Experimental investigation of influence of surface roughness of crystals on diffraction of x rays in conditions of total external reflection

R. M. Imamov, E. A. Kondrashkina, D. V. Novikov, and S. A. Stepanov

Institute of Crystallography, Academy of Sciences of the USSR

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The authors investigate the influence of coarse and fine scale surface roughness of crystal surfaces on x-ray diffraction in conditions of total external reflection (XDTER). They suggest a method of taking account of the surface relief of specimens in processing a diffraction experiment, based on the use of roughness parameters determined from curves of specular reflection of x rays measured simultaneously with curves of XDTER. They show that for standard semiconducting plates after superfinish surface treatment, and also after ion implantation, roughness does not play an important part in experiments to determine the thickness of amorphous surface layers by the method of integral XDTER curves.

1. INTRODUCTION

In recent years to investigate the structure of thin surface layers of crystals, successful use has been made of the method of x-ray diffraction in conditions of total external reflection (XDTER). On the basis of measurements of the integral intensity of XDTER there was determined the thickness of surface amorphous layers of silicon in the range 1-100 nm, formed by oxidation[1,2] mechanical damage,[2] and ion implantation.[3]

On the other hand, it is known that the most perfect surfaces of semiconducting crystals, processed to the high 14th class, have roughness with a depth of the irregularities $R_z \gtrsim 30$ nm (All-Union State Standard GOST 2789-73). In this connection there arises the question of to what extent the surface roughness of the test specimens influences the experimental integral curves of XDTER. In other words, it is necessary to elucidate how we can determine the parameters of the roughness and take account of them in processing diffraction measurements. This article describes an experimental investigation of this problem.

To determine the effect of roughness we can use x-ray reflectometry (total external reflection of x rays outside the conditions of diffraction). Since in this method the angles made by the x-ray beams with the surface, and the depth of penetration of the radiation into the crystal are practically the same as for diffraction in conditions of TER, it is natural to suppose that the character of the influence of roughness in both cases will be identical. On the other hand, the curves of specular reflection of x rays are not sensitive to the crystal structure, therefore it is much simpler to extract the roughness parameters from them than directly from diffraction measurements.

The influence of surface roughness of the specimens on the results of x-ray reflectometry were studied in detail earlier (see Refs. 4-11 and survey in Ref. 12). As shown in Refs. 5-8, the character of this influence depends on the ratio of the mean dimension ℓ of the irregularities along the surface of the crystal and the critical parameter $\ell_{cr} = \lambda/\phi_c^2 \sim 1$-$10$ μm (λ is the wavelength of the x radiation and Φ_c is the critical angle of TER).

If $\ell \ll \ell_{cr}$ the irregularities can be regarded as small-scale. Such surface roughness plays the part of a layer with reduced density and leads to reduction of the critical angle Φ_c (Ref. 8).

In the case of large-scale irregularities ($\ell \gg \ell_{cr}$), the surface of the crystal can be represented in the form of statistically distributed microareas of various orientations. The scatter of the orientations causes widening of the differential curves of specular reflection in comparison with the angular divergence of the incident beam and corresponding smoothing of the integral TER curves.[6,7]

To compare the actions of roughness on the curves of specular reflection of x rays and diffraction in the conditions of TER, we investigated four crystals with different types of surface roughness.

2. EXPERIMENT

The test specimens were silicon plates with orientation of the (111) surface, diameter 76 mm, and thickness about 350 μm.

Specimen I was an initial perfect substrate, the surface of which was processed by the standard method, including superfinishing treatment of the test side to the 14th class of roughness. Specimen II was the same substrate, but subjected to implantation of heavy thallium ions with energy 100 keV and dose $5\cdot10^{13}$ ions/cm².

Specimens III and IV were model structures, qualitatively corresponding to fine- and coarse-scale roughness. The surfaces of these crystals were polished with various abrasives, and then etched to remove the layer with disturbed crystal structure. As a result, the surface of specimen III had marked small-scale roughness (Fig. 1a) the tangential dimension of which was about 5 μm, i.e., less than the characteristic dimension $\ell_{cr} = \lambda/\Phi_c^2 \approx 10$ μm for a silicon specimen with Cu Kα radiation. The depth of the relief of specimen III, measured with an MII-4 optical interferometer, was about 300 nm. Specimen IV had slight fine-scale roughness (Fig. 1b) but possessed microrelief, the parameters of which, measured on the MII-4, were: depth about 300 nm. tangential characteristic dimension $\ell = 200$ μm $\gg \ell_{cr}$.

The x-ray measurements were made on a spectrometer which was described in Ref. 13.

The specimen, placed in the Laue diffraction position (220), was rotated about the vertical axis of

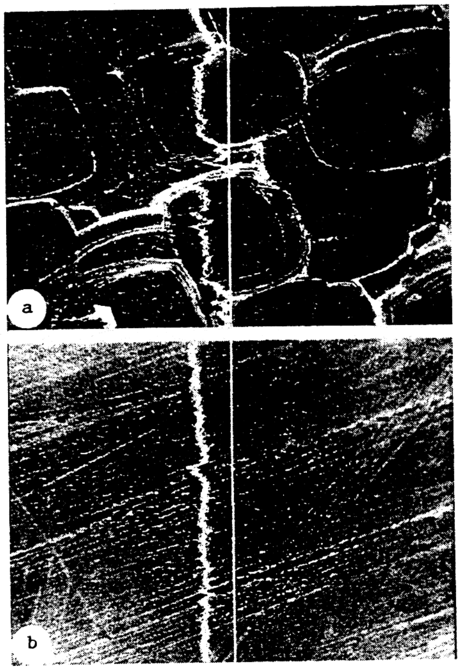

FIG. 1. Electron micrographs of surfaces of specimens III (a) and IV (b). × ~ 1000. Micrographs obtained in Hitachi H-500H electron microscope with scanning attachment operating in secondary electron regime.

the goniometer (Fig. 2). Thus there was a change in the angle Φ_0 of incidence of the x-ray beam on the crystal and detectors 2 and 3 registered the integral curves of TER and XDTER respectively. The fact that the specimen was established in the diffraction position did not affect the curve of specular reflection, since owing to the large vertical divergence of the incident x-ray beam (about 2°) only a very small (about 0.1%) part of the waves satisfied the diffraction condition.

To plot the differential curves of TER ahead of detector 2 (Fig. 2), we set up an additional silicon analyzer crystal in the (111) Bragg diffraction position.[14] The horizontal divergence of the incident beam, measured with its aid, was about 0.3'. The differential curves of TER were measured at Φ_0 = 3' (for specimen III) and Φ_0 = 6' (for specimens I, II, and IV) by rotating the analyzer crystal and detector 2 about the vertical axis of the principal goniometer.

Note that the use of the analyzer crystal also enabled us to increase the accuracy of registration of the position of the specimen, in which Φ_0 = 0', to ±0.1'.

The results of experimental investigation of specimens I-IV are shown in Fig. 3 and Table I.

3. DISCUSSION OF RESULTS

3.1. Method of taking account of roughness. Comparison of the curves for specimens III and IV with the curves for perfect crystal 1 in Fig. 3 demonstrates the identity of the influence of roughness on the form of the integral curves of specular reflection and diffraction of x rays in the conditions of TER. Large-scale roughness causes the same blurring of the curves in Fig. 3a-b, and small-scale roughness causes equal compression on the angular scale toward smaller angles.

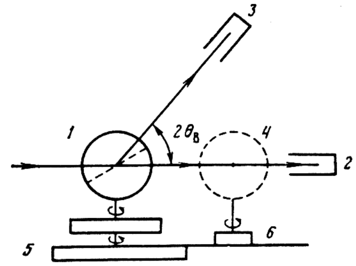

FIG. 2. Scheme of experimental apparatus for measurement of curves of specular reflection of x rays and diffraction in conditions of TER. 1) Specimen; 2, 3) detectors; 4) analyzer crystal; 5) main goniometer; 6) additional goniometer.

Comparative analysis of the intensities of the TER and XDTER curves reveals that roughness leads to a decrease in the coefficient of specular reflection both in the case of micro and macro surface relief, and also to a decrease in the coefficient of diffraction reflection from specimen IV with large-scale roughness. At the same time the intensity of the diffraction curve from specimen III is anomalously large (curve 3 in Fig. 3b is halved in intensity). However, most likely this anomaly is due not to roughness but to the presence of a residual mosaic layer on the surface of specimen III, which gives an increase in the integral coefficient of diffraction reflection.[15] Thus the influence of surface roughness on the integral intensity of TER and XDTER is also identical.

Table I contains the effective values of the critical angle of TER (Φ_c^e) and the mean angular scatter of the sections of the surfaces of the specimens ($\phi_{1/2}$), determined respectively from the integral and differential curves of x-ray TER. As one would expect, the least critical angle corresponds to specimen III with small-scale surface roughness, while the scatter of orientations is maximal for specimen IV with large-scale roughness. Therefore, as we verified, the changes in the TER and XDTER

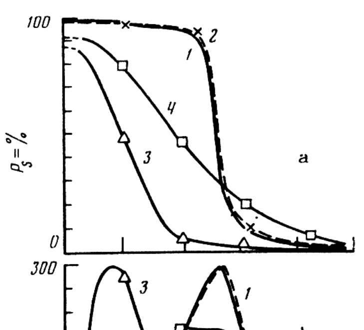

FIG. 3. Integral curves of TER (a) and XDTER (b) for specimens I-IV (curves 1-4 respectively). I) Intensity of diffraction reflection; P_S) coefficient of specular reflection. Solid curves, experimental; dashed, theoretical.

TABLE I. Surface Roughness Parameters of Test Specimens of Silicon, Determined from Integral and Differential Curves of TER of X-Rays

Characteristic, arcmin	Specimen No.			
	I	II	III	IV
Φ_c^e	13.2	13.2	5.5	10.2
Φ	0.7	0.7	6.0	9.8

curves under the action of surface irregularities are identical, so that the roughness characteristics obtained with the aid of TER can be used in processing the diffraction experiment.

So as to take account of fine-scale roughness, it is necessary to replace in the calculations parameters proportional to density of the test crystal by their effective values determined by the experimentally measured critical angle Φ_c^e. In place of the tabulated values of the polarizabilities χ_0 and χ_h we must substitute in the diffraction relations $\chi_0^e = (\Phi_c^e)^2$ and $\chi_h^e = \chi_h(\chi_0^e/\chi_0)$.

To take account of the large-scale roughness it is necessary to calculate the intensities of diffraction reflections from arbitrarily orientated parts of the crystal surface and then average them over the distribution of orientations found with the aid of TER.

Consider an arbitrary part of the surface with a small angle of deviation from the mean position δ and azimuth of deviation θ_p reckoned from the reciprocal lattice vector. It is easy to show that if diffraction on an area with mean orientation is characterized by the angles $\langle\Phi_0\rangle$, $\langle\Phi_h\rangle$, and $\langle\phi\rangle$, then for the given area these angles will have the values

$$\Phi_{0,h}=\langle\Phi_{0,h}\rangle+\delta\sin(\theta_n\pm\theta_p),\quad \varphi=\langle\varphi\rangle-\delta\cos\theta_p. \qquad (1)$$

Here Φ_0 is the angle of incidence, Φ_h is the angle of emergence of the diffracted wave, and ϕ is the angle of inclination of the reciprocal lattice vector to the surface (see Refs. 1-3).

Assuming that the parameters δ and θ_p are random variables with distribution function $G(\delta, \theta_p)$, we can represent the resulting diffracted intensity for reflection from a rough surface in the form

$$I(\langle\Phi_0\rangle,\langle\Phi_h\rangle,\langle\varphi\rangle)\sim\int_0^\infty\int_0^\infty d\Phi_0\,d\Phi_h P_h^s(\Phi_0,\Phi_h,\varphi)\,(\Phi_0/\langle\Phi_0\rangle)\,G(\delta,\theta_p). \qquad (2)$$

Here P_h^s is the coefficient of diffraction reflection from a plane surface, calculated, for example, in Ref. 2; $\Phi_0/\langle\Phi\rangle$ is a weighting factor corresponding to the increase in the flux of x-radiation arriving at the area with rise in the angle of incidence.

The function $G(\delta, \theta_p)$ is determined from the TER curves. In particular, if on the surface of the crystal there is no directed relief, then we can use $G(\delta,\theta_p)\sim\exp\{-(2\delta/\Phi_{1/2})^2\}$, determining $\Phi_{1/2}$ from the half-width of the differential curves.

3.2. Reliability of data obtained from crystals with perfect surfaces. As seen from Fig. 3a, the integral curves of specular reflection of x rays for a perfect crystal I and an ion-doped specimen II practically coincide with the theoretical curve of TER, calculated for a perfect crystal with an ideal surface. The maximum of the coefficient of specular reflection for specimens I and II is 97%, which also indicates the high quality of the surfaces of these specimens.

From Table I it follows that the corrections for surface roughness for specimens I and II do not exceed about 5% ($(\Phi_c^{id}-\Phi_c^e)/\Phi_c^{id}\sim 0.7\%$, $\Phi_{1/2}/\Phi_c^{id}\sim 5\%$, where $\Phi_c^{id} = 13.3'$ for crystals of silicon). At the same time the diffraction curve for specimen II differs from the curve for specimen I and the theoretical curve for a perfect crystal (Fig. 3b). The change in the curve from specimen II (sharp decrease in intensity toward small Φ_0) does not coincide with the changes observed on the curves from specimens III and IV, and consequently is not related to the roughness. However, the curve from specimen II is well interpreted with the aid of the model of a surface amorphous layer of thickness L_{am} = 15 nm and partial amorphization of the specimen distributed to a depth of L_{am}^{eff} = 100 nm. The presence of an amorphous layer and a transitional, partly amorphized, region is confirmed by electrophysical and electron-microscope measurements, and the parameters L_{am} and L_{am}^{eff} are correlated with the results of these measurements, and also with the theoretical length of the mean projected flight of the thallium ions with energy 100 eV in the silicon.[16]

Similarly from the curve of XDTER from specimen I we determine the presence of an amorphous surface layer about 2 nm thick. However, in this case the effect is less marked, and therefore for more precise measurement of the thickness we must take account of the surface roughness.

Thus, analyzing the results in Refs. 1-3, we can conclude that surface roughness does not play an important part in experiments on x-ray diffraction in the conditions of TER, made on standard semiconducting crystals, and the obtained information on layers 2-100 nm thick is reliable.

This conclusion does not mean that the surface relief of the test crystals has a depth less than 2 nm. Here we have to do with a problem which has been more than once noted in investigations of TER and Bragg diffraction of x rays (see Refs. 4-10, 17, 18). Owing to the small amplitude of interaction with matter, x-ray beams as it were "smooth" the surface, i.e., they give a low estimate of the roughness and averaged information along the surface of the crystal. For example, in Ref. 7 it was experimentally found that TER of x rays with specular reflection coefficient 97% is observed from a surface with irregularities of depth about 20 nm, although according to the criteria of visible-light optics TER of x rays from such a surface is impossible. The problem requires detailed theoretical analysis.

In conclusion we may again emphasize that, as shown by the experimental results, roughness of the surfaces of standard semiconducting crystals with treatment quality of the 14th class has little influence on the optics of glancing x rays, and can be neglected.

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