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Synthesis and Characterization of β-Ga_2O_3/por-GaAs/mono-GaAs Heterostructures for Enhanced Portable Solar Cells

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This study comprehensively details the successful synthesis of a β-Ga_2O_3/por-GaAs/mono-GaAs heterostructure designed for portable solar cells. Employing a combination of electrochemical etching and high-temperature oxygen annealing, we engineered a heterostructure that exhibits both crystalline and amorphous phases. XRD, SEM, and Raman spectroscopy analyses confirmed the formation of crystalline β-Ga_2O_3 and GaAs, with the porosity in the GaAs layer enhancing light absorption and charge collection. The potential of the heterostructure to improve photovoltaic performance is attributed to the inherent stability of Ga_2O_3 and the increased surface area provided by the porous GaAs.

Keywords: β-Ga_2O_3, por-GaAs, mono-GaAs, heterostructure, solar cells, electrochemical etching, oxygen annealing, XRD, SEM, Raman spectroscopy.

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Introduction

Solar energy is foundational to sustainable electricity production, offering a clean alternative to fossil fuels with the potential for significant global carbon emissions reduction [1,2]. The versatility of solar energy is particularly evident in the development of portable devices, ranging from consumer electronics [3, 4] to remote sensing equipment [5, 6], which require reliable and efficient energy sources for operation in diverse environments.

Due to its abundant availability and well-established manufacturing processes, silicon has dominated the solar cell market [7 - 9]. Cadmium telluride (CdTe) has also been utilized, offering advantages such as lower material costs and higher absorption coefficients [10, 11]. However, efforts to enhance efficiency and versatility have led to the exploration of A3B5 semiconductors, such as InP [12 - 14] and GaAs [15 – 17]. These materials are known for their exceptional electronic properties and are actively used in photovoltaic technologies, demonstrating the evolving material landscape of the industry [18 - 20].

Recently, the focus has shifted to oxide materials, recognized for their stability and wide bandgap energy, making them suitable for high-efficiency solar cells [21 - 24]. Among them, β-Ga_2O_3 has garnered attention as a promising candidate for solar cell applications due to its wide bandgap that allows effective ultraviolet light absorption while remaining transparent to visible light [25, 26]. This characteristic is particularly advantageous for heterostructural solar cells, combining different materials to utilize their complementary properties, enhancing the overall performance [27, 28].

Developing heterostructural solar cells has been particularly intense, prompting efforts to achieve higher efficiency and greater flexibility in device design [29, 31]. These cells offer several advantages, including adapting the bandgap energy through combinations of materials and improving light absorption and conversion efficiency [32, 33]. Despite the potential, the creation of β-Ga_2O_3/GaAs

heterostructures has encountered challenges, mainly due to lattice mismatch between the constituent layers, which can lead to poor material quality and decreased device performance [34, 35]. Furthermore, the high cost of synthesis methods such as molecular beam epitaxy [36] and metal-organic chemical vapor deposition [37] has posed a significant barrier to the widespread application of these advanced materials in solar cell technology.

In response to these challenges, our study proposes an innovative approach to overcoming the limitations associated with lattice mismatch and high manufacturing costs. Adding an intermediate por-GaAs buffer layer effectively reduces the tension between layers, creating a more coherent and defect-free interface. Additionally, we employ a simple method of electrochemical etching followed by annealing in an oxygen atmosphere, offering a cost-effective and scalable solution for synthesizing β-Ga_2O_3/GaAs heterostructures. This method eliminates economic constraints and enhances the heterostructure's structural integrity, paving the way for its application in portable solar cells, where efficiency, durability, and affordability are paramount.

I. Materials and Methods

Materials

The primary material for forming Ga_2O_3 layers was monocrystalline semiconductor gallium arsenide (GaAs) plates, grown by the Czochralski method. These n-type, Sb-doped plates had a carrier concentration of $2.3x10^{18}$ cm^{-3} and were oriented in the (111) direction. Plates sized 10x20x2 mm were thoroughly polished to achieve a mirror finish. Before the experiment, they were cleaned using acetone, ethyl alcohol, and deionized water to ensure a contaminant-free surface for further etching and annealing.

Electrochemical Etching

The formation of the heterostructure began with electrochemical etching in acidic solutions using a standard three-electrode electrochemical cell, which included a chloroacetate reference electrode, a platinum cathode, and a gallium arsenide plate serving as the anode. A Teflon stirrer was used to remove bubbles from the sample surface. In the first stage of the experiment, a porous layer was created on the GaAs surface using a nitric acid solution (HNO_3:H_2O = 1:4), with the system subjected to a constant current (DC) at a voltage of 7 V. The total etching duration was 10 minutes.

In the second stage of the experiment, the solution was modified by introducing ethanol (C_2H_5OH:HNO_3:H_2O = 1:1:4), and the etching continued under a constant current voltage of 7 V for 10 minutes.

Oxygen Annealing

The final stage of the synthesis process included oxygen annealing in a JetFirst furnace, which was conducted at 650°C for 20 minutes. This stage aimed to ensure the crystallization of the formed layers and saturate them with oxygen.

Characterization

Morphological analysis was performed using scanning electron microscopy (SEM) using the SEO-SEM Inspect S50-B microscope. The phase state was analyzed through Raman spectroscopy using the RENISHAW inVia Reflex micro spectrometer.

To study the structural characteristics of the heterostructures, X-ray diffraction (XRD) spectroscopy on the Dron-3M diffractometer in the angle range of $2\theta = 10° – 60°$ was conducted.

II. Results

SEM Analysis

Figure 1 presents the surface morphology of the synthesized Ga_2O_3/por-GaAs/mono-GaAs heterostructure. The SEM image reveals a highly textured surface morphology characteristic of a porous structure. The field of view is dominated by a network of pores of various shapes and sizes, indicating an uneven etching process that leads to a non-uniform distribution of pores. The pores appear randomly distributed across the surface, with some regions showing a high density of closely packed pores while others appear more sparsely populated.

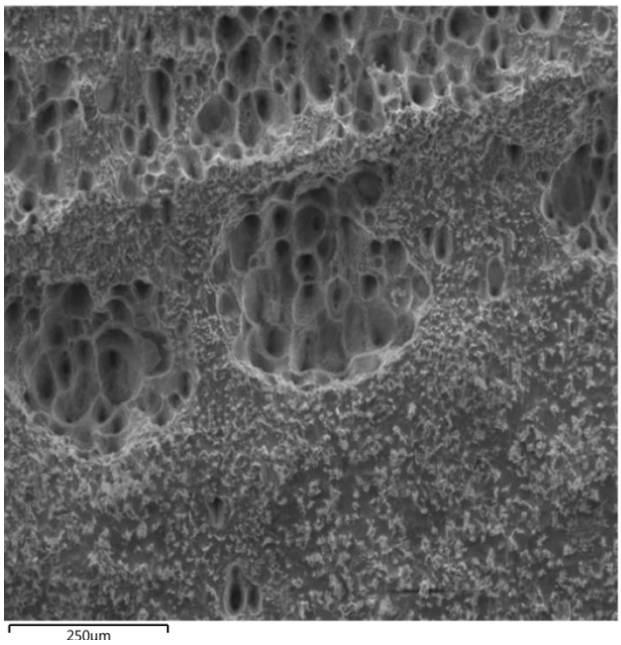

Fig. 1. SEM image of the β-Ga_2O_3/por-GaAs/mono-GaAs surface.

The pores have irregular shapes, ranging from nearly round to elongated and interconnected. The edges of the pores are not smooth but rather rough and jagged, indicating the intensive etching that underlies the formation of these features. Inside the pores are indications of substructures or smaller pores that could significantly affect the overall surface area.

The SEM analysis of the synthesized β-Ga_2O_3/por-GaAs/mono-GaAs heterostructure reveals a complex surface morphology characterized by various pore sizes and types. The surface is dominated by large, cone-shaped pores with diameters varying from 8 to 25 μm. These larger pores exhibit loose walls composed

of smaller pores, indicating a hierarchical porosity. This macroporous structure contributes significantly to the overall surface area, enhancing the heterostructure's light absorption capability and, consequently, its efficiency in solar cell applications.

Two distinct types of smaller pores are observed within the walls of these larger pores. There are smaller pores near the surface with diameters ranging from 1 to 1.5 μm. These mesopores are densely packed around the larger macropores, adding to the roughness and increasing the effective surface area of the GaAs layer. Closer to the bottom of the larger pores, an additional set of even smaller pores is evident, with diameters between 100 and 200 nm. These nanopores form a more compact network within the porous walls, further enhancing the material's surface area.

The interpore walls, which separate these various pores, have a thickness ranging from 70 to 100 nm. These walls consist of smaller crystallites and voids, creating a secondary level of mesoporosity within the material.

The areas between the pores are covered with a granular texture, which may indicate a secondary phase or residue from the etching solution. These granules vary in size and appear less ordered than the pores, adding complexity to the surface topology.

Raman Analysis

Figure 2 presents the Raman scattering spectrum of the β-Ga_2O_3/por-GaAs/mono-GaAs heterostructure.

The Raman peak at approximately 200 cm^{-1} is attributed to the longitudinal optical (LO) phonon mode of Ga_2O_3. In the Raman scattering spectrum, LO-phonon modes indicate lattice vibrations where atoms move parallel to the direction of phonon propagation. The LO phonon mode in Ga_2O_3 is typically strong due to the polar nature of the Ga-O bonds, leading to significant electro-optical interaction. The presence of this LO mode without any significant shift or broadening suggests that the Ga_2O_3 layer in the heterostructure is relatively free from strain and well-aligned with the underlying structure, which is crucial for the efficient integration of the Ga_2O_3 layer into the heterostructure without introducing additional scattering centers or trap states that could hinder charge carrier mobility.

The 267 cm^{-1} and 291 cm^{-1} peaks are associated with GaAs transverse optical (TO) and longitudinal optical (LO) phonon modes. The TO phonon mode, represented by the peak at 267 cm^{-1}, involves atomic vibrations perpendicular to the direction of propagation and is sensitive to the bonds and symmetry in the crystal lattice. In GaAs, this mode is usually clearly defined due to the covalent nature of the bond and the crystal symmetry of the zinc blend structure [38].

The electric field influences the LO phonon mode at 291 cm^{-1} due to the polar nature of the GaAs lattice. LO-phonon modes can interact with free carriers (Fröhlich interaction), which is significant in doped semiconductors and can affect the charge carrier dynamics [39].

The higher-order combination modes observed at 417 cm^{-1}, 640 cm^{-1}, and 760 cm^{-1} are associated with vibrational phenomena in the Ga_2O_3 part of the heterostructure.

The peak at 417 cm^{-1} can typically be attributed to the high-phonon E2 mode of Ga_2O_3. This mode is related to the vibrations of atoms in the crystal lattice, which are perpendicular to the c-axis of the material's crystal structure. This mode indicates a well-ordered lattice and is often used as a fingerprint for identifying the monoclinic β-phase of Ga_2O_3 [40].

The 640 cm^{-1} peak corresponds to the A1(TO) phonon mode, a transverse optical mode along the c-axis. This is a unique characteristic of β-Ga_2O_3, resulting from the asymmetric stretching of Ga–O bonds in the lattice. It is sensitive to stoichiometry and the presence of defects or impurities in the lattice, which can create local electric fields affecting the Raman shift of this mode.

The peak at 760 cm^{-1} is likely the A1(LO) Ga_2O_3 mode, involving longitudinal optical vibrations along the c-axis. This mode is particularly sensitive to lattice disorder and often broadens in the presence of defects or dislocations [41]. The appearance and intensity of this peak can provide information about the crystal quality and the presence of internal or external defects in the Ga_2O_3 layer.

The observed broadening of the Raman scattering peaks and noise throughout the spectrum can be attributed to the high porosity of the heterostructure. Porosity inherently introduces a significant degree of disorder at the microscopic level, manifesting as broadened Raman peaks. This is because the porous structure disrupts the uniformity of the crystal lattice, leading to changes in bond lengths and angles, which, in turn, affects the vibrational modes of the crystal.

In porous materials, the surface-to-volume ratio significantly increases, enhancing the Raman signal and contributing to peak broadening due to the increased likelihood of surface defects and irregularities. Additionally, the porous nature can lead to localized changes in strain and stress within the material, as the pores can act as stress concentrators. This variability in the local lattice environment is likely to cause the observed noise in the Raman spectrum, as different regions of the material contribute differently to the overall signal.

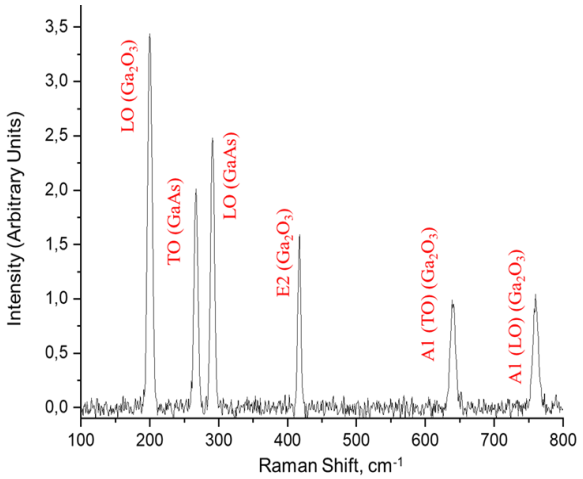

Fig. 2. – Raman spectrum of the β-Ga_2O_3/por-GaAs/mono-GaAs heterostructure.

XRD Analysis

The XRD spectrum of the Ga_2O_3/por-GaAs/mono-GaAs heterostructure demonstrates a combination of

crystalline and amorphous characteristics, indicating a complex interaction of different material phases within the system (Table 1, Figure 3). Crystalline peaks are identifiable at 2θ angles of 18.9°, 34.5°, 39.4°, and 55.0° can be attributed to the β-Ga_2O_3 phase, correlating with (010), (002), (102), and (110) planes, respectively (JCPDS Card No. 41-1103). These peaks confirm the crystalline presence of β-Ga_2O_3; however, the broader and less intense nature compared to those of single-crystal β-Ga_2O_3 suggests a partial crystal structure with a degree of disorder or defects within the crystal lattice. This phenomenon is likely a result of the integration process within the heterostructure, which can induce lattice distortion or defects across different material interfaces.

A broad background signal in the spectrum indicates the presence of amorphous regions within the heterostructure, distinguishing it from its monocrystalline counterparts. The amorphous phase contributes to the scattering of the non-crystalline background in the XRD spectrum, highlighting the material's structural complexity.

Peaks associated with GaAs are noticeable at 2θ angles around 27.3°, 45.5°, and 54.8°, corresponding to the (111), (220), and (311) planes (JCPDS Card No. 32-0389). The peak broadening, especially compared to monocrystalline GaAs, indicates a reduction in X-ray diffraction coherence due to the porous structure. Such broadening signifies variations in lattice parameters and micro deformations, characteristic of materials with high porosity.

Table 1.

X-ray Diffractometric Reflections Analysis of the β-Ga_2O_3/por-GaAs/mono-GaAs Heterostructure

Peak Position (2θ)	Plane (hkl)	Phase Identified
18.9°	(010)	β-Ga_2O_3
27.3°	(111)	GaAs
34.5°	(002)	β-Ga_2O_3
39.4°	(102)	β-Ga_2O_3
45.5°	(220)	GaAs
54.8°	(311)	GaAs
55.0°	(110)	β-Ga_2O_3

The crystallite size of the β-Ga_2O_3 layer was estimated using the Scherrer equation, which relates the full width at half maximum (FWHM) of the X-ray diffraction peaks to the size of the coherent scattering domains. The average crystallite size calculated from the FWHM of the XRD peaks is approximately 4.86 nm. This value provides insight into the material's crystalline quality and potential microstrains.

It is important to note that the crystallite size derived from the FWHM of the XRD spectrum does not correspond to the pore size or the size of the interpore spaces observed in the SEM analysis. Crystallites refer to the coherent scattering regions within a crystalline material, while pores are voids that do not contribute to the diffraction pattern.

In conclusion, the XRD analysis indicates that the β-Ga_2O_3/por-GaAs/mono-GaAs heterostructure comprises high-quality crystalline regions of both β-Ga_2O_3 and GaAs, as evidenced by the symmetrical peaks observed in the diffractogram. The presence of these symmetrical peaks, with a full width at half maximum (FWHM) in the range of approximately 3-4 degrees 2θ for both β-Ga_2O_3 and GaAs, suggests that the β-Ga_2O_3 layer is primarily polycrystalline with a minor interspersion of the amorphous phase. This polycrystalline nature is further supported by the broadening of the peaks, which indicates the presence of stresses and defects within the crystal lattice, as well as variations in crystallite size.

These structural characteristics, including the presence of amorphous regions and the observed polycrystalline nature, are critical for understanding the electronic and optical properties of the heterostructure. The porosity of the GaAs layer and the integration of the two materials contribute to the structural disorder, which can influence the charge transport and light absorption capabilities, directly impacting the performance and efficiency of the heterostructure when applied in solar cell technologies.

Therefore, optimizing the performance of heterostructural solar cells requires precise control over porosity and the minimization of lattice defects. This includes careful management of crystallite size and lattice stress to ensure a balanced combination of crystalline and amorphous phases tailored to enhance the heterostructure's efficiency and durability in photovoltaic applications.

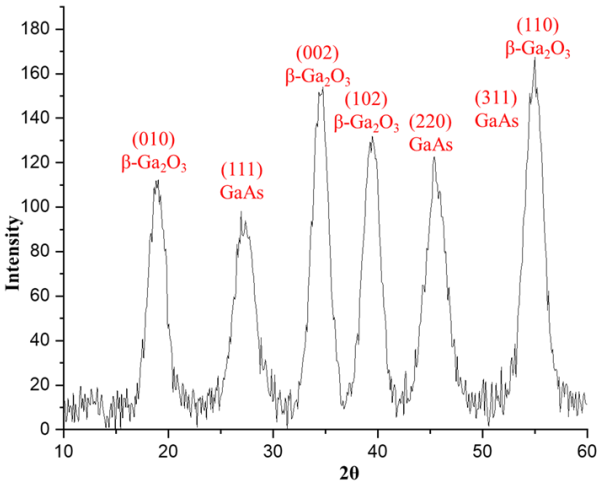

Fig. 3. - XRD spectrum of the β-Ga_2O_3/por-GaAs/mono-GaAs heterostructure.

III. Discussion

The formation mechanism of β-Ga_2O_3 under the specific synthesis conditions adopted in this study can be attributed to controlled etching and annealing processes. Initially, gallium and arsenic, constituents of GaAs, are etched from the crystal surface during the first stage, with arsenic being etched out faster [42]. This selective etching facilitates the formation of a porous structure in the GaAs layer, crucial for the subsequent formation of β-Ga_2O_3.

Following adding ethanol to the etching solution, a significant change in the chemical dynamics at the GaAs surface occurs. Ethanol acts as a mild organic solvent,

affecting the etching process by altering the local chemical environment. This potentially reduces the oxidation rate of gallium, thus allowing for a more controlled formation of the Ga_2O_3 layer. The presence of ethanol can also modify the surface chemistry, influencing the nucleation centers of Ga_2O_3 and promoting the formation of a more uniform layer.

Subsequent annealing in an oxygen atmosphere is vital as it leads to oxygen saturation and recrystallization of the structure. Any residual surface arsenic likely evaporates during this high-temperature annealing, leaving a purer Ga_2O_3 layer. This recrystallization stage is critically important for improving the crystalline quality of Ga_2O_3 as it helps heal defects and enhance the material's structural integrity.

For portable solar cells, the synthesized heterostructure offers several advantages. The porous GaAs layer provides a large surface area, facilitating better light absorption and, in turn, a more significant accumulation of photogenerated charge carriers. The β-Ga_2O_3 layer is a wide-bandgap material that allows for efficient absorption of ultraviolet light while being transparent to visible light, potentially leading to an extended solar cell spectral response. Moreover, using Ga_2O_3 enhances the element's stability, providing resistance to environmental factors critical for the longevity of portable devices.

The ability to synthesize such a complex structure through an economically efficient and scalable process aligns with the industry's pursuit of more sustainable and economically viable solutions for solar energy. Future work will likely focus on optimizing the material properties and device architecture to maximize conversion efficiency and pave the way for commercialization in the portable electronics market.

Conclusions

In this study, we fabricated a β-Ga_2O_3/por-GaAs/mono-GaAs heterostructure through electrochemical etching to create a porous GaAs layer followed by oxygen annealing to form the crystalline top layer of β-Ga_2O_3. X-ray diffraction (XRD) confirmed the heterostructure's structural integrity and crystalline quality were confirmed by X-ray diffraction (XRD), which identified characteristic crystalline peaks, and Raman scattering spectroscopy, which revealed specific phonon modes associated with Ga_2O_3 and GaAs. Scanning electron microscopy (SEM) was employed to verify the morphology and porosity of the layers. This heterostructure shows promise for portable solar cells due to its enhanced light absorption capabilities and stability provided by the Ga_2O_3 layer.

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Синтез та характеристика β-Ga_2O_3/por-GaAs/mono-GaAs гетероструктур для покращених портативних сонячних елементів

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У даному дослідженні детально описано успішний синтез β-Ga_2O_3/por-GaAs/mono-GaAs гетероструктури, розробленої для портативних сонячних елементів. Застосовуючи комбінацію електрохімічного травлення та високотемпературного окислення киснем, ми створили гетероструктуру, яка має як кристалічні, так і аморфні фази. Аналізи за допомогою рентгенівської дифракції (XRD), скануючої електронної мікроскопії (SEM) та раманівської спектроскопії підтвердили утворення кристалічних фаз β-Ga_2O_3 та GaAs, причому пористість у шарі GaAs підвищує поглинання світла та збір заряду. Потенціал гетероструктури для покращення фотогальванічних характеристик обумовлений властивою стабільністю Ga_2O_3 та збільшеною площею поверхні, забезпеченою пористим GaAs.

Ключові слова: β-Ga_2O_3, por-GaAs, mono-GaAs, гетероструктура, сонячні елементи, електрохімічне травлення, окислення киснем, XRD, SEM, раманівська спектроскопія.