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Correlated polaritonic emission from a semiconductor microcavity

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We experimentally demonstrate the existence of intensity correlations between different zones of the far field emission from a semiconductor microcavity, that is, between polariton fields with different transverse wave vectors. The origin of these correlations is attributed to a new pair scattering process from two distinct polariton modes populated by cw resonant pumping.

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I. INTRODUCTION

In strong-coupling semiconductor microcavities [1] the system is described in terms of "mixed" exciton-photon quasiparticles, the cavity polaritons. These composite bosons have very interesting properties. Due to their photon component, polaritons with a given transverse wave vector \mathbf{k} can be directly created by an incident laser beam with the appropriate energy and momentum. For the same reason, the angular distribution of the emission provides information about the polariton population along the dispersion curve. The exciton part is responsible for the coupling between polariton modes via the Coulomb interaction, which is at the origin of polaritonic nonlinearities. The understanding of these nonlinearities was greatly improved by the results of Savvidis et al. [2], who demonstrated parametric amplification of a probe beam at normal incidence, when a polariton population was created at a specific wave vector \mathbf{k}_p by a resonant pump beam. Detection of a weak "idler" beam with wave vector $2\mathbf{k}_p$ allowed to identify the nonlinear mechanism as parametric scattering of two polaritons from the pump mode into a signal-idler pair $\{\mathbf{k}_p, \mathbf{k}_p\} \Rightarrow \{0, 2\mathbf{k}_p\}$ [3]. The system has many similarities with an optical parametric oscillator [4], which were underlined by the demonstration of parametric oscillation [5], phase coherence of the nonlinear emission [6] and more recently optical bistability [7].

In this paper, we report on the experimental investigation of a different scattering process. We study the nonlinearities arising from the interaction of two distinct pump modes, populated by resonant cw pumping. For this we use two pump beams resonant with the lower polariton branch, having opposite planar wave vectors $\{\mathbf{k}_p, -\mathbf{k}_p\}$. Our scheme is comparable to the one used by Huang et al. [8]. They created an excitonic reservoir at large wavevectors by two pump beams with opposite incidence angle ($\pm 45^\circ$), and stimulated the scattering towards the bottom of the upper branch with a seed at normal incidence, resonant with the lower polariton branch. In contrast with the experiment by Huang et al., in which scattering involving both polariton branches is demonstrated, we study scattering processes that involve only the lower polariton branch. For this, we set

the pump beam at smaller angles ($\simeq 6^\circ$), with results in a more efficient excitation of the system, because the small- \mathbf{k} polariton states have a larger photon component with respect to the large- \mathbf{k} excitonic states. We look for cross scattering processes, in which one polariton is annihilated from each of the two pump modes to create a pair of signal-idler polaritons. Momentum conservation requires that the signal and idler have opposite wave vectors, while the resonance condition imposes $|k| = |k_p|$. As a consequence, all pair scattering processes $\{\mathbf{k}_p, -\mathbf{k}_p\} \Rightarrow \{\mathbf{k}, -\mathbf{k}\}$ with the condition $|k| = |k_p|$ are possible. Nonlinear emission is expected on a ring in the far field plane, the diameter of which is fixed by the pump wave vector. It is well known that resonant Rayleigh scattering (RRS) has the same far field pattern [9]. Since the parametric process creates polaritons with opposite wave vector in pairs, we expect to detect intensity correlations in the emitted light field between points located at both extremities of a diameter in the far field pattern. On the contrary, the different parts of the ring emission produced by RRS are not correlated. By measuring intensity correlations in the far field emission, the contribution of the nonlinear process can be evidenced.

II. EXPERIMENTAL SETUP

The microcavity sample is described in detail in Ref. [10]. It is a high quality factor 2λ GaAs/AlAs cavity, with three low indium content $\text{In}_{0.04}\text{Ga}_{0.96}\text{As}$ quantum wells, one at each antinode of the cavity mode. The Rabi splitting energy is 5.1 meV. The experimental setup is shown in Fig. 1.

The microcavity sample is cooled at 4K in a cold finger cryostat. The laser source is a cw, single mode Ti:Sa laser. Two pump beams resonant with the lower polariton branch are focused on the sample on a spot of 80 μm diameter with the same angle of about 6° . This angle is chosen so as to avoid competition with the one-pump scattering channel $\{k_p, k_p\} \Rightarrow \{0, 2k_p\}$ the efficiency of which is maximal at the so-called "magic angle" i.e. about 12° for this sample. The far field of the microcavity emission in transmission is obtained using a 50mm lens and imaged on a CCD camera (fig 2 (a), (b)). Two independently movable screens allow to select a part of

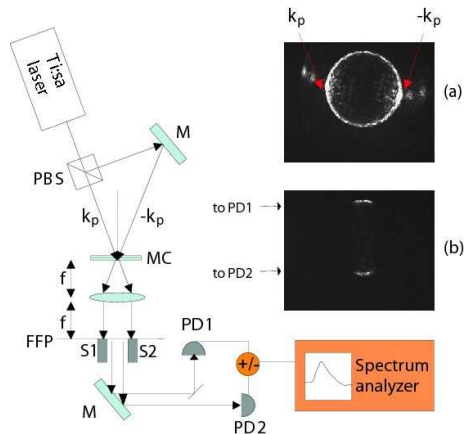


FIG. 1: Experimental setup. Two pump beams with opposite planar wave vectors k_p and $-k_p$ are focused on the microcavity sample MC. The microcavity emission is collimated by a lens (focal length $f = 50$ mm). The emission pattern is a ring in the far field plane (FFP) (a). Two movable screens S1 and S2 are used to eliminate the transmitted pump beams and select the emission localized at both extremities of the vertical diameter of the ring (b). The upper part of the emission is detected by photodiode PD1, the lower part by photodiode PD2. The photocurrent sum and difference are then spectrally analyzed.

the far field emission. The pair of selected beams is subsequently detected by two identical photodiodes. The two photocurrents are amplified and their sum or difference is sent to a spectrum analyzer.

III. RESULTS

The far field pattern of the emission shows a bright ring corresponding to the condition $|k| = |k_p|$ (see Fig. 1 (a)). As explained above, it can be attributed to both RRS and polariton pair scattering. In order to estimate the contributions of these two mechanisms, we measured the steady state intensity detected by each photodiode when only one of the pump beams was present, for each pump beam. Then we compared it to the intensity detected when both pumps were simultaneously present. This straightforward procedure already yields evidence for a nonlinear response of the system, because the intensity detected with both pumps is higher than the sum of the individual contributions previously measured. We can roughly evaluate the relative amount of the pair scattering with respect to RRS under the approximation that the three processes involved (pair scattering, RRS of the $+\mathbf{k}_p$ polaritons and RRS of the $-\mathbf{k}_p$ polaritons) are independent. The contribution of the pair scattering is then $I_{ps} = I_{tot} - I_{k_p} - I_{-k_p}$, where I_{tot} (respectively I_{k_p} , I_{-k_p}) is the intensity of the emission ring with two pump beams on (respectively with only the $+\mathbf{k}_p$, $-\mathbf{k}_p$ pump beam on). In Fig. 2 we plot the quantity I_{ps}/I_{tot} as a function of the pump power. The cavity-exciton detuning δ is fixed

at -0.45 meV. We see that the relative contribution of the pair scattering increases linearly with the pump power, and it becomes equal to RRS for a pump power of about 20 mW.

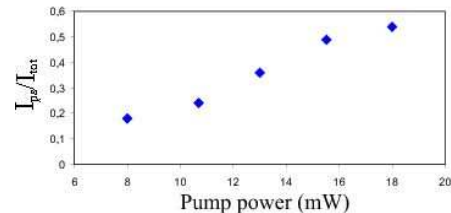


FIG. 2: Relative contribution of the pair scattering to the total emission, defined as I_{ps}/I_{tot} , as a function of the pump power.

The measurement of intensity correlations between the emissions at \mathbf{k} and $-\mathbf{k}$ provides reliable experimental evidence of the pair scattering; since emissions are expected to be uncorrelated in the case of RRS and strongly correlated in the case of pair scattering, the degree of correlation will depend on the relative contribution of these two mechanisms. To do this we select the emission from opposite regions of the emission ring, using the movable screens, and measure the noises of the photocurrent sum and difference, as shown in Fig. 1. A typical result is shown in Fig. 3 (a), for a cavity-exciton detuning $\delta = -0.45$ meV and an incident power of 24 mW (corresponding to about 2×10^8 excitons/cm²) for each pump beam.

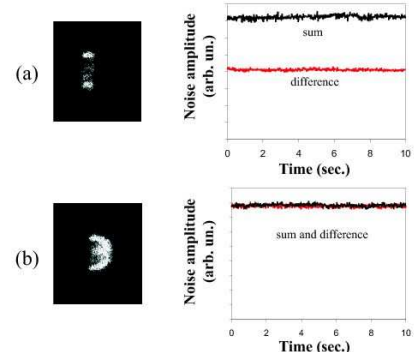


FIG. 3: Noises of the photocurrent sum and difference for two different positions of the screens S1 and S2 in the far field plane. The thickness of the ring in the images is due to the fact that we have used a diffusion screen. The upper half of the pattern is detected by PD1, the lower half by PD2, as shown in fig 1. (a) The selected zones lie on a diameter of the far field ring. Sum and difference of the photocurrents have different noise levels, and thus the two photocurrents are correlated. (b) The selected zones do not lie on a diameter of the far field ring. Sum and difference of the photocurrents have the same noise levels, and thus the two photocurrents are not correlated.

The noise of the photocurrent difference is much lower

than that of the sum, indicating a significant intensity correlation. In all the experiments described here, the frequency of analysis is fixed at the same value of 4 MHz. We did not observe any frequency dependence in the available range (2-30 MHz). This is due to the fact that the width of the correlation spectrum is expected to be of the same order as the width of the polariton resonance, i.e. a few tens of GHz.

We have performed further experiments in order to demonstrate that these correlations are due to parametric pair scattering from the two pump modes. In the first type of experiment, we select regions that are not symmetric with respect to the center of the ring (see Fig. 3 (b)). The noises of the photocurrent sum and difference are equal, which means that the two photocurrents are completely uncorrelated. Indeed, we have measured nonzero intensity correlations only when the selected parts of the ring lie on the same diameter, that is, when the polaritons at the origin of the emission have opposite wave vectors.

In the second type of experiment, starting from a situation where significant correlations are present, we block one of the pump beams. Then we observed again that sum and difference noises were exactly equal: the two parts of the emission ring that were previously correlated are completely independent when one pump beam is blocked. This confirms directly that the different parts of the RRS ring are uncorrelated. We have measured nonzero intensity correlations only in presence of both pump beams. From these results we conclude that the origin of the observed correlations is indeed the nonlinear process $\{k_p, -k_p\} \Rightarrow \{k, -k\}$.

In fig. 4 we show the response of the system when the pump power is increased. Fig. 4 (a) reports the contribution to the emission due respectively to RRS and to pair scattering. From these data fig. 2 has been obtained. Fig. 4 (b) shows the corresponding variations of the noises of the photocurrent sum and difference. As the contribution of the pair scattering increases, the noise of the photocurrent sum rises rapidly while the noise of the photocurrent difference remains quite low. For quantitative analysis, we define the normalized intensity correlation between the beams $\{\mathbf{k}, -\mathbf{k}\}$ as

$$C_{k,-k}(\Omega) = \frac{S_{k,-k}(\Omega)}{\sqrt{S_k(\Omega)S_{-k}(\Omega)}} \quad (1)$$

where $S_{k,-k}$ is the intensity correlation spectrum (defined as the Fourier transform of the correlation function $C_{k,-k}(\tau) = \langle \delta I_k(t) \delta I_{-k}(t + \tau) \rangle$) and S_k, S_{-k} are the intensity noise spectra of the two beams (defined as the Fourier transform of the intensity autocorrelation function $C_I(\tau) = \langle \delta I(t) \delta I(t + \tau) \rangle$). This quantity verifies $|C_{k,-k}(\Omega)| \leq 1$ and is equal to 1 for perfect correlations and -1 for perfect anticorrelations. The correlation spectrum $S_{k,-k}$ can be easily deduced from the noises of the intensity sum (S_+) and difference (S_-): $S_{k,-k} = (S_+ - S_-)/4\sqrt{S_k S_{-k}}$, and the noise spectrum

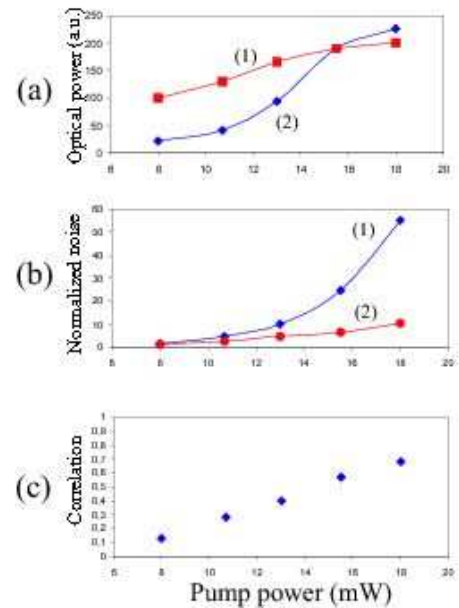


FIG. 4: (a): RRS (1) and pair scattering emission (2). (b): noise power of the photocurrent sum (1) and of the photocurrent difference (2) for increasing pump power. The curves are taken at a fixed noise frequency of 4 MHz and normalized at the standard quantum limit (dashed line). (c): correlation coefficient of the two photocurrents. All the quantities are measured as a function of the pump power.

of each beam can be measured directly by blocking the other one.

The normalized intensity correlation is plotted as a function of pump power in fig. 4(c). As the pump power increases and the pair scattering emission becomes important, the correlations between the two parts of the ring build up to a value bigger than 0.6.

IV. DISCUSSION

These results, while demonstrating the polariton parametric process $\{k_p, -k_p\} \rightarrow \{k, -k\}$ and showing that strong correlation can be obtained, raise a few important issues that require further study.

The first issue concerns the parametric oscillation threshold. Indeed, in the same way as in experiments using one pump beam, the system is expected to oscillate above some threshold intensity. The fact that the intensity noise of the sum increases and correlations build up suggests that threshold is gradually approached but not reached. Because of the circular symmetry of the system, all the possible $\{k, -k\}$ signal-idler pairs (with $|k| = |k_p|$) are expected to have the same oscillation threshold. However, it has been shown that in optical parametric oscillators two signal-idler pairs cannot oscillate simultaneously [11]. If this reasoning can be extended to our system, one should expect that when the oscillation threshold

is reached, a transition should occur and only one pair should oscillate. Then one should observe two bright spots on a diameter of the $|k| = |k_p|$ circle in the far field of the emitted light. Since we observe an uniform spatial intensity distribution over the far field ring, we conclude that the system is below oscillation threshold.

Another issue is the dependence of the parametric scattering on the polarization of the pump beams. All the results presented here were obtained with linearly polarized pump beams, respectively horizontally for the beam at k_p and vertically for the beam at $-k_p$. We have tested several other configurations, for example with the two pumps linearly polarized along the same directions, or circularly polarized, and found that the polarization of the pump beams has little effect on the pair scattering efficacy. This can be explained observing that for every given combination of spin orientations of the polaritons created by the pump beams, it is possible to scatter into a signal-idler pair that conserves the total spin. Such a behavior differs from what is observed in pump-probe experiments [2, 12].

We finally discuss the possibility of achieving quantum correlations between the signal and idler beam intensities [13]. It should be noted that signal-idler correlations are much more easily measured with our geometry with respect to the usual one with only one pump beam. This is due to the perfect symmetry between signal and idler: they have opposite wave vectors, hence the same photon content and the same linewidth. On the contrary, in the one-pump mechanism the idler polariton has a small photon fraction ($\simeq 0.05$) and is coupled to large- \mathbf{k} exciton modes. As a result, the emitted idler beam is much weaker than the signal beam at $\mathbf{k} = \mathbf{0}$ (the intensity ratio can vary between 10 and 10^4 depending on the sample characteristics), with a larger linewidth and a larger angular divergence. In addition, the signal and idler energies are separated by a few meV. This asymmetry makes it extremely difficult to evidence quantum correlations between the signal and idler beams [14]. From this point of view, our scheme is a promising candidate for the generation of quantum correlated polariton modes.

Putting aside any other interaction mechanisms, parametric pair scattering is expected to produce quantum correlated polariton fields inside the microcavity. The correlations between the intensities of the emitted light beams are then reduced because only the photon part of the polariton field is detected, which corresponds to losses [14]; however, they retain a quantum character. If the beams have equal intensities, the quantum character of correlations can be evidenced by showing that the noise of the intensity difference is below the standard quantum limit [15] (even though the intensity noise of each beam can be extremely high; in particular the fluctuations of the signal and idler beams are expected to diverge in the vicinity of the parametric threshold). In our experimental results it can be seen that the noise of the intensity difference is above the standard quantum limit (fig. 4(b)). Several factors can be at the origin of the

loss of the quantum character of the correlations. First, the signal-idler correlations are expected to be maximal in the vicinity of threshold; further studies are required in order to locate precisely the parametric threshold and find the optimal conditions for the correlation measurement. More importantly, the simple picture described above neglects all other interaction mechanisms involving polaritons. Among them, RRS plays an important role, because the scattered light is superimposed on the parametric emission in the ring $|k| = |k_p|$. Since the intensities of the scattered light in two opposite directions $\{k, -k\}$ are uncorrelated, the contribution of RRS to the intensity noises of the selected beams at $\{k, -k\}$ does not cancel out when the noises are subtracted, which diminishes the measured correlation. A promising direction for future investigations is to use slightly different excitation geometries, in which the emission patterns of RRS and parametric pair scattering would not overlap [16]. Moreover, it is also well known that polariton modes can couple to neighboring modes (in energy and wave vector) by absorption or emission of acoustic phonons. Even though this relaxation process is less important at low temperature, it is still the main source of resonant photoluminescence [10, 17] which adds some background excess noise to the parametric emission [18]. Since the luminescence intensities in two opposite directions $\{k, -k\}$ are uncorrelated, the excess noises do not cancel out. It is difficult to estimate whether the contribution of this process is large enough to suppress the quantum correlations between the emitted beams; however the results of ref.[18] are quite encouraging, showing that quantum noise reduction can be achieved in spite of phonon relaxation.

V. CONCLUSION

We have reported the experimental observation of a novel scattering process. As a result of the crossed non-linear interaction between two distinct polariton modes, resonantly populated by two pump beams, strong correlations are observed between different parts of the far field emission, originating from polariton modes with opposite wave vectors. This result constitutes a very interesting complement to the experiments on parametric polariton oscillation using one pump beam, since in our case the huge asymmetry between the signal and idler mode is completely suppressed. The measured correlations are *classical*, meaning that the noise of the signal-idler intensity difference is above the standard quantum limit. However, the idea of using two pump beams is very versatile and can be easily extended to other geometries that may prove more advantageous. In particular, it constitutes a promising first step towards the generation of quantum correlated polariton modes.

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