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# Impact of Gamma, Neutron, Ion, and Electron Irradiation on Structure and Properties of Graphene

*Biliak R. V.*

Lviv Polytechnic National University, Lviv, Ukraine

E-mail: [roman.v.biliak@lpnu.ua](mailto:roman.v.biliak@lpnu.ua)

This article discusses studies of the effects of various types of radiation, including  $\gamma$ -radiation, neutron, ion, and electron irradiation, on graphene and graphene-based devices. The study of graphene's response to radiation is crucial because of its potential applications in fields such as nuclear power and space exploration, where the impact of radiation is significant. This paper discusses recent experiments conducted to investigate the effects of  $\gamma$ -radiation on graphene layers and devices based on graphene, which revealed changes in graphene layer spacing, defect formation, and electrical characteristics. Similarly, studies of the effect of neutron irradiation on graphene demonstrate its resistance to such radiation, with graphene-based sensors retaining functionality even after exposure to high neutron flux. Moreover, studies of ion irradiation reveal the ability to modify the structure of graphene, although it causes significant damage. The electron irradiation creates defects, which in turn reduced noise levels in graphene-based devices, a unique characteristic not observed in traditional materials. Conclusions were also drawn regarding the effect of temperature on graphene, and it was found that elevated temperatures contribute to the reduction of defects through annealing, demonstrating the self-healing properties of graphene. The paper concludes by emphasizing graphene's resistance to radiation and its potential for use in high radiation environments where traditional materials may not be able to withstand. The results show that graphene-based sensors and devices can maintain functionality even in the presence of defects caused by radiation, which opens up promising prospects for applications in nuclear energy and space research. Further in-situ studies are recommended to better understand the real-time effects of radiation on device functionality. Overall, graphene is an excellent candidate for various applications due to its unique properties and radiation resistance.

*Keywords:* graphene;  $\gamma$ -radiation; neutron irradiation; electron irradiation; ion irradiation; self-healing effect; nuclear fusion

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## Introduction

As of today, graphene is one of the most actively researched materials worldwide [1]. The interest in this material is driven by its unique properties and two-dimensional structure [2, 3]. One key research direction for graphene is the study of the impact of irradiation on its structure and properties [4]. This issue is particularly relevant due to plans for using graphene-based devices and materials in nuclear reactors, the space industry, and other fields where the level of radiation exposure is significant, and conventional semiconductor devices cannot withstand such conditions [5–7]. Based on literature sources on the graphene structure, it can be concluded that it interacts weakly with radioactive radiation [8–10].

Thermonuclear fusion requires the use of new materials and sensors for accurate measurement of the magnetic field and plasma control in the reactor,

especially in the DEMO reactor. Considering the high neutron flux [11] and temperature conditions that can reach 350°C during the reaction [12], traditional Hall sensors may not cope with this task. For this reason, graphene-based sensors [11] are considered one of the possible alternatives.

Recently, SpaceX successfully launched the “Fourth Transporter” mission [13], which, in addition to commercial purposes, also included scientific research. Students from the Netherlands and Chile developed devices incorporating graphene elements to study the effects of space travel and space conditions on graphene components. Graphene elements will undergo a series of tests to check their performance and resistance to extreme conditions of the space environment, such as vibrations, radiation, and temperature changes. The obtained data will be used to develop sensors and instruments for navigation and astronomical observations in space [13].

## 1 Impact of $\gamma$ -radiation

Studying the impact of  $\gamma$ -radiation on graphene and micro- and nanoelectronic devices based on it is crucial due to their potential applications in the space industry and nuclear energy. Ensuring the reliability and long-term operation of such equipment requires understanding how  $\gamma$ -radiation affects various components and developing devices and materials resistant to this type of radiation.

In their research [14], Zhang and colleagues investigated the influence of  $\gamma$ -radiation on graphene layers. Graphene oxide was first obtained using the improved Hummers method, and then graphene oxide was exfoliated to obtain graphene oxide. Subsequently, graphene oxide was transformed into graphene using sodium citrate. The prepared graphene underwent heat treatment in a nitrogen atmosphere and was later irradiated with  $\gamma$ -rays from a  $\text{Co}^{60}$  source at a dose of 100 kGy with an accumulation rate of 2 kGy/h. Gamma irradiation resulted in a reduction in the distance between graphene layers but increased the size of the graphite crystal and the number of layers. The authors suggest that this increase was likely caused by cross-linking opened due to gamma irradiation [14].

Other researchers investigated the impact of  $\gamma$ -radiation on graphene-based devices. Xu and colleagues [15] studied the effects of  $\gamma$ -radiation on graphene/Si Schottky diodes.  $\text{Co}^{60}$  served as the radiation source, and the devices were irradiated without electrical bias. The devices were irradiated up to a dose of 10 kGy at a dose rate of 0.5 Gy/s. The research revealed an increase in reverse current after irradiation.  $\gamma$ -radiation disrupted C=C bonds in graphene and Si-O bonds in  $\text{SiO}_2$ . Photoelectron spectroscopy showed the formation of defects in the form of unpaired Si bonds.  $\gamma$ -irradiation of graphene/Si Schottky diodes led to the degradation of their electrical characteristics, including a reduction in the Schottky barrier height, resulting in a significant increase in reverse current [15].

Another team of scientists led by Xi [16] investigated the impact of  $\gamma$ -radiation on energy-independent memory using graphene nanodisks Fig. 1.  $\text{Co}^{60}$  was also used as the radiation source, and it was found that irradiation caused electron loss through photoemission. Despite significant shifts in the graphene and surrounding oxide conduction zone, data retention after irradiation remained satisfactory. The researchers concluded that  $\gamma$ -radiation significantly affects devices in programmed states, but data retention after irradiation is acceptable, with devices exhibiting good data retention even after irradiation with a dose of 10 kGy [16].

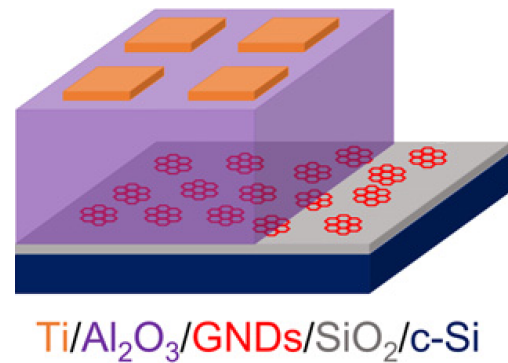


Fig. 1. Scheme of graphene nanodisk memory [16]

Similar results were obtained by a team of scientists from the Brookhaven National Laboratory [17] on Long Island, New York, USA. The team studied the impact of  $\gamma$ -quanta on the graphene field-effect transistor (GFET) schematic representation of the transistor shown in Fig. 2.  $\text{Co}^{60}$  served as the traditional radiation source, and the radiation dose was approximately 1 kGy/h, similar to previous studies. The study found that transistors exhibit high sensitivity to this type of radiation.  $\gamma$ -radiation resulted in a decrease in electron mobility in graphene and a shift of the Dirac point towards positive p-doping.

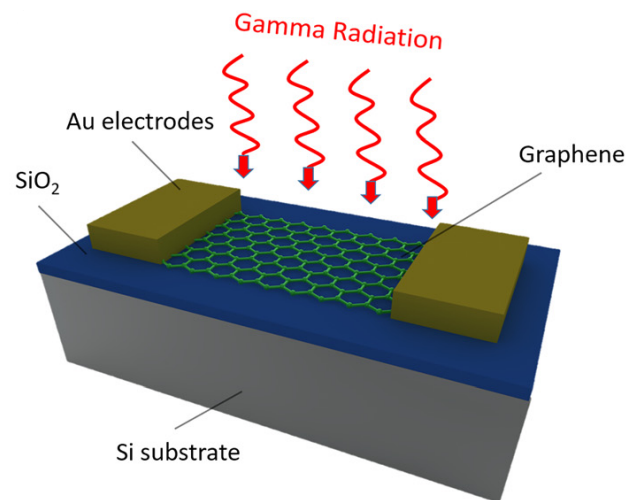


Fig. 2. Schematic representation of The graphene field-effect transistor (GFET) [17]

Raman spectroscopy indicated an increase in the D-peak to G-peak intensity ratio after  $\gamma$ -irradiation, suggesting an increase in disorder in monolayer graphene [17].

Fan and his colleagues [18] investigated the impact of gamma irradiation on a Hall sensor based on graphene.  $\text{Co}^{60}$  was used for irradiation with a dose rate of 50 rad(Si)/s. The samples were irradiated without applied bias voltage. Different doses were obtained by varying the contact time. Hall measurements were conducted at various irradiation doses: 100, 300, 500, and 1000 krad(Si) [18]. The sensitivity of the sensor decreased with increasing irradiation doses. For example, at a dose of 1 Mrad(Si), the sensitivity at

a bias current of 50  $\mu\text{A}$  decreased to 65.1% of the initial value [18]. The carrier mobility also changed during irradiation, decreasing from an initial average value of 823.3  $\text{cm}^2/\text{Vs}$  to 523.05  $\text{cm}^2/\text{Vs}$  at doses up to 1 Mrad(Si) [18].

As a result, it can be confidently stated that this type of irradiation affects the physical and electrical properties of graphene. The acquired defects degrade graphene properties, and this type of radiation influences the interactions between graphene and other substances within the device. It is noteworthy that, after such testing, all devices retained their functionality.

## 2 Impact of Neutron Irradiation

It is crucial to understand the impact of neutron irradiation on graphene, as this material exhibits higher radiation resistance [19]. This characteristic makes it promising for use in various sensors in the space industry and, particularly, in nuclear energy, including future thermonuclear fusion reactors.

Eapen and colleagues [20] from the University of North Carolina investigated the effects of neutron irradiation using the PULSTAR reactor with a flux of  $2.8 \times 10^{-9}$  dpa/s. The irradiation was performed at  $\sim 325$  K with an average fast neutron flux of  $2 \times 10^{12}$  n/( $\text{cm}^2 \cdot \text{s}$ ). The Raman spectroscopy results showed an increase in the intensity of the D peak after irradiation, indicating the formation of new defects in graphene. X-ray photoelectron spectroscopy (XPS) results indicated changes in the chemical structure of irradiated graphene samples [20]. Specifically, a high content of  $\text{sp}^3$  bonds was noted, likely formed during heating and cooling. These  $\text{sp}^3$  bonds may suggest the formation of new chemical bonds and defects in graphene.

In 2019, Bolshakova et al. [19] conducted the world's first study on the influence of neutron irradiation on Hall sensors based on monolayer graphene. The study was conducted in-situ, meaning information was collected from Hall sensors located in a magnetic field in the operating reactor. The neutron flux in the reactor was  $1.5 \times 10^{20}$  n/( $\text{m}^2 \cdot \text{s}$ ). Graphene for Hall sensors was grown using the CVD method on a copper foil, and the substrate was made of  $\text{Al}_2\text{O}_3$ . The experiment lasted for 48 hours. After irradiation, the sensitivity of the sensors remained unchanged, indicating that graphene-based sensors are resistant to neutron irradiation.

Another research team led by Semir El-Ahmar [6] also investigated the effects of neutron irradiation on Hall sensors based on graphene passivated with a 100 nm  $\text{Al}_2\text{O}_3$  layer. They used hydrogen-intercalated graphene grown on a semi-insulating high-purity single-crystal 4H-SiC(0001) substrate by the CVD method. Irradiation was performed at the National Center for Nuclear Research (NCBJ) using the

MARIA research nuclear reactor. MARIA is a high-performance device for extracting nuclear energy with a thermal neutron flux up to  $2.5 \times 10^{14}$   $\text{cm}^{-2}\text{s}^{-1}$  and fast neutrons up to  $1.0 \times 10^{14}$   $\text{cm}^{-2}\text{s}^{-1}$ . The samples were irradiated with fast neutrons at a fluence of  $\sim 6.6 \times 10^{17}$   $\text{cm}^{-2}$  for an exposure time of 121 hours. Electrical measurements were taken before and after irradiation, and the temperature during irradiation did not exceed 300°C [6].

Raman spectroscopy revealed a characteristic defect peak D, indicating that the neutron flux penetrates through the  $\text{Al}_2\text{O}_3$  layer to graphene, creating defects. Hall effect measurements showed that the structure after a nuclear reaction retained the characteristics of graphene, which would not have been the case if the sheet had undergone intensive chemical bonding with the substrate [6]. This indicates the stability of graphene to neutron irradiation. As a result of the study, scientists found that the defect density was seven orders of magnitude lower than the fluence, suggesting that graphene has a small cross-section for neutrons [6].

Neutron irradiation leads to structural changes in graphene, including the formation of defects and alterations in the chemical structure. However, Hall sensors in two studies [6, 19] did not lose their functionality, demonstrating the resilience of graphene to neutron irradiation.

## 3 Impact of Ion Irradiation

The study of the impact of ion irradiation on graphene is essential, as this type of radiation can be used to create defects in graphene in the form of nanopores [21] and alter its properties. Ion irradiation is also a common technique in the production of semiconductor devices [22]. Ions can be either atoms with unpaired electrons or whole molecules with unpaired charges [23].

A research team led by Kichul Yoon [24] investigated the influence of ion irradiation on graphene using the Zeiss ORION NanoFab ion microscope, operating at a voltage of 25-27 kV and using helium and neon gas atoms as ions for irradiating CVD-grown graphene. Researchers observed the formation of various types of defects, including single-atom vacancy defects (monovacancies) and Stone-Wales defects (STW).

Xu [25] and the team also studied the impact of low-energy (less than 50 eV) ion irradiation on graphene. Monolayer graphene samples were irradiated with various ions, including boron, nitrogen, and fluorine, with different energies and fluences. Analysis of the samples before and after irradiation was performed using X-ray photoelectron spectroscopy (XPS), X-ray spectroscopy, Raman spectroscopy, and scanning electron microscopy [25].

The research revealed that low-energy ion irradiation can be successfully used to dope graphene with other atoms, but it also introduces damage and defects into the graphene [25]. The level of damage depends on the type of ion and fluence. Boron irradiation caused the least damage among the three types of ions considered, as this atom is the lightest. Nitrogen and fluorine irradiation at high fluences led to significant changes in the structure and properties of graphene, including its transformation into amorphous carbon [25].

Vazquez and the team proposed using this type of irradiation for graphene modification, specifically for creating nanopores, which could further be applied in filtration and sensing devices [21].

Ion irradiation causes serious damage to the two-dimensional structure of graphene, a conclusion supported by other researchers as well [24–27].

## 4 Impact of Electron Irradiation

Scanning electron microscopy and transmission electron microscopy are crucial tools for studying micro and nanoscale objects, making the investigation of electron irradiation's effects on graphene important due to potential damage to the studied samples. Additionally, this type of radiation can be employed to control graphene properties, such as adding functional groups, altering electronic structure, or manufacturing graphene devices through electron beam lithography (EBL) [28].

I. Childres and colleagues [29] presented data on the impact of electron irradiation on the electrical transport properties of monolayer graphene and the performance of devices based on this material. The irradiation resulted in a decrease in charge carrier mobility in graphene and the appearance of the D-peak in Raman spectroscopy, indicating defects induced by irradiation [29].

Studying the impact of electron irradiation on graphene devices revealed two main effects: a reduction in the charge-neutral point (CNP) for substrate-supported graphene and a decrease in graphene mobility with the appearance of the D-peak in Raman spectroscopy, suggesting irradiation-induced defects [29].

Similar conclusions were drawn by other researchers, including Yangbo Zhou and colleagues [30], who investigated mechanically exfoliated graphene layers irradiated with electron beams ranging from 1 keV to 30 keV. Irradiation doses were controlled by the beam current (from  $0 \pm 1$  pA at 2 keV to  $130 \pm 1$  pA at 30 keV). The study revealed that electron irradiation can lead to changes in the electrical characteristics of graphene devices, such as increased resistance and reduced charge carrier mobility. The effects of irradiation depended on electron energy and dose, with high-energy electrons and large doses causing significant changes in graphene properties [30].

Electron irradiation can also have a positive impact on the operation of graphene-based devices. Hossain and all [31] investigated the influence of electron irradiation on graphene and 1/f noise (flicker noise). The level of 1/f noise is an important indicator for assessing material suitability for practical devices in various fields. The authors demonstrated an interesting feature of 1/f noise in graphene: it decreases with an increase in the concentration of defects resulting from electron irradiation [31]. They found that bombarding graphene devices with 20 keV electrons can reduce the spectral noise density by an order of magnitude. The authors analyzed this noise reduction using mobility mechanisms and fluctuations in the number of charge carriers. The research results showed that electron irradiation introduces defects into graphene, confirmed by the appearance of disorder peaks in Raman spectroscopy. Interestingly, the noise level in graphene decreases after irradiation, contrary to the behavior of traditional materials where irradiation typically increases noise levels [31]. This noise reduction is explained by decreased mobility caused by defect introduction. In graphene, mobility is limited by long-range Coulomb scattering from charged defects, even at room temperature, unlike traditional materials where phonon scattering typically determines mobility limitations [31].

In conclusion, electron irradiation influences graphene, altering its electronic properties and structure. It is important to use scanning and transmission electron microscopes cautiously for graphene investigation and electron beam lithography (EBL) in the fabrication of graphene devices. Prolonged exposure can lead to a deterioration of graphene device properties, so optimal conditions should be adhered to for preserving its characteristics.

## 5 Impact of Temperature on Graphene

Understanding how temperature affects graphene and graphene-based devices is crucial for predicting and comprehending whether such devices can operate in high-temperature environments, such as nuclear fusion reactors.

Studies have shown that elevated temperatures can restore the properties of graphene and reduce defects that accumulate in samples during irradiation [6, 14, 25, 31]. The processing temperature in experiments was around 300°C. Performing several annealing cycles proves to be more effective than a single annealing process [6]. These findings indicate that graphene possesses self-healing properties for defects induced by irradiation through annealing.

## Conclusion

Summarizing the obtained results, it is noteworthy that all types of radiation have an impact on the structure of graphene, inducing defects in the material. It is evident that an increase in fluence and particle energy leads to a higher concentration of defects in graphene. An important conclusion is that graphene-based sensors, even in the presence of defects, continue to function effectively, demonstrating the resilience of graphene to radiation. This opens up possibilities for the application of graphene in high-radiation environments where traditional semiconductors may not be efficient.

Radiation exposure also allows for the modification of the graphene structure, creating pores and altering the interactions between graphene and substrates. A significant outcome is the reduction in the  $1/f$  noise level upon irradiation, in contrast to the increased noise observed in classical semiconductors. Post-irradiation material treatment contributes to defect reduction, indicating graphene's ability to compensate for them.

It is crucial to conduct more in-situ studies, particularly measuring sensor characteristics during irradiation, rather than solely before and after, to gain a more precise understanding of the impact of radiation on device functionality. Considering the unique properties and radiation resistance of graphene, this material stands out as an excellent candidate for applications in nuclear energy and space exploration.

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## Вплив гамма, нейтронного, іонного та електронного опромінення на структуру та властивості графену

Біляк Р. В.

У статті розглядаються дослідження впливу різних видів радіації, зокрема  $\gamma$ -проміння, нейтронного, іонного та електронного опромінення, на графен та пристрої на основі графену. Вивчення реакції графену на радіацію має вирішальне значення через його потенційне застосування в таких галузях, як ядерна енергетика та освоєння космосу, де вплив радіації є значним.

У статті обговорюються нещодавні експерименти, проведені для дослідження впливу  $\gamma$ -проміння на шари графену та пристрої на його основі, які виявили зміни у відстані між шарами графену, виникнення дефектів і зміну електричних характеристик.

Аналогічно, дослідження впливу нейтронного опромінення на графен демонструють його стійкість до такого проміння, причому сенсори на основі графену зберігають функціональність навіть після впливу високого нейтронного потоку. Більше того, дослідження іонного опромінення виявляють здатність модифікувати структуру графену, хоча воно і спричиняє значні пошкодження. Електронне опромінення утворює дефекти, що в свою чергу знизило рівень шуму в пристроях на основі графену, що є унікальною характеристикою, яка не спостерігається в традиційних матеріалах.

Також були зроблені висновки щодо впливу температури на графен, при цьому виявлено, що підвищені температури сприяють зменшенню дефектів шляхом відпалу, демонструючи властивості графену до самовідновлення.

На завершення, стаття підкреслює стійкість графену до радіації та його потенціал для використання у високорадіаційних середовищах, де традиційні матеріали можуть не витримати. Отримані результати свідчать про те, що сенсори і пристрої на основі графену можуть зберігати функціональність навіть за наявності дефектів, спричинених радіацією, що відкриває багатообіцяючі перспективи для застосування в ядерній енергетиці та космічних дослідженнях. Для кращого розуміння впливу радіації в реальному часі на функціональність пристроїв рекомендується проводити подальші натурні дослідження. Загалом, графен є чудовим кандидатом для різних застосувань завдяки своїм унікальним властивостям і радіаційній стійкості.

*Ключові слова:* графен;  $\gamma$ -опромінення; нейтронне опромінення; електронне опромінення; іонне опромінення; ефект самозаліковування; термоядерний синтез