Reproducibility and Stability of X-ray Reflectivity Technique: GaAs/AlAs Multilayer Case

Mari Mizusawa and Kenji Sakurai

National Institute for Materials Science, Sengen, Tsukuba, Ibaraki 305-0047 Japan

Abstract. X-ray reflectometry is a highly reliable method for investigating layered materials along their depth non-destructively. This note examines how the analysis is reproducible and stable by comparing the data taken repeatedly, by two different machines, for two samples that have essentially the same structure.

INTRODUCTION

The X-ray reflectivity technique gives information on the density, thickness of each layer and roughness of the surface and each interface of thin films and multilayers [1,2]. As the method uses interference of X-rays inside the thin films, one can determine the layer thickness with a fairly high degree of precision. The technique has been routinely used even in industry for checking the thickness of specific layers in final products. However, since the measurements are performed at a very small angle, the experimental data are easily influenced by the alignment and other experimental conditions. Another issue is how the data agree each other when different types of instruments are employed for measuring the same sample. In addition, curve fitting, based on the least mean squares algorithm, does not always give true parameters. There should be any number of local minima, and generally, the solutions are not unique. Therefore, in a realistic analysis, one would encounter many factors that could mislead the analysis. The aim of the present research is to examine the level of reproducibility and stability of the X-ray reflectivity technique.

EXPERIMENTAL

The samples measured are GaAs/AlAs superlattices, where 3-layer pairs are MBE grown on the GaAs substrate. The materials are stable and have sharp interfaces. Even in the1970s, the structure was studied using the X-ray reflectivity technique [3]. It is also known that the thickness of the layers, except the top surface, is fairly stable in 1~3 years range, and can be used as a sort of standard reference materials [4]. The two samples, A and B, were cut from the same wafer, and therefore the layered structure is basically identical. The measurements were done by two different X-ray reflectometers, X and Y. Their main specifications are listed in Table 1. Both the samples A and B were measured by X-ray reflectometers X and Y. The measurements were repeated 5 times. Each time, the sample was removed once, then mounted again and the reflectometer was realigned. The angular range was not taken so widely this time, because the interest of the present research is placed on the thickness of each layer, rather than the details of the top surface (formation of thin oxide layer, density gradient etc), of which influences are enhanced in quite higher angle region. The higher end of the glancing angle was set as 30~40 mrad (ca. 0.24~0.33 $Å^{-1}$ in q_z). The measuring times for one reflectivity curve using reflectometers X and Y were 50 min and 40 min, respectively. The obtained X-ray intensity at each point was normalized by the direct beam intensity, which was measured when the sample was removed from each reflectometer.

TABLE 1 – Specification of reflectometers and measurement conditions		
	Reflectometer X	***Reflectometer Y***
X-Ray Source	Copper rotating anode, Small focus 0.03mm(H) × 3mm(V), 40kV-80mA, Bias –320V	
X-Ray Wavelength	Cu $K\alpha_1$ (Si(111) channel-cut)	Cu $K\alpha_1$+ $K\alpha_2$ (W/B_4C multilayer monochro)
Goniometer	NIMS original 1-axis precise goniometer for sample rotation and a translational stage for detector	Rigaku RINT ATX-G 2-axes goniometer
Sample Holding	Vacuum chuck	
Detector	YAP:Ce scintillation detector	NaI:Tl scintillation detector
Entrance Slit	0.05mm(H) × 2.5mm(V)	0.05mm(H) × 2mm(V)
Receiving Slit	0.1mm(H) × 10mm(V)	0.1mm(H)
Remarks	High resolution	High intensity Large angle range

DATA ANALYSIS

In the present case, a good layered model was already available from the beginning. Generally, however, one of the biggest problems for data analysis is finding a good model (i.e., How many layers? What is the approximate thickness? etc. But sometimes the sample is not an ideal multilayer.). The Fourier transform technique [5-7] is a simple and good way to build the first model, which can be used as a starting point. The wavelet transform technique [8-10] is another promising way to start the analysis without depending on a model. In the present study, after an initial look using the above mentioned methods, standard least-mean-square fitting using the Simplex method was carried out. Here, the model used is a 9 media multilayer including substrate and air. That is, the existence of an additional thin low-density top layer (around 10 Å) is assumed on 3 pairs of GaAs and AlAs layers that are deposited on a GaAs single crystal substrate. As the layer density and the interface roughness are correlated each other, in the present analysis, the density of each layer is fixed to make it easier to see the reproducibility and stability of the analysis. As the same measurements were repeated 5 times, the analysis was also done for those 5 data sets. Then the average and the standard deviation were calculated for each parameter. In addition, to avoid falling into the local minima during the fitting, dependence of the residual sum of squares was examined by slightly shifting values for each parameter.

RESULTS AND DISCUSSION

All raw experimental data are summarized in Fig.1. Note that the angular ranges are not the same, and the number of Bragg peaks observed is 4 and 5, for reflectometers X and Y, respectively. As will be shown later, however, the analysis gives very good agreement in the thickness of each layer. Figure 2 shows the typical results of Fourier transform. This clearly indicates that each layer has a very similar thickness at ca. 95 Å. One can see also that the total thickness is around 586.5 Å. In this way, initial values for the layered model were selected properly. Reproducibility of the reflectivity data can be confirmed by this simple procedure.

The parameters obtained by least-mean-square fitting are listed in Tables 2 and 3. As is clearly seen, the standard deviation is fairly small both for layer thickness (< 0.2~0.3Å in many cases) and for roughness (< 1Å). This means the reproducibility of the same measurement, using the same sample, and the same reflectometer with independent alignment, is very good. In addition, the variance for layer thickness caused by employing a different reflectometer is smaller than 0.8Å except in the case of the top layer (3~4Å) and the beneath AlAs layer (1.7Å) for Sample A, and the bottom AlAs layer (1.5Å) for Sample B. As the sensitivity to those layers is generally affected by the angle range of the measurement, the error could be due to this reason rather than to some other instrumental differences. The differences in layer thickness between Samples A and B were smaller than 0.9Å (reflectometer X)

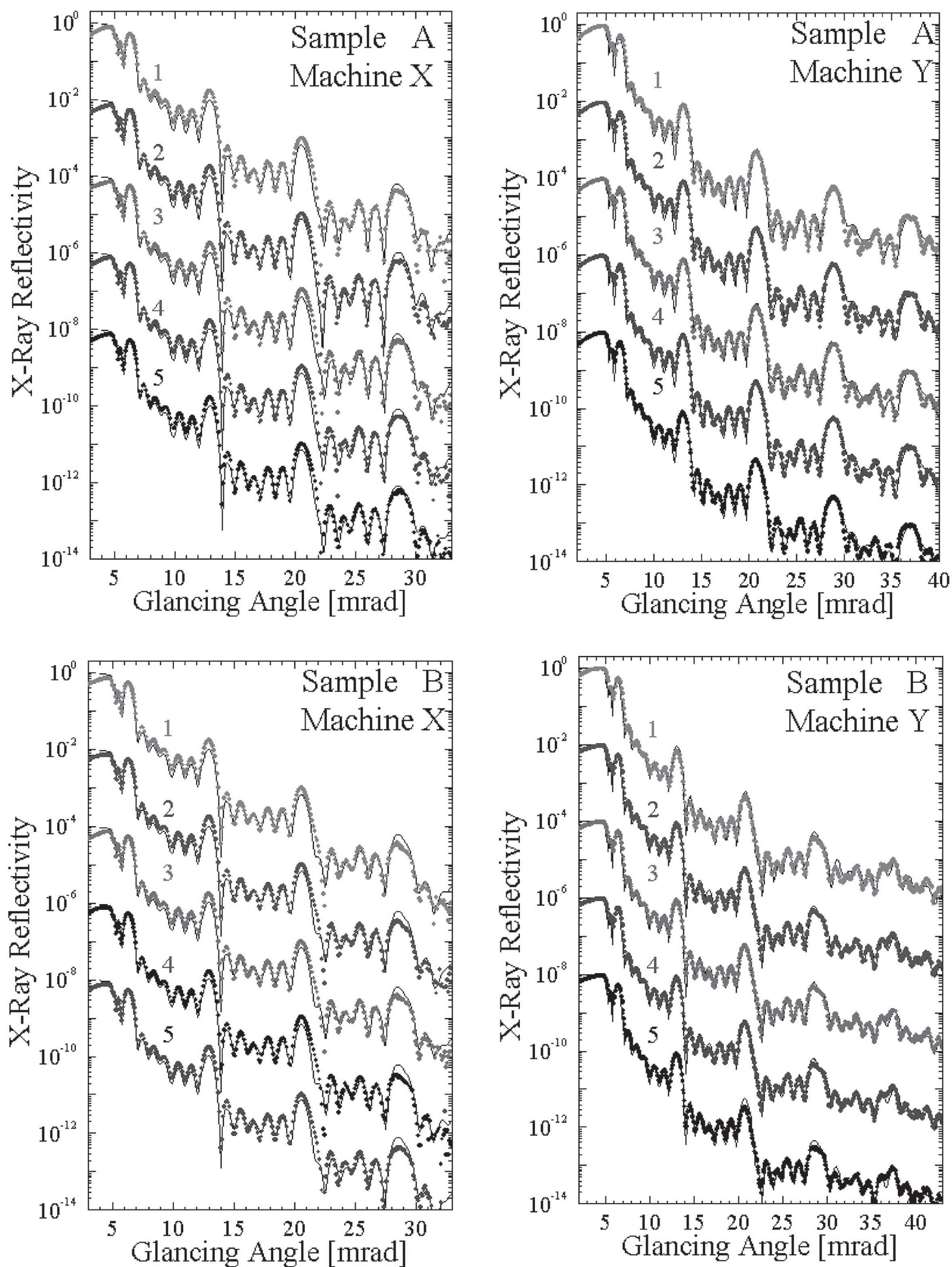

FIGURE 1. X-ray reflectivity data for two samples, A and B, both of which have basically the same structure, i.e., 3 layer pairs of GaAs and AlAs deposited on single crystal GaAs substrate. Two independent reflectometers X and Y are employed. The measurement was repeated 5 times. The sample was removed once and realigned for each measurement. Closed circle and solid line correspond to experimental and fitted data, respectively.

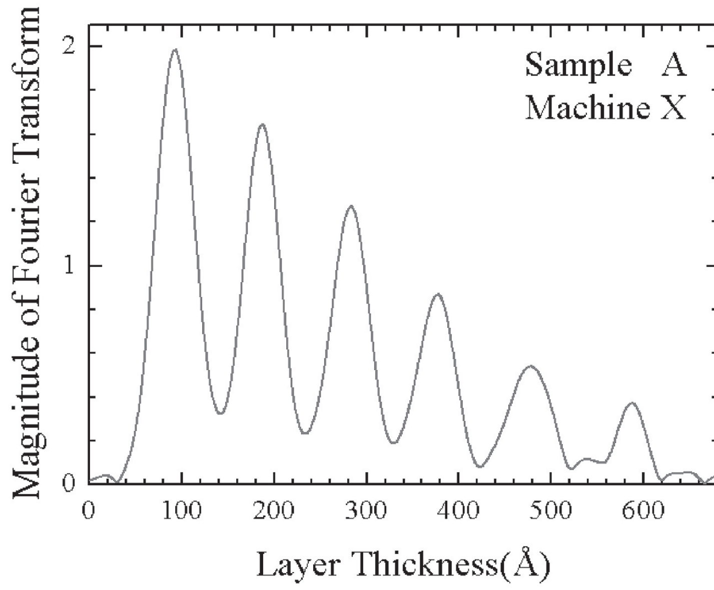

FIGURE 2. Fourier analysis of X-ray reflectivity data (Sample A, No.1 measured by reflectometer X).

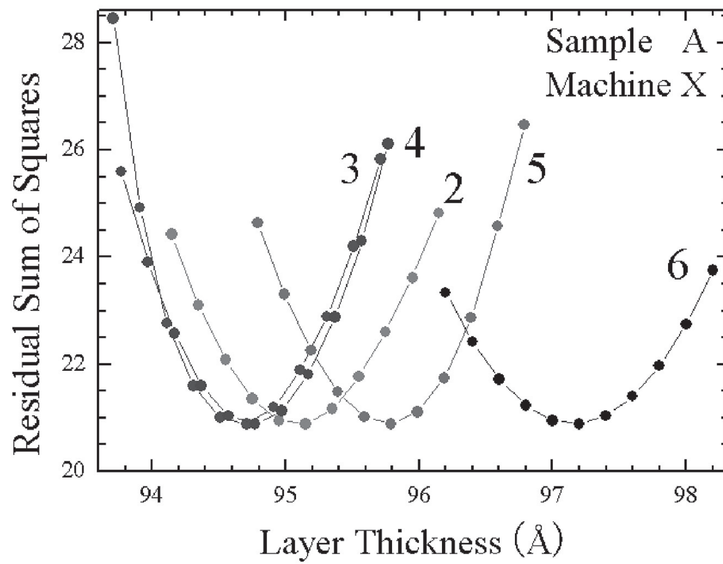

FIGURE 3. Sensitivity of least-mean-square fitting. 2, 3, 4, 5, and 6 corresponds to the layer of AlAs (beneath the top GaAs layer) , GaAs, AlAs, GaAs, and AlAs (just on the substrate), respectively. The data is for Sample A. No.1 measured by reflectometer X.

TABLE 2 – Summary of the determination of layer thickness

	Sample A		***Sample B***	
	X	***Y***	***X***	***Y***
GaAs with low-density top layer	***108.8 ± 0.39 (13.3+95.4)***	***104.6 ± 0.52 (15.2+89.4)***	***108.8 ± 0.17 (12.1+96.7)***	***105.6 ± 0.32 (14.8+90.8)***
AlAs	***94.9 ± 0.15***	***93.2 ± 0.25***	***94.2 ± 0.21***	***94.4 ± 0.27***
GaAs	***94.7 ± 0.07***	***94.3 ± 0.06***	***94.7 ± 0.18***	***94.1 ± 0.27***
AlAs	***95.0 ± 0.22***	***94.4 ± 0.14***	***95.2 ± 0.14***	***94.8 ± 0.50***
GaAs	***95.7 ± 0.18***	***95.5 ±0.08***	***95.2 ± 0.14***	***94.4 ± 0.32***
AlAs	***97.0 ± 0.19***	***96.2 ±0.08***	***96.1 ± 0.15***	***94.6 ± 0.52***

TABLE 3 – Summary of the determination of interface roughness				
	Sample A		*Sample B*	
	X	*Y*	*X*	*Y*
1 (Surface)	***5.7 ± 0.2***	***6.4 ± 0.2***	***5.1 ± 0.1***	***4.0 ± 0.3***
2	***6.6 ± 0.2***	***7.6 ± 0.3***	***5.3 ± 0.2***	***5.9 ± 0.2***
3	***3.2 ± 0.8***	***2.8 ± 0.2***	***3.7 ± 0.2***	***4.2 ± 0.1***
4	***4.0 ± 0.6***	***3.6 ± 0.1***	***4.7 ± 0.3***	***4.5 ± 0.2***
5	***2.7 ± 0.4***	***3.9 ±0.2***	***3.6 ± 0.4***	***4.1 ± 0.5***
6	***4.7 ± 0.3***	***5.5 ±0.1***	***5.0 ± 0.3***	***4.8 ± 0.3***
7	***3.6 ± 0.5***	***5.4 ±0.1***	***4.2 ± 0.3***	***4.4 ± 0.2***
8	***4.8 ± 0.4***	***7.0 ±0.2***	***5.6 ± 0.2***	***5.3 ± 0.1***

or 1.6Å (reflectometer Y).

During the fitting calculation, surface and interface roughness are likely to be forced to assume the role of correction factors, and therefore they are not as stable as layer thickness. However, the errors for the same sample with different reflectometers are not so large, i.e., within 1.1Å (Sample A) and 2.2Å (Sample B). Those for samples A and B are within 1.3Å (reflectometer X) or 2.4Å (reflectometer Y). In contrast, Figure 3 shows how each layer thickness is sensitive to the fitting process. This clearly demonstrates that the calculation can find each parameter with ±0.1Å precision. The R-factor is 5~8 % in the present case, and the fit is not still perfect. It has been found that further improved fit is possible by modifying the model, but the values for layer thickness do not change.

CONCLUSIONS

The reproducibility and stability of the X-ray reflectivity technique were examined by comparing the data taken 5 times repeatedly by two different machines, for two samples that have essentially the same structure. In addition to the 3 layer pairs of GaAs and AlAs, a low-density thin top layer has been assumed for the analysis. The results of independent measurement for the same sample, using the same reflectometer, are reproducible within 0.2~0.3Å for layer thickness and 1Å even for roughness. An error larger than 1Å was obtained for the thickness of specific layers when a different reflectometer was employed, but this could be due to differences in covering angle range rather than some other instrumental differences. As layer thickness is more stable than roughness, a potential problem is how to treat the top layer. The differences found between both samples, measured by the same reflectometer, are small for almost all layers, but some layers show a of difference up to 1.6Å. The present results show that thickness of each layer, except the top layer, can be well reproducibly determined with the measurements in reasonably limited angle range. On the other hand, it has been found that the fit can be further improved by considering some gradation of density, or some asymmetric roughness. Such parameters depend on the model to be considered and might not be stable, however, one should note that the technique can give other parameters, in particular, thickness of each layer with fairly good reproducibility and stability even when such effects are neglected.

ACKNOWLEDGMENTS

The authors would like to thank Dr T. Fujimoto (AIST, Japan) for providing GaAs/AlAs samples. Part of this research is connected to the VAMAS TW2/A10 project [11], conducted by Professors L Depero and E. Bontempi (Brescia University, Italy).

REFERENCES

1. J. Daillant and A. Gibaud Eds., “X-ray and Neutron Reflectivity: Principles and Applications”, Springer (1999).
2. K. Stoev and K. Sakurai, Spectrochim. Acta **B54,** 41 (1999).
3. L. L. Chang, A. Segmuller, L. Esaki; , Appl. Phys. Lett., **28,** 39 (1976).
4. I. Kojima, B. Li and T. Fujimoto, Journal of NIMC, **7,** 335 (1999). (in Japanese)
   http://www.aist.go.jp/NIMC/publication/journal99/7-6-3.html
5. K. Sakurai and A. Iida, Jpn. J. Appl. Phys. **31,** 113 (1992).
6. K. Sakurai and A. Iida, Adv. X-ray Anal. **35,** 813 (1992).
7. F.Bridou and B.Pardo, J. X-Ray Sci. Tech., **4.** 200 (1994).
8. E. Smiegel and A. Cornet, J. Phys. **D33,** 1757 (2000).
9. I.R. Prudnikov, R.J. Matyi and R.D. Deslattes, J. Appl. Phys. 90, 3338 (2001).
10. O.Starykov and K.Sakurai, Appl. Surf. Sci., **244,** 235 (2005).
11. See, http://dimgruppi.ing.unibs.it/chimica/VAMAS/vamas.htm