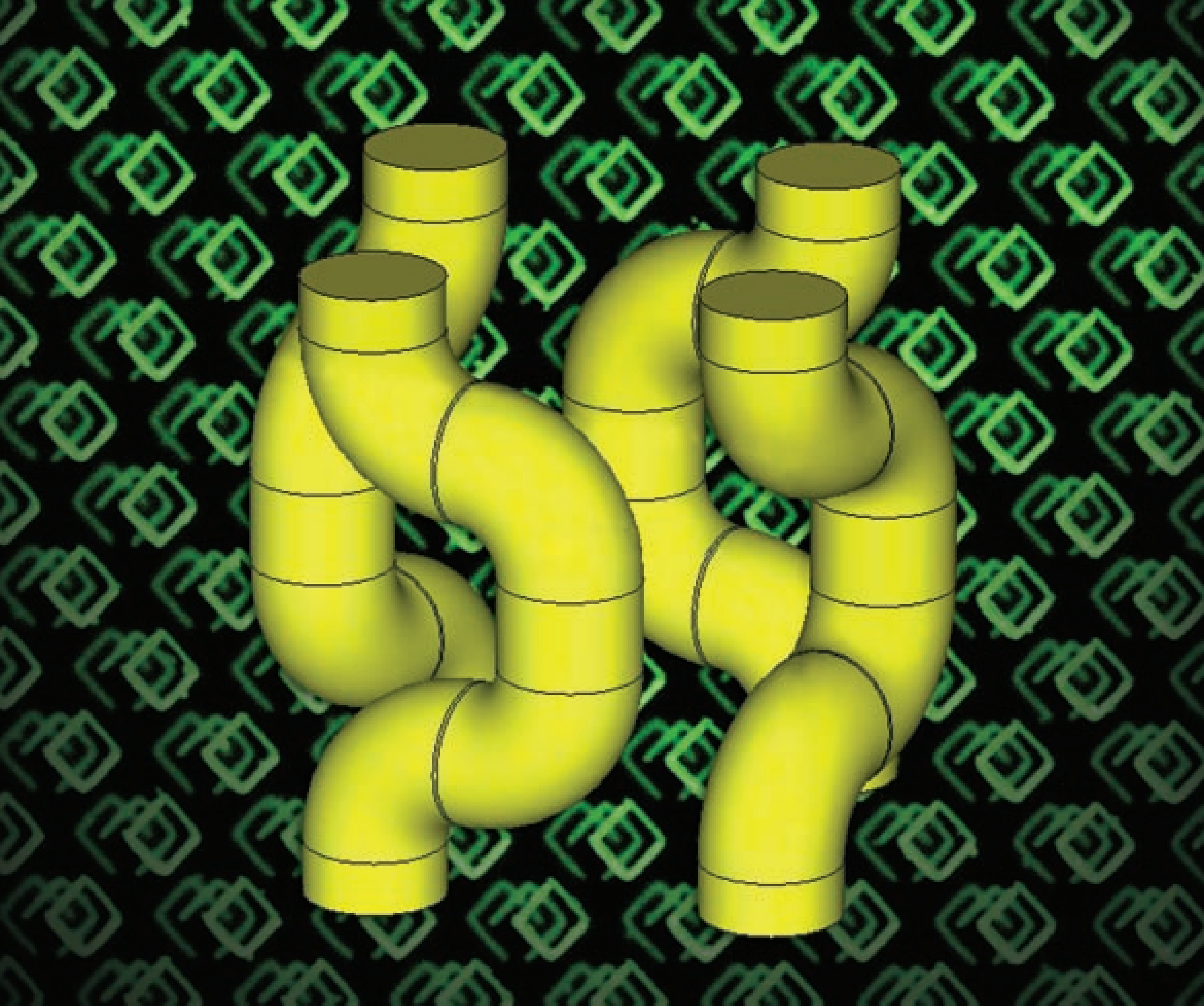
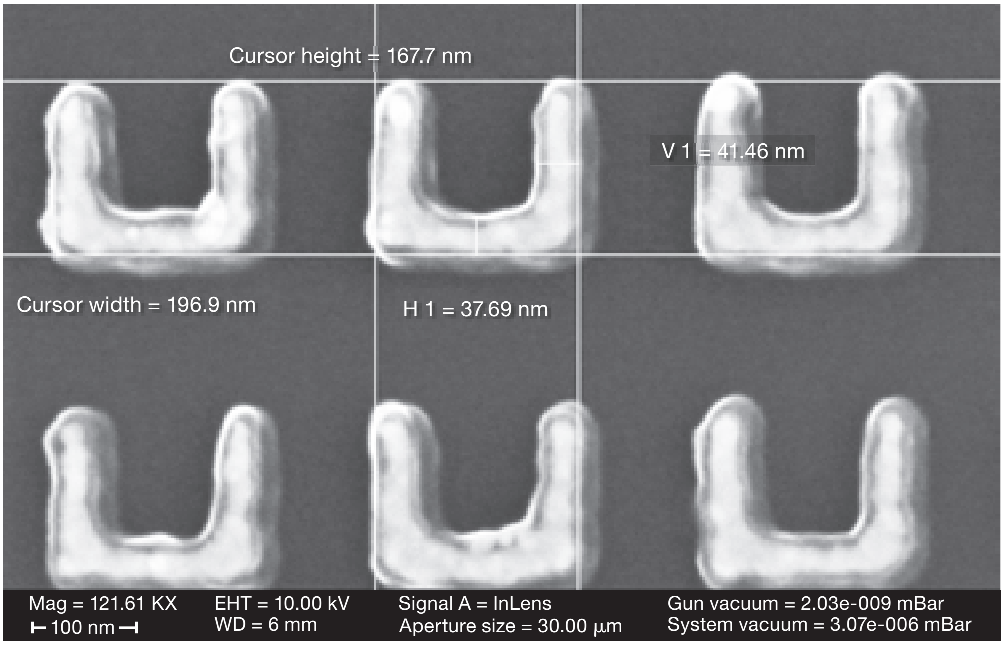


The Magical World of Photonic Metamaterials

Ekmel Ozbay

Metamaterials have opened a world of new possibilities that would have seemed like science fiction a decade ago, including subwavelength imaging, super lenses, perfect lenses and cloaking. This article explores the progress of a field that is making optical magic.



SRR structures: Photo of nanoscale split ring resonator structures fabricated in Bilkent University.

Starting in high school physics, we learn that light is a combination of electric and magnetic fields. As light propagates through matter, conventional materials only react to the electric field, resulting in most common optical effects, including refraction, diffraction and imaging.

Forty years ago, Victor Veselago pondered the question of whether matter also interacts with the magnetic field of light. He then tabulated the materials according to the sign (positive and negative) of their electric permittivity (ε) and magnetic permeability (μ). When both electric and magnetic properties were negative ($\varepsilon<0$ and $\mu<0$), he showed that the solution of the Maxwell equations resulted in an index of refraction with a negative sign.

Veselago had to wait more than 30 years for his theoretical predictions to be confirmed experimentally—with the engineering of a new type of structure called metamaterials. The seminal work of Sir John Pendry provided the blueprint for the realization of metallic-based resonant structures called split-ring resonators (SRRs) that exhibit $\mu<0$ at specific resonance frequencies. Smith et al. combined an array of SRRs ($\mu<0$) and an array of metallic wires ($\varepsilon<0$) in order to create double-negative composite metamaterials. They were then used in a wedge-type structure to experimentally demonstrate negative refraction at microwave frequencies.

In such cases, the famous "right-handed rule" between the electric and magnetic fields becomes left-handed. Thus, these materials are known as left-handed materials (LHMs) or negative index materials (NIMs). Scientists have demonstrated that—at least in theory—metamaterials can also be used to achieve magical applications such as subwavelength imaging, super lenses, perfect lenses, cloaking, etc. Although the early experiments were performed at microwave frequencies, it took only a few years for scientists to downscale these structures to optical frequencies. These nano-scale metamaterials are now called photonic metamaterials.

Magnetism at optical frequencies

Magnetism is usually associated with radio frequencies. However, the SRR structure suggested by Sir John Pendry can be downscaled in order to achieve magnetism at optical frequencies. Using electron beam nanolithography techniques, SRRs can be fabricated with dimensions reaching nanometer scales. The three connected metallic rods, which make the U-type SRR structure, act as a nano-inductor, while the gap between the ends of the U-shape act like a nano-capacitor. This nano-LC circuit acts as an LC resonator with a resonance frequency at optical frequencies. More important, the typical size of these resonant inclusions is approximately 10 times smaller than the vacuum wavelength of the light at the resonance frequency. The electromagnetic properties of such optical-scale subwavelength structures can then be evaluated by using an effective medium approximation.

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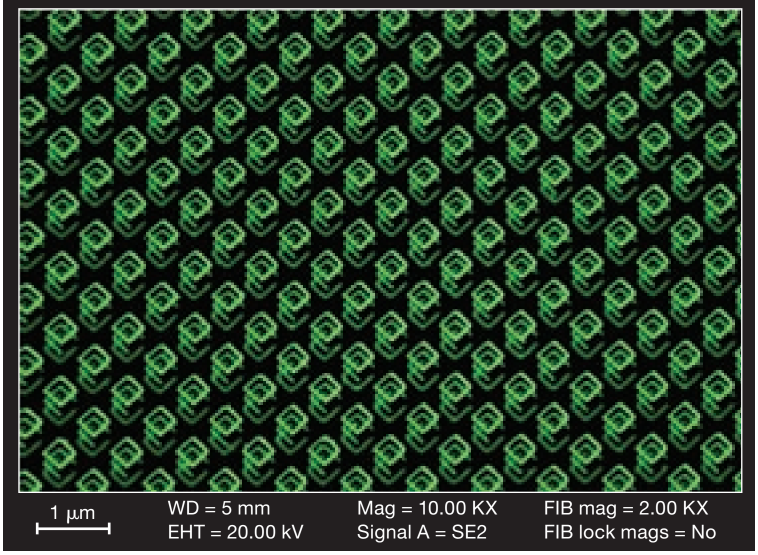

[Four-layer stacked photonic metamaterial (200 x 200 μm^2)]

Although a single-layer SRR structure can easily be constructed on a dielectric surface, it is relatively difficult to stack these structures due to the tight alignment tolerance requirements. Harald Giessen and his group have recently reported a new method in which metamaterials in the near-infrared spectral region can be fabricated using a layer-by-layer technique. Robust alignment marks are made on the sample, allowing for the accurate alignment of the electron beam lithography. Once a single layer is fabricated, planarization using a special polymer warrants a flat surface for the next lithography step.

In principle, an arbitrary number of subsequent layers can be made in this way, with arbitrary unit cells as well as the spatial arrangements of subsequent layers. The figure above shows a 200 × 200 μm^2 four-layer split-ring resonator structure with a 90-degree twist between subsequent layers. In these stacked structures, the coupling of the electric and magnetic resonances becomes crucial for understanding the spectra of the transmitted light. The hybridization of electric and magnetic resonances cause spectra that resemble energy states in molecular physics.

Photonic metamaterials with a negative index

Conventional SRRs provide a clever way to achieve magnetism at optical frequencies. In order to excite the magnetic resonance of the SRR, the incoming light should propagate in a direction that is parallel to the SRR plane. This is necessary in order for the magnetic field to couple to the SRR, which will induce a current that will in turn create a magnetic field in the reverse direction. In most photonic applications, the optical beam is usually perpendicular to the surface. This means that we need an SRR perpendicular to the substrate plane, so that the incident beam can excite the magnetic resonance of the SRR structure.

However, the conventional U-type SRR geometry is quite difficult to fabricate and stack in a vertical direction. Vladimir Shalaev and his co-workers at Purdue University came up with a solution. They modified the classical SRR into a coupled nanostrip structure. The coupled nanostrips were fabricated by electron beam lithography techniques. The geometries of the samples were first defined with an electron beam writer on a glass substrate with a thin layer of indium tin oxide.

Then, the researchers used standard electron beam deposition in order to produce a stack of silver and alumina layers. Using the coupled nanostructures with various dimensions, the Purdue team was able to create metamaterials that exhibited magnetic response at optical frequencies across the entire visible spectrum. The "rainbow magnetism" obtained from these structures provides a universal building block for producing controllable optical magnetism for various implementations.

Although the coupled nanostrip structure results in magnetism at even shorter wavelengths, it still does not create the desired negative-index behavior. In order to achieve a negative index at optical frequencies, the coupled nanostrip structures need to be integrated and stacked with metallic nanowires. If the nanowires are fabricated on the same plane as the coupled nanorods, the structure resembles that of a fishnet.

Martin Wegener and his colleagues at Karlsruhe University have recently reported a three-layer "fishnet" metamaterial. The silver-based samples were fabricated by standard electron-beam lithography and a lift-off procedure. The lattice constant along the stacking direction was much smaller than the wavelength of light. This means that the effective-medium approximation is justified.

Although the integration of nanowires and coupled nanostrips create a double negative medium, they do not automatically guarantee a negative index. The complex refractive index of the metamaterials can only be numerically calculated by applying retrieval methods to the reflectance and transmittance characteristics. Using these numerical retrieval methods, researchers showed that the three-layer metamaterial exhibited a negative index value of -2.2 around 1,410 nm.

X. Zhang's group at the University of California at Berkeley has significantly improved the stacking and measurement of

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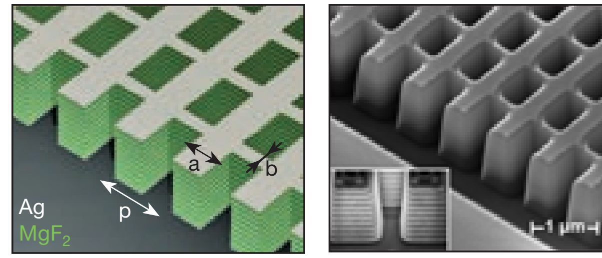
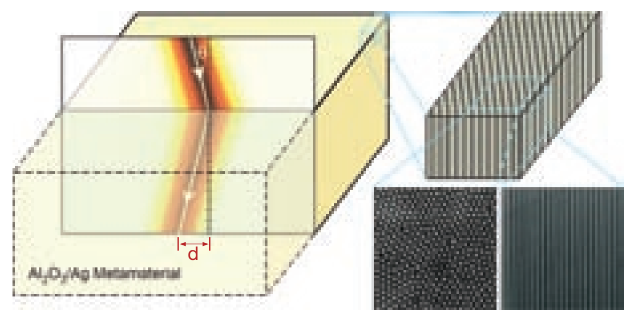
[Fishnet metamaterials]



Schematic and SEM image of fabricated 10-layer fishnet photonic metamaterial structure.

[Silver nanowire metamaterials]



Left: Schematic of negative refraction from air into the silver nanowire metamaterials. Right: SEM images of the nanowires embedded in an alumina matrix.

fishnet structures. They reported a 10-layer fishnet metamaterial structure, which was fabricated on a multilayer metal-dielectric stack using focused ion beam milling. This process resulted in nanometer-sized features with a high aspect ratio. The proposed multilayer fishnet pattern was milled on 21 alternating films of silver and magnesium fluoride, resulting in 10 functional layers.

A nano-prism was created in the multilayer fishnet stack. It was then used to measure the refractive index of the planar metamaterial. Instead of relying on numerical retrieval methods, the researchers measured the refractive index of the structures by observing the refraction angle of the light passing through the prism by Snell's Law. This method provides a direct and unambiguous determination of the refractive index, as the refraction angle depends solely on the phase gradient that the light beam experiences when refracted from the angled output face.

The UC Berkeley group used a tunable light source to determine the refractive index at different wavelengths. They focused the beam on the sample and placed a charged coupled device (CCD) camera in the Fourier plane. Then they measured the beam shift resulting from the light bending at the prism output to determine the index of refraction at different wavelengths, ranging from 1,200 to 1,775 nm.

The refractive index varied from $n = 0.63$ at 1,200 nm to $n = -1.23$ at 1,775 nm. As the wavelength increased, the beam shift resulting from the prism refraction changed from positive to negative, indicating a transition from a positive index in the shorter wavelengths to a negative index in the longer ones. At 1,475 nm, the index of refraction approached a value of zero, where the beam does not acquire any phase while propagating in the metamaterial.

In a related article, the Berkeley group reported observations of negative refraction in metallic photonic-crystal-based metamaterials composed of silver nanowires embedded in alumina. The figure on the bottom right shows a schematic of negative refraction from air into the silver nanowire metamaterial and the SEM images of the nanowires embedded in an alumina matrix. The researchers measured the sample by illuminating a collimated diode laser beam at different incident angles.

The transmitted light was then mapped by scanning the bottom surface. The group refractive indices of the metallic photonic-crystal-based metamaterial were then shown to be -4.0 and 2.2 for TM and TE light, respectively. As expected from a photonic-crystal-based metamaterial, the phase refractive index of the metamaterial remains positive; this is in contrast to left-handed metamaterials. For normal incidence, the light intensity only decays at a rate of 43 percent for each micron thickness in the medium at 780 nm—which corresponds to a few orders of magnitude improvement over the loss factors that are achieved from left-handed metamaterials reported at optical wavelengths.

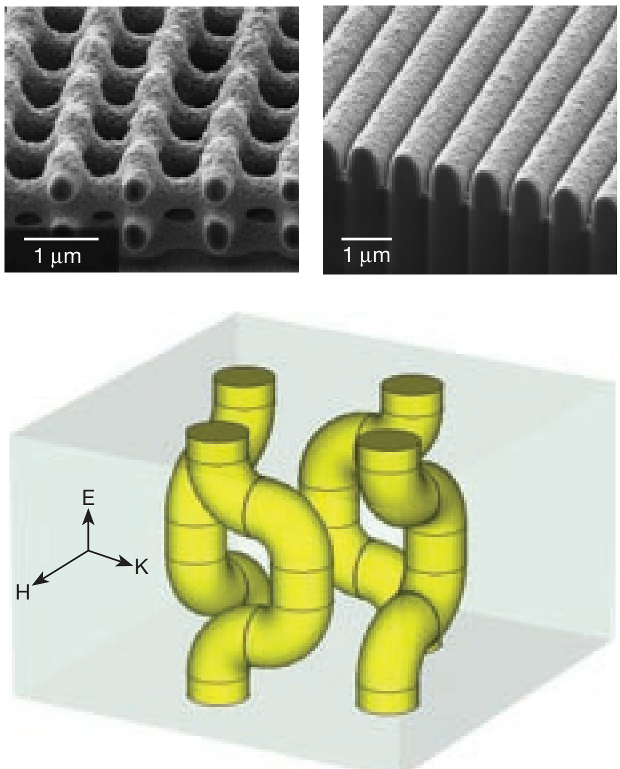
Bragg-type scattering is the major physics underlying the negative refraction in metallic photonic crystals. When compared to left-handed metamaterials, these structures possess relatively lower loss and a broader spectral range for a span of incident angles. Similar to dielectric-based photonic crystals, metallic photonic-crystal-based metamaterials can support propagating waves with large wave vectors that are evanescent in air or dielectrics, enabling the manipulation of visible light at the subwavelength scale. This property will be rather useful in a number of applications, including waveguiding, imaging and optical communications.

Isotropic photonic metamaterials

As discussed earlier, photonic metamaterials are usually fabricated by well-established 2D fabrication technologies, such as electron-beam lithography, focused ion beam etching, evaporation of metal films, etc. The reported structures have a few layers, with the exception of Zhang's 10-layer fishnet metamaterial structures. However, all of the reported photonic metamaterials exhibit a negative index along a certain propagation direction, essentially making them one-dimensional metamaterials.

On the other hand, isotropic 2D and 3D bulk photonic metamaterial designs with low absorption and high transmission are needed to explore all of the potential applications of photonic metamaterials. Direct laser writing (DLW) is a

[Metamaterials fabricated by direct laser writing]



Top: Electron micrographs of photonic metamaterial structures using direct laser writing. Bottom: A photonic metamaterial that can be grown by laser beam deposition. The structure exhibits a negative index along two directions.

promising technique for fabricating truly 3D large-scale photonic metamaterials. Researchers at the University of Karlsruhe have recently demonstrated the feasibility of this method at near-infrared frequencies assisted with silver chemical vapor deposition.

The samples go through 10 chemical vapor deposition cycles, resulting in an estimated silver thickness of approximately 50 nm. The figure above shows electron micrographs of the structures made by using these methods. The coating is uniform around the structures, even in 3D; this is in sharp contrast to the usual 2D evaporation process. The films are somewhat granular but they are all connected. This crucial aspect is demonstrated by the fact that the silver films exhibit good DC conductivity, by a reflectance of $R > 95\%$ in the 1 to 4 μm wavelength range.

The researchers used retrieval methods to determine the electromagnetic properties of the metamaterials grown by DLW. The magnetic permeability exhibits the anticipated negative values around the 100 THz frequency range. This clearly shows that the fabrication of a magnetic metamaterial using DLW and silver CVD is possible. However, the influence of bi-anisotropy of the fabricated structure results in a positive index of refraction. Therefore, there is a clear need to come up with a better design that is suitable for DLW.

In a new paper, Costas Soukoulis and his colleagues offer an effective solution to this problem. They theoretically analyze and demonstrate the first truly bulk NIM design, which is feasible to fabricate with DLW at around telecom wavelengths. The blueprint design shows the unit cell for a 2D photonic metamaterial consisting of two pairs of SRRs. Magnetic permeability and electric permittivity are simultaneously negative in the region of negative index of refraction.

The operation frequency of the metamaterial is close to the telecom band (150 THz) with a THz bandwidth of about 20. The designed structure has a 2D isotropy in the sense that the propagation is in two directions with fixed polarization and a negative index of refraction. Combined with such new and suitable theoretical blueprints, the DLW and silver CVD techniques can be used to overcome the major hurdles in the field of photonic metamaterials.

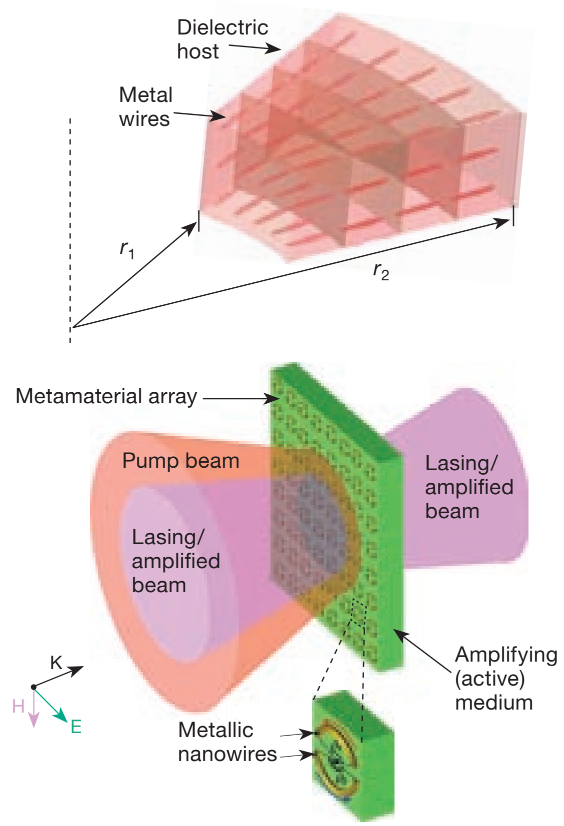
Cloaking at optical frequencies

Early experiments have shown that it is possible to design a metamaterial around an object, such that an incoming wave can be totally reconstructed on the other side of the same object. In a sense, the metamaterial acts like a cloak that makes the object inside invisible to the outside. However, the design used at microwave frequencies cannot be implemented for an optical cloak. In order to achieve cloaking at optical frequencies—which is certainly of particular interest because optical frequencies are where the word "invisibility" is conventionally defined—Vladimir Shalaev suggested the design of a new non-magnetic cloak. It consists of wires, all of which are perpendicular to the cylinder's inner and outer interfaces.

The spatial positions of the nanowires do not have to be periodic. The non-magnetic nature of the new design eases the pain of constructing gradient magnetic metamaterials in three-dimensional space and, therefore, paves the way for the realization of cloaking devices at optical frequencies. The proposed design can be generalized to cloaks with other metal structures, such as chains of metal nanoparticles or thin continuous or semicontinuous metal strips.

The complex refractive index of the metamaterials can only be numerically calculated by applying retrieval methods to the reflectance and transmittance characteristics.

[Blueprint for an invisibility cloak]



Top: A cylindrical cloak consists of wires that are all perpendicular to the cylinder's inner and outer interfaces. The spatial positions do not have to be periodic. Bottom: The "lasing spaser" structure consists of a gain medium slab (green) supporting a regular array of metallic asymmetric split-ring resonators.

It can also be adopted for wavelengths other than the optical spectral ranges, including the infrared and microwave. However, the achievable invisibility with the proposed cloak is not perfect due to the impedance mismatch that is associated with the reduced material specifications and the inevitable loss in a metal-dielectric structure. The optical losses associated with the metallic inclusions have always been pointed out as one of the major limitations of photonic metamaterials. More research is needed to design photonic metamaterials with lower propagation losses. Using active media could be one way to develop lossless photonic metamaterials, where the losses in metallic structures will be compensated by the gain obtained from the active media. Besides cloaking applications, this will also enable a family of new photonic metamaterial devices, including the superlens, where the spatial resolution could be brought close to the ultimate limit stemming from the finite electron velocity—possibly as small as just a few nanometers.

Nikolay Zheludev and his co-workers recently suggested another application for photonic metamaterials. This is called a "lasing spaser" that can create a narrow diversion coherent source of electromagnetic radiation. In-phase plasmonic oscillation in individual resonators leads to the emission of spatially and temporarily coherent light propagating in the direction normal to the array. The lasing spaser allows for the high amplification and lasing in a very thin layer of material with a more modest gain level, making it a very practical proposition.

The thin-layer geometry is a desirable feature for some highly integrated devices and for heat management and integration. The amplification and lasing frequency is determined by the size of the ring and may be tuned to match the luminescence resonances in a large variety of gain media. This, therefore, makes the lasing spaser a generic concept for many applications.

Concluding thoughts

Photonic metamaterials represent one of the most active research areas within optics and photonics. Thanks to recent experimental and theoretical work, the optical magic behind these structures has already been partially unveiled. In the future, researchers will create optical materials with new and unusual properties that will stretch the bounds of our imaginations even further.

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OSA Member **Ekmel Ozbay** (ozbay@bilkent.edu.tr) is with the Nanotechnology Research Center at Bilkent University, in Ankara, Turkey.

[References and Resources]

>> V.G. Veselago. Soviet Physics Uspekhi **10**, 509 (1968).
>> J. Pendry. Phys. Rev. Lett. **85**, 3966 (2000).
>> D.R. Smith et al. Phys. Rev. Lett. **84**, 4184 (2000).
>> R.A. Shelby et al. Science **292**, 77 (2001).
>> E. Cubukcu et al. Nature **423**, 604 (2003).
>> E. Cubukcu et al. Phys. Rev. Lett. **91**, 2140 (2003).
>> S. Linden et al. Science **306**, 1351 (2004).
>> E. Ozbay. Science **311**, 189 (2006).
>> D. Schurig et al. Science **314**, 977 (2006).
>> W. Cai et al. Opt. Express **15**, 3333 (2007).
>> W. Cai et al. Nature Photon. **1**, 224 (2007).
>> G. Dolling et al. Opt. Lett. **32**, 551 (2007).
>> W.V. Shaalev et al. Nature Photon. **1**, 414 (2007).
>> C.M. Soukoulis et al. Science **315**, 47 (2007).
>> N. Liu et al. Nature Materials **7**, 31 (2008).
>> J. Valentine et al. Nature **454**, 7247 (2008).
>> J. Yao et al. Science **321**, 930 (2008).
>> M.S. Rill et al. Nature Materials **7**, 543 (2008).
>> C.M. Soukoulis et al. J. Phys.: Condens. Matter **20**, 304217 (2008).
>> N.I. Zheludev et al. Nature Photon. **2**, 351 (2008).