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Durnev, M.; Kavokin, A. V.; Yamamoto, Y.; Forchel, A.; Kamp, M.; Höfling, S.; Schneider, C.; Fischer, J.; Brodbeck, S.; Savenko, I. G.; Shelykh, I. A.; Chernenko, A.; Rahimi-Iman, A.; Kulakovskii, V. D.; Reitzenstein, S.; Kim, N. Y.; Amthor, Matthias

2014

Schneider, C., Fischer, J., Amthor, M., Brodbeck, S., Savenko, I. G., Shelykh, I. A., et al. (2013). Exciton-polariton lasers in Magnetic Fields. Proc. SPIE 8993, Quantum Sensing and Nanophotonic Devices XI, 899308-.

<https://hdl.handle.net/10356/98621>

<https://doi.org/10.1117/12.2038484>

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Exciton-Polariton Laser in Magnetic Fields

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ABSTRACT

Polariton lasers do not rely on stimulated emission of cavity photons, which sets stringent conditions on the threshold current in a conventional laser. Indeed, it has been demonstrated in optically pumped systems, that bosonic polariton lasers can outperform standard lasers in terms of their threshold power. The polaritons, which are part light and part matter quasiparticles, can undergo a condensation process into a common energy state. The radiated light from such a system shares many similarities with the light emitted from a conventional photon laser, even though the decay of the polaritons is a spontaneous process.

We discuss properties of polariton lasers and condensates in GaAs based microcavities. Special emphasis is given to the system's response to an applied magnetic field. We introduce the magnetic field interactions as a reliable tool to distinguish a polariton laser from a conventional photon laser device. In particular, we will discuss the first successful realization of an electrically pumped polariton laser, which marks a promising step towards the exploitation of polaritonic devices in the real world. We believe that our work can be extended to devices operated at room temperature by transferring the technology to large bandgap semiconductors, or even to GaAs samples with a modified layer design.

Keywords: Exciton-Polaritons, Bose-Einstein Condensate, Microcavity

1. INTRODUCTION

Quantum well (QW) microcavity exciton-polaritons are quasi-particles characteristic of the strong light matter coupling regime in a microcavity [1,2]. They are composite bosons, consisting of part light and part matter. When polaritons are formed, photons and excitons are hybridized, and their properties can be widely tailored to resemble a photon or an exciton. Being bosonic quasi-particles, they can undergo a dynamical condensation above a critical particle density [3–5]. Such a condensation in a solid state environment is an interesting topic on its own sake, and a long sought after goal for decades in the solid state community. Compared to uncoupled excitons, polaritons bear one major advantage: Due to the hybridization with photons, the effective polariton mass can be several orders of magnitude smaller than the electron mass [2], which simultaneously relieves the requirements on particle density and sample temperature. Indeed, in the meantime, condensation of exciton-polaritons in microcavities has been observed up to the room temperature in appropriate material systems, such as high-bandgap semiconductors (GaN, ZnO) [6,7] or organic compounds [8]. Via the excitonic part, the polaritons can interact rather strongly with each other, which facilitates the ground-state condensation via stimulated scattering. The condensate of polaritons is characterized by an off diagonal long range order (spatial coherence), it exhibits properties strongly related to superfluidity (frictionless flow) and the formation of vortices, vortex-pairs and solitons [9-10]. Furthermore, the exciton's spin degree of freedom adds further interest in the investigation of polariton condensates. In particular, the exciton spin components can be Zeeman-split in the presence of a magnetic field, which again results in a strong modification of the light polarization emitted from the condensate. The full paramagnetic screening of this Zeeman-splitting (the so-called "spin-Meissner effect") has been predicted in 2006 [11], however its fully consistent and conclusive experimental evidence is still missing. This is most likely a result of

the requirement for thermal equilibrium, which can hardly be satisfied in the polariton system as a result of the short lifetime. Nevertheless, several groups have reported strong non-linear effects in magnetooptical spectra of exciton-polaritons.

A full quenching of the Zeeman-splitting has been reported by Larionov et al. [12] and Walker et al. [13]. However, in both demonstrations, the effect was accompanied by anomalies in the polarization of the polariton condensate, leading to an alternative interpretation of the observed quenching. As we will show in this article, we have carried out magneto-optical experiments on a microcavity sample in the presence of an external magnetic field under optical and electrical pumping. We study the Zeeman-splitting, circular polarization degree and diamagnetic shift of the cavity polariton mode in the low excitation regime, polariton lasing regime and photon lasing regime above the Mott density in the QWs. All observed features in the experiment carried out under optical exciton injection can be qualitatively explained within the same model based on the assumption of thermal equilibrium within each of the polariton spin components, but no equilibrium between two spin components. Such a regime is likely to be observed in spinor bosonic systems where the energy relaxation rate strongly exceeds the spin relaxation rate.

The interest in electrically injected polariton condensates arises from the fact that exciton-polaritons can decay from the condensate by the leakage of photons from the microcavity, which produces monochromatic and coherent light. In contrast to stimulated emission in a weak coupling microcavity, fulfillment of the Bernard-Duraffourg condition (related to the non-equilibrium nature of population inversion via a splitting of the quasi-Fermi levels in a diode) are relaxed in the polariton system, which directly points out the possibility to build a coherent light emitting device with a very low threshold, and consequently with low power consumption [14].

Indeed, the basic capability of a polariton laser operating in the *strong coupling regime* to outperform a photon laser operating in the weak coupling regime has been indicated multiple times in optically pumped systems at cryogenic temperatures (including planar GaAs based microcavity samples [15,16] micropillar cavities [17], and photonic crystal polariton lasers [18]). To date, many milestone experiments towards the realization of functional electrically driven polariton devices were realized, including GaAs-based polariton light emitting diodes operating up to room temperature [19]. Here, we significantly extend these important accomplishments into the regime of polariton lasing. We can clearly discriminate our prototype device from a conventional VCSEL laser by studying its response to an applied magnetic field, yielding unambiguous evidence of the persistence of the strong coupling regime across the laser threshold.

This paper is organized as follows: Section 2 gives a description of the device fabrication technology and characterization procedure, both for optically and electrically injected polariton devices. Section 3 focuses on the investigation of optically generated polariton condensates in the presence of an external magnetic field. In section 4, we will demonstrate polariton laser operation under electric current injection. Conclusions are given in section 5.

2. DEVICE DESIGN AND CHARACTERIZATION

In this paper we present the results of studying two different samples, one designed for polariton lasing and condensation under optical pumping, and another one designed to facilitate electrical injection of polaritons.

2.1 Microcavity for optical pumping

The high-Q microcavity sample designed for optically pumped polariton lasing is a planar Fabry-Perot microresonator grown by molecular beam epitaxy. Four GaAs quantum-wells with a thickness of 13 nm, separated by 4nm thick AlAs layers, are integrated at the antinode of the optical eigen mode of a $\lambda/2$ AlAs cavity. Two further stacks of 4 QWs are integrated into the first eigen mode intensity maximum of the structure at the second mirror interface. The cavity is surrounded by AlAs/Al_{0.2}Ga_{0.8}As distributed Bragg reflectors with 23/27 mirror pairs in the top/bottom. A cross-

sectional scanning electron microscope image of the cleaved edge from such a sample is shown in Fig. 1a). The Q-factor of the sample under investigation is experimentally determined to exceed 10000 by etching micropillars into the sample and locally probing the linewidth from the cavity resonance. Such a high Q-factor enables us to pump the MC under non-resonant, continuous wave (CW) excitation at normal incidence. The excitation energy of the pump laser is set to the energy of the first interference fringe of the Bragg-reflectors. This is typically about 100meV above the exciton energy. In order to generate a comparably homogeneous polariton occupation over the area of the pump spot, we injected the carriers non-resonantly via a top hat shaped laser spot of 40 μm diameter. The sample temperature was set to $T = 5\text{K}$ to reduce the influence of phonons. The pump laser is linearly polarized, however it can be expected that the polarization properties of the polaritons in the condensate remain widely unaffected by the polarization of the pump laser due to multiple phonon scattering events in the relaxation process. To avoid heating, we used an optical chopper which produces 1 ms laser pulses. All optical experiments were carried out in a Fourier-space photoluminescence setup, similar to the one used by Lai et al. [20]. This allows us to directly extract the energy over k-dispersion of the polaritons. We use a $\lambda/4$ plate and a linear polarizer to select either the σ^+ - or σ^- polarized component of the emission.

2.2 Microcavity for electrical pumping

The sample for electrical injection of exciton-polaritons comprises a four-fold stack of 8 nm thick $\text{In}_{0.15}\text{Ga}_{0.85}\text{As}$ QWs, separated by 6 nm thick GaAs barriers. The QWs are integrated into an undoped one λ -thick (281 nm) GaAs cavity layer. The cavity is centered between 23 and 27 GaAs (64 nm) / AlAs (71 nm) mirror pairs in the top and bottom DBR layers, respectively. The doping in the mirrors was symmetrically tapered from 1×10^{18} to $3 \times 10^{18} \text{ cm}^{-3}$ in both the n-type and p-type doped sections, as schematically indicated by the grey scale variation in Fig. 1b). In the topmost two p-type mirror pairs, the doping concentration was ramped up to a value of $2 \times 10^{19} \text{ cm}^{-3}$. Delta doped layers (sheet density: 10^{12} cm^{-2}) were included at every second interface in both the top and the bottom DBR section to improve the electrical properties of the devices. The epitaxial wafer was patterned into circular pillar structures with a diameter of 20 μm by reactive ion etching. After sample planarization by benzocyclobutene, ring-shaped p-contacts (Ti-Au) were evaporated at the upper facet of the pillars. The close stack of four InGaAs QWs guarantees a homogeneous vertical carrier injection into each QW. The n-contact on the backside of the wafer consists of a AuGe-Ni-Au alloy (200 nm/70nm/500nm).

The Q-factor of the devices was experimentally determined via optical spectroscopy, it achieves 6300. This does not reach the empty cavity limit, which can have values in excess of 10000 also for doped structures with an equivalent amount of Bragg mirrors [21], but is, most likely, limited by the weak absorption tail of the QWs in the cavity.

Figure 1. a) Cross section electron micrograph of a polariton microcavity designed for optical pumping. The QWs are distributed in the intrinsic AlAs cavity layer in three stacks, each containing 4 QWs. **b)** Schematic drawing of the polariton laser device designed for electrical pumping. The DBR segments are doped by Si (n-type) and C (p-type), and the device is patterned into 20 μm diameter circles to avoid current spreading in the electrical pumping process.

3. OPTICALLY PUMPED CONDENSATE

3.1 Polariton condensation under optical injection

Figure 2. Energy-momentum dispersions at 0T (left columns) and 5T (right columns) in the linear regime (a), the polariton condensate (b) and the weak coupling laser regime (c). d)-f) The corresponding polarization resolved spectra at $k=0$, for magnetic field strength between 0T and 5T. The Zeeman-splitting can be resolved below threshold (d) and in the condensate (e), however not in the weak coupling laser regime (f).

We will first address the power dependent emission characteristics of the optically driven polariton cavity. Characteristic k -space resolved emission spectra of three fundamentally different emission regimes are plotted in Fig. 2a-2c). Each spectrum is normalized to maximum intensity to enhance visibility. In the spectrum in 2a) which is recorded far below any nonlinearity threshold of our system, one can observe a LP-dispersion with an exciton-photon detuning of $\Delta = E_c - E_x = -6.5$ meV. Applying a magnetic field of 5T (right side of Fig. 2a) results in a change of the detuning from 6.5 to -7.7 meV, along with the increase of the Rabi-splitting from 10.1 meV to 10.5 meV. Once the injection power is increased, the device can be driven into a second regime. The corresponding polariton distribution in the energy and momentum space is shown in Fig. 2b). One can observe a very significant narrowing of the linewidth and the angular dispersion of the polariton cloud. Furthermore, the energy ground state is slightly blueshifted with respect to the linear regime in Fig. 2a). In order to access the third emission regime, we have to apply pulsed optical excitation (50ps pulse width) to overcome limitations in the power of the excitation laser. Due to a congruence of the emission energy with the bare, uncoupled photon mode, we attribute this regime to weak coupling lasing (photon lasing) (Fig. 2(c)). The full input-output characteristic was recorded under pulsed excitation and is shown in Fig. 3. The three accessible regimes are distinctly separated by two non-linearities, which can be attributed to the transition from the linear polariton regime to the regime of polariton lasing characterized by the stimulated scattering towards a macroscopically occupied quantum state (the condensate), and from the polariton lasing regime into the weak coupling photon lasing regime.

Figure 3. a) Input-output characteristics of the cavity device, recorded under pulsed excitation. The three regimes are separated by two distinct thresholds.

3.2 Magnetic field properties of polariton condensates

Based on the aforementioned descriptions, we assess the properties of our optically driven polariton system in an external magnetic field which was applied in the Faraday configuration along the growth axis of the layered structure. In particular, we will focus on the Zeeman splitting of the ground state energy in the three different emission regimes.

We compare the dispersions of exciton-polaritons at 0T and B=5T (right) in Fig. 2a-c. We note a diamagnetic shift throughout the entire dispersion in the polaritonic regime (Fig. 2a and b). Due to the presence of the magnetic field, the exciton energy shifts by about 1.2 meV, and the Rabi-splitting increases to about 10.5 meV, which results in a change of the detuning from 6.5 to ~7.7 meV at 5T. Polarization resolved spectra of the ground-state are shown in Fig. 2 d-f). In fact, in the linear regime (Fig. 2d), we can only observe a small Zeeman splitting of the polariton mode as a result of the large photonic content of the polaritons, which increases approximately linearly with the applied magnetic field (magnified spectra are shown in the inset of 5T). In stark contrast, the behaviour of condensed polaritons is fundamentally different. As shown in Fig. 2e), the mode-splitting remains hardly observable up to a critical magnetic field (B_{crit}) and increases linearly for higher fields. In the regime shown in Fig. 2f), which we attribute to the weak coupling lasing, we cannot resolve any mode-splitting or diamagnetic mode shift. This strongly suggests that the polaritonic origin of the emission is lost, and that the stimulated emission of a photon laser generated by recombination of uncorrelated electron-hole pairs is the driving force in this regime. As we will discuss later in this article, this fundamentally different response of the different phases on the magnetic field is a supreme tool to distinguish between standard microcavity laser emission and polariton lasing.

We will now discuss the polariton emission in the magnetic field in detail. The extracted values for the Zeeman splitting in the linear phase and the polariton lasing phase are shown in Fig. 4a) as functions of the magnetic field. For the linear polaritons, the data can well be reproduced by the simple formula

$$E_z = g_{eff} \mu_B B \quad (1)$$

With g_{eff} as the effective g factor which depends critically on the exciton-photon detuning (and hence the magnetic field). Fitting yields a rather small value of $g_{eff} \sim 0.16$ as a result of the red detuning. In the polariton condensate, no Zeeman splitting can be unambiguously extracted up to a value of B~3T. For larger magnetic fields, the Zeeman-splitting changes its sign and magnitude significantly. The degree of circular polarization of the polariton condensate is shown in Fig. 4b for increasing magnetic fields, exhibiting a monotonous increase.

In order to quantitatively understand this anomalous behaviour of the polariton g-factor, we consider a condensate of two spin components, which are in thermal equilibrium, but the spin up system being out of equilibrium with the spin down system. This is justified by long spin relaxation times of exciton-polaritons in microcavities with a strong negative photon-exciton detuning as indicated in [22]. At zero temperature, the free energy in such a scenario reads:

$$F = -\mu_+ n_+ - \mu_- n_- + \frac{\alpha_1 + \alpha_2}{4} n^2 + (\alpha_1 - \alpha_2) S_z^2 - \mu_B g_{eff} B S_z \quad (2)$$

In this framework, μ_- and μ_+ denote the chemical potentials corresponding to the spin up (down) population n_+ (n_-). α_1, α_2 are the interaction constants for polaritons with parallel and antiparallel spins. $S_z = \frac{n_+ - n_-}{2}$ is related to the circular polarization of the condensate via $\rho = \frac{2S_z}{n}$.

Minimizing the free energy over S_z allows us to link the Zeeman splitting $\Delta = \mu_1 - \mu_2$ with the degree of circular polarization ρ via

$$\Delta = \mu_B g_{eff} B - (\alpha_1 - \alpha_2) n \rho \quad (3)$$

Based on this equation, we can qualitatively reproduce our experimental data (shown in Fig 4c) based on an assumed circular polarization degree (Fig. 4d) which nicely compares to the experimental data. In this quasi-equilibrium model the occupation of the Zeeman-levels is not governed by thermal equilibrium (such as in the original theoretical work by Rubo et al [11]) but it is determined by the degree of circular polarization (Fig. 4b). The observed competition between interaction blue-shifts of the individual Zeeman branches and the Zeeman splitting itself leads to the peculiar response of our out-of-equilibrium system. Note that while the concentration of polaritons n can hardly be obtained with a good precision, the term $(\alpha_1 - \alpha_2)n$ can be estimated from the blue-shift of the lowest energy peak of the polariton emission. Such estimation yields $\alpha_1 - \alpha_2 = 5.2 \times 10^{-12}$ meV cm² in our case.

Figure 4. **a)** Zeeman-splitting in the linear regime and condensate as a function of the magnetic field. **b)** Extracted degree of circular polarization. **c)** Theoretically reproduced splitting based on the circular degree of polarization shown in **d)**.

4 Electrically pumped polariton laser

In analogy to the previous chapter, we will first discuss power dependent emission characteristics from the electrically driven device. In this case, we study a polariton resonator with a diameter of $20\mu\text{m}$, etched into a post geometry (see chapter 2). Carriers are injected into the intrinsic cavity in the direct current (DC) mode, while the sample is held at a temperature of 8K. In order to stabilize the excitons in the InGaAs QWs, we apply an external magnetic field (5T in Fig. 5). The detuning of the selected device amounts to $\Delta = \sim -6.4\text{ meV}$, which is slightly larger than the Rabi-splitting of 6 meV (at 5T). We note, that in agreement with on the tendency observed in optically pumped devices, the polariton laser threshold is reduced at negative detunings due to the accelerated dynamics of polariton scattering into the ground state. The corresponding energy-momentum dispersions of the device are shown in Fig. 5a) for the linear regime, 5b) for the polariton lasing regime and 5c) for the regime which we relate to weak coupling lasing. While the emission in the linear regime is dominated by a strong luminescence from the bottleneck region at finite k -values, the shape of the dispersion dramatically changes once a threshold is overcome: In Fig. 5b, most emission is recorded from the energy ground-state, which indicates the onset of stimulated scattering in our device. The emission energy monotonously blueshifts until a second transition is crossed when it reaches the energy of the uncoupled photon. In this regime, the emission is dominated by resolution limited narrow resonances of high brightness, as expected for a conventional microcavity laser diode.

Figure 5. a) Energy-momentum dispersion characteristics of a polariton laser diode driven under CW conditions at an applied magnetic field of 5T, in the linear regime. b) The polariton laser regime, c) the weak coupling photon laser regime.

The input-output characteristic of the device is shown in Fig. 6a). The intensity was extracted by integrating over a momentum range of $k = 0 \pm 0.1\mu\text{m}^{-1}$. It features two pronounced non-linearities, corresponding to the qualitative transitions shown in Fig. 5. When we trace the energy of the ground-state as a function of the excitation current (Fig. 6b), we observe a blueshift of the polariton resonance until the second threshold is reached. These features are all consistent with our assumption that the strong coupling regime is preserved across the first non-linearity of the input-output characteristic. In order to unambiguously prove this assumption, we perform polarization resolved investigations at various excitation powers. As discussed in section 3.2, the extracted Zeeman splitting in the three regimes of our diode should fundamentally differ, and directly reflect the nature of the particles involved in the lasing process.

Figure 6. a) Input-output characteristics of the electrically pumped polariton diode, featuring two pronounced thresholds (recorded at 5T). b) Corresponding ground-state energy versus pump-current. The ground-state emission monotonously blueshifts until it pins to the bare cavity mode above the second threshold.

The waterfall diagram in Fig. 7a) shows polarization resolved spectra (σ_+ and σ_-) for varying excitation powers (all recorded at 5T). For the sake of visibility, each spectrum was normalized. The spectra which correspond to the two identified thresholds are indicated in the graph. In fact we reveal a persistence of the Zeeman-splitting of the emission feature across the first threshold, until the second threshold is crossed. The extracted values of the Zeeman-splitting as a function of the excitation power are plotted in Fig. 7b). Once the first threshold is crossed, the extracted values of the Zeeman splitting continuously decrease, which is most likely caused by an interplay of two effects: First, due to the continuous increase of B_{cr} with the density of polaritons, a reduction of the Zeeman splitting detected at the fixed magnetic field of 5T is expected (if the effective g -factor remains unaffected by the excitation power). Second, the transition into the weak coupling regime is continuous, and accompanied by a successive bleaching of excitons. This would, again, result in the decrease of Zeeman splitting due to the increasingly photonic nature of the mode which emits light.

Figure 7. a) Polarization resolved spectra at various injection currents. The continuous blueshift up to the second threshold, the linewidth narrowing above the first threshold and the persisting Zeeman-splitting beyond the first threshold is clearly seen in the data. **b)** Zeeman splitting dependence on the pump current density is an unambiguous prove of the polariton's hybrid nature.

5. CONCLUSION

We have reported on the magnetic field properties of polariton lasers, both in the optical and electrical injection modes. For optically injected polariton condensates, we have identified three fundamentally different operating regimes which can be attributed to a linear polariton emission, emission of light by a polariton condensate in the strong coupling regime and the photon laser emission in the weak coupling regime. Each phase responds fundamentally differently to an external magnetic field. In particular, in the important regime of polariton condensation, we have developed a non-equilibrium model which serves to qualitatively understand the anomalies of our experimental findings.

The characteristic non-trivial behavior of the Zeeman splitting in a polariton laser is a smoking gun for this lasing mode allowing to distinguish it from a standard VCSEL. We have demonstrated a first prototype of an electrically driven exciton-polariton laser [23]. In congruence with a similar report [24] it features a characteristic double threshold, an emission blueshift beyond the first threshold followed by a stable emission energy beyond the second threshold. Most importantly, we could directly monitor the persistence of the strong coupling regime through the first threshold via the characteristic Zeeman-splitting of the ground-state emission.

We believe that our study can be extended to room temperature polariton laser devices based on large bandgap semiconductors or sophisticated GaAs based structures.

ACKNOWLEDGEMENTS

The authors thank A. Wolf, M. Emmerling T. Süner, I. Lederer and A. Schade for technical assistance. The authors would like to thank the State of Bavaria, the National Science Foundation and by JSPS through its FIRST program for financial support. MD and AVK acknowledge support from the Russian Ministry of Science and Education (contract No. 11.G34.31.0067). AVC acknowledges support from the Russian Foundation for Basic Research. I.G.S. acknowledges support from the Eimskip foundation. I.A.S. acknowledges support from the 'Center of excellence in polaritonics', IRSES SPINMET and POLAPHEN projects. A.R.-I. acknowledges a German National Academic Foundation fellowship. The authors thank for experimental and technical support.

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