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Surface plasmon polariton dispersion relation at organic/dielectric/metal interfaces

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ABSTRACT

The dispersion relation of a hybrid photonic–plasmonic system consisting of a light emitting thin organic para-Hexaphenylene (*p*-6P) layer separated by a dielectric gap from a plasmonic silver film is investigated using leakage radiation spectroscopy. Experimental studies are complemented by numerical simulations for the same structure but in the complementary, inverted configuration. The numerical simulations use as input ellipsometric measurements providing optical constants of *p*-6P in a wide spectral range. From a comparison between calculated and measured dispersion curves it is concluded that the molecular excitons in the *p*-6P film, created in the course of ultraviolet illumination of the sample, are de-excited via excitation of a plasmonic mode of the metal film.

1. Introduction

Hybrid structures, which combine both plasmonic and photonic components, have attracted much attention during the last decade due to their great potential for the development of ultra-compact photonic circuits [1–4]. One of the main routes in this direction is to find active surface plasmon based elements, which can convert optical signals to plasmonic signals and vice versa [4]. To obtain this, many different passive methods like slit or grating couplers [5,6] as well as active methods like electrical excitation [7], have been presented. Among others, a number of systems based on organic nanofibers has been proposed and successfully exploited, where organic nanofibers were used as local surface plasmon polaritons (SPPs) sources [8,9] or dielectric-loaded SPP waveguides [10–12]. These alternative materials, with inherent tailorable linear and nonlinear optical and optoelectronic properties [13–15], have proved a great potential for further development in the field of active nanoplasmonics.

In this article, we present experimental studies of a hybrid system consisting of a thin organic para-Hexaphenylene (*p*-6p) layer separated by a dielectric gap layer from a silver surface. SPPs in the silver film can be excited via several methods. Usually they are excited in the Kretschmann configuration by light undergoing total internal reflection at a prism surface covered with a metallic film. In this case, the tangential component of the wave vector of the TM (transverse magnetic) field on the dielectric prism is equal to that of the surface waves. Another popular method is to use a metallic grating instead of the prism. The hybrid system of the present approach allows for excitation of SPPs on a flat silver surface upon direct UV illumination of molecular emitters located in the proximity of the metal surface. Specifically, non-radiative relaxation of molecular excitons created in the *p*-6P film launches SPPs modes in the substrate.

To demonstrate that SPPs have been generated we use a *reversed* Kretschmann configuration, with a glass hemisphere. In this configuration a SPP excited on the metal–dielectric interface, leaks energy out through the metal film into glass at angles larger than the critical one for total internal reflection. This effect is often referred to as *leakage radiation* or *surface plasmon coupled emission* [16–18]. Performing these measurements for different wavelengths we create dispersion relations for SPPs. For comparing these measurements with theory we performed numerical modeling of the plasmonic modes which can exist in the multi-layer sample structure.

The results of the simulations are very sensitive to the *p*-6P complex index of refraction which is known only approximately [19]. Since our simulation approach requires very precise values of both n and k, we have measured those quantities in the wavelength of interest by spectroscopic ellipsometry.

2. Experimental

2.1. Sample preparation

The samples used in the experiment consisted of a 0.5 mm thick BK-7 glass plate (square 10 mm × 10 mm), coated on one side

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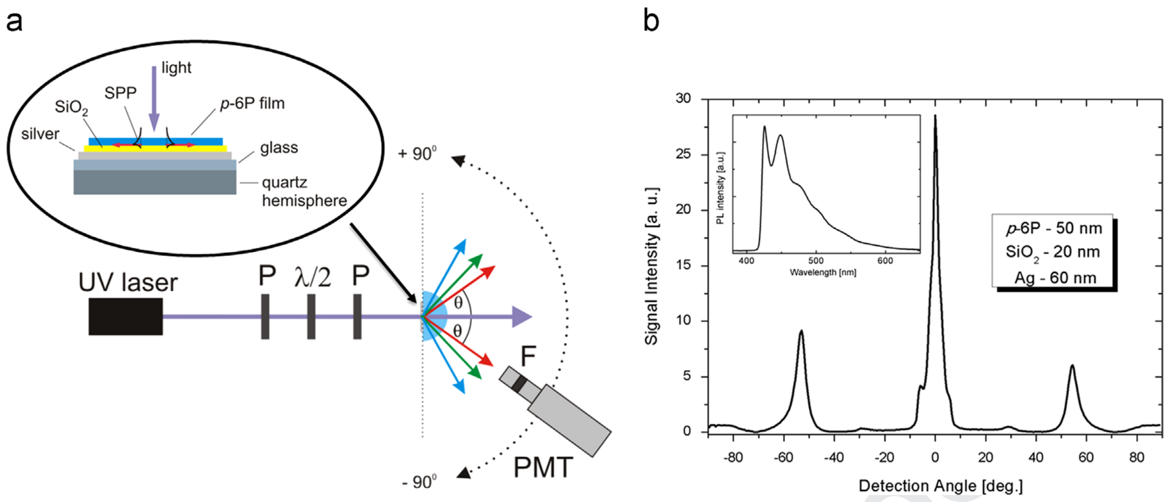

Fig. 1. (a) Leakage radiation spectroscopy setup: *P*, polarizer; *λ*/2, half-wave plate; *F*, filter; PMT, photomultiplier; inset: zoom on the half-sphere with the sample mounted on the flat side. (b) Typical experimental signal obtained at 470 nm during the angular scan; inset: photoluminescence (PL) spectrum of *p*-6P molecules upon UV excitation.

with a 60 nm silver layer, which was evaporated by electron beam metal deposition. For each sample, we added a layer of *p*-6P thin film, separated from the metal surface by a dielectric gap of 20 nm SiO$_2$. *p*-6p films were deposited by sublimation from a custom-built Knudsen cell at a temperature of 680 K and with evaporation rates of 1 Å/s. Evaporation rate and nominal thickness of the organic film were monitored by water cooled Inficon XTC/2 quartz crystal microbalance.

2.2. Leakage radiation spectroscopy

Each sample was placed on the flat surface of a 10 mm radius quartz hemisphere with the help of an immersion oil ($n = 1.5000 \pm 0.0001$). The excitation of the *p*-6P film was performed by light from a He–Cd 325 nm *p*-polarized laser. To fulfill the conditions of leakage spectroscopy the excitation light was directed under normal incidence to the surface and the scattered light was detected on the other side of the hemisphere by a photomultiplier tube (PMT) mounted on a motorized rotating arm. The angular resolution was 0.1 degree and we performed measurements in steps of 0.5 degree in the range from -90 to $+90$ degrees. The chosen detection spectral range 420–675 nm, overlapping with the *p*-6P photoluminescence band (inset of Fig. 1b), was ensured by a set of various narrow band-pass filters (FWHM 10 nm) mounted separately in front of the PMT (Fig. 1a). A typical scan showing the leakage radiation signal as a function of detection angle is shown in Fig. 1b. The side peaks correspond to leakage emission whereas the central peak results from transmitted direct laser light.

2.3. Spectroscopic ellipsometry

In order to achieve a high accuracy of the numerical simulations we performed spectroscopic ellipsometry to find precise values of both n and k of the *p*-6P thin film. We have exploited this method in the range 400–800 nm using a commercial spectroscopic ellipsometer (Angstrom Sun Technologies, SE200BM) with rotating polarizer Fig. 2.

In our experimental approach, in order to reduce the number of unknown parameters, which could increase the uncertainty of complex index of refraction measurement, we have simplified our system by depositing a *p*-6P film directly on a flat Silicon substrate. The thickness of the film was carefully checked by means of atomic force microscopy. To minimize measurement uncertainties caused by possible film non-uniformity we have used three different spots on the sample surface, at two different incident angles of 65 and 70 degrees, resulting in six different data sets. For each set we have used the Levenberg–Marquardt algorithm for evaluation of the data. Since the thickness of the film was precisely measured and optical indices for Silicon are readily accessible, the only unknown parameter of the whole system was the complex refractive index of the *p*-6P film. Its wavelength dependence was modeled by the Cauchy dispersion law with the following expressions for the coefficients *n* and *k*:

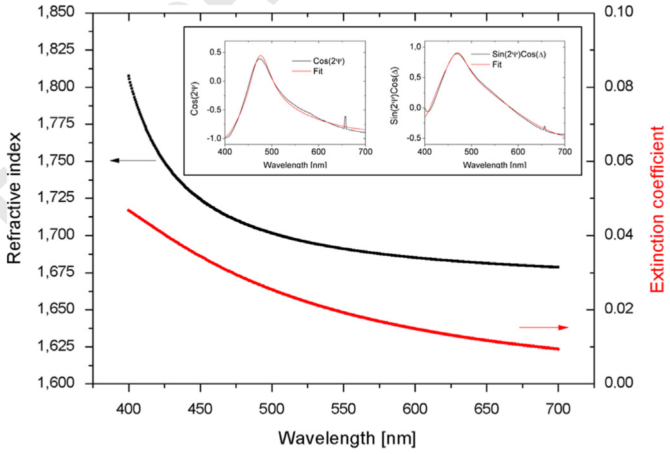

Fig. 2. Complex dispersion relation for a para-haxaphenylene thin film (thickness 200 nm) on Silicon. Inset: typical experimental data from ellipsometer obtained at 65 degrees, where *Ψ* and *Δ* are ellipsometric angles.

$$\begin{aligned} n(\lambda) &= A + B/\lambda^2 + C/\lambda^4 + D/\lambda^6, \\ k(\lambda) &= E + F/\lambda^2 + G/\lambda^4 + H/\lambda^6, \end{aligned} \tag{1}$$

where *A*–*D* are the Cauchy coefficients and *E*–*H* are the Cauchy extinction coefficients which are to be obtained. The measured complex dispersion relation for a *p*-6P thin film is plotted in Fig. 3 and the measured Cauchy coefficients are presented in Table 1.

3. Numerical simulations

The plasmonic modes in our system were investigated numerically by simulating the attenuated total reflection (ATR) of a plane electromagnetic wave in the Kretschmann configuration. Due to the boundary conditions the wave vector component parallel to

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