Coherently driving a single quantum two-level system with dichromatic laser pulses

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The excitation of individual two-level quantum systems using an electromagnetic field is an elementary tool of quantum optics, with widespread applications across quantum technologies. The efficient excitation of a single two-level system usually requires the driving field to be at the same frequency as the transition between the two quantum levels. However, in solid-state implementations, the scattered laser light can dominate over the single photons emitted by the two-level system, imposing a challenge for single-photon sources. Here, we propose a background-free method for the coherent excitation and control of a two-level quantum system using a phase-locked dichromatic electromagnetic field with no spectral overlap with the optical transition. We demonstrate this method experimentally by stimulating single-photon emission from a single quantum dot embedded in a micropillar, reaching single-photon purity of 0.988(1) and indistinguishability of 0.962(6). The phase-coherent nature of our two-colour excitation scheme is demonstrated by the dependence of the resonance fluorescence intensity on the relative phase between the two pulses. Our two-colour excitation method represents an additional and useful tool for the study of atom–photon interaction, and the generation of spectrally isolated indistinguishable single photons.

The coherent control of a single two-level quantum system using an electromagnetic field, by tuning the frequency, amplitude and envelope of the field, is the elemental tool of quantum optics: resonant atom–photon interaction underpins interesting phenomena such as Rabi oscillation\textsuperscript{1}, Ramsey interference\textsuperscript{2}, Autler–Townes splitting\textsuperscript{3} and the Mollow triplet\textsuperscript{4}. Meanwhile, it has also become ubiquitous in quantum information technologies\textsuperscript{5}, used extensively for the initialization, manipulation and measurement of quantum bits in various physical systems including trapped ions\textsuperscript{6}, quantum dots\textsuperscript{7–9,14–20} from quantum dots, which have shown near-unity single-photon purity and indistinguishability, and high extraction efficiency\textsuperscript{21–23}. As the pumping laser spectrally overlaps with the single photons, polarization filtering was usually used to suppress the laser background; however, this reduced the system efficiency of the single-photon source\textsuperscript{21–23} by at least 50%. The quests for large-scale boson sampling\textsuperscript{24} and optical quantum computing\textsuperscript{25} require near-unity single-photon efficiency, and thus novel methods for optical control of single quantum emitters. One possibility is to use coherent two-photon excitation\textsuperscript{26} to drive the cascaded biexciton–exciton transition in a quantum dot, where a record-high single-photon purity has been reported\textsuperscript{27,28}. However, in such a three-level system, the emitted single photons do not have single polarization. Moreover, the intrinsic time correlation between the biexciton and exciton radiative decay can reduce the photon indistinguishability\textsuperscript{29}. Despite recent progress in such a system, the generation of single-photon sources with simultaneous combination of single polarization, near-unity indistinguishability and efficiency was proven difficult.

Theory on two-colour excitation

Here, we propose and demonstrate a new way to coherently drive a quantum two-level system using a dichromatic field that has no spectral overlap with the transition frequency. As schematically shown in Fig. 1a, the dichromatic pulse consists of two sidebands, which have the same pulse envelope $e(t)$ and a fixed phase difference $\delta \varphi$ and are symmetrically red-detuned and blue-detuned by $\Delta \omega$ from the atomic transition frequency $\omega_{eg}$. A phase-coherent combination of the two sidebands, as shown by the mathematical relation

$$e(t) = \frac{e(t)}{2} \cos((\omega_{eg} + \Delta \omega)t + \delta \varphi) + \frac{e(t)}{2} \cos((\omega_{eg} - \Delta \omega)t)$$

$$= e(t) \cos\left( \frac{\Delta \omega t + \delta \varphi}{2} \right) \cos\left( \frac{\omega_{eg} t + \delta \varphi}{2} \right)$$

effectively cancels the detuning, and results in a resonant pulse with a new envelope $E(t) = e(t) \cos(\Delta \omega t + \delta \varphi/2)$, as shown in Fig. 1b. The dichromatic pulse yields an effective Rabi frequency proportional to $E(t)$. The time-integrated pump pulse area can be suitably engineered to $\pi$, which will deterministically invert the population. As the detuning can be set much larger than the linewidth of the...
resonance fluorescence, the single photons can be spectrally isolated from the well-separated laser background with a bandpass filter.

Assuming a Gaussian-shaped envelope, the time-resolved Rabi frequency is plotted in Fig. 1c for different ratios of the detuning ($\Delta \omega$) to the full-width at half-maximum of the sideband ($\text{FWHM}$), where a more rapid oscillation is observed for increasing $\Delta \omega / \text{FWHM}$ ratios. Owing to the parity-alternating oscillations, the laser power to reach a $\pi$-pulse is expected to increase for larger detunings. In addition, the relative phase between the red and blue sidebands gives rise to a new quantum interference phenomenon and a controlling knob for the two-level system beyond the conventional one-pulse resonant excitation. As illustrated in Fig. 1d, the phase $\delta \varphi$ can control the relative position of the positive and the negative parts of the Rabi frequency within the envelope, thereby modulating the time-integrated pulse area.

The coherent dynamic process could be conceptually visualized as the motion of the well-known Bloch vector. While the red-detuned or blue-detuned sideband alone cannot efficiently drive the two-level system along the prime meridian of the Bloch sphere (Fig. 1e), a phase-coherent combination of the two sidebands together can achieve a deterministic population inversion through an oscillating trajectory as shown in Fig. 1f.

Experimental arrangement

Our scheme is in principle applicable in various scenarios involving a single two-level quantum system interacting with electromagnetic fields. Here, we report an experimental demonstration. The two-level system is a single InGaAs quantum dot embedded inside a 2.5-µm-diameter micropillar, cooled to 3.6 K. The micropillar cavity features a relatively low quality factor of ~1,000 such that its cavity bandwidth can accommodate the spectral width of the dichromatic pulse. A confocal microscope is used for laser excitation of the quantum dot and collection of the emitted resonance fluorescence.

First, we prepare a phase-locked dichromatic pulse using a 4f optical system that consists of two identical lenses of focal length $f = 20$ cm, as shown in Fig. 2a. Picosecond pulses with a width of 3 ps from a Ti:sapphire laser are diffracted by the first grating; that is, the different frequency components are distributed to different spatial directions. A block placed in the Fourier plane intercepts the laser scattering by an extremely high extinction ratio of $10^{11}$ (Supplementary Fig. 1). The transmission efficiency of the second grating is measured to be 26.4%; in future, this system could be replaced by commercially available ultranarrowband optical filters with a bandwidth as narrow as 0.1 nm and a near-unity transmission rate.

Population inversion

Next, we send the dichromatic pulse to excite a single-electron-charged quantum dot with the centre of the dichromatic pulse being tuned to be resonant with the atomic transition, and test its performance in population inversion. Figure 3a plots the pulsed resonance fluorescence single-photon intensity as a function of the driving optical field strength. For a comparison, the data from the conventional resonant excitation using a 3 ps laser pulse and cross-polarization extinction are also presented in Fig. 3a. The dichromatic excitation reaches a full population inversion at a pump power of 115 nW, 8.5 times larger than the monochromatic excitation. The single-photon count under the dichromatic excitation, after correcting for the transmission loss of the second 4f system, is 1.74 times higher than that using the monochromatic excitation and cross-polarization. Such an enhancement is due to the removal of the polarization filtering that usually sacrifices ~50% of the single photons. Ideally, the efficiency enhancement should be a factor of 2.

![Fig. 1](image-url)
The additional loss might be due to cutting out of the phonon side-band\(^3\) by the 4\(f\) system and asymmetry of the two sidebands of the dichromatic pumping pulse.

An important distinction of the solid-state emitter from an ideal two-level system is the presence of phonons. Phonon-assisted far off-resonant optical excitations have been demonstrated in previous work\(^3\)\(^4\), but require high pump power (typically \(\approx 100\) times the \(\pi\)-pulse power with resonant excitation). We perform controlled experiments by isolating only the red or blue sideband to pump the quantum dot. Figure 3b shows the single-photon intensity as a function of pump power. With a laser power of \(\approx 115\) nW, which corresponds to the \(\pi\)-pulse of the two-colour excitation, the isolated phonon-assisted blue and red sideband excitation, however, can achieve only 15\% and 7\% of population inversion, respectively, in agreement with the theoretical model\(^3\). These data reinforce our model in Fig. 1 that shows that because the two-colour pulses are phase-locked, they should be effectively viewed as a resonant single-colour pulse.

Phase control of the single photons

Next, we send the two-colour pulse through a stable Sagnac interferometer as shown in Fig. 4a, where the red and blue sidebands are split into two paths with an adjustable time delay (thus, the phase \(\phi\)) and then recombined on the output beamsplitter. Figure 4b shows the detected resonance fluorescence counts as a function of the relative phase for two examples of driving field strength. In the weak excitation regime (bottom panel), the population of the excited state is approximately proportional to the effective input pulse area that is modulated by the relative phase \(\phi\); thus, the oscillation of the resonance fluorescence counts with the pulse delay is close to a sinusoidal function. This phenomenon can also be described as the coherent superposition of the excited-state wavefunction created by two pulses in the low-excitation limit\(^3\). In the strong excitation regime (upper panel), the photon counts are dependent on the effective pulse area. The photon counts will drop when the pulse area is larger than the \(\pi\)-pulse as the relative phase \(\phi\) varies, so a dip will appear at the peak of the sinusoidal oscillation. A two-dimensional map of the phase-sensitive interference fringes at varying phase delay and excitation strength is shown in Fig. 4c, which is in good agreement with the numerical simulations (see Supplementary Information) in Fig. 4d, showing a gradual transition from a sinusoidal fringe in the weak power regime to a more complex structure in the strong power regime. We note that the minimum value in Fig. 4b does not drop to zero, which is mainly due to the phonon-induced dephasing\(^3\) and remaining asymmetry of the red and blue pulses (such as the amplitude and linewidth) in our experiment so that the effective pulse area starts from a non-zero value.

Photon purity and indistinguishability

Finally, we study the effect of this new coherent control technology on the purity and the indistinguishability of the emitted single photons, two key parameters for optical quantum computation\(^1\)\(^2\)\(^3\), boson sampling\(^1\)\(^2\) and quantum networks\(^3\). We first
Fig. 3 | Single-photon intensity as a function of the driving strength. a, The Rabi oscillations under one-colour and two-colour excitations. Under two-colour excitation, the single-photon intensity oscillates near sinusoidally as the pump power increases (purple circles). Compared with one-colour excitation (open squares), the maximum rate is 1.74 times higher after correcting the transmission efficiency of the 4f system, while the excitation power is 8.5 times higher. See Supplementary Information for the detailed description and loss budget of the optical elements. b, The single-photon intensity as a function of the driving power under dichromatic excitation (purple circles), and isolated phonon-assisted blue (blue symbols) and red (red symbols) sideband excitation. With increasing driving power, the counts climb slowly under the blue and red sideband excitation. When the two-colour excitation reaches the population inversion under the driving power of ~115 nW, the blue and red sideband excitation can achieve only 15% and 7% of the population inversion, respectively.

Fig. 4 | Phase-dependent resonance fluorescence under dichromatic driving. a, A Sagnac interferometer was employed to stably control the phase of the dichromatic laser pulses. First, two filters at two different paths filter the red- and blue-detuned pulse, respectively. Then, one phase shifter was inserted into one of the paths to adjust the relative phase. b, Under low excitation (bottom panel), the quantum interference of probability waves excited by the red- and blue-shifted pulses gives rise to sinusoidal oscillation of the photon counts when the phase changes. Under strong excitation (top panel), the photon counts depend on the effective pulse area. When the pulse area is larger than \( \pi \), there will be a dip at the peak of the sinusoidal oscillation. c, Phase-dependent quantum interference and Rabi oscillations under different excitation power. d, Theoretical simulation of the phase-dependent quantum interference and Rabi oscillations.
test the purity by second-order coherence measurements. Under π-pulse excitation and at zero relative phase between the two sidebands, we observe a vanishing multiphoton probability of $g^2(0) = 0.012 \pm 0.001$ in the collected photons (Fig. 5a). The photon indistinguishability is tested by using Hong–Ou–Mandel interference between two consecutively emitted single photons at a time delay of 13 ns. Figure 5b plots the time-delayed histograms of normalized counts for two photons prepared at orthogonal and parallel polarization, where the counts for the latter are markedly suppressed at zero time delay. From Fig. 5b, a degree of indistinguishability of $0.964 \pm 0.006$ is obtained between the two π-pulse-excited single photons. A closer inspection of the co-polarized two-photon counts around the zero delay shows a dip. This is due to temporal filtering by ultrafast timing resolution (~20 ps) of the superconducting nanowire single-photon detectors, which increase the interference visibility at the dip to $0.982 \pm 0.006$. These results indicate a high level of photon coherence using the dichromatic pulse excitation, with a similar quality to the purity and indistinguishability data obtained from the one-colour resonant excitation9,14–20 (see Supplementary Information). Finally, we note that the high degree of photon indistinguishability suggests that the dichromatic excitation induces no additional time jitter, thus further excluding the model of phonon-assisted excitation. In the future, the two-colour excitation method could be combined with other techniques such as side excitation14,39 and polarized microcavities40 to generate optimal single-photon sources.

Online content
Any methods, additional references, Nature Research reporting summaries, source data, statements of code and data availability and associated accession codes are available at https://doi.org/10.1038/s41567-019-0585-6.

Data availability
The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

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 References
1. Rabi, I. I., Millman, S. & Kasch, P. The molecular beam resonance method for measuring nuclear magnetic moments. The magnetic moments of $^6\text{Li}$, $^7\text{Li}$ and $^9\text{F}$. Phys. Rev. 55, 526–535 (1939).
2. Ramsey, N. F. A molecular beam resonance method with separated oscillating fields. Phys. Rev. 78, 695–699 (1950).
3. Autler, S. H. & Townes, C. H. Stark effect in rapidly varying fields. Phys. Rev. 100, 703–722 (1955).
4. Mollow, B. R. Power spectrum of light scattered by two-level systems. Phys. Rev. 188, 1969–1975 (1969).
5. Bennett, C. H. & DiVincenzo, D. P. Quantum information and computation. Nature 404, 247–255 (2000).
6. Leibfried, D., Blatt, R., Monroe, C. & Wineland, D. Quantum dynamics of single trapped ions. Rev. Mod. Phys. 75, 281–324 (2003).
7. Bonadeo, N. H. et al. Coherent optical control of the quantum state of a single quantum dot. Science 282, 1473–1476 (1998).
8. Press, D., Ladd, T. D., Zhang, B. & Yamamoto, Y. Complete quantum control of a single quantum dot spin using ultrafast optical pulses. Nature 456, 218–221 (2008).
9. Lodahl, P., Mahmoodian, S. & Stobbe, S. Interfacing single photons and single quantum dots with photonic nanostructures. Rev. Mod. Phys. 87, 347–400 (2015).
10. Weber, J. R. et al. Quantum computing with defects. Proc. Natl Acad. Sci. USA 107, 8513–8518 (2010).
11. Clarke, J. & Wilhelm, F. K. Superconducting quantum bits. Nature 453, 1031–1042 (2008).
12. Zrenner, E. et al. Coherent properties of a two-level system based on a quantum-dot photodiode. Nature 418, 612–214 (2002).
13. Stevater, T. H. et al. Rabi oscillations of excitons in single quantum dots. Phys. Rev. Lett. 87, 133603 (2001).
14. Michler, P. et al. A quantum dot single-photon turnstile device. Science 290, 2283–2285 (2000).
15. Santori, C., Fattal, D., Vučković, J., Solomon, G. S. & Yamamoto, Y. Indistinguishable photons from a single-photon device. Nature 419, 594–597 (2002).
16. Muller, A. et al. Resonance fluorescence from a coherently driven semiconductor quantum dot in a cavity. Phys. Rev. Lett. 99, 187402 (2007).
17. Vamivakas, A. N., Zhao, Y., Lu, C.-Y. & Atature, M. Spin-resolved quantum-dot resonance fluorescence. Nat. Phys. 5, 198–202 (2009).
18. He, Y.-M. et al. On-demand semiconductor single-photon source with near-unity indistinguishability. Nat. Nanotechnol. 8, 213–217 (2013).
19. Buckley, S., Rivore, K. & Vučković, J. Engineered quantum dot single-photon sources. Rep. Prog. Phys. 75, 126503 (2012).
20. Senellart, P., Solomon, G. & White, A. High-performance semiconductor quantum-dot single-photon sources. Nat. Nanotechnol. 12, 1026–1039 (2017).
21. Ding, X. et al. On-demand single photons with high extraction efficiency and near-unity indistinguishability from a resonantly driven quantum dot in a micropillar. Phys. Rev. Lett. 116, 020401 (2016).
22. Somaschi, N. et al. Near-optimal single-photon sources in the solid state. Nat. Photon. 10, 340–345 (2016).
23. Wang, H. et al. Near-transform-limited single photons from an efficient solid-state quantum emitter. Phys. Rev. Lett. 116, 213601 (2016).
24. Aaronson, S. & Arkhipov, A. The computational complexity of linear optics. In Proc. 43rd Annu. ACM Symp. Theory of Computing 333–342 (ACM, 2011).
25. Kok, P. et al. Linear optical quantum computing with photonic qubits. Rev. Mod. Phys. 79, 135–174 (2007).
26. Müller, M. et al. On-demand generation of indistinguishable polarization-entangled photon pairs. Nat. Photon. 8, 224–228 (2014).
27. Schweikert, L. et al. On-demand generation of background-free single photons from a solid-state source. Appl. Phys. Lett. 112, 093106 (2018).
28. Hanschke, L. et al. Quantum dot single-photon sources with ultra-low multi-photon probability. npj Quantum Inf. 4, 43 (2018).
29. Huber, T. et al. Measurement and modification of biexciton–exciton time correlations. Opt. Express 21, 9890–9898 (2013).
30. Wang, H. et al. On-demand semiconductor source of entangled photons which simultaneously has high fidelity, efficiency, and indistinguishability. Phys. Rev. Lett. 122, 113602 (2019).
31. Hoon, H. et al. Interplay of Rabi oscillations and quantum interference in semiconductor quantum dots. Phys. Rev. Lett. 88, 087401 (2002).
32. Grange, T. et al. Reducing phonon-induced decoherence in solid-state single-photon sources with cavity quantum electrodynamics. Phys. Rev. Lett. 118, 253602 (2017).
33. Iles-Smith, J. et al. Phonon scattering inhibits simultaneous near-unity efficiency and indistinguishability in semiconductor single-photon sources. Nat. Photon. 11, 521–526 (2017).
34. Gheorghiu, M., Barth, A. M. & Aspelmeyer, M. Proposed robust and high-fidelity preparation of excitons and biexcitons in semiconductor quantum dots making active use of phonons. Phys. Rev. Lett. 110, 147401 (2013).
35. Quilter, J. H. et al. Phonon-assisted population inversion of a single InGaAs/GaAs quantum dot by pulsed laser excitation. Phys. Rev. Lett. 114, 137401 (2015).
36. Förstner, J. et al. Phonon-assisted damping of Rabi oscillations in semiconductor quantum dots. Phys. Rev. Lett. 91, 127401 (2003).
37. Pan, J. W. et al. Multiphoton entanglement and interferometry. Rev. Mod. Phys. 84, 777–838 (2012).
38. Humphreys, P. C. Deterministic delivery of remote entanglement on a quantum network. Nature 558, 268–273 (2018).
39. Ates, S. et al. Post-selected indistinguishable photons from the resonance fluorescence of a single quantum dot in a micropillar. Preprint at https://arxiv.org/abs/1809.10992 (2018).

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Author contributions
C.-Y.L. and J.-W.P. conceived the idea and designed the experiment. C.-Y.L. and J.-W.P. performed the experiment. C.S. and S.H. grew the quantum dot samples. Y.-M.H., H.W., C.W., X.D., J.Q., Z.-C.D., S.C., J.-P.L., R.-Z.L. and C.-Y.L. performed the theoretical modelling and analysed the experimental data. C.-Y.L. wrote the paper with C.-Y.L. and J.-W.P. and C.-Y.L. performed the experiment. C.S. and S.H. grew the quantum dot samples. Y.-M.H., H.W., C.W., X.D., J.Q., Z.-C.D., S.C., J.-P.L., R.-Z.L. and C.-Y.L. performed the experimental. C.-Y.L. and J.-W.P. supervised the whole project.

Competing interests
The authors declare no competing interests.

Additional information
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