This is the accepted manuscript made available via CHORUS. The article has been published as:

**Multimode Strong Coupling in Cavity Optomechanics**
P. Kharel, Y. Chu, D. Mason, E. A. Kittlaus, N. T. Otterstrom, S. Gertler, and P. T. Rakich
Phys. Rev. Applied **18**, 024054 — Published 19 August 2022
DOI: [10.1103/PhysRevApplied.18.024054](https://doi.org/10.1103/PhysRevApplied.18.024054)
Multimode strong coupling in cavity optomechanics

P. Kharel1,1,∗ Y. Chu,1,2 D. Mason,1 E. A. Kittlaus,1 N. T. Otterstrom,1 S. Gertler,1 and P. T. Rakich1

1Department of Applied Physics, Yale University, New Haven, Connecticut 06511, USA and Yale Quantum Institute, Yale University, New Haven, Connecticut 06520, USA
2Department of Physics, ETH Zürich, 8093 Zürich, Switzerland and Quantum Center, ETH Zürich, 8093 Zürich, Switzerland

Optomechanical systems show great potential as quantum transducers and information storage devices for use in future hybrid quantum networks. In this context, optomechanical strong coupling can enable efficient, high bandwidth, and deterministic transfer of quantum states. While optomechanical strong coupling has been realized at optical frequencies, it has proven difficult to identify a robust optomechanical system that features the low loss and high coupling rates required for more sophisticated control of mechanical motion. In this paper, we demonstrate strong coupling in a Brillouin-based bulk cavity optomechanical system in both the single and multimode strong coupling regime, which leads to a useful device for both applications in quantum information and for investigating decoherence phenomenon in bulk acoustic wave resonators. Using nontrivial mode hybridizations in the strong coupling regime, we create hybridized photonic-phononic modes with lifetimes that are significantly longer than those of the uncoupled system. This surprising lifetime enhancement, which results from the interference of decay channels, showcases the use of multimode strong coupling as a general strategy to control extrinsic decoherence mechanisms. Moreover, phonons supported by such BAW resonators have a collection of properties, including high frequencies, long coherence times, and robustness against thermal decoherence, making this optomechanical system particularly enticing for applications such as quantum transduction and memories. Hence, this system provides access to phenomena in a previously unexplored regime of optomechanical interactions and could serve as an important building block for future quantum devices.

I. INTRODUCTION

As is the case for many quantum-optical systems, optomechanical devices exhibit novel physical behaviors and acquire new useful capabilities when they enter the so-called “strong coupling regime” [1]. In this regime, the coupling rate between light and motion becomes faster than both the optical and mechanical dissipation rates, which is necessary for applications such as quantum transduction [2, 3] and memories [4, 5]. Since light is the natural carrier of quantum information over long distances [6], and mechanical motion efficiently couples to many quantum systems [7–9], a robust and coherent interface between light and mechanical motion could be a useful building block in hybrid quantum networks for long-distance communications [10] or modular quantum computation [11]. In such systems, strong coupling permits operation in the regime where transduction bandwidth can be maximized. Beyond quantum communication, strong coupling is also necessary for applications that seek to utilize the information stored in the optical mode[12] rather than released from the cavity such as in hybrid quantum systems where light is coupled to individual atoms or quantum dots [13, 14].

Only a few optomechanical devices have entered the regime of optomechanical strong coupling due to technical challenges associated with realizing low-loss systems that can also robustly support high coupling rates. Radiation pressure has been used to achieve strong coupling between THz-frequency optical modes and MHz-frequency mechanical modes within micromechanical systems [15, 16]. However, if we seek to utilize optomechanical systems as a quantum resource, it is advantageous to instead utilize high frequency (GHz) phonon modes; this is because higher frequencies yield lower thermal decoherence at any given temperature, enable faster quantum operations, and allow access to the mechanical ground state using standard refrigeration techniques. Despite the many successes of GHz-frequency micro- and nanoptomechanical systems [17], it remains challenging to reach strong coupling in such systems due to practical limits [18, 19] on the circulating photon number, which in turn limit the cavity-enhanced coupling rate.

Through an alternative approach, Brillouin interactions have been used to demonstrate strong coupling to GHz-frequency mechanical modes of a fused-silica whispering gallery mode resonator [20]. This strategy permits resonant driving of the optical mode, and the use of macroscopic fused-silica based resonators having low material- and surface-induced absorption alleviates some of the technical challenges associated with laser heating, making large coupling rates more readily accessible. While this recent demonstration illustrates the advantages of Brillouin-based coupling in macroscopic systems, low acoustic dissipation rates–necessary to store information in the mechanical mode–are difficult to achieve in glasses at cryogenic temperatures. In particular, two-level tunnelling-state systems, which are intrinsic to silica, produce excess dissipation and noise at cryogenic
temperatures [21, 22], complicating the prospects for efficient quantum operations in such systems. However, a promising way to address this challenge could be to realize strong coupling between optical cavity modes and the long-lived high-frequency (>10 GHz) phonon modes supported by crystalline bulk acoustic wave (BAW) resonators [23].

Separate from the opportunities presented by strong coupling, another direction with significant untapped potential in cavity optomechanics—more generally in quantum information science—involves the exploration of coupled multimode systems. Such systems have already given rise to the observation of a wide variety of interesting physical phenomena, such as optomechanical dark modes [24, 25], synchronization of mechanical frequencies [26], topological dynamics [27], and non-reciprocity [28]. Achieving strong coupling within a multimode system could lead to a wealth of new capabilities including storage of light through control of bright and dark states [29] and exploration of topological phonon transport [30, 31]. Furthermore, in the quantum regime, multimode strong coupling could open the door to generation of multipartite mechanical entanglement [32–34] and the implementation of a quantum simulator for many-body bosonic systems [35, 36].

In this paper, we utilize Brillouin interactions to demonstrate strong coupling between a single optical mode and one or more high-frequency (12.6 GHz) modes of a crystalline bulk acoustic resonator at cryogenic temperatures. Our system combines a high-finesse optical resonator with a low-loss, crystalline BAW resonator, which can be reconfigured so that the optical mode is strongly coupled to either a single acoustic mode or several acoustic modes. Using both frequency and time-domain measurements, we quantify the parameters of the system and explore its dynamics. In the single mode case, we achieve an optomechanical coupling rate of $g_m = 2\pi \times (7.2 \pm 0.1)$ MHz, which exceeds both the optical dissipation rate of $\kappa = 2\pi \times (4.43 \pm 0.02)$ MHz and the mechanical dissipation rate of $\Gamma_m = 2\pi \times (66 \pm 3)$ kHz. In the multimode case, the coupling rates exceed the acoustic free spectral range of $\delta = 2\pi \times (610\pm10)$ kHz, meaning we enter the multimode strong coupling regime. In this regime, strong coupling produces a new set of optomechanical “dark modes” with linewidths that are a factor of five less than the smallest dissipation rate, $\Gamma_m$, of the uncoupled system. We show that this intriguing phenomenon can be explained by the destructive interference of radiative loss channels for the dark modes.

II. CAVITY OPTOMECHANICAL SYSTEM

Our optomechanical system consists of a planar quartz crystal that is placed within a Fabry-Pérot optical cavity with high reflectivity (99.9%) optical mirrors (Fig. 1a). At a temperature of $\sim 10$ K, long-lived longitudinal acoustic modes within the quartz crystal are reflected from the planar surfaces of the crystal to form a series of macroscopic standing wave acoustic modes similar to the standing wave electromagnetic modes formed within a Fabry-Pérot optical cavity. A high-frequency acoustic mode within the BAW resonator (formed by the quartz crystal) can mediate coupling between two distinct longitudinal modes of the optical cavity through Brillouin interactions when energy conservation and phase matching requirements are satisfied (Fig. 1b). For crystalline z-cut quartz at cryogenic temperatures and optical modes near 1550 nm, such interactions occur for a narrow band of acoustic modes near 12.6 GHz (see Supplementary Information section I [38]).

This multimode coupling can be described by the in-


Figure 2. Optomechanical strong coupling to a single mechanical mode. (a) Probe laser transmission spectra taken at various control laser powers. Dashed lines show expected values of $g_m$ for the dominantly coupled acoustic mode, extrapolated from fits to low-power spectra with $\sqrt{P_m} < 0.6 \text{ mW}^{1/2}$ (see Supplementary Information section II [38]). Lower and upper panels (inset i - ii) show spectra in the regimes of weak and strong coupling, respectively. In the strongly coupled case, the normal-mode splitting indicated is due to the dominantly-coupled acoustic mode, but OMIT features are visible from other acoustic modes that are still weakly coupled. (b) Probe transmission taken at various cryostat temperatures. Dashed red line shows the value of $\Delta$ at each temperature, obtained by fitting the data to the theoretical expression for probe transmission (see Supplementary Information section II [38]). Panels (inset iii - iv) show spectra in the regimes of weak and strong coupling, respectively. In the strongly coupled case, the transmission spectrum develops additional features in the strong coupling regime. Since a BAW resonator supports multiple acoustic modes with regular frequency spacing ($\delta$) that is smaller than the optical dissipation rate ($\kappa$), it is important to go beyond the minimal model of a single optical mode coupled to a single phonon mode. This is because, more than one acoustic modes can simultaneously mediate coupling between the same pair of optical modes (Fig. 1b). Therefore, in addition to normal-mode splitting of a single strongly coupled acoustic mode ($\Omega_m$), we expect several OMIT dips to arise from weak coupling to a multitude of acoustic modes (e.g. $\Omega_{m-2}, \Omega_{m-1}, \Omega_{m+1}, \Omega_{m+2}$) as seen in Fig. 1d.

III. EXPERIMENTAL RESULTS

A. Strong coupling to a single acoustic mode

We first present experimental measurements of strong optomechanical coupling when our system is configured to couple predominantly to a single acoustic mode. For the lowest control laser power, the transmission spectrum seen in Fig. 2a.i reveals a single OMIT dip at $\Omega_m = 2\pi \times 12.591 \text{ GHz}$. Through these low power measurements in the weak coupling regime, we extract $\kappa = 2\pi \times (4.43 \pm 0.02) \text{ MHz}$, $\Gamma_m = 2\pi \times (66 \pm 3) \text{ kHz}$, and $g_m^0 = 2\pi \times (23 \pm 1) \text{ Hz}$ (See Supplementary Information section II [38]). Note that the asymmetric line shape seen in Fig. 2i (inset) is characteristic of leaky
The temporal response observed using this method help to elucidate some subtle features that emerge from the optical and acoustic modes, indicating that our system is in the strong coupling regime. For mechanical oscillators with non-zero thermal occupations, it is relevant to consider not only how the coupling rate compares to the dissipation rates, but also to the total (thermal) decoherence rate, $\gamma_{\text{th}} = n_{\text{th}} \Gamma_m$. In this way, one can define the quantum cooperativity, $C_q = 4 g_m^2 / \kappa \gamma_{\text{th}}$. Achieving $C_q > 1$ indicates that quantum state transfer between photons and phonons can occur at a much faster rate than the mechanical decoherence. This opens the door for many quantum-coherent protocols, including efficient and low-noise quantum transduction of information between the optical and acoustic domain [40, 41].

To characterize $\gamma_{\text{th}}$ (and thus $C_q$), one must measure the mechanical bath occupation, which we accomplish through a series of calibrated thermometry measurements of undriven thermal motion. By carefully characterizing our optical detection path with an optical calibration tone, and extracting optomechanical scattering rates with a driven measurement, we can reliably calibrate measured RF voltage spectra into units of mechanical quanta necessary to extract the effective phonon occupation number (see Supplementary Information section III for details [38]). From these measurements, we extract a thermal bath occupation of $n_{\text{th}} = 25 \pm 1$, yielding a thermal decoherence rate of $2\pi \times (1.6 \pm 0.1)$ MHz.

To understand our prospects for reaching high quantum cooperativities, we must investigate the possibility of spurious mode heating at higher powers (and higher coupling rates). Absorbed light has been observed to cause excessive heating of the mechanical mode [18, 41], limiting the performance of state-of-the-art quantum optomechanical experiments [2, 43]. By comparison, this bulk crystalline system has several properties that could prove advantageous in this regard. For example, the high-purity quartz crystals used within our optomechanical system have exceedingly low material absorption [44]. At the same time, the macroscopic crystalline substrate has high thermal conductivity ($>20$ W cm$^{-1}$ K$^{-1}$) which peaks around 10 Kelvin [45] and also has good thermal anchoring to the cryostat, helping to minimize any temperature changes produced by any deposited heat. To quantify the degree of possible laser heating, we repeat our calibrated thermometry experiments in the presence of an auxiliary ‘heating laser’ that is used to drive a separate optical mode (not participating in the optomechanical process).

Within the uncertainty of our measurements, we observe that the presence of a strong heating laser does not alter the thermal decoherence rate of the mechanical mode at input optical powers of 150 mW, which is comparable with the highest control laser powers used to demonstrate strong coupling (see Supplementary Information section III for details [38]).

These measurements confirm that we can reach optomechanical scattering rates ($4g_m^2/\kappa$) which exceed the thermal decoherence rate ($\gamma_{\text{th}}$) by over an order of magnitude, corresponding to the regime of strong quantum cooperativity (or $C_q > 1$). With the parameters demonstrated here, ground-state sideband cooling experiments should be straightforward, though one must carefully manage and model the multimode interactions and coupled dissipation channels. In this direction, it may be more straightforward to work with a plano-convex crystal [23], which offers a higher acoustic finesse, such that multimode interactions are less relevant.

Next we demonstrate time-domain control and utilize it to study the dynamics of our system under a pulsed probe signal. These pulsed operations are not only an important step toward implementing deterministic state transfer for quantum transduction and information storage, but also allow us to perform accurate measurements of the timescales for coherent and dissipative dynamics within our optomechanical system [41]. We probe the time domain dynamics of our system by pulsing a weak probe light ($\omega_1$) while maintaining a strong continuous drive at $\omega_1$ (Fig. 3a). A heterodyne signal resulting from the interference between the control and the probe light transmitted through the cavity provides phase-sensitive detection of the probe light as a function of time (See Supplementary Information section I [38]).

The temporal response observed using this method help to elucidate some subtle features that emerge from multimode strong coupling. These time-domain measurements, shown in Fig. 3b, were performed at the
strong control laser is continuously on-resonance with the optical mode at \( \omega_1 \) to turn on the optomechanical coupling (see Fig. 1b). A short probe pulse excites the optical mode at \( \omega_2 = \omega_1 + \Omega_m \) and the response of the system is then recorded as a function of time (see Supplementary Information section I [38]). (b) Time-domain measurements taken at the same set of cryostat temperatures as in Fig. 2b. Note that in these measurements, the probe frequency is centered at \( \Omega_m \), but has a large enough bandwidth to excite the optical mode even when it is detuned. (c) Probe transmission as a function of time after probe pulse is turned on (top) and zoom in (bottom) showing oscillations at \( \pi/g_m \) and exponential decay with timescale \( \tau \approx 70 \text{ ns} \).

Figure 3. Time-domain measurements of strong coupling. (a) Schematic of the time-domain measurement. A strong control laser is continuously on-resonance with the optical mode at \( \omega_1 \) to turn on the optomechanical coupling (see Fig. 1b). A short probe pulse excites the optical mode at \( \omega_2 = \omega_1 + \Omega_m \) and the response of the system is then recorded as a function of time (see Supplementary Information section I [38]). (b) Time-domain measurements taken at the same set of cryostat temperatures as in Fig. 2b. Note that in these measurements, the probe frequency is centered at \( \Omega_m \), but has a large enough bandwidth to excite the optical mode even when it is detuned. (c) Probe transmission as a function of time after probe pulse is turned on (top) and zoom in (bottom) showing oscillations at \( \pi/g_m \) and exponential decay with timescale \( \tau \approx 70 \text{ ns} \).

So far, we have demonstrated robust strong coupling between a single optical mode and a single high frequency acoustic mode in our Brillouin-based bulk cavity optomechanical system. Next, we show that this system can be reconfigured to achieve multimode strong coupling regime, which leads to useful device physics for both applications in quantum information and investigating decoherence phenomena in bulk acoustic resonators. To enter the multimode strong coupling regime, we strongly couple a single optical mode to three acoustic modes (Fig. 4a). We accomplished this by tuning the optical wavelength to select a different pair of optical modes, which changes the spatial overlap between the optical and acoustic modes (See Supplementary Information section I [38]). The transmission spectrum taken at low power (Fig. 4b.i) reveals three OMIT dips. As before, theoretical fits to OMIT spectrum at low powers allow us to extract coupling rates \( g_1 = 2\pi \times (4.9 \pm 0.1) \text{ MHz} \), \( g_2 = 2\pi \times (4.0 \pm 0.1) \text{ MHz} \) and \( g_3 = 2\pi \times (3.7 \pm 0.1) \text{ MHz} \), as well as dissipation rates \( \kappa = 2\pi \times (2.52 \pm 0.08) \text{ MHz} \) and \( \Gamma_m = 2\pi \times (67 \pm 10) \text{ kHz} \) (See Supplementary Information section II [38]). In the strong coupling regime, we observe four distinct peaks in the transmission spectrum seen in Fig. 4b.ii. These peaks represent the four eigenmodes produced by the hybridization of the optical mode \((a_2)\) with the three dominant phonon modes \(b_1, b_2, \) and \(b_3\).

To understand the nature of these four new eigenmodes, we start by considering a simpler case of a single optical mode \((a_2)\) coupled strongly to two phonon modes \((b_1, b_2)\) separated by \(2\delta\). Furthermore, we assume that \(g_1 = g_2 = g, \Gamma_1 = \Gamma_2 = \Gamma, \) and \(\Omega_{1,2} = \Delta \mp \delta\). The Hamiltonian of this three coupled oscillator system in the basis of \(a_2, b_1, \) and \(b_2\) is given by

\[
H_{\text{eff}} = \begin{bmatrix}
\Delta - i\kappa/2 & -g & -g \\
-g^* & \Omega_1 - i\Gamma/2 & 0 \\
-g^* & 0 & \Omega_2 - i\Gamma/2
\end{bmatrix}.
\]

This effective Hamiltonian can be diagonalized to obtain three eigenmodes of the hybridized system (see Supplementary Information section V [38]). In the limit of large \(g\), these eigenmodes become two ‘bright’ modes \(B_{\pm} = \frac{1}{\sqrt{2}}(b_1 + b_2)\) at frequencies \(\omega_{\pm} = \Delta \pm \sqrt{2}g\) with dissipation rates \(\kappa_{\pm} = \kappa/2\) and one ‘dark’ mode \(D = \frac{1}{\sqrt{2}}(b_1 - b_2)\) at frequency \(\omega_D = \Delta\) with a dissipation rate \(\kappa_D = \Gamma\). Notice that the bright modes are formed from the superposition of both the optical and the acoustic modes whereas the dark mode lacks an optical mode component, meaning that it does not couple
to light. The dynamics of such a system, and the existence of such bright and dark modes, has been explored in an electromechanical system using a GHz frequency microwave resonator strongly coupled to two MHz frequency micromechanical oscillators [29]. However, this regime of coupling has not been previously accessible for optomechanical systems.

Extending the Hamiltonian in Eq. (3) to treat the case of a single optical mode coupling to three acoustic modes, we now expect four eigenmodes of the hybridized system seen in Fig. 4a. Of these eigenmodes, the two broad peaks correspond to the bright modes, whereas the two narrow peaks correspond to the dark modes. This analysis leads us to expect the decay rates of these dark modes to approach the mechanical decay rate \( \Gamma_m = 2\pi \times 67 \text{ kHz} \). However, high-resolution measurements of such modes at the highest control laser powers (seen in Fig. 4b.iv) reveal decay rates \( \Gamma_d \approx 10.9 \text{ µs} \) confirms that both eigenmodes, which are hybridized excitations of both light and sound, have lifetimes that are significantly longer than the optical and mechanical lifetimes of the uncoupled system.

The measured decay time of \( \tau_d \approx 10.9 \text{ µs} \) confirms that both eigenmodes, which are hybridized excitations of both light and sound, have lifetimes that are significantly longer than the optical and mechanical lifetimes of the uncoupled system.

black The observed dissipation reduction phenomenon can be understood as a form of coherent cancellation that occurs when mechanical modes—which couple into a common reservoir—become strongly hybridized. To understand this, we note that the Brillouin-active phonon modes supported by this crystal structure can be viewed as leaky modes that lose energy through coupling (or radiation) to a band of phonon modes within the flat-flat crystal geometry. Hence, it is natural to decompose the total decay rate (\( \Gamma \)) into distinct radiative (\( \Gamma_{\text{rad}} \)) and intrinsic (\( \Gamma_{\text{int}} \)) contributions, such that \( \Gamma = \Gamma_{\text{rad}} + \Gamma_{\text{int}} \). Importantly, this radiation constitutes a common bath, and as such \( \Gamma_{\text{rad}} \) will appear as both a decay term and a dissipative coupling [47, 48] between different mechanical modes (see Supplementary Information section V [38]).

In the simple case of two acoustic modes treated above, this would correspond to the addition of an off-diagonal term \( -\gamma \Gamma_{\text{rad}} \) linking modes \( b_1 \) and \( b_2 \). In this case, the resulting decay rate of the dark mode, \( D \), becomes \( \kappa_D = (\Gamma - \Gamma_{\text{rad}}) + \kappa \delta^2 / 2g^2 \). If radiative loss was the only decay channel, we would thus find \( \kappa_D \rightarrow 0 \) as \( g \) becomes large. In reality, \( \kappa_D \rightarrow \Gamma_{\text{int}} \), which may be set by surface scattering and absorption due to imperfections in the crystal. At cryogenic temperatures, \( \Gamma_{\text{int}} \) can be very small within pristine crystals [23, 49], opening the possibility of extremely long-lived dark modes. We note that analogous line-narrowing phenomena due to interference of decay pathways has been investigated in fluorescence spectra of a V-type atomic system [50] as well as in a circuit quantum electrodynamics (cQED) platform [51].

Measurement of the linewidths of the two dark modes as a function of power (Fig. 4d) agrees well with the theoretical description of our system presented above. Theoretical fits to the data, for the case of three phonon modes, were performed by numerically diagonalizing the effective Hamiltonian that includes the radiative coupling terms. Only \( \Gamma_{\text{int}} = 2\pi \times 5 \text{ kHz} \) is taken as a fit parameter (see Supplementary Information section V [38]) and is consistent with independent measurements of acoustic damping in quartz crystals at cryogenic temperatures [23, 52]. Note that we observe a larger than expected linewidth of the dark modes at the highest powers, which could be due to a deviation from the linear dependence of \( g \) on \( \Gamma_{\text{int}} \) (see Supplementary Information section III [38]). Nevertheless, these experiments clearly demonstrate that, through the formation of hybridized modes, multimode strong coupling becomes a powerful tool to dynamically manipulate decoherence pathways in optomechanical systems.

IV. DISCUSSION

These results demonstrate that optomechanical coupling to a multitude of high-frequency, low-loss phonons within BAW resonators present intriguing opportunities for investigating classical and quantum phenomena in the multimode strong coupling regime. Moreover, the time-domain measurements presented in this paper represent an important step towards optical control of bulk acoustic phonons for quantum transduction and the generation of non-classical mechanical states. While our system is already in the quantum-coherent strong-coupling regime necessary to observe quantum effects, a number of improvements can be made to achieve robust quantum control of phonons and realize the aforementioned goals.

First, it is possible to directly initialize such high-frequency (12.6 GHz) phonons in their quantum ground states at temperatures \(< 1 \text{ K} \) by using a standard dilution refrigerator. Decreasing \( \kappa \) by improving mirror reflectivity to 99.99% and utilizing low-loss crystalline substrates with larger Brillouin gain (such as TeO₂) could enable access to the strong coupling regime at \(< 100 \mu\text{W} \) input powers. These improvements, along with low duty-cycle pulsed operation of the control laser with micro-Watt average powers could make operation in dilution refrigerators feasible. More importantly, it would be beneficial to design a fiber-coupled optical cavity system with piezo-tunable crystal position to minimize stray optical reflections as well as to provide enhanced control over coupling to one or more phonon modes.

These improvements could offer avenues for utilizing multimode optomechanical interactions for future applications in quantum information and metrology. For in-
Figure 4. Optomechanical strong coupling to three mechanical modes. (a) Diagram of linearized optomechanical coupling between an optical mode and three acoustic modes (top), corresponding expected spectra of probe laser transmission in the weak and strong coupling regime (middle), and the coupling rate under a strong laser drive (bottom). Strong coupling between an optical mode and three acoustic modes gives rise to four hybridized excitations of light and sound with two narrow resonances corresponding to optical ‘dark’ modes and the two broad resonances corresponding to optical ‘bright’ modes. (b) Probe laser transmission spectra taken at various $P_m$. The right panel shows a zoom-in of this spectra around frequency $f_0 = 12.684$ GHz. Lower and upper panels (inset i-iv) show spectra in the regimes of weak and strong coupling, respectively. In the strongly coupled case (insets ii and iv), two narrow resonances are observed, corresponding to the optomechanical dark modes. (c) Time-domain measurement of dark modes. Inset shows the pulse sequence for the control and probe lasers. (d) Measured linewidth of the two dark modes at various control laser powers and fits to theory with $\Gamma_{\text{int}}/2\pi = 5$ kHz as described in the main text. The shaded region corresponds to the theoretically-predicted linewidth of the two dark modes when we vary the fit parameter $\Gamma_{\text{int}}/2\pi$ from 3 kHz to 7 kHz.

stance, it may be possible to adiabatically transfer quantum optical states, such as single-photons, to the long-lived dark states for quantum information storage. Moreover, it may be possible to use the nontrivial mode hybridization to generate entangled mechanical states of the resonator by the simultaneously swapping optical excitations to multiple strongly coupled acoustic modes [53]. In addition, as shown by the line-narrowing phenomena we observed, strong coupling between light and acoustic modes within BAW resonators could be used to explore and mitigate acoustic dissipation mechanisms. More generally, it has been shown that acoustic waves within BAW resonators couple strongly to a variety of other quantum systems such as superconducting qubits [54], defect centers [55], and microwave fields [9]. Therefore, deterministic control of bulk acoustic waves using light in the strong coupling regime could be a valuable tool for manipulating quantum information and exploring physical phenomena in hybrid quantum systems.

ACKNOWLEDGMENTS

The authors thank Jack G. E. Harris, Robert J. Schoelkopf, and Liang Jiang for insightful discussions regarding coherent phenomena and for generous contribution of technical expertise and experimental resources. We also thank Luke Burkhart, Vijay Jain, and Yizhi Luo for helpful discussions and valuable feedback. This work was initially supported by the U.S. Office of Naval Research under award number N00014-17-1-2514 and completed under U.S. Department of Energy, Office of Science through award number DE-SC0019406. Y.C. was also supported by the Army Research Office under award number W911NF-14-1-0011. E.A.K. received support from the Packard Fellowship for Science and Engineering. N.T.O. acknowledges support from the National Science Foundation Graduate Research Fellowship under Grant number DGE1122492. The authors of this paper are contributors to patent application no. 62/465101 related to Bulk Crystalline Optomechanics, which was submitted by Yale University.

Author contributions: P.K., Y.C., and E.A.K. performed the experiments under supervision of P.T.R.
P.K., Y.C., and D.M. analyzed the data and developed analytical theory under the guidance of P.T.R.. P.K. designed and built the experimental apparatus to perform the measurement with support from Y.C. and P.T.R.. E.A.K., N.T.O., and S.G. aided in the development of experimental techniques. All authors participated in the writing of this manuscript. P.K and Y.C. contributed equally to this work.

**Competing interests:** The authors declare that they have no competing interests.

**Data and materials availability:** All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data available from authors upon request.

[1] M. Aspelmeyer, T. J. Kippenberg, and F. Marquardt, “Cavity optomechanics,” Rev. Mod. Phys. **86**, 1391–1452 (2014).

[2] M. Mirhosseini, A. Sipahigil, M. Kalaee, and O. Painter, “Superconducting qubit to optical photon transduction,” Nature **588**, 599–603 (2020).

[3] R. D. Delaney, M. D. Urney, S. Mittal, B. M. Brubaker, J. M. Kindem, P. S. Burns, C. A. Regal, and K. W. Lehnert, “Superconducting-qubit readout via low-backaction electro-optic transduction,” Nature **606**, 489–493 (2022).

[4] V. Fiore, Y. Yang, M. C. Kuzyk, R. Barbou, L. Tian, and H. Wang, “Storing optical information as a mechanical excitation in a silica optomechanical resonator,” Phys. Rev. Lett. **107**, 133601 (2011).

[5] K. Stannigel, P. Komar, S. Habraken, S. Bennett, M. D. Lukin, P. Zoller, and P. Rabl, “Optomechanical quantum information processing with photons and phonons,” Phys. Rev. Lett. **109**, 013603 (2012).

[6] H. J. Kimble, “The quantum internet,” Nature **453**, 1023 (2008).

[7] P. Rabl, P. Cappellaro, M. G. Dutt, L. Jiang, J. Maze, and M. D. Lukin, “Strong magnetic coupling between an electronic spin qubit and a mechanical resonator,” Physical Review B **79**, 041302 (2009).

[8] A. D. O’Connell, M. Hofheinz, M. Ansmann, R. C. Bialczak, M. Lenander, E. Lucero, M. Neeley, D. Sank, H. Wang, M. Weides, et al., “Quantum ground state and single-phonon control of a mechanical resonator,” Nature **464**, 697–703 (2010).

[9] X. Han, C.-L. Zou, and H. X. Tang, “Multimode strong coupling in superconducting cavity piezoelectromechanics,” Phys. Rev. Lett. **117**, 123603 (2016).

[10] K. Stannigel, P. Rabl, A. S. Sørensen, P. Zoller, and M. D. Lukin, “Optomechanical transducers for long-distance quantum communication,” Phys. Rev. Lett. **105**, 220501 (2010).

[11] K. Lee, B. Sussman, M. Sprague, P. Michelberger, K. Reim, J. Nunn, N. Langford, P. Bustard, D. Jaksh, and I. Walmsley, “Macroscopic non-classical states and terahertz quantum processing in room-temperature diamond,” Nat. Photonics **6**, 41–44 (2012).

[12] Y.-D. Wang and A. A. Clerk, “Using interference for high fidelity quantum state transfer in optomechanics,” Phys. Rev. Lett. **108**, 153603 (2012).

[13] K. Hammerer, M. Wallquist, C. Genes, M. Ludwig, F. Marquardt, P. Treutlein, P. Zoller, J. Ye, and H. J. Kimble, “Strong coupling of a mechanical oscillator and a single atom,” Phys. Rev. Lett. **103**, 063005 (2009).

[14] J. P. Reithmaier, G. Sek, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. Keldysh, V. Kulakovskii, T. Reinecke, and A. Forchel, “Strong coupling in a single quantum dot–semiconductor microcavity system,” Nature **432**, 197 (2004).

[15] S. Gröblacher, K. Hammerer, M. R. Vanner, and M. Aspelmeyer, “Observation of strong coupling between a micromechanical resonator and an optical cavity field,” Nature **460**, 724–727 (2009).

[16] E. Verhagen, S. Delégilse, S. Weis, A. Schliesser, and T. J. Kippenberg, “Quantum-coherent coupling of a mechanical oscillator to an optical cavity mode,” Nature **482**, 63–67 (2012).

[17] M. Eichenfield, J. Chan, R. M. Camacho, K. J. Vahala, and O. Painter, “Optomechanical crystals,” Nature **462**, 78 (2009).

[18] J. Chan, T. M. Alegre, A. H. Safavi-Naeini, J. T. Hill, A. Krause, S. Gröblacher, M. Aspelmeyer, and O. Painter, “Laser cooling of a nanomechanical oscillator into its quantum ground state,” Nature **478**, 89–92 (2011).

[19] R. Riedinger, S. Hong, R. A. Norte, J. A. Slater, J. Shang, A. G. Krause, V. Anant, M. Aspelmeyer, and S. Gröblacher, “Non-classical correlations between single photons and phonons from a mechanical oscillator,” Nature **530**, 313–316 (2016).

[20] G. Enzian, M. Szczykulska, J. Silver, L. Del Bino, S. Zhang, I. A. Walmsley, P. Del’Haye, and M. R. Vanner, “Observation of brillouin optomechanical strong coupling with an 11 ghz mechanical mode,” Optica **6**, 7–14 (2019).

[21] S. Hunklinger and W. Arnold, “Ultrasonic properties of glasses at low temperatures,” Phys. Acoustics **12**, 155–215 (1976).

[22] O. Arcizet, R. Rivière, A. Schliesser, G. Anetsberger, and T. J. Kippenberg, “Cryogenic properties of optomechanical silica microcavities,” Phys. Rev. A **80**, 021803 (2009).

[23] W. Renninger, P. Kharel, R. Behunin, and P. Rakich, “Bulk crystalline optomechanics,” Nat. Phys. **14**, 601–607 (2018).

[24] Q. Lin, J. Rosenberg, D. Chang, R. Camacho, M. Eichenfield, K. J. Vahala, and O. Painter, “Coherent mixing of mechanical excitations in nano-optomechanical structures,” Nat. Photonics **4**, 236 (2010).

[25] C. Dong, V. Fiore, M. C. Kuzyk, and H. Wang, “Optomechanical dark mode,” Science **338**, 1609–1613 (2012).

[26] G. Heinrich, M. Ludvig, J. Qian, B. Kubala, and F. Marquardt, “Collective dynamics in optomechanical arrays,” Phys. Rev. Lett. **107**, 043603 (2011).

[27] H. Xu, D. Mason, L. Jiang, and J. Harris, “Topological energy transfer in an optomechanical system with exceptional points,” Nature **537**, 80 (2016).

[28] F. Ruesink, M.-A. Miri, A. Alu, and E. Verhagen, “Non-reciprocity and magnetic-free isolation based on optome-
mechanical interactions,” Nat. Commun. 7, 13662 (2016).

[29] F. Massel, S. U. Cho, J.-M. Pirkkalainen, P. J. Hakonen, T. T. Heikklä, and M. A. Sillanpää, “Multimode circuit optomechanics near the quantum limit,” Nat. Commun. 3, 987 (2012).

[30] V. Peano, C. Brendel, M. Schmidt, and F. Marquardt, “Topological phases of sound and light,” Phys. Rev. X 5, 031011 (2015).

[31] H. Xu, L. Jiang, A. A. Clerk, and J. G. E. Harris, “Non-reciprocal control and cooling of phonon modes in an optomechanical system,” Nature 568, 65–69 (2019).

[32] M. J. Weaver, D. Newsom, F. Luna, W. Löfler, and D. Bouwmeester, “Phonon interferometry for measuring quantum decoherence,” Phys. Rev. A 97, 063832 (2018).

[33] L. M. d. Lépinay, C. F. Ockeloen-Korppi, M. J. Woolley, and M. A. Sillanpää, “Quantum mechanics–free subsystem with mechanical oscillators,” Science 372, 625–629 (2021).

[34] S. Kotler, G. A. Peterson, E. Shojaee, F. Lecocq, K. Ci- cak, A. Kwiatkowski, S. Geller, S. Glancy, E. Knill, R. W. Simmonds, J. Aumentado, and J. D. Teufel, “Direct observation of deterministic macroscopic entanglement,” Science 372, 622–625 (2021).

[35] M. Ludwig and F. Marquardt, “Quantum many-body dynamics in optomechanical arrays,” Phys. Rev. Lett. 111, 073603 (2013).

[36] M. J. Hartmann, “Quantum simulation with interacting photons,” J. Opt. 18, 104005 (2016).

[37] P. Kharel, G. I. Harris, E. A. Kittlaus, W. H. Renninger, N. T. Otterstrom, J. G. Harris, and P. T. Rakich, “High-frequency cavity optomechanics using bulk acoustic phonons,” Science Advances 5, 1–9 (2019).

[38] See Supplemental Material at [URL will be inserted by publisher] for additional experimental details and references 56-61.

[39] S. Weis, R. Rivière, S. Delégîse, E. Gavartin, O. Arcizet, A. Schliesser, and T. J. Kippenberg, “Optomechanically induced transparency,” Science 330, 1520–1523 (2010).

[40] A. H. Safavi-Naeini, D. Van Thourhout, R. Baets, and R. Van Laer, “Controlling phonons and photons at the wavelength scale: integrated photonics meets integrated phononics,” Optica 6, 213–232 (2019).

[41] S. M. Meenehan, J. D. Cohen, G. S. MacCabe, F. Marsili, M. D. Shaw, and O. Painter, “Pulsed excitation dynamics of an optomechanical crystal resonator near its quantum ground state of motion,” Physical Review X 5, 041002 (2015).

[42] A. H. Safavi-Naeini, J. Chan, J. T. Hill, S. Gröblacher, H. Miao, Y. Chen, M. Aspelmeyer, and O. Painter, “Laser noise in cavity-optomechanical cooling and thermometry,” New Journal of Physics 15, 035007 (2013).

[43] R. Riedinger, A. Wallucks, I. Marinković, C. Löschnauer, M. Aspelmeyer, S. Hong, and S. Gröblacher, “Remote quantum entanglement between two micromechanical oscillators,” Nature 556, 473–477 (2018).

[44] D. Pinnow and T. Rich, “Development of a calorimetric method for making precision optical absorption measurements,” Applied optics 12, 984–992 (1973).

[45] R. Zeller and R. Pohl, “Thermal conductivity and specific heat of noncrystalline solids,” Physical Review B 4, 2029 (1971).

[46] Y. Chu, P. Khare, W. H. Renninger, L. D. Burkhart, L. Fruanzio, P. T. Rakich, and R. J. Schoelkopf, “Quantum acoustics with superconducting qubits,” Science 358, 199–202 (2017).

[47] A. Metelmann and A. A. Clerk, “Nonreciprocal photon transmission and amplification via reservoir engineering,” Phys. Rev. X 5, 021025 (2015).

[48] K. Fang, J. Luo, A. Metelmann, M. H. Matheny, F. Mar- quardt, A. A. Clerk, and O. Painter, “Generalized nonreciprocity in an optomechanical circuit via synthetic magnetism and reservoir engineering,” Nat. Phys. 13, 465–471 (2017).

[49] S. Galliou, M. Goryachev, R. Bourquin, P. Abbé, J. P. Aubry, and M. E. Tobar, “Extremely low loss phonon-trapping cryogenic acoustic cavities for future physical experiments,” Sci. Rep. 3, 2132 (2013).

[50] P. Zhou and S. Swain, “Quantum interference in resonance fluorescence for a driven v atom,” Phys. Rev. A 56, 3011 (1997).

[51] N. M. Sundaresan, Y. Liu, D. Sadri, L. J. Szöcs, D. L. Underwood, M. Malekakhlagh, H. E. Türeci, and A. A. Houck, “Beyond strong coupling in a multimode cavity,” Phys. Rev. X 5, 021035 (2015).

[52] P. Kharel, Y. Chu, M. Power, W. H. Renninger, R. J. Schoelkopf, and P. T. Rakich, “Ultra-high-q phononic resonators on-chip at cryogenic temperatures,” APL Photonics 3, 066101 (2018).

[53] M. J. Weaver, F. Buters, F. Luna, H. Eerkens, K. Heeck, S. de Man, and D. Bouwmeester, “Coherent optomechanical state transfer between disparate mechanical resonators,” Nature communications 8, 824 (2017).

[54] Y. Chu, P. Kharel, T. Yoon, L. Fruznio, P. T. Rakich, and R. J. Schoelkopf, “Creation and control of multi-phonon Fock states in a bulk acoustic-wave resonator,” Nature 563, 666–670 (2018).

[55] E. MacQuarrie, T. Gosavi, A. Moehle, N. Jungwirth, S. Bhave, and G. Fuchs, “Coherent control of a nitrogen-vacancy center spin ensemble with a diamond mechanical resonator,” Optica 2, 233–238 (2015).

[56] D. F. Walls and G. J. Milburn, Quantum optics (Springer Science & Business Media, 2007).

[57] F. Marquardt, J. P. Chen, A. A. Clerk, and S. M. Girvin, “Quantum Theory of Cavity-Assisted Sideband Cooling of Mechanical Motion,” Physical Review Letters 99, 093902 (2007).

[58] H. J. Carmichael, “Quantum trajectory theory for cascaded open systems,” Physical Review Letters 70, 2273–2276 (1993).

[59] A. A. Clerk, M. H. Devoret, S. M. Girvin, F. Marquardt, and R. J. Schoelkopf, “Introduction to quantum noise, measurement, and amplification,” Reviews of Modern Physics 82, 1155–1208 (2010).

[60] D. Cardimona, M. Raymer, and C. Stroud Jr, “Steady-state quantum interference in resonance fluorescence,” Journal of Physics B: Atomic and Molecular Physics 15, 55 (1982).

[61] E. Nichelatti and G. Salvetti, “Spatial and spectral response of a Fabry–Perot interferometer illuminated by a Gaussian beam,” Applied Optics 34, 4703–4712 (1995).