Optomechanical single photon frequency division multiplexing

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Today, digital data transmission relies on the parallel transmission of information employing wavelength (WDM) or frequency division multiplexing (FDM)¹,²,³. For quantum communication, such superchannels enable the creation of hyperentangled states. Nonlinear optics has been employed to harness this quantum resource by creating an entangled state of a quantum register formed by different teeth of a frequency comb⁴,⁵. However to date, frequency combs⁶–⁹ and derived quantum technologies⁴,⁵ are generated by photonic means, rigorously described by Maxwell’s classical theory of electromagnetism. Here we report on the realization of a tunable frequency comb of non-classical light, single photons. This comb is emitted from a single semiconductor quantum dot (QD) by coherent optical and surface acoustic waves (SAWs) acting on its exciton transition. The coherent optomechanical interaction mixes optical frequencies at \( \omega_{\text{opt}}/2\pi \approx 330 \text{ THz} \) and radio frequencies of phonons at \( \omega_{\text{SAW}}/2\pi \geq 1 \text{ GHz} \). Thus, a comb of sidebands is generated in the scattered light spectrum, precisely split by \( \omega_{\text{SAW}} \). We show that the nonlinear interaction between two mutually coherent SAW fields at \( \omega_{\text{SAW}}^{(1)} \) and \( \omega_{\text{SAW}}^{(2)} = 2\omega_{\text{SAW}}^{(1)} \) creates a highly stable and tunable comb. We observe frequency rungs of scattered photon intensities in the time domain for deliberately set detunings \( \Delta_{\text{SAW}}/2\pi \) as small as 50 \( \mu \text{Hz} \). This extremely stable quantum frequency comb may enable
practical quantum technologies, for instance quantum clock synchronization\textsuperscript{10,11} or frequency comb spectroscopy\textsuperscript{12,13} at the single photon level.

When a QD is dynamically strained by the oscillating mechanical field of a SAW, it experiences a time-modulation due to the deformation potential\textsuperscript{14}. Furthermore, the monochromatic nature and phase stability of the SAW preserve the outstanding coherent properties of the QD\textsuperscript{15}. Via the optomechanical interaction, discrete numbers of SAW phonons can be absorbed from or emitted to the SAW field giving rise to coherent inelastic Stokes- and anti-Stokes scattering, respectively. Because SAWs can be readily excited at gigahertz frequencies, which exceed the natural linewidth of the dot’s excitonic transitions\textsuperscript{16} this hybrid quantum dot optomechanical platform is placed in the resolved sideband regime\textsuperscript{17}. The latter is imperative for the frequency multiplexing of the single photons emitted from the dot in focus here. The experimental setup for coherent optomechanical spectroscopy of single QDs is illustrated in Figure 1a. 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 the excitation of SAWs of different frequencies over three frequency bands simply by applying the respective resonant radio frequency (rf) voltage. The exciton transition of a single QD is resonantly excited by a narrow band 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\textsuperscript{18}. Thus, in the resolved sideband regime, the narrow zero phonon line (ZPL) of the QD at energy $E_x$ splits into a series of phononic sidebands (PSBs) precisely spaced by $\omega_{\text{SAW}}$\textsuperscript{17,19} as shown schematically in Figure 1b. Figure 1c shows typical scattered photon spectra of a single QD strained by SAWs of three different $\omega_{\text{SAW}}$. The recorded spectra are plotted (dark lines) as a function of frequency $\omega_s = \omega - E_x / \hbar$, relative to the QD’s exciton transition. The recorded spectra show the expected creation of a comb of emission lines split precisely by the set $\omega_{\text{SAW}}/2\pi$. Moreover, all spectra are well reproduced by theoretical calculations (light lines). In our model, we consider the QD exciton as an optically driven two-level system, described by the Hamiltonian $H = (E_x + \Delta E(t))\sigma_z + \frac{\hbar}{2} \Omega (\sigma_+ + \sigma_-)$ with the time dependent shift of the transition energy $\Delta E(t) = \hbar \Delta \cdot \cos(\omega_{\text{SAW}} t)$. In these expressions, $\Omega$ denotes the Rabi frequency induced by the optical driving of the QD exciton and $\Delta$ the amplitude of the
SAW modulation. $\sigma_\pm = \sigma_x \pm i \sigma_y$ are the raising and lowering operators of the exciton two-level system with $\sigma_i$ denoting the Pauli spin matrices.

**Figure 1 | Frequency tunable PSBs.** a, Experimental implementation comprising a semiconductor QD embedded in a Bragg-type microcavity. The QDs exciton transition is resonantly excited by a laser tuned to its exciton transition at $E_X$. SAWs are generated by IDTs on the sample surface to dynamically strain the QD giving rise to inelastic Stokes and anti-Stokes scattering processes. b, Energy level diagram of the QD’s SAW-modulated exciton transition being resonantly excited by a laser. Due to stimulated SAW phonon absorption and emission, the ZPL breaks up in a series of PSBs split by the phonon energy $\hbar \omega_{SAW}$. c, Measured (dark lines) and calculated (light lines) scattered light spectra of a single QD dynamically strained by SAWs of three different SAW frequencies $\omega_{SAW}/2\pi$. The exciton decay $\gamma/2\pi$ and the SAW amplitudes $\Delta/2\pi$ used in the calculations are given in the plot. d, Measured PSB position as a
function of electrical frequency applied to the IDT. The grey lines mark the expected 
\( \omega_{rf} = m \cdot \omega_{SAW} \) dependence of the PSBs.

The multi-passband IDT\(^{20} \) enables elegant tuning of the frequency splitting simply by 
setting the frequency of the electrical signal. Figure 1d demonstrates the frequency 
tuning obtained in our approach. The splitting observed in the optical domain 
(horizontal axis) of the PSB faithfully follows the tuning of the electrical signal frequency 
\( \omega_{rf}/2\pi \) (vertical axis) of constant power \( P_{rf} = +14 \) dBm. In this range of frequencies, 
we cover passbands of the first, second and third overtone of the IDT (see Methods 
section), respectively. Across these frequency bands, the IDT generates SAWs and 
consequently PSB are observed at the programmed \( \omega_{rf} = m \cdot \omega_{SAW} \), with \( m = 0, \pm 1, \pm 2, \ldots \) The number of observed PSB decreases with increasing \( \omega_{rf} \) due to less 
efficient SAW generation and a reduction of the mean phonon number in the coherent 
SAW field.

![Figure 1](image1.png)

**Figure 1** | SAWs generated by IDT. The splitting of the PSB is shown for different 
SAW frequencies. The grey lines mark the expected \( \omega_{rf}/2\pi \) dependence of the PSBs.

![Figure 2](image2.png)

**Figure 2** | Nonlinear \( \omega_{SAW} - 2\omega_{SAW} \) phonon coupling and phase matching. a, 
Energy level diagram of the QD resonantly excited by a laser and two SAWs with 
\( \omega_{SAW}^{(2)}/2\pi = 2 \cdot \omega_{SAW}^{(1)}/2\pi \). Two series of PSBs are generated with every second of the 
\( \omega_{SAW}^{(1)} \) PSBs being in resonance with an \( \omega_{SAW}^{(2)} \) PSB. Arrows mark \( \omega_{SAW}^{(1)} \) (red) and \( \omega_{SAW}^{(2)} \) (blue) absorption (up arrow) and emission processes (down arrow). Dark and light 
arrows mark 2-phonon processes to reach the \( +1 \cdot \omega_{SAW}^{(1)} \) and \( -1 \cdot \omega_{SAW}^{(1)} \) PSBs,
respectively. b, Normalized scattered photon intensity as a function of relative optical frequency and relative phase $\phi$ between the two SAWs. The experimental data (upper panel) exhibits clear anti-phased oscillations of the $+1 \cdot \omega_{\text{SAW}}^{(1)}$ and $-1 \cdot \omega_{\text{SAW}}^{(1)}$ PSBs. This observed phase-matching is well reproduced by theory (lower panel) for the given exciton decay rate $\gamma/2\pi$ and SAW amplitudes $\Delta^{(1)}/2\pi$ and $\Delta^{(2)}/2\pi$.

Next, we demonstrate tunability of the PSB intensities harnessing nonlinear two-phonon couplings. This is achieved by combining two rf signals and connecting them to the IDT. We set $\omega_{\text{SAW}}^{(1)}/2\pi = 0.6775 \text{ GHz}$ and $\omega_{\text{SAW}}^{(2)}/2\pi = 1.3550 \text{ GHz}$. As depicted schematically in Figure 2a, we now generate two frequency combs of PSBs split by $\omega_{\text{SAW}}^{(1)}/2\pi$ and $\omega_{\text{SAW}}^{(2)}/2\pi = 2 \cdot \omega_{\text{SAW}}^{(1)}/2\pi$, respectively. Most importantly, only every second level of the $\omega_{\text{SAW}}^{(1)}$ comb is lined up with the levels of that of $\omega_{\text{SAW}}^{(2)}$. The nonlinear optomechanical interaction between the QD and the $\omega_{\text{SAW}}^{(1)}$ and $\omega_{\text{SAW}}^{(2)}$ SAW phonons renders sum and difference SAW frequency processes possible. Figure 2a illustrates processes leading to emission of photons in the $+1 \cdot \omega_{\text{SAW}}^{(1)}$ PSB. For a single $\omega_{\text{SAW}}^{(1)}$ comb, absorption of a single $\omega_{\text{SAW}}^{(1)}$ SAW phonon (red up arrow) is the only possible first order process leading to this emission and two-phonon processes would not give contributions to this line. However, when $\omega_{\text{SAW}}^{(1)}$ and $\omega_{\text{SAW}}^{(2)}$ SAW phonons are interacting with the QD, the $+1 \cdot \omega_{\text{SAW}}^{(1)}$ PSB becomes accessible by the combination of absorption of a single $\omega_{\text{SAW}}^{(2)}$ SAW phonon (blue up arrow) and emission of a single $\omega_{\text{SAW}}^{(1)}$ SAW phonon (red down arrow). Conversely, emission of photons in the $-1 \cdot \omega_{\text{SAW}}^{(1)}$ PSB arises from the opposite combination of processes, namely emission of one $\omega_{\text{SAW}}^{(2)}$ SAW phonon (light blue down arrow) and absorption of one $\omega_{\text{SAW}}^{(1)}$ SAW phonon (light red up arrow). Analogous to mixing of optical waves in nonlinear optics, also two-phonon processes have to obey underlying phase-matching conditions. In Figure 2b, we plot the intensity of the scattered photons in false-colour representation as a function of relative phase $\phi$ between the $\omega_{\text{SAW}}^{(1)}$ and $\omega_{\text{SAW}}^{(2)}$ SAWs (horizontal axis) and the relative optical frequency. Importantly, $\phi$ can be simply tuned via the relative phase of the mutually coherent electrical signals exciting the two SAWs. In this proof-of-principle experiment, the rf power levels are set such that the $\pm 1 \cdot \omega_{\text{SAW}}^{(1)}$ PSBs dominate the spectra. The upper panel shows the experimental data, which is compared to
calculations using our theoretical model extended to two SAWs in the lower panel. As \( \phi \) is tuned, the \( +1 \cdot \omega^{(1)}_{\text{SAW}} \) and \( -1 \cdot \omega^{(1)}_{\text{SAW}} \) PSBs undergo clear \( 2\pi \)-periodic but anti-phased intensity oscillations. This observation directly reflects the aforementioned tunable phase-matching condition. For \( \phi = n \cdot \pi, \ n = 0, 1, 2, ... \) the intensity is distributed equally between positive and negative sidebands resulting in a symmetric spectrum. In contrast, for \( \phi = (2n + \frac{3}{2}) \cdot \pi \), the positive PSBs are preferentially generated and respectively for \( \phi = (2n + \frac{1}{2}) \cdot \pi \) the negative PSBs. Remarkably, experimental observations (upper panel) and prediction of our analytical theory (lower panel) are in excellent agreement using SAW amplitudes \( \Delta^{(1)}/2\pi \) and \( \Delta^{(2)}/2\pi \), and a spontaneous emission rate \( \gamma/2\pi \) as given in the plot. Thus, our experimental results are fully supported by theory and underpin, that nonlinear SAW mixing is a key requirement to continuously tune the intensities of the time-averaged PSB spectrum. We note that this tunability cannot be achieved by simply combining two counterpropagating SAWs of identical frequency. In this case, discussed in SFig 1 of the Supplementary Material, a standing wave is generated and tuning \( \phi \) just moves the position of the nodes across the QD. As expected, we observe the same intensities of \( |m| \cdot \omega^{(1)}_{\text{SAW}} \) PSBs with positive or negative \( m \) for all \( \phi \), while lacking the possibility to deliberately suppress or enhance one or the other.
Figure 3 | Frequency ring generation and stability analysis. 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 $T_\phi = 2\pi/\Delta_{\text{SAW}} \approx 5.55 \, \text{hours}$. c, Colour-coded intensity of the $+1\cdot \omega_{\text{SAW}}^{(1)}$ 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 $T_\phi$.

Finally, we study the frequency stability of our hybrid nonlinear optomechanical scheme. To this end we deliberately introduce a small, yet finite detuning $\Delta_{\text{SAW}}$ to the
SAW such that \( \omega_{\text{SAW}}^{(2)} = 2\omega_{\text{SAW}}^{(1)} + \Delta_{\text{SAW}} \). The resulting PSB alignment is shown schematically in Figure 3a. As depicted, introducing \( \Delta_{\text{SAW}} \) is equivalent to a time-varying relative phase \( \phi(t) \) of the \( \omega_{\text{SAW}}^{(2)} = 2\omega_{\text{SAW}}^{(1)} \) nonlinear mixing scheme. 

\[
\omega_{\text{SAW}}^{(1)}/2\pi = 0.65 \text{ GHz} \quad \text{and} \quad \omega_{\text{SAW}}^{(2)}/2\pi = 1.30 \text{ GHz}
\]

are set with the precision of state-of-the-art rf electronics. Thus, the accessible sub-hertz frequency range of \( \Delta_{\text{SAW}} \) can be significantly smaller than the linewidth of the laser (\( \delta \omega_{\text{laser}}/2\pi \approx 300 \) kHz) and that of the exciton transition (\( \delta \omega_{\text{x}}/2\pi \approx 1 \) GHz). As proof-of-principle, we set \( \Delta_{\text{SAW}}/2\pi = 50 \) µHz, corresponding to a time-dependent phase \( \phi(t) = \Delta_{\text{SAW}} t \), and detect the scattered photon spectrum as a function of time over one full cycle of this oscillation. We plot the recorded intensity in false-colour representation as a function of time \( t \) (horizontal axis) and the relative optical frequency \( \omega_s \) (vertical axis). The experimental data exhibits a clear oscillation of the intensities of the PSB and the ZPL with a period of precisely \( T_{\phi} = 2\pi/\Delta_{\text{SAW}} \approx 5.55 \) hours. Remarkably, this demonstrated frequency rung is only \( 7.6 \cdot 10^{-14} \omega_{\text{SAW}}^{(1)} \) and \( 1.6 \cdot 10^{-19} \frac{E_{\text{x}}}{\hbar} \), underlining the outstanding stability of our hybrid nonlinear optomechanical scheme. Again, the frequency rung of the sideband intensity is fully tunable. This is studied in detail in Figure 3c. Here the intensity of the \(+1\cdot\omega_{\text{SAW}}^{(1)}\) PSB is plotted in false-colour representation as a function of time (horizontal axis) and the SAW detuning \( \Delta_{\text{SAW}}/2\pi \) (vertical axis). As the detuning is tuned \(-5 \) Hz \( \leq \Delta_{\text{SAW}}/2\pi \leq +5 \) Hz the period of the oscillation detected in the time-domain faithfully changes accordingly, creating the inversion symmetric pattern in Figure 3c. This pattern can be seen even more clearly in Figure 3d, which is a zoom-in on the regime of small detunings \(-0.5 \) Hz \( \leq \Delta_{\text{SAW}}/2\pi \leq +0.5 \) Hz of the data shown in Figure 3c.

In conclusion, we demonstrated frequency domain multiplexing of single photons emitted from a semiconductor QD. Our scheme relies on SAWs, one of the very few phononic technologies of industrial relevance\(^{23}\). The employed nonlinear optomechanical paradigm enables fully fledged coherent frequency transduction over 19 orders of magnitude, proven by the observation of a 50 µHz intensity oscillation of the 330 THz photons emitted by the QD. While our particular implementation is based on epitaxial QDs made of III-V compound semiconductors, defect centers in diamond\(^{24}\), silicon carbide\(^{25}\) or two-dimensional materials\(^{26}\) have already been proven to be well-suited to be interfaced with SAWs. Moreover, the driving optomechanical
interaction can be drastically enhanced by embedding the quantum emitter in phononic or optomechanical resonators\textsuperscript{25} building on demonstrated monolithic integration on GaAs\textsuperscript{27}. Alternatively, the heterointegration of the semiconductor on LiNbO\textsubscript{3}\textsuperscript{28,29} harnesses the strong piezomechanical\textsuperscript{30} and nonlinear optical properties\textsuperscript{31} of the host substrate and the large optomechanical coupling of semiconductor\textsuperscript{23}.

Methods

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. Multiharmonic IDTs\textsuperscript{20} (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 $\lambda = 9.91\mu$m, or, in order to realize frequency chirped transducers\textsuperscript{20}, with a wavelength linearly varying from $\lambda_0 = 8.94\mu$m to $\lambda_1 = 10.38\mu$m along the length ($L_{\text{IDT}} = 400\mu$m) of the IDT. The first design allows SAW generation at a fundamental frequency of about 338 MHz and at overtones of 677 MHz, 1015 MHz 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).

Acousto-optical spectroscopy

For measurement the sample was mounted in a 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 cw 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 collected light is spectrally filtered using a piezo-tunable Fabry Perot etalon (free spectral range $FSR = 60$ GHz, finesse $F = 263$) 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, that 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 signal were added using a standard rf-power combiner and applied to the IDT\textsuperscript{21}. For the data shown in the Supplementary Information, two IDTs of the same design were used to generate counterpropagating SAWs.

For the measurements presented in Figure 3(c, d) the emission of the $+1 \cdot \omega_{\text{SAW}}^{(1)}$ PSB was filtered by the etalon and the time evolution of the intensity was recorded in real time over a period of 20 seconds.

**Theory**

We describe the QD as a two-level system and obtain the resonance fluorescence\textsuperscript{32} by calculating the correlation function $G(t, t + \tau) = \langle \sigma_+(t) \sigma_-(t + \tau) \rangle$. 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 $E_{\chi}$. In addition, we take into account the spontaneous recombination rate $\gamma$ 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 $\delta_{\text{e}}/2\pi = 0.41$GHz for all simulations) we obtain for the scattered light intensity $I(\omega) = 2 \text{Re} \left( \frac{1}{T} \int_{0}^{T} \int_{-\infty}^{\infty} \int_{0}^{\infty} F^*(t - \tau) F(t - \tau - s) G(\tau, \tau + s) e^{i\omega s} \, ds \, d\tau \, dt \right)$, where $F(t) = \exp(-\delta_{\text{e}}|t|)$ is the Fourier transform of $F$ and $T$ is the period of the SAW field.

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Author contributions

H.J.K., D.W. and P.M. instigated the project. M.W. designed and fabricated devices, built experimental setup and performed experiments. M.W., D.W., M.N. and H.J.K. analysed the experimental data. D.W., P.M. and T.K. developed the theoretical model. K.M. and J.J.F. performed crystal growth and pre-characterization of the semiconductor heterostructure. H.J.K., D.W., P.M. and M.W. wrote the manuscript with input from all authors.
Supplementary Figure 1 | Standing wave modulation. a, Standing wave excitation by counterpropagating SAWs. Changing the relative phase $\phi$ between the two SAWs shift the standing wave pattern across the position of the QD. For the QD being not at the position of a node, a symmetric modulation occurs. b, Normalized scattered photon intensity as a function of relative optical frequency (horizontal axis) and relative phase $\phi$ (in degrees) between the two SAWs (vertical axis) showing the expected symmetric modulation. c, Extracted intensities of the PSB (left panel) and the ZPL (right panel). In the left panel, red and blue lines show data of the negative and positive PSBs, respectively.