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Determination of stress in Au/Ni multilayers by symmetric and asymmetric X-ray diffraction

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A stress analysis was done for Au/Ni multilayers prepared by molecular beam epitaxy and by thermal evaporation. The lattice parameters in the growth and in-plane directions of multilayer constituents were directly determined by the analysis of the symmetric and asymmetric X-ray diffraction (XRD) profiles. The $\theta-2\theta$ XRD profiles were interpreted using the 1D model of non-ideal superlattice structure, whereas the asymmetric XRD profiles using the 3D model. Both models were based on the Monte Carlo simulation and on the kinematical theory of scattering. It was shown that considerable stress was encountered in multilayers with in-plane structural coherence.

1. Introduction

Strains in multilayered systems are now important in device technology and in basic research. Their determination is one of more important elements of structural analysis because they can influence the physical properties of multilayers [1]. The most often used X-ray diffraction technique to determine strains is the $\sin^2\Psi$ method. This method was first employed to determine strains in thin films and homogenous materials. Today, this method is also applied to superlattices [2] – [4].

We observe two kinds of reciprocal space maps in the high-angle asymmetric X-ray diffraction profiles of multilayers obtained by different technologies. Generally, we can devide them into maps which have many satellite peaks and do not reveal them. As it was proved in the work [5] that difference is connected with the in-plane structural coherence of sublayers. The presence of satellite peaks on the reciprocal space maps of superlattice indicates a strong in-plane coherence in these systems. It is evident that for systems with the in-plane coherence of sublayers, the $\sin^2\Psi$ method cannot be applied. In this case these profiles should be interpreted with the aid of a theoretical model. The application of the $\sin^2\Psi$ method for multilayers not having the in-plane coherence between sublayers, like that given in [6], may lead to considerable errors. Also in that case, it is desirable to use

theoretical models to determine strains. The aim of this paper is to show the method for strain determination in multilayered systems by making a fit of symmetric X-ray diffraction (SXRD) and asymmetric X-ray diffraction (AXRD) profiles. We present the results obtained for the Au/Ni multilayers. The system reveals large misfit of the lattice parameters (14%).

2. Experimental

The Au/Ni multilayers were deposited by two different techniques: molecular beam epitaxy (MBE) and thermal evaporation in high vacuum. The MBE films are grown on the GaAs substrate in room temperature in systems with basic pressure 10^{-8} Pa. The superlattice period was repeated 25 times and the sample was covered by 700 Å Au layer. In the Au/Ni system deposited by thermal evaporation we used a glass as substrate. The superlattice period was repeated 21 times. The structural characterization was performed by high resolution X'Pert (Philips) diffractometer with CuK_α using $\theta-2\theta$ geometry and reciprocal space maps. For each sample we measured $\theta-2\theta$ in low-angle and high-angle range and reciprocal space maps near the (113) Au and Ni bulk peak positions collected as $\omega-2\theta$ scans.

3. Results and discussion

It is well known that high-angle SXRD profiles contain information on the structure of superlattice in the growth direction. The period of superlattice Λ and the average interplanar distance $\bar{d}$ are directly available from satellite peak positions [4]. We can obtain other parameters, including interplanar distances of multilayer constituents interpreting these profiles with the aid of a one-dimensional (1D) model, such as the model of non-ideal superlattice structure [4]. This program looks for the best fit of calculated and measured profiles, mainly by changing interplanar distances. To interpret the AXRD profiles in the high-angle range, a three-dimensional (3D) model for the calculation of X-ray scattering profiles should be used [5]–[7]. A large number of structural parameters appearing in the 3D model makes the direct fitting of the AXRD profiles impossible. To reduce the number of these parameters we load parameters obtained from the analysis of the SXRD profile. Then by changing other parameters, we look for the best fit of the experimental reciprocal space map.

The low-angle SXRD measurements confirmed the existence of well-defined periodic structure for both samples. The computer program SlerfWin was used for the thorough analysis of the high-angle SXRD profiles (see [4], for details of the program). We use that program because simulation parameters are directly linked with the parameters of our 3D model [7]. Results of these calculations are given in the Table, where Λ denotes the period of multilayers, d_{Au}, d_{Ni} are interplanar distances in sublayers in the growth direction, n_{Au}, σ_{Au}, n_{Ni}, σ_{Ni} are the numbers of monolayers (ML) and their standard deviations, σ_{int} is the standard deviation of interfaces, D_z and σ_z, respectively, are the average grain size and its standard

Table. The parameters deduced from analysing SXRD profiles by SlerfWin.

Parameter	Au/Ni (001)	Au/Ni (111)
d_{Au} [Å]	2.0420	2.3640
d_{Ni} [Å]	1.6040	2.0600
n_{Au} [ML]	11.5	14.68
n_{Ni} [ML]	3.9	18.12
σ_{Au} [ML]	0.2	1.2
σ_{Ni} [ML]	0.2	1.2
D_z [Å]	270	0
σ_z [Å]	140	45
σ_{int} [ML]	0.4	0.3
Λ [Å]	29.62	72.04

deviation. These results proved the (001) and (111) to be growth directions, respectively for samples deposited by MBE and the thermal evaporation. The interplanar distances (d_{Au}, d_{Ni}) in the growth direction for constituents in both samples are larger than in bulk materials. For Au this is the expected behavior. However, for Ni the increase in interplanar distance is connected with the segregation of Au, which occurs when Ni grows on top of Au, and consequently with the increasing free-stress parameter in nickel sublayers. This effect in Au/Ni multilayers was discussed many times before [2], [8].

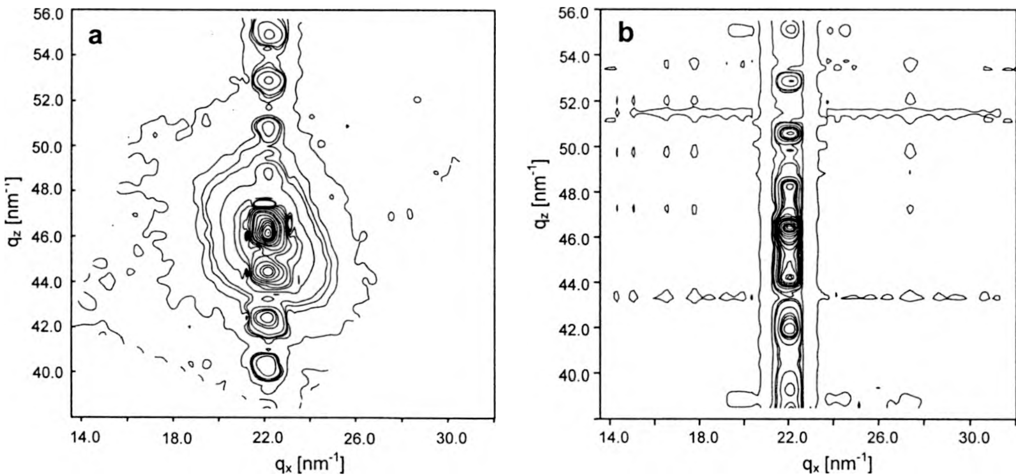

Fig. 1. Experimental (a) and calculated (b) the reciprocal space maps around (113) Au peak position for Au/Ni (001) multilayers.

Next, taking into account the fact that in Au/Ni (001) exists a strong in-plane coherence and that the satellite peaks are situated along one line, $q_x = \text{const}$, we assume that the interplanar distance in both sublayers is equal. Then we look for the best fit of the experimental reciprocal space map by changing lattice parameters in

plane. The result is shown in Fig. 1. The deformations calculated in relation to bulk materials are equal: $\varepsilon_{Au\perp} = 0.13\%$, $\varepsilon_{Au\parallel} = 1.93\%$, $\varepsilon_{Ni\perp} = 8.96\%$, $\varepsilon_{Ni\parallel} = 13.51\%$. Such a large value for Ni sublayers is not surprising if we take into account the fact that Ni sublayers are very thin and that the presence of Au atoms in Ni sublayers may be significant. Moreover, this result suggests that we should very cautiously use this method and the results should be confirmed by other means.

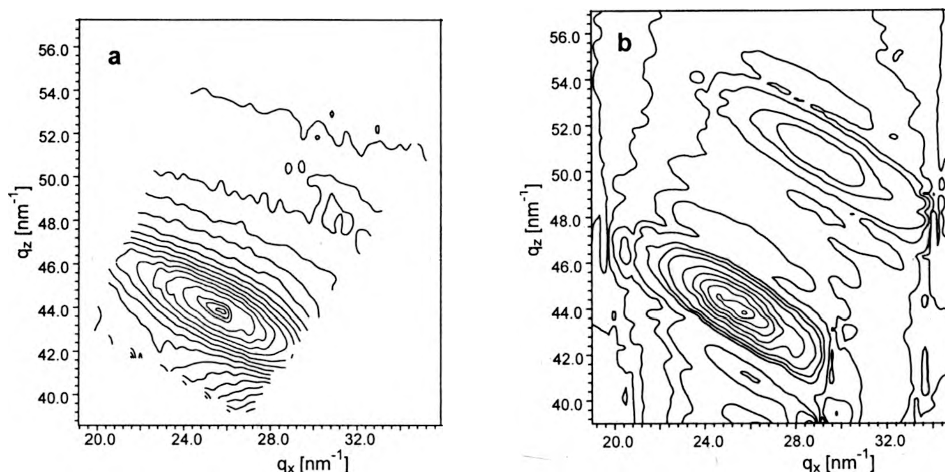

Fig. 2. Experimental (a) and calculated (b) the reciprocal space maps around 113 Au peak position for Au/Ni(111) multilayers.

In contrast to Au/Ni(001) system, for Au/Ni(111) we cannot determine interplanar distances in Ni sublayers because the peak near Ni bulk position is invisible. Additionally we notice that it does not influence (or the influence is too small) the peak position near bulk Au. It allows us to find the peak position only by changing the lattice parameters in Au sublayers. The result is shown in Fig. 2. In that case the stress depends on the direction in the sample and is smaller than for sample Au/Ni(001). We obtain the following results for gold sublayers: $\varepsilon_{Au<111>} = 0.39\%$, $\varepsilon_{Au<1\bar{1}0>} = 0.34\%$, $\varepsilon_{Au<\bar{1}\bar{1}2>} = 0.0\%$.

4. Conclusions

In this work it has been demonstrated that the proposed method, based on the analysis of the SXRD profiles and the reciprocal space maps, can be applied to strain determination. The high angle reciprocal space maps can be analyzed by the 3D model. While interpreting the SXRD profiles the 1D model is sufficient.

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