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Epitaxial metal nanocrystal-semiconductor quantum dot plasmonic nanostructures

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1. Introduction

Without any doubt the most fascinating application of plasmonics is in active structures like nanolasers [1,2], optical transistors [3,4] and photonic switches [5], where a metal nanostructure is combined with a single quantum emitter, for example a semiconductor quantum dot (QD). Such hybrid nanostructures can easily be scaled to sub-wavelength dimensions. However, fabrication of such devices requires single-nanometer precise control of the separation of the metal nanostructure and the emitter, due to extremely high confinement of the electromagnetic field of the surface plasmon resonance (SPR) supported by the metal nanostructures. Our approach to solve this problem is based on the alignment of epitaxial metal nanocrystals on near surface InAs QDs all grown by molecular beam epitaxy (MBE). It has been demonstrated for In [6], as well as Ag nanocrystals [7], the latter metal being the material of choice for plasmonic applications, as it offers lowest resistive losses. What is more, this approach, being based on standard epitaxial growth technology, offers the possibility of integration of active plasmonic structures with existing photonic and electronic semiconductor devices.

Here we report the self-alignment of epitaxial Ag nanocrystals on epitaxial near surface InAs QDs grown on GaAs (100) substrates by MBE. The SPR wavelengths of the Ag nanocrystals can be tuned over a wide range, covering the emission wavelength of the QDs. Low temperature Photoluminescence (PL) measurements reveal enhanced intensity of the emission, with lines from individual QDs being resolved.

2. Experimental procedure

All samples were grown in a solid-source MBE system with an attached second deposition chamber for Ag evaporation. The process to obtain aligned metal-QD hybrids is shown schematically in Fig 1. After native oxide desorption a 200 nm GaAs buffer layer was grown at 580 °C. InAs QDs were deposited at 500 °C and capped with a 3-5 nm thin GaAs layer. The deposition of Ag was done at 250-300 °C with the coverage of 0.5-2 nm. The sample morphology was characterized by tapping-mode atomic force microscopy (AFM) and the SPR wavelength was measured using differential reflectivity (DR). PL measurements were performed at 10K in a micro-PL setup with excitation wavelength of 635 nm.

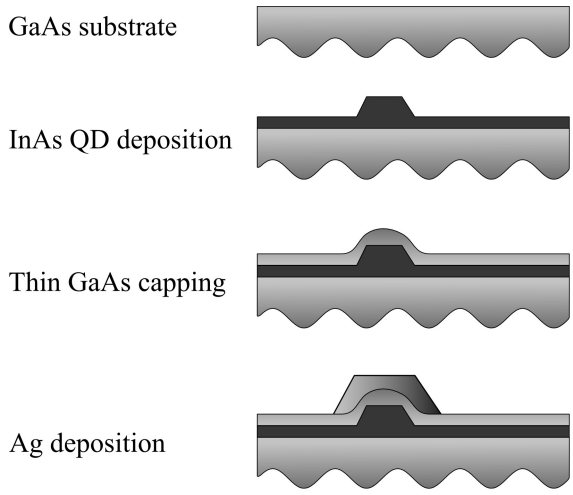

Fig. 1. Deposition steps to obtain Ag nanocrystal/InAs QD self-aligned structures.

3. Results and discussion

Deposition of Ag on a clean GaAs surface results in formation of epitaxial metal nanocrystals with a hut shape and clear facets, as shown in Fig 2a. Reflection high-energy electron diffraction measurements (RHEED) reveal that the

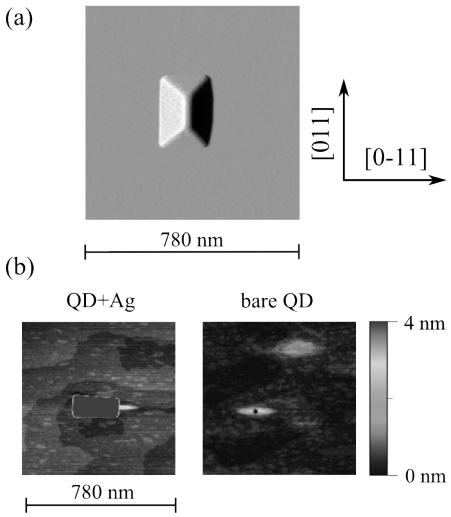

Fig. 2. (a) AFM image of a single Ag nanocrystal revealing clear faceting. (b) Single metal-QD hybrid structure (left) and a reference image of a single near-surface QD (right).

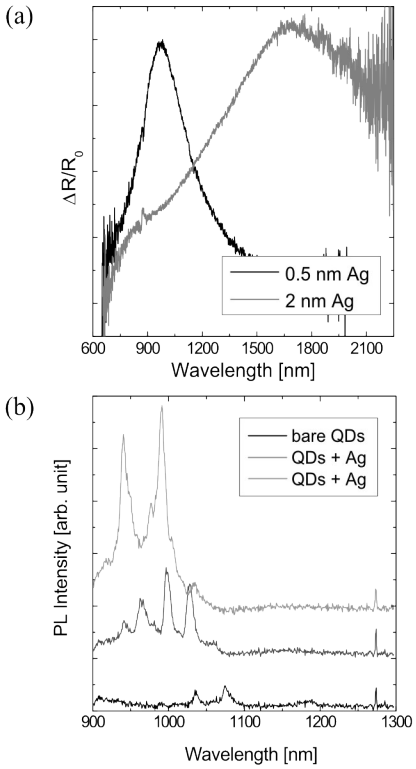

Fig. 3. (a) DR spectra of samples with different Ag coverage. (b) Micro-PL spectra of a sample with metal-QD hybrids and of a reference sample with only bare QDs. The two measurements of the sample with Ag are taken at different positions.

nanocrystals have a well-defined orientation with respect to the substrate. Careful analysis of the AFM data reveals, that the Ag nanocrystals nucleate on top of near-surface QDs, as presented in Fig 2b. Interestingly, the nucleation on presence of the near surface QDs changes the preferential elongation direction of the metal nanocrystals from [011] to [0-11], what suggests the possibility of template based shape control. By changing the average nanocrystal size upon variation of coverage and deposition temperature, the SPR wavelength, shown in Fig 3a, can be tuned over a wide spectral range covering the important 1.3 μm and 1.5 μm telecom bands.

The micro-PL measurements show clear enhancement of the emission from individual QDs (Fig. 3c). This is not the case for a reference sample without metal nanocrystals. The relatively large linewidths, as compared with deeply buried QDs, are due to the close proximity of the semiconductor surface. What is more, it is also possible to observe an enhancement of the bulk GaAs emission, which is attributed to SPR-enhanced absorption of the exciting light.

4. Conclusions

We demonstrated epitaxial growth of Ag nanocrystals on GaAs by MBE. By changing deposition parameters, such as coverage and temperature, their size and, thus, the SPR resonance was tuned over a wide range covering the important telecom wavelength bands. When the Ag nanocrystals were deposited on near-surface InAs QDs they self-aligned on top of the QDs. This showed a way to synthesize hybrid metal-semiconductor nanostructures with control of the lateral, owing to the self-alignment, and vertical, due to the capping layer thickness, QD to metal separation. PL measurements revealed clear intensity enhancement resolved down to the individual QD level. We believe, that the superior control of size, density, and position demonstrated here for epitaxial metal nanocrystals is highly relevant to exploit plasmonic effects in future solid-state devices for applications in sensing, spectroscopy, imaging, solar energy conversion, light emitting diodes, nanophotonic circuits, and many others.

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