

Near-field microscopy: Optical antennae enable nanoscale microscopy and spectroscopy

Sharply pointed metal tips act as optical antennae that can produce optical images with resolutions to 10 nm and permit many forms of spectroscopy.

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Each generation of microscopes, from confocal to scanning-electron to transmission-electron, has brought better resolution, until finally atomic wavefunctions of atoms themselves are visible. However, spatial resolution alone has limited importance if it isn't complemented by a means to identify the composition of the sample.

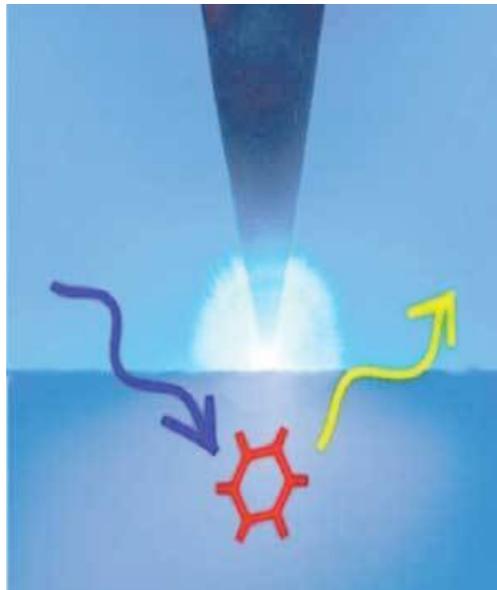
While spatial resolution refers to the question “where things are,” it does not answer anything about “what things are.” The latter question forms the traditional field of spectroscopy. Naturally, it is desirable to combine spectroscopy and high-resolution microscopy with the ultimate goal of resolving and chemically identifying each molecule within a sample. In this regard, antenna-based optical spectroscopy is a promising approach.

Standard optical microscopy uses conventional optics to direct and focus light onto a sample. While this simple concept might seem to leave little room for improvement, clearly this is not so, as evidenced by the recent microscopy revolution that gave birth to powerful techniques such as multiphoton microscopy, structured-illumination microscopy, stimulated-emission-depletion (STED) microscopy, coherent anti-Stokes Raman-scattering (CARS) microscopy, and fluorescence self-interference microscopy. Some of these techniques achieve very high spectroscopic information while others render high spatial resolution. But, because conventional optics redirects the wavefronts of propagating radiation, it is unable to sample the high spatial frequencies associated with evanescent waves and imposes a limit on either spatial resolution or spectroscopic information.

Optical analog of radio antennae

Antenna-based optical microscopy is a form of near-field optical microscopy that uses the analog to well-established concepts developed in the radio-wave and microwave regimes.¹ Essentially, an antenna efficiently converts the energy of free-propagating radiation to localized energy, and vice versa. For example, the antenna of a cell phone is used to concentrate the energy of incoming radiation onto a receiver chip with dimensions much smaller than the wavelength of the incoming radiation. Because it is a reciprocal device, the antenna also helps to efficiently release the energy from the receiver chip. Antennae are key enablers for many technologies, from satellite communication to baby phones. The introduction of the antenna concept into the optical-frequency regime will likely bring new technological applications.

In microscopy, an optical antenna basically replaces a conventional focusing lens (objective), thereby concentrating external laser radiation to dimensions much smaller than the diffraction limit of light (on the order of half the wavelength of light). In the technique, the localized antenna field is brought close to a sample surface (see Fig. 1). The response due to the local optical interaction is converted by the same antenna into propagating radiation and subsequently spectrally analyzed using standard optical instrumentation. A hyperspectral image of the sample surface can be recorded by guiding the antenna pixel-by-pixel over the sample surface and recording for each image pixel a corresponding spectrum.



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FIGURE 1. In antenna-based optical spectroscopy, an antenna, such as a metal tip or particle, localizes the incident radiation and gives rise to a local optical interaction with a sample surface. The response is converted by the same antenna into propagating radiation and subsequently spectrally analyzed.

Typically, a sharply pointed metal tip is used as an optical antenna, but other antenna geometries, such as particles and bowties, have also been studied. Antenna-based spectroscopy has been demonstrated using IR absorption, Raman scattering, coherent anti-Stokes Raman scattering, and fluorescence.²⁻⁹ Spatial resolutions down to 10 nm have been achieved while retaining the full power of optical spectroscopy. In one example, near-field Raman scattering using a bare metal tip as an optical antenna captures images of a single-walled carbon nanotube.

While radio and microwave antennae are pervasive in our lives, their optical analog has appeared only recently. Fabrication difficulties are largely to blame, since specifications would require tolerances smaller

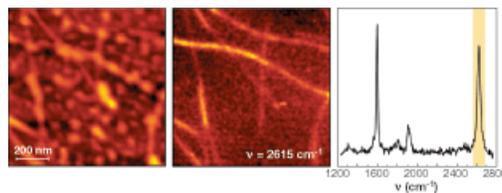
than 10 nm. However, this length scale becomes increasingly accessible with the use of novel top-down nanofabrication tools (focused-ion-beam milling, electron-beam lithography, and so on) and bottom-up self-assembly schemes. Therefore, the fabrication of optical-antenna structures is becoming a possibility not only for optical microscopy and spectroscopy, but also for optoelectronic-device architectures.

More complex than radio

A challenge associated with optical antennae is the fact that the penetration of radiation into metals can no longer be neglected and that collective electron resonances (surface plasmons) must be considered. These collective excitations make a direct downscaling of traditional antenna designs not possible and demand the careful study of surface modes in metal nanostructures (plasmonics).

The range of applications of antenna-based optical spectroscopy would be greatly enhanced if the technique were able to image underneath the surface of a sample. The structural characterization of buried nanoscale features in a nondestructive way is central to the understanding of biological processes and for the characterization of solid-state devices whose functional units are protected from interactions with the environment (capping layers). Antenna-based spectroscopy may seem to be limited to surface characterization. However, the localized fields at the antenna apex extend a couple of nanometers beyond the metal surface of the antenna and are able to penetrate through capping layers.

Using this property, the Nano-Optics group at the University of Rochester has recently demonstrated that carbon nanotubes buried underneath a 10 nm silicon dioxide layer can be imaged and spectroscopically identified via Raman scattering (see Fig. 2).¹⁰ The nanoscale imaging of subsurface features, their chemical identification, and three-dimensional reconstruction is the central theme of a project funded by the Air Force Office of Scientific Research (Arlington, VA) and including seven participating investigators from different universities.¹¹



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FIGURE 2. Near-field Raman imaging of a single-walled carbon-nanotube sample reveals topography showing a network of carbon nanotubes overgrown with water droplets (left). A Raman image of the same sample is recorded by integrating, for each image pixel, the photon counts that fall into a narrow spectral bandwidth centered around $\nu = 2615 \text{ cm}^{-2}$ (center). The Raman-scattering spectrum recorded on top of the nanotube appears at right, with the narrow spectral band of the integrated photons shown in yellow.

Once mature, antenna-based spectroscopy will be able to provide high-resolution chemically specific maps of surfaces and subsurface features. Potential applications include biomedical imaging and sensing, stress analysis and dopant profiling in integrated circuits, and chemical analysis of nanocomposite materials.

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Laser Focus World July, 2006

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