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OMVPE of InAs quantum dots on an InGaP surface

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ABSTRACT

The organometallic vapor phase epitaxy of InAs quantum dots has been investigated by comparing the effect the underlying surface has on the quantum dot physical characteristics. Atomic force microscopy measurements were used to identify the InAs QDs coalesce to significantly larger size when deposited on an InGaP surface compared to a GaAs surface. Quantitative assessment of the total QD volume on different surfaces such as GaAs, InGaP, and GaAsP implicates the role of indium in the underlying surface for the increase in QD size on InGaP surfaces.

1. Introduction

The most common QD/matrix material combination investigated to date is the InAs/GaAs system. The 7.8% compressive strain between InAs and GaAs enables QD formation in the Stranski–Krastanow (S–K) mode wherein a thin wetting layer forms two-dimensionally, prior to formation of three-dimensional, coherent islands. Control of the size of InAs QDs can allow 3D confinement of carriers resulting in discrete densities of states. The InAs/GaAs QD system has been studied for varied applications including telecommunication wavelength lasers on GaAs substrates [1,2] and extended spectrum solar cells [3,4]. QDs have also recently been explored for potentially highly efficient PV concept known as intermediate band solar cell (IBSC) [5], which has the potential to exceed 40% efficiency under appropriate conditions.

A main requirement for the IBSC is a matrix bandgap of 1.93 eV [5]. GaAs is therefore unsuitable as a host material in terms of demonstrating the maximum potential of the IBSC. $In_{0.48}Ga_{0.51}P$ lattice matched to GaAs (subsequently referred to as InGaP) has a bandgap as high as 1.91 eV and therefore is a better candidate for investigation of QD physics with respect to IBSC.

A wide range of approaches has been investigated to control the QD size and optical properties within the InAs/GaAs system. Considerable control of QD morphology and properties has been achieved using approaches including epitaxy condition modification [6–8], InGaAs interlayers or dots-in-a-well [9–11], and the use of Sb-containing materials [12,13]. An additional means to control QD morphology is the use of substrate misorientation. Vicinal substrates are commonly used in the manufacturing of III–V solar cells as a means to control the morphology and ordering of the top cell InGaP [14]. The use of vicinal substrates has been shown to be successful in controlling the morphology of InAs QD on GaAs surfaces [15,16], and therefore, is expected to play a significant role in modifying the morphology of InAs QD on an InGaP surface.

InAs QDs within an InGaP matrix are an important materials' system to investigate as an alternative means to adjust the QD properties. A higher band offset between the QD and matrix material is expected to minimize thermal escape from QD [17,18]. InAs QDs grown in an Al_xGaAs matrix exhibit an expected shift in emission wavelength as a function of the bandgap of the matrix material [19]. In addition, the higher bandgap barrier successfully reduced the thermal escape at 300 K. However, AlGaAs suffers from sensitivity to oxygen incorporation especially at lower growth temperatures typical of QDs. Elevated temperatures generally required for AlGaAs tend to degrade QD performance. In contrast, the aluminum-free material

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InGaP can be grown at lower temperatures and exhibits less oxygen incorporation than AlGaAs particularly at the lower growth temperatures commonly used with QD active regions.

An earlier report studied the molecular beam epitaxy (MBE) growth of InAs QD on an InGaP surface [20]. This study highlighted a dramatic change in the InAs nucleation and growth properties when deposited on InGaP surfaces. In particular, the critical thickness required for InAs QD nucleation decreased considerably when deposited on an InGaP surface. The use of a GaAs interlayer was also successful at modifying the subsequent InAs QD properties [20,21].

In this paper we study the organometallic vapor phase epitaxy (OMVPE) of InAs QDs on GaAs and InGaP surfaces. The OMVPE process is exclusively used for commercial production of III–V solar cells and therefore is an appropriate platform to perform QD research. Successful demonstration of QD-enhanced solar cells on a commercial OMVPE reactor, such as used here, makes any transition into production straightforward.

We find that, consistent with previous reports using MBE, the physical properties of InAs QDs are dramatically different when deposited directly on an InGaP surface. In particular, the diameter and height are significantly larger on an InGaP surface compared to a GaAs surface. Comparing to other reports on various surfaces, it is concluded that the indium component at the surface is responsible for the increased size of QDs on the InGaP surface. The change in QD epitaxy kinetics must be accounted for when optimizing the growth conditions on an InGaP surface.

2. Experimental

The epitaxial structure was grown by OMVPE on a 2 in. diameter n-type GaAs substrate with a misorientation of 2° toward (1–10). The OMVPE reactor is a 3×2 in. Veeco D125 using standard methyl-based organometallic sources together with arsine and phosphine. The undoped epitaxial structure consists of a 200 nm GaAs buffer layer grown at 620 °C followed by a 100 nm InGaP layer grown at 590 °C. The growth was interrupted for 300 s under PH_3 overpressure to reduce and stabilize the temperature prior to QD growth. Self-assembled InAs QDs were grown directly on the InGaP surface at 485 °C using a V/III of 17. Following a 60-s H_2-only growth interrupt, the sample was cooled to room temperature. The InAs QDs were uncapped to facilitate atomic force microscopy (AFM) measurements. The InAs deposition time was varied to control the InAs nominal thickness between 1.1 and 1.8ML. The InAs growth temperature was varied between 465 and 485 °C.

The surface QDs were characterized using a Veeco D3100 AFM in the tapping mode. $1 \times 1\ \mu m^2$ scans were taken at the center point of a 2-in. diameter wafer. QD size, density and height statistics were obtained using an imaging processing software, SPIP (Image Metrology) and these characteristics were examined as a function of the epitaxy conditions. Histograms of the QD physical characteristics were obtained using a fixed bin size of 0.3 nm for height data and 1.0 nm for diameter data. Additional AFM scans along the wafer diameter were also obtained to provide a rough, but useful, indication of the overall uniformity of the QD epitaxy process. The data shown here correspond to the center point of the 2-in. diameter wafer, but the trends discussed are identical regardless of wafer position.

3. Results

Fig. 1 shows AFM images from the center of each wafer for four different samples A–D. Table 1 shows the experimental conditions for each sample as well as the quantitative size characteristics for each image. The sample designation (A–D) corresponds to Fig. 1(a–d). Table 1 indicates the average values for the density, diameter, and height for each AFM scan in Fig. 1(a–d). Sample A uses conditions for our standard InAs/GaAs strain balanced structure that routinely provides increased short-circuit current in QD-GaAs solar cells [22] including enhanced overall AM0 efficiency [23]. Sample A, therefore, represents the QD size targets for efficient photovoltaic performance.

Fig. 1(a) exhibits a QD density of $4.82 \times 10^{10}\ cm^{-2}$ with QDs aligned with the step edges. InAs QD on GaAs surfaces generally exhibit a monomodal diameter distribution with larger, coalesced QDs eliminated through epitaxy condition optimization [23]. Fig. 1(b–d) shows AFM images of InAs QDs grown on an InGaP surface and illustrates a marked difference between QD epitaxy on an InGaP surface compared to the GaAs surface. The overall size of the InAs QDs is significantly larger when grown on the InGaP surface as indicated by Fig. 1(b–d).

Fig. 1(b) (Sample B) simply duplicates the same QD epitaxy conditions as Fig. 1(a) but deposits the QD on an InGaP surface. Fig. 1(b) shows a significant increase in the average diameter, in addition to significant coalescence compared to QDs grown on the GaAs surface.

Fig. 2a and b shows the height and diameter histograms for samples A and B respectively. Both histograms for sample A illustrate a narrow, single-mode distribution in height and diameter for optimized QD epitaxy on a GaAs surface. The histograms for sample B exhibit significantly larger mean values and wider distributions for both diameter and height. The mean and standard deviations (values in parenthesis) are calculated and tabulated in Table 1.

The QD diameter increases from 15.1 nm on a GaAs surface (Fig. 1a) to mean diameter of 34.1 nm on InGaP including considerable coalescence. The height also increases from 2.3 nm on GaAs to 5.8 nm on InGaP with heights as large as 20 nm on the InGaP surface.

In general a bi-modal distribution and coalescence is observed on a GaAs surface when excess InAs is deposited. Reducing the QD deposition time in sample D (Fig. 1d) by 40% is expected to reduce the overall amount of InAs available for QD formation and therefore reduce the QD size. Fig. 1d and Table 1 illustrate the expected reduction, although the diameter remains considerably larger than the GaAs surface in sample A. Coalescence remains significant, with mean diameter of 18.8 nm, but