Study of the opto – electronic coupling in some complementary metallic – dielectric metamaterials used as biosensors

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Abstract. Using of the metamaterials improves the sensitivity and resolution of sensor devices, having multiple applications in biomedicine (imaging, biomarkers, telemedicine, etc.). We have studied the effect of different types of ordering in the lattices of nanostructures inside the metamaterial samples. Structures consisting of metallic atoms (tens of nanometers diameter) arranged relatively periodically in a dense dielectric matrix have been analyzed by simulation methods. We have focused on the opto-electronic coupling phenomena and the interactions between plasmons and the applied field. A study of the electron scattering processes was performed, the simulational data being used to calculate the reflectance and permittivity in function of the wavelength, in the range of 500…2300 nm. The electromagnetic properties of the metamaterial samples depending on particles shape, dimensions and on the reports between metallic inclusions dimensions and the dimensions of the dielectric matrix where are included have been studied and new configurations were proposed in order to improve the metamaterial sensors performance. We expect an increment in the response to the exciting field of about 7%, in a more dense state of spins characterising the new material samples.

1. Introduction
The metallic – dielectric metamaterials used for the transducer part of the biosensors represent new conceived material structures with special properties. Using of the metamaterials improves the sensitivity and resolution of sensor devices, having multiple applications in biomedicine (imaging, biomarkers, real-time diagnostics, telemedicine, drug delivery and so on). High performances can be achieved at imaging contrast agents, wearable health monitors, biomarker detection, spectroscopic labels and rulers, etc.

Sensors working on the basis on the surface plasmon resonance (SPR) are one of the most performing optical biosensors, with high sensitivity of more than a hundred nm/RIU (refractive index unit), if we refer to the sensitivity to refractive index change inside the fluidic medium for detection.

In literature different structures of metamaterial biosensors have been proposed, with sensitivity of about 70…100 nm/R.I.U [5,7,10,13]. We have studied the effect of different types of ordering in the lattices of nanostructures inside the metamaterial samples for improving these performances. We expect an increment in the response to the exciting field of about 7%, in a more dense state of spins characterising the new material samples.
2. Characterization of the method

The analyzed device is a plasmonic biosensor type (figure 1), which detects the refractive index of biomolecules. This type of sensors with metamaterials can be used in the domain of 100–800 THz and has the sensitivity of tens of nm to hundred of nm per RIU (refractive index unit) [1-4]. The device sensitivity could be further improved by optimizing the metamaterial structure and decreasing the permittivity of the substrate.

2.1 Details of the structure

For biosensing devices are suitable the complementary metallic – dielectric metamaterials with subwavelength separations of meta-atoms. The metallic constituents’ dimensions are comparable to the resonant wavelength of the exciting field. Inclusions of gold, aluminum, silver and even copper have been considered, which represent the nano-components on an alumina or silica substrate.

Many structures have already experimented, having different shape of the metallic inclusions into the dielectric substrate (e.g. cylindrical and spherical inclusions, nanorods of different shapes, in PI, U, T, rings, etc., placed in periodical configurations, and so on [5-7]).

Starting from the idea of the split-ring-resonator metamaterials configurations, where one or two concentric metallic bands of different shape (ring, U, etc.) are distributed periodical on a surface, we have conceived and simulated a metamaterial structure with periodical elements inside, presenting an ordering hierarchy. Spherical metallic inclusions of different diameter between 20 and 80 nm are distributed randomly in the nodes of hexagons placed in the dielectric walls of cavities. These cavities have approximately spherical and elliptic shape, with maximum long axis of 100 – 600 nm. The bees’ honeycomb structure (figure 2) was considered as model. In the honeycomb unit cell, with different dimensions inside the volume, we have hierarchy of first order (figure 3).

For manufacturing the metamaterial, the dielectric matrix with irregular honeycomb structure and cavities can be obtained by superimposing successive layers. In every layer, the metallic spherical inclusions are added.

Figure 1. Schematic representation of a plasmonic biosensor (after https://bitesizebio.com/34644/biosensor-chips-surface-plasmon-resonance/)

Figure 2. The irregular honeycomb structure model for the sensing metamaterial (image from https://honeybeesuite.com/wp-content/uploads/2016/01/Feral-hive-Mike-Riter.jpg).

Figure 3. The considered hierarchy of the honeycomb unit cell of the metamaterial [14]. Metallic inclusions are placed in the inside (2nd order) hexagon nodes.
The effective permittivity of the composed metamaterial can be calculated with the formula [7]:

\[
\varepsilon_{r,\text{eff}} = 1 - \frac{\omega_p^2}{\omega^2}
\]

(1)

where \(\omega_p\) is the plasma frequency and \(\omega = 2\pi f\) is the frequency of the propagating electromagnetic wave. The condition: \(\omega < \omega_p\) implies \(\varepsilon_{\text{eff}} < 0\). The plasma frequency depends on the metallic nano-components geometry and nature and can be determined using the simulation data. The HFSS program (Ansys Technologies) was used for our structural simulation. A model for the metamaterial was conceived, by reproducing the material at microscopic level, starting with the unit cell and reconstructing a thin film which represents the metamaterial sensor layer (Figure 4).

We have used a test configuration presented in Figure 5. Exposure of the metamaterials samples has occurred in a multi-mode channel waveguide, having dimensions of \(w = 5.6 \mu\text{m}, h = 3.2 \mu\text{m}\). With help of the HFSS program, the samples analysis was performed in the domain of 500…2300 nm for the incident field wavelength.

The effect of different types of ordering in the lattices of nanostructures inside the metamaterial samples was studied. Our purpose was to predict the metamaterial configuration which to ensure an improvement of the sensitivity, selectivity, and resolution of the biosensor across broader spectral regions, as wide as possible in the considerate frequency range.

![Figure 4. Configuration of the detection zone of the biosensor with metamaterials.](image)

![Figure 5. The multi-mode channel waveguide with the metamaterial sample.](image)

2.2 The phenomenon - opto–electronic coupling

Considering the biosensor structure illustrated in figure 4, changes in the refractive index that take place in the dielectric medium of the optical structure of the sensor are detected.

The surface plasmons, which represent the free electron oscillations at the interface between the metamaterial and the fluidic medium for detection, are tuned by the modifying the metamaterial constituents’ geometry and nature. Localized surface plasmons are excited in the metamaterial layer with an electromagnetic field having the frequency near the plasma frequency of the metallic constituents inside [8-10].

The surface plasmons are extremely sensitive to the refractive index modifications in the dielectric medium, within the penetration depth of the evanescent field [11,12].

We have focused on the opto-electronic coupling phenomena and the interactions between plasmons and the applied field. A study of the electron scattering processes was performed, the simulational data being used to calculate the reflectance and permittivity for the device material.

3. Results and discussions

Study of the biosensor response depends on the opto–electronic coupling at structure level. A resonant coupling between plasmon and photonic modes is of maxim interest from the point of view of the effects.

Structural details of the considered metamaterials have been presented in paragraph 2.1 and on the graphs were indicated the substances chosen for exemplification (selective graphs were presented for
Simulations were iterated until reproducible results have been obtained. Simulation method was tested until errors of a few percents have been obtained for the data reported for similar structures in literature [5,7,10,13]. We mention that the simulation model represents the intellectual property of the authors and can be published after patent.

Material reflectance (fraction of incident electromagnetic power reflected at an interface) and electric permittivity in function of the wavelength, in the range of 500...2300 nm, have been determined and represented on graphs, using the simulation data. Results are given in figures 6 and 7.

Curve evolutions are depending on concentration of the constituents and on incidence angle of the incident light. One observes that, higher the concentration of metallic insertions, higher the material reflectance. The reflectance is almost double in the case of the Au inclusion in comparison with the materials with Al inclusions. What is important is that peaks and valleys on curves are not accentuated, the minima caused by geometrical resonances being of not very low magnitude in comparison with the average envelope.

The metamaterial structure keeps its periodic character, but the dimensions of the geometric unit cell (the hexagon with hierarchy) are variable. In the same time, diameters of the spherical metallic inclusions are also distributed in an interval. Due to this fact, the effects of geometrical resonances can be partially avoided. The minima of reflectance are not so accentuated like in the case of a singular set of dimensions for the structural unit components. As a result, the material keeps its metamaterial properties concerning the wave propagation, but the unwanted resonant valleys on the graphs for some characteristic quantities can be flattened.

![Figure 6](image.png)

**Figure 6.** Reflectance in function of the wavelength, for different types and concentrations c of the metallic inclusions in the substrate. For clarity reasons, only the average envelope of the complete curve was represented in the most cases.

The substrate nature influence does not change the casual evolution of the result. No significant differences for alumina substrate occur.

Real and imaginary parts of the effective electric permittivity in function of the frequency were given in figure 7, in the frequency range of 130…600 THz (which represents the correspondent of 500…2300 nm wavelength).

The electromagnetic properties of the metamaterial samples depends on particles shape, dimensions and on the reports between metallic inclusions dimensions and the dimensions of the dielectric matrix where are included. Graphs are indicating that dissipations within the medium (proportional with ε'') increase when the inclusion concentration increases. In the same time, the stored energy within the medium (related with ε') decreases in the same conditions.
Figure 7. Real and imaginary parts of the electric effective permittivity of the metamaterial sensor layer, for different values of the metallic inclusions in the substrate.

Sensitivity $S$ of the metamaterial biosensor (relation (2)) was calculated and represented on graphs in Figure 8. We have calculated the sensitivity to refractive index change (shift in the resonant wavelength $\lambda_{res}$ per refractive index ($n$) unit) at the interface with the fluidic medium for detection. Values of about 100 nm/R.I.U. (R.I.U. = refractive index unit) can be achieved if we set properly the metamaterial configuration.

$$S = \frac{\partial \lambda_{res}}{\partial n} \ [nm / RIU]$$

(2)

Figure 8. Sensitivity of the metamaterial biosensor in function of the wavelength, for the considered types of metallic inclusions in the dielectric substrate (silica, respectively alumina).

Due to the variable dimensions of the geometric unit cell inside the metamaterial structure, the character of a curve with punctual maxima disappears in the case of the biosensor sensitivity. Flat
maxima occur in a wide domain of wavelength values, which is convenient for the practical operation of the device. The alumina substrate demonstrates itself less performing than the silica substrate material, the results being somehow comparable.

4. Conclusions
A biosensor type based on metamaterial structures has been analyzed in this paper. The electromagnetic properties of the metamaterial sensor layer depends on particles shape, dimensions and is influenced on the reports between metallic inclusions dimensions and the dimensions of the dielectric matrix where are included. New configurations have been proposed, in order to improve the metamaterial sensors performance. A structure of irregular honeycomb and first hierarchy, with inclusions of varied dimensions placed in the nodes was tested by simulation. The purpose was to avoid the evolution with maxima and minima caused by the geometrical resonances in the case of the physical quantities which describes the metamaterial behavior (reflectance, permittivity). This desiderate was realized in a considerable percentage and evolutions with flat maxima and minima having low variation in magnitude were encounted for the material parameters. An increasing of the biosensor sensitivity occurs, of more than 20 % on specific frequency domains, in comparison with the data reported in literature for the composed structures but with a singular set of dimensions. We expect an increment in the response to the exciting field of about 7%, in a more dense state of spins characterizing the new material samples.

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