Preparation and analysis of plasmonic Ag thin films deposited by Ionized Jet Deposition method and Magnetron Sputtering method

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Abstract

Plasmonics is an advanced technology in the field of optical sensors. A methodology for the preparation of silver thin films suitable for plasmonics by Ionized Jet Deposition is developed. The RF Magnetron sputtering method is used for comparison as a well mastered technique. The roughness, relative permittivity and growth rate of the layers are analyzed using atomic force microscopy, attenuated total reflection and ellipsometry methods. Samples prepared by Magnetron Sputtering show slightly better properties, but the preparation by Ionized Jet Deposition can be further optimized.

Keywords: Surface Plasmonic Resonation; Ionized Jet Deposition; Magnetron Sputtering; thin films.

Introduction

In the past two decades, plasmonic has progressed to a mature technology allowing the manipulation of light in various applications, including sensorics. Plasmonic oscillations are oscillations of free electron gas in metals, which strongly influence the optical properties (reflectancy and transmittancy) of thin metal layers. Plasmonic sensors offer a versatile state-of-the-art optical sensors which can be used for example as distributed gas sensors.

As plasmonic properties rely on free electron gas, the elements from I.B. group - Cu, Ag, Au - whose metal phases exhibit high free electron concentrations, play an important role in plasmonic structures. It is necessary to investigate the manufacturing of plasmonic Ag layers, which are often used as a base layer in plasmonic multilayer structures in sensorics due to Ag's superior plasmonic properties.

Ionized jet deposition method (IJD) is a novel method belonging to the group of physical vapor deposition (PVD) techniques. The method is based on generating short pulses of electrons with energies between $5 - 20$ kV, which ablate the target material in an evacuated chamber. The plasma plume of the target material then travels across the chamber towards the substrate and forms the thin film. Due to its simplicity and relatively low operating expenses, the IJD method is a good candidate for upscaling and industrial use of thin films deposition.

In order to have good surface plasmon resonation properties the films need to have very smooth and consistent surface and have consistent low thickness (in the order of tens nm). We investigated the manufacturing of plasmonic Ag thin films using the IJD method and optimised deposition parameters to create films with desired thickness and surface properties. We compared our samples with plasmonic Ag layers manufactured using the well established Magnetron sputtering (MS) method.

Materials and Methods

Substrate, deposition material. Common laboratory glass and fused sillica were used as substrates. Pure Ag targets were used for the deposition of thin Ag films. TODO: původ materiálů

Ionized Jet Deposition. The IJD deposition system JetDep 100 (manufactured by Noivion s. r. o. and Czech Vacuum s. r. o., Czech Republic) was used in the preparation of thin Ag films. The IJD deposition parameters common for all prepared samples were as follows: The pulse frequency was set to 100 Hz, the distance between substrate and target was set to 110 mm, the distance between the IJD head and target was set to 3 mm. The mean primary electron beam spot diameter amounted to 1 mm on the target. The substrate temperature was set to 300 K. A gas mixture of Ar+6 vol. % H2 was used as the IJD working gas (purity 99.999%, provided by Linde Technoplyn, s.r.o., Czech Republic). The initial pressure before deposition and the working pressure were set in the range of $(3,0-4,1)\times10^{-3}$ Pa and $(5,2-7,2)\times10^{-2}$ Pa, respectively. The deposition voltage was varied between $9-12$ kV and deposition time between $20-60$ min. Microscope slides were used as substrates.

Magnetron Sputtering. The control samples of thin Ag films were prepared using magnetron sputtering system installed in the Institute of Physics, Czech Academy of Sciences. The power of RF source was set to 100 W, the sputtering target was water-cooled and substrate was at a room temperature. The working pressure was set to 1 Pa, Ar was used as the working gas. In two cases, a predeposition of AgO_x were performed by using a mixture of Ar and O in the ratio of 2 : 1. The gases were of purity 99.999%, provided by Linde Technoplyn, s.r.o., Czech Republic. Microscope slides and fused silica were used as substrates.

Sample analysis. The root mean square (RMS) roughness of both sample series were analyzed using Atomic Force Microscopy (LiuteScope equipped with the Akyiama probe, manufactured by NenoVision s. r. o.). The thickness, relative permitivity and EMA Roughness (calculated according to Bruggeman model of effective media approximation[1]) were analyzed using Attenuated Total Reflection method in the Kretschmann configuration (laser $\lambda = 632,8$ nm, BK7 prism and photodiode detector) and Ellipsometry method (M-2000X manufactured by J. A. Woollam Co., Inc).

The AFM data were analyzed using the Gwyddion[1] software in version 2.61 in order to evaluate the RMS roughness of the samples. The ATR data were processed using the tmm Python package[2] in version 0.1.8 in order to evaluate the surface plasmon properties of the prepared films and calculate thickness, EMA rougness and relative permitivity. The ellipsometry data analysis was performed using the CompleteEASE software in version 6.56 in order to evaluate the surface plasmon properties of the prepared films and calculate thickness, EMA rougness and relative permitivity.

[1] `http://gwyddion.net/`
[2] `https://pypi.org/project/tmm/`

Results and Discussion

Roughness

Using magnetron sputtering method, a lower roughness was achieved, which is essential factor for excitation of surface plasmon-polariton. The lower roughness was achieved mainly by switching to fused silica substrates and short (2 s) predeposition of AgO_x, because it nucleates on the substrate by layers and Ag nucleates on it by layers, while Ag on the substrate nucleates by island [2], so probably a similar improvement is possible in the IJD method.

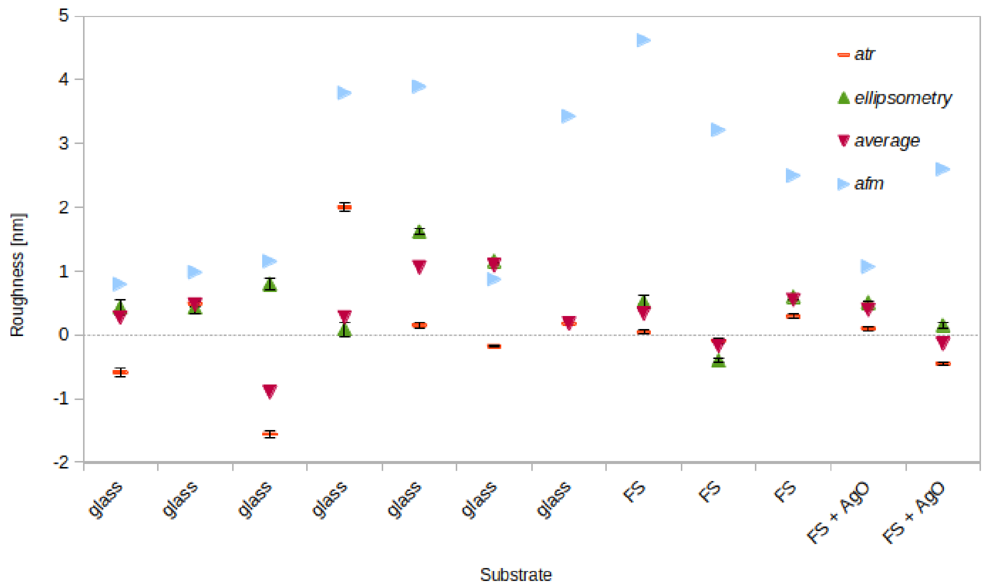

Figure 1: Magnetron sputtering series: Rouhness on different substrates

Relative permitivity

At $\lambda = 632,8$ nm, the relative permitivity achieved was close to relative permitivity of bulk Ag $(-18,281 + 0,48108\mathrm{i})$[3], which indicates purity and low porosity of thin Ag films. The average relative permitivity of the IJD series was $(-13,44 \pm 0,02) + (0,960 \pm 0,004)\mathrm{i}$ on microscope slides. The Magnetron Sputtering series achieved $(-17,17 \pm 0,03) + (0,716 \pm 0,007)\mathrm{i}$ on microscope slides, $(-16,74 \pm 0,03) + (0,840 \pm 0,009)\mathrm{i}$ on fused silica and $(-15,50 \pm 0,02) + (0,886 \pm 0,009)\mathrm{i}$ on fused silica with AgO_x predeposition. According to literature [2], the AgO_x should reduce to pure Ag during the deposition, but the relative permitivity value achieved suggests that it might not happen fully. The relative permitivity values of Magnetron Sputtering series compared to IJD series suggests that the Magnetron Sputtering method achieves better quality films (e. g. due to lower porosity).

[3] `https://refractiveindex.info/`

Growth rate

The average film growth rate using the IJD method was $(16,33 \pm 0,02)$ pm/s, increasing with the deposition voltage as expected. The average film growth rate using the Magnetron Sputtering method was $(1,1673 \pm 0,0004)$ nm/s, which is cca $60\times$ faster than the IJD method, but the deposition process is so fast it starts to be a problem to precisely control the thickness of the samples by manipulating the deposition time, while the IJD method being slower on the other hand offers better thickness control. The IJD deposition rate can be increased by increasing the deposition voltage, but it would probably also increase the roughness, thus decrease the quality of the samples.

Conclusions

At this time, the Magnetron Sputtering method offers better Ag thin films plasmonic properties, but it is better established method than the IJD method and the Ag deposition process is well optimized, so it is possible that by further development and optiomization of the IJD deposition process we could achieve similar results. The further research should focuse on deposition using the IJD method and fused silica as a substrate, alternatively also with the AgO_x predeposition, in order to decrease roughness and increasing the deposition voltage in order to increase growth rate.

References

[1] D. A. G. Bruggeman. Berechnung verschiedener physikalischer Konstanten von heterogenen Substanzen. I. Dielektrizitätskonstanten und Leitfähigkeiten der Mischkörper aus isotropen Substanzen. *Annalen der Physik*. 1935; **416**(7):636-64. Available from: `https://onlinelibrary.wiley.com/doi/abs/10.1002/andp.19354160705`.

[2] J. Bulíř, M. Novotný, A. Lynnykova and J. Lančok. Preparation of nanostructured ultrathin silver layer. *Journal of Nanophotonics*. 2011; **5**(1):051511. Available from: `https://doi.org/10.1117/1.3562568`.