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Highly directional plasmonic nanolaser based on high-performance noble metal film photonic crystal

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ABSTRACT

A fundamental problem in the integration of photonic elements is the problem of the light localization and the creation of nanolocalized laser sources of radiation. A new approach in the miniaturization of lasers is the approach based on using plasmon fields instead of photon fields. Plasmons arise from the interaction of the oscillations of the electron density and the electromagnetic fields that excite them. Accordingly, the electromagnetic effects caused by these fields occur in the subwavelength region near the surfaces, namely, in the nanometer range. Therefore, the approach allows to overcome the diffraction limitation on the laser size. Plasmonic nanolaser is a nanoscale (at least in one dimension) quantum generator of nanolocalized coherent plasmon fields. The nanoscopic in all three dimensions plasmon nanolaser has a different name: SPASER (Surface Plasmon Amplification by Stimulated Emission of Radiation). It is based on patterned metal film. The precision of formed structures and the dielectric properties of the metal are critical factors in determining any plasmonic device performance. Surface and morphology inhomogeneities should be minimized to avoid SPP scattering during propagation and etching anisotropy. Moreover, the metal should have high conductivity and low optical absorption to enhance optical properties and reduce losses. Some researchers focused on developing new low-loss materials (nitrides, highly-doped semiconductors, semiconductors oxides, or two-dimensional materials), but silver and gold are the most commonly used materials in optics and plasmonics due the lowest optical losses in visible and near infrared wavelength range. Recently, we have presented plasmonic nanolaser built on ultra-smooth silver films. Nanoscale structure in metallic films are typically fabricated by a two-step process. Metals are first deposited using evaporation or sputtering on a substrate and then patterned with focused-ion-beam milling or e-beam lithography and dry etching. If the deposited films are polycrystalline, etch rates vary for different grain orientations and grain boundaries. Therefore, the patterned structures could differ from each other. One of the possible solutions is to deposit single-crystalline metals, which will be etched more uniformly and lead to precise structures. Another approach deals with large grain (>300 nm) polycrystalline film preparation. The fabricated silver films showed ultra-low losses (40 cm^{-1}). Built on it a plasmonic laser demonstrated the lasing at 628 nm with a linewidth of 1.7 nm and a directivity of 1.3.

Keywords: plasmonic film, silver, transparent substrate, SPASER, photonic crystal, high-performance film, e-beam deposition.

1. INTRODUCTION

Nanoscale localized sources of optical fields give benefits for numerous existing applications of nanoplasmonics. It could be used for ultrasensitive detection and spectroscopy, single-molecule sensitivity fluorescence, coupling of light to waveguide nanostructure [1]. However, achievement of a high quality plasmonic nanolaser is still challenging task so

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far, because of large metal cavity losses [2]. One of possible sensor implementations is nanolaser based on surface plasmon amplification by stimulated emission of radiation (SPASER).

Surface plasmon resonance (SPR) and localized surface plasmon resonance (LSPR) play important role in sensorics. They have found their application in medical diagnostics [3], environmental monitoring [4], and food safety [5]. Plasmonic sensors effects could be based on metal nanoparticles, metal nanostructures or their combinations.

The precision of plasmonic structures and materials optical properties are critical factors in determining the plasmonic devices performance. Plasmonic nanostructures are typically fabricated by a two-step process. First, metals are deposited by evaporation or sputtering on a substrate and then patterned with focused-ion-beam milling or e-beam lithography and dry etching.

High-energy focused ion beam (FIB technology) dopes film areas close to created topology, and etched film redeposition deteriorates the optical properties of formed structures. Moreover, devices for practical application require high aspect ratio structures and large area topologies on standard wafers. Basic FIB principles do not allow it to be an efficient instrument for these purposes. Nanoscale implementation of optically qualitative silver structures with precisely controlled shape and profile depth is a huge challenge. In this work, we propose to use e-beam lithography and dry etching instead of FIB.

In addition, the metal should have high conductivity and low optical absorption to enhance optical properties and reduce losses. Silver and gold are the most commonly used materials in optics and plasmonics due to the lowest optical losses in a visible and near infrared wavelength ranges [6]. Moreover, surface and morphology inhomogeneities should be minimized to fabricate high quality device and to avoid SPP scattering during propagation and etching anisotropy.

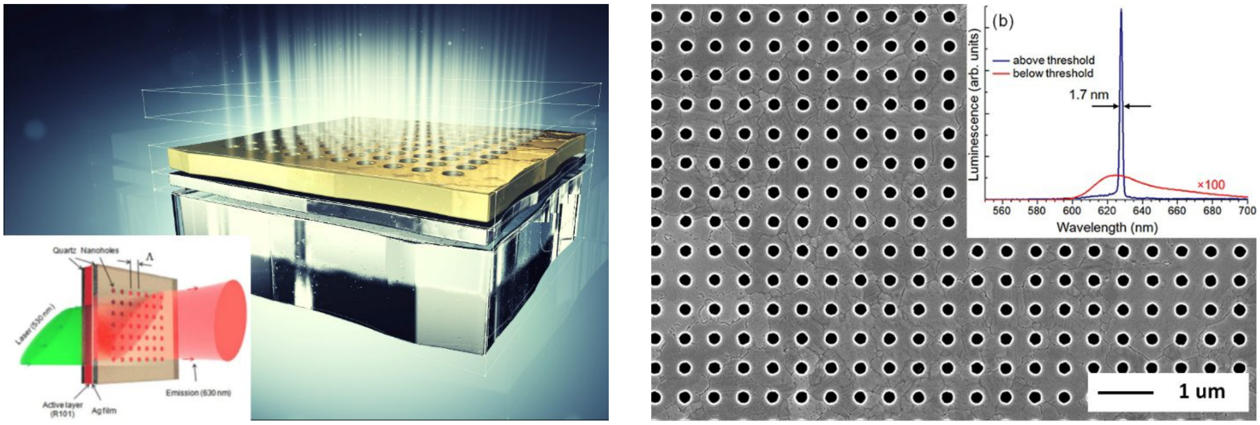
To satisfy this requirements, we have already performed plasmonic device fabrication based on a single-crystalline silver films [7]. This technology allows fabricating continuous, damage-free, single-crystalline, with root mean square (RMS) roughness less than 0.5 nm and perfect optical properties at thicknesses down to 15 nm. However, deposition of a single-crystalline silver films used in this technology is compatible only with matched substrates of Si <100>, <110>, <111> and mica.

It is important to notice that a big part of plasmonic devices has to be fabricated on a transparent in the visible and near-IR ranges substrates, which are not compatible with a silver epitaxial growth. Especially, it is important for some sensors schemas, which require exposure from the other side of analyte supply [8, 9]. To fulfill this requirements, we focused our research on a noble thin films on transparent substrates: quartz and sapphire.

2. PLASMONIC NANOLASER BASED ON A SILVER FILM ON TRANSPARENT SUBSTRATE

Different successful SPASER experimental schemas have been reported [1, 2, 9]. In this article we perform fabrication process of SPASER [9] acting as a sensor (Figure 1a).

The fabricated SPASER is based on a planar waveguide made of a liquid DMSO (solution of R101 dye in dimethyl sulfoxide) layer deposited on a plasmonic crystal (Figure 1b). Array of nanoholes created in a 100-nm-thin silver film deposited onto a quartz substrate is used as a plasmonic crystal. The optimal parameters of the plasmonic crystal have been numerically calculated. To compensate the uncertainty in calculated values of the material parameters, we fabricated and studied structures with nanoholes of the varying pitch Λ (545, 555, 565, 575, and 585 nm) and diameter (150, 175, and 200 nm).



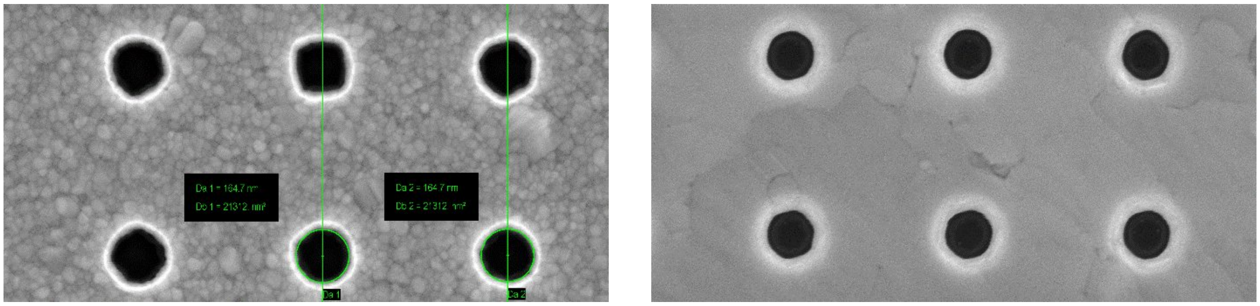
a)

b)

Figure 1 – a) Sensor SPASER model and operating scheme; b) fabricated array of holes SEM image and measured radiation spectra

The nanohole arrays in the silver films are fabricated by electron-beam lithography and dry etching (Figure 1). The array of holes is patterned on a 100-nm thick silver film, as it is shown in Fig. 1b. This film is deposited on the UV-grade quartz substrate (RMS < 1 nm).

The two alternative silver film structures were tested for SPASER fabrication: nanocrystalline with a grain size less than 30 nm (Figure 2a) and polycrystalline with large grains, mean grain size of which is more than 300 nm (Figure 2b).



a)

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Figure 2 – SEM image of holes: a) etched in nanocrystalline film; b) etched in polycrystalline film with large grains

Both films have a good optical properties (in comparison with published by Johnson and Christie (JC) [10], presented in Figure 5) and are useful for fabrication of nanostructures with precisely controlled shape. For 300-nm grain size, the shape of the thin film photonic crystal holes was still better than for 30-nm grain size film. Hole circularity factor calculated using ImageJ software is 0.90 comparing to 0.84. That is why, large grain structure was chosen to fabricate final device.

Fabrication

FIB is commonly used for such plasmonic structures in thin metal films. It is well known, that the main disadvantage of FIB is significant time of process. To increase the volume of fabricated samples e-beam lithography with RIE are used (Figure 3). The fabrication process is a classical planar technology. Nanoholes array was fabricated by reactive ion etching of silver film through spin-coated PMMA mask structured by 50kV e-beam lithography.

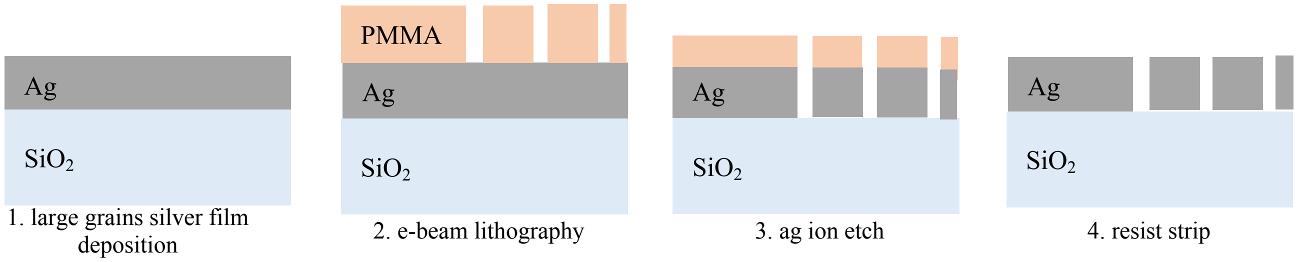
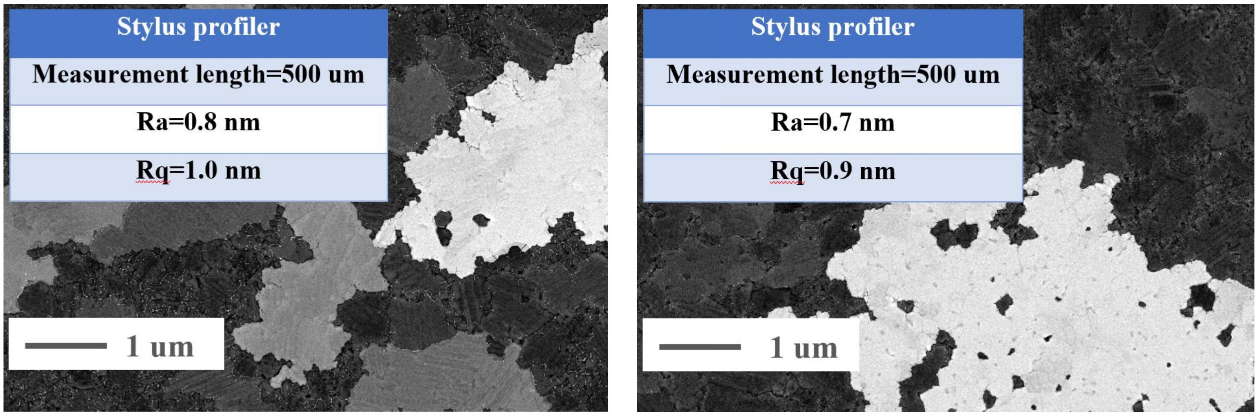


Figure 3 – SPASER fabrication scheme

First, silver thin film should be deposited on the quartz substrate. Deposition process includes electron-beam evaporation on the quartz substrate and subsequent annealing. Silver films surface morphology is monitored by scanning electron microscopy (SEM), showing that more than 90% surface filled with grains with linear size bigger than 300 nm (Figure 4). We have proposed the special annealing procedure, which allows increasing crystalline size and obtaining perfect roughness of the silver film. This procedure is compatible with both quartz and sapphire substrates (Figure 4). The silver films surface roughness is measured be means of stylus profilometry with 38 nm radius tip, yielding RMS roughness less than 1.2 nm. Optical properties are measured by spectroscopic ellipsometry (Figure 4).



Stylus profiler
Measurement length=500 um
Ra=0.8 nm
Rq=1.0 nm

Stylus profiler
Measurement length=500 um
Ra=0.7 nm
Rq=0.9 nm

a)

b)

Figure 4 – Annealed silver thin film SEM image : a) on quartz substrate b) on sapphire substrate

It is known, that silver thin films optical and plasmonic properties dramatically depends on its crystalline structure. Larger crystalline size provides better properties [11, 12]. We also observed it previously in our research [13].

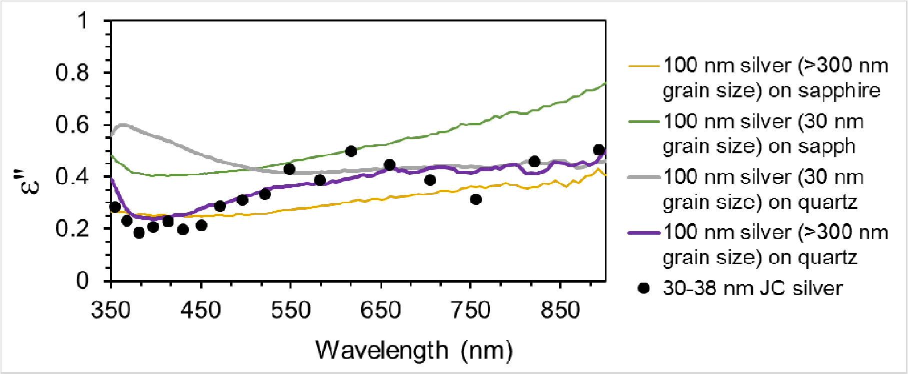


Figure 5 – Spectroscopic ellipsometry of fabricated films

An alternative way for fabricating of the high quality plasmonic films on substrates, which are not compatible with epitaxial growth, is a template-stripped technology [12]. It should be noted, that these technologies have several limitations, which complicates further e-beam lithography. Template stripping requires the use of optical adhesive to glue a film on a new substrate, which is obstructive for device fabrication.

The thin films with large grains are good for further device fabrication. A mean crystalline size of annealed silver thin film is 300 nm. It exceeds the typical SPASER hole size and allows to eliminate borders etching effects for most of the holes.

After deposition, the silver film is capped by a 10-nm-layer of SiO_2. The use of the SiO_2-film solves two problems: it protects silver from degradation and prevents quenching of excited dye molecules. The quenching leads to an undesirable heating of the silver film, which could lead to sample destruction.

Silver thin film is deposited on a diced and properly cleaned quartz samples. After film deposition a thin PMMA resist layer is spin-coated on it. Then, the samples are specially baked at low temperature, because standard softbake at 170 °C causes fast degradation of the thin silver film. On each sample, 12 structures with a few millimeters gap between them are exposed by e-beam lithography. Maximum size of the e-beam tool write field (500um x 500um) is used to save the maximum pitch accuracy. It helps to avoid stitching errors after e-beam tool stage movements. Pre-exposure alignment and calibration procedures allow avoiding distortion of holes shape and pitching errors caused by aberration of beam near the corners of a write field. It guarantees a high uniformity of elements array and consequently photonic crystal figure-of-merit.

For etching 2-step ICP silver etch process in argon plasma [7] with excellent defined regimes for each part of the film thickness that was used. It consists of 2-step etch. First step is an Ar-sputtering process, performed in the 100/300 W mode (ICP / RF power), that removes 75% of the film thickness and almost does not damage the PMMA resist mask. Provided etch selectivity of this process is approximately 0.8:1 (silver: PMMA). At the second step, a Ar-sputtering is carried out at higher ICP power 300/300 W (ICP / RF power) to the full thickness.

Photonic crystal fabrication technology provided to achieve the following SPASER parameters: radiation pattern extended in the direction perpendicular to the extension direction of the pumping region with the minimal directivity is 1.3˚, the orthogonal direction is 4.1˚ and a very narrow spectral line with the half-width of 1.7 nm emerges at 628 nm.

Experimental Section

The surface cleaning procedure is a crucial for preparing the high-quality silver films. Both the sapphire and quartz substrates are processed with the 2-step cleaning. First, the wafers are immersed into a hot (140˚C) solution of H_2SO_4:H_2O_2 (4:1) for 10 min, rinsed in DI water for 2 min and dried by nitrogen gun.

The silver films were deposited on UV-grade quartz substrates and C-cut sapphire substrates using 6 kW e-beam evaporator (Angstrom Engineering) with a base pressure lower than 3×10^{-8} Torr [14]. All the films were grown using 5N (99.999%) pure silver. Films were deposited with the rate of 1.0 Å s^{-1} measured with a quartz monitor at approximate source to substrate distance of 30 cm. Deposition is done in two steps. First, silver film is deposited on the substrate. At the second step evaporation is stopped and the substrate is heated up to 60-150˚C.

Dielectric functions of the silver films were measured by a multi-angle spectroscopic ellipsometer Sentech SER 800 (Sentech Instruments Measured spectral wavelength range was from 380 nm to 1000 nm. The spectra were acquired at four angles of incidence: 40˚, 50˚, 60˚, and 70˚. Calculation of dielectric functions was made by solving the inverse ellipsometric problem.

The stylus profiler KLA Tencor P17 was used. Durasharp stylus with 38 nm tip radius was used. All the measurements were done with 0.5 mg taping strength, scan rate was 2 $\mu m\cdot s^{-1}$,and the scanning line length was 20 µm.

In order to check the quality and uniformity of the deposited layers silver films surfaces after deposition were investigated by means of scanning electron microscope Zeiss Merlin with Gemini II column. All SEM images were obtained using in-lens detector and the accelerating voltage 5 kV and working distance from the sample to detector from 1 to 4 mm. Magnifications 3000, 7000, 15000 and 50000 were used to fully analyze samples.

For e-beam lithography Raith 50 kV e-beam tool with maximum write field of 500 µm with high speed pattern generator is used. ICP Ar-sputtering etch is done with Oxford Instruments PlasmaPro100 system.

CONCLUSION

We have implemented fabrication of high quality silver thin films on transparent substrates. More than 90% of silver films surface have linear size of grains more than 300 nm. Measured RMS is less than 1.2 nm. Obtained from spectroscopic ellipsometry optical properties are compared with those reported by Johnson and Christy and even better in desired wavelength 628 nm. Most of film grain have size higher than typical used SPASER-topology hole size, that allows to eliminate borders etching effects for most of structure elements.

Developed technology including nanolithography and etching procedures provided fabrication of high-performance nanostructures, such as noble metal film photonic crystal on transparent substrate, for high quality planar nanolaser. At 628 nm it shows lasing linewidth of 1.7 nm and a directivity of 1.3°. It could be used for a high-resolution sensorics in different areas and could be a mighty platform for lab-on-chip realization.

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REFERENCES

[1] Stockman, M. I., “Spasers explained,” Nature Photonics 2, 327 (2008).

[2] Meng, X., Liu, J., Kildishev, A. V. and Shalaev, V. M., “Highly directional spaser array for the red wavelength region,” Laser Photonics Rev. 8, 896-903 (2014).

[3] Jain, P. K., Huang, X., El-Sayed, I. H. and El-Sayed, M. A., “Noble metals on the nanoscale: optical and photothermal properties and some applications in imaging, sensing, biology, and medicine,” Accounts of Chemical Research 41, 1578-1586 (2008).

[4] Wei, H., Abtahi, S. M. H. and Vikesland, P. J., “Plasmonic colorimetric and SERS sensors for environmental analysis,” Environmental Science: Nano 2, 120-135 (2015).

[5] Craig, A. P., Franca, A. S. and Irudayaraj, J., “Surface-enhanced Raman spectroscopy applied to food safety,” Annual review of food science and technology 4, 369-380 (2013).

[6] West, P. R., Ishii, S., Naik, G. V., Emani, N. K., Shalaev, V. M. and Boltasseva, A., “Searching for better plasmonic materials,” Laser Photonics Rev. 4, 795-808 (2010).

[7] Rodionov, I. A., Baburin, A. S., Zverev, A. V., Philippov, I. A., Gabidulin, A. R., Dobronosova, A. A., ... & Baryshev, A. V., “Mass production compatible fabrication techniques of single-crystalline silver metamaterials and plasmonics devices,” In Metamaterials, Metadevices, and Metasystems (Vol. 10343, p. 1034337). International Society for Optics and Photonics (2017).

[8] Nguyen, H. H., Park, J., Kang, S. and Kim, M., “Surface plasmon resonance: a versatile technique for biosensor applications,” Sensors 15, 10481-10510 (2015).

[9] Melentiev, P., Kalmykov, A., Gritchenko, A., Afanasiev, A., Balykin, V., Baburin, A. S., ... & Dorofeenko, A. V., “Plasmonic nanolaser for intracavity spectroscopy and sensorics,” Applied Physics Letters 111, 213104 (2017).

[10] Johnson, P. B. and Christy, R. W., “Optical constants of the noble metals,” Phys. Rev. B 6, 4370-4379 (1972).

[11] Drachev, V. P., Chettiar, U. K., Kildishev, A. V., Yuan, H. K., Cai, W. and Shalaev, V. M., “The Ag dielectric function in plasmonic metamaterials,” Optics express 16, 1186-1195 (2008).

[12] McPeak, K. M., Jayanti, S. V., Kress, S. J. P., Meyer, S., Iotti, S., Rossinelli, A. and Norris, D. J., “Plasmonic films can easily be better: rules and recipes,” ACS Photonics 2, 326-333 (2015).

[13] Baburin, A. S., Ivanov, A. I., Ryzhikov, I. A., Trofimov, I. V., Gabidullin, A. R., Moskalev, D. O., Panfilov, Y. V. and Rodionov, I. A., “Crystalline structure dependence on optical properties of silver thin film over time,” PIERS – Spring 17490831 (2017).

[14] Baburin, A. S., Gabidullin, A. R., Zverev, A. V., Rodionov, I. A., Ryzhikov, I. A., Panfilov, Y. V., “Silver Films Deposited by Electron-Beam Evaporation for Application in Nanoplasmonics,” Herald of the Bauman Moscow State Tech. Univ. Instrum. Eng. 6, 4–14 (2016).