Surface plasmon resonance based on graphene-metal-graphene structure: recurrence relation theory

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The last years, graphene has opened exciting new fields in graphene plasmonics, due to the graphene's unique optoelectronic properties such as long-lived collective excitation, extreme optical confinement in graphene plasmonics and extraordinary light-matter interactions in metamaterials. Therefore, these excellent properties make graphene a favorable candidate for novel plasmonic devices and potential applications in photonics, optoelectronics and sensor technologies. In this work, theoretical investigations are carried out to in Graphene-Metal-Graphene structure for enhanced surface plasmon resonance based on the recurrence relations' method. We find that the graphene-metal-graphene structure supports both high-energy optical plasmon oscillations and out-of-phase low energy acoustic charge density excitations. Since a high performance of surface plasmon resonance excitations should exhibit a large depth of dip (small reflectivity), the minimum of reflectivity in the hybrid structure can be manipulated dynamically by changing the thickness of the metallic film, the number of the graphene layers and the dielectric proprieties of the surrounding dielectric materials. Based on this principle, different kinds of plasmonic sensors have been designed in previous years.

Keywords: Graphene plasmonics; plasmons; reflectance.

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1. Introduction

Surface plasmons (SPs) are transverse magnetic (TM) polarized electromagnetic waves coupled with charge density oscillations (plasmons) traveling along metal-dielectric interface. The electric field associated with these oscillations decay exponentially into the dielectric medium making plasmons extremely sensitive to the refractive index of the medium. When the wave vector of the incident light matches the wave vector of the SP wave, the SP resonantly couples with the incident light and a remarkable electric field enhancement can be realized, and the so called SP resonance occurs. Usually surface plasmons can be excited via evanescent waves in the Kretchmann configuration, utilizing high-index prisms, where the wavevector mismatch between vacuum and SP is compensated [1]. Once the SP is excited in the Kretchmann configuration, partially of the energy associated to the incident of the electromagnetic radiation will be transferred to the SP, and a sharp minimum is observed in the reflectance versus angle (or wavelength) curve. The ability of controlling strong light-matter interaction through surface plasmons in metals has driven the field of plasmonics. Additionally, increasing research has been carried out to investigate and manipulate SP for new sensing functionalities. Based on this principle, different architectures of plasmonic sensors involving metals have been designed in previous years [2,3]. However, the major obstacle in developing plasmonic applications is dissipative loss, which limits the propagation length of surface plasmons and broadens the bandwidth of surface plasmon resonances and their surfaces easily oxidize degrading their plasmonic characteristics [4]. To overcome these shortcomings, some new structures have been proposed to investigate surface plasmon resonances for new plasmonic materials; among them, graphene has emerged as an alternative, unique two- dimensional material able to extend the field of plasmonics for terahertz to mid-infrared applications [4-7]. Graphene is a two-dimensional material made of carbon atoms arranged in hexagonal lattice. Graphene material has attracted tremendous attention due to its physical properties including high electrical and thermal conductivity, optical transparency, and controllable plasmon properties. Two-dimensional plasmons in graphene exhibit unique optoelectronic properties and mediate extraordinary light-matter interactions. Therefore, these excellent properties make graphene a favorable candidate for novel plasmonic devices and potential applications in photonics, optoelectronics, and in sensor technologies [8]. Hence, the hybridization of graphene-metal metamaterials plays an important role in the field of plasmonics and exploring the interactions of the plasmon modes in multilayer graphene structures coupled via Coulomb interaction with metallic substrates offer new opportunities for applications and fundamental studies of collective electron excitations in plasmonic metamaterials for biological and chemical sensing [9-17], photodetectors [18] and optoelectronics [19,20].

In this paper, we report a theoretical investigation of SPs of hybrid metal graphene structures for surface plasmons resonances applications. Specifically, we consider a graphene-metal-graphene metamaterial surrounded by a semi-infinite materials of dielectric constants ε_1 and ε_2, respectively, see Fig. 1. Solving Poisson equation for the electric potential and

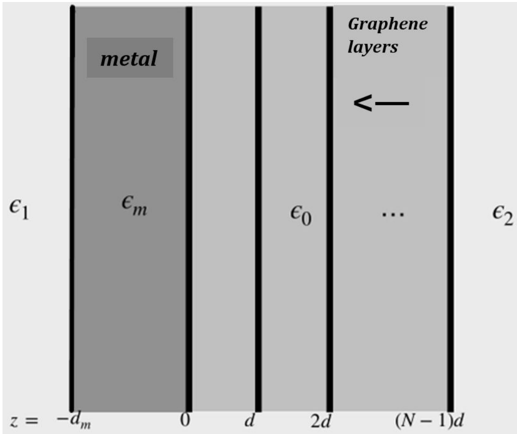

FIGURE 1. Schematic representation of a metal-graphene metamaterial surrounded by a semi-infinite materials with dielectric constants ε_1 (left) and ε_2 (right), respectively. The incident radiation is on the metallic surface at $z = -d_m$, the separation of the graphene layers is d embedded in a material of dielectric constant ε_0.

and applying standard boundary conditions at the interfaces between the different layers, the amplitudes associated with the electric potential between different layers satisfy a linear recurrence relation of second order and consequently, an analytical expression for the dielectric function of the metamaterial can be obtained. The zeros of the dynamic dielectric function lead the dispersion relation of the plasmon modes. It is important to mention that in the past years the transfer matrix method has become a standard theoretical model to study optical properties and charge density excitations in non-interacting graphene layers. Thus, the recurrence method offers an alternative method to study plasmon modes in multilayer structures. It is found that the SPs of the hybrid structure depend significatively on the number of graphene layers, the thickness of the metallic film and the dielectric properties of the environment. Thus, it is important to point out that the reflectivity property depends not only on the angle (or wavelength) of the incident radiation but also on the geometrical parameters of the metamaterial and the sensing medium. It is worth to mention that our results may elucidate the SPs resonances sensor of the graphene-metal-graphene structured numerically investigated in Ref. [5].

2. Theoretical approach

In this work, we consider a periodic system consisting of a finite graphene monolayer stack; each layer with an electron density n spatially separated a distance d on a substrate constituted by a metal film of thickness d_m. The layered graphene system is represented by an array of N graphene layers located ay $z = ld(l = 0, 1, N-1)$ inserted in a material with background dielectric constant ε_0. The substrate constituted by a graphene-metal composite film occupy the space $-dm < z < -0$. The graphene-metal-graphene metamaterial is surrounded by a semi-infinite material with dielectric constants ε_1 and ε_2, respectively, see Fig. 1.

In previous works, a transfer matrix method has been applied to structures consisting of periodically arranged graphene layers for optical calculations [21]. In this paper, we adopt an alternative theoretical method for plasmons in periodic graphene layered structures [22].

The collective electronic excitations can be found by solving Poisson equation for the electrostatic potential as $\nabla^2\varphi(r,t) = 0$ between graphene layers where $\varphi(z)e^{i(qx-\omega t)}$ and the electrostatic potential between l and $l+1$ layers is given as $\varphi_1(z) = A_l e^{-q(z-ld)} + B_l e^{q(z-ld)}$ where A_l and B_l represent the coefficients of the forward and backward propagation wave, standard electromagnetic boundary conditions at $z = ld$, yields

$$A_{l-1}e^{-qd} + B_{l-1}e^{qd} = A_l + B_l$$
$$-A_{l-1}e^{-qd} + B_{l-1}e^{qd} - (-A_l + B_l) = 2V_q\Pi(q,\omega)$$
$$\times(A_{l-1}e^{-qd} + B_{l-1}e^{qd}), \quad (1)$$

where $V_q = 2\pi e^2/\varepsilon_0 q$ is the Fourier transform of the two-dimensional Coulomb interaction and $\Pi(q,\omega)$ is the graphene electron polarizability calculated in Refs. [23,24]. Equation (1) can be written as

$$\begin{pmatrix} A_l \\ B_l \end{pmatrix} = T \begin{pmatrix} A_{l-1} \\ B_{l-1} \end{pmatrix},$$
$$T = \begin{pmatrix} (1 + V_q\Pi(q,w))e^{-qd} & V_q\Pi(q,w)e^{qd} \\ -V_q\Pi(q,w)e^{-qd} & (1 - V_q\Pi(q,w))e^{qd} \end{pmatrix}, \quad (2)$$

here, T represents the transfer matrix connecting the coefficients at adjacent graphene layers with unit determinant and $Tr(T) = 2(\cos hqd - Vq\Pi(q,\omega)\sin hqd) = 2R$. From Eqs. (2), it is possible to obtain the following linear recurrence relation of second order with constant coefficients,

$$A_{l+2} - 2RA_{l+1} + A_l = 0. \quad (3)$$

Recurrence relations given by Eq. (3) can be solved using standard methods, the solutions of which can be written in terms of the electrostatic amplitudes A_0 and B_0 as

$$A_l = \frac{-1}{\operatorname{sen}\theta}\Big[A_0 \sin(l-1)\theta$$
$$- A_0T_{11}\sin l\theta - B_0T_{12}\sin l\theta\Big], \quad \text{and}$$
$$B_l = \frac{-1}{\operatorname{sen}\theta}\Big[B_0 \sin(l-1)\theta$$
$$- B_0T_{22}\sin l\theta - A_0T_{21}\sin l\theta\Big], \quad (4)$$

where the coefficients T_{nm} are the elements of the transfer matrix, the angle θ is defined as $\cos\theta = R$, $|R| < 1$.

According to Fig. 1, the existence of graphene-metallic substrate in the range $-dm < z < 0$ and the dielectric boundaries where the graphene-metal-graphene structure is immersed, the electrostatic potential outside the hybrid structure is written as

$$\varphi(z) = \begin{cases} Ce^{qz} & Z < -d_m \\ N_m e^{-qz} + M_m e^{qz} & -d_m < z < 0 \\ De^{-qz} & z > (N-1)d \end{cases}, \quad (5)$$

matching the electromagnetic boundary conditions at the interfaces $z = 0$, $z = -dm$ and $z = (N-1)d$, leads the relationship between the electrostatic coefficients A_0, B_0, A_{N-2} and B_{N-2};

$$\frac{A_0}{B_0} = \frac{(1 - \varepsilon_m/\varepsilon_0 + 2V_q\Pi(q,\omega))e^{2qd_m} + (1 + \varepsilon_m/\varepsilon_0 + 2V_q\Pi(q,\omega))F(q,\omega)}{(1 + \frac{\varepsilon_m}{\varepsilon_0} - 2V_q\Pi(q,\omega))e^{2qd_m} + (1 - \frac{\varepsilon_m}{\varepsilon_0} - 2V_q\Pi(q,\omega))F(q,\omega)} = H(q,\omega), \quad (6)$$

$$\frac{A_{N-2}}{B_{N-2}} = \frac{-1 - \frac{\varepsilon_2}{\varepsilon_0} + 2Vq\Pi(q,\omega)}{-1 + \frac{\varepsilon_2}{\varepsilon_0} - 2Vq\Pi(q,\omega)} = \frac{e^{2qd}}{G(q,\omega)}, \quad (7)$$

$$F(q,\omega) = \frac{-\frac{\varepsilon_1}{\varepsilon_0} + \frac{\varepsilon_m}{\varepsilon_0} + 2V_q\Pi(q,\omega)}{\frac{\varepsilon_1}{\varepsilon_0} + \frac{\varepsilon_m}{\varepsilon_0} - 2V_q\Pi(q,\omega)}, \quad (8)$$

with $\varepsilon_m(\omega) = \varepsilon_\infty$-$\omega_p^2/\omega^2$ the dielectric permittivity of the metal and ω_p and ε_∞ the plasmon frequency and dielectric constant at infinite frequency, respectively. The condition for the self-sustaining collective oscillations (plasmons) occur when the amplitudes of the electrostatic potential A_m and $B_m \neq 0$ while the effective dielectric constant of the multilayer graphene-metal-graphene $\varepsilon(q,\omega)$ vanishes. An analytic expression for $\varepsilon(q,\omega)$ is calculated making use of Eqs. (6) and (7) in Eq. (4) as

$$\begin{aligned} \varepsilon(q,\omega) &= G(q,\omega)H(q,\omega)[\sin(N-3)\theta - T_{11}\sin(N-2)\theta] \\ &- [G(q,\omega) + H(q,\omega)]T_{12}\sin(N-2)\theta \\ &- [\sin(N-3)\theta - T_{22}\sin(N-2)\theta]e^{2qd} = 0. \end{aligned} \quad (9)$$

In general, numerical results for the plasmon dispersion can be presented by solving numerically Eq. (9) for a system consisting of a finite number of graphene layers N coupling via Coulomb interaction with the electron gas in the graphene-metal as shown in Fig. 1. However, in the THz region, which is our interest, we take the long wavelength approximation such that $qd \ll 1$. Under this approximation $\theta \sim q$ and [24]

$$\Pi(q,\omega) = \frac{E_f}{\pi}\frac{q^2}{\omega^2}\left(1 - \frac{\omega^2}{E_f^2}\right). \quad (10)$$

Here E_f is the Fermi energy of the two-dimensional electron gas in monolayer graphene. Thus, Eq. (9) reduces to

$$\begin{aligned} \varepsilon(q,\omega) &= G(q,\omega)H(q,\omega)[N - 3 - T_{11}(N-2)] \\ &- [G(q,\omega) + H(q,\omega)T_{12}(N-2) \\ &- [N - 3 - T_{22}(N-2)]e^{2qd} = 0, \end{aligned} \quad (11)$$

whose solution for the plasmon optical mode is written as

$$\omega^2 = \frac{4(N+1)e^2E_f}{\varepsilon_1 + \varepsilon_2}q + \frac{d_m\omega_p^2}{\varepsilon_1 + \varepsilon_2}q, \quad (12)$$

where the first term of the right hand in Eq. (12) represents the optical plasmon energy of a monolayer graphene times the graphene number of layers $N + 1$ in the hybrid structure, *i.e.*, the electron density oscillations of the stacked graphene layers are in phase coupled via Coulomb interaction with the symmetric plasmon modes in the metal film. Equation (12) also shows how a graphene induces a shift of surface-plasmon resonances of metal films, which agrees with the experimental results reported in Ref. [14].

3. Numerical results

Surface plasmons in graphene-metal-graphene can be significantly influenced by many-particle involving interaction between electrons and plasmons and besides that, they can be tuned continuously by manipulating the thickness of the metal, the dielectric properties of the external medium, etc. The dispersion curves of the surface plasmons on graphene-metal-graphene structures, Eq. (11), reveal these essential features. Therefore, the interaction of light with surface plasmons can be enhanced leading a great ability for biological and chemical sensing applications. Usually surface plasmon can be excited via evanescent waves in the Kretschmann configuration, utilizing high-index prisms, where the wavevector matching between incident light and surface plasmon is compensated. Once surface plasmon is excited in the Kretschmann configuration, a sharp minimum is observed in the reflection coefficient versus incident angle (or wavelength) curve.

In the present work we consider a p-polarized light incident on the graphene-metal substrate, and to determine the reflectance of plasmon resonances as a function of the incident angle, we solve the Fresnel problem for the graphene-metal-graphene structure as developed in Ref. [21].

As depicted in Fig. 1, if the $x - y$ plane is the interface plane, for wave propagation in the x direction, the magnetic field is polarized along the y direction and can be written in the form $H = (0, Ue^{-ikz} + Ve^{ikz}, 0)e^{ikx-i\omega t} = (0, \Phi, 0)$ and the electric field associated with this electromagnetic wave is $E = -c/i\omega\varepsilon(-\partial_z\Phi, 0, \partial_x\Phi)$ with $k^2 = \varepsilon\omega^2/c^2 - \kappa^2$ and c the light velocity in vacuum. Furthermore, the propagation of light across the set of interfaces formed by a graphene-metal graphene must satisfy the standard electromagnetic boundary conditions, *i.e.*, the tangential component of E is continuous and H discontinuous. Thus, the relationship between the field components U, V at the first and last interface of the layered structure are related by a 2×2 transfer matrix M, namely

$$\begin{pmatrix} U_1 \\ V_1 \end{pmatrix} = M \begin{pmatrix} U_{N+1} \\ V_{N+1} \end{pmatrix}, \tag{13}$$

where M is the characteristic transfer matrix of the combined metal-graphene stack and can be obtained from the following relation for p-polarized incident light [25]:

$$M = R_{1\to 2} P(d_2) R_{2\to 3} P(d_3) \ldots P(d_N) R_{N\to N+1}, \tag{14}$$

where the electric and magnetic field in different layers are related through the propagation matrix $P(d_{m+1})$

$$P(d_{m+1}) = \begin{pmatrix} e^{-ik_{m+1}d_{m+1}} & 0 \\ 0 & e^{ik_{m+1}d_{m+1}} \end{pmatrix}, \tag{15}$$

and the transmission matrix

$$R_{m\to m+1} = \frac{k_{m+1}}{2\varepsilon_{m+1}} \begin{pmatrix} \frac{\varepsilon_m}{k_m} + \frac{\varepsilon_{m+1}}{k_{m+1}} + \frac{4\pi}{\omega}\sigma(\omega) & -\frac{\varepsilon_m}{k_m} + \frac{\varepsilon_{m+1}}{k_{m+1}} - \frac{4\pi}{\omega}\sigma(\omega) \\ -\frac{\varepsilon_m}{k_m} + \frac{\varepsilon_{m+1}}{k_{m+1}} - \frac{4\pi}{\omega}\sigma(\omega) & \frac{\varepsilon_m}{k_m} + \frac{\varepsilon_{m+1}}{k_{m+1}} - \frac{4\pi}{\omega}\sigma(\omega) \end{pmatrix}. \tag{16}$$

Here $\sigma(\omega)$ is the surface conductivity of graphene which considers the contribution of the interband and intraband electronic transitions

$$\sigma_{\text{intra}} = \frac{2ie^2 k_B T}{\pi\hbar^2(\omega + i\Gamma)} \ln\left[2\cosh\left(\frac{E_f}{2k_B T}\right)\right], \tag{17}$$

and

$$\sigma_{\text{inter}} = \frac{e^2}{4\hbar}\left[\frac{1}{2} + \frac{1}{pi} + \arctan\left[\frac{\hbar\omega - E_f}{2k_{BT}}\right] - \frac{i}{2\pi}\ln\frac{(\hbar\omega + E_f)^2}{(\hbar\omega + E_f)^2 + (2K_B T)^2}\right], \tag{18}$$

where e is the electron charge, k is the Boltzmann's constant, $\hbar$ is the reduced Planck's constant, T is the temperature, and ω is the angular frequency of incident light and Γ is the relaxation frequency. Once the transfer matrix is known, we can calculate the optical properties of the multilayer graphene-metal structure. Assuming the incident light is on the metallic slab, $z = 0$, it can be easily shown that the reflection coefficient is given by the elements of the matrix M, $r = M_{21}/M_{11}$ and the reflectivity $R = |r|^2$.

In Fig. 2 the reflectivity of the metamaterial is depicted as a function of the angle associated with the incident radiation for metal thickness increasing from 50 to 70 nm. One important note from this plot is the optimum thickness value such that the resonance dip in the reflectivity curve becomes the sharpest with a reflection minimum close to zero. Furthermore, as can be seen from the plot, optimum thickness value is around 70 nm at a resonance angle near to 0.88 radians ($\sim$ 50 deg), and a wavelength of the radiation $\lambda = 632$ nm, the dielectric constant of the prism $\varepsilon_1 = 2.25$, the plasma frequency of the metal $\omega_p = 1.37 \times 10^{16}$ sec^{-1}, the graphene Fermi energy $E_f = 0.5$ eV and the dielectric constant of the sensing medium $\varepsilon_2 = 1.2$, here $N = 1$.

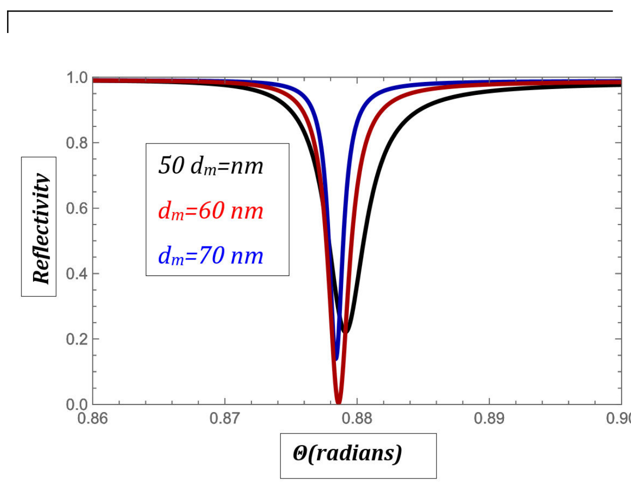

FIGURE 2. Reflectivity spectra of metal-graphene structure as a function of the incident angle of the incident radiation for different thickness of the metallic film with $\lambda = 632$ nm, $\varepsilon_1 = 2.25$, $\varepsilon_2 = 1.2$, $E_f = 0.5$ eV, $\omega_p = 1.37 \times 10^{17}$ sec^{-1}, $N = 1$.

Another parameter that affects the performance (minimum reflectivity) of the proposed optical metamaterial is the wavelength of the incident radiation. In Fig. 3, the reflectivity intensity with respect to the incident angle is shown for the configuration of highest sensitivity ($d = 60$ nm) with three different wavelengths. The reflectivity intensity exhibits maximum dip at $\lambda = 800$ nm and with $\lambda = 632$ nm and $\lambda = 550$ nm, respectively. These results indicate a shift of the resonance dip in the reflectivity showing that wavelength of the incident radiation can be optimized to enhance the sensitivity surface plasmon resonance sensor as shown numerically in Ref. [5].

Surface plasmon resonance can be also used to detect biological molecules [26]. This feature is used to design opti-

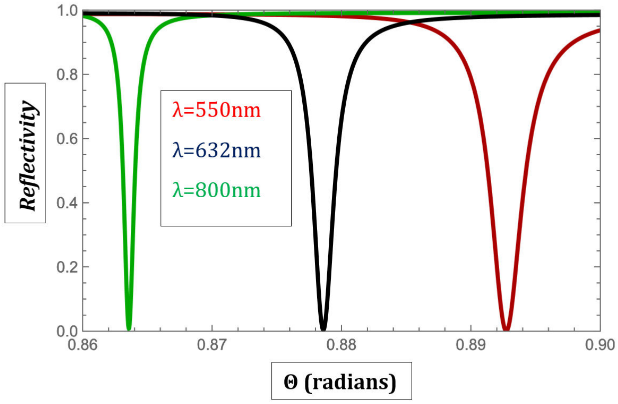

FIGURE 3. Surface plasmon resonance reflectivity of metal-graphene metamaterial as a function of the incident angle for different incident light wavelength with metallic film thickness $d_m = 60$ nm. The other parameters of structure are the same as in Fig. 2.

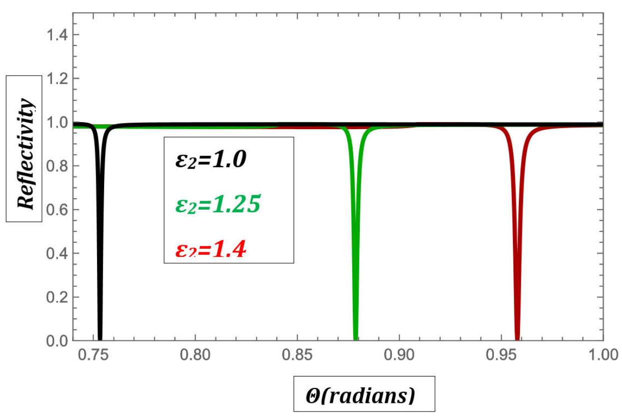

FIGURE 4. Shows the reflectivity curves as a function of incident angle of the incident radiation for different dielectric permittivity of adsorbed organic molecules on graphene surface with $d_m = 60$ nm. The other parameters of metal-graphene structure are the same as in Fig. 2.

cal biosensors that can measure the refractive index when the biomolecules become adsorbed on the graphene surface and create a layer of refractive index higher than that of the air (ε_2 in Fig. 1) resulting in a change in the resonance angle. Figure 4 plots the theoretical reflectivity against the resonance angle for different refractive index of the sensing medium. As can be seen, there is a shift of the reflectivity depth dip on the incident angle for different dielectric constants of the sensing medium. Therefore, this plasmonic metamaterials are very sensitive to the changes of refractive index of the dielectric media in the vicinity of the graphene layer.

Finally, Fig. 5 shows the variations of the reflectivity with the number of the graphene layers N and the graphene layers

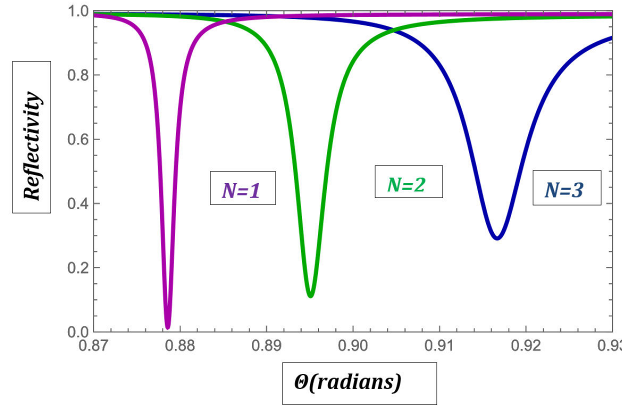

FIGURE 5. Reflectivity spectra of the metal-graphene metamaterial as a function of the incident angle for different number of the graphene layers. The other parameters are the same as Fig. 2.

separation such that $qd \ll 1$(the frequency range is $0.1 - 10$ THZ) . These results indicate that the number of graphene layers can be optimized to enhance the performance of the hybrid metamaterial structures-based surface plasmon biosensor. As can be seen, when the number of graphene layers increases there is a shift of the resonance peak and a reduction in the amplitude of the resonance dip. Thus, a maximuum transfer of energy of the incident radiation corresponds to one graphene layer, $N = 1$.

4. Conclusions

In this study, the collective electronic excitations in metal-layered graphene structure were investigated. Long-range Coulomb interactions in the metamaterial lead new set spectra of surface plasmons, which depend on a certain characteristic parameter of each material in metal-layered graphene structure. At long wavelength ($qd \ll 1$) the optical plasmons emerge in this metamaterial structure characterized by a square root dependence on the momentum, which can be excited under the Kretschmann coupling configuration. Since a high performance of surface plasmon resonance should exhibit large depth of dip (small reflectivity), the minimum of the reflectivity in the hybrid configuration can be manipulated dynamically by changing, the thickness of the dielectric film, the dielectric properties of the environment, the number of graphene layers, etc. which is an efficient method to realize high sensing properties for the metamaterial. As an application, we have shown that metal-graphene stack is a sensitive THz plasmonic biosensor to study graphene layer interactions with biomolecules characterized with their own dielectric permittivity [5,26].

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