Dr. Gabriel Christmann NanoPhotonics Centre Department of Physics University of Cambridge JJ Thomson Ave, Cambridge, CB3 OHE Phone: +44 (0)1223 337446 e-mail: gprmc2@cam.ac.uk

Scientific report

POLATOM ESF Research Networking Programme

I. Purpose of the visit

There is currently a strong on-going collaboration between the group of Professor Pavlos Savvidis at the University of Crete, the visited institution, and the group of Professor Jeremy Baumberg at the University of Cambridge, the home institution. This has led to the publication of numerous journal articles in the past years.

Building up on the experience of the Crete group on polariton-based electrically injected devices, named polariton light emitting diodes (LEDs)¹, we used similar structures to apply electric field to quantum wells (QWs) inserted inside optical microcavities. This has led to the observation of several new effects in semiconductor microcavities. By applying electric field to microcavities with sets of weakly coupled QW pairs we are able, by a careful alignment of electronic levels, to initiate LO-phonon assisted electron tunnelling, which in turn allows switching on and off the parametric scattering process^{2,3}. This could enable the realization of ultrafast light modulators. We also proposed and demonstrated the concept of dipolaritons, which are observed in strongly coupled microcavities using asymmetric double quantum wells (ADQWs) as active region⁴. This new type of polaritons exhibits a strong dipole moment, and therefore should help to lower the threshold of polariton condensation. In addition such structures pave the way to the observation of many unusual effects. The resulting three-state system yields dark polaritons analogous to those in atomic systems or optical waveguides offering new possibilities for electromagnetically induced transparency, and adiabatic photonic to electronic transfer⁵.

In parallel to these electrically controlled samples through our collaboration we obtained a numerous results on unprocessed sample. Using a distributed Bragg reflector (DBR) structure with QWs inserted in the Bragg stack we demonstrated strong coupling between QW exciton and the Bragg mode as well as parametric amplification⁶. By studying a high quality (Q) factor microcavity we also observed a collection of new effects in polariton condensates: spontaneous oscillations⁷, sunflower patterns⁸, and vortex lattices⁹.

The purpose of the visit is to push further the above-mentioned results, and identify new samples and directions to explore.

As mentioned in the grant application it would be highly desirable to study the effect of electrical control and of dipolaritons on the nonlinear properties. We observed that these samples are plagued by an important electron tunnelling outside of the quantum wells at the bias that are commonly used for our measurements. This creates a strong channel for non-radiative losses. It is desirable to work with samples with higher barriers preventing these unwanted losses. Furthermore, our results on polariton condensation showed the crucial need of high quality factor microcavity to observe the nonlinear effects while staying in the strong coupling regime. It is then required to combine the high quality samples of the polariton condensation experiment with electrically connected structures to explore the possibility of controlling polariton condensation.

During this visit we therefore studied a sample with GaAs/AlGaAs triple QWs barriers and high quality factor, with doping which is necessary for the realization of a polariton LED, and would be promising for electrical control of polariton lasing.

Furthermore to extend polariton physics to new types of systems we explored the possibility to observe the strong coupling regime with generalized Brewster angle modes¹⁰.

II. Description of the work carried during the visit

A. High Q factor polariton LED

In order to realize an electrically injected polariton laser or to study electrical control on nonlinear effects such as polariton lasing, it is highly desirable to work on high quality factor samples with doping of the DBRs in order to have a *p-i-n* junction. In this context a new microcavity sample has been grown at the University of Crete. It consists in an Al_{0.15}Ga_{0.85}As cavity layer with four sets of three GaAs/Al_{0.15}Ga_{0.85}As QWs inserted at the antinodes of the electric field. QW and barrier thicknesses are 9 nm and 12 nm, respectively. The cavity layer is surrounded by a 35 pair *n*-type bottom DBR and a 32 pair *p*-type top one. Finally the sample is processed into mesas with a ring shaped Ti/Pt electrode deposited after a second etch step to contact the lower *p* layers, improving the series resistance. This allows to electrically injecting polaritons in forward bias or to apply electric field in reverse bias.



Figure 1. Photoluminescence images versus angle images at different sample bias.

Initial measurements on this sample showed a very fast tuning versus bias of the anticrossing features on the angle dispersions, as observed in figure 1. The considered bias changes correspond to fields of several kV/cm, which would not lead to significant exciton energy shifts by quantum confined Stark effect in single quantum wells. Such shifts would correspond more to indirect transitions between adjacent quantum wells, which is confirmed by the observation of more than one anticrossings on the dispersions of figure 1 (possibly the signature of a direct and an indirect transition. The problem with this explanation is that given the structure design, especially the QW barrier height coupling between QWs is not expected and therefore the indirect transitions are expected to have negligible oscillator strength due to very weak overlap between electron and hole wavefunctions.

During this visit we tried to assess whether it is possible to have signature of these indirect transitions in the current sample, by checking if the actual sample properties corresponds to the one specified during growth. In addition we developed a simple model describing what type of bias dependant and angle dependant dispersions are expected in case significant tunnelling coupling in QWs is present.

B. Strong coupling with generalized Brewster angle

Quarterwave layer stacks are widely used to produce high reflectivity dielectric mirrors, called DBRs, for numerous applications. In our case for example they are the basic building block of our semiconductor microcavity samples. It is widely known that these stacks have different properties in transverse electric (TE) and transverse magnetic (TM) polarizations. In particular, as shown in figure 2a) and b), these DBRs show at very large angle a resonant mode in TM polarisation while no mode is observed in TEⁱ. In fact the more pair the DBR has, the closer to 90 degree the mode is, the mode ultimately converging to a surface mode for infinite number of pairs. This mode has been described as generalized Brewster angle¹⁰.



Figure 2. Typical DBR angular dependant reflectivity images (GaN/AlInN material parameters). a) TE polarization. b) TM polarization, the generalized Brewster angle mode is circled. c) High angle reflectivity image for the sample in Ref. 6 with QW oscillator strength (simulation).

We propose to take advantage of this mode to do light matter coupling. The sample we used is the one in which we observed strong exciton photon coupling of Bragg polaritons⁶. It consists in an optimized unfolded microcavity in which QWs are incorporated into a Bragg mirror stack. The sample is a stack of quarter wavelength thick alternating high refractive index ($n_{GaAs} \sim 3.5$) GaAs layer and of a smaller effective refractive index ($n_{eff} \sim 3.2$) pseudolayer AlAs/GaAs/InGaAs/GaAs/AlAs. The 10 nm wide In_{0.1}Ga_{0.9}As QWs are placed symmetrically inside the pseudolayer at the antinodes of the electric field. Such sample should also exhibit strong exciton photon coupling with this generalized Brewster angle mode, as seen on the simulation in figure 2c).

During this visit we tried to measure these high angle modes through reflectivity, which can be quite challenging as they are above 89 degrees. We also tried to see if strong exciton photon coupling can be observed.

ⁱ In case the top DBR layer is made of the low refractive index material the mode is observed for TE polarization.

III. Description of the main results obtained

A. High Q factor polariton LED

In order to assess the effect of quantum well coupling on the field dependant dispersion curves the following simple model was developed similarly to what we used in Ref. 5. We considered our triple well MC structure with possibility to observe both Rabi splitting due to exciton photon coupling (constant Ω) and also tunnel coupling (constant J) between electronic levels. Through tunnelling coupling the QWs, which would be independent otherwise, start to mix, which enables the possibility to see strong coupling with "indirect excitons". We write the following matrix Hamiltonian:

$$H = \begin{bmatrix} E_{e1} & J/2 & 0\\ J/2 & E_{e2} & J/2\\ 0 & J/2 & E_{e3} \end{bmatrix}$$

Where $E_{e1...3}$ are the energies of the electronic level of QW number 1 to 3. The eigenvalues of this matrix give the triple coupled well energy levels, and the corresponding eigenvectors tell for each eigenvalue the fraction of each well. The next step is to calculate the strong exciton photon coupling. We consider that each of the three previously calculated electronic eigenstate is likely to form exciton with the hole levels (we only consider heavy hole and neglect tunnelling for holes). This leads to 9 exciton levels, the matrix Hamiltonian including coupling to the cavity mode is therefore 10x10. For the exciton photon coupling constant we add renormalize the Ω coupling constant with the fraction of the electron eigenstate which is in the same QW than the corresponding hole level. We are finally able to quickly calculate field dependant dispersions as a function of exciton photon coupling.

We also checked if the barrier composition was matching the specified one. For that we performed photoluminescence on a cleaved facet to see the emission energy of AlGaAs. The energy we measured is in fact indicating a lower Al content than expected (around 10%). This indicates that QW coupling is indeed possible and that we are likely to observe indirect transitions. At the moment it is however difficult to explain our experimental data with the simple model above. The results on this sample are still under investigation.

B. Strong coupling with generalized Brewster angle

We performed high angle reflectivity on the Bragg polariton sample to see whether we are able to observe this generalized Brewster angle mode, and strong exciton photon coupling with it. The high angle reflectivity setup is described in figure 3a). We use a halogen lamp focused just on/slightly above the edge of the sample using a lens. Through this lens numerical aperture we illuminate the sample with a small angular range close to 90 degree. We collect this reflected light through a lens, and by placing a collection optical fibre in the backfocal plane of this lens we are able to collect reflected light from a very small angle range close to 90 degree. We then analyse the reflected light in a spectrometer. All the measurements are performed in a cryostat at 16 K.

Figure 3b) displays polarization dependant high angle reflectivity spectra at negative detuning. The generalized Brewster angle mode is clearly observed for TM polarization and disappears in TE polarization as expected. We are therefore able to probe this mode with our setup. To explore strong exciton photon coupling with such a mode we move across the sample wedge in order to

bring the mode energy in resonance with the exciton. When we do that, as illustrated in figure 3c) we observe a clear anticrossing, which proves strong exciton photon coupling for the first time which such a mode.



Figure 3. a) Schematic description of the experimental setup. b) Polarization dependant reflectivity of the sample at high angle. c) Position dependant reflectivity spectra in TM polarization. Lines are guide to the eye.

IV. Future collaborations

The two institutions have a long history of collaboration and we have several on-going projects in which we will collaborate in the future. In this section I will only describe future collaborations in the continuity of what has been done during the visit.

A. High Q factor polariton LED

As mentioned above our understanding of this sample is still not complete and further work will be done in collaboration. We have already decided to perform photocurrent measurements in the University of Cambridge (the home institution), were we have a tuneable low linewidth continuous wave titanium Sapphire laser. The advantage of such measurement is that it is more sensitive to detect indirect transitions than the nonresonant photoluminescence as previously observed on our ADQW samples¹¹.

B. Strong coupling with generalized Brewster angle

We will continue to collaborate on structures based on this generalized Brewster angle. Indeed current sample was not optimized for operation with this mode; it was optimized for Bragg polaritons. In particular QWs were inserted with all the DBR pairs which is clearly not optimum as the field distribution of this mode is mostly close to the surface. New samples with only QWs In the top layer would be highly desirable. Furthermore it would be nice to observe emission on such samples. We will therefore try to observe photoluminescence on such samples.

V. Projected publications

After this visit we hope to write one publication for each of the samples that have been studied. For the high Q factor polariton LED sample we clearly need further measurements, as our understanding is currently incomplete. On the other hand for the generalized Brewster angle polaritons we are very close to write a paper as we clearly observe strong coupling with this mode for the first time.

VI. Other comments

I would like to thank everyone at the University of Crete for welcoming me during these six weeks and in particular Pavlos Savvidis, Peter Eldridge, Simos Tsintzos, Panos Tsotsis, and Marina Tzanakaki.

I would also like to thank the European Science Foundation Polatom Research Networking Programme for funding me during these six weeks.

- ⁶ A. Askitopoulos, L. Mouchliadis, I. Iorsh, G. Christmann, J. J. Baumberg, M. A. Kaliteevski, Z. Hatzopoulos, and P. G. Savvidis *Phys. Rev. Lett.* **106**, 076401 (2011).
- ⁷ G. Tosi, G. Christmann, N. G. Berloff, P. Tsotsis, T. Gao, Z. Hatzopoulos, P. G. Savvidis, and J. J. Baumberg *Nature Physics* **8**, 190 (2012).
- ⁸ G. Christmann, G. Tosi, N. G. Berloff, P. Tsotsis, P. S. Eldridge, Z. Hatzopoulos, P. G. Savvidis, J. J. Baumberg, *Phys. Rev. B* **85**, 235303 (2012).
- ⁹ G. Tosi, G. Christmann, N. G. Berloff, P. Tsotsis, T. Gao, Z. Hatzopoulos, P. G. Savvidis, J. J. Baumberg Submitted to *Nature Communications*.
- ¹⁰ H. F. Mahlein *J. Opt. Soc. Am.* **64**, 647 (2012).

¹¹ C/ Coulson, G. Christmann, P. Cristofolini, C. Grossmann, J. J. Baumberg, S. I. Tsintzos, G. Konstantinidis, Z. Hatzopoulos, and P. G. Savvidis, *Submitted to Phys. Rev. B*.

¹ S. I. Tsintzos, N. T. Pelekanos, G. Konstantinidis, Z. Hatzopoulos, and P. G. Savvidis, *Nature* **453**, 372 (2008).

² P. G. Savvidis, J. J. Baumberg, R. M. Stevenson, M. S. Skolnick, D. M. Whittaker, and J. S. Roberts, *Phys. Rev. Lett.* **84**, 1547 (2000).

³ G. Christmann, C. Coulson, J. J. Baumberg, N. T. Pelekanos, Z. Hatzopoulos, S. I. Tsintzos, and P. G. Savvidis *Phys. Rev. B* **82**, 113308 (2010).

⁴ G. Christmann, A. Askitopoulos, G. Deligeorgis, Z. Hatzopoulos, S. I. Tsintzos, P. G. Savvidis, and J. J. Baumberg *Appl. Phys. Lett.* **98**, 081111 (2011)

⁵ P. Cristofolini, G. Christmann, A. Askitopoulos, G. Deligeorgis, Z. Hatzopoulos, S. I. Tsintzos, P. G. Savvidis, and J. J. Baumberg, Science **336**, 704 (2012).