

Ultrafast dynamics of reconfigurable mode switching of polariton condensates revealed in a tunable ZnO microcavity

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Mode switching in exciton-polariton condensates represents a significant phenomenon that illustrates the non-Hermitian properties and out-of-equilibrium dynamics of exciton-polaritons. In this research, we experimentally demonstrate controllable mode switching in a tunable ZnO microcavity at room temperature, where increasing pump power drives the condensate from one lower polariton branch to another of higher energy. This unconventional up-switching is explained by an imbalance in the densities of the active excitonic reservoirs, governed by both loss and relaxation efficiency. Ultrafast spectroscopy reveals a several-picosecond delay between the condensates, providing direct evidence for this mechanism. Our results provide important insights into the fundamental physics of Bose-Einstein condensation in dissipative systems, enable control of polariton condensation through reservoir engineering, and open routes to room-temperature polaritonic devices exploiting nonlinear switching.

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I. INTRODUCTION

The strong coupling between excitonic states in semiconductors and photons confined within cavities leads to the emergence of exciton-polariton quasiparticles, which exhibit distinctive hybrid qualities of both light and matter [1,2]. These hybrid quasiparticles enable macroscopic quantum phenomena, such as Bose-Einstein condensation [3–5] and quantum fluids of light [6–8], and they pave the way

for the development of on-chip devices due to the interplay of excitonic nonlinearity and photonic coherence [9–13]. Room-temperature polariton condensates have been achieved in microcavities made from materials like GaN [14], ZnO [15], perovskite [16–18], and organic compounds [19,20]. Among these, ZnO stands out for its notable properties, including a high exciton binding energy (60 meV) [21], strong oscillator strength [22], and substantial Mott density. Beyond enabling room-temperature polariton condensation [23], ZnO-based microcavities facilitate intriguing observations, such as parametric scattering [24,25], quantum state manipulation [26], and polariton switching [27].

Numerous significant findings in polariton condensation involve precise control over the exciton reservoir and the mechanisms of polariton relaxation. A particularly remarkable phenomenon is mode switching, which is uniquely connected to the interplay between kinetics and thermodynamics during the condensation process [28,29]. Typically, when multiple lower polariton branches (LPBs) coexist in a cavity, polariton condensation transitions from the LPB

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at a higher energy to one at a lower energy as the pumping power increases [15,30], facilitating relaxation into the thermodynamics favored state through enhanced kinetics of polariton-polariton scattering [31]. Consequently, achieving anomalous up-switching for comprehensive control over polariton condensation dynamics would necessitate innovative physical mechanisms and advanced photonic techniques. Furthermore, insights into ultrafast dynamics related to the switching mechanism in condensation are still lacking.

In this work, we present experimental evidence of controllable switching between polariton condensates in a tunable ZnO microcavity at room temperature. In addition to the conventional down-switching behavior, we observe an anomalous switch of polariton condensation toward the high-energy branch as the pump power increases. We demonstrate that this unusual switching arises from an imbalance in the exciton density accumulation efficiency of the active reservoirs, manifested as a distinct picosecond-scale delay in the condensation dynamics between the two polariton branches. The flexibility of the microcavity system allows for fine-tuning between the two regimes of condensation dynamics, enabling full control over multibranch polariton condensation. Our findings reveal a fundamental mechanism whereby the efficiency of active exciton reservoirs acts as an additional degree of freedom in the polariton condensation process, which could be advantageous for future on-chip polaritonic devices leveraging nonlinear interactions.

II. RESULTS AND DISCUSSION

The microcavities were formed using one-dimensional ZnO microrods synthesized by chemical vapor deposition. ZnO powder (99.9% purity) and carbon powder (99.8% purity) were mixed in a 1:1 mass ratio and loaded into a quartz boat [32,33]. A silicon wafer was placed above the precursor mixture to serve as the growth substrate. The quartz boat was inserted into an open-tube furnace and heated from room temperature to 1050 °C over 60 min, followed by a 60 min dwell at the growth temperature. During the process, carbothermal reduction of ZnO generated Zn vapor, which reacted with oxygen in the furnace atmosphere to form ZnO and deposit onto the silicon substrate. After naturally cooling to room temperature, the substrate was removed, revealing dense, villouslike ZnO microrod structures.

The scanning electron microscope (SEM) image of the microrod under investigation [shown in the inset of Fig. 1(a)] reveals atomically smooth surfaces and a uniform hexagonal cross section, which support whispering gallery modes (WGMs), quasi-WGMs, and Fabry-Pérot (FP) modes [34–36]. In this study, the WGMs are the primary focus and are characterized by quantized energy levels in the azimuthal direction, defined by the azimuthal mode number N , along with parabolic dispersion in the axial direction. The strong coupling between excitons and cavity photons results in the formation of upper and lower polariton branches, with polariton condensation occurring at the energy minima of the LPBs [37,38]. Typically, hot excitons are generated from hot carriers excited by the pump, which then relax to the exciton reservoirs before further relaxing to the bottom of the LPB through polariton-phonon and polariton-polariton

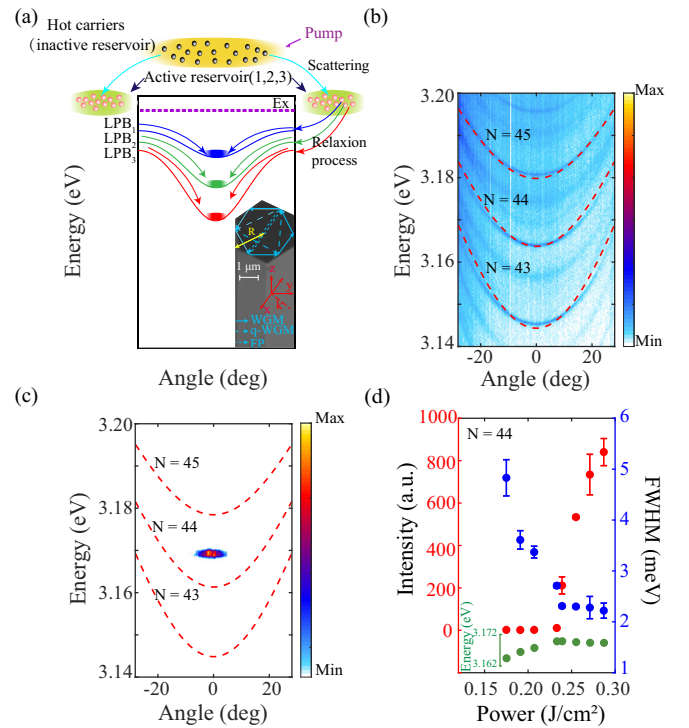


FIG. 1. (a) Diagram illustrating the nonlinearity of polariton condensates. The inset presents a SEM image of a typical ZnO microrod, alongside the optical modes within the ZnO microcavity, specifically WGM, q-WGM, and FPM. (b) Angle-resolved PL spectra in steady state acquired using a 325 nm continuous wave laser for excitation; the red dashed lines represent fitted dispersion curves for the WGMs. N represents the azimuthal number. (c) Steady-state angle-resolved PL spectra gathered from a 355 nm laser pump above the condensate threshold. In this context, N represents the number of modes. (d) The intensity at $k_{\parallel} = 0$ (solid red dots), its FWHM (solid blue dots), and energy blue shift (solid green dots) plotted against excitation fluence.

interactions. Specifically, in the presence of multiple LPBs, each LPB is associated with its own active exciton reservoir formed through relaxation from a significantly hotter common reservoir known as the inactive reservoir [39–42], as illustrated in Fig. 1(a). The thickness gradient of the microrod along the axial direction enables fine-tuning of the cavity resonances, mode spacing, and excitonic fractions for each LPB simply by adjusting the position of the pumping spot. The polariton condensation process is influenced by the interaction between thermodynamics and kinetics. The LPB with the most negative detuning is thermodynamically favored for Bose-Einstein condensate (BEC) due to its small effective mass linked to a high photonic fraction. However, this same branch is not kinetically favored, as the relaxation time from the exciton reservoir to the bottom of the LPB often exceeds the polariton lifetime, preventing condensation. Consequently, under a given pump power, polaritons condense at the optimal branch with intermediate detuning, arising from this intricate interplay between thermodynamics and kinetics [28,31]. Figures 1(b)–1(d) experimentally demonstrate this mechanism, showing multiple LPBs measured in steady-state angle-resolved photoluminescence (PL) spectra, with polariton condensation occurring on the optimal LPB corresponding

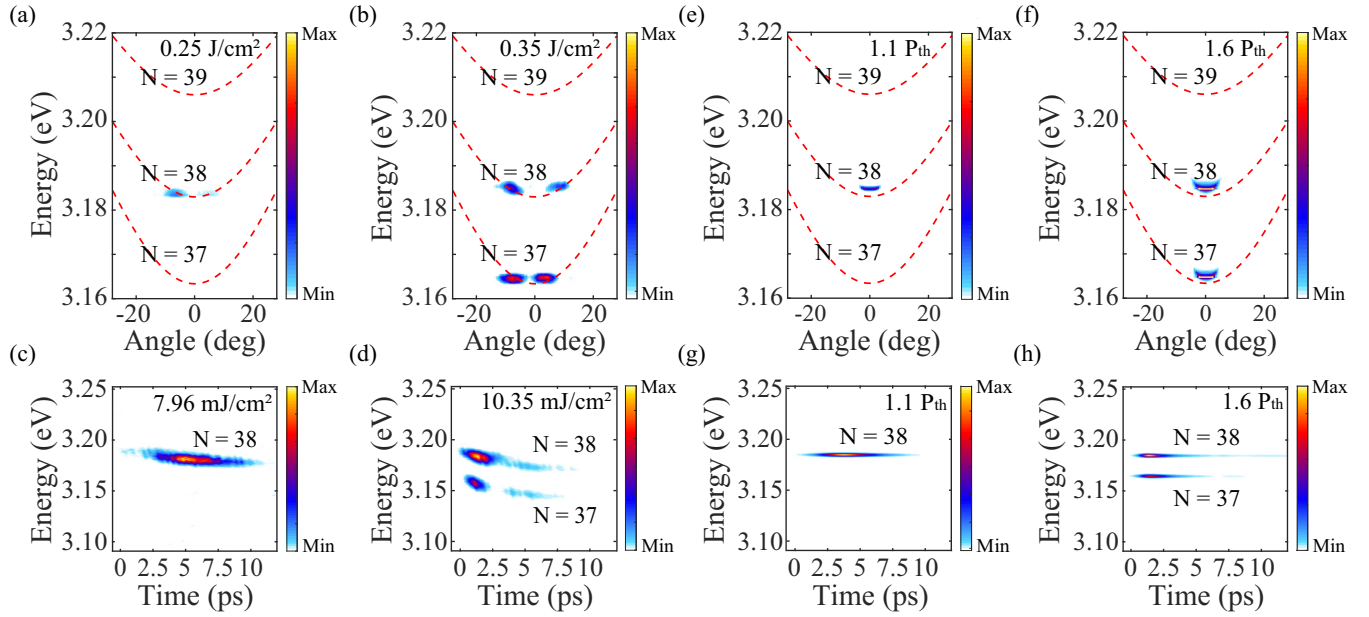


FIG. 2. Polariton condensate and the dynamics of various modes in scenario 1. (a), (b) Steady-state angle-resolved PL spectra obtained at excitation fluence of 0.25 and 0.35 J/cm², respectively. (c), (d) Angle-integrated PL distribution over time and energy at excitation fluence of 7.96 and 10.35 mJ/cm². (e), (f) Steady-state angle-resolved PL spectra obtained by numerical calculation. (g), (h) Temporal dynamics of polariton condensates for different branches obtained by numerical calculation.

to $N = 44$. The upper polariton branches are not clearly resolved in the angle-resolved PL spectra, consistent with previous reports [15,23,24,43,44]. Details of the experimental setup and the characteristics of polariton condensation are described in Appendix A.

An intriguing phenomenon is the switching between different branches of polariton condensation as the pump power increases. The underlying principle is the following: As the density of excitons in the reservoir increases, the rate of polariton-polariton interactions also escalates, resulting in a larger relaxation rate. This allows the polaritons to attain lower energy states within their finite lifetimes. Enhanced relaxation facilitates condensation at a more thermodynamically favorable LPB, which is characterized by a greater negative detuning and smaller effective mass. This effect has been confirmed experimentally in ZnO planar microcavities [15] and can be theoretically simulated by solving the density rate equations in conjunction with the Gross-Pitaevskii (GP) equation, with specifics provided in Appendix B. In these simulations, a crucial parameter τ is introduced in Eqs. (B2) and (B3), representing the efficiency of exciton relaxation from the inactive reservoir to each active reservoir, thereby indicating the exciton accumulation rate in the bottleneck region. To illustrate this standard mode switching, one must adjust the cavity length—either the circumference or radius of the microrod—to ensure an appropriate energy separation between the two LPBs. This separation must be neither too close, which would trigger simultaneous multiple lasing, nor too distant, which would make the lower LPB kinetically unreachable. Fortunately, this tunability is achievable due to the radius gradient of the microrod (see Supplemental Material [45]).

Figure 2 showcases a typical instance of conventional mode switching, with the microrod radius approximately

1.03 μm . At a relatively low excitation fluence of 0.25 J/cm², condensation occurs at the optimal LPB with $N = 38$. However, upon increasing the excitation fluence of 0.35 J/cm², the polariton condensation shifts to the $N = 37$ branch, as illustrated in Figs. 2(a) and 2(b). The “halo” shapes of the condensates result from the small pump point creating a polaritonic barrier at the center, causing the polariton condensate to acquire nonzero momenta [49]. It is worth noting that the higher lasing threshold at $+k$ compared to $-k$, as well as the momentum-space displacement of the $N = 37$ branch relative to $N = 38$, can be attributed to photonic disorder (due to sidewall roughness, tapering fluctuations, defects, refractive index inhomogeneity, etc.) along the tapered ZnO microrod (see related discussion in Ref. [15]). Time-resolved measurements were performed using femtosecond angle-resolved spectroscopic imaging (FARSI). The sample was excited by 350 nm pump pulses with a duration of 70 fs, generated from a Ti:sapphire laser system via frequency conversion. The PL signal at 800 nm was gated by a birefringent Kerr medium and focused through a 15 $\times$ objective onto the ZnO microcavity, enabling simultaneous subpicosecond temporal and momentum-resolved detection based on the transient Kerr effect. The results are presented in Figs. 2(c) and 2(d). Both LPBs display similar temporal dynamics, characterized by a pronounced redshift during condensation. Importantly, the experimentally observed redshift primarily arises from energy relaxation mediated by phonon emission (see discussion in Ref. [50]), a mechanism not captured by the GP model. Specifically, the condensations develop at nearly identical rates [shown in Fig. 2(d)] and decay according to the lifetime associated with each LPB. This similar growth rate signifies that the active reservoirs of both LPBs demonstrate comparable efficiency τ for polariton condensation, a behavior that

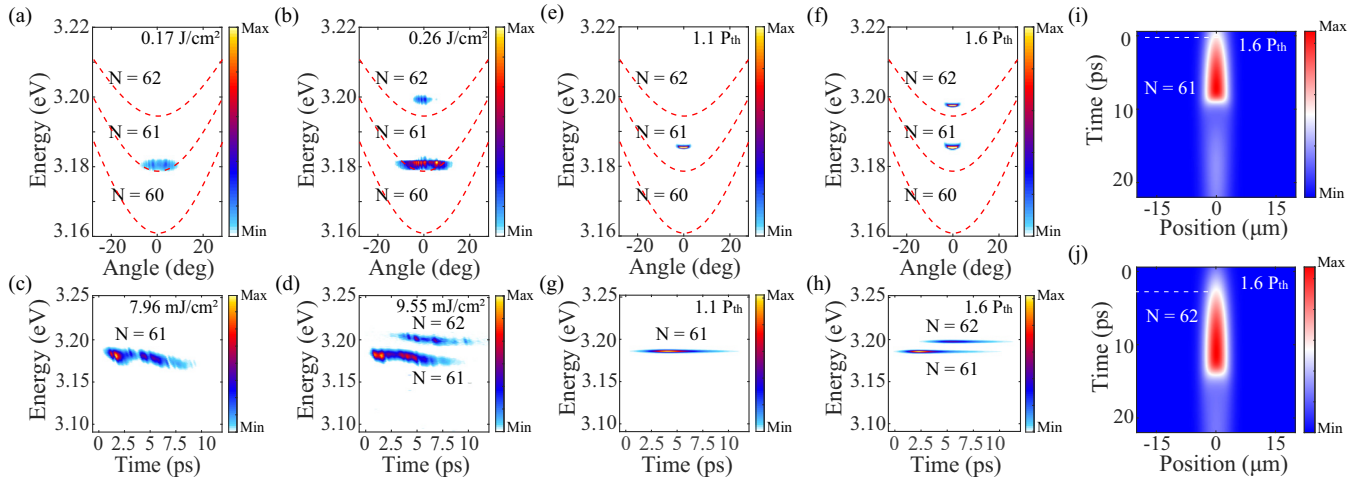


FIG. 3. The polariton condensate and the dynamics of various modes in the second scenario. (a), (b) Steady-state angle-resolved PL spectra recorded at excitation fluence of 0.17 and 0.26 J/cm², respectively. (c), (d) Angle-integrated PL distribution over time and energy at excitation fluence of 7.96 and 9.55 mJ/cm². (e), (f) Steady-state angle-resolved PL spectra obtained by numerical calculation. (g), (h) Temporal dynamics of polariton condensates across different branches obtained by numerical calculation. (i), (j) Temporal variation in the density of the active reservoir for $N = 61$ and $N = 62$ with different loss rates of $\gamma_c = 1 \text{ ps}^{-1}$ and $\gamma_c = 1.01 \text{ ps}^{-1}$.

theory also accurately predicts, as demonstrated in Figs. 2(g) and 2(h) through both steady-state angle-resolved and time-resolved analyses.

To date, our physical understanding of polariton condensation as a balance between kinetics and thermodynamics consistently indicates a down-switching behavior with increasing pump power. In reality, there have been few experimental or theoretical demonstrations of up-switching to a condensation that resembles a more excitonic state. To investigate this unexpected switching phenomenon, we adjusted our cavity to a different resonant condition with a microrod radius of 2.23 μm , with the measurement results illustrated in Fig. 3. We refer to this scenario as Cavity II, while the situation examined in Fig. 2 is designated as Cavity I. A comparison of the LPB dispersions is provided in Figs. 4(a) and 4(b). Notably, the $N = 61$ in Cavity II shares an energy level similar to the $N = 38$ LPB in Cavity I, suggesting that it is the most suitable branch for polariton condensation. Indeed, at a relatively low excitation fluence of 0.17 J/cm², only the $N = 61$ branch shows condensation, as depicted in Fig. 3(a). However, as the pump fluence increases, the behavior diverges from that observed in Cavity I. Instead of down-switching, polariton condensation shifts upward to $N = 62$ [Fig. 3(b)], characterized by a larger effective mass (indicating a greater excitonic fraction) and thus being thermodynamically less favorable.

To comprehend these findings, we must juxtapose the conditions in Cavity II with those in Cavity I. By analyzing the $N = 60, 61$, and 62 branches in Cavity II against the $N = 37, 38$, and 39 branches in Cavity I [as shown in Figs. 4(a) and 4(b)], it becomes apparent that the highest LPB of Cavity II ($N = 62$) is significantly less excitonic and much closer to the optimal LPB ($N = 61$) compared to that of Cavity I ($N = 39$). Concurrently, the lowest LPB of Cavity II ($N = 60$) is substantially more distant from the optimal LPB than Cavity I ($N = 37$). This observation provides a qualitative rationale: The thermodynamically favored state ($N = 60$) is too far from the exciton reservoir for down-switching to

occur, while the state with the largest effective mass can still achieve the critical density for BEC given sufficiently high pump power.

However, this simplistic interpretation only clarifies the preference of the highest branch over the lowest; it does not ensure that the transition from the middle (optimal) branch to the highest branch takes place. To gain a clearer understanding, we conducted theoretical calculations using the same model applied to Cavity II, assuming similar exciton densities for the active exciton reservoirs involved with both $N = 61$ and $N = 62$. The results, displayed in Figs. 4(c)–4(f), unexpectedly reveal that the $N = 62$ branch is predicted to be the optimal one instead of $N = 61$, and down-switching still occurs from $N = 62$ to $N = 61$, which starkly contrasts with our experimental findings. This discrepancy indicates that an undiscovered physical mechanism must be integrated into the theoretical framework to fully elucidate the observed switching dynamics.

To investigate the origin of the anomalous mode switching, we examine the properties of the active exciton reservoirs, with particular focus on the efficiency of exciton density accumulation. As illustrated in Fig. 1(a), nonresonant pumping first creates an inactive exciton reservoir, which subsequently relaxes into the active reservoirs associated with each LPB. Polariton condensation occurs once the active reservoir reaches a critical density that supports sufficient polariton population and relaxation via polariton-polariton scattering. Consequently, the rate of exciton accumulation in the active reservoir strongly influences the overall relaxation rate and the condensation dynamics. Two primary effects govern the accumulation rate of the active reservoir density n_A : (B1) the relaxation efficiency from the inactive to the active reservoir, characterized by τ in the rate equations, and (B2) the condensate loss rate, γ_c , in the GP equation, which sets the condensate density and feeds back to n_A through saturable gain [51]. The loss can originate from the cavity photon lifetime, nonradiative exciton decay, or exciton reabsorption. Crucially, the

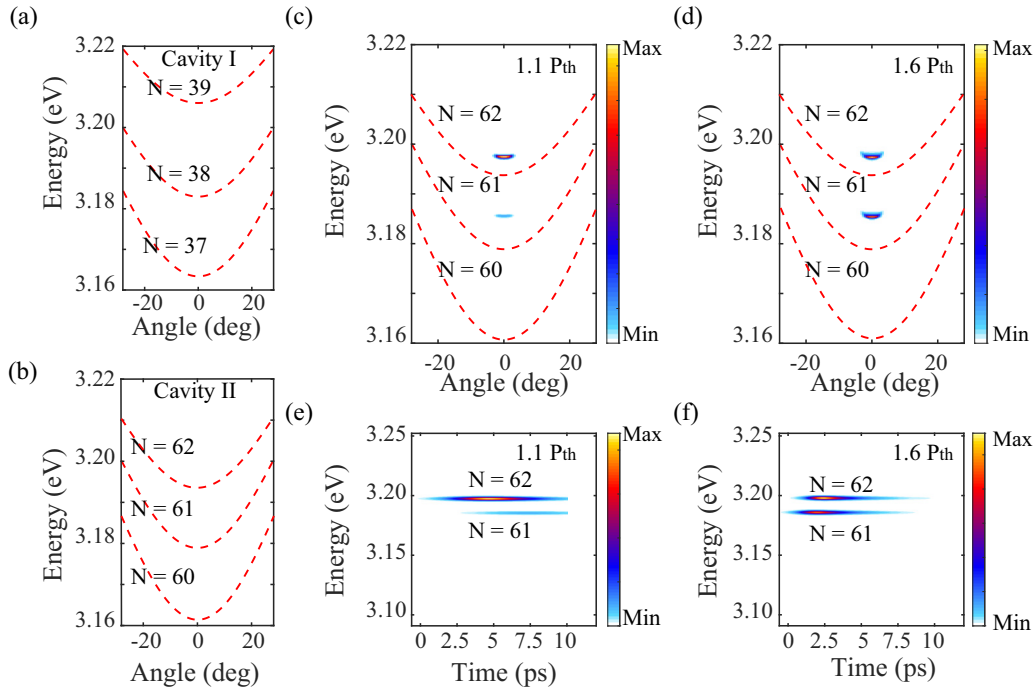


FIG. 4. (a), (b) The dispersion of the lower polariton branches in scenarios 1 (Cavity I) and 2 (Cavity II). (c), (d) Steady-state angle-resolved PL spectra for scenario 2 under conditions of identical relaxation rates (τ) for the LPBs ($N = 61$, $N = 62$), (e), (f) Temporal dynamics of polariton condensates corresponding to panels (c) and (d).

observed up-switching requires a disparity between the active reservoir densities of the two LPBs rather than assuming them to be identical. By introducing slightly different values of τ and γ_c for the two LPBs (see Table I in Appendix B), our simulations reproduce the experimentally observed up-switching between the $N = 61$ and $N = 62$ branches [Figs. 3(e) and 3(f)] and simultaneously yield a clear picosecond-scale delay in the buildup of n_A [Figs. 3(i) and 3(j)].

Notably, the imbalance of n_A leads directly to a measurable delay in polariton condensation, as revealed by time-resolved FARS measurements. Figures 3(c) and 3(d) show a clear delay of approximately 3.1 ps between the condensations on the $N = 62$ and $N = 61$ branches, providing direct evidence of the slower reservoir accumulation efficiency for $N = 62$. This interpretation is further supported by our theoretical calculations [Figs. 3(g) and 3(h)], which reproduce a comparable ~ 3.5 ps delay. Such an observable time delay cannot be explained without invoking an imbalance of the active reservoirs.

One key question is whether up-switching can occur if both branches have the same n_A . In theory, to maintain the lower branch as the optimal choice at relatively low pump power, a difference in n_A is needed. However, this necessary difference diminishes when the two branches are closer in energy, and simultaneously, the disparity in condensation thresholds between them decreases. Thus, if the cavity length is sufficiently large to allow for very small mode spacing, up-switching with balanced n_A becomes theoretically feasible according to the density rate equations that consider two active reservoirs. On the other hand, with extremely small mode spacing, numerous LPBs could exist around the optimal detuning, invalidating the model of only two active reservoirs. In this scenario, as

multiple branches share similar condensation thresholds, the system transitions into a regime characterized by multimode competition and multimode lasing presenting a situation entirely distinct from the up-switching process.

In summary, we have experimentally shown the reversible mode switching effect in exciton-polariton condensation. The unique up-switching behavior observed is linked to the uneven density to the active reservoirs, a finding strongly supported by ultrafast time-resolved spectroscopy. It is worth noting that a different type of up-switching behavior has been reported in organic microcavities, where switching is achieved by modifying the excitonic oscillator strength using an external electric field [52]. This mechanism is fundamentally different from the one studied here, as it does not support down-switching under the reported operating conditions. The connection between polariton mode switching and the density imbalance to different active reservoirs provides valuable insights into the fundamental physics underlying polariton condensation dynamics, which could significantly influence research on Bose-Einstein condensates of dissipative particles. The ability to reverse mode switching facilitates precise control over the dynamics of polariton condensation at picosecond scales, paving the way for innovative techniques in future polaritonic on-chip devices that leverage nonlinearity.

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TABLE I. Parameters used for numerical simulation.

LPB	$N = 37$	$N = 38$	$N = 61$	$N = 62$
Effective mass (m_e)	1.806×10^{-5}	2.367×10^{-5}	2.095×10^{-5}	2.566×10^{-5}
Relaxation rate (ps^{-1})	0.1	0.1	0.1	0.09
Loss rate (ps^{-1})	NA	NA	1	1.01

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Z.S. and F.L. initialized the project. C.H. fabricated the samples. M.Z., J.F., and F.P. performed the experiments and carried out the simulations. X.M. carried out the numerical modelling. M.Z., Z.S., F.L., and X.M. analyzed the data. All the authors contributed to the discussion of the results and wrote the manuscript.

The authors declare no competing interests.

DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

APPENDIX A: EXPERIMENTAL SETUP AND DEMONSTRATION OF POLARITON CONDENSATION

For steady-state experiments, we generated exciton-polaritons using two lasers: a 325 nm He-Cd gas laser (low-power) and a 355 nm pulsed laser (1.1 ns pulse width, 1 kHz rate) for high-intensity pumping. Polariton properties were measured using a custom microangular system with a $40 \times$ UV objective lens (NA = 0.47) and CCD-equipped spectrometer at room temperature.

APPENDIX B: THEORETICAL ANALYSIS AND REPRODUCTION

We utilized the Gross-Pitaevskii model to describe the dynamics of polariton condensates:

$$i\hbar \frac{\partial \Psi_j(\mathbf{r}, t)}{\partial t} = \left[E_j - \frac{\hbar^2}{2m} \nabla_{\perp}^2 - i\hbar \frac{\gamma_c}{2} + g_c |\Psi_j(\mathbf{r}, t)|^2 + \left(g_r + i\hbar \frac{R}{2} \right) n_{A,j}(\mathbf{r}, t) + g_r n_l(\mathbf{r}, t) \right] \times \Psi_j(\mathbf{r}, t) \quad (\text{B1})$$

Here, $\Psi_j(\mathbf{r}, t)$ is the polariton field with the index j representing different polariton branches (here, we consider two active polariton branches with $j = 1, 2$), E_j is the ground energy of the j -th polariton branch, m is the effective mass, $\gamma_c = 1 \text{ ps}^{-1}$ is the loss rate, $g_c = 6 \times 10^{-3} \text{ meV } \mu\text{m}^2$ is the polariton-polariton interaction, $g_r = 2g_c$ is the polariton-reservoir interaction, and $R = 0.1 \text{ ps}^{-1} \mu\text{m}^2$ is the condensation rate. To better characterize our specific system with condensation occurring in two lower polariton branches, we consider two types of reservoirs: the active reservoir $n_{A,j}$, providing direct gain to the j -th branch condensation, and the inactive reservoir n_l , being directly created by a nonresonant pump and allowing polaritons to relax to the active reservoirs. The dynamics of the reservoirs under nonresonant excitation satisfy the density equations:

$$\frac{\partial n_{A,j}(\mathbf{r}, t)}{\partial t} = \tau_j n_l(\mathbf{r}, t) - \gamma_A n_{A,j}(\mathbf{r}, t) - R |\Psi_j(\mathbf{r}, t)|^2 \times n_{A,j}(\mathbf{r}, t), \quad (\text{B2})$$

$$\frac{\partial n_l(\mathbf{r}, t)}{\partial t} = -\tau_l n_l(\mathbf{r}, t) - \tau_2 n_{A,j}(\mathbf{r}, t) - \gamma_l n_l(\mathbf{r}, t) + P(\mathbf{r}, t). \quad (\text{B3})$$

In this context, $\gamma_A = 0.1 \text{ ps}^{-1}$ and $\gamma_l = 0.01 \text{ ps}^{-1}$ represent the loss rates of the two reservoirs, while τ_j denotes the relaxation rate of polaritons transitioning from the inactive reservoir to two active ones. The pulsed pump $P(\mathbf{r}, t)$ used in the numerical simulations features a Gaussian distribution in both real space ($2 \mu\text{m}$) and time (10 ps).

For the down-switching mechanism, i.e., Fig. 2, we consider the parameters are the same but the effective masses, see Table I which are obtained from experiments. In this scenario, polaritons favor to further occupy the branch below the already condensed one, which has a smaller effective mass, as the pump intensity increases, and both condensations build up at almost the same time [see Figs. 2(e)–2(h)]. The up-switching mechanism with time delay between the two condensations is ascribed to the density of the active reservoir, including loss and relaxation rate. Here, we summarize the relaxation rates τ_j corresponding to the two condensations.

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