Photonic/Plasmonic Structures from Metallic Nanoparticles in a Glass Matrix

O.Kiriyenko[*,1], *W.Hergert*[1], *S.Wackerow*[1], *M.Beleites*[1] *and H.Graener*[1]
[1] *Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, Friedemann-Bach-Platz 6, 06108 Halle, Germany*
[*] *oleksiy.kiriyenko@physik.uni-halle.de*

Abstract: Glass containing nanoparticles is a promising material for various photonic applications due to the unique optical properties mainly resulting from the strong surface plasmon resonance (SPR) of the silver nanoparticles. The characteristics of the resonance can be modified by varying size, shape and concentration of the particles. A finite element method (FEM) implemented in the software Comsol Multiphysics is used to calculate effective permittivities of this composite material. Two-dimensional structures of rods characterized by an effective permittivity have been considered. In contrast to the preparation of photonic crystals in silicon the deviations from the ideal structures are more important for the structures under consideration here, because such structures cannot be manufactured with the same accuracy like the silicon counterparts. Such deviations are a variation of the radii of the rods and a variation of the filling factor. It was investigated how such deviations influence the transmission of the structures.

Keywords: photonic/plasmonic structures, silver nanoparticles, effective permittivity, waveguides.

1 Introduction

In the last years the interest in nanostructures for optical applications increased dramatically. Metallic nanoparticles in a glass matrix can be used to tailor different optical properties of photonic/plasmonic structures. DC electric field-assisted dissolution can be used as a method to structure glass templates containing such nanoparticles [1]. As a result of the procedure photonic crystal slabs of hexagonal or square symmetry are obtained. The structure is formed as a two-dimensional (2D) lattice of regions of a composite material containing the metallic nanoparticles. The particles are usually spherical but can be changed in shape by mechanical stress or intensive laser pulses. A basic ingredient for a theoretical analysis of functional elements, prepared on the basis of such a material, is the calculation of the optical properties of the composite material. Based on such knowledge, waveguide structures can be designed.

2 Properties of silver nanoparticles

According to the experimental results spherical and ellipsoidal silver nanoparticles have to be considered. Spherical silver nanoparticles have usually a radius of $R = 15-20nm$. Ellipsoidal nanoparticles can be described by a geometrical factor $L = a/c$. L is the relation between the two non-equal semi-axes $(a \neq b = c)$. The position of the ellipsoidal nanoparticles in the glass matrix is random, with parallel major semi-axes due to the method of preparation. The optical properties of such a system are defined by the ensemble of nanoparticles and are dependent on the distribution in the matrix. Effective medium theory is used to describe such a random distributions of metallic nanoparticles. Instead of the ensemble of particles itself, a homogeneous medium with a certain effective permittivity ε_{eff}, which leads to the same optical properties is considered. For ellipsoidal nanoparticles this effective permittivity depends on the direction and is a tensor of rank two. If the ellipsoidal nanoparticles are oriented along the x-axis, the tensor ε_{eff} is diagonal and the diagonal elements are: $\varepsilon_{xx}^{eff} \neq \varepsilon_{yy}^{eff} = \varepsilon_{zz}^{eff}$. The permittivity of the silver nanoparticles is described by Drude's model.

$$\varepsilon = \varepsilon_b + 1 - \frac{\omega_p}{\omega^2 + i\gamma\omega} \qquad (1)$$

Here ω_p, ε_b, γ represent the bulk plasma frequency, the contribution of interband transitions and all nonconduction electron contributions to the permittivity, and the damping of the electron oscillations. [2] The effective permittivity is a complex function of frequency. The imaginary part characterizes absorption of electromagnetic radiation by silver nanoparticles.

3 Calculation of effective permittivity

For the calculation of the effective permittivity an finite element method (FEM) has been used (cf. also [5, 4]). The electric field is applied to the composite, located in the plane capacitor. The energy density of the electric field between plates of capacitor for a homogeneous medium is given by:

$$W = \frac{1}{2}\varepsilon_0 \varepsilon \frac{S}{h} U_0^2 \tag{2}$$

The density of electric energy W of the composite in the capacitor is obtained from the minimization of the functional F with potential $\varphi(\mathbf{r})$ from:

$$W = F[\varphi(\mathbf{r})] = \frac{1}{2}\varepsilon_0 \int_V \varepsilon(\mathbf{r})[\nabla\varphi(\mathbf{r})]^2 d^3r \tag{3}$$

The local electrostatic potential is calculated from the following boundary-value problem:

$$\nabla \cdot [\varepsilon(\mathbf{r})\nabla\varphi(\mathbf{r})] = 0 \tag{4}$$

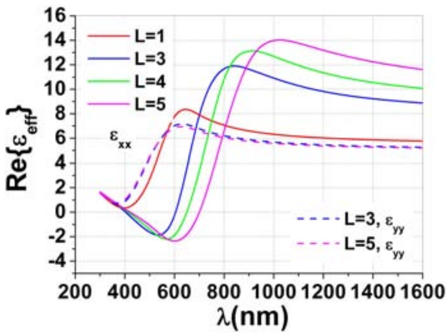

Figure 1: $\Re\varepsilon_{xx}^{eff}$ and $\Re\varepsilon_{yy}^{eff}$ for different shapes of nanoparticles. The filling factor is $f = 0.3$.

If ε for one of the constituents of the composite is complex, the density of energy W is a complex function too. Real and imaginary parts of ε_{eff} can be found independently. For a given L ($L = 1, 2, ..., 5$) a series of 100 randomly constructed ensembles of nanoparticles are investigated. The effective permittivity is found by an average over the set of ensembles. If $L \neq 1$ the tensor components of ε_{eff} were calculated.

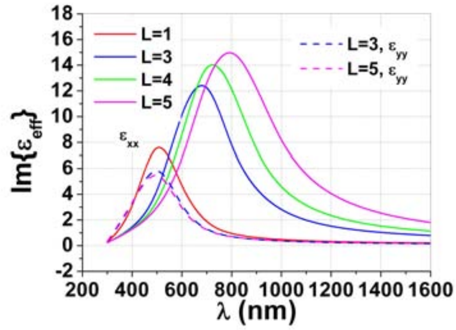

Figure 2: $\Im\varepsilon_{xx}^{eff}$ and $\Im\varepsilon_{yy}^{eff}$ for different shapes of nanoparticles. The filling factor is $f = 0.3$.

From Fig. 1 and Fig. 2 can be seen that ε_{xx}^{eff} grows with increasing factor L. The other diagonal element ε_{yy}^{eff} depends on geometry of particles very weakly because the absolute value of b is fixed. $\varepsilon_{xx}^{eff} > \varepsilon_{yy}^{eff}$ holds for any geometry factors for the real and image parts. The region of absorption of electromagnetic waves becomes broader with increasing L for ε_{xx}^{eff} element. The red line corresponds to spherical nanoparticles.

4 Plasmonic structures and statistical analysis

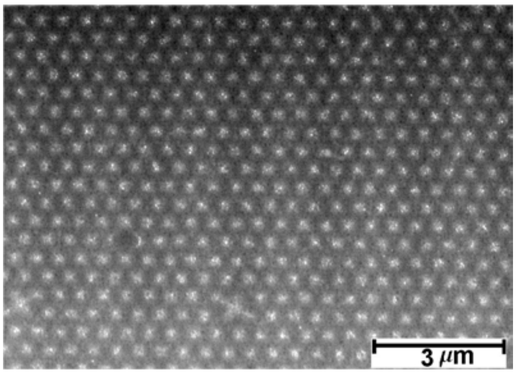

Figure 3: Experimental realization of a periodic plasmonic structure with $2D$ hexagonal lattice. The lattice constant is $0.5\mu m$. The white areas are the composite regions containing silver nanoparticles.

Starting from the base material two-dimensional photonic crystals can be constructed. Fig. 3 shows a photonic crystal realized in the glass matrix containing silver nanoparticles.

Fig. 3 demonstrates also, that the structure is not perfect. The nanocomposite areas, named rods further, are not of equal shape and tend to be square instead of circular. In a first approximation it was assumed, that the rods are represented by circles with different radii. The distribution of the radii in the sample is approximated by a Gaussian distribution (cf. Fig. 4).

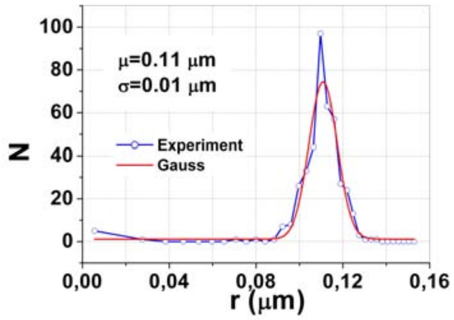

Figure 4: The distribution of radii in the structure shown in Fig.3. Experimental results (blue) are fitted by a Gaussian distribution (red) The average radius for this sample is $R = 0.11\mu m$

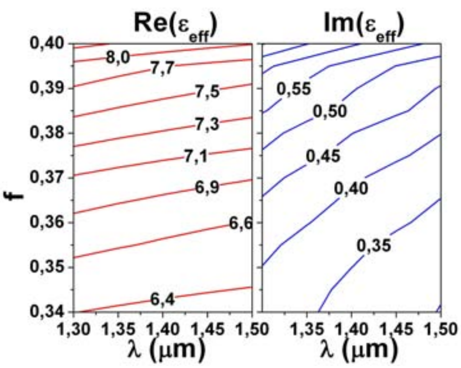

Figure 5: The effective permittivity ε_{eff} as a function of wavelength and filling factor. Results for spherical nanoparticles. The range of wavelength is $1.4 < \lambda < 1.5\mu m$

A waveguide on the basis of such a structure can be constructed if one row of rods is removed. This can be realized from the very beginning using a structured mask in the preparation of the sample or later by means of laser radiation. The localization of the electromagnetic field in a waveguide is possible for this lattice if the contrast of permittivity between the rods and the glass matrix will be larger than $\Re\varepsilon_{rods} - \varepsilon_{glass} > 4.5$. This contrast corresponds to a filling factor of $f = 0.37$. The effective permittivity ε_{eff} corresponding to different filling factors is shown in Fig. 5.

It is supposed that the filling factor for the nanocomposite has a Gaussian distribution with average $f = 0.37$. The data will be used for the calculation of the electromagnetic field in the non-ideal waveguides.

5 Photonic crystals and Y-waveguide

First, an ideal two-dimensional photonic crystal with hexagonal lattice structure is considered. In contrast to the experimental realization the structure is translational invariant in z-direction. The lattice constant of this structure is $a = 0.5\mu m$. The dielectric medium is glass and the permittivity of the rods is equal to the real part of ε_{eff}, i.e. no absorption is considered. It is assumed that the rods contain spherical nanoparticles. The medium inside the rods is isotropic. On the basis of this structure Y-waveguides are investigated numerically. The source of electromagnetic radiation is located on the left side of waveguide. We will consider electromagnetic waves with TM polarization. For this polarization the electromagnetic wave has a z component of electric field E_z and x, y components of magnetic field H_x, H_y.

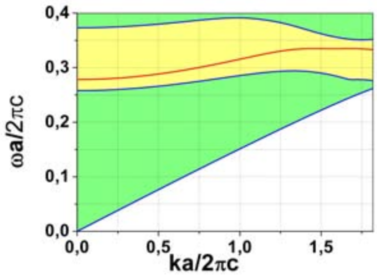

Figure 6: Projection of bands on waveguide the direction Γ-K in the first Brillouin zone together with mode of a linear waveguide (red line). Calculation is performed by means of MPB [3].

The photonic bandstructure of the crystal containing a linear defect is calculated.

Fig. 6 shows the bandstructure of the ideal structure projected on the direction Γ-K of the first Brillouin zone together with the bands of the crystal containing a defect. The calculation is performed for the TM mode. Inside the band-gap (yellow area) a defect mode can be seen.

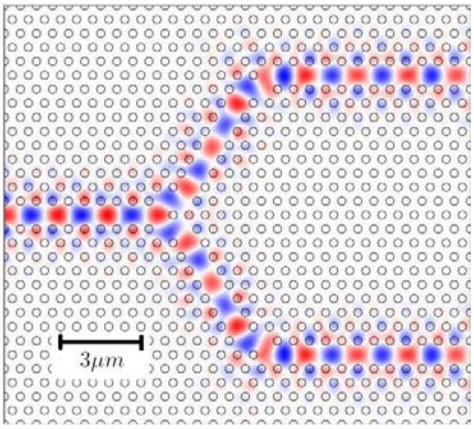

Figure 7: Ideal Y-waveguide in $2D$ photonic crystal with hexagonal lattice structure . The lattice constant is $a = 0.5\mu m$. E_z component of the electromagnetic field is shown.

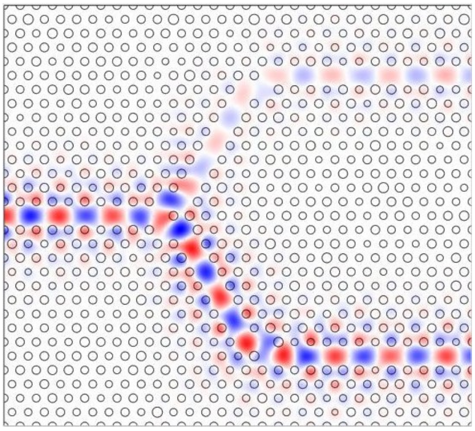

Figure 8: Y-waveguide like in Fig. 7, but the radii have a Gaussian distribution with mean value $r = 0.11\mu m$.

Electromagnetic waves with frequencies in the band-gap region cannot propagate in direction Γ-K in the ideal crystal. The frequencies of the band gap region correspond to wavelengths between $1.4-1.5\mu m$. The defect mode allows for the propagation of electromagnetic waves in the waveguide. The localization of electromagnetic wave is studied for a wavelength of $\lambda = 1.43\mu m$.

Fig. 7 shows the ideal waveguide structure. All radii are equal and the filling factor is $f = 0.37$ for all rods. Propagation of electromagnetic waves is symmetric, i.e. the transmission is equal in both branches. T

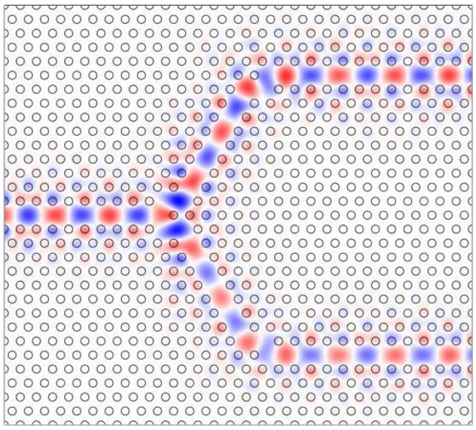

Figure 9: Y-waveguide like in Fig. 7, but the filling factor has a Gaussian distribution with mean value $f = 0.37$.

The waveguide in Fig. 8 contains rods with different radii. The radii have the distribution given in Fig. 4. The propagation of EM-waves becomes asymmetric. The waveguide of Fig. 9 has equal radii for all rods but the filling factor varies. The influence of fluctuations of the filling factor is smaller than deviations of the radii from the ideal value.

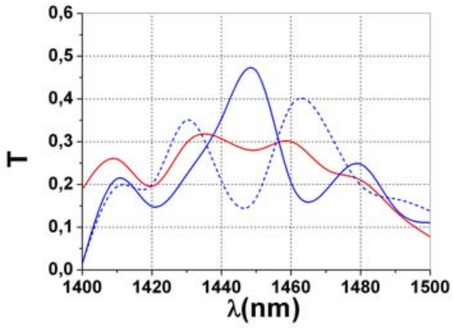

Figure 10: Transmission coefficient for the ideal structure (red) and the two branches (blue) for structure with Gaussian distribution of radii.

The transmission coefficient T for these cases has been calculated. The mean value of total transmittance is not very large ($T_{sum} = 0.6$ for the ideal structure) while the contrast of permittivity between rods and medium is small. For all the calculations $\Im\varepsilon = 0$ is assumed. For real systems the imaginary part is different from zero. Calculations of transmission for this case were also performed. The mean value of T tends to zero for all wavelengths. The reason of this results is absorbtion of EM waves by the metallodielectric rods.

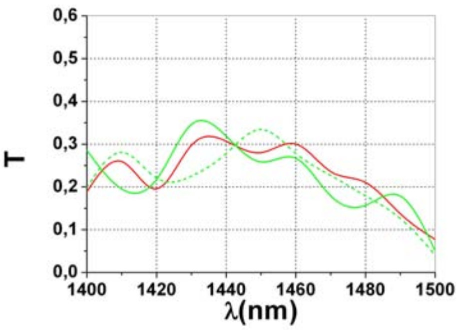

Figure 11: Transmission coefficient for ideal structure (red) and the two branches (green) for structure with Gaussian distribution of filling factor.

6 Conclusion

The finite element method (FEM) allows to calculate the effective permittivity of nanocomposite materials. Due to the method particles of any shape can be considered in the calculations. The effective permittivity is calculated as an average over many ensembles with random positions of the nanoparticles. Based on calculate effective permittivities waveguides in photonic/plamonic structures can be considered. The influence of deviations from the ideal structures like those experimentally obtained is studied numerically.

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