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Effects of germanium doping on the behavior of oxygen and carbon impurities and impurity-related complexes in Si

C.A. Londos [a,*], A. Andrianakis [a], V.V. Emtsev [b], G.A. Oganesyan [b], H. Ohyama [c]

[a] University of Athens, Solid State Physics Section, Panepistimiopolis Zografos, Athens 157 84, Greece
[b] Ioffe Physicotechnical Institute of the Russian Academy of Sciences, Politeknicheskaya ul. 26, 194021 St. Petersburg, Russia
[c] Kumamoto National College of Technology, 26592, Nishigoshi, Kumamoto 861-1102, Japan

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ABSTRACT

The production and annealing of oxygen and carbon related defects in electron-irradiated Ge-doped Czochralski silicon have been studied by means of IR spectroscopy. The results demonstrate that the role of Ge changes with increasing Ge content. At low Ge concentrations, much less than $10^{20}\,cm^{-3}$, Ge atoms act as temporary traps for vacancies preventing their annihilation with self-interstitials. At high Ge concentrations, Ge atoms form clusters which tend to attract vacancies and self-interstitials, enhancing their mutual annihilation. As a result, for low Ge concentrations the production of oxygen-related defects (VO) and carbon-related defects (C_iO_i, $C_iO_i(Si_I)$, C_iC_s) tend to increase as a function of Ge content, while for high Ge concentrations tend to decrease. The annealing of the oxygen-related defects VO_n ($1 \leq n \leq 6$) is also affected by the Ge presence. The ratio of the VO defects that convert to VO_2 is reduced in Ge doped material. However, this ratio increases for the conversions $VO_n \rightarrow VO_{n+1}$ ($3 \leq n \leq 5$), respectively. The annealing temperature of VO defect and the growth temperature of VO_2 defect decrease versus Ge content. On the other hand, the thermal stability of the carbon-related defects was found to be insensitive to the presence of Ge. It was also found that the carbon precipitation process shows some marked differences in irradiated Ge-doped Si, in comparison with the undoped material.

1. Introduction

Silicon is the material-of-choice for most of the nowadays electronic and microelectronic applications. During crystal growth, various impurities are unintentionally introduced in the Si lattice. The most important among them are the oxygen impurity incorporated at interstitial sites and the carbon impurity incorporated at substitutional sites. Although both are electrically inactive, upon preparation of the material for various applications they give rise to electrically active complexes which affect material properties and behavior. Important stages of material processing are irradiations or/and implantations and thermal treatments. The production, evolution and reactions of oxygen and carbon related defects in Si are topics that material science attempts to elucidate in every detail.

Important characteristics for a large variety of applications are the thermal stability and the radiation hardness of silicon. Upon thermal treatments, for instance at 450 °C, oxygen containing thermal donors [1,2] are introduced which affect the electrical properties of the material. Thermal treatments at higher temperatures in the range (700–1100 °C) introduce precipitates [1,3,4] which affect the mechanical, thermal and electrical properties in the material. Upon irradiation at room temperature, the primary defects produced, that are vacancies and self-interstitials, are efficiently trapped [5–8] by oxygen and carbon impurities, respectively, leading to the formation of various complexes, most of them electrically active. It is well-known that isovalent impurities reduce the generation of secondary radiation defects, thus improving radiation hardness. They also affect the formation of thermal donors and precipitates and therefore can be used to control the thermal stability of the material.

Important among isovalent impurities in Si is germanium. It supresses the formation of thermal donors but it seems to enhance oxygen precipitation [9]. Thus, it can potentially affect the thermal stabilty of the material. This element is incorporated in the Si lattice at substitutional sites [10] and due to its larger covalent tetrahedral radii than that of Si ($r_{Ge} = 1.22$ Å, $r_{Si} = 1.17$ Å) it produces compressive elastic strains in the lattice which are relieved by the capture of vacancies. On the other hand, carbon which is another isovalent impurity in Si, when present in the material also at substitutional sites [10], due to its smaller covalent tetrahedral radii ($r_C = 0.77$ Å) gives rise to tensile strains, which are relieved by the capture of self-interstitials. Thus germanium and carbon are effective sinks for vacancies and self-interstitials, respectively, and therefore when

* Corresponding author. Tel./fax: +30 2107276726.
E-mail address: hlontos@phys.uoa.gr (C.A. Londos).

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both are present in Cz–Si, they could affect the production and the interactions of radiation-induced defects. Consequently, the radiation hardness of the material could be affected.

The shrinkage of devices dimensions, introduces stresses which affect the behavior of various defects. Since Ge introduces internal stresses in the Si lattice it would be interesting to investigate its influence on defects. Any acquired knowledge would be beneficial for defect control and defect engineering. Additionally, external pressure either uniaxial or hydrostatic, is widely used to investigate various phenomena generally in solids [11–15]. Therefore, the knowledge acquired by the study of the Ge induced strains in the Si lattice could be also beneficial in relation with the understanding of phenomena related to the application of external hydrostatic pressure and vice versa.

The most important radiation-induced defects in Cz–Si containing carbon are the VO and the C_iO_i and C_iC_s defects. Upon annealing a family of oxygen related VO_n defects is formed [5,6]. Also, on high irradiation doses $C_i(Si_I)$, $C_iC_s(Si_I)$ and $C_iO_i(Si_I)$ complexes are formed [7,16]. In this work, we studied the effect of Ge doping on the production, the evolution, the interactions and the reactions of the main oxygen and carbon-related defects in electron-irradiated Si.

2. Experimental details

Czochralski grown germanium-doped silicon (Cz–Si:Ge) samples containing carbon were used in this work. The samples were cut from prepolished wafers and they have dimensions of $20\times10\times2\,mm^3$. Their initial germanium, carbon, and oxygen concentrations are cited in Table 1. The Ge concentrations were estimated from the mass ratio of Si and Ge in the melt. The oxygen concentration was determined from the 1107 cm^{-1} IR band using a calibration coefficient of $3.14\times10^{17}\,cm^{-2}$ [17]. The carbon concentration was determined from the 605 cm^{-1} IR band using a calibration coefficient of $1.0\times10^{17}\,cm^{-2}$ [18].The samples were irradiated with 2 MeV electrons, with a dose of $5\times10^{17}\,cm^{-2}$, using the Dynamitron accelerator at Takasaki-JAERI (Japan). After the irradiation all the samples were subjected to isochronal anneals up to 900 °C, in steps of $\Delta T\approx10\,°C$ and 20 min duration. After each annealing step, the IR spectra were recorded at room temperature by means of a FT-IR spectrometer with a resolution of 1 cm^{-1}.

3. Experimental results and discussion

3.1. *The effect of Ge on the production of VO, C_iO_i, C_iC_s, $C_iO_i(Si_I)$ defects*

Fig. 1 shows the IR spectra in the region of 500–1200 cm^{-1} of the Cz–Si:Ge-8 sample before and after electron irradiation; hereafter we shall use short names for the Ge-doped samples, for instance Ge-8 instead of Cz–Si:Ge-8. The spectra of all the other samples are very similar apart from the amplitudes of the respective bands. The well-known bands of the O_i (513, 1107 cm^{-1}) and C_s (605 cm^{-1}) impurities as well as the radiation-induced bands VO (830 cm^{-1}), C_iO_i (862 cm^{-1}), $C_iO_i(Si_I)$ (936, 1020 cm^{-1}) and C_iC_s(546 cm^{-1}) are shown. The concentrations of these defects are cited in Table 1. The calibration coefficients were taken $6.25\times10^{16}\,cm^{-2}$ [19] for the VO band, $1.1\times10^{17}\,cm^{-2}$ [19] for the C_iO_i band, $3.81\times10^{16}\,cm^{-2}$ [19] for the $C_iO_i(Si_I)$ band and $1.5\times10^{15}\,cm^{-2}$ [20] for the C_iC_s band. Fig. 2 shows the concentration of the VO (a), the C_iO_i (b), C_iC_s (c) and $C_iO_i(Si_I)$ (d) defects, respectively, versus Ge content. To facilitate the discussion the samples were organized in three groups. Group I contains the samples Cz–Si, Ge-2 and Ge-6 with $[C_s]_o<2\times10^{16}\,cm^{-3}$, labeled carbon-lean samples. Group II contains the samples Ge-1, Ge-3, Ge-4 and Ge-7 with $[C_s]_o$ in the range 2×10^{16} to $1\times10^{17}\,cm^{-3}$ labeled carbon-moderate doped samples. Group III contains the samples Ge-5 and Ge-8 with $[C_s]_o$ around $2\times10^{17}\,cm^{-3}$, labeled carbon-rich samples.

In the carbon-lean samples of Group I the main produced defect is the VO defect. A gradual increase in the concentration of VO defect is observed versus Ge content (Fig. 2a). It is well-known, that Ge atoms can effectively compete with O in Si in trapping the vacancies [21,22]. At temperatures higher than 200 K these Ge-V defects are not stable. We argue that at room temperature irradiation, Ge atoms act as temporary traps for vacancies [23] thus preventing their direct annihilation with self-interstitials. As a result, the production of the VO defects increases.

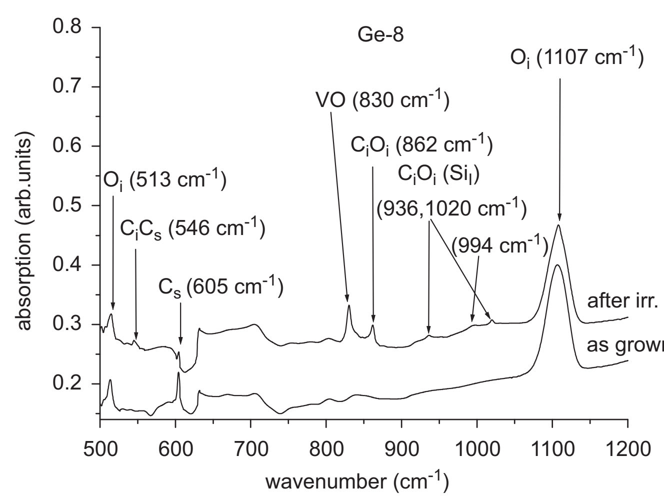

Fig. 1. Fragments of the IR spectra of the Ge-8 sample in the range 500–1200 cm^{-1} prior and after irradiation.

Table 1
Ge content, oxygen and carbon concentrations before the electron irradiation, as well as the concentrations of VO, C_iO_i, C_iC_s, and $C_iO_i(Si_I)$ defects in the samples used in our study.

Sample name	[Ge] (cm^{-3})	$[O_i]_o$ ($10^{17}\,cm^{-3}$)	$[C_s]_o$ ($10^{16}\,cm^{-3}$)	[VO] ($10^{16}\,cm^{-3}$)	$[C_iO_i]$ ($10^{16}\,cm^{-3}$)	$[C_iC_s]$ ($10^{14}\,cm^{-3}$)	$[C_iO_iSi_I]$ ($10^{15}\,cm^{-3}$)
Cz–Si	0	9.56	<2.0	2.35	[a]	[a]	[a]
Ge-1	1×10^{17}	9.60	2.0	3.40	1.55	1.25	2.96
Ge-2	7×10^{17}	6.50	<2.0	2.50	[a]	[a]	[a]
Ge-3	1×10^{18}	10.00	3.0	3.50	1.95	1.30	3.20
Ge-4	4×10^{18}	5.55	10.0	3.60	2.55	1.31	3.29
Ge-5	1×10^{19}	6.74	20.0	5.00	4.20	1.30	3.34
Ge-6	5×10^{19}	7.60	<2.0	3.10	[a]	[a]	[a]
Ge-7	1×10^{20}	8.77	3.7	3.75	2.30	1.35	3.81
Ge-8	2×10^{20}	7.70	18.0	4.25	3.85	1.20	3.16

[a] Below detection limit.

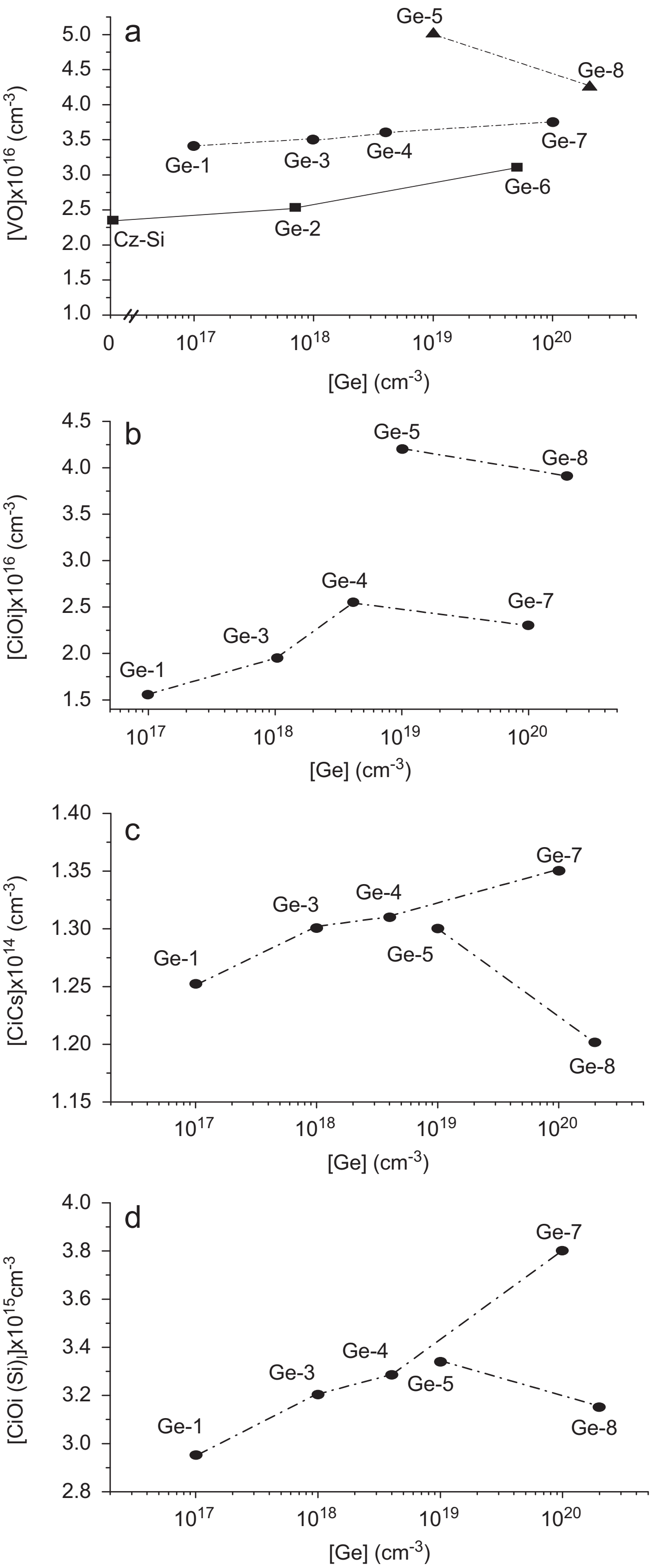

Fig. 2. The concentration of the VO (a), C_iO_i (b), C_iC_s (c) and $C_iO_i(Si_I)$ (d) defects versus [Ge].

In the carbon-moderate doped samples of Group II we have both the production of the oxygen- and carbon-related defects. By inspection of Fig. 2a–d it is observed that the concentration of the VO defect as well as the concentrations of the C_iO_i, C_iC_s and $C_iO_i(Si_I)$ defects, show a gradual increase versus Ge content. We notice that carbon is an effective trap for self-interstitials. The results could be explained in the same line of thought as that for the samples of Group I. We argue that the reduction of the annihilation rate between vacancies and self-interstitials due to the Ge presence allows for more vacancies to be captured by O_i atoms and more self-interstitials by C_s atoms, leading finally to an increase in the concentrations of the oxygen-related (VO) and carbon-related (C_iO_i, C_iC_s and $C_iO_i(Si_I)$) defects, respectively [23].

In the carbon-rich samples of group III it is observed that the concentrations of the VO, C_iO_i, C_iC_s and $C_iO_i(Si_I)$ defects are smaller in the samples with $[Ge] = 2\times10^{20}\,cm^{-3}$ than those of the samples with $[Ge] = 1\times10^{19}\,cm^{-3}$, respectively. Thus, in the group III samples a tendency of decreasing the concentrations of oxygen and carbon related radiation defects are observed. We note that earlier studies [24] on Ge-doped Si have reported significant differences in the photoluminescence spectra between samples with $[Ge] = 1\times10^{19}$ and $2\times10^{20}\,cm^{-3}$. The phenomenon was ascribed to the inhomogeneous distribution of the Ge atoms in the Si lattice and their tendency, when at large concentrations, to form clusters and introduce strains in the lattice. Vacancies and self-interstitials produced by the irradiation are trapped by these clusters and they are annihilated. Thus the annihilation rate of the primary defects is enhanced. As a result less vacancies and self-interstitials are available and therefore the production of VO and C_iO_i, C_iC_s and $C_iO_i(Si_I)$ defects is reduced [23].

3.2. *The effect of Ge on the thermal evolution of VO, C_iO_i, C_iC_s, $C_iO_i(Si_I)$ defects*

Fig. 3 displays the IR spectra in the region of 800–1050 cm^{-1} of the sample Ge-8 at several selected temperatures of annealing. The well-known bands VO_2 (888 cm^{-1}), VO_3 (904, 968, 1000 cm^{-1}) and VO_4 (985, 1010 cm^{-1}) defects [25,26] are shown, together with the bands at 967, 1005, 1037, 1051 cm^{-1} which were attributed to VO_5/VO_6 defects [27,28] or/and V_mO_n ($n>4$) defects [29]. The band at 994 cm^{-1} has been tentatively attributed to a VO structure perturbed by C and Ge neighboring atoms [23].

Fig. 4(a) and (b) shows the thermal evolution of the VO, VO_2, VO_3, VO_4 defects in Cz–Si and Ge-8 samples, respectively. The evolution of the VO_5/VO_6 or/and V_mO_n ($n>4$) is also shown in Fig. 4(b). The signals of these defects are very weak in the spectra of the Cz–Si samples and their evolution cannot be shown in Fig. 4(a).

By inspection of Fig. 4(a) and (b), it is immediately seen that the onset of the annealing of VO defect and the onset of the

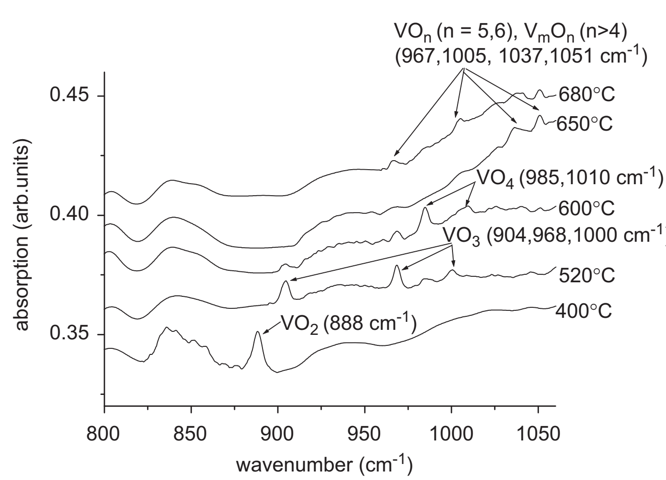

Fig. 3. Fragments of the IR spectra of the Ge-8 sample in the range 800–1050 cm^{-1} at various selective temperatures of the isochronal anneal sequence.

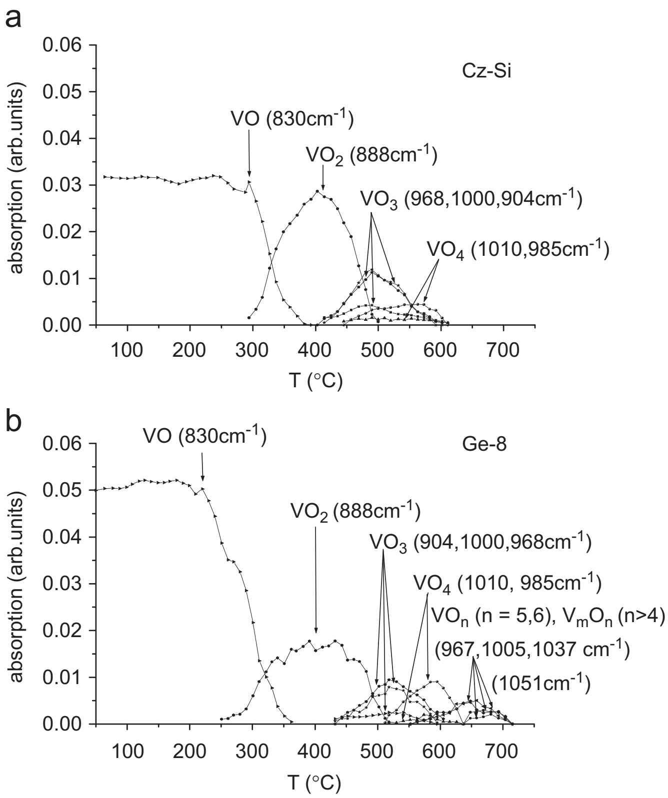

Fig. 4. The thermal evolution of the VO ($830\,cm^{-1}$), VO_2 ($888\,cm^{-1}$), VO_3 (904, 968, $1000\,cm^{-1}$), VO_4 (985, $1010\,cm^{-1}$) and VO_5/VO_6 or/and V_mO_n ($n>4$) (967, 1005, 1037, $1051\,cm^{-1}$) IR bands of the Cz–Si (a) and Ge-8(b) samples.

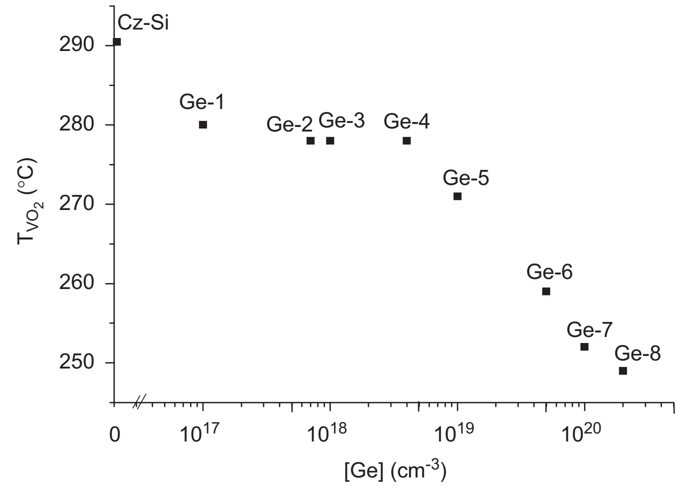

Fig. 5. The growth temperature of the VO_2 defect versus [Ge].

production of the VO_2 defect occur at markedly lower temperatures in the heavily doped Ge-8 sample than in the undoped Cz–Si sample; compare the temperatures $T_{VO}=290\,^{\circ}C$ and $T_{VO_2}=290\,^{\circ}C$ in the Cz–Si sample with the temperatures $T_{VO}=220\,^{\circ}C$ and $T_{VO_2}=250\,^{\circ}C$ in the Ge-8 sample, respectively. Fig. 5 shows the production temperature of the VO_2 defect versus Ge content. A gradual decrease of the T_{VO_2} is observed, which amounts to $\sim\Delta T=40\,^{\circ}C$ between the Ge-8 and the Cz–Si samples. We consider that this temperature lowering is related with the compressive strains introduced by Ge atoms in the Si material [30–32]. The VO_2 defect forms mainly when migrating VO centers are captured [5,6,25] by O_i atoms ($VO+O_i\rightarrow VO_2$). As a result of the induced strains due to the Ge presence, the potential barrier for the migration of the VO defect is reduced. This facilitates the movement of the VO centers and therefore the production of the VO_2 defects at lower temperatures. The phenomenon is manifested better for Ge concentrations in the range 10^{19}–$2\times10^{20}\,cm^{-3}$ (Fig. 5). The higher the Ge content of the material, the higher the induced strains and therefore the lower the production temperature of the VO_2 defect. Noticeably, a faster decay of the VO defect has been reported [33,34] in the IR spectra of the Si material subjected to high temperature, high-hydrostatic pressure treatments prior to irradiation. The phenomenon was attributed to pressure-induced decrease of the potential barrier for the migration of the VO defect, resulting in the production of the VO_2 defect, at a lower temperature.

We also observe (Fig. 4(b)) that in the Ge-8 sample the onset of the annealing of the VO defect occurs at an even lower temperature than that of the corresponding growth of the VO_2 defect. However, it is known [30,35] that self-interstitials emanated from large clusters in the course of annealing, also participate in reactions with VO defects ($VO+Si_I\rightarrow O_i$). We have argued [30,31] that the binding of the self-interstitials at the clusters is sensitive to strains in the Si lattice. Thus, a reduction of the binding energy of the self-interstitials due to the strains induced by the Ge presence could trigger the reaction $VO+Si_I\rightarrow O_i$ at a lower temperature.

Furthermore, it is clear by comparison of Fig. 4(a) and (b), that the concentration of the VO_2 is much lower in the Ge-doped Ge-8 sample than that in the Cz–Si sample. This is a result [30] of the enhanced contribution of the reaction $VO+Si_I\rightarrow O_i$ over the reaction $VO+O_i\rightarrow VO_2$ in the disappearance of the VO defect in the Ge-doped samples, therefore leaving less VO defects to convert to VO_2 defects. On the other hand, the conversion ratio of the $VO_n\rightarrow VO_{n+1}$ ($3\leq n\leq 5$) or/and V_mO_n ($n>4$) defects is larger in the Ge-doped Si. However, at temperatures higher than $450\,^{\circ}C$, oxygen atoms begin to diffuse as well as oxygen dimers are formed [36,37]. The latter species are very mobile at this temperature range. Additionally, the diffusivity of oxygen is enhanced [38] in Ge-doped Si. Consequently, the reaction rates of the formation of the VO_n ($n=5$ and 6), V_mO_n ($n>4$) defects are enhanced. It is worth noting that due to the enhanced formation of the VO_n ($n=5$ and 6), V_mO_n ($n>4$) defects we are able to monitor their IR bands in the spectra of the Ge-8 sample (Fig. 4(b)) in comparison with the IR spectra of Cz–Si where only traces of these bands exist and their evolution cannot be presented (Fig. 4(a)).

3.3. The thermal evolution of C_s impurities

Fig. 6 exhibits the thermal evolution of carbon impurity in the electron irradiated samples: Ge-4 (lightly Ge-doped), Ge-8 (heavily Ge-doped) selected from Table 1 as well as Cz–Si:0 (free Ge sample). As it is seen, a partial restoration of the isolated substitutional carbon occurs starting at $\sim500\,^{\circ}C$ and continuing up to the onset of the decay of the defect ($\sim700\,^{\circ}C$) from the IR spectra. The sources of the additional carbon atoms could be attributed [39] to large $C_N(Si_i)_M$ complexes which are formed from reactions among vacancies, self-interstitials and carbon atoms at elevated temperatures. It has been suggested [40] that with increasing annealing temperature a fraction of the $C_N(Si_i)_M$ complexes could trap extra self-interstitials and transform to SiC-based precipitates. Another fraction of them could react with vacancies and self-interstitials leading to the release of carbon atoms and the final recovery of this impurity. In the case of Ge presence, this recovery stage is substantially suppressed. We have

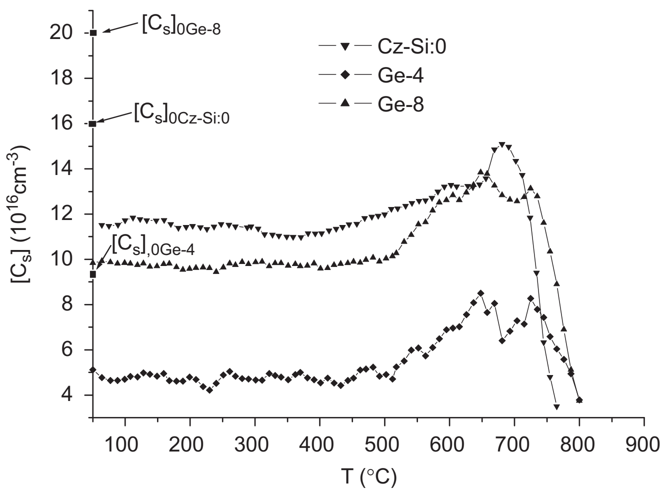

Fig. 6. The thermal evolution of the C_s (605 cm^{-1}) in Cz–Si:0, Ge-4 and Ge-8 samples.

argued [40] that, since Ge atoms could serve [23] as temporary traps for vacancies, the recovery of carbon substitutional atoms released from $C_N(Si_i)_M$ complexes is suppressed. Additionally, the local strains introduced by Ge impurities in the Si lattice also impede the restoration of carbon in the Ge-doped samples, in comparison with the Ge free samples. It is worth noting that the complete loss of carbon occurs in the Ge-doped irradiated Si at a slightly higher temperature ($\sim$40 °C) than that of the undoped material (Fig. 6).

4. Conclusions

We have reported studies on the effect of Ge doping on the behavior of oxygen-related and carbon-related defects in electron-irradiated Si. Firstly, it was found that for samples with [Ge] less than 10^{20} cm^{-3} Ge atoms, due to their tendency to serve as temporary traps for vacancies, reduce the annihilation rate of the primary defects. However, for high Ge concentration [Ge] $\approx 2\times10^{20}$ cm^{-3}, Ge atoms tend to form large clusters which, due to the induced strain fields, facilitate the annihilation of the primary defects. As a result, for low Ge concentrations the production of the VO, C_iO_i, C_iC_s and $C_iO_i(Si_I)$ defects increased as a function of Ge content, although for high Ge concentrations 2×10^{20} cm^{-3} the production of the above radiation defects decreases versus Ge content. Secondly, it was observed that the annealing temperature of the VO defect and the temperature of the growth of the VO_2 defect are gradually reduced versus Ge content. Furthermore, the conversion ratio $VO\rightarrow VO_2$ decreases versus [Ge] although, the conversion ratio $VO_n\rightarrow VO_{n+1}$ ($3\leq n\leq 5$) increases. Finally, it was found that the carbon precipitation process in irradiated Ge-doped Si is markedly affected, in comparison with the undoped material.

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