# Coherent and incoherent polaritonic gain in a planar semiconductor microcavity

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The gain processes in a semiconductor microcavity in the strong coupling regime have been studied by pump-probe experiments in transmission geometry. It is demonstrated that the nonlinear signal consists of two contributions, a coherent and an incoherent one. In agreement with recent reports, the coherent gain is identified as a parametric amplification process that is driven by the probe field and stimulates the scattering of polaritons into the  $\mathbf{k}_{\parallel}=0$  states. We attribute the incoherent gain to scattering of randomly distributed polaritons in the predominantly excitonic part of the lower polariton branch into states with zero wave number in the lower branch. Both processes are characterized by their polarization dependence and their sensitivity to the spectral position of the pump laser beams. They also show a pronounced threshold behavior versus the pump power.

## I. INTRODUCTION

During the last decade, the optical properties of semiconductor microcavities (MC's) have been a central topic of semiconductor research. These structures typically consist of single or multiple quantum wells placed between two high reflectance Bragg mirrors. One of the most intriguing properties of these structures is the recovery of the importance of polaritonic effects for the coupling of the quantum well exciton mode to the photon mode of the cavity.<sup>1</sup> Unlike in bulk, for quantum wells embedded in a homogeneous medium polaritonic effects can often be neglected due to the breaking of the translational invariance normal to the well plane. In contrast to bulk semiconductors, the energy of the optical mode in MC's and thus its separation from the exciton can be varied. This opens the possibility of tailoring the dispersion relations of the polaritons.<sup>2</sup>

Recently, nonlinear optical gain phenomena in microcavities have attracted considerable interest. Such processes observed for high particle densities where the strong coupling of exciton and photon is broken and the dynamics can be described by Fermi's golden rule are well investigated and a much discussed topic in the literature.<sup>3-14</sup> Here we study gain processes in the strong coupling regime,<sup>6,7</sup> in which interest has been boosted by claims of the so-called boser effect:<sup>8</sup> the polaritonic emission is greatly enhanced by scattering of excitations with large in-plane wave numbers  $\mathbf{k}_{\parallel}$  to states with  $\mathbf{k}_{\parallel} = 0$ . The scattering is stimulated by the  $\mathbf{k}_{\parallel} = 0$ polariton population. Recent studies reported evidence for this effect, because in photoluminescence measurements nonlinear emission from the lower polariton branch was observed even for low polariton densities, where the cavity is within the regime of strong exciton-photon still interaction.<sup>9,10</sup> In these investigations nonresonant excitation far above the band gap was applied to the cavity. In similar experiments the emission from the lower polariton branch was found to be increased, when the polaritons were resonantly excited into the upper polariton branch.<sup>12</sup> Furthermore, a strong correlation between the polarization of the excitation and amplification of the emission was found.<sup>12</sup>

However, these studies did not address the time evolution

of the observed amplification. Very recently, time resolved studies of the polaritonic amplification were reported experimentally and theoretically. The authors focus on coherent gain only.<sup>13,14</sup> They suggest a so-called parametric polariton amplifier, where the gain is attributed to stimulated polariton-polariton scattering due to coherent wave mixing. In this article we present a study of the time evolution of incoherent amplification processes in the strong coupling regime. We investigate the dynamics of gain mechanisms by pump-and-probe experiments: In these studies either the lower or the upper polariton branch is pumped selectively. Furthermore, the influence of relative circular polarization of the pulses was analyzed. The role of inter- and intrabranch scattering processes in the gain action is explored. As the origin of the incoherent gain we propose a probe-beaminduced scattering mechanism of polaritons from the predominantly excitonic part of the lower polariton dispersion relation into  $\mathbf{k}_{\parallel} = 0$  states.

# **II. EXPERIMENTAL DETAILS**

We have investigated a microcavity sample containing a single 7 nm wide  $In_xGa_{1-x}As$  (x=0.14) quantum well (QW) at the antinode of a  $\lambda$  GaAs cavity. The top (bottom) mirror consists of 21 (23) pairs of distributed Bragg reflectors with a reflectivity of 99.5%. The cavity length and thus the detuning  $\Delta = E_c - E_x$  between the cavity mode of energy  $E_c$  and the heavy hole exciton mode of energy  $E_x$  can be varied by changing the position of the laser spot on the sample. At resonance ( $\Delta = 0$ ) the transmission spectrum shows a Rabi splitting of 3.8 meV and linewidths of 1.0 meV and 1.2 meV [full width at half maximum (FWHM)] for the upper (UPB) and lower (LPB) polariton branches, respectively.

Pump-and-probe experiments were performed in transmission geometry using a femtosecond mode-locked Ti:sapphire laser with a repetition rate of 82 MHz. The spectrally broad probe pulse had a width of  $\sim$  30 meV (FWHM) and a duration of  $\sim$  80 fs. The pump pulse was spectrally tailored using a grating and a slit for exciting the polaritons in the upper or the lower polariton branch selectively. The spectrally narrow pump pulse had a width of  $\sim$  1.5 meV

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FIG. 1. Differential transmission signal  $(\Delta T/T_0)$  as function of energy for zero detuning when the pump beam is pumping the lower polariton branch. The dashed (dotted) line displays the differential transmission spectrum when pump and probe beam are cocircularly (anticircularly) polarized. These spectra are recorded at zero delay between pump and probe. The dash-dotted line shows the nonlinear signal for cocircular excitation at a delay of 50 ps. In addition, the normalized linear transmission of the probe is also shown (solid line). For clarity, the spectral region of the upper branch has been enlarged by a factor of 25.

(FWHM) corresponding to a duration of ~1.6 ps, limiting the time resolution of the setup. Both pulses were focused onto the same spot of the sample, having a diameter of roughly 50  $\mu$ m. In all experiments the probe beam hits the sample perpendicular to the surface (with an accuracy of  $\pm 1^{\circ}$ ). If not stated otherwise the angle between pump and probe beam was fixed at  $\Theta \sim 8^{\circ} \pm 1^{\circ}$  and the probe power was chosen to be less than 0.1 mW. The transmission was spectrally analyzed by a monochromator and detected by a liquid nitrogen cooled charge-coupled device camera as a function of photon energy *E* and delay  $\tau_{delay}$  between the two pulses. The sample was kept in a helium bath at a temperature of 2 K in all experiments.

## **III. PUMPING THE LOWER POLARITON BRANCH**

### A. Experimental data

In the following the results for the situation in which polaritons were excited in the LPB only will be discussed. Figure 1 shows the differential transmission at zero delay for different polarization conditions, when the cavity is in resonance ( $\Delta = 0$ ). The differential transmission signal is presented as  $\Delta T/T_0$ , where  $T_0$  is the transmission of the probe pulse in the absence of the pump (solid line in Fig. 1) and  $\Delta T = T$ (with pump) $-T_0$ . The pump power used in this experiment was 5.4 mW. For cocircular excitation (dashed



FIG. 2. (a) Gain spectra from the lower polariton branch for different delay times  $\tau_{delay}$ . The delay time was varied between  $\tau$ = 0 (upper trace) and 30 ps (lower trace) in steps of 1 ps. The LPB was pumped by a  $\sigma^+$  polarized beam; the probe beam was  $\sigma^+$ polarized as well. The arrows mark the spectral positions of the coherent gain and of the linear transmission signal. (b) Gain spectra of the LPB for cocircular excitation for pump powers between 0.2 and 4.8 mW. The  $P_{pump}$  has been increased in equal steps. The dotted line shows the gain at 2.0 mW pump power.

trace), i.e., both pulses are  $\sigma^+$  polarized, we observe a pronounced gain<sup>15</sup> in the LPB at zero delay. The transmitted signal is amplified by more than one order of magnitude. In contrast, no gain occurs at the position of the UPB as can be seen in the magnified section of Fig. 1. The gain shows a strong dependence on the relative polarization of the two beams. When the probe polarization is switched to  $\sigma^-$ , no nonlinear signal is observed in either polariton branch (dotted trace).

The spectral position of the gain in cocircular configuration lies slightly above the LPB energy of the empty cavity. However, this shift is small compared to the Rabi splitting. Since the cavity is in resonance, the uncoupled cavity mode would be located 1.9 meV above the lower polariton branch. From these observations we conclude that the cavity is in the strong coupling regime at all excitation conditions used in the present studies. We can therefore exclude high density effects such as excitonic bleaching from our considerations.

Gain can be observed for longer delay times between the two pulses of the same circular polarization, also. This can be seen from the pump-and-probe trace recorded for a delay of 50 ps which is shown in Fig. 1 by the dash-dotted line. The LPB shows considerable nonlinear transmission, while there is no gain in the UPB. However, in comparison to zero delay the gain is reduced by a factor of 5. Remarkably, at



FIG. 3. Spectrally integrated gain as function of delay for zero detuning, when the LPB is pumped. The solid dots represent the gain for cocircular excitation, while open dots show the gain for anticircular polarization. The solid line shows the fitted gain intensity and is calculated from the sum of coherent and incoherent gain. The dotted (dashed) line displays the coherent (incoherent) gain contributions. The inset shows the polarization degree of the polaritons as a function of delay time.

these long delay times gain is also observed for anticircular polarization of the two beams, as will be discussed later.

Figure 2(a) shows the evolution of the spectrally resolved gain as a function of the delay. Here we focus on the spectral region of the LPB to make the spectral shift discussed above more evident. The arrows mark the spectral positions of the gain at  $\tau_{delay} = 0$  and that of the linear transmission signal. The gain occurring for small delays is located above the spectral position of the LPB ( $E_{LPB}$ ). With increasing delay time it fades away and the slower gain of regime 2 appears in the spectra exactly at  $E_{LPB}$ . Thus the gain contains two energetically different components although the limited spectral resolution and the finite polariton linewidth in the present experiment prevent a clear separation of the two gain contributions. The twofold nature of the amplification process will be treated in more detail in the next section. The lower panel of Fig. 2 shows gain spectra recorded at zero delay for varying pump excitation powers  $P_{pump}$ , where the pump power has been increased in equal steps from 0.2 mW to 4.8 mW. The gain remains low for excitation powers  $P_{pump}$ <2.0 mW (dotted line in Fig. 2) and then rises strongly. Most strikingly, the energy at which this gain occurs does not depend on the pump excitation power, confirming that the spectral blueshift is not related to high density effects.

Figure 3 shows the evolution of the gain in the LPB with delay time. The solid dots represent the spectrally integrated intensity of the differential transmission as a function of the delay between the laser pulses, when the lower polariton branch is cocircularly pumped and probed. First, an instantaneous rise of the amplification is observed; then the gain decays. For the decay, two different regimes can clearly be distinguished. At delay times  $\tau_{delay} < 5$  ps a very fast decrease is observed. For longer delay times ( $\tau_{delay} > 20$  ps) we find a significantly slower decay on time scales of

 $\tau_{decay} \sim 50$  ps. The time evolution of the differential transmission has been studied for anticircular excitation, as well. The open dots in Fig. 3 show the differential transmission for  $\sigma^+ \sigma^-$  polarized beams. Under these conditions the magnitude of the gain is low compared to the cocircular case and shows a contrary time evolution. For small delays, no amplification is observed. Only for  $\tau_{delay} > 20$  ps does the amplification rise slowly, remaining well below the cocircular signal.

#### **B.** Discussion

From the decay behavior and the spectral positions of the gain as a function of the delay time it can be concluded that two different processes contribute to the amplification in cocircular configuration. The ultrafast gain in the first regime is in agreement with the observations reported recently by Savvidis et al.<sup>13</sup> In their pump-and-probe experiments gain that can be as large as two orders of magnitude appears when the direction of the pump beam relative to the cavity normal is tuned to a "magic" angle  $\Theta$ . Very recently, a theoretical model has been developed that can explain these observations:<sup>14</sup> The amplification arises from a parametric scattering of polaritons which are generated by the pump beam into the  $\mathbf{k}_{\parallel} = 0$  polariton states. The scattering is stimulated by coherent wave mixing of the pump with the probe beam, which leads to a giant amplification of the probe signal.

Since the pump beam hits the sample at an angle  $\Theta$ , it generates a polariton population in the lower polariton branch with a wave number  $\mathbf{k}_{\parallel}^{0}$  corresponding to this angle. The probe polaritons at  $\mathbf{k}_{\parallel} = 0$  stimulate the scattering of two of these  $\mathbf{k}_{\parallel}^{0}$  polaritons; one is transferred into a  $\mathbf{k}_{\parallel}=0$  state, while the other one is scattered into a state with  $2\mathbf{k}_{\parallel}^{0}$  (the so-called idler state) to fulfill momentum conservation. In addition, the energy has to be conserved in the scattering process:  $E(2\mathbf{k}_{\parallel}^{0}) - E(\mathbf{k}_{\parallel}^{0}) = E(\mathbf{k}_{\parallel}^{0}) - E(\mathbf{k}_{\parallel} = 0)$ . We note that in particular the energy conservation can be fulfilled due to change of the dispersion relation by polariton formation only. In contrast, momentum and energy conservation cannot be fulfilled for a pure exciton mode with its quadratic dispersion relation. These conservation conditions are satisfied in the sample investigated for a pump angle of  $\Theta \sim 8^{\circ}$  when the cavity is in resonance, corresponding to the angle of incidence of the pump beam in these experiments. The blueshift of the gain as compared to the linear LPB transmission originates from exciton-exciton interaction<sup>14</sup> and it does not depend on the pump power. Furthermore, coherent wave mixing can occur for polariton populations of the same circular polarization only. Therefore ultrafast coherent gain is not observed for the case of anticircular polarization of the pump and probe beams.

From the resonant scattering process a significant polariton population arises also in the idler state. This population at  $2\mathbf{k}_{\parallel}^{0}$  has been tested by monitoring the emission at an angle of  $2\Theta$ . Figure 4 shows emission spectra for different detection angles close to  $2\Theta$ . For these measurements the detuning has been changed to  $\Delta = -2$  meV, in order to resolve the idler state more clearly. For this detuning the resonant angle is increased to  $\Theta = 9.5^{\circ}$ . The spectrum is dominated by scattered light from the pump beam. When the



FIG. 4. Emission spectra of the lower polariton branch for different detection angles relative to the cavity normal. The detection direction was varied around the angle corresponding to the wave number  $2\mathbf{k}_{\parallel}^{0}$  of the idler state.

angle is tuned to the direction corresponding to  $2\mathbf{k}_{\parallel}^{0}$ , an emission line appears in the spectrum at an energy  $E(2\mathbf{k}_{\parallel}^{0})$ . This gives striking proof for the stimulated scattering process and its strong angle dependence. The emission from the idler is, however, considerably weaker than that from the  $\mathbf{k}_{\parallel}=0$ state which is observed normal to the sample ( $\Theta=0^{\circ}$ ) (see Fig. 1). This is very easily conceivable since the state at  $2\mathbf{k}_{\parallel}^{0}$ has already strong excitonic character. Therefore the population of this state is quickly reduced by the efficient scattering into the excitonlike portion of the polariton dispersion. An additional peak that is observed below the pump peak is most likely due to the emission of charged excitons (trions). It may also be caused by the emission of leaky modes.<sup>16</sup>

As indicated in Fig. 2(b), the gain shows a pronounced threshold as a function of the pump power  $P_{pump}$ . This be-



FIG. 5. Pump power dependence of the gain in the LPB in the first regime recorded at  $\tau_{delay}=0$  (hollow dots) and of the gain in the second time regime recorded at  $\tau_{delay}=20$  ps (full dots).

havior is summarized in Fig. 5, which shows the pump power dependence of the coherent gain at  $\tau_{delay} = 0$  (hollow dots). For  $P_{pump} \leq 2$  mW the gain is very low, and negligible as compared to the linear transmission signal. For higher powers gain appears, which first increases abruptly as  $P_{pump}$  is increased above 2 mW. For  $P_{pump}$  above about 4 mW the gain tends to saturate. The reduced increase of the gain for higher  $P_{pump}$  indicates that the dominant portion of the polaritons created by the pump beam are scattered into the  $\mathbf{k}_{\parallel} = 0$  state. The observed behavior is in good agreement with the theoretically predicted dependence, as is the lack of dependence of the spectral position of the ultrafast gain on the pump power [Fig. 2(b)].<sup>14</sup> In Fig. 5 also the corresponding dependence of the gain at a delay time of  $\tau=20$  ps is plotted (full dots). Surprisingly, its behavior is very similar to that of the coherent gain: It also shows a pronounced threshold, which occurs at about the same value for  $P_{pump}$  as in the case of the coherent gain. The gain at long delay times saturates for high excitation powers, as well.

The long lifetimes of the amplification cannot be explained within the framework of the coherent parametric amplification process but give evidence of additional gain mechanisms. Since we can observe gain even after delay times that exceed the typical dephasing times of polaritons<sup>19</sup> by more than one order of magnitude, the amplification at longer delay times is due to incoherent processes. This gives rise to the assumption that the observed gain is a superposition of the coherent and incoherent amplification phenomena. To analyze the time evolution in more detail, the gain intensities due to the two processes need to be separated from each other. This was done by the following procedure: Initially only polaritons with nonzero wave number are created by the pump beam at  $\mathbf{k}_{\parallel}^{0}$ . The coherent polaritons undergo a stimulated scattering process due to coherent wave mixing, which can occur as long as the coherence of the polaritons created by the probe beam is maintained. The rise of the coherent gain is given by the overlap of the pump and probe laser pulses and has been modeled by a Gaussian pulse with a width of 1.25 ps half width at half maximum. The decay behavior, on the other hand, depends on the laser pulse overlap and on the exponential decay of the coherent population. For simplicity, we have neglected the influence of the finite pulse width and have modeled the decay of the coherent amplification by a simple exponential decay with a decay constant  $au_{deph}$  corresponding to the dephasing time of the cavity polaritons (dotted line in Fig. 3). The time evolution of the incoherent gain process  $I_{incoh}$  is more complex. In our model it can be described with the following relation:

$$I_{incoh}(\tau) = \alpha [1 - \exp(-\tau/\tau_{deph})] \exp(-\tau/\tau_{incoh}).$$
(1)

Here  $\tau_{incoh}$  is the decay time of the incoherent amplification process, which is given by the spontaneous relaxation of polaritons and the emission into leaky modes. The first term describes the buildup of the incoherent polariton population, which can be calculated from the dephasing behavior of the initially coherent polariton population. The temporal evolutions of the coherent and incoherent gain are fitted simultaneously, where the time constants as well as the proportionality constant  $\alpha$  are taken as variable parameters. The solid line in Fig. 3 shows the results of the fitting procedure. It is



FIG. 6. (a) Magnitude of the coherent gain versus probe power  $P_{probe}$ . (b) Decay time of the incoherent gain as a function of  $P_{probe}$ .

calculated from the sum of both gain phenomena. In addition the dotted (dashed) trace displays the time evolution of the coherent (incoherent) amplification process. We find good agreement between the model and the experimental data. The incoherent portion of the gain rises fast, with a fitted dephasing time of  $\approx 1$  ps. This is followed by slow decay where the decay constant  $\tau_{incoh}$  is  $\approx 44$  ps. This long time constant reflects the polariton bottleneck that has been reported in the literature:<sup>23</sup> For polaritons in the excitonic part of the LPB the spontaneous relaxation into states with wave numbers close to zero is strongly reduced, because of the lack of final scattering states.

We can now analyze the dynamic properties of the pump polaritons. For zero delay between the two pulses, coherent wave mixing dominates the gain process and no incoherent polariton population is present in the resonator. If the delay between the arrival of the probe and that of the pump pulse increases, the pump polaritons undergo scattering processes and become predominantly incoherent. These incoherent polaritons than cause the amplification of the probe pulse when scattered into the  $\mathbf{k}_{\parallel}=0$  state as will be discussed below.

Further insight into the dynamic behavior of cavity polaritons can be obtained from a polarization analysis. The coherent gain was found to be of purely cocircular origin. In the case of the incoherent amplification, gain can be observed for anticircular polarization and long delay times also (open dots in Fig. 3). The incoherent gain for cocircular excitation



FIG. 7. Polariton dispersion relations for zero detuning. The suggested scattering mechanisms are indicated by arrows. The positions of the solid dots indicate the initial states. The inset shows the gain (for pumping the UPB) as a function of cavity detuning. The dashed line is a guide to the eye.

reaches its maximum while the gain for anticircular excitation is still zero. The amplification in this case occurs at significantly longer delay times and rises as a function of  $\tau_{delay}$ . These observations allow one to conclude that the probe beam can be amplified by polaritons of the same polarization only. For anticircular excitation the spin of the pump-induced polaritons must be flipped via long range exciton-exciton exchange interaction before gain can occur.<sup>20,21</sup> Its slow rise therefore reflects the polariton spin flip time. To investigate the underlying spin dynamics in more detail the time evolution of the spin polarization ( $\mathcal{P}$ ) of the polaritons has been evaluated. This spin polarization is defined by

$$\mathcal{P} = \frac{I^{++} - I^{+-}}{I^{++} + I^{+-}},$$

where *I* is the spectrally integrated differential transmission. The superscripts indicate the polarizations of the exciting pulses, ++ for cocircular and +- for anticircular excitation conditions. The inset of Fig. 3 shows a logarithmic plot of *P* as a function of  $\tau_{delay}$ . While at zero delay  $\mathcal{P}$  equals 1, it shows an exponential decay for delay times larger than 10 ps. From the decay time we can estimate a spin flip time of  $\tau_{SF}=63\pm10$  ps. This time is in good agreement with the rise time of the  $\sigma^+\sigma^-$  signal in Fig. 3.

In addition to the pump power dependence we investigate the role of the probe power in the coherent gain process. The probe power dependence is shown in Fig. 6(a). When the probe power is increased the amplification rises to reach a more or less constant level. For very low probe intensities no gain can be observed. We also investigate the dependence of the temporal evolution of the incoherent gain on the power of the probe beam  $P_{probe}$ . From Fig. 6(b) it can be concluded that  $P_{probe}$  has only a little influence on the time evolution of the gain. The decay constant  $\tau_{decay}$  remains unchanged in the recorded range.

Having obtained a complete set of experimental data, we will now propose a model that can explain the observations made for the time and polarization dependence of the incoherent amplification. Any explanation of the observed lightinduced mechanism must take into account the pronounced correlations between the pump and probe beams that are evident in the polarization selection rules for the gain. In order to obtain an amplification of the probe beam, the number of polaritons at the bottom of the lower polariton branch has to be increased due to polariton scattering into  $\mathbf{k}_{\parallel}=0$  states. Scattering involving phonons (e.g., two polaritons with wave numbers  $+\mathbf{k}_{\parallel}'$  and  $-\mathbf{k}_{\parallel}'$  could be scattered to  $\mathbf{k}_{\parallel} = 0$  with the excess energy emitted as an acoustic phonon) can be excluded, because this scattering channel would not show any dependence on the polarization of the probe polaritons. Therefore the nonlinearities must arise from polaritonpolariton scattering, since this scattering mechanism is dominantly determined by the interparticle exchange interaction and thus occurs between polaritons of the same spin orientation only.<sup>22</sup>

The picture that we envisage at longer delay times is the following: The probe beam creates a polariton population at zero wave number, which will trigger scattering processes of incoherent pump polaritons to the  $\mathbf{k}_{\parallel} = 0$  states. Different scattering channels exist for these processes; for example, polariton-polariton scattering can occur between the initial states  $+\mathbf{k}_{\parallel}'$  and  $-\mathbf{k}_{\parallel}'$  and the final  $\mathbf{k}_{\parallel}=0$  states with one lying in the LPB and the other one in the UPB, as indicated by the solid arrows in Fig. 7. Also, higher order processes might contribute: The energy released in a scattering process  $(+\mathbf{k}_{\parallel}',-\mathbf{k}_{\parallel}') \rightarrow (\mathbf{k}_{\parallel}=0,\mathbf{k}_{\parallel}=0)$  in the LPB will be transferred to a probe polariton that is scattered to the  $\mathbf{k}_{\parallel} = 0$  state in the upper branch.<sup>17</sup> This scattering process is sketched in Fig. 7 by the dotted arrows. In these considerations we concentrate on pump polaritons with large in-plane momentum. Due to the enhanced density of states in this region, it is expected that these states are the most relevant ones for scattering processes in the incoherent regime. Polaritons in the lower parts of the LPB will be emitted within a few picoseconds and therefore are irrelevant for processes at long delay times.

The model proposed above for the scattering processes is able to describe the experimental findings. However, the underlying mechanism of how the probe polaritons trigger the scattering of pump polaritons into  $\mathbf{k}_{\parallel} = 0$  states is not yet understood. A potential explanation might be provided by assuming that after long delay times wave mixing processes are also possible, e.g., the probe beam introduces coherence into the system. Parametric mixing should be possible between this wave and all other components with suitable phases with which constructive interference can occur. This wave mixing might lead to the amplification of the probe beam through the scattering processes discussed above. A related process might also provide an explanation of the strong nonlinear emission observed recently in nonresonant cw studies:<sup>9,10</sup> self-induced coherence in the  $\mathbf{k}_{\parallel}=0$  state, which is similar to the buildup of coherence in a semiconductor laser after injecting incoherent electron-hole pairs,



FIG. 8. Integrated differential transmission versus delay, when the upper polariton branch is excited. The cavity detuning is  $\Delta = -2$  meV. The solid (open) dots indicate co- (anti)circular polarization. The inset shows the normalized linear spectrum (solid line) and the differential transmission spectrum (dotted line) for cocircular excitation at a delay 40 ps.

might enable the wave mixing, as well. Future theoretical studies have to test the validity of these proposed mechanisms. Also, further experiments, e.g., on the temperature dependence of gain phenomena, are desirable.

In principle the suggested scattering mechanisms should lead to a gain in the upper polariton branch, as well. Our experiments show no or only little gain in the UPB. One reason can be the effective depopulation of  $\mathbf{k}_{\parallel}=0$  states in the UPB via interbranch scattering processes. This fast depopulation is in agreement with previous experimental studies reported in the literature.<sup>18</sup>

From the above considerations it is also evident that the amplification should have a strong dependence on the detuning of the cavity: The energy difference between the  $\mathbf{k}_{\parallel}=0$  states of the upper and lower polariton branches must be smaller than twice the energy separation between the excitonic part of the lower polariton branch dispersion and its  $\mathbf{k}_{\parallel}=0$  state ( $\Delta E$  in Fig. 7). In particular, for positive detuning this process should be strongly suppressed. Indeed, our experiments confirm the expected detuning dependence, as can be seen in the inset of Fig. 7.

#### **IV. PUMPING THE UPPER POLARITON BRANCH**

The picture that we have developed has to be tested for modified experimental conditions also. So far, the pump pulses were spectrally tailored to excite polaritons in the LPB to allow for the angle resonant amplification process. In the next step we increase the energy of the pump pulse, to excite polaritons resonantly in the upper branch. The observed differential transmission spectrum for cocircularly polarized pulses at  $\tau_{delay} = 40$  ps is shown in the inset of Fig. 8 (dotted line). The upper polariton branch is seen in the enlarged section. The solid line represents the linear transmission. No gain can be observed at the spectral position of the upper polariton branch. The absence of gain processes at the spectral position of the upper branch again indicates a very efficient interbranch scattering from the upper into the lower polariton branch. Polaritons from the upper branch are likely to be scattered into states with large  $\mathbf{k}_{\parallel}$  in the lower branch, since the density of states is strongly enhanced when the lower polariton branch becomes predominantly excitonic.

In contrast, when analyzing the differential transmission at the spectral position of the lower branch, we observe an amplification. However, the gain is considerably weaker compared to the amplification observed for pumping the lower branch directly. It reaches a maximum of  $\sim 60\%$  only. To obtain further insight into the dynamics of polariton scattering and the gain processes involved we again analyze the time evolution of the observed nonlinear signal and its polarization dependence. Figure 8 shows the integrated differential transmission at the spectral position of the lower polariton branch as a function of the delay. The pump and probe beams were cocircularly polarized for solid dots and anticircularly for open dots. The cavity detuning is slightly negative ( $\Delta = -2$  meV). In both cases there is no gain at short delay times, since the polaritons have to undergo scattering into the lower polariton branch first. In the cocircular case it remains close to zero for the first 5 ps and then rises to reach a maximum after 60 ps. This is followed by a surprisingly slow decay, where the decay time is calculated to be  $\tau_{decay} = 0.5$  ns from an exponential fit. This is about one order of magnitude longer than for pumping the lower branch. The long time scales involved in the gain prove that the amplification processes are purely of incoherent origin in this case.

For anticircular polarization the gain rises considerably more slowly as function of the delay. It is still close to zero when the cocircular amplification is at its highest value, and it reaches maximum intensity after 200 ps. Then it remains on a constant level for the rest of the recorded delay range. Again, the anticircular amplification remains below the gain observed for cocircular excitation. This indicates that the polarization dependence of this gain process is the same as in the case of pumping the lower polariton branch: The polaritons injected by the pump beam must undergo a spin flip before they can contribute to the amplification. The spin flip time can be estimated by analyzing the rise time of the anticircular signal, assuming that the scattering time from the upper into the lower branch is comparatively small. We find a spin flip time of  $\tau_{SF} = 60 \pm 10$  ps, which is in perfect agreement with the results obtained above.

However, when pumping the upper branch the decay times of the incoherent gain are increased by about an order of magnitude in comparison with lower polariton branch pumping. The principle difference between the two processes is the way of populating the LPB. When pumping the lower branch, on the one hand, the polaritons are injected at low  $\mathbf{k}_{\parallel}$ . In this case the polaritons that participate in the incoherent gain predominantly originate from the pump population. Even after the polariton distribution becomes randomized by polariton-polariton or polariton-phonon scattering, a considerable portion of the polaritons will still be concentrated in a region of the k space with rather moderate wave numbers. Therefore the probability of finding a polariton pair with wave numbers  $(+\mathbf{k}_{\parallel}', -\mathbf{k}_{\parallel}')$  which can undergo scattering to  $(\mathbf{k}_{\parallel}=0,\mathbf{k}_{\parallel}=0)$  is relatively large. On the other hand, when pumping the upper branch, fast relaxation of the excitations from the upper branch into states with large  $\mathbf{k}_{\parallel}$  of the lower branch occurs and the polaritons are spread widely in momentum space. Therefore the probability of finding a polariton pair  $(+\mathbf{k}_{\parallel}', -\mathbf{k}_{\parallel}')$  is rather small, leading to the much longer lasting decay of the gain.

### **V. CONCLUSIONS**

In conclusion, we have studied coherent and incoherent amplification phenomena in a semiconductor microcavity by pump-and-probe spectroscopy. We have proposed a model based on polariton-polariton scattering, which provides a consistent description of the experimental observations. The gain processes show characteristic dependencies on the excitation energy and the polarizations of the laser beams as well as on the detuning of the cavity. They also show a pronounced threshold behavior as the power of the pump beam is varied. The dynamic properties of the gain depend critically on the distribution of the polaritons generated by the pump beam. In particular, the data indicate that polaritons in the excitonic part of the lower polariton branch cause the incoherent gain. We are hopeful that the experimental results presented here might trigger additional theoretical analysis that will help to obtain a more detailed understanding of the incoherent gain processes.

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