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RAMAN SPECTROSCOPY OF GAMMA-IRRADIATED SILICON DOPED WITH RHODIUM

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This research investigates the impact of gamma-ray irradiation and rhodium (Rh) contamination on the crystalline structure of silicon through Raman spectroscopy. The presence of rhodium in silicon single crystals induces slight structural changes and introduces new elements in the Raman spectral profile. Notably, the intensity of the silicon signature peak at 521 cm^{-1} shows a marked increase, while its full width at half maximum (FWHM) remains essentially unchanged. This rise in peak intensity is likely due to enhanced bonding interactions within the silicon lattice resulting from the incorporation of Rh. Additionally, the new Raman signal at 245 cm^{-1} in the Si<Rh> spectra is attributed to elemental rhodium and the formation of RhSi compounds. Furthermore, irradiation of n-Si with 1.25 MeV gamma rays and a dose of 107 rad disrupts and amorphizes the silicon crystal structure and creates radiation-vacancy defects. The irradiation results for the doped samples indicate that the addition of rhodium atoms reduces the amorphous content in silicon and enhances its crystalline structure.
Keywords: *Silicon; Rhodium; Doping; Irradiation; Gamma quantum; Raman spectroscopy*
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INTRODUCTION

Silicon (Si) plays a vital role in microelectronics, serving as the foundation for integrated circuits, transistors, and various semiconductor components. Nevertheless, when exposed to radiation – such as in space environments, nuclear reactors, or areas with ionizing radiation – its structural properties can undergo considerable alterations. These changes lead to degradation of its electrical, mechanical, and optical properties, which restrict its use in extreme conditions [1-3].

The study of radiation effects in silicon is essential for the development of radiation-hardened semiconductor devices used in the military, aerospace, and nuclear industries. In addition, radiation exposure is also used in controlled doping of silicon, making it an active subject of scientific research [4,5].

When silicon is irradiated with various types of radiation (gamma rays, X-rays, electrons, neutrons, heavy ions), the crystal structure of the material changes. This is due to the ionization of atoms, the formation of interstitial defects, and vacancy complexes. The main mechanisms of destruction of the silicon structure include: Ionization processes, Dislocation defects and lattice damage, and Amorphization of the structure [6].

One effective way to increase the radiation resistance of silicon is to alloy it with transition elements such as gold (Au), platinum (Pt), rhodium (Rh), iridium (Ir), etc. These elements play a key role in reducing the concentration of radiation defects and minimizing their negative impact on the material's electrical properties [7,8].

The study of the effect of radiation on the structure of silicon has both theoretical and applied significance. Radioprotective technologies and methods for increasing silicon's resistance to radiation exposure are important areas in microelectronics and materials science.

The radiation effects on Si under the influence of γ-quanta have also been well studied. Research shows that high-energy γ-radiation causes the formation of point defects and partial amorphization at high total doses, which is manifested by a decrease in the intensity of the crystalline peak and an enhancement of the phonon continuum in the low-frequency region [9-11].

Considerable attention in the literature has been devoted to the influence of transition metal impurities on defect formation in silicon. Elements such as Ni, Co, Pt, and Pd can interact with radiation defects, form stable metal-silicide phases, and act as recombination centers for vacancies and interstitial atoms [12]. It has been shown that silicide formation increases lattice stability against radiation damage and reduces the degree of amorphization [13].

Platinum-group metals, including rhodium, are strong silicide-forming elements. Although there are significantly fewer studies on the Rh–Si system than on the Pt–Si or Pd–Si systems, existing data indicate that rhodium can form stable compounds with silicon and influence the evolution of defects in the matrix [14,15]. Similar studies have shown that Pt and Pd increase the radiation resistance of silicon by suppressing the accumulation of the amorphous phase [16], suggesting a similar analogy and mechanisms for Rh.

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This study aims to investigate the defect structure of silicon doped with rhodium and exposed to gamma radiation, utilizing Raman spectroscopy as the primary analytical method.

Raman spectroscopy is an optical method of material analysis based on the inelastic scattering of monochromatic light (usually a laser) by molecules of the material being studied. This method provides information on vibrational, rotational, and other low-energy states of molecules, making it a powerful tool for studying the structure and properties of materials. This article presents results on the change in the structure of rhodium-doped silicon irradiated with gamma quanta.

EXPERIMENTAL PART

The research was conducted on n-type silicon (n-Si) with a specific resistivity of 40 Ω·cm, fabricated using the Czochralski method. The initial doping concentration of phosphorus in the n-Si single crystals ranged from 8×10^{13} to 7×10^{14} cm^{-3}. Rhodium was incorporated into the silicon via diffusion, which entailed placing rhodium atoms on the silicon surface inside evacuated quartz ampoules. This diffusion process was performed at temperatures ranging from 1100 to 1200°C for durations of 3 to 5 hours. Following the diffusion, the samples were subjected to different cooling rates. [17,18].

The original (n-Si) and doped samples (n-Si<Rh>) were irradiated with gamma quanta with an energy of 1.25 MeV, a dose of 10^7 rad using a ^{60}Co gamma source at the INP ASUz.

The samples were examined using Raman spectroscopy with a 785 nm laser, employing an Ocean Optics Raman spectrometer capable of analyzing the spectral range from 0 to 2000 cm^{-1}. All spectral measurements were carried out at room temperature. Additionally, a Carl Zeiss EVO10 scanning electron microscope (Zeiss, Oberkochen, Germany) was used to analyze the elemental composition and obtain micrographs of the samples.

RESULTS AND DISCUSSIONS

Figure 1 displays the Raman spectra of the original silicon, rhodium-doped silicon, and gamma-irradiated samples. In the spectrum of the undoped silicon, prominent peaks are observed at 305 cm^{-1} and 521 cm^{-1}, along with a broad band spanning from 913 to 1039 cm^{-1} – features that are indicative of the cubic crystalline phase of silicon. It is established [19] that silicon with a diamond cubic structure exhibits a primary first-order Raman-active phonon mode near 520 ±1 cm^{-1}, associated with the longitudinal optical (LTO) mode, along with additional weaker features resulting from long-range translational symmetry. The peak near 304 cm^{-1} corresponds to the longitudinal acoustic (LA) phonon mode, as noted in references [20, 21]. The broad band observed in the 913–1039 cm^{-1} range has been experimentally detected in both single-crystalline [22] and nanocrystalline silicon [20, 21, 23], and is associated with the scattering of multiple transverse optical (2TO) phonons and their overtone vibrations [19, 24].

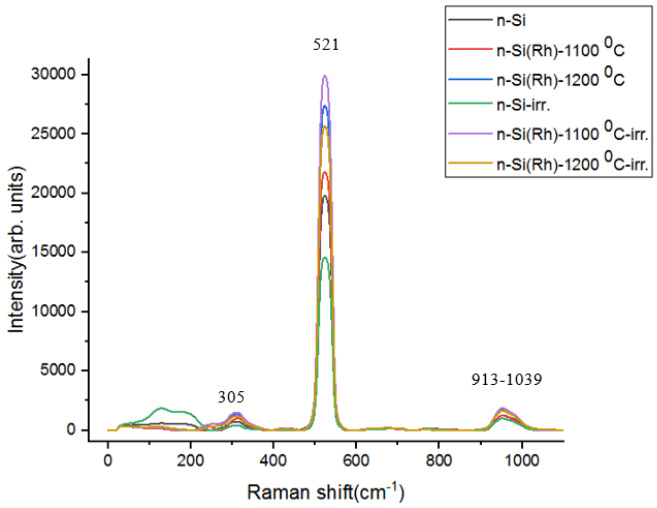

Figure 1. Raman spectra of silicon doped with rhodium and irradiated with gamma quanta

Additional doping of silicon with rhodium results in subtle modifications and the emergence of new vibrational features in the Raman spectra. In the doped samples, the intensity of the primary peak at 521 cm^{-1} increases – by a factor of 1.17 at 1100°C and 1.55 at 1200°C – while the full width at half maximum (FWHM) remains nearly unchanged (Fig.2b). This shows that after doping, rhodium can form complexes with defects in silicon, which reduces inelastic losses of scattered light and increases the scattering yield, strengthens the bonds in the structure of the silicon crystal lattice due to the incorporation of Rh atoms.

In addition, a new peak with low intensity at 245 cm^{-1} Raman shift (Fig. 2a) appears in the Raman spectrum of doped samples. The formation of this peak is due to the introduction of rhodium atoms into the crystal structure of silicon and the formation of rhodium silicide (RhSi) after high-temperature diffusion. Rhodium is recognized as one of the few transition metal impurities in silicon that can induce surface turbidity [25]. This effect is primarily attributed to rhodium's high diffusion coefficient and significant solubility in silicon. Rapid cooling (quenching) of rhodium-doped silicon

samples from high diffusion temperatures down to room temperature results in the formation of electrically active rhodium atoms occupying substitutional positions within the silicon lattice.

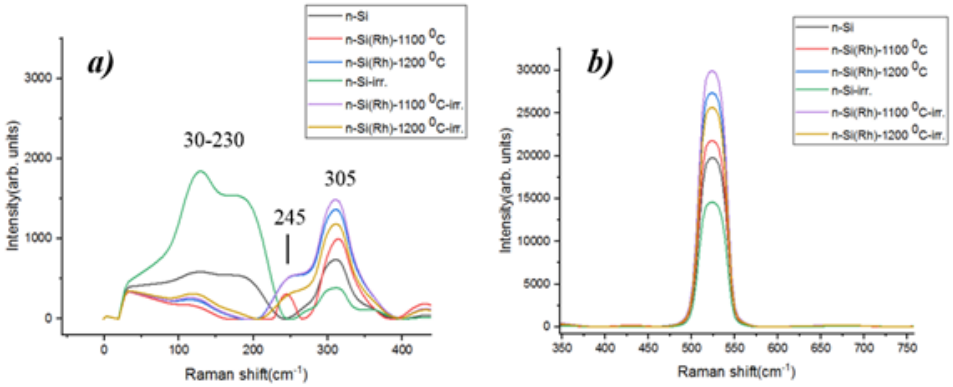

Figure 2. Fragments (a and b) of Raman spectra of single-crystal silicon, doped with rhodium and irradiated with gamma-rays.

Further analysis confirmed the presence of rhodium in the single-crystal samples through X-ray spectral analysis, which revealed a rhodium concentration of 0.68 atomic percent (at.%) or 2.4 weight percent (wt.%). As shown in the energy-dispersive spectra in Figure 3, the samples also contain oxygen and carbon atoms, with concentrations of 1.23 at.% (1.82 wt.%) and 1.84 at.% (1.32 wt.%), respectively, alongside the detected rhodium.

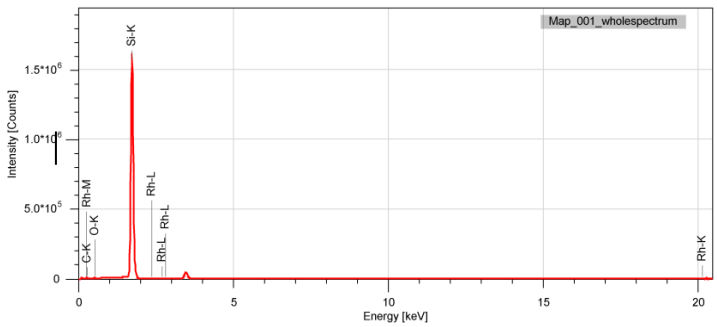

Figure 3. Energy dispersive spectra of a single crystal of silicon doped with rhodium

The change in doping temperature had almost no effect on the concentration of rhodium atoms on the silicon surface. The distribution map (Fig. 4) of impurity atoms in silicon shows that, after high-temperature diffusion, rhodium atoms formed micro- and nanostructures that were uniformly distributed on the silicon surface.

Furthermore, all studied samples were irradiated with 1.25 MeV gamma rays to a dose of 10^7 rad. Some changes in the Raman spectrum of the irradiated samples were determined. After gamma irradiation, the intensity of the main Raman peak at 521 cm^{-1} in the original silicon sample decreases by approximately 1.35 times, indicating a degradation of the crystalline structure. Furthermore, the broad peak observed in the 30–230 cm^{-1} range – associated with the amorphous phase of silicon – becomes more pronounced in the irradiated sample, suggesting an increase in structural disorder.

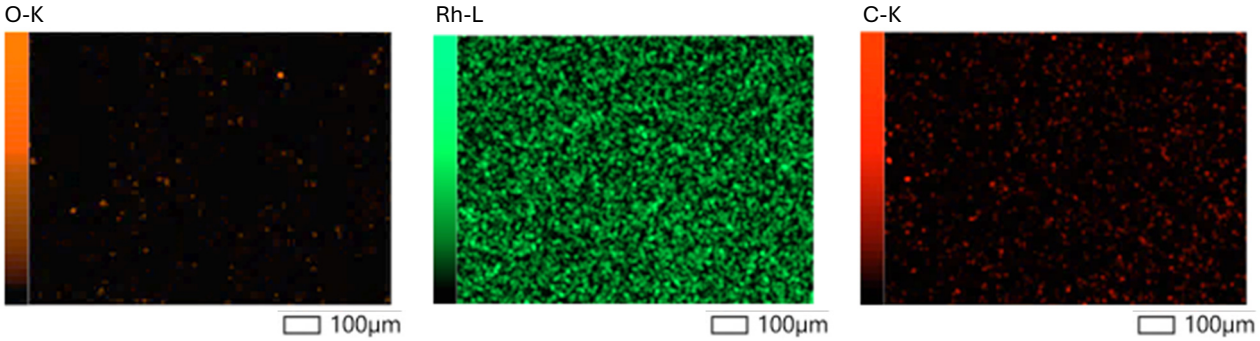

Figure 4. Micrographs of the surface of a Si single crystal after Rh doping with distribution maps

As noted by the authors in [26], exposing silicon to various forms of radiation results in the formation of radiation-induced defects – such as A-centers, divacancies, and E-centers – that introduce deep energy levels within the silicon bandgap. Among these, the vacancy–oxygen complex (A-center or VO) is one of the most prominent defects. This complex creates an acceptor level at approximately Ec – 0.17 eV. When a vacancy forms, it can trap an oxygen atom, which then shifts into the vicinity of the vacancy, residing near the vacant lattice site without wholly occupying a substitutional position [27].

Divacancy is the second most concentrated radiation defect in irradiated silicon. At high irradiation doses, when the accumulation of A-centers is limited, divacancy becomes the dominant radiation defect in concentration [28]. The next radiation defect is considered to be a vacancy-phosphorus complex (E-center), which creates a deep level in the forbidden zone of silicon with an ionization energy of Ec – 0.43 eV. All these radiation-induced defects are considered traps for the main charge carriers in silicon and worsen its electrophysical properties, leading to changes in the crystal structure. Based on [26-28], we can assume that irradiation of silicon leads to the creation of radiation defects, disruption of the crystal structure, and amorphization.

A summary of existing scientific data confirms that introducing transition metals and forming silicide phases can significantly increase silicon's resistance to γ-irradiation, reducing the concentration of structural defects and preventing the destruction of crystalline order. The experimental results obtained for Rh-doped silicon are consistent with the general trend identified in the literature.

The Raman spectra of the rhodium-doped (Si<Rh>) samples subjected to irradiation show an enhanced intensity of the primary silicon peak at 521 cm^{-1}, accompanied by a significant decrease in the broad band within the 30–230 cm^{-1} range. This suggests an improvement in the crystalline structure and a reduction in the amorphous content. This suggests a positive structural effect from the rhodium doping. Transition metals like rhodium are highly chemically active and can interact with radiation-induced defects such as vacancies and interstitials, promoting their recombination and thereby lowering the overall defect concentration [29]. Rhodium atoms can form stable complexes with these defects, effectively preventing them from participating in charge-carrier recombination [30]. Based on these findings, it can be concluded that incorporating rhodium into the silicon lattice reduces radiation-induced defect density and enhances the crystal's structural integrity.

CONCLUSIONS

Raman spectroscopy analysis revealed two distinct peaks at 305 cm^{-1} and 521 cm^{-1}, along with a broad band between 913 and 1039 cm^{-1} – all characteristic of cubic-phase silicon. Additional rhodium doping alters the spectral profile, leading to the appearance of a new peak at 245 cm^{-1}, attributed to the formation of rhodium silicide.

Using a scanning electron microscope, the concentration of impurities on the silicon surface was determined. It was found that, after high-temperature diffusion, rhodium atoms form micro- and nanostructures and are uniformly distributed on the silicon surface.

Further exposure of n-type silicon (n-Si) to gamma radiation with an energy of 1.25 MeV and a total dose of 10^7 rad results in a reduction of the main Raman peak at 521 cm^{-1} and the emergence of a broad feature within the 30–230 cm^{-1} range. These changes indicate damage and partial amorphization of the silicon crystal lattice. However, in irradiated rhodium-doped silicon (n-Si<Rh>), a different behavior is observed: the intensity of the main peak at 521 cm^{-1} increases, while the broad band associated with structural disorder in the 30–230 cm^{-1} region nearly disappears. These results suggest that incorporating rhodium atoms into the silicon matrix mitigates radiation-induced amorphization and enhances the preservation or recovery of the crystalline structure.

Conflict of Interest

The authors declare that there is no conflict of interest regarding the publication of this paper.

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РАМАНІВСЬКА СПЕКТРОСКОПІЯ ГАММА-ОПРОМІНЕНОГО КРЕМНІЮ, ЛЕГОВАНОГО РОДІЄМ

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Це дослідження досліджує вплив гамма-опромінення та забруднення родієм (Rh) на кристалічну структуру кремнію за допомогою раманівської спектроскопії. Присутність родію в монокристалах кремнію викликає незначні структурні зміни та вводить нові елементи в спектральний профіль раманівського розсіювання. Примітно, що інтенсивність піку кремнієвої сигнатури при 521 $см^{-1}$ демонструє помітне збільшення, тоді як його повна ширина на половині висоти (FWHM) залишається практично незмінною. Це збільшення інтенсивності піку, ймовірно, пов'язане з посиленням зв'язків у кремнієвій решітці внаслідок включення Rh. Крім того, новий раманівський сигнал при 245 $см^{-1}$ у спектрах Si<Rh> пояснюється елементарним родієм та утворенням сполук RhSi. Крім того, опромінення n-Si гамма-променями з енергією 1,25 МеВ та дозою 107 рад призводить до порушення та аморфізації кристалічної структури кремнію та до створення радіаційно-вакансійних дефектів. Результати опромінення легованих зразків показують, що додавання атомів родію зменшує вміст аморфного шару в кремнії та покращує його кристалічну структуру.

Ключові слова: *кремній; родій; легування; опромінення; гамма-квант; раманівська спектроскопія*