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Ultrasoft X-ray Spectroscopy Investigation of the Model System Si–SiO_2

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Abstract—SiO_2 surface films with different thicknesses (ranging from 20 to 630 Å), grown on a crystal silicon substrate, have been investigated by the method of reflection and scattering of ultrashoft X-rays. It is shown on the basis of a simultaneous analysis of the Si$L_{2,3}$ reflection spectra and the scattering indicatrix that the critical angle θ_c for total external reflection for SiO_2 at $\lambda = 57$ Å lies in the range 4.5°–5°. The angular dependence of the thickness of the surface layer that forms the specular reflection is obtained. It is shown that the surface layer, whose thickness corresponds to the penetration depth of the radiation into the material with glancing angle close to the critical value θ_c, plays a large role in the formation of the anomalous scattering peak (Yoneda peak).

Ultrasoft X-ray spectroscopy is a promising experimental method for investigating the atomic and electronic structure of matter [1]. The small penetration depth of the radiation in matter (tens–hundreds of angstroms) and the local character of the photon absorption process in a specific atom in a multiatomic system make this method surface-sensitive [2, 3].

The purpose of the present work is to determine the depth of the near-surface layer of matter forming the reflected and scattered radiation in a two-phase system (surface layer + substrate) in the ultrasoft X-ray range. The system Si–SiO_2 with various thicknesses of the SiO_2 surface film, ranging from 20 to 630 Å, was chosen as such a system. This choice is due not only to the existence of details, differing in type and energy position, of the fine structure of the Si$L_{2,3}$ reflection spectra for crystalline Si and SiO_2 but also the well-known importance of this system in the production of various objects in the microelectronic industry.

All investigations were performed on an RSM-500 spectrometer-monochromator in a special camera attachment [4] using the bremsstrahlung and characteristic radiations from a tungsten anode. The energy resolution of the details of the spectra in the region of the Si$L_{2,3}$ ionization threshold was $\Delta E \simeq 0.3$ eV, and the angular resolution of the scattering indicatrix was 1.2°. The radiation was detected with a VEU-6 channel-type secondary-electron multiplier with a CsI photocathode. Wafers of a KDB (111) 4° silicon single crystal, which were 380 μm thick and 30 mm in diameter and were oxidized in a dry-oxygen atmosphere at $T = 1050$°C, were investigated. The thickness of the surface SiO_2 films grown was monitored using ellipsometry on an LÉF-2 apparatus.

Figure 1 shows the experimental Si$L_{2,3}$ reflection spectra for various SiO_2 thicknesses on a silicon substrate for various glancing angles θ_0 of the radiation. All systems were studied in a wide range of angles $4° \le \theta_0 \le 16°$, i.e., inside and outside the region of total external reflection of X-rays. The spectra presented are normalized to the maximum B (≃108.9 eV), with the exception of the spectrum for a system with a 20 Å thick surface film. We note that the fine structure of the reflection spectrum of crystalline Si is characterized by the presence of the details *a*–*g* (energy range 100–104 eV), while the spectrum of crystalline SiO_2 is characterized by the details *A*–*C* (105–120 eV). Thus, the presence of only one group of details in the spectrum corresponds to the case where the reflected radiation is formed by a single-phase system. The manifestation of details of both groups in the spectrum attests to reflection from a substrate + surface layer system. The fact that the contribution of the components of this system to the reflection is different should cause the corresponding group of details in the spectrum to dominate. It is natural to compare the thickness of the film for which details characteristic for a silicon spectrum, specifically, the bands *b* and *d*–*f*, appear in the reflection spectrum with the corresponding value of *d*—the penetration depth of the radiation in the material. Thus, for an Si–SiO_2 system with different thicknesses of the surface film, details characteristic of the substrate spectrum are present in the spectra for $\theta_0 = 4°$ right up to a thickness of 85 Å, and for glancing angles $\theta_0 = 8°$, 12°, and 16° they are present up to thicknesses 140, 190, and 260 Å, respectively. We also note the observed correlation of the spectra for different thicknesses of the surface layer but for different glancing angles. For example, the relative intensity distribution for the spectrum for a 85 Å thick film for $\theta_0 = 4°$ is virtually identical to that of the spectrum obtained for a 260 Å thick film for $\theta_0 = 16°$. This shows that the relative contributions of the surface

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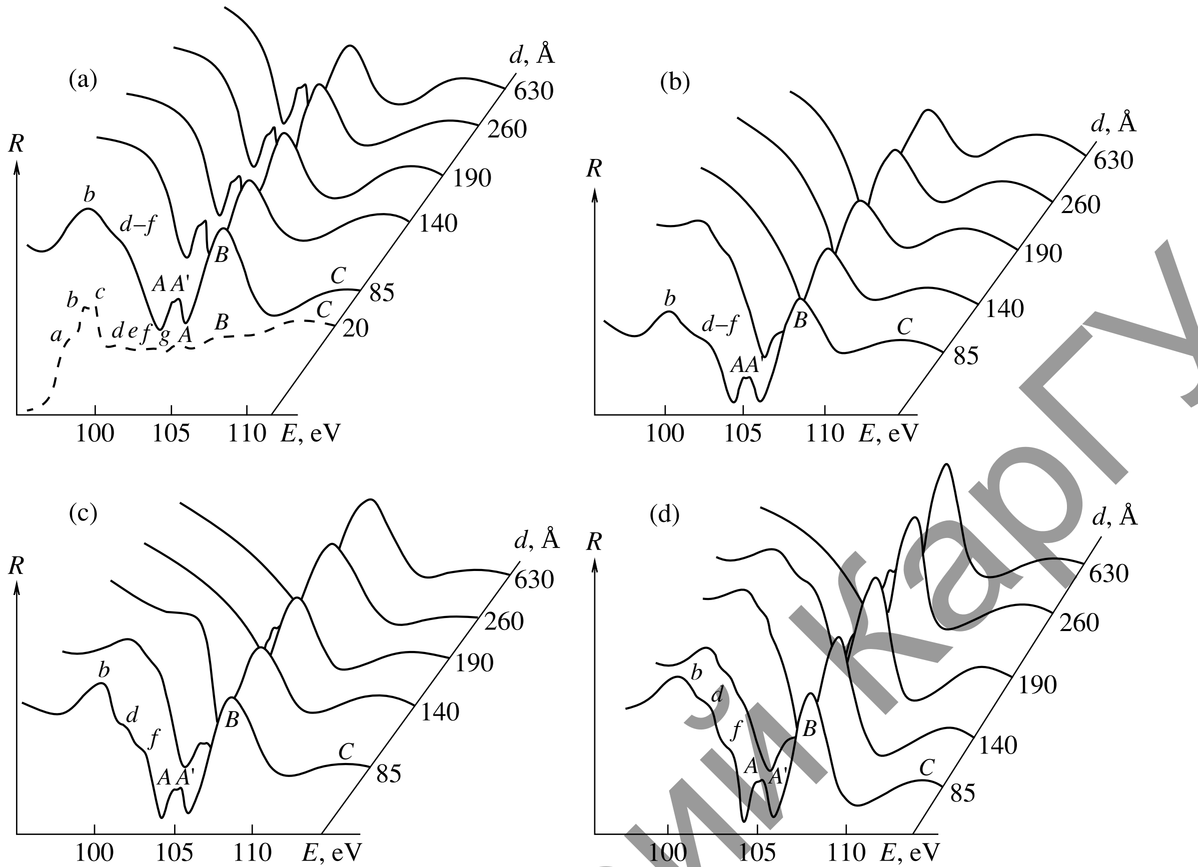

Fig. 1. Si$L_{2,3}$ reflection spectra for the system Si–SiO_2: θ_0 = (a) 4°; (b) 8°; (c) 12°; (d) 16°; *d* is thickness of the surface SiO_2 film.

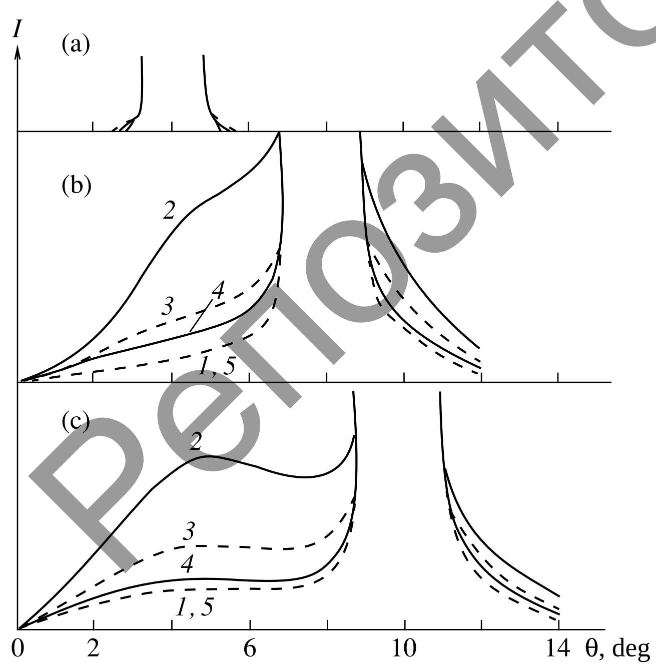

Fig. 2. Scattering indicatrix for the system Si–SiO_2 for λ = 57 Å for SiO_2 surface films with various thicknesses ((*1*) 20; (*2*) 85; (*3*) 140; (*4*) 190; (*5*) 630 Å): θ_0 = (a) 4°; (b) 8°; (c) 10°.

film and substrate to the reflected radiation are identical in these spectra.

The angular distribution of the scattered radiation (scattering indicatrix) from the surfaces of two-phase systems was also studied in this work. It is known [5, 6] that for glancing angles less than the critical angle θ_c, i.e., in the region of total external reflection, the scattering indicatrix carries information primarily about the roughness of the surface. In the range of angles $\theta_0 > \theta_c$, the characteristic features of the atomic spectra of the near-surface regions of materials can be judged from the form of the scattering indicatrix. For the samples investigated with different thicknesses of the surface film, it was found that in the region of total external reflection (θ_0 = 4°) the forms of the phase functions are essentially identical (Fig. 2a). Outside the region of total external reflection (θ_0 = 8°, 10°) an anomalous scattering peak appears in the scattering indicatrix (Yoneda peak) (Figs. 2b, 2c). In the figures presented, the detected radiation intensity was normalized to the intensity of the specularly reflected radiation. As one can see, the angular position of the Yoneda peak lies in the range 4.5°–5°, and the relative intensity is substantially different for different samples. The angular position of the anomalous scattering peak corresponds to

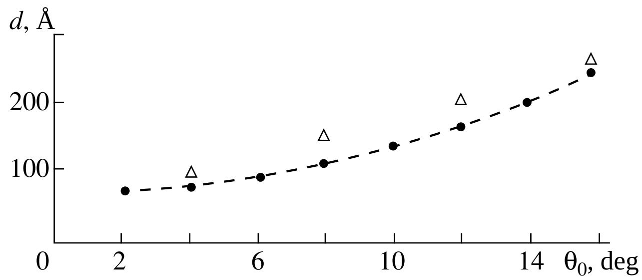

Fig. 3. Angular dependence of the formation depth of a specularly reflected beam in SiO_2: dots—calculation, triangles—experiment.

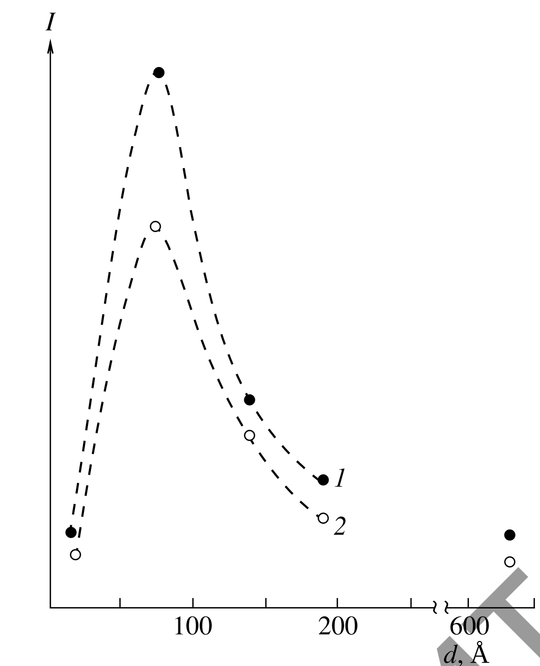

Fig. 4. Relative intensity of the anomalous scattering peak for SiO_2 surface films ($\lambda = 57$ Å) with different thicknesses: θ_0 = (*1*) 8°, (*2*) 10°.

the value of the critical angle θ_c [5], so that the critical angle for SiO_2 for $\lambda = 57$ Å can be assumed to fall within the indicated limits.

The angular dependence of the thickness of the surface layer forming the specularly reflected radiation in silicon dioxide can be constructed on the basis of the spectra obtained. This dependence is displayed in Fig. 3 together with the computed curve. The calculation was performed using the formula [7]

$$d = \frac{\lambda}{2\sqrt{2\pi}}$$

$$\times\left[\sqrt{(\sin^2\theta_c - \sin^2\theta_0)^2 + \gamma^2} + \sin^2\theta_c - \sin^2\theta_0\right]^{-1/2},$$

where $\gamma = (\lambda/2\pi)\mu$, for the following values of the parameters: $\theta_c = 9°$, $\lambda = 117.4$ Å, and $\mu = 10^5$ cm^{-1}. The value of the critical angle was chosen on the basis of the fact that it is directly proportional to the wavelength of the radiation [8]. It is obvious that the theoretical and experimental curves agree well with one another in the entire range of comparison.

Figure 4 displays the dependence of the relative intensity of the Yoneda peak on the thickness of the SiO_2 film. As one can see, this dependence is distinctly nonmonotonic, and the maximum corresponds to a layer thickness of 85 Å. As inferred in [5], the largest relative intensity of the Yoneda peak can appear in cases where the vacuum-material boundary possesses a so-called transitional layer, i.e., the spatial structure of the material changes in a certain layer near the surface. The fact that the intensity of the Yoneda peak is greatest for $d = 85$ Å indicates that the largest structural changes occur in the layer forming this peak. On the other hand, the penetration depth of the radiation in the material for $\theta_0 = 4°–5°$ is close to $d = 85$ Å. It can be asserted on this basis that primarily a layer whose thickness corresponds to the penetration depth of the radiation in the material for $\theta_0 = \theta_c$ (in this case $\theta_c = 4.5°–5°$, $d = 60–80$ Å) influences the relative intensity of the anomalous scattering peak.

In summary, in the present work the thickness of the surface layer forming the specularly reflected radiation in silicon dioxide was determined for various glancing angles θ_0 (4°–16°) on the basis of experimental Si$L_{2,3}$ spectra for the system Si–SiO_2. The distinct nonmonotonic character established for the relative intensity of the Yoneda peak in the scattering indicatrix as a function of the thickness of the SiO_2 surface layer shows that the layer of matter corresponding to the radiation penetration depth in matter for $\theta_0 = \theta_c$ has the dominant effect on the form of this peak. This circumstance could make it possible to extract additional information when studying the characteristics of radiation scattered from the surface of various substances.

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