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Single photon emission from ODT passivated near-surface GaAs quantum dots

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Epitaxially grown semiconductor quantum dots (QDs) are one of the most promising candidates for efficient quantum light sources. Excellent optical properties can typically be ensured only if these so-called 'artificial atoms' are buried deep inside the semiconductor host material (≥ 100 nm). The optical properties of semiconductor micro- and nanostructures are greatly affected by their surface. Defects in the crystal lattice usually result in additional electronic states in the bandgap (so-called surface states) which is detrimental for radiative recombination processes and charge carrier transport. Quantum dots grown close to the surface are prone to charge carrier fluctuations and trap states on the surface, degrading the brightness, coherence and stability of the emission. However, some hybrid nanophotonic devices with near-field coupling, such as QDs coupled with surface plasmons or a single photon transistor, require a very small distance (< 25 nm) between the plasmon and the QD emitter. This raises the need for effective surface passivation methods to recover the optical properties for near-surface QDs.

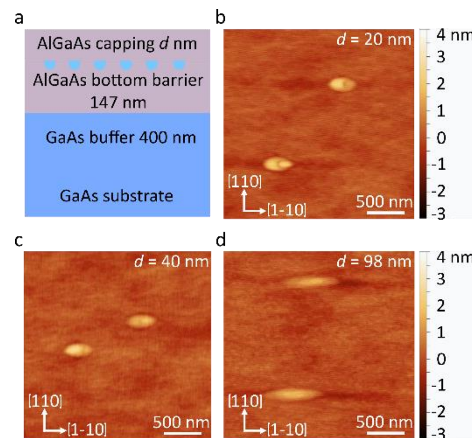

Fig. 1: (a) Schematic of the sample structure. (b)-(d) Atomic force microscopy (AFM) images of GaAs QDs with a capping layer thickness of d=20 nm, 40 nm, 98 nm, respectively. The elevated structures on the surface indicate the location of the nanoholes below the capping layer.

The GaAs QD sample is grown on an undoped GaAs (001) substrate with solid source molecular beam epitaxy via the local droplet etching method. The GaAs QDs are in-filled in the Al droplets etched nanoholes and then capped with AlGaAs barrier layer with a thickness of 98 nm, 40 nm, and 20 nm, respectively, as shown in Fig. 1(a). Fig. 1(b)-(d) show the surface morphology of the as-grown samples investigated with an atomic force microscope (AFM). With 20 nm capping, the dips in the bumps are still observable, showing the location of the nanoholes which are partially in-filled by QDs. With increasing the capping thickness from 40 nm to 98 nm, only bumps remain on the surface. This is caused by the capillary force of the nanoholes and surface reconstruction. The ability to observe the QDs location with 20 nm capping is an advantage for the realization of hybrid QD based devices cupled to surface plasmonic nanoparticles, without any need for optical positioning.

The analysis of optical properties of GaAs QDs with three capping thicknesses before and after ODT passivation is based on the measurement of the micro-photoluminescence (PL) spectra. All the spectra show a dominant neutral exciton peak and several charged exciton peaks at the red-shifted side, which is a fingerprint of single GaAs QD emission. By reducing the capping thickness, the intensity of the exciton peaks of the as-grown sample is significantly decreased and the linewidth is broadened. This indicates the presence of extra decay channels caused by surface states as well as the influence of electric fields from the charge carrier fluctuations at the surface via the DC Stark effect. After ODT passivation, it is difficult to observe any change from the PL spectra for the sample with 98 nm

capping. For 40 nm and 20 nm, the intensity of the exciton emission is enhanced after passivation. Even at a low capping thickness of 20 nm, the different excitonic peaks can be clearly resolved. The statistic results of the linewidth and peak intensity is shown in Fig. 2(a)-(b).

Fig. 2 (c) and (d) show the second-order autocorrelation measurement of the as-grown and passivated sample with 20 nm capping. Both samples are pure single photon emitters, proven by the low values of $g^{(2)}(0) = 0.016 \pm 0.015$ and 0.042 ± 0.014, respectively. These values have not been corrected for detector dark counts. The autocorrelation measurement on the as grown sample shows a slight bunching effect. This is attributed to blinking in the exciton emission induced by trap states. After ODT passivation, the bunching effect is not observed, indicating a reduction of the density of trap states. In addition, the anti-bunching dip is broadened after ODT passivation. On the one hand, this is a clear sign that the non-radiative transition channels have been reduced and the lifetime of the exciton is prolonged, consistent with the previous linewidth measurements. On the other hand, the passivation may lead to a stronger phonon bottleneck in the QDs, since excited exciton recombination dynamics can be affected by surface states. Broadening of the anti-bunching dip can then be attributed to slower excited exciton relaxation.

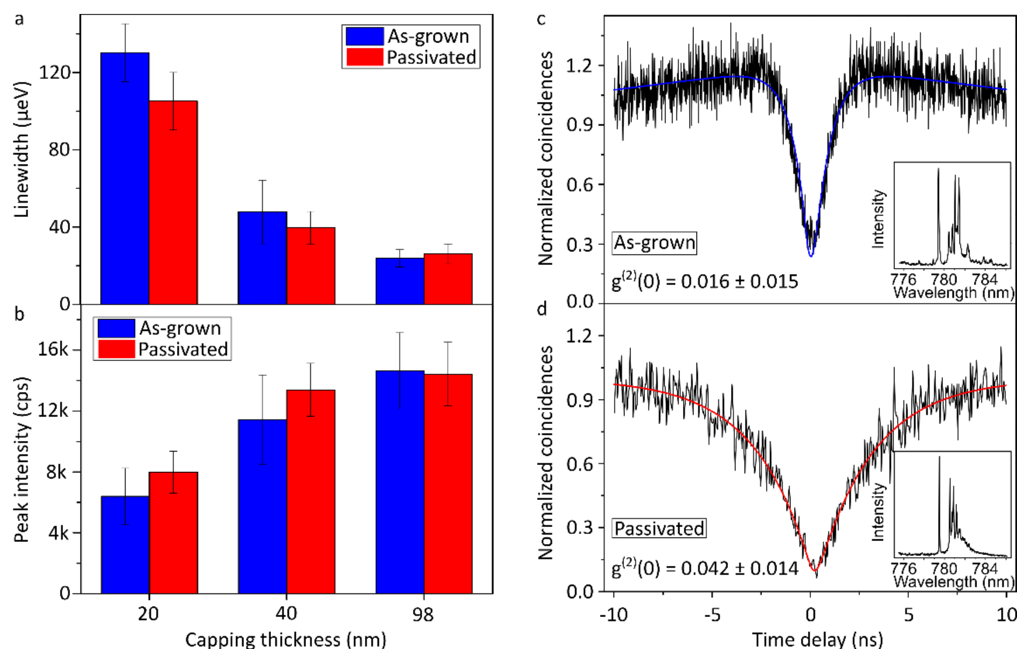

Fig. 2: Left panel: Comparison of linewidth (a) and saturated peak intensity (b) of the neutral exciton emission from GaAs near-surface QD samples before and after passivation. The linewidth is 23.9/26.1 µeV for 98 nm capping, 47.8/39.6 µeV for 40 nm capping and 130.3/105.1 µeV for 20 nm capping for the as grown/passivated samples, respectively. Right panel: Second-order autocorrelation measurement of as grown (c) and passivated (d) sample with 20 nm capping. The anti-bunching dip at zero-time delay indicates single photon emission. Inset: PL spectra of the measured QDs.

In conclusion, we have shown that the quantum dot emission is not influenced by surface states when the dots are buried around 100 nm beneath the surface. By reducing the quantum dot to surface distance from 40 nm to 20 nm, surface states have stronger influence on the QD emission. The addition of non-radiative decay channels and charge fluctuations results in linewidth broadening and fluorescence quenching. By applying chemical passivation with ODT, the surface states are partially eliminated, inducing a partial recovery of linewidth and intensity. The single photon emission properties and excitonic spectral features are very well preserved even for 20 nm capping. The presence of blinking in the single photon emission is reduced with ODT passivation. The passivation of near surface quantum dots paves the way for hybrid nanophotonic devices where near-field coupling is needed, such as surface plasmon coupling. Although ODT can partially passivate the surface states, the surface states cannot be totally eliminated, which motivates the investigation of more efficient methods for complete passivation of the semiconductor surface. Applying more reactive sulphur compounds such as $(NH_4)_2S_x$ may yield better passivation.

For further information see: X. Cao, J. Yang, P. Li, Y. Zhang, E. P. Rugeramigabo, B. Brechtken, R. Haug, M. Zopf, F. Ding, *Single photon emission from ODT passivated near-surface GaAs quantum dots*, Appl. Phys. Lett. **118**, 221107 (2021)