THE EVALUATION OF STRUCTURE PARAMETERS OF A Mo/Si SUPERLATTICE USING X-RAY SCATTERING DATA AND A GENETIC ALGORITHM

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ABSTRACT

Specular and nonspecular x-ray scattering are used to study the mesoscopic structure of interfaces of Mo/Si superlattices which have period of thirty or fewer. The sample was deposited on a Si wafer by ion beam sputtering. The data are explained quantitatively by using a distorted-wave Born approximation including correlating behaviour of interface roughness both in a lateral and vertical direction. A genetic algorithm technique was used to determine the structural parameters of the sample. The fundamental concepts of a genetic algorithm are explained. The method was found to be robust and flexible compared to conventional optimization techniques.

INTRODUCTION

The chemical abruptness and physical smoothness of the inner interfaces of Mo/Si superlattices (SLs) consisting of alternating Mo and Si layers play a key role in the performance of x-ray optical elements. The knowledge of the value and lateral character of interface roughness and its correlation, along with the compositional density profile of superlattices permits one to predict the reflective features and durability of multilayered x-ray mirrors, to control the growth process and improve preparation technology. The replication character of interfacial roughness in a vertical direction may influence the optical properties of a mirror if the correlation length is of an magnitude comparable to the operating wavelength of the mirror. The perfection of a SL depends on the method of fabrication. The most widely used techniques are magnetron [1, 2] and ion beam [3] sputtering, and evaporation [2]. Mo/Si multilayers (ML) produced by magnetron sputtering and evaporation were extensively studied [1, 2, 4] in the recent decade. In the present work, a ML prepared by ion beam sputtering is investigated by x-ray methods.

X-ray scattering measurements in specular and off-specular geometries effectively characterize the interface morphology [5-7]. Whereas the specular signal being recorded in θ-2θ scans yields the electron density profile in the direction normal to the sample surface, the diffuse scattering reveals details of lateral ordering of the interface roughness as well as the character of vertical replications of roughness from one interface to another.

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The above described sample parameters can be obtained from x-ray reflectivity and diffuse scattering data using various mathematical tools to refine the data. A typical refinement procedure uses linear or nonlinear optimization methods to minimize the difference between experimental data and simulated theoretical curves:

$$\chi^2 = \sum_{i=1}^{N_{\text{exp}}} \left( \frac{x_{i,\text{meas}} - x_{i,\text{cal}}}{x_{i,\text{meas}}} \right)^2,$$

where $N_{\text{exp}}$ is the number of experimental points, $x_{i,\text{meas}}$ and $x_{i,\text{cal}}$ are measured and calculated x-ray intensities, respectively. Commonly used classical calculus-based and hill-climbing procedures have tendency to be trapped in local minima of $\chi^2$ and can result in wrong modelling of sample parameters. Recently invented (see, e.g., [8]) genetic algorithm (GA) combines the advantages of a stochastic search with the intelligent strategy of solution finding that makes this technique very reliable in finding of true global minimum of $\chi^2$. This algorithm mimics the main principle of the evolution theory by C. Darwin, - “survival of fittest”. According to the results of present work, the GA is found to be robust and reliable in finding of unique solution for x-ray data analysis.

**DISCUSSION**

A Mo/Si superlattice consisting of 30 bilayers with a total thickness of approximately 3000 Å was probed by x-rays. Scattered specular and nonspecular signals were recorded by using an “Advanced Thin Film X-ray System, ATX-G”, made by the Rigaku Corporation. The substrate was not chemically cleaned prior the deposition of ML and the root-mean-square surface roughness was estimated by specular reflectivity measurements to be equal to 4 Å. The basic pressure in the deposition chamber was typically 2x10^6 Pa and the Argon pressure during the deposition was held around 2x10^2 Pa. The Argon ion gun was operated at 1000 V with an ion beam current of 50 mA. The Si and Mo plates, both of high purity, were mounted on a water-cooled holder and a particular target was selected by rotating the holder. Substrates were rotated similar to the planetary motion used for ML thickness uniformity, which is assumed to be ±1% for both samples. The first layer on the substrate was Mo, and then Si and Mo were deposited alternatively in 30 pairs with a nominal period of 97 Å. Layer thickness was controlled by deposition time.

The theory used for simulation of reflectivity and rocking curves takes into account a dynamical coherent scattering and use of a distorted-wave Born approximation [9] for calculating the incoherent (diffuse) component. The attenuation of scattered intensity due to the rough interfaces is given by Vidal and Vincent [10], and the auto-correlation function $C_f(R)$ and cross-correlation function $C_{ij}(R)$ describing the lateral and vertical correlation of roughness, respectively, are chosen as:

$$C_f(R) = \sigma_j^2 \exp \left[-(R/A_j)^{2H_j}\right] = \sigma_j^2 C_0(R); \quad C_{ij}(R) = \sigma_i \sigma_j \exp \left(-|z_i - z_j|/\xi_z\right)$$

where $\sigma_j$ is rms roughness of j-interface, $A_j$ and $\xi_z$ are the lateral and vertical correlation lengths, $H_j$ is the Hurst parameter determining the fractal dimension of surface, and $z_j$ is the coordinate of
j-interface. The form of cross-correlation function includes the assumption that vertical correlation is independent on lateral frequency of roughness. The model of roughness (1) can be varied from completely vertically uncorrelated ($\xi_z = 0$) to almost a conformal one for the values of $\xi_z >> |z_i - z_j|$. The coupling of sample parameters which have to be extracted from the fitting of reflectivity and diffuse scattering data makes it necessary to analyze both specular and nonspecular data simultaneously. The instrumental function of diffractometer and resolution effects have been taken into account in similar way as in work [11].

For a detailed description of a GA fitting procedure, the reader is referred to a book by D.Goldberg [8]; here we only highlight GA principles. The first step towards the application of GA for x-ray analysis is the formalization of the physical parameters of a sample, i.e. the coding of real physical parameters into objects of genetic algorithm. The binary string representation is the most widely used technique for this purpose, and we utilize it here for the transformation of physical parameters to discontinuous space. Thus, the continuous scale of physical parameter $x^j$ is divided into a discrete set of variables $j$ ($j = 0 \ldots N$) corresponding to the values of physical variable $x^j$:

$$x^j = x^j_{\text{min}} + j\Delta x^j; \quad \Delta x^j = (x^j_{\text{max}} - x^j_{\text{min}})/2^N,$$

where the extremum is searched in the interval $[x^j_{\text{min}} .. x^j_{\text{max}}]$. Using this simple transformation, the density of probable values for parameter $x^j$ can be easily managed by choosing the appropriate value of $N$. Then value $j$ can be encoded to a binary string during the computer simulations to unify the basis for all fitted parameters, which may have different physical dimensions, in general case. By analogy to biology, the separate physical parameter in a discrete representation, or its binary equivalent, is called a gene. Every point of the phase space of the sample parameters is equivalent to the set of genes $X = (x^1, x^2, \ldots x^j)$ which is called in GA chromosome, or individual. In the computer code a chromosome is represented by an array of binary strings (vertical bars in Fig.1). Thus, the chromosome reproduces a unique state of the sample model determined by the above mentioned physical parameters to be refined from reflectivity and diffuse scattering data.

The second step of GA is composing of a limited population of size $N_p$ from these chromosomes and evolving it according to the biological laws (Fig.1). The GA technique implements three principal operators governing the evolution of a population: mating (crossover), selection and mutation, and a variety of parameters for these operators. There are different types of the selection of chromosomes from a population for subsequent their mating. The current literature identifies the following rules of selection: roulette wheel selection, rank selection, tournament selection and some of their modifications. In the example below we

![Figure 1. Single evolution cycle of genetic algorithm. Transition from Generation $i$ to Generation $(i+1)$.
](attachment:image.png)
vertical correlation lengths are approximately of the same order, $\Lambda = 700 \, \text{Å}$ and $\xi_z = 500 \, \text{Å}$. Measured and fitted theoretical x-ray intensities, plotted in Figure 3, illustrate good agreement of theory and experiment. Panel (a) demonstrate x-ray beam reflected in specular direction, whereas panel (b) shows non-specular x-ray diffuse scattering together with the section of specular beam, both recorded in $\omega/2\theta$ scans (theoretical curves contain diffuse signal only, for clarity). The parameters fitted indicate that the sample possesses a replicated roughness, spreading from substrate-multilayer interface to all successive ones. The strong confinement of diffuse intensity within resonant diffuse scattering (RDS) sheets in the reciprocal space map (Fig.4) confirms this result [12]. The lateral structurization of interfaces within a ML is close to Gaussian (Hurst parameter $H \approx 1$) and represented by a short and quite small waviness of roughness. The thickness of intermediate Mo-on-Si and Si-on-Mo layers is typical for Mo/Si superlattices grown by other techniques. Finally, the absolute value of roughness rms points out the abrupt enough interfaces between all sublayers, that indicates the x-ray mirror will have a good reflectivity and produce a high resolution image.

Table 1. Sample parameters giving best agreement of theory and experiment.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thickness $t_{Mo}$, Å</td>
<td>35</td>
</tr>
<tr>
<td>Thickness $t_{Si}$, Å</td>
<td>36</td>
</tr>
<tr>
<td>Thickness $t_{Mo-on-Si}$, Å</td>
<td>12</td>
</tr>
<tr>
<td>Thickness $t_{Si-on-Mo}$, Å</td>
<td>8</td>
</tr>
<tr>
<td>Roughness $\sigma_{Mo}$, Å</td>
<td>2</td>
</tr>
<tr>
<td>Roughness $\sigma_{Si}$, Å</td>
<td>5</td>
</tr>
<tr>
<td>Lateral correlation length $\Lambda$, Å</td>
<td>700</td>
</tr>
<tr>
<td>Vertical correlation length $\xi_z$, Å</td>
<td>500</td>
</tr>
<tr>
<td>Hurst parameter $h$</td>
<td>=1</td>
</tr>
</tbody>
</table>

Figure 3. (a) specularly reflected intensity, and (b) diffuse scattering measured in $\omega/2\theta$-scans for different offsets $2\theta$. The theoretical data are given by solid lines and experiment is represented by open dots. Curves are shifted progressively for clarity.
use tournament selection where a random group or pair of chromosomes is selected from the population, and the fittest among these individuals take part in mating. In the mating procedure, the selected pair of chromosomes produces offspring, the number of which is the parameter of crossover scheme. The offspring inherit the genes from both parent chromosomes and there are different ways to carry out the crossover. The simplest single-point crossover shown in Fig.1 by dotted lines cuts both parents at a random gene and combines four cut segments into two offspring where parts of different parents are swapped. The crossover operation is performed with some probability $p_{\text{cross}}$ which is the internal parameter of the GA influencing the algorithm convergence. For example, if the parent chromosomes $(x_1', x_2', x_3', x_4')$ and $(y_1', y_2', y_3', y_4')$ are selected and the random cut point is chosen at the third gene, the offspring take a shape $(x_1', x_2', x_3', y_3', y_4')$ and $(y_1', y_2', y_3', x_4')$. Other types of crossover used in GA are two-point crossover (cutting of chromosomes in two points) and uniform crossover (the genes are copied from parents to children randomly with some parametric probability $p_{\text{uni}}$).

A subsequent mutation operator is responsible for permanent exploration of new regions in phase space and the prevention of a population from collapse and degradation. Using the jump mutation procedure, the randomly selected gene $x_j'$ changes its value to any from the range $[x_{j, \text{min}}', x_{j, \text{max}}']$ with probability $p_{\text{mut}}$.

Above described operations of selection, mating and mutation being repeated $m$ times ($2m$ is the number of individuals in population) yields a new generation. To complete a single evolution cycle, the fitness function is calculated for every individual of new generation. After performing of such an $M$ evolution cycles, the final population contains the true sample model with a high probability, as was proven by numerous applications of GA. At the final stage, the procedure of post-hybridization is used to improve, locally, the selected solution (see flowchart Fig.2). For this purpose we utilized the Levenberg-Marquardt method for a nonlinear least-square problem. One important advantage of GA must be mentioned here. Due to the natural symbiosis of random search and intelligent solution finding strategy, a GA method is not sensitive to the choice of initial guess for sample parameters, contrary to other fitting techniques. Moreover, the boundaries of search intervals for parameters may be chosen in a quite wide range, making us sure that true solution is included in search area.

As a result of refinement procedure, the layer thicknesses, interfacial roughness rms, vertical and lateral correlation lengths and density profiles were extracted from fitting of the experimental data (see Table I). The basic period of the superlattice is assumed to consist of four layers, polycrystalline Mo and amorphous Si plus Mo-on-Si and Si-on-Mo interdiffused layers. The thicknesses of these layers are 35 Å, 36 Å, 12 Å and 8 Å, respectively. The roughness is greater in Si-based interfaces (~5 Å) in comparison with Mo-based layers (~2 Å).
CONCLUSIONS

We investigated the density profile of a superlattice and the interface morphology of a Mo/Si multilayer grown on a Si wafer by ion beam sputtering. Interdiffusion layers were detected between basic Mo and Si layers, and the interfacial roughness is found to be vertically correlated. Exact values of roughness rms and its vertical and lateral correlation lengths are extracted from the data by theoretical simulations. The large vertical correlation length indicates the quasi-conformal replication behaviour of roughness. The genetic algorithm applied to parameters refinement is proven to be versatile and robust. The combination of GA with classical gradient optimization methods improves the procedure of final solution search.

REFERENCES

