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FIELD-DEPENDENT NEGATIVE-U PROPERTIES FOR ZINC-RELATED CENTRE IN SILICON

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Field-dependent quenching and sign reversal of photocapacitance have been observed in Zn doped p-type silicon. These findings are interpreted as evidence for self-compensation and metastability properties of the Zn-related centre in silicon. In order to test for this possibility, we consider the effect observed within the framework of the negative-U concept and find that the zinc double-acceptor centre exhibits field-dependent negative-U behaviour. A model that accounts for the properties for the Zn-related centre is proposed; it is based upon the dependence of the electron-vibration interaction constant, which is governed by the linear and quadratic Stark effects on charge states of the centre.

Keywords: A. semiconductors, C. impurities in semiconductors, D. trapping, E. photoelectron spectroscopies

1. Introduction

Deep defects in semiconductors typically have charge and spin correlations that are compensated by local low-symmetry lattice distortions [1–3]. Anderson has shown [4] that the outcome of the compensation for the Coulombic repulsion of two electrons trapped at a deep centre should be a negative$-U$ defect if the centre can trap the electrons such that the second is more strongly bound than the first. This variety of deep defects is then an analogue of the Cooper pair with a small correlation radius. Two versions of the negative-U concept have been proposed to date: a 'soft' version for elastic centres in which electron pairing takes place in the presence of a large lattice relaxation due to the dynamic Jahn–Teller effect [4–6], and a 'hard' version for centres which occur in a rigid lattice where the Coulombic repulsion is compensated, instead, via tunnelling effected by the centre moving between its site and off-site positions [7–10]. Applied, respectively, to chalcogenide glasses [11, 12] and crystalline semiconductors [1–3, 13, 14], both versions have proved successful in understanding the phenomena of self-compensation and metastability, which are characteristic of deep centres present in these materials. In addition to its negative-U character, the deep centre is a system where the interrelationship between electron-vibration interaction and charge correlations gives rise to linear and quadratic Stark effects [14, 15]. In the presence of an external electric field, these effects may greatly enhance, or conversely, depress negative-U properties [14].

In the present paper we report, for the first time, field-dependent negative-U properties of the Zn-related centre in crystalline p-type silicon.

Zinc is one of the more extensively studied double acceptors in silicon of both conductivity types [16–20]. It has been studied by both the thermally stimulated current (TSC) technique [16] and the method of thermal scan of the high frequency junction capacitance (TSCAP) [17]. As a result, the energies of thermal emission of holes from Zn acceptor levels, both of which lie within the band gap, have been determined; they are 0.316 eV and 0.617 eV for $(0/-)$ and $(-/--)$ levels, respectively. The microscopic identity of the centre has yet to be established, though. Two main features of zinc centres are seen in the experimental results obtained by Yau and Sah [16] and Herman and Sah [17]. One is a strong field dependence of the thermal emission rate for holes from the $(0/-)$ level; this dependence can be readily accounted for in terms of the quadratic Stark effect at a deep centre [14]. The second is a trend for the values for thermal emission rates of holes from the two levels to cross with decreasing strength of the electric field applied. In addition, the authors report the detection of a level at $E_v + 0.167$ eV (which they failed to identify, however) at field strengths below the value associated with the crossing. These findings promoted us to surmise that Zn double-acceptor centres have field-dependent negative-U properties which are only displayed at weak electric fields. The $(0/-)$ and $(-/--)$ levels of the centre would lie in negative-U ordering, forming in the silicon band gap the following sequence: $(0/-)-E_v+0.316$ eV and $(-/--)-E_v+0.167$ eV, which cannot be observed at higher electric fields because of the energy barriers induced by the Stark effect between the charge states of the Zn-related centre.

In the present study, a proof of this surmise has been obtained through photocapacitance experiment.

The negative-U ordering for $(0/-)-E_v+0.62$ eV and $(-/--)-E_v+0.5$ eV optical transitions, recorded for the Zn-related centre in p-type silicon at low electric fields, is observed to be enhanced by the corresponding negative-U ordering for thermal emission energies to produce a dramatic increase in the hole emission rate from the Zn^--state, and to give rise to photocapacitance quenching. We interpret these results as being due to a spontaneous negative-U reaction of the form: $2Zn^- \rightarrow Zn^{--} + Zn^0$ which leads to self-compensation for centres with levels inverted according to negative-U ordering. The disappearance of the quenching effect and the reversal of the photocapacitance sign, which are observed at higher electric fields, are interpreted as a result of the negative-U reaction being suppressed by the Stark effect which causes crossing in the energy values for hole emission from the first and the second levels of the Zn-related centre. The change from the negative to a positive sequence of activation energies observed when the electric field was increased provides convincing evidence of the field-dependent negative-U character for the Zn-related centre in silicon.

2. Experiment

The studies were made on n^+p junctions, fabricated by the conventional planar technology, with had impurity concentration profiles being nearly uniform. These diodes duplicate those used in previous TSC and TSCAP studies [16, 17]. Boron was used to provide a shallow acceptor majority impurity. Diffusion of zinc was carried out in sealed quartz ampoules evacuated to 10^{-6} Torr. The B doping doses and zinc diffusion temperatures were chosen so that the concentration of Zn in the samples amounted to 10% of that of the majority impurity [17].

The capacitance measurements were performed with a Boonton 72B capacitance meter on samples cooled to 92 K in a cold finger cryostat, with cooling carries out under reverse bias conditions. Variations in photocapacitance were photogenerated by allowing the light from an incandescent lamp outfitted with a double monochromator to access the sample through a quartz window in the cryostat.

3. Results

The TSCAP studies indicate [16, 17] that zinc gives rise to two interrelated deep acceptor levels in the energy gap of p-type silicon. It should, then, be possible to induce emission of holes by illuminating the samples with impurity light, thus providing a basis for photocapacitance studies. The results of an experiment carried out along these lines are shown in figures 1 and 2.

Following a zero bias pulse which set the initial hole occupancy, reverse bias was applied that caused most levels in the depletion region to be filled with holes. If the temperature conditions of the experiment are appropriate for thermal emission of holes from either or both impurity levels, then one should expect a gradual rise in the capacitance signal. This is just what was observed at $V_R = 20$ V, figure 1(b). No such rise, however, was observed when $V_R = 1$ V was applied, figure 1(a). Instead, after the 'short-circuiting' step, the photocapacitance continued to decline dramatically at a rate that could be made still faster by optical

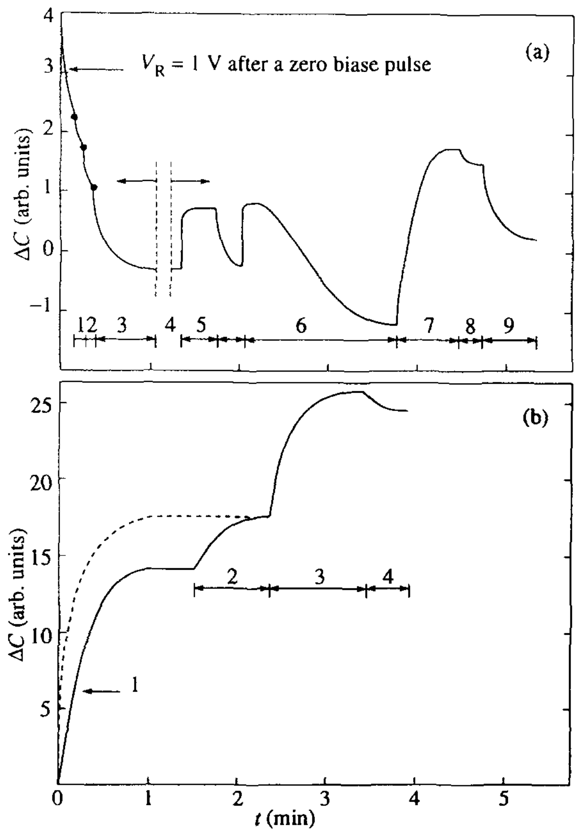

Figure 1: Decay of the photocapacitance signal after the 'short-circuiting' step where a zero bias pulse is followed by the application of reverse bias. (a) $V_R = 1$ V, (b) $V_R = 20$ V (a) 1-3→ after a zero bias pulse, 1—$h\nu = 0.3$ eV, 2—0.35 eV, 3—0.4 eV, 5—0.5 eV, 6—0.55 eV, 7—0.62 eV, 8—0.9 eV, 9—1.05 eV. (b) 1—after a zero bias pulse, 2—$h\nu = 0.55$ eV, 3—0.65 eV, 4—1.05 eV; dashed line—after a zero bias pulse $h\nu = 0.55$ eV.

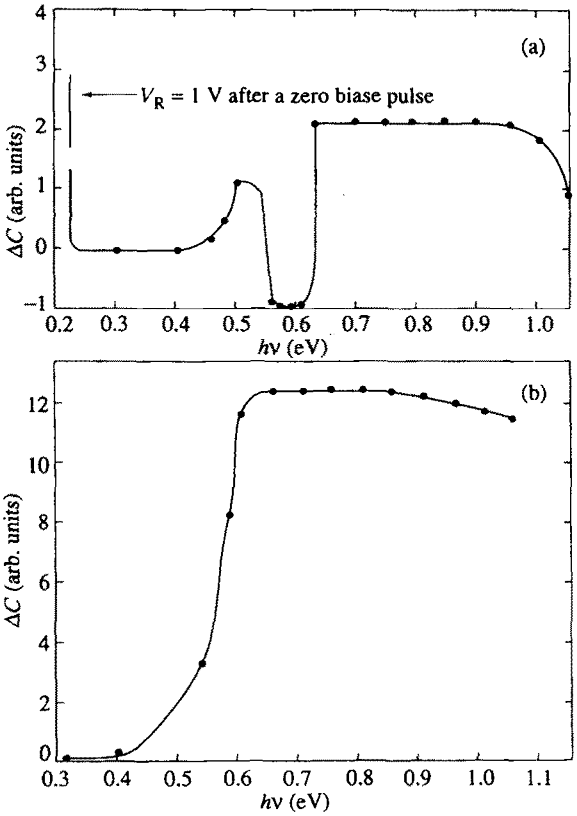

Figure 2: Spectral dependence of photocapacitance for p-Si:Zn (a) $V_R = 1$ V, (b) $V_R = 20$ V.

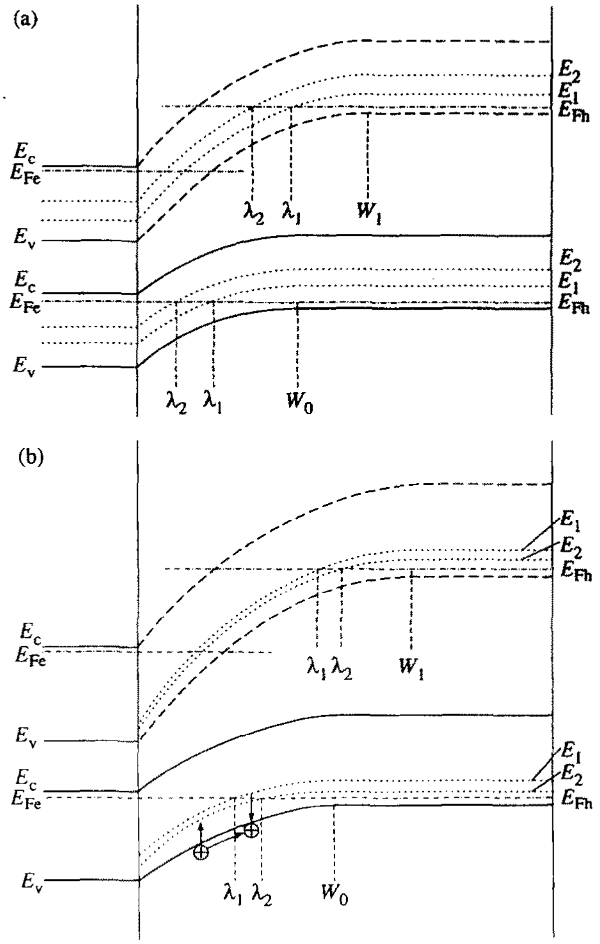

Figure 3: Band diagram of an n^+p junction in Zn- and B-doped p-type silicon for $V_R = 0$ (dashed line) and $V_R = 1$ V (solid line). Points mark the zinc levels. E_{Fh} and E_{Fe} are the Fermi quasi-levels for holes and electrons, respectively, at $V_R = 1$ V. (a) data from [14], (b) the negative-U ordering in the acceptor levels. E_1 and E_2 are the energies for $(0/-)$ and $(-/--)$ thermal transitions, respectively.

pumping ($0.3 \leq h\nu < 0.5$ eV), figure 1(a). All defects with normal level ordering that populate the lower half of the energy gap would normally be expected to induce photo- and thermoemission of holes (with the corresponding enhancement in the photocapacitance signal). Therefore, the behaviour exhibited by zinc at weak electric fields, which is anomalous in that zinc tends to stabilize the emitted holes within the depletion region, should be interpreted as the result of a negative-U reaction (of thermal or optical origin): $2Zn^- \rightarrow Zn^{--} + Zn^0$ (92 K, $0.3 \leq h\nu < 0.5$ eV), and in any case, could serve as a signature of a defect with negative-U level ordering.

The enhancement of the photocapacitance signal at $h\nu \geq 0.5$ eV, figures 1(a) and 2(a), may will be taken as evidence of the hole photoemission induced by $(-/--)$ optical transitions from residual Zn^- states located close to the bound of the depletion region, figure 3.

An exciting effect has been observed at $h\nu > 0.55$ eV, figures 1(a) and 2(a). At this energy the photocapacitance signal first showed an increase with time of optical pumping, and then a decrease with the final magnitude being less than its initial value. This observation constitutes convincing evidence for metastability as a fundamental property of the Zn-related centre in silicon, which could be a consequence of the photodissociation of the residual Zn^- states as a result of $(-/--)$ photoionization energy changes depending on the distance between the Zn-related centre and the bound of the depletion region, figure 3: at $h\nu = 0.5-0.55$ eV $\rightarrow Zn^- + h\nu \rightarrow Zn^{--} + h$; at $h\nu = 0.55-0.62$ eV $\rightarrow 2Zn^- + h\nu \rightarrow Zn^{--} + Zn^0$: $Zn^- + h\nu \rightarrow Zn^{--} + h$, $Zn^- + h \rightarrow Zn^0$.

Further increase of the pumping light energy at $V_R = 1$ V, figure 1(a), led to an enhancement in the photocapacitance, due to an increased rate for the $(0/-)$ transitions, and it was only at much higher illumination energies, which enabled the electrons being emitted from charged zinc centres to pass into the conduction band, that a somewhat slower rise of the photocapacitance was observed, figure 1(a).

It should be noted that the reduced photocapacitance observed at weak electric fields with a zero bias pulse off (figure 1(a)) could conceivably be due to the emission of minority carriers trapped at zinc centres lying near the outer boundary of the n^+p junction, figure 3. Indeed, evidence from experiments on Si:Au and GaAs containing A and B centres [21] indicates that such minority carriers of this kind apparently contribute to the drop in the photocapacitance signal which occurs under reverse bias when these carriers would normally be undergoing emission, of a thermal or an optical variety. From figure 3 it is evident, on the other hand, that the magnitude of the above enhancing effect decreases dramatically with increasing reverse bias. Considering that the energies necessary for holes to be emitted from zinc centres via the thermal channel are 0.316 eV and 0.617 eV [17], it is reasonable to suppose that only few minority carriers could be trapped at zinc centres, and that their contribution to the observed reduction in the photocapacitance signal (figure 1(a)) should therefore be marginal at best. Following the negative-U approach the Zn-related centre in p-type silicon, described in the Introduction, the minority carriers trapped at near-surface centres would not be expected, as figure 3(b) suggests, to undergo any appreciable emission under the present experimental conditions (92 K) and, thus, could not possibly have any observable effect on the post-'short-circuiting' drop in the photocapacitance signal, figure 1(a).

It follows that the likeliest candidate for a mechanism that would account for the precipitous photocapacitance drop in the negative-U reaction mentioned above, viz. $2Zn^- \rightarrow Zn^{--} + Zn^0$. This reaction comes into effect with lowering the sample temperature from 293 K to 92 K, and it gives rise to Zn-related centres in the Zn^--state when operating beyond the depletion region, whereas centres generated within the depletion region are all in the Zn^{--}-state. (The value of the ΔC quenching after the 'short circuiting' step is proportional to the difference between λ_1 and λ_2), figure 3(b). As a result of equalisation in total charge between the inside and the outside of the depletion region that sets in upon application of a zero bias pulse, the Zn^{--}-states within the depletion region convert to Zn^--states through hole capture $Zn^{--} + h \rightarrow Zn^-$. When reverse bias is then applied, the negative-U reaction gains a substantial impetus, making the depletion region narrower and causing the concomitant drop in the photocapacitance signal observed in the experiment, figure 1(a) and figure 3. After the 'short-circuiting' step the residual Zn^--states remain only near the bound of the depletion region and dissociate at $h\nu = 0.55-0.62$ eV ($2Zn^- + h\nu \rightarrow Zn^{--} + Zn^0$), figures 1(a) and 3.

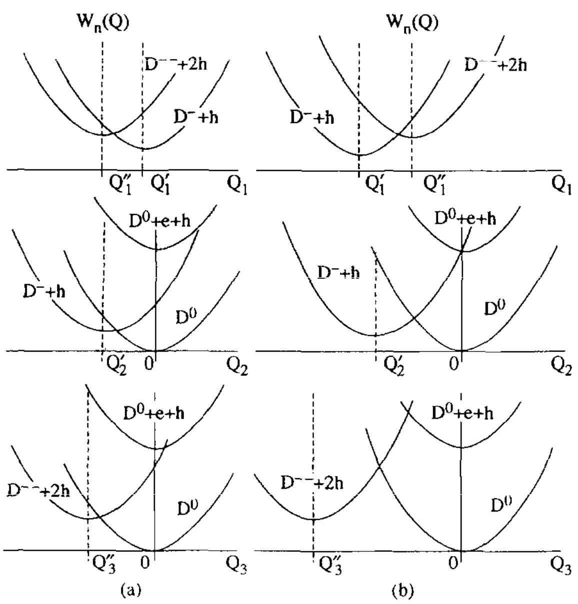

Figure 4: Two-electron adiabatic potentials and the equivalent one-electron band diagram for the negative-U Zn-related centre in p-type silicon. (a) $\mathbf{E} = 0$; (b) $\mathbf{E} \parallel [111]$. Configuration coordinates Q_1, Q_2 and Q_3 correspond to the directions in the silicon lattice (see figure 5).

At stronger electric fields ($V_R = 20$ V), the behaviour of zinc charge states responsible for variations in the photocapacitance signal were found to be consistent with the conventional positive-U picture, figure 1(b) ($Zn^{--} + h \rightarrow Zn^- \Longleftrightarrow Zn^- \rightarrow Zn^{--} + h$).

The spectral dependences obtained for quenching and regeneration of the photocapacitance, figures 2(a) and (b), also suggest an important role for the electric field in inhibiting the negative-U reaction, and thus, the effects of metastability and self-compensation observed for the Zn-related centre in p-type silicon. This spectral information forms a basis for the model of the negative-U defect to be described in the next section.

4. The model

The basic principles of the model for zinc double acceptor centre, an off-centre with three charge states as a function of the configurational coordinate (figure 4), can best be explained in terms of two-electron adiabatic potentials [14, 15]. The model suggests that the reconstructed double acceptor, $(Zn_iV_{Si})^{--} = D^{--}$, is located in the C_{3V}-tetrahedral interstitial position, while the $(Zn_iV_{Si})^- = D^-$ and $(Zn_iV_{Si})^0 = D^0$ states are also located in interstices and satisfy the symmetry conditions imposed, respectively by C_{2V} and D_{2d} (figures 4 and 5). Since the electron-vibration interaction (EVI) constant is a nonmonotonic function of the number of electrons at the centre, any change in the centre's charge state will cause the reconstructed defect to tunnel from one of its equilibrium positions to the other. This tunnelling movement by the centre between the C_{3V}, C_{2V} and D_{2d}-symmetry positions is to compensate for the Coulombic repulsion of electrons in a sufficiently rigid lattice [14]. Because of its negative-U properties, the orthorhombic Zn centre in the D^--state is unstable under the C_{2V}-symmetry conditions, and should therefore transform through a spontaneous dissociation reaction to either D^{--} or D^0-state, thus changing its symmetry position to C_{3V} or D_{2d}, respectively. The centre may, however, subsequently revert to its initial state, having absorbed or emitted a charge particle owing to impurity light illumination, figures 1 and 2.

The equivalent band diagram shown in figure 4 incorporates two levels, one for the background ($E_g - I_1$) and one

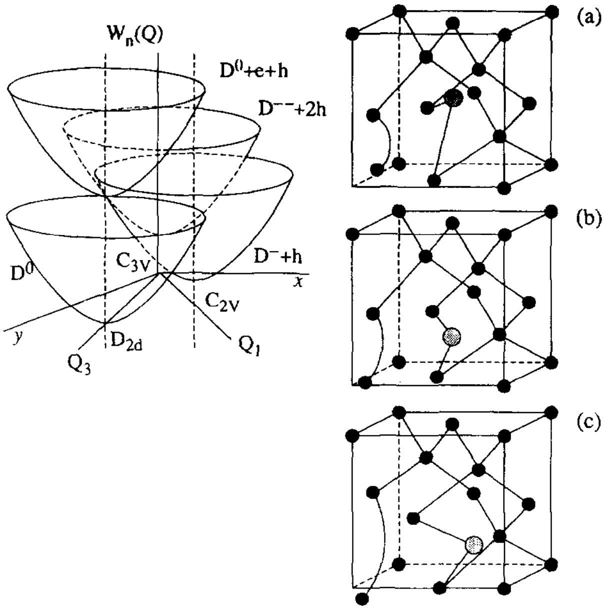

Figure 5: Model of a reconstructed double acceptor centre in silicon (a) D^{--}-state; (b) D^{-}-state; (c) D^{0}-state. Three-dimensional two-electron adiabatic potentials for different charge states of the Zn-related centre in p-type silicon (C_{3V}, C_{2V} and D_{2d} sequence).

for the correlated ($E_g - \Delta I$) electron, so as to allow for the centre's charge states; here $I_2 = I_1 + \Delta I$.

By virtue of its rationale, which is based on the principles set forth in the Introduction with regard to the interpretation of the TSCAP data for I_1, I_2 and ΔI in p-Si:Zn [16, 17] in terms of the negative-U concept, figure 4 provides evidence in favour of negative-U ordering not only for optical transitions of the form $(0/-) - h\nu = 0.62$ eV, $(-/--) - h\nu = 0.5$ eV, but for the thermally-induced change of the charge state of the Zn-related centre as well. It thus explains the possibility of the above-described thermal self-compensation to the Zn^{-}-states after the 'short-circuiting' ($D^{--} + h \to D^{-}$) step:

$$\left.\begin{array}{l} D^{-} \to D^{--} + h \;\; (92\,K) \\ D^{-} + h \to D^{0} \end{array}\right\} 2D^{-} \to D^{--} + D^{0} \qquad (1)$$

A consequence of reaction (1), which is a negative-U reaction, is that the depletion region is narrowed (figure 3) and there is the corresponding reduction in the capacitance at weak electric fields (figure 1(a)) which can be accelerated by pumping with $0.3\,eV \leq h\nu < 0.5$ eV light (figure 1(a)), causing holes to be emitted from the Zn^{-}-state

$$D^{-} + h + h\nu \to D^{0} \qquad (2)$$

In general, as a consideration of reaction (1) suggests, there are two ways in which a hole can be trapped as a result of the negative-U reaction. The capture of an emitted hole by the Zn^{-}-centre can occur at the edge of the depletion zone, or else it may result from the 'tail effects' and then the captured hole is one that originates from the rest of the depletion region.

The narrowing of the depletion region can be eliminated, with the photocapacitance signal at weak fields restored to its previous value, if, as was pointed out earlier, the energy of the pumping light is 0.5 eV, in which case $(-/--)$ transitions occur, figure 4

$$D^{-} + h\nu \to D^{--} + h \qquad (3)$$

The use of the pumping light with energies exceeding 0.55 eV, however, gives rise to photodissociation of the residual Zn^{-}-centres

$$2D^{-} + h\nu \to D^{--} + D^{0} \Longleftrightarrow \qquad (4)$$

$$\Longleftrightarrow D^{-} + h\nu\;(0.55\text{–}0.62\,eV) \to D^{--} + h \qquad (5)$$

$$D^{-} + h \to D^{0} \qquad (6)$$

as a result of $(-/--)$ photoionization energy dependence on the distance between the Zn^{-}-centre and the bound of the depletion region, figure 3: at $h\nu = 0.5\text{–}0.55$ eV the photoholes are only emitted from residual Zn^{-}-centres located close to the bound of the depletion region, whereas at $h\nu = 0.55\text{–}0.62$ eV all residual Zn^{-}-centres begin to create photoholes within the depletion region thereby stimulating the Auger process of the type (4)–(6). Thus, the competition between reactions (5) and (6) would account for the observation of double quenching experienced by the photocapacitance signal at $0.55 < h\nu < 0.62$ eV, figures 2(a) and 4.

These results are in good agreement with the photo-EPR data that identify the negative-U properties for the Si-NL36 orthorhombic Zn-related centre (C_{2V} -symmetry) [22–25]. The coincidence of the quenching and regeneration spectra of the Zn^{-} -state, which is obtained using both techniques, shows that the Zn-related centre observed by the photocapacitance method represents the direct analog of the Si-NL36 Zn-related centre. It should also be noted that several trigonal (ZnFe, ZnCr), monoclinic I (ZnCu) and orthorhombic Zn-related centres have been found using EPR technique [24]. The identification of their models was made by special isotopic doping with ^{67}Zn, ^{57}Fe, etc. magnetic isotopes [22–25]. It is important that only the Si-NL36 centre (Zn^{-}-state) was created by impurity light illumination and revealed negative-U properties, while the pairs of zinc with other transition metals showed themselves in the EPR measurements without optical pumping [22–25].

Figure 4 suggests that there is a definite relationship between the rigidity of deep centres and, therefore, the probability of the above described processes of photocapacitance quenching, regeneration, and sign reversal on the one hand, and the magnitudes of the ratio of the Q_1, Q_2 and Q_3 values, on the other. The Stark effect contributes significantly to making these values highly sensitive to the electric field applied. Indeed, as can be seen in figure 4, the negative-U properties of the Zn-related centre are suppressed under the action of the linear and quadratic Stark effects [14, 15] which arise from the electric field applied and induce the shifts of the adiabatic potentials of the charged states (D^- and D^{--}) from the fixed C_{3V} and C_{3V} interstitial positions. The magnitudes of these shifts responsible for the metastable behaviour of the Zn-related centre depend on its charge state and the angle at which the electric field $\mathbf{E}$ is inclined to the [110] and [111] crystallographic directions: $\delta Q_1'' = (2eE/\kappa)\cos\Theta_1$; $\Theta_1 = [\mathbf{E} \parallel [111]]$; $\delta Q_2' = (eE/\kappa)\cos\Theta_2$; $\Theta_2 = [\mathbf{E} \parallel [110]]$, where κ is the force constant for the local vibration mode of the defect. Thus, despite the negative-U ordering of the lower and the upper levels, the metastability induced by the Stark effect prevents spontaneous dissociation of the D^--state, figures 1(b), 2(b) and 4. A subsequent optical pumping promotes the enhances in the photocapacitance signal because of the effective photoemission of holes: $D^0 + h\nu \to D^{--} + h$; $D^- + h\nu \to D^{--} + h$.

By fitting the experimental values obtained in figures 1 and 2 to the fixed separations between the 'ideal' C_{3V}, C_{2V} and D_{2d} lattice positions, it is possible to determine the direction for the short-range anisotropic Stark interaction and hence to confirm the C_{2V}-symmetry of the Zn^- unstable state.

It is to be noted that zinc centres residing in the depletion region are subject to an electric field differential, present there with respect to the outer boundary of the n^+p junction. For this reason, even at $V_R = 1$ V, the negative-U reaction can only extend over the areas of smooth band bending (figure 3), leaving the centres that lie near the outer boundary of the n^+p junction metastable and hence unaffected from the stand-point of spontaneous dissociation ($2D^- \to D^{--} + D^0$). For higher values of the reverse bias and the consequent greater acuteness in band bending, the number of metastable Zn^--centres would be expected to grow at the expense of the negative-U centres; this seems indeed to be the case, as the increased scope for photocapacitance sign reversal shown in figure 1(a) and (b), suggests.

Further TSCAP studies to be carried in weak electric fields are needed to establish the exact values for the energy required to maintain negative-U ordering in the case of thermal $(-/--)$ and $(0/-)$ transitions.

5. Summary

Photocapacitance experiments, conducted at various electric fields, have been employed to determine whether zinc double acceptors in p-type silicon are negative-U centres. The self-compensatory and metastable nature of the Zn-related centre has been identified by analysing of two effects observed in the present work: field-dependent quenching and sign reversal of photocapacitance. The negative-U ordering in the $(0/-)$ and $(-/--)$ levels has been found for both optically- and thermally-induced emission. It has been shown that the linear and quadratic Stark effects are involved in the field-dependence of negative-U ordering for deep levels of the Zn-related centre which tunnels between its off-centre positions as a result of the addition of one or two electrons. The experimental results and those from model studies are combined to demonstrate that the Zn-related centre in p-type silicon has negative-U properties.

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