Letter

Swing-up dynamics in quantum emitter cavity systems: Near ideal single photons and entangled photon pairs

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In the SUPER scheme (Swing-UP of the quantum EmitteR population), excitation of a quantum emitter is achieved with two off-resonant, red-detuned laser pulses. This allows the generation of high-quality single photons without the need of complex laser stray light suppression or careful spectral filtering. In the present work, we extend this promising method to quantum emitters, specifically semiconductor quantum dots, inside a resonant optical cavity. A significant advantage of the SUPER scheme is identified in that it eliminates re-excitation of the quantum emitter by suppressing photon emission during the excitation cycle. This, in turn, leads to almost ideal single-photon purity, overcoming a major factor typically limiting the quality of photons generated with quantum emitters in high-quality cavities. We further find that for cavity-mediated biexciton emission of degenerate photon pairs, the SUPER scheme leads to near-perfect biexciton initialization with very high values of polarization entanglement of emitted photon pairs.

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Semiconductor quantum dots (QDs) have been intensely studied as sources of single photons and entangled photon pairs [1–17]. The most efficient generation of highest-quality photons [18–21], however, as well as their extraction as optical information carriers [22,23], remain challenges on the road to using QDs as on-demand photon sources in quantum information processing architectures [24–26]. A number of different excitation and photon extraction strategies have been explored and demonstrated over the years [27–33], however, each of the different approaches typically comes with specific limitations or difficulties [13,19–21,34]. To achieve spectral separation of excitation lasers and emitted photons, in the last few years, excitation using dichromatic pulses has moved into the spotlight [11,12,35]. The recently introduced SUPER (Swing-UP of the quantum EmitteR population) scheme follows a similar approach [36–38], avoiding from the outset typical problems brought about by near-resonant optical excitation. In the SUPER scheme, by use of two off-resonant red-detuned laser pulses, phonon scattering is minimized at low temperatures [39,40], and spectral filtering for photon detection is easily performed [22,23,41].

Here we extend this promising excitation method to a quantum emitter, specifically a semiconductor quantum dot, placed inside a resonant optical resonator as sketched in Fig. 1. In that setup, for near-resonant optical excitation with a Gaussian laser pulse, cavity-accelerated photon emission (and then re-excitation) would occur already during the excitation cycle, spoiling the single-photon character of the emission [2,42–44]. For the SUPER excitation scheme, here we theoretically show that this emitter re-excitation is greatly suppressed via a laser-induced AC-Stark shift, leading to almost ideal single-photon purity even for high-quality cavities approaching strong coupling. This way the SUPER scheme allows us to overcome a major drawback that limits the quality of photons generated with quantum emitter cavity systems. We further investigate the SUPER scheme for excitation of the biexciton state with subsequent cavity-mediated generation of pairs of degenerate polarization entangled photons [14,45]. We show that in contrast to pulsed resonant two-photon excitation, the SUPER scheme avoids populating the cavity such that polarization entanglement is not reduced by the laser excitation [46].

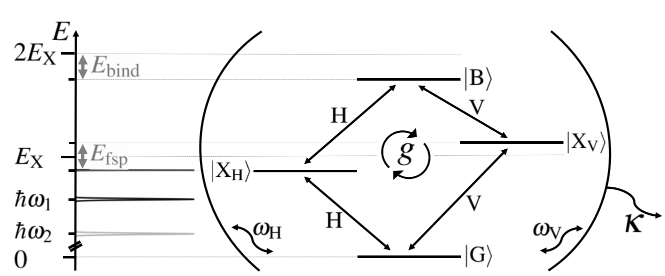

FIG. 1. Schematics of quantum dot cavity system. Electronic states considered are ground state, $|\mathrm{G}\rangle$, two exciton states, $|\mathrm{X_H}\rangle$ and $|\mathrm{X_V}\rangle$, with fine structure splitting E_{fsp}, and biexciton state, $|\mathrm{B}\rangle$, with binding energy E_{bind}. Electronic transitions are coupled to linearly polarized cavity modes with frequencies ω_{H} and ω_{V}, with coupling strength g and cavity photon loss κ. Spectra of the two red-detuned pulses of the SUPER scheme with H polarization at $\hbar\omega_1$ and $\hbar\omega_2$ and exciton emission are indicated. Energy differences shown are not to scale.

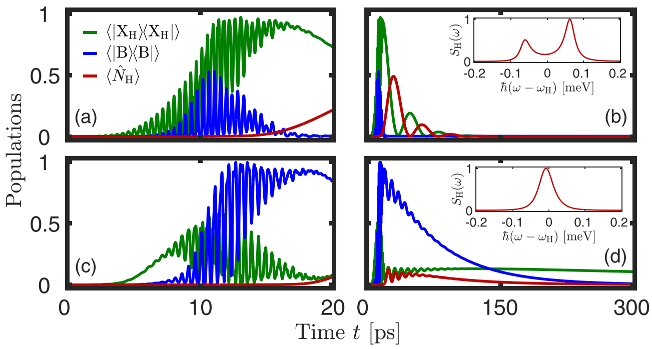

FIG. 2. Dynamics of SUPER excitation. Populations of H exciton (green), biexciton (blue), and H cavity mode (red). (a) and (b) Excitation of H exciton with cavity at X → G transitions for $E_{\text{bind}} = 3$ meV. Laser parameters as in first set of Table I. (c) and (d) Excitation of biexciton with cavity at two-photon B → G transition with $E_{\text{bind}} = 1$ meV. Laser parameters as in fourth set of Table I. (a) and (c) show the initial excitation period, (b) and (d) show time evolution up to 300 ps. The laser pulses are 3 ps long, centered at 10 ps. Cavity coupling is $g = 66\,\mu$eV, cavity loss $\hbar\kappa = g$. Insets in (b) and (d) show normalized H mode emission spectra $S_{\text{H}}(\omega)$ with frequencies relative to cavity frequency ω_{H}.

The SUPER scheme was proposed in Ref. [36], demonstrated in Refs. [37,38] and further analyzed in Ref. [47]. For excitation of the quantum emitter, the SUPER scheme uses a superposition of two off-resonant, red-detuned laser pulses, with laser field amplitude

$$\Omega(t) = \frac{\Omega_1}{\sqrt{2\pi\sigma_1^2}} e^{-t^2/(2\sigma_1^2)} e^{-i\omega_1 t} + \frac{\Omega_2}{\sqrt{2\pi\sigma_2^2}} e^{-(t-\Delta t)^2/(2\sigma_2^2)} e^{-i\omega_2(t-\Delta t)+i\phi}. \quad (1)$$

We use horizontal (H) polarization of this laser; Ω_i denotes pulse area, σ_i duration and ω_i frequency of respective pulses with $i = 1, 2$. Δt denotes temporal shift and ϕ phase shift between pulses ($\phi = 0$ unless otherwise noted). When a quantum emitter (in the simplest case a two-level system) is excited from the ground state with an appropriate choice of pulse parameters, hereafter referred to as parameter sets, a Swing-UP of the quantum EmitteR population (SUPER) to the excited state is observed, initializing the emitter (cf. Fig. 2 for results with cavity). With both pulses being on the low-energy side of the resonant transition, at low temperatures, even in condensed matter environments, adverse phonon influences in the excitation process are minimized [36,39,40], and spectral filtering of emitted photons is easily achieved.

Quantum dot cavity system. We model the lowest electronic excitations of a semiconductor QD as a four-level system [2,48] as sketched in Fig. 1, with typical parameters for high-quality InGaAs QDs [34]. We note that the specific choice of system parameters does not influence the main results and conclusions. We include the electronic ground state $|\text{G}\rangle$, two orthogonal exciton states $|\text{X}_\text{H}\rangle$ and $|\text{X}_\text{V}\rangle$, and the biexciton state $|\text{B}\rangle$. The two excitons have energies $E_{\text{X}_{\text{H,V}}} = E_\text{X} \mp \frac{E_{\text{fsp}}}{2}$ with $E_\text{X} = 1.366$ eV and fine structure splitting $E_{\text{fsp}} = 2\,\mu$eV. We note that the exact choice of fine structure splitting only

TABLE I. Examples of parameter sets for excitation. First two sets excite the H exciton for a QD with $E_{\text{bind}} = 3$ meV, the latter two sets excite the biexciton for a QD with $E_{\text{bind}} = 1$ meV. Pulse detunings are relative to G → X transition energy, $\hbar\Delta_i = \hbar\omega_i - E_{\text{X}_\text{H}}$.

$\hbar\Delta_1$ (meV)	$\hbar\Delta_2$ (meV)	Ω_1 (π)	Ω_2 (π)	σ_1 (ps)	σ_2 (ps)	Δt (ps)
−8	−17.53	32.00	32.01	3.61	3.42	0.01
−5	−11.3	25	33.33	4.0	4.0	0.0
−5	−12.14	30	30	1.37	1.37	0.0
−5	−12.99	36.88	36.88	3.0	3.0	4.74

has little influence on our excitation and photon emission results. However, for larger fine structure splittings overall lower degrees of polarization entanglement are observed, see, e.g., Ref. [45]. The biexciton has a binding energy of $E_{\text{bind}} = 3$ meV unless stated otherwise. The electronic transitions are coupled to the respective orthogonal linearly polarized cavity modes with frequencies ω_i, $i = \text{H,V}$, with coupling rate g and photon loss κ. We note that for the laser pulses in Eq. (1) it is implicitly assumed that they couple to an additional light mode and drive the electronic transitions in the H polarization channel. Time evolutions are calculated solving the von Neumann equation [48,49]. Correlation functions measuring the quality of generated photons are calculated using the quantum regression theorem [50]. Further details on theoretical modeling are given in Sec. I, Ref. [51]. One aspect we do not address in the present work is the influence of direct pumping of the cavity mode on the quantum properties of emitted photons. We note that with significantly off-resonant laser pulses this effect is expected to be relatively weak and at low temperature mostly of adiabatic nature. For a realistic investigation of this matter, detailed knowledge of the specific photonic environment would be needed. The latter could then together with the laser pulses be systematically optimized to achieve optimal emissive state initialization and emission, following Refs. [18,33].

First, we show that targeted excitation of one of the energy levels of the QD is possible and that the cavity mode does not hinder SUPER excitation. For initialization of either exciton or biexciton we study two different cases. In the first case, the H exciton is the target state for excitation while the QD is in a cavity with modes resonant with the X → G transitions; $\hbar\omega_i = E_i$, $i = \text{H, V}$. In the second case, the biexciton state is the target state with the cavity resonant with the two-photon B → G transition; $\hbar\omega_{\text{H,V}} = \frac{E_\text{B}}{2}$ for generation of polarization entangled photons [14,45,48]. To increase resonance enhancement of the relevant two-photon transition, a biexciton binding energy of 1 meV is used in the latter case.

In a first step, we identify ideal excitation parameters (by simple parameter sweeps) such that near-unity population of respective target states, H exciton or biexciton, is achieved. Following the general rules for SUPER excitation [36,47], for example, the parameter sets listed in Table I fulfill these criteria. To emphasize that parameter combinations are not unique for exciton or biexciton excitation, respectively, in Table I, we give two possible parameter sets for either case; the first two sets excite the H exciton, the last two sets excite the biexciton. For a given target excitation, we find a parameter sensitivity similar to resonant excitation. Slightly higher sensitivity to the pulse width was found for biexciton excitation. We note that while virtually no interpulse phase dependence is observed for the first two and the last parameter set of Table I, the third parameter set is significantly phase dependent. Generally, we find that phase insensitivity is found for parameters for which the product of interpulse detuning $\Delta\omega = \omega_1 - \omega_2$ and average pulse width $\overline{\sigma} = \frac{|\sigma_1+\sigma_2|}{2}$ fulfills $\hbar\Delta\omega \cdot \overline{\sigma} \gtrsim 20\,\text{meVps}$. In the main text, we only show results for phase-insensitive parameter sets with zero phase difference between pulses, $\phi = 0\,\pi$. Further discussion is given in Sec. IV, Ref. [51].

We now focus on excitation dynamics. Figures 2(a) and 2(b) show the case, where we excite the H exciton (first set from Table I) while Figs. 2(c) and 2(d) show the targeted biexciton excitation (fourth set from Table I). The two pulses are centered around 10 ps and about 3 ps long; Figs. 2(a) and 2(c) only show the main excitation window. Insets in Figs. 2(b) and 2(d) show the cavity H mode emission spectra [52], with coupling strength $g = 66\,\mu\text{eV}$ and $\hbar\kappa = g$. Results for lower cavity coupling, $\hbar\kappa = 4g$, look qualitatively very similar; see Fig. A.1, Ref. [51]. The swing-up dynamics are very similar to the observations without cavity [47]. However, if we compare these swing-up results with resonant Gaussian π-pulse excitation (not shown), one very important difference is that the cavity does not get populated with photons during SUPER excitation, neither for exciton [Fig. 2(a)] nor for biexciton [Fig. 2(c)] excitation. For the exciton this is due to the AC-Stark shift [53] induced by the strong, detuned pulses in the SUPER scheme. After the excitation, we observe the expected vacuum Rabi oscillations. During the excitation, the QD transition is shifted out of resonance with the cavity and consequently no significant photon emission occurs during excitation (see Sec. III, Ref. [51] for details). This is the main reason, why the swing-up is very insensitive to the cavity parameters, with no need to adjust excitation parameters for different cavities. Premature emission and then emitter re-excitation, as observed for resonant excitation [2,42–44], is suppressed. Below we show that this turns out to be one major advantage of the SUPER scheme as, for the parameters of the present study, it leads to almost perfect single-photon purity even approaching strong emitter-cavity couplings. We note that for cavities with broader resonances this effect is expected to be quantitatively reduced.

Next we turn to biexciton excitation. Using a two-photon resonant Gaussian pulse, a transfer of H exciton excitation to the H polarized cavity mode is observed during excitation (not shown; see, e.g., Ref. [32]). This causes a different emission behavior in the differently polarized H or V modes, inducing which-path information and reducing polarization entanglement. Below we show that for the SUPER scheme, where photons are created equally in H and V cavity modes, for degenerate two-photon emission into the cavity, polarization entanglement is insensitive to the excitation.

The insets in Fig. 2 show the cavity emission spectra for the two cases of exciton (b) and biexciton (d) excitation. For exciton excitation a significant asymmetry is observed, again induced by the AC-Stark shift, consistent with Sec. III, Ref. [51], and a vacuum Rabi splitting is observed. For biexciton excitation, no significant asymmetry is found.

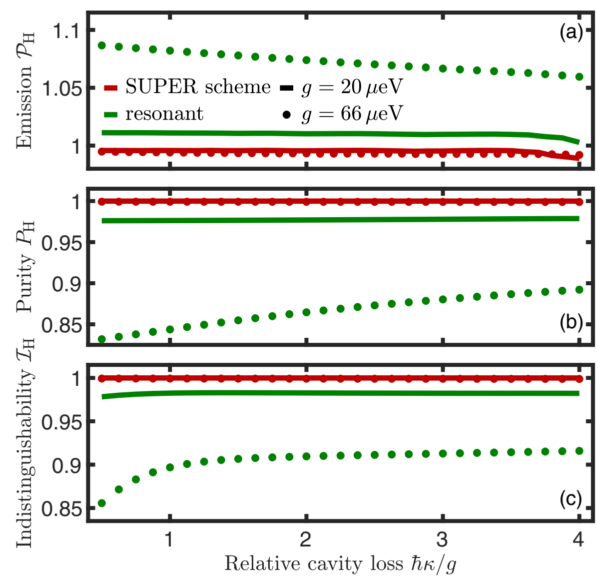

FIG. 3. Single-photon generation. Quality measures for photons emitted from the H polarization cavity mode for a cavity resonant with the X → G transition. Excitation with the SUPER scheme (red; first set of Table I) or with a resonant Gaussian π-pulse (green; width $\sigma = 3.5$ ps), respectively, for two different QD-cavity couplings $g = 20\,\mu\text{eV}$ (solid lines) and $g = 66\,\mu\text{eV}$ (dotted). Shown are (a) emission probability $\mathcal{P}_H$, (b) photon purity P_H, and (c) photon indistinguishability $\mathcal{I}_H$.

Single-photon source. Let us now investigate the potential of SUPER excitation of a single-photon emitter. To most efficiently extract the emitted single photons the cavity is resonant with the X → G transition [as in Figs. 2(a) and 2(b)]. We now analyze the quality of photons emitted from the H cavity mode. Besides photon emission probability $\mathcal{P}_i$ [41], we consider the purity P_i as a measure for the single-photon character [36], and photon indistinguishability $\mathcal{I}_i$ [54]. Definitions of these quantities are given in Sec. I, Ref. [51]. Results are shown in Fig. 3 for different cavity loss rates $\hbar\kappa \in [\frac{1}{2}\,g, 4\,g]$ and fixed cavity couplings $g = 20\,\mu\text{eV}$ (solid lines) and $g = 66\,\mu\text{eV}$ (dotted). In Fig. 3, we compare data for the emitter excited with the SUPER scheme (red; first parameter set of Table I) with data for resonant excitation with a Gaussian π-pulse with width $\sigma = 3.5$ ps (green). For excitation with the Gaussian pulse we consistently find emission probabilities larger than unity. This is because of premature photon emission into the cavity mode while the pulse is still present and then re-excitation of the quantum emitter (most pronounced for small cavity loss κ and large coupling g). For SUPER excitation, the emission probability is consistently just slightly below the ideal value of unity with an insignificant decline for increasing cavity loss κ. For excitation with the Gaussian pulse this decline is significantly more pronounced, especially for larger cavity coupling $g = 66\,\mu\text{eV}$. Apart from showing the insensitivity of the SUPER scheme to the cavity parameters, this indicates, that no re-excitation occurs for excitation with the SUPER scheme (in contrast to the resonant excitation). For the SUPER scheme, this can be understood by the AC-Stark shift induced by the off-resonant pulses, shifting the X → G transition out off resonance with the cavity mode during excitation (cf. spectrally resolved emission shown in Sec. III, Ref. [51]). This is also reflected in the single-photon purity in Fig. 3(b). For the SUPER scheme we find purity of almost exactly 1 for both cavity coupling rates and all cavity loss rates. When using resonant excitation, the purity increases with increasing cavity loss (more pronounced for larger cavity coupling g), however, it remains significantly below unity. This is consistent with emission probabilities in Fig. 3(a) being larger than unity as consequence of re-excitation events. The indistinguishability in Fig. 3(c) shows similar trends as the purity. And again, the SUPER scheme convincingly outperforms resonant excitation. Very similar results are found for SUPER excitation of a two-level system inside a resonant cavity (not shown). For completeness, we also note that the photon quality for the resonantly driven emitter is generally decreased with increasing pulse width (not shown).

These results showcase the superiority of SUPER excitation (over resonant excitation) leading to cavity-enhanced emission of single photons with near-ideal quality that are spectrally well separated from the excitation laser. In the present study we focus on the intrinsic differences brought about by different optical excitation methods, loss mechanisms that may largely differ for different quantum emitter systems and cavity designs such as dephasing, radiative loss, and phonon-induced cavity feeding are not discussed in detail here.

Entangled photon emission. While SUPER excitation of the quantum dot biexciton was discussed above, we now investigate the properties of (polarization entangled) photons emitted into the cavity mode at half the biexciton energy in the resonant degenerate two-photon emission process [14,45,48], which was previously shown to be rather insensitive to fine structure splitting [14,55]. We note that the emission configuration can be extended to two-color schemes with two cavity modes per polarization channel, spectrally tuned to either two-color two-photon emission [55] or in resonance with the single-photon transitions. As a measure of polarization entanglement, we calculate the concurrence $\mathcal{C}$ [56–58] and emission occurs into two polarization channels, with probabilities $\mathcal{P}_{\mathrm{H,V}}$ [41]; definitions are given in Sec. I, Ref. [51]. As discussed above, here $E_{\mathrm{bind}} = 1\,\mathrm{meV}$ and $g = 66\,\mu\mathrm{eV}$. Figure 4 shows results for different cavity loss rates $\hbar\kappa \in [\frac{1}{2}\,g, 4\,g]$. Besides the results for SUPER excitation (red; fourth parameter set of Table I), as references, we include results for two-photon resonant biexciton excitation with a Gaussian pulse (green; $\hbar\omega = \frac{E_{\mathrm{B}}}{2}$, pulse area $\Omega = 3.3\,\pi$, width $\sigma = 3\,\mathrm{ps}$), and results for an initially excited biexciton (blue). For the latter, the emission probability equals unity for both H and V mode. For SUPER excitation very similar values are obtained, with values for the H mode just slightly below unity over the entire parameter range. For two-photon resonant excitation, emission probabilities in the two cavity modes significantly differ, with values greater (less) than unity for the H mode (V mode) and pronounced changes with the cavity loss rate. In this case, the excitation process leads to residual population in the H mode, significantly also reducing the observed degree of polarization entanglement as measured by the concurrence; see Fig. 4(b). This behavior further increases for larger pulse widths (not shown). In Fig. 4(b), we observe the highest

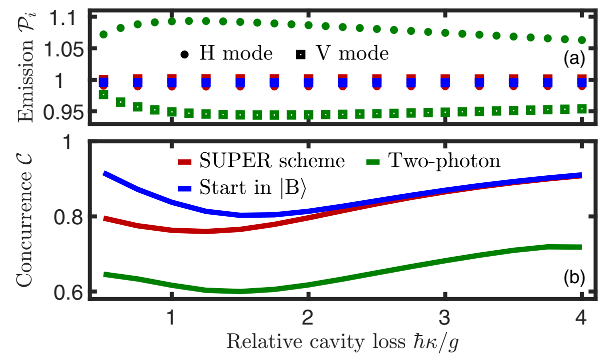

FIG. 4. Polarization entangled photon pairs for cavity resonant with degenerate B → G two-photon transition for $g = 66\,\mu\mathrm{eV}$. Shown are biexciton excitation with the SUPER scheme (red; fourth set of Table I), with a resonant Gaussian pulse (green; pulse area $\Omega = 3.3\,\pi$, width $\sigma = 3\,\mathrm{ps}$, $\hbar\omega = \frac{E_{\mathrm{B}}}{2}$), and for initially excited biexciton (blue). Shown are (a) emission probabilities $\mathcal{P}_{\mathrm{H,V}}$ of photons from H (points) and V (squares) mode, respectively, and (b) concurrence $\mathcal{C}$.

concurrence values for the idealized scenario of the initially excited biexciton. However, this ideal case outperforms the SUPER excitation only for the case of small cavity loss κ, with the SUPER scheme being a close second for larger κ.

In conclusion, we have analyzed and demonstrated the potential that the SUPER scheme offers for excitation of photonic quantum emitters inside an optical cavity. Semiconductor quantum dots serve as a typical example here. While high excitation fidelities of emmissive states (exciton and biexciton, respectively) are achieved for wide parameter ranges, quantum properties of emitted single photons and polarization entangled photon pairs are by far superior to resonant excitation. As one main result, for single-photon generation we find that photon emission and emitter re-excitation during excitation are entirely suppressed. This holds true even for systems approaching the strong coupling regime where photon emission occurs on timescales comparable to duration of excitation pulses. For SUPER excitation, this leads to very high and near-ideal single-photon purity and indistinguishability over the entire parameter range investigated.

Finally, we note that during review of the present paper two related papers on quantum emitter resonator systems were published [59,60]. In Ref. [60], it was shown that phonons do not have a significant influence on photon qualities in two-photon emission when exciting with the SUPER scheme. Reference [35] was significantly extended during review of our own paper (cf. Refs. [35,59]). In the final version [59], the phonon influence on SUPER excitation was shown to not significantly affect the quality of emitted single photons. Very recently also high-quality single-photon emission from a trion state initialized by swing-up excitation in a circular Bragg reflector was demonstrated experimentally [61].

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[1] X. Ding, Y. He, Z.-C. Duan, N. Gregersen, M.-C. Chen, S. Unsleber, S. Maier, C. Schneider, M. Kamp, S. Höfling, C.-Y. Lu, and J.-W. Pan, 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).

[2] L. Hanschke, K. A. Fischer, S. Appel, D. Lukin, J. Wierzbowski, S. Sun, R. Trivedi, J. Vučković, J. J. Finley, and K. Müller, Quantum dot single-photon sources with ultra-low multi-photon probability, npj Quantum Inf. **4**, 43 (2018).

[3] L. Schweickert, K. D. Jöns, K. D. Zeuner, S. F. C. da Silva, H. Huang, T. Lettner, M. Reindl, J. Zichi, R. Trotta, A. Rastelli, and V. Zwiller, On-demand generation of background-free single photons from a solid-state source, Appl. Phys. Lett. **112**, 093106 (2018).

[4] Y. Chen, M. Zopf, R. Keil, F. Ding, and O. G. Schmidt, Highly-efficient extraction of entangled photons from quantum dots using a broadband optical antenna, Nat. Commun. **9**, 2994 (2018).

[5] D. Huber, M. Reindl, S. F. Covre da Silva, C. Schimpf, J. Martín-Sánchez, H. Huang, G. Piredda, J. Edlinger, A. Rastelli, and R. Trotta, Strain-tunable GaAs quantum dot: A nearly dephasing-free source of entangled photon pairs on demand, Phys. Rev. Lett. **121**, 033902 (2018).

[6] J. Liu, R. Su, Y. Wei, B. Yao, S. F. C. d. Silva, Y. Yu, J. Iles-Smith, K. Srinivasan, A. Rastelli, J. Li, and X. Wang, A solid-state source of strongly entangled photon pairs with high brightness and indistinguishability, Nat. Nanotechnol. **14**, 586 (2019).

[7] H. Wang, H. Hu, T.-H. Chung, J. Qin, X. Yang, J.-P. Li, R.-Z. Liu, H.-S. Zhong, Y.-M. He, X. Ding, Y.-H. Deng, Q. Dai, Y.-H. Huo, S. Höfling, C.-Y. Lu, and J.-W. Pan, On-demand semiconductor source of entangled photons which simultaneously has high fidelity, efficiency, and indistinguishability, Phys. Rev. Lett. **122**, 113602 (2019).

[8] D. Huber, M. Reindl, Y. Huo, H. Huang, J. S. Wildmann, O. G. Schmidt, A. Rastelli, and R. Trotta, Highly indistinguishable and strongly entangled photons from symmetric GaAs quantum dots, Nat. Commun. **8**, 15506 (2017).

[9] M. Müller, S. Bounouar, K. D. Jöns, M. Glässl, and P. Michler, On-demand generation of indistinguishable polarization-entangled photon pairs, Nat. Photon. **8**, 224 (2014).

[10] H. Wang, Z.-C. Duan, Y.-H. Li, S. Chen, J.-P. Li, Y.-M. He, M.-C. Chen, Y. He, X. Ding, C.-Z. Peng, C. Schneider, M. Kamp, S. Höfling, C.-Y. Lu, and J.-W. Pan, Near-transform-limited single photons from an efficient solid-state quantum emitter, Phys. Rev. Lett. **116**, 213601 (2016).

[11] Y.-M. He, H. Wang, C. Wang, M.-C. Chen, X. Ding, J. Qin, Z.-C. Duan, S. Chen, J.-P. Li, R.-Z. Liu, C. Schneider, M. Atatüre, S. Höfling, C.-Y. Lu, and J.-W. Pan, Coherently driving a single quantum two-level system with dichromatic laser pulses, Nat. Phys. **15**, 941 (2019).

[12] Z. X. Koong, E. Scerri, M. Rambach, M. Cygorek, M. Brotons-Gisbert, R. Picard, Y. Ma, S. I. Park, J. D. Song, E. M. Gauger, and B. D. Gerardot, Coherent dynamics in quantum emitters under dichromatic excitation, Phys. Rev. Lett. **126**, 047403 (2021).

[13] B. Jonas, D. Heinze, E. Schöll, P. Kallert, T. Langer, S. Krehs, A. Widhalm, K. D. Jöns, D. Reuter, S. Schumacher, and A. Zrenner, Nonlinear down-conversion in a single quantum dot, Nat. Commun. **13**, 1387 (2022).

[14] S. Schumacher, J. Förstner, A. Zrenner, M. Florian, C. Gies, P. Gartner, and F. Jahnke, Cavity-assisted emission of polarization-entangled photons from biexcitons in quantum dots with fine-structure splitting, Opt. Express **20**, 5335 (2012).

[15] *Quantum Optics with Semiconductor Nanostructures* , edited by F. Jahnke (Woodhead Publishing, Philadelphia, 2012).

[16] S. Strauf, N. G. Stoltz, M. T. Rakher, L. A. Coldren, P. M. Petroff, and D. Bouwmeester, High-frequency single-photon source with polarization control, Nat. Photon. **1**, 704 (2007).

[17] N. Tomm, S. Mahmoodian, N. O. Antoniadis, R. Schott, S. R. Valentin, A. D. Wieck, A. Ludwig, A. Javadi, and R. J. Warburton, Photon bound state dynamics from a single artificial atom, Nat. Phys. **19**, 857 (2023).

[18] T. Praschan, D. Heinze, D. Breddermann, A. Zrenner, A. Walther, and S. Schumacher, Pulse shaping for on-demand emission of single Raman photons from a quantum-dot biexciton, Phys. Rev. B **105**, 045302 (2022).

[19] J. Iles-Smith, D. P. S. McCutcheon, A. Nazir, and J. Mørk, Phonon scattering inhibits simultaneous near-unity efficiency and indistinguishability in semiconductor single-photon sources, Nat. Photon. **11**, 521 (2017).

[20] A. J. Ramsay, A. V. Gopal, E. M. Gauger, A. Nazir, B. W. Lovett, A. M. Fox, and M. S. Skolnick, Damping of exciton Rabi rotations by acoustic phonons in optically excited InGaAs/GaAs quantum dots, Phys. Rev. Lett. **104**, 017402 (2010).

[21] T. Huber, A. Predojević, H. Zoubi, H. Jayakumar, G. S. Solomon, and G. Weihs, Measurement and modification of biexciton-exciton time correlations, Opt. Express **21**, 9890 (2013).

[22] N. Somaschi, V. Giesz, L. De Santis, J. C. Loredo, M. P. Almeida, G. Hornecker, S. L. Portalupi, T. Grange, C. Antón, J. Demory, C. Gómez, I. Sagnes, N. D. Lanzillotti-Kimura, A. Lemaítre, A. Auffeves, A. G. White, L. Lanco, and P. Senellart, Near-optimal single-photon sources in the solid state, Nat. Photon. **10**, 340 (2016).

[23] Y.-J. Wei, Y.-M. He, M.-C. Chen, Y.-N. Hu, Y. He, D. Wu, C. Schneider, M. Kamp, S. Höfling, C.-Y. Lu, and J.-W. Pan, Deterministic and robust generation of single photons from a single quantum dot with 99.5% indistinguishability using adiabatic rapid passage, Nano Lett. **14**, 6515 (2014).

[24] M. Varnava, D. E. Browne, and T. Rudolph, How good must single photon sources and detectors be for efficient linear optical quantum computation? Phys. Rev. Lett. **100**, 060502 (2008).

[25] Y.-X. Gong, X.-B. Zou, T. C. Ralph, S.-N. Zhu, and G.-C. Guo, Linear optical quantum computation with imperfect entangled photon-pair sources and inefficient non–photon-number-resolving detectors, Phys. Rev. A **81**, 052303 (2010).

[26] T. Jennewein, M. Barbieri, and A. G. White, Single-photon device requirements for operating linear optics quantum computing outside the post-selection basis, J. Mod. Opt. **58**, 276 (2011).

[27] M. Reindl, J. H. Weber, D. Huber, C. Schimpf, S. F. Covre da Silva, S. L. Portalupi, R. Trotta, P. Michler, and A. Rastelli, Highly indistinguishable single photons from incoherently excited quantum dots, Phys. Rev. B **100**, 155420 (2019).

[28] S. E. Thomas, M. Billard, N. Coste, S. C. Wein, Priya, H. Ollivier, O. Krebs, L. Tazaïrt, A. Harouri, A. Lemaitre, I. Sagnes, C. Anton, L. Lanco, N. Somaschi, J. C. Loredo, and

P. Senellart, Bright polarized single-photon source based on a linear dipole, Phys. Rev. Lett. **126**, 233601 (2021).

[29] P.-L. Ardelt, L. Hanschke, K. A. Fischer, K. Müller, A. Kleinkauf, M. Koller, A. Bechtold, T. Simmet, J. Wierzbowski, H. Riedl, G. Abstreiter, and J. J. Finley, Dissipative preparation of the exciton and biexciton in self-assembled quantum dots on picosecond time scales, Phys. Rev. B **90**, 241404(R) (2014).

[30] H. Jayakumar, A. Predojević, T. Huber, T. Kauten, G. S. Solomon, and G. Weihs, Deterministic photon pairs and coherent optical control of a single quantum dot, Phys. Rev. Lett. **110**, 135505 (2013).

[31] S. Stufler, P. Machnikowski, P. Ester, M. Bichler, V. M. Axt, T. Kuhn, and A. Zrenner, Two-photon Rabi oscillations in a single $In_xGa_{1-x}As/GaAs$ quantum dot, Phys. Rev. B **73**, 125304 (2006).

[32] D. Bauch, D. Heinze, J. Förstner, K. D. Jöns, and S. Schumacher, Ultrafast electric control of cavity mediated single-photon and photon-pair generation with semiconductor quantum dots, Phys. Rev. B **104**, 085308 (2021).

[33] D. Bauch, D. Siebert, K. D. Jöns, J. Förstner, and S. Schumacher, On-demand indistinguishable and entangled photons using tailored cavity designs, Adv. Quantum Technol. **7**, 2300142 (2024).

[34] D. Heinze, D. Breddermann, A. Zrenner, and S. Schumacher, A quantum dot single-photon source with on-the-fly all-optical polarization control and timed emission, Nat. Commun. **6**, 8473 (2015).

[35] L. Vannucci and N. Gregersen, Phonon-decoupled di-chromatic pumping scheme for highly efficient and indistinguishable single-photon sources, arXiv:2209.07770v1.

[36] T. K. Bracht, M. Cosacchi, T. Seidelmann, M. Cygorek, A. Vagov, V. M. Axt, T. Heindel, and D. E. Reiter, Swing-up of quantum emitter population using detuned pulses, PRX Quantum **2**, 040354 (2021).

[37] Y. Karli, F. Kappe, V. Remesh, T. K. Bracht, J. Muenzberg, S. Covre da Silva, T. Seidelmann, V. M. Axt, A. Rastelli, D. E. Reiter, and G. Weihs, SUPER scheme in action: Experimental demonstration of red-detuned excitation of a quantum dot, Nano Lett. **22**, 6567 (2022).

[38] K. Boos, F. Sbresny, S. K. Kim, M. Kremser, H. Riedl, F. W. Bopp, W. Rauhaus, B. Scaparra, K. D. Jöns, J. J. Finley, K. Müller, and L. Hanschke, Coherent dynamics of the swing-up excitation technique, arXiv:2211.14289.

[39] J. H. Quilter, A. J. Brash, F. Liu, M. Glässl, A. M. Barth, V. M. Axt, A. J. Ramsay, M. S. Skolnick, and A. M. Fox, Phonon-assisted population inversion of a single InGaAs/GaAs quantum dot by pulsed laser excitation, Phys. Rev. Lett. **114**, 137401 (2015).

[40] C. Roy and S. Hughes, Polaron master equation theory of the quantum-dot Mollow triplet in a semiconductor cavity-QED system, Phys. Rev. B **85**, 115309 (2012).

[41] C. Gustin and S. Hughes, Efficient pulse-excitation techniques for single photon sources from quantum dots in optical cavities, Adv. Quantum Technol. **3**, 1900073 (2020).

[42] K. Müller, A. Rundquist, K. A. Fischer, T. Sarmiento, K. G. Lagoudakis, Y. A. Kelaita, C. Sánchez Muñoz, E. del Valle, F. P. Laussy, and J. Vučković, Coherent generation of nonclassical light on chip via detuned photon blockade, Phys. Rev. Lett. **114**, 233601 (2015).

[43] K. A. Fischer, L. Hanschke, M. Kremser, J. J. Finley, K. Müller, and J. Vučković, Pulsed Rabi oscillations in quantum two-level systems: Beyond the area theorem, Quantum Sci. Technol. **3**, 014006 (2018).

[44] K. A. Fischer, L. Hanschke, J. Wierzbowski, T. Simmet, C. Dory, J. J. Finley, J. Vučković, and K. Müller, Signatures of two-photon pulses from a quantum two-level system, Nat. Phys. **13**, 649 (2017).

[45] D. Heinze, A. Zrenner, and S. Schumacher, Polarization-entangled twin photons from two-photon quantum-dot emission, Phys. Rev. B **95**, 245306 (2017).

[46] T. Seidelmann, C. Schimpf, T. K. Bracht, M. Cosacchi, A. Vagov, A. Rastelli, D. E. Reiter, and V. M. Axt, Two-photon excitation sets limit to entangled photon pair generation from quantum emitters, Phys. Rev. Lett. **129**, 193604 (2022).

[47] T. K. Bracht, T. Seidelmann, Y. Karli, F. Kappe, V. Remesh, G. Weihs, V. M. Axt, and D. E. Reiter, Dressed-state analysis of two-color excitation schemes, Phys. Rev. B **107**, 035425 (2023).

[48] Y. Ota, S. Iwamoto, N. Kumagai, and Y. Arakawa, Spontaneous two-photon emission from a single quantum dot, Phys. Rev. Lett. **107**, 233602 (2011).

[49] C. Roy and S. Hughes, Influence of electron–acoustic-phonon scattering on intensity power broadening in a coherently driven quantum-dot–cavity system, Phys. Rev. X **1**, 021009 (2011).

[50] H. Carmichael, *Statistical Methods in Quantum Optics 1: Master Equations and Fokker-Planck Equations* (Springer-Verlag, Berlin, 1999).

[51] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevResearch.6.L012017 for details on theoretical modelling and numerical implementation. The swing-up dynamics in our model system are described in detail, also showing state evolutions and spectra for weaker cavity coupling. Additionally, theoretical analyses of the phase shift dependence of the SUPER scheme and the light-field induced spectral shifts of the transition energies are provided. All references are already included in the main text.

[52] I. M. Mirza and S. J. van Enk, Single-photon time-dependent spectra in quantum optomechanics, Phys. Rev. A **90**, 043831 (2014).

[53] M. Cosacchi, J. Wiercinski, T. Seidelmann, M. Cygorek, A. Vagov, D. E. Reiter, and V. M. Axt, On-demand generation of higher-order Fock states in quantum-dot–cavity systems, Phys. Rev. Res. **2**, 033489 (2020).

[54] C. Gustin and S. Hughes, Pulsed excitation dynamics in quantum-dot–cavity systems: Limits to optimizing the fidelity of on-demand single-photon sources, Phys. Rev. B **98**, 045309 (2018).

[55] D. Heinze, Quantenoptik mit Halbleiterquantenpunkten (German) [Quantum optics with semiconductor quantum dots], Ph.D. thesis, Universität Paderborn (2018).

[56] W. K. Wootters, Entanglement of formation of an arbitrary state of two qubits, Phys. Rev. Lett. **80**, 2245 (1998).

[57] M. Cygorek, F. Ungar, T. Seidelmann, A. M. Barth, A. Vagov, V. M. Axt, and T. Kuhn, Comparison of different concurrences characterizing photon pairs generated in the biexciton cascade in quantum dots coupled to microcavities, Phys. Rev. B **98**, 045303 (2018).

[58] T. Seidelmann, M. Cosacchi, M. Cygorek, D. E. Reiter, A. Vagov, and V. M. Axt, Different types of photon entanglement

from a constantly driven quantum emitter inside a cavity, Adv. Quantum Technol. **4**, 2000108 (2021).

[59] L. Vannucci and N. Gregersen, Highly efficient and indistinguishable single-photon sources via phonon-decoupled two-color excitation, Phys. Rev. B **107**, 195306 (2023).

[60] T. K. Bracht, M. Cygorek, T. Seidelmann, V. M. Axt, and D. E. Reiter, Temperature-independent almost perfect photon entanglement from quantum dots via the SUPER scheme, Optica Quantum **1**, 103 (2023).

[61] R. Joos, S. Bauer, C. Rupp, S. Kolatschek, W. Fischer, C. Nawrath, P. Vijayan, R. Sittig, M. Jetter, S. L. Portalupi, and P. Michler, Triggered telecom C-band single-photon source with high brightness, high indistinguishability and sub-GHz spectral linewidth, arXiv:2310.20647.