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Interaction of Silver Nanoparticles with a Substrate Under Plasmonic Resonance Conditions

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***Abstract*—The interaction between metal nanoparticles and substrates under plasmonic resonance conditions plays a crucial role in various optical applications. In this study, we research the impact of substrate material on the optical response of silver nanoparticles under surface plasmon resonance conditions. Using theoretical modeling based on the quasi-static dipole approximation, we explore how the dielectric constant of the substrate affects the extinction cross-section spectra of silver nanoparticles as a function of nanoparticle size and distance from the substrate surface. The calculation results show significant shifts in the extinction peak and enhancements in the extinction cross-section values when considering different substrate materials, including cellulose, indium tin oxide and silver. It was found that substrates with higher dielectric constants induce larger shifts in the extinction peak towards longer wavelengths and lead to increased extinction cross-section values at the operating wavelength. Furthermore, it was found that the orientation of the external electric field relative to the substrate surface influences the magnitude of these shifts. The results of the study show that while changing the size of the nanoparticles has minimal effect on the position of the extinction peak, increasing nanoparticle size significantly enhances the maximum extinction cross-section values. Additionally, varying the distance between the nanoparticles and the substrate surface causes shifts in the extinction spectra, with larger shifts observed for substrates with higher dielectric constants. These findings provide valuable insights into the design and optimization of plasmonic structures for various optoelectronic applications. By understanding the nanoparticle-substrate interactions and their optical properties, our theoretical study aids in the prediction of optical responses and the development of tailored optical structures for enhanced productivity of their usage. Overall, this study highlights the importance of substrate material selection and nanoparticle-substrate interactions in engineering plasmonic systems for advanced optical applications, paving the way for the design of efficient and optimized optoelectronic devices and sensors.**

Keywords — silver nanoparticles; extinction cross section; substrate; localized surface plasmon resonance.

I. INTRODUCTION

Metal nanoparticles are characterized by extraordinary optical properties in the visible wavelength range [1]. Due to precise nanoscale control and the presence of free electrons in the metal nanoparticles, strong light-matter interaction occurs [2]. As a result, the conduction electrons in nanoparticles oscillate at the frequency of the external field, which is known as localized surface plasmon resonance [3]. Localized surface plasmons can cause strong scattering and absorption of electromagnetic radiation, which enhances the local electromagnetic fields surrounding the nanoparticles, providing a wide field for research and applications. The properties of localized surface plasmon resonance are widely used in various optoelectronic devices [4], biological and chemical detection [5], Raman amplification [6], etc.

The majority of both theoretical and experimental studies in the field of plasmonics are mainly focused on noble metals nanostructures [7-9], and less often on semi-metals [10, 11]. All of them exhibit plasmon resonance in the visible and near-infrared spectral ranges, which has led to their widespread practical use. The wavelength of localized surface plasmon resonance can be changed over such a wide range of wavelengths by controlling the size, shape, and dielectric properties of plasmonic nanoparticles [1, 12]. Over the past decades, considerable attention has been paid to the study of the effect of the shape of plasmonic nanoparticles on

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their extinction spectrum [13, 14]. Many scientific papers have been published on the fabrication and application of nanoparticles in the shape of triangles [15], cubes [16], ellipsoids [17], stars [18], etc. However, spherical plasmonic nanoparticles are still the most studied and used.

One of the critical parameters affecting the resulting optical response of plasmonic nanoparticles is the dielectric constant of the substrate [19]. The presence of a substrate near the surface of a nanoparticle affects its optical properties due to the induced field in the substrate, although this effect is often neglected in the literature [20]. The gap between the nanoparticle and the substrate can be a source of near-field enhancement [21, 22], which has an impact on the resultant gain, for example in Surface-Enhanced Raman Spectroscopy (SERS). Therefore, it is very important to have a clear understanding and research the interaction of plasmonic nanoparticles with the substrate under resonance conditions.

The interaction of nanoparticles with dielectric substrates can be considered using a quasi-static approximation if the nanoparticle size is significantly smaller than the radiation wavelength [23]. In the case of a metallic substrate, numerical approaches need to be used since surface plasmon polaritons will be generated and propagate along the substrate surface. These methods have shown good correlation with the experimental results and numerical approaches [24]. However, there are few studies in the literature on the influence of the substrate material on the plasmonic characteristics of nanoparticles.

Understanding the plasmonic coupling between nanoparticles and their substrate is fundamental for designing and optimizing optoelectronic structures. In this paper we present the results of theoretical studies of the effect of the substrate material on the extinction cross section spectra of silver nanoparticles under surface plasmon resonance conditions as a function of nanoparticle size and distance from the surface, with the aim of applying them to SERS. The calculations have been carried out by means of the image dipole approach within the quasi-static approximation.

II. Mathematical model

The quasi-static dipole approximation is reasonable for spherical or ellipsoidal particles with dimensions much smaller than the incident wavelength. At the boundary where the nanoparticle size is much smaller than the incident wavelength, it is exposed to a mostly homogeneous field throughout the occupied space. Thus, the interaction of light with a nanoparticle can be described in terms of electrostatics. This approach is known as quasi-static approximation [20]. In this approximation, incident homogeneous electric field **E** leads to formation of electric dipole moment inside the nanoparticle.

When an external homogeneous electric field $\mathbf{E}_\perp$ is applied perpendicularly to a nanoparticle on a substrate, the induced charges at the interface between the substrate and the environment change the resulting electric field, which polarizes the sphere. This total electric field has a non-homogeneous character, which leads to an effective multipole polarization of the sphere [25, 26]. Although higher multipole moments change the resulting optical response of the sphere, the sphere's dipole moment determines the main characteristics of the optical spectra. The perpendicular component of the dipole moment is equal:

$$\mathbf{p}_\perp = \varepsilon_0 \varepsilon_m \alpha_\perp^{eff} \mathbf{E}_\perp, \quad (1)$$

where ε_0 is the absolute dielectric constant, ε_m is the dielectric constant of the environment, and $\alpha_\perp^{eff}$ is the perpendicular component of the effective polarization tensor of the nanoparticle.

In order to determine the influence of the substrate on the optical response of the nanoparticle, we consider the dipole representation approach. In this model, the dipole moment associated with the nanoparticle induces electric charges on the substrate, which can be represented by a new dipole moment, namely the image of the dipole moment inside the substrate. It can be assumed that this is a system of two electromagnetically coupled spheres, the sphere and its image in the substrate.

Thus, the total dipole moment of the two dipoles will determine the effective polarizability of the system of the two coupled spheres. Then, the effective polarizability of the sphere over the substrate is determined as follows [25]:

$$\alpha_\perp^{eff} = \left[\frac{V(\varepsilon(\lambda)-\varepsilon_m)}{\varepsilon_m+L_\perp(\varepsilon(\lambda)-\varepsilon_m)}\right]\frac{2\varepsilon_s(\lambda)}{(\varepsilon_s(\lambda)+\varepsilon_m)}, \quad (2)$$

where V is the volume of the nanoparticle, $\varepsilon_s(\lambda)$ is the dielectric constant of the substrate.

The electrical depolarization coefficient $L_\perp$ is equal:

$$L_\perp = \frac{1}{3}\left[1-\frac{1}{4}\left(\frac{d}{R}\right)^3\left(\frac{\varepsilon_s(\lambda)-\varepsilon_m}{\varepsilon_s(\lambda)+\varepsilon_m}\right)\right], \quad (3)$$

where d is the distance between the particle and the substrate, R is the radius of the nanoparticle.

In the case when the external electric field component $\mathbf{E}_\parallel$ is applied in parallel to the substrate, the effective polarizability of two coupled dipoles is determined by the equation:

$$\alpha_\parallel^{eff} = \left[\frac{V(\varepsilon(\lambda)-\varepsilon_m)}{\varepsilon_m+L_\parallel(\varepsilon(\lambda)-\varepsilon_m)}\right]\frac{2\varepsilon_s(\lambda)}{(\varepsilon_s(\lambda)+\varepsilon_m)}, \quad (4)$$

where the depolarization coefficient L_‖ is equal:

$$L_\perp = \frac{1}{3}\left[1-\frac{1}{8}\left(\frac{d}{R}\right)^3\left(\frac{\varepsilon_s(\lambda)-\varepsilon_m}{\varepsilon_s(\lambda)+\varepsilon_m}\right)\right]. \quad (5)$$

Dissipative radiation losses of a nanoparticle are the result of absorption and scattering, which are studied by their cross-sections C_{abs} and C_{scat}, respectively. In

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the quasi-static approximation, C_{abs} and C_{scat} are determined by the following formulas:

$$C_{abs} = kIm(\alpha_{\perp,\parallel}^{eff}), \quad (6)$$

$$C_{scat} = \frac{k^4}{6\pi}\left|\alpha_{\perp,\parallel}^{eff}\right|^2, \quad (7)$$

where k is the incident wave vector modulus.

III. RESULTS AND DISCUSSION

The extinction cross-sections of spherical silver nanoparticles deposited on three different surfaces (cellulose, indium tin oxide (ITO), and silver) were calculated using the quasi-static dipole approximation (Fig. 1). Since this approximation is only valid for particles much smaller than the working wavelength [27], the size of the nanoparticles studied was used for calculations in the range 5 to 55 nm. The refractive index of cellulose was calculated using the Sellmeier dispersion formula from [28]. The dielectric constant of ITO was calculated by the description of the experimental data from [29] with a seventh order polynomial. The dielectric constant of silver was calculated by using the analytical relations as given in [30]. For the initial calculations, the distance between the nanoparticles and the substrate was assumed to be 1 nm and the radius of the nanoparticles was assumed to be 55 nm. Since the size of such a nanoparticle is rather large, we calculated the extinction cross section of the nanoparticle, which is determined by the sum of the absorption and scattering cross sections. The environment in which the nanoparticles were placed was assumed to be the air, which has a refractive index of 1.028.

All calculations were performed for three cases, including the calculation of nanoparticle extinction cross sections without considering the substrate influence and including the substrate influence when the external electric field component **E** is applied both perpendicularly and parallel to the surface of the substrate containing nanoparticles. Modeling results showed that the dielectric constant of the nanoparticle-deposited substrate is a key parameter that significantly affects the resulting optical response. Without considering the influence of the substrate, the peak of the extinction cross section is at the wavelength of 0.337 µm, and the maximum extinction value reaches 14 cm^{-2}. Taking into account the dielectric constant of the substrate material, we obtain a shift of the extinction peak to the long wavelength region for both variants of the applied external electric field **E**$_\perp$ and **E**$_\parallel$. The higher the dielectric constant of the substrate material, the greater the nanoparticle extinction maximum shift. In the case where the external electric field component E is applied perpendicular to the nanoparticle, the shift is 9 nm for the cellulose substrate, 29 nm for the ITO, and 61 nm for the silver. The shift is 4 nm for the cellulose substrate, 13 nm for ITO, and 33 nm for silver when the external electric field component E is applied in parallel.For the three substrate materials studied, the maximum value of nanoparticle extinction cross section is larger when the external electric field component E is applied perpendicularly. In conclusion, using a substrate with a higher refractive index to deposit silver nanoparticles not only shifts the extinction peak, but also significantly increases the value of the extinction cross section.

It should be noted that changing the size of the nanoparticle has virtually no effect on the position of the extinction peak for the three substrate materials studied and when they are omitted (Fig. 2).

During calculations, the nanoparticle size varied from 5 to 55 nm. Only the maximum value of the absorption cross section increased with increasing nanoparticle size. When the nanoparticle radius increased from 30 to 55 nm, the maximum extinction cross section increased more than 30 times for both components of the applied external electric field in all cases of substrate materials.

In contrast to changing the nanoparticle radius, changing the distance between the nanoparticle and the substrate surface does not lead to a significant increase in the extinction cross section, but causes the plasmon peak to shift to the long wavelength region (Fig. 3). The distance between the nanoparticle and the substrate surface was varied from 0.5 to 2 nm, since at larger distances the particle can be considered as unbound to the substrate.

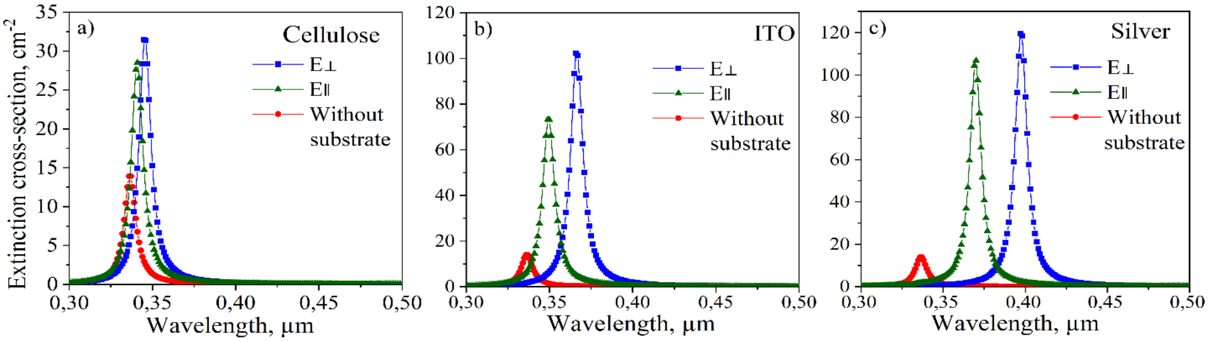

Fig. 1 Extinction cross sections of nanoparticles placed on cellulose (a), ITO (b) and silver (c) substrates. The radius of the nanoparticles is 55 nm. The distance between the nanoparticle and the substrate is 1 nm. The particles are placed in an air environment.

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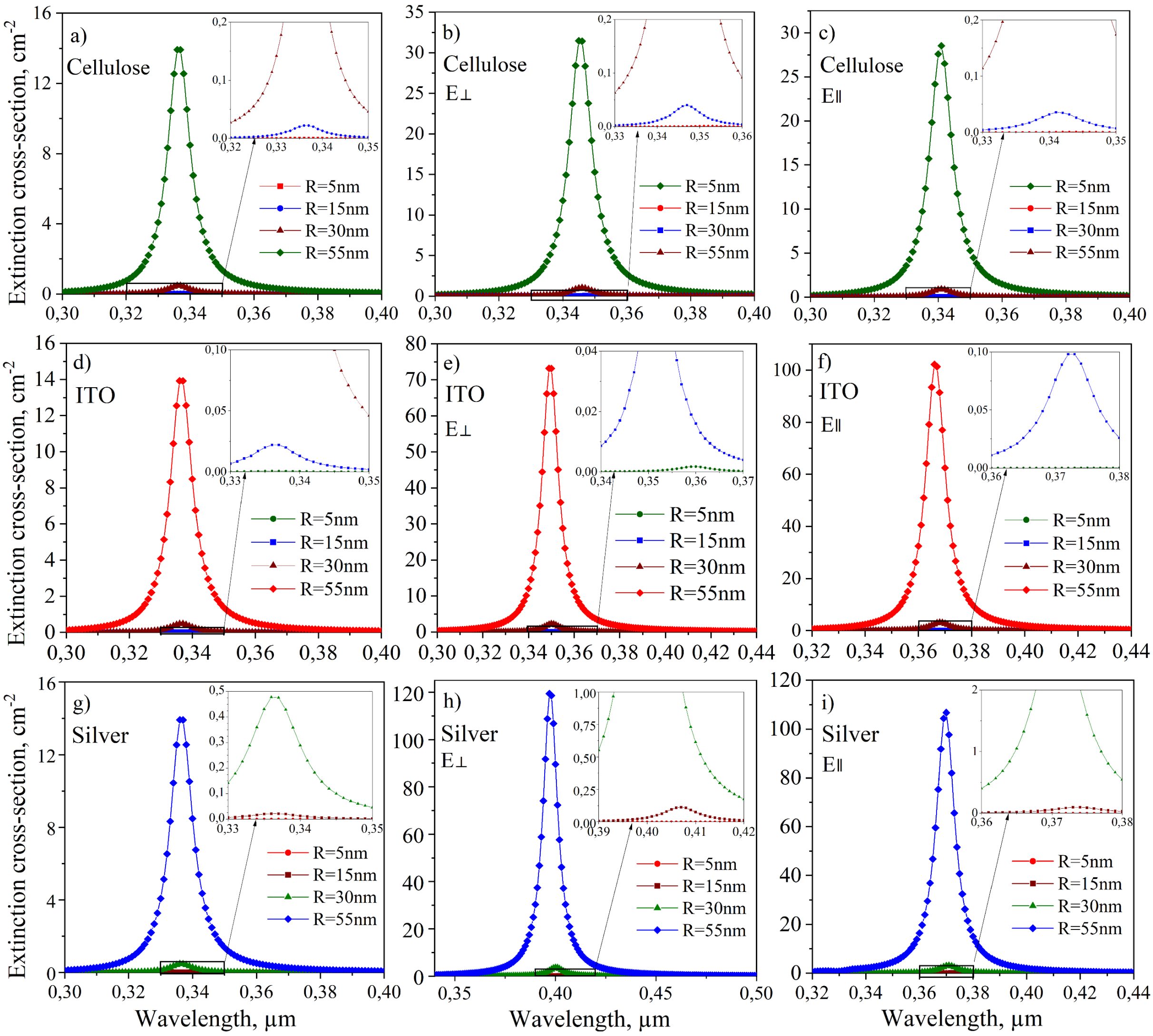

Fig. 2 Extinction cross sections of nanoparticles placed on cellulose (a-c), ITO (d-f), and silver (g-i) substrates. The radius of the nanoparticles varied from 5 to 55 nm. The distance between the nanoparticle and the substrate was 1 nm. The particles are placed in the air.

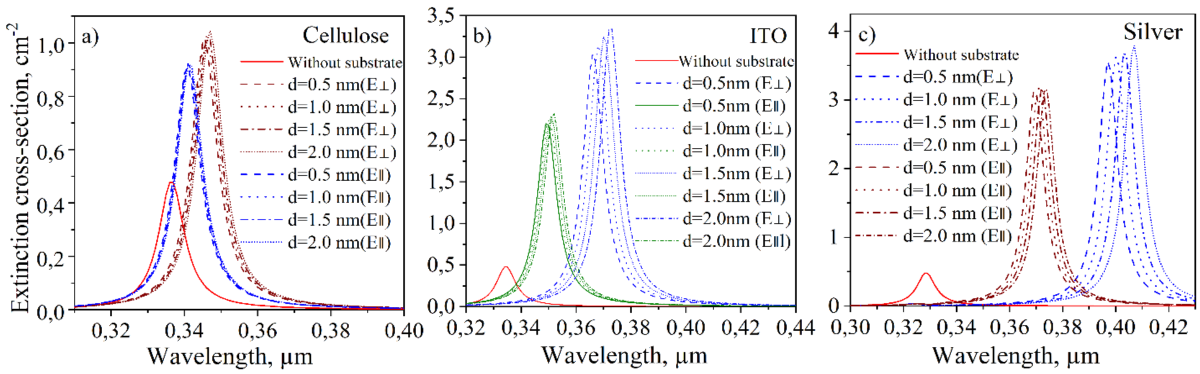

Fig. 3 Extinction cross sections of nanoparticles placed on cellulose (a), ITO (b), and silver (c) substrates. The nanoparticle radius is 30 nm. The distance between the nanoparticle and the surface varied from 0.5 to 2 nm. The particles are placed in the air.

In the case of the cellulose substrate, the shift of the extinction spectra is insignificant, in the order of 1 nm. For the ITO substrate, a shift of approximately 5nm and for the silver substrate, a shift of 10nm can be seen for a 1.5nm change in the distance of the nanoparticle from the substrate surface. It should be noted that these figures refer to the case when the external electric field component E is applied in a perpendicular direction. For all substrate materials studied, when the external electric field component E is applied in parallel, the shift is much smaller. Thus, in the design and fabrication of silver

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nanoparticle-based optical structures, the dielectric constant of the substrate material is of key importance.

Conclusions

The extinction cross-sectional spectra of silver nanoparticles under surface plasmon resonance conditions, depending on the nanoparticle size and the distance between the nanoparticles and the substrate surface, have been calculated considering the effect of the presence of the substrate on which they are deposited based on the quasi-dipole approximation. The nanoparticle substrates used were cellulose, ITO and silver. The results of numerical experiments have shown that the material (permittivity) of the substrate with deposited silver nanoparticles is crucial for the design and fabrication of optical structures based on them. It was found that a significant shift of the peak of the extinction cross section, as well as an increase of its value at the operating wavelength, is mainly caused by surfaces with high dielectric constant.

The calculations presented here provide insight into the interaction between nanoparticles and the surface on which they are deposited, and will be useful in the prediction of optical properties and in the design of optimized optical structures for many technological applications, including SERS.

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Взаємодія срібних наночастинок з підкладкою в умовах плазмонного резонансу

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***Анотація*—Взаємодія між наночастинками металів і підкладками в умовах плазмонного резонансу відіграє вирішальну роль у різних оптичних застосуваннях. У цій роботі ми досліджуємо вплив матеріалу підкладки на оптичний відгук наночастинок срібла в умовах поверхневого плазмонного резонансу. Використовуючи теоретичне моделювання на основі квазістатичного дипольного наближення, ми дослідили, як діелектрична проникність підкладки впливає на спектри перерізу екстинкції наночастинок срібла в залежності від розміру наночастинок і відстані від поверхні підкладки. Проведені розрахунки показують значні зсуви піку екстинкції та збільшення перерізу екстинкції при розгляді різних матеріалів підкладок, зокрема целюлозу, оксид індію та олова і срібло. Було виявлено, що підкладки з більшою діелектричною проникністю спричиняють більший зсув піку екстинкції в бік довших довжин хвиль і призводять до збільшення значень перерізу екстинкції на робочій довжині хвилі. Більше того, було встановлено, що орієнтація зовнішнього електричного поля відносно поверхні підкладки впливає на величину цих зсувів. Результати дослідження показують, що хоча зміна розміру наночастинок має мінімальний вплив на положення піку екстинкції, збільшення розміру наночастинок значно збільшує максимальні значення перерізу екстинкції. Крім того, зміна відстані між наночастинками і поверхнею підкладки спричиняє зсуви в спектрах екстинкції, причому більші зсуви спостерігаються для підкладок з більшими значеннями діелектричної проникності. Ці результати дають цінну інформацію для проектування та оптимізації плазмонних структур для різних оптоелектронних застосувань. Розуміння взаємодії наночастинок з підкладкою та їх оптичні властивості допомагає прогнозувати оптичні відгуки і розробляти оптичні структури для покращення продуктивності їх використання. Загалом, це дослідження підкреслює важливість вибору матеріалу підкладки та взаємодії наночастинок з підкладкою в інженерних плазмонних системах для передових оптичних застосувань, прокладаючи шлях до розробки ефективних і оптимізованих оптоелектронних пристроїв і сенсорів.**

Ключові слова — наночастинки срібла; переріз екстинкції; підкладка; локалізований поверхневий плазмонний резонанс.