STUDY OF SURFACE PLASMON RESONANCE STRUCTURE WITH As_2S_3 AMORPHOUS CHALCOGENIDE COMPOUND WAVEGUIDE

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In this paper we present theoretical and experimental results regarding the analysis of optimum conditions to couple the light within a BK7-Au-As2S3-Air plasmonic compound. We first determined the characteristic equation for the plasmonic waveguide structure consisting in four thin layers by solving the system of Helmholtz equations and applying continuity conditions at the three interfaces of the structure. Subsequently, numerical calculations have been done to obtain the propagation constant and the field distribution within the structure layers. The plasmonic mode confined to the metal interface corresponds to TM0 waveguide mode. The structure supports higher order waveguide modes which can be exited, for several dielectric thicknesses, using low refractive index prism. The investigations emphasize that the value of the resonance angle is in very good accordance with our theoretical findings. Plasmonic waveguide which contains As2S3 as active optical material is a promising configuration for optical memory and switching devices due to well-known photo-induced phenomena in such materials.

Keywords: Surface Plasmon Resonance, Amorphous Chalcogenide Materials, As_2S_3 thin films.

1. Introduction

The ability of light to transmit information is enormous, due to the fact that the light frequency as the electromagnetic wave overpass 100 THz. Beyond of this, in optics there exists the simple possibility of information parallel processing by Fourier transform with the aid of a lens. As to date the technology of the optical fiber fabrication with losses smaller than 0.2 dB/km is already realized. One can foresee human desire to use the light as support for the transmission of information. Regrettably, the photonic devices dimensions are restricted from the miniaturization by diffraction of light limit, these one being established for the conventional optical devices at a level of half of the wavelength. In the visible spectral range diffraction limit constitutes on cca 250 nm, which is very much when compared to the actual electronic component

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dimensions, and which have already reached values less than 100 nm. There is a very large interest for metal-insulator structures, because they support surface plasmon-polariton resonance which may confine the light near surface at dimensions shorter than the wavelength. This phenomenon led very soon to the development of plasmonic sensors [1-15]. As was demonstrated earlier, the surface plasmon resonance (SPR) is very sensitive, up to 10^{-5} changes of the refractive index. The results being particularly impressive in the case of the biological selective sensors [16,18], the chalcogenide glasses are used for the determination of blood group [17]. The light irradiation enhances the efficiency of mass-transport in amorphous chalcogenide materials [19-21] due to localized plasmons in the presence of gold nanoparticles. Chalcogenide glasses, like As2S3 are promising media for active plasmonic devices [22]. Our paper proposes to develop the characteristic equation in a general form for the four layers IMII plasmonic structures. Subsequently, we present several numerical simulations and experimental results concerning surface plasmon resonance in four layers (SPR) configuration which contains low refractive index prism, gold film and high refractive index film made of amorphous As2S3 as a waveguide. The experimental results regarding coupling of the light with plasmonic waveguide modes made of amorphous As2S3 are presented.

2. Surface plasmon-polaritons, basic of phenomenon

Surface plasmon-polariton (SPPs) are electromagnetic waves that represent the coupling of the electromagnetic wave with the free oscillations of the electrical charges from the conductor. The equations which describe the phenomenon are derived from the Maxwell's equations. From the Maxwell's equations, assuming a harmonic time dependence of the field, the Helmholtz type wave equation can be obtained for the electric field:

$$\nabla^2 \vec{E}(\vec{r}) + \frac{\varepsilon\omega^2}{c^2} \vec{E}(\vec{r}) = 0 \tag{1}$$

where the dielectric constant ε of the structure depends on the spatial coordinate. If the waveguide layers are oriented along the z direction, x is along the transverse profile of the waveguide and the y is perpendicular to the incidence plane, we can derive the following wave equation for TE modes:

$$\frac{\partial^2 \vec{E}_y}{\partial x^2} + \left(k_0^2 \varepsilon(x) - \beta^2\right)\vec{E}_y = 0 \tag{2}$$

where $k_o = \omega/c$ is the wave vector of the propagating wave in vacuum and the dielectric constant is a function of x transverse position. The complex parameter $\beta = k_z$ is called the propagation constant of the traveling waves.

A similar equation can be derived for the magnetic field. Thus, the wave equation for TM modes will be:

$$\frac{\partial^2 \vec{H}_y}{\partial x^2} + \left(k_0^2 \varepsilon - \beta^2\right)\vec{H}_y = 0 \tag{3}$$

The simplest geometry which sustains surface plasmon polaritons (SPPs) is that of a single flat interface between a dielectric and a metal. Then, the solution of equation (3) leads to the dispersion equation $\beta = f(\omega)$:

$$\beta = \frac{\omega_{sp}}{c_0}\sqrt{\frac{\varepsilon_{r,1}\varepsilon_{r,2}}{\varepsilon_{r,2} + \varepsilon_{r,1}}} \tag{4}$$

$$\varepsilon_{r,2} = 1 - \frac{\omega_p^2}{\omega_{sp}^2}$$

The amplitude of the wave decreases exponentially with distance from the interface, as presented in Fig. 1.

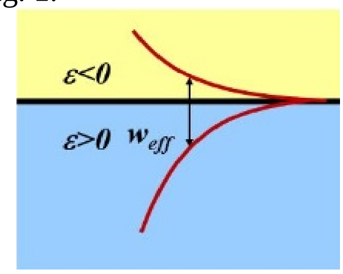


Fig.1. Exponential amplitude decay from interface

Two conditions are needed so that the metal-dielectric interface to supports SPPs [23]:

a) SPPs can be excited only by TM polarized incidence wave (no surface modes can exist for TE polarization). For TM polarization, the electric field has a component perpendicular to the insulator-metal (IM) interface, that can cause the oscillation of free electrons.

b) The real parts of the dielectric constant of the metal ε_{1r} and dielectric ε_{2r} are of opposite sign and satisfy the conditions: Re $\{\varepsilon_{1r}\} < -\varepsilon_{2r}$. The amplitude of the electromagnetic field decreases exponentially when departing from the IM interface. The penetration depth is around ½ of the wavelength in dielectric (that is 250-300 nm) and 10-15 nm in metal. Thus, the electromagnetic field can propagate along the metal surface, being strongly confined at the IM interface.

In surface plasmon resonance (SPR), p-polarized (i.e. TM polarized) incident light excites an electron-density surface plasmon wave (SPW) along the metal–dielectric interface by satisfying plasmon resonance condition:

$$k_0 n_p \sin(\theta) = \frac{\omega}{c}\sqrt{\frac{\varepsilon(\omega)}{\varepsilon(\omega)+1}} \tag{5}$$

The existence of surface wave was predicted long time ago. However, the modern beginning of this domain was put forward in the 1968 year, by Kretschmann and Raether [24], which purposed the method of plasmonic surface waves excitation by evanescent waves. Using of prism and incident beam in total reflection conditions permits matching of wave vectors for both: the electromagnetic wave and surface plasmon polariton wave. Strong interaction take place at a special incidence angle and the SPW can be excited at nanoscale level.

The reflection coefficient of the IMII type structure containing four layers (corresponding to Prism-Metal-Chalcogenide-Air configurations) can be derived by using Fresnel formula for light reflection at the layer's interfaces. The reflectivity Rp for the p-polarized wave in such case may be calculated by using a Johnson-matrix approach:

$$M = \prod_{k=2}^{N-1} M_k$$

$$M_k = \begin{bmatrix} \cos\beta_k & (-i\sin\beta_k)/q_k \\ (-i\sin\beta_k)q_k & \cos\beta_k \end{bmatrix} \tag{6}$$

$$R_p = \left| \frac{(M_{11} + M_{12}q_N)q_1 - (M_{21} + M_{22}q_N)}{(M_{11} + M_{12}q_N)q_1 + (M_{21} + M_{22}q_N)} \right|^2$$

where M_{11} , etc. are the components of M-matrix. The method is applicable to any number of layers.

Similar equations can be derived for the reflectivity Rs for the s-polarized wave.

3. Characteristic equations for four layers structure

Several SPR optical sensing structures were developed by using 4-layer structures in which the ambient medium (e.g. air) is the last one. In a recent investigation the authors [10] proposed and experimentally demonstrated the SPR light modulation by use Ga-La-S light sensitive vitreous film. Amorphous chalcogenide materials have many applications in modern optoelectronics due to photo-induced modifications of optical constants.

However, the chalcogenide materials have high value of refractive index, for example (2.45-2.50) in case of As_2S_3, and it is difficult to select a material for prism, as it needs the refractive index to be higher. However, the chalcogenide film can form a planar waveguide. The different modes of the waveguide may have different propagation constants, some of which may satisfy the resonance conditions.

We have studied the SPR and the sensibility to the refractive index changes in 4-layer structure which contains As_2S_3 chalcogenide films. Photo-induced changes of the refractive index may destroy the resonance conditions and thus a large variation in reflected signal intensity will occur.

The structure configuration and the refractive indices distribution are presented in Fig. 2.

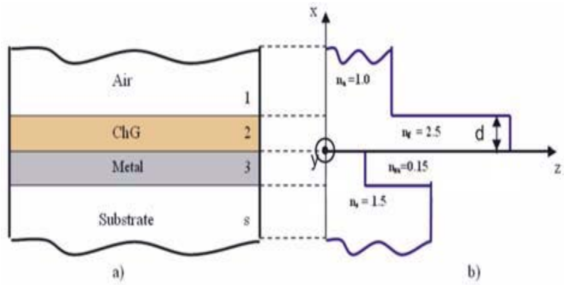


Fig. 2. SPR in four layers configuration.
a) The structure. b) The refractive indices distribution.

The plasmonic waveguides contain at least one metallic medium that has a complex-valued dielectric constant. As a result, the waveguide propagation constants are complex numbers. Finding the numerical solutions for the propagation constants within these structures is cumbersome. So that finding of a suitable analytical form for the characteristic equation, when is possible, appears to be a necessity.

The way is to solve the Helmholtz wave equation (2 or 3) that results from the Maxwell's equations. The equation is valid for each layer and the continuity conditions should be applied. The following formula was obtained, which is the characteristic equation enabling the identification of $\omega = f(\beta)$ features:

$$\left(\frac{1+\frac{r_1-r_4}{r_1+r_4}e^{-2k_1\cdot a}}{1+\frac{r_2+r_3}{r_2-r_3}e^{2k_2 d}}\right)=\left(\frac{r_1}{r_2}\right)\cdot\left(\frac{1-\frac{r_1-r_4}{r_1+r_4}e^{-2k_1\cdot a}}{1-\frac{r_2+r_3}{r_2-r_3}e^{2k_2 d}}\right). \qquad (7)$$

Here, $k_i = \sqrt{\beta^2 - k_o^2\varepsilon_i}$, $i = 1,2,3,4$, represent the wave vectors, $k_0 = \omega/c$ is the wave vector of the propagating wave in vacuum, $r_i=k_i/\varepsilon_i$, $r_{ij}=r_i/r_j$, $k_{ij}=k_i/k_j$, a and d are the thicknesses of the Au and respectively chalcogenide films.

After finding solutions and making transformations with complex numbers, it has been obtained the simplest form of the characteristic equation, used for numerical simulations, namely:

$$d = \frac{\arctan(f) + \arctan(g) + m \cdot \pi}{\bar{k}_2}$$

$$\text{with } f = \frac{t-g}{1+tg} \text{ and } t = \tan(\bar{k}_2 \cdot d) = \frac{f+g}{1-fg}, \tag{8}$$

where f and g depend on the layer's physical parameters and m is an integer, which represents the modes number. The above shown characteristic equation for the four layers structure has the same form with the equation obtained earlier by Marcuse [25] for the three-layer lossless waveguides. The difference consists of more general values for the coefficients f and g. Also, the numerical simulations have been done in the field of complex numbers.

4. Numerical simulations

The characteristic equation was implemented in MATLAB to find the propagation constant. The propagation constant was inserted into the characteristic equation to calculate k_i and, subsequently, the field intensities for the TM modes.

We considered that the incident laser wavelength is 632.8 nm and the four layers off the IMII structure are as follows: layer 4-BK77, layer 1-Au, layer 2-As_2S_3, layer 3-Air.

The thickness of the Au film is considered a=50 nm and the chalcogenide film thickness (d) is varied between 200 and 16000 nm. In our simulations we considered the refractive index of the chalcogenide film $n_2 = 2.45$.

The gold optical constants are taken from Rakic [26] measurements. The real part o of the effective refractive index $N_{eff} = \beta/k_0$ as a function of chalcogenide film thickness d for the TM modes are presented in Fig. 3 (top).

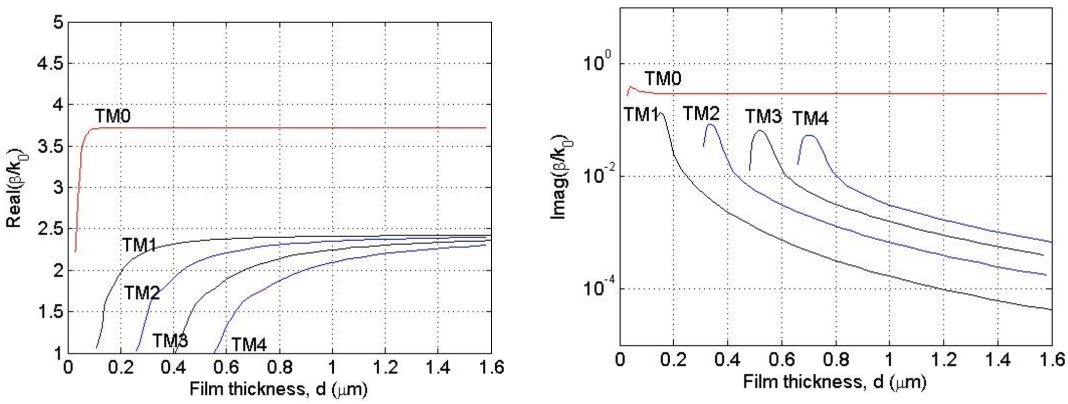


Fig.3. The propagation constants for TM modes: real part (top) and imaginary part (below) of propagation constant.

The mode that can be coupled in planar waveguide by a BK7 prism depend on the As_2S_3 thickness. Only the modes which have the effective refractive index in the range [1, 1.51] can be excited. The higher values could not be excited due to the requirement of synchronization which implies the condition $n_p \sin\theta = N_{eff}$. For instance, the mode TM0 may never be excited, and only one mode can be coupled for the selected chalcogenide film thickness with the BK7 prism.

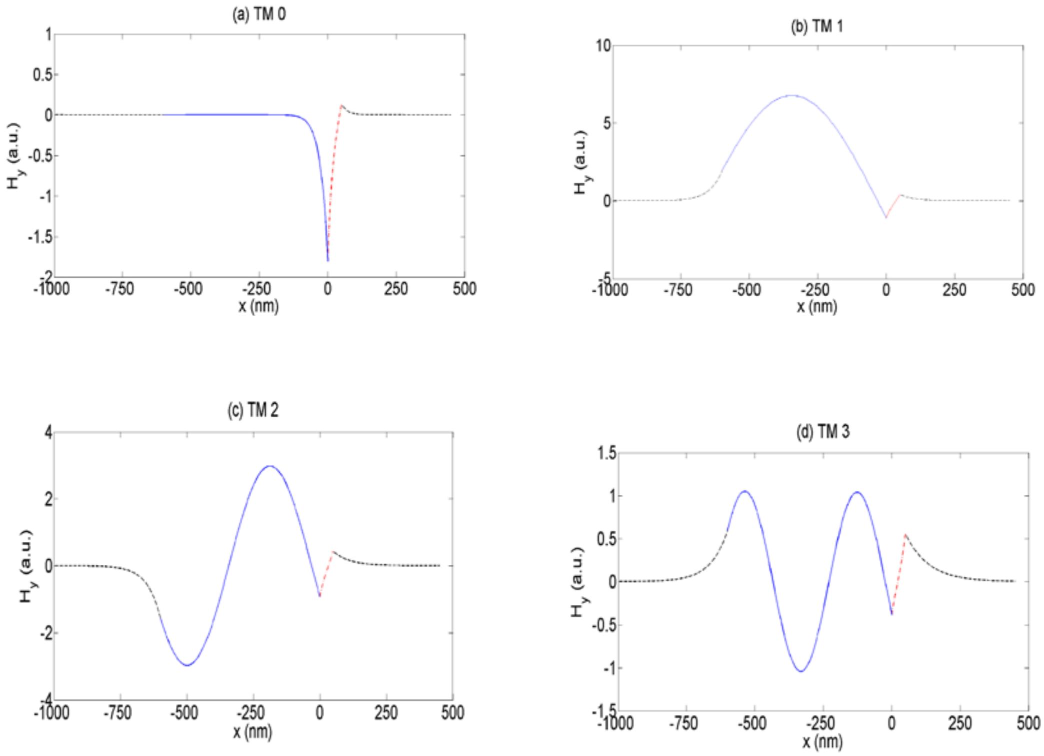
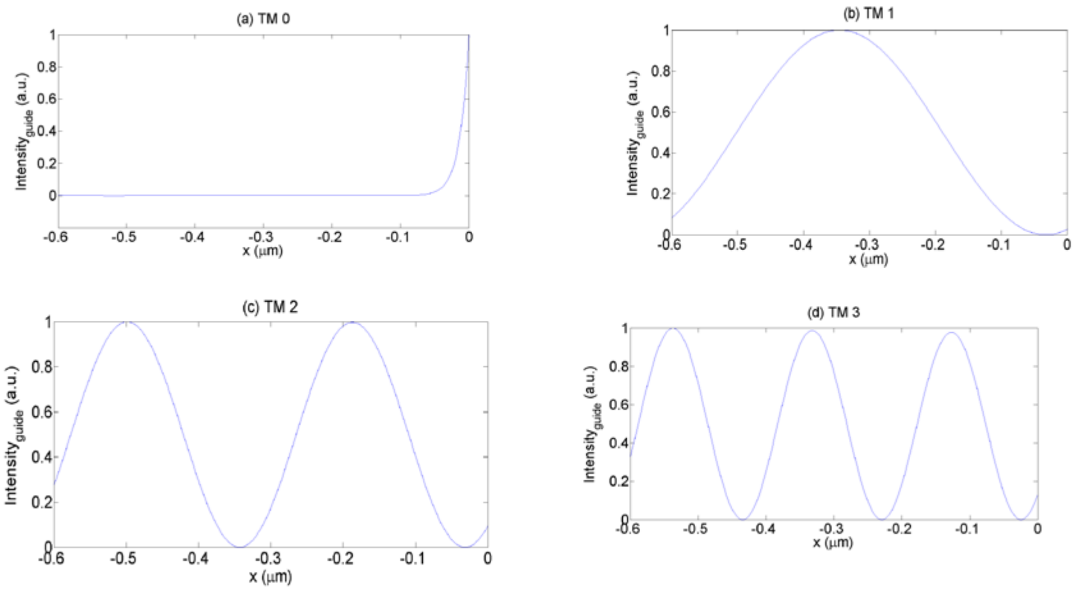


Fig.4. The magnetic field within the four regions of the structure for different TM modes: TM 0 (a), TM1 (b), TM2 (c) and TM3 (d) modes.

The simulation presented in Fig. 4 correspond to d = 600 nm. The blue curves represent the field within the chalcogenide film and the red curves represent the field within the metallic layer. We can see that TM0 is confined to the metal interface. The higher TM modes may have non-zero field at the chalcogenide-air interface. The simulation of light intensity distribution within the 600 nm thick chalcogenide layer is presented below in Fig. 5.



.Fig.5. Light intensity distribution within the 600 nm thick chalcogenide layer for TM 0 (a), TM1 (b), TM2 (c) and TM3 (d) modes. The simulation parameters were: a = 50 nm, λ = 632.8 nm

5. Experimental results

An experimental setup, shown in Fig. 6, was fitted to check the validity of thee model. The polarization of the laser is set in the plane of incidence so that TM modes can be investigated. The plasmonic structure consists of a chipset made up of a glass slide on which a thin (~ 50 nm thickness) gold film is deposited.

We used a XANTTEC bare gold film. The real thickness of the gold film measured by profilometer was 46 nm. AFM micros copy analysis showed a good homogeneity of the surface of the gold film and a roughness of 2.0-2.55 nm.

Over the gold film, an amorphous chalcogenide As_2S_3 film was deposited by thermal evaporation in vacuum of $6.6\cdot10^{-4}$ Pa. As_2S_3 granular material for evaporation was placed in the tantalum resistive boat.

The As_2S_3 film thickness measured by the profilometer varies between 600 to 630 nm on the 110x10 mm surface. This complex Au-As_2S_3 chipset is attached to the BK7 prism by using Cargile microscope immersion oil. SPR resonance curves are presented on Fig.7: Red curve-simulation and blue curve-experiment.

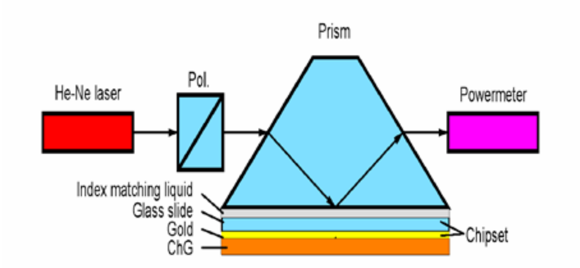


Fig.6. Experimental setup for the investigation of SPR.

The proposed configuration has two advantages: a) metal-dielectric structure does not need to be deposited directly on the prism base, which is more expensive than a slide and b) there is no need to deposit the gold film - it is used a good quality commercial one.

The prism with the attached plasmonic structure is mounted on a rotating with the axis perpendicular to the plan of the figure.

Thorlabs NR360S continuous rotation stage with stepper motor actuator and 1 arcsec resolution was used in order to ensure the measurement of the plasmon resonance peak with the requirement accuracy.

The radiation reflected by the plasmonic structure is measured by a digital powermeter model Gentec Solo PE. Experimental graph representing the reflected light intensity depending on the incidence angle on the prism base is shown in Fig.7 (blue). The minimum reflectance corresponds to the incidence angle of 48.2^0.

The experimental results were provided in two steps: In the first step the structure used both for calculations and for experiments was a three layers configuration: BK7 (prism) - Au (46 nm film) - Air. The glass BK7 is a well-known optical material with the refractive index 1.5151.

The measured (by profilometer) thickness of the gold film was 46 nm. The gold film was a commercial one dedicated to plasmonic bio-sensor applications. The film was obtained by the electron gun evaporation. The optical constants (n, k) of gold may be found from different sources.

In the second step the determined values of the gold optical constants (n = 0.196 and k = 3.256) was used to investigate the four layers plasmonic resonance structure.

The best fitted curve presented in Fig. 7, the red curve, was obtained by using the following parameters for chalcogenide film: the thickness 600 nm (the profilometer data indicated 626 nm ± 2%) and the refractive index 2.43.

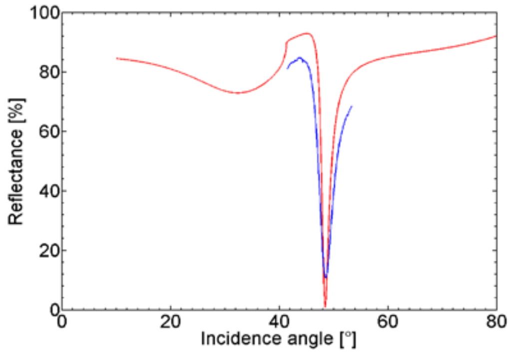


Fig.7. SPR picks: Red curve: simulation and blue – experiment

This value of fitted refractive index complies with the value 2.41 measured by waveguide m-line spectroscopy [27]. According to the simulations presented in Fig. 3 the measured resonance angle corresponds to the TM4 waveguide mode.

6. Conclusions

The characteristic equation was obtained analytically in the case of a Dielectric-Metal-Dielectric-Air four layers structure. We demonstrated that the characteristic equation has a similar form with the characteristic equation written by Marcuse for the three-layered step refractive index waveguides.

The characteristic equation was solved numerically. As a result, we determined the propagation constant β as a function of chalcogenide film thickness for BK7-Au-As_2S_3-Air plasmonic structure.

The four layers structure becomes similar with a three layers structure when the metallic layer thickness is increased up to 100 nm.

The experiment show that the reflectance of light supports a dip and sharp resonance minimum (Fig.7) at a certain angle. The dip value and position depend on the optical constants of the metal and dielectric.

Consider the metal optical properties as being constant, the SPR angle depends only on the dielectric constants connected to the complex refractive index by the relation $\varepsilon = (n - jk)^2$. The other particularity that can be mentioned is the sensibility only to the properties of the medium situated close to the interface. This phenomenon can be used for sensing optical devices.

The plasmonic structure containing a finite thick metal film and a finite thick dielectric film supports TM modes. For some well-established thicknesses, the plasmonic structure with high refractive indexes (n~2.45) materials like As_2S_3 may be excited by using a low refractive index prism made of BK7 glass. This allows the use of common microscope slide with deposited gold films and link them together using immersion oil.

Unlike the lossless waveguide where the fundamental mode is nearly centered for the plasmonic waveguides the TM0 mode is close to the metal interface. The higher modes as TM3, for example, have considerable portion of the field linked to the external dielectric interface. The importance of using chalcogenide amorphous films lies in the fact that they can be used as light sensitive optical material which supports photo-induced changes of the refractive index.

The provided experiment has demonstrated the realization of such structures.

The structure presents interest for the development of the optical switches, 2D memories or sensing elements that use optical active materials such as amorphous chalcogenides.

Acknowledgments

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