Supporting Information

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SI Text

Optimum Silver Film Thickness

Using numerical modeling, the thickness of a silver film on pyramid structures is roughly optimized. Fig. S1 shows the LDOS enhancement with respect to the dipole in a bare pyramid structure and the extraction efficiency of plasmonic modes as a function of the thickness of the silver film. Both are averaged over the spectral range of our QDs. In this work, for the demonstration of self-aligned nanofocusing, we chose a 40-nm-thick film, which is optimum for both LDOS enhancement (~19 for truncated pyramid QDs emitting at ~500 nm) and extraction efficiency (~16%). Extraction efficiency would increase with increasing wavelength (e.g., ~70% at 580 nm).

Temperature Dependence of Single QD Emission

Fig. S2 shows the temperature dependence of the PL measured for a single silver-coated pyramid structure. The sharp QD line is superimposed on the broadband emission from facet quantum wells (QWs). It gradually broadens with increasing temperature to merge into the QW background at around 200 K. We believe that measurements could be extended to room temperature in optimized structures, as recently demonstrated by Holmes et al. (1) with strongly confined InGaN pyramid QDs (height < 1 nm, compared with 5–7 nm in this work).

Quantitative PL Measurements of Spontaneous Emission Enhancement

Optical pumping and decays of pyramid QDs with and without the silver film are schematically represented in Fig. S3. Carriers are photogenerated in continuum bands of apex QD and facet QWs. Thus, QDs are excited nonresonantly through their continuum bands, and carrier transfers from nearby quantum wells (located at close pyramid facets). Efficiency of this nonresonant pumping would probably change upon deposition of the silver film. However, under saturation pumping condition, photoluminescence properties of QDs are independent of the pumping efficiency.

As-grown QDs spontaneously decay through a radiative channel with a rate Γ_r and a nonradiative channel with a rate $\Gamma_{nr}(T)$. With a silver film, QDs strongly interact with plasmonic modes, and we have a modified spontaneous decay channel owing to plasmonic modes, characterized by a decay rate Γ_p^{Ag} (here, spontaneous decay of QD into nanofocused plasmonic modes is much stronger than direct radiative decay into free space). In turn, plasmonic modes can be scattered into the far field with a rate κ_r^p or absorbed in the near field (heat loss) with a rate κ_{nr}^p, which are temperature independent because real and imaginary parts of the refractive index of silver are temperature independent in case of thin films (2). Nonradiative decay of exciton in QDs with silver film would have both temperature-dependent and -independent nonradiative decay processes. $\Gamma_{nr}^{Ag}(T)$ is related to thermally activated carrier escape out of the QD, and Γ_q^{Ag} is related to the metallic (nonplasmonic) quenching (e.g., interband absorption, electron scattering losses, and electron–hole excitations) (3). In this study, we assume that Γ_q^{Ag} is negligible in our system because Γ_q^{Ag} is usually dominant at the distance between metal and emitter below 5 nm, whereas our distance between QD and metal surface is sufficiently long (~10 nm) (3, 4). In addition, the large increase (× 13) in the saturated PL intensity of QDs under CW pumping with silver film indicates our decay rate enhancement is dominated by Purcell enhancement (Γ_p^{Ag}) rather than quenching into metal surface (Γ_q^{Ag}).

We can define PL intensity of QD as follows:

$$\mathrm{I_{PL}} = a \times \mathrm{P_{laser}} \times \eta_{\mathrm{in}} \times \mathrm{IQE} \times \eta_{\mathrm{ext}} \times \pi_0,$$

where a is a constant depending on the micro-PL setup, $\mathrm{P_{laser}}$ is a laser pumping power, η_{in} is an in-coupling efficiency of the excitation laser on QD, IQE is internal quantum efficiency of QD, η_{ext} is a light extraction efficiency to the far field, and π_0 is the far-field radiation pattern of QD within the measuring solid angle of the micro-PL setup.

Under saturation pumping conditions at low temperature where $\Gamma_{nr}^{Ag}(T)$ and Γ_q^{Ag} are negligible ($\mathrm{IQE} = [\Gamma_p^{Ag}/(\Gamma_p^{Ag} + \Gamma_{nr}^{Ag}(T) + \Gamma_q^{Ag})] = 1$; Fig. 3*D*), it can be shown that:

For CW pumping, the saturation pumping power is directly proportional to the SE decay rate, so that

$$\mathrm{CW_0} \propto a \times \Gamma_r \times \eta_{\mathrm{ext0}} \times \pi_0,$$

for the saturated PL intensity of as grown QDs,

$$\mathrm{CW_1} \propto a \times \Gamma_p^{Ag} \times \eta_{\mathrm{ext1}} \times \pi_1,$$

for the saturated PL intensity of silver-coated QDs, where a is a constant depending on the micro-PL setup, η_{ext0} and η_{ext1} ($=\kappa_r^p/\Gamma_p^{Ag}$) are light extraction efficiencies to the far field for as-grown QDs and silver-coated QDs, respectively. Note that the saturated PL intensity is governed by both SE rate and out-coupling efficiency ($\eta_{\mathrm{ext}} \times \pi$).

For pulsed pumping (when pulse duration time is much longer than QD's decay times), the time-integrated PL decay signal is independent of the SE decay rate, so that

$$\mathrm{P_0} \propto b \times \eta_{\mathrm{ext0}} \times \pi_0,$$

for the saturated PL intensity of as grown QDs,

$$\mathrm{P_1} \propto b \times \eta_{\mathrm{ext1}} \times \pi_1,$$

for the saturated PL intensity of silver-coated QDs, where b is a constant depending on the micro-PL setup. Note that the saturated PL intensity is governed by only out-coupling efficiency.

Combining these relationships, we can express enhancement of the spontaneous emission rate by the following:

$$\Gamma_p^{Ag} \Big/ \Gamma_r = (\mathrm{CW_1}/\mathrm{CW_0}) \times (\mathrm{P_0}/\mathrm{P_1}).$$

Experimentally, we measured $\mathrm{CW_1}/\mathrm{CW_0} = 13 \pm 8$ and $\mathrm{P_1}/\mathrm{P_0} = 0.5 \pm 0.3$, which yields $\Gamma_p^{Ag}/\Gamma_r = 26 \pm 22$. The very large data dispersion is due to the strong dependence of LDOS and extraction efficiency on the pyramid sharpness (Fig. 4).

Spontaneous emission enhancement can also be deduced from decay time measurements. The temperature dependence study of QD dynamics shows that $\Gamma_{nr}^{Ag}(T)$ can be neglected at low temperature (Fig. 3*D*). Therefore, the measured decay times at low temperature are given by the following:

$$\tau_0 \approx 1/\Gamma_r,$$

measured decay time of as grown QDs,

$$\tau_1 \approx 1 \Big/ \Gamma_p^{Ag},$$

measured decay time of silver-coated QDs.

Now the spontaneous emission enhancement can be expressed in terms of measured decay times by the following: $\Gamma_p^{Ag}/\Gamma_r = \tau_0/\tau_1$. Experimentally, we measured $\tau_0 = 4.4 \pm 2.2$ ns and $\tau_1 = 0.2 \pm 0.1$ ns, yielding $\Gamma_p^{Ag}/\Gamma_r = 22 \pm 16$, in good agreement with saturation PL measurements.

1. Holmes MJ, Choi K, Kako S, Arita M, Arakawa Y (2014) Room-temperature triggered single photon emission from a III-nitride site-controlled nanowire quantum dot. *Nano Lett* 14(2):982–986.
2. Mayy M, Zhu G, Mayy E, Webb A, Noginov M (2012) Low temperature studies of surface plasmon polaritons in silver films. *J Appl Phys* 111(9):094103.
3. Pelton M, Bryant GW (2013) *Introduction to Metal-Nanoparticle Plasmonics* (Wiley, Hoboken, NJ), Vol 5.
4. Ford GW, Weber W (1984) Electromagnetic interactions of molecules with metal surfaces. *Phys Rep* 113(4):195–287.

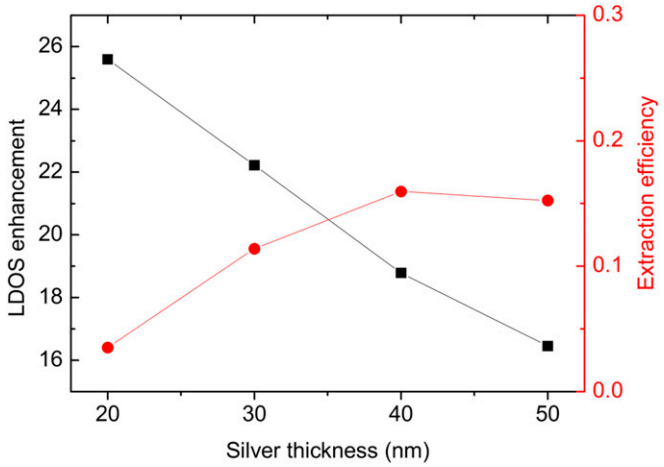

Fig. S1. The LDOS and extraction efficiency for a dipole in a truncated pyramid with $h/h_0 = 0.8$, emitting at ~500 nm, as function of the silver film thickness.

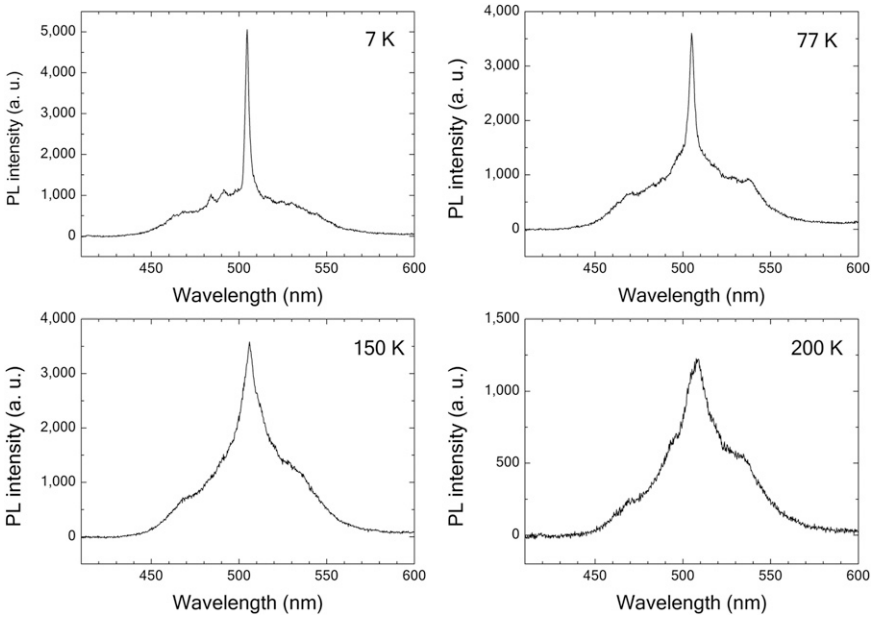

Fig. S2. Temperature dependence of the PL from a single silver-coated pyramid structure. The sharp line and broad band are due to emissions from the QD at the pyramid apex and from facet QWs, respectively.

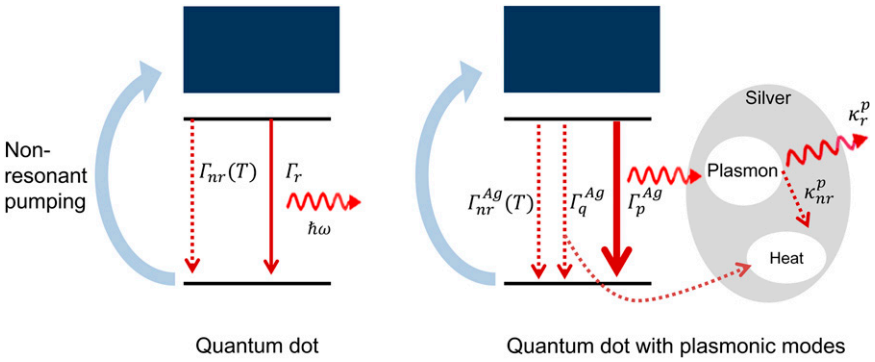

Fig. S3. Schematic representation of pyramid QDs with and without the silver film. Γ_r is spontaneous radiative decay rate, $\Gamma_{nr}(T)$ is nonradiative decay rate, Γ_p^{Ag} is spontaneous emission rate into plasmonic modes, $\Gamma_{nr}^{Ag}(T)$ is nonradiative decay rate with metal film (thermally activated carrier escape out of the QD), Γ_q^{Ag} is metallic (nonplasmonic) quenching, κ_{nr}^p is nonradiative (heat) optical loss of plasmonic modes, κ_r^p is radiative decay rate of plasmonic modes into far field. The QD is nonresonantly pumped (light blue arrows). This nonresonant pumping could be changed in the presence of the silver film (not represented in the figure).