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Four wave mixing oscillation in a semiconductor microcavity: Generation of two correlated polariton populations

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We demonstrate a novel kind of polariton four wave mixing oscillation. Two pump polaritons scatter towards final states that emit two beams of equal intensity, separated both spatially and in polarization with respect to the pumps. The measurement of the intensity fluctuations of the emitted light demonstrates that the final states are strongly correlated.

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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 [2]. 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 [3]. 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. [4], 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 $\{\mathbf{k}_p, \mathbf{k}_p\} \Rightarrow \{0, 2\mathbf{k}_p\}$ [5]. The system has many similarities with an optical parametric oscillator (OPO) [6], further underlined by the demonstration of parametric oscillation [7], phase coherence of the nonlinear emission [8] and more recently optical bistability [9]. A remarkable property of OPOs above the oscillation threshold is the generation of two bright quantum correlated beams [10]. In principle, polariton parametric oscillation should lead to the same effect and generate two macroscopically populated polariton modes (the signal and idler polaritons) which are quantum correlated [11, 12]. In practice, however, a major problem arises because of the asymmetry between the signal and idler modes. Due to its large wave vector, the idler has a small photonic fraction, typically $\simeq 5\%$. Moreover, its energy is very close to the bare exciton energy, and this allows a very efficient relaxation towards large $\mathbf{k}$ excitonic states. As a result, the idler has a large linewidth compared to the signal, and a much weaker intensity (the intensity ratio is typically $\sim 10^2 - 10^4$ [7, 13, 14]). This makes it very difficult to measure the correlations of the signal and idler above the oscillation threshold. Even

though a signature of quantum pair correlations has been recently shown in the regime of multimode parametric scattering below the oscillation threshold [15], to the best of our knowledge a direct evidence of signal-idler correlations in the oscillation regime is lacking.

In this paper, we describe a polariton four wave mixing oscillator that generates equilibrated signal and idler beams. Above the oscillation threshold, these beams have the same intensity, since they are produced by polariton states having opposite wave vector, and hence the same photonic fraction and linewidth. Furthermore, they are spatially separated and linearly polarized, with orthogonal polarization with respect to the pumps: These features greatly simplify the optical detection, since they permit to filter out the pump light and the secondary emission produced by elastic scattering (resonant Rayleigh scattering). We have measured the intensity correlations of the two beams, finding that almost perfect correlations are present. In the strong coupling regime, extracavity photon fields carry the ampli-

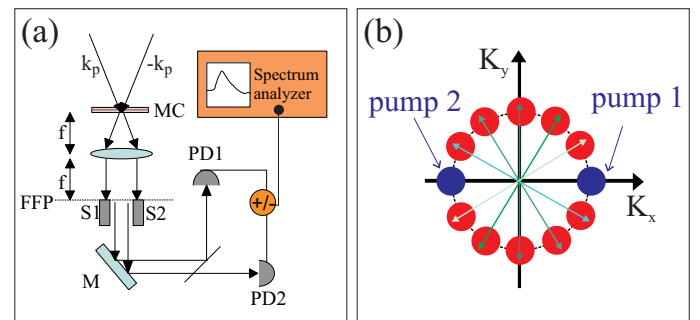


FIG. 1: (a) Experimental setup. Two pump beams $\mathbf{k}_p$ and $-\mathbf{k}_p$ are focused on the microcavity sample MC. The microcavity emission is collimated by a lens ($f = 50$ mm). Two movable screens S1 and S2 placed in the far field plane (FFP) eliminate the transmitted pump beams. The upper part of the emission is detected by photodiode PD1, the lower part by photodiode PD2. The photocurrent sum and difference are then spectrally analyzed. (b) Representation in $\mathbf{k}$ -space of the expected scattering processes. Correlated pairs are connected by arrows.

tude and phase information of the intracavity polariton fields; therefore, our results demonstrate the generation of two strongly correlated macroscopic polariton populations, in which the electronic excitations in the semiconductor quantum well are involved.

The experimental setup is shown in fig. 1(a). We use two pump beams resonant with the lower polariton branch, having opposite in-plane wave vectors $\{\mathbf{k}_p, -\mathbf{k}_p\}$. For crossed scattering processes involving one polariton from each pump mode, momentum conservation requires that the signal and idler have opposite wave vectors, while energy conservation imposes $|\mathbf{k}| = |\mathbf{k}_p|$. As a consequence, all pair scattering processes $\{\mathbf{k}_p, -\mathbf{k}_p\} \Rightarrow \{\mathbf{k}, -\mathbf{k}\}$ with the condition $|\mathbf{k}| = |\mathbf{k}_p|$ are in principle allowed. Nonlinear emission is expected on a circle in the far field plane, whose diameter is fixed by the pump wave vector (see fig. 1(b)). The incidence angle of the pump beams is of about 6° . This value is chosen so as to avoid competition with the one-pump scattering channel $\{\mathbf{k}_p, \mathbf{k}_p\} \Rightarrow \{\mathbf{0}, 2\mathbf{k}_p\}$ the efficiency of which is maximal at the so-called "magic angle" i.e. about 12° for this sample. The two linearly TM-polarized (polarization in the incidence plane $\leftrightarrow$) pumps are focused on the microcavity sample on a spot of $80 \mu\text{m}$ diameter. The polarization resolved far field of the transmitted microcavity emission is obtained using a 50mm lens and

imaged on a CCD camera. Two independently movable screens allow to select a part of the far field emission, which is subsequently detected by two identical photodiodes. The two photocurrents are amplified and their sum or difference is sent to a spectrum analyzer. The microcavity sample is cooled at 4K in a cold finger cryostat. The sample is described in detail in Ref.[16]. 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 . Polariton linewidths are in the $100\text{-}200 \mu\text{eV}$ range. The laser source is a cw Ti:Sa laser, intensity stabilized, with 1 MHz linewidth.

In fig.2 we show the far field emission detected on the TE-linear polarization, *orthogonal* to the pump polarization, for several values of the pump power. For low pump power (a-b), the emitted TE-polarized light is weak and roughly uniformly distributed on the elastic circle. For higher pump power (c-d), two bright spots located on the vertical diameter appear, breaking the circular symmetry of the emission. This surprising feature is a strong indication that the oscillation threshold has been reached. Above threshold, polaritons do not redistribute uniformly over the elastic circle, rather they scatter preferentially towards states localized around a well defined diameter, the vertical one. As a result, two macroscopic polariton populations build up in these states (fig.2(c-d)). We note that related phenomena of polarization inversion in the stimulated scattering regime have been recently observed [17, 18, 19]. Our results can be explained by taking into account the polariton spin dynamics [20, 21]; this issue will be discussed in detail in a forthcoming paper.

In fig. 3, we show the optical power emitted by the upper and lower polariton populations as a function of the pump power. Oscillation threshold is reached at about 20 mW of pump power. Everywhere in text, "pump power" means the sum of the power of each pump beam. Above threshold, the power ratio of the two beams is equal to

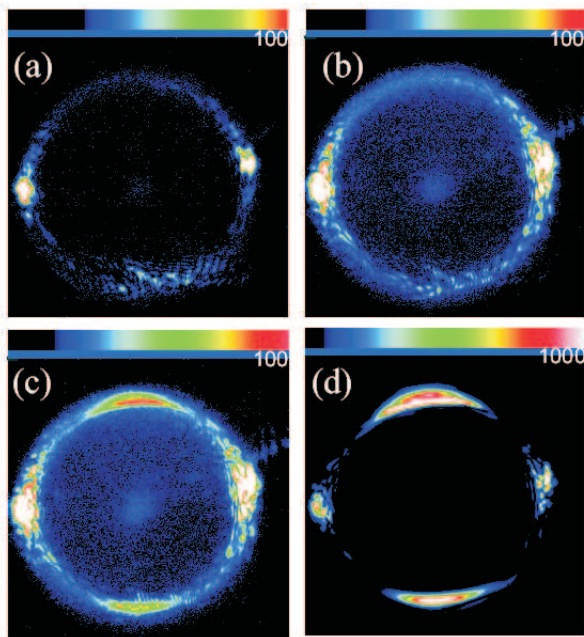


FIG. 2: (a) Far field emission in the TE-polarization for a total pump power of: (a) 3 mW ; (b) 19 mW ; (c) 27 mW ; (d) 39 mW . Cavity-exciton detuning $\delta = 0$. At low pump power (a-b), light intensity is roughly uniform on the elastic circle (except for the two pump spots, lying approximately on the horizontal diameter). At high pump power (c-d), scattered polaritons accumulate around the vertical diameter.

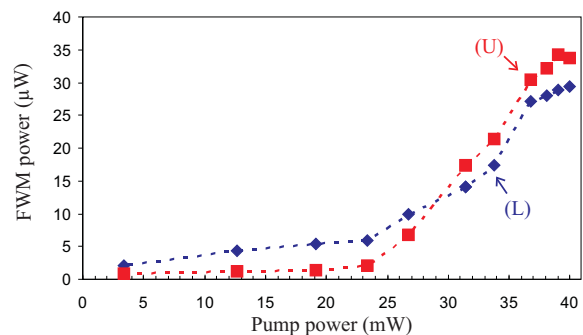


FIG. 3: Optical power of the upper (U) and lower (L) beam as a function of the pump power. Cavity-exciton detuning $\delta = 0$.

about 1.15. We stress that this situation is very different from the conventional "magic-angle" configuration, where the signal beam is $10^2 - 10^4$ times stronger than the idler [7, 13, 14].

In the case of four-wave mixing emission, the two conjugated beams are expected to exhibit extremely strong intensity correlations, since every scattering process from the two pumps must create one polariton in each of the states on the vertical diameter, in order to fulfill energy-momentum conservation. We have verified this feature by measuring the intensity correlations of the two beams. In order to do so, we have detected the beams with two identical photodiodes. The generated photocurrents are electronically added or subtracted; the noise spectra of the photocurrent sum and difference are measured with a spectrum analyzer, as shown in Fig. 1. 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 experimentally available frequency 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. The normalized intensity correlation between the beams $\{\mathbf{k}, -\mathbf{k}\}$ is defined as

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

where $S_{\mathbf{k},-\mathbf{k}}$ is the intensity correlation spectrum (defined as the Fourier transform of the correlation function

$C_{\mathbf{k},-\mathbf{k}}(\tau) = \langle \delta I_{\mathbf{k}}(t) \delta I_{-\mathbf{k}}(t + \tau) \rangle$) and $S_{\mathbf{k}}, S_{-\mathbf{k}}$ 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 $|C_{\mathbf{k},-\mathbf{k}}(\Omega)| \leq 1$ and is equal to 1 for perfect correlations and -1 for perfect anticorrelations.

One can have access to the normalized intensity correlation $C_{\mathbf{k},-\mathbf{k}}$ by measuring the noise spectra of the intensity sum S_+ , of the intensity difference S_- , and of each beam $S_{\pm\mathbf{k}}$. In fact, these quantities satisfy the following equation:

$$C_{\mathbf{k},-\mathbf{k}} = (S_+ - S_-) / 4\sqrt{S_{\mathbf{k}}S_{-\mathbf{k}}} \quad (2)$$

The normalized intensity correlation is plotted as a function of pump power in Fig. 4(b). One recognizes a clear threshold behaviour of the correlation coefficient: for low pump powers the correlation is weak, while it jumps abruptly at values close to 1 at ~ 15 mW of pump power. Above this value, the correlation does not exhibit a marked dependence on the pump power. By comparing Fig. 4(b) with the emitted average power for this series of experimental data (Fig. 4(a)), one can see that the correlation increases abruptly at the four wave mixing oscillation threshold. Such a high value of $C_{\mathbf{k},-\mathbf{k}}$ proves that polariton four wave mixing allows the generation of two macroscopic polariton populations that are strongly correlated. Note that the correlations cannot in any way be due to the fact that the two pump beams are produced by the same laser. In fact, we have carefully checked that the intensity fluctuations of the two pump beams are completely uncorrelated at the frequency of analysis of 4 MHz. This is due to the fact that the noise level of the pump laser is the standard quantum limit (at a sufficiently high frequency). Therefore, the pump beams cannot be at the origin of the correlations between the two emitted beams. Correlations are instead generated by the four wave mixing process.

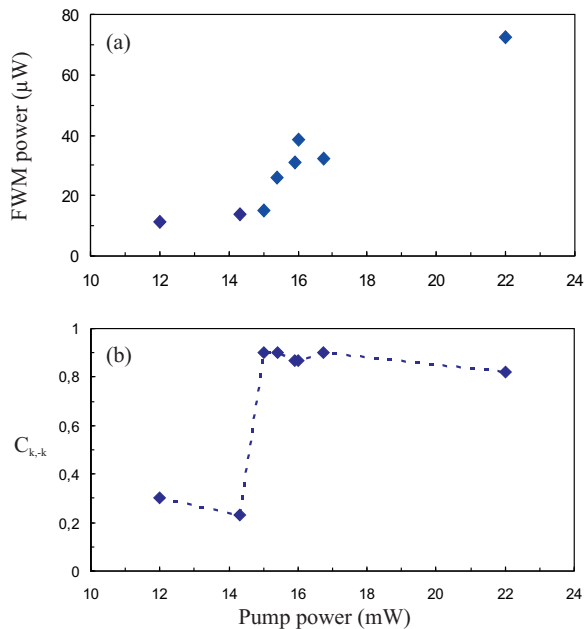


FIG. 4: (a) Four wave mixing optical power as a function of the pump power. (b) Normalized intensity correlation $C_{\mathbf{k},-\mathbf{k}}$ as a function of the pump power.

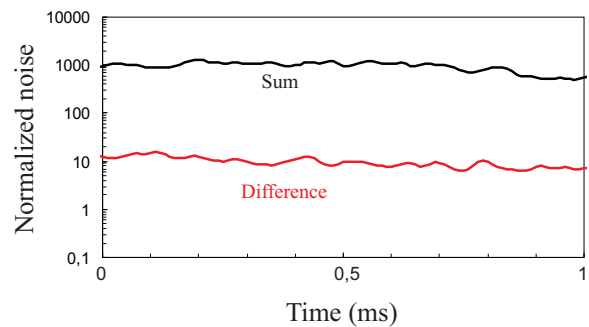


FIG. 5: Noise power of the photocurrent sum and of the photocurrent difference. The curves are taken at a fixed noise frequency of 4 MHz and normalized to the standard quantum limit.

In fig. 5, we show an example of the noise spectra of the sum and difference of the photocurrents. The curves are taken at a pump power slightly above the oscillation threshold. Since each individual beam is extremely noisy, a small unbalance can strongly affect the noise of the photocurrents difference; therefore, we have slightly attenuated the upper beam in order to compensate for the power unbalance and minimize the noise of the difference [22]. The photocurrent sum is extremely noisy (10^3 the standard quantum limit); on the contrary, the beams fluctuations almost perfectly cancel out when the photocurrents are subtracted, and difference noise is 10^2 times lower than the sum noise. The corresponding correlation coefficient is $C_{\mathbf{k},-\mathbf{k}} = 0.98$.

In conclusion, we have demonstrated a novel kind of polariton four wave mixing oscillation. In contrast with the so-called magic angle configuration, our scheme is based on the parametric interaction of two distinct pump modes with opposite wave vectors. We have shown that above the oscillation threshold, pump polaritons scatter towards final states orthogonally polarized with respect to the pumps, and localized around a well defined diameter of the elastic circle in the far field. Furthermore, we have demonstrated that the final states of the scattering process are strongly correlated. We want here to comment about the fact that, despite the extremely high value of $C_{\mathbf{k},-\mathbf{k}}$, the correlations are not *quantum* (since the difference noise is above the standard quantum limit). We believe that this fact may be interpreted as follows. As one can see in Fig. 4(b), the normalized intensity correlation $C_{\mathbf{k},-\mathbf{k}}$ is quite low below the oscillation threshold, and jumps abruptly at almost 1 just above threshold. Indeed, this is in contrast with the expected behavior for a parametric oscillator, which exhibits extremely strong correlations below threshold. This indicates that the few μW of optical power detected below threshold (see Fig. 4(a)) are not due to parametric luminescence below threshold; they come instead from other processes, which give an uncorrelated background emission, that becomes dominant at low pump power. Above threshold, this uncorrelated emission may reduce the observed correlations. If our interpretation is correct, present results may be improved in a configuration in which the FWM emission is not exactly at the same energy as the pumps [15], and therefore do not lie on the elastic circle, where the background emission is expected to be the strongest.

Our results indicate that the achievement of quantum correlated and entangled polaritons may be within reach in the near future.

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- [1] C. Weisbuch, M. Nishioka, A. Ishikawa, Y. Arakawa, "Observation of the coupled exciton-photon mode splitting in a semiconductor quantum microcavity", *Phys. Rev. Lett.* **69**, 3314 (1992).
 - [2] A. Kavokin, G. Malpuech, "Cavity polaritons", Elsevier, Oxford (2003); *Semicond. Sci. Technol.* **18**, S279-S434, Special Issue on Microcavities, edited by J.J. Baumberg and L. Viña (2003); *Phys. Stat. Sol. (b)*, **242**, (11), Special Issue: Physics of Semiconductor Microcavities, edited by B. Deveaud (2005).
 - [3] R. Houdré, C. Weisbuch, R. P. Stanley, U. Oesterle, P. Pellandini, M. Illegems, "Measurement of Cavity-Polariton Dispersion Curve from Angle-Resolved Photoluminescence Experiments", *Phys. Rev. Lett.* **73**, 2043 (1994).
 - [4] P. G. Savvidis, J. J. Baumberg, R. M. Stevenson, M. S. Skolnick, D. M. Whittaker, J. S. Roberts, "Angle-resonant stimulated polariton amplifier", *Phys. Rev. Lett.* **84**, 1547 (2000).
 - [5] C. Ciuti, P. Schwendimann, B. Deveaud, A. Quattropani, "Theory of the angle-resonant polariton amplifier", *Phys. Rev. B* **62**, R4825 (2000).
 - [6] J. J. Baumberg, P. G. Savvidis, R. M. Stevenson, A. I. Tartakovskii, M. S. Skolnick, D. M. Whittaker, J. S. Roberts, "Parametric oscillation in a vertical microcavity: a polariton condensate or micro-optical parametric oscillator", *Phys. Rev. B* **62**, R16247 (2000).
 - [7] R. M. Stevenson, V. N. Astratov, M. S. Skolnick, D. M. Whittaker, M. Emam-Ismael, A. I. Tartakovskii, P. G. Savvidis, J. J. Baumberg, J. S. Roberts, "Continuous wave observation of massive polariton redistribution by stimulated scattering in semiconductor microcavities", *Phys. Rev. Lett.* **85**, 3680 (2000).
 - [8] G. Messin, J.-Ph. Karr, A. Baas, G. Khitrova, R. Houdré, R. P. Stanley, U. Oesterle, E. Giacobino, "Parametric polariton amplification in semiconductor microcavities", *Phys. Rev. Lett.* **87**, 127403 (2001).
 - [9] A. Baas, J.-Ph. Karr, M. Romanelli, A. Bramati, E. Giacobino, "Optical bistability in semiconductor microcavities in the nondegenerate parametric oscillation regime: analogy with the optical parametric oscillator", *Phys. Rev. B* **70**, 161307(R) (2004).
 - [10] A. Heidmann, R. J. Horowicz, S. Reynaud, E. Giacobino, C. Fabre, and G. Camy, "Observation of quantum noise reduction on twin laser beams", *Phys. Rev. Lett.* **59**, 2555-2557 (1987).
 - [11] J.-Ph. Karr, A. Baas, E. Giacobino, "Twin polaritons in semiconductor microcavities", *Phys. Rev. A* **69**, 063807 (2004).
 - [12] P. Schwendimann, C. Ciuti, A. Quattropani, "Statistics of polaritons in the nonlinear regime", *Phys. Rev. B* **68**, 165324 (2003).
 - [13] M. Saba, C. Ciuti, J. Bloch, V. Thierry-Mieg, R. André, Le Si Dang, S. Kundermann, A. Mura, G. Bongiovanni, J. L. Staehli and B. Deveaud, "High-temperature ultrafast polariton parametric amplification in semiconductor microcavities", *Nature* **414**, 731 (2001).
 - [14] R. Butté, M. S. Skolnick, D. M. Whittaker, D. Bajoni, and J. S. Roberts, "Dependence of stimulated scattering in semiconductor microcavities on pump power, angle, and energy", *Phys. Rev. B* **68**, 115325 (2003).

- [15] S. Savasta, O. Di Stefano, V. Savona, W. Langbein, "Quantum complementarity of microcavity polaritons", *Phys. Rev. Lett.* **94**, 246401 (2005).
- [16] R. Houdré, C. Weisbuch, R. P. Stanley, U. Oesterle, M. Ilegems, "Nonlinear emission of semiconductor microcavities in the strong coupling regime", *Phys. Rev. Lett.* **85**, 2793 (2000).
- [17] K. V. Kavokin, P. Renucci, T. Amand, X. Marie, P. Senellart, J. Bloch, B. Sermage, "Linear polarization inversion: a signature of Coulomb scattering of cavity polaritons with opposite spins", *Phys. Stat. Sol.(c)* **2**, 763 (2005).
- [18] P. Renucci, T. Amand, X. Marie, P. Senellart, J. Bloch, B. Sermage, K. V. Kavokin, "Microcavity polariton spin quantum beats without a magnetic field: a manifestation of Coulomb exchange in dense and polarized polaron systems", *Phys. Rev. B* **72**, 075317 (2005).
- [19] G. Dasbach, C. Diederichs, J. Tignon, C. Ciuti, Ph. Roussignol, C. Delalande, M. Bayer, A. Forchel, "Polarization inversion via parametric scattering in quasi-one-dimensional microcavities", *Phys. Rev. B* **71**, 161308(R) (2005).
- [20] I. A. Shelykh, A. V. Kavokin, G. Malpuech, "Spin dynamics of exciton polaritons in microcavities", *Phys. Stat. Sol.(b)* **242**, 2271 (2005).
- [21] A. V. Kavokin, G. Malpuech, M. Glazov, "Optical spin Hall effect", *Phys. Rev. Lett.* **95**, 136601 (2005).
- [22] M. Romanelli, E. Giacobino, and A. Bramati, "Optimal intensity noise reduction in multimode vertical-cavity surface-emitting lasers by polarization-selective attenuation", *Opt. Lett.* **29** (14), 1629 (2004).