Nanophotonics: shrinking light-based technology

A. Femius Koenderink,[1] Andrea Alù,[1,2] and Albert Polman[1]

[1]Center for Nanophotonics, FOM Institute AMOLF
Science Park 104, 1098 XG Amsterdam, the Netherlands
[2]Department of Electrical and Computer Engineering, University of Texas at Austin
1616, Guadalupe Street, Austin TX 78712, USA

Abstract

The study of light at the nanoscale has become a vibrant field of research, as researchers now master the flow of light at length scales far below the optical wavelength, largely surpassing the classical limits imposed by diffraction. Using metallic and dielectric nanostructures precisely sculpted into 2D and 3D nanoarchitectures, light can be scattered, refracted, confined, filtered, and processed in fascinating new ways, impossible to achieve with natural materials and in conventional geometries. This control over light at the nanoscale has not only unveiled a plethora of new phenomena, but has also led to a variety of relevant applications, including new venues for integrated circuitry, optical computing, solar, and medical technologies, setting high expectations for many novel discoveries in the years to come.

Introduction

Optics and the science of light is a lively field of research that continues to surprise decade after decade with fundamental breakthroughs and disruptive applications. Communications technology has been revolutionized by the invention of the laser and the optical fiber, incandescent light bulbs are being replaced by efficient solid-state lighting, and solar energy technologies are on their way to price parity with fossil-fuel based power generation. A large number of these developments has resulted from increased control over the flow of light at length scales smaller than the wavelength. Squeezing light to nanoscale dimensions also opens the prospect of dense optical integrated circuits, which may overcome fundamental challenges related to bandwidth and energy dissipation in today's electronic integrated circuit technology. More broadly, the field of nanophotonics aims at overcoming Abbe's diffraction limit, developing technology able to manipulate light on a deep-subwavelength scale. As photons are shrunk to the nanometer scale, ultimately approaching the scale of the wave function of electrons, fundamental new science is expected, and important technological advances appear. In this article we review recent highlights in the science and applications of nanophotonics, focusing on the ultraviolet/visible/near-infrared spectral range, and provide an outlook for the bright future of this research field.

Photonic crystals

The initial concept for on-chip miniaturization of light dates back to the late 1990's, when photonic crystals – periodic structures fabricated from high refractive index materials like Si or GaAs – were proposed and realized (Fig. 1A). As the periodicity in these structures approaches the wavelength of light a photonic bandgap can appear, analogous to the energy bandgap in a semiconductor. The propagation of light with a frequency in the band gap is then forbidden, except in localized regions created by a well-designed break in periodicity, such as line defects that can guide light, or point defects that confine light. Band structure engineering gives exquisite control over light dispersion, i.e., over the relation between its frequency ω and its effective propagation constant $k=2\pi/\lambda$, and thereby also over how fast signals of different wavelengths propagate, as given by the group velocity $d\omega/dk$.

Slowing down light in waveguides, and confining it in optical nanocavities, can be used to create optical memories and enhance light switching and manipulation schemes based on ultrafast modulation of the local refractive index. Impressively, data transmission with just one femto-Joule of energy expenditure per bit was recently demonstrated using this photonic crystal platform, and as many as a 100 individually addressable and switchable optical cavities have been multiplexed to reach a low-power optical memory with nanosecond storage time (*1*).

Plasmonics

Confining light waves is much more difficult than confining electron waves because there is no optical equivalent to the Coulomb force, based on which deep electron traps can be created. Instead, the only parameter to shape light is the material dielectric constant ($\varepsilon=n^2$, with n the refractive index), which is limited to the range n=1.3-4.0 for nearly all dielectrics in the visible spectral range. Photonic crystals provide the ultimate confinement achievable with transparent materials: light can be routed in waveguide circuits exactly at the diffraction limit. To go beyond, new strategies are necessary, one of which is provided by noble metals with large negative dielectric constants. This is a consequence of the fact that metals contain a large density of unbound electrons, which experience no restoring force upon being driven by an oscillating electric field. Light can propagate at a metal-dielectric interface in the form of surface plasmon polaritons, hybrid waves of photons and charge oscillations sustained by the electrons near the interface. When two of such interfaces are brought together in a metal-insulator-metal waveguide, light is strongly confined in the dielectric gap between them, allowing light confinement well beyond Abbe's limit (Fig. 1B). Moreover, as the gap shrinks, not just the lateral confinement increases, but also the effective in-plane wavelength is reduced as the dispersion of light is increasingly determined by the metal properties. For visible light with a free-space wavelength of 450-650 nm, an effective wavelength as small as 50 nm has been observed (*2*), and routing of light in such waveguides is subject only to the diffraction limit for this effective wavelength. The strong confinement in the dielectric gap also enhances the interaction with nonlinear or active materials placed in the gap. This has been exploited to realize plasmon lasers with modes that are tightly confined in two or three dimensions (*3*).

In a recent series of breakthroughs for nanophotonics, it was demonstrated that light can be hybridized with electrons in layers that are just one atomic layer thick, such as in graphene. Plasmons have been excited on graphene at mid-infrared frequencies (*4,5*), with wavelengths that are shrunk relative to free-space by a factor of ~50. Graphene can be electrostatically gated to locally control its carrier concentration, and thereby its dielectric constant, promising a versatile platform for dynamically controlling plasmon propagation and reaching the ultimate confinement limit for optical integrated circuits.

Antennas for light

The unprecedented level of light concentration offered by plasmonic nanostructures suggests interesting new perspectives to interface light and matter, possibly even down to the level of illuminating a single molecule with a single photon. This level of control becomes possible as light is shrunk in all three dimensions. Optical antennas are nanophotonic elements designed to achieve this functionality, transducing free-space, far-field radiation to localized electromagnetic energy. The simplest nano-antenna is a single metal nanoparticle whose free electrons can support localized plasmon resonances at visible wavelengths, implying that its far-field excitation can result in a strongly localized near-field response. Reciprocally, an optically excited nanoparticle can efficiently radiate light in a controlled way.

A small isolated nanoparticle scatters as an electric point dipole, with a well-defined but broad angular distribution of radiation. Yet, tools borrowed from RF antenna design have allowed far-reaching control over the directivity of light radiation and scattering from proper arrangements of nanoantennas, and conversely over how far-field radiation drives near-field focusing in properly designed nanoclusters. Following these principles, antennas coupled to a single quantum emitter can result in highly-directional beaming of spontaneous emission (*6*) and phased array antennas coupled to efficient single-photon sources can serve as light steering elements. Over 1000-fold brightness enhancement per fluorophore has been recorded in single-molecule experiments aimed at improving fluorescence microscopy in biological systems (*7*). Besides engineering emission directivity, antennas open a completely new regime for light-matter interaction strengths, especially in the case of dimer antennas with narrow gaps, through which plasmon modes can efficiently interact (Fig. 1C). A quantitative measure of the light-matter interaction enhancement around a nanoantenna is the Purcell factor: the rate of spontaneous emission increase due to enhanced photon mode density in the antennas optical near field. Recently, a Purcell factor as high as 1000 was reported in metallic nanoantennas with gaps in the order of just a few nm (*8*). The capability of plasmon antennas to transduce between light and molecules has large relevance for microscopy, as in the case of observation of single molecules, and infrared vibrational spectroscopy using field enhancement at sharp metal tips (*9*).

Shrinking light and quantum photonics

As light is shrunk to smaller and smaller scales, its interaction with matter occurs over volumes that eventually become comparable to a single atomic unit cell. In these situations, classical models for describing optical interactions fail. In the dimer nanoantenna of Fig. 1C, for instance, classical electromagnetic theory would predict a diverging light intensity in the gap as the distance between particles approaches zero. For gaps below a few nanometers, however, quantum tunneling effects start dominating and the non-locality of the electron wave function, which extends across the gap, fundamentally limits the overall light intensity enhancement (*10*). As nanofabrication tools allow finer and finer control, such nanoscale quantum effects become increasingly relevant in experiments.

At these scales, also our understanding of quantum optical effects faces new challenges to properly describe the interaction of single emitters with single plasmons. For example, the Purcell factor has needed rewriting in plasmonics as the conventional definition of a resonant mode, and its associated cavity volume, must be revised as light shrinks to deep-subwavelength volumes where optical absorption losses and quantum effects become dominant (*11*). Furthermore, in the case of very high Purcell factors, the characteristic spontaneous emission time can become comparable to a single cycle of the electromagnetic field, in which case Fermi’s Golden Rule breaks down (*12*).

In this context, it is currently being debated whether the quantum mechanical regime of strong coupling can be reached with plasmons and single emitters. When a single emitter releases its energy as a quantum of light near a nanoresonator, the decay is not irreversible. Instead, the coupling may be made sufficiently strong to enable Rabi oscillations, coherently exchanging the excitation (*13*). This regime may enable ultrafast integrated quantum circuits based on single photons, with plasmons and emitters taking the role of information carriers and processors (*14*). The current state of the art in integrated quantum photonics has been reached in photonic crystals, where single photon sources with near-unity efficiency have been reported (*15*), setting the stage for single-photon transistors and quantum-logic gates based on single-photon nonlinearities (*16*). These remarkable advances are opening a new paradigm for scalable, integrated quantum photonics.

Shrinking vector fields to boost magnetism and chirality

Antennas not only localize the photon energy density but also provide a way to control the vector nature of light at the nanoscale. The relevance of magnetic effects in optics is conventionally neglected for small objects, as material magnetization rapidly vanishes as the frequency grows. This also implies that chiral effects, like circular dichroism and optical rotary power are inherently weak in ordinary matter, as they require that distinct optical responses, such as an electric dipole plus a magnetic dipole response, or an electric dipole plus a quadrupole response, occur simultaneously in the same object with a controlled phase lag. Such magnetic and magneto-electric effects can be boosted to become as strong as conventional electric responses by setting up scattering resonances in loop-shaped and helical antennas that carry circulating optical currents, or by pairing achiral resonances in strongly asymmetric geometries (*17*, *18*). These nanostructures may allow enhancement in the local chirality of the optical field such that enantiomers, i.e., molecular stereoisomers that are mirror images of each other, may be optically screened at the single molecule level by placing them near a chiral antenna (*19*). Furthermore, helicity-dependent near-field enhancements may be used to control spin-polarized optical transitions.

Metamaterials and metasurfaces

The unprecedented opportunity to localize light at deep-subwavelength scales has not only been applied to isolated optical nanoresonators, but has also led to the synthesis of optical metamaterials - artificial materials with an unusual optical response, formed by ordered or disordered collections of resonant nanoscale plasmonic scattering elements. Many unusual bulk optical responses have been theoretically predicted and experimentally verified based on complex 3D metallodielectric architectures, using nanoantennas as their basic elements. Remarkable nanofabrication advances have allowed the realization of nanostructured materials with unusual optical responses such as for example a metamaterial slab composed of 3D chiral helix antennas (Fig. 1D) (*20*), offering large selectivity to circular polarization with a broadband response in an optical frequency band where optical components filtering circular polarization do not exist. The fact that constituent antennas can carry both a strong electric and a strong magnetic response gives the opportunity to reach optical properties far outside the scope of naturally available refractive indices, permittivities and permeabilities. Such materials can for example refract light in unexpected directions, as demonstrated in various geometries, most recently in the ultraviolet spectral range (*21*). Negative refraction has been a precursor to the broad paradigm of transformation optics (*22*), most popular for enabling invisibility cloaks that can make objects undetectable in a certain frequency band by wrapping them in a metamaterial shell with suitably graded electric and magnetic response. Strong frequency dispersion, as well as absorption that takes place in the constituent antennas, are fundamental constraints of passive metamaterials that have so far limited their applicability in practical applications.

The need to overcome loss, combined with the continuous drive towards integration and large-area fabrication, has inspired a recent shift from 3D metamaterials towards 2D optical metasurfaces (Fig. 1E). Metasurfaces are planarized, ultrathin patterned artificial surfaces that are designed to mimic the functionalities of conventional optics and metamaterials in two dimensions, avoiding absorption losses by light propagation in the third dimension. Strongly localized optical resonances induced by plasmonic nanoantennas enable abrupt phase and amplitude discontinuities, which can be tailored at will across the surface to provide a controllable transverse gradient, inducing anomalous refraction, reflection, and control over subwavelength structure of the impinging wavefront (*23*,*24*). This enables the realization of ultrathin lenses, beam steering devices, and generation of orbital angular momentum of light, to name just a few examples. A promising trend in metasurface design is the use of arrays of dielectric nanoscatterers that show geometric Mie-type resonances, avoiding absorption losses (*25*,*26*).

Recent work has shown that flat and ultrathin metamaterials may be used to engineer to a large extent both the spatial and spectral response of the impinging wavefront. In this sense, metasurfaces and metamaterials can operate as all-optical circuitry, processing the impinging signals at the nanoscale with nanocircuit elements that may form thin metasurfaces acting as complex operators (*27*). These concepts may lead to all-analog filtering, signal processing, and even computing functionalities performed as light interacts with these devices (*28*).

Probing nanoscale optical fields

Measuring light confinement is inherently difficult, as the diffraction limit of conventional microscopy places a lower limit to imaging resolution. Yet, advances in nanophotonics research require new techniques to excite materials and probe them at the nanoscale. While super-resolution microscopy tools like PALM, STORM and STED have made it possible to circumvent Abbe's limit, they work best for specimens that are almost transparent and that can be functionalized with proper fluorescent markers. Nanophotonics research, instead, often requires a point detector, or a point source of vector fields that can be brought into the optical near field. Near-field scanning microscopy uses a sharp raster scanning probe to approach a nanostructure of interest to within 10 nm from its surface. The probe either acts as a scatterer that converts near fields into far field light (*4,5*), or as a fiber-coupled source or detector. This technique has become so advanced in recent years that electric and magnetic field components of light can be separately imaged at a length scale far below the wavelength, and at femtosecond time scales (Fig. 2A) (*29*). Scanning emitter lifetime imaging microscopy can probe the optical density of states in 2D using a single fluorescent source attached to the end of a sharp fiber probe (*30*). Electron beam induced excitation in a scanning/transmission electron microscope is another highly controllable way to study optical modes and resonances in polarizable metallic and dielectric nanostructures with deep-subwavelength spatial resolution (Fig. 2B). Cathodoluminescence spectroscopy enables spatial mapping of the radiative density of states in 2D using the light emitted when a beam of fast electrons impinges on a sample (*31*). Conversely, electron energy loss spectroscopy (EELS) maps the kinetic energy lost by electrons during their interaction with photonic structures. EELS and CL have recently been demonstrated in tomography mode, imaging the localized surface plasmon modal field distribution of Au nanoparticles in 3D (*32,33*). Electron beam induced optical images provide a spatial resolution of around 5 nm, on par with the best optical super-resolution microscopy.

Practical applications of nanophotonics

Nanophotonics has already delivered many of the original promises dating back to when this research field started to develop around ten years ago. One of the earliest nanophotonics discoveries to transcend the lab were chemically synthesized silica-core/Au-shell nanoparticles for applications in medical diagnostics and therapy (*34*). When introduced into the blood stream, these 'nanoshells' are preferentially trapped in malicious tissues. When irradiated with a laser tuned to their plasmon resonance, the particles are locally heated, destroying the cells. This concept is presently being tested in clinical trials on humans for cancer treatment.

Molecular sensing and spectroscopy has motivated the drive to controllably shrink light to the size of a single molecule. It was found that optical hotspots in rough metal surfaces arising due to localized surface plasmons result in a strong enhancement of the molecular Raman scattering signal, forming the basis for surface enhanced Raman scattering (SERS), a well-established spectroscopy tool in chemistry. In a similar area, new sensors have been proposed based on the plasmon resonance wavelength shift caused by different molecular species placed in the plasmonic near field. Antibody-specific chemistry may also be used to bind or cluster plasmon particles, using the fact that gold is easily functionalized through thiol chemistry; commercial pregnancy tests today use this concept. An intriguing application of plasmonic light focusing is in DNA sequencing, in which a DNA molecule is transported through a small hole in a metal film, with the aim to read out the base-pair sequence by the subsequent detection of fluorescent markers that are selectively bound to different base pairs (*35*).

Lasers, solid-state lighting and photovoltaics are also important fields in which nanophotonic structures have enabled new designs for improved functionality. Photonic crystal lasers have now become so advanced that they can deliver power in the Watt range (Fig. 3A) (*36*). The emission of light-emitting diodes (LEDs) is strongly enhanced if a suitably designed periodic array of Ag nanoparticles is embedded in the light-emitting phosphor (Fig. 3B). These nanostructures help to efficiently couple and scatter light from the UV pump LED into the phosphor material and, at the same time, aid the directional outcoupling of the phosphor emission which enhances LED efficiency and brightness (*37*). Conversely, periodic and aperiodic metasurfaces composed of resonant plasmonic or dielectric nanoparticles can lead to improved light coupling and trapping into solar cells, supporting increased photovoltaic energy conversion efficiency, as well as thinner cell designs that can be made at lower cost (Fig. 4C) (*25*). Recently, efficient solar cells were realized using InP nanowires (*38*). Here, the small radius of the wire enables efficient collection of electrical carriers. At the same time, optical resonances in the nanowire can lead to light concentration, increasing the photovoltage (*39*).

Metal nanowire networks have been developed as transparent electrically conducting coatings. Even though these nanowires suffer from Ohmic losses due to plasmon excitation, they show a beneficial tradeoff between optical transmission and electrical conduction, and can be made at relatively low cost (Fig. 4D) (*40*). These nanowire networks are already finding applications in solar cells and computer display and tablet technology, replacing the commonly used, expensive and brittle indium-tin-oxide as a transparent top-contact.

The unique near field focusing provided by plasmonic nanostructures has also found applications in heat-assisted magnetic recording (HAMR) for data storage, a technique in which the magnetic phase change in a recording medium is facilitated by a transient temperature increase (Fig. 3E). This increase is induced by an optical hotspot resulting from nanofocusing of plasmons onto a magnetic film (*41*). Plasmonic hole arrays have been demonstrated as color multiplexers in CCD imaging systems (Fig. 3F) (*42*). Here, light within specific wavelength bands is guided to the matching light collection pixels on a CCD array and then converted to electrical signals. This represents an important advantage over the use of conventional color filters, in which a significant portion of the impinging light is lost by absorption.

Future developments and perspectives

Nanophotonics provides a diverse set of tools to build on: photonic crystals offer ultimate dispersion control and low-loss storage, while plasmonics is the platform of choice to control light on ultrafast timescales and ultrasmall length scales matching optical interactions with single molecules. Metamaterials and metasurfaces provide the ultimate control over all properties of light. With the great control over light at the nanoscale that has now been achieved, the nanophotonics community is in an ideal position to bring the field another step further, by coupling light with other degrees of freedom at the nanoscale. Hybrid nanophotonics revolves around the simultaneous control of tightly confined light, and phonons, electrons, spins, and/or excitons interacting with light. For example, when optical fields and acoustic vibrations are co-localized at the nanoscale, light can be used to control mechanical motion, and vice versa, with optomechanical coupling strengths that are not achievable with other geometries (Fig. 4A). These interactions have for example been used for laser cooling of a nanomechanical resonator to its quantum ground state (*43*). It has even been suggested that plasmon-enhanced SERS can be described by the dynamic backaction of the plasmon on a molecule’s vibration, paving the way to a new form of molecular quantum optomechanics (*44*). Future hybrid nanophotonic systems hold the promise to couple electron spins with light, enabling integrated networks for quantum nanophotonics, for example using single-photon emission by color centers in diamond or defect centers in novel materials such as SiC (*45*).

Nanophotonics is also at the brink of making key contributions to the development of novel energy conversion mechanisms. The recently discovered plasmo-electric effect in metal nanoparticles and hole arrays (*46*), wherein optical illumination directly creates an electric potential by off-resonant excitation of a plasmonic structure (Fig. 4B), is being further explored to investigate how electrical power can be best generated. Another intriguing challenge is to harvest the excess energy of hot electrons excited in optically excited plasmonic nanostructures (*47*). This may find applications in energy harvesting and also plasmon-assisted surface catalysis for the generation of fuel (e.g. ethanol, hydrogen) from sunlight. More generally, plasmon-assisted photochemistry and catalysis is a research field with many opportunities to be explored.

An initial motivating factor for nanophotonics research was its direct impact on optoelectronic integration, aiming to bring together electronic and photonic length scales. The development of novel nanostructures that enable a nonreciprocal flow of light paves the way to on-chip all-optical isolation, in which light can only propagate in one direction, one of the missing components towards a fully integrated light-based communication system. Recent proposals and experimental demonstrations have shown that non-reciprocity can be indeed achieved using a suitably designed spatio-temporal modulation of the local permittivity in waveguides and microring resonators (Fig. 4C) (*48,49*). Another interesting direction in this context lies in the exploration of the photonic equivalent of topological insulators. In such structures, unidirectional flow of light and reflection-less propagation robust to disorder may be achieved by designing periodic metasurfaces that support topologically nontrivial band diagrams (*50*).

Two further important ingredients for opto-electronic integration are access to strong nonlinearities within a very compact footprint, and dynamic tunability and adaptability (*51*). The nonlinear optical response of materials is typically weak, and therefore large volumes are required to realize a measurable response. The recent observation of a giant nonlinear response from plasmonic metasurfaces coupled to intersubband transitions in semiconductors may open exciting venues for nonlinear nanophotonics (*52*). Similarly, one may ask how metasurfaces can enable new schemes for dynamically tunable integrated photonics. For example, can electrically-tunable metasurfaces be used to steer and multiplex light on a chip, thereby integrating an optical fiber communication system within an electronic integrated circuit? Also, how can "metatronic" optical signal processing and computing circuits be built and reconfigured using building blocks with discrete optical functions (Fig. 4D)? Novel 2D materials and heterostructures based on graphene, MoS_2, and WSe_2 will play an important role in bringing light to the nano-scale, enabling 2D electrically-tunable integrated optical nanocircuits (Fig. 4E) (*53,54*). It will be interesting to see to what degree of integration complexity optical and electronic functionality can ultimately be achieved on a chip using these materials. A decisive factor will be to find the right tradeoff between light confinement and optical absorption losses.

The unique control over light at the nanoscale that scientists have achieved in recent years leads to a continuous stream of new fundamental insights in the interaction of light with matter at a deep subwavelength scale. There is no doubt that this intense research activity promises a bright future for nanophotonics in the years to come.

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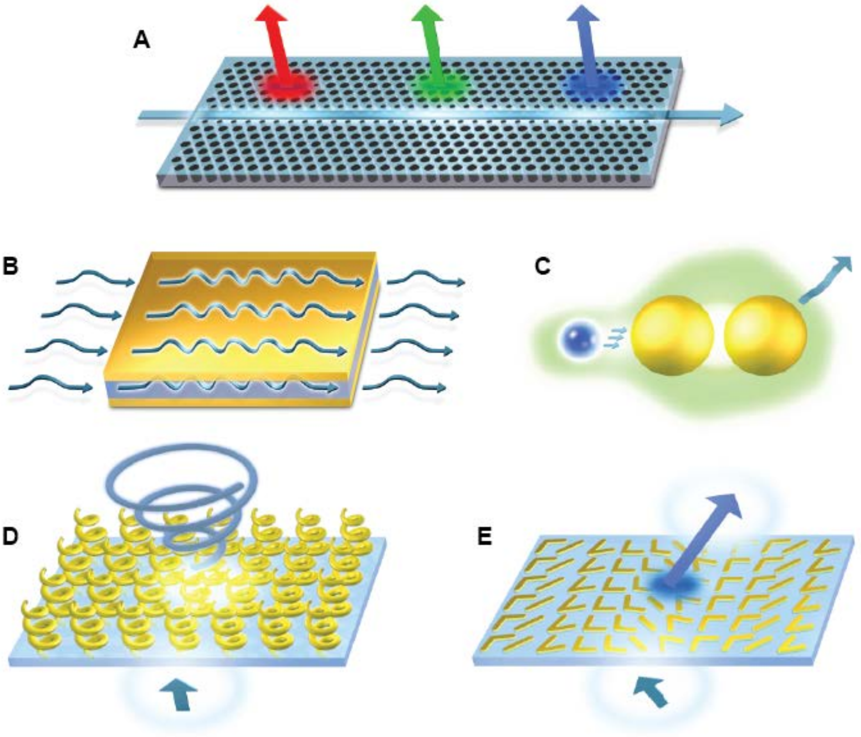

Fig. 1. Nanophotonic architectures. (**A**) 2D photonic crystal waveguide coupled to resonant cavities serves as a wavelength division multiplexer. (**B**) Metal-insulator-metal surface plasmon polariton waveguide strongly confines light and shrinks the wavelength. (**C**) Plasmonic dimer nanoantenna coupled to an optical emitter creates directional emission of light. (**D**) Metasurface composed of chiral antennas offers selectivity to circularly polarized light. (**E**) Optical metasurface composed of graded plasmonic or dielectric antenna geometries enables wavelength-dependent control over the reflection and refraction of the optical wave front.

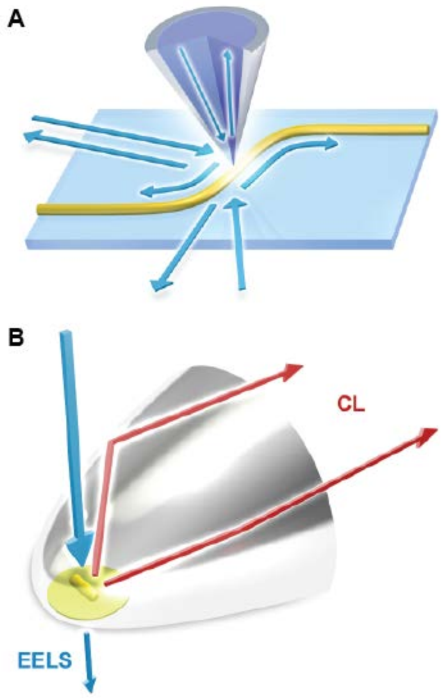

Fig. 2. Nanophotonic characterization techniques. (**A**) Near-field scanning optical microscopy, either in transmission, collection or scattering mode, can measure the magnetic and electric field components of light at a 10-100 nm spatial resolution in the femtosecond time domain. (**B**) Resonant modes and light dispersion of plasmonic and dielectric nanostructures can be probed by electron irradiation: either by collecting the induced radiation (cathodoluminescence) or by measuring the electron energy loss spectrum (EELS). The spatial resolution of these techniques is determined by the electron beam width: ~10 nm.

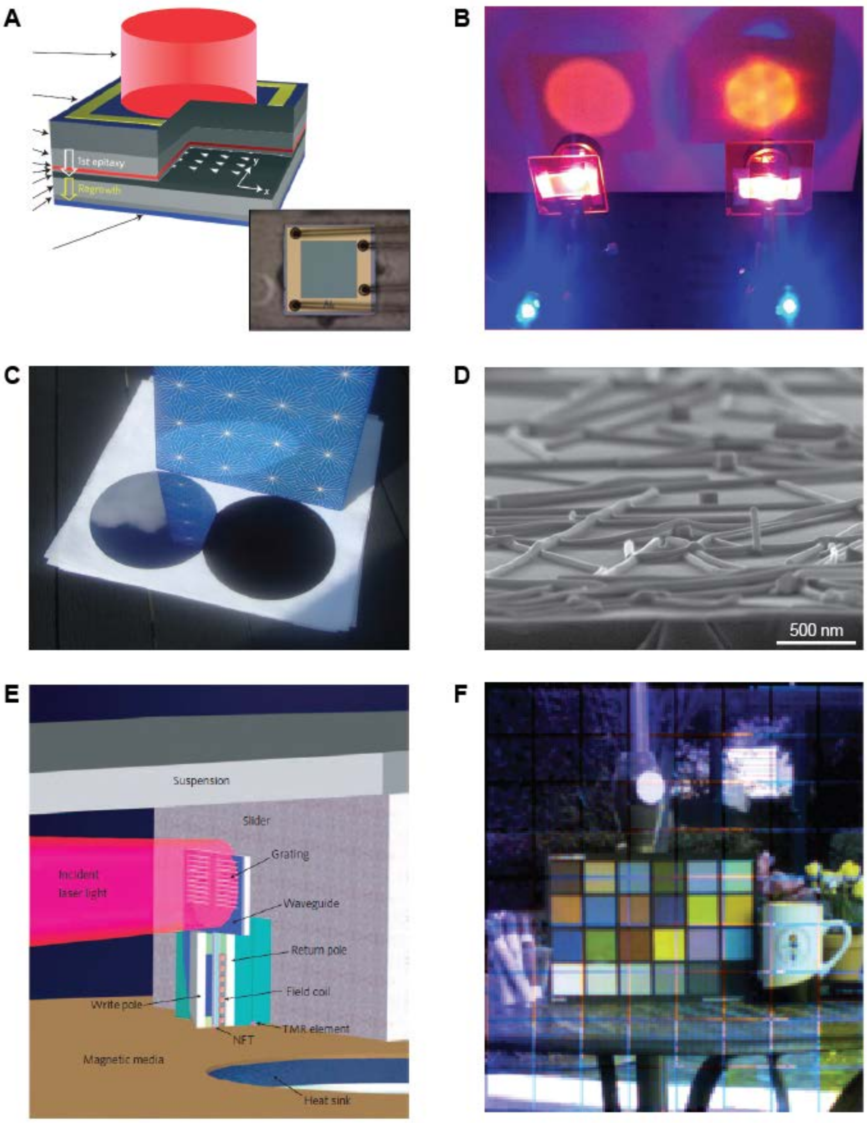

Fig. 3. Nanophotonic technologies and applications. (**A**) High-power photonic crystal laser (reprinted with permission from Ref. 36, copyright 2014, Nature Publishing Group). (**B**) Light emitting diode without (left) and with (right) enhanced emission due to light scattering from embedded Ag nanoparticles (image A. Nikitin, after Ref. 37, reprinted with permission, copyright 2013, Nature Publishing Group). (**C**) Black silicon wafer with (right) and without (left) dielectric optical metasurface for enhanced light coupling and trapping in solar cells (reprinted with permission from Ref. 25, copyright 2012, Nature Publishing Group). (**D**) Transparent conducting nanowire network made from randomly dispersed chemically grown single-crystal Ag nanowires (reprinted with permission from Ref. 40, copyright 2012, Nature Publishing Group). (**E**) Schematic of heat-assisted magnetic recording head with plasmonic light focusing taper (reprinted with permission from Ref. 41, copyright, 2009, Nature Publishing Group). (**F**) Image taken with plasmonic CCD chip with integrated plasmonic nanohole array for wavelength multiplexing (reprinted with permission from Ref. 42; copyright, 2013, American Chemical Society).

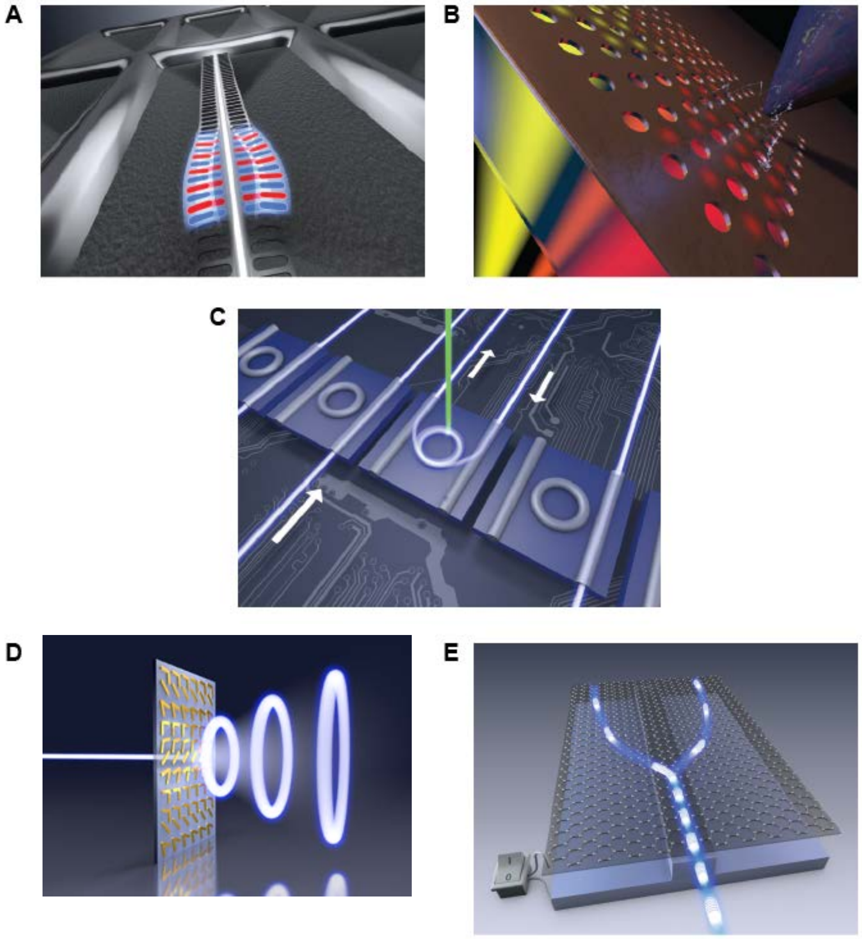

Fig. 4. Nanophotonics future challenges and research fields (**A**) Hybrid nanophotonics: coupling light with other degrees of freedom such as e.g. mechanical motion. (**B**) Plasmo-electric effect converting sunlight to electrical power using all-metal nanostructures. (**C**) Non-reciprocal optical integrated circuit enables logic functions for optical computing. (**D**) Electrically tunable optical metasurface serves as wavelength division multiplexer in optical communication and computing networks. (**E**) Electrically-tunable 2D graphene opto-electronic integrated circuit brings together optical and electronic length scales. Cartoons inspired by Refs. 23,24,27,43,46,49, and 54.