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2005 J. Phys.: Condens. Matter 17 S2247

(http://iopscience.iop.org/0953-8984/17/22/012)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **17** (2005) S2247–S2253 doi:10.1088/0953-8984/17/22/012

Annealing of defects in irradiated silicon detector materials with high oxygen content

M Mikelsen[1], E V Monakhov[1], G Alfieri[1], B S Avset[2], J Härkönen[3] and B G Svensson[1]

[1] Department of Physics, Physical Electronics, University of Oslo, PO Box 1048 Blindern, N-0316 Oslo, Norway
[2] SINTEF ICT, PO Box 124 Blindern, N-0314 Oslo, Norway
[3] Helsinki Institute of Physics, University of Helsinki, PO Box 64, FIN-00014, Finland

Received 6 October 2004
Published 20 May 2005
Online at stacks.iop.org/JPhysCM/17/S2247

Abstract
A deep level transient spectroscopy (DLTS) study of electrically active defects in electron irradiated silicon detectors has been performed. Two types of materials have been studied and compared: carbon-lean magnetic Czochralski (MCZ-) Si, and high purity, diffusion oxygenated float-zone (DOFZ-) Si. In both materials we observed an earlier reported shift in position of peaks associated with the divacancy (V_2) at 250–325 °C, indicating a gradual transition from V_2 to the divacancy–oxygen complex (V_2O). Heat treatments at higher temperatures reveal a difference in annealing behaviour of defects in DOFZ- and MCZ-Si. It is observed that VO and V_2O anneal with a higher rate in DOFZ-Si. The appearance of a hydrogen related level only in the DOFZ-Si reveals a small presence of H and it is suggested that the difference in annealing behaviour is due to defect interaction with H in the DOFZ-Si. Our findings also suggest that dissociation may be a main mechanism for the annealing of V_2O in MCZ-Si.

1. Introduction

The characterization and study of irradiation induced defects is of key importance in the search for new and radiation tolerant detector materials. One particular type of material that has received recent interest is oxygen enriched silicon. It was demonstrated by the CERN RD48 collaboration that diffusion oxygenated float-zone (DOFZ) Si showed considerably improved radiation tolerance as compared to ordinary float-zone (FZ) Si [1, 2].

Czochralski (CZ-) Si has a higher oxygen concentration than DOFZ-Si. It is expected that such high concentrations of O will influence the formation of defects during irradiation, in particular for high doses where oxygen depletion can occur. High resistivity CZ-Si suited for detector application has only become available in recent years after recent developments in the crystal growth utilizing magnetic techniques [3]. There is a lack of data for this material regarding radiation induced defects.

Table 1. Survey of the samples used in the study.

Sample	Doping ($P\ cm^{-3}$)	Carbon (cm^{-3})	Oxygen (cm^{-3})
MCZ-Si	5.5×10^{12}	$\leqslant 10^{16}$	$(5–10) \times 10^{17}$
DOFZ-Si	5.0×10^{12}	$(2–4) \times 10^{16}$	$(2–3) \times 10^{17}$

Deep level transient spectroscopy (DLTS) studies of irradiation induced defects in DOFZ Si have revealed pronounced concentrations of divacancy (V_2) and vacancy–oxygen (VO) complexes. In studies of the thermal stability of these defects, a gradual shift in the position of the V_2 peaks has been observed at 250–300 °C. This shift has been interpreted as formation of a new centre, identified as a divacancy–oxygen pair (V_2O) with two acceptor levels close to those of V_2, as V_2 anneals out [4]. A strong support for this identification is that the formation rate of the new levels and the annealing rate of V_2 have been found to be proportional to the oxygen content [5]. Laplace-DLTS studies have also shown that the formation of the new levels and the annealing of V_2 have an almost one-to-one correlation [6]. Recently, an observation of V_2O formation has also been reported in carbon-lean CZ-Si with a typical oxygen concentration of 10^{18} cm^{-3} and presumably low concentrations of other impurities [7].

2. Experimental details

In this work we have used DLTS to study irradiation induced acceptor type defects and their evolution under isochronal heat treatment. Two different Si detector materials have been investigated; high purity DOFZ-Si and C-lean magnetic Czochralski (MCZ) Si.

Two sets of samples were studied: the first set was prepared from n^- MCZ-Si. During fabrication of this material a magnetic field is utilized to control the distribution of charged impurities in the melt. The second set was prepared from n^- DOFZ-Si. The manufacture of this material includes a step where high purity FZ-Si is O enriched: the FZ wafers are dry-oxidized for 21 h at 1200 °C, and a subsequent anneal is then performed at 1200 °C in a N_2 atmosphere where O atoms from the pre-formed SiO_2 surface layer diffuse into the bulk Si. For all samples boron and phosphorus implantations with post-implant annealing were performed to form $p^+n^-n^+$ diodes. As a final step aluminium contacts were deposited. Secondary ion mass spectroscopy (SIMS) has been used to determine the oxygen and carbon concentrations (see table 1 for a summary), where also the electron concentration in the n^- layer is given. The samples were irradiated with 15 MeV electrons to a dose of of 4×10^{12} cm^{-2} for MCZ-Si and 2×10^{12} cm^{-2} for DOFZ-Si.

The DLTS measurements were made using a set-up described elsewhere [8]. In short, the temperature of the sample was scanned between 77 and 280 K, and the measured capacitance transients were averaged in intervals of 1 K. The DLTS signal was extracted by using a lock-in type of weighting function, and different spectra were obtained corresponding to six rate windows ranging from $(20\ ms)^{-1}$ to $(640\ ms)^{-1}$. The isochronal annealing was performed in steps lasting 15 min at temperatures ranging from 50 to 400 °C. DLTS scans were done before and after each step.

3. Experimental results and discussion

Figure 1 shows DLTS spectra for both types of samples before and after heat treatments at $\leqslant$250 °C. For both samples one observes three dominant peaks labelled E1, E2 and E3.

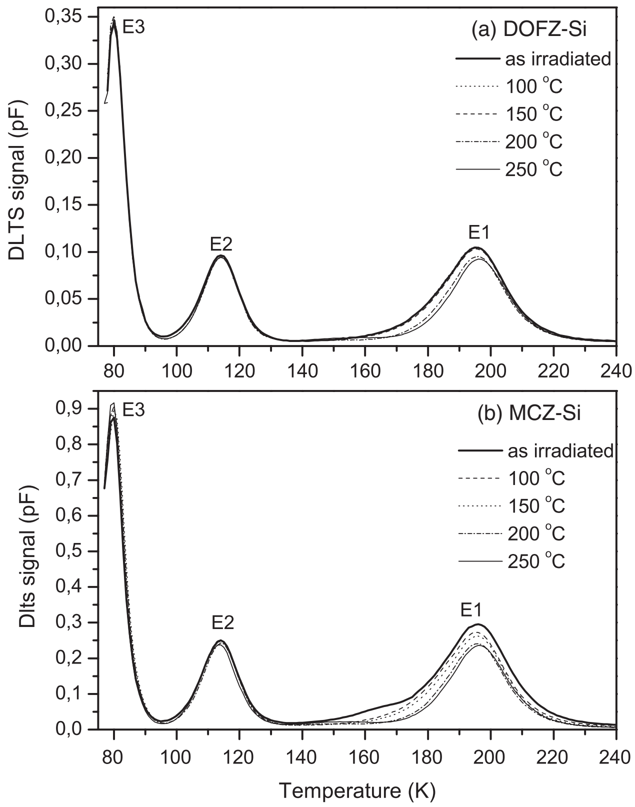

Figure 1. DLTS spectra for as-irradiated and annealed ($\leqslant$250 °C) n-type MCZ and DOFZ-Si samples.

The corresponding activation enthalpies are 0.43, 0.23 and 0.18 eV. Based on previous work we identify E1 as the singly negatively charged divacancy, V_2 $(0/-)$, which may have a small admixture of the vacancy–phosphorus pair, VP [9–11]. E2 is identified as the doubly negatively charged divacancy, V_2 $(-/=)$ [9, 10, 12], and E3 as the VO pair with a possible minor contribution from the carbon interstitial–carbon substitutional complex [11, 13, 14]. In the as-irradiated MCZ-Si we also notice a minor peak at 162 K. This peak disappears at 50 °C, and its identity is currently not known.

For both MCZ- and DOFZ-Si the annealing behaviour in the temperature range $\leqslant$200 °C is similar: E2 and E3 remain relatively stable, while there is a gradual decrease of E1 until it reaches approximately the same amplitude as E2 at 200 °C. The interpretation is that the E2 and E3 peaks are relatively 'pure', and correspond solely to the V_2 $(-/=)$ and VO $(0/-)$ transitions. E1 is likely to be a composite, where V_2 $(0/-)$ is the major contributor. The other constituents to E1 start to anneal at 50 °C, and at 200 °C E1 is a relatively pure divacancy peak.

Figure 2 shows DLTS spectra after annealing at temperatures in the range 250–350 °C. Similarly to that reported in [4, 5], a gradual shift in the divacancy related peaks occurs, which has been interpreted as a transition from V_2 to V_2O. This behaviour is observed in both DOFZ- and MCZ-Si. At 325 °C the positions of the peaks stabilize, suggesting that V_2 is annealed out. In [4, 5] it is concluded that the E1 and E2 peaks are mainly V_2O related at 325 °C. A difference in amplitude suggests, however, that other minor defects also may contribute to the

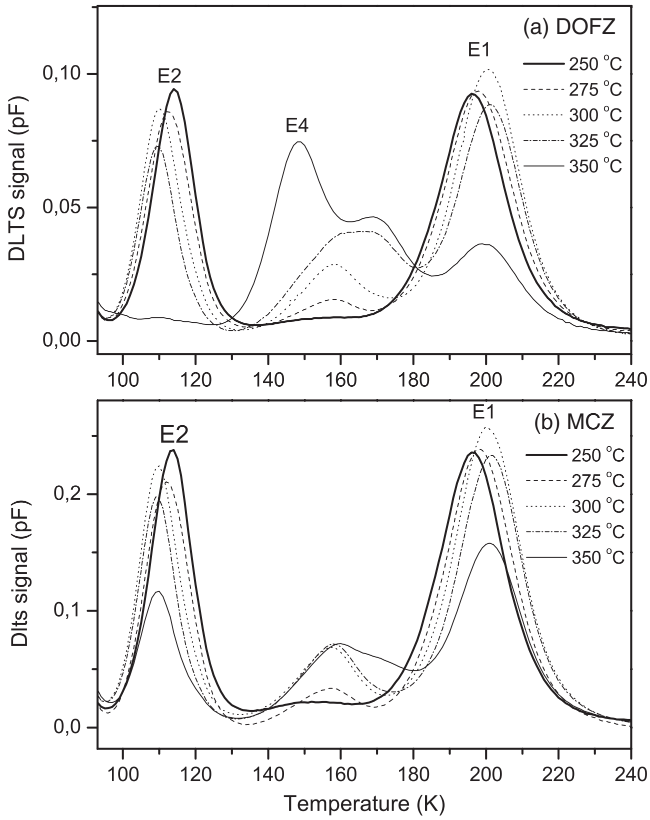

Figure 2. DLTS spectra after annealing at temperatures between 250 and 350 °C, revealing a shift in the V_2 related peaks, and interpreted as a transition from V_2 to V_2O.

single acceptor level (∼200 K). The activation enthalpies of V_2O (0/−) and V_2O (−/=) are 0.47 and 0.23 eV, respectively. We notice a minor peak at ∼160 K after annealing at 275 °C present in both samples, and a peak at ∼170 K after annealing at 300 °C primarily pronounced in the DOFZ samples. The origins of these peaks have not been identified.

After annealing at the intermediate temperatures (250, 275 and 300 °C), the E1 and E2 peaks are a mixture of V_2 and V_2O. By using the obtained energy and capture cross-sections for the V_2 and V_2O levels in the 200 and 325 °C annealed samples, respectively, the overlapping peaks are fitted to the sum of the two DLTS signals by varying only the amplitudes of V_2 and V_2O. We thereby obtain the individual amplitudes for V_2 and V_2O. Within experimental accuracy, the results of this fit agree with previous Laplace-DLTS studies [6]. The results can be seen in figure 4, which shows the DLTS amplitudes for different defects after annealing steps between 200 and 400 °C.

The DLTS spectra for the MCZ- and DOFZ-Si are similar up to 325 °C. After the annealing at 350 °C, however, there are several differences: in the MCZ-Si a decrease in the V_2O concentration and a slight increase in the VO concentration takes place, while in DOFZ-Si the VO concentration decreases slightly. The E2 peak disappears while a fraction of E1 remains at 375 °C. We interpret this as V_2O annealing out and a second level appearing or remaining at the E1 position. Another feature in DOFZ-Si is the appearance of a level labelled E4 (figure 3). This level is absent in the MCZ samples and occurs with an activation enthalpy

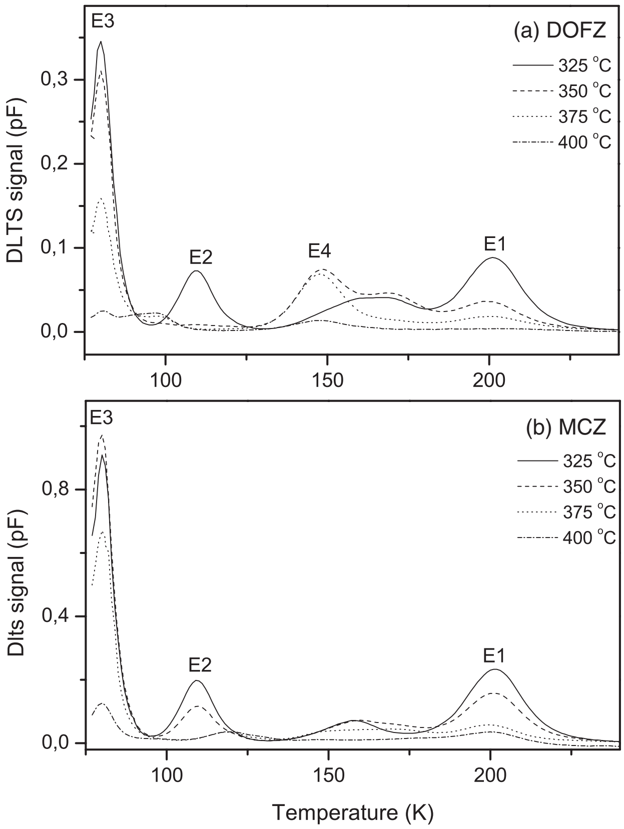

Figure 3. DLTS spectra for samples annealed between 325 and 400 °C.

of 0.32 eV. Based on previous studies this level is identified as the vacancy–oxygen–hydrogen complex, VOH [15–19].

At 375 °C, V_2O has also annealed out in the MCZ-samples. There is also a decrease in the intensity of the VO peak in both types of samples. It is interesting to note that this decrease is larger in the DOFZ-Si than in the MCZ-Si. VOH remains stable at 375 °C, but after annealing at 400 °C it is hardly detectable. At 400 °C both materials exhibit a strong decrease in the VO concentration.

The appearance of VOH shows the presence of H in DOFZ-Si, which can explain the observed differences in defect annealing in the two materials. Indeed, H is known to be mobile at elevated temperatures and to interact strongly with defects having dangling bonds. The relative drop in VO concentration after annealing at 350 °C and at 375 °C is substantially larger in the DOFZ samples compared with the MCZ samples, and the concurrent growth of VOH suggests the reaction $VO + H \rightarrow VOH$, where H is the mobile species. The results also indicate that the annealing of V_2O in DOFZ-Si is H assisted: $V_2O + H \rightarrow V_2OH$. Since the electrical properties of V_2O are similar to those of V_2, it may be reasonable to assume that the properties of V_2OH are similar to those of V_2H. It is known that V_2H has a level at ~0.43 eV which overlaps with V_2 (0/−) [20, 21]. We therefore tentatively identify the ~200 K level observed in DOFZ-Si at 350 °C as V_2OH, exhibiting an activation enthalpy of ~0.44 eV and an apparent capture cross section of $\sim 7 \times 10^{-15}$ cm^2.

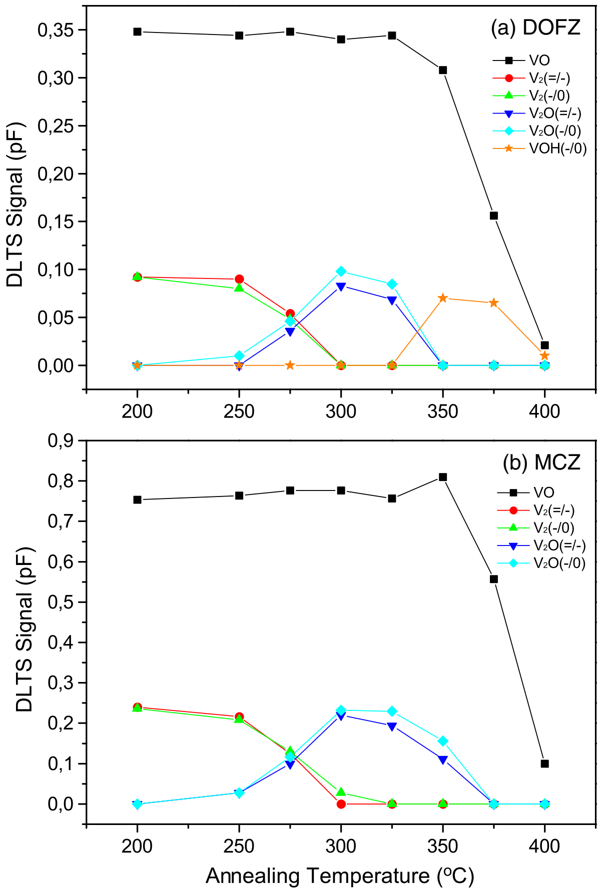

Figure 4. Amplitudes of DLTS peaks corresponding to different defects. The amplitudes of V_2 and V_2O in the overlapping region were obtained by fitting.

(This figure is in colour only in the electronic version)

In MCZ-Si it is interesting to note that the small (but significant) increase in the VO concentration at 350 °C correlates with a decrease in the V_2O concentration, which implies that the dissociative reaction $V_2O \rightarrow VO + V$ is an important annealing mechanism. This mechanism for V_2O annealing has also been predicted in theoretical studies [22].

4. Summary

In summary, we have investigated irradiation induced defects in detector grade DOFZ-and MCZ-Si and their behaviour under subsequent isochronal annealing between 50 and 400 °C. The two materials behave in a similar manner in the range $\leqslant$325 °C, where the earlier reported transition of V_2 to V_2O is observed. However, at higher temperatures the presence of H in DOFZ-Si is revealed by the emerging VOH centre, which is not detected in the MCZ samples. The data also show that VO and V_2O anneal at a faster rate in the DOFZ-Si, presumably caused by interaction with migrating H-atoms. As a result, a new level with an activation enthalpy of $\sim$0.44 eV occurs when V_2O disappears in DOFZ-Si and this level is tentatively ascribed to V_2OH. In MCZ-Si, our findings suggest that dissociation of V_2O ($V_2O \rightarrow VO + V$) is an important annealing mechanism.

Acknowledgments

The authors thank The Norwegian Research Council (NFR) for financial support, and Okmetic for providing MCZ-Si samples. The work was partly done within the framework of the RD50 CERN collaboration.

References

[1] Lindström G *et al* (The RD48 collaboration) 2001 *Nucl. Instrum. Methods* A **465** 60
[2] Lindström G *et al* (The RD48 collaboration) 2001 *Nucl. Instrum. Methods* A **466** 308
[3] Savolainen V, Heikonen J, Ruokolainen J, Anttila O, Laakso M and Paloheimo J 2002 *J. Cryst. Growth* **243** 243
[4] Monakhov E V, Avset B S, Hallén A and Svensson B G 2002 *Phys. Rev.* B **65** 233207
[5] Alfieri G, Monakhov E V, Avset B S and Svensson B G 2003 *Phys. Rev.* B **68** 233202
[6] Monakhov E V, Alfieri G, Avset B S, Hallén A and Svensson B G 2003 *J. Phys.: Condens. Matter* **15** S2771
[7] Markevich V P, Peaker A R, Lastovskii S B, Murin L I and Lindstom J L 2003 *J. Phys.: Condens. Matter* **15** S2779
[8] Svensson B G, Ryden K-H and Lewerentz B M S 1989 *J. Appl. Phys.* **66** 1699
[9] Kimerling L C 1977 *Radiation Effects in Semiconductors 1976* ed N B Urli and J W Corbett (Bristol: Institute of Physics Publishing) p 221
[10] Svensson B G and Willander M 1987 *J. Appl. Phys.* **62** 2758
[11] Brotherton S D and Bradley P 1982 *J. Appl. Phys.* **53** 5720
[12] Evwaraye A O and Sun E 1976 *J. Appl. Phys.* **47** 3776
[13] Song L W, Zhan X D, Benson B W and Watkins G D 1990 *Phys. Rev.* B **42** 5765
[14] Keskitalo N, Hallén A, Lalita J and Svensson B G 1997 *Defects and Diffusion in Silicon Processing* (*Mater. Res. Soc. Symp. Proc.* No. 469) ed T Diaz de la Rubia, S Coffa, P A Stolk and C S Rafferty (Pittsburg, PA: Materials Research Society) p 233
[15] Pellegrino P, Lévêque P, Lalita J, Hallén A, Jagadish C and Svensson B G 2001 *Phys. Rev.* B **64** 195211
[16] Bonde Nielsen K, DobaMCZewski L D, Goscinski K, Bendesen R, Andersen O and Bech Nielsen B 1999 *Physica* B **273/274** 167
[17] Peaker A R, Evans-Freeman J H, Rubaldo L, Hawkins I D, Vernon-Parry K and DobaMCZewski L 1999 *Physica* B **273/274** 243
[18] Tokuda Y and Seki T 2000 *Semicond. Sci. Technol.* **15** 126
[19] Lévêque P, Pellegrino P, Hallén A, Svensson B G and Privitera V 2001 *Nucl. Instrum. Methods Phys. Res.* B **174** 297
[20] Bonde Nielsen, Dobaczewski L D, Goscinski K, Bendesen R, Andersen O and Bech Nielsen B 1999 *Physica* B **273/274** 167
[21] Lévêque P, Hallén A, Svensson B G, Wong-Leung J, Jagadish C and Privitera V 2003 *Eur. Phys. J.: Appl. Phys.* **23** 5
[22] Coutinho J, Jones R, Öberg S and Briddon P R 2003 *Physica* B **340–342** 523