

Supplementary Materials for
**Single-photon controlled phase shifts with colloidal quantum dots at
room temperature**

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Sci. Adv. **12**, eaee1340 (2026)
DOI: [10.1126/sciadv.aee1340](https://doi.org/10.1126/sciadv.aee1340)

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S1 Mode modulation of the photonic crystal-Fabry-Pérot (PC-FP) hybrid cavity

The simulation model was established using the Finite-Difference Time-Domain (FDTD) method. An electric dipole source was adopted to simulate the emission of quantum dots. The simulation unit was defined as the region marked by the red dashed box in Fig. 1B. Periodic boundary conditions were applied in the x and y directions, with periods of 674 nm and 337 nm, respectively, and the diameter of the photonic crystal (PC) cylinders was set to 196 nm. Perfectly matched layer (PML) boundary conditions were employed in the z-direction. The material parameters of SiO₂ and Cr were taken from the built-in database of the simulation software, while the refractive indices of SiN and SU-8 were set to 1.95 and 1.57, respectively. We scanned the thickness of SU-8 in the range of 2.0–3.0 μm , and the wavelength-dependent Purcell factor of the system is shown in Fig. S1A. It shows that multiple cavity modes exist within the wavelength range of 730–830 nm, and the modes exhibit a red shift as the SU-8 thickness increases. Considering that the microcavity mode should match the central emission wavelength of colloidal quantum dots (CQDs) at 780 nm, we finally selected an SU-8 thickness of 2.37 μm .

With the SU-8 thickness fixed at 2.37 μm and the PC cylinder diameter set to 196 nm, we simulated the electric field distribution at the mode wavelength of 780 nm. The electric field distribution of the PC-FP cavity structure under excitation by an X-polarized electric dipole source is shown in Fig. S1B. Under X-polarization, the electric field is mainly localized around the Cr layer, with an electric field enhancement factor of only 5. In contrast, under Y polarization as shown in Fig. 1E, the electric field is distributed within the SU-8 layer, forming a stable BIC with an electric field enhancement factor of 12. Considering that the CQDs layer (red circles in Fig. 1B) is located on the top surface of SU-8, the Y-polarized source is more suitable for our requirements.

Furthermore, we simulated the Purcell enhancement characteristics of the system with and without the PC structure, as shown in Fig. S1C. The system without the PC structure consists only of a simple SU-8 slab on SiO₂, and its simulated Purcell factor spectrum is represented by the blue curve in Fig. S1C. It shows that after removing the modulation effect of the PC structure, the Purcell factor of the system decreases significantly, with almost no obvious Purcell peaks and no distinct BICs observed. For the structure with PC modulation, the Purcell factor spectrum is shown by the black curve. Purcell enhancement appears at 726 nm, 780 nm, and 838 nm, with the Purcell factor reaching 4.6. The Q factor at the wavelength of interest (780 nm) was calculated using the Q-factor analysis group in the simulation software and was found to be 63.

The Q-factor of the microcavity mode around 780nm measured in our experiment is approximately 25, whereas the Q-factor obtained from the cavity mode of the Purcell enhancement spectrum simulated by FDTD method is 63. Considering the effects of fabrication imperfections and surface roughness, the theoretical value is somewhat higher than the experimental one, which is within a reasonable range. In our calculation of the conversion from pump power to photon number, we used a Q-factor of 100 for the microcavity mode. This Q-factor corresponds to the pump wavelength of 532 nm. For our planar microcavity structure, the Q-factor generally increases at shorter wavelengths; therefore, it is reasonable to take $Q = 100$ for the 532 nm pump light.

The electric field distribution of the PC-FP cavity structure at 780 nm, as excited by a Y-polarized electric dipole source, was simulated using FDTD with periodic boundary conditions in the transverse plane and PML boundaries along the z-direction (Fig. 1E).

The simulation reveals that the electric field is predominantly confined within the SU-8 layer, where 5.5 bright lobes are resolved as a result of the coupling between the z-directed FP resonances and the in-plane photonic crystal modes, with an SU-8 thickness of 2.37 μm . This lobe number deviates from the standard FP standing-wave prediction, reflecting the strong modulation imposed by the underlying photonic crystal. A field antinode—that is, the maximum field intensity—is located at the top surface of the SU-8 layer, which allows for enhanced emission and strong coupling when colloidal quantum dots (CQDs) are placed at this position.

In the experimental configuration, CQDs are deposited near this top antinode at the SU-8 interface, where pronounced local field enhancement is present. Radiation from the dipole source (indicated by the white concentric circles in Fig. 1E), after being coupled into the microcavity, is modulated by the cavity such that its radiation pattern conforms to the mode distribution of the hybrid structure, yielding a non-zero spatial overlap with the cavity mode—that is, the CQD emission resonantly couples with the microcavity mode. Although the spatial extent of a conventional dipole is far smaller than the entire cavity mode volume and thus cannot achieve full spatial overlap with the entire high-order standing wave over the whole cavity, strong exciton–photon coupling can nevertheless be established through the locally enhanced electric field. In addition, the evanescent field extending from the SU-8/air interface into the air layer overlaps with the surface-layer quantum dots, which, together with the dominant cavity field at the top antinode, supports stable light–matter strong coupling between the CQDs and this high-order hybrid longitudinal mode along the z-axis.

S2 Controlled phase measurement of high concentration CQDs

We comprehensively investigated the influence of the probe-light wavelength on the controlled phase modulation effect generated by pump light. Figs. S2–S5 present the original interference signals excited by pump light at corresponding characteristic powers, as well as the comparison results of controlled phase changes with and without pump-light excitation, for probe-light wavelengths of 770 nm, 780 nm, 793 nm, and 830 nm, respectively. In Figs. S2A–S5A, the black hollow scatter points correspond to the measured original interference signals under three different pump powers. The specific power parameters are as follows: 10.94 μW , 48.6 μW , and 158.2 μW for 770 nm; 7.4 μW , 89.4 μW , and 400.6 μW for 780 nm; 19.76 μW , 101 μW , and 222.4 μW , and 824.0 μW for 793 nm; and 42.2 μW , 436.6 μW , and 865.2 μW for 830 nm. The corresponding average pump photon numbers n_c are as follows: 0.23, 1.02, and 3.33 for 770 nm; 0.16, 1.88, and 8.43 for 780 nm; 0.42, 2.13, and 4.68 for 793 nm; and 0.89, 9.19, and 18.20 for 830 nm. It can be clearly observed from the experimental data that the intensity of the interference signal at each wavelength strictly follows the sine function law, showing a typical periodic fluctuation characteristic. To accurately characterize this variation behavior, the sine function was uniformly used to fit the experimental measurement data at each wavelength, and the fitting results are presented as the blue curves in each figure. Comparing the fitting curves under different pump powers, it is found that with the gradual increase of pump power, the sine fitting curves corresponding to all wavelengths show an obvious right-shift trend. This phenomenon is a direct reflection of the regular change in the controlled phase induced by pump-light excitation, confirming the universal effectiveness of pump power in regulating the phase of interference signals. To more intuitively reveal the phase modulation effect of pump power at each wavelength, interference curves with no pump light and with two characteristic pump powers were selected for precise comparison for

each probe-light wavelength (Figs. S2B–S5B, S2C–S5C). In each figure, the blue curve corresponds to the interference signal without pump-light excitation, while the red curve represents the controlled interference signal obtained after excitation by pump light at the characteristic power. The phase difference between the interference curves with and without pump-light excitation was quantitatively calculated via sine function fitting. The specific pump powers and corresponding phase changes are as follows: at 770 nm, the phase change is 1.13π under an average pump photon number of 1.77 and increases to 1.4π at $n_c = 2.08$; at 780 nm, it is 0.96π at $n_c = 0.37$ and rises to 1.4π at $n_c = 1.88$; at 793 nm, it reaches 0.4π at $n_c = 1.11$ and further increases to 1.39π at $n_c = 8.39$; and at 830 nm, it is 1.05π at $n_c = 4.78$ and grows to 1.42π at $n_c = 18.20$.

We systematically investigated the correlation between pump power and controlled phase change. By quantifying the phase change values relative to the unpumped state ($0 \mu\text{W}$) for different pump powers across all probe-light wavelengths, we established the evolutionary relationship of phase change with pump power, as illustrated in Figs. S2D–S5D. In each figure, the red scatter points represent the measured phase change data under varying pump power conditions, revealing a distinct exponential growth trend of phase change with increasing pump power for all probe-light wavelengths. The blue curves correspond to the fitting results based on an exponential function, and their excellent agreement with the measured data at each wavelength not only validates that the phase modulation effect of pump power universally adheres to this exponential growth behavior but also provides reliable experimental evidence and data support for future precise phase modulation across different wavelength scenarios.

S3 Controlled phase measurement of single CQD

A single CQD was site-specifically introduced via the soft probe method. Fig. S6 presents the experimental setup of the soft probe method, microscopic images, and scanning electron microscopy (SEM) micrographs. The image contains five groups of PC and transmission waveguide structures with different dimensional parameters, among which the fourth group clearly illustrates the operation of introducing a CQD via the soft probe. In addition, the PC structure highlighted by the red dashed box in the figure is shown in the magnified SEM image in Fig. S6B.

Based on strong coupling between single CQD and FP cavities, we comprehensively investigated the dependence of the probe-light phase on pump power at different probe-light wavelengths. Figs. S7–S9 present the original interference signals under pump-light excitation with corresponding characteristic powers at probe wavelengths of 770 nm, 780 nm, and 790 nm, as well as the comparative results of controlled phase changes with and without pump-light excitation. In Figs. S7A–S9A, the black hollow scatter points correspond to the measured interference signals under three different pump power conditions, corresponding to specific average photon numbers: 0.02, 0.29, and 0.69 at 770 nm; 0.28, 0.57, and 1.07 at 780 nm; 0.08, 0.49, and 0.96 at 790 nm, respectively. From the experimental data, it can be clearly observed that the interference signal strength at each wavelength strictly follows the sine function form, exhibiting typical periodic fluctuation characteristics. To accurately characterize this change pattern, a sine function was uniformly used to fit and analyze the experimental measurement data at each wavelength. The fitting results are presented as blue curves in each figure. Comparing the fitting curves under different pump powers, it is found that as the pump power changes, the sine fitting curves corresponding to all wavelengths undergo significant shifts. To more intuitively reveal the phase modulation effect of pump power at

various wavelengths, for each probe-light wavelength, we selected the interference curves under three conditions for precise comparison: without pump light and under two characteristic pump powers (Figs. S7B–S9B, S7C–S9C). The blue curves in each figure correspond to the interference signal without pump-light excitation, while the red curve represents the controlled interference signal after characteristic power pump-light excitation. The phase difference between the interference curve with and without pump-light excitation was quantitatively calculated through sine function fitting. The specific averaged pump photon numbers and corresponding phase changes are as follows. At 770 nm, with 0.37 as the reference pump photon number, the phase changes are -0.12π and -0.10π under pump photon numbers of 0.53 and 0.69, respectively. At 780 nm, the phase change under a pump photon number of 0.09 is 0.47π , which increases to 1.04π at $n_c = 0.88$. At 790 nm, the phase change is 0.33π under a pump photon number of 0.56, which increases to 1.03π at $n_c = 0.96$.

We systematically investigated the correlation between pump power and controlled phase change based on strong coupling between single CQDs and PC-FP cavities. By calculating the phase change at different pump powers relative to the state without pump light at various probe-light wavelengths, we obtained the evolution relationship of phase change with pump power (photon numbers), as shown in Figs. S7D–S9D. The red scatter dots in each figure represent the measured phase change data under different power conditions. The results show that, at all wavelengths except 770 nm, the phase change exhibits a significant exponential increase with rising pump photon number. The blue curve is the fitting result based on an exponential function, showing good consistency with the measured data at various wavelengths. This not only verifies that the modulation effect of pump power on phase generally follows an exponential growth behavior but also provides a reliable experimental basis and data support for precise phase regulation across different wavelength scenarios in the future.

S4 Evidence for single CQD

A single CQD can sustain sufficient emission intensity for either phase-shift measurements (typically >1 hour) or antibunching measurements (typically several tens of minutes), but not both sequentially on the same emitter, due to photobleaching. In our experiments, priority was given to the phase measurement, and thus it was not possible to obtain $g^{(2)}(0)$ data (due to photobleaching) on the same CQD used for the phase characterization. Therefore, in the experimental design, priority was given to ensuring the completeness and reliability of the core experiment—the phase measurement.

Evidence I: Blinking and spatial isolation characteristics of the CQD

On the CQD used for phase measurement, we quickly recorded its blinking behavior before the phase measurement (Fig. S10). The PL blinking on/off ratio was found to be $\sim 10^4$, with the bright-state intensity reaching as high as 10^4 counts/s. Moreover, the dark-state duration was measured to exceed 5 s—a feature that is impossible for a cluster of quantum dots, as clustered emitters cannot exhibit such long dark-state periods. The blinking trace shown was obtained after subtracting a constant background offset, which is mainly attributed to the residual fluorescence from the surrounding environment or substrate.

In addition, spatial isolation of the quantum dot was verified on the confocal micro-spectroscopy setup. During emission measurement, when the stage was laterally moved by more than 1 μm (the focal spot size of the 100 $\times$ objective is $\sim 1 \mu\text{m}$) away from the

position of maximum emission intensity, the CQD emission rapidly decayed and disappeared, confirming that the quantum dot is an isolated single particle.

Evidence II: Systematic single-emitter validation of the CQD system

Prior to this work, extensive antibunching characterization of single CQDs was performed. The colloidal quantum dots used in this study (Thermo Fisher Scientific) have been systematically characterized by our research group. Our group's work on CQD-based single-photon sources was consistently used CQDs from this same supplier. Antibunching measurements were performed for CQDs on various substrates (glass, SiO₂/Si, sapphire, SU-8, photonic crystal structures, etc.), yielding single-emitter behavior, with $g^2(0) < 0.5$ in the vast majority of measurements and best values reaching $g^2(0) < 0.02$. Some results were published in peer-reviewed papers (3, 36–41) reporting blinking and antibunching measurements on these CQDs. In those works, dozens of independent single-CQD measurements have been performed, using material sources and preparation protocols identical or similar to those in the present work.

This extensive experimental foundation elevates the single-CQD claim from anecdotal evidence to a systematically validated property of our well-characterized sample system. The blinking CQD measured in this work was prepared using the same protocol, and its blinking characteristics (on/off ratio, intensity distribution) are indistinguishable from those of confirmed single emitters.

Evidence III: Blinking in the literature

The use of PL blinking as evidence for single-CQD emission is widely accepted in the field. Fluorescence blinking is a typical optical signature of isolated single quantum dots, as confirmed by multiple classic experiments: multi-quantum-dot aggregates, due to statistical averaging of the on/off behavior of individual dots, typically exhibit no clear long-duration complete OFF dark states, but rather only minor intensity fluctuations (51–54).

Notably, an important reference for this work—the pioneering study on phase control with single InGaAs quantum dots—also mentions blinking behavior of single quantum dots and does not provide antibunching measurements for the specific QD used for phase characterization (11). This indicates that in scenarios where the integrity of the core physics experiment must be prioritized, relying on comprehensive information including blinking features is a reasonable and long-established experimental strategy in the field.

S5 Feasibility verification of the experimental measurement method

In our experiments, the controlled phase shift was obtained by acquiring multiple interference curves while scanning the piezoelectric positioning stage, with each phase point taking several minutes to measure. Consequently, a full set of measurements from low to high pump power for a single figure required approximately 30 minutes. To minimize potential photobleaching effects of the CQDs and to facilitate timely comparison of the controlled phase shifts at different wavelengths, we performed only one measurement of the controlled phase shift per wavelength and then successively measured the controlled phase shifts at different wavelengths. Although the phase measurements were performed only once, we have experimentally demonstrated the repeatability and reliability of the results through the following evidence.

As a test experiment, we performed a comparison experiment: in a strong coupling system, a significant controlled phase shift was observed under pump-light excitation (as shown in the main text and Figs. S2-5), whereas in a similar CQDs–microcavity system without pumping, no noticeable phase change was observed over a similar measurement duration, as shown in Fig. S11(A). In the CQDs–microcavity strong coupling system, a significant controlled phase shift was observed under optical pumping. Physically, strong coupling arises when the cavity mode is tuned close to the exciton resonance of the CQDs, enabling the observation of the corresponding phase change. However, in an empty cavity without CQDs, no noticeable phase change was observed over a similar measurement duration even with similar pump powers, as shown in Fig. S11(B).

As a validation of repeatability, we performed two independent measurements of the phase shift as a function of pump power using CQDs coupled to a microcavity and observed consistent trends in the phase variation with pump power. We performed repeated measurements of the controlled phase change for two types of strong-coupling systems—namely, a microcavity containing high-concentration CQDs and a single CQD–microcavity system—as shown in Figs. S12(A) and S12(B). The results show that, across different measurement runs within the same system, the measured data follow the same trend as the pump power is varied. This confirms the reliability of our experimental setup and measurement system for repeatable measurements of the controlled phase change.

S6 Device fabrication deviation and gate fidelity

In our sample replication process, multiple dimensional parameters are designed, and by adjusting the exposure dose and photonic crystal structural parameters, microstructures with high dimensional accuracy can be fabricated, achieving a dimensional deviation within 2.5% (<5 nm).

Based on the <5 nm dimensional deviation evaluated from the actual SEM images of structures fabricated by electron beam lithography (EBL) and reactive ion etching (RIE), the relative impact on cavity mode shift was assessed. We simulated the Purcell factor of composite cavities with varying photonic crystal radii (Fig. S13). FDTD modeling reveals that the structural diameter deviation of <5 nm (deviation ratio <2.5%) translates into a cavity mode shift of ≤ 0.2 nm. This mode shift has a negligible effect on the phase shift in single-stage operations. As shown in Fig. 2D of the main text, within a 20 nm spectral window around the resonance center (770–790 nm), the probe wavelength lies in a steeply varying phase-response regime, rendering the system largely insensitive to such a small mode shift.

Whether this cavity mode shift produces a significant effect depends on the specific application scenario and the required fidelity. With a mode shift of ≤ 0.2 nm, the corresponding controlled-phase deviation is less than 0.016π (estimated from Fig. 4B of the main text). From this, the impact on fidelity for cascaded quantum gates can be estimated. Considering only the effect of microfabrication deviations on fidelity (neglecting all other factors), the relevant figure of merit is the induced phase deviation, not the absolute mode shift. With a measured phase deviation of $\delta\phi < 0.016\pi$: Single-gate fidelity: $F_1 = \cos^2(\delta\phi/2) = \cos^2(0.008\pi) = 99.94\%$.

N -cascaded gate fidelity: $F_N = F_1^N \approx [1 - (\delta\phi)^2/4]^N$.

For $N = 2, 3, 10$, and 100 , the fidelities are 99.88%, 99.82%, 99.40%, and 94.1%, respectively. For $N \leq 10$ cascaded operations, the fidelity exceeds the 99% threshold

typically required for fault-tolerant quantum computing. This confirms that our fabrication tolerance supports near-term quantum information processing (QIP) applications with realistic circuit depths.

Photonic crystal structures were fabricated in multiple batches using EBL and RIE. For a fixed designed structure, the fabrication process window is optimized through gradient exposure dose testing and multi-parameter structural scanning to compensate for dimensional deviations caused by process fluctuations. Through repeated validation across multiple fabrication runs, this approach consistently yields microstructures that meet the design specifications. Within the allowable process deviation range (<5 nm), the target structural parameters can be reliably achieved.

At the current stage, our research-level micro/nano fabrication process (EBL and dry etching) offers high dimensional control precision and repeatability, meeting the development requirements for multi-cascaded microcavity arrays in quantum information processing. This precision ensures mode matching between individual probe photons and each cavity stage in the cascade, thereby maintaining stable coherent transmission of single photons over extended propagation paths.

The characteristic dimensional deviation of <5 nm achieved by EBL and dry etching is comparable to, or even better than, typical CMOS foundry processes for photonic devices. Standard CMOS-compatible silicon photonics processes typically exhibit dimensional deviations of 5–10 nm (i.e., 2–3% relative deviation). Considering the fidelity variation caused by such process deviations as analyzed above, even a 10 nm process deviation would still yield acceptable fidelity (e.g., for $N = 10$, $F_{10} \approx 98.74\%$). Therefore, our devices are also feasible for future foundry fabrication.

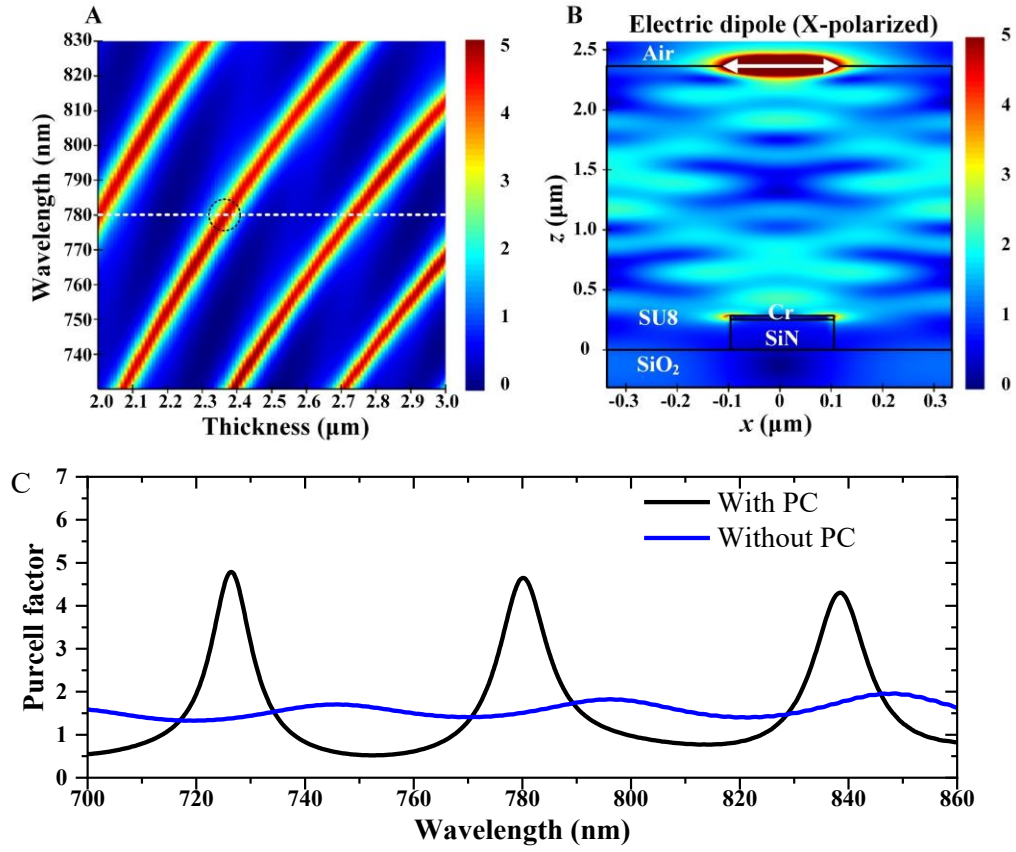


Fig. S1. Mode modulation of the PC-FP hybrid cavity.

(A) The wavelength-dependent Purcell factor of the system as the SU-8 thickness is scanned from 2.0 μm to 3.0 μm ; (B) Electric field distribution excited by an X-polarized electric dipole source; (C) Comparison of Purcell factors with and without the PC structure.

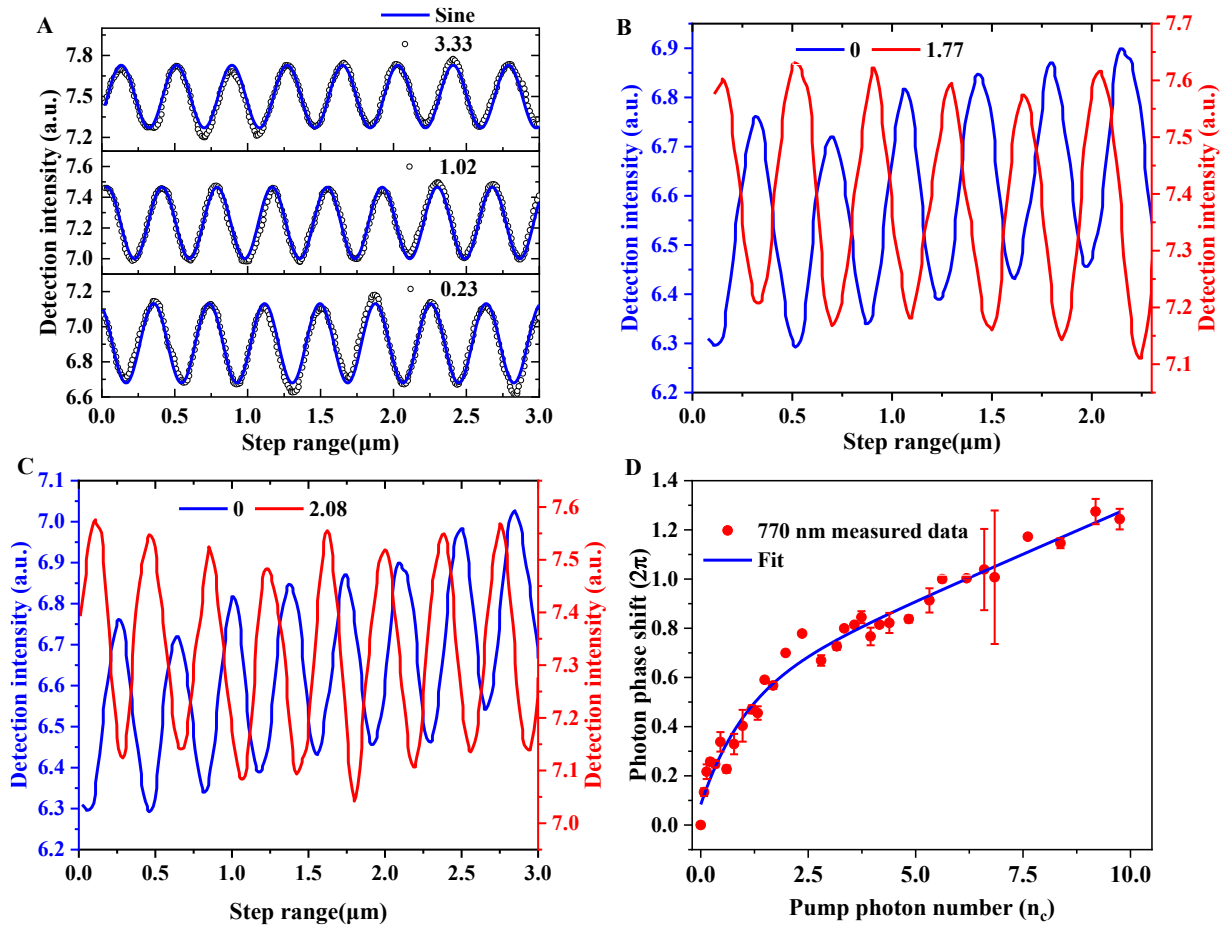


Fig. S2. Dependence of the phase shift on pump power at a probe-light wavelength of 770 nm.

(A) Original interference curves at average pump photon numbers of 0.23, 1.02, and 3.33; (B) Comparison of interference curves between pump photon numbers of 0 and 1.77; (C) Comparison of interference curves between pump photon numbers of 0 and 2.08; (D) Variation of the systematic phase with pump power. The fitting residuals (error bars) of phase shift are also displayed in the figure (the same applies to the following figures).

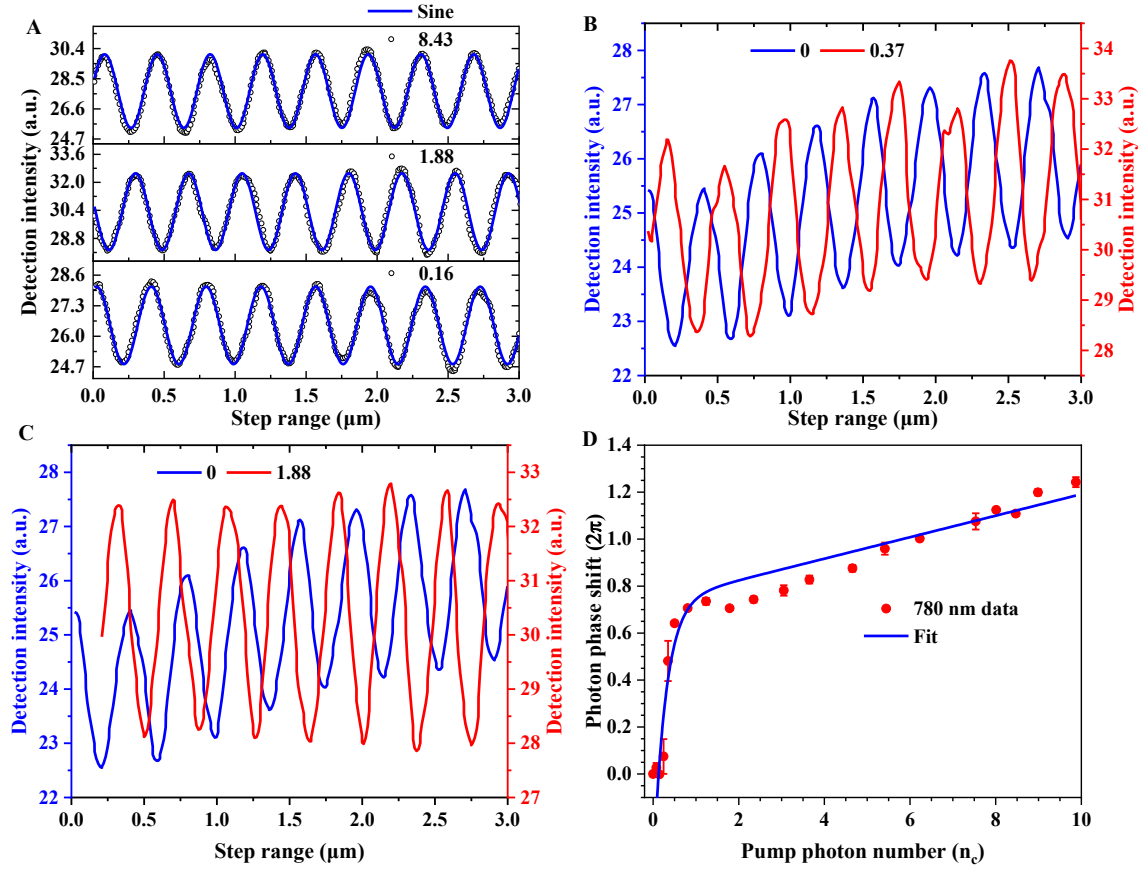


Fig. S3. Dependence of the phase shift on pump power at a probe-light wavelength of 780 nm.

(A) Original interference curves at pump photon numbers of 0.16, 1.88, and 8.43; (B) Comparison of interference curves between pump photon numbers of 0 and 0.37; (C) Comparison of interference curves between pump photon numbers of 0 and 1.88; (D) Variation of the systematic phase with pump photon number.

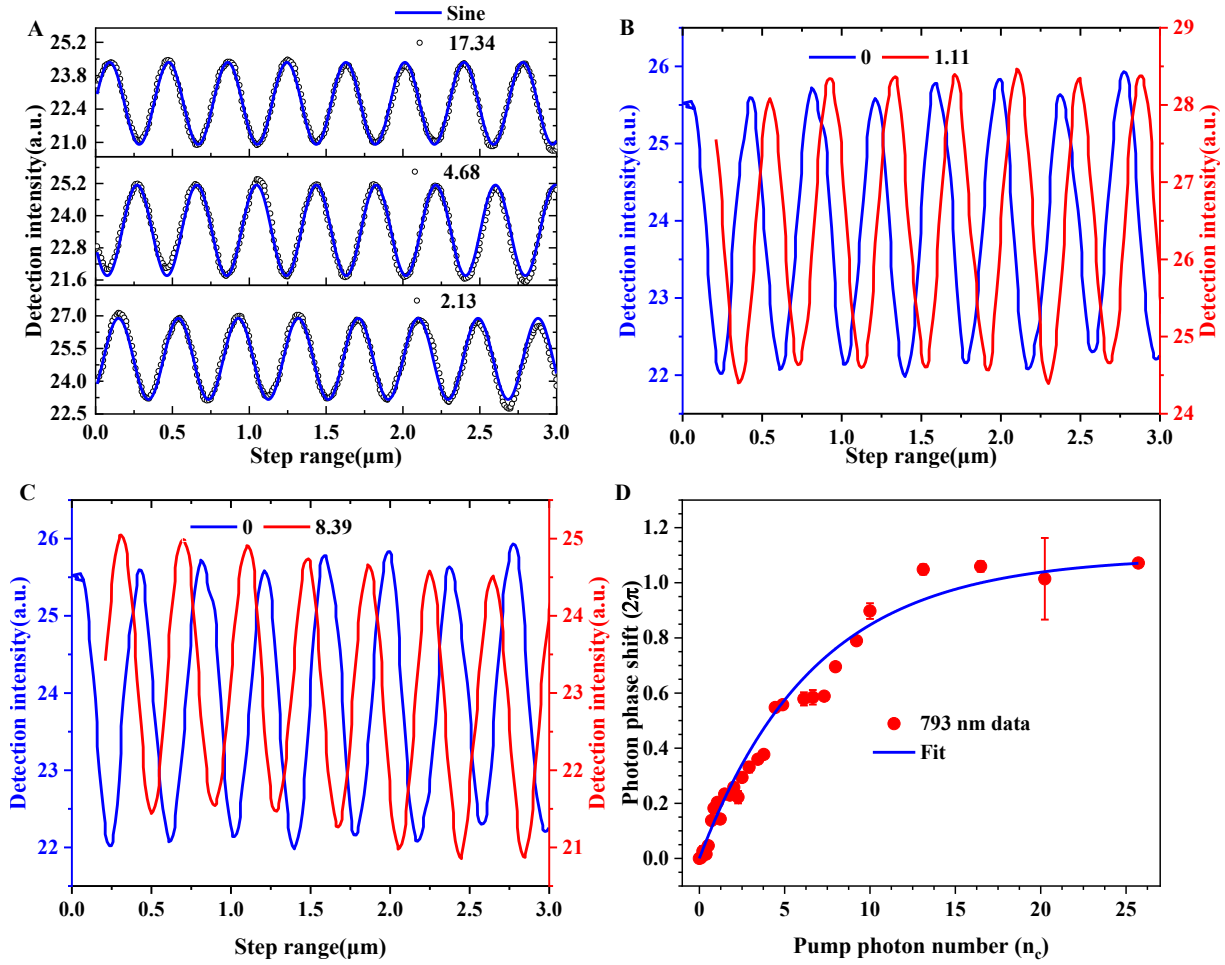


Fig. S4. Dependence of the phase shift on pump power at a probe-light wavelength of 793 nm. (A) Original interference curves at pump photon numbers of 2.13, 4.68, and 17.34; (B) Comparison of interference curves between pump photon numbers of 0 and 1.11; (C) Comparison of interference curves between pump photon numbers of 0 and 8.39; (D) Variation of the systematic phase with pump photon number.

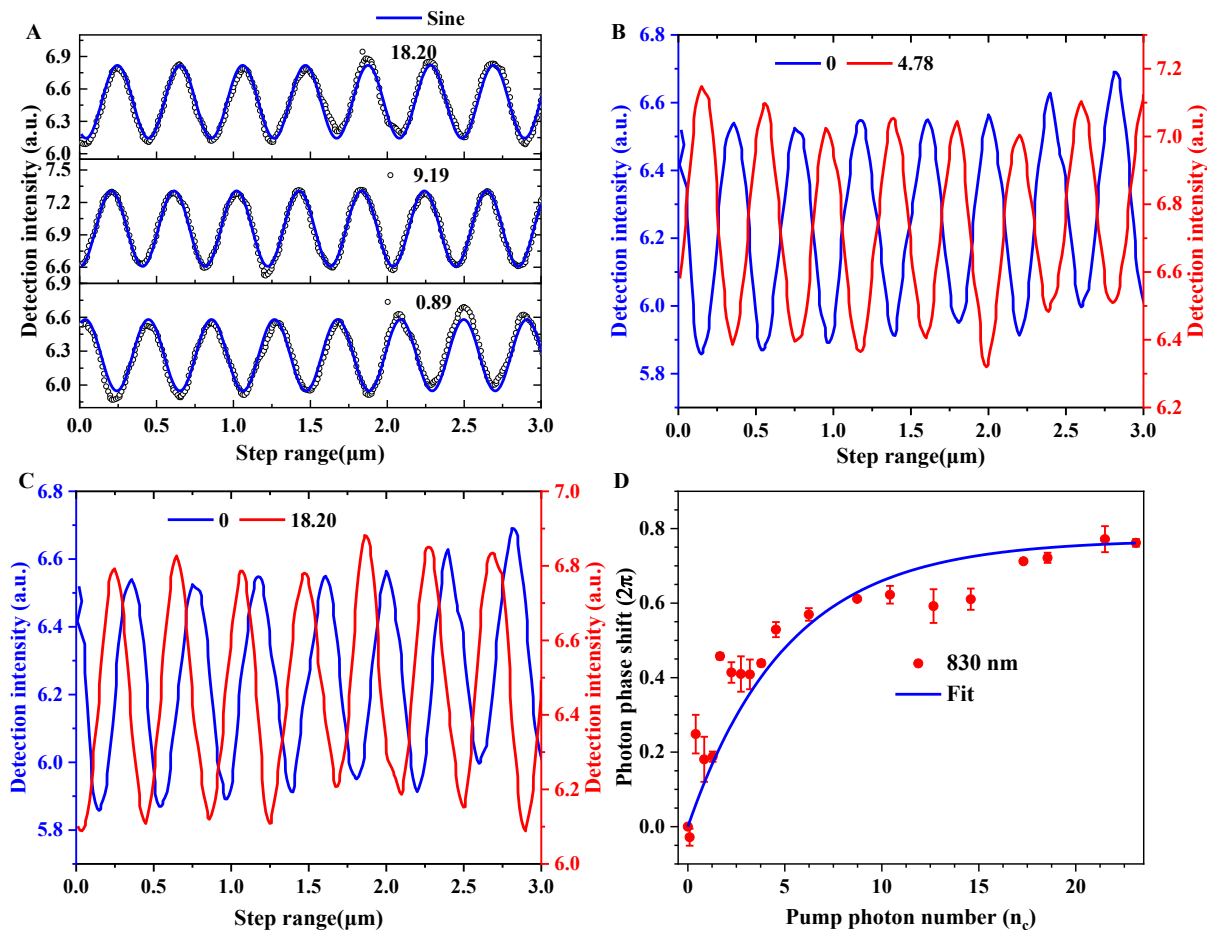


Fig. S5. Dependence of the phase shift on pump power at a probe-light wavelength of 830 nm.

(A) Original interference curves at pump photon numbers of 0.89, 9.19, and 18.20; (B) Comparison of interference curves between pump photon numbers of 0 and 4.78; (C) Comparison of interference curves between pump photon numbers of 0 and 18.20; (D) Variation of the systematic phase with pump photon number.

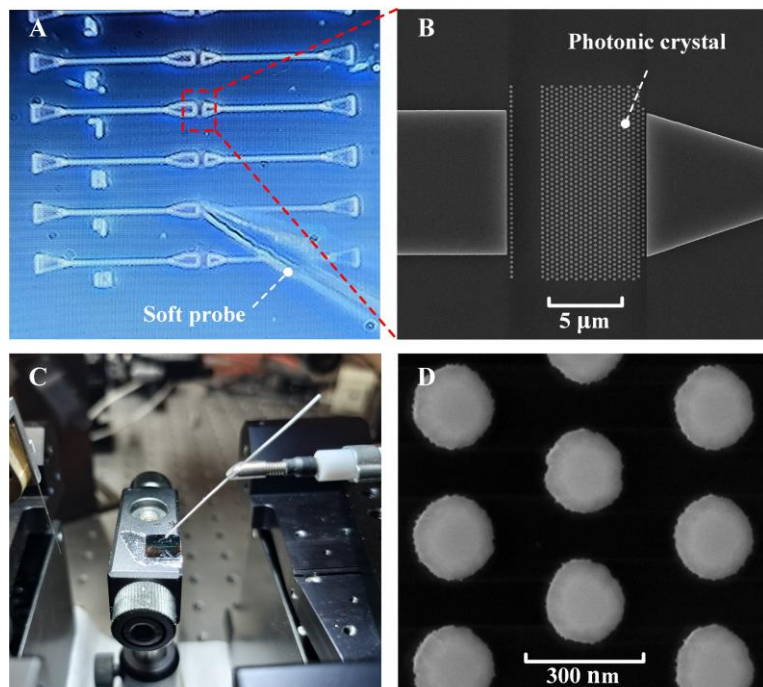


Fig. S6. Preparation of single CQDs via the soft probe method.

(A) Micrograph of a single CQDs introduced onto a PC structure using the soft-probe method; (B) The SEM image of the PC structure; (C) Schematic diagram of the experimental setup; (D) Magnified SEM image of the PC structure.

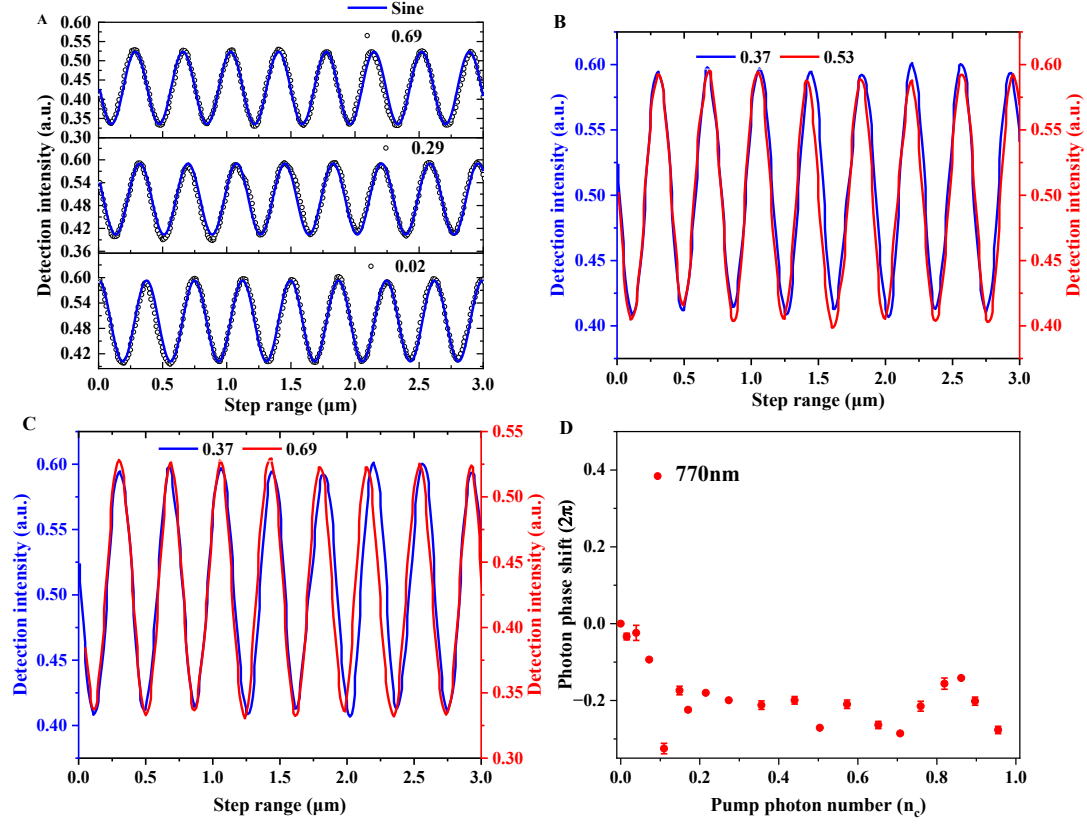


Fig. S7. Dependence of the phase shift on pump power at a probe-light wavelength of 770 nm. (A) Original interference curves at pump photon numbers of 0.02, 0.29, and 0.69; (B) Comparison of interference curves between pump photon numbers of 0.37 and 0.53; (C) Comparison of interference curves between pump photon numbers of 0.37 and 0.69; (D) Variation of the systematic phase with pump photon number.

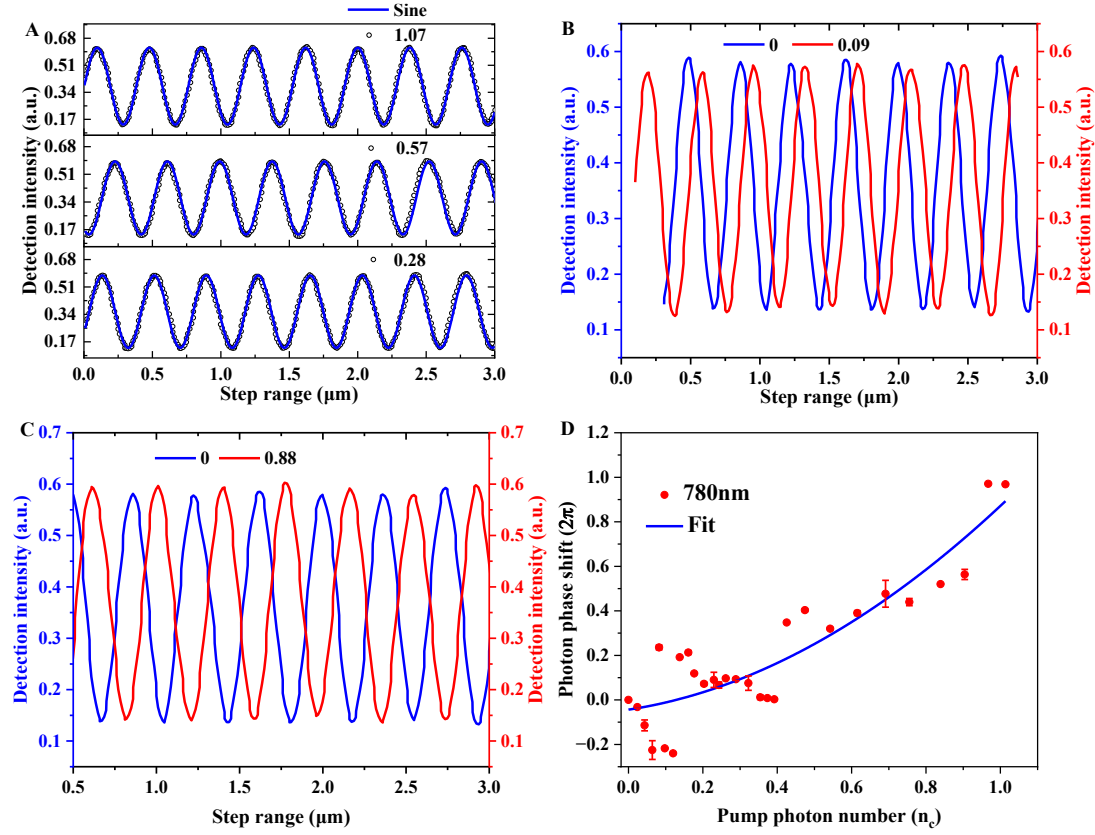


Fig. S8. Dependence of the phase shift on pump power at a probe-light wavelength of 780 nm.

(A) Original interference curves at pump photon numbers of 0.28, 0.57, and 1.07; (B) Comparison of interference curves between pump photon numbers of 0 and 0.09; (C) Comparison of interference curves between pump photon numbers of 0 and 0.88; (D) Variation of systematic phase with pump photon number.

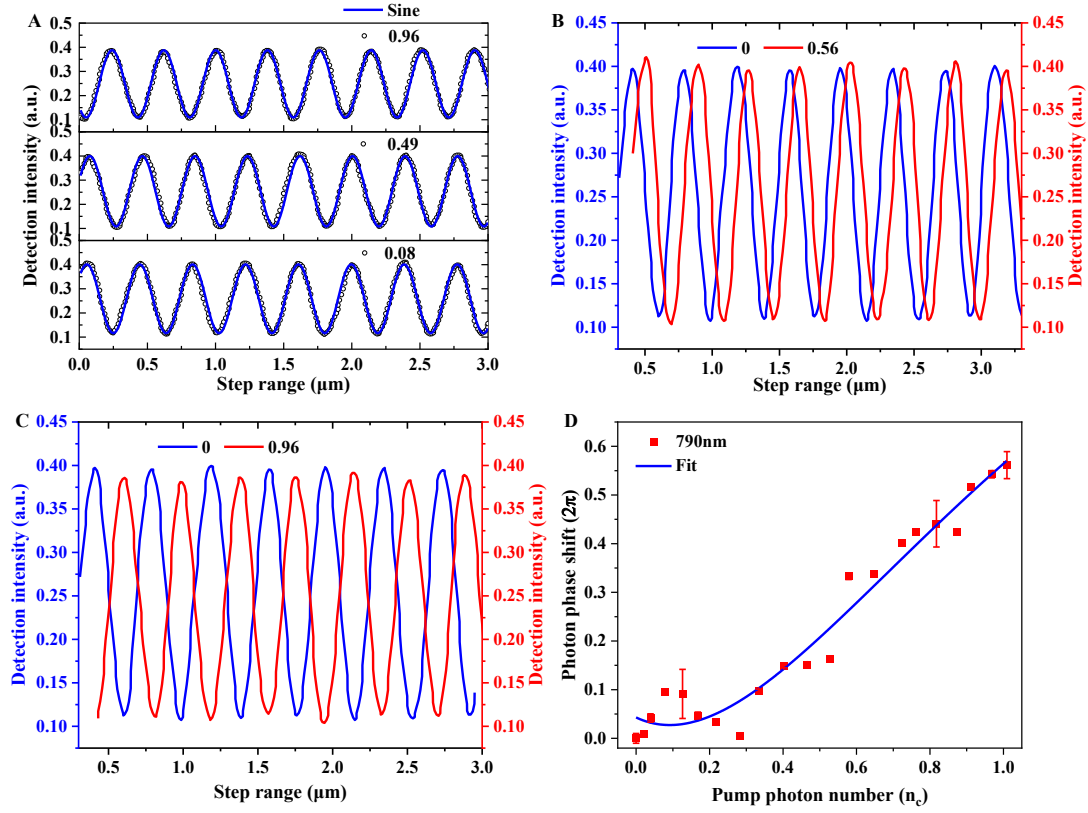


Fig. S9. Dependence of phase shift on pump power at a probe-light wavelength of 790 nm.

(A) Original interference curves at pump photon number of 0.08, 0.49, and 0.96; (B) Comparison of interference curves between pump photon numbers of 0 and 0.56; (C) Comparison of interference curves between pump photon numbers of 0 and 0.96; (D) Variation of systematic phase with pump photon number.

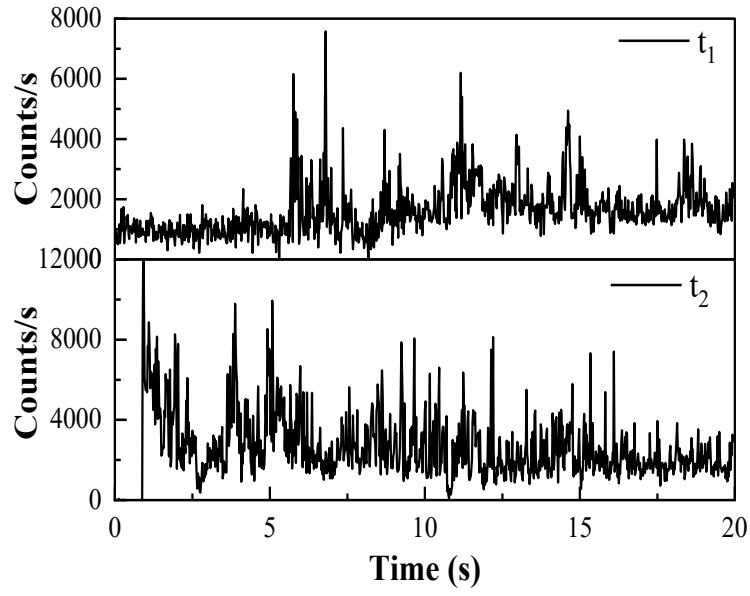


Fig. S10. Blinking traces of the same CQD used for phase measurement in Fig. 4 of the main text, recorded during two separate time intervals (t_1 , t_2).

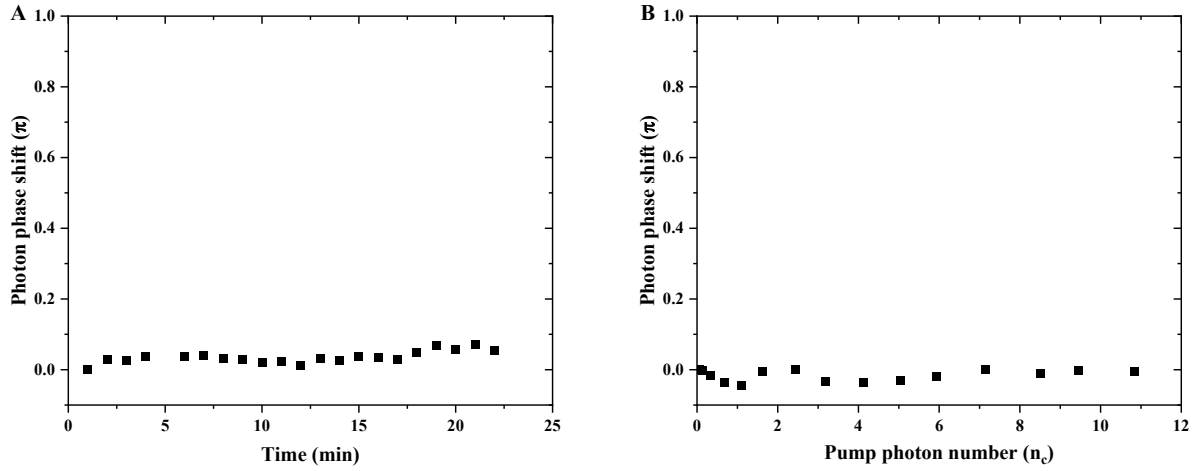


Fig. S11. Comparison measurements of phase shifts without pumping and without CQDs. (A) Measurements of the phase change in the absence of pumping. (B) Measurements of the controlled phase change in an empty cavity without CQDs.

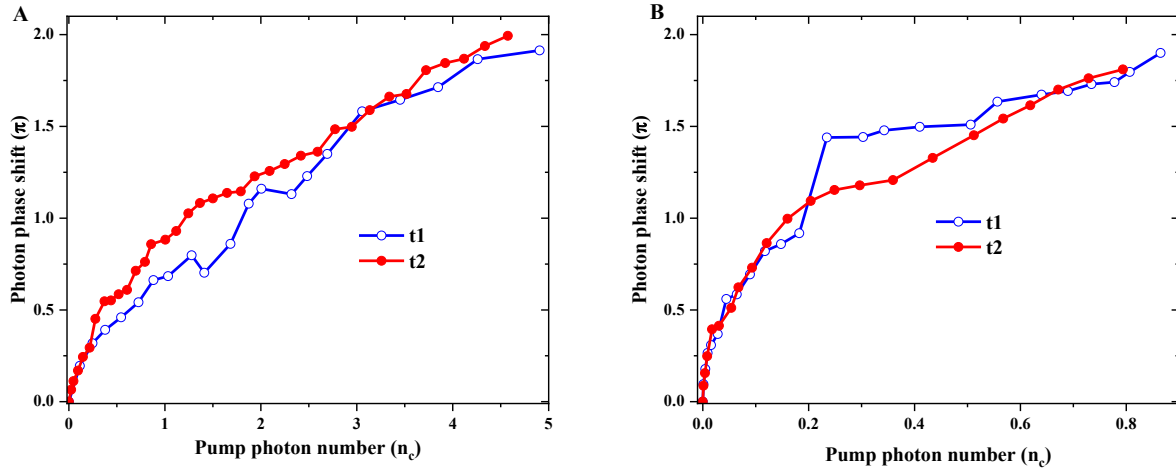


Fig. S12. Two repeated measurements of the controlled phase change in strongly coupled systems. (A) Two repeated measurements of the controlled phase change in a high-concentration CQDs coupled microcavity; (B) Two measurements of the controlled phase change with a single CQD coupled microcavity.

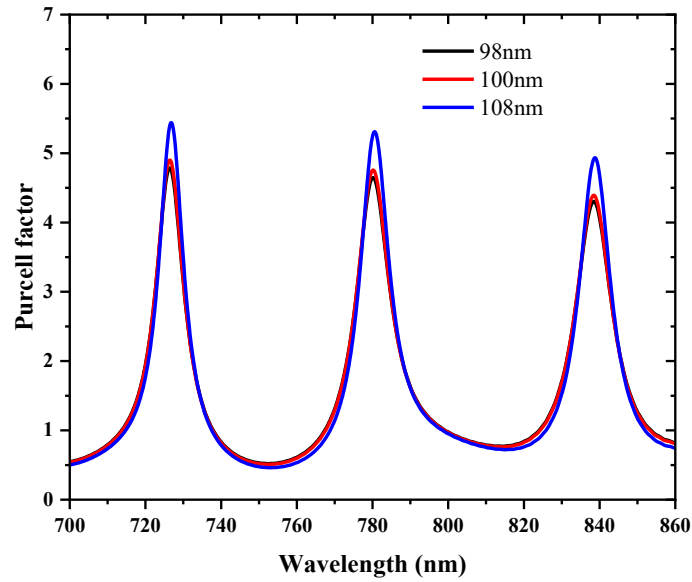


Fig. S13. Calculated hybrid cavity Purcell factor with different rods of photonic crystal. The symbol values of 98 nm, 100 nm, 108 nm represent the radius of fabricated rods.

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