



Iryna Yaremchuk,
Tetiana Bulavinets,
Yuriy Smachylo,
Yuriy Mysiuk,
Volodymyr Fitio,
Pavlo Stakhira

MODELING OF THE TUNABLE PLASMONIC PROPERTIES OF SPHERICAL AND ELLIPSOIDAL SILVER NANOPARTICLES IN THE MATRIX OF AN ORGANIC SEMICONDUCTOR

The object of research is the tunable plasmonic properties of spherical and ellipsoidal silver nanoparticles in the organic semiconductor matrix. The average absorption cross-sections, scattering cross-sections, and optical radiation efficiency of spherical and ellipsoidal silver nanoparticles have been simulated. The long-wave statistical approach has been used to model the optical parameters of the assembled spherical and ellipsoidal nanoparticles. Statistical averaging is used here, where absorption and scattering are considered from an "effective" particle with statistical properties. This approach avoids complex calculations considering the details of the spectral characteristics of single nanoparticles of different shapes. Taking into account the fact that the nanocomposite matrix will contain ensembles of spherical nanoparticles of different sizes, the peak of their absorption and scattering cross sections will be shifted to the short wavelength region of the spectrum compared to ensembles of the same spherical nanoparticles. In addition, there is a slight increase in the absorption cross-section and a decrease in the scattering cross-section, confirming the presence of smaller nanoparticles. A study was made of a composite material containing a randomly dispersed ensemble of silver ellipsoidal nanoparticles of the same and different shapes and sizes in an organic semiconductor matrix. An ensemble of identical ellipsoidal nanoparticles is characterized by the presence of two plasmon peaks, which corresponds to the characteristics of a single ellipsoidal nanoparticle. A completely different situation is observed if to consider that the nanocomposite will contain an ensemble of ellipsoidal nanoparticles of different shapes and sizes. Such nanoparticles will be characterized by a plasmon peak for both the absorption and scattering cross-sections. This can be explained by the fact that as the size of ellipsoidal nanoparticles decreases, the distance between the peaks responsible for the longitudinal and transverse modes of plasmon excitation decreases. An increase in the shape distribution leads to a broadening of the absorption and scattering cross-section spectra. The efficiency of the optical radiation increases as the size distribution increases. It is shown that a change in the refractive index of an organic semiconductor matrix mainly affects only the value of the scattering cross-section of an ensemble of ellipsoidal nanoparticles dispersed in it. This research is a preliminary step to studying the influence of these particles on the properties of organic light-emitting structures.

Keywords: silver, spherical and ellipsoidal nanoparticles, plasmon resonance peak, absorption cross-section, scattering cross-section, nanoparticles assembly.

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1. Introduction

The absorption and scattering of electromagnetic radiation by plasmonic nanoparticles have attracted considerable interest due to its potential to enhance the light-matter interaction in a number of applications. Nanoparticles improve light coupling in solar cells through efficient scattering [1, 2] and can increase the radiative recombination rate or internal quantum efficiency of organic light-emitting diodes [3, 4]. This is explained by the presence of a localized surface

plasmon resonance in the nanoparticles. It is associated with the coherent oscillation of the free electrons of the metal under the action of an external electromagnetic field. The plasmon resonance leads to a significant enhancement of the electromagnetic field around the nanoparticles and depends on the size and shape of the nanoparticles as well as the properties of the medium, which can be used to modify the optical properties of materials [5].

Noble metal nanoparticles, especially silver nanoparticles, have recently attracted considerable interest due to their

unique optical properties in the visible spectrum [6, 7]. Molecules of organic components located near the surface of metal nanoparticles are exposed to local electromagnetic fields. Accordingly, the radiative and non-radiative rates of intramolecular electron transitions either increase or decrease depending on the distance between the nanoparticles and the organic molecules [8, 9]. The enhancement of the efficiency of organic phosphorescent light-emitting diodes is a result of the coupling between the localized plasmon resonance of silver nanoparticles and excitons in the emitting layer [10, 11]. The overlap of the localized surface plasmon resonance spectra of the silver nanoparticles and the emitting device is the main factor that enhances its brightness [12]. The semiconducting properties of some coupled polymers can be enhanced by incorporating various metal nanoparticles such as gold, platinum and silver into polymer matrices [13, 14].

Thus, plasmonic silver nanoparticles exhibit many new phenomena in their inherent electromagnetic interactions, which can be used to control the mobility of charge carriers in semiconductor materials, increase the efficiency of light-emitting devices, etc. However, most of the work is usually aimed at studying the effects related to the absorption or scattering of light by single nanoparticles [15]. In practice, we are dealing with particle ensembles rather than single particles. Ensembles of nanoparticles have different sizes, shapes, and compositions. They can also constantly change their spatial position and orientation. All these factors determine the characteristics of the interaction of light with particle ensembles and the medium in which they are dispersed.

Considering the above issues, *the aim of research* is to identify the electromagnetic absorption, scattering, and optical radiative efficiency of randomly distributed silver nanoparticles of spherical and ellipsoidal shapes of different sizes as a preliminary step to studying the influence of these particles on the properties of organic light-emitting structures.

2. Materials and Methods

The computational model for studying the optical properties of an ensemble of metal nanoparticles with chaotic orientation in a dielectric medium is based on a statistical approach in the long-wave approximation [16]. In the long-wave approximation, the effective dielectric function of any ensemble of nanoparticles can be represented in an integral form [17]. Statistical averaging is used here, taking into account the absorption and scattering from an "effective" particle whose properties are statistical in nature. This approach avoids complex calculations that take into account the details of the spectral characteristics of individual nanoparticles of different shapes. In addition, the model used gives a more adequate description of the absorption and scattering of light for the corresponding experimental sample than that given by the usual calculations for idealized systems of the same particles, whose shape is strictly specified. It is known that the spectra of particles of any complex shape can be approximated by appropriately averaged spectra of ellipsoidal particles [18], so let's consider in detail the analytical expressions for determining the spectral characteristics of an ensemble of randomly placed ellipsoids.

The average absorption and scattering cross sections for an ensemble of randomly placed identical ellipsoids (spheres) can be written as follows:

$$\langle C_{abs} \rangle = k \langle V \rangle \text{Im} \langle \alpha \rangle, \quad (1)$$

$$\langle C_{scat} \rangle = \frac{k^4 \langle V^2 \rangle}{6\pi} |\langle \alpha \rangle|^2, \quad (2)$$

where $k = \frac{2\pi\sqrt{\epsilon_h}}{\lambda}$ is the wavenumber of the medium surrounding the nanoparticle with a dielectric constant ϵ_h , $\langle V \rangle$ is the average volume of the nanoparticle, and $\langle \alpha \rangle$ is the average (effective) polarization value of the nanoparticle, which is determined as follows:

$$\text{Im} \langle \alpha \rangle = \frac{1}{3} \text{Im} (\alpha_1 + \alpha_2 + \alpha_3), \quad (3)$$

$$|\langle \alpha \rangle|^2 = \frac{1}{3} (|\alpha_1|^2 + |\alpha_2|^2 + |\alpha_3|^2). \quad (4)$$

Polarization along the j for a single volume is determined by the formula:

$$\alpha_j = \frac{\epsilon_m - 1}{1 + L_j (\epsilon_m - 1)}, \quad (5)$$

where ϵ_m is the dielectric constant of the nanoparticle material, L_j is the geometric parameter of the ellipsoid, which is equal to:

$$L_i = \frac{a_1 a_2 a_3}{2} \int_0^\infty \frac{dq}{(a_i^2 + q) f(q)}, \quad (6)$$

where $f(q) = \sqrt{(q + a_1^2)(q + a_2^2)(q + a_3^2)}$, $a_1 a_2 a_3$ are the semi-axes of the ellipsoid.

It should be noted that only two of the three described geometric factors are independent, since $\sum L_i = 1$, and satisfy the condition $L_1 < L_2 < L_3$. In the case of the sphere $L_1 = L_2 = L_3$.

The effective absorption cross-section for an ensemble of nanoparticles with different ellipsoidal shapes is equal:

$$\begin{aligned} \langle C_{abs} \rangle &= k \langle V \rangle \text{Im} \langle \alpha \rangle = \\ &= \frac{2}{\Delta^2} k \langle V \rangle \text{Im} \left[\left(A + \frac{1}{3} + \frac{2}{3} \Delta \right) \text{Im} \left(\frac{A + \frac{1}{3} + \frac{2}{3} \Delta}{A + \frac{1}{3} - \frac{2}{3} \Delta} \right) \right], \end{aligned} \quad (7)$$

where $A = 1/(\epsilon_m - 1)$, Δ is a shape distribution that changes from 0 to 1.

The effective scattering cross-section of an ensemble of nanoparticles with different ellipsoidal shapes is determined as follows:

$$\begin{aligned} \langle C_{scat} \rangle &= k^4 \langle V \rangle^2 \langle \tilde{C}_{scat} \rangle = \\ &= \frac{k^4 V^2}{\Delta^2} \left\{ 2r_+ [\arctan(r_+) - \arctan(r_-)] - \ln \left(\frac{1+r_+^2}{1-r_-^2} \right) \right\}, \end{aligned} \quad (8)$$

where

$$r_+ = \frac{\eta + 2\Delta}{\theta}, \quad r_- = \frac{\eta - \Delta}{\theta}, \quad (9)$$

$$\eta = 1 + 3Re(\beta), \quad (10)$$

$$\theta = 3Im(\beta), \quad (11)$$

$$\beta = \frac{1}{(\epsilon_m - 1)}. \quad (12)$$

A good reference point for the selection of metal nanoparticles to increase the efficiency of optoelectronic devices is the optical radiation efficiency. This parameter indicates the fraction of energy that is lost and re-radiated by the particle and is defined as follows:

$$\eta_{Eff} = \frac{\langle C_{scat} \rangle}{\langle C_{scat} \rangle + \langle C_{abs} \rangle}. \quad (13)$$

The described theoretical methods of studying the optical parameters of an ensemble of nanoparticles make it possible to assess the influence of changes in the shape of the nanoparticles and the parameters of the surrounding medium on their optical response and to effectively adjust the resonance of the surface plasmons, shifting them to the desired region of the spectrum.

3. Results and Discussion

Let's first consider a composite material containing identical spherical nanoparticles randomly dispersed in the matrix. Silver was chosen as the nanoparticle material and

the dielectric constant value was taken from [19]. The organic semiconductor Alq3 with a refractive index of 1.77 was used as the matrix [20]. It is one of the most common and commercially available organic semiconductors. The radius of the nanoparticles was chosen to be 50 nm. The plasmon peak for an ensemble of such nanoparticles is at a wavelength of 0.433 μm (Fig. 1, *a, b*, black curve), which coincides with the position of the peak of a single spherical nanoparticle. A different picture is observed in the case of a finely dispersed nanocomposite, in which the inclusions are slightly non-spherical and distributed in shape, that is, the distribution of the shape $\Delta=0.1$ (Fig. 1, *a, b*, red and blue curves).

Taking into account the fact that the nanocomposite matrix will contain ensembles of spherical nanoparticles of different sizes, it can be seen that the peak of their absorption and scattering cross sections is shifted by 80 nm towards the short wavelength region of the spectrum. There is also a slight increase in the absorption cross section and a decrease in the scattering cross section, confirming the presence of smaller nanoparticles. It is known that nanoparticles with a smaller surface area are characterized by higher absorption compared to scattering and vice versa. At the same time, for both types of nanoparticle ensembles, the efficiency of optical radiation is the same from 0.342 μm and further into the red region of the spectrum. This suggests that such ensembles of nanoparticles could be used throughout the visible wavelength range.

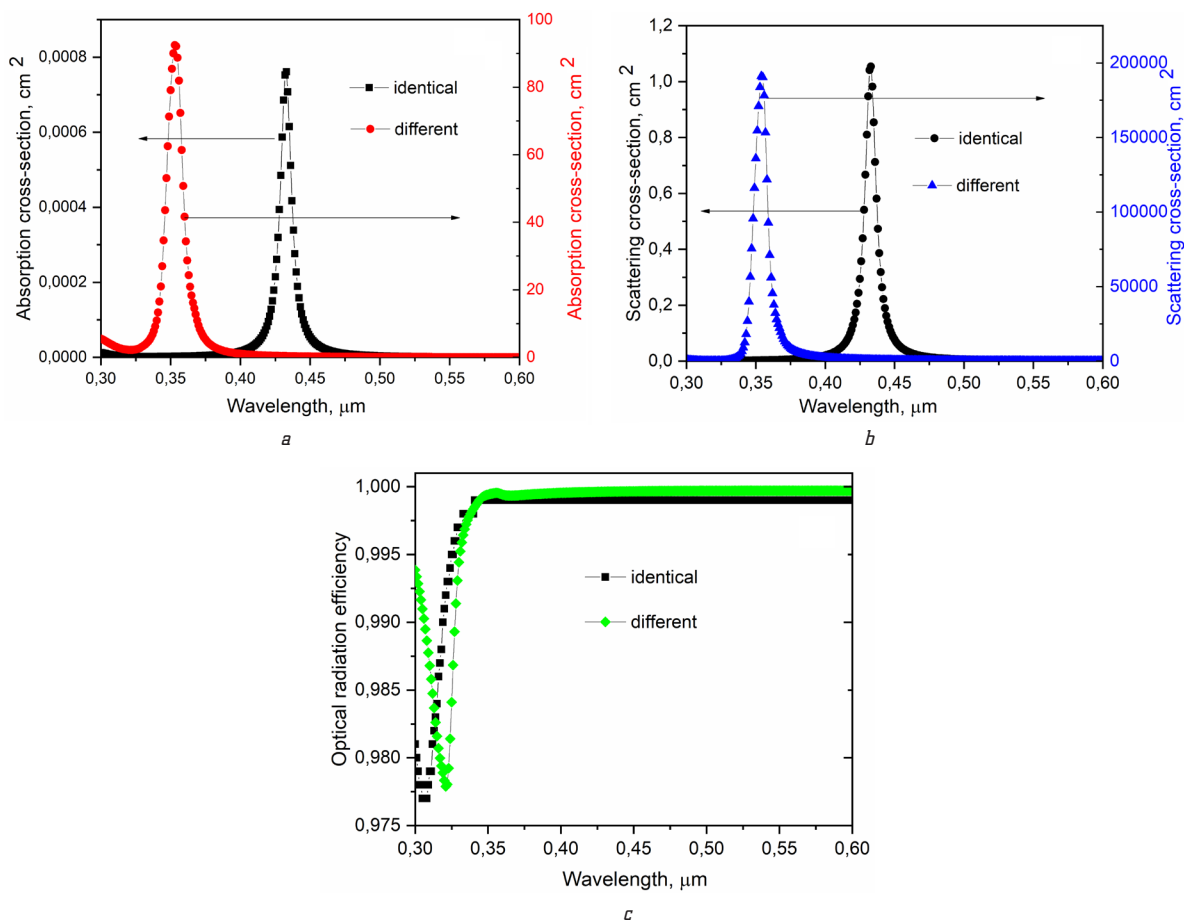


Fig. 1. Spectra of an ensemble of spherical nanoparticles of identical and different size and shape ($\Delta=0.1$): *a* – absorption cross of sections; *b* – scattering; *c* – optical radiation efficiency

Recently, in addition to size, the shape of nanoparticles has become a key regulator for the exploitation of their plasmonic properties. The absorption process can cause spontaneous reorientation of non-spherical particles [21]. Accordingly, attention has been drawn to ellipsoidal nanoparticles as they are characterized by longitudinal and transverse modes of plasmon excitation. Consider the case of a composite material containing an ensemble of identical ellipsoidal nanoparticles randomly dispersed in an organic semiconductor matrix. For the purpose of the calculations, it is assumed that the major axis of the ellipsoid is $\alpha_1=50$ nm and the two minor axes are equal to each other and are $\alpha_2=\alpha_3=40$ nm (Fig. 2).

An ensemble of identical ellipsoidal nanoparticles placed in an organic matrix is characterized by the presence of two plasmon peaks at a wavelength of 0.420 μm and 0.470 μm , corresponding to the calculated properties for a single ellipsoidal nanoparticle. The optical radiation efficiency of such an ensemble of nanoparticles is characterized by a minimum value at a wavelength of 0.450 μm .

A completely different situation is observed if to consider that the nanocomposite will contain ellipsoidal nanoparticles of smaller size and different shapes (Fig. 3). Such nanoparticles will be characterized by a plasmon peak for both the absorption and scattering cross sections. This can be explained by the fact that as the size of the ellipsoidal nanoparticles decreases, the distance between the peaks responsible for the longitudinal and transverse modes of plasmon excitation decreases.

Thus, the plasmon spectra of nanoparticles of different sizes will overlap. It is worth noting that even a small change in the shape of nanoparticles ($\Delta=0.1$ will not only

lead to the formation of a peak, but also to a shift of the spectrum towards the short wavelength region of the spectrum compared to an ensemble of identical nanoparticles). An increase in the shape distribution leads to a broadening of the absorption and scattering cross section spectra.

Calculations show that if the composite contains nanoparticles with a large size distribution ($\Delta=0.9$), such an ensemble of nanoparticles will absorb and scatter light in almost the entire visible range. However, the intensity of the absorption will decrease by almost five times and the scattering will also decrease by a quarter. The efficiency of optical radiation increases as the size distribution increases.

As mentioned above, the work considered ensembles of nanoparticles dispersed in a commercially available organic semiconductor matrix with a refractive index of 1.77. However, organic semiconductor materials that can be successfully combined with plasmonic nanoparticles are now being intensively synthesized and researched. In general, the refractive index of these materials is between 1.70 and 1.80. Spectra of absorption cross-sections, scattering and efficiency of optical radiation of an ensemble of ellipsoidal nanoparticles as a function of the refractive index of an organic semiconductor are shown in Fig. 4.

Changing the refractive index of the organic semiconductor matrix will mainly affect the size of the scattering cross section of the ensemble of ellipsoidal nanoparticles dispersed in it. An increase in the refractive index will lead to an increase in the amplitude of the scattering cross section, but will not affect the shape of the spectrum. The efficiency of the optical radiation is virtually unaffected by the choice of organic semiconductor matrix material.

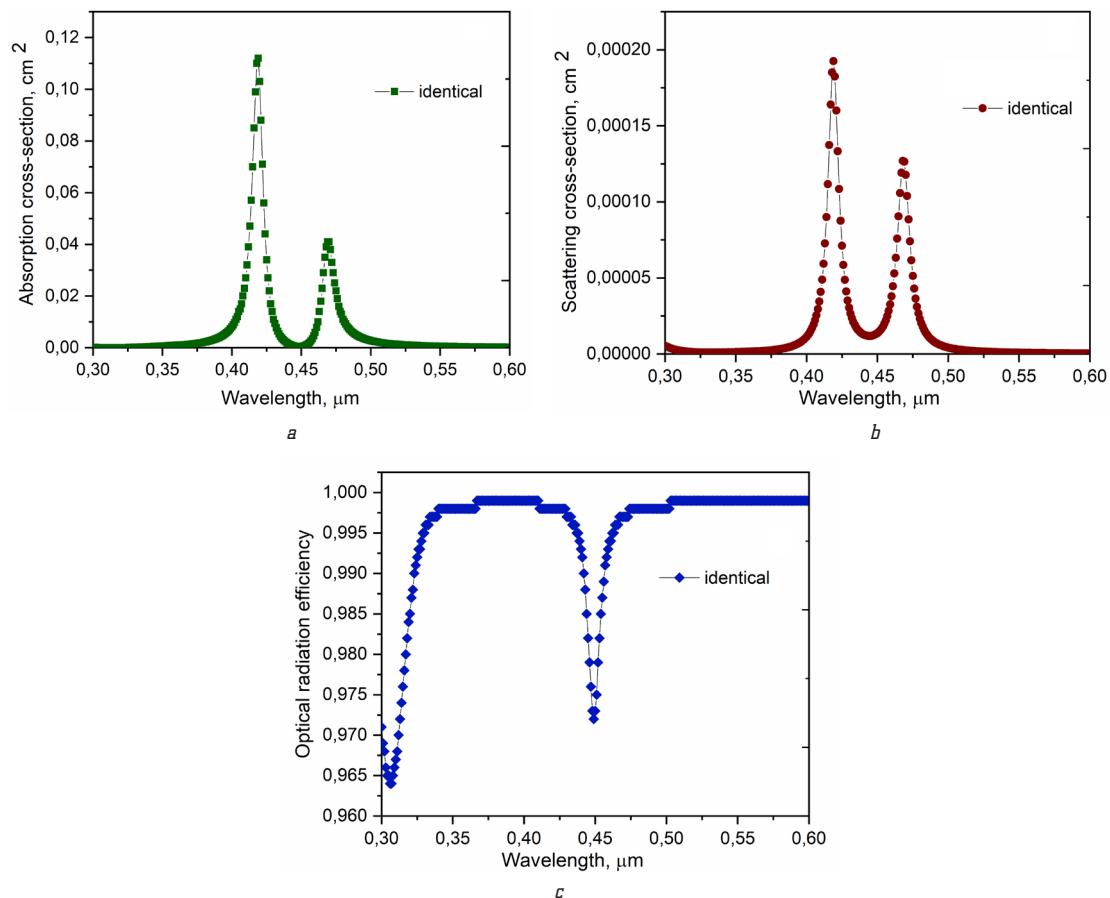


Fig. 2. Spectra of an ensemble of identical ellipsoidal nanoparticles: *a* – absorption cross of sections; *b* – scattering; *c* – optical radiation efficiency

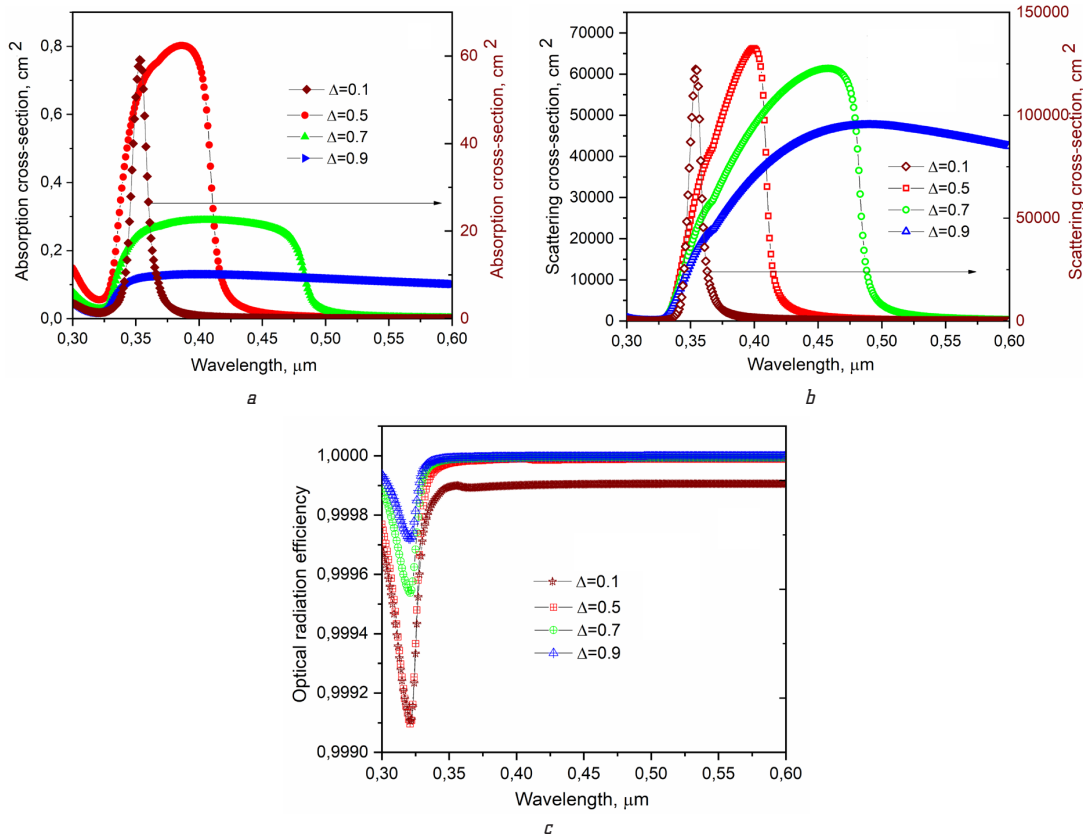


Fig. 3. Spectra of an ensemble of nanoparticles with different ellipsoidal shapes: *a* – absorption cross of sections; *b* – scattering; *c* – optical radiation efficiency

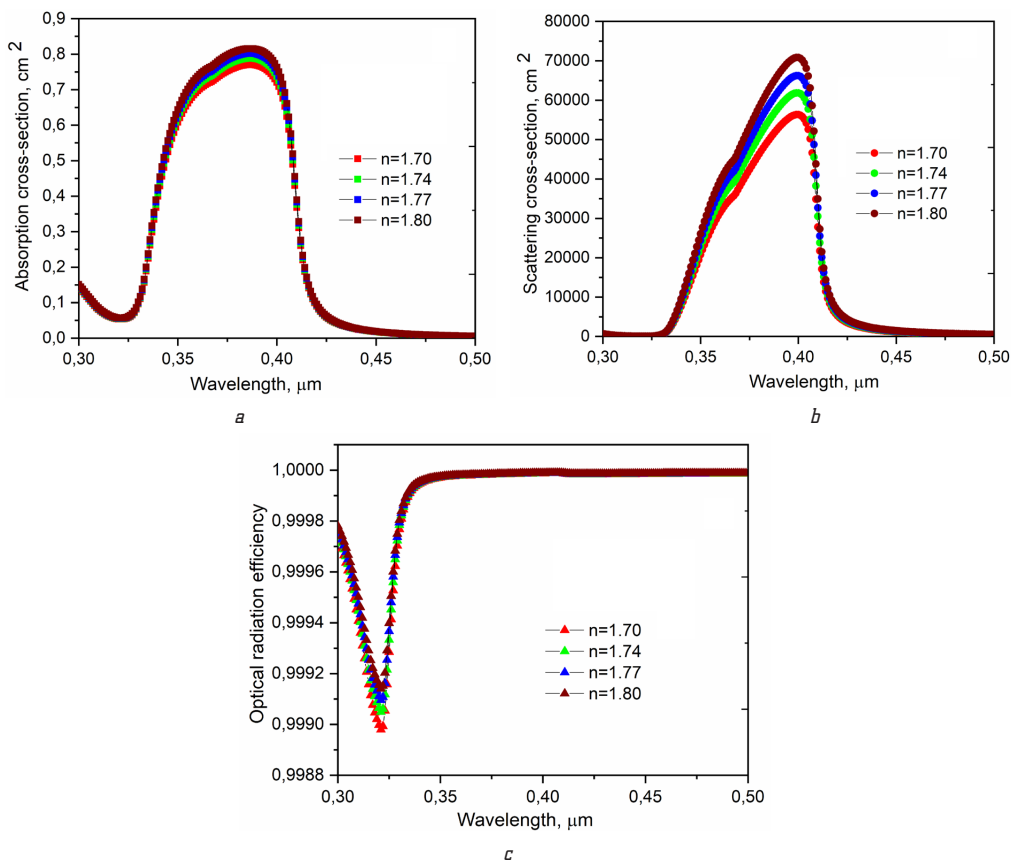


Fig. 4. Spectra of an ensemble of nanoparticles with different ellipsoidal shapes ($\Delta = 0.5$) depending on the refractive index of the organic semiconductor: *a* – absorption cross of sections; *b* – scattering; *c* – optical radiation efficiency

The practical significance of the obtained results lies in the possibility of increasing the efficiency, tuning the spectral characteristics, reducing the power consumption and increasing the lifetime of organic diodes due to the tunable plasmonic properties of spherical ellipsoidal silver nanoparticles in the matrix of an organic semiconductor. Their properties can be optimized by tuning the plasmonic resonance to the appropriate wavelengths.

One of the major research limitations is the precise control of nanoparticle shape, size, and distribution. Effective control of plasmonic properties requires high precision in the synthesis of nanoparticles with the desired geometry, since small changes in shape or size can greatly affect the plasmonic resonance.

Marital law and active hostilities caused interruptions in the power supply and complicated the work of scientists, which could lead to delays even in theoretical research.

Considering the above, further research can focus on the structure of silver nanoparticles and their orientation, which will allow further tuning of plasmonic properties for specific optical applications. This will enable the creation of organic diodes with improved properties, such as increased quantum yield, reduced light absorption, and enhanced brightness for use in displays and lighting devices.

4. Conclusions

In this work, the absorption and scattering cross sections as well as the optical radiation efficiency of an ensemble of spherical and ellipsoidal nanoparticles dispersed in an organic semiconductor matrix have been calculated. It is shown that the peak of the absorption and scattering cross sections of an ensemble of spherical nanoparticles of different sizes is shifted to the short wavelength region of the spectrum from 0.433 to 0.342 μm . In addition, there is a slight increase in the absorption cross section and a decrease in the scattering cross section, confirming the presence of smaller nanoparticles. An ensemble of ellipsoidal nanoparticles of different sizes and shapes is characterized by a single plasmon peak for both the absorption and scattering cross sections at the, unlike nanoparticles of the same size which are characterized by longitudinal and transverse modes of plasmon excitation. There are peaks at 0.354 μm for $\Delta=1$, 0.385 μm for $\Delta=0.5$, 0.415 μm for $\Delta=0.7$, and 0.440 μm for $\Delta=0.9$ in case of absorption cross sections. It was found that the change in refractive index of the organic semiconductor matrix mainly affects only the size of the scattering cross section of the ensemble of ellipsoidal nanoparticles dispersed in it. This research is a preliminary step towards studying the effect of these particles on the properties of organic light-emitting structures.

Conflict of interest

The authors declare that they have no conflict of interest in relation to this study, including financial, personal, authorship or other, which could affect the study and its results presented in this article.

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Data availability

The manuscript has no associated data.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies in the creation of the presented work.

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- ✉ **Iryna Yaremchuk**, Doctor of Technical Sciences, Professor, Head of Department of Electronic Engineering, Lviv Polytechnic National University, Lviv, Ukraine, e-mail: iryna.y.yaremchuk@lpnu.ua, ORCID: <https://orcid.org/0000-0002-7072-5950>
-
- Tetiana Bulavinets**, PhD, Associate Professor, Department of Electronic Engineering, Lviv Polytechnic National University, Lviv, Ukraine, ORCID: <https://orcid.org/0000-0001-6898-3363>
-
- Yuriy Smachylo**, PhD Student, Department of Electronic Engineering, Lviv Polytechnic National University, Lviv, Ukraine, ORCID: <https://orcid.org/0009-0007-2225-1993>
-
- Yuriy Mysiuk**, PhD Student, Department of Electronic Engineering, Lviv Polytechnic National University, Lviv, Ukraine, ORCID: <https://orcid.org/0009-0002-1056-323X>
-
- Volodymyr Fitio**, Doctor of Technical Sciences, Professor, Department of Electronic Engineering, Lviv Polytechnic National University, Lviv, Ukraine, ORCID: <https://orcid.org/0000-0001-6086-4087>
-
- Pavlo Stakhira**, Doctor of Technical Sciences, Professor, Department of Electronic Engineering, Lviv Polytechnic National University, Lviv, Ukraine, ORCID: <https://orcid.org/0000-0001-5210-415X>
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- ✉ Corresponding author