Controlling Light on the Nanoscale

John B. Pendry*

(*Invited Review*)

Abstract—In many ways light and nanoscience do not mix well. By convention light can be focussed to a spot no smaller than about a micron whereas nano structures by definition are three orders of magnitude smaller in scale. However recent theoretical advances show how to control light at the nanoscale, provided we can find the correct materials for our devices. I shall describe these new theories, and how they enable us to concentrate light to better than a nanometre. In this way light can detect single molecules, and the huge concentrations of optical energy can force photons to interact with one another which they normally do not do.

1. INTRODUCTION

Light is an ancient subject of man's curiosity and has been exploited for many practical purposes. Key to harnessing light is to understand how it moves. Snell told us how light refracts at an interface between two media as shown in Fig. 1.
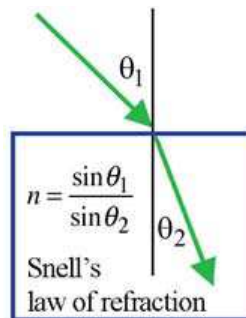

Figure 1. One of the earliest laws of optics is attributed to Snell. It relates the change in angle as light passes from air into another medium to the 'refractive index', n, of the medium.

His law has been used to design optical systems which enable us to view very small objects. However it soon became apparent that there are limitations to how small an object could be resolved: nothing much smaller than the wavelength of light can be seen in a conventional microscope, no matter how precisely it is manufactured. To understand how to control light on the sub micron scale, on the nanoscale we need to look inside the wavelength at the individual constituents of light. The first clue to this came from Michael Faraday who showed that a changing magnetic field produced an electric field. See Fig. 2. His laws of induction were formalised by James Clerk Maxwell who completed the
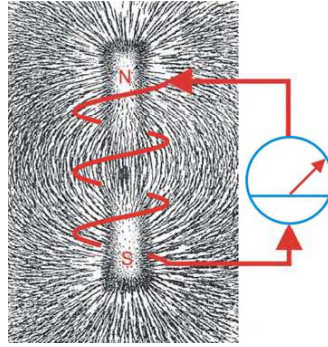
Figure 2. Faraday imagined a magnet to be surround by a force field, the magnetic lines of force. If the magnet is moved these magnetic field lines induce a current in any wire which the lines cut across. Here we show a solenoid into which a bar magnet is being inserted.

theory of light by adding a second complementary rule: a changing electric field produces a magnetic field. These two laws enable light to be understood as a dance between ever changing electric and ever changing magnetic fields, each sustaining the other as they move infinite distances through space. It is these internal components of light, particularly the electrical component, that we must address if we are to manipulate light on the nanoscale.

A third influential character in the story of light is Albert Einstein. He had a highly original theory of gravity: in contrast to Newton's theory of forces acting at a distance, Einstein postulated that in the presence of gravitation, space is distorted. He measured this distortion by a quantity called the 'metric' which measures how much space has been crushed or expanded. In this distorted space massive objects no longer travel in a straight line. In particular even light which has no mass, is affected by this distortion of space and its path can be bent by a massive object such as the sun or a black hole. In fact Einstein's theory has a very simple interpretation in three dimensional space: as far as light is concerned Einstein's metric is a refractive index. In the next section we bring together these ingredients to make a new approach to the control of light, exact at the level of Maxwell's equations and which can be used to control light on the nanoscale.

2. TRANSFORMATION OPTICS

Snell's law of refraction takes no account of the wave nature of light and has nothing to say about the electromagnetic components of light. Yet it still has a role as an approximate working tool of optics. This persistence is due to the intuitive nature of the law working as it does in terms of rays of light which we can picture moving through a lens. This picture we exploit to understand how changing the design will affect the trajectories of the rays. In contrast Maxwell's equations are an exact description of classical light but fail to give that vital picture required by our intuition before it can begin to create. It is this dilemma that transformation optics is designed to resolve.

First we must cast aside the concept of a ray and work instead with the ingredients of Maxwell's equations: the electric and magnetic fields. We recognise that although we have discarded the rays, an equally appealing picture is to be had in terms of the electric and magnetic lines of force. These are the elements that our new theory is designed to manipulate, retaining the pictorial element so vital to our intuition.

Suppose we want to change the configuration of fields. The fields are of course embedded in three dimensional space, and if we believe Einstein we are free to imagine that this space can be distorted. The distortion will carry the lines of force with it as shown in Fig. 3 where a field line starts off as straight, Fig. 3(a), but as the space is distorted, Fig. 3(b), changes its shape. Coordinates are drawn so that the distortion can be followed and which can be thought of as a transformation of coordinates. Einstein has given us a formula for the new metric of the distorted space. Fortunately we do not need to distort space for light because he also told us that changing the refractive index has the same effect.
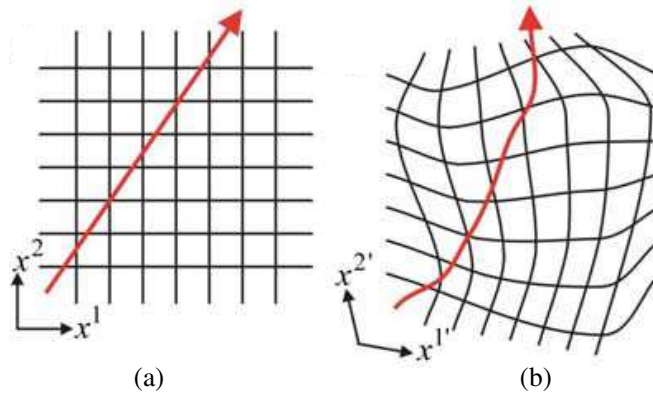
Figure 3. To the left we show in red a line of force (electric or magnetic) whose direction we wish to change to the shape shown on the right. By distorting the space through which the line travels, as if the space were made of rubber, we can direct the line of force almost as we please. The spatial distortion dictates the material parameters we must use to ensure that the field lines satisfy Maxwell's equations. Coordinate lines are shown in black: (a) x^j and (b) $x^{j'}$ in the distorted frame.

To get a little formal for a moment: the refractive index, n, is given by the electric and magnetic response of the material, ε, μ,

$$n = \sqrt{\varepsilon\mu} \tag{1}$$

and the spatial distortion $x^{j'}\left(x^j\right)$ determines the new values of electromagnetic response,

$$\begin{aligned}
\varepsilon'^{i'j'} &= \left[\det\left(\Lambda\right)\right]^{-1} \Lambda_i^{i'} \Lambda_j^{j'} \varepsilon^{ij} \\
\mu'^{i'j'} &= \left[\det\left(\Lambda\right)\right]^{-1} \Lambda_i^{i'} \Lambda_j^{j'} \mu^{ij}
\end{aligned} \tag{2}$$

where,

$$\Lambda_j^{j'} = \frac{\partial x^{j'}}{\partial x^j} \tag{3}$$

So once we have decided where the field lines go and determined $x^{j'}\left(x^j\right)$, Equation (2) tells us what material parameters we must use if Maxwell's equations are to be obeyed. More technical details can be found in [1, 2].

3. TRANSFORMATION OPTICS AND INVISIBILITY

To illustrate the power of transformation optics we can use this new tool to design a cloak of invisibility. We require that the cloak is finite, and that it surrounds the object to be hidden.

In Fig. 4, we illustrate our strategy. Our objective is to exclude all light from the interior of the circle by using a finite cloak to deflect the light around the circle, but leaving its trajectory unchanged outside the cloak. In this way an external observer can neither see the content of the circle, nor be aware that a cloak is present because it creates no disturbance in the light seen by the observer. Transformation optics is the ideal tool for this problem. Imagine that we can compress all of the 'space' inside the circle into the cloak (remember that Einstein said that space could be distorted) taking the field lines along with the compression. This leaves an empty circle into which we can place anything we choose and no light can see it. Also no disturbance of space outside the cloak has taken place and therefore there is no disturbance in the fields which constitute incident light We reported this cloak to *Science* in 2006 [3]. A different scheme based on conformal mapping was also published at the same time [4].

From the compression of the coordinates, and using Equation (2) we can calculate the material parameters of the cloak. Needless to say they are somewhat unusual and set a challenge to experimentalists. How that challenge has been met is the subject of the next section.
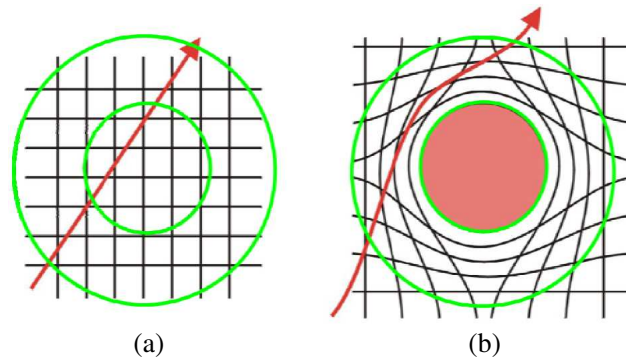
Figure 4. (a) Any object placed within the circle will be seen by incident light represented by a red field line. (b) All of the 'space' within the inner circle has been forced outside the circle and compressed into a cloak contained by the outer circle. Coordinate lines in black record this distortion which carries all the field lines with it excluding them from the interior. Note that outside the cloaking region we do not distort space and therefore the field lines are unaltered. No observer can detect that any disturbance has taken place.

4. METAMATERIALS

There remains the problem of finding a material with the required values of ε, μ. Nature gives us a limited palette of material parameters so often there are devices that cannot be made because the right materials cannot be found. It was to address this problem that I came up with the concept of a metamaterial. A conventional material is made up of atoms so that its macroscopic response to an applied field is an average over the billions of individual atomic responses. In fact the individual units do not have to be atoms or even molecules, they can be any tiny unit provided that the unit's size is much less than the wavelength of light and the light sees an average response. Fig. 5 illustrates the concept, in this instance introducing minute metallic rings into the structure.
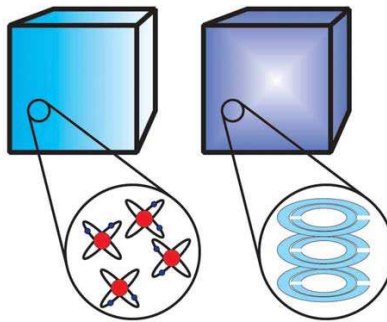

Figure 5. The concept of a metamaterial. On the left we show a conventional material containing only atoms and molecules. On the right is a material containing artificial structures whose shape, rather than composition determines how they respond to applied fields.

So we are not bound to simply varying chemical composition when changing the material properties but have the additional parameter of structure to play with [5, 6]. This concept has been exploited to build the first cloak which was designed to work at radar frequencies — the sub wavelength structures required by the metamaterial are much easier to fabricate because the wavelength is of the order of a few centimetres [5]. See Fig. 6.

More recently the metamaterial technology has been extended to visible light. Fig. 7 shows another metamaterial structure, this time with sub micron dimensions. It is designed not for the purposes of cloaking but to produce a material property never observed in natural materials: a negative refractive index. In the next section the extraordinary effects of negative refraction will be described.
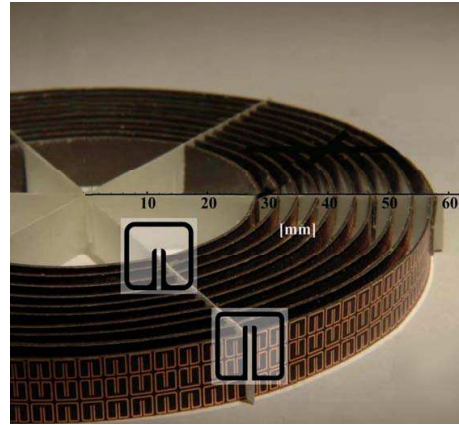
Figure 6. The first experimental realisation of a cloak of invisibility. The cloak operates at radar wavelengths of the order of 10 cm and is built of a metamaterial comprising millimetric copper loops, shown inset, whose shape is varied with the radius in order to tune the material properties.
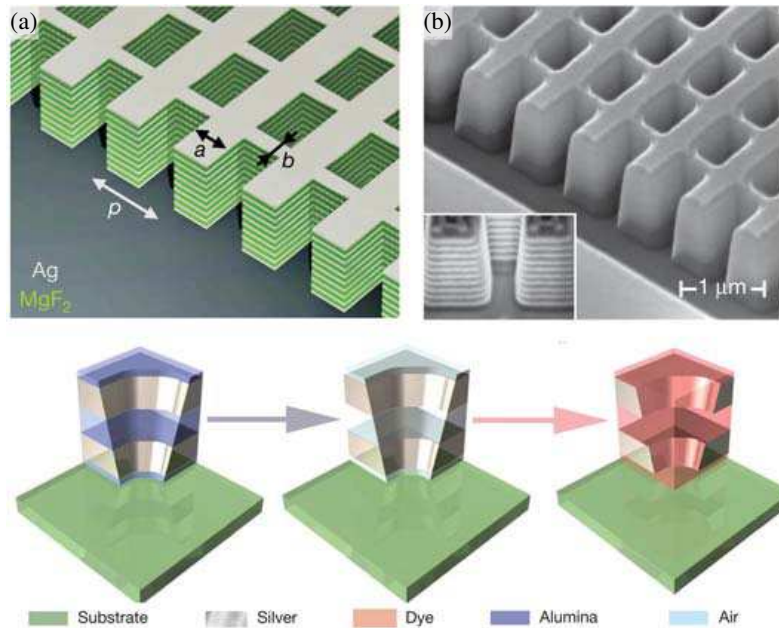

Figure 7. An example of a metamaterial known as the 'fishnet' structure. It consist of alternate layers of alumina and silver punctured by a periodic set of holes as shown diagrammatically on the left and in an electron micrograph on the right. In some realisations a dye is incorporated to act as an amplifying medium compensating for any loss in the silver [8, 9].

5. NEGATIVE REFRACTIVE INDEX AND THE PERFECT LENS

Some time ago Victor Veselago predicted that if a material had a negative response both to electric and to magnetic fields ($\varepsilon < 0$, $\mu < 0$), it would refract light in a negative direction: see Fig. 8 [10]. By negative response we mean that the material polarises in the opposite direction to the applied field. He showed theoretically that some extraordinary effects follow from this property, but was never able to find a material satisfying the negative index requirements.

There the story rested until the advent of metamaterials which had been shown to create materials with $\varepsilon < 0$, and another set of materials with $\mu < 0$. The USD team [11] picked up on these results
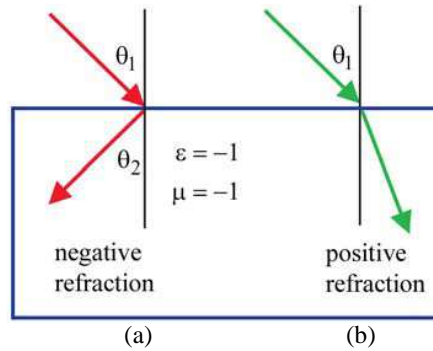
Figure 8. (a) A material with a negative index of refraction bends light to the same side of the normal as the incident wave. (b) In contrast to normal, positive, refraction where the light refracts to the opposite side of the normal.

and combined them to create the first material with $\varepsilon < 0$, $\mu < 0$ and hence a negative refractive index according to the Veselago prescription.

Why should a negative index of refraction be such an extraordinary thing? I remind the reader that as far as light is concerned Einstein equated the index of refraction with the metric of a space. The metric measures how much space is crushed and therefore a negative refraction index means that light sees space as being turned inside out creating a sort of optical antimatter. A medium with a negative refractive index appears to 'annihilate' the adjoining regions of space. This strange effect can be seen in the ability of negatively refracting materials to focus light.
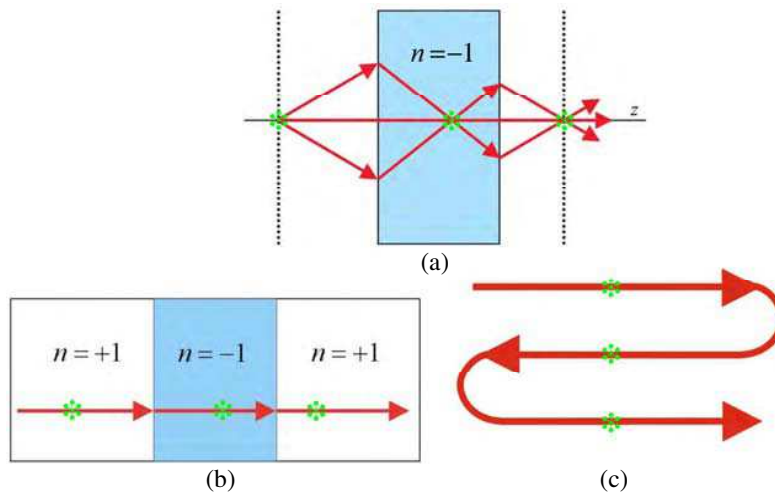

Figure 9. (a) A slab of metamaterial with a negative refractive index refracts rays diverging from a focus (green star) back towards the axis forming a second focus, and then a third focus after leaving the slab. (b) We see a schematic of light passing through the lens. However light acts as though negative refraction creates a piece of negative space so that (c) is the trajectory from the light's point of view: light behaves as though it is travelling backwards inside the metamaterial.

Figure 9 shows how a planar slab of negatively refracting material bends rays of light back towards the axis, bringing them to a focus inside the material, and to a second focus on the far side. Transformation optics interprets this focussing in terms of an effectively negative space seen by the light. Negative refraction cancels the effects of neighbouring positive refraction reversing the divergence so that light follows a trajectory that bends back on itself: the focal point is passed three times in this picture.
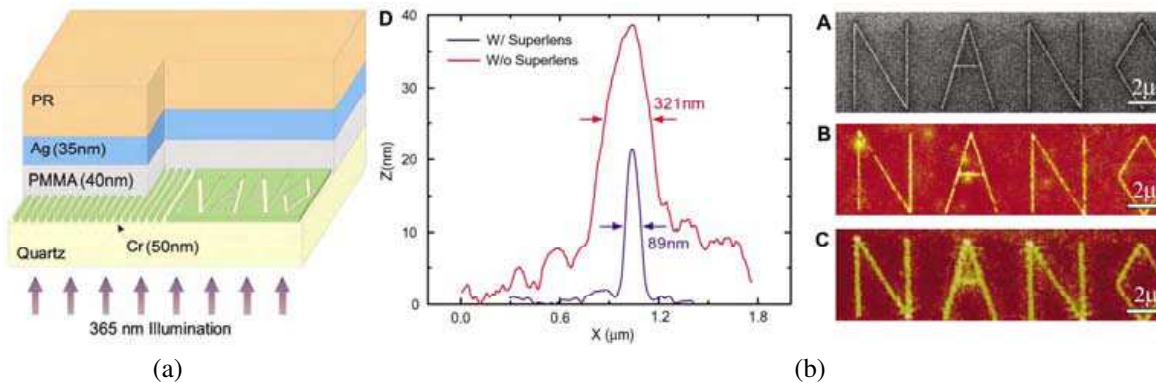
Figure 10. (a) The objects to be imaged are inscribed into chrome as an array of 60 nm wide slots of 120 nm pitch. The image is recorded on a photoresist placed on the far side of the superlens. (b) A: focussed ion beam image of the object; B: atomic force microscope scan of the image; C: atomic force microscope scan with the superlens removed. Centre: D: a typical cross section of the line width scanned with and without the superlens in place.

Clearly this is a most unusual lens. The focussing effect was first noted by Veselago who applied Snell's law to show how rays are refracted. However the new interpretation using transformation optics is more profound because it is not an approximate treatment, but exact at the level of Maxwell's equations. Hence when we deduce that there is a focus, that is an exact statement and the focus has to be perfect. In 2000, I showed that the Veselago lens will focus light as perfectly as we can make the lens with no restrictions from the wavelength of light [12]. This is a remarkable result that blows apart the wavelength restriction on controlling light and opens the way to using light an the nanoscale.

To test this new theory scientists at UCB built an approximation to the negatively refracting lens in the form of a slab of silver [13, 14]. Their object was a 60 nm wide grating, and the word 'nano', etched into a layer of chromium shown on the left of Fig. 10. Above the object was a spacer layer 40 nm thick and above that the 35 nm thick silver lens. The lens acts so as to annihilate the spacer layer producing an image immediately above the silver. Finally, to record the image, a layer of photo resist sits on top of the silver.

After exposure to 365 nm wavelength light, the photoresist is stripped away from the rest of the experiment, developed, and scanned with an atomic force microscope to reveal the image shown on the right of Fig. 10. Also shown in the centre is a scan through one of the lines in the image illustrating the factor of six improvement in resolution due to the lens. Their results confirm that light can be focused much more precisely than the wavelength of 365 nm would suggest. The resolution is now dictated by imperfections in construction of the lens not by the wavelength

6. HARVESTING LIGHT ON THE NANOSCALE

The transformation optics tool can be exploited to design nanoscale optical devices. Here we show how to gather light incident on a structure and concentrate the radiation in less than a square nanometre. The difference between harvesting and focussing is that focussing maps an object point by point into an image, whereas harvesting gathers all the incident light and concentrates it onto a single point. Both face the same challenges of resolution, but harvesting is a simpler task and therefore this is the area in which most progress has been made. A review of this work can be found in [15].

Transformation optics as a design tool gives us great flexibility because in the initial stages we need not worry about the shape of our device since we can transform it into another quite different shape using the usual rules. Hence we start by considering a very simple device which grabs radiation emitted by a point source, such as an excited atom. Fig. 11(a) shows a waveguide cut into silver which supports very short wavelength excitations on the silver surface. Known as surface plasmons these comprise oscillations of the conduction electron density in the silver. Inside the waveguide a dipole source emits

radiation which is captured by the surface plasmons and radiated away to infinity. So in step one we have captured the radiation, but not concentrated it.

Next we apply a two dimensional coordinate transformation formally known as an inversion which maps points at infinity to the origin and vice versa. In the notation of Fig. 3:

$$x^{1\prime} = \frac{x^1}{\sqrt{(x^1)^2 + (x^2)^2}}, \quad x^{2\prime} = \frac{x^2}{\sqrt{(x^1)^2 + (x^2)^2}} \tag{4}$$

where the origin in the x^j frame is chosen to be the centre of the dipole. The transformation maps the infinite planes into two cylinders which touch at the point mapped from infinity in the first frame. The dipole is now removed to infinity and produces a uniform electric field acting on the cylinders.

This type of transformation is a 'conformal' transformation and leaves the dielectric properties of silver unchanged.

Figure 11(b) shows the resulting structure which performs the task we set ourselves: light incident on the structure is captured by the cylinders and shepherded towards the touching point. Just as light in the original structure never reaches infinity, in the transformed structure light never reaches the touching point, slowing down as it moves around the circumference of the cylinder. Fig. 11(c) shows the electric field intensity on the surface of a cylinder as a function of angle. This leads to a concentration of energy: just as traffic on a motorway becomes more densely packed when the speed is reduced. We have created a sort of 'black hole' into which radiation vanishes never to emerge.
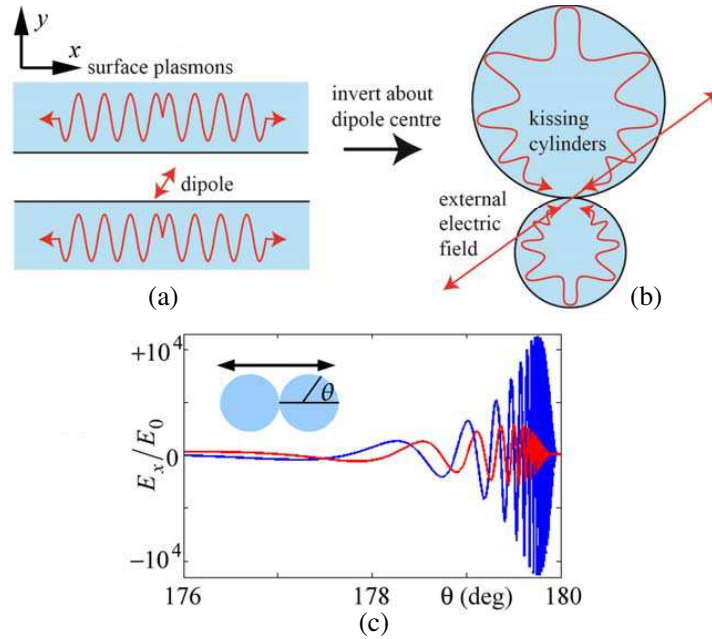

Figure 11. (a) A 2D waveguide cut into a slab of silver metal captures radiation from a dipole source. (b) After inversion about the centre of the dipole, the dipole source is now at infinity producing a uniform incident field, and the waveguide is mapped into two silver cylinders touching at a point which before inversion sat at infinity. (c) A plot of the electric fields relative to the incident field as a function of angle around the surface of one of the cylinders. The cylinders are approximately 20 nm in diameter.

Ideally the energy density would rise to infinity at the touching point but this is prevented by several effects one of which is the resistive loss in the silver to which the light as more vulnerable as it slows down, eventually killing the enhancement effect. This results in the collapse of enhancement seen in Fig. 11(c). Nevertheless the field achieves enormous enhancement before the collapse: more than a factor of 10^3 in amplitude and therefore more than 10^6 in energy density. This has potential to greatly enhance sensing of molecular species down to the single molecule level with potential applications in biosensing.

Experimental realisation of these theoretical predictions is challenging: surfaces need to be made as smooth as possible, much more highly polished than conventional mirrors which are merely flat to slightly better than a micron. Here the requirement is smoothness to less than a nanometre. A team at Duke university has achieved these exacting conditions in a recently published experiment [16]. Fig. 12 shows their set up.
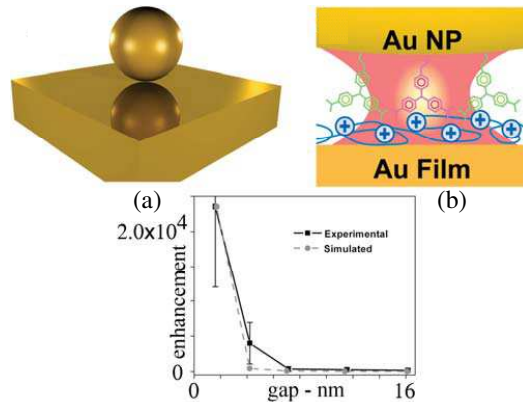

Figure 12. (a) A gold sphere separated from the gold film by a few nanometres. (b) The spacing is controlled by a layer of molecules that can be 1, 2, 3, ... layers thick. Embedded in the layer are some other molecules that give off a characteristic Raman signal when illuminated. This enables the local intensity of light to be measured. (c) The enhancement of the Raman signal as a function of the separation.

A gold sphere is placed at a precise distance from a flat gold surface, controlled by a known thickness of molecular layers. Light is shone onto the system and is concentrated at the point of closest approach where its intensity can be measured by the response of Raman active molecules embedded in the separating film. These exhibit a characteristic loss which depends on the local intensity enhancement.

The results are shown at the bottom of the figure indicating an enhancement of up to 2×10^4 of the Raman signal at the closest distance of approach which in this experiment was 2 nm. As experiments become more refined and distances more precisely defined, the separation between the surfaces will be reduced, with enhancements converging on theoretical predictions.

7. CONCLUSIONS

Breaking the diffraction limit has opened new vistas for exploitation of light on the nanoscale. To some extent theory has raced ahead of experiment, but the challenges are being met and new technology developed. It is an exciting time of nanoscale light. Further reading is to be had in [17–19].

REFERENCES

1. Ward, A. J. and J. B. Pendry, "Refraction and geometry in Maxwell's equations," *Journal of Modern Optics*, Vol. 43, 773–93, 1996.
2. Schurig, D., J. B. Pendry, and D. R. Smith, "Calculation of material properties and ray tracing in transformation media," *Optics Express*, Vol. 14, No. 21, 9794–9804, 2006.
3. Pendry, J. B., D. Schurig, and D. R. Smith, "Controlling electromagnetic fields," *Science*, Vol. 312, 1780–1782, 2006.
4. Leonhardt, U., "Optical conformal mapping," *Science*, Vol. 312, 1777–1780, 2006.
5. Schurig, D., J. J. Mock, B. J. Justice, S. A. Cummer, J. B. Pendry, A. F. Starr, and D. R. Smith, "Demonstration of a metamaterial electromagnetic cloak at microwave frequencies," *Science*, Vol. 314, 977–980, 2006.

6. Pendry, J. B., A. J. Holden, D. J. Robbins, and W. J. Stewart, "Magnetism from conductors and enhanced non-linear phenomena," *IEEE Trans. Micr. Theory and Techniques*, Vol. 47, 2075, 1999.

7. Pendry, J. B., A. J. Holden, W. J. Stewart, and I. Youngs, "Extremely low frequency plasmons in metallic mesostructures," *Phys. Rev. Lett.*, Vol. 76, 4773, 1996.

8. Valentine, J., S. Zhang, T. Zentgraf, E. Ulin-Avila, D. A. Genov, G. Bartal, and X. Zhang, "Three dimensional optical metamaterial exhibiting negative refractive index," *Nature*, Vol. 455, 376, 2008.

9. Xiao, S., V. P. Drachev, A. V. Kildishev, X. Ni, U. K. Chettiar, H. K. Yuan, and V. M. Shalaev, "Loss-free and active optical negative-index metamaterials," *Nature*, Vol. 466, 735, 2010.

10. Veselago, V. G., "The electrodynamics of substances with simultaneously negative values of ϵ and μ," *Soviet Physics USPEKI*, Vol. 10, 509, 1968.

11. Smith, D. R., W. J. Padilla, D. C. Vier, S. C. Nemat-Nasser, and S. Schultz, "Composite medium with simultaneously negative permeability and permittivity," *Phys. Rev. Lett.*, Vol. 84, 4184, 2000.

12. Pendry, J. B., "Negative refraction makes a perfect lens," *Phys. Rev. Lett.*, Vol. 85, 3966, 2000.

13. Fang, N., H. Lee, C. Sun, and X. Zhang, "Sub-diffraction-limited optical imaging with a silver superlens," *Science*, Vol. 308, 534, 2005.

14. Blaikie, R. J. and D. O. S. Melville, "Imaging through planar silver lenses in the optical near field," *J. Opt. A: Pure Appl. Opt.*, Vol. 7, S176, 2005.

15. Pendry, J. B., A. Aubry, D. R. Smith, and S. A. Maier, "Transformation optics and subwavelength control of light," *Science*, Vol. 337, 549–552, 2012.

16. Hill, R. T., et al., "Leveraging nanoscale plasmonic modes to achieve reproducible enhancement of light," *Nano Letters*, Vol. 10, 4150, 2010.

17. Pendry, J. B., "Negative refraction," *Contemporary Physics*, Vol. 45, 191–202, 2004.

18. Pendry, J. B., "Metamaterials and the control of electromagnetic fields" *Coherence and Quantum Optics IX*, edited by N. P. Bigelow, J. H. Eberly, and C. R. Stroud, Jr. (OSA Publications, 2009), 42–52, 2008.

19. Aubry, A. and J. B. Pendry, "Transformation optics for plasmonics," *Active Plasmonics and Tuneable Plasmonic Metamaterials*, A. Zayats and S. Maier (eds.), John Wiley & Sons, 2013.