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Production and evolution of A-centers in n-type $Si_{1-x}Ge_x$

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The vacancy-oxygen pair (*VO* or A-center) in *n*-type $Si_{1-x}Ge_x$ crystals (x = 0, 0.025, 0.055) has been studied using infrared (IR) spectroscopy. It is determined that the *VO* production is suppressed in the case of *n*-type $Si_{1-x}Ge_x$ as compared to Si. It is observed that the annealing temperature of the *VO* defect in $Si_{1-x}Ge_x$ is substantially lower as compared to Si. The decay of the *VO* (830 cm^{-1}) band, in the course of 20 min isochronal anneals, shows two stages: The onset of the first stage is at ∼180 °C and the decrease of the *VO* signal is accompanied in the spectra by the increase of the intensity of two bands at ∼834 and 839 cm^{-1}. These bands appear in the spectra immediately after irradiation and were previously correlated with (*VO*-Ge) structures. The onset of the second stage occurs at ∼250 °C were the 830 cm^{-1} band of *VO* and the above two bands of (*VO*-Ge) decrease together in the spectra accompanied by the simultaneous growth of the 885 cm^{-1} band of the VO_2 defect. Interestingly, the percentage of the *VO* pairs that are converted to VO_2 defects is larger in the $Si_{1-x}Ge_x$ samples with intermediate Ge content (x = 0.025) as compared with Si (x = 0) and with the high Ge content samples (x = 0.055). The results are discussed in view of the association of *VO* pairs with Ge. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4795812]

I. INTRODUCTION

The technological requirement to replace Si with higher mobility substrates and the introduction of high-*k* dielectrics that substitute SiO_2 has regenerated the interest in the application of $Si_{1-x}Ge_x$ and related group IV alloys.[1,2] Apart from their higher carrier mobilities as compared to Si, the possibility for band-gap engineering constitute these materials advantageous for numerous applications, especially for optoelectronics and high speed devices.[3,4] The understanding of the defects introduced in $Si_{1-x}Ge_x$ crystals during the various stages of material fabrication is technologically important.

The study of oxygen and oxygen-related defects such as the A-center in $Si_{1-x}Ge_x$[5,6] is rather limited as compared to Si.[7–9] The A-center is an important defect pair in Si and its control is necessary to optimize devices.[10] Additionally, the *VO* defect is also electrically active in $Si_{1-x}Ge_x$ material,[6] and therefore, any information about its properties and behavior would be beneficial for increasing the quality of devices. In $Si_{1-x}Ge_x$, experimental studies revealed that oxygen interstitial (O_i) atoms preferentially bond with Si atoms rather than Ge atoms.[11] Notably, most of the investigations on the issue were limited to low Ge-content $Si_{1-x}Ge_x$.[6] Importantly, previous studies indicated that there is a propensity for the vacancy (*V*) to form near at least one Ge atom.[12] In particular Sihto *et al.*[12] used positron annihilation spectroscopy (PAS) to determine that there is an energy gain when a Ge atom replaces a Si atom next to a *V* in strained $Si_{1-x}Ge_x$. These results were further supported by the density functional theory (DFT) calculations of Chroneos *et al.*[12]

In $Si_{1-x}Ge_x$ as well as in Ge-doped Si material the Ge content influences the diffusion of oxygen and the conversion of the *VO* pairs to VO_2 clusters.[5,13,14] This is consistent to investigations of isovalent-doped Si, where there is an impact of doping on the production and successive conversion of oxygen-vacancy clusters.[15,16] In the present study, infrared spectroscopy is used to investigate the impact of Ge content on the production, evolution, and annealing of A-centers in *n*-type $Si_{1-x}Ge_x$ (x = 0, 0.025, 0.055).

II. EXPERIMENTAL DETAILS

The samples used in this study were cut from *n*-type $Si_{1-x}Ge_x$ wafers, which were grown by the Czochralski technique[17,18] (Institute of crystal growth, Berlin, Germany). The Ge concentration of the used samples was x = 0.025 and x = 0.055, plus a reference sample of x = 0 for comparison purposes. These *n*-type crystals were doped with phosphorus with a concentration of $[P] = 1 \times 10^{15}\,cm^{-3}$ during growth. The oxygen concentration of the samples was in the range of $6–9 \times 10^{17}\,cm^{-3}$ and their carbon concentration in the range $2–3 \times 10^{16}\,cm^{-3}$. The concentrations of P and Ge were determined by the provider. The samples were irradiated with 2 MeV electrons at T = 95 °C and a fluence of $5 \times 10^{17}\,cm^{-2}$ (Takasaki Jaeri Dynamitron Accelerator, Japan). Additionally, they were subjected to 20 min isochronal anneals from 50 to 550 °C with a step of 10 °C. After each annealing step, the IR spectra were measured at room temperature using a Fourier Transform IR spectrometer (FTIR).

III. RESULTS AND DISCUSSION

Figure 1 shows the IR spectra of the Si and $Si_{1-x}Ge_x$ (x = 0, 0.025, 0.055) samples considered here recorded just after irradiation and at a characteristic temperature of 350 °C, in the course of the 20 min isochronal anneals. It is well-known that in Cz-Si containing carbon, the vacancies

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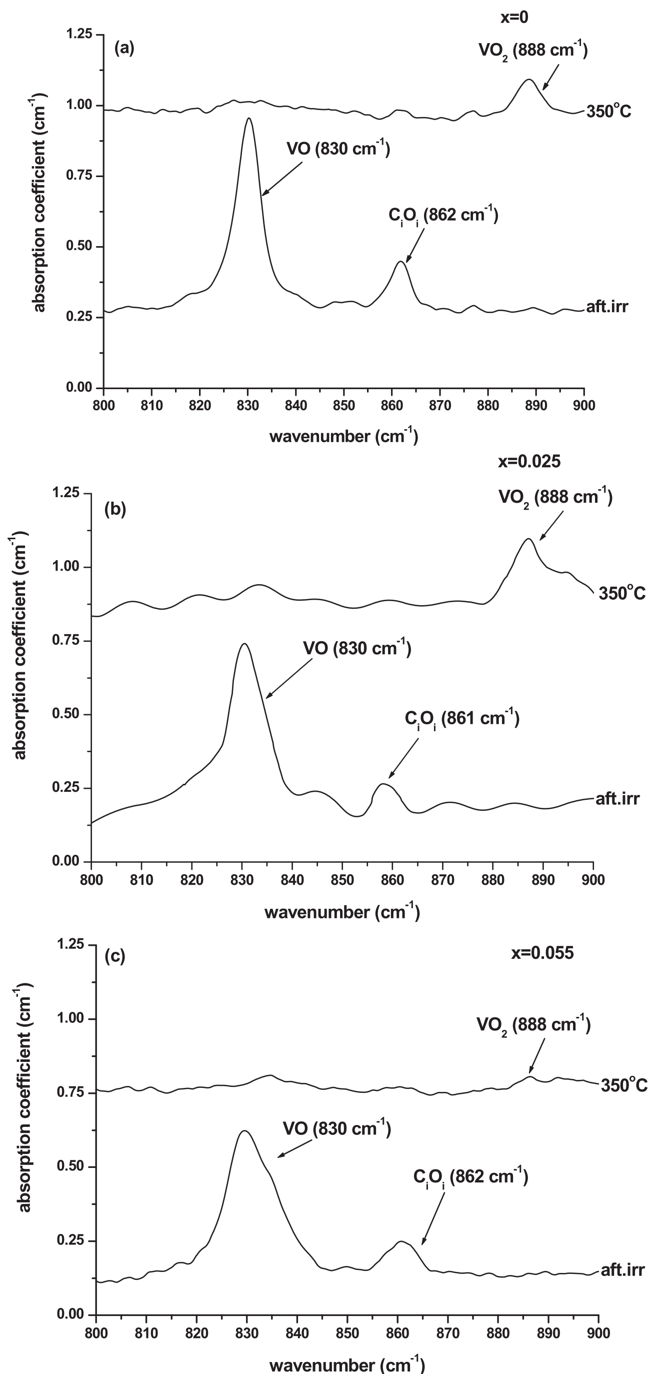

FIG. 1. Segments of the IR spectra of the *n*-type $Si_{1-x}Ge_x$ crystals (x = 0 (a), 0.025 (b), 0.055 (c)) samples after irradiation and at 350 °C, in the course of the 20-min isochronal anneals sequence.

that survive annihilation with self-interstitials (Si_I's) are captured by oxygen impurities to form *V*O pairs ($V + O_i \rightarrow VO$). On the other hand, the survived Si_I's are captured by carbon atoms converting them to carbon interstitials ($C_s + Si_I \rightarrow C_i$), which are very mobile and are readily captured by oxygen atoms to form the C_iO_i defect ($C_i + O_i \rightarrow C_iO_i$). The same is expected in $Si_{1-x}Ge_x$, and this is manifested in the spectra after irradiation by the presence of the IR bands at 830 cm^{-1} and 862 cm^{-1} related with the *V*O and C_iO_i defects, respectively (Fig. 1). The latter defect is out of the scope of the present investigation. Upon annealing, the well-known 888 cm^{-1} band of the VO_2 pair is observed. The latter defect is formed upon annealing of the *V*O defect and its subsequent trapping by O_i atoms according to the reaction $VO + O_i \rightarrow VO_2$. In Fig. 1 we also present the IR spectra at 350 °C. This is a characteristic temperature where the *V*O band has almost completely disappeared from the spectra and the VO_2 band at 888 cm^{-1} is now present. Notably, the amplitudes of the *V*O band are larger in the sample without Ge (x = 0). With the increase of Ge the strength of the *V*O band decreases (compare spectra with x = 0.025 and x = 0.055). Furthermore, the larger the x the more disturbed the shape of the *V*O band. Fig. 2 exhibits the evolution with the annealing temperature of the *V*O and the VO_2 defects of the samples used in these studies. Regarding the amplitudes of the *V*O defect, it is immediately seen that they decrease with the increase of the Ge content, something mentioned above from the inspection of Fig. 1. Regarding the VO_2 defect we observe that it begins to grow from ∼250 °C onwards, indicating its relation with the *V*O defect which is substantially decreased from this temperature ($VO + O_i \rightarrow VO_2$). Significantly, the VO_2 band continues to grow even after the disappearance of the *V*O band from the spectra at around ∼360 °C. This is a strong indication for the existence of alternative mechanisms and reaction processes that contribute also in the formation of the VO_2 defect. These mechanisms and the fact that the concentration of VO_2 defect at x = 0 is smaller than that of x = 0.025 and larger than that of x = 0.055 are further discussed below. At first sight, we observe that the *V*O defect in the $Si_{1-x}Ge_x$ samples begins to decay at around 180 °C in comparison with the substantial higher temperature of ∼250 °C in the Si sample. Additionally, an inspection of the spectra (refer to Fig. 1) reveals that after irradiation the *V*O peak is broader in the case of the $Si_{1-x}Ge_x$ samples as compared with Si. This is an indication that the *V*O band in $Si_{1-x}Ge_x$ is perturbed due to the presence of other bands. Thus the evolution of the bands depicted in Fig. 2 might not reveal all of the details of the involved processes and further analysis of the available data is necessary for a more accurate representation of the phenomena taking place. The shape of the *V*O band is distorted and changes with the

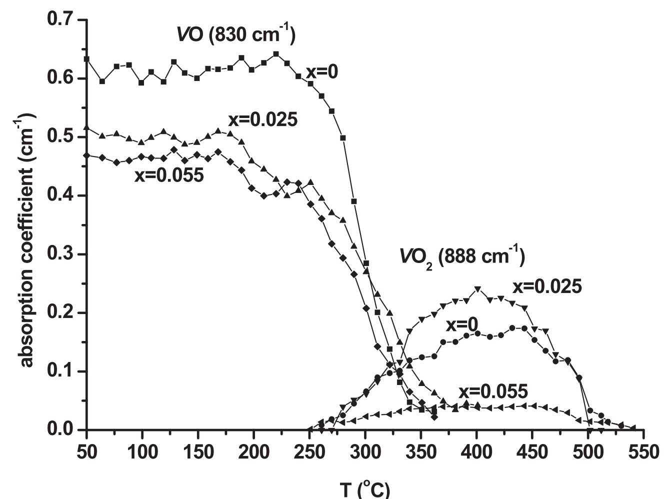

FIG. 2. The thermal evolution of *V*O and VO_2 in *n*-type $Si_{1-x}Ge_x$ crystals (x = 0, 0.025, 0.055) in the course of the 20 min isochronal anneals sequence.

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increase of x due to the presence of other bands in the same spectral range. Apparently, the overlapping of various bands in the same frequency range is a serious additional obstacle in determining their identity, their thermal evolution and stability and their properties in general. Moreover, when there are weak peaks in the same spectra range their signal could be masked completely by stronger signals. A usual way to resolve this issue and recover the contribution of each individual component in the total signal is by making computer deconvolution using Lorentzian profiles. Fig. 3 is the Lorentzian deconvolution of the A-center frequency range of the IR spectra depicted in Fig. 1 for the x = 0.055 sample. The analysis shows the existence of two other bands at ~834 and ~839 cm^{-1}. It is clearly seen that with the increase of annealing temperature the amplitude of the bands changes. At around ~200 °C, the amplitude of all the bands is more or less the same (refer to Fig. 3) indicating how important is the analysis of complex bands in their constituents. The 834 and ~839 cm^{-1} pair of bands has been detected previously.[5] To be precise the two bands are shown in Fig. 3 of Ref. 5 without reporting their frequencies there, but in the text they are identified with frequencies at 834.6 and 839.2 cm^{-1}. They were attributed to *V*O pairs being situated near Ge atoms in the first neighbor position (839 cm^{-1}) (denoted by $(VO\text{-}Ge)_1$ thereafter) and the second neighbor position (834 cm^{-1}) (denoted by $(VO\text{-}Ge)_2$ thereafter). The (*V*O-Ge) association has also been investigated[6] by Deep Level Transient Spectroscopy (DLTS) coupled with DFT calculations. These results confirmed fully the attribution of the two IR bands at 839 and 834 cm^{-1} to the $(VO\text{-}Ge)_1$ and $(VO\text{-}Ge)_2$ structures, respectively. As a result of the formation of (*V*O-Ge), the *V*O production in the $Si_{1-x}Ge_x$ (x = 0.025, 0.055) samples is lower than that in Si.

Fig. 4 shows the evolution of the 830, 834, and 839 cm^{-1} bands as a function of temperature in the course of 20 min isochronal anneal sequence. Since it is more likely for the Ge atom to be situated in the second nearest position with respect to *V*O than in the first, it is expected that more $(VO\text{-}Ge)_2$ defects form than $(VO\text{-}Ge)_1$ and this is further experimentally demonstrated by the fact that the IR band of 834 cm^{-1} correlated with

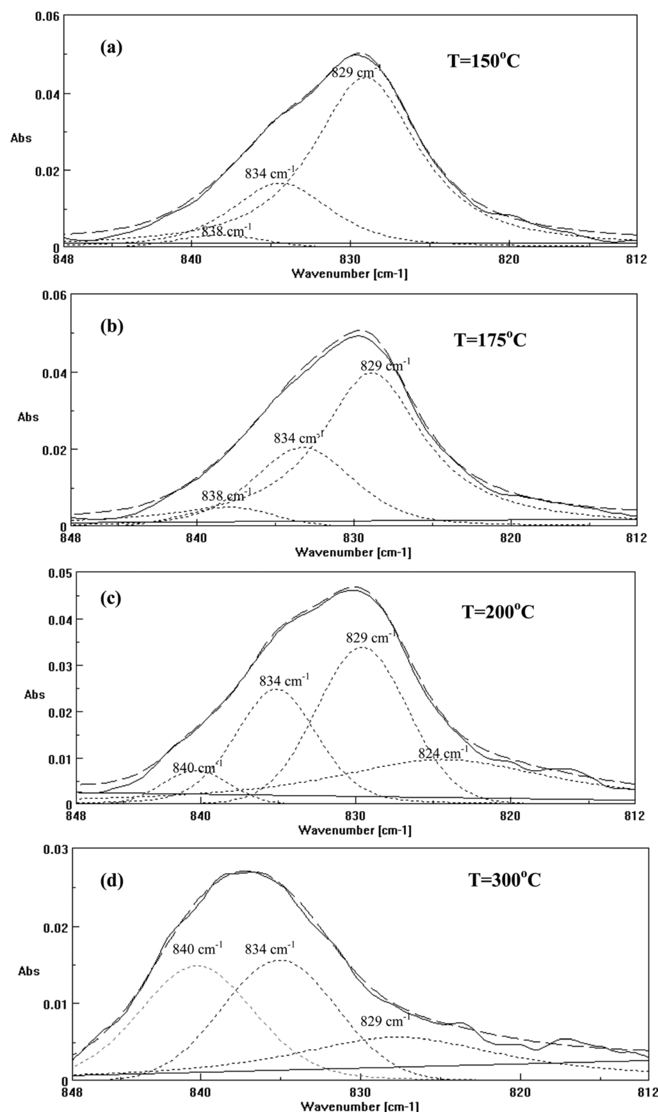

FIG. 3. Deconvolution of the A-centre region IR bands into Lorentzian profiles, at selected temperatures (150 °C (a), 175 °C (b), 200 °C (c) and 300 °C (d) of the isochronal anneal sequence for the x = 0.055 sample. Solid lines represent experimental results; dashed lines the best fitting curves; dotted lines the Lorentzian profiles.

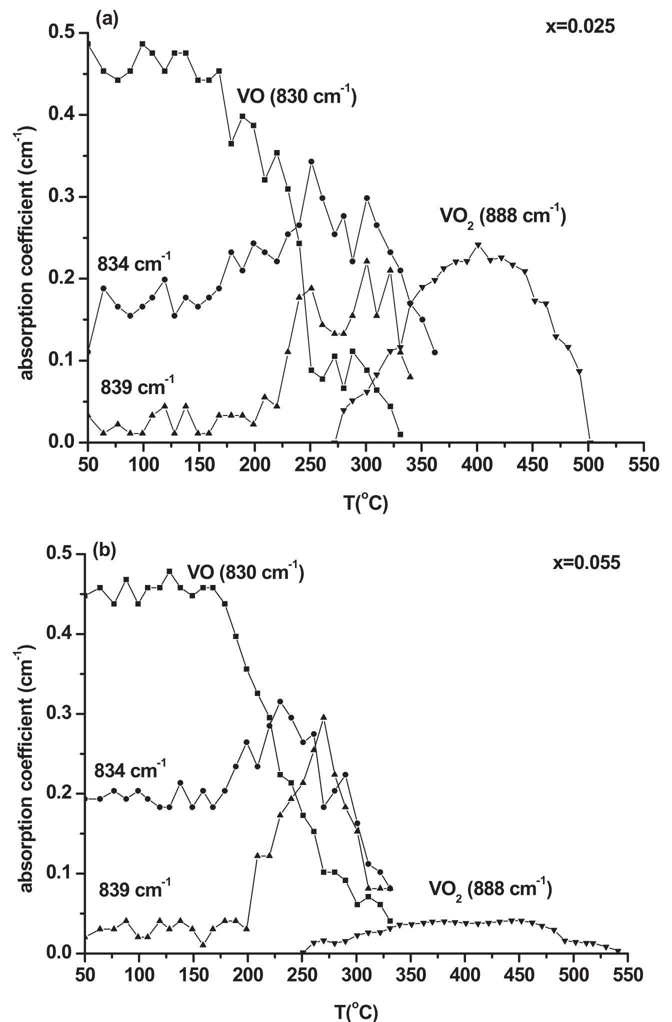

FIG. 4. The evolution of the *V*O, $(VO\text{-}Ge)_1$ and $(VO\text{-}Ge)_2$ bands in *n*-type $Si_{1-x}Ge_x$ crystals (x = 0, 0.025, 0.055) in the course of the 20 min isochronal anneals sequence.

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the $(V\text{O-Ge})_2$ structure is stronger than that of $839\,\text{cm}^{-1}$ band of $(V\text{O-Ge})_1$ structure. Notably, comparing the intensities of the two bands, we are not able from the present data to make any definite conclusion regarding their dependence with respect to the Ge content. One would expect their amplitudes to be stronger in the sample with the higher Ge content, due to the higher probability of *V*O pairs to associate with Ge atoms. Indeed, in the temperature range after the irradiation up to $\sim$180 °C, the amplitude of the stronger band at $834\,\text{cm}^{-1}$ is higher on the average in the $x = 0.055$ sample than that of the $x = 0.025$ sample. However, above that temperature the intensities of the bands increase until they reach in both samples almost similar values (refer to Fig. 4). In the case of the other weaker band at $839\,\text{cm}^{-1}$, the picture is less clear. Apparently, further investigation is needed involving samples with larger Ge content to clarify this issue.

It is observed from Fig. 4, that the *V*O peak begins to decay at around $\sim$180 °C, in the $x = 0.025$ and 0.055 samples, a temperature that is substantially lower than that of the Si sample ($\sim$250 °C). Simultaneously the intensity of the bands at 834, $839\,\text{cm}^{-1}$ increases. This is an indication of further (*V*O-Ge) formation due to localization of *V*O pairs near Ge atoms.[5] In $Si_{1-x}Ge_x$ material the Si-Ge and Ge-Ge bonds are weaker as compared to the Si-Si bonds in Si.[19] Consequently, the *V*O pair is expected to migrate more easily in the $Si_{1-x}Ge_x$ lattice. The migrating *V*O pairs are trapped by the Ge atoms forming $(V\text{O-Ge})_1$ and $(V\text{O-Ge})_2$ structures. Importantly, the Ge concentration is much larger than the O_i concentration, which favors the formation of (*V*O-Ge) structures. Furthermore, the trapping of *V*O pairs by Ge is consistent with previous DFT results, which calculated that the association of *V*O pairs is very strong with Ge atoms in Ge-doped Si.[20]

Around 250 °C, both the *V*O and (*V*O-Ge) bands begin to decrease. Their decay is accompanied in the spectra by the growth of a band at $885\,\text{cm}^{-1}$ attributed to the VO_2 cluster (via the reaction $VO + O_i \rightarrow VO_2$). However, the shape of the VO_2 band is disturbed (Fig. 1(b)) indicating the existence of additional bands in this spectral region. Indeed, deconvolution to Lorentzian profiles (Fig. 5) shows that two other bands at $\sim$898 and $893\,\text{cm}^{-1}$ contribute to the initial band of VO_2. These two bands have been previously detected[21] and attributed to $(VO_2\text{-Ge})$ structures. We envisage that at 250 °C (*V*O-Ge) structures begin to anneal out contributing to the formation of VO_2 and $(VO_2\text{-Ge})$ structures which give rise to a disturbed IR band around $885\,\text{cm}^{-1}$. The temperature of annealing of the *V*O-Ge structures is higher as compared to *V*O by about 70 °C and this reflects the extra thermal energy required to overcome the binding of the *V*O pair to the Ge atom.

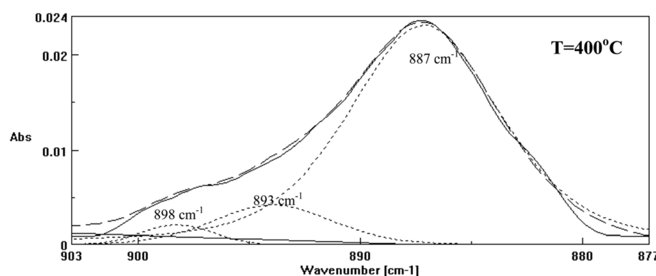

FIG. 5. Deconvolution of the VO_2 defect region IR bands into Lorentzian profiles, at a representative temperature of the isochronal anneal sequence for the $x = 0.025$ sample. The solid line represents experimental results; dashed lines the best fitting curves; dotted lines the Lorentzian profiles.

Fig. 6 shows the absorption coefficient of *V*O and VO_2 defects as a function of Ge content. The IR spectra were recorded at room temperature and therefore absorption coefficient values for the *V*O and VO_2 defects for each sample refer to this temperature. The production of *V*O decreases with the increase of Ge consistently with previous reports.[5,6] Interestingly, we determine that the formation of VO_2 defects is more pronounced in the intermediate value of Ge ($x = 0.025$) than that of Ge ($x = 0.055$). A complete understanding of the above reported results requires to take into account all the reaction processes and the respective mechanisms involved in the annealing of the *V*O band and the corresponding growth of the VO_2 band. Thus, the shape of the curve (b) in Fig. 6 essentially reflects the impact of Ge, on the mechanism that governs the two main reactions ($VO + O_i \rightarrow VO_2$, $VO + Si_I \rightarrow O_i$), which mainly contribute to the *V*O annealing and its conversion to the VO_2 defects. Ge plays a key role on the balance between the two reactions, probably affecting the availability of self-interstitials participating in the second reaction thus determining the percentage of *V*O pairs that convert to VO_2 defects. However, although the *VO* related bands disappear from the spectra at $\sim$330 °C, the VO_2 band continues to increase up to $\sim$400 °C. This is a strong indication that its formation involves additional reaction processes. The phenomenon particularly seen for the $x = 0.025$ sample (Fig. 4(a)), but for the $x = 0$ sample as well (Fig. 1), has been also reported[22] in Ge-doped Si. It has been suggested[23] that reactions involving oxygen dimmers, as well as divacancies[22] may provide additional channels for the VO_2 formation in Si. It is not unreasonable to extend the possibility of such reactions to take place also in $Si_{1-x}Ge_x$ material. It is important to note that the VO_2 band has contributions from the $(VO_2\text{-Ge})$ bands (Fig. 5). In that respect, the VO_2 curve in Fig. 6 represents the total

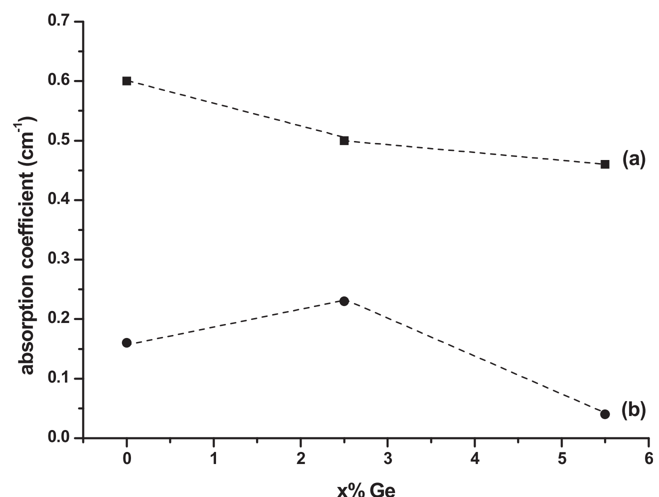

FIG. 6. The production of *V*O pairs (a) and the formation of VO_2 defects (b) with respect to the Ge content for *n*-type $Si_{1-x}Ge_x$ crystals ($x = 0$, 0.025, 0.055).

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number of VO_2 and VO_2-related structures formed in the course of annealing of the VO pair and not only those formed by the reaction $VO + O_i \rightarrow VO_2$.

Importantly, it has been previously established by Lindström and Svensson[8] that about half of the VO pairs are not converted to VO_2 defects. This may indicate that besides a number of VO pairs that dissociate ($VO \rightarrow V + O_i$), another part may be trapped to unknown sinks that cannot be detected. Upon annealing at temperatures above 330 °C, these structures may dissociate liberating additional VO that contribute to the further formation of VO_2 defects. Remarkably, the increase of the VO_2 defects above 330 °C is practically insignificant in the case of the sample with $x = 0.055$ (Fig. 4(b)), indicating that the above mentioned mechanisms contributing to the further VO_2 formation are largely suppressed in the case of the sample with high Ge content $x = 0.055$. The phenomenon of the VO_2 formation is more complicated than it was initially anticipated and further investigations are necessary to clarify the issue.

IV. SUMMARY

FTIR spectroscopy was used to investigate the production and evolution of A-centers in *n*-type $Si_{1-x}Ge_x$ ($x = 0$, 0.025, 0.055). It is determined that A-centers in $Si_{1-x}Ge_x$ anneal at lower temperatures as compared to Si, whereas their concentrations is lower. Both these effects can be attributed to the weaker bonds (Si-Ge and Ge-Ge) present in $Si_{1-x}Ge_x$.

Lorentzian analysis has shown that the total VO signal in the IR spectra consists of the following contributions: (i) VO surrounded by Si atoms up to the next nearest neighbor sites, (ii) $(VO\text{-Ge})_1$ that is VO with Ge atom(s) only at the nearest neighbor site, and (iii) $(VO\text{-Ge})_2$ that is VO with Ge atom(s) at the next nearest neighbor site. The association of VO with Ge to form these $(VO\text{-Ge})_1$ and $(VO\text{-Ge})_2$ structures is strong, leading to higher annealing temperatures for these centers. This in turn is consistent with previous DFT results in Ge-doped Si.[20]

When annealing $Si_{1-x}Ge_x$ at temperatures ~180 °C, the VO pairs migrate (this is a lower temperature compared to Si) and they begin to associate with the Ge atoms in the lattice. The driving force for this association is the attraction of the V (in the VO pair) with the Ge atom. This is reflected by the increase of the $(VO\text{-Ge})_1$ and $(VO\text{-Ge})_2$ structures as the VO pairs decrease (Fig. 4). The next step occurs at around 250 °C where the VO pairs and the $(VO\text{-Ge})_1$ and $(VO\text{-Ge})_2$ structures anneal out and contribute to the formation of VO_2 and $(VO_2\text{-Ge})$ defects.

The absorption coefficient a_{vo2} is larger for $x = 0.025$ than $x = 0.055$ an indication of the impact of Ge on the balance between the reactions $VO + O_i \rightarrow VO_2$, $VO + Si_I \rightarrow O_i$ that mainly control the anneal of VO defect. Importantly, Ge seems to play a significant role on the availability of self-interstitials thus affecting the involved processes related in the formation of the VO_2 defect. Furthermore, the presence of Ge leads to the formation of $(VO_2\text{-Ge})$ structures. Finally, it is suggested that when Ge content increases in the $Si_{1-x}Ge_x$ material, alternative reaction channels that also lead to the VO_2 defect formation are substantially suppressed.

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