

From metamaterials to metadevices

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Metamaterials, artificial electromagnetic media that are structured on the subwavelength scale, were initially suggested for the negative-index 'superlens'. Later metamaterials became a paradigm for engineering electromagnetic space and controlling propagation of waves: the field of transformation optics was born. The research agenda is now shifting towards achieving tunable, switchable, nonlinear and sensing functionalities. It is therefore timely to discuss the emerging field of metadevices where we define the devices as having unique and useful functionalities that are realized by structuring of functional matter on the subwavelength scale. In this Review we summarize research on photonic, terahertz and microwave electromagnetic metamaterials and metadevices with functionalities attained through the exploitation of phase-change media, semiconductors, graphene, carbon nanotubes and liquid crystals. The Review also encompasses microelectromechanical metadevices, metadevices engaging the nonlinear and quantum response of superconductors, electrostatic and optomechanical forces and nonlinear metadevices incorporating lumped nonlinear components.

For some years the metamaterials paradigm has mostly been considered as a means of engineering the electromagnetic response of passive micro- and nanostructured materials by engaging resonance excitations such as localized plasmonic modes. Remarkable results have been achieved in this way: they include, for instance, negative-index media that refract light in the opposite direction from that of conventional materials, chiral materials that rotate the polarization state of light hundreds of thousands of times more strongly than natural optical crystals, and structured thin films with remarkably strong dispersion that can slow light in much the same way as resonant atomic systems with electromagnetically induced transparency.

The ever-increasing demand for faster information transfer and processing drives efforts to remove the bottleneck between fibre-based optical telecommunication networks and electronic data handling and routing, improving data storage and developing parallel data processing operating in a compact space. Fulfilment of these tasks will require strong and fast nonlinearities for switching light with light, and much improved control of the electromagnetic properties of matter with external stimuli such as electric signals.

In this Review we illustrate that many of these functionalities may be greatly enhanced by hybridizing functional matter with metamaterials, by exploiting nonlinearity of the metamaterial framework itself, and by taking advantage of the changing balance of forces in systems with building blocks smaller than the wavelength of light. This leads to the concept of metadevices, a logical extension of the metamaterial paradigm, where interactions are nonlinear and responses are dynamic. Here we envisage that the future platform for highly integrated electromagnetic signal processing and distribution will emerge that will combine nonlinear, memory and switchable functionalities with transformation optics' ability to guide light via the engineered electromagnetic space, using metamaterials with spatially variable parameters.

Reconfigurable metadevices

Active tunability and switching of electromagnetic characteristics of metamaterials can be achieved by altering the shape of individual metamolecule resonators, or by manipulating the near-field interactions between them. The latter can be attained by changing

the relative position of rows of the metamolecular structure¹ or by displacing arrays of metamolecules forming a three-dimensional metamaterial lattice² (Fig. 1a).

The potential of microelectromechanical systems (MEMS) for electromagnetic metamaterials was initially recognized for the tuning of transmission lines^{3,4}. Shortly after the first publications on MEMS filters, microelectromechanical actuators were applied to reconfigure metamolecules⁵, in the expectation that controlling the resonant properties of the individual elements could be used to make tunable negative-refractive-index metamaterial arrays. Reconfigurable metamaterials at terahertz frequencies were first produced by fabricating planar arrays of split-ring resonators on bimaterial cantilevers designed to bend out of plane in reaction to a thermal stimulus (Fig. 1b). A marked change of the electric and magnetic response was observed as the split-ring resonators synchronously reoriented within their unit cells⁶. Similar thermally activated structures may be used in infrared and terahertz detectors. A number of terahertz metamaterial designs with electrically activated MEMS switches have since been demonstrated^{6–8}. One of the most elaborate designs is an array of pairs of asymmetric split-ring resonators, one fixed to the substrate and the other patterned on a movable frame⁹ (Fig. 1c). Here the reconfigurable metamaterial and the supporting structures (microactuators, anchors, supporting frames and so on) are fabricated on a silicon-on-insulator wafer using deep reactive-ion etching. By adjusting the distance between the two rings using the micromachined actuators, the strength of dipole-dipole coupling can be tuned continuously, allowing efficient tailoring of the electromagnetic response. The reconfiguration of metamolecules also allows switching of polarization eigenstates of this anisotropic metamaterial.

Terahertz metamaterial structures that use flexing microelectromechanical cantilevers to tune the resonance frequency were suggested recently¹⁰. The cantilevers are coated with a magnetic thin film that can be actuated by an external magnetic field, enabling continuous control of the resonance frequency over a large frequency range¹⁰. Manufacturing arrays of plasmonic resonators on flexible, stretchable polymer substrates offers a practical way to dynamically tune the response of photonic metamaterials^{11–13}, and also allows them to be fabricated on curvilinear shapes

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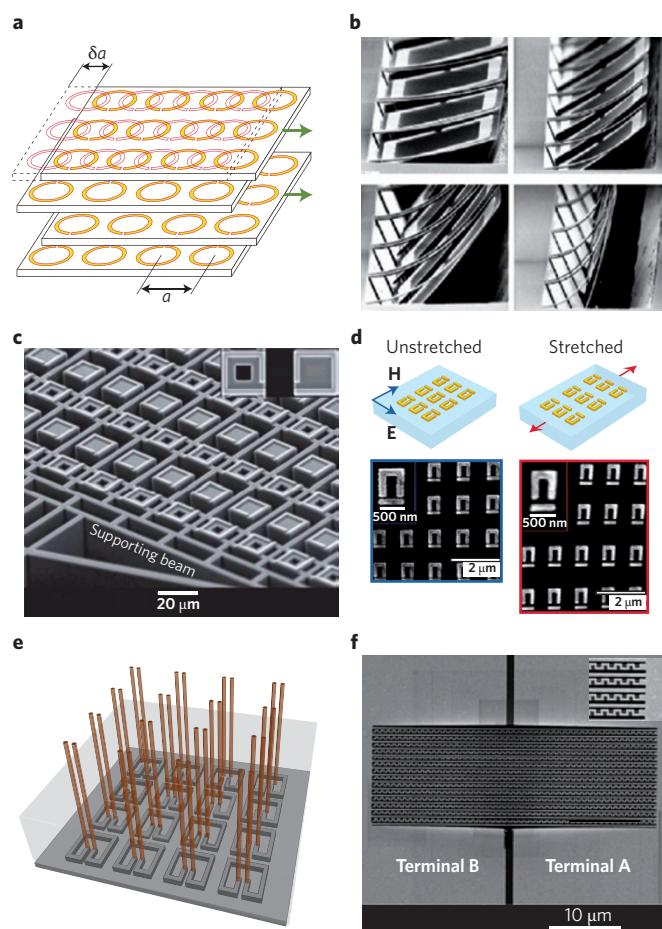


Figure 1 | Reconfigurable metamaterials. **a**, A metamaterial that is tuned by shifting the metamolecular planes of the lattice by δa relative to each other (artist's impression)². **b**, Arrays of metamolecules of a terahertz metamaterial are switched from one configuration to the other by thermal activation (scanning electron micrograph, SEM)⁶. **c**, Terahertz metamaterial can be dynamically tuned by manufacturing the metamaterial array on a MEMS-driven silicon platform⁹. Inset: SEM showing the metamolecule consisting of static and movable split rings. **d**, Photonic metamaterials manufactured on polymer films can be tuned by manipulation of resonator components by stretching the substrate¹¹. **e** (**H**) shows the direction of the incident electric (magnetic) field. **e**, Schematic impression of a microwave metamaterial that can be switched by injecting liquid metallic mercury into the capillary array in the shape of split-ring resonators¹⁴. **f**, Plasmonic metamaterials manufactured on dielectric strings cut from a silicon nitride membrane can be driven electrostatically to megahertz frequencies¹⁵. The inset is an SEM that shows a fragment of the array at the terminal end. Figure reproduced with permission from: **a**, ref. 2, © 2009 AIP; **b**, ref. 6, © 2009 APS; **c**, ref. 9, © 2011 Wiley; **d**, ref. 11, © 2010 ACS; **e**, ref. 14, © 2009 AIP; **f**, ref. 15, © 2012 OSA.

(Fig. 1d). Microfluidics can also be used to reconfigure microwave metadevices when a conductive liquid (such as mercury) is injected into the network of metamolecules, changing their electromagnetic spectra¹⁴ (Fig. 1e).

Engineering dynamically reconfigurable photonic metamaterials with metamolecular features on the scale of tens of nanometres is a formidable technological challenge. However, working on the nanoscale also has some important advantages. The elastic forces and the forces of inertia acting on metamaterial elements scale differently with size in such a way that the mechanical frequencies of the system reach high values for submicrometre structures. Moreover, the electrostatic force, which is inversely proportional to

the distance, becomes the dominant force at the nanoscale: potential differences of only a few volts induce a force that can overcome the elastic response of a metamaterial framework. A photonic metamaterial driven by electrostatic forces¹⁵ has been developed, consisting of a gold plasmonic nanowire pattern fabricated on a dielectric membrane (Fig. 1f). Operating in the optical telecommunication range of wavelengths, it can be used as a megahertz-bandwidth modulator consuming only a few microwatts of power and can also perform non-volatile switching, providing high-contrast transmission change.

It is clear that the technological solutions developed in MEMS, NEMS and micro/nanofluidics technologies will have considerable impact on metadevices in the future. The mechanical oscillation frequencies of nanoscale components could be in the gigahertz range, allowing properly engineered arrays of metamolecules based on subwavelength-sized cantilevers to be driven at high bandwidth. In some applications this approach can directly compete with electro-optical modulators, simultaneously offering low-voltage operation and lending itself to high-density integration.

Electro-optical metadevices

An active metadvice capable of efficient real-time control of radiation with electric signals was first developed for the terahertz part of the spectrum¹⁶. It consisted of a gold metamaterial array fabricated on a semiconductor substrate. The array and substrate together effectively formed a Schottky diode, where dielectric properties of the substrate can be controlled by injection and depletion of carriers. An electric signal applied to the metamaterial affects the high-frequency conductivity of the substrate in critical areas near the metamolecules and thus affects their resonant response (Fig. 2a). This approach allows modulation of the terahertz transmission by 50%. These types of hybrid metamaterial devices offer frequency and modulation bandwidth potentially up to 10 MHz (ref. 17). A multi-pixel 4×4 voltage-controlled spatial modulator for terahertz beams has been developed using this type of active terahertz metamaterials (Fig. 2b). In the modulator each pixel is an array of subwavelength-sized split-ring resonator elements fabricated on a semiconductor substrate, and is independently controlled by applying an external voltage. The spatial modulator has a uniform modulation depth of around 40% and negligible crosstalk at the resonant frequency. It can operate at room temperature under small voltages, with low power consumption¹⁸. Researchers have also demonstrated that carrier photogeneration in the silicon substrate supporting a chiral terahertz metamaterial can lead to a switching of its optical activity in the form of reversed circular dichroism¹⁹.

A very substantial change in the dielectric properties of a nanometre-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 metamaterial²⁰. Ferroelectrics can also be engaged in tuning a metamaterial response²¹. Graphene (Fig. 2c) is another favourite for constructing metamaterials^{22,23} with electro-optical capability, in particular in the infrared and terahertz domains, by exploiting the modification of the electromagnetic response by an applied voltage²⁴. Such a terahertz electro-optical modulator, consisting of engineered graphene microribbon arrays, was recently demonstrated (Fig. 2d)²⁴. The graphene plasmon resonances were reported to have remarkably large oscillator strengths, resulting in prominent room-temperature optical absorption peaks. Moreover, the graphene's response can be tuned over a broad terahertz frequency range by electrostatic doping.

The main advantage that metamaterial technology can bring to electro-optical modulation is achieving deep modulation in thin, often subwavelength, metadevices. In many cases such metadevices can operate at low voltages, which is clearly a competitive advantage over conventional technology exploiting bulk and expensive electro-optical crystals.

Liquid-crystal metadevices

Tunability and a strongly nonlinear response can be achieved in metamaterials by infiltrating them with liquid crystals²⁵. Electrical control of negative permeability in microwave metamaterials infiltrated with nematic liquid crystals was experimentally demonstrated for a periodic array of split-ring resonators²⁶, showing a reversible change of the transmission resonance with a maximum shift of about 210 MHz (Fig. 2e). A similar approach was later applied to demonstrate the tunability of wire-pair²⁷ and fishnet²⁸ microwave metamaterials, where the external electric field changes the orientation of infiltrated molecules, leading to an effective index variation within the negative-index regime. In a similar way, the tunability can be achieved with magnetic field²⁹.

Mastering control in the near-infrared and optical regimes is a much harder task, but thermal³⁰ and ultraviolet-irradiation-induced tunability of optical metamaterials with liquid crystals has been shown experimentally³¹. Light-induced control of fishnet metamaterials infiltrated with liquid crystals was also achieved recently³² using a metal–dielectric (Au–MgF₂) sandwich nanostructure on a glass substrate infiltrated with a nematic liquid crystal (Fig. 2f). In such a device the transmission can be modulated up to 30% by both the electric voltage and incident optical power at the telecommunications wavelength of 1,550 nm.

Liquid crystals are a robust, proven and affordable technology offering a highly practical solution for controlling metamaterial devices when ambient temperatures and speed of operation are not critical issues, given that in most liquid crystals the response relaxation time is in the millisecond ballpark.

Phase-change metadevices

A radical change in the arrangement of atoms is called a structural phase transition, or phase change. Phase-change functionality of semiconductor chalcogenide glass has been used for decades in optical compact disks and DVDs, where the rewritable memory function is underpinned by a transition from amorphous to crystalline phase. Phase-change functionality in polymorphic metals can also provide a way to achieve nanoscale optical and plasmonic switching devices that can be fast and require little energy to activate³³. Depending on the regime of stimulation and confinement of the active medium, phase changes can be either reversible or irreversible.

The first phase-change nanocomposite material for nonlinear optics and nonlinear plasmonics was created by grain-boundary penetration of gallium into the network of domains of an aluminium film³⁴. Here, continuous and reversible changes occur through the intermediate coexistence of two different phases of gallium³⁵. The change may be induced in a few picoseconds, and it relaxes back on a timescale of microseconds or nanoseconds. This optically and temperature-driven composite metamaterial forms a mirror-like interface with silica and shows an exceptionally broadband phase-transition-based switching response to optical excitation. It operates from the visible to near-infrared part of the spectrum and exhibits ~20% reflectivity change at optical fluence of about 1.5 mJ cm⁻² with sub-100-ns response time.

Another important example of a phase-change medium is vanadium dioxide (VO₂), which shows a phase transition of a percolative nature in which 5–10-nm metallic puddles emerge and grow in the insulating host (Fig. 3a). It has attracted considerable attention as an active medium for hybrid metamaterial structures^{36–39}. Hybridizing vanadium dioxide with a metamaterial shows 20% temperature-activated tuning of the transmission in the terahertz range. Similar switching has also been demonstrated in the near-infrared using a dual-bar gold metamaterial array³⁸. A form of electrically activated memory function and persistent frequency tuning of a metamaterial, which allows lasting modification of its response by using a transient stimulus, have also been demonstrated in a hybrid VO₂ metadvice in the terahertz part of the spectrum⁴⁰.

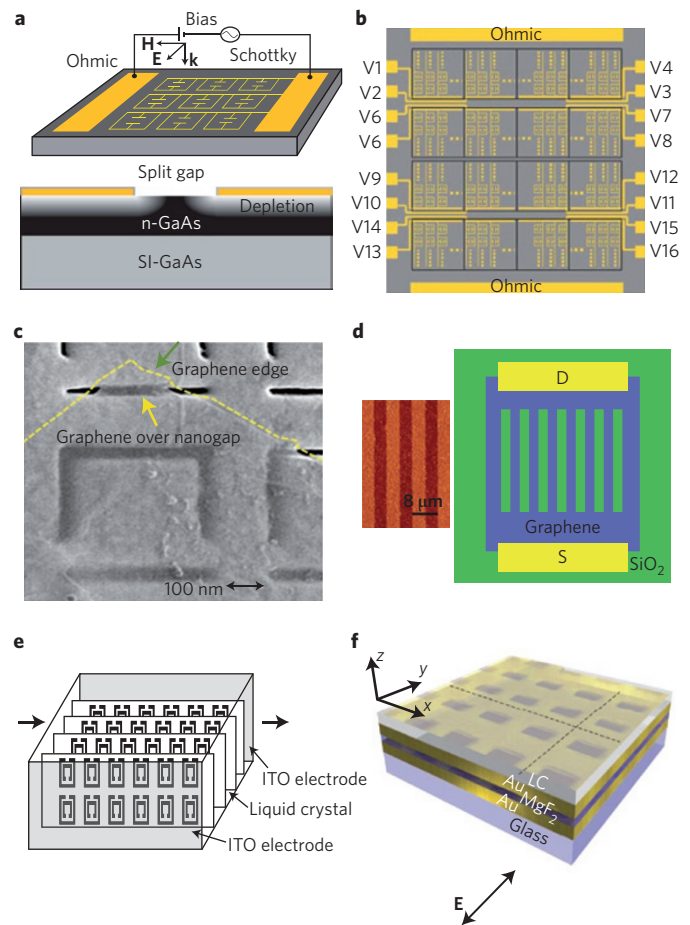


Figure 2 | Electro-optical and liquid-crystal metadevices. **a**, A terahertz metamaterial modulator fabricated on a semiconductor substrate (artist's impression) can be controlled by injection and depletion of carriers in response to an electric bias¹⁶. A vertical cross-section of the metamaterial structure near the resonator gap is shown below. **b**, Terahertz spatial light modulator based on metamaterial array elements that are individually addressable by electrical signals¹⁸. **c**, Graphene covering a metamaterial strongly modifies its plasmonic spectrum. A helium ion microscope image shows a fragment of the gold nano-slit array partially covered by graphene^{22,71}. **d**, Right: A terahertz electro-optical modulator consisting of graphene microribbons with electrical terminals on a dielectric substrate (artist's impression)²⁴. Left: An atomic force microscope image of the graphene array. S and D are the source and drain terminals. **e**, A negative-permeability microwave metamaterial consisting of an array of split rings infiltrated with nematic liquid crystals (artist's impression) can be continuously and reversibly adjusted by an applied electric field²⁶. ITO, indium tin oxide. **f**, Optical nonlinearity of photonic fishnet metamaterials (artist's impression) infiltrated with nematic liquid crystal (LC). The material's optical properties can be tuned by an electric field³². Figure reproduced with permission from: **a**, ref. 16, © 2006 NPG; **b**, ref. 18, © 2009 AIP; **c**, ref. 71, © 2011 AIP; **d**, ref. 24, © 2011 NPG; **e**, ref. 26, © 2007 AIP; **f**, ref. 32, © 2012 AIP.

Combining the phase-change technology of chalcogenide glass semiconductors with metamaterials is a promising direction that offers high-contrast, near-infrared, electronically and optically addressable gating and switching. Its technological importance lies in the wide availability of chalcogenide glass production for optical data storage and the potential of integration with the future silicone and chalcogenide glass photonics technologies. Switching has been demonstrated by exploiting the frequency shift of a narrowband Fano resonance mode of a plasmonic planar metamaterial that

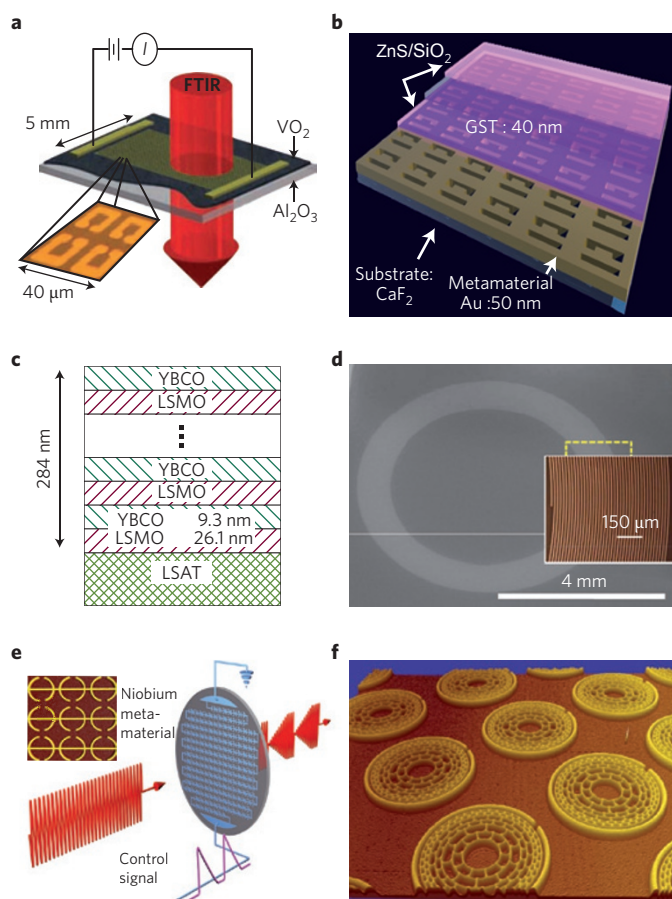


Figure 3 | Phase-change and superconducting metadevices. **a**, Metal-oxide memory hybrid-metamaterial device exploiting the temperature-driven phase transition of a vanadium oxide film (artist's impression)⁴⁰. FTIR, Fourier transform infrared spectroscopy. **b**, Structure of the metamaterial all-optical switch using phase-change chalcogenide glass GST: the hybrid device consists of a planar gold plasmonic metamaterial on a silicon nitride membrane, covered with chalcogenide glass⁴¹. **c**, Negative-refraction, millimetre-wave metamaterial nanostructure based on ferromagnet-superconductor superlattice⁵². YBCO, yttrium barium copper oxide; LSMO, lanthanum strontium manganese oxide. **d**, A spiral metamolecule made from superconducting niobium is about 700 times smaller than its resonance wavelength⁵³. **e**, Controlling sub-terahertz transmission of niobium superconducting metamaterial with electric current running through the network of metamolecules (inset shows optical image of the network of metamolecules connected by the control wire)⁵⁵. **f**, Millimetre-wave quantum metamaterial exploiting the flux exclusion effect in superconductors manufactured from high-critical-temperature superconductor YBCO (profile image)⁶¹. Figure reproduced with permission from: **a**, ref. 40, © 2009 AAAS; **b**, ref. 41, © 2010 AIP; **c**, ref. 52, © 2005 APS; **d**, ref. 53, © 2011 IEEE; **e**, ref. 55, © OSA; **f**, ref. 61, © 2012 NPG.

was induced by a change in the dielectric properties of an adjacent 200-nm-thick film of chalcogenide glass⁴¹. The material used was a new gallium lanthanum sulphide chalcogenide glass, which was bistable and silicon-on-insulator compatible. An electrically stimulated transition between amorphous and crystalline forms of the glass brings about a 150-nm shift in the near-infrared resonance, providing transmission modulation with a contrast ratio of 4:1 in a layer of subwavelength thickness. One of the advantages of this technology is that devices may be structurally engineered to operate at any wavelength throughout the visible and infrared spectral range down to 11 μm . Recently, fully reversible bidirectional optically activated switching has been demonstrated in a gold plasmonic nanostructure

combined with a conventional germanium–antimony–tellurium (GST) chalcogenide glass⁴², making this technology compatible with real-world photonic applications (Fig. 3b).

Phase-change technology is gaining recognition in photonics applications. When non-volatile switching is required, its combination with metamaterials offers high contrast switching response with response times as low as nanoseconds and beyond⁴³ in devices of subwavelength thicknesses.

Superconducting metadevices

Negative dielectric constants and the dominant kinetic resistance make superconductors an intriguing plasmonic medium⁴⁴. Applications of superconducting metamaterials are, however, 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 microwave and terahertz metamaterials where superconductors replace metals in conventional metamaterial designs. This includes the use of niobium⁴⁵ and patterned high-critical-temperature perovskite-related cuprates and niobium nitride^{46–51}. Negative refraction in a multilayer stack of ferromagnetic and superconducting thin films (Fig. 3c) has also been demonstrated⁵². Because a niobium thin film shows low losses at cryogenic temperatures, it allows the development of a metamaterial with extremely compact metamolecules that are as small as 1/658 of the free-space wavelength and have resonances with quality factors in excess of 5,000 (ref. 53; Fig. 3d).

Tuning of metamaterial resonances by temperature and external magnetic field is easy to achieve^{45,47,50,51,54}. A superconducting metamaterial in the form of an interlinked network of subwavelength resonators can also be dynamically controlled by passing electrical current through it⁵⁵: using a niobium metamaterial it was possible to achieve 45% intensity modulation at a carrier frequency of 100 GHz (Fig. 3e). The mechanism underpinning this functionality is a combination of the suppression of superconductivity by magnetic field and heat created by the control current. Superconducting metamaterials can be used in temperature-controllable slow light devices⁵⁶ and other applications⁵⁷.

Moreover, in superconducting metamaterials, it will be possible to switch from the plasmonic excitations of conventional metamaterial devices to quantum excitations underpinned by flux quantization and quantum interference effects. Indeed, the iconic object of metamaterials research — the ubiquitous split-ring metamolecule — has much in common with the fundamental superconductive element, the Josephson junction ring. An array of Josephson rings could be a truly quantum metamaterial, where each metamolecule is a multilevel quantum system supporting phase qubits^{58–60}. However, Josephson junction devices require extremely high-quality nanofabrication and sub-kelvin temperatures. It was recently suggested⁶¹ that a much simpler quantum superconducting metamaterial could be constructed that would exploit the magnetic flux quantization for switching, but would not require Josephson junctions (Fig. 3f). This metamaterial is an array of split-ring resonators enclosing a nest of superconducting rings⁶¹. To achieve a quantum regime of switching, it exploits the quantum exclusion of the oscillating magnetic field penetrating the superconducting rings when a magnetic field is generated by the current in the outer split ring driven by the incident wave.

A superconducting metamaterial can also be used to control static magnetic fields^{62–64}. An anisotropic magnetic metamaterial consisting of an array of superconducting plates can be used for non-intrusive screening of weak d.c. magnetic fields. Moreover, a purposely designed cylindrical superconductor–ferromagnetic bilayer can cloak uniform static magnetic fields⁶⁵.

Superconducting metamaterials offer a radically new base for data processing and quantum information technologies.

Superconductors not only provide far lower losses, but also allow access to the extreme sensitivity of the superconducting state to external stimuli such as heat, electric and magnetic fields, light, current and mechanical stress. The cryo-cooling requirement is no longer a serious technological limitation as compact cryo-devices are now widely deployed in telecommunications and sensing installations.

Ultrafast photonic metadevices

Metamaterials in which metal nanostructures are hybridized with nonlinear and switchable dielectric or semiconductor layers and controlled by ultrafast optical pulses provide much faster switching than is attainable with MEMS/NEMS repositioning of parts, phase-change or voltage-driven carrier injection, liquid crystals or superconductivity modulation. A change in the refractive index or absorption in the layer adjacent to a plasmonic metamaterial array induced by the intense light modifies the plasmon spectrum of the nanostructure. 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 graphene implanted into the fabric of the metamaterials.

The interaction of ultrafast optical pulses with metamaterial was initially studied for optical modulation of their terahertz responses: shunting the capacitive region of the metallic split-ring network by injecting optical carriers into the supporting ErAs/GaAs superlattice leads to a deep modulation of terahertz transmission characteristics of the planar metamaterial, with recovery time on the picosecond scale^{66,67}.

In the optical part of the spectrum the plasmonic resonance field enhancement created by the metamaterial network may be used to enhance the nonlinear response of the adjacent dielectric or semiconductor layer^{68,69} (Fig. 4a). A threefold improvement of pump–probe response was observed in the near-infrared part of the spectrum in a fishnet metamaterial manufactured on an α -silicon substrate exhibiting up to 60% light-induced modulation at an excitation fluence of about 0.5 mJ cm^{-2} . Here the relaxation of nonlinearity is controlled by the electron relaxation time and happens within 2 ps (ref. 68).

Semiconductor carbon nanotubes are highly nonlinear media in their own right, where nonlinearity is associated with the optical saturation of excitonic transitions. Hybridization of single-walled carbon nanotubes with plasmonic metamaterials makes a photonic medium with an exceptionally strong ultrafast nonlinearity operating in the regime of plasmon–exciton coupling⁷⁰ (Fig. 4b). More than tenfold enhancement of the nonlinearity of nanotubes was achieved in a plasmonic nanostructure supporting Fano-type resonances, so a fluence of only $40 \mu\text{J cm}^{-2}$ can create a 10% optical modulation with relaxation time less than 500 fs.

If the nonlinearity of carbon nanotubes has an essentially resonant nature, graphene is highly attractive as a medium with extremely broadband and fast nonlinear response. Unfortunately the achievable nonlinear transmission changes of atomic-thickness graphene films (which absorb only about 2% of incident light) are very small, in the region of 10^{-4} , even at intensities close to optical breakdown. The broadband nonlinear optical response of graphene can, however, be resonantly enhanced by more than an order of magnitude through hybridization with a plasmonic metamaterial⁷², while retaining an ultrafast response time of about 1 ps (ref. 71). Transmission modulation close to 10% has been seen at a pump fluence of $30 \mu\text{J cm}^{-2}$ (ref. 71). This approach allows the engineering of graphene's nonlinearity at a prescribed wavelength within a broad wavelength range, enabling applications in optical switching and pulse shaping.

In terms of data-processing applications, one of the most important figure of merit is the product of the fluence necessary to

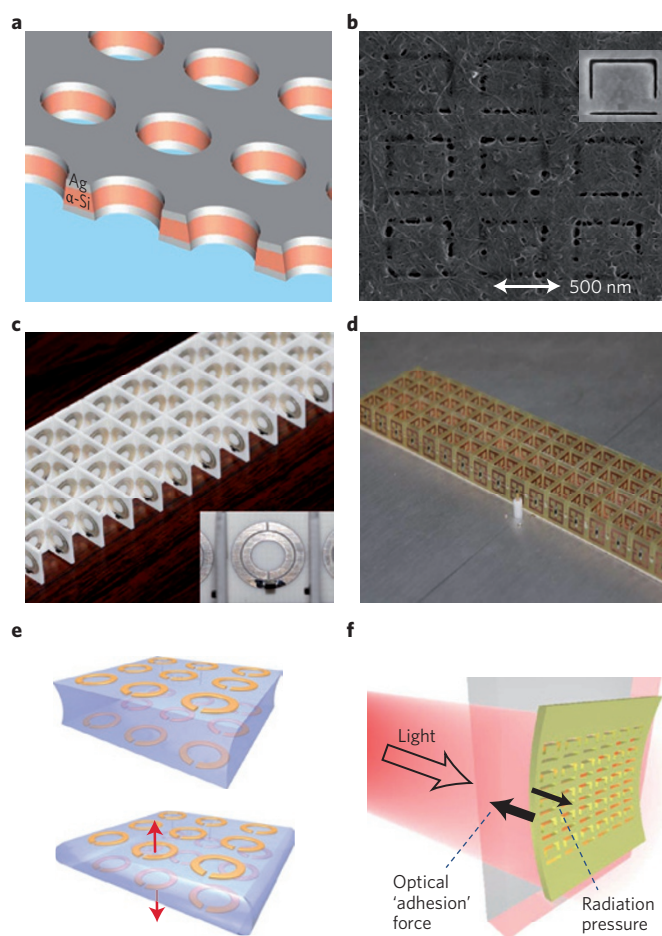


Figure 4 | Ultrafast metadevices, varactor metamaterials and electromagnetic forces.

a, A fishnet photonic metamaterial with sub-picosecond optical response exploits fast carrier dynamics in the α -Si substrate⁶⁹. **b**, A plasmonic metamaterial functionalized with carbon nanotubes shows strong sub-picosecond exciton–plasmon nonlinearity in the near-infrared part of the spectrum⁷⁰. **c**, A nonlinear magnetic metamaterial with lumped varactor diodes embedded in the metamolecules⁷⁸. Inset: a unit cell of the metamaterial before deposition of carbon nanotubes. **d**, Nonlinear electric metamaterials with varactor diodes⁸⁰. **e**, A magnetoelastic metamaterial⁹⁹ is driven by the Ampère's force between excited metamolecules. Artist's impression of a metamaterial slab before (top) and after (bottom) illumination with light (the red arrows indicate the electromagnetic forces acting between the metamaterial elements). **f**, Optical gecko toe: artist's impression of a metamaterial film attracted by a beam of light to a dielectric surface¹⁰². Figure reproduced with permission from: **a**, ref. 69, © 2009 ACS; **b**, ref. 70, © 2010 APS; **c**, ref. 78, © 2008 OSA; **d**, ref. 80, © 2009 AIP; **e**, ref. 99, © 2012 NPG; **f**, ref. 102, © 2012 APS.

create an acceptable modulation contrast and the recovery time of response. Remarkably, in the optical part of the spectrum the metal metamaterial framework itself provides a source of nonlinearity that delivers one of the best performances. Indeed, metamaterial nanostructuring of a thin gold film increases the two-photon interband resonant absorption nonlinearity 300-fold, creating arguably one of the fastest and brightest nonlinear optical medium currently known⁷². Moreover, depending on the spectral position with respect to the plasmonic resonance, nonlinearity may change sign, thus switching from nonlinear dissipation to enhanced transmission. At a fluence of about $270 \mu\text{J cm}^{-2}$, modulation of up to 40% can be achieved with a recorded relaxation time that has a fast component

of about 100 fs (ref. 72). Enhanced nonlinearity of a gold metamaterial has been used to demonstrate a giant effect of intensity-dependent polarization rotation that is millions of times stronger than previously observed in natural crystalline materials⁷³.

In metals, optical excitation close to the edge of the Fermi level also offers a strong, but more slowly relaxing, nonlinear response. In this regime a metal nanowire metamaterial provides a 1-ps relaxation time with a good modulation contrast⁷⁴ at fluence of 7 mJ cm^{-2} .

Strong and fast nonlinearities are needed for controlling light with light because, according to the fundamental Huygens superposition principle, light beams travelling in a linear medium will pass through one another without mutual disturbance. Indeed, the field of photonics is based on the premise that controlling light signals with light requires intense laser fields to facilitate beam interactions in nonlinear media, where the superposition principle can be broken. This premise was recently challenged with help of a plasmonic metamaterial⁷⁵: it was demonstrated that two coherent beams of light of arbitrarily low intensity can interact on a metamaterial layer of nanoscale thickness in such a way that one beam modulates the intensity of the other. The interference of beams can eliminate the plasmonic Joule losses of light energy in the metamaterial or, in contrast, can lead to almost total absorption of light. Applications of this phenomenon may lie in ultrafast all-optical pulse-recovery devices, coherence filters and terahertz-bandwidth light-by-light modulators.

Enhancement of ultrafast nonlinearities with metamaterials arguably offers the brightest and fastest nonlinear media with potential ground-breaking applications for terahertz-rate all-optical data processing as well as ultrafast optical limiters and laser saturable absorbers.

Nonlinear metadevices with varactors

Tuning metamaterials by taking advantage of the nonlinear response of lumped elements integrated into metamolecules of left-handed metamaterials was first suggested theoretically^{76,77} and later implemented experimentally^{78,79}. A similar approach has been explored for other types of metamaterial systems^{17,80–82}. For instance, resonance of the split-ring metamolecule can be controlled by adding the capacitance of a varactor diode in series with the distributed capacitance of the resonator when placed at a point of maxima in the electric currents^{83,84}. At low powers a split ring with embedded varactor exhibits nonlinearity of the second and third order^{85,86}, whereas at higher powers the nonlinear response becomes multivalued or bistable^{84,87}.

An example of a nonlinear magnetic metamaterial operating at microwave frequencies is shown in Fig. 4c. Here the varactor diodes are placed in each element of the composite structure^{83,87}. By selecting the operating frequency to be near resonance, one can dynamically change the transmission properties of the metamaterial, for instance from opaque to transparent, by varying the input power. If a point-like dipole source is placed near the metamaterial, the metamolecules closer to the source will experience stronger fields, making the metamaterial more transparent, which can be seen as beaming of radiation emerging from the metamaterial⁷⁸. A similar approach has been applied to the design of a tunable nonlinear electric response⁸⁰ (Fig. 4d).

Intensity-dependent (nonlinear) polarization rotation can also be achieved with a varactor-based chiral metadvice⁸⁸. This polarization effect is almost negligible in any natural crystals, whereas in the nonlinear metamaterial the response of the structure has strongly resonant features caused by the excitation of currents in the left-handed metamolecule by the left-handed circularly polarized wave. At the same time, the right-handed circularly polarized wave does not noticeably excite any resonances in the structure. Changing the power of the incident wave shifts the resonance of

the gyrotropic response to a higher frequency, and also leads to asymmetric transmission in the forward and backward directions⁸⁸.

The quadratic nonlinear response of the varactor-loaded metamaterials⁸⁹ can be used in various parametric processes, including phase conjugation⁹⁰, three- and four-wave mixing⁹¹, and second-harmonic generation in quasi-phase-matched⁹² and doubly resonant⁹³ structures. An unusual effect of phase matching between the backward and forward waves can also be realized in metamaterials that display negative refraction, giving rise to a range of exotic transmission and reflection features⁹². In applications that involve second-harmonic generation in metamaterials, momentum conservation can be satisfied in a process that creates backward harmonic radiation^{94,95}. For other applications of lumped nonlinear devices in metamaterials see references 96–98.

Integration of lumped electronic components with metamaterial offers a highly efficient playground for modelling nonlinear systems and also provides a straightforward way of developing highly nonlinear and switchable media for the microwave part of the spectrum.

Metadevices driven by electromagnetic forces

In metamaterials composed of an anisotropic lattice of resonant elements, such as split-ring resonators or capacitively loaded ‘meta-atoms’, the currents induced in the resonators not only affect each other through mutual inductance, but also result in Ampère’s force between the resonators (Fig. 4e), which is attractive, provided that the neighbouring currents are in phase. If the resonators are allowed to move, this force will displace them from their original positions, thus changing their mutual impedance, which in turn affects the current amplitudes and interaction forces. The balance is maintained by a restoring Hooke force, which originates from the elastic properties of the host medium. It enables the electromagnetically induced forces to change the metamaterial structure, dynamically tuning its effective properties. Reconfigurable metamaterials exploiting this force have been branded magnetoelastic metamaterials⁹⁹. Related ideas involving conformational nonlinearity of chiral spiral metamolecules¹⁰⁰ and metamaterials with a gold nanowire pairs have also been discussed recently¹⁰¹.

A strong light-driven force may be generated when a plasmonic metamaterial is illuminated in close proximity to a dielectric or metal surface¹⁰² (Fig. 4f). This near-field force can exceed radiation pressure and Casimir forces to provide an optically controlled adhesion mechanism mimicking the gecko toe: at illumination intensities of just a few tens of nanowatts per square micrometre, it is sufficient to overcome the Earth’s gravitational pull, thus offering a new opportunity for designing metadevices driven by electromagnetic forces.

The proliferation of nanostructured materials is magnifying the role of electromagnetic forces. In the near future, these could become a practical source of optical nonlinearity and photonic switching.

Outlook

Future technologies will demand a huge increase in photonic integration and energy efficiency far surpassing that of bulk optical components and silicon photonics. Such a level of integration can be achieved by embedding the data-processing and waveguiding functionalities at the material’s level, creating the new paradigm of metadevices. We argue that robust and reliable metadevices will allow photonics to compete with electronics not only in telecommunication systems, but also at the level of ‘Photonics Inside’ consumer products such as mobile phones or automobiles. The main challenges in achieving this vision will be in developing cost-efficient fabrication and device integration technologies.

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Author Contributions

N.I.Z initiated the sections on reconfigurable, electro-optical, phase change, superconducting and ultrafast metadvice; Y.S.K initiated the sections on liquid crystal metadvice, nonlinear metadvice with varactors and metadvice driven by electromagnetic forces; both authors contributed equally to editing.

Competing financial interests

The authors declare no competing financial interests.