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Low-temperature irradiation-induced defects in *p*-type germanium

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Besides two well-known dominant peaks, which have been earlier correlated with the single monovacancy and gallium interstitial defect, a number of small lines appear in deep level transient spectroscopy (DLTS) spectra measured *in situ* after particle irradiation at low temperatures in *p*-type germanium. Three of the most pronounced have been studied to some detail in the present investigation by combining DLTS and high-resolution Laplace DLTS. These three lines are called H70, H280 and H290, where the H refers to their hole-trap nature, and the numbers to their apparent enthalpy for hole emission relative to the valence-band edge. The H70 trap is most probably a primary defect, and its observed energy level is found to be an acceptor level. Possible defects related to these DLTS lines are discussed with special emphasis on the divacancy defect in germanium.

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I. INTRODUCTION

Continued progression toward smaller feature size of silicon metal-oxide-semiconductor transistors in very-large-scale-integrated circuits has resulted in a quest for higher channel mobilities. The exploitation of Ge, which shows considerably higher low-field hole and electron mobilities, is one of the most prominent solutions of this task. However, to be able to utilize the full potential of Ge many issues should be understood. One of the most important clues is related to the elementary point defects in germanium, which have attracted intensive research for a long time.

In our previous studies we demonstrated that *in situ* deep level transient spectroscopy (DLTS) and high-resolution Laplace DLTS are crucial for the study of electrical properties of elementary point defects created in low-temperature electron irradiated *p*- and *n*-type Ge.[1,2] We demonstrated that four lines appeared in the DLTS spectrum in the case of electron-irradiated *n*-type Ge. The first peak, FP, was shown to stem from the Frenkel pair. The other dominant peak, $SbV^{--/-}$, was correlated with the well-known double-acceptor state of the E center. Besides these dominant peaks the lines labeled A and B were concluded to originate from different charge states of the self-interstitial. The situation is more complex in electron-irradiated *p*-type Ge. In this case two dominant peaks were also observed in the conventional DLTS spectrum. These peaks were assigned to the double-acceptor state of the monovacancy and to the single donor state of interstitial gallium. However, besides these two dominant peaks some minor features were also observed. These minor peaks were not further discussed in Refs. 1 and 2; they play, however, the lead role in the present investigation.

II. EXPERIMENTAL PROCEDURE

N^+p diodes for this study were prepared from gallium doped, oxygen and carbon lean Ge crystals from UMICORE. A n^+ top layer was grown by molecular beam epitaxy (MBE) and mesa diodes were made by photolitography and chemical etching following the procedure described in Ref. 3. The gallium concentration was 4×10^{14} cm^{-3} in the *p*-type Ge substrate. Electron, proton, and alpha-particle irradiations were done at 2 MeV to different doses as specified below while the diodes were held at 22 K. Beam-current densities were about 200 nA/cm^2 for electrons whereas they were significantly reduced to be about 10 nA/cm^2 for both protons and alpha particles. The irradiated samples were subjected to thermal isochronal annealing in the temperature range of 50–200 K. *In situ* conventional deep level transient spectroscopy and high-resolution Laplace DLTS were used to analyze the resulting deep electronic levels. No deep levels were detected prior to irradiation.

III. EXPERIMENTAL RESULTS

Figure 1(a) shows a conventional DLTS spectrum recorded *in situ* after low-temperature 2 MeV electron irradiation to a dose of 4×10^{14} cm^{-2} at 22 K. This condition resulted in the appearance of two dominant peaks labeled $V(--/-)$ and $Ga(0/+)$, and three minor features labeled H70, H280, and H290, respectively. As mentioned above the two dominant lines are well known, and have been assigned previously to the double-acceptor level of the vacancy $V(--/-)$ and to the interstitial gallium defect $Ga_i(0/+)$ formed as a result of the Watkins-replacement mechanism in irradiated *p*-type Ge.[1,2] No discussion as to the origin of the H70, H280, and H290 peaks were presented in the previous works. After a short annealing at room temperature the DLTS spectrum shown in Fig. 1(a) is significantly simplified. In this case only the $Ga_i(0/+)$ line remains in the spectrum whereas the other peaks observed just after irradiation disappear. This is in a good agreement with our previous observations. Indeed, as shown in Ref. 1 the $V(--/-)$ defect is stable at temperatures below 200 K and afterwards it starts to migrate forming complex defects such as, for example, the E center. Also this is consistent with results reported in Ref. 4 in which low-temperature alpha-particle irradiated n^+p-mesa

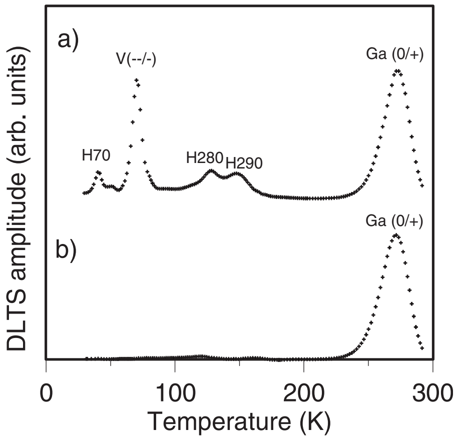

FIG. 1. DLTS spectra of n^+p-mesa diodes after 2 MeV electron irradiation to a dose of 4×10^{14} cm^{-2} at 22 K. Spectrum (a) was recorded *in situ* after the electron irradiation whereas spectrum (b) was recorded after a short annealing under reverse bias at room temperature. The DLTS settings were: $e_n=20$ s^{-1}, $V_R=-8$ V, $V_p=-2$ V (a) and $e_n=20$ s^{-1}. The duration of the filling pulse was 3 ms in both cases.

diodes of Ge were investigated by conventional deep level transient spectroscopy and high-resolution Laplace deep level transient spectroscopy after a short annealing at room temperature. None of the DLTS peaks shown in that work was observed before a temperature of about 210 K was reached. On the other hand, all primary defects created after alpha-particle irradiation were shown to anneal out at a temperature significantly lower than room temperature.

Figure 2 shows the Arrhenius plots of T^2-corrected hole emission rates derived from Laplace DLTS measurements for the H70, H280, and H290 peaks. The electronic levels corresponding to these defects are characterized by an apparent enthalpy for hole emission of $\Delta E_{pe}(\text{H70})=0.07$ eV, $\Delta E_{pe}(\text{H280})=0.28$ eV, and $\Delta E_{pe}(\text{H290})=0.29$ eV and an apparent capture cross section of $\sigma_{pa}(\text{H70})=1.3\times10^{-15}$ cm^2, $\sigma_{pa}(\text{H280})=7\times10^{-15}$ cm^2, $\sigma_{pa}(\text{H290})=1.4\times10^{-13}$ cm^2, respectively. The apparent capture cross sections are assumed to be temperature independent for all these defects.

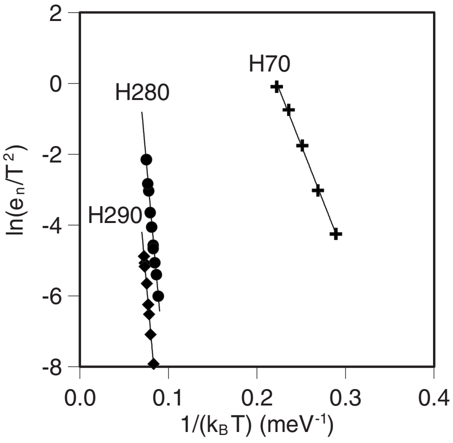

FIG. 2. Arrhenius plots of the H70, H280, and H290 signals depicted in Fig. 1(a).

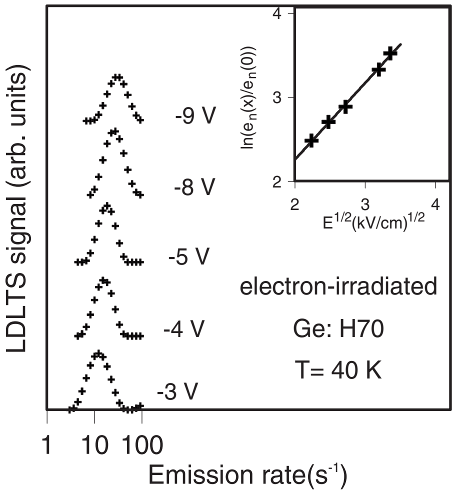

FIG. 3. Laplace DLTS spectra recorded at 40 K for different values of the bias voltage applied to the diode. The inset shows electric-field dependence of emission rates of the H70 peak demonstrating its acceptor character where the shift in rate as a function of square root of the electric field inside the depletion layer of the diode is shown.

In order to reveal a donor or acceptor character of the H70, H280, and H290 traps we examined the dependence of the Laplace DLTS signal emission rates on the electric field for these defects. As seen in Fig. 3 the H70 line shifts toward higher emission rates with increasing reverse bias applied to the diode. The inset in this figure shows the emission rates of the H70 defect plotted on a logarithmic scale as a function of the square root of the electric field inside the depletion layer of the diode. The observed linear dependence is in good agreement with the Poole-Frenkel effect, which describes the increase of the thermal emission rate of carriers in an external electric field due to the lowering of the barrier, associated with the Coulomb attraction.[5] Therefore, one can conclude that the H70 trap is, at least, negatively charged before it captures a hole from the valence band and, therefore, this defect introduces either a single acceptor or double-acceptor state into the band gap of Ge.[6] Moreover, in this case the single acceptor state seems to be the most plausible candidate due to the fact that the apparent capture cross section of H70 is about 10^{-15} cm^2. No field effect was observed for the H280 and H290 lines even in the case when four times higher doping level was used.

Thus, from these results both H280 and H290 seem to behave as donorlike defects in p-type Ge. However, taking into account the much higher apparent capture cross section observed for these defects compared to that of H70 this assumption seems to be very unlikely. On the other hand the absence of a field effect for acceptorlike defects in p-type Ge can be related to the following phenomenon. In the frame of the Poole-Frenkel effect, the emission rate should increase in an electric field as

$$\ln[e(E)/e(0)] \propto \varepsilon_{\mathrm{PF}}/k_B T, \qquad (1)$$

where the lowering of the barrier $\varepsilon_{\mathrm{PF}}$ is given by $\varepsilon_{\mathrm{PF}}=2\mathrm{sqrt}(Zq^3E/\varepsilon)$. Here, Z is the charge of the defect, q is the

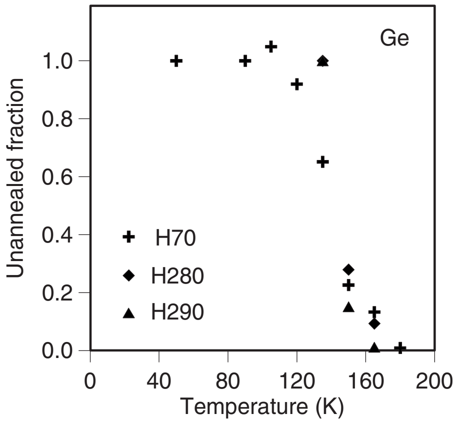

FIG. 4. The normalized change in concentration of the H70, H280, and H290 traps upon 10 min isochronal annealing steps. During each step the bias was on.

electron charge, E is the electric-field strength and ε is the dielectric constant.[6,7]

Hence the Poole-Frenkel effect is more pronounced for levels observed at low temperature. This statement can also be valid when different energy levels of defects, e.g., single and double acceptors are observed at different temperatures. Thus, for example, if the single acceptor state is observed at about 40 K and the double-acceptor state appears at about 150 K one can expect that the field effect for the single acceptor state should be more pronounced (about 2.7 times) as compared to that observed for the double-acceptor state at the higher temperature (see the above equation). Due to this weaker field effect it is not possible to rule out the presence of field effects for H290 and H280 even with the highest applied bias. Therefore, based on the apparent capture cross sections it is possible that also H290 and H280 are acceptor-like defects in p-type Ge.

Figure 4 shows the normalized change in concentration of the H70 trap upon 15 min isochronal annealing steps. During each step the bias was on. Corresponding points for H280 and H290 are included as well. Here one should keep in mind that the annealing temperature of H280 and H290 is very close to 150, i.e., the temperature range in which these defects were observed with conventional DLTS as well as Laplace DLTS. Assuming that the annealing temperature of a defect is taken as the temperature at which the population is reduced to 50% then in the present case the annealing temperature of H70 is about 140 ± 5 K whereas it is about 147 ± 5 K for H280 and H290. If one assumes that a part of H280 and H290 anneals out before they appear in the DLTS spectrum then very similar anneal temperatures are found for all three defects. On the other hand, intensities of the DLTS peaks corresponding to these defects do not always follow to those observed for the H70 peak, as one can expect if these peaks originate from the same defect. This was observed after proton and alpha-particle bombardments and after electron irradiation in samples with about four times higher doping level. Thus, it is a plausible case that H70, H280 and H290 belong to different defects.

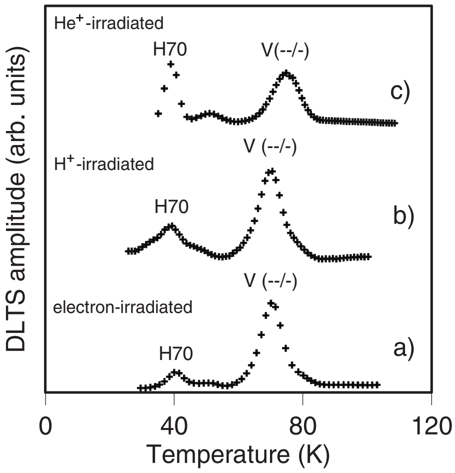

FIG. 5. DLTS spectra of $n^{+}p$-mesa diodes after, and 2-MeV electron (a), 2 MeV proton (b) and 2 MeV alpha-particle (c) irradiations to a dose of 4×10^{14}, 1×10^{10}, and 1×10^{10} cm^{-2}, respectively, at 22 K.

Figure 5 shows a part of the DLTS spectra of $n^{+}p$-mesa diodes after irradiation with 2 MeV electrons (a), 2 MeV protons (b), and 2 MeV α-particles (c) at 20 K. The conventional DLTS spectra recorded during the heating up from 100 K to room temperature after 2 MeV H and 2 MeV He implantation were very complicated with a large number of unstable peaks; hence, we cannot unambiguously conclude how the intensity of the H280 and H290 peaks changes in this case, and for the sake of clarity we do not present this part of the DLTS spectrum in Fig. 5. In the case of 2 MeV protons and α particles the low-temperature part of the spectra are similar to the one shown in Fig. 5(a) after 2 MeV electron irradiation. Thus, the two dominant peaks labeled H70 and $V(--/-)$ were observed in all cases. From the extracted finger prints of the different DLTS peaks of Fig. 5(b), it can be concluded that these peaks stem from the same defects as those in Fig. 5(a). However, the $V(--/-)$ line in Fig. 5(c) is shifted by about 5 K compared to those in Figs. 5(a) and 5(b). Nevertheless, due to the identical annealing behavior we suggest that these peaks produced by proton and α-particle irradiation originate from the same defects as those produced by electron irradiation. The observed shifts are probably related to the higher local strain in the crystal lattice following by the alpha-particle irradiation compared with those observed after proton and electron irradiation. In support of this idea, a similar shift was also observed for the $V(--/-)$ line in proton-irradiated p-type Ge with doses by a factor of about five higher than those presented in Fig. 5(b).

As seen in Fig. 5 using heavier particles results in an increase of the H70 peak relative to the $V(--/-)$ line. This demonstrates that H70 is a more complex defect compared to the monovacancy in Ge. Of course, one should remember that $V(--/-)$ is the double-acceptor state of the monovacancy observed close to the temperature range in which this defect is able to migrate assisted by an electric field.[1] However, the electric field in these $n^{+}p$-mesa diodes is only on the order of 10^{3} V/cm. Due to such a low field the field assisted drift is suppressed, and we can therefore use this peak as a reference level in the present investigation.

IV. DISCUSSION

In the following, we address the possible identification of the observed lines. First of all we can rule out that different charge states of single interstitials can be the origin of these peaks. As seen in Fig. 1 the intensity of $Ga_i(+/0)$ is much higher than those of H70, H280, and H290. Further it is believed that $Ga_i(+/0)$ is formed as a result of the Watkins-replacement mechanism. In this case, one would expect that the number of interstitials should be comparable to that of interstitial gallium observed as $Ga_i(+/0)$ whereas the ratio between $Ga_i(+/0)$ and H70 is about 20 in the present investigation. Using similar arguments one can rule out the monovacancy as the origin of these defects. On the other hand H70, H280, and H290 appear in the DLTS spectrum before the monovacancy and the self-interstitial anneal out in Ge. This means that none of H70, H280 and H290 can be related to defects having either the single self-interstitial or the monovacancy in their structure. In low-temperature electron-irradiated *p*-type Ge only simple defects such as self-interstitials, monovacancies, divacancies, and di-interstitials are expected to be created. As these become mobile additional simple complexes can form. Due to this, one can expect that H70, H280, and H290 are either primary defects created in the collision cascade just after electron irradiation or secondary defects, which are formed by diffusion of irradiation-induced defects at temperatures lower than the diffusion temperature of the vacancy and self-interstitial in Ge. To our knowledge no defects, which could diffuse faster than monovacancies and self-interstitials, have been observed in pure Ge. In this case H70, H280, and H290 seem to be primary defects and could be related to either the divacancy or the di-interstitial.

However, no electrically active levels of the di-interstitial have been unambiguously identified in Si and Ge. Due to this, in the following we will concentrate on the discussion of possible precursors of the observed peaks with special emphasis on the divacancy defect in germanium.

Thus, three divacancy levels have been observed in pure Si: a single donor level $E(0/+)=E_V+0.2$ eV, a single acceptor level $E(-/0)=E_C-0.42$ eV and a double-acceptor level $E(--/-)=E_C-0.23$ eV. Using *ab initio* density functional studies Coutinho *et al.*[8] predicted that also three levels of the divacancy should exist in Ge. A single donor level was predicted to be close to the valence band at about $E(0/+)=E_V+0.08$ eV whereas two acceptor levels were predicted to be close to the middle of the band gap: $E(-/0)=E_C-0.27$ eV and $E(--/-)=E_C-0.4$ eV. In agreement with these predictions, in several experimental studies the divacancy was suggested to have an energy level in the upper half of the Ge bandgap.[9–11] In our previous work, however, we demonstrated that none of the earlier observed traps, which are stable at room temperature, could be related to the divacancy in neither *p* nor *n*-type Ge irradiated with electrons and alpha particles.[4,12] Based on these arguments and in analogy with Si where the divacancy is one of the dominant peaks observed in the DLTS spectrum after low-temperature electron irradiation we assume that this defect anneals out before room temperature is reached in Ge. On the other hand no levels, which could be correlated with the divacancy, were observed in *n*-type Ge irradiated with electrons at 22 K[1] and, therefore, the conclusion that the divacancy has no electrical levels in the upper half of the band gap could be drawn.[4] Following this idea one can expect that divacancy-related levels are in the lower part of the band gap.

As mentioned above, according to theoretical predictions the divacancy should introduce three electrically active levels into the band gap of Ge.[8] However in this case the donor level of V_2 was predicted to be very close to the valence band (about 0.08 eV). Taking into account the uncertainty of *ab initio* calculations in determination of the position of the energy levels it is possible that this donor level can either be resonant with the valence band or too shallow to be observed using the capacitance techniques. Assuming that the donor level is resonant with the valence band is in good agreement with the results obtained by av Skardi *et al.*[13] They followed the position of three energy levels of the divacancy in epitaxially grown, strain-relaxed $Si_{1\text{-}x}Ge_x$ alloys as a function of x for $0 \leq x \leq 0.5$. Generally, it is accepted that the reduction of the band gap is entirely reflected in the valence-band shift in such alloys.[14] Following this idea they concluded that the donor level of the divacancy, which is located about $E_V+0.2$ eV in pure Si, remains pinned to the conduction band in $Si_{1\text{-}x}Ge_x$ alloys. This means that this donor level becomes gradually shallower with increasing x and, moreover, it was found to become resonant with the valence band at about $x=0.5$. On the other hand two acceptor levels in the upper part of the band gap were shown to follow each other and remain approximately equidistant from each other by a so-called Hubbard energy of 0.2 eV. The single acceptor level was found to cross midgap at $x \approx 0.25$ and, afterward, it appeared as a level in the lower part of the band gap. Following the trends observed by av Skardi *et al.*[13] one can assume that the single donor level of the divacancy should be resonant with the valence band whereas the single and double-acceptor levels should be located in the lower part of the band gap of pure Ge. Moreover, similar to silicon these two acceptor levels could be expected to be separated by a Hubbard energy of about 0.2 eV. Then considering the field dependence of the emission rate observed for the H70 peak one can tentatively assign this signal to the single acceptor state of V_2 whereas the double-acceptor state of the divacancy should be located near the middle of the band gap. In support of this assignment is the observation of Fig. 5 in which H70 was concluded to be a more complex defect than the monovacancy. Irradiating with heavier particles such as H^+, the defect density in the collision cascade along the particle track will be denser[15] than in the case of electrons, increasing thereby the probability of forming divacancies as a primary defect.

If it is assumed that H70 is the single acceptor state of the divacancy, from the reasons mentioned above we rule out that H280 or H290 might be candidates for the double-acceptor state of the divacancy. However, in this case the double-acceptor state could be located deep enough in the band gap and, therefore, cannot be observed in the DLTS spectrum due to the possible low annealing temperature of this defect. Indeed, if H290 having an apparent capture cross section about 10^{-13} cm^2 is observed about 150 K in the DLTS spectrum then due to variation in the capture cross section one can imagine a situation when the double-acceptor state of the divacancy with an approximately similar activation energy could be shifted toward higher temperatures in the DLTS spectrum.

In Ref. 16 Stein found an absorption band, which he tentatively attributed to an internal electronic transition of the divacancy in Ge. This signal was found to anneal out at a higher temperature (200 K) that has been observed in the present work. Stein used optical spectroscopy studies following H^+, O^+, and B^+ ion bombardment to a dose of about 10^{15} cm^{-2} with an energy of 400 keV whereas an electron irradiation with significantly lower doses was used in the present investigation. Although a difference of 50 K in anneal temperature is significant, it could well be the same defect observed by Stein and in the present work considering the very different defect concentrations in the two cases.

An anneal temperature of 150 K for the divacancy is much smaller than that predicted by density functional theory (DFT) cluster calculations. Thus, in Ref. 17 Janke *et al.* estimated energy barriers for migration and dissociation of the divacancy. The dissociation energy, consisting of the binding energy between two vacancies plus the migration energy of a single vacancy, was determined to be about 1.5–1.7 eV, whereas the migration barrier of the divacancy was calculated to be about 1.1 eV. This corresponds approximately to a thermal stability of the divacancy up to 420 K.

How is it then possible that the divacancy in Ge anneals at such a low temperature as suggested from the present investigation? The most plausible explanation is the following: after the irradiation with 2 MeV electrons at cryogenic temperatures the diode capacitance increases significantly for the whole range of applied bias. This is correlated with the introduction of a large concentration of negative charge into the depletion region of the n^+p diode, which adds up with the negatively charged dopant (Ga^-). In Ref. 2 we demonstrated that this observation can only be due to the presence of an equal number of doubly negatively charged monovacancies[1,18] and singly positive charged self-interstitials[1,18] which are believed to be the dominant defects after electron irradiation. On the other hand, a short annealing at about 200 K can significantly reduce the number of these negatively charged traps observed in the CV profile. Irradiating with electrons to doses as specified above, the Fermi level is located below 0.08 eV from the valence band at about 150 K and, thus, the divacancy should be neutral at this temperature in p-type Ge. On the other hand, the self-interstitials are known to be either neutral or at least positively charged[1,18] which leads to the absence of a Coulombic repulsion between them and divacancies. Considering these arguments, it is possible that when the self-interstitials become mobile at 150 K they annihilate with the divacancies. Thus, we suggest that due to the presence of both vacancies and self-interstitials in the crystal lattice after low-temperature electron irradiation these experimental observations suggest a different annealing mechanism than what has been considered by theory.

Moreover, one cannot also exclude that the H70 level might be linked with the double-acceptor state of the divacancy. In this case both the single donor and single acceptor states are assumed to be resonant with the valence band. Indeed, just after electron irradiation two negatively charged vacancies could be stabilized by the surrounding strain, which could allow overcoming the Coulombic repulsion between defects having identical charges. Following this idea the divacancy should be created as a primary defect and is very unstable leading to the low annealing temperature of this defect.

Finally, we cannot determine the origin of the H280 and H290 lines, however, due to their similar annealing behavior it is possible that the levels belong to different charge states of the same defect. In this case one can only speculate that the H280 and H290 peaks might be due to the di-interstitial or a complex having a di-interstitial in its structure.

V. CONCLUSION

In the present study we demonstrated that the H70 peak observed in the conventional DLTS spectrum in electron-irradiated p-type Ge could originate from one of the acceptor states of the divacancy or a di-interstitial-related defect. The trap is characterized by an apparent enthalpy for hole emission of $\Delta E_{pe}(\mathrm{H70})=0.07$ eV and an apparent capture cross section of $\sigma_{\mathrm{pa}}(\mathrm{H70})=1.3\times10^{-15}$ cm^2. If H70 is related to the single acceptor state of the divacancy then the double-acceptor state of the divacancy is rather located much deeper in the band gap and cannot be observed due to the low annealing temperature of the divacancy. Contrary to the Si case the donor state of the divacancy is believed to be resonant with the valence band in Ge. This model seems to be consistent with *ab initio* density functional studies in which also two acceptor levels were predicted to be deep acceptor traps in the band gap of Ge. The discrepancy in the thermal stability of the divacancy compared to DFT cluster calculations is explained by the presence of self-interstitials which, when they become mobile, can annihilate with the divacancies.

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