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High-Quality Ultrathin Gold Layers For Use In Plasmonic And Metamaterials Applications

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ABSTRACT

The propagation of electromagnetic waves can be manipulated at the nanoscale by surface plasmons supported by ultra thin metal layers. An adhesion layer, with thickness in the order of few nanometerss is used for depositing ultra thin metal gold layers. Cr and Ti are the most popular metallic adhesion layers. Apart from them, a non metallic silane based wetting layer like (3-Aminopropyl)trimethoxysilane (APTMS) can be used. The behaviour of the propagating surface plasmons due to the influence of these adhesion layers has not been thoroughly investigated. To study the influence of the adhesion layers on propagating plasmons for use in plasmonic and metamaterial applications,we experimentally compared the performances of the ultra-thin gold layers using Cr and APTMS adhesion layers and without any adhesion layer. We show that the gold layers using APTMS adhesion exhibit short range surface plasmon polaritons (SR-SPPs) with characteristics close to the theoretical calculations, considering an ideal gold film.

Keywords: APTMS adhesion layer, ultra thin gold film, low roughness, plasmonics

1. INTRODUCTION

Plasmonics is playing a key role in the field of nanophotonics with a wide range of applications in nanofocussing,[1] optical biosensing,[2] enhanced emission of light.[3] This is possible due to collective oscillations of electron plasma in the metal that have the ability to focus electromagnetic waves at nanoscale in metal-dielectric structures.

In the visible to near infra-red wavelengths regime, silver (Ag) and gold (Au) are known to be the best plasmonic materials due to their high plasma frequency. Au is mainly used in the field of plasmonics applications due to its chemical stability. The most commonly used adhesion layers for Au are Ti and Cr which are metallic but non metallic options like organosilane compounds[4] can be used.The reported roughness of the Au layers using these different adhesion layers ranges from 0.30 nm to 1 nm.[5] However, even after successful fabrication of ultrathin films, introduction of these typically 1-2 nm thick metallic adhesion layers may affect the optical performance of the structures by introducing extra absorption or scattering of the localized surface plasmon polaritons.

One of the applications of ultra-thin Au film is the realization of anisotropic materials like hyperbolic metamaterials (HMMs).[6] HMMs are used for various applications in nano imaging, biosensing and spontaneous emission engineering.[7] Fabrication of multilayer HMMs in the visible and near infra red requires ultra-thin and ultra-smooth films with thicknesses in the range of 15 nm.[2] Therefore, the quest for adhesion layer to obtain thinner and smoother Au layers with non damping optical performance is crucial.

In this work, we show that silane based wetting layer, APTMS can achieve high quality ultra-thin gold layers with low roughness 0.30 nm rms which supports the short range surface plasmon polaritons with large wavevectors when compared experimentally with a Cr adhesion layer and without any adhesion layer.

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2. METHODS

2.1 Au Nanolayer Fabrication

The samples are fabricated on Si wafers with thick SiO_2 (8 μm). They are initially cleaned in Piranha solution (70 % H_2SO_4 and 30% H_2O_2) for 20 minutes to remove any organic residues.The deposition of APTMS adhesion layers starts with the immersion into 2.5 % APTMS in IPA solution for 3 hours.[4,8] For the metallic ahdesion layer, 1 nm thick layer Cr are deposited using e-beam deposition at a very slow deposition rate of 2Å/s .Then, the Au layers with thicknesses of 8, 10 and 18 nm are sputtered onto the samples with a constant deposition rate of 10 Å/s. We deposited another 1 nm thick layer of APTMS and Cr respectively on top of the Au films following the same procedure to maintain the symmetry. 200 nm SiO_2 layer was deposited on top of the Au films using sputtering to maintain the symmetry of the mode and also to allow for excitation of plasmons using the Otto configuration.

The Au layer qualities are characterized using SEM and AFM. The various RMS roughness of the layers are listed in table 1. Fig 1 shows the SEM images for the Au layers with different adhesion layers with thickness of 8nm.

Table 1. RMS Roughness of Gold Layers with different thickness and adhesion layer

	Au-8nm	Au-10nm	Au-18nm
1nm APTMS	0.30	0.31	0.32
1nm Cr	0.40	0.41	0.36
No adhesion	1.24	0.63	0.51

The Au films with the APTMS adhesion layer are the smoothest, with a root mean square (RMS) surface roughness of 0.30 nm followed by the Au films with the Cr adhesion layer with a surface roughness RMS of 0.40 nm. From the SEM and AFM characterization, it is seen clearly that the Au without adhesion layer has the highest roughness of 1.2 nm rms and it can be correlated from the SEM image where the layer without adhesion layer has pin holes and cracks due to the percolation threshold limit of the gold layer.

2.2 Optical Characterization

The spectroscopic reflection measurements were performed using a ZnSe semi-cylinder as a high refractive index prism to excite SR-SPPs.The incidence angle was varied from 33 to 55 degrees with 2 degrees interval by rotating the mechanical stage. The light source was a NKT Photonics, super continuum broadband laser allowing for measurements between 600 and 1750 nm wavelength range. Each incidence angle was measured 10 times to eliminate the mechanical stage uncertainties: 5 times when increasing the incidence angle and 5 times when decreasing the angle. The reference of the reflection spectrum is the measurement of the reflected beam at 55 degrees incidence angle without sample to ensure total internal reflection (TIR).

2.3 Numerical Simulation

The simulations in Figure 2(a-c) are conducted by the scattering matrix formalism in order to calculate the reflectance from the multilayer structures.[9] The model structure consists of 5 layers: ZnSe prismSiO_2 (200 nm thick)Au nanofilm (8, 10, 18 nm)SiO_2 (8 μm)Si substrate. The reflectance simulation was conducted with TM-polarized incident light with the wavelength range from 600 nm-1750 nm and the angle of incidence from 33°-55°.

3. RESULTS AND DISCUSSION

Gold layers of thicknesses of 8, 10 and 18 nm with three different configurations-with an APTMS adhesion layer, Cr adhesion layer and without any adhesion layer were fabricated to compare their performances.

We performed a series of reflection spectroscopic measurements to excite and detect SR-SPPs by using the Otto-configuration with high index ZnSe prism.The measured dispersion diagrams for SR-SPPs are obtained for
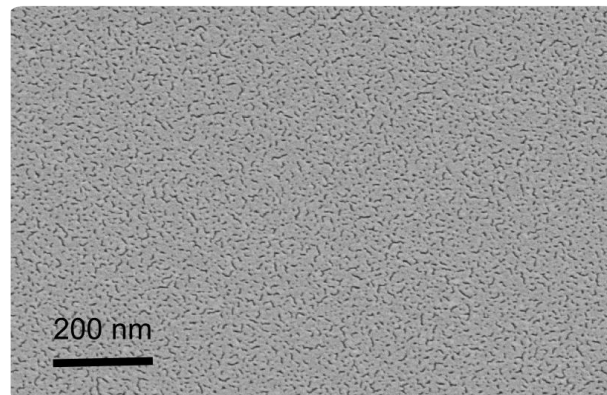
a

200 nm

b

200 nm

c
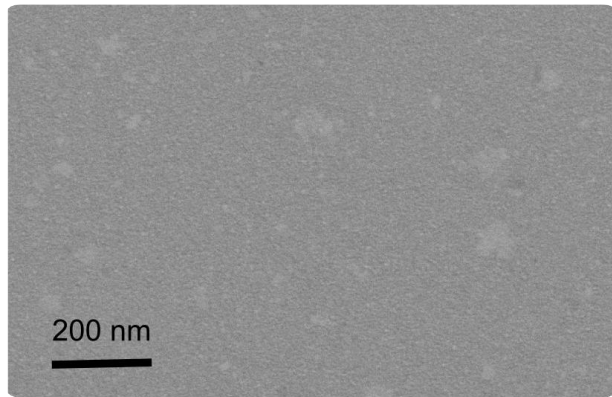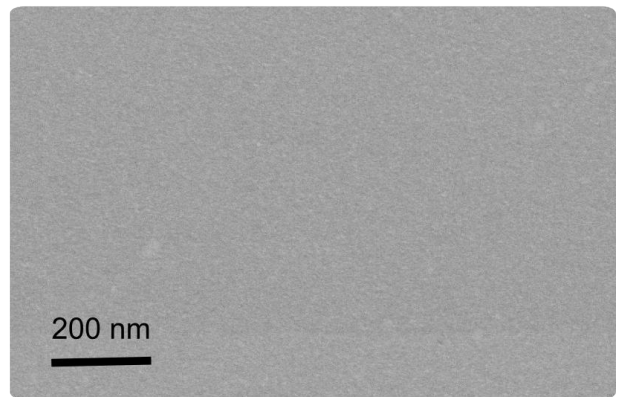
200 nm

Figure 1. SEM images of (a)1 nm APTMS adhesion layer (b) 1 nm Cr adhesion layer (c) and without adhesion layer for the thickness of Au 8nm
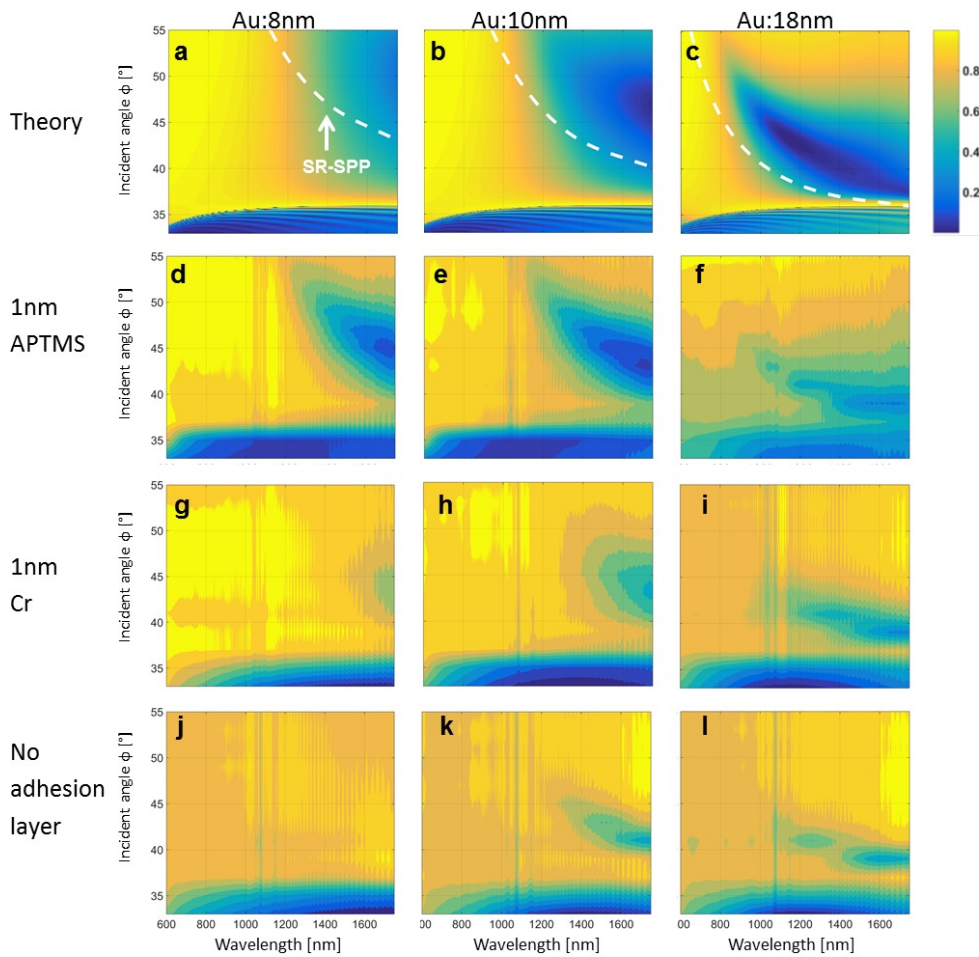
Figure 2. Dispersion diagram of SR-SPPs on ZnSe prism-SiO$_2$ (200 nm)-adhesion layer (1 nm)-Au-adhesion layer (1 nm)-SiO$_2$ (8 μm)-Si substrate structures. Simulations (a-c) and measurements with APTMS adhesion layers (d-f), 1 nm Cr adhesion layers (g-i) and without adhesion layer (j-l), for the thickness of Au 8, 10 and 18 nm, respectively. The white dashed lines in (a)-(c) are theoretical dispersion of SR-SPPs on infinitely thick SiO$_2$-Au-SiO$_2$ layers. The critical angle between the ZnSe prism and the SiO$_2$ layer divides the high and low reflectance regions.

three different sets of samples: 1nm thick APTMS as adhesion layer (figure 2d-f), 1nm thick Cr adhesion layer (figure 2g-i),and with no adhesion layer (figure 2j-l) compared to theoretical simulations using scattering matrix method considering an ideal uniform Au film embedded in SiO_2 (figure 2a-c). The low reflection region beneath 35 degrees is due to the non fulfillment of the total internal reflection conditions below the critical angle between the ZnSe prism and the SiO_2 layer. The oscillations visible in the experimental measurement spectra above 1100 nm are due to Fabry-Perrot interference in the experimental setup and are not related to the sample's response and the noise close to 1064 nm visible in the experimental dispersion diagram is due to normalization errors arising from the spectral characteristics of the light source.

For the 8 nm thick Au film, it can be seen from figure 2d that the experimental dispersion curve for APTMS adhesion layer is much more vivid and reflection dips corresponding to SR-SPPs are clearly visible . However, for the Cr adhesion layer, the reflectance dip associated with SR-SPPs cannot be observed below 1600 nm wavelength at all (figure 2g).SR-SPPs suffer the extra damping introduced by the metallic adhesion layer. As a result, SR-SPPs for larger angles of incidence are not supported. For the no adhesion layer, there is no any reflection dip visible for the case of the 8 nm film (figure 2j). This can be understood in terms of the percolation threshold of the Au film on the SiO_2 layer, further sustained by the high Au layer roughness of more than 1 nm RMS as shown in table 1. In the case of no adhesion layer, the plasmons propagate when the layers start to be continuous around 10nm which can be seen from figure 2k .

The role of the adhesion layers become more prominent when the when the thickness of the gold film is below 10 nm where the SR-SPP is highly localized. However, due to the addition of the lossy Cr metallic adhesion layer, the optical properties of the Au film are deteriorated, which is clearly visible by comparing figure 2d,e and figure 2g,h. In comparison with the simulation results, it can be seen that the dispersion curves of ultra-thin Au films with APTMS have better agreement compared to the rest. For example, in the case of 10 nm Au film with Cr, the plasmon dips are less pronounced. From this point of view, it is clear that the Au film with APTMS follows more closely the optical characteristics of the ideal Au film. Especially for the thickness of the gold film below 10 nm, the choice of the adhesion layer makes a distinct difference for the existence of SR-SPP modes for the shorter wavelength range and large wavevector region.For the thicker 18nm Au layer, the dispersion curves of all the cases looks similar to each other which is due to the fact that the volume ratio of adhesion layer and Au is low.

CONCLUSION

We conducted comprehensive experimental and theoretical studies on the influence of adhesion layers on propagating SR-SPPs plasmonic modes on Au films in three configurations: with Cr adhesion layer, with APTMS adhesion layer and without any adhesion layer. We found that using Cr as an adhesion layer greatly influences the optical properties of the SR-SPP due to its extra absorptions and scattering. For the Au films with the APTMS adhesion layers, the SR-SPP dispersion follows closely to the one theoretically calculated for completely flat pure Au layers, showing that APTMS is the better adhesion layer between oxide and Au if one aims to support highly localized propagating plasmon modes.

ACKNOWLEDGMENTS

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