



ELLIPSOMETRIC REGISTRATION OF COUPLING OF SURFACE AND LOCALIZED PLASMONS

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Abstract

Ellipsometry was used to register coupling of surface and localized plasmons in the system with gold nanoparticles deposited on a gold film. It is known that coupling of modes results in their hybridization and splitting of dispersion relations of separated modes, which crosses without coupling. Dispersion relations were obtained from ellipsometric measurements made as in standard configuration with external reflection as in Kretschmann configuration with internal reflection and direct excitation of surface plasmon. Those dispersions demonstrate expected splitting and allow to speak about strong coupling and Rabi splitting.

1 Experimental

Spherical gold nanoparticles of 50nm diameter from "Nanopartz" were deposited by absorption on a gold layer of about 40nm thickness. Gold layer was evaporated directly on BK7 glass slide and, for more uniform adsorption of nanoparticles, monolayer of dithiol (C10S2H22) was preliminary adsorbed on gold surface. Result of the adsorption is shown in Figure 1.



Figure 1 Nanoparticles on the gold surface.

In addition to the standard measurement configuration with external reflection ellipsometric measurements were made in Kretschmann configuration with internal reflection too. For such measurements additional 90^o prism from BK7 glass was optically contacted to the glass slide by spectrally matched immersion liquid from Cargille Co. Ellipsometry provides two so-called ellipsometric angles Ψ and Δ bearing amplitude and phase information [1]. As by definition $tg\Psi=|r_p|/|r_s|$ where r_i are Fresnel reflection coefficients, behaviour of the angle Ψ reflects extremums of reflection coefficients allowing to follow conditions of the energy transfer to modes of the system of interest.

2 Results and Discussion

Spectra of the angle Ψ for different angles of incidence measured for the system with nanoparticles independently on the measurement mode demonstrate two additional minima in comparison to the results for the clean gold film. Those minima correspond to the conditions of the excitation of additional modes created in the system after deposition of nanoparticles. Dispersion relations for those modes restored from the position of all recorded minima are shown in Figure 2.





A number of lines are presented in Figure 2. Two inclined dashed lines correspond to plane wave in air or BK7 glass. They separate the whole space into three areas where the area left to the light line in air is accessible by standard measurements with external reflection, the area between two light lines is accessible by internal reflection in Kretschmann geometry, any the last area is not accessible in our experiement. Horizontal lines are eye guides for the position of dispersionless localized modes of deposited nanoparticles. It is worth to note that deposition of spherical nanoparticles on a substrate lifts the degeneration of the localized plasmon of a spherical metallic nanoparticle and shifts it to lower energy. Black line corresponds to the recorded surface plasmon for clean gold film. It is seen that the visibility of this mode is restricted but it continues in both sides.

Three different branches created by the hybridisation of localized and surface plasmons are well seen and shown by colours. Lines correspond to the dispersion recorded by internal reflection as stars to the dispersion recorded by external reflection. It is interesting to notice that hybridisation increases the range of visibility of modes to low energies.

At least two other interesting results are visible in Figure 2. Dispersion recorded in both measurement modes well coincide and the mode recorded by external reflection is dispersive in contrary to the case of nanoparticles on a glass [2]. It happens because of the hybridization and reflects the existence of the surface plasmon felt in this way even at external reflection. The second worth result is the splitting about 0.2eV between modes coded by red and green what allows to speak about possible strong coupling of two plasmons and Rabi splitting between them.

The origin of the third mode at ~2.55eV is questionable. From one side it looks like the third mode also split with surface plasmon what is visible as deviation of the green mode from surface plasmon for clean gold. From other side this deviation may be explained by standard reaction of surface plasmon on material deposited on the surface. Also there should be no dipole mode of spherical gold nanoparticles at that energy. So, this question demands additional investigations.

3 Conclusions

Ellipsometry demonstrates good ability in the investigation of plasmonic systems and coupling of different plasmonic modes. Obtained results demonstrate splitting of coupled modes and allow to speak about strong coupling of localized and surface plasmons in the investigated system. Additionally results demonstrate that in such a system surface plasmon is exhibited even at external reflection at grazing incidence due to the noticed hybridisation.

4 References

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