Quantitative Analysis of the Intensity of Interference Fringes of X-Ray Diffraction Line Profiles

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X-Ray diffraction spectra of thin films may show characteristic interference fringe patterns which wash out with increasing surface roughness. From the intensity of the secondary maxima quantitative data on the roughness can be derived. Model calculations are presented assuming that the distribution of heights about the mean surface level is Gaussian. The theory is applied to explain the diffraction spectra of single-crystal silver films. It is shown that some uncertainty in the results remains due to the influence of lattice distortions.

1. Introduction

In two previous papers [1, 2] we have shown that useful information on surface roughness of thin films can be derived from the analysis of interference fringe patterns in the flanks of x-ray diffraction peaks. The evaluation procedure was restricted to highly uniform films where the 4th and 5th order secondary maxima were well resolved. In practical cases, however, often only secondary maxima of lower order can be detected with sufficient accuracy. Even for such films roughness data can be obtained if a larger inhomogeneity of the surface is taken into account and if the disturbing influence of instrumental broadening on the diffraction line profiles is eliminated. It is the aim of the present paper to extend the theory in the described manner.

Starting point of our calculation is the assumption that the distribution of heights about the mean surface level is Gaussian. Such a distribution has already been improved for thin film surfaces by other authors [3, 4]. The theoretical results are compared with experimental data obtained for thin silver films deposited on silicon substrates. Due to the relatively weak chemical silver/silicon interaction [5] is was expected that these films contain only little lattice distortions. Moreover, a gold single-crystal [6] was at our disposal for the exact determination of the instrumental broadening (gold has nearly the same lattice constant as silver).

2. Theory

X-ray diffraction measurements on an ideal film of N parallel lattice planes lead to an interference fringe pattern whose intensity is given by [7]

$$I_N(\varphi) = N^2 u^2 (\varphi^{-1} \sin \varphi)^2 , \qquad (1)$$

where

$$\varphi = N b a \Delta\theta , \qquad (2)$$

and

$$a = 2 \pi \lambda^{-1} \cos \theta . \qquad (3)$$

In these equations b is the distance between lattice planes, u their scattering amplitude, λ the wavelength of the x-rays and $\Delta\theta$ the deviation from Bragg's angle θ.

Vacuum deposited thin films deviate from the ideal case in that they consist of portions of varying thicknesses. In our previous paper [2] a simple meander model was studied assuming that the films consist of two equally sized portions of thickness differing by a step height around a mean thickness, where the step height was typically one or two inter-lattice plane distances. Although this model seemed reasonable for highly uniform films, a more realistic picture is needed for the description of rougher surfaces. We start with generalizing (1) for an arbitrary distribution of N around the mean film thickness. The total intensity $I(\varphi)$ is then given by

$$I(\varphi) = \sum_N f(N) I_N(\varphi) , \qquad (4)$$

where $f(N)$ is the fraction of the film with N lattice planes. Here we sum up intensities instead of amplitudes considering a subdivision of the monocrystal-

line film into small coherent domains which scatter incoherently with respect to one another [8]. Such a model may be suitable for films with a marked mosaic structure where the mean lateral spacing of surface steps is equal or larger than the spacing of the low angle grain boundaries. All informations on the structure of our films [9–11] are uptil now in agreement with this picture. For a Gaussian distribution $f(N)$ we obtain

$$I(\Delta\theta) = \frac{\sum\limits_{N} N^2 e^{-(Nb-d)^2/\delta^2} (N b a \Delta\theta)^{-2} \sin^2(N b a \Delta\theta)}{\sum\limits_{N} N^2 e^{-(Nb-d)^2/\delta^2}},$$ (5)

normalized to $I(\Delta\theta = 0) = 1$. δ is the scattering of the Gaussian distribution function, d the mean thickness of the film. If the sum over N is replaced by an integral one can easily check that the heights of secondary maxima and minima of $I(\Delta\theta)$ do not depend on d but only on the ratio $r = \delta/d$. The quantity r is a measure for the roughness of the

surface but higher values are obtained in comparison to the roughness $R = D/2d$ derived from a meander model where D is the step height [2].

Figure 1 shows four theoretical curves calculated on the basis of (5) for $r = 0$, 10%, 20% and 30%, respectively. The intensity I is plotted as a function of $\Delta\theta$. Obviously the maxima of higher order are less pronounced at higher roughness. Taking into account this result we propose to determine r from experimental data by computing the quantities

$$J_n(r) = \{I_n^{\max}(r) - I_{n+1}^{\min}(r)\}/I_n^{\max}(r=0).$$ (6)

Here $I_n^{\max}$ is the intensity of the secondary maximum of n-th order, $I_{n+1}^{\min}$ is the neighbouring minimum. $I_n^{\max}(r=0)$ can be easily calculated from (5); the result is shown in Figure 1a. The advantage of calculating differences of relative intensities according to (6) is that some unknown background noise in the experimental profiles is eliminated automatically. Particularly, a possible influence of
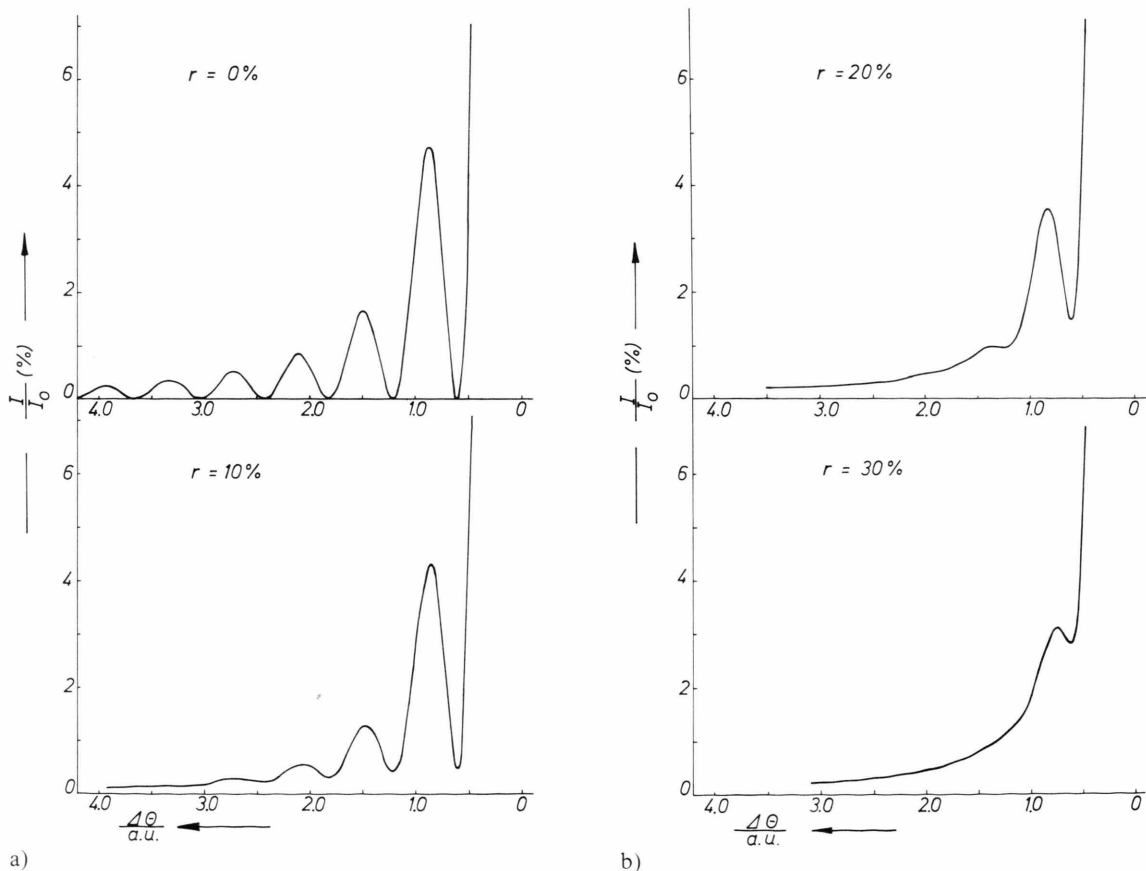

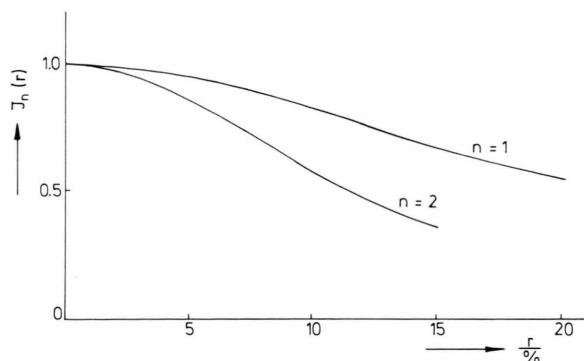Fig. 1. Interference fringe patterns calculated on the basis of (5).

Fig. 2. The quantity J_n of (6), which describes the intensity of the secondary maxima, in dependence on surface roughness r. n is the order of the secondary maxima.

coherent scattering of portions of the film is drastically reduced by this procedure.

Figure 1 shows that for films with roughnesses of 10% or more it seems to be fruitful to evaluate only the first or the second order secondary maximum. The results of the calculation of $J_n(r)$ are given in Figure 2. It is interesting to note that the half width β of the main peak also decreases with increasing roughness. The relative change amounts to more that 10% for rough films and, therefore, cannot be neglected in the determination of film thickness from x-ray line broadening [7]. To the best of our knowledge this error source has not been previously discussed in the literature.

Moreover, the calculation reveals that the main peak shape is also influenced by roughness. The shape shifts from the theoretical sine-like profile of (1) towards a Cauchy profile with increasing roughness. This may be one of the reasons that Cauchy profiles often describe experimental peaks of thin films [12, 13] better than sine-like profiles.

3. Comparison with Experimental Data

The theory is applied to the interpretation of the (111) diffraction peak of a 17 nm thick single-crystal silver film. This film has been deposited on a (111) oriented silicon substrate at room temperature under UHV conditions. Details of the preparation method and the highly sensitive $\theta-2\theta$-scanning unit used here have been described elsewhere [8]. A cobalt tube was used as the radiation source in the present case ($\lambda = 0.17902$ nm).

The measured spectrum is shown in Figure 3. The thickness of 17 nm had been determined from the fringe spacing of the secondary minima [15] as well as by direct vibrating-quartz-monitor control. Two secondary maxima can be detected on each side of the peak but their intensity is slightly asymmetric. Similar spectra have been reported by other authors [13, 15, 16]. In order to minimize the influence of twin faults [13] we have restricted ourselves in evaluating the low-angle side of the peak as drawn in Fig. 4 on a magnified scale (full curve).

Analyzing with the help of (6) leads to $J_1 = 0.85$ and $J_2 = 0.66$, respectively. From Fig. 2 we obtain $r = 9\%$ in both cases. The theoretical curve calculated from (5) for $r = 9\%$ is also drawn in Fig. 4 (dashed curve). Obviously the agreement between the experimental and the theoretical spectrum is rather bad. Particularly in the environment of the first secondary maximum stronger deviations are evident which indicate that instrumental broadening cannot the neglected in the present case.

In order to get a better coincidence with the theoretical curve, we have extracted the physical profile $I_{Phys}(\psi)$ from the measured one $I_{exp}(\varphi)$ by
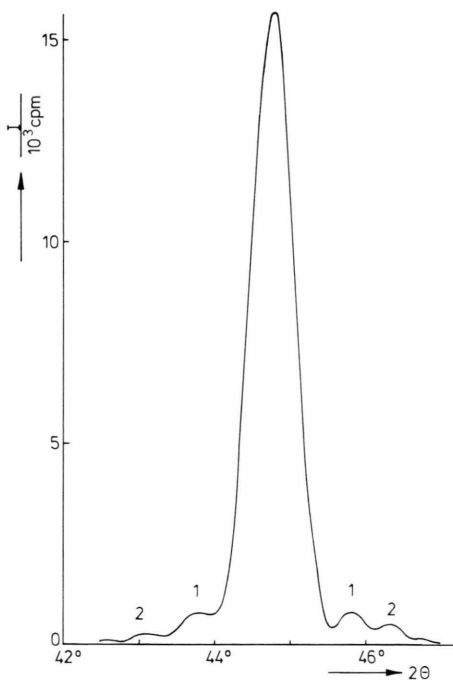

Fig. 3. Peak profile measured for a 17 nm thick silver film. The numbers indicate that two secondary maxima can be detected on each side of the peak.
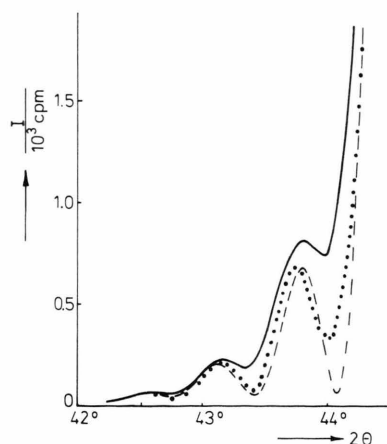
Fig. 4. Experimental data for a 17 nm thick silver film (solid curve) plotted on a magnified scale as compared to Figure 3. The physical profile obtained after deconvoluting with the instrumental profile of Fig. 5 is characterized by the dotted curve. The dashed curve was calculated on the basis of (5) for $r = 9\%$.

deconvoluting with the instrumental broadening g by means of a Fourier analysis according to the Stokes method [17], i.e.

$$I_{\exp}(\varphi) = \int I_{\text{Phys}}(\psi)\, g\,(\varphi - \psi)\, \mathrm{d}\psi\,. \qquad (7)$$

Figure 5 shows the profile $g\,(\theta)$ obtained for our instrument by calibrating with a (111) oriented
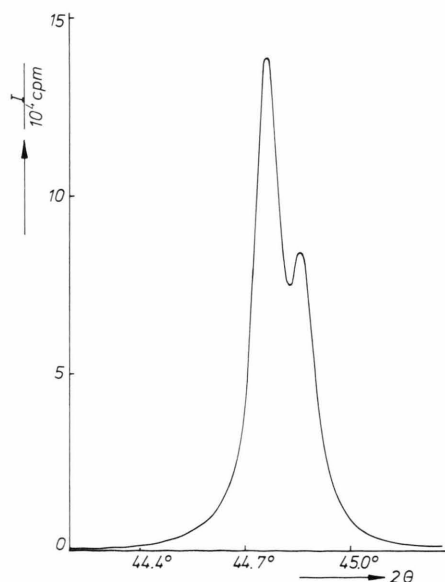

Fig. 5. Instrumental broadening profile as measured for a (111) oriented gold single-crystal sample.

single-crystal gold sample. To demonstrate the effect of instrumental broadening on the results we additionally present in Fig. 4 the corrected profile I_{Phys} as calculated from (7) (dotted curve). Obviously this profile is in better agreement with the theoretical curve. The quantitative evaluation with the help of (6) now leads to $J_1 = 0.83$ and $J_2 = 0.72$, respectively. From Fig. 2 we read out $r = 10\%$ and $r = 8\%$, respectively, which is in the average identical with the $r = 9\%$ obtained for the uncorrected profile. Thus it becomes evident once more that the evalution with the help of (6) is rather insensitive on profile broadening.

The deviations between the corrected profile and the theoretical curve near the first minimum can be traced back to a lattice fault broadening the magnitude of which can be estimated from the disagreement of the thickness derived from the fringe spacing with that derived from the peak broadening of I_{Phys} [18]. A quantitative treatment is only possible if the amount and special distribution of the lattice faults are known. Then, an additional deconvolution is necessary. A symmetrical Gaussian distribution often used in the literature to describe strain broadening [19] is considered to be unsuitable to explain the results in the present case. The shape of the experimental profile (Fig. 3) should rather be ascribed to twin faults which cause the distribution function to become asymmetric [20, 21].

4. Conclusions

A theoretical interpretation of x-ray interference fringe patterns is presented assuming that the distribution of heights about the mean surface level is Gaussian. In the frame work of this model quantitative data on roughness can be determined even in cases where only a few secondary maxima are detectable against the background noise. It should be mentioned, however, that the r-values characterize the volume effective for diffraction and, therefore, cannot be related directly to roughness factors measured by gas adsorption experiments [9].

Deviations between the theoretical and experimental peak shapes are mainly due to the instrumental line broadening and to the influence of lattice faults and imperfections. Note that the effect of instrumental broadening is the more pronounced the thicker the films are. Some other sources of error, however, should be kept in mind while using

Fig. 2: (a) gradients of the thickness in the film due to an oblique deposition geometry, (b) reduction of the illuminated volume with increasing diffraction angle [2], (c) influence of coherent scattering which may cause the background level to move, and (d) deviations of the distribution function of surface roughness from Gaussian type which may occur particularly for very rough films [4].

Nevertheless, results of general importance can be derived from the theoretical model presented here. The half widths of the peaks decrease with increasing roughness, simultaneously the peak shape shifts towards a Cauchy profile. For films with

r-values higher than 20% the fringe patterns with $n > 1$ wash out totally so that a roughness determination may only be possible by evaluation of the first order secondary maximum. Then, however, deconvolution is absolutely necessary, and all the restrictions mentioned above must be considered in detail.

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