feature article

From near-field optics to optical antennas

Lukas Novotny

Nanoscale probes that convert light into localized energy or vice versa form the basis for diffraction-unlimited imaging and intriguing light–matter interactions.

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In a prophetic letter to Albert Einstein on 22 April 1928, Irish scientist Edward Hutchinson Synge proposed a microscopic imaging method in which an optical field scattered from a tiny gold particle could be used as a radically new light source. The gold particle, Synge argued, would act as a local probe of some sample of interest, with the scattered light transmitted through the sample and into a detector. The sample's image could then, at least in principle, be obtained by raster scanning the particle over the sample surface while continuously recording the detected light's intensity.

Along with a sketch of the proposal, shown in figure 1, Synge wrote, "By means of the method the present theoretical limitation of the resolving power in microscopy seems to be completely removed and everything comes to depend on technical perfection." The resolution was limited not by the wavelength of light, he realized, but by the particle's size. A month later Einstein replied that although Synge's implementation appeared *prinzipiell unbrauchbar* ("essentially unusable"), the basic ideas seemed correct and he should publish the article, which Synge then did.[1]

Synge's proposal was truly visionary. It conceived the concept of scanning probe microscopy and formed the basis for near-field optics, the study of nonpropagating optical fields and their interaction with matter. The proposal was also well ahead of its time and had to wait more than a half century for experimental verification. In 1982, a year after the invention of the scanning tunneling microscope at IBM's research lab in Switzerland, Dieter Pohl and colleagues there recorded the first optical scans at a resolution below the diffraction limit. Before then, Synge's proposal had been reinvented several times and proof-of-concept experiments using acoustic waves, radio waves, and microwaves had been performed.

In the early 1980s, much of the research concentrated on high-resolution microscopy and spectroscopic characterization of materials. That work provided the first spatial maps of surface plasmons—collective electron density oscillations—and a route for studying single-molecule emission and pulse propagation in optical waveguides. Near-field optics subsequently branched into various other fields, such as plasmonics, and today elements of it have been absorbed into the wider field of nano-optics and nanophotonics. (See the PHYSICS TODAY articles by Mark Stockman, February 2011, page 39, and by Thomas Ebbesen, Cyriaque Genet, and Sergey Bozhevolnyi, May 2008, page 44.)

Intriguingly, Synge's particle can be viewed not simply as a light scatterer but as an optical antenna, a mesoscopic structure that efficiently converts the energy of incident optical radiation into localized energy or vice versa, depending on whether the antenna is configured as a receiver or a transmitter. The connections between near-field optics and antenna theory are the focus of many ongoing studies, as we'll see later.

The near field

In conventional optical imaging and spectroscopy, an object is typically irradiated by a light source and the scattered or

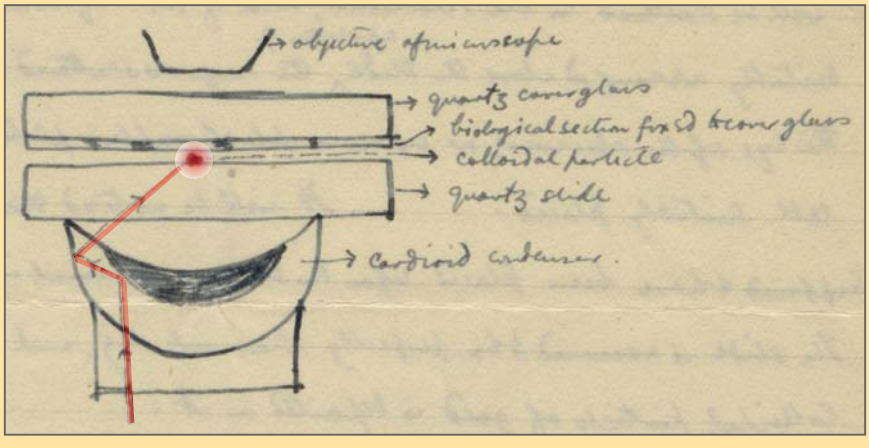

Figure 1. In an April 1928 sketch sent to Albert Einstein, Edward Hutchinson Synge proposed a new microscopy method: using a tiny gold particle (red) between two quartz slides to scatter incident light from below onto a sample. Light that didn't strike the particle would be totally internally reflected, and an objective lens of a microscope could be positioned to accept some of the gold-scattered light. That arrangement, Synge wrote, could be used to image a biological specimen fixed to the top cover slip at a resolution below the diffraction limit. (Courtesy of the Albert Einstein Archives, Hebrew University of Jerusalem, Israel.)

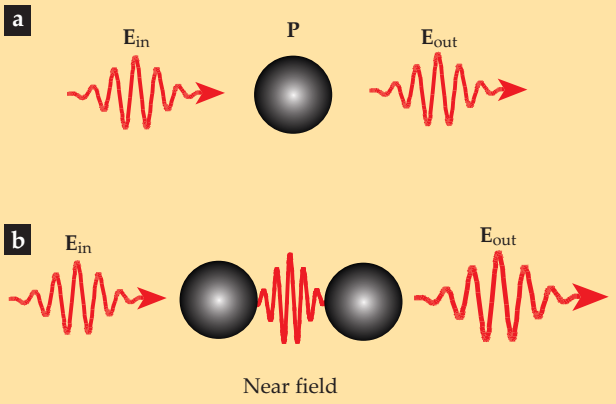

Figure 2. (a) In the canonical optics problem, incident radiation $\mathbf{E}_{in}$ polarizes an object, and the induced polarization **P** gives rise to emitted radiation $\mathbf{E}_{out}$ through, for example, scattering or photoluminescence. **(b)** In the canonical near-field optics problem, the object is split into a probe and a sample, and the emitted field is a measure for the mutual near-field interaction between them.

emitted light is recorded by a detector. That situation, illustrated in figure 2a, constitutes the canonical optics problem: The incident field $\mathbf{E}_{in}$ induces in the object polarization currents, which in turn give rise to an emitted field $\mathbf{E}_{out}$. In the canonical near-field optics problem, on the other hand, the object is split into two parts, as shown in figure 2b. One part is referred to as the probe and the other part as the sample.

Typically, the probe is engineered to exploit the unique properties of metal nanostructures at optical frequencies to localize incident radiation and enhance the light–matter interaction with the sample. For example, when a light wave is incident on a tiny gold probe, as in Synge's proposal, the incident field periodically displaces the probe's electrons with respect to the lattice. Near resonance, that charge oscillation gives rise to a greatly amplified electric field just outside the probe, which then affects the nearby sample.

The field $\mathbf{E}_{out}$ emitted by the combined system is detected and serves as a source of information from which the sample properties are reconstructed. Manipulating the relative separation between probe and sample adds to the information content. A near-field image is recorded by raster scanning the probe over the sample, or vice versa, while continuously detecting the emitted field. Typically, an image is represented as a two-dimensional scan $f[\mathbf{E}_{out}(x,y)]$, where f stands for a function of the field, such as intensity or an interferometric signal. The enhanced resolution, which can be as fine as 10 nm if visible or IR light is used, originates from the conversion of nonpropagating field components confined on the sample surface to propagating radiation in the presence of the probe.

How it works

A near field can arise from primary sources, such as electrically driven currents, or secondary sources, such as induced polarization currents. The choice of basis in which to express the field depends on the geometry of that source. For example, the fields near a planar sample surface are conveniently expanded in a plane-wave basis, also known as the angular spectrum representation, and can be described as a superposition of propagating plane waves and evanescent waves, whose field amplitudes decay exponentially from the surface. The localized evanescent waves are also the source of the fields' high spatial frequencies.

Interestingly, evanescent waves are not orthogonal, and quantization can be accomplished only if the fields that generate the evanescent waves are also accounted for. For example, for a plane interface irradiated under total internal reflection, Charles Carniglia and Leonard Mandel at the University of Rochester showed that the incident wave, reflected wave, and evanescent wave form an orthogonal set of modes that can be quantized similar to the free radiation field. Consequently, the quanta of evanescent fields are always connected to their sources and there are no purely evanescent photons in the plane-wave basis.

The localization of optical fields is more conveniently studied in the multipole basis. The multipole expansion generates a near field, an intermediate field, and a far field. The near field typically dominates the region $kR \ll 1$, where k is the wavenumber and R the distance from the source (primary or secondary). Retardation of phase and energy propagation can often be neglected in the near-field zone, and light-matter interactions can be studied in the quasi-static approximation. When $kR \approx 1$, the character of the fields changes and the fields become neither evanescent nor freely propagating. And in the far-field zone ($kR \gg 1$), the fields become detached from their sources and propagate as free radiation. To describe the birth of a photon, Ole Keller uses the term "photon embryo" to denote a state of the field that has not yet detached from its source.[2]

On an elementary level, one can think of a near-field measurement as an electromagnetic interaction between a pair of two-level atoms—sample atom S and probe atom P. A question of central importance is, What is the probability for a system initially in a state $|S = 1, P = 0, F = \text{vacuum}\rangle$ (sample atom excited,

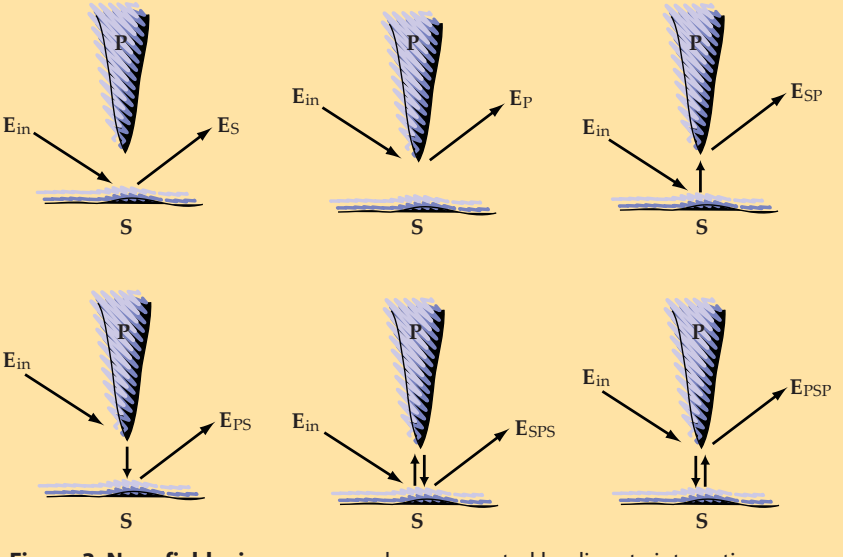

Figure 3. Near-field microscopy can be represented by discrete interactions between probe *P* and sample *S*, which locally scatter incident radiation $\mathbf{E}_{in}$. The total emitted field $\mathbf{E}_{out}$ can thus be written as a sum of interaction terms $\mathbf{E}_{out} = \mathbf{E}_S + \mathbf{E}_P + \mathbf{E}_{SP} + \mathbf{E}_{PS} + \mathbf{E}_{SPS} + \mathbf{E}_{PSP} + \ldots$, in which the sublabel for each term designates the order in which objects (the sample or probe) scatter or emit radiation.

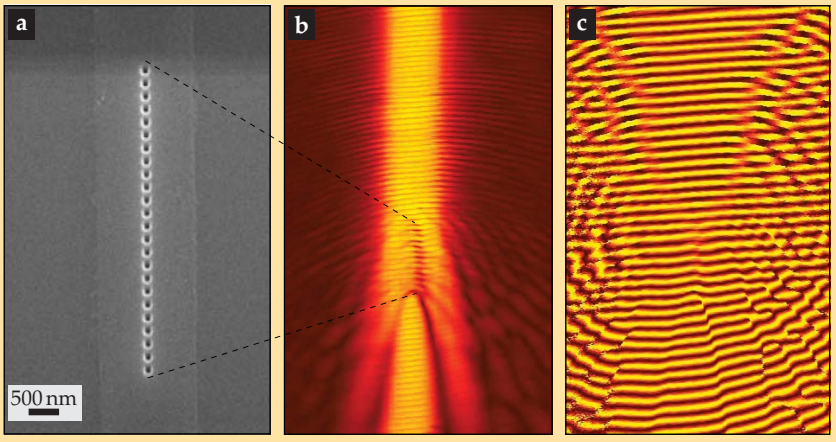

Figure 4. Light injected into a ridge waveguide containing a linear series of holes, shown here as an electron micrograph **(a)**, propagates along the guide as an electromagnetic mode that scatters during each encounter with a hole. An aperture probe can be used to locally pick up the field distribution of the scattered light just above the waveguide. Using heterodyne near-field microscopy, researchers from the University of Twente interfered that signal with a frequency-shifted reference signal to image the scattered light's amplitude **(b)** and phase **(c)** as a function of position. (Adapted from ref. 5.)

probe atom in the ground state, and no external field, except for vacuum fluctuations) to change at some later time to the state $|S = 0, P = 1, F = \text{vacuum}\rangle$? As theorists pointed out more than a decade ago, the problem leads to a noncausal result, which implies that it is not possible to know the initial and final states of both atoms.[3] Indeed, one can infer a causal relationship between them only if the final states of sample and field are not specified, a result that has implications for near-field measurements. A measurement of the system alters the near field and consequently captures information on the probe–sample interaction, not on the sample per se.

Light–matter interactions

More precisely, a near-field measurement captures the field $\mathbf{E}_{out}$ generated by the currents, primary or secondary, that define the probe–sample system. The combined system can be represented in terms of a single Green function $\mathbf{G} = \mathbf{G}_P + \mathbf{G}_S$, where $\mathbf{G}_P$ and $\mathbf{G}_S$ are the Green functions of probe and sample, respectively. The problem, however, is that $\mathbf{G}_S$ is defined in the presence of the probe and does not reflect the properties of the sample alone. To understand the mutual interaction of probe and sample, one can think of the near-field interaction in terms of a perturbation series, similar to a Born series in standard scattering problems. As illustrated in figure 3, the field $\mathbf{E}_{out}$ can then be written as a sum of discrete interactions:[4]

$$\mathbf{E}_{out} = \mathbf{E}_S + \mathbf{E}_P + \mathbf{E}_{SP} + \mathbf{E}_{PS} + \mathbf{E}_{SPS} + \mathbf{E}_{PSP} + \ldots,$$

where $\mathbf{E}_S$ and $\mathbf{E}_P$ are fields emitted or scattered from sample and probe, respectively; $\mathbf{E}_{SP}$ is the field scattered by the sample and then emitted by the probe; $\mathbf{E}_{PS}$, in turn, describes the reverse—the field scattered by the probe and emitted by the sample. Additional interactions contribute as higher-order terms in the series.

Fortunately, near-field microscopy exploits prior knowledge of probe and sample properties, which makes it possible to greatly suppress most terms in the series. For example, the signal measured by photon scanning tunneling microscopy (PSTM), in which photons—or more accurately, evanescent waves confined to a surface—"tunnel" from the sample to the probe, is dominated by the term $\mathbf{E}_{SP}$. In contrast, the signal measured by illumination-mode aperture-probe microscopy, in which the sample is excited by the field from a small hole at the probe's tip, is dominated by the $\mathbf{E}_{PS}$ term.

Individual interactions at sample and probe may involve different frequencies. Tip-enhanced Raman scattering (TERS), which relies on laser light to excite the vibrational modes of the sample, involves $\mathbf{E}_{PSP}$, but with input and output frequencies that differ by an amount corresponding to the energy of vibrational modes. Depending on the signal to be measured—whether from Rayleigh scattering, fluorescence, Raman scattering, or some nonlinear response of a system—one has to account for the coherent or incoherent sum of interaction orders.

It is convenient to divide different interaction schemes into two subgroups—a "strong probe" group, which comprises $\mathbf{E}_{PS}$, $\mathbf{E}_{PSP}$, and their related higher orders, and a "weak probe" group, which comprises $\mathbf{E}_{SP}$, $\mathbf{E}_{SPS}$, and so forth. In the strong probe regime, as the name implies, incident radiation interacts more strongly with the probe than with the sample; the opposite is true in the weak probe regime. Although qualitative, the classification scheme helps researchers interpret experimental near-field data.

Of modes and microscopy

Near-field optics makes it possible to study localized light–matter interactions and to visualize fields confined to a surface, such as waveguide modes and surface polaritons, electromagnetic waves that propagate at the interface between two media. Early work in near-field optics paved the way for several related developments, including nanoplasmonics, single-molecule spectroscopy, and short-range heat transfer. To convey the richness of those research fields, I discuss a few illustrative examples.

The $\mathbf{E}_{SP}$ term dominates if the probe is used to locally collect or scatter the fields near a sample surface. That measurement mode has been used to spatially map the propagation of evanescent waves near dielectric surfaces, to visualize the scattering of near fields at edges and sharp features, and to characterize the modes of waveguide structures. Nearly a decade ago, to simultaneously record amplitude and phase distributions, the groups of L. (Kobus) Kuipers and Niek van Hulst combined near-field microscopy with heterodyne interferometry. Figure 4 shows one of their representative data sets, obtained using a light-collecting aperture probe, from a ridge waveguide perturbed with a series of holes.[5] The phase map reveals discontinuities and singularities in addition to variations in phase velocity and dispersion. The same groups also performed time-resolved near-field measurements to visualize and study the propagation of femtosecond laser pulses in waveguide structures and photonic crystals.

Other groups have performed similar measurements. Fritz Keilmann and coworkers, for example, used heterodyne

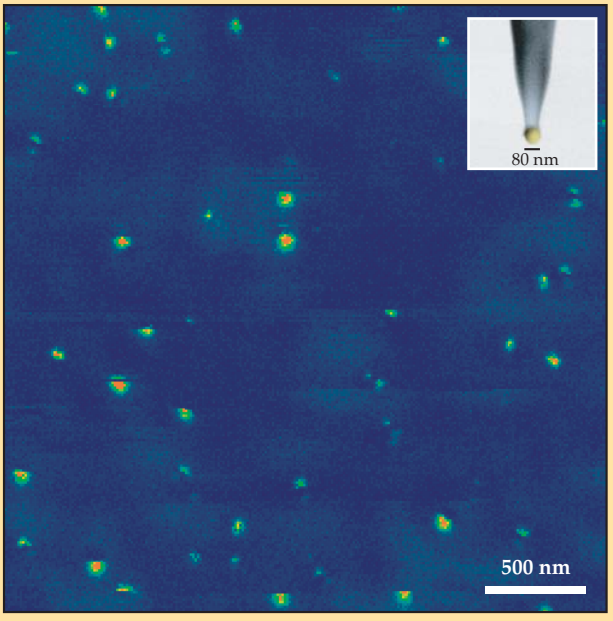

Figure 5. Single molecules on a surface. The local field near a laser-irradiated gold nanoparticle (inset) is used to excite a sample of fluorescent molecules. The single-molecule fluorescence intensities in the scanned image represent the gold particle's local field distribution. (Courtesy of Shawn Divitt, Institute of Optics, University of Rochester.)

near-field microscopy at IR frequencies to record refractive and absorptive properties at the surfaces of materials such as polymers and semiconductors.[6] To characterize optoelectronic materials or devices in the past, researchers have typically treated the device in question as a black box and simply related the output signals to input signals. Near-field maps, such as those in figure 4, reveal the inner workings of a material and thus offer richer information.

Researchers have also implemented $\mathbf{E}_{SP}$-based microscopy to record spatial maps of elementary material excitations, such as excitons and polaritons. In the mid 1990s groups led by Othmar Marti and Paul Dawson used PSTM to record the first images of surface plasmon polaritons (SPPs) on a metal surface.[7] Since then, near-field microscopy has become a standard tool for characterizing surface plasmons and visualizing the enhanced, localized fields known as hot spots in metal nanostructures.

Surface plasmons are also observed in doped semiconductors, but their resonances are typically in the IR or THz regime. To profile dopant distributions in integrated semiconductor devices and to study SPPs in polar materials, such as silicon carbide, Rainer Hillenbrand and coworkers extended near-field microscopy to lower frequencies.[8] Other groups have also made similar instrumentation advances for high-resolution imaging of materials undergoing phase transitions and for the study of Anderson localization in disordered metal films (for more on Anderson localization, see the article by Ad Lagendijk, Bart van Tiggelen, and Diederik Wiersma in PHYSICS TODAY, August 2009, page 24).

Fluorescence and Raman imaging

The interaction described by $\mathbf{E}_{PS}$ is dominant if the probe is used in illumination-mode to excite the sample. With an aperture probe in 1994, Eric Betzig and colleagues recorded the first single-molecule fluorescence images on a surface.[9] Although spectra of single molecules had been detected years earlier, it was near-field imaging that jump-started the field of single-molecule biophysics. Ironically, shortly after Betzig's breakthrough experiment, researchers realized that ordinary light microscopy can also detect the fluorescence from single molecules, and most of today's single-molecule studies no longer rely on near-field optics. Nevertheless, near-field single-molecule experiments offer greater spatial resolution and allow researchers to correlate fluorescence images with simultaneously recorded topographical maps of surface molecules. Those advantages, however, come at the cost of significantly higher experimental complexity.

Figure 5 shows a fluorescence image of single Nile blue molecules dispersed on a glass surface. In that case, a laser-irradiated gold particle at the end of an apertureless probe locally excited the molecules. The fluorescence emitted by the molecules was then collected through a distant lens. Such imaging has been applied to a variety of systems, including conjugated polymers and membrane proteins. Indeed, recent work from the groups of María García-Parajo and Christiane Höppener demonstrated that it is possible to image and identify single proteins in intact cell membranes and to follow their diffusion and localization with other proteins.

The fluorescence patterns in figure 5 represent the characteristic field distributions that support the $\mathbf{E}_{PS}$ assignment. However, reducing the scattering series in the equation on page 49 to a single term is generally a rough approximation. For example, the fluorescence of a molecule in very close proximity to an optical probe can be quenched, or turned off as the probe absorbs the molecule's energy, a process that requires corrections to the $\mathbf{E}_{PS}$ term.

Fluorescence imaging typically requires adding fluorescent molecules of choice to a sample, a process known as la-

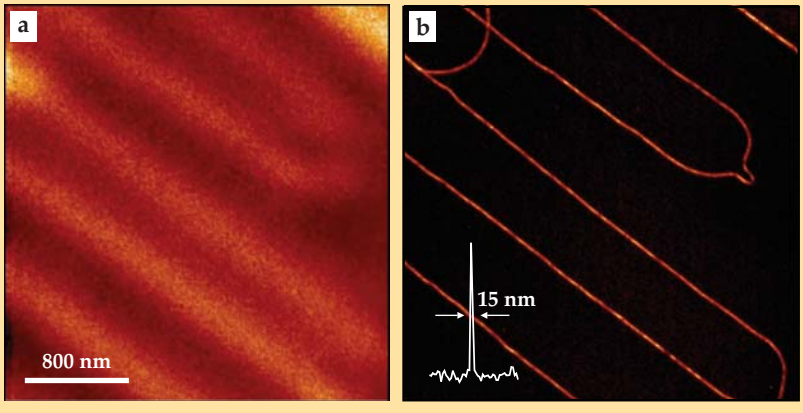

Figure 6. Single-wall carbon nanotubes, imaged **(a)** by confocal Raman microscopy and **(b)** by tip-enhanced Raman scattering (TERS). In both cases, image contrast comes from the intensity of the Raman G band at $\nu = 1580\ \text{cm}^{-1}$, a carbon–carbon stretching vibrational mode. The superior signal-to-noise ratio and resolution of TERS stems from using near-field light from a sharp gold tip scanning close to the nanotube sample to locally enhance Raman scattering. (Adapted from ref. 17.)

beling—the use of antibodies or molecules engineered to bind to a particular target molecule. That labeling could be eliminated by spectroscopically accessing the intrinsic material properties of a sample. Such chemical specificity is provided by Raman scattering and its nonlinear family of techniques, which measure the vibrational modes of materials. Raman scattering is a weak process, but about four decades ago researchers discovered that it could be enhanced by orders of magnitude by exploiting the local fields near metal nanostructures. The technique that emerged, surface enhanced Raman scattering, made it possible to detect the vibrational modes of a single molecule (see the article by Katrin Kneipp in PHYSICS TODAY, November 2007, page 40) and inspired the development of TERS.[10] In TERS, an irradiated metal tip localizes incident radiation on a target molecule and reradiates the molecule's response. Accordingly, the tip acts both as a receiving and a transmitting antenna, characteristic of $\mathbf{E}_{PSP}$ in the interaction series.

As an example of imaging specific to $\mathbf{E}_{PSP}$, figure 6 shows a TERS image of a single-wall carbon nanotube and, for comparison, the corresponding confocal Raman image. The contrast in both Raman images reflects the intensity of a normal-mode vibrational frequency band at 1580 cm^{-1}. The TERS image resolution—about 15 nm, judging by the cross section through the nanotube—and signal-to-noise ratio are much greater than the confocal Raman image of the same sample thanks to the much smaller effective sample area that interacts with the light field.

Although the near-field classification scheme illustrated in figure 3 assumes an incident light field $\mathbf{E}_{in}$, optical near fields may also be produced by other means, such as charge recombination or heating. Ten years ago Jean-Jacques Greffet and collaborators predicted that heat transfer between two separate bodies can be drastically enhanced if the two bodies interact via their surface phonon polaritons, the vibrational analog of surface plasmons.[11] The group showed that near-field heat transfer can be viewed as a phonon tunneling effect and described by a distance-dependent heat conductance. Near-field heat transfer has since been systematically investigated and has found application in thermally assisted magnetic recording, which currently achieves a data density as high as 1 Tbit/cm^2.

There are many more near-field stories to tell. Near-field optics has been used to measure magnetic fields at optical frequencies, to map out the mode structure in metamaterials, to localize defects in materials, and to trap and manipulate nanoparticles and viruses. Researchers have also shown that the near-field spectrum of a thermal emitter differs from its blackbody spectrum measured in the far field and that the surface of a blackbody can be structured so that its thermal near fields are converted to coherent radiation. Near fields have been exploited for controlling and enhancing nonlinearities in materials and for probing their dipole-forbidden transitions. Moreover, because an optical near field is confined in space, it can be viewed as an ultrasmall optical cavity. Such cavities open new opportunities for ultrasensitive detection and quantum control.

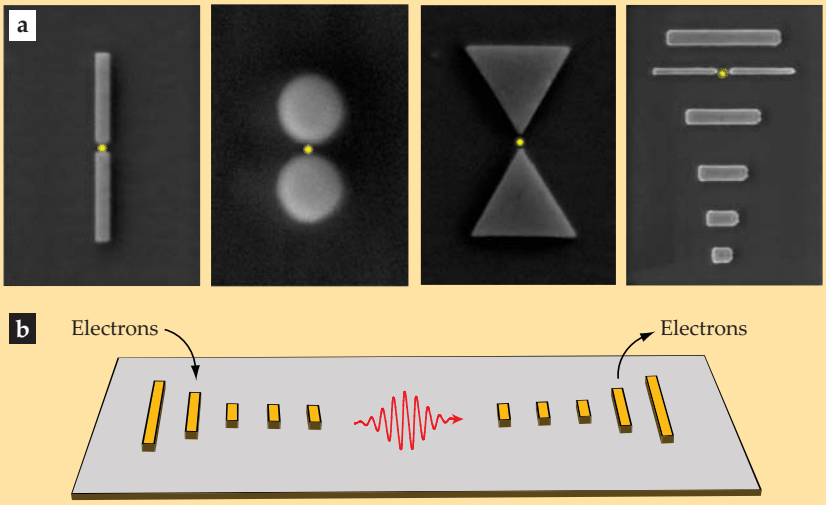

Figure 7. (a) Scanning electron micrographs of various optical antennas: a gap antenna that radiates as an effective linear dipole; a Hertzian dimer antenna,[18] which provides superior radiation efficiency; a bow-tie antenna, whose geometry provides broadband reception or transmission; and a Yagi–Uda antenna, which yields a highly directional signal due to the interplay between the different elements. In each case the antenna (gray) either couples incident light onto, or efficiently reradiates energy out of, a molecular receiver or transmitter (yellow). **(b)** An antenna can also be configured as an optical interconnect, in this case performing transduction between electrical current and optical radiation.

Optical antennas

Near-field optics has many parallels with antenna theory.[12] The common features are evident in the canonical near-field optics problem outlined in figure 2b. By simply removing the outgoing light path, one ends up with a schematic diagram of a receiving antenna: One object assumes the function of a receiver and the other represents the antenna. Alternatively, removing the incoming light path but leaving the outgoing light path intact yields a schematic representation of a transmitting antenna. Because an antenna is defined by its function, namely its ability to convert incident radiation to localized energy and vice versa,[13] Synge's nanoparticle can be viewed as an optical antenna. It takes incoming freely propagating radiation and localizes it on a sample, the receiver.

Conceptually, optical antennas fulfill the same function as their radio and microwave analogs, but with properties that differ in important ways. At radio frequencies, metals behave like perfect conductors, with a skin depth—the extent to which a field penetrates the metal surface—that is negligible compared to the size of the antenna. But at optical frequencies, the skin depth is appreciable, on the scale of tens of nanometers, and the metal behaves like a plasma strongly coupled to the incident light. The antenna geometry thus scales with an effective wavelength that differs from the wavelength of incident light.[13]

Moreover, because of their small size, receivers and transmitters cannot be wired to antenna elements in the traditional fashion. Instead, interconnects become part of the antenna design;[14] in the extreme limit, receivers and transmitters become discrete quantum objects such as molecules, quantum dots, or tunnel junctions that couple to the antenna by the transfer of energy or charge.

In practice, optical antennas are fabricated using both top-down nanofabrication techniques and bottom-up synthesis. Figure 7 shows several examples fabricated by focused ion-beam and electron-beam lithography. Over the past years

many different antenna structures have been constructed and studied, including self-similar (or fractal), bow-tie, cross, half-wave, monopole, and patch antennas. Traditionally, engineers have controlled optical radiation by manipulating the wavefronts of free propagating radiation with mirrors, lenses, and diffractive elements, but optical antennas make it possible to control light on the subwavelength scale. That capability makes them attractive components for integrated optoelectronics.[15]

As a mesoscopic structure that enhances the light–matter interaction, an optical antenna operates in a territory where quantum optics and optical engineering meet and where microscopic and macroscopic concepts overlap. For example, an antenna's impedance can be represented in terms of the electromagnetic density of states,[16] its radiation efficiency in terms of the quantum yield, and its aperture in terms of the absorption cross section.[13] Current research into optical antennas is being driven in part by the promise of controllably enhancing the performance and efficiency of photodetection, light emission, and sensing. For example, the active area of antenna-coupled photodetectors can be made very small, which improves their signal-to-noise ratio, response time, and efficiency.

The highly localized fields near an optical antenna also hold promise for interacting with matter in a way that violates the usual dipole selection rules and opens experimental access to momentum-forbidden transitions. Such interactions have the potential to enrich optical spectroscopy and boost the efficiencies of optical sensing and detection.

Originally developed as a microscopy tool, near-field optics has steadily matured in the quest for ever-higher resolution of molecules, plasmons, and localized fields. But it has also matured with the concomitant advancement of technology and researchers' ability to shrink and manipulate the shapes of probes at ever-smaller dimensions. Optical antennas are the most recent offspring of near-field optics and technology, but others are certain to come.

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NOMINATIONS ARE INVITED FOR THE

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Nominations are to be submitted as a single pdf file including a letter of nomination, two letters supporting the nomination, a curriculum vitae of the nominee, and a proposed citation for the award. Nominations must be received by 15 August 2011. Copies of the nomination material should be sent to the award committee chair Chris Quigg at quigg@fnal.gov and to the Assistant to the AIP Corporate Secretary, Ms. Melissa Poleski, at mpoleski@aip.org.

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