Tailorable spectral dispersion of copper-nickel 1D plasmonic crystals

Marko Obradov, Zoran Jakšić, *Senior Member, IEEE*, Ivana Mladenović, Slobodan Vuković, Goran Isić, Dana Vasiljević Radović, Jelena Lamovec

***Abstract*— Plasmonic nanocomposites are a new class of materials that offers unprecedented opportunities to tailor the optical response, including the possibility to design their spectral and spatial dispersion at will. This includes the optical parameters rarely or never met in nature, which opens a path toward plasmonic metamaterials and the wide new area of transformation optics. Responsible for such a unique behavior are bound surface modes propagating along interfaces between materials with different signs of relative dielectric permittivity known as surface plasmon polaritons (SPP). Most metals possess negative relative permittivity in optical range due to the existence of free electron plasma. However, they also exhibit large absorption losses and are bound to a given spectral range defined by the metal itself, which is the reason why alternative plasmonic materials are being actively sought upon. One possible way to extend the toolbox of available materials is to use alternating metal-dielectric or metal-metal layers – the one-dimensional plasmonic crystals. Typically gold and silver are used for the metal part due to their large conductance and generally favorable properties. In this contribution we perform an analysis of the suitability of the use of copper for plasmonic nanocomposites. Its oxidation, the main barricade towards its more widespread use in plasmonics, is avoided by combining it with nickel. We utilize ab initio analysis by 2D finite element modeling and realistic material parameters to assess different electromagnetic modes. Tailorability of the response is attained by simple changing of the Cu to Ni fill factor. The analyzed Cu-Ni plasmonic crystals are convenient for simple, low cost biochemical sensors and superabsorbers.**

Marko Obradov is with Centre of Microel. Technologies, Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, 11000 Belgrade, Serbia (e-mail: marko.obradov@nanosys.ihtm.bg.ac.rs).

Zoran Jakšić is with Centre of Microelectronic Technologies, Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, 11000 Belgrade, Serbia (e-mail: jaksa@nanosys.ihtm.bg.ac.rs).

Ivana Mladenović is with Centre of Microel. Technologies, Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, 11000 Belgrade, Serbia (e-mail: ivana@nanosys.ihtm.bg.ac.rs).

Slobodan Vuković is with Centre of Microel. Technologies, Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, 11000 Belgrade, Serbia (e-mail: svukovic@nanosys.ihtm.bg.ac.rs).

Goran Isić is with Center for Solid State Physics and New Materials, Institute of Physics Belgrade, University of Belgrade, Pregrevica 118, 11080 Belgrade, Serbia (e-mail: isicg@ipb.ac.rs).

Dana Vasiljević Radović is with Centre of Microelectronic Technologies, Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, 11000 Belgrade, Serbia (e-mail: dana@nanosys.ihtm.bg.ac.rs).

Jelena Lamovec is with Centre of Microel. Technologies, Institute of Chemistry, Technology and Metallurgy, University of Belgrade, Njegoševa 12, 11000 Belgrade, Serbia (e-mail: jejal@nanosys.ihtm.bg.ac.rs).

***Index Terms*— Plasmonics; Heterometallics; Optical Multilayers; Biochemical sensors; Superabsorbers**

I. INTRODUCTION

INTERFACES between materials with contrasting dielectric permittivity are of special interest in the field of electromagnetics [1, 2] since they ensure the appearance of a number of modes. Maybe the best example are the interfaces between materials with negative and with positive relative dielectric permittivity, the simplest case being an interface between semi-infinite metal and dielectric. In this case the interface between metal and dielectric acts as a waveguide supporting a two-dimensional surface hybrid mode propagating along the interface – the surface plasmon polariton (SPP) [2]. SPPs are p-polarized and confined to the interface, exponentially decaying in both perpendicular directions. By producing metal-dielectric (nano)composites multitudinous interfaces are created and the individual modes from each one of them will interact, resulting in a wealth of new modes. The part of electromagnetics dealing with the properties, design and utilization of such structures and modes is called plasmonics [2, 3].

Most notable and readily observed characteristic of SPPs is that they have the optical frequency while possessing much smaller wavelengths, i.e. their dispersion curves are located way outside the light cone. This results in extreme field localizations on the subwavelength scale [4, 5]. However, the mismatch between the large wavevectors of the bound waves (SPP) and the much smaller wavevectors of the propagating waves imposes the need for additional impendence matching between the structure and the environment. A possible approach to ensure this matching is the design of plasmonic structures with embedded diffractive gratings, for instance metallic layers with periodic pattern of grooves or openings on their surface [6]. A special subclass of plasmonic materials are superlattices in which the dielectric permittivity varies periodically along one, two or all three spatial dimensions – the plasmonic crystals. Even the simplest 1D plasmonic crystals offer a number of available electromagnetic modes.

The possibility to tailor the dispersive properties of the plasmonic nanocomposites through modification of their geometry and optical parameters brought to an almost complete control of the optical space, resulting in what is now known as transformation optics [7]. Practical applications of plasmonic structures are numerous and include chemical and biological sensors [8], superabsorbers and superlenses [9, 10], cloaking devices [11], etc.

Most commonly used plasmonic constituents are noble metals like gold and silver. These metals possess a plasma frequency fixed in the UV-vis part of the optical spectrum and exhibit high absorptive losses around that frequency. This is the reason why alternative plasmonic materials are actively being researched, some of them with properties tunable by design, for instance highly doped semiconductors and transparent conductive oxides (TCO) [12].

We consider here a heterometallic plasmonic crystal consisting of nickel and copper layers, the structure not previously investigated in literature. The idea is to generate a new plasmonic material with its optical/plasmonic parameters continually tunable by modifications of its layer thickness ratio. This should lead to a tailorability of the obtainable plasma frequency between the frequencies of the two constituent metals, as well as in the possibility to tune and adjust the dispersion curves of the obtained multilayers. We limit ourselves to one-dimensional structures – multilayer films with periodically alternating strata. We consider the nickel-copper multilayers numerically, utilizing the finite element program package Comsol Multiphysics. The model we generate is two-dimensional.

II. Theory

We investigate heterometallic plasmonic crystals consisting of copper and nickel layers shown in Fig. 1. Layers are uniform and homogenous and described by their respective relative dielectric permittivity ε(ω) and by their respective thicknesses (d_1 and d_p for nickel and d_2 for copper layers). At optical frequencies the magnetic permeability of materials is μ=1.

For most metals in the optical range, including Ni and Cu, relative dielectric permittivity is well described by the lossy extended Drude model [2]

$$\varepsilon(\omega) = \varepsilon_\infty - \frac{\omega_p^2}{\omega^2 + i\gamma\omega}, \tag{1}$$

ε_∞ is the asymptotic dielectric permittivity and $\gamma=1/\tau$ is the characteristic frequency related to the damping of electron oscillations due to collisions, where τ is the relaxation time of the electron gas and ω_p is the plasma frequency, determined by the concentration of free carriers

$$\omega_p = \frac{ne^2}{m^* \varepsilon_0} \tag{2}$$

where n is the electron concentration, e is the free electron charge ($1.6\cdot10^{-19}$ C), ε_0 is the dielectric permittivity of vacuum ($8.854\cdot10^{-12}$ F/m), and m^* is the effective mass of electrons.

We used surface diffractive gratings readily formable by etching one of the two metal constituents (copper) to provide the necessary impendence matching between the propagating waves and the SPPs bound to the heterometallic interface.

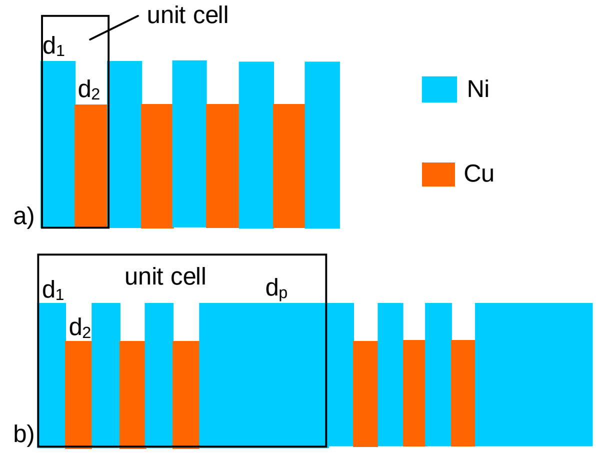


Fig. 1. Geometry of heterometallic plasmonic crystals: a) alternating Cu and Ni layers and b) alternating Cu and Ni layers with a four wavelength thick nickel layer after every three Ni-Cu pairs.

Practical implementation of such diffractive gratings in our structures is rather straightforward and involves dipping the entire multilayer in a solvent that will selectively remove only one material. Impedance matching is readily described by the wave vectors of the diffracted mode k_g, the propagating wave k_p and the surface mode k_{spp} [13]. The diffracted mode wave vector k_g is determined by the diffractive grating constant $A = d_1 + d_2$ for the simple case shown in Fig. 1.a as

$$k_g = \pm m \frac{2\pi}{A}\ , \tag{3}$$

where m is an integer. Coupling of the propagating wave to the SPP occurs when following condition is met:

$$\vec{k}_{spp} = \vec{k}_g + \vec{k}_p\ , \tag{4}$$

where k_p is the in-plane wave vector of the propagating wave

$$k_p = \frac{\omega}{c} \sin\theta\,, \tag{5}$$

where ω is the angular frequency, c is the speed of light in the medium above the plasmonic surface and θ is the incident angle.

III. Results and discussion

We examined optical properties of plasmonic crystals described in Fig. 1. using the finite element method. To this purpose we applied the RF module of Comsol Multiphysics software package. The structure in Fig. 1a consists of alternating $d_1 = 200$nm thick nickel and $d_2 = 300$ nm thick copper layers. The structure in Fig. 1b has the same d_1 and d_2, but there is a four wavelength thick ($d_p=4\lambda$) nickel layer after every three Ni-Cu pairs. For the purpose of our simulation the plasmonic crystals are considered infinite in the direction perpendicular to the layers. We assumed that the structure was surrounded by air. The parameters of Drude model describing the relative permittivity of the metals are taken from literature [15].

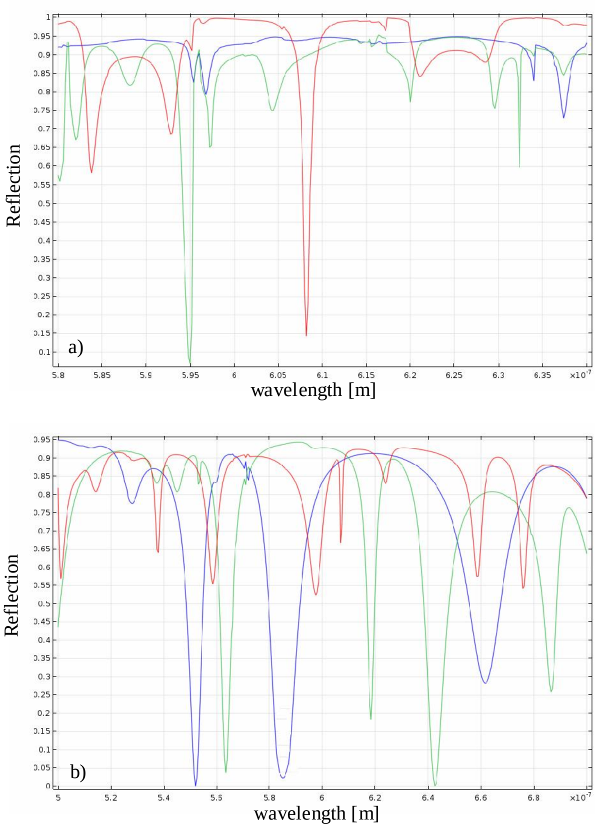


Fig. 2. Dispersive properties of heterometallic plasmonic crystal for different incident angles: normal incidence (blue), 30º incident angle (green) and 60º incident angle (red); a) for structure shown in Fig.1a with d_1=200nm and d_2 = 300 nm; b) for structure shown in Fig.1.b with d_1 = 200 nm, d_2 = 300 nm and $d_p = 4\lambda$.

Numerical simulation obtains the spatial field distributions as well as the scattering parameters for a plane wave incident on the structure surface at various angles. Two ports were added above and below the unit cell, each of them parallel to the surface. One port was active, allowing the incident light to enter the structure through it. Floquet boundary conditions simulate plasmonic crystal infinite dimensions via the unit cell periodicity. Parametric sweep of the wavelength of the incident light was used to determine the spectral dependence of the scattering parameters and the spatial distribution of the electromagnetic fields.

It is readily seen from Fig. 2 that both structures exhibit rich modal behavior. For the structure shown in Fig. 1a two dominant resonant dips in reflection can be observed at 595 nm and 608 nm for incident angles of 30º and 60º respectively. Structure behaves as a standard diffractive grating and incident light couples to the plasmonic modes when the impendence matching conditions are met. Since there is no transmission through the structure almost all incident radiation will be absorbed at resonant frequencies.

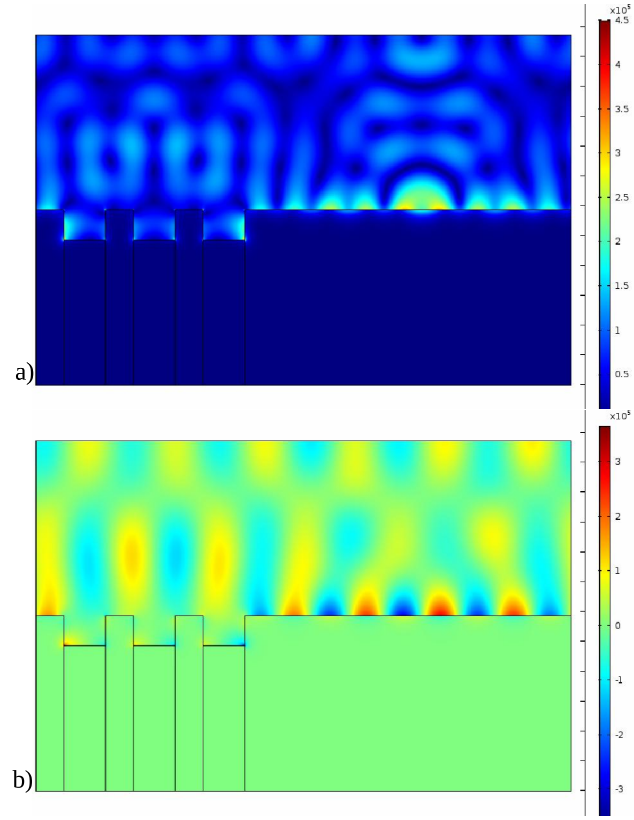
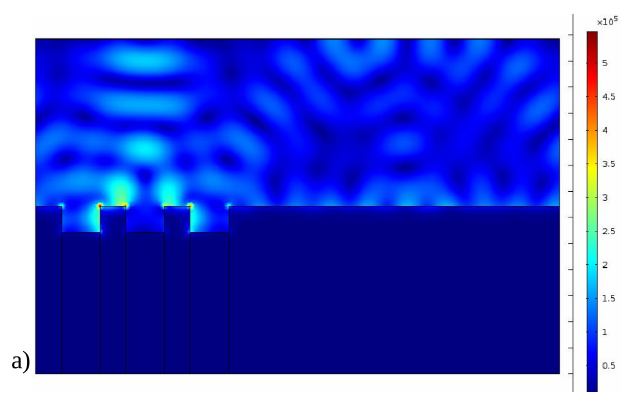


Fig. 3 Electric field spatial distribution for d_1 = 200 nm, d_2 = 300 nm and $d_p = 4\lambda$ structure at 584 nm and normal incidence; a) electric field intensity, b) component of the electric field vector in the direction perpendicular to the surface.

When we increase the complexity of the structure by practically superimposing two diffractive gratings (Fig. 1b) we can observe significant increase in the wave pattern complexity. As seen in Fig. 2b there are now several resonant dips in the reflection characteristics connected with coupling between the propagating light wave and the plasmonic modes. It is especially strong for normal and 30º incidence.



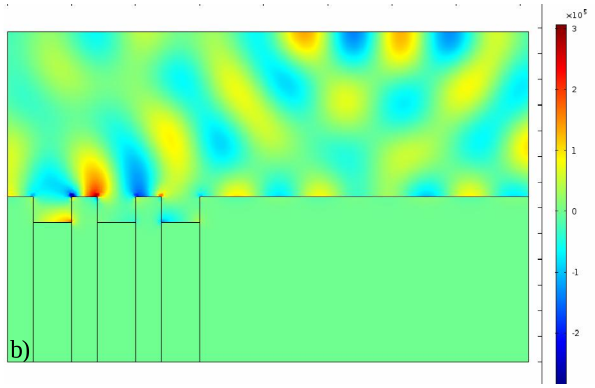


Fig. 4. Electric field spatial distribution for d_1 = 200 nm, d_2 = 300 nm and $d_p = 4\lambda$ structure at 642 nm and 30º incident angle; a) electric field intensity, b) component of the electric field vector in direction perpendicular to the surface.

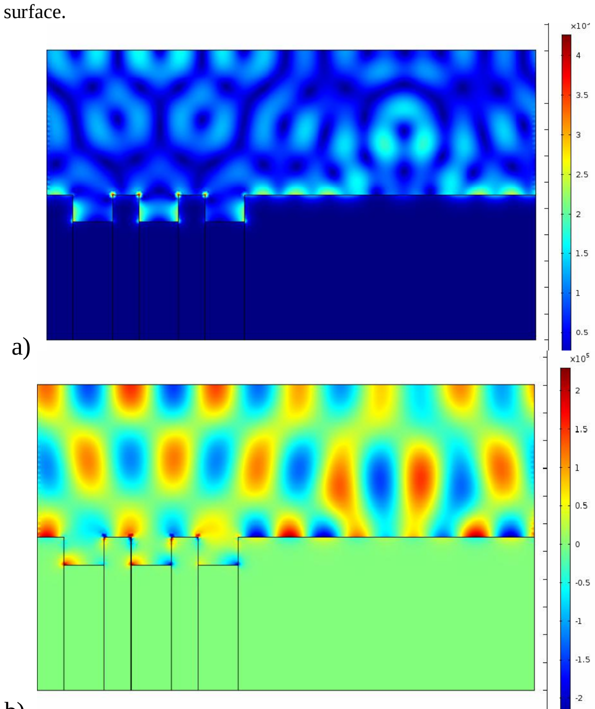


Fig. 5. Electric field spatial distribution for d_1 = 200 nm, d_2 = 300 nm and $d_p = 4\lambda$ structure at 552 nm and normal incidence; a) electric field intensity, b) component of the electric field vector in direction perpendicular to the surface.

In Fig. 3 spatial field distributions for 584 nm and normal incidence are shown. Fig. 3a shows the electric field intensity, whereas Fig. 3b shows only the electric field component in the direction perpendicular to the surface. It is readily observed that the incident light predominantly couples into the SPPs which propagate along the platform. Following the same pattern Fig. 4 shows the spatial field distributions at 642 nm for a 30º incident angle. In this case light is localized primarily in the subgrating with the higher spatial frequency, which is manifested as pronounced edge effects.

Fig. 5 shows the spatial field distributions at 552 nm for normal incidence. Unlike to the previous two cases light localization occurs on both subgratings, including the copper channels between nickel layers.

IV. Conclusion

We analyzed numerically some specific features of the spectral dispersion of 1D heterometallic plasmonic crystals. We started with a simple structure consisting of alternating layers of copper and nickel. When a diffractive grating was embedded into the plasmonic crystal surface we observed narrow resonant dips in reflectivity of the structure, which is related to the incident light coupling with plasmonic modes due to the grating-based impedance matching. These narrow resonant dips in reflection offer an excellent contrast when dispersion relation shifts due to the presence of an adsorbent on the structure surface, pointing out to the possible use in ultrasensitive chemical sensing. The choice of materials and dimensions ensures that the operating wavelengths are in the green/red part of the visible spectrum. We also proposed to further functionalize the structure by superimposing two diffractive gratings within the same plasmonic crystal. As a result additional resonant wavelengths were achieved (dips in the spectral reflectance). Since there is no transmission through the structure, all of the incident light will be absorbed when coupling to plasmonic modes occurs. Thus the structure can be used as a superabsorber. Additionally plasmonic modes of the structure can be selective of the individual subgratings, leading to light localization in different parts of the structure. Thus we have shown that our structures exhibit a potential for simple, low cost, easily fabricated biochemical sensors and superabsorbers. Large absorption losses observed in heterometallics are no issue here and actually even prove themselves to be beneficial both in the case of superabsorbers (directly improving their efficiency) and that of sensors (large contrasts of reflection coefficient and multiple resonances). In our further work we intend to modify the structures with a goal to achieve broadband multidirectional superabsorption.

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