



5. Mikrofotografie frézovaných vzorků 5 a 6 potvrzují účinek plastické deformace povrchových vrstev vzniklé mechanickou interakcí materiálu s nástrojem, která je v případě sousledného frézování výraznější a sáhá do hloubky cca 50  $\mu\text{m}$ .

6. Výsledky stanovení mřížkových parametrů povrchových vrstev zkoumaných vzorků uvedených v tab. 2 ukazují, že elektroerozivní hloubení vede ve všech případech k hodnotám nižším než frézování. Této skutečnosti, která je zřejmě důsledkem difúze atomů elektrody (grafit, měď) do povrchových vrstev obráběného vzorku, bude věnována pozornost při dalších měřeních.

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## Literatura

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## X-RAY DIFFRACTION FROM POLYCRYSTALLINE MULTILAYERS WITH FLAT INTERFACES IN GRAZING-INCIDENCE GEOMETRY

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### Abstract

X-ray diffraction from polycrystalline multilayers in grazing-incidence geometry is a new method to measure the crystallite size and its distribution as a function of depth in multi-layer thin films. The principle relies on the idea that when X-rays are scattered at an interface the incident and scattered waves create a standing wave whose periodicity can be varied and thereby enhance the scattering at certain depths. By varying the angle of incidence the penetration depth is changed.

This has been used for mono-crystalline semiconductor wafer samples and for very thin (30 nm) polycrystalline layers. Numerical examples are shown for multi-layer consists of 14 periods, each containing a 4 nm thick Nb layer and a 11 nm thick Al layer.

### I. Introduction

X-ray powder diffractometry is a standard method for obtaining the size of crystallites in industrial materials. However in general the method is suited to obtaining the size normal to the surface using the focusing Bragg-Brentano geometry. To estimate the general shape of crystallites in bulk samples, several reflections are required, however this method is less suited to thin films that in general exhibit significant texture. Hence these methods are essentially bulk methods, i.e. they assume a random distribution of orientations and become difficult in the analysis of thin films when the crystallite size in the plane of the interface cannot be determined. Access to scattering planes normal to the interface is possible at grazing angles of incidence. This in-plane scattering geometry relies on the refractive index of X-rays being less than unity, such that there is a critical angle for total external reflection. Thus the X-rays can enter through the sample surface, scatter from planes normal to

the surface plane and then scatter out through the same surface. By varying the angle of incidence the penetration depth is varied. This has been used extensively in the study of mono-crystalline semiconductor wafer samples and can also be shown to work for very thin (30nm) polycrystalline layers. Although some average crystallite size is determined, knowledge of the variation as a function of depth in thin films can be very valuable.

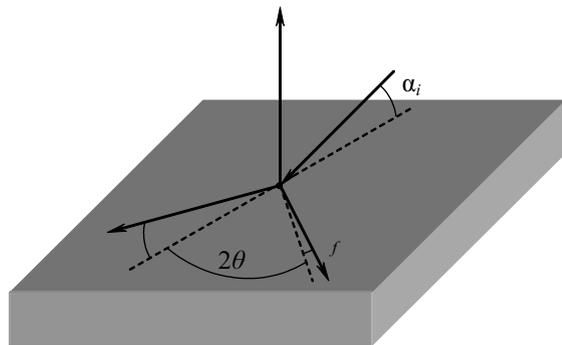
This paper describes a new method to measure the crystallite size and its distribution as a function of depth in multi-layer thin films. The principle relies on the idea that when X-rays are scattered at an interface the incident and scattered waves create a standing wave whose periodicity can be varied and thereby enhance the scattering at certain depths.

### II. Theoretical description

In the in-plane grazing-incidence geometry, both the angle of incidence  $\theta_i$  of the incident radiation, and the angle of exit  $\theta_f$  of the scattered beams are comparable with the critical angle  $\theta_c$  of total external reflection of X-rays (Fig. 1). Thus, the interaction of x-rays with matter is rather strong and it cannot be described by a simple kinematical approximation. On the other hand, the polycrystalline grains can be assumed much smaller than the extinction length  $\lambda_{\text{ext}}$  of x-rays in the material, so that the individual grains scatter kinematically.

We describe the scattering process using the distorted-wave Born approximation (DWBA) [1], in which the total scattering potential  $\hat{V}(\mathbf{r})$  occurring in the wave equation:

$$(\nabla^2 + K^2)E(\mathbf{r}) = \hat{V}(\mathbf{r})K(\mathbf{r}); K = \frac{2\pi}{\lambda} \quad (1)$$



**Fig. 1.** Grazing incidence diffraction, the crystallographic plane is perpendicular to crystal surface.

is divided in two terms  $\hat{\mathbf{V}} = \hat{\mathbf{V}}_A + \hat{\mathbf{V}}_B$ . The first term describes the undisturbed system, the latter is the disturbance. Within the DWBA, the undisturbed wave equation with  $\hat{\mathbf{V}}_A$  is solved exactly, while the scattering from  $\hat{\mathbf{V}}_B$  is assumed kinematical.

In simple model with flat interfaces  $\hat{\mathbf{V}}_A$  corresponds to an amorphous multi-layer and we assume that the primary x-ray beam  $E_i$  is scattered by  $\hat{\mathbf{V}}_A$  which generates a standing wave-field  $E_A^{(1)}$  in the multi-layer; this wave-field is diffracted in the polycrystalline grains due to  $\hat{\mathbf{V}}_B$ . The diffracted wave-field,  $E_A^{(2)}$  is scattered again by  $\hat{\mathbf{V}}_A$  and finally, the measured wave-field  $E_f$  results. In this choice,

we completely neglect the scattering from interface roughness.

We assume a sample consisting in  $N$  layers. The wave-field  $E_{Aj}^{(1)}(\mathbf{r})$  in layer  $j$  is generated by the incident wave consisting of transmitted and reflected components and their amplitudes can be calculated using the well-known matrix formalism [2]. The second, independent solution  $E_{Aj}^{(2)}(\mathbf{r})$  of Eq. (1) (where  $\hat{\mathbf{V}} = \hat{\mathbf{V}}_A$ ) is chosen so that its vacuum incident wave equals the actual scattered wave  $E_f$  and this solution is time inverted.

The diffuse part of differential cross-section of the diffraction from polycrystalline grains is [1]:

$$\frac{d}{d_{diff}} = \frac{1}{16} \left\langle \frac{2}{B} \right\rangle \quad (2)$$

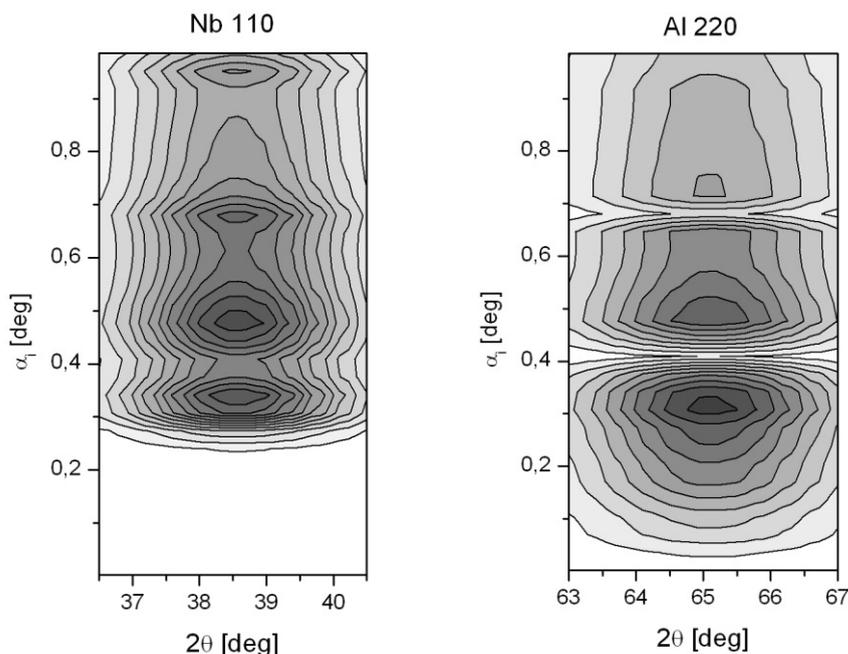
where  $\langle \rangle$  denotes the averaging over a statistical ensemble of random positions  $\mathbf{R}_{jn}$  (position vector of grain  $n$  in layer  $j$ ), of the grains, random directions of  $\mathbf{h}_{jn}$  (reciprocal lattice vector of grain), and over random grain sizes. The matrix elements  $B$  is:

$$B = \left\langle E_A^{(2)} | \hat{\mathbf{V}}_B | E_A^{(1)} \right\rangle \quad (3)$$

and disturbance  $\hat{\mathbf{V}}_B$  comprises the crystal structure of individual polycrystalline grains.

### III. Numerical examples

In the simulations, we have assumed the multi-layer consists of 14 periods, each contains a 4 nm thick Nb layer and a 11 nm thick Al layer. We also assume that the polycrystalline grains are spherical, each grain has the same radius  $R$ , grain positions are completely uncorrelated,



**Fig. 2.** Calculated intensity for Nb110 and Al220, mean grain size  $R = 2$  nm,  $f = 0,5^\circ$ .



all grain orientations have the same probability (no texture).

Figure 2 shows an intensity map calculated in the coordinates  $(2\theta; \varphi)$ , assuming  $\varphi = 0,5^\circ$  where  $2\theta$  is the in-plane scattering angle. In this map, a part of the Debye ring of Nb110 and Al220 is depicted. The width of the ring in the  $2\theta$  direction is inversely proportional to the grain radius  $R$ . Along the Debye ring, the intensity modulation follows the dependence of the intensity of the wave-field on  $d_i$ ; the period of the modulation is inversely proportional to the multi-layer period. The rings Nb110 and Al220 have different structures; these differences are caused by different intensities of the standing wave in Nb and Al layers. Below  $d_c$ , the penetration depth of the incoming beam is very small and no diffraction from the grains takes place.

Figure 3 shows the intensity scans for  $2\theta = \text{const.}$  calculated for Nb110 and Al220 diffraction and reflection on assuming multi-layer.

#### IV. Conclusions

Model with flat interfaces (and expectations in paragraph III.) is too simplified to describe real experimental data. The most important point of the further work is description of the scattering process from interfaces roughness. The next problems are the texture, influence of the random size of the grains, etc.

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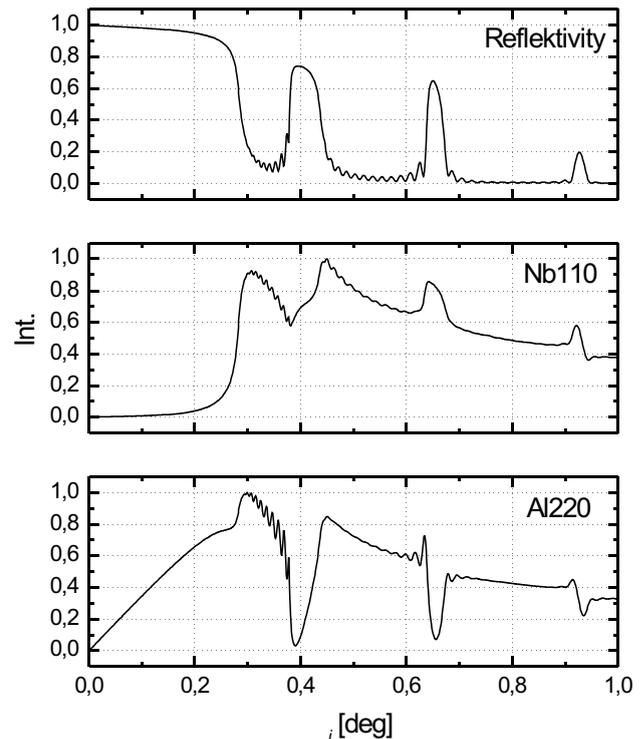


Fig.3. The intensity scans for  $2\theta = \text{const.}$  for Nb110 and Al220 and reflection on multi-layer, mean grain size  $R = 2 \text{ nm}$ ,  $\varphi = 0,5^\circ$ .

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## TANTALONIOPATES IN CASSITERITE: INCLUSIONS OR EXSOLUTIONS?

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#### Keywords

Tantaloniobates, cassiterite, exsolution process, inclusion, pegmatite

#### Abstract

Tantaloniobates in cassiterite from the Annie-Claim Pegmatite (Canada) and pegmatites in the Czech Massif were studied by the following methods: light microscopy, single-crystal X-ray diffraction, electron microprobe, and transmission electron microscopy (TEM). A concentration gradient around inclusions (electron microprobe results) and a defined structural orientation of inclusions within the cassiterite matrix (X-ray and electron diffraction results) were observed. This confirms that in the cases studied, tantaloniobates in cassiterite formed as a result of an exsolution process.

#### Introduction

In natural samples, it is sometimes difficult to differentiate between inclusions that were trapped during crystal growth and products of post-crystallization exsolution processes. In order to understand the process of rock formation, the inclusions/exsolution have to be studied in detail. Presently, we studied cassiterite from the Annie-Claim Pegmatite (Canada) and pegmatites in the Czech Massif. The following methods were employed: light microscopy, single-crystal X-ray diffraction, electron microprobe and transmission electron microscopy (TEM).

The word "inclusion" is being used loosely. In general, an inclusion can be any solid or fluid phase enclosed in the surrounding matrix. This covers inclusions *sensu stricto*, which are randomly trapped by a crystal during its growth, as well as precipitates that form by exsolution in the subsolidus. Hereafter, we shall use the term "inclusion" in non-genetic context, whereas the term "inclusion *sensu stricto*" as opposed to terms "precipitate" or "exsolution" will allude to the formation process involved.