

COHERENTLY STIMULATED BRILLOUIN SPECTROSCOPY

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ABSTRACT  
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This dissertation explores novel ways in which light and sound interact in optical waveguides to enable laser cooling of traveling-wave phonons and to enhance phonon spectroscopy. First, we establish how anti-Stokes Brillouin scattering can be harnessed to cool acoustic excitations in a continuous fiber, extending laser cooling concepts beyond discrete cavity optomechanical systems. By using a liquid-core optical fiber (LCOF) filled with  $\text{CS}_2$ , we achieve significant reduction in the phonon population through spontaneous anti-Stokes processes. This demonstration marks the first instance of in-fiber traveling-wave phonon cooling.

Second, we introduce a Coherently Stimulated Brillouin Spectrometer (CoBS) that employs a four-wave mixing scheme comprising pump, probe, driven Stokes, and observed signal fields to dramatically improve sensitivity, especially for short, low-gain samples. By relaxing traditional phase-matching constraints, the CoBS approach enables a  $10^6$  increase in scattered power over mm- to cm-scale lengths and demonstrates sub- $10\text{ fW}$  sensitivity. We validate its utility with measurements in both high- and low-gain media, opening new possibilities for ultrathin-film and microfluidic analyses.

Lastly, this dissertation investigates the theoretical and experimental conditions under which ambient Brillouin-driven Raman-like modes might be observed. These Brillouin-induced Raman modes could provide a bridge between conventional Raman spectroscopy and the acousto-optic phenomena characteristic of traveling-wave phonons. While not yet fully realized, the groundwork here points to future avenues in which engineering the geometric and material properties of optical waveguides can yield both net phonon cooling and advanced hybrid phononic devices. Collectively, these results broaden our understanding of optomechanical coupling in fibers and pave the way for low-noise photonic systems, quantum acoustic memory elements, and highly sensitive, broadband spectroscopic tools.

*Science is temporal arbitrage; invest in it.*

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## List of Acronyms

**AC** alternating current

**AOM** acousto-optic modulator

**AMO** atomic, molecular, and optical

**BPF** bandpass filter

**BYU** Brigham Young University

**C** Cooling metric

**CARS** Coherently stimulated Anti-Stokes Raman Scattering

**CINT** Center for Integrated Nanotechnologies

**CoBS** Coherently stimulated Brillouin Spectrometer

**CW** Continuous Wave

**DC** direct current

**EDFA** Erbium-Doped Fiber Amplifier

**FBG** fiber Bragg grating

**FP** fiber port

**FWHM** full-width half-max

**IM** fiber-optic intensity modulator

**LANL** Los Alamos National Laboratory

**LCOF** liquid-core optical fiber

**LO** local oscillator

**ORNL** Oak Ridge National Laboratory

**PC** polarization controller

**PCF** photonic crystal fiber

**PBS** polarizing beam splitter

**PVD** physical vapor deposition

**RBW** resolution bandwidth

**RF** radio frequency

**RFSA** radio-frequency spectrum analyzer

**SNL** Sandia National Laboratory

**SBS** Stimulated Brillouin Scattering

**SMF-28** single mode fiber 28

**SNR** signal-to-noise ratio

**UHNA3** Ultra High Numerical Aperture 3

**UHNA7** Ultra High Numerical Aperture 7

**VOA** Variable Optical Attenuator

**WGM** whispering-gallery-mode

# Chapter 1

## Introduction

Optomechanics is the study of light-matter interactions; it is the study of how the intangible (light) can affect change in the tangible (matter) and vice versa. Injecting light into a material under specific conditions allows for an exchange of energy to occur between the light and the mechanical oscillations of the material which leaves the mechanical energy of the material altered. This interaction can be controlled to deposit or withdraw mechanical energy into/from a system and thus leave the system in a more, or less, mechanically energetic state respectively. The same interaction can also be harnessed for passive observation of material properties. Mechanical systems from bulk to atomic scales can be probed and characterized by light through retrieval of the inelastically scattered light resulting from interaction with the material. This retrieved light contains embedded information about the energy exchange that occurred, which, when considered as part of a population of scattering events, reveals natural resonances of a mechanical system.

Optomechanics comprises a broad range of phenomena involving the interaction of optical and mechanical systems, from basic photothermal absorption to more complex nonlinear processes. This section offers a brief overview of notable optomechanical phenomena. Photothermal absorption is the process by which light is absorbed by a material, leading to an increase in temperature of the material and consequent changes in the material's dimensions (thermal expansion) or refractive index (thermo-optic effect). This effect has applications in optical switches, actuators, and sensors.<sup>1, 2</sup> Photothermal therapy in medicine is an emerging application of this effect, where light is used to target and heat specific areas, causing localized damage to diseased tissue<sup>3</sup>. This technique becomes especially effective when combined with nanoparticle-enhanced absorption, allowing for dramatically increased absorption in ultra-localized zones within the body.<sup>4</sup>

Light scattering, in its many forms, is also an optomechanical process as it involves the interaction of an optical field with the fluctuation, motion, or vibration of matter. Rayleigh scattering, perhaps the most well-known example, is the elastic scattering of light by particles much smaller than the wavelength of the incident light, leading to scattering in possibly a new direction but without a change in wavelength. It is responsible for the blue color of the sky because the efficiency of Rayleigh scattering is inversely proportional

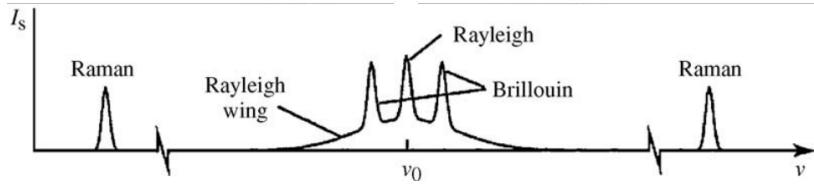
to the fourth power of the wavelength ( $\lambda$ ) of the light ( $\frac{1}{\lambda^4}$ ) and so shorter (blue) wavelengths are scattered much more than longer (red) wavelengths by the molecules in the atmosphere.<sup>5</sup>

Raman scattering is the interaction of light with vibrational and rotational modes within a material (often molecular), resulting in scattered light with frequencies that are shifted from the incident light. This inelastically scattered light provides insights into the material's molecular structure and properties. Raman scattering is widely used in chemical and material science for identifying chemical compounds, analyzing molecular structures, and studying molecular dynamics. It finds application in the characterization of pharmaceuticals, monitoring changes in biological tissues for medical diagnostics, and investigation of stress and temperature distributions in engineering materials, among others.<sup>6, 7</sup>

Brillouin scattering, around which much of the work detailed in this document is centered, is the scattering of light with acoustic phonons or coherent traveling density waves in a material, resulting in scattered light with a frequency that is slightly shifted from the incident light. This inelastically scattered light reveals mechanical properties of the material such as its bulk and elastic moduli. This phenomenon is used in materials science to measure elastic properties and viscoelasticity of materials, in fiber optic sensing to monitor temperature and strain over large distances, and in physics to study phase transitions and mechanical properties of crystals, liquids, and gases.<sup>8–10</sup>

Rayleigh-wing scattering is the broad, smooth extension of the Rayleigh scattering spectrum that results from interactions with low-frequency excitations in a material, providing insights into dynamic processes like rotational and translational diffusion of molecules that make up a material. This scattering is particularly useful in studying the dynamics of complex fluids, gases, and soft materials, where it can reveal information about molecular orientation, diffusion rates, and interactions within the medium. Applications include the analysis of atmospheric phenomena, characterization of liquid crystals, and investigations into the properties of polymers and biological materials, aiding in the understanding of their behavior at the molecular level.<sup>11, 12</sup>

Figure 1.1 shows the relative domains of typical frequency shifts for Rayleigh, Rayleigh-wing, Brillouin, and Raman scattering. Rayleigh-wing scattering is broad and shares part of its domain with Brillouin scattering. This makes sense because for any given molecule and within the timescale that it occurs, diffusive translational motion can be thought of as indistinguishable from motion caused by traveling density waves that host Brillouin scattering. In this way, Rayleigh-wing scattering represents a sporadic distribution of fleeting, localized Brillouin scattering. Of course, the difference between incoherent diffusion of molecules and coherently traveling acoustic modes within a material is an important distinction. However, this thought experiment offers a perspective for bridging the gap between Rayleigh-wing and Brillouin scattering and for understanding their common frequency domains. Moreover, it serves as a reminder of the rich continuum of



**Figure 1.1:** Relative domains of typical frequency shifts for Rayleigh, Rayleigh-wing, Brillouin, and Raman scattering. Figure adapted from Boyd Nonlinear Optics (2020).<sup>13</sup>

material behavior and responses that affect light scattering as opposed to the distinct categories we ascribe for convenience. This is a core concept of the research described in further chapters.

Returning to other optomechanical phenomena beyond scattering processes, the momentum of photons can exert forces on objects, leading to phenomena like radiation pressure, optical tweezing, and optical trapping. These effects are widely used in manipulating microscopic particles, biological cells, and atoms, enabling studies of single molecules, cold atoms, and quantum computing elements.<sup>14–16</sup>

The final category of optomechanical interactions to be noted here is that of nonlinear optical phenomena. Second harmonic generation, parametric oscillation, and four-wave mixing all feature the interaction between light and material nonlinearities that lead to the generation of new light frequencies.<sup>13</sup> The Kerr effect is the change in the refractive index of a material in response to an applied electric field, which can be induced optically with sufficient intensities of light. In general, nonlinear optical responses of materials are often only accessible with the use of high intensity laser light. This is emphasized by the fact that the field of nonlinear optics can be traced back to the discovery of second-harmonic generation in 1961<sup>17</sup>, just one year after the first demonstration of the laser by American physicist Theodor Maiman.<sup>18</sup> These nonlinear effects provide the foundation for a range of technologies, including high-speed optical communication systems, frequency converters, and lasers for materials processing.<sup>17, 19, 20</sup> Also included in nonlinear optical phenomena is electrostriction. Electrostriction is a reversible material deformation induced by an electric field, which can be generated by light in electro-optic materials. This effect is quadratic, scaling with the square of the applied electric field, and hence a nonlinear optical effect. At sufficiently high intensities, electrostrictive forces serve to enhance Brillouin scattering whereby the scattered light electrostrictively reinforces the acoustic wave that caused its scattering, leading to a nonlinear positive feedback loop known as Stimulated Brillouin Scattering (SBS). Photostriction is a related phenomenon that occurs when light absorption causes a change in the lattice structure of a material, leading to mechanical strain. It combines photovoltaic and piezoelectric effects and can be seen as optically induced strain. These effects are utilized in designing optical modulators, tunable photonic devices, and smart materials that respond to light.<sup>21, 22</sup>

This dissertation is organized into three core chapters, each centered on harnessing traveling-wave phonon

dynamics through Brillouin scattering. Chapter 2 presents the first experimental demonstration of laser cooling of a continuum of traveling acoustic phonons in a macroscopic medium via anti-Stokes Brillouin scattering. By leveraging the strong acousto-optic coupling in a liquid fiber core, we observe a power-dependent suppression of thermal phonons corresponding to a  $>20\text{ K}$  reduction of the acoustic mode's effective temperature. This result extends the paradigm of continuous-mode optomechanical cooling, previously only achieved in chip-scale waveguides, to a meter-scale fiber system, highlighting the universality and scalability of traveling-wave phonon manipulation. Chapter 3 introduces an ultra-sensitive four-wave-mixing phonon spectrometer (CoBS), which we developed to probe Brillouin interactions with unprecedented sensitivity. The Coherently stimulated Brillouin Spectrometer (CoBS) instrument uses coherent pump–probe stimulation to amplify phonon signals, achieving fW detection limits that enable Brillouin scattering measurements in extremely short interaction lengths. Producing  $\sim 10^6$  more scattered power than conventional Brillouin scattering techniques, this advanced methodology provides a new window into phonon spectra and dynamics, laying the experimental groundwork for observing extremely subtle phononic phenomena. Chapter 4 explores the emergence of Raman-like phonon modes induced by traveling-wave Brillouin interactions across a range of material platforms. By systematically shrinking the acoustic path length (from meters to sub-millimeter scales) and enhancing phonon reflectivity in media including doped optical fibers, bulk liquids, thin films of tellurium dioxide, and integrated waveguides, we probe the crossover between the continuous Brillouin regime and discrete standing-wave vibrational modes. These experiments approach the regime in which traveling acoustic waves “freeze” into bounded modes; where traveling-wave Brillouin scattering morphs into Raman-like phonon resonances. This pursuit effectively unifies the Brillouin (traveling-wave) and Raman (standing-wave) paradigms of light–sound interaction, providing new opportunities in cavity-free phononics and coherent phonon devices at room temperature. Taken together, the work and results presented here underscore traveling-wave Brillouin scattering as a versatile tool for phonon manipulation, from optical cooling of phonon populations to high-precision phonon spectroscopy and the generation of novel phononic modes at room temperature. Brillouin-mediated light–sound coupling enables these diverse advances and links them to the broader context of hybrid photonic–phononic technologies. By demonstrating control over traveling phonons in continuous media, developing instrumentation to detect minute phonon signals, and pushing toward the Brillouin–Raman convergence, this work contributes to emerging frontiers in both classical and quantum acoustics. In particular, this work suggests new avenues for acousto-optic devices with reduced noise (through phonon cooling) and lay groundwork for quantum acoustic control in room-temperature systems. The subsequent chapters detail these contributions, collectively advancing the goal of manipulating sound waves with light in unprecedented ways.





## Chapter 2

### Laser Cooling of Traveling-Wave Phonons in an Optical Fiber

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*This chapter elaborates on experiments and results related to the demonstration of optomechanical cooling of traveling-wave phonons in an optical fiber, which have been published as an article by the same name in Physical Review Applied by the author (Johnson et al. 2023).<sup>23</sup> Any discrepancies, omissions, or errors that may exist between the published paper and this dissertation chapter are the sole responsibility of the author, as the text, analyses, and interpretations herein represent an independent and original presentation of the work.*

#### 2.1 Introduction

Over the past half-century, light-based techniques have dramatically reshaped how we control mechanical motion. Laser cooling has become indispensable across many branches of physics, allowing researchers to reduce thermal motion in ions, atoms, and mesoscopic oscillators to previously unimaginable levels. From the earliest proposals and demonstrations of Doppler cooling in dilute gases<sup>24</sup> to the realization of ultracold quantum gases in Bose–Einstein condensates,<sup>25</sup> the ability to reduce thermal motion with light has profoundly transformed atomic, molecular, and optical (AMO) physics, enabling breakthroughs ranging from high-precision atomic clocks to quantum state preparation.<sup>26</sup> Equally impactful is the extension of laser cooling concepts into solid-state systems. In rare-earth-doped optical crystals, for example, anti-Stokes fluorescence cooling has now approached cryogenic temperatures, cooling Yb-doped solids down to  $\sim 87\text{ K}$

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(the boiling point of liquid argon) and projected to reach 77 K (liquid nitrogen) in the near future.<sup>27</sup> Such optical refrigeration offers a vibration-free route to cryocoolers and has steadily advanced since the first demonstration in 1995.<sup>28</sup> Meanwhile, in mesoscopic mechanics, radiation-pressure laser cooling of solid-state oscillators has opened new frontiers. By coupling light to discrete mechanical modes in optical or microwave cavities, researchers can damp mechanical vibrations to the point of quantum ground-state occupancy.<sup>29</sup> Milestone experiments achieved sub-single-phonon populations in nanomechanical resonators cooled by laser light<sup>29</sup>, laying the groundwork for quantum metrology and hybrid quantum devices. From AMO systems to solid-state platforms, laser cooling has become a cornerstone technique for reducing thermal noise and accessing novel quantum regimes.

Given this context, there is intense interest in applying laser cooling to optomechanical systems, where light interacts with vibrational modes of a material. The established paradigm in optomechanics involves discrete mechanical oscillators coupled to optical cavities.<sup>30</sup> In a typical cavity-optomechanical system (e.g., a tiny mirror or membrane attached to a cavity) the radiation pressure force provides a parametric coupling between an optical resonance and a mechanical mode.<sup>30</sup> By tuning a laser slightly red of a cavity resonance (i.e. the anti-Stokes sideband), photons can scatter inelastically from the mechanical vibration, gain energy (up-shift) by absorbing a phonon, and promptly leave the cavity, carrying away thermal energy. This sideband cooling process leads to a reduced phonon population in the mechanical mode, provided the anti-Stokes scattering dominates over the Stokes (down-shift) process that would add phonons. Using this approach, a variety of mechanical oscillators from gram-scale mirrors to nanoscale beams have been cooled near their ground state of motion.<sup>29</sup> A landmark example is the laser cooling of a 3.7 GHz silicon nanobeam mode to an average occupancy of  $\langle n \rangle \approx 0.85$  (15% of one quantum) starting from 20 K environment.<sup>29</sup> Such achievements, along with comprehensive theoretical frameworks,<sup>30</sup> firmly established the state-of-the-art: *discrete* mechanical modes can be laser-cooled via engineered light-matter interactions in optical cavities.

Early optomechanical cooling experiments focused on isolated resonant modes, but recent work has pushed toward cooling a *continuum* of vibrational degrees of freedom, in particular, traveling-wave phonons in extended media. One route to bridging this gap is to use whispering-gallery-mode (WGM) microresonators, which support both localized optical modes and traveling acoustic waves confined around the periphery. In 2012, Bahl et al. demonstrated the first observation of Brillouin optomechanical cooling in a silica microcavity.<sup>31</sup> In that system, light circulating in the WGM resonator underwent spontaneous Brillouin scattering with an acoustic whispering-gallery wave. Because the scattering was in the forward direction (co-propagating light and sound), it accessed a low-frequency acoustic mode that had much lower damping than the usual high-frequency phonons of spontaneous backscattering. Satisfying this low acoustic dissipation condition (achieving phonon lifetimes longer than the optical storage time) allowed the optical field to cool

the mechanical mode rather than amplify it. The experiment revealed two regimes: the normal Stokes process where light amplifies acoustic vibrations, and a Brillouin cooling regime where anti-Stokes scattering dominantly *damps* the acoustic motion. This result was remarkable since Brillouin interactions in bulk or fiber had long been known primarily as a heating (Stokes) process; the microresonator provided the multi-mode, low-loss environment needed to tip the balance towards net cooling.

Even more surprising was the recent demonstration that one can cool a continuum of phonon modes in a completely non-resonant, traveling-wave configuration.<sup>32</sup> Otterstrom et al. (2018) showed for the first time that laser cooling is possible in a continuous optomechanical waveguide, without any optical cavity or discrete mechanical resonance. In their experiment, light propagating through a 2.3 cm silicon photonic waveguide was observed to reduce the thermal occupancy of a band of acoustic phonons via Brillouin scattering. The key was to leverage intermodal Brillouin scattering, wherein the pump laser in one waveguide mode scatters to an anti-Stokes photon in a different optical mode while annihilating a phonon. This phase-matched anti-Stokes process selectively damped those phonons, cooling a spectral band by over 30 K relative to room temperature. Conceptually, the continuous-waveguide regime has an inherent advantage: the Stokes (phonon-creating) and anti-Stokes (phonon-annihilating) processes occur on different phonon modes in an extended medium. Thus, any unavoidable Stokes scattering does not re-excite the very phonon mode being cooled, avoiding the direct competition that limits cooling of a single isolated mode. Still, achieving cooling in a traveling-wave system demands stringent conditions. The optical wave must interact sufficiently strongly with the acoustic waves (high acousto-optic coupling), and the phonons must remain dissipated long enough for the cooling scattering to outpace thermal re-population and for the light to exit the system carrying that stolen mechanical energy with it. In the silicon waveguide used by Otterstrom et al., engineered cross-section geometry provided large Brillouin gain, and the small device length ensured that anti-Stokes photons escaped the system faster than the phonons could dissipate, fulfilling these requirements.<sup>32</sup> This demonstration has spurred growing interest in continuous optomechanical cooling, including new theoretical proposals to dynamically control phonon baths in waveguide systems.<sup>33</sup>

*Laser cooling of traveling-wave phonons in an optical fiber* is a natural next goal in this progression. Optical fibers represent the quintessential one-dimensional continuous medium for light and sound, with applications ranging from fiber lasers to quantum information transfer. Cooling phonons in fiber could, for example, suppress thermally driven noise (e.g. guided-acoustic-wave Brillouin noise) that limits the frequency stability of fiber lasers and squeezes light in fiber interferometers.<sup>34</sup> It could also enable new in-fiber acousto-optic devices with reduced thermal noise, or even allow one to prepare traveling phonon wavepackets in low-entropy states for quantum phononics. However, extending optomechanical cooling to standard fibers poses several distinct challenges that were absent in chip-scale or microresonator systems. First, conventional

single-mode fibers have relatively weak light–sound coupling. The Brillouin gain coefficient  $g_B$  in fused silica fiber is on the order of  $5 \times 10^{-11} \text{ m W}^{-1}$ ,<sup>35, 36</sup> many orders of magnitude smaller than in highly confinement-enhanced waveguides (such as silicon nanowires or resonant structures). This means achieving appreciable anti-Stokes scattering in a fiber typically requires either very long interaction lengths or high optical power.

Secondly, the acoustic modes in a fiber have finite lifetimes that can be quite short, especially for the high-frequency phonons usually involved in Brillouin scattering. In backwards spontaneous Brillouin scattering with a  $1.5 \mu\text{m}$  pump, these phonons are typically  $\sim 9\text{--}12 \text{ GHz}$  sound waves in silica, which experience significant acoustic damping (linewidths of tens of MHz) and thus live only on the order of nanoseconds.<sup>37</sup> Such brief phonon lifetimes make it difficult to achieve net cooling of a phonon mode: the anti-Stokes process must remove phonons faster than they are thermally replenished. Essentially, one needs phonon  $Q$ - (quality) factors high enough that the phonon lifetime exceeds the transit time of light through the interaction region. Meeting this condition in a meter-scale fiber (where light transit is only a few nanoseconds) is non-trivial. Prior experiments in resonators or short waveguides addressed this by using lower-frequency acoustic modes with inherently longer lifetimes<sup>31</sup> or by effectively shortening the optical interaction length, but in a long fiber the default acoustic damping is too strong to allow cooling in the usual Brillouin regime.

These challenges help explain why, until now, laser cooling of phonons had not been realized in a fiber; no prior studies have achieved phonon cooling in an optical fiber. This chapter details how we overcome these obstacles and presents the results we achieved. By using a specially engineered liquid-core fiber platform, we satisfy the acousto-optic coupling and phonon dissipation requirements and achieve over  $20 \text{ K}$  of cooling in an optical fiber with modest pump power. This marks the first demonstration of laser cooling of a continuum of phonons in fiber and extends the reach of optomechanical cooling to macroscopic length scales. Through this work, we bridge the gap between chip-scale optomechanics and fiber optics, enabling new low-noise acousto-optic technologies and fundamental studies of traveling-wave phonons in the quantum regime.

## 2.2 Optomechanical Cooling

### 2.2.1 Physical Mechanism

Optomechanical laser cooling can be viewed as a coupling between a discrete or traveling-wave phonon mode and an external reservoir capable of removing vibrational energy. When light interacts with sound through anti-Stokes scattering (where photons are upshifted in frequency by absorbing phonons), the optical field acts as an effective “damping channel,” allowing mechanical excitations to exit the system in the form of higher-energy scattered photons. This additional pathway for phonon annihilation increases the net decay rate of the targeted mechanical mode, thereby reducing its occupation number below the thermal equilibrium

level. Conceptually, one can think of this interaction much like placing a fingertip on a vibrating bell: the bell (analogous to the mechanical mode) is no longer isolated but is now coupled to a broader set of mechanical modes in one’s hand, enabling rapid dissipation of vibrational energy into the surroundings. In laser cooling, the “finger” is replaced by the optical continuum associated with the anti-Stokes scattering process, which carries away phonon energy in the form of upshifted photons rather than mechanical vibrations.

This coupling arises from momentum and energy conservation between the pump photon at frequency  $\omega$ , the phonon at frequency  $\Omega$ , and the scattered photon at  $\omega + \Omega$ . Although the pump itself may be extremely narrow in linewidth, its interaction with the phonon mode broadens the resulting scattered light analogous to how a short-lived mechanical oscillator necessarily exhibits a broader frequency spectrum. The faster the phonon mode decays via anti-Stokes scattering, the more rapidly the mechanical amplitude (phonon occupation) drops, and thus the “effective” linewidth of that mode broadens. In other words, the very mechanism that damps the phonon occupation also manifests as a broadened distribution of scattered photon frequencies centered at  $\omega + \Omega$ . This is again analogous to the finger placed on the vibrating bell: the mechanical-mechanical coupling of the finger to the bell damps the bell’s vibration by offering a broad continuum of mechanical modes (a mechanical bath) for the bell’s vibration to dissipate into. Left behind in the finger is a broad distribution of mechanical modes, with the linewidth of this mechanical frequency distribution corresponding to the rate of damping (dissipation rate). Likewise, the applied optical field (laser) *opto*-mechanically couples to the phonon mode, lowering the phonon occupancy of the mechanical mode by offering a broad continuum of *optical* modes (a “light bath”) for the phonons to dissipate into. Produced in the scattered light is a broad distribution of *optical* modes, with the linewidth of this optical frequency distribution indicating the dissipation rate of the phonon mode.

Importantly, this does not require the intrinsic mechanical properties of the medium to change. Instead, once the optical field is present, the formerly isolated mechanical mode sees a new “escape route” for its energy. In a parametric picture, the simultaneous conservation of energy and momentum among the three waves enables phonons to be converted into higher-frequency photons that promptly leave the system, carrying away the vibrational quanta. The result is an enhanced net decay rate: intrinsic mechanical damping of the host medium plus the new optical damping channel coupled to the system. If these processes occur faster than the phonons can be re-supplied by thermal fluctuations (the natural dissipation rate of the medium), the phonon population is driven to a lower effective temperature.

While anti-Stokes scattering provides the mechanism for laser cooling, the inverse Stokes heating process simultaneously occurs. Spontaneous backward Brillouin scattering interacts with longitudinally traveling acoustic waves (phonons) according to both of these complementary processes, but the specific phonon mode is distinct for each. Figures 2.1a and 2.1b offer respective illustrations of these Stokes and anti-

Stokes scattering processes. In the Stokes process, an incident pump photon scatters with a *retreating* phonon, annihilating the *photon* and creating both an additional phonon and a backscattered photon at the difference energy. Both energy and momentum must be conserved for this process, demanding these Stokes components satisfy

$$\omega_S = \omega_P - \Omega, \quad -k_S = k_P + q, \quad (2.1)$$

where  $\omega_S$ ,  $\omega_P$ , and  $\Omega$  are the angular frequencies of the Stokes, pump, and phonon waves, respectively, and  $k_S$ ,  $k_P$ , and  $q$  are their corresponding wavevectors. This can be understood by analogy of the incident light experiencing a doppler *down-shift* in frequency as though the photon were reflected from a *retreating* mirror (phonon). The Stokes processes results in increased phonon occupancy of the respective longitudinal mode of the material, and thus this process results in optomechanical heating. Optical energy is donated to the material in the form of mechanical (thermal) energy.

The anti-Stokes process is the inverse process (Figure 2.1b), whereby an *approaching* phonon scatters with a pump photon, annihilating the *phonon* and creating a backscattered photon at the additive energy. In the anti-Stokes process, the incident light experiences a doppler *up-shift* in frequency as if, to continue the analogy, the photon were reflected from an *approaching* mirror (phonon). Phase matching requirements demand the anti-Stokes components satisfy

$$\omega_{aS} = \omega_P + \Omega, \quad -k_{aS} = k_P - q, \quad (2.2)$$

where  $\omega_{aS}$  and  $k_{aS}$  are the respective angular frequency and wavevector of the backscattered anti-Stokes light.

### 2.2.2 Spectral Signatures of Cooling and Heating

Optomechanical cooling reduces the phonon occupation of a given mechanical mode. Evidence of this phenomenon is offered by changes in two key spectral features of the backscattered light: linewidth and amplitude. Spectral linewidth is a measure of the mechanical dissipation rate of the phonon mode. A broader linewidth corresponds with a faster mechanical dissipation rate of a given mode as per the time-energy uncertainty principle: greater dissipation means shorter phonon lifetime, which introduces an uncertainty in the phonon frequency. Thereby, increasing the dissipation rate (cooling) broadens the spectral linewidth; con-



**Figure 2.1:** Illustration of optomechanical heating and cooling processes. Figure 2.1a shows an incident photon of frequency  $\omega$  scattering with a retreating phonon of frequency  $\Omega$ , resulting in the annihilation of the incident photon and the creation of both an additional retreating phonon of frequency  $\Omega$  and a backwards propagating photon down-shifted in frequency ( $\omega_{\text{Stokes}} = \omega - \Omega$ ). Figure 2.1b shows the inverse process, whereby an incident photon,  $\omega$ , scatters with an approaching phonon,  $\Omega$ , annihilating the incident photon and the phonon to produce a backwards propagating photon up-shifted in frequency ( $\omega_{\text{anti-Stokes}} = \omega + \Omega$ ).

versely, decreasing the dissipation rate (heating) narrows the spectral linewidth. For spontaneous Brillouin scattering this is given by<sup>32</sup>

$$\Gamma_{\pm} \approx \Gamma_0 \left( 1 \pm \frac{G}{4} \right), \quad (2.3)$$

where  $\Gamma_0$  is the mechanical dissipation rate at thermal equilibrium,  $\Gamma_{\pm}$  is the effectively broadened (+) or narrowed (-) dissipation rate due to an anti-Stokes or Stokes process, respectively. Here,  $G$  is the Brilouin scattering process gain factor  $G = G_B P_P L$ , where  $G_B$  is the effective Brilouin gain ( $\text{W}^{-1} \text{m}^{-1}$ ),  $P_P$  is the pump power, and  $L$  is the effective length (see Section B.2 in Appendix B). Equation 2.3 is related to the effective temperature change of the mechanical mode via the quantized phonon dissipation rate (phonons dissipated per second) at equilibrium, given by

$$Q = n_{\text{th}} \Gamma_0, \quad (2.4)$$

where  $\Gamma_0$  is the mechanical dissipation rate at thermal equilibrium and  $n_{\text{th}}$  is the mean thermal occupancy (phonon count) of the mode at equilibrium given by the Bose-Einstein distribution, or the Planck distribution function for phonons,<sup>38</sup>

$$n_{\text{th}} = \frac{1}{e^{\frac{\hbar\Omega_B}{k_B T_0}} - 1}. \quad (2.5)$$

Here,  $\hbar = h/2\pi$  is the reduced Planck constant,  $\Omega_B$  is the angular phonon frequency,  $k_B$  is the Boltzmann constant, and  $T_0$  is equilibrium (room) temperature in kelvin. Laser cooling of the phonon mode increases the effective dissipation rate of the mode by offering a new escape channel by which phonons may exit the mode (converting to the optical domain). This altered dissipation rate gives the effective temperature via Equation 2.4,

$$n_{\text{eff}}\Gamma_{\pm} \approx n_{\text{th}}\Gamma_0, \quad (2.6)$$

where  $n_{\text{eff}}$  is the effective phonon mode occupancy of the cooled (+, widened dissipation rate) or heated (-, narrowed dissipation rate). For GHz phonon frequencies near room temperature,  $k_B T \gg \hbar\Omega_B$ , allowing the first order approximation

$$e^{\frac{\hbar\Omega_B}{k_B T}} \approx 1 + \frac{\hbar\Omega_B}{k_B T}, \quad (2.7)$$

which gives, for Equation 2.5,

$$n_{\text{th}} \approx \frac{k_B T_0}{\hbar\Omega_B}. \quad (2.8)$$

Inserting this approximation in Equation 2.6, and in combination with Equation 2.3, we find two independent approximations for the ratio of effective to equilibrium temperatures of the cooled phonon mode,

$$\frac{\Gamma_+}{\Gamma_0} \approx \frac{T_0}{T_{\text{eff}}} \approx 1 + \frac{G}{4}. \quad (2.9)$$

Rearranging for fractional degrees cooled ( $\Delta T/T_0$ ), where  $\Delta T = T_0 - T_{\text{eff}}$  is degrees cooled from equilibrium, we find (to first order approximation)

$$\frac{\Gamma_+}{\Gamma_0} - 1 \approx \frac{\Delta T}{T_0} \approx \frac{G}{G + 4}. \quad (2.10)$$

Spectral amplitude provides additional evidence of optomechanical cooling. Spectral amplitude is a measure of the scattered power produced from pump light scattering with phonons occupying a given mechanical mode. Higher phonon occupancy within a given mode provides a higher rate of scattering and thus a higher measured spectral amplitude of the backscattered light. Increasing pump power reduces the phonon population occupying the anti-Stokes mode and thereby lessens the otherwise scattered power, measured as spectral amplitude. The inverse process occurs for the Stokes process; increasing pump power increases the phonon population occupying the Stokes mode and thereby increases the otherwise scattered power, measured from spectral amplitude.

In the spontaneous Brillouin scattering process, however, scattered power increases not only with phonon population (linked to temperature via Equation 2.5), but also with pump power  $P_P$  (see Equation B.50 in Appendix B.2). As a result, both the Stokes and anti-Stokes spectral amplitudes increase with increasing pump power. Despite this, however, spectral amplitudes still provide evidence of laser cooling. Asymmetric growth rates of the relative Stokes and anti-Stokes spectral amplitudes with increasing pump power indicate underlying cooling of the anti-Stokes phonon mode relative to the Stokes. The anti-Stokes scattering process (phonon annihilation) leads to a sublinear growth trend in peak spectral amplitude even as increasing pump power raises the overall peak amplitude. Conversely, the Stokes scattering process (phonon creating) leads to a superlinear growth trend in peak spectral amplitudes as pump power raises. These divergent trends in peak spectral amplitudes are a direct result of the optomechanically-induced altering of the thermal state of the respective modes and provide key evidence of laser cooling.

To expand on this, one might imagine a process whereby a pump laser cools a medium through the anti-Stokes process while an independent probe laser is simultaneously co-injected and allowed to scatter with the same phonon mode being cooled by the pump. In this pump-probe arrangement, increasing pump power induces further cooling of the anti-Stokes mode just as before, but with the addition of a probe that may be held at constant power. Critically, while backscattered *pump* light would still feature an overall growth trend in peak spectral amplitude (proportional to pump power), scattered power of the *probe* would only be sensitive to the underlying phonon occupancy (temperature) of the mode. As such, for the anti-Stokes (cooling) process, peak amplitudes of the probe spectra would actually be seen to *decrease* with increasing pump power, commensurate with the mode being increasingly cooled by the pump. As such, this pump-probe spectroscopy regime offers a direct measure of phonon occupancy, and thus direct and independent confirmation of laser cooling occurring.

## 2.3 Methods

While spontaneous Brillouin scattering processes naturally alter phonon populations (and thus mode temperatures), practical demonstration and detection of these effects pose significant challenges. Foremost among these is the requirement to remain in the spontaneous regime: we wish to probe the natural thermal phonons in a medium, so we cannot rely on artificially driving the mechanical modes to enhance scattered power (see Appendix B.2, and specifically Figure B.1, for a comparison of scattered power produced by different Brillouin techniques). Although stimulated Brillouin scattering (SBS) is often employed to boost signal levels, it actively drives phonon populations via injected optical fields, and thus no longer measures the intrinsic thermal phonons. In practical terms, SBS occurs when the overall process gain factor  $G = G_B P_P L$  (where  $G_B$  is the effective Brillouin gain,  $P_P$  the pump power, and  $L$  the effective length) far exceeds unity ( $G \gg 1$ ).

To demonstrate optomechanical cooling (i.e., an anti-Stokes process that lowers phonon energy), one must detect scattered light from a mode whose phonon population has been reduced. This is intrinsically difficult because a decreased phonon population means fewer scattering events, and hence diminished backscattered light. Consequently, an ideal testbed should provide sufficiently high single-pass gain (overall gain factor near unity) to enable clear detection, yet remain below the threshold that would drive the process into the stimulated regime. Although the Stokes and anti-Stokes processes address distinct phonon modes in a traveling-wave system, allowing any mode to become stimulated generates large driven phonon populations that can obscure or counteract spontaneous cooling. Achieving this balance ensures that measurements reflect genuine spontaneous cooling of a thermally populated phonon mode, rather than an artifact of optically driven phonons.

In addition to the requirement that the overall process gain be near but below unity, temporal constraints impose further critical conditions. Specifically, the rate at which phonons are removed from the system must exceed the rate at which they are replenished by the thermal bath to ensure net cooling of the anti-Stokes mode. This condition demands that backscattered photons leave the system rapidly, carrying away the extracted mechanical energy. A mean-field analysis (see Appendix A of Johnson et al. 2023<sup>23</sup>) shows that the relevant depletion rate is  $4v_g/L$ , where  $v_g$  is the group velocity of the anti-Stokes light and  $L$  is the system length. This must exceed the mechanical dissipation rate  $\Gamma_0$ , which represents the natural return of the phonon mode to thermal equilibrium. Hence, a suitable platform for demonstrating optomechanical cooling of traveling-wave phonons must fulfill the fast escape condition  $4v_g/L > \Gamma_0$ . Meeting this requirement on system length, however, directly conflicts with the desire for a sufficiently high overall gain factor, illustrating the delicate balance needed for effective cooling.

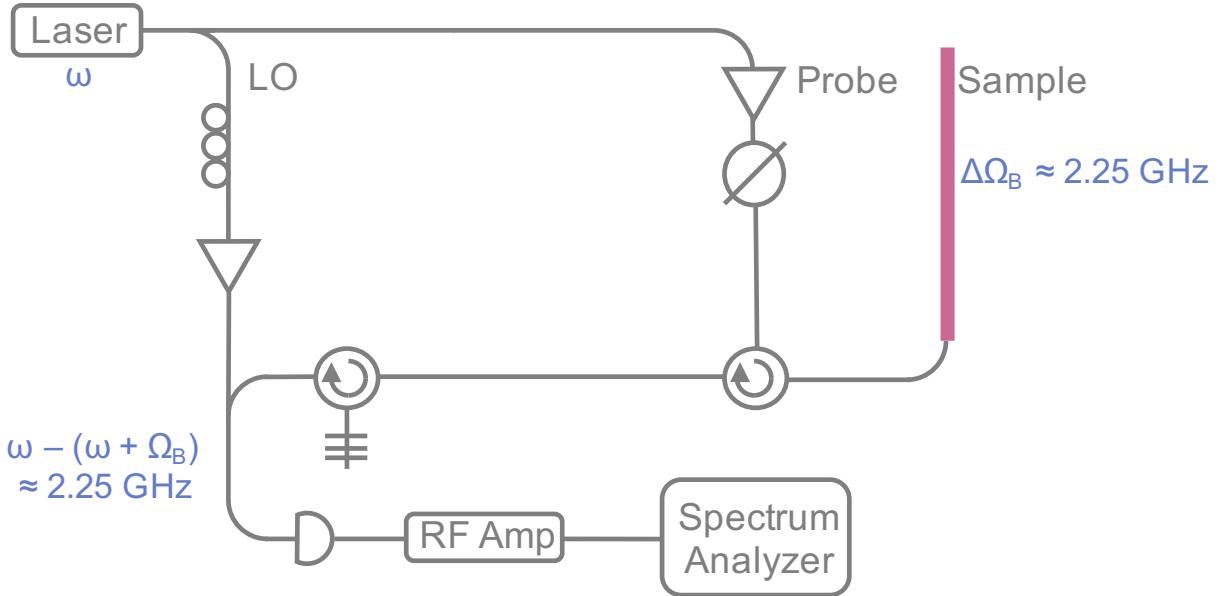


**Figure 2.2:** Schematic of liquid-core optical fiber (LCOF) design. A length of single mode fiber 28 (SMF-28) is arc-spliced to 5-10 cm of Ultra High Numerical Aperture 7 (UHNA7) fiber, with a post-arc process applied to taper the larger SMF-28 core down to the smaller UHNA7 core for better mode matching and coupling efficiency. The UHNA7 fiber is angle-cleaved and fusion-spliced to a flat-cleaved hollow-core fiber via a heated filament in a Vytran fusion splicer system. The angle cleave results in a splice that only partially fuses the two fibers, leaving a pathway for liquid to enter the hollow core fiber via capillary action once submerged. A mirrored splice configuration on the other end of the length of hollow-core fiber allows air to escape as the fiber fills, and a reverse taper again provides improved mode matching for the light to recouple into SMF-28.

### 2.3.1 CS<sub>2</sub>-Filled Liquid-Core Optical Fiber

To demonstrate optomechanical cooling of traveling wave phonons, we utilize 1 m of liquid-core optical fiber filled with carbon disulfide, first developed by Kieu et al. (2012).<sup>39</sup> This platform features large optomechanical coupling due to the high electrostrictive response of CS<sub>2</sub><sup>13</sup> and the small effective area defining acousto-optic mode overlap offered by the small 0.9 μm core radius of the capillary fiber. These characteristics of the LCOF system combine to produce an effective Brillouin gain coefficient  $G_B > 2 \text{ W}^{-1} \text{ m}^{-1}$ , enabling sufficient scattering within the relatively short length required to satisfy the fast-escape condition  $4v_g/L > \Gamma_0$  ( $4v_g/L \approx 0.82 \text{ GHz}$  and  $\Gamma_0 \approx 2\pi \times 97 \text{ MHz} \approx 0.61 \text{ GHz}$ ).<sup>23</sup> Additionally, this CS<sub>2</sub>-filled LCOF system provides excellent acoustic and optical guidance due to the large electromagnetic and acoustic impedance differential between the CS<sub>2</sub> core and surrounding silica.<sup>40</sup>

Figure 2.2 shows a schematic of the LCOF design. Coupling into and out of the LCOF is facilitated by a short length of tapered UHNA7 for better mode matching between the 4.8 μm core radius SMF-28 and the 0.84 μm core radius of the capillary fiber. An angled cleave of the UHNA7 fiber allows for a small wedge gap to remain after fusion splicing to the capillary fiber. This gap allows liquid CS<sub>2</sub> to enter and fill the hollow core of the fiber via capillary action. Appendix A.1 describes the fabrication and filling processes of the LCOF and details key insights which contributed to significant reductions in failure rate as well as time and material cost of sample preparation.



**Figure 2.3:** Schematic of experimental setup for Experiment A. In this experiment, a Continuous Wave (CW) pump laser emitting at  $1.55\text{ }\mu\text{m}$  is amplified, passed through a circulator, and injected into the  $\text{CS}_2$ -LCOF. Backscattered light is routed to a bandpass filter (BPF) for selection of Stokes or anti-Stokes frequencies and sent to a detector. A local oscillator (LO) is synthesized from the pump laser for heterodyne detection, whereby a polarization controller is used to align the polarization of the LO to that of the backscattered signal. The signal passes through a radio frequency amplifier before being sent to a radio-frequency spectrum analyzer (RFSA) for collection. Pump power is controlled by a Variable Optical Attenuator (VOA) placed just after the Erbium-Doped Fiber Amplifier (EDFA). Stokes and anti-Stokes spectra are collected sequentially for each pump power by adjusting the placement of the BPF.

### 2.3.2 Cooling Experiment A: Spontaneous Brillouin Cooling

We conducted two independent experiments to demonstrate and verify laser cooling of traveling wave phonons in our  $\text{CS}_2$ -LCOF platform. The first experiment (Experiment A) employs a pump laser to launch  $1.55\text{ }\mu\text{m}$  light into the LCOF. The backscattered Stokes and anti-Stokes components are filtered and collected by a detector. In this experiment, evidence of cooling is given by symmetry breaking between the amplitudes and widths of the Stokes and anti-Stokes spectra as pump power is varied. Backscattered pump light is shifted in frequency by a band of frequencies centered at the resonant Brillouin frequency of the LCOF ( $\Omega_{\text{B,LCOF}} \approx 2\pi \times 2.27\text{ GHz}$ ).

Figure 2.3 shows a schematic diagram of the experimental setup for Experiment A. A CW pump laser emitting at  $1.55\text{ }\mu\text{m}$  ( $\omega_{\text{P}}$ ) is amplified by an EDFA and its power is controlled by a VOA. This light is subsequently routed via a circulator to the  $\text{CS}_2$ -LCOF, where some of the light backscatters within the length of the LCOF. Backscattered light is frequency up-shifted for anti-Stokes ( $\omega_{\text{AS}} = \omega_{\text{P}} + \Omega_{\text{B}}$ ) and down-shifted for Stokes ( $\omega_{\text{S}} = \omega_{\text{P}} - \Omega_{\text{B}}$ ) by a band of frequencies centered around the Brillouin frequency

of the LCOF ( $\Omega_{B,LCOF} \approx 2.27$  GHz). Backscattered light is routed by circulators to a 5 GHz BPF to allow selection of either the Stokes or anti-Stokes light while also reducing unwanted noise. The filtered signal is then incident on a photodiode detector sensitive to GHz frequencies. A LO is synthesized from the pump laser, polarization controlled, amplified, and combined with the signal pre-detector for heterodyne detection. Output from the detector is amplified with an radio frequency (RF) amplifier and sent to an RFSA for data collection.

To conduct the experiment, pump power was varied from 10 mW to 290 mW in increments of 10 mW, as measured pre-injection into the LCOF, and respective Stokes and anti-Stokes spectra for each pump power were collected sequentially by tuning the placement of the BPF. For both the Stokes and anti-Stokes modes, five repeated measurements of each spectra and the background (pump off) were collected for each pump power, with each measurement comprising an average over 100 250 ms sweeps at a resolution bandwidth (RBW) of 1 MHz. Optical transmission through the entire LCOF sample used for the published data was measured to be 17%, giving  $100\sqrt{0.17} \approx 41\%$  single-pass transmission through the first splice assuming equal transmission through both. This provided a scaling factor for obtaining intrafiber powers from injected pump power ( $P_{intrafiber} = \sqrt{0.17}P_p$ ) which was used for data processing and analysis for the publication.

### 2.3.3 Cooling Experiment B: Pump-Probe Verification

A second experiment (B) was conducted to directly probe cooling via pump-probe spectroscopy, whereby a separate probe laser is held at constant power while a pump laser is varied to affect cooling in the LCOF. In Experiment B, backscattered light from the probe laser provides direct evidence of cooling through spectral changes in amplitude and linewidth as pump power is independently varied. To achieve independence of the pump and probe, probe light was controlled to be polarized orthogonal to the pump.

Figure 2.4 shows a schematic of the experimental design for Experiment B. Starting from a single CW laser operating at  $1.55\text{ }\mu\text{m}$ , we generate a pump  $\omega_p$ , probe  $\omega_{Pr}$ , and LO. A polarizing beam splitter (PBS) combines the pump and probe for co-injection into the LCOF. The pump light is amplified by an EDFA, and its polarization is adjusted so that it reflects at the PBS for launching into the LCOF. The probe light passes through a circulator and is polarization-adjusted to transmit through the PBS for injection into the LCOF. A 99-1 splitter directs 1% of the combined pump and probe light to a second PBS for monitoring of respective powers injected into the LCOF.

Backscattered light from both the pump and the probe is shifted in frequency by approximately the Brillouin frequency of the LCOF ( $\Omega_{B,LCOF} \approx 2.27$  GHz). These backscattered signals exhibit a spectral



**Figure 2.4:** Schematic of experimental setup for Experiment B. A pump, probe, and LO are generated from a single CW laser operating at  $1.55\text{ }\mu\text{m}$ . Pump light is amplified and directed through a VOA and polarization controller (PC), where its polarization is adjusted to reflect at the polarizing beam splitter (PBS). Probe light passes through a VOA for careful power control across measurement sets before being routed by a circulator to the PBS. Polarization of the probe light is adjusted for transmission through the PBS for co-injection with the pump into the LCOF. A 99-1 splitter directs 1% of the total pump and probe light to a second PBS for monitoring of respective powers injected into the LCOF. Backscattered Stokes and anti-Stokes components of both the pump and the probe retrace to the first PBS, where the respective polarization states of each ensure a re-separation of backscattered pump and probe light. The probe light is filtered by a tunable BPF and heterodyned with the LO prior to detection. Detector output is amplified by an RF amplifier and fed to an RFSA for data collection.

linewidth corresponding to the mechanical dissipation rate in the phonon mode (conversely, finite phonon lifetime). The polarization state of the backscattered pump and probe light is recovered after retracing their paths, permitting only probe light to transmit through the PBS for filtration and heterodyne detection. Output from the detector is again amplified by an RF amplifier and routed to an RFSA for data collection. With the probe power inside the LCOF held constant, changes in the amplitude and width of the probe spectra provide direct evidence of laser cooling from varying pump powers within the LCOF. The frequencies and wavevectors involved in both the pump and the probe scattering processes satisfy similar phase matching conditions given by Equation 2.2. Because this pump-probe technique provides a direct measure of laser cooling, only the anti-Stokes spectra need to be collected. In conducting the experiment, five repeated measurements of both the anti-Stokes spectra and the background (pump off) were collected for each pump power, with each measurement comprising an average over 100 250 ms sweeps at a RBW of 1 MHz.

Notably, this experiment relies on careful control of the polarization states of the pump and the probe. Upon backscattering within the LCOF, their respective polarization states are recovered after retracing their

paths to the PBS, which ensures only probe light transmits through the PBS for filtration and heterodyne detection. Output from the detector is amplified by an RF amplifier and routed to an RFSA for data collection. In this way, by holding the probe power constant inside the LCOF, changes in the amplitude and width of the backscattered probe spectra provide direct and independent evidence of laser cooling from varying pump powers within the LCOF.

## 2.4 Results

### 2.4.1 Cooling Experiment A Results

Figures 2.5 and 2.6 show the respective Stokes and anti-Stokes spectra gathered across a range of intra-fiber pump powers, from 4.12 mW to 119.57 mW. Both Stokes and anti-Stokes spectra sets are centered at approximately  $f_B = 2.27 \text{ GHz}$ , representing the respective up- and downshift from the pump frequency by the Brillouin frequency of the CS<sub>2</sub>-LCOF at 1.55 μm. For increasing pump power, we see an asymmetry in evolution of peak spectral densities and of the Stokes and anti-Stokes spectra, a key fingerprint of optomechanical cooling. Figure 2.7 plots the lorentzian-fitted Stokes and anti-Stokes peak amplitudes for each pump power. A dashed line showing a linear trend was obtained by applying a linear fit of all data points. Solid lines show the theoretical super- and sublinear scattered power for the Stokes and anti-Stokes processes, respectively, and were calculated using Equations A27 and A28 in Appendix A of Johnson et al. (2023)<sup>23</sup>. Figure 2.7 shows that peak amplitudes of both processes are in good alignment of the theoretical trend, with anti-Stokes peaks increasing sublinearly and Stokes peaks increasing superlinearly.

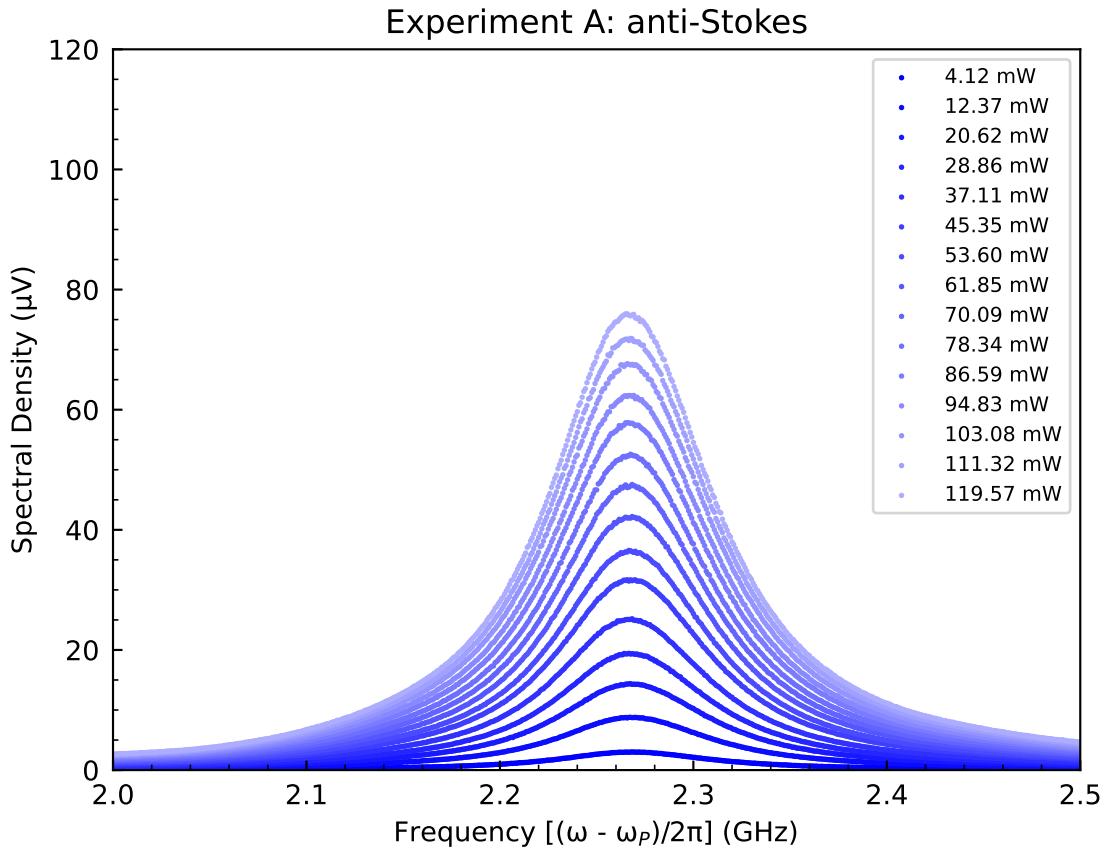
This is consistent with one of the key signatures of laser cooling: as higher pump powers further reduce the phonon occupation within the anti-Stokes mode (cooling), scattered power falls farther short of the otherwise linear relationship with pump power (Equation B.50). This sublinear trend in scattered power produces the same trend in backscattered light incident on the detector and reveals itself in the data as a sublinear trend in peak spectral density with increasing pump power. The opposite effect occurs in the Stokes process; as higher pump powers further increase the phonon occupation within the Stokes mode (heating), scattered power rises further above the otherwise linear relationship with pump power (again given by Equation B.50). This superlinear trend in scattered power produces the same trend in backscattered light incident on the detector and reveals itself in the data as a superlinear trend in peak spectral density with increasing pump power. These respective super- and sublinear growth trends in peak amplitudes of the Stokes and anti-Stokes spectra as pump power increases provide a spectral fingerprint of optomechanical cooling of the anti-Stokes phonon mode within the LCOF.

Linewidths of the spectra shown in Figures 2.5 and 2.6 also exhibit key spectral signatures of laser cooling



**Figure 2.5:** Stokes spectra for a range of pump powers obtained for Experiment A via spontaneous backwards Brillouin scattering. Intrafiber pump powers are displayed and were calculated from a measurement of 17% total through-fiber transmission and assuming equal losses through each of the two splices bookending the LCOF. To obtain each spectra, five repeated measurements of both the Stokes spectra and the background (pump off) were collected for each pump power, with each measurement comprising an average over 100 250 ms sweeps at a RBW of 1 MHz. Plotted are the resulting background-subtracted spectra, with uncertainties smaller than the data point markers.

(and heating for the Stokes mode). With increasing pump power, the linewidths of the anti-Stokes spectra broaden, indicating a higher dissipation rate of phonons into the “optical bath” (cooling). The inverse trend is seen for the Stokes mode: with increasing pump power, the linewidths of the Stokes spectra narrow, indicating a reduced phonon dissipation rate (heating). Figure 2.8 plots the Stokes and anti-Stokes linewidths for each pump power, obtained from a lorentzian fit. Solid lines provide a linear fit to the respective Stokes and anti-Stokes measured linewidths. The secondary y-axis provides a temperature scale in degrees K, with room temperature (taken to be 293 K) centered at the equilibrium linewidth of the LCOF. Section A.3.1 provides a tabulation of the extracted fit parameters and their associated uncertainties for each of the Stokes and anti-Stokes spectra for all pump powers.



**Figure 2.6:** Anti-Stokes spectra for a range of pump powers obtained for Experiment A via spontaneous backwards Brillouin scattering. Intrafiber pump powers are displayed and were calculated from a measurement of 17% total through-fiber transmission and assuming equal losses through each of the two splices bookending the LCOF. To obtain each spectra, five repeated measurements of both the anti-Stokes spectra and the background (pump off) were collected for each pump power, with each measurement comprising an average over 100 250 ms sweeps at a RBW of 1 MHz. Plotted are the resulting background-subtracted spectra, with uncertainties smaller than the data point markers.

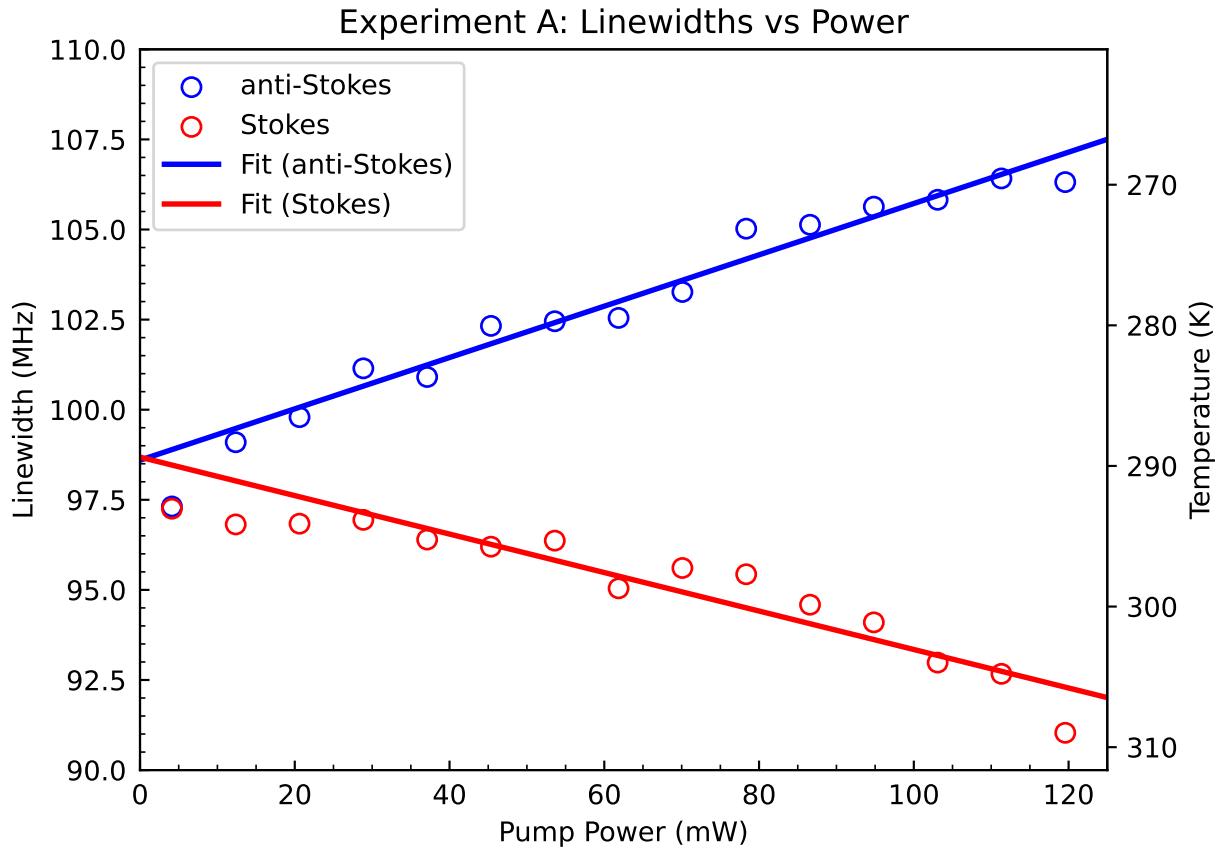
Using Equation 2.3 and the respective broadened and equilibrium anti-Stokes linewidths at the highest intrafiber pump power ( $\Gamma_{P_p} = 119.57 \text{ mW} = 106.32 \text{ MHz}$ ) as compared to the lowest ( $\Gamma_{P_p} = 4.12 \text{ mW} = 97.3 \text{ MHz}$ ), we find an effective Brillouin gain  $G_B \approx (3.1 \pm 0.5) \text{ W}^{-1} \text{ m}^{-1}$ . Using this gain factor, we find (right side of Equation 2.10) the temperature of the band of traveling wave phonons in the LCOF to be reduced by  $\Delta T \approx (25 \pm 2) \text{ K}$  from room temperature. Using instead the phonon occupancy method (left side of Equation 2.10) with the same respective linewidths, we find  $\Delta T \approx (27 \pm 2) \text{ K}$ , which agrees with the prior calculation and is independent of estimated Brillouin gain and intrafiber pump powers. All measured values and their associated uncertainties are tabulated for reproducibility in Tables A.1 and A.2 in Appendix A, Section A.3. The published article associated with this work reports a conservatively calculated Brillouin



**Figure 2.7:** Lorentzian-fitted peak amplitudes of the observed Stokes and anti-Stokes spectra. The dashed line showing a linear trend was obtained by applying a linear fit of all data points. The solid lines show the theoretical super- and sublinear scattered power for the Stokes and anti-Stokes processes, respectively, and were calculated using Equations A27 and A28 in Appendix A of Johnson et al. (2023)<sup>23</sup>. Section A.3.1 provides a tabulation of the extracted fit parameters and their associated uncertainties for each of the Stokes and anti-Stokes spectra for all pump powers.

gain of  $2.3 \text{ W}^{-1} \text{ m}^{-1}$ , also obtained from measured linewidths and Equation 2.3. This discrepancy arises from slight differences in measured linewidths of the anti-Stokes spectra, which we attribute to variations among weighted fitting algorithms and resulting uncertainties. Using this reported value of  $G_B \sim 2.3 \text{ W}^{-1} \text{ m}^{-1}$  for the Brillouin gain along with the right side of Equation 2.10 recovers the conservatively reported  $\sim 21 \text{ K}$  of cooling.

Figure 2.9 plots a simulated Brillouin gain spectrum  $G_B (\text{W}^{-1} \text{ m}^{-1})$ , obtained from finite element simulations<sup>23</sup> of the optical and acoustic modes of the LCOF, overtop the 4.12 mW pump power (nearest to natural equilibrium) anti-Stokes spectrum. While the y-scaling between the two spectra is arbitrary, the lorentzian profile of the gain spectrum, and specifically the center frequency and natural-equilibrium linewidth, is pre-



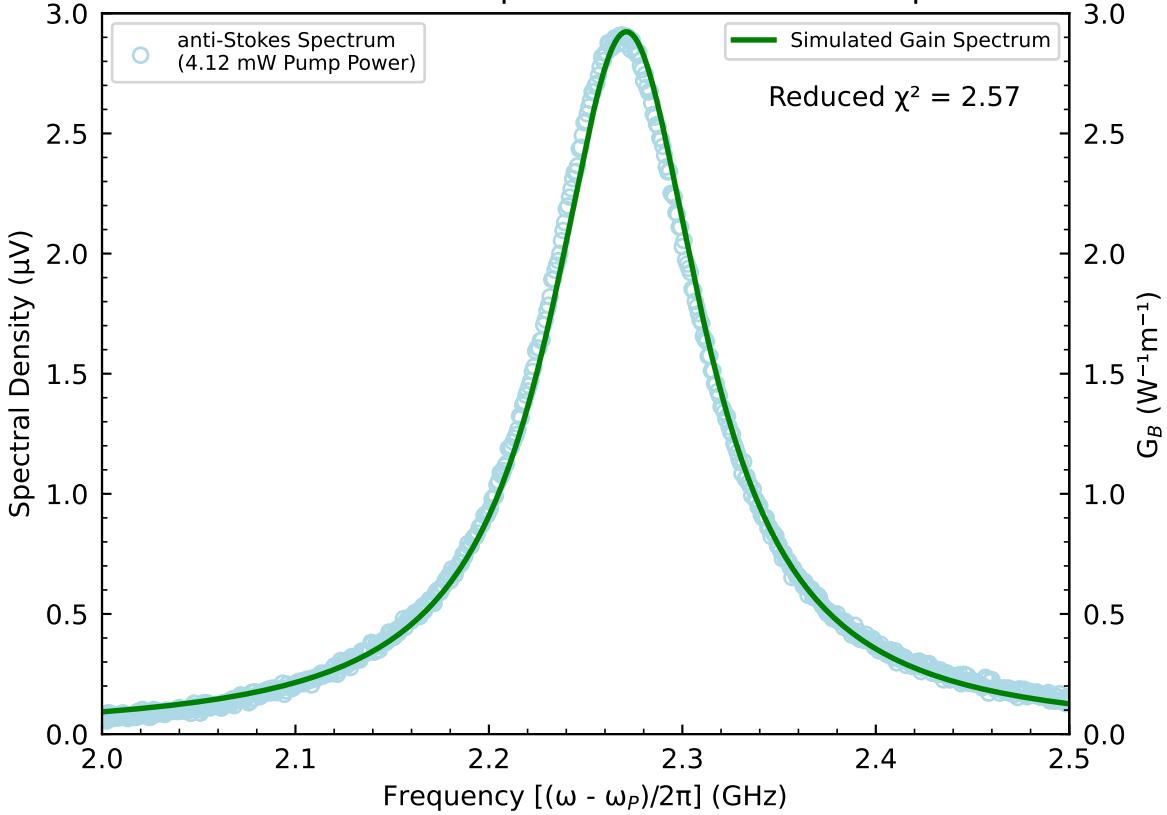
**Figure 2.8:** Stokes and anti-Stokes linewidths for each pump power, obtained from a lorentzian fit. Solid lines provide a linear fit to the respective Stokes and anti-Stokes measured linewidths. The secondary y-axis provides a temperature scale in degrees K, with room temperature (taken to be 293 K) centered at the equilibrium linewidth of the LCOF. Section A.3.1 provides a tabulation of the extracted fit parameters and their associated uncertainties for each of the Stokes and anti-Stokes spectra for all pump powers.

dictive of scattered power expected from the LCOF across the frequency range. An uncertainty-weighted reduced  $\chi^2$  test for goodness of fit of the simulated gain spectrum to the observed anti-Stokes spectrum yields a value of 2.57. Reduced  $\chi^2$  values close to unity indicate a good fit.

#### 2.4.2 Cooling Experiment B Results

Anti-Stokes scattered probe power was collected for four pump powers launched into the LCOF, providing intrafiber pump powers of 0.0 mW, 22.7 mW, 45.4 mW, and 68.0 mW. Figures 2.10, 2.11, 2.12, and 2.13 show these resulting spectra, respectively, along with associated weighted Lorentzian fit for each. The plotted data has been binned within 10 MHz bins for clarity, with the Lorentzian fits and associated parameters having been derived from the unbinned data. Visual inspection confirms an excellent fit for each of the Lorentzian

### Experiment A: Simulated Gain Spectrum Predicts Lineshape

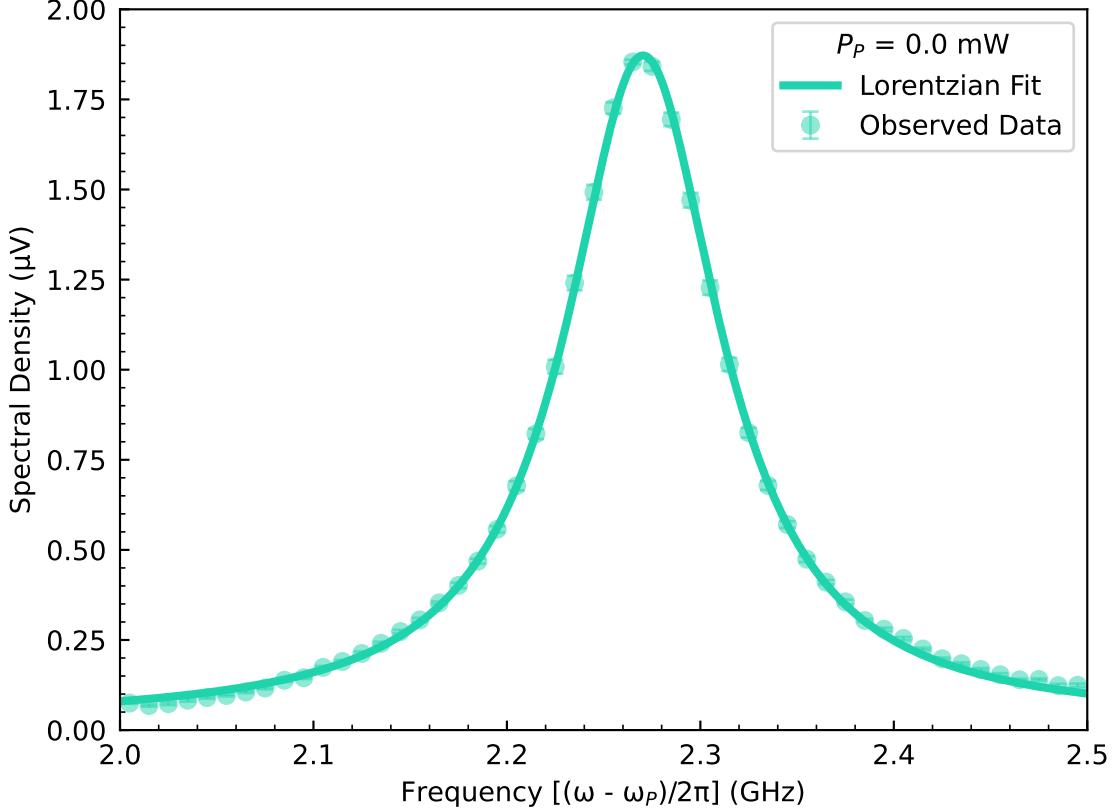


**Figure 2.9:** A simulated Brillouin gain spectrum  $G_B$  ( $\text{W}^{-1} \text{m}^{-1}$ ), obtained from finite element simulations<sup>23</sup> of the optical and acoustic modes of the LCOF, overtop the lowest power (nearest to natural equilibrium) anti-Stokes spectrum (the 4.12 mW pump power spectrum). While the y-scaling between the two spectra is arbitrary, the lorentzian profile of the gain spectrum, and specifically the center frequency and natural-equilibrium linewidth, is predictive of scattered power expected from the LCOF across the frequency range. An uncertainty-weighted reduced  $\chi^2$  test for goodness of fit of the simulated gain spectrum to the observed anti-Stokes spectrum yields a value of 2.57. Reduced  $\chi^2$  values close to unity indicate a good fit.

fits to their respective data, which have been plotted with error bars representing  $1\sigma$  of the spectral density for each data point. All measured and calculated values are tabulated in Table A.3 in Appendix A, Section A.3.2, including solved fit parameters and their associated uncertainties.

Figure 2.14 plots just the resulting fits for each spectra, showing a clear decrease in peak spectral amplitude with increasing pump power, consistent with the theory (see Equation B.50 in Appendix B.2, with constant power yet decreasing modal temperature). Figure 2.15 plots these peak spectral heights, derived from the fits, with their associated  $1\sigma$  uncertainties. A linear fit has been applied, showing good correlation with the decreasing trend in peak spectral densities with increased pump power (cooling) corresponding to a lowering of the temperature of the anti-Stokes phonon mode. Figure 2.16 plots the spectral linewidths derived

### Experiment B: anti-Stokes Power Spectrum $P_P = 0.0 \text{ mW}$

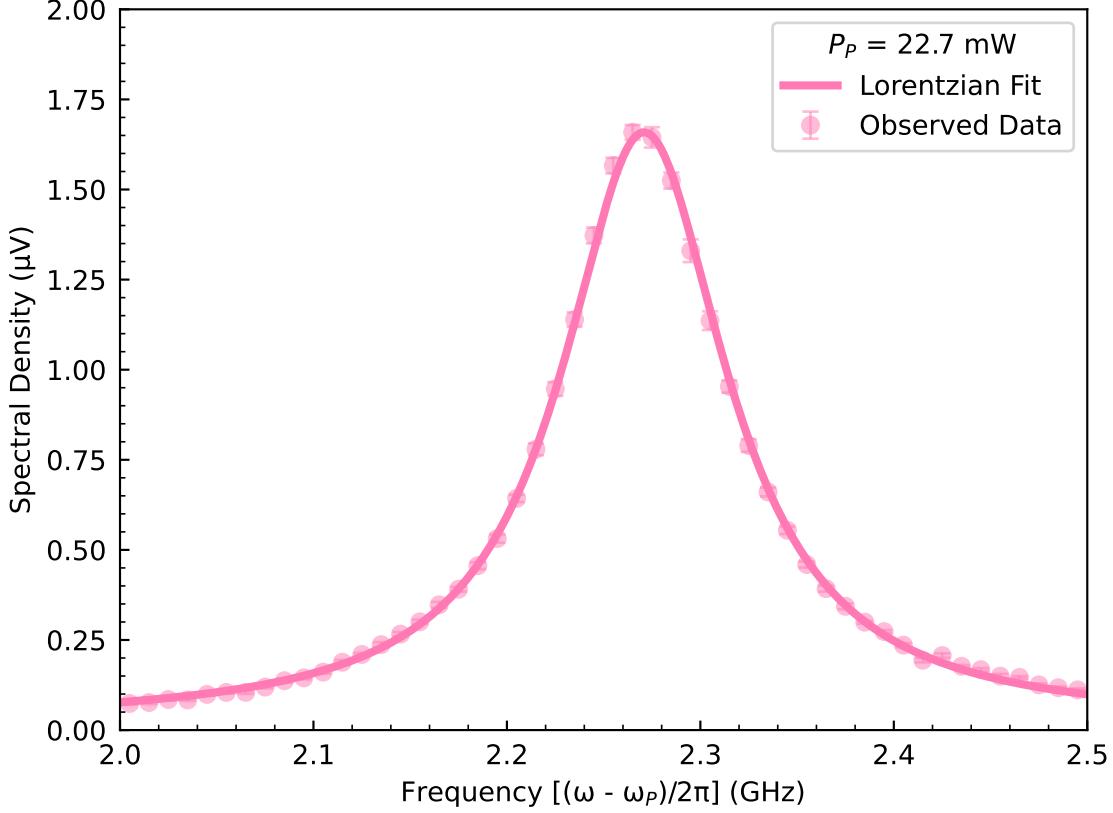


**Figure 2.10:** Anti-Stokes probe spectra collected for 0.0 mW intrafiber pump power in Experiment B. Data is binned within 10 MHz bins and displayed with error bars representing  $1\sigma$  of spectral density. A Lorentzian fit has been computed based on the unbinned data and visual inspection confirms an excellent fit to the observed spectra. In conducting the experiment, five repeated measurements of both the anti-Stokes spectra and the background (pump off) were collected for each pump power, with each measurement comprising an average over 100 250 ms sweeps at a RBW of 1 MHz. The spectral data shown here are the resulting background-subtracted spectra, with uncertainties propagated accordingly.

from the fits presented in Figure 2.14, with their associated  $1\sigma$  uncertainties. A linear fit has been applied, showing fair correlation given the  $1\sigma$  uncertainties displayed. The linear increase in spectral linewidths with increasing pump power (cooling) indicates, as with Experiment A, an increased dissipation rate due to laser cooling of the anti-Stokes phonon mode.

We can apply the same quantitative analysis for the data gathered in Experiment B as was performed for Experiment A. Using the 0.0 mW and 68.0 mW pump power spectral linewidths for the equilibrium and maximally cooled dissipation rate, respectively, in Equation 2.3, we find a Brillouin gain of  $G_B \approx (8 \pm 2) \text{ W}^{-1} \text{ m}^{-1}$ , taking the  $2\sigma$  confidence interval in linewidths and allowing a 10% uncertainty in pump power within the LCOF. Applying this gain factor to the right side of Equation 2.10, we find the traveling-

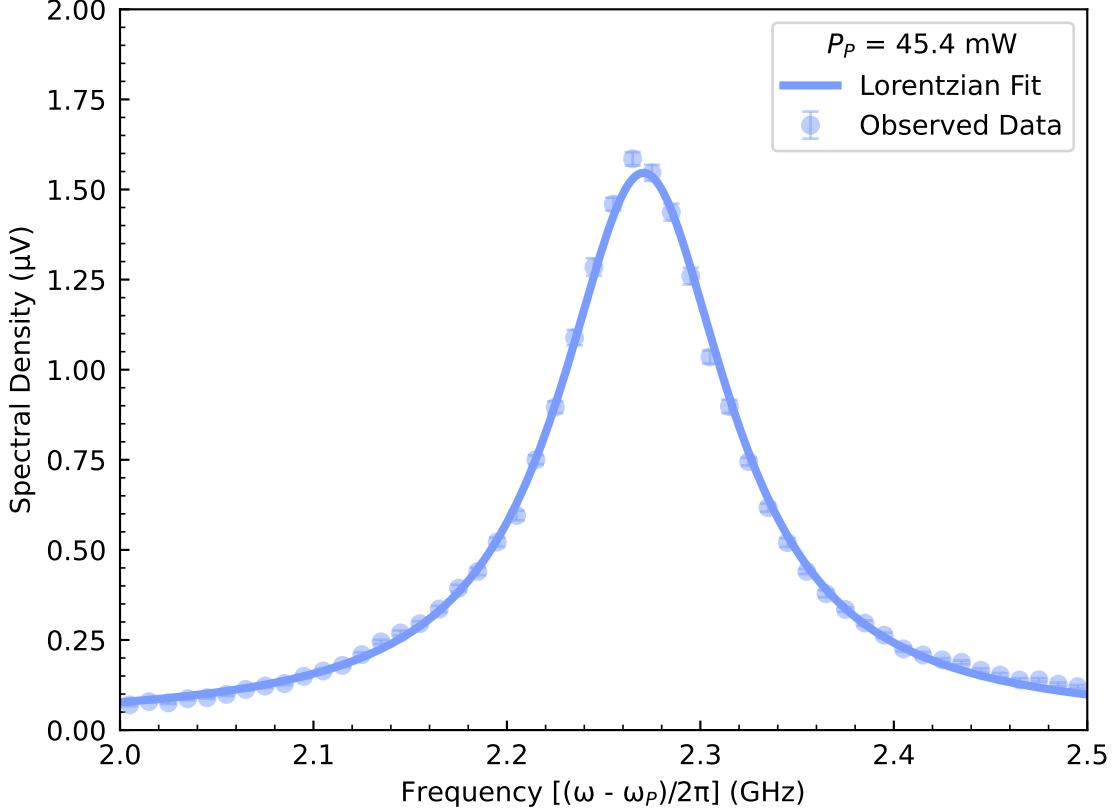
**Experiment B: anti-Stokes Power Spectrum**  
 $P_P = 22.7 \text{ mW}$



**Figure 2.11:** Anti-Stokes probe spectra collected for 22.7 mW intrafiber pump power in Experiment B. Data is binned within 10 MHz bins and displayed with error bars representing  $1\sigma$  of spectral density. A Lorentzian fit has been computed based on the unbinned data and visual inspection confirms an excellent fit to the observed spectra. In conducting the experiment, five repeated measurements of both the anti-Stokes spectra and the background (pump off) were collected for each pump power, with each measurement comprising an average over 100 250 ms sweeps at a RBW of 1 MHz. The spectral data shown here are the resulting background-subtracted spectra, with uncertainties propagated accordingly.

wave phonons in the anti-Stokes mode of the LCOF to have been cooled by  $\Delta T \approx (35 \pm 2)$  K. Using the quantized thermal dissipation rate method to calculate degrees cooled (left side of Equation 2.10), we find  $\Delta T \approx (36 \pm 2)$  K of cooling, which is in agreement with the prior calculated value using the Brillouin gain approach. The discrepancy among Brillouin gain factors for the LCOF calculated from Experiment A and Experiment B is not expected and is explored in the next section. This Brillouin gain factor is a consequence of both the linewidths and the corresponding pump powers inside the LCOF, seen visually as the slopes of Figure 2.8 for Experiment A and Figure 2.16 for Experiment B.

### Experiment B: anti-Stokes Power Spectrum $P_P = 45.4$ mW

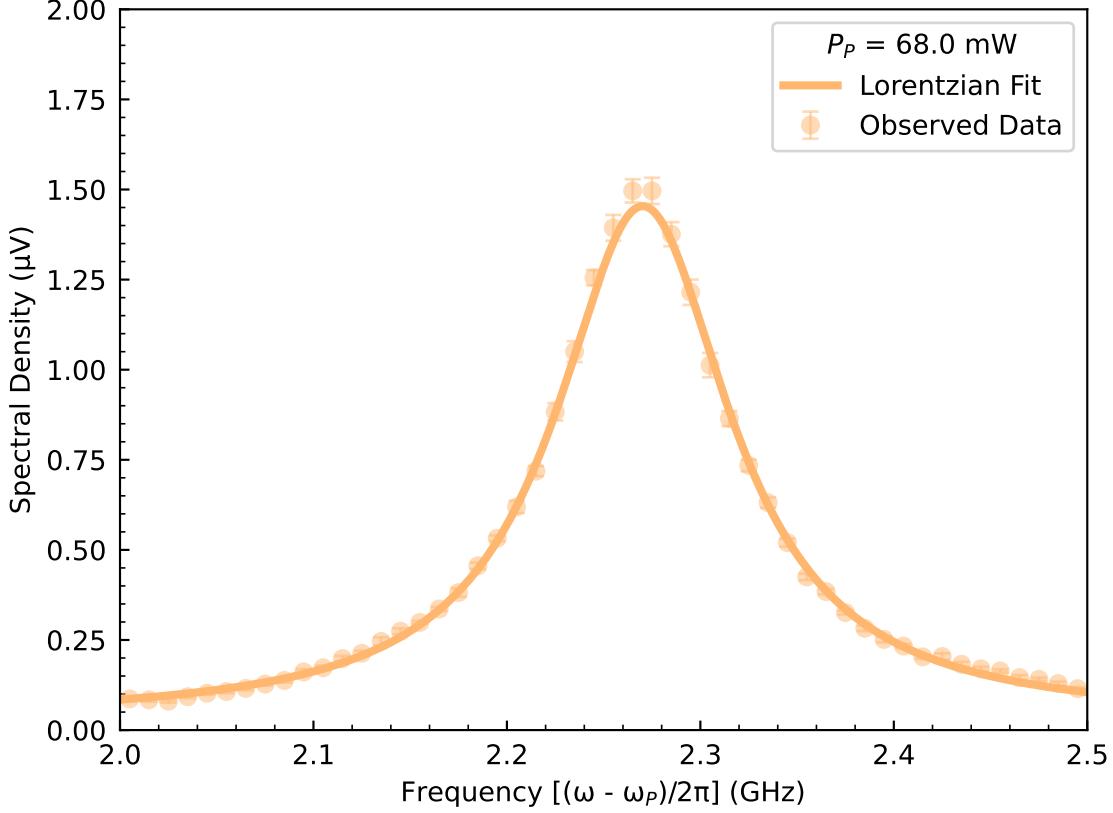


**Figure 2.12:** Anti-Stokes probe spectra collected for 45.4 mW intrafiber pump power in Experiment B. Data is binned within 10 MHz bins and displayed with error bars representing  $1\sigma$  of spectral density. A Lorentzian fit has been computed based on the unbinned data and visual inspection confirms an excellent fit to the observed spectra. In conducting the experiment, five repeated measurements of both the anti-Stokes spectra and the background (pump off) were collected for each pump power, with each measurement comprising an average over 100 250 ms sweeps at a RBW of 1 MHz. The spectral data shown here are the resulting background-subtracted spectra, with uncertainties propagated accordingly.

#### 2.4.3 Evidence of Transmission Degradation

After comparing the anti-Stokes linewidths measured in Experiments A and B, we conclude that significant splice transmission degradation occurred in the interim. Experiment B was performed first (with a through-fiber transmission of 17%), followed by Experiment A the next day. If the experimental conditions were unchanged between the two experiments, the slopes of the anti-Stokes linewidth versus pump power would be identical (Figures 2.8 and 2.16). Instead, Experiment B exhibits a noticeably steeper slope than Experiment A. A reanalysis shows that reducing the assumed transmission for Experiment A so the two slopes match would require the total fiber transmission to have dropped from 17% down to approximately

**Experiment B: anti-Stokes Power Spectrum**  
 $P_P = 68.0 \text{ mW}$



**Figure 2.13:** Anti-Stokes probe spectra collected for 68.0 mW intrafiber pump power in Experiment B. Data is binned within 10 MHz bins and displayed with error bars representing  $1\sigma$  of spectral density. A Lorentzian fit has been computed based on the unbinned data and visual inspection confirms an excellent fit to the observed spectra. In conducting the experiment, five repeated measurements of both the anti-Stokes spectra and the background (pump off) were collected for each pump power, with each measurement comprising an average over 100 250 ms sweeps at a RBW of 1 MHz. The spectral data shown here are the resulting background-subtracted spectra, with uncertainties propagated accordingly.

3%. While large, degradation of this level is plausible and is supported by several further points of analysis.

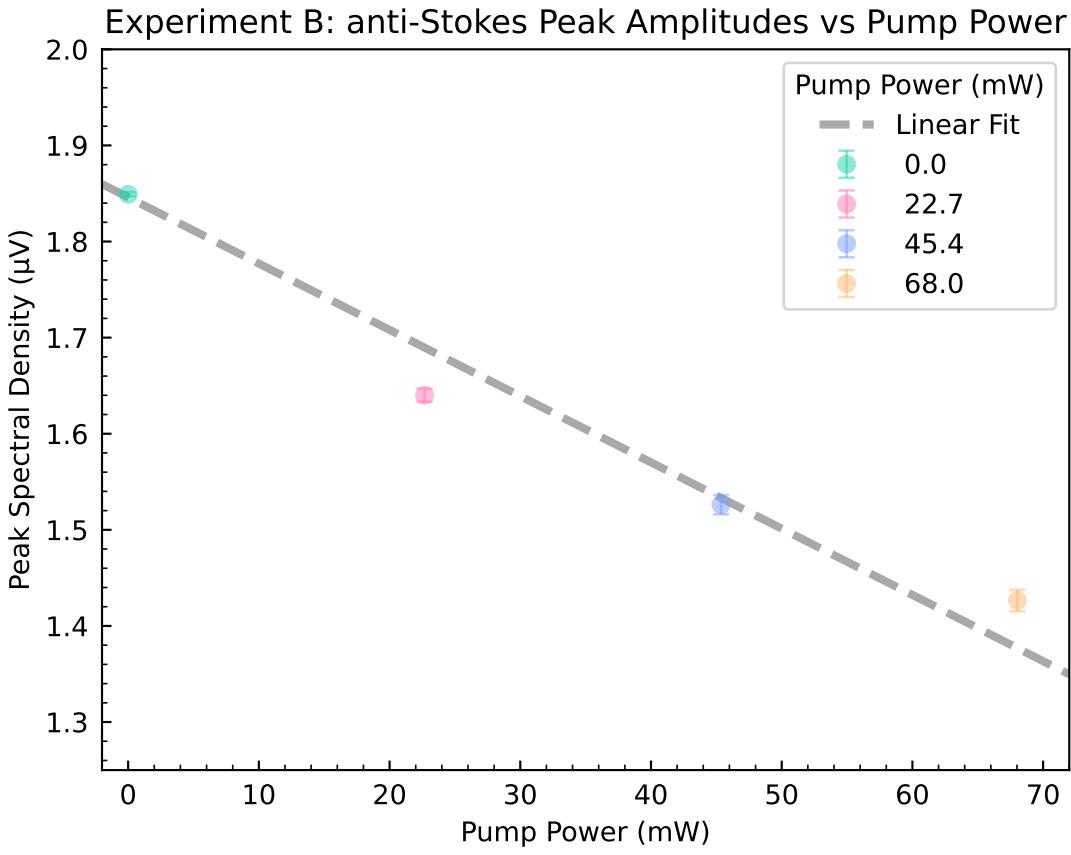
First, the optical performance of the splices bookending the LCOF, already very delicate and susceptible to damage from normal handling, are vulnerable to any particulates becoming lodged between the cores of the UHNA7 and LCOF. While epoxy is among the more chemically resistant adhesives, any particulate contamination adhered to the surface of the epoxy or reservoir walls introduced from the environment during fabrication could easily become dislodged and settle into the splice opening. Some chemical dissolution of the epoxy within the reservoir by the CS<sub>2</sub> after the prolonged soaking period (24 h) is also possible. Additionally, any unfortunate mechanical stress incurred during handling of the LCOF is liable to cause damage and incur significant optical loss through one of the splices. Optical losses of this rate and magnitude, while not



**Figure 2.14:** Computed Lorentzian fits from the observed anti-Stokes probe spectra gathered for Experiment B, shown in Figures 2.10, 2.11, 2.12, and 2.13, with consistent coloring across plots corresponding to intrafiber pump power. Fits are identical to those displayed with their respective observed spectra, with the fit parameters computed from the unbinned spectra for each intrafiber pump power. The decreasing trend in peak spectral density (proportional to phonon population) with increasing pump power (cooling) provides direct evidence of laser cooling of a band of traveling wave phonons within the LCOF.

common, have been observed in several prior LCOF samples.

Standard experimental procedure would catch this degradation in performance, however for this particular LCOF sample, the exit fiber immediately following the end splice broke off irreparably after testing transmission, making it no longer possible to remeasure the full through-fiber transmission during Experiment A. In other words, if transmission had declined between experiments, we had no direct means of detecting it. Additionally, as seen in Figures 2.3 and 2.4, the pump power was recorded prior to launch into the LCOF for both experiments, providing no indication of change in performance of the splice between experiments. Hence, applying the same optical loss estimate to a splice that had experienced transmission degradation in the interim would result in an overestimation of the actual power inside the LCOF during

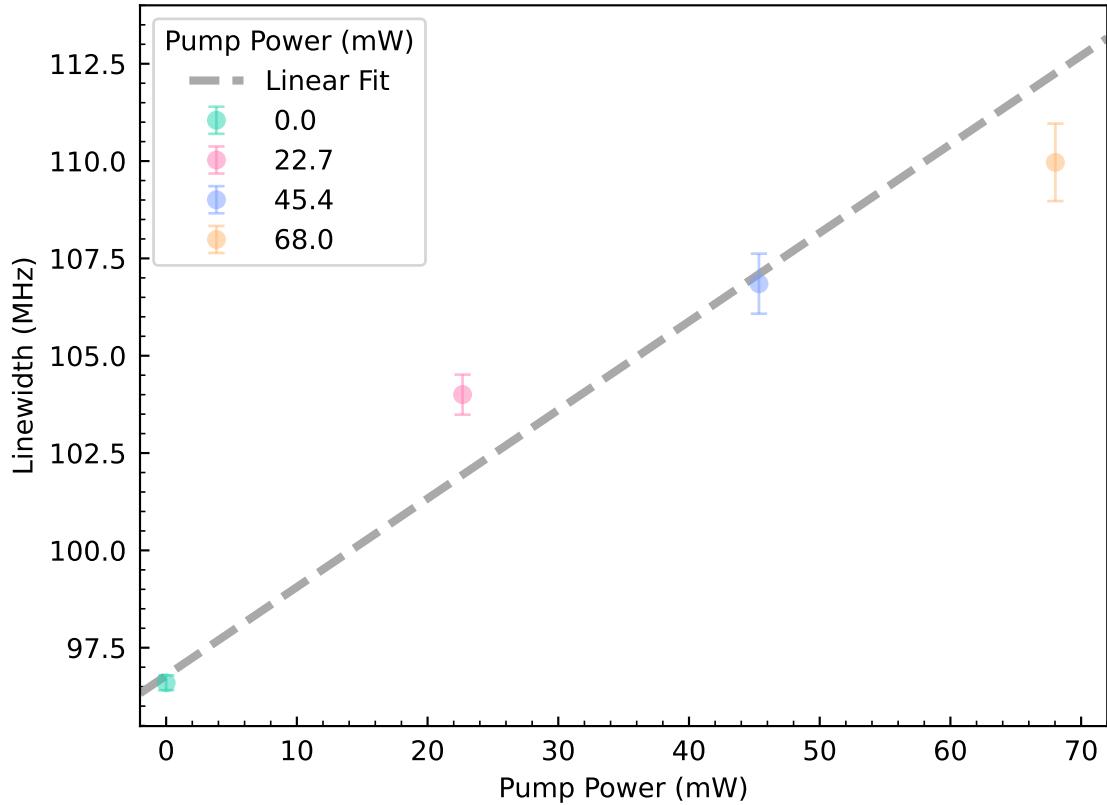


**Figure 2.15:** Plot of peak spectral density amplitudes for the anti-Stokes spectra gathered for Experiment B, corresponding to the four intrafiber pump powers used for the experiment. Values have been extracted from solved fit parameters for the Lorentzian fits displayed in Figure 2.14, as well as individually with their respective observed spectra (Figures 2.10, 2.11, 2.12, and 2.13). A linear fit has been applied to the data, showing a fair correlation to the data given the  $1\sigma$  uncertainty representation for peak spectral density. The peak spectral amplitude is expected to decrease linearly with decreasing temperature of the mechanical mode due to anti-Stokes Brillouin cooling, consistent with Equation B.50 for scattered power of the spontaneous Brillouin scattering process.

Experiment A (conducted after Experiment B).

Further quantitative analysis supports this hypothesis as well. As we reported in Johnson et al. (2023),<sup>23</sup> Experiment A yields a Brillouin gain of approximately  $2.3 \text{ W}^{-1} \text{ m}^{-1}$ , calculated using Equation 2.3. This value is smaller than the  $(6 \pm 1) \text{ W}^{-1} \text{ m}^{-1}$  reported by Behunin et al. in 2019<sup>40</sup> for a near-identical LCOF geometry and fabrication method. As stated previously, if instead we calculate the Brillouin gain based on the linewidths and corresponding intrafiber pump powers seen in the earlier Experiment B, we find the Brillouin gain to be approximately  $(8 \pm 2) \text{ W}^{-1} \text{ m}^{-1}$ , which is in good agreement with the prior published value from the literature. Again, in estimating the uncertainty here, we have taken the  $2\sigma$  confidence level

### Experiment B: anti-Stokes Linewidths vs Pump Power



**Figure 2.16:** Plot of the full-width half-max (FWHM) linewidths of the anti-Stokes spectra gathered for Experiment B, corresponding to the four intrafiber pump powers used for the experiment. Values have been extracted from solved fit parameters for the Lorentzian fits displayed in Figure 2.14, as well as individually with their respective observed spectra (Figures 2.10, 2.11, 2.12, and 2.13). A linear fit has been applied to the data, showing a fair correlation to the data, given the  $1\sigma$  uncertainty representation for peak spectral density. Effective anti-Stokes linewidths are expected to increase linearly with increased pump power (cooling) according to Equation 2.3, absent pump depletion or any saturation effects. Discrepancies with a linear trend among the data presented with their associated vertical uncertainties are likely due to an underestimation of uncertainty in intrafiber pump power, which would be represented by some horizontal uncertainty associated with each data point. In the possible event of thermal saturation effects in the LCOF, or unlikely pump depletion, we would expect to see linewidths exhibit a sublinear trend above a critical threshold of intrafiber pump power. While it is possible some minor saturation occurred at these powers, a more conservative accounting of uncertainties in both linewidths and intrafiber pump powers is sufficient to explain a linear trend.

in linewidths and allowed a 10% uncertainty in intracore pump powers. The natural conclusion is that for Experiment A we overestimated the intrafiber pump power due to the undetected splice transmission degradation between experiments. This produced an apparent smaller dependence of linewidth on pump power in Experiment A and a consequent underestimation of the true gain of the LCOF.

Finally, the linewidths of Experiment B appear to exhibit a slight sublinear trend with power (Figure 2.16), indicating the possibility of a saturation effect occurring at higher pump power. However any saturation effect or similar which could cause this trend should then also be activated at the higher pump powers used in Experiment A, if the optical transmission were the same across experiments. If instead the transmission of the LCOF entrance splice had been significantly greater for the earlier-run Experiment B than for Experiment A, the powers inside the LCOF in Experiment B could have been similar to or even greater than those in Experiment A. This might explain why Experiment B linewidths appear to exhibit slightly sublinear growth with pump power despite the lower pump powers launched into the LCOF while Experiment A does not seem to exhibit this same trend, despite the use of higher nominal pump powers. This is consistent with an estimated factor of 5 drop in transmission through the LCOF splice between experiments, as indicated by the discrepancy in linewidth vs power slopes of each experiment.

In summary, all these lines of evidence strongly indicate that the LCOF splice transmission fell substantially in the time between Experiments B and A, by an approximate factor of five. This fully reconciles the differing slopes in the anti-Stokes linewidth data and explains the otherwise surprising discrepancy in inferred Brillouin gain compared to values published in the literature for identical  $\text{CS}_2$ -filled LCOFs. This conclusion implies even higher performance of our LCOF example than was previously understood and reported in Johnson et al. (2023):<sup>23</sup> the amount of cooling achieved in Experiment A (unaffected by this reanalysis) was accomplished with far less pump power.

## 2.5 Discussion

### 2.5.1 The C-Value: A Standardized Cooling Metric

Cooling results are often reported in degrees kelvin cooled, sometimes only implicitly stated from room temperature. While this is understood by researchers closely following laser cooling, it can be misleading to those who are looking to apply these published techniques to systems not starting at room temperature. A lack of clarity of cooling performance as being a function of starting temperature may cause buyer's remorse among researchers who expend resources employing the technique only to realize less than advertised absolute degrees cooling of a material below room temperature. Even for the more careful researcher this can introduce frustration in the literature review process. To address this, we propose a normalized cooling metric be adopted to report the fractional degrees cooled relative to initial temperature. We introduce the *Cooling metric ( $C$ )*, or *C-value*, expressed as a percentage of degrees cooled from initial temperature,

$$C = \frac{\Delta T}{T_0}. \quad (2.11)$$

Reporting the  $C$ -value of a cooling technique offers greater transparency, clarity, and universal applicability of performance across cooling techniques and application fields. Recent discussions in the optical refrigeration community have similarly called for standardized cooling metrics to ensure fair, reproducible comparisons of cooling results.<sup>41</sup> Indeed, reporting only absolute degrees cooled may significantly *underrepresent* the performance of a given technique if applied to a material above room temperature (as is often the case for data center and information processing applications). Adopting the  $C$ -value for cooling performance will expand the perceived viability and utility of laser cooling techniques to researchers outside of optomechanical physics and related fields, potentially spurring greater investment interest in the advancement of laser cooling techniques. As such, we report a  $C$ -value of 12%, achieved with 68.0 mW of intrafiber pump power (Experiment B).

By contrast, the recent result by Martinez et al. (Max Planck Institute, 2024)<sup>42</sup> demonstrated a much larger  $\Delta T \approx 219$  K cooling from room temperature, an unprecedented  $C$ -value of 73% cooling of the phonon mode's temperature. This emphasizes the Max Planck group's achievement of removing nearly three-quarters of the thermal energy from that acoustic mode, a level on par with the best solid-state optical cryocoolers, which have reached  $\sim 91$  K from ambient ( $\sim 70\%$  cooling) in Yb-doped crystals.<sup>41</sup> The fractional metric thus allows us to compare techniques across disparate systems such as optical fibers and bulk crystals on equal footing for their cooling effectiveness. As approaches push toward lower starting temperatures (e.g. pre-cooled or cryogenic environments), absolute  $\Delta T$  naturally shrinks, but a high  $C$ -value remains notable. Adopting the  $C$ -value, in combination with a commitment to publishing sufficiently rigorous experimental context,<sup>41</sup> yields a transparent gauge of performance across diverse temperature regimes and material platforms.

### 2.5.2 Parallel Achievement: Max Planck Institute

The principles demonstrated in this chapter can be directly compared to parallel work by other groups using different fiber platforms. In particular, Martinez et al. (2024)<sup>42</sup> reported optoacoustic cooling of traveling phonons in a tapered chalcogenide photonic crystal fiber (PCF). In their experiment, a 50 cm long PCF with a microstructured, tapered core was used to enhance optomechanical coupling. Like our system, the Max Planck group employed Brillouin backscattering in a continuous waveguide to achieve phonon cooling of traveling-wave phonons. The underlying mechanism is the same: anti-Stokes scattering removes

thermal phonons by converting them into higher-energy photons that promptly leave the system. Both platforms therefore rely on traveling-wave Brillouin interactions and leverage the fast escape of anti-Stokes light compared to phonon thermalization times to cool a band of acoustic modes.

Despite these similarities in principle, there are important differences in the experimental implementation. The PCF platform uses a solid chalcogenide fiber with a patterned microstructure, which was tapered to a small core diameter. The combined material and geometric parameters yield an acoustic mode at 7.38 GHz (higher than the 2.27 GHz acoustic band in our liquid-core fiber) and a very large effective Brillouin gain coefficient  $G_B \approx 180 \text{ W}^{-1} \text{ m}^{-1}$ . The higher acoustic frequency in the PCF means each phonon carries more energy ( $\hbar\Omega_B/k_B \approx 0.35 \text{ K}$  vs 0.1 K for our system), but also that the mean thermal occupation at room temperature is lower ( $k_B T/\hbar\Omega_B \approx 10^3$  vs.  $10^4$  in our lower frequency mode). The two experiments also operated at different pump power: our work used a modest  $(68 \pm 7) \text{ mW}$  of laser power to achieve  $(36 \pm 2) \text{ K}$  of cooling (Experiment B), whereas the PCF results employed a maximum pump power of nearly 250 mW to achieve the reported 219 K cooling from room temperature, or a  $C$ -value of 74.7%.<sup>42</sup>

### 2.5.3 Pathways to Net Cooling

While the experiments to date (including this work and Martinez et al.) achieve cooling of a phonon mode relative to the ambient lattice temperature, the phonon population of the overall system remains uncooled due to the inverse process (Stokes) simultaneously depositing energy into the Stokes phonon mode. Net cooling would imply extracting heat faster than it is replenished by both thermalization (the natural dissipation rate of the system) *and* Stokes scattering of the Stokes phonon mode. Engineering a system to reach this regime is a challenge, but we envision several pathways that could tip the balance toward net cooling.

One strategy is to bias the gain in favor of anti-Stokes scattering within a single transit of the medium, by suppressing or preventing Stokes processes through waveguide design. In a standard fiber, any pump photon has some probability to scatter into a Stokes photon, creating a phonon, which counteracts cooling. If we can reduce the likelihood of Stokes scattering while allowing anti-Stokes scattering to proceed, the net effect in one pass will be phonon removal without equivalent phonon creation. For example, one could design a fiber or waveguide that does not efficiently guide the Stokes-shifted wavelength, similar to a photonic bandgap fiber that places the Stokes frequency in a bandgap. In such a fiber, a pump photon could still undergo anti-Stokes scattering, but a would-be Stokes photon falls outside the guided modes and is effectively not generated.

Another approach is to incorporate an out-of-plane coupling mechanism to an absorbing medium or a frequency cutoff for only the Stokes. By asymmetrically enhancing loss for Stokes, one can bias the single-

pass Brillouin interaction toward net phonon annihilation. By diverting the Stokes field away from the guided mode through an engineered out-of-plane coupling, we can effectively “dump” the Stokes into a loss channel while preserving the anti-Stokes within the waveguide core, thus favoring net phonon removal. For instance, one might introduce a side-etched structure or grating that phase-matches only to the Stokes frequency, routing Stokes photons into an absorbing or radiative mode. Meanwhile, the anti-Stokes remains confined to the original waveguide, continuing to interact with the target phonon mode. Such a design ensures that Stokes photons, once generated, are immediately evacuated before they can drive the reverse (phonon creation) process. When combined with sufficiently strong anti-Stokes gain, this asymmetric loss channel could break the symmetry of phonon creation and annihilation within a single transit, thereby accelerating phonon cooling. Although this approach demands precise control over waveguide geometry and materials to selectively scatter and absorb Stokes without degrading anti-Stokes propagation, it holds promise for realizing net cooling in practical fiber architectures.

Another promising avenue for enhancing net phonon cooling in continuous Brillouin systems is the deliberate engineering of a group-velocity bias between the Stokes and anti-Stokes waves. By differentiating their propagation speeds, one introduces a temporal asymmetry in interaction strength that preferentially enables phonon annihilation over phonon creation. Some works explore this idea via enhancement of the optical density-of-states at the anti-Stokes frequency via slow-light or resonance effects while reducing it at the Stokes frequency.<sup>31, 43</sup> Crucially, however, any group velocity bias strategy must be designed with the phonon lifetime and effective system length in mind, as effective cooling demands that anti-Stokes photons carry away energy faster than the phonon mode thermally repopulates.

#### 2.5.4 Applications to Ground State Cooling

Reaching lower phonon occupancies and ultimately approaching the quantum ground state of a traveling acoustic wave mode is not only a scientific milestone but also an enabling step for quantum applications. Cooling traveling-wave phonons toward the ground state means drastically reducing their thermal noise, to the point that quantum mechanical effects of the phonons emerge. In the quantum regime, a traveling acoustic phonon mode in an optical fiber can serve as a memory or intermediary for quantum states of light. Photons can be coherently converted to phonons and back, effectively storing the photonic state in a mechanical degree of freedom. Coupling light to acoustic phonons has already been used to demonstrate all-optical signal storage and delay in classical contexts. For example, light pulses can be fully converted into acoustic waves and retrieved after a storage interval.<sup>44</sup> If those acoustic waves are in a near-ground-state (low-noise) condition, one could perform the same feat at the single-photon level, realizing a broadband quantum

memory at telecom wavelengths. Such a memory would be reversible and coherent, capable of storing quantum information (amplitude and phase of a single-photon wavepacket) for microsecond durations.<sup>44</sup> Moreover, a low-phonon-temperature waveguide could function as a quantum optical delay line or buffer with minimal added noise, preserving quantum states during the delay.

If the traveling acoustic mode approaches its ground state, one can begin to prepare and manipulate non-classical phonon states. This could include generating single phonons on demand, phonon Fock states, or entangled states between phonons and photons. One exciting prospect is creating entangled states between a traveling phonon and a propagating photon. The cooled phonon mode would have low enough noise to allow high-fidelity entanglement generation via stimulated scattering processes. In fact, a recent proposal by Zhu et al. (2024) suggests that even without ground-state preparation, one can generate entangled photon–phonon pairs in a fiber using pulsed Brillouin processes.<sup>45</sup> Their scheme relies on a two-pump configuration to create correlated Stokes and anti-Stokes photons along with a phonon, and remarkably it does not require the phonon to start in the ground state. If the phonon mode were already near its ground state, one can imagine the entanglement rates and purity could be even higher. Beyond two-party entanglement, cooled traveling phonons could facilitate multi-mode quantum state engineering, for example, creating squeezed phonon states.<sup>46</sup> The ability to manipulate a continuum of phonon modes at the quantum level might also enable quantum acoustic signal processing, where information is encoded in phononic wavepackets and processed (filtered, delayed, interfered) with quantum coherence.<sup>47</sup>

Achieving ground-state cooling of a macroscopic, traveling-wave phonon mode will likely require a hybrid approach: combining conventional cryogenics with our laser cooling techniques. Cavity optomechanics experiments regularly precool the device to liquid helium temperatures before applying laser cooling to reach the quantum ground state,<sup>48</sup> and the same idea applies to continuous systems. Placing the optical fiber or waveguide in a cryostat lowers the baseline phonon occupancy, dramatically reducing the gap to the ground state. Laser cooling could then be applied to overcome the remaining thermal quanta.

### 2.5.5 Conclusion

In conclusion, the work presented in this chapter establishes a robust foundation for fiber-based optomechanical cooling of traveling-wave phonons, pushing the boundaries of what can be achieved in continuous-wave Brillouin systems. By demonstrating Brillouin anti-Stokes cooling in a carefully engineered LCOF platform, we have shown that laser cooling need not be relegated to highly specialized chip waveguides or microresonators. The advantageous optomechanical properties of the LCOF enabled clear evidence of laser cooling under ambient conditions and marks the first demonstration of its kind in a fiber format. This proof of prin-

ciple opens up numerous opportunities for fiber-based applications, including precision metrology, low-noise acousto-optic devices, and quantum information schemes requiring mechanically cooled states. By demonstrating the capacity to cool traveling-wave phonons in an optical fiber, this work lays the groundwork for future low-noise photonic devices and advanced acousto-optic technologies, marking a significant step toward quantum-limited acoustic control.



## Chapter 3

### A Coherently Stimulated Phonon Spectrometer

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#### 3.1 Abstract

We present a novel coherently stimulated Brillouin spectrometer utilizing a detuned pump-probe design that exploits a relaxation of phase-matching requirements at small lengths, enabling room-temperature traveling-wave phonon spectroscopy at the micrometer scale with sub-10 fW sensitivity. This approach overcomes the limitations of traditional stimulated Brillouin techniques, particularly regarding phase-matching constraints and spatial resolution. We validated our instrument's sensitivity with 1 cm of Ultra High Numerical Aperture 3 (UHNA3) fiber and 100  $\mu\text{m}$  of bulk carbon disulfide liquid, demonstrating its capability to measure Brillouin scattering in materials with low Brillouin gain or, with particular advantage, small effective lengths. This advancement opens new possibilities for nanometer-scale Brillouin spectroscopy and the development of nano-acousto-optic devices.

#### 3.2 Introduction

Brillouin scattering, the inelastic interaction between light and acoustic phonons, is a fundamental phenomenon for probing the mechanical and structural properties of materials at microscopic scales. In spontaneous Brillouin scattering, thermally excited acoustic phonons scatter incident light, causing frequency shifts that reveal information about the material's elastic properties and acoustic modes.<sup>13</sup> However, the weak signal inherent to spontaneous Brillouin scattering often demands long acquisition times and limits spatial resolution, posing challenges for high-resolution material characterization.

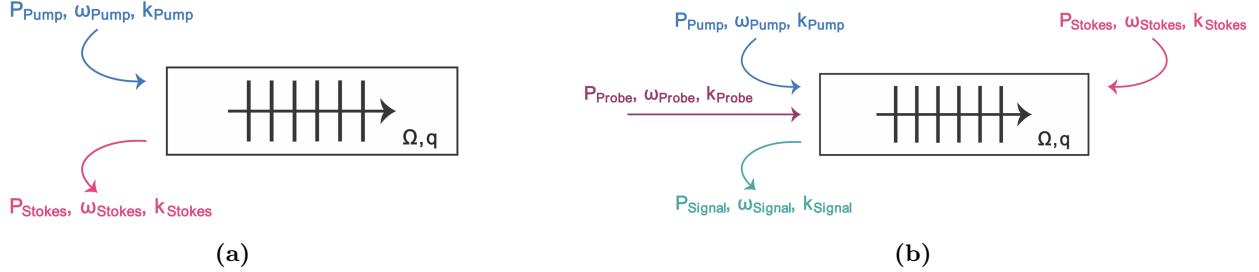
Stimulated Brillouin Scattering (SBS) uses intense optical fields to amplify the acoustic wave through a

nonlinear optical process.<sup>49</sup> In SBS, a strong pump laser interacts with a counter-propagating Stokes wave in the medium, generating acoustic phonons via electrostriction. As the phonon population grows, it further amplifies the Stokes wave. In turn, that amplified Stokes wave interferes with the pump and reinforces the acoustic field, creating a positive feedback loop that drives exponential amplification. This mechanism enables more efficient excitation and detection of acoustic phonons and underpins numerous applications in optical signal processing, sensing, and high-resolution spectroscopy including mechanobiology.<sup>50–59</sup>

However, conventional SBS techniques struggle with short samples or materials of inherently low Brillouin gain and strict phase matching requirements continue to pose a challenge.<sup>60–62</sup> Because the Stokes amplitude depends on the product of the Brillouin gain coefficient, pump power, and interaction length, small volumes often yield weak signals unless extremely high optical powers are used. Moreover, while backward SBS sends the scattered wave in the opposite direction of the pump, parasitic reflections and pump leakage can still obscure the Stokes signal, demanding careful optical isolation and sometimes elaborate filtering. These constraints make it difficult to measure thin films and micro- and nanoscale devices, particularly if high optical intensities risk damaging sensitive samples. As a result, standard SBS approaches are not easily adapted to sub-centimeter lengths or low-gain media, prompting the need for new methods that maintain high sensitivity in short interaction regions.

To overcome these challenges, researchers have explored various approaches.<sup>63–68</sup> Techniques based on optical cavities increase the effective interaction length, but require precise alignment and are sensitive to environmental fluctuations.<sup>69</sup> Forward Brillouin scattering methods, such as those demonstrated by Kittlaus et al.,<sup>70</sup> offer relaxed phase-matching conditions but introduce increased modal complexity. Meanwhile, coherent probe beam amplification can boost sensitivity, yet it can introduce additional noise and complexity because phase noise in laser sources can cause significant gain fluctuations.<sup>71</sup>

Here, we demonstrate a detuned pump–probe design that relaxes the usual phase-matching constraints at short lengths. This approach offers a new route to measure traveling-wave phonons in sub-centimeter or even micrometer-scale samples at room temperature with unprecedented sensitivity. We demonstrate the capabilities of the instrument by measuring Brillouin scattering in 1 cm of UHNA3 fiber and 100 μm of bulk carbon disulfide (CS<sub>2</sub>) liquid. These measurements highlight the instrument’s ability to characterize materials with low Brillouin gain or small effective lengths. The development of this Coherently stimulated Brillouin Scattering (CoBS) spectrometer opens new avenues for nanometer-scale Brillouin spectroscopy and advances the characterization and design of nano-acousto-optic devices. It holds promise for pushing research in material science, photonics, and sensing technologies toward higher spatial resolution and sensitivity, marking a significant step toward practical, room-temperature Brillouin-based spectroscopy and sensing solutions.



**Figure 3.1:** (a) Illustration of standard (three-wave) stimulated Brillouin scattering (SBS), showing a single optical pump field  $P_{\text{Pump}}$ , the backscattered Stokes field  $P_{\text{Stokes}}$ , and the acoustic (phonon) wave of frequency  $\Omega$  and wavevector  $q$ . Here,  $\omega_i$  and  $k_i$  represent the frequency and wavevector of the optical fields, respectively. (b) Illustration of Coherently stimulated (four-wave) Brillouin Scattering (CoBS), where an additional optical probe  $P_{\text{Probe}}$  mixes with the pump to generate both the Stokes wave and a new signal  $P_{\text{Signal}}$ , mediated by the same acoustic wave. We refer to this as four-wave because four optical fields (pump, probe, Stokes, signal) participate, although one may also count the acoustic wave as a “fifth” wave.

### 3.3 Theoretical Framework

#### 3.3.1 Coherently Stimulated Four-Wave Brillouin Scattering

Traditional SBS, illustrated in Figure 3.1a, is a three-wave mixing process in which incident pump laser light of frequency  $\omega_{\text{Pump}}$  inelastically scatters from a traveling-wave phonon of frequency  $\Omega$  to produce light that is frequency-shifted by the phonon frequency. In the Stokes process the phonon is retreating from the incident laser light and the scattered light is shifted down in frequency ( $\omega_{\text{Stokes}} = \omega_{\text{Pump}} - \Omega$ ). Spatial overlap of the backscattered light with the incident laser light allows for interference of the two optical fields to produce a frequency at the difference of the two ( $\omega_{\text{Pump}} - \omega_{\text{Stokes}}$ ). Since this difference frequency is exactly equal to the frequency of the acoustic field  $\Omega$ , the beating of the incident pump light with the backscattered Stokes light produces an electrostrictive reinforcement of the acoustic wave. This driving of the acoustic wave in turn increases the scattering rate of the incident pump light, producing a positive feedback process and an exponential increase of the amplitude of the backscattered Stokes wave.

Figure 3.1b illustrates coherently stimulated four-wave Brillouin scattering for the Stokes process, mediated by an acoustic wave. We introduce a strong, controlled external Stokes wave of frequency  $\omega_{\text{Stokes}}$  that drives electrostrictive reinforcement of the acoustic field in the medium. The backscattered Stokes light is normally collected in an SBS process, but the external Stokes laser overwhelms it. To resolve this, we inject light of a distinct frequency  $\omega_{\text{Probe}}$  from an additional external laser which copropagates with the Pump and backscatters in the medium from the strongly driven acoustic field. This produces backscattered Signal light to be collected ( $\omega_{\text{Signal}} = \omega_{\text{Probe}} - \Omega$ ) which is spectrally distinct from the high-powered Stokes laser light.

To describe this interaction and characterize the performance of the instrument, we derive the coupled-

wave equations for the four-wave mixing process in Appendix B.1. These equations describe the relationship between the optical fields and the acoustic field in the material and result in the following expression for the scattered power of the backscattered signal,

$$P_{\text{Sig}} = \frac{1}{4}(G_B L)^2 P_P P_S P_{\text{Pr}} \text{sinc}^2 \left( \frac{\Delta k L}{2} \right), \quad (3.1)$$

where  $P_P$ ,  $P_S$ , and  $P_{\text{Pr}}$  are the powers of the pump, Stokes, and probe lasers, respectively.  $G_B$  is the effective Brillouin gain, given by

$$G_B = \frac{g_0}{A_{\text{eff}}^{\text{ao}}} \frac{\left(\frac{\Gamma_B}{2}\right)^2}{(\Omega - \Omega_B)^2 + \left(\frac{\Gamma_B}{2}\right)^2}, \quad (3.2)$$

with the on-resonance gain factor of the material given by

$$g_0 = \frac{\gamma_e^2 \omega^2}{n v c^3 \rho_0 \Gamma_B}. \quad (3.3)$$

Here,  $\gamma_e$  is the electrostrictive constant,  $\omega$  is the pump frequency,  $n$  is the refractive index of the material,  $v$  is the sound speed of the material,  $c$  is the speed of light,  $\rho_0$  is the mean density of the material, and  $\Gamma_B$  is the Brillouin linewidth, or phonon dissipation rate, of the material. In Equation 3.2,  $\Omega_B$  is the resonant Brillouin frequency of the material,  $A_{\text{eff}}^{\text{ao}}$  is the effective acousto-optic area of overlap within the material,  $\Delta k$  is the wavevector mismatch between the optical fields, to be discussed next, and  $L$  is the effective length of the material.

### 3.3.2 Phase Matching Relaxation

In all nonlinear optical processes, efficiency is maximized when phase matching conditions are satisfied. A frequency mismatch (energy unconservation) or a wavevector mismatch (momentum unconservation) each result in drastically reduced efficiency of a given process.<sup>72</sup> This can be seen by Equation 3.1, where the wavevector mismatch,  $\Delta k$ , is contained within a  $\text{sinc}^2$  function. This  $\text{sinc}^2$  term thereby defines the phase matching bandwidth of the system, notably scaling with effective interaction length  $L$ .

We apply this wavevector mismatch allowance to the pump and probe waves ( $\Delta k = k_{\text{Pump}} - k_{\text{Probe}}$ ) so that the backscattered signal is different than the applied Stokes wave. This choice allows for selection of the signal and rejection of the Stokes with a bandpass filter. Expressed in terms of wavelengths, this gives

$$\Delta k = \frac{4\pi n \Delta \lambda}{\lambda_{\text{Pump}} \lambda_{\text{Probe}}} \approx \frac{4\pi n \Delta \lambda}{\lambda_{\text{Pump}}^2}. \quad (3.4)$$

We can apply this to the phasematching bandwidth term to find the fraction of maximum scattered power,  $\Phi$ , that can be expected for a given interaction length,  $L$ , and phase mismatch  $\Delta\lambda$  between the pump and probe,

$$\Phi \equiv \text{sinc}^2 \left( \frac{2\pi n \Delta \lambda L}{\lambda_{\text{Pump}}^2} \right). \quad (3.5)$$

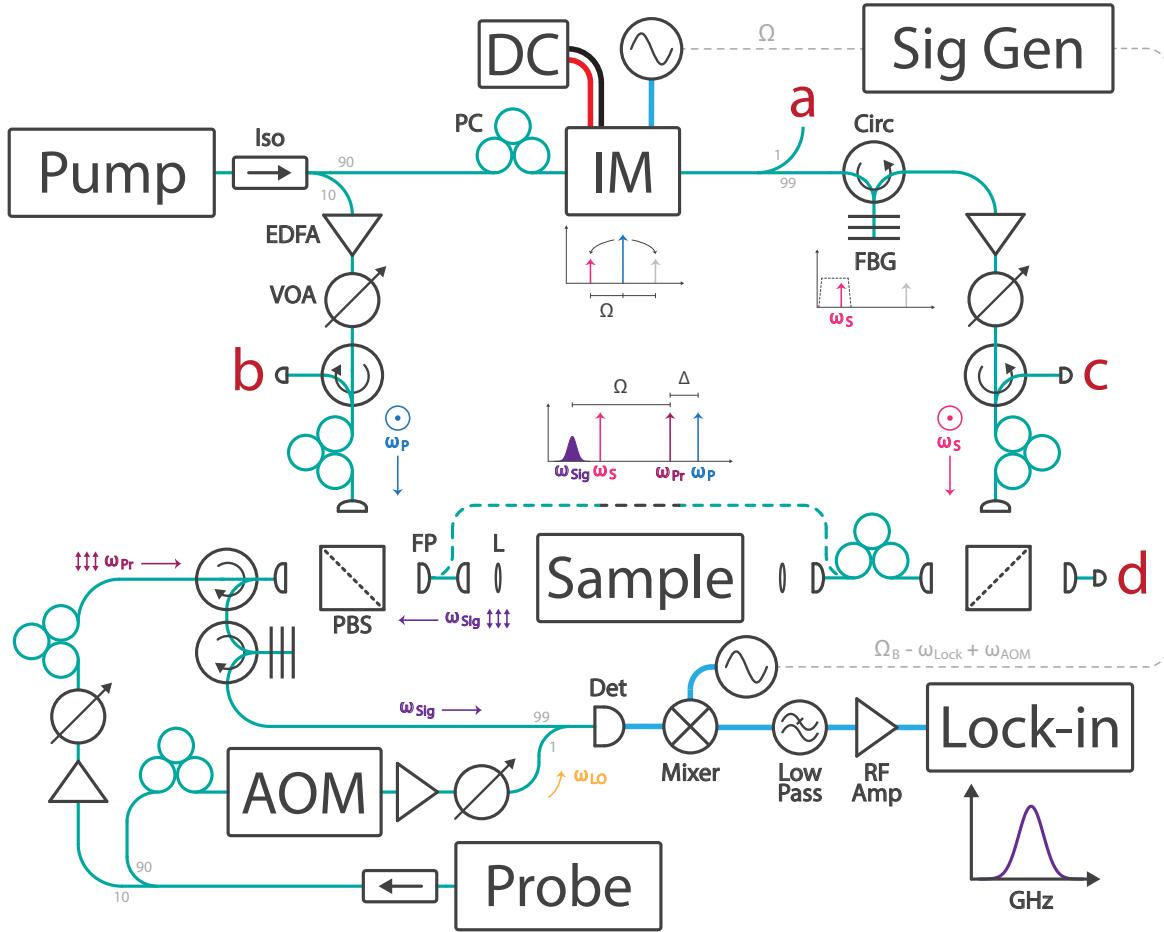
Using this expression for  $\Phi$ , we see that for an effective length of 1 m, a wavelength mismatch of only 0.6 pm from a typical wavelength of 1.55  $\mu\text{m}$  pump light in UHNA3 fiber drops the scattered power to half of the maximum. However, for shorter effective lengths the wavevector mismatch becomes more forgiving; a 36 pm mismatch preserves 82.5% of the maximum signal for a length of 1 cm under identical conditions. This separation translates to about 4.5 GHz, providing enough spectral separation for the backscattered signal to be isolated from the applied Stokes light.

Furthermore, for decreasing lengths, Equation 3.5 predicts an increase in the fraction of maximum signal produced, given equivalent pump–probe detuning, as the  $\text{sinc}^2$  function is sampled closer to its peak center. Alternatively, as length decreases, the probe may be further detuned from the pump and still achieve the same fraction of the maximum signal as for longer lengths, perhaps offering a slight advantage in noise reduction. It should be noted that the scattered power, as given by Equation 3.1, scales with the square of the effective length. Thus, while smaller lengths allow for the ability to capture a larger fraction of this maximum scattered power, the actual amount of scattered power decreases dramatically as length decreases.

## 3.4 Methods

### 3.4.1 Instrument Design

Figure 3.2 shows the instrument’s design. A pump and Stokes wave is synthesized from a single tunable laser source for coherent stimulation of a sample. The pump wave ( $\omega_{\text{Pump}}$ ) is amplified by an Erbium-Doped Fiber Amplifier and passed through a Variable Optical Attenuator for power control. The output is then polarization-controlled to reflect at a polarizing beam splitter for injection into the sample. For Stokes synthesis, an alternating current signal ( $\Omega$ ) is supplied to an fiber-optic intensity modulator with carrier frequency nulled and a tunable filter is used to select the lower-frequency Stokes side band ( $\omega_{\text{Pump}} - \Omega$ ). This Stokes light is then amplified by an EDFA, passed through a VOA, and polarization-controlled to reflect



**Figure 3.2:** Design schematic of the Coherently stimulated Brillouin Spectrometer (CoBS). A tunable Continuous Wave (CW) laser at approximately  $1.55\text{ }\mu\text{m}$  emits light that passes through an isolator (Iso) and a splitter, diverting 10% to a 27 dBm Erbium-Doped Fiber Amplifier (EDFA) followed by a Variable Optical Attenuator (VOA). This pump light ( $\omega_P$ ) is polarization-controlled to reflect off a polarizing beam splitter (PBS) and is recoupled to fiber via a fiber port (FP), then directed to the sample either by direct fiber coupling or through a pair of FPs and lenses (L) for free-space samples. After passing through the sample, the pump light traverses a corrective polarization controller that mitigates fiber twists and bends before reflecting off a second PBS, where it is routed to port (c) for power monitoring. To synthesize the Stokes wave, a 90% split from the original pump is processed through a fiber-optic intensity modulator (IM) and a fiber Bragg grating (FBG), generating a Stokes sideband downshifted from the pump by  $\Omega$ . This frequency shift is swept via a signal generator to capture  $\Omega_B$ . A 99/1 splitter provides a tap at port (a) to optimize Stokes synthesis. The Stokes wave ( $\omega_S$ ), amplified by a 1 W EDFA and VOA-controlled, counter-propagates along the pump path and is monitored at port (b). A second tunable CW laser, detuned from the pump, generates the probe wave ( $\omega_{Pr}$ ), which is amplified by a 1 W EDFA, attenuated variably, and polarization-controlled to pass through the initial PBS where it is incident on the sample. Backscattered signal light ( $\omega_{Sig}$ ) transmits back through the PBS, while unscattered probe light transmits to a power meter at port (d). A circulator parts the signal from the probe path, with an FBG filtering out any unwanted noise or Stokes light. Finally, the signal is heterodyned with an EDFA-amplified, acousto-optic modulator (AOM)-shifted local oscillator (LO) derived from the probe laser and directed to a photodiode for detection. The resulting radio frequency (RF) signal is mixed with an alternating current (AC) LO supplied by the signal generator, which sweeps synchronously with the Stokes synthesis frequency, and collected by a lock-in amplifier for data processing.

at a second PBS for counter-propagation to the pump through the sample.

A separate tunable laser is used to supply a probe wave ( $\omega_{\text{Probe}} = \omega_{\text{Pump}} + \Delta k$ ) and local oscillator. Probe light is amplified by an EDFA and passed through a VOA and a polarization controller aligns the polarization for transmission through the first PBS whereby it copropagates with the pump through the sample. Backscattered light exits the sample and transmits back through the first PBS, whereas the orthogonally polarized Stokes light reflects at this same point to be diverted to a tap for power monitoring. The backscattered signal ( $\omega_{\text{Signal}} = \omega_{\text{Probe}} - \Omega$ ) then routes through two subsequent circulators for spectral filtering by a 5 GHz bandpass tunable filter. This filter allows the desired backscattered signal to pass while rejecting any reflected probe light as well as any reflected, transmitted, or backscattered light from the pump or Stokes waves that was not already diverted by the PBS.

The filtered signal then heterodynes via a 99-1 splitter with the LO which is frequency-upshifted by an acousto-optic modulator ( $\omega_{\text{LO}} = \omega_{\text{Probe}} + \omega_{\text{AOM}}$ ) and controlled to be copolarized with the signal. Of the resulting frequencies from the heterodyne process, only the difference frequency term is considered, as all others are beyond the range of detection. This heterodyned signal ( $\omega_{\text{Signal}} = \Omega + \omega_{\text{AOM}}$ ) is then captured by a photodiode detector and heterodyned again by a radio frequency mixer with a second AC signal ( $\Omega + \omega_{\text{AOM}} - \omega_{\text{Lock}}$ ), where  $\omega_{\text{Lock}}$  is a fixed-frequency to be detected by a lock-in amplifier set to this frequency after being passed through a low-pass filter and amplified by an RF amplifier. Synchronous sweeping of both AC signals, each involving  $\Omega$ , allows for  $\omega_{\text{Lock}}$  to remain fixed throughout measurement over a frequency range.

### 3.4.2 Experimental Techniques

We optimized the signal-to-noise ratio (SNR) of the instrument through specific design choices and device settings. Our setup simultaneously generates pump, Stokes, and probe optical fields for coherently stimulated Brillouin scattering. The pump laser provides  $\sim 45$  mW total output, of which 10% is split and amplified to  $\sim 0.5$  W for the pump field; the remaining 90% is frequency-shifted and amplified to  $\sim 1$  W for the Stokes field. Likewise, the probe laser also outputs  $\sim 45$  mW, with 10% amplified to  $\sim 1$  W for the probe field and the remaining 90% reserved for the LO. To combine the backscattered signal and LO with minimal loss, we use a 99/1 splitter instead of a typical 50/50, preserving 99% of the signal. The LO is therefore amplified to  $\sim 230$  mW so the total optical power at the detector remains below the 2.4 mW damage threshold. After detection, the electronic signal is mixed with a 17 dBm AC reference and further amplified by 23 dBm before input to the lock-in amplifier. We find that running both the pump and probe lasers in “whisper” mode (as opposed to “dither”) significantly enhances the measured SNR.

We use a Zurich Instruments HF2LI 50 MHz lock-in amplifier whose demodulator settings are carefully tuned to maximize SNR. A 10 MHz reference clock from the signal generator is fed into the lock-in to synchronize timing. The input-signal range, which sets the analog input amplifier’s gain, should exceed the measured signal (including any direct current (DC) offset) by at least a factor of two. This is best achieved by using the lock-in software’s auto feature, which continuously adjusts the range over a rolling 100 ms window. We set the input coupling to AC, insert a high-pass filter to remove DC components, and choose  $1\text{ M}\Omega$  input impedance. For noise suppression, we also engage the lock-in’s eighth-order low-pass filter (roll-off  $48\text{ dB Oct}^{-1}$ ) and sample the data at  $1.84\text{ MSa s}^{-1}$ , the maximum rate available.

Further SNR improvements are gained by narrowing the lock-in’s low-pass filter bandwidth to match both the sub-Hz natural linewidth of the heterodyne signal and the thermally-driven frequency drift of the apparatus. After a  $\sim 30$  min warm-up, we observe less than 100 Hz of drift in the detected signal frequency, so we typically set a 100 Hz low-pass bandwidth for multi-hour measurements. For shorter scans ( $< 15$  min), we can reduce this bandwidth to 40 Hz if needed. In addition to linewidth variability, the signal’s center frequency can shift due to thermal changes in the AOM and related electronics. Although  $\Omega$  is nominally controlled to sub-hertz precision by the signal generator, our AOM’s shift  $\omega_{\text{AOM}}$  drifts from 40 MHz up to  $\sim 40.000\,82$  MHz over roughly 30 min. Once at thermal equilibrium, the AOM remains stable within  $\pm 50$  Hz, enabling a reliable lock-in frequency reference and minimal filter bandwidth. This stability is crucial for repeatable, high-resolution Brillouin measurements.

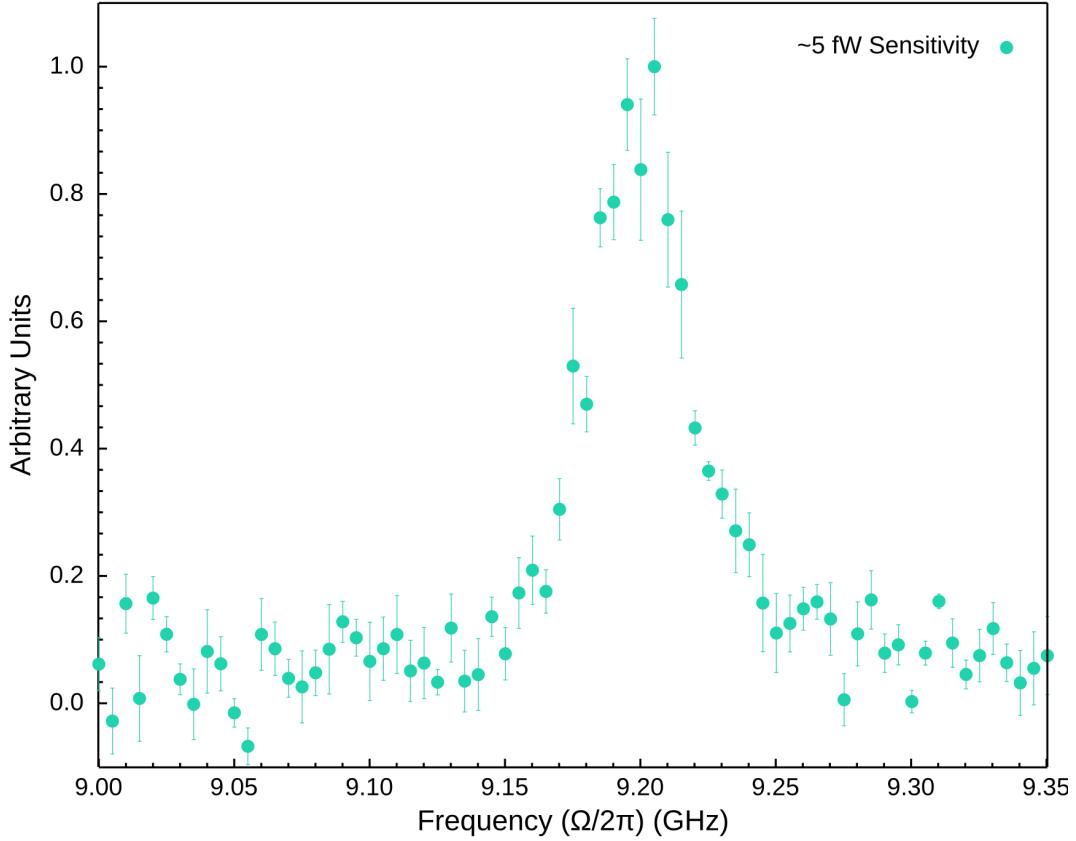
## 3.5 Results

### 3.5.1 Instrument Sensitivity

We begin by testing the sensitivity of the instrument as a way of defining a performance metric for the instrument which can be used to indicate what material, power, and length combinations might be possible to measure. From Equation 3.1, the sensitivity of the instrument is the minimum scattered power,  $P_{\text{Sig}}$ , to produce a statistically significant measurement. To determine this, we target a specific length,  $L$ , of a sample of known effective Brillouin gain,  $G_B$ . We keep the pump-probe detuning,  $\Delta\lambda$ , constant across measurements and record the pump, Stokes, and probe optical powers to calculate the scattered power. Starting with sufficient optical powers to produce a clearly distinguishable measurement, we gradually reduce the optical powers until the sensitivity floor is reached.

To serve as our sensitivity testbed, we prepared 1 cm of Nufern’s Ultra High Numerical Aperture 3 fiber, a well-studied fiber with known effective Brillouin gain.<sup>73</sup> Additionally, UHNA3 fiber offers several properties that make it ideal for this task of unambiguous detection of the Brillouin signal as it diminishes with each

### CoBS Measurement: 1 cm UHNA3



**Figure 3.3:** Observed spectra demonstrating  $P_{\text{Signal}} \approx 5 \text{ fW}$  sensitivity of the instrument. Experimental parameters for this measurement are tabulated in Table 3.1. In obtaining the spectra, five repeated measurements of both the signal and background (probe off) were collected at a 100 Hz resolution bandwidth (RBW), dwelling for 1 s at each 5 MHz frequency step. Plotted is the resulting background-subtracted spectrum. Uncertainties represent  $1\sigma$  standard error of the mean. By comparing the peak amplitude at resonance to the off-resonance baseline, we estimate an SNR greater than 5, which, under a normal-noise assumption, corresponds to a  $5\sigma$  significance level (99.99994% confidence).

subsequent reduction in optical powers. First, it offers a Brillouin shift that is spectrally far from that of the single mode fiber 28 (SMF-28) which constitutes much of the instrument. This ensures that the Brillouin response of the sample is not conflated with the Brillouin response of the instrument itself. Additionally, the core of UHNA3 fiber features a high concentration of germanium which improves the optical and acoustic guidance in the fiber as a result of the large refractive index difference between core and cladding. Finally, UHNA3 fiber offers a high optomechanical nonlinear response, with an effective Brillouin gain of  $0.6 \text{ W}^{-1} \text{ m}^{-1}$  measured at room temperature.<sup>73</sup> This gain factor is larger than that of SMF-28 by an order of magnitude.

<sup>74</sup>

Figure 3.3 presents a measurement in which the instrument's sensitivity reaches  $P_{\text{Sig}} = 2 \text{ fW}$ . Table 3.1 provides the experimental parameters used to gather this measurement and calculate the sensitivity. Each

trace is the average of five consecutive scans, and an average of five background scans has been subtracted to isolate the signal. Error bars represent the standard error ( $1\sigma$  of the mean). By comparing the peak amplitude at resonance to the off-resonance baseline, we estimate an SNR greater than 5. Under a normal-noise assumption, an SNR of 5 corresponds to a  $5\sigma$  significance level (99.7% confidence). Achieving this 5 fW threshold demonstrates the feasibility of measuring weaker signals in materials with lower Brillouin gain or smaller effective lengths.

$G_B$ (W $^{-1}$ m $^{-1}$ )	L (m)	P <sub>P</sub> ( $\mu$ W)	P <sub>S</sub> ( $\mu$ W)	P <sub>Pr</sub> (mW)	$\Delta\lambda$ (pm)	$\Phi$
0.6	0.01	506	504	2.01	20	0.90

**Table 3.1:** Experimental parameters used in observing  $P_{\text{Sig}} \approx 5$  fW sensitivity of the Coherently stimulated Brillouin Spectrometer (CoBS) instrument. The resulting spectra is shown in Figure 3.3.

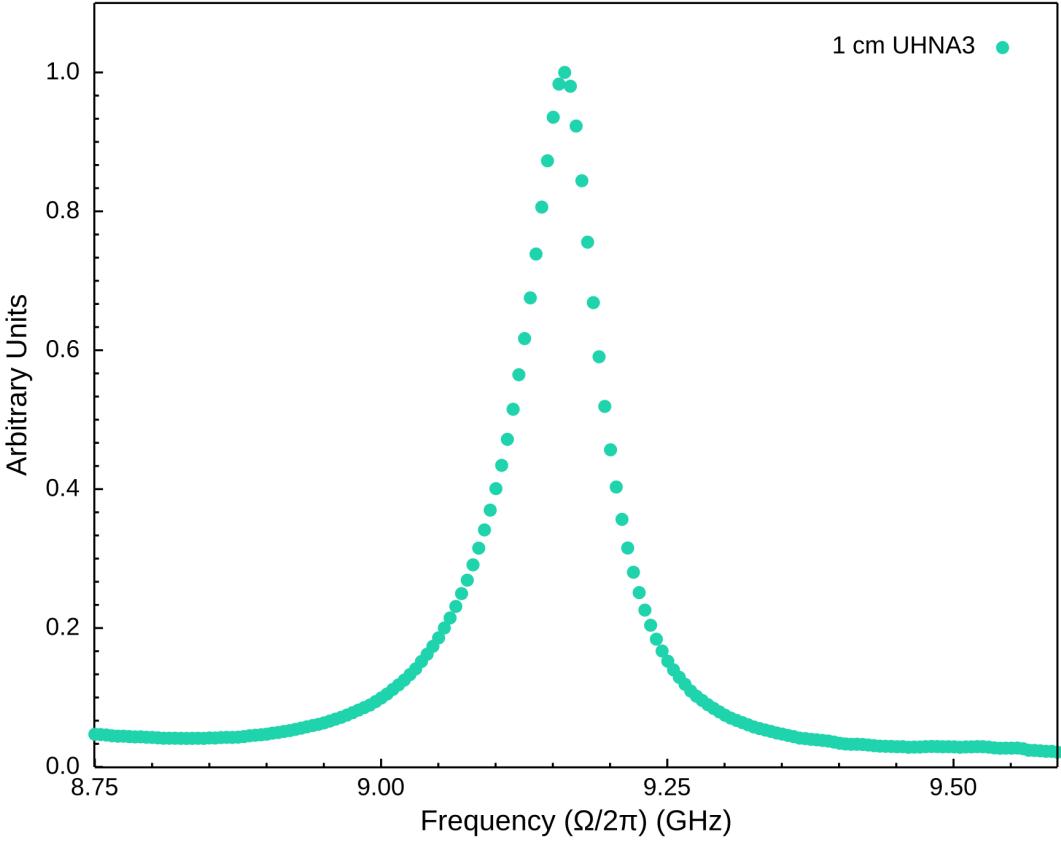
### 3.5.2 Measurements

We demonstrate the capabilities of the instrument on two common sample classes: fiber and bulk material. For a fiber sample we again choose UHNA3 for its higher nonlinear response and excellent optical and acoustic guidance. In contrast to the sensitivity measurements, we now seek to demonstrate the full measuring capabilities of the instrument and so apply all available optical power ( $\sim 1.5$  W) to maximize the backscattered signal from the sample. We target the same 1 cm segment of UHNA3 fiber as was used for determining sensitivity.

Figure 3.4 shows the spectral profile captured for 1 cm of UHNA3 fiber, revealing the expected lorentzian profile consistent with Equation 3.2. The peak amplitude of the spectra occurs at 9.1704 GHz, indicating the Brillouin resonance frequency of the longitudinal traveling-wave mode in the fiber. The full-width half-max (FWHM) linewidth of the measurement is 80 MHz and provides a measure of the phonon dissipation rate. Both values match what is seen in the literature for SBS measurements of UHNA3 fiber.<sup>73</sup> The data shown are a background-subtracted average of five successive measurements taken over ten minutes with error bars corresponding to  $1\sigma$  of the mean.

To achieve this measurement of UHNA3 fiber, the instrument design was altered to include only fiber-coupled segments connecting the fiber ports between the two PBSs. We set the pump laser wavelength to 1549.000 nm and the probe laser wavelength to 1549.020 nm, giving a frequency mismatch of approximately 2.5 GHz. The pump–probe mismatch is chosen to be only as large as needed to allow the edge of the passband of the probe filter to split the backscattered pump and probe light, thus rejecting any backscattered light from the pump laser and accepting only the backscattered signal from the probe laser. We placed the

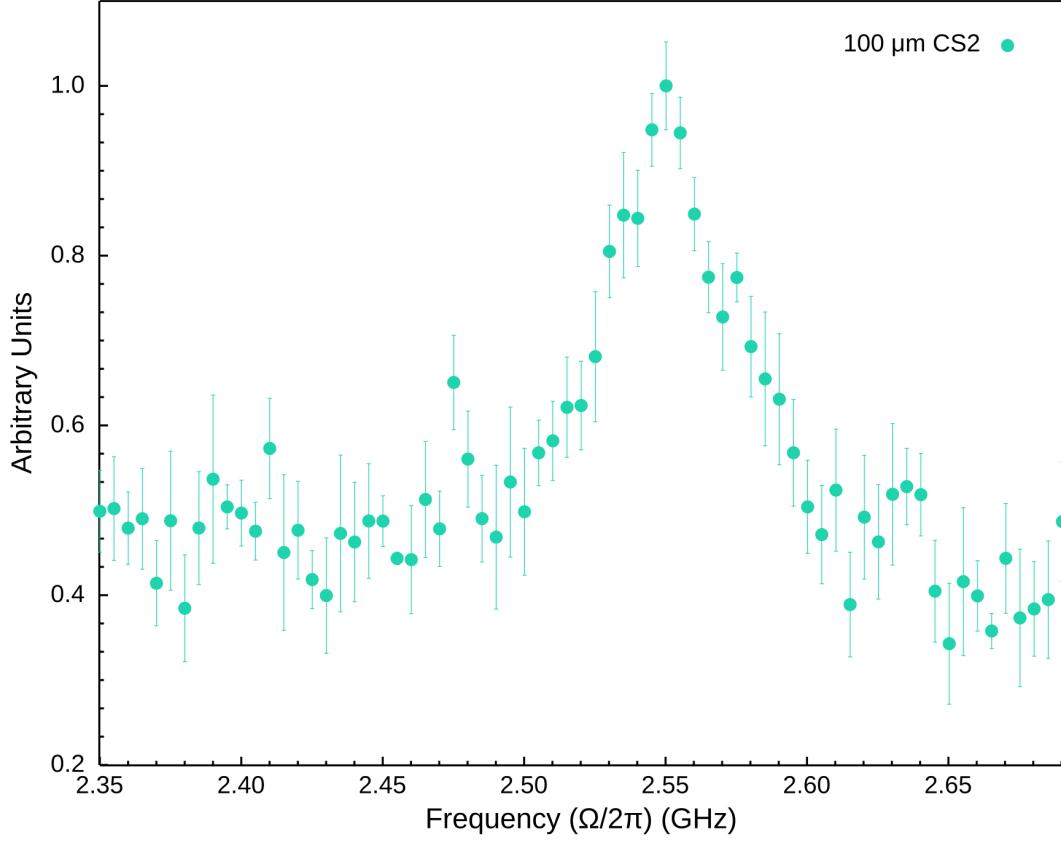
### CoBS Measurement: 1 cm UHNA3



**Figure 3.4:** Observed spectra of 1cm UHNA3 fiber captured at maximum operating optical powers  $P_P P_S P_{Pr} \sim 0.25 \text{ W}^3$ . The data exhibits the familiar Lorentzian profile for frequency-dependent dissipation probed by Brillouin scattering. This response is reflected in the CoBS process gain  $G_{\text{CoBS}} = \frac{1}{4}(G_{BL})^2 P_P P_S P_{Pr}$  given in Equation 3.1, with Equation 3.2 for effective Brillouin gain offering the off-resonant Lorentzian lineshape. In obtaining the spectra, five repeated measurements of both the signal and background (probe off) were collected at a 100 Hz RBW, dwelling for 1 s at each 5 MHz frequency step. Plotted is the resulting background-subtracted spectrum, with  $1\sigma$  uncertainties smaller than the data point markers.

Stokes filter at 1549.073 nm, an offset of approximately 9.18 GHz from the pump laser to capture the Stokes sideband from the intensity modulator. This corresponds to the center of the measured frequency range and was chosen to allow the Stokes sideband output from the intensity modulator to remain within the pass band of the Stokes filter as the RF signal fed to the intensity modulator is swept through the full measurement range. The probe filter was set to 1549.109 nm, an offset of approximately 11.18 GHz from the probe laser, to capture the Stokes-shifted backscattered signal from the probe. The center frequency of the backscattered signal is of course shifted 9.18 GHz from the probe laser, however an extra offset of 2 GHz is chosen for improved rejection of any pump light as the pass band of our filter extends approximately 2.5 GHz on either side of center.

### CoBS Measurement: 100 $\mu\text{m}$ CS<sub>2</sub>



**Figure 3.5:** Observed spectra of 100  $\mu\text{m}$  liquid CS<sub>2</sub> captured at maximum operating optical powers  $P_{\text{P}}P_{\text{S}}P_{\text{Pr}} \sim 0.25 \text{ W}^3$ . Within the uncertainties, the data exhibits the familiar Lorentzian profile for frequency-dependent dissipation probed by Brillouin scattering. This response is reflected in the CoBS process gain  $G_{\text{CoBS}} = (G_{\text{BL}})^2 P_{\text{P}}P_{\text{S}}P_{\text{Pr}}$  given in Equation 3.1, with Equation 3.2 for effective Brillouin gain offering the off-resonant Lorentzian lineshape. In obtaining the spectra, five repeated measurements of both the signal and background (probe off) were collected at a 100 Hz RBW, dwelling for 1 s at each 5 MHz frequency step. Plotted is the resulting background-subtracted spectrum. Uncertainties represent  $1\sigma$  standard error of the mean.

For a free-space bulk example we target liquid CS<sub>2</sub> for its exceptionally high Brillouin gain factor of 1.5 m GW<sup>-1</sup>.<sup>13</sup> Figure 3.5 reveals the Brillouin signal of bulk CS<sub>2</sub> liquid contained in a 100  $\mu\text{m}$  path length cell. To our knowledge, measurement of longitudinal Brillouin scattering at this scale has not been reported in the literature. A scattered power comparison would reveal that achieving such a measurement using traditional SBS techniques would require excessively high optical powers or cooling the material to cryogenic temperatures, which, of course, would be prohibitive for carbon disulfide in the liquid state.

For this measurement of CS<sub>2</sub>, the pump and probe laser wavelengths were set to 1548.808 nm and 1548.898 nm, respectively. The short path length of the sample significantly broadens  $\Phi$ , the sinc<sup>2</sup> term defining the phasematching bandwidth, allowing for further separation of the pump and probe wavelengths

for improved signal isolation without significant reduction in scattered power of the signal produced in the  $\text{CS}_2$ . Specifically, the additional pump–probe wavelength separation of 70 pm employed for this measurement compared to the UHNA3 measurement results in a negligible 0.045% reduction in scattered power. This additional separation contributes meaningfully, however, to improved rejection of pump light by the probe filter and thus higher SNR of the signal.

Placement of the Stokes filter is critical for measurements of materials that give small Brillouin shifts, such as with  $\text{CS}_2$  2.55 GHz shift. We offset our 5 GHz bandwidth Stokes filter an additional 2 GHz to ensure the nearby carrier signal and anti-Stokes sideband from the intensity modulator are rejected and only the Stokes sideband is allowed to pass. For the measurement shown in Figure 3.5, this corresponds to a Stokes and probe filter placement of 1548.844 nm and 1548.934 nm, respectively.

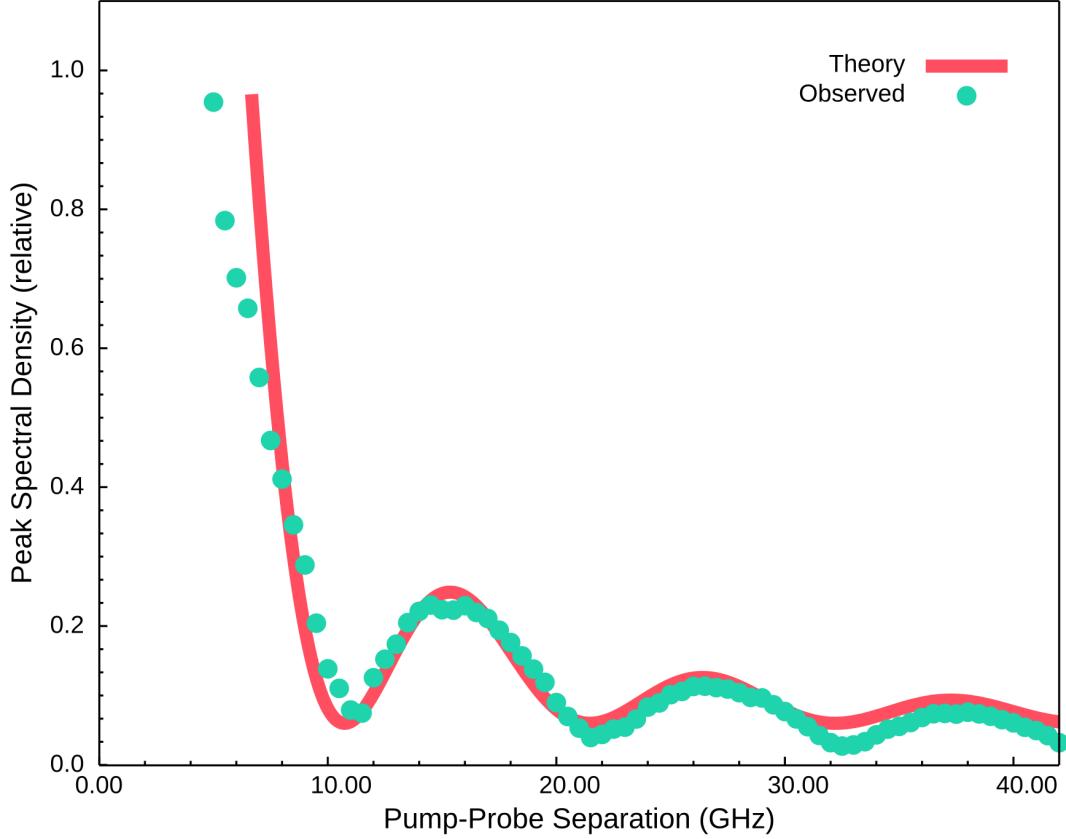
### 3.5.3 Phase Matching Bandwidth

To characterize the phase matching tolerance of the instrument for a given length of sample, we performed an additional experiment whereby we took a series of measurements of 1 cm of UHNA3 at constant optical powers while letting the detuning of the pump and probe lasers vary. In the language of Equation 3.1, this experiment holds  $G_B$ ,  $L$ ,  $P_P$ ,  $P_S$ , and  $P_{Pr}$  constant while letting  $\Delta k$  vary. For the experiment to support the validation of Equation 3.1, we would expect the peak amplitudes of these measurements to trace out a  $\text{sinc}^2$  profile, also given by Equation 3.5. Figure 3.6 shows the results of this experiment. We performed 75 measurements between 5 GHz and 42 GHz pump–probe detuning, at 0.5 GHz intervals. We found peak amplitudes by fitting each spectra with a Fano profile (see Section 3.5.4) and represented each peak as a data point in Figure 3.6. The theoretical  $\text{sinc}^2$  function matching the parameters used in the experiment is shown on the plot with a solid red line.

### 3.5.4 Fano-Resonant Asymmetries at Small Signals

Under certain conditions where the resonant Brillouin amplitude approaches the background continuum level, we observe an asymmetric, Fano-type line-shape.<sup>75–78</sup> These Fano distortions can shift the apparent peak frequency, complicate simple Lorentzian fitting, and affect the extracted linewidth in small-signal measurements.<sup>79</sup> To properly handle these occurrences, it is necessary to understand when they are likely to arise with this technique and how they may be corrected for or controlled. Fano resonances arise when a discrete resonance (in our case, the Brillouin mode) interferes with a continuum background (e.g., noise floor or broad, non-resonant scattering). When the resonance amplitude is no longer much larger than the continuum, the interference leads to an asymmetric line-shape described by the Fano formula,<sup>75</sup>

### 1 cm UHNA3 Phase-Matching Bandwidth



**Figure 3.6:** Spectral peaks of 75 observed spectra of  $(1.0 \pm 0.1)$  cm UHNA3 across a range of pump-probe detunings  $(\omega_{\text{Pump}} - \omega_{\text{Probe}})/2\pi$  from 5 GHz-42 GHz in 0.5 GHz increments. Values are obtained from uncertainty-weighted Fano fits of the observed spectra (see Appendix B.3). The solid theoretical line shows the expected theoretical trend (a  $\text{sinc}^2$  lineshape) given by Equation 3.5, plotted here for the experimental parameters used in collecting the data and a fiber length of  $\sim 0.9$  cm. Section B.3.1 in Appendix B.3 offers an extended discussion of the observed spectra and contains a plot of the full set of 75 spectra (Figure B.3.1). In obtaining each spectra, five repeated measurements of both the signal and background (probe off) were collected at a 100 Hz RBW, dwelling for 1 s at each 5 MHz frequency step. Uncertainty-weighted Fano fits were computed from the resulting background-subtracted spectra. Plotted here are the fit-extracted spectral peaks, with  $1\sigma$  uncertainties smaller than the data point markers.

$$I(\omega) \propto \frac{(q + \epsilon)^2}{1 + \epsilon^2}, \quad (3.6)$$

where  $\epsilon \equiv (\Omega - \Omega_B)/(\Gamma_B/2)$  is the dimensionless detuning from the Brillouin peak (measured in half the spectral linewidth) and  $q$  is the Fano asymmetry parameter. The Fano asymmetry parameter captures the ratio of the resonant scattering to the background scattering amplitudes as well as the relative phase of each. It can be defined as

$$q = \frac{(\text{resonant amplitude})}{(\text{background amplitude})} \cot(\Delta\phi), \quad (3.7)$$

where  $\Delta\phi$  is the phase difference between the oscillation driven by the discrete resonant mode and that of the background continuum.<sup>76, 80, 81</sup>

In the event of Fano interference, the complex background amplitude often varies slowly with energy and can be seen as having a fixed phase reference, while the resonant amplitude undergoes a rapid  $\pi$  phase change as the energy passes through the resonance.<sup>76</sup> When the resonant and background contributions are exactly out of phase at the resonance frequency (phase difference  $\Delta\phi = \pi/2$ , meaning destructive interference), the spectral line exhibits a dip to zero (anti-resonance) at the discrete resonance frequency, corresponding to  $q = 0$  in Equation 3.6. If they are exactly in phase ( $\Delta\phi = 0$  or  $\pi$ , fully constructive interference at resonance), the resonance appears purely Lorentzian without asymmetry and  $|q| \rightarrow \infty$ . For intermediate phase offsets, one lobe of the resonance is enhanced while the other is suppressed, yielding an asymmetric peak or dip with finite  $q$ . The sign of  $q$  indicates the direction of asymmetry (which wing of the resonance is enhanced). A positive  $q$  means the discrete mode *leads* the background in phase. In this case, the spectral profile has a sharp rise on the low-frequency side and a more gradual fall-off on the high-frequency side. Conversely, a negative  $q$  means the discrete mode *lags* the background in phase, leading to a flipped orientation featuring a sharp rise on the *high*-frequency side and a gradual fall-off on the *low*-frequency side.

We first noticed this behavior appearing in our small-length CS<sub>2</sub> data, where a small shift in probe wavelength revealed an asymmetric line-shape. For the measurements of 100 μm CS<sub>2</sub> (Figure 3.5) and 1 cm UHNA3 fiber at low power (Figure 3.3), the amplitude of the resonant Brillouin peak is on the order of that of the non-resonant continuum, giving a strong chance for Fano interference. Whenever  $\frac{I_{\text{res}}}{I_{\text{bkg}}} \approx 1$ , the parameter  $q$  as given by Equation 3.6 can become finite rather than  $\pm\infty$  in the limit that the background is negligible, and Fano interference arises. To explore this further, we performed a similar phase-matching experiment as was done for 1 cm UHNA3 (see Phase Matching Bandwidth subsection), this time with 1 mm of CS<sub>2</sub>. Results from this experiment are presented in Appendix B.3 and offer examples of line shapes with pronounced asymmetry and featuring clear characteristic morphologies associated with Fano interference. These pronounced distortions in spectral line shape for small signal measurements underscore the role Fano interference in small resonant amplitudes relative to the background.

Because our instrument offers sub-10 fW sensitivity signal amplitudes have the potential to approach the order of the background continuum amplitude. For this reason, and because the instrument offers an advantage in measurements of samples of short length (<10 m) (see Appendix B.2), Fano effects may often arise with usage of this technique and must be handled appropriately. This includes properly fitting data

with a Fano profile as opposed to a Lorentzian to ensure accurate capture of relevant parameters such as linewidth, resonant frequency, and peak amplitude. Beyond fitting and parameter extraction, it is important to be mindful that these effects are likely to occur in ambitious measurements at the limits of equipment sensitivity. Expectation and proper handling of Fano effects in measurements of this nature ensures that they may be more easily recognized and confirmed, as the data is likely to present a spectrum that deviates considerably from the standard Lorentzian. Appendix B.2 offers a comparative analysis of two example spectra featuring highly assymetric profiles fitted with a naïve Lorentzian vs. a more appropriate Fano function.

In some cases, and of particular interest for ambitious measurements, Fano interference may even *boost* the measured peak above the naïve Lorentzian amplitude (i.e., ‘amplify’ it) when  $q \neq 0$ . In principle, a Fano-type lineshape can exhibit a locally higher peak amplitude than a pure Lorentzian if the discrete Brillouin response constructively interferes with the background continuum near the resonance. Crucially, this does not represent net energy gain but rather a redistribution of intensity through interference. Although the resonance peak may appear taller, the continuum also contributes noise and can partially interfere destructively elsewhere, so the global SNR may or may not improve. Nonetheless, our technique offers the ability to dynamically tune the phase of the Brillouin response relative to the background via adjustment of the probe laser wavelength. This interference-tuning of the continuum and discrete components allows some control over constructive or destructive interference. Moreover, we can achieve this without sacrificing independent control of the pump–probe detuning: by simultaneously shifting the pump laser in step with the probe, we can preserve the desired phase-matching bandwidth while optimizing the relative phase for Fano interference.

In the phase-matching bandwidth experiment on 1 cm of UHNA3 fiber (Figure 3.6), several effects (noise floor, alignment drifts, fiber dispersion, etc.) slightly distort the ideal  $\text{sinc}^2$  response. Near the troughs of the  $\text{sinc}^2$  function and for larger pump–probe detunings, the measured Brillouin peaks are weaker and exhibit small spectral asymmetries (see Appendix B.3). This is consistent with a Fano-type distortion in which the Brillouin amplitude and background continuum are comparable, allowing interference to skew the lineshape and shift the peak away from the naïve Lorentzian center. Consequently, a simple Lorentzian fit underestimates the true peak amplitude in these regimes. By fitting a standard Fano profile, we more accurately capture the asymmetric peak and its shifted center frequency.

### 3.6 Conclusion

In conclusion, we have introduced a coherently stimulated Brillouin spectrometer utilizing a detuned pump-probe design to achieve high sensitivity and room-temperature operation in  $\mu\text{m}$ -scale samples. This approach successfully overcomes the spatial resolution limitations imposed by conventional SBS methods, demonstrating sub-10 fW sensitivity in UHNA3 fiber and enabling Brillouin measurements in bulk liquid carbon disulfide with unprecedented efficiency. By relaxing phase-matching constraints, this instrument opens new possibilities for characterizing nanoscale material properties and developing nano-acousto-optic devices in standard laboratory settings without the need for cryogenic environments. Moving forward, our methodology could facilitate advancements in high-resolution phonon spectroscopy and inspire further innovations in the study of material mechanics at the microscale, reinforcing the broader applicability of Brillouin-based techniques across materials science, photonics, and sensing technologies.



## Chapter 4

### Brillouin-Induced Raman Modes and Device Exploration

#### 4.1 Introduction

This chapter explores progress towards the goal of demonstrating Brillouin-induced Raman-like modes at room temperature. We aim to show that acoustic traveling-wave phonons generated by a Brillouin scattering process in a confined medium can organize into discrete standing-wave vibrational modes. This goal represents a convergence of several themes in modern photonics: cavity optomechanics, coherent phonon control, and nonlinear light–matter interactions. Cavity optomechanics has traditionally focused on discrete mechanical resonances such as drumhead membranes or whispering-gallery-mode (WGM) microresonators to achieve effects like laser cooling of vibrational modes or phonon lasing.<sup>29, 30, 82, 83</sup> In parallel, Brillouin scattering provides a route to manipulate traveling-wave acoustic phonons in extended media, as demonstrated by our Brillouin-based laser cooling in optical fiber (a “cavity-less” system).<sup>23, 31, 32, 50</sup> The research in this chapter aims to bridge these domains, leveraging traveling-wave Brillouin scattering in a finite-length system so that the phonons become self-confined, resembling the vibrational modes that give rise to Raman scattering.

This investigation is grounded in the broader context of controlling phonons and light at the mesoscale. High-coherence phonons are garnering interest for precision metrology and quantum information,<sup>84, 85</sup> spurring new strategies for phonon coherent optomechanical interactions.<sup>30, 82</sup> Earlier chapters of this dissertation advanced this frontier: Chapter 2 demonstrated the laser cooling of propagating acoustic phonons in an optical fiber, extending optomechanical control to continuous media at room temperature,<sup>23</sup> and Chapter 3 introduced a novel Brillouin spectrometer (CoBS) which offers especially high sensitivity for short ( $<1\text{ cm}$ ) lengths over traditional Stimulated Brillouin Scattering (SBS). Building on such results, the present work tackles the next challenge: inducing standing-wave acoustic modes in a bulk-like sample through Brillouin processes. Achieving this at room temperature would be significant, as to date strong acoustic mode formation has been mostly limited to cryogenic systems where phonon lifetimes are long.<sup>32, 86</sup> By pursuing Brillouin-induced modes under ambient conditions, we push toward practical phonon devices and new regimes of light–sound

interaction without the strict need for optical cavities.<sup>87</sup>

## 4.2 From Traveling-Wave to Raman-Like Standing-Wave Modes

### 4.2.1 Review of Brillouin and Raman Scattering

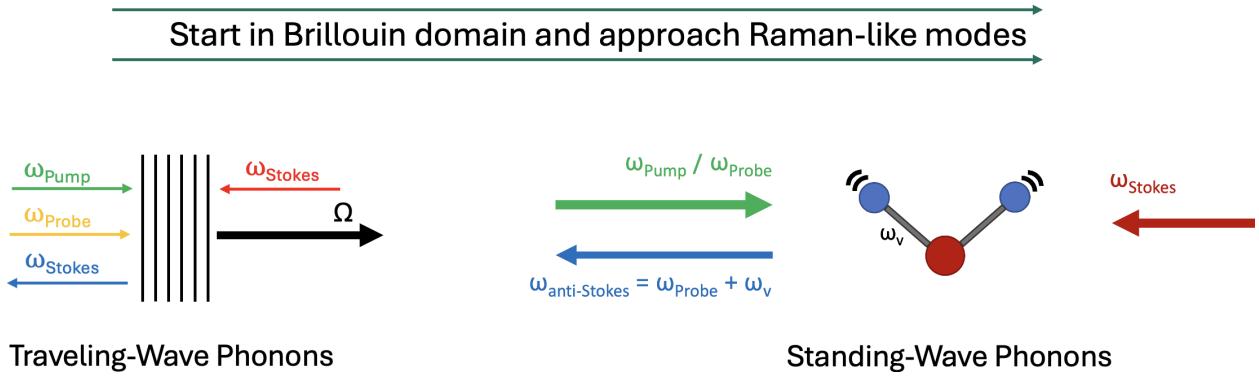
Brillouin scattering and Raman scattering are two related light-matter interactions that involve inelastic scattering of photons by phonons, but they differ in the nature of the phonons involved. In Brillouin scattering, an incident photon exchanges energy and momentum with a long-wavelength acoustic phonon (a propagating sound wave in the medium). In contrast, Raman scattering typically involves optical phonons or molecular vibrations, which are localized oscillations (e.g., bond vibrations within a molecule or internal lattice vibrations) rather than a continuum acoustic wave. In other words, Brillouin scattering is mediated by traveling acoustic waves in a bulk material, whereas Raman scattering probes standing-wave intramolecular or lattice modes. This fundamental difference is reflected in the frequency scales: acoustic phonons have relatively low frequencies (GHz or below) and are responsible for the small Stokes/anti-Stokes shifts in Brillouin spectra, whereas optical phonons and molecular vibrations have much higher frequencies (THz) yielding the larger shifts seen in Raman spectra (see Figure 1.1).<sup>88</sup> It is also reflected in momentum conservation conditions: Brillouin scattering requires phase-matching between the optical wave and an acoustic phonon of a particular wavevector, essentially picking out a traveling phonon mode with a definite momentum. Raman scattering, on the other hand, often involves phonons with near-zero wavevector (e.g., zone-center optical modes in crystals or whole-molecule vibrations) due to the selection rule of crystal momentum conservation in perfect lattices.<sup>89</sup>

Despite these distinctions, the line between Brillouin and Raman processes can blur in certain situations. If the medium lacks long-range order or the phonon coherence length is short, the usual momentum-selection rules break down. Shuker and Gammon (1970)<sup>90</sup> showed that in amorphous materials, translational symmetry is lost and essentially all vibrational modes can participate in light scattering. In such cases, even low-frequency acoustic-like modes (normally the realm of Brillouin) become “Raman-active.” This insight explained the observation of broad low-frequency Raman scattering (the so-called boson peak) in glasses by attributing it to acoustic vibrations made allowable by disorder.<sup>91-98</sup> Conversely, in a small or confined system, the vibrational modes are discrete rather than forming a continuous acoustic band. In effect, they approach the molecular limit where each normal mode can scatter light akin to a Raman transition. Thus, one can view Brillouin and Raman scattering as two limits of the same fundamental interaction, distinguished by whether the phonons act as continuous waves or as localized normal modes.

Spatial confinement of acoustic waves can convert the Brillouin regime into a Raman-like regime. In

an unbounded or large medium, acoustic phonons exist over a continuum of frequencies and wavevectors (a traveling-wave picture). But if the medium is bounded (e.g., a micron-scale particle or a cavity of finite length) the acoustic field must satisfy boundary conditions, leading to a set of allowed eigenmodes. Classic elasticity theory by Lamb (1882)<sup>99</sup> provided the first analysis of this: a homogeneous elastic sphere supports only certain quantized vibration modes (classified into spheroidal and torsional families) dictated by the sphere's finite size and free-surface boundary condition. These quantized vibration modes are true standing-wave modes confined to the sphere. Subsequent work extended Lamb's theory to include various effects such as surface tension and clamping for small particles, but the essential result is that a finite object has discrete acoustic eigenmodes. Experimentally, Duval et al. (1986)<sup>100</sup> performed a notable demonstration by observing very-low-frequency Raman scattering from nanometer-sized microcrystals embedded in glass. The Raman spectra showed distinct peaks whose frequencies scaled inversely with the particle diameter, exactly as expected for Lamb's vibrational modes in a sphere. Duval identified these peaks as the confined acoustic eigenmodes ("particle vibrational modes") of the microcrystallites, which had become Raman-active. This exemplified how a Brillouin-like acoustic wave (here, a sphere's breathing or shear wave) can become Raman-like when spatially confined. When an acoustic phonon is restricted by boundaries, it is no longer a freely propagating wave but rather a normal mode with a discrete frequency, effectively turning a Brillouin interaction into a Raman-style interaction with a set of allowed modes.

Figure 4.1 offers a conceptual illustration of the transition from a traveling acoustic wave to standing-wave vibrational modes. In a bulk material (left), light scatters from a continuum of acoustic waves (Brillouin scattering), whereas in a confined medium (right), only discrete phonon modes are allowed, producing Raman-like spectral lines. Spatial confinement and acoustic reflections thus shift the scattering from the Brillouin regime toward a Raman-like regime. Contemporary research in cavity optomechanics and related fields has leveraged this wave-to-mode transition as well. For instance, Renninger et al. (2018)<sup>101</sup> showed that by shaping the geometry of a cm-scale crystal, one can support long-lived acoustic standing-waves even in a bulk solid. At low temperature, the phonon coherence length in their system exceeded the system size, and the crystal effectively behaved as a giant "phonon cavity" with high-Q (quality) acoustic modes ( $Q = \Omega_B/\Gamma_B$ ).<sup>101, 102</sup> These modes were accessed via Brillouin interactions, blurring the line between traditional Brillouin and Raman: the process was Brillouin in origin (photoelastic coupling to sound waves) but the phonons were in discrete cavity modes like Raman vibrations. These studies underscore that confined acoustic phonons can take on the character of Raman modes.



**Figure 4.1:** Conceptual illustration of the transition from a traveling acoustic wave to standing-wave vibrational modes. In a bulk material or extended media (left), light scatters from a continuum of traveling acoustic waves (Brillouin scattering), whereas in the molecular vibrations of atomic bonds (right), only discrete standing-wave phonon modes are allowed (Raman-like scattering). The left diagram is a conceptual visualization of the CoBS process for a longitudinally-traveling phonon  $\Omega$  (see Chapter 3) while the right diagram gives a conceptual visualization of an analogous Coherently stimulated Anti-Stokes Raman Scattering (CARS) process with the standing-vibrations  $\omega_v$  among bonded atoms in a molecule.

#### 4.2.2 Brillouin-Induced Raman Modes

In a traditional SBS experiment, a pump laser drives an acoustic wave through electrostriction, and the scattered Stokes light is down-shifted by the acoustic frequency  $f_B$ , given by<sup>13</sup>

$$f_B = \frac{2v_s n}{\lambda}, \quad (4.1)$$

where  $v_s$  is the longitudinal sound speed,  $n$  is the refractive index, and  $\lambda$  is the optical frequency. In an unbounded or extended medium, the frequency response of the scattered light is determined by material properties (sound speed and optical dispersion) and the acoustic wave can be thought of as a traveling grating moving through the medium. If the medium is shortened such that the acoustic wave can reflect off the sample boundaries, the traveling acoustic waves can auto-interfere to form a standing-wave pattern in the medium. Under these system conditions, a phonon induced by SBS will propagate to the sample end, reflect (assuming a high acoustic impedance mismatch at the boundary), and retraverse the medium in the reverse direction. If the length  $L$  between acoustic interfaces is a half-integer number of acoustic wavelengths, the forward- and backward-traveling phonon waves can interfere to form a standing wave pattern (i.e., a resonance), in essence trapping the traveling phonon in the cavity defined by the sample. This standing wave in turn acts like a stable grating, enhancing the scattering of light at well-defined frequencies corresponding to its resonances. We term these resonant phonon excitations “Brillouin-induced Raman modes” to reflect

their hybrid character.

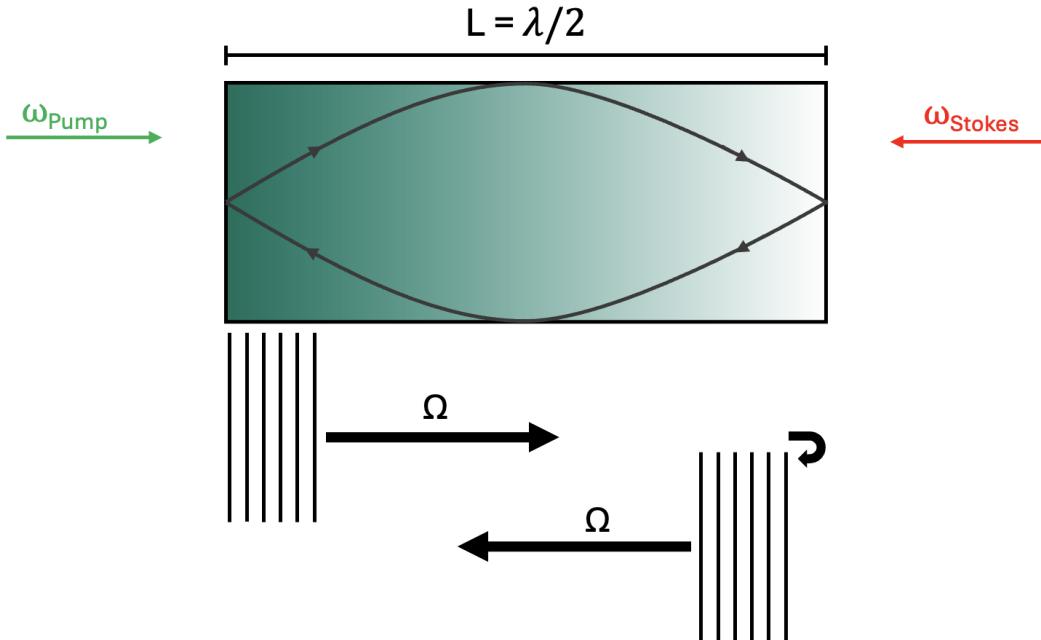
In more concrete terms, the finite-length system behaves like an acoustic Fabry-Pérot cavity. For a longitudinal acoustic mode, the condition for a standing wave is that an integer number  $n$  of half-wavelengths equals the round-trip length:  $\frac{n\lambda_s}{2} = L$ . Equivalently, the allowed acoustic frequencies are

$$f_R = \frac{n v_s}{2L}, \quad (4.2)$$

where  $v_s$  is the sound velocity in the medium and  $n = 1, 2, 3, \dots$  indexes the mode order. The lowest-frequency mode ( $n = 1$ ) has a fundamental frequency  $f_1 \approx \frac{v_s}{2L}$ , and higher modes are integer multiples of this fundamental (assuming a simple one-dimensional confinement). Thus, instead of a single Brillouin shift  $f_B$ , one expects a ladder of equally spaced phonon modes in the scattering spectrum, resembling a Raman vibrational progression. The spacing  $\Delta f$  between adjacent modes is approximately  $\Delta f \approx \frac{v_s}{2L}$ , set solely by the cavity length and sound speed. Figure 4.2 illustrates why this is so. For example, if  $L = 10$  mm in a medium where  $v_s = 5000$  m s<sup>-1</sup>, the fundamental mode would be  $f_1 \approx 250$  kHz and overtones at 500 kHz, 750 kHz, etc. In principle, a sufficiently short and high-Q acoustic cavity could produce a comb of multiple GHz-range lines (analogous to molecular vibrational Raman lines) out of what would ordinarily be a single broad Brillouin gain peak.

These Brillouin-induced modes would manifest as distinct peaks in the spectrum of the scattered light. In addition to the usual broadened Brillouin spectrum (with width given by acoustic damping), one would see a series of sharp lines at  $f_1, f_2, f_3, \dots$  around the center scattered frequency. Such a spectrum would be clear evidence that the phonon field is not only a freely propagating wave but also oscillating in discrete standing patterns (i.e., an optically driven acoustic resonator within the material). This is conceptually similar to stimulated Raman scattering in a molecule, where one can get a cascade of Stokes lines corresponding to  $1\hbar\omega_{\text{vib}}, 2\hbar\omega_{\text{vib}}, 3\hbar\omega_{\text{vib}}$  energy shifts if the pump is intense. Here the “molecular vibration” is replaced by an acoustic cavity mode. If the pump power is high enough to drive the acoustic mode into the nonlinear regime, one might even observe multiple orders of Stokes (and anti-Stokes) as the phonon population builds up in those modes.

The idea of SBS-driven acoustic modes has parallels in prior work. In the cryogenic experiment of Renninger et al. mentioned earlier,<sup>101</sup> a bulk acoustic mode was driven via Brillouin scattering in a cm-scale quartz crystal. At low temperatures, they observed ultra-narrow acoustic resonances (indicative of discrete modes) in place of a broad Brillouin response, confirming that acoustic coherence across the entire sample can indeed produce a modal spectrum. In our case, we seek to do this at room temperature by engineering a



**Figure 4.2:** Illustration showing how geometry and sound speed of the material determine the allowed acoustic frequencies, given by Equation 4.2. Traveling-wave phonons around the material's Brillouin frequency shift  $f_B$  are transduced within the medium via the Coherently stimulated Brillouin Spectrometer (CoBS) process. These phonons traverse the length to reach the boundary, where a high acoustic impedance mismatch causes them to reflect at the interface and retraverse the length of the medium in the reverse direction. The spatio-temporal overlap of counterpropagating traveling-wave phonons causes them to interfere, creating an acoustic standing-wave pattern within the material at discrete frequencies determined by the length and sound speed of the material. Shown in the illustration is the half-wavelength fundamental ( $n = 1$ ) mode, however in general, harmonics near the material's mechanical resonance frequency  $f_B$  would be excited in the medium.

shorter effective acoustic cavity. We aim to accomplish this by taking advantage of our Coherently stimulated Brillouin Spectrometer (CoBS), which offers  $\sim 10^6$  improvement in scattered power for  $\sim \text{cm}$  lengths over traditional SBS (see Appendix B.2 for a comparison of scattered power across Brillouin techniques, and specifically Figure B.1 for a comparison by effective length  $L$ ).

### 4.2.3 Key Parameters and Feasibility

Observing Raman-like standing-wave modes via SBS hinges on several key parameters of the system. We identify three especially critical factors: (1) the scattered power, determined by the effective material Brillouin gain and length as well as the optical powers; (2) the acoustic dissipation in the medium, which limits the mean phonon travel distance (coherence length); and (3) the acoustic boundary reflectivity, determined by impedance mismatch at interfaces, which enables the phonons to reflect and form standing-waves. These

parameters together determine whether the phonon will remain a distributed traveling excitation or collapse (blur) into discrete modes. Chapter 3 describes the CoBS instrument, showing that the scattered power as a result of the CoBS process is given by Equation 3.1, stated here again as

$$P_{\text{Signal}} = \frac{1}{4}(G_B L)^2 P_{\text{Pump}} P_{\text{Stokes}} P_{\text{Probe}} \Phi, \quad (4.3)$$

where  $G_B$  is the material-dependent effective (acousto-optic overlap-adjusted) Brillouin gain factor given by Equation 3.2,  $L$  is the effective length,  $P_i$  are the optical powers of the pump, Stokes, and probe waves, respectively, and  $0 < \Phi < 1$  is a phase-matching relaxation term (given by Equation 3.5) that captures the pump-probe detuning on which the instrument relies (in general,  $\Phi$  increases for smaller  $L$ ). A material's Brillouin gain coefficient  $g_0$  ( $\text{W m}^{-1}$ ), or overlap-adjusted effective gain  $G_B$  ( $\text{W}^{-1} \text{m}^{-1}$ ), sets how strongly the phonons are driven for given  $P_{\text{Pump}}$ ,  $P_{\text{Stokes}}$ , and  $P_{\text{Probe}}$  optical powers over an interaction length  $L$  in the CoBS process (see Chapter 3, and specifically Equation 3.1), given here again by

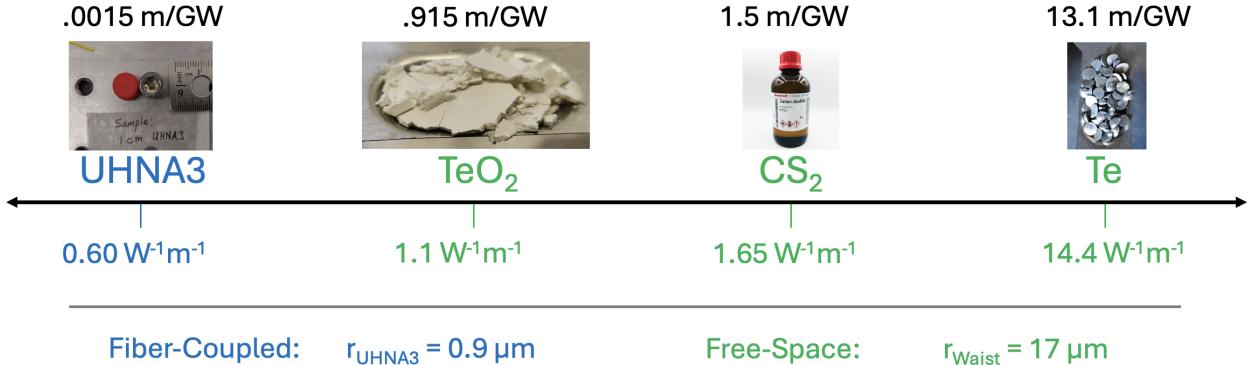
$$G_B = \frac{g_0}{A_{\text{eff}}^{\text{ao}}} \frac{\left(\frac{\Gamma_B}{2}\right)^2}{(\Omega - \Omega_B)^2 + \left(\frac{\Gamma_B}{2}\right)^2}. \quad (4.4)$$

Here,  $\Gamma_B$  is the angular Brillouin linewidth,  $\Omega$  ( $\Omega_B$ ) is the (resonant) angular acoustic frequency,  $A_{\text{eff}}^{\text{ao}}$  is the effective area (acousto-optic mode overlap), and  $g_0$  is the Brillouin gain coefficient given by

$$g_0 = \frac{\gamma_e^2 \omega^2}{n v_s c^3 \rho_0 \Gamma_B}, \quad (4.5)$$

where  $\gamma_e$  is the electrostrictive constant,  $\omega$  is the angular optical frequency,  $n$  is the refractive index,  $v_s$  is the speed of sound in the material,  $c$  is the speed of light, and  $\rho_0$  is the mean density of the material. Short samples demand very high effective Brillouin gain to achieve significant scattered power in a small  $L$ . Certain tellurium-based materials, for instance, can offer gain factors orders of magnitude higher than silica,<sup>35, 103</sup> allowing measurable scattered power in sub-mm cavities. Figure 4.3 shows a graphic which summarizes key gain parameters for the materials investigated in this study, including germanium-doped fiber (UHNA3), tellurium dioxide ( $\text{TeO}_2$ ), carbon disulfide ( $\text{CS}_2$ ), and tellurium (Te).

Even if phonons are driven strongly, they must live long enough (i.e., have a low enough dissipation rate, or high enough acoustic  $Q = \frac{\Omega_B}{\Gamma_B}$ ) to form a standing wave. At room temperature, intrinsic damping can limit phonon  $Q$ s to  $\sim 10^3$ – $10^4$  in many solids,<sup>114, 115</sup> implying attenuation lengths of mm to cm for GHz



**Figure 4.3:** Brillouin gain ( $g_0$ ,  $\text{m GW}^{-1}$ ) and acousto-optic overlap-adjusted Brillouin gain ( $G_B = \frac{g_0}{A_{\text{eff}}^{\text{ao}}}$ ,  $\text{W}^{-1}\text{m}^{-1}$ ) of the materials investigated in this study. While the material gain (top row text) of Ultra High Numerical Aperture 3 (UHNA3) fiber is  $10^3$  lower than the other materials, the effective area is confined tightly within its  $0.9\text{ }\mu\text{m}$  radius core, greatly improving the acousto-optic response for given length and optical power. The lenses of our free-space optical setup offer comparatively less tight confinement of the light, with the beam waist having radius  $r_{\text{Waist}} = 17\text{ }\mu\text{m}$  (bottom row text). The effective gain (middle row text) is ultimately the important gain parameter, as it accounts for both the material gain and the area over which light and sound interact (see Equations 4.4 and 4.5).<sup>13, 23, 73, 101, 104–113</sup>

frequencies. Ideally, the sample length  $L$  should be comparable to or less than half the attenuation length so that phonons undergo multiple round trips. This is considerably more difficult at room temperature than at cryogenic temperatures, where  $Q_s$  can exceed  $10^7$ .<sup>101, 116</sup>

Finally, the phonon must reflect rather than escape at the boundaries. Acoustic reflection is determined by the acoustic impedance mismatch between two materials at an interface, with acoustic impedance given by

$$Z_{\text{ac}, i} = \rho_i v_{s,i}, \quad (4.6)$$

where  $\rho_i$  is the mean density and  $v_{s,i}$  is the longitudinal speed of sound in a given material  $i$ . Acoustic reflection is then given by

$$R = \frac{Z_1 - Z_2}{Z_1 + Z_2}. \quad (4.7)$$

A large acoustic impedance mismatch (e.g., a free surface with air on one side) can approach nearly 100% reflection.<sup>86, 117</sup> For example, the acoustic reflection between  $\text{TeO}_2$  ( $\rho \approx 5670\text{ kg m}^{-3}$ ,  $v_s \approx 4260\text{ m s}^{-1}$ ) and air ( $\rho \approx 1.02\text{ kg m}^{-3}$ ,  $v_s \approx 343\text{ m s}^{-1}$ ) is  $R = 0.999$ , or 99.9%. Smaller acoustic impedance mismatches result

is lower acoustic reflection, for example  $R = 0.005$ , or 0.5% between Te ( $\rho \approx 6240 \text{ kg m}^{-3}$ ,  $v_s \approx 2610 \text{ m s}^{-1}$ ) and selenium (se) ( $\rho \approx 4810 \text{ kg m}^{-3}$ ,  $v_s \approx 3350 \text{ m s}^{-1}$ ). Designing the sample with two opposing highly acoustically reflective boundaries is essential to generating a Raman-like standing-wave mode in the medium. In practice, however, partial reflections at each (or even just one) end may suffice so long as the net round-trip reflectivity is high enough to sustain a mode.

In short, we want to create a high- $Q$  acoustic resonator inside a Brillouin-active medium at room temperature: strong enough acousto-optic driving to excite the phonons, low enough damping to maintain them, and robust boundary reflections to confine them. Meeting all of these conditions can be challenging. However, the high ( $\sim 5 \text{ fW}$ ) sensitivity and unique short path length advantage of our CoBS instrument (described in Chapter 3 and Section B.2) sparks new motivation, as it provides a novel technique tailored for observing small-scale scattering phenomenon. Feasibility estimates and initial CoBS measurements indicate that by utilizing ultra-high-gain media such as Te<sup>35, 103</sup> or liquid CS<sub>2</sub><sup>13</sup>, and ensuring at least one boundary is acoustically reflective, one can push toward Brillouin-induced Raman modes even under ambient conditions.

The sensitivity of our instrument, representing the minimum scattered power that may be detected, has been measured at  $P_{\text{Signal}} \approx 5 \text{ fW}$  (see Section 3.5.1, and specifically Table 3.1 together with Equation 3.1 and Figure 3.3 for validation of this claim). Using this sensitivity value for  $P_{\text{Signal}}$  we can rearrange Equation 4.3 to solve for the minimum length  $L$  we can expect to observe scattering within for given optical powers, material gain, and pump-probe detuning:

$$L = \frac{2}{G_B} \sqrt{\frac{P_{\text{Signal}}}{P_{\text{Pump}} P_{\text{Stokes}} P_{\text{Probe}} \Phi}}, \quad (4.8)$$

where  $0 < \Phi < 1$  and generally increases for smaller  $L$ . For  $P_{\text{Signal}} \approx 5 \text{ fW}$  sensitivity and maximum optical powers  $P_{\text{Pump}} P_{\text{Stokes}} P_{\text{Probe}} \approx 0.25 \text{ W}^3$  under typical conditions, Equation 4.8 predicts, for example, the ability to observe scattering within  $\sim 500 \text{ nm}$  of UHNA3 fiber ( $G_{B, \text{UHNA3}} = 0.6 \text{ W}^{-1} \text{ m}^{-1}$ ). Equation 4.8 makes clear that minimizing the observable scattering length is accomplished by any of: bumping applied optical powers, improving instrument sensitivity, or choosing a higher gain scattering medium.

In what follows, we detail the experimental platforms and theoretical modeling that guided our attempts to observe discrete phonon modes in high-gain materials. Although a conclusive demonstration proved elusive, the conceptual framework is robust and provides a foundation for ongoing efforts. By shrinking the acoustic path length, maximizing phonon reflectivity, and exploiting strong CoBS gain, one can approach the regime where traveling-wave Brillouin scattering morphs into Raman-like standing-wave modes. This pursuit effectively unifies the traveling-wave and standing-wave paradigms of light-sound interaction, pro-

viding new opportunities in cavity-free phononics, resonant optomechanics, and coherent phonon devices at room temperature.

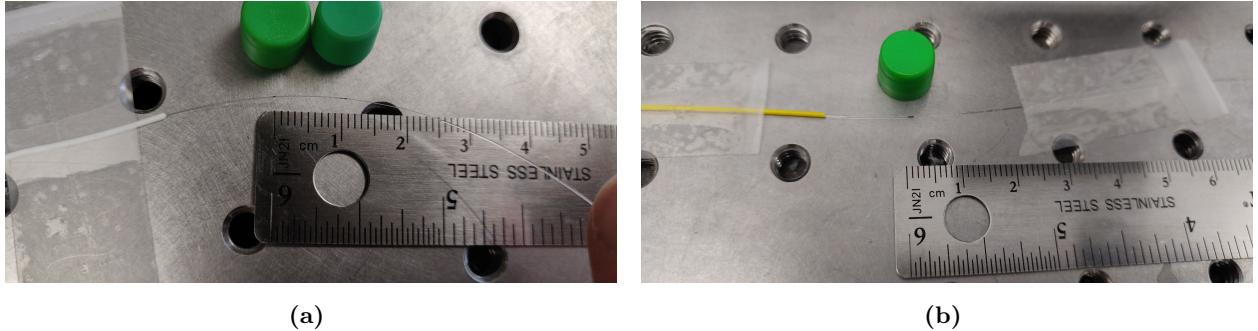
## 4.3 Results

In this section we detail results and progress toward measuring Brillouin-induced Raman modes, in chronological order of investigation. The overall approach was to start deep in the continuous Brillouin domain of longitudinally-traveling phonons in bulk material and steadily approach the discrete Raman domain by measuring incrementally shorter lengths of materials well-suited to the task. This investigative study began with user-friendly fiber-coupled lengths of UHNA3 fiber, progressed quickly to increasingly challenging free-space measurements of materials such as liquid CS<sub>2</sub> and thin films of TeO<sub>2</sub> and Te, and ultimately arrived at fiber-to-chip coupling into specialized suspended waveguides. We present here both the successes along the way as well as key insights gained from setbacks, with special focus on illuminating the reasoning behind critical pivots in our investigation.

### 4.3.1 Ge-Doped Optical Fiber

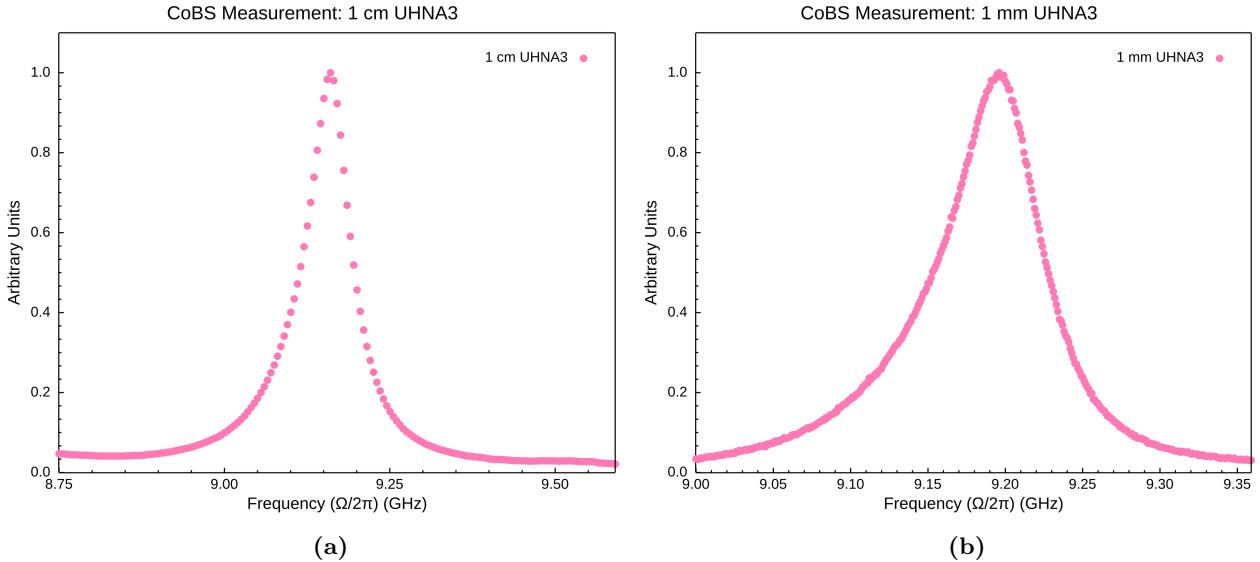
To begin with a clear measurement firmly in the Brillouin domain, we target (short) lengths of the well-understood UHNA3 fiber.<sup>73</sup> UHNA3 features a Brillouin frequency shift distinct from the single mode fiber 28 (SMF-28) of the apparatus ( $f_{B,\text{UHNA3}} = 9.18\text{ GHz}$  vs  $f_{B,\text{SMF28}} = 10.85\text{ GHz}$ ), making it easily distinguishable from instrument background. Additionally, its small core and high Ge concentration (~44 wt.%) make for tight acousto-optic overlap ( $A_{\text{eff}}^{\text{ao}}$ ) and good guidance of both light and sound, respectively. Figure 4.3 shows the key gain figures for UHNA3. While its material gain ( $g_0 = 0.0015\text{ m GW}^{-1}$ ) is low, the acousto-optically adjusted effective Brillouin gain ( $G_B = \frac{g_0}{A_{\text{eff}}^{\text{ao}}} = 0.60\text{ W}^{-1}\text{ m}^{-1}$ ) is sufficiently high to allow a measurement in sub-mm lengths for typical optical powers (Equation 4.8).

Figure 4.4 shows pictures of measured 1 cm and 1 mm lengths of UHNA3 fiber (Figures 4.4a and 4.4b, respectively) spliced into the SMF-28 of the sample stage of the CoBS apparatus. Figure 4.5a shows the observed spectra for this 1 cm segments of UHNA3 captured at maximum available optical powers  $P_P P_S P_{Pr} \approx 0.25\text{ W}^3$ . The peak spectral amplitude has been normalized to unity and spectral density is expressed in Arbitrary Units for clean presentation, a standard practice which acknowledges the arbitrary nature of the optical-to-electrical conversion unique to each photodiode detector.  $1\sigma$  uncertainties are smaller than the data point markers. Figure 4.5a showcases a replication measurement taken more recently, after orders of magnitude sensitivity and performance improvements to the CoBS instrument, however the measurement gathered initially also represented a strong successful measurement of 1 cm UHNA3, prompting an advance



**Figure 4.4:** Photographs of (a) 1 cm and (b) 1 mm lengths of UHNA3 fiber, indicated by black markings and spliced into the SMF-28 of the CoBS instrument. Samples were fabricated by first splicing a longer segment of UHNA3 to SMF-28 and marking the splice location such that the UHNA3 could subsequently be cleaved to the appropriate length as measured from the marking of the first splice.

to attempting a measurement of 1 mm UHNA3.



**Figure 4.5:** Observed background-subtracted spectrum obtained through a CoBS measurement of (a) 1 cm of UHNA3 fiber (pictured in Figure 4.4a) and (b) 1 mm of UHNA3 fiber (pictured in Figure 4.4b), captured at maximum operating optical powers  $P_{PP}P_S P_{Pr} \sim 0.25 \text{ W}^3$ , with  $1\sigma$  uncertainties smaller than the data point markers. The spectrum shows strong Fano asymmetry (see Section B.3 in Appendix B), indicating interference of a weak signal with the broad background continuum. This is an older measurement, taken prior to orders of magnitude sensitivity improvements, and the Fano effects seen here indicate this was near the sensitivity limit of the instrument at the time.

This 1 mm UHNA3 measurement is shown in Figure 4.5b, which features slight Fano asymmetry in line-shape indicative of signal-background interference in a low-signal regime (see Section B.3 in Appendix B). Unlike Figure 4.5a, which was recaptured more recently, after orders-of-magnitude sensitivity and performance improvements to the CoBS instrument, the spectra shown in Figure 4.5b dates to the time of its

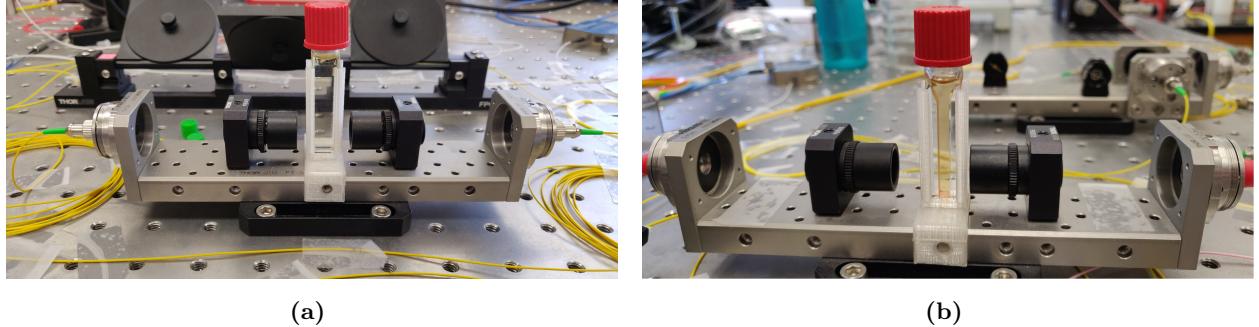
relevance to this investigation. The slight Fano asymmetry indicates that this measurement, while very clear and of high SNR, was captured from a signal which was near the order of the background continuum. This places the measurement not far from the sensitivity limit of the instrument at the time. Nonetheless, Figure 4.5b represents a clear successful measurement of 1 mm UHNA3 fiber, prompting a continuation on the path to shorter lengths and Brillouin-induced Raman modes.

### 4.3.2 Free-Space Optics with Liquid CS<sub>2</sub>

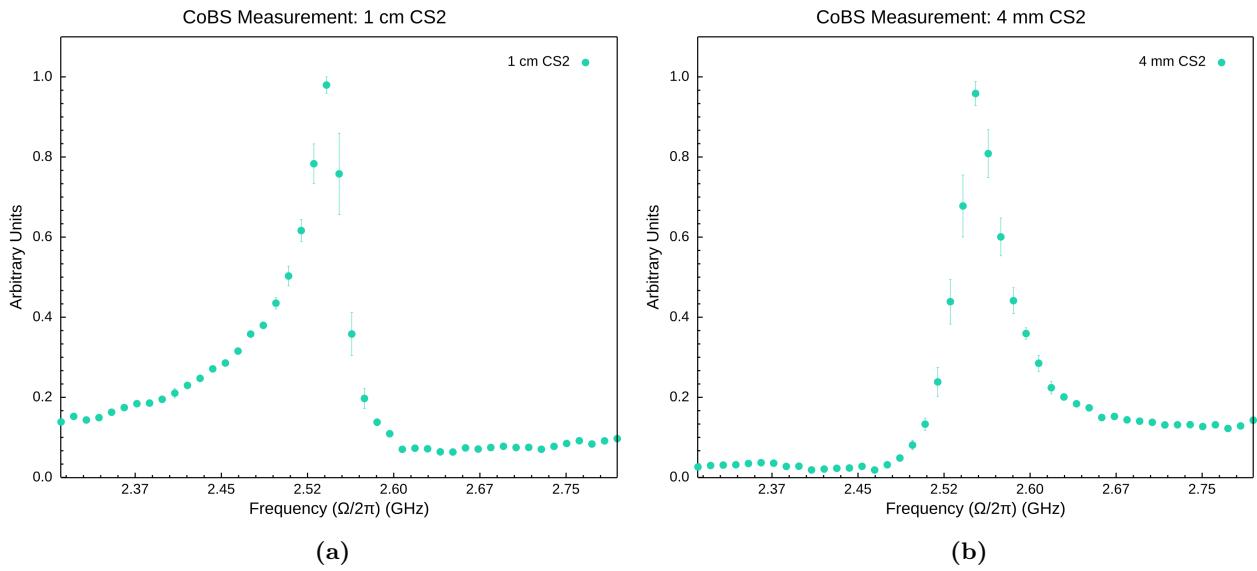
Having exhausted the shortest practical lengths of optical fiber reasonably fabricatable, a regime change to free-space optics in the CoBS sample stage was required to progress our investigation further. To smooth this transition, we first targeted meaningful lengths of the high-gain liquid CS<sub>2</sub>. From Figure 4.3, CS<sub>2</sub> has a material Brillouin gain of  $\sim 1.5 \text{ m GW}^{-1}$ , which is  $10^3$  higher than that of UHNA3 fiber, however this material advantage is offset by the much looser acousto-optic mode overlap afforded by the beam waist produced by our lenses ( $A_{\text{eff, Free-Space}}^{\text{ao}} = \pi(17 \mu\text{m})^2$  vs  $A_{\text{eff, UHNA3}}^{\text{ao}} = \pi(0.9 \mu\text{m})^2$ ). The resulting effective Brillouin gain of liquid CS<sub>2</sub> in the free-space optical setup of our CoBS instrument is  $G_B \approx 1.65 \text{ W}^{-1} \text{ m}^{-1}$ , nearly a factor of 3 greater than that of UHNA3. This combined with an initially targeted length  $L = 1 \text{ cm}$  provided a sufficiently large target for a gentle transition from the fiber-coupled to free-space regime.

Figure 4.6 shows photographs of a 1 cm-by-4 mm path length cuvet of liquid CS<sub>2</sub> mounted in the sample stage between two lenses. Light is guided from the fiber of the apparatus into free space, collimated and focused by the fiber ports and diffraction-limited lenses on either side of the fiber bench upon which the sample sits. Figure 4.6a shows the cuvet placed in the 1 cm path length configuration while Figure 4.6b shows it rotated 90° for the 4 mm path length configuration.

Figure 4.7 shows the observed spectra from CoBS measurements of 1 cm and 4 mm liquid CS<sub>2</sub> photographed in Figure 4.6. Both measurements showcase significant Fano asymmetry, again indicative of signal-background interference in a low-signal regime (see Section B.3 in Appendix B). The strong Fano interference in both measurements emphasizes the proximity to the sensitivity limit of the CoBS instrument at the time these spectra were observed. Each progression to lower-signal (less scattered power  $P_{\text{Signal}}$ , from Equation 4.3) represents iterative sensitivity and performance improvements to the CoBS instrument, ranging from insights about the importance of well-cleaned and polished pigtail ends throughout the apparatus to operating setting discoveries such as setting both the pump and probe lasers to “whisper” instead of “dither” mode, as well as component upgrades such as a higher performance detector and higher power Erbium-doped fiber amplifiers (EDFAs) (see Chapter 3 for more details on the CoBS instrument and Section 3.4.2 for specifics on performance optimization).



**Figure 4.6:** Photographs of (a) 1 cm and (b) 4 mm lengths of liquid  $\text{CS}_2$  in the beam path of the CoBS instrument. The single 1 cm-by-4 mm path length cuvet allowed for both measurements shown in Figure 4.7 via  $90^\circ$  rotation within its mount.



**Figure 4.7:** Observed background-subtracted spectrum obtained through a CoBS measurement of (a) 1 cm of liquid  $\text{CS}_2$  (pictured in Figure 4.6a) and (b) 4 mm of liquid  $\text{CS}_2$  (pictured in Figure 4.6b), with  $1\sigma$  uncertainties smaller than the data point markers. The spectrum shows strong Fano asymmetry (see Section B.3 in Appendix B), indicating interference of a weak signal with the broad background continuum. This is an older measurement, taken prior to orders of magnitude sensitivity improvements, and the Fano effects seen here indicate this was near the sensitivity limit of the instrument at the time.

With proof-of-principle free-space functionality demonstrated with the successful measurement of 1 cm and 4 mm bulk liquid  $\text{CS}_2$  in a cuvet, all basic experimental infrastructure was in place to pursue Brillouin-induced Raman modes in much shorter length samples. The newly-achieved  $\sim 10 \text{ fW}$  sensitivity limit of the instrument around this time, experimentally-obtained from reduced-power measurements of 1 cm UHNA3 fiber, supported the feasibility of measuring scattering in far shorter interaction lengths. With this in mind, we made the ambitious leap to thin films.

### 4.3.3 TeO<sub>2</sub> Thin Film

Table 4.1 presents key values for material properties of TeO<sub>2</sub> relevant to observing Brillouin-induced Raman modes for both the crystalline and amorphous thin film forms. To observe Raman-like modes in a confined medium, we must be able to sufficiently excite traveling-wave phonons via the CoBS process in an effective length equal to at most half the mean travel distance of the phonons in that material (coherence length  $L_{coh}$ ). Phonon coherence length is a function of the material's mechanical dissipation rate, or Brillouin linewidth  $L_{coh} = \frac{2\pi v_s}{\Gamma_B} = \tau_p v_s$ , where  $\tau_p$  is the phonon lifetime  $\tau_p = 2\pi\Gamma_B^{-1}$ . Setting the effective length  $L$  at half of the phonon coherence length ensures the phonons make at least one round-trip in the material before dissipating (decohering), representing a critical requirement in observing Brillouin-induced Raman modes. In the table, a CoBS scattered power estimate is given (via Equation 4.3), corresponding to half the phonon coherence length of the material. This figure (consistent for  $L_{coh}/2$  and in pW reported units across all similar material property tables presented in this chapter) offers a reference for the minimum sensitivity performance required from the CoBS instrument if Raman modes are to be observed in the material. For example, if Raman modes are to be observed in crystalline TeO<sub>2</sub>, the effective length of the sample used to measure them must be at most  $L_{eff, sample} = \frac{L_{coh}}{2} = 215 \mu m$  and CoBS must be capable of 3500 pW sensitivity (which it achieves by  $\sim 10^3$ ). In the table,  $\tau$  is the phonon lifetime ( $\tau = (2\pi \cdot \Gamma_B)^{-1}$ ),  $v_{s, long}$  is the longitudinal sound speed,  $n$  is the refractive index, and  $\Omega_B$  and  $\Omega_{R, 1 \mu m}$  are the Brillouin frequency shift (Equation 4.1) and the first harmonic ( $n = 1$ ) of the fundamental  $L_0$  Raman mode for  $L = 1 \mu m$  (Equation 4.2), respectively. The reported first harmonic Raman mode is also consistent across all material property tables in this chapter as calculated for  $L = 1 \mu m$ .

Table 4.1 shows that to measure Brillouin-induced Raman modes in an amorphous thin film of TeO<sub>2</sub>, the film may be at most  $\frac{L_{coh}}{2} \approx 41 \mu m$  (notably beyond the standard thickness considered to be a “thin film”) and CoBS must be capable of  $P_{CoBS, \frac{L_{coh}}{2}} \approx 137 \text{ pW}$  sensitivity. This makes clear the viability of TeO<sub>2</sub> as a good material platform for observing discrete Raman modes with the well-suited CoBS instrument. For a typical thin film of  $1 \mu m$  thickness the required CoBS sensitivity increases to  $\sim 76 \text{ fW}$ , still within the measured sensitivity limit, with excited phonons making on average  $\sim 80$  round trips within the film before dissipating.

To fabricate TeO<sub>2</sub> thin film samples we collaborated with Dr. John Gibbs' Nanotechnology Laboratory and its members to perform PVD of controlled films onto glass substrates. TeO<sub>2</sub> powder was initially attempted, via the recommended thermal evaporation method, however the fine TeO<sub>2</sub> powder was observed to ballistically repel from the thermal evaporation boat while still in solid powder form before it had the chance to melt into the liquid phase. To resolve this, we attempted a pretreatment procedure of applying

<b>TeO<sub>2</sub></b>	<b>Γ<sub>B</sub><sup>a</sup></b> (MHz)	<b>τ</b> (ns)	<b>v<sub>s, long</sub><sup>b</sup></b> (m s <sup>-1</sup> )	<b>n<sup>c</sup></b>	<b>L<sub>coh</sub></b> (μm)	<b>P<sub>CoBS, L<sub>coh</sub>/2</sub></b> (pW)	<b>Ω<sub>B</sub></b> (GHz)	<b>Ω<sub>R, 1 μm</sub></b> (GHz)
<b>Crystal</b>	2π·10	100	4260	2.2	430	~3.5 × 10 <sup>3</sup>	2π·12.1	2π·2.13
<b>Thin Film</b>	~2π·50 ± 10	~20	~4150	2.27	~83	~137	~2π·12.2	~2π·2.08

a: 101, b: 107–112, c: 113

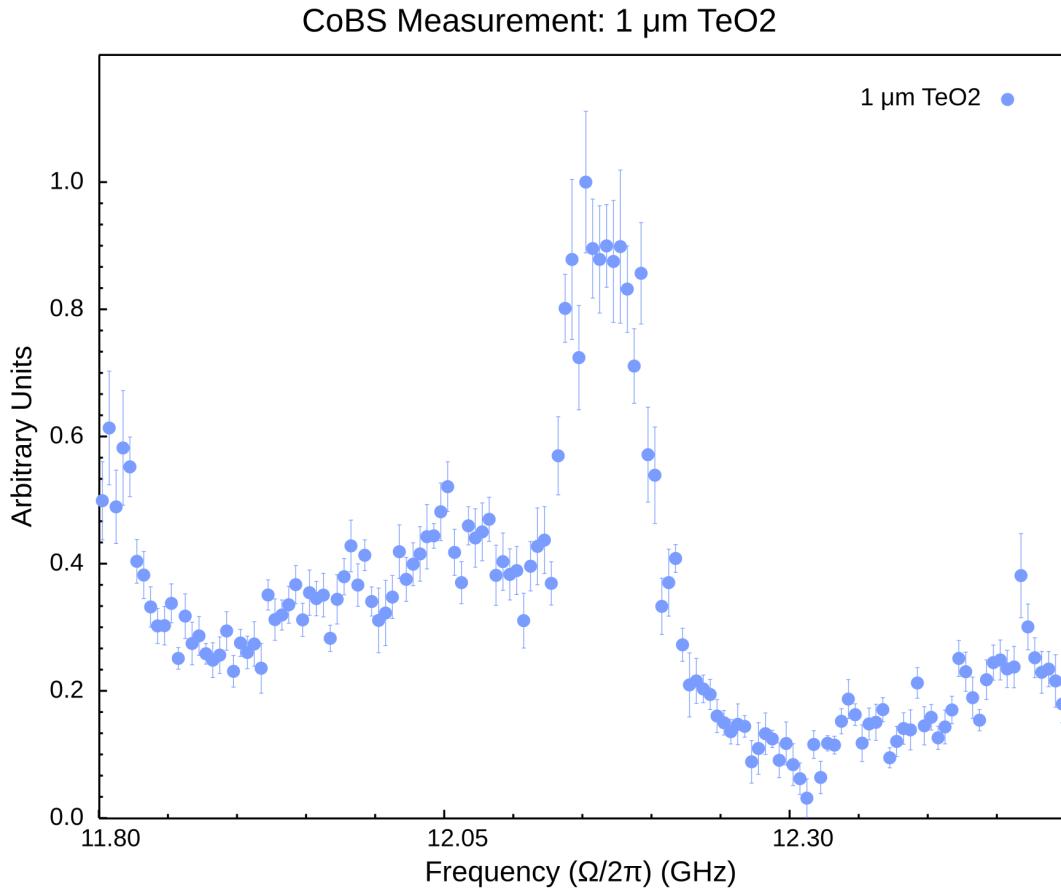
**Table 4.1:** Material parameters for TeO<sub>2</sub> relevant to observing Brillouin traveling-wave modes and Raman standing-wave modes. The first row gives values for crystalline TeO<sub>2</sub><sup>101, 113</sup>, while the second row reports measured values of our 1 μm TeO<sub>2</sub> thin film obtained from Figures 4.9 and 4.10. Here, Γ<sub>B</sub> is the angular Brillouin linewidth (phonon dissipation rate) and the inverse of phonon lifetime τ = (2π·Γ<sub>B</sub>)<sup>-1</sup>, v<sub>s, long</sub> is the longitudinal sound speed, and n is the refractive index. We take the refractive index for an amorphous thin film of TeO<sub>2</sub> as the average of the crystalline-TeO<sub>2</sub> ordinary (n(o) = 2.2) and extraordinary (n(e) = 2.34) refractive indices (n<sub>avg</sub> ≈ 2.27), giving an expectedly reduced longitudinal sound speed v<sub>s, film</sub> ≈ 4150 m s<sup>-1</sup> in the imperfect lattice of the physical vapor deposition (PVD)-deposited film. l<sub>coh</sub> is the phonon coherence length (mean travel distance), scattered power for a CoBS process (P<sub>CoBS</sub>), reported here for L<sub>coh, crystalline</sub> = 2 · 215 μm and L<sub>coh, thin film</sub> = 2 · 41.5 μm TeO<sub>2</sub>, scales with L<sup>2</sup> (Equation 4.3). P<sub>CoBS</sub> improves slightly in the thin film due to the higher refractive index, but worsens significantly by faster dissipation Γ<sub>B</sub>. Finally, Ω<sub>B</sub> is the angular Brillouin frequency shift (Equation 4.1), and Ω<sub>R, 1 μm</sub> is the first harmonic (n = 1) of the fundamental L<sub>0</sub> Raman-like mode for L = 1 μm (Equation 4.2).

high-compression forces to the TeO<sub>2</sub> powder with a hydraulic press. The resulting compressed plates of TeO<sub>2</sub> powder (pictured in Figure 4.3) maintained stability in the thermal boat under vacuum and heating conditions, indicating initial success. However, the tungsten thermal boat twice melted together with the compressed TeO<sub>2</sub> plates, halting the deposition and preventing the completion of the sample.

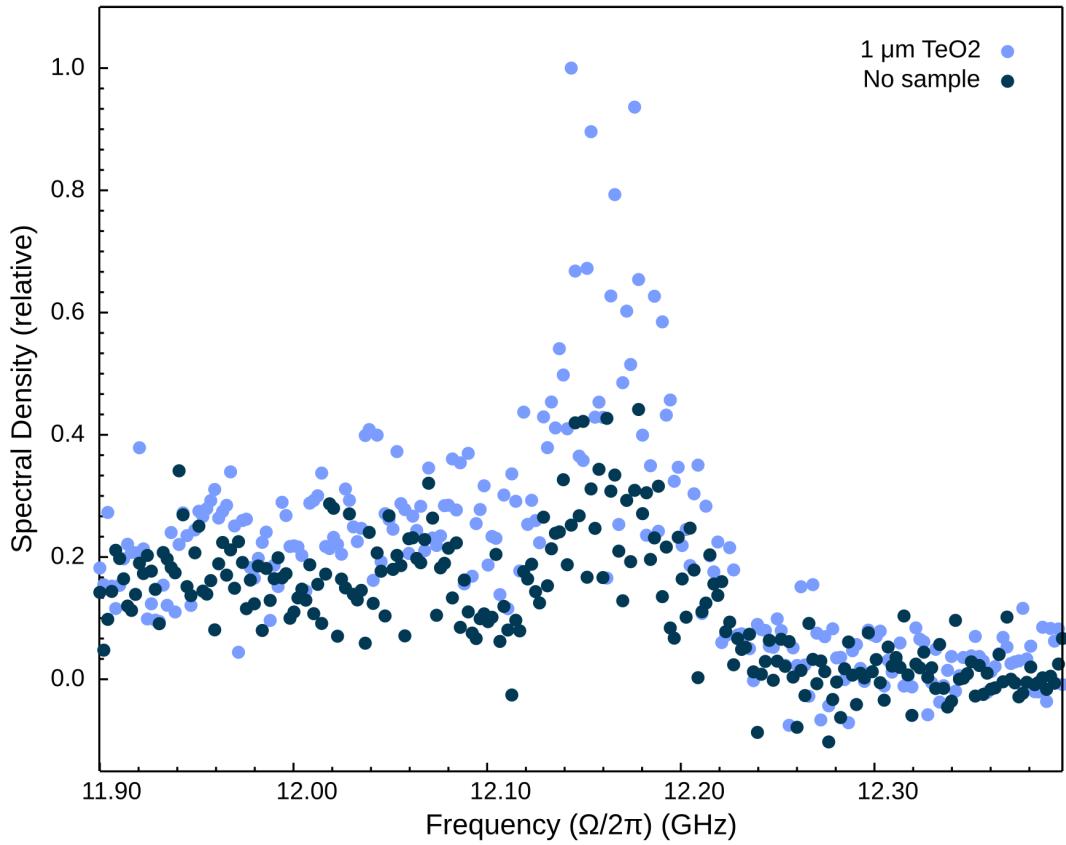
To surmount this issue, we resolved to deposit Te metal and apply a post-deposition annealing treatment to oxidize the Te into the desired TeO<sub>2</sub>. This fabrication procedure proved successful. Initial deposition rate stability issues were resolved by inserting custom electrical circuit logic into the deposition chamber voltage controls. The circuit logic provided added sensitivity of deposition parameters by introducing a switchable “course” and “fine” voltage control mode, with the fine setting containing the phenomenologically-found sweet-spot of resistance for thermally depositing Te metal. In our experience, thermally depositing μm films of Te metal via multiple successive deposition-annealing cycles of annealable sublayers, occurring over ∼20 h, and keeping the deposition rate steady with continuous adjustment of the fine-control knob is akin to rolling a ball lengthwise down the outside of a pipe of diameter equal to the ball’s. Nevertheless, this procedure proved a reliable method of producing TeO<sub>2</sub> thin films deposited onto glass slides which could easily be inserted and secured in the beam path of the CoBS instrument. Figure 4.8 shows an image of a glass slide with a deposited thin film of TeO<sub>2</sub> which has been successfully annealed, converting the mirror-shine appearance



**Figure 4.8:** Glass slide with a  $1\text{ }\mu\text{m}$  thin film of  $\text{TeO}_2$  deposited via PVD and subsequently annealed.

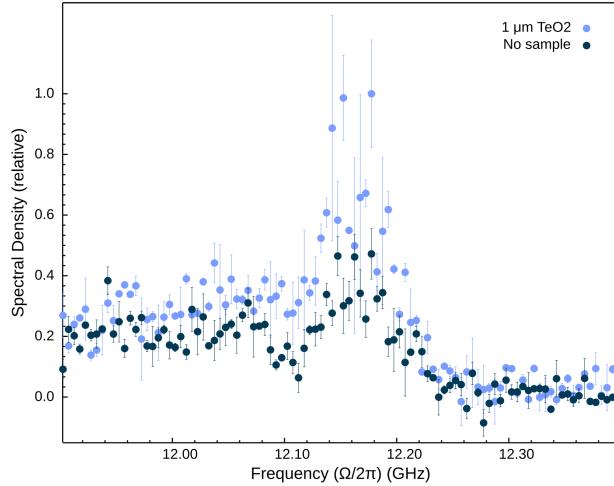


### CoBS Measurement: 1 $\mu\text{m}$ TeO<sub>2</sub>



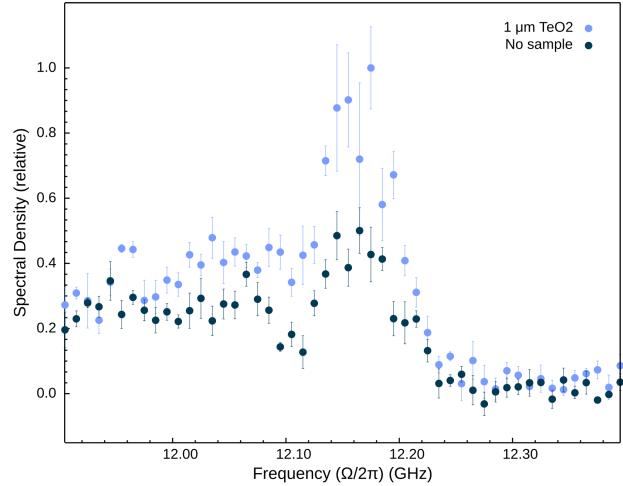
(a) Raw resolution.

### CoBS Measurement: 1 $\mu\text{m}$ TeO<sub>2</sub>



(b) 5 MHz binning.

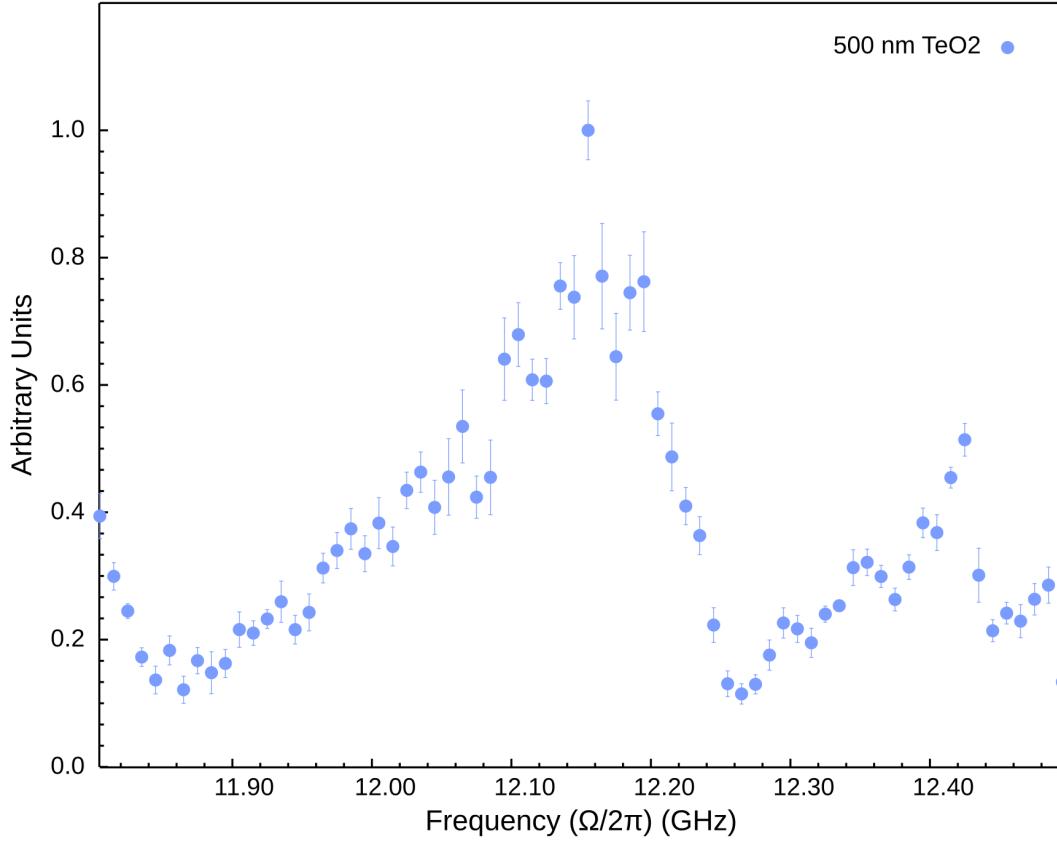
### CoBS Measurement: 1 $\mu\text{m}$ TeO<sub>2</sub>



(c) 10 MHz binning.

**Figure 4.10:** Observed background-subtracted spectra obtained through a CoBS measurement of 1  $\mu\text{m}$  of TeO<sub>2</sub> captured at maximum operating optical powers  $P_{\text{P}}P_{\text{S}}P_{\text{Pr}} \approx 0.25 \text{ W}^3$ , with  $1\sigma$  uncertainties. Each panel shows the same data set under different binning resolutions.

### CoBS Measurement: 500 nm TeO<sub>2</sub>



**Figure 4.11:** Observed background-subtracted spectrum obtained through a CoBS measurement of 500 nm of TeO<sub>2</sub> captured at maximum operating optical powers  $P_{\text{P}}P_{\text{S}}P_{\text{Pr}} \approx 0.25 \text{ W}^3$ , with  $1\sigma$  uncertainties.

Figure 4.9 shows a CoBS measurement of a 1  $\mu\text{m}$  thin film of TeO<sub>2</sub>. From reported sound speed and refractive index for TeO<sub>2</sub>,<sup>107–113</sup> Equation 4.1 predicts a Brillouin frequency shift for TeO<sub>2</sub> to be  $f_{\text{B}} \approx (12.4 \pm 0.5)$  GHz. An independent measurement was performed on a second 1  $\mu\text{m}$  thin film of TeO<sub>2</sub> prepared in the same way. For this second measurement, after capturing the TeO<sub>2</sub> film spectra, a repeated measurement was immediately performed in which the TeO<sub>2</sub> sample was removed and a measurement was taken under identical parameters. Figure 4.10 shows the result of these two measurements in three variations in resolution via binning: unbinned raw resolution (Figure 4.10a), 5 MHz bins (Figure 4.10b), and 10 MHz bins (Figure 4.10c). While perfectly crystalline TeO<sub>2</sub> features a smaller linewidth  $\sim 10$  MHz,<sup>101</sup> dissipation within a rough amorphous thin film of varying grain size and imperfect crystal structure would feature significantly higher dissipation (wider linewidth) than its crystalline counterpart. These same imperfections would contribute to a slightly slower sound speed in the deposited TeO<sub>2</sub>, pushing the Brillouin frequency shift slightly higher (see Equation 4.1). As previously noted, a measurement of 1  $\mu\text{m}$  TeO<sub>2</sub> would require at least as great as  $\sim 76$  fW, which is within the capability of the demonstrated  $\sim 5$  fW sensitivity of our

Te	$\Gamma_B^a$ (MHz)	$\tau$ (ns)	$v_{s, \text{long}}^b$ (m s <sup>-1</sup> )	n <sup>c</sup>	L <sub>coh</sub> (μm)	P <sub>CoBS, 500 nm</sub> (pW)	$\Omega_B$ (GHz)	$\Omega_{R, 1 \mu\text{m}}$ (GHz)
<b>Bulk</b>	$\sim 2\pi \cdot 10$	100	$\sim 2610$	4.58	261	$\sim 80 \times 10^{-3}$	$2\pi \cdot 15.4$	$2\pi \cdot 1.31$

a: 119–123, b: 119–124, c: 118, 125

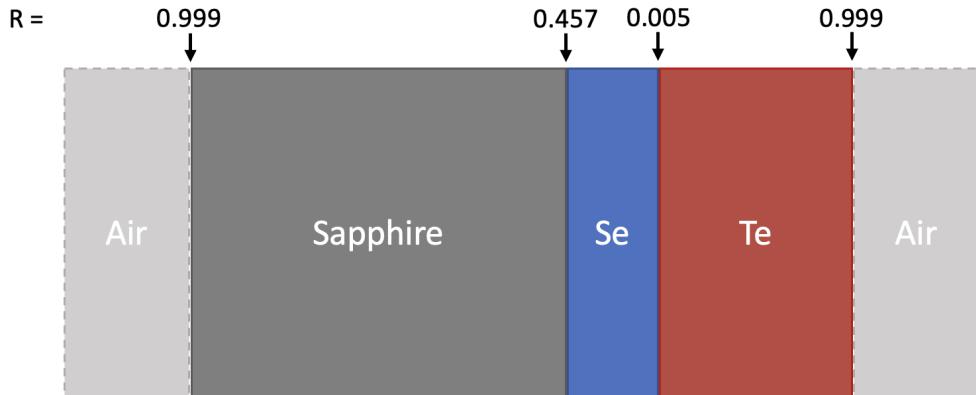
**Table 4.2:** Material parameters for Te relevant to observing Brillouin traveling-wave modes and Raman standing-wave modes, obtained from published values for bulk Te as well as for Te thin films. Here,  $\Gamma_B$  is the angular Brillouin linewidth (phonon dissipation rate) and the inverse of phonon lifetime  $\tau = 2\pi\Gamma_B^{-1}$ ,  $v_{s, \text{long}}$  is the longitudinal sound speed,  $n$  is the refractive index,  $l_{\text{coh}}$  is the phonon coherence length (mean travel distance), and  $P_{\text{CoBS}}$  is the scattered power for the CoBS process, reported here for 500 nm Te, and scales with  $L^2$  (Equation 4.1). Finally,  $\Omega_B$  is the angular Brillouin frequency shift (Equation 4.1), and  $\Omega_{R, 1 \mu\text{m}}$  is the first harmonic ( $n = 1$ ) of the fundamental  $L_0$  Raman-like mode for  $L = 1 \mu\text{m}$  (Equation 4.2). While CS<sub>2</sub> and TeO<sub>2</sub> are transparent at 1.55 μm, Te is absorptive here. However, transmission becomes meaningful through thin films, with 1 μm of deposited Te permitting 11.4% and 500 nm permitting 29% of 1.55 μm light to transmit.<sup>118</sup> This extra ~70% loss (for each of the three optical fields) has been accounted for in the scattered power value for the CoBS process listed in the table.

CoBS instrument. Further measurements are required to confirm a measurement of a 1 μm deposited thin film of TeO<sub>2</sub> with the CoBS instrument.

Figure 4.11 shows a spectrum obtained for a 500 nm thin film of TeO<sub>2</sub>. A pronounced background noise profile dominates the spectrum, with no significant spectral features indicative of a measurement of TeO<sub>2</sub>. While the scattered power produced from 500 nm of TeO<sub>2</sub> ( $P_{\text{CoBS}, 500 \text{ nm TeO}_2} \approx 19 \text{ fW}$ ) remains just within the sensitivity limit of the instrument, in practice, a host of non-ideal experimental conditions could prevent the instrument from performing at its full sensitivity limit. In search of other options, and having worked extensively with Te metal over so many hours of PVD depositions, we began researching the possibility of using un-annealed Te over its oxidized counterpart due to the meanwhile discovered tremendously higher material gain factor offered by Te the metal over its oxidized state TeO<sub>2</sub>.

#### 4.3.4 Te Thin Film

Table 4.2 gives the relevant parameters for Te metal. While Te is absorptive at 1.55 μm, transmission becomes meaningful as films become thinner, with a 1 μm thin film of Te permitting 11.4% of 1.55 μm light to transmit and a 500 nm film permitting 29%.<sup>118</sup> The  $n = 4.58$  refractive index of Te contributes to a massive material Brillouin gain of  $g_0 = 13.1 \text{ m GW}^{-1}$ , translating to an acousto-optic overlap-adjusted effective Brillouin gain  $G_B = 14.4 \text{ m}^{-1} \text{ W}^{-1}$  (see Figure 4.3). With no successful measurements for TeO<sub>2</sub> thin films achieved, this much higher gain of Te presented an enticing opportunity.



**Figure 4.12:** Illustration of the material layers of a Te thin film sample. Sapphire ( $\text{Al}_2\text{O}_3$ ) substrates are primed with a 10 nm deposited layer of Se by collaborators Dr. Alessandro Mazza of Los Alamos National Laboratory (Los Alamos National Laboratory (LANL)) and the Center for Integrated Nanotechnologies (Center for Integrated Nanotechnologies (CINT)) and Dr. Matthew Brahlek of Oak Ridge National Laboratory (Oak Ridge National Laboratory (ORNL)). This Se layer acts as an adhesion layer between the sapphire and the Te film. Acoustic reflectivity values at each interface as a result of the acoustic impedance mismatch between materials are shown along the top of the illustration for each material boundary.



**Figure 4.13:** Evidence of oxidization of the 500 nm Te thin film in the circular beam spot due to  $\sim 70\%$  absorption of the  $1.55 \mu\text{m}$  optical fields.

Initial CoBS measurements of Te thin films resulted in evident ablation of the film from the glass substrate, with poor adhesion between the Te and glass slide the determined cause. Collaborators at the Center for Integrated Nanotechnologies (CINT), a joint center affiliated with LANL and Sandia National Laboratory (SNL), lent the insight that a thin adhesion layer of Se is recommended between Te deposited onto glass. Se-primed sapphire substrate samples were kindly prepared and sent to us by Dr. Alessandro Mazza of LANL and Dr. Matthew Brahlek of ORNL, ready for depositions of Te to be performed ontop of a 10 nm Se layer. The Se adhesion layer proved successful in adhering the Te. A diagram of the material layering is shown in Figure 4.12. A 500 nm thin film of Te was fabricated after estimations of a feasible measurement of the material showed a required sensitivity  $\sim 15\times$  the sensitivity limit of the instrument. During measurement, it was discovered that the energy of the optical powers incident on the sample (of which  $\sim 71\%$  is absorbed

$\text{CS}_2$	$\Gamma_B^a$ (MHz)	$\tau$ (ns)	$v_{s, \text{long}}^b$ ( $\text{m s}^{-1}$ )	$n^c$	$L_{\text{coh}}$ ( $\mu\text{m}$ )	$P_{\text{CoBS}, L_{\text{coh}}/2}$ (pW)	$\Omega_B$ (GHz)	$\Omega_{R, 1\mu\text{m}}$ (GHz)
<b>Liquid</b>	$2\pi \cdot 90 \pm 10$	10	1150	1.59	$13 \pm 2$	$\sim 7.2$	$2\pi \cdot 2.54 \pm 0.03$	$2\pi \cdot 0.575$

a: 13, 23, 105, 106, b: 13, 23, 40, 126, c: 13, 23

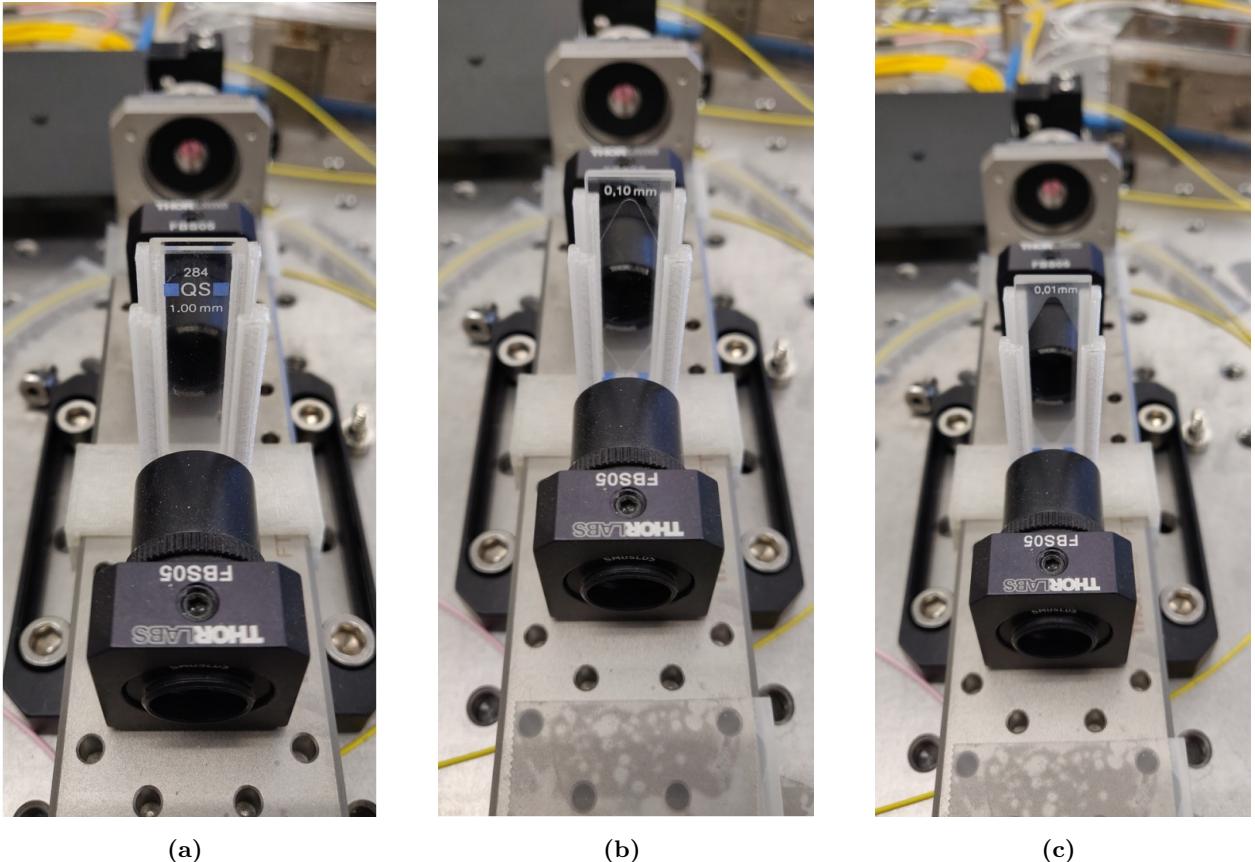
**Table 4.3:** Material parameters for bulk liquid  $\text{CS}_2$  relevant to observing Brillouin traveling-wave modes and Raman standing-wave modes, obtained from published values as well as our own observations shown in Figures 4.7a, 4.7b, 4.15a, and 4.15b. Here,  $\Gamma_B$  is the angular Brillouin linewidth (phonon dissipation rate) and the inverse of phonon lifetime  $\tau = 2\pi\Gamma_B^{-1}$ ,  $v_{s, \text{long}}$  is the longitudinal sound speed,  $n$  is the refractive index,  $l_{\text{coh}}$  is the phonon coherence length (mean travel distance), and  $P_{\text{CoBS}}$  is the scattered power for the CoBS process, reported here for  $L_{\text{coh}} = 2 \cdot 6.5\mu\text{m}$   $\text{CS}_2$ , and scales with  $L^2$  (Equation 4.3). Finally,  $\Omega_B$  is the angular Brillouin frequency shift (Equation 4.1), and  $\Omega_{R, 10\mu\text{m}}$  is the first harmonic ( $n = 1$ ) of the fundamental  $L_0$  Raman-like mode for  $L = 1\mu\text{m}$  (Equation 4.2).

as previously discussed) caused significant heating in the thin film of Te, resulting in oxidization of the Te, changing its state into  $\text{TeO}_2$  within the beam spot. Figure 4.13 shows a photograph which captures this resulting oxidized state of the Te thin film after sitting for a measurement in the beam path of the instrument.

With this result, we began a broader reanalysis of the choice of platform for observing Brillouin-induced Raman modes at room temperature. Figure 4.12 also shows acoustic reflectivity values  $R$  for each interface of a Te sample, calculated by Equations 4.7 and 4.6. While the Te-air boundary offers near complete acoustic reflection, owing to the large acoustic impedance mismatch between Te and air, the acoustic reflectivity is near zero between Te and Se and only  $\sim 50$

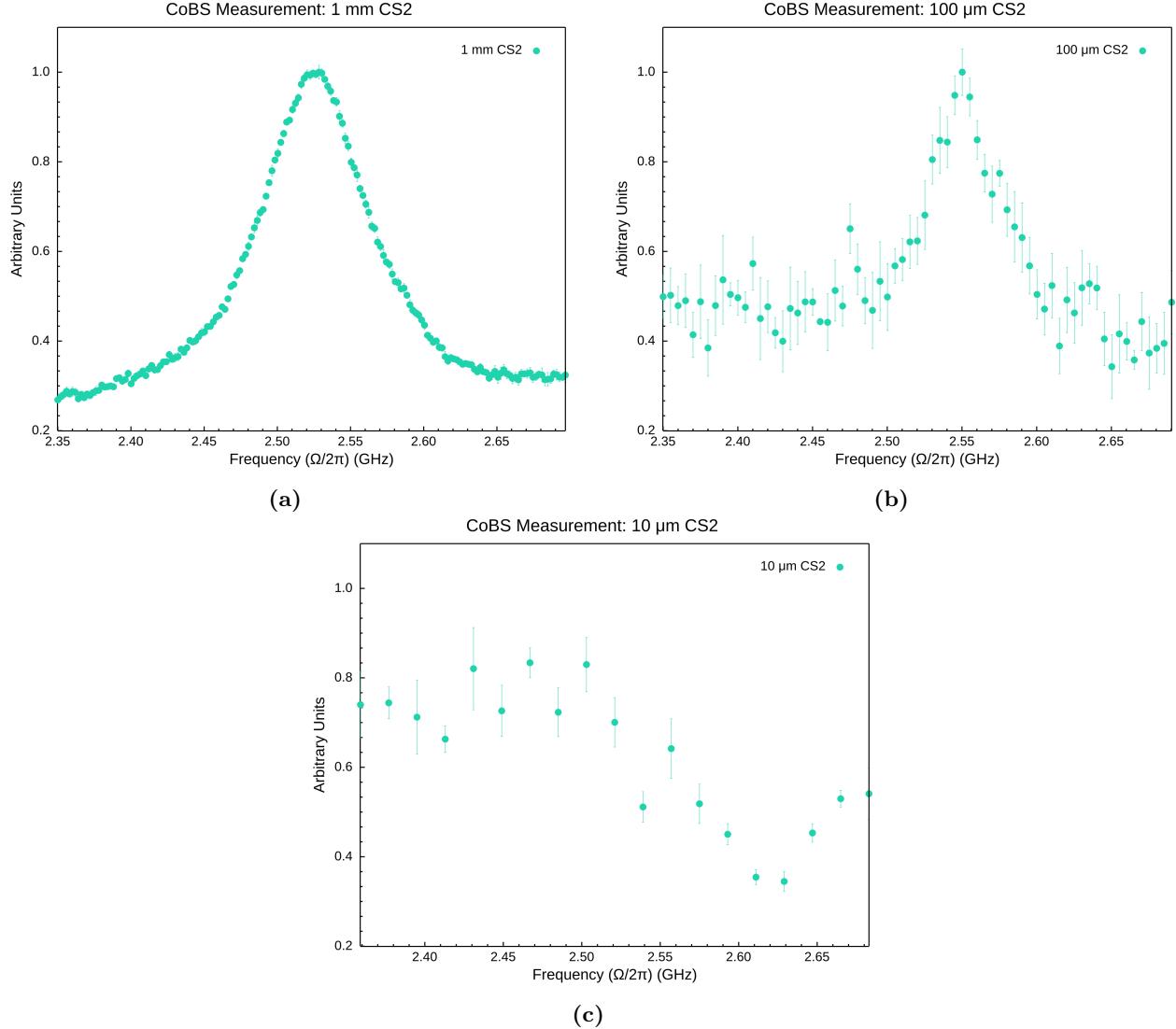
#### 4.3.5 $\text{CS}_2$ Micrometer Cell

Table 4.3 gives the relevant material parameters for achieving Brillouin-induced Raman modes in  $\text{CS}_2$ . Notably, the phonon dissipation rate within  $\text{CS}_2$  is nearly  $10\times$  greater than that of  $\text{TeO}_2$  or Te, at  $\Gamma_B = (90 \pm 10)$  MHz. This, in combination with its slower speed of sound  $V_s = 1150\text{ m s}^{-1}$ , results in a short phonon coherence length  $L_{\text{coh}} = (13 \pm 2)\mu\text{m}$  and a correspondingly high sensitivity requirement of  $P_{\text{CoBS}, 6.5\mu\text{m}} \approx 7.2\text{ pW}$ . While this is smaller than the half-coherent-length sensitivity required for  $\text{TeO}_2$  ( $6.5\mu\text{m}$  vs  $41.5\mu\text{m}$  for  $\text{TeO}_2$ ), making for a harder measurement to observe Raman modes in  $\text{CS}_2$  than  $\text{TeO}_2$  by this metric, the reduced complexity of the  $\text{CS}_2$  experimental platform as compared to the amorphous thin films presented as attractive. Additionally, the air- $\text{CS}_2$ -air boundaries represent near total acoustic confinement at the interfaces, and the narrower Raman mode spacing (multiples of  $f_{R, \text{CS}_2} = 575\text{ MHz}$ , as opposed to  $f_{R, \text{TeO}_2} = 2.13\text{ GHz}$ ) in combination with the wider linewidth of  $\text{CS}_2$  together offered the possibility of more achievable length-tuning for Raman mode placement near the Brillouin frequency shift.



**Figure 4.14:** Three  $\text{CS}_2$  cells of different path lengths (1 mm, 100  $\mu\text{m}$ , and 10  $\mu\text{m}$ ) secured in the beam path of the Coherently stimulated Brillouin Spectrometer.

Small path length cells were obtained for 1 mm, 100  $\mu\text{m}$ , and 10  $\mu\text{m}$  path lengths. Figure 4.14 show photographs of each of these cells inserted into the sample holder and installed into the sample stage of CoBS. Progressive measurements were attempted, with each 10 $\times$  decrease in effective length  $L$  corresponding to a 100 $\times$  decrease in scattered power produced, and thereby a 100 $\times$  higher sensitivity required for a successful measurement (see Equation 4.3). Figure 4.15a shows the observed spectra collected from a measurement of 1 mm liquid  $\text{CS}_2$ . This spectrum is part of a set of measurements collected on the same day and which helped reveal the Fano interference behavior common to CoBS measurements approaching the sensitivity limit. While the spectrum in Figure 4.15a shows mild to no Fano asymmetry, spectra corresponding to other pump-probe detunings display strong Fano asymmetry (see Section B.3.2 in Appendix B for a description of the experiment and Figure B.3 for the full measurement set), indicative of a measurement near the sensitivity limit of the instrument at the time. Figure 4.15b shows the spectrum obtained from a CoBS measurement of 100  $\mu\text{m}$  liquid  $\text{CS}_2$ . The signal-to-noise ratio (SNR) of this measurement is significantly lower than that of the 1 mm liquid  $\text{CS}_2$  spectrum, as expected from the 100 $\times$  reduction in scattered power produced as a result



**Figure 4.15:** Observed background-subtracted spectra obtained through CoBS measurements of (a) 1 mm, (b) 100  $\mu$ m, and (c) 10  $\mu$ m liquid CS<sub>2</sub> (pictured in Figure 4.15c), captured at maximum operating optical powers  $P_P P_S P_{Pr} \approx 0.25 \text{ W}^3$ . All measurements have  $1\sigma$  uncertainties smaller than the data point markers.

of the 10 $\times$  reduction in effective length. Finally, Figure 4.15c shows the spectrum obtained from a CoBS measurement attempt of 10  $\mu$ m liquid CS<sub>2</sub>, binned to 20 MHz bins. While the scattered power produced remains theoretically within the sensitivity limit of the instrument, a successful measurement of 10  $\mu$ m liquid CS<sub>2</sub> could not be obtained, likely due to variations in experimental conditions. Furthermore, due to the short phonon mean travel distance  $L_{coh} = (13 \pm 2) \mu\text{m}$ , the 10  $\mu$ m path length cell would still have been insufficiently short to exhibit Brillouin-induced Raman modes. A  $\sim$ 5  $\mu$ m path length cell would be required for this result.

#### 4.3.6 Suspended Silica Rib Waveguide

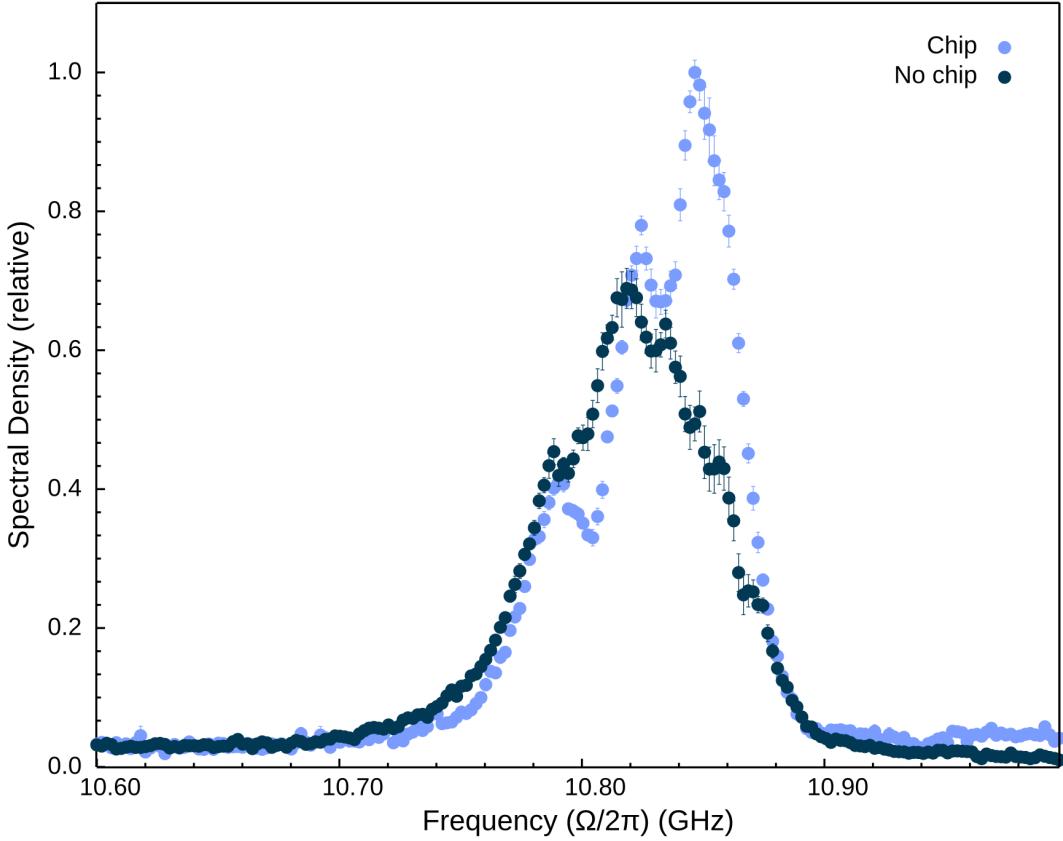
An exciting collaborative connection was made with Dr. Aaron Hawkins' Semiconductor Devices and Microfabrication group at Brigham Young University (BYU) which opened up new possibilities for photonic-phononic interactions and control of phonons at room temperature. The Hawkins group specializes in advanced chip-waveguide and device design and fabrication, introducing the possibility for a collaboration with a widened sandbox of available materials and platform designs. We received suspended silica rib waveguides for initial investigation and proof of principle. The goal was to measure backward Brillouin scattering in the silica rib of these suspended waveguides with the CoBS instrument. The main challenge of this task became evident immediately: getting light into the chip. The method available was via careful fiber-to-chip alignment to align the cone of light output from the fiber end for injection into the silica rib waveguide. Initial attempts were unsuccessful due to imprecise XYZ control and lack of experiential knowledge. After many weeks of unsuccessful attempts and several video consultations with the Hawkins group for guidance, a visit to the Hawkins group at BYU in Provo, UT was arranged to gain hands-on learning and experience in the laboratories and facilities at BYU. This friendly and illuminating trip immediately shored up the lacking knowledge and experience, allowing for immediate success getting light into the chip upon returning home. However, the silica rib waveguide closely shares many properties with the silica SMF-28 which comprises much of the CoBS apparatus as well as the entirety of the sample stage where all interacting light and sound waves overlap.

Initial measurements of the waveguide captured a spectral feature which seemed to match the known spectral response of the SMF-28 ( $\Omega_B \approx (10.8 \pm 0.5)$  GHz). However, with perseverance and improved alignment of light into and out of the chip waveguide, distinct spectral features were observed which differentiated the response of the chip waveguide from the background measurement performed with chip waveguide removed. Figure 4.16 shows spectra gathered subsequently with and without the chip waveguide present in the instrument. A clear feature of the chip at  $\sim 10.84$  GHz rises above the background SMF-28 response, centered at  $\sim 10.82$  GHz. This plausible measurement represents a significant step towards harnessing the opportunity of specialized chip-waveguide design to control phonons at room temperature and potentially observe Brillouin-induced Raman modes.

#### 4.3.7 Elastically-Suspended Photonic-Phononic Waveguide

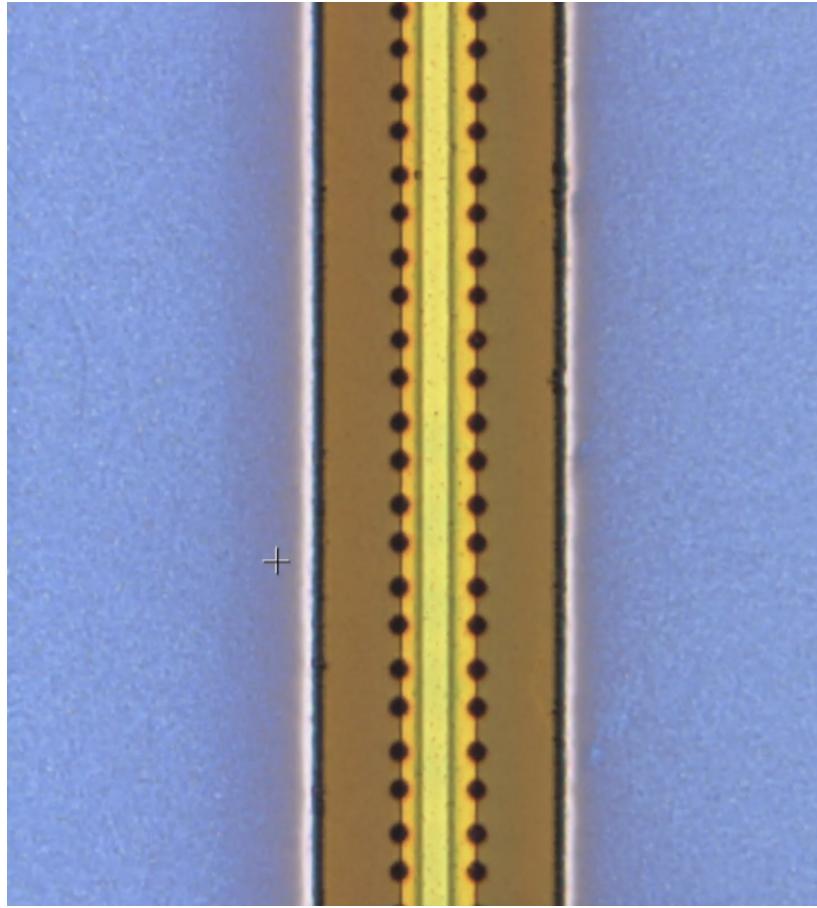
In this section we provide analysis of expected optomechanical behavior from a specialized photonic-phononic waveguide designed and fabricated by the Hawkins group at BYU. This device features a silica rib suspended on a thin memberane of elastic SU-8 polymer, primarily used as a negative photoresist which acts as a

## CoBS Measurement: 1 cm Suspended Silica Rib Waveguide



**Figure 4.16:** CoBS measurement of a suspended silica rib waveguide for a backward scattering process. A subsequent measurement with the chip waveguide removed is also plotted. The spectral response of the SMF-28 which comprises much of the sample stage of the instrument is visible without the chip, however a notable signal from the waveguide can be seen above the background. In obtaining the spectra, five repeated measurements of both the signal and background (probe off) were collected at a 100 Hz resolution bandwidth (RBW), dwelling for 1 s at each 5 MHz frequency step. Plotted is the resulting background-subtracted spectrum. Uncertainties represent  $1\sigma$  standard error of the mean.

sacrificial layer in forming complex layered designs. Through collaborative discussions sharing ideas and specific domain knowledge between us and the Hawkins group, Dr. Hawkins cleverly proposed we may see a large mechanical response from harnessing the elastic nature of the SU-8 for optomechanical interactions. In this proposed design scheme, stimulation of a breathing mode in the silica rib would induce a drumhead-like mode in the SU-8 suspension. In parallel to this design behavior, we also proposed to design notches or other similar acoustic boundary feature to support acoustic guidance down the length of the waveguides. Improved acoustic guidance would produce stronger opto-mechanical interactions between light and sound along the length of the waveguide due to more tightly confined mode overlap ( $A_{\text{eff}}^{ao}$ ). These lengthwise notches or other similar feature, we proposed, might be designed to also act as lengthwise acoustic resonators. These resonant acoustic cavity notches would trap traveling-wave phonons stimulated in the silica rib and

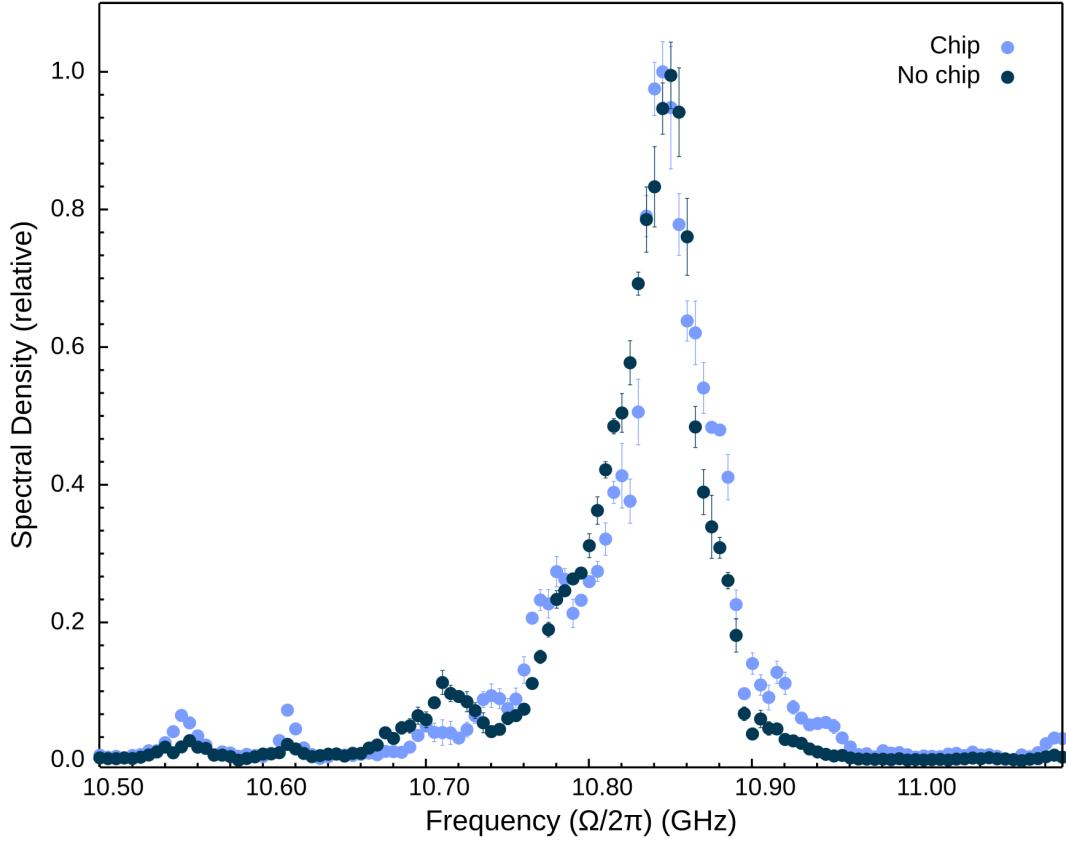


**Figure 4.17:** Microscope image of the elastically-suspended photonic-phononic waveguide designed by the Hawkins group at BYU. The brown region shows the SU-8 suspension, with the yellow region comprising the silica rib layers. The dark circles seen running lengthwise on either side of the silica rib are holes which, if made to be evenly spaced in a future design, would offer a clean n-multiplication of acoustic resonant cavities for longitudinally-traveling phonons (Brillouin-induced) to be trapped within the cavities to produce standing-wave patterns according to the effective material sound speed and hole spacing (Raman-like modes) according to Equation 4.2.

effectively produce a controllable n-multiple of short path length samples lengthwise along both sides of one chip device of desired half-coherence-length spacing. These design features were indeed implemented, and successful proof of principle elastically-suspended photonic-phononic waveguides were created and sent to us for testing.

Figure 4.17 shows a top-down microscope image of the elastically-suspended photonic-phononic waveguide device designed by the Hawkins group at BYU. Dark circles seen running lengthwise on either side of the silica rib are holes which, if made to be evenly spaced in a future design, would offer a clean n-multiplication of acoustic resonant cavities for longitudinally-traveling phonons (Brillouin-induced) to be trapped within the cavities to produce standing-wave patterns according to the effective material sound speed and hole

## CoBS: 1 cm Elastically-Suspended Photonic-Phononic Waveguide



**Figure 4.18:** CoBS measurement of the elastically-suspended photonic-phononic waveguide for a backward scattering process. A subsequent measurement with the chip waveguide removed reveals a similar spectral profile, as expected from the identical resonant frequency response of the SMF-28 which comprises much of the apparatus, especially in the sample region of the instrument where all three optical powers overlap. In obtaining the spectra, five repeated measurements of both the signal and background (probe off) were collected at a 100 Hz RBW, dwelling for 1 s at each 5 MHz frequency step. Plotted is the resulting background-subtracted spectrum. Uncertainties represent  $1\sigma$  standard error of the mean.

spacing (Raman-like modes) according to Equation 4.2. Figure 4.18 plots the spectra collected from an initial test CoBS measurement of backward Brillouin scattering in the elastically-suspended silica rib of the photonic-phononic waveguide. While some spectral features differ between the “Chip” and “No chip” measurements, additional measurements and testing are required to confirm or deny the significance of these apparent features.

In what follows, we present preliminary theoretical estimates of the key mechanical modes and optomechanical couplings predicted for the elastically-suspended photonic-phononic waveguide under development. The structure under consideration consists of a polymeric (SU-8) membrane that is tensioned during high-temperature curing and supports a rib waveguide above it. This combined membrane-and-rib geometry gives rise to multiple distinct mechanical resonances that can be excited and probed optically via the CoBS

instrument operating in the forwards scattering configuration. We summarize below the principal modes, referred to as “drumhead” modes for the membrane and “breathing” modes for the rib, and estimate rough frequency ranges in which they are expected to appear. We first consider the rib portion of the waveguide, having characteristic lateral width and thickness of 4 cm and 6 cm, respectively. One can treat the smallest cross-sectional dimension, denoted  $d$ , as setting the approximate half-wavelength condition for its fundamental breathing mode. The simplest estimate for such a half-wavelength mechanical resonance is given equivalently by Equation 4.2, restated here as

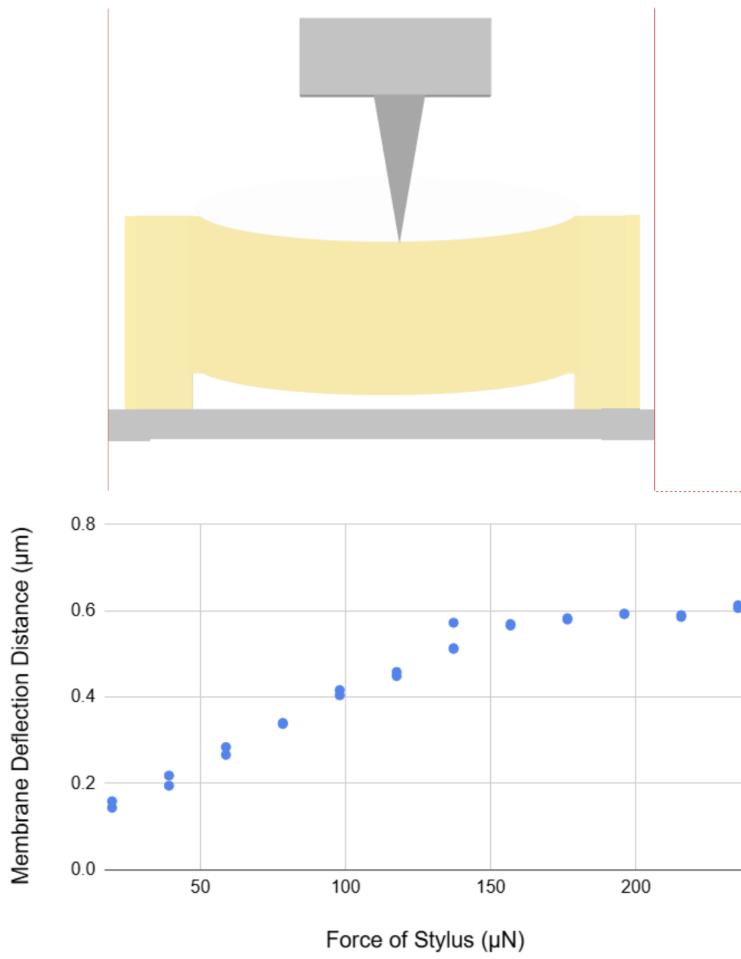
$$f_{\text{rib,breathe}} \approx \frac{v_s}{2d}, \quad (4.9)$$

where  $v_s$  is the speed of sound in the silica rib and  $d$  is the lateral width. Taking the rib to be 6  $\mu\text{m}$  wide and assuming a speed of sound in the range  $5 \text{ km s}^{-1}$  to  $6 \text{ km s}^{-1}$ , this places the fundamental rib breathing mode near several hundred MHz (estimates range from 400 MHz to 500 MHz). Higher-order modes can arise (integer multiples of the half-wavelength condition), and so in practice one expects a series of possible breathing modes in the 100s of MHz to low-GHz range.

In principle, when optically excited via the forward CoBS process, the rib breathing mode can couple to and drive displacement of the underlying elastic polymer membrane. Conversely, membrane stimulation at or near the rib’s breathing-frequency range (according to the membrane’s thickness) can induce motion in the rib. Thus, the device design permits either the drumhead mode of the membrane or the rib breathing mode to be excited (off-resonance if necessary) and potentially induce mechanical vibration in the other. Beneath the rib, the polymer membrane is suspended over an open region which has been etched away, and is anchored on either side. We thus expect a classical drumhead family of modes in which the membrane oscillates out-of-plane. The tension provided by the fabrication curing process indicates that the membrane vibrates in accordance with a tension-dominated regime. For a square membrane of dimension  $L$ , under tension per unit thickness  $T$  and with mass density  $\rho$  and thickness  $h$ , the fundamental frequency (1,1) can be approximated by<sup>127–129</sup>

$$f_{\text{membrane, drumhead (1,1)}} \approx \frac{v_s}{\sqrt{2}L}, \quad (4.10)$$

where  $v_s$  is the *transverse* sound speed of the membrane, given by



**Figure 4.19:** Needle profilometer force-deflection measurements (illustrated above) performed on the suspended polymer membrane of the photonic-phononic waveguide. The inverse slope of the data (taken to be linear) is proportional to the tension of the membrane ( $m \propto T$ ). Using  $m \approx 300 \text{ N m}^{-1}$ , we estimate the tension of the membrane to be  $T \approx 2 \text{ mN m}^{-1}$ , giving a transverse sound speed  $v_s \approx 1 \text{ m s}^{-1}$  via Equation 4.11. Figure and data courtesy of Adams et al., Hawkins group, *manuscript in prep.*

$$v_s = \sqrt{\frac{T}{\rho_A}} = \sqrt{\frac{\sigma h}{\rho h}} = \sqrt{\frac{\sigma}{\rho}}, \quad (4.11)$$

where  $\rho_A$  and  $\rho$  are the areal and volumetric mass density of the membrane, respectively, and  $\sigma$  is the stress on the membrane (Pa) corresponding to the tension per unit membrane thickness ( $\sigma = T/h$ ).

Modeling the polymer membrane as a square membrane, we can estimate the fundamental membrane drumhead mode using a mean density of SU-8 ( $\rho = 1190 \text{ kg m}^{-3}$ )<sup>130</sup> and needle profilometer tension measurements performed on the membrane. Figure 4.19 plots deflection for various stylus force values applied to

the membrane, as measured by Adams et al. of the Hawkins group (*manuscript in prep*) for the elastically-suspended photonic-phononic device under consideration. The inverse of the slope of these data (taken to be linear,  $m \approx 300 \text{ N m}^{-1}$ ) allows for the membrane's approximate tension to be found via  $T \approx \frac{L}{4}m \approx 2 \text{ mN m}^{-1}$  for a square tension-dominated membrane with side length  $L$  and a small central point load.<sup>128, 129</sup> For a 2  $\mu\text{m}$ -thickness with Equation 4.11, this gives an approximate transverse sound speed of a very slow  $v_s \approx 1 \text{ m s}^{-1}$ . Inserting this into Equation 4.10 with length  $L \approx 25 \mu\text{m}$ , we anticipate the fundamental drumhead mode resonant frequency of the SU-8 membrane to lie around 30 kHz. Meanwhile, the rib breathing mode are predicted to lie around the mid-100s of MHz. Given this 4-order-of-magnitude discrepancy in resonant frequencies, the two mechanical modes are not expected to exhibit mechanical coupling, even when accounting for the possibility of off-resonant driving. To have the two resonances approach each other, one might alter the fabrication parameters to try significantly increasing the resulting tension of the membrane, perhaps by experimenting with different bake times or temperatures and membrane thicknesses. Alternatively, or additionally, shortening the length  $L$  across which the membrane spans from clamp-to-clamp would minimally nudge its resonant frequency higher.

## 4.4 Discussion

In this chapter, we have investigated the potential for creating Raman-like vibrational modes via Brillouin interactions in a variety of photonic and phononic structures. By examining how traveling-wave phonons can be transformed into standing-wave resonances, we bridge the gap between conventional Brillouin physics (characterized by traveling acoustic phonons) and a regime reminiscent of Raman scattering (characterized by standing-wave vibrations). Our experimental and theoretical efforts illustrate both the challenges and the promising pathways for realizing stable, high-quality, Brillouin-induced Raman modes in waveguides, crystals, and fibers. Below, we synthesize the key findings and discuss future directions.

### 4.4.1 Pathways to Brillouin-Induced Raman Modes

A central goal has been to identify platforms capable of generating high- $Q$  standing-wave vibrational modes, analogous to conventional Raman-active modes, by leveraging Brillouin coupling in materials where both the optical and the acoustic (or elastic) degrees of freedom can be engineered. These results highlight how geometry, acoustic impedance contrasts, and overall device configuration govern the competition between traveling- and standing-wave phonons. Here, we group the ideal platforms by three broad categories: waveguides,  $\text{TeO}_2$  crystals, and fiber-based approaches. A promising strategy for forming Raman-like resonances emerges from waveguide geometries that confine both light and sound. Notably, rib waveguides of  $\text{TeO}_2$  (or

other similar high-Brillouin-gain chalcogenide material composition) are attractive due to the high photoelastic coefficients of tellurite and chalcogenide glasses, coupled with the possibility of incorporating evenly spaced square holes to pin acoustic modes. The fabrication of a long rib waveguide on the order of centimeters can allow an extended optical interaction length while introducing a phononic band structure via an engineered hole pattern. This pattern can create a 1D phononic bandgap, effectively confining traveling-wave phonons to form standing waves at selected frequencies. Tellurite- or other chalcogenide-based waveguides combine strong Brillouin gain with a sufficiently low acoustic damping rate to enable coherent standing-wave formation at room temperature (Section 4.3.3). As in earlier suspended photonic-phononic waveguides (Section 4.3.7), the tension in the membrane can significantly alter the acoustic mode spectrum. Proper design of periodic corrugations and sidewalls ensures that back-reflection of phonons gives rise to well-defined modes at the material Brillouin frequency shift. By tuning geometric parameters such as hole spacing and their proximity to the rib waveguide, it is possible to achieve a strong acousto-optic overlap while allowing the transduced traveling-wave phonons to “spill” out to the holes for selective Raman-like mode conversion.

Although many of our demonstrations focused on thin-films of  $\text{TeO}_2$ , a bulk or thick-crystal platform is also a compelling route for Brillouin-induced Raman effects.  $\text{TeO}_2$  boasts among the largest photoelastic coefficients for chalcogenide or oxide glasses, making it an exemplary host for strong acousto-optic coupling. In a bulk crystal with polished facets or embedded reflectors, backward Brillouin scattering may transition into a standing-wave configuration within the CoBS sensitivity limits, allowing observable Raman-like resonances. Relative to many polymeric or silica systems, crystalline  $\text{TeO}_2$  can exhibit reduced phonon damping, particularly for carefully oriented crystal axes. This intrinsic advantage can facilitate a higher mechanical  $Q$  and a narrower linewidth (longer phonon coherence length  $L_{\text{coh}}$ ), key ingredients for stable Brillouin-induced Raman modes (see Table 4.1). Realizing a clear set of discrete “Raman-like” lines in a bulk crystal requires controlling reflections at each interface. Spurious acoustic reflections or partial mode mismatch can excite multiple unwanted phonon branches, broadening the spectrum. Future work could integrate an acoustic Bragg grating within the crystal or deposit patterned reflectors on crystal faces to support more robust mode confinement.

Finally, optical fibers remain one of the most convenient photonic platforms for combining long interaction lengths with precise control over acoustic guidance. Introducing a series of periodic notches or refractive index modulations forms an acoustic mirror inside the fiber, providing a route to localized standing-wave phonons. Unlike conventional uniform-core fibers where traveling acoustic waves dominate, these fiber Bragg structures could reflect phonons at targeted frequencies. Once the phonon reflectivity is sufficiently high, coherent standing waves may arise. Implementing an acoustic fiber Bragg grating must balance strong reflection of phonons (to produce standing-wave modes) with minimal introduction of optical loss. While

doping or microstructuring the fiber is feasible, the potential for scattering or wavefront distortion may ultimately impact optical guidance.

Collectively, these three categories ( $\text{TeO}_2$  waveguides,  $\text{TeO}_2$  crystals, and notched acoustic fibers) demonstrate overlapping yet distinct methods for accessing Brillouin-induced Raman modes. Each system can be fine-tuned for specific applications:  $\text{TeO}_2$  waveguides for integrated photonics where footprint is critical; bulk crystals for high- $Q$  acoustic experiments; and fiber-based geometries for long-distance coupling and simpler device integration.

#### 4.4.2 Conclusion

By mapping the transition from traveling-wave Brillouin phonons to standing-wave Raman-like modes across these diverse platforms, we underscore the broad adaptability of optomechanical coupling at room temperature. The demonstrations in this chapter affirm that controlling acoustic boundary conditions, whether via physical reflectors, periodic perturbations, or intrinsic interface mismatches, lies at the center of generating stable, high- $Q$  vibrational modes. The overarching theme of the work presented in this document is control of phonons at room temperature. The progress reported here highlights that we do not need to confine ourselves exclusively to classical Raman systems or purely Brillouin-based traveling-wave scenarios. Instead, it is possible to strategically blend the strengths of each to realize new types of hybrid resonances with unique spectral signatures. Ultimately, these Brillouin-induced Raman modes broaden the scope of optomechanical interactions. They show that even modest structural modifications, whether in waveguides, bulk crystals, or specialized fibers, can tip the balance away from purely traveling acoustic waves toward fully resonant, discrete modes. This tunability promises new device functionalities at telecom and near-infrared wavelengths, where controlling phonons in tandem with photons stands to accelerate advances in sensing, signal processing, and quantum photonics.





## Chapter 5

### Conclusion

Throughout this dissertation, we have explored how Brillouin-based optomechanics can be harnessed for both practical device applications and fundamental studies of light–matter interactions. Specifically, we demonstrated that traveling-wave phonons in a liquid-core optical fiber can be cooled significantly through the anti-Stokes Brillouin scattering process. This result marks an important step in transitioning from the established model of discrete cavity modes toward the more general case of continuous-waveguide phonons, showing that key regimes of laser cooling can be accessed without relying on highly engineered photonic microstructures.

In tandem, this dissertation introduced the concept of a Coherently stimulated Brillouin Scattering (CoBS) spectrometer, which relaxes traditional phase-matching constraints and dramatically increases measurement sensitivity for short-length or low-gain samples (offering a  $\sim 10^6$  increase in scattered power for  $\sim 1$  cm lengths). By incorporating a four-wave mixing scheme (pump, probe, driven Stokes, and backscattered signal) the spectrometer opens new avenues for measuring the mechanical modes of thin films, microscale liquid volumes, and emerging photonic–phononic waveguides. This flexibility is crucial for advancing applications ranging from precision material characterization to integrated acousto-optic devices.

Beyond traveling-wave phonons, we also proposed how Brillouin-induced Raman-like modes might be generated and observed at room temperature, potentially bridging the gap between conventional Raman spectroscopy and Brillouin acoustics. Although direct experimental realizations of these room temperature hybrid modes remain unobserved, the theoretical groundwork and device prototypes described here highlight the promise of such approaches for both fundamental and applied research. Ultimately, these efforts underscore the rich variety of phononic phenomena accessible when one leverages strong acousto-optic interactions in engineered media.

Looking ahead, one important direction will be to pursue net optomechanical cooling of fiber by minimizing or routing away unwanted Stokes heating contributions. Achieving such robust cooling could open pathways to ground-state phonon occupancy, enabling quantum acoustic memory and photon–phonon entan-

lement protocols. In tandem, refining the CoBS method to push detection limits and spatial resolution even further, perhaps by introducing additional  $n$ -pairs of pump and Stokes driving fields within small detunings to preserve coherent multi-wave driving of a single phonon mode with relaxed phase-matching allowances. Such a scheme would offer scattered power gains in high-gain parameter domains paired with modest interaction lengths. With the demonstrated and proposed advances described in this document, additional device architectures across the fields of quantum information, ultralow-noise photonics, and advanced sensing may be unlocked. In this way, the work presented herein lays the foundation for continued innovation at the intersection of photonics, phononics, and materials science resting on control of traveling-wave phonons at room temperature.





## Appendix A

### Supplementary Information for Chapter 2

#### A.1 Fabrication of CS<sub>2</sub>-Filled Liquid-Core Optical Fiber

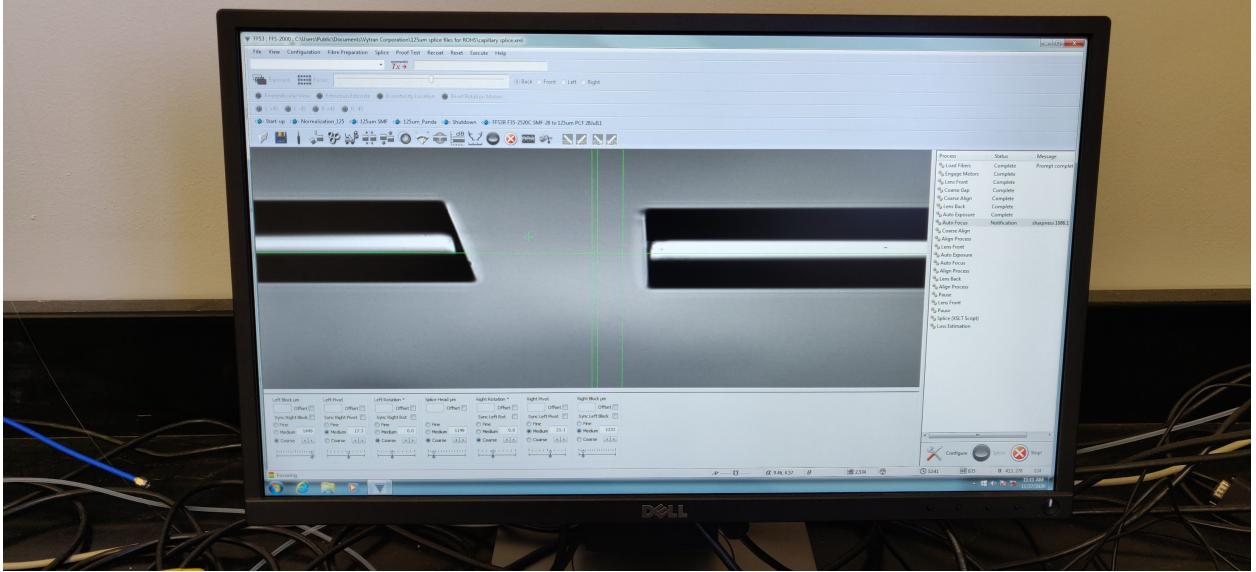
Fabrication of the liquid-core optical fiber (LCOF) followed the methods described by Kieu et al. (2012).

<sup>39</sup> Figure 2.2 in the main text illustrates the overall schematic of the LCOF, showing how single mode fiber 28 (SMF-28) and Ultra High Numerical Aperture 7 (UHNA7) fibers flank a hollow core segment. This section details the splicing, assembly, and filling processes in fabricating the CS<sub>2</sub>-filled LCOF.

##### A.1.1 Splicing

Before integrating the hollow-core fiber, a short length ( $\sim 0.5$  m) of Corning SMF-28 was stripped, flat-cleaved, and arc-spliced to 5–10 cm of UHNA7 fiber which was fully stripped of its coating to prevent up-and-down bending of the fiber end in subsequent horizontal alignment with the Vytran fusion splicer. A tapering effect was produced by repeating the arc four times in order to gradually narrow the SMF-28 core (9.2  $\mu\text{m}$  diameter) down to the smaller UHNA7 core (2.4  $\mu\text{m}$  diameter), improving coupling efficiency. Prior to and following the splice, optical transmission was monitored to ensure maximum transmission was preserved.

After the preliminary SMF-28–UHNA7 splice, the UHNA7 fiber was angle-cleaved to ensure a gap remained for CS<sub>2</sub> to be able to enter the hollow-core fiber after fusion-splicing the two fiber segments. An angle-cleaver with an adjustable torque and tension mechanism was used for this process. The angle-cleaved SMF-28-UHNA7 segment was then loaded into the Vytran fusion splicing system. The Vytran fusion splicing station and its vacuum pump were turned on, and the system’s software interface was opened. Approximately two minutes was allowed for the vacuum level to stabilize. The left fiber clamp block was pivoted fully to the left, and the UHNA7 fiber was placed so that its freshly cleaved tip was centered over the reference circle at the splice head. While the vacuum held the fiber in place, the system’s camera view was used to rotate the fiber such that the sharpest angle of the UHNA7’s end face was in the camera’s view. A small “flag” (a piece of unstripped fiber with a small tape handle on one end) was used to help the clamp grip

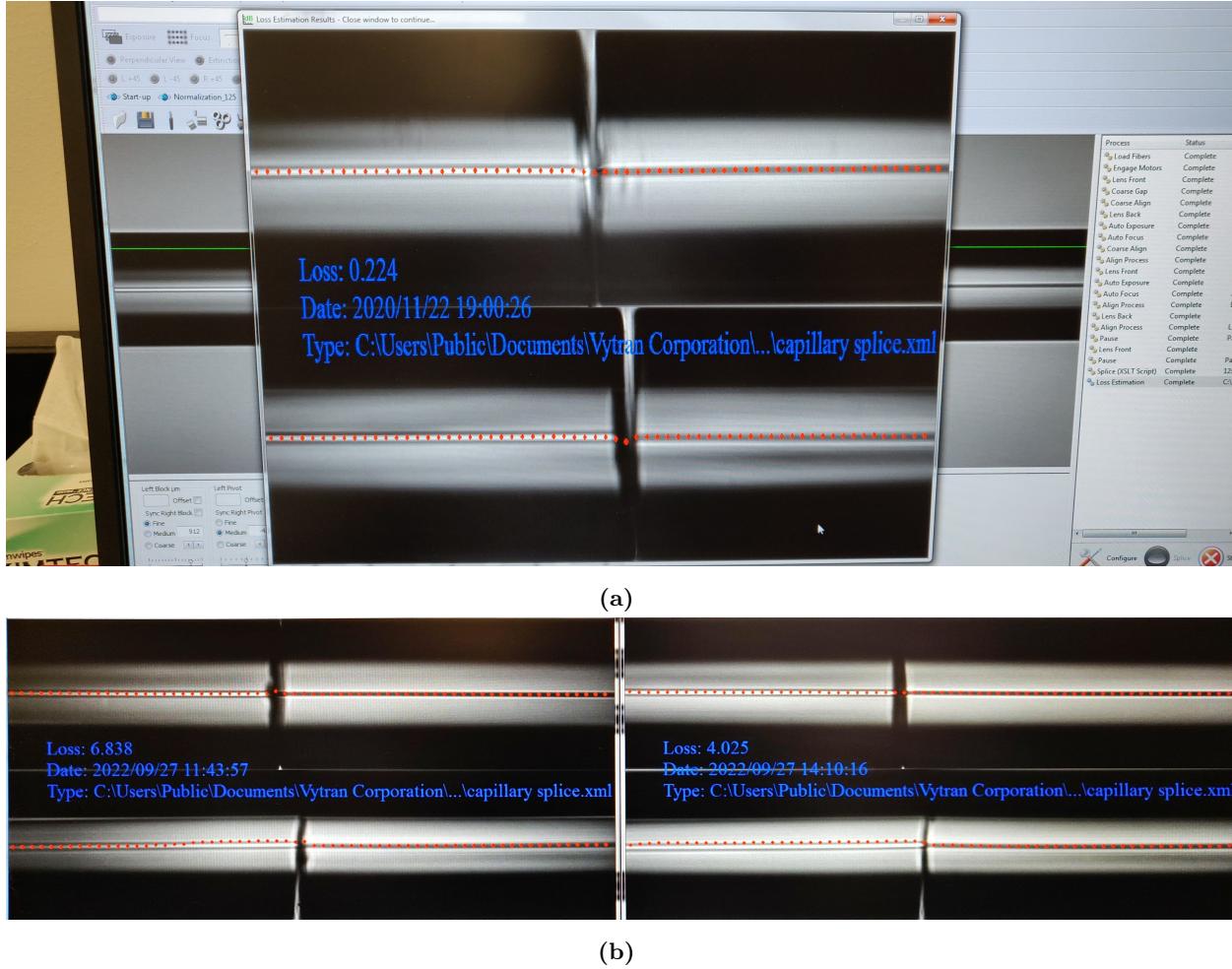


**Figure A.1:** Picture of Vytran software interface camera imaging system showing a microscope view of the two fiber ends pre-splice (Section A.1.1). The left angle-cleaved fiber is the UHNA7 fiber. The right flat-cleaved fiber is the hollow-core fiber. Subsequent alignment processes, first automatic then manual fine-tuning, align the fibers in xy space for optimal fusion-splicing and optical transmission once filled.

the bare UHNA7 fiber securely. The left block was pivoted back and forth until the UHNA7 tip was near the correct horizontal position. A folded piece of support paper and two boxes of Kimtech Kimwipes on the benchtop were used to prevent the fiber from drooping and to minimize the risk of breakage during handling (see Figure A.3).

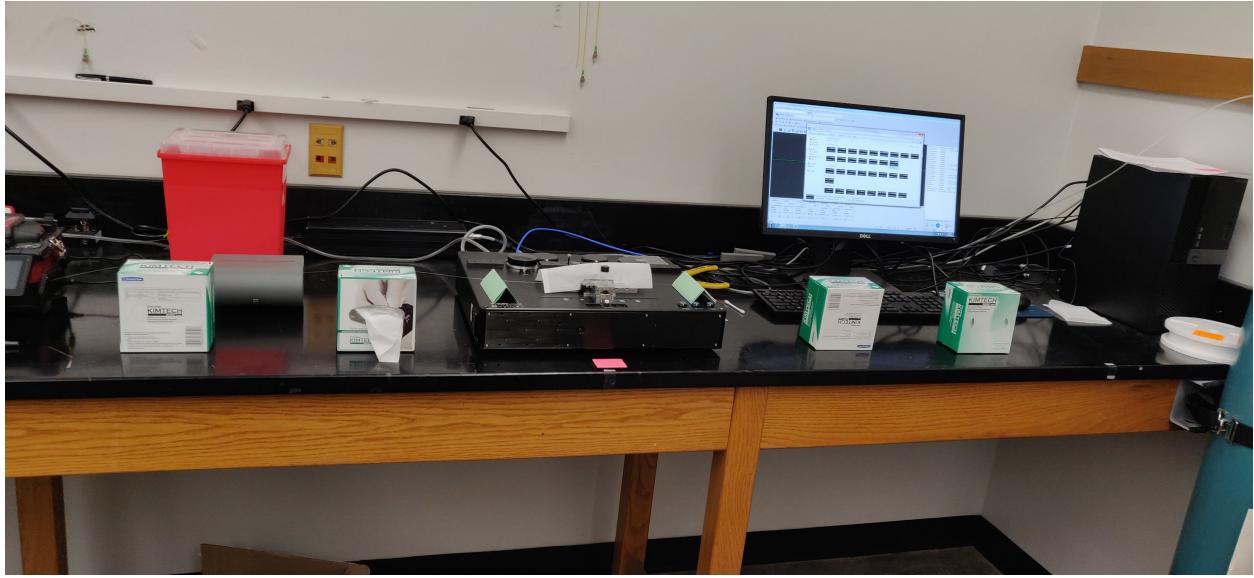
Once the UHNA7 fiber was properly angle-cleaved, the hollow-core fiber segment was prepared for splicing. The hollow-core fiber coating was dissolved with acetone and wiped away to expose the bare glass, which was then cleaved flat. The right clamp block was pivoted to its stop position, and the hollow-core fiber was placed so its end was centered over the splice head's circle, ensuring no physical contact with the UHNA7 fiber. As with the UHNA7, a flag of unstripped fiber was placed above the hollow-core fiber to facilitate clamping. The right clamp block was slowly pivoted toward the center until the fiber tips were close but still not touching. Like before, the fiber was supported with folded paper and Kimwipe boxes to prevent bending.

The next step was to form the final fusion splice between the UHNA7 fiber and the hollow-core segment within the fusion splicer. With both fibers supported and extending naturally from opposite blocks, the splicer's auto-alignment was initiated. The system brought the fibers within viewing distance so the operator could adjust vertical and horizontal positioning. Using the splicer's camera interface, the UHNA7 core was lined up with the hollow-core's central region. Small vertical and horizontal translations ensured good overlap. The left fiber (UHNA7 side) was carefully advanced until the tips touched, then retracted two clicks



**Figure A.2:** Example images of LCOF splices (Section A.1.1). Figure (a) shows an earlier example of a splice which featured a large visible wedge-shaped gap for liquid to enter the hollow-core fiber. While gaps of this size were sometimes successfully transferred onto a slide, further investigation showed gaps of this scale to reduce optical transmission through the splice. This fact became critical later for collecting the data for the pump-probe experiment. Figure (b) shows images of the two splices featured on the ultimate sample which was used to gather final data for publication.

for optimal separation during splicing. Precise final positioning of the fiber ends ensured that a small gap remained in the hollow core to accommodate liquid infiltration during the filling process. Simultaneously, this same fiber end positioning ensured the cladding regions were fused sufficiently to preserve splice integrity under the delicate handling steps that followed. An argon flow was introduced to shield the fusion region from contaminants, and the fusion filament heating was applied. Images were taken and saved for future review of the splice. If alignment or splicing was inadequate, the fibers were separated, re-cleaved if necessary, and the procedure repeated. After confirming a good splice, the argon was turned off and the splicer was shut down.



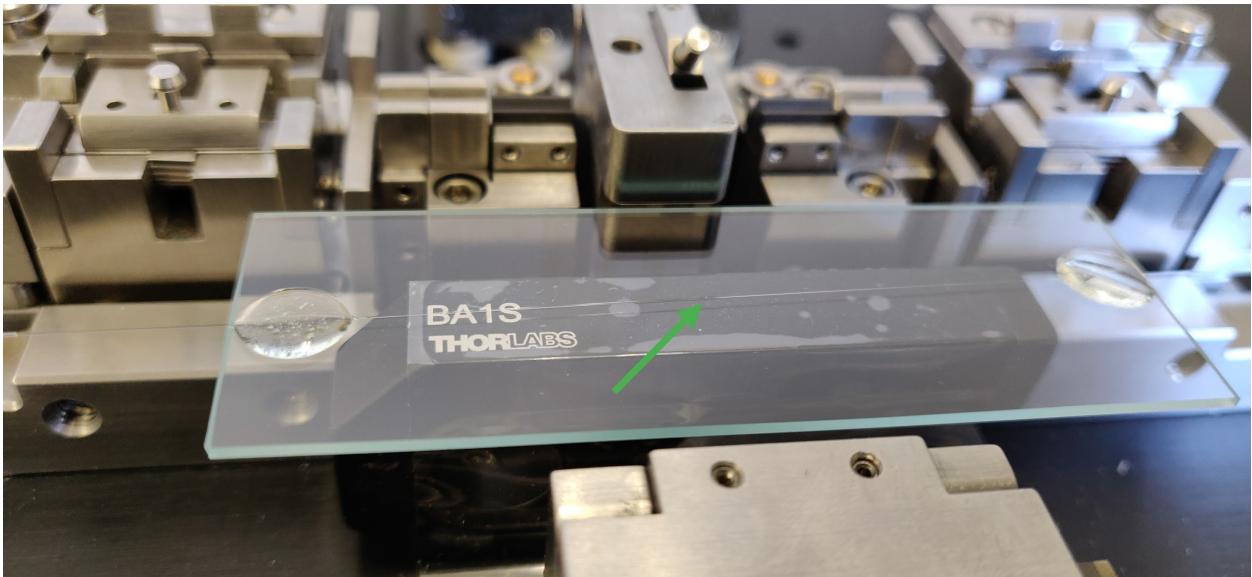
**Figure A.3:** Picture of both lengths of fiber braced by folded paper and two Kimwipe boxes on either side to prevent bending and flexing while transferring the delicate splice onto a glass slide (Section A.1.1).

### A.1.2 Reservoir Assembly

Because the final splice assembly must be encapsulated for protection, it was mounted on a clean microscope slide. The slide was scrubbed with soap and a toothbrush under running hot water, then thoroughly rinsed and dried with an air gun. Clean handling was crucial, so the slide was placed inside a folded Kimwipe for safekeeping until needed.

A small glass vial was used to form a protective enclosure over the splice region and allow liquid to fill the hollow core. Wearing gloves and safety glasses, a new glass vial was secured carefully by hand and cut at low rotational speed on a saw. The cut removed only the bottom portion of the vial so that the main body of the vial remained relatively long. The cut edge was smoothed and flattened with a file block, and two shallow, wide straddle-gaps were added on opposite sides of the new opening. These gaps would later accommodate the fiber on the slide. Finally, the vial was scrubbed with dish soap and a pipe cleaner, rinsed, dried with compressed air, and recapped until needed.

Upon completing the angle splice, the fiber assembly needed to be moved gently off the splicer onto the prepared microscope slide. With the splicer's vacuum pump still on to hold the splice securely, the magnetic latches clamping the fiber on either side of the splice to the translation/rotation stages were carefully lifted. Tweezers were used to remove any fiber flags, and wooden craft sticks helped lift the fibers from the block grooves to free the fiber from any stuck position within the grooves. The splicer's vacuum pump was switched off and the vacuum seal securing the fibers on either end of the splice was allowed to gradually release as air slowly equilibrated the pressure differential. The splice head and camera assembly remained overtop the

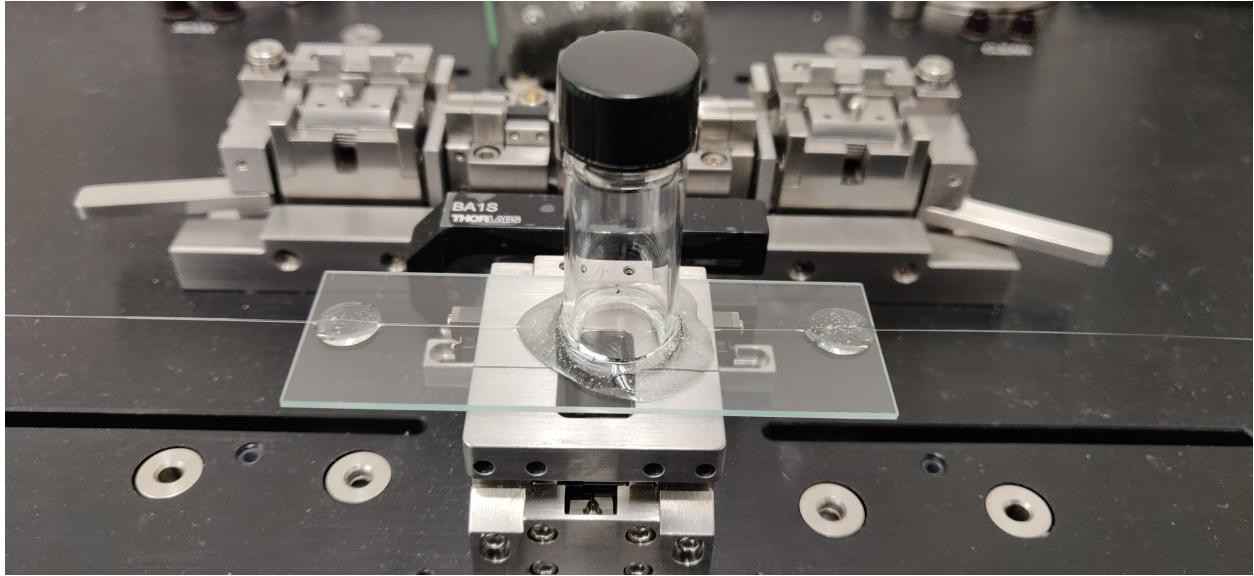


**Figure A.4:** Picture of splice successfully transferred onto a glass slide and tacked on either side with drops of epoxy (Section A.1.1). An arrow points to the location of the critical splice between the angle-cleaved UHNA7 fiber and the flat-cleaved hollow-core fiber.

splice for monitoring the camera feed on the computer screen. After approximately five minutes the camera feed would show the splice quickly move out of focus, indicating that the entire length of fiber was free and ready to be handled. With the splice head lifted, the slide was positioned directly in front of the splice and the fiber was transferred carefully in a smooth motion. Two small drops of epoxy were placed on each side of the splice region to tack the fiber down. This was allowed to cure for at least five minutes.

The splice region was enclosed in a glass vial to protect the hollow-core's interior and to facilitate later filling with liquid. The prepared vial was placed over the splice region by sliding one straddle-gap around the fiber first, then tilting it so the second gap aligned. Gloves were worn to prevent transferring skin oils onto the vial. A fresh mixture of two-part epoxy was prepared in roughly equal proportions and thoroughly mixed for 10–15 seconds to ensure a uniform bond. Generous epoxy was applied around the vial's perimeter where it contacted the slide. Care was taken to avoid epoxy wicking into the splice itself. The assembly was then left undisturbed for at least five minutes to cure.

Once the epoxy had set, the slide was labeled with the date, splice reference, and relevant notes using a marker. For long-term handling, the slide and its fibers were taped to a poster board to facilitate transport and prevent accidental breakage from jostling. This procedure ensured a robust, low-loss transition from SMF-28 to UHNA7 to the hollow-core fiber. The careful cleaning steps, controlled splicing environment with argon shielding, and meticulous handling minimized the risk of fiber breakage and guaranteed a clean optical interface. Encapsulating the splice with a modified glass vial on a microscope slide allowed easy



**Figure A.5:** Picture of a complete splice assembly (Section A.1.2). The hardened epoxy around the base of the vial securely holds the cut and notched vial onto the slide, forming a sealed reservoir around the critical splice. The reservoir is later filled with liquid CS<sub>2</sub> by easy removal of the screw-off cap in order to submerge the critical splice and begin the filling process of the hollow-core fiber via capillary action.

manipulation of the hollow core's environment, which was crucial for subsequent liquid filling and optical characterization experiments.

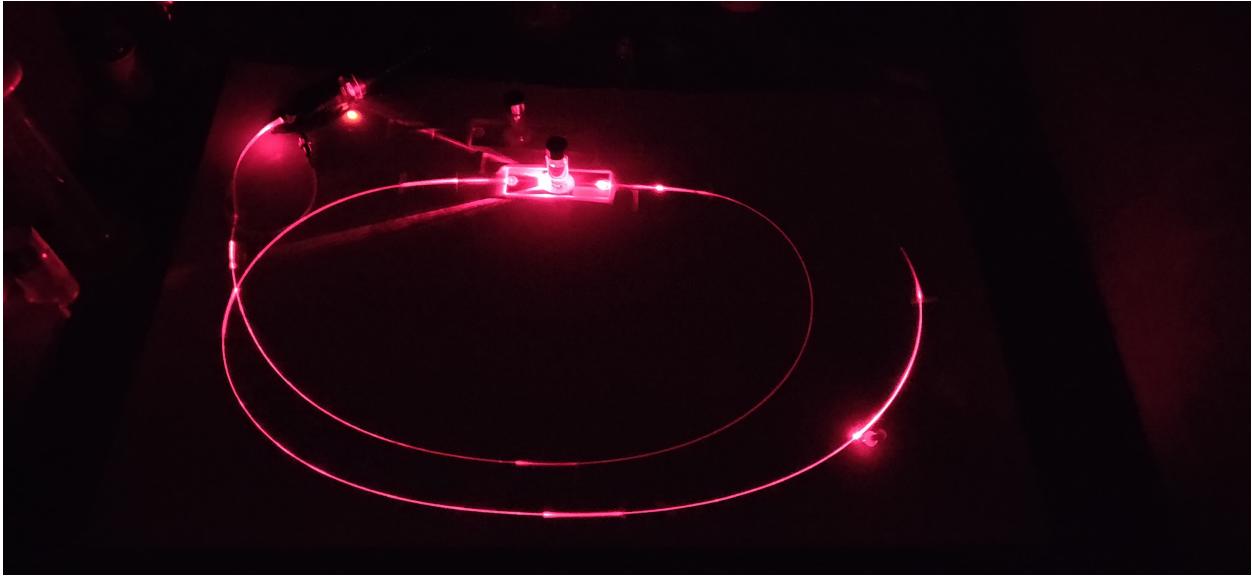
Finally, this entire procedure was repeated on the opposite side of the hollow-core fiber to complete the LCOF assembly. Each splice was carefully optimized to be robust enough to guide light but also unfused enough to allow CS<sub>2</sub> to enter the hollow core via capillary action.

### A.1.3 Filling with Carbon Disulfide

Once both ends of the hollow-core fiber were successfully spliced to their respective fiber pigtails, the next critical step involved filling the hollow core with carbon disulfide. Because CS<sub>2</sub> is highly volatile and poses health risks if inhaled, all operations were carried out in a fume hood with proper protective equipment.

The prepared LCOF sample, securely taped to a poster board, was transferred to the fume hood. A small red laser source was connected to the input pigtail; this red beam served as an in situ indicator of the fluid-filling front. The vial on the input side was uncapped, and the vial on the opposite side was loosened to prevent pressure buildup within the fiber. This arrangement ensured that as CS<sub>2</sub> entered the hollow core, displaced air could escape through the opposite vial, allowing continuous capillary flow.

To deliver the CS<sub>2</sub>, a syringe was first used to extract an adequate volume from a sealed supply bottle. The needle tip was then removed and replaced with a  $\mu\text{m}$ -level particulate filter, thereby minimizing the

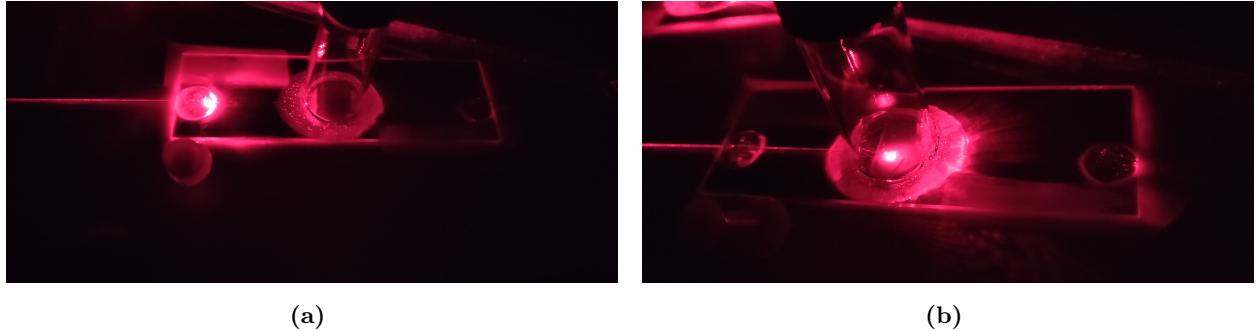


**Figure A.6:** Picture of a complete sample under the fume hood with lights dimmed and red laser light injected into the end of the sample for monitoring (Section A.1.3). The red light is partially guided along the length of fiber which has filled with liquid CS<sub>2</sub> and a thumb tack marks the progress of the CS<sub>2</sub>-air interface.

introduction of debris that could obstruct the hollow core. By gently angling the syringe, CS<sub>2</sub> was dispensed along the interior wall of the vial rather than dripping directly onto the delicate splice region. This careful approach reduced mechanical shocks, which could otherwise fracture or misalign the newly formed splice. Once the vial was filled, it was recapped promptly to prevent evaporation.

If a given splice was imperfect (fully sealed at the fiber core rather than partially open) it prevented CS<sub>2</sub> from flowing in. Under these circumstances, no visible progression of the fluid meniscus would appear in the red laser beam path, confirming an unsuccessful splice. In contrast, if the splice was partially open, capillary action would begin immediately, typically drawing CS<sub>2</sub> several centimeters into the hollow core within seconds. The interface between the CS<sub>2</sub> and the air still occupying the rest of the fiber showed up as a faintly scattering “dot” in the path of the red beam. By darkening the room, this dot could be easily observed and tracked. Once the fiber was fully filled, the second vial was also filled with filtered CS<sub>2</sub>, then capped. If the second reservoir was filled prematurely, an air bubble would be trapped in the final short length of hollow-core fiber and the resulting transmission through the full length of the LCOF would be reduced to nearly 0%.

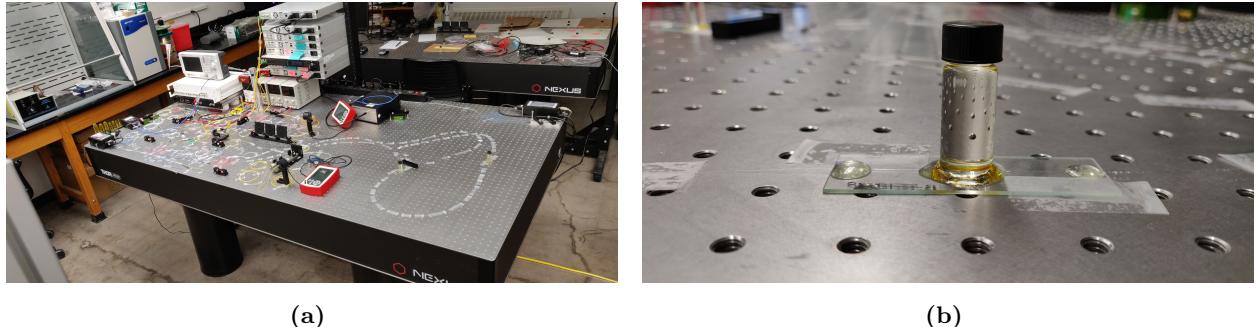
Figure A.6 shows a picture of a sample in the process of filling with CS<sub>2</sub>. Progress is marked by a thumbtack next to the CS<sub>2</sub>-air interface. With the lights turned off, the dot of scattered light at this interface was a reliable visible indicator. Figures A.7a and A.7b show images of a sample that has nearly finished and fully finished filling, respectively. The filling state captured by Figure A.7a indicates a likely



(a)

(b)

**Figure A.7:** Images of an LCOF sample in the filling process (Section A.1.3). Figure (a) shows the  $\text{CS}_2$ -air interface a few centimeters from the end of the length hollow-core fiber, indicated by the red dot of scattered light just underneath the epoxy tack. From this position, the meniscus will typically reach the exit splice in approximately 4 hours. Figure (b) shows evidence of a fully-filled LCOF sample, indicated by the red dot of scattered light having reached the exit splice. If the pictured reservoir were to be filled prematurely, an air bubble would be locked in, reducing full transmission through the sample to nearly 0%.



(a)

(b)

**Figure A.8:** Images of a fully finished  $\text{CS}_2$ -LCOF sample (Section A.1.4). Figure (a) shows a sample secured with tape to an optical table and integrated into an optical setup. Liberal use of tape ensured the safety of the sample as well as reduced vibrations and minimized changes in the polarization of light travelling through the sample. Figure (b) shows one splice assembly secured directly to the optical table via tape. Transfer of all parts of the sample from the poster board directly onto the optical table proved critical for eliminating noise and polarization drift issues with the pump-probe experiment.

time to finish filling of approximately 4 hours.

#### A.1.4 Typical Optical Performance

Optical transmission at each splice typically ranged from 5–25%. The final LCOF used to gather the experimental results published in the paper associated with this chapter achieved full throughput transmission of about 17%. This suggests each splice individually offered an average of approximately 41% transmission of the incident light, assuming each splice provided equal transmission. In the demonstration of optomechanical cooling of traveling-wave phonons in this LCOF, the light collected at the detector is backscattered within the sample and thus passes back through the same splice that the incident pump light enters through.

This round-trip backscattered LCOF transmission was treated as approximately equal to the full throughput transmission monitored to be exiting the sample, as light passes through two splices of assumed equal transmission in each case.

Figure A.8 shows two images of a complete LCOF secured to an optical table with Scotch tape and integrated into an optical setup. In later stages of experimentation it was discovered that a large amount of noise and changes in polarization of the light passing through the sample would be incurred if the LCOF were left attached to the poster board used in fabrication and transport. Despite taping all edges of the poster board securely down onto the optical table, minor flexing of the board from small temperature and humidity changes or air drafts in the room caused sufficient instability in the signal to delay successful data collection for the pump-probe experiment (Section 2.4.2).

Over periods of days, some evaporation was inevitable; replenishing the vials every two to three days extended the operational lifetime of the sample. Despite occasional epoxy degradation at the splice site, which might reduce forward transmission, the backscattering experiments remained unaffected except for a loss of forward transmission monitoring. In most cases, weeks-long sample preservation and continued experimentation was feasible with proper maintenance.

## A.2 Improvements in Yield and Efficiency of LCOF Samples

A key refinement that greatly improved sample fabrication success rates was the adoption of extremely careful post-splice handling. Previously, standard procedure was to lift the newly spliced fiber assembly without ensuring that each side was completely free of the splicer's grooves. This led to abrupt bending or sudden snapping out of the clamp grooves, which almost invariably broke delicate splices that were only partially fused. The implementation of several measures helped to avoid this failure mode. First, folded-paper sawhorses and two small Kimwipe boxes on each side of the splice were placed to support the fibers and reduce vibration or flexing. Before switching off the vacuum seal, the clamp latches were gently lifted and small wooden sticks were used to ease the fibers out of the translation/rotation block grooves, ensuring there was no latent twisting or bending. Throughout these steps, the fibers were kept as close as possible to their natural resting position, minimizing stress that would be transferred to the just-completed splice. This careful approach not only reduced the chance of breakage but also allowed for the feasibility of splices that remained sufficiently "open" for  $\text{CS}_2$  to enter the hollow core, substantially increasing the proportion of successfully filled samples.

In using the slow-rotation saw to cut and form notches in the glass vial, simple improvements in notch geometry greatly reduced breakage of splices while placing the vial on the slide overtop the delicate splice,

and straddling the fibers on either side. Cutting notches to be ~1 cm (10-20 times the width of the fiber) reduced the risk of inadvertently contacting the fibers and breaking the delicate splice during vial placement. Additionally, cutting the notches to be shallow (approximately 5 times the width of the fiber) prevented epoxy from running through the notch and sealing the splice. Previous standard procedure were opposite to this geometry as a natural result of the narrow width of the saw blade and the depth to which it could easily be allowed to cut.

A key insight in the filling procedure was learned by performing a long observation of the filling process for one sample. Previous standard procedure was to stop monitoring after 8 or 12 hours, assuming the fill process had ceased or completed but subsequently reversed if the fluid had not reached the far end by that time. Through extended trials, it was discovered that in successful splices, the fill front progresses at a nonlinear, steadily decreasing rate, and that a one-meter fiber segment might take more than 24 hours to completely fill. In one particularly instructive case, continuous monitoring for 27 hours without leaving the room as a sample filled confirmed that the CS<sub>2</sub> front never reversed; it merely advanced extremely slowly in the final length to reach the other end. Alarms were set for inspection every 90 minutes throughout the night in the final hours of monitoring the sample under the fume hood. Having observed and documented this behavior, the standard practice became allowing the sample to sit undisturbed for at least a full day or more, ensuring the fiber was completely saturated before concluding success or failure. This observation effort and resulting insight halted the regular production of significant waste of both material and time in the fabrication process of LCOF samples.

A final insight that improved the yield of successfully filled samples and mitigation of wasted materials was selecting the less certain end of a prepared sample for filling with CS<sub>2</sub>. If that splice was sealed, only that end would need to be re-fabricated. Collectively, these simple method improvements in sample fabrication led to significantly shorter fabrication times and dramatically reduced material waste. Hollow-core fibers, which cost on the order of ten dollar per meter, were previously scrapped in large quantities when deemed “dead.” By implementing patient monitoring, careful handling, and mindful filling procedures each length of fiber was used more efficiently, saving significant time and expense while producing more effective samples.

### A.3 Tabulated Fit and Uncertainty Values Derived from the Observed Data

#### A.3.1 Cooling Experiment A Tabulated Values

**Table A.1:** Measured anti-Stokes parameters for Experiment A. Here  $P_P$  is the nominal pump power, measured via a 1% tap just prior to launching into the LCOF, and  $P_{\text{Intra}}$  is the power actually guided within the fiber. Amplitude, Linewidth, Center, and Offset are the peak spectral density, FWHM linewidth, center frequency, and vertical baseline offset, respectively, obtained from a Lorentzian fit of the data. Uncertainties are  $1\sigma$ .

$P_P$ (mW)	$P_{\text{Intra}}$ (mW)	Amplitude ( $\mu\text{V}$ )	Linewidth (MHz)	Center (GHz)	Offset ( $\mu\text{V}$ )
10.00	4.12	$3.015 \pm 0.004$	$97.3 \pm 0.1$	$2.2690 \pm 0.0001$	$-0.006 \pm 0.002$
30.00	12.37	$8.747 \pm 0.006$	$99.10 \pm 0.05$	2.2690	$0.015 \pm 0.002$
50.00	20.62	$14.22 \pm 0.01$	$99.79 \pm 0.04$	2.2690	$0.032 \pm 0.002$
70.00	28.86	$19.12 \pm 0.02$	$101.15 \pm 0.05$	2.2690	$0.015 \pm 0.003$
90.00	37.11	$24.92 \pm 0.01$	$100.91 \pm 0.03$	2.2680	$0.039 \pm 0.003$
110.00	45.35	$31.27 \pm 0.02$	$102.33 \pm 0.03$	2.2690	$0.029 \pm 0.003$
130.00	53.60	$36.06 \pm 0.02$	$102.45 \pm 0.03$	2.2690	$0.016 \pm 0.003$
150.00	61.85	$41.59 \pm 0.02$	$102.55 \pm 0.03$	2.2690	$0.118 \pm 0.003$
170.00	70.09	$46.78 \pm 0.02$	$103.27 \pm 0.03$	2.2690	$0.086 \pm 0.003$
190.00	78.34	$51.86 \pm 0.03$	$105.02 \pm 0.03$	2.2690	$-0.071 \pm 0.004$
210.00	86.59	$57.04 \pm 0.02$	$105.13 \pm 0.03$	2.2690	$0.058 \pm 0.004$
230.00	94.83	$61.42 \pm 0.03$	$105.63 \pm 0.03$	2.2690	$0.093 \pm 0.004$
250.00	103.08	$66.64 \pm 0.04$	$105.83 \pm 0.03$	2.2690	$0.148 \pm 0.005$
270.00	111.32	$70.63 \pm 0.04$	$106.42 \pm 0.04$	2.2690	$0.205 \pm 0.006$
290.00	119.57	$74.68 \pm 0.04$	$106.32 \pm 0.03$	2.2690	$0.314 \pm 0.006$

**Table A.2:** Measured Stokes parameters for Experiment A, in the same format as Table A.1.

$P_P$ (mW)	$P_{\text{Intra}}$ (mW)	Amplitude ( $\mu\text{V}$ )	Linewidth (MHz)	Center (GHz)	Offset ( $\mu\text{V}$ )
10.00	4.12	$3.203 \pm 0.005$	$97.3 \pm 0.1$	$2.2690 \pm 0.0001$	$0.020 \pm 0.002$
30.00	12.37	$9.686 \pm 0.006$	$96.82 \pm 0.05$	2.2680	$0.046 \pm 0.002$
50.00	20.62	$15.874 \pm 0.009$	$96.84 \pm 0.04$	2.2680	$0.073 \pm 0.002$
70.00	28.86	$21.87 \pm 0.01$	$96.94 \pm 0.04$	2.2680	$0.066 \pm 0.002$
90.00	37.11	$28.47 \pm 0.01$	$96.40 \pm 0.03$	2.2680	$0.102 \pm 0.002$
110.00	45.35	$37.46 \pm 0.02$	$96.20 \pm 0.03$	2.2680	$0.084 \pm 0.003$
130.00	53.60	$43.36 \pm 0.03$	$96.37 \pm 0.03$	2.2680	$0.023 \pm 0.003$
150.00	61.85	$50.96 \pm 0.02$	$95.04 \pm 0.02$	2.2680	$0.174 \pm 0.003$
170.00	70.09	$57.94 \pm 0.03$	$95.61 \pm 0.02$	2.2680	$0.058 \pm 0.003$
190.00	78.34	$65.46 \pm 0.04$	$95.43 \pm 0.03$	2.2680	$0.029 \pm 0.004$
210.00	86.59	$73.49 \pm 0.04$	$94.59 \pm 0.02$	2.2680	$0.166 \pm 0.003$
230.00	94.83	$81.56 \pm 0.04$	$94.10 \pm 0.02$	2.2680	$0.170 \pm 0.004$
250.00	103.08	$90.64 \pm 0.06$	$92.98 \pm 0.03$	2.2680	$0.272 \pm 0.005$
270.00	111.32	$98.22 \pm 0.04$	$92.67 \pm 0.02$	2.2680	$0.316 \pm 0.004$
290.00	119.57	$107.04 \pm 0.05$	$91.04 \pm 0.02$	2.2680	$0.557 \pm 0.006$

### A.3.2 Cooling Experiment B Tabulated Values

**Table A.3:** Measured anti-Stokes parameters for Experiment B. Here  $P_P$  is the nominal pump power, measured via a 1% tap just prior to launching into the LCOF,  $P_{\text{Intra}}$  is the power actually guided within the fiber, and  $P_{\text{Pr}}$  is the probe power, also measured just prior to launching into the LCOF. Amplitude, Linewidth, Center, and Offset are the peak spectral density, FWHM linewidth, center frequency, and vertical baseline offset, respectively, obtained from a Lorentzian fit of the data. Uncertainties are  $1\sigma$ .

<b><math>P_P</math></b> (mW)	<b><math>P_{\text{Intra}}</math></b> (mW)	<b><math>P_{\text{Pr}}</math></b> (mW)	<b>Amplitude</b> ( $\mu\text{V}$ )	<b>Linewidth</b> (MHz)	<b>Center</b> (GHz)	<b>Offset</b> ( $\mu\text{V}$ )
0.00	0.00	10.00	$1.849 \pm 0.002$	$96.6 \pm 0.2$	2.2700	$0.0230 \pm 0.0009$
55.00	22.68	10.00	$1.640 \pm 0.007$	$104.0 \pm 0.5$	$2.2710 \pm 0.0001$	$0.019 \pm 0.001$
110.00	45.35	10.00	$1.53 \pm 0.01$	$106.9 \pm 0.8$	$2.2710 \pm 0.0002$	$0.020 \pm 0.002$
165.00	68.03	10.00	$1.43 \pm 0.01$	$110 \pm 1$	$2.2700 \pm 0.0003$	$0.028 \pm 0.002$

## Appendix B

### Supplementary Information for Chapter 3

#### B.1 Coupled-Wave Equations

Here we derive the coupled wave equations that describe Coherently stimulated Brillouin Scattering (CoBS) involving a pump, Stokes, probe, and backscattered optical field given respectively by

$$\tilde{E}_P(z, t) = A_P e^{i(k_P z - \omega_P t)} + c.c., \quad (\text{B.1})$$

$$\tilde{E}_S(z, t) = A_S e^{i(-k_S z - \omega_S t)} + c.c., \quad (\text{B.2})$$

$$\tilde{E}_{Pr}(z, t) = A_{Pr} e^{i(k_{Pr} z - \omega_{Pr} t)} + c.c., \quad (\text{B.3})$$

$$\tilde{E}_{Sig}(z, t) = A_{Sig} e^{i(-k_{Sig} z - \omega_{Sig} t)} + c.c., \quad (\text{B.4})$$

and a common acoustic field given by

$$\tilde{\rho}(z, t) = \rho_0 + \rho(z, t) e^{i(qz - \Omega t)} + c.c., \quad (\text{B.5})$$

where  $\Omega = \omega_P - \omega_S$  and  $q = k_P - k_S \approx 2k_P$ .

### B.1.1 Acoustic Field

As in the case of SBS<sup>13</sup>, we start by assuming that the material obeys the acoustic wave equation,

$$\frac{\partial^2 \tilde{\rho}}{\partial t^2} - \Gamma' \nabla^2 \frac{\partial \tilde{\rho}}{\partial t} - v_s^2 \nabla^2 \tilde{\rho} = \nabla \cdot \vec{f}, \quad (\text{B.6})$$

where  $v_s$  is the sound speed in the material and  $\Gamma'$  is a damping parameter given by

$$\Gamma' = \frac{1}{\rho} \left[ \frac{4}{3} \eta_s + \eta_b + \frac{\kappa}{C_p} (\gamma - 1) \right], \quad (\text{B.7})$$

where  $\eta_s$  and  $\eta_b$  are the shear and bulk viscosity coefficients of the material, respectively. The source term on the right side of Equation B.6 is the divergence of the electrostrictive force:

$$\vec{f} = \nabla p_{st} = \nabla \cdot \left[ -\frac{1}{2} \epsilon_0 \gamma_e \left( \langle \tilde{E}_P \cdot \tilde{E}_S \rangle + \langle \tilde{E}_{Pr} \cdot \tilde{E}_{Sig} \rangle \right) \right], \quad (\text{B.8})$$

which yields, assuming the acoustic wave varies slowly in time,

$$\nabla \cdot \vec{f} = \epsilon_0 \gamma_e q^2 (A_P A_S^* + A_{Pr} A_{Sig}^*) e^{i \Delta k z}, \quad (\text{B.9})$$

where  $\Delta k = (k_{Pr} - k_{Sig}) - (k_P - k_S)$  is the phase mismatch between the four optical fields. Only two electrostrictive terms survive after accounting for the orthogonal polarization of the pump and Stokes fields with respect to that of the probe and backscattered signal. Inserting this electrostrictive force term (Equation B.9) and the acoustic field (Equation B.5) into the acoustic wave equation (Equation B.6), we find

$$-2i\Omega \frac{\partial \rho}{\partial t} - \Gamma' 2iq^2 \Omega \rho - 2iqv_s^2 \frac{\partial \rho}{\partial z} = \epsilon_0 \gamma_e q^2 (A_P A_S^* + A_{Pr} A_{Sig}^*) e^{i \Delta k z}, \quad (\text{B.10})$$

which can be restated in terms of the Brillouin linewidth,  $\Gamma_B = q^2 \Gamma'$ , as

$$-2i\Omega \frac{\partial \rho}{\partial t} - 2i\Omega \Gamma_B \rho - 2iqv_s^2 \frac{\partial \rho}{\partial z} = \epsilon_0 \gamma_e q^2 (A_P A_S^* + A_{Pr} A_{Sig}^*) e^{i \Delta k z}. \quad (\text{B.11})$$

Given the phonon dispersion relations  $\Omega_B = |q_B|v_s$  and  $\Omega^2 = q^2(v_s^2 - i\Omega\Gamma')$ , Equation B.11 can be rewritten as

$$-2i\Omega \frac{\partial \rho}{\partial t} + (\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B) \rho - 2iqv_s^2 \frac{\partial \rho}{\partial z} = \epsilon_0 \gamma_e q^2 (A_P A_S^* + A_{Pr} A_{Sig}^*) e^{i\Delta kz}. \quad (\text{B.12})$$

We take the common assumption that the phonon propagation distance is small compared to the distance over which the source term varies significantly, which allows the spatial derivative term in Equation B.12 to be dropped. We further assume steady-state conditions such that the time derivative term also vanishes, leaving

$$(\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B) \rho = \epsilon_0 \gamma_e q^2 (A_P A_S^* + A_{Pr} A_{Sig}^*) e^{i\Delta kz}. \quad (\text{B.13})$$

We thus find the acoustic field amplitude to be

$$\rho(z, t) = \epsilon_0 \gamma_e q^2 \frac{(A_P A_S^* + A_{Pr} A_{Sig}^*) e^{i\Delta kz}}{\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B}. \quad (\text{B.14})$$

### B.1.2 Optical Fields

We now turn to the spatial evolution of the optical fields, described by the wave equation,

$$\frac{\partial^2 \tilde{E}_i}{\partial z^2} - \frac{n^2}{c^2} \frac{\partial^2 \tilde{E}_i}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2 \tilde{P}_i}{\partial t^2}, \quad (\text{B.15})$$

where  $i$  denotes the four optical fields, namely: pump, Stokes, probe, and the backscattered signal. The total nonlinear polarization that gives rise to the source term in the wave Equation is given by

$$\tilde{P} = \epsilon_0 \Delta \chi \tilde{E} = \epsilon_0 \Delta \epsilon \tilde{E} = \epsilon_0 \rho^{-1} \gamma_e \tilde{\rho} \tilde{E}. \quad (\text{B.16})$$

The parts of  $\tilde{P}$  that can act as phase-matched source terms for the optical fields are

$$\tilde{P}_P = p_P e^{i(k_P z - \omega_P t)} + c.c. = \frac{1}{2} \epsilon_0 \rho_0^{-1} \gamma_e \rho A_S e^{i(k_P z - \omega_P t)}, \quad (\text{B.17})$$

$$\tilde{P}_S = p_S e^{i(-k_S z - \omega_S t)} + c.c. = \frac{1}{2} \epsilon_0 \rho_0^{-1} \gamma_e \rho^* A_P e^{i(-k_S z - \omega_S t)}, \quad (\text{B.18})$$

$$\tilde{P}_{\text{Pr}} = p_{\text{Pr}} e^{i(k_{\text{Pr}} z - \omega_{\text{Pr}} t)} + c.c. = \frac{1}{2} \epsilon_0 \rho_0^{-1} \gamma_e \rho A_{\text{Sig}} e^{i(k_{\text{Pr}} z - \omega_{\text{Pr}} t)} e^{i\Delta kz}, \quad (\text{B.19})$$

$$\tilde{P}_{\text{Sig}} = p_{\text{Sig}} e^{i(-k_{\text{Sig}} z - \omega_{\text{Sig}} t)} + c.c. = \frac{1}{2} \epsilon_0 \rho_0^{-1} \gamma_e \rho^* A_{\text{Pr}} e^{i(-k_{\text{Sig}} z - \omega_{\text{Sig}} t)} e^{-i\Delta kz}. \quad (\text{B.20})$$

Inserting the optical fields (Equations B.1-B.4) and phase-matched source terms (Equations B.17-B.20) into Equation B.15, we obtain

$$\frac{\partial A_P}{\partial z} + \frac{n}{c} \frac{\partial A_P}{\partial t} = \frac{i\omega_P \gamma_e}{2nc\rho_0} \rho A_S, \quad (\text{B.21})$$

$$-\frac{\partial A_S}{\partial z} + \frac{n}{c} \frac{\partial A_S}{\partial t} = \frac{i\omega_S \gamma_e}{2nc\rho_0} \rho^* A_P, \quad (\text{B.22})$$

$$\frac{\partial A_{\text{Pr}}}{\partial z} + \frac{n}{c} \frac{\partial A_{\text{Pr}}}{\partial t} = \frac{i\omega_{\text{Pr}} \gamma_e}{2nc\rho_0} \rho A_{\text{Sig}}, \quad (\text{B.23})$$

$$-\frac{\partial A_{\text{Sig}}}{\partial z} + \frac{n}{c} \frac{\partial A_{\text{Sig}}}{\partial t} = \frac{i\omega_{\text{Sig}} \gamma_e}{2nc\rho_0} \rho^* A_{\text{Pr}}, \quad (\text{B.24})$$

We again assume steady-state conditions, allowing the time derivative term to be dropped. Plugging in the acoustic field amplitude (Equation B.14), we arrive at the coupled-amplitude wave equations for the optical fields,

$$\frac{\partial A_P}{\partial z} = \frac{i\epsilon_0 \omega_P q^2 \gamma_e^2}{2nc\rho_0} \frac{(A_P |A_S|^2 + A_{\text{Pr}} A_{\text{Sig}}^* A_S) e^{i\Delta kz}}{\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B}, \quad (\text{B.25})$$

$$\frac{\partial A_S}{\partial z} = -\frac{i\epsilon_0\omega_S q^2 \gamma_e^2}{2nc\rho_0} \frac{(|A_P|^2 A_S^* + A_{Pr} A_{Sig}^* A_P) e^{-i\Delta kz}}{\Omega_B^2 - \Omega^2 + i\Omega\Gamma_B}, \quad (\text{B.26})$$

$$\frac{\partial A_{Pr}}{\partial z} = \frac{i\epsilon_0\omega_{Pr} q^2 \gamma_e^2}{2nc\rho_0} \frac{(A_P A_S^* A_{Sig} + A_{Pr} |A_{Sig}|^2) e^{i\Delta kz}}{\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B}, \quad (\text{B.27})$$

$$\frac{\partial A_{Sig}}{\partial z} = -\frac{i\epsilon_0\omega_{Sig} q^2 \gamma_e^2}{2nc\rho_0} \frac{(A_P A_S^* A_{Pr} + |A_{Pr}|^2 A_{Sig}^*) e^{-i\Delta kz}}{\Omega_B^2 - \Omega^2 + i\Omega\Gamma_B}. \quad (\text{B.28})$$

We drop the very small signal amplitude terms on the right side of Equations B.25-B.28 and integrate each along the effective length to get the amplitudes,

$$A_P = \frac{i\epsilon_0\omega_P q^2 \gamma_e^2}{2nc\rho_0} \frac{|A_P|^2}{\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B} \frac{e^{i\Delta k L} - 1}{i\Delta k}, \quad (\text{B.29})$$

$$A_S = -\frac{i\epsilon_0\omega_S q^2 \gamma_e^2}{2nc\rho_0} \frac{|A_P|^2 A_S^*}{\Omega_B^2 - \Omega^2 + i\Omega\Gamma_B} \frac{e^{-i\Delta k L} - 1}{-i\Delta k}, \quad (\text{B.30})$$

$$A_{Pr} = \frac{i\epsilon_0\omega_{Pr} q^2 \gamma_e^2}{2nc\rho_0} \frac{A_P A_S^* A_{Sig}}{\Omega_B^2 - \Omega^2 - i\Omega\Gamma_B} \frac{e^{i\Delta k L} - 1}{i\Delta k}, \quad (\text{B.31})$$

$$A_{Sig} = -\frac{i\epsilon_0\omega_{Sig} q^2 \gamma_e^2}{2nc\rho_0} \frac{A_P A_S^* A_{Pr}}{\Omega_B^2 - \Omega^2 + i\Omega\Gamma_B} \frac{e^{-i\Delta k L} - 1}{-i\Delta k}. \quad (\text{B.32})$$

We focus on the signal amplitude given by Equation B.32, noting that close to resonance, the denominator of the middle term containing  $\Omega$  can be approximated as,

$$\Omega_B^2 - \Omega^2 + i\Omega\Gamma_B \approx \Omega_B(\Omega - \Omega_B + i\Gamma_B), \quad (\text{B.33})$$

giving

$$A_{\text{Sig}} = -\frac{i\epsilon_0\omega_{\text{Sig}}q^2\gamma_e^2}{2nc\rho_0} \frac{A_P A_S^* A_{\text{Pr}}}{\Omega_B(\Omega - \Omega_B + i\Gamma_B)} \frac{e^{-i\Delta k L} - 1}{-i\Delta k}, \quad (\text{B.34})$$

and in fact on resonance the expression reduces to

$$A_{\text{Sig}} = -\frac{i\epsilon_0\omega_{\text{Sig}}q^2\gamma_e^2}{2nc\rho_0} \frac{A_P A_S^* A_{\text{Pr}}}{\Omega_B i\Gamma_B} \frac{e^{-i\Delta k L} - 1}{-i\Delta k}. \quad (\text{B.35})$$

Using  $q = 2k_P = 2\omega n/c$  and  $q = \Omega_B/v_s$ , we can express the leading terms as

$$A_{\text{Sig}} = -\frac{\epsilon_0\omega^2\gamma_e^2}{c^2 v_s \rho_0 \Gamma_B} A_P A_S^* A_{\text{Pr}} \frac{e^{-i\Delta k L} - 1}{-i\Delta k}, \quad (\text{B.36})$$

where we have dropped the signal designator on  $\omega$ . Defining the Brillouin gain factor,  $g_0$ , as Boyd does,

$$g_0 = \frac{\gamma_e^2 \omega^2}{n v_s c^3 \rho_0 \Gamma_B}, \quad (\text{B.37})$$

reduces this expression to

$$A_{\text{Sig}} = -\epsilon_0 n c g_0 A_P A_S^* A_{\text{Pr}} \frac{e^{-i\Delta k L} - 1}{-i\Delta k}. \quad (\text{B.38})$$

The intensity of the backscattered signal is given by the magnitude of the time-averaged Poynting vector, given by

$$I_i = 2n\epsilon_0 c |A_i|^2, \quad i = \text{P, S, Pr, Sig} \quad (\text{B.39})$$

which produces for the signal intensity

$$I_{\text{Sig}} = 2\epsilon_0 n c (\epsilon_0 n c g_0)^2 |A_P|^2 |A_S^*|^2 |A_{\text{Pr}}|^2 \left| \frac{e^{-e\Delta k L} - 1}{-i\Delta k} \right|^2 = 2\epsilon_0^3 \epsilon_0 n^3 c^3 g_0^2 \frac{I_P}{2\epsilon_0 n c} \frac{I_S}{2\epsilon_0 n c} \frac{I_{\text{Pr}}}{2\epsilon_0 n c} \left| \frac{e^{-e\Delta k L} - 1}{-i\Delta k} \right|^2. \quad (\text{B.40})$$

The squared modulus term containing  $\Delta k$  can be reduced as

$$\begin{aligned} \left| \frac{e^{-i\Delta k L} - 1}{-i\Delta k} \right|^2 &= \frac{(e^{-i\Delta k L} - 1)(e^{i\Delta k L} - 1)}{(\Delta k)^2} = \frac{L^2}{(\Delta k L)^2} \left[ 2 - 2 \left( \frac{e^{i\Delta k L} + e^{-i\Delta k L}}{2} \right) \right] \\ &= \frac{2L^2(1 - \cos \Delta k L)}{(\Delta k L)^2} = \frac{4L^2 \sin^2 \left( \frac{\Delta k L}{2} \right)}{(\Delta k L)^2} = \frac{L^2 \sin^2 \left( \frac{\Delta k L}{2} \right)}{\left( \frac{\Delta k L}{2} \right)^2} = L^2 \text{sinc}^2 \left( \frac{\Delta k L}{2} \right), \end{aligned} \quad (\text{B.41})$$

giving as a final expression for backscattered signal intensity,

$$I_{\text{Sig}} = \frac{1}{4}(g_0 L)^2 I_P I_S I_{\text{Pr}} \text{sinc}^2 \left( \frac{\Delta k L}{2} \right). \quad (\text{B.42})$$

To find the power of the backscattered signal, we would integrate this intensity over the effective area (acousto-optic mode overlap). For a uniform area  $A_{\text{eff}}^{\text{ao}}$ , this gives

$$P_{\text{Sig}} = I_{\text{Sig}} A_{\text{eff}}^{\text{ao}} = \frac{1}{4}(g_0 L)^2 \frac{A_{\text{eff}}^{\text{ao}}}{A_{\text{eff}}^{\text{ao}}} I_P \frac{A_{\text{eff}}^{\text{ao}}}{A_{\text{eff}}^{\text{ao}}} I_S \frac{A_{\text{eff}}^{\text{ao}}}{A_{\text{eff}}^{\text{ao}}} I_{\text{Pr}} \text{sinc}^2 \left( \frac{\Delta k L}{2} \right) A_{\text{eff}}^{\text{ao}}, \quad (\text{B.43})$$

or,

$$P_{\text{Sig}} = \frac{1}{4}(G_B L)^2 P_P P_S P_{\text{Pr}} \text{sinc}^2 \left( \frac{\Delta k L}{2} \right), \quad (\text{B.44})$$

where,

$$G_B = \frac{g_0}{A_{\text{eff}}^{\text{ao}}}. \quad (\text{B.45})$$

It can be seen that off resonance, the  $\Omega$  term from Equation B.34 goes to a lorentzian form after taking the squared modulus for intensity.

## B.2 Scattered Power Comparison to Traditional Brillouin Scattering Processes

This appendix provides a comparative analysis of the scattered power produced by our instrument to that of standard Brillouin scattering processes—that is, spontaneous and stimulated Brillouin scattering. The difference in behavior of our instrument from the traditional techniques arises due to the coherent stimulation of the acoustic mode by the pump and Stokes fields, producing a 4-wave-coupled-amplitude interaction that yields much higher scattered powers in smaller lengths. While our instrument is particularly well suited to small interaction lengths due to enhanced phase-matching relaxation, it maintains production of a significant amount of scattered power at greater lengths as well (greater than 1 m). This is because the reduction in scattered power from the breakdown of phase-matching relaxation at greater lengths is perfectly counterbalanced by the quadratic dependence on length in the overall scattered power, as seen in Equation 3.1. At very large lengths (greater than 1 km), the instrument is ultimately limited by the coherence length of the lasers employed, as the process relies on the coherent stimulation of the phonon mode and thus the mutual coherence of the pump and Stokes fields over the interaction length. Here we offer an exploration into the respective performance of each technique across the entire meaningful length scale, from nanometers to kilometers.

Despite shared dependence on basic Brillouin scattering principles, the three techniques compared here (spontaneous-, stimulated-, and coherently stimulated Brillouin scattering) yield significantly different scattered power for identical experimental parameters. At small lengths, the high-gain threshold for optical stimulation of the material fluctuations is often not achievable without the use of extremely large optical powers. This prevents the system from entering a process of exponential growth of the scattered Stokes light indicative of stimulated Brillouin scattering.<sup>13</sup> In this low-gain regime, any scattered Stokes light is spontaneously scattered from thermal fluctuations of the material, or from quantum-mechanical fluctuations of materials at the ground state. The low-gain regime is defined by an overall process gain factor, denoted by  $G = G_P P_P L$ , which is much less than unity ( $G \ll 1$ ). Here,  $G_B$  is the effective Brillouin gain in  $\text{W}^{-1} \text{m}^{-1}$  ( $G_B = \frac{g}{A_{\text{eff}}^{\text{ao}}}$ ),  $P_P$  is the pump power, and  $L$  is the effective length. This spontaneous scattering process follows a linear growth trend described by Boyd et al in 1990<sup>131</sup> as

$$R = \frac{\langle |E_S|^2 \rangle}{\langle |E_P|^2 \rangle} = (\bar{n} + 1)g\hbar\omega_S\Gamma_B \frac{L}{4A_{\text{eff}}^{\text{ao}}}, \quad (\text{B.46})$$

where  $R$  is the reflectivity, or the ratio of scattered Stokes intensity to incident pump intensity, and  $\bar{n} = (e^{\frac{\hbar\Omega_B}{k_B T}} - 1)^{-1}$  is the mean number of phonons occupying the mode due to thermal fluctuations of the material.<sup>38</sup> Rearranging this Equation and converting to effective Brillouin gain,  $G_B$ , and power by applying the effective area, we arrive at the scattered power of the Stokes spontaneous Brillouin scattering process,

$$P_S = \frac{1}{4} G_B P_P L \hbar \omega_S \Gamma_B (\bar{n} + 1). \quad (\text{B.47})$$

At room temperature and typical Brillouin frequencies in the GHz range, the quantity  $k_B T \gg \hbar \Omega_B$ , allowing

$$e^{\frac{\hbar \Omega_B}{k_B T}} \approx 1 + \frac{\hbar \Omega_B}{k_B T} \quad (\text{B.48})$$

to be a good approximation. We thus find that

$$(\bar{n} + 1) \approx \bar{n} \approx \frac{k_B T}{\hbar \Omega_B}. \quad (\text{B.49})$$

Inserting this reduced quantity into Equation B.47, we arrive at a convenient expression for the scattered power of the Stokes spontaneous Brillouin scattering process,

$$P_{S, \text{SpontBS}} = \frac{G_B P_P L \omega_S \Gamma_B k_B T}{4 \Omega_B}. \quad (\text{B.50})$$

It may be noted that the derived expression for the low-gain spontaneous regime here matches the form reported by Kharel et al. in 2016<sup>132</sup> for the complementary forward scattering process. While our backward scattering geometry differs from the forward scattering geometry discussed by Kharel et al., the underlying physics of light coupling to thermally excited acoustic modes is the same.

Next we turn to the high-gain regime leading to a stimulated Brillouin scattering process. This regime is defined by an overall process gain factor,  $G = G_P P_P L$ , that is much greater than unity ( $G \gg 1$ ). For organic liquids, this crossover threshold from spontaneous to stimulated regimes occurs in the range of  $20 < G < 25$ ,<sup>131</sup> whereas for typical lengths of single mode fiber it can be lower<sup>53</sup> owing to the small effective area compared to longer effective lengths of fiber typically used.

The reflectivity of a stimulated Brillouin scattering process in the high-gain regime is given by<sup>131</sup>

$$R = \frac{\langle |E_S|^2 \rangle}{\langle |E_P|^2 \rangle} = \frac{Y}{\sqrt{\pi}} \frac{e^G}{G^{\frac{3}{2}}}, \quad (\text{B.51})$$

where  $Y$  is the reflectivity of the low-gain (spontaneous) regime given above and  $G$  is the overall process gain factor,  $G = G_P P_P L$ . Again, converting to the effective Brillouin gain,  $G_B$ , and power by applying the effective area, we solve for the scattered power of the Stokes field,

$$P_{S, \text{StimBS}} = \frac{G_B P_P L \omega_S \Gamma_B k_B T}{4 \sqrt{\pi} \Omega_B} \frac{e^G}{G^{\frac{3}{2}}} \quad (\text{B.52})$$

This expression captures the exponential growth in scattered power as any parameter within the overall process gain factor,  $G = G_B P_P L$ , increases. However, this exponential growth can only continue while the pump is not significantly undepleted. Once the scattered power described by Equation B.52 grows to a significant fraction of the driving pump power, the exponential increase in scattered Stokes power asymptotically approaches the pump power. For very large  $G$ , virtually all of the pump energy is converted to scattered Stokes energy in a complete transfer process.<sup>13</sup> To account for pump depletion, we numerically solve the transcendental Equation derived in Boyd's Nonlinear Optics which describes the effects of pump depletion, given here in terms of power as

$$P_S(L) = \frac{P_S(0)x(1-x)}{e^{G_B P_P(0)L(1-x)} - x}, \quad (\text{B.53})$$

where  $x = P_S(0)/P_P(0)$ , or the ratio of the unknown Stokes power at the end of its journey through the medium ( $z = 0$ ) to the known pump power at the beginning (also  $z = 0$ ). This solution for  $x$ , specific to system parameters such as length, offers via its definition the solution to the unknown power of the scattered Stokes light at the end of its traversal through the effective length, given as

$$P_S(0) = xP_P(0). \quad (\text{B.54})$$

The solution to this numeric approach to scattered power in the high-gain (stimulated) Brillouin scattering regime with pump depletion effects at large  $G$  is plotted for varying effective lengths in Figure B.1, along with the analytical solutions derived previously for the low-gain (spontaneous) regime and our coherently stimulated Brillouin spectrometer (CoBS) given by Equation 3.1. System parameters used to generate the plot for each of the three processes are provided in Tables B.1 and B.2. Wherever possible, the parameters shared by all three Brillouin scattering processes were kept consistent, while quantities unique to each process were assigned their respective values.

**Table B.1:** Parameters relevant to the CoBS process for the example Ultra High Numerical Aperture 3 (UHNA3) fiber.  $G_B$  is the effective Brillouin gain,  $P_P$  is the pump power,  $P_S$  is the Stokes power,  $P_{Pr}$  is the probe power, and  $\Delta\lambda$  is the wavelength detuning of the probe from the pump.

**Coherently Stimulated Brillouin Scattering Process Model System Parameters**

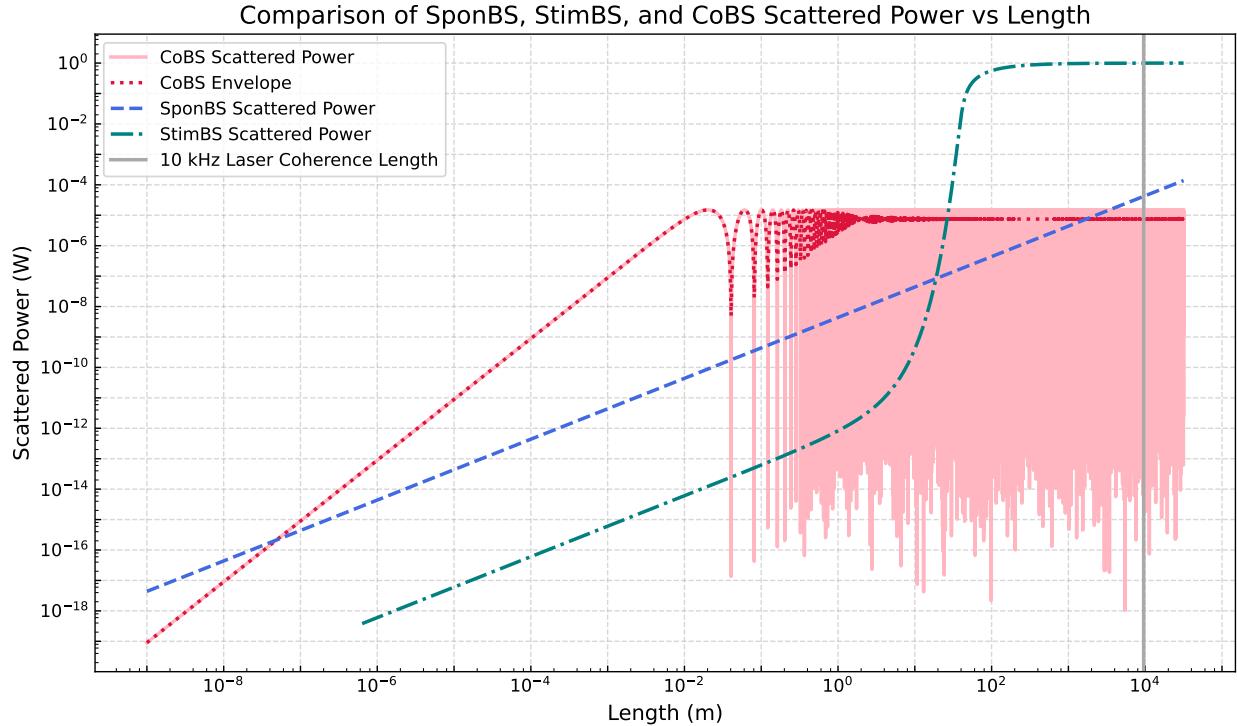
$G_B$ (W $^{-1}$ m $^{-1}$ )	$P_P$ (W)	$P_S$ (W)	$P_{Pr}$ (W)	$\Delta\lambda$ (pm)
0.6	1	1	1	20

At lengths beyond a centimeter, the phase-matching relaxation of the CoBS process begins to break down, and the specific choice in pump and probe detuning becomes critical. This corresponds to a narrowing of

**Table B.2:** Parameters relevant to the spontaneous and/or stimulated backward Brillouin scattering processes for the example UHNA3 fiber.  $G_B$  is the Brillouin gain coefficient,  $P_P$  is the pump power,  $\omega$  is the optical angular frequency,  $\Gamma_B$  is the acoustic damping rate,  $k_B$  is Boltzmann's constant,  $T$  is the temperature, and  $\Omega_B$  is the acoustic angular frequency.

Spontaneous and Stimulated Scattering Process Model System Parameters								
$G_B$ (W $^{-1}$ m $^{-1}$ )	$P_P$ (W)	$P_{S,seed}$ (pW)	$n$	$\lambda_P$ (nm)	$\Gamma_B$ (MHz)	$k_B$ (J K $^{-1}$ )	T (K)	$\Omega_B$ (GHz)
0.6	1	1	1.48	1549	$2\pi \cdot 80$	$1.38 \times 10^{-23}$	295	$2\pi \cdot 9.18$

the  $\text{sinc}^2$  function given in Equation 3.5. The scattered power beyond this length rises and falls according to the oscillations of the  $\text{sinc}^2$  function far from the origin. As length increases continuously beyond 1 m, the scattered power oscillates with increasing frequency and ceases to offer practical significance. To better visualize the scattered power offered by the instrument in this region, we have computed the envelope of scattered power. In a laboratory setting, the appropriate pump and probe detuning would be selected for the specific sample length being measured such that the scattered power function lies on a local peak of the  $\text{sinc}^2$  function.



**Figure B.1:** Comparison of scattered power from traditional Brillouin scattering processes and CoBS.

Figure B.1 shows the advantage that CoBS offers compared to the traditional Brillouin processes for the example medium of UHNA3 fiber. For lengths up to about 50 m and down to as low as 100 nm, the

coherently stimulated process employed by our instrument offers superior scattered power, with the relative advantage peaking for a length just under 1 cm. At this length, the gain factor  $G$  places the traditional process within the low-gain (spontaneous) regime, and thus the scattered power generated is only on the order of 10s of pW. In contrast, the scattered power for the same system offered by our instrument is on the order of 10s of  $\mu$ W, exceeding that of the spontaneous process by a factor of a million. This is, of course, the most ideal case for this system, however it can be seen from Figure B.1 that CoBS offers orders of magnitude more scattered power than either traditional process through a wide range of lengths.

### B.3 Observance of Fano-Resonant Asymmetries at Small Signals

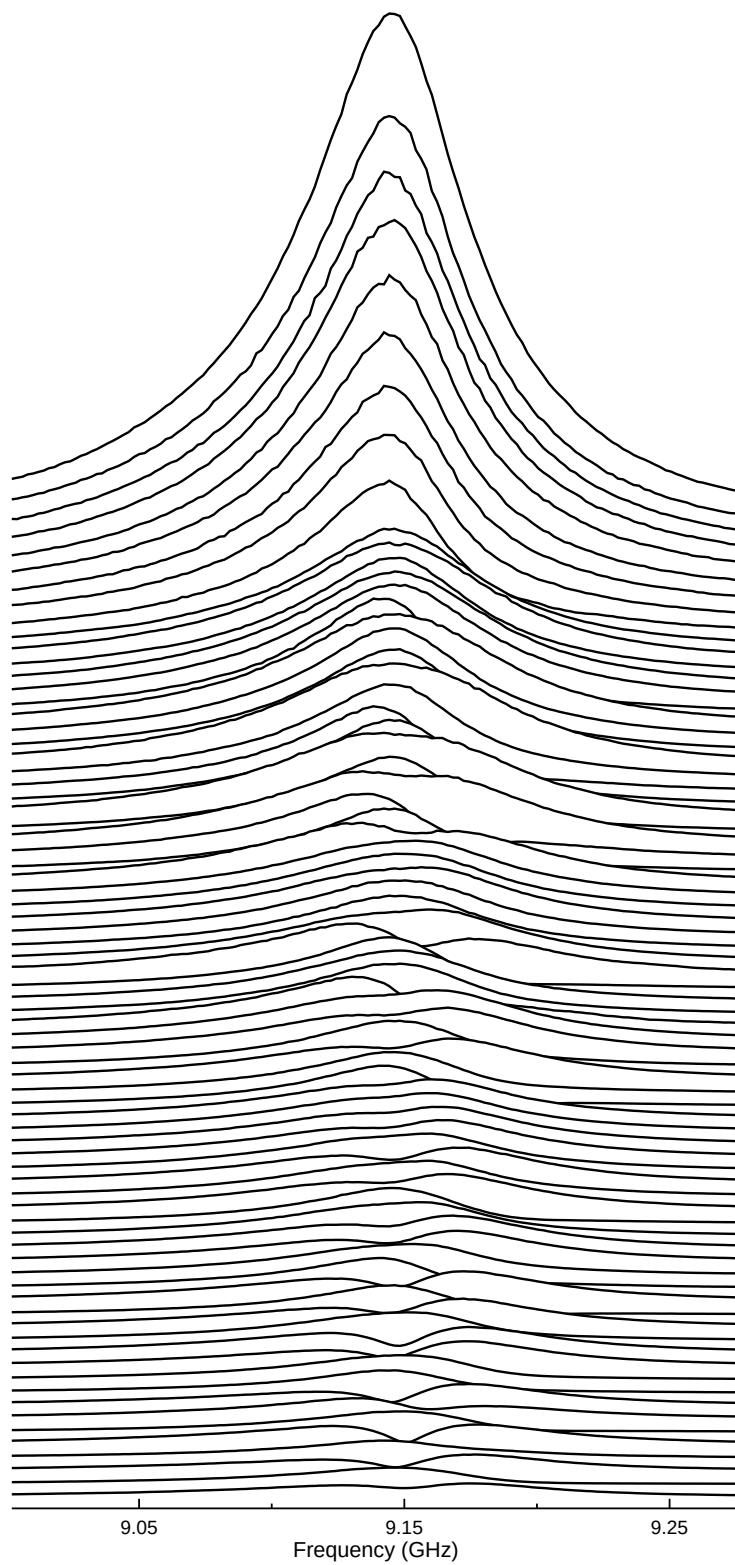
In *Fano-Resonant Asymmetries at Small Signals* (Section 3.5.4 in the main text), we discussed how Fano-type interference can distort Brillouin line shapes in situations where the resonant Brillouin amplitude becomes comparable to the background continuum. We focus here on two experiments (A and B) that reveal these Fano asymmetries especially clearly. Experiment A is a measurement series using the same 1 cm UHNA3 fiber referenced in the main discussion, for which the main text showed only the fitted amplitudes (Figure 3.6). Here we show the full spectra, illustrating the emergence of asymmetries at lower amplitude conditions. Experiment B is a distinct measurement involving a short ( $\sim 1$  mm) bulk liquid sample of carbon disulfide ( $\text{CS}_2$ ) that we briefly mentioned in Section 3.5.4 but did not detail. This experiment was performed specifically to further probe the unexpected Fano-like distortions observed in Experiment A. In each case, we outline the experimental setup, present the spectra, and highlight the appearance of Fano resonances. These observations corroborate the theoretical discussion of Fano line shapes (Section 3.5.4) and provide insight into when and why they are most prominent.

#### B.3.1 Fano Experiment A: Extended 1 cm UHNA3 Fiber Spectra

In Section 3.5 of the main text, we introduced a phase-matching experiment on 1 cm of UHNA3 fiber in which the pump-probe detuning was varied from 5 GHz to 42 GHz in 0.5 GHz increments. There, we reported only the resulting peak amplitudes, showing how they follow a  $\text{sinc}^2$  dependence on detuning (Figure 3.6 in the main text). However, each measurement in that scan also yields a full Brillouin spectrum (75 in total). Here, we present all 75 spectra to illustrate how the line shape transitions from nearly Lorentzian (when the Brillouin peak amplitude greatly exceeds the background continuum) to distinctly Fano-like (when the two amplitudes are comparable). We used the same setup and procedure described in *Phase Matching Characterization* in Section 3.5 of the main text. As the pump-probe detuning increases, the phase-matching term  $\text{sinc}^2(\Delta kL/2)$  oscillates through peaks and troughs, causing the Brillouin peak amplitude to rise and fall. When the amplitude is sufficiently large, the Brillouin mode dominates the continuum and the spectrum appears nearly Lorentzian; when it drops to the order of the background amplitude, strong interference skews the line shape into a Fano-like profile.

Figure B.2 highlights the progressive shift from Lorentzian to asymmetric line shapes. Near 5 GHz detuning (top spectra), the resonant amplitude is large relative to the background, giving a classic Lorentzian peak ( $|q| \rightarrow \infty$ ) at the resonance frequency ( $\sim 9.17$  GHz). By contrast, at detunings between  $\sim 15$ - $20$  GHz, where the  $\text{sinc}^2$  factor is near a local minimum, the peak amplitude falls to roughly the same level as the continuum, and Fano interference is observed. Interestingly, as the detuning is increased further, and the

amplitude rises again on a subsequent “lobe” of the  $\text{sinc}^2$  function, the spectra partly recover a Lorentzian shape. This cyclical behavior persists, with each local maximum yielding a near-Lorentzian profile and each local minimum reintroducing a strong Fano distortion. These observations confirm the relationship between Brillouin peak amplitude and continuum interference described in *Fano-Resonant Asymmetries at Small Signals* in Section 3.5. When the Brillouin amplitude significantly exceeds the background, the discrete phonon resonance dominates, resulting in little or no asymmetry ( $|q| \rightarrow \infty$ ). Once the two amplitudes become comparable, Fano interference skews the line shape, shifting the apparent peak frequency slightly and altering the slope on either side of the resonance. Analyzing selected spectra with both Lorentzian and Fano fits indicates that ignoring these distortions can lead to up to a 5–10% misestimation of peak amplitude in the “trough” (low-amplitude) sets. This underscores the importance of employing a Fano model in small-signal measurements where the Brillouin peak may not tower over the background. A comparative analysis of a Lorentzian vs. Fano fit function applied to highly assymetric spectra is explored in the following section, for data gathered from Experiment B.



**Figure B.2:** All measured Brillouin spectra for 1 cm UHNA3 at pump-probe detuning steps of 0.5 GHz from 5 GHz (top spectrum) to 42 GHz (bottom spectrum). Each trace is offset for clarity. The resulting asymmetries highlight the characteristic Fano-resonant behavior under low signal conditions.

### B.3.2 Fano Experiment B: 1 mm CS<sub>2</sub> Spectra and Fano Distortions

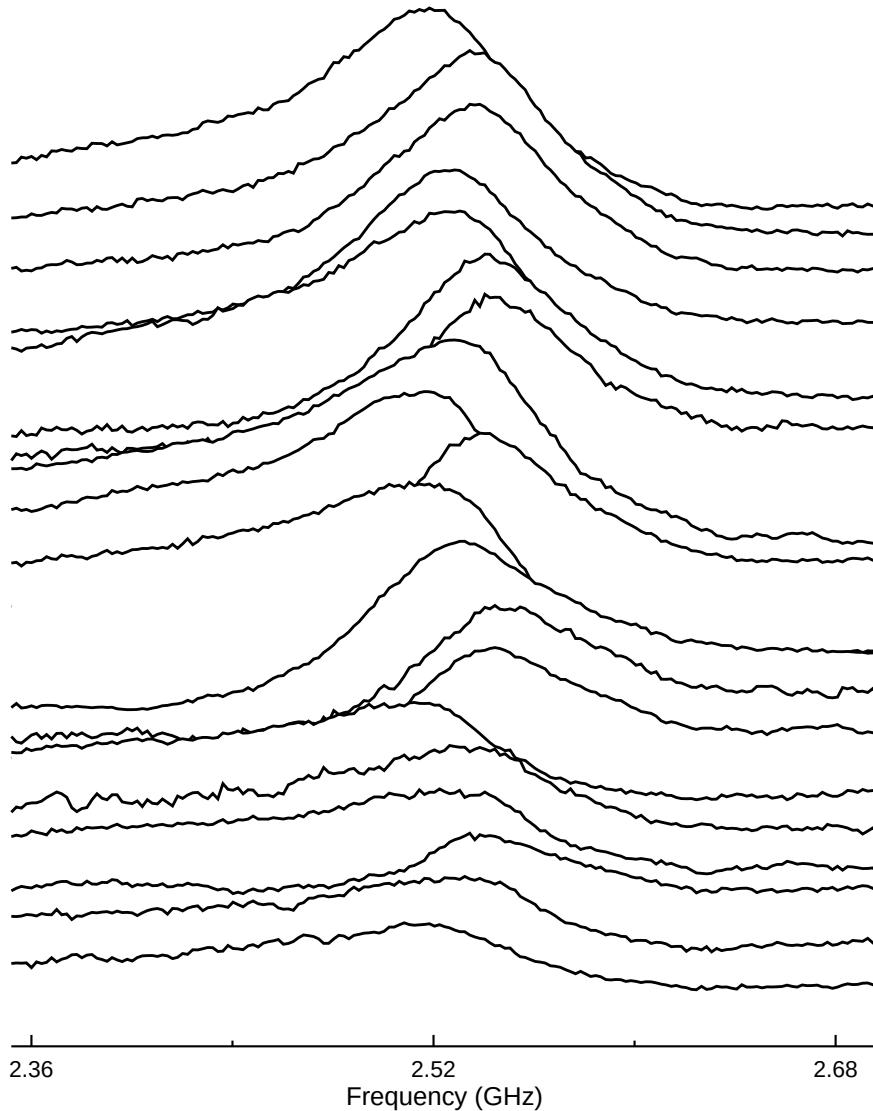
We now turn to measurements on a 1 mm-thick cell of CS<sub>2</sub> in a free-space geometry, complementing the 1 cm UHNA3 fiber results (Experiment A). Both experiments used comparable sub-Watt optical powers (on the order of  $\sim$ 60–70 mW pump,  $\sim$ 25–30 mW Stokes, and  $\sim$ 40–50 mW probe). However, unlike Experiment A, which probed a 1 cm fiber with 0.5 GHz detuning increments from 5 GHz to 42 GHz, here the detuning is stepped in 0.25 GHz increments between 10 GHz and 14 GHz. Because the CS<sub>2</sub> sample is an order of magnitude shorter (1 mm vs. 1 cm), its phase-matching bandwidth (sinc<sup>2</sup> profile) is roughly ten times wider, making these 0.25 GHz steps effectively twenty times finer than the 0.5 GHz steps used in the fiber experiment. This reduced range of detunings within a broader sinc<sup>2</sup> profile produce measured peaks all of similar amplitude to one another, as opposed to the dynamic evolution of peaks in the 1 cm UHNA3 fiber data.

Figure B.3 shows all 17 spectra obtained at detuning increments of 0.25 GHz, presented in order of increasing detuning from top spectrum to bottom spectrum. Each trace is offset vertically for clarity, with the topmost spectrum corresponding to 10 GHz and the bottom spectrum corresponding to 14 GHz detuning of the pump and the probe. A change in the detuning of the pump and probe via adjustment of the probe laser wavelength produces a change in phase of the resonant Brillouin signal. This changing resonant Brillouin phase relative to the background continuum produces spectra with different Fano-resonant distortions corresponding to specific values of the Fano parameter,  $q$ , as discussed in *Fano-Resonant Asymmetries at Small Signals* in the main text (Section 3.5.4). Fano-resonant asymmetries are seen in nearly every spectrum of this liquid experiment, indicating that the background continuum is competing strongly with the Brillouin amplitude in all measurements.

The Brillouin amplitudes featured in this experiment are an order of magnitude lower compared to the highest amplitudes seen in Experiment A. This is caused by the order of magnitude shorter sample length of CS<sub>2</sub> (1 mm) as compared to the 1 cm length of UHNA3 fiber used in Experiment A. The  $\sim$ 1000 times higher Brillouin gain offered by the CS<sub>2</sub> ( $1.5 \text{ m GW}^{-1}$ ) is discounted significantly by the  $\sim$ 350 times larger effective area offered by the beam waist in the free-space optical setup compared to the core of UHNA3 fiber used in Experiment A ( $\sim 17 \mu\text{m}$  radius beam waist vs.  $\sim 0.9 \mu\text{m}$  radius core of UHNA3). The effective Brillouin gain of the CS<sub>2</sub> used in Experiment B is thus a net  $\sim 3$  times greater than that of the UHNA3 fiber. From Equations 3.1 and 3.2 in the main text, scattered power scales with the square of both the length and the effective Brillouin gain of the sample ( $P_{\text{Sig}} \propto L^2 G_B^2$ ). Cumulatively, this makes for an approximate order-of-magnitude signal reduction for similar optical powers and pump-probe detuning.

However, both experiments (A and B) feature a sweep through a range of pump-probe detunings, with Experiment B featuring a step size effectively 25 times finer than that of Experiment A. This further cor-

robordinates these two data sets, as the signal reduction from near-center peak to a side trough of the  $\text{sinc}^2$  profile is also on the order of a 10 times reduction (Figure 3.6). This places the signal amplitudes of the  $\text{CS}_2$  spectra from Experiment B (all near *peak-center* of its  $\text{sinc}^2$  profile) on the same order as the signal amplitudes near the *troughs* of the UHNA3 fiber spectra from Experiment A. These spectra all share strong Fano asymmetries, indicating as they should that they all sit in a similar signal amplitude range: small enough that the background continuum competes but does not dominate over the signal.



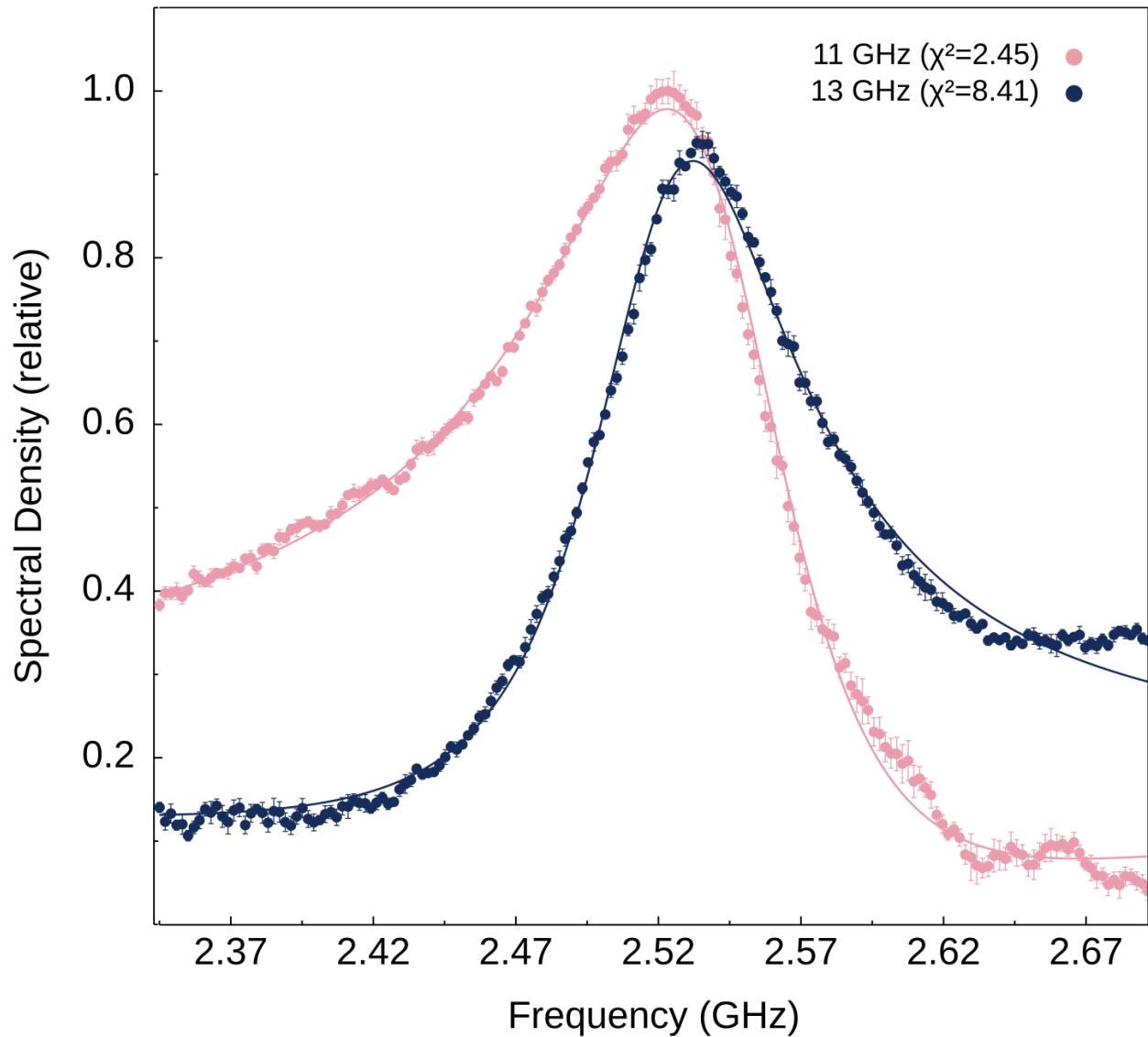
**Figure B.3:** All measured Brillouin spectra for 1 mm  $\text{CS}_2$  at pump-probe detuning steps of 0.25 GHz from 10 GHz (top spectrum) to 14 GHz (bottom spectrum). Each trace is offset for clarity.

To illustrate the strong distinction in line-shape of a spectra resulting from a positive vs. negative  $q$  value, we focus on two particular detunings that yielded notably skewed line shapes: 11 GHz and 13 GHz. Figures B.4 and B.5 compare the spectra for these two detunings normalized relative to the slightly larger

peak amplitude of the 11 GHz spectra. The line-shape of the 11 GHz spectrum exhibits a sharper rise on the higher-frequency side and a gentler roll-off on the lower-frequency side, indicative of  $q < 0$ , whereas that of the 13 GHz spectrum is skewed oppositely, featuring a sharper low-frequency side and a softer high-frequency roll-off, suggesting  $q > 0$ . Figure B.4 shows these two spectra with uncertainty-weighted Fano function fits applied, with the corresponding reduced  $\chi^2$  values reported in the legend. The Fano fits yield reduced  $\chi^2$  values of 2.45 and 8.41 for the 11 GHz and 13 GHz spectra, respectively (reduced  $\chi^2$  values near unity indicate a good fit to the data). Figure B.5 shows these same spectra with naïve Lorentz function fits applied and their corresponding reduced  $\chi^2$  evaluations of goodness-of-fit. These Lorentzian fits yield reduced  $\chi^2$  values of 39.45 and 146.5 for the 11 GHz and 13 GHz spectra, respectively, indicating that the Lorentzian function is a very poor fit to the data.. The poor fit of the Lorentzian function is due to its inherent symmetry, whereas the underlying data exhibit strongly asymmetric lineshapes. These results clearly demonstrate the superiority of the Fano model in capturing the asymmetry of small signals produced by the instrument due to Fano interference with the background continuum. This, in turn, emphasizes the importance of applying a Fano fit function when asymmetries arise to accurately extract valuable spectra parameters such as peak amplitude, center frequency, and linewidth from the data.

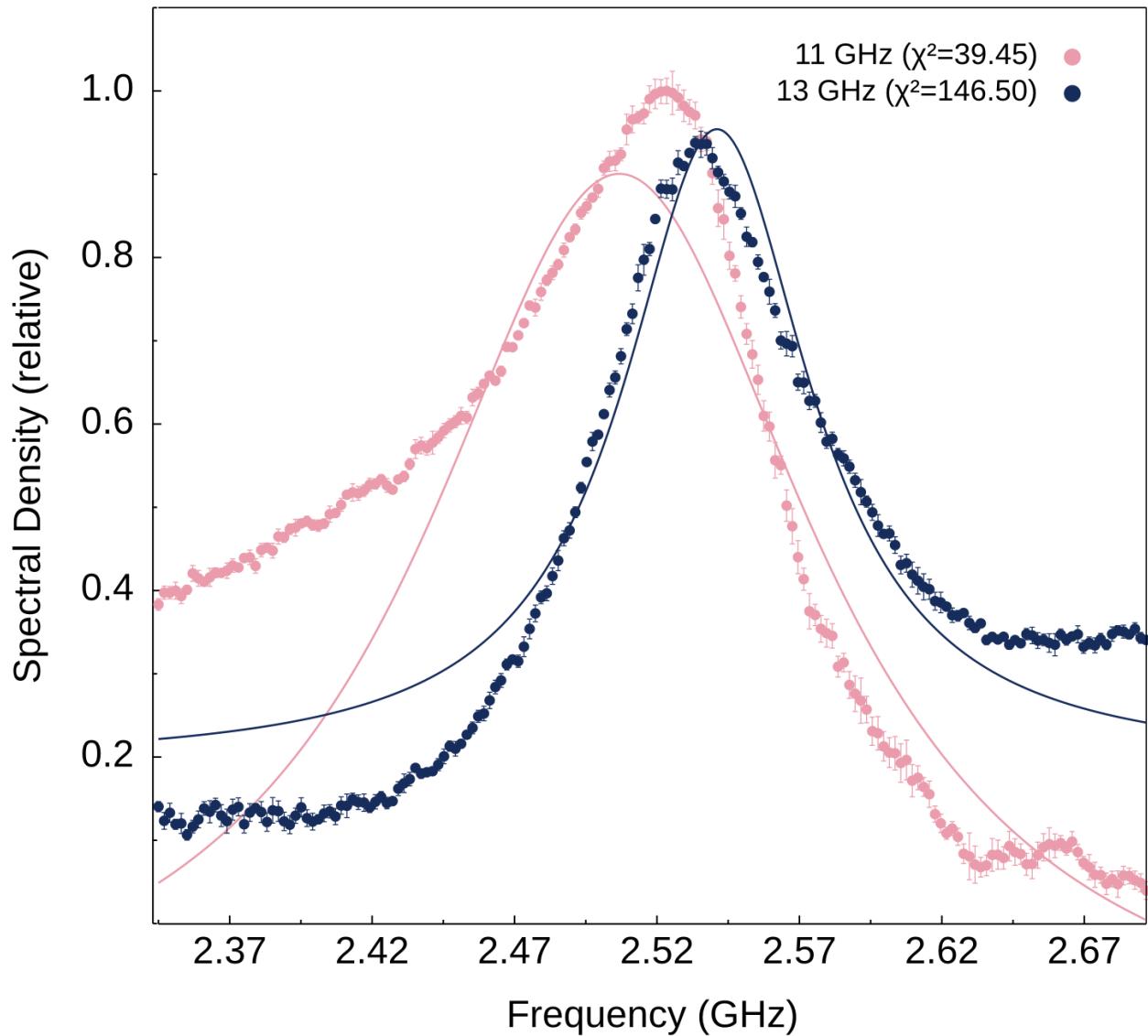
Notably, in the 11 GHz case, our Fano fit reveals a local amplitude slightly higher than what the Lorentzian fit suggests, sometimes referred to as a “peak boost.” In essence, partial *constructive* interference between the discrete Brillouin response and the broad continuum locally raises the amplitude, although it does not imply any net energy gain. As mentioned in *Fano-Resonant Asymmetries at Small Signals* (Section 3.5), this effect can aid in detecting weak resonances if the background is not too noisy. One could, in principle, tune the phase relationship to maximize this interference near the resonance, possibly producing a sharper or taller peak for certain values of  $q$  than would be achieved without interference with the background. This Fano interference-tuning of the discrete mode relative to the background can be done dynamically via adjustment of the probe laser wavelength and, critically, can be adjusted independently from the phase-matching bandwidth tuning (pump-probe detuning). This ability to dynamically adjust the discrete-continuum interference is an elegant and notable feature, as in typical systems this is adjusted via changes in physical geometry or material doping of the sample.<sup>80, 81, 133</sup>

## 1 mm CS<sub>2</sub> CABS



**Figure B.4:** Comparison of representative spectra at 11 GHz and 13 GHz, showing the positive vs. negative  $q$  asymmetry in 1 mm CS<sub>2</sub>. A Fano function fit has been applied to each spectra, with  $\chi^2$  value for each fit listed in the plot legend.

## 1 mm CS<sub>2</sub> CABS



**Figure B.5:** Comparison of representative spectra at 11 GHz and 13 GHz, showing the positive vs. negative  $q$  asymmetry in 1 mm CS<sub>2</sub>. Here, a naïve Lorentz function fit has been applied to each spectra, with  $\chi^2$  value for each fit listed in the plot legend. These spectra show strong Fano-resonant asymmetry and thus the standard Lorentz function offers a poor fit for these spectra, quantified by the  $\chi^2$  evaluation metric for goodness of fit as compared to the same evaluation of the Fano function fit.

To better convey the cyclical evolution of the 75 measured UHNA3 spectra from Experiment A, several animated GIFs have been generated. The following is a link to the GitHub repository which hosts these files, along with all raw data, measurement logs, and plots generated in support of this work (see See Appendix C):

<https://github.com/HamletTheHamster/Plotting-Data-in-Go>

The reader will find several GIFs which step through each spectrum in ascending pump–probe detuning at various frame rates. Readers are encouraged to view them for an animated perspective on how the spectra evolve with increasing pump–probe detuning, with special attention to transitions between Lorentzian and Fano-distorted line shapes.

## B.4 CoBS Mini Experiment: Equal Contribution of Pump, Stokes, and Probe

Equation 3.1 gives the somewhat unintuitive result that the powers of the Pump, Stokes, and Probe waves contribute equally to the resulting scattered power of the Signal and invites verification with a miniexperiment. Initially, this experiment was motivated by a practical consideration: determination of whether the placement of a high power amplifier on any specific line of the setup (Pump, Stokes, or Probe) would offer any advantage over another.

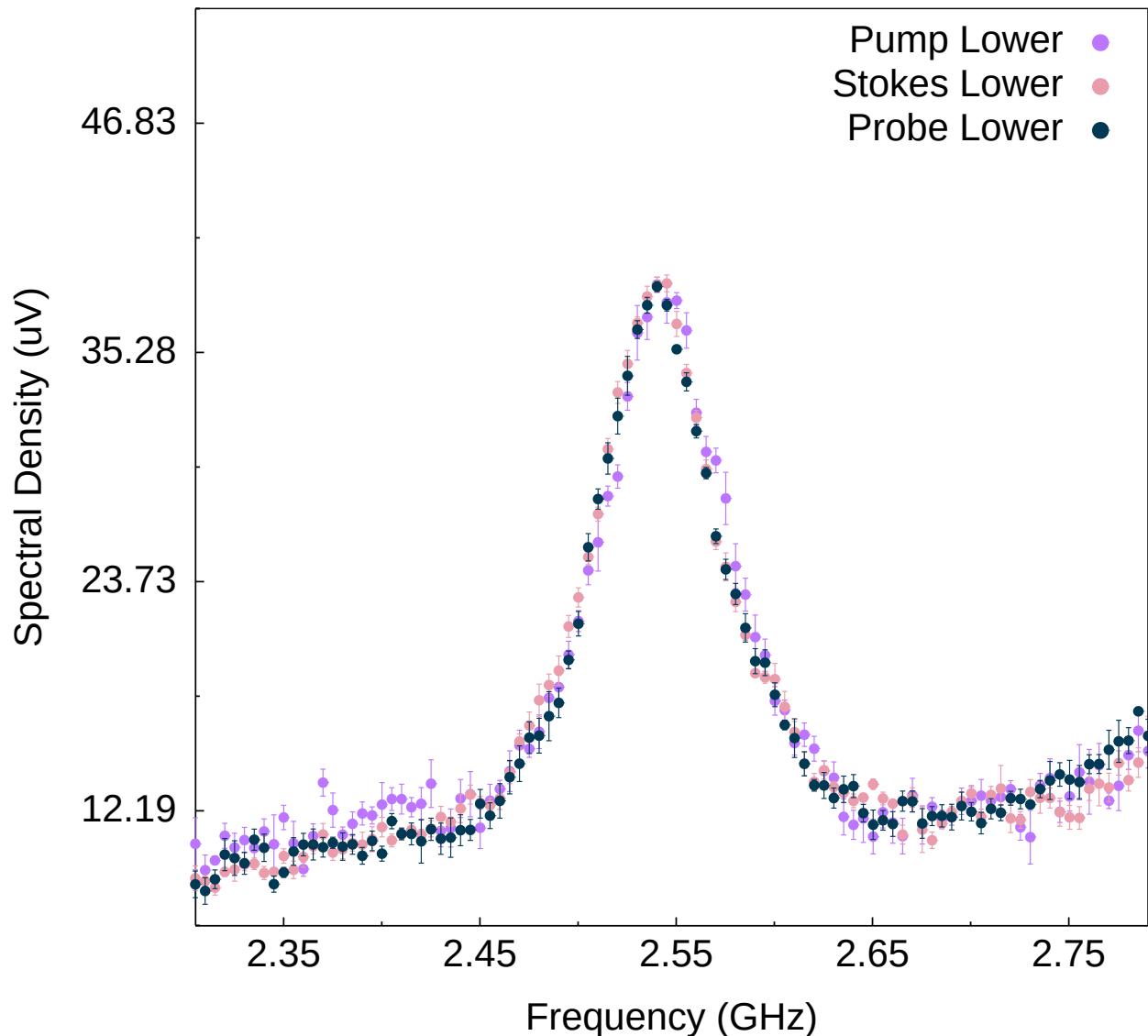
To test this, we conducted a controlled experiment with a 1 mm CS<sub>2</sub> sample. For each measurement, one of the three source powers (Pump, Stokes, or Probe) was systematically reduced by 75% while holding the others constant and ensuring consistent experimental conditions across trials. Table B.3 shows the respective powers for each source during the three measurements, along with the multiplicative total contribution of the three powers for each measurement towards the generation of scattered power of the Signal.

Measurement	Pump Power (mW)	Stokes Power (mW)	Probe Power (mW)	Total (mW <sup>3</sup> )
Pump Lower	19.190	32.210	54.560	$3.372 \times 10^4$
Stokes Lower	76.600	8.020	54.650	$3.359 \times 10^4$
Probe Lower	76.600	32.530	13.480	$3.359 \times 10^4$

**Table B.3:** Power values for each source (Pump, Stokes, Probe) across the three measurements, with the multiplicative total power for each setup.

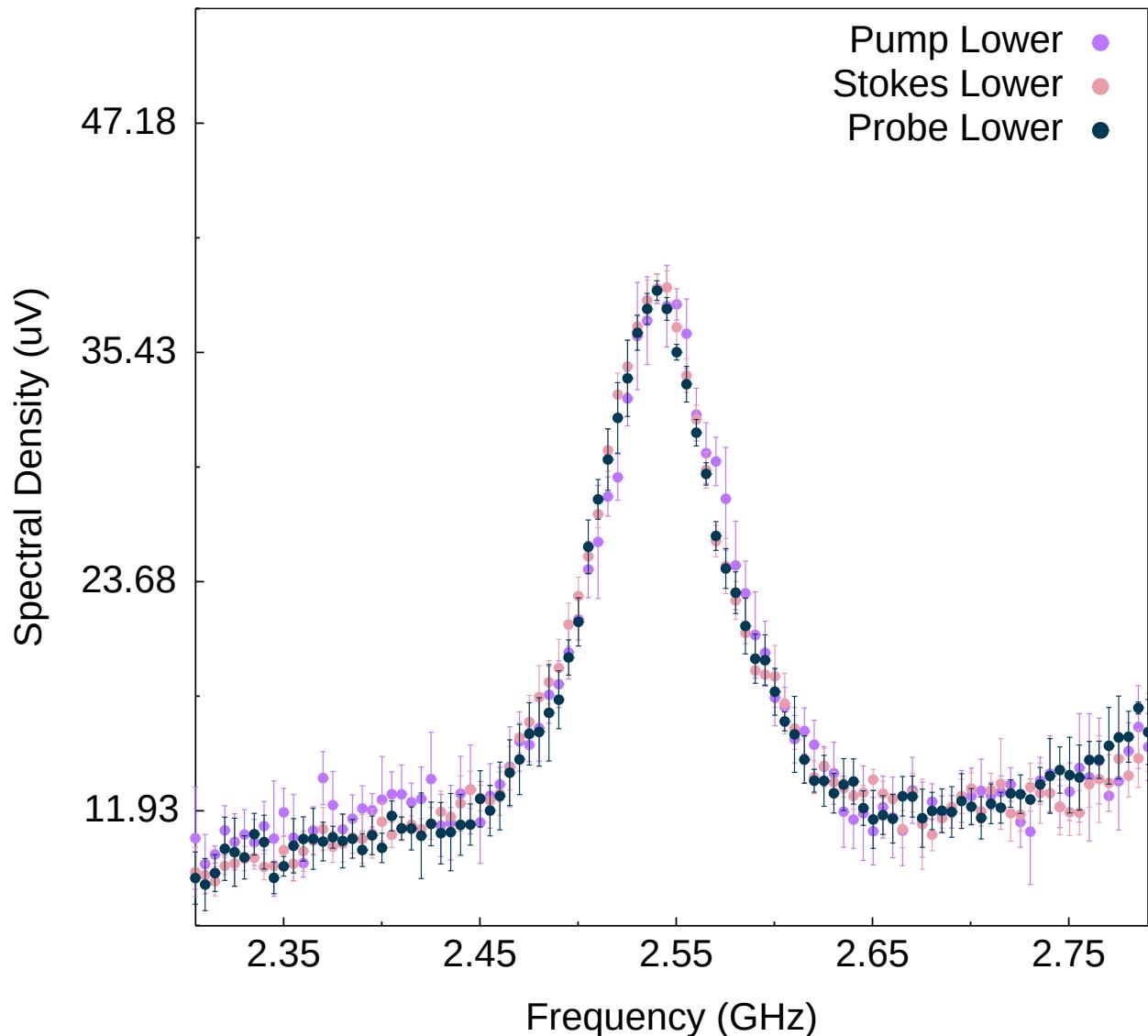
Figure B.6 displays the average results from these three measurements, plotted with error bars representing one standard deviation of the mean. For increased certainty, Figure B.7 presents the same data with error bars extended to two standard deviations, providing additional confidence in the reproducibility of the results. This experiment confirms that the scattered Signal power indeed depends equally on each of the three contributing wave powers, as expected from the theoretical framework. Consequently, boosting the power of any of the three sources affects the Signal power equally, allowing flexibility in pragmatic design across any of the three lines. Ultimately, this result reinforces the reliability of Equation 3.1 for predicting Signal power across a range of power distributions within practical settings.

## 1 mm CS<sub>2</sub> CABS



**Figure B.6:** Scattered power spectra with varying proportions of pump, Stokes, and probe power and error bars representing one standard deviation of the mean for each measurement.

## 1 mm CS<sub>2</sub> CABS



**Figure B.7:** Scattered power spectra with varying proportions of pump, Stokes, and probe power and error bars extended to two standard deviations of the mean for each measurement.

## Appendix C

### Code and Data Availability

#### C.1 Plotting Data In Go

Data plots presented in this dissertation were generated using custom programs written by the author in Golang or Python. The source code for these program, as well as all data collected and plots generated in support of the work presented in this dissertation, are publicly available on GitHub via the following link:

<https://github.com/HamletTheHamster/Plotting-Data-in-Go>

A video visualizing the evolution of data collection and development of functionality of the plotting program is available on YouTube:

<https://www.youtube.com/watch?v=PdoWVoJx2Vg>



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