Negative-Index Metamaterials: Going Optical

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(Invited Paper)

***Abstract*—The race toward engineering metamaterials comprising of negative refractive indexes in the optical range started with the realization of negative-index materials for gigahertz frequencies six years ago. Sheer miniaturization of the gigahertz 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. We review these recent results and summarize how to unambiguously retrieve the effective refractive index of thin layers from data accessible to measurements. Numerical simulations show that a composite material comprising of silver strips and a gain-providing material can have a negative refractive index of -1.3 and 100% transmission, simultaneously.**

***Index Terms*—Left-handed materials, metamaterials, nanoparticle plasmon resonance, negative refractive index.**

I. Introduction

Refractive index is the most fundamental parameter to describe the interaction of electromagnetic radiation with matter. It is a complex number $n = n' + in''$, 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 counterintuitive properties. For example, light, which is refracted at an interface between a positive-index material (PIM) and a negative-index material (NIM), is bent in the "wrong" way with respect to the normal. In this case, the group and phase velocities are antiparallel, the wave and Pointing 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 NIMs. Theoretical work on negative phase velocity dates back to Lamb (in hydrodynamics) [1] or Schuster (in optics) [2] and was considered in more detail by Mandel'shtam [3] and Veselago [4]. A historical survey referring to these and other early papers can be found in [5]

In general, left-handed materials do not exist naturally, with some rare exceptions like bismuth in a metallic waveguide that shows $n'<0$ at a wavelength of $\lambda \approx 60\ \mu$m [6]. However, no naturally existing NIM has been discovered for the optical range of frequencies. Therefore, it is necessary to turn to man-made or artificial materials, which are composed in such a way that the averaged (effective) refractive index is less than zero: $n'_{\text{eff}}<0$. One material that can display such properties is the photonic crystal (PC) [7]–[11]. 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, superresolution, which has been predicted by Pendry [12], is not achievable with photonic band-gap materials because their periodicity is in the range of λ. A thin slab of a photonic crystal restores only 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 [13]–[15]. 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 nanocrafted.

A possible—but not the only—approach to achieve a negative refractive index in a passive medium is to design a material where the (isotropic) permittivity $\varepsilon = \varepsilon' + i\varepsilon''$ and the (isotropic) permeability $\mu = \mu' + i\mu''$ obey the inequality

$$\varepsilon'|\mu| + \mu'|\varepsilon| < 0. \tag{1}$$

This leads to a negative real part of the refractive index $n = n' + in'' = \sqrt{\varepsilon\mu}$. [16]. Relation (1) is satisfied, if $\varepsilon'<0$ 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 (1) is fulfilled and which therefore show a negative real part of the refractive index n'.

Relation (1) only holds for passive media. The following more general relationships between the refractive index, the impedance $Z = \sqrt{\mu/\varepsilon}$, and the permittivity and permeability provide the requirements for the NIMs that may also contain gain components:

$$2Re\left(\frac{1}{Z^*}\right)n' = \varepsilon' + \mu'\frac{|\varepsilon|}{|\mu|} \tag{2}$$

$$2n'n'' = \varepsilon'\mu'' + \mu'\varepsilon''. \tag{3}$$

The directions of the Pointing vector and wave vectors are governed by the signs of $Re(1/Z^*)$ and n', respectively [17]. Hence, the sign of $\varepsilon' + \mu'|\varepsilon|/|\mu|$ in (2) should be negative for

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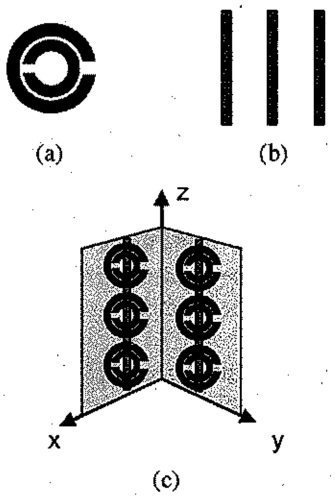


Fig. 1. (a) Magnetically resonant ($\mu' < 0$) metal structure: two counterfacing split rings of subwavelength dimensions (SRR). (b) Electrically resonant ($\varepsilon' < 0$) metallic structure: metal rods. (c) Combination of both structures results in a negative-index metamaterial $n' < 0$.

the NIMs. Equation (3) requires $\varepsilon'\mu'' + \mu'\varepsilon''$ being negative for an absorbing effective medium and positive for a gain NIM.

Until now, we have considered only the isotropic media where ε and μ are complex scalar numbers. It has been shown that in the case of the anisotropic media, where ε and μ are tensors, a negative refractive index is feasible even if the material placed in a waveguide shows no magnetic response ($\mu = 1$). For example, the condition $n'<0$ can be achieved for an uniaxial dielectric constant with $\varepsilon_x = \varepsilon_\perp<0$ and $\varepsilon_y = \varepsilon_z = \varepsilon_P>0$ [6], [18]. We will not focus on the anisotropic media at present despite it being a very promising approach. This is mainly because a negative index for optical frequencies has only been achieved following the approach of magnetically active media.

The paper is organized as follows: In Section II, we recall how to achieve magnetic activity for gigahertz frequencies using metallic split-ring resonators (SRRs) and how the SRRs have been successively scaled down to shift the magnetic resonance up to terahertz 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. This opens the way for new design outlines making active use of localized plasmonic effects, as will be outlined in Section III. Metamaterials containing metal nanostructures as magnetically active components usually show low transmission due to reflection and absorption. In Section IV, we develop an impedance-matched design to suppress reflection. In Section V, we add a gain material to compensate for losses and finally obtain a fully transparent layer of the negative-index metamaterial.

II. DOWNSCALING SRRs

The first recipe to design a magnetically active material was suggested by Pendry in 1999 [19]: Two concentric split rings that face in opposite directions and that 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 that 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 assure that the wavelength of the resonance is larger than the dimensions of the SRR.

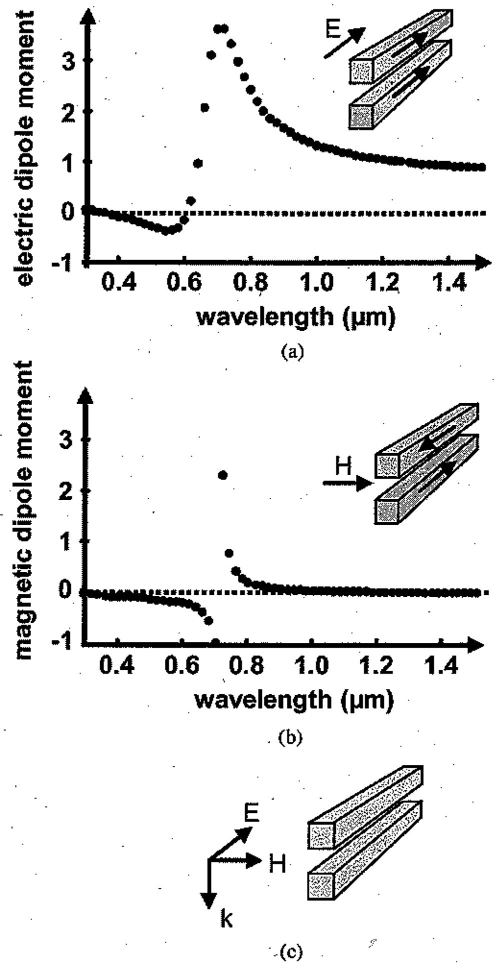
Following this theoretical prediction, Schultz and coworkers combined the SRRs with a material having negative electric response in the 10-GHz range and consisting of metallic wires, in order to reduce the charge carrier density and hence shift the plasmonic response from optical frequencies down to gigahertz frequencies [Fig. 1(b) 20]. The outcome was the first-ever metamaterial with simultaneously negative real parts of permeability and permittivity [21] and consequently with a negative refractive index at approximately 10 GHz [Fig. 1(c)] [22], [23]. This time onward, the race to push left handedness to higher frequencies was open. The gigahertz-resonant SRRs had a diameter of several millimeters, but size reduction led to a higher frequency response. The resonance frequency has been pushed up to 1 THz using this scaling technique [24], [25].

An alternative to double the SRRs is to fabricate only one SRR facing a metallic mirror and use its mirror image as the second SRR [26]. Using that technique, the resonance frequency has been shifted to 50 THz. In order to increase the frequency even more, simply downscaling the geometrical dimensions with wavelength becomes questionable because localized plasmonic effects must be considered. However, localized plasmons open a wide field of new design opportunities. For example, a double C-shaped SRR is not required any more. Originally, the double C-shaped structure was necessary in order to shift the resonance frequency to sufficiently low frequencies so that the requirement of subwavelength dimension could be fulfilled. In the optical range, however, localized plasmons help to shift resonance frequencies to lower energies and consequently, the doubling of the split ring is not necessary [27]. The first experimental proof that single SRRs show an electric response at 3.5 μm (85 THz) was provided in 2004 by Linden and coworkers [28] and it was concluded that the magnetic response of single SRRs should be found at the same frequency. Meanwhile, the electric resonance frequencies of single SRRs has even been pushed to the important telecom wavelength of 1.5 μm [29], [30]. Other approaches to creating metamaterials with magnetic activity that make use of localized plasmonic resonances and abandon the classical SRR shape completely, will be discussed in Section III.

III. METAMATERIALS USING LOCALIZED PLASMONIC RESONANCES

A. Metal Nanorods

It was mentioned by Lagarkov and Sarychev [31] that a pair of noble metal nanorods can show a large paramagnetic response, and it was first pointed out by Podolskiy *et al.* [32] that such a pair of noble metal nanorods is also capable of a *diamagnetic response* at 1500 nm. In this publication, it was predicted for



(a)

(b)

(c)

Fig. 2. Response of a pair of gold nanorods to radiation, simulated with coupled dipole approximation technique. (a) Electrical dipole moment, the 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) Pair of rods illuminated from above with TM polarization. Pair of rods will have a double negative response to the field.

the first time that materials containing such pairs of rods can show a negative n' even at wavelengths in the visible region. The issue has been discussed in more detail by Panina *et al.* [27] and also by Podolskiy *et al.* [33], [34]. Fig. 2 depicts this by showing a pair of nanorods that show a negative response to an electromagnetic plane wave. The two gold rods are separated by a distance far less than the wavelength. The diameter of the cross section of the rods is also much less than the wavelength and the length of the rods may be, but need not be, in the range of half a wavelength. An ac electric field parallel to both the rods induces parallel currents in both the 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 electrodynamically coupled rods. Fig. 2(a) shows the induced electric dipole moment for the specific dimensions reported in [34]: 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 that will 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 electrodynamically coupled rods, as shown in Fig. 2(b) [34]. In terms of the 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) [33].

So far, the electromagnetic response has been discussed in terms of coupled plasmonic resonances. An alternative way of looking at it is by considering 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 [31], [35].

It is important that both the resonances, the dipolar electric, and the dipolar magnetic resonance are at similar wavelengths. This requires that the coupling between the two rods should not be too strong, otherwise, the two resonances would be split further apart. It is seen in Fig. 2(a) and (b) that there is a certain range of wavelength (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) [transverse magnetic (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 telecommunication wavelength of 1500 nm) was reported for the first time in [36]. 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 dimensions than the lower rod. A scanning electron microscope (SEM) image of a single pair and its dimensions are shown in Fig. 3(a). Pairs of nanorods are periodically repeated as depicted in Fig. 3(b) and shown by an SEM image in Fig. 3(c). Fig. 3(d) shows the unit cell of the periodic arrangement and gives more dimensions. A full description of the sample and its preparation, is given in [37] and [38]. Reflections spectra of a sample containing pairs of gold nanoparticles have been explained assuming a negative permeability in [39]. Although a negative refractive index was not claimed in that work, in our opinion [40], even a negative permeability in the visible range cannot be demonstrated with the sample proposed in [40].

Fig. 4 shows the results obtained in [36] 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 time-domain (FDTD) method. It is clearly seen that the real part of the refractive index becomes negative in the wavelength

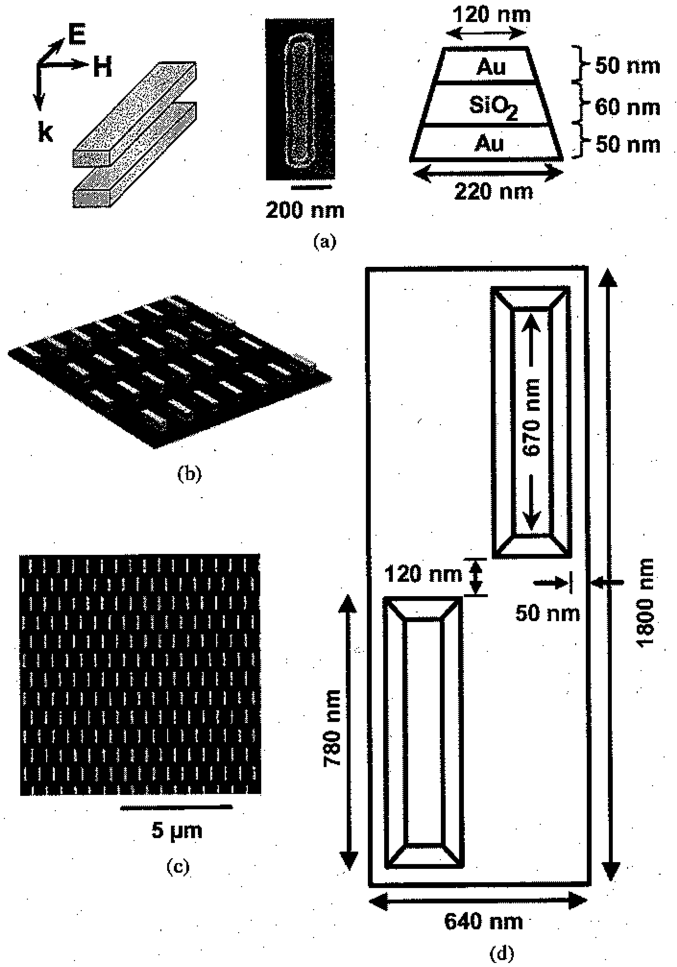


Fig. 3. (a) *Left to right*: Scheme of nanorod pair and proper light polarization for negative index, SEM image, dimensions. (b) Scheme of the arrangement of nanorod pairs. (c) SEM image of arranged nanorod pairs. (d) Dimensions of the arrangement (one unit cell is shown).

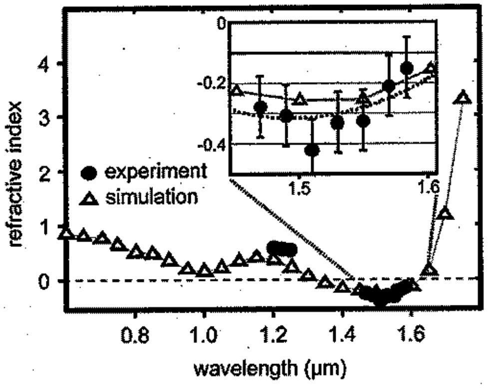


Fig. 4. Real part of the refractive index of a layer of nanorod pairs, as shown in Fig. 3. Data that are restored from experimentally determined transmission, reflection, and phase measurements (*full circles*). FDTD simulation (*open triangles*). Zoom of the region of negative refraction (*inset*). *Dashed line* is a least-square fit to the experimental data. Refractive index of $n' = -0.3 \pm 0.1$ was determined.

range from approximately 1400 to 1600 nm, which includes the important telecommunication band at 1500 nm. The inset gives a closer look at that frequency range. The experimental data proving that $n' = -0.3 \pm 0.1$ was obtained [36].

It turns out to be nontrivial 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 the air (top) and a glass substrate (bottom) can be determined uniquely as discussed in [38] using transfer matrices

$$n = \frac{1}{k\Delta} \arccos \frac{1 - r^2 + n_s t^2}{[1 + n_s - (1 - n_s) r] t}, \tag{4}$$

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

$$t = \sqrt{T}\, e^{i\tau}, \quad r = \sqrt{R}\, e^{i\rho}. \tag{5}$$

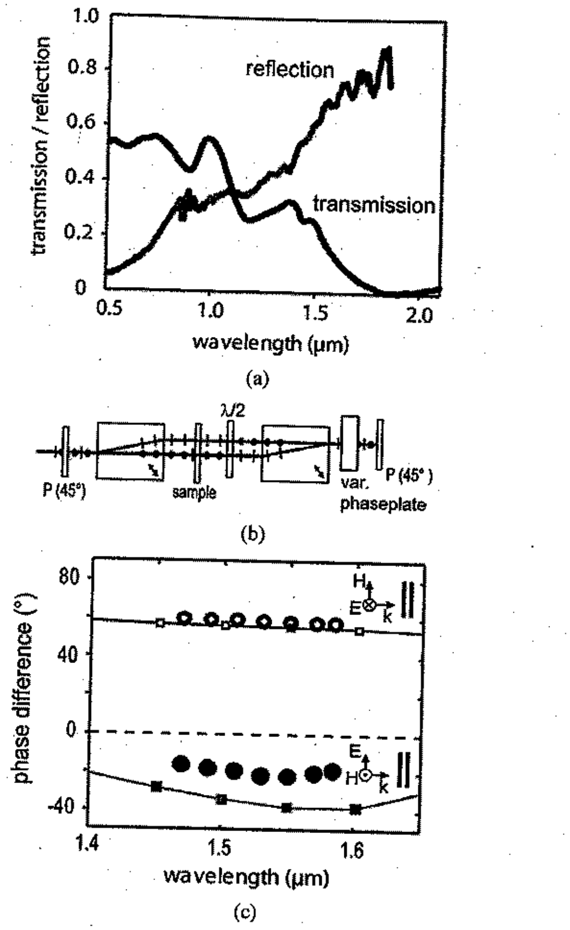
The importance of the phase measurements in addition to T and R was emphasized in [37] where it was shown that two similar, but not identical, samples of pair of nanorods can show the same values T and R (within experimental error) but greatly differ in τ. Consequently, one sample was found to have negative n', while for the other one, n' is positive.

Fig. 5(a) shows the transmission and reflection spectra of the negative-index metamaterial of Fig. 3. The absolute phase shifts were measured with a "walk off" interferometer. The beam of a tunable semiconductor laser was split into two orthogonally polarized beams. One beam passed through the negative-index metamaterial of thickness Δ while the other beam was used as a reference and passed only through the glass substrate at a spot not covered by the metamaterial [37], [Fig. 5(b)]. The beams were recombined behind the glass substrate. The measured phase difference $\tau - k\Delta$ between the beam passing through the thin film and the reference beam propagating only through air of the same thickness Δ allows to determine τ using interferometry [Fig. 5(c)]. The phase shifts in reflection (ρ) were obtained for both polarizations in a similar way.

In [36], it was found that the phase τ is delayed in the metamaterial by approximately 60° compared to air in the case of the transverse electric (TE) polarization (electric field perpendicular to the plane of the rods). In contrast, τ is advanced by approximately 20° in the case of the TM polarization, as shown in Fig. 5(c) [36]. The advancement of τ for the TM polarization can be used as 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 (3) [38].

Nevertheless, one can use pure phase measurements to make an estimate for n', as was pointed out in [38]. In the case of low reflection ($R \ll 1$)

$$n' \approx \frac{\tau}{k\Delta} \tag{6}$$

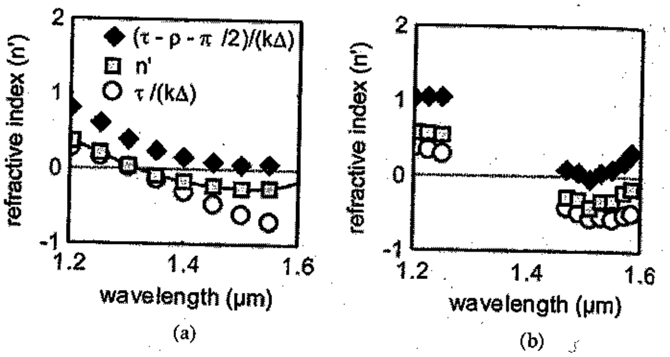


(a)

(b)

(c)

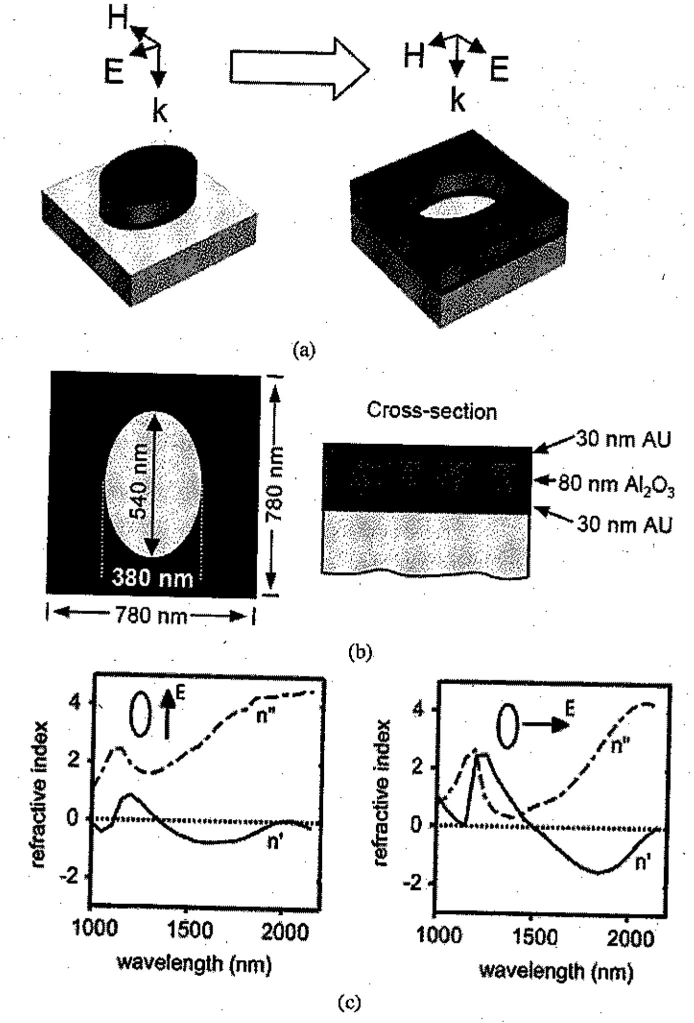
Fig. 5. (a) Measured transmission and reflection spectra of the sample shown in Fig. 3. (b) Setup for phase measurements. (c) Phase difference τ for two polarizations. *Circles* are measured values, *squares* and *lines* are from simulation. The light is delayed in the case of the TE polarization (H-field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



(a)

(b)

Fig. 6. Real part of the refractive index as determined by the exact formula (2) (*squares*) or by phase only assumptions according to (5) (*full diamonds*) or (4) (*open circles*). (a) Numerical simulations. (b) Experimental results.



(a)

(b)

(c)

Fig. 7. (a) Nanoellipse consisting of two 30-nm thick ellipses of gold separated by 80 nm of Al_2O_3 (*left*). Elementary cell of coupled elliptic voids (*right*). (b) Dimensions of the voids. The voids are repeated periodically in two dimensions. (c) Refractive index $n = n' + in''$ for light polarized parallel (*left*) or perpendicular (*right*) to the long axis of the voids, as obtained from FDTD simulations.

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

$$n' \approx \frac{\tau - \rho - \frac{\pi}{2}}{k\Delta} \tag{7}$$

holds. These two formulae indeed give an upper and lower bound to the correct value of n' according to (2) and (3) (Fig. 6) [38].

B. Voids

An interesting approach to negative-index metamaterials is to take the inverse of a resonant structure [41], e.g., a pair of voids as the inverse of a pair of nanorods [42]–[44]. 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 were performed to determine the refractive index of void metamaterials [38]. The dimensions were chosen according to Fig. 7(b) in the simulations in order to match the dimensions of the experimental sample reported in [44].

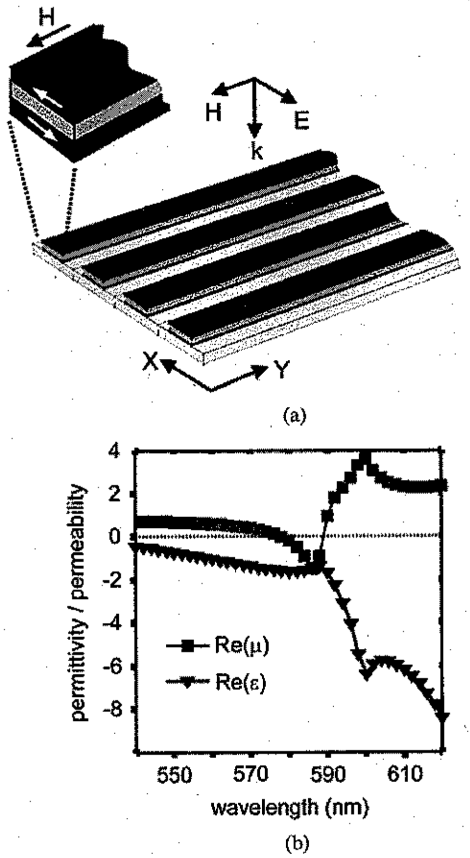
These simulations were carried out for both the cases of polarization: the electric field oriented along the long axis of the elliptical voids and perpendicular to it. It is seen that n' becomes negative in both cases. However, the effect is more pronounced if the electric field is oriented along the 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 earlier, where the imaginary part of the refractive index was 3 [36]. 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 [42]. The imaginary part n'' is large in that case. However, it has been shown that further optimization can reduce n'' substantially [44].

IV. 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 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 Section V. In the present section, we will concentrate on the second issue, which is impedance matching. The impedance is given by $Z^2 = (Z' + iZ'')^2 = \mu\varepsilon^{-1}$ and it is required that the impedances match at a boundary between the two media in order to eliminate reflection at normal incidence. This condition is well known for microwaves and replaces Brewster's law for optical frequencies if $\mu \neq 1$ [27]. The impedance is matched at a boundary between a negative-index metamaterial and air if $Z' \rightarrow 1$, and in the metamaterial, if $Z'' \rightarrow 0$.

In Fig. 8(a), we introduce a metamaterial where the conditions $Z \rightarrow 1 + 0i, n' < -1$, and $n'' < 1$ hold simultaneously for a wavelength in the visible region. The structure consists of pairs of coupled silver strips. Both strips are 280-nm wide (x-direction), 16-nm thick, and they are infinitely long in the y-direction. The two silver strips are separated in the z-direction by a 65-nm thick layer of Al_2O_3. The pairs of strips are periodically repeated in the x-direction with a period of 500 nm. We assume the presence of 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 the z-direction). The electric field is polarized in the x-direction. The magnetic field, which is parallel to the strips, induces antiparallel currents in the two silver strips, as indicated in the magnified inset of Fig. 8(a) by the two white arrows. This leads to a magnetic response of the structure. We use FEMFD calculations to determine the spectra of the electrodynamic constants. Fig. 8(b) shows the real parts of the permittivity (triangles) and of 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., $Z' > 0.5$ and eventually reaches 1 at 586 nm, and simultaneously $|Z''| < 0.5$ in the range 570–585 nm [Fig. 9(c)]. In total, this leads to a reflectance of less than 10% at 584 nm.



(a)

(b)

Fig. 8. (a) Double silver strips, separated by Al_2O_3. The strips are infinitely long in the y-direction and are periodically repeated in the x-direction. The H field is oriented in the y-direction. Currents in both the strips are antiparallel (*white arrows* in the magnified inset) if the H-field is polarized in the y-direction. (b) Real parts of the permittivity and permeability as simulated with FEMFD.

The absorption seems to have a local maximum at 586 nm. However, it does not reproduce in the spectrum of n''. This is mainly because the reflection at the interface between the 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. Even so, it accounts for almost 90% of the losses in the range of the "reflectance window" at 584 nm. In summary, the present section shows that a metamaterial consisting of pairs of silver strips, as depicted in Fig. 8(a), can form an almost impedance-matched NIM for visible light. The transmission is limited to 50% almost solely due to absorption, while reflection losses play a minor role.

V. GAIN, COMPENSATING FOR LOSSES

It has recently been pointed out that energy can be transferred from gain material to surface plasmon polaritons [45]–[50] or

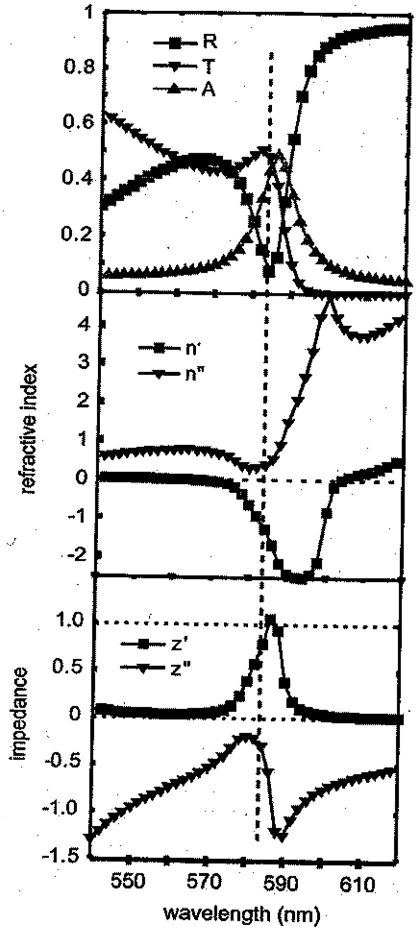


Fig. 9. Spectra of several optical constants of the structure shown in Fig. 8. (*Upper panel*) reflection R, transmission T, and absorption A spectra. (*Middle panel*) real and imaginary parts of the refractive index. (*Lower panel*) real and imaginary parts of the impedance. The *vertical dashed line* at 584 nm indicates the spectral region where the reflection is minimal, the transmission is high, the imaginary part of the refractive index is only 0.4 while the real part is negative and the real part of the impedance is close to 1, indicating impedance matching to air. The spectra were determined using FEMFD simulations.

to plasmons in metal nanostructures [51], [52] using stimulated emission. Specifically, continuous thin films of metal were used to confine lasing modes in quantum cascade lasers to the gain region and also to guide the lasing modes by surface plasmon modes [45], [46]. Ramakrishna and Pendry suggested that gain materials such as semiconductor laser materials be stapled between the negative index (or metal) layers of stacked near-field lenses [53] in order to remove absorption and improve resolution. The requirement of a perfect near-field lens, where thin layers of PIMs and NIMs are alternated is that $\varepsilon_{\mathrm{P}} = -\varepsilon_{\mathrm{N}}$ and simultaneously $\mu_{\mathrm{P}} = -\mu_{\mathrm{N}}$, where the subscripts denote material constants of positive (P) and negative (N) materials. This requirement naturally includes the conditions $\varepsilon''_P = -\varepsilon''_N$ and $\mu''_P = -\mu''_N$, i.e., the positive layers must provide gain in order to optimize the lens [53].

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: $n'' = (\lambda/4\pi)(\alpha - g)$.

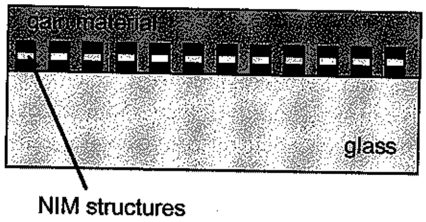


Fig. 10. NIM (e.g., double rods), filled with a gain medium, e.g., a solid solution of dye molecules in a matrix.

Further, instead of alternating the layers of NIMs and PIMs, we propose to "submerge" the negative-index structures (containing, e.g., metal nanorods) in the gain media, as shown in Fig. 10. For example, this could be achieved by spin coating a solution of laser dye molecules or π-conjugated molecules on top of the negative-index structures. Applying semiconductor nanocrystals could be an alternative approach.

One might question whether the metal nanostructures nullify any attempt to amplify the electromagnetic fields using gain materials in their close vicinity because gold nanoparticles are well known to quench fluorescence in an extremely efficient manner [54], [55]. In contrast, however, working solid state and organic 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 was 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 metalfree DFB gratings) to be operative [56]. In the case of infrared quantum cascade lasers (QCL), a wave guiding metallic layer was shown to be beneficial for the laser power output [46]. 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 negative-index metamaterials.

We would like 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. 10). 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 an absorption of approximately 45% [Fig. 9(a)]. Applying Beer–Lambert'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 $g = 3 \times 10^4\ \mathrm{cm}^{-1}$ 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 $\sigma_{\text{SE}} = 3 \times 10^{-16}$ cm^2 [57] and therefore the concentration of excited dye molecules should be 170 mM. Alternatively, semiconductor nanocrystals (NC) such as CdSe could be applied. It has been shown in [58] 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 the densely packed nanocrystal films can show gain of the order of $g \approx 10^5$ 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 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 been pointed out by Kim *et al.* [59] and by Lawandy [52] that a gain medium and localized plasmonic resonances 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 pair of strips, as in Fig. 8, shows a much larger response to an incoming electric field as the same metal structure without gain.

In the example considered earlier, 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 following model [Fig. 11(a)]: We took the same structure as shown in Fig. 8, but now we filled the gaps between the double silver strips with a material that provides a fixed amount of gain between 0 and 15×10^3 cm^{-1}. Fig. 11(b)–(d) shows the transmittance (T), reflectance (R), refractive index (n' and n''), and impedance (Z' and Z'') as a function of gain (g). We found that with a gain of 12×10^3cm^{-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)]. Further, the impedance which was already matched quite well without the gain medium (Fig. 9) improves further when gain is applied, i.e., $Z' \approx 1$ and $Z'' \approx 0$ for $g = 12 \times 10^{-3}$cm^{-1} [Fig. 11(d)]. The exact results for a gain of $g = 12 \times 10^{-3}$cm^{-1} are $n' = -1.355, n'' = -0.008, Z' = 0.89, Z'' = 0.05, T = 100.5\%$, and $R = 1.6\%$.

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 [52]. 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 [52]. This will allow the design of NIMs of less overall metal content. The density of pairs of rods may be reduced, 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 given elsewhere. We also did not include dispersion of the gain material because we performed our preliminary FEMFD studies at a single wavelength (584 nm) only. The effects of dispersion of gain materials, especially the fact that each gain material necessarily introduces increased losses at the excitation band, will also be addressed elsewhere.

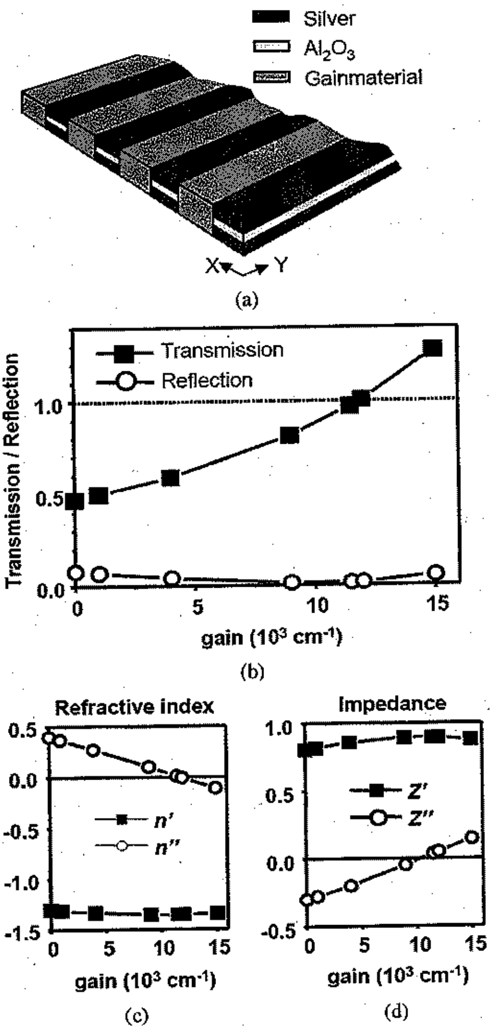


Fig. 11. (a) Same sample as in Fig. 8(a), but with gain-providing material between the double silver strips. Air is assumed above and below the layer, and the layer is irradiated with a plain wave (584 nm) from above, H-field polarized along the y-direction. (b) Transmission and reflection as a function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

VI. CONCLUSION

Very recently, metamaterials have been designed that show a negative real part of the refractive index at the telecommunication wavelength of 1500 nm or 200 THz. Keeping in mind that it only took five years to come from 10 GHz up to 200 THz, we have no doubt that a negative refractive index metamaterial will soon be available also for the visible range. We have shown in numerical simulations that two key remedies are now available to overcome major obstacles that currently limit the development of optical NIMs: 1) impedance matching designs are capable enough to suppress high reflectance and 2) gain materials embedded in metallic nanostructures can fully compensate for absorptive losses while still retaining the negative refractive index.

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