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Determination of surface parameters of solids by methods of X-ray total external reflection

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Abstract. The series of GaAs and SiO_2 samples with the specially prepared one- and two-dimensional surface reliefs have been investigated by the methods of integral and differential curve total external reflection of X-rays. The direct and inverse problem was solved, taking into consideration data obtained by the method of atomic-force microscopy: the theoretical curves of total external reflection are calculated and parameters describing a surface relief of the samples are restored.

Keywords: total external reflection of X-rays, surface relief, rocking curves, fractals.

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1. Introduction

The methods based on the phenomenon of X-ray total external reflection (TER) allow one to investigate surfaces with values of average deviation of roughness R_a<10 nm [1]. The high resolution of the TER methods is achieved due to angles of grazing incidence and short X-ray wavelength (0.1–0.3 nm). Surface parameters are determined as a result of solution of the inverse problem by TER methods using experimental reflection curves. Taking into consideration the ambiguousness of this solution, there is a question of result correctness in the TER methods. The purpose of this work is to develop new approaches to investigation of surfaces by methods of integral curves (IC) and differential curves (DC) in the case of TER [2] using data obtained by the method of atomic-force microscopy (AFM) [3].

2. Experimental details

In the IC TER method, the integrated signal intensity reflected from a sample and a part of a primary incident beam is registered by a detector. The sample rotated around the vertical axes of a goniometer with the θ angle range from 0 to ≈ 0.5° with respect to the direction of the primary beam.

In the DC TER method, the crystal - analyser is used for the determination of angular intensity distribution in the reflected beam satisfying the Bragg condition. Rocking curves were analysed in dependence on the angle α in this method. To research the surface relief with oriented roughness, the sample was turned in vertical plane by an angle ν.

Surface studies by TER methods were carried out using specially developed hardware and software automated complex for measurements based on the X-ray diffractometer DRON-3M [4]. Experimental conditions were as follows: X-ray tube BSV25-Cu, silicon monochromator and analyser, reflection (220) of $CuK_{\alpha 1}$-radiation.

3. Theory

The approximation of geometric X-ray optics was used for the theoretical analysis of our experimental results. In this approximation, the surface profile of a sample was divided into flat areas (microparts) sloped under an angle γ to the medial line of the profile. The Fresnel formulae were used [5] for determination of X-ray reflection coefficient I_f on the surface microrelief. The beam scattering on the surface roughness was described by distribution function of slopes $F_r(\gamma)$, giving probability of micropart slopes under the angle γ.

IC TER method

The normalised intensity of beams reflected from a sample (I_K) and those reaching the detector (I_P) are described by expressions:

$$I_K(L_K,\theta) = 2\int_0^{L_K\theta/2} I_1(y_0)dy_0 \int_{\gamma_{\min}}^{\gamma_{\max}} I_f(\theta+\gamma)F_r(\gamma) * I_1(\eta)d\gamma\,, \quad (1)$$

$$I_p(L_K,\theta) = I_0 - \int_0^{L_K\theta/2} I_1(y_0)dy_0\,, \quad (2)$$

where L_K is the length of the sample; I_0 is the half of the primary beam intensity. The function $I_1(y_0)$ describes space distribution of the primary beam intensity. The angular scattering of the primary beam are taken into account by the convolution $F_r(\gamma)$ with function $I_1(\gamma)$. Thus, integrated intensity of rays reaching the detector is given by:

$$I_D(L_K,\theta) = I_K(L_K,\theta) + Ip(L_K,\theta)\,. \quad (3)$$

DC TER method

The intensity reflected from the analyser is described by the formula

$$I_A(\theta,\alpha) = \int_{\alpha_{\min}}^{\alpha_{\max}} I_f(\alpha/2)F_r(\alpha/2-\theta) * G(\sigma_\alpha)d\alpha\,, \quad (4)$$

where $G(\sigma_a)$ is the normally disrtibuted function describing a rocking curve obtained from the analyser without sample. The convolution $G(\sigma_a)$ with a distribution function of slopes $F_r(\gamma)$ takes into account angular and spectral width of the primary beam, dispersion and imperfection of the analyser for the rays scattering by the analyser.

4. Experimental results and discussion

The samples GaAs#1-#3 with the specially treated one- and two-dimensional surface relief and the samples SiO_2#1-#3 that were put through the superthin chemical and mechanical treatment have been investigated.

Analysis of relief parameters obtained by AFM method

Parameters of sample profiles defined by the AFM method are represented in Table 1, where: R_a is the average deviation of profile heights from the base plane, s_y is the root mean square of profile heights; C_m is a maximum amplitude of profile harmonics; T_m is a period of profile harmonics with maximum amplitude; C_v is a relative amplitude of profile harmonics (it is equal to ratio of C_m and sum of amplitudes of all harmonics). The profile was decomposed by the Fourier series on the space frequencies for the determination of parameters C_m, C_v, T_m.

Analysis of the experimental IC and DC TER

The integral and differential curves TER for the samples GaAs and SiO_2 are shown in Fig. 1 and Fig. 2. DC were investigated at various angles h of the sample in the vertical plane for the definition of a primary orientation of relief roughness, and the value of relief asymmetry A_P was analysed:

$$A_p = B_v / B_{v+90°}\,, \quad (5)$$

where B is the halfwidth of the DC. At perpendicular orientation of relief roughness to the direction of the primary beam (Fig. 2a) the maximum value A_P (angle $\eta = 0$) is obtained, and the parallel orientation of roughness (Fig. 2b) is responded by angle $\eta = 90°$.

The asymmetry A_K of the experimental DC and also fractal dimension D_f were defined for the entire analysis. The asymmetry of the curve A_K was determined as a ratio

Table 1. Profile parameters for samples GaAs, obtained by the AFM method.

Samples	R_a, nm	σ_y, nm	C_m, nm	T_m, µm	C_v
#1	2.411	2.875	2.108	3.568	0.125
#2	4.390	5.544	2.847	0.762	0.058
#3	2.984	3.799	2.554	11.665	0.097

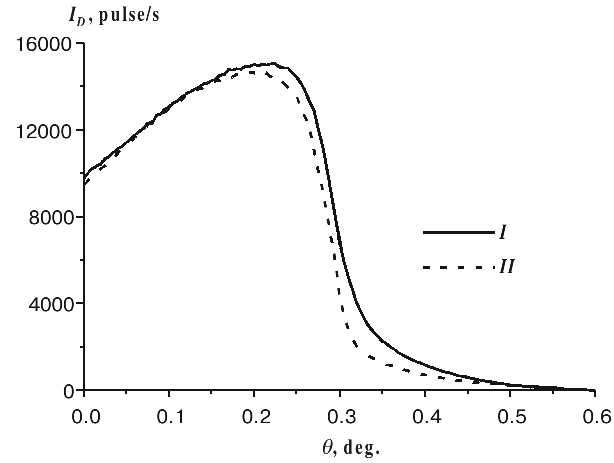

Fig. 1. Experimental IC TER for GaAs#1: I – ν = 0°, II – ν = 90°.

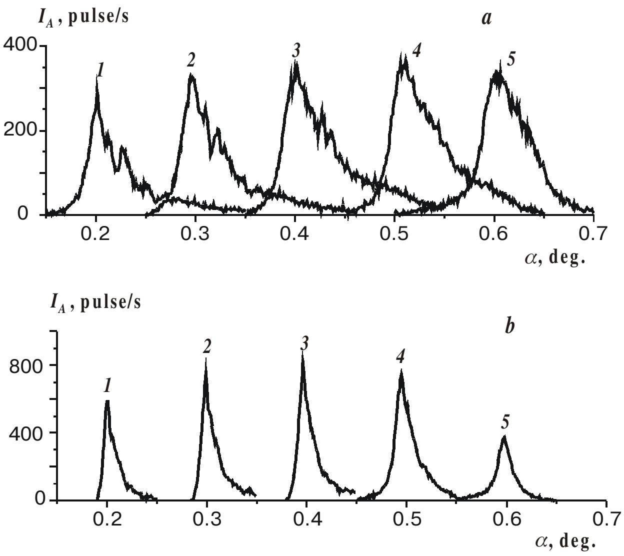

Fig. 2. Experimental DC TER for GaAs#1: a) ν = 0°; b) ν = 90°: *1* – θ = 0.1°; *2* – θ = 0.15°; *3* – θ = 0.2°; *4* – θ = 0.25°; *5* – θ = 0.3°.

of square of the DC at $\alpha > 2\theta$ to the square of the DC at $\alpha < 2\theta$. Fractal dimension D_f [6] describing complexity of the form of curve was measured by the method of the triangle length [7]. The obtained values D_f of the DC, namely the average value D_f on θ (Fig. 3), are used at calculation of maximum amplitude of the profile harmonics C_m. The values D_f were not taken into account at small angles ($\theta < 0.1°$) because of distortion D_f caused by shadowing microparts of the profile, and at large angles ($\theta > 0.3°$) – by absorption.

The set of values $B(\theta)$, $A_K(\theta)$, $D_f(\theta)$ describing parameters of the studied surface was relief obtained for each sample from experimental DC. The existing data correlation of the DC TER and AFM methods allows to express parameters of the surface profile through performances of the DC using the empirical formulae. When fitting parameters of the empirical formulae, the sum of squared differences for parameters of calculated profiles and parameter of profiles obtained by AFM method was minimised.

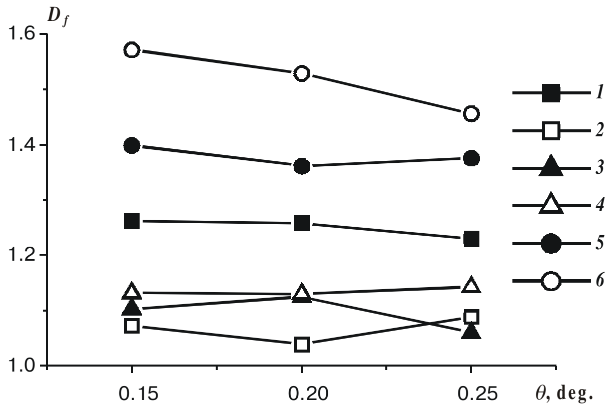

Fig. 3. Fractal dimension D_f of the DC TER for samples GaAs#1-#3: *1* – #1, ν = 0°; *2* – #1, ν = 90°; *3* – #2, ν = 0°; *4* – #2, ν = 90°; *5* – #3, ν = 0°; *6* – #3, ν = 90°.

The experimental dependences of the halfwidth $B(q)$ in Fig. 4a are satisfactorily approximated directly with the slope coefficient b_1. Based on AFM data, the correlation between the value of the slope coefficient b_1 and the period of profile harmonics T_m in Fig. 4b was described by the following empirical relation:

$$T_m(b_1) = k_1 \cdot 2^{b_1} + g_1 \exp\left(\frac{-(b_1 - m_1)}{2d_1}\right) + g_2 \exp\left(\frac{-(b_1 - m_2)}{2d_2}\right), \qquad (6)$$

where k_1, g_1, g_2 are weight factors equal to 0.48; 6.0; 5.16, respectively; m_1, m_2 are mathematical representations for the Gauss function equal to 1.07 and 3.23, respectively; d_1, d_2 are dispersions equal to 1.15 and 0.92, respectively.

The values of asymmetry $A_K(\theta)$ of the DC (Fig. 4c) at magnification θ sequentially come nearer up to some magnitude at $\theta = 0.25°$. It enabled us to take the value A_K at a given θ into account for the analysis. The correlation between values A_V and relative amplitude of harmonics profile C_V was found (Fig. 4d) when passing from the coefficient of asymmetry A_K to the parameter of relative asymmetry A_V. It behaves in accord with the following rule: $A_V = A_K$, at $A_K > 1$; $A_V = \sqrt{1/A_K}$, at $A_K < 1$. Then, in view of AFM data, the dependence $C_V(A_V)$ is described by the expression

$$C_V = c_1 + c_2 A_V , \qquad (7)$$

where c_1 = 0.0092; c_2 = 0.047.

Using the data of DC TER and AFM methods, the value of mean deviation of the profile roughness R_a was expressed by the empirical formula:

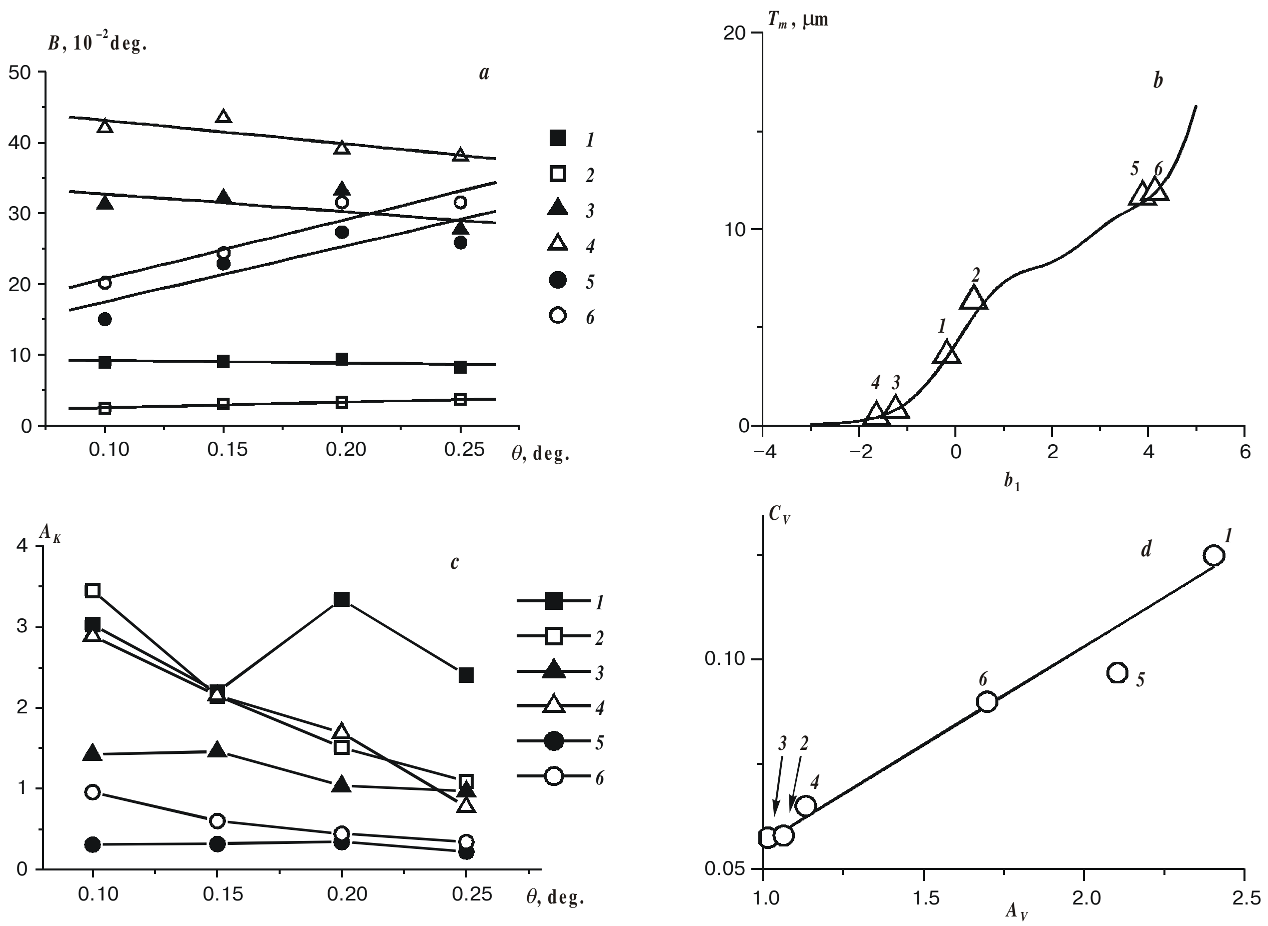

Fig. 4. Correlation of DC parameters and profiles for samples GaAs#1-#3: *a* – slope dependence $B(\theta)$ DC; *b* – correlation slope b_1 of halfwidth of the DC and the period of the profile harmonics T_m; *c* – $A_K(\theta)$ asymmetry of the DC dependence; *d* – correlation of relative asymmetry A_V of the DC and relative amplitude of the profile C_V: *1* – #1, $v = 0°$; *2* – #1, $v = 90°$; *3* – #2, $v = 0°$; *4* – #2, $v = 90°$; *5* – #3, $v = 0°$; *6* – #3, $v = 90°$.

$$R_a = r_0 \cdot B \cdot T_m \frac{1}{\mu_s C_V}, \tag{8}$$

where $r_0 = 9.2 \cdot 10^{-4}$; B is the mean value of DC halfwidth. The scale coefficient μ_s intended for correction of R_a values can be expressed by the formula:

$$\mu_s = 1 - \frac{1}{\exp[(T_m - \Lambda\mu_1)/(\Lambda\mu_2)] + 1}\mu_3, \tag{9}$$

where $\mu_1 = 0.42$; $\mu_2 = 0.057$; $\mu_3 = 0.922$, Λ is the thickness of layer of half slacking of the X-ray beam.

In the case of a sinusoidal profile, the surface amplitude of space harmonics is $C_m = (\pi/2)R_a$. Generally maximum amplitude of harmonics for the real profile is described by the expression:

$$C_m = \frac{\pi}{2} \cdot R_a (C_V)^{p_1} (D_f)^{p_2}, \tag{10}$$

where $p_1 = 0.38$; $p_2 = 0.9$.

Calculation of the IC and DC on the base of AFM data (direct task)

The integral (Fig. 5a) and differential (Fig. 5b) curves of the TER were calculated on the base of profile data obtained by AFM method. The correction of the surface profile and distribution function of slopes $F_r(\gamma)$ was carried out for matching the theoretical and experimental curves. The profile was multiplied by the scale coefficient μ_s in the course of the correction. The correction of the distribution function of slopes $F_r(\gamma)$ took into account repeated reflections from the surface, asymmetry of the DC and diffusion of DC form in dependence on the angle θ.

Determination of surface parameters on the base of IC TER

The following parameters of sample surface were determined from experimental IC: the critical angle θ_{CE} and averaged deviation R_a (Tab. 2). Taking into account the theoretical density of the sample material ρ, theoretical θ_C and experimental θ_{CE} value of the critical angle, the

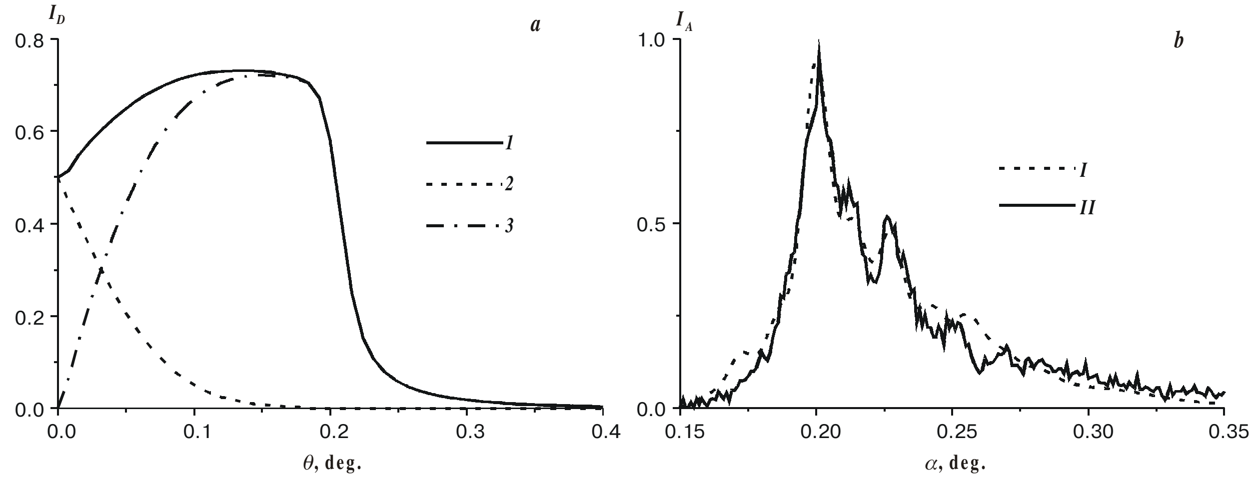

Fig. 5. Calculation of IC and DC: *a* – theoretical IC for SiO_2#1 at $\nu = 0°$: *1* – full intensity; *2* – intensity of a beam which passes through the sample; *3* – intensity reflected from the sample; *b* – DC for GaAs#1 at $\nu = 0°$: I – theoretical DC on the base of AFM profile; II – experimental DC.

Table 2. Surface parameters of samples, obtained by the IC, DC and AFM methods.

	ν,°	θ_{CE},°	ρ, g/cm^3	$\Delta\rho$,%	R_a, nm			T_m, μm	
					AFM	IC	DC	AFM	DC
GaAs*		0.3051	5.001						
#1	0	0.300	4.8352	–3.32	2.411	2.40	2.48	3.57	3.47
#1	90	0.2997	4.8255	–3.51	1.43	1.73	1.87	6.35	5.56
#2	0	0.3003	4.8449	–3.13	4.39	4.99	5.01	0.76	0.79
#2	90	0.2994	4.8159	–3.70	3.56	4.24	3.05	0.42	0.40
#3	0	0.2988	4.7966	–4.09	2.984	3.94	2.44	11.67	11.38
#3	90	0.2986	4.7902	–4.22	3.24	3.80	3.33	11.91	11.85
SiO_2*		0.236	2.65						
#1	0	0.2276	2.4647	–6.99	0.263	0.31	0.34	0.81	0.80
#2	0	0.2348	2.6231	–1.01	0.54	0.71	0.62	0.69	0.72
#3	0	0.2342	2.6097	–1.52	1.851	1.37	1.13	0.83	0.81

GaAs*, SiO_2* – theoretical values for GaAs and SiO_2, respectively.

density of the surface layer can be described by the following expression:

$$\rho_E = \rho\left(\frac{\theta_{CE}}{\theta_C}\right)^2 . \qquad (11)$$

The minor diminution of density of the surface layer (Tab. 2) was revealed for the majority samples, that is satisfactory agreed with data of other researches [8].

Determination of surface parameters on the base of DC TER

The fractal approach is effective for describing the relief and profile of the real surface in many cases [9]. Therefore, the fractal profile representing a sum of 8 sinusoids was used to solve the inverse problem in DC TER method. Amplitudes and periods of harmonics were set as follows [6]:

$$A_1 = C_m;\ T_1 = T_m;\ A_n = A_{n-1}V_A;\ T_n = T_{n-1}V_T , \qquad (12)$$

where $V_A = 0.67$, $V_T = 0.65$.

Fitting the theoretical DC to the experimental one was made after representation of entry conditions. The amplitudes of the profile harmonics were used as fitting parameters. During the fitting process, it was possible to receive a good agreement of the calculated profile (Fig. 6b) and the profile restored by AFM method (Fig. 6a). The basic parameters of the sample profiles obtained by the AFM and DC TER methods are represented in Tab. 2. The fragment of the surface relief of the sample (Figs 6c,d) is restored using profile parameters obtained by DC method.

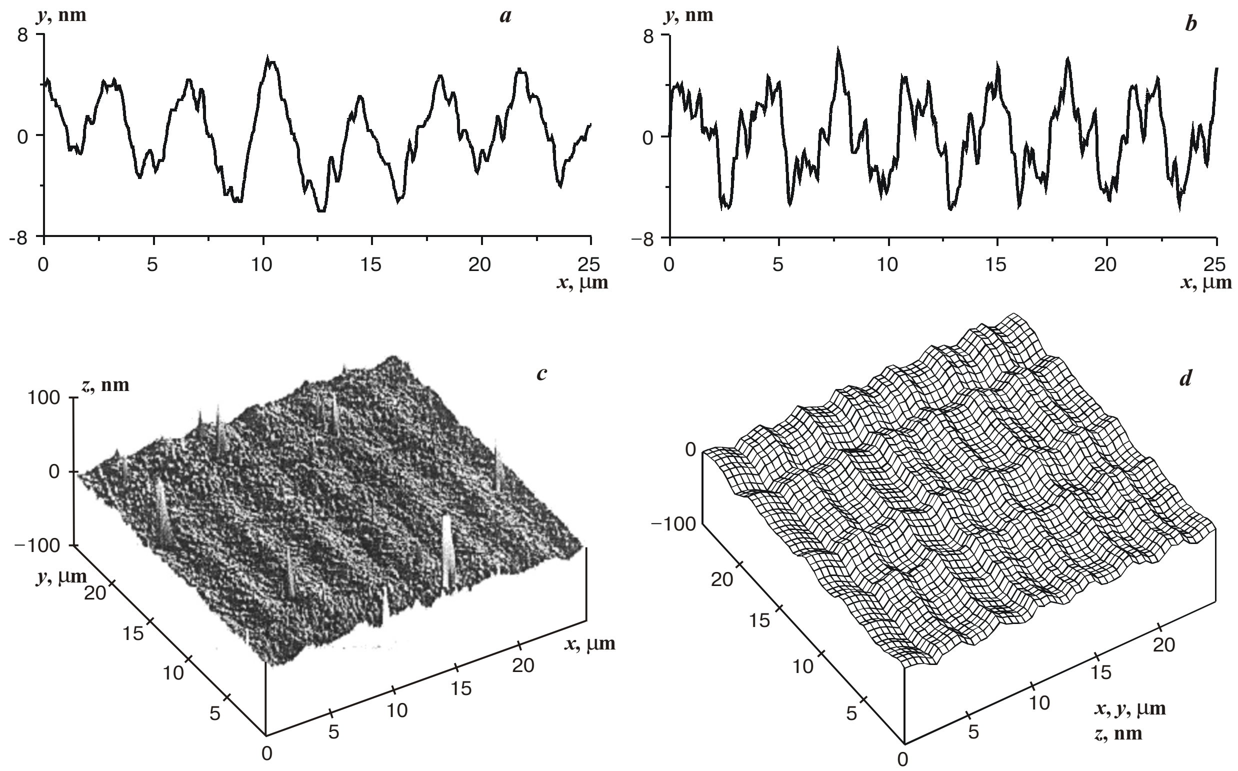

Fig. 6. Sample GaAs#1: *a*, *b* – profiles obtained by the AFM and DC TER methods; *c*, *d* – relief, obtained by the AFM and DC TER methods.

5. Conclusions

Thus, using DC TER method, it is obviously possible to restore the relief parameters for researched samples. The relief parameters and the density of the surface layer of the samples were determined using IC TER method. The good agreement of the surface relief parameters obtained by the TER and AFM methods is reached as a result of correction of the parameters describing IC and DC TER.

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