Influence of Metal Deposition on Exciton—Surface Plasmon Polariton Coupling in GaAs/AlAs/GaAs Core—Shell Nanowires Studied with Time-Resolved Cathodoluminescence

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ABSTRACT: The coupling of excitons to surface plasmon polaritons (SPPs) in Au- and Al-coated GaAs/AlAs/GaAs core—shell nanowires, possessing diameters of ~100 nm, was probed using time-resolved cathodoluminescence (CL). Excitons were generated in the metal coated nanowires by injecting a pulsed high-energy electron beam through the thin metal films. The Purcell enhancement factor (FP) was obtained by direct measurement of changes in the temperature-dependent radiative lifetime caused by the nanowire exciton-SPP coupling and compared with a model that takes into account the dependence of FP on the distance from the metal film and the thickness of the film covering the GaAs nanowires.

KEYWORDS: GaAs/AlAs core—shell nanowires, surface plasmon polariton, cathodoluminescence

An important field in nanotechnology research is the design of plasmonic metal/semiconductor composite structures to obtain novel optical device properties for a variety of applications in biological labeling/sensing,† light-emitting diodes,† light-emitting lasers,† and solar cells.† By an appropriate choice of a metal possessing a surface plasmon frequency, ωsp, which is reasonably close to the excitonic transition energy of the nanocrystal, exciton coupling to surface plasmon polaritons (SPPs) can be enhanced. The enhancement of the photoluminescence (PL) efficiency of InGaN/GaN quantum well (QW) samples coated with thin films of Ag has been presented before.7–10 The results demonstrate an enhancement of the internal quantum efficiency by transfer of energy between excited electron–hole (e−h) pairs and propagating SPPs. A similar SPP-enhanced PL has been observed in the GaAs/AlGaAs QW system in which samples were coated with Au.11 Notably, SPP-enhanced emission was also observed in CdSe quantum dots (QDs) that were dispersed on evaporated Au films12 and GaN/AlN QD samples prepared using thin Ag films.13

In particular, one-dimensional III−V nanowires may play an important role in the development of electronic devices at the nanoscale as well as in the aforementioned photonic applications.14–20 Light absorption and resonant optical modes in nanowires can be tuned by altering the diameter and cross-sectional shape.21,22 Semiconductor core—shell nanowires, in which a narrower band gap core is embedded within a wider band gap material, are of significant importance for fabricating devices with enhanced optical properties or ballistic electronic transport properties.13,14 Detailed time- and polarization-resolved PL studies of GaAs/AlGaAs core—shell nanowires demonstrate the long exciton lifetimes and enhanced luminescence efficiency provided by the AlGaAs shell.24–26 The III−V nanowires are commonly nucleated and grown via a vapor–liquid–solid (VLS) mechanism using Au droplets or by self-assisted catalyst-free growth.27–29 In this work, we investigated the optical properties and carrier relaxation dynamics of Al- and Au-coated GaAs/AlAs/GaAs core−shell nanowires dispersed on Si substrates using time-resolved cathodoluminescence (CL). The nanowires were grown by self-assisted VLS growth in a high purity molecular beam epitaxy (MBE) system. The metal-coated nanowires, having diameters of ~100 nm, provide a system in which the electron−hole pairs can be created, diffuse, and recombine in varying proximity to the surface. The variable distance from the surface, at which excitons recombine, results in a range in the expected Purcell enhancement factor (FP) which describes the enhanced radiative recombination rate due to coupling of the excitons to the SPP modes of the metal film. The SPPs can be converted to free-space photons if the propagating SPPs in the
thin metal film are scattered by the surface/interface roughness or grain boundaries of the polycrystalline metal film, owing to the momentum change associated with the scattering that enables a matching with the $\omega$ vs $k$ light dispersion relation.\textsuperscript{9,10} Due to the opacity of the metal with respect to light/laser excitation from the top surface, CL is shown to be a particularly useful probe for the metal/nanowire (or more generally for metal coated nanostructure systems) in that the high-energy electron beam can easily penetrate the metal film and create excess $e^-$–$h^+$ pairs in the semiconductor nanostructure.

The GaAs/AlAs/GaAs nanowires were grown by molecular beam epitaxy (MBE)\textsuperscript{28–30} using the self-assisted VLS method on (111)-oriented silicon bearing a native oxide layer. After water removal at $\sim$200 °C, the Si wafer was outgassed overnight at high temperature ($\sim$600 °C) in a separate chamber, before being transferred into the MBE growth chamber. The growth was initiated by condensation of Ga at the point defects in the SiO$_2$ layer and carried out at $\sim$640 °C and a group V/III (As$_4$/Ga) ratio of $\sim$100. Uniform diameter GaAs nanowires were grown with a high aspect ratio, no significant tapering, and a pure zinc-blende structure, as revealed by careful transmission electron microscopy (TEM) analysis. For growth of the uniform $\sim$6 nm AlAs shell and $\sim$12 nm GaAs capping layer, the temperature was lowered to 520 °C. The GaAs capping layer protects the AlAs shell from oxidation when exposed to air.

A typical scanning electron microscopy (SEM) image of the as-grown GaAs/AlAs/GaAs core–shell nanowires can be seen in Figure 1a. Nanowires, having widths of $\sim$100 nm and lengths of $\sim$12 $\mu$m, were harvested and dispersed onto heavily doped $p$-type Si wafers coated with a $\sim$20-nm-thick layer of Al to minimize the effects of charging during the CL measurements. A characteristic hexagonal prism shape was observed for such wires, consistent with previous studies of the shape of self-assisted MBE-grown GaAs nanowires.\textsuperscript{30} Slightly tilted SEM top views of a single GaAs/AlAs/GaAs core–shell nanowire showing the tip and the base are presented in Figure 1b and c, respectively, which clearly reveal the hexagonal shape. A typical TEM image of a single GaAs/AlAs/GaAs core–shell wire taken from the same sample, showing a uniform structure with no defects, is presented in Figure 1d. The inset on the left shows an electron diffraction pattern revealing the zinc-blende structure. The inset on the right shows a high-resolution TEM image of a cross-section taken from a GaAs/AlAs/GaAs core–shell nanowire possessing the same nominal structure but having slightly different thicknesses, displaying the typical hexagonal shape.

Figure 1. SEM image of the as-grown GaAs/AlAs/GaAs core–shell nanowires taken from a side view in (a). The nanowires exhibit a uniform aspect ratio greater than $\sim$100. Slightly tilted SEM top views of a single GaAs/AlAs/GaAs core–shell nanowire showing the tip and the base in (b) and (c), respectively, which clearly reveal the hexagonal shape. A typical TEM image of a single GaAs/AlAs/GaAs core–shell wire taken from the same sample, showing a uniform structure with no defects, is presented in (d). The inset on the left shows an electron diffraction pattern revealing the zinc-blende structure. The inset on the right shows a high-resolution TEM image of a cross-section taken from a GaAs/AlAs/GaAs core–shell nanowire possessing the same nominal structure but having slightly different thicknesses, displaying the typical hexagonal shape.

observed with the aluminum deposition showing more spots and hillocks, possibly owing to differences in grain size and morphology. To better quantify the roughness of the metal films on the nanowires, we performed atomic force microscopy (AFM) measurements for the three cases, as shown in Figure 2d–f. A line scan analysis of the AFM images, with scans along a line parallel to the nanowire length near the top, is shown in Figure 2g. The root-mean-square (RMS) roughness is 0.54, 1.05, and 1.31 nm for typical scans along the bare, Au-, and Al-covered nanowires, respectively. The roughness observed for the bare nanowire is attributed to possible variations in the oxide layer thickness. A variety of feature sizes, ranging from $\sim$10 to $\sim$50 nm is observed in the AFM images and line scans for the metal coated nanowires. CL imaging and spectroscopy were previously used to determine the propagation distance of SPPs near the surface plasmon resonance of Ag and Au films, and the propagation lengths varied from a few hundred nanometers to several micrometers.\textsuperscript{31,32} Therefore, differences in the polycrystalline morphology, surface roughness, and size scale between the Al- and Au-coated wires are expected to be small in comparison to the wavelength of light and typical SPP
propagation lengths. Such roughness is not expected to influence the CL intensity measurements.

The CL experiments were performed on a modified JSM-5910 scanning electron microscope (SEM). The CL detection system has been described previously in detail and measures emission in a far-field detection geometry. Electron beam energies ($E_b$) ranging from 7 to 15 keV and e-beam currents ($I_b$) ranging from 0.3 to 2 nA were used to excite and create excess carriers in the GaAs nanowires. A GaAs:Cs photomultiplier tube operating in the 380–890 nm spectral range enabled photon counting of the luminescence which was dispersed by a monochromator with a 0.25 m focal length. The spectral resolution was set to ~1 nm. The samples were mounted in the SEM on a variable temperature and vibration isolated cold stage that was cooled with a closed-cycle cryocooler and were maintained at temperatures ranging from 50 to 300 K. Time-resolved CL experiments were performed by the method of delayed coincidence in an inverted single photon counting mode. Electron beam pulses of 50 ns width with a 1 MHz repetition rate were used to excite the sample.

Sets of CL spectra and CL decay transients of the GaAs near band-edge (NBE) emission for e-beam excitation of a single bare nanowire are shown in Figure 3 for temperatures in the 50–210 K range. The intensities of the CL spectra in Figure 3a have been normalized so that each spectrum displays approximately the same peak height, and scaling factors are shown to indicate the relative CL intensity for each temperature. As the temperature increases, the CL spectra are observed to red-shift in accordance with the temperature dependence of the GaAs bandgap, and the CL spectra broaden from 20.4 to 54.3 meV as the temperature is raised from 50 to 210 K. CL transients of the bare and Au-covered nanowires are shown in Figures 3b and c, respectively. The transients were acquired for emission wavelengths near the CL peak intensity at each temperature. The carrier lifetimes were extracted from the slopes of the CL transients at each temperature, assuming a single exponential decay whose fits are illustrated by the red lines running through the data in Figures 3b and c. The carrier lifetimes depend on the temperature and generally increase in the temperature range shown. The deposition of Au on the nanowires is found to markedly reduce the carrier lifetimes relative to the values for the bare nanowires, as shown in Figure 2.
Figures 3b and c. An analysis of the temperature dependence of the radiative lifetimes for the bare, Au-, and Al-covered wires is presented below.

To account for variations among the wires, we have performed a statistical analysis of the integrated CL intensities ($I$) and carrier decay times ($\tau$) for emission from single bare, Au-, and Al-covered wires for $T = 50$ K. The e-beam was injected into each individual nanowire through the metal film at a point above its center. The dispersal of wires on the Si substrate with random orientations has allowed for a statistical analysis of the integrated CL emission intensity for various orientation angles ($\theta$) relative to the optical axis of the CL detection system.34 By comparing coated nanowires with two different metals, whose deposition geometries are nearly identical, we can compare the effects of exciton-SPP coupling and luminescence intensity enhancements for the Au and Al metal coatings. Since the Au/GaAs surface plasmon resonance energy ($\hbar\omega_{SP} = 1.804$ eV) is much closer to the GaAs band gap energy ($\hbar\omega_{GaAs} = \sim 1.51$ eV at 50 K) than in the case of Al/GaAs ($\hbar\omega_{Al} = 2.700$ eV), as determined by the calculated $\omega$ vs $k$ dispersion relations described below, we would expect a CL intensity enhancement for the Au-covered wires relative to that for the Al-covered wires. Histograms of the integrated NBE CL emission for $T = 50$ K are shown in Figure 4a–c for the bare, Au-, and Al-covered nanowires, with each panel summarizing the mean and standard deviation of the integrated CL intensities and lifetimes. The Au-covered nanowires exhibit an average CL intensity that is $\sim 3.1$ times greater than that for the Al-covered nanowires, consistent with an enhancement in the radiative recombination rate due to exciton-SPP coupling. The absence of nanowires in the $40^\circ$–$50^\circ$ range in Figure 4a–c is purely accidental, as the nanowires were dispersed with random angles on the Si substrates for the bare, Au-, and Al-covered nanowire samples. We note that, despite the nearly identical internal reflection geometries for the Au- and Al-covered wires, the reflectivity of Al is lower than the reflectivity of Au by $\sim 12\%$ at $\lambda = 820$ nm due to an interband transition in Al,35 and as such this cannot account for the observed large differences in the integrated CL intensities. Thus, the CL intensity enhancement likely results from an enhanced exciton-SPP coupling in the Au-covered wires.

In addition, a statistical analysis of the carrier lifetimes of the GaAs nanowires was performed, and histograms of the decay times measured at $T = 50$ K for the bare, Au-, and Al-covered wires are shown in Figure 4d–f. These results show that the average lifetimes ($\tau$) for the bare, Al-covered, and Au-covered wires are $\sim 2.0$, $1.3$, and $1.1$ ns, respectively. Moreover, we have performed time-resolved CL measurements and CL integrated intensity measurements, $I(T)$, for temperatures ranging from 50 to 300 K to determine the temperature dependence of the radiative lifetime ($\tau_R$) and nonradiative lifetime ($\tau_{NR}$). We have extracted $\tau_R(T)$ from $I(T)$ using a procedure which we have previously described for time-resolved CL studies of InGaN/GaN quantum wells, presented in the Supporting Information.36 The results for $\tau(T)$ and $\tau_R(T)$ are shown in Figure 5a and b, respectively, for the bare, Al-covered, and Au-
covered wires. To assess the enhancement as a result of exciton coupling to SPPs, we obtain the energy-dependent Purcell enhancement factors as

\[ F_P(\omega) = \frac{\tau_R(\text{bare})}{\tau_R(\text{metal})} \]

which are shown in Figure 6 for the time-resolved CL measurements of the bare, Al-covered, and Au-covered GaAs/AlAs/GaAs core–shell nanowires.

To develop a reasonable model and calculate \( F_P(\omega) \) for the metal–nanowire system, we first observe that the diameter of the nanowire (~100 nm) is large compared with (i) the...
penetration depth of the SPP field into the semiconductor near resonance and (ii) the typical exciton dipole to metal film separation, both of which are \( \sim 10 \) nm for SPP-induced field enhancements in quantum heterostructures and nanostructures.\(^6\) Therefore, we expect that the largest enhancement of CL intensity and decrease in radiative lifetime will occur in regions of the nanowire where excitons recombine in closest proximity to the metal films. Assuming a small curvature of the wire for excitons recombining within \( \sim 10 \) nm of the metal, we expect that the two-dimensional (2D) SPP energy dispersion model for a metal film covering a flat dielectric material will provide a reasonable model from which the \( \omega \) vs \( k \) dispersion relations can be calculated for both Au- and Al-covered wires.\(^3\)

From the solution of Maxwell’s equations with a matching of boundary conditions at the interfaces of the vacuum/metal/GaAs system, we have calculated the \( \omega \) vs \( k \) dispersion relations which include the effects of losses for both Al and Au films possessing varying thicknesses on GaAs, as shown in Figure 7.

**Figure 7.** Calculated \( \omega \) vs \( k \) surface plasmon dispersion relations for various thicknesses (as indicated by the arrows) of Au (a) and Al (b) deposited on GaAs. The effects of losses are taken into account by including the imaginary part of the dielectric constants for Au, Al, and GaAs.

We have used the energy dependence of the dielectric constants (i.e., \( \varepsilon(\omega) = \varepsilon'(\omega) + i\varepsilon''(\omega) \)) of GaAs, Au, and Al that are found in the literature,\(^4\)\(^-\)\(^43\) and the formalism for the SPP dispersion relation calculation has been previously described.\(^7\)\(^,\)\(^8\)\(^,\)\(^44\) The Purcell factor, \( F_p \), is given by

\[
F_p(z) = 1 + \frac{\pi c^3}{4} \frac{|E(z)|^2}{\omega^2} \int_\infty^z u_k(\omega, z) dz \rho_{2D}(\omega)
\]

where \( u_k(\omega, z) \) is the electric energy density for each material region and \( E(z) \) is the un-normalized plasmon electric field at a distance \( z \) from the metal/GaAs interface.\(^7\)\(^,\)\(^8\) The 2D density of plasmon states per unit area can be calculated from the dispersion relations in Figure 7 and is given by \( \rho_{2D}(\omega) = (1/4\pi^2)\partial^2 / \partial \omega^2. \) For a dielectric in the transparency region (i.e., \( \varepsilon''(\omega) \ll \varepsilon'(\omega) \)), the energy density of the electric field in the layer is given by \( u_k(\omega, z) = [\partial(\omega\varepsilon''/\partial\omega)]/|E(z)|^2/8\pi^2. \)\(^3\)

However, for metals with dissipation, the expression for the energy density is \( u_k(\omega, z) = (\varepsilon'(\omega) + 2\omega\varepsilon''/\gamma)|E(z)|^2/8\pi^2 \) where \( \gamma \) is the damping constant which can be estimated from the Drude model.\(^46\)\(^,\)\(^47\) The propagation of electromagnetic waves along the metal/GaAs interface yields analytical solutions for the in-plane electric field, \( E_{\text{in}} \), in the \( i \)th layer (such that \( i = 1 \) to 3 represent the vacuum, metal, and GaAs regions, respectively) of the form \( E_{\text{in}}(\omega) = \exp(ikz)\exp(-ic\varepsilon_0^{1/2}/\gamma) \) with \( k_0^2 = k_0^2 - (\omega^2/c^2) \). The denominator of eq 1 can easily be integrated to yield an analytical solution for \( F_p(z) \) that is given by

\[
F_p(z) = 1 + \frac{\pi c^3}{4} \left[ \frac{e^{-2k_0z^2}}{k_0^2} + \frac{e^{-2k_0z^2}}{k_0^2} \left[ 1 - e^{-2k_0z^2} \right] + \frac{\partial(\omega\varepsilon''/\partial\omega)}{\partial\omega} \right]
\]

Using the appropriate dielectric functions of Au, Al, and GaAs,\(^40\)\(^-\)\(^43\) we have plotted \( F_p(z) \) for metal thicknesses \( t \) of 10, 20, and 30 nm and for \( \hbar\omega = 1.42 \) eV \( (T_{\text{GaAs}} \approx 300 \text{ K}) \), as shown in Figure 8a. As expected from the energy differences between the GaAs NBE emission at room temperature \( (\sim 1.42 \text{ eV}) \) and the surface plasmon frequencies for Au \( (\hbar\omega_{\text{Au}} \approx 1.804 \text{ eV}) \) and for Al \( (\hbar\omega_{\text{Al}} \approx 2.700 \text{ eV}) \), the values of \( F_p(z) \) in close proximity to the metal (i.e., for \( z < \sim 10 \text{ nm} \)) for Al/GaAs are at least an order of magnitude larger than those for Al/GaAs.

To develop a model for the average Purcell enhancement factor, \( \langle F_p \rangle \), for exciton recombination in the metal-covered wires subject to the present e-beam excitation conditions, we expect that a relatively uniform e–h pair excitation density occurs across the cross-section of the wire \( (\sim 100 \text{ nm}) \) owing to (i) the size of the e–h generation volume for a beam energy of 15 keV and (ii) the minority carrier diffusion length, both of which are \( \sim 1 \mu\text{m} \). Therefore, in the simplest model, we calculate the effective Purcell enhancement factor \( \langle F_p \rangle \) as an area average over the 2D cross-section of the wire (neglecting recombination in the thin AlAs shell). We considered nanowire cross-sectional shapes of hexagonal, circular, and square, and for comparison, as illustrated in the inset of Figure 8a for the first two shapes. The dimensions of the nanowire used in the calculations are given by \( r_0 = 50 \text{ nm}, r_1 = 32 \text{ nm}, \) and \( r_2 = 38 \text{ nm} \) for a 100 nm diameter GaAs/AlAs/GaAs core–shell nanowire possessing a \( \sim 12\)-nm-thick GaAs capping layer surrounding a \( \sim 6\)-nm-thick AlAs shell. In this model, we assume that half of the wire is covered by a thin metal film of variable thickness \( t \), whereas the other half is bare due to shadowing during the metal deposition. The local thickness of the metal on the nanowire surface is taken as \( t = t_0 \cos \alpha \), where \( \alpha \) is the angle between the impinging metal beam and surface normal during the metal deposition. The results of the averaging for the case of hexagonal nanowires are shown in Figure 8b in which \( \langle F_p \rangle \) is plotted vs \( \hbar\omega \) for Au and Al covered wires for film thicknesses \( t_0 \) ranging from 10 to 30 nm. The energy range of the GaAs NBE emission for temperatures between 50 and 300 K is shown on the graph for comparison. As observed in Figure 8b, the expected average Purcell factor...
for $\hbar \omega = 1.42$ eV is reduced in comparison to the maximum values near the metal/GaAs interface ($z \approx 0$) shown in Figure 8a. This is due to the assumption of a roughly uniform density for $e^-h$ pair generation and recombination over the cross-sectional area of the wire.

Moreover, in Figure 9a, we show the calculated values of $\langle F_P \rangle$ for the energy range associated with the NBE emission in GaAs from 50 to 300 K. The calculations of $\langle F_P \rangle$ for a metal thickness of $t_0 = 20$ nm are shown for nanowires possessing hexagonal, circular, and square cross sections, for comparison. For the circular cross-section, two cases were considered in which the metal film thickness was both constant ($t = t_0$) and allowed to vary as $t = t_0 \cos \alpha$ (i.e., a tapered film coverage, as illustrated in the inset of Figure 8a). The calculations for the hexagonal and circular (tapered) cross sections yield the largest values of $\langle F_P \rangle$ in Figure 9a as a result of local enhancements in $F_P$ caused by the proximity of excitons to segments of the Au films having reduced thicknesses ($t \ll 20$ nm). While the calculations agree qualitatively with the experimental results in that the values of $\langle F_P \rangle$ for the Au-covered nanowires are larger than those for the Al-covered nanowires at all energies, it is clear that the model...
predicts an $\langle F_0 \rangle$ which is considerably larger (by a factor of 5–10) than the experiment.

To account for this discrepancy, we first note that an oxide layer of 2–3 nm in thickness surrounds the outer GaAs capping layer of the nanowire. Therefore, we observe that when Au or Al is deposited on the nanowires, the actual minimum separation between the metal layer and the excitons in the nanowires is at least equal to the thickness of the oxide layer. However, additional effects caused by the absence of a complete surface passivation may influence the average minimum separation distance between the metal film and the excitons. The presence of the oxide layer may induce surface/interface states in the gap, create a depletion layer, and lead to small band-bending effects near the oxide/GaAs interface. Band-bending near the surface of semiconductors has the well-known effect of creating a “dead-layer” near the surface in which the density of photoexcited or e-beam excited electron–hole pairs is significantly reduced. Such effects have recently been observed in GaAs and ZnO nanowires.46,49 Unfortunately, without sufficient information regarding the Fermi-level pinning near the oxide/GaAs surface the effective width of such a “dead-layer” cannot be reliably estimated. Instead, we have included an additional step in our model for the calculation of $\langle F_0 \rangle$ in which the minimum effective metal–exciton separation ($t_{\text{eff}}$) is allowed to vary from 0 to 10 nm, as shown in Figure 9b for an energy of $\hbar \omega = 1.42$ eV (i.e., close to the room-temperature band gap of GaAs). In this modified model, $t_{\text{eff}}$ describes the minimum effective metal–exciton separation, due to a combination of the oxide layer thickness and band-bending effects near the oxide/GaAs interface, before the plasmon field begins to act on the excitons. The calculations in Figure 9b show an exponential decay in $\langle F_0 \rangle$ as a function of $t_{\text{eff}}$ for the four cross-sectional wire geometries, which we believe is a plausible explanation for the reduced values of the experimental $\langle F_0 \rangle$ relative to the values for the ideal case shown in Figure 9a in which $t_{\text{eff}} = 0$. Consequently, the calculated values in our simple model approach the maximum experimental values of $\langle F_0 \rangle \approx 3$ for $t_{\text{eff}} = 10$ nm. Thus, the assumption of an effective metal–exciton separation of $\sim 10$ nm appears to minimize the discrepancy between experiment and theory in these results. The calculations are also observed to be strongly dependent on the nanowire diameter, cross-sectional shape, and on local film thickness variations which depend on the metal evaporation geometry.

In conclusion, the coupling of excitons to SPPs in Au- and Al-coated GaAs/AlAs/GaAs core–shell nanowires was probed using time-resolved CL. Excitons were generated in the metal-coated nanowires possessing nearly identical geometries by injecting a pulsed high-energy electron beam through the thin metal films. The average Purcell enhancement factor, $\langle F_0 \rangle$, was obtained by the direct measurement of the changes in the integrated NBE CL emission intensities and the temperature-dependent radiative lifetimes. To analyze the coupling of excitons to SPPs, a model was presented for $\langle F_0 \rangle$ which takes into account the dependence of $F_0$ on the distance from the metal film and the thickness of the film covering the GaAs nanowires possessing various cross-sectional shapes. The present study suggests a more general approach for treating metal-covered nanostructures in which exciton diffusion in the mesoscopic regime can influence the observed plasmonic effects.

**NOTE ADDED AFTER ASAP PUBLICATION**

Equations 1 and 2 have been updated. The revised version was re-posted on April 2, 2013.