SUPPLEMENTARY INFORMATION

Manuscript entitled "Excitonic Optical Tamm States: a step towards a full molecular-dielectric photonic integration"

Authors: S. Núñez-Sánchez, M. López-García, M. M. Murshidy, A. G. Abdel-Hady, M. Y. Serry, A. M. Adawi, J.G. Rarity, R. Oulton, W.L. Barnes

1.-High reflectance in heavily doped polymer films as a function of oscillator strength

Observation of a metallic lustre in a thin film is possible when the value of the real part of the permittivity of the material is negative enough. In a material described by a Lorentzian expression the restricted wavelength range where the real part of the permittivity of a material is negative to show a high reflectance response, depends strongly on the oscillator strength. The figure 1 shows the simulated reflection using an iterative recursive Fresnell method for two different thin films with different oscillator strengths. The figure 1.a corresponds to the optical properties used in the modelling part of the paper (f_0=1). Figure 1.b corresponds to the equivalent model for our experimental thin excitonic thin films (f_0=0.4).

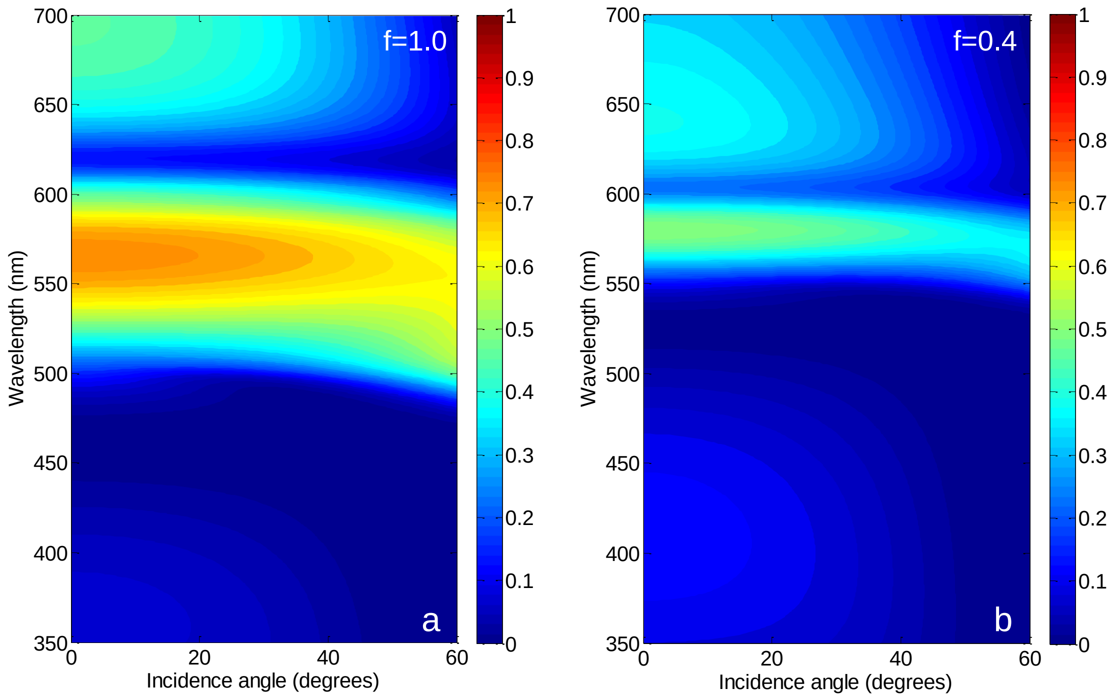

Figure 1. P-polarized reflectance for a 200 nm thin film with two different oscillator strengths: a) f=1 b) f=0.4.

2.- Determination and analysis of the dispersion curves of Exciton Optical Tamm States (EOTS)

2.1- Determination of dispersion curves by reflectance simulations

We have studied different methods in order to obtain the dispersion curves of the EOTS and to determine the cut-off conditions. The first method used is based on a qualitative observation on the comparison of the optical response of a bare DBR and the EOTS supporting structure (DBR covered with the J-aggregate layer). The Figure 2 shows the simulated reflectance at different angles obtained by recursive Fresnel formulation for a bare DBR and an EOTS-structure. The DBR is formed by 9 pairs of SiO2/SiN3 layers with a central wavelength of 520 nm. As it is well known, the optical response of a bare DBR shows a reflectance values close to the 100% in the stop band wavelength range with Fabry-Perot oscillations in the laterals. However, in case of EOTS, a dip in reflectance is observed at those wavelengths for which excitation of the OTS is possible. This condition can be only fulfilled at wavelengths lower than the low energy band edge of the bare DBR. Therefore, we may be able determine the mode cut-off wavelengths simply following the position of the dip in the reflectance inside the stop band of the bare DBR.

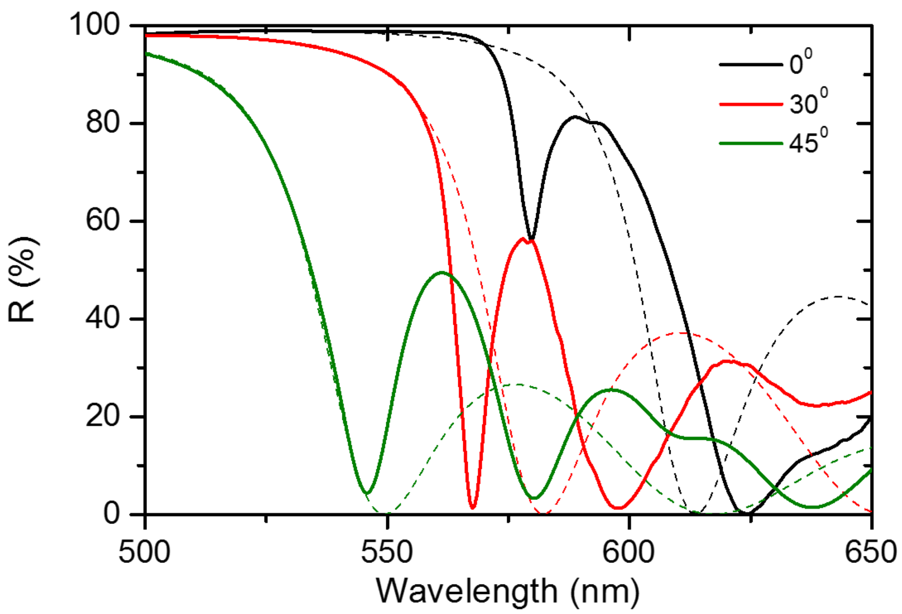

Figure 2. P-polarized reflectance of a DBR and an EOTS-structure using a DBR formed by 9 pairs of SiO2/SiN3 layers of with a central wavelength of 520 nm. The optical properties of the 200 nm excitonic layer correspond to the real properties or our excitonic material. Labels indicate the incidence angle.

We have established a numerical method to determine the dispersion curves by reflectance simulations. We have defined a new function that is the subtraction of the reflectance of the bare DBR from the reflectance of the EOTS-structure (Subtraction(λ,angle)=R_{DBR}-R_{EOTS}). The Figure 3 shows the subtraction function as a function of the wavelength and the reflectance for the bare DBR and the EOTS structure at 20 degrees. When the mode condition is fulfilled, the subtraction function will have a local maximum. In our method we determine the wavelength associated to the local maxima of the substraction function at different incident angles, only taking into account wavelengths within DBR stop band.

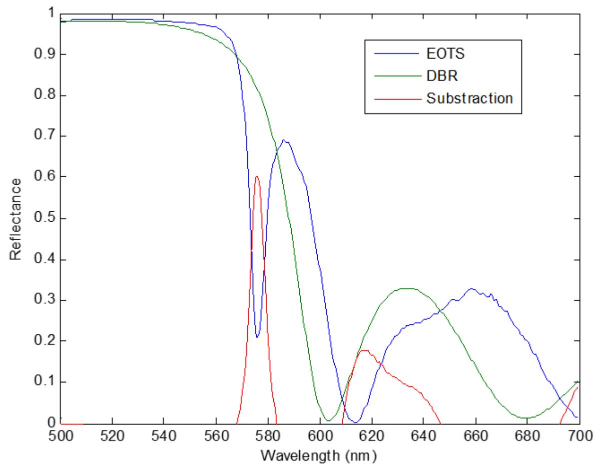

Figure 3. P-polarized reflectance at 20 degrees for a DBR with a central wavelength of 520 nm and an EOTS structure with a 200nm of our TDBC material on top the DBR. The value of the subtraction function has been also included.

The contour plots of the figure 4 show the reflectance values obtained for a DBR with a central wavelength of 520 nm, the associated EOTS structure and the substation function as a function of the wavelength and incident angle. We have superposed a red curve that corresponds to the dispersion curved obtained following the local maxima of the substation. In this case, the cut-off wavelengths at shorter wavelengths is mainly established by the band edge of the DBR.

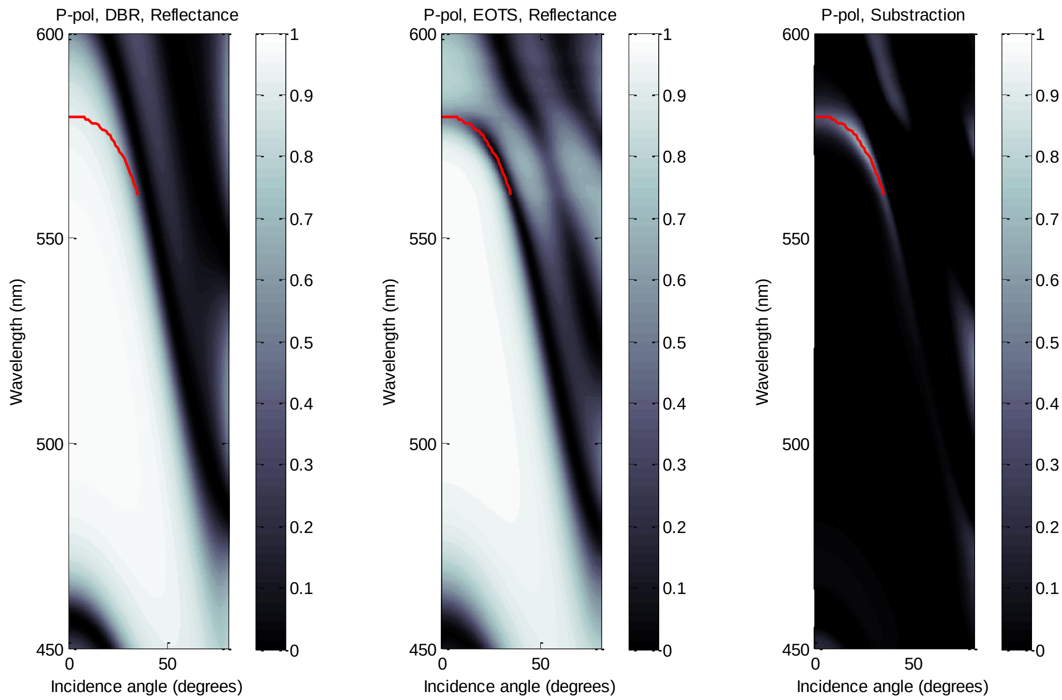

Figure 4. Contour plot of the P polarized reflectance of a) DBR formed by 9 layers of SiO2/SiN3, b) EOTS structure with the same DBR. c) Values of the substraction function for the same structures. The red line corresponds to the estimated dispersion curves.

2.2. Determination of dispersion curves by mode condition equation

From Kaliteevski et al [1] we know that the condition in the interface DBR-thin film for an eigenmode associated to a Tamm Optical State is: $r_{left}r_{right} = 1$. Where r_{left} and r_{right} are the Fresnell amplitude coefficients at the interface DBR-thin film. Therefore we could estimate the dispersion curves from the two Fresnel amplitude coefficients at different angles and analysing at which wavelengths the product of the two is close to one.

This equation has been established for an ideal situation, that is, two perfect mirrors with reflectance of 100%. However, in our case the DBR is formed by only 9 layers and the thin film shows a high reflectance, but never close to the total reflection condition (usually around 60% as maximum depending of the angle, see Figure 1). Therefore, we decided to estimate the dispersion curve as the position of the local maximum of the product=r_{left}*r_{right} (see Figure 5).

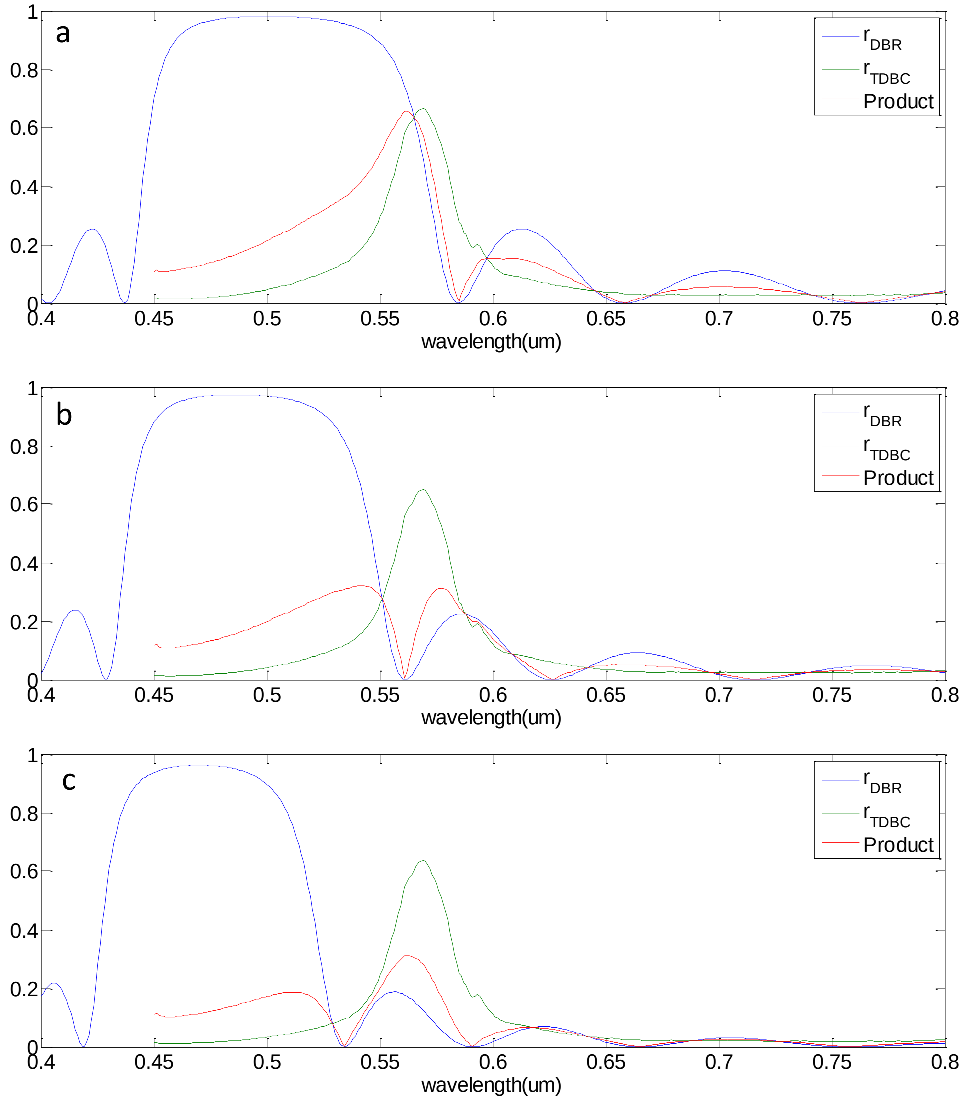

Figure 5. Amplitude of the Fresnel coefficients for a bare DBR with a central wavelength of 500 nm (r_{right}), a EOTS structure formed by the same DBR and 200 nm of a excitonic thin film (r_{left}) and the product of the two coefficients at different incidence angles. a) normal incidence, b) 26 degrees, c) 40 degrees.

The product function of the amplitude of the two Fresnel coefficients shows a minimum around the band edge position of the DBR. Specifically the product function shows two local maxima for larger and shorter wavelengths than the DBR band edge. We observe than when the mode condition is fulfilled (as at normal incidence, Figure 5a) the absolute maximum of the function product is located for wavelengths bellow the band edge. However, when the EOTS is not excitable, as at 40 degrees (see Figure 4c), the absolute maximum of the function product is located at larger wavelengths than the band edge. There is a transition region where the two local maximums are pretty similar, where we expect a hybrid mode (as at 26 degrees, Figure 5b). Therefore we could estimate at different angles (or wave vectors) the mode condition as the wavelength values where the local maximum of the product function is below the DBR band edge.

2.3. - Comparison of the dispersion curves obtained for the two methods with the experimental data

If we compare the dispersion curves obtained for the same EOTS structure but using the two different methods, we could observe that the two curves are similar but not exactly the same (Figure 10). The main difference is that the dispersion curve obtained from the product establish the mode condition at higher energies than the one by reflection. This difference is because product method searches the shorter wavelength where the mode condition is fulfilled, therefore it will follow the edge of the feature of the dip in reflectance. However the mode condition estimated by reflection follows the minimum in reflectance. Independently on this difference cut-off conditions are equivalent. In all the figures of the paper we have decided only show the dispersion curves obtained from the reflection simulations.

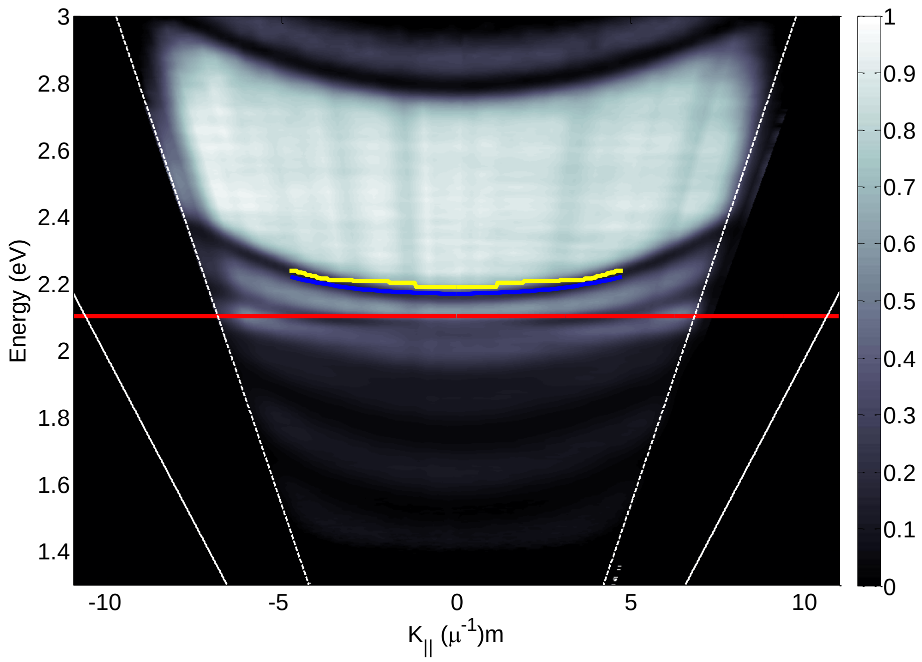

Figure 6. Experimental reflectance of the EOTS structure with a DBR band centre at 500 nm. The dispersion curve obtained from substraction function of the EOTS simulations and the bare DBR is shown in blue color. The yellow line is the dispersion curve estimated by the location of the maximum of the product of the Fresnel coefficients.

3- Verification of the obtained wavelength cut-off values. Analysis of field profiles.

In order to verify that our analysis and estimation of the cut-off wavelengths of the EOTS was correct we have plotted the intensity field profiles for the EOTS modes and compare with the intensity field profiles of the band edge mode of the bare DBR at the same angle. The figure 7 shows the reflectance values for a DBR with a central band at 500 nm and its equivalent EOTS structure. The estimated EOTS dispersion curve shows a cut-off wavelength of 557 nm at 26 degrees, determined when the substraction function cross the bare DBR edge. If we analysed the features on the reflectance simulated for the EOTS structure in figure 7.b it is not evident cut off wavelength position. From only reflection simulations of the EOTS (Figure 1.b) it is not clear when mode condition is dismissed and the minimum observed corresponds to the DBR stop band edge.

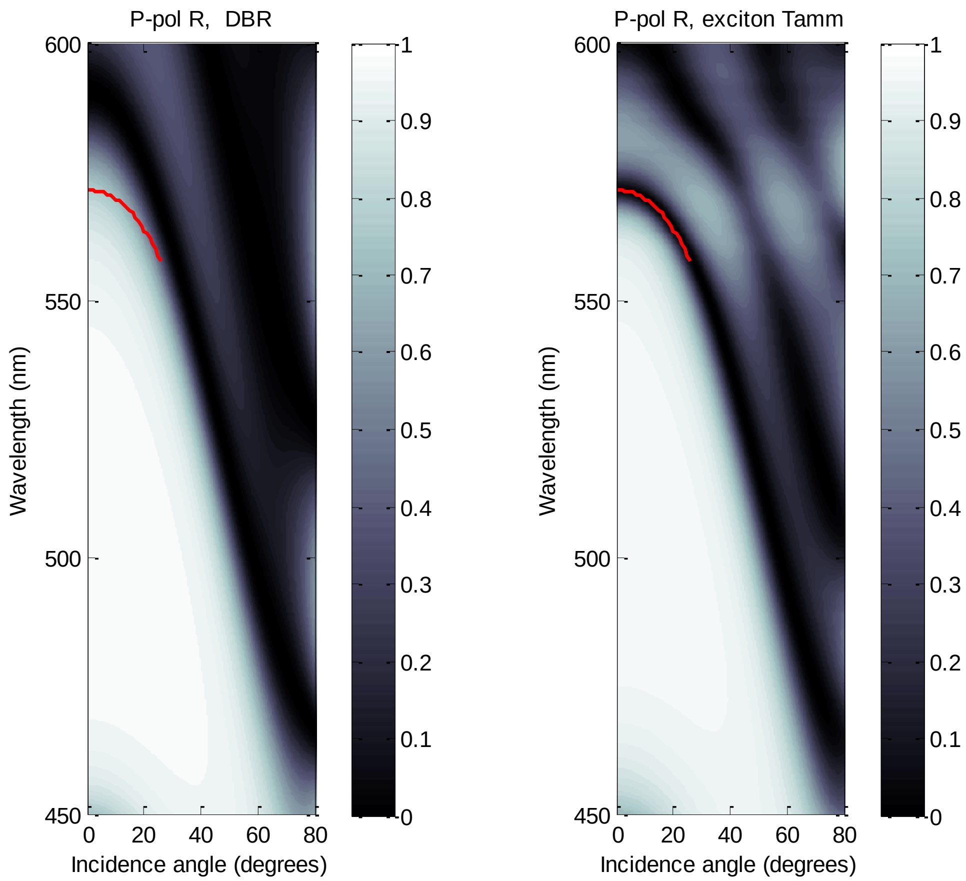

Figure 7. Contour plot of the P polarized reflectance of a) DBR with a central wavelength of 500 nm, b) EOTS structure with the same DBR and a 200 nm of our excitonic reflector. The red lines correspond to the estimated dispersion curves.

The electric intensity profiles for the EOTS mode and the band edge mode in the bare DBR at normal incidence are shown in the Figure 8. The intensity profile of the electrical field of an Optical Tamm State along the photonic structure is well establish on the literature. [1,2] The electrical intensity profile decays exponentially in the positive and negative directions from the interface between the DBR and the thin film. The most characteristic features are an oscillating decay within the DBR structure due to the periodic change of the refractive index and a faster decay in the thin film medium. In contrast, DBR edge mode should be a symmetric oscillating field as it can be observed in Figure 8.

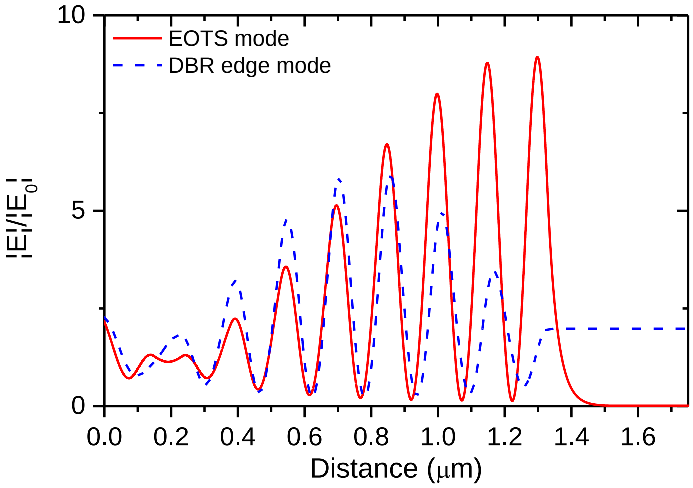

Figure 8. Electric intensity field profiles at normal incidence for at EOTS (wavelength=571nm) and a DBR edge mode (wavelength=590 nm) at normal incidence.

For the structure represented on Figure 7, from 0 degrees to 20 degrees the intensity field profiles supported by EOTS structure are similar to the characteristics of a OTS, confirming that the minimum observed corresponds to the EOTS mode condition. However from 20 to 30 degrees the mode transforms into an hybrid mode between a OTS mode and an DBR edge mode (see Figure 9).

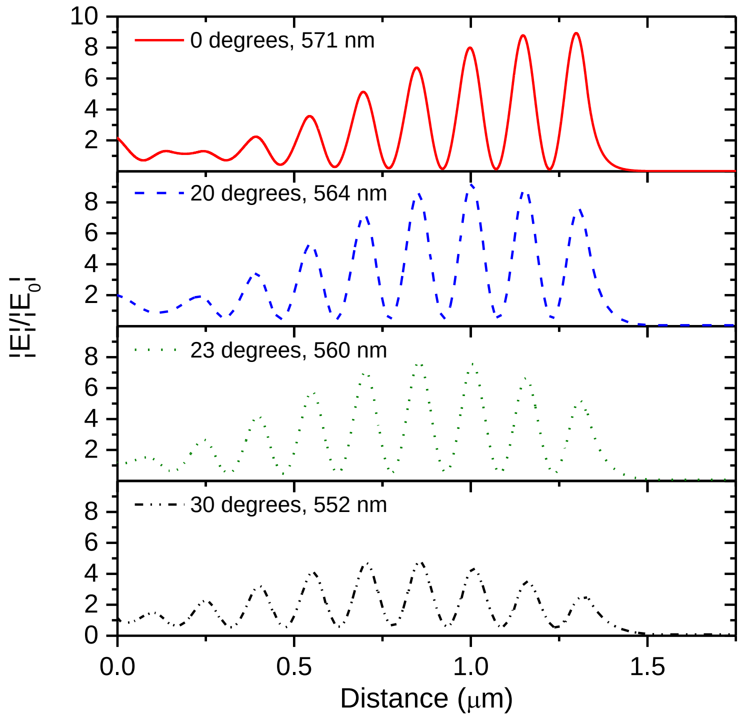

Figure 9. Electric intensity field profiles for at EOTS structure at different incident angles at mode condition (0, 20, 23 degrees) or DBR edge conditions (30 degrees).

The cut-off wavelength for this EOTS mode has been estimated around 26 degrees at 557 nm The mode associated to the minimum observed in the reflectance of the EOTS structure for larger angles and shorter wavelengths follows clearly the tendency of an edge mode without any exponential decay envelope, as it is shown in Figure 9 at 30 degrees.

4. Splitting between TE and TM modes of EOTS

As we mentioned on the paper, OTS are characterized by the splitting of between TE and TM polarized modes that increases quadratically as a function of the in-plane wave-vector. As OTS, EOTS dispersion curves follows the same tendency, with the difference that in the EOTS, the limited metal-like band causes different cut-off wavelengths between TE and TM modes for the same structure (see Figure 10).

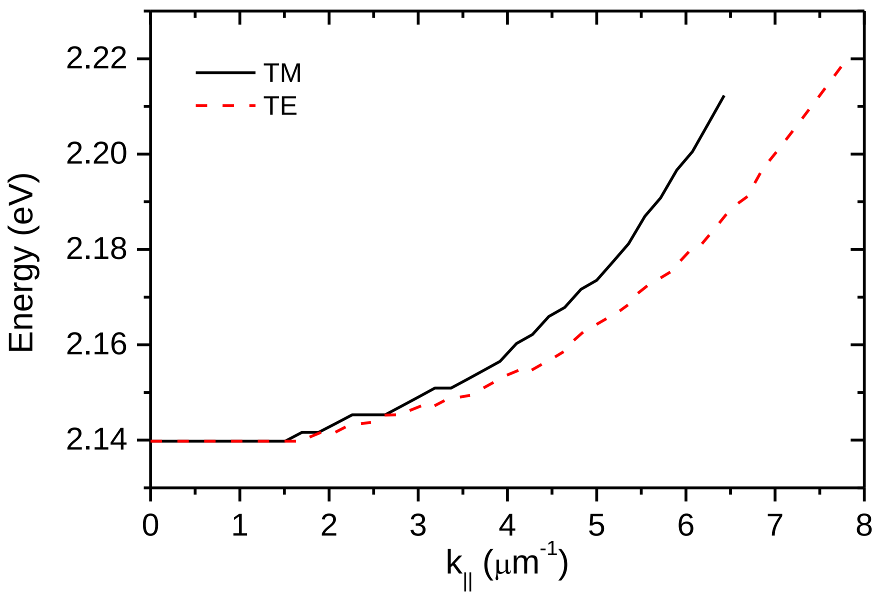

Figure 10. Dispersion curves of TE and TM modes estimated by reflection simulations for a EOTS structure formed by a DBR with a central wavelength of 520 nm and an excitonic layer of 200 nm.

REFERENCES

[1] M. Kaliteevski, I. Iorsh, S. Brand, R. Abram, J. Chamberlain, a. Kavokin, and I. Shelykh, Phys. Rev. B **76**, 165415 (2007).

[2] A. Kavokin, I. Shelykh, and G. Malpuech, Phys. Rev. B **72**, 233102 (2005).