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## Transient dual-energy lasing in a semiconductor microcavity

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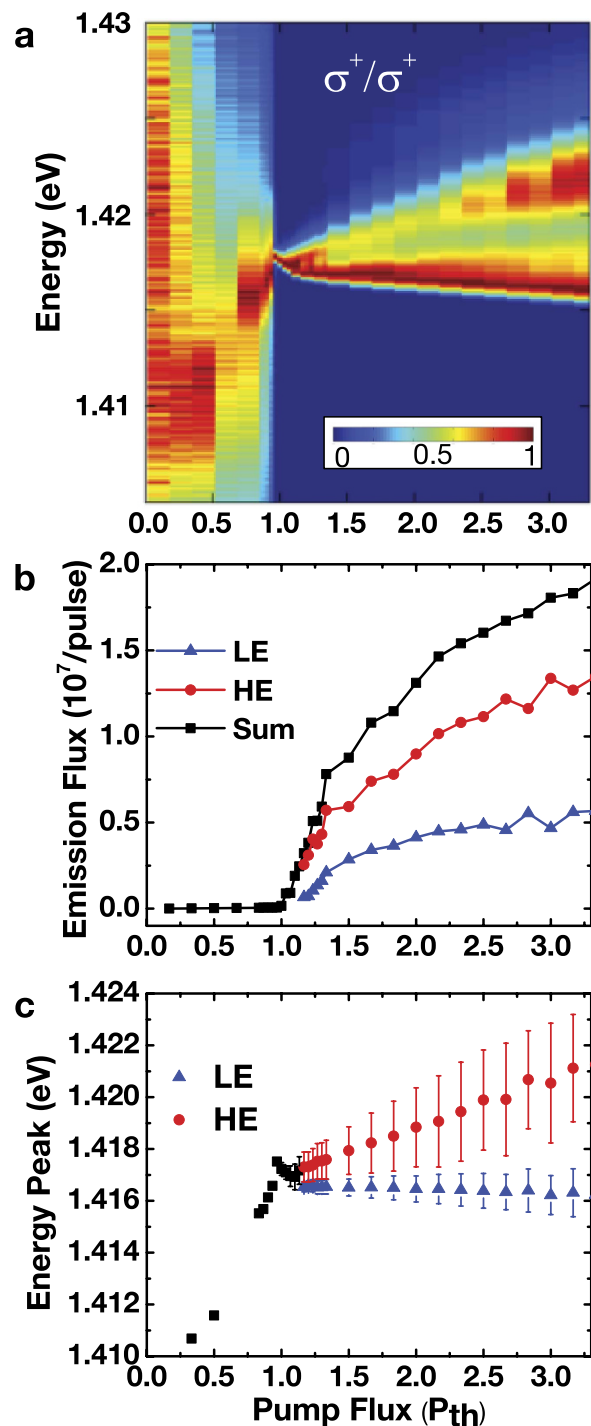
**We demonstrate sequential lasing at two well-separated energies in a highly photoexcited planar microcavity at room temperature. Two spatially overlapped lasing states with distinct polarization properties appear at energies more than 5 meV apart. Under a circularly polarized nonresonant 2 ps pulse excitation, a sub-10-ps transient circularly polarized high-energy (HE) state emerges within 10 ps after the pulse excitation. This HE state is followed by a pulsed state that lasts for 20–50 ps at a low energy (LE) state. The HE state is highly circularly polarized as a result of a spin-preserving stimulated process, while the LE state shows a significantly reduced circular polarization because of a diminishing spin imbalance.**

The spectral characteristics of a conventional semiconductor laser are typically fixated to static composition structures<sup>1–4</sup>. As a result, the lasing energy is generally single-valued, with a sub-1-meV linewidth. Nevertheless, a simultaneous two-state lasing effect with well-separated lasing wavelengths has been demonstrated in quantum dot lasers<sup>5–7</sup> and nanocrystal lasers<sup>8</sup> when the intraband carrier relaxation between the ground and excited states quasi-0D nanoscale gain medium is finite. However, the lasing energies in these quantum-dot lasers are still fixated to a static cavity structure. Another essential characteristic of a semiconductor laser is polarization. In edge-emitting semiconductor lasers, laser radiation is typically linearly polarized as determined by the polarization-dependent reflectivity of the cavity. By contrast, in vertical-cavity surface-emitting lasers (VCSELs), the lasing polarization is inherently unstable and is affected by gain or crystalline anisotropy<sup>9,10</sup> of the gain medium or cavity. VCSELs typically give laser radiation linearly polarized either along the [110] or [110] crystallographic direction, but can exhibit complex polarization switching<sup>10–12</sup> and bistability<sup>13,14</sup> effects depending on the cavity structure and carrier density.

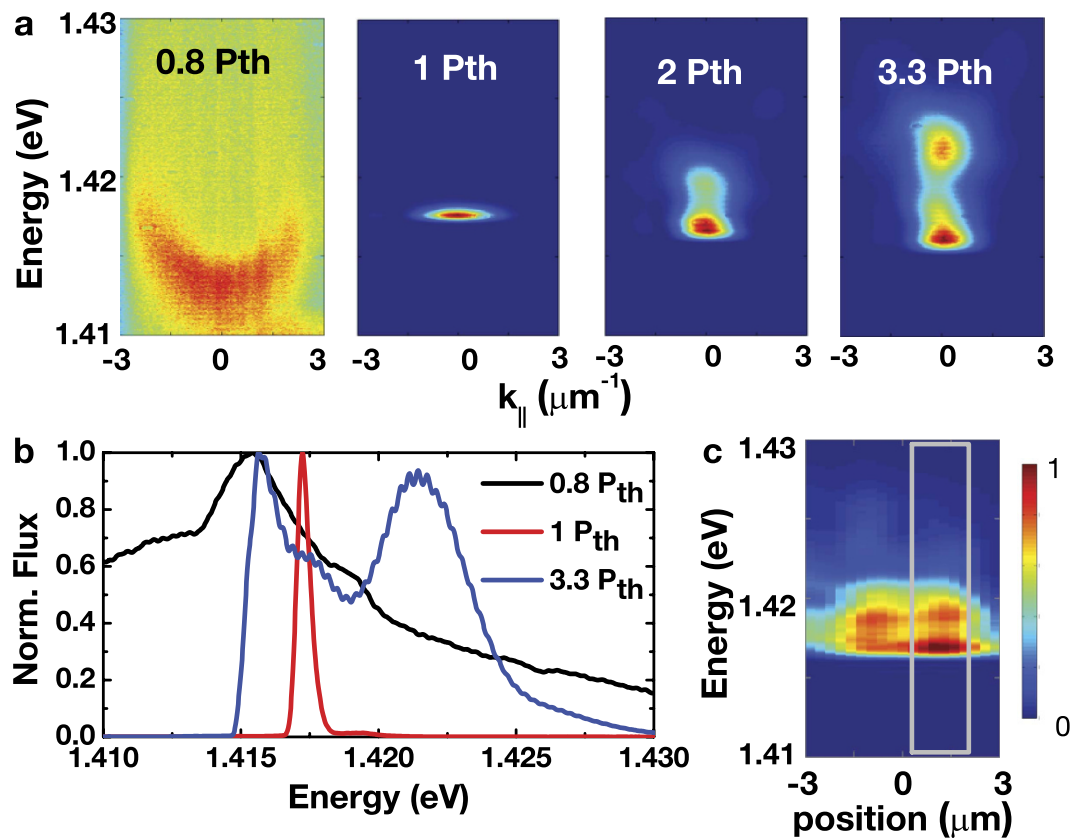
In this study, we report optically controlled dual-energy (two-state) lasing in a highly photoexcited planar Fabry-Pérot semiconductor microcavity in which the gain media, In<sub>0.15</sub>Ga<sub>0.85</sub>As quantum wells (QWs), are positioned at the antinodes of the cavity light field<sup>15</sup>. The sample is *nonresonantly* photoexcited by circularly polarized 2 ps laser pulses at  $E_p = 1.58$  eV, which is about 250 meV above the QW bandgap ( $E_g' \approx 1.33$  eV; the *el-hhl* transition between the first quantized electron and heavy-hole levels) and 180 meV above the bare cavity resonance ( $E_c \approx 1.40$  eV). Above a critical photoexcited density ( $n_c$ ), near-unity circularly-polarized ~10 ps pulsed radiation appears within 10–20 ps after the pulse excitation. The spin-polarized lasing is attributed to the spin-dependent stimulation of cavity-induced correlated electron-hole (*e-h*) pairs formed near the Fermi edge of the high-density *e-h* plasmas<sup>16</sup>. The lasing energies of the samples studied range from  $E_\mu = 1.40$  eV to 1.42 eV, which are near the transition energy between the second quantized energy levels in QWs ( $E_g'' \approx 1.41$  eV; the *e2-hh2* transition), but not locked to the bare cavity resonance  $E_c$ .

In spatially localized areas, an additional low-energy (LE) lasing state with distinct polarization properties appears at well-separated energies of up to 5 meV apart when the photoexcited density increases to a bifurcation density ( $n_b \approx 1.1 n_c$ ) (Fig. 1). The two transient lasing states with distinct spectral and polarization characteristics emerge sequentially in time. Temporally, radiation from the high-energy (HE) spin-polarized state commences within 10 ps after pulse excitation and lasts for ~10 ps. The LE state follows the HE state and lasts for 20–50 ps. Spectrally, slightly above the threshold, the HE state has a

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**Figure 1. Spectral characteristics of dual-energy (two-state) lasing.** (a) Normalized time-integrated spectra of the co-circular ( $\sigma^+/\sigma^+$ ) emission component for  $|k_{\parallel}| < 3 \mu\text{m}^{-1}$ . (b) Emission flux of the HE state (solid red circles), the LE state (solid blue triangles), and the sum (solid black squares). (c) Peak energies (solid shapes) and linewidths (error bars) of the HE state (solid red circles) and LE state (solid blue triangles). The peak energies below the threshold are represented by the solid black squares. The emission fluxes, peak energies, and linewidths are determined by fitting of the spectra with multiple-Gaussian functions. The photoexcited density at the threshold pump flux ( $P_{th}$ ) is  $n_c \approx 2 \times 10^{12} \text{ cm}^{-2}$  per quantum well per pulse.



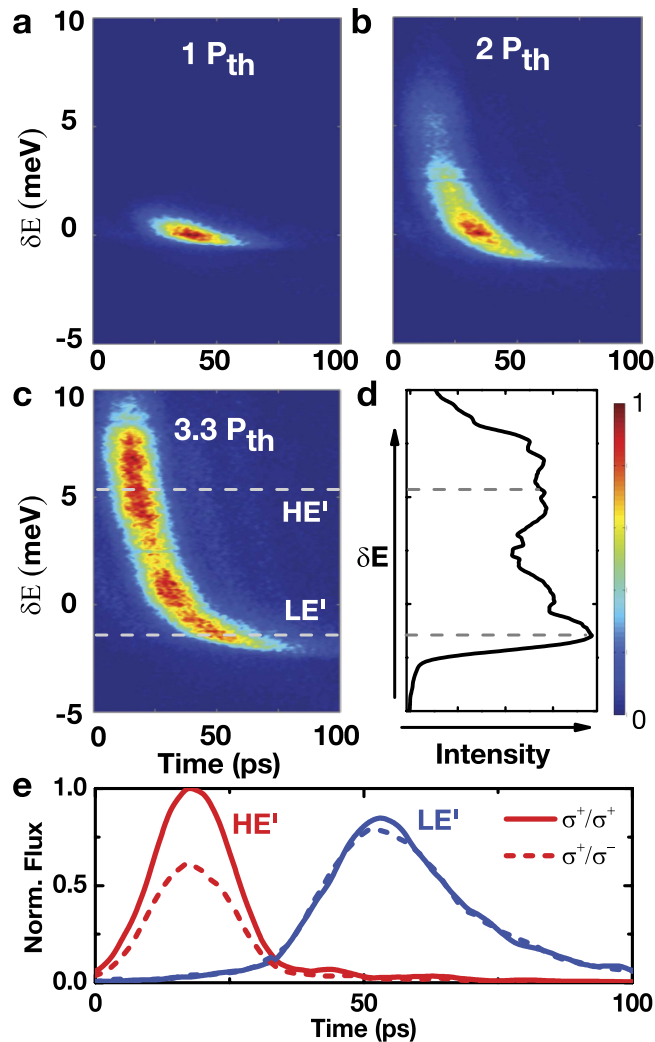
**Figure 2. Two-state lasing in a microcavity.** (a) Angularly resolved ( $k$ -space) imaging spectra of the co-circular ( $\sigma^+/\sigma^+$ ) emission component at  $P = 0.8, 1.0, 2.0,$  and  $3.3 P_{th}$ . (b) Time-integrated spectra at  $0.8$  (solid black line),  $1.0$  (solid red line) and  $3.3 P_{th}$  (solid blue line). (c) Real-space ( $r$ -space) imaging spectra at  $2.0 P_{th}$ . The  $k$ -space spectra shown in (a,b) are measured through a pinhole, as represented by the gray box.

time-integrated linewidth  $\lesssim 3$  meV, whereas the LE state has a linewidth of less than 1 meV. With increasing photoexcited density, the HE state blueshifts about 5 meV, while the LE state redshifts less than 1 meV. As a result, the energy separation between these two states depends on the photoexcited density. Moreover, the two lasing states have distinct polarization properties that are largely determined by the spin imbalance in the  $e$ - $h$  plasmas. Slightly above the lasing threshold, the HE state gives fully circularly polarized radiation as a result of a spin-preserving stimulated process. With an increasing photoexcited density, the HE state becomes a spinor-like state and gives pulsed radiation of positive (+) and negative (−) helicity with a spin-splitting of about 1 meV. By contrast, the LE state appears 20–50 ps after the pulse excitation and exhibits a vanishing degree of circular polarization because of the diminishing spin imbalance.

## Results

We first analyse the density-dependent energy shifts and linewidths. Fig. 1a shows the co-circular  $\sigma^+/\sigma^+$  luminescence spectra  $[S^+(E)]$  at  $k_{\parallel} \approx 0$  as a function of pump flux. Above the bifurcation pump flux  $P_b \equiv 1.1 P_{th}$ , radiation bifurcates into a doublet (*two-state lasing*) with a dominant HE state. The emission flux of the LE state reaches a plateau near above  $1.5 P_{th}$ , whereas the emission flux of the HE state continues to increase linearly with the pump flux. Spectrally, the HE and LE states display distinct energy shifts with increasing photoexcited density. Figure 1c shows the peak energies of the two states with increasing pump flux. Below  $P_b$ , the emission is single-peaked and blueshifts by 6 meV when the pump flux increases from  $0.5 P_{th}$  to  $\sim 1.1 P_{th}$ . When the pump flux increases gradually from  $1.0 P_{th}$  to  $3.5 P_{th}$ , the HE state blueshifts linearly by 5 meV, whereas the LE state redshifts by less than 0.5 meV. This result indicates that the energy difference between the HE and LE states increases with increasing photoexcited density. Above  $P_b$ , the linewidth of the HE state increases from  $\sim 0.3$  meV at  $P_{th}$  to 3 meV at  $4 P_{th}$ . On the other hand, the LE state remains spectrally narrow with a linewidth of 1 meV or less.

Next, we study the spectral characteristics in  $k$ -space and  $r$ -space. Figure 2a shows the  $k$ -space imaging spectra of the co-circular emission component for selected pump fluxes. A nearly parabolic energy versus in-plane momentum ( $E$  vs.  $k_{\parallel}$ ) dispersion curve emerges slightly below the threshold ( $P \approx 0.8 P_{th}$ ). At the threshold (pump flux  $P = P_{th}$ ), the radiation becomes directional (angular-spread  $\Delta k_{\parallel} \leq 1 \mu\text{m}^{-1}$ )

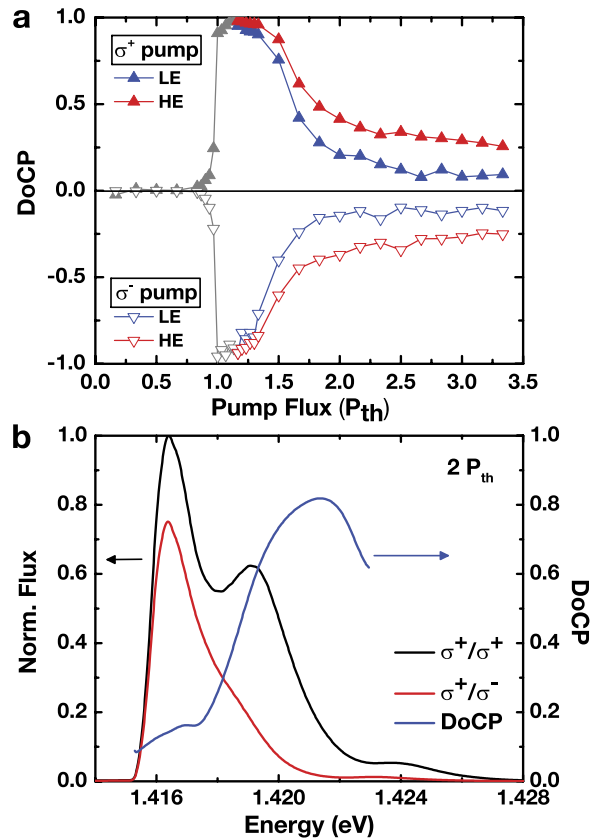


**Figure 3. Dynamics.** (a–c) Time-dependent spectra of the co-circular component at  $k_{\parallel} \approx 0$  under  $P = 1.0$ ,  $2.0$ , and  $3.3 P_{th}$ . The energy  $\delta E$  is measured with respect to  $1.415$  eV, the lasing energy at the threshold. (d) The time-integrated spectrum obtained from (c). (e) Polarized time-dependent luminescence spectra of the HE state at  $\delta E = 5$  meV [HE' as indicated in (c)] (solid and dashed red lines) and the LE state  $\delta E = -5$  meV [LE' as indicated in (c)] (solid and dashed blue lines) for  $P = 3.3 P_{th}$ .

and spectrally narrow (linewidth  $\Delta E \leq 0.3$  meV) (Fig. 2a,b). The time-integrated emission spectra appear to bifurcate into a doublet when the pump flux is increased above  $2 P_{th}$ . The two lasing states are spatially overlapped in-plane, as evidenced in the  $r$ -space imaging spectra (Fig. 2c).

The spectral doublets that appears in time-integrated spectroscopy measurements are in fact temporally separated, as shown in the time-dependent polarized luminescence spectra at  $k_{\parallel} \approx 0$  (Fig. 3). When two-state lasing occurs, luminescence from the high-energy HE state appears within 10 ps after pulse excitation and decays with a time constant  $\tau'_d \sim 10$  ps. After the HE state diminishes, the low-energy LE state appears and then decays with a time constant  $\tau''_d \sim 30 - 50$  ps.

In Fig. 4, we study the polarization properties of the two lasing states. Figure 4a shows the  $\bar{p}_c$  of the HE and LE states as a function of pump flux. The  $\bar{p}_c$  of the HE state rises from nearly zero to unity at threshold, remains above 0.9 for  $P_{th} < P < 1.2 P_{th}$ , and decreases gradually to 0.25 at  $4 P_{th}$ . The LE state appears with a sizable  $\bar{p}_c$  slightly above  $1.1 P_{th}$ , and shows rapidly diminishing  $\bar{p}_c$  for  $P \gtrsim 1.7 P_{th}$ . Moreover, an apparent spin-dependent energy splitting of  $\sim 1$  meV between the co-circular and cross-circular components of the HE state appears, as demonstrated by the selected polarized emission spectra at  $2.0 P_{th}$  shown in Fig. 4b (see also ref. 16). The spectrally resolved  $DoCP(E) = \bar{p}_c(E)$  also shows the energy-dependent  $\bar{p}_c$ , in which the HE state reaches a maximal  $\bar{p}_c(E) \approx 0.8$ , even at a high photoexcited density. Such a high  $\bar{p}_c(E)$  occurs despite the sub-10-ps spin relaxation times of electrons and holes in InGaAs/GaAs QWs, a result that is indicative of sub-10-ps carrier cooling and spin-dependent stimulated processes. The rapid cooling results in a sizable spin imbalance in the cavity-induced correlated  $e$ - $h$



**Figure 4. Polarization Properties.** (a) Stationary degree of circular polarization ( $DoCP = \bar{p}_c$ ) of the HE state (solid and open red triangles) and LE state (solid and open blue triangles) under circularly polarized  $\sigma^+$  pump (upper part, solid triangles) or  $\sigma^-$  pump (lower part, open triangles).  $DoCP = \bar{p}_c = (A^+ - A^-)/(A^+ + A^-)$ , where  $A^{\pm}$  refers to the areas of co-circular ( $A^+$ ) and cross-circular ( $A^-$ ) components of the HE and LE spectral peaks and are obtained by fitting of the time-integrated spectra (Fig. 1) with multiple-Gaussian functions. The  $\bar{p}_c$  of the HE states increases from zero to near unity for  $P_{th} < P < 1.2P_{th}$  and decreases gradually to 0.25. The  $\bar{p}_c$  of the LE states is relatively high ( $>0.8$ ) initially, but it decrease rapidly to less than 0.1 when the pump flux is increased above  $2.0P_{th}$ . (b) The time-integrated polarized spectra  $S^{\pm}(E)$  of the co-circular ( $\sigma^+/\sigma^+ \rightarrow S^+(E)$ , black curve) and the cross-circular ( $\sigma^+/\sigma^- \rightarrow S^-(E)$ , red curve) components at  $P = P_{th}$ . The energy-dependent  $DoCP(E) = \bar{p}_c(E) = [S^+(E) - S^-(E)]/[S^+(E) + S^-(E)]$  (blue curve) shows a maximal  $\bar{p}_c(E) \approx 0.8$ , which is significantly larger than the spectrally averaged  $\bar{p}_c$  shown in (a).

pairs formed near the Fermi edge of the high-density plasmas, whereas the spin-dependent stimulation of these  $e-h$  pairs amplifies such optical spin polarization in the presence of non-radiative loss<sup>16</sup>. By contrast, the LE state has a vanishing  $\bar{p}_c$  because of the lack of spin imbalance at a time delay of 50 ps or more after the optical injection of the spin-polarized carriers.

## Discussion

In this study, the HE state is dominated by a macroscopic population of “hot”  $e-h$  pairs near the Fermi edge via the spin-dependent stimulation. The LE state forms in a confined area of a lateral dimension of  $\sim 2-5 \mu\text{m}$ , which is defined by natural crystalline in-plane inhomogeneities (Fig. 2). After the radiative recombination of these “hot”  $e-h$  pairs, the remaining “cold”  $e-h$  pairs form the LE state that gives spectrally narrow and directional radiation. Therefore, the formation of the LE state hinges on the population of the remaining “cold”  $e-h$  pairs, which typically becomes significant in samples in which the chemical potential  $\mu$  can exceed the bare cavity resonance significantly (i.e.,  $\mu \sim E_g'' \gtrsim E_c$ ).

First, we discuss the density and time dependence of the peak energies of the HE and LE states. In the highly photoexcited microcavity studied here, high-density  $e-h$  plasmas of density  $\approx 1-5 \times 10^{12} \text{cm}^{-2}$  per QW per pulse are formed following *nonresonant* 2 ps pulse excitation as a result of rapid ( $<10$  ps) energy dissipation through optical phonons. When the chemical potential of the  $e-h$  plasma  $\mu$  approaches  $E_c$ , the average refractive index near  $E_c$  can be considerably modified; the result is a sizable blueshift of the effective cavity resonance ( $E_c'$ ). The peak energy of the HE state increases with the pump flux (Fig. 1),

which can be understood as a result of the light-induced refractive index change and the consequent effective cavity resonance shift<sup>17</sup>. Furthermore, under pulse excitation, the effective cavity resonance  $E'_c$  decreases over time toward the bare cavity resonance  $E_c$  owing to the temporal decay of the reservoir carriers (Fig. 3). Slightly above the threshold, the HE state emerges as a burst of spectrally broad  $\sim 10$  ps pulsed radiation, spurred by the stimulation of the majority of the optically active carriers. This rapid depletion of the optically active  $e$ - $h$  pairs in the reservoir precipitates a drop in  $E'_c$ . Afterwards, radiation from the LE state ensures from a local confinement that is replenished with  $e$ - $h$  carriers via spatial diffusion (Figs 2 and 3).

The dual-energy lasing occurs in a locally confined area when the surrounding  $e$ - $h$  pairs are able to diffuse and replenish the area to above the lasing threshold density, even after the depletion of a majority of the optically active  $e$ - $h$  pairs by the HE state. The prolonged replenishment of  $e$ - $h$  pairs from the reservoir causes a temporal redshift of the LE state over  $\gtrsim 50$  ps (Fig. 3). The increasing pump flux results in an apparent time-integrated spectral redshift (Figs 1c and 3b,c). At cryogenic temperatures, similar multiple bursts of radiation have also been observed in localized exciton-polariton condensates formed in a spatially inhomogeneous microcavity. These dynamic relaxation oscillations appear as the result of the interplay between carrier diffusion (gain) and Bose stimulation (depletion)<sup>18</sup>.

Next, we consider the polarization properties, which are affected by the interplay of energy relaxation, spatial diffusion, and spin relaxation. Under circularly polarized nonresonant optical pumping, the HE state is spin-polarized and displays a splitting between the major (co-circularly polarized  $\sigma^{+/+}$ ) and minor (cross-circularly polarized  $\sigma^{+/-}$ ) components of approximately 1 meV<sup>19</sup>. The spin-dependent energy splitting of the HE state is attributed to the spin-dependent stimulation of spin-polarized  $e$ - $h$  pairs within the  $\sim 10$  ps spin relaxation time. The density of the spin-polarized  $e$ - $h$  pairs optically injected by the nonresonant optical pump then reaches the lasing threshold, resulting in co-circularly polarized radiation with the same helicity as the pump. With the increased photoexcited density, the spin-flipped  $e$ - $h$  pairs also reach the lasing threshold, and results in laser radiation with the opposite helicity (i.e., cross-circularly polarized). The laser radiation energy is related to the mean energy of the spin-polarized  $e$ - $h$  pairs; therefore, there is an energy splitting between the co-circularly and cross-circularly polarized lasing components. By contrast, the LE state appears more than 20 ps after the pulse excitation exhibits a diminishing *DoCP* (Figs 3c-e and 4b) owing to the sub-10-ps spin relaxation time of the  $e$ - $h$  pairs.

Spectral functions resembling the two-state lasing spectra presented here have been calculated for  $e$ - $h$  systems in the presence of a cavity by Littlewood *et al.*<sup>20</sup> and Ogawa *et al.*<sup>21,22</sup>. Excitations in exciton condensates<sup>20,23–25</sup> or BCS-like  $e$ - $h$  states<sup>21,22,26–29</sup> can lead to spectral multiplets. Moreover, analogous frequency shifts and doublings of the optical excitations have been reported in Bose gases<sup>30–32</sup>, in which condensate and thermal components coexist. In general, the doublet in the absorption spectra for normal and condensate components in a cold Bose gas is fundamentally related to the two-fluid model of superfluid helium<sup>33</sup> and the two-component superconductivity model of high  $T_c$  superconductors<sup>34</sup>. In contrast to the aforementioned states of thermal equilibrium, the transient dual-energy lasing presented here is a non-equilibrium phenomenon, in which carrier injection by photoexcitation, energy relaxation, spatial diffusion, and dissipation by both spontaneous and stimulated processes should all be considered. In general, a coupled  $e$ - $h$ -photon system like the one described here can exhibit diverse energy distributions and polarization properties. For example, the crossover from photon (i.e., weak coupling limit) to exciton-polariton (i.e., strong coupling limit) lasing with decreasing photoexcited density has been studied in terms of the spectral and angular distributions, dynamics, and polarizations of emissions from a microcavity at cryogenic temperatures<sup>28,35–38</sup>. In semiconductor quantum wells, 10–100 ps bursts of superfluorescence<sup>39,40</sup> have been observed in high-density magneto-plasma in the absence of a cavity structure<sup>41–44</sup>.

To clarify the mechanisms of the two-state lasing effect found in the samples studied here, it is necessary to conduct further temporally and spatially resolved spectroscopies to characterize the energy distribution and relaxation of nonradiative  $e$ - $h$  carriers. For example, one could use a pump-probe spectroscopy to show any time- or density-dependent resonances of the high-density  $e$ - $h$  plasma that is coupled to the cavity light field. Eventually, it will become possible to develop multiple-pulse lasing with energies and polarizations that can be controlled by external optical stimulus<sup>19</sup> and lateral confinement<sup>17</sup>.

## Methods

The undoped microcavity consists of a  $\lambda$  cavity sandwiched within two Bragg mirrors with alternating AlAs/GaAs layers with thickness of  $\lambda/4$ . The active layer includes three sets of three In<sub>0.15</sub>Ga<sub>0.85</sub>As/GaAs (6-nm/12-nm) multiple quantum wells (QWs) embedded at the antinodes of the cavity light field within a  $\lambda$  GaAs cavity. The QW bandgap ( $E'_g$ ) is tuned through a rapid thermal annealing process at 1010 °C–1090 °C for 5–10 s, in which  $E'_g$  blueshifts up to about 20 meV as a result of the diffusion of gallium ions into the QWs. A Fourier transform optical system is used for angle-resolved ( $k$ -space) and space-resolved ( $r$ -space) luminescence spectroscopy and polarimetry. The polarizations of the excitation and emission are controlled and analysed with liquid crystal devices. A circularly polarized light (pump or luminescence) with angular momentum  $\pm \hbar$  along the pump laser wavevector  $\hat{k} \parallel \hat{z}$  is defined as  $\sigma^\pm$ . Time-resolved luminescence and polarimetry are performed with a combination of an imaging



spectrometer and ps streak camera system. Further descriptions of the composition structure and the experimental setups are provided elsewhere<sup>15</sup>.

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## Author Contributions

F.H., W.X. and C.W.L. conducted the experiments and analysed the results, Y.L. and S.L. grew the sample, C.W.L. conceived the experiments and wrote the manuscript. All authors reviewed the manuscript.

## Additional Information

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