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Diffuse scattering of neutrons in thermally oxidized silicon

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Abstract

Diffuse scattering of thermal neutrons in thermally oxidized silicon was studied using the triple-crystal diffraction method. The observed scattering shows a strong asymmetry characteristic for the vacancy type of defects. The mean size of the distorted region around these lattice defects is determined as (220 ± 20) nm. The diffuse scattering can be connected with the growth of the stacking faults in the oxidized silicon.

Keywords: Diffuse neutron scattering; Lattice defects; Silicon; Triple-crystal diffractometer

1. Introduction

Properties and reliability of microelectronic devices are influenced by the real structure (crystal lattice defects) of the material. Point defect clusters like oxygen precipitates may have detrimental effects on a device if they grow during a thermal treatment into its electronically active region. However, if they grow into non-active regions they can have the desirable effect of an internal impurity gettering. Therefore, the study of the defect growth during the treatment of semiconductor materials both in the surface region and in the bulk of a crystal is of essential importance for the device technology.

The diffuse scattering of X-rays in as-grown and thermally treated silicon single crystals was studied in a number of papers. Double-crystal [1] and triple-crystal [2] diffraction techniques were used. Scattering caused by growth defects [3], oxygen clusters [1], and dislocation loops [4] was observed. From the study of the diffuse scattering, information can be obtained for the mean size and the symmetry of defects and the predominant sign of the internal strain caused by the defects [5].

However, due to the high absorption the X-ray diffraction is not effective for a study of defects in the bulk of the crystal. Therefore, other types of radiation with a higher penetration are used for the investigation of imperfections of silicon, too. The diffuse scattering of γ-rays was used to search different kinds of defects in Si, e.g. irradiation-induced defects after the heat treatment [6]. The depth profile of the defect structure was studied and an alteration of the strain sign was observed.

The use of a neutron triple-crystal diffractometer for diffuse scattering studies was proposed in [7].

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In [8] the high sensitivity of neutron scattering to the defects in the volume of the crystal was shown by comparing the data obtained with X-rays and neutrons from the same crystal. The extinction (or Pendellösung) length characterizes the material depth for the formation of *coherent wave fields* in the crystal. For neutrons it is usually in the order of 100 μm at a Bragg reflection and, therefore, about one order of magnitude larger than that for X-rays. This provides the possibility of studying defects within a larger size range and/or a smaller value of stress than it could be achieved with X-rays. The very low absorption of neutrons in silicon ensures that the whole volume of the crystal can contribute to the *diffuse scattering* intensity. In the present paper a study of the diffuse scattering from thermally oxidized silicon is reported.

2. Theoretical considerations

According to the theory of the diffuse scattering [5] the angular dependence of the intensity of the diffuse peak could reveal the characteristics of the defects of the crystal. Here, we consider the region near the Bragg peak, where the deviation $\boldsymbol{q}$ from the Bragg position is small compared to the reciprocal lattice vector $\boldsymbol{h}$; $\boldsymbol{q}$ is the difference between the scattering vector $\boldsymbol{K}$ (momentum transfer vector of the incident to the scattered neutron beam at the sample) and the reciprocal lattice vector $\boldsymbol{h}(q = |\boldsymbol{q}| = 4\pi \sin(\theta_B)\sin(\theta)/\lambda$ with θ being the angle between the momentum transfer vector and the near-by reciprocal lattice vector and λ the wavelength of the neutron beam). This is the so-called Huang region [5] of the diffuse scattering.

The region of the diffuse scattering around the reciprocal lattice point is extended in all three dimensions. In double- and triple-crystal experiments an intensity integrated over the direction perpendicular to the scattering plane is measured because the vertical divergency is greater than the extension of the diffuse scattering range. In the following the intensity integrated over a direction perpendicular to the exit wave vector $\boldsymbol{K}_h$ (in the scattering plane) is considered. Then only the scattering vector component q_0 parallel to $\boldsymbol{K}_h$ is varied in the scattering experiment. Because $\theta \ll \theta_B$ $q_0 \approx q\cos(\theta_B)$, where

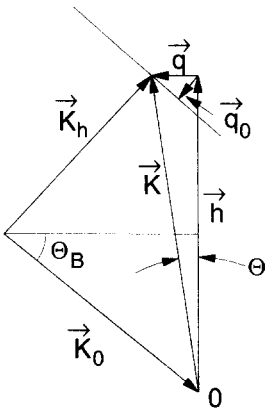

Fig. 1. Vector diagram of scattering in the vicinity of the reciprocal lattice point of the sample. In the real case $\theta \ll \theta_B$.

θ_B is the Bragg angle at the sample. In Fig. 1 the vector diagram of scattering in the vicinity of the reciprocal lattice point of the sample is shown.

According to theoretical considerations given by Dederichs [5], the symmetric part of the integrated diffuse intensity $I_s(q_0)$ in the region of small q_0 ($q_0 < 1/R_0$, R_0 is the mean defect radius) can be expressed by

$$I_s(q_0) = A \cdot \ln(e^{1/2}/q_0 R_0).$$

The q_0-axis intercept of the linear part of the plot of $I_s(q_0)$ versus $\ln(q_0)$ determines the mean radius of the defect. The asymmetric part of the diffuse intensity gives information about the sign of the strain field around the defects and so it is possible to distinguish between either interstitial type or vacancy type of defects.

In the vicinity of the Bragg position the intensity scattered from a nearly perfect crystal (a crystal with a small content of defects) consists generally of different components: coherent intensity produced by the wave field in the crystal and diffuse intensity caused by the scattering on defects of different kinds like thermal vibrations, defect clusters, surface roughness, etc. Whereas the direction of the coherently scattered beam varies with the sample orientation (in a small range near the Bragg angle) according to the dynamical theory, the diffuse intensity is concentrated near the angular position of a kinematically diffracted beam. In order to separate these components of the scattered beam

an analyzer crystal can be used. Then the diffuse and coherent scattering (main peak) components occur in the rocking curve of the analyzer as separated peaks together with the pseudo peak (for the terminology see [4, 2]) which is the resolution function of the instrument.

3. Experimental

Samples under investigation were 1 mm thick slices of a Czochralski-grown dislocation free ingot. Some of them were thermally oxidized at 1000°C for 100 min and the SiO_2 film was 93 nm thick. Both sides of slices were oxidized to avoid the elastic bending of the crystal [9].

The measurements of the diffuse scattering of thermal neutrons were carried out at the triple-axis spectrometer for perfect crystal studies in the Kurchatov Institute, Moscow. The (1 1 1) reflection of silicon crystals with (1 1 1) orientation of the surface was used at monochromator, sample and analyzer in $(n, -n, n)$ position. The neutron wavelength was 0.156 nm. Analyzer rocking curves were taken at a constant angular deviation from the exact Bragg position of the sample. Because of the small signal-to-background ratio, only the scattering concentrated near the Bragg peak ($\theta < 40''$, instrumental peak width about $2''$) could be observed.

Triple-crystal diffraction patterns of the initial non-oxidized crystal show only the coherent (main)peak and the pseudo peak, any diffusely scattered intensity does not exist (Fig. 2).

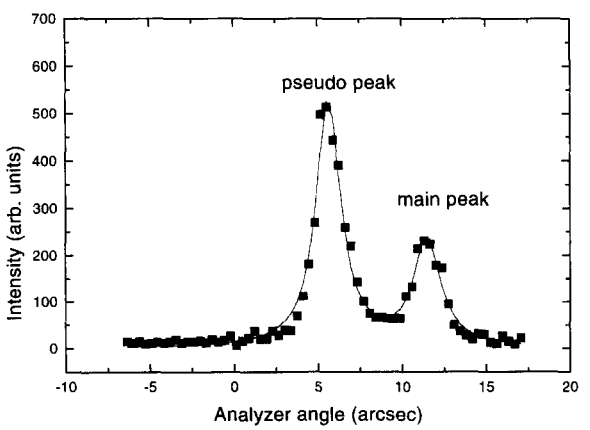

Fig. 2. Analyzer rocking curve for the initial non-oxidized (as-grown) sample at an angular deviation of 6.0 s of arc from the exact Bragg position of the sample.

Typical rocking curves of the analyzer crystal for the oxidized sample are presented in Fig. 3. The diffuse peak is observed both for positive and negative angular deviations of the sample orientation from the exact Bragg position. The dependence of the diffuse peak integral intensity on the sample angular deviation (on the value of q_0) is given in Fig. 4. The intensity of the diffuse peak for negative values of the angular deviation is much greater than that for positive ones. The symmetric part of the diffuse intensity $I_s = \frac{1}{2}(I_{meas}(+\theta) + I_{meas}(-\theta))$ is plotted versus $\ln(|\theta|)$ in Fig. 5. I_{meas} is the measured diffuse intensity; for deviations $\theta > 8''$ the values of $I_{meas}(+\theta)$ vanish.

The dependence shown in Fig. 5 is nearly linear and the intercept of the straight line with the $\ln(\theta)$ axis at $\theta = 41''$ gives a mean size of $R_0 = (220 \pm 20)$ nm for the distorted lattice region caused by the clusters. The strong asymmetry of the

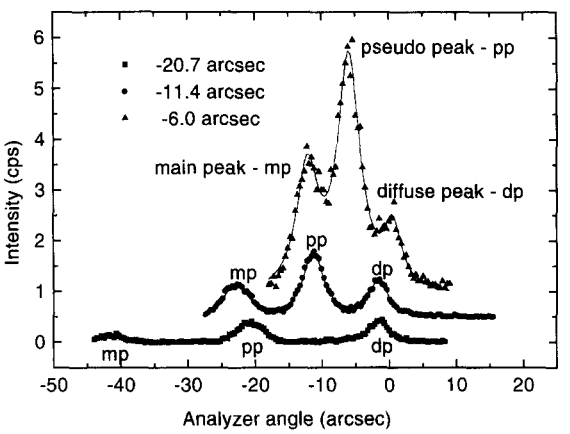

Fig. 3. Analyzer rocking curves for the oxidized sample at angular deviations of -20.7, -11.4, and -6.0 s of arc from the exact Bragg position of the sample. According to the usual terminology [2] the main peak (mp) corresponds to a successive reflection of a neutron beam from the angular centre of the monochromator crystal curve at the sample crystal reflection curve wings, whereas the pseudo peak (pp) is caused by a reflection of the neutron beam at the monochromator crystal reflection curve wings followed by the reflection at the sample crystal angular reflection centre. The diffuse peak (dp) intensity is the intensity scattered at the distorted crystal regions of the sample. (For a convenient diagram values of 0.5 cps and 1.0 cps are added to the intensity of the curves for angular deviation of -11.4 and -6.0, respectively.)

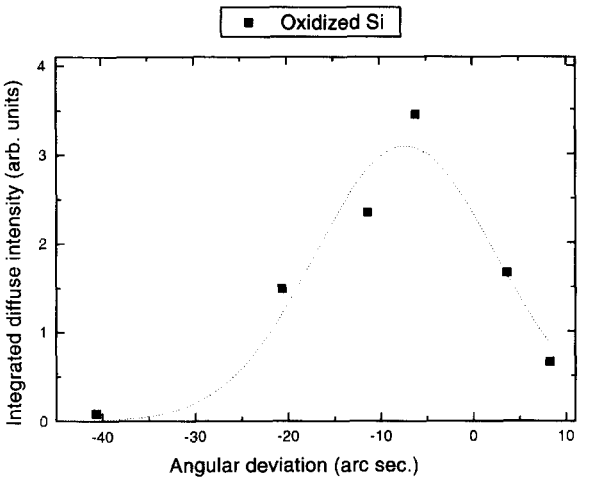

Fig. 4. Dependence of the integrated intensity of the diffuse peak on the angular deviation of the oxidized sample.

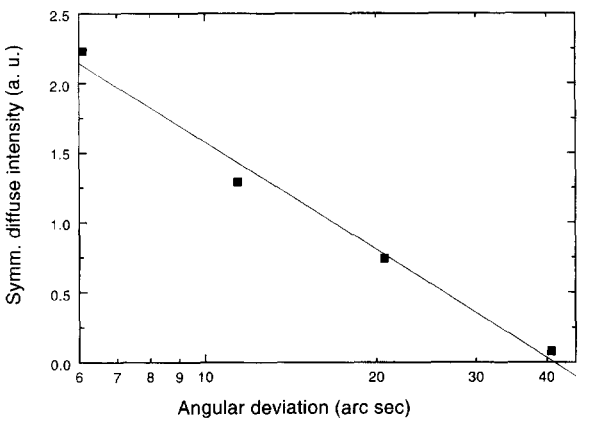

Fig. 5. Plot of the symmetrical part of the integrated intensity of the diffuse peak versus logarithm of the angular deviation of the sample. The intercept gives the value of $R_0 = (220 \pm 20)$ nm for the radius of the distorted crystal lattice region around the cluster.

diffuse scattering can be explained by the tensile stress field in the volume of crystal which produces higher intensity at negative angular deviations indicating a vacancy type of defects.

4. Discussion

According to [10] thermal oxidation is known to favour the growth of stacking faults of the intrinsic type in silicon. The mean size of these faults observed by TEM was about 250 nm [11]. An intrinsic stacking fault represents itself a withdrawal of a part of an atomic plane. The formation of intrinsic-type stacking faults is caused by the general mechanism of silicon oxidation. For the reaction localized on the surface the silicon atoms must move from the bulk to the surface, and this process is the source of vacancy generation in the bulk, the condition for the stacking fault formation. So we can decide that the stacking faults are the origin of the scattering observed in the present study. The mean defect radius derived from our data is close to that of [11]. Also a similar mean defect size of 230 nm has been obtained in [1], where the diffuse scattering from oxygen clusters in Czochralski-grown Si after the heat treatment at 975°C for 5 h has been studied. So according to the results obtained by different methods the effective size of defects in silicon single crystals after oxidation is in the range 200–250 nm. Different asymmetry of the diffuse scattering pattern in [1] and of the present study is the indication of the presence of different types of defects (oxygen clusters and stacking faults).

The results of this study show the feasibility of defect structure investigation in silicon by measurements of the diffuse scattering of thermal neutrons.

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