



A Roadmap for Metamaterials

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Metamaterials have rapidly advanced over the past few years—from being a paradigm for engineering unique electromagnetic properties to forming a material base for functional devices with tuneable, switchable and nonlinear capabilities. In the future, they will allow for dynamic quantum-effect-enabled systems offering exciting applications that we have not yet imagined.

The quick and widespread proliferation of new nanofabrication techniques has opened many opportunities for researchers to create artificial media known as metamaterials, which are designed to control and interact with electromagnetic waves. Research in this field is developing at an incredible pace. In natural solids, optical response is determined by the quantum energy-level structure of the constituent atoms or molecules. By contrast, the electromagnetic properties of metamaterials are derived from the resonant characteristics of the subwavelength plasmonic resonators from which they are constructed.

The metamaterial paradigm is an incredible one that promises groundbreaking new functionalities such as invisibility and imaging with unlimited resolution. It allows the tailoring of boundary conditions, frequency and spatial dispersion of electromagnetic response and thereby leads to applications in slow light and asymmetric transmission devices, polarization control and absorption management. The next stage of development will be the widespread use of active (gain-assisted, controllable and nonlinear) metamaterials and metamaterials for sensing and energy applications.

We anticipate that another radical advance in expanding the metamaterial paradigm will occur when the arrays of classical plasmonic resonators found in today's metamaterials are replaced with arrays of superconducting quantum interference devices to create truly quantum artificial electromagnetic media. Controlling the fabric of "electromagnetic space" (and thus light propagation) with metamaterials that have coordinate-dependent parameters offers additional technological opportunities, which are not feasible with conventional homogeneous optical materials. Known as "transformation optics," this idea offers new solutions for sophisticated lenses, interconnect applications, waveguides and "mirage" (cloaking) devices.

Laser sources

The idea of combining gain media with metamaterials has attracted increasing attention within the research community. This new generation of metamaterials will play a key role in developing novel laser sources.

Indeed, it is now experimentally confirmed that hybridizing a gain medium (semiconductor quantum dots or quantum well structures) with a plasmonic metamaterial can lead to a multi-fold-intensity increase and a narrowing of their photoluminescence spectra. The luminescence

enhancement is a clear manifestation of the quantum Purcell effect, and it can be controlled by a metamaterial's design.

This is an essential step towards the development of metamaterial-enhanced gain media and the “lasing spaser”: a “flat” laser with emission fueled by

plasmonic excitations in an array of coherently emitting metamolecules. In contrast to conventional lasers that operate at wavelengths of suitable natural atomic or molecular transitions, the lasing spaser's emission wavelength can be controlled by metamolecule design.

We envision that, in the future, electrically pumped semiconductor gain media will provide a practical solution for metamaterial-based lasers at visible and telecom frequencies, while quantum cascade semiconductor amplifiers show promise for tackling losses and providing gain in the infrared. Electrically and optically pumped graphene is expected to show strong plasmonic amplification in the terahertz part of the spectrum.

Adding gain to metamaterials also compensates for the joule losses that damp plasmons in metal nanostructures. Lowering losses is crucial for the performance of metamaterial-based negative-index devices, waveguides, spectral filters, delay lines and, in fact, practically any application of metamaterials.

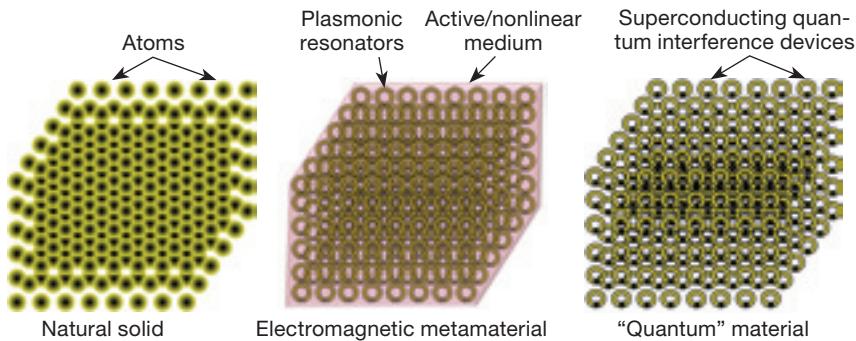
Switchable metamaterials

Switchable and tuneable metamaterials are other rapidly expanding areas of research. Indeed, the development of nanophotonic all-optical data processing circuits depends on the availability of fast and highly responsive nonlinear media that react to light by changing their refractive index and absorption. This is difficult to deliver in nanoscale-size devices using electronic or molecular nonlinearities, where stronger responses often come at the expense of longer reaction times and where the optical path through the nonlinear medium is shorter than the wavelength of light.

When high speed switching is not the prime objective, metamaterials can be reliably and reversibly controlled by microelectromechanical (MEMS) actuators that reposition parts of the metamolecules. This has been convincingly demonstrated for terahertz and far-infrared metamaterials. Reconfigurable optical metamaterials require moving components on the scale of a few tens of nanometers (NEMS actuators) to realize a profound change in optical properties.

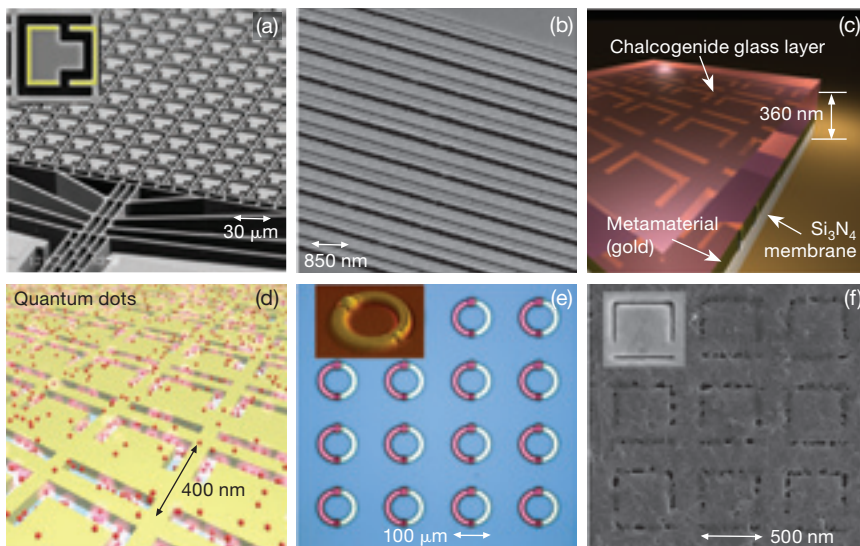
Metamaterials in which metal nanostructures are hybridized with nonlinear and switchable dielectrics or semiconductors provide a way to achieve changes faster than they can be achieved by mechanical repositioning of parts.

[Metamaterials: back to nature]



Today's photonic metamaterials are nanofabricated arrays of classical plasmonic resonators that mimic and surpass the electromagnetic properties of natural solids. Additional functionalities can be achieved by hybridizing these structures with nonlinear, gain and switchable media. In the future, arrays of quantum interference devices will form “quantum metamaterials” that provide a much closer analogy to natural crystals.

[Metamaterials go active and dynamic]



(a) MEMS actuators can reposition parts of the metamolecule (inset) in an array, allowing the tuneability of infrared transmission and reflection spectra. (b) Reconfigurable NEMS photonic metamaterial fabricated on bimorph membranes are tuneable by temperature. (c) Switchable metamaterial with phase-change chalcogenide glass active layer. (d) Optical luminescence of semiconductor quantum dots in a plasmonic array is enhanced manifold. (e) Array of Josephson junction quantum interference devices is a prototype quantum metamaterial. (f) Plasmonic metamaterial array provides an order-of-magnitude enhancement of the ultrafast optical nonlinearity of carbon nanotubes.

Images courtesy of (clockwise from top left) Liu Ai Qun, NTU, Singapore; Bruce Ou, Zsolt Samson, Eric Plum, Roger Buckingham, Andrei Nikolayenko, Univ. of Southampton

Graphene is another favorite that promises to add electro-optical capability to metamaterials, in particular in the IR and terahertz domains, by exploiting the spectral shift of electromagnetic response driven by applied voltage.

This can lead to a strong change in the resonant transmission and reflection of the hybrid. Prime candidates for hybridization with metamaterials are semiconductors and semiconductor multiple-quantum-well structures used as substrates for a metallic framework, carbon nanotubes and fullerenes implanted into the fabric of the metamaterials and organic nonlinear media.

Scientists have already demonstrated the ability to change a metamaterial's response at terahertz frequencies by injection or optical generation of free-carriers into a gallium-arsenide substrate. Recent experiments show that the ultrafast nonlinear response of silicon can be strongly enhanced by adding a metamaterial layer. Single-wall semiconductor carbon nanotubes deposited on metamaterials exhibit an order-of-magnitude higher nonlinearity than the already extremely strong response of the nanotubes themselves, due to a resonant plasmon-exciton interaction.

So-called "phase-change" materials are prime agents for switching: chalcogenide glasses have been used in rewritable optical disk technology for several decades. They provide fast and reproducible changes in optical properties in response to excitation. This functionality is underpinned by phase transitions between crystalline and amorphous states and may be engaged by optical or electrical stimulation: A nanoscale metamaterial electro-optical

switch using chalcogenide glass has been demonstrated.

Similar properties are exhibited by transition metal oxides, in particular vanadium dioxide. In another example, the transition between different metastable phases in polymorphic elemental gallium leads to dramatic change in dielectric and plasmonic properties, making it another candidate for use in switchable metamaterials, alongside with liquid crystals.

A very substantial change in the dielectric properties of a nanometer-thick layer may be achieved in conductive oxides through the injection of free-carriers, which should be enough to control resonant transmission in a hybrid metamaterial. Graphene is another favorite that promises to add electro-optical capability to metamaterials, in particular in the IR and terahertz

domains, by exploiting the spectral shift of electromagnetic response driven by applied voltage. A recent demonstration of the magnetic control of plasmons in layered structures of ferroelectric and noble metals can also be translated to the tuning of metamaterials.

Sensor applications

Sensor applications represent another rapidly growing area in metamaterials research. For instance, asymmetrically split ring resonators supporting high-quality Fano resonances or metamaterial arrays of nanoscale antennas are well suited to detecting low-concentration analytes such as sugar, hydrogen, etc., through variations in their transmission and reflection characteristics. A single molecular layer of graphene, for example, can induce a multifold change in the transmission of a metamaterial. Plasmonic metamaterial nanostructures can also be used to improve light-harvesting solutions, permitting a considerable reduction in physical thickness and improved efficiency in solar photovoltaic absorber layers.

Superconducting and quantum metamaterials

Superconducting metamaterials have recently emerged to offer a radically new paradigm for data processing and information technologies. They will provide a dramatic reduction of losses, accompanied by access to the extreme sensitivity of the superconducting state to external stimuli and the exceptional nonlinearity of superconductors (orders of magnitude higher than *p-n* junctions), enabling low energy switching at the subattojoule level.

Negative dielectric constants and dominant kinetic resistance also make superconductors an intriguing plasmonic media. Moreover, a fundamental change in the nature of information carriers is produced by superconductivity: In some implementations, it will be possible to switch from the classical excitations of conventional plasmonic and metamaterial devices to quantum excitations

[Conventional metamaterials vs. future "quantum metamaterials"]

	Conventional metamaterials (current paradigm)	Superconducting metamaterials (future paradigm)
Building block of the technology	Split ring—classical plasmonic resonator	Josephson quantum-mechanical interference device
Excitation/information carrier	Plasmon	Supercurrent/quantum qubit
Mode of operation	Analog	Quantized ("digital")
Frequency range	From microwave to optical	From microwave to terahertz
Advantages	Simplicity	Tuneability and opening of the quantum horizon: Quantum level of operation, extremely high nonlinearities
Main challenges	Losses and challenging fabrication for applications in the visible part of the spectrum	Sophisticated fabrication and need for cryo-cooling

underpinned by flux quantization and quantum interference effects. Indeed, the classical object of metamaterials research—the ubiquitous split-ring metamolecule—has much in common with the fundamental unit of superconductivity, the Josephson junction ring.

An array of superconducting Josephson rings could be a truly quantum metamaterial, where each metamolecule is a multilevel quantum system supporting phase qubits. However, applications of superconducting metamaterials will be limited to the microwave domain for niobium-based metamaterials, and to the terahertz spectral domain if high-temperature superconductors are used. This is because higher frequencies destroy the superconducting phase.

Researchers have demonstrated the fabrication of metamaterials from niobium films and the use of patterned high-critical-temperature perovskite-related cuprates. However, the manufacture of large metamaterial arrays of Josephson

Another promising approach is membrane projection lithography, which enables the creation of nearly arbitrary metamaterial unit cells with metal inclusions along each of the coordinate axes.

rings is a highly sophisticated process that is not yet widely available. The table on p. 33 presents a comparison between conventional metamaterials that support classical plasmonic excitations and future superconducting metamaterials. Note that the cryo-cooling requirement for superconductors is no longer a serious technological limitation as compact cryo devices are now widely deployed in telecommunications and sensing equipment.

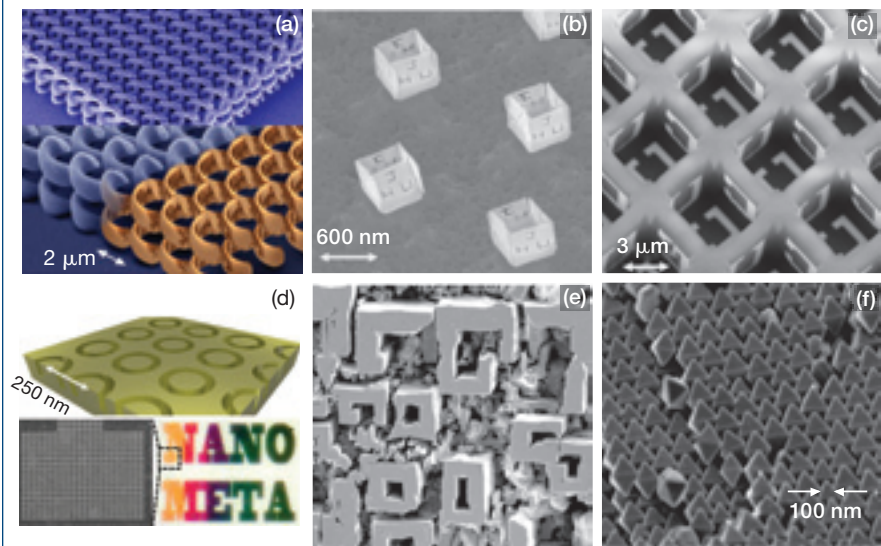
Fabrication

No progress in metamaterials research will be possible without further developments in fabrication. New techniques will be needed to achieve close-to-molecular perfection in nanostructures, and they must be inexpensive. We need to go beyond electron-beam lithography, focused ion beam milling and nanoimprint. Most of the research so far has dealt with quasi-two-dimensional metamaterial structures or few-layer assemblies. The real challenge is to create truly volume metamaterials, and a great deal of inspiring and creative effort is now being concentrated on that. This includes prototyping of metamaterials using sophisticated two-photon resist polymerization techniques followed by metalizing the dielectric framework or directly releasing metallic silver from a silver halide through a two-photon absorption process.

Another promising approach is membrane projection lithography, which enables the creation of nearly arbitrary metamaterial unit cells with metal inclusions (such as split-rings) along each of the coordinate axes. Recently introduced three-dimensional indented (“intaglio”) or raised (“bas-relief”) subwavelength “continuous-metal” metamaterials offer interesting opportunities for controlling the electromagnetic properties of surfaces. They may easily be manufactured by focused ion-beam milling or high-throughput nanoimprint lithography. The fabrication of complex volume metamaterial structures also opens radical new opportunities such as the exploitation of intriguing toroidal electromagnetic excitations.

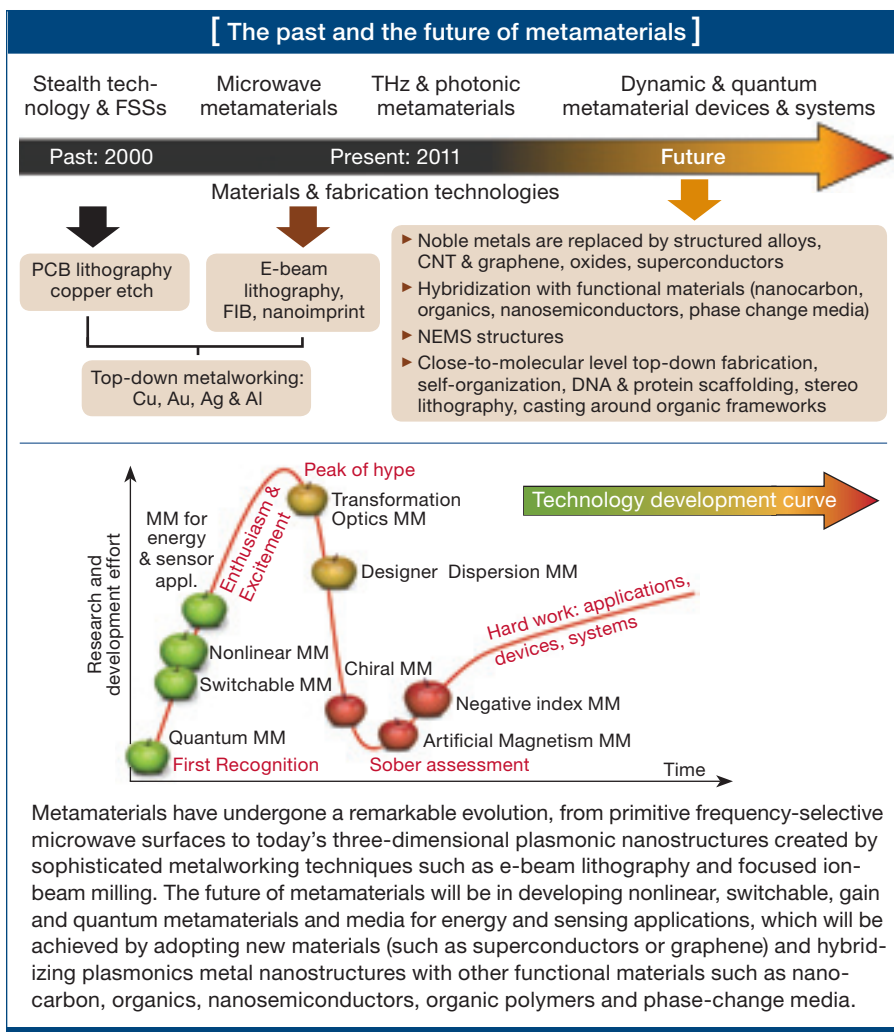
Some research groups are exploring self-assembly strategies for fabricating metamaterials. For instance, one may overcome the limitation of inherently two-dimensional lithographic processes by transforming prepatterned lithographic templates into mechanically robust and precisely patterned three-dimensional nanoscale structures. This can be done by curving, rotating, aligning and bonding them using forces derived from a minimization of surface area of liquefying or coalescing metallic grains.

[3-D metamaterials and new approaches to fabrication]



(a) Chiral metamaterial fabricated by direct laser writing. (b) Arrays of 3-D metamolecules self-assembled with lithographically defined “hinged” surface patterns. (c) Infrared metamaterial defined by projection lithography on the walls of a dielectric cage structure. (d) A new type of “Intaglio” 3-D metamaterial structure milled into a gold surface by a focused ion beam changes the color of the metal. (e) Split-ring metamaterial-like structure fabricated by directional solidification of eutectics. (f) Self-assembled plasmonic colloidal nanocrystal arrays.

Images courtesy of (clockwise from top left) Andre Radke and Harald Giessen, Univ. of Stuttgart and Martin Wegener, Univ. of Karlsruhe; David Gracias, Johns Hopkins Univ.; Bruce Burckel and Igal Brener, Sandia Labs; Kevin Macdonald, Univ. of Southampton; Dorota Pavlak, IEM; Peidong Yang, UC Berkeley



The directional solidification of eutectics leads to the formation of wire-like and split-ring-like structures. This has been engaged for the manufacture of metamaterial-like composite solids at a size scale that is suitable for the infrared, terahertz and microwave metamaterials. However, real progress in photonic metamaterials will require new techniques that occupy a position between chemical processes controlled by self-organizing forces on the truly molecular level and the less accurate, top-down methods of metamaterial metalworking, which have the advantage of being able to build metamaterials to almost any blueprint. They will be expected to work with a variety of materials.

These techniques may include bioengineering approaches such as DNA scaffolding or protein-driven

crystallization for fabricating nanoparticle superlattices, the self-assembly of semiconductor quantum dots and magnetic nanocrystals or the casting of metal nanostructures around liquid crystalline frameworks and colloidal frames. Moreover, even metals themselves can

be replaced as agents of conductivity by self-assembled carbon nanotubes or patterned graphene, allowing for stretchable and flexible metamaterials. Some oxides, semiconductors and perovskites can be used for infrared applications.

Shifting paradigms

Metamaterials were initially perceived as a means of developing new and unusual electromagnetic properties (such as a negative index of refraction) via structuring, assuming continuous ("analog") and linear regimes of metamaterial response. In other words, we thought of metamaterials as, first and foremost, materials.

The current trend is to think of metamaterials as devices, where the structuring of metal and the hybridization with functional agents brings new functionality and response becomes tuneable, switchable or nonlinear. In the near future, we will be able to enter the field of quantum metamaterials. Moreover, by exploiting the concept of transformation optics, metamaterials with spatially variable parameters and active metamolecular switches imbedded in the strategic location will allow combining complex quantum-level switching and memory functions with the waveguiding of electromagnetic radiation across the body of a metamaterial volume. Rather than materials or devices, we will begin to think of metamaterials as dynamic systems. ▲

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