Study of ultrafast Rabi flopping in colloidal quantum dots at room temperature

Xingsheng Xu^{1,2} [✉], Zhen Zhen^{1,2}, Siyue Jin^{1,2} & Jie Ren^{1,2}

The interaction between high-intensity ultrashort optical pulses and materials has led to a number of fascinating optical phenomena, including Rabi flopping and self-induced transparency. Until now, there have been few reports on ultrashort coherent pulse propagation and reshaping in semiconductor materials. Here we investigate Rabi flopping and Rabi splitting in colloidal quantum dots with Fabry-Perot cavity of SU8/Si. The Rabi flopping phenomenon is monitored via the pump-probe differential reflection spectroscopy. A high excitation power reshapes the temporal oscillations so that the fast Fourier transform spectra display several peaks. The photoluminescence spectrum by continuous-wave excitation splits under a proper incident angle, and the splitted photoluminescence spectrum is generally consistent with the amplitude of differential reflectivity as function of wavelength. These results demonstrate that both of the temporal oscillations and the splitting of the continuous-wave excited photoluminescence spectra are due to strong coupling between colloidal quantum dots and the Fabry-Perot cavity.

¹State Key Laboratory of Integrated Optoelectronics, Institute of Semiconductors, Chinese Academy of Sciences, Beijing 100083, P. R. China. ²Center of Materials Science and Optoelectronics Engineering, University of Chinese Academy of Sciences, Beijing 100049, P. R. China. [✉]email: xsxu@semi.ac.cn

The interaction between ultrafast pulsed light and matter in a cavity is an important research topic^{1–3}. The interaction between short optical pulses and matter leads to Rabi oscillation⁴, which involves strong coupling. Strong coupling between the light and emitters in a microcavity can be quantified by the vacuum Rabi frequency, and the most prominent form of coherent regulation of such a coupled system is Rabi oscillation or Rabi flopping. An example of this phenomenon is the temporal oscillations in the CuCl microcavity at various incident pump angles, which were detected by transmission-type electro-optic sampling⁵. The subpicosecond fast Fourier transform (FFT) spectrum of the oscillating signal was explained by Rabi oscillations between the middle polariton branch and the lower polariton branch⁵.

Quantum dots in a cavity form a basic platform for investigating Rabi oscillation⁶. Quantum dots are important materials for optoelectronic devices⁷, and single quantum dots in cavities have been extensively studied for quantum information applications⁸. The study of transient coherent phenomena (e.g., Rabi oscillation) in quantum dots produced by Stranski–Krastanov growth usually requires low temperatures⁹, which limits the practical applications. An alternative to Stranski–Krastanov-grown quantum dots is colloidal quantum dots (CQDs), which are easily synthesized by the wet chemical method¹⁰, and a single CQD can serve as a single-photon source at room temperature (RT)¹¹.

CQDs usually have a broad photoluminescence (PL) spectrum at RT¹². Both ensemble effects (inhomogeneous line broadening) and coupling to the bath (homogeneous line broadening) result in fast exciton dephasing in CQDs, thus challenging the detection of transient coherence^{13,14}. Such ultrafast coherent processes in CQDs, including real-time Rabi oscillation¹⁵ and carrier-wave Rabi flopping¹⁶, were rarely reported by using femtosecond spectroscopy. In addition, continuous-wave (CW) excitation realized strong coupling between the CQDs and planar cavities at RT¹⁷, which is indicative of a large transition dipole moment in CQDs. The strong coupling between the CQDs and photonic modes split the reflectivity spectrum¹⁷ or the PL spectrum¹⁸, which is similar to self-induced transparency¹⁹.

Previous studies have investigated the dipole–dipole interaction in electromagnetically induced transparency in polaritonic band-gap materials doped with an ensemble of four-level nanoparticles^{19,20}. In these studies, the researchers recognized that the probe field-induced dipoles and dipole–dipole interaction (DDI) in the quantum emitters with metallic nanorod shells through investigating two-photon fluorescence of the material^{19,20}. The results showed that the two-photon fluorescence spectrum split from one peak to three peaks due to dressed states created in the system by strong DDIs²⁰. Other similar researches have been done on hybrid materials with quantum dots or other quantum emitters^{21,22}.

This present study investigates strong coupling and Rabi flopping in a CQDs-planar-cavity system excited by femtosecond pulses at RT. The resulting ultrafast coherent processes are monitored via differential reflectivity of CQDs on a low-refractive-index/high-refractive-index hybrid material. It reveals Rabi flopping in a strong coupled CQDs-planar-cavity system. The splitting of the CW PL spectrum as a function of incident angle suggests that strong coupling occurs between the excitons in CQDs and the planar cavity in the CQDs/SU8/Si material. The Rabi frequency obtained from CW excitation is comparable to that obtained from transient differential reflectivity measurements. These results demonstrate that Rabi flopping is caused by strong coupling between the excitons in CQDs and the planar

cavity formed by the SU8 layer on Si substrate. Moreover, carrier-wave Rabi flopping may occur at high pump intensity.

Results and discussion

Ultrafast transient processes in CQDs excited at 770 nm. The transient signals of the differential reflectivity (shown in Fig. 1a–d) of CQDs on SU8/Si were measured by using the pump-probe method, which is described in the Methods section. The period of the temporal oscillations in the transient signal varies substantially with pump power. Figure 1a–d show that, for pump powers of 0.177, 3.0, 56.0, and 82.5 mW, the maximum intervals in the four oscillations are 158, 127, 112, and 89 fs, and the minimum intervals are 104, 44, 51, and 66 fs, respectively. At low pump power, the oscillation period is large, whereas the oscillation becomes nonperiodic at high pump power, where a mixture of oscillation with large and small periods appears, and the small-period oscillation plays an important role.

Figure 1a shows the transient differential reflectivity at a pump power 177 μ W, and the oscillation period is as large as 158 fs. Applying a fast-Fourier transform (FFT) to the transient spectrum produces the spectrum shown in Fig. 1e, in which a main spectral peak appears with small wrinkles at higher frequencies. The temporal oscillation in Fig. 1a is a Rabi oscillation^{5,15}, and the peak in the FFT spectrum gives the Rabi frequency of 5.79 THz, which corresponds to a Rabi splitting energy of 23.9 meV. Increasing the pump power to 1.5 mW produces the differential reflectivity shown in Supplementary Fig. 1a, which shows that the intervals between the adjacent transient peaks decrease and become nonuniform in comparison with those in Fig. 1a. The typical oscillation interval in Supplementary Figure 1a is 82 fs. The corresponding FFT spectrum in Supplementary Fig. 1e displays several spectral peaks, with the main peak being at the high frequency of 13.3 THz. Upon increasing the pump power to 3.0 mW (Fig. 1b), the FFT spectrum becomes a broadband (Fig. 1f) with a high-frequency edge at 16 THz followed by a small spectral peak at 21 THz. Increasing the pump power to 56.0 mW (Fig. 1c) produces three main peaks in the FFT spectrum (Fig. 1g), notably, a strong peak appears at 16.9 THz (69.85 meV), a slightly smaller peak appears at 10.9 THz (45 meV), and the third smaller spectral peak appears at 8 THz (33 meV). Upon increasing the pump power to 82.5 mW, the three main spectral peaks shift to 6.15, 13.3, and 20 THz. Although the peak at 20 THz weakens, it is broader than the other two peaks. Moreover, some small peaks appear at high frequencies, as shown in the inset of Fig. 1h. Generally, the frequency of the FFT spectral peak increases with the pump power increasing.

As shown in Fig. 1 and Supplementary Figure 1, the amplitude of the temporal oscillation and the amplitude of the FFT spectrum increase with the pump power increasing. In addition, the intervals between temporal oscillations become small and irregular, which leads to peaks at higher frequencies in the FFT spectrum (see Supplementary Note 1). With increasing pump power, the number of peaks in the FFT spectrum increases, with smaller peaks appearing both at higher and lower frequencies. Moreover, increasing the pump power clearly stretches the periods of some of the temporal oscillations and produces a pronounced beating in the coherent signal²³. These dynamics are associated with several FFT spectral peaks appearing at high and low frequencies beside the main spectral peak, as shown in Fig. 1e–h and Supplementary Fig. 1e–h. This is the spectral fingerprint of carrier-wave Rabi flopping^{23,24}. Increasing the pump intensity higher still may cause a more complex nonlinear effect in

Fig. 1 Temporal oscillations and fast Fourier transform (FFT) spectra from colloidal quantum dots (CQDs) pumped at various powers by femtosecond pulse at 770 nm. **a–d** Differential transient reflectivity as function of delay time between pump and probe light; **e–h** Corresponding FFT spectra pumped at wavelength 770 nm with indicated powers (177 μW , 3.0 mW etc.). Insert in **g** and **h** is the FFT spectrum at high frequency. R represents reflectivity, $\Delta R/R$ represents differential reflectivity, fs represents femtosecond, and THz is the acronym of TeraHertz, which is the frequency unit.

which more spectral peaks appear, or the spectrum broadens due to four-wave mixing.

The interaction strength between light and matter can be characterized by the Rabi frequency, which is linearly proportional to the dipole moment and the electric-field amplitude²⁵. With increasing pump power, the Rabi frequency increases ($\Omega_R \propto E_0$), and the Rabi period decreases. The temporal oscillations of the differential signals are attributed to Rabi oscillations of the CQDs in the planar cavity formed by a 3- μm -thick SU8 film on Si. The thickness of the SU8 film can only support optical modes in the visible and near-infrared region. Usually, the RT exciton emission spectrum of CQDs in the visible or near-infrared is broad, and transient coherent processes in CQDs, such as Rabi oscillation, are difficult to be observed. However, in our experiment, the CQDs emission couples evanescently to the Fabry-Perot (F-P) mode supported by the SU8 film on the Si substrate. In addition, as described in detail below, the F-P mode in SU8/Si is a quasi-bound state in the continuum, which enhances the

light-matter interaction and makes transient coherent processes possible.

Ultrafast transient processes in CQDs excited at 710 nm. Figure 2a, Supplementary Figure 2a–c, and Supplementary Figure 3a–c show the transient differential reflectivity of CQDs/SU8/Si when pumped at 710 nm and at various pump powers. When the sample was pumped at 315 μW , the transient signal is weak, and the signal near-zero delay time oscillates with a period of 215 fs. Figure 2b shows the corresponding FFT spectrum of the transient signal; a main spectral peak appears at approximately 4.6 THz. At a pump power of 524 μW , the transient signal oscillates with a period of approximately 200 fs (Fig. 2a, Supplementary Fig. 2b). The FFT spectrum in Fig. 2b (Supplementary Fig. 2e) has a main peak at 4.55 THz and a small peak at 9.1 THz. As the pump power was increased to 3.4 mW, the oscillating transient signal changes shape (Fig. 2a, Supplementary Fig. 3a), and the corresponding FFT spectrum develops a main spectral peak at 4.1 THz

Fig. 2 Transient differential reflectivity of colloidal quantum dots (CQDs) on SU8/Si with femtosecond pump pulse at wavelength 710 nm. **a** Transient differential reflectivity ($\Delta R/R$) and **b** corresponding fast Fourier transform (FFT) spectra under 710 nm pump with various powers (315 μ W, 524 μ W etc.). Different pump power is denoted by different line pattern. Acronyms of fs and THz are the same as those defined as in the caption of Fig. 1.

and a small spectral peak at 10.95 THz. The high-frequency peak is more obvious when pumped at 3.4 mW than when pumped at 524 μ W. The interval between the two peaks becomes larger, that is, the main peak redshifts while the small peak blueshifts. Pumped at 7.0 mW, the oscillation period becomes small (see Fig. 2a and Supplementary Fig. 3b) and the main peak in the FFT spectrum appears at 4.58 THz, with a small peak at 12.6 THz (Fig. 2b and Supplementary Fig. 3e). In addition, a new small peak appears at 2.29 THz. Being pumped at 14.0 mW, the FFT spectrum displays two small peaks at high frequency (Fig. 2b and Supplementary Fig. 3f). The appearance of the small peaks near the main spectral peak is due to carrier-wave Rabi flopping²⁴ (see Supplementary Note 2).

As shown in Fig. 3a–c, when the pump powers were increased to 28.0, 51.7, and 62.9 mW, the temporal oscillation curves are relatively smooth and periodic, and their corresponding FFT spectra have only one main peak (Fig. 3e–g). As the pump power increases further, the CQDs produce shaped oscillations again, as shown in Fig. 3d with pump power 75.0 mW, and the small, high-frequency spectral peak appears again, as shown in Fig. 3h. The basic temporal oscillation of transient differential signal is Rabi oscillation¹⁵ or Rabi flopping²⁶ (see Supplementary Note 4). In Fig. 3, under high-power excitation, the electron is pumped to high-excited states, so that electron transitions mainly occur between high-excited states and the ground state. In this case, the strong exciton-cavity coupling requires higher pump power than that used in Fig. 2. Therefore, the FFT spectra display only a single spectral peak when pumped at high power, as shown in Fig. 3e–g. As the pump power increases more, the transient signal becomes asymmetric, and the FFT spectrum again develops more spectral peaks in the high-frequency region. As the pump was turned on, the coupled system started from the ground state and gradually became more excited, increasing as it occupied the higher level²⁷. However, once the system had become fully excited, the pump wave stimulated the emission of radiation and the system returned to the ground state²⁷. Then the whole process started again. This is the Rabi flopping²⁷, and it occurs at the Rabi frequency.

Strong coupling of CQDs under continue-wave excitation.

With the same sample of CQDs on SU8/Si as used in Figs. 1–3, the PL and reflection spectral images were acquired by using a confocal optical system. A CW laser at 532 nm was focused on the sample by an objective lens with 40 $\times$ magnification and a numerical aperture of 0.6, and the spectral image was detected by a monochromator with a CCD. The spectral image can be

decomposed into several spectral lines as a function of incident angle, which gives the angle-resolved PL and reflectivity spectra. Figure 4a compares the PL spectrum and the reflectivity spectrum at an incident angle of 0° with pump power 1.5 mW. The structure of the sample is simple but special: it consists of an F-P cavity in the form of a SU8 film with a low-refractive index on a Si substrate with a high-refractive index, with the CQDs coated on the surface of the SU8 film. The reflectivity spectrum in Fig. 4a has two peaks centered at 782.27 and 806 nm that correspond to two F-P modes. Two PL spectral peaks appear at 758.23 and 808.42 nm, on each side of the reflectivity peak at 782.27 nm. The reflectivity spectrum is measured by using a broadband Halogen light source, which is too intense to distinguish the reflectivity spectral peak in the short-wavelength region with the CCD camera. The calculated reflectivity spectrum (Fig. 4b) has a peak at approximately 715 nm, which is entirely consistent with the measured reflectivity spectrum (Supplementary Fig. 4d). The two peaks of the measured PL spectrum also sandwich the peak in the calculated reflectivity spectrum (cf. Fig. 4b and Supplementary Fig. 4d). This reflectivity peak corresponds to an F-P mode in the SU8/Si material (see Supplementary Note 3).

The calculated reflectivity spectrum in Supplementary Figure 4b shows that normally reflectivity spectrum from SU8/Si is opposite that from freestanding SU8 (i.e., the SU8 film in air). The peaks in the reflectivity spectrum from SU8/Si correspond to the F-P modes. The PL spectrum of the CQDs on SU8/Si collected at various angles splits into two main peaks (Supplementary Fig. 4c–e), while the PL spectrum has only one broad peak for CQDs on bulk quartz (Supplementary Fig. 4f). Therefore, the two PL spectral peaks sandwiching the reflectivity peak result from PL spectral splitting by the CQDs on SU8/Si and is due to the strong coupling between the excitons in the CQDs and the F-P mode in SU8/Si. The measured two PL bands at 758 and 808 nm in Figs. 4a and 4b correspond to two polariton branches. The PL spectrum acquired at 0° has three peaks, with the small peak at 713.9 nm resulting from the enhancement by the F-P mode corresponding to the reflectivity peak at 715 nm in Fig. 4b.

As shown in Fig. 4b and Supplementary Figure 4f, the PL spectra collected at various angles can be fit by multiple Gaussians. Figure 4c plots the peak energies from the PL and reflectivity spectra around the exciton emission as a function of incident angles. The positive and negative numbers correspond to the incident angles of the objective lens. The PL peak energies displayed by the solid squares fall above and below the energy of the reflectivity peak (hollow circles). The energy of the reflectivity

Fig. 3 Transient differential reflectivity and corresponding fast Fourier transform (FFT) spectra with femtosecond high-power pump at 710 nm. **a–d** Transient differential reflectivity ($\Delta R/R$) and **e–h** corresponding FFT spectra with pump power 28.0 mW (**a, e**), 51.7 mW (**b, f**), 62.9 mW (**c, g**), 75.0 mW (**d, h**).

peak can be fit by the F-P mode equation

$$E = \frac{\alpha}{\sqrt{1 - \left(\frac{\sin \theta}{n}\right)^2}}, \quad (1)$$

where E is the energy of the F-P cavity mode and n is the refractive index of the cavity material. In the fitting, $n = 1.58$, which is the refractive index of SU8. The gray solid line in Fig. 4c shows the fit, which is consistent with the energies of the reflectivity peaks. The two curves for the energy of the PL peaks can be fit by using

$$E_{\pm} = \frac{(E_m + E_{QD})}{2} \pm 0.5 \sqrt{(E_m - E_{QD})^2 + g^2}, \quad (2)$$

where E_m is the angular-dependent photon mode, E_{QD} is the exciton transition energy, and g is the energy of the Rabi splitting. The exciton transition energy (dashed line) crosses the energy of the F-P mode at approximately $\pm 10^\circ$, and these two fitted solid lines anticross at $\pm 10^\circ$, which is indicative of strong coupling between the

CQD exciton and the F-P mode in SU8/Si. The blue solid line and the red solid line are fittings to the two polariton branches. The Rabi splitting obtained is 100 meV, which corresponds to a Rabi frequency of 24.2 THz. This Rabi frequency obtained from CW excitation is comparable with those obtained from the FFT spectra from the transient differential reflectivity shown in Figs. 1–3.

In the pump-probe experiment, we changed the pump wavelength and maintain almost the same pump power of 5.5 mW to measure the differential reflectivity of the material (see Supplementary Fig 5). The maximum amplitude of the differential reflectivity signal $\Delta R/R$ as a function of excitation wavelength is compared in Fig. 4d with the PL spectrum at incident angle 0° . At 713.9 nm, a PL peak corresponds to a peak in $\Delta R/R$, which means that the F-P mode enhances the PL and the $\Delta R/R$. The PL peaks at 758 and 808 nm correspond to two polariton branches under 532 nm CW excitation. The PL spectral peaks are nearly in accord with the peaks of $\Delta R/R$, which means that, when the excitation wavelength approaches the polariton wavelength, the differential reflectivity ($\Delta R/R$) is enhanced.

Fig. 4 Strong coupling between colloidal quantum dots (CQDs) and Fabry-Perot (F-P) cavity upon continuous-wave pumping. **a** Photoluminescence (PL) spectrum (red solid line) from CQDs on SU8/Si compared with reflectivity (RL) spectrum (blue dashed line) from SU8/Si at normal incidence. **b** PL spectrum (black solid line) from CQDs on SU8/Si and calculated reflectivity spectrum (blue dashed line) from 3- μ m-thick SU8/Si and at normal incidence. The short-dotted lines are fittings to multiple Gaussians. **c** Central peak energies of the PL spectra and the reflectivity spectra as a function of incident angle. Solid and open squares represent the measured PL peak energies (denoted by symbols of PL1_Exp and PL2_Exp), and open circles represent the measured reflectivity peak energies (denoted by RL_Exp). The gray curve is a fitting to Eq. (1) for the F-P mode (denoted by RL_Fit), and the blue and red solid curves are fittings to Eq. (2) for strong coupling (denoted by PL_Fit). The horizontal dashed line shows the exciton transition energy. **d** Comparison of PL spectrum (red dashed line, denoted by PL_Ag0) from CQDs on SU8/Si at normal incidence with the maximum amplitude of the differential reflectivity ($\Delta R/R$) as function of wavelength. $\Delta R/R$ was measured under the same pump power (5.5 mW) at different wavelength with femtosecond pump pulse.

However, some mismatch still remains between the CW polaritons and the $\Delta R/R$, which may be due to the differences in excitation wavelength and the characterization of the excitation laser. The polaritons in Figs. 4c and 4d are excited by CW pumping, whereas $\Delta R/R$ in Fig. 4d and Supplementary Figure 5 are excited by femtosecond pulses. Moreover, under femtosecond-pulse excitation at the right wavelength with a suitable pump power, the transient signal of $\Delta R/R$ is reshaped, and more spectral peaks appear in the FFT spectrum, which is due to carrier-wave Rabi flopping.

Comparing the transient signals and the FFT spectra for pulse excitation at 770 nm in Fig. 1 and Supplementary Figure 1 with those for pulse excitation at 710 nm in Fig. 2 (also see Supplementary Fig. 2–3) show that, the transient signals and the FFT spectra for pulse excitation at 770 nm are much more sensitive to the pump power than those for pulse excitation at 710 nm. In the same range of pump power, the transient signals pumped by 770 nm are stronger than those pumped by 710 nm, and the oscillation periods by 770 nm are smaller. Consequently, the FFT spectra for pulse excitation at 770 nm have several spectral peaks but only a single main peak for pulse excitation at 710 nm. When pumped at 770 nm with a power of 3.0 mW, several strong peaks appear in the FFT spectrum, even at a frequency higher than 10 THz. However, with a similar pump power at 710 nm, the main spectral peak still appears below 10 THz, and only one small spectral peak appears at high-frequency region. The maximum amplitude of the temporal oscillation in $\Delta R/R$ shown in Fig. 4d occurs near 770 nm, which is approximately between the upper (UP) polariton branch and the peak of the F-P mode. The temporal oscillation of differential reflectivity signals come from Rabi oscillations¹⁵, their periods and amplitudes are related to the polaritons. The efficiency of Rabi oscillations is determined by the absorption and the field of the cavity mode. Moreover, the transient

signal is produced by femtosecond pump laser, so the temporal nature of the excitation should also be considered. Therefore, the shaped transient differential reflectivity signals and the corresponding FFT spectra at different pump wavelengths and different pump powers result from the combined effect of absorption, F-P resonance, and the time-derivative of the excitation pulse. Because pump wavelength at 770 nm is nearly resonant with the exciton emission wavelength and the cavity mode at 781 nm, and is not far from the polariton wavelength in the trilayer CQDs/SU8/Si structure, so the amplitude of the transient $\Delta R/R$ signal around 770 nm is enhanced.

The FFT spectra in Fig. 1 due to femtosecond pumping at 770 nm reveal the energies of the spectral peaks approximately in the range of 24–70 meV. Furthermore, the anti-crossing phenomenon of the 532 nm, CW pumped PL spectra (see Fig. 4c) gives a Rabi frequency of 100 meV. Therefore, the Rabi frequency obtained from spectral splitting by CW excitation is comparable to that obtained from temporal oscillations created by femtosecond excitation. There is some difference between them, which is understandable because the Rabi frequency is proportional to the excitation power, and the absorption and excitation powers of CW and pulsed excitation differ. This result confirms that the PL spectral splitting upon CW excitation has the same origin as the transient differential reflectivity oscillations from pulse excitation: they are both due to the strong coupling between the CQDs excitons and the F-P modes in the simple low-refractive-index/high-refractive-index bilayer.

The F-P mode formed in the low-refractive-index film on a high-refractive-index substrate is a quasi-bound state in the continuum¹⁸. The schematic drawing of the material of SU8/Si is shown in Fig. 5a, and Fig. 5b indicates that the high-refractive-index Si substrate under SU8 can be approximated as a mirror. When the condition of the F-P mode in the SU8 film is satisfied,

Fig. 5 Schematic structure of the SU8/Si material. **a** Schematic of the structure of SU8/Si material, where n_1 , n_2 , and n_3 represent the refractive indexes for air, SU8, and Si, respectively; R1 and R2 denote the reflectivity from the surface of SU8 and that from the Si surface, respectively. **b** Effective cavity formed in SU8/Si bilayer, where the bottom Si with large refractive index (n_3) is effective as a mirror. Light in the Fabry-Perot (F-P) cavity leaks out via two pathways as shown by the blue arrow line and the red arrow line.

the optical phase for optical resonance is $2k\pi$, where k is an integer. The light of the F-P optical mode is lost via two pathways: the transmission R1 from the upper surface of SU8 (shown by the blue arrow in Fig. 5a) and the reflectivity R2 from the surface of Si (shown by the black arrow in Fig. 5a). Due to the high-refractive index of Si, the reflectivity R2 in the second pathway has an additional phase of π , and the total phase difference between R2 in the second pathway and R1 in the first pathway is $2k\pi + \pi$. The phase difference between light transmitted from the air-SU8 interface and that reflected from the SU8-Si interface is $2k\pi + \pi$, which leads to destructive interference¹⁸. Therefore, loss to the mode in the SU8 film is low, and the F-P mode in the low-refractive-index/high-refractive-index configuration is a quasi-bound state in the continuum. Light satisfying the F-P resonance condition thereby produces a peak in total reflectivity, as displayed by Supplementary Figure 4a. The bound state in the continuum has low loss and a long lifetime. The evanescent coupling of the exciton emission from CQDs on SU8/Si to the quasi-bound state in the continuum increases the lifetime of the CQDs excitons. This can explain the occurrence of the transient coherence of the material.

In fact, the strong coupling between excitons and photons might be mediated by the DDI. To understand this, we compare the above results with those of the DDI in quantum emitters^{19,20}, where the core-shell hybrid structure included a core of metallic nanoparticles and a shell of an ensemble of quantum emitters²⁰. The surface plasmon polariton field could be induced by the external (probe) field, and the dipoles were induced subsequently in the ensemble of emitters. Therefore, anomalous DDI formed through interaction between induced dipoles in the quantum emitters^{20,28}. As a result, the PL spectrum, as well as the scattering cross section, split into two peaks mainly due to the strong coupling between excitons and anomalous DDI²⁸. In similar research, a theory was developed for the PL and absorption coefficient in nanohybrid structures made of an ensemble of metallic nanoparticles and core-shell quantum emitters (CdSe/ZnS CQDs). Excitons in the quantum emitters interact with surface plasmon polaritons via the DDI. The results show that PL quenching and enhancement occur mainly due to the DDI in the ensemble²⁹. In our experiment, the main effect on the PL is the interaction between the CQDs and photonic modes (quasi-bound states in the continuum) in an F-P cavity, where the electric field of the F-P modes is strong enough to induce strong coupling in the CQDs. The Rabi splitting is due to strong coupling between CQDs and photonic modes. According to the formula for the Rabi frequency ($\Omega_R \propto E_0$), if the strength of the

DDI electric field²⁸ is comparable to that of the photonic modes, then strong coupling will be enhanced and the Rabi splitting. With the help of semiclassical theory, the exciton coupling with the photonic mode is approximated in terms of the DDI, but no evidence indicates that a DDI electric field is generated and cannot be ignored in our material as reported in previous works^{19,28}.

In conclusion, we have studied both transient differential reflectivity using femtosecond pump-probe spectroscopy and CW PL spectrum of CQDs/SU8/Si trilayer material. The temporal oscillations of differential reflectivity created by femtosecond pump pulses are identified as Rabi oscillations, and their FFT spectra reveal several THz frequencies corresponding to transitions between polariton branches due to strong coupling between the excitons in CQDs and the F-P modes in the hybrid structure. Pumping with high power reshapes the oscillation in the differential reflectivity, which produces multiple peaks in the FFT spectra due to carrier-wave Rabi flopping. The amplitude of the Rabi flopping as a function of wavelength is basically consistent with the splitted PL spectrum. These results demonstrate that Rabi flopping revealed by ultrafast transient differential reflectivity is due to strong coupling between excitons and F-P modes in the low-refractive-index/high-refractive-index material, and the F-P modes are quasi-bound states in the continuum in the bilayer material.

Methods

Sample preparation. The SU8 was spin-coated on a Si substrate to form a 3- μm -thick layer of SU8 on Si. The CQDs (Qdot™ 800 ITC™) were purchased from Thermo Fisher and consisted of CdTeSe cores and ZnS shells. The CQDs with a high concentration of 1×10^{-8} M were coated onto the SU8/Si surface by drop-casting.

Pump-probe measurement. A femtosecond laser is used to excite the CQDs on a SU8/Si structure and then another femtosecond laser pulse was used to probe the structure (pump-probe setup) under ambient conditions. The femtosecond laser produced 140 fs pulses at a repetition rate of 80 MHz, and the wavelengths for the pump light and for the probe light were the same and were tunable. The probe laser was focused onto the CQDs normal to the SU8 surface, while the pump light was focused by the same lens but at an angle of approximately 5° with respect to the surface normal. The focal length of the pump lens was 150 mm. The probe powers were 51 μW , 160 μW for wavelengths 710 and 770 nm, respectively. The pump power remained higher than the probe power at corresponding wavelength. A half waveplate was inserted into the pump optical path to vary the pump polarization so that the pump and probe polarizations were orthogonal. A polarizing beam splitter in the optical path for collecting the probe reflection was oriented to be orthogonal to the pump, thereby only transmitting the probe. The emission from the CQDs evanescently coupled to the SU8 film, which formed an active cavity. Under different excitation powers and wavelengths, the differential reflectivity was acquired as a function of pump-probe delay time. The frequency-

domain spectra were then obtained by FFT of the differential reflectivity. In the measurement of pump-probe spectroscopy, the time step of the delay time between the pump pulse and probe pulse was adjusted as a suitable value to obtain proper transient differential reflectivity signal. The time step of the delay time between the pump pulse and probe pulse was chosen as 6.66 fs for pump wavelength 710 nm with different pump powers, and the time step was 3.33 fs for other pump wavelengths.

Photoluminescence spectroscopy. The PL spectra from the CQDs on SU8/Si were acquired by using a confocal system, and the PL spectra were detected and analyzed by using a spectrometer from Princeton Instruments. An angle-resolved PL measurement setup was constructed that projects the objective Fourier plane onto the spectrometer configured with a CCD chip to capture the far-field image. All measurements were made at RT, and the sample was excited by a nonresonant CW laser at 532 nm with pump power 1.5 mW, as measured just in front of the microscope objective. We acquired the spectral image by using an objective and then decomposed it into spectra at different incident angles. The objective magnification was $40\times$ with a numerical aperture of 0.60, corresponding to an angular range of -36.9° – 36.9° . The PL spectra at various incident angles were projected onto the momentum range on the CCD. The PL spectra at various angles were then extracted from the CCD chip. The polariton dispersion relation was mapped by fitting the PL spectra at various angles to a bi-Gaussian function. Varying the pump distance (i.e., the distance between the surface of the lens and the sample surface) will vary the amplitude of the PL spectral peaks and the spectral structure and change the incident angle. When the pump distance is less than the focal length of the focal lens, the incident angle range is smaller than the angle range corresponding to the numerical aperture.

Data availability

All data are available in the main text or in the supplementary materials.

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Author contributions

X.S.X. conceived the experiment, performed the measurements, carried out data analysis, and wrote the manuscript. Z.Z. wrote the data-collection software for the pump-probe measurement and helped optimize the figures. S.Y.J. prepared the sample. J.R. took part in discussions. All authors reviewed the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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Correspondence and requests for materials should be addressed to Xingsheng Xu.

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