# Ultra-thin gold films

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# Ultra-thin gold films: towards 2D metals for photonic and optoelectronic applications

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**Abstract.** Fabrication of continuous ultra-thin (<10 nm) and ultra-smooth gold films on different substrates is reported. Using a variety of electrical, optical and structural characterization techniques, we show that monolayer $MoS_2$ can be superior to more conventional adhesion/seeding layers as an optical substrate for realizing ultra-thin gold films. Our results show that optical losses in ultrathin gold films increase with decreasing thickness due to the fine-grained structure and the presence of a small number of voids, however, they exhibit metallic properties down to a thickness of 3-4 nm.

## 1. Introduction

Gold is a particularly important material for many applications in optics, electronics, photonics, plasmonics, and biosensing as it possesses a number of favorable electrical, optical, physical and chemical properties [1,2]. It is well-known that the growth of continuous and ultrathin gold films on different substrates, such as glass, silicon oxide, silicon nitride, graphene etc., is notoriously difficult due to the poor wetting of gold to these materials [3-5]. The growth kinetics of metal films is generally determined by the adsorption and diffusion behavior of metal adatoms on the substrate. A small ratio of the adsorption energy of metal adatoms on the substrate to the bulk cohesive energy of the metal and low diffusion barrier for an adatom favor the three-dimensional island growth behavior also known as the Volmer-Weber growth mode [3]. Within the framework of this growth model, the formation of a metal film is associated with the following stages: nucleation of islands, island growth, island impingement and coalescence, percolation, and channel-filling to finally form a continuous thin

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film. To reduce the percolation threshold of ultrathin gold films, adhesion or seed layers of Ti, Cr, Ni, Pt or Ge are commonly used. However, these adhesion layers significantly affect the optical and electrical properties of ultra-thin metal nanostructures [6, 7]. Here, we report on the fabrication of ultra-thin gold films on different types of substrates (including $SiO_2$, graphene and monolayer $MoS_2$). The gold films were fabricated using standard vacuum deposition methods at room temperature. We have undertaken numerous experiments with different deposition regimes (to study the change in the optical and electrical properties of ultra-thin gold films with respect to differences in structural features) and focused on observing how the different under-layers affect the growth kinetics and characteristics of continuous ultra-thin gold films.

## 2. Materials and methods

As substrates for thin metal films deposition, we used:

- Chemically cleaned $SiO_2$/Si substrate;
- CVD-grown monolayer graphene transferred by a poly(methyl methacrylate) (PMMA)-assisted transfer method onto silicon wafers with a 285 nm $SiO_2$ coating (from Graphene Laboratories Inc. (New York, NY, USA));
- Atmospheric pressure CVD (APCVD)-grown full area coverage MoS2 monolayers on silicon wafers with a 285 nm SiO2 coating (from 2D semiconductors Inc.).

An electron beam evaporator Nano Master NEE-4000 was used to simultaneously deposit Au films of different thicknesses on top of all three substrates. The deposition was performed at room temperature using gold pellets with a purity of 99.999% (Kurt J. Lesker). The base pressure in the vacuum chamber before the evaporation process was as low as $\approx 5\times10^{-7}$ Torr, and it increased to $\approx 2\times10^{-6}$ Torr during evaporation. The high deposition rate of about 5 Å/s and a resulted mass film thickness were controlled during deposition by a quartz-crystal sensor mounted in the vacuum chamber. The ultra-thin Au films thickness was also independently determined by step height AFM measurements and the root-mean-square (RMS) surface roughness value was determined for each film. Spectroscopic ellipsometry (SE) has been applied to study the optical properties of ultrathin Au films deposited on monolayer $MoS_2$ samples, graphene and pure SiO2/Si substrates [8, 9]. The dielectric function spectra were evaluated from data measured using a variable-angle spectroscopic ellipsometer (WVASE®, J. A. Woollam Co.) operating in the wavelength range of 300 – 3300 nm. Parameters of $\Psi$ and $\Delta$ were measured at angles of incidence of 60°, 65° and 70°. To investigate the nano-morphology of the films we used SEM (JEOL JSM-7001F). Finally, sheet resistances of all deposited Au films were measured by the four-point probe system (Jandel RM3000).

## 3. Results and discussion

Scanning electron micrographs in Figure 1 demonstrate the surface morphology of 4-nm-thick Au films on $MoS_2$, graphene and $SiO_2$ substrates (obtained at the same deposition process). The difference in film morphologies can be characterized by the metal filling fraction. As shown in Figure 1, Au films on $SiO_2$ primarily consist of isolated metal nano-islands (merging into a percolating film with multiple voids at the film thickness of 9 nm). While, in comparison, Au films on graphene and $MoS_2$ exhibit structure of closely linked clusters and almost continuous structure with tiny voids at 4 nm, correspondingly. Such differences in structure are a result of different kinetic processes of Au film growth on different substrates [10]. As well as SEM results, a significant difference in structural morphology of Au films grown on different substrates is also observed in the analysis of AFM scans.

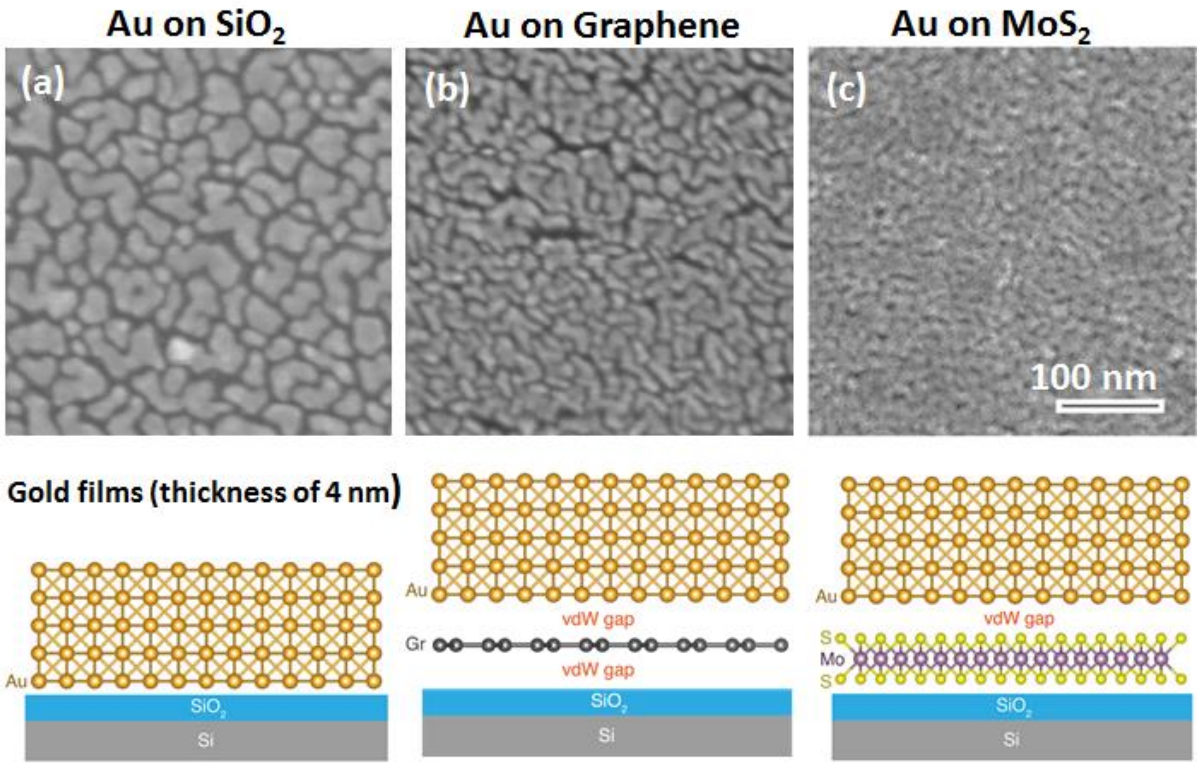


**Figure 1.** SEM images (scale bar = 100 nm) of the surface morphologies of gold film on monolayer $MoS_2$, graphene and SiO2/Si substrates along with schematic illustrations of the fabricated films structure. The films thickness is found to be close to ~4 nm. All images are 400 nm across.

AFM imaging demonstrates that gold films of 4 nm on $MoS_2$ are more densely packed and smoother than those formed on $SiO_2$ and graphene. Thus, RMS surface roughness value of 0.22 nm is determined for Au on $MoS_2$ (with a three-fold decrease in the average roughness compared to $SiO_2$-based films). Next, to determine the effective permittivities of the gold films, we performed ellipsometric measurements and fitted the acquired data with a model comprising film thicknesses measured by AFM. The most interesting findings are obtained for ultra-thin gold films formed on $MoS_2$. Our results demonstrate that an increase in the gold film thickness from 2 to 9.0 nm is accompanied by a significant change in both real ε' and imaginary ε'' dielectric parts. Transition of ε' from a positive value (insulator) to negative (metal) determines the film percolation threshold [10], that is confirmed by the electrical measurements (showing the nonconducting behavior of 2 nm-thick film and conductive from 3 nm) and analysis of the morphology carried out by SEM. As a final independent check of the film quality, the scattering scanning near-field optical microscopy (s-SNOM) was applied for probing local electromagnetic response of the gold films under investigation. It is found, that our SNOM observations are also consistent qualitatively with the wavelength-dependent features in the far-field experiments discussed previously.

## 4. Conclusions

Ultra-thin gold films deposition on different substrates using standard vacuum deposition methods at room temperature is studied. By extensive optical, electrical and structural characterization, we have demonstrated that ultrathin gold films on MoS2 exhibit continuous smooth morphology and Drude plasmonic response at thicknesses down to 3-4 nm. The results obtained can advance the use of high-quality ultrathin Au films for fabrication of the optoelectronic and nanophotonic devices.

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