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The unique properties of surface plasmon polaritons, such as strong field confinement and local field enhancement effects, make them ideal candidates to enhance and shape the emission of luminescent nanoparticles. Of these nanoparticles, quantum dots are highly versatile, suitable for vastly different applications due to their size and material tunability. In many cases however, the emission wavelength of the quantum dots is fixed after manufacturing, allowing no control over the in situ emission properties. Here, we show fully optical, in situ tunability of the emission wavelength of quantum dots, with shifts of over 30 nm, employing surface plasmon polaritons to control the emission wavelength. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4724327]

The combination of surface plasmon polaritons (SPPs) with emitters, such as dye molecules1–5 rare-earth ions,6,7 and quantum dots,8 is a heavily studied field of optics. The unique properties of SPPs, such as a strong field confinement, a resonant behavior, and the presence of local field enhancement,9,10 explain the broad interest. Of all types of emitters, quantum dots have been shown to be versatile luminescent nanoparticles, suitable for various applications in plasmonics, such as loss compensation and gain,11–14 coupling to plasmonic antennas,15 and imaging of near fields of plasmonic waveguides and bends.16 Moreover, broader applications like in vivo imaging of cells and animals,17,18 single emitter studies,15,19,20 and fundamental research of quantum effects8,20,21 are also possible with quantum dots. The versatility of quantum dots stems from the fact that their absorption and emission properties can be selected by varying the material and size22,23 of the individual particles, allowing the particles to be designed to a particular application. However, in many cases, the emission wavelength of the quantum dots is fixed after fabrication, and one has no control over the in situ emission properties. Here, we show fully optical, in situ tunability of the wavelength of the emission of quantum dots with SPPs as the excitation and wavelength control. The shift in wavelength we obtain with our method is over 30 nm and therefore can be employed with frequency-selective devices to allow, e.g., switchable selective emission redirection.24

Our approach employs basically a classic Kretschmann configuration,9 with either a 532 nm frequency-doubled continuous wave (CW) neodymium yttrium aluminium garnet (Nd:YAG) laser for excitation and wavelength control, or a combination of a CW 405 nm diode laser for excitation and a 740–850 nm tunable titanium-sapphire (Ti:Sapph) laser for emission wavelength control. See Figure 1 for a schematic depiction of the setup. Three solutions of quantum dots in toluene were made, each with a different concentration. Subsequently, quantum dots were spin coated onto glass substrates containing a 50 nm thin silver film. The quantum-dot concentration was 20 μM, 40 μM, and 60 μM, respectively, and the samples in this study are referred to with these concentrations.25

The SPP reflectance minima for p-polarized light with a wavelength of 532 nm were determined for samples with silver and quantum dots applied, as a function of angle. See Figure 2. The minimum in reflectance for the samples with 20 μM, 40 μM, and 60 μM quantum-dot concentrations occurred at 45.6° ± 0.2°, 49.2° ± 0.2°, and 55.6° ± 0.2°, respectively. The plasmonic nature of the interaction of p-polarized light with the sample is clearly visible in Figure 2. In accordance with theory,9 for samples with a layer of quantum dots obtained from a more concentrated solution, the plasmon resonance shifts to higher angles due to the effective refractive index of the quantum dots that form the layer, which is greater than that of air.8 The inset in Figure 2 shows the reflectance curve of the 60 μM sample for s-polarized light, which indeed does not show any sign of the existence of SPPs: The reflectance is larger than 94% ± 3% for all angles of incidence. A similar behavior was observed for the

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We pose that the tunability in our approach stems from the fact that the band gap of the semiconductor quantum dots depends on the local temperature. The temperature dependence of the band gap of semiconductors and semiconductor quantum dots can be approximated by

\[ E_g(T) = E_{g0} - \alpha \left( \frac{T^2}{T + \beta} \right), \]

where \( E_g \) is the temperature-dependent band gap, \( E_{g0} \) is the band gap at 0 K, \( T \) is the temperature in Kelvin, and \( \alpha \) and \( \beta \) are experimentally determined parameters.

The absorbed power by each sample, irradiated upon by the 532 nm laser, is calculated by multiplying the incident power by 1–\( R \), with \( R \) the reflection coefficient as shown in Figure 2. It is assumed that the temperature of the sample and the absorbed power are linearly dependent on each other, and the temperature rise due to the absorbed power is calculated under the assumptions that the prism is at room temperature (293 K) and that heat transfer by convection is
FIG. 5. Shift of the emission peak (in eV) of the quantum dot samples as a function of the power of the surface plasmon, both for SPP caused (a) and independent (b) excitation of the quantum dots. (a) Shift of the emission peak of the 20 μM, 40 μM, and 60 μM quantum dot samples as a function of the power of a 532 nm laser, inducing SPPs in the sample while simultaneously exciting the quantum dots in the thin film on top of the silver. With increasing power of the induced SPP, the emission of the quantum dots shifts towards the red. For the 20 μM quantum dot sample, a shift in band gap of 34.3 nm ± 0.9 nm was observed, or, in electronvolts, a shift of nearly 100 meV. (b) Shift of the emission peak of the 60 μM and 40 μM quantum dot samples as a function of the power of a tunable Ti:Sapph laser, inducing SPPs in the sample. The excitation power of the 405 nm laser is kept constant at 1 mW. This shows that the shift can be controlled independently from the quantum dot excitation power. Data for two samples are shown, each at two wavelengths of the SPP-inducing laser. These 40 μM and 60 μM samples are the same samples as in (a). Lines are guides for the eye.

FIG. 4. The quantum dot emission wavelength shifts as a function of the laser power inducing the SPP, both for SPP caused (a) and independent (b) excitation of the quantum dots. (a) Shift of the emission peak of the 20 μM, 40 μM, and 60 μM quantum dot samples as a function of the power of a 532 nm laser, inducing SPPs in the sample while simultaneously exciting the quantum dots in the thin film on top of the silver. With increasing power of the induced SPP, the emission of the quantum dots shifts towards the red. For the 20 μM quantum dot sample, a shift in band gap of 34.3 nm ± 0.9 nm was observed, or, in electronvolts, a shift of nearly 100 meV. (b) Shift of the emission peak of the 60 μM and 40 μM quantum dot samples as a function of the power of a tunable Ti:Sapph laser, inducing SPPs in the sample. The excitation power of the 405 nm laser is kept constant at 1 mW. This shows that the shift can be controlled independently from the quantum dot excitation power. Data for two samples are shown, each at two wavelengths of the SPP-inducing laser. These 40 μM and 60 μM samples are the same samples as in (a). Lines are guides for the eye.

negligible. The change in the emission peak wavelength (in eV) versus the increase in temperature of the sample, both compared to the case of the lowest excitation power (200 mW), is shown in Figure 5, which also contains plots of Eq. (1) with the constants taken from literature.27–29 The plots show the minimum and maximum change possible given the constants from literature and their respective error bounds. At temperatures close to the boiling temperature of octadecylamine, the surface group of the quantum dots, the measured data deviate from the theoretical curves. However, for lower temperatures, the overlap of the quantum dot emission wavelength with the theoretical curves is excellent. The likely range of temperature increase, above room temperature, makes this scheme highly attractive for plasmolectric devices.

In conclusion, we have shown surface plasmon polariton-controlled emission wavelength control of quantum dots, spin-coated onto a thin silver film. By varying the power of the surface plasmon, we repeatedly and reversibly have shifted the wavelength of the emission of the quantum dots towards longer wavelengths, with observed shifts of up to 34 nm. The control of the emission wavelength is independent from the excitation of the quantum dots, as is shown by employing two separate lasers: one for the excitation of the quantum dots and one to induce surface plasmon polaritons in the silver film. On the other hand, we have shown that the effect can be realized also with one laser only, providing simultaneously the quantum dot excitation and the emission wavelength control. We pose that the control of the emission wavelength is due to dissipation of power, in the form of surface plasmon polaritons, causing local heating. The rise in temperature consequently shifts the band gap of the quantum dots to lower energetic values. With a model for the heating of the sample due to surface plasmon dissipation, the local temperature elevation is calculated. The measured shift in emission peak wavelength versus temperature shows a considerable overlap with theoretical curves, obtained from literature. Our results demonstrate the power...
of SPPs in the control of emitters and introduce an accurate, highly controllable method for \textit{in situ} tuning of the quantum dot emission wavelength.

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