Research article

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The experimental evidence of a strong coupling regime in the hybrid Tamm plasmon-surface plasmon polariton mode

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Abstract: Total internal reflection ellipsometry was employed for the excitation and study of hybrid Tamm plasmon-surface plasmon polaritons mode. Simple optical methodology using optical filters to cut the part of incident light spectra was proposed. Using optical filters measured energy spectra was divided into two parts where in each range only one branch of the hybrid TPP-SPP plasmonic mode was excited directly by the incident light. Present experimental studies have shown, that if the investigated system is in strong coupling, this is always enough to excite only one component of the hybrid excitation. Thus, its dispersion relation will be the same as when the excitation is done with a whole spectrum. In the case of the TPP-SPP hybrid mode where strong coupling is realized only in p-polarized light, the fitting results have shown that the strongest coupling was at the point where the noninteracting TPP and SPP curves should be crossing. The obtained Rabi splitting for the hybrid TPP and SPP modes in BK7 prism/1D PC TiO$_2$/SiO$_2$ (60 nm/110 nm)/TiO$_2$ (30 nm)/Au (40 nm) multilayered structure was about 105 meV.

Keywords: hybrid Tamm plasmon-surface plasmon polaritons; strong coupling; total internal reflection ellipsometry.

1 Introduction

Recently much attention has been paid to the study of strong coupling between plasmon active metal nanostructures and various emitters such as excitons in semiconductors in dyes or photochromic molecules [1–4]. Nanostructures which are able to support energy exchanges in strong coupling regimes are promising systems for a new generation of nanolasers (spasers) [5, 6], ultra-sensitive optical biosensors [7], influencing chemical reaction rates [8, 9] and in quantum information processing [10]. The main feature of this strong coupling regime is that the energy exchange between the plasmons and the emitters occurs during a coherent time [11]. This means that in such coupled systems, the coupling strength between the plasmons and the excitons exceeds the damping rate and as a result, hybrid modes are created [12]. These hybrid modes can be achieved when the interactions are sufficiently strong, and the energy spectrum is modified, compared to spectral position of single resonances in noncoupled systems. These single excitations of the plasmons and excitons are then modified into new normal modes, the so called hybrid modes, which are polaritons formed partially from plasmons and excitons [13].

However, the existence of hybrid modes have been also shown between two different plasmonic excitations – surface plasmon polaritons (SPP) and Tamm plasmon polaritons (TPP) [14]. The well-known SPP is a surface electromagnetic wave propagated at the interface of a semitransparent metal layer (commonly gold or silver) and the dielectric. The SPP is a nonradiative electromagnetic mode, thus in order to excite the SPP waves, a glass prism or grating coupler is usually used in order to match the in-plane wave vectors of the incident light and the plasmons in the metal [15]. It should be noted that such SPPs can be excited only in TM polarization. Another type of electromagnetic surface waves which exist between the metal and photonic crystals are the so called Tamm plasmon polaritons (TPP) [16]. TPPs are optical states similar to the electron states proposed by Tamm [17]. These can be
In the case of a photonic crystal, the Bragg reflections form the photonic stop band for the photons, which resembles the energy band gap of the real crystals. In contrast to the propagated SPPs, the TPPs are nonpropagating states and can be excited in both TM and TE polarizations, like the Bloch surface waves on the interfaces between PC and dielectric [18]. The TPPs have an in-plane wave vector which is less than the wave vector of light in a vacuum, thus a direct optical excitation of these TPPs is possible [19], contrary to the SPPs, where the light wave vector is always smaller than the SPP.

When the conditions for these excitations are satisfied for both the resonances of the SPPs and TPPs, hybrid modes of both nonoverlapped excitations can be generated. Such situations can be realized whenever the glass prism is optically connected to a PC on which a thin metal layer is deposited. In such systems, the TPP and SPP resonances exist in a broad range of angles of light incident (AOI) to the prism. At certain AOIs to the prism corresponding to the same wavevectors at which both the SPPs and TPPs can be excited, the reflectance spectrum is changed, and the dispersion relations of both excitations are modified. These changes in the reflectance and dispersion relations indicate in most cases a strong coupling regime between the TPPs and SPPs. It should be noted that these alterations of the plasmonic resonances in the wavelength spectra also indicate their dynamics in the time domain at frequencies that correspond to their splitting into the hybrid mode [20]. This leads to an exchange of energy between SPPs and other oscillators (TPP, quantum dots, dye molecules and others) during the time of the coherence. Pump-probe spectroscopy was used to investigate the dynamics of such SPP oscillations strongly coupled with such other emitters [21]. However, the decay times for the plasmon modes are about \( \sim 100 \) fs, which indicate that the coherent energy exchanges are even shorter in time. Thus, the observation of these processes becomes rather challenging [22, 23], while the measurement of the reflection-transmission spectra in the wavelengths (frequency domain) remains much simpler. On the other hand, the analysis of the experimental reflectance spectra can be rather complicated and ambiguous as the strong coupling can be confused with other effects such as Fano-like interference (or exciton-induced transparency) [24].

In this study, we present unambiguous experimental evidence of such strong coupling between the TPP and SPP resonances in the hybrid TPP-SPP mode by tuning of the reflectance spectra with the optical filters, using the total internal reflection ellipsometry (TIRE) method [25, 26].

### 2 Methods

The sample used for the hybrid TPP-SPP mode excitation consisted of a 1D PC and a TiO\(_2\) layer about 30 nm thick with a thin gold layer (\( \sim 40 \) nm) deposited on its top. The PC was made of six alternating TiO\(_2\) (\( \sim 60 \) nm) and SiO\(_2\) (\( \sim 110 \) nm) bilayers, deposited onto a BK7 glass substrate by means of ion beam sputtering. The thickness of the metal had to be thin enough for the coupling of the TPP and the SPP to take place in a hybrid mode. The structure described above was measured using spectroscopic ellipsometry (SE). The ellipsometer used for these measurements was a J. A. Woollam RC-2 with two rotating compensators. The light source was a Xenon lamp with a spectral range of 210–1700 nm.

For the excitation of the hybrid TPP-SPP mode, a total internal reflection (TIR) configuration of the spectroscopic ellipsometry with a 45° prism coupler was used (Figure 1). Three different TIRE spectra were measured (Figure 2). The first was the full spectra (Figure 2, gray curve) where both TPP and SPP components manifested themselves at the 533 and 641 nm wavelengths, respectively, and were measured. Different optical filters were used to excite only one resonance (either the TPP or SPP). Due to this, some parts of source light were cut. An optical filter Schott VG 14 (F2) was used to cut a part of the white light source in order to leave only the TPP resonance (Figure 1, blue curve). Afterward, the SPP component (Figure 1, red curve) in the hybrid mode was left and the TPP was cut with filter Schott OG 590 (F1). All three types of the spectra were measured in light angles of incidence in the range of 40.5–49.5° and the experimental dispersion relations were determined.

### 3 Results and discussion

This strong coupling regime has been found and widely studied in the various exciton and surface plasmon supporting systems [3, 13, 21]. Nanostructures supporting Tamm plasmons have also begun to be widely used to couple these surface states with excitons [27–29]. It has been found [14] that both the TPP and SPP surface modes can be excited simultaneously on the same metal layer and interact with each other when suitable conditions are satisfied. In such cases, a new hybrid TPP-SPP mode appears and the anticrossing of the TPP and SPP resonances has been revealed [14, 30, 31].

When the energy exchange between the two harmonic oscillators exceeds the damping rate, the system is in a strong coupling regime. Under such conditions, the original resonant frequencies of the single oscillators are modified and a new hybrid mode appears in which the coupled oscillators are inextricably linked with each other [24]. In this study, the simultaneously excited TPP and SPP components corresponded to the two coupled oscillators. The definition of the strong coupling regime [20] implies that if the external excitation source has a frequency range suitable to excite only one component (the TPP or SPP),
still both parts of the hybrid mode should be generated, because one component excited by an external source will coherently transfer energy to the second component of the coupled oscillators. As a result, the behavior of the directly excited plasmonic components should follow the dispersion relation of the hybrid mode instead of the uncoupled ones.

To confirm this statement, the experimentally measured energy spectra (Figure 2) was divided into two parts where in each range only one branch of the hybrid TPP-SPP plasmonic mode was excited directly by the incident light. The variable angle TIRE measurements were conducted to evaluate and compare the dispersion relations of the separate components with the general spectra. The general spectra are presented as a reflectance map of the TM-polarization and the separate components marked as the dots on the map in Figure 3(a). Also the other modes have contribution to the whole optical response. The dispersion curves lower than 1.75 eV corresponds to the edge of the photonic stop band and interference of the Bragg mirror. As can be clearly seen, both components (TPP and SPP), excited separately, generated exactly the same energies as in the general spectra when both components were excited simultaneously. The modeled uncoupled TPP and SPP dispersion curves are presented as the white crossed curves in Figure 3(a), while the separately excited TPP and SPP branches (dots) follow the dispersion relation of coupled general spectra (map). Such behavior can be realized only in the case when the separate excitations of the hybrid plasmonic modes are influenced by the presence of the other components. This, however, implies that the system should be in the strong coupling regime, as otherwise, the separate plasmonic components should follow the dispersion relations of their uncoupled excitations.

The strong coupling between the TPP and SPP leads to the alteration of their initial frequencies. Thus due to the anticrossing effect, the two plasmonic branches form the gap in the frequency spectra called the vacuum Rabi splitting [20]. The value of the Rabi splitting allows one to...
determine whether the system is in a strong coupling regime or not. In the field of plasmonics and photonics, this strong coupling regime is usually defined as the range of splitting which exceeds the linewidths of the two coupled systems. It has been reported that in order to more precisely evaluate the coupling strengths between the two coupled oscillators, it is necessary to analyze a plot of the wave-vector vs. energy, instead of the angle of incidence vs. energy, which is directly obtained from the reflectance measurements [3]. The exact Rabi splitting for the hybrid TPP and SPP modes can be seen in Figure 3(b) and was about 105 meV. It should be noted that for the hybrid TPP-SPP plasmonic modes, the differences in the gap when the angle of incidence is monitored instead of the wave-vector can be overestimated by more than three times.

As has been shown before [12], the interference effect usually makes a noticeable contribution to the optical response and can distort the peaks of the coupled excitations. In order to evaluate the coupling strength and to distinguish the strong coupling from the weak and the other interference effects such as Fano resonances [12], a fitting procedure of the whole spectra needs to be conducted. The coupled plasmonic excitations were modeled as two Lorentz oscillators which influence each other with equal strength [32]:

\[
\begin{align*}
\ddot{x}_{TPP} + \gamma_{TPP} \dot{x}_{TPP} + \omega_{TPP}^2 x_{TPP} &= F + g x_{TPP} \\
\ddot{x}_{SPP} + \gamma_{SPP} \dot{x}_{SPP} + \omega_{SPP}^2 x_{SPP} &= F + g x_{SPP}
\end{align*}
\]

(1)

Where \( g \) is the coupling strength, \( \omega_j \) the resonance frequencies, and \( \gamma_j \) the damping terms that correspond to the linewidths of the TPPs and SPPs, respectively. \( F = F_0 e^{i \omega t} \) was the external driving force. We looked for solutions in the form of \( x_j = X_j e^{i \omega t} \). Thus, the resulting complex amplitudes for the oscillators were:

\[
\begin{align*}
X_{TPP} &= F_0 \frac{w_{SPP} + g}{w_{TPP} w_{SPP} - g^2} \\
X_{SPP} &= F_0 \frac{w_{TPP} + g}{w_{TPP} w_{SPP} - g^2}
\end{align*}
\]

(2)

Where

\[
W_j = -\omega_j^2 + j \gamma_j \omega + \omega_j^2
\]

(3)

Assuming there was no detuning and no damping, the resulting hybridized frequency for the oscillators is given by:

\[
w_\pm = \frac{\sqrt{\omega_{TPP}^2 + \omega_{SPP}^2 + \sqrt{4 g^2 + (\omega_{TPP}^2 - \omega_{SPP}^2)^2}}}{\sqrt{2}}
\]

(4)

We treated \( \omega_j = \omega_j(\theta) \) as a function of the angle of incidence. Furthermore, to determine whether the investigated multilayer structure was in a strong coupling regime or not, the coupling strengths between two coupled plasmonic oscillators were analyzed by regression analysis (fitting).

The value of the strength of the coupling parameter \( g \), however, does not give a direct answer as to whether the system is in strong coupling regime or not since the system is in the strong coupling regime only when parameter \( g \) is larger than [12]:

\[
g > \frac{1}{4} \left( \gamma_{TPP} + \gamma_{SPP} \right)
\]

(5)
Thus, for a more precise evaluation of the strong coupling parameters, only the TM-polarized reflectance spectrum was used for the regression analysis instead of the total reflection intensity spectra, where both the TE- and TM-polarizations had contributions to the optical response. This was done because the coupling between the Tamm plasmon polariton and the surface plasmon polariton can only be realized for TM-polarized light. In the intensity reflection spectra, the TE-polarization, which is not involved in the coupling with the surface plasmon, distorts and partially masks the two coupled plasmonic oscillators related to the hybrid TPP-SPP mode. This can be clearly seen from the spectra in Figure 4, where the sharp dip at the 1.88 eV in the curve with total reflectance is related to the Tamm plasmon for TE-polarization. The fitting results (Figure 5) of the TM-polarized reflection $R_p$ at different AOI show that the strength parameter $g$ had larger values than right side of the inequality (2), indicating the strong coupling regime (Table 1). The residual square or coefficient of determination (COD) of the fitting varied from 0.97622 to 0.99782.

It should be noted that applied rather simple coupled oscillator model do not take into account the multiple coupling with other modes such as edge of the photonic stop band of the Bragg mirror. This fact has influence to the evaluation of strong coupling parameter $g$ and fitting errors (Figure 5 (inset), Table 1) which increases at the angles of incidence ($43.98^\circ$, $44.32^\circ$, $45.68^\circ$, and $46.02^\circ$) further from the anticrossing point (Figure 3, white dashed lines). However, the fitting procedure was sensitive enough to evaluate whether the system is in a strong coupling regime or not between TPP and SPP branches.

### 4 Summary

Summarizing, we propose a simple optical methodology using optical filters which gives unambiguous experimental evidence of the strong coupling regime in the hybrid TPP-SPP mode. Present experimental studies have shown, that if the investigated system is in strong coupling, this is always enough to excite only one component of the hybrid excitation. Thus, only a part of the incoming light and its dispersion relation will be the same as when the excitation is done with a whole spectrum. In the case of the TPP-SPP hybrid mode where strong coupling is realized only in TM-polarized light, the fitting results have shown that the strongest coupling will be at the point where the noninteracting TPP and SPP curves should be crossing. As the plasmonic widely applied for realizing the strong coupling and many of these excitations are polarization dependent, the evaluation of the coupling strengths can be better conducted in a spectrum where only the state of the light polarization responsible for the strong coupling is involved. Polarization based optical methods such as spectroscopic ellipsometry thus can serve as advanced optical methods having the ability to analyze in detail the strong coupling effect. Application of strong coupling of hybrid TPP-SPP excitations could be valuable due to the reduction of energy losses in the metals and tunability in...
the desired spectral range. If one component is excited at resonant wavelength, the other is also present and can be controlled by strong coupling between TPP and SPP. The energy conversion between TPP and SPP shows potential applications of hybrid TPP-SPP modes for integrated photonic devices.

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### References


