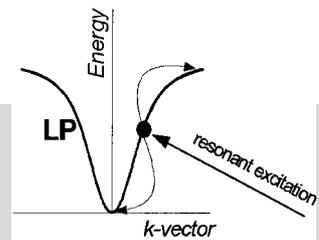


Stimulated Polariton Scattering in Semiconductor Microcavities: New Physics and Potential Applications

By Alexander I. Tartakovskii, Maurice S. Skolnick,*
Dmitrii N. Krizhanovskii, Vladimir D. Kulakovskii,
R. Mark Stevenson, Raphaël Butté, Jeremy J. Baumberg,
David M. Whittaker, and John S. Roberts

Stimulated polariton scattering in semiconductor microcavities is reported. The phenomena arise from the bosonic character of the coupled exciton–photon modes of the system. The ability to manipulate and control the polariton dispersions in microcavities is shown to be a key factor underlying the new observations. The potential of such phenomena to form the basis of a new type of coherent light source (“polariton” lasers) and highly efficient optical parametric oscillators is discussed.



1. Introduction

Semiconductor microcavities are micrometer scale photonic structures in which quantum wells (QWs) are embedded within a high finesse Fabry–Perot cavity, the whole structure being prepared by high precision, modern crystal growth techniques.^[1] In such structures vertical confinement of both excitons in the quantum wells and of light within the Fabry–Perot

cavity results in strong and controllable light-matter interactions unachievable in bulk semiconductors. This control has opened up a new field of exciton–polariton physics, where key features of the interacting exciton–photon system can be tailored by sample design. Most importantly for the physics described here, the dispersion curves of the coupled two-dimensional (2D) exciton–photon modes, exciton–polaritons (termed cavity polaritons), differ from those of their bulk analogues since the confinement of light results in a finite energy at $k = 0$. This property, combined with the controllable dispersion and the extremely low density of polariton states,^[1] has recently allowed a variety of new phenomena to be observed, including final state stimulation and a new condensed phase with macroscopic coherence, which have the potential to lead to new devices including very low threshold optical parametric oscillators and a coherent light source based on stimulated polariton scattering.

A schematic diagram of a typical structure is shown in Figure 1a. The structure consists of a $3\lambda/2$ GaAs cavity (λ is the wavelength of light in the medium) surrounded by 20 (below) and 17 (above) layers of $\text{Al}_{0.13}\text{Ga}_{0.87}\text{As}/\text{AlAs}$ high reflectivity Bragg mirrors (BMs). The quantization of light in the vertical direction with free propagation within the plane leads to the approximately quadratic photon dispersion shown in Figure 1b. Two sets of three $\text{In}_{0.06}\text{Ga}_{0.94}\text{As}$ quantum wells are embedded within the GaAs cavity and provide 2D excitonic

[*] Prof. M. S. Skolnick, Dr. A. I. Tartakovskii,^[+] Dr. R. M. Stevenson, Dr. R. Butté
Department of Physics and Astronomy, University of Sheffield
Sheffield S3 7RH (UK)
E-mail: M.Skolnick@sheffield.ac.uk

D. N. Krizhanovskii, Prof. V. D. Kulakovskii
Institute of Solid State Physics, Russian Academy of Sciences
Chernogolovka 142432, Moscow reg. (Russia)

Prof. J. J. Baumberg
Department of Physics and Astronomy, University of Southampton
Southampton SO17 1BJ (UK)

Dr. D. M. Whittaker
Cavendish Laboratory, University of Cambridge
Cambridge CB3 0HE (UK)

Dr. J. S. Roberts
Department of Electronic and Electrical Engineering
University of Sheffield
Sheffield S1 3JD (UK)

[+] Second address: Institute of Solid State Physics, Russian Academy of Sciences, Chernogolovka 142432, Moscow reg., Russia.

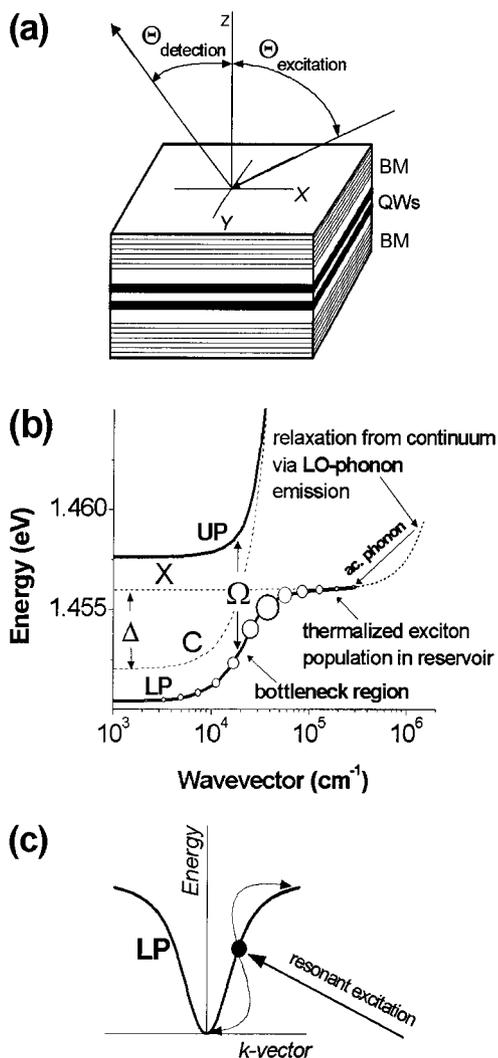


Fig. 1. a) Schematic diagram of the sample and excitation/detection geometry. b) Polariton (thick lines) and uncoupled cavity mode (C) and exciton (X) dispersions. The mechanisms responsible for relaxation following non-resonant excitation are indicated. The polariton population is represented schematically by the open symbols, with the maximum occurring at the edge of the polariton trap. c) Schematic diagram for resonant excitation into the polariton trap.

states which are also confined in the vertical direction. Provided the broadenings of both the exciton and photon states are small compared to their characteristic interaction energy, Ω (see Fig. 1b, the vacuum Rabi splitting $\Omega \approx 6$ meV for the sample investigated), the strong coupling limit^[2] is achieved where new quasi-particles arise, termed cavity (exciton)–polaritons. As a result of the coupling, pronounced anti-crossing of the exciton and cavity mode dispersions is observed leading to the formation of new polariton branches with dispersion relations possessed by neither photons nor excitons alone (Fig. 1b). Most notably the lower polariton branch exhibits a dispersion that is photon-like at small wavevector and exciton-like at large wavevector, with a point of inflection in the dispersion between these two extremes, as shown in Figure 1b. It is this dispersion which leads to much of the new physics reported in the present paper. It gives rise to a trap

for polaritons as depicted in Figure 1c (a trap in k -space as opposed to the real-space traps employed in atomic physics to study condensation phenomena), whose depth is controllable by changing the detuning Δ , i.e., the energy separation between the uncoupled exciton and photon modes.

Although the constituent electron–hole components of excitons have fermionic character, excitons have integer spin and are expected to exhibit bosonic properties. Since photons are bosons, exciton–photon coupled modes (polaritons) will also exhibit bosonic properties. For bosonic particles large populations of individual states are allowed by their fundamental symmetry properties^[3] (as opposed to fermions which must obey the Pauli exclusion principle). Since the rate for any quantum mechanical transition is proportional to $(1 + N_{\text{final}})$, where the 1 describes spontaneous processes, and N_{final} describes stimulation of the transition by occupation of the final state, bosonic particles exhibit the property of stimulation of transitions by final state occupation. Such stimulation underlies the operation of lasers, where photon emission is stimulated by macroscopic occupation of the photon modes of the cavity. In this work we demonstrate stimulation of the scattering of polaritons, which in turn gives rise to the new phenomena we describe. Such stimulation could, in principle, occur also for excitons. However, the polariton mass is $\approx 10^{-4}$ greater than that for excitons,^[1] and hence the polariton density of states is very small compared to that for excitons. As a result, it is very much easier to achieve macroscopic state occupancies for polaritons than for excitons, at total densities well below the screening limit where excitons ionise into electron–hole pairs and hence no longer exhibit bosonic properties. As will be discussed in the present paper, specific resonant excitation conditions^[4–9] facilitate very greatly the efficient population of low energy states in the trap necessary to achieve large N_{final} .

Remarkably, the unusual shape of the lower polariton dispersion (Figs. 1b,c) permits new energy and momentum conserving polariton–polariton scattering processes, not possible for either excitons or photons alone.^[4–8] Such processes can be initiated when resonant excitation is employed close to the point of inflection of the lower polariton (LP) branch, as shown in Figure 1c. In this regime the structure acts like an optical parametric oscillator^[6] with efficient conversion of the laser photons into macroscopic populations of two new polariton modes (see Fig. 1c). Such an excitation geometry enables the observation of bosonic stimulation effects and the creation of polariton condensates.

2. Experimental Techniques

Very importantly, the photons and hence the polaritons have a finite lifetime in the cavity, and therefore their population can be probed directly in well-controlled photoluminescence (PL) measurements.^[1] Since the in-plane wavevector k is directly related to the external angle θ by $k = (\omega/c)\sin(\theta)$,

the distribution of the polariton population can be studied directly in measurements of the PL signal at different angles to the sample normal, as shown in Figure 1a. Furthermore, the polaritons can be injected at specific points of the dispersion by varying the angle of incidence of the tunable laser, and the polariton occupation simultaneously probed by varying the detection angle. The experiments were carried out in cryostats with wide angular access in both excitation and detection channels, with angular resolutions in both cases of 1° . A tunable continuous wave (CW) Ti-sapphire laser was used for excitation. The PL was collected by a fiber mounted on a rotating rail and then detected with a high resolution monochromator and nitrogen cooled charge-coupled device (CCD).

3. Non-resonant Excitation

The emission phenomena under non-resonant excitation conditions are first described, to set the scene before moving to the main part of the paper. Under non-resonant conditions at an energy of 1.56 eV, photocreated excitons first relax rapidly by longitudinal-optical (LO) phonon emission (<1 ps), followed by slower (~ 1 ns) acoustic phonon relaxation to the high density of high k exciton states (the exciton reservoir), as shown schematically on Figure 1b. The excitons then relax from the reservoir into the region of strongly coupled polariton states, as also indicated on Figure 1b. However, the increasing escape rate (~ 1 ps $^{-1}$) from the cavity as the LP states become increasingly photon like with decreasing k leads to very inefficient filling of the states at the bottom of the polariton trap. This is shown by the results in Figure 2a where the integrated intensity of the angle-resolved PL for a range of excitation densities is presented. At densities less than 20 W/cm 2 the PL signal from the LP branch peaks at $\sim 18^\circ$, with a marked depletion at smaller angle. This is a signature of the relaxation bottleneck for polaritons, first discussed for bulk materials,^[10] and then for microcavity polaritons by Tassone and co-workers,^[11] before its recent definitive observation for III-V^[12] and II-VI^[13] microcavities. For the higher power of 80 W/cm 2 the PL intensity by contrast peaks at 0° and then decreases smoothly to higher angle, showing that the bottleneck is suppressed at higher exciton densities. The emission at $k = 0$ shows a marked super-linear increase with increasing laser power (Fig. 2b). These results can be explained purely in terms of exciton-exciton pair scattering without any contribution from final state stimulation.^[12,14]

Further increase of power beyond those in Figure 2a leads to a rapid loss of strong coupling, accompanied by a marked blue shift of the emission to the energy position of the uncoupled cavity mode. The loss of strong coupling due to exciton screening at exciton reservoir densities^[15] of $\sim 2 \times 10^{11}$ cm $^{-2}$ is followed by conventional (photon) lasing in the system, as shown in Figure 2b. The results of Tartakovskii et al.,^[12] supported by Senellart et al.,^[14] show that some pre-

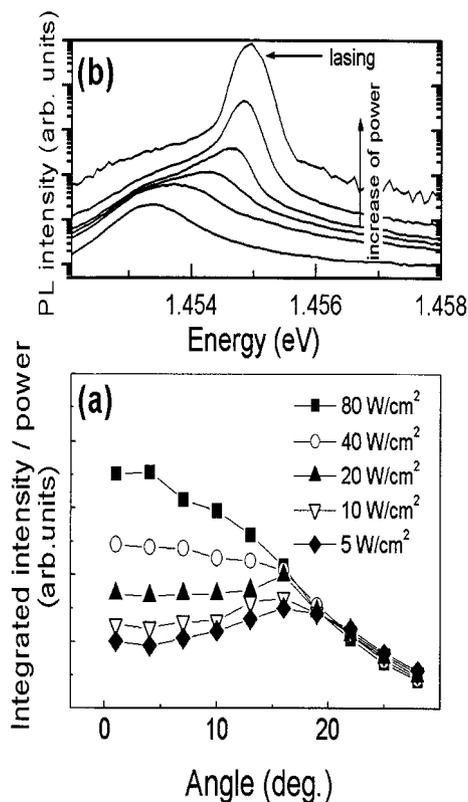


Fig. 2. a) Angular distribution of polariton PL measured for a range of non-resonant excitation powers at $T = 34$ K. b) Non-resonantly excited PL spectra recorded over a wide range of powers. Lasing is observed at the highest powers, blue-shifted by ~ 2 meV relative to the low power PL maximum.

vious claims^[16,17] of stimulated polariton scattering (boson behavior),^[17,18] under conditions of non-resonant excitation were premature. We return to this point below.

4. Resonant Excitation

Very different, striking behavior is observed when resonant excitation into the lower polariton branch is employed. In this case polaritons are injected into the polariton trap without direct population of the exciton reservoir. Clear evidence for final state stimulation is obtained with the system remaining in the strong coupling regime. Strong line narrowing is found to accompany very strong, superlinear (near exponential) increases in $k = 0$ intensity, characteristic of a process with gain, stimulated by transitions to a final state with macroscopic occupancy.

The experiments are performed in the geometry shown schematically in Figures 1a and c and discussed in the introduction. Figure 3a presents $\Theta = 0^\circ$ PL spectra with the laser incident at 16° and tuned into resonance with the LP state. For excitation powers $P < 100$ W/cm 2 the PL spectrum (0.5 meV linewidth) is very similar to that in Figure 2b (at low power). However for $P > 100$ W/cm 2 the form of the spectrum changes dramatically and a new, strongly nonlinear very narrow peak (linewidth < 0.07 meV) appears about 0.3 meV

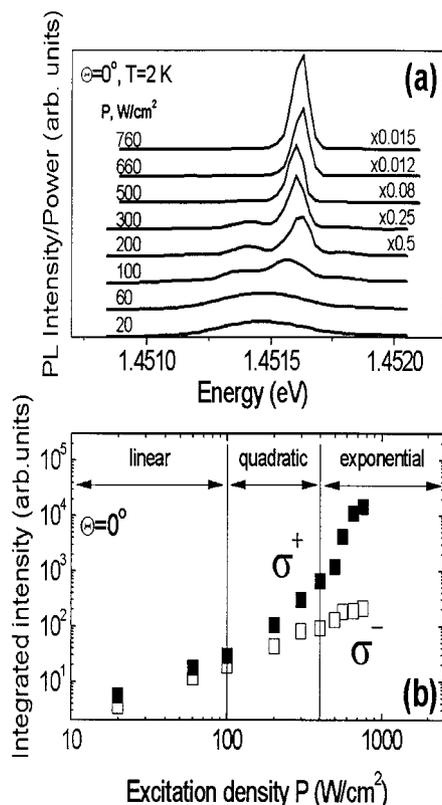


Fig. 3. a) σ^+ -polarized $\theta = 0^\circ$ emission recorded for a range of powers of σ^+ resonant excitation at the point of inflection of the lower polariton branch, as depicted in Figure 1c. b) Integrated intensity of $\theta = 0^\circ$ emission for both σ^+ and σ^- polarizations versus excitation power. Three distinct regimes are clearly seen in the behavior of the σ^+ component.

above the low power PL peak. The small blue shift is negligible by comparison with the magnitude of the Rabi splitting Ω demonstrating the persistence of strong coupling in the nonlinear regime.

Remarkably, if circularly polarized (σ^\pm) excitation is employed, the nonlinearities are found only for emission co-polarized with the laser photons. This is clearly seen in Figure 3b where the power dependence of the PL intensities for σ^+ (solid symbols) and σ^- (open symbols) polarizations are shown. At low power the intensities in both polarizations are similar indicating rapid spin flip relative to the polariton energy relaxation time. With increasing power the σ^+ emission grows very rapidly showing an exponential increase with power above a threshold of $400\text{ W}/\text{cm}^2$. The σ^- emission by contrast remains in the linear regime resulting in nearly complete (>98 %) circular polarization of the $\theta = 0^\circ$ LP emission.

The following three factors clearly indicate the stimulated character of the process observed: i) the strong (nearly exponential) nonlinearity, ii) the occurrence of the nonlinearity for only one spin-state, iii) drastic line narrowing. The first two observations are only possible if the filling rate of the $k = 0$ state is proportional to its occupation factor. The line narrowing corresponds to a long coherence time, which can only arise for a macroscopically occupied mode of the dense polariton system. Indeed the occupancy of the $k = 0$ polariton mode has

been estimated to be close to unity at the threshold of exponential growth in Figure 3b, characteristic of a bosonically stimulated process, reaching as high as 100 at the highest powers employed.^[4]

In the experiments above the angle of excitation is at the point of inflection of the lower polariton dispersion curve (Fig. 1c). Excitation at this angle permits parametric pair scattering of the pump polaritons (Fig. 1c) with conservation of energy and momentum. Clear evidence for such processes is obtained from angular dependent studies. As shown in Figure 4a by the open symbols, at low power the emission peaks at an energy and k -vector around those of the laser excitation, and decreases towards lower and higher k states.

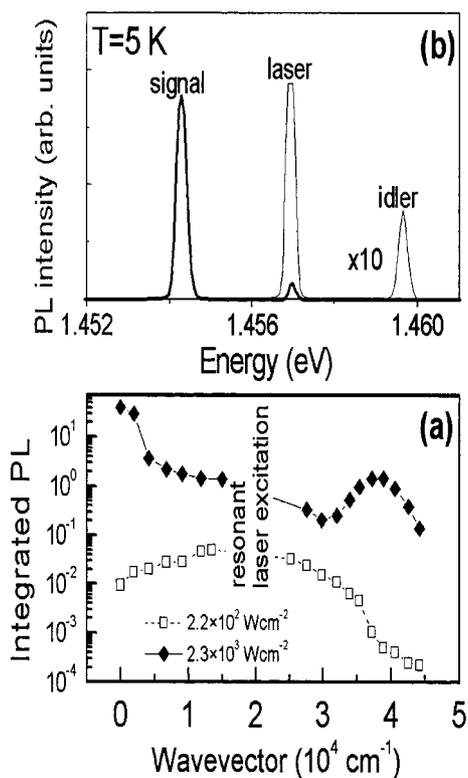


Fig. 4. a) Distribution in k -space of integrated PL intensity at low and high excitation power for resonant excitation (open and solid symbols, respectively). b) Emission spectra detected at $\theta = 0^\circ$ (thick line) and 33° (thin line) for high resonant excitation powers above threshold. The appearance of the “signal” peak is accompanied by strong “idler” emission.

The situation changes fundamentally with increasing power (solid symbols in Fig. 4a). Two strong maxima appear in the emission pattern at $k = 0$ and $2k_{\text{laser}}$, where k_{laser} is the in-plane k -vector of the laser photons. Furthermore a very marked asymmetry in the emission is observed in the angular distribution^[7] (not shown in Fig. 4). The peak in the polariton distribution at $2k_{\text{laser}}$ occurs only for $k > 0$, while the distribution for $k < 0$ remains similar to that at low power. Marked changes also occur in the spectrum at $k = 2k_{\text{laser}}$, with a sharp emission peak observed above the same threshold found for the $k = 0$ peak. Spectra above threshold are shown in Figure 4b, detected at $\theta = 0^\circ$ and 33° (thick and thin lines, respectively). The peak at 1.457 eV (marked “laser”) arises from scattered

laser light, and is situated exactly at the midpoint in energy between the two sharp emission peaks at $\Theta = 0^\circ$ (“signal”) and $\Theta = 3^\circ$ (“idler”), confirming energy conservation in the scattering process.

The angular dependent measurements show that the strong nonlinear behavior observed for resonant excitation has a very different origin to that for non-resonant excitation, and that it arises from a polariton–polariton scattering process in the strong coupling regime to states at $k = 0$ and high k . The scattering process to two specific points in k -space has many of the characteristics of a polariton condensation,^[4,19] to a final state with macroscopic occupancy (greater than 100 at the highest powers employed).^[4] Most notably, the specific form of the polariton dispersion curve shown in Figure 1c permits stimulated energy and momentum conserving pair scattering of the pump polaritons to $k = 0$ and $2k_{\text{las}}$, and hence highly efficient population of the $k = 0$ state.

The stimulated scattering was first reported in two beam pump-probe experiments.^[5,20] In this case a strong pump pulse (1 ps) was incident on the sample at the point of inflection of the LP branch. Only weak nonlinearity of the $k = 0$ emission was found for excitation by the pump pulse alone due to fast escape of the photocreated polaritons before sizeable $k = 0$ populations were able to build up. Injection of a second weak probe at $k = 0$ was found to lead to stimulated scattering from k_{las} to $k = 0$ with very large gains being found. In the CW excitation experiments described here, by contrast the process is self-stimulated, with sufficient $k = 0$ population building up to lead to strong stimulation without the need for injection of a second probe pulse.^[21]

5. Discussion

The results above show that carrier–carrier (pair) scattering processes (exciton–exciton,^[12,14,22] exciton–polariton, and polariton–polariton^[4–9]) play a determining role in the polariton distributions which result from both non-resonant and resonant excitation of semiconductor microcavities. For non-resonant excitation most of the photocreated carriers relax into the exciton reservoir with inefficient population of the small k states where exciton–photon coupling occurs. Attempts to further increase the low k polariton population in the present structures, and hence to achieve stimulation, lead to even greater densities of excitons in the reservoir and eventually to exciton screening.

By contrast for resonant excitation, polaritons are injected directly into the trap formed by the strongly coupled states, with negligible population of the exciton reservoir at low T ; stimulated scattering of the bosonic polariton particles to the $k = 0$ states is achieved whilst remaining firmly in the strong coupling regime. The polariton state has macroscopic coherence, as indicated by its very narrow linewidth, and emits photons at a rate controlled by the photon lifetime in the cavity, determined by the finesse of the Fabry–Perot cavity. Such stimulated scattering followed by photon escape has the

potential to lead to a new form of light source, which we term a polariton “laser”.

In order to achieve polariton lasing under conditions of non-resonant excitation it is necessary that $k = 0$ occupancies greater than one be achieved before strong coupling is lost due to exciton screening from the high density of excitons inevitably created in the exciton reservoir. From the results above it is clear that this situation cannot be achieved in GaAs III–V structures, due mainly to the low densities for exciton screening determined by the relatively small exciton binding energy (8 meV). II–VI,^[23] GaN, and organic^[24] microcavities all offer potentially more favorable systems for the achievement of polariton stimulation under non-resonant conditions, due to their significantly greater exciton binding energies. However, the larger oscillator strengths of exciton transition in these materials lead to much greater (by up to a factor of 10) splittings between the polariton branches, which in turn may lead to less efficient scattering to the low k polariton states, due to the large energy loss required to populate these states from the reservoir. Nevertheless it should be noted that early experiments on II–VI microcavities did provide possible evidence for polariton lasing before collapse of the system to the weak coupling regime,^[23] thus giving optimism that such a polariton laser can be achieved in systems with large exciton binding energy.

The resonantly excited microcavities may also have considerable potential as highly efficient, triply resonant parametric oscillators^[6] and ultrafast switches driven by weak switching pulses at normal incidence.^[5] For these applications structures with large polariton branch splittings are also likely to be highly advantageous. Such devices will operate at room-temperature^[24] and can produce very large energy shifts between laser and signal of up to 5 % of the pump energy, with conversion efficiencies in the range of 10 to 20 % of the power coupled into the structure.

6. Conclusions

We have shown that a host of new phenomena are observable in semiconductor microcavities. These new phenomena arise from the bosonic nature of the polariton quasi-particles and from the ability to manipulate the dispersions of the polaritons by structure design. By contrast with a photon laser where the photon emission process is stimulated, in the present case polariton scattering to a macroscopically occupied coherent mode is stimulated, from which coherent photons are emitted by leakage through the Bragg mirrors. Such structures have potential to lead to highly efficient light sources where the emitting state is populated very efficiently and rapidly by stimulated scattering, and to highly efficient optical parametric oscillators.

[1] For reviews see M. S. Skolnick, T. A. Fisher, D. M. Whittaker, *Semicond. Sci. Technol.* **1998**, *13*, 645. G. Khitrova, H. M. Gibbs, F. Jahnke, M. Kira, S. W. Koch, *Rev. Mod. Phys.* **1999**, *71*, 1591.

- [2] C. Weisbuch, M. Nishioka, A. Ishikawa, Y. Arakawa, *Phys. Rev. Lett.* **1992**, *69*, 3314.
- [3] Such macroscopic occupations for bosons underly the phenomenon of Bose–Einstein condensation, and of photon stimulation in a laser.
- [4] R. M. Stevenson, V. N. Astratov, M. S. Skolnick, M. Emam-Ismail, A. I. Tartakovskii, D. M. Whittaker, P. G. Savvidis, J. J. Baumberg, J. S. Roberts, *Phys. Rev. Lett.* **2000**, *85*, 3680.
- [5] P. G. Savvidis, J. J. Baumberg, R. M. Stevenson, D. M. Whittaker, M. S. Skolnick, J. S. Roberts, *Phys. Rev. Lett.* **2000**, *84*, 1547.
- [6] J. J. Baumberg, P. G. Savvidis, R. M. Stevenson, M. S. Skolnick, A. I. Tartakovskii, D. M. Whittaker, J. S. Roberts, *Phys. Rev. B* **2000**, *62*, R16247.
- [7] P. G. Savvidis, J. J. Baumberg, R. M. Stevenson, M. S. Skolnick, D. M. Whittaker, J. S. Roberts, *Phys. Rev. B* **2000**, *62*, R13278.
- [8] A. I. Tartakovskii, D. N. Krizhanovskii, V. D. Kulakovskii, *Phys. Rev. B* **2000**, *62*, R13298.
- [9] R. Houdré, C. Weisbuch, R. P. Stanley, U. Oesterle, M. Ilegems, *Phys. Rev. Lett.* **2000**, *85*, 2793.
- [10] J. Toyozawa, *Suppl. Prog. Theor. Phys.* **1959**, *12*, 11.
- [11] F. Tassone, C. Piermarocchi, V. Savona, A. Quattropani, P. Schwendimann, *Phys. Rev. B* **1997**, *56*, 7554. F. Tassone, Y. Yamamoto, *Phys. Rev. B* **1999**, *59*, 10830.
- [12] A. I. Tartakovskii, M. Emam-Ismail, R. M. Stevenson, M. S. Skolnick, V. N. Astratov, D. M. Whittaker, J. J. Baumberg, J. S. Roberts, *Phys. Rev. B* **2000**, *62*, R2283.
- [13] M. Müller, J. Bleuse, R. André, *Phys. Rev. B* **2000**, *62*, 16886.
- [14] P. Senellart, J. Bloch, B. Sermage, J. Y. Marzin, *Phys. Rev. B* **2000**, *62*, R16263.
- [15] R. Butté, G. Dellaleau, A. I. Tartakovskii, M. S. Skolnick, V. N. Astratov, J. J. Baumberg, G. Malpuech, A. di Carlo, A. V. Kavokin, J. S. Roberts, unpublished.
- [16] P. Senellart, J. Bloch, *Phys. Rev. Lett.* **1999**, *82*, 1233.
- [17] H. Cao, S. Pau, J. M. Jacobson, G. Björk, Y. Yamamoto, A. Imamoglu, *Phys. Rev. A* **1997**, *55*, 4632.
- [18] A. Imamoglu, R. J. Ram, *Phys. Lett. A* **1996**, *214*, 193.
- [19] The similarities and differences between the polariton condensation by stimulated scattering and Bose–Einstein condensation where the condensation results from cooling of the system is discussed in ref [4].
- [20] Theoretical studies of the parametric scattering are reported in C. Ciuti, P. Schwendimann, A. Quattropani, *Phys. Rev. B* **2001**, *63*, 041303. D. M. Whittaker, *Phys. Rev. B* **2001**, *63*, 193305.
- [21] In a very recent paper by J. Erland, V. Mizeikis, W. Langbein, J. R. Jensen, J. M. Hvam, *Phys. Rev. Lett.* **2001**, *86*, 5791 on very high quality samples with photon lifetimes of ~10 psp, stimulation in ultrafast measurements was observed in the presence of the pump alone.
- [22] R. Huang, F. Tassone, Y. Yamamoto, *Phys. Rev. B* **2000**, *61*, R7854.
- [23] L. S. Dang, D. Heger, R. André, F. Boeuf, R. Romestain, *Phys. Rev. Lett.* **1998**, *81*, 3920.
- [24] D. G. Lidzey, D. D. C. Bradley, T. Virgili, A. Armitage, M. S. Skolnick, S. Walker, *Phys. Rev. Lett.* **1999**, *82*, 3316.