The method of electro-induced lithography for Tamm plasmon observation

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Abstract. Here we reported the results on a formation of a multilayered metallic lithographic pattern repeating the probe trajectory of an atomic force microscope (AFM). The features of a growth on the surface of a Bragg mirror have been studied in the details. The precise control of the topological parameters of the metal track used as a waveguide for propagation of light-matter coupling states provides the control of Tamm plasmon energy. The possible applications of the obtained plasmon-polariton structures for nanophotonics and plasmonics were discussed.

1. Introduction

The evolution of modern computer technologies requires the development of new devices based on new physical principles that makes it possible to speed up the execution of logical operations in a significant way. One of the possible solutions to solve this problem is a creation of completely optical elements and schemes. Recently, there has been an increasing interest in creating a base of new elements for computing devices designed to replace existing electronic components by forming metallic nanostructures of noble metals that have unique optical properties when localized plasmon resonances are excited. A special interest in the study of such structures is associated with numerous applications [1-3]. The localized mode of the electromagnetic field is called the Tamm plasmon (TP) by analogy with the localized electronic states on the crystal surface predicted by Tamm [4]. Unlike the ordinary surface plasmon, Tamm plasmon can be directly optically excited at any angle of incidence both in TM and in TE polarization [5], since its dispersion in the plane lies entirely in the light cone. But the main advantage of Tamm plasmons is associated with the low level of losses that arises from the fact that the field is concentrated mainly at the depth of the Bragg mirror, due to which the width of the Tamm plasmon line is an order of magnitude lower than that of the surface plasmon. Excitation of localized plasmon resonances of metallic nanoparticles or clusters in the visible and near infrared regions of the spectrum, characterized by an increase in local electromagnetic fields, leads to an intensification in nonlinear optical effects, which allow us to speak of the possibility of using localized states to create active photonic elements. Thus, the usage of plasmon-polariton nanostructures for optical elements will significantly reduce the size of integrated schemes in which the excitation and directed propagation of surface electromagnetic waves will be realized.
The described properties of TE and TM of Tamm modes demonstrate a strong energy dependence on the geometric parameters of the waveguide. In this connection, there is a need for precise control of these parameters, and, consequently, new high-precision methods of creating metal structures are needed.

2. The method of electro-induced lithography

There is a plenty of methods for structure creation on some surfaces according to a pre-prepared template. However, the most of them requires an expensive equipment, high-precision leveling operations, exposure, etching, etc. A probe lithography is a well-known and relatively inexpensive method for a forming of nanoscale structures. A lithographic pattern in our case has been formed during a dissociation procedure of chemical compounds under effect of applied external field by means of a scanning probe microscope on the basis of the platform of NTEGRA Aura probe nanolaboratory in the contact mode of AFM operation [6].

Bragg mirrors (BM) containing of 18-20 GaAs / AlGaAs layers were used as a substrate for the local formation of a surface nanorelief. The cleaned substrates were placed into a container isolated from solar light to saturate with an alcohol solution of Au or Ag metal salts. The initial alcohol index was of 96%. The saturation time has been varied from 1 hour to 7 days, changing the saturation degree of a substrate. The probe and a substrate played electrode roles that initiated an electrochemical reaction in the water meniscus under a conducting needle (see Fig. 1).

![Figure 1](image-url)

**Figure 1.** The scheme of a structure growth during an electro-induced lithography: a) an initial position of the elements, where (1) – probe; (2) – BM; (3) – Me\(_x\)(NO\(_3\))\(_y\) salt dissolved in C\(_2\)H\(_5\)OH solution; (4) – the adsorbed water layer on both surfaces of a substrate as well as a conductive probe; b) the probe and the substrate are in contact during the lithographic procedure: the alcoholic solution of a metal salt dissociates with decomposition to the formation of water and metallic ions (5), which are attracted to the substrate by a potential difference and a volatile gas NO\(_2\); (6). On the surface of the substrate, the ions are reduced to Me/Me\(_2\)O\(_x\) (5); c) the result of the lithographic operation.

The precision of this method is due to the accuracy of the movement of the AFM probe that is as 0.1-1 nm in the plane of a sample. The main advantage of the method is a high-precision deposition with a nanometer level of a spatial resolution due to the formation of a lithographic pattern, formed directly under the tip of the probe. The process of a deposition of a metal nanolayer is determined by the value of a probe-substrate bias voltage, which is most often set in the range of 6-10 V, as well as the scanning speed. However, the minimum size of the growing element in the scanning plane is limited by the diameter of the water meniscus formed between the substrate and the probe where the chemical reaction proceeds. The increase of a thickness of a metal structure is achieved by step by step repeating of the lithography process, the duration of the pulses applied to the needle and the intensity of the electric field, which depends on the amplitude of the voltage pulse.
3. Results and its discussion
An important parameter affecting the growth of metal structures is a concentration of the saline solution of a metal. Within the framework of this work, five kinds of solutions of different ratio between distilled water to alcohol and metallic salt of AgNO₃ were prepared to determine the most suitable composition. To form the lithographic pattern, a single pass vector lithography was used at a speed of 0.03 Hz (see Fig. 2). The results were recorded from a sample of five measurements, the data was averaged and listed in table 1.

![Image of metallic nanopatterns obtained at different concentrations of C₂H₅OH, other parameters remain the same: (a) 48%; (b) 72%; (c) 24%.

The width of the resulting structures changes as well as the height with the variation in the percentage composition of alcohol in the solution. This effect is associated with an increase in the amount of water in the solution, and hence the thickness of the adsorbed water layer on the surface of the substrate, which in turn has an effect on the formation of an aqueous meniscus between the substrate and the probe. As a result, there is a strong broadening of the lines of the lithographic track and blurring of the boundaries (Fig. 2(c)).

![Image of template for lithography. Measured parameters are a height (h) and a width (w).

### Table 1. Influence of the concentration of AgNO₃ on the growth of structure.

| №  | C₂H₅OH, % | Weight AgNO₃, g | h, nm | w, nm |
|----|-----------|-----------------|------|------|
| 1  | 48        | 0.278           | 1.6  | 89   |
| 2  | 72        | 0.278           | 2    | 205  |
| 3  | 24        | 0.278           | 3    | 256  |
| 4  | 72        | 0.160           | -    | -    |
| 5  | 72        | 0.398           | 3    | -    |

At the first step we determined the optimum ratio between constituent elements of a saturation solution. We formed a homogeneous lithographic pattern with keen boundaries in a case of a percentage composition of alcohol of 72% in a solution to a salt mass of 0.278 g that used as optimum
values of the height and width of the grown nanolayer in future. The significant decreasing of a mass of silver salt in solution stopped a nanostructure growth because of the lower concentration of the dissociating substance in the alcohol solution. However, a critical increasing of salt concentration in solution leaded to an irreversible effect; the surface of a substrate became extremely sensitive to any applied external action that often leaded to the uncontrollable formation of a thin continuous film on the entire surface of the BM, rather than a true lithographic pattern under effect of a moving AFM probe.

The Figure 4 shows a 3D view of a typical metallic nanostructure of a height of about 2.5 nm that was grown in a raster lithography regime according to a pre-prepared template at a scanning speed of 0.25 Hz.

The resulting layer is sufficiently homogeneous of $R_a = 0.2$ nm. Multiple repetition of the pattern trajectory makes it possible to vary of thickness of a multilayer metallic nanostructure. For nanolayer formation we have used a optimum ratio between an alcoholic solution of 72% and the mass of the silver salt of 0.278 g. The lithography has been made on a vector mode. The height of the resulting structure was 1.4 nm after the first pass-lithography, in further the structure growth occurred linearly depending on a number of passes with a subsequent increasing in the layer thickness of 0.1 nm. This approach is perfectly suited for a precise control in thin metal nanolayers production.

To observe the TP propagation three samples with extended metal waveguide on the BM surface have been grown: 5 nm, 50 nm, 500 nm thicknesses. The incidence angle of the exciting radiation on the BM-metal interface was about 22.5° by the lateral illumination of the sample (Fig. 5) that allowed us to observe TP-modes in a one-dimensional structure.
Figure 5. Simplified optical functional diagram of the NTEGRA Spectra PNL with the side illumination module employed: 1 – triple input unit; 2, 3, 4, 6, 7 – mirrors; 5÷97% mirror; 8 – beam expander; 9 – turret with three sets of changeable edge or notch filters; 10, 11 – lenses; 12, 13 – adjustable pinhole; 14 – diffraction gratings; 15, 16 – adjustable density neutral filter; 17, 18 – polarizer; 19 – filter cartridge; 20 – OTS; 21 – focusing module; PMT – photomultiplier tube; CCD – camera; APD – avalanche photodiode; PMT - photoelectric multiplier; CCD - CCD camera; APD - avalanche photodiode; SPM – SPM.

Excitation of surface states at the edges of the band was provided in the near field mode using a scanning near-field optical microscope Ntegra Spectra produced by NT-MDT company. The Figure 6 shows the distribution of electromagnetic waves in the near-field of the metal tracks of different thicknesses. The metallic layer of 5 nm height was actually transparent in the used geometry for the exciting radiation (Fig. 6(a)) that leaded to a total reflection of a light from the BM substrate. The thickest metallic waveguide of 500 nm was not suitable for excitation of Tamm plasmon states (Fig. 6(c)). In this case the reflection of the pump radiation predominantly occurred from the metallic layer and there were only small visible fluctuations in the field intensity due to the presence of inhomogeneities on the metallic layer surface. According to the theoretical modelling the optimum height for TP propagation through a metallic layer of 50 nm. One can see the significant manifestations of the light-field intensity maxima on the Figure 6(b) that correspond to the formation of light-matter coupling states.

Figure 6. The distribution of the electromagnetic wave response recorded in the near-field of the metallic waveguide with different thickness: (a) 5 nm; (b) 50 nm; (c) 500 nm.

4. Conclusion
In this paper we have applied the method of electro-induced lithography to prove it as reliable and technically simple tool for performing lithographic operations which does not requires the creation of special conditions such as high humidity, a special substrate with a thin layer of well oxidized metal, exposure, etching, etc. The variation in the solution concentration as the saturation time of the substrate affects to the quality of the resulting metallic nanolayers. The precise control of the topological parameters of the metal multilayer track would possess a possibility to provide control of the TP energy. We offer using of obtained nanostructures demonstrating unique optical effects for creating a new-generation element base: high-speed data transmissions, optical computer elements and optical data recording, processing and displaying systems.

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