Materials Science in Semiconductor Processing 83 (2018) 1–11

Contents lists available at ScienceDirect

Materials Science in Semiconductor Processing

journal homepage: www.elsevier.com/locate/mssp

Production and annealing of the paramagnetic defects in as-grown and oxygen doped floating zone silicon irradiated with high fluence 3.5 MeV and 27 MeV electrons

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ARTICLE INFO

Keywords:
Electron spin resonance
Silicon
Irradiation defects
Multivacancies

ABSTRACT

The production and thermal stability of the irradiation paramagnetic point defects (IPPDs) in the as-grown, standard float-zone silicon (STFZ) and oxygen doped float-zone silicon (DOFZ) irradiated at room temperature with high fluence 3.5 MeV (1×10^{17} cm^{-2}) low energy and 27 MeV (2×10^{16} cm^{-2}) high energy electrons were investigated by Electron Spin Resonance (ESR). The nature and concentration of the IPPDs identified in the as-irradiated and isochronally annealed up to 300 °C samples by ESR measurements under intense above the gap 1.06 μm *in-situ* laser illumination, were found to depend on oxygen concentration and electrons energy. While irradiation of STFZ with electrons produced as main IPPDs, besides divacancies, larger tetravacancy and pentavacancy cluster defects, irradiation of DOFZ resulted mainly in divacancies, vacancy-oxygen and interstitial oxygen-carbon impurity pairs. The observed variation in the nature of the main resulting IPPDs with the concentration of incorporated oxygen is explained by differences in the dominant defects production mechanisms. Thus in STFZ with lower oxygen concentration (1×10^{16} cm^{-3}) the dominant production mechanisms are the direct defects formation from a chain of neighboring vacancies by a cascade of secondary recoils across the path of the high energy irradiating particle and the divacancies diffusion and trapping/aggregation. Meanwhile, in DOFZ with larger oxygen content (1.2×10^{17} cm^{-3}) the primary vacancy and interstitial trapping by the oxygen and carbon impurities is the dominant defect production mechanism. Variations in the concentration and nature of the IPPDs observed during isochronal annealing are discussed in terms of defects thermal activated diffusion and recombination processes.

1. Introduction

Silicon (Si) based radiation detectors are used on a large scale in fundamental research, such as elementary particles and nuclear physics, or in research with photons and radiation in free electron laser experiments [1]. In the case of the Large Hadron Collider (LHC) at the European Nuclear Research Center CERN and its High Luminosity upgrade (HL-LHC) the use of crystalline Si in pixel and microstrip detectors for particle tracking applications requires to improve its bulk radiation hardness to radiation damage produced by the larger intensity, high energy charged hadrons, resulting in the degradation of the detectors performance [2–4]. The expected increase in the luminosity of HL-LHC up to 10^{35} cm^{-2} s^{-1} requires an increased radiation hardness up to a fluence of 10^{16}n$_{eq}$ cm^{-2} for fast signal collection, one order of magnitude higher than current detector technologies [5].

The radiation induced changes in the properties of the crystalline Si used in the tracking detectors are due to the formation of irradiation defects, leading to modifications of the electrical properties. The structure of some of the radiation induced electrically active defects (EADs), which proved to have a direct impact on the performance of the sensors operating at room temperature (RT), is still under debate [3,6–8]. Investigations are therefore required to determine the formation, structure and interaction of the irradiation defects which are produced at room temperature (RT) by charged and neutral particle beams at high radiation fluence. Among the experimental techniques used in investigating the defects in semiconductors [9], Electron Spin/Paramagnetic Resonance (ESR/EPR) spectroscopy offers comprehensive information about the presence and structure of the irradiation paramagnetic point defects (IPPDs), about the role played by existing impurities, in particular the oxygen and carbon, in their production, stability and recombination properties [10–13]. In the particular case of crystalline Si, which is by far the best ESR investigated elementary

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https://doi.org/10.1016/j.mssp.2018.04.003
Received 30 January 2018; Received in revised form 20 March 2018; Accepted 3 April 2018

Table 1
Characteristics of the electron irradiated Si samples investigated by ESR.

Si samples	Energy of irradiation	Fluence	Impurity concentrations	Size
STFZ	3.5 MeV	1×10^{17} cm^{-2}	c(O) = 1×10^{16} cm^{-3}	$4 \times 1.8 \times 0.3$ mm^3
STFZ	27 MeV	2×10^{16} cm^{-2}	c(C) = 2×10^{15} cm^{-3}	
			c(P) = 1×10^{12} cm^{-3}	
DOFZ	3.5 MeV	1×10^{17} cm^{-2}	c(O) = 1.2×10^{17} cm^{-3}	
DOFZ	27 MeV	2×10^{16} cm^{-2}	c(C) = 2×10^{15} cm^{-3}	
			c(P) = 1×10^{12} cm^{-3}	

semiconductor, a large number of IPPDs have been already identified. Their spectra parameters and proposed structural models reference data are tabulated in the Landolt-Boernstein Database [14] and in the Defect Dat@base from the University of Tsukuba, Japan [15]. The properties of the EADs in silicon were discussed and tabulated in Ref. [16].

Here we present the results of a comparative study concerning the nature of the IPPDs produced at RT in n-type (P-doped) oxygen-lean, standard STFZ and oxygen doped float-zone silicon DOFZ by irradiation with high fluence monochromatic electron beams of low energy (3.5 MeV) (fluence 1×10^{17} cm^{-2}) and high energy (27 MeV) (fluence 2×10^{16} cm^{-2}), as well as their production properties during further thermal isochronal annealing up to 300 °C. To our knowledge no such a study has been reported yet. Up to now, the large majority of ESR investigations have been performed on STFZ irradiated with low energy electrons (1–4 MeV), resulting in the detailed characterization of the divacancy in various paramagnetic charge states, namely G7/$[V_2]^-$, G6/$[V_2]^+$, NL11/$[V_2]^\circ$ and of the vacancy-oxygen B1/A/$[V\text{-}O]^-$ defect [17–23]. ESR investigations have also been performed on Czochralski grown silicon (CzSi) with two orders of magnitude higher oxygen concentration, irradiated with ~ 2 MeV electrons at fluence values from 1×10^{15} cm^{-2} to 1.2×10^{19} cm^{-2} [24–26]. The observed formation in such samples, besides divacancies, of the B1/A/$[V\text{-}O]^-$ and A14/$[V_2\text{-}O]$ defects with subsequent formation by annealing at higher temperatures of the $[V_n\text{-}O_m]$ (n, m > 1) type centers was explained by the vacancy migration and their excellent trapping by oxygen impurities [27]. One should also mention the ESR investigation in the production rate of the G7/$[V_2]^-$ and B1/A/$[V\text{-}O]^-$ defects *vs.* electrons energy up to 56 MeV, at fluence of up to 6×10^{16} cm^{-2} (for 1.41 MeV electrons) in both p- and n-type CzSi [28,29]. None of these investigations on electron irradiated oxygen rich CzSi or oxygen lean STFZ [22,26] reported the formation of larger vacancy cluster defects $[V_n]$ ($n > 2$) such as A4/$[V_3]^-$, A3/$[V_4]^-$ or P1/$[V_5]^-$, observed in neutron irradiated CzSi and FzSi [30–33]. Very recently [34] we identified in the ESR spectra of STFZ irradiated with high fluence 27 MeV electrons, recorded at $T < 150$ K under *in-situ* illumination with intense above the gap 1.06 μm laser light, besides divacancies, small cluster tetravacancy and pentavacancy defects. The production of such small cluster defects was attributed to both direct formation and thermal activated diffusion and aggregation of divacancies, processes expected to be active according to theoretical predictions.

We show here that the nature and structure of the IPPDs produced by high fluence 3.5 MeV low and 27 MeV high energy electrons in oxygen lean STFZ and oxygen doped DOFZ depend on the electrons energy and concentration of oxygen. Thus, the STFZ irradiated with 3.5 MeV electrons contains besides divacancies, comparable amounts of cluster pentavacancy but not tetravacancy defects, as observed in 27 MeV irradiated STFZ [34]. Meanwhile, the oxygen doped DOFZ, with one order of magnitude increased oxygen concentration, irradiated in similar conditions as STFZ, contains in both cases besides divacancies, comparable amounts of vacancy-oxygen and oxygen-carbon pair defects. The observed difference between the production of the multivacancy cluster defects at lower and higher irradiation energy in STFZ, as well as their absence in electron-irradiated DOFZ are discussed in terms of IPPDs production mechanisms.

2. Experimental details

2.1. *Samples preparation and irradiation*

The investigations were performed on samples originating from a Si wafer of 0.3 mm thickness and 100 mm diameter of oxygen lean standard float-zone, n-type (P-doped), high resistivity (ρ = 3–4 kohm cm) single-crystal silicon, with (100) oriented polished surfaces, from Wacker-Chemitronik GmbH. Such Si-wafers are routinely used in producing tracking detectors at CERN. Their impurity concentration values: c(O) = 1×10^{16} cm^{-3}; c(C) = 2×10^{15} cm^{-3}, c(P) = 1×10^{12} cm^{-3} were in the previously reported range for this material (Table 4.1 from Ref. [16]). Oxygen enrichment was performed on platelets of 15×15 mm^2 cut from the STFZ wafer, which were double side implanted with 3 MeV energy O^{17} (70% abundance) ions (each side with 1×10^{14} cm^{-2}), followed by annealing for 72 h in a pure nitrogen atmosphere at 1150 °C. According to SIMS measurements the oxygen concentration in the resulting DOFZ samples was 1.2×10^{17} cm^{-3}. STFZ and DOFZ platelets of 10×10 mm^2 and 5×5 mm^2 were further irradiated at RT with 27 MeV (fluence 2×10^{16} cm^{-2}) and 3.5 MeV electrons (fluence 1×10^{17} cm^{-2}) using the irradiation facilities from the Metrology Institute PTB, Braunschweig, Germany [35] and Belarusian State University BUS, Belarus [36] respectively. The characteristics of the samples for the presently reported ESR investigations, cut from the irradiated platelets, are given in Table 1.

2.2. *ESR methodology*

The ESR methodology is similar to the one described in Ref. [34]. The ESR samples, cut with the longest axis parallel to one of the < 110 > crystal axes, were inserted in calibrated ultrapure silica ESR sample tubes of 2 mm inner diameter with closed bottom. Between the measurements the samples were kept, together with the remaining irradiated platelets, in a freezer at $T < -25$ °C. The ESR measurements were performed in the Q (34 GHz) band microwave frequency, from RT down to 10 K, with an ELEXSYS-E500Q (Bruker) spectrometer equipped with a probe head offering a nominal sensitivity of 1.2×10^9 spins/Gauss, inserted in a cryostat operating from 3.6 K up to RT. Details about the equipment and magnetic field calibration procedures are given in Ref. [37]. *In-situ* above the gap optical excitation was done with the 1.06 μm coherent light provided by a thermally stabilized pig tailed laser diode model LPS-1060-FC from ThorLabs, of 60 mW maximum output power, directed through optical fibers to the ESR sample inserted in the ESR probe head [34]. ESR spectra were recorded with the magnetic field rotated in a (110) plane, in steps of 5° between the < 100 > and < 110 > directions. An estimated ± 0.75° orientation accuracy of the recorded spectra was obtained.

The ESR measurements were performed on samples as-irradiated and further subjected to isochronal/pulse annealing firstly from RT to 150 °C and afterwards in steps of 50 °C, from 150 °C to 300 °C, for 30 min each. From the analysis of the resulting ESR spectra recorded after each annealing step one can evaluate the temperature induced changes in the nature and concentration of the observed IPPDs [38]. The as-irradiated samples exhibited low intensity, isotropic EPR spectra, attributed [39] to paramagnetic surface and impurity related