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X-Ray Reflectivity Studies on Buried GaAs Quantum Dots: Non-Destructive Determination of Depth and Density

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GaAs quantum dots buried under capping GaAs and $Al_{0.3}Ga_{0.7}As$ layers have been evaluated by X-ray reflectivity. Some oscillating structures caused by interferences have been observed. Since the frequency corresponds to the layer thickness, Fourier analysis has been employed to determine the depth of the GaAs dot layer, which forms the sharp interface. It has been demonstrated that this technique is useful to check whether the dots are successfully prepared or not. Furthermore, the data provide information on the average deposition amount of GaAs over the whole sample as well as the composition fluctuation along the depth. It was found that dot density was determined by those values. The advantages of the non-destructive nature of the present method are discussed.

Key words: interface, multilayer, nano-structure, specular reflection, grazing-incidence

1. INTRODUCTION

Quantum dots (QDs) are microscopic metal or semiconductor boxes that hold a certain number of electrons and thus have a wide number of potential applications in developing optoelectronic devices, such as laser diodes [1]. They are grown on substrates or on deposited thin films as island-type nano-crystalline particles, of which the size and height are in the order of 3~30 nm. Demand has grown for the introduction of specific powerful non-destructive tools to explore the structures, because in many cases QDs are buried under the capping layer, thus making observation difficult by modern atomic-resolution microscopes like STM [2] and AFM [3].

It is well-known that X-rays are totally reflected on flat and smooth surfaces, since the refractive index for X-rays is slightly smaller than 1. Reflectivity measured as a function of the glancing angle shows a pattern strongly correlated to the electron density distribution along the depth, from nm to μm. Therefore, the technique has been used as a powerful method to study surface and buried interfaces of thin films [4]. In particular, when the sample is a layered material, interference fringes are observed in the profile. The analysis gives both layer thickness and information on the interface, i.e., grading, diffusion and roughness. The non-destructive nature of the technique is clearly extremely important for applications.

In this research, X-ray reflectivity was employed to evaluate GaAs quantum dots buried under the capping GaAs and $Al_{0.3}Ga_{0.7}As$ layers. The emphasis was placed on obtaining average information, such as the depth position, deposition amount, and dot density over the whole sample, rather than the properties of individual QDs.

2. DESCRIPTION OF ANALYTICAL PROCEDURES

X-ray reflectivity is theoretically sensitive to the change in electron density according to depth [5]. When the sample has interfaces, which generate gaps in the refractive index, interference fringes are observed due to multiple reflections. One can obtain the thickness of each layer in a multilayer by analyzing the frequency components of the fringes [6]. Figure 1 schematically shows the structure of the GaAs QD studied in the present research. Although there are many layers deposited on the GaAs substrate, the bottom $Ga_{0.7}Al_{0.3}As$ buffer is as thick as 500 nm, and can be

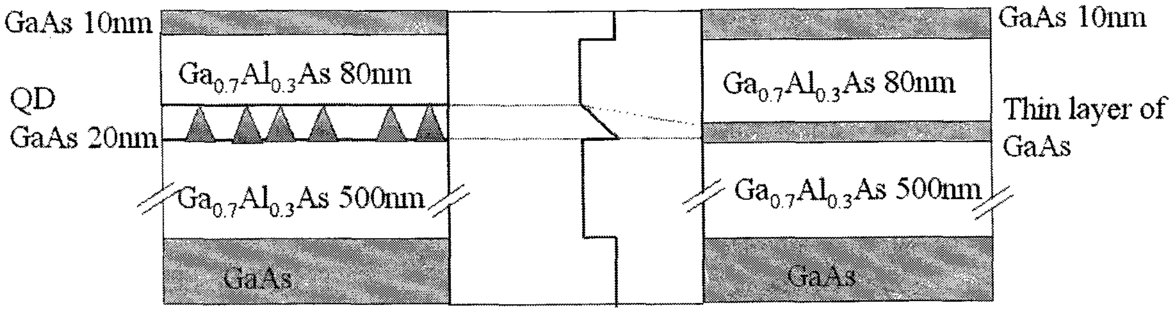

Fig. 1 Schematic diagram of real QD structure (left), depth profile of refractive indexes of ideal structure (middle), and assumed model for reflectivity calculation (right).

regarded as a bulk. The pyramidal QDs are grown on this buffer, and then those islands are covered with the capping layer with a total thickness of 90 nm. In Fig.1, the real part of the refractive index, which correlates to the electron density, is shown as a function of the depth. Although QDs are not uniform both along the depth and lateral directions, one should note that the bottom parts of QDs are essentially sharp interfaces, and the other parts give the gradation. Since X-ray reflectivity looks only at such a depth profile, a laterally uniform and graded depth layer can be used as an equivalent model for the analysis of a real QD layer.

Experimentally obtained X-ray reflectivity for QDs can be analyzed as follows: (i) the depth position of the QD layer is determined by Fourier analysis, and (ii) the total deposition amount of GaAs QDs and (iii) the gradation of the concentration ratio of GaAs and GaAlAs along the depth are evaluated by curve fitting. When the size and height of the pyramidal QDs are assumed as S and t, respectively, dot density N can be given as N=3d/St where d is the thickness corresponding to the total deposition amount of GaAs QDs and can be obtained by assuming a simple uniform GaAs layer. This is valid for quite small QDs, i.e., t and d should be the same order. For large QDs, it is crucial to consider the gradation. One can assume a linearly graded layer of a mixture of GaAs and $Ga_{0.7}Al_{0.3}As$. Then N=C/S where C is the relative concentration of GaAs at the bottom of the GaAs dot layer. Other parameters, such as the thickness and surface/interface roughness of each layer, are obtained by curve fitting.

3. RESULTS AND DISCUSSION

GaAs QD specimens as schematically shown in Fig.1 [7] were prepared using the liquid phase epitaxy method [8]. Prior to growing QDs, a wetting layer of around 1.75 ML of GaAs was deposited on the $Ga_{0.7}Al_{0.3}As$ buffer. The annealing temperature was ca. 600 °C. The size and height of QDs are quite large, at around 20nm. The size of the sample for X-ray reflectivity measurement is 10mm×15mm.

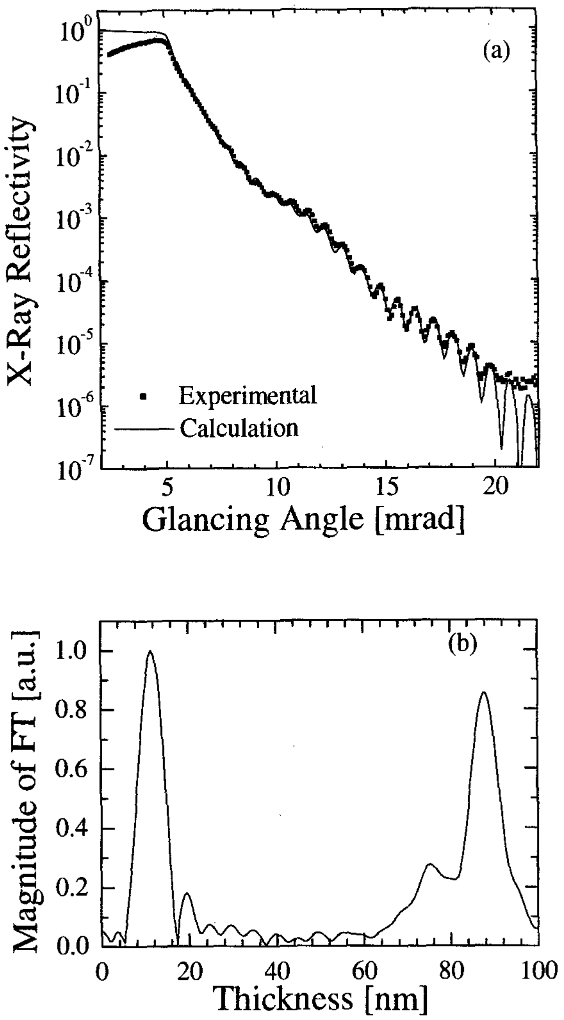

Fig.2 (a) Experimental (closed squares) and calculated (solid line) results of specular reflectivity. (b) Fourier transformation of (a) as a function of scattering vector $(q^2-q_c^2)^{1/2}$.

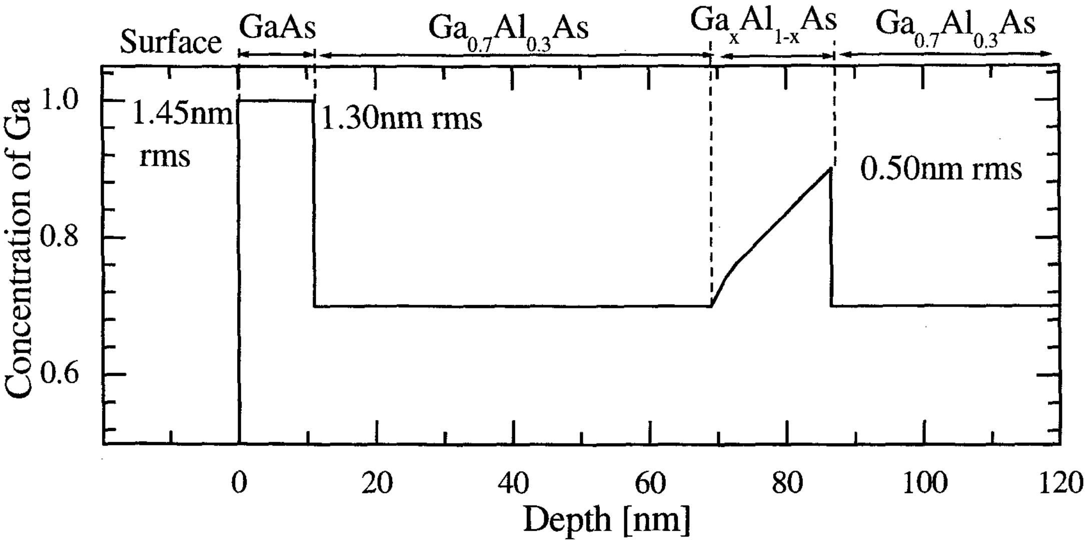

Fig.3 Composition of simulation model used in Fig. 2(a) is shown as a function of concentration of Ga. Layer including QD is shown as hatched part.

Figure 2 (a) shows the typical X-ray reflectivity for a CuKα_1 line, 0.154 nm measured by normal $\theta/2\theta$ scan. The critical angle is approximately 5.4 mrad, corresponding to the density of 5.32 g/cm^2, which agrees well with the bulk GaAs. Total reflection takes place below the critical angle and the experimental maximum was 66.3% (smaller than the theoretical value, ~96%), indicating that the sample surface could have a slightly curved shape. The data exhibit clearly visible interference oscillations, which include different frequency components, corresponding to the different layer thicknesses. As shown in Fig.2 (b), Fourier transform of the oscillating part [9] reveals that the pattern is derived from the top GaAs layer (12 nm), and the $Ga_{0.7}Al_{0.3}As$ capping layer plus GaAs QDs (total 78 nm). One should note that the bottom parts of the GaAs QDs form a sharp interface between the capping layer and beneath the buffer layer. The depth position of this interface is ca. 90 nm from the surface.

In order to investigate the influence of the total deposition amount of GaAs QDs, calculations were carried out. The model used for calculation is shown in Figure 3. Figure 4(a) shows reflectivity curves for a model that assumes a uniform GaAs layer of a different thickness. It was found that 0.6 nm can explain the experiments well. In the present case, the size of QDs is quite large, making it necessary to perform the calculation assuming the linear gradation of the mixture layer of GaAs and $Ga_{0.7}Al_{0.3}As$. Here, the total amount of GaAs is considered as equivalent to the 0.6 nm uniform layer. The relative concentration of GaAs at the bottom of the QD layer was determined as 0.66. When the QD size is around 20 nmx20nm, dot density can be evaluated as 1.6x10^{11} /cm^2. Other parameters obtained through curve fitting are summarized in Table I. The final fitted curve is shown in Fig.2(a).

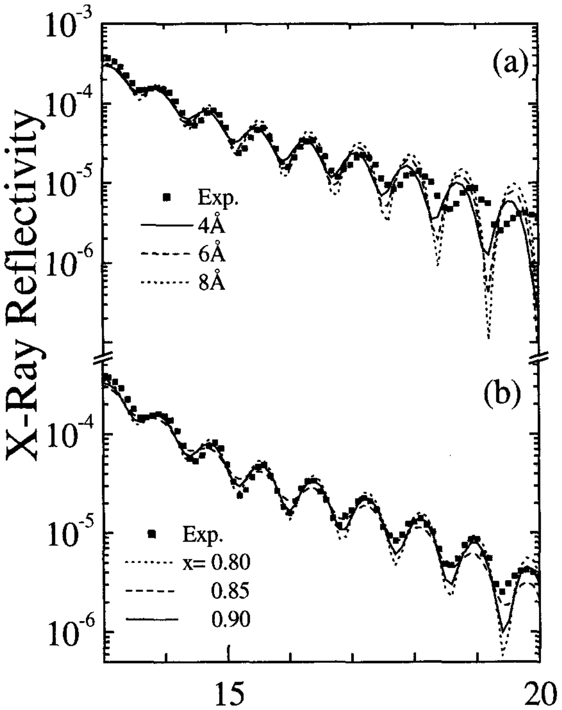

Fig.4 Estimation of thickness and grading of the QD layer. (a) thin-film models with thickness 0.4, 0.6, and 0.8 nm. (b) grading linearly from $Al_{0.3}Ga_{0.7}As$ to $Al_{1x-1}Ga_xAs$(x=0.90, 0.85 and 0.80).

Table I Estimation of nanostructure of the quantum dots. Aspect ratio is assumed as 1.

Total deposition amount	0.6 nm
Height	17.5 nm
Dot density	1.6x10^{11}/cm^2
Distance	22 nm

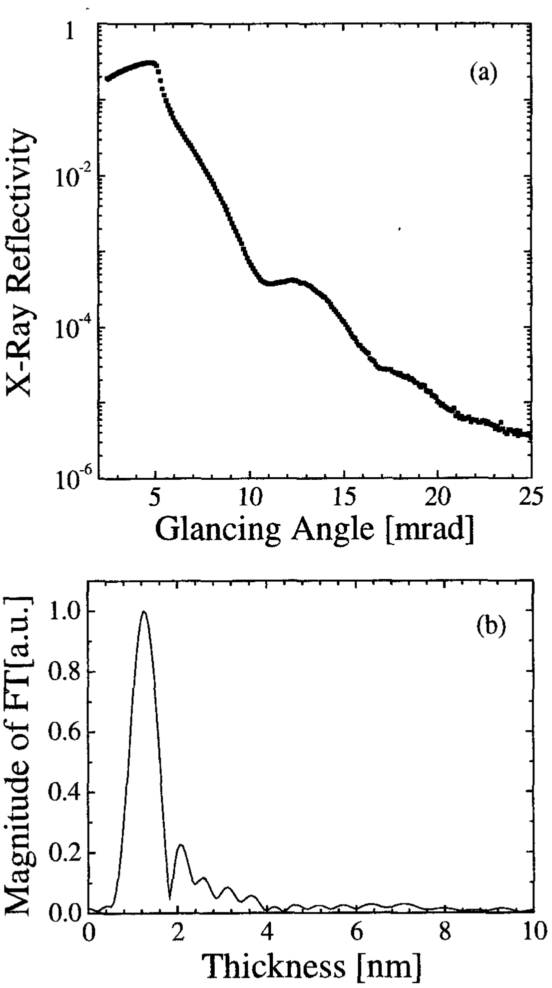

Fig.5 (a) Experimental results of specular reflectivity for another QD sample (wetting layer was not prepared in prior to QD growth). (b) Fourier transformation of (a) as a function of scattering vector $(q^2-q_c^2)^{1/2}$.

The technique can be employed simply to see whether the QD layer is formed successfully or not. Figure 5 (a) shows the X-ray reflectivity of the sample, the design structure of which is almost the same as shown in Fig.1. The only difference is that the wetting layer of GaAs has not been deposited before growing QDs. When compared with Fig.2(a), one can see the interference fringes are different. As shown in Fig.5(b), Fourier transform of the oscillating part has only one single peak corresponding to the top GaAs layer (11 nm). This suggests that the interface between the capping and buffer layers is not sharp, and that there are probably too few QDs to form the layer.

Generally, non-specular X-ray reflection, which is sometimes called diffuse scattering or GISAXS (grazing-incidence small angle X-ray scattering), can give information on the lateral inhomogeneous structure. When applied to QDs, the size, height and distance between dots can be analyzed [10,11]. In the present study, however, data acquisition was not successful, because the scattering X-ray intensity was too weak due to the fairly thick capping layer. In addition, quite high QDs cast shadows, resulting in a loss of information.

4. CONCLUSION

X-ray reflectivity was employed to evaluate the buried nanostructure of QDs. The depth position, the total amount of QDs, and the concentration gradation were determined successfully. The reflectivity includes the interference fringe corresponding to the thickness of the capping $Ga_{0.7}Al_{0.3}As$ plus GaAs QD layers. The technique is feasible for examining whether the formation of QDs is successful or not.

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