An investigation of the parametric amplification and its coherent control in a semiconductor microcavity is presented. The time and angle resolved pump and probe experiments show that several ps after pumping parametric scattering is still phase coherent. The experimental data are in qualitative agreement with the numerical data obtained from a relatively simple theoretical model based on three polarisation components, pump, probe, and idler. This model also predicts that under particular circumstances, the polaritons can scatter back from the signal and idler to the pump.

1. Introduction

Ten years ago the strong coupling regime between quantum well (QW) excitons (Xs) and microcavity (MC) photons has been observed [1]. Since then, Fabry–Perot semiconductor MCs have been intensively investigated [2]. The mixed X–photon states, the polaritons, can be excited optically by external photons sent to the planar structure. It has not been straightforward to understand the linear optical properties of MCs [3, 4]. The MCs were initially strongly affected by disorder in the mirror and QW structures. The latter leads to inhomogeneous X broadening, which of course strongly influences the polariton spectra and in extreme cases even destroys the strong coupling regime [5–7]. Moreover, this disorder causes also Rayleigh scattering in the MC, whereby the part due to the QWs is resonant [8–10]. The spread of the polaritons, after resonant excitation by light with oblique incidence, along a ring in the wavevector plane parallel to the QW, is one of the most spectacular features of resonant Rayleigh scattering [11, 12]. It is directly related to the strong coupling regime and the particularities of the polariton dispersion that ensue. The particular shape of the polariton dispersion is also at the origin of the observed very long polariton dephasing times [13]. Thorough experimental and theoretical investigations were needed to understand the interplay of strong coupling, disorder effects, and the combination of both [14, 15] on the optical properties and polariton dynamics in MCs.

The effects of strong excitation in MCs, which are featuring a very rich phenomenology, can be divided into two groups. The first one concerns excitonic non-linearities...
which in principle could be observed also directly in QWs outside a MC. Since a MC acts as a narrow spectral filter, inside such a device it is possible to excite very selectively only fundamental Xs and to avoid perturbations caused by free electron–hole pairs. In this way, it has been possible e.g. to observe directly the resonant Stark effect of QW Xs [16]. The phenomena of the second group are connected to the strong coupling regime, i.e. polaritonic non-linearities are concerned. They are mainly due to the peculiar properties of the inplane dispersion of MC polaritons. Their effective mass is four orders of magnitude smaller than that of Xs. This property, together with the long dephasing times, makes polaritons good candidates for the observation of bosonic effects [17]. In fact, occupation numbers per mode of up to $10^4$ can easily be reached by selective coherent excitation of a few polariton modes [18, 19]. In other words, quantum degeneracy can be reached while the polariton–polariton interactions are still quite weak and therefore the fermionic character of the (still strongly correlated) electrons and holes, which together with the photons are components of the polaritons, do not perturb.

In this paper, we discuss parametric amplification (PA) in MCs and its coherent control. Huge PA can be observed in a degenerate and coherent polariton system, the amplification factor depends sensitively on sample quality and in particular on the polariton line width. This phenomenon, besides its high scientific interest, appears also to be well suited for optoelectronic applications, such as high-speed switches and amplifiers. First, we present how effective stimulated polariton scattering can be achieved in angle, energy, and time resolved pump and probe experiments. Most of the article is devoted to the coherent control of PA, by laser pulses that are much weaker than the emission they control. The experimental results are compared with theoretical calculations based on a three-component polariton model [20].

2. Parametric Amplification

The exciton–photon interaction is important only for in-plane wave vectors $k$ close to zero. The dispersion of the two polariton branches is sketched in Fig. 1, for the case of perfect resonance between X and photon mode. The lower branch features a narrow valley with a minimum at $k = 0$ and reaches the X energy for wave vectors that are bigger than the reciprocal X radius. Polaritons occupying states in the central valley interact only weakly with other polaritons, excitons, or phonons, because for such processes energy and momentum conservation is difficult to achieve. These polaritons are thus isolated from the other excitations of the cavity and the crystal, and have therefore very long dephasing times [13, 21, 22].

There is an exception to this isolation. Two polaritons occupying the states with $k = k_m$, at an energy about halfway between the bottom and the top edges of the valley, can easily scatter to $k \approx 0$ and $k \approx 2k_m$ since energy and momentum are conserved for this process (Fig. 1). In fact, this process can be stimulated, e.g. by coherently occupying the polariton states at $k \approx 0$. Or, in other words, an MC can work as an optical parametric amplifier, if it is pumped with a laser beam incident at the so called magic angle corresponding to $k = k_m$. By probing at normal incidence ($k \approx 0$, signal) a “magically” excited MC, at liquid He temperature probe gain as high as 5000 has been observed [18, 19], and also the emission at $k \approx 2k_m$ (idler) has been detected. Let us also mention that the dynamics of this PA process seems to be quite fast; the gain decays, with increasing pump–probe delay, with a characteristic time of roughly 1 ps which is much
less than the escape time of the photons from the cavity. We attribute this to a rapid dephasing of the dense pump polariton gas [11, 15]. Due to the above-mentioned isolation effect, the interaction rate of the polaritons remains relatively low also at higher temperatures. Therefore, PA occurs also at higher temperatures, it has been observed up to 220 K in a II–VI MC containing CdTe/Cd(Mg, Te) QWs [19]. A detailed investigation of the temperature dependence in different samples revealed that PA can occur as long as the thermal energy \( k_B T \lesssim E_X \), the X binding energy. This means that it is the thermal dissociation of the Xs that prevents PA from occurring, the presence of free carriers strongly perturbs the Xs and thus also the polaritons. It is expected that a MC containing a material having an X binding energy of say 40 meV should feature PA also at room temperature.

3. Coherent Control of the Parametric Amplification The fastest and most efficient way to manipulate optical excitations in semiconductors is by coherent control [23, 24]. In an experiment using this technique, first an ensemble of excitations having all practically equal energy and phase are created, typically by a short laser pulse. As long as the excitations are not subject to relaxation processes, with subsequent laser pulses a substantial fraction of the excitations can be switched to other states having different properties. The short dephasing times of excitations in semiconductors often represent a serious problem. As mentioned above, MC polaritons in the strong coupling regime have long dephasing times, and are thus particularly suited for doing coherent control experiments [25, 26]. In the following we demonstrate how parametric scattering in a MC can be controlled coherently.

3.1 Experimental The investigated sample is a high-quality λ MC, the spacer layer is made of pure GaAs, at its centre, where the electric field of the fundamental mode is strongest, a single In\(_{0.03}\)Ga\(_{0.97}\)As quantum well, 7.5 nm thick, has been inserted. At liquid He temperatures, the QW X has an energy of 1.487 eV (slightly below the GaAs band gap). The two Bragg mirrors consist of 20 and 26.5 GaAs and Al\(_{0.1}\)Ga\(_{0.9}\)As \(\lambda/4\) layers. At resonance, the upper and lower polariton modes have a spectral width of about 0.3 and 0.13 meV, respectively, while the polariton splitting amounts to about 3.5 meV. Thus, the strong X–photon coupling regime is well established.
The experimental setup is shown in Fig. 2. The pump and probe pulses, which initially have a temporal length of 0.15 ps, are obtained from a Ti:sapphire laser using a beam splitter. The part of the beam used for the pump is sent through an adjustable spectral filter having a bandwidth of about 2 meV. The pulses emerging from this filter, about 1 ps long, are adequate to selectively pump a small number of states on one of the two polariton branches. With the probe pulses, having a spectral width of some 15 meV, all the states that are influenced by the strong X–photon coupling can be reached. The angle of incidence of the pump beam on the sample can be adjusted using a mechanical goniometer, normally it is set to the magic angle of the investigated MC. The probe pulses are divided into two phase-controlled pulses by a Michelson interferometer. The coarse delay can be varied by translating one of the mirrors using a μm screw, the relative phase is tuned using piezoelectric elements, and the required mechanical stability is obtained by monitoring the interferences of a separate cw HeNe laser beam [27]. Our setup permits to control the relative phase of the two pulses with a precision of $\pi/40$. The probe beam is reflected by the sample, dispersed by a spectrometer and detected (time integrated) by a CCD camera. The light leaving the sample from its backside can also be collected by a lens and imaged on a CCD, permitting to measure the angular distribution of the emissions.

Figure 3 shows the emission of the MC from its backside. When only the pump beam excites the sample at the magic angle, “spontaneous” parametric scattering occurs, leading to signal ($k \approx 0$) and idler ($k \approx 2k_m$) emissions. If, contemporaneously with the pump pulses, the MC is probed by single pulses at normal incidence, the polariton states at $k \approx 0$ are coherently occupied which stimulates parametric scattering of the
pump polaritons, therefore, the signal and idler emissions are strongly enhanced. Let us remark that the number of probe photons penetrating into the cavity is much smaller than that of the signal polaritons generated by spontaneous scattering of the pump polaritons (Fig. 3), nevertheless the former have a very strong stimulation effect. We note also that the incidence angles do not exactly match the ideal values, which causes the emissions related to the spontaneous and stimulated processes to occur in slightly different directions. Moreover, the signal peak due to PA is narrower than those due to pump and probe alone.

3.2 Experimental results and features  Figure 4a shows probe reflection spectra obtained with one single probe pulse hitting the sample at the same time as the pump pulse. The pump angle has been chosen to be about $10^\circ$ for maximum PA, and the photon density employed here is about $10^{12}$ photons/(cm$^2$ pulse) which creates roughly $10^{10}$ polaritons/(cm$^2$ pulse) at $k \approx k_m$. The probe photon density of about $2 \times 10^8$ generates roughly $1.5 \times 10^6$ polaritons/(cm$^2$ pulse) in the lower polariton branch at $k \approx 0$ (a similar density is created in the upper polariton branch). The cavity is slightly detuned, i.e. the empty cavity photon mode at zero in-plane wave-vector (1.4860 eV) has a slightly lower energy than that of the bare exciton energy (1.487 eV). The maximum amplification reaches about 60, and the spectral position of the gain peak is slightly shifted with respect to that of the unperturbed polariton, as predicted by theory [20]. The probe is weak enough so as not to affect the gain.

The arrival of the second probe pulse on the sample has a very strong influence on the amplification process, depending on its phase and on its delay. Figures 4b–d show the peak gain as a function of the relative phase. When there is no delay $\Delta t$ between the two probe pulses, the amplification can be controlled totally, i.e. by varying the relative phase between the two probe pulses, the gain changes in a reproducible way between zero and a maximum value. The fact that the amplification can be turned off completely tells us that the temporal and spatial superpositions of two probe beams on the sample are perfect. The oscillations are however not always sinusoidal as one could expect from an interference effect. Close to maximum amplification the gain vs. phase curve is flatter than a sine, as if there were some saturation effect. Strong oscillations in...
the gain also occur when the second pulse is delayed with respect to the coinciding first probe and pump pulses. The oscillations persist for delays of several ps. The amplitude of the oscillations in the reflected signal is in any case much larger than the intensity of the probe that controls them. The surprisingly long coherence times that can be observed even at relatively large excitation densities are a characteristic property of the polaritons occupying the states inside the above mentioned valley close to $k = 0$ of the lower polariton branch.

We define the contrast $C(\Delta \tau)$ of the oscillations on the signal as a function of delay between the two probe pulses,

$$C = \frac{G_{\text{max}} - G_{\text{min}}}{G_{\text{max}} + G_{\text{min}}},$$

where $G_{\text{min}}$ and $G_{\text{max}}$ are the maximum and minimum intensities of the emitted signal. $C(\Delta \tau)$ is a measure of how efficient the coherent control can be at a delay $\Delta \tau$ after the initial excitation. As can be seen from Fig. 5, the contrast decays in a non-trivial way with increasing delay. For relatively weak pump powers, it first remains about constant for a few ps, and decays only for bigger delays. The decay becomes faster with increasing pump intensities, however, the contrast without pump decays more rapidly than in the case of the weakest pump intensity we investigated. This is a signature of the stimulated parametric scattering process which transfers coherent polarisation from the pump to the signal at $k = 0$.

![Fig. 4. Coherent control of the parametric amplification. a) Probe reflectivity spectra with (full line) and without (dotted line) pump excitation, only one probe pulse hits the sample; b), c), and d) peak gain as a function of the relative phase between the two probe pulses. The first probe pulse temporally coincides with the pump pulse, the second one is delayed by $\Delta \tau = 0$, 2, and 4 ps, respectively](image-url)
It is interesting to investigate how signal, idler, and pump are correlated. Figure 6 shows the light intensities leaving the sample on the backside, as a function of emission angle and of the relative phase of the two probe pulses. The coarse delay between the two pulses amounts to 2 ps. The emission angle is measured with respect to the sample normal, i.e. the signal appears at about zero angle, the pump at about 9°, and the idler, much weaker, at slightly more than twice the pump angle. When the two probe pulses are in phase (modulo 2π), i.e. when the gain on the signal is maximal, also the (time integrated) idler emission is strongest, while the pump intensity is minimal. And when the two probe pulses are out of phase by π, the signal and idler emissions are almost completely quenched, and the pump is strongest. Such features are expected when the above described stimulated parametric polariton scattering process occurs. Further,

![Figure 6](online colour). Coherent control of the angular distribution of the light emitted in transmission geometry from the back of the microcavity. The surface plot shows the emission intensity as a function of the emission angle and the relative phase of the two probe pulses. The rough delay Δτ between the two phase controlled pulses is 2 ps. Signal and idler intensities are in phase, while that of the pump is in antiphase with respect to the probe. The signal and pump emissions (up to an angle of 14°) were attenuated by four orders of magnitude. In order to evidence the effects of the coherent control on the emitted pump intensity, the probe density employed here was higher (5 × 10⁸ polaritons/(cm²pulse)) than that in the reflection experiments, while the pump density was lower (∼10¹¹ polaritons/(cm²pulse))

Fig. 5. Contrast C (see Eq. (1)) of the coherent control oscillations versus delay between the two probe pulses, the first probe pulse is synchronous with the pump. Each curve corresponds to a different pump intensity. The dotted line has been taken without pump
since the effect of the two out of phase probe pulses cancels almost completely, there are signal and idler emissions caused by spontaneous polariton–polariton scattering. They appear at angles that differ somewhat from those of the stimulated process, in a similar way as can be observed when only the pump excites the MC (Fig. 3).

4. Comparison with Theory  A theoretical model for the MC parametric polariton amplifier has been developed by Ciuti et al. [20]. In its simplest version, the model accounts for three polarisation fields on the lower polariton branch of the MC, the pump $P_{k_p}$ at $k = k_p = k_{\text{magic}}$, the signal $P_0$ at $k = 0$, and the idler $P_{2k_p}$ at $k = 2k_p$. The temporal behaviour of these three polarisations are described by

\[ i\hbar \frac{dP_0}{dt} = (\tilde{E}(0) - i\gamma_0) P_0 + E_{\text{int}}P_{2k_p}^0 P_{k_p}^2 + F_{\text{probe}}(t), \]  

(2)

\[ i\hbar \frac{dP_{k_p}}{dt} = (\tilde{E}(k_p) - i\gamma_{2k_p}) P_{k_p} + 2E_{\text{int}}P_{k_p}^0 P_{2k_p} + F_{\text{pump}}(t), \]  

(3)

\[ i\hbar \frac{dP_{2k_p}}{dt} = (\tilde{E}(2k_p) - i\gamma_{2k_p}) P_{2k_p} + E_{\text{int}}P_0^0 P_{k_p}^2, \]  

(4)

In the first terms of the right-hand side of the above equations, the polariton energies $\tilde{E}(k)$ are renormalised with respect to the unperturbed energies $E(k)$. Due to the presence of an intense pump field $P_{k_p}$, $\tilde{E}(k)$ is blue-shifted by the repulsive polariton–polariton interactions,

\[ \tilde{E}(k) = E(k) + 2V_{kk_0} |P_{k_p}|^2, \]  

(5)

where the effective interaction potential $V_{kk_0}$ acts on two polaritons having initial wave vectors $k$ and $k'$ and exchanging momentum $p$. $V_{kk_0}$ is a sum of a direct contribution, a boson exchange contribution, and one due to the saturability of the exciton transition [20]. The damping rates $\gamma_k$ that also appear in the first terms on the right hand side include polariton dephasing and escape from the cavity. The second terms describe parametric scattering of the pump polaritons to signal and idler polaritons, and vice versa. The scattering rate depends again on the polariton–polariton interaction potential

\[ E_{\text{int}} = \frac{1}{2}(V_{k_0k_0k_{k_0}} + V_{k_{k_0}k_{k_0}k_{k_0}}). \]  

(6)

The inhomogeneous terms are the pump and probe driving fields.

These coupled equations have been integrated numerically. For the calculations discussed here, the following numerical parameters have been assumed: $\gamma_0 = \gamma_{k_p} = \gamma_{2k_p} = 0.15$ meV (which means that polariton damping is essentially caused by polariton escape from the MC); a polariton splitting of $3.5$ meV (as for our MC containing one In$_{0.03}$Ga$_{0.97}$As quantum well); exciton and cavity-mode energies $1.480$ eV (at $k = 0$); duration of the probe pulses $150$ fs; duration of the pump pulses $4$ ps; zero delay between pump and first probe pulses, delay between first and second probe pulses $2$ ps.

Figure 7 shows the temporal dependence of the signal, in a simple pump–probe experiment using only one probe pulse, and the probe and pump pulses hit the sample at the same time. The probe to pump intensity ratio is of the order of $10^{-4}$. The sharp rise close to $t = 0$ is caused by the penetration of the short probe pulse into the cavity.
Without pump, the probe polarisation just decays exponentially. The relatively long pump pulses lead to correspondingly slow changes in $|P_0|^2$ caused by the parametric scattering. With increasing pump intensity, this contribution of parametric scattering builds up more rapidly. Moreover, there is a clearly visible threshold behaviour for increasing pump intensity, since $-\gamma_0 P_0$ competes with $E_{int} P_{2k_p} P_{k_p}^2 / k_p^2$, see Eq. (2).

In Fig. 8 the calculated polariton dynamics during a coherent control experiment is shown. To be able to see the effects of the probe pulses on the pump, the pump to probe intensity ratio has been chosen to be as high as 0.1. When the two probe pulses coincide, the temporal behaviour of $P_0$ is similar to what can be seen in Fig. 7, and the number of idler polaritons just smoothly rises and reaches a maximum at about 5 ps. Moreover, the maximum signal and idler polarisations are nearly equal, as expected when there is strong PA. When the second probe pulse is delayed and the two polarisations interfere constructively inside the MC, at $t = 2$ ps a second step occurs in $|P_0|^2$ which is about three times as high as the first one (the two probe pulses have the same amplitude), and which brings the signal to the same intensity as in the case of undelayed pulses. Also for the idler polarisation $P_{2k_p}$ depending exclusively on parametric scattering, the slope becomes abruptly steeper at 2 ps. At about 5 ps also the idler reaches practically the same maximum intensity as when the two probe pulses coincide. In the case of destructively interfering probe polarisations, and for the example shown in Fig. 8, the number of signal polaritons left in the MC at 2 ps is slightly lower than that created by the second pulse, i.e. the resulting signal polarisation is strongly reduced and its phase is shifted by $\pi$. A phase shift of $\pi$ corresponds to a change of the sign of $P_0$ which leads to a sign change in the scattering rates transferring polarisation out of the pump and into the idler, but not for the polarisation transfer into the signal (Eqs. (2)–(4)). This means that the polarisations transferred from the pump to idler and signal, during the time interval between about 2 and 3 ps, both interfere destructively with the ones that are already there, and since parametric scattering conserves energy this means that polaritons are scattered back from idler and probe to the pump. This is clearly visible on the probe and idler polarisations shown in Fig. 8, in the interval between 2 and 3 ps $P_0$ and $P_{2k_p}$ decrease rapidly. For $t > 3$ ps, i.e. when the phase of $P_0$
has switched back ($|P_0|^2$ does not go to zero at the moment when the phase shifts by $\pi$). This is due to the slight shift of the probe energy as the number of pump polaritons varies.), parametric scattering from the pump to signal and idler takes place again, even though this happens at a reduced rate it leads again to a buildup of idler and signal polarisations.

The oscillations on the signal vs. relative phase of the probe pulses observed in a time integrated coherent control experiment can be extracted from the theoretical model by calculating the total energy emitted by the signal, $E_{\text{signal}} \propto \gamma_0^2 \int_0^\infty |P_0(t)|^2 \, dt$. The re-

Fig. 8. Calculated emission intensities of signal, pump, and idler as a function of time, in a coherent control experiment. The results are shown for two delays between the two probe pulses, $\Delta \tau = 0$ and $\Delta \tau = 2$ ps, and for constructive and destructive interferences of the two probe polarisations. The sample is excited at the pump angle with a relative intensity of 0.9 (see Fig. 7). The chosen numerical parameters are given in the text.
Results are shown in Fig. 9a for different delays $\Delta \tau$ between the probe pulses. Spontaneous parametric scattering and polarisation losses due to dephasing have been neglected. Qualitatively, the oscillations observed in the experiment (Fig. 4) are reproduced by the theoretical model, however, the “saturation” effect observed in the experiments is absent. As far as the contrast of these oscillations is concerned (Fig. 9b), the calculations reveal the same trend as observed in the experimental data: The contrast decays more rapidly when the pump is absent and when it is strong, than when the pump is relatively weak. To explain this, we neglect the effect of the idler for a moment, and note that for full contrast the amplitude of the polarisation created by the second probe pulse has to be equal to that of signal polaritons present in the cavity at the time when the second pulse penetrates the MC. This is only the case for a certain “critical” pump intensity at which parametric scattering compensates the escape rate of signal polaritons. When we take into account the effect of the idler which keeps parametric scattering going even when $P_0 = 0$, this critical pump intensity will just be somewhat reduced. In the examples shown in Fig. 9a, and in Fig. 7 for the relative pump densities of 0.85 and 1.0, the pump intensity is close to the critical value.

5. Conclusions Semiconductor microcavities that are in the strong coupling regime can indeed work as efficient and rapidly responding parametric amplifier. From our results it becomes obvious that probe pulses, that are much weaker than the amplified light leaving the sample, can control PA in a MC. There is an optimum pump intensity at which coherent control is most efficient, i.e. at which the interference of the probe

Fig. 9. Calculated signal gain, in time integration, in a coherent control experiment. The first probe pulse is synchronous with the pump. a) Gain vs. control phase, for three different delays $\Delta \tau$ between the two probe pulses; b) contrast $C$ of the coherent control oscillations versus delay $\Delta \tau$ between the two probe pulses. Each curve corresponds to a different intensity, the scale of the relative pump intensities is the same as in Fig. 7. The chosen numerical parameters are given in the text.
polarisations is still substantial for delays $\Delta t$ between the two probe pulses of several ps. The relatively simple theoretical model [20], based on just three polarisation components (pump, signal, and idler) which are coupled in third order via exciton–exciton interaction, permits to understand the dynamics of stimulated parametric scattering. This model predicts that the system can be prepared in such a way that polaritonic scattering goes backwards from the signal and idler to the pump. More experimental and theoretical investigations are needed to confirm these findings.

Acknowledgements We thank J. Bloch, M. A. Dupertius, A. Quattropani, P Schwendimann, Le Si Dang and R. Houdré for stimulating discussions. This work has been supported in part by the Fonds National de le Recherche Scientifique.

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