Design and Comparative Study of Hybridized Monolayered Gallium Arsenide Based Bio-Sensors for Alzheimer 's disease Early Identification

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Abstract. Extensive research on alternative methods that may be helpful in replacing the conventional methods used for detection of Alzheimer's Disease. To cope up with dangerously increasing cases of dementia and alzheimer's disease worldwild for past some years, academia is actively involved in designing methods to detect this fatal disease well in advance. A family of III-V compound semiconductors Gallium Arsenide (GaAs) monolayer has a large band gap and honeycomb structure, making it an attractive options for sensing applications. In this work, the adsorption behavior of three molecules— Butylated hydroxytoluene , Pivalic acid, and Phenylenediamine on GaAs monolayers, which are known as Volatile Organic Compound (VOC) for detection of Alzheimer's disease, were examined using density functional theory (DFT). For every GaAs@gas system, the band structure, density of states (DOS), charge transfer (Q_T), adsorption energy (E_{ads}) and Recovery Time (τ) were calculated as part of the analysis. The band gap was reduced significantly after adsorption in the GaAs monolayer. Interestingly, Butylated hydroxytoluene displayed the highest adsorption energy on the GaAs monolayer out of all the gases examined. And also based on the displayed recovery time butylated hydroxytoluene may be suitable as a bio-sensor for Alzheimer's disease due to its comparatively lower recovery time. According to these results, the GaAs monolayer doped with these gas molecules may successfully function as a bio sensor to detect alzheimer's disease in its early stages.

Keywords: Bio Sensor, Alzheimer's Disease, Density Function Theory, two dimensional, density of states, adsorption energy, charge transfer

1 Introduction

About 60 to 70 percent of dementia cases worldwide are caused by Alzheimer's disease (AD) [1], a neurodegenerative illness that is debilitating and the most frequent type of dementia. The symptoms, which first manifest as trouble recalling recent events, gradually increase to include impaired language, confusion, mood swings, and a progressive loss of physiological functions that ultimately results in death. It primarily affects older persons. Amyloid plaques and neurofibrillary tangles build up in the brain, which stops neurons from working normally and causes damage that can't be fixed. It is projected that approximately 7.2 million Americans 65 and older— roughly one in nine in this age group—will have Alzheimer's dementia in the United States alone in 2025[10]. Given the severity of the issue, a number of nations have set a deadline for finding a treatment or possible cure to address AD[10]. The frequency is particularly high in people 75 years of old or above, and about two-thirds of those afflicted are female. The impact of the disease on mortality has significantly increased in recent decades; between 2000 and 2022, the death rate from Alzheimer's disease more than doubled, despite a minor drop in heart disease deaths. Alzheimer's places a significant strain on patients, carers, and healthcare systems. In 2024, unpaid carers will have provided over 19 billion hours of care, valued at over $413 billion. With healthcare and long-term care expenses expected to exceed $384 billion in 2025 and perhaps rising to about $1 trillion by 2050, the economic impact is substantial. Its genetic and environmental risk factors have been thoroughly studied, and new diagnostic and therapeutic approaches have been developed, but there is still no cure. The various treatments aim to control symptoms and reduce the progression of the illness. The number of people affected by Alzheimer's disease is predicted to increase dramatically from the current 50 million to 152 million by 2050[11], underscoring the urgent need for more research, better healthcare practices, and social support networks to address this growing public health concern. This introduction highlights the scope of Alzheimer's disease as a complex medical, financial, and social problem that necessitates coordinated interdisciplinary approaches.

Electrochemical biosensors have become a vital tool for early identification of Alzheimer's disease (AD) which is mainly diagnosed through the symptoms of cognitive decline. The need for early diagnostic methods is packed by the increasing prevalence of AD, which at present, is the cause of illness for a few millions of people worldwide and is predicted to be three times as much in the next 30 years[12]. The traditional ways of diagnosis which include imaging and cerebrospinal fluid (CSF) analysis are usually invasive, expensive, and lengthy, thus impractical for widespread use[12]. On the other hand, electrochemical biosensors provide a solution that is attractive owing to their features viz. high sensitivity, specificity, quick response times, and low cost of use[13]. These biosensors deploy nanomaterials like gold nanoparticles (AuNPs), carbon-based nanostructures, reduced graphene oxide (rGO) to facilitate electron transfer, enhance bioreceptor immobilization, and amplify the signal[12,13]. The biosensors have the capability to identify the specific biomarker sequences and distinguish them from the non-specific analogs that is how they ensure that the specificity is high[13,15]. Overall, electrochemical biosensors constitute a major breakthrough in moving towards point-of-care testing for Alzheimer’s disease and thus, the implementation of

earlier intervention strategies and better therapeutic regimens become possible besides the reduction of healthcare expenses and diagnostic burdens[13,14].

The insufficiencies of presently utilized biosensors for detecting Alzheimer's disease in its early stage via the identification of volatile organic compounds (VOCs) encompass problems with sensitivity and specificity since the concentration of VOCs in the exhaled breath or other bio fluids can be extremely low and can be affected by various external factors[14]. Moreover, the accuracy and repeatability of VOC-based biosensors are usually restricted by environmental factors and biological variability among patients. These biosensors also have problems with clinical validation, regulatory approval, and standardization, which gradually approach cessation of their use in clinical settings. Besides that, when biosensors are combined with digital health technologies, there are privacy and data security issues, and ethical considerations need to be resolved before their widespread use[15]. In general, the restrictions still pose a barrier to their current effectiveness and practical use in the early detection of Alzheimer's, albeit they are potentially non-invasive and could be convenient.

Gallium Arsenide (GaAs) is widely recognized as a better biosensor material because it has outstanding electronic properties, for example, it has very high electron mobility (about 7467 $cm^2/V•s$), which allows it to be ultra-sensitive and very rapid in detecting. Biosensors based on GaAs, in particular those with high-electron-mobility transistors (HEMT), are free from nanomaterial modifications and external reference electrodes, thus, the operational procedures are simplified and the reproducibility is enhanced. Moreover, they can be processed by traditional semiconductor manufacturing methods, thus, they can be easily scaled up and integrated into point- of-care diagnostic platforms. In brief, GaAs biosensors can be very sensitive, fast, and handy for the detection of disease biomarkers at an early stage when compared to other biosensor materials.

The band gap and density of states properties of a 2D GaAs (Gallium Arsenide) biosensor structure for early detection of Alzheimer's disease [2] are used to increase the sensitivity as well as specificity. The band gap of GaAs is the one that allows the generation of electron-hole pairs efficiently under small perturbations, while the density of states specially prepared for the 2D structure thus facilitates the improved charge carrier dynamics and interaction with Alzheimer's biomarkers, making electrochemical or electronic detection at very low concentrations possible. Such a combination in 2D GaAs biosensors makes it possible to perform rapid, label-free, and highly sensitive detections which are indispensable for early Alzheimer's diagnosis.

In the realm of sensors [3], hybridized Gallium Arsenide based devices have demonstrated high sensor responses towards gas molecules ranging from small-sized toxic air molecules to large-sized volatile organic compounds (VOCs) molecules, related to Alzheimer’s diseases (AD) [3]. The identification and validation of such VOCs biomarkers hold significant importance in advancing the ability to diagnose AD in its early stages, allowing for timely interventions and planning appropriate prognosis [3].

2 Computational Details

In this work, we use first principles density functional theory calculations to investigate the electronic properties of GaAs 2D Van der Waals heterostructure [4]. Material selection is predicated on the hexagonal symmetry of the monolayers, which reduces the interface's lattice mismatch. It is very crucial in determining if an interface configuration is energetically favorable. To perform the simulations [26] we used QuantumWise software. The exchange and correlation functionals were treated in the PerdewZunger (LDA) generalized Meta gradient approximation, with a fundamental function TB09LDA. PulayMixer algorithm is used. We employed a density mesh cutoff of 105 Hartree to describe the plane waves which are incorporated into the basis set. A Gammacentered $1 \times 3 \times 3$ k-mesh is employed to assess the electronic characteristics of the interface.. Gaussian smearing is used with a smearing width of 0.05 eV [4]. The energy convergence threshold to achieve self- consistency was set to 10^6 eV [4]. All crystal structures were relaxed until the forces on atoms are less than 0.0001 eV/A [4].

Three gas molecules which are adsorbed on substrate material have following adsorption energy formula:

$$E_{ads} = E_{GaAs@gas} - E_{GaAs} - E_{gas} \quad (1)$$

The term " E_{ads} " refers to the energy that is produced when gas molecules act on surface of substrate material. The symbols $E_{GaAs@gas}$, E_{GaAs} , and E_{gas} represent the overall system energy following gas adsorption, gas molecules' energy value as well as the overall energy of the substrate material after optimization, in that order [5]. A process that is spontaneous, exothermic, and energy-positive [5] is indicated by a negative adsorption energy value ($E_{ads} < 0$), In contrast, an $E_{ads} > 0$ denotes an energy-consuming and endothermic process.

The band gap can be calculated as the difference between the bottom valence band and the highest conduction band.

$$E_g = V_{V.B.} - V_{C.B.} \quad (2)$$

Furthermore, Eq. (2) defines the change transmission between the gas molecules and substrate material.

$$Q_T = Q_a - Q_b \quad (3)$$

Where Q_a and Q_b represent the net charge carried by gas molecule that was separated and the charges quantity that are carried by the gas upon adsorption, respectively. The charge exchange from molecules of gas to the substrate is represented by the positive value of Q_T .

The following equation [6] can be used to calculate the recovery time by the transition state theory and Van't Hoff–Arrhenius expression:

$$\tau = \frac{1}{A \exp^{(E_{a/K_B T})}} \tag{4}$$

Where (E_a) is the energy required to break a chemical bond for desorption, and it is thought to be equal to (E_{ad}), and T represents temperature, (K_B) represents Boltzmann constant which is 1.38×10^{-23} $m^2kgs^{-2}K^{-1}$. As a result, the attempt frequency A is frequently thought of as a typical value that falls in range of 10^{12}–10^{13} s^{-1} [7], which is taken to be the range according to transition state theory.

3 Discussions and Results

3.1 Hybridized GaAs structural properties:

We have employed a GaAs cell with 24 Ga, 24 As and 12 H atoms to assess the adsorption performance of the pure GaAs monolayer, as seen in Fig. 1(a). The structure's lattice parameters are a = 4 Å, b = 12 Å, and c = 37.62 Å. The GaAs bond length in a low buckling honeycomb monolayer structure is 2.45 Å. With a 0.44 eV band gap. These findings are consistent with earlier research [26]. To account for additional adsorption scenarios various molecular to the GaAs monolayer, as demonstrated in Fig. 2. The optimized supercell configuration of the hybridized GaAs single layer is depicted in Fig. 1(a).

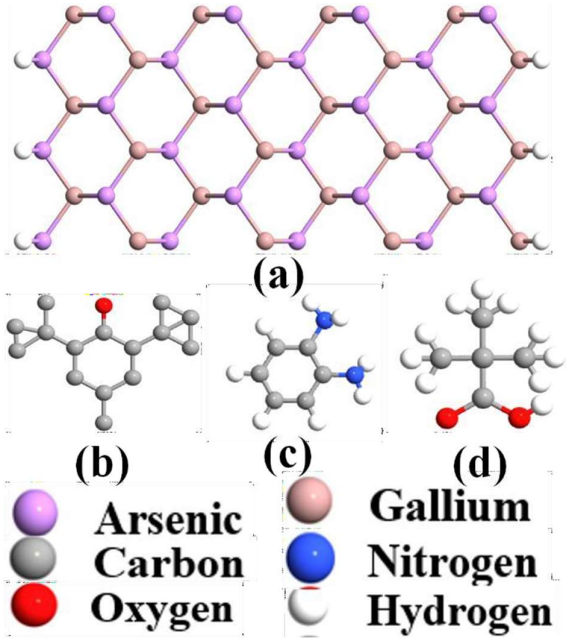


Fig. 1. (a) 2-D Hybridized GaAs monolayer, (b) Butylated hydroxytoluene($C_{15}H_{24}O$), (c) Phenylenediamine($C_6H_8N_2$), (d) Pivalic acid($C_5H_{10}O_2$)2

3.2 Butylated hydroxytoluene($C_{15}H_{24}O$) molecule adsorption on GaAs monolayer

$C_{15}H_{24}O$ adsorption on the hybridized GaAs monolayer is depicted in Fig. 2(a). The adsorption parameter is tabulated into Table 1. The adsorption energy of the $C_{15}H_{24}O$ doped on GaAs monolayer is obtained to be -0.17 eV. The gas of Butylated hydroxytoluene ($C_{15}H_{24}O$) is adsorbed to Gallium Arsenide (GaAs) molecule. After adsorption, two bonds are generated: the Arsenic–carbon (As–C) bond (2.08Å) and the Gallium-carbon (Ga-C) bond (2.07Å). The transfer of charge of +0.146 e from $GaAs$ to $C_{15}H_{24}O$, is also seen in Table 1. So, $C_{15}H_{24}O$ on the surface of the GaAs sheet acts as donor of electrons.

$$Ga_{24}As_{24} + C_{15}H_{24}O \rightarrow Ga_{24}As_{24}@C_{15}H_{24}O + E_{ad}(-0.17eV) \quad (5)$$

3.3 Pivalic acid($C_5H_{10}O_2$) molecule adsorption on GaAs monolayer

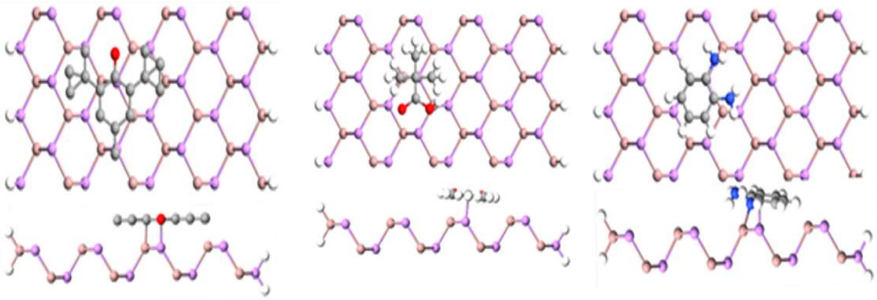
Fig. 2(b) illustrates the adsorption of $C_5H_{10}O_2$ on the hybridized GaAs surface. Table 1 contains a list of the adsorptive parameter. The $C_5H_{10}O_2$ doped GaAs system's adsorption energy has been determined to be -0.81eV. Furthermore, After adsorption, one bond is generated: the Arsenic–carbon (As–C) bond (2.07Å). The transfer of charge of -0.028 e from $C_5H_{10}O_2$ to $GaAs$, is also seen in Table 1. So, $C_5H_{10}O_2$ on the exterior surface of the GaAs sheet acts as acceptor of electrons.

$$Ga_{24}As_{24} + C_5H_{10}O_2 \rightarrow Ga_{24}As_{24}@C_5H_{10}O_2 + E_{ad}(-0.81eV) \quad (6)$$

3.4 Phenylenediamine ($C_6H_8N_2$) molecule adsorption on GaAs monolayer

Fig. 2(c) illustrates the adsorption of $C_6H_8N_2$ on the hybridized GaAs surface. Table 1 contains a list of the adsorptive parameter. The $C_6H_8N_2$ doped GaAs system's adsorption energy has been determined to be -0.71eV. After adsorption, two bonds are generated: the Arsenic–carbon (As–C) bond (1.84Å) and the Gallium- nitrogen (Ga-N) bond (1.95 Å). The transfer of charge of -0.24 e from $C_6H_8N_2$ to $GaAs$, is also seen in Table 1. So, $C_6H_8N_2$ on the surface of the GaAs sheet acts as an acceptor of electrons

$$Ga_{24}As_{24} + C_6H_8N_2 \rightarrow Ga_{24}As_{24}@C_6H_8N_2 + E_{ad}(-0.71eV) \quad (7)$$



(a) (b) (c)

Fig. 2. 2-D structure of (a) $Ga_{24}As_{24}@C_{15}H_{24}O$, (b)$Ga_{24}As_{24}@C_5H_{10}O_2$ (c) $Ga_{24}As_{24}@C_6H_8N_2$

4 Properties of the adsorbent system's electrochemistry

4.1 Band Structure Study

The band configuration of the 2-D GaAs single layer exhibits a direct band gap of 1.4 eV, as displayed in Fig. 3(a). Following the adsorption of $C_{15}H_{24}O$, the band gap decreases to 1.26 eV, as presented in Fig. 3(b). This reduction brings the valence band close to Fermi level, consequently in p-type semiconducting behavior. Upon the adsorption of $C_5H_{10}O_2$, the band gap shrinks slightly to 0.75 eV, as depicted in Fig. 3(c), due to the conduction band moving closer to Fermi level, leading to n-type semiconductor characteristics. The adsorption of $C_6H_8N_2$ also reduces the band gap to 0.94 eV, as displayed in Fig. 3(d), indicating additional changes of the electronic structure. These findings suggest that the adsorption of $C_{15}H_{24}O$, $C_5H_{10}O_2$, and $C_6H_8N_2$ substantially modifies the electronic structure of the 2-D GaAs monolayer, enabling tailored modifications to its semiconducting properties.

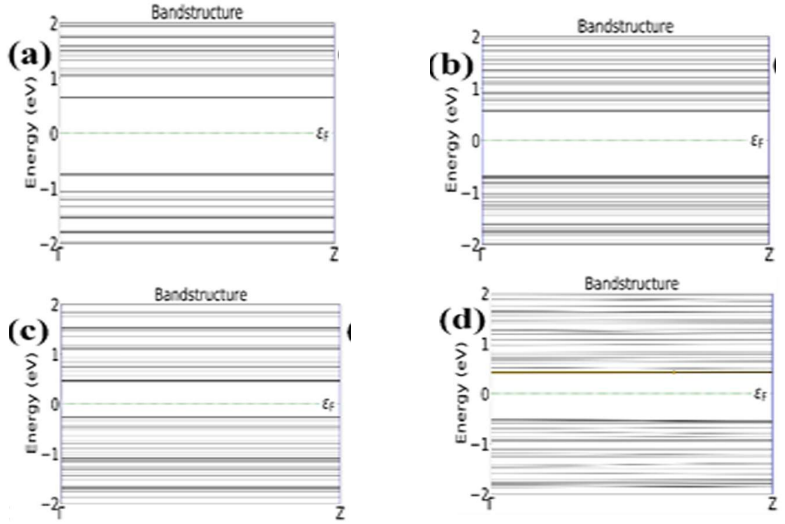


Fig. 3. Band structure of (a) GaAs, (b) $Ga_{24}As_{24}@C_{15}H_{24}O$ (c)) $Ga_{24}As_{24}@C_5H_{10}O_2$ (d) $Ga_{24}As_{24}@C_6H_8N_2$

4.2 Density of states

Fig.4 (a to d) illustrates the density of states (DOS) plotting of GaAs, $Ga_{24}As_{24}@C_{15}H_{24}O$, $Ga_{24}As_{24}@C_5H_{10}O_2$ and $Ga_{24}As_{24}@C_6H_8N_2$ respectively. In Fig. 4 (a), the DOS for GaAs shows a notable absence of state of energy in the range of -0.75 eV to 0.65 eV. In Fig. 4 (b), the $Ga_{24}As_{24}@C_{15}H_{24}O$ configuration exhibits a gap in the DOS between -0.695eV and 0.568 eV. Similarly, Fig. 4 (c) reveals that $Ga_{24}As_{24}@C_5H_{10}O_2$ lacks DOS energy states in the range of -0.276 eV to 0.479 eV. Fig.4 (d) indicates that for $Ga_{24}As_{24}@C_6H_8N_2$, there are no DOS energy states between -0.524 eV and 0.42 eV. These findings confirm successful adsorption of GaAs, $Ga_{24}As_{24}@C_{15}H_{24}O$, $Ga_{24}As_{24}@C_5H_{10}O_2$ and $Ga_{24}As_{24}@C_6H_8N_2$ gases, with the GaAs single layer exhibiting heightened sensitivity to $C_5H_{10}O_2$, there by indicating its potential effectiveness as a bio sensor for Alzheimer's disease.

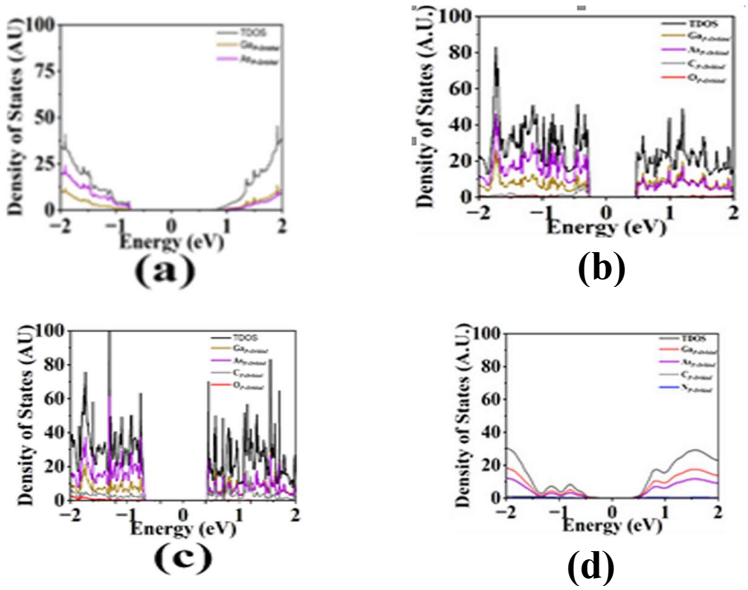


Fig. 4. DOS plot of (a) GaAs, (b) $Ga_{24}As_{24}@C_{15}H_{24}O$ (c) $Ga_{24}As_{24}@C_5H_{10}O_2$ (d)$Ga_{24}As_{24}@C_6H_8N_2$

Table I:Comparision Table

Adsorption System	E_{ads} (*eV*)	E_g (*eV*)	*d*(Å)	$Q_T(e)$	Style	Recovery Time (S)
$Ga_{24}As_{24}@C_{15}H_{24}O$	-0.17	1.26	As-C (2.08), Ga-C (2.07), As-C (2.08)	0.146	Donor	6.9122×10^{-10}
$Ga_{24}As_{24}@C_5H_{10}O_2$	-0.81	0.75	As-C (2.07)	-0.028	Acceptor	33.88
$Ga_{24}As_{24}@C_6H_8N_2$	-0.71	0.94	As-C (1.84), (Ga-N) 1.95	-0.24	Acceptor	0.7237

Where, Q_T stands for charge transfer, E_g for energy band gap, and E_{ads} for adsorption energy. For GaAs monolayer E_g=1.4 eV.

4.3 Reusability Analysis of GaAs@Gas

The reusable nature of the bio-sensitive material is necessary to consider when using bio sensors in practical uses. Consequently, it is essential to compute as well as analyze the recovery time. The recovery time is typically determined using equation [4]

$$\tau = \upsilon^{-1} \exp\left(\frac{-E_{ads}}{K_B T}\right) \tag{8}$$

Where, E_a, which is said to be equivalent in value E_{ads} to, is the amount of adsorption energy that adsorption must overcome ϑ is the attempt frequency (10^{12} s^{-1}), τ is the absolute temperature, and K_B is equals to the Boltzmann constant. The recovery time calculated at the room temperature (298K). Based on Fig. 5 and Table 1, Clearly, the GaAs monolayer demonstrates excellent potential for trapping Pivalic acid, as these gases exhibit prolonged retention times, indicating a high recovery time. Conversely, GaAs doped with $C_{15}H_{24}O$ may be suitable as a bio-sensor for Alzheimer's disease due to its comparatively lower recovery time. In the previous study, Titanium carbide MXenes used to detect the butylated hydroxytolune showed a very long recovery period of about 2.35×10^13s[16]. In this present study when Gallium arsenide (GaAs) monolayer is used to detect the same butylated hydroxytolune in this work to investigate Alzheimer's biomarkers, the recovery time is significantly reduced to 0.69 ns, indicating a considerable improvement in sensor performance.

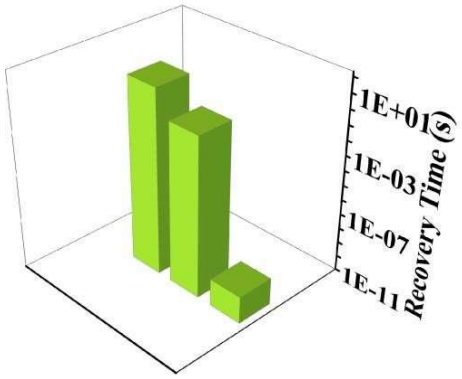


Fig. 5. Recovery time comparison of GaAs monolayer to(left to right) $C_5H_{10}O_2$, $C_6H_8N_2$, $C_{15}H_{24}O$

5 Conclusion

The implementation of pivalic acid, phenylenediamine, and butylated hydroxytoluene doped hybridized GaAs monolayers is a viable technique for the invention of ultra-sensitive biosensors targeting Alzheimer's disease detection. According to density functional theory (DFT) computations, substantial changes in electronic properties of GaAs monolayers, mainly in the band gap as well as density of states (DOS), have been reported. In this way, the organic molecules that are used for doping act as a medium for effectively tuning the band gap, thus, the sensitivity and selectivity of the material with respect to biomolecular interactions pertinent to Alzheimer's biomarkers are considerably increased. The modified electronic structure through the designed mechanism enables the transfer of charges as well as the transduction of the signal, which are factors leading to the accomplishment of the correct detection at the molecular level. Besides that, the changes in DOS also represent the improvement of surface reactivity and the increase of binding affinity, which are the main requirements addressed for the stability of biosensing performance.

The hybridized monolayers constitute a stable, reproducible, and scalable platform that harmonizes chemical specificity and excellent electronic properties. The theoretical argument here serves as a signal that such doped GaAs sensors have an incredible potential in the early-stage diagnosis of diseases and, moreover, help in developing biosensing technologies based on nanomaterials.

Next experimental trials and optimization would be the steps towards portable, non-invasive, and inexpensive devices for the monitoring of Alzheimer’s disease progression. This, in turn, would be assisting in the enhancement of patient outcomes by facilitated timely intervention.

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