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Coupling of single InGaAs quantum dots to the plasmon resonance of a metal nanocrystal

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The authors report the coupling of single InGaAs quantum dots (QDs) to the surface plasmon resonance of a metal nanocrystal. Clear enhancement of the photoluminescence (PL) in the spectral region of the surface plasmon resonance is observed which splits up into distinct emission lines from single QDs in micro-PL. The hybrid metal-semiconductor structure is grown by molecular beam epitaxy on GaAs (100) utilizing the concept of self-organized anisotropic strain engineering for realizing ordered arrays with nanometer-scale precise positioning of the metal nanocrystals with respect to the QDs. © *2010 American Institute of Physics*. [doi:10.1063/1.3467853]

Coupling of a single quantum emitter to an electromagnetic resonator is of fundamental importance to control its optical properties such as radiative lifetime, absorption cross-section, and nonlinear susceptibilities. It is most widely achieved by placing the emitter in a dielectric cavity.[1–3] Recently there is an increasing interest in using metal nanostructures as electromagnetic resonators to confine light well below its wavelength through the excitation of surface plasmons.[4–7] Quantum dots (QDs) are an important class of quantum emitters in solid state enabling scalable quantum functional devices and circuits with ultimate performance at the single or few electron and photon levels.[8,9] However, placing a QD in the vicinity of a metal nanostructure requires nanometer-scale precise control over the mutual position and separation. For too small separation the QD emission is quenched due to nonradiative energy transfer while for too large separation the plasmonic effects will be lost.[7] In the case of colloidal QDs chemical self-assembly based on long organic molecules that bind to the functionalized surface of metal nanoparticles is often used.[10] For epitaxial systems we have recently developed a very promising approach, which is based on strain correlated growth.[11] Ordered QD structures are first fabricated by self-organized anisotropic strain engineering of strained superlattice (SL) templates and local strain recognition.[12] The same strain recognition mechanism is then applied for the alignment of metal nanocrystals on top of the QD arrays. The required precise position control is hence achieved laterally due to the QD ordering and vertically by thin separation layers between the QDs and metal nanocrystals and enhanced QD emission in ensemble measurements has already been observed.

Here we report enhanced emission from single InGaAs QDs due to coupling to the surface plasmon resonance (SPR) of an In nanocrystal. Micro-photoluminescence (micro-PL) measurements at low temperatures reveal intense sharp lines from single QDs in the spectral region of the SPR. Without In nanocrystals there is no emission enhancement and apart from the spectral region of the SPR no emission from individual QDs is resolved due to the density of the QDs being much larger than that of the In nanocrystals. Realization of plasmon enhanced emission from single QDs is the basis for exploring and utilizing the quantum nature of light at deep subwavelength nanometer length scales.

The samples were grown by solid source molecular beam epitaxy (MBE) on undoped, singular GaAs (100) substrates. First, a 200 nm thick GaAs buffer layer was grown at 580 °C. Next, a 15 period InGaAs/GaAs SL template was deposited to obtain one-dimensional QD ordering on top.[12] Each SL period consisted of a 2.3 nm $In_{0.4}Ga_{0.6}As$ QD layer grown at 540 °C immediately capped with 0.7 nm GaAs, annealing for 2 min at 580 °C, and a 12 nm GaAs layer grown at 580 °C. On the SL template a single layer of 2.3 nm $In_{0.4}Ga_{0.6}As$ QDs was deposited, that was capped with 3 nm GaAs. Afterwards the samples were cooled down to 120 °C. During cooling down the As valve was closed. After considerable growth interruption until the pressure was below 2×10^{-9} Torr, In nanocrystals with 4 monolayers In amount were deposited.[13] For reference samples without In nanocrystals and with varying GaAs cap layer thicknesses were grown. The growth rates of GaAs and InAs were 0.054 and 0.0375 nm/s. The morphology of the samples was characterized by a tapping-mode atomic force microscope (AFM) under ambient conditions. Low temperature micro-PL measurements of the InGaAs QD arrays were performed with the samples placed in a He-flow cryostat. The 632.8 nm line of a helium–neon laser served as excitation source. The PL was dispersed by a single 1/4-m monochromator and detected by a liquid nitrogen cooled InGaAs photodiode array. The spatial resolution was around 2 μm. The SPR of the In nanocrystals was measured by differential reflectivity spectroscopy[14] at room temperature.

A schematic of the structure is shown in Fig. 1 including the SL template, InGaAs QD arrays, and In nanocrystals. Due to multiple stacking and annealing in SL template formation, wirelike InGaAs structures form due to the anisotropic properties of the GaAs (100) surface and strain-driven migration.[12] The InGaAs QDs order in linear arrays on top of the SL template due to local strain recognition. The In nanocrystals align on top of the InGaAs QDs. This alignment is again strain driven as it is not affected by the GaAs layer on top of the QDs smoothing the surface.[11] Morphology related ordering would be lost after capping, which is not the case as seen in the AFM image in Fig. 2.

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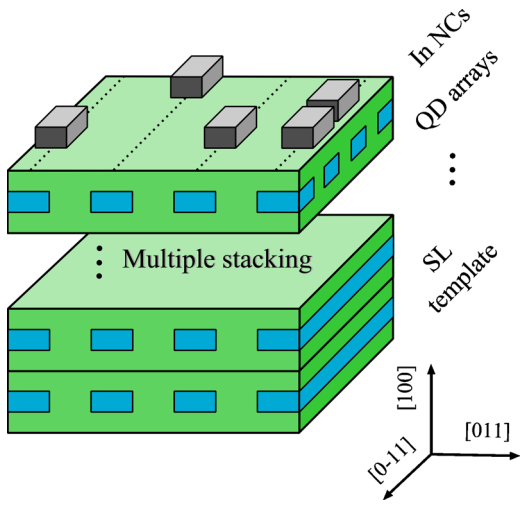

FIG. 1. (Color online) Schematic of the sample structure.

The growth conditions of the In nanocrystals are chosen to obtain a relatively low density of ~5 μm^{-2} (average base size and height are 40 and 30 nm). This is advantageous to resolve plasmon enhanced emission from single QDs in micro-PL as the SPR-related local field enhancement is extremely short-ranged.[7] Especially in high-index materials such as GaAs it extends only a few nanometers. In this respect the one-dimensional QD ordering together with the vertical alignment of the In nanocrystals is essential. It guarantees that there is always a QD at the optimal distance from the In nanocrystal. The micro-PL spectra taken at 150 K at different positions of the sample are shown in Fig. 3. The inset shows ensemble PL together with a reference spectrum of a sample without In nanocrystals. The peak QD PL intensity at 1240 nm of the reference sample is typically a factor 2 higher. Most important, there is a clear enhancement of the PL emission around 1000 nm which coincides with the spectral region of the SPR. This enhanced emission is split up into distinct lines in micro-PL, indicating the emission from single QDs.

The measurement temperature of 150 K is chosen as it is the temperature where the most distinct PL enhancement is observed. At lower temperature the PL of the SL template dominates while at higher temperature the enhancement becomes less pronounced, though visible up to room temperature. No enhanced PL emission is observed for the reference

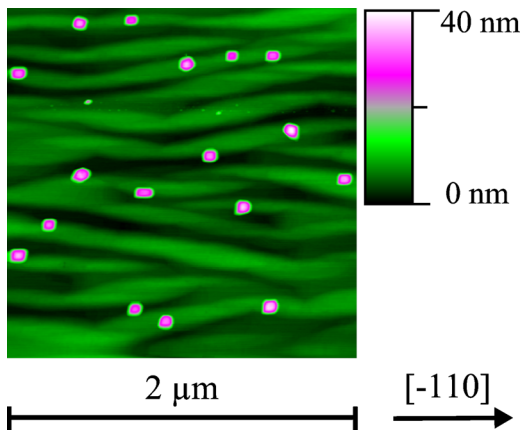

FIG. 2. (Color online) 2×2 μm^2 AFM image of the investigated structure.

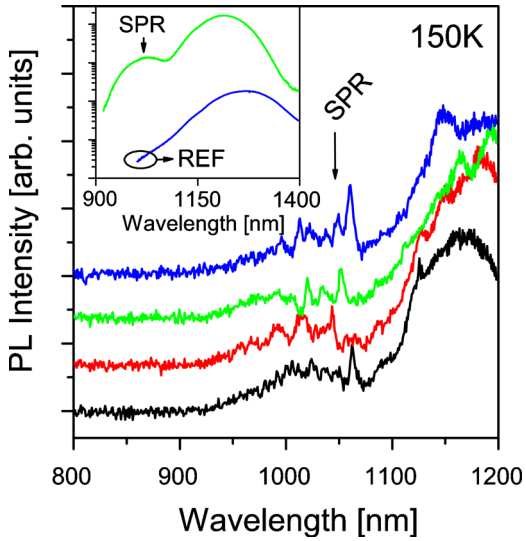

FIG. 3. (Color online) Micro-PL spectra taken at 150 K at different positions of the sample. Inset shows ensemble-PL of the investigated and reference (without In nanocrystals) samples. The curves are vertically offset for clarity.

sample without In nanocrystals. Moreover, enhanced PL emission is not observed for similar structures with increased GaAs cap layer thickness and when the SPR of the In nanocrystals is detuned with respect to the QD PL by changing the size of the In nanocrystals (depending on In amount and growth temperature). The linewidth of the emission from the single QDs is 3–5 nm. This relatively broad linewidth is typical for single surface QDs or QDs close to processed interfaces due to surface states or defects creating a fluctuating charge environment affecting the QD energy levels.[15] Most remarkable, the splitting up of the micro-PL is only observed in the spectral region of the SPR. This additionally confirms that it is due to enhanced emission only from those single QDs that are coupled to the SPR of the In nanocrystals which intentionally have a low density. Otherwise the QD density is too high that emission from individual QDs can be resolved for the present spatial resolution. As mentioned, the one-dimensional QD ordering is beneficial for the observation of SPR-QD coupling because it provides a continuous variation in the QD-metal separation, as presented in Fig. 4. Nevertheless, the number of lines observed in micro-PL is much smaller than the number of metal nanocrystals in the laser spot, because only very few QDs are both spatially and spectrally matched for optimum enhancement.

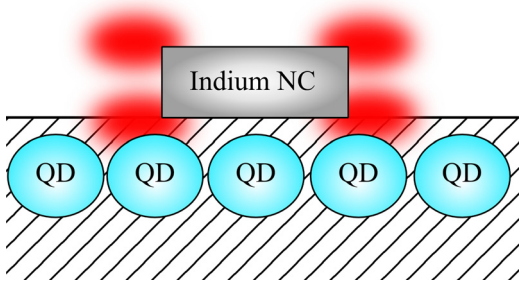

FIG. 4. (Color online) Schematic cross-section of the InGaAs QD arrays and In nanocrystals together with the plasmon field, indicating the importance of the one-dimensional ordering for optimum SPR-QD coupling.

We have reported the coupling of single InGaAs QDs to the SPR of an In nanocrystal. The hybrid metal-semiconductor structure was grown by MBE utilizing the concept of self-organized anisotropic strain engineering for realizing ordered arrays with nanometer-scale precise lateral and vertical positioning of the In nanocrystals with respect to the QDs. Clear enhancement of the emission in the spectral region of the SPR was observed. In low-temperature micro-PL this emission splits up into distinct lines from single QDs opening the door for the exploitation and utilization of the quantum nature of light at deep subwavelength nanometer length scales.

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