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Interfaces correlation effect in 2D GaAs/AlAs heterostructures

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Abstract. A single 4.5-nm GaAs quantum well (QW) is grown into GaAs/AlAs superlattices (SL). For a sample with 120-s growth interruptions (GI) at the GaAs surfaces we have exciton linewidth the same as for best quality GaAs/AlAs thin QW with long time GI at bottom and top interfaces. For the sample with 120-s and 20-s GI at the GaAs and AlAs surfaces, respectively, the smallest linewidth (1.4 meV) occurs when the AlAs thickness is exactly an integer number of monolayers (ML). Then each AlAs surface almost reproduces the large-island GaAs surface just below it (interface correlation effect), providing a better AlAs surface on which the QW is grown.

The low-temperature exciton linewidth of GaAs/AlAs QW's is generally believed to be dominated by well-thickness fluctuations [1, 2, 6, 3]. The best QW grown with GI have macrorough interfaces with large lateral size and 1 ML height islands (holes). At the typical growth conditions, an inhomogeneous linewidth is dominated by bottom interface of the QW (AlAs surface). It has a smaller scale of macroroughness than top interface of GaAs QW because a surface diffusion length of Al much shorter than Ga. In this paper we show that due to shorter Al diffusion length it is possible for the surface of a thin AlAs layer is to reproduce the GaAs surface on which it was grown. Such interface correlations make the bottom interface of QW almost as the top one and decrease exciton linewidth.

Molecular beam epitaxy was used to grow the samples on (001) oriented substrates with miscut angle $< 0.1^{\circ}$. The samples were grown without substrate rotation resulting in large lateral gradients of the GaAs and AlAs layer thicknesses. Two samples NMSL15 and NMSL17 have the same structure and contain two single GaAs QW's. A control QW1 nominally of 25 ML GaAs with 35 ML AlAs barriers was grown without any GI. The second QW2 nominally of 16 ML GaAs was grown in the middle of a 80-period SL (8 ML GaAs, 6 ML AlAs). Both samples had 120-s GI's at GaAs surfaces of the SL and QW2. The sample NMSL15 had 20-s GI's also at AlAs surfaces of the SL and QW2.

The differential reflection (dR/dE)/R and photoluminescence (PL) excitonic spectra were taken at T = 10 K. GaAs layer thicknesses in QW2 (G_{QW2}) could be kept constant by scanning along a contour of constant QW1 exciton energy. Then the A_{SL} variation was defined by the SL period variation which is measured from the folded acoustic phonon Raman shifts, with an accuracy ± 0.05 ML.

In the NMSL17 sample grown without GI after AlAs (Fig. 1(a)), there are three sharp excitonic resonances in the reflection spectra (solid lines) with energy splittings approximately same as calculated for QW2 width differing by 1 ML (monolayer splitting). The linewidth (2 meV) to monolayer splitting (\approx 14 meV) ratio is \approx 0.15, the



Fig 1. The differential reflection (dR/dE)/R (solid lines) and the PL (dotted lines) exciton spectra of QW2 for the NMSL17 (a) and NMSL15 (b) as a function of AlAs layers thickness of SL (A_{SL}) at the constant thickness of QW2 (G_{QW2}).

same as for the best quality GaAs/AlAs QW's, grown with long GI's after GaAs and AlAs [2]. Hence, the lateral scale of macroroughness for both of the QW2 interfaces (top and bottom) is much larger than the exciton diameter (≈ 20 nm). It is very important to note that the excitonic energies and intensities are changed continuously with AlAs thickness and repeat when *A* changes by 1 ML (but the linewidths and multiplet weighted average are almost unchanged). Hence, the local thicknesses of QW2 in the NMSL17 is noninteger in general (but its difference is integer) and increasing when A_{SL} decrease. Measurements along other directions of the sample show that the relative intensities are periodically dependent upon $G_{QW2} + A_{SL}$ (this is the distance between top QW2 surface and GaAs surface just bellow QW2) with the period 1 ML.

The behavior of the NMSL15 sample grown with GI after AlAs, Fig. 1(b), is strikingly different. The excitonic resonance amplitude and width now depend strongly on the AlAs thickness. For noninteger A_{SL} the peaks are shifted to higher energies and broadened to 4–5 meV. Hence, at noninteger A_{SL} GI strongly decreases the lateral scale of macroroughness of the AlAs surfaces. When A_{SL} is an integer, the lines are quite narrow, 1.4 meV, with the record value of a linewidth to monolayer splitting ratio (≈ 0.1). Hence, for integer A_{SL} GI brought the improving of bottom interface of QW (evidently because its microroroughness decreased after GI). But even for integer A_{SL} the AlAs surface is not perfect after GI. The satellite peak just below the main peak for integer A_{SL} in Fig. 1(b) shows that array of a uniform small holes is presented on a large AlAs



Fig 2. Schematic illustration of the interfaces in GaAs/AlAs heterostructures. Gray, light gray and white colors specifies AlAs, GaAlAs and GaAs respectively. The interfaces tilt angle is 0.01° .

planes (this holes are $\approx 7 \,\text{nm}$ in size and occupy $\approx 10\%$ of the area by our estimates).

For discussion about the exciton confinement effect we have two different scales: exciton diameter $(D_{ex} \approx 20 \text{ nm})$ in the QW plane and monolayer height (1 ML = 0.283 nm) across this one. The key structural parameter is the surface diffusion length $L_{D,Ga}$ (or $L_{D,AI}$). Without GI diffusion length is much shorter than D_{ex} so surface is microrough and its local height z coincidences with its average height Z (the bottom interface on Fig. 2(a). After long time GI $L_{D,Ga} \gg D_{ex}$ and macroroughness takes place on the GaAs surface (the top interface on Fig. 2(a). Thus, the local height of the i-th surface can take one of the two values: $z_i = [Z_i]$ or $z_i = [Z_i] + 1$) with probabilities $1 - \{Z_i\}$ and $\{Z_i\}$ respectively, where rectangular and figure brackets denote integer and fractional parts of the average height of surface Z_i in ML. The value $\{Z_i\}$ determinate the filling factor of the i-th surface. For $\{Z_i\} = 0$ the surface is completed and clean. Islands are formed on the surface for $0 < \{Z_i\} < 0.5$, and holes for $0.5 < \{Z_i\} < 1$. A lateral size of islands (holes) depends on diffusion length and filling factor.

Now it is very important to notice that for a real (001) substrate an average interface height Z_i shifts up in the growth direction by a lot of atomic layers within the light beam spot. Even for a 0.01° error in locating the (001) plane result in ≈ 30 ML shift up in the average height of surface across the light spot diameter ≈ 50 mkm. Therefore all values for the local filling factor of the i-th surface can be assumed equally probable. So, to calculate the allowed local QW thicknesses and the areas they occupy, one must average over $0 < \{Z_i\} < 1$ and to take into account that the local filling factor of two uncorrelated macrorough surfaces are related by $\{Z_{i+1}\} = \{Z_i + T\}$, where T is the average distance between them. Result is dependent on interfaces correlation.

Four interface models with GI after GaAs surfaces are shown in Fig. 2. There is no GI after AlAs and no correlation with the previous interface in Fig. 2(a); therefore the slope of the microrough AlAs surface is that of the average GaAs surface. The macroroughness on the GaAs interface as shown on Fig. 2(a) give two QWs sets where the thicknesses change saw-like from $G_{QW} + 1$ to G_{QW} and from G_{QW} to $G_{QW} - 1$. This model without interface correlation predicts a continuous thickness distribution that does not agree with the discrete reflection spectra of Fig. 1(a).

In the Fig. 2(b) the microrough surface of the thin AlAs layer correlates with the previous GaAs surface. One can see that the QW local thicknesses (g_{QW}) are discrete but noninteger $(g_{QW} = [T] + \delta n - A_{SL})$ where $T = G_{QW} + A_{SL}$. For noninteger T the model gives four values g_{QW} ($\delta n = -1, 0, 1, 2$). For integer T there is only three g_{QW}

 $(\delta n = -1, 0, 1)$. Areas and hence excitonic oscillator strength for regions with different g_{QW} depend on the fractional part of *T*. Thus, the interface correlation effect explains the basic tendencies in the exciton spectra of NMSL17 (Fig. 1(a)), namely number of component (4 in sum at PL and (dR/dE)/R spectra for $\{T\} = 0.2 \div 0.5$), oscillator strength dependencies on the $G_{\text{QW}} + A_{\text{SL}}$ and continuous energy shift up for increasing A_{SL} .

A GI on the AlAs surface could destroy the correlation with the previous GaAs one if $L_{D,AI}$ is of the same order as $L_{D,Ga}$. If so, the QW local thicknesses depend only on QW average thickness ($g_{QW} = [G_{QW}] + \delta n$) but do not depend on AlAs thickness. Since this is not the case NMSL15 we conclude that $L_{D,AI} \ll L_{D,Ga}$, so there must be two lateral scales of fluctuations on the AlAs surface (Fig. 2(c) in general: the larger one repeats the large islands on the GaAs surface, the smaller consists of macrorough islands (holes) when A_{SL} is noninteger (then excess of Al must go somewhere but $L_{D,AI}$ is too small). For integer A_{SL} , large flat regions on the AlAs surfaces are complete, and each AlAs surface repeats the GaAs surface below (Fig. 2(d)). Interface correlation with exactly the right amount of Al narrows the exciton line width at the points with integer AlAs thickness on sample NMSL15.

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