



Structural, optical and plasmonic sensing characteristics of graphene quantum dots/gold nanolayered film in contact with dopamine solution

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Abstract

Graphene quantum dots (GQDs) have captured a considerable attention in biomedical field due to their unique structure-related properties. In this work, GQDs monolayer film was coated on gold thin film and integrated into surface plasmon resonance spectroscopy (SPR). The plasmonic sensing properties of GQDs/Au nanostructured layer in contact with varied concentrations of dopamine (DA) solution were evaluated. Increasing DA concentrations increased the changes in the resonance angle. This sensing platform showed a good sensitivity of $0.332^\circ/\text{nM}$ throughout a linear range of 0.01–100 nM, as well as a high binding affinity of $1.610 \times 10^9 \text{ M}^{-1}$. The optical properties of GQDs film were precisely determined by fitting the experimental curves to theoretical data formula using the transfer matrix method (TMM). The fitting results showed that the n value of the GQDs film was 1.3049 and its thickness was 7.22 nm in the absence of DA solution. The binding of DA to the SPR chip, as evidenced by the structural analysis of the film using FTIR and AFM, increased the n value and thickness of the GQDs film. These findings revealed the obvious changes in the structural and optical characteristics of this GQDs film after interaction with DA, and confirmed the potential of this material in DA sensing when combined with SPR spectroscopy.

Keyword Graphene quantum dots · Neurotransmitters · Surface plasmon resonance · Refractive index sensor · Sensitivity enhancement

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1 Introduction

Recently, several studies have revealed that graphene quantum dots (GQDs) films and/or coatings have prospective uses in biomedical (Liu et al. 2017; Qian et al. 2014; Xiao et al. 2016; Li et al. 2017; Zhu et al. 2012a), optical (Zubair et al. 2015; Kim and Kim 2017; Zhang et al. 2018; Tang et al. 2013; Das et al. 2015; Zhu et al. 2012b), and energy applications (Sudhagar et al. 2016; Zhu et al. 2014; Yan et al. 2010a; Majumder et al. 2016; Moon et al. 2017; Protich et al. 2016; Diao et al. 2017), which will influence our quality of life and draw substantial economic interest. The exciton Bohr radius of graphene is infinite (Yan et al. 2010b). GQDs, on the other hand, is a zero-dimensional material obtained by converting two-dimensional graphene. As a result, the quantum confinement and edge effects appeared. Because of the quantum confinement effect, GQDs have several unique features, such as their distinctive fluorescence capabilities found by Pan et al. (2010). If GQDs are to be employed in a variety of applications, the ability to adjust their characteristics is critical. Moreover, GQDs have a high solubility. This is because GQDs have a significant edge effect that may be modified by functional groups. Additionally, GQDs show different chemical and physical characteristics when compared to other carbon-based materials, such as carbon dots, carbon nanotubes, fullerene and graphene (Tian et al. 2018). Along with the structural properties of GQDs thin films, it is critical to precisely characterize the optical properties and thicknesses of GQDs films, on which their appealing qualities depend for their many applications (Sandu 2012; Majhi and Kuri 2020). Thus far, several approaches have been proposed for this purpose, including laser feedback interferometry (Xu et al. 2014, 2015), ellipsometry (McCrackin et al. 1963; Elizalde et al. 1986; Pristinski et al. 2006), prism coupler (Kirsch 1981; Hou and Mogab 1981; Ding and Garmire 1983), and surface plasmon resonance (SPR) technique (Fen et al. 2011; Rosso et al. 2014; Salvi and Barchiesi 2014; Kamal Eddin et al. 2022a, 2023a; Noda and Hayakawa 2016).

SPR spectroscopy has received significant attention and demonstrated effectiveness as an optical, label-free, and high throughput technique due to its potential for real time detection of heavy metal ions (Lopes et al. 2021; Fen et al. 2013, 2012, 2015; Fen and Yunus 2013a; Fauzi et al. 2020; Ramdzan et al. 2020), glucose (Omidniaee et al. 2022; Rosddi et al. 2021; Panda et al. 2020; Yuan et al. 2018; Kim et al. 2021; Hossain and Talukder 2021; Hakami et al. 2021), DNA (Pal et al. 2018; Haque and Rouf 2021; Shushama et al. 2017; Schneider et al. 2013; Kumar et al. 2019; Azab et al. 2018), hemoglobin (Singh et al. 2021; Mostufa et al. 2021; Sumantri et al. 2020; Mohanty and Sahoo 2016; Heidarzadeh 2020; Duanghathaiornsuk et al. 2020), neurotransmitters (Kamal Eddin et al. 2021, 2022b, c, 2023b; Dutta et al. 2011; Abd Manaf et al. 2017; Yuan et al. 2019), viruses (Omar et al. 2020, 2019; Omar and Fen 2017; Brun et al. 2015; Chang et al. 2018; Cairns et al. 2019; Chung et al. 2005), gases (Nuryadi and Mayasari 2016; Wei et al. 2016; Srivastava et al. 2016), and other targets (Kamal Eddin et al. 2020; García-Aljaro et al. 2008; Verma et al. 2015; Kamaliev et al. 2016) with good reliability and high performance. SPR phenomenon is the oscillation of the charge density at the interface of a metal film and a dielectric (Mao et al. 2015; Maurya et al. 2015; Elmahdy et al. 2022; Singh and Prajapati 2019; Li and Chen 2013; Haiwei et al. 2016; Islam et al. 2021). The high sensitivity of SPR spectroscopy to the boundary conditions enables it to detect the small changes in the medium refractive index induced after the adsorption of the target molecules on the surface of the active layer (Hong et al. 2015; Mukhtar et al. 2016; Xia et al. 2019; Kuo and Chang 2011; Kumar et al. 2021; Elsayed et al. 2017; Zhou et al. 2011). Due to the need to develop the SPR

technique itself, employing the surface plasmons to measure the optical properties as well as the thickness of thin films has gained considerable interest (Bruijn et al. 1990; Hoffmann et al. 1996; Kapoor et al. 2019; Yang et al. 2021; Nur et al. 2019; Kim et al. 2018). Because the reflected light carries information about the used film, the optical properties and thickness of the thin film could only be determined indirectly by mathematical processing of the experimental data (Kamal Eddin et al. 2022a, 2023a; Daniyal et al. 2022; Meradi et al. 2022). Wave propagation in one-dimensional structures may be studied using the transfer matrix method (TMM), which is based on Fresnel's theory. It allows for reflection and transmission computations as well as guided mode evaluations in multilayered systems. TMM treats Fresnel reflection and transmission at the interface of two media as one matrix and light propagation in a particular medium as another. This method provides information on electromagnetic wave propagation through ideal multilayer structures by multiplying matrices (Balili 2012; Tiwari et al. 2015; Chiu et al. 2007; Mudgal et al. 2020; Pandey 2021; Nisha et al. 2019).

In this work, a GQDs/Au nanostructured layer was integrated to SPR spectroscopy to interact with different concentrations of the neurotransmitter dopamine (DA). This was possible due to GQDs' exceptional chemical stability, biocompatibility, and low toxicity, as well as their graphene-like properties, including a substantial surface area and strong surface bonding, making them excellent for diverse biosensing applications (Duhan and Obrain 2023). Furthermore, SPR provides sensitive, real-time, label-free detection of DA. Additionally, unlike electrochemical methods, SPR is less susceptible to interference from other electroactive species. Moreover, it avoids electrode fouling, a common issue that can significantly impact the performance and reliability of electrochemical DA sensing (Kamal Eddin et al. 2022b). This study primarily focused on evaluating the sensor's performance. In addition, the experimentally acquired SPR curves were then computationally processed to analyze the optical properties of the GQDs/Au bilayer structure and determine the thickness of the GQDs film. The reported studies on DA sensors did not investigate DA binding behaviour on the sensor surface using structural measurements. So, the structural analysis of the sensor film prior to and following DA injection was achieved utilizing FTIR spectroscopy and atomic force microscopy (AFM), which confirmed the attachment of DA to GQDs/Au nanostructured layer.

2 Materials and methods

2.1 Materials and reagents

Graphene quantum dots (GQDs) with concentration of 1 mg/mL, and dopamine hydrochloride with molecular weight of 189.64 g/mol were obtained from Sigma-Aldrich. The glass cover slips of 24 × 24 mm with thickness between 0.13 and 0.16 mm and the triangular prism (refractive index of 1.77861) were provided by Menzel-Glaser, Germany. Norland index matching liquid (IML) with refractive index of 1.52 at 589 nm and low viscosity was bought from Norland (USA). This liquid monomer was used to eliminate the reflection losses associated with the glass-air interface. Acetone was used to thoroughly clean the prism and cover slips, assuring that their surfaces were not contaminated and that no leftover adsorbents that may affect the accuracy of the measurements. Throughout experiments, the deionized water (DW) was utilized for dilution.

2.2 Preparation of target solution

To produce 0.5 M of DA solution, 1.896 g of DA powder were dissolved in 20 mL of DW. To dilute DA solution, DW was used to obtain several samples with various low concentrations based on this formula ($M_1V_1 = M_2V_2$).

2.3 Chip modification

The glass cover slip was cleaned with acetone before coating gold thin film on its surface utilizing a K575X sputter coater from Quorum Technologies Ltd (West Sussex, UK). The duration of coating was 67 s using an applied current 20 mA and voltage 2.2 kV. After getting the gold thin films, 0.5 mL of GQDs was distributed evenly on the centre of the gold film's surface. The sensor film (GQDs/Au) was then deposited at high angular velocity of 2000 rpm using spin coating technique (P-6708D). The spin time was 30 s. The prepared GQDs/Au bilayer thin film was left for few hours at room temperature before its incorporation to SPR system.

2.4 Experimental procedure

The sensing performance of GQDs/Au bilayer film towards DA was examined and assessed utilizing a homemade prism based SPR spectroscopy designed in Kretschmann configuration as shown in Fig. 1. The angular interrogation technique was used, where the optical system included a 5 mW He-Ne laser (632.8 nm) with spot diameter of 0.8 mm was employed as excitation source, a light chopper with frequency of 188 Hz, a linear polarizer,

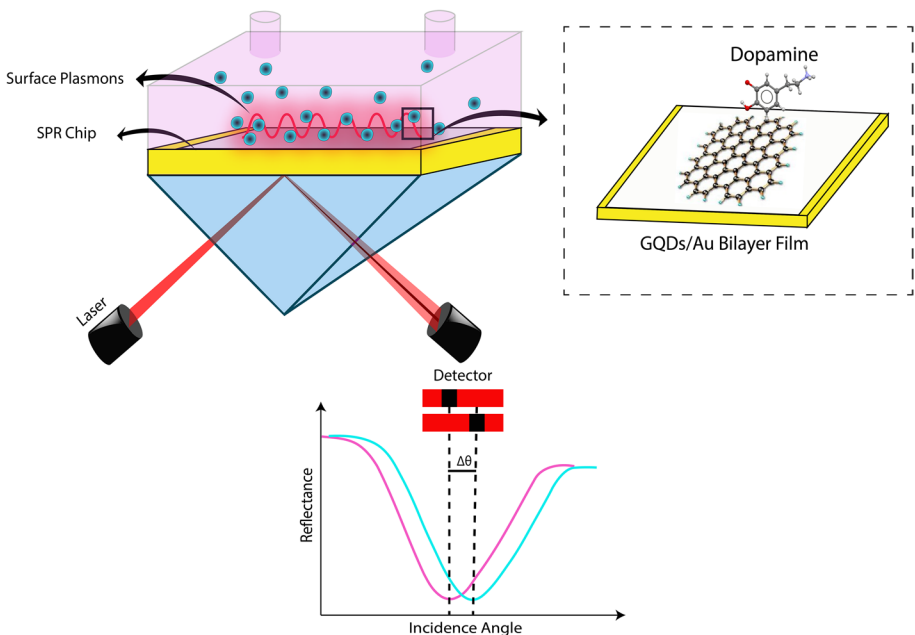


Fig. 1 SPR spectroscopy in Kretschmann configuration

a small pinhole, a prism (triangular with a refractive index of 1.77861), an optical rotating platform powered by a motion controller with a resolution of 0.001° (Newport model MM 3000), a photodetector, as well as a lock-in amplifier. The SPR chips were adherent to the prism side by the index matching liquid and a flow cell containing the target solution contacted the surface of SPR chip. Following that, SPR experiments were performed in the dark. DW was injected into the attached cell to contact the GQDs/Au bilayer film structure and obtain the reference signal. The incidence angles were scanned and the reflectance was measured as a function of incidence angle. As the incidence angle increased to reach the critical angle, the total internal reflection occurred, and the intensity of the reflected light at the interface was around 100%. As the angle increased further, surface plasmons were generated at the interface and the reflected intensity was therefore dropped. The intensity of reflected light from the film surface reached a minimum at the resonance angle. After that, SPR measurements were continued for DA solution of different concentrations.

2.5 Structural analysis techniques

FTIR spectra of GQDs/Au thin film were obtained in the range $400\text{--}4000\text{ cm}^{-1}$ utilizing ALPHA II FTIR Spectrometer before and after interactions with DA solution. The FTIR analysis was performed in ATR mode. The topographical measurements of the thin films and the analysis of roughness changes of GQDs films after interaction with DA were done using a Bruker Dimension Edge AFM with $5\text{ }\mu\text{m}\times 5\text{ }\mu\text{m}$ scanning size. The Peak Force Tapping mode was used with AFM tip's radius of curvature $< 10\text{ nm}$.

3 Result and discussion

3.1 FTIR analysis

FTIR spectrum of GQDs thin film before interaction with DA is shown in Fig. 2 (black spectrum). The peaks appeared at 3848 and 3742 cm^{-1} are attributed to O–H stretching vibration. The peak at 3116 cm^{-1} was attributed to the stretching vibration of O–H and N–H (Teymourinia et al. 2017; Choppadandi et al. 2021). The peaks located around 2882 , 2382 , 2148 , and 2083 cm^{-1} correspond to the stretching vibration of C–H, C=O, C≡C, and C–N, respectively (Ananthanarayanan et al. 2014; Tashkhourian and Dehbozorgi 2016; Wang et al. 2016; Costa et al. 2018; Sadrolhosseini et al. 2020), and the peaks at 2013 and 1768 cm^{-1} were imputed to the stretching of C=O (Teymourinia et al. 2017; Choppadandi et al. 2021; Bokare et al. 2020). The peaks at 1693 and 1528 cm^{-1} were related to the stretching vibrations of C=C and C=O bonds, respectively (Tan et al. 2016; Zhao et al. 2016). The peak centered at 1341 cm^{-1} was assigned to the stretching vibration of C–H and the bending vibration of C–N bond (Tashkhourian and Dehbozorgi 2016; Yuan et al. 2014; Yan et al. 2015), and the peak at 1192 cm^{-1} was attributed to the stretching vibration of C–O bond and the stretching vibrations of C–N groups in amines (Bokare et al. 2020; Abbas et al. 2020). In addition, the peaks appearing at 1079 , 1028 and 603 cm^{-1} were due to the stretching vibrations of C–O, C–O–C and bending vibrations of C–H, respectively (Bokare et al. 2020; Zhao et al. 2016; Yan et al. 2015).

After introducing DA, FTIR spectrum recorded for GQDs film (red spectrum) reveals that a few peaks showed a decrease in intensity (3848 , 3742 , 1528 and 603 cm^{-1}) owing to the overlap with the stretching vibrations of N–H, while the intensity of the peak at 2148

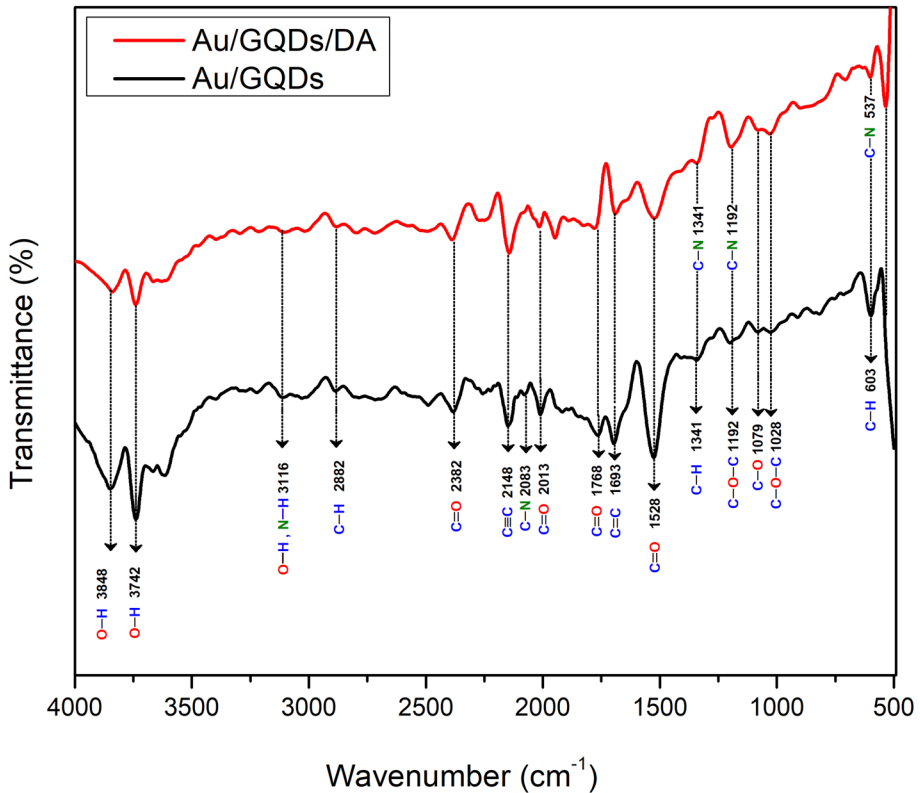


Fig. 2 FTIR spectra of GQDs/Au nanolayered film prior to and following the contact with DA

cm^{-1} was increased. Also, the peaks located at 1341 and 1192 cm^{-1} became more obvious due to C–N stretching vibrations. There was a new peak appeared at 537 cm^{-1} due to the amine C–N stretching (Wang et al. 2016). These results validated the DA-GQDs film interaction and demonstrated that when DA was added, the functional groups of GQDs changed. This confirms that DA was bound to the sensor film's surface and changed its refractive index.

3.2 Surface morphology of GQDs/Au nanolayered film

Before DA injection, the surface morphology of a GQDs film was scanned. The obtained 2D image as shown in Fig. 3a reveals the granular structure and distribution of GQDs on the surface of Au thin film, and the 3D AFM image (Fig. 3c) of GQDs film shows nanoneedles distributed regularly with maximum height of 5.3 nm. However, as shown in Fig. 3b, DA adsorption on the sensor chip affected its granular structure, reducing the number of nanoneedles and decreasing their maximum height to roughly 3.7 nm (Fig. 3d). Furthermore, the sensor surface's average roughness R_a was decreased from 0.801 nm to 0.755 nm, and R_q was reduced from 1.340 nm to 1.030 nm after DA injection. The considerable change in sensor film morphology and roughness following DA introduction confirms DA binding to GQDs thin film.

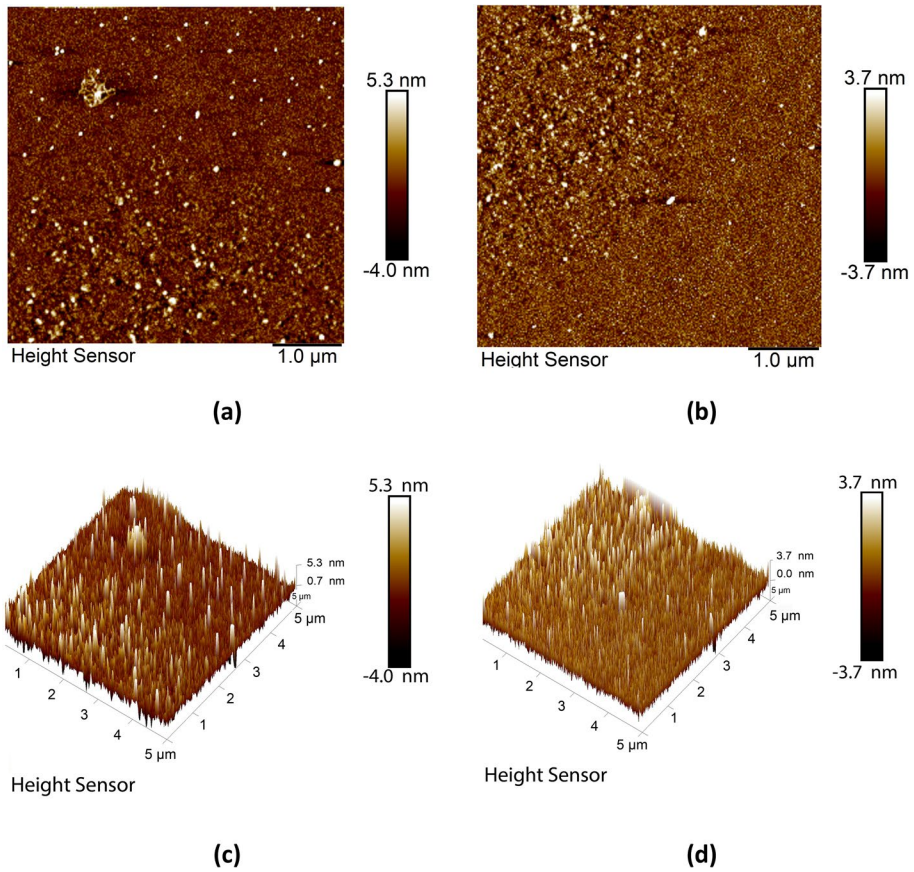


Fig. 3 AFM images of GQDs thin film: (a) 2D image before interaction with DA; (b) 2D image after interaction with DA; (c) 3D image before interaction with DA; and (d) 3D image after interaction with DA

3.3 Optical Characterization of GQDs/Au film

The thickness and refractive index of the GQDs/Au nanolayered film were determined through fitting the SPR experimental curves to theoretical data formula using Fresnel's Equation as shown in Fig. 4a–g (Fen and Yunus 2012). The simulation was done based on TMM in MATLAB. In Kretschmann setup, the multilayered structure GQDs/Au was positioned between the triangular prism and the DA solution. At both interfaces where the boundary conditions are met, reflection coefficient r can be expressed by:

$$r = \frac{m_{21} + m_{22}\gamma_2 - m_{11}\gamma_0 - m_{12}\gamma_2\gamma_0}{m_{21} + m_{22}\gamma_2 + m_{11}\gamma_0 + m_{12}\gamma_2\gamma_0} \quad (1)$$

Here m_{11} , m_{12} , m_{21} and m_{22} denote the elements of the transfer matrix, and γ_i can be obtained from the following formula:

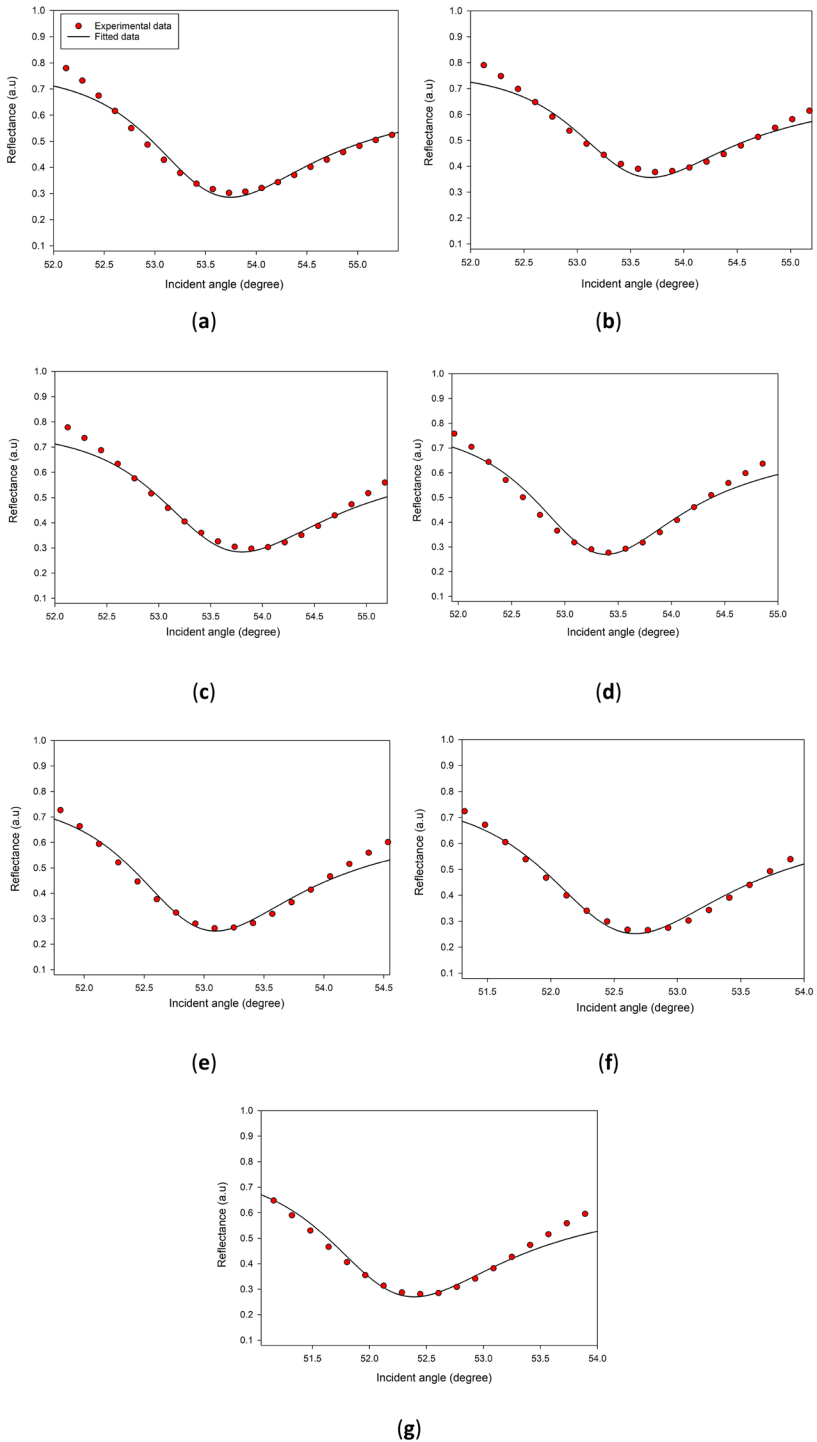


Fig. 4 Fitted and experimental reflectance curves for GQDs/Au nanolayered film exposed to DA solution with concentrations of: (a) 0 pM; (b) 1 pM; (c) 10 pM; (d) 100 pM; (e) 1 nM; (f) 10 nM; and (g) 100 nM

$$\gamma_i = \frac{n_i}{\cos(\theta_{ii})} \sqrt{\epsilon_0 \mu_0} \quad (2)$$

where ϵ_0 and μ_0 are the permittivity and permeability of free space, and $i=0, 1, 2$. The reflectivity (R) can be obtained using the formula below:

$$R = rr^* \quad (3)$$

The gold film's refractive index was found to be in good agreement with recent investigations (Fen and Yunus 2013b; Omar et al. 2022), where the n and k were 0.1950 and 3.6820, respectively, and thickness was 57.70 nm. The n and k values of DA solutions were the same as those of DW for concentrations lower than 10 pM. While for higher concentrations, the k value became 0.0030. The fitting yielded the n value of 1.3049 and k value of 0.0000 for GQDs film contacting DW with a thickness of 7.22 nm. As shown in Table 1, the interaction between the sensor chip and DA clearly had an influence on both the n value and the thickness of the GQDs monolayer film. The change in the sensing layer refractive index following contact with varied concentrations of DA solutions was clear through the angular shift of SPR dips.

This table shows the increased change in the n value of GQDs film as DA concentrations rose, which increased the change in the resonance angular shifts. This demonstrates the importance of GQDs thin film in enhancing sensor sensitivity to DA.

3.4 Sensing properties of DA on GQDs film

In our previous work, we have investigated the capability of SPR sensor based on bare gold to detect DA, and our results demonstrated that Au based SPR is insensitive to DA (Omar et al. 2020). Using GQDs/Au thin film, SPR measurements were conducted for DW first, then DA solutions of 1 fM, 1 pM, and 1 nM were introduced one by one into the flow cell to perform measurements and specify the concentration of DA that can be detected by this sensor film. SPR angle was 53.843° when DW contacted GQDs/Au sensing layer. Following that, by inserting DA solution at concentrations of 1 fM and 1 pM, the resonance occurred at 53.843°, the same as with DW. As DA concentration was increased from 1 pM to 1 nM, SPR dip was shifted to the left and the resonance took place at an angle of 53.011°. Because the SPR dip shifted significantly and the angular shift was around 0.830° when DA concentration increased from 1 pM to 1 nM, the measurements were performed for graduated concentrations between 1 pM and 1 nM, and continued for higher

Table 1 Refractive index and thickness values of GQDs monolayer film, the change of the real part of the refractive index Δn , and the resonance angle shift $\Delta\theta$

DA concentration (nM)	$n (\pm 0.0001)$	$k (\pm 0.0001)$	d (nm) (± 0.01)	Δn
0.000	1.3049	0.0000	7.22	0.0000
0.001	1.3049	0.0000	7.22	0.0000
0.01	1.2758	0.0000	6.83	0.0291
0.1	1.2448	0.0000	6.12	0.0601
1	1.2066	0.0000	5.92	0.0983
10	1.2023	0.0000	4.87	0.1026
100	1.1980	0.0000	4.63	0.1069

Table 2 The resonance angle and angular shift of the SPR dips for GQDs film in contact with DA solutions

DA concentration (nM)	SPR angle (deg)	$\Delta\theta$ (deg)
0.000	53.843	0.000
0.001	53.843	0.000
0.01	53.841	0.002
0.1	53.287	0.556
1	53.011	0.832
10	52.733	1.110
100	52.457	1.386

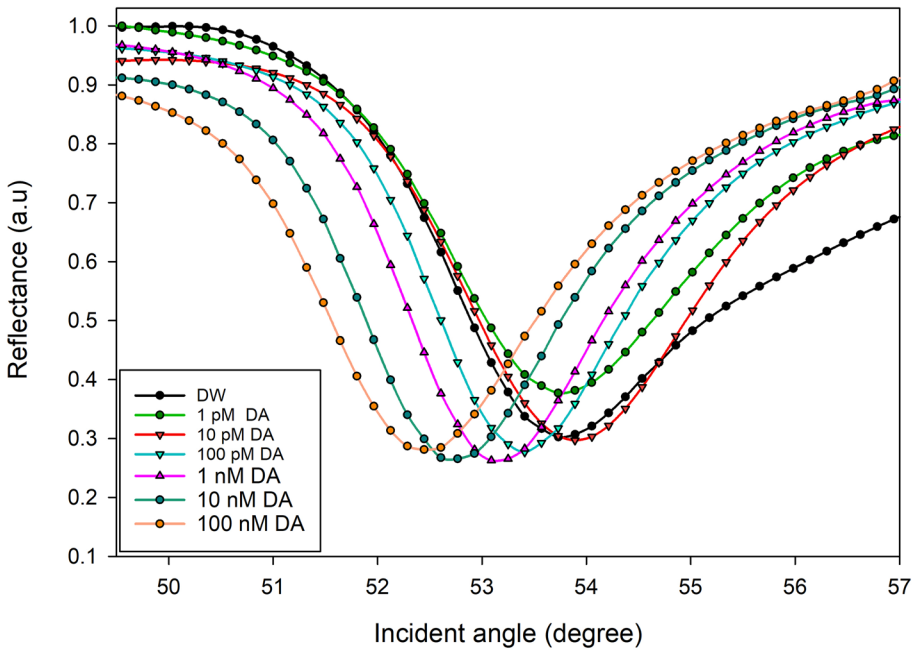


Fig. 5 SPR reflectivity curves acquired experimentally for GQDs/Au nanolayered film subjected to DA solution with concentrations from 1 pM to 100 nM

concentrations, to determine which concentration caused the first shift of the SPR dip. Using another GQDs/Au thin film, the resonance happened at an angle of 53.843° for both DW and 1 pM of DA. When 10 pM of DA was inserted into the attached cell, the SPR dip shifted slightly to lower angle at 53.841°. While, for 100 pM DA, the resonance happened at 53.287° and the angular shift was 0.556° as indicated in Table 2. When DA concentration was raised to 1 nM, the SPR reflectance curve remained shifted by 0.832° from the baseline as shown in Fig. 5. For 10 nM DA, the SPR dip shifted to lower angle of 52.733°. Clearly, the higher concentration of 100 nM of DA solution induced the greatest SPR dip shift of 1.386°.

The correlation between DA concentrations and the resonance angle shift of GQDs/Au based SPR sensor is shown in Fig. 6. The linear fitting yielded a good sensitivity of 0.332°/nM for this GQDs based SPR sensor towards DA ranging from 0.01 to 100 nM, with an R²

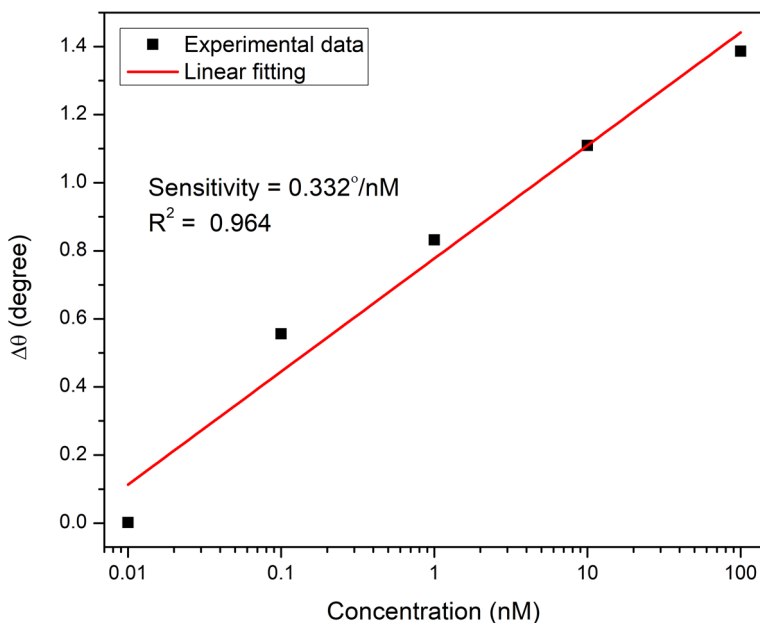


Fig. 6 The linear fitting for the change of resonance angle with DA concentrations

value of 0.964 and a LOD of 0.01 nM. Compared to previous reports on GQDs-based biosensors for DA detection, our sensor has demonstrated the ability to detect even lower concentrations of DA. For instance, Yan et al. (2015) introduced a photoelectrochemical biosensor employing GQDs-TiO₂, which demonstrated acceptable accuracy and precision in DA detection (Yan et al. 2015). Their biosensor exhibited an extensive linear range, spanning from 0.02 to 105 μ M, with LOD of 6.7 nM. In the study by Zhou et al. (2015), a fluorescence sensor for DA detection was introduced, utilizing polypyrrole PPy/GQDs core/shell hybrids (Zhou et al. 2015). These composites demonstrated robust fluorescence emission, with an enhancement of up to threefold compared to pristine GQDs. The developed sensor enabled highly sensitive DA determination through a decrease in fluorescent intensity upon the addition of DA. It exhibited excellent linearity within the range of 5–8000 nM, boasting a detection limit of 10 pM. Zhao et al. (2016) presented a fluorescence sensor based on GQDs (Zhao et al. 2016). Their sensor exhibited a linear correlation between quenching efficiency and DA concentration, falling within the range of 0.25–50 μ M, with a LOD of 0.09 μ M. Pang et al. (2016) employed a hydrothermal method to synthesize graphene quantum dots (GQDs) (Pang et al. 2016). These GQDs were then incorporated into a GQDs-Nafion composite to modify a glassy carbon electrode for use in an electrochemical sensor designed for dopamine (DA) detection. The interaction and electron communication between GQDs and DA were enhanced through π - π stacking forces. Nafion served as an anchoring agent, enhancing the stability and reproducibility of the GQDs on the electrode surface. This GQDs-Nafion composite exhibited a linear detection range spanning from 5 nM to 100 μ M, with an LOD of 0.45 nM for DA detection. Baluta et al. (2017) developed a fluorescence-based strategy for DA detection (Baluta et al. 2017). Their approach involved the formation of polydopamine (poly(DA)) on the surface of GQDs and utilized enzyme-laccase for substrate oxidation. Under optimized conditions, this method

exhibited strong analytical performance, featuring high sensitivity and selectivity across a broad linear range. Notably, it achieved a low LOD of 80 nM. The electrochemical sensor developed by Ben Aoun (2017) by modifying a nanostructured carbon screen-printed electrode with a chitosan/nitrogen-doped GQDs nanocomposite exhibited a high sensitivity of $418 \mu\text{AmM}^{-1} \text{cm}^{-2}$ with LOD of 0.145 μM in broad dynamic range (1–200 μM) (Ben Aoun 2017). Arumugasamy et al. (2020) developed a ratiometric electrochemical biosensor using GQDs combined with acid-functionalized multiwall carbon nanotubes (MWCNTs) on a glassy carbon electrode surface (Arumugasamy et al. 2020). Their sensor exhibited good electrocatalytic activity for DA oxidation, covering a dynamic linear range of 0.25–250 μM , with a low detection limit of 95 nM. Chatterjee et al. (2022) synthesized Boron and Sulfur co-doped GQDs (BS-GQDs) and utilized them as a label-free fluorescence-based sensor for the exceptionally sensitive and selective detection of DA. When DA was introduced, BS-GQDs displayed significant fluorescence intensity quenching within a broad concentration range of DA (0–340 μM), achieving LOD of 3.6 μM (Chatterjee et al. 2022). This SPR-based sensor clearly outperforms existing detection methods employing the same material (GQDs) and its composites in constructing the sensing platform.

In order to study the binding affinity of GQDs/Au based sensor towards DA, the non-linear fitting was applied to the experimental results based on Langmuir and Freundlich isotherm model as shown in Fig. 7. The Langmuir and Freundlich model's equation is as follows (Vijayaraghavan et al. 2006):

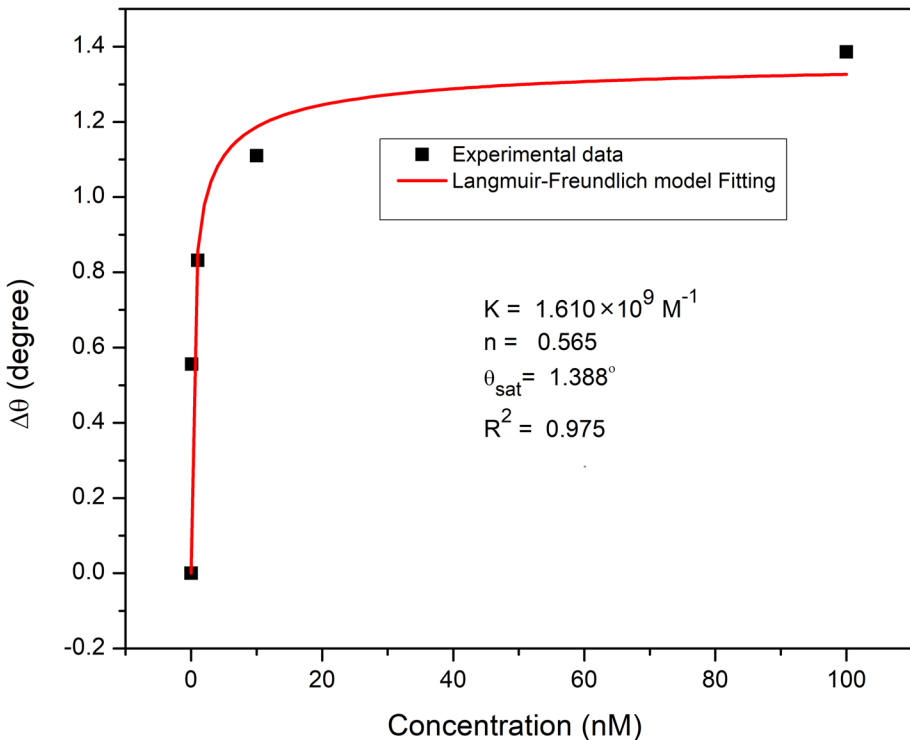


Fig. 7 Experimental and fitting data to Langmuir and Freundlich model for the adsorption of DA on GQDs/Au nanolayered film

$$\Delta\theta = \frac{\Delta\theta_{max}KC^n}{1 + KC^n} \quad (4)$$

where $\Delta\theta_{max}$ represents the maximum value of the resonance angle shift, K indicates the affinity constant, C is the concentration of the analyte, and n represents the system heterogeneity index.

Langmuir and Freundlich isotherm model was well suited to the experimental results with K value of $1.610 \times 10^9 \text{ M}^{-1}$ and correlation coefficient R^2 of 0.975. Langmuir and Freundlich exponent value was 0.565, and the $\Delta\theta_{max}$ value produced from this model was so close to value obtained experimentally (1.386°).

All SPR curves were fitted to Gaussian model in order to calculate their full width half maximum (FWHM) values. The FWHM value obtained for the reference signal was 3.143° with detection accuracy of $0.318 \text{ (deg}^{-1}\text{)}$, where the detection accuracy is inversely related to FWHM (Ge et al. 2022). The measurements conducted with DA resulted in SPR curves that were narrower than that for DW, where the obtained value for 1 pM DA was 2.671° as shown in Table 3. This suggests that injecting DA solution to touch the sensor film improved detection accuracy. This might be attributed to sensor film deterioration with increased DA concentrations, which reduced film thickness and FWHM, where the primary resonance experienced a shift. When DA concentrations were increased to 100 pM, the FWHM values continued to fall while the detection accuracy increased to $0.398 \text{ (deg}^{-1}\text{)}$. The injection of 1 nM DA resulted in an FWHM value of 2.605° , which thereafter dropped to 10 nM. The signal-to-noise ratio (SNR) is calculated by multiplying the resonance angle shift and the detection accuracy (Cennamo et al. 2013; Daniyal et al. 2018). The variation in SNR and detection accuracy values as a function of DA concentrations is shown in Fig. 8. The refractive index of the sensor film significantly changed with increasing DA concentrations, which shifted the SPR dips. As a consequence, the signals noise was decreased and SNR for this sensor were increased.

The strong affinity of DA for the GQDs sensing layer can be attributed to noncovalent interactions between the hydroxyl and carboxyl groups present on the GQDs and the diols, amine functional groups, and phenyl structure in DA. Additionally, π - π stacking forces further bolster the interaction between DA and the GQDs film (Ben Aoun 2017). These combined interactions contribute to the effective detection of DA by this sensor.

Table 3 The values of FWHM, detection accuracy and SNR of the developed sensor

DA Concentration (nM)	FWHM (deg)	Detection accuracy (deg^{-1})	SNR
0.000	3.143	0.318	0.000
0.001	2.671	0.374	0.000
0.01	2.649	0.377	0.001
0.1	2.514	0.398	0.221
1	2.605	0.384	0.319
10	2.496	0.401	0.445
100	2.886	0.347	0.480

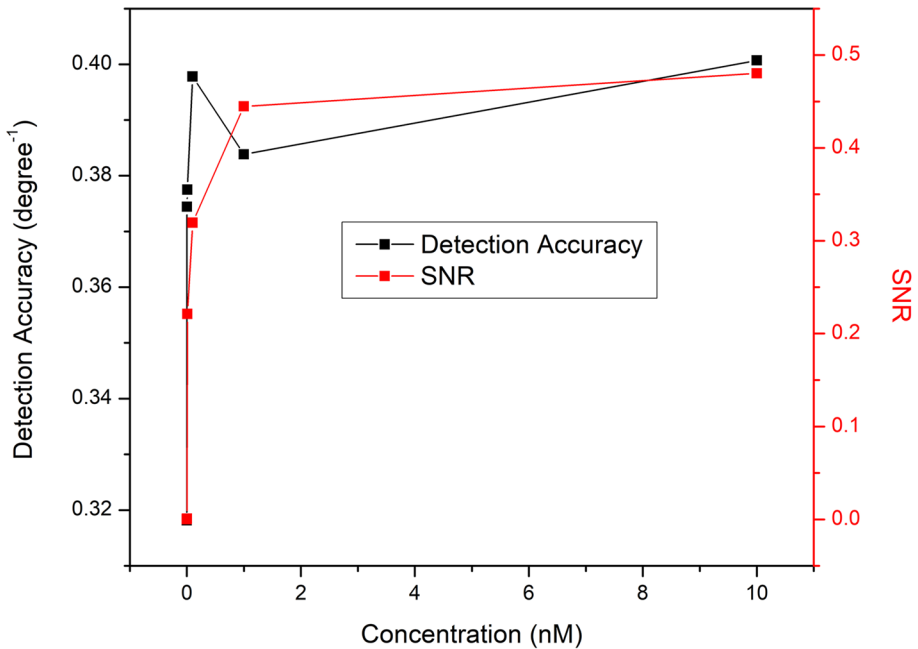


Fig. 8 Variations of the detection accuracy and SNR with DA concentration

4 Conclusions

To conclude, GQDs thin film was prepared and integrated into SPR spectroscopy. Its sensing properties towards DA were investigated for various concentrations of DA solution ranging from 0.01 to 100 nM. Experimentally, the angular shifts of SPR dips were observed when DA concentrations were increased owing to the adsorption of DA on the surface of GQDs film which led to its morphological changes as was verified by FTIR and AFM analysis. The optical properties and thickness of this thin film were determined through fitting the experimental SPR curves to theoretical data based on TMM. This GQDs film combined with the plasmonic based sensing platform proved its efficiency in detecting induced variations in the refractive index of the sensing medium when the thin film was in contact with low concentrations of DA.

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Data availability All data required to reproduce these findings are included into the paper.

Declarations

Competing interests The authors declare no competing interests.

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