Self-assembled In$_{0.5}$Ga$_{0.5}$As quantum dots on GaP

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We demonstrate the growth and luminescence of coherently strained In$_{0.5}$Ga$_{0.5}$As self-assembled quantum dots on GaP. Cross-sectional and planar-view transmission electron microscopy confirmed the dislocation-free nature of the In$_{0.5}$Ga$_{0.5}$As quantum dots and GaP cap layers. Intense photoluminescence from the quantum dots was measured at 80 K and was visible to the unaided eye in ambient lighting. The photoluminescence results show that emission energy can be controlled by varying the In$_{0.5}$Ga$_{0.5}$As deposition thickness. In combination with recent advances in the growth of GaP on Si, the In$_{0.5}$Ga$_{0.5}$As quantum dots demonstrated here could enable monolithic optoelectronic integration on Si. © 2010 American Institute of Physics. [doi:10.1063/1.3522647]

The monolithic integration of III–V optoelectronics with Si is critical for applications such as photonic integrated circuits and optical interconnects. Recently, several groups have reported growth of GaP on Si with minimal antiphase disorder and low densities of extended defects. However, GaP itself is not useful for most photonic applications due to its indirect bandgap. Several methods for the epitaxial growth of direct-bandgap material on GaP have therefore been investigated. One approach takes advantage of the band-bowing effect in the dilute nitride material GaNAsP. The ability to tune the lattice constant and bandgap in this quaternary alloy system permits the pseudomorphic growth of direct-bandgap Ga(NAsP) quantum wells (QWs) on GaP. Lasing under pulsed mode current injection at ~940 nm from Ga(NAsP) QWs has been demonstrated from 80 to 278 K. However, the relatively high threshold currents of 1.5 and 42 kA/cm$^2$ at 80 and 278 K, respectively, indicate that nonradiative recombination may still be significant in these metastable alloys.

A second approach toward realizing GaP-based light emission involves the use of self-assembled quantum dots (SAQDs). Several groups have investigated the growth of both InP and In-rich InGaP SAQDs on GaP. Most reported photoluminescence (PL) from such SAQDs with typical peak energies ($E_{\text{peak}}$) of 1.8–2.1 eV and full width at half maxima (FWHM) of 70–160 meV. Both Hatami et al. and Williams et al. reported room temperature operation of visible light emitting diodes (LEDs) using InP/GaP and InGaP/GaP SAQDs, respectively. However, no lasers based on such SAQDs have yet been demonstrated and the band offset between the dots and matrix has been suggested as the reason for this. Junno et al. theorized a type-II band offset with no electron confinement after failing to observe PL from InP/GaP SAQDs. Dewitz et al. proposed that the conduction band offset was likely to be type-I but with a very small barrier for electrons. The lack of electron confinement in InP/GaP SAQDs presents a fundamental obstacle to the attainment of high-efficiency laser diodes.

In addition, several studies of InAs SAQDs growth on GaP have been reported. Leon et al. demonstrated InAs SAQDs on GaP with a broad size distribution and corresponding broad PL emission at 77 K ($E_{\text{peak}}=1.7$ eV, FWHM ~215 meV). In contrast, neither Junno et al. nor Guo et al. were able to observe PL from InAs/GaP SAQDs. At ~10%, the lattice mismatch of InAs on GaP is significantly greater than the 4%–7% mismatch for InAs grown on either InP or GaAs, and Guo et al. attributed the absence of luminescence to nonradiative recombination due to dislocations. Tanaka and co-workers used cross-sectional transmission electron microscopy (XVTEM) to show that InAs dots grown on GaP by organometallic vapor phase epitaxy were heavily relaxed by misfit dislocations and stacking faults.

In$_{x}$Ga$_{1-x}$As/GaP SAQDs could offer several advantages over both InP/GaP and InAs/GaP SAQDs. First, the bandgap of bulk In$_{x}$Ga$_{1-x}$As is lower than that of bulk InP for $x > 0.05$, meaning that the confinement of one or both carriers in In$_{x}$Ga$_{1-x}$As/GaP SAQDs may be stronger than in InP/GaP SAQDs. Second, unlike InAs, the lattice mismatch of In$_{x}$Ga$_{1-x}$As on GaP can be tuned to values of 4%–7% for $x = 0.07–0.50$, allowing a wider window for dislocation-free three-dimensional (3D) growth and greater emission tunability.

In this letter, we report on the growth, structure, and luminescence properties of In$_{0.5}$Ga$_{0.5}$As SAQDs on GaP. In situ observations by reflection high-energy electron diffraction (RHEED) indicated that the In$_{0.5}$Ga$_{0.5}$As SAQDs grow by the Stranski–Krat assessing (SK) mode with a wetting layer critical thickness of ~1.9 monolayers (ML). Using XVTEM, we found the In$_{0.5}$Ga$_{0.5}$As dots and GaP capping layer to be free of dislocations and stacking faults. Visible PL at 80 K was measured for all samples and $E_{\text{peak}}$ could be tuned from 1.88–2.00 eV (660–620 nm) by varying the In$_{0.5}$Ga$_{0.5}$As deposition thickness from 2.0 to 11.0 ML. In addition to the goal of optoelectronic integration with Si, the reddish-orange luminescence from the In$_{0.5}$Ga$_{0.5}$As/GaP SAQDs described here may prove to be useful for epitaxial, transparent-substrate LEDs.

Growth was carried out using solid source molecular beam epitaxy on undoped, on-axis GaP(001) substrates. To enable efficient sample heating, we indium-mounted the GaP samples on Si handle wafers prior to growth. The structural properties of the buried In$_{0.5}$Ga$_{0.5}$As SAQDs were determined by XVTEM and plan-view TEM (PVTEM). We prepared samples for TEM using standard mechanical polishing.

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and Ar-ion milling techniques. For TEM analyses we used a Tecnai F20 operated at 200 keV. For PL measurements, samples were cooled to 80 K and optically pumped with a 100 mW, 3.07 eV (404 nm) diode laser, while spectra were collected by an Ocean Optics USB2000 spectrometer.

After oxide desorption at 650 °C, all growths started with an undoped 150 nm GaP buffer at 590 °C using a P$_2$/Ga beam equivalent pressure (BEP) ratio of ~0.5 and a growth rate of 0.4 ML/s. During growth of the GaP buffer, the RHEED pattern showed a streaky (2 × 4) reconstruction [Fig. 1(a)], indicating a smooth two-dimensional (2D) growth mode. Ex situ atomic force microscopy of GaP buffers confirmed a smooth morphology and yielded a root mean square roughness of 0.85 nm over a 10 × 10 μm$^2$ area. Various thicknesses of In$_{0.5}$Ga$_{0.5}$As were then deposited at 490 °C, with an As$_2$/Ga(Bin) BEP ratio of ~40 and a growth rate of 0.2 ML/s. In Ga and Ga beam fluxes were calibrated by a combination of RHEED intensity oscillations and high-resolution x-ray diffractometry. We chose an In$_{1−x}$Ga$_x$As composition of x = 0.5 to mimic the lattice mismatch of ~7% for the well-established growth of InAs SAQDs on GaAs. Significant interdiffusion on both the cation and anion sublattices is likely, altering the composition of the SAQDs to In$_{0.5}$Ga$_{0.5}$As$_{1−x}$P$_x$ with x < 0.5 and y > 0; references to the In$_{0.5}$Ga$_{0.5}$As SAQDs in this work are to the nominal, as opposed to the actual, composition. After initiating In$_{0.5}$Ga$_{0.5}$As growth, the RHEED pattern remained streaky for deposition thicknesses <1.9 ML, indicating planar growth. As In$_{0.5}$Ga$_{0.5}$As thickness was increased further, the RHEED pattern became spotty [Fig. 1(b)], corresponding to a transition from 2D to 3D growth. This change in growth mode indicates that In$_{0.5}$Ga$_{0.5}$As/GaP SAQDs form via the SK growth mode. PVTEM images shown three (2 × 4) bright-field XVTEM image in Fig. 1(d) shows three SAQDs buried beneath the GaP cap. Their strain contrast matches that typically seen in InAs/GaAs SAQDs under similar imaging conditions. Several other two-beam conditions were used to confirm that both the SAQDs and the GaP cap layer were free from dislocations. A two times magnified portion of the main figure appears as an inset to Fig. 1(d). It shows the presence of an In$_{0.5}$Ga$_{0.5}$As wetting layer and confirms the SK growth mode. PVTEM images (not shown) showed no Moiré fringes within the dots, further verifying their dislocation-free nature. From PVTEM, we also estimated a dot density of 1.5 × 10$^{10}$ cm$^{-2}$, with an average diameter of 45 nm and a variance of 11 nm.

Figure 2 compares the 80 K PL spectra from an epitaxial GaP buffer and a sample containing 5.0 ML In$_{0.5}$Ga$_{0.5}$As SAQDs, both grown on pieces from the same starting wafer. The epitaxial GaP buffer layer showed only weak defect luminescence with several broad, overlapping peaks. In contrast, the PL spectrum from the In$_{0.5}$Ga$_{0.5}$As SAQD sample exhibited a single peak centered at 1.94 eV (640 nm) with FWHM ~115 meV (38 nm). The PL integrated intensity from the SAQD sample was more than an order of magnitude greater than that from the GaP buffer and could be clearly seen through laser goggles in room light. We speculate that the radiative recombination process in the SAQDs is significantly faster than other defect-mediated radiative recombination occurring either in the substrate or the epitaxial GaP layers.

Comparing the emission energy of 1.94 eV with the 0.84 eV bandgap of bulk In$_{0.5}$Ga$_{0.5}$As at 80 K (Ref. 21) reveals an energy shift of ~1.1 eV that results from a combination of quantum confinement, compressive strain, and interdiffusion. In contrast, the energy shifts in In$_{0.5}$Ga$_{0.5}$As/GaAs SAQDs were previously reported to be ~0.4 eV. The higher energy gain in our dots likely results from the interplay of higher compressive strain and diffusion of P into the nominal In$_{0.5}$Ga$_{0.5}$As/GaP SAQDs, both of which would raise the en-

![Image](https://via.placeholder.com/150)
energy of the expected emission peak. InAs/GaAs SAQDs, grown at a nominally identical mismatch strain to our In$_{0.5}$Ga$_{0.5}$As/GaP SAQDs demonstrated here could enable monolithic optoelectronic integration on Si.

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FIG. 3. (Color online) (a) PL spectra at 80 K from buried SAQDs for various In$_{0.5}$Ga$_{0.5}$As deposition thicknesses. (b) E$_{\text{peak}}$ and FWHM as a function of In$_{0.5}$Ga$_{0.5}$As coverage. E$_{\text{peak}}$ moves to lower energy with greater In$_{0.5}$Ga$_{0.5}$As deposition thickness. The extracted FWHM for all peaks is in the range of 115–125 meV. Nonuniformity in SAQD size is likely to be the major contributing factor to FWHM.