



Optomechanical wave mixing by a single quantum dot

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Wave mixing is an archetypical phenomenon in bosonic systems. In optomechanics, the bidirectional conversion between electromagnetic waves or photons at optical frequencies and elastic waves or phonons at radio frequencies is building on precisely this fundamental principle. Surface acoustic waves (SAWs) provide a versatile interconnect on a chip and thus enable the optomechanical control of remote systems. Here we report on the coherent nonlinear three-wave mixing between the coherent fields of two radio frequency SAWs and optical laser photons via the dipole transition of a single quantum dot exciton. In the resolved sideband regime, we demonstrate fundamental acoustic analogues of sum and difference frequency generation between the two SAWs and employ phase matching to deterministically enhance or suppress individual sidebands. This transfer between the acoustic and optical domains is described by theory that fully takes into account direct and virtual multiphonon processes. Finally, we show that the precision of the wave mixing is limited by the frequency accuracy of modern radio frequency electronics. © 2021 Optical Society of America under the terms of the [OSA Open Access Publishing Agreement](#)

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1. INTRODUCTION

The phenomenon of wave mixing is well known in nonlinear optics [1] and has been widely employed in numerous other wave phenomena [2–5]. Also, in optomechanics, coherent transduction between optical and radio frequencies has been achieved [6,7], recently in the limit of single vibrational and optical quanta [8]. In this field, surface acoustic waves (SAWs) [9] provide a versatile bus enabling the control of remote systems on a chip, both in the classical regime [10,11] and approaching the quantum domain [12]. Thus, SAWs are attractive to implement quantum gates and transduction in hybrid architectures [13–15] with most progress being made for superconducting artificial atoms [16–18] and spins of defect centers [19–21]. However, the relevant quantum mechanical transitions in these artificial atoms can be addressed by microwave photons or phonons that are of the same energy. For fully fledged transduction to optical frequencies, quantum emitters in the relevant spectral domain are attractive because of their atomic-like, i.e., quantum mechanical, nature. Here semiconductor quantum dots (QDs) are attractive because their transitions are naturally in the visible and near-infrared spectral region and can be further tuned by size and composition [22]. Furthermore, QDs can be embedded in electrically active devices [23–27], and the emitted single photons [28] and entangled photon pairs [29]

can be efficiently extracted by optical cavities [30,31] or interfaced with optical fibers [32]. Moreover, QDs couple to the dynamic strain of a SAW via the deformation potential [33–35]. Because SAW frequencies may even exceed the dot's natural linewidth, the emission spectrum splits up into a series of phononic sidebands (PSBs) [36,37]. These properties, in combination with the ability to coherently address the optical transition with lasers [5,38], make QDs ideally suited for coherent SAW-based optomechanical schemes overcoming the presumed detrimental effect of phonons on the coherence of the exciton [39–42].

Here we report on coherent nonlinear three-wave mixing between the coherent fields of two $\omega_{\text{SAW}}/2\pi \geq 1$ GHz SAWs and $\omega_{\text{opt}}/2\pi \approx 330$ THz optical laser photons via the dipole transition of a single QD exciton. To this end, the optical transition of this quantum emitter is dynamically modulated by up to two SAWs via deformation potential coupling, and the exciton transition is simultaneously driven by a resonant laser. Because our system is operated in the resolved sideband regime, sum and difference frequency generation processes between the two SAWs occur, which are directly observed in the scattered photon spectrum. This transfer between acoustic and optical domains obeys phase-matching conditions between the two SAWs, enabling the deliberate enhancement or suppression of optical spectral

components. We develop a full theoretical model considering both direct and virtual multiphonon processes, which is found to be in excellent agreement with our experimental findings. Finally, we show that the precision of the wave mixing is limited by the frequency accuracy of modern radio frequency electronics.

2. RESULTS

A. Frequency Tunable Phononic Sideband Generation

The experimental setup for coherent optomechanical spectroscopy of single QDs is illustrated in Fig. 1(a). It comprises a GaAs/AlAs Bragg-type semiconductor microcavity with a layer of self-assembled In(Ga)As QDs at the antinode of the optical field. A multi-passband interdigital transducer (IDT), lithographically patterned on the sample surface, facilitates tunable excitation of SAWs of frequency ω_{SAW} over four different frequency passbands. This is achieved by simply applying a radio frequency (rf) voltage, the frequency of which, ω_{rf} , is faithfully mapped onto the frequency of the resulting SAW, ω_{SAW} . Further details on the IDT designs and their characteristics are included in Section 1 of Appendix A and Supplement 1, Section S2. In the following, only three of the four passbands are exploited in

our experiment. The generated coherent acoustic field of the SAW propagates over distances $>2000 \mu\text{m}$ at the surface to the position of the optically interrogated QD. As shown in Fig. 1(a), the QD is dynamically strained by the SAW. The oscillating deformation potential induces an ω_{SAW} -periodic spectral modulation of the optical transition [33–35] of amplitude $\hbar\Delta$ given by $E_X + \Delta E(t) = E_X + \hbar\Delta \cdot \cos(\omega_{\text{SAW}}t)$. For the SAW amplitudes applied in our experiment, any additional modulation due to a piezoelectrically induced Stark shift [43] can be excluded. The exciton transition of a single QD is additionally excited by a narrowband continuous-wave laser, and the resonantly scattered photons are detected. Low optical pump powers are used to ensure that the coherence of the scattered single photons is determined by that of the laser [44]. Further details on the experiment are included in Section 2 of Appendix A. First, we confirm the anti-bunched nature of the emitted light by measuring the second-order correlation function $G^{(2)}(\tau)$ for the laser being detuned from the optical transition E_X . The experimental data plotted in dark red in Fig. 1(b) show two characteristic features, namely, a $T_{\text{SAW}} = 2\pi/\omega_{\text{SAW}} = 1.476 \text{ ns}$ periodic oscillation due to the dynamic modulation of the two-level system [37] and reduced coincidences, and photon anti-bunching with $G^{(2)} < 0.5$

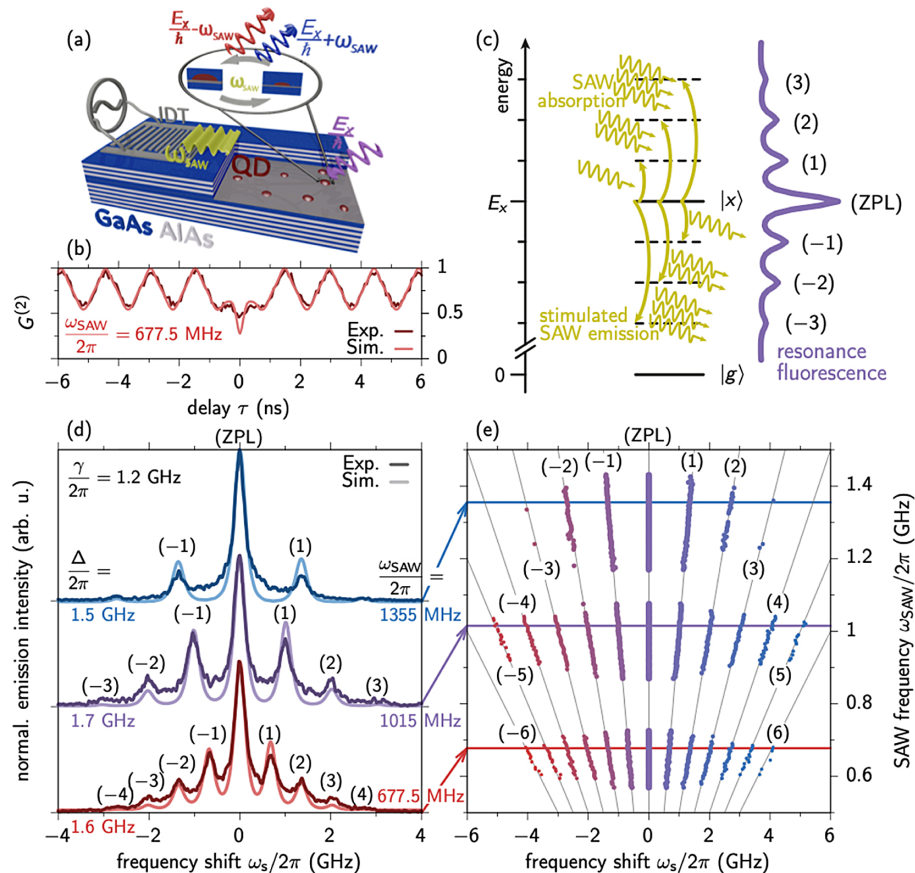


Fig. 1. Frequency tunable PSBs. (a) Experimental implementation comprising a semiconductor QD embedded in a Bragg-type microcavity. The QD exciton transition is excited by a laser tuned close to or in resonance with its exciton transition at E_X . SAWs are generated by IDTs on the sample surface to dynamically strain the remotely positioned QD. (b) Second-order correlation function $G^{(2)}(\tau)$ of a dynamically strained QD proving single-photon statistics. (c) Energy level diagram of the QD's SAW-modulated exciton transition being resonantly excited by a laser. Due to inelastic Stokes and anti-Stokes scattering processes, i.e., stimulated SAW phonon absorption and emission, the ZPL breaks up into a series of PSBs split by the phonon energy $\hbar\omega_{\text{SAW}}$. (d) Measured (dark lines) and calculated (light lines) scattered light spectra of a single QD dynamically strained by SAWs of three different SAW frequencies $\omega_{\text{SAW}}/2\pi$. The exciton decay $\gamma/2\pi$ and the SAW amplitudes $\Delta/2\pi$ used in the calculations are given in the plot. (e) Measured PSB position as a function of electrical frequency ω_{rf} applied to the IDT. The data nicely show the three passbands of the IDT separated by narrow stopbands. The gray lines mark the expected $\omega_s = m \cdot \omega_{\text{SAW}}$ dependence of the PSBs.

at $\tau = 0$ indicative for single-photon emission. We note that only a moderate reduction of $G^{(2)}$ below 0.5 is observed due to the limited time resolution of our detectors. Other exemplary $G^{(2)}(\tau)$ measurements can be found in Section 3 of Supplement 1. Second, we study the system in the frequency domain, where it is set in the resolved sideband regime. To this end, we tune the driving laser in resonance with the QD's optical transition and employ this resonant excitation condition for all experiments presented in the remainder of the paper. With these settings, the narrow zero phonon line (ZPL) of the QD at energy E_X splits into a series of PSBs due to Stokes and anti-Stokes scattering. In the former and latter process, a discrete number of SAW phonons is emitted into or absorbed from the coherent bosonic field of the SAW, respectively. As shown schematically in Fig. 1(c), these PSBs are precisely spaced by the energy of the driving SAW phonon $\hbar\omega_{\text{SAW}}$. Figure 1(d) shows typical scattered photon spectra of a single QD affected by SAWs with three different values of ω_{SAW} . The recorded spectra are plotted as dark lines as a function of the frequency shift $\omega_s = \omega - E_X/\hbar$, relative to the QD's exciton transition. The spectra show the expected creation of a series of emission lines split precisely by the selected $\omega_{\text{SAW}}/2\pi$. Our experimental data do not show any signatures of coherence loss such as line broadening. These data show that, apart from the single-photon nature of the emission, the QD behaves like a driven classical dipole that is modulated in time. Therefore, the sideband spectrum shows generic features characteristic of modulated light sources, similar to those demonstrated in a recent experiment on dynamic electrical modulation of quantum emitters [45]. Moreover, the $G^{(2)}(\tau)$ experiment in Fig. 1(b) and all spectra in Fig. 1(d) are well reproduced by simulations shown as light lines. In short, our theoretical model considers the QD exciton as an optically driven two-level system, described by the Hamiltonian $H = \frac{1}{2}(E_X + \Delta E(t))\sigma_z + \frac{1}{2}\hbar\Omega(\sigma_+ + \sigma_-)$ with the time-dependent shift of the transition energy $\Delta E(t) = \hbar\Delta \cdot \cos(\omega_{\text{SAW}}t)$. In these expressions, Ω denotes the Rabi frequency induced by the optical driving of the QD exciton and $\hbar\Delta$ the amplitude of the SAW modulation introduced above. $\sigma_{\pm} = \frac{1}{2}(\sigma_x \pm i\sigma_y)$ are the raising and lowering operators of the exciton two-level system with σ_i denoting the Pauli spin matrices. Further details are included in Appendix A, Section 3 and Supplement 1, Section 1. For the $G^{(2)}(\tau)$ data in Fig. 1(b) we considered a laser detuning of $1.6 \cdot \hbar\omega_{\text{SAW}}$ and found the best agreement for a relative SAW amplitude of $D = \Delta/\omega_{\text{SAW}} = 3$. The quantities for Fig. 1(d) are given next to each curve. Moreover, our theoretical model reproduces the experimental data almost perfectly using the same parameters as for the unmodulated QD. In particular, no additional pure dephasing rate is required. Owing to the multi-passband design of the employed transducers, the PSBs are fully tunable over a wide range of ω_{SAW} values. We demonstrate this in Fig. 1(e), where the measured positions of the PSBs are plotted as a function of the optical frequency shift ω_s and the SAW frequency ω_{SAW} . Here the IDT is driven by the electrical signal of frequency ω_{rf} and constant power $P_{\text{rf}} = +14$ dBm, and it generates SAWs with $\omega_{\text{SAW}} = \omega_{\text{rf}}$ across three of its wide frequency bands. These bands are separated by narrow stopbands where the IDT does not convert the electrical signal to a SAW. Details on the IDT design are provided in Appendix A, Section 1 and Supplement 1, Section 2. Consequently, PSBs are observed at the programmed frequencies $\omega_s = m \cdot \omega_{\text{SAW}}$, with $m = 0, \pm 1, \pm 2, \dots$. The number of resolved PSBs decreases with increasing ω_{rf} due to less

efficient SAW generation and a reduction of the mean phonon number in the coherent SAW field, reflected formally by the reduction of the relative SAW amplitude $D = \Delta/\omega_{\text{SAW}}$. Clearly, the PSB positions faithfully reproduce $\omega_{\text{SAW}} = \omega_{\text{rf}}$ in the optical domain, proving that the PSB spectrum can be easily tuned by changing ω_{rf} .

B. Sum and Difference Frequency Generation

In the next step, we demonstrate sum and difference frequency generation by using the QD exciton to mix two SAWs and the optical field, which thus leads to an optically induced acoustic nonlinearity in an otherwise linear medium. To this end, we generate two SAWs with frequencies $\omega_{\text{SAW}}^{(1)}$ and $\omega_{\text{SAW}}^{(2)}$ as described in Appendix A and record the scattered photon spectrum. In Fig. 2, we present two distinct cases. In the first case, the frequency of the second SAW is precisely twice that of the first SAW, i.e., $\omega_{\text{SAW}}^{(1)} = 1\omega_0$ and $\omega_{\text{SAW}}^{(2)} = 2\omega_0$, where ω_0 is the reference frequency. In the second case, we chose $\omega_{\text{SAW}}^{(1)} = 2\omega_0$ and $\omega_{\text{SAW}}^{(2)} = 3\omega_0$. We begin with $\omega_{\text{SAW}}^{(1)}/2\pi = 0.6775$ GHz and $\omega_{\text{SAW}}^{(2)}/2\pi = 1.3550$ GHz $= 2\omega_0/2\pi$ in Figs. 2(a) and 2(b), a scenario that is an acoustic equivalent to second-harmonic generation in nonlinear optics. As shown in the level diagram in Fig. 2(a), we expect that the $+1$ and $-1 \cdot \omega_0$ PSBs can be generated by a set of processes combining well-defined numbers of $1\omega_0$ (red arrows) and $2\omega_0$ (blue arrows) phonons. Up and down arrows indicate phonon absorption (anti-Stokes) and phonon emission (Stokes) processes, respectively. Combinations resulting in optical emission into the $+1$ or $-1 \cdot \omega_0$ PSB are indicated by dark and light arrows, respectively. Obviously, there exists the established single-phonon processes as for the case of a single SAW field shown in the left part. The next higher processes generating the $\pm 1 \cdot \omega_0$ PSBs require two phonons. Specifically, the $+1 \cdot \omega_0$ PSB is generated by the process marked by dark arrows in Fig. 2(a). It comprises absorption of a single $\omega_{\text{SAW}}^{(2)} = 2\omega_0$ phonon and emission of a single $\omega_{\text{SAW}}^{(1)} = 1\omega_0$ phonon. The underlying sum and difference frequency generation have to obey a phase-matching condition because they occur due to coherent wave mixing. This means that the intensity in each sideband depends on the relative phase ϕ of the two SAWs. In the following we reference this relative phase such that for $\phi = 0$ the two-phonon process described above is constructive. The opposite process, comprising the emission of one $\omega_{\text{SAW}}^{(2)}$ phonon and the absorption of one $\omega_{\text{SAW}}^{(1)}$ phonon, is marked by the light arrows in the figure and leads to pronounced emission into the $-1 \cdot \omega_0$ PSB. When $\phi = \pi$, the latter process is enhanced while the process dominating for $\phi = 0$ leading to $+1 \cdot \omega_0$ is suppressed. This expected phase matching is confirmed in our experiment, in which both frequency components are simultaneously generated by applying the two driving electric signals with the corresponding frequencies of powers $P_{\text{rf}}^{(1)} = -6$ dBm and $P_{\text{rf}}^{(2)} = -2$ dBm to the same IDT. In the data plotted as blue lines in Fig. 2(b), we clearly observe the expected anticorrelated enhancement and suppression of the two-phonon frequency mixing processes, leading to emission into the $+1$ or $-1 \cdot \omega_0$ PSB. For $\phi = 0$ (upper panel), the $+1 \cdot \omega_0$ PSB marked by the arrow is the dominant PSB while the $-1 \cdot \omega_0$ PSB is almost completely suppressed. When we set $\phi = \pi$ (lower panel), the situation is completely reversed and the $-1 \cdot \omega_0$ PSB (marked by arrow) is enhanced while the $+1 \cdot \omega_0$ PSB vanishes. We note that the asymmetry of the spectra is direct evidence for nonlinear and coherent mixing in our case of the two

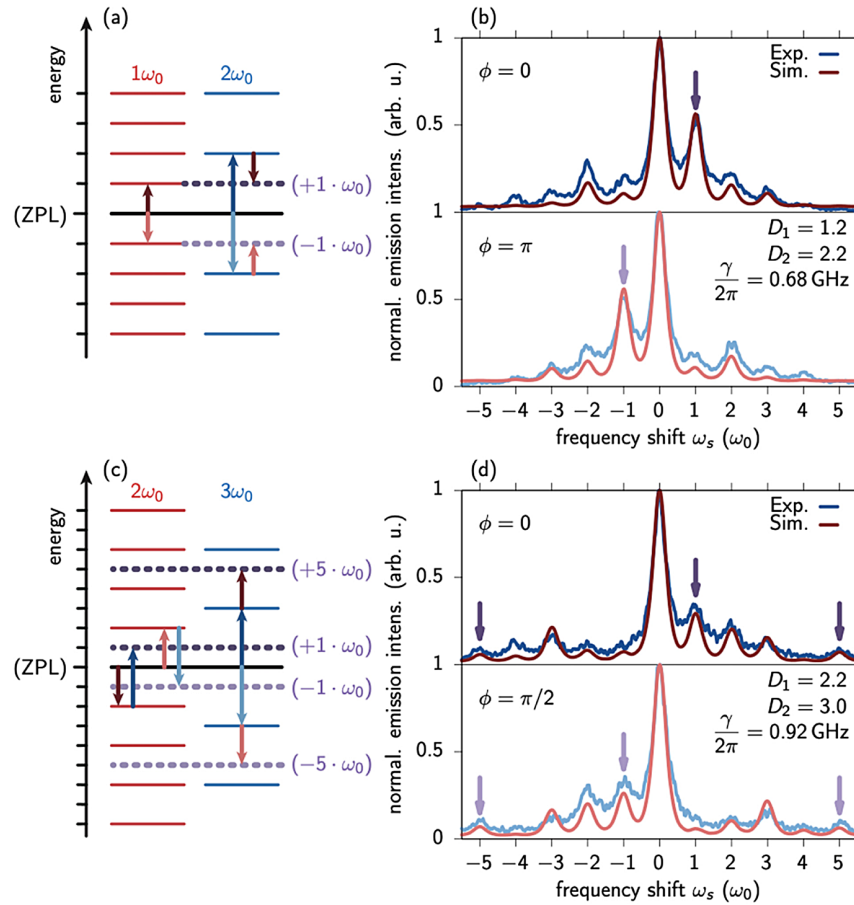


Fig. 2. Sum and difference frequency generation. (a) and (c) Energy level diagrams indicating sum and difference SAW frequency generation processes for the frequencies for (a) $\omega_{\text{SAW}}^{(2)} = 2\omega_{\text{SAW}}^{(1)}$ and (c) $2\omega_{\text{SAW}}^{(2)} = 3\omega_{\text{SAW}}^{(1)}$. (b) and (d) Measured (blue lines) and calculated (red lines) scattered light spectra corresponding to the two set SAW frequencies in (a) and (c), respectively. By tuning the relative phase between the two SAWs from $\phi = 0$ (upper panels) to [lower panel (b)] $\phi = \pi$ and [lower panel (d)] $\phi = \pi/2$, the intensities of different PSBs can be tuned due to phase matching.

bosonic acoustic fields and the laser's optical field through the QD's optical dipole. Furthermore, it is an acoustic analogue to the well-known phase matching in nonlinear optics and to the elimination of spectral components by all-optical quantum interference of two laser fields in an optical two-level system [46,47]. Note that in the case of incoherent mixing, symmetric spectra are expected with the combined PSBs' intensities being the sum of that when the SAWs are applied individually. The abovementioned sum and difference frequency generation is further corroborated by data for two SAW fields with commensurate frequencies $\omega_{\text{SAW}}^{(1)}$ and $\omega_{\text{SAW}}^{(2)}$ such that $\omega_{\text{SAW}}^{(1)}/2\pi = 2\omega_0/2\pi = 0.92$ GHz and $\omega_{\text{SAW}}^{(2)}/2\pi = 3\omega_0/2\pi = 1.38$ GHz. For this more general setting, no single-frequency SAW is generated at $\omega_{\text{SAW}} = \omega_0$, and thus emission of the $\omega_s = \pm 1 \cdot \omega_0$ PSBs is only possible via two-phonon processes with $\omega_s = \pm(\omega_{\text{SAW}}^{(2)} - \omega_{\text{SAW}}^{(1)})$. A similar situation holds for the $\pm 5 \cdot \omega_0$ PSBs, for which $\omega_s = \pm(\omega_{\text{SAW}}^{(2)} + \omega_{\text{SAW}}^{(1)})$ has to be fulfilled. In the level diagram in Fig. 2(c), these processes are marked by arrows, and the single-frequency forbidden PSBs are indicated by the dashed lines. Again, in experiment both frequency components are generated by electrical signals [$P_{\text{rf}}^{(1)} = 2$ dBm and $P_{\text{rf}}^{(2)} = 14$ dBm] applied to the same IDT. In the measured scattered photon spectra plotted in blue in Fig. 2(d), emissions of the $\pm 1 \cdot \omega_0$ and $\pm 5 \cdot \omega_0$ PSBs are clearly resolved. This observation is unambiguous, and it is direct evidence of the coherent

nonlinear sum and difference frequency generation of the two SAWs. Furthermore, the relative intensities of all PSBs are again fully tunable via the relative phase ϕ between the two SAWs. The upper and lower panels compare experimental spectra plotted in red for $\phi = 0$ and $\phi = \pi/2$, respectively. Again, similar to the data in Fig. 2(b), the curves are asymmetric, manifesting coherent wave mixing. Interestingly, for the selected phases, a pronounced asymmetry exists for the $\pm 1 \cdot \omega_0$ PSBs, while, in contrast, the $\pm 5 \cdot \omega_0$ PSBs exhibit similar intensities as indicated by the arrows. We extended our theoretical model to treat the experimentally employed dual acoustic modulation using relative SAW amplitudes $D_1 = \Delta^{(1)}/\omega_{\text{SAW}}^{(1)}$ and $D_2 = \Delta^{(2)}/\omega_{\text{SAW}}^{(2)}$, and a spontaneous emission rate $\gamma/2\pi$ as given in the plot. Note that we had to choose slightly different decay rates to fit the measured spectra which were recorded from different QDs. However, given the large parameter space, the determined combinations of values are reasonable choices. The relative phase ϕ is defined into the SAW modulation with the larger frequency. The simulated spectra are plotted in red in Figs. 2(b) and 2(d) and faithfully reproduce the experimental data. Most strikingly, the phase matching underlying the different sum and difference frequency generation processes are in excellent agreement.

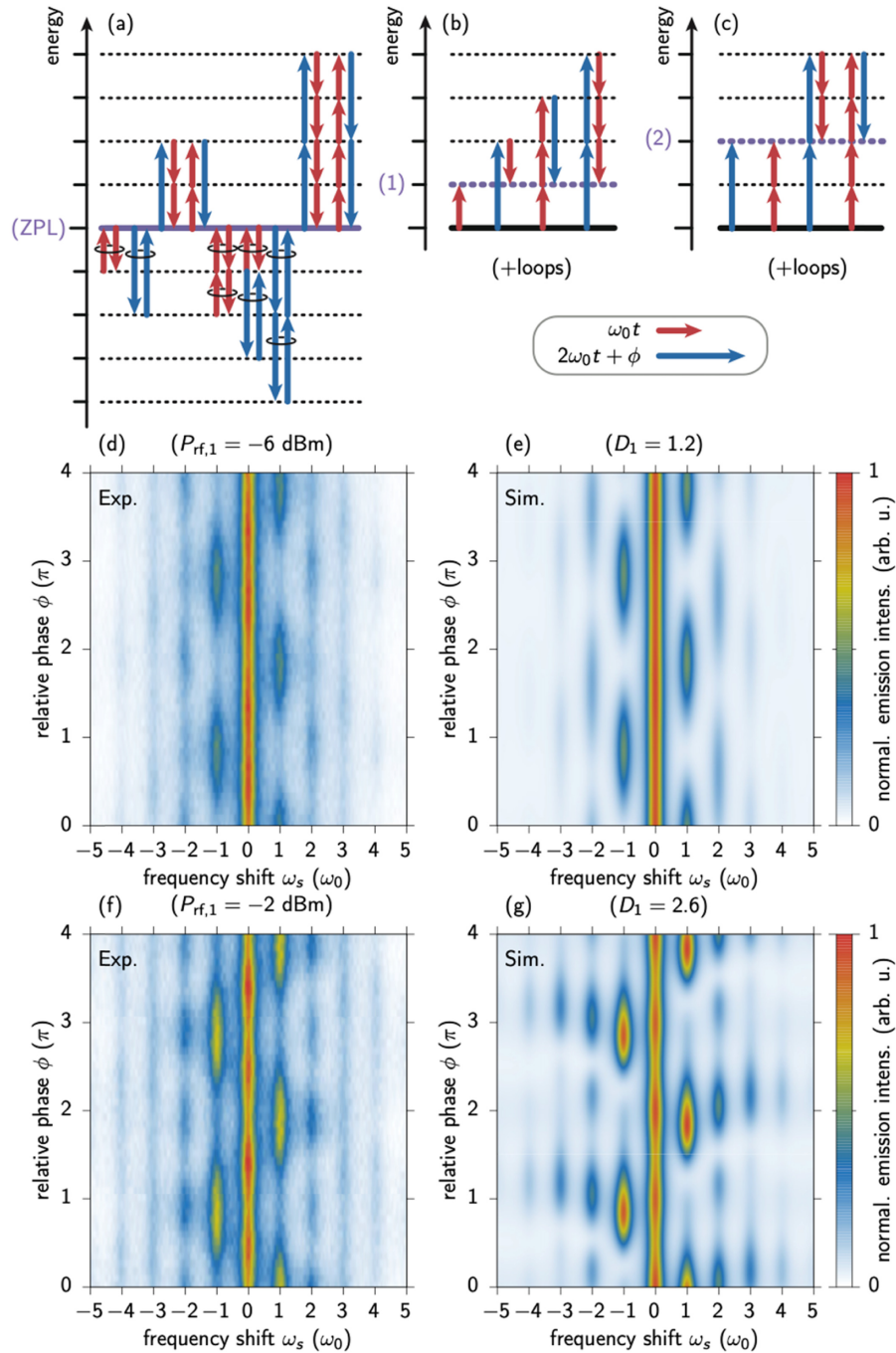


Fig. 3. Phase matching. (a)–(c) Energy level diagrams indicating the lowest $\omega_{\text{SAW}}^{(1)} = \omega_0$ (red) and $\omega_{\text{SAW}}^{(2)} = 2\omega_0$ (blue) phonon absorption (up arrows) and emission (down arrows) processes leading to photon emission into the (a) ZPL and the (b) first and (c) second PSBs, respectively. Virtual loop processes are indicated by coupled up and down arrows. All processes including loops are omitted in (b) and (c). (d) and (e) Relative phase dependence of the PSB spectrum in experiment and simulation, respectively. The normalized detected and calculated light intensities are color coded and plotted as a function of ω_s and ϕ . (f) and (g) Same as (d) and (e) but for a larger amplitude of the $1 \cdot \omega_0$ SAW as given in the plot.

C. Phase Matching

We continue addressing the phase matching (ϕ dependence) and higher-order wave-mixing processes for the case of $\omega_{\text{SAW}}^{(2)} = 2\omega_{\text{SAW}}^{(1)}$ both in experiment and theory. An analogous supporting analysis for the $2\omega_{\text{SAW}}^{(2)} = 3\omega_{\text{SAW}}^{(1)}$ case is included in Section 4.C of Supplement 1. We begin with higher-order processes giving rise to emission into the ZPL as well as the $+1 \cdot \omega_0$ and the $+2 \cdot \omega_0$ PSBs. Figures 3(a)–3(c) show the corresponding level diagrams

where the $\omega_{\text{SAW}}^{(1)}$ transitions are marked as red arrows and those with $\omega_{\text{SAW}}^{(2)}$ phonons as blue arrows. From left to right the number of involved phonons increases. Importantly, an $\omega_{\text{SAW}}^{(2)}$ process carries a relative phase ϕ with respect to an $\omega_{\text{SAW}}^{(1)}$ process as indicated in the figure. Figure 3(a) shows the transitions giving rise to emission at the ZPL. Since this signal requires that the energies of the involved phonons precisely cancel, at least two phonons, either $\omega_{\text{SAW}}^{(1)}$ or $\omega_{\text{SAW}}^{(2)}$, are required. In the coherent mixing process shown

on the left for both frequencies, two phonons form a loop with one phonon being emitted and one phonon being absorbed. In these loops, phonon emission and absorption contribute relative phases of opposite sign. Thus, this precise cancellation ensures that such loops only contribute to the phase dependence by a constant amplitude. This is no longer the case for the next higher processes, which involve three phonons. As shown in the second process of Fig. 3(a), one $\omega_{\text{SAW}}^{(2)}$ phonon is absorbed (blue up arrow) and two $\omega_{\text{SAW}}^{(1)}$ phonons are emitted (red down arrows), or vice versa. These processes do not involve any loops but a single $\omega_{\text{SAW}}^{(2)}$ process and therefore contribute a phase ϕ . The next higher processes are also shown in the level diagram and involve two loops, i.e., four phonons. Based on these arguments, we do not expect a pronounced ϕ modulation of the ZPL for low SAW amplitudes since such a modulation requires at least three phonons. In Fig. 3(b) we get back to the modulation of the $+1 \cdot \omega_0$ PSB and consider higher-order processes contributing to emission into this PSB. The next higher-order transition after that discussed in context of Fig. 2 [absorption of $\omega_{\text{SAW}}^{(2)}$ and emission of $\omega_{\text{SAW}}^{(1)}$] involves four phonons, emission of a single $\omega_{\text{SAW}}^{(2)}$, and absorption of three $\omega_{\text{SAW}}^{(1)}$ quanta. This process is followed by that shown in the right side of Fig. 3(b) formed by absorption of two $\omega_{\text{SAW}}^{(2)}$ and emission of three $\omega_{\text{SAW}}^{(1)}$ quanta. Finally, Fig. 3(c) shows an analogous consideration for emission into the $+2 \cdot \omega_0$ PSB. The lowest possible process is the absorption of an $\omega_{\text{SAW}}^{(2)}$ phonon. The next higher multiphonon processes resulting in a modulation of this PSB require four or five phonons, namely, the process of the absorption of two $\omega_{\text{SAW}}^{(2)}$ and emission of two $\omega_{\text{SAW}}^{(1)}$ quanta and that of emission of one $\omega_{\text{SAW}}^{(2)}$ and absorption of four $\omega_{\text{SAW}}^{(1)}$ quanta, respectively. These considerations show that the phase dependences of the different PSBs change when increasing the SAW amplitudes as higher phonon processes become more likely. Note that there exist several higher processes for emission of the $+1 \cdot \omega_0$ and $+2 \cdot \omega_0$ PSBs, which include loops introduced in the discussion of the ZPL modulation. These loops are omitted in Figs. 3(b) and 3(c) since they do not contribute a net phase. However, they are fully considered in our theoretical model. The complete set of transitions is discussed in Section 4.A of Supplement 1.

Next, we study the discussed higher-order processes in experiment and theory. We begin with the case of weak acoustic modulation. In Fig. 3(d), we present the measured optical emission of a single QD dynamically strained by two SAWs excited with powers of $P_{\text{rf}}^{(1)} = -6$ dBm and $P_{\text{rf}}^{(2)} = -2$ dBm. In this regime, we expect that processes involving a single phonon of each frequency will dominate. The normalized emission intensity is color coded and plotted as a function of ω_s (horizontal axis) and ϕ tuned over two full periods, $[0, 4\pi]$ (vertical axis). As expected, the $+1$ and $-1 \cdot \omega_0$ PSBs are the dominant PSBs. The intensities of these two PSBs exhibit precisely the expected 2π -periodic modulation. They are also perfectly anticorrelated when ϕ is tuned, which follows from the interplay between $\omega_{\text{SAW}}^{(1)}$ and $\omega_{\text{SAW}}^{(2)}$ components as explained in Section 1 of Supplement 1. Moreover, the ZPL's intensity remains almost constant when ϕ is scanned. As detailed above, this is expected because at least three phonons are required to induce a ϕ dependence. The $+2$ and $-2 \cdot \omega_0$ PSBs are less pronounced and are slightly modulated with a 2π -periodicity stemming from the trivial single $\omega_{\text{SAW}}^{(2)}$ phonon process. Next, we compare the experimental data to

our theoretical model, which includes all possible multiphonon processes contributing to the wave mixing. These processes include in particular those shown in Figs. 3(a)–3(c). A complete overview of the relevant processes up to the fifth order is included in Supplement 1 (Fig. S6) [19–21]. Figure 3(e) shows the results of the simulations using normalized SAW amplitudes $D^{(1)} = \Delta^{(1)}/\omega_{\text{SAW}}^{(1)} = 1.2$ and $D^{(2)} = \Delta^{(2)}/\omega_{\text{SAW}}^{(2)} = 1.5$ and an exciton decay rate $\gamma/2\pi = 0.81$ GHz. To better compare the results of our simulation to the experimental data, the simulation data in Fig. 3(e) are plotted in the same way as the experimental data in Fig. 3(d). Clearly, the result of our simulation is in excellent agreement with the experimental data and fully reproduces all characteristic features. In particular the phase-matching condition for emission into the $+1$ or $-1 \cdot \omega_0$ PSB is faithfully reproduced by our model.

The case of higher SAW amplitudes is studied in Fig. 3(f) (experiment) and Fig. 3(g) (theory). In experiment, we increased $P_{\text{rf}}^{(1)}$ to -2 dBm and kept $P_{\text{rf}}^{(2)} = -2$ dBm unchanged compared to the first case. Under these experimental conditions, the ZPL now exhibits a clear π -periodic modulation. This type of modulation is in fact expected for the combination of the two three-phonon processes in Fig. 3(a), in which each $\omega_{\text{SAW}}^{(2)}$ phonon contributes a ϕ dependence. In the same way, the additional structure on the $\pm 2 \cdot \omega_0$ PSB can be understood. In the next order, a three-phonon process with one $\omega_{\text{SAW}}^{(2)}$ phonon and an $\omega_{\text{SAW}}^{(1)}$ loop also leads to the 2π -periodicity also weakly resolved in Figs. 3(d) and 3(e). However, for larger SAW amplitudes, the four-phonon process, absorption (emission) of two $\omega_{\text{SAW}}^{(2)}$ phonons and emission (absorption) of two $\omega_{\text{SAW}}^{(1)}$ phonons, contributes significantly. This process involves two $\omega_{\text{SAW}}^{(2)}$ phonons, which gives rise to the observed π -periodicity. Again, the corresponding simulations using our theoretical model [$D^{(1)} = \Delta^{(1)}/\omega_{\text{SAW}}^{(1)} = 2.6$, $D^{(2)} = \Delta^{(2)}/\omega_{\text{SAW}}^{(2)} = 1.5$] shown in Fig. 3(g) nicely reproduce all characteristic π - and 2π -periodic features of the experimental data. The only minute deviation is a slight phase shift of the ZPL in comparison to the experiment in Fig. 3(f).

D. Precision

Finally, we study the sensitivity of our hybrid nonlinear optomechanical coupling scheme to variations of mechanical frequency. To this end we deliberately introduce a small yet finite detuning $\Delta\omega_{\text{SAW}}$ to the SAW such that $\omega_{\text{SAW}}^{(2)} = 2\omega_0 + \Delta\omega_{\text{SAW}}$. The resulting PSB alignment is shown schematically in Fig. 4(a). As depicted, introducing $\Delta\omega_{\text{SAW}}$ is equivalent to a time-varying relative phase $\phi(t)$ of the $\omega_{\text{SAW}}^{(2)} = 2\omega_{\text{SAW}}^{(1)}$ nonlinear mixing scheme. Owing the precision of state-of-the-art rf electronics, $\omega_{\text{SAW}}^{(1)}/2\pi = 0.65$ GHz and $\omega_{\text{SAW}}^{(2)}/2\pi = 1.30$ GHz can be set with a microhertz (μHz) resolution. Thus, the accessible subhertz frequency range of $\Delta\omega_{\text{SAW}}$ can easily be significantly smaller than the linewidth of the laser ($\delta\omega_{\text{laser}}/2\pi \leq 300$ kHz) and that of the exciton transition ($\delta\omega_X/2\pi \approx 1$ GHz). As proof of principle, we set $\Delta\omega_{\text{SAW}}/2\pi = 50$ μHz , corresponding to a time-dependent phase $\phi(t) = \Delta\omega_{\text{SAW}}t$, and detect the scattered photon spectrum as a function of time over one full cycle of this oscillation. In Fig. 4(b), we plot the recorded intensity in false-color representation as a function of time t (horizontal axis) and the relative optical frequency shift ω_s (vertical axis). The experimental data exhibit a clear oscillation of the intensities of the PSB and the ZPL

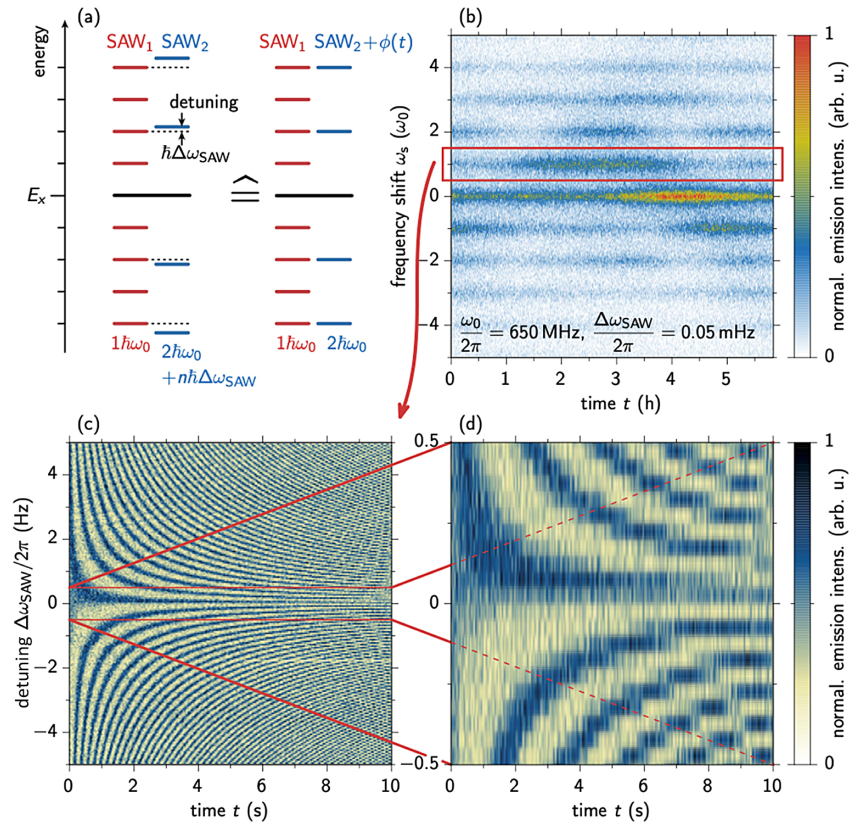


Fig. 4. Frequency ring generation. (a) Energy level diagram for finite detuning. The detuning $\Delta_{\text{SAW}}/2\pi$ gives rise to a time-dependent phase of the $\omega_{\text{SAW}} - 2\omega_{\text{SAW}}$ resonance condition. (b) Time evolution of the PSB spectrum for $\Delta_{\text{SAW}}/2\pi = 50 \mu\text{Hz}$. The observed period of the intensity oscillations matches the expected $T_\phi = 2\pi/\Delta_{\text{SAW}} \cong 5.55 \text{ h}$. (c) Color-coded intensity of the $+1 \cdot \omega_0$ PSB as a function of time and $\Delta_{\text{SAW}}/2\pi$, confirming the anticipated increase of the oscillation period as $|\Delta_{\text{SAW}}/2\pi|$ increases. (d) Zoom-in to the central part of the data shown in (c) (red box), highlighting the central symmetry of the observed pattern confirming the faithful mapping of $\Delta_{\text{SAW}}/2\pi$ onto T_ϕ .

with a period of precisely $T_\phi = 2\pi/\Delta\omega_{\text{SAW}} \cong 5.55 \text{ h}$. Remarkably, this demonstrated frequency rung is only $7.6 \cdot 10^{-14} \omega_{\text{SAW}}^{(1)}$ and $1.6 \cdot 10^{-19} E_X/\hbar$, underlining the outstanding stability of our hybrid nonlinear optomechanical scheme. Again, the frequency rung of the sideband intensity is fully tunable via Δ_{SAW} , which is studied in detail in Fig. 4(c). Here the intensity of the $+1 \cdot \omega_0$ PSB is plotted in false-color representation as a function of time (horizontal axis) and the SAW detuning $\Delta_{\text{SAW}}/2\pi$ (vertical axis). As the detuning is varied between -5 Hz and $+5 \text{ Hz}$, the period of the oscillation detected in the time domain faithfully changes accordingly, creating the pattern in Fig. 4(c) that is inversion symmetric with respect to the $\Delta\omega_{\text{SAW}} = 0$ line. This pattern can be seen even more clearly in Fig. 4(d), which is a zoom-in on the regime of small detunings $-0.5 \text{ Hz} \leq \Delta\omega_{\text{SAW}}/2\pi \leq +0.5 \text{ Hz}$ of the data shown in Fig. 4(c). An instructive theoretical reproduction of this pattern is given in Supplement 1, Section 4.D.

3. DISCUSSION

We have demonstrated optomechanical nonlinear three-wave mixing of two mutually coherent SAW fields and the optical field of a laser via the optical transition of a single QD. The phase matching underlying the interference enables deterministic enhancement or suppression of the different PSBs, which is faithfully detected in the resonantly scattered photon spectrum and accounted for by

our theory. While our particular implementation is based on epitaxial QDs made of III-V compound semiconductors, many other systems like defect centers in diamond [19,21], silicon carbide [20] or two-dimensional materials [48] have already been proven to be well suited to be interfaced with SAWs. Our observations are in excellent agreement with classical modulation theory, and the underlying principle can be transferred to other types of modulation mechanism, in particular electrical fields [45]. In the context of hybrid quantum technologies, the driving optomechanical interaction can be drastically enhanced by embedding the quantum emitter in phononic or optomechanical resonators, building on demonstrated monolithic integration on GaAs [10,49]. Ultimately, coherent reciprocal microwave-optical quantum state transduction comes into reach. For example, a single SAW phonon emitted by a microwave superconducting qubit can be transmitted on chip [18] to a single QD or another type of quantum emitter that converts it to a single photon employing the scheme demonstrated here. The required cooperativity can be achieved, however, only if the QD is efficiently coupled to an acoustic resonator. In our experiment, we demonstrated the transfer from the phononic to the photonic domain. To prove bidirectional transduction, i.e., back action on the phononic spectrum detected by the IDT, embedding of the QDs in acoustic resonators or waveguides is imperative. Furthermore, our SAW modulation paves the way toward dynamically reconfigurable control of integrated quantum photonic devices [50–53], for instance, for sideband-resolved single-photon routing. Alternatively, the heterointegration of the

semiconductor on LiNbO₃ harnesses the strong piezomechanical and nonlinear optical properties of the host substrate and the large optomechanical coupling of the compound semiconductor [54–57].

APPENDIX A: METHODS

1. SAMPLE DESIGN

The sample was grown by solid-source molecular beam epitaxy. It contains a single layer of In(Ga)As QDs embedded in a planar optical cavity formed by two distributed Bragg reflectors (DBRs) of 8 and 15 alternating layers of AlAs and GaAs on the top and bottom, respectively. The QDs were grown with low surface density to enable selective optical excitation of and detection from a single dot. The cavity resonance was designed to match the emission band of the QDs. The cavity quality factor of $Q \approx 150$ is sufficiently high to increase the light–matter interaction. At the same time, the corresponding cavity linewidth of $\Delta f \geq 2.2$ THz is sufficiently large to avoid dynamic modulation of the cavity resonance with an amplitude exceeding the cavity linewidth [58–60]. This ensures that we can exclude any time modulation of the Purcell effect as reported in our previous work on high- Q cavities [61]. Multiharmonic IDTs (Ti 5 nm, Al 50 nm) were patterned by standard electron beam lithography in a lift-off process directly on top of the sample to facilitate SAW excitation at the fundamental frequency and three overtones. The IDTs were either fabricated with a constant wavelength of $\lambda = 9.91 \mu\text{m}$ [34], or, in order to realize frequency chirped transducers [62], of wavelength linearly varying from $\lambda_0 = 8.94 \mu\text{m}$ to $\lambda_1 = 10.38 \mu\text{m}$ along the length ($L_{\text{IDT}} = 400 \mu\text{m}$) of the IDT. The first design allows SAW generation at a fundamental frequency of about 338 MHz and at overtones of 677, 1015, and 1355 MHz, while the second design enables SAW generation across four frequency bands, nominally spanning from 300–350 MHz (fundamental), 600–700 MHz (first overtone), 900–1050 MHz (second overtone), and 1200–1400 MHz (third overtone). Additional details on the IDT design and characterization can be found in [62–64] and in Supplement 1, Section 2.

2. ACOUSTO-OPTICAL SPECTROSCOPY

For measurement, the sample was mounted in a closed-cycle optical cryostat and kept at a temperature of $T = 5$ K. Single QDs are optically accessed perpendicular to the sample surface/planar cavity by a confocal microscope setup and resonantly excited by a continuous-wave frequency-tunable laser (linewidth $\delta\omega_{\text{laser}}/2\pi \leq 100$ kHz). The resonance fluorescence signal is collected in the same direction, and, in order to suppress reflected laser light, excitation and detection are cross-polarized with respect to each other. The second-order correlation function $G^{(2)}(\tau)$ was recorded with a fiber-based Hanbury Brown and Twiss setup equipped with Si avalanche single-photon detectors with a time resolution < 300 ps. Due to the limited time resolution, $G^{(2)}(\tau)$ was measured for a finite optical detuning with the laser being resonant with the turning point of the sinusoidal oscillation in the time domain [37]. To resolve the PSB, the collected light is spectrally filtered using a piezo-tunable Fabry–Perot etalon (free spectral range FSR = 60 GHz, finesse $\mathcal{F} = 263$) and detected by a single-photon avalanche photodetector (SPAD). The temperature

of the etalon was stabilized using a proportional-integral-derivative (PID) control loop.

The electrical signals used to excite SAWs were provided by two independent signal generators, which allowed for tuning of signal amplitude, frequency, and relative phase. To ensure a stable phase-lock between both SAWs, the two signal generators were referenced to a common 10 MHz oscillator. Both output signals were added using a standard rf-power combiner and applied to the IDT [34]. Power levels are chosen such that only the lowest PSBs are observed. For the data shown in Section 4.B of Supplement 1, two IDTs of the same design were used to generate counterpropagating SAWs.

3. THEORY

We describe the QD as a two-level system and obtain the resonance fluorescence by calculating the correlation function $G^{(1)}(t, t + \tau) = \langle \sigma_+(t)\sigma_-(t + \tau) \rangle$ [65]. The QD is coupled to a classical light field using rotating wave and dipole approximations, and the influence of the strain of the SAW waves is introduced as an energy modulation of the transition energy E_X via deformation potential coupling [66]. In addition, we consider the spontaneous recombination rate γ of the exciton state. The temporal evolution is calculated using the Master equation in Lindblad form, and the two-point correlation function G is found by quantum regression. In the experiment, the resonance fluorescence signal emitted by the QD is spectrally filtered by an etalon. Using a Lorentzian filter function $F(\omega)$ (spectral width $\frac{\delta\omega}{2\pi} = 0.41$ GHz for all simulations), we obtain for the scattered light intensity

$$I(\omega) = 2\text{Re} \frac{1}{T} \int_0^T \int_{-\infty}^{\infty} \int_0^{\infty} F^*(t - \tau) F(t - \tau - s) \times G(\tau, \tau + s) e^{i\omega s} ds d\tau dt,$$

where $F(t) = \exp(-\delta_e |t|)$ is the Fourier transform of $F(\omega)$, and T is the period of the SAW field. Further details are included in Supplement 1, Section 1.

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Supplemental document. See Supplement 1 for supporting content.

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