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Spatially resolved In and As distributions in InGaAs/GaP and InGaAs/GaAs quantum dot systems

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InGaAs/GaP QD self-assembled quantum dot (QD) systems are of tremendous interest, for GaP can be directly grown on Si, enabling true monolithic integration of optoelectronics with Si technology [1]. Promising optical properties such as light emitting diodes (LEDs) at room temperature [2] and lasing at 80 K [3] have been demonstrated in these systems. It is critical that we develop a detailed microscopic understanding of the QD structures and elemental distributions of InGaAs/GaP QD systems because the band structure and band alignment, and thus the optical properties, depend on the detailed atomic structure of the QDs and spatial distributions of each element (In, Ga, As) [4-6], particularly In [7].

So far, two-beam TEM imaging has been the popular choice to image these QDs (Figure 1). In addition, the In distribution in these QDs has been inferred from lattice constant changes or diffraction contrast in TEM [8] or STM [9]. These indirect measurements can be potentially inaccurate due to factors such as strain relaxations in thin TEM samples and other elements (As, Ga, or P) affecting the lattice constant. Low-loss electron energy loss spectroscopy (EELS) was used to report the elemental distribution [10], but careful post subtraction of the zero loss peak is necessary. Therefore, direct measurements of In, Ga, and As in InGaAs/GaAs and InGaAs/GaP QD systems are highly desired.

We directly measure two-dimensional (2D) inhomogeneous distributions of In and As in InGaAs QDs grown in the GaAs and GaP matrix, using energy-dispersive X-ray spectral imaging in a scanning transmission electron microscope (EDX-STEM). The 2D chemical maps were acquired within several minutes, made possible by the combination of the high brightness electron source and high-solid-angle area, quadrant EDX detector, dramatically increasing the signal to noise ratio of the EDX maps.

Our main finding is that, in InGaAs/GaP QD systems, the In elemental distribution does not closely follow the QD structures while the As profile does (Figure 2). In contrast, for InGaAs/GaAs QD systems, In is concentrated within the QDs and along the interfacial wetting layer (Figure 2). Comparing the InGaAs/GaAs (Figure 2j-m) and InGaAs/GaP (Figure 2c-g) QD systems, In is more broadly distributed in the GaP matrix. Our findings show that the In elemental profile is much more complicated than previously assumed and that replacing the GaAs matrix with GaP for monolithic integration with Si results in different elemental profiles of InGaAs QDs. These findings challenge our current understanding of the band structure model of InGaAs/GaP QD systems [4, 6]. The detailed analysis of elemental distributions of InGaAs QDs on GaAs and GaP gives us a better understanding of the atomic structure of the InGaAs QDs, which can lead to improved QD structures and better optical properties by optimizing the growth parameters.

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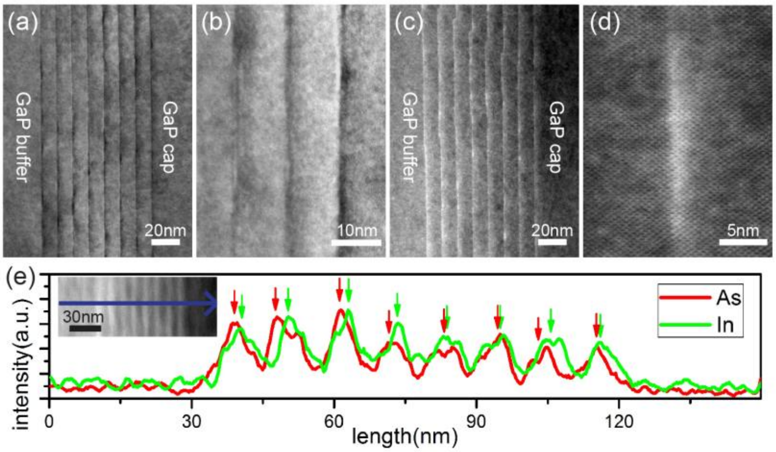

Figure 1. Two-beam (002) and HAADF-STEM imaging of 8 stacks of InGaAs/GaP QDs grown on GaP/Si. (a) and (b) show two-beam (002) TEM images where the dark regions point to QD locations due to local strain. (c) and (d) show HAADF-STEM images of the same sample. (e) shows elemental line profiles of As and In across the 8 stacks of QD layers.

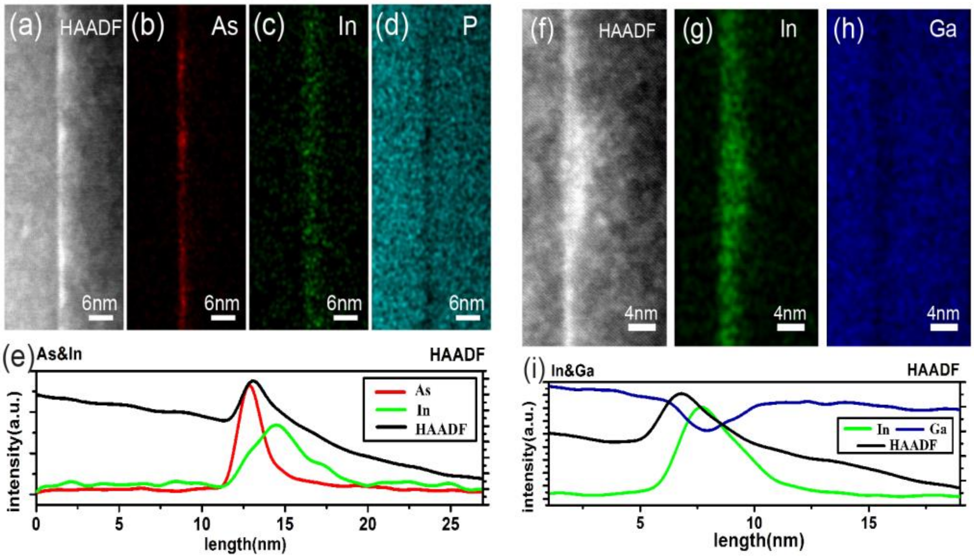

Figure 2. HAADF images and chemical maps of In and As in InGaAs/GaP(a-e) and GaAs(f-i) QD samples. From the maps, it is clear that the As distribution follows the formation of QDs while In is more broadly distributed. (e) and (i) show line profiles of the HAADF intensity, As, and In, obtained from averaging the 2D maps vertically.