Thin gold layers – experimental optical properties and their correlation with classical Drude, non-local and ab-initio calculations

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Abstract: We present our latest findings regarding the optical properties of thin Au films and their relation with currently available models, from classical Drude, with thickness correction, to non-local estimations and ab-initio calculations predictions.

Thin metallic layers, in particular Au, are the main building block in plasmonic and metamaterials community [1]. However, there have been very few measurements of their optical properties, especially regarding the dependence on the film thickness [2].

Using a non-metallic adhesion layer, we showed that we can obtain ultra-smooth and thin Au layers [3] supporting surface plasmon polariton propagation characteristics very close to the theoretical ones [4]. Here, we present their experimental permittivity and compare it to several available models from literature.

The experimental data was obtained on layers between 8 and 22 nm thick, using ellipsometry measurements in the range of 675 to 1750nm to limit the influence of the interband transitions and use a simple Durde model.

In general, the dependence of the collision energy with thickness t is considered to be in the form of $\Gamma(t) = \Gamma_0 + A * v_f / t$ [5]. For nano-spheres, the free factor A is assumed to be unity [6]. In the case of our nano-layers the best fit is for a factor A of 0.06, significantly smaller (Fig 1(a)). As second finding, no clear trend in the behaviour of the plasma energy, as defined by the Drude model (Fig 1(b)) was observed.

The GNOR non-local model [7] shows a variation of the plasma energy within the error of the measure and a general trend of the collision energy that matches our data. The model from [8] predicts a change of the plasma energy that does not consistently match our experimental data. Ab-initio calculations on particles with diameters smaller than 3 nm show collision energies having a similar trend as the one observed experimentally [9].

We present experimental data and their comparison to different theoretical models for Au layer permittivity with no other metallic influence. Involving metallic adhesion layers would complicate the problem manifold [10]. To conclude, the size-dependent damping of Au was much smaller than expected, and there was no measurable plasmon energy change. Further analysis, especially for thicknesses below 10nm is required.

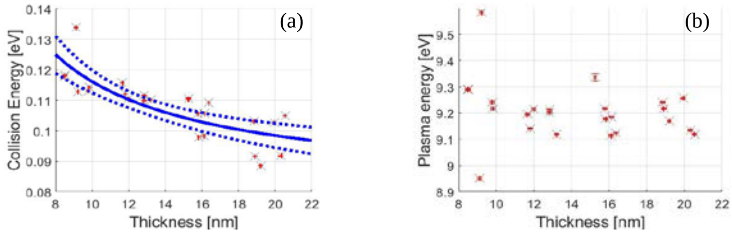

Fig. 1 The fitted values of the collision (a) and plasma (b) energy from the measured ellipsometer data. The full line is the best fit with the $1/t$ dependence. Dotted lines show the 95% confidence interval. The computed error bars (red lines) are too small to be visible. The fitting was made between 675 to 1750nm, to minimise the influence of the Lorentz terms.

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