Single dot spectroscopy of site-controlled InAs quantum dots nucleated on GaAs nanopyramids

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Single InAs quantum dots, site-selectively grown by a patterning and regrowth technique, were probed using high-resolution low-temperature microphotoluminescence spectroscopy. Systematic measurements on many individual dots show a statistical distribution of homogeneous linewidths with a peak value of ~120 μeV, exceeding that of unpatterned dots but comparing well with previously reported patterning approaches. The linewidths do not appear to depend upon the specific facet on which the dots grow and often can reach the spectrometer resolution limit (<100 μeV). These measurements show that the site-selective growth approach can controllably position the dots with good optical quality, suitable for constrained structures such as microcavities. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2790498]

Semiconductor quantum dots (QDs) continue to be the subject of intensive research due to their atomlike density of states that makes them appealing for applications in photonics and quantum information.[1] Particularly promising is their use as single photon sources[2,3] toward quantum cryptography applications, potentially overcoming the limited speed and efficiency of existing approaches. This is because QDs can feature very narrow linewidths, i.e., long coherence times, and because they can simultaneously be monolithically embedded in high *Q* optical resonators for enhanced light-matter coupling.

However, self-assembled QDs, while possessing high optical quality, suffer from high densities, nucleation at random sites, and coupling to a wetting layer. These characteristics often limit the capabilities of the dots in device engineering. Thus, techniques aimed at site-selective growth have been vigorously investigated, ultimately aiming at deterministic placement of a single dot at a given spatial location. Site-controlled single dot emission has been studied in various material systems such as InAs/GaAs,[4,5] InAs/InP,[6,7] InGaAs/AlGaAs,[8] and GaAs/AlGaAs.[9,10] In these studies, a key requirement was that the site-selective pattering does not lead to a deterioration of the dots' optical quality, i.e., homogeneous linewidths, which are intimately related to their emission efficiency and their ability to couple to microcavities.

Here, we report on the statistical analysis of the homogenous linewidths of single site-controlled QDs in the InAs/GaAs system, which, in principle, can work at telecommunication wavelengths and with which most solid-state cavity quantum electrodynamics (QED) experiments are realized. In cavity QED, where a single QD is to be coupled to a high *Q*, low mode volume optical microcavity, the dot's position is crucial, thus renewing interest in site-selective growth methods. Recently, for example, deterministic positioning in a photonic crystal membrane has been achieved by using tracer dots to determine the dot's position before etching the cavity around it.[11] Site-selective growth could streamline this process, and promising attempts to achieve this have been reported.[6,12] Site-selectively coupled QD/microcavity systems could also be realized with alternative approaches such as Bragg reflectors[13] or microdisks,[14] which are promising for strong coupling and few QD lasers.

In our low-temperature experiments, hundreds of photoluminescence (PL) emission peaks [such as those shown in Fig. 1(b)] from single QDs grown on pyramidal structures with different limiting crystal facets were systematically measured. We find that these PL linewidths are widely scattered from resolution-limited values below 100 μeV up to values of a few meV, showing a distribution of the homogeneous linewidths peaked at ~120 μeV. Our data suggest that these linewidths do not depend on the size and shape of the quantum dots, nor on the facet on which they nucleate, indicating that the broadening is associated with materials processing.

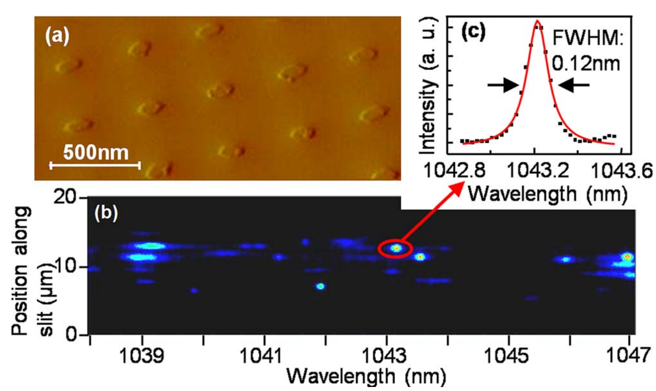

FIG. 1. (Color online) (a) AFM image of capped sample. (b) Typical spectrum recorded with 180 s integration time. The *y* axis corresponds to the spatial axis along the spectrometer slit while the *x* axis denotes wavelength. Each peak is from a single QD emission. The peaks are most likely from the ground state due to the low temperature and the excitation intensity. (c) Intensity profile of one QD state with a Lorentzian line shape.

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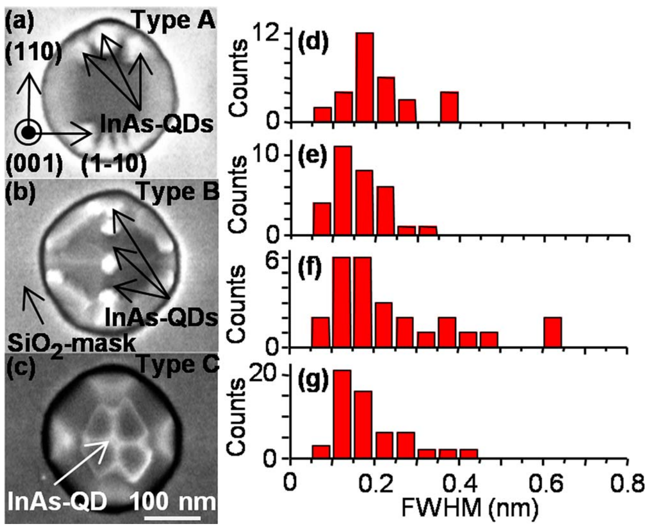

FIG. 2. (Color online) [(a)–(c)] Morphology of patterned QDs grown on the three different kinds of pyramids. The histograms show the distribution of the linewidth for a certain set of pyramids: (d) data from type A, (e) data from type B, and [(f)–(g)] data from type C. Each bin in a histogram covers a range of 0.05 nm. The data were taken at a temperature of 6 K.

The sample was grown with metal organic chemical vapor deposition under low pressure (60 Torr) on a GaAs (001) substrate. The substrate was initially covered with a SiO_2 mask in which a two dimensional array of circular openings of 230 nm in diameter and a pitch of 330 nm were etched using interferometric lithography and dry etching. In these holes, three different types of pyramidal GaAs buffers, A, B, and C, each with different facets, were grown. The InAs QDs were subsequently grown on the pyramids. The patterned QDs whose morphology has been previously characterized by scanning electron microscopy can be seen in Figs. 2(a)–2(c). Size, shape, and number of dots per pyramid are different for each kind of pyramid but consistent within each set. For example, pyramids of type C are limited by facets of {111}, {011}, and {113} groups and a (001) apex on which one large single dot nucleates. For PL measurements, the dots were capped with InGaAs and GaAs. The sample growth and the morphology of the pyramids and dots are described elsewhere in more detail.[15]

The micro-PL measurements were conducted from 6 to 55 K. A frequency doubled diode pumped Nd:yttrium aluminum garnet laser ($\lambda = 532$ nm) was used to excite the dots above the GaAs bandedge. The excitation intensity was held constant throughout the measurements at around 2.4 W/cm^2 to ensure that the dots' ground states are predominantly observed. The imaging scheme utilized by our setup preserves the spatial information along one axis which is the ordinate in the spectrum.[16] To confirm that the location probed contained patterned pyramids, we also recorded additional surface topography images as shown in Fig. 1(a), with atomic force microscopy (AFM) prior to our measurements. From raw data such as those shown in Fig. 1(b), the dots' line profiles and linewidths were extracted. We could observe peaks whose linewidths reached the resolution limit of our spectrometer (<100 μeV) as well as peaks with widths in the order of meV. Only peaks with an unambiguous Lorentzian line shape were considered in the statistics on the full width at half maximum (FWHM) of the peaks. For example, the peaks at 1041.9, 1043.2, and 1043.6 nm in Fig. 1(b) have very clear Lorentzian intensity profiles. With the silicon charge coupled device we used, emission peaks from single dots could be found in the spectral region between 950 and 1150 nm wavelengths. From the measured emission peaks, we extracted 145 Lorentzian-shaped linewidths of dots from all three kinds (A, B, and C) of pyramids to see if the FWHM depends on the shape and size of the dots or on the facet on which they nucleate. Our results show that the distributions of the FWHM [see Figs. 2(d)–2(g)] resemble each other for all three sets of pyramids. The distribution is always peaked at linewidths around 0.1–0.2 nm which correspond to energies of about 120–220 μeV. No indications of a dependence of the FWHM on the emission energy or pump power were observed. Although our measurements do not cover the whole spectral range over which the QDs emit light (limited detector sensitivity range), the lack of an energy dependence of the linewidth suggests that the measured linewidths used in the statistics are representative of all QDs.

These linewidths are similar to the results of other groups on similar material systems. Linewidths of 140 μeV have been measured in patterned InGaAs/AlGaAs QDs grown on inverted AlGaAs pyramids.[8] For patterned GaAs/AlGaAs dots grown in lithographically defined nanoholes, homogenous linewidths of 135 μeV have been reported.[10] Our result shown in Figs. 2(d)–2(g) and the previous results mentioned above are all broader than those found in unpatterned self-assembled InAs/GaAs dots for which the width is routinely below 50 μeV.[17] As the widths do not seem to be dependent on the dots' shape, size, or nucleation facet, it is likely that this broadening is due to the patterning process. Such broadening is well-known in micro- and nanocavities such as micropillars or photonic crystals where dots are positioned near the etched interfaces. In those cases, broadening is likely due to the coupling of excitons to surface states, which results in dephasing and/or spectral diffusion.[18] Similarly, in our case, we suspect that the existence of unpassivated surface states at the GaAs interface introduced in the etching process is the cause of broadening. Nevertheless, our results show that the optical quality of single QDs is sufficiently high on all three types of pyramids, with long enough dephasing times T_2 for quantum optical device applications. The small heights of the pyramids between 30 and 90 nm make this method of site-selective QD growth suitable for applications involving microcavities. For such purposes, the pyramids are shallow enough that they can be overgrown with distributed Bragg reflectors[19] to form a three dimensional cavity or be used in photonic crystal membranes.[12] The wide distribution of the linewidths among dots is likely due to inhomogeneities of the solid-state matrix in proximity of the quantum dots caused by the etching of the sample that roughens the interface.

Temperature-dependent measurements of the single dot PL linewidth have also been carried out. Starting at 6.8 K, the temperature was incremented up to 55 K and the line shape and linewidth were recorded. As shown in Fig. 3, a single patterned QD linewidth has a remarkably similar temperature dependence to that of an unpatterned QD. It is characterized by a thermal activation, usually involving a Bose-Einstein occupation number due to coupling to phonons.[20] The sudden increase of the linewidth at 40 K is in good agreement with the results found for unpatterned QDs.[21] This temperature activation plays an important role in achieving room temperature device operation (or at least thermoelectri-

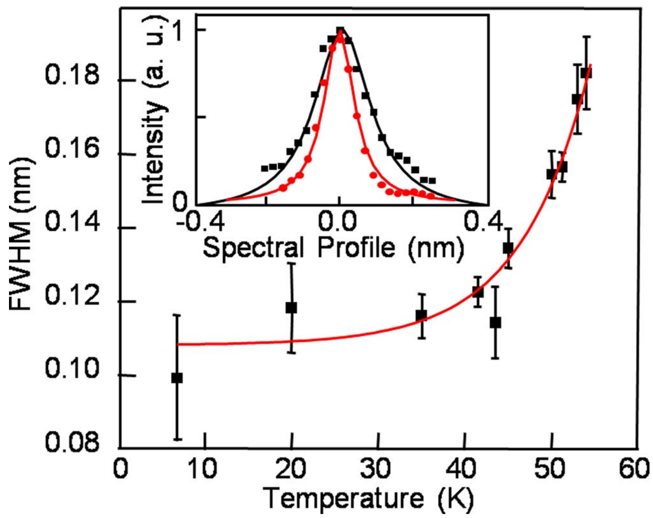

FIG. 3. (Color online) Temperature dependence of the FWHM of one QD. The linewidth increases suddenly at 40 K. Inset: intensity profiles at 6.8 K (FWHM=0.1 nm) and 45 K (FWHM=0.18 nm). Uncertainty in Lorentzian curve fitting represents the dominant contribution to the error bars shown.

cally cooled environments that can reach ~150 K). For example, the spontaneous emission enhancement in cavity-coupled single photon sources saturates as the QD linewidth exceeds the cavity linewidth. We finally note that ensembles of these patterned QDs exhibit strong PL at room temperatures.[15]

In summary, single dot PL measurements have been performed on different types of site-controlled InAs/GaAs QDs. The linewidths, whose broadening is most likely primarily due to the etching process, prove that the patterned InAs/GaAs QDs are of good quality and could be used in optoelectronic devices such as single quantum emitter light sources. Better control of materials growth and processing should enable smoother interfaces so that the patterned QDs' quality can be substantially improved. The small size of the buffers makes this method of site-controlled QD growth suitable for embedding into microcavities, and future efforts will target this direction.

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