involve zero angular-momentum change
for a strained semiconductor, the photons
should be entangled in right and left
circular-polarization states. So far, the
most widely used source of entangled
photons for quantum information
research uses spontaneous parametric
down-conversion9,10, a process in which
pump photons from a laser are converted
into signal and idler photons in a non-
centrosymmetric material. Because the
down-conversion process is non-resonant
(no electronic states are ever populated)
and optically nonlinear, even with
femtosecond laser pumping and phase
matching, it is orders of magnitude weaker
than the resonant process discussed by
Hayat et al. Semiconductor 2PE is based
on a resonant emission process, involving
a potentially high electron density that
can be excited by incoherent optical or
electrical pumping processes. And, just
as appropriately designed microcavities
can be used to enhance the parametric
down-conversion process as well as narrow
the bandwidth of emission, 2PE from
semiconductors can be similarly enhanced
and tailored, using a cavity or embedded
photonic crystal, while suppressing single-
photon emission.

The early results are promising. It’s
possible that a compact, electrically
pumped, efficient source of entangled
photons may make the laser-pumped
parametric crystal redundant in the
same way that the transistor superseded
the vacuum tube. However, much work
remains to be done.

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OPTICAL ANTENNAS

Redirecting single molecules

Optical antennas have already been shown to dramatically enhance molecular excitation
and emission processes. Now, a compelling new study illustrates how they can redirect
the emission of single molecules.

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Controlling the direction of light
emission is often a critical step
in achieving intense optical
sources — just ask anyone who has ever
aligned the mirrors in a laser cavity. After
light leaves the cavity, directionality may
appear to be an inherent property
of electromagnetic radiation. However, a
laser’s intense, spatially coherent beam
is produced by directing the collective
decay of innumerable excited electrons
into a well defined mode. As researchers
scale down photonic devices and explore
emission from individual quantum dots
and single molecules, it has become
abundantly clear that the light emitted by
a discrete photon source is neither easy
to observe nor direct. On page 234 of this
issue, Tim Taminiau and colleagues in the
laboratory of Niek van Hulst at ICFO, the
Institute of Photonic Sciences in Barcelona,
demonstrate how an optical antenna can
help to both observe and redirect light
emission from single molecules.

Single molecules are generally
directional emitters. In free space, the
electric-dipole transitions in most
fluorescent molecules emit light of a
fixed polarization into well-defined
dipolar modes. In 1993, Eric Betzig and
Robert Chichester capitalized on the
directional nature of dipolar transitions
to produce a seminal work in the field
of single-molecule detection2. Using a
near-field scanning optical microscope,
they imaged the fluorescent emission
of discrete carbocyanine molecules
immobilized on a polymer substrate.
Rather than observing similar near-field
images for each molecule, they discovered
that each dipole produced a unique image.
The strength with which each molecule
was excited depended on its position and
orientation relative to the fields emerging
from the near-field probe. By investigating
the excitation efficiency as a function of
position, Betzig and Chichester were able
to determine the precise orientation of each
molecular dipole.

As near-field techniques improved, it
became clear that nearby objects, including
the probe itself, could dramatically
influence the emission process. In
2000, Gersen et al. demonstrated that
proximity to the metallized aperture of a
near-field probe could shift the angular
emission of single molecules from side to
side3. This subtle change suggested that
local optical perturbations could help
shape the emission of a single molecule.
However, the redirection of angular
emission was dependent on the original
dipole orientation, and the dominant
emission shape was still determined by the
molecule itself. To truly redirect molecular
emission requires coupling the original
molecule to a second, much more efficient
radiating system.

In the microwave and radiowave
regime, efficient and directional
electromagnetic radiation is often
mediated by an antenna. Although any
time-varying current can serve as the
source of electromagnetic radiation, some
sources are much more efficient than others.
For example, a hertzian dipole (an infinitesimal
oscillating current source) is much less
efficient at radiating energy than the longer,
half-wavelength, dipole antenna. The added
length of the dipole antenna enables the
current source to more effectively couple
to free-space radiation modes (Fig. 1a).

By connecting an infinitesimal current
source to an elongated dipole antenna, the
resulting electromagnetic radiation can be
both amplified and redirected.

Over the past few years, considerable
progress has been made in realizing
resonant optical analogues to conventional
antenna designs4. In particular, the dipole
modes supported by metal nanoparticles

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have proven to be very efficient optical antennas\textsuperscript{5,6}. They have been able to modify the excitation efficiency, quantum yield and decay rates of single fluorescent molecules. However, as a result of their radial symmetry, metal nanoparticles can support dipole modes in all directions, and the dominant emission modes tend to vary with relative molecule orientation and particle position. To reorient molecular emission along a fixed direction, a resonant optical antenna with a fixed emission pattern is needed.

By incorporating an elongated metal rod onto the tip of an apertured near-field optical probe, this is precisely what Taminiau \textit{et al.} have achieved: they have coupled individual molecular dipole sources to a highly directional optical antenna\textsuperscript{7}. Given the finite conductivity of metals at visible frequencies\textsuperscript{7}, as well as the tip’s proximity to the extended metal coating of the near-field probe, the length of the optical antenna used for their experiments deviates from the classical half-wavelength antenna. However, in understanding the observed redirection of molecular emission, the elongated rod may be modelled as a dipole antenna. As the antenna is scanned over the fluorescent molecule, the emission pattern is effectively reoriented.

Exploiting the tip-on-aperture geometry of their near-field probe, Taminiau \textit{et al.} were able to selectively probe emission from individual carbocyanine molecules as they were coupled and uncoupled to the optical antenna. Scanning the near-field probe over the sample produced two distinct but adjacent images for each molecule: (1) a broad emission pattern characteristic of the uncoupled molecule excited by the central aperture fields far from the optical antenna, and (2) a highly focused pattern emitted when each molecule became coupled to the optical antenna. Using a polarizing beam splitter and two photodetectors, the researchers were able to monitor the relative polarization of molecular emission. Although the uncoupled emission patterns confirmed that the molecules were oriented in a variety of directions, the coupled emission patterns demonstrated a uniformity of polarization consistent with a dipole emitter oriented along the antenna’s long axis (Fig. 1b). Comparing the coupled and uncoupled emission patterns, the team presents compelling evidence that an optical antenna can redirect the emission from a single molecule.

These experimental results will enable many new studies in the field of single-molecule detection. Indeed, the probe design and comparative analysis of intrinsic and perturbed molecular emission provide essential tools for a more rigorous level of experimental study. Moreover, the robust fabrication scheme developed for these resonant antenna probes ensures their future use by other researchers.

The ability to direct and shape molecular emission could also have profound implications for nanophotonic devices and single-photon sources. In particular, as we attempt to harness the quantum nature of single-photon events, antenna designs to control spontaneous emission offer complements to the traditional stimulated-decay engineering of laser cavities. By coupling single molecules to optical antennas and perhaps optical antenna arrays\textsuperscript{8}, it might be possible to truly shape the emission of single photons. When demonstrating the first room-temperature, molecular, single-photon sources, Brahim Lounis and William Moerner specifically noted the difficult relationship between directionality and intensity for molecular emission\textsuperscript{8}. “Photons,” they wrote, “are emitted into a range of solid angles, which can limit the detection efficiency.” As a possible solution, they suggested coupling single molecules to an optical cavity to help “reduce losses, change the emission pattern, or modify the emission lifetime.” Since the publication of these words in 2000, optical antennas have made significant advances towards modifying emission lifetimes and, now, changing the emission pattern. If designs could be developed that reduce losses, resonant optical antennas might truly become nanoscale alternatives to resonant optical cavities.

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