Spontaneous mirror-symmetry breaking in coupled photonic-crystal nanolasers

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Multi-cavity photonic systems, also known as photonic molecules, exhibit multi-well potentials that may prove useful for advanced quantum and nonlinear optics. A key phenomenon arising in double-well potentials is the spontaneous breaking of inversion symmetry, with a transition to two localized states in the wells, which are mirror images of each other. Although a few theoretical studies have addressed mirror-symmetry breaking in micro- and nanophotonic systems, no experimental evidence has been reported to date. Here, we demonstrate spontaneous mirror-symmetry breaking through a pitchfork bifurcation in a photonic molecule composed of two coupled photonic-crystal nanolasers. The coexistence of localized states is shown by switching them with short pulses. This offers exciting prospects for the realization of ultra-compact, integrated, scalable optical flip-flops. Analysis suggests that such symmetry breaking should be possible with a small number of intracavity photons and is thus suitable for quantum correlation devices.

Spontaneous symmetry breaking (SSB) unifies a range of physical mechanisms through which a given symmetric system ends up in an asymmetric state. It explains many central questions from particle and atomic physics to nonlinear optics (the Goldstone boson and the Higgs mechanism), phase transitions in Bose–Einstein condensates (BECs), metamaterials, bifurcations in lasers, photorefractive media, to mention just a few. Symmetry breaking may also lead to important technological advances, such as novel optical devices based on parity-time (PT) symmetry (see ref. 14 and references therein). In nature, in pyramidal molecules like ammonia, the most paradigmatic symmetry is spatial reflection in a double-well potential (DWP). SSB underlies transitions from delocalized to confined states within the wells. In photons, such a mechanism is possible provided that second-order nonlinearities overcome photon tunnelling. In this work we experimentally show SSB in a photonic molecule provided by two evanescently coupled photonic-crystal nanolasers. Switchable localized modes with broken mirror-symmetry will be demonstrated. This can be regarded as a nanoscale version of a laser flip-flop, in which the memory is incoherently pumped and there is no coherent beam driving the device, as in conventional bistability. This paves the way for the realization of ultra-small flip-flop optical memories based on SSB.

Our photonic molecule is a symmetric, light-emitting DWP given by two adjacent optical defects—or nanocavities—in a photonic-crystal lattice (Fig. 1a). The dynamics of the photonic molecule nanolaser with embedded semiconductor quantum wells is modelled using coupled-mode equations for the left (L) and right (R) cavity fields ($a_{L,R}$), normalized such that $|a|^2$ is the photon number:

$$\frac{da_{L,R}}{dt} = -i\omega_{L,R}a_{L,R} + \frac{1 + i\alpha}{4}V_s G(\mathcal{N}_{L,R})^2 a_{L,R} + \gamma L_{L,R} + \text{spont}$$

where $\omega = 1/\tau$ is the empty cavity field damping rate, $\omega_{L,R}$ are the resonant optical frequencies, $\alpha$ is the phase-amplitude coupling (Henry) factor of the quantum well laser, $V_s$ is the volume of the active medium, $G(\mathcal{N})$ is the carrier density-dependent gain function, $K$ is the inter-cavity evanescent coupling constant or photon tunnelling rate, $\gamma$ is the dissipative coupling or loss splitting rate, and ‘spont’ stands for spontaneous emission. We further assume that the dynamics of $\mathcal{N}_{L,R}$ is ruled by standard rate equations.

For identical cavities (that is, $\omega_{L,R} = \omega_{L,R}$), two eigenmodes exist, bonding ($a_{L}(t) = a_{R}(t)$) and antibonding ($a_{L}(t) = -a_{R}(t)$). In the laser regime, only the eigenmode minimizing optical losses is stable, usually the antibonding mode (threshold power, $P_T^{a}$). Neglecting the spontaneous emission for simplicity, the photon number will increase linearly above threshold as $|a_{L,R}(t)|^2 = |a_{0,L,R}|^2 + S_S^{a}$, where $S_S$ is a characteristic intracavity photon number and $S_S$ is the pump power. The laser regime begins at $\alpha = \alpha_{th}$. Here, SSB instabilities occur when $K$ is lower than a critical value $K_c (|K| < |K_c|)$, where $K_c$ is a characteristic intracavity photon number and $P_T^{a}$ is the pump power. It can be shown that SSB instabilities occur when $K$ is lower than a critical value $K_c (|K| < |K_c|)$, where $K_c$ is a characteristic intracavity photon number and $P_T^{a}$ is the pump power. In the absence of cavity detuning $K$ is related to the mode splitting ($\Delta \omega_{split}$) as $K = \Delta \omega_{split}/\delta$, where $\delta$ is the cavity linewidth, and $\alpha = 5$. Hence, a good DWP candidate for demonstrating SSB with low pump power has to fulfill the condition that mode splitting be on the order of the resonance width, that is, $K/\tau < 1$. Finely controlling the coupling strength in photonic molecules is thus key to achieving SSB. We implement such control—combined with efficient laser emission and free-space photon collection—by means of an original photonic-crystal cavity design, as explained in the following.

Our photonic-crystal molecule was formed from two evanescently coupled photonic-crystal L3 cavities (that is, with three holes missing in the FK direction of a triangular lattice) in a semi-conductor free-standing membrane (Fig. 1a,b). With the aim of realizing high-quality-factor ($Q = \omega_{th}/2$) cavities with improved beaming and controlled coupling strength, three design tools were
used: (1) the end holes of each L3 cavity were shifted and shrunk to increase the theoretical Q-factors up to a value of $\sim 10^5$ (blue holes in Fig. 1a,b); (2) the radii of neighbouring holes were modified to confine radiated photons within a $\sim 30^\circ$ emission cone$^{21-23}$ (red holes in Fig. 1a,b); and (3) the hole size of the central row of holes (green in Fig. 1a,b) was modified to control the coupling strength towards $K\tau \sim 1$ (refs 24,25). Both single and coupled cavities were etched on InP membranes containing InGaAs/InGaAsP quantum wells (see Methods). Resonant wavelengths were $\lambda \approx 1,540$ nm and measured Q-factors at transparency were $Q = 4,970$ ($\tau \approx 8$ ps) for single and $Q = 4,300$ ($\tau \approx 7$ ps) for coupled cavities. These Q-factors are lower than those given in (1), above, due to the strong beaming effect introduced in (2), and represent a good compromise between a low laser threshold and reduced onset for SSB ($\Delta P_{\text{c}} \propto Q$).

Mode splitting was measured by means of room-temperature microphotoluminescence spectroscopy. A continuous-wave (c.w.) pump beam was focused to a $\sim 2.2$-µm-diameter spot at the centre of the photonic molecule. Two modes could be observed: antibonding ‘AB’ (ground state) and bonding ‘B’ (excited state)$^{26,27}$. 

**Figure 1 | Photonic molecule**

a, Three-dimensional sketch of the photonic molecule composed of two coupled L3 photonic-crystal nanocavities (lattice period $a = 425$ nm; hole radius $r = 0.266a$). Embedded quantum wells are represented by a yellow line. b, Scanning electron microscopy image of the fabricated sample. Coloured circles highlight the modified holes. Blue holes ($r_{\text{blue}} = r - 0.06a$), shifted in the ΓK direction by $\Delta r_{\text{blue}} = 0.15a$, increase the cavity Q-factor. Red holes ($r_{\text{red}} = r + 0.05a$) improve beaming of the radiated photons. Green holes ($r_{\text{green}} = r - 20\%$) control the coupling strength. Orange holes ($r_{\text{orange}} = r_{\text{red}} - 20\%$) combine red and green effects. c, Split energy levels of the photonic molecule. Insets: far-field emission profiles (axes $k_x$ and $k_y$) of bonding (B, left) and antibonding (AB, right) modes.

**Figure 2 | Laser emission of coupled nanocavities**

a, Spectral measurements as a function of pump power when pumping at the centre of the photonic molecule. Both modes (B, short wavelength; AB, higher wavelength) are observed, but only AB undergoes laser emission. b, AB laser mode output versus input power. Green filled squares: experimental measurements obtained from the spectral peaks in a, for increasing input power. Black line: numerical solution of coupled laser rate equations. Black open square: pitchfork bifurcation. Coloured crosses: second broken parity state. Coloured open squares: first broken parity state. Coloured crosses: second broken parity state. Open circles: bifurcations leading to oscillations. Insets: illustrations of the DWP with the unique stable solution before bifurcation (left inset) and the two coexisting solutions after bifurcation (right inset).
The antibonding nature of the ground state in this molecule can be explained from the Bloch mode-effective refractive index of the underlying coupled W1 waveguide system (with two adjacent missing rows of holes in the TK direction). The effective refractive index of the antisymmetric mode is larger than the index of the symmetric mode, which is the opposite of the usual situation in standard geometries\(^5\). Far-field patterns showing intensity maxima (B) and minima (AB) at \(k = 0\) are shown in Fig. 1c. From a laser-mode splitting of 1.4 nm (Fig. 1c), the coupling parameter is found to be \(K = 3.3\) (see Supplementary Section IIA).

A solitary nanocavity laser exhibits an S-shaped output versus input power curve, with its sharpness related to the spontaneous emission factor \(\beta\). What is the expected behaviour for two evanescently coupled nanolasers when pumped at the centre of the photonic molecule? Of the two hybrid modes, the lasing mode is the one with lower optical losses (that is, AB), while the B mode is strongly attenuated (Fig. 2a). In Fig. 2b we present the AB maxima (green symbols) superimposed on the steady-state solutions of equations (1), with \(\beta = 0.017\) given by a fit of the experimental points (black line, see Supplementary Information). An S-shaped curve for the AB mode is observed up to \(P_p = 1.33P_{th}\). Within this range the solution is delocalized in the DWPs (Fig. 2b, left inset). Above this value two branches of steady-state solutions arise (plus a third, the destabilized AB mode), corresponding to two coexisting solutions: the 'high left cavity' together with the 'low right cavity' (L1R0' from now on) and conversely L0R1 (Fig. 2b, right inset). Unlike the AB mode, these new solutions have no defined parity: an SSB instability takes place at \(P_p = 1.33P_{th}\) in the form of a pitchfork bifurcation (Fig. 2b, black square). The two new branches (upper and lower) remain stable up to \(P_p = 1.37P_{th}\), where the system undergoes secondary instabilities (Hopf bifurcations), leading to ultrafast oscillations (predicted frequencies of \(\approx 150-180\) GHz depending on the pump power) that are larger and close to the tunnelling frequency of \(K/\pi \approx 148\) GHz. The former can be related to a.c. Josephson oscillations\(^4\). For a lasing AB mode in the presence of self-focusing nonlinearities (positive nonlinear refractive index above quantum well transparency), such an SSB scenario is only possible for a specific sign of the coupling parameter such that the lasing AB mode is redshifted (\(K > 0\)). The L3 photonic molecule implemented here meets this requirement.

The spectral measurements presented in Fig. 2 show a saturation of the integrated output laser power for \(P_p > 1.4P_{th}\), consistent with proximity to an SSB bifurcation. To experimentally investigate SSB, fast time detection with spatial filtering of individual cavity outputs was set up. Two identical avalanche photodiodes (APDs) coupled to single-mode optical fibres were used to collect left and right cavity signals simultaneously. The diffraction-limited collection area was smaller than the intercavity distance (\(d = 1.47\) µm) such that less than 10% crosstalk was observed (see Methods). The modulated pump beam (50 kHz, 30 ns rise time), impinging the sample with a peak power of \(\approx 6\) mW, was aligned at the centre of the photonic molecule.

Figure 3a presents a sequence of simultaneous outputs from both cavities. Segments of alternating 'low-red/high-blue' and 'high-red/low-blue' peaks are observed. We averaged out these time traces by superimposing events using a peak detection algorithm (see Methods). The results are shown in Fig. 3b. Two types of events are clearly identified: L1R0 and L0R1. These are plotted in Fig. 3b as a function of time and in Fig. 3c as a function of instantaneous pump power, together with a numerical integration of equations (1). The AB mode builds up from noise, with a laser threshold of \(P_{th} \approx 2.7\) mW, and increases up to \((P_p - P_{th})/P_{th} \approx 0.7\) \((P_p \approx 4.5\) mW), where the two distinct branches of output states appear. Note that the lower power branch, instead of decreasing monotonically as \(P_p\) increases, increases again for \((P_p - P_{th})/P_{th} > 1\). This is in good agreement with the fast oscillations observed in the
Numerical solutions, the amplitude of which increases with pump power (see Supplementary Information). Experimentally, oscillations at \( \sim 100 \) GHz are filtered out by the APD bandwidth and only their mean value is measured.

A most important question is whether our scenario corresponds to SSB or to explicit symmetry breaking induced by small changes in a control parameter. First, cavity-to-cavity detuning can be neglected in our photonic molecule, because it is smaller than the experimental error (\( \Delta \omega / 2 \pi < 12 \) GHz). Next, by adding Langevin noise into equations (1) we verify that spontaneous emission dominates over a relative pump-cavity spatial jitter as large as 500 pm (see Supplementary Information). Within the experimental bandwidth of SSB experiments (100 Hz–30 MHz), we measured \( \sim 70 \) pm spatial jitter on the cavity plane (see Methods), on the order of the shot noise limit. As a result, fast (\( \sim 50 \) kHz) jumping between states is most probably due to spontaneous emission fluctuations in the nanolasers. The only way to experimentally prove that our scenario indeed corresponds to SSB, that is, that the L1R0 and L0R1 states do coexist, is to be able to switch from one to the other within the 30 ns pump ramp, as will be done in the following.

An additional short (\( \sim 100 \) ps) pulse laser, synchronous with the pump beam modulation, is focused into either cavity while the position of the pump beam is kept fixed at the centre of the photonic molecule. Figure 4a shows the initial (non-perturbed, top) situation. As the short pulse is aligned at the left cavity, two cases are observed: either the intensity in the left cavity was low before the arrival of the short pulse (that is, the L0R1 state anticipated the pulse perturbation), in which case L0R1 survives (Fig. 4a, centre, continuous lines), or the L1R0 state anticipated the pulse perturbation, in which case L0R1 switches to L1R0, while L1R0 remains unchanged. Such a switching mechanism relies on carrier saturation in the high-intensity cavity due to stimulated emission, so only an increase in carrier density induces a power drop-off, simultaneously raising the power in the other cavity. In the theoretical model, the short-pulse excitation is simulated by adding a Dirac delta perturbation in the carrier variables, resulting in a step-like local increase in the carrier density.

When the perturbation laser is shifted to the right cavity the situation is reversed: L0R1 switches to L1R0, while L1R0 remains unchanged. Such a switching mechanism relies on carrier saturation in the high-intensity cavity due to stimulated emission, so only an increase in carrier density induces a power drop-off, simultaneously raising the power in the other cavity. In the theoretical model, the short-pulse excitation is simulated by adding a Dirac delta perturbation in the carrier variables, resulting in a step-like local increase in the carrier density.

The short-pulse laser can also be used to stabilize spontaneous jumps such that photon trapping can be measured with a slower two-dimensional detector (Fig. 4b), further illustrating photon confinement around the cavities. This demonstration of coexistence via optical switching constitutes an experimental proof of SSB in the photonic molecule. Switching mirror images is a clear advantage in the context of optical memory applications (that is, optical flip-flops\(^7\)). Compared to microdisk-laser photonic molecules\(^26\), the InP-based photonic-crystal molecule benefits from a smaller footprint and potentially lower energy consumption\(^15\). In addition, the SSB mechanism circumvents the need for a coherent driving beam in standard injection-locked laser memories\(^35\). Note that different applications emerge from PT symmetry breaking in which only one mode survives\(^39\), for instance the single-mode laser in a microring\(^31\). Pitchfork bifurcated states are bistable, which clearly assesses the memory capabilities of our photonic molecule device. Ultimately, electrical pumping would both improve performance and allow independent control of cavity resonances\(^31\).

In the present experiments, the inferred photon number in each cavity at the onset of SSB is \( |a|^2 \approx 100 \) (the normalization photon number is \( S_{\text{norm}} \approx 135 \), see Supplementary Information), which is compatible with the signal level measured by the APD detectors (see Methods). Shrinking the middle row hole radius \( r_{\text{open}} \), by just a few percent would result in a reduced mode splitting, from \( K_T = 3.3 \) to, for example, \( K_T \approx 0.7 \). The bifurcation point would then decrease to \( \Delta P_c = K_T / \alpha \approx 0.1 \). This means that the pitchfork bifurcation point would be shifted towards the laser threshold. In such conditions, the photon number in each cavity, \( |a|^2 \approx S_{\text{norm}} \Delta P_c \), becomes \( \sim 10 \) at the SSB instability. Quantum interference in photonic molecules in the presence of (even modest) nonlinearities is expected to leave its fingerprints on the quantum correlations of the laser photons\(^4\). This photonic-crystal molecule might then constitute a building block for a new class of light-emitting nanosources with strong photonic correlations.

**Methods**

**Sample and set-up description.** A 265 nm InP membrane containing four InGaAs/InGaAsP quantum wells (photoluminescence centred at \(-1.510 \) nm, FWHM = 63 nm) was bonded to a silicon substrate by means of a 400 nm benzocyclobutene layer. After standard photonic-crystal fabrication, chemical etching provided a suspended membrane (1 \( \mu \)m air gap). The photonic molecule was pumped at \( \lambda = 808 \) nm with a c.w. single-mode fibre-coupled laser diode (Lumics L808M100), modulated by a 120 MHz (Agilent N150A, minimum rise time \( \sim 2 \) ns) waveform generator (pulses of a few tens of nanoseconds, repetition rates from 10 to 200 kHz). A \( \times 100 \) magnification, 0.95 numerical aperture (NA) and infrared antireflection-coated microscope objective (Olympus MPLAN \( \times 100 \) IR) was used to focus the pump to a 2.2-\( \mu \)m-diameter spot on the sample. Its relative position with respect to the cavities was adjusted (1 nm resolution) by means of a nanopositioning stage.
The sample was removed and the microscope objective replaced by an pump beam stability. Perturbation pulse. Averages were subsequently performed (Fig. 4). Restricted to a time window running from the pump pulse to the occurrence of the statistical that beyond 150 kHz mechanical noise can be neglected and only unavoidable quadrant position detector (Thorlabs PDQ80A, 150 kHz bandwidth). We assume lens. The pump beam displacements were measured at its focal plane with a Taking into account the focal length of the microscope objective (1.8 mm), the in the SSB experiments.

peak detection algorithm. Time traces in each channel were recorded simultaneously using a 13 GHz bandwidth oscilloscope (Lecroy WaveMaster 813Zi) in the form of a sequence of 50 consecutive, 50 ns time windows (one output pulse per window); 500 pulses were then recorded for each cavity in one shot. Under SSB conditions, two types of event were observed: a high pulse in cavity L (Ch1) and a low pulse in cavity R (Ch2), called L1R0, or a low pulse in cavity L (Ch1) and a high pulse in cavity R (Ch2) (L0R1). Such states appear either in long clusters (up to hundreds of pulses) or in segments of rapidly alternating L1R0–L0R1 events (few tens of pulses). We associate long clusters to a small long-lived drift (typically due to mechanical vibrations). Segments with alternating events (600 μs duration, 31 pulses, 50% L1R0 and 50% L0R1), picked up in Fig. 3a, are attributed to spontaneous switching. A peak detection algorithm with a threshold (75% of the peak amplitude) was implemented in Ch2 to discriminate L0R1 and L1R0. Averages were performed over each type of event (Fig. 3b). When applying a short (~100 ps) perturbation pulse for the demonstration of coexistence, peak detection was restricted to a time window running from the pump pulse to the occurrence of the perturbation pulse. Averages were subsequently performed (Fig. 4).

Pump beam stability. The spatial jitter of the pump beam was measured as follows. The sample was removed and the microscope objective replaced by an f = 2,000 mm lens. The pump beam displacements were measured at its focal plane with a quadratic position detector (Thorlabs PD300A). 150 kHz bandwidth. We assume that beyond 150 kHz mechanical noise can be neglected and only unavoidable statistical fluctuations are present. Within the bandwidth of the experiment (100 Hz–30 MHz) we found a main peak of mechanical noise at ~300 Hz with an r.m.s. displacement of ~70 nm, corresponding to an angular fluctuation of ~35 rad. Taking into account the focal length of the microscope objective (1.8 mm), the spatial jitter of the pump beam on the photonic molecule was found to be ~65 pm in the SSB experiments.

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Author contributions
A.L. and A.M.Y. conceived and designed the experiments. P.H. and S.H. performed the experiments. F.R.G. and I.S. fabricated the samples. P.M. provided technical assistance. A.M.Y. performed theoretical analysis and numerical simulations. A.L., P.H. and A.M.Y. co-wrote the paper.

Additional information
Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to A.M.Y.

Competing financial interests
The authors declare no competing financial interests.