$, is exploited in ‘plasmonics’. Indeed, 10-100 nm sized gold and silver particles have polarizabilities far in excess of their physical volume $V$ at frequencies where $\epsilon = -2$, which is associated with a collective response of the free electron plasma. Similarly, metamaterial building blocks are polarizable metal structures which contain magnetic polarizability in addition to an electric polarizability. Therefore, one can consider metamaterial scatterers, such as split ring resonators, as a sum of two objects with an electric dipole moment $p$ and a magnetic dipole moment $m$. Fig. 1.3 (b) shows how the incident electric field induces charge separation across a split ring gap, giving rise to the in plane electric dipole moment $p$. The current oscillating in the ring will give rise to the out of plane magnetic dipole moment $m$. Section 1.3 discusses this mechanism in detail.
1.3 Metamaterial building blocks: split ring resonator

The principle to realize metamaterials with unusual $\epsilon$ and $\mu$ is to assemble dense arrays of nano-scatterers with strong electric and magnetic polarizability that, when combined, render a strongly polarizable quasi-homogeneous medium. Therefore, it is important to remain at lattice spacings $< \lambda/2$, so that diffractive effects as in photonic crystals and gratings do not occur. We call this the effective medium limit, since the $E$ and $H$ only ‘see’ a homogeneous material. Realizing negative $\epsilon$ is relatively easy, since free electron metals by themselves supply negative $\epsilon$ at optical frequencies. However, they still maintain $\mu = 1$ at optical frequencies. In 1999, Pendry *et al.* [27] suggested that a response to the magnetic field $H$ could be evoked via resonances of metal rings. Since then, many efforts have been aimed at further strengthening the magnetic response of metamaterial scatterers by introducing double-gap or double split ring structures [28, 29]. In the previous section we have analyzed how an electric response arises in sub-wavelength scatterers via induced dipole moments. In order to understand how the magnetic response arises in metamaterials, we examine a case of an archetypical metamaterial building block, the split ring resonator (SRR).

A split ring resonator exhibits strong resonances in the visible and infra-red regime as an electric and magnetic response to both components of the electromagnetic field of light. The magnetoelastic response is electrostatically described as an $LC$ circuit with a parallel plate capacitance $C$ and inductance $L$ of an $N$-loop coil with $N = 1$. We can calculate the $LC$ frequency of a split ring by a simple electrostatic formula

$$\omega = \frac{1}{\sqrt{LC}}$$

(1.13)

where $L$ is the inductance and $C$ the capacitance of the Au ring. Assuming the
Introduction

‘coil’ has one loop, the inductance of a solenoid becomes

\[ L = \frac{\mu_0 A_{\text{loop}}}{t}, \]  

(1.14)

where \( A_{\text{loop}} \) is the area of the loop. The parallel-plate capacitance is given by

\[ C = \varepsilon \frac{w t}{d}, \]  

(1.15)

where \( w, d \) and \( t \) are the dimensions of the capacitor as noted in Fig. 2.2 (a). The incident magnetic field \( \mathbf{H} = H_0 e^{-i\omega t} \) can drive a current in the ring that sets up a voltage over the loop via Faraday’s law

\[ V = - \oint \frac{d\mathbf{H}}{dt} \mu_0 dA = i\omega \mu_0 H_0 e^{-i\omega t} A_{\text{loop}}. \]  

(1.16)

Vice versa is also true. Incident electric field can excite charge separation across the SRR gap, consequently inducing a current in the ring, as shown in Fig. 1.3 (b). The current induced in the ring depends on the impedance \( Z \) via Ohm’s law \( V = I \cdot Z \), with the impedance of an \( LC \) circuit given by

\[ Z = i\omega L + \frac{1}{i\omega C} + R, \]  

(1.17)

where \( R \) is the Ohmic resistance of the circuit. The current sets up a magnetic dipole moment \( m = IA_{\text{loop}} \) pointing out of the loop. Since evidently, \( \omega = 1/\sqrt{LC} \propto \text{size} \), scaling down split rings pushes the resonance to optical frequencies [30].

The first metamaterial made of cm-sized double-SRRs resonant structures was reported in [28, 31], where negative refraction was demonstrated in the microwave regime. The promise of NIMs in the visible part of the electromagnetic spectrum, i.e., to create transformation optical devices such as cloaks and near field lenses, has prompted state-of-the art fabrication of split rings with very small dimensions [31–35]. However, due to the material properties of the most commonly used metal, Au, size scaling ultimately reaches its limit at frequencies approaching the Au plasma frequency [34]. Though the choice of metal determines the \( LC \) frequency limit, resonance frequencies have remained above 750 nm [34].

Negative refractive index has furthermore been studied in literature in structures fabricated in metal-dielectric multilayers [36–39], such as rod-pairs [40] and coaxial waveguides [41]. Shalaev et al. [40] have reported on a NIM in the optical regime (1.5 \( \mu \)m) consisting of pairs of parallel Au nanorods. These cut-wire pair structures consist of two noble metal wires separated by a dielectric, as shown in Fig. 1.4 (a), where the light incident along the stacking axis induces an
1.4 Metamaterial building blocks as scatterers

Many researchers have recently aimed to achieve negative refractive index by stacking metamaterial building blocks in densely packed arrays. Due to the existence of both electric and magnetic dipoles, these scatterers are expected to couple strongly in arrays. Such coupling can at the same time result in unwanted complications when predicting and engineering $\epsilon$ and $\mu$, yet also can provide valuable insight in the relative strengths of the electric and magnetic dipole moments. The key question of this thesis is to identify what the polarizability that underlies the electric and magnetic response is, and how dipole-dipole coupling can be quantified. We wish to quantify how large the polarizability $\alpha$ of metamaterial building blocks is, and to identify if any magnetic character is evident in scattering. Experiments outside the domain of effective media have appeared only recently. These include experiments by Husnik et al. [46], and Banzer et al. [47] that quantify the extinction cross section of single split rings under differently polarized illumination, experiments where split ring resonators act as near field probes [48, 49], as well as a range of experiments on coupled systems. These experiments include extinction measurements on split ring dimers [50] that point at resonance hybridization, as...
Introduction

as well as reports of magnetization waves [51], structural and geometrical chirality in arrays, as evident in, e.g., massive circular dichroism [52–59], and chiral effects in split ring stereo-dimers studied by Liu et al. [60]. In many instances, metamaterial building blocks are in fact very strong scatterers with large cross sections [46, 61–63], comparable to the large cross sections of plasmonic structures. Therefore, metamaterial building blocks are excellently suited to construct magnetic antennas, array waveguides and gratings in which electric and magnetic dipoles couple and form cooperative excitations, in analogy to the functionality imparted by plasmon hybridization [64].

In order to introduce the measures that quantify how strongly nanoscatterers scatter, let us consider the simple textbook problem of a nanoparticle placed in a homogeneous electromagnetic field. The nanoparticle will either absorb part of the light, or it will cause light to be scattered and propagate in different directions. These phenomena both take power out of the incident beam, which is termed ‘extinction’. Energy conservation means that extinction equals absorption plus scattering

\[
\text{Extinction} = \text{Absorption} + \text{Scattering}. \tag{1.18}
\]

Let us now suppose the particle is so small, that it can be viewed as just a polarizable point dipole

\[
p = \alpha E_{\text{in}}. \tag{1.19}
\]

The amount of energy removed from the incident beam by the dipole is called the extinction. It is equal to the cycle-average work done per unit time by the incident field \(E_{\text{in}}\) to drive the dipole \(p\),

\[
W = \langle \langle \text{Re} E_{\text{in}} \cdot \text{Re} \frac{dp}{dt} \rangle \rangle. \tag{1.20}
\]

Assuming a harmonic time dependence in the driving \(E_{\text{in}}(t) = E_0 e^{-i\omega t}\) the time derivative of the dipole becomes \(\frac{dp}{dt} = -i\omega \alpha E_0 e^{-i\omega t}\), where \(E_0\) is the amplitude of the incident electric field and \(\omega\) is the driving frequency. Taking the real part of \(E_{\text{in}}\) and \(\frac{dp}{dt}\), the work done per cycle average becomes

\[
W = \langle \langle \text{Re} E_{\text{in}} \cdot \text{Re} \frac{dp}{dt} \rangle \rangle = \omega \text{Im}(E_0^* \alpha E_0). \tag{1.21}
\]

The loss of energy from the beam due to extinction normalized to the incident intensity is the extinction cross section \(\sigma_{\text{ext}} = W / I\) [65]. In case of a simple spherical electric dipole scatterer, polarizability \(\alpha\) is a scalar, and the extinction cross section (units of area) is given by

\[
\sigma_{\text{ext}} = 4\pi k \text{Im} \alpha. \tag{1.22}
\]
We will see in Chapter 3 how the extinction cross section depends on a tensorial and magnetoelectric $\alpha$. Once excited, the dipole re-radiates a certain field $E_{\text{out}}$ in a process called scattering. However, some of the radiation will be absorbed by the dipole, i.e., lost as heat in the scatterer, leading to a relation

$$\sigma_{\text{ext}} = \sigma_{\text{abs}} + \sigma_{\text{scatt}}, \quad (1.23)$$

where $\sigma_{\text{abs}}$ is absorption cross section and $\sigma_{\text{scatt}}$ is the scattering cross section, all in units of area ($m^2$). The scattering cross section quantifies how much power an induced dipole radiates. It is obtained by dividing the angle integrated radiated intensity of an oscillating dipole by the incident intensity, to get

$$\sigma_{\text{scatt}} = \frac{8\pi^2}{3} k^4 |\alpha|^2. \quad (1.24)$$

The ratio of scattering to extinction is called the albedo $a$ of the particle. It tells us how much light that was taken out of the beam the particle actually re-radiates. In case of a lossless scatterer, $\sigma_{\text{ext}} = \sigma_{\text{scatt}}$, the albedo equals one. This means that the scattering resonance loses energy only due to radiative losses [65]. Material absorption losses limit the albedo to $a \leq 1$. Eq. (1.22) and (1.23) are best known in the context of Rayleigh scattering for very small scatterers where $\alpha$ is simply given by Eq. (1.12). In this regime, the equations imply the well known $\lambda^{-4}$ increase of scattering strength with wavelength, and the fact that scattering scales as $V^2$, while absorption scales linearly with particle volume $V$. In other words, the smallest particles tend to just absorb, while bigger objects are strong scatterers and weak absorbers.

A fact that is not so broadly appreciated is that Eq. (1.22) and (1.23) hold equally for very strong dipole scatterers that have cross sections comparable to $\lambda^2$, though in such cases the polarizability as in Eq. (1.12) must be modified to include radiative damping. We will generalize this strongly scattering dipolar description common for plasmon scatterers [66–69] to metamaterials in Chapter 3.

### 1.5 This thesis

The aim of this thesis is to characterize the magnetoelectric response of metamaterial building blocks, specifically split rings, and to quantify scattering and extinction of single building blocks as well as coupled systems. At the start of this work, in 2008, several groups had just recently reported on achieving negative $\mu$ using split rings [31, 33, 70]. Our quest does not revolve around engineering $\mu$, but concentrates on identifying what the underlying polarizability $\alpha$ is. To this end, we have fabricated periodic arrays of split ring resonators with sub-wavelength lattice
Introduction

spacing and different dimensions along \( x \) and \( y \) directions in Cartesian coordinate space. We present experimental proof of strong electric and magnetic dipole-dipole coupling of SRRs in such arrays in Chapter 2. Chapter 3 is a theoretical study of the polarizability of individual SRRs. We have developed an electrodynamic scattering model that describes the magnetoelectric response of SRRs and predicts their extinction cross sections and radiation patterns. The theory shows strong support for our experimental findings from Chapter 2 on coupling in arrays as evidenced by direct comparison in Chapter 4. Furthermore, optical activity due to the magnetoelectric coupling in the polarizability of geometrically achiral scatterers, as previously reported for molecules, is explored in Chapter 5. In Chapter 6 we describe a custom built Fourier microscope set up to image radiation patterns of single photonic structures. This set up can be used to probe radiation patterns of individual and coupled plasmonic as well as metamaterial scatterers, a possibility that is outlined in Chapter 7. Here we propose further experiments for quantifying the magnetic response, coupling and optical activity of SRRs, as well as potential applications.
Bibliography