

Towards a room-temperature polariton amplifier

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Received 4 November 2002, in final form 11 May 2003

Published 3 September 2003

Online at stacks.iop.org/SST/18/S325

Abstract

Microcavity exciton polaritons, the fundamental optical excitations of semiconductor microcavities with quantum wells inside, have been proposed as promising candidates for observing stimulated scattering, condensation and other phenomena related to the bosonic nature of excitons. Having a light mass, quantum degeneracy of polaritons can be reached at low densities and high temperatures. But the radiative time of polaritons is very short (in the picosecond range) and usually prevents an efficient thermalization and cooling of the excited cloud of polaritons. A ‘coherently driven condensate’, not corresponding to a thermal equilibrium, but featuring multiple occupation of single-particle states, can however be created by an external laser source resonantly exciting polaritons. Under this condition, stimulated parametric scattering of polaritons can provide huge optical gain on a weak probe pulse shined on the sample. In this work we demonstrate that this phenomenon can survive at temperatures close to room temperature and could be achieved in the future even above this limit. Clever sample designs favour the thermal robustness of polariton parametric amplification, but from the experimental data it turns out that the parameter that ultimately limits the highest temperature for polariton parametric scattering is the exciton binding energy.

1. Introduction: polariton dispersion and parametric amplification

In a gas of particles at low temperature and high density, the wavefunctions of individual particles start to significantly overlap, giving rise to non-classical collective states. In these states the particles lower their total energy through phase transitions that develop a macroscopic order. Such collective states are responsible for phenomena such as superconductivity and Bose condensation. It is a long-lasting goal of semiconductor optics to investigate collective states developed by a low-temperature gas of excitons, as the ‘exciton condensation’ could produce an optically active medium with exceptional properties, such as extremely low lasing threshold, production of squeezed light and negligible dissipation [1–6]. But excitons can be considered bosons only at low densities, while at high densities the fermionic character of the electrons and holes that compose excitons dominates and prevents the exciton condensation. On the other side, one has to raise the exciton density to achieve the condensation and it turns out that at temperatures of the order of 1 K the density necessary

to achieve quantum degeneracy is of the same order of the density at which the bosonic approximation is no more valid.

One of the approaches that have been proposed to ease the exciton condensation is to place the excitons inside a semiconductor microcavity [7, 8]. Semiconductor microcavities are realized by sandwiching the optically active semiconductor layer (a quantum well in our case) between two high-reflectivity mirrors. The energy of the cavity resonance is determined by the distance between the two mirrors. If the cavity is chosen to be energy-resonant with the exciton transition of the quantum well, the radiative decay of excitons is strongly enhanced and can become the main scattering channel. This regime is called ‘strong-coupling’ regime and is characterized by the fact that the dipole coupling energy between exciton and photon is larger than both the exciton and cavity-photon linewidths.

In the strong-coupling regime, the normal modes of the optical excitations of the microcavity are no more excitons and photons, but quantum mixtures of these two states, called cavity polaritons [9]. This can be seen as the counterpart of the oscillations in the time domain between exciton and

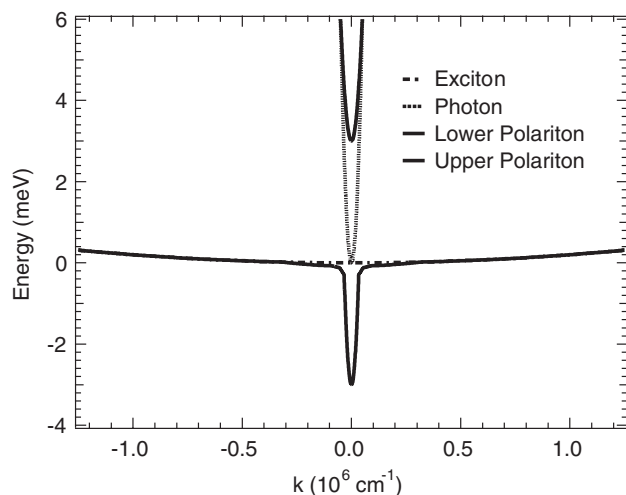


Figure 1. Sketch of the dispersion curve of exciton, cavity photon and polaritons, i.e. the energy of the modes as a function of the momentum in the cavity plane (in-plane momentum).

photon. A major consequence of the strong coupling is that the energy degeneracy between the two normal modes is lifted by the exciton–photon dipole coupling, and the polaritons are separated in two branches by an energy splitting and are called upper and lower polaritons.

The energy of the cavity photon mode changes with the angle of light propagation (with respect to the normal incidence), modifying also the strength of the exciton–photon coupling. Therefore the lower polariton energy dispersion as a function of the in-plane momentum k has a non-trivial shape (see figure 1). For k -vectors larger than the photon momentum, the dispersion curve is the smooth excitonic one, while for the radiative region, where the momentum is about equal to or smaller than the photon momentum, the energy dispersion abruptly becomes very steep. This implies that polaritons have a very light effective mass, some 10^4 – 10^5 times smaller than the exciton mass. The mass being very low, polaritons have a much larger spatial extent (or de Broglie wavelength) than excitons. This is important for quantum degeneracy, which is achieved when the inter-particle spacing is comparable to the de Broglie wavelength. In the k -space, the low mass corresponds to a low density of states per energy unit. For polaritons, the quantum degeneracy can be achieved for excitation densities 4–5 orders of magnitude lower than for excitons. In other words, the polariton density of states is very low compared to the exciton one and few polaritons per unit area are enough to occupy the available states. The effects related to the Pauli exclusion principle grow as the excitation density and it turns out that they are negligible at the density necessary to reach the quantum degeneracy of polaritons, meaning that polaritons reach the quantum degeneracy in a regime where they can be still considered as interacting bosons. If the density is kept constant, a light mass favours the degeneracy and condensation at higher temperatures.

However, it is not easy to create a quantum-degenerate gas of polaritons by letting them cool to the lattice temperature after a non-resonant optical excitation. The main obstacle is an intrinsic characteristics of the polariton states, the fast radiative decay, due to the hybridization between exciton and

photons. The radiative decay is typically of the order of a picosecond. This time is fast compared to the time necessary to populate the polariton states starting from a reservoir of free carriers or excitons, meaning that polariton states are emptied more rapidly than they can be occupied, and a degenerate distribution in thermal equilibrium is not likely to be achieved.

A different approach to study the non-classical properties of polaritons is to resonantly inject a dense and non-thermal polariton gas in the microcavity by shining a laser beam whose wavelength is resonant with the polariton absorption. Gas of polaritons can be resonantly created featuring macroscopic coherence and multiple occupation of single-particle states. These properties being similar to those of a spontaneous condensate, we can consider that a ‘coherently driven condensate’ is created by laser excitation. One should be aware of the fundamental differences between this ‘coherent condensate’ and a spontaneous Bose condensate, where the coherence is generated by a spontaneous symmetry breaking and is not injected by an external source.

The polariton dispersion curve can be accessed by tuning the incidence angle and the energy of the exciting laser beam, as the component of the photon momentum parallel to the plane of the cavity is transmitted to the polaritons excited inside the cavity. Due to the short polariton lifetime, the dynamics of polaritons has to be studied with ultrafast spectroscopy techniques, usually by exciting the microcavity with pulses from a mode-locked titanium:sapphire laser. In the typical experimental configuration, a pump pulse excites the sample at a variable angle and a weak probe pulse detects the stimulated dynamics at the polariton band bottom, i.e. at normal incidence. To change the excitation angle of the pump beam without affecting the delay between pump and probe pulses, we employed a mechanical goniometer [10]. The energy of the polaritons changes with the angle according to the polariton dispersion. Therefore the energy of pump polaritons does not coincide with the energy of probe polaritons at the band bottom. The contemporaneous excitation of both states can be done with a pulsed laser whose spectral width is larger than half the polariton splitting. To selectively excite the polaritons on the lower branch, the pump spectrum can be tailored to match the polariton absorption line.

Detecting the reflection of the probe pulse, one can monitor the effects induced by the pump pulse as a function of its excitation angle. For a particular pump angle, a sharp line appears in the probe reflection spectrum featuring optical gain, much more intense than the incident probe, as shown in figure 2. The amplification of the reflection corresponds to a net gain on the probe [10]. The amplification has a sharp angular resonance and occurs only for a particular angle: if the pump excites the sample few degrees away from the angle of maximum gain, the gain is already completely suppressed. The angular resonance is related to the parametric nature of the gain [11, 12]. To satisfy energy and momentum conservation, the pump polaritons have to scatter in pairs. One polariton scatters into the probe state, thus loses energy and momentum, while the other one takes away the difference in energy and momentum and generates emission of light at a larger angle, as shown in figure 3. The angular resonance is due to the fact that only at the inflection point of the polariton dispersion it is possible to conserve exactly both energy and

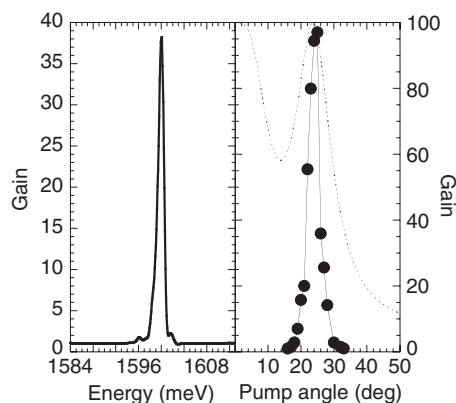


Figure 2. *Left panel:* Reflection spectrum of the probe beam in presence of the pump incident at an angle of 23° . A large nonlinear amplification is apparent. The measurement is taken at 77 K. *Right panel:* Angular resonance of the amplification (the pump density is higher than that in panel (a), so that the peak values of the gain cannot be compared with what observed in the spectrum). The dashed line is the function $F(k_p) = 2E_{k_p} - E_0 - E_{2k_p}$ representing the energy difference between two pump polaritons and the sum of one signal and one idler as a function of the pump angle, as explained in the text. The maximum gain occurs near the angle where the phase matching is optimum.

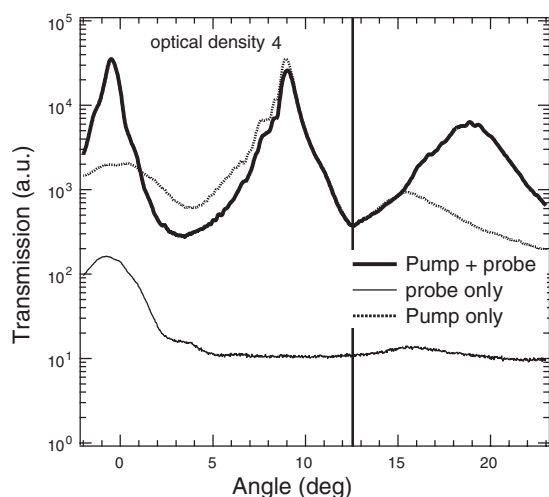


Figure 3. Emission of the microcavity on the substrate side (opposite to the excitation side) as a function of the emission angle (the detection is spectrally integrated). The photon component of the polaritons is low at large angles, therefore the emission intensity is reduced for growing angles. The emission intensity at the pump angle is comparable to the emission at zero angle, even if the number of pump polaritons is larger than the number of polaritons at $k = 0$. In order to see the idler branch on the same intensity scale as pump and signal, we had to attenuate by four orders of magnitude the emission at angles smaller than 12.5° . In this measurement the probe is not exactly at normal incidence, but at a small negative angle.

momentum for the pair scattering. If the pump angle does not correspond to this inflection point, no high-energy state is available for the pair scattering and the amplification is suppressed. Usually, one calls the polaritons at zero angle the signal branch, those with higher energy the idler branch, in analogy with the optical parametric amplifier.

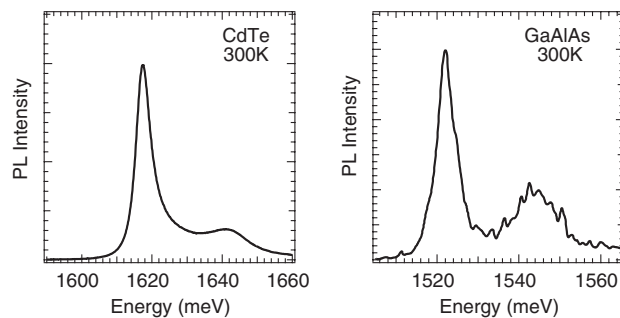


Figure 4. Room-temperature photoluminescence spectra for two large-splitting samples. The GaAlAs sample is the one with 36 quantum wells (see the text for the details of the sample structure). The samples are excited in the absorption continuum with a weak cw laser. The intensity ratio between the two polaritons is determined not only by the detuning but also by the energy difference between the two branches, which makes the lower polariton more populated than the upper one at thermal equilibrium.

2. High-temperature polariton amplification

The optical amplification arising from polariton parametric scattering is very efficient and immediately after its discovery [10] attracted the attention of a large community of researchers [13–20]. A fundamental issue is to establish under which physical condition the parametric amplification can take place and what are the effects of temperature on it. It is intuitive that by making the polariton trap deeper the amplification should be stable at higher temperatures. Therefore to obtain the same amplification at higher temperatures (i.e. with a broader polariton line) one needs a higher polariton splitting. The idea is to substantially increase the number of quantum wells and to put some of them also in the first few layers of the Bragg mirrors, where the electric field is almost as intense as in the cavity [21]. In this way, the number of quantum wells can be increased without diminishing the electric field acting on them. With this arrangement, a sample with 36 GaAs quantum wells has a 20 meV polariton splitting. A similar sample with only 12 quantum wells gave a polariton splitting as large as 15.3 meV. An alternative way of increasing the polariton splitting with a standard cavity design is to use a semiconductor material with a larger exciton oscillator strength, e.g., a II–VI compound. Among the II–VI compounds, CdTe has not the highest exciton oscillator strength, but high-quality CdTe microcavities can be grown with well-established growth processes. The sample we investigated is a 3λ cavity containing six stacks of four quantum wells each. The polariton splitting is as high as 25 meV.

All three mentioned samples are in the strong-coupling regime up to room temperature, meaning that the coupling of excitons with light is larger than the coupling with phonons at room temperature. This feature confirms that these samples are well suited for the investigation of high-temperature polariton amplification. Typical photoluminescence spectra taken at room temperature are shown in figure 4: the two peaks corresponding to the two polariton energies are well resolved, but the linewidth of the peaks is not negligible in comparison to the splitting.

The first remarkable consequence of the large polariton splitting is a very high parametric amplification [22]. All

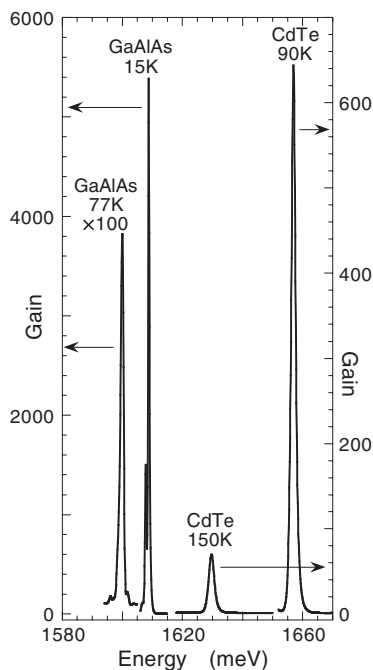


Figure 5. Gain spectra for different temperatures in the GaAs (12 wells) and CdTe-based samples. When temperature increases, the polariton mode gets broader and consequently the gain peak gets smoother, broader and lower. Pump photon densities incident on the sample are typically of the order of 10^{13} photons (cm^2 pulse) (but the polariton densities created are much lower).

three samples show at low temperature unprecedented light amplification. The highest recorded gain value is more than 5000, for the GaAs sample with 12 wells at temperatures lower than 15 K, as shown in figure 5. This performance represents an improvement by almost two orders of magnitude with respect to the amplification initially reported [10]. It is obtained under resonant excitation with 250 fs laser pulses.

The distinctive features of parametric amplification have been checked in all three samples. The gain always peaks at an energy slightly higher than that of the linear polariton absorption. The gain energy is anyway far away from that of the empty-cavity mode, a condition ensuring that the gain occurs in the strong coupling regime. The gain has an angular resonance, and the ‘magic’ angle, different in each sample, always corresponds to the excitation of polaritons at the inflection point of the polariton dispersion. The gain dependence on the pump power has a very sharp threshold.

It has to be realized that this huge light amplification takes place in a very thin device. Including all Bragg mirrors the microcavity is only about $5 \mu\text{m}$ thick. Why is the gain so high in the high-splitting samples? With growing pump densities the lower polariton branch is broadened in energy by the excitation-induced dephasing and is shifted towards the blue by the repulsive polariton–polariton interaction. The same polariton–polariton interaction that generates the amplification, at higher densities saturates the oscillator strength of the exciton transition, causes the break-down of the strong-coupling regime and limits the maximum pump excitation density and therefore the maximum parametric amplification. A possible explanation of the giant amplification is therefore that the larger is the polariton

splitting the higher is the pump density needed to break the strong-coupling regime and the less important is the impact of broadening of the polariton line induced by the pump excitation. Recently, a quantitative many-body theory has been developed to explain why the large splitting increases so much the maximum gain [23]. The polariton–polariton interaction is treated beyond the mean-field approximation by including four-particle terms. The many-body terms essentially provide an additional dephasing that limits the gain. Owing to the particular shape of the polariton dispersion, not only the scattering between polaritons and phonons or polaritons and disorder are inhibited, but also the polariton–polariton scattering is suppressed, except for the scattering channel leading to the parametric amplification. The larger the splitting, the more efficient is the suppression of all the scattering channels competing with the parametric one.

Figure 6 shows the parametric gain versus temperature. Even if the gain decays rapidly with temperature, the large polariton splitting proves being extremely beneficial also for the high-temperature operation. In the CdTe sample, having the largest polariton splitting among the investigated samples, gain can be measured up to 220 K. Also the value measured in the GaAs samples is remarkable, as amplification can be obtained up to 125 K, thus above the liquid nitrogen temperature. As a comparison, figure 6 shows the temperature dependence of the gain in a low-splitting InGaAs sample containing one quantum well only. This sample has an outstanding splitting-to-linewidth ratio, but the polariton splitting is just 3.5 meV. Already at 60 K, the parametric amplification is suppressed.

The large splitting is necessary for the high-temperature operation. Yet it does not seem to be the only parameter which determines the highest temperature at which the parametric process can occur, as in the two GaAs samples, having a different splitting, the gain disappears at the same temperature. Moreover, the CdTe sample has a 25 meV splitting (instead of 20 meV), but shows gain up to an almost double temperature than the GaAs samples. Figure 6 suggests that the other relevant parameter is the exciton binding energy, as in all the samples the gain vanishes at a temperature such that the thermal energy kT (k is Boltzmann’s constant) is of the order of the exciton binding energy (see [22] for more details on this topic).

One could imagine that the hybridization with photons is sufficient to make polaritons ‘condense’, even if the electronic transition associated with polaritons generates fermionic excitations such as electrons and hole. It has been proposed in some theoretical models [24] that polariton condensation can occur in microcavities embedding quantum dots, even if the optical excitation of quantum dots creates perfect fermions. In our case, a scenario originally proposed by Keldysh (see, e.g., the review by Keldysh in [1]) could be possible, where electrons and holes are free particles, but above a certain threshold density undergo a ‘BCS-like’ phase transition, as the Cooper pairing of one electron and one hole is energetically favoured by the appearance of an energy gap whose amplitude is much lower than the exciton binding energy. But the fact that the exciton binding energy plays such a fundamental role in the high-temperature polariton amplification is a clear indication that the phenomenon takes place only if excitons exist as bound

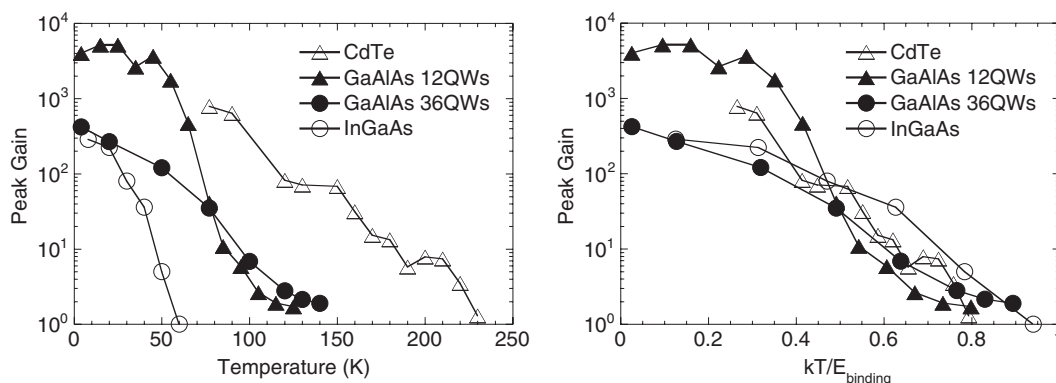


Figure 6. Peak gain (maximum measured gain value) versus temperature for the different samples. In the right panel, the temperature is renormalized by the exciton binding energy ($E_{\text{binding}} \simeq 5, 13, 25$ meV, respectively, for the InGaAs, AlGaAs and CdTe quantum wells).

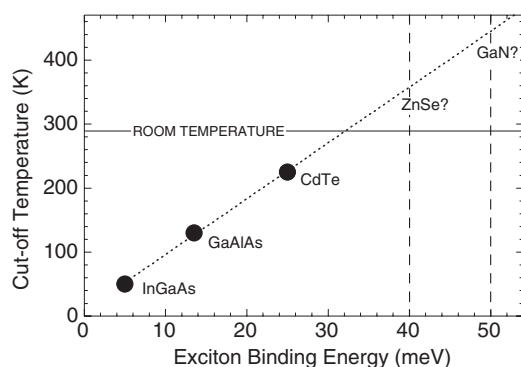


Figure 7. Cut-off temperatures for the gain in different materials as a function of the exciton binding energy (lower x -axis, filled circles). The broken line is a linear fit to the data. The experimental points fit very well with a linear dependence on the exciton binding energy. This dependence does not change significantly if one takes different (but still reasonable!) definitions of cut-off temperature, such as the inverse logarithmic slope of the last points on each curve of gain versus temperature. The vertical dotted lines mark the typical exciton binding energies in ZnSe- and GaN-based quantum wells.

particles. This rules out the possibility of the ‘BCS-like’ scenario, confirming that the interaction between electron and hole dominates and the excitons as bosonic quasi-particles are already formed at densities lower than the critical density for quantum degeneracy.

3. Physical interpretation: Bose condensate or optical parametric amplifier?

Concerning the physical interpretation of the polariton parametric amplification, it is important to establish the exact role of the bosonic statistics. In principle, wave-mixing phenomena just require that the involved waves are coherent, not that the corresponding quantum operators obey boson commutation rules, as also fermions can undergo parametric scattering. In order to decide experimentally whether or not the polariton parametric amplification is related to the bosonic statistics, the number of polaritons in each quantum state during the amplification has to be determined. This can be done by measuring the number of photons coming out in the probe direction within a certain solid angle, e.g., in the case of the highest recorded gain, for the GaAlAs sample at 15 K

(figure 5). The experimental parameters needed for this estimate are the probe-beam angle spread (0.5°) and the probe photon density injected into the cavity (2×10^6 photons cm^2 pulse). This probe density is already sufficient to populate each state around $k = 0$ with approximately 100 polaritons [22] (on how to count the available polariton states see [13]). As the gain peak is approximately two times narrower than the polariton, when the gain is 5000, the light intensity emitted in the probe direction after the parametric amplification is 2500 times that absorbed from the probe. The resulting occupancy on the lowest energy state is of the order of 10^5 polaritons per state. At higher temperatures and in different samples the occupation numbers that can be calculated are somewhat lower, but still much higher than 1. Under cw excitation, occupation numbers higher than 100 were measured [13]. It has to be remarked that the exciton fraction of polaritons in the fundamental state is relevant, typically between 20 and 50%, meaning that during the polariton amplification more than 10^4 excitons occupy the same quantum state. This estimate leads to the conclusion that *even if the parametric scattering itself is not related to the bosonic statistics of polaritons, the efficiency of the amplification is due to the possibility to occupy each quantum state with many polaritons.*

The high occupation number and the stimulated scattering are phenomena analogous to the ones occurring in cold atom gases (see, e.g., [1]). However, even if many polaritons occupy the same quantum state and the threshold in the amplification seems to mark a phase transition, the phenomenon has relevant fundamental differences with respect to the atomic Bose–Einstein condensation. The system is far from a thermal equilibrium, as the polaritons along the branch are created by resonant optical excitation, undergo a coherent stimulated scattering and then re-radiate outside the cavity before they can thermalize. The resonant pumping introduces a coherent reservoir of pump polaritons, whereas in the Bose–Einstein condensation there is a spontaneous symmetry breaking that lowers the total energy of the system and creates the coherence in the condensate out of an incoherent (thermal) atom reservoir. In this sense, the experiments with non-resonant creation of an incoherent exciton reservoir have much more similarities with the Bose condensation [25–28].

A closer analogy can be drawn between the polariton parametric amplification and the atom–wave mixing.

The polariton parametric amplification is a wave-mixing phenomenon half way between the classical optical parametric amplification, involving the photons coupled by the nonlinear susceptibility of a crystal, and the atom-wave mixing [29–31], involving only coherent matter waves. Recent experiments [32] demonstrate the phase coherence of the polariton parametric amplification, i.e. the fact that not only the emission is coherent, but that there is a precise phase relationship between pump, signal and idler. If the ultrafast scattering process could lead to very efficient all-optical amplifiers and switches, the phase coherence of polariton parametric scattering could lead to devices with new capabilities, such as emitters of squeezed light or entangled photons.

4. Conclusions

Polariton parametric scattering in semiconductor microcavities can generate a huge optical amplification on a weak laser beam. The main characteristics of the parametric amplification, ultrafast dynamics, narrow spectrum and angular resonance, originate from the peculiarities of the polariton gas coherently created in the microcavities, which realizes a non-equilibrium and non-thermal ‘coherent boson condensate’ with occupations much higher than one particle per state. In this work we review the experiments demonstrating that the polariton parametric amplification can survive almost up to room temperature (220 K). The same sample design that maximizes the thermal stability of the gain also maximizes the gain efficiency, leading to amplification factors higher than 5000. The key parameter determining the thermal stability of the amplification appears to be the binding energy of the quantum well excitons in the microcavity. As long as the excitons exist as bound and bosonic quasi-particles the parametric amplification is possible.

Acknowledgments

R André, J Bloch, G Bongiovanni, Le Si Dang, A Mura, U Oesterle, V Thierry-Mieg participated in our experimental work, provided us with most of the samples we employed

(GaAlAs samples from Paris and CdTe samples from Grenoble) and discussed with us the results. This work was supported by the Swiss National Science Foundation.

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