All-optical coherent control of vacuum Rabi oscillations

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When an atom strongly couples to a cavity, the two systems can coherently exchange a single quantum excitation through the process of vacuum Rabi oscillation. Controlling this process enables precise synthesis of non-classical light, which plays a central role in quantum information and measurement. Although this control has been realized in microwave-frequency devices, it has been difficult to achieve at optical frequencies, which are essential for quantum communication and metrology. Here, we demonstrate coherent control of vacuum Rabi oscillation in an optical frequency device. We use a photonic molecule composed of two coupled nanocavities to simultaneously achieve strong coupling and a cavity-enhanced a.c. Stark shift. The Stark shift tunes a single quantum dot onto resonance with the photonic molecule on picosecond timescales, creating fast coherent transfer of energy between an atomic and photonic excitation. These results enable ultrafast control of light–matter quantum interactions in a nanophotonic device platform.

Strong interactions between single atoms and photons play a central role in the fields of quantum information and measurement. They are essential for transmitting quantum information over long distances in quantum networks1–3, and can be harnessed to generate non-classical states of light4–7, which are highly desirable for quantum metrology applications6. Optical cavities can create strong atom–photon interactions by confining light to a small mode volume. The strong coupling regime represents the extreme limit of these interactions, where a single quantum of energy coherently oscillates between an atomic and photonic excitation through the process of vacuum Rabi oscillation.

Previous work investigating single atomic systems strongly coupled to optical cavities usually operated in the static limit where the coupling between the two systems remained constant. Strong coupling was detected either in the form of vacuum Rabi splitting7,8, or by direct time-domain observation of vacuum Rabi oscillations11. Recently, several works have reported significant progress in controlling the dynamical response of atomic systems strongly coupled to cavities for applications such as optical switching12–15, reversible storage of photonic qubits16,17 and hybrid quantum information processing18,19. However, these experiments all operated in the adiabatic limit where control was applied on timescales that are slow compared to the vacuum Rabi frequency.

When the interaction between an atom and cavity changes on a timescale that is fast compared to the vacuum Rabi frequency, the system enters the regime of diabatic rapid passage. This regime enables deterministic transfer of a single quantum of energy between an atomic excitation and a cavity photon by coherent manipulation of vacuum Rabi oscillation dynamics. Circuit quantum electrodynamics (QED) devices operating at microwave frequencies have effectively implemented this coherent manipulation to demonstrate remarkable capabilities such as Fock-state generation20 and the synthesis of arbitrary photonic wavefunctions21. At optical frequencies, however, diabatic control of vacuum Rabi oscillations between a single atomic system and a cavity remains challenging due to a lack of good methods to rapidly tune the coupling between the two systems.

In this Article we report a method to coherently control vacuum Rabi oscillations diabatically in an optical frequency device. We achieve this capability by utilizing a single quantum dot embedded in a photonic molecule composed of two coupled optical nanocavities22–26. The photonic molecule supports a pair of optical modes, one that strongly couples to the quantum dot and a second that creates a cavity-enhanced a.c. Stark shift27. We use the a.c. Stark shift to rapidly tune the quantum dot onto resonance with the strongly coupled mode on picosecond timescales and control vacuum Rabi oscillation dynamics. We demonstrate transfer of excitation between the cavity mode and quantum dot, and show that this process is coherent. Furthermore, we implement coherent control of light–matter states, an important building block for synthesizing photonic wavefunctions. We attain these capabilities using an integrated semiconductor nanophotonic device structure that could enable the controlled generation of quantum states of light at gigahertz rates.

Coherent control of Rabi oscillations

To understand how a Stark shift enables coherent control of vacuum Rabi oscillations we consider a simplified model of the system described by the level structure shown in Fig. 1a. We restrict our attention to the first excitation manifold, a valid approximation when the system is weakly excited. The quantum dot is treated as a two-level atomic system and we adopt the notation $|G,n\rangle$ and $|E,n\rangle$ to denote quantum states where the atom is in its ground and excited state, respectively, and the cavity contains $n$ photons. In the figure, $\Delta$ is the detuning between the atom and cavity, and $g$ is the cavity–quantum dot coupling strength. An off-resonant optical pulse, expressed as a classical Rabi frequency $\Omega(t)$, excites the quantum dot with detuning $\Delta$. The optical pulse induces an a.c. Stark shift that controls the detuning between the dot and cavity. We assume $\Delta \gg \Omega(t)$ for all time so that the off-resonant pulse only interacts with the quantum dot through a virtual transition. In this limit, the...
atom–cavity system evolves according to the following effective Hamiltonian (Supplementary Section I):

$$
H_{\text{eff}} = \hbar \Delta a \sigma_+ \sigma_- + \hbar g \left( a^\dagger \sigma_+ + a \sigma_- \right)
$$

where $a$ is the bosonic annihilation operator for the cavity mode, $\sigma_+$ and $\sigma_-$ are the quantum dot dipole raising and lowering operators, and $\Delta = \Delta_c - (2|G(t)|^2/\Delta)$ is the cavity–quantum dot detuning in the presence of an a.c. Stark shift.

For the Hamiltonian described in equation (1), states $|G,0\rangle$ and $|G,1\rangle$ comprise a two-level system that can be coherently controlled by the Stark field $\Omega(t)$ by rapidly modulating their relative detuning. The effect of the Stark field is particularly simple to derive in the ideal limit of diabatic rapid passage, where the field turns on and off instantaneously. For the special case where only the quantum dot is excited and the Stark shift tunes it onto cavity resonance, the probability that the system occupies state $|G,1\rangle$ after the Stark pulse is $P(G,1) = \sin^2(\theta)$, where $\theta$ is the Stark pulse duration (Supplementary Section II). Thus, the quantum dot excitation coherently transfers to the cavity, with perfect transfer occurring at the condition $2\pi \sigma = \pi + m2\pi$, where $m$ is an integer. We obtain the same results for the quantum-dot excitation when the cavity is initially excited, indicating that this process can transfer excitation to either system.

**A photonic molecule strongly coupled to a quantum dot**

Achieving a large a.c. Stark shift using the excitation scheme illustrated in Fig. 1a is challenging, because the field $\Omega(t)$ drives the cavity mode off-resonance. This means that the majority of the field reflects and only a small amount of power drives the quantum dot. Recently, we have demonstrated that a photonic molecule can solve this problem\(^2\). A photonic molecule is composed of two cavities coupled by a fast photon tunnelling interaction\(^{12,24}\). These photonic structures exhibit two non-degenerate modes, one that strongly couples to the quantum dot and a second that can induce a cavity-enhanced a.c. Stark effect\(^2\).

We utilized a photonic crystal implementation of a photonic molecule to achieve controlled transfer. Figure 1b presents a scanning electron microscopy (SEM) image of the fabricated device, which is composed of two coupled photonic-crystal cavities, as well as the calculated modes (see Methods). The photonic molecule exhibits two coupled modes composed of symmetric

**Figure 1 | Device design and experimental set-up.** a, Schematic of quantum level structure of a cavity–quantum dot system in the presence of a strong off-resonant pulse denoted by the classical Rabi frequency $\Omega(t)$. The pulse induces an a.c. Stark shift that optically tunes the quantum dot on-resonance with the cavity. b, Scanning electron microscopy image of the fabricated photonic-crystal molecule composed of two evanescently coupled photonic-crystal cavities (scale bar, 1 $\mu$m), together with finite-difference time-domain calculations showing the $E_x$ component of the anti-symmetric (bottom) and symmetric (top) modes of the device. c, Experimental set-up for all performed measurements. BS, beamsplitter; PBS, polarizing beamsplitter; OL, objective lens; HWP, half-wave plate; SMF, single-mode fibre; SPCM, single-photon-counting modules.

**Figure 2 | Characterization of photonic molecule modes.** a, Reflection spectrum of the photonic molecule, recorded at 45 K, showing the two coupled cavity modes (M1 and M2) and the quantum dot (QD). b, Reflection spectrum around mode M1 as a function of temperature. The quantum dot tunes across the cavity mode, exhibiting an anti-crossing.
and anti-symmetric combinations of the individual cavity modes (top and bottom, respectively, in Fig. 1b).

We characterized the mode structure of the fabricated device using a continuous-wave broadband light-emitting diode (LED) that acted as a white light source. Figure 1c shows the complete experimental set-up used for all reported measurements, which is described in further detail in the Methods. Figure 2a presents the reflection spectrum of the device, taken at 45 K. The spectrum shows two peaks corresponding to the coupled cavity modes, denoted M1 (at \( \omega_c = 925.84 \) nm) and M2 (at \( \omega_c = 927.48 \) nm), which are spectrally separated by 565 GHz. The spectrum shows an additional peak corresponding to a quantum dot resonance that is red-detuned from mode M1 (labelled QD in the figure). We note that another quantum dot can be seen to couple to mode M2, but is far detuned from mode M1 and plays no role in our measurements. From the spectrum we calculated the quality factor \( Q \) of modes M1 and M2 to be 18,500 and 12,000, respectively, corresponding to energy decay rates of \( \kappa_1/2\pi = 17.7 \) GHz and \( \kappa_2/2\pi = 28.3 \) GHz.

Figure 2b presents the cavity reflection spectrum near the resonance of mode M1 as a function of temperature. The quantum-dot resonance redshifts with increasing temperature and exhibits an anti-crossing when tuned across the cavity resonance. We calculated a coupling strength of \( g/2\pi = 8.1 \) GHz by fitting the reflection spectrum when the quantum dot was resonant with the cavity to the theoretically predicted spectrum (Supplementary Section III). The coupling strength satisfies the condition \( 4g > \kappa_1 \), indicating that we are operating in the strong coupling regime. We note that the vacuum Rabi period, given by

\[
\tau_r = \frac{\pi}{\sqrt{\pi^2 - (\kappa/4)^2}} = 74 \text{ ps}
\]

in the presence of cavity losses, is much longer than the cavity lifetime of mode M2 given by \( \tau_c = 1/\kappa_2 = 6 \) ps. Thus, we can dynamically excite mode M2 to induce a Stark shift on timescales that are much shorter than \( \tau_c \).

Transfer of excitation between a quantum dot and cavity

To demonstrate that we can coherently control the strongly coupled system on fast timescales, we utilized picosecond optical pulses both for excitation and to induce a Stark shift (see Methods). We first investigated energy transfer from the quantum dot to the cavity. We set the detuning between the two systems to \( \Delta \omega = 18 \) GHz, which is sufficiently large to ensure that the stationary states of the system are well-approximated by the bare quantum dot and cavity modes. An excitation pulse drives the quantum dot resonance and a subsequent Stark pulse drives the resonance of mode M2. The Stark pulse induces a pulsed a.c. Stark shift that transfers energy to the cavity on timescales that are short compared to the decay rate of the quantum dot exciton. We then allowed the system to radiatively decay and measured the total emission spectrum to determine the fraction of light emitted at the cavity frequency.

Figure 3a presents the measured cavity emission spectrum as a function of \( \Delta \tau \), the time delay between excitation and the Stark pulse. For \( \Delta \tau < 0 \) the cavity emits a constant background that is independent of the Stark pulse delay. This background is due to partial spectral overlap between the excitation pulse and cavity resonance, as well as non-resonant energy transfer of the quantum-dot excitation. However, when \( \Delta \tau > 0 \) we observe enhanced emission at...
Coherent Rabi oscillations

We next investigated the energy transfer as a function of Stark laser power. We excited the quantum-dot resonance and fixed $\Delta \tau$ to the peak transfer point observed in Fig. 3b. Figure 4a plots the measured spectrum as a function of average Stark field power (determined after the focusing objective). The spectrum shows out-of-phase oscillations between the quantum dot and cavity emission. These oscillations become more clear when we plot the intensity at the resonant frequency of the cavity ($\Delta \lambda = 0$) and quantum dot ($\Delta \lambda = 0.07 \text{ nm}$) as a function of Stark field power, as shown in Fig. 4b. The model described by the Hamiltonian in equation (1) provides insight regarding the origin of these oscillations. From this model, we can show that $P_{\text{QD}, \text{C}} \propto \sin^2(\omega_\tau/2)$, where $\omega_\tau = \sqrt{(2\Omega^2/\Delta - \Delta_c)^2 + 4\gamma^2}$ is the vacuum Rabi frequency of the Stark-shifted quantum dot and $\Omega$ is the amplitude of the Stark pulse, expressed as a classical Rabi frequency. By increasing the Stark pulse amplitude we increase $\omega_\tau$, thereby modulating the number of oscillations the system undergoes, which demonstrates the coherence of the transfer process.

Figure 4c,d shows the numerical solution to the master equation, which exhibits good agreement with the measured results. We plot these figures as a function of $\Omega_{\text{C}}^2$, where $\Omega_\text{C}$ is the peak amplitude of the time-varying Stark pulse (assumed to be a Gaussian pulse), again expressed as a classical Rabi frequency. Optimal transfer occurs at $\Omega_{\text{C}}/2\pi = 107 \text{ GHz}$, which corresponds to a peak Stark shift of 40 GHz. We note that there is a deviation in anti-correlation between the dot and cavity intensity at higher Stark powers, which is due to non-adiabatic corrections (Supplementary Section V).
Coherent control of light–matter states

To further demonstrate coherent control of the strongly coupled system we performed a Ramsey-type experiment. In contrast to the previous results where the quantum dot and cavity were detuned, we performed the Ramsey measurement when the two systems were on-resonance. In this regime, the two stationary states of the system are the polariton modes $|P_{\pm}\rangle = (|G,1\rangle \pm |E,0\rangle)/\sqrt{2}$. We chose this operating condition because both polaritons have equal decay rates, which leads to the best Ramsey interference contrast. We excited the polariton states simultaneously with a short 2 ps optical pulse, then let the system freely evolve for a fixed amount of time $\Delta \tau$ so that the polaritons dephased by $\phi = 2\pi (\Delta \tau/\tau_c)$. A Stark pulse subsequently mixed the two polariton states, creating a Ramsey interference effect. Using the Hamiltonian in equation (1), we show that the final occupation probabilities for the polaritons after the Stark pulse are given by $P_{\pm} = (1/2)(1 \mp \sin \phi \sin \omega_c \tau)$, where $\tau$ is once again the Stark pulse duration (Supplementary Section VI). The oscillatory dependence on $\phi$ is the signature of Ramsey interference.

Figure 5a shows the measured spectrum as a function of $\Delta \tau$. The two resonances observed in the spectrum for $\Delta \tau < 0$ correspond to the dressed-state polaritons. For $\Delta \tau > 0$, the Stark pulse induces a clear energy transfer. Figure 5b plots the emission intensity at the centre wavelength of the lower and upper polaritons. The out-of-phase oscillation between the two polariton intensities is the signature of Ramsey interference. These oscillations fall off after one period due to decay of the polariton states. Figure 5c,d shows the calculated results using master equation formalism, which again exhibit good agreement with the experimental results.

Discussion

In summary, we have demonstrated picosecond optical coherent control of vacuum Rabi oscillation. We utilized a photonic molecule to simultaneously achieve strong coupling and a cavity-enhanced a.c. Stark shift that enabled us to modulate the cavity–quantum dot detuning on picosecond timescales. By rapidly Stark-shifting the quantum dot onto cavity resonance, we demonstrated controlled transfer of excitation between the two systems. Oscillations in the transfer efficiency as a function of the vacuum Rabi frequency established the coherence of the process. We also demonstrated coherent control of light–matter states through a Ramsey-type measurement. This method enables fast probing and control of light–matter interactions in an integrated nanophotonic device structure.

Further improvements in transfer efficiency can be achieved by increasing the cavity–quantum dot coupling strength and reducing cavity losses using deterministic alignment and better cavity designs. Current state-of-the-art quantum dot cavity QED systems have already achieved $g/k > 3$ (ref. 37). Such coupling strengths may be sufficient to control higher-order photon number
states when combined with the control technique we demonstrate here. Our results could ultimately provide a path towards gigahertz-rate controlled synthesis of non-classical light at optical frequencies.

Methods

Experimental set-up. The sample was mounted in a low-temperature, closed-cycle refrigerator with variable temperature down to 3.5 K, and excited using either a continuous-wave broadband LED or pulsed Ti:sapphire lasers. For dynamic measurements, the two pulsed lasers were synchronized in time using a lock-to-clock system to generate the excitation and Stark shift pulses. The delay between the two laser pulses was varied by changing the delay in the phase-locked loop of the lock-to-clock circuit. The reflected signal from the cavity–quantum dot system was collected using polarization-selective spectroscopy with a half-wave plate (HWP) and a polarization beamsplitter (PBS). Light was injected with vertical polarization, which was rotated 45° relative to the cavity-mode polarization axis. Light that coupled to the cavity experienced a polarization rotation and therefore partially transmitted through the PBS. The transmitted signal was sent either to a spectrometer for spectral characterization, or to a Hanbury–Brown–Twiss interferometer for second-order correlation measurements using two single-photon-counting modules (SPCMs) and a 50/50 beamsplitter.

Device design and fabrication. The modes of the photonic molecule were calculated using a numerical finite-difference time-domain method using commercial software (Lumerical). The device design consisted of a two-dimensional array of air-holes in a triangular lattice with a radius of 70 nm and period of 240 nm. Cavities were formed by removing three holes and shifting adjacent holes to optimize the quality factor. Device fabrication was performed on an initial wafer consisting of a 160 nm GaAs membrane on top of a 1-μm-thick AlGaAs sacrificial layer. The GaAs membrane contained a single layer of InAs quantum dots at the centre (with quantum dot density of 30 μm⁻²) embedded in GaAs photonic-crystal structures. Photonic crystals were fabricated using electron-beam lithography, followed by chlorine-based inductively coupled plasma etching and a chemical wet etch to remove the sacrificial layer.

Continuous-wave reflectivity measurement. The fabricated sample was mounted in a liquid-helium cryostat and optically excited from the out-of-plane direction using a broadband laser diode with emission between 900 and 1,000 nm. One of the two cavities in the molecule was predominantly excited, and the emission from the same cavity was isolated using a spatial filter. Because of the strong hybridization of the two modes, the exact choice of cavity to excite does not matter, as each mode is a nearly pure superposition of the individual cavity modes. The signal reflected from the cavity was isolated using cross-polarization reflection spectroscopy and sent to a spectrometer with a resolution of 0.02 nm. For the anti-crossing spectrum in Fig. 2b, the temperature of the photonic crystal device was precisely tuned between 40 and 45 K.

Picosecond optical pulse excitation. Two time-synchronized Ti:sapphire lasers were used as the excitation field and the Stark shift field. A phase-locked loop in the synchronization circuit controlled the delay between the two pulses. For the data shown in Figs 3a,b and 4, the excitation pulse was obtained by filtering 8 ps pulses from a Ti:sapphire laser using a fibre Fabry–Perot filter to increase the pulse duration to ~15 ps. The filter bandwidth of the Fabry–Perot filter was varied as it was tuned. Thus, the exact pulse duration varied from 10 to 22 ps depending on the specific setting of the filter. The average excitation power was 20 nW. The Stark pulse was filtered to a 22 ps pulse duration using a free-space grating spectrometer (pulse duration measured using an autocorrelator), with an average excitation power of 120 nW. For the data in Fig. 3c,d, the excitation pulse was generated by filtering an 8 ps pulse using a fibre Fabry–Perot filter and set to an average power of 40 nW. The Stark pulse was an 8 ps pulse directly generated by the laser with an average power of 150 nW. The results shown in Fig. 5 were obtained using a 2 ps excitation pulse with an average power of 40 nW and an 8 ps Stark pulse with average power of 150 nW, both obtained directly from the laser without filtering. For all experiments, the delay between the excitation and Stark pulses was calibrated using a high-time-resolution avalanche photodiode with temporal resolution of ~30 ps to determine the zero delay point.

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Author contributions
E.W. conceived and designed the experiment. R.B. and T.C. designed and fabricated the device, conducted experiments and carried out analysis. K.R.C., E.W., T.C. and R.B. performed theoretical simulations. R.B. and E.W. wrote the manuscript, with input from all authors. G.S.S. grew the quantum-dot wafer. E.W. supervised the work.

Additional information
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Competing financial interests
The authors declare no competing financial interests.