Long-lived guided phonons in fiber by manipulating two-level systems

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The synthesis of ultra-long lived acoustic phonons in a variety of materials and device geometries could enable a range of new coherent information processing and sensing technologies; many forms of phonon dissipation pose a barrier to this goal. We explore linear and nonlinear contributions to phonon dissipation in silica at cryogenic temperatures using fiber-optic structures that tightly confine both photons and phonons to the fiber-optic core. When immersed in helium, this fiber system supports nearly perfect guidance of 9 GHz acoustic phonons; strong electrostrictively mediated photon-phonon coupling (or guided-wave stimulated Brillouin scattering) permits a flexible form of laser-based phonon spectroscopy. Through linear and nonlinear phonon spectroscopy, we isolate the effects of disorder-induced two-level tunneling states as a source of phononic dissipation in this system. We show that an ensemble of such two-level tunneling states can be driven into transparency—virtually eliminating this source of phonon dissipation over a broad range of frequencies. Experimental studies of phononic self-frequency saturation show excellent agreement with a theoretical model accounting for the phonon coupling to an ensemble of two-level tunneling states. Extending these results, we demonstrate a general approach to suppress dissipation produced by two-level tunneling states via cross-saturation, where the lifetime of a phonons at one frequency can be extended by the presence of a high intensity acoustic beam at another frequency. Although these studies were carried out in silica, our findings are quite general, and can be applied to a range of materials systems and device geometries.

I. INTRODUCTION

Access to new regimes of classical and quantum dynamics hinge upon our ability to create and manipulate ultra-long lived coherent excitations in electromagnetic, optical, and phononic domains [1-9]. In particular, ultra-long-lived phonon modes have been identified as a crucial new resource by quantum information, optomechanics, and precision metrology communities [6-10]. To this end, a variety of systems, ranging from nano- and micro-scale phononic devices to resonator technologies of centimeter-scale, have harnessed remarkable phonon coherence times [2-3, 8, 11-19]. However, radical improvements in performance are possible if technical and fundamental sources of dissipation are mastered [3, 8, 9, 22-24]. This realization has spawned a resurgence of interest in the fundamental origins of phonon dissipation at cryogenic temperatures [21-23]. A ubiquitous source of dissipation arises from disorder-induced defects. Some of such defects possess quantized energy spectra, and can exchange energy with electromagnetic, optical, and phononic fields [21-22, 29, 37]. When phonon-active, such defect states can absorb and emit phonons just as atoms absorb and emit light. Dissipation by such defects pose a fundamental limit to phonon lifetimes in acoustic media at cryogenic temperatures. Phonon-active defect states have been extensively studied in [highly disordered] amorphous media [25, 26, 28, 35-41]; however, their deleterious effects also appear within highly ordered crystalline systems and in systems with material interfaces [8, 9, 21]. This form of dissipation bars access to new regimes of classical and quantum dynamics, central to a range of emerging technologies [8, 9].

In this work, we examine the dynamics of an ensemble of phonon-active defects using a guided wave geometry that produces tight confinement of both light and acoustic waves (Fig 1). Strong photon-phonon coupling within this system permits noninvasive optical excitation and interrogation of high frequency (≈9.2 GHz) phonons. Tight confinement of acoustic modes produce high phonon intensities (20 W/m²) in the core of this waveguide, permitting frequency selective nonlinear phonon spectroscopy with modest (∼mW) optical powers over a range of temperatures (1.1-300K). Building on established models, we elucidate the nature of phonon-defect interactions in our system, allowing us to extract defect density, coupling strength, and a range of other parameters that capture the dynamics of the defect ensemble. Using the nonlinear phonon dynamics, we show that this ensemble of defects can be driven into transparency in the strong-field limit; through two different saturation mechanisms, we show that defect-induced dissipation can be suppressed by an estimated factor 45 at 1.1 Kelvin. In this limit, defect-induced dissipation is a negligible source of loss within our guided-wave system; remarkably high phononic Q-factors (>12,000) and decay lengths (>1mm) are achieved. Building on these findings, more sophisticated schemes could provide a path toward radically enhanced coherence times [42] in a range of systems, as the basis for emerging quantum information technologies.

Phonon dissipation can be suppressed by lowering the system temperature (below 150K in our system), and may in itself be a viable strategy to achieve low phonon losses in crystalline material which have low defect densities. However, for amorphous (and even crystalline) materials such as silica or silicon nitride, a temperature will be reached below which the acoustic damping will cease...
Two individual wells characterized by an asymmetric double-well potential for the generalized coordinate $x$ with well-separation $d$, barrier height $V_b$, and the average oscillation frequency of the two individual wells $\omega_c$. Given the finite barrier height tunneling occurs between the two-wells and is quantified by the tunneling strength $\Delta_0$ (defined below). Hence, the energy eigenstates of an uncoupled TLS are symmetric and anti-symmetric superpositions of position states and are split by energy $E = \sqrt{\Delta^2 + \Delta_0^2}$.

The two-level nature of this decay channel leads to non-linearity in the phonon dynamics \[^{[35, 38–41]}\] and provides a way to break through the dissipation floor established by resonant absorption.

We demonstrate that by working with high phonon intensities resonant absorption can be saturated, significantly extending phonon lifetimes. This is accomplished in one of two ways: through self-frequency saturation and cross-frequency saturation. In the first case (self-frequency saturation), the amplitude acoustic mode (at a single frequency) is increased to drive the defects into transparency. In the case of cross-frequency saturation, the lifetime of an arbitrarily weak phonon beam (with a small number of phonons) is extended through spectral hole burning. This is achieved by driving the ensemble of defects out of equilibrium with a high intensity auxiliary phonon beam (at a different frequency).

Two-level tunneling states have generated much contemporary interest in their own right for the possibility of beneficial implications. In particular, it has been shown that they can engender the dynamics of nonlinear electromechanical systems with nonlinearity, even in the single phonon regime \[^{[43]}\], and their relatively long coherence times have facilitated their use as a quantum memory \[^{[33]}\]. In light of this recent interest, our exploration of TLSs and their manipulation may provide valuable information for a variety fields.

II. OVERVIEW

A. Acoustic guidance in fiber

Our system is a 2.2 cm segment of ge-doped Nufern UHNA-3 optical fiber. This fiber’s high germanium concentration ($\sim 44$ wt%) guides light exceptionally and produces a large longitudinal sound speed contrast between core $v_{L,\text{core}} = 4,740 \pm 68$ m/s \[^{[47]}\] and cladding $v_{L,\text{clad}} = 5,944$ m/s \[^{[48]}\] that promotes acoustic guidance. However, ideal guidance is not expected since mode conversion occurs at the core-clad interface \[^{[49]}\] leading to mode leakage since the shear wave velocity in the cladding $v_{S,\text{clad}} = 3,764$ m/s \[^{[48]}\] is smaller than $v_{L,\text{core}}$.

To understand the limits of acoustic guidance we studied the axial-radial acoustic modes of our system with full vectorial simulations and semi-analytical calculations. We solved for acoustic eigenfrequencies of a core region (diameter 1.8 $\mu$m) embedded in a finite cladding (diameter 125 $\mu$m) surrounded by liquid helium. A perfectly absorbing boundary condition was implemented adiabatically in the liquid region at large separation from the core to prevent spurious reflections from the simulation boundaries. We employed a shear velocity in the core region $v_{S,\text{core}} \sim 3092$ m/s which was estimated by interpolating between the values of pure silica and germania, and the liquid region was ascribed the acoustic properties of liquid helium. The core, cladding and liquid were assumed lossless other than the absorbing layer used to model mode leakage.

Acoustic energy that enters the liquid region leaves the system irreversibly and hence reduces the acoustic guidance. The signature of this leakage is an imaginary part to the eigenfrequencies that quantifies the dissipation rate. Our simulations correctly predict the acoustic frequency and yield negligible leakage (much smaller than the dissipation rate observed in experiment). Hence, we conclude that the dissipation of phonons in our system is due almost entirely to internal friction of the fiber as opposed to leakage. In this sense we describe our phonons as well-guided.

It should be emphasized that acoustic guiding in fiber is not guaranteed. Germanium doping has the fortunate consequence that increased doping leads to higher index
a) High temperature transparency

b) Low temperature absorption

c) Saturation-induced transparency

- TLSs resonant with acoustic wave

- Acoustic wave amplitude

acoustic damping due to resonant absorption

resonant absorption saturated at high intensity

FIG. 2. Resonant absorption of phonons of angular frequency \( \Omega \) by TLSs (illustrated by green spheres). a) At high temperatures \((k_B T \gg \hbar \Omega)\) the TLSs addressed by the acoustic wave have almost equal probabilities of being in the excited or ground states, and hence resonant absorption is compensated by stimulated phonon emission. b) At low temperatures \((k_B T \ll \hbar \Omega)\) the TLSs addressed by the acoustic wave condense to their ground states. At sufficiently low acoustic intensities TLSs, having resonantly absorbed a phonon, decay via spontaneous emission thus attenuating the incident acoustic wave in the process. c) At low temperatures and high phonon intensities a given TLS is interrogated by multiple phonons during its excited state lifetime. Hence, an excited TLS may decay via stimulated phonon emission which preserves the coherence of the incident phonon. At high intensities resonant absorption is compensated by stimulated emission.

of refraction while simultaneously decreasing sound speed \[48, 50\], thus enhancing the confinement of light and sound simultaneously. Titanium dioxide and phosphorous pentoxide are other dopants which simultaneously enhance guiding of light and sound, while boron trioxide and fluorine doping enhance the confinement of light while raising the sound speed \[51\]. Fibers doped with the former will possess leaky acoustic modes. At room temperature, where phonons are strongly damped in silica, one may posit that the guiding of sound in fiber plays a minor role as the mean free path is on the order of the acoustic mode field diameter. However, the existence of higher order guided phonon modes can be observed at room temperature \[52, 53\] which shows that acoustic modes in fiber cannot be completely understood in terms of bulk properties without considering boundaries. We have also simulated axial-radial modes in a system composed of a core embedded in an infinite cladding region. In this case the computed acoustic leakage rate exceeds the observed phonon dissipation rate suggesting that the cladding liquid interface plays an important role in the observed acoustic guidance. In light of this fact it is important note that the leakage rate we derived from simulation for the finite fiber case is only a lower bound as we have not accounted for nonuniformity in the geometry of the fiber over its length. Irregularities in cladding and core diameters will contribute to inhomogenous broadening that will manifest as an effective increase in the leakage rate.

B. Stimulated Brillouin Scattering

The strong confinement and guidance of optical and acoustic modes (see Fig. 3) allows for the efficient excitation of phonons via stimulated Brillouin scattering (SBS) and which provides a means to characterize our system \[54–56\].

SBS involves the conversion of a pump photon into a phonon and a Stokes photon with respective angular frequencies \(\omega_p, \Omega, \omega_S\), and wavevector magnitudes \(k_p, q, k_S\) (see Fig. 1 c-d). For backward SBS, 1D phase matching between the non-linear polarization currents and the optical fields \[55\] provide the following relations between the frequencies and wavevector magnitudes

\[
\begin{align*}
\omega_p &= \omega_S + \Omega \quad (1) \\
k_p &= q - k_S. \quad (2)
\end{align*}
\]

The minus sign in front of \(k_S\) in the second equation above indicates that the Stokes photon and the pump counterpropagate (the phonon and the pump copropagate). For a given pump frequency we can approximate the phonon frequency by assuming linear dispersion for light \((\omega = (c/n)k)\) and sound \((\Omega = vq)\)

\[
\Omega \approx \frac{2nv}{c} \omega_p. \quad (3)
\]
where $n$ is the effective index of refraction, $v$ is the modal sound speed, and $c$ is the speed of light. When the acoustic dissipation dominates over optical losses the power of the steady-state Stokes field in space is described by the following equation

$$\frac{dP_S}{dz} = G_B P_p P_S - \alpha_S P_S$$

where $P_S$ is the power in the Stokes field, $P_p$ is the power in the pump field, $G_B$ is the Brillouin gain factor, and $\alpha_S$ is the spatial loss rate for the photons. Generally, $G_B$ is a function of the difference frequency between the pump and Stoke’s field’s, $\omega_{IM}$, which is experimentally set by an intensity modulator (IM) and need not be on resonance, i.e. $\omega_{IM} \neq \Omega$.

$$G_B(\omega_{IM}) = g_B \frac{(\Gamma/2)^2}{(\Omega - \omega_{IM})^2 + (\Gamma/2)^2}$$

where $g_B$ is the peak Brillouin gain, and $1/\Gamma$ is the phonon lifetime.

Balanced detection is employed to determine the amplification of the Stokes field as it traverses the fiber under test (FUT). For this purpose we work expressly in the weak signal regime, given by $G_B P_p L \ll 1$ where the change in the Stokes field power across the fiber $\Delta P_S$ is small, and can be approximately given by

$$\Delta P_S \approx G_B(\omega_{IM}) P_p P_S L = \frac{(\Gamma/2)^2 g_B P_p P_S L}{(\Omega - \omega_{IM})^2 + (\Gamma/2)^2}$$

after solving Eq. 4, where $L$ is the length of the FUT, and where we have dropped a negligible correction arising from the optical loss. Balanced detection is a direct measurement of $\Delta P_S$, and in the weak signal regime provides a Lorentzian frequency response which supplies the peak Brillouin gain, and the phonon frequency and lifetime.

C. Phonon dissipation in glasses

Dissipation of phonons in glasses has several origins: from multi-phonon interactions to scattering by defects. Among the latter of these are hypothesized TLSs that attenuate acoustic waves through the processes of resonant scattering and relaxation absorption. To understand the origins of these two mechanisms we lay out the tunneling state theory and derive its consequences for the dynamics of phonons.

A tunneling state is characterized by a double-well potential of asymmetry $\Delta$ and the overlap energy $\Delta_0 = h\omega_c e^{-\lambda}$ where $\omega_c$ is roughly the average zero point energy of the two wells and $\lambda = \sqrt{2m\hbar \Delta} c$ characterizes the extent of wave function overlap between the two wells with $m$ the mass of the atom(s) comprising the TLS, $V$ the barrier height, and $d$ the ‘distance’ between the double-well’s minima (see Fig. 1b). Given a finite barrier height the atom(s) may tunnel between the two minima of the potential. At low temperatures $k_B T \ll h\omega_c$, when the atom would be in the ground state of either well, a TLS can be effectively represented by the Hamiltonian

$$H_{TLS} = \frac{1}{2} \left( \Delta \frac{\Delta_0}{\Delta_0 - \Delta} \right)$$

where the states $|L\rangle = (0, 1)$ and $|R\rangle = (1, 0)$ correspond with position states with the particle localized in the left (‘L’) or right (‘R’) well and which we approximate as being orthogonal. Diagonalizing the Hamiltonian above gives the corresponding energy eigenstates

$$|e\rangle = \frac{\Delta_0}{\sqrt{2E(E + \Delta)}} \left[ |L\rangle + \frac{\Delta + E}{\Delta_0} |R\rangle \right]$$

$$|g\rangle = \frac{\Delta_0}{\sqrt{2E(E - \Delta)}} \left[ |L\rangle + \frac{\Delta - E}{\Delta_0} |R\rangle \right]$$

which reveal that the stationary states of a free TLS are spatial superpositions between the wells and have energies $\pm \frac{1}{2} E \pm \frac{1}{2}\sqrt{\Delta^2 + \Delta_0^2}$, see Fig. 1c). The parameters $\Delta$ and $\lambda$ are assumed to be uniformly distributed which results in a density of states for the TLS parameters of $G(\Delta, \lambda) = P$, or in terms of the overlap energy $f(\Delta, \Delta_0) = P/\Delta_0$, where $P$ is constant. This assumption of uniformity correctly predicts the linear in temperature behavior of the specific heat and the anomalous thermal conductivity of glasses at low temperatures [38-41].
The local potential experienced by a given TLS is determined by its neighbors in the glass matrix. Hence, any strain which produces relative shifts of neighboring atoms will inevitably perturb the double-well potential of a given TLS. The dominant contribution to the coupling arises from strain-induced shifts in the asymmetry $\Delta$, and hence leads to a perturbation of the diagonal elements of the Hamiltonian in Eq. 1.

$$H = H_{\text{ph}} + \sum_j \left[ \frac{1}{2} (E_j + D_j : \xi_j (r_j)) \sigma_{z,j} + M_j : \xi_j (r_j) \sigma_{x,j} \right]$$

where $\xi_{ab}$ is the elastic strain, $\frac{1}{2} \partial^2 \xi_{ab} = \gamma_{ab}$ is the deformation potential characterizing the linear response of the TLS potential to strain, 'a' and 'b' denote spatial components of the strain tensor, and the Einstein summation convention for repeated indices is used.

Upon diagonalization of the full Hamiltonian, including the strain-induced perturbation of the asymmetry, we find

$$H = H_{\text{ph}} + \sum_j \left[ \frac{1}{2} (E_j + D_j : \xi_j (r_j)) \sigma_{z,j} + M_j : \xi_j (r_j) \sigma_{x,j} \right]$$

where abbreviated notation $A : B \equiv A^{ab} B_{ab}$ has been introduced, the sum over $j$ counts all TLSs in the glass at various positions $r_j$, with energy splitting $E_j$ and coupling parameters $D_j$ and $M_j$, the 2 x 2 matrices representing the Hamiltonian for the TLSs have been replaced by Pauli matrices $\eta_i$, where $i = \{x,y,z\}$, and $D_j^{ab} = 2(\Delta_j/E_j) \gamma_{ab}$ and $M_j^{ab} = (\Delta_0/E_j) \gamma_{ab}$. $H_{\text{ph}}$ is the free Hamiltonian for the phonons

$$\sum_{q,\eta} \hbar \Omega \eta_q (b_{q\eta}^\dagger b_{q\eta} + 1/2)$$

where the sum counts modes with wavevector and polarization $q$ and $\eta$, and $b_{q\eta}$ and $\Omega \eta_q$ are the $q\eta$-mode annihilation operator and frequency, respectively. The decomposition of the strain field into normal modes, here in plane waves, reveal the connection with the annihilation operator $b_{q\eta}$

$$\xi_{ab}(x) = \frac{i}{2} \sum_{q,\eta} (q_a \hat{r}_{q\eta}(q) + q_b \hat{r}_{q\eta}(q)) \sqrt{\frac{\hbar}{2 \Omega \eta_q \rho V}} e^{i q \cdot x} b_{q\eta} + H.c. \tag{11}$$

where $V$ is the volume of the system, $\rho$ is the material density, $\hat{r}_{q\eta}(q)$ is a unit vector for $\eta$-polarized phonons, and $H.c.$ stands for Hermitian conjugate.

As a final remark regarding the underpinnings of the theory, the deformation potential tensor of each TLS is generally unique and orientation dependent; we ignore this complication and replace $\gamma_{ab}$ with an averaged effective linear response of the TLS potential that we define by its contraction on the strain

$$\gamma : \xi(x) \equiv i \sum_{q,\eta} q \gamma \eta \sqrt{\frac{\hbar}{2 \Omega \eta_q \rho V}} e^{i q \cdot x} b_{q\eta} + H.c. \tag{12}$$

where $\gamma_0$ are constant. This assumption states that the potential deformation of a given TLS is independent of the incident phonon’s propagation direction $q$.

**D. TLS lifetime**

Before we begin our investigation of TLS-induced effects upon the phonons we outline some of the phonon-induced effects upon the TLSs which play a role in our later analysis. The most important of these effects is that interaction with the phonons leads to a finite TLS upper state lifetime.

For a TLS of asymmetry $\Delta$ and overlap energy $\Delta_0$ Fermi’s golden rule gives the excited state decay rate (see Appendix)

$$\frac{1}{\tau} = \sum_{\eta} \frac{\gamma_{\eta}^2 E \Delta_0^2}{\nu_{\eta}^2} \coth \left( \frac{E}{2k_B T} \right) \tag{13}$$

where the sum over $\eta$ counts decay channels corresponding with the various phonon polarizations, and $T$ is the temperature of the phonon bath. The polarization dependence of the deformation potential and the sound speed are accounted for in $\eta$ suffices. For fixed energy $E$ the lifetime $\tau$ has the minimum value $\tau_{\text{min}}$ obtained from Eq. 13 by taking $\Delta_0 \rightarrow E$.

**E. Resonant phonon absorption by TLSs**

Tunneling states can resonantly interact with phonons in three ways: stimulated absorption, and spontaneous and stimulated emission. The relative magnitude of each of these processes determines the phonon dissipation rate and depends on temperature and the intensity of the acoustic field.

1. **Weak fields**

First, we consider weak fields, meaning that the mean free time between TLS-phonon interactions is long compared to the excited state lifetime of the TLS, and which will be defined quantitatively below. For such low intensity acoustic waves of angular frequency $\Omega \eta_q$ and polarization $\eta$ the golden rule can be used to find the TLS-induced decay rate (see Appendix)

$$\Gamma_{\text{res}} \eta_q = \frac{\pi P_{\gamma}^2 \Omega \eta_q}{\rho v_{\eta}^2} \tanh \left( \frac{\hbar \Omega \eta_q}{2k_B T} \right), \tag{14}$$

$$\frac{\pi P_{\gamma}^2 \Omega \eta_q}{\rho v_{\eta}^2} \left( \frac{P_g}{(i)} - \frac{P_e}{(ii)} \right)$$
and is characterized by two regimes occurring at high and low temperatures. This is elucidated by the temperature dependence of the phonon decay rate on the thermal population inversion \( P_c - P_g \) of the TLSs at energy \( h\Omega_{\text{q}} \) and temperature \( T \), where \( P_c \) and \( P_g \) are the equilibrium probabilities to find a TLS in the excited and ground state, respectively. At low temperatures \( h\Omega \gg k_B T \) the decay rate is maximized since the TLSs are found entirely in their ground state \((P_g \to 1)\), and thus (i) stimulated absorption dominates. In the low intensity regime TLSs that have absorbed phonons decay through spontaneous emission which attenuates coherent phonon beams. As the temperature is raised the probability to find the TLS in its excited state grows which opens the possibility for (ii) stimulated phonon emission which amplifies the sound amplitude. At high temperatures these two processes compensate each other exactly and resonant absorption is suppressed.

2. Strong fields

At high acoustic intensities perturbation theory is no longer adequate to calculate the phonon decay rate. In this regime one must solve the coupled Heisenberg equations of motion for the TLSs and the acoustic field given by

\[
\dot{\sigma}_{z,j} = \frac{2}{\hbar} M_j : \xi(r_j) \sigma_{y,j} \tag{15}
\]

\[
\dot{\sigma}_{y,j} = \frac{1}{\hbar} (E_j + D_j : \xi(r_j)) \sigma_{x,j} - \frac{2}{\hbar} M_j : \xi(r_j) \sigma_{z,j} \tag{16}
\]

\[
\dot{\sigma}_{x,j} = -\frac{1}{\hbar} (E_j + D_j : \xi(r_j)) \sigma_{y,j} \tag{17}
\]

\[
\dot{b}_{q\eta} = -i\Omega_{q\eta} b_{q\eta} - \frac{1}{2} \sum_j g_{q\eta,j} (\Delta_j \sigma_{z,j} + 2\Delta_{0,j} \sigma_{x,j}) \tag{18}
\]

where \( g_{q\eta,j} \equiv \frac{\gamma_j}{\pi E_j} q \sqrt{\frac{\hbar}{2\Delta_{q\eta} q^2} e^{-iqr_j}} \).

When the acoustic field is driven strongly at angular frequency \( \omega \approx \Omega_{\text{q}} \) the physics can be greatly simplified. Under these conditions we focus our attention solely on the classical steady-state dynamics of the \( q\eta \)-mode. We account for the remaining, thermally populated, phonon modes with phenomenological damping terms and ‘Langevin’ forces that influence the TLS dynamics. Strong driving also allows the use of the rotating wave approximation (RWA) that we implement by decomposing the strain field and the \( x \) and \( y \) Pauli operators into positive and negative frequencies i.e. \( \xi = \xi^{(+)} + \xi^{(-)} \) where \( \xi^{(\pm)} \propto e^{\pm i\omega t} \), and similarly \( \sigma_{x,j} = \sigma_{x,j}^{(+)} + \sigma_{x,j}^{(-)} \) where \( \sigma_{x,j}^{(\pm)} \propto e^{\pm i\omega t} \). The operator \( \sigma_{y,j} \) is similarly decomposed and \( \sigma_{z,j} \) is assumed to be time-independent in the steady-state limit.

After these simplifications the coupled equations of motion for the driven phonon amplitude \( \beta_{q\eta} \), and the mean values for the TLS operators, \( \langle \sigma_{i,j} \rangle \equiv S_{i,j} \), reduce to

\[
\dot{S}_{z,j} = -\frac{1}{T_1} (S_{z,j} - w_{0,j}) + \frac{2}{\hbar} M_{\eta,j} \left[ \zeta_{q\eta}^{(-)}(r_j) S_{y,j}^{(+)} + H.c. \right] \tag{19}
\]

\[
\dot{S}_{y,j}^{(+)} = -\frac{1}{T_2} S_{y,j}^{(+)} + \frac{1}{\hbar} E_j \zeta_{x,j}^{(+)} - \frac{2}{\hbar} M_{\eta,j} \zeta_{q\eta}^{(+)}(r_j) S_{z,j} \tag{20}
\]

\[
\dot{S}_{x,j}^{(+)} = -\frac{1}{T_2} S_{x,j}^{(+)} - \frac{1}{\hbar} E_j \zeta_{y,j}^{(+)} \tag{21}
\]

where the TLS phenomenological decay rates \( 1/T_1 \), quantifying the upper state lifetime, and \( 1/T_2 \), characterizing the dephasing rate, arise from the interaction with the thermal phonon field, and additionally in the latter from spectral diffusion of a given TLS’s oscillation frequency \( \frac{1}{\hbar}(E_j - D_j : \xi(r_j)) \) as the static background strain field is modified by spin flips of neighboring TLSs. The strain amplitude of the \( \psi\eta \)-mode \( \zeta^{(+)}_{q\eta} \) is given by the coefficient of \( b_{q\eta} \) in Eq. 11 with \( b_{q\eta} \) replaced by \( \beta_{q\eta} \), \( M_{\eta,j} \) is \( \gamma_j |\Delta_j/E_j| \), and \( F_{q\eta} \) is the magnitude of an external drive at angular frequency \( \omega \). The thermal equilibrium value of \( S_{z,j} \) given by \( w_{0,j} \equiv -\tanh(E_j/2k_B T) \) plays the role of a ‘Langevin’ force in this system of equations by ensuring the return to thermal equilibrium in the absence of driving.

In steady-state and noting that \( \zeta^{(+)} = \zeta^{(-)} \dagger \) the equation for \( S_{z,j} \) can be solved in terms of \( \zeta_{q\eta}^{(+)} \) giving

\[
S_{z,j} = \frac{w_{0,j}}{1 + 4M_{\eta,j}^2 T_1 T_2 \left| \zeta_{q\eta}^{(+)} \right|^2 \left( \frac{1}{1 + i\omega T_2} + \frac{1}{1 + i\Lambda T_2} \right)} \tag{20}
\]

where \( \delta_j \equiv E_j/\hbar - \omega \) and \( \Sigma_j \equiv E_j/\hbar + \omega \). Eq. 20 is the nonequilibrium steady-state value for the population inversion at energy \( E_j \) under driving by an acoustic beam at angular frequency \( \omega \). Eq. 20 can be used to find the steady-state solution for \( S_{x,j} \) which can then be plugged into the equation of motion for the phonon annihilation operator to give the TLS-influenced phonon dynamics

\[
i(\Omega_{q\eta} - \omega) \beta_{q\eta} \approx F_{q\eta} + \sum_j \frac{M_{\eta,j}^2 q^2 T_1}{2\hbar\Omega_{q\eta} \rho V} \left[ 1 + iT_2 \delta_j - \frac{1}{1 - iT_2 \Sigma_j} \right] S_{z,j} \beta_{q\eta} \tag{21}
\]

The backreaction of the TLSs on the phonons is characterized by a complex susceptibility \(-i\Delta w_{q\eta} - \frac{i}{2} \Gamma_{q\eta}^{\text{res}} \) that modifies the phonon dynamics in two ways: The real part results in dissipation of the phonon beam by resonant absorption, and the imaginary part induces a frequency shift (to be discussed below).
The sum over the various TLSs can be performed by assuming the validity of the ergodic hypothesis which states that the volume average is equal to the ensemble average of the TLSs in the thermodynamic limit i.e.,
\[
\frac{1}{V} \sum_j J(\Delta_j, \Delta_{0,j}, E_j) \rightarrow \int d\Delta \int d\Delta_0 P/\Delta_0 F(\Delta, \Delta_0, E)
\]
where \( F \) represents the summand and recall that \( P/\Delta_0 \) is the TLS density of states. By assuming that \( T_2 \) is insensitive to \( \Delta_0 \), using the ergodic hypothesis, and by working in ‘polar’ coordinates \((E, \phi), \) i.e \( \Delta = E \cos \phi \) and \( \Delta_0 = E \sin \phi \), the dissipation rate is given by
\[
\Gamma_{qh}^{\text{res}} \approx \frac{P\gamma_q^2 \Omega_{q\eta}}{\hbar \rho v_0^2} \int_{-\infty}^{\infty} dE \frac{T_2 \tanh(E/2k_B T)}{1 + (\frac{E}{\hbar} - \omega)^2 T_2^2 + \frac{\delta T_{\text{min}} T_2}{\hbar} (\xi_{q\eta}^2)^2}
\]
where \( \omega T_2 \gg 1 \) has been used, and \( T_1 \) is taken to be approximately given by Eq. [13]. For \( \omega T_2 \gg 1 \), the integrand is sharply peaked for \( E \approx \omega \). Assuming that \( T_2, \tau_{\text{min}}, \) and \( \tanh(E/2k_B T) \) vary little over the range \( E \in h(\omega - 1/T_2, \omega + 1/T_2) \) the integral may be approximated as
\[
\Gamma_{qh}^{\text{res}} \approx \frac{\pi P\gamma_q^2 \Omega_{q\eta} \tanh(h\omega/2k_B T)}{\rho v_0^2} \sqrt{1 + J}/J_c
\]
To arrive at this result we’ve used the expression for the acoustic intensity \( J = 2\rho v_0^2 |\xi_{q\eta}^2|^2 \), and we’ve introduced the critical intensity
\[
J_c = \frac{\hbar^2 \rho v_0^2}{2\gamma_q^2 \tau_{\text{min}} T_2} \approx \frac{\hbar^2 \rho v_0^2}{2\gamma_q^2 T_1 T_2}
\]
that demarcates the boundary between high and low intensity. With \( f(\Delta, \Delta_0) \) and Eq. [13] the density of TLSs as a function of \( E \) and \( \tau \) can be derived. This distribution is highly peaked near \( \tau_{\text{min}} \) (see Eq. 3.8 of [11]), and thus we approximate \( T_1 \gg \tau_{\text{min}} \) as evidenced on the far right in Eq. [21].

As with the case of weak fields, this result for the phonon dissipation can be interpreted as a competition between stimulated emission and absorption. In the strong field case however the effective population inversion for the TLSs which interact with a phonon of frequency \( \omega \) is given by the nonequilibrium steady-state value
\[
P_e - P_y = -\frac{\tanh(h\omega/2k_B T)}{\sqrt{1 + J}/J_c}
\]
At high intensities \( J/J_c \gg 1 \) a TLS, excited through resonant absorption, will encounter several phonons within its upper state lifetime. Thus, stimulated emission can become a dominant decay channel and resonant absorption will be suppressed.

A similar calculation can be performed for the frequency shift \( \Delta \omega_{q\eta} \)
\[
\Delta \omega_{q\eta}(T) - \Delta \omega_{q\eta}(T_0) \approx -\frac{P\gamma_q^2 \Omega_{q\eta}}{\rho v_0^2} \left[ \ln \left( \frac{h\Omega_{q\eta}}{k_B T} \right) \right] - \text{Re} \Psi \left( \frac{1}{2} + \frac{h\Omega_{q\eta}}{2\pi k_B T} \right) - (T \rightarrow T_0)
\]
where \( \Psi \) is the digamma function.

**F. Relaxation absorption**

In addition to resonant absorption TLSs can attenuate coherent acoustic waves via relaxation absorption. This process results from a modulation of the TLS’s energy splitting by the presence of a time-dependent strain field \( E_j(t) \rightarrow E_j + D_j : \xi(t) \). As \( E_j(t) \) changes, the TLS can equilibrate with the surroundings by absorbing or releasing energy. Thus, the equilibrium population \( w_{0,j} \) in Eq. [19] possesses a time-dependent component, i.e., \( w_{0,j} \rightarrow w_{0,j}(t) = w_{0,j} + \frac{\partial w_{0,j}}{\partial E_j} D_j : \xi(t) \), which acts as a drive field for the level inversion, and leads to the decoherence of the perturbing field \( \xi_{ab}(t) \).

In taking the RWA to arrive at Eqs. [19] we have dropped a \( S_{z,j} \)-dependent drive term in the equation of motion for the phonon amplitude. However, when the time dependence of the level inversion is taken into account this term now couples to the phonons and leads to dissipation and a frequency shift.

The steady-state amplitude of the time-dependent component of \( S_{z,j} \) that dominantly couples to \( \beta_{q\eta} \) is given by
\[
\delta S_{(+)}^{z,j} = \frac{1}{1 - i \omega T_1} \frac{\partial w_{0,j}}{\partial E_j} D_{q\eta,j} \xi(+) \rho_{v\eta} (r_j)
\]
where \( \delta S_{z,j} \equiv S_{z,j} - w_{0,j} \). Accounting for this additional drive term in the equation of motion for the phonons leads to
\[
(i(\Omega_{q\eta} - \omega + \Delta \omega_{q\eta}) + \frac{1}{2} \Gamma_{q\eta}^{\text{res}}) \beta_{q\eta} = F_{q\eta}
\]
\[
- \frac{i}{V} \sum_j \frac{D_{q\eta,j}^2 \Omega_{q\eta}}{4\rho v_0^2 T_1} 1 - i \omega T_1 \frac{\partial w_{0,j}}{\partial E_j} \beta_{q\eta}.
\]
Just as in the case of resonant absorption, the effect of relaxation absorption is quantified by the complex susceptibility \(-i \Delta \omega_{q\eta}^{\text{rel}} - \frac{1}{2} \Gamma_{q\eta}^{\text{rel}} \). Here we focus on the relaxation-induced damping coefficient
\[
\Gamma_{q\eta}^{\text{rel}} = -\frac{2}{V} \sum_j \frac{D_{q\eta,j}^2 \Omega_{q\eta} w_{0,j} T_1}{4\rho v_0^2 1 + \omega^2 T_1^2} \int_{0}^{\infty} \frac{dE}{E_j}. \]
To evaluate $\Gamma_{q\eta}^{\text{rel}}$ we employ the ergodic hypothesis and note an important observation about the relative magnitudes of the probing frequency and the inversion decay in our experiment. Assuming that $T_1$ is given by the upper state lifetime of the TLSs then it takes a minimum value calculated by taking $\Delta_0 \to E$ in Eq. 13. Furthermore, the factor $\partial \ln \rho / \partial E$ exponentially suppresses contributions to the integrals from $E$ bigger than $k_B T$. Hence, for the frequencies of interest in our experiment it can be shown that $\omega T_1 \gg 1$ over the integration range contributing to $\Gamma_{q\eta}^{\text{rel}}$. Thus, a Taylor expansion of the integrand of $\Gamma_{q\eta}^{\text{rel}}$ is justified for small $1/\omega T_1$ and results in

$$
\Gamma_{q\eta}^{\text{rel}} = \frac{P_{\gamma q}^2}{\rho v^2 k_B T} \int d\Delta \int d\Delta_0 \Delta^2 \frac{1}{\Delta_0 E^2} \cosh^2 \frac{E}{2k_B T} 
$$

where we’ve taken $\Omega_{q\eta} / \omega = 1$ and where we’ve replaced $T_1$ with $\tau$. The integrals can performed giving

$$
\Gamma_{q\eta}^{\text{rel}} \approx \frac{\pi^3}{24} \frac{P_{\gamma q}^2}{\rho v^2 k_B T^2} \left( \sum_{\eta'} \frac{\gamma_{\eta\eta'}}{e^2} \right) (k_B T)^3. \quad (31)
$$

In the following we use the formulae of this section to analyze the contribution of TLSs to the dissipation of phonons in our system.

III. EXPERIMENTAL SETUP

The system is characterized by pump-probe measurements performed using balanced detection and lock-in amplification. Our apparatus consists of a 1548.963 nm source that is split into two optical lines (See Fig. 4); one reserved to act as a pump, and the second to act as a probe. The pump line is subsequently amplified and pump and probe polarizations are aligned. The pump beam is then power modulated at the fixed frequency $\Omega_{\text{mod}}$ for lock-in detection before being sent through the FUT. The probe beam is sent through an intensity modulator which upon exit is filtered to isolate a single side-band. To achieve common mode noise rejection using balanced detection, the probe beam is split into two arms. One arm passes probe light through the FUT and is amplified via SBS whereas the other arm acts as a reference. A variable attenuator on the reference arm is adjusted so that both probe arms have the same optical power in the absence of gain via SBS. The difference in power of the balanced probe arms yields the net gain experienced by the probe beam passing through the sample.

Modulation of the pump power generates a fixed-frequency side-band of the amplified probe which is detected with a lock-in amplifier. To measure Brillouin gain spectra (BGS) the modulation frequency $\omega_{1M}$ is swept across the Brillouin resonance generating a Lorentzian response.

The temperature of the fiber is controlled using a double walled cryostat, and a large copper heat sink. The samples are mounted in a shallow mail slot passing through the block. The block size was chosen to ensure temperature stability, and to allow a minimum of 10 Brillouin lineshape measurements per 100 mK as the block warms slowly to room temperature. The system is cooled using liquid Helium and evaporative cooling resulting in lowest achievable temperatures in the neighborhood of 1 K.

To study the saturation of losses arising from the TLSs the intensity of the sound field inside the FUT was swept at fixed temperature. In the weak signal regime the steady-state phonon intensity follows the powers of the optical fields

$$
J(\omega_{1M}) \approx \frac{v}{\omega_{1M}} \frac{1}{A_{\text{eff}}} G_B(\omega_{1M}) P_p P_S L, \quad (32)
$$

where $A_{\text{eff}}$ is the phonon mode area. Hence, the phonon intensity can be controlled by changing the power of the optical driving fields. By sequential area of the variable optical attenuators on pump and probe lines the phonon intensity generated through SBS can be swept through more than 4 decades of dynamics range. The maximum pump and probe powers were used to determine the 2.2 cm length of the FUT; this length ensures that all measurements were performed in the weak-signal regime. This fiber length allowed access to high phonon intensities while simultaneously preventing inhomogeneous broadening from non-linearity induced by strong backward scattering via SBS.
IV. RESULTS

A. Temperature and Intensity Dependence of Phonon Losses

Brillouin gain spectra corresponding to the fundamental acoustic mode in a 2.2 cm segment of UHNA-3 fiber were continuously acquired as the system slowly warmed to room temperature from 4.17 K. Pump and probe powers were set to 35 mW and 0.550 mW, respectively to ensure SBS measurements in the weak signal regime. The BGS were binned in 100 mK steps, averaged, and fit to the Lorentzian model given by Eq. 6. We determined the frequency of the acoustic mode Ω and the dissipation rate Γ as a function of temperature. The red data points in Figure 5a show the dissipation rate of the fundamental acoustic mode as a function of temperature. Three representative BGS are shown in Fig. 5d. To judge the relative importance of resonant absorption by TLSs we have plotted the ratio $P_\gamma/P_0$ as a red curve (top).

For low phonon intensities ($J \ll J_c$) the linewidth begins to level off below 4K (red points in Figs. 5a & 5b) and then begins to increase as the temperature is lowered. This gray region in Figs. 5a & 5b, where $P_\gamma/P_0 < 0.9$, indicates the temperature range where resonant absorption by TLSs begins to dominate the acoustic damping. After cooling the system to 1.1 K, we let the fiber system slowly warm up to 4.17 K. For each 100 mK rise in temperature we acquire BGS as a function of phonon intensity. The data is analyzed by binning and averaging as described above, and results in dissipation rate as a function of both temperature and phonon intensity. This data is presented in Figs. 5a & 5b: shows phonon dissipation rate as a function of temperature. The three sets of data correspond with three distinct settings of the optical powers that generate phonons of low, moderate, and high intensities (as compared to $J_c$). Normalized BGS for the three settings is shown in Fig. 5c: for the lowest temperature of 1.1 K. Figure 5c shows phonon dissipation rate as a function of phonon intensity for three different temperatures (black, orange and tan points). It is qualitatively clear from Figs. 5a & 5b: that at temperatures lower than 4 K phonon dissipation is suppressed at high phonon intensity; this indicates saturation of the resonant absorption due to TLSs and is made evident by the theoretical plot of $P_\gamma/P_0$ (at top), computed using Eq. 33.

In Figs. 5f, g we compare our measurements with the tunneling state model. In Fig. 5g: a model of the phonon dissipation rate given by

$$\Gamma = \Gamma^{res}_{q_0} + \Gamma_0$$

(33)

is fit to the data where $\Gamma_0$ is an offset parameter that represents all intensity independent background acoustic losses including relaxation absorption, Rayleigh scattering, phonon-phonon interactions, etc. Three parameters are employed to fit the data: 1) $P_\gamma J_0$ ($\gamma_L$ being the deformation potential for longitudinal waves), 2) $J_c$, and 3) the offset $\Gamma_0$. The parameters $P$ and $\gamma_L$ are assumed to be constant. In distinction, $J_c$ and $\Gamma_0$ are expected to depend upon temperature. In particular, $J_c$ scales inversely with the effective decay rates $T_1 T_2$. We expect $T_1 \approx \tau_{\text{min}}$, but an estimation of the temperature dependence of $T_2$ is beyond the scope of this work. Instead we fit $J_c$ (gray dots in Fig. 5f) to a power law $J_c = a T^b$ that is shown as a solid gray line in [Fig. 5]. In Fig. 5f: the measured shift in Brillouin frequency (gray points) referenced to $T_0 = 1.09$ K is compared to Eq. 26.

The fitted values obtained from Fig. 5g were used as inputs. The offset $\Gamma_0$ is plotted as blue points in Figs. 5g and contains all background losses which we model as

$$\Gamma_0 = \Gamma^{rel}_{q_0} + \Gamma_{\text{Background}}$$

(34)

where $\Gamma_{\text{Background}}$ is a constant representing the background dissipation floor for our system. The model is fit to the data by adjusting the offset and the value of the longitudinal deformation potential $\gamma_L$ (we assume that the transverse deformation potential is given by $\gamma_T^2 \approx 2 \gamma_L^2$). $P_\gamma J_0^2$ is held fixed to the value obtained from the data analysis in Fig. 5c.

The model parameter values obtained from this analysis are tabulated in Tab. 1 and their values are compared with those of vitreous silica. There are several remarks in order. The data of Fig. 5g is well-described by the tunneling state model, and the computed Brillouin frequency shift using the fitted value of $P_\gamma J_0^2$ from Fig. 5g compares well with our measurements. Relaxation absorption accounts well for the intensity independent background, and the fitted value for $\gamma_L$ agrees well with silica. The fitted parameters and their theoretical relationships can also be used to estimate the density of TLSs $P$ and the dephasing rate $T_2$.

As a final note, the values of $T_1$ and $T_2$ should not be directly compared with those of vitreous silica listed in Tab. 1. The phonon frequencies and the temperatures were entirely different from our experiment. $\tau_{\text{min}}$ at $T = 20$ mK and 0.68 GHz for the parameters we find for ge-doped silica is 277 µs.

B. Phononic Spectral Hole Burning

The non-linear dynamics induced by the coupling to TLSs mediates an interaction between phonons with different frequencies. We experimentally study this effect by adding, auxiliary, pump and probe beams to our system in addition to the pump and probe of the setup in Fig. 4 from hereon the primary beams (see Fig. 5 & Fig. 7). The auxiliary beams are synthesized from a separate tunable laser source and chosen to excite phonons of angular frequency $\Omega_{\text{aux}}$, different than the primary phonon frequency, and propagating in the opposite di-
FIG. 5. a) Dissipation rate of the fundamental acoustic mode at low intensity as a function of temperature. b) Phonon dissipation rate as a function of temperatures below 6 K. The three data sets below 4 K correspond with different pump and probe power settings corresponding with low, moderate, and high phonon intensities, as compared to $J_c$. c) Phonon dissipation rate as a function of acoustic intensity. Each curve has a fixed temperature. d) Brillouin gain spectra as a function of temperature. e) Normalized Brillouin gain spectra as a function of intensity at 1.1 K. f) Comparison of the measured Brillouin frequency vs. temperature with the theoretical prediction given by Eq. [26]. g) Critical intensity $J_c$ and background losses $\Gamma_0$ as a function of temperature. The parameters for the theory can be found in Tab.1.

The primary and auxiliary optical beams were polarized orthogonally to one another.

Qualitatively, we expect that auxiliary phonons will saturate resonant absorption by TLSs in an intensity-dependent bandwidth, i.e. a ‘spectral hole’ in the phonon absorption, centered on $\Omega_{aux}$. As $J_{aux}$, the auxiliary phonon intensity, is increased well above the critical intensity we expect the spectral hole to increase in size (see Fig. 7).

To measure phononic spectral hole burning the dissipation rate of the primary phonon beam was measured as a function of frequency and intensity of the auxiliary phonon beam. Auxiliary pump wavelengths of 1537.963 nm, 1547.963 nm and 1552.963 nm, were used to generate auxiliary phonon beams with angular frequencies of $(2\pi)9259.8$ MHz, $(2\pi)9199.8$ MHz and $(2\pi)9170.8$ MHz respectively. Figure 7 shows the primary phonon dissipation rate as a function of the estimated $J_{aux}$. As we do not measure BGS for the auxiliary beam we have no measurement of the various parameters in Eq. [32] used to estimate the acoustic intensity. Instead, we approximate $J_{aux}$ by using the measured properties of the primary phonon beam. The data clearly demonstrate that a cross-saturating auxiliary phonon beam at high inten-
Acoustic dissipation imposed by defect states presents a barrier to attaining low loss acoustic modes that are critical to a range of technologies. To overcome/understand these challenges we explored defect-induced dissipation in a guided wave system using nonlinear phonon spectroscopy. Theory-experiment comparison using the tunneling state model quantifies the influence of defects in our system. We found that the large phonon lifetimes, made possible by the tight acoustic confinement, permit access to the TLS-induced regime of nonlinear phonon dynamics. Here we demonstrated two strategies for quashing phonon dissipation. The first is self-saturation where TLS resonant absorption is driven to transparency as the intensity of the acoustic beam becomes large. And, the second technique is cross-saturation, or spectral hole burning, where low dissipation for weak acoustic beams, i.e. those with low occupation number, can be achieved when the system is driven by an additional intense, spectrally-resolved, acoustic beam. Saturation of TLS losses with these techniques can result in dissipation rate shifts as large as $\frac{\pi P \gamma_p^2 Q_{\eta_i}}{\rho \nu_i^2} \approx \frac{44}{\Omega - 9\text{GHz}}$ MHz which when used in conjunction with cross-saturation provides a means to tune phonon lifetimes within a broad range.

At low temperatures defect scattering dominates the phonon dissipation rate as a result of the low thermal occupancy of the phonon modes we probe and the lack of anchor losses. At the highest intensities we encounter a dissipation floor $\Gamma_0$ due to relaxation absorption $\Gamma_{\text{rel}}$ and a roughly temperature independent offset $\Gamma_{\text{Background}}$. There are several possible explanations for $\Gamma_{\text{Background}}$ such as: inhomogeneous broadening due to irregularities in the fiber geometry along its length, acoustic leakage due to imperfect guiding, and Rayleigh scattering due to disorder and scattering by dopants. Large amounts of inhomogeneous broadening are unlikely given the relatively short length of the fiber and Lorentzian shape of the BGS, and acoustic leakage was estimated in simulations to be too small to explain $\Gamma_{\text{Background}}$. Rayleigh scattering, however is a likely explanation from the observed background as the fiber is doped with a large concentration of germanium that will produce large stochastic variations of the density and sound speed in the fiber core, and furthermore $\Gamma_{\text{Background}}$ is insensitive to temperature and intensity. For the lowest temperatures achieved in our experiment we estimate relaxation absorption to make up 6% of the dissipation rate floor. Hence, we believe we have reached the neighborhood of the smallest phonon dissipation rates possible in UHN3 fiber which we estimate to be near $(2\pi)650$ kHz. By working at lower frequencies (as Rayleigh scattering scales with the $\Omega^4$), or with fibers exhibiting less Rayleigh scattering much longer phonon lifetimes may be possible.

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FIG. 7. a) Illustration of optical and acoustic beams involved in phononic spectral hole burning. b) Cartoon of primary phonon absorption as a function of \( \Omega_{\text{aux}} - \Omega \) at three auxiliary phonon intensities. The auxiliary beam has a small influence on the primary beam absorption at low intensities. As \( J_{\text{aux}} \) is raised above \( J_c \), the primary phonon absorption is quashed in a range centered on the auxiliary phonon frequency which grows as \( J_{\text{aux}} \) is raised. The colored vertical lines represent the three \( \Omega_{\text{aux}} - \Omega \) settings used in our measurements. c) Linewidth of the primary phonon beam as a function of the estimated intensity of the auxiliary phonon beam. The auxiliary phonon frequencies used in our experiment: \{9199, 9199.9170\} (MHz) \( J_{\text{aux}} \) is estimated by using the known optical powers of the auxiliary beams and Eq. (32) where the gain and line width of the primary beam are used in place of the unknown auxiliary beam quantities. d) Illustration of the generation of the counter propagating primary and auxiliary acoustic waves and their resulting interference.

VI. APPENDIX

A. Derivation of TLS lifetime

In this appendix we derive the lifetime of the \( j \)th TLS using Fermi’s golden rule. We begin with matrix elements for upward transition of the TLS from the ground \( |g_j\rangle \) to the excited state \( |e_j\rangle \)

\[
\langle e_j, n_{q\eta} + 1|M_j : \xi(r_j)\sigma_{z,j}|g_j, n_{q\eta}\rangle = \frac{\hbar}{2\Omega_{q\eta}\rho V} e^{i\eta\mathbf{r}_j} \Delta_0 \sqrt{n_{q\eta}}
\]

where \( n_{q\eta} \) is the number of phonon quanta in the \( q\eta \)-mode. The matrix element for downward transitions is given by

\[
\langle g_j, n_{q\eta}|M_j : \xi(r_j)\sigma_{z,j}|e_j, n_{q\eta}\rangle = -\frac{\hbar}{2\Omega_{q\eta}\rho V} e^{-i\eta\mathbf{r}_j} \Delta_0 \sqrt{n_{q\eta} + 1}.
\]

After averaging over the initial (thermal) phonon state and summing over all final states that contribute to the two processes the golden rule gives the upper transition rate as

\[
C_{g\rightarrow e} = \sum_{\eta} \frac{\gamma_{\eta}^2 \Delta_{g,i}^2}{2\pi\rho V} \frac{1}{e^{\Delta_0 / kT} - 1} - 1
\]

where the Debye phonon density of states has been used \( g_{\eta}(E) = E^2/(2\pi^2\hbar^3v_0^3)\). The rate \( T_{e\rightarrow g} \) can similarly be computed and is identical to \( T_{g\rightarrow e} \) if the factor \( (\exp(E_j/kBT) - 1)^{-1} \) is taken to \( (\exp(E_j/kBT) - 1)^{-1} + 1 \).

These two rates can be combined to give the time rate of change for probability of the TLS to be in its excited state

\[
P_e = \frac{P_{e\rightarrow g} - P_{g\rightarrow e} + P_{e\rightarrow g} + P_{g\rightarrow e}}{\tau_e} = \left( C_{g\rightarrow e} + C_{e\rightarrow g} \right) P_e + T_{g\rightarrow e}
\]

since \( P_e + P_g = 1 \) which gives the decay rate for the excited state as \( C_{g\rightarrow e} + C_{e\rightarrow g} \) written explicitly in Eq. (33).
B. Derivation of the phonon lifetime via Fermi’s golden rule

In this section we derive the phonon decay rate for weak fields using the golden rule. The transition rates for an increase in the number of quanta in the $q_0$-phonon mode from $n_{q_0}$ to $n_{q_0} + 1$ as well as the decay rate from $n_{q_0}$ to $n_{q_0} - 1$ are given by

$$C_{n_{q_0} \rightarrow n_{q_0} + 1} = \frac{1}{V} \sum_j P_e \frac{\Delta^2 \gamma_j^2}{E_j} \frac{\pi q^2}{\Omega_{q_0}\rho} (n_{q_0} + 1) \delta(E_j - \hbar\Omega_{q_0})$$

$$C_{n_{q_0} \rightarrow n_{q_0} - 1} = \frac{1}{V} \sum_j P_g \frac{\Delta^2 \gamma_j^2}{E_j} \frac{\pi q^2}{\Omega_{q_0}\rho} n_{q_0} \delta(E_j - \hbar\Omega_{q_0}).$$

(39)

We employ the ergodic theorem to make the replacement $\sum_j \int d\Delta \int d\Delta_0 f(\Delta, \Delta_0)$. The integrals over $\Delta$ and $\Delta_0$ can be done giving the total rate of change of $n_{q_0}$

$$\dot{n}_{q_0} = \lambda[P_e (n_{q_0} + 1) - P_g n_{q_0}]$$

(40)

where $\lambda(P_e - P_g)$ is given by Eq. 14. The under-braces denote the terms contributing to; (i) stimulated phonon emission, (ii) spontaneous phonon emission, and (iii) stimulated phonon absorption. From the expression above it is clear that when $P_g \rightarrow 1$ that the TLSs predominantly attenuate the acoustic wave. Resonant absorption is suppressed at either high temperature where $P_e = P_g$, or high intensities where $n_{q_0} \gg 1$ and $P_e \rightarrow P_g$. However, the physics of the latter case can only be elucidated by working with the full dynamics governed by the Bloch equations.

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