Behaviour of SPP waves frequency spectrum for dielectric/metal/dielectric structures

Petro Kostrobiy, Vitalii Polovyi, Iryna Ryzhha
Lviv Polytechnic National University, Lviv, 12. Stepana Bandery Str. Lviv
Vitaliy.Y.Polovyi@lpnu.ua

Abstract: In this paper, the authors studied the influence of different phenomena and materials properties on the frequency spectrum of the surface plasmon-polariton (SPP) waves behaviour in "dielectric/metal/dielectric" structures. Among them are spatial dispersion, quantization of energy levels, electron-electron interaction (Coulomb correlations) thickness of a metal layer. Established that for atomically thin metal films mentioned effects affect the frequency spectrum significantly. Mentioned that taking into account quantum-sized effects reveals a specific oscillatory picture of the spectrum that repeats one obtained for chemical potential. Proposed model yields results that have a quite good agreement with an experimental data.

Keywords: SURFACE PLASMON-POLARITONS, FREQUENCY SPECTRUM, ATOMICALLY THIN METAL FILMS, QUANTUM-SIZED EFFECTS, COLOUMB CORRELATIONS

1. Introduction

The basic application of surface plasmon-polariton waves in so-called plasmon structures (i.e., structures in which propagate SPP) is the development on their basis of compact highly sensitive optical waveguides [1,2], plasmon biosensors [3-5] designed to study the presence of impurities, the effect of drugs in a real time, etc. which work is based on the surface plasmons resonance effect [6] and laser stethoscopy devices [7].

Also, since plasmons are characterized by a high localization and intensity increase at the nanoscale, the control of the light at the nanoscale is a promising field. In addition, it should be noted that another important application of plasmonics is the use as a conductive material of a planar graphene [8-12], which is characterized by a high activity of charge carriers [10,13] and, therefore, leads to less energy losses [11,12,15].

Another promising application of plasmonics that will be of most interest to us in this paper is the use of metal-dielectric heterogeneous structures to excite plasmon waves, such as dielectric/metal, metal/dielectric or dielectric/metal/dielectric [1,15-23]. Usage of different materials for metals or dielectrics makes it possible to obtain plasmon waves with different integral characteristics, such as frequency or length of propagation.

Given the variety of application, various properties of SPP waves and existence of necessary tools for research, both mathematical and experimental, this topic also attracts the attention of many experimental scientists [22-27].

In this paper we focused our attention on a dielectric/metal/dielectric structures, when a metal is an atomically thin film (ATMF) with a thickness up to 100 nm.

2. Prerequisites and means for solving the problem

A structure we simulated propagation of SPP waves in consists of two dielectrics with sandwiched ATMP between (Fig. 1). The coordinate for the strucuted is chosen so the plane divides ATMP on two even parts. SPP wave will propagate along X axis.

According to this we have the following mathematical boundaries for the areas:

\[
\Omega = \begin{cases}
\Omega_1, & z < -1/2, \\
\Omega_2, & -1/2 < z < 1/2, \\
\Omega_3, & z > 1/2.
\end{cases}
\]

Dielectric permittivity of insulators \( \varepsilon_1(\omega) \) and \( \varepsilon_2(\omega) \) are taken in the high-frequency approximation, thus, they are functions of the time variable / frequency. On the contrary, the dielectric permittivity of a metal depends on both time variable and spatial coordinates \( \vec{r}, t \).

\[
\varepsilon(\vec{r}, \vec{r}, t - t').
\]

Here we assume that external charges (stimuli) \( \rho \) in the area of ATMF are absent, so the Maxwell system of equations are such

\[
div\vec{D} = 0, div\vec{B} = 0, \\
rot\vec{H} = \frac{\partial \vec{D}}{\partial t}, rot\vec{E} = \frac{\partial \vec{B}}{\partial t}.
\]

Here \( \vec{H} \) and \( \vec{E} \) are vectors of magnetic and electric strength and \( \vec{B} \) and \( \vec{E} \) are vectors of electric and magnetic flux density.

As we mentioned before in our model, we considered a spatial dispersion in the area of ATMF. In this case a connection between \( \vec{B} \) and \( \vec{E} \) vectors are non-local as process we investigate is non-stationary by nature [28]

\[
\vec{D}(\vec{r}, t) = \int d\vec{r}' \int dt' \varepsilon(\vec{r}, \vec{r}', t - t') \vec{E}(\vec{r}', t').
\]

Also, we considered a transverse magnetic (TM) polarization of vectors \( \vec{H} \) and \( \vec{E} \) vectors as this type of polarization SSPs exists. Then we have the following form of these vectors:

\[
\vec{H} = (0, H_y, 0), \vec{E} = (E_x, 0, E_z).
\]

After applying a Fourier transform to equations (3) and (4) and considering (5) we obtain a system of wave equations

\[
\frac{\partial^2 H_y}{\partial z^2} + (k_y^2 \varepsilon_1(\omega) - k_z) H_y = 0, \quad z < -\frac{1}{2},
\]

\[
\frac{\partial^2 H_y}{\partial z^2} + (k_y^2 \varepsilon_2(\omega) - k_z) H_y = 0, \quad -\frac{1}{2} < z < \frac{1}{2},
\]

\[
\frac{\partial^2 H_y}{\partial z^2} + (k_y^2 \varepsilon_3(\omega) - k_z) H_y = 0, \quad z > \frac{1}{2}.
\]

The solution the equations for areas \( \Omega_1 \) and \( \Omega_3 \) is straightforward and simple. As for the equation in the ATMF \( \Omega_2 \) we have an equation with a non-constant coefficient \( k_y^2 \varepsilon_2(\omega) = k_z \) and, therefore, cannot be solved analytically. Thus, we needed to make some additional assumption about \( \varepsilon_2(\omega) \) to proceed with a problem solution.
3. Solution of the examined problem

Based on results obtained in [29] for a metal layer, we considered a model of an electron gas in a symmetric rectangular potential well of an infinite depth. The dielectric function based on the diagonal component of the dielectric permittivity tensor has the following form for this model:

\[ \varepsilon_z(\omega, z) = \left( 1 - \frac{\omega_p^2}{\omega^2 + \omega_0^2} \right) \left( k_F^2 - \alpha^2 \right) \varepsilon(\omega, z)^2. \]  

(7)

Here \( n_e \) – electron concentration in a metal, \( \omega_p = \sqrt{\frac{4\pi n_e e^2}{m_e}} \), \( k_F \) – the Fermi wave vector [30], \( \alpha \) – quantum numbers, \( n_{\text{max}} \) – a number of bound states (energy levels).

The presence of surfaces is described by potential

\[ U(z) = \begin{cases} \infty & \text{if } z \leq 0, z \geq l, \\ 0 & \text{if } 0 < z < l, \end{cases} \]  

(8)

where \( l \) – width of a potential well. Function

\[ \psi_n(z) = \left( \frac{2}{\pi} \right)^{1/2} \sin(\alpha z) \]  

(9)

is a wave function of an electron in metal, \( \alpha = (k_x, k_y) \), \( \alpha = (\alpha_x, \alpha_y, \alpha_z) \) - the solution of the Schrödinger equation with a Dirichlet boundary conditions

\[ \frac{d^2 \psi_n(z)}{dz^2} + U(z) \psi_n(z) = W \psi_n(z), \]  

(10)

\[ \lim_{z \to \pm \infty} \psi_n(z) = 0. \]  

(11)

\( W \) – is a full energy.

Equation (10), (11) with the potential (8) yield the expression for the wave function [29]:

\[ \psi_n(z) = \left( \frac{2}{\pi} \right)^{1/2} \sin(\alpha z) \quad \text{if } 0 < z < l, \]  

(12)

\[ \psi_n(z) = 0 \quad \text{if } z \leq 0, z \geq l. \]

Quantum numbers \( \alpha \) and maximum number of bound states \( n_{\text{max}} \) are given by relations:

\[ \alpha_n = \frac{2n}{l}, \quad n_{\text{max}} = \left\lfloor \frac{k_F}{\pi} \right\rfloor \]  

(13)

where \( \lfloor \cdot \rfloor \) is a ceiling function and \( k_F \) is a magnitude of the Fermi wave vector, \( \mu \) is a chemical potential [30].

It is known that for satisfying the conditions of electroneutrality the width of the potential well due to the presence of an exponential "tail" of the electron density in dielectrics near the surface of ATMF (so called quantum spill-out effect) does not coincide with the boundaries of ATMF and depends on the penetration depth of electrons into the dielectric [29]. As shown in [30]

\[ l = l_{\text{slab}} + 2d, \]  

(14)

\[ d = \frac{\pi}{k_F} + \frac{\pi^2}{k_F}. \]  

(15)

is a function of \( k_F \) and a geometric thickness of a ATMF \( l_{\text{slab}} \). Combining together (14) and (15) for \( l \) we have [30]:

\[ l(k_F) = \frac{l}{2} + \frac{3\pi}{2k_F} + \frac{16\pi^2}{24\pi k_F l + 25\pi^2}{2k_F}. \]  

(16)

It should be mentioned that in a proposed model we consider Coulomb correlations but only through their influence on the chemical potential \( \mu \) and, consequently, the number of quantization levels \( n_{\text{max}} \) in the dielectric function of ATMF and neglecting their influence on other parts of the system.

Considering all the above we have an expression for a dielectric function \( \varepsilon_z(z, z', \omega) \) in ATMF

\[ \varepsilon_z(z, z', \omega) = \varepsilon_z(z, \omega) \delta(z - z'), \]  

(17)

In [31] we showed that the dielectric permittivity function (17) and a wave function of electron (9) significantly depend on coordinates only at the dielectric-metal and metal-dielectric interfaces. Means that (17) significantly varies from constant only at boundaries of the potential well. So, we can make the following assumption about (17)

\[ \varepsilon_z(z, z', \omega) = \varepsilon_z(l, \omega), \]  

(18)

\[ \varepsilon_z(l, \omega) = \frac{1}{l} \int_0^l \varepsilon_z(z, \omega) dz = \]  

\[ = 1 - \frac{\omega_p^2}{\omega^2 + \omega_0^2} \sum_{n=1}^{n_{\text{max}}} (k_F^2 - \alpha^2) |\phi_n(z)|^2, \]  

(19)

\[ |\phi_n(z)|^2 = \frac{1}{l} \int_0^l |\phi_n(z)| dz. \]  

(20)

In this case, we limited ourselves to considering the first equation of the system

\[ \frac{\partial^2 H_z(x)}{\partial z^2} + (k_F^2 \varepsilon_z(l, \omega) - k_F^2) H_z(x) = 0. \]  

(21)

When the wave equation has such form, the dispersion relation can be easily found and has the following form [31]

\[ e^{-\pi h z^2} = \frac{1}{k_F^2} \left( \frac{1}{z_1} + \frac{1}{z_2} + \frac{1}{z_3} \right) \]  

(22)

\[ k_F = \sqrt{k_F^2 - k_0^2}. \]  

(23)

4. Results and discussion

Study of the obtained model were conducted for such structures "SiO2/Ag/Si", "Vacuum/Ag/Si", "Vacuum/Ag/Al2O3" and, characteristics of the metal are taken from [29] and of the dielectrics from [32] (Tab. 1). The results of simulation are shown in Fig. 2-5.

<table>
<thead>
<tr>
<th>Table 1: Properties of dielectrics for considered structures.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Structure</td>
</tr>
<tr>
<td>SiO2/Ag/Si</td>
</tr>
<tr>
<td>Vacuum/Ag/Si</td>
</tr>
<tr>
<td>Vacuum/Ag/Al2O3</td>
</tr>
</tbody>
</table>

Fig. 2. Dependence of plasmon spectrum on metal thickness for "Vacuum/Ag/Si" structure (with the Coulomb correlation).

Fig. 3. Frequency spectrum for "SiO2/Ag/Si". Without the Coulomb correlations - red surface, with the Coulomb correlations - blue surface.
also largely depends on the process of propagation of SPPs waves in ATMF. It is important to note that the inclusion of Coulomb correlations also leads to a significant improvement in the frequency spectrum of SPPs compared to the classical approach and correct considering of the condition of electroneutrality for a non-interacting system of electrons. This is explained by the fact that in the case of an interacting system, the influence of quantum size effects increases significantly [29]. The oscillation pattern of the spectrum with an increase in the film thickness becomes less noticeable Fig.2, which coincides with the corresponding behavior of a chemical potential [30], the decay speed of the oscillation peaks also largely depends on the Wigner-Seitz radius. It is worth noting the dependence of the spectrum on the dielectrics surrounding ATMF which can be seen on the Wigner-Seitz surface, with the Coulomb correlations - blue surface.

5. Conclusions

The results show that even a rather “rough” consideration of the Coulomb correlations, namely, their influence on the chemical potential leads to significant changes in the frequency spectrum of SPPs waves in ATMF. This is explained by the fact that in the case of an interacting system, the influence of quantum size effects increases significantly [29]. The oscillation pattern of the spectrum with an increase in the film thickness becomes less noticeable Fig.2, which coincides with the corresponding behavior of a chemical potential [30], the decay speed of the oscillation peaks also largely depends on the Wigner-Seitz radius. It is worth noting the dependence of the spectrum on the dielectrics surrounding ATMF which can be seen by comparing the data given in Fig.3,4,5.

It is important to note that the inclusion of Coulomb correlations also leads to a significant improvement in the agreement with the experimental data Tab.2, and this proves the need to take these correlations into account when modeling the processes of propagation of SPPs waves in ATMF.

### Table 2: Comparison of the model simulation with an experimental data.

<table>
<thead>
<tr>
<th>$k_{x} \ (\text{nm}^{-1})$</th>
<th>$A \ (\text{eV})$</th>
<th>$B \ (\text{eV})$</th>
<th>$E \ (\text{eV})$</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0278</td>
<td>0.696</td>
<td>1.288</td>
<td>0.62</td>
</tr>
<tr>
<td>0.0487</td>
<td>0.889</td>
<td>1.789</td>
<td>0.8</td>
</tr>
</tbody>
</table>

**Fig. 4.** Frequency spectrum for “Vacuum/Ag/Si”. Without the Coulomb correlations - red surface, with the Coulomb correlations - blue surface.

**Fig. 5.** Frequency spectrum for “Vacuum/Ag/Al$_2$O$_3$”. Without the Coulomb correlations - red surface, with the Coulomb correlations - blue surface.

### 6. References

4. J. Liu, M. Jalali et al., Analyst - The Royal Society of Chemistry, **145** 2, 364-384 (2020)
14. S. V. Borisikina, T. A. Cooper et al., Advances in Optics and Photonics, **9**, 4, 775-827 (2017)
21. V. G. Achanta, Reviews in Physics, **5**, 100041 (2020)
27. Gao, Xi and Cui, Tie Jun, Nanotechnology Reviews, **5**, 1, 297-303, (2010)
35. V. G. Achanta, Reviews in Physics, **5**, 100041 (2020)
41. Gao, Xi and Cui, Tie Jun, Nanotechnology Reviews, **5**, 1, 297-303, (2010)