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Empowering Metamaterials with Gain and Nonlinearities

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Abstract: Calculations show that matched impedance and compensated losses due to optimized design and gain material, respectively, can lead to 100% transmission. Extraordinary nonlinear-optical properties originating from contra-directed wave and Poynting vectors are discussed.

The refractive index ($n = n' + in''$) is the key parameter in the interaction of light with matter. While n' has generally been considered to be positive, the condition $n' < 0$ does not violate any fundamental physical law, and materials with negative index have some remarkable properties. For example, vectors $\vec{E}$, $\vec{H}$, and $\vec{k}$ form a left-handed system and such materials are synonymously called "left handed" or negative-index materials (NIMs). No naturally existing NIM is known so far in the optical range and it is necessary to create artificial materials (metamaterials) in which the effective refractive index (n'_{eff}) is negative. A truly negative index $n'_{eff} < 0$ can only be achieved in metamaterials with structural dimensions far below the wavelength; for optical wavelengths such materials must be nano-crafted. A possible approach to create a NIM is to design a material where the effective isotropic properties (permittivity $e = e' + ie''$ and permeability $m = m' + im''$) obey the equation $e'|m| + m'|e| < 0$, which is always satisfied, if $\varepsilon' < 0$ and $\mu' < 0$.

The GHz resonant SRRs first demonstrated by Smith, Schults et al [1] had a diameter of several millimeters, but size reduction leads to a higher frequency response. The resonance frequency has been pushed up to 1THz using this scaling technique [2]. An alternative to double SRRs is a single SRR facing a metallic mirror, where the resonance frequency was shifted to 50 THz [3]. But further downscaling becomes uncertain due to localized plasmonic effects, which nonetheless open yet another design opportunities. Thus, a double SRR is not required any more; The first experimental proof that single SRRs show an electric response at ~100 THz was provided in 2004 by Linden and coworkers [4]. The resonance of U-shaped structures has even been pushed to the important telecom wavelength of 1.5 µm [5]. The experiments above were critical for the field; still no negative refraction was accomplished because those structures were missing components providing both negative epsilon and negative μ.

The first experimental demonstrations of negative refraction were accomplished for pairs of metal rods [6,7] and for the inverted system of pairs of dielectric voids in metal [8]. In [6] it was predicted, that materials containing pairs of coupled metal rods can show a negative n' even for visible wavelengths. Two gold rods are separated by a distance far less than the wavelength. An AC electric field parallel to both rods induces parallel currents in both rods. The magnetic field, which is oriented perpendicular to the plane of the rods, causes anti-parallel currents in the two rods. This can be considered as a dipolar magnetic mode. Above, the electromagnetic response has been discussed in terms of coupled plasmonic resonances. An alternative way of looking at it is that the anti-parallel currents in the rods and the displacement currents at the ends of the two rods form inductors, while the gaps at the ends form two capacitors. The result is a resonant LC-circuit with a current loop [6].

The Purdue team observed a negative refractive index $n' = -0.3 \pm 0.1$ at the telecommunication wavelength in their ebeam fabricated sample of paired rods [6]. We note that the imaginary part of the refractive index n'' was significantly larger than the real part $|n'|$ so that the figure of merit $F = |n'| / n''$ was low in this first experiment. The paired pillars design used in [7] is essentially equivalent to the one described above, with the magnetic and electric responses related to the symmetric and asymmetric currents in the pillar pairs, respectively. This group was first to report the optical magnetism and negative refraction in the visible range [7].

An interesting approach to NIMs is to use the inverse of a resonant structure [8], e.g. a pair of voids as the inverse of a pair of nanorods [6]. Instead of a pair of metal nano-ellipses separated by an oxide, which are similar to the pair of rods, two thin films of metal are separated by an oxide and mounted on a glass substrate.

Then, elliptically shaped voids are etched in the films, thus forming the inverse of the original paired metal ellipses structure. Both samples, in accord with the Babinet principle, should have similar resonance behavior if the orientation of the electric and magnetic fields are also interchanged. Using this approach, Zhang et al. have obtained negative refractive index in the range $\lambda = 1.8$ μm, with a relatively large figure of merit reaching $F = 2$ [8].

A negative refractive index with the largest F in the optical range was achieved in [9] for the fishnet structure. The self-supporting fishnet structure can be thought of as pairs of metal strips, providing a negative permeability, and metal wires giving a negative permittivity; alternatively it could be also viewed as pairs of rectangular dielectric voids in parallel metal films. This group observed a large negative refractive index, reaching $n' = -2$ at $\lambda = 1.5$μm, and an impressive figure of merit reaching $F = 3$ at $\lambda = 1.4$μm. By using the pulsed interferometry, they were also able to measure both the phase and group velocities of light in their structure [9].

All NIMs using plasmon resonant metallic elements have two distinct problems: high reflection and absorptive losses, both reducing the overall transmission through the metamaterial. The reflection can be suppressed by an optimized design with matched impedance. We show an example of optimized NIM where the conditions Z ® $1 + 0 \times i$, n¢< -1, and $|n$¢¢$| < 1$ hold simultaneously for a visible wavelength [10]. The NIM is arranged of coupled silver strips separated by a dielectric spacer. For this NIM, the simulated transmission has a local maximum of 51% at 582 nm. The impedance is matched quite well from 582 to 589 nm, i.e. $Z' > 0.5$ and reaching 1 at 586 nm with $|Z''| < 0.5$ in the range 570 - 585 nm. We have shown that a given design can be optimized to an almost impedance matched NIM for visible light. The transmission is limited to 50% almost solely due to absorption.

These absorptive losses (in terms of a large n¢¢) are the second major difficulty, since the ohmic losses are generally large due to localized plasmon resonances. To overcome this difficulty we propose supplying energy from gain material in NIMs using stimulated emission [10]. We simulated the same impedance-matched structure, but now we embedded a material that provides a fixed amount of gain between 0 and $15 \cdot 10^3$ cm^{-1}. We found that at a gain of $12 \cdot 10^3$ cm^{-1} the structure becomes transparent, while the real part of the refractive index n¢ is almost unaffected by the gain material. Moreover, the impedance which has already been matched quite well without the gain medium improves further when gain is applied, i.e. Z¢» 1 and Z¢¢» 0 for $g = 12 \times 10^{-3}$cm^{-1}.

Extraordinary nonlinear-optical properties originating from contra-directed wave and Poynting vectors are investigated. The feasibility of light-controlled transparency, cavityless oscillation and generation of counter-propagating entangled right- and left-handed photons is shown [11].

In summary, we have shown that two key remedies are now available to overcome major obstacles that currently limit the development of optical negative-index materials (1) impedance matching designs are capable to suppress high reflectance, and (2) gain materials embedded in metallic nanostructures can compensate for absorptive losses while still retaining the negative refractive index. Novel nonlinear optical properties in NIMs are also investigated.

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