Origin of Spectral Brightness Variations in InAs/InP Quantum Dot Telecom Single Photon Emitters

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Long-distance quantum communication relies on the ability to efficiently generate and prepare single photons at telecom wavelengths. Low density InAs quantum dots on InP surfaces are grown in a molecular-beam epitaxy system using a modified Stranski-Krastanov growth paradigm. This material is a source of bright and indistinguishable single photons in the 1.3 μm telecom band. Here, exploration of the growth parameters are presented as a phase diagram, while low-temperature photoluminescence and atomic resolution images are presented to correlate structure and spectral performance. This work identifies specific stacking faults and V-shaped defects that are likely causes of the observed low brightness emission at 1.55 μm telecom wavelengths. The different locations of the imaged defects suggest possible guidance for future development of InAs/InP single photon sources for c-band, 1.55 μm wavelength telecommunication system.

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I. INTRODUCTION

The desire to synthesize InAs quantum dots (QDs) that operate at conventional optical telecommunication wavelengths is long-standing.[1,2] More recently, the desire has evolved to use InAs QDs as a source of individual and indistinguishable photons as a building block for emerging optical quantum information systems. Self-assembled InAs QDs is a leading technology for creating the single photon source because they have demonstrated single photon operation as determined through the low value of the single photon second order correlation value, $g_1^{(2)}(0) \leq 0.5$, using a Hanbury-Brown and Twiss measurement, and indistinguishable photons as determined by a high value of the interference visibility that is calculated from differences in the two-photon second order correlation function, $[g_a^{(2)}(0) - g_b^{(2)}(0)]/g_b^{(2)}(0) > 0$, in a Hong-Ou-Mandel measurement. Some demonstration of single photon sources accomplished with InAs QDs sandwiched between AlGaInAs layers,[3] and InAs QDs grown on a GaAs wetting layer.[4] Both metal organic chemical vapor deposition (MOCVD)[5] and molecular beam epitaxy (MBE)[6] have been used to produced InAs QDs on InP. All of these demonstrations are with QD materials with dot densities around 50 μm^{-2}, which is significantly lower than that used for QD lasers.

Many current experiments in quantum information are conducted at wavelengths near 1-μm as a result of the emission spectrum of InAs QD nanostructures grown on GaAs substrates. With motivation to leverage the components and installed infrastructure in the 1.55 μm telecommunication band the constraints on the operating wavelength of the single photon source in quantum information devices are increasing. In contrast to InAs on GaAs, InAs QDs grown on InP are attractive because of the smaller misfit strain and therefore perceived ability to grow larger QDs that emit at telecom wavelengths. Depending on the growth conditions, both MOCVD and MBE have produced QDs with shape anisotropy resulting in quantum dashes or elongated sticks with high QD densities that exhibit large inhomogeneous broadening and non-distinct spectral emission lines, which are both less desirable characteristics for single photon sources in optical quantum information systems.

The materials reported here are produced using solid source molecular beam epitaxy using a modified Stransk-Krastanov growth mode to produce circular InAs QDs on InP with a controllable dot density.[7] These QDs emit strongly at wavelengths near 1.3-μm and have been incorporated into a heterostructure design that has been fabricated into a number of devices that leverage the indistinguishable single photons produced. These devices include: 1) a nanophotonic cavity demonstrating a Purcell enhanced spontaneous emission rate of up to 4 and bright single photon emission ($g_1^{(2)}(0) \sim 0.08$) that exhibits indistinguishable visibilities of 67% with post-selection;[8] 2) demonstration of two-photon interference from two independent cavity-coupled emitters on the same chip that are spaced 15-μm apart with resonance matching better than 3 μeV;[9] 3) hybrid integration of QDs to a silicon photonic device using a nanometer resolution pick-and-place technique to enable an integrated photonics Hanbury-Brown and Twiss measurement;[10] and 4) incorporation of two QDs in a single nanophotonic device to create quantum interactions between the QDs that create super-radiant emission.[11] The single photon emission characteristics reported in these references are indicative of the materials reported here as they are grown with the same recipe and during the same growth campaign.

The specific growth recipe developed for the materials reported here does not follow the conventional Stranski-Krastanov growth process. While some details of the growth recipe have been reported[7] the parameter space and dependence on growth temperature and overpressure have not. Here, a phase diagram presents the results from exploring these conditions and variation in the formation temperature of the QDs using the multi-temperature growth process described herein.

To achieve the performance requirements of quantum communications, i.e., small single- and two-photon second order correlation functions at zero delay, thermal noise needs to be eliminated; devices are therefore expected to operate at cryogenic temperatures. Reports of strong emission from macroscopic room temperature PL do not necessarily correspond to strong low-temperature microscopic PL of isolated dots.[7,12] Therefore, characterization of spectral variations in the emission wavelengths at low-temperature is measured and presented.

Characterization of QD materials using probe corrected scanning transmission electron microscopy (STEM)[13] provides a means to investigate the nature of microscopic structure defects and their origin that transmission electron microscopy has difficulty resolving.[14,15] Here, we report on atomic resolution STEM images that lead to the identification and origin of crystal defects that are likely to preclude MBE grown InAs QDs on InP to efficiently operate at the 1.55 μm telecommunications c-band. By identifying the specific mechanisms, several approaches to minimizing these defects and possibly extending QD emission wavelengths are suggested.

II. GROWTH

Low-density ($< 50/\mu m^{-2}$) strained InAs QDs are grown on InP using a modified-SK-growth mode in a V-80H solid source molecular beam epitaxy system with both arsenic and phosphorous valved crackers and two effusion cells loaded with indium in order to grow InAs and InP with different growth rates without the need to ramp cell temperatures. Substrate temperatures are measured with an optical pyrometer that is calibrated to the temperature of oxide desorption from GaAs substrates at 583^{o}C.

Quantum dots are grown on (100)-oriented InP substrates (either S- or Fe-doped) that have had the native oxide thermally desorbed in the growth chamber under a phosphorous overpressure and growth of an approximately 200-nm-thick unintentionally-doped InP buffer layer. The growth process of the InAs dots is a multiple-temperature recipe that makes extensive use of kinetically controlled group-V exchange.[7]

Quantum dots are grown on a P_2 stabilized InP surface after the substrate temperature is stabilized at 525^{o}C. The group-V overpressure is changed to As_2 and 10 s later approximately 0.8 ML of InAs is deposited at a growth rate of 0.56Å/s and a V/III BEP ratio of ∼40. The InAs surface is annealed at 525^{o}C for 120 s. The wafer is then cooled under As_2 overpressure at a rate of 15^{o}C/min to 450^{o}C. During this cooling process the reflection high energy electron diffraction (RHEED) reconstruction changes from a streaky 2x4 pattern, to a pattern that contains both chevrons and streaky 4x2 pattern.[7] The temperature of this transition is dependent on the As-overpressure.

Numerous experiments were performed while monitoring the RHEED pattern with the electron beam parallel to either the [110] or [$\bar{1}$10] directions using various values of As_2 over-pressure. By repeating each experiment twice and monitoring the diffraction pattern from both orientations the temperature boundary of the surface reconstruction and the formation temperature of spots indicating QD formation is determined. In general, the streaky 2x4 reconstructed surface changes to a 4x2 reconstructed surface and chevrons associated with QDs appear at the same temperature. The results of these experiments are summarized in figure 1. The preferred orientation of the non-rotating substrate for these experiments is for the electron beam to be parallel to the [110]. This orientation expresses the 4x peaks at

FIG. 1. (Color online) Phase diagram of the surface reconstruction and topology as determined from RHEED

high temperatures, which disappear at the surface reconstruction transition temperature.

Fortuitously, these experiments were completed on a single substrate which is possible through the reversibility of group-V exchange. Following each QD phase boundary experiment, the arsenic source was turned off, and a phosphorous overpressure was introduced. This causes the InAs to convert into InP. Following a 5 min P_2-soak, 100-300 nm of InP was grown resulting in a streaky 2x4 surface. Following these multiple experiments, a final set of InAs QDs were grown in accordance to the standard recipe. The dot shapes and density were then measured with AFM and determined to be consistent to other samples that are grown on pristine substrates.

Buried QDs are grown for optically active samples. Following stabilization of the substrate at 450^oC under an As_2 flux, the overpressure is changed to P_2, and the sample is soaked for 28 s. This step removes the InAs wetting layer. Next, a thin layer of InP that is only 2.7-nm thick partially buries the QDs. The sample is soaked under P_2 flux for 60 s to remove the top of the QDs with the goal of changing the natural QD shape into a quantum disk, that has been shown to decrease the inhomogeneous broadening. Finally a 100-nm thick InP cap is grown to completely bury the QDs. More details about the double-layer capped growth sequence can be found in previous reports including the observation that conversion of InP into InAs results in the final sample having more InAs than what is provided during the 0.8 ML deposition.[7]

III. LOW TEMPERATURE OPTICAL CHARACTERIZATION

Optical characteristics of these QD samples is an important aspect of evaluating fitness of the epitaxial materials. Figure 2 shows low temperature micro-photoluminescence (PL) measurement results of QDs in the as-grown film collected with a confocal microscope. The QD samples are cooled to 4 K using a low vibration closed cycle cryostat and optically pumped with a 785 nm continuous wave laser. The confocal microscope is constructed with an objective lens that has a numerical aperture of 0.7, producing a pump laser spot size of 1 μm. The spectrometer is fitted with a liquid-nitrogen-cooled InGaAs array.

FIG. 2. Low temperature optical characteristics of (a) single quantum dot bi-exciton luminescence, (b) low-temperature macro luminescence indicating of a region containing a bright quantum dot and of a (c) region not-containing bright quantum dots.

Each measurement illuminates approximately 20 QDs. The luminescence shown in 2(b) shows one of the longest emission wavelengths observed with high brightness as indicated by high count rates, while 2(c) is an example of another region where no bright emission is observed. It has been observed that bright QDs have low-temperature emission wavelengths that are less than 1400 nm. The observed emission wavelength boundary is the driving motivation for all single photon nanophotonic device demonstrations to be completed in the 1300-nm telecommunication band. While only three spectra are selected for presentation, numerous measurements are made per wafer and over a dozen wafers have been evaluated. The trend is consistent among all wafers produced even those grown under various As-overpressures.

Because of the low QD density, it is common for several QDs to show bright emission. However, inhomogeneous broadening in the QD ensemble causes the emission wavelengths of the isolated QDs to be spectrally distinct. In figure 2(c), a high resolution scan of the emission from a single QD is shown. The measured full width at half maximum of both the single exciton, X, and double exciton, XX, peaks are less than system resolution of approximately 50 μeV. The biexciton binding energy is 1.4 meV.

IV. SCANNING TRANSMISSION ELECTRON MICROSCOPY

The high resolution electron microscopy was performed on an aberration-corrected 200 keV JEOL ARM instrument in STEM mode with a high angle annular dark field (HAADF) detector. The studied specimens were prepared using a gallium focused ion beam (FIB) and inspected with the electron beam propagating in the $\langle 110 \rangle$ direction. All of the images presented here are representative of imaged QDs. There is no direct correlation between emission and structure of individual QDs because the technical challenges to measure the emission of a specific QD, mark it, prepare a TEM specimen, and then image that same QD are too great for the scope of this project.

The combination of low QD density and nominal 100-nm thickness of the TEM foils

FIG. 3. Scanning transmission electron micrograph of a typical dome shaped quantum dot with wetting layer.

FIG. 4. Double capped InAs quantum dot with the wetting layer partially removed resulting in a quantum dot with the top of the dot removed.

allows cross-sectional investigation of isolated QDs, with the caveat that most of the imaged area of the prepared TEM specimens do not contain any QDs. Since only a few QDs were found, the images presented here should be interpreted as representative. The QD shown in figure 3 shows an as-formed QD that has been covered with a single cap, i.e. the top of the QD has not been removed. The dome shape is clearly evident as is the tapering thickness that decreases down to a single monolayer wetting layer. This shape is consistent with other reports of InAs QDs grown in InP.[16] While not shown clearly in these figures, the wetting layer is indeed one monolayer thick supporting the interpretation of the room temperature PL[7] and that it is only partially removed by the initial P_2 soak at 450^oC even though the wetting layer no longer expresses a peak in the room temperature macroscopic PL spectrum.

The effect of the double-layer capped heterostructure is evident in figure 4. These images show that the arsenic that is removed from the top of the InAs QD is not completely removed from the sample. Rather, the arsenic is moved around with a high degree of reincorporation into the InP capping layer. This redistribution and reincorporation is most likely the mechanism that prohibits the observation peaks in the PL spectrum associated with families of QDs with integer numbers of monolayers that was reported by Sakuma, *et al.*[17] At this time, it is not clear if the lack of ML QD families is a result of incomplete optimization of the MBE recipe, or if the kinetics of MOCVD produce a superior environment for removing the top of the QD during the double capping procedure.

Images of defect structures are shown in figures 5 and 6. Figure 5 shows a V-shaped growth defect that originates inside some QDs. V-shaped defects are dislocations that separate into two partial dislocations that glide on opposite {111} planes. Figure 6 shows a stacking fault that originates in the InP near a QD and extends above the QD.

FIG. 5. Scanning transmission electron micrograph of a quantum dot expressing a V-shaped defect that originates inside the InAs quantum dot.

FIG. 6. Scanning transmission electron micrograph of a stacking fault in the vicinity of a quantum dot.

V. DISCUSSION

The experiments that quantified the growth window study and enabled the phase diagram of the surface reconstruction and QD formation temperatures were motivated by the desire to modify the shape of the InAs QD with the goal of improving the brightness of the larger QDs. Ultimately, the QDs appeared to form similar shapes with statistics that did not significantly vary from previous reports. From the perspective of growth science, it is interesting to note that the surface reconstruction occurs simultaneously with the formation of the QDs over a wide range of As-overpressure.

Both defects that have been observed in these samples, V-shaped defects and stacking faults, are associated with increased rates of non-radiative recombination. The extended V-shaped defect has been characterized using deep level transient spectroscopy in ripened InAs QDs grown on GaAs and shown to form a mid-gap state with an activation energy of 0.52 eV.[14] Other structural studies of InAs QDs on GaAs also associated larger QDs with these defect structures, even though high QD densities and non-atomic resolution electron microscopy obscured the exact origins of these defects in previous studies.[15,18]

The atomic resolution imaging provided by this study shows a correlation between the specific origin of the defect and the nature of the defect. As illustrated by the two exemplar defects in Figure 5 and 6, the V-shaped defects are observed to originate from within the InAs QD, while the stacking faults originate near the QD in the InP capping material. This observation and interpretation is consistent with the relative stacking fault energy of the materials where InAs has a stacking fault energy of 30 mJm^{-2} that is higher than that of InP (18.83 mJm^{-2}).[19] Higher stacking fault energy materials tend to relieve strain by dislocation formation and slip, while lower stacking fault energy materials tend to form stacking faults. Interestingly, the relative difference in stacking fault energy is greater in the InAs/GaAs system (GaAs stacking fault energy is 45 mJm^{-2})[19] suggesting that V-shaped defects form in the GaAs near the InAs QDs, while stacking faults form in the QD. The observation that these extended defects form both inside and near the InAs QDs suggest that their formation may not only be introduced during the QD forming stage, but may form during the growth of the capping structure.

The observation of these defects in the atomic resolution images (that are know to form more readily in highly strained materials) infer that the larger QDs are more likely to contain these defects than smaller QDs. Additionally, quantum confinement of carriers make the smaller QDs emit at shorter wavelengths. It is reasonable to connect the two statements and conclude that the smaller QDs that have a shorter emission wavelength are less likely to express these defects and emit brightly, while the larger QDs with a larger degree of strain are more likely to have these defects and not emit.

The solution to minimizing the formation of these extended defects may be to anneal the structures for a longer period of time after the InAs QDs have had their tops removed. With the observation that some defects form outside of the QD, it is reasonable to consider the effect of confining strain formed when the capping layer is grown. That is, larger QDs may be defect free if an alloyed cap is used that has a lattice constant slightly larger than InP, thereby reducing the stress boundary in the area around the InAs QD. A final alternative may be to abandon the use of SK growth and adopt a strain-free substrate-encoded size-reducing epitaxy growth approach.[20]

VI. CONCLUSIONS

Exploring the substrate temperature and arsenic overpressure dependencies of InAs quantum dot growth on InP with a modified Stranski-Krastanov growth technique are unsuccessful in manipulating quantum dot shape and long wavelength efficiency. Atomically resolved high resolution electron microscopy identified extended defects in InAs quantum dots that express reduced emission efficiency at long wavelengths. The observation that these extended defects form both inside and near the InAs dots suggest that the capping layer plays a critical role in defect formation.

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