Stimulated Raman adiabatic passage in optomechanics

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Stimulated Raman adiabatic passage (STIRAP) describes adiabatic population transfer between two states coherently coupled via a mediating state that remains unoccupied. This renders STIRAP robust against loss in the mediating state, leading to profound applications in atomic- and molecular-beam research \([1,2]\), trapped-ion physics \([3]\), superconducting circuits \([4]\), other solid-state systems \([5,6]\), optics \([7]\), in entanglement generation \([8]\) and qubit operations \([9]\). STIRAP in optomechanics has been considered for optical frequency conversion where a mechanical mode provides the mediating state \([10,12]\). Given the advances of optomechanical devices with exceptionally high mechanical-quality factors \([13\textsuperscript{-}15]\), STIRAP between mechanical modes has the prospect of generating macroscopic quantum superposition and of supporting quantum information protocols \([16]\). An optical cavity mode can mediate the coupling between mechanical modes, without detrimental effects of optical losses. We demonstrate STIRAP between two mechanical modes of a phononic membrane-in-the-middle system with an efficiency of 86% and immune against photon loss through the mediating optical cavity.

The transfer of population between states of mechanical resonators and/or optical modes at the same or at disparate frequencies in optomechanical systems has been addressed in a variety of ways including topological operations \([17]\), direct coupling through a waveguide \([18]\), coupling through two optical modes \([19]\), coupling through internal resonance \([20]\) and optical-microwave photon conversion via a mechanical mode \([21]\). Furthermore, optomechanical systems with multiple membranes have attracted attention recently \([22,23]\). The concept of STIRAP has been theoretically considered in the context of optomechanics \([10,11,24]\). Here we present the first optomechanical implementation of STIRAP with its distinct advantage of being insensitive to losses in the mediating state.

STIRAP concerns a Λ-type level scheme as shown in Fig. 1a. In the triply rotating frame at frequencies \(\omega_1 = E_1/h\) for states \(\psi_i, i = 1, 2, 3\), the Hamiltonian is:

\[
\hat{H}(t) = \frac{\hbar}{2} \begin{pmatrix}
0 & \Omega_{12}(t) & 0 \\
\Omega_{12}(t) & 0 & \Omega_{23}(t) \\
0 & \Omega_{23}(t) & 0
\end{pmatrix},
\]

with \(\Omega_{12}\) and \(\Omega_{23}\) the Rabi frequencies resulting from two driving fields at frequencies \((E2-E1)/h\) and \((E2-E3)/h\). One of the three instantaneous eigenstates has eigenvalue 0 and does only include states \(\psi_1\) and \(\psi_3\):

\[
\Phi_0(t) = \cos \theta(t) \psi_1 - \sin \theta(t) \psi_3,
\]

with \(\tan \theta(t) = \Omega_{12}(t)/\Omega_{23}(t)\). STIRAP is based on the adiabatic following of \(\Phi_0(t)\) by slowly varying \(\theta(t)\) from \(\theta(-\infty) = 0\) to \(\theta(\infty) = \pi/2\). Thus, the system can be adiabatically transferred from \(\psi_1\) to \(\psi_3\) without occupying state \(\psi_2\). Fig. 1b shows a typical time evolution of \(\Omega_{12}(t)\) and \(\Omega_{23}(t)\) and Fig. 1c shows the energy eigenvalues corresponding to the three eigenstates \(\Phi_+(t), \Phi_0(t), \) and \(\Phi_-(t)\). The remarkable aspect of STIRAP is that one starts with driving two unoccupied states 2 and 3, before coupling 1 and 2, and also ends with driving two unoccupied states 1 and 2. This driving pulse sequence, often described as “counter-intuitive”, together with the adiabaticity condition \(\sqrt{\Omega_{12}(t)^2 + \Omega_{23}(t)^2} \gg \theta\) prevents the lossy mediating state from being occupied throughout the transfer process.

Figure 1. STIRAP scheme in optomechanics. a, The general three-level setting. b, The pulse sequence for the driving fields. c, The resulting energy eigenvalues for the instantaneous eigenstates \(\Phi_+(t), \Phi_0(t), \) and \(\Phi_-(t)\). STIRAP explores the properties of \(\Phi_0(t)\) given in Eq. (2). d, The optomechanical implementation contains a cavity mode at frequency \(\omega_{\text{cav}}\), two driving fields \(\omega_1, \omega_2\) on the driving fields, red bars corresponding to the sidebands on \(\omega_{1,2}\) and blue bars corresponding to the sidebands on \(\omega_{1,2}\). Two sidebands match \(\omega_{\text{cav}}\). In the case of \(\Phi_0(t)\) the states \(\psi_1\) and \(\psi_3\) are out of phase leading to destructive interference of the sidebands that overlap with \(\omega_{\text{cav}}\).
The Hamiltonian in Eq. (1) can be realized in multimode optomechanics where states 1 and 3 are mechanical excitations with frequencies ω₁ and ω₂ and state 2 is an optical cavity mode at ω_{cav}, see Fig. 1d. Two optical fields at ω_{i,a} = ω_{cav} - ω_i for i = 1, 2 are introduced in order to create the beamsplitter interaction \( \hat{a} \hat{b}_i^\dagger + \hat{a}^\dagger \hat{b}_i \) that couples the mechanical modes to the cavity mode, where \( \hat{a}(\hat{a}^\dagger) \) and \( \hat{b}_i(\hat{b}_i^\dagger) \) are the photon and phonon annihilation (creation) operators.

The optical mode can be represented by the operator \( \hat{a} = \bar{a} + \delta \hat{a} \), where \( \bar{a} \) is the average coherent amplitude due to the optical fields sent into the cavity and \( \delta \hat{a} \) is the fluctuating term. Each mechanical mode produces two sidebands with frequencies \( \omega_{cav} \pm \omega_i \) that acquire much larger amplitudes than the other sidebands. Taking into account only those two sidebands and including mechanical and optical loss rates, \( \Gamma_i \) and \( \kappa \), the time evolution of the state vector \( \psi(t) = (\hat{b}_1(t), \delta \hat{a}(t), \hat{b}_2(t))^T \) is given by

\[
i \frac{d\psi(t)}{dt} = \begin{pmatrix}
-\frac{i \Gamma_1}{2} & g_1(t) & 0 \\
g_1^*(t) & -\frac{i \Gamma_2}{2} & g_2(t) \\
0 & g_2^*(t) & -\frac{i \Gamma_2}{2}
\end{pmatrix} \psi(t), \tag{3}
\]

in the rotating wave approximation and the linearized approximation with \( g_i \) being the optomechanical multiphoton coupling for mechanical modes \( i = 1, 2 \), see Figure 3. Experimental optomechanical STIRAP. a, Left scale: phonon population as a function of time, red dots correspond to averaged measurements for mode 1, blue dots for mode 2, both divided by the phonon population of mode 1 in the beginning of the transfer. Light red and light blue regions represent the phonon populations with statistical uncertainties obtained from simulations without free fit parameters. Right scale: multiphoton optomechanical couplings \( g_1(t) \) red line, \( g_2(t) \) blue line, calculated from measured pulse intensities. The driving field pulses have a nearly Gaussian profile with the standard deviation parameter \( \sigma \) and separation \( \Delta t \), but their beginning and ending are smoothly truncated to zero. Black stars correspond to the phonon populations used to calculate transfer efficiency (5% of the peak voltage sent to AOM). b, Measured phases of mode 1 (red) and mode 2 (blue) in the rotating frame. There are four time domains with distinct behavior of phases: in domain 1 \( g_1(t) = 0 \) and the phase of mode 1 is defined by the excitation used to drive it, while mode 2 is in its thermal motion, thus the difference between the phases is random; in domain 2 STIRAP starts and the phase of mode 2 adjusts itself until the sidebands at \( \omega_{cav} \) become \( \pi \) out of phase; in domain 3 the phase of the locked mechanical modes changes due to the optomechanically induced frequency shift from field \( \omega_{cav} \) (unmatched sidebands); in domain 4 the read-out signal of mode 2 becomes much less than the read-out noise.

Figure 2. Optomechanical setup. a, A transparent dielectric membrane patterned with a phononic crystal is placed in the middle of a high-finesse optical cavity. Shift of the membrane along the axis of the cavity changes the cavity resonance frequency, causing coupling of light in the cavity to vibrational modes of the membrane. b, Simulated displacement of a mechanical mode of the defect of the phononic crystal. The mode is localized as its frequency lies in the band gap (mode 1, initially excited). c, Simulated displacement of the 3.3 drumhead mode of the full membrane (mode 2). This second mode was selected because it has an appropriate mechanical frequency and quality factor and has a maximum amplitude at the center. This allows both modes to be aligned for optimal coupling to the same cavity mode.
Figure 4. **Transfer efficiencies under different parameters of the optical pulses.** a, The efficiency as a function of the ratio of the separation between the peaks of the pulses $\Delta t$ and the Gaussian parameter $\sigma$. Positive values of $\Delta t$ correspond to the field at $\omega_{L2}$ being sent before the field at $\omega_{L1}$. b, Maximal transfer efficiencies as a function of $\sigma$. In a) and b) the red circles show measured efficiencies in individual runs, black circles are the simulated efficiencies, and the black lines are guides for the eyes. c and d show the experimental (c) and predicted (d) transfer efficiency as a function of the Gaussian parameter $\sigma$ and separation $\Delta t$. The horizontal row of dots in c) and d) correspond to the data shown in a), while the vertical row of dots correspond to the data shown in b). The transfer process for the parameters corresponding to the open circle in c) and d) is shown in Fig. 3.

Supplementary Information. Equation (3) is identical to Eq. (1) in the absence of losses and with the Rabi frequencies $\Omega_{12}$ and $\Omega_{23}$ corresponding to $2g_1$ and $2g_2$. STIRAP is realized by adiabatically transitioning from $\psi_1$ to $\psi_3$ by changing from $g_2(-\infty)/g_1(-\infty) \to \infty$ to $g_2(\infty)/g_1(\infty) \to 0$. This is satisfied for two Gaussian driving pulses that are temporally shifted (Fig. 1b). Note that optical cooling of the mechanical modes is absent in the $\Phi_0(t)$ state, despite the presence of the red-detuned optical fields, due to destructive interference of the side-bands at $\omega_{cav}$.

Experimentally we demonstrate the state transfer in the membrane-in-the-middle (MIM) configuration [25], where a membrane with low optical absorption is placed in the center of a high-finesse optical cavity with $\kappa/2\pi = 54$ kHz (including membrane), see Fig. 2. A displacement of the membrane along the optical axis leads to a shift in the optical cavity transmission described by the interaction Hamiltonian $\hat{H}_{int} = -\hbar g_0 \hat{a}^\dagger \hat{a} \hat{b} + \text{h.c.}$, where $g_0$ is the single photon optomechanical coupling. The membrane is a highly stressed 25 nm thick SiN film lithographically patterned with a phononic crystal with a defect in its center suspended on a silicon frame [14]. There are two types of mechanical modes: whole membrane drumhead modes and modes localized near the phononic crystal defect with frequencies in the phononic crystal bandgap. Vibrational energy of the drumhead modes is mainly lost in the bending regions where the membrane is connected to the frame [14, 26]. The modes localized near the defect possess enhanced quality factors by 1-2 orders of magnitude compared to the drumhead modes [14]. STIRAP is implemented using the fundamental mode of the defect with frequency $\omega_{1}/2\pi = 1.25$ MHz and quality factor $Q = 1.3 \cdot 10^7$ (mode 1, Fig. 2b), and the 3.3 drumhead mode with $\omega_{2}/2\pi = 0.22$ MHz and $Q = 1.2 \cdot 10^6$ (mode 2, Fig. 2c). The modes are coupled to the optical cavity with single-photon couplings of $g_{01}/2\pi = 1.5 \pm 0.1$ Hz and $g_{02}/2\pi = 1.0 \pm 0.1$ Hz. STIRAP is very sensitive to the double-photon detuning $\Delta_{2ph} = \omega_{L1} + \omega_1 - (\omega_{L2} + \omega_2)$, therefore the two optical fields are created by amplitude modulation (see Supplementary Information) of a single 1064 nm laser field. The membrane is in a vacuum chamber with pressure below...
Next we investigate the dependence of the transfer efficiency on the parameters of the process. First the time shift between the optical pulses $\Delta t$ is varied, see Fig. 4. The adiabaticity condition is satisfied increasingly better with longer pulses such that for pulses with $\sigma = 100$ ms only 2\% of the initial phonon population in mode 1 is lost through the population and decay of the optical mode. Nevertheless the efficiency starts to decrease for $\sigma \gtrsim 25$ ms due to the mechanical decay of the modes and cooling of the mode $i$ by the field $j$, $i \neq j$, setting the upper bound on the transfer efficiency. A comparison of the experimental and the simulated efficiencies is depicted in Fig. 4 c) and d) respectively when $\Delta t$ and $\sigma$ are varied.

A signature of STIRAP [16] is strong sensitivity of the transfer efficiency to the two-photon detuning $\Delta_{2\text{ph}} = (\omega_{L1} + \omega_1) - (\omega_{L2} + \omega_2)$ given $\Delta_{1\text{ph}} = 0$, compared to the sensitivity to the single-photon detuning $\Delta_{1\text{ph}} = \omega_{\text{cav}} - (\omega_{L1} + \omega_1)$ given $\Delta_{2\text{ph}} = 0$, see Fig. 5. The frequency scale for the two-photon detuning is set by the duration of the transfer process: $\Delta_{2\text{ph}} \sim \pi/T_{\text{transfer}}$, implying that the sidebands at $\omega_{\text{cav}}$ accumulate a phase difference of $\pi$ during the transfer and consequently no longer interfere destructively. The simulated curve for transfer efficiency vs. $\Delta_{2\text{ph}}$ is shifted by $\sim 2$ Hz due to the corrections caused by the unmatched sidebands as confirmed by experimental data. The frequency scale for $\Delta_{1\text{ph}}$ is set by the optical cavity linewidth $\kappa$: deviations of $\Delta_{1\text{ph}}$ from zero lead to decreased amplitudes of the matched sidebands, having an effect equivalent to a decrease of the intensity of fields 1 and 2 and causing a violation of the adiabaticity condition.

The highest transfer efficiency we observe in our system is $0.86 \pm 0.03$ which is limited by the intrinsic mechanical decay of the mechanical modes and by the cross-talk between mechanical mode at $\omega_i$ and field $\omega_{Lj}$, $i \neq j$. In general, the STIRAP scheme in optomechanics can result in transfer efficiencies close to 1 provided the difference

$10^{-6}$ mbar at room temperature.

STIRAP with parameters tuned for optimal efficiency is shown in Fig. 3b. The measurement of a typical transfer process has the following sequence: mode 1 is excited to an amplitude much higher than its thermal occupation. During its free decay the two optical pulses are sent and the excitation of mode 1 is transferred to mode 2. The transfer starts with the beginning of pulse 1 (red) and finishes with the end of pulse 2 (blue), these moments are denoted by dashed vertical lines. The transfer efficiency is calculated as the ratio of the phonon population in mechanical mode 2 at the end of the transfer to the phonon population in mechanical mode 1 at the beginning of the transfer (black stars). A theoretical model without free fit parameters was developed in the classical limit to simulate the transfer process taking into account the corrections due to the other sidebands and the measured profiles of the light pulses (see Supplementary Information).

In our realization of STIRAP using coherent state populations, i.e. in the classical regime, the phases of the mechanical modes during the transfer can be continuously monitored, see Fig. 3b. The simplified model based on Eq. (3) predicts that the phases of the two mechanical modes become locked (in the triply rotating frame) such that the resulting sidebands on the two driving fields at $\omega_{\text{cav}}$ are $\pi$ out of phase resulting in destructive interference in the cavity mode. We observe the phase locking during the onset of STIRAP but record an overall phase shift during the transfer. This shift is a consequence of the unmatched sidebands (Fig. 1d) that are not accounted for in the simplified model.

Figure 5: Transfer efficiencies for non-zero single- and two-photon detuning. a. The efficiency as a function of the two-photon detuning $\Delta_{2\text{ph}}$ with zero single-photon detuning. The vertical line is the horizontal position of the peak of a Gaussian fit to the measured data. b. The efficiency as a function of the single-photon detuning $\Delta_{1\text{ph}}$ with zero two-photon detuning. In a) and b) the red circles are measured efficiencies in individual runs, and the shaded regions are simulated efficiencies with statistical uncertainties.
between the frequencies of the mechanical modes is much larger than the cavity linewidth, while being in the weak coupling regime, with peak multiphoton optomechanical couplings being much larger than the inverse of the transfer duration, and with slow enough mechanical decay:

$$|\Omega_1 - \Omega_2| \gg \kappa \gg \max g_i(t) \gg 2\pi/T_{\text{transfer}} \gg \Gamma_i.$$  

(4)

This sets stringent requirements on the experimental realization of STIRAP in optomechanics. Other experimental challenges are the accurate control of 1- and 2-photon detunings, circumventing detrimental effects of the unmatched sidebands, and proving stable subwavelength positioning of the membrane to maximize the coupling strength.

This letter presents the first optomechanical implementation of STIRAP with a maximum transfer efficiency of 86 ± 3%. It is expected that STIRAP in optomechanics will become as important as STIRAP in other branches of physics.

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[1] U. Gaubatz, P. Rudecki, M. Becker, S. Schiemann, M. Külz, and K. Bergmann, Chemical Physics Letters 149, 463 (1988).
[2] P. Pillet, Physical Review A - Atomic, Molecular, and Optical Physics 48, 1 (1993).
[3] J. L. Sørensen, D. Møller, T. Iversen, J. B. Thomsen, F. Jensen, P. Staanum, D. Voigt, and M. Drewsen, New Journal of Physics 8 (2006), 10.1088/1367-2630/8/11/261.
[4] K. S. Kumar, A. Vepsäläinen, S. Danilin, and G. S. Paraoanu, Nature Communications 7, 1 (2016).
[5] H. Goto and K. Ichimura, Physical Review A - Atomic, Molecular, and Optical Physics 75, 1 (2007).
[6] D. A. Golter and H. Wang, Physical Review Letters 112, 1 (2014).
[7] S. Longhi, Laser and Photonics Reviews 3, 243 (2009).
[8] J. Simon, H. Tanji, S. Ghosh, and V. Vuletic, Nature Physics 3, 765 (2007).
[9] K. Toyoda, K. Uchida, A. Noguchi, S. Haze, and S. Urabe, Physical Review A - Atomic, Molecular, and Optical Physics 87, 1 (2013).
[10] Y. D. Wang and A. A. Clerk, Physical Review Letters 108, 1 (2012).
[11] L. Tian, Physical Review Letters 108, 1 (2012).
[12] C. Dong, V. Fiore, M. C. Kurzyk, and H. Wang, Science 338, 1609 (2012).
[13] R. A. Norte, J. P. Moura, and S. Gröblacher, Physical Review Letters 116, 1 (2016) arXiv:1511.06235.
[14] Y. Tsaturyan, A. Barg, E. S. Polzik, and A. Schiessl, Nature Nanotechnology 12, 776 (2017).
[15] S. M. Meenehan, J. D. Cohen, G. S. MacCabe, F. Marsili, M. D. Shaw, and O. Painter, Physical Review X 5, 1 (2015) arXiv:1503.05135.
[16] N. V. Vitanov, A. A. Rangelov, B. W. Shore, and K. Bergmann, Reviews of Modern Physics 89, 1 (2017).
[17] H. Xu, D. Mason, L. Jiang, and J. G. Harris, Nature 537, 80 (2016).
[18] K. Fang, M. H. Matheny, X. Luan, and O. Painter, Nature Photonics 10, 489 (2016).
[19] M. J. Weaver, F. Buters, F. Luna, H. Eerkens, K. Heeck, S. De Man, and D. Bouwmeester, Nature Communications 8, 1 (2017).
[20] C. Chen, D. H. Zanette, D. A. Czaplewski, S. Shaw, and D. López, Nature Communications 8, 1 (2017) arXiv:1612.00490.
[21] R. W. Andrews, R. W. Peterson, T. P. Purdy, K. Cicak, R. W. Simmonds, C. A. Regal, and K. W. Lehnert, Nature 10, 321 (2014) arXiv:1310.5276.
[22] P. Piergentili, L. Catalini, M. Bawaj, S. Zippilli, N. Malossi, R. Natali, D. Vitali, and G. D. Giuseppe, New Journal of Physics 20 (2018), 10.1088/1367-2630/aa85f arXiv:1805.09669.
[23] C. Gätter, J. P. Moura, W. Haaxman, R. A. Norte, and S. Gröblacher, Nano Letters 18, 7171 (2018) arXiv:1809.05372.
[24] D. Garg, A. K. Chauhan, and A. Biswas, Physical Review A 96 (2017), 10.1103/PhysRevA.96.023837 arXiv:1706.05869.
[25] J. D. Thompson, B. M. Zwickl, A. M. Jayich, F. Marquardt, S. M. Girvin, and J. G. Harris, Nature 452, 72 (2008).
[26] S. Schmidt, K. D. Jensen, K. H. Nielsen, and A. Boisen, Physical Review B - Condensed Matter and Materials Physics 84, 1 (2011).
[27] R. W. Drever, J. L. Hall, F. V. Kowalski, J. Hough, G. M. Ford, A. J. Munley, and H. Ward, Applied Physics B Photophysics and Laser Chemistry 31, 97 (1983).
[28] A. M. Jayich, J. C. Sankey, B. M. Zwickl, C. Yang, J. D. Thompson, S. M. Girvin, A. A. Clerk, F. Marquardt, and J. G. Harris, New Journal of Physics 10 (2008), 10.1088/1367-2630/10/9/095008 arXiv:arXiv:0805.3723v1.
[29] M. Aspelmeyer, T. J. Kippenberg, and F. Marquardt, Reviews of Modern Physics 86, 1391 (2014).
Stimulated Raman adiabatic passage in optomechanics: Supplementary Information

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I. OPTICAL SETUP

The motion of the membrane is read out via the light fields generated by a probe laser at $\omega_{\text{probe}}$ (10 µW) locked to the transmission resonance frequency of the optical cavity via the Pound-Drever-Hall technique (PDH) \textsuperscript{24}. In order to measure the instantaneous oscillation displacement of a mechanical mode $\alpha$, the reflection signal is demodulated at $\omega + \omega_{\text{ EOS}}$, where $\omega_{\text{EOS}} = 10$ MHz is the detuning frequency of the sidebands in the PDH locking scheme. The driving light fields generated by the pump laser at $\omega_{\text{pump}}$ is locked to the probe laser by a phase-locked loop, see Fig. S1. The light fields from the two lasers are measured by a fast photodetector and the beating signal is mixed with a reference microwave signal, supplied by an RF generator. The resulting signal is sent to a proportional-integral-differential controller (PID) which adjusts the frequency of the pump laser. The difference between the lasers frequencies is kept at $\omega_{\text{probe}} - \omega_{\text{pump}} = 2\text{FSR} + \Delta \sim 3$ GHz because the dispersion curves of membrane-in-the-middle systems are parallel for all odd and all even resonances \textsuperscript{25}. This ensures a well-defined cavity resonance detuning of the pump laser in spite of drifts in the membrane position along the optical axis ($\sim 10$ nm/hour). The pump light fields have orthogonal polarization to the probe light fields in order to minimize interference of both fields at the reflection photodetector. To excite a membrane mechanical mode, an AC voltage ($\sim 10$ mV) at its mechanical frequency is applied to a needle placed close to the defect of the membrane ($\sim 0.5$ mm).

II. MEMBRANE POSITIONING

The dispersion curves of a membrane-in-the-middle system are parallel for the curves separated by 2FSR provided the membrane is positioned exactly in the middle of the cavity. For a small displacement $z$ of the membrane from the center, the free-spectral range changes as $2\text{FSR} - 2\text{FSR}_{\text{middle}} \propto 2\text{FSR}_{\text{middle}} \frac{z}{L} \sin \frac{\pi z}{L}$, where $L$ is the length of the cavity and $\lambda$ is the wavelength. The membrane holder is mounted on a tip-tilt stage with 3 vacuum compatible motors (1 step $\sim 20$ nm). To minimize the influence of the membrane drift along the optical axis, the membrane was moved towards the middle of the cavity to $z \sim 30$ µm by measuring 2FSR as a function of $z$, which provides an estimate for the direction and amplitude of the movement. To further minimize the influence of membrane drifts, we use a piezo element to bring the membrane to the position where 2FSR has a local maximum as a function $z$. This position coincides with the maximum optomechanical coupling strength. As a result an average drift of the membrane during a measurement run of 1 hour causes an acceptable change of $2\text{FSR} \sim 5$ kHz. When the actual experiment is running, we use the piezo to bring the membrane back to the position of maximum 2FSR every hour.

III. MEMBRANE FABRICATION

We begin the fabrication process of the devices with a commercially supplied 525 µm thick silicon wafer coated on both sides with 25 nm of LPCVD high-stress silicon nitride. We pattern the phononic crystal structure into the nitride on one side through the use of standard photolithography. During a second photolithography step, we use an IR contact aligner to pattern a square hole in photoresist on the opposite side of the chip. A subsequent Bosch etch step etches through the exposed silicon underneath the phononic crystal. After cleaning the chip in piranha solution, we release the phononic crystal membrane by wet etching the remaining 100 µm of silicon using KOH at 80°C. We perform a final clean by submerging the chip in HF for 1 minute and then we extract it out of IPA and allow it to dry through evaporation.

IV. THEORY

Here we derive Eq. (3) of the main text. We start from the optomechanical equations of motion \textsuperscript{29} in the presence of two mechanical modes $\hat{b}_i$ and two coherent driving fields at $\omega_{l1}$ and $\omega_{l2}$ with the condition $\omega_{l1} + \omega_1 = \omega_{l2} + \omega_2 = \omega_{\text{cav}}$, where $\omega_i$ is the frequency of mechanical mode $i$, $i=1,2$. In the linearized approximation and in the frame rotating at $\omega_{\text{cav}}$, the total intracavity light fields $\hat{a}$ is

$$\hat{a} = |\bar{a}_1(t)|e^{i(\omega t + \phi_1)} + |\bar{a}_2(t)|e^{i(\omega t + \phi_2)} + \delta \hat{a}, \quad (S1)$$

where $\bar{a}_i$ is the amplitude of the intracavity field due to driving field $i$, $\phi_i$ is a constant and $\delta \hat{a}$ is a fluctuating term. The evolution of $\delta \hat{a}$ is given by

$$\delta \ddot{a} = -\frac{k}{2} \delta \dot{a} + i \{G_1 \dot{x}_1 + G_2 \dot{x}_2 \} \delta \hat{a}, \quad (S2)$$

where $G_i$ is the optical frequency shift per displacement of the mechanical mode $\dot{x}_i = x_{zpm,i}(\dot{b}_i + \dot{b}_i^\dagger)$ with $x_{zpm}$ being the zero-point motion of mode $i$. Neglecting the
thermal occupation of the environment, the mechanical modes evolve as

\[ \dot{\hat{b}}_i = \left( -\frac{\Gamma_i}{2} - i\omega_i \right) \hat{b}_i + ig_0\hat{a}^\dagger \hat{a}, \quad (S3) \]

\[ \hat{c}_i = -\frac{\Gamma_i}{2} \hat{c}_i + ig_0|\alpha_i|\hat{a}, \quad (S6) \]

\[ \delta \hat{a} = -\frac{\kappa}{2} \delta \hat{a} + iG_1 x_{zpm,1}|\alpha_1|\hat{c}_1 + iG_2 x_{zpm,2}|\alpha_2|\hat{c}_2, \quad (S7) \]

Using the multiphoton optomechanical coupling \( g_i(t) = G_i x_{zpm,1}|\alpha_1(t)| \), changing notation \( \hat{c} \to \hat{b} \) and \( \delta \hat{a} \to -\delta \hat{a} \) we get Eq. (3) of the main text

\[ \dot{\hat{b}}_i = -i\frac{\Gamma_i}{2} \hat{b}_i + g_i(t)\delta \hat{a}, \quad (S8) \]

\[ i\delta \hat{a} = -i\frac{\kappa}{2} \delta \hat{a} + g_1(t)\hat{b}_1 + g_2(t)\hat{b}_2, \quad (S9) \]

and \( \kappa \) are modified in the following way:

\[ -i\frac{\Gamma_1}{2} \to -i\frac{\Gamma_1}{2} - i\frac{\Gamma_{\text{opt}12}(t)}{2} + \delta \omega_{\text{opt}12}(t), \quad (S10) \]

\[ -i\frac{\kappa}{2} \to -i\frac{\kappa}{2} + \Delta_{\text{2ph}}, \quad (S11) \]

\[ -i\frac{\Gamma_2}{2} \to -i\frac{\Gamma_2}{2} - i\frac{\Gamma_{\text{opt}21}(t)}{2} + \delta \omega_{\text{opt}21}(t) - \Delta_{2\text{ph}}, \quad (S12) \]

where \( \Gamma_{\text{opt}ij} \) and \( \delta \omega_{\text{opt}ij} \) are the optomechanically induced damping rate and frequency shift of mode \( i \) due to light fields at \( \omega_{L,j} \) [16] [29].
V. GENERATION OF DRIVING PULSES

As mentioned in the main text, fluctuations in the difference of the frequencies of the two driving pulses must be much less than $1/T_{\text{transfer}}$ for the adiabaticity condition to be satisfied\cite{16}. We achieve this by generating both driving pulses from the same pump laser by frequency shift, see Fig. S2. An AC voltage with frequency $\omega_{\text{AOM}}$ generates two light fields in the first diffraction maximum of the acousto-optical modulator (AOM) with frequencies $\omega_{\text{pump}} \pm \omega_{\text{AOM}}$. In order to independently address both frequencies required for the state transfer ($\omega_{L1}$ and $\omega_{L2}$), we send two electronic pulses to the AOM with frequencies $\omega_{\text{AOM},i}$, $i=1,2$ and Gaussian envelopes generated by an arbitrary function generator (ArbFunGen). The pump laser detuning $\Delta = 3.5$ MHz is chosen so that $\omega_{Li} = \omega_{\text{pump}} + \omega_{\text{AOM},i} = \omega_{\text{cav}} - \omega_i$ for mechanical modes at $\omega_i$, $i=1,2$, and the effect on the transfer process of the other pair of light fields at $\omega_{\text{pump}} - \omega_{\text{AOM},i}$ and harmonics $\omega_{\text{pump}} + k \cdot \omega_{\text{AOM},i}$, $k=2,3,4,...$ is negligible. The measured amplitude of the 2nd harmonics is much smaller than that of the 3rd harmonics, as is represented by the arrows labeled “harm.” in Fig. S2. To check the effect of the harmonics, we excite the mechanical modes to a level much higher than the thermal motion and we send driving pulses individually during the mechanical decay. With the above shown value for $\Delta$, mode 2 is not affected by the pulse sent to the AOM at $\omega_{\text{AOM},1}$ within detection sensitivity, while the measured effect of the pulse at $\omega_{\text{AOM},2}$ on mode 1 agrees well with the theoretically predicted optomechanical effect from the light fields at $\omega_{L2}$.

VI. CALIBRATION PROCEDURE

The transfer efficiency is defined in the main text as the ratio of the phonon population in mode 2 at the end of the transfer process to the phonon population in mode 1 at the beginning. The number of phonons in a mechanical mode $\langle \hat{b}_i \hat{b}_i^\dagger \rangle = 0.5 \propto u_i^2 \propto R_i^2$, where $u_i$ is the amplitude of oscillation, and $R_i$ is the amplitude of the demodulated reflection signal measured at $\omega_i + \omega_{\text{EOM}}$. Thus the transfer efficiency is

$$\text{Eff}_{1\rightarrow 2} = \frac{k_2 R_2^2(t_{\text{end},1\rightarrow 2})}{k_1 R_1^2(t_{\text{beginning},1\rightarrow 2})},$$

(S13)

where the state is transferred from mode 1 to mode 2 and $k_i$ are coefficients of proportionality. Let us consider the reverse transfer $2 \rightarrow 1$. The product of transfer efficiencies

$$\text{Eff}_{1\rightarrow 2} \text{Eff}_{2\rightarrow 1} = \frac{R_2^2(t_{\text{end},1\rightarrow 2})}{R_1^2(t_{\text{beginning},1\rightarrow 2})} \frac{R_2^2(t_{\text{end},2\rightarrow 1})}{R_1^2(t_{\text{beginning},2\rightarrow 1})},$$

(S14)

does not have any coefficients of proportionality, thus it can be measured directly without any calibration. For the parameters of the transfer $\sigma = 25$ ms and $\Delta t/\sigma = 1.25$, this product is measured to be $0.73 \pm 0.05$. This implies that we demonstrate a transfer efficiency of at least $\sqrt{\text{Eff}_{1\rightarrow 2}\text{Eff}_{2\rightarrow 1}} = 0.855 \pm 0.03$, independently of the model and calibrations. A numerical solution of the full model shows that for the above chosen $\sigma$ and $\Delta t$, the efficiencies $\text{Eff}_{1\rightarrow 2}$ and $\text{Eff}_{2\rightarrow 1}$ differ by 0.01, which amount to the transfer efficiency from the defect mode to the 3.3 mode being $0.86 \pm 0.03$, see Fig. S3.

The AOM used to shape the driving pulses has a nonlinear intensity vs voltage response, which causes the actual temporal profile of the pulse’s intensity to deviate from a Gaussian shape. Another consequence of this nonlinearity is that the sum of intensities of individual pulses is not equal to the intensity of the pulse resulting from two Gaussian pulses being added and sent to the AOM. To account for these undesired effects, we measured the time profiles of the multiphoton optomechanical coupling $g_i(t)$ as follows. We excite mode 1 to a level much higher than the thermal occupation. During the mechanical decay, we send a single short Gaussian pulse $g_1(t,\sigma)$ to the cavity, with frequency $\omega_{\text{AOM}}$ and the same peak intensity as used for the STIRAP measurements. We adjust $\sigma$
Figure S3. **Representative single runs of state transfer from mode 1 to mode 2 (left) and in the opposite direction (right).** Left scale, thick lines: phonon population as a function of time, red line corresponds to mode 1, blue line to mode 2, both divided by the phonon population of mode 1 in the beginning of the transfer. Right scale, thin lines: multiphoton optomechanical couplings $g_1(t)$ red line, $g_2(t)$ blue line. The driving field pulses have a nearly Gaussian temporal profile, but their beginning and ending are modified such that they have zero amplitude outside the pulse. Vertical lines indicate the beginning and ending of the transfer process. Black stars correspond to the phonon populations used to calculate transfer efficiency.

for this pulse so that exactly half of the initial excitation energy is lost due to the optomechanical damping. This gives $\sigma_{1/2} = 0.12 \pm 0.01$ ms. Numerical solution of Eq. (3) for such a pulse gives the peak value of the pulse $\max g_1(t) \sim 2$ kHz. Next a similar procedure is followed for mode 2, but $\sigma$ of the pulse is set equal to $\sigma_{1/2}$, and the peak value of the pulse is set so that exactly half of the initial excitation of mode 2 is lost after the pulse $g_2(t, \sigma_{1/2})$ at $\omega_{L2}$. This gives the estimate of $\max g_2(t) \sim 2$ kHz and the required voltage amplitude sent to AOM in the pulse.

To get the actual temporal profile of $g_i(t)$, we measure in transmission the time profiles of the intensities of the pulses used for the transfer, with $\sigma = 25$ ms and all the values of $\Delta t/\sigma$ used for the measurements (-1:0.25:4). In order to measure the exact temporal intensity profile of both STIRAP pulses individually, while both pulses are simultaneously applied (STIRAP sequence), the pump laser detuning $\Delta$ is adjusted such that $\omega_{cav} - \omega_{L1} \sim \kappa$, while $\omega_{L2} + \omega_2 = \omega_{L1} + \omega_1$ as always, making $|\omega_{cav} - \omega_{L2}| \gg \kappa$. Therefore the transmitted light consists almost exclusively of the intensity at $\omega_{L1}$. To correct for the small fraction of light at $\omega_{L2}$, we send this pulse individually with the same detunings, and subtract the measured transmission from the case when both pulses are present. We follow the same procedure in order to measure the individual intensity of light at $\omega_{L2}$. The measured intensity profiles of the pulses are used in the numerical simulations presented here and in the main text.

[1] U. Gaubatz, P. Rudecki, M. Becker, S. Schiemann, M. Kühl, and K. Bergmann, Chemical Physics Letters 149, 463 (1988).
[2] P. Pillet, Physical Review A - Atomic, Molecular, and Optical Physics 48, 1 (1993).
[3] J. L. Sørensen, D. Moller, T. Iversen, J. B. Thomsen, F. Jensen, P. Staanum, D. Voigt, and M. Drewsen, New Journal of Physics 8 (2006), 10.1088/1367-2630/8/11/261.
[4] K. S. Kumar, A. Vepsäläinen, S. Danilin, and G. S. Paraoanu, Nature Communications 7, 1 (2016).
[5] H. Goto and K. Ichimura, Physical Review A - Atomic, Molecular, and Optical Physics 75, 1 (2007).
[6] D. A. Golter and H. Wang, Physical Review Letters 112, 1 (2014).
[7] S. Longhi, Laser and Photonics Reviews 3, 243 (2009).
[8] J. Simon, H. Tanji, S. Ghosh, and V. Vuletic, Nature Physics 3, 765 (2007).
[9] K. Toyoda, K. Uchida, A. Noguchi, S. Haze, and S. Urabe, Physical Review A - Atomic, Molecular, and Optical Physics 87, 1 (2013).
[10] Y. D. Wang and A. A. Clerk, Physical Review Letters 108, 1 (2012).
[11] L. Tian, Physical Review Letters 108, 1 (2012).
[12] C. Dong, V. Fiore, M. C. Kuzyk, and H. Wang, Science 338, 1609 (2012).
[13] R. A. Norte, J. P. Moura, and S. Gröblacher, Physical Review Letters 116, 1 (2016), arXiv:1511.06235.
[14] Y. Tsaturyan, A. Barg, E. S. Polzik, and A. Schliesser, Nature Nanotechnology 12, 776 (2017).
[15] S. M. Meenehan, J. D. Cohen, G. S. MacCabe, F. Marsili, M. D. Shaw, and O. Painter, Physical Review X 5, 1 (2015), arXiv:1503.05135.
[16] N. V. Vitanov, A. A. Rangelov, B. W. Shore, and K. Bergmann, Reviews of Modern Physics 89, 1 (2017).
[17] H. Xu, D. Mason, L. Jiang, and J. G. Harris, Nature
[18] K. Fang, M. H. Matheny, X. Luan, and O. Painter, Nature Photonics 10, 489 (2016).

[19] M. J. Weaver, F. Buters, F. Luna, H. Eerkens, K. Heeck, S. De Man, and D. Bouwmeester, Nature Communications 8, 1 (2017).

[20] C. Chen, D. H. Zanette, D. A. Czaplewski, S. Shaw, and D. López, Nature Communications 8, 1 (2017). [arXiv:1612.00490]

[21] R. W. Andrews, R. W. Peterson, T. P. Purdy, K. Cicak, R. W. Simmonds, C. A. Regal, and K. W. Lehnert, Nature Physics 10, 321 (2014). [arXiv:1310.5276]

[22] P. Piergentili, L. Catalini, M. Bawaj, S. Zippilli, N. Malossi, R. Natali, D. Vitali, and G. D. Giuseppe, New Journal of Physics 20 (2018), 10.1088/1367-2630/aad85f. [arXiv:1805.09699]

[23] C. Gärtner, J. P. Moura, W. Haaxman, R. A. Norte, and S. Gröblacher, Nano Letters 18, 7171 (2018). [arXiv:1809.06372]

[24] D. Garg, A. K. Chauhan, and A. Biswas, Physical Review A 96 (2017), 10.1103/PhysRevA.96.023837. [arXiv:1706.05869]

[25] J. D. Thompson, B. M. Zwickl, A. M. Jayich, F. Marquardt, S. M. Girvin, and J. G. Harris, Nature 452, 72 (2008).

[26] S. Schmid, K. D. Jensen, K. H. Nielsen, and A. Boisen, Physical Review B - Condensed Matter and Materials Physics 84, 1 (2011).

[27] R. W. Drever, J. L. Hall, F. V. Kowalski, J. Hough, G. M. Ford, A. J. Munley, and H. Ward, Applied Physics B Photophysics and Laser Chemistry 31, 97 (1983).

[28] A. M. Jayich, J. C. Sankey, B. M. Zwickl, C. Yang, J. D. Thompson, S. M. Girvin, A. A. Clerk, F. Marquardt, and J. G. Harris, New Journal of Physics 10 (2008), 10.1088/1367-2630/10/9/095008. [arXiv:arXiv:0805.3723v1]

[29] M. Aspelmeyer, T. J. Kippenberg, and F. Marquardt, Reviews of Modern Physics 86, 1391 (2014).