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Supercrystalization Produced With X-Rays and Applications

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ABSTRACT: *The X-rays influence on NaCl and KCl very fast crystallization in a saturated water solution and introduction of phosphorus and boron in crystalline silicon at room temperature were investigated. These new technologies or superdiffusion of added impurities do not harm the Si lattice and can be used for production of a new kind electronics devices and solar cells of high quality. Using the superdiffusion generated by soft X-rays we investigated the incorporation of carbon into silicon and produced the diodes based on p-type Si and SiC junctions formed by irradiation with X-rays. The reverse and exponential forward currents of produced diodes with SiC-Si junctions are more stable by heating than silicon diodes.*

PACS 78.70.Ck – X-ray scattering
PACS 32.80.Hd – Auger effect
PACS 61.72.-y – Defects and impurities in crystals; microstructure

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I. INTRODUCTION

We applied soft X-rays for generation of metastable vacancies in silicon crystal. This is sufficient for practical realization of new kind of superdiffusion [1], for introduction of boron and phosphorus in crystalline silicon [2] at room temperature. The proposed technologies do not harm Si lattice because for displacement of impurities atoms in crystal silicon is used Auger effect taking part in displacement of only one crystal's lattice's atom and therefore we can produce electronic devices with less defects.

Also it was found that irradiation of a water solution of NaCl [3] and KCl [4] has significant influence on two-step processes of crystallization. The irradiation of the solution produces metastable radicals of water and excited seeds which stimulate a very fast crystallization. The fast growth of crystals can be explained by creation of an irradiated crystallization centers on their surfaces (Fig. 1) and vacancy-interstitial pairs in the volume of growing crystals. By using the founded supercrystallization technologies we obtained that the linear rate of crystallization of NaCl were increased [3] about 100 thousand times.

II. SUPERDIFFUSION OF CARBON INTO CRYSTALLINE SILICON

By using the superdiffusion we investigated the incorporation of carbon into silicon what has interest for production of the new electronic devices, such as SiGeC hetero bipolar transistors, light emitters and diodes.

Usually the epitaxial layers of silicon carbide are growing [5] in silane (SiH_4) and propane (C_3H_8) gasses on silicon wafers (100) with surface's temperature of 1350 ^{0}C. The thickness of layers of typical samples which are grown by standard methods for time of 1h is between 2 and 3 μm.

On the surface of *p* type Si substrate we vaporized carbon by using tapered rods. These rods with closed tapered ends were placed in a vacuum chamber. Through them we applied the electrical current of 100-300 A for about 2-5 seconds. The closed ends of the rods are heated until become white (approximately till 1000 ^{0}C) and carbon atoms in a vaporized phase will be deposited on surface of silicon wafer forming polycrystalline film. The thickness of produced silicon carbide is about 0.35 μm.

The samples of the Czochralski type silicon crystals covered with 0.1 μm thickness layer of carbon were irradiated by $t = 2h$ time with X-rays using 8 kV voltage and 20 mA current from copper anode of Russian diffractometer DRON-3M. We obtained increasing of substitutional carbon concentration $0.68 \cdot 10^{16} cm^{-3}$ in the irradiated sample. Formed silicon carbide precipitations produce polycrystalline film presented in Fig. 2. The obtained precipitates (Fig. 2) of polytype Si5C3 can be used for production of electronics devices based on silicon carbide.

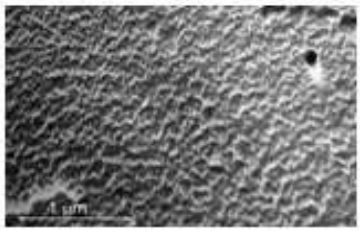

Fig. 1. KCl surface irradiated with X-ray.

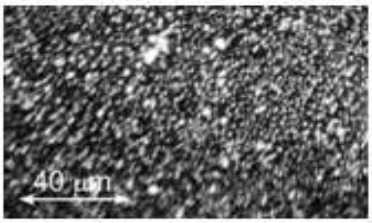

Fig. 2. Silicon carbide precipitates

The X-ray photons absorbed in the surface region of crystal silicon in the inner *K* shells of Si atoms produce photoelectrons and Auger electrons generating mobile vacancies and its complexes. X-rays influence on the point defects production in Cz-Si was detected by infrared absorption. Using transmittance measurements we obtained increasing $\Delta\alpha$ = 0.068 cm^{-1} of absorption coefficient at band 605 cm^{-1} after irradiation of 1 mm thickness sample of Cz-Si crystal covered with epitaxial 0.1 μm layer of carbon. After multiplying $\Delta\alpha$ on the calibration factor $\sigma_C = 1.0 \cdot 10^{17} cm^{-2}$ we founded increasing of C_S atoms concentration [6] on $0.68 \cdot 10^{16} cm^{-3}$ by superdiffusion of interstitial silicon atoms' C_i in a silicon crystal at room temperature.

After irradiation with X-Rays we measured the increasing of absorption coefficients. By using FTIR spectroscopy [7] the produced mobile complexes [8] *VO*, VO^- with absorption bands at 836 cm^{-1}, 885 cm^{-1} and metastable complexes VO_2 at 1002 cm^{-1} were found**.** The latter values are determined by the efficiency of vacancies production after irradiation with soft X-rays. Here we have the production of photoelectrons and Auger electrons generating charged, neutral vacancies and mobile interstitials I_{Si} atoms of silicon. After reactions $VO + I_{Si} \rightarrow O_i$, $C_s + I_{Si} \Leftrightarrow C_i$ the mobile interstitials O_i, C_i, I_{Si} were obtained. The increasing of IR absorption $\Delta\alpha = \alpha_x - \alpha_o$ of the irradiated point defects and complexes' are calculated using FTIR infrared measurements. We obtained the superdiffusion of carbon atoms from epitaxial layer into the Si sample. The movement of interstitials O_i, C_i, I_{Si} produces the growth of the layer of a polycrystalline silicon carbide presented in Fig. 2 at the surface of Si crystal. The small crystallites are growing because their surface energy is greater than the surface energy of silicon. Multiplying the changes of absorption coefficients $\Delta\alpha$ at some bands and corresponding calibration factor [9] $\sigma \cdot 10^{17}$ cm^{-2} we can obtain concentrations of the generated complexes, corresponding point defects at some bands and concentrations of substitutional C_s atoms (at band 605 cm^{-1}), interstitials (922, 932 cm^{-1}) of carbon C_i and complexes C_iO_i (866, 1116 cm^{-1}). The produced Si-C bonds at 722 and 814 (or 817) cm^{-1} [8], [9] are represented by increasing of absorption coefficients $\Delta\alpha$ =1.191 at band 812 cm^{-1}. We obtained the increasing of concentration by $6.041 \cdot 10^{17}$ cm^{-3} of interstitial oxygen atoms [9] after irradiation from the absorption band 1107 cm^{-1}. We also calculated [9] increasing of density of substitutional carbons' atoms C_s by $1.189 \cdot 10^{17}$ cm^{-3} at band 605 cm^{-1} in MCZ-Si sample after 2 h irradiation using Cu anode with applied voltage of 8 kV and tube current of 20 mA for measured increasing of absorption coefficient by $\Delta\alpha$ =1.189 and the calibration factor $1.00 \cdot 10^{17}$ cm^{-2}.

III. PRODUCTION OF THE NEW KIND DIODES

The p-type Si crystal of 0.3 mm thickness covered with 0.1 μm C layer was irradiated by X-rays with voltage U_a = 7 *kV* and current *I* = 7 *mA* of Cu anode by time *t* = 27 *h* with difractometer DRON-3M for production of photoelectrons and Auger electrons for generating charged, neutral vacancies and *Si* interstitials I_{Si}. After reactions $VO + I_{Si} \rightarrow O_i$, $C_s + I_{Si} \Leftrightarrow C_i$ were obtained the mobile interstitials O_i, C_i, I_{Si} which formed *SiC* layers denoted like spectrum 1 (atomic composition 80.81% *Si* , 19.19 % *C*) and Spectrum 2 (92.09% *Si*, 7.91% *C)*with common width 390.4 nm presented in Fig. 3 measured by quantum scanning electron microscope (FEI company). The obtained structure formed high resistivity n-type layer of precipitations [1] consisting **of** crystallites Si5C3 (structure defined by Smart Lab Rigaku diffractometer) and Si crystal. That enables us to form p-n junctions (see Fig. 2) by using Au contacts on SiC and SiC-Si-Al junctions as indicated in Fig. 4 presenting parameters of X-ray irradiations using Cu anode. The p-Si substrate of the diode is 0.5 mm of with and 40 Ω · cm of resistivity.

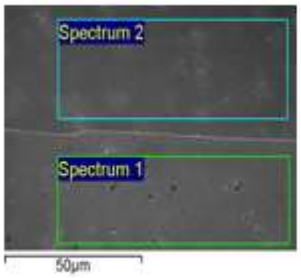

Fig. 3. Atomic compositions *Si* and *C*

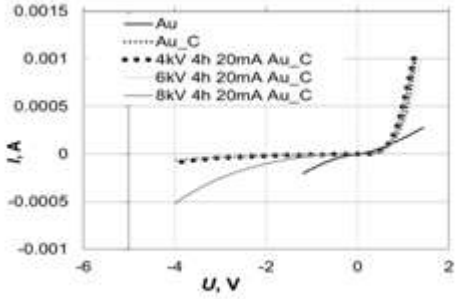

Fig. 4. Volt-Ampere characteristics of pn junctions

The volt ampere characteristics of p-n junctions (presented in Fig. 4) depends on parameters of irradiation (voltage, time, current) defining the atomic composition of p *Si and SiC* junctions. *Au* is used as upper contact for *SiC* layer and lower contact from *Al* with p type *Si.* We can see that in the first non-irradiated case the contact *Au* and *Si* like the p-n junction was not obtained. The second volt-ampere characteristics of the diodes *Au_C* are presented for the following four cases of irradiation with Cu anode: for time of 4 h for current 20 mA in X-ray tube and voltages 4 kV, 6 kV, 8 kV. The flow of electrons in opposite direction represented in Fig. 4 is very small for irradiation with 4 kV and 6 kV voltages and produced diodes allow the flow of current in only one direction. Not so good diode was obtained using irradiation at 8 kV voltages. These results coincide with the decreasing of the X-ray's absorption coefficients for increasing energies [10]. As absorption coefficients are proportional to cross-sections of photoelectric emission of electrons' the probability of vacancies production in Si lattice has its maximum at 4 keV energy's irradiation. That allows us easy to explain the formation of maximum width of SiC layer in the process of our diodes production. We measured the temperature dependences of volt-ampere characteristics presented in Fig. 4 of p-n junctions and obtained that exponential forward and reverse saturation currents at 100 ^{0}C stay not changed comparing with 20 ^{0}C. After heating the sample till 200 ^{0}C the currents are increasing in both directions and then if we cool it till 20 ^{0}C the voltage break down. The diodes produced according proposed technologies have significantly large temperature stability then silicone diodes. Also the degradation of produced diodes with greater width of SiC layers occurs at significantly higher temperatures.

IV. CONCLUSION

Instead of superdiffusion a growth of epitaxial layer can be produced by the chemical vapor deposition [11] by using the doping densities ($10^{14} – 10^{19}$ cm^{-3}) in n and p-type materials controlling the C/Si ratio. In SiC both atoms tetrahedrally bounded with covalent bonds by sharing electrons pairs in sp^3 hybrid orbitals displaying Si-C pairs in three different positions and layers [11] which produce over 200 polytypes or different structures. Standard technique for Si bulk growth is the seeded sublimation at high temperatures [11]

above 2000 C and up to 15 % carbon can be dissolved in a Si melt at 2800 C. It is interesting that we used X-rays for SiC polycrystalline layer production with C atoms concentration 19.19% in Si and C substrate at room temperature.

For the epitaxial growth of SiC layers can be used the method of chemical vapor deposition by employing the gases C_3H_8, SiH_4 and C_3H_8 or C_2H_4 as precursors. Like the carrier gas can be applied H_2 and Ar. The crystallization rate at temperatures 1500-1650 ^{0}C is 3-15 μm/h. From the [10] and Fig. 3 we obtained that using superdiffusion of carbon into silicon generated by 10 times' of greater X-rays intensity we obtained the similar rate of growth 11 μm/h. This fact allows us to expect the successful practical applications using X-rays for production of electronic devices.

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