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Revealing the radiation-induced effects in silicon by processing at enhanced temperatures–pressures

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Abstract

Effect of processing at up to 1400 K under Ar hydrostatic pressure (HP) equal to 1.1 GPa for oxygen-containing Czochralski grown silicon (Cz-Si) irradiated with neutrons (energy $E = 5$ MeV, dose $D = 1 \times 10^{17}$ cm^{-2}) or γ-rays ($E = 1.2$ MeV, $D = 1000$ Mrad) on oxygen clustering and precipitation has been investigated by electrical, X-ray, infrared absorption, and photoluminescence methods. Depending on irradiation conditions, processing of irradiated Cz-Si, especially under HP, results in creation of oxygen-containing defects. Such processing of irradiated Cz-Si is helpful for revealing its irradiation-related history.

Keywords: Czochralski silicon; Irradiation; Microstructure; Hydrostatic pressure

1. Introduction

Irradiation of oxygen-containing Czochralski grown silicon (Cz-Si) basic material for integrated circuits, sensors, etc. with neutrons, electrons, or γ-rays results in creation of silicon interstitials, vacancies, and vacancy–oxygen complexes, V_xO_y. Subsequent annealing of irradiated Cz-Si at sufficiently high temperature (HT) causes the massive precipitation of oxygen interstitials (O_i's), especially if performed under enhanced hydrostatic pressure (HP). As shown by Hallberg and Lindström (1992) and Misiuk et al. (2005a, b), oxygen-related defects are formed at HT, or, especially, at the HT–HP conditions. The V_xO_y and similar irradiation-induced defects act as nucleation centres for clustering of O_i's. In other words, the HT–HP treatment of Cz-Si results in enlargement of radiation-induced disturbances and so creates the specific microstructure. As it follows from the papers by Misiuk et al. (2005a, b) and Londos et al. (2005), post-irradiation processing of Cz-Si under HP can help in revealing its irradiation-related history. Revealing the irradiation-induced effects in Cz-Si irradiated with fast neutrons or γ-rays by processing at enhanced temperature–pressure is now investigated in more detail.

2. Experimental

Three kinds of Si samples of $14 \times 6 \times 2$ mm^3 dimensions, with different initial concentration of O_i's, were cut from Cz-Si rods (Table 1) and irradiated with neutrons (energy $E = 5$ MeV, dose $D = 1 \times 10^{17}$ cm^{-2}) or γ-rays using a ^{60}Co source ($E = 1.2$ MeV, $D = 1000$ Mrad). In what follows, symbol N is added for labelling the samples irradiated with neutrons (NT1, NT2); the γ-irradiated sample is labelled as γT3. The reference T1, T2, T3 and irradiated NT1, NT2, γT3 samples were subjected to processing for 5 or 10 h in inert Ar atmosphere at temperatures

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up to 1400 K, under atmospheric pressure (10^5 Pa) and HP = 1.1 GPa.

The microstructure and related sample properties were investigated by electrical (capacitance–voltage, C–V, measurements), high resolution X-ray diffraction (to record reciprocal space maps, XRRSMs), Fourier transform infrared (FTIR) absorption, and photoluminescence (PL, measured at 6 K, excitation with Ar laser, $\lambda = 488$ nm) methods.

Table 1
Characteristics of investigated samples: crystallographic orientation, O_i concentration (c_o), conductivity type, concentration of carriers (electrons, N_e, or holes, N_h), and kind of irradiation: with neutrons (N) or γ-rays (γ)

Sample	Orientation	c_o (cm^{-3})	Conductivity	$N_{e,h}$ (cm^{-3})	Irradiation
T1	001	11×10^{17}	p	3×10^{14}	N
T2	111	9.5×10^{17}	n	2.5×10^{14}	N
T3	111	8.1×10^{17}	n	2.5×10^{15}	γ

Table 2
Concentration of electrons in conduction band ($N_e \times 10^{15}$ cm^{-3}) in reference and irradiated samples processed for 10 h at 723 K under 10^5 Pa and 1.1 GPa

Pressure/sample	T1	NT1	T2	NT2	T3	γT3
10^5 Pa	2.5	2.0	1		5.4	5.3
1.1 GPa	10	4.5	3	0.75	11	13

Enhanced concentration of electrons in the conduction band of Cz-Si processed at 623–823 K is related to a creation of the chain-like oxygen complexes, exhibiting thermal donor (TD) activity (Lee et al., 2001). High-resolution X-ray diffractometry delivers information on the structural perfection of processed single crystalline silicon (Lal et al., 2000). The concentration of O_i's in the Cz-Si lattice (c_o) was determined by FTIR absorption measurements before and after the treatments: the presence of O_i's results in IR absorption near 1107 cm^{-1}. The PL emission at about 0.807 eV (the D1 PL line) from processed Cz-Si is related to the presence of dislocations created at the oxygen precipitate/Si matrix boundary (Pizzini et al., 2000).

3. Results and discussion

Post-irradiation processing at 723–923 K of the irradiated Cz-Si samples (NT1, NT2, γT3) did not affect markedly their microstructure in comparison with the case of the same treatment applied to the reference samples.

The effect of treatments at 723 and 923 K on the O_i content in the reference as well as in irradiated samples was the same within an accuracy of the applied FTIR method ($\pm 0.5 \times 10^{17}$ cm^{-3}).

Processing of the reference Cz-Si at 723 K resulted in the creation of TDs, much enhanced in the case of treatment under HP (Table 2). In the case of irradiated Cz-Si, the concentration of electrons in the conduction band (N_e) was distinctly lowered

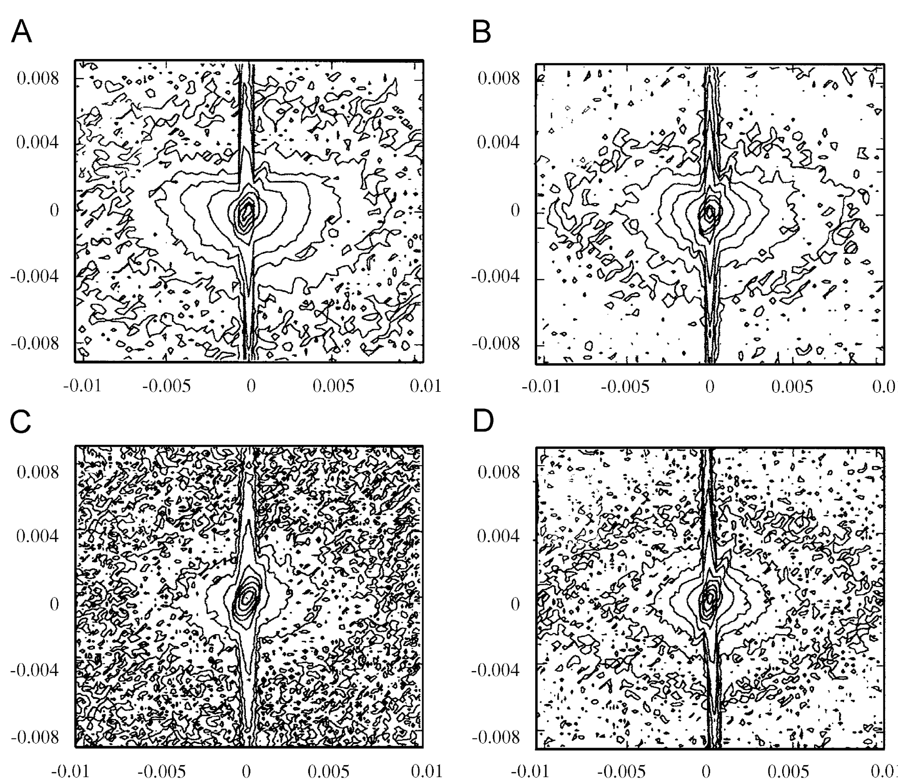

Fig. 1. XRRSMs taken for 333 reflection from T3 (A and C) and γT3 samples (B and D), as prepared (A, B) and processed for 10 h at 923 K under 1.1 GPa (C, D). Axes x and y are in relative reciprocal lattice units (rlu), Δq_x and Δq_y ($10\,nm^{-1}$).

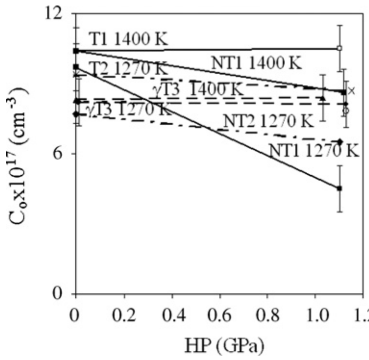

Fig. 2. Dependence of c_o on HP applied at processing for 5 h of T1, T2, T3, NT1, NT2, and γT3 samples at 1270 and 1400 K. Error bars are indicated.

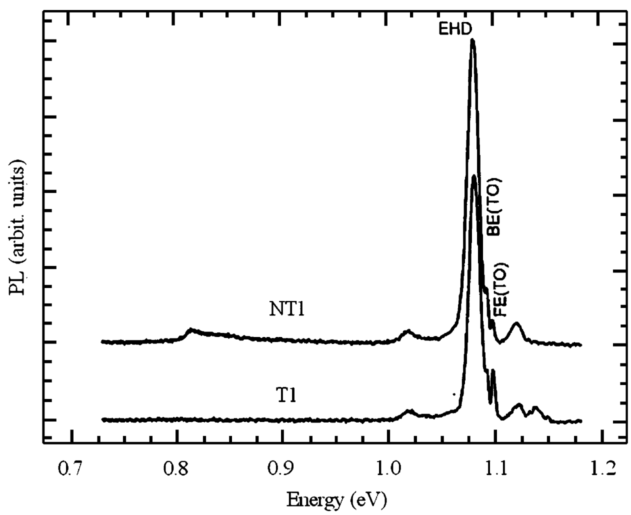

Fig. 3. PL spectra of T1 and NT1 samples processed for 5 h at 1270 K under 10^5 Pa. EHD means emission related to electron–hole droplets; BE(TO) and FE(TO)—transverse optical phonon replicas of boron bound exciton and of free exciton, respectively.

(Table 2). This is related to the presence of acceptors produced at irradiation.

Processing at 923 K of T3 and γT3 samples, both under 10^5 Pa and HP, resulted in the decreased X-ray diffuse scattering intensity evidencing out-annealing of some grown-in as well as of irradiation-induced defects (Fig. 1). This out-annealing was similar in the case of neutron-irradiated samples.

Contrary to the case of processing at 723–923 K, the treatment of irradiated Cz-Si at 1270–1400 K affects much stronger the resulting microstructure, especially if performed under HP (Figs. 2–7).

The O_i's concentration in the reference and irradiated samples, subjected to processing at 1270 and 1400 K, is presented in Fig. 2. Processing of T1 and NT1 samples, with the highest initial c_o value (Table 1), at 1270–1400 K under 1.1 GPa resulted in substantially decreased concentration of O_i's. The c_o value decreased in effect of the formation of oxygen-containing clusters and precipitates (agglomerates).

HP-induced removal of O_i's from the Si lattice was clearly detectable also for neutron-irradiated NT2, most pronounced

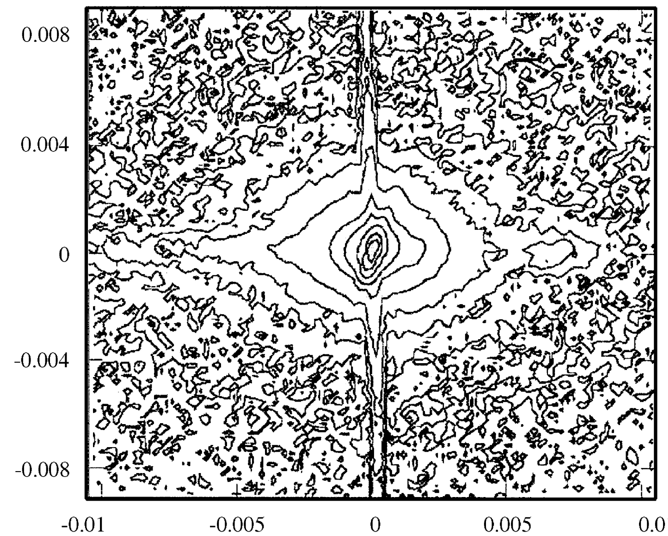

Fig. 4. XRRSMs for 333 reflection from NT2 sample processed for 5 h at 1270 K under 1.1 GPa. Axes x and y are given in relative reciprocal lattice units (rlu), Δq_x and Δq_y (10 nm^{-1}).

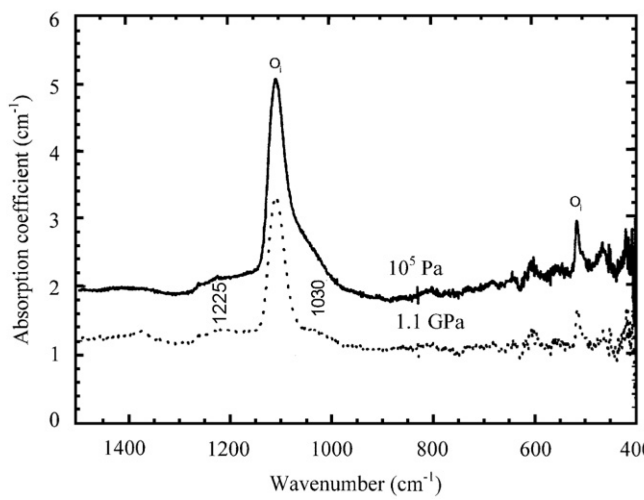

Fig. 5. Fourier transform absorption spectra (FTIR, taken at 300 K) of NT2 samples processed for 5 h at 1270 K under 10^5 Pa and 1.1 GPa.

after processing at 1270 K (Fig. 2). Much smaller effect of HP on oxygen precipitation has been detected for the T3 and γT3 samples (Cz-Si with the lowest initial c_o value, Table 1).

The presence of the PL peak at 0.81 eV (the so-called D1 line) for the NT1 sample processed under 10^5 Pa at 1270 K (Fig. 3) evidenced the presence of dislocations at the SiO_{2-x}/Si boundary. This PL line was of distinctly higher intensity for the NT1 and NT2 samples treated at 1270 K under 1.1 GPa. This means that O_i's precipitating on the irradiation-induced nucleation centres form relatively large misfitting SiO_{2-x} agglomerates.

Features of XRRSM for the NT1 and NT2 samples processed at 1270 K under HP (Fig. 4) can be interpreted as indicating the presence of plate-like oxygen precipitates. These precipitates are formed by oxygen interstitials, as evidenced by

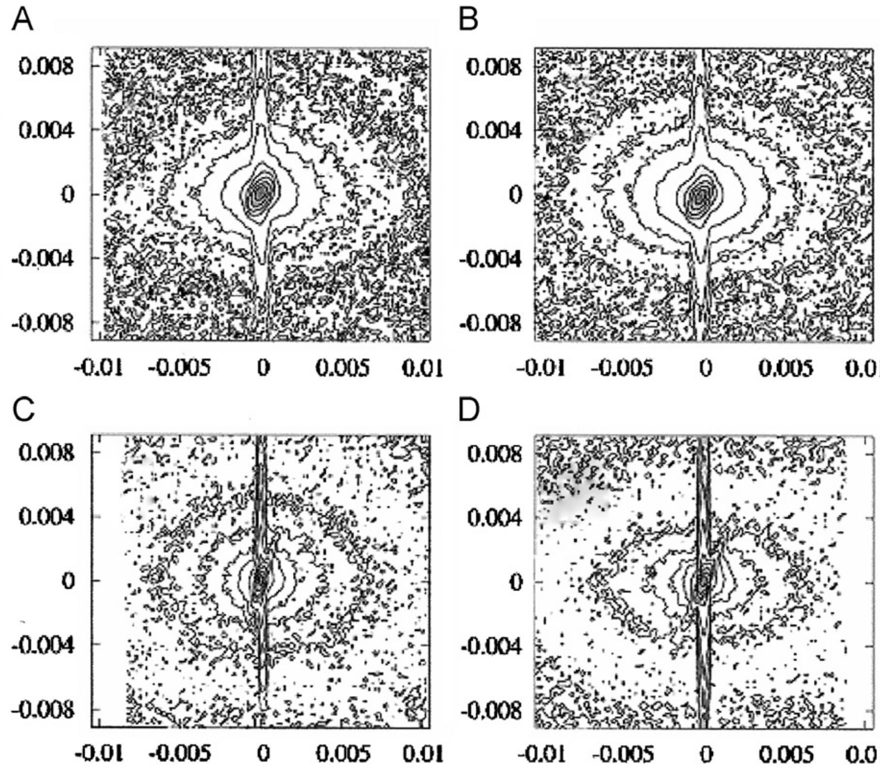

Fig. 6. XRRSMs for 333 reflection from T3 (A and C) and γT3 (B and D) samples processed for 5 h under 1.1 GPa at 1270 K (A and B) and at 1400 K (C and D). Axes x and y are given in relative reciprocal lattice units (rlu), Δq_x and Δq_y ($10\,nm^{-1}$).

decreased absorption at $1107\,cm^{-1}$ corresponding to the decreased concentration of O_i's (Fig. 5). This precipitation takes place at the nucleation sites, created by the HP-induced activation of irradiation-induced point defects and their clusters (Misiuk et al., 2005a,b).

The decrease in c_o is accompanied with the increased IR absorption at $1225\,cm^{-1}$ and, especially, at $1030\,cm^{-1}$ (Fig. 5); these features have been reported to be related to oxygen precipitates (Surma et al., 2005).

Irradiation with neutrons affects precipitation of O_i's at 1270 K much stronger than γ-rays (Fig. 2), at least for the irradiation doses applied in this study. Loss of interstitial oxygen from γT3 processed for 5 h at 1270 K under 1.1 GPa was equal to about $0.5 \times 10^{17}\,cm^{-3}$ only. As it follows from XRRSMs, γ irradiation affects the transformation of defects in γT3 samples processed under HP in a specific way (Fig. 6).

X-ray diffuse scattering increased meaningfully for γT3 sample processed under HP at 1270 K (Fig. 6B) in comparison to that observed for the similarly processed reference T3 sample (Fig. 6A). Oxygen-related defects responsible for X-ray diffuse scattering in processed γT3 are small, of below 10 nm dimension.

Processing of the NT1 samples at 1400 K under HP resulted in distinctly enhanced oxygen precipitation, contrary to the case of similarly processed reference T1 (Fig. 2). Irradiation-induced precipitation of O_i's with the creation of SiO_{2-x} agglomerates was evidenced by enhanced IR absorption at about 1030 and $1225\,cm^{-1}$ (compare Fig. 7C with Fig. 7A and B); this confirms the creation of oxygen-deficient silicon dioxide precipitates. As indicated by Misiuk et al. (2005b), well resolved defects with large strain were observed in the synchrotron topographs for neutron-irradiated Cz-Si treated at 1400 K under HP. Neutron irradiation and subsequent processing at 1400 K under 1.1 GPa produced distinctly resolved large defects, most probably composed of SiO_{2-x} cores surrounded by dislocation loops.

In γT3 samples processed at 1400 K, HP-induced oxygen precipitation was much smaller ($\Delta c_o \leqslant 0.5 \times 10^{17}\,cm^{-3}$, Fig. 2); only minor processing-related differences in the microstructure have been detected by X-ray topography and X-ray diffuse scattering (Fig. 6C and D).

When Cz-Si is irradiated with energetic particles (or photons) of sufficiently high energy, self-interstitials and vacancies (the Frenkel pairs) are generated. These defects partly annihilate just during irradiation. Remaining (surviving annihilation) vacancies (V's) and interstitials (Si_i's) interact with impurities present in Cz-Si, among which O_i's are the strongest attractors. Thus vacancy–oxygen pairs (VO's) are created, as well as V_xO_y and larger clusters, the latter especially in neutron-irradiated Si.

In the case of Cz-Si irradiated with neutrons, processing under HP affects strongly its microstructure, related to oxygen clustering and precipitation, and so allows for insight into irradiation-related history of such objects.

In the case of Cz-Si irradiated with γ-rays and electrons (Misiuk et al., 2005a, b), the HP-induced effects are compar-

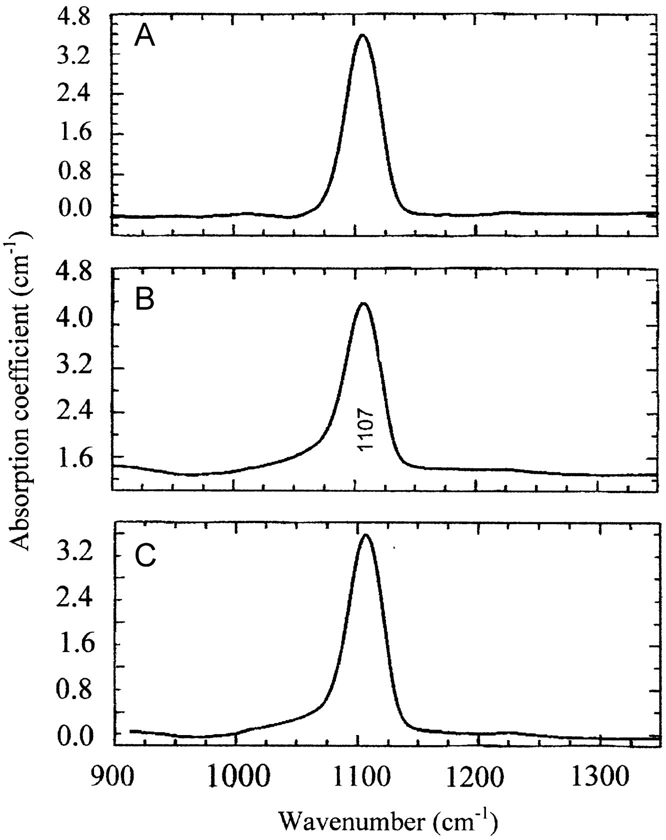

Fig. 7. FTIR absorption spectra (taken at 300 K) of T1 (A and B) and NT1 (C) samples processed for 5 h at 1400 K under 10^5 Pa (A) and 1.1 GPa (B and C). Absorption at about 1030 and 1225 cm^{-1}, related to SiO_{2-x} precipitates, contributes to total absorption (compare Fig. 5).

atively weak. Still the treatment of such samples under HP, especially at about 1270 K, also helps in revealing the irradiation-induced defects.

4. Conclusions

Processing at enhanced temperature–HP of oxygen-containing Cz-Si irradiated with neutrons or γ-rays contributes to revealing of the irradiation-induced defects. Appropriate HT–HP processing results in a specific enlargement of these defects being helpful in their recognition. This enlargement depends on the kind of irradiation (more pronounced for irradiation with neutrons), irradiation energy and dose (more pronounced for higher doses), and on the initial concentration of interstitial oxygen in silicon (more pronounced for Cz-Si with high concentration of O_i's). One can hope to apply this phenomenon for radiation detection/dosimetry e.g. in space and for military applications.

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