involves 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-conversion[5,10], 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source of entangled photons may make the laser-pumped parametric crystal redundant in the same way that the transistor superseded the vacuum tube.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[1].

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 detection[2]. 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 side[3]. 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 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 designs[4]. In particular, the dipole modes supported by metal nanoparticles

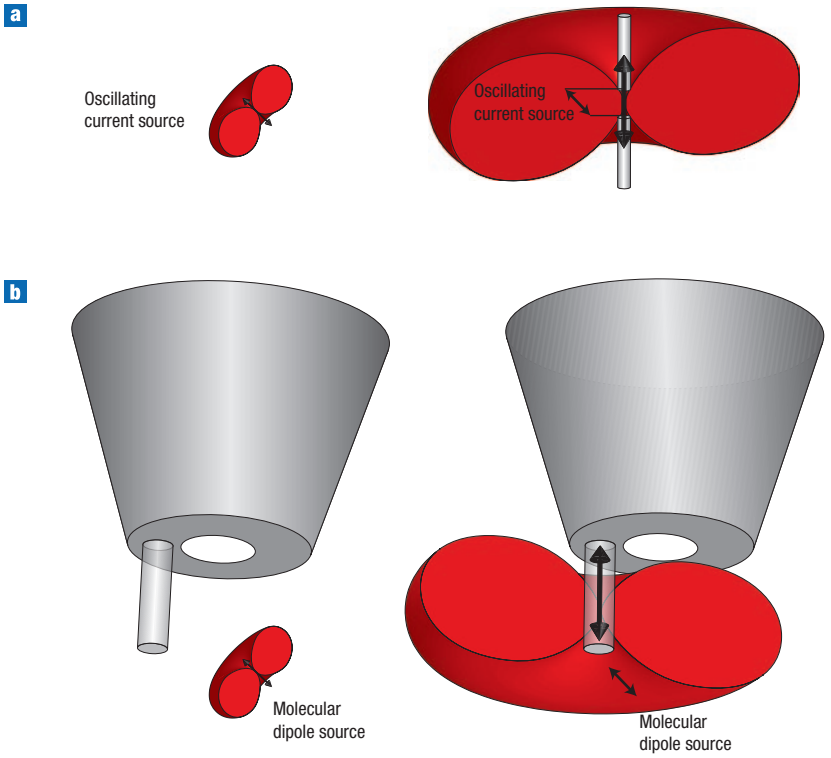

Figure 1 Antennas amplifying and redirecting emission. **a**, An oscillating point source (left) is not an efficient radiator, but when that current source is wired to a half-wavelength antenna (right), emission from the coupled system is amplified and redirected into the dipolar mode of the antenna. **b**, Likewise, the emission from a single fluorescent molecule (left) can be amplified and redirected by coupling to the optical antenna at the tip of a near-field probe (right). Note that by coupling the molecule to a highly directional antenna, the coupled system acquires the directionality of the antenna.

have proven to be very efficient optical antennas[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 *et al.* have achieved: they have coupled individual molecular dipole sources to a highly directional optical antenna[1]. Given the finite conductivity of metals at visible frequencies[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 *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[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[9]. "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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