Strong coupling of single quantum dots with low-refractive-index/high-refractive-index materials at room temperature

Xingsheng Xu1,2* and Siyue Jin1,2

Strong coupling between a cavity and transition dipole moments in emitters leads to vacuum Rabi splitting. Researchers have not reported strong coupling between a single emitter and a dielectric cavity at room temperature until now. In this study, we investigated the photoluminescence (PL) spectra of colloidal quantum dots on the surface of a SiO2/Si material at various collection angles at room temperature. We measured the corresponding reflection spectra for the SiO2/Si material and compared them with the PL spectra. We observed PL spectral splitting and regarded it as strong coupling between colloidal quantum dots and the SiO2/Si material. Upper polaritons and lower polaritons exhibited anticrossing behavior. We observed Rabi splitting from single-photon emission in the dielectric cavity at room temperature. Through analysis, we attributed the Rabi splitting to strong coupling between quantum dots and bound states in the continuum in the low-refractive-index/high-refractive-index hybrid material.

INTRODUCTION

A cavity can control the spontaneous emission of a quantum dot (QD) and the cavity quantum electrodynamics regime (1, 2). Studying vacuum Rabi splitting for a single QD with a single photon level is important, including the mechanisms of vacuum Rabi splitting and the essential criteria and conditions for realization of vacuum Rabi splitting between single QDs and microcavities (3). In a true strong coupling regime, as few as one or two photons can successfully realize the nonlinear quantum optics process, which has application in the control of the emission direction of a single-photon source, turnstiles of indistinguishable single photons, quantum phase gates, quantum entanglement, quantum-state transfer devices, and quantum physics in the quantum information regime (4–11). If the coupling strength between a cavity and a two-level atom (or a QD) exceeds the atom decay rate, then the spectrum [photoluminescence (PL) or transmission] splits and vacuum Rabi splitting occurs. Vacuum Rabi splitting is a fundamental quantum phenomenon, and the implementation of solid-state single-oscillator vacuum Rabi splitting will lead to true strong coupling (12). Vacuum Rabi splitting could also occur in many atoms or many QD systems (13), where the behavior will be classical.

For a cavity, important factors for strong coupling usually include a small mode volume V and a high Q. Plasmonic resonators have a mode volume that is small enough to produce strong coupling, which researchers usually study using scattering measurement (14). Scattering measurements alone were insufficient for confirming strong coupling, as the results were unclear because both a strong coupling effect and an intermediate coupling effect could produce two spectral peaks in the scattering spectrum (15). Because PL is incoherent light, the Fano interference phenomenon cannot appear with PL light. Splitting of the PL spectrum into two peaks can only occur in the strong coupling process, and thus, researchers recognized this phenomenon as a decisive sign of Rabi splitting.

For colloidal QDs (CQDs), researchers found strong coupling in an open microcavity formed by a silver mirror (16). The large oscillator strength and fluorescence quantum yield has contributed to the strong coupling effect. Giebink et al. (17) reported strong exciton-photon coupling with CQDs in a one-dimensional microcavity with a Q factor of 250. They found exciton-photon coupling energies between 32.8 and 24.6 meV. The coupling between the CQDs and the microcavity was large because the oscillator strength of the transition in the CQDs was large. Researchers also found strong coupling for two-dimensional atomic and dielectric cavities based on PL spectra (18). The PL spectra could be extracted from a charge-coupled device (CCD) at various in-plane momenta, corresponding to the emission angle. The researchers observed an anticrossing behavior with a Rabi splitting of 23.5 meV, the key signature of the strong coupling regime. The emission was still classical light, not single-photon emission (18). Strong exciton-photon couplings in one-dimensional microcavities have also been reported for other active materials, mainly on the basis of reflection (RL) spectra (19, 20).

Until now, only a few reports on PL splitting for demonstrating the strong coupling between single CQDs and a nanocavity at room temperature have been presented; one example has no results for the anticrossing phenomenon (15), and the other two investigated tip-enhanced strong coupling (21, 22). These few examples of strong coupling of single photons according to PL splitting were based on a plasmonic nanocavity. Until now, strong coupling between a single emitter and a dielectric cavity at room temperature has not been reported. Compared with a plasmonic cavity, a dielectric cavity has very small absorption loss, has efficient off-chip coupling, can achieve a high Q, and is easy to integrate. These characteristics are very useful for realizing large-scale photonic integrated circuits and quantum integrated chips. In addition, bound states can exist above the continuum threshold. These special states are called bound states in the continuum (BICs) (23) and appear as a resonance that does not decay. Researchers have demonstrated photonic BICs in photonic crystal lasers (24), photonic crystal surfaces (25), waveguide arrays...
Recently, researchers theoretically (28) and experimentally (29) demonstrated that photonic BICs existed in low-refractive-index materials on high-refractive-index substrates. These low-refractive-index/high-refractive-index hybrid structures, optical dissipation is prohibited because of the destructive interference of various dissipative channels under certain geometric parameters. Researchers theoretically reported and demonstrated the application of photonic BIC in strong coupling in microwave experiments (30, 31). The researchers demonstrated that strong coupling was related to BICs when the radiative losses were almost suppressed because of destructive interference, and the giant radiative lifetime of a BIC allowed them to Engineer the exciton-polariton lifetime, enhancing it by two orders of magnitude compared to a bare exciton (31).

In this study, by combining CQDs and a low-refractive-index/high-refractive-index (SiO$_2$/Si) hybrid dielectric material, we investigated the PL spectra from CQDs on the hybrid material. Strong coupling and Rabi splitting between a single CQD and a Fabry-Perot (F-P) cavity of the dielectric SiO$_2$/Si material occurred at room temperature. We observed two-peak structure PL spectra and anticrossing of the exciton and photon modes. In our simple structure, the low-refractive-index SiO$_2$ material worked as an F-P resonator, and we treated the bottom silicon as a reflecting mirror. The phase shift at the SiO$_2$/Si RL boundary was $\pi$ due to the high-refractive-index index of silicon, and waves in different leakage channels destructively interfered, drastically decreasing the radiation loss. An F-P BIC occurred in the simple hybrid low-refractive-index/high-refractive-index structure, and we attributed the PL spectral splitting of CQDs on SiO$_2$/Si to strong coupling between excitons and the F-P BIC.

RESULTS

Strong coupling of CQDs with a high concentration

In the experiment, we introduced CQDs with a high concentration of $1 \times 10^{-8}$ M onto a SiO$_2$/Si surface by the drop-casting method, and single CQDs were introduced onto a SiO$_2$/Si surface by the drop-dragging method (32), as mentioned in Materials and Methods. First, we investigated the PL spectral image by a Princeton Instrument (PI) spectrometer with a sensitive CCD using a 40×/0.6 numerical aperture (NA) objective. We could measure the PL spectral image of CQDs at the Fourier plane of the objective using the spectrometer with the CCD. Figure 1A shows a raw angle-resolved spectral image, where the brightness represents the PL intensity, the horizontal axis represents the wavelength, and the vertical axis represents the in-plane momentum (collection angle). We can decompose the spectral image into a series of spectral lines for various in-plane momentums, which correspond to different collection angles of the objective. For the spectral image in Fig. 1A, the working distance between the objective and the surface of the sample is nearly the focal distance, where the collection angle was equal to the angle corresponding to the NA of the objective. The NA of the objective was 0.6, and the corresponding maximum collection angle was 36.9°. Therefore, as we measured the PL spectral image of the CQDs on the surface of SiO$_2$/Si, we could uniformly decompose the spectral lines into angles ranging from $-36.9^\circ$ to $36.9^\circ$.

As the angle increases from 0° to 14° (Fig. 1, B and C, and fig. S1), the F-P mode becomes far from the exciton energy of 1.893 eV (PL central wavelength of 655 nm). The PL spectrum splits at angle of 0°, where the F-P mode is close to the exciton energy. Two spectral peaks appear at 1.937 eV (wavelength of 640.29 nm) with a spectral width of 61.6 meV (20.36 nm) and at 1.862 eV (wavelength of 666.13 nm) with a spectral width of 60.6 meV (21.69 nm). At an angle of 14°, the PL spectrum remains similar to the original spectrum of the CQDs with a single peak, where the CQDs almost do not couple to the F-P mode. As the angle decreases from 14° to 11.8°, a small spectral peak appears in the large energy region at 1.974 eV (628.166 nm) with a spectral width of 46.4 meV (14.77 nm), while a large spectral peak occurs at 1.886 eV (657.476 nm) with a spectral width of 51.8 meV (18.05 nm). As the angle further decreases, the PL intensity of the peak in the short-wavelength region increases. In the angle range less than 14°, the PL spectra all split into two spectral peaks (fig. S1). As we tune the F-P mode to near the exciton transition energy, we observe a strong coupling effect (7.1°, 4.7°, and 0°), where the PL spectrum splits into two obvious PL spectral peaks.

Figure 1D shows some of the measured spectra for various collection angles, where the vertical dotted line represents the original exciton energy peak at 1.893 eV. In the angle range from 14° to 33° (Fig. 1D and fig. S2), the central peak of the F-P mode varies with the collection angle; as we tune the F-P mode close to the exciton transition energy at an angle of 18.9°, the spectrum splits into two spectral peaks, although the peak at low energy is small. As we tune the F-P mode to be resonant with the exciton transition at an angle of 23.6°, the PL spectrum exhibits obvious spectral splitting. As the angle increases beyond the resonance angle to an angle of 30.6° (fig. S2D), the coupling strength decreases, and the splitting of the PL spectrum reduces compared to that at resonance (33). When the angle increases from 14° to 33°, a small peak gradually develops in the low-energy region, and the spectrum gradually splits into two peaks. In this range, the PL spectrum changes from one peak to two peaks. At the angle of 14°, only one spectral peak occurs at 1.894 eV (654.7 nm) with a spectral width of 51.4 meV (17.75 nm). At an angle of 25.9°, two spectral peaks occur at 1.9375 eV (wavelength of 639.99 nm) with a spectral width of 71.5 meV (23.63 nm) and at 1.8628 eV (wavelength of 665.67 nm) with a spectral width of 53 meV (18.94 nm).

Generally, for a passive material without CQDs, the variation of the F-P modes of the SiO$_2$ slab with collection angle will obey the rule given by the following equation

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

where $E$ is the energy of the F-P cavity mode and $n$ is the refractive index of the material.

By fitting the measured PL spectra to a bi-Gaussian function, we could collect the obtained peak energies as a function of angle, which are shown by the scattering symbols in Fig. 1E. We can divide the peak energies into two angle ranges: 0° to 14°, where two peak energies are located on the two sides of the exciton energy, and 14° to 33°, where two peak energies are also located on the two sides of the exciton energy. The two groups of experimental PL peak energies are hybrid states formed by exciton and photons. The following equation can describe the transition energies of the coupled hybrid states (34).
where $E_m$ is the angular-dependent photon mode, $E_{QD}$ is the exciton transition energy, and $\Delta$ is the coupling strength.

After fitting these two peak transition energies as a function of collection angle to the strong coupling formula in Eq. 2, we can obtain an upper polariton line and a lower polariton line, as shown by the solid lines in Fig. 1E. The horizontal dotted line represents the exciton energy, and the dashed line is the F-P mode fitted by Eq. 1. The transition energies of CQDs in the angle range of 14° to 33° exhibit an anticrossing behavior, and the curves show an anticrossing point at an angle of approximately 24.5°. On the basis of the splitting spectra and curves fitted to the strong coupling formula, these two peak energies (transition energies) originate from strong coupling between CQDs and the F-P mode of the SiO$_2$/Si. In the angle range of 14° to 33°, the Rabi splitting energy obtained by fitting the two transitions to the strong coupling formula reaches 74 meV, which is equal to the interval between the two transition energies at resonant angle 24.5°. Similarly, the Rabi splitting energy in the angle range of 0° to 14° is 76 meV.

**Strong coupling between single CQDs and F-P modes**

We also investigated the PL spectra from low-concentration CQDs on SiO$_2$/Si. To explore the polariton dispersion of single CQDs on SiO$_2$/Si, we measured the angle-resolved PL spectral image. A 100×0.85 NA objective focused the pump light, and the PL was collected by the same lens and sent to a PI spectrometer with a Si-CCD detector, which was sensitive enough to measure single-photon emission spectra and spectral images. The measured PL spectra at specific collection angles correspond to specific in-plane momentum in an angle range from 0° to 21°, as shown in Fig. 2 (A to D). As the F-P mode is far from the exciton transition energy, at an angle of 0°, the PL spectrum almost remains as a single peak, and the central peak energy is located at 1.894 eV, which is just the exciton transition energy.
energy. When we tune the F-P mode to be close to the exciton transition energy at an angle of 7°, the spectrum splits into two spectral peaks, although splitting of the peaks is not obvious. When we tune the F-P mode to be nearly resonant with the exciton transition, obviously strong coupling is observed in the PL spectrum at an angle of 10.7°. For the angles at which the exciton energy is near resonant or resonant with the F-P mode, the PL spectrum from single CQDs usually displays a two-peak structure. As the angle increases to beyond the resonance angle, such as at an angle of 21°, the coupling strength decreases and the splitting of the spectra is reduced compared with that at resonance.

By fitting the PL spectra to a bi-Gaussian function, we can map the polariton dispersion relation of the single CQDs, as shown in Fig. 2E. The minimum peak distance between the two prominent energy.

![Fig. 2. Rabi splitting from a single CQD on SiO₂/Si.](image-url)

PL spectra from a single CQD on 3-μm SiO₂/Si for different collection angles: (A) 0°, (B) 7°, (C) 10.7°, and (D) 21°. The green solid lines are lines fitted by a bi-Gaussian function. (E) Variation in the split PL peak energies from a single CQD on 3-μm SiO₂/Si with angle (°). The scattered symbols show the experimental results of spectral peaks. The solid lines are fits to the experimental data by the strong coupling formula. The dotted line is the exciton energy, and the gray line is the F-P mode. (F) Antibunching phenomenon of a single CQD on SiO₂/Si. The horizontal dotted line represents the value of 0.5 for the second-order correlation function.
features occurs at an angle of approximately 12.5°. The two polariton branches feature the characteristic anticrossing behavior with a Rabi splitting of 42 meV at the angle of 12.5°, where the F-P cavity photon is resonant with the exciton (35).

The following formula gives a criterion for strong coupling (36)

$$g^2 > (\gamma_r - \gamma_i)^2/6$$

where $g$ is the exciton-photon coupling parameter, $\gamma_r$ is the linewidth of the cavity, and $\gamma_i$ is the linewidth (corresponding to the emission rate) of the exciton of CQDs. From the split PL spectra for single CQDs, RL spectra for the F-P cavity and their fitted parameters in Fig. 2, $\gamma_r = 40$ meV, $\gamma_i = 64$ meV, $g = 42$ meV, and $g = 42 > \frac{(\gamma_r - \gamma_i)^2}{6} = \frac{40^2}{6} = 6$; therefore, the formula (Eq. 3) is fulfilled, and strong coupling occurs.

To determine whether the low-concentration CQDs for strong coupling are single CQDs, we characterized the second-order correlation function using a Hanbury Brown and Twiss (HBT) setup and a time-correlated single-photon counting (TCSPC) system. We measured the relationship between the coincidence count rate (normalized) and the delay time, as shown in Fig. 2F, where an obvious dip appears at zero time delay. When we fitted this normalized coincidence curve by the second-order correlation function $g^{(2)}(t)$, the value of $g^{(2)}(0) < 0.4$ at zero time delay demonstrated that the CQD used is a single QD, and the corresponding PL is single-photon emission.

We also measured PL spectral splitting from some other single CQDs on 3-μm SiO$_2$/Si, as shown in fig. S4 (A to D). For comparison, fig. S4E shows a PL spectrum of CQDs without PL splitting on 3-μm SiO$_2$/Si. We also measured the PL intermittence (fluorescence blinking) from a CQD using the same low-concentration CQDs on SiO$_2$/Si, as shown in fig. S4F. The blinking is obvious, which demonstrates that the measured CQD is a single QD.

DISCUSSION

For the planar SiO$_2$/Si cavity that we used, the mode volume is comparable to that of the open microcavity formed by the silver mirror (15, 16) or the distributed-Bragg-reflection (DBR) cavity (17), where the mode volume is similar to or larger than that we used here. The difference is that the dielectric membrane of SiO$_2$/Si used in this study has a low-refractive-index/high-refractive-index structure. The interface between the low-refractive-index and high-refractive-index materials forms an RL mirror with a high reflectivity. The membrane with a low-refractive index will form an F-P cavity with a bottom RL face of the low-refractive-index/high-refractive-index interface and a top RL face of the air/low-refractive-index interface. In planar semiconductor waveguide structures with QDs or quantum wells, only 1 to 4% of the light can escape from the planar surface because of the large difference between the refractive indices of the semiconductor and air. However, in our case, with CQDs on the surface of the SiO$_2$ membrane, the PL from the CQDs couples to the bottom F-P cavity of SiO$_2$ through an evanescent wave. When the SiO$_2$ membrane is sufficiently thick, the F-P cavity can enhance the emission from the CQDs. Therefore, we can easily detect the PL, including the PL from strong coupling. The membrane with a low-refractive index is on a high-refractive-index bottom substrate, which is an important ingredient for producing strong coupling. Whether the hybrid material structure contains other physical mechanisms that produce strong coupling is worth exploring.

We measured both PL and RL spectral images for CQDs on 4-μm SiO$_2$/Si by a PL spectrometer with a CCD using a 40×/0.6 NA objective as shown in Fig. 3 (A and B), where the pump distance was less than the focal length. The PL spectral image splits into two PL peaks near 655 nm, while the RL spectral image shows only one peak near 655 nm. We extracted the spectral images as a series of spectral lines at different collection angles, as shown in Fig. 3 and fig. S5. At an angle of 0°, as shown in Fig. 3C, two PL spectral peaks appear at 1.9297 eV with a width of 47.95 meV (15.97 nm) and at 1.8418 eV with a width of 49.4 meV (18.06 nm). An RL peak at 1.8855 eV with a width of 64.6 meV (22.53 nm) is located between the two PL spectral peaks. The other spectra in Fig. 3D at an angle of 8.3° show similar phenomena. We fit the PL spectra at different angles to a bi-Gaussian function. Figure 3 (E and F) shows the spectral peak energies as a function of collection angle. In the angle range from 0° to 15°, both a high-energy branch and a low-energy branch appear. We also collected the peak energies of the RL spectra at the corresponding angles and compare them in Fig. 3F (shown by the hollow circles). We can well fit the RL spectral peak energies to the F-P mode formula of Eq. 1, which demonstrates that the RL spectral peaks of the SiO$_2$/Si material correspond to the F-P modes. The RL spectral peaks are located between the two polariton peaks, close to the exciton energy, which means that the photon mode is nearly resonant with the exciton energy in the measured angle range; therefore, the PL spectra all obviously split. By fitting these two PL peak energies to the strong coupling formula of Eq. 2, we can obtain the Rabi splitting energy as 85 meV. In Fig. 3F, the spectra at these collection angles all split into two spectral peaks. One PL spectral peak of the exciton splits into two PL spectral peaks because of the strong coupling between the exciton and the F-P mode. Because of the strong coupling effect, we regard the high-energy branch as the upper polariton and the low-energy branch as the lower polariton.

To explain the above strong coupling phenomena, Fig. 4 (A and B) shows a schematic of the low-refractive-index/high-refractive-index material, where a slab of SiO$_2$ with thickness $h$ sits on a Si substrate with a thickness of 500 μm and the refractive indices are $n_1 = 1$, $n_2 = 1.45$, and $n_3 = 3.4$. For this low-refractive-index material on a high-refractive-index substrate, the modes in the low-refractive-index membrane are usually above the light line; thus, the modes are in the continuum. In our low-refractive-index/high-refractive-index hybrid structure, we can consider the high-refractive-index (silicon) substrate to be a reflecting mirror. The upper low-refractive-index membrane (SiO$_2$) supports an F-P mode along the vertical $y$ direction. Light in the F-P cavity leaks in the upper direction into the air (channel A) and in the lower direction into the high-refractive-index material (Si substrate). Since the refractive index of Si is much higher than that of SiO$_2$, the main part of the down-going light is reflected, and part of the light is transmitted through the above SiO$_2$ membrane into the air (channel B), interfering with the light from channel A. Because the boundary of the high-refractive-index material under the low-refractive-index material (SiO$_2$/Si) adds a phase shift of $\pi$ to the RL wave, the light from channel B differs in phase from the light from channel A by $\pi$, the destructive interference drastically decreases the radiation loss, and then, the F-P BIC in SiO$_2$/Si occurs. The magnitudes of the waves from the two channels differ because of the different RL coefficients of the SiO$_2$ surface and the Si surface. If the waves in these two channels have the same magnitude, then the destructive interference will
eliminate the radiation loss and the lifetime of the mode will be infinite (37).

We consider the case of vertical light incidence. When \( k_y d = 2m\pi \), an F-P resonance occurs for transverse magnetic (TM) modes, where \( k_y \) is the wave vector of the \( y \) component of the TM modes, matching that of the TE modes; \( d \) is the thickness of the low-refractive-index film; and \( m \) is an integer. According to (28, 29), the loss \( \alpha \) of the TM mode is inversely proportional to the attenuation length, which we can express as

\[
\alpha \propto \cos^2 \left( \frac{k_y d}{2} \right) \frac{d^2}{\lambda^2} \quad (4)
\]

When \( k_y d = 2m\pi \) is satisfied, \( \alpha \) becomes infinitesimal, and then, the BIC correspondingly occurs. For the hybrid structure shown in Fig. 4A, because of the refractive index \( n_3 \) being >\( n_2 \), when the transverse electric (TE) light is reflected because of the interface between \( n_1 \) and \( n_2 \), the phase shift will be \( \pi \), and then, the incident TE light becomes reflected TM light. Because TM light can meet the F-P resonance condition, the BIC of the TM light occurs in the hybrid structure. When the light is incident with an angle, such as \( \theta \) relative to the membrane with refractive index \( n_2 \), the wave vector of the \( y \) component for the TM light is \( \frac{k_y h}{\sin^2 \theta} \), and the wave vector for the TE light is \( \frac{k_x h}{\sin \theta} \). This means that both the TE light and the TM light can form F-P oscillations. Therefore, for both TE light and TM light, the F-P mode can form a BIC with a defined incident angle, and an F-P BIC can occur in the low-refractive-index/high-refractive-index hybrid structure.

To verify the BIC occurrence, we designed and simulated a SiO_2/Si waveguide with a width of 3 \( \mu \)m, a height of 5 \( \mu \)m, and a length of 1 mm of SiO_2, and the transmittance of the TE mode is approximately 92\%.

Fig. 3. PL spectral splitting compared with the RL spectra. Strong coupling between the CQDs and 4-\( \mu \)m SiO_2/Si based on a characterization of the spectral image. (A) PL spectral image. (B) RL spectral image of 4-\( \mu \)m SiO_2/Si. Extracted PL spectra compared with the RL spectra for different angles: (C) 0° and (D) 8.3°. (E) Peak energies as function of collection angle. (F) Angular dispersion of the coupled exciton transition energies in the F-P cavity formed by SiO_2/Si. The horizontal dotted line at 1.893 eV is the CQD exciton transition energy in a solvent. The scattered symbols show the experimental results. The gray line shows the angular dispersion of the photon mode for the F-P cavity without CQDs. The solid line is a fit to the data using the strong coupling formula.
Another case of a high-refractive-index material is an silicon-on-insulator (SOI) material, as shown in fig. S7 (A and B), where the refractive indices are \( n_1 = 1, n_2 = n_4 = 1.45, \) and \( n_3 = n_5 = 3.4; \) the top SiO\(_2\) membrane thickness is \( h = 3 \) \( \mu \text{m}; \) the SiO\(_2\) waveguide width is \( w = 1.5 \) \( \mu \text{m}; \) and the thickness of the embedded SiO\(_2\) is \( t = 1 \) \( \mu \text{m}. \) Figure S7C shows the calculated transmission and mode distribution for the SiO\(_2\)/SOI waveguide. We found that the transmission after a length of 1 mm almost does not decrease. The transmittance of the TE mode of the SiO\(_2\) waveguide is approximately 98.8%, and the propagation loss is 0.53 dB/cm. Figure S7D shows the TE field distribution at the exit sections of the waveguide after 1-mm propagation, where the field of the optical mode is still mainly concentrated in the SiO\(_2\) waveguide. Therefore, if we adopt a thin Si membrane as a mirror reflector, such as a low-refractive-index SiO\(_2\) waveguide on an SOI material, then the BIC can easily form. We believe that the use of this hybrid low-refractive-index/SOI structure will be a good way to produce a BIC, and it is a promising integration platform for strong coupling between an emitter and a BIC.

Compared with the structure in (38), the low-refractive-index SiO\(_2\) membrane is a planar waveguide (F-P cavity), similar to the side-coupled waveguide in (38), and the Si surface is equivalent to a mirror reflector (shown in Fig. 4B), similar to the semi-infinite waveguide array in (38), in which the existence of an F-P BIC was verified theoretically and experimentally. We calculated the RL spectra of a 3-\( \mu \text{m} \) SiO\(_2\)/Si material and 3-\( \mu \text{m} \) SiO\(_2\)/air at a collection angle of 0°, as shown in Fig. 4C. According to the calculation, the RL spectrum of SiO\(_2\)/Si is opposite to that of SiO\(_2\)/air because the phase shift for the RL light between the interface of SiO\(_2\)/Si is \( \pi \). From other experimental observations (17), the conventional F-P mode for a high-refractive-index material surrounded by a low-refractive-index material is characterized by a dip in its corresponding RL spectrum (e.g., SiO\(_2\)/air in Fig. 4C). However, the RL spectral peaks from SiO\(_2\)/Si in Figs. 3 (A to D) and 4C correspond to F-P modes. That is, the F-P modes are exhibited as spectral peaks in the RL spectra of the low-refractive-index/high-refractive-index hybrid structure. This feature is consistent with a previous analysis of an F-P BIC (26); as the F-P resonance condition is approached, the transmission coefficient decreases and approaches zero, while the RL coefficient increases and approaches 1. As shown in Fig. 3, the measured central RL spectral peak, representing the F-P mode, is located near the center of the two split PL peaks. These F-P modes in SiO\(_2\)/Si are BICs that occur in the SiO\(_2\) F-P cavity on Si. The Q factor of the BIC is not high but is comparable to that of the PL spectrum of CQDs, which is important for generating strong coupling. As the RL at the Si boundary is not perfect and the magnitudes of the two leakage channels are different, the Q factor of the BIC is not high.

We measured the spectra in Fig. 4D and fig. S8 by an angle-resolved PL setup similar to that in (39). We measured the PL spectrum from CQDs on 3-\( \mu \text{m} \) SiO\(_2\)/Si and the RL spectrum of 3-\( \mu \text{m} \) SiO\(_2\)/Si for the same collection angle and the PL spectrum from CQDs on a silicon substrate; the results are compared in Fig. 4D. The spectrum of the CQDs on the silicon substrate does not split; the spectrum maintains a single peak that we can fit by one Gaussian function.
with a center at 655.01 nm (1.8931 eV). This spectrum represents the original exciton emission spectrum of the CQDs. The vertical dashed line represents the exciton peak emission wavelength. The RL spectral peak of 3-μm SiO₂/Si at 657.54 nm overlaps with the exciton emission wavelength center at 655 nm. From the measured PL spectrum of CQDs on 3-μm SiO₂/Si, two PL spectral peaks are located at 641.99 and 672.67 nm, the PL spectrum splits, and Rabi splitting occurs.

When we change the low-refractive-index SiO₂ above to another material with a low refractive index, such as SU8 on a Si substrate, after CQDs are introduced onto SU8, the PL spectrum at a particular angle also splits, as shown in fig. S8A; a RL spectrum peak from SU8/Si, representing the F-P mode, is located at the center of the two split PL peaks. If the refractive index of the material under the membrane is smaller than that of the above membrane, such as for SU8/SiO₂, then we cannot observe spectrum splitting (fig. S8B). Moreover, from a calculation, the electrical field on the upper film surface of the hybrid low-refractive-index/high-refractive-index material is stronger than that of the high-refractive-index/low-refractive-index material. For CQDs on SU8/Si, at a particular collection angle, near the exciton emission wavelength, the PL spectrum splits, in which an RL peak appears near the exciton emission wavelength, while the PL spectra in the long-wavelength region and short-wavelength region are significantly enhanced because of the F-P resonance corresponding to the PL peaks (figs. S8, C and D). In our experiment, the PL of CQDs on the low-refractive-index material on a high-refractive-index material is significantly enhanced compared to the PL of CQDs on silicon or a thick glass, as shown in fig. S8E. The light leakage into the high-refractive-index substrate is low. The enhancement in the PL is also related to the F-P BIC.

The material structure used in this study is simple: We introduced CQDs onto a dielectric membrane with a low refractive index (SiO₂ and SU8) on a substrate with a high refractive index (Si) and observed PL spectral splitting for the CQDs; however, the PL spectra from the CQDs on a silicon substrate and on a high-refractive-index/low-refractive-index hybrid material do not split. These results demonstrate that the PL spectral splitting derives from the F-P cavity mode supported by the low/high-refractive-index structure. The F-P BIC is believed to form for a low-refractive-index material on a high-refractive-index substrate (28, 29). In this hybrid material, the optical loss drastically decreases because of the destructive interference among relevant light waves with particular geometric parameters. Specifically, the RL spectral peaks that we measured and calculated for SiO₂/Si and SU8/Si are not simple F-P modes but F-P BICs (38, 40) in the hybrid structure. The PL is enhanced because of the Purcell effect based on the F-P BIC in the low-refractive-index/high-refractive-index material, and the Rabi splitting that we obtained on the basis of the PL spectra is due to the strong coupling between the exciton in the CQDs and the F-P BIC in the hybrid structure of the low-refractive-index/high-refractive-index material. By combining BICs with dielectric cavities, we can control single CQDs or other quantum emitters with a long lifetime to enhance the coupling strength between emitters and BICs to the point where it exceeds the rates of quantum decoherence in the system (30), and then, we can relatively easily reach quantum coherence. This low-refractive-index/high-refractive-index configuration overcomes the fundamental limitations of photonic architectures and material systems (28) for photonic integrated circuits and quantum photonics at room temperature.

In conclusion, we measured the PL spectral images from CQDs on SiO₂/Si and the RL spectral images from SiO₂/Si, and we observed and analyzed PL spectral splitting at various collection angles. We obtained the polariton dispersion based on the PL spectral splitting peak energies, which exhibited an anticrossing phenomenon. The Rabi splitting was 42 meV for single CQDs at room temperature. We analyzed the occurrence of the F-P BIC in the low-refractive-index/high-refractive-index material and the interaction between CQDs and BICs. We regarded the PL spectral splitting as strong coupling between the CQDs and the F-P BIC in the low-refractive-index material on the high-refractive-index substrate. We have found the first evidence of strong coupling between a single photon and a BIC in a dielectric membrane based on PL spectra at room temperature, which will provide an important platform for quantum integrated circuits and quantum information sciences.

**MATERIALS AND METHODS**

We used CQDs from Thermo Fisher Scientific in our experiment. We introduced CQDs with a high concentration of 1 × 10⁻⁸ M onto a SiO₂/Si surface by the drop-casting method. To obtain single-photon emission from a single CQD, we introduced CQDs at a low concentration of 1 × 10⁻⁹ M onto a SiO₂/Si surface by the drop-dragging method (32). We measured the PL spectra under a confocal system and detected and analyzed spectra by a Pı spectrometer. We constructed an angle-resolved PL measurement setup that images the objective Fourier plane onto a spectrometer with a CCD chip in the far-field imaging configuration (21). We measured the PL spectra from the samples at room temperature under excitation by a nonresonant continuous wave laser at a wavelength of 532 nm with an excitation power of 1.5 mW. We measured the spectral image with an objective and then decomposed it into the spectra at different collection angles. The magnification of the objective was 100× with an NA of 0.85, which corresponds to the collection angle range of 0° to 58.2°. Another objective was a 40x objective with an NA of 0.6, corresponding to an angle range of 0° to 36.9°. The PL at various collection angles was projected onto the momentum range on the CCD. We could extract the PL spectra at various angles from the spectra image by the CCD chip. By fitting the PL spectra at various angles to a bi-Gaussian function, we could map the polariton dispersion relation. As the pump distance, i.e., the distance between the surface of the focus objective and the surface of the sample, changed, the PL intensities of the spectral peaks and the spectral structure changed. With changing pump distance, the collection angle range changed. When the pump distance was smaller than the focal length, the collection angle range became smaller than the angle range corresponding to the NA.

In the PL experiment of single CQDs, to cover a large emission angle, we used a high magnification (100×) microscope objective with an NA of 0.85. The polariton’s in-plane momentum is proportional to sin(θ), with θ being the PL emission angle, which allows us to project a collection angle range (a momentum range) of up to the angle range corresponding to the NA onto the CCD connected to the spectrometer in the far-field imaging configuration. We first used this spectral imaging method to obtain angle-resolved spectra for the characterization of single QDs here, which was demonstrated to be an effective method. We measured PL blinking phenomenon and antibunching effects of single-photon emission from single CQDs with an HBT setup and a TCSPC system.
