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D.M. SCHAADT✉
S. KRAUSS
R. KOCH
K.H. PLOOG

# Stress evolution during growth of bilayer self-assembled InAs/GaAs quantum dots

Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5–7, 10117 Berlin, Germany

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**ABSTRACT** We investigated the stress evolution during molecular-beam epitaxy of bilayer InAs/GaAs(001) quantum dot (QD) structures in real time and with sub-monolayer precision using an in-situ cantilever beam setup. During growth of the InAs at 470 °C a stress of 5.1 GPa develops in the wetting layer, in good agreement with the theoretical misfit stress. At a critical thickness of 1.5 monolayers the strain is relieved by the QD formation. In the case of InAs/GaAs bilayer structures, the second InAs layer grows identical to the first for GaAs spacer thicknesses exceeding $\sim 13$ nm. For thinner spacers the critical thickness for the 2D/3D transition in the second layer decreases. The stress of the second InAs layer does not reach the value of the first, indicating that InAs QDs grow on partially strained areas due to the strain field of the previous InAs layer.

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## 1 Introduction

Multilayer structures of self-assembled InAs/GaAs quantum dots (QDs) separated by GaAs spacer layers have attracted considerable research interest due to the prospect of fabricating improved optoelectronic devices. If the spacer layers are thin enough, InAs/GaAs QDs align vertically [1]; their size distribution is more uniform leading to a decreased emission line width and an increased emission intensity [2]. Various studies have indirectly shown that the vertical alignment of QDs in subsequent InAs/GaAs layers is the result of the strain field of the respective QD layer underneath [1, 3–6]. From in-situ measurements of the strain during growth, Fuster et al. [7] concluded that the formation of quantum wires in the InAs/InP system proceeds via stress-driven processes. In the present study, we employ a UHV cantilever beam technique to directly measure the stress evolution during QD growth. Our experimental setup is capable of measuring the stress in situ and in real time with a precision in the sub-monolayer range. During growth of the InAs wetting layer the observed stress is in good agreement with the theoretical misfit stress; strain is relieved when the QDs nucleate. The stress in the second QD layer and the critical thickness for QD formation decrease as the GaAs spacer thickness is reduced. Then, the QDs preferentially grow on partially strained areas due to the strain field of the previous InAs layer.

## 2 Experimental details

The experiments were performed in a III–V semiconductor molecular-beam epitaxy chamber with a base pressure of $1 \times 10^{-10}$ mbar. It is equipped with a cantilever beam device using GaAs(001) substrates with dimensions of $25 \times 5 \times 0.15\,\mathrm{mm}^3$. The substrates are clamped only on one side, thus allowing the free end to bend. The deflection is determined in real time during film deposition by a capacitance technique combined with lock-in-assisted signal detection. Using a modified form of Stoney's equation [8], the film force, defined as the force per film width, is calculated from the substrate bending. Note that the slope in a plot of the film force vs. film thickness corresponds to the instantaneous film stress. The QDs were prepared at 470 °C (thermocouple reading) on epi-ready GaAs(001) substrates, which were coated after oxide desorption at 605 °C with a 500-nm-thick GaAs buffer layer. X-ray diffraction was used to determine the GaAs and InAs growth rates ($\sim 0.1$ nm/s and $\sim 0.01$ nm/s, respectively) by depositing five layers of $\mathrm{In}_{0.13}\mathrm{Ga}_{0.87}\mathrm{As}/\mathrm{GaAs}$ at 470 °C and fitting the resulting (004) X-ray peak to a simulated spectrum [9]. For atomic force microscopy, 2.5-monolayer (ML)-thick uncapped films were deposited; for photoluminescence investigations the QDs were capped at 470 °C by a 10-nm-thick GaAs layer.

## 3 Results and discussion

Figure 1a shows an atomic force microscopy (AFM) image of an uncapped InAs QD film. The QDs are clearly visible as bright spots in the AFM image. The average QD height is 10 nm and the base width is 75 nm, respectively; the QD density is $4 \times 10^9\,\mathrm{cm}^{-2}$. As reported in the literature and corroborated by our own measurements, QD size and shape change during overgrowth [10]. Photoluminescence spectra of a GaAs capped film show two peaks at $E_{\mathrm{h1}} = 1.184 \pm 0.001\,\mathrm{eV}$ and

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✉ Fax: +49-30-20377-201, E-mail: dschaadt@pdi-berlin.de

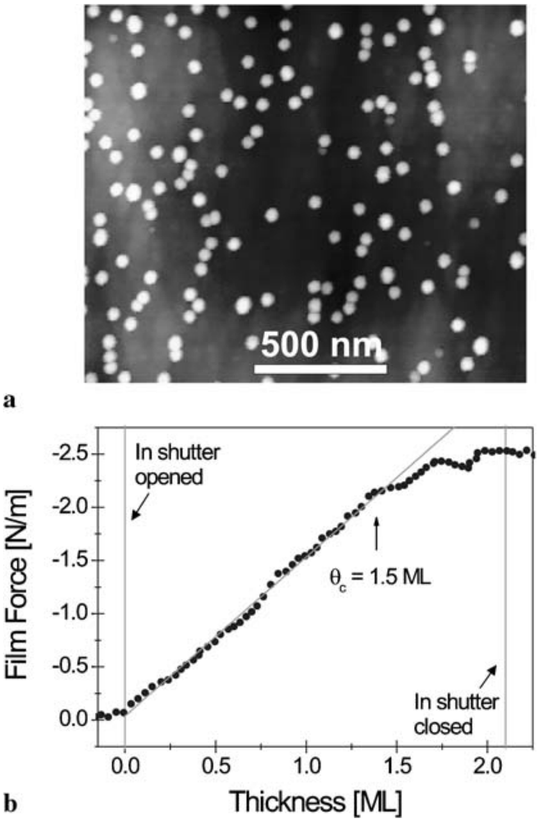

**FIGURE 1** (**a**) AFM image of an uncapped 2.3-ML-thick InAs film grown on GaAs(001) at 470 °C showing QD formation. (**b**) Respective film forces measured in real time as a function of the InAs thickness; the linear slope of the force curve up to the critical thickness $\theta_c$ of 1.5 ML corresponds to a stress of 5.1 GPa, in good agreement with the theoretical misfit stress

$E_{h2} = 1.228 \pm 0.002$ eV, which correspond to the ground state and the first excited hole state, both in reasonable agreement with values reported for samples prepared under similar conditions (see e.g. [11]).

Figure 1b displays the film force measured during growth of the film depicted in the AFM image of Fig. 1. Upon opening of the In shutter, a linear increase in film force is observed up to a thickness of 1.5 ML. During further InAs deposition, the slope decreases to zero. AFM images reveal that no InAs dots are formed at InAs thicknesses below 1.5 ML, thus suggesting that the thickness at which the slope decreases for the first time indicates the critical thickness $\theta_c$ at which the QDs nucleate. The observed $\theta_c = 1.5$ ML indeed is in very good agreement with the values reported in the literature for QD growth at 470 °C (see e.g. [12]).

Quantitatively, our experiments yield a force value of $2.3 \pm 0.1$ N/m at $\theta_c = 1.5$ ML, which corresponds to a stress of 5.1 GPa. Using linear elasticity, the misfit stress of an epitaxial InAs film is calculated by $\sigma_{\text{misfit}} = (c_{11} + c_{12} - 2c_{12}^2/c_{11})(a_{\text{InAs}} - a_{\text{GaAs}})/a_{\text{InAs}}$, where the $c_{ij}$ are the elastic constants, and $a_{\text{InAs}}$ and $a_{\text{GaAs}}$ are the lattice constants of InAs and GaAs, respectively. With $c_{11} = 83.3$ GPa, $c_{12} = 45.3$ GPa, $a_{\text{InAs}} = 0.606$ nm, and $a_{\text{GaAs}} = 0.565$ nm, the misfit stress at 470 °C is 5.3 GPa. The experimentally determined stress of the InAs wetting layer value therefore is identical – within experimental error ($< 10\%$) – with the theoretical misfit stress calculated with the elastic constants of the bulk.

This is indeed a surprising result as it seems to imply that the elastic properties of a monolayer are already bulk-like and that the influence of surface stress effects is negligible. It can be understood, however, by a more detailed look at the involved surface processes. When InAs is deposited onto the $c(2 \times 4)$ reconstructed surface of GaAs(001) the reconstruction is lifted, but an analogous structure is formed in the InAs wetting layer. Due to the close resemblance of the two semiconductor materials the surface stress of the two compounds probably is almost identical, so that no net surface stress contribution is obtained when the InAs wetting layer is deposited. An analogous argument holds for the elastic constants, in accordance with previous studies of Ge/Si(001) and Ge/Si(111) [13–15].

We remark that our experimental findings do not agree with recent stress measurements of InAs/GaAs(001) QDs by García et al. [12]. They determined a wetting layer stress of only 4.1 GPa, which is significantly smaller than both our value and the theoretical misfit stress, and suggested a model in which not all arriving In is incorporated into the InAs film but rather floats on the surface, thus not contributing to any stress build-up. We want to emphasize that we carefully calibrated our experimental setup with respect to thickness and stress measurements. The error of growth rates of InAs and GaAs obtained from fitting the experimental X-ray diffraction data to simulated spectra (see above) is better than 2%. To further corroborate this calibration we compared the measured film forces of the $In_{0.13}Ga_{0.87}As$/GaAs layer-by-layer film with the value theoretically expected from the misfit. Consistent with analogous experiments reported for $In_{0.2}Ga_{0.8}As$ [16], good agreement in the growth rate and stress is achieved. In addition, the cantilever beam setup was calibrated by magnetic measurements that quantitatively reproduce the expected magnetization of thick Fe and MnAs films [17]. In view of the correctness of our data, we speculate that the difference between our and García's results arises from the different growth conditions (higher growth rate, temperature and growth pauses).

In order to investigate the strain coupling between two InAs QD layers, we have grown bilayer structures consisting of two 2.5-ML-thick InAs layers separated by GaAs spacers with different thicknesses of 3.3, 6.6, and 13.3 nm. Due to the thermal inertia of cantilever beam devices, these experiments had to be performed at constant substrate temperature. Otherwise, thermal drift impairs the required resolution of the stress measurements. In addition, to avoid Ostwald ripening of the InAs dots and diffusion between InAs and GaAs layers, we did not perform any annealing at the growth or higher temperatures.

The film force measured during growth of a bilayer with a GaAs spacer layer thickness of 6.6 nm is shown in Fig. 2. During the growth of the GaAs

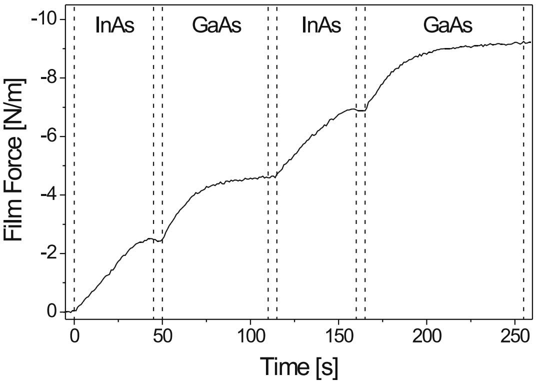

**FIGURE 2** Film force vs. time during the growth of a bilayer InAs/GaAs(001) film with a spacer layer thickness of 6.6 nm. Periods of growth interruptions (5 s) are indicated by *vertical lines*

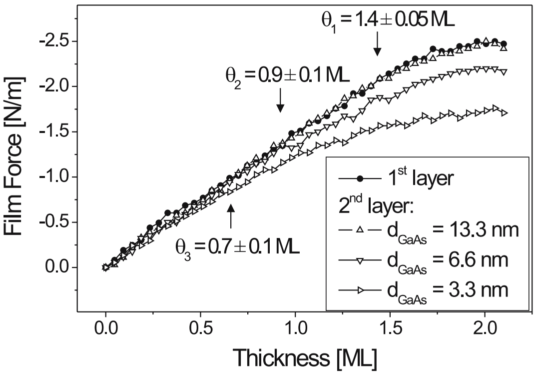

**FIGURE 3** Film force vs. time measured during InAs deposition for three GaAs spacer layer thicknesses. $\theta_1$, $\theta_2$, and $\theta_3$ indicate the critical thickness of the InAs films with 13.3-, 6.6-, and 3.3-nm-thick GaAs spacers

spacer layer, we observe additional build-up of stress indicating a straining of the QDs and the wetting layer. AFM reveals that GaAs accumulates around the QDs [10], which reverses their strain relaxation.

Figure 3 provides a comparison of the force curves measured during subsequent InAs deposition. The curves overlap for a GaAs spacer thicker than ~ 13 nm. As the spacer layer thickness is reduced, the film force during the growth of the second InAs layer does not reach the value of the first layer, indicating that the InAs dots grow on partially strained areas. In addition, the critical thickness for a 2D/3D transition in the second layer decreases with decreasing GaAs spacer thickness. Both results clearly demonstrate that the growth of the second InAs layer is dominated by the strain field of the previous InAs layer. Due to the chosen growth conditions, the surface of the GaAs spacer consists of mound-like structures, which can be understood by a step-edge barrier limited growth [18–20]. This probably explains why the strain coupling is only observed up to ~ 13 nm in our experiments, compared to ~ 40 nm for optimized spacer layer growth [1].

## 4 Conclusions

We have measured directly the stress evolution during molecular-beam epitaxy growth of bilayer InAs/GaAs(001) quantum dot structures. During growth of the InAs wetting layer the observed stress is in good agreement with the theoretical misfit stress; strain is relieved when the QDs nucleate. Film force curves measured during growth of bilayer structures with varying GaAs spacer layer thickness show that as the spacer layer thickness is reduced the film force during the growth of the second InAs layer does not reach the value of the first layer. The critical thickness for a 2D/3D transition in the second layer decreases with decreasing GaAs spacer thickness. This suggests that the growth of QDs in the second InAs layer is driven by a strain field due to the QDs in the first InAs layer.

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