

Optical antenna enhanced spontaneous emission

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Atoms and molecules are too small to act as efficient antennas for their own emission wavelengths. By providing an external optical antenna, the balance can be shifted; spontaneous emission could become faster than stimulated emission, which is handicapped by practically achievable pump intensities. In our experiments, InGaAsP nanorods emitting at ~ 200 THz optical frequency show a spontaneous emission intensity enhancement of $35\times$ corresponding to a spontaneous emission rate speedup $\sim 115\times$, for antenna gap spacing, $d = 40$ nm. Classical antenna theory predicts $\sim 2,500\times$ spontaneous emission speedup at $d \sim 10$ nm, proportional to $1/d^2$. Unfortunately, at $d < 10$ nm, antenna efficiency drops below 50%, owing to optical spreading resistance, exacerbated by the anomalous skin effect (electron surface collisions). Quantum dipole oscillations in the emitter excited state produce an optical ac equivalent circuit current, $I_o = q\omega|x_o|/d$, feeding the antenna-enhanced spontaneous emission, where $q|x_o|$ is the dipole matrix element. Despite the quantum-mechanical origin of the drive current, antenna theory makes no reference to the Purcell effect nor to local density of states models. Moreover, plasmonic effects are minor at 200 THz, producing only a small shift of antenna resonance frequency.

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Antennas emerged at the dawn of radio, concentrating electromagnetic energy within a small volume $\ll \lambda^3$, enabling nonlinear radio detection. Such coherent detection is essential for radio receivers and has been used since the time of Hertz (1). Conversely, an antenna can efficiently extract radiation from a subwavelength source, such as a small cellphone. Despite the importance of radio antennas, 100 y went by before optical antennas began to be used to help extract optical frequency radiation from very small sources such as dye molecules (2–10) and quantum dots (11–14).

In optics, spontaneous emission is caused by dipole oscillations in the excited state of atoms, molecules, or quantum dots. The main problem is that a molecule is far too small to act as an efficient antenna for its own electromagnetic radiation. Antenna length, l , makes a huge difference in radiation rate. An ideal antenna would preferably be $\lambda/2$, a half-wavelength in size. To the degree that an atomic dipole of length l is smaller than $\lambda/2$, the antenna radiation rate $\Delta\omega$ is proportional to $\omega(l/\lambda)^3$, as given by the Wheeler limit (15). Spontaneous emission from molecular-sized radiators is thus slowed by many orders of magnitude, because radiation wavelengths are much larger than the atoms themselves. Therefore, the key to speeding up spontaneous emission is to couple the radiating molecule to a proper antenna of sufficient size.

Since the emergence of lasers in 1960, stimulated emission has been faster than spontaneous emission. Now the opposite is possible. In the right circumstances, antenna-enhanced spontaneous emission could become faster than stimulated emission. Theoretically, very large bandwidth >100 GHz or >1 THz is possible when the light emitter is coupled to a proper optical antenna (16).

Metal optics have been able to shrink lasers to the nanoscale (17–20), but high losses in metal-based cavities make it increasingly difficult to achieve desirable performance. Metal structures have also been used to enhance the spontaneous emission rate, such as by coupling excited material to flat surface plasmon

waves (21–28). Flat metal surfaces are far from ideal antennas, resulting in low radiation efficiencies and large ohmic losses. Semiconductor emitters have been further limited by large surface recombination losses and by processing difficulties at the extremely small dimensions. Semiconductor experiments (29, 30) show weak antenna–emitter coupling, with the antenna enhancement sometimes masked by metal-induced elastic scattering that enhances light extraction from the semiconductor substrate. Light extraction alone can increase optical emission by $4n^2$, as often used in commercial light-emitting diodes (LEDs), without necessarily modifying the spontaneous emission rate (31, 32).

In this article, we elucidate the physics of antenna-enhanced spontaneous emission, using a traditional antenna circuit model, not the Purcell effect (33) nor a local density-of-states model (34). We use the circuit approach to analyze for the maximum possible spontaneous emission enhancement in the presence of spreading resistance losses (35) and the nonlocal anomalous skin effect (36) in the metal.

We experimentally tested an optical dipole antenna, coupled to a “free-standing” 40-nm nanorod of semiconductor material. Thus far, optical emission measurements show a $>115\times$ antenna spontaneous emission rate enhancement factor compared with no antenna at all. At smaller dimensions, circuit theory predicts a spontaneous emission rate enhancement $>10^4\times$, but at the penalty of decreased antenna efficiency. Nonetheless, we will derive that $>2,500\times$ rate enhancement should be possible, while still maintaining antenna efficiency $>50\%$.

Antenna Physics

An antenna converts free space electromagnetic waves to ac currents in circuits and vice versa. At radio frequencies and at optical frequencies the antenna physics are quite similar. There are three important questions about antennas:

Significance

Since the invention of the laser over 50 y ago, stimulated emission has been stronger and far more important than spontaneous emission, the ordinary light we are accustomed to. Indeed spontaneous emission has been looked down upon as a weak effect. Now a new science of enhanced spontaneous emission is emerging that makes spontaneous emission faster than stimulated emission. This new science depends upon the use of optical antennas to increase the spontaneous emission rate. Antennas emerged at the dawn of radio for concentrating electromagnetic energy to a small volume. Despite the importance of radio antennas, 100 y went by before optical antennas began to be used to help extract optical frequency radiation from very small sources such as dye molecules and quantum dots.

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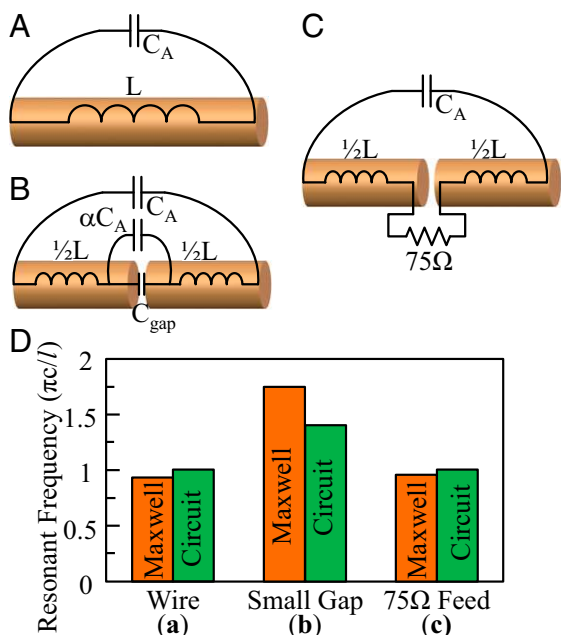


Fig. 1. Circuit models for common antenna geometries. (A) A straight wire. (B) A straight wire with a center gap, where a molecule or quantum dot could be inserted. (C) The case of a straight wire with a 75Ω load at the center. (D) The fundamental resonance frequency is in units of πcl , where l is the wire length. Orange bars are calculated from Maxwell's equations and green bars from a circuit model. In the calculation, the wire length $l = 1.5$ m, the wire diameter $= 2r = 1$ mm, and the gap spacing $= 0.5$ mm. The metal conductivity was taken to be $4.56 \times 10^5/\Omega\text{cm}$, which at 100 MHz translates to a complex dielectric constant, $\epsilon_m = 1 - j8.2 \times 10^9$. From Table 1, $L = (\mu_0/\pi^2)l \times \ln(l/r) = 1.4 \mu\text{H}$, $C_A = \epsilon_A l \times \ln(l/r) = 1.82$ pF, $C_{\text{gap}} = \epsilon_0 \pi r^2/d = 55.6$ fF. The stray capacitance of the wire region adjacent to the gap is αC_A , where $\alpha = 1$.

- For a given rms ac current, $I_{\text{rms}}(\omega)$, how much electromagnetic wave power P will be launched? Because radiated power is proportional to current squared, the question reduces to finding the coefficient R in $P = I^2 R$, where $R \equiv R_{\text{radiation}}$ or R_{rad} is called radiation resistance. $R_{\text{radiation}} = (2\pi/3)(l/\lambda)^2 \sqrt{(\mu_0/\epsilon_0)}$ for constant current in a wire length l , where $\sqrt{(\mu_0/\epsilon_0)} \sim 377\Omega$ is the fundamental impedance of free space. $R_{\text{radiation}}$ is derived in most antenna books (37, 38).
- What is the cross-section for an antenna capturing electromagnetic wave power from free space optical intensity and transferring it to a matched load? Surprisingly, antenna capture

cross-section has a universal answer for all antennas, no matter how small: $\text{Area} = \lambda^2/\Omega$, where λ is the resonant electromagnetic wavelength, and Ω is the acceptance solid angle of the antenna. $\Omega = 4\pi$ steradians for an isotropic antenna leading to a capture cross-section $\text{Area} = \lambda^2/4\pi$, whereas for dipoles $\Omega = (2/3)4\pi = 8\pi/3$ steradians, because z -directed radiation parallel to the dipole is forbidden, whereas x and y are allowed. A more highly directive antenna captures proportionately more power from an incident plane wave. By time reversal, captured power $P \equiv \text{Intensity} \times \text{Area}$ equals reradiated power $= I^2 R_{\text{radiation}} = V^2/R_{\text{radiation}}$. Thus, in a receiving antenna, there is an effective optical frequency voltage source $V_{\text{rms}}(\omega) = 2\sqrt{(\text{Intensity} \times \text{Area} \times R_{\text{radiation}})}$, where the 2 accounts for voltage division between $R_{\text{radiation}}$ and matched R_{load} . With $R_{\text{load}} = 0$, the reradiation cross-section $\text{Area} = 3\lambda^2/2\pi$. The equivalent circuit of a transmitting antenna will be shown (see Fig. 3).

- Every metallic structure is resonant at some frequency and is, in effect, an LC resonator, with distributed inductance L and capacitance C . Because self-oscillating currents lead to some electromagnetic radiation, every metal object is also an antenna. In optics we often seek the highest possible Q factor, but in antennas we seek the opposite because we desire radiation losses. Harold Wheeler (15) answered the question, What is the lowest Q factor that can be achieved in an antenna? This is called the Wheeler limit: $(\omega/\Delta\omega) \equiv Q \sim (3/4\pi^2)(\lambda/l)^3$, where l is the longest linear dimension of the antenna.

Thus, a small antenna is a poor antenna, with a high Q , but nonetheless still capturing over an $\text{Area} \sim \lambda^2/4\pi$, which can actually be much larger than its physical area. For example, a dipole atom is a very small resonator and has a high Q according to the Wheeler limit, but it scatters electromagnetic waves with a cross-sectional area, $\text{Area} = 3\lambda^2/2\pi$, much larger than the atomic size.

The lowest LC resonant frequency of a metallic object is usually related to its size. That is only the first of many electromagnetic resonances that extend all of the way to high optical frequencies, at which point the resonances could be influenced by kinetic inductance and assume some plasmonic character.

The oscillating ac currents associated with the LC resonators can radiate electromagnetic energy. Thus, all metal objects act as antennas to some degree, converting ac currents to radiated power.

For a given resonance, there is insight to be gained by visualizing the distributed inductance and capacitance as a circuit. In Fig. 1A we illustrate an LC circuit model associated with the lowest resonance of a straight wire, with $C_A = \epsilon_A l \times \ln(l/r)$ modeling the tip-to-tip capacitance of the wire, as documented in

Table 1. Antenna circuit parameters

Gap capacitance		Current induced in antenna	
Blunt tips (39)	$C_{\text{gap}} = \frac{\epsilon_0 A}{d}$	Parallel plate current induced by ac dipole (40)	$I_0 = \frac{q\omega x_0}{d}$
Round tips (41)	$C_{\text{gap}} = \frac{\pi\epsilon_0 r}{2} \left[\ln\left(\frac{l}{d}\right) + 2\gamma \right]$		
Lumped reactance terms		Resistance terms	
Faraday (38) inductance	$L_f = \frac{\mu_0}{\pi} l \times \ln\left(\frac{l}{r}\right)$	Radiation (38) resistance	$R_{\text{rad}} = \frac{2\pi}{3} Z_0 \left(\frac{l_{\text{eff}}}{\lambda}\right)^2 n_A$
Kinetic (42) inductance	$L_k = \frac{l_{\text{eff}}}{A} \text{Re} \left\{ \frac{1}{\omega^2 \epsilon_0 (1 - \epsilon_m)} \right\}$	Ohmic (42) resistance	$R_{\text{ohmic}} = \frac{l_{\text{eff}}}{A} \text{Im} \left\{ \frac{1}{\omega \epsilon_0 (1 - \epsilon_m)} \right\}$
Antenna (38) capacitance	$C_A = \frac{\epsilon_A l}{\ln\left(\frac{l}{r}\right)}$	Spreading (35, 42) resistance	$R_{\text{spread}} = \frac{1}{\beta d} \text{Im} \left\{ \frac{1}{\omega \epsilon_0 (1 - \epsilon_m)} \right\}$

Shown are circuit parameters of wire antennas, where r = wire radius, l = antenna length, l_{eff} = effective antenna length accounting for current $\rightarrow 0$ at ends; for a half-wave antenna $l_{\text{eff}} = 0.64l$; A = antenna wire cross-sectional area, $Z_0 \equiv \sqrt{(\mu_0/\epsilon_0)}$, the impedance of free space; x_0 = optical ac peak dipole moment length centered in a gap-spacing d ; βd is the diameter on the adjacent electrodes over which the dipole currents spread ($\beta = 1.6$ for flat electrodes); n_A and ϵ_A are the refractive index and dielectric constant of the medium surrounding the antenna; ϵ_m = metal relative dielectric constant; and ϵ_g = dielectric constant in the gap.

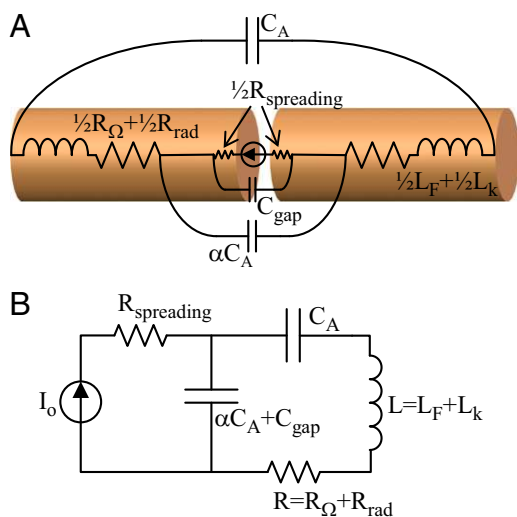


Fig. 3. Optical antenna circuit model. (A) Circuit components of an optical antenna, where R_{Ω} is the ohmic resistance, R_{rad} is the radiation resistance, L_k is the kinetic inductance, L_F is the Faraday inductance, C_{gap} is the internal gap capacitance $= \epsilon_0 \pi r^2 / d$ for blunt tips of radius r , C_A is the external tip-to-tip capacitance, and αC_A is the stray capacitance between internal tips. We find $\alpha C_A \sim C_A$. (B) The same circuit in a simplified schematic.

that a small gap d is desired, but not to the point that $C_{\text{gap}} > \alpha C_A$. R_{rad} and $|1/\omega \alpha C_A|$ are closely related to Z_0 . There is also an $R_{\text{rad}}/(R_{\text{ohmic}} + R_{\text{rad}})$ impedance matching term (46).

The efficiency from the circuit model of Fig. 3 is

$$\text{Efficiency} = \frac{R_{\text{rad}}}{R + R^2 (C_A/L) (\alpha + C_{\text{gap}}/C_A) (1 + \alpha + C_{\text{gap}}/C_A) R_{\text{spread}}},$$

where $R \equiv R_{\text{ohmic}} + R_{\text{rad}}$. Neglecting the spreading resistance, R_{spread} , the efficiency is that of a normal resistive voltage divider. At small d , R_{spread} is increased by surface collisions (the anomalous skin effect), and R_{spread} limits the efficiency.

Experiment

To demonstrate antenna-enhanced spontaneous emission, many of our experiments have used the arch antenna, as shown in Fig. 5. This antenna works by introducing an inductor across the gap that can be used to diminish the effective gap capacitance of the antenna. InGaAsP is the spontaneous light-emitting material. The antenna was created by wet etching an InGaAsP film, parallel to crystal planes to create high aspect ratio sidewalls. The InGaAsP surface was coated with 3 nm TiO_2 , using atomic layer deposition to provide electrical isolation between the InGaAsP surface and the gold antenna. Finally, a bar of gold was then deposited perpendicularly over the square nanorod to create the antenna and the inductor.

The fabricated structures were characterized by measuring the amount of light emitted. Increased spontaneous emission intensity, in competition with nonradiative losses, directly measures spontaneous emission enhancement. This measure of spontaneous emission rate is complementary to lifetime measurements. Lifetime can be influenced by emitters coupling to surface plasmons or optical cavities, which may not radiate, the energy still being lost as heat (21, 27), whereas the intensity method demands a knowledge of relative pump efficiency, relative emission pattern, and possible difference in internal quantum efficiency near the antenna.

In our experiment the dominant nonradiative mechanism is surface recombination. The quantum efficiency (QE) of the emitter is equal to the ratio of the radiative recombination rate to the

surface recombination rate. With equivalent pumping and surface recombination conditions, an increase in optical emission is therefore a direct measurement of the increased rate of spontaneous emission into the far field. Although the nonradiative losses are undesirable in a device, they are a type of clock for fundamental measurements.

To ensure that nonradiative rates dominate, the InGaAsP surface was not treated with any form of passivation layer. Bare InGaAsP layers have been rigorously studied before and have been shown to have a surface recombination velocity of $\sim 3 \times 10^4$ cm/s (47, 48). Considering the ~ 34 -nm width of the InGaAsP nanorods, this corresponds to a surface recombination lifetime of ~ 28 ps, which should be compared with InGaAsP's unenhanced spontaneous emission lifetime of >10 ns. The nanorods are covered in a 3-nm TiO_2 layer that increases the surface recombination rate by an additional factor of 3 (*Supporting Information*) and prevents direct contact between the InGaAsP and the gold antennas. This ensures that the only change in nonradiative recombination will be from ohmic losses in the antenna and not from changes in surface recombination. Although chemical surface passivation or cladding layers would greatly increase the light emission from this structure, they would impair the proper measurement of antenna properties, which is the main goal of this study.

It is also necessary to eliminate mechanisms that enhance external quantum efficiency without increasing the spontaneous emission rate, most notably improved light extraction. For a traditional LED, the majority of light gets trapped in the semiconductor and only a very small emission angle can escape. By using a textured surface or grating, the light extraction can be increased by up to $4n^2$, where n is the refractive index of the semiconductor (31). Arrays of optical antennas can provide scattering extraction enhancement, thereby increasing light output, but without increasing the emission rate. By completely removing the substrate and leaving only very small, subwavelength semiconductor islands, the possibility of scattering light extraction enhancement is removed.

To excite carriers in the semiconductor nanorods, a Ti:Sapphire laser at $\lambda = 720$ nm center wavelength, 120 fs pulse width, 13.3 MHz repetition rate, and 20 μW average power was focused onto the sample. The spot size of the pump laser was $\sim 2 \mu\text{m}$, within which approximately four devices were probed simultaneously. The pump was polarized off axis to the antenna to prevent antenna-enhanced pumping. Spontaneously emitted light was collected

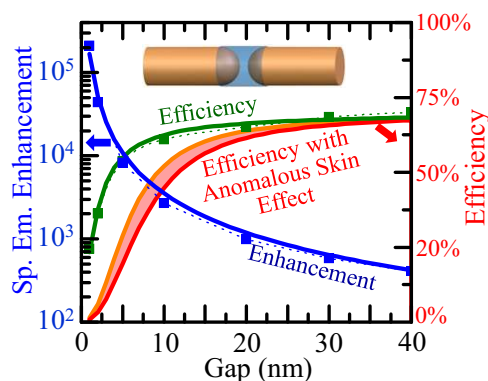


Fig. 4. Enhancement (blue) and efficiency (green) of an Au-wire antenna (radius $r = 20$ nm) with rounded inner tips and a semiconductor filled gap (light blue, refractive index $= 3.4$). The harmful effect of anomalous skin effect is plotted as the shaded red region bounded by $\delta = 1$ (orange) and by $\delta = 0.5$ (red). Solid lines are derived from the circuit model. Solid boxes connected by dotted lines are obtained from 3D FDTD simulations (CST; Lumerical). The antenna length l is adjusted to maintain 200 THz resonant frequency. For rounded electrodes, the current spreading factor $\beta \sim 0.5$ is a weak function of d/r in this range.

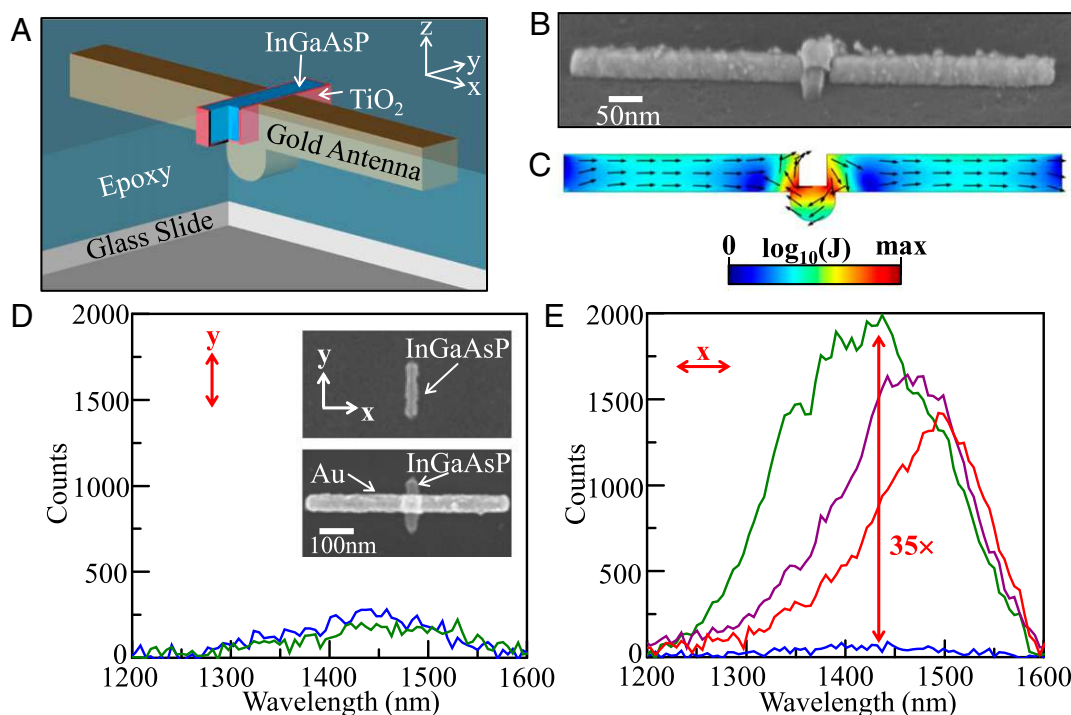


Fig. 5. Antenna-enhanced spontaneous emission from InGaAsP nanorods. (A) Schematic cutaway view of an arch-antenna-coupled InGaAsP nanorod, isolated by TiO_2 , and embedded in epoxy. (B) Perspective SEM of antenna-coupled nanorod before substrate removal. (C) Current density of the arch-antenna electromagnetic mode showing the antiparallel current in the arch compared with the arms of the antenna. (D) Optical emission polarized in the unenhanced y direction for bare nanorod (blue) and antenna-coupled nanorod (green). (Inset) Top-down SEM image of antenna-coupled and bare nanorod. (E) Optical emission polarized in the enhanced x direction for a bare nanorod (blue) and from nanorods coupled to different antenna lengths: 400 nm (green), 600 nm (purple), and 800 nm (red) in length.

through the pump lens and passed through a polarizer to discriminate light polarized parallel or perpendicular to the antenna. The light was resolved by a spectrometer (Princeton Instruments Acton SP2300i) and detected on a cooled InGaAs CCD (Princeton Instruments OMA V).

Fig. 5D shows the optical emission spectrum collected for the two orthogonal emission polarizations for both the bare and the antenna-coupled InGaAsP nanorods. A 400-nm-long, 50-nm-wide antenna has little effect on the spontaneous emission polarized in the unenhanced direction, indicating that the presence of the antenna does not affect the pumping or surface recombination of the semiconductor nanorod. An improvement of 35 \times , however, is seen for light emitted polarized in the x direction (Fig. 5E) relative to the bare nanorod. The enhancement is spectrally broad, spanning almost 200 nm of vacuum wavelength, indicating an antenna Q on the order of ~ 5 . As the antenna arms are lengthened the antenna resonance red shifts and a corresponding shift in the enhanced spectrum can be observed. The red shift of the antenna resonance is affected by the LC matching arch. Longer antennas show spectral mismatch between the antenna resonance and the InGaAsP spontaneous emission peak.

Ideally, carriers generated in the nanorod would equilibrate with volume under the arch of the antenna, where the highest enhancement occurs. Unfortunately, the diffusion length is insufficient compared with the nanorod length. Because the recombination lifetime is limited to ~ 10 ps by surface recombination, the diffusion length for holes in InGaAsP can be calculated as ~ 16 nm, using 1/10th the bulk hole mobility of $10 \text{ cm}^2/\text{Vs}$ (49). Consequently, a large portion of the carriers will never see the antenna arch before recombining. Given that the exposed InGaAsP arms are ~ 50 nm long, compared with a diffusion length of ~ 16 nm, only $\sim 32\%$ of the carriers will diffuse to the antenna hotspot. Fabricating shorter nanorods will reduce this effect, but the devices

in this study were limited by alignment tolerances. Taking this factor into consideration, the observed enhancement ratio of 35 \times corresponds to an actual enhancement greater than 115 \times .

Fig. 4 predicts an $\sim 400\times$ enhancement factor for a 40-nm gap at the central vertex of the antenna. In the experiment, the optical electric field drops across ~ 6 nm of TiO_2 before reaching the ~ 34 nm of InGaAsP. Only a fraction of the antenna voltage drops across the semiconductor. The optical voltage division ratio is $34/(34 + 6\gamma) = 0.74$, where $\gamma \sim 2$ is the dielectric constant ratio of InGaAsP/ TiO_2 . Thus, the predicted $400\times$ enhancement factor becomes $400(0.74)^2 \sim 220\times$, where light emission depends on electric field amplitude squared. The remaining experimental/theoretical discrepancy of $\sim 1/2$ could be associated with spatial averaging away from the optimal central spot under the arch antenna.

Because half-wave antennas can have radiation $Q \sim 1$, radiation competes well with ohmic losses. We calculate an antenna efficiency of 66%.

In summary, we have demonstrated an enhanced spontaneous emission rate from InGaAsP nanorods coupled to an optical antenna, directly observing spontaneous emission intensity enhancement of 35 \times . This corresponds to a spontaneous emission rate enhancement of 115 \times .

Directly modulated semiconductor lasers are $\sim 200\times$ faster than spontaneously emitting light-emitting diodes. If we can increase the optical antenna enhancement ratio to 200 \times , that will be a landmark in the competition between spontaneous emission and stimulated emission.

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