Towards measuring bandgap inhomogeneities in InAs/GaAs quantum dots

S Kadkhodazadeh¹, M J Ashwin², T S Jones² and D W McComb¹

¹Department of Materials and London Centre for Nanotechnology, Imperial College London, UK, SW7 2AZ
²Department of Chemistry, University of Warwick, Coventry, UK, CV4 7AL

d.mccomb@imperial.ac.uk

Abstract. In this paper we report initial results from structural and optical investigations on bilayers of InAs/GaAs quantum dots (QDs) grown by molecular beam epitaxy with different GaAs spacer layer thicknesses. We have used bright field imaging in transmission electron microscopy and high angle annular dark-field imaging in scanning transmission electron microscopy to study strain and compositional correlations in these QD structures. Our goal is to utilise valence electron energy-loss spectroscopy (VEELS) to probe the bandgap and optical properties of the quantum dots. We discuss the influence of Čerenkov losses on VEELS and approaches to mediating their effect on bandgap measurements.

1. Introduction
Multilayer InAs/GaAs quantum dots (QDs) are promising materials for achieving the technologically important 1.3 – 1.5 µm emission wavelength in optoelectronics and data communication applications. Optical properties of these QDs are determined by their chemical and structural properties such as shape, size, composition and strain. Investigating these properties has been the focus of many theoretical and experimental studies [1-4]. Surface techniques such as atomic force microscopy (AFM), scanning tunnelling microscopy (STM) and reflection high energy electron diffraction (RHEED) have been used extensively to characterise InAs/GaAs QDs but these techniques cannot be used to investigate the dots after they have been capped. Transmission electron microscopy (TEM) can provide structural and compositional information on both capped and uncapped QDs. However, interpretation of on-zone bright field (BF) TEM images of such structures is complicated by the contrast arising from the strain fields around the QDs, due to the ~7 % lattice mismatch between InAs and GaAs. This means that properties such as size and shape are difficult to determine unambiguously from BF TEM images [5]. In this work we have used high-angle annular dark field (HAADF) imaging in a scanning transmission electron microscope (STEM) to obtain information on the structural properties of bilayer InAs/GaAs QDs. Our goal is to perform electron energy-loss spectroscopy (EELS) in our monochromated STEM to gain insight into the spatial homogeneity of the electronic and optical properties of the QDs such as the bandgap energy and complex dielectric constant. We discuss complications caused to this analysis by relativistic effects and how they can be mediated.

2. Experimental details
The samples were grown on epi-ready GaAs(001) substrates using a Gen II molecular beam epitaxy (MBE) system. After growing a GaAs buffer layer on the substrate at 580 °C, 2.5 ML of InAs was
deposited at 500 ºC followed by a GaAs spacer layer. A second InAs layer was then deposited under the same conditions and the structure was capped with 100 nm of GaAs. The temperature was kept constant throughout the growth of the active layers. Four identical samples were grown, but with different GaAs spacer layer thicknesses of \( S = 60 \) nm, 20 nm, 10 nm and 5 nm.

TEM samples were prepared by mechanical grinding and polishing followed by dimple-grinding and argon ion milling in a Gatan precision ion polishing system (PIPS) at 4.0 keV (reduced to 2.5 keV for final stage of milling). The samples were examined using a 200 kV LaB\(_6\) filament JEOL FX2010 microscope and a 300 kV STEM (FEI Titan 80-300) fitted with a monochromated field emission gun (FEG).

3. Results and discussion

3.1. Structural properties

Producing uniformly sized InAs/GaAs QDs is of utmost importance for most device fabrication purposes. It has been reported that in multilayer structures the interaction between the strain fields around the dots can provide a mechanism for controlling the size and position of the dots in subsequent layers [6-8]. Vertical stacking of the islands often has to be utilised in order to achieve high number density of the dots for optimal performance in optical devices. One parameter that can be used to tune this strain-induced vertical correlation between islands in multilayer structures is the GaAs spacer layer thickness, \( S \).

Fig 1 BF cross-sectional [110] TEM images of bilayer InAs/GaAs QD samples with \( S = \) (a) 60 nm, (b) 20 nm, (c) 10 nm and (d) 5 nm.

Figures 1 (a) – (d) show [110] BF TEM images of cross-sections of bilayers of InAs/GaAs QDs with \( S \) varying between 60 and 5 nm. Our observations are in general agreement with previous reports in the literature [1, 6, 8]. In the 60 nm sample, (a), the dots formed in the first and second layer do not exhibit any vertical spatial correlation, \( i.e. \) the dots in the second layer grow independently of the dots
in the first layer. In the 10 nm and 5 nm samples, (c) and (d), the second layer dots are formed directly above the dots in the first layer and have similar dimensions. The dots in the 20 nm sample, (b), on the other hand exhibit a partial correlation, i.e. some dots in the second layer are aligned with dots in the first layer and some are not. Given that the origin of vertical correlation is the strain field induced in the GaAs spacer layer by the underlying QD, the partial correlation observed in sample (b) suggests that all of the dots in the first layer do not have the same magnitude of strain field around them. This non-uniformity of the strain fields around the first layer dots could be caused by non-uniformity in either their size or their composition, or both. Further investigation on the size and composition of the first layer dots needs to be carried out to determine this.

As indicated earlier the presence of a strain field around the QDs can have a large influence on the contrast observed in the BF on-zone or two beam TEM images. This means that determining the size of highly strained dots using these imaging techniques can result in a significant overestimate of the actual size. In HAADF imaging in STEM it is possible to select electrons scattered to large angles and the resulting contrast is no longer strongly influenced by the strain field around the QDs. Figure 2 (a) is a BF TEM image of the \( S = 20 \) nm sample taken along the [110] zone-axis and (b) is a STEM image of the same region of the sample. The numbers on the images indicate the same QDs in each case. The dots appear to be lens-shaped and the length of the base is similar in both the TEM and STEM images (\( \sim 25 \) nm). However, the height of the QDs appears to be significantly greater in TEM image (\( \sim 8\) nm) than in the STEM image (\( \sim 5 \) nm).

![Fig 2 (a): BF [110] zone-axis TEM and (b) STEM HAADF image of bilayer InAs/GaAs quantum dots with \( S = 20 \) nm. The numbers (1-4) in the images correspond to the same features.](image)

3.2. Bandgap measurement in EELS

Determining the bandgap and other optical properties of semiconductors using EELS has been a popular topic since the development of monochromated TEMs because unlike other techniques, EELS allows measuring such properties with a high spatial resolution [9-11]. In principle, the first rise in an EEL spectrum corresponds to the bandgap and it can be determined simply by subtracting the zero-loss peak from the spectrum. However, in semiconducting materials with high refractive indices such as GaAs, the speed of electrons exceeds the speed of light in the material and this causes electrons to lose energy by emitting Čerenkov radiation. Čerenkov losses complicate measurements by contributing to the EELS signal at energies below the bandgap. The influence of relativistic losses on VEELS and experimental methods of mediating them have been discussed in detail in the literature [10-11]. The energy-loss probability of fast electrons penetrating a thin foil at a normal incident angle
can be modelled using the formula derived by Kröger [12]. Figure 3 (a) and (b) show the effect of sample thickness and accelerating voltage on the energy-loss spectrum of GaAs. As it is shown in fig 3 reducing the accelerating voltage of the electron source in TEM or specimen thickness reduce but do not eliminate relativistic effects in GaAs.

One approach of suppressing Čerenkov losses is to take advantage of their small scattering angle. Čerenkov radiation has a very small scattering angle (<0.01 mrad) associated with them compared with typical collection angles used in EELS (~ 10 mrad). This means that under typical experimental conditions in EELS, Čerenkov radiation is independent of the collection angle used. Therefore, two EEL spectra of the same intensity but collected with different collection angles, β₁ and β₂ (~ 10 mrad) can be subtracted from each other to remove the Čerenkov contributions. This method is discussed in detail in [11].

Fig 3: Simulated VEEL spectra of GaAs using Kröger’s equation [12] showing contributions from Čerenkov and surface losses to the spectrum for different accelerating voltage (a) and specimen thickness (b).

4. Conclusions
In conclusion, we have investigated structural properties of InAs/GaAs quantum dots using imaging in TEM and HAADF in STEM. Size and vertical position of the dots in multilayers can be manipulated by taking advantage of strain-induced nucleation in the top layer dots. This was observed in bilayer quantum dots with a GaAs spacer layer thickness less than 20 nm. We also introduced bandgap mapping using VEELS and complications associated with GaAs caused by relativistic effects and presented preliminary modelled data based on the Kröger equation.

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