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Observation of a long-range electric drift of negatively charged vacancies in the space-charge regions of Au/n-Si Schottky diodes

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Abstract. In-depth distributions of P_sV and C_iP_s complexes formed in the space-charge regions of Au/n-Si Schottky diodes irradiated with fast electrons in a special electrical regime have been investigated. In the course of the irradiation the diodes were periodically driven into the reverse-bias state with square-shaped voltage pulses of fixed amplitude alternating with the forward-current pulses at a 2 MHz frequency. Non-flat profiles of P_sV concentration have been revealed by deep-level transient spectroscopy measurements, while the interstitial defects were found to have a constant density. The result is regarded as strong evidence for the long-range (approximately 0.1–1 μm) field-assisted drift of negatively charged vacancies (V^-) across the depleted region. The diffusion length of V^- evaluated from the drift model (about $(3.8\pm0.4)\times10^{-2}$ μm for the float-zone silicon used with a phosphorus content of 5×10^{15} cm^{-3}) was found to be consistent with the value expected from the theory of diffusion-limited reactions.

1. Introduction

The electrical polarization of diode structures combined in some cases with thermal treatments has been successfully used in order to observe a drift motion of several impurity and defect centres in silicon and some other semiconductors [1–19]. However, no unambiguous observations of a long-range electric drift of primary radiation defects (RDs) (in particular, vacancies) in silicon have been yet reported in the literature; although models assuming electromigration have been widely used for the description of non-uniform distributions of secondary defects formed in the space-charge regions (SCRs) or near external interfaces [20–22], the proposed interpretations were obviously not alternative-free nor did they include a comprehensive analysis of other possible reasons which could cause the origin of inhomogeneities.

For example, an increased concentration of a vacancy-related deep level at the boundary between an SCR and p-type quasi-neutral bulk (QB), which was attributed in [20] to the field-assisted drift of vacancies, could be merely an apparent feature caused by uncertainties in the profiling in the presence of an abrupt gradient of deep-level concentration (analogous to that described in [23]). A detailed review of our early studies made on n-type Si [22, 24] was given in [25]. Although both the spatial distributions of RDs formed in the SCRs and the time characteristics of their formation process have been thoroughly investigated, we could not conclude unambiguously whether it is a drift motion of vacancies or the charge dependence of the Frenkel pair separation probability that causes the lower introduction

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rates of RDs in the SCR compared with the QB. The latter, however, seems to be more reasonable, since too high vacancy diffusion coefficients (about 10^{-5} cm^2 s^{-1}) were needed to describe the experimental results from the viewpoint of the drift model. In addition, it has been realized that peaks of RD concentration appearing at the SCR boundary with the QB could be the result of mere diffusion motion of primary defects whose diffusion length in a depleted region exceeded that in QB due to the difference in their charge states in the two regions.

The procedure which has been commonly used heretofore for making an attempt to observe a vacancy drift in SCRs consisted in a study of RD accumulation in Si diode structures which were reverse-biased with a constant voltage when irradiated [20, 22]. However, the main limiting factor for realization of long-range electromigration of charged vacancies in such conditions is too short a period (compared with the free-vacancy lifetime) during which non-zero non-equilibrium charge states of a vacancy in the SCR do exist (even if these do really appear just after the separation of the Frenkel pairs, which in itself was the point of numerous discussions (see, for example, [26–28])). Indeed, the charged vacancies in the SCR lose their charge over times $\tau \simeq 10^{-11}$ s (V^+), $\tau_e \simeq 10^{-8}$ s (V^{2+}) [29] and $\tau_e \simeq 10^{-6}$–10^{-5} s (V^-) (see below), thus limiting their drift length l_E in fields of about 10^{-5} V cm^{-1} to be less than 2×10^{-2} μm† (if a special mechanism of combined vacancy motion with the carriers is not to be supposed [25, 28]). In the present work we have proposed and employed much more favourable experimental conditions for observation of the vacancy drift in the SCRs of Schottky barriers. These conditions are as follows. In the course of irradiation the square-shaped pulses of the reverse-bias voltage are applied to the diode, each followed by an interval during which the forward current is passed through the structure. The pulse-period-to-pulse-duration ratio equals 2 while the repetition frequency is chosen to satisfy the condition

$$\tau_c < f^{-1}/2 < \tau_e \tag{1}$$

where τ_c and τ_e are the electron capture and emission times, respectively. The condition (1) provides an occupancy of vacancies of close to 100% with electrons in the near-contact region for the half-period during which the forward current flows, and a weak V^- discharge over the time of field being applied. The condition $\tau_c < \tau_e$ corresponds to a negative steady charge state of vacancies in the QB of the sample and is fulfilled when the Fermi level lies above $E_{V^-} \simeq E_c - (0.27 \div 0.30)$ eV [25, 28, 32]. If such conditions are realized, half the vacancy lifetime τ (when the forward bias is applied) is spent in maintaining the negative (characteristic to QB) charge state of vacancies in the SCR while the other half (when the diode is reverse-biased) is used to give V^- drift. When oxygen-free silicon with $N_{P_s} \simeq 5 \times 10^{15}$ cm^{-3} is used in the experiment, one should expect vacancies to drift over about 0.5 μm in a field of about 10^5 V cm^{-1}. This statement can easily be demonstrated by using the relationship $l_E = \mu_V E\tau/2 = eEl_V^2/2kT$ and substituting $(4\pi N_{P_s} R_C)^{-1/2}$ for the vacancy diffusion length ($R_C = e^2/4\pi\varepsilon_a kT$ is the Coulomb capture radius). The above value arises on the assumption that the vacancy lifetime is limited by the diffusion-controlled capture of vacancies at P_s^+ [33]. The formation of non-flat distributions of vacancy-related complexes with the characteristic length of inhomogeneity of about 0.5 μm can easily be revealed by deep-level transient spectroscopy profiling. This was the purpose of the present work.

† Estimation according to $l_E = \mu_V E\tau_e$, $\mu_V = eD_V/kT$ with $D_V = (0.9 \div 2.2) \times 10^{-4} \exp(-E_a/kT)$ cm^2 s^{-1}, the activation energy E_a being taken to be equal to 0.25 eV and 0.48 eV for V^- and $V^{+(2+)}$, respectively [30–32].

2. Experimental details

In our experiment we used Au/n-Si Schottky barriers obtained by thermovacuum evaporation of gold onto the etched wafers of float-zone silicon with $N_{P_s} \simeq 5.5 \times 10^{15}$ cm^{-3} and $N_{O_i} \lesssim 5 \times 10^{15}$ cm^{-3}†. The irradiations have been performed using a pulsed cyclic accelerator of microtron type [34] with an average current of fast electrons of 0.2 μA cm^{-2} ($E_e = 3.5$ MeV; $t_{pulse} = 3$ μs; $f' = 100$ Hz). The electrical regime of the diodes under irradiation was as described above. The reverse-bias voltage pulses followed at a frequency of 2 MHz. No synchronization between the pulsed biasing and irradiation was used. Several diodes have been irradiated with different amplitudes of the reverse-bias viltage ranging from 0 to 35 V. The density of forward current during the pulse separations amounted to 50–100 μA cm^{-2}. The choice of the frequency satisfied condition (1). To see this, let us take into account the estimation of cross section for the electron capture at a neutral vacancy, $\sigma_n \simeq 5 \times 10^{-17}$–$10^{-16}$, which was made in [25, 28]. Then for τ_c and τ_e we obtain $\tau_c \simeq (\sigma n v_t)^{-1} \simeq 0.18 \div 0.36$ μs (here $n = N_{P_s}$ is the concentration of free electrons in QB, and v_t their thermal velocity), $\tau_e \simeq (\sigma_n b_n T^2)^{-1} \exp((E_c - E_{V^-})/kT) \simeq 0.8 \div 5.5$ μs ($b_n \simeq 6.6 \times 10^{21}$ cm^{-2} s^{-1} K^{-2} [36]). The spatial distributions of P_sV complexes were measured immediately after the irradiation while those of C_iP_s complexes were taken a day after, a period sufficient for full annealing of interstitial carbon C_i. A typical deep-level transient spectrum of the irradiated silicon and the details of measurements have been published elsewhere [37].

3. Results and discussion

Shown in figure 1 are the measured distributions of P_sV and C_iP_s complexes. It is seen that flat profiles of P_sV concentration are formed at the investigated depths ($x \geqslant 0.2$–0.3 μm) for the forward-bias irradiated diodes, while non-uniform profiles appear for the diodes irradiated under pulsed bias. In the latter case the profiles $N_{P_sV}(x)$ manifest a lower P_SV concentration (compared with the bulk density) in the high-field region and a higher concentration in the space-charge layer immediately adjacent to the bulk. In both cases, C_iP_s complexes are distributed uniformly at the probed depths, their introduction rates in the QB and in the SCR being the same. It should be noted that the observed distributions of the vacancy-related defects can be understood as resulting from the electric-field drift transport of negatively charged vacancies across the depletion region towards the bulk and their accumulation near the inner edge of the depleted region owing to the lower drift velocity at $x \simeq h$.

Since in the general case the drift of charged vacancies is one possible reason which can influence the accumulation of RDs in the presence of the electric field, let us consider arguments proving a negligible contribution of other possible mechanisms of inhomogeneity appearance in the case under study.

These alternative mechanisms are the following:

(1) field and charge dependences of Frenkel pair separation probability [25, 28, 38];
(2) charge dependence of the P_sV formation rate constant [28].

The observed flatness of C_iP_s concentration profiles (see figure 1) provides evidence against a noticeable contribution of mechanism (1). Indeed, since the diffusion length of C_i in the samples under study does not exceed about 0.2 μm [28], the uniformity of the

† Evaluation according to [35].

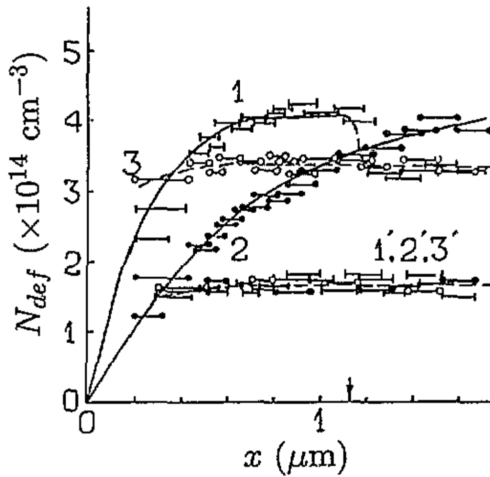

Figure 1. In-depth distributions of P_sV (curves 1–3) and C_iP_s (curves 1′–3′) complexes in Au/n-Si Schottky diodes irradiated in the pulsed electric regime (see text) (curves 1, 2, 1′ and 2′) and under a constant forward current (curves 3 and 3′). The broken lines are drawn through experimental 'points' for better visualization of the profiles; the solid curves represent the result of calculations according to equation (3). For curves 1 and 1′, $U = -4$ V and, for curves 2 and 2′, $U = -35$ V. The dose of irradiation is 1×10^{15} cm^{-2}.

profiles is a good indication that no essential variation in generation rate of free vacancies over the depth had taken place. In the case when mechanism (1) is operative, the profiles N_{P_sV} and $N_{C_iP_s}$ should have similar forms.

*Taking into account the similarity of vacancy charge states in the bulk and in the near-*contact region as well as our estimates of the vacancy charge effect on the introduction rate of P_sV complexes in the material under study [28], we can conclude that mechanism (2) *cannot contribute noticeably too. This conclusion is additionally supported by the spatial* uniformity of vacancy charge states in the intermittently depleted region, by virtue of which mechanism (2) cannot account for irregularities of P_sV concentration profiles within the SCR (sufficiently far from its boundaries).

Let us compare now the measured P_sV concentration profiles with those expected from V^- redistribution under the action of the electric field. The latter can easily be derived from the kinetic equation for vacancies in the quasi-stationary approximation which can be obtained by averaging the exact time equation over the period exceeding the vacancy lifetime [22]:

$$0 = (\alpha^- D_{V^-} + \alpha^0 D_{V^0})\frac{\partial^2 \bar{N}_V}{\partial x^2} - \frac{\beta}{2}\mu_{V^-}\frac{\partial}{\partial x}(E(x)\bar{N}_V) + g - \bar{N}_V\left(\frac{\alpha^-}{\tau^-} + \frac{\alpha^0}{\tau^0}\right). \quad (2)$$

From the kinetic equation for P_sV complexes

$$\frac{\partial N_{P_sV}}{\partial t} = \bar{N}_V\left(\frac{\alpha^-}{\tau_P^-} + \frac{\alpha^0}{\tau_P^0}\right),$$

where $E(x) = (2(U_b+U)/h)\times(1-x/h)$ is the linearly decreasing electric field ($U_b = 0.8$ V is the built-in bias; $h = (2(U_b + U)/e^2\varepsilon_a N_{P_s})^{1/2}$ is the SCR width), D_{V^-} and D_{V^0} are the diffusion coefficients of V^- and V^0, respectively, $\alpha^-\bar{N}_{V^-}$ and $\alpha^0\bar{N}_{V^0}$ are the time-averaged concentrations of negatively charged and neutral vacancies, respectively ($\bar{N}_V$ being their total concentration), $\beta\bar{N}_V$ is the V^- concentration averaged over the duration of the field application, g is the mean generation of free vacancies, τ^- and τ^0 are the free-vacancy

lifetimes in V^- and V^0 charge states, respectively, and τ_P^- and τ_P^0 are the lifetimes with respect to the capture at P_s^+.

When writing equation (2) we took into account that only the V^- and V^0 charge states of vacancies are significant in the steady-state regime (when the charge states of vacancies in the near-contact region vary periodically with time), i.e. $\alpha^- + \alpha^0 \simeq 1$. Neglect of the diffusion term in equation (2) (the so-called drift approximation applicable within the high-field region of the space-charge layer) leads to the following solution which satisfies the boundary condition $\bar{N}_V(0, t) = 0$:

$$\bar{N}_V(x) = g\tau_{eff} \frac{h}{h - l_E} \left[1 - \left(\frac{h-x}{h}\right)^{(h-l_E)/l_E}\right],$$

where $\tau_{eff} = (\alpha^-/\tau^- + \alpha^0/\tau^0)^{-1}$, $l_E = [e(U+U_b)/kT](l_{eff}^2/h)$ and $l_{eff} = (\beta D_V - \tau_{eff})^{1/2}$.

It follows from this that the P_sV complexes are distributed according to

$$N_{P_sV} = \begin{cases} kN_{P_sV}(\infty) \dfrac{h}{h - l_E} \left[1 - \left(\dfrac{h-x}{h}\right)^{(h-l_E)/l_E}\right] & x < h \\ N_{P_sV}(\infty) & x > h \end{cases} \tag{3}$$

where $k = (g^{SCR}/g^{QB})$ $(\tau_{eff}^{QB}/\tau_{eff}^{SCR})$ $(\tau_{P\,eff}^{SCR}/\tau_{eff}^{QB})$ (the indices QB and SCR denote the values related to the bulk and to the layer $0 < x < h$, respectively, $N_{P_s}(\infty)$ is the bulk concentration, and $\tau_{P\,eff} = (\alpha^-/\tau_P^- + \alpha^0/\tau_P^0)^{-1}$).

In order to estimate the values of α^-, α^0, β and their ratios α^0/α^- and α^-/β, which will be needed later, let us consider the equation describing the kinetics of charge states of a vacancy in the region $0 < x < h$:

$$\frac{\partial N}{\partial t} = \begin{cases} \dfrac{N}{\tau_c} - N^- \left(\dfrac{1}{\tau_c} + \dfrac{1}{\tau_e}\right) & 0 < t < \frac{T}{2} \\ -\dfrac{N^-}{\tau_e} & \frac{T}{2} < t < T. \end{cases} \tag{4}$$

Here N^- is the concentration of negatively charged vacancies, and $T = f^{-1}$ is the period of the frequency; the leading edge of a forward-current pulse corresponds to the moment $t = 0$. As a solution of equation (4) that we are seeking (which is periodic in time), we obtain

$$\frac{N^-}{N} = \begin{cases} \dfrac{1}{1+\tau_c/\tau_e} \left\{1 - \dfrac{1-\exp(-T/2\tau_e)}{1-\exp(-T/\tau_e - T/2\tau_c)} \exp\left[-\left(\dfrac{1}{\tau_c} + \dfrac{1}{\tau_e}\right)t\right]\right\} & 0 < t < \frac{T}{2} \\ \dfrac{1}{1+\tau_c/\tau_e} \left(1 - \dfrac{1-\exp(-T/2\tau_e)}{1-\exp(-T/2\tau_c - T/\tau_e)}\right) \exp\left(\dfrac{T-t}{\tau_e}\right) & \frac{T}{2} < t < T. \end{cases}$$

Performing the appropriate averagings, we get

$$\beta = \frac{1}{1+\tau_c/\tau_e} \frac{2\tau_e}{T} \left[\exp\left(\frac{T}{2\tau_e}\right) - 1\right] \left(1 - \frac{1-\exp(-T/2\tau_e)}{1-\exp(-T/2\tau_c - T/\tau_e)}\right)$$

$$\alpha^- = \frac{\beta}{2} + \frac{1}{2(1+\tau_c/\tau_e)} \left(1 - \frac{1-\exp(-T/2\tau_e)}{1-\exp(-T/2\tau_c - T/\tau_e)}\right) \frac{2\tau_c}{T} \frac{1}{1+\tau_c/\tau_e} \times \left[1 - \exp\left(-\frac{T}{2\tau_c} - \frac{T}{2\tau_e}\right)\right].$$

With the above-mentioned uncertainties in τ_c and τ_e, we obtain the following ranges of possible values for α^-, α^0, β, α^-/β and α^0/α^-:

$$0.67 \leqslant \alpha^- < 1 \qquad 1 < \alpha^-/\beta \leqslant 1.03$$

$$0 < \alpha^0 \leqslant 0.33 \qquad 0 < \alpha^0/\alpha^- \leqslant 0.50$$
$$0.66 \leqslant \beta < 1.$$

The profiles $N_{P_s}(x)$ calculated according to equation (3) and exhibiting the best agreement with the experimental data at $k = 1$ and $l_{eff} = 0.034$ μm are shown in figure 1 by the solid lines. A discontinuity in the P_sV distributions at $x = h$ stemming from the drift approximation used should be smoothed in the experimental profiles both because of diffusion and because the P_sV concentration is measured within a layer of a certain finite width. This is shown by a broken part of the solid line in figure 1. The value $k = 1$ is chosen taking into account that $g^{SCR} \simeq g^{QB}$, $\tau_P^{SCR} \simeq \tau_P^{QB}$ and $\tau_{eff}^{SCR} \simeq \tau_{eff}^{QB}$ (because of the prevailing fraction of negatively charged vacancies and because τ_{eff} is determined mainly by the capture of vacancies at phosphorus donors). Hence, taking into consideration the inequality $\tau^- < \tau^0$, we have the following estimation for the diffusion length of V^- $l_{V^-} = (D_{V^-}\tau^-)^{1/2}$: $(\alpha^-/\beta)^{1/2} l_{eff} < l_{V^-} < ((1 + \alpha^0/\beta)\alpha/\beta)^{1/2} l_{eff}$. With the ranges obtained for α^-/β and α^0/α^-, we obtain $l_{V^-} \simeq (3.8 \pm 0.4) \times 10^{-2}$ μm. It is noteworthy that the closeness of this result to the value $(D_{V^-}\tau_P^-)^{1/2} = (4\pi N_{P_s} R_C)^{-1/2} \simeq 5.4 \times 10^{-2}$ μm is consistent with preferential vacancy trapping at phosphorus. The somewhat lower experimental value observed indicates V^- trapping at other background species (such as interstitial oxygen).

4. Summary

The formation of non-uniform depth distributions of P_sV complexes in Au/n-Si Schottky diodes subjected to intermittently reversed biases under irradiation has been observed. We attribute this observation to the field-assisted drift of negatively charged vacancies in the SCRs. The drift interpretation is supported by the simultaneous observation of flat distributions of interstitial defects (C_iP_s). Good agreement between the observed P_sV concentration profiles and the calculated profiles has been achieved. The V^- diffusion length derived from the best fit is found to be consistent with the value determined by the V^- Coulomb capture at P_s^+ donors.

Finally, it should be noted that the experimental procedure proposed in the present work can be applied to a number of other sufficiently mobile deep centres which although charged in the QB possess no charge in the depleted regions, provided that their electronic levels are sufficiently deep. Note here that, since the fulfilment of the left-hand side of inequality (1) is not really necessary for the successful realization of the method, in the case of its application to centres with unknown charge emission rate the repetition frequency f of the pulses should be chosen to be as high as possible†, keeping pulse period-to-pulse duration ratio constant. It is easy to understand that in the case when $\tau_c > f^{-1}/2$ the approach to steady-state (periodic in time) kinetics of charge states will take not one, as in the opposite case, but several periods of the frequency. The novel procedure would make it possible to gain new information about the migration characteristics (diffusion lengths, coefficients, lifetimes, etc) of species by monitoring their motion in the depletion region of a reverse-biased diode structure.

† This maximal frequency is restricted from above by the demand on the shape of pulses, which requires that the duration of the edges of pulse should be small compared with the period of frequency and is determined by both the diode's capacitance and the output resistance of the pulse generator employed. The total duration of both edges of the pulse in our experiments did not exceed 15–20% of the period.

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