



THESIS DEFENSE: Highly Percolated Ultra-Thin Metal Films for Reconfigurable Metasurfaces

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There is currently significant scientific and technological interest in ultrathin materials, with at least one dimension in the nm range or even smaller, since they possess features not achievable with their thicker counterparts. The large electrical tunability and extreme field confinement in graphene and other two-dimensional (2D) materials provide a clear example of unique electrical and optical properties that can be used to demonstrate unprecedented devices. Besides 2D materials, ultra-thin metal films (UTMFs) are emerging as potential game-changers in the field of optoelectronics. However, the standard deposition of metals via scalable fabrication methods, such as sputtering or evaporation, results in the formation of disconnected metal islands at small thicknesses. Therefore, substrate surface energy modification techniques are generally required to promote early percolation of the metal

films. In addition, the tuning of the physical properties of UTMFs after fabrication has been elusive for a long time due to the large charge carrier density of metals together with the difficulties in obtaining continuous films at sufficiently small thicknesses. Recently, nm-thick silver (Ag) and gold (Au) UTMFs have been obtained by using seed layers. Thanks to the small film thickness achieved through this technique, large electro-optical tunability of plasmons supported by UTMFs through electrolyte gating was demonstrated for the first time.

Following these advances, throughout this thesis we introduce, for the first time to our knowledge, a new deposition technique consisting of seeding noble metal films with a sub-nanometric layer of cupric oxide. Such a technique promotes the early percolation of Ag and Au on silicon dioxide and calcium fluoride while leaving unaltered the optical properties of the substrate. The reported technique is scalable and is potentially expandable to the growth of other noble metals, therefore showing great potential for use in actual technological applications. The morphological, electrical and optical properties of the resulting Ag and Au UTMFs are also studied as a function of the thickness, demonstrating their high quality when conductive films with high transparency and/or ultra-smooth surfaces are required.

Regarding UTMF tunability, firstly we show a new way to tune Au UTMFs by photoreduction. This technique is used to demonstrate that transparent conductive and IR plasmonic surfaces can be continuously tuned and, if oxidation is added, can be cycled back and forth. The functional tuning enables reconfigurable UTMF structures to be achieved, and also has the potential to trimming their response to specific working points. This method is thus important for the large-scale deployment of such surfaces as one can compensate for material non-uniformity, as well as morphological and structural dimension errors occurring during fabrication. Secondly, we demonstrate reversible changes to the optical properties of infra-red (IR) semi-reflective and plasmonic surfaces by combining Au UTMFs with electrolyte gating, particularly focusing on measurements in a reflection geometry. The results demonstrate the potential of the method for electro-optical tunable devices in the IR regime, and extend its applicability to situations in which working in a reflection configuration is required or beneficial. Examples include smart windows or chemical- or bio-sensing applications such as surface-enhanced infra-red absorption (SEIRA).

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