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ZnO thin film characterization by X-ray reflectivity optimization using genetic algorithm and Fourier transformation

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ABSTRACT

Zinc oxide (ZnO) thin film was fabricated by sol–gel spin coating method on glass substrate. X-ray reflectivity (XRR) and its optimization have been used for characterization and extracting physical parameters of the film. Genetic algorithm (GA) has been applied for this optimization process. The model independent information was needed to establish data analyzing process for X-ray reflectivity before optimization process. Independent information was exploited from Fourier transform of Fresnel reflectivity normalized X-ray reflectivity. This Fourier transformation (Auto Correlation Function) yields thickness of each coated layer on substrate. This information is a keynote for constructing optimization process. Specular X-ray reflectivity optimization yields structural parameters such as thickness, roughness of surface and interface and electron density profile of the film. Acceptable agreement exists between results obtained from Fourier transformation and X-ray reflectivity fitting.

1. Introduction

New technology amplifications in addition to new applications outlooks in optoelectronic and spintronic instruments motivated a supplementary research interest in ZnO studies. Due to the capacity usages as transparent and conducting electrode materials for flat panel displays electrodes [1], surface acoustic instruments [2], optical wave-guides [3] and gas sensors [4], the attention in zinc oxide (ZnO) thin films has newly increased.

Fabrication of zinc oxide thin film can be carried out with different techniques such as chemical vapor deposition [5], sputtering [6] and sol–gel process [7]. Among these techniques sol–gel technique contests with the others due to its low cost and this technique is a process well suited to large scale production [8].

There is a wide use of thin layered films with layer thickness in some nanometer range of thickness in modern technology and industry. Also manufacturing of high quality light emitting devices with longer operational lifetime needs smooth interface between hetero-junction structures. Controlling of structural parameters such as roughness at the surface and interface is essential in making high-performance devices, based on low-dimensional structures such as thin films and quantum wells. So analyzing the structure of the thin films is technologically very significant. Material characterization enables the improvement of new materials, structures and technologies. By developing instrumentation for improved measurement methods, there is an analogous need for the progress of techniques in order to operate these tools to their maximum benefits.

One of the most excellent techniques to study the structural and physical aspects of thin films is X-ray reflectivity (XRR). XRR is well established and nondestructive technique used for extraction density, thickness and roughness of surface and interface of thin film structures. In grazing incidence X-ray reflectivity (GIXR) technique, the X-ray beam is incident on the film at grazing angle and the interfered reflected beam is aggregated by X-ray detector. Presence of interfaces in the film causes interference process. The thickness of the layers causes interference periods and the amplitude of the interference oscillations depend on the both interfacial roughness and the electron density difference between the layers [9].

A typical way to characterize the structural parameters of a film from its X-ray reflectivity is to construct a model that we expect logically describes its structure and from which we can simulate X-ray reflectivity. By calculating the differences between the experimental and simulated curves, using a number of fitness functions, the model fitted by some optimization methods in order to minimize the difference between the two curves. This procedure is repeated until the difference between the two curves is arbitraged to be sufficiently small, at which point we believe that the model to be an accurate representation of the structure [10].

A disadvantage of the X-ray reflectivity fitting for exploiting structural parameters of thin films is that more than one electron

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density profile may be used to generate a single reflectivity result. Therefore we require model independent information about the system to estimate a close model of the system for correct analysis of the data.

Fourier transformation process of the X-ray reflectivity is the quick method to obtain useful model independent information concerning to the real space structure. Use of the Fourier transformation of X-ray reflectivity (Auto Correlation Function – ACF) can prepare useful layer thickness measurements for constructing fitting procedure. The combined use of Fourier analysis and fitting procedures would make X-ray reflectivity data more practicable for realistic analysis of thin films.

Durand [11] showed that the procedure based on fast Fourier transform (FFT), applied to X-ray reflectometery (XRR) and high resolution X-ray diffractometry (HRXRD) techniques, is a quick and powerful method for the evaluation of individual thicknesses inside complicated semiconductor heterostructures. He explained the ability of this method by reporting X-ray reflectometry study on superlattices, multiple quantum wells, and other complicated structures. Gerasimenko et al. [12] applied Fourier, Fractal and wavelet analysis for exploration of nanoscale $CoSi_2$ structures in Si fabricated by ion synthesis. The relationship between fractal dimensions and degree of surface cobalt disilicide ordering was established and the correlation between Fourier and fractal analysis was shown. Itoh and Yamauchi [13] characterized Surface morphology of pentacene thin films and their substrates with under-layers by applying fast Fourier transformation on the atomic force microscopy (AFM) data. The power values of power spectral density (PSD) for the AFM digital data were determined by the fast Fourier transform (FFT) algorithms instead of the root-mean-square (rms) and peak-to-valley value.

Fitting algorithm, which minimizes the discrepancy between theory and experiment, is the main part of iteration process. Classical gradient based optimization procedures show acceptable performance but remain unreliable due to trapping in local extreme. In contrast genetic algorithms (GAs) combine the advantages of stochastic search with intelligent strategy of solution finding. Application of GA in science and engineering has made this technique to be robust and effective [14].

Surface morphology and its acceptable characterization play an essential role in the study of thin films from different aspects. Generally each existing and future applications of thin films need specific optical, electrical, chemical and mechanical properties, which almost all strongly depend on the surface quality of the film. For this reason in this study Auto Correlation Function of Zinc Oxide thin film, obtained by calculating the Fourier transformation of the ratio of reflectivity data and Fresnel reflectivity, was applied for extracting layer thicknesses of zinc oxide thin film. Total thickness information of the film is the starting point for constructing fitting procedure between the experimental and theoretical X-ray reflectivity. Genetic algorithm was applied for optimization of the fitness function between logarithmic experimental and theoretical X-ray reflectivity. Structural parameters such as roughness of surface and interface, layer thickness and electron density profile were extracted by fitting procedure. The underlying motivation for the study is founded on increased interest in ZnO films due to their importance in a range of technological applications.

2. Experimental

2.1. Sample preparation

A zinc oxide thin film was fabricated by sol–gel spin coating method [15]. For each layer, the film was preheated at 275 °C for 10 min and annealed at 350 °C for 1 h. The deposition was repeated five times to obtain five-layered film of zinc oxide. X-ray reflectivity measurement was performed using Bede GXR1 reflectometer at Durham University, Physics Department. The specular reflectivity curve was recorded with $\theta - 2\theta$ scan.

2.2. X-ray reflectivity

X-ray reflectivity is a method used to characterize the surface structure of materials irrespective of their crystalline perfection. Hence this technique can be applied to crystalline, polycrystalline and amorphous materials. Application of this technique for thin films provides information about thickness, roughness and electron density in the film.

By impinging X-ray beam (I_0) with a grazing incident angle on the film, a reflectivity is specified as

$$|r_{12}|^2 = \frac{I}{I_0} \tag{1}$$

Here I_0 and I are incident and reflected X-ray intensities.

The recursive formula for reflectivity is [16]

$$r_{i,i+1} = \left[\frac{r_{i+1,i+2} + F_{i,i+1}}{r_{i+1,i+2} \times F_{i,i+1} + 1}\right] \times a_i^4 \tag{2}$$

where

$$F_{i,i+1} = \left[\frac{g_i - g_i}{g_i + g_{i+1}}\right] \times \exp\left(\frac{-8\pi g_i g_{i+1} {\sigma_{i+1}}^2}{\lambda^2}\right),$$

$$a_i = \exp\left(\frac{-i\pi g_i d_i}{\lambda}\right)$$

$$g_i = \sqrt{{n_i}^2 - \cos(\theta)^2} = \sqrt{(1 - \delta_i + i\beta_i)^2 - \cos\theta^2}$$

where θ, λ, d_j and σ_j are incident angle, X-ray wavelength, j-th layer thickness and surface roughness respectively.

The recursive equation was first obtained by Parrat for X-ray reflectivity simulation [16]. The roughness term was introduced in the framework of the Distorted Wave Born Approximation (DWBA) [17]. This expression indicates that the reflectivity profile will have series of minimum and maximum giving interface fringes, called Kiessig fringes, and the successive maxima in q-space ($q = 4\pi/\lambda \sin\theta$) is inversely related to the thickness of the film.

For exploiting structural parameters of film GA optimization was performed in order to minimize the fitness function. The selection of a suitable fitness function is crucial for data-fitting procedure independent of the optimization method used. A number of fitness functions can be assumed, but in the case where a measured and a calculated curve are compared, a fitness function consisting of the Root Mean Squared Error (RMSE) of measured and the calculated data has been observed to work well in practice [10]. We used the mean-squared error of the log transformed data as a fitness function

$$E = \frac{1}{N-1}\sum_{j=1}^{N}\left[\mathrm{Log}I_{\mathrm{exp},j} - \mathrm{Log}I_{cal,j}\right]^2 \tag{3}$$

where N is number of data points.

Before starting optimization, the overall thickness of the film was extracted by ACF of normalized X-ray reflectivity. For fitting the program with the experimental data, the zinc oxide thin film is considered to be made of a number of slabs of same thickness with varying electron density. Electron density in each slab and roughness of each interface are fitting parameters. After optimization process, position of peaks in electron density profile versus depth indicates thickness of individual layers in the film.