

Self-aligned epitaxial metal-semiconductor hybrid nanostructures for plasmonics

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Self-aligned epitaxial metal-semiconductor hybrid nanostructures for plasmonics

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We demonstrate self-alignment of epitaxial Ag nanocrystals on top of low-density near-surface InAs quantum dots (QDs) grown by molecular beam epitaxy. The Ag nanocrystals support a surface plasmon resonance that can be tuned to the emission wavelength of the QDs. Photoluminescence measurements of such hybrid metal-semiconductor nanostructures reveal large enhancement of the emission intensity. Our concept of epitaxial self-alignment enables the integration of plasmonic functionality with electronic and photonic semiconductor devices operating down to the single QD level. © 2011 American Institute of Physics. [doi:10.1063/1.3596460]

Nanometer-scale precise alignment of hybrid nanostructures is a fundamental challenge in modern nanotechnology. One of the most widely targeted applications is in plasmonics when coupling a semiconductor quantum dot (QD) to a metal nanostructure. Metal nanostructures support electromagnetic modes called localized surface plasmon resonances (SPRs) that enable confinement of light at deep subwavelength length scales and induce huge local field enhancements.¹ Coupling of QDs to such localized modes allows engineering of their optical properties. Those hybrid nanostructures find applications in novel optical devices including nanolasers or spasers^{2,3} and optical transistors.⁴ All above mentioned functionalities rely on near-field coupling, so fabrication requires control of the metal-emitter distance within a nanometer, which has so far been demonstrated employing colloidal QDs.⁵⁻⁷

We achieve this control by the self-alignment of epitaxial Ag nanocrystals on near-surface InAs/GaAs QDs grown by molecular beam epitaxy (MBE). This is the most significant advance compared to the previously demonstrated alignment of In nanocrystals⁸ as Ag is the material of choice for plasmonics exhibiting the lowest resistive losses of all metals. Moreover, it reveals the generality of our concept of epitaxial self-alignment. In fact, epitaxial self-alignment is well known for the correlated stacking of QDs due to strain mediation⁹⁻¹² but has so far not been applied to hybrid systems. We precisely match the density of the Ag nanocrystals with that of the QDs. We also demonstrate tuning of the Ag nanocrystal SPR by changing the nanocrystal size and accurately control the metal-QD distance by the thickness of the GaAs cap layer on the QDs. Finally we demonstrate strong enhancement of the emitted light intensity of the QDs.

All samples were grown by solid-source MBE on singular (100) oriented, undoped GaAs substrates. After oxide removal under As₄ flux at 580 °C, a 200 nm GaAs buffer layer was grown. Ag nanocrystals were grown on both bare GaAs and capped InAs QDs. InAs QDs were grown following the

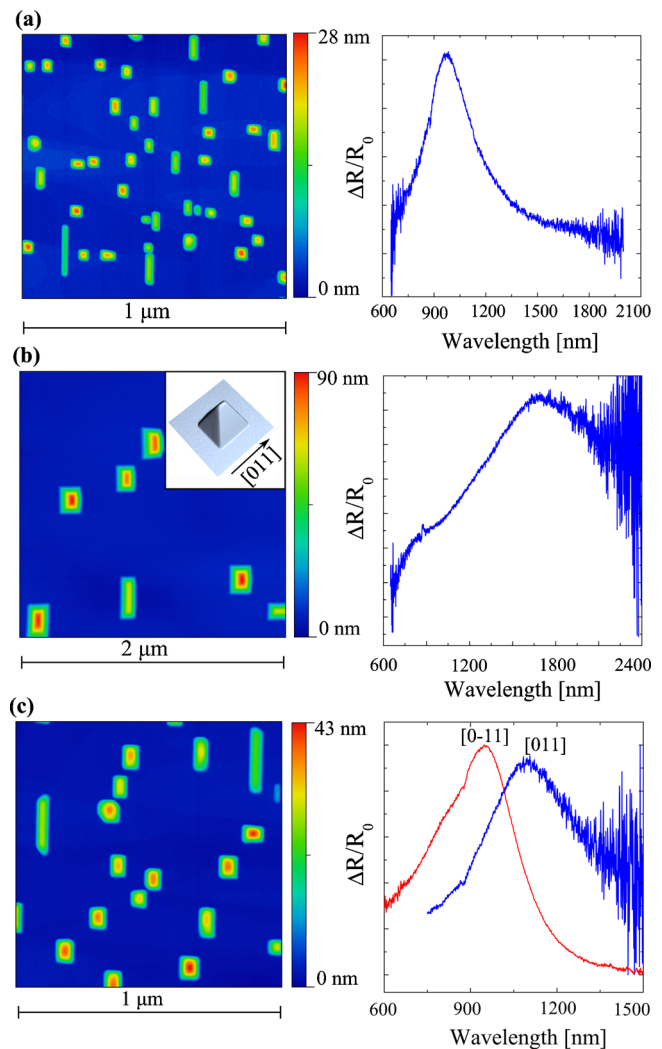


FIG. 1. (Color online) (a) AFM image and DR spectrum of 0.5 nm Ag nanocrystals deposited at 250 °C. (b) AFM image and DR spectrum of 2 nm Ag nanocrystals deposited at 300 °C. The Inset shows a three-dimensional AFM image of a single Ag nanocrystal at enlarged scale. The nanocrystal elongation is along [011]. (c) AFM image and DR spectra of 1 nm Ag nanocrystals deposited at 275 °C. The DR spectra are shown for light polarized parallel and perpendicular to the long axis of the Ag nanocrystals, i.e., along [011] and $[0\bar{1}1]$. The large difference of the signal-to-noise ratio is due to the polarization characteristics of the diffraction grating.

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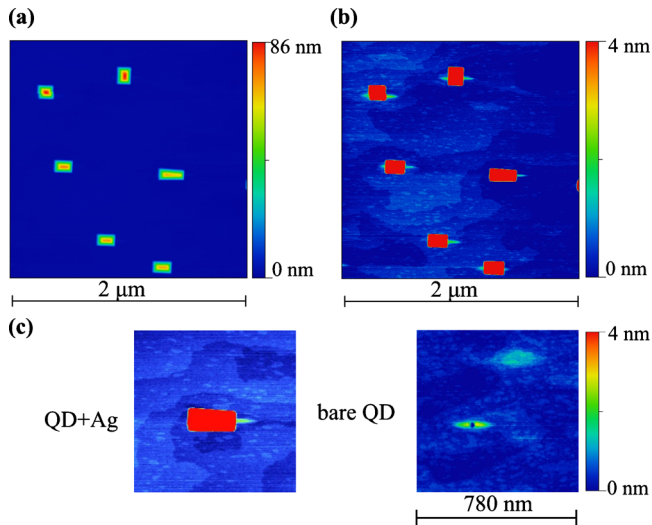


FIG. 2. (Color online) (a) AFM image of Ag nanocrystals deposited on InAs QDs. The nanocrystal elongation is along $[0\bar{1}1]$. (b) The same image with expanded height scale. (c) Close-up of an individual Ag nanocrystal-QD pair and a reference image of a single InAs QD with 3 nm GaAs cap layer.

droplet epitaxy approach.¹³ One monolayer In was deposited at 100 °C to form In nanocrystals, which were transformed into InAs QDs by annealing under As_4 flux initially at 100 °C and then at 450 °C, where the QDs were capped by 3 nm GaAs. For Ag nanocrystal growth the samples were transferred into an attached metal MBE chamber. The substrate temperature was kept between 250 and 300 °C, the growth rate was 0.1 nm/min, and the Ag coverage amounted to 0.5–2 nm. The morphology of the samples was characterized by tapping-mode atomic force microscopy (AFM) under ambient conditions. The SPRs of the Ag nanocrystals were measured by differential reflectance (DR) spectroscopy.¹⁴ A halogen lamp and a double quarter-meter monochromator were used as a tunable light source and the light reflected from the sample surface was detected by a PbS photoresistive sensor. Photoluminescence (PL) measurements were performed with the samples placed in a continuous-flow He cryostat at 10 K. A long working distance objective (NA 0.5) was used to excite the samples and collect the emitted light. A 630 nm semiconductor laser was used as excitation source. The PL was dispersed by a single quarter-meter monochromator and detected by a liquid-nitrogen cooled InGaAs photodiode array.

Figures 1(a)–1(c), left panels, present AFM images of the morphology of the Ag nanocrystals on GaAs for various growth conditions. The hut-shaped nanocrystals exhibit a clearly faceted surface and tend to elongate in the $[011]$ direction, see inset in Fig. 1(b), what reveals that they have a well-defined epitaxial relation with the substrate. Formation of epitaxial metal nanocrystals on semiconductor surfaces is not uncommon and has been reported long ago in the Ag/Si(100) materials system.¹⁵ The size and density of the Ag nanocrystals are easily controlled by varying the deposition temperature and coverage. With increase in the coverage, the average island size increases and with increase in the substrate temperature, the density decreases due to higher adatom mobility. The lower density results in increased nanocrystals size, which can be compensated by lowering the coverage.

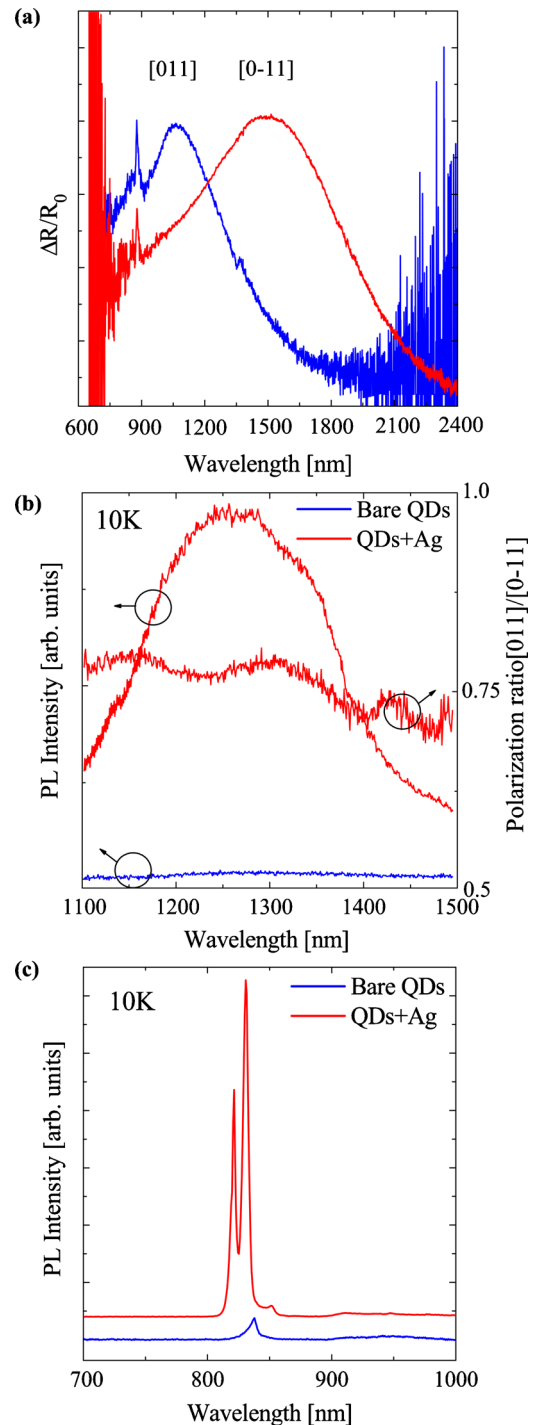


FIG. 3. (Color online) (a) DR spectra for light polarized along $[011]$ and $[0\bar{1}1]$ of the sample with Ag nanocrystal-QD hybrids. The sharp feature around 900 nm is due to the GaAs band gap. (b) Low-temperature PL spectra of the QDs of the metal-QD hybrid structures (including the intensity ratio of the linear polarized PL along $[011]$ and $[0\bar{1}1]$) and of a reference sample with only near-surface QDs. (c) Low-temperature PL spectra of the metal-QD hybrid structures and of a reference sample without Ag nanocrystals.

As also shown in Figs. 1(a)–1(c), right panels, the SPRs of the Ag nanocrystals, measured by DR spectroscopy shift from 1 to 1.7 μm due to increasing nanocrystal size determined by the growth conditions. Though certainly not the limit this wavelength range is of particular interest covering the second and third telecom bands at 1.3 and 1.5 μm . As mentioned above, the Ag nanocrystals tend to elongate. This

results in strong linear polarization dependence of the SPR response, shown in Fig. 1(c) with peak separations of 100–200 nm for light polarized parallel and perpendicular to the long axis. The long wavelength resonance for the light polarized along the long axis is broader, which we attribute to larger inhomogeneous broadening caused by larger size variation in this direction.

When Ag is not deposited on bare GaAs but on InAs QDs capped with a thin GaAs layer, the Ag nanocrystals nucleate right on top of the QDs. The thickness of the GaAs cap layer controls their separation. This is evident when comparing the AFM topography data shown in Figs. 2(a)–2(c). The morphological feature corresponding to a QD after capping has an elongated shape and height of 3–4 nm, see right panel in Fig. 2(c). On the sample with Ag such features protrude from beneath the nanocrystals having a height around 80 nm. They are visible when the height scale is expanded in Fig. 2(b) and left panel of Fig. 2(c) compared to Fig. 2(a). No such features are observed for Ag deposited on the bare GaAs surface. From careful analysis of the AFM images we conclude that every Ag nanocrystal has a QD beneath, so the probability of alignment is 100%. This is achieved since the Ag nanocrystal density is matched within 10% to the QD density, $1.7 \mu\text{m}^{-2}$ versus $1.8 \mu\text{m}^{-2}$. The corresponding Ag coverage and growth temperature are 0.5 nm and 400 °C. Interestingly, in case of deposition on top of the QDs the direction of the Ag nanocrystal elongation is changed from [011] to $[0\bar{1}1]$, the latter being the direction of elongation of the QDs. It is thus evident that both nucleation and growth of the Ag nanocrystals are modified by the presence of the QDs revealing a route to template-based shape control.

The optical properties of the samples with hybrid metal-semiconductor nanostructures are clearly different from those of the samples with solely QDs or Ag nanocrystals. Due to the different direction of elongation of the Ag nanocrystals the polarization properties of the SPR modes are reversed. The nanocrystals exhibit SPR peaks at 1.5 and $1.1 \mu\text{m}$ for light polarized along $[0\bar{1}1]$ and [011], shown in Fig. 3(a). Most important, the presence of the Ag nanocrystals results in a large intensity enhancement of about one order of magnitude of the PL of the QDs underneath the Ag nanocrystals centered at $1.25 \mu\text{m}$, shown in Fig. 3(b). A similar enhancement of the intensity is found for the bulk GaAs emission, see Fig. 3(c). The increased PL intensity can in principle arise from an increase in the light absorption, spontaneous emission rate, or both. Moreover a positive balance of radiative and nonradiative recombination has to be maintained. For the QDs this is provided by the 3 nm GaAs

cap layer. Taking into account that the areal coverage of Ag is about 2.8% and that the SPR mode volume extends only over a few nanometer it is unlikely that the observed PL enhancement of bulk GaAs is due to enhanced emission or scattering. In addition, there is only a weak trend of the linear polarization behavior of the QD PL following that of the SPRs, shown in Fig. 3(b). For purely SPR enhanced emission a stronger polarization dependence of the PL would be expected.¹⁶ Hence, we attribute the PL enhancement mainly to near-field enhanced absorption of the exciting laser light, also in resonance with the SPR, and thus greater carrier injection into GaAs and the QDs, where the alignment is essential.

In conclusion, Ag nanocrystals were grown epitaxially on GaAs by MBE. Their size and density was tuned by varying the substrate temperature and coverage resulting in strong shifts of the SPRs at telecom wavelengths. When deposited on near-surface InAs QDs the Ag nanocrystals self-align on top of the QDs. This opened the door to synthesize hybrid metal-semiconductor nanostructures with precise control of the lateral, due to the self-alignment, and vertical, due to the cap layer, QD to metal separation. PL measurements revealed clear intensity enhancement, which was attributed to SPR-enhanced absorption of the exciting light.

- ¹S. A. Maier and H. A. Atwater, *J. Appl. Phys.* **98**, 011101 (2005).
- ²D. J. Bergman and M. I. Stockman, *Phys. Rev. Lett.* **90**, 027402 (2003).
- ³M. Noginov, G. Zhu, A. Belgrave, R. Bakker, V. Shalaev, E. Narimanov, S. Stout, E. Herz, T. Suteewong, and U. Wiesner, *Nature (London)* **460**, 1110 (2009).
- ⁴M. Stockman, *J. Opt.* **12**, 024004 (2010).
- ⁵O. Kulakovich, N. Strekal, A. Yaroshevich, S. Maskevich, S. Gaponenko, I. Nabiev, U. Woggon, and M. Artemyev, *Nano Lett.* **2**, 1449 (2002).
- ⁶T. Pons, I. L. Medintz, K. E. Sapsford, S. Higashiya, A. F. Grimes, D. S. English, and H. Mattoussi, *Nano Lett.* **7**, 3157 (2007).
- ⁷R. K. Kramer, N. Pholchai, V. J. Sorger, T. J. Yim, R. Oulton, and X. Zhang, *Nanotechnology* **21**, 145307 (2010).
- ⁸A. Urbańczyk, G. J. Hamhuis, and R. Nötzel, *Appl. Phys. Lett.* **96**, 113101 (2010).
- ⁹J. He, R. Nötzel, P. Offermans, P. M. Koenraad, Q. Gong, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, *Appl. Phys. Lett.* **85**, 2771 (2004).
- ¹⁰J. Tersoff, C. Teichert, and M. G. Lagally, *Phys. Rev. Lett.* **76**, 1675 (1996).
- ¹¹T. Mano, R. Nötzel, G. J. Hamhuis, T. J. Eijkemans, and J. H. Wolter, *J. Appl. Phys.* **95**, 109 (2004).
- ¹²Q. Xie, A. Madhukar, P. Chen, and N. P. Kobayashi, *Phys. Rev. Lett.* **75**, 2542 (1995).
- ¹³T. Chikyow and N. Koguchi, *Jpn. J. Appl. Phys., Part 2* **29**, L2093 (1990).
- ¹⁴R. Lazzari, S. Roux, I. Simonsen, J. Jupille, D. Bedeaux, and J. Vlioger, *Phys. Rev. B* **65**, 235424 (2002).
- ¹⁵M. Hanbücken, M. Futamoto, and J. Venables, *Surf. Sci.* **147**, 433 (1984).
- ¹⁶H. Mertens, J. S. Biteen, H. A. Atwater, and A. Polman, *Nano Lett.* **6**, 2622 (2006).