Research article

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Polariton lasing and energy-degenerate parametric scattering in non-resonantly driven coupled planar microcavities

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Abstract: Multi-level exciton-polariton systems offer an attractive platform for studies of non-linear optical phenomena. However, studies of such consequential non-linear phenomena as polariton condensation and lasing in planar microcavities have so far been limited to two-level systems, where the condensation takes place in the lowest attainable state. Here, we report non-equilibrium Bose–Einstein condensation of exciton-polaritons and low threshold, dual-wavelength polariton lasing in vertically coupled, double planar microcavities. Moreover, we find that the presence of the non-resonantly driven condensate triggers interbranch exciton-polariton transfer in the form of energy-degenerate parametric scattering. Such an effect has so far been observed only under excitation that is strictly resonant in terms of the energy and incidence angle. We describe theoretically our time-integrated and time-resolved photoluminescence investigations by an open-dissipative Gross–Pitaevskii equation-based model.

1 Introduction

Strong coupling between excitons and an optical mode leads to the formation of a hybrid quasiparticle called a polariton [1, 2]. The polaritons can be accessed and manipulated by their light component while the matter, excitonic component is responsible for the non-linear nature of the light–matter coupling and gives the polaritons the ability to interact.

In particular, the excitonic component is at the origin of polariton scattering processes, either of polariton–polariton type or related to interaction with the environment [3, 4]. In semiconductor microcavities, non-degenerate parametric scattering becomes efficient when a non-resonantly created high polariton density accumulates at the bottleneck region of the dispersion curve or when the resonant excitation is adjusted to the so-called magic angle of incidence [5]. The bosonic nature of polaritons enables their massive occupation of a single quantum state. Stimulated parametric scattering to the final state leads to such fascinating and consequential effects such as Bose–Einstein condensation of polaritons [6, 7], achievable at ultra-low excitation power and a higher temperature than in cold-atomic systems [8]. Photons emitted in radiative recombination of the condensate inherit its coherence and phase. By analogy to the conventional laser, the emission arising from the condensate is described by the term polariton lasing [9, 10].
Studies of polariton condensation and related effects such as spontaneous coherence, lasing, or superfluidity have increasingly become a focus of condensed matter and optics research in recent years. However, they have so far been mostly limited to single microcavities, either planar [7, 10, 11] or microstructured [12–16], and were related to intrabrancho, non-degenerate polariton parametric scattering.

Extension of studies of polariton-related non-linear phenomena to such effects as energy or momentum degenerate polariton parametric scattering [17, 18], localized to delocalized phase transitions of a photon [19], non-reciprocity based optical isolation [20, 21], parity-time symmetry breaking [22], or the generation of non-classical states of light [23, 24] requires implementation of a multiple-level polariton system. Such a system has so far been realized in a set of vertically coupled planar microcavities [18, 25] or a semiconductor microrod [26–28]. Offering a high degree of tunability, vertically coupled microcavities have been studied mostly in the linear regime [20, 25, 29–33], while the non-linear regime was addressed in the exclusive context of resonantly driven parametric scattering [17, 18, 34]. In particular, such fundamental effects, as polariton condensation and lasing, or parametric polariton scattering under non-resonant excitation, have so far not been observed and studied in a vertically coupled, planar microcavity system.

In this letter, we implement vertically coupled double planar microcavities, each embedding quantum wells (QW), to observe non-linear polariton phenomena under non-resonant excitation. In the two-level polariton systems, the fast relaxation from the upper to the lower branch precludes polariton condensation in the upper polariton state. Here, we obtain polariton condensation not only in the lowest, but also in the upper polariton branch. Emission dynamics measurements reveal that the population of two condensates, differing in energy and excitonic content, do not reach the maximum simultaneously at the same point in the structure. We introduce a theoretical model, assuming the existence of two polariton reservoirs, an active and an inactive one, which accurately describes both the time-integrated and time-resolved data. In particular, the model confirms different formation and recombination dynamics of the two condensates and indicates that the interplay between the relaxation and loss kinetics governs the condensation process. Moreover, we show that the polariton condensate created via non-resonant excitation in the upper polariton branch triggers energy degenerate parametric scattering from the bottom of the upper branch to a high wave vector within the lowest polariton branch. In this way, the condensate acts equivalently to a pump laser tuned to resonance with the bottom of the polariton branch.

2 Materials and methods

We investigate a molecular beam epitaxy grown sample with two \( \frac{3 \lambda}{2} \text{Cd}_{0.77}\text{Zn}_{0.13}\text{Mg}_{0.10}\text{Te} \) microcavities coupled via a semi-transparent distributed Bragg reflector (DBR) (see Figure 1a). The DBRs are made of alternating \( \text{Cd}_{0.77}\text{Zn}_{0.13}\text{Mg}_{0.30}\text{Te} \) and \( \text{Cd}_{0.43}\text{Zn}_{0.07}\text{Mg}_{0.50}\text{Te} \) layers lattice-matched to MgTe [33, 35–37]. Three quantum wells are placed at the anti-node of the electric field of each of the microcavities: 12 nm wide (Cd,Zn)Te:Mn QWs in the lower and 10 nm wide (Cd,Zn)Te QWs in the top microcavity. The microcavities and the DBRs are wedge-like, meaning that the absolute thickness of the layers of the sample changes when varying position across the sample’s surface, whereas the ratio of the thicknesses of all the layers stays constant. This allows us continuously to tune the energy of the optical modes in a 100 meV range.

Figure 1: (a) Sketch of a vertically coupled, double microcavity structure. The top and the bottom microcavities contain a set of three (Cd,Zn)Te or (Cd,Zn)Te:Mn QWs, respectively. (b) and (c) Cross-sectional energy-dispersive X-ray spectroscopy (EDX) images of the sample showing the spatial distribution of magnesium, the content of which controls the energy bandgap and refractive index of the layers. (d) A high angle-annular dark-field scanning transmission electron microscope image with EDX compositional mapping of magnesium in the region of the microcavity with three QWs visible.
wide range by adjusting the position on the sample surface while keeping the cavity–cavity coupling strength unaltered.

The results of the characterization of the structure by scanning transmission electron microscopy (STEM) are shown in Figure 1b–d. Consecutive close-up views of the microcavity structure present sharp interfaces of the DBR and QW layers, testifying to the high structural quality of the sample.

The sample is placed in a liquid-helium flow or pumped-helium cryostat and cooled down to 8 K or 1.5 K, respectively. The emission is pumped non-resonantly at \( E_{\text{exc}} = 1.72 \text{ eV} \) (\( \lambda_{\text{exc}} = 720 \text{ nm} \)) at normal incidence using a pulsed Ti:sapphire laser operating in femtosecond mode. The excitation power is adjusted using neutral density filters. We use a CCD or a streak camera combined with a grating spectrometer (1200 grooves/mm) as a detector for time-integrated and time-resolved measurements, respectively. The laser beam is focused onto the sample surface to a spot of 1–2 μm diameter, and the signal is collected with a microscope objective (NA = 0.7) or an aspheric lens (\( f = 3.1 \text{ mm}, \text{NA} = 0.68 \)) for time-integrated or time-resolved measurements, respectively. By adjusting a set of lenses, we switch between acquiring angle-integrated signal and Fourier space imaging. The in-plane photon momentum wave vector is recorded over a range of values up to 4.2 μm⁻¹. Reflectivity spatial mapping of the structure is performed using a halogen lamp as the light source, with a lens (focal length of 500 mm) shifted in the sample plane (step of 0.06 mm). The size of the light spot on the sample surface is 0.1 mm in this case.

### 3 Experimental results

Figure 2 shows the reflectivity spectra recorded for consecutive positions on the sample along the microcavities’ thickness gradient. The coupled, spatially delocalized optical modes of the structure \( C_s \) and \( C_{AS} \) shift in energy with the variation of the position on the sample, entering in resonance with consecutive excitonic transitions. The heavy-hole excitons, \( X_1 \) in the (Cd,Zn,Te) QWs and \( X_2 \) in the (Cd,Zn,Te):Mn QWs at 1640 meV and \( X_1 \) in the (Cd,Zn,Te):Mn QWs at 1644 meV, as well as the light-hole exciton \( X_{LH} \) in the (Cd,Zn,Te):Mn QWs at around 1663.5 meV are seen. The energies of the polariton levels are simulated within the framework of a coupled oscillator model (see Supplementary Note 1), as indicated by the white lines. The presence of strong coupling conditions is manifested in Figure 2 by the anticrossing of the polaritonic resonances [20, 38]. Matching of the calculated energies to the experimental ones allows us to determine the coupling constant \( \Omega_\parallel = 12 \text{ meV} \pm 0.3 \text{ meV} \) between the mode of the top microcavity and \( X_1 \), as well as \( \Omega_\parallel = 10.6 \text{ meV} \pm 0.3 \text{ meV} \) or \( \Omega_{1LH} = 9.8 \text{ meV} \pm 0.4 \text{ meV} \) between the mode of the bottom microcavity and \( X_2 \) or \( X_{LH} \), respectively. Modes of the top and bottom microcavity are coupled with a constant \( k_{1L} = 12.8 \text{ meV} \pm 0.2 \text{ meV} \), governing the \( C_{AS} \) and \( C_s \) energy separation. One should note here that, unlike in the case of the two-level polaritonic system, the map in Figure 2 shows a crossing of coupled (polariton) and uncoupled (optical modes and exciton states) levels. Such behaviour is a property of any system involving a number of polariton levels higher than two.

The non-parabolic dispersion of polariton branches observed in non-resonantly pumped photoluminescence (see Figure 3), recorded at a position on the sample of 0.7 mm (see Figure 2), further confirms the strong light–matter coupling regime in the studied structures. For excitation density \( P_{\text{exc}} = 0.8 P_{\text{th}} \) (Figure 3a), the emission from the bottom of the upper polariton branch at 1629 meV dominates the spectrum. The much weaker emission from the lower branch is stretched along the dispersion curve in its bottleneck region. It results from two coexisting effects – accumulation of polaritons scattered from active reservoirs and the Rayleigh scattering of polaritons from the bottom of the upper branch. When the excitation density is increased up to \( P_{\text{exc}} = 3.4 P_{\text{th}} \) (Figure 3b), a strong emission limited to the close vicinity of \( k_\parallel = 0 \) is observed from both the upper and the lower branch. Due to the relatively small size of the excitation spot and the resulting high density reservoir of photo-created carriers, the polaritons are partially ejected out of the bottom of the dispersion curves [39]. The qualitative change in the spectrum

![Figure 2](image-url)
shape at $k_\parallel = 0$ with increasing excitation power density is depicted in Figure 3c.

In order to trace in detail the impact of the excitation density on the emission properties of the studied structure, a systematic measurement of input–output dependence is performed with a focus on the emission at $k_\parallel = 0$ from the two lowest polariton levels. The emission intensity of both levels increases non-linearly by more than three orders of magnitude across the threshold at around $P_{\text{th}} = 25 \text{ kW cm}^{-2}$ (see Figure 4a). With further increase of the power density, the emission from the upper level saturates, while the signal from the lower level increases linearly. This behaviour is well reproduced using an open-dissipative model described in Section 4. Crossing the threshold is assisted by a narrowing (Figure 4b) and blueshift (Figure 4c) of the emission in the case of both levels. The higher exciton content in the polariton, the larger magnitude of polariton–polariton interactions, hence the blueshift of the more excitonic upper level is larger than that of the more photonic lower level. Such properties of the emission allow us to attribute the massive occupation of the bottom of the polariton branches shown in Figure 3 upon crossing the threshold $P_{\text{th}}$ to the effect of polariton condensation [7, 10, 40].

In the studied four-level system, polariton condensation and lasing occur at the two lowest levels, in contrast to the typical case of a two-level polariton system, where condensation takes place in the ground state, which is at the bottom of the lowest branch. Moreover, the condensation threshold for both the lower and upper polariton levels is comparable, unlike what was previously observed in ZnO microwire-based multimode systems [41]. As Figure 4a shows, tuning of excitation power allows for a steering of the relative emission intensity of the upper and lower levels. In particular, switching of the dominant line of the dual-wavelength polariton lasing from the lower to the upper branch is achievable.

To answer the question of how two condensates of different energy may coexist at the same point of the sample, we perform emission dynamics measurements. The temporal cross-sections of the spectra obtained under non-resonant excitation at the energy of the upper and lower polariton levels extracted from streak camera images are shown in Figure 5. For excitation density below $P_{\text{th}}$, the
population of the upper level (that with a higher excitonic content) builds up first and quickly decays. Once it vanishes, the population of the lower level (with a higher photonic content) builds up. The same behaviour is observed for excitation density above \( P_{\text{th}} \); however, the intensity of the lower level is much higher than that of the upper level, and an oscillatory character of the decay curve is observed. Thus, the condensates of different energy share a common threshold but do not emerge simultaneously.

The experimental data presented in Figure 5a and b are very well described by the calculated time transients shown, respectively, in Figure 5c and d, both in terms of temporal dynamics and intensity. It should be stressed that the same values of the parameters are used in the description of both time-integrated and time-resolved photoluminescence presented in Figures 4a and 5, respectively. The values of the parameters ensuring agreement with the experimental data are provided in Supplementary Note 2. The scheme showing the two lowest polariton energy branches along with the transitions included in the model is presented in Figure 5e.

Upon the formation of the polariton condensate in the upper level, a signal emerges at discrete points at high \( k_\parallel \) of the lowest polariton branch, as seen in the momentum-resolved emission visible in Figure 3b. These discrete points are distributed symmetrically around \( k_\parallel = 0 \), at \( k_\parallel = -4.1 \text{ \mu m}^{-1} \) and \( k_\parallel = 4.1 \text{ \mu m}^{-1} \), at an energy of 1630.5 meV, that is the energy of the condensate formed in the upper polariton level. We interpret the presence of such points as a manifestation of energy degenerate parametric scattering from the upper to the lower polariton branch, i.e. the annihilation of two polaritons with a wave vector \( k_\parallel = 0 \) and creation of two polaritons with wave vectors \( k_l \) and \( k_r \). Such a process fulfills the condition \( k_r + k_l = 0 \) while keeping the polariton energy unchanged. The scattering is facilitated by negative detuning conditions and the resulting relatively low exciton content in the polaritons, which diminishes the absorption. Point-like emission at \( k_l \) and \( k_r \) appears only for excitation density above \( P_{\text{th}} \) and is not observed below \( P_{\text{th}} \), as further described in Supplementary Note 3. This indicates that the presence of a polariton condensate is a prerequisite for the observed energy degenerate parametric scattering of polaritons. As shown in Supplementary Figure 2, the parametric scattering threshold exceeds slightly the polariton lasing threshold. Signals from the scattered pairs at \( k_l \) and \( k_r \) show the same threshold value. In the previous studies on planar coupled microcavities, energy degenerate parametric scattering was achieved by tuning the excitation to a strict resonance with the bottom of the upper polariton branch \([17, 42, 43]\). Our work shows, in contrast, that a non-resonantly driven polariton condensate provides an alternative to the previously used resonant pumping. The use of a non-resonant excitation for preparation of the condensate enables a clean observation of the scattering effects, thanks to the spectral separation of the excitation and the measured signal. It is also promising for implementation of robust, polariton-based entangled photon sources.
4 Theoretical model describing observed emission intensities

In order to describe the dependence of the emission intensity and dynamics on the excitation power, an open-dissipative Gross–Pitaevskii equation-based model (see Figure 5e), inspired by Refs. [44–46], is implemented. The model assumes that electron–hole pairs created by the non-resonant, pulsed excitation with equal efficiency in both microcavities [33] accumulate in an inactive reservoir.

From the inactive reservoir, they either decay or relax to an active reservoir in the high energy and momentum region of the lower and upper polariton dispersions [45, 47–50]. The polaritons then relax towards the bottom of a given branch. When the polariton density is high enough, stimulated scattering to the minimum of the polariton branch accelerates the relaxation and, eventually, induces the condensation of polaritons in the bottom of the branch. The model also includes a direct transfer from the upper to the lower polariton branch at \( k_\parallel = 0 \).

The inactive reservoir is described by the following rate equation:

\[
\frac{\partial n(t)}{\partial t} = - (\gamma_1 + r_U + r_L) n(t) + P(t),
\]

where \( \gamma_1 \) is the decay rate of the inactive reservoir population; \( n(t) \), \( r_U \) and \( r_L \) are the transfer rates from the inactive reservoir to the active reservoir in the upper (i.e. second-lowest) and the lower (the lowest) polariton branches, respectively. The \( P(t) \) is the pump in the form of a Gaussian pulse with a temporal width of 10 ps.

The active reservoirs \( n_{A_U}(t) \) and \( n_{A_L}(t) \) are the polariton populations accumulating in the high energy and momentum regions of the lower and upper polariton dispersion, respectively [45, 47–49]. The rate equations for \( n_{A_U}(t) \) and \( n_{A_L}(t) \) are:

\[
\frac{\partial n_{A_U}(t)}{\partial t} = - (\gamma_{A_U} + R_U |\psi_U(t)|^2) n_{A_U}(t) + n(t)r_U, \tag{2}
\]

\[
\frac{\partial n_{A_L}(t)}{\partial t} = - (\gamma_{A_L} + R_L |\psi_L(t)|^2) n_{A_L}(t) + n(t)r_L, \tag{3}
\]

where \( \gamma_{A_U} \) and \( \gamma_{A_L} \) are the decay rates, and \( R_U \) and \( R_L \) are the transfer rates of the active reservoir to the bottom of the upper and the lower polariton branches, respectively.

The polariton wave functions in the upper and lower branches are described by the following equations, where the gain is provided by the active reservoir population \( n_A \):

\[
i\hbar \frac{\partial \psi_U(t)}{\partial t} = \left( -\frac{\hbar^2 \nabla^2}{2m_U} + g_R n_A(t) + g_L |\psi_U(t)|^2 \right) \psi_U(t) - \frac{i\hbar}{2} \left[ \gamma_{C_U} - R_U n_{A_U}(t) + R_{UL} |\psi_L(t)|^2 \right] \psi_U(t),
\]

\[
i\hbar \frac{\partial \psi_L(t)}{\partial t} = \left( -\frac{\hbar^2 \nabla^2}{2m_L} + g_R n_A(t) + g_L |\psi_L(t)|^2 \right) \psi_L(t) - \frac{i\hbar}{2} \left[ \gamma_{C_L} - R_L n_{A_L}(t) - R_{UL} |\psi_L(t)|^2 \right] \psi_L(t).
\]

The factors \( \gamma_{C_U} \) and \( \gamma_{C_L} \) in Eqs. (4) and (5) are the radiative decay rates from the bottom of the upper and the lower polariton branches, respectively, while \( R_{UL} \) is the transfer rate from the upper to the lower polariton branch. The \( R_{UL} \) represents all the processes starting from the bottom of the upper polariton branch and finishing in the bottom of the lower polariton branch, either occurring with the conservation of the polariton in-plane momentum or not. Near \( k_\parallel = 0 \), the effective masses of polaritons related to the lowest two polariton branches are equal to \( m_U = 7.84 \times 10^{-2} m_e \) and \( m_L = 5.60 \times 10^{-2} m_e \). The values of the masses are obtained by fitting a parabolic dependence to the respective polariton dispersion for small values of the photon wave vector. The constants \( g \) and \( g_R \) denote the strength of the polariton–polariton and the exciton-polariton interactions, respectively. Assuming that the exciton–exciton coupling constant is \( g_{\text{exc}} \approx \frac{4\pi e^2}{c A} \) (Ref. [51]), we estimate the interaction constants for each polariton branch (as denoted by the respective indices) using the formula \( g_R = |x|^2 g_{\text{exc}} \) and \( g = |x|^4 g_{\text{exc}} \), where \( |x|^2 \) is the excitonic fraction of the respective polariton at \( k_\parallel = 0 \). In the case of non-resonantly pumped polariton condensates considered in this work, the interaction of polaritons with the exciton reservoir is stronger than the polariton–polariton interactions.

The emission intensity versus time following the excitation pulse of the upper and lower polariton branches (shown in Figure 5c and d) is calculated as \( \psi_C(t) |\psi_C(t)|^2 \) and \( \psi_L(t) |\psi_L(t)|^2 \), respectively. Integration over time of these intensities gives the time-integrated intensities (presented in Figure 4). A common factor accounting for the (unknown) excitation and detection efficiency of the experimental setup multiplies all the calculated intensities to ensure they match with the measured intensities. Very good agreement between the experimental data and the calculation is seen in Figures 4a and 5.

In view of our simulations, the relative temporal order of the emission from the bottom of the lower and upper branch is governed by the relationship between the rates
of transfer from the active reservoir to these levels, independently of the excitation power. Namely, emission from the lower level prior to emission from the upper level is induced by a higher transfer rate from the active reservoir to the lower level than to the upper level. The relative emission intensity ratio of the lower to the upper level is affected by both the transfer rates and the decay times of the polariton population of these levels, i.e. below the condensation threshold, the shorter the lifetime the stronger the emission of a given level for a reasonably wide range of values and ratios of the transfer rates. Above \( P_{\text{th}} \), the role of the transfer rates becomes dominant. In turn, the saturation of the emission intensity from the upper level above \( P_{\text{th}} \) results from the efficient transfer of the condensate from the upper to the lower level. A possible mechanism of the interbranch transfer at \( k_{||} = 0 \) is energy relaxation assisted by the emission of acoustic phonons or momentum degenerate polariton scattering [18]. Oscillations in the emission decay are a consequence of the presence of the inactive reservoir and competition in the polariton population built-up in the upper and lower polariton branches.

5 Conclusions and outlook

We have observed non-resonantly driven polariton condensation and lasing in a four-level system formed in a strongly coupled double microcavity structure. Emission from the two lowest polariton branches features non-linear behaviour with a common threshold in input–output characteristics. The time-resolved measurements reveal that the condensates at these two branches do not form simultaneously following the excitation pulse, but rather emerge and decay subsequently one after the other. We have introduced the open-dissipative Gross–Pitaevskii equation-based model that properly describes the time-resolved and time-integrated emission intensities and gives insight into the processes governing the polariton dynamics in a multilevel system. Moreover, we show that with the formation of the condensate in the upper of the two lowest branches, energy-degenerate polariton parametric scattering from the upper to the lower branch is launched.

A high degree of tunability of the investigated system should allow extension of the studies towards a variety of polariton related effects. In particular, three-dimensional polariton hopping, with a transfer occurring in two directions: parallel and perpendicular to the microcavities’ plane or temporal properties of energy- and/or momentum degenerate polariton pair scattering under non-resonant pumping might be addressed. Manganese doping of the QWs in one of the microcavities opens perspectives for a quite natural extension of the present study to spin-related phenomena, such as conservation of spin in parametric scattering, control of the polariton condensation using a magnetic field or non-trivial Berry curvature generation [52]. A microstructurization of the structure by either dry or focused ion beam etching would open exciting prospects for studies of non-linear effects in the dynamically developing field of cavity-polariton lattices [53, 54]. Apart from the novelty value for fundamental research, the present work is also of high importance for practical applications, e.g. a realization of tunable multi-wavelength sources of coherent light with an ultra-low lasing threshold or development of integrated, all-optical devices performing logic operations [55–58].

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