Broadband Electrical Permittivity of Gold for Plasmonics and Nano-Optics Applications

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Abstract: We measure the electrical permittivity of different bulk and film gold samples by spectroscopic ellipsometry from 200 nm to 20 µm, resolving inconsistencies on plasmon resonances, lifetime, and SPP propagation associated with imprecise current literature values.

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1. Introduction

Resonances of plasmonic particles or optical antennas, nonlinear light-matter interactions, photoluminescence lifetimes, nanoparticle coupling efficiency, surface plasmon excitation or propagation, and metamaterial behavior all rely critically on the exact values of the complex electrical permittivity $\tilde{\varepsilon}(\omega)$ of the materials involved. In noble metal plasmonics, for example, especially the extrinsic particle plasmon properties like the size dependence of resonance or plasmon lifetime depend sensitively on the difference in permittivity between the particle and the surrounding medium. Despite the importance of accurate knowledge of $\tilde{\varepsilon}(\omega)$, only a few precise measurements are available. Yet, with thin film optical devices varying in crystallinity, film thickness, substrate, surface contamination, and surface roughness, long standing questions remain regarding how $\tilde{\varepsilon}(\omega)$ varies with sample preparation method and the resulting film morphology.

Previous data measured on poorly characterized samples or using different techniques vary drastically or contain significant errors or scatter, contributing to inconsistencies between data sets. For example, the expected surface plasmon propagation length [1] on a gold/vacuum interface at 10 µm wavelength varies between ca. 10 and 40 mm, depending on the choice of data used for the calculation ([2] and [3], respectively).

Here, we provide accurate electrical permittivity data for bulk gold in the form of single-crystal, evaporated thin film, and template-stripped thin film. The permittivity is measured by spectroscopic ellipsometry over a broad spectral range from 200 nm to 20 µm. The data resolves many inconsistencies that arise from the use of the previous measurements, and are crucially relevant for many plasmonics and metamaterial applications. Important variations are observed with surface roughness and crystallinity depending on sample preparation.

2. Spectroscopic ellipsometry measurements

Three bulk and thick film gold surfaces were measured. These included a 1 mm thick Au (111) single crystal (SC) (MaTeck GmbH), a 200 nm thick thermally evaporated film on a glass substrate, and a 200 nm thick template-stripped Au film on a glass slide [4]. The thickness of 200 nm was chosen in excess of the skin depth of ~20 nm where a thin gold film exhibits bulk behavior [5].

The complex refractive index, $\tilde{N} = n + ik$, was measured as a function of optical wavelength using a variable angle spectroscopic ellipsometer (VASE) or IR-VASE (both from J. A. Woollam, Inc.) for the 200 nm - 2 µm and 2 µm – 20 µm regions, using steps of 10 nm and 32 cm^{-1}, respectively. Complex permittivity values were calculated from the refractive index with $\tilde{\epsilon} = \epsilon_1 + i\epsilon_2 = \tilde{N}^2$.

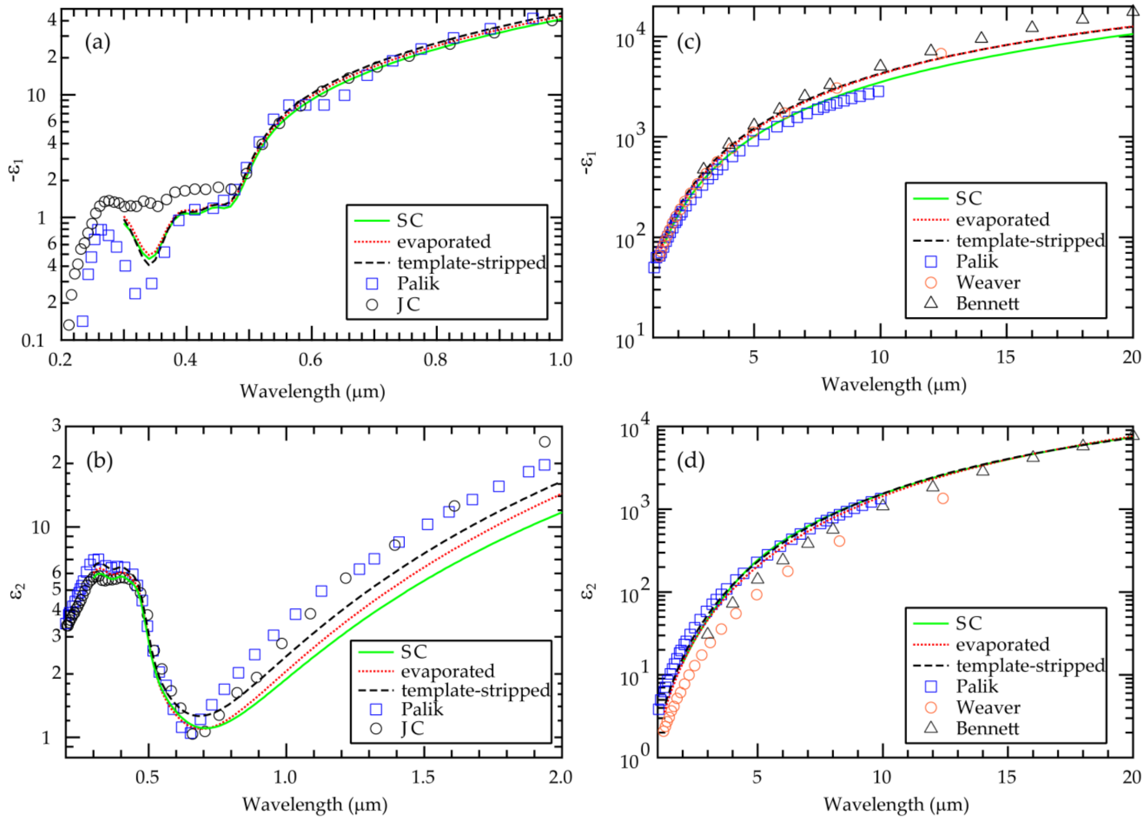

Fig. 1. Real part of permittivity (log scale) in the visible (a) and IR (b) and imaginary part in the visible (c) and IR (d) of a 200 nm thick Au film evaporated on glass, template-stripped 200 nm thick Au film, and single crystal (SC) Au, in comparison with previous experimental data from Johnson and Christy [5], Palik [2],Weaver, and Bennett (given by Ordal [3]). Variations between the samples indicate a strong dependence on crystallinity, particularly at longer wavelengths.

3. Results and Discussion

Figure 1 shows permittivity data (log scale) measured for the SC, evaporated, and template-stripped samples in the wavelength range of 200 nm – 20 μm. Data from Johnson and Christy [5], Palik [2], Weaver, and Bennett (given in [3]) are shown for comparison. All three samples agree well with respect to the real permittivity in the visible (a), and they are in good agreement with JC above about 500 nm. At shorter wavelengths, in the region of interband sp-d band transitions, JC deviates significantly. Palik, meanwhile, exhibits an anomaly centered at about 650 nm. Conversely, the imaginary part (b) shows good agreement at short wavelengths, but the permittivities begin to vary at about 600 nm, with JC and Palik systematically too high toward longer wavelengths. In the IR, the measured real permittivity values (c) are within the large range given by previous measurements. The evaporated and smooth template-stripped samples show nearly identical behavior, while the SC has a lower negative permittivity, indicating a dependence on crystallinity, but not surface roughness. For the imaginary part (d), while the three samples show good agreement with each other, particularly at long wavelengths, indicating that loss in the IR has a low dependence on sample preparation, their trend is steeper than Palik's, crossing to higher permittivity at about 5 μm.

The variability with degree of crystallinity observed, indicates that depending on the desired accuracy of model calculations for plasmonic device performance, metamaterials, plasmon-enhanced Raman or fluorescence spectroscopy, measurement of the dielectric permittivity might be necessary for the sample material used.

4. References

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