RESIDUAL STRESS AND OPTIMIZATION OF LATTICE PARAMETERS IN STRONGLY TEXTURED ZnO THIN FILMS

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1. Introduction

Residual stresses are commonly generated in thin films during the deposition process and can influence the film properties. In this article, the impact of the lattice parameters on the evaluation of the measured residual stress is analyzed and the algorithm which finds the optimal lattice parameters of the hexagonal ZnO is proposed. In this work, strong texture is presented in all ZnO samples. For that reason, crystallite group method is considered for the evaluation stress. For the optimization of the lattice parameters genetic algorithm was used. The advantage of the genetic algorithm is its speed, simplicity and its ability to untie from the local extreme and approach the global extreme. The whole principle of genetic algorithm is described in [1].

2. Background

Diffractometric determination of residual stresses is based on the measurement of the interplanar distance d_{mer}^{hkl} of selected lattice planes and their comparison without stress values d_0^{hkl}. Their relative difference, which is identical with the value of the deformation in the measured direction determined by the angles φ, ψ, is evaluated

$$\varepsilon_{\varphi\psi}^{hkl} = \frac{d_{mer}^{hkl} - d_0^{hkl}}{d_0^{hkl}} \tag{1}$$

Equilibrium values of interplanar spacing are calculated from table values of lattice parameters by

$$d_0^{hkl} = \left(\frac{H^2}{a_0^2} + \frac{l^2}{c_0^2}\right)^{-1/2} \tag{3}$$

for tetragonal and hexagonal materials. A parameter $H^2 = h^2 + k^2$ for tetragonal symmetry and $H^2 = \frac{4}{3}(h^2 + hk + k^2)$ for hexagonal symmetry.

From these relations, we can see that the variables $\varepsilon_{\varphi\psi}^{hkl}$ are affected by the used lattice parameters. The question is how "correct" lattice parameters influence the stress evaluation. For the correct determination of the stress (strain) components, the slope of $\varepsilon_{\varphi\psi}^{hkl} = f(\sin^2\psi)$ is decisive in particular. Differences in the table values of the lattice parameters are small, and linearized dependencies can be used $\varepsilon_{\varphi\psi}^{hkl}(a_0, c_0)$ for determination their influence on the values $\varepsilon_{\varphi\psi}^{hkl}$. In the case of tetragonal and hexagonal materials for small variations of deformation, the following applies:

$$\Delta\varepsilon = \frac{\mathrm{d}\varepsilon}{\mathrm{d}d_0}\left(\frac{\partial d_0}{\partial a_0}\Delta a_0 + \frac{\partial d_0}{\partial c_0}\Delta c_0\right) =$$

$$= -\frac{d_{mes}}{d_0^2}\left(\frac{d_0^3 H^2}{a_0^3}\Delta a_0 + \frac{d_0^3 l^2}{c_0^3}\Delta c_0\right) = \tag{3}$$

$$= -d_{mer} d_0 \left(\frac{H^2}{a_0^2}\frac{\Delta a_0}{a_0} + \frac{l^2}{c_0^2}\frac{\Delta c_0}{c_0}\right)$$

It can be seen that changing the lattice parameter Δa_0, the points corresponding to the diffraction with $h = k = 0$ are not affected. Likewise, changing Δc_0 doesn't effect on position of $\varepsilon_{\varphi\psi}^{hkl}$, where $l = 0$. For a detailed examination of the influence of changes in both lattice parameters, it is appropriate to introduce new independent parameters Δz and ϵ, where changes a_0 and c_0 are expressed as follows:

$$a_0 = (1 + \Delta z)(1 + \epsilon)a_{in} \tag{4}$$

$$c_0 = (1 + \Delta z)(1 - 2\epsilon)c_{in} \tag{5}$$

where a_{in} and c_{in} are starting values of lattice parameters. It is easy to convince that changing the parameter Δz leads to a uniform change in the dimensions of the elementary cell without changing shape of cell. On the contrary, changing the parameter ϵ causes a change in cell shape (changing the ratio c_0/a_0).

$$\varepsilon_{xx} = \varepsilon_{yy} = \frac{\Delta a_0}{a_0} = \epsilon, \qquad \varepsilon_{zz} = \frac{\Delta c_0}{c_0} = -2\epsilon \tag{6}$$

If the ratio c_0/a_0 changing

$$\Delta\varepsilon = -d_{mer} d_0 \left(\frac{H^2}{a_0^2} - 2\frac{l^2}{c_0^2}\right)\epsilon \tag{7}$$

In case, standard $\sin^2\psi$ method, when only one diffraction hkl is used for the measurement, the equation (7) is constant and changing ϵ parameter leads to shift of $\varepsilon_{\varphi\psi}^{hkl}$ values. The choice of equilibrium lattice parameters therefore does not affect the slope of $\varepsilon_{\varphi\psi}^{hkl} = f(\sin^2\psi)$ or measurement stress. However, if different hkl diffractions are used to construct the dependence $\varepsilon_{\varphi\psi}^{hkl} = f(\sin^2\psi)$, it can be seen from the expression (7) that the different combination of indices lead to different values of $\Delta\varepsilon_{\varphi\psi}^{hkl}$ for the same value of parameter ϵ. The points corresponding to the diffraction $hk0$ and $00l$ are moved in the opposite direction. The choice of "correct" values a_0 and c_0 can influence the scattering of points in the graph $\sin^2\psi$, but also slope.

On the other hands, this "sensitivity" can be used to optimize lattice parameters and residual stress, simultaneously. For optimization can be used least squares method

$$R = \sum_{i=1}^{n}\left(\varepsilon_i^{exp} - \varepsilon_i^{fit}\right)^2 \tag{8}$$

where $\varepsilon_i^{exp} \equiv \varepsilon_{\varphi\psi}^{hkl}(a_0, c_0)$ are values from Eq. (1) and ε_i^{fit} correspond to

$$\varepsilon_i^{fit} = P\sigma_{\parallel}(a_0, c_0) + Q\sigma_{\parallel}(a_0, c_0)\sin^2\psi \tag{9}$$

where parameters P and Q dependence from elastic coefficient of the material.
For strongly textured thin films and crystallite group method:

$$P = 2S_{13}, \quad Q = S_{11} + S_{12} - 2S_{13} \tag{10}$$

and ψ_0

$$\psi_0 = \arcsin\sqrt{\frac{-2S_{13}}{S_{11} + S_{12} - 2S_{13}}} \tag{11}$$

Parameters P, Q and ψ_0 are the same for all diffractions. [2]

3. Results

The parameters ϵ and Δz were used as input variables, which are related to the lattice parameters according to (4) and (5). Using these, a population (number matrix) was created, which represents the first generation at the beginning of the program. Based on typical genetic operations, such as the selection of the strongest individual, mutation and crossing, the nth generation has been acquired. If the nth generation did not differ from n-1, the condition for completing the algorithm was fulfilled.
Optimization algorithm was written in Matlab. A population of 30 individuals was used for the calculation. For the calculation were used starting values of lattice parameters ZnO, $a_{in} = 0{,}324982$ nm and $c_{in} = 0{,}520661$ nm [3]. The optimal lattice parameters are calculated in Table 1.

Table 1 Optimized lattice parameters of ZnO

Sample	*a* [nm]	*c* [nm]
ZnO_A	0,325013	0,520495
ZnO_B	0,325003	0,520562
ZnO_C	0,325055	0,520471
ZnO_D	0,325017	0,520490
ZnO_N47	0,324941	0,520824
ZnO_NZ6	0,325034	0,520465
ZnO_NZ9	0,324936	0,520725

4. Conclusion and discussion

The resulting lattice parameter values listed in Table 1 differ only slightly between the "start" values a_{in} a c_{in}. However, the application of the described optimization procedure showed that in principle it is possible to simultaneously determine the residual stress and the lattice parameters of the analyzed materials whose symmetry is less than cubic. For the analysis, however, it is necessary to use a method that combines several diffractions for the construction of dependence $\varepsilon_{\varphi\psi}^{hkl} = f(\sin^2\psi)$. It is important to note that the results of analysis are influenced by the choice of equilibrium lattice parameters.

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References:
[1] Sekaj I., Evolučné výpočty a ich využitie v praxi, Bratislava: IRIS (2014), ISBN 80-89018-87-4.
[2] Novák P., Analýza reziduálnych napätí v tenkých vrstvách pomocou rtg difrakcie pri malom uhle dopadu, Dissertation thesis, (2017).
[3] Morkoc H., Zinc Oxide – Fundamentals, Materials, and Device Technology, WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim (2009), ISBN 978-3-527-40813-9