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Evidence of formation of tetravacancies in uniformly oxygen irradiated n-type silicon

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ABSTRACT

High purity n-type silicon single crystal with resistivity in the order of 4000 Ω cm has been irradiated with high-energy oxygen ions at room temperature up to a fluence of 5E15 ions/cm^2. The energy of the beam was varied from 3 to 140 MeV using a rotating degrader to achieve a depthwise near-uniform implantation profile. Radiation induced defects and their dynamics have been studied using positron annihilation spectroscopy along with isochronal annealing up to 700 °C in steps of 50 °C for 30 min. After annealing the sample at 200 °C for 30 min, formation of silicon tetravacancies has been noticed. The formation of the tetravacancies was found to be due to agglomeration of divacancies present in the irradiated sample. An experimentally obtained positron lifetime value of 338 ± 10 ps has been reported for silicon tetravacancies, which has a very close agreement with the value obtained from recent theoretical calculations. The tetravacancies were found to dissociate into trivacancy clusters upon further annealing. The trivacancies thus obtained were observed to agglomerate beyond 400 °C to form larger defect clusters. Finally, all the defects were found to anneal out after annealing the sample at 650 °C.

1. Introduction

Radiation damage study in silicon crystal is important as radiation-induced defects substantially affect the behavior of the devices fabricated out of it. Nuclear radiation detectors, when placed in radiation environment, show gradual changes in their vital properties with time, like deterioration of resolution, increase in leakage current, etc., eventually prohibiting their long-term use as measurement devices [1–3]. Several types of radiation-induced defects have been identified till date in silicon. Defects like monovacancies (*V*), divacancies (V_2), vacancy–oxygen (*V–O*) complexes and vacancy phosphorus (*V—P*) complexes have been studied in great detail. Techniques like electron paramagnetic resonance [4], infrared spectroscopy [5], deep level transient spectroscopy [6] and positron annihilation spectroscopy (PAS) [7] have been heavily employed to identify and characterize these defects.

Theoretical calculations [8–10] do support the fact that formation of vacancy complexes like V_2, V_3, V_4, V_5, V_6, etc., is quite possible in silicon. But except in the case of V_2, very few experimental data are available in the existing literature regarding the nature of higher order vacancy complexes. Divacancies can be readily introduced into the silicon crystal using electron or other ion-irradiation; hence, have been studied in great detail [11,6]. Higher order vacancy complexes can be studied when there is a possibility of agglomeration of smaller vacancy clusters to form a higher order vacancy complex. The agglomeration process can take place during irradiation or during post irradiation treatment like thermal annealing when smaller vacancy clusters acquire sufficient mobility upon heat treatment [12]. But again, as pointed out by Haley et al. [13], vacancy clustering process on thermal treatment depends on several factors such as the cluster size, vacancy interaction range, vacancy concentration, etc., leading to uncertainty in the formation of a particular vacancy cluster.

The present article reports the formation of four-vacancy clusters, also known as tetravacancies that are denoted by V_4, during annealing of oxygen irradiated FZ grown n-type silicon crystal. Deliberate introduction of oxygen in silicon crystal is a very effective technique to fabricate radiation-hardened detectors [14]. High-energy oxygen ions can be used to get them implanted deep inside the crystal, which is needed for fabrication of a practical radiation-hardened detector, but the process of ion-implantation introduces several defects in the crystal. In order to remove these defects it is required to anneal the irradiated sample prior to detector fabrication. The thermal annealing in the present case has been carried out in intervals of 50 °C and the radiation induced defects have been studied at each step. Each annealing step was of 30 min duration. This type of measurement facilitates the study of defect dynamics and determination of actual temperature of a particular type of defect to anneal out. The defects have been characterized using positron annihilation lifetime spectroscopy (PALS) and Doppler broadening of positron annihilation radiation

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(DBPAR) spectroscopy. Positron annihilation spectroscopy is a very effective tool to detect and identify vacancy type defects and their complexes in semiconductors [7].

In the present article, we report the experimentally obtained positron lifetime in silicon tetravacancy. The experimentally obtained value has been found to be in very good agreement with the results of most recent theoretical calculations. The formation mechanism and stability of the tetravacancies have been discussed and were compared with existing reports in the literature on tetravacancies in silicon. From the available theoretical calculations, the structure of the tetravacancies detected in the present study is proposed to be of "part-of-a-hexagonal-ring" (PHR) configuration.

2. Experimental details

N-type detector-grade silicon wafer (resistivity $\sim$4000 Ω cm) of thickness around 500 μm and diameter about 15 mm were sliced from an ingot along the ⟨1 1 1⟩ crystal plane. The crystal was irradiated with 140 MeV oxygen ions (O^{6+}) up to a fluence of 5×10^{15} ions/cm^2 using the beam-line of Variable Energy Cyclotron Centre (VECC), Kolkata, India. The energy of the beam from the cyclotron was varied using a rotating degrader. The rotating degrader is a rotating mechanical wheel containing slots fitted with aluminium absorbers of different thicknesses placed in the path of the beam. As a result, the energy of the outgoing beam from the degrader varied in a regular cyclic manner from 140 to 3 MeV. The details of the experimental set up can be found in Ref. [15]. The outgoing beam energies and their corresponding ranges in the sample have been illustrated in Fig. 1.

For PALS and DBPAR measurements a 12 μCi ^{22}Na positron source (deposited on aluminium foil) has been used. The PALS system used in this work was a standard fast–fast coincidence setup with two identical 1 in. tapered-off BaF_2 scintillator detectors fitted with XP2020Q photomultiplier tubes. The time resolution obtained using ^{60}Co source with ^{22}Na gates was 290 ps. A total of more than 1 million counts were recorded for each lifetime spectrum. All lifetime spectra were analyzed using PATFIT 88 program [16] after background and source corrections. A total of 15% contribution from the source and an average background count of 20 per channel have been incorporated as corrections during the analysis. The source correction was carried out following the procedure adopted by Staab et al. [17]. Up to an annealing temperature of 600 °C, all lifetime spectra of the irradiated samples were deconvoluted using one Gaussian resolution function with two exponential lifetimes. Beyond 600 °C, a single component fit gave satisfactory results. DBAR spectra were recorded for two hours using a single HPGe detector with an energy resolution of 1.7 keV for 662 keV gamma rays from a ^{137}Cs standard source. Doppler broadened energy spectra were analyzed using the code SP (version 1.0) [18] to calculate the *S*-parameter. The *S*-parameter has been calculated as the ratio of number of counts integrated under a central segment spreading over $\pm$0.8 keV around the centroid of the 511 keV annihilation peak to the number of counts under the full spectrum.

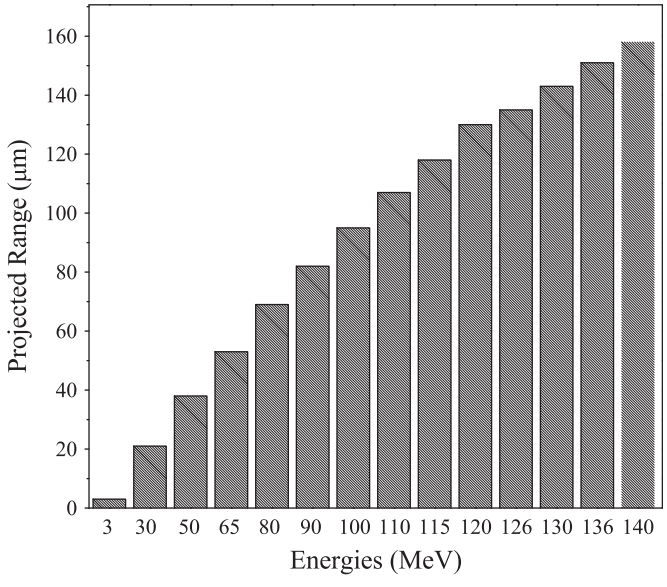

Fig. 1. Beam energies obtained from the rotating degrader and their corresponding ranges in silicon.

The isochronal annealing treatments were carried out in the temperature range 100–700 °C in intervals of 50 °C. Each annealing was carried out in vacuum ($\sim10^{-5}$ mbar) for 30 min. Several data (PALS as well as DBPAR) were retaken and reanalyzed and the resolution and peak position were monitored intermittently to check the reproducibility of the results and the stability of the spectrometer. A reasonably good reproducibility and stability of the spectrometer was observed over the entire period of experiment. During the deconvolution process the variance of a fit was minimized to obtain the best fit and at the same time the standard deviations of the individual lifetime parameters were checked not to give abnormally large values even if it shows a lesser variance in the overall fit.

3. Results and discussion

3.1. Doppler broadening of annihilation radiation studies

The Doppler broadening of the 511 keV annihilation gamma peak reflects momentum distribution of the annihilating electrons. In open volume defects, because of significantly low momentum of the valence electrons, the momentum distribution of annihilating electrons shifts to smaller values; hence, Doppler broadening of the annihilation line decreases [19]. Therefore, the annihilation peak of a defect-rich sample becomes higher and narrower compared with that of a defect-free sample, when both the peaks are normalized to equal area. As a result of this, *S*-parameter of a defect-rich sample becomes higher compared with that of a defect-free sample. However, the value of *S*-parameter so obtained, is the weighted average of *S*-parameters for positrons annihilating in the defect-free bulk region (S_b) as well as those trapped in the vacancy type defects (S_d). S_d can be calculated using the relations

$$S=(1-f)S_b+f\,S_d \tag{1}$$

where

$$f=K/(K+\tau_b^{-1}) \tag{2}$$

and K is the trapping rate of positrons in the vacancy type defects obtained from lifetime measurements (will be discussed in details in the next section) and τ_b is positron lifetime in bulk.

Fig. 2 shows the variation in S_d of the irradiated sample as a function of annealing temperature. It increases sharply at 200 °C, then falls to lower value and then maintains a more or less constant value up to 400 °C and again increases gradually up to an annealing temperature of 500 °C. Since *S*-parameter is sensitive to both concentration and size of the defects, the observed increase in the defect related *S*-parameter values may be due to vacancy agglomeration or may be due to increase in concentration of a particular type of defect as well. A confirmatory answer can be obtained by studying the deconvoluted lifetime parameters (to be discussed in the next section). Beyond 500 °C, the defect related