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Chapter 9

Thin metal-dielectric nanocomposites with a negative index of refraction

by

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§ 1. Introduction

The race in engineering metamaterials comprising a negative refractive index in the optical range has been fueled by the realization of negative index materials for GHz frequencies 6 years ago. Sheer miniaturization of the GHz resonant structures is one approach. Alternative designs make use of localized plasmon-resonant metal nanoparticles or nanoholes in metal films. Following this approach, a negative refractive index has been realized in the optical range very recently. These results are reviewed and summarized in this chapter. The chapter addresses the critical question on how to unambiguously retrieve the effective refractive index of a given thin nanostructured layer from data accessible to measurements and reveals difficulties in cascading individual layers in a bulk material. The major focus is made on overcoming absorptive losses. Numerical simulations show that a composite material comprising silver strips and a gain providing material can have a negative refractive index and 100% transmission, simultaneously. The chapter has a special focus on simple and intuitive ways of designing negative index metamaterials (NIMs) where efficient magnetic resonators, which are used to provide the required negative permeability, are combined with bulk or semicontinuous metal elements to provide the necessary permittivity.

1.1. The index of refraction

The refractive index is the most fundamental parameter to describe the interaction of electromagnetic radiation with matter. It is a complex number $n = n' + in''$ ($i = \sqrt{-1}$) where n' has generally been considered to be positive. While the condition $n' < 0$ does not violate any fundamental physical law, materials with negative index have some unusual and counter-intuitive properties. For example, light, which is refracted at an interface between a positive and a negative index material, is bent in the "wrong" way with respect to the normal, group and phase velocities are antiparallel, wave and Poynting vectors are antiparallel, and the vectors $\vec{E}$, $\vec{H}$, and $\vec{k}$ form a left-handed system. Because of these properties, such

materials are synonymously called "left handed" or negative-index materials. Theoretical work on negative-phase velocity dates back to Lamb (1904, in hydrodynamics) or Schuster (1904, in optics) and was considered in more detail by Mandel'shtam (1945) and Veselago (1968). A historical survey referring to these and other early works can be found in Holloway et al. (2003). Important discussions on terminology and later works is done by Caloz and Itoh (Caloz and Itoh, 2006).

In general, left-handed materials do not exist naturally, with some rare exceptions like bismuth that being placed in a waveguide shows $n' < 0$ at a wavelength of $\lambda \approx 60 \mu\text{m}$ (Podolskiy et al., 2005a). However, no naturally existing negative index material is known so far in the optical range of frequencies. Therefore, it is necessary to turn to man-made, artificial materials, which are composed in such a way that the averaged (effective) refractive index is less than zero, $n'_{\text{eff}} < 0$. Such a material can be obtained using photonic crystals (PC) (Kosaka et al., 1998; Gralak et al., 2000; Notomi, 2000; Luo et al., 2002; Berrier et al., 2004). However in this case, the interior structure of the material is not subwavelength. Consequently, PCs do not show the full range of possible benefits of left-handed materials. For example, super-resolution, which has been predicted by Pendry (2000), is not achievable with photonic band gap materials because their periodicity is in the range of λ . A thin slab of a photonic crystal only restores small k -vector evanescent field components because the material can be considered as an effective medium only for long wavelengths, and large k -vector components are not restored (Luo et al., 2003; Smith et al., 2003; Lu et al., 2005). A truly effective refractive index $n'_{\text{eff}} < 0$ can be achieved in metamaterials with structural dimensions far below the wavelength. Metamaterials for optical wavelengths must therefore be nano-crafted.

A possible – but not the only – approach to achieve a negative refractive index is to design a material where the (isotropic) permittivity $\epsilon' = \epsilon' + i\epsilon''$ and the (isotropic) permeability $\mu = \mu' + i\mu''$ obey the equation

$$\epsilon''|\mu| + \mu'|\epsilon| < 0, \quad (1.1)$$

This leads to a negative real part of the refractive index $n = \sqrt{\epsilon\mu}$ (Depine and Lakhtakia, 2004). Equation (1.1) is satisfied, if $\epsilon' < 0$ and $\mu' < 0$. However, we note that this is not a necessary condition. There may be magnetically active media (i.e., $\mu \neq 1$) with a positive real part μ' for which eq. (1.1) is fulfilled and therefore show a negative n' .

To date, we only considered isotropic media where ϵ and μ are complex scalar numbers. It has been shown that in a waveguide with an

anisotropic media, where ϵ and μ are tensors, a negative refractive index is feasible even if the material shows no magnetic response ($\mu = 1$). If, for example, $\epsilon_L < 0$ and $\epsilon_{\parallel} > 0$, then $n' < 0$ can be achieved (Podolskiy and Narimanov, 2005; Podolskiy et al., 2005a). Despite the fact that using anisotropic waveguide is a very promising approach, we will not focus on that topic here. This is mainly because so far a negative index for optical frequencies has only been achieved following the approach of magnetically active media.

This chapter is organized as follows: first, we recall how to achieve magnetic activity for GHz frequencies using metallic split-ring resonators (SRR) and how the SRRs have been successively scaled down to shift the magnetic resonance up to THz frequencies. When the optical range is approached, the finite skin depth of metals as well as localized plasmonic resonances must be considered in addition to the simple geometric scaling of metallic structures. Then, we discuss approaches to new design outlines, which are making active use of localized plasmonic effects. Metamaterials containing metal nanostructures as magnetically active components usually show low transmission due to reflection and absorption. We demonstrate an impedance-matched design to suppress reflection. To compensate for losses and obtain a fully transparent layer of NIM we add a gain material. A special focus is made on simple semi-analytical approaches to the simulation of optical NIMs. Finally, intuitive ways of designing NIMs are shown, where efficient magnetic resonators, which are used to provide the required negative permeability, are combined with bulk or semicontinuous metal elements to provide the necessary permittivity.

1.2. Downscaling split ring resonators

The first recipe of how to design a magnetically active material was suggested by Pendry in 1999 (Pendry et al., 1999). Two concentric split rings that face in opposite directions and are of subwavelength dimensions were predicted to give rise to $\mu' < 0$ (fig. 1(a)). One can regard this as an electronic circuit consisting of inductive and capacitive elements. The rings form the inductances and the two slits as well as the gap between the two rings can be considered as capacitors. A magnetic field, which is oriented perpendicular to the plane of drawing, induces an opposing magnetic field in the loop due to Lenz's law. This leads to a diamagnetic response and hence to a negative real part of the permeability. The capacitors (the two slits and the gap between the rings) are necessary to

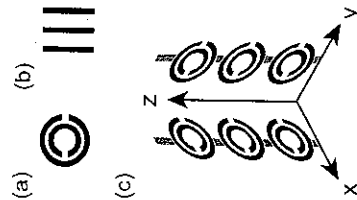


Fig. 1. (a) Magnetically resonant ($\mu' < 0$) metal structure: two counter-facing split rings of subwavelength dimensions (split ring resonator, SRR). (b) Electrically resonant ($\epsilon' < 0$) metallic structure: metal rods. (c) A combination of both structures results in a negative index metamaterial ($n' < 0$).

assure that the wavelength of the resonance is larger than the dimensions of the SRR.

Soon after this theoretical prediction, Schultz and coworkers combined the SRRs with a material that shows negative electric response in the 10 GHz range and consists of metallic wires in order to reduce the charge-carrier density; and hence shift the plasmonic response from optical frequencies down to GHz frequencies (fig. 1(b)) (Pendry et al., 1996). The outcome was the first-ever metamaterial with simultaneously negative real parts of the permeability and permittivity (Smith et al., 2000) and consequently a negative refractive index of approximately 10 GHz (fig. 1(c)) (Shelby et al., 2001a, b). From then onwards the race to push left-handedness to higher frequencies was open. The GHz resonant SRRs had a diameter of several millimeters, but size reduction led to a higher frequency response. The resonance frequency was pushed up to 1 THz using this scaling technique (Yen et al., 2004; Moser et al., 2005).

1.3. Metamaterials using localized plasmonic resonances

1.3.1. Metal nanorods

It was mentioned by Lagarkov and Sarychev (1996) that a pair of noble metal nanorods can show a large paramagnetic response, and it was first pointed out by Podolskiy et al. (2002) that such a pair of noble metal nanorods is also capable of a *diamagnetic response* at 1500 nm. In the

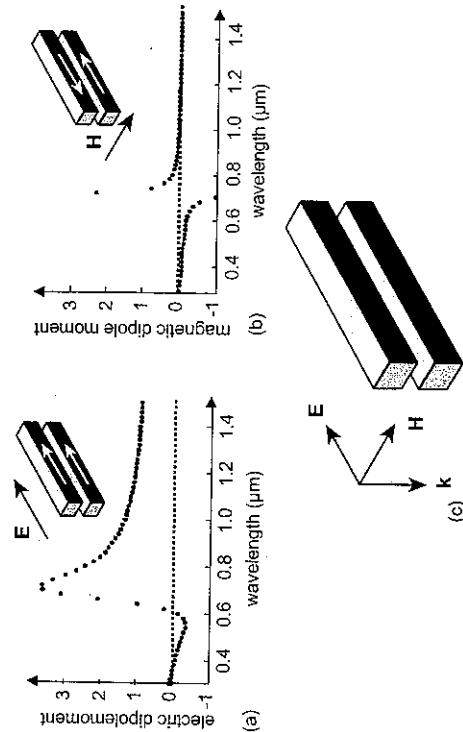


Fig. 2. Response of a pair of gold nanorods to radiation, simulated with coupled dipole approximation technique. (a) Electrical dipole moment: electric field oriented parallel to the axis of the rods. (b) Magnetic dipole moment: magnetic field oriented perpendicular to the plane of the rods. (c) A pair of rods illuminated from above with TM polarization. The pair of rods will have a double negative response to the field.

publication by Podolskiy et al. (2002), it was predicted for the first time that materials containing such pairs of rods can show a negative n' even for visible wavelengths. This issue has been discussed in more detail by Panina et al. (2002) and also by Podolskiy et al. (2003, 2005b). It is illustrated in fig. 2 how a pair of nanorods can show a negative response to an electromagnetic plane wave. Two gold rods are separated by a distance far less than the wavelength. The diameter of the cross-section of rods is also much less than the wavelength and length of the rods, but does not need to be in the range of half the wavelength. An alternating electric field parallel to both rods will induce parallel currents in both rods, which are in phase or out of phase with the original electric field, depending on whether the wavelength of the electric field is longer or shorter than the wavelength of the dipolar eigenresonance of the electro-dynamically coupled rods. Figure 2(a) shows the induced electric dipole moment in case of the following specific dimensions as has been reported in Podolskiy et al. (2005b): a rod length of 162 nm, a diameter of 32 nm (assuming cylindrically shaped rods), and a distance of 80 nm.

Let us now consider the magnetic field, which shall be oriented perpendicular to the plane of the rods. This magnetic field will cause antiparallel currents in the two rods as shown in fig. 2(b). This can be

considered as a dipolar magnetic mode. The magnetic response will be dia- or paramagnetic depending on whether the wavelength of the incoming magnetic field is shorter or longer than the dipolar magnetic eigenfrequency of the electro-dynamically coupled rods (fig. 2(b), after Podolskiy et al., 2005b). In this description in terms of coupled plasmonic resonances the magnetic dipole resonance appears at the same wavelength as the electric quadrupole resonance. However, the latter does not contribute to the electromagnetic radiation in the direction given in fig. 2(c) (Podolskiy et al., 2003).

So far, the electromagnetic response has been discussed in terms of coupled plasmonic resonances. An alternative way of looking at it is that the antiparallel currents in the rods and the displacement currents at the ends of the two rods form a current loop or an inductance, while the gaps at the ends form two capacitors. The result is a resonant LC-circuit (Lagarkov and Sarychev, 1996; Engheta et al., 2005).

It is important that both resonances, the dipolar electric and the dipolar magnetic, are at similar wavelengths. This requires that the coupling between the two rods should not be too strong, otherwise the two resonances split further apart. It is seen in figs. 2(a) and 2(b) that there is a certain range of wavelengths (between 500 and 600 nm) where both, the induced electric and the induced magnetic dipole moments are opposing the incident fields. Hence, an electromagnetic plane wave impinging from above and with E and H oriented as shown in fig. 2(c) (TM polarization) will induce a double negative response.

To the best of our knowledge, the unambiguous measurement of a negative refractive index in the optical range (specifically, at the optical telecom wavelength of 1500 nm) was reported for the first time by Shalaev et al. (2005a, b). The metamaterial in which the negative refractive index was achieved is outlined in fig. 3. Pairs of nanorods were fabricated on a glass substrate using electron beam lithography. The actual structure of the gold nanorod doublets is shown in fig. 3(a). The nanorods are 50 nm thick, stacked on top of the glass substrate, and a 50 nm thick SiO_2 layer is used as a spacer. The upper rod is smaller in dimension than the lower. A scanning electron microscope (SEM) picture of a single pair and its dimension are shown in fig. 3(a). Pairs of nanorods are periodically repeated as depicted in fig. 3(b) and shown by a SEM micrograph in fig. 3(c). Figure 3(d) shows a unit cell of the periodic arrangement and gives more dimensions. A full description of the sample and its preparation is given in Shalaev et al. (2005a), Drachev et al. (2006), and Kildishev et al. (2006). An attempt to explain the reflection spectra of a similar sample containing pairs of gold nanoparticles has been made

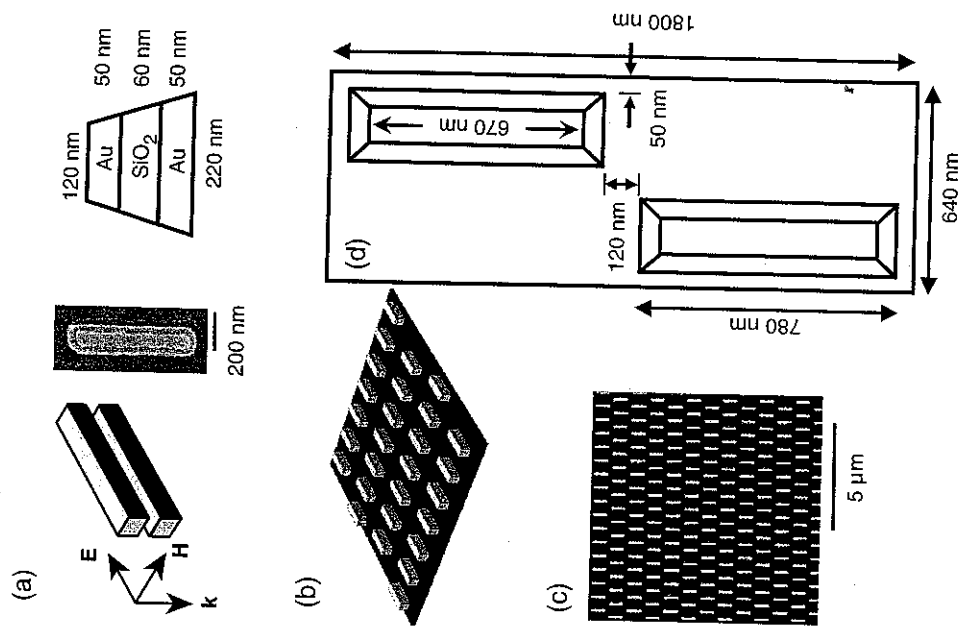


Fig. 3. (a) Left to right: scheme of nanorod pair and proper light polarization for negative index, SEM image, and dimensions. (b) Scheme of the arrangement of nanorod pairs. (c) SEM image of arranged nanorod pairs. (d) Dimensions of the arrangement (one unit cell is shown).

assuming a given dispersion of μ and ϵ (Grigorenko et al., 2005). However negative refractive index has been achieved in that work.¹

¹ Although the authors claim a negative magnetic response achieved in their metamaterial in visible range, validation of their result with our 3D FDTD code and an independent study performed at Penn State University using a hybrid finite element - boundary integral (PFEBI) technique showed only a feeble always-positive magnetic response (to be discussed elsewhere).

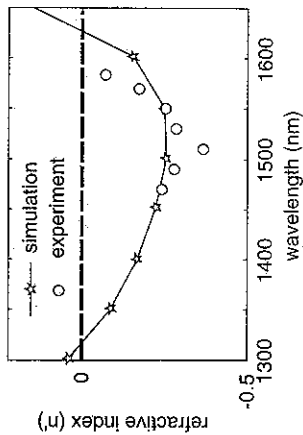


Fig. 4. Region of negative refraction for the real part of the refractive index of a layer of nanorod pairs shown in Fig. 3. Circles represent data, which are restored from experimentally determined transmission, reflection, and phase measurements. Stars represent FDTD simulation. A refractive index of $n' = -0.3 \pm 0.1$ was determined.

Figure 4 shows the results obtained in Shalaev et al. (2005a) for the real part of the refractive index of the metamaterial shown in fig. 3. The full circles show experimental results and the open triangles give the results as obtained from simulations using the finite difference method in time domain (FDTD). It is clearly seen that the real part of the refractive index becomes negative in the wavelength range from approximately 1400 to 1600 nm, which includes the important telecommunication band at 1500 nm. The figure gives a closer look to this frequency range. The experimental data prove that $n' = -0.3 \pm 0.1$ was obtained in Shalaev et al. (2005a).

It turns out to be not trivial to experimentally determine the exact value of the refractive index for a thin film. In the present case, the film of negative refraction was only 160 nm thick. Therefore, the straightforward method of determining n by applying Snell's law to the incoming and refracted beams cannot be used. A different method to unambiguously determine the refractive index requires the measurement of the transmission T , the reflectance R , and the absolute phases of the transmitted and reflected electric fields τ and ρ , respectively. If these four quantities are measured, the refractive index $n = n' + in''$ in a thin, passive ($n'' > 0$) film sandwiched between air (top) and a glass substrate (bottom) can be determined uniquely as has been discussed in Smith et al. (2002) and Kildishev et al. (2006) using transfer matrices

$$n = \frac{1}{k\Delta} \arccos \frac{1 - r^2 + n_s^2}{[1 + n_s - (1 - n_s)r]t}, \quad (1.2)$$

where $k = 2\pi/\lambda$ is the wave vector of light in vacuum, Δ the thickness of the thin film, n_s the refractive index of the glass substrate, and r and t the

complex reflection and transmission coefficients, respectively:

$$t = \sqrt{T} e^{i\tau}, \quad r = \sqrt{R} e^{i\rho}. \quad (1.3)$$

Figure 5(a) shows the transmission and reflection spectra of the NIM of fig. 3. In order to measure the absolute phase, the beam of a tunable semiconductor laser was split into two orthogonally polarized beams, where one beam passed through the NIM of thickness Δ while the other was used as a reference and passed only through the glass substrate at a spot not covered by the metamaterial (Drachev et al., 2006) (fig. 5(b)). The beams were recombined behind the glass substrate. The phase difference between the beam passing through the thin film and the reference

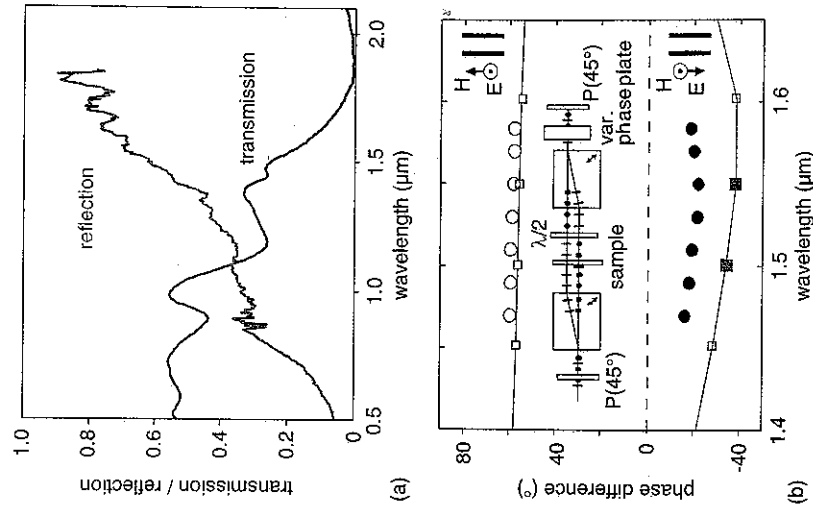


Fig. 5. (a) Measured transmission and reflection spectra of the sample shown in fig. 3(b). Setup for phase measurements. (b) Phase difference in the two light paths as shown in the inset. Circles are measured values, quadrangles and lines are from simulation. The light is delayed in case of TE polarisation (H-field parallel to rod pair, open symbols). In contrast, the phase is advanced in case of TM polarisation.

beam propagating only through air of the same thickness Δ was determined using interferometry (inset in fig. 5(b)). The phase τ was delayed in the metamaterial by approximately 60° compared to air in case of TE polarization (electric field perpendicular to the plane of rods). In contrast, τ was advanced by approximately 20° in case of TM polarization (fig. 5(c), Shalaev et al., 2005a). The phase shifts in reflection ρ were obtained for both polarizations in a similar way. The advancement of τ for TM polarization was an indirect evidence of $n' < 1$. However, to unambiguously prove that $n' < 0$, the complete set (T, R, τ, ρ) must be obtained, so that n can be reconstructed using eq. (1.3) (Kildishev et al., 2006).

Nevertheless, one can use pure phase measurements to make an estimate for n' as has been pointed out in Kildishev et al. (2006). In the case of low reflection ($R \ll 1$), the following equation holds:

$$n' \approx \frac{\tau}{k\Delta}, \quad (1.4)$$

while in the limit of strong reflection ($R \approx 1$) the following equation holds:

$$n' \approx \frac{\tau - \rho - \frac{\pi}{2}}{k\Delta}. \quad (1.5)$$

These two formula indeed give an upper and lower bound to the correct value of n' according to eqs. (1.2) and (1.3) (see fig. (6)) (Kildishev et al., 2006).

1.3.2. Voids

It is an interesting approach for NIMs to take the inverse of a resonant structure (Falcone et al., 2004), e.g., a pair of voids as the inverse of a pair of nanorods (Zhang et al., 2005a, b, 2006). The basic idea is illustrated in fig. 7(a). Instead of a pair of metal nanoellipses separated by an oxide, which are similar to the pair of rods in fig. (2), two thin films of metal are separated by an oxide and mounted on a glass substrate. Then, an elliptically shaped void is etched in the films (fig. 7(a), right-hand side), thus forming the negative of the original-paired metal ellipse structure (fig. 7(a), left-hand side). Both samples should have similar resonance behavior if the orientation of the electric and magnetic fields are also interchanged. FDTD simulations have been performed to determine the refractive index of void metamaterials (Kildishev et al., 2006). The dimensions were chosen according to fig. 7(b) in the simulations in order to match the dimensions of the experimental sample reported in Zhang et al. (2006).

The simulations were carried out for both cases of polarizations: the electric field oriented along the long axis of the elliptical voids and perpendicular to it. It is seen that n' becomes negative in both cases; however,

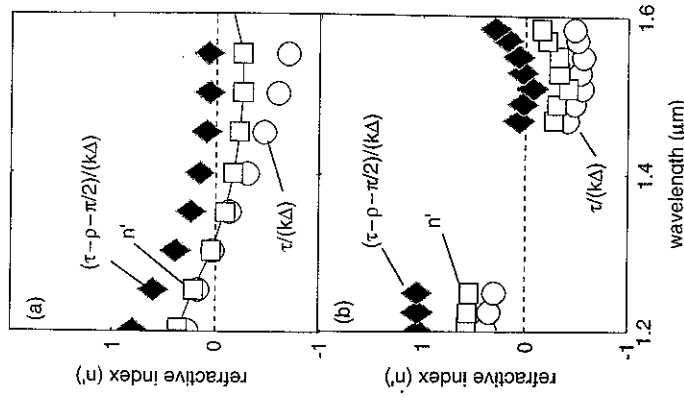


Fig. 6. Real part of the refractive index as determined by the exact formula (eq. (1.2), squares) or by phase-only assumptions according to eq. (2.3) (full diamonds) or eq. (1.4) (open circles). (a) Numerical simulations. (b) Experimental results.

the effect is more pronounced if the electric field is oriented along the short axis (fig. 7(c)). Furthermore, at approximately 1600 nm the real part of n is negative while the imaginary part is less than 1 indicating lower losses compared to the double-rod sample discussed before, where the imaginary part of the refractive index was 3 (Shalaev et al., 2005a). Experimental measurements with samples similar to those sketched in fig. 7(a), but with spherical voids instead of elliptical voids, confirmed a negative n' at a wavelength of 2 μm (Zhang et al., 2005b). The imaginary part n'' was large in that case, however it has been shown that further optimization can reduce n'' substantially (Zhang et al., 2006).

1.4. Pairs of metal strips for impedance-matched negative index metamaterials

Metamaterials using plasmon-resonant metal nanoparticles have two distinct problems, each of them reducing the overall transmission through the

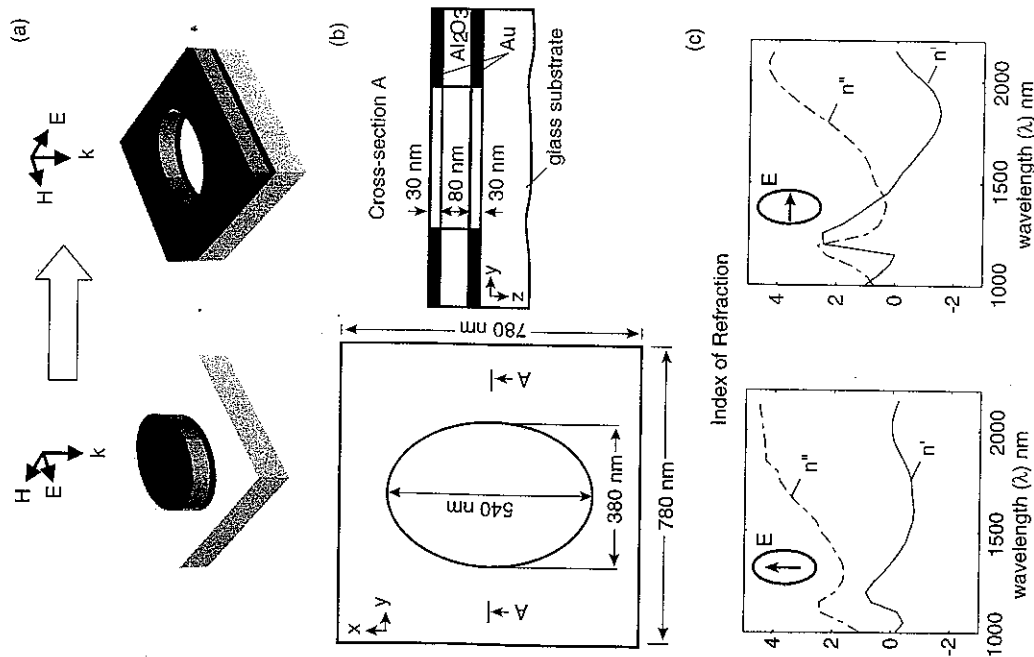


Fig. 7. (a) Left: nano-ellipse consisting of two 30 nm thick ellipses of gold separated by 80 nm of Al_2O_3 . Right: An elementary cell of coupled elliptic voids. (b) Dimensions of the voids. The voids are repeated periodically in 2D. (c) Refractive index $n = n' + i n''$ for light polarized parallel (left) or perpendicular (right) to the long axis of the voids as obtained from FDTD simulations.

metamaterial. The first one is absorptive losses (in terms of a large n''), because ohmic losses are generally large due to the excitation of localized plasmon resonances in the nanostructures. A possible solution to this problem will be discussed in the next section. In this section we will concentrate

on the second issue, which is impedance matching. The impedance is given by $\eta^2 = (\eta' + i\eta'')^2 = \mu\epsilon^{-1}$ and it is required that the impedances match at a boundary between two media in order to eliminate reflection. This condition is well known for microwaves and replaces Brewster's law for optical frequencies if $\mu \neq 1$ (Panina et al., 2002). Impedance is matched at a boundary between a NIM and air, if $\eta' \rightarrow 1$ and $\eta'' \rightarrow 0$ in the metamaterial.

In fig. 8(a) we introduce a metamaterial where the conditions $\eta \rightarrow 1 + 0i$, $n' < -1$, and $n'' < 1$ hold simultaneously for a visible wavelength. The structure consists of pairs of coupled silver strips. Both strips are 280 nm wide (x direction), 16 nm thick, and are infinitely long in the y direction. The two silver strips are separated in z direction by a 65 nm thick layer of Al_2O_3 . The pairs of strips are periodically repeated in x direction with a period of 500 nm. We assume air above and below the layer of strips. In our finite element frequency-domain (FEMFD) simulations this layer of metamaterial is illuminated from above with plane waves at normal incidence (along z direction). The electric field is polarized in x direction. The magnetic field, which is parallel to the strips, induces antiparallel currents in the two silver strips. This leads to a magnetic response of the structure. We use FEMFD calculations to determine the spectra of the permittivity (triangles) and the permeability (squares). It is seen that both are negative at wavelengths between 580 and 590 nm.

The spectra of the reflectance, transmission, absorption, refractive index, and impedance are displayed in fig. 9. It can be seen in fig. 9(a) that the transmission has a local maximum of 51% at 582 nm. This is because the reflection has a local minimum and the absorption is limited. Indeed, the impedance is matched quite well from 582 to 589 nm, i.e., $n' > 0.5$ and eventually reaching 1 at 586 nm, and simultaneously $|n''| < 0.5$ in the range 570–585 nm (fig. 9(c)). In total, this leads to a reflectance of less than 10% at 584 nm.

The absorption seems to have a local maximum at 586 nm, however it does not reproduce the spectrum of n'' . This is mainly because the reflection at the interface between air and the metamaterial hinders the electromagnetic radiation from entering the metamaterial at longer wavelengths, and therefore, the effective absorption of radiation inside the metamaterial is low for longer wavelengths. Still, it accounts for almost 90% of the losses in the range of the “reflectance window” at 584 nm. In summary of this section, we have shown that a metamaterial consisting of pairs of silver strips as depicted in fig. 8(a) can form an impedance-matched negative index material for visible light. The transmission is limited to 50% almost solely due to absorption, while reflection losses play a minor role.

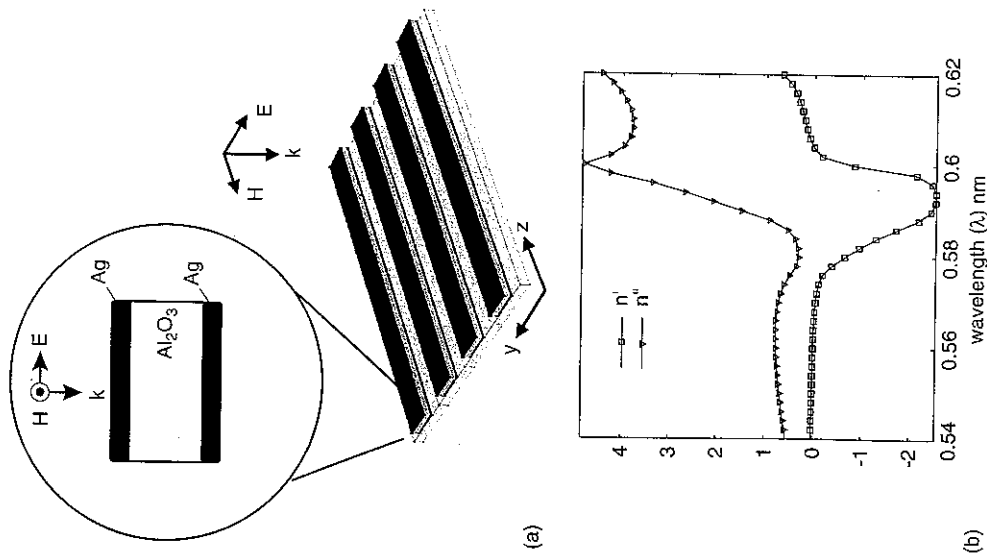


Fig. 8. (a) Double silver strips, separated by Al_2O_3 . The strips are infinitely long in y direction and periodically repeated in x direction. The H -field is oriented in the y direction. Currents in the strips are antiparallel if the H -field is polarized in y direction. (b) Real and imaginary parts of the refractive index (n' and n'') simulated with FEMFD.

1.5. Gain, compensating for losses

It has been pointed out recently that energy can be transferred from a gain material to surface plasmon polaritons (Sudarkin and Demkovich, 1989; Tredicucci et al., 2000; Avrutsky, 2004; Nezhad et al., 2004; Seidel et al., 2005) or to plasmons in metal nanostructures (Bergman and

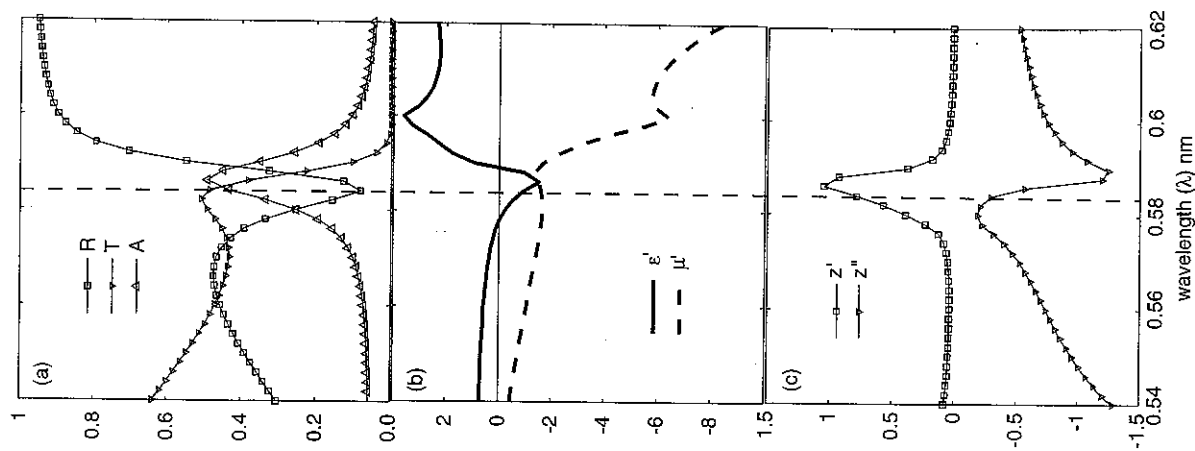


Fig. 9. Spectra of several optical constants of the structure shown in Fig. 8. (a) Reflection R , transmission T , and absorption A spectra. (b) Real parts of permittivity (ϵ') and permeability (μ') and imaginary part of the impedance. The vertical dashed line at 584 nm indicates a spectral region where the reflection is minimal, the transmission is high, and the real parts of permittivity and permeability are close to -1 , while the real part of the impedance is close to $+1$, indicating impedance matching to air.

Stockman, 2003; Lawandy, 2004) using stimulated emission. Specifically, continuous thin films of metal have been used to confine lasing modes in quantum cascade lasers to the gain region and also to guide the lasing modes by surface plasmon modes (Tredicucci et al., 2000). Ramakrishna and Pendry suggested to staple gain materials such as semiconductor laser materials in between the negative index (or metal) layers of stacked near-field lenses (Ramakrishna and Pendry, 2003) in order to remove absorption and improve resolution. The requirement of a perfect near-field lens, where thin layers of positive and negative index materials are alternated, is that $\epsilon_P = -\epsilon_N$ and simultaneously $\mu_P = -\mu_N$, where the subscripts denote materials constants of positive (P) and negative (N) materials. This requirement naturally includes the conditions $\epsilon_P'' = -\epsilon_N''$ and $\mu_P'' = -\mu_N''$, i.e., the positive layers must provide gain in order to optimize the lens (Ramakrishna and Pendry, 2003).

In our discussion we would like to turn to the refractive index rather than the permittivity and the permeability, because the absorption (α) and gain (g) coefficients are more straightforwardly connected to the refractive index $n'' = \lambda(\alpha - g)/4\pi$. Further, instead of alternating negative and positive index materials we propose to "submerge" the negative index structures (e.g., containing metal nanorods) in gain media as shown in fig. 10. This could be achieved, for example, by spin-coating a solution of laser dye molecules or π -conjugated molecules on top of the negative index structures. Applying semiconductor nanocrystals (NCs) would be an alternative approach.

One might question whether the metal nanostructures nullify any attempt to amplify electromagnetic fields using gain materials in their close vicinity because gold nanoparticles are well known to quench fluorescence in an extremely efficient manner (Dulkeith et al., 2002; Imahori et al., 2004). In contrast, however, working solid state and organic

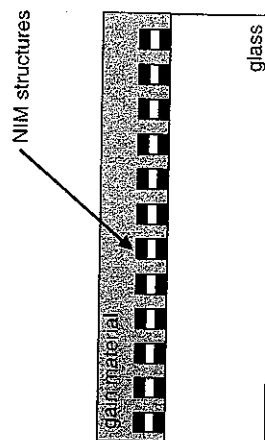


Fig. 10. Negative index metamaterial (e.g., double rods), filled with gain medium, e.g., a solid solution of dye molecules in a matrix.

semiconductor lasers show that sufficient gain can be provided so that in devices containing metal layers or metal nanoparticles the losses can be compensated. For instance, it has been shown that an optically pumped organic laser comprising a metal-nanoparticle distributed feedback (DFB) grating needs only a marginally increased pumping threshold (compared to organic lasers with metal-free DFB gratings) to be operative (Stehr et al., 2003). In the case of infrared quantum cascade lasers (QCL), a wave guiding metallic layer was shown to be beneficial for the laser power output (Tredicucci et al., 2000). This astonishing result is due to an increased overlap of the surface plasmon-guided mode profile with the gain region (the quantum cascade structure, in this case). This overlap offsets the increased losses (compared to a metal-free QCL) resulting from surface plasmon excitation. The net effect is an overall improved laser performance. We therefore conclude that it should indeed be feasible to use gain materials in order to compensate for the losses introduced by the resonant plasmonic metal nanoparticles in NIMs.

We want to give a specific example on the basis of the sample shown in figs. 8 and 9. For the moment, we assume that the metal strips are submerged in a 200 nm thick layer of gain material (fig. 11(a)). We further assume that the gain material and the metal strips do not influence each other. This is an assumption that certainly needs to be discussed, but for the moment we shall assume that the gain of the material is not influenced by the metal strips. At the wavelength of least reflectance (due to impedance matching, $\lambda = 584$ nm), the strip material shows absorption of approximately 45% (fig. 9(a)). Applying Lambert-Beer's law and assuming that the absorptive loss should be fully compensated by the 400 nm thick gain layer, it turns out that a gain of $g = 3 \times 10^4 \text{ cm}^{-1}$ is required. Let us further assume that we use Rhodamine 6G dissolved in some optically inert polymer. Rhodamine 6G has a stimulated emission cross-section of $\sigma_{SE} = 3 \times 10^{-16} \text{ cm}^2$ (Holzer et al., 2000) and therefore the concentration of excited dye molecules should be 170 mM. Alternatively, semiconductor nanocrystals such as CdSe NCs could be applied. It has been shown in Leatherdale et al. (2002) that the absorption cross-section per NC volume can be as large as 10^5 cm^{-1} . Because g and α are usually of similar magnitude, we conclude, that densely packed nanocrystal films can show gain in the order of $g \approx 10^5 \text{ cm}^{-1}$.

It is seen that the dye or nanocrystal concentrations need to be quite high to compensate for the losses. However, we have assumed in our rough estimation that the gain of the material in between the metal strips is not affected by the local fields in the vicinity of these metal strips. These fields can be quite high due to nanoplasmonic resonances. In fact, it has

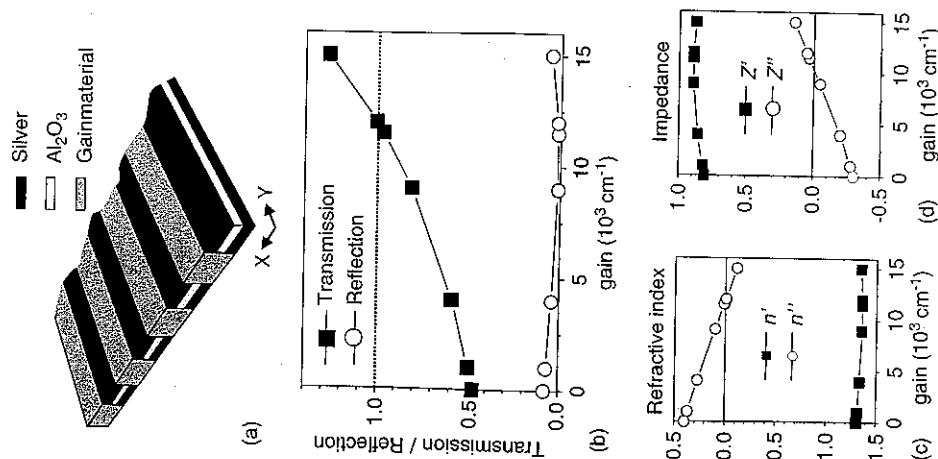


Fig. 11. (a) Same sample as in Fig. 8(a), but with gain providing material in between the double silver strips. Air is assumed above and below the layer, and the layer is irradiated with a plane wave (584 nm) from above, H-field polarized along the y direction. (b) Transmission and reflection as a function of the gain. At $g = 12,000 \text{ cm}^{-1}$, gain and losses cancel each other. Interestingly, the reflection shows also a minimum at $g = 12,000 \text{ cm}^{-1}$. (c, d) Refractive index and impedance as a function of gain. $n' \approx -1.35$ for all investigated gain levels.

been pointed out by Kim et al. (1999) and Lawandy (2004) that a gain medium and localized plasmonic resonance may lead to extremely high effective polarizabilities of the combined system. Therefore, the possibility may arise that each pair of gold nanorods as shown in fig. 2, or each strip as in fig. 8, shows a much larger response to an incoming electric field as the same metal structure in air.

In the example given above we have neglected that the gain material is in intimate contact with the silver strips. In order to get a better picture, we applied FEMFD simulations on the model shown in fig. 11(a). We took the same structure as shown in fig. 8, but now filled the gaps in between the double silver strips with a material that provides a fixed amount of gain between 0 and $15 \times 10^3 \text{ cm}^{-1}$. Figures 11(b-d) show the transmittance (T), reflectance (R), refractive index (n' and n''), and impedance (η' and η'') as a function of gain (g). We found that at a gain of $12 \times 10^3 \text{ cm}^{-1}$ the structure becomes transparent (fig. 11(b)), while the real part of the refractive index n' is almost unaffected by the gain material (fig. 11(c)). Furthermore, the impedance that has already been matched quite well without the gain medium (fig. 9) improves further when gain is applied, i.e., $\eta' \approx 1$ and $\eta'' \approx 0$ for $g = 12 \times 10^3 \text{ cm}^{-1}$ (fig. 11(d)). The exact results for a gain of $g = 12 \times 10^3 \text{ cm}^{-1}$ are $n' = -1.355$, $n'' = -0.008$, $\eta' = 0.89$, $\eta'' = 0.05$, $T \approx 100.5\%$, and $R \approx 1.6\%$.

Actually, if a critical magnitude of gain is surpassed, the polarizability and the field enhancement do not depend on nanoparticle shape or material any longer, but are solely limited by gain saturation in the gain medium (Lawandy, 2004). At present, we have not included gain saturation in our model. It could be envisioned that the gain material does not “simply” restore energy, which is lost due to absorption by the metal nanostructures, but it becomes an instrumental element of the NIM, e.g., heavily increasing the negative response of the pairs of nanorods. This will allow the design of NIMs of less overall metal content. The density of pairs of rods or the size of each pair may be reduced, while the overall effective negative response of the metamaterial remains strong. This exciting field certainly needs more consideration, which will be discussed elsewhere.

§ 2. Optical characteristics of cascaded NIMs

A possible approach to designing NIMs is a periodic array of elementary coupled metal-dielectric resonators. This work takes a closer look at approaches, which simultaneously provide fast calculation of the field inside a given metamaterial arranged of elementary periodic layers and calculation of its effective parameters. First, consider a simplified approach to defining equivalent optical properties of an elementary NIM layer (Kildishev et al., 2006). With this approach, an effective refractive index is ascribed to a layer of NIM, as if it were a layer of homogeneous medium. This assumption suggests that the periodic structure of NIM does not diffract the incident plane wave, and a classical direct problem of plane-wave propagation

through a multilayer structure of homogeneous materials can be used in accordance with Born and Wolf (Born and Wolf, 1964). For a given monochromatic incident light it is then possible to measure complex reflectance and transmittance coefficients (r and t) and then unambiguously retrieve the refractive index of the NIM sample. This effective parameter can be conveniently retrieved from a characteristic matrix of a homogeneous film at normal incidence (Kildishev et al., 2006).

Provided that the characteristic matrix ($\mathbf{M}$) is decomposed using a diagonal matrix of eigenvalues ($\mathbf{A}$) and a matrix of eigenvectors ($\mathbf{V}$) as $\mathbf{M} = \mathbf{V}\mathbf{A}\mathbf{V}^{-1}$, the transmission through an elementary layer is given by

$$\mathbf{Q}_0 = \mathbf{M}\mathbf{Q}, \quad (2.1)$$

where

$$\mathbf{Q} = \begin{pmatrix} H_2 \\ \eta_0^{-1} E_2 \end{pmatrix}, \quad \mathbf{Q}_0 = \begin{pmatrix} H_1 \\ \eta_0^{-1} E_1 \end{pmatrix},$$

$$\mathbf{V} = \begin{pmatrix} 1 & 1 \\ -\eta & \eta \end{pmatrix} = \begin{pmatrix} 1 & 0 \\ 0 & \eta \end{pmatrix} \begin{pmatrix} 1 & 1 \\ -1 & 1 \end{pmatrix}, \text{ and}$$

$$\mathbf{A} = \begin{pmatrix} e^{mk\Delta} & 0 \\ 0 & e^{-mk\Delta} \end{pmatrix}$$

E_1 , and H_1 are the field values at the source-side interface and E_2 , H_2 are the values at the output interface, Δ is a thickness of the film, $k = 2\pi/\lambda$, (λ is free-space wavelength), $\eta = \sqrt{\mu/\epsilon}$ and $n = \sqrt{\mu\epsilon}$ are the effective impedance and the refractive index of the film, respectively, and $\eta_0 = \sqrt{\mu_0/\epsilon_0}$ is the impedance of free-space.

With this simple assumption it is thought that a cascaded bulk material can be arranged using a stack of q elementary layers with an effective transformation matrix $\mathbf{M}^q = \mathbf{V}\mathbf{A}^q\mathbf{V}^{-1}$. In general, this straightforward approach assumes that the spatial harmonics of each layer interact only with the same harmonics of other layers in the stack. In essence, this loose assumption ignores any transformation of a given incident harmonic into the spatial harmonics of different order, which are either reflected or transmitted.

To illustrate this issue, consider another approach to obtaining effective parameters of a multilayer NIM arranged of thin infinite elementary layers with periodic distribution of elementary materials. Essentially, the enhanced method follows the recipe for a classical case of stratified media (e.g., Luneburg, 1964).

2.1. Bloch-Floquet waves in cascaded layers

A simpler 2D example is used here to illustrate the approach, just because derivations for the spatial harmonic analysis (SHA) in 2D are less difficult. Consider a single period (l) of an infinite interface of a free-space domain with the domain of a material characterized by a set of stepwise continuous permittivity values ($\epsilon_{1,1}$, $\epsilon_{2,1}$, $\dots$), as shown in fig. 12(a). Introduce a local coordinate system, with the unit normal ($\hat{x}$), the unit transverse vector ($\hat{z}$) and the tangent unit vector ($\hat{y}$). Provided that a TM

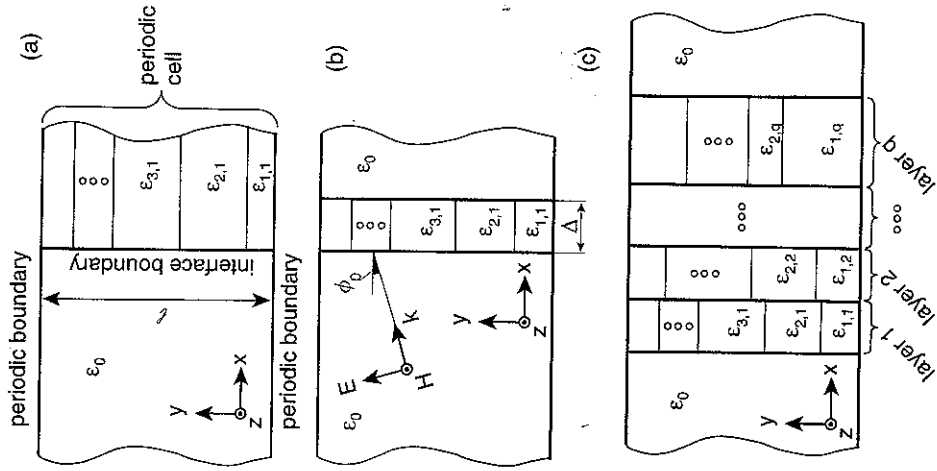


Fig. 12. (a) Interface of an elementary layer of NIM with free space. (b) An isolated elementary layer. (c) A cascaded multilayer NIM.

boundary-value problem is taken ($\vec{H} = \hat{z}h$), then only the tangential components of the $\vec{H}$ and $\vec{E}$ field distributions at the interfaces are required in this case. Then, consider two scalar fields (h and d) as the distribution of the transverse magnetic field ($h = \hat{z} \cdot \vec{H}$) and the distribution of the electric field ($d = \hat{y} \cdot \vec{E}$). A monochromatic Maxwell equation $\vec{y} \cdot (\nabla h \times \hat{z}) = -i\omega d$ couples the fields

$$d = (i\omega)^{-1} h'. \quad (2.2)$$

Here the normal derivative of h is denoted as h' , where $h' = \hat{x} \cdot \nabla h$.

The core of any SHA approach is the transformation of the fields from a physical space to spatial spectral space using available proper functions (g''). Provided that h and g'' are sets of discrete values obtained at a uniform grid on l , these sets are considered as two vectors (h and g''). The following sum: $\sum_{p=-p_{\max}}^{p_{\max}} g''^* g''$ is considered here as a scalar product of two vectors (g'', g'') _{l} , where p is a point of the grid on l arranged of $\tilde{p} = 2p_{\max} + 1$ points. Note that the proper functions g'' are orthonormal on l , i.e., $(g'', g'') = \delta(m - v)$.

Isolate, for example, the first elementary layer as shown in fig. 12(b). The magnetic field is defined by $h_1 = \sum_{m=-\infty}^{\infty} g''^m c_{r,1}^m + g''^m c_{t,1}^m$, where g''^m and g_r^m are transmitted and reflected elementary fields of order m , respectively. In essence, the field h_1 is decomposed into elementary fields (the Bloch-Floquet waves), which are orthonormal on l . In a truncated approximation, $|m| < m_{\max}$, the vectors g''^m can form a $\tilde{p} \times \tilde{m}$ matrix g , $\tilde{m} = 2m_{\max} + 1$, and the complex magnitudes of reflected and transmitted fields ($c_{r,1}^m$ and $c_{t,1}^m$) can be taken as the components of two different $\tilde{m}$ -dimensional vectors $c_{r,1}$ and $c_{t,1}$. Then, the magnetic field in a matrix form is defined by $h_1 = g_{t,1} c_{t,1} + g_{r,1} c_{r,1}$.

The Bloch-Floquet theorem allows for the separation of variables, $g_t = v_1 u_1$ and $g_r = v_1 u_1^{-1}$, where u is a $\tilde{m} \times \tilde{m}$ matrix exponential, $u_1 = \exp i k_{x,1} x$, of a proper values matrix, $k_{x,1}^m$; and v_1 is a $\tilde{p} \times \tilde{m}$ matrix constructed of orthogonal vectors v_1^m . For the free-space case, indices in u , v , and k_x^m are dropped, and the proper functions u are defined through $k_x^m = [1 - (k_y^m)^2]^{1/2}$, where $k_y^m = m\lambda l^{-1} + \sin \phi_0$ and ϕ_0 is the angle of incidence (shown in fig. 12(b)). The wavefront $v'' = \tilde{p}^{-1/2} \exp i k_y^m k y$ is just an orthonormal Fourier component of the m -th order.

2.2. Eigenvalue problem

To obtain both $k_{x,1}$ and v_1 for a given elementary layer, where the permittivity of elementary materials (a piecewise continuous function ε_1) is periodic in y direction, but constant in x direction, it is necessary to

attain an eigenvalue problem formulation. Since in this case (Luneburg, 1964),

$$k^2 \varepsilon_1 h_1 + \nabla^2 h_1 - f_1 \partial_y h_1 = 0, \quad (2.3)$$

where f_1 is the logarithmic derivative, $f_1 = \varepsilon_1' \varepsilon_1^{-1}$, ε_1 and ε_1^{-1} are $\tilde{p} \times \tilde{p}$ diagonal matrices, and k^2 a scalar.

Then, the above equation can be rewritten using $h_1 = v_1 (u_1 c_{t,1} - u_1^{-1} c_{r,1})$. Next, introducing a_1 as a $\tilde{m} \times \tilde{m}$ matrix mapping an orthogonal basis v_1 into the free-space basis v ($v_1 = v a_1$), substituting $i(v^{-1} f_1 v) k_y$ with $i(k_y v^{-1} \varepsilon_1 v - v^{-1} \varepsilon_1 v k_y) w^{-1} \varepsilon_1^{-1} v k_y$ and using γ_1 for $v^{-1} \varepsilon_1 v$ and i for the $\tilde{m} \times \tilde{m}$ identity matrix, eq. (2.3) is further simplified as

$$a_1 k_{x,1}^2 a_1^{-1} = \gamma_1 (i - k_y \gamma_1^{-1} k_y). \quad (2.4)$$

The transform a_1 is required because in contrast with the free-space case, each wavefront v_1^m in an elementary inhomogeneous layer is not a single Fourier component anymore; but as a "physical function"² it still can be expressed as a superposition of the Fourier components. Note that eq. (2.4) is written in an eigenvalue form since $k_{x,1}^2$ is a diagonal matrix. The equation can be solved either numerically or analytically for both $k_{x,1}^2$ and a_1 , provided that γ_1 and k_y are known.

2.3. Mixed boundary-value problem

Transverse magnetic field continuity together with the conservation of the tangential electric field gives the standard boundary conditions (BC), $h_0 = h_1$, $\varepsilon_1 h_0' = h_1'$, where the pairs h , h' represent the magnetic field and its normal derivative just before and after the interface; ε_1 is the value of permittivity. First, to simplify the notations (and further programming) a matrix nomenclature is defined as

$$\mathbf{n}(m_1, m_2) = \begin{pmatrix} m_1 & m_2 \\ m_2 & m_1 \end{pmatrix}, \quad \mathbf{d}(m_1, m_2) = \begin{pmatrix} m_1 & 0 \\ 0 & m_2 \end{pmatrix}, \quad \mathbf{i} = \begin{pmatrix} i & -i \\ i & i \end{pmatrix}, \quad (2.5)$$

$$\mathbf{s}(v_1, v_2) = \begin{pmatrix} v_1 \\ v_2 \end{pmatrix},$$

where $\mathbf{d}$, $\mathbf{n}$, and $\mathbf{i}$ are partitioned square matrices, 0 , i are two $\tilde{m} \times \tilde{m}$ matrices (the null matrix and the identity matrix, respectively), and m_1, m_2 are general square matrices of the same size; $\mathbf{s}$ is a stacked vector made of two equal vectors (v_1, v_2) with $\tilde{m}$ components.

² That is, a piecewise continuous function with limited variation on l .

After using $h_0 = v(u_{c,0} - u^{-1}c_{r,0})$, $h_1 = v a_1(u_1 c_{x,1} - u_1^{-1}c_{r,1})$, and taking the normal derivatives, a spectral form of the BC is

$$c_0 = s_1 c_1. \quad (2.6)$$

Here $c_0 = \mathbf{d}(u, u^{-1})s(c_{i,0}, c_{r,0})$, $c_1 = \mathbf{d}(u_1, u_1^{-1})s(c_{x,1}, c_{r,1})$, and $s_1 = \mathbf{i}^{-1}\mathbf{d}(a_1, k_x^{-1}\gamma_1^{-1}a_1 k_{x,1})\mathbf{i}$.

At the second interface (as shown in fig. 12(b)), the equation for the elementary layer is given by

$$b_1 c_1 = s_1^{-1} c_2. \quad (2.7)$$

Here $b_1 = \mathbf{d}(\beta_1, \beta_1^{-1})$, the matrix exponential $\beta_1 = \exp i k_{x,1} \delta_1$ adjusts the phases for a scaled thickness of layer ($\delta_1 = 2\pi\Delta_1/\lambda$).

Combining eqs. (2.6) and (2.7) gives the following form:

$$c_0 = s_1 b_1^{-1} s_1^{-1} c_2. \quad (2.8)$$

Since k_x is given as a common matrix for all layers, a possible alternative is employing the following normalization $\mathbf{c} = i\mathbf{c}$, where the upper and lower partitions of $\mathbf{c}$ correspond to a magnetic component and a normalized electric component, respectively. These Fourier components are both continuous across any interlayer interface and form the basis for wave matching. From now on the arguments in $\mathbf{d}(\tilde{a}^{-1}, \tilde{k}_x^{-1} \tilde{a}^{-1} \tilde{\gamma})$ are dropped and a subscript is added to denote a layer number starting from the source side; thus, eq. (2.8) is simplified to

$$\mathbf{c}_0 = \mathbf{d}_1 \mathbf{b}_1^{-1} \mathbf{d}_1^{-1} \mathbf{c}_2, \quad (2.9)$$

where the linear operators $\mathbf{d}_1 = \mathbf{d}(a_1, k_x^{-1}\gamma_1^{-1}a_1 k_{x,1})$ and $\mathbf{b}_1 = i\mathbf{b}_1 \mathbf{i}^{-1}$ are individual for each layer with a given distribution of elementary materials (γ_1), and defined matrices of the proper values $k_{x,1}$, and the proper vectors a_1 .

If the trivial case of a uniform slab of permittivity ϵ_1 is taken, then $a_1 = \mathbf{i}$, $\gamma_1 = \epsilon_1 \mathbf{i}$ and a generalized analog of eq. (2.1) is

$$\begin{pmatrix} \tilde{h}_0 \\ \tilde{h}'_0 \end{pmatrix} = \begin{pmatrix} \mathbf{i} & \mathbf{o} \\ \mathbf{o} & \eta_1 \end{pmatrix} \begin{pmatrix} \mathbf{i} & -\mathbf{i} \\ \mathbf{i} & \mathbf{i} \end{pmatrix} \begin{pmatrix} \beta_1^{-1} & \mathbf{o} \\ \mathbf{o} & \beta_1 \end{pmatrix} \begin{pmatrix} \mathbf{i} & -\mathbf{i} \\ \mathbf{i} & \mathbf{i} \end{pmatrix}^{-1} \quad (2.10)$$

$$\begin{pmatrix} \mathbf{i} & \mathbf{o} \\ \mathbf{o} & \eta_1^{-1} \end{pmatrix} \begin{pmatrix} \tilde{h}_2 \\ \tilde{h}'_2 \end{pmatrix},$$

where the tangential fields are the correspondent matrices of the Fourier transforms, $\tilde{h} = v^{-1}h$, $\tilde{h}' = (kv)^{-1}h'$, $\eta_1 = \text{diag}(\epsilon_1^{-1}k_x^m)$ (from eq. (2.4), $k_{x,1}^2 = \epsilon_1 \mathbf{i} - k_y^2$, and $k_{x,1}^m = [\epsilon_1 - (k_y^m)^2]^{1/2}$). Then for example, validation of eq. (2.10) for a plane wave at normal incidence gives a familiar result,

shown earlier in eq. (1.1). Note that in general, the following identity holds:

$$\begin{pmatrix} \tilde{h}_0 \\ \tilde{h}'_0 \end{pmatrix} = \begin{pmatrix} a_1 & \mathbf{o} \\ \mathbf{o} & k_x^{-1}\gamma_1^{-1}a_1 k_{x,1} \end{pmatrix} \begin{pmatrix} \cos k_{x,1}\delta_1 & -i\sin k_{x,1}\delta_1 \\ -i\sin k_{x,1}\delta_1 & \cos k_{x,1}\delta_1 \end{pmatrix} \begin{pmatrix} \tilde{h}_2 \\ \tilde{h}'_2 \end{pmatrix}, \quad (2.11)$$

where $\cos k_{x,1}\delta_1$ and $\sin k_{x,1}\delta_1$ are arranged of the adequate matrix exponentials.

2.4. A simple validation test

A simplified single-layer model for validating the simulation method is shown in fig. 13(a). The sample structure is intentionally made of a very thin metallic grating (with 10 nm thickness). The grating is arranged of 400 nm gold strips separated by narrow strips of silica, the period of the structure is 480 nm. A large aspect ratio of the metallic strips and large electric resonance at about 1.2 μm are among the main challenges of the test model. To obtain a good set of the reference data the structure was simulated using a commercial software package with fifth-order finite elements. The validity of the FEM solution was verified by using the same model with different levels of additional meshing refinement and an adaptive solver. The results were stable upon the use of 41,000 degrees of freedom (field variables), where the bulk of the resources had been spent for the free-space buffer, nonreflecting layers, and adequate meshing at the corners.

In contrast to FEM, the spatial harmonic analysis appeared much more efficient. The problem was stabilized after the use of 11 eigenvalues with the calculation time of about 100 times less (4s) versus the FEM solver with the same number of wavelength points and the same hardware. It should be noted that the amount of simulation time using SHA is approximately proportional to the total number of elementary layers and scales approximately as the square (or cube) of the total number of eigenvalues in 2D (or 3D) problems, while the performance of FEM solvers is decreasing very moderately with the increase in the total number of stacked layers.

Both models appeared to be quite sensitive to the material properties of the metal. Although in both cases, the interpolated complex refractive

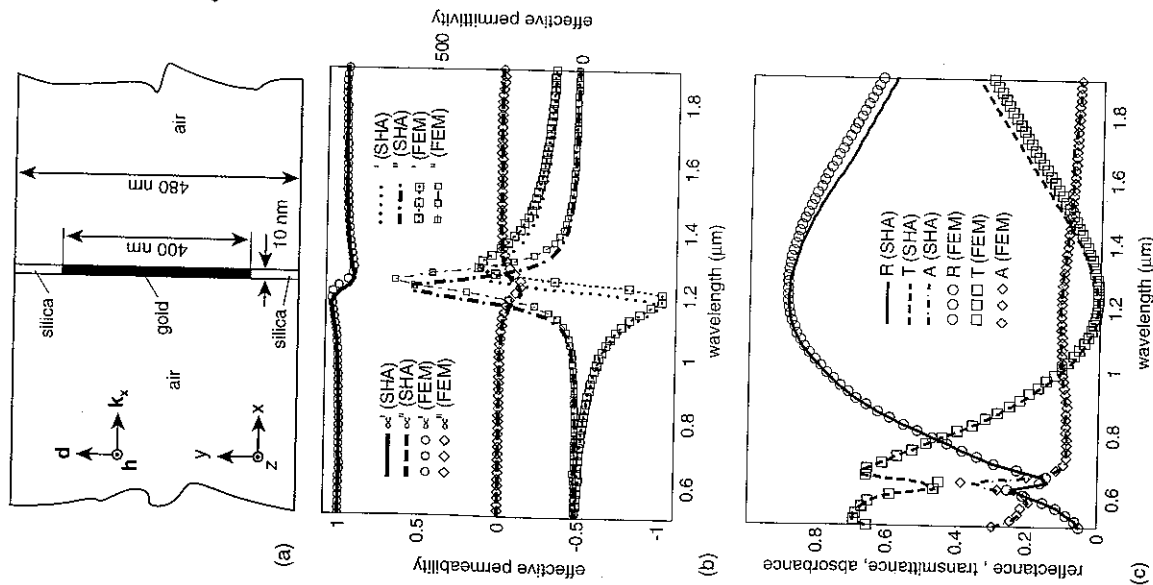


Fig. 13. (a) Sketch of a resonant elementary layer. (b) Effective permeability and permittivity obtained for the elementary layer using FEM and SHA (using 11 eigenvalues). (c) Comparison of the reflection, transmission, and absorption spectra obtained in simulations using FEM and SHA (11 eigenvalues).

index was based on the experimental table of Johnson and Christy (1972). In our opinion, the overall differences in the effective characteristics shown in figs. 13(b) and 13(c) are most likely due to the differences in the interpolation approach used in the FEM solver.

Above a simple validation of the modeling approach, the test model of fig. 13(a) reveals all the typical features of the periodic structures with localized plasmonic resonances, e.g., at the same wavelength the electric resonance is always accompanied by a satellite magnetic antiresonance and vice versa.

2.5. Cascading the elementary layers

Using eq. (2.8) cascading a subset of q elementary layers (depicted in fig. 12(b)) gives

$$c_0 = w_q c_q. \quad (2.12)$$

Here $w_q = \prod_{v=1}^q \tilde{w}_v$, $\tilde{w}_v = s_v b_v^{-1} s_v^{-1}$ is an elementary transform due to v -th layer and q the total number of layers in the subset stack.

2.6. Reflection and transmission coefficients

The major workload in the above method falls on the calculation of the proper values and vectors ($k_{x,v}$ and a_v) for each elementary layer. Once the values are obtained, the characteristic matrices of each layer are arranged as $s_v b_v^{-1} s_v^{-1}$.

Introducing the transformations $r_v c_{t,v} = c_{t,v}$, $t_v c_{t,v} = c_{t,q}$ (with the initial values given by $t_q = i$ and $r_q = 0$), and then from eq. (2.12) the matrices of spatial spectral reflectance and transmittance are defined as

$$\begin{aligned} r_{v-1} &= (w_v^{1,0} + w_v^{1,1} \rho_v) (w_v^{0,0} + w_v^{0,1} r_v)^{-1}, \\ t_{v-1} &= t_v (w_v^{0,0} + w_v^{0,1} r_v)^{-1}, \end{aligned} \quad (2.13)$$

where the characteristic matrix is partitioned as

$$\tilde{w} = \begin{pmatrix} w^{0,0} & w^{0,1} \\ w^{1,0} & w^{1,1} \end{pmatrix}.$$

The partitions are calculated using s_v , which is a matrix with symmetrical partitions, $s_v = \mathbf{n}(s_v^0, s_v^1)$ with $s_v^0 = \frac{1}{2}(k_x^{-1} \gamma_v^{-1} a_v k_{x,v} + a_v)$ and $s_v^1 = \frac{1}{2}(k_x^{-1} \gamma_v^{-1} a_v k_{x,v} - a_v)$ and a similar matrix $s_v^{-1} = \mathbf{n}(s_v^0, s_v^1)$ with

$\tilde{s}_v^0 = \frac{1}{2}(k_{x,v}^{-1}a_v^{-1}\gamma k_x + a_v^{-1})$ and $\tilde{s}_v^1 = \frac{1}{2}(k_{x,v}^{-1}a_v^{-1}\gamma k_x - a_v^{-1})$, then

$$\begin{aligned} w_v^{0,0} &= s_v^0 \beta_v^{-1} \tilde{s}_v^0 + s_v^1 \beta_v \tilde{s}_v^1, & w_v^{0,1} &= s_v^0 \beta_v^{-1} \tilde{s}_v^1 + s_v^1 \beta_v \tilde{s}_v^0, \\ w_v^{1,0} &= s_v^1 \beta_v^{-1} \tilde{s}_v^0 + s_v^0 \beta_v \tilde{s}_v^1, & w_v^{1,1} &= s_v^1 \beta_v^{-1} \tilde{s}_v^1 + s_v^0 \beta_v \tilde{s}_v^0. \end{aligned} \quad (2.14)$$

Thus, for example, a single-layer structure is calculated as follows: $t_0 = (w_1^{0,0})^{-1}$, $r_0 = w_1^{1,0} (w_1^{0,0})^{-1}$; then, the transmitted and reflected Bloch-Floquet waves are $c_{\alpha,1} = t_0 c_{\alpha,0}$ and $c_{\gamma,0} = r_0 c_{\alpha,0}$.

2.7. Discussions

It also follows from the analysis of eqs. (2.8)–(2.14) that:

- (i) None of asymmetric multilayer composites can be effectively described either by the simplified homogenization approach (eq. (2.1)) or through its generalized analog (eq. (2.10)). (A multilayer composite is asymmetric, if it contains an odd number of elementary layers, and the layers are not mirror-symmetric relative to the central layer; all structures with even number of distinct layers are always asymmetric.)
- (ii) Effective optical parameters (including an effective negative refractive index) obtained in a single symmetric subset of elementary layers may not guarantee the same effective parameters in a bulk material arranged of identical subsets, not just because of absorptive losses, but also due to new interactions of near-field waves introduced by the use of cascading.

To illustrate (i) consider a classical example of a subset structure with two homogeneous lossless layers (Born and Wolf, 2002, p. 72). The characteristic matrix of two layers with thicknesses Δ_1 and Δ_2 , and indices n_1 and n_2 , is computed using eq. (2.10) as $w_2 = \tilde{w}_1 \tilde{w}_2$, with $\tilde{w}_1 = \mathbf{d}_1 \mathbf{b}_1^{-1} \mathbf{d}_1^{-1}$ and $\tilde{w}_2 = \mathbf{d}_2 \mathbf{b}_2^{-1} \mathbf{d}_2^{-1}$. The effective characteristic matrix (w_{eff}) of an equivalent single layer, which is defined as

$$\mathbf{w}_{\text{eff}} = \begin{pmatrix} \mathbf{w}_{\text{eff}}^{0,0} & \mathbf{w}_{\text{eff}}^{0,1} \\ \mathbf{w}_{\text{eff}}^{1,0} & \mathbf{w}_{\text{eff}}^{1,1} \end{pmatrix} = \begin{pmatrix} \cos n_{\text{eff}} k_x \delta & -i \eta_{\text{eff}} \sin n_{\text{eff}} k_x \delta \\ -i \eta_{\text{eff}}^{-1} \sin n_{\text{eff}} k_x \delta & \cos n_{\text{eff}} k_x \delta \end{pmatrix}, \quad (2.15)$$

(using a scaled thickness $\delta = k(\Delta_1 + \Delta_2)$ and the effective index n_{eff} , should be equal to the characteristic matrix of the double layer w_2 . To be equivalent to w_{eff} , the product $w_2 = \tilde{w}_1 \tilde{w}_2$, must have identical diagonal partitions since $w_{\text{eff}}^{0,0} = w_{\text{eff}}^{1,1}$ in eq. (2.15). This is true only if the product

commutes, i.e., $\tilde{w}_1 \tilde{w}_2 = \tilde{w}_2 \tilde{w}_1$, leaving the only trivial case of $n_1 = n_2$ possible. Therefore, even a simple stack of two distinct lossless films cannot be adequately modeled by a single effective layer. Physically, the condition $\tilde{w}_1 \tilde{w}_2 = \tilde{w}_2 \tilde{w}_1$ means that the effective parameters of a multilayer NIM should not depend on the side that is chosen for illumination, i.e., its structure should be symmetric.

Note that although $\tilde{w}_1 \tilde{w}_2 \tilde{w}_3 = \tilde{w}_3 \tilde{w}_2 \tilde{w}_1$ is always true for any triple-layered structure, since the first and the last layers are equal ($\tilde{w}_1 = \tilde{w}_3$), the homogenization of $w_3 = \mathbf{d}_1 (\mathbf{b}_1^{-1} \mathbf{d}_{12} \mathbf{b}_2^{-1} \mathbf{d}_{12}^{-1} \mathbf{b}_1^{-1}) \mathbf{d}_1^{-1}$ is not very simple even for the structure with homogeneous elementary layers.

Now to exemplify (ii) consider a cascaded structure arranged of identical symmetric substructures. A three-layer substructure is of natural choice here, since the majority of known structures falling in this category (as shown in figs. 2, 3, 7, and 8) are $w_3 = s_1 b_1^{-1} s_{12} b_2^{-1} s_{12}^{-1} b_1^{-1} s_1^{-1}$, where $s_{12} = s_1^{-1} s_2 = i^{-1} \mathbf{d}_1^{-1} \mathbf{d}_2$. The diagonally partitioned matrix $\mathbf{d}_1^{-1} \mathbf{d}_2$ is responsible for interactions between the layers. Cascading p triple-layer substructures suggests taking the p -th power of the characteristic matrix w_3 . Although the result is straightforward since $(w_3)^p = s_1 (b_1^{-1} s_{12} b_2^{-1} s_{12}^{-1} b_1^{-1})^p s_1^{-1}$, it is clear that new interactions of near-field waves introduced by cascading will change the effective properties of the cascaded structure in comparison to those of the initial three-layer substructure, unless it is possible to write w_3 as $w_3 = s_{\text{eff}} b_{\text{eff}} s_{\text{eff}}^{-1}$, where b_{eff} is a diagonal matrix of effective eigenvalues and s_{eff} a matrix of effective eigenvectors.

§ 3. Combining magnetic resonators with semicontinuous films

The above method built on SHA has been usefully applied to the practical problem of optimizing nanostrip magnetic resonators in combination with homogeneous and later with semicontinuous metal films (Chettiar et al., 2006). A unit cell of the geometry is shown in fig. 14(a) along with the orientation of the incident field. It consists of two main parts, a pair of nanostrips, which act as a magnetic resonator giving negative permeability, and two metal films adding negative permittivity. Note that resonant conditions are required to give negative permeability. In contrast, negative permittivity is given by the bulk metal of the metal films. The silica layer in the left represents the substrate over which the structure is fabricated. The thin layer of silica on the right is to protect the top metal layer from oxidation or other possible degradations.

It is preferable to use noble metals (gold and silver) because of losses. But the metal films create a problem when designing an NIM at the

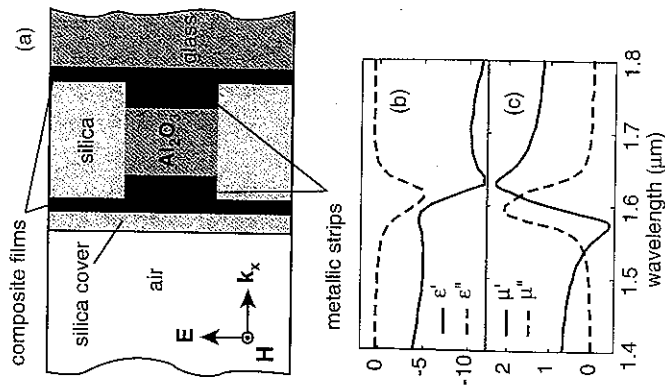


Fig. 14. (a) Schematic representation of the NIM structure. (b) Effective permittivity for a representative structure. (c) Effective permeability.

telecommunication wavelength of 1.5 μm . Alas, at a wavelength of 1.5 μm both gold and silver have highly negative permittivity, while the nanostrip magnetic resonator cannot provide a permeability that is comparably negative. This results in a huge impedance mismatch causing low transmittance. The problem can be circumvented by using a thinner metal film. But the metal film thickness cannot be reduced arbitrarily as the minimum thickness depends critically on the material properties and fabrication limitations.

But instead of using bulk metal, the films can be formed of a mixture of silica and metal. These materials are known as semicontinuous metal films. Such a film can be fabricated using very basic techniques. The permittivity of semicontinuous metal films is described by the effective medium theory (EMT). According to EMT, semicontinuous metal films have a permittivity that is much less negative as compared to bulk metal films. Hence, a semicontinuous metal film is thick enough to be fabricated easily while having a permittivity similar in magnitude as the permeability created by magnetic resonators. This is critical for impedance matching.

The structures were simulated using SHA. Selected results were verified with the results obtained using a commercial FEM solver. The effective material properties were extracted from the simulated reflection and transmission data. Figures 14(b) and 14(c) show the effective permittivity and permeability, respectively, for sample geometry. For this case the semicontinuous metal films had a metal filling fraction of 65% and the metal was silver. We note that for some wavelengths both permittivity and permeability have negative real parts.

Figure 15(a) shows the effective refractive index and we note that the real part of the refractive index is negative between 1500 and 1650 nm. Figure 15(b) shows the transmittance and reflectance spectra. The transmittance spectrum has a maximum value of 27% at 1570 nm. The refractive index has a value of $-1.85 + 0.93i$ at the wavelength of

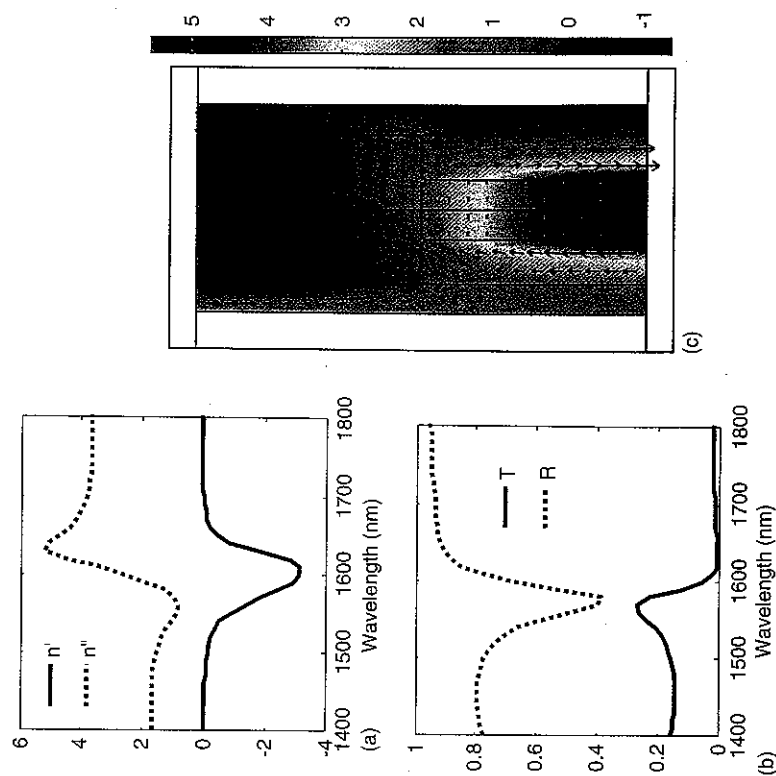


Fig. 15. (a) Effective refractive index for a representative structure. (b) Transmittance and reflectance spectra. (c) Arrows of electric field vectors plotted on top of the gray scale map of the normalized magnetic field.

maximum transmittance. A related structure was simulated in a commercial FEM solver to obtain the field plots, which would provide us with a much better understanding of the nature of the resonance. The structure was simulated at a wavelength of 1570 nm and the resulting quiver plot of the displacement vector field is shown in figure 15(c). For saving time, only one half of the structure was simulated along with proper application of mirror boundary conditions. From the arrow plot it is clear that there is a strong magnetic resonance due to the two coupled silver strips, which is instrumental in providing the negative permeability.

3.1. Sensitivity of the design

The basis structure designed to operate at $1.5\ \mu\text{m}$ is shown in fig. 16(a). The structure is a combination of a periodic metal-dielectric composite (PMD) and a semicontinuous silver film (SSF). The PMD is arranged of infinitely long silver strips separated by an alumina spacer. To prevent aging of the film, an additional layer of silica is added on top of the upper SSF. (The effect of the protective layer has been investigated and appears to be of limited importance.)

The structure demonstrates both negative permittivity (fig. 16(b)) and permeability (fig. 16(c)) about the target wavelength of $1.5\ \mu\text{m}$. To explore sensitivity of the design to geometrical parameters a number of geometrical characteristics have been changed. For example, fig. 16 shows permittivity (fig. 16(b)), permeability (fig. 16(c)), and refractive index (fig. 16(d)) spectra changing according to the variation of the width W of the PMD shown in fig. 16(a). For example, the spectra in figs. 16(b–e) are depicted for the variation of W ranging from 260 to 380 nm with 20 nm stepping. Simulations demonstrate stable negative refractive index near $1.5\ \mu\text{m}$ (illustrated in the inset, fig. 16(e)).

3.2. Conclusion

Recently, metamaterials have been designed that show a negative real part of the refractive index at the telecom wavelength of 1500 nm or 200 THz. Keeping in mind that it took only 5 years to come from 10 GHz to 200 THz, we have no doubt that a negative refractive index metamaterial will be soon available also for the visible range. We have shown in numerical simulations that the following two key remedies are now available to overcome major obstacles that currently limit the

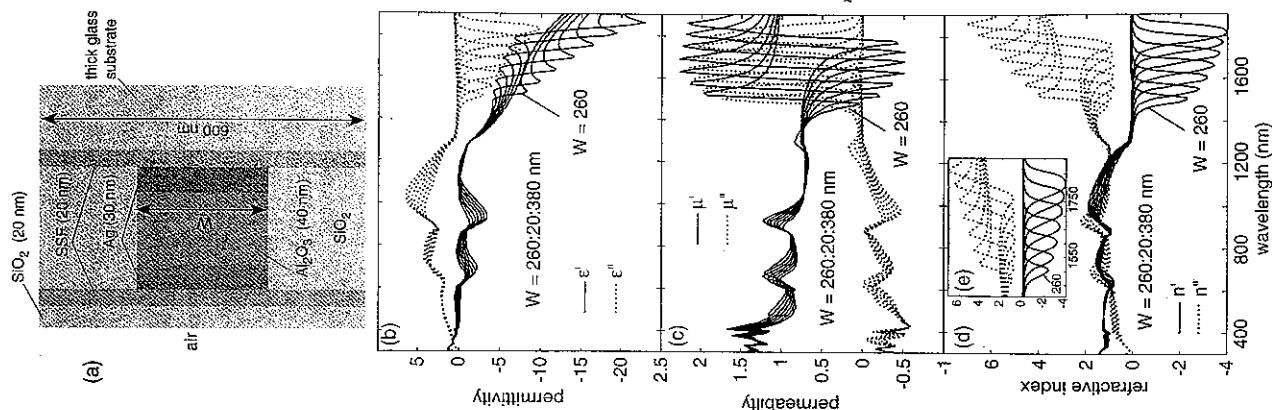


Fig. 16. NIM with semicontinuous films. (a) Basis structure. (b) Permittivity spectra. (c) Permeability spectra. (d) Refractive index spectra for different widths of the silver strips. (e) Zoomed region of negative refraction.

development of optical NIM: (1) impedance-matching designs are capable of suppressing high reflectance, and (2) gain materials embedded in metallic nanostructures can fully compensate for absorptive losses while still retaining the negative refractive index.

The instrumental idea of the new design of fig. 16(a) is based on combining narrow-band resonant PMDs and broadband nonresonant films. While a PMD provides negative effective permeability, an adequate negative effective permittivity is achieved due to nonresonant SSFs.

The approach has been accomplished in the following steps addressing major difficulties of the design process:

- the basis periodic structure (PMD) has electric and magnetic responses at different wavelengths. A remedy is to use different substructures in order to obtain negative magnetic and electric responses at the same wavelength;
- any basis periodic resonant structure exhibits a complementary anti-resonance. While for example, a normal magnetic resonance of μ' shown in fig. 9(b) is paired with the anomalous electric resonance (antiresonance) at the target wavelength of 584 nm, a normal resonance of ϵ' (fig. 13(b)) of an electrically resonant periodic structure of fig. 13a comes with the anomalous magnetic resonance (μ' in fig. 13(b)) at about 1.2 μm . These anti-resonances could decrease (and at times nullify) the effect of magnetic resonance provided that an isolated element of PMD is both magnetically and electrically resonant at the same target wavelength. Therefore, it is essential to use a substructure that has no electrical antiresonance. Silver films are used to add negative permittivity since silver provides negative permittivity naturally (at nonresonant condition);
- the interaction between the silver film and the magnetic substructure gives rise to additional resonances. Therefore, the film is fused with the magnetic substructure to minimize those additional resonances;
- a homogeneous silver film provides an intense negative permittivity and the permeability provided by the magnetic substructure (PMD) cannot match it. As a result, in impedance mismatch causes excess reflection. That is why; semicontinuous metal films with controlled effective permittivity are used instead of continuous metal films.

The design starts with the initial optimization of a given PMD to obtain an optimal negative magnetic response for the structure. Once the negative magnetic response is preliminary optimized, metal films are added to provide an adequate negative electric response. The combined

multilayered structure requires additional optimization due to the interaction of the elementary layers of PMDs with metallic strips.

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