Periodic Report on Simulation and Experimental Results

Plasmon Enhanced Photonics (PLEAS)

12 Month Report
Autumn 2007
www.eu-pleas.org

IST-FP6-034506

PLASMON ENHANCED PHOTONICS

OBJECTIVES OF THIS REPORT

This report represents the work carried out within the European project PLEAS (ISA-FP6-034506) on the 'Simulation and Design of Plasmonic Enhancing Structures'. After 12 months large advances have been made in our understanding of plasmon structures appropriate for light emitting diodes (LEDs) and photodetectors. The work presented here has been approved for public release by all partners. For up-to-date results please see the project website (www.eu-pleas.org).

PLASMONS IN LIGHTING INDUSTRY (PREVIOUSLY STATE OF THE ART)

Plasmonic phenomena have been investigated on light-emitting devices previously, focussing on the modification of the spontaneous emission by placing the metallic structure very close (< 50nm) to the emitter. The light can then be directly generated in a surface plasmon, which is then coupled to radiation by the appropriate structuring. In this way, non-radiative recombination processes can be suppressed. However, this technique is only beneficial for very low internal quantum efficiencies, initially most of the interest was for organic-LEDs and hence not applicable to high-efficiency state-of-the-art LEDs.

CONCEPT AND DESIGN OF PLASMON ENHANCING STRUCTURES

The state of the art in plasmonic phenomena such as enhanced transmission, beaming apertures, and simple light harvesting structures are just the start of research into plasmonic concepts that can enhance emission and photodetection. Many topics are to be addressed: what happens to enhanced transmission structures when placed on high index substrates? How do guided modes couple through sub-wavelength apertures? How can reflection losses be removed in light harvesting structures. The main differences between emitters and detectors is that plasmon enhanced structures, must be fully conductive for emitters and therefore only continuous structures will be considered, while for detectors the plasmon structures may be discontinuous as a conductive contact is not required.

The main topics which will be studied for emitter applications are hole arrays. For detectors light harvesting structures and circular gratings will be studied as well as field enhancing structures such as particles.

MODELLING

The modelling effort provides the theoretical framework, required to further understand the plasmonic phenomena of different devices. There are two main aspects which were concentrated

(1) The fundamental properties .

(2) Design and fabrication guidelines.

GENERAL MODELLING TECHNIQUES AND IMPROVEMENTS

Two numerical methods have been developed and used to simulate plasmonic structures. Each one of them with its own advantages and disadvantages.

- Mode-matching. This method is virtually exact for perfect conductors. The great advantage is that it provides readily the relevant modes involved in the optical phenomena. This results in an extremely fast computation time and therefore several parameters can be scanned easily. This method is very useful for understanding the fundamental mechanisms involved in enhanced transmission phenomena.

- Finite difference time domain (FDTD). This method is very efficient for complex system. It is the best method available for studying enhanced transmission at optical frequencies. The main disadvantage is that it is very time-consuming compared to the mode matching technique. Furthermore, accurate transmission spectra cannot be calculated for an infinite hole array illuminated at non normal incidence.

Within the first year of this project the mode matching method has been extended to include surface impedance boundary conditions. When these bound conditions are included the mode matching method has been shown to provide semi-quantitative results for real metals in the optical regime.

RECTANGULAR HOLES FOR LIGHT EXTRACTION FROM LEDS

There are many problems which need to be addressed when looking at light extraction of LEDs using plasmonic structures: angular emission, effect of substrate, metal compatibility with standard LED processing. One of the key ways in which to improve extraction efficiency of LEDs is to obtain angle independent emission. In standard LEDs a lot of attention has been placed on light extraction e.g. back reflectors, and surface roughness. However, there is still a large proportion of the light is reflected back into the structure. This light can be re-absorbed and remitted in the LED, the percentage of which depends on the internal quantum efficiency of the device.

Recent theoretical works predicted that the transmission of light through a single rectangular hole presents a higher transmittance than a square or circular hole with the same area. One of the resonances close to the cut-off has a peak transmittance controlled by the ratio between the long (a_y) and short (a_x) sides of the rectangle, see figure 1. If the metal layer is composed of a Perfect Electrical Conductor (PEC), this transmission peak develops at wavelength λ equal to $2a_y$ for the case of a p-polarized plane wave ($\lambda=2a_x$ for an s-polarized plane wave). It should be noted that, for the case of one single rectangular hole the resonance peak related to the cut-off does not shift when the angle of incidence is increased.

A theoretical analysis of an infinite array of rectangular holes was made in order to verify the possibility of a transmission peak independent of the angle of incidence. In particular, the objectives of the work consists of a study of the physical mechanisms underlying the phenomenon of extraordinary optical transmission to find the best set of geometrical parameters for future experimental investigations for industrial and commercial integrations.

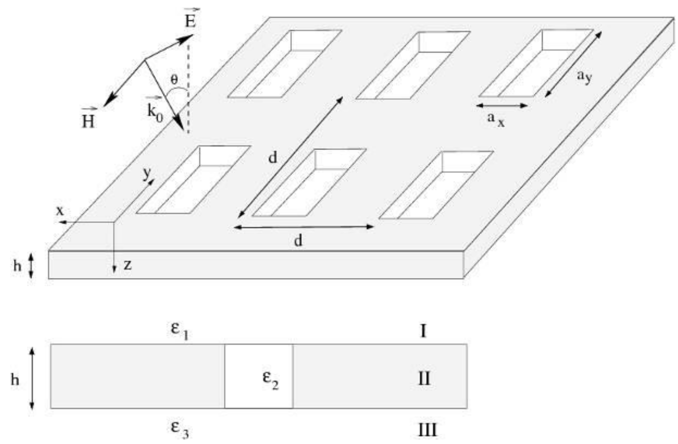

Figure 1: Diagram of an infinite array of rectangular holes of side a_x and a_y perforated on a metal film of thickness h. The structure is illuminated by an incident p-polarized plane wave. The electric field vector is pointing along the y-axis.

For this study a modal expansion technique coupled to the surface-impedance boundary conditions to simulate real metal at optical frequencies was used. This numerical method allows the study of normal incidence illumination as well as a non-normal incidence plane wave while keeping a reasonable computing time and a good comparison with full calculations and the FDTD was performed, see figure 2. In this case, medium I and medium III are made of vacuum or air i.e. n_1=n_3=1.

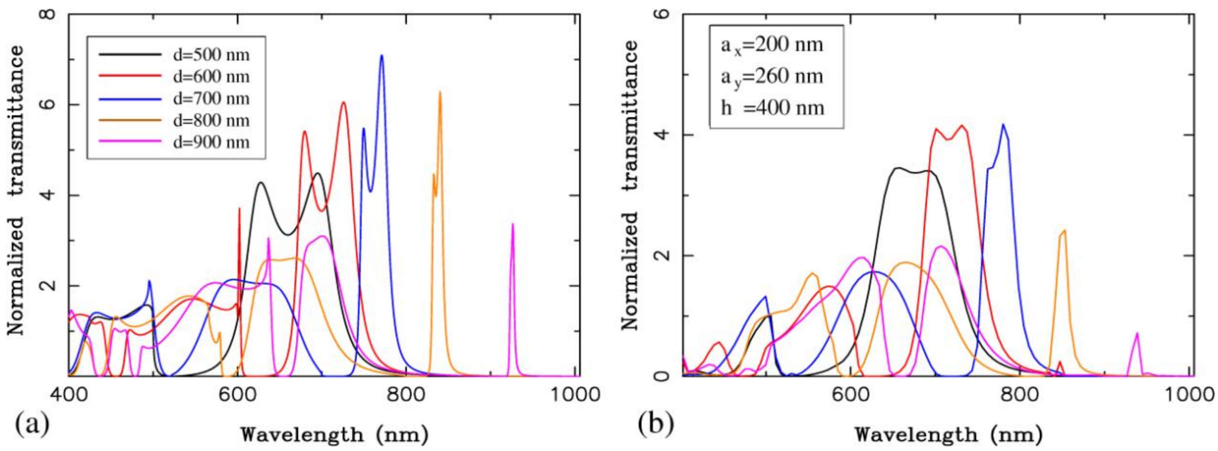

Figure 2: Comparison between the modal expansion technique (a) and FDTD calculations (b) for a p-polarized normal incidence plane wave and a real metal (Ag).

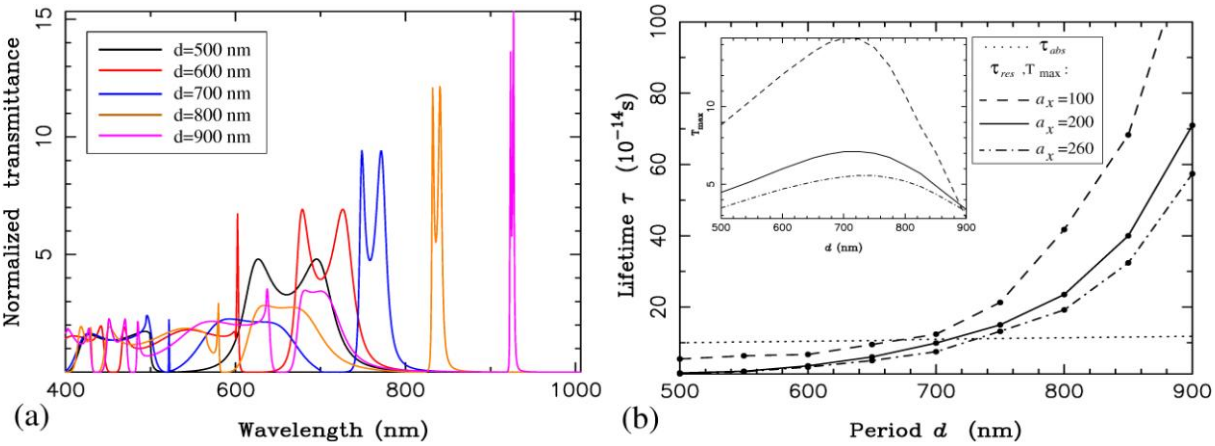

Figure 3: (a) Normalized-to-area transmittances calculated for different values of the lattice period d. Absorption of silver is neglected. (b) Lifetimes of the resonant process τ_{res} and for absorption, τ_{abs} (dotted line) versus period of the hole array.

From figure 2, it is clear that our simplified theoretical formalism is able to capture accurately the main features observed in the FDTD-spectrum. The locations and linewidths of the several peaks are well reproduced within our approach. The heights of the transmission peaks are higher in our approach, mainly due to the fact that absorption within the vertical walls of the holes is neglected. It is also important to note that the theoretical results of panel (b) are in very good agreement with the experimental data [A. Degiron and T.W. Ebbesen, J. Opt. A: Pure Appl. 7, No 2, S90-S96 (2005)]. We can note that the modal expansion and FDTD spectra present a kind of optimum value for d when looking at the evolution of the transmission peaks with the period of the array.

For the chosen parameters this optimum d is close to the cut-off wavelength, 695nm. Naively, this could imply that the optimum d appears when the resonant wavelengths of the two mechanisms (SPP and cut-off resonance) coincide. However, panel (b) of figure 2 demonstrates that the explanation is due to absorption by the metal. If the absorption in the metal is neglected, the heights of the transmission peaks grow with d like d^2, as it would correspond to a perfect transmission (100%) per unit cell. As explained previously, absorption along the walls of the holes is not taken into account in our approximated model. However, we have checked that for all d analyzed in this study, E-field intensity maxima are located at the horizontal metallic surfaces, where SIBCs are imposed within our modeling. Therefore, to consider only absorption on top and bottom surfaces of the metallic film is a reasonable approximation.

By looking at the linewidth of the transmission peaks with no absorption (Panel (a) figure.3), we can extract the lifetime associated with the resonant process, t_{res} . From the knowledge of the dielectric function of the metal, we can estimate the time taken for a photon to get absorbed, t_{abs}. It is expected that optimum d would appear where $t_{res}(d) = t_{abs}(d)$. When $t_{res}(d)$ is much smaller than $t_{abs}(d)$, photons are mainly transmitted and they are not absorbed by the metal. Absorption plays a minor role in the transmission process and the normalized-to-area transmittance at resonance increases when d is increased, as seen in figure 3. In the other limit ($t_{abs}(d) < t_{res}(d)$), photons are absorbed by the system before the resonance is built up. As $t_{res}(d)$ grows rapidly with d, a decrease of the transmittance at resonance versus d is expected to occur in this limit.

Absorption by metal layer is detrimental for LED extraction. This study reveals that the dependence to the absorption by the metal is less important for transmission peaks related to the cut-off than for SPP peaks due to grating. This result suggests that the cut-off peak has to be used and optimized for LED operation. Calculations are underway to know if these conclusions obtained for rectangular hole arrays can be generalized for all kind of hole shapes (annular, slits,...).

ANNULAR HOLE ARRAYS FOR HARVESTING LIGHT ONTO PHOTODETECTORS

A theoretical analysis of an infinite array of annular holes was performed in order to verify the possibility of a transmission peak independent of the angle of incidence which may improve light extraction in LEDs. Optical transmission through arrays of annular holes has been considered in the literature but mainly at normal incidence. Moreover, the effect of a dielectric substrate has not yet been analysed in detail as it is an important issue in plasmonic devices for solid state lighting devices. Additionally, the optical properties of annular waveguides are essential for the optimisation of the bulls' eye geometry, therefore the calculation for hole arrays helps to understand how to deal mathematically with these modes. The bull's eye geometry with an annular structure is thought to be potentially very useful for harvesting structures which may improve the signal to noise ratio in photodetectors. For this study, the modal expansion technique was used. In this first approach perfect conductor boundary conditions were used. In the future, surface impedance boundary conditions will be incorporated in the calculation.

As the metal is considered as a PEC, all results can be made applicable to different frequency regimes simply by scaling all length scales by the same factor. The unit of length is the period d. Figure 4 shows the transmittance through an array of annular holes with external radius a=0.45 and internal radius b=0.4, when the array is illuminated by p-polarised plane waves impinging at different angles. The metal thickness considered in the results presented is w=0.4, for a d=600nm which corresponds to 240nm, these are typical experimental parameters.

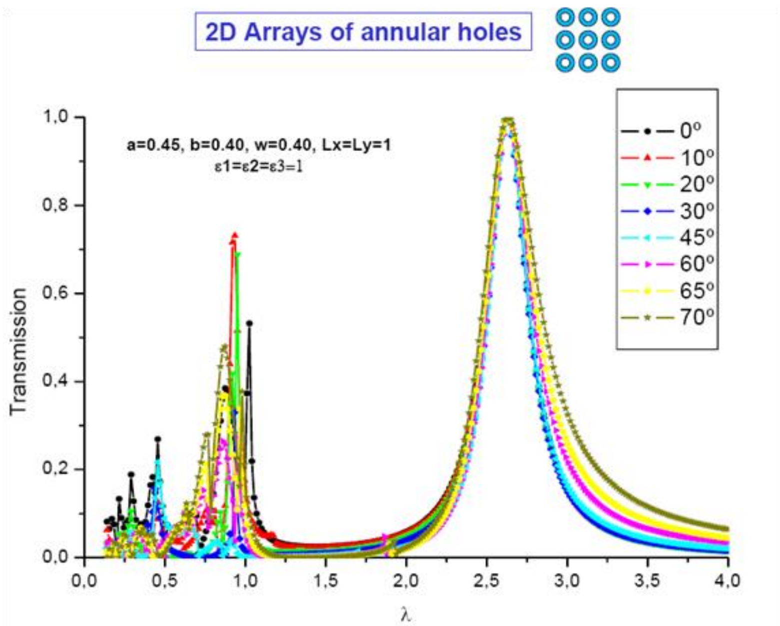

Figure 4 Dependence with angle of incidence for the normalized-to-area transmittance through an array of annular hole, for the case of a=0.45, b=0.4.

These calculations show that, as well as the typical extraordinary transmission (EOT) peak at wavelengths close to the period; there is another EOT peak, which is almost independent of angle of incidence. This peak occurs close to the cut-off wavelength of the annular hole. Recall that the cut-off wavelength for a circular hole of radius an is $\lambda^{circ}_c=2\lambda a/1.84$. Surprisingly, an annular hole defined by internal radius b, as well as external radius a, has a longer cut-off wavelength, even though the field is confined to a smaller spatial region. Actually, this is the situation for the mode TE11, for which $\lambda^{annular}_c \approx 2\lambda a$ in the limit b → a. Additionally, the annular hole has a TEM propagating mode with no cut-off, which has not been considered here because its modal shape is radial, and as such it couples very poorly to a plane wave (the overlap with a normal incidence plane wave would be zero, due to symmetry).

For typical parameters in the device, the values for a and b considered in figure 4 are too close for the fabrication capabilities as the equivalent distance is 30nm. However it was considered as a limiting case.

EXPERIMENT

To characterise the behaviour of the plasmon enhancing structures through surface and optical measurements, there are several techniques which are available in the consortium including techniques for both the near field, such as confocal microscope, scanning near-field optical microscopy and standard far field measurements such as spatially resolved transmission spectrum. Some of the optical experiments were specifically adapted for characterising photodetectors. Characterisation techniques such as scanning electron microscope (SEM) and atomic force microscopy (AFM) are essential tools to characterise the plasmonic structures.

The main experimental investigations for the first year were

(1) Surface roughness and the effect on plasmon structuring.

(2) Characterisation of plasmon properties of metals compatible with plasmon structures.

(3) Spectral measurements of harvesting structures for photodetectors.

SURFACE ROUGHNESS

Contacts for LEDs are not normally just gold but often contain a zinc or aluminium layer designed to decreased resistance to obtain good adhesion and avoid diffusion. However, plasmons in aluminium and general fabrication techniques are less advanced. Three layers were investigated.

- 100nm of gold
- 100nm of aluminium
- 20nm of aluminium and 80nm of gold on top

It can be seen from figure 5 that the aluminium layers have an adverse effect on the grain size. Whilst gold, far left of figure 5, shows a route mean square (RMS) roughness of only 2.3nm adding even a 20nm layer give and increase to near twice this of 3.5nm Moreover pure aluminium roughness is 4 times greater than gold.

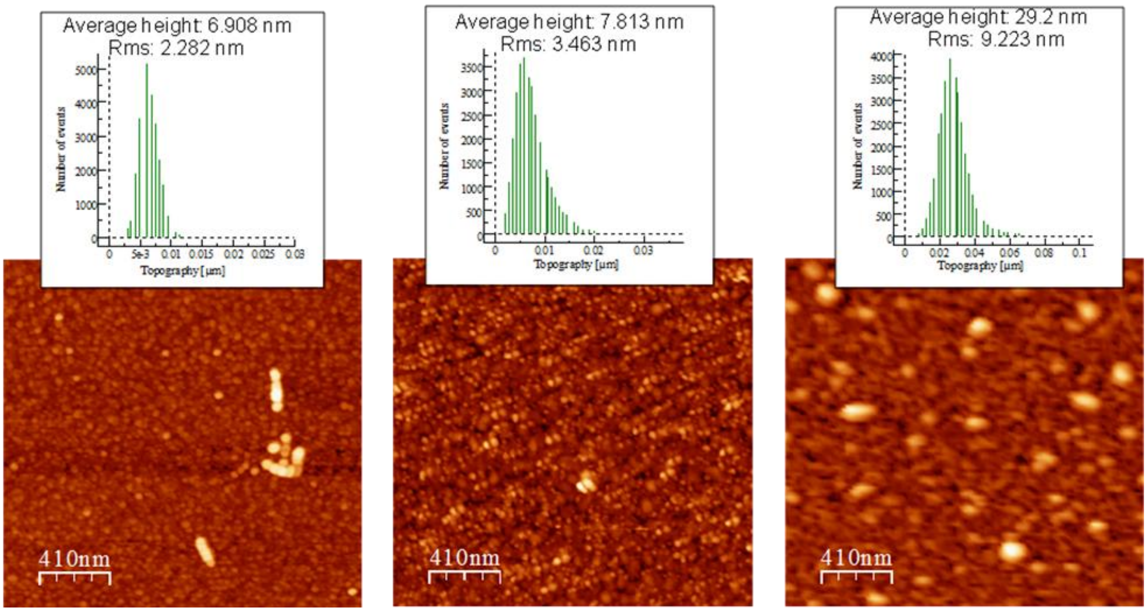

Figure 5: AFM images from left to right of 100nm Au, 20nm Al and 80nm Au and 100nm of Al metal layers. The grain size can clearly be shown to increase as the content of aluminium increases. The inset shows a statistical analysis of the average height and the RMS roughness taken over a constant surface area.

The effect of roughness on fabricating plasmonic structures at the nanoscale was shown to be very important. This is due to the fact that when etching using focussed ion beam (FIB) the different grain sizes etch at different rates. This results in an inverse relationship between grain size and feature resolution which can be etched, see figure 6.

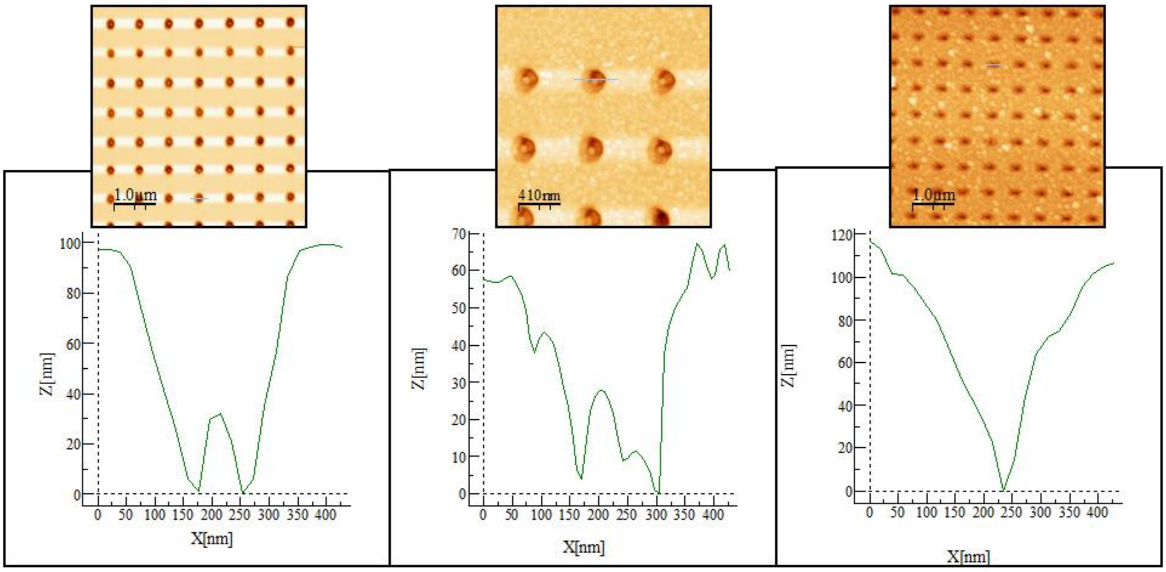

Figure 6: SEM images from left to right of 100nm Au, 20nm Al and 80nm Au and 100nm of Al. When fine details are etched using FIB in these layers it can be seen that the resolution obtainable is directly linked to the grain size. For the smoothest layer Au, far left, it can be seen that a features of 100nm can be etched. These features completely disappear in the pure aluminium layer, far right.

CHARACTERISATION CONTACT METALS

Plasmons in aluminium are less studied than those in gold and in general fabrication techniques are less advanced in view of this layers of gold, aluminium & gold (Al/Au 20/80) and aluminium where characterised. Ellipsometry measurements were carried out on the layers in order to have information on the different complex dielectric functions (ε). For the aluminium-gold layer, the complex dielectric function is essentially that of the top layer of gold. For the pure aluminium layer, when one compares the measured values for the aluminium ε with that found in the literature, as illustrated on figure XX, we notice that the aluminium layer has been oxidised. Pure aluminium cannot be used in a practical device due to this oxidation.

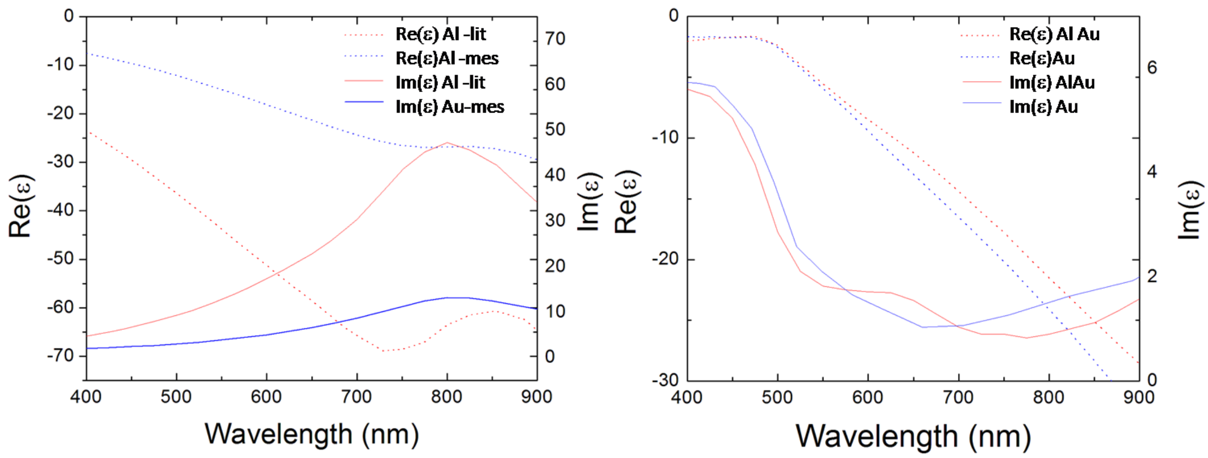

Figure 6: Ellispsometry measurements which compare the measured values of the imaginary and real dielectric complex function of silver and gold with literature values, taken from Palik. LHS: The measured values of aluminium (20nm) gold (80nm) are similar to that of pure gold.

OPTIMISATION OF BULLS' EYE STRUCTURES FOR PHOTODETECTORS

Bulls' eye structures, consisting of a central aperture surrounded by concentric grooves, were first reported several years ago but the progress in understanding of the extraordinary transmission phenomenon since then warrants a new look at these structures and how they can be optimized for the purpose of photodetectors. The bulls' eye structures have several parameters that can be varied such as the period, the groove depth and width, the number of grooves, etc., and these have been analyzed. This is illustrated in figure 7, where the transmission intensity is plotted as a function of the central hole diameter. As can be seen, at diameters less than 350nm, the transmission is very weak while above this diameter size the transmission increases rapidly.

Considering the resonance wavelength of ca. 720nm, these two regimes correspond to a transition through the cut-off wavelength of the hole. In other words, as the diameter of the central hole is larger than half the resonance wavelength, light can freely propagate through the aperture. For the purpose of photodetectors there is no need to have a tiny hole and one should clearly choose one that is as large as possible without sacrificing other spectral features.

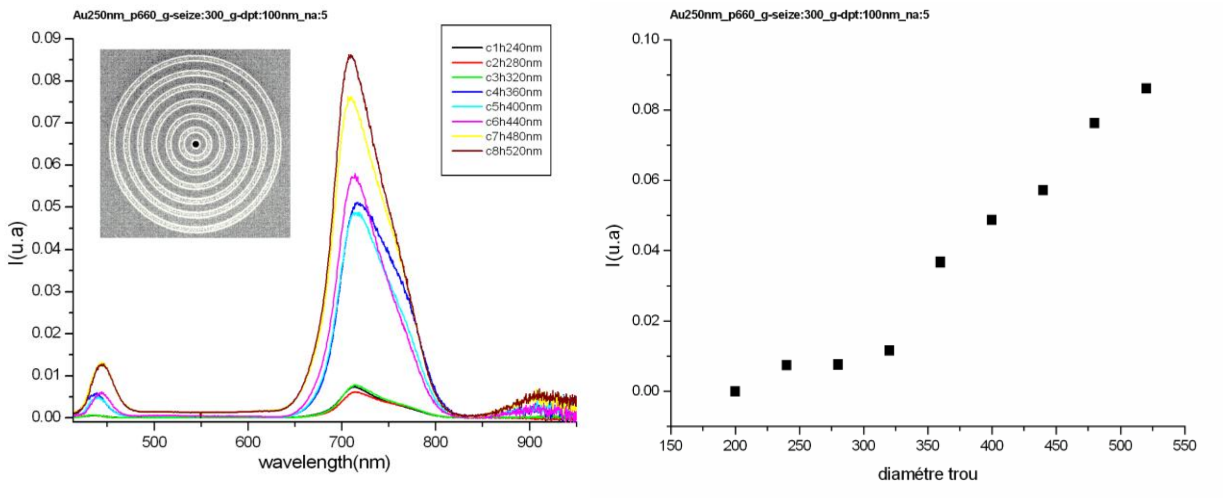

Figure 7: Transmission of Bulls' eye structure (insert) as a function of hole diameter. The structures were prepared in 250nm thick silver on glass substrate.

CONCLUSIONS

In this first year important utilities were put in place both for modelling and experimental work.

Several important areas for the implementation of plasmon structures to obtain improved lighting devices have been addressed. Both rectangular and annular structures were theoretically studied as possible grid structures which could be used to both inject current and increase light emission in LEDs. The angular dependence of annular structures was studied and it was shown that no angular dependence is seen for the high transmission peak at the cut-off wavelength. The fabrication of plasmonic structures was also investigated and it was shown that when etching plasmonic structures it is imperative to obtain small grain sizes if high resolution structures are needed. Metals which can be used, such as aluminium with a capping gold layer, as contacts have been investigated and both their surface roughness and their dielectric functions were measured. Bulls' eye structures were fabricated and measured with varying central hole diameters.

THE CONSORTIUM

The fact that the PLEAS project stretches from the fundamentals of plasmons to the industrial application is reflected in the consortium members..

Institute	Country	Project leader
• Centre Suisse d'Electronique et de Microtechnique SA (CSEM)	Switzerland	Ross Stanley (Co-ordinator)
• Universidad Autonoma de Madrid (UAM)	Spain	Francisco J. Garcia-Vidal
• Universidad de Zaragoza (UNIZAR)	Spain	Luis Martin-Moreno
• Osram Opto Semiconductors GmbH (Osram OS)	Germany	Reiner Windisch
• The Queen's University of Belfast (QUB)	United Kingdom	Anatoly Zayats
• Technische Universität Dresden (TUD)	Germany	Lukas M. Eng
• Université Louis Pasteur de Strasbourg (ULP)	France	Thomas Ebbesen
• SAGEM Défencse Sécurité (SAGEM)	France	Eric Agostini

For more details on the consortium, and other aspects of the project please visit www.eu-pleas.org.