Active Nanoplasmonic Metamaterials: A Route to Perfect Lenses, Stopped Light and Nanolasers

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Introduction

Up until the discovery of surface enhanced Raman scattering in 1974 [1] , metals were seen to be optically uninteresting [2]. However, in recent years, interest has grown significantly in the interaction of metallic nanoparticles with light due to the diverse range of interesting effects observed. This renewed interest, coupled with the advent of modern fabrication techniques [3] and a drive to integrate photonics with nanoelectronics [4] has led to metal optics becoming one of the most actively researched areas of nanoscience [5]. One particular application borne out of this resurgence is that of metamaterials, artificial materials carefully structured in order to engineer physical properties not observed in nature. The possibility of engineering a negative refractive index in particular gives rise to a number of interesting effects, including optical cloaking, trapped light and the potential to construct a 'perfect', diffraction limit breaking lens [6]. However, these novel materials suffer greatly from large ohmic losses in the visible light regime, making it necessary to search for strategies to compensate before their true potential for numerous applications can be reached. In this essay we present one such strategy, that of employing active (gain-enhanced) nanoplasmonic metamaterials, first realised experimentally just a few years ago [7]. We will give a brief overview of the theoretical background of negative refractive index materials and problems associated with their operation in the optical regime, followed by an outline of the working principles of active metamaterials. Key advances in the area are highlighted, with a particular emphasis on the potential impact and varied applications that could be made possible by this important advance.

What are metamaterials?

Metamaterials are materials that exhibit properties that are not seen (or have not been seen yet) in naturally occurring materials. One particular class is negative refractive index materials. These were originally proposed by Victor Veselago almost half a century ago[8]. Before Veselago's paper refractive indices were assumed to always be positive. John Pendry was the first to propose a practical way to create a negative refractive index material in his paper "Negative refraction makes a perfect lens", published in 2000 [9]. He suggested that a simple negative index material could be constructed from an array of metal wires and split rings, and that such a structure/material could be used as a perfect lens.

The refractive index of a material is defined as $n = \sqrt{\epsilon\mu}$ where n is the refractive index and ε and μ are the dielectric permittivity and magnetic permeability respectively. If it were possible to engineer structures so that they had both negative permittivity and permeability, then those materials would exhibit negative refractive indices. Metals are typically used because at certain frequencies they naturally have a negative dielectric permittivity [10]. Free electrons at the surface of a metal behave like a plasma in the presence of an electromagnetic field. The permittivity of an ideal plasma is given by $\epsilon = 1 - \frac{{\omega_p}^2}{\omega^2}$, so that metals with a plasma frequency greater than the frequency of the electromagnetic filed will have negative permittivity. In Pendry's simple system, the metal rods naturally contribute the required negative permittivity, while the negative permeability arises from magnetic dipole resonance in the loops, forming a structure with an overall negative refractive index.

However the use of metallic elements in these systems is not without issues. Metals have a finite skin depth that characterises the distance electromagnetic waves can penetrate as a function of frequency. In the optical regime the system size is usually very small. It is possible for the system dimensions to be smaller than the skin depth, leading to high losses [11]. Losses arise due to scattering of the accelerated electrons, either from other electrons, from phonons or from lattice defects and are particularly significant in the optical regime. These losses limit the potential uses of negative refractive index materials.

Strategies to compensate the loss in these systems are needed. One obvious method to mitigate this is to optimise the system geometry by adjusting the size and shape of the components [12]. However this approach is limited because losses can only be minimised and not completely eliminated, since there are still unavoidable intrinsic losses in the metals. Another proposed strategy which has been fairly successful is to use all dielectric components [13-15]. This directly eliminates the high ohmic losses associated with metals at optical frequencies, a phenomenon associated with free charges not present in dielectric materials. Another promising strategy that has recently emerged [16-18] is the use of a gain medium to compensate for losses over a specific frequency range. This method is of particular interest since the incorporation of plasmonic materials has the potential to compensate for losses while also creating inherent feedback, which could allow for the fabrication of nanoplasmonic lasers.

Active nanoplasmonic metamaterials

In 2010, S. Wuernster et al. [17] presented their mathematical model for a double fishnet structured metamaterial with a gain medium (Rhodamine-800 dye) incorporated into its structure. The probe wavelength used was 710nm and their key result is illustrated in figure 1. Note that when the gain medium is included this results in a decrease in the imaginary part of the refractive index (and therefore the absorption coefficient) around the probe wavelength. This is indicates that the losses at this wavelength have been reduced, an effect which scales with increasing pump intensity (gain). However, this effect is only present in the region of interest; which here is ≈706-718nm, where a negative refractive index is observed. The losses are not eliminated, but rather are relocated spectrally to a region where the real part of the refractive index is positive (e.g. >720nm). This is a consequence of the Kramers-Kronig relations relating the real and imaginary part of the refractive indices. Thus the losses can be reduced at certain wavelengths only, and are simply 'passed on' to another region of the spectrum. The bandwidth over which a negative index material with compensated losses is realisable is therefore limited. However, this theoretical result does demonstrate a complete compensation of loss in a region ≈706-714nm (where both the real and imaginary parts of the refractive index are negative) for a pump pulse of 2kV/cm, which is a promising development.

The first experimental demonstration of this result was published by S. Xiao et al. later that year. Using the same structure and gain medium, they were able to demonstrate a compensation of losses and light amplification in the range 715-735nm. The loss compensation mechanism here is relatively straightforward: dye molecules are excited by a sufficiently powerful pump pulse, a population inversion is formed inside them. A probe pulse is chosen such that its wavelength coincides with the stimulated emission wavelength of the dye molecules and is amplified sufficiently to compensate for losses. It should be noted that both the papers discussed above used dye molecules as their gain medium. A more desirable approach would be to use a solid-state medium in order to enable optical pumping and increase potential for large-scale production [2]. Although full-loss compensation has been presented as being theoretically possible [19] there has not been an unambiguous experimental demonstration that this is the case. Ultimately, the success of active nanoplasmonic metamaterials may rely on finding better plasmonic materials [20]. Despite this, other novel ideas (such as surface plasmon lasers, known as spasers) related to gain-enhanced metamaterials have experienced an enormous growth of ideas recently due to these developments. Additionally, should full-loss compensation be successfully demonstrated in the future, it could have a profound impact on a number of areas including optical computing and even the biomedical sciences due to the varied prospective applications of these materials.

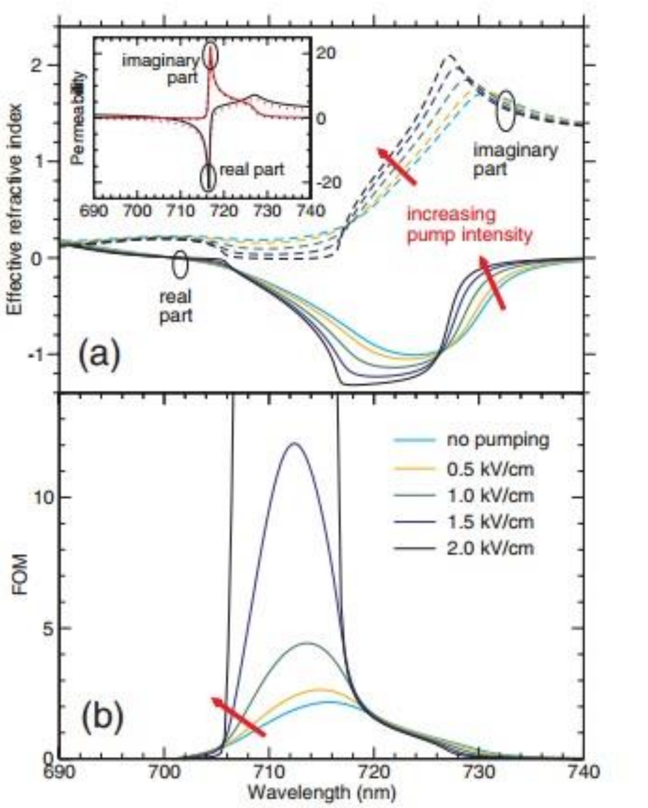

Figure 1 From S. Wuernster et al. [17]. a) shows the real and imaginary parts of the refractive index for different pump intensities as a function of wavelength. b) shows the figure of merit (FOM) versus the wavelength, where the FOM is a ratio of the real and imaginary parts of the refractive index. Note that in a region ≈706-714nm for high pump intensity, both the real and imaginary parts of the refractive index are negative, indicating loss compensation.

Stopped light and 'trapped rainbows'

One particularly interesting phenomenon achievable using negative index materials is stopped or slowed light, otherwise known as the 'trapped rainbow' effect [6]. This effect is achieved by propagating a guided electromagnetic wave through an ordinary waveguide into a tapered, negative refractive index guide surrounded by ordinary dielectric cladding. Inside both waveguides, the wave undergoes successive Goos-Hänchen shifts, an optical phenomenon whereby light undergoes a small lateral shift when it is totally internally reflected at the boundary with the positive index cladding [21]. For a positive waveguide, this shift occurs along the direction of propagation, but for a negative refractive index waveguide this shift is reversed, such that it opposes the direction of propagation [22]. In this way, the guided waves are slowed down inside the metamaterial. Additionally, due to the anomalous frequency dispersion of the metamaterial, different frequency components will be stopped at different critical thicknesses of the metamaterial layer. For example, the longest (red) wavelength components will be stopped inside the thinnest part of the waveguide, while the shortest (blue) wavelength components are stopped at the thicker end. As such, the use of a tapered, negative refractive index waveguide allows both the trapping of light inside the material, as well as its spatial separation into different frequency components (colour constituents) forming a trapped rainbow. Potential applications of this work include data storage, raising the possibility of

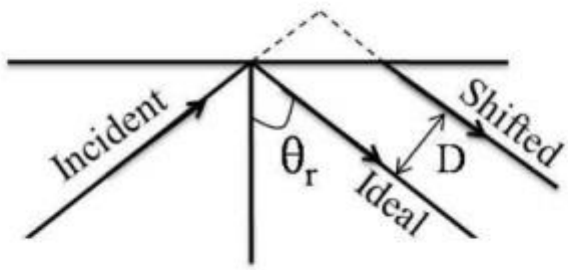

Figure 2 Taken from [21], an illustration of the Goos-Hänchen shift as it occurs in a positive refractive index material. In the case of a negative index material, the direction of this shift is reversed [22].

realising quantum optical memories or an all-optical network router [23] to increase the operating speed of optical fibre networks. While the stopping of light via other approaches had already been experimentally demonstrated prior to the publication of the first paper proposing this method [6], these methods had significant limitations such as requiring the use of ultra-hot or ultra-cold gases [24] or not preventing pulses from travelling down the fibre with a high enough efficiency [25].

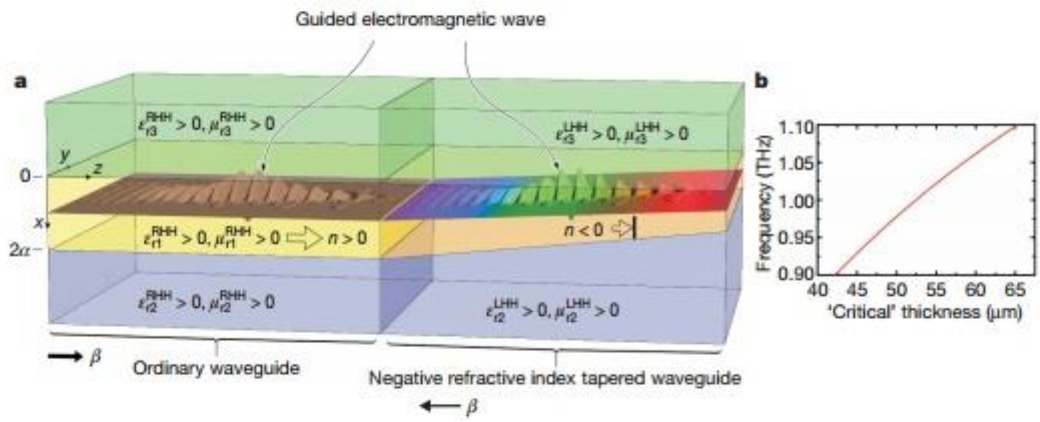

Figure 3 An illustration of the trapped rainbow concept from [6]. In a), the left hand side is an ordinary, positive refractive index waveguide, while the right hand side contains a tapered negative index material layer in its core. The tapering of this layer allows light to be stopped via successive Goos-Hänchen shifts at a certain critical thickness that is dependent on the frequency of the spectral component (b)).

Despite this, obstacles exist to achieving the trapped rainbow effect experimentally. The key issue is that of losses in the visible region of the spectrum for negative refractive index materials, where several authors have argued that even if very small amounts of loss are introduced into the system that the trapping of light in this way becomes theoretically impossible [26]. However, this does not mean that systems cannot be engineered to overcome this limitation, and the use of active plasmonic metamaterials provides a possible route for achieving this [27-28]. A schematic showing an example of a proposed structure is shown in figure 4. The use of gain enhanced metamaterials for this application also opens up a new possibility, that by not only compensating for losses but also amplifying the optical modes, this stopping of light could be exploited in low threshold cavity-free nanolasers [29].

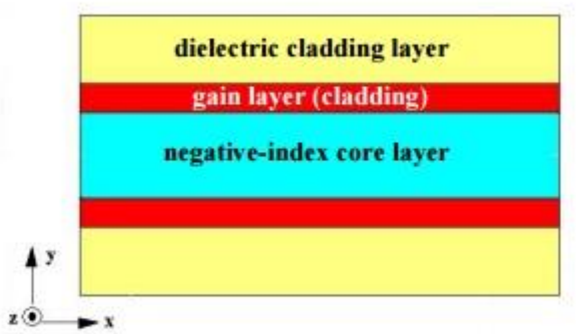

Figure 4 A schematic showing a possible design for a gain compensated waveguide, taken from [27].

Spasers and cavity-free nanolasers

The term 'spaser' was first coined back in 2003 [30], when Bergman and Stockman presented their theoretical description of the quantization of surface plasmon modes leading to stimulated emission. It differs from an ordinary laser in the sense that its transitions are radiationless, the excitation energy is directly coupled to the surface plasmons and transformed into their quasistatic field energy. The realisation that amplification can occur by increasing the gain to the point where it outweighs dissipative losses in an active plasmonic metamaterial [31] has renewed interest in this area. In fact, it has been argued that the threshold for a transition to a spaser state is identical to the regime where full loss compensation is possible [32] since the full compensation (or overcompensation) of losses leads to an instability, which, provided the gain is not depleted in some other way, leads to lasing action. An active metamaterial also supports dark modes [2]. Dark modes are modes that do not radiate or radiate weakly and are not directly excitable from free space. They cannot, for normal incidence be directly excited from free space, however, they are easily excited through spontaneous emission once the gain medium has been pumped to inversion. These modes act to deplete the gain, but in some cases can even lase themselves. The lasing of dark surface plasmon states is often referred to as spasing action.

The first spaser operating at visible wavelengths was demonstrated experimentally in 2009 [33]. This system was based on the use of gold nanoparticles measuring just 44nm across clad in dye-doped silicon dielectric cell. Researchers reported laser action under the influence of strong optical pumping, with an emission linewidth of around 6nm. Although numerous proposals for the construction of an optically coherent metamaterial emitters have been made [34-36], a system based on this idea has yet to be experimentally realised. A key benefit to achieving this would be that a design based on metamaterials would not have to be limited to being sub-wavelength in size in all three dimensions [2]. Additionally, one key problem is in the desirability of using ultra-short pulses for pumping, which is difficult to achieve via electrical injection due to a tendency to cause heat generation. Since the metals used in active plasmonic metamaterials are effective heat sinks, a successful realisation of such a system could provide a route to overcoming this problem. Several promising developments have recently been made in the area, most notably a proposal for a cavity-free nanolaser [37]. The elimination of the need for an optical cavity provides an alternative route for creating a sub-wavelength architecture. The construction of nanolasers on these scales is a potentially very important development with far reaching impact. While potential applications such as optical computing or signalling are an obvious consequence, other less obvious applications may also be found. For example, it has been suggested that the research may also have an impact on the biomedical sector due to the possibility for such devices to be embedded in synthetic tissue in order to provide sensors that can communicate signals to the nervous system [38]. The wide range of potential implications of this research highlights the significance of the development of active plasmonic metamaterials with application to light trapping and nanolasing.

Superlenses: Imaging beyond the diffraction limit

Conventional lenses are unable to produce finely detailed images due to the diffraction limit, which states that the resolving ability of a lens is limited by the spreading out of light rays due to diffraction. These lenses only focus far-field radiation, which prevents them from resolving objects smaller than approximately half the wavelength of visible light. However, there is information about the small scale structure contained in the evanescent near-field, which decays rapidly with distance and is therefore not detectable with conventional lenses. If it were possible to amplify and focus this near-field radiation then we would be able to view sub-wavelength structures.

A superlens made from specially designed metamaterials could offer a solution to these problems. When light rays from a point source enter a conventional positive index lens they refract towards the normal, whereas a ray in a negative index material will bend all the way back across the normal. This means that rays travelling through a negative index metamaterial will meet and form a reversed image at a focal point inside the lens, and then meet again at second focal point after exiting to produce a reconstructed image of the object. Additionally Pendry showed that evanescent waves travelling through a negative refractive index lens will actually be amplified [9], instead of decaying as they would do in a positive index material. Together these two properties imply that it is possible to create a superlens capable of producing a perfect image formed from both the propagating and evanescent waves, which effectively takes the optical field of an object and transfers it completely to the opposite side of the lens.

The challenge of course is to build metamaterials for which losses don't prevent imaging of small details. One suggestion[39] is to form a lens from two slabs that each have opposite permittivity and permeability. The combined optical effect of these slabs is to cancel each other out, so that an object on one side will have its image perfectly recreated on the other. The advantage to this system is that with two slabs the difficult problems, that of designing a negative refractive index material and that of designing a gain material, can be separated.

In the future an optical microscope built using superlenses would open up the possibility of high resolution real-time imaging of system on the nanometre scale. The ability to study complex processes occurring inside live biological cells would be of huge importance in the study of biology and medicine, paving the way to new discoveries in much the same way that ability to view the microscopic world with conventional lenses did. Superlenses offer not just the possibility of imaging on small scales but also of focussing light beyond the diffraction limit. Superlens based optical lithography would enable the creation of nanoscale features far smaller than anything possible with conventional optics, from single nanometre scale microchip features to ultra-high density optical data storage.

Outlook

Active metamaterials show promise for realising many metamaterial applications that are currently limited by losses, from perfect lenses to optical storage and switching. There are however still obstacles to be overcome. Practical demonstration of loss compensation has not been achieved because attempts to compensate result in lasing [32]. While this is an interesting result in itself, it currently limits the usefulness of such systems with regard to loss compensation. This could lead to a drive for new, improved plasmonic materials by the nanoscience community [20] which in turn could spin-off some new, exciting physics and improve scope for technological applications of plasmons. Recent developments show that loss compensation via the incorporation of a gain medium into metamaterial structures may be within reach, and it is likely that the successful implementation of these systems will be critical in the creation of a wide range of new technologies. Gain enhanced negative index metamaterials could lead to advances in real-time high resolution imaging, with the potential to revolutionise the study of cellular processes. This, coupled with their potential for controlling light on the nanoscale make active plasmonic metamaterials a vital development for the future of nanoscience, with far reaching implications across science as a whole.

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